SPECTRAL PROFILES

OF

CERTAIN ATOMIC LINES

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CONTENTS

ABSTRACT

Several calculations have been carried out using the transition operator technique.

The radiative decay in a two-level quantum system with excited state coupled by an external perturbation has been investigated and an expression obtained for P(t), the probability of finding the atom in its excited state at time t. This is seen to possess a steady value for $\gamma t >> 1$ and a third term which decays at nearly half the rate of its second.

Next radiative decay of an atom with two close-lying excited states is considered and $|b_q|^2$, the spectral density for spontaneous emission from the uppermost level, calculated. This shows that the proximity of the upper two levels makes the line-shape non-Lorentzian.

Heisenberg equations of motion were then derived for the transitions operators of an ℓ-level atom undergoing radiative decay, the possibility of overlapping pairs of levels being ignored. These master equations were then used to obtain the spectral profiles of certain atomic lines, employing the so-far avoided Markoff approximation.

First the spectral density of the scattered radiation was found for transitions between levels $5P_{3/2}$ and 4S (ground state), and 6S and $5P_{3/2}$, in the potassium atom when driving fields of arbitrary strength coupled levels 6S and $4P_{3/2}$, and $4P_{3/2}$ and 4S. Triple-peaked profiles were obtained.

Spectral profiles for emission were then derived for transitions between levels 3P and $1S_{1/2}$ (ground state), and 3P and $2S_{1/2}$ in the hydrogen atom when a driving field of arbitrary strength coupled levels $2S_{1/2}$ and 3P. The former profile was double the latter quadruple-peaked.

Lastly master equations were derived for an atom, with two excited levels undergoing radiative decay, when allowance was made for the

possibility of their overlapping. A driving field of arbitrary magnitude coupled the uppermost and ground states. The spectrum for the scattered light resulting from transitions between the uppermost and ground state was found and graphs computed for various field strengths and separations of the excited states.

CHAPTER I

INTRODUCTION

A. Summary

Several calculations concerning radiative decay have been carried out. They involve use of the transition operator technique described by Lehmberg in references [1] and [2].

In Section B of the present chapter a review is given of the background to the calculations. This review explains the terminology and the derivations of certain equations used in the following chapters and is given so that it may be referred back to in the subsequent chapters and detailed explanations need not be given in the body of the text. It is not an original part of the thesis.

Chapter VIII complements Chapter I Section B in giving a discussion of the various approximations used in the thesis and their validity. It also contains reasons for choosing our method of approach and points out the limitations of the Lehmberg method, on which the major part of the calculations is based.

The first two calculations described in Chapters II and III, are outlined in the attached papers published jointly with Dr. L. M. Bali [3], [4]. In Chapter II, [3], radiative decay in a two-level system with excited state coupled by an external perturbation has been considered. The quantum system is simultaneously coupled by a quantised radiation interaction, describing the decay, and by a classical external perturbation. As in Lehmberg's papers [1], [2] the approximation used is that no appreciable secular change occurs in the atomic states during the times of the order of an atomic period. The solutions are valid for times very much greater than the reciprocal of the atomic resonance frequency and these times may or may not be very much greater than the atomic life-time. The problem is essentially the same as that of Keller and Robiscoe [5] but their theory has been worked out for a time scale very much greater than the atomic life-time, a restriction we have avoided. They also assume the classical external perturbation to be less than the atomic level separation. We do not necessarily assume this but we do assume $\mu(= |\lambda|^2/\omega^2)$ and $\beta(= 2\gamma/\omega) << 1$ and neglect powers higher than their first order, where λ is a c-number coupling parameter, γ is the decay constant and $\omega = \omega_0 - \Omega$, where ω_0 is the level separation and Ω the frequency shift. Our more complete solutions lead to entirely different conclusions from those of Keller and Robiscoe about the effect of the external perturbation on the radiative decay of the system, our most important conclusion being that the equations result in the non-appearance of the new type of modulation factor discovered by Keller and Robiscoe [5]. Our solutions have the advantage of enabling evaluation of expressions for the state populations to be made in a very simple and direct way and they do lead to conclusions similar to those derived, for the three-level problem, by Fontana and Lynch [6].

In Chapter III, [4], the radiative decay of an atom with two close-lying excited states is considered and the effects on the line shape of spontaneous emission of the atom as a result of the presence of this second excited state is calculated on the assumption no direct transitions can occur between the two close-lying states. Mollow and Miller [7] have shown, in detail, how the effect of spontaneous emission of an atom can be described by considering coupling of the atom to a "bath" of harmonic oscillators representing modes of the electromagnetic field. They consider a two-level atom. Lehmberg's method [1], [2] is an improvement on this as it does not require the use of the Markoff approximation or that the coupling between the atom and the bath be sufficiently weak to consider its effect up to second order of the coupling constants. The resulting equation is the same as that of Morozov and Shorygin [8], derived using the Heitler-Ma method, when all decay rates are assumed to be equal. It shows that inclusion of the exchange of virtual photons between the excited levels causes a change in the emission line contours which becomes increasingly noticeable when their separation, $\Delta \gg$ the decay constants γ. This change involves a shifting of the peak intensity to left or right depending on whether the atom is initially in the uppermost or next to uppermost state respectively.

The spectral profile for the atomic decay (essentially the Fourier transform of a two-time correlation function) is obtained without use of the fluctuation-regression theorem. This method is therefore useful for describing situations where calculations based on perturbation theory become invalid; e.g. in the presence of a very intense radiation field such as that of a laser.

In Chapter IV we consider the problem of a multi-level atom undergoing radiative relaxation. Heisenberg equations of motion are derived for the transition operators using, as previously, a Hamiltonian obtained under both dipole and rotating wave approximations for an atom at the origin of co-ordinates. Contrary to Lehmberg [1], we do not restrict ourselves to specific regions of the spectrum, but rather consider the broad spectrum limit. The possibility of overlapping pairs of levels is ignored in order to simplify the equations, although in Chapter VII this is allowed for in the relatively simple case of the three-level atom. The master equations so derived are used to obtain a general expression for the 2-time atomic correlation function, employing the so-far-avoided Markoff approximation in the manner of Mollow's paper [9]. This general expression is used in Chapters V and VI to calculate the spectral density of the scattered light for various transitions in potassium and hydrogen atoms. The use of the Markoff approximation, instead of Lehmberg's approach is necessary at this stage since the equations otherwise require the introduction of various tedious complications for their solution. This is due to the fact that multiplying the transition operator equations, modified by the addition of driving terms, on the right by vacuum state $|0\rangle$ for all q photons does not reduce the number of equations and lead to easy solution as in |2|. Our calculations nevertheless still have the advantage over Mollow's of not being restricted to second order in the coupling constants.

In Chapter V the spectral density of the scattered radiation is found for transitions between (a) levels $5P_{3/2}$ and 4S (ground state) and between (b) levels 6S and $5P_{3/2}$ in the potassium atom, when strong driving fields couple levels 6S and $4P_{3/2}$, $4P_{3/2}$ and 4S. In both cases (a) and (b) triple-peaked spectral profiles are obtained. The model used consists of 10 of the levels between level 4S (ground state) and level 6S; i.e. ignoring the degeneracy of levels 4S, 5S, 4D and 6S. The initial time t' is kept arbitrary in both cases so that we can consider the overall effect of several multiphoton processes and not just single processes of emission or absorption.

Next, in Chapter VI, we apply the general equations to the specific case of emission spectra in the hydrogen atom when strong radiation couples levels $2S_{1/2}$ and 3P. The two cases considered are the spectral densities of the emitted light for transitions from (a) state 3P to $1S_{1/2}$ (ground state) and from (b) state 3P to state $2S_{1/2}$ when the atom is naturally initially in state 3P. Five of the levels between $1S_{1/2}$ (ground state) and 3P are considered, namely levels $1S_{1/2}$, $2P_{1/2}$, $2S_{1/2}$, $2P_{3/2}$ and 3P. In case (a) a double-peaked and in (b) a 4-peaked profile was obtained.

The results of the calculations for the potassium atom are compared with numerous experimental papers on potassium, [10] to [31], bearing in mind that our calculations refer to the overall effect of all the individual types of transitions mentioned in these papers. In ref. [11], the origin of the 3 lines obtained for transitions (b) is said to be a 2- or 3-photon processes. In [28], it is stated that "the observed effects are connected with the splitting of the atomic levels in the external field" as described by the wave function in their expression (1) which shows the splitting of the non-degenerate states of the atom, namely states $4P_{3/2}$ and $6S_{1/2}$, due to the external field. In [29] also they stress "the field origin of absorption line splitting at the $4S_{1/2}$ - >$5P_{3/2,1/2}$ transition". They also point out in [29] that when the emission intensity is high enough, distinction between processes involving different numbers of photons becomes groundless. In other words, the perturbation theory approach becomes invalid, as stated earlier. In particular, they use the structure of the $4S_{1/2} - 5P_{3/2}$ (λ = 4044 Å) absorption line, as an example, to show that 1 -, 2 - and 3-photon processes are "mixed-up" in an intense resonance field so that under these conditions it is more relevant to talk about "a SINGLE process of violet absorption in which the line structure is interpretated as a result of FIELD SPLITTING of atomic levels". They point out that "such an approach is in full accord with the spirit of non-linear spectroscopy" and they regard their data "as an experimental verification of one of its main theses". In reference [29], equation (A.8.) for the atomic absorption spectrum also contains a set of equidistant TRIPLETS. In other papers the theoretical stress is on the calculation of cross-sections and population densities.

The results obtained for the hydrogen atom are compared with those of Zernik [32] and Rautian and Sobel'man [33]. In Zernik's paper, he neglects spontaneous decay from level 3P to 2S when using the strong signal theory whereas we haven't neglected it entirely. However our results cannot truly be compared with his, since he considers the 2-photon transition $2S_{1/2}$ to $1S_{1/2}$ through the intermediate level 3P, whereas we consider only the SINGLE photon processes 3P to $1S_{1/2}$ and 3P to $2S_{1/2}$. We could only consider such SINGLE photon processes since we restricted the initial time to $t' = 0$. It was necessary to consider the processes as starting at level 3P and that the atom had been excited to that level by the field long ago. Had we not done this but assumed instead the atom to be initially in level $2S_{1/2}$, the spectral densities for emission would have been identically zero so that the only alternative would be to consider t' to be arbitrary as in the previous chapter. In the paper of Rautian and Sobel'man, a hypothetical atomic system is considered where $\gamma_{32} << \gamma_2$ (in their notation), but we do not assume this, though for weak fields, we assume $\dfrac{\gamma_{53}}{\gamma_{53} + \gamma_{51} - \gamma_{32}}$ to be small, i.e. in their notation $\dfrac{\gamma_{32}}{\gamma_{32} + \gamma_{31} - \gamma_2}$ small, i.e. $\gamma_{32} << \gamma_{31} - \gamma_2$ since Zernik says that spontaneous decay from $3 \rightarrow 2$ is negligible, i.e. $\gamma_{31} >> \gamma_2$. We do not specifically assume $\gamma_{32} << \gamma_2$. Our calculation is an improvement on these two papers in that it removes these two restrictions but all the same it unfortunately only covers single photon emission processes. We point out how we could modify our calculations to take into account the 2-photon process at the end of the chapter.

In the following chapter, Chapter VII, we derive equations of motion for a general 3-level atom allowing for the possibility of overlapping of the upper two levels and from these derive an expression for the power spectrum of the scattered light for transitions between the uppermost level, 3, and lowermost level, 1, when a strong driving field, of arbitrary magnitude, couples levels 3 and 1. Curves for the spectral profiles are computed for various separations of levels 3 and 2 and for various magnitudes of the driving field for the case of resonance when all decay rates have equal magnitude, as in Chapter III, i.e. $\gamma_{31} \doteq \gamma_{21} \doteq \Gamma_{31} \doteq \Gamma_{21}$ $(= \gamma)$. A similar model has been used by Apanasevich et al. [34] where he considers levels 3 and 2 to be always degenerate. In contrast to use, he allows transitions to occur between levels 3 and 2. He also limits himself to consideration of a weak field so that he can use a linear approximation to obtain two equations, i.e. equations (2). On the other hand, we have performed a more exact treatment involving 9 coupled equations, which is not limited to weak fields.

The computed spectral profiles show that the inclusion of "cross-type" decay constants, Γ_{31} and Γ_{21}, substantially affects the spectral profiles for the scattered radiation particularly for values of ω_3 around $\omega_3 \doteq 1/2G$, i.e. when the separation of the excited levels is of the order of $\lambda\mathcal{E}_0 = (\hat{e}_{CD} \cdot \underline{r}_{31})\mathcal{E}_{CD}/\hbar$. For this region the profile no longer corresponds to that obtained by Mollow in ref. 9, i.e. a central peak with 2 side-peaks of equal and lower intensity symmetrically spaced about it. For values $\lambda\mathcal{E}_0 > \gamma$, when $\omega_3 < 1/2G$ the LH peak begins to dominate over the RH one, until at 1/2G there are no emission peaks at all. When $\omega_3 > 1/2G$ the LH peak decreases and the RH one increases until the Mollow-type situation is again obtained. This phenomenon requires experimental verification. Calculations taking into account the "cross-type" decay rates do not appear to have been carried out except for weak fields, viz. ref. 34.

Finally we should like to further emphasise that the methods used here are preferable to existing methods, particularly the perturbation approach, especially when dealing with problems involving strong incident radiation.

B. Background [35]

The calculations set out in the following chapters involve the use of the non-relativistic Dirac formulation of quantum mechanics. Before we commence these calculations it would seem fit to explain certain terms which are frequently used.

1.B.1. Hermitian operators

Firstly, most of the operators we shall use, with the notable exception of the boson operators, are hermitian, i.e. they are real linear operators representing dynamical variables which give real numbers when measured. They possess the property of self adjointness so that if L is a hermitian operator

$$L = L^{+} \tag{1.B.1}$$

also

$$\langle p|L|b\rangle = \langle b|L|p\rangle^{*} \tag{1.B.2}$$

for any state vectors $|b\rangle$ and $|p\rangle$. There are two important theorems for all hermitian operators. One being that the eigenvalues of linear hermitian operators are REAL ($\ell = \ell^{*}$), and the other being that two eigenvectors of a linear hermitian operator L belonging to different eigenvalues are orthogonal ($\langle \ell' | \ell'' \rangle = \delta_{\ell',\ell''}$). The latter theorem is an expression of the orthonormality relation for eigenvectors and applies to the eigenvalue problem in which the norm of vectors, i.e. $\langle \ell' | \ell' \rangle$ is finite, viz.

$$\langle \ell' | \ell'' \rangle = \delta_{\ell,\ell''} \qquad \left.\begin{array}{l}\text{where } \delta_{i,j} \text{ is the Kronecka} \\ \text{delta defined by} \\ \delta_{i,j} = \begin{cases} 1 & \text{if } i = j \\ 0 & \text{if } i \neq j \end{cases}\end{array}\right\} \tag{1.B.3}$$

The completeness or closure relation for discrete, as opposed to continuous, eigenvalues of an observable is

$$\sum_{\ell} |\ell\rangle\langle\ell| = I \qquad \text{where } I \text{ is the identity operator.} \qquad (1.B.4)$$

These hermitian operators having such a complete set of eigenvectors $\{|\ell\rangle\}$ are called observables. The symbol $\{|\ell\rangle\}$ signifies that the set of vectors is complete and also that the set may be regarded as a particular set of orthogonal unit basis vectors in the sense of

$$\langle\ell'|\ell''\rangle = \delta_{\ell',\ell''} \qquad (1.B.5)$$

1.B.2. Matrices

Ket and bra vectors, viz. $|\rangle$ and $\langle|$ respectively, and linear operators in a space have a matrix representation. The trace of a square finite matrix A is defined as the sum of the diagonal elements, viz.

$$\mathrm{Tr}(A) = \sum_i A_{ii} \qquad (1.B.6)$$

where Tr is the abbreviation for trace and A_{ii} is the ith diagonal element. The trace of a product of finite square matrices is invariant under cyclic permutations, i.e.,

$$\mathrm{Tr}(ABC) = \mathrm{Tr}(BCA) = \mathrm{Tr}(CAB) \qquad (1.B.7)$$

1.B.3. Representations

There are three pictures of quantum mechanics: the Schrödinger picture, the Heisenberg picture (which we shall use), and the interaction picture.

In the Schrödinger picture

(1) observables (p, q and H), which are hermitian operators, are time independent

(2) eigenvectors of operators p_s, q_s and H_s are stationary (time

independent) vectors $|p'\rangle$s, $|q'\rangle$s, $|E\rangle$s and may be taken as basis vectors to represent operators and state vectors, i.e. basis vectors are stationary

(3) the dynamical state vector $|\psi_S(t)\rangle$ moves.

In the Heisenberg picture

(1) operators are time-dependent $A_H(t)$

(2) basis vectors move

(3) state vectors remain stationary $|\psi_H(t_0)\rangle$.

In the interaction picture

(1) operators are time-dependent $A_I(t)$

(2) basis vectors move

(3) state vectors move $|\psi_I(t)\rangle$, this marking its sole difference from the Heisenberg picture.

All these descriptions of quantum mechanics are physically equivalent and so any one can be used depending on which is convenient for a particular situation.

The state vectors in the Schrödinger and Heisenberg pictures are related, by definition, by

$$|\psi_S(t)\rangle = U(t,t_0)|\psi_H(t_0)\rangle \tag{1.B.8}$$

where subscript H designates Heisenberg picture and S, Schrödinger picture.

The average value of an operator A_S, when it is known with centainty that system is in state ψ, is

$$\left.\begin{aligned}\langle A\rangle &= \langle\psi_S(t)|A_S|\psi_S(t)\rangle && \text{in the Schrödinger picture}\\ &= \langle\psi_H(t_0)|U^+A_SU|\psi_H(t_0)\rangle &&\\ &= \langle\psi_H(t_0)|A_H(t)|\psi_H(t_0)\rangle && \text{which is the average value of A at time t in the Heisenberg picture}\end{aligned}\right\} \tag{1.B.9a}$$

since we define the operator in the Heisenberg picture by

$$A_H(t) = U^{+}(t,t_0)A_S U(t,t_0) \tag{1.B.9b}$$

thus making the average the same in both pictures. The latter transformation is called a similarity transformation when $U^{+} = U^{-1}$, i.e. when U is unitary, and is valid even if A_S has an explicit time dependence.

The form of U, the transformation operator, depends on whether the system is conservative or non-conservative, i.e. whether the Hamiltonian, H, is time independent <u>$H \neq H(t)$</u> <u>(conservative system)</u> or time dependent <u>$H = H(t)$</u> <u>(non-conservative system)</u>. We shall be considering non-conservative systems.

For a conservative system

$$U(t,t_0) = \exp\left[\frac{-iH(t-t_0)}{\hbar}\right] \tag{1.B.10}$$

and for a non-conservative system

$$U(t,t_0) = 1 - {}^{i}/_{\hbar}\int_{t_0}^{t} H(t')dt' + \left({}^{-i}/_{\hbar}\right)^2 \int_{t_0}^{t} H(t')dt' \int_{t_0}^{t'} H(t'')dt'' + \ldots \tag{1.B.11}$$

since U is unitary

$$U^{+}U = UU^{+} = I. \tag{1.B.12}$$

The equation of motion for an observable A in the Heisenberg picture can be obtained by differentiating both sides of the equation for the transformation law with respect lot. It is necessary to also use the equation

$$i\hbar \frac{dU}{dt} = HU \tag{1.B.13}$$

and its adjoint

$$-i\hbar \frac{dU}{dt} = U^{+}H \qquad (1.B.14)$$

and (1.B.12).

Thus,

$$i\hbar \frac{dA_H}{dt} = \left[A_H, H_H\right] + i\hbar \frac{\partial A_H}{\partial t} \qquad (1.B.15)$$

and if A_S has no explicit time dependence

$$\frac{dA_H}{dt} = \frac{1}{i\hbar} \left[A_H, H_H\right]. \qquad (1.B.16)$$

This is the formula we shall be using to derive Heisenberg equations of motion for non-conservative systems.

1.B.4. Boson operators

We shall now consider the properties and nature of boson operators. First we recall that particles in nature having the property that any number may occupy the same dynamical states are called BOSONS. Examples of such particles are light quanta (photons), elastic vibrations in crystals (phonons), α-particles, etc. We shall be interested in photons and phonons only. Boson creation and annihilation operators, a^{+} and a respectively, obey the commutation relation

$$\left[a, a^{+}\right] = 1 \qquad (1.B.17)$$

a^{+} is also known as the raising operator since when it operates on oscillator state $|n\rangle$, containing n quanta, it generates state $|n+1\rangle$, containing n + 1 quanta, i.e.

$$a^{+}|n\rangle = \sqrt{n+1}\,|n+1\rangle. \tag{1.B.18}$$

Similarly a is known as the lowering operator since

$$a|n\rangle = \sqrt{n}\,|n-1\rangle \quad \text{where n is an integer} > 0, \tag{1.B.19}$$

and

$$a|0\rangle = 0 \tag{1.B.20}$$

$N = a^{+}a$ is known as the number operator since

$$N|n\rangle = n|n\rangle \qquad \text{where } n = 0, 1, \ldots, \infty \tag{1.B.21}$$

n being the number of quanta in the wave.

Other commutation relations can be derived from eq. (1.B.17)

$$[a, a^{+}a] = a \tag{1.B.22a}$$

$$[a^{+}, a^{+}a] = -a^{+} \tag{1.B.22b}$$

Although operators a and a^{+} are non-hermitian the Heisenberg equations of motion still apply, viz.

$$\frac{da_H}{dt} = \frac{1}{i\hbar}\left[a_H, H_H\right] \tag{1.B.23a}$$

$$\frac{da_H^{+}}{dt} = \frac{1}{i\hbar}\left[a_H^{+}, H_H\right] \tag{1.B.23b}$$

For a single boson, of frequency ω, we shall see later that the Hamiltonian is

$$H = \hbar\omega(a^{+}a + {}^{1}/_{2}) \tag{1.B.24}$$

where the zero point energy ${}^{1}/_{2}\hbar\omega$ can be ignored.

Thus for H given in eq. (1.B.24)

$$\frac{da_H}{dt} = -i\omega a_H \tag{1.B.25}$$

$$\frac{da_H^+}{dt} = i\omega a_H^+ \tag{1.B.26}$$

and the solutions are

$$a_H(t) = U^+(t,0)a_S U(t,0) = a_S e^{-i\omega t} \tag{1.B.27a}$$

$$a_H^+(t) = U^+(t,0)a_S^+ U(t,0) = a_S^+ e^{i\omega t} \tag{1.B.27b}$$

where $U(t,0) = e^{-i\omega a^+ a}$ $\quad U^+(t,0) = e^{i\omega a^+ a}$

The state $|n\rangle$ can be generated by using eq. (1.B.18) and applying the operator a^+ to the ground or vacuum state $|0\rangle$ n times. Hence

$$|n\rangle = \frac{a^{+n}}{\sqrt{n!}} |0\rangle \tag{1.B.28}$$

From the general theory, the orthonormality relations are

$$\langle n'|n''\rangle = \delta_{n'n''} \tag{1.B.29}$$

i.e. boson eigenstates are orthogonal

and the completeness relation is

$$\sum_{n=0}^{\infty} |n\rangle\langle n| = I \tag{1.B.30}$$

i.e. boson eigenstates are complete.

Since the norm of these vectors is finite, they form a complete set of basis vectors for a Hilbert space.

The energy eigenvalues are

$$E_n = \hbar\omega(n + {}^1/_2) \tag{1.B.31}$$

where $n = 0, 1, 2, \ldots, \infty$.

Classically any positive value of energy may be obtained when energy is measured but quantum mechanically only discrete values may be obtained. In the limit of large n (n is called a quantum number) the discrete character of eq. (1.B.31) is not noticeable and the quantum result becomes the classical result. Since $\hbar \hat{=} 10^{-27}$ erg. sec., $\hbar\omega$ is small up to optical frequencies where quantum features become important. Since we will be working in the optical region it is obvious that a quantum approach is vital.

1.B.5. Normal ordering (see also Chapter VIII, Section 7)

In order to enable solution of problems involving non-conservative systems, i.e. where $H = H(t)$, without use of cumbersome iterated solution, a powerful operator technique is employed. This is known as normal ordering. In any simple product term of creation and annihilation operators, the product is a normal one if all annihilation operators appear to the right of all creation operators, e.g. if ℓ and m are integers

$$a^{+\ell}a^{m} \text{ is a normal product} \qquad (1.B.32a)$$

$$a^{m}a^{+\ell} \text{ is not.} \qquad (1.B.32b)$$

1.B.6. Quantisation of the radiation field (a brief discussion of the remaining sections of this chapter is given in ref. 36)

The quantisation of the electromagnetic (e.m.) field by Dirac [37] enabled the synthesis of the two aspects of radiation and also explained them in a unified way. The first aspect, the wave-like properties of light radiation, is apparent in interference and diffraction experiments and the second aspect, the particle-like properties, is apparent when the radiation is absorbed or emitted by atoms. In Louisell ([35] pp. 149-153) it is shown how the e.m. field is quantised in a cavity.

This is done by showing that the classical radiation field in a vacuum is equivalent to an infinite set of harmonic oscillators suggesting that the radiation field should be quantised in the same way as a harmonic oscillator.

The radiation field in a source-free cavity, i.e. in a vacuum, may be described classically in m.k.s. units by the vector potential $\underline{A}(\underline{r},t)$ which obeys the wave equation

$$\underline{\nabla}^2\underline{A}(\underline{r},t) = \frac{1}{C^2}\frac{\partial^2\underline{A}(\underline{r},t)}{\partial t^2} \qquad (1.B.33)$$

The Coulomb guage in which

$$\underline{\nabla}.\underline{A} = 0 \qquad (1.B.34a)$$

and the scalar potential

$$V = 0 \qquad (1.B.34b)$$

is assumed.

The electric and magnetic fields are

$$\underline{E}(\underline{r},t) = \frac{\partial A}{\partial t} \qquad (1.B.35)$$

$$\underline{B}(\underline{r},t) = \underline{\nabla} \times \underline{A} \qquad (1.B.36)$$

We may expand $\underline{A}$ in the form

$$\underline{A}(\underline{r},t) = \frac{1}{\sqrt{E_0}} \quad q_\ell(t)\underline{u}_\ell(\underline{r}) \qquad (1.B.37)$$

where
$$\frac{d^2q}{dt^2} + \omega_\ell^2 q_\ell = 0 \qquad (1.B.38)$$

$$\nabla^2\underline{u}_\ell(\underline{r}) = \left(\frac{\omega_\ell}{C}\right)^2 \underline{u}_\ell(\underline{r}) = 0 \qquad (1.B.39)$$

When we solve eq. (1.B.39) in a cavity with perfectly conducting walls, we obtain a set of normal modes. The resulting equations for the fields will then represent standing waves. But we still find it convenient to represent the fields in terms of plane travelling waves, and so we shall write the vector potential as a linear superposition of plane waves in the form

$$\underline{A}(\underline{r},t) = \sum_{\ell} \sum_{\sigma=1}^{2} \sqrt{\frac{\hbar}{2\omega_\ell \epsilon_0 \tau}}\, \hat{e}_{\ell\sigma}\{a_{\ell\sigma} \exp\left[i(\underline{k}_\ell.\underline{r} - \omega_\ell t)\right] + a^{+}_{\ell\sigma}\exp\left[-i(\underline{k}_\ell.\underline{r} - \omega_\ell t)\right] \tag{1.B.40}$$

In the above expression τ is the volume of the cavity, which is assumed to be cubic, i.e. $\tau = L^3$, although its shape will be seen to have no effect on the derisations. For radiation in FREE SPACE we will let $\tau \to \infty$ after the calculations are complete.

In the expression for $\underline{A}(\underline{r},t)$, $\hat{e}_{\ell\sigma}$ and the numbers $a_{\ell\sigma}$ and $a_{\ell\sigma}^{+}$ are constants. $\hat{e}_{\ell\sigma}$ will also be assumed to be real throughout our calculations. The vector $\underline{k}_\ell$ is the propogation constant and since each term in the series (1.B.40) must satisfy the wave equation

$$\underline{k}_\ell^2 = \frac{\omega_\ell^2}{c^2} \tag{1.B.41}$$

From the Conlomb guage condition $\underline{\nabla}.\underline{A} = 0$, we obtain the transversatility condition

$$\hat{e}_{\ell\sigma}.\underline{k}_\ell = 0 \tag{1.B.42}$$

showing that $\underline{E} = -\frac{\partial A}{\partial t}$ and $\underline{A}$ are transverse to the direction of propagation in absence of sources.

Vectors $\hat{e}_{\ell 1}$ and $\hat{e}_{\ell 2}$ are UNIT vectors specifying the polarisation of the plane wave. Each is INDEPENDENT of the other and thus the total field in (1.B.40) is summed over BOTH polarisations. They are chosen to be perpendicular for convenience so that

$$\hat{e}_{\ell 1}\cdot\hat{e}_{\ell 2} = 0 \tag{1.B.43}$$

We can also define a unit vector $\hat{k}_\ell$ by

$$\hat{k}_\ell = \frac{\underline{k}_\ell}{|\underline{k}_\ell|} \tag{1.B.44}$$

Thus eqs. (1.B.43) and (1.B.44) become

$$\hat{e}_{\ell\sigma}\cdot\hat{e}_{\ell\sigma'} = \delta_{\sigma\sigma'} \qquad \sigma,\sigma' = 1,2 \tag{1.B.45a}$$

$$\hat{e}_{\ell\sigma}\cdot\hat{k}_\ell = 0 \tag{1.B.45b}$$

In order to make the modes DISCRETE it is convenient to require the vector potential to satisfy periodic boundary conditions on opposite faces of the cavity. Thus if $\hat{i}$, $\hat{j}$ and $\hat{k}$ are 3 unit vectors along the cube edges, the position vector is $\underline{r} = x\hat{i} + y\hat{j} + z\hat{k}$ and the propagation vector is $\underline{k}_\ell = k_{\ell_x}\hat{i} + k_{\ell_y}\hat{j} + k_{\ell_z}\hat{k}$ then as periodic boundary conditions require that

$$\underline{A}(\underline{r} + L\hat{i}, t) = \underline{A}(\underline{r} + L\hat{j}, t) = A(\underline{r} + L\hat{k}, t) \tag{1.B.46}$$

they are satisfied if

$$\underline{k} = \frac{2\pi}{L}(\ell_1\hat{i} + \ell_2\hat{j} + \ell_3\hat{k}) \tag{1.B.47}$$

where ℓ_1, ℓ_2 and ℓ_3 are integers from $-\infty \rightarrow +\infty$.
Thus the propagation constants $k_{\ell x} = \frac{2\pi}{L}\ell_1$ etc. are restricted to a discrete set of infinite values, and

$$\sum_\ell = \sum_{\ell_1=-\infty}^{\infty}\sum_{\ell_2=-\infty}^{\infty}\sum_{\ell_3=-\infty}^{\infty} \tag{1.B.48}$$

The result of this is that the field can now be described by a COUNTABLY infinite set of variables whereas previously the electric and magnetic fields were determined by the values A_x, A_y, A_z at each point at time t and these values were UNCOUNTABLY infinite.

For every triplet (ℓ_1,ℓ_2,ℓ_3) there are 2 travelling modes - one for each polarisation σ according to eq. (1.B.40) so that $(\ell_1,\ell_2,\ell_3,\sigma)$ signifies a mode of a given polarisation. There are also forward and backward modes, since if we let $(\ell_1,\ell_2,\ell_3) \rightarrow (-\ell_1,-\ell_2,-\ell_3)$

then $$\underline{k}_{-\ell} = -\underline{k}_{\ell} \qquad (1.B.49)$$

and $$\omega_\ell \equiv \omega_{-\ell} \qquad (1.B.50)$$

It is obvious that $\underline{A}(\underline{r},t)$ is real since $\hat{e}_{\ell\sigma}$ is real

and $$a_{\ell\sigma} \exp\left[i(\underline{k}_\ell.\underline{r} - \omega_\ell t)\right] = \{a_{\ell\sigma} + \exp\left[-i(\underline{k}_\ell.\underline{r} - \omega_\ell t)\right]\}^* \qquad (1.B.51)$$

If we now let

$$a_{\ell\sigma}(t) = a_{\ell\sigma}e^{-i\omega_\ell t} \qquad a^+_{\ell\sigma}(t) = a^+_{\ell\sigma}\, e^{i\omega_\ell t} \qquad (1.B.52)$$

and $$\underline{u}_{\ell\sigma}(\underline{r}) = \frac{\hat{e}_{\ell\sigma}\exp(i\underline{k}_\ell.\underline{r})}{\sqrt{\tau}} \qquad \underline{u}^*_{\ell\sigma}(\underline{r}) = \frac{\hat{e}_{\ell\sigma}\exp(-\underline{k}_\ell.\underline{r})}{\sqrt{\tau}} \qquad (1.B.53)$$

where the latter 2 variables satisfy the orthonormality relations

$$\int_{\text{cavity}} \underline{u}^*_{\ell\sigma}(\underline{r}).\underline{u}_{\ell'\sigma'}(\underline{r})d\tau = \delta_{\ell\ell'}\delta_{\sigma\sigma'} \qquad (1.B.54)$$

then we may use the following expressions for variables $a_{\ell\sigma}$ and $a^+_{\ell\sigma}$ in terms of $p_{\ell\sigma}$ (momentum) and $q_{\ell\sigma}$ (co-ordinate) to describe the field

$$a_{\ell\sigma} = \frac{1}{\sqrt{2\hbar\omega_\ell}} \qquad (\omega_\ell q_{\ell\sigma} + ip_{\ell\sigma}) \qquad (1.B.55a)$$

$$a^{+}_{\ell\sigma} = \frac{1}{\sqrt{2\hbar\omega_\ell}} \qquad (\omega_\ell q_{\ell\sigma} - ip_{\ell\sigma}) \tag{1.B.55b}$$

Thus $\underline{A}(\underline{r},t)$ and $\underline{E} = -\frac{\partial \underline{A}}{\partial t}$ are now expressible in terms of <u>canonically conjugate variables</u>, and the electric field is

$$\underline{E}(\underline{r},t) = i \sum_{\ell,\sigma} \sqrt{\frac{\hbar\omega_\ell}{2\epsilon_0\tau}}\, \hat{e}_{\ell\sigma} \{a_{\ell\sigma}(t)e^{i\underline{k}_\ell\cdot\underline{r}} - a^{+}_{\ell\sigma}(t)e^{-i\underline{k}_\ell\cdot\underline{r}}\} \tag{1.B.56}$$

and since the magnetic field $\underline{H} = \frac{\nabla \times \underline{A}}{\mu_0}$

$$\underline{H}(\underline{r},t) = \frac{-1}{c\mu_0} \sum_{\ell,\sigma} \sqrt{\frac{\hbar\omega_\ell}{2\epsilon_0\tau}}\, (\hat{e}_{\ell\sigma} \times \hat{k}_\ell)\{a_{\ell\sigma}(t)e^{i\underline{k}_\ell\cdot\underline{r}} - a^{+}_{\ell\sigma}(t)e^{-i\underline{k}_\ell\cdot\underline{r}}\} \tag{1.B.57a}$$

where

$$\nabla \times \left[\hat{e}_{\ell\sigma}e^{\pm i\underline{k}_\ell\cdot\underline{r}}\right] = \mp i(\hat{e}_{\ell\sigma} \times \underline{k}_\ell)e^{\pm i\underline{k}_\ell\cdot\underline{r}} \equiv \mp \frac{i\omega_\ell}{c} (\hat{e}_{\ell\sigma} \times \hat{k}_\ell)e^{\pm i\underline{k}_\ell\cdot\underline{r}} \tag{1.B.57b}$$

(N.B. so far $a_{\ell\sigma}$ and $a^{+}_{\ell\sigma}$ commute classically though later we identify them as boson operators.)

At this point we will convert the formulas into the units we shall use in the rest of the thesis, namely those used also by Lehmberg except that $\hbar$ will not be put equal to 1. For convenience we will show how conversions may be made between the units used in our main references with the help of ref. [38], pp. 729-743.

Table 1.B.1

CONVERSION TABLE

Rationalised M.K.S.	Unrationalised e.m.u. and e.s.u.		Unrationalised mixed Gaussian c.g.s. system
LOUISELL[35]	LEHMBERG ($\hbar = 1$)[1,2]	THIS THESIS	MOLLOW[7,9] etc
$4\pi\epsilon_o\lvert\underline{E}\rvert^2$	$\lvert\underline{E}\rvert^2$	$\lvert\underline{E}\rvert^2$	$4\pi\lvert\underline{E}\rvert^2$
$4\pi/\mu_o\,\underline{B}^2 = 4\pi\mu_o\lvert\underline{H}\rvert^2$	$\lvert\underline{B}\rvert^2$	$\lvert\underline{B}\rvert^2$	$4\pi\lvert\underline{B}\rvert^2$
$ia_{l\sigma}$	a_q	$a_{l\sigma}$	
$-ia^+_{l\sigma}$	a^+_q	$a^+_{l\sigma}$	
$d\tau$	d^3r	d^3r	
τ	V	V	
$\underline{E} = i\sum_{l,\sigma}\sqrt{\frac{\hbar\omega_l}{2\epsilon_o\tau}}\,\hat{e}_{l\sigma}a_{l\sigma}(t)e^{i\underline{k}_l\cdot\underline{r}}$ + h.c.	$\underline{E} = \sum_q\sqrt{\frac{2\pi\omega_q}{V}}\,\hat{e}_q a_q e^{i\underline{k}_q\cdot\underline{r}}$ + h.c.	$\underline{E} = \sum_{l,\sigma}\sqrt{\frac{2\pi\hbar\omega_l}{V}}\,\hat{e}_{l\sigma}a_{l\sigma}e^{i\underline{k}_l\cdot\underline{r}}$ + h.c.	
$\underline{B} = \frac{1}{c}\sum_{l,\sigma}\sqrt{\frac{\hbar\omega_l}{2\epsilon_o\tau}}\,\hat{k}_l\times\hat{e}_{l\sigma}a_{l\sigma}(t)\times e^{i\underline{k}_l\cdot\underline{r}}$ + h.c.	$\underline{B} = \sum_q\sqrt{\frac{2\pi\omega_q}{V}}\,\hat{k}_l\times\hat{e}_q a_q e^{i\underline{k}_q\cdot\underline{r}}$ + h.c.	$\underline{B} = \sum_{l,\sigma}\sqrt{\frac{2\pi\hbar\omega_l}{V}}\,\hat{k}_l\times\hat{e}_{l\sigma}a_{l\sigma}e^{i\underline{k}_l\cdot\underline{r}}$ + h.c.	
ϵ_o replaced by $1/4$ →		ϵ_o replaced by 1 →	
μ_o replaced by 4		μ_o replaced by 1	
$H = \frac{1}{2}\int_{\text{cavity}}(\epsilon_o\underline{E}^2 + \mu_o\underline{H}^2)d$	$H = 1/8\int(\lvert\underline{E}\rvert^2 + \lvert\underline{B}\rvert^2)d^3r$		

Thus, using the following plane-wave representation for $\underline{E}$ and $\underline{B}$:

$$\underline{E}(\underline{r},t) = \sum_{\ell\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma} \quad \{a_{\ell\sigma} e^{i\underline{k}_\ell\cdot\underline{r}} + a^+_{\ell\sigma} e^{-i\underline{k}_\ell\cdot\underline{r}}\} \tag{1.B.58}$$

$$\underline{B}(\underline{r},t) = \sum_{\ell\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{k}_\ell \times \hat{e}_{\ell\sigma} \quad \{a_{\ell\sigma} e^{i\underline{k}_\ell\cdot\underline{r}} + a^+_{\ell\sigma} e^{-i\underline{k}_\ell\cdot\underline{r}}\} \tag{1.B.59}$$

We can derive the Hamiltonian for the field in the cavity, i.e.

$$H_R = \frac{1}{8\pi} \int_{\text{cavity}} (|\underline{E}|^2 + |\underline{B}|^2) d^3r \tag{1.B.60}$$

and following the procedure outlined in Appendix A of Louisell [35] we finally obtain

$$\left.\begin{aligned} H_R &= \frac{1}{2} \sum_{\ell,\sigma} \hbar\omega_\ell (a_{\ell\sigma} a^+_{\ell\sigma} + a^+_{\ell\sigma} a_{\ell\sigma}) \\ &= \frac{1}{2} \sum_{\ell,\sigma} (p^2_{\ell\sigma} + \omega^2_\ell q^2_{\ell\sigma}) \end{aligned} \quad \begin{array}{l}\text{(the order of a} \\ \text{and a}^+ \text{ being kept} \\ \text{since they will} \\ \text{later be treated} \\ \text{as non-commuting} \\ \text{operators)}\end{array} \right\} \tag{1.B.61}$$

In order to quantise the radiation field hermitian operators are associated with variables $p_{\ell\sigma}$ and $q_{\ell\sigma}$. Since photons are bosons we may postulate that <u>$q_{\ell\sigma}$ and $p_{\ell\sigma}$ satisfy boson commutation relations.</u> Quantisation is necessary to show the particle nature of light. In terms of non-hermitian operators $a_{\ell\sigma}$ and $a^+_{\ell\sigma}$ these relations are

$$[a_{\ell\sigma}, a^+_{\ell'\sigma'}] = \delta_{\ell,\ell'}\, \delta_{\sigma,\sigma'} \tag{1.B.62}$$

i.e. they commute for different oscillators because they are independent,

also $$[a_{\ell\sigma}, a_{\ell'\sigma'}] = 0 = [a^+_{\ell\sigma}, a^+_{\ell'\sigma'}] \tag{1.B.63}$$

Hence

$$H_R = \sum_{\ell\sigma} \hbar\omega_\ell a^+_{\ell\sigma} a_{\ell\sigma} + \frac{1}{2}\hbar\omega_\ell \tag{1.B.64}$$

but the zero-point energy can be neglected since one can change the level from which the energy is measured. Even if it is left in it would not affect the Heisenberg equations of motion. Thus we use

$$H_R = \sum_{\ell,\sigma} \hbar\omega_\ell a^+_{\ell\sigma} a_{\ell\sigma} \tag{1.B.65}$$

Photons are bosons and so satisfy the earlier results given for bosons except that now instead of one oscillator there are an infinite number of independent field oscillators.

Since each cavity mode is <u>independent</u>, a <u>complete</u> set of state vectors may be written as a <u>simple product</u> of state vectors for each mode; i.e. a state vector for the radiation field may be written as

$$|n_1> |n_2> \ldots |n_\infty> = |n_1, n, \ldots, n_\infty> \tag{1.B.66}$$

where each subscript 1, 2, ... stands for the quartet of integers $(\ell_1, \ell_2, \ell_3, \sigma)$. Also the state vector for an assembly of <u>non-interacting</u> bosons must be symmetric under the interchange of any 2 of the bosons.

The effect of $a_{\ell\sigma}$ and $a^+_{\ell\sigma}$ on the state vectors of eq. (1.B.66) is given by

$$\left.\begin{aligned}
a^+_{\ell\sigma}|\ldots, n_{\ell\sigma}, \ldots> &= \sqrt{n_{\ell\sigma}+1}\,|\ldots, n_{\ell\sigma}+1, \ldots> \\
a_{\ell\sigma}|\ldots, n_{\ell\sigma}, \ldots> &= \sqrt{n_{\ell\sigma}}\,|\ldots, n_{\ell\sigma}-1, \ldots> \\
a_{\ell\sigma}|\ldots, 0, \ldots> &= 0 \\
N_{\ell\sigma}|\ldots, n_{\ell\sigma}, \ldots> &= n_{\ell\sigma}|\ldots, n_{\ell\sigma}, \ldots>
\end{aligned}\right\} \tag{1.B.67}$$

and with this choice, these state vectors are normalised to unity. As in the case of the single oscillator, these operators are in the

Schrödinger picture but may be generalised to the Heisenberg picture; e.g. Heisenberg equations of motion for $a_{\ell\gamma}(t)$ are

$$
\begin{aligned}
i\hbar \frac{da_{\ell\sigma}(t)}{dt} &= \left[a_{\ell\sigma}(t), H_H\right] \\
&= -i\omega_\ell a_{\ell\sigma}(t) \quad \text{for } H_H = H_R \text{ the radiation field in a vacuum}
\end{aligned}
\tag{1.B.68}
$$

also $$a_{\ell\sigma}(t) = U^{+}(t,t_0)a_{\ell\sigma}\, U(t,t_0) \tag{1.B.69}$$

We shall consider what happens in the free space limit $L \to \infty$. In this limit when we sum over discrete values of ℓ (ℓ_1,ℓ_2,ℓ_3) the values ℓ_1/L, ℓ_2/L, ℓ_3/L became practically continuous and we may replace sums by integrals so that

$$\frac{1}{L^3}\sum_{\ell} (\qquad) \underset{L\to\infty}{\longrightarrow} \frac{1}{(2\pi)^3}\iiint_{-\infty}^{\infty} dk_x dk_y dk_z (\qquad) \tag{1.B.70}$$

since $k_x = 2\pi\ell_1/L$ etc.,

and if we transform (k_x, k_y, k_z) from rectangular to spherical polar co-ordinates by means of

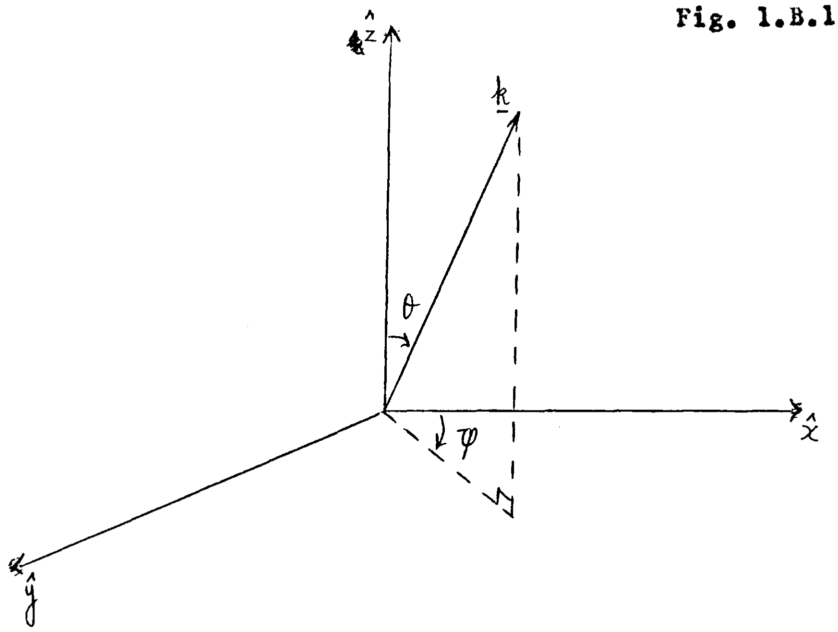
$$\underline{k} = k\,(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta) \tag{1.B.71}$$

so that the element of volume in $\underline{k}$ space is

$$dk_x dk_y dk_z = k^2 dk \sin\theta\, d\theta d\phi \equiv k^2 dk d\Omega_{\hat{k}} \tag{1.B.72}$$

where $\Omega_{\hat{k}}$ is the element of solid angle about direction of propagation.

Fig. 1.B.1



Then

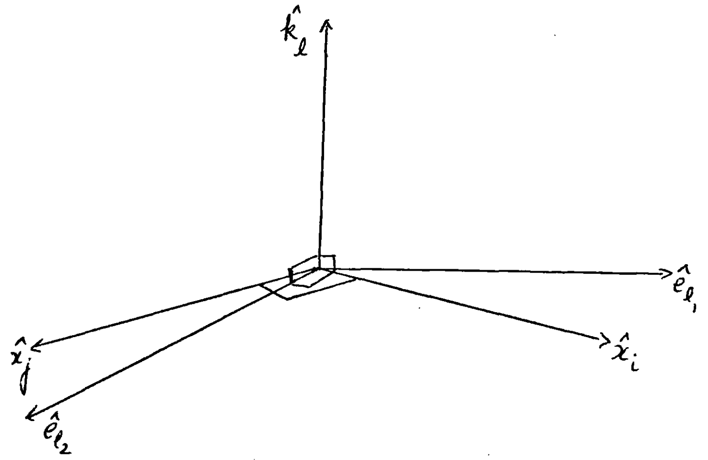
$$\frac{1}{L^3}\sum_{\ell}(\quad) \xrightarrow[L\to\infty]{} \frac{1}{(2\pi)^3}\int_0^\infty k^2 dk \int_0^\pi \sin\theta d\theta \int_0^{2\pi} d\psi(\quad)$$

$$\rightarrow \frac{1}{(2\pi)^3}\int_0^\infty k^2 dk \int_0^{4\pi} d\Omega_{\hat{k}}(\quad)$$

$$\rightarrow \frac{1}{(2\pi c)^3}\int_0^\infty \omega^2 d\omega \int_0^{4\pi} d\Omega_{\hat{k}}(\quad) \qquad (1.B.73)$$

The summation over polarisation index σ will be done as follows,

$\hat{k}_\ell$, $\hat{e}_{\ell 1}$, $\hat{e}_{\ell 2}$ are all mutually perpendicular Fig. 1.B.2



If $\hat{x}_i$ and $\hat{x}_j$ are unit vectors along x_i and x_j axes respectively then

$$(\hat{e}_{\ell,1})_i = \cos(\hat{e}_{\ell,1}.\hat{x}_i)$$
$$(\hat{e}_{\ell,2})_j = \cos(\hat{e}_{\ell,2}.\hat{x}_j) \qquad (1.B.74)$$
$$(\hat{k}^\ell)_i = \cos(\hat{k}_\ell.\hat{x}_i)$$

and by the properties of direction cosines

$$(\hat{e}_{\ell,1})_i(\hat{e}_{\ell,1})_j + (\hat{e}_{\ell,2})_i.(\hat{e}_{\ell,2})_j + (\hat{k}_\ell)_i.(\hat{k}_\ell)_j = \delta_{ij}$$

or

$$\sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma})_i.(\hat{e}_{\ell\sigma})_j = \delta_{ij} - (\hat{k}_\ell)_i.(\hat{k}_\ell)_j \qquad (1.B.75)$$

$$= \delta_{ij} - \frac{(\underline{k}_{\ell i}).(\underline{k}_{\ell j})}{k_\ell^2}$$

1.B.7. Interaction of radiation with matter

In the Dirac theory of radiation an atom (A) and the radiation field (R) with which it interacts are considered as a single system and the energy of this system is represented by:

(1) the energy of the atom alone, H_A

(2) the energy of the radiation field alone, H_R

(3) a small term = the coupling energy between atom and field, V_{AR}, since atom and field affect one another.

The non-relativistic Hamiltonian for a 1-electron atom in the presence of a radiation field, is

$$H = \frac{1}{2m}(\underline{p} - e\underline{A})^2 + eV(\underline{r}) + H_R \qquad (1.B.76)$$

where electron spin is neglected since energies are not considered to be sufficiently large enough for relativistic effects to be important. In ref. 40 , p. 176 it is said that:

> "Even for heavy atoms the energy of the K-shell is still $\ll mc^2$ and the relativistic correction, though appreciable for Ur and X-rays emitted in transitions to the K-shell, does not seriously affect the results."

We shall anyway not be considering heavy atoms here.

Equation (1.B.76) also applies to single valency atoms such as alkali metals, e.g. potassium, which we consider in Chapter V, as well as to hydrogen which we consider in Chapter VI.

e   is the charge on the electron

m   is the mass of the electron

$V(\underline{r})$ is the potential in which the atom is situated at position $\underline{r}$ (i.e. Conlomb interaction between electron and nucleus)

$\underline{p}$ is the electron momentum

$\underline{A}$ is the vector potential describing the radiation field.

In the Coulomb guage $\nabla.\underline{A} = 0$, hence

$$H = H_A + H_R + V_{AR}^{(1)} + V_{AR}^{(2)} \tag{1.B.77a}$$

$$= H_0 \qquad + V_{AR}^{(1)} + V_{AR}^{(2)} \tag{1.B.77b}$$

where $$H_A = \frac{\underline{p}^2}{2m} + eV(\underline{r}) \tag{1.B.78a}$$

$$H_R = \frac{1}{8\pi} \int_{\text{cavity}} (|\underline{E}|^2 + |\underline{B}|^2) d^3r \tag{1.B.78b}$$

$$= \sum_{\ell,\sigma} \hbar\omega_\ell \, (a_{\ell\sigma}^\dagger a_{\ell\sigma} + \frac{1}{2})$$

when the radiation field is quantised and source-free.

$$H_0 = H_A + H_R \tag{1.B.78c}$$

is the unperturbed Hamiltonian.

$$V_{AR}^{(1)} = \frac{-e}{m} \underline{A}.\underline{p} \tag{1.B.79}$$

represents interaction between electron $\underline{p}$ and radiation field $\underline{A}$ and is $\ll H_0$. It is of first order in coupling constant e.

$$V_{AR}^{(2)} = \frac{e^2}{2m}\underline{A}^2 \qquad (1.B.80)$$

represents mutual interaction between different radiation oscillators of the radiation field through coupling of the electron to the field. $V_{AR}^{(1)} >> V_{AR}^{(2)}$ is the only term of importance in our applications. If we use mixed Gaussian units the denominator of $V_{AR}^{(2)}$ is mc^2 and so is very large, making it obvious why $V_{AR}^{(2)}$ is so small. $V_{AR}^{(2)}$ is important in dispersion and Compton scattering.

The Hamiltonian is in the Schrödinger picture and so we must use $\underline{A}$ in the Schrödinger picture also and then solve the Schrödinger equation of motion. This cannot be solved exactly and a perturbation approach is used which results in an infinite set of coupled equations for the probability amplitudes. Various approximation techniques can then be used to solve these.

The techniques used depend on the size of the interaction times t involved compared with the coupling constant, g, in $V_{AR}^{(1)}$. N.B. in (1.B.79) $g \propto e$.

(1) In the case of (a) <u>absorption</u> of radiation by atoms
and (b) <u>spontaneous emission</u> by excited atoms
the interaction times, $\underline{t << g^{-1}}$
and a solution involving <u>perturbation expansion</u> in powers of
$\gamma t << 1$ is adequate.

(2) In the case of the theory of <u>natural line width</u>
interaction times, $\underline{t >> g^{-1}}$
and development in powers of $gt > 1$ will converge too slowly to be useful. In this case <u>Wigner-Weisskopf[41] approximation</u> (W-W approximation) can be used to obtain approximate results.

N.B. In the theory of emission and absorption of radiation by atoms their energy levels can be assumed infinitely sharp even though it is known from experiment that emission and absorption lines have finite width owing to the reaction of the radiation field on the atom. This reaction is caused since the emitting atom generates a radiation field which reacts back on the atom causing the emitted spectral line to have a natural line width. It is with this reaction which we shall be concerned and so this reason we shall not be using a perturbation approach.

Time-dependent perturbation theory (P.T.) is valid only for times t short enough that initial state does not change during the course of the interaction. An initially excited atom is bound to change its state and so in order to measure the energy-level separation a sufficient time must be allowed to allow its state to change significantly (according to Heisenberg's uncertainty principle). Perturbation theory is therefore inadequate and one alternative is the W-W approximation.

As an alternative to the W-W method which involves solving the Schrödinger equations of motion, we use the method of Lehmberg which involves solving Heisenberg equations of motion using the Hamiltonian in the Heisenberg representation where $a_{\ell\sigma}$ and $a^{\dagger}_{\ell\sigma}$ are time-dependent. We are particularly interested in radiation damping which results in atoms having a finite line width and so it is obvious that perturbation theory (P.T.) is invalid here as stated above. The initial state will change during the course of the interaction and this is another reason for not using P.T. In Mollow and Miller's paper they point out that 1st. order P.T. only holds good when the density operator for the atom corresponds to a PURE state throughout the interaction. This is just another way of saying that the initial atomic state must not change. We shall explain why this is so later.

1.B.8. Phenomenological model for loss mechanism

When an atom (A) is coupled to a radiation field (R) it spontaneously decays with a finite lifetime which is responsible for the natural line width of the atom. This decay can be visualised as a single quantum system (the atom) coupled to a large number of harmonic oscillators (the radiation field) into which the energy of the atom goes and is thus dissipated. We know that this process involves a loss mechanism (L) and so in order to find the quantitative effect of spontaneous emission a phenomenological model for L must be found. It will be equivalent to, though not the same as, the single atom (A) coupled to the radiation field (R) discussed in Section 1.B.7.

Non-rigorous treatment

The model for L we shall use is based on the model used for the natural theory of line width and is that also used by Senitzky.[42]

The Hamiltonian for a single mode, frequency ω_0, of the radiation field in a cavity is

$$H_A = \hbar\omega_0 A^\dagger A \tag{1.B.81}$$

(see eq. (1.B.61) where $\omega_\ell = \omega_0$ and σ is not specified).

(This single mode will later be taken to represent the atom, hence subscript A.) The solution of the Heisenberg equation of motion for A(t)

$$\frac{dA}{dt} = \frac{1}{i\hbar}\left[A, H_A\right] = -i\omega_0 A \tag{1.B.82}$$

is $$A(t) = Ae^{-i\omega_0 t} \tag{1.B.83}$$

If we put in the cavity a phenomenological loss term by analogy with a circuit resistance we then obtain damped solutions of the form

$$A(t) = Ae^{-i\omega_0 t - \gamma/2t} \tag{1.B.84a}$$

$$A^\dagger(t) = A^\dagger e^{i\omega_0 t - \gamma/2t} \tag{1.B.84b}$$

by replacing ω_0 by $\omega_0 + i\gamma/2$. But the commutator of A and $A^\dagger$ is then

$$\left[A(t),A^\dagger(t)\right] = e^{-\gamma t} \tag{1.B.85}$$

since $[A,A^\dagger] = 1$ since A and $A^\dagger$ are boson operators. Thus for times $t \ll \gamma^{-1}$ all is well but when $t \gg \gamma^{-1}$ the commutator in (1.B.85) approaches zero, violating the uncertainty principle. Since we are interested in times $t \gg \gamma^{-1}$, an ordinary damping term is not an adequate phenomenological model for loss since the Heisenberg operator equations should be identical in form to the classical equations of motion. This model only accounts for the action of the cavity on the loss but we require one which also allows for reaction of the loss back on the cavity.

N.B. If the life-time of the excited atom is very large, i.e. $\gamma^{-1} \gg t$ (the interaction time) we could account for emission and absorption by 1st. order P.T. neglecting reaction of radiation field back on the atom but when the lifetime is short $\gamma^{-1} \ll t$ we must take into account the reaction of the field on the atom which results in the natural line width.

In order to describe a loss cavity, we now let a single cavity mode represent the atom and let it be coupled to elastic vibrations in a dielectric material. These vibrations are expanded in normal modes and their energy is then equivalent to a large number of "elastic" oscillators which are then quantised as were the em oscillators earlier. The quanta are known as PHONONS for the elastic waves and as they obey Box-Einstein statistics their Hamiltonian (cf. eq. (1.B.65) is

$$H_L = \hbar \sum_{\ell,\sigma} \omega_\ell \, a^\dagger_{\ell\sigma} a_{\ell\sigma}$$

$$= \hbar \sum_q \omega_q \, a^\dagger_q a_q \qquad (1.B.86)$$

There are about 10^{23} atoms in a solid and at the initial time all are at the same frequency $\omega_q = \omega_{q_0}$. When the atoms are coupled in a solid $\underline{\omega_q}$ spreads into a band of frequencies[1] ω_q about the initial frequency ω_{q_0}. Thus

$$\sum_q = \sum_{q=1}^{10^{23}} \qquad (1.B.87)$$

Because the frequencies are very closely spaced with density $\rho(\omega_q)$, we shall be able to replace sums over q by integrals when convenient. [cf. ref. 1, eq. (2.13) and ref. 2, eq. (11) where this is done more rigorously after the manner of Section 3, eq. (1.B.73).]

In Mollow and Miller's paper [7] the atom is said to be damped by its coupling to a (ZERO-temperature) "bath" of harmonic oscillators (L) which are taken to represent modes of the em field (R). In this way the damping mechanism represents the effect of spontaneous emission. According to Lehmberg's 1st. paper it is not necessary either for the bath to be at zero temperature or to specify the initial state of the bath at all although we do find the latter to be necessary.

To summarise, the phonons are thus equivalent to an ensemble of harmonic oscillators in thermal equilibrium with a heat bath at temperature T. We also assume that the dielectric is in a cavity with a mode of frequency $\omega \simeq \omega_q$..

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1. We shall see in Chapter VIII, Section 8 that, because of the BROAD nature of the spectrum of loss oscillators, we may use the Markoff approximation when desired. It is also important in other respects also as we shall see.

Phenomenological model for loss mechanism - schematically represented

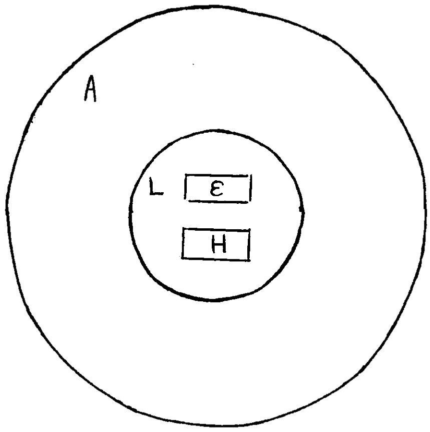


Fig. 1.B.3

A - cavity with mode of frequency $\omega_0 \simeq \omega_q$ representing the atom. In fact a radiation field is emitted by the atom and this field then reacts back on the atom. It is the mode of this field which represents the atom.

L - dielectric material containing elastic vibrations whose energy is quantised in PHONONS. This represents the radiation field. In fact the damping reservoir L is composed of phonons equivalent to ε in thermal equilibrium with H.

ε - ensemble of harmonic oscillators.

H - heat bath at temperature T.

L is referred to as R, the radiation field,[1] in Lehmberg [1], [2], and also elsewhere in this thesis when we refer to it as the e.m. radiation field. At this point it is referred to as L only to make clear its significance as a loss mechanism. L is referred to as B, the bath, in Mollow's papers [7], [9].

---

1. According to von Foerster [42], normal modes of the e.m. radiation field in a macroscopic cavity are sufficiently numerous and have a sufficiently dense spectrum that they serve as a heat bath for the atom.

A is referred to as S in Lehmberg [1], [2] and later on in this thesis when it is referred to as the atom cavity system.

Before the two systems L and A areccoupled, the Hamiltonian for the loss oscillators (phonons) and the cavity mode (photons) as

$$\left.\begin{aligned} H_0 &= H_A + H_L \\ &= \hbar\omega_0 A^\dagger A + \hbar \sum_q \omega_q a_q^+ \quad a_q \end{aligned}\right\} \tag{1.B.88}$$

where $[A,A^\dagger] = 1$,[1] $[A,A] = [A^\dagger,A^\dagger] = 0$ and the A's and a_q's all commute with each other. Here a_q and $a_q^\dagger$ are PHONON annihilation and creation operators respectively. We assume that H_0 describes the state of the whole system before $t = 0$ when the cavity mode (e.m. field), (A) is uncoupled from the lattice modes (elastic field, L).

---

1. In fact for an atom with two levels this would be an anti-commutator if, for levels $|i\rangle$ and $|j\rangle$, where $j > i$, the operators A and $A^\dagger$ are identified as

$$\left.\begin{aligned} A &= |i\rangle\langle j| = P_{ij} \\ A^\dagger &= |j\rangle\langle i| = P_{ji} \quad \text{(see Lehmberg |1| for definition of P's)} \end{aligned}\right\} \tag{1.B.89}$$

and then $[A,A^\dagger] = P_{ii} - P_{jj} \neq 1$, whereas the anti-commutator $[A,A^\dagger]_+ = P_{ii} + P_{jj} \neq 1$.

If A and $A^\dagger$ are defined by eq. (1.B.89) then they are pseudo-spin operators for the atom and have entirely different properties from boson operators (cf. pp. 81-84, ref. [35]) and it is because fermions obey the Pauli exclusion principle that they do not satisfy the relation $[A,A^\dagger] = 1$.

Fermion operators also satisfy

$$A^{\dagger 2} = A^2 = 0$$

$$\left.\begin{aligned} &\text{since} \quad A^{\dagger 2} = |j\rangle\langle i|j\rangle\langle i| = \delta_{ij}|j\rangle\langle i| = 0 \text{ for } i \neq j \\ &\text{and} \quad A^2 = |i\rangle\langle j|i\rangle\langle j| = \delta_{ji}|i\rangle\langle j| = 0 \text{ for } i \neq j \end{aligned}\right\} \tag{1.B.90}$$

Further properties of these operators are given on pages 131-134 of ref. [35].

In eq. (1.B.88) A and $A^\dagger$ are identified as boson operators for the atom since the atom is represented as a cavity mode of frequency $\omega_0 \simeq \omega_q$.

Let $\rho^{(A)}(0)$ be the density operator describing the ensemble of radiation oscillators at $t = 0$, while $\rho^{(L)}(0)$ describes the ensemble of loss oscillators. $\rho^{(L)}(0)$ is specified in Louisell, p. 257 [35] as a Boltzmann distribution, since L is assumed to be in thermal equilibrium at temperature T.

$$\rho^{(L)}(0) = \frac{e^{-\beta H_L}}{\mathrm{Tr}(e^{-\beta H_L})} \tag{1.B.91}$$

where $\beta = (KT)^{-1}$ and K is Boltzmann's constant. We shall specify $\rho^{(A)}(0)$ later.

In order for the e.m. field (A) to interchange energy with the lattice vibrations (L) there must be a coupling between the two fields e.g. for an ionic lattice this would be accomplished by charges on the ions interacting with the e.m. field, whereas in a crystal there may be dipoles that interact with the field. We shall, in fact, be considering atomic vapours in which the atomic dipoles interact with the field but, no matter what the mechanism for coupling, the simplest energy preserving Hamiltonian is

$$V_{AL} = \sum_q \hbar(g_q A a_q^\dagger + g_q^* a_q^\dagger A) \tag{1.B.91}$$

where the operators are normally ordered. (Compare eq. (1.B.91) with $V_{AR}^{(1)}$ of (1.B.79) which is of the same order.) This is the interaction Hamiltonian in the resonant form. The coupling coefficients g_q will later be assumed REAL. They are small compared with ω_0 or ω_q and will depend on the parameters involved in the actual coupling mechanism, such as charge, or dipole moments of crystal atoms (see rigorous treatment). Physically, the above phenomenological coupling terms <u>couple significantly only those phonons for which $\omega_q \simeq \omega_0$, the cavity mode frequency.</u>

Eq. (1.B.91) is hermitian, but a term of the form

$$g_q a_q^{\dagger} A^{\dagger} + g_q^{*} A a_q \tag{1.B.92}$$

is also hermitian and of the same order in the strength of coupling. It is not included since its effect would be small. When there is no coupling, $g_q = 0$ and, from eq. (1.B.88)

$$\begin{aligned} \frac{dA(t)}{dt} &= \frac{1}{i\hbar}\left[A, H_0\right] = -i\omega_0 A \\ \frac{da_q(t)}{dt} &= \frac{1}{i\hbar}\left[a_q, H_0\right] = -i\omega_q a_q \end{aligned} \tag{1.B.93}$$

hence $\qquad A(t) = Ae^{-i\omega_0 t}$

and $\qquad a_q(t) = a_q e^{-\omega_q t} \qquad$ (1.B.94)

Since $g_q << \omega_q$ [1] $\simeq \omega_0$, we expect that when the coupling is turned on A and a_q on the RHS of (1.B.94) will be only slowly varying functions of time, i.e. little altered by the small coupling terms. Thus,

$$\begin{aligned} a_q^{\dagger}(t)A^{\dagger}(t) &\simeq e^{i(\omega_0+\omega_q)t} \\ &\simeq e^{2i\omega_q t} \\ a_q^{\dagger}(t)A(t) &\simeq e^{i(\omega_q-\omega_0)t} \\ &\simeq e^{0} \\ &\simeq 1 \end{aligned}$$

I.E. terms in (1.B.91) are approximately d.c. whilst those in (1.B.92) are rapidly varying and according to von Foerster[43] only contribute to physical processes as higher order terms and can therefor be neglected. In fact the interaction will be in effect for many cycles of ω_q so that

---

1. $g_q \sim \omega_q^{1/2}$ (see eq. (1.B.99).

terms in eq. (1.B.92) will almost average to zero compared with those of (1.B.91) and so the former are neglected in order to simplify the equations. This is known as the rotating-wave approximation, the importance of which will be discussed in Chapter VIII, Section 3. When the coupling becomes stronger, i.e. when g_q is large,[1] neglecting these extra terms would not be justified.

We assume that the interaction starts at $t = 0$ and that the Hamiltonian for a single cavity mode coupled to many oscillators is approximately given by

$$H = H_0 + V_{AL}$$

$$= \hbar\omega_0 A^\dagger A + \hbar \sum_q \omega_q a_q^\dagger a_q + \hbar \sum_q (g_q A^\dagger a_q + g_q^* a_q^\dagger A) \qquad (1.B.95)$$

Table 1.B.2

Notation of main references:				
	Louisell [35]	Lehmberg [1],[2]	Mollow [7],[9]	Here
Atom	Single cavity mode (photon) Density operator ρ_f	Atomic system, S. Density operator $\rho^{(S)}$	Atom Density operator, ρ_a	A, later S Density operator $\rho^{(S)}$
Loss Mechanism	Loss oscillators (photons). Density operator ρ_L	Damping reservoir, R. Density operator $\rho^{(R)}$	Bath Density operator $\|0\rangle_{BB}\langle 0\|$	L, later R Density operator $\rho^{(R)}$

1. We shall see that $g_q = \left(\frac{2\pi\omega_q}{\hbar V}\right)^{1/2} (\hat{e}_q \cdot \underline{p})$ eq. (1.B.99), so that g_q would be large, if p, $\omega_q^{1/2}$ and $\hat{e}_q \cdot \hat{p}$ are large and $V^{1/2}$ is small, although the latter will not occur since we shall be considering free-space where $V \to \infty$.

Rigorous treatment

According to Lehmberg[2], V_{AL}, the Hamiltonian for the coupling between the e.m. field and the lattice vibrations, can be written, in the dipole approximation (see Chapter VIII, Section 2 for a discussion on this), as

$$V_{AL} = -\underline{E}(\underline{r},t).\underline{p}(A + A^{\dagger})$$

$$= -\underline{E}(\underline{r},t).\underline{d}(t) \qquad \text{in Mollow's notation}^{[9]} \qquad (1.B.96)$$

where $\underline{d}(t) = \underline{p}(A(t) + A^{\dagger}(t))$ is the dipole moment operator for the atom and $\underline{p} = \langle j|\underline{d}|i\rangle$. $\underline{E}(\underline{r},t)$ is given by (1.B.58), viz.

$$\underline{E}(\underline{r},t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma}\{a_{\ell\sigma}e^{i\underline{k}_\ell\cdot\underline{r}} + a^{\dagger}_{\ell\sigma}e^{-i\underline{k}_\ell\cdot\underline{r}}\} \qquad (1.B.97)$$

for transverse plane wave modes.

Substituting we obtain

$$V_{AL} = -\sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma}.\underline{p}\{a_{\ell\sigma}e^{i\underline{k}_\ell\cdot\underline{r}} + a^{\dagger}_{\ell\sigma}e^{-i\underline{k}_\ell\cdot\underline{r}}\}\,(A + A^{\dagger})$$

$$= -\sum_{\ell,\sigma} \hbar g_{\ell\sigma}\,\{e^{i\underline{k}_\ell\cdot\underline{r}}\,A^{\dagger}a_{\ell\sigma} + e^{-i\underline{k}_\ell\cdot\underline{r}}\,a^{\dagger}_{\ell\sigma}A$$

$$+ e^{i\underline{k}_\ell\cdot\underline{r}}\,Aa_{\ell\sigma} + e^{-i\underline{k}_\ell\cdot\underline{r}}\,A^{\dagger}a^{\dagger}_{\ell\sigma}\} \qquad (1.B.98)$$

where $g_{\ell\sigma} = \sqrt{\frac{2\pi\omega_\ell}{\hbar V}}\,\hat{e}_{\ell\sigma}.\underline{p}$ and therefore $g_{\ell\sigma}$ is real for $\underline{p}$ real, since $\hat{e}_{\ell\sigma}$ has already been assumed real. (1.B.99)

For the atom at the origin of co-ordinates in the dipole and R.W. approximations

$$V_{AL} = -\sum_{\ell,\sigma} \hbar g_{\ell\sigma}(A^{\dagger}a_{\ell\sigma} + a^{\dagger}_{\ell\sigma}A) \qquad (1.B.100)$$

which, but for the negative sign, which could be absorbed in the definition of $g_{\ell\sigma}$ by the introduction of a phase factor $e^{i\pi}$, agrees with (1.B.95). We shall see, in Chapter II, that phase factors in the Hamiltonian have no effect.

In Appendix I we shall show what happens when the Hamiltonian is used with

$$V_{AL} = -\sum_{\ell,\sigma} \hbar g_{\ell\sigma}(S\, a_{\ell\sigma} e^{i\underline{k}_\ell\cdot\underline{r}} + e^{-i\underline{k}_\ell\cdot\underline{r}}\, a^\dagger_{\ell\sigma} S) \tag{1.B.101}$$

where $S = A + A^\dagger$.

It is shown there that this more rigorous treatment only results in negligible high frequency terms, and frequency shift modifications, which we can neglect, as they are not our main concern.

Hence the complete Hamiltonian for a 2-level atom, considering radiation damping, in the dipole approximation, is

$$H = \hbar\omega_0 A^\dagger A + \frac{1}{8\pi}\int (|\underline{E}|^2 + |\underline{B}|^2)d^3r - \underline{E}(\underline{r},t).\underline{p}(A + A^\dagger) \tag{1.B.102}$$

where H, A, $A^\dagger$, $\underline{E}$, $\underline{B}$ are all time-dependent in the Heisenberg picture. This becomes, on using eqs. (1.B.58) and (1.B.59), for operators normally ordered,

$$H = \hbar\omega_0 A^\dagger A + \sum_{\ell,\sigma} \hbar\omega_\ell a^\dagger_{\ell\sigma} a_{\ell\sigma} - \sum_{\ell,\sigma} \hbar g_{\ell\sigma}(S a_{\ell\sigma} e^{i\underline{k}_\ell\cdot\underline{r}} + a^\dagger_{\ell\sigma} S e^{-i\underline{k}_\ell\cdot\underline{r}}) \tag{1.B.103}$$

when the zero point energy is discarded,

where
$$g_{\ell\sigma} = \sqrt{\frac{2\pi\omega_\ell}{\hbar V}}\, \hat{e}_{\ell\sigma}\cdot\underline{p}$$

$S = A + A^\dagger$

V is the normalisation volume

$\hat{e}_{\ell\sigma}$ is the unit polarisation vector

$\hat{k}_\ell$ is the propagation vector

$\hat{e}_{\ell\sigma}.\hat{k}_{\ell} = 0$, for the Coulomb guage, i.e. $\underline{E}$ and $\underline{B}$ transverse to direction of propagation $\hat{k}_{\ell}$.

$a_{\ell\sigma}$ and $a^{\dagger}_{\ell\sigma}$ are the annihilation and creation operators of the ℓ^{th} mode of polarisation σ, and $[a_{\ell\sigma}, a^{\dagger}_{\ell\sigma}] = \delta_{\ell\sigma}\delta_{\ell'\sigma'}$, since they are boson operators.

According to von Foerster, [43] in the simplest case, the internal structure of the atom is irrelevant and its energy levels are sufficiently widely spaced that only 2 levels are important: a ground state and an excited state. If the atomic states are $|i\rangle$, the ground state, and $|j\rangle$, the single excited state of energy $\hbar\omega_0$ then transitions between these states can be described mathematically by the operators,

$$\left.\begin{aligned} A &= |i\rangle\langle j| = P_{ij}\text{, the lowering operator for the atom} \\ A^{\dagger} &= |j\rangle\langle i| = P_{ji}\text{, the raising operator for the atom} \\ A^{\dagger}A &= |j\rangle\langle j| = P_{jj} \end{aligned}\right\} \quad (1.B.104)$$

where P_{ij} and P_{ji} are transition operators as defined in ref. [1].

$$A|j\rangle = |i\rangle\langle j|j\rangle = |i\rangle$$

$$A^{\dagger}|i\rangle = |j\rangle\langle i|i\rangle = |j\rangle$$

and this shows why A and $A^{\dagger}$ are referred to as lowering and raising operators respectively.

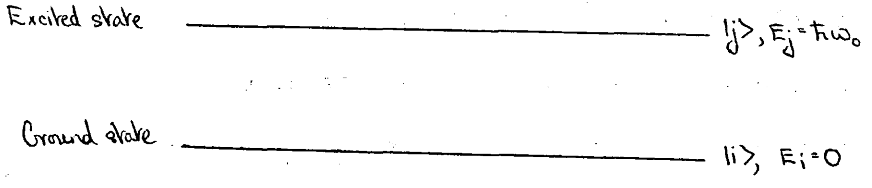


Fig. 1.B.4

Since the atomic states form a complete set, i.e.:

$$|i\rangle\langle i| + |j\rangle\langle j| = I$$

one can expand an arbitrary operator in terms of basis operators

$$|i\rangle\langle i| = AA^{\dagger}$$

and $$|j\rangle\langle j| = A^{\dagger}A$$

We shall be considering the dipole matrix element for the atom, $\underline{p} = \langle j|e\underline{r}|i\rangle$, to be real. Since the matrix is hermitian $\underline{p} = \underline{p}_{ji} = \underline{p}_{ij}^{*}$ and so far $\underline{p}$ to be real this means that $\underline{p} = \underline{p}_{ji}^{*}$ also so that in all further equations the ordering of the indices of $\underline{p}$ will be considered unimportant.

Finally, in this section, we shall give the form of the Hamiltonian to be used in out calculations. It includes the R.W.A. and assumes the atom to be at the origin of co-ordinates, since, when its interaction with other atoms is negligible, its position relative to them has no significance (see Chapter VIII, Section 2, where this is shown to be true in the long wavelength limit). The Hamiltonian is

$$H = \hbar\omega_0 A^{\dagger}A + \sum_{\ell,\sigma} \hbar\omega_{\ell}\, a_{\ell\sigma}^{\dagger}a_{\ell\sigma} - \sum_{\ell,\sigma} \hbar g_{\ell\sigma}(A^{\dagger}a_{\ell\sigma} + a_{\ell\sigma}^{\dagger}A) \qquad (1.B.105)$$

1.B.9. Density matrix formation

According to Mollow and Miller,[7] the coupling of the atom (S) to the bath (R) makes it necessary to describe the state of the atom by means of a density operator, since an initially pure state becomes mixed under the influence of the damping mechanism. (In Chapter VIII, Section 6 various papers for, and one against, considering the states becoming mixed are discussed.)

As pointed out by Lehmberg,[1] in most treatments, three assumptions are made:

(i) R has a BROAD continuum of modes coupled more or less uniformly to S

(ii) The initial full density operator can be written as

$$\rho(0) = \rho^{(S)}(0)\rho^{(R)}(0) \tag{1.B.106}$$

where $\rho^{(S)}(0)$ describes the initial state of S, which is an arbitrary mixed state, and $\rho^{(R)}(0)$ is the thermal equilibrium distribution for R which is in a pure state.

Eq. (1.B.106) means that at t = 0, before the loss oscillators are coupled to the cavity, system S and reservoir R are independent and so the density operator factorises into a direct product.

(iii) R is only slightly affected by its interaction with S.

The last assumption need not be used in Lehmberg's method and is made only so that the approximation

$$\rho(t') = \rho^{(S)}(t')\rho^{(R)}(0), \tag{1.B.107}$$

namely the Markoff approximation, which will be discussed in Chapter VIII, Section 8, can be used to replace the actual density operator $\rho(t')$ by the factorised expression $\rho^{(S)}(t')\rho^{(R)}(0)$ when it occurs in second order terms. According to Mollow and Miller,[7] the criterion given for this approximation is that excitations induced in the bath R by its interaction with the atom S remain small throughout the experiment and so, as far as its effect on the atom is concerned, the state of R at any time may be approximated by its initial state, i.e. $\rho^{(R)}(t') = \rho^{(R)}(0)$. This is true when the reservoir, R, is very large, i.e. in other words, if the radiation field is very strong.

If H and $\rho(t)$ represent the complete Hamiltonian and density operator respectively and if S is described by basis states $|m\rangle$ and eigenstates $\{m\}$ then the components of the reduced density operator can be written in the Heisenberg picture as:

$$\left.\begin{aligned} \rho^{(S)}_{\ell,m}(t) &= \langle \ell | Tr_R \rho(t) | m\rangle \\ &= Tr\{\rho(0) P_{m,\ell}(t)\} \\ &= \langle P_{m,\ell}(t)\rangle \end{aligned}\right\} \qquad (1.B.108)$$

where Tr_R denotes trace over reservoir co-ordinates only, and Tr denotes trace over both reservoir and system co-ordinates.

Eq. (1.B.108) can be explained more fully as follows:

$\rho^{(S)}_{\ell,m}(t) = \langle \ell | Tr_R \rho(t) | m\rangle$, since $Tr_R \rho(t) = \rho^{(S)}(t)$ and $Tr_S \rho(t) = \rho^{(R)}(t)$

$= \langle \ell | \sum\limits_R \langle R | \rho(t) | R\rangle | m\rangle$ from eq. (1.B.6)

$= \sum\limits_R \langle \ell, R | \rho(t) | R, m\rangle$

$= \sum\limits_{S,R} \langle \ell | S\rangle \langle S, R | \rho(t) | R, m\rangle$, since the eigenstates of S form a complete set, i.e. $\sum\limits_S |S\rangle\langle S| = I$

$= \sum\limits_{S,R} \langle S, R | \rho(t) | R, m\rangle \langle \ell | S\rangle$

$= \sum\limits_{S,R} \langle S, R | \rho(t) | m\rangle \langle \ell | R, S\rangle$

$= \sum\limits_{S,R} \langle S, R | \rho(t) P_{m\ell}(0) | R, S\rangle$

$= Tr_{S,R}\{\rho(t) P_{m,\ell}(0)\}$

$= Tr_{S,R}\{U\rho(0)U^{\dagger} P_{m,\ell}(0)\}$, since $\rho(t) = U\rho(0)U^{\dagger}$ as will be shown in eq. (1.B.125)

$$= \mathrm{Tr}_{S,R}\{\rho(0)U^{\dagger}P_{m\ell}(0)U\},$$ since $\mathrm{Tr}(AB) = \mathrm{Tr}(BA)$ (see eq. (1.B.7))

$$= \mathrm{Tr}_{S,R}\{\rho(0)P_{m,\ell}(t)\},$$ from eq. (1.B.96), i.e. $P_{m,\ell}(t)_H = U^{\dagger}P_{m\ell}(0)_S U$

$$= \langle P_{m,\ell}(t)\rangle$$ by definition

$P_{m,\ell}(t)$ is known as a generalised projection operator or transition operator and is defined in the Heisenberg picture as

$$P_{m,\ell}(t) = |m,t\rangle\langle\ell,t| = e^{i/\hbar Ht}|m\rangle\langle\ell|e^{-i/\hbar Ht} \tag{1.B.109a}$$

and in Chapter II is put equal to $A_m^{\dagger}(t)A_\ell(t)$, where the atomic operators satisfy $\left[A_\ell, A_m\right]_+ = 0$, $\left[A_\ell, A_m^{\dagger}\right] = \delta_{\ell,m}$.

Also

$$P_{m,\ell}(0) = |m\rangle\langle\ell| \tag{1.B.109b}$$

where it is assumed that $|m\rangle = |m,t=0\rangle$ and $\langle\ell| = \langle\ell,t=0|$.

In the formalism presented in Lehmberg's 1st. paper[1] the dynamical properties of the damped system are calculated from $P_{m,\ell}(t)$ rather than $\rho(t)$. Damped equations of motion can be derived for all $P_{m,\ell}(t)$ if R consists of a BROAD-land distribution of harmonic oscillators even if S is a multi-level system. (We shall consider multi-level systems in later chapters.) The initial states of R need not be specified and the treatment need not be restricted to 2nd. order in the S-R coupling, i.e. to 2nd. order in g_q.

If R is a collection of harmonic oscillators, then one can derive damped equations of motion for the amplitude operators of S, without explicitly using assumptions (ii) or (iii). Only <u>unperturbed reservoir</u>

co-ordinates appear in these equations, the perturbation due to S being entirely absorbed in the damping constant and frequency shift. Similar damped equations are derived in Chapters V onwards for the reduced density-matrix elements of multi-level atoms which provide a convenient and nearly exact starting point for studying their interaction with known radiation fields.

Although the initial state of the damping radiation need not be specified, when it is, the transition operator equations lead immediately to those for the reduced density matrix as we shall show using assumption (ii) also.

(1) We find equations of motion for $P_{m,\ell}(t)$,

(2) we derive from them equations of motion for

$X_{m,\ell}(t) = {}_R\langle i|P_{m,\ell}(t)|i\rangle_R$,

(3) and, from these, equations of motion for

$\rho^{(S)}_{\ell,m}(t) = \sum_S \langle S|X_{m,\ell}(t)\rho^{(S)}(0)|S\rangle$ where $|i\rangle_R$ is the initial state of R.

Now

$$\left.\begin{aligned}
\rho^{(S)}_{\ell,m}(t) &= \mathrm{Tr}_{R,S}\{\rho(0)P_{m,\ell}(t)\} = \langle P_{\ell,m}(t)\rangle && \text{from eq. (1.B.108)}\\
&= \mathrm{Tr}_{R,S}\{P_{m,\ell}(t)\rho(0)\} && \text{since } \mathrm{Tr}(AB) = \mathrm{Tr}(BA) \text{ see (1.B.7)}\\
&= \sum_{R,S} \langle R,S|P_{m,\ell}(t)\rho(0)|S,R\rangle\\
&= \sum_{R,S} \langle R,S|P_{m,<}(t)\rho^{(S)}(0)\rho^{(R)}(0)|S,R\rangle && \text{from (1.B.106)}\\
&= \sum_{R,S} \langle R,S|P_{m,\ell}(t)\rho^{(S)}(0)|i\rangle_R\,{}_R\langle i|S,R\rangle\\
&\text{since } \rho^{(R)}(0) = |i\rangle_R\,{}_R\langle i|, \text{ i.e. reservoir is in a pure state at } t = 0.
\end{aligned}\right\} \quad (1.B.110)$$

$$= \sum_{R,S} {}_R\langle i|R\rangle\langle R,S|P_{m,\ell}(t)\rho^{(S)}(0)|i\rangle_R|S\rangle$$

$$= \sum_S {}_R\langle i|\langle S|P_{m,\ell}(t)\rho^{(S)}(0)|i\rangle_R|S\rangle$$

since $\sum_R |R\rangle\langle R| = I$, i.e. reservoir basis states form a complete set.

$$\boxed{\rho^{(S)}_{\ell,m}(t) = \sum_S \langle S|{}_R\langle i|P_{m\ell}(t)|i\rangle_R\,\rho^{(S)}(0)|S\rangle}$$

$$= \sum_S \langle S|X_{m\ell}(t)\rho^{(S)}(0)|S\rangle$$

From this we conclude that we never need all the information contained in $\rho(t)$ (see p. 184, ref. 34) but only need the reduced density operator defined by

$$\rho^{(S)}(t) = \mathrm{Tr}_R\,\rho(t).$$

N.B. $a_q|i\rangle_R = \sqrt{i}\,|i-1\rangle$ where i = no. of photons in reservoir mode at $t = 0$

or if $|i\rangle_R = |0\rangle_R$ vacuum state

$a_q|0\rangle_R = 0$

We shall be considering the initial state of R to be a vacuum state when we consider external fields classically. But when we consider driving fields in Chapters V onwards we shall consider them both classically in which case $|i\rangle_R$ equals $|0\rangle_R$, and also quantum mechanically using Glauber's notation. Both methods will be seen to give the same result except that Glauber's approach is more consistent.

We might add that in the case of a WEAK field Mollow and Miller [7] say that one can assume the atom remains in a <u>pure</u> state and add a phenomenological damping term [as in their eq. (3.13)] to the equation for the time derivatives of the amplitude of the excited state. They use a 1st. order P.T. approximation and assume the density operator for the atom corresponds to a pure state, i.e.

$$\left.\begin{aligned} &\rho^{(S)'}(t) = |t\rangle_{S}{}_{S}\langle t| \quad \text{for an \underline{initially unexcited} atom} \\ \text{where}\quad &|t\rangle_S = |i\rangle_S + \alpha'(t)|j\rangle_S \\ \text{and}\quad &\alpha'(t) = \langle j|\rho^{(S)'}(t)|i\rangle \\ & = \mathrm{Tr}\left[\rho^{(S)'}(t)A\right] \\ \text{and}\quad &\rho^{(S)'}(t)\overset{M.A.}{\approx}\rho^{(S)'}(t)|0\rangle_{R}{}_{R}\langle 0| \end{aligned}\right\} \quad (1.B.111)$$

But in the general case they point out that the initially unexcited atom becomes MIXED as a result of coupling to its bath R.

We shall be considering in Chapters V onwards the effects of fields which are not necessarily weak and so even were the atom initially unexcited (which it probably won't be anyway) P.T. cannot be used in this way.

From the above explanations some meaning of density operators has emerged but we shall explain their properties and importance a little further before proceeding to use them in the following chapters.

Basically density operators, first introduced by von Neumann, [44], [45], are necessary, because, in both classical and quantum mechanics, though for different reasons, a lack of knowledge of the system under study requires a statistical approach. In quantum mechanics this lack is due to the fundamental nature of the disturbance caused by the measure itself. There are 2 main situations:

(1) in which the state of the system can be represented by a definite wave function or state vector $|\psi\rangle$ and hence the system is said to be in a <u>pure</u> state. I.E. we have as much knowledge about the system as is allowed quantum mechanically.

(2) the state of the system is not completely known and it is said to be in a mixed state.

In case (1) each system in the ensemble, similar to the one under study, is the same, whereas in case (2), each system is in a different possible state weighted by a probability, p_ψ, according to some partial knowledge of the system.

For case (1) the ensemble average of an observable A, which is the same as the value of A for the system, is

$$\langle A\rangle_\psi = \frac{\langle\psi|A|\psi\rangle}{\langle\psi|\psi\rangle} = \langle\psi|A|\psi\rangle \quad \text{for } \langle\psi|\psi\rangle = 1 \tag{1.B.112}$$

whereas for case (2) if there is a probability p_ψ that the system is in state $|\psi\rangle$ then

$$\left.\begin{aligned} \langle A\rangle &= \sum_\psi p_\psi \langle A\rangle_\psi \\ &= \sum_\psi p_\psi \langle\psi|A|\psi\rangle \end{aligned}\right\} \tag{1.B.113a}$$

where $\langle\psi|\psi\rangle = 1$ (1.B.113b)

is assumed.

N.B. different states $|\psi\rangle$ are not necessarily orthogonal, i.e. $\langle\psi|\psi'\rangle$ is not known.

$\sum_\psi$ is over all possible states $|\psi\rangle$ of the system.

Also $p_\psi \geq 0$ and $\sum_\psi p_\psi = 1$ (properties of probability) (1.B.113c)

The density operator, ρ, was introduced in order to aid in calculating ensemble averages. It is defined by

$$\rho = \sum_{\psi} |\psi\rangle \, p_{\psi} \, \langle\psi| \tag{1.B.114}$$

for mixed states, and describes an ensemble of quantum systems.

Since ρ is an operator it can have a matrix representation, e.g. for an ensemble of harmonic oscillators with basis vectors represented by energy eigenvectors $\{|n\rangle\}$ we can write eq. (1.B.114) in this representation, as the density matrix

$$\langle n|\rho|n'\rangle = \sum_{\psi} \langle n|\psi\rangle p_{\psi} \langle\psi|n'\rangle \tag{1.B.115}$$

The density operator has various properties which are proved in ref. 35, pp. 222 onwards.

(i) $\quad \mathrm{Tr}\rho = 1 \qquad$ (1.B.116)

(ii) $\quad \mathrm{Tr}|\psi\rangle\langle Q| = \langle Q|\psi\rangle \qquad$ (1.B.117)

N.B. A trace may be taken in any representation, viz.

$\mathrm{Tr}|\psi\rangle\langle Q| = \sum_{n} \langle n|\psi\rangle\langle Q|n\rangle$ as long as the set $\{|n\rangle\}$ is complete.

(iii) $\quad \langle A\rangle = \mathrm{Tr}\rho A \qquad$ (1.B.118)

Also $\quad \langle f(A)\rangle = \mathrm{Tr}\rho f(A)$

(iv) $\quad \rho = \rho^{\dagger}$ i.e. ρ is hermitian $\qquad$ (1.B.119)

(v) $\quad \rho$ is positive definite

since if $|\chi\rangle$ is any bet

$$\left.\begin{aligned} \langle\chi|\rho|\chi\rangle &= \sum_{\psi} p_{\psi} \langle\chi|\psi\rangle\langle\psi|\chi\rangle \\ &= \sum_{\psi} p_{\psi} \left|\langle\psi|\chi\rangle\right|^2 \\ &\geq 0 \quad \text{since } p_{\psi} \geq 0 \end{aligned}\right\} \tag{1.B.120}$$

and real since p_{ψ} is real and $|\langle\psi|\chi\rangle|^2$ is real.

Therefore diagonal matrix elements of ρ are always real and positive.

Since $\quad Tr\rho = 1$

we also have $\quad \sum_n \langle n|\rho|n\rangle = 1 \quad$ i.e. $\sum_n \rho_{nn} = 1$

so each diagonal element of ρ in any representation possesses a real value between 0 and 1, i.e. $0 \leq \rho_{nn} \leq 1$.

We can now define a density operator as a positive definite hermitian operator of trace 1 which can represent an ensemble. It may be written in form (B.9.9).

(vi) Trρ is invariant under a unitary transform.

If S is the unitary transformation matrix

$$S^\dagger = S^{-1}$$

and if $\quad S\rho S^\dagger = \rho'$

then ρ' is diagonal.

$$TrS\rho S^\dagger = Tr\rho'$$

but $\quad TrS\rho S^\dagger = Tr\rho S^\dagger S \quad$ since trace is invariant under cyclic permutation (see eq. (1.B.7)

$\quad = Tr\rho \quad$ since $S^\dagger S = S^{-1}S = I$ (see eq. (1.B.12)

$$\therefore \quad Tr\rho = 1 = Tr\rho' \tag{1.B.121}$$

(vii) In diagonal representation the diagonal matrix elements of ρ' are ρ'_{nn}; they are real and satisfy

$$\sum_n \rho'_{nn} = 1 \quad \text{and} \quad 0 \leq \rho'_{nn} \leq 1$$

thus if ρ' is hermitian, positive definite and possesses a trace 1, it can be written in its diagonal representation (where $|n\rangle = S|\psi\rangle$) as

$$\rho' = \sum_n \rho'_{nn} |n\rangle\langle n| \tag{1.B.122}$$

where ρ'_{nn} are the non-degenerate eigenvalues.

The date vectors $\{|n\rangle\}$ form a complete orthogonal set

since $\quad Tr\rho' = 1$

and $\quad \sum_n \rho'_{nn} = 1$

and also since eigenvalues of hermitian operators are real and pos itive

$$0 \leq \rho'_{nn} \leq 1$$

ρ'_{nn} therefore possesses the properties of statistical weights and therefore ρ' is a density operator of a mixture of states $|n\rangle$ each with weight $\rho'_{nn} = P'_\psi$.

(viii) $\quad Tr\rho^2 \leq 1 \qquad$ (1.B.123)

since $\quad Tr\rho^2 = Tr\rho'^2 = \sum_n \rho'^2_{nn} \leq (\sum_n \rho'_{nn})^2 = (Tr\rho')^2 = 1$

where $\quad \sum_n \rho'^2_{nn} \leq (\sum_n \rho'_{nn})^2$

is a general mathematical inequality.

It can be shown by means of a procedure similar to that used in deriving (1.B.17) that the equation of motion for $\rho(t)$ is

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}\left[H, \rho(t)\right] \tag{1.B.124}$$

N.B. ρ is a function of time in the Schrödinger picture although most other variables are not in this picture.

In deriving this, we use the fact that $\rho(t)$, for a statistical mixture, undergoes a unitary transformation as time progresses, i.e.

$$\rho_S(t) = \sum_\psi p_\psi |\psi_S(t)\rangle\langle\psi_S(t)| = \sum_\psi p_\psi U|\psi_H(t_0)\rangle\langle\psi_H(t_0)|U^\dagger$$

from (1.B.9)

$$\boxed{\rho_S(t) = U(t,t_0)\rho_H(t_0)U^\dagger(t,t_0)} \qquad (1.B.125)$$

where $\rho(t_0)$ is the initial density operator

$$\rho_H(t_0) = \sum_\psi p_\psi |\psi(t_0)\rangle\langle\psi(t_0)| \qquad (1.B.126)$$

In the Schrödinger picture the expectation value of $A_S(t_0)$ at time t_0 is

$$\langle A_S(t_0)\rangle = \sum_\psi p_\psi \langle\psi_S(t)|A_S(t_0)|\psi_S(t)\rangle = \mathrm{Tr}\rho_S(t)A_S(t_0) \qquad (1.B.127a)$$

On transforming to the Heisenberg picture by means of (1.B.125)

$$\langle A\rangle = \mathrm{Tr}(U\rho_H(t_0)U^\dagger A_S(t_0))$$

$$= \mathrm{Tr}(\rho_H(t_0)A_H(t)) \qquad (1.B.127b)$$

since $U^\dagger A_S(t_0)U = A_H(t)$.

Thus we again see that, whether we evaluate ensemble averages in either picture, the result is the same. In our applications the Heisenberg picture is easier.

In the case of the system being in a pure state $|\psi\rangle$, $p_\psi = 1$ and $p'_\psi = 0$ for all $\psi' \neq \psi$. Every system of the ensemble is in state $|\psi\rangle$ and therefore

$$\rho = \rho_\psi = |\psi\rangle\langle\psi| \qquad (1.B.128)$$

Hence

$$\left.\begin{aligned}
&\rho_{\psi}^2 = \rho_{\psi} \\
&\mathrm{Tr}\rho_{\psi} = 1 \\
&\mathrm{Tr}\rho_{\psi}^2 = \mathrm{Tr}\rho_{\psi} = 1
\end{aligned}\right\} \qquad (1.B.129)$$

N.B. For a mixed state $\mathrm{Tr}\rho^2 \leq 1$.

So the necessary and sufficient condition that a density operator represents a pure state is that

$$\mathrm{Tr}\rho^2 = 1,$$

assuming that it can be proved that ρ is hermitian, positive definite and $\mathrm{Tr}\rho = 1$ also.

When the ensemble represents a pure state, then

$$\left.\begin{aligned}
\langle A\rangle &= \mathrm{Tr}\rho A \\
&= \mathrm{Tr}\sum_{\psi'} p_{\psi}' |\psi'\rangle\langle\psi'|A \\
&= \sum_{n,\psi'} \langle n|p_{\psi}'|\psi'\rangle\langle\psi'|A|n\rangle \\
&= \sum_{n,\psi'} \langle\psi'|A|n\rangle\langle n|\psi'\rangle p_{\psi}' \\
&= \sum_{\psi'} \langle\psi'|A|\psi'\rangle p_{\psi}' \\
&= \langle\psi|A|\psi\rangle
\end{aligned}\right\} \qquad (1.B.130a)$$

since $\sum_{\psi'} p_{\psi'} = \delta_{\psi\psi'}$ (1.B.130b)

as given also in (1.B.112), i.e. ensemble averages, for systems in a definite state $|\psi\rangle$, are ordinary quantum ensemble averages.

Therefore density operators are useful in calculating ensemble averages whether one has complete (see (1.B.130a)) or incomplete knowledge (see (1.B.127)) of the state of the system.

CHAPTER II

A TWO LEVEL SYSTEM WITH EXCITED STATE COUPLED BY A CLASSICAL EXTERNAL PERTURBATION [3]

A. Heisenberg equations of motion

We shall consider a 2-level quantum system S to be simultaneously coupled by a quantised radiation interaction R, describing the decay, and in addition, by a classical time independent external perturbation, E. The bath of oscillators R are assumed to be closely spaced in frequency such that their frequencies overlap the atomic resonance frequency, ${}_{o}$.

For comparison of notation with that of later Chapters V, VI and VII, where method (i), based on Mollows classical treatment of the external field is used, see Appendix II.

We shall use the following Hamiltonian for the entire system of atom, bath and external perturbation:-

$$\left.\begin{array}{lll}
 & H = H_o + H_I & \\
\text{where} & H_o = H_s + H_R & \text{is the unperturbed Hamiltonian} \\
\text{and} & H_I = V_{sR} + V_{sE} & \text{is the interaction Hamiltonian} \\
 & H_s = \hbar\omega_o\, A^+ A & \text{is the Hamiltonian for the atomic system} \\
 & H_R = \hbar \sum_{l,\sigma} \omega_l\, a^+_{l\sigma}\, a_{l\sigma} & \text{is the Hamiltonian for the bath of loss oscillators} \\
 & V_{sR} = \hbar A^+ \sum_{l,\sigma} g_{l,\sigma}\, a_{l\sigma}\, e^{i\theta_l} + \hbar \sum_{l,\sigma} a^+_{l\sigma}\, e^{-i\theta_l} A & \\
 & & \text{is the Hamiltonian for the interaction between the atoms and bath in resonant form} \\
 & V_{sE} = \hbar\lambda'\, e^{-i\psi} A^+ + \hbar\lambda'\, e^{i\psi} A & \text{is the Hamiltonian for the interaction between atom S and external perturbation E in resonant form.}
\end{array}\right\} \quad (2.A.1)$$

$g_{\ell\sigma}$ and λ' are c-number coupling parameters. (It is shown elsewhere that they are indentifiable as $g_{\ell\sigma} = \sqrt{\frac{2\pi\,\omega_\ell}{\hbar V}}\;\hat{e}_{\ell\sigma}\cdot\underline{p}$, see eq. (1B99), and $\lambda' = -\lambda\varepsilon_{OD}\,C^{i\psi}$ and $\lambda'^{*} = -\lambda\varepsilon_{OD}\,e^{-i\psi}$, see eq.(II.8), when the perturbation is a time independent electric field, $\underline{E}_D = 2\epsilon_{OD}\,\hat{e}_{OD}$)

θ_ℓ and ψ denote arbitrary phases.

We now derive the Heisenberg equations of motion ofor $A(t) = P_{ij}(t)$, the number operator $Q(t) = A^\dagger(t)\,A(t) = P_{jj}(t)$ and $a_{\ell\sigma}(t)$, using eq. (2.A.1.) and the relations

$$\left[A,\, A^\dagger\right]_+ = 1 \qquad (2.A.1a.)$$

$$\left[a_{\ell\sigma},\, a^\dagger_{\ell'\sigma'}\right] = \delta_{\ell\sigma,\,\ell'\sigma'} \qquad (2.A.1b.)$$

and

$$\left[A,\, A^\dagger\right] = 1 - 2Q \qquad (2.A.1c.)$$

$$\left[A^\dagger,\, A^\dagger A\right] = -A^\dagger = \left[A^\dagger,\, Q\right] \qquad (2.A.1d.)$$

$$\left[A,\, A^\dagger A\right] = A = \left[A,\, Q\right] \qquad (2.A.1e.)$$

$$\dot{a}_{\ell\sigma} = i/\hbar\left[H,\, a_{\ell\sigma}\right] \text{ from (1.B.68.) since } \left[H,\, a_{\ell\sigma}\right] = -\left[a_{\ell\sigma},\, H\right]$$

$$= -\,i\omega_\ell\, a_{\ell\sigma} - i g_{\ell\sigma}\, A e^{-i\theta_\ell}$$

NB. H is time dependent in the Heisenberg picture.

with formal solution[46].

$$a_{\ell\sigma}(t) = a_{\ell\sigma}(0)\, e^{-i\omega_\ell t} - i g_{\ell\sigma}\int_0^t dt'\, e^{-i(\omega_\ell(t-t') + \theta_\ell)} \times A(t') \qquad (2.A.2.)$$

and hermitian conjugate

$$a^\dagger_{\ell\sigma}(t) = a_{\ell\sigma}(0)\;\; e^{i\omega_\ell t} + i g_{\ell\sigma}\int_0^t dt''\, e^{i(\omega_\ell(t-t') + \theta_\ell)} \times A(t''). \qquad (2.A.3.)$$

$$\dot{A}(t) = i/\hbar\;\; H,\, A(t) \quad \text{from (1.B.82) where } H \text{ is now total Hamiltonian}$$

$$= i\omega_o\, A(t) + i\,2\,(Q(t)-\tfrac{1}{2}\sum_{\ell\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t)\, e^{i\theta_\ell} + i\,2\lambda'\, e^{-i\psi}(Q(t)-\tfrac{1}{2}) \qquad (2.A.4.)$$

$$\dot{Q}(t) = i/\hbar \left[H, Q(t)\right]$$

$$= - i A(t) \sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(t) e^{i\theta_\ell} + i \sum_{\ell,\sigma} g_{\ell\sigma} a e_\sigma^\dagger (t) e^{-i\theta_\ell} A(t)$$
$$- i A^\dagger(t) \lambda' e^{-i\psi} + i\lambda'^* e^{i\psi} A(t) \qquad (2.A.5.)$$

Consider

$$\sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(t) = \sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(0) e^{-i\omega_\ell t} - i \sum_{\ell\sigma} g_{\ell\sigma}^2 \int_0^t dt' e^{-i(\omega_\ell (t-t')+\theta_\ell)} A(t') \qquad (2.A.6.)$$

Now put, for the quantised part of the field:-

$$\underline{E}_{(+)}^{(0)} (0, t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}} \hat{e}_{\ell\sigma} a_{\ell\sigma}(0) e^{-i\omega_\ell t}$$

$$\underline{E}_{(-)}^{(0)} (0, t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}} \hat{e}_{\ell\sigma} a_{\ell\sigma}^\dagger(0) e^{i\omega_\ell t}$$

i.e. fields are in the Schrodinger representation since operators $a_{\ell\sigma}$ and $a_{\ell\sigma}$ are time independent (2.A.7.)

$$\underline{E}^{(0)} (0, t) = \underline{E}_{(+)}^{(0)} (0, t) + \underline{E}_{(-)}^{(0)} (0, t)$$

N.B. $\underline{E}_{+}^{(0)\dagger} (0, t) = \underline{E}_{(-)}^{(0)} (0, t)$

so that we can rewrite (2.A.6.) as

$$\sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\omega_\ell}{\hbar V}} \hat{e}_{\ell\sigma} \cdot \underline{p}\, a_{\ell\sigma}(0) e^{-i\omega_\ell t} - i \sum_{\ell,\sigma} g_{\ell\sigma}^2 \int_0^t dt' e^{-i(\omega_\ell (t-t')+\theta_\ell)} A(t')$$

$$= \sum_{\ell,\sigma} \frac{1}{\hbar} \underline{E}_{+}^{(0)} (t) \cdot \underline{p} - i \sum_{\ell,\sigma} g_{\ell\sigma}^2 \int_0^t dt' e^{-i(\omega_\ell (t-t')+\theta_\ell)} A(t') \qquad (2.A.8.)$$

If we now consider the interaction to take place in free space $V \rightarrow \infty$ and therefore, according to (1.B.73),

$$\frac{1}{V} \sum_\ell (\;) \rightarrow \frac{1}{(2\pi c)^3} \int_0^\infty \omega^2 \, d\omega \int_0^{4\pi} d\Omega_\ell \, (\;)$$

Naturally

$$\sum_{\sigma} = \sum_{\sigma=1}^{2} \text{ always.}$$

We have dropped the subscripts ℓ on ω and θ since ω is now the variable of integration. Thus (2.A.8.) becomes

$$\sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t) \frac{1}{\hbar} \sum_{\ell,\sigma} \underset{\rightarrow +}{E}^{(0)}(t) \cdot \underline{p} = -i \frac{1}{(2\pi c)3} \int_{0}^{\infty} \frac{2\pi\omega}{\hbar}\, \omega^2\, d\omega \int_{0}^{4\pi} d\Omega_{\hat{k}} \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot p)^2$$

$$x \int_{0}^{t} dt' e^{-i(\omega(t-t')+\theta)}\, A(t')$$

$$= -i \frac{p^2}{4\pi^2 c^3 \hbar} \int_{0}^{\infty} d\omega\, \omega^3 \int_{0}^{4\pi} d\Omega_{\hat{k}} \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot \hat{p})^2$$

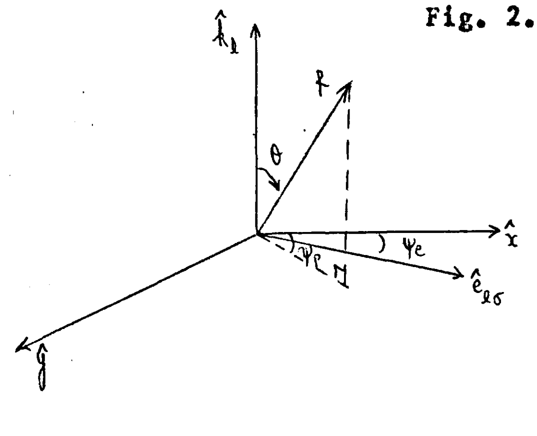
$$x \int_{0}^{t} dt' e^{-i(\omega(t-t')+\theta)} A(t')$$

(2.A.9.)

To find $\sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot \underline{p})^2 = (\hat{e}_{\ell 1} \cdot \underline{p})^2 + (\hat{e}_{\ell 2} \cdot \underline{p})^2$

we choose rectangular coordinate axes $\hat{x}$, $\hat{y}$ and $\hat{k}_{\ell}$, where $\hat{k}_{\ell}$ is the direction of propogation of the photon, and $\hat{x}$ and $\hat{y}$ are any arbitrary directions perpendicular to each other and in a plane perpendicular to $\hat{k}_{\ell}$. Since $\hat{e}_{\ell\sigma} \cdot \hat{k}_{\ell} = 0$ in the Coulomb gauge, $\hat{e}_{\ell\sigma}$ is in the (x, y) plane, $\underline{p}$ is not in any fixed direction.

Fig. 2.A.1



Using spherical polar coordinates[39] as in the above diagram, we can write

$$\hat{e}_{\ell\sigma} = \hat{x} \cos \psi_e + \hat{y} \sin \psi_e$$

$$\hat{p} = \hat{x} \cos \psi_p \sin \theta + y \sin \psi_p \sin \theta + \hat{k}_e \cos \theta$$

$$\therefore \quad \hat{e}_{\ell\sigma} \cdot \hat{p} = \cos \psi_e \cos \psi_p \sin \theta + \sin \psi_e \sin \psi_p \sin \theta$$

$$= \cos (\psi_e - \psi_p) \sin \theta$$

$$(\hat{e}_{\ell\sigma} \cdot \hat{p})^2 = \cos^2 (\psi_e - \psi_p) \sin^2 \theta$$

Now for each mode ℓ there are 2 polarisations, hence there are 2 polarisation vectors $\hat{e}_{\ell 1}$ and $\hat{e}_{\ell 2}$ and since these are both in the (x, y) plane and perpendicular to each other (see eq (1.B.4.3.) $\hat{e}_{\ell 1} \cdot \hat{e}_{\ell 2} = 0$) they can be used as axes to replace $\hat{x}$ and $\hat{y}$.

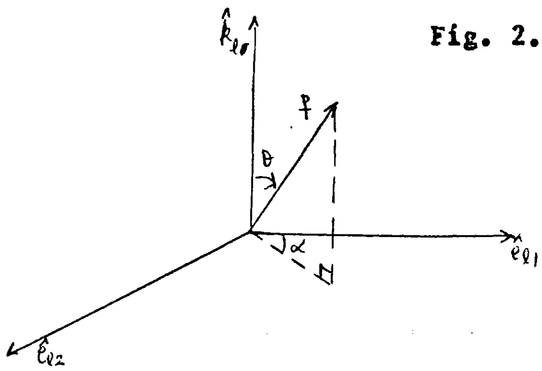


Fig. 2.A.2

Then

$$\hat{p} = \hat{e}_{\ell 1} \cos \alpha \sin \theta + \hat{e}_{\ell 2} \sin \alpha \sin \theta + \hat{k}_{\ell} \cos \theta$$

$$\therefore \quad \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot \hat{p})^2 = (\hat{e}_{\ell 1} \cdot \hat{p})^2 + (\hat{e}_{\ell 2} \cdot \hat{p})^2$$

$$= \cos^2 \alpha \sin^2 \theta + \sin^2 \alpha \sin^2 \theta$$

$$= \sin^2 \theta$$

$$= 1 - \cos^2 \theta$$

$$= 1 - (\hat{p} \cdot \hat{k}_{\ell})^2 \qquad (2.A.10.)$$

Substituting (2.A.10.) in (2.A.9.) we obtain

$$\sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t) - \frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}^{(0)}_{(+)}(t).p = \frac{-\,i\,p^2}{4\pi^2 c^3 \hbar}\int_0^\infty d\omega\,\omega^3 \int_0^{4\pi} d\Omega_{\hat{k}}(1-(\beta.k)^2)\int_0^t$$

$$\times\ dt' e^{-i(\omega(t-t')+\theta)}\, A(t') \qquad (2.A.11)$$

$$\text{Now} \int_0^{4\pi} d\Omega_{\hat{k}} = \int_0^{\pi} \sin\theta \; d\theta \int_0^{2\pi} d\psi \qquad (2.A.12.)$$

$$\text{Hence} \int_0^{4\pi} d\Omega_{\hat{k}} \left(1 - (\hat{k}.\beta)^2\right) = \int_0^{\pi} \sin\theta(1-\cos^2\theta)d\theta \int_0^{2\pi} d\psi$$

$$= 2\pi \int_{-1}^{1} (1-x^2)\, dx \qquad \text{where} \quad x = \cos\theta$$

$$= 8\pi/3 \qquad (2.A.13.)$$

Thus eq.(2.A.11.) becomes

$$\sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t) - \frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}^{(0)}_{(+)}(\underline{r},t).p = -\frac{i2p^2}{3\pi c^3 \hbar}\int_0^\infty d\omega\,\omega^3 \int_0^t dt' e^{-i(\omega(t-t')+\theta)} A(t')$$

$$(2.A.14.)$$

Now according to Lehmberg[1], although the ω integral extends to ∞ the dipole approximation begins to break down as $\omega \to \omega_B = c/a_B \sim 10^{10}/10^{-8} = 10^{18}$ per sec. where a_B is the Bohr radius, i.e. the radius of the ground state of the hydrogen atom, and so ω_B is therefore related to the hydrogen atom and is $\gg \varepsilon_{yz}$ for any other atom with levels $|y\rangle$ and $|z\rangle$ where $y > z$. In a more exact treatment, p should be replaced by some function $p(\omega)$ which decreases exponentially for $\omega > \omega_B$

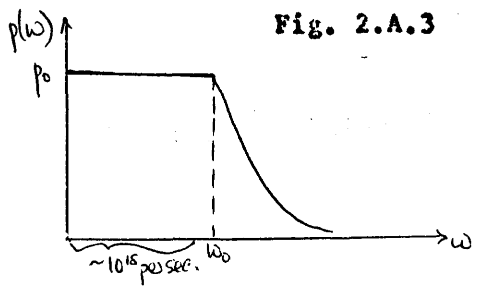


Fig. 2.A.3

$$p(\omega) = p_o e^{-a\omega} \quad \text{for } \omega > \omega$$

$$p(\omega) = p_o \quad \text{for } 0 < \omega < \omega$$

i.e. the integral effectively cuts off around $\omega \triangleq \omega_B$, i.e. we should have $\int_0^{\omega_B} d\omega$ and not $\int_0^{\infty} d\omega$. For a given atom the most important values of t' ∴ lie within a region on the order of ω_B^{-1} ($= 10^{-18}$ sec) around t i.e. $(t'-t)_{max.} = \omega_B^{-1}$. Since $\omega_B >> \omega_o$ (in fact for the Nα line

$\lambda = 10^{-5}$ cm.

∴ $\omega_o \approx 10^{15}$ sec.$^{-1}$

so that important values of t' lie within a region $\omega_B^{-1} << \tau$ around t)

we can replace

$$A(t') \text{ by } A(t)\, e^{-i\omega_o(t'-t)} \tag{2.A.15a.}$$

$$\text{and} \quad A\dagger(t') \text{ by } A\dagger(t)\, e^{-i\omega_o(t'-t)} \tag{2.A.15b.}$$

In fact $A(t)\, e^{-i\omega_o(t'-t)}$ has a maximum value $A(t)\, e^{-i\omega_o\omega_B^{-1}}$ but since ω_o/ω_B is very small ($10^{-18}\,\omega_o$ i.e. 10^{-3} for the Na line) even when we consider $(t'-t)$ at its maximum for all t' it still only effects $A(t)$ very slightly and this modification could be either added on so that

$$A(t') \rightarrow A(t) + A(t)\, e^{-i\omega_o/\omega_B}$$

or $\quad A(t') \rightarrow A(t)\, e^{-i\omega_o(t'-t)}$ and is substituted in and integrated over t'.

Either method makes little difference.

With replacement (2.A.15a.), (2.A.14.) becomes:-

$$\sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t) - \frac{1}{\hbar} \sum_{\ell,\sigma} \underset{\rightarrow}{E}^{(0)}(t).\underline{p} = \frac{-2p^2 A(t) e^{-i\theta}}{3\pi c^3 \hbar} \int_0^{\infty} d\omega\, \omega^3 \int_0^t dt' e^{-i((\omega-\omega_o)(t-t'))} \tag{2.A.16.}$$

Now

$$\int_0^t dt' e^{-i(\omega \mp \omega_o)(t-t')} = \left[\frac{e^{-i(\omega \mp \omega_o)(t-t')}}{i(\omega \mp \omega_o)} \right]_0^t$$

$$= \left(\frac{\sin(\omega \mp \omega_o)t}{\omega \quad \omega_o}\right) - i\left(\frac{1-\cos(\omega \mp \omega_o)t}{\omega \mp \omega_o}\right)$$

$$\xrightarrow[\omega_o t \;>>\; 1]{} \pi\,\delta(\omega \mp \omega_o) - i\,\frac{p}{\omega \mp \omega_o} \qquad \text{(2.A.17.)}$$

according to ref. 40 p.66-69, where p is the principal part.

The last approximation is made since we are only interested in times $t >> \omega_o^{-1}$ (i.e. 10^{15} sec. for Na)

Hence

$$\sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}(t) - \frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}_{(+)}^{(0)}(t).\underline{p} = \frac{-i2p^2\, A(t)\, e^{-i\theta}}{3\pi c^3\hbar}\left[\int_0^\infty d\omega\, \omega^3 \pi\, \delta(\omega-\omega_o) - i\int_0^\infty d\omega\, \omega^3 \frac{p}{\omega-\omega_o}\right]$$

$$= \frac{-i2p^2\, A(t) e^{-i\theta}}{3\pi c^3\hbar}\left[\pi\omega_o^3 - i\int_0^\infty d\omega\, \frac{\omega^3}{\omega-\omega_o}\right] \qquad \text{(2.A.18.)}$$

since $\int_0^\infty d\omega\, f(\omega)\, \delta(\omega-\omega_o) = f(\omega)$ is not singular at $\omega = \omega_o$ ^{2} and is true since ω_o is positive and is included in the integral $0 \to \infty$.

In the second term p can be ignored as no singularity is involved.

Now

$$\int_0^\infty d\omega\, \frac{\omega^3}{\omega \mp \omega_o} = \int_{\mp\omega_o}^\infty dx\, \frac{(x \pm \omega_o)^3}{x} \qquad \text{where } x = \omega \mp \omega_o$$

$$= \left[\frac{1}{3}x^3 \mp \frac{3\omega_o}{2}x^2 \mp \omega_o^3 \log e\, x\right]_{\mp\omega_o}^\infty \qquad \text{(2.A.19.)}$$

and this diverges unless we introduce a cut-off around $\omega_B = c/a_B \sim 10^{18}$ so that the upper limit is ω_B. Since we know that $\omega_B >> \omega_o$ this is a reasonable approximation and it makes the integral finite

N.B. $\int_0^\infty d\omega \frac{\omega^3}{\omega-\omega_o}$ has a singularity but the same reasoning applies.

Thus (2.A.18.) becomes

$$\sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(t) = +\frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}^{(0)}_{(+)}(t)\cdot\underline{p} - i\left[\frac{2p^2\omega_o^3}{3\hbar c^3} - i\frac{2p^2}{3\pi\hbar c^3}\int_0^\infty d\omega \frac{\omega^3}{\omega-\omega_o}\right] \times A(t)e^{-i\theta}$$

$$= +\frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}^{(0)}_{(+)}(t)\cdot\underline{p} - i\left[\tfrac{1}{2}\gamma - i\Omega_-\right]A(t)e^{-i\theta} \qquad (2.A.20.)$$

where $\gamma = \frac{4p^2\omega_o^3}{3\hbar c^3}$ is the decay constant,

$$\text{and } \Omega_{\pm} = \frac{\gamma}{\omega_o^3}\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega \pm \omega_o} = \frac{\gamma}{k_o^3}\int_0^\infty \frac{dk}{2\pi}\frac{k^3}{k \pm k_o} \qquad (2.A.21.)$$

is the frequency shift, where $k = \omega/c$, $k_o = \omega_o/c$.

The hermitian conjugate is:-

$$\sum_{\ell,\sigma} g_{\ell\sigma} a_{\ell\sigma}(t) = \frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}^{(0)}_{(-)}(t)\cdot\underline{p} + i(\tfrac{1}{2}\gamma + i\Omega_-)A^\dagger(t)e^{i\theta} \qquad (2.A.22.)$$

Substituting (2.A.20.) and (2.A.22.) in (2.A.4.) and (2.A.5.), we obtain

$$A(t) = -i\omega_o A(t) + i\frac{2}{\hbar}\sum_{\ell,\sigma} e^{i\theta\ell}(Q(t)-\tfrac{1}{2})\underline{E}^{(0)}_{(+)}(t)\cdot\underline{p} - (\tfrac{1}{2}\gamma - i\Omega_-)A(t)$$
$$+ i2\lambda' e^{-i\psi}(Q(t)-\tfrac{1}{2}) \qquad (2.A.23.)$$

since $Q(t)\,A(t) = 0$

$$Q(t) = -\frac{i}{\hbar}A^\dagger(t)\sum_{\ell,\sigma} e^{i\theta_\ell}\underline{E}^{(0)}_{(+)}(t)\cdot\underline{p} + \frac{i}{\hbar}\sum_{\ell,\sigma}\underline{E}^{(0)}_{(-)}(t)\cdot\underline{p}\,A(t)$$
$$- \gamma Q(t) - i\lambda' A^\dagger(t)e^{-i\psi} + i\lambda'^* A(t)e^{i\psi} \qquad (2.A.24.)$$

$$A(t) = i\,2\,(Q(t)-\tfrac{1}{2})(q' + \lambda' e^{-i\psi}) - (\tfrac{1}{2} + i\ \omega)A(t) \qquad (2.A.25.)$$

$$Q(t) = -iA^\dagger(t)(q' + \lambda' e^{-i\psi}) + i(q' + \lambda'^* e^{i\psi})A(t) - \gamma Q(t) \qquad (2.A.26.)$$

where $q = 1/\hbar \sum_{\ell,\sigma} e^{i\theta_\ell} \underline{E}_{(+)}^{(0)}(t) \cdot \underline{p} = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\omega_\ell}{\hbar V}}\, \hat{\varepsilon}_{\ell\sigma} \cdot \underline{p}\, a_{\ell\sigma}(0)$

$$\times\, e^{-i(\omega_\ell t-\theta_\ell)} = \sum_{\ell,\sigma} g_{\ell\sigma}\, a_{\ell\sigma}^{(0)}\, e^{-i(\omega_\ell t-\theta_\ell)} \qquad (2.A.27a)$$

and $q'^{\dagger} = 1/\hbar \sum_{\ell,\sigma} e^{-i\theta_\ell} \underline{E}^{(0)}(t) \cdot p = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\omega_\ell}{\hbar V}}\, \varepsilon_{\ell\sigma} \cdot \underline{p}\, a_{\ell\sigma}^{\dagger}(0)$

$$\times\, e^{i(\omega_\ell t-\theta_\ell)} = \sum_{\ell\sigma} g_{\ell\sigma}\, a_{\ell\sigma}^{\dagger}(0)\, e^{i(\omega_\ell t-\theta_\ell)} \qquad (2.A.27b)$$

(The only difference between the definitions or q' and $q'^{\dagger}$ here and those of q_{mn} and q_{mn}^{+} in Chapters IV, V, VI, VII, is a phase factor. N.B. q's are always time dependent).

$B(t) = i(q' + \lambda' e^{-i\psi})$ is the term quoted in ref. 3, and $\omega = \omega_0 - \Omega_-$.

We shall now calculate equations for

$$X(t) = \langle i_{ph} |A(t)| i_{ph} \rangle \qquad (2.A.28)$$

$$Y(t) = \langle i_{ph} |Q(t)| i_{ph} \rangle \qquad (2.A.29)$$

where the initial radiation state is assumed to be a vacuum photon state $|0\rangle_R$.

N.B. $q' |0\rangle_R = 0$ and ${}_R\langle 0| q'^{\dagger} = 0$

since $a_{\ell\sigma} |0\rangle_R = 0$ and ${}_R\langle 0|\, a_{\ell\sigma}^{\dagger} = 0$.

Hence

$$\dot{X}(t) = i\, 2\, \lambda' e^{-i\psi}\, Y(t) \quad - i\, \lambda' e^{-i\psi} \quad -(\tfrac{1}{2}\gamma + i\omega)\, X(t) \qquad (2.A.30.)$$

$$\dot{Y}(t) = -i\, \lambda' e^{-i\psi} \quad X(t)^{\dagger} \quad + i\, \lambda'^{*} e^{i\psi}\, X(t) \qquad -\gamma Y(t) \qquad (2.A.31.)$$

We shall now calculate equations for $\sigma(t)$ and $P(t)$

where $\sigma(t) = \mathrm{Tr}\ \rho(t)\, A$ and $P(t) = \mathrm{Tr}\ \rho(t)\, Q$. (2.A.32.)

As explained in the introduction Sec.A.9 eq. (1.B.108).

$$\sigma(t) = \rho^{(S)}(t)_{ji} \quad \text{and} \quad P(t) = \rho^{(S)}(t)_{jj} \quad \text{and} \quad \sigma^*(t) = \rho^{(S)}(t)_{ij} \tag{2.A.33}$$

since

$$\left.\begin{aligned}
\sigma(t) &= \sum_{S,R} \langle S,R|\rho(t)|i\rangle\langle j|S,R\rangle \quad \text{and} & P(t) &= \sum_{S,R} \langle S,R|\rho(t)|j\rangle\langle i|i\rangle\langle j|R,S\rangle \\
&= \sum_{S,R} \langle j|S\rangle\langle S,R|\rho(t)|i,R\rangle & &= \sum_{S,R} \langle S,R|\rho(t)|j\rangle\langle j|R,S\rangle \\
&= \sum_{R}\langle j,R|\rho(t)|i,R\rangle & &= \sum_{S,R} \langle j|S\rangle\langle S,R|\rho(t)|j,R\rangle \\
&= \mathrm{Tr}_R\langle j|\rho(t)|i\rangle & &= \sum_{R}\langle j,R|\rho(t)|j,R\rangle \\
&= \rho^{(S)}(t)_{ji} & &= \rho^{(S)}(t)_{jj}
\end{aligned}\right\} \tag{2.A.34}$$

Thus P(t) is the probability of finding the atom in its excited state $|j\rangle$ at time t. $\sigma(t)$ is the off-diagonal matrix element of the reduced density operator $\rho^{(S)}(t) = \mathrm{Tr}_R\rho(t)$ and P(t) is the diagonal one.

According to (B.9.5)

$$\rho^{(S)}(t)_{ji} = \sigma(t) = \sum_S \langle S|X(t)\rho^{(S)}(0)|S\rangle \quad \text{and} \quad \rho^{(S)}(t)_{jj} = P(t) = \sum_S \langle S|Y(t)\rho^{(S)}(0)|S\rangle$$

$$\tag{2.A.35}$$

From eqs. (2.24), (2.25) and the hermitian conjugate of (2.24) we obtain coupled linear differential equations

$$\dot{\sigma}(t) = \dot{\rho}^{(S)}(t)_{ji} = i2\lambda' e^{-i\phi}P(t) - i\lambda' e^{-i\phi} \quad -(\tfrac{1}{2}\gamma + i\omega)\sigma(t) \tag{2.A.36}$$

$$\dot{\sigma}^*(t) = \dot{\rho}^{(S)}(t)_{ij} = -i2\lambda'^* e^{i\phi}P(t) + i\lambda'^* e^{i\phi} \quad -(\tfrac{1}{2}\gamma - i\omega)\sigma^*(t) \tag{2.A.37}$$

$$\dot{P}(t) = \dot{\rho}^{(S)}(t)_{jj} = -i\lambda' e^{-i\phi}\sigma^*(t) + i\lambda'^* e^{i\phi}\sigma(t) - \gamma P(t) \tag{2.A.38}$$

since $\quad \sum_S \langle S|\rho^{(S)}(0)|S\rangle = \rho^{(S)}(0)_{ii} + \rho^{(S)}{}_{jj} = 1$

N.B. If $\lambda' = 0$:

$$\sigma(t) = \sigma(0)e^{-(\frac{1}{2}\gamma - i\omega)t}$$

$$P(t) = P(0)e^{-\gamma t}$$

i.e. where there is no external perturbation $\sigma(t)$ decays at half the rate of $P(t)$. See Mollow and Miller[7], p. 469 where this is shown to be an essential feature of the damping process.

B. Time dependence of the occupation of the excited state

Equations (2.A.36) to (2.A.38) can be solved exactly by using the method of Laplace transforms. If we define the Laplace transform of A(t) as

$$\left.\begin{aligned} L(A(t)) &= \int_{t'}^{\infty} dt e^{-st} A(t) = \hat{A}(S) \\ &\text{where the inverse is} \\ L^{-1}(A(t)) &= \frac{1}{2\pi i} \oint e^{st} \hat{A}(S) dS, \end{aligned}\right\} \qquad (2.B.1)$$

$$\left.\begin{aligned} \text{then} \quad L(\dot{A}(t)) &= \int_{t'}^{\infty} dt e^{-st} \dot{A}(t) \\ &= \Big[A(t)e^{-st}\Big]_{t'}^{\infty} + S \int_{t'}^{\infty} e^{-st} A(t) \\ &= -A(t')e^{-st'} + S\, L(A(t)) \\ &= -A(t')e^{-st'} + S\, \hat{A}(S) \end{aligned}\right\} \qquad (2.B.2)$$

If $A(t) = A$, a constant independent of time,

$$L(A) = A \int_{t'}^{\infty} dt e^{-st}$$

$$= A \left[\frac{e^{-st}}{-S}\right]_{t'}^{\infty} \qquad (2.B.3)$$

$$= A \frac{e^{-st'}}{S}$$

We shall consider the initial time to be $t' = 0$ (see Chapter VIII, Section 9) since we shall assume the atom to be initially in the excited state $|j\rangle$ so that we may find the characteristics of the decay process from level $|j\rangle$. Thus we assume $P(0) = 1$, $\sigma(0) = \sigma^*(0) = 0$.

Thus if we let

$$\varepsilon = \lambda' e^{-i\psi}, \quad \varepsilon^* = \lambda'^* e^{i\psi} \qquad (2.B.4)$$

then on taking Laplace transforms of eqs. (2.A.36) to (2.A.38) we obtain:

$$(S + \tfrac{1}{2}\gamma + i\omega)L(\sigma) = 2i\varepsilon L(p) - i\varepsilon\frac{1}{S} + \sigma(0) \qquad (2.B.5)$$

$$(S + \tfrac{1}{2}\gamma - i\omega)L(\sigma^*) = -2i\varepsilon^* L(p) + i\varepsilon^*\frac{1}{S} + \sigma^*(0) \qquad (2.B.6)$$

$$(S + \gamma) \quad L(p) = -i\varepsilon L(\sigma^*) + i\varepsilon^* L(\sigma) + p(0) \qquad (2.B.7)$$

Solving for $L(p)$ we obtain

$$\left.\begin{aligned} L(p) = 2|\varepsilon|^2 \frac{(S+\frac{1}{2}\gamma)}{SZ} &+ \frac{(S + \frac{1}{2}\gamma + i\omega)(S + \frac{1}{2}\gamma - i\omega)}{Z} p(0) \\ &+ i\varepsilon^* \frac{(S + \frac{1}{2}\gamma - i\omega)}{Z}\sigma(0) - i\varepsilon\frac{(S + \frac{1}{2}\gamma + i\omega)}{Z}\sigma^*(0) \end{aligned}\right\} \qquad (2.B.8)$$

where

$$\left.\begin{aligned} Z = (S + \tfrac{1}{2}\gamma + i\omega)((S + \gamma)(S + \tfrac{1}{2}\gamma - i\omega) + 2|\varepsilon|^2) \\ + 2|\varepsilon|^2(S + \tfrac{1}{2}\gamma - i\omega) \\ = (S - S_1)(S - S_2)(S - S_3) \end{aligned}\right\} \qquad (2.B.9)$$

Since $P(0) = 1$ (i.e. we are only considering the emission process) and $\sigma(0) = \sigma^*(0) = 0$

$$L(p) = 2|\varepsilon|^2 \frac{(S + \frac{1}{2}\gamma)}{SZ} + \frac{(S + \frac{1}{2}\gamma + i\omega)(S + \frac{1}{2}\gamma - i\omega)}{Z} \quad (2.B.10)$$

Writing $Z = (S - x_1)(S - (x_2 + ix_3))(S - (x_2 - ix_3))$, where $s_1 = x_1$, $s_2 = x_2 + ix_3$, $s_3 = x_2 - ix_3$, and x_1, x_2 and x_3 are real, since we shall see that , p and a are real (see eqs. (2.B.13) and (2.B.17), we can separate (2.B.10) into partial fractions and obtain the inverse Laplace transform:

[N.B. If $L(A(t)) = X\frac{1}{S+S_1}$, then $L^{-1} = A(t) = Xe^{-s_1 t}$ when $X \neq X(t)$ follows from (2.B.1), i.e. $L(Xe^{-s_1 t}) = X\int_0^\infty dt e^{-(S+S_1)t} = \frac{X}{S+S_1}$ for $t' = 0$.]

$$P(t) = \left[\frac{|\varepsilon|^2 \gamma}{-x_1(x_2-ix_3)(x_2+ix_3)}\right]$$

$$+ \left[\frac{2|\varepsilon|^2(x_1+\frac{1}{2}\gamma) + x_1((x_1+\frac{1}{2}\gamma) + i\omega)((x_1+\frac{1}{2}\gamma) - i\omega)}{x_1((x_1-x_2) + ix_3)((x_1-x_2) - ix_3}\right] e^{x_1 t}$$

$$+ \left[\frac{2|\varepsilon|^2(x_2+\frac{1}{2}\gamma)-ix_3)+(x_2-ix_3((x_2+\frac{1}{2}\gamma)-i(x_3-\omega))((x_2+\frac{1}{2}\gamma)-i(x_3+\omega))}{-2ix_3(x_2-ix_3)((x_2-x_1)-ix_3)}\right] e^{(x_2-ix_3)t}$$

$$+ \left[\frac{2|\varepsilon|^2((x_2+\frac{1}{2}\gamma)+ix_3)+(x_2+ix_3)((x_2+\frac{1}{2}\gamma)+i(x_3+\omega))((x_2+\frac{1}{2}\gamma)+i(x_3-\omega))}{(x_2+ix_3)((x_2-x_1)+ix_3)2ix_3}\right] e^{(x_2+ix_3)t}$$

(2.B.11)

It is now necessary to solve the equation for Z using the method outlined in Appendix III. Using the same notation as there

$$\left.\begin{aligned} a &= 2\gamma \\ b &= \frac{5}{4}\gamma^2 + \omega^2 + 4|\lambda'|^2 \\ c &= \gamma(\frac{1}{4}\gamma^2 + \omega^2 + 2|\lambda'|^2) \end{aligned}\right\} \quad (2.B.12)$$

N.B. $|\varepsilon|^2 = |\lambda'|^2$ (this is equivalent to $\frac{1}{4}\mathcal{G}^2$ of later chapters when the perturbation is caused by a constant electric field $\underline{E}_D$)

Letting $\mu^2 = \dfrac{|\lambda'|^2}{\omega^2}$ and $\beta = \dfrac{2\gamma}{\omega}$

$$\left.\begin{aligned} a &= \omega\beta \\ b &= \omega^2(\frac{5}{16}\beta^2 + 1 + 4\mu^2) \\ c &= \omega^3\, \tfrac{1}{2}\beta(\frac{1}{16}\beta^2 + 1 + 2\mu^2) \end{aligned}\right\} \quad (2.B.13)$$

We also know from eqs. (III.9), (III.7) and (III.2), that

$$\left.\begin{aligned} S_1 &= x_1 \qquad = (\alpha + \beta) - \frac{1}{3}a \\ S_2 &= x_2 + ix_3 = \tfrac{1}{2}\{(\alpha + \beta) - i\sqrt{3}(\alpha \pm \beta)\} - \frac{1}{3}a \\ S_3 &= x_2 - ix_3 = -\tfrac{1}{2}\{(\alpha + \beta) + i\sqrt{3}(\alpha + \beta)\} - \frac{1}{3}a \\ \text{where} \quad \alpha &= \left[\frac{-G + \sqrt{G^2 + 4H^3}}{2}\right]^{1/3} \\ \beta &= \left[\frac{-G - \sqrt{G^2 + 4H^3}}{2}\right]^{1/3} = -H\alpha^{-1} \\ \text{and} \quad G &= \frac{2}{27}a^3 - \frac{1}{3}ab + c \\ H &= \frac{1}{3}(\frac{-1}{3}a^2 + b) \end{aligned}\right\} \quad (2.B.14)$$

Hence $$G = \frac{1}{27 \times 32}\, \omega^3\beta(\beta^2 - 9 \times 32X_4 + 27 \times 16X_2)$$

and $$H = \frac{1}{144}\, \omega^2(48X_4 - \beta^2) \tag{2.B.15}$$

where $$X_n = 1 + n\mu^2$$

It is necessary to introduce certain approximations in order to solve the cubic equation as cube roots cannot be found exactly. If we consider $\beta << 1$ (i.e. $\omega >> 2\gamma$ or $(\omega_0 - \Omega) >> 2\gamma$, which means the natural line width of the excited state is small compared with the energy separation, when the small frequency shift is included), and ignore then powers higher than 1st. order in β

then

$$\alpha = \frac{\omega}{6}\left[6\{-3\beta(1-2\mu^2) + 4\sqrt{3X_4^3}\}\right]^{1/3}$$

$$\beta = \frac{-\omega^2}{3}\, X_4\, \alpha^{-1} \tag{2.B.16}$$

and can also be written

$$\alpha = \frac{\sqrt{3}}{3}\, \omega\, X_4^{\frac{1}{2}}\left[1 - \frac{\sqrt{3}}{4}\, \beta(1 - 2\mu^2)X_4^{-3/2}\right]^{1/3}$$

$$\beta = \frac{-\sqrt{3}}{3}\, X_4^{\frac{1}{2}}\left[1 - \frac{\sqrt{3}}{4}\, \beta(1 - 2\mu^2)X_4^{-3/2}\right]^{-1/3} \tag{2.B.17}$$

$\therefore$

$$\alpha \pm \beta = \frac{\sqrt{3}}{3}\, X_4^{\frac{1}{2}}\omega\left[1 - \frac{\sqrt{3}}{4}\beta(1-2\mu^2)X_4^{-3/2}\right]^{1/3} \mp \frac{\sqrt{3}}{3}\, X_4^{\frac{1}{2}}\left[1 - \frac{\sqrt{3}}{4}\beta(1-2\mu^2)X_4^{-3/2}\right]^{-1/3} \tag{2.B.18}$$

If this is expanded by means of the Binomial theorem and terms in β^2 and higher are again neglected

$$\alpha + \beta = \frac{-1}{6}\beta\omega(1-2\mu^2)X_4^{-1}$$
$$\alpha - \beta = \frac{2\sqrt{3}}{3}\omega X_4^{\frac{1}{2}} \tag{2.B.19}$$

Hence on substituting in (3.44)

$$x_1 = \frac{-1}{3}\omega\beta\left[1 + \tfrac{1}{2}(1-2\mu^2)X_4^{-1}\right]$$
$$x_2 = \frac{-1}{3}\omega\beta\left[1 - \tfrac{1}{4}(1-2\mu^2)X_4^{-1}\right] \tag{2.B.20}$$
$$x_3 = \omega X_4^{\frac{1}{2}}$$

Hence to 1st. order in β

$$P(t) = (X_1-1)X_2^{-1}$$
$$+ X_2^{-1}X_3X_4^{-1}\exp(-X_2X_4^{-1}\gamma t) \tag{2.B.21}$$
$$+ \left[(X_2-1)X_4^{-1}\cos(X_4^{\frac{1}{2}}(\omega t+\beta X_4^{-2}X_{5/2}))\right]\exp(-\tfrac{1}{2}X_4^{-1}X_6\gamma t)$$

If we now assume $\mu^2 << 1$ (i.e. $|\lambda'|^2 << \omega^2$ or $\hbar|\lambda'| << \hbar\omega$, i.e. the perturbation energy small in comparison to the energy of separation of the atomic levels)[1] and ignore powers higher than 1st. order in μ, we obtain

$$P(t) = \mu^2$$
$$+ (1-3\mu^2)\exp(-(1-2\mu^2)\gamma t) \tag{2.B.22}$$
$$+ 2\mu^2\cos(\omega' t + \beta)\exp(-\tfrac{1}{2}(1+2\mu^2)\gamma t)$$

where $\omega' = (1+2\mu^2)\omega$ [2]

1. Keller and Robiscoe[5] cannot derive an equation for P(t) at all without this approximation, whereas we can, viz. eq. (2.B.21).

2. The correction to ω, accidentally omitted in reference 1, indicates that there is oscillation at the Rabi frequency, since the population is continuously oscillating between the two levels.

[N.B. Neither (2.B.21) nor (2.B.22) contain any dependence on the arbitrary phases θ_ℓ and ϕ.]

If $\lambda' \to 0$, i.e. $\mu^2 \to 0$, then $P(t) = e^{-\gamma t}$, the exponential decay solution. If $\gamma \to 0$, i.e. $\beta \to 0$, $P(t) = (1-2\mu^2) + 2\mu^2 \cos((1+2\mu^2)\omega t + \beta)$ the quantum oscillator solution. This is true also for eq. (2.B.21). Both these results are consistent with the conclusions of Keller and Robiscoe's paper[5] in which they treat essentially the same problem. They use the Wigner-Weisskopf approximation and are limited to a time scale >> the atomic lifetime, γ^{-1}, which we are not (our equations are valid for times $t \gg \omega_0^{-1}$). Eq. (43) of their paper in our notation is:

$$P(t) \simeq \left[1 - 4\mu_0^2 \sin^2 \tfrac{1}{2}\omega_0' t\right] \exp\left[-(1+2\mu^2)\gamma t\right] \exp\left[-\beta_0 \mu^2 \sin \omega_0' t\right]$$
$$+ \left[4\mu_0^2 \sin^2 \tfrac{1}{2}\omega_0' t\right]\left[1 - \exp(-(1+2\mu^2)\gamma t) \exp(-\beta_0 \mu^2 \sin \omega_0' t)\right]$$

where $\mu_0 = \hbar|\lambda'|/\hbar\omega_0$ (2.B.23)

$\beta_0 = \frac{2\gamma}{\omega_0}$

and $\omega_0' = (1+2\mu^2)\omega_0$ i.e. the frequency shift Ω_- is neglected

I.E. $$P(t) \simeq 4\mu_0^2 \sin^2 \tfrac{1}{2}\omega_0' t$$
$$- 4\mu_0^2 \sin^2 \tfrac{1}{2}\omega_0' t \exp(-(1+2\mu^2)\gamma t)\exp(-\beta_0 \mu^2 \sin \omega_0' t) \qquad (2.B.24)$$

They point out that parameters μ_0 and β_0 respectively give the strength of the external perturbation, $\hbar|\lambda'|$, and the radiation interaction, $2\hbar\gamma$, with respect to the unperturbed binding energy, $\hbar\omega_0$. ω_0' and $\gamma' = (1+2\mu^2)\gamma$ are the perturbed counterparts of the j-i state separation ω_0 and the j-state decay rate γ. From their equation they conclude that when $2\hbar\gamma \to 0$

$$P(t) \to 1 - 4\mu_o^2\sin^2\tfrac{1}{2}\omega_o't \tag{2.B.25}$$

while when $\hbar|\lambda'| \to 0$

$$P(t) \to e^{-\gamma t} \quad \text{the W-W solution} \tag{2.B.26}$$

We have shown that our equation will also tend to (2.B.26) for $\lambda' \to 0$ and that when $\gamma \to 0$ we get a quantum oscillation solution, although not of exactly the same form as that of Keller and Robiscoe [5].

The main differences between eqs. (2.B.22) and (2.B.23) are

(i) for $\gamma t >> 1$ (2.B.22) gives a steady state solution $P(t) = \mu^2$ whereas (2.B.23) gives a quantum oscillation solution $P(t) = \mu_o^2\left[4\sin^2\tfrac{1}{2}\omega_o't\right]$ described in fig. 3 of ref. [5],

(ii) the 3rd. term in (2.B.22) decays at $\tfrac{1}{2}(1+2\mu^2)$, nearly half the rate of the second, $(1-2\mu^2)\gamma$, owing to the mixing of the diagonal (P) and off-diagonal (σ) matrix elements caused by coupling with the external perturbation. (When there is no perturbation present we saw that $\sigma(t)$ decayed at half the rate of $P(t)$. In (2.B.23) the decay terms all decay at the same rate, namely $(1+2\mu^2)\gamma$. This is noticeable also in eq. (2.B.21) where the rates are $\tfrac{1}{2}X_4^{-1}X_6$ and $X_4^{-1}X_2$.

Keller and Robiscoe [5] expect the new type of modulation factor which remains for $\gamma t >> 1$ will also be found for a 3-level quantum system in which the same external perturbation couples both the upper 2 levels and the lower 2 levels. In Fontana and Lynch's [6] paper, they consider the radiative decay of an atom with 2 excited states coupled by an external perturbation but there is no coupling between the lower 2 levels but this could be considered on the basis of their theory to see whether Keller and Robiscoe's hypothesis holds, but we doubt it, since Fontana and Lynch's equations for the 2-level atom embody similar characteristic features of the decay process described by our eq. (2.B.22) and noted in (i) and (ii), viz. their eq. (24):

$$
\begin{aligned}
|b_f(t)|^2 = {} & |a_1(\epsilon)|^2\{1+e^{-(X_o+X_j-Q)t} - 2e^{-\frac{1}{2}(X_o+X_j-Q)t}\cos\left[\tfrac{1}{2}(\epsilon-P)t\right]\} \\
& + |a_2(\epsilon)|^2\{1+e^{-(X_o+X_j+Q)t} - 2e^{-\frac{1}{2}(X_o+X_j+Q)t}\cos\left[\tfrac{1}{2}(\epsilon+P)t\right]\} \\
& + 2a(\epsilon)\{e^{-(X_o+X_j)t}\cos(Pt+\theta)+\cos\theta \qquad (2.B.27) \\
& - e^{-\frac{1}{2}(X_oX_j-Q)t}\cos\left[\tfrac{1}{2}(\epsilon-p)t+\theta\right] \\
& - e^{-\frac{1}{2}(X_o+X_j+Q)t}\cos\left[\tfrac{1}{2}(\epsilon+p)t-\theta\right]
\end{aligned}
$$

which is equivalent to $(1-P(t))$ when their intermediate level is ignored. This expression contains pure exponential decay rates $(X_o+X_j\mp Q)$ and modulated decay rates of $\frac{1}{2}(X_o+X_j\mp Q)$ i.e. half the pure one and, (X_o+X_j). In our case we get a pure exponential decay rate of $(1-2\mu^2)\gamma$ and a modulated one of $\frac{1}{2}(1+2\mu^2)\gamma$ which is approximately half. For $(X_o+X_j\mp Q)t \gg 1$, $\frac{1}{2}(X_o+X_j\mp Q)t \gg 1$ and $(X_o+X_j)t \gg 1$

$$|b_f(t)|^2 \doteq |a_1(\epsilon)|^2 + |a_2(\epsilon)|^2 + 2a(\epsilon)\cos\theta$$

i.e. it also reaches a steady state value and not an oscillating one.

N.B. Fontana and Lynch use the Heitler-Ma formalism [40],[41] but Keller and Robiscoe do not treat the classical external perturbation exactly as we do.

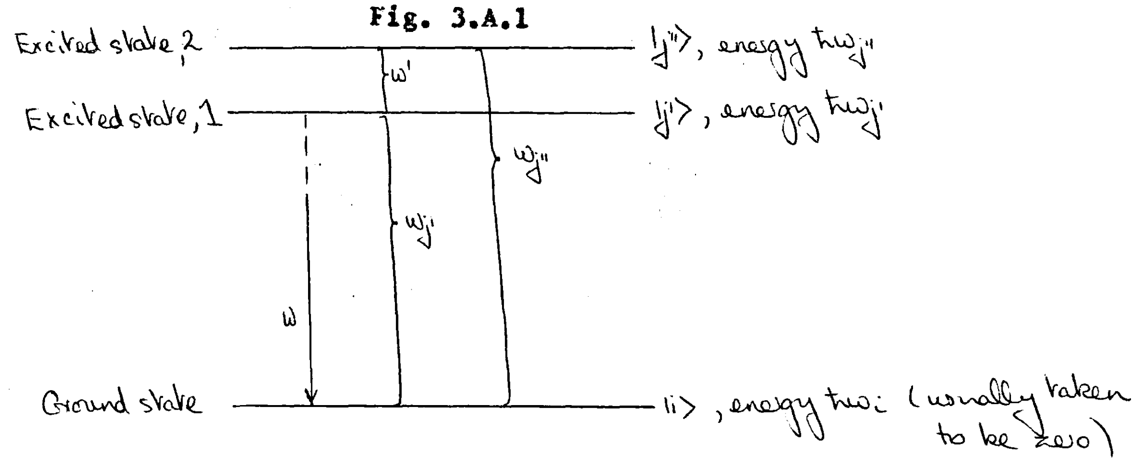
CHAPTER III

AN ATOM WITH TWO CLOSE-LYING EXCITED STATES COUPLED ONLY BY THE RADIATION FROM ITS TRANSITIONS[4]

A. Heisenberg equations of motion

We now go on to consider an atom (lying at the prigin of co-ordinates so that $\underline{r} = \underline{0}$) which has 2 excited states $|j''\rangle$ and $|j'\rangle$ coupled to a ground state $|i\rangle$ by a quantised multi-mode e.m. field. We shall not be introducing any external classical perturbation or considering the initial radiation state to be anything other than a vacuum state and so the problem we are treating is simply that of undriven spontaneous emission. We shall denote the energy separations between states $|j''\rangle$ and $|i\rangle$ by $\epsilon_{j''} - \epsilon_i \equiv \omega_{j''}$ and between states $|j'\rangle$ and and $|i\rangle$ by $\epsilon_{j'} - \epsilon_i \equiv \omega_j'$ and between $|j''\rangle$ and $|j'\rangle$ by $\epsilon_{j''} - \epsilon_{j'} \equiv \omega' = \omega_{j''} - \omega_{j'}$

Fig. 3.A.1



We can write the Hamiltonian, in the dipole approximation, as

$$
\begin{aligned}
H = \hbar \sum_{\alpha=i,j',j''} \epsilon_\alpha P_{\alpha\alpha} + \frac{1}{8\pi}\int (|\underline{E}|^2 + |\underline{B}|^2)d^3r &- \underline{E}\cdot\underline{p}_{j'i}(P_{ij'} + P_{ij'}{}^\dagger) \\
&- \underline{E}\cdot\underline{p}_{j''i}(P_{ij''} + P_{ij''}{}^\dagger) \qquad (3.A.1) \\
&- \underline{E}\cdot\underline{p}_{j''j'}(P_{j'j''} + P_{ij''}{}^\dagger)
\end{aligned}
$$

If we now neglect direct transitions between $|j''\rangle$ and $|j'\rangle$, by assuming the dipole moment $\underline{p}_{j'j''} = 0$ (i.e. if quantum numbers for levels j' and j" do not satisfy the condition for allowed transitions, namely $\ell_{j'} - \ell_{j''} = \pm 1$) we obtain, on substituting for $\underline{E}$ and $\underline{B}$ from eqs. (1.B.58) and (1.B.59) when $\underline{r} = 0$

$$H = \hbar \sum_{\alpha=i,j',j''} \epsilon_\alpha P_{\alpha\alpha} + \hbar \sum_{\ell,\sigma} \omega_\ell a^\dagger_{\ell\sigma} a_{\ell\sigma} - \hbar \sum_{\ell,\sigma} g_{\ell\sigma j'}(S_1 a_{\ell\sigma} + a^\dagger_{\ell\sigma} S_1) - \hbar \sum_{\ell,\sigma} g_{\ell\sigma j''}(S_2 a_{\ell\sigma} + a^\dagger_{\ell\sigma} S_2) \tag{3.A.2}$$

where $\quad S_1(t) = P_{ij'}(t) + P^\dagger_{ij'}(t)$

and $\quad S_2(t) = P_{ij''}(t) + P^\dagger_{ij''}(t)$

Also $\quad g_{\ell\sigma j'} = \sqrt{\dfrac{2\pi\omega_{\ell j'}}{\hbar V}}\, \hat{e}_{\ell\sigma}\cdot\underline{p}_{j'i} = K_{\ell j'}\hat{e}_{\ell\sigma}\cdot\underline{p}_{j'i}$

$$\text{and} \quad g_{\ell\sigma j''} = \sqrt{\frac{2\pi\omega_{\ell j''}}{\hbar V}}\, \hat{e}_{\ell\sigma}\cdot\underline{p}_{j''i} = K_{\ell j''}\hat{e}_{\ell\sigma}\cdot\underline{p}_{j''i} \tag{3.A.3}$$

where $\quad K_{\ell j'} = \sqrt{\dfrac{2\pi\omega_{\ell j''}}{\hbar V}}$ and $K_{\ell j''} = \sqrt{\dfrac{2\pi\omega_{\ell j''}}{\hbar V}}$

and $\underline{p}_{j'i}$ and $\underline{p}_{j''i}$ are real dipole matrix elements

$\underline{p}_{j'i} = \langle j'|e\underline{x}|i\rangle$, $\underline{p}_{j''i} = \langle j''|e\underline{x}|i\rangle$.

[N.B. Subscripts on ω_ℓ indicate whether the frequencies are for photon transitions between j' and i or between j" and i.]

The remaining terms have their usual meaning. If we omit high frequency terms $P_{ij'}a_{\ell\sigma}$, $P_{ij''}a_{\ell\sigma}$, $a^\dagger_{\ell\sigma}P^\dagger_{ij'}$, $a^\dagger_{\ell\sigma}P^\dagger_{ij''}$ then in the R.W.A. the Hamiltonian becomes

$$H = \hbar \sum_{\alpha=i,j',j''} \epsilon_\alpha P_{\alpha\alpha} + \hbar \sum_{\ell,\sigma} \omega_\ell a^\dagger_{\ell\sigma} a_{\ell\sigma} - \hbar \sum_{\ell,\sigma} g_{\ell\sigma j'} (P^\dagger_{ij'} a_{\ell\sigma} + a^\dagger_{\ell\sigma} P_{ij'}) - \hbar \sum_{\ell,\sigma} g_{\ell\sigma j''} (P^\dagger_{ij''} a_{\ell\sigma} + a^\dagger_{\ell\sigma} P_{ij''}) \tag{3.A.4}$$

Using Hamiltonian (3.A.4) we obtain the following Heisenberg equations of motion

$$\dot{a}_{\ell\sigma} = -i\omega_\ell a_{\ell\sigma} + ig_{\ell\sigma j'} P_{ij'} + ig_{\ell\sigma j''} P_{ij''} \tag{3.A.5}$$

$$\dot{P}_{ij''} = -i\omega_{j''} P_{ij''} - iP_{j'j''} \sum_{\ell,\sigma} g_{\ell\sigma j'} a_{\ell\sigma} - i(P_{j''j''} - P_{ii}) \sum_{\ell,\sigma} g_{\ell\sigma j''} a_{\ell\sigma} \tag{3.A.6}$$

$$\dot{P}_{ij'} = -i\omega_{j'} P_{ij'} - iP_{j''j'} \sum_{\ell,\sigma} g_{\ell\sigma j''} a_{\ell\sigma} - i(P_{j'j'} - P_{ii}) \sum_{\ell,\sigma} g_{\ell\sigma j'} a_{\ell\sigma} \tag{3.A.7}$$

Equation (3.5) has formal solution

$$a_{\ell\sigma}(t) = a_{\ell\sigma}(0) e^{-i\omega_\ell t} + ig_{\ell\sigma j'} \int_0^t dt' e^{-i\omega_\ell (t-t')} P_{ij'}(t') + ig_{\ell\sigma j''} \int_0^t dt'' e^{-i\omega_\ell (t-t'')} P_{ij''}(t'') \tag{3.A.8}$$

Consider

(i) $\quad \sum_{\ell,\sigma} g_{\ell\sigma j'} a_{\ell\sigma}(t)$ and (3.A.9a)

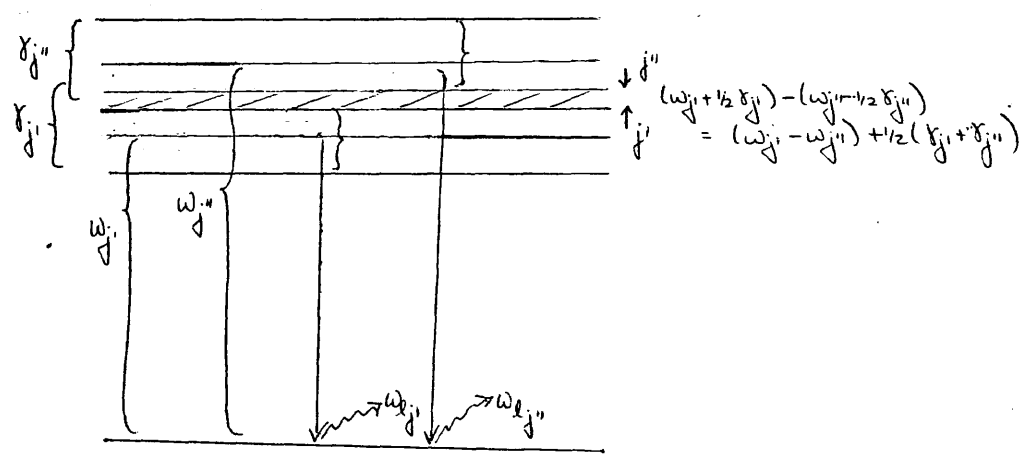
(ii) $\quad \sum_{\ell,\sigma} g_{\ell\sigma j''} a_{\ell\sigma}(t).$ (3.A.9b)

As in Chapter II when $V \to \infty$

$$
\left.\begin{aligned}
&\text{(i)}\quad \sum_{\ell,\sigma} g_{\ell\sigma j'} a_{\ell\sigma}(t) - \frac{1}{\hbar}\sum_{\ell,\sigma} \underline{E}_{+}^{(0)}(t)\cdot\underline{p}_{j'i} \\
&= +\, i\, \frac{p_{j'i}^{2}}{4\pi^{2}C^{3}\hbar}\int_{0}^{\infty} d\omega\,\omega^{3}\int_{0}^{4\pi} d\Omega_{\hat{k}}\sum_{\sigma=1}^{2}\left(\hat{e}_{\ell\sigma}\cdot\hat{\underline{p}}_{j'i}\right)^{2} \\
&\quad \times \int_{0}^{t} dt'\, e^{-i\omega(t-t')} P_{ij'}(t') \\
&\quad +\, i\, \frac{p_{j'i}p_{j''i}}{4\pi^{2}C^{3}\hbar}\int_{0}^{\infty} d\omega\,\omega^{3}\int_{0}^{4\pi} d\Omega_{\hat{k}}\sum_{\sigma=1}^{2}\left(\hat{e}_{\ell\sigma}\cdot\hat{\underline{p}}_{j'i}\right)\left(e_{\ell\sigma}\cdot\hat{\underline{p}}_{j''i}\right) \\
&\quad \times \int_{0}^{t} dt''\, e^{-i\omega(t-t'')} P_{ij''}(t'')
\end{aligned}\right\} \quad (3.A.9c)
$$

N.B. In the 2nd term on the R.H.S. we have integrated over frequencies comments photon transitions between levels j'' and i and i and j' and vice versa, i.e. $g_{\ell\sigma j'}g_{\ell\sigma j''} \propto \sqrt{\omega_{\ell j'}\omega_{\ell j''}}$ is taken as $\propto \omega_\ell$, for a certain range of frequencies common to both photon transitions between levels j'' and i and j' and i.

Fig. 3.A.2



For the shaded region where levels j'' and j' overlap $\omega_{\ell j'} = \omega_{\ell j''} = \omega_\ell$. The area of overlap is common to both levels j' and j'' so that if a photon is absorbed to or emitted from that region it can be considered as going to or originating from either level and we can say that photon is (1) emitted by level j'' and absorbed by j' or (2) a photon is emitted by level j' and absorbed by j''.

$\omega_{\ell j'} = \omega_{\ell j''} = \omega_\ell$ for frequencies $\omega = \omega_{j'} + \frac{1}{2}\gamma_{j'}$ to $\omega_{j''} - \frac{1}{2}\gamma_{j''}$

But when $\omega_{\ell j'}$ has values $\omega_{j'} - \frac{1}{2}\gamma_{j'}$ up to $\omega_{j''} - \frac{1}{2}\gamma_{j''}$

and when $\omega_{\ell j''}$ has values $\omega_{j'} + \frac{1}{2}\gamma_{j'}$ up to $\omega_{j''} + \frac{1}{2}\gamma_{j''}$

$\omega_{\ell j'} \neq \omega_{\ell j''}$ and the photons $\hbar\omega_{\ell j'}$ and $\hbar\omega_{\ell j''}$ are NOT common to both levels j' and j".

In calculating $\Gamma_{j'}$ and $\Gamma_{j''}$ we integrate only over the shaded region and let ω_ℓ take values 0 to ∞ in the free space limit $V \to \infty$ when the spectrum of ω_ℓ becomes continuous.

$$\underline{\text{To find } \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'i})(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''i})} = (e\ {}_1\cdot p\ {}_j \tag{3.A.10}$$

$$\underline{= (\hat{e}_{\ell 1}\cdot\hat{p}_{j'i})(\hat{e}_{\ell 1}\cdot\hat{p}_{j''i}) + (\hat{e}_{\ell 2}\cdot\hat{p}_{j'i})(\hat{e}_{\ell 2}\cdot\hat{p}_{j''i})}$$

<u>when $p_{j'i}$ and $p_{j''i}$ are in different directions.</u>

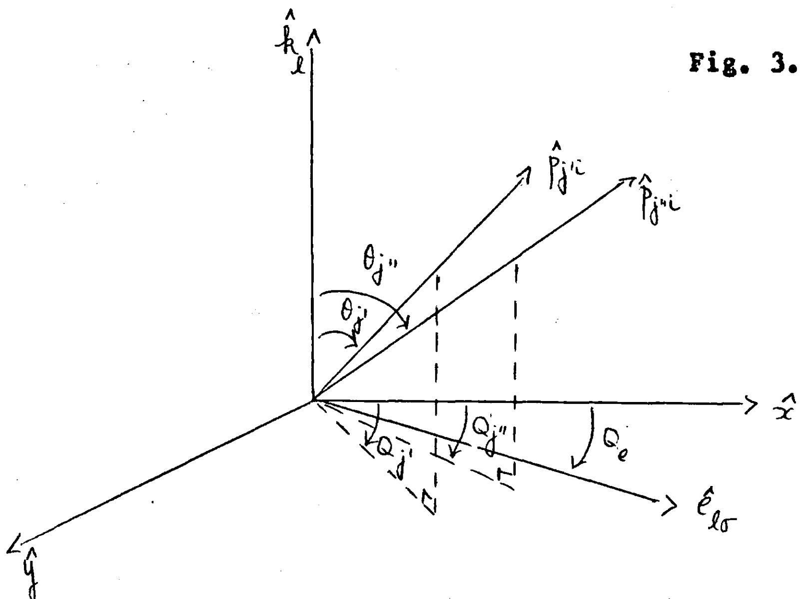


Fig. 3.A.3

As in Chapter II we choose rectangular co-ordinate axes $\hat{x}$, $\hat{y}$ and $\hat{k}_\ell$. Then we can write the vectors in terms of spherical polars [39] as

$$\hat{e}_{\ell\sigma} = \hat{x}\cos Q_e + \hat{y}\sin Q_e$$

$$\hat{p}_{j'i} = \hat{x}\sin\theta_{j'}\cos Q_{j'} + \hat{y}\sin\theta_{j'}\sin Q_{j'} + \hat{k}\cos\theta_{j'} \qquad (3.A.11)$$

$$\hat{p}_{j''i} = \hat{x}\sin\theta_{j''}\cos Q_{j''} + \hat{y}\sin\theta_{j''}\sin Q_{j''} + \hat{k}\cos\theta_{j''}$$

$\therefore$

$$\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'i} = \cos Q_e\sin\theta_{j'}\cos Q_{j'} + \sin Q_e\sin\theta_{j'}\sin Q_{j'}$$

$$= \sin\theta_{j'}\cos(Q_e - Q_{j'})$$

$$\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''i} = \cos Q_e\sin\theta_{j''}\cos Q_{j''} + \sin Q_e\sin\theta_{j''}\sin Q_{j''} \qquad (3.A.12)$$

$$= \sin\theta_{j''}\cos(Q_e - Q_{j''})$$

$$(\hat{e}_{\ell 1}\cdot\hat{p}_{j'i})(\hat{e}_{\ell 1}\cdot\hat{p}_{j''i}) = \sin\theta_{j'}\sin\theta_{j''}\cos(Q_{e1} - Q_{j'})\cos(Q_{e1} - Q_{j''})$$

$$(\hat{e}_{\ell 2}\cdot\hat{p}_{j'i})(\hat{e}_{\ell 2}\cdot\hat{p}_{j''i}) = \sin\theta_{j'}\sin\theta_{j''}\cos(Q_{e2} - Q_{j'})\cos(Q_{e2} - Q_{j''}) \qquad (3.A.13)$$

$$= \sin\theta_{j'}\sin\theta_{j''}\cos((Q_{e1}+270) - Q_{j'})\cos((Q_{e1}+270) - Q_{j''})$$

because $Q_{e2} = Q_{e1} + 270^{o}$ since $e_{\ell 1}$ and $e_{\ell 2}$ are perpendicular.

Also $$\hat{p}_{j'i}\cdot\hat{p}_{j''i} = \sin\theta_{j'}\sin\theta_{j''}\cos(Q_{j'} - Q_{j''}) + \cos\theta_{j'}\cos\theta_{j''} \qquad (3.A.14)$$

$$\sum_{\sigma}(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'i})(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''i}) = \sin\theta_{j'}\sin\theta_{j''}\Big[\cos(Q_{e1} - Q_{j'})\cos(Q_{e1} - Q_{j''})$$

$$+ \sin(Q_{j'} - Q_{e1})\sin(Q_{j''} - Q_{e1})\Big] \qquad (3.A.15)$$

since $\cos(270^{\circ} - a) = -\sin(180^{\circ} - a) = -\cos(90^{\circ} - a) = -\sin a$

$\therefore$

$$\sum_\sigma(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'i})(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''i}) = \sin\theta_{j'}\sin\theta_{j''}\cos\left[(Q_{e_1}-Q_{j'})-(Q_{e_1}-Q_{j''})\right]$$

$$= \sin\theta_{j'}\sin\theta_{j''}\cos(Q_{j''}-Q_{j'})$$

$$= \sin\theta_{j'}\sin\theta_{j''}\cos(Q_{j'}-Q_{j''}) \qquad (3.A.16)$$

$$= \hat{p}_{j'i}\cdot\hat{p}_{j''i} - \cos\theta_{j'}\cos\theta_{j''}$$

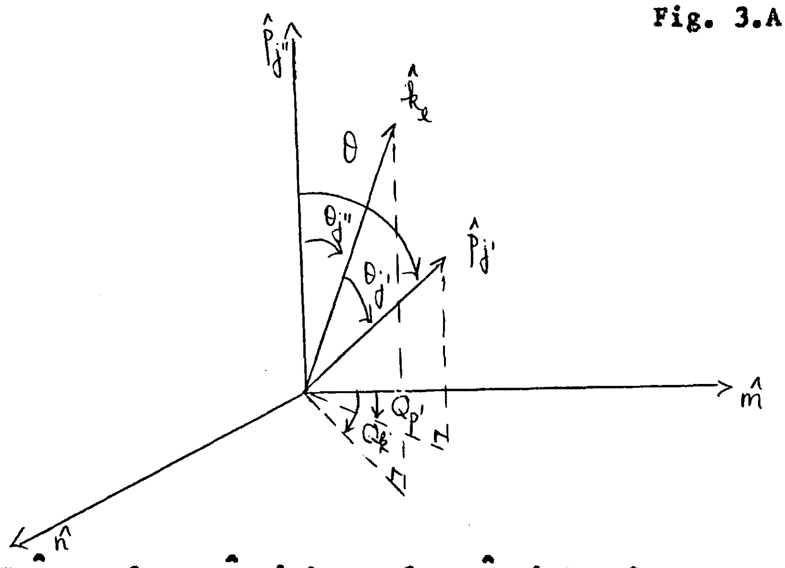
$$= \cos\theta - \underline{\cos\theta_{j'}}\cos\theta_{j''}$$

Now in order to find

$$I = \int_0^{4\pi} d\Omega_{\hat{k}} \sum_\sigma(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'i})(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''i}) = \int_0^{4\pi} d\Omega_{\hat{k}}\{\hat{p}_{j'i}\cdot\hat{p}_{j''i}-\cos\theta_{j'}\cos\theta_{j''}\} \qquad (3.A.17)$$

we need to consider the adjoint figure and define vectors with respect to the new axes $\hat{m}$, $\hat{n}$, $\hat{p}_{j''}$ (alternatively $\hat{m}$, $\hat{n}$, $\hat{p}_{j'}$) where $\hat{m}$ and $\hat{n}$ can be $\hat{e}_{\ell_1}$ and $\hat{e}_{\ell_2}$.

Fig. 3.A.4



$$\hat{k}_\ell = \hat{p}_{j''}\cos\theta_{j''} + \hat{m}\sin\theta_{j''}\cos Q_k + \hat{n}\sin\theta_{j''}\sin Q_k$$

$$\hat{p}_{j'} = \hat{p}_{j''}\cos\theta + \hat{m}\sin\theta\cos Q_{p'} + \hat{n}\sin\theta\sin Q_{p'} \qquad (3.A.18)$$

$$\therefore \quad \hat{k}_{\ell}\cdot\hat{p}_{j'} = \underline{\cos\theta_{j'}} = \cos\theta_{j''}\cos\theta + \sin\theta_{j''}\sin\theta\cos(Q_k - Q_{p'}) \qquad (3.A.19)$$

Hence

$$\sum_{\sigma}(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j'})(\hat{e}_{\ell\sigma}\cdot\hat{p}_{j''}) = \cos\theta - \cos\theta_{j''}\left[\cos\theta_{j''}\cos\theta + \sin\theta_{j''}\sin\theta\cos(Q_k - Q_{p'})\right]$$

$$= \cos\theta(1-\cos^2\theta_{j''}) - \sin\theta\cos\theta_{j''}\sin\theta_{j''}\cos(Q_k - Q_{p'}) \qquad (3.A.20)$$

$$= \cos\theta\sin^2\theta_{j''} - \sin\theta.\tfrac{1}{2}\sin 2\theta_{j''}.\cos(Q_k - Q_{p'})$$

$$\therefore \; I = \int_0^{4\pi} d\Omega_{\hat{k}}\left[\cos\theta\sin^2\theta_{j''} - \sin\theta\tfrac{1}{2}\sin 2\theta_{j''}\cos(Q_k - Q_{p'})\right]$$

$$= \int_0^{2\pi} dQ_k \int_0^{\pi} dQ_{j''}\sin\theta_{j''}\left[\cos\theta\sin^2\theta_{j''} - \sin\theta\tfrac{1}{2}\sin 2\theta_{j''}\cos(Q_k - Q_{p'})\right] \qquad (3.A.21)$$

The 2nd. term gives zero on integration over Q_k since

$$\int_0^{2\pi} dQ_k(\cos Q_k\cos Q_{p'} - \sin Q_k\sin Q_{p'})$$

$$= \cos Q_{p'}\int_0^{2\pi} dQ_k\cos Q_k - \sin Q_{p'}\int_0^{2\pi} dQ_k\sin Q_k \qquad (3.A.22)$$

$$= \cos Q_{p'}(0) - \sin Q_{p'}(0)$$

$$\therefore \qquad I = \cos\theta\int_0^{2\pi} dQ_k\int_0^{\pi} d\theta_{j''}\sin\theta_{j''}(1-\cos^2\theta_{j''})$$

$$= (\hat{p}_{j'i}\cdot\hat{p}_{j''i})\,\frac{8\pi}{3} \quad \text{as in (2.A.13)} \qquad (3.A.23)$$

Hence (3.A.9) becomes, when we proceed as in Chapter II eqs. (2.A.15) etc., and take $P_{ij'}(t') = P_{ij'}(t)e^{-i\omega_{j'}(t'-t)}$

$$P_{ij''}(t') = P_{ij''}(t)e^{-i\omega_{j''}(t'-t)}$$

and $\omega_{j'}t \gg 1$ and $\omega_{j''}t \gg 1$

$$\sum_{\ell,\sigma} g_{\ell\sigma j'} a_{\ell\sigma}(t) = \frac{1}{\hbar} \sum_{\ell,\sigma} \underline{E}_+^{(0)}(t).\underline{p}_{j'} + i(\tfrac{1}{2}\gamma_{j'} - i\Omega_{\underline{j}'})P_{ij'}(t) + i(\tfrac{1}{2}\Gamma_{j''} - i\Omega'_{\underline{j}''})P_{ij''}(t) \qquad (3.A.24a)$$

$$\text{where } \gamma_{j'} = \frac{4p_{j'}^2\omega_{j'}^3}{3\hbar c^3}, \quad \Omega_{\underline{j}'} = \frac{\gamma_{j'}}{\omega_{j'}^3}\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega-\omega_{j'}};$$

$$\text{and } \Gamma_{j''} = \frac{4(\underline{p}_{j'}\cdot\underline{p}_{j''})\omega_{j''}^3}{3\hbar c^3}, \quad \Omega'_{\underline{j}''} = \frac{\Gamma_{j''}}{\omega_{j''}^3}\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega-\omega_{j''}}. \qquad (3.A.24b)$$

N.B. $\underline{p}_{j'} = \underline{p}_{j'i}$ and $\underline{p}_{j''} = \underline{p}_{j''i}$. In deriving $\Gamma_{j'}$ and $\Gamma_{j''}$ we have integrated over those frequencies which are common to photon transitions between levels j" to i and i to j' and vice versa. If levels j" and j' are far apart these cross terms and hence $\Gamma_{j'}$ and $\Gamma_{j''}$ can be neglected. For them to be important ω' must be of the order of the natural line width of level j" or j'. Similarly

$$\sum_{\ell,\sigma} g_{\ell\sigma j''} a_{\ell\sigma}(t) = \frac{1}{\hbar} \sum_{\ell,\sigma} \underline{E}_+^{(0)}(t).\underline{p}_{j''} + i(\tfrac{1}{2}\Gamma_{j'} - i\Omega'_{\underline{j}'})P_{ij'}(t) + i(\tfrac{1}{2}\gamma_{j''} - i\Omega_{\underline{j}''})P_{ij''}(t) \qquad (3.A.25a)$$

$$\text{where } \gamma_{j''} = \frac{4p_{j''}^2\omega_{j''}^3}{3\hbar c^3}, \quad \Omega_{\underline{j}''} = \frac{\gamma_{j''}}{\omega_{j''}^3}\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega-\omega_{j''}}$$

$$\text{and } \Gamma_{j'} = \frac{4(\underline{p}_{j'}\cdot\underline{p}_{j''})\omega_{j'}^3}{3\hbar c^3}, \quad \Omega'_{\underline{j}'} = \frac{\Gamma_{j'}}{\omega_{j'}^3}\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega-\omega_{j'}} \qquad (3.A.25b)$$

$$\text{Therefore } \frac{\gamma_{j'}}{\gamma_{j''}} = \frac{p_{j'}^2\omega_{j'}^3}{p_{j''}^2\omega_{j''}^3} \text{ and } \frac{\Gamma_{j'}}{\Gamma_{j''}} = \frac{\omega_{j'}^3}{\omega_{j''}^3} \qquad (3.A.26)$$

Substituting eqs. (3.A.24a) and (3.A.25a) in eqs. (3.A.6) and (3.A.7) we obtain

$$\dot{P}_{ij''}(t) = -(\tfrac{1}{2}\gamma_{j''} + i(\omega_{j''}-\Omega_{\underline{j''}}))P_{ij''} - (\tfrac{1}{2}\Gamma_{j'}-i\Omega'_{\underline{j'}})P_{ij'}$$
$$-\frac{i}{\hbar}\sum_{\ell,\sigma}\left[P_{j'j''}\underline{E}_{+}^{(0)}(t).\underline{p}_{j'} + (P_{j''j''}-P_{ii})\underline{E}_{+}^{(0)}(t).\underline{p}_{j''}\right] \quad (3.A.27)$$

Similarly

$$\dot{P}_{ij'}(t) = -(\tfrac{1}{2}\gamma_{j'} + i(\omega_{j'}-\Omega_{\underline{j'}}))P_{ij'} - (\tfrac{1}{2}\Gamma_{j''}-i\Omega'_{\underline{j''}})P_{ij''}$$
$$-\frac{i}{\hbar}\sum_{\ell,\sigma}\left[P_{j''j'}\underline{E}_{+}^{(0)}(t).\underline{p}_{j''} + (P_{j'j'}-P_{ii})\underline{E}_{+}^{(0)}(t).\underline{p}_{j'}\right] \quad (3.A.28)$$

These eqs. may also be written in terms of $q_{j'}$ and $q_{j''}$, where

$$q_{j'} = \frac{1}{\hbar}\sum_{\ell,\sigma}\underline{E}_{+}^{(0)}(t).\underline{p}_{j'} = \sum_{\ell,\sigma} g_{\ell\sigma,j'}a_{\ell\sigma}(0)e^{-i\omega_\ell t}$$
$$q_{j''} = \frac{1}{\hbar}\sum_{\ell,\sigma}\underline{E}_{+}^{(0)}(t).\underline{p}_{j''} = \sum_{\ell,\sigma} g_{\ell\sigma,j''}a_{\ell\sigma}(0)e^{-i\omega_\ell t} \quad (3.A.29)$$

N.B. $q_{j'}$ and $q_{j''}$ are time dependent.

Also the frequency shifts $\Omega_{\underline{j'}}, \Omega_{\underline{j''}}, \Omega'_{\underline{j'}}, \Omega'_{\underline{j''}}$ can be absorbed into the definitions of $\omega_{j'}, \omega_{j''}, \Gamma_{j'}$ and $\Gamma_{j''}$ so that

$$\omega_{j'_o} = \omega_{j'} - \Omega_{\underline{j'}}$$
$$\omega_{j''_o} = \omega_{j''} - \Omega_{\underline{j''}}$$
$$\tfrac{1}{2}\Gamma_{j'_o} = \tfrac{1}{2}\Gamma_{j'} - i\Omega'_{\underline{j'}} \quad (3.A.30)$$
$$\tfrac{1}{2}\Gamma_{j''_o} = \tfrac{1}{2}\Gamma_{j''} - i\Omega'_{\underline{j''}}$$

In fact these frequency shifts can be neglected completely and can be reintroduced if required at a later stage.

B. The line-shape of spontaneous emission

In order to calculate the line-shape of spontaneous emission of the atom, we use a method based on the approaches of Lehmberg[1],[2] and Glauber[48],[49],[50] in order to avoid use of the Markoff approximation and the fluctuation regression theorem. The spectral profile of the atomic decay is essentially given by the Foiner-transform of a 2-time atomic correlation function. This is explained in ref. 48, where Glauber says that the energy spectrum can be derived from the 1st. order correlation function. We shall outline how this is done below, using Glauber's references.

If we substitute in the L.H.S. of eq. (10.11) of ref. [49] for $\underline{E}^{(-)}(\underline{r},t)$ and $\underline{E}^{(0)}(\underline{r},t')$ expanded in plane-wave modes, using our notation,

$$\text{i.e.}\quad \underline{E}(\underline{r},t) = (\underline{E}^{(+)}(r,t) + \underline{E}^{(-)}(\underline{r},t) \qquad \text{from (II.11)} \tag{3.B.1}$$

$$\left.\begin{aligned} \text{where } \underline{E}^{(+)}(\underline{r},t) &= \sum_{\ell,\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\,\hat{e}_{\ell\sigma}a_{\ell\sigma}(t)e^{i\underline{k}_\ell\cdot\underline{r}} \\ \underline{E}^{(-)}(\underline{r},t) &= \sum_{\ell,\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\,\hat{e}_{\ell\sigma}a^{\dagger}_{\ell\sigma}(t)e^{-i\underline{k}_\ell\cdot\underline{r}} \end{aligned}\right\}\ \text{from (II.10)} \tag{3.B.2}$$

we obtain

$$\left.\begin{aligned} I &= 2\int \underline{E}^{(-)}(\underline{r},t).\underline{E}^{(+)}(\underline{r},t')d\underline{r} \\ &= 2\int \sum_{\ell,\sigma}\ \sum_{\ell',\sigma'} \frac{2\pi\hbar}{V}(\omega_\ell\omega_{\ell'})^{\frac{1}{2}}(\hat{e}_{\ell\sigma}.\hat{e}_{\ell'\sigma'})e^{-i(\underline{k}_\ell-\underline{k}_{\ell'}).\underline{r}} \\ &\quad\times a^{\dagger}_{\ell\sigma}(t)a_{\ell'\sigma'}(t')d\underline{r} \end{aligned}\right\} \tag{3.B.3}$$

According to Louisell[35] p. 155, eq. (4.88)

$$I' = \int_{\text{cavity}} \underline{U}^{*}_{\ell\sigma}(t).\underline{U}_{\ell'\sigma'}(\underline{r})d\tau = \delta_{\ell\ell'}\delta_{\sigma\sigma'}$$

where $\underline{U}_{\ell\sigma}(\underline{r}) = \dfrac{\hat{e}_{\ell\sigma}e^{i\underline{k}_{\ell}.\underline{r}}}{\sqrt{\tau}}$

and $\underline{U}^{*}_{\ell\sigma}(\underline{r}) = \dfrac{\hat{e}_{\ell\sigma}e^{-i\underline{k}_{\ell}.\underline{r}}}{\sqrt{\tau}}$

(where τ is the normalisation volume) (3.B.4)

since $$I' = \frac{(\hat{e}_{\ell\sigma}.\hat{e}_{\ell'\sigma'})}{\tau} \int_{\text{cavity}} e^{-i(\underline{k}_{\ell}-\underline{k}_{\ell'}).\underline{r}}d\underline{r}$$

$$= (\hat{e}_{\ell\sigma}.\hat{e}_{\ell'\sigma'})\ \delta_{\ell\ell'}$$

$$= \hat{e}_{\ell\sigma}.\hat{e}_{\ell\sigma'}\ \delta_{\ell\ell'}$$

$$= \delta_{\sigma\sigma'}\delta_{\ell\ell'} \text{ (since } e_{\ell\sigma}.e_{\ell\sigma'} = \delta_{\sigma\sigma'}\text{(B.6.13a)}$$

Hence $$I = (4\pi)\sum_{\ell,\sigma} \hbar\omega_{\ell}\, a^{\dagger}_{\ell\sigma}(t)a_{\ell\sigma}(t') \text{ for \underline{free} fields } (V\to\infty) \quad (3.B.5)$$

[This is 4π x Glauber's expression (10.11) since Glauber is using the same units as Mollow viz. the unrationalised mixed Gaussien c.g.s. system (Chapter I, Section B6).]

Now taking a statistical average of both sides of this equation

$$\tfrac{1}{2}\langle I\rangle = \int \langle\underline{E}^{(-)}(\underline{r},t).\underline{E}^{(+)}(\underline{r},t')\rangle d\underline{r} = \sum_{\mu}\int G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r},t')d\underline{r} \quad (3.B.6)$$

i.e. the 1st. order 2-time correlation function

$$\left.\begin{aligned} G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r},t') &= \langle \underline{E}^{(-)}(\underline{r},t).\underline{E}^{(+)}(\underline{r},t')\rangle \\ &= \mathrm{Tr}\{\rho(0)\underline{E}^{(-)}(\underline{r},t).\underline{E}^{(+)}(\underline{r},t')\} \end{aligned}\right\} \quad (3.A.7)$$

Rewriting,

$$\left.\begin{aligned} \sum_{\mu} \int G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r},t')d\underline{r} &= (4\pi)^{\frac{1}{2}} \sum_{\ell,\sigma} \hbar\omega_{\ell} \langle a^{\dagger}_{\ell\sigma} a_{\ell\sigma}\rangle e^{i\omega_{\ell}(t-t')} \\ &= (4\pi)^{\frac{1}{2}} \sum_{\ell,\sigma} \hbar\omega_{\ell} \langle n_{\ell\sigma}\rangle e^{i\omega_{\ell}(t-t')} \end{aligned}\right\} \quad (3.B.8)$$

where $\langle n_{\ell\sigma}\rangle = \langle a^{\dagger}_{\ell\sigma} a_{\ell\sigma}\rangle = \mathrm{Tr}\{\rho(0)a^{\dagger}_{\ell\sigma}(t)a_{\ell\sigma}(t)\}$ is the average number of photons in the ℓ,σ mode and gives a measure of the intensity of its excitation. Thus we see that the Fourier representation of the volume integral of $\sum_{\mu} G^{(1)}_{\mu\mu}$ identifies the energy spectrum $\hbar\omega_{\ell}\langle n_{\ell\sigma}\rangle$ quite generally. But when the fields can be represented by stationary density operators it is simpler to extract the energy spectrum $W(\omega)$ from the correlation function. This we will now do.

According to Glauber's eq. (14.9) ref. [50]

$$G^{(1)}_{\mu\nu}(\underline{r}t,\underline{r}'t') = \langle \underline{E}_{\mu -}(\underline{r},t)\underline{E}_{\nu +}(\underline{r}'t')\rangle$$

$$= \sum_{\ell,\sigma} \sum_{\ell',\sigma'} \{\frac{2\pi\hbar}{V}(\omega_{\ell}\omega_{\ell'})^{\frac{1}{2}} e^{-i(\underline{k}_{\ell}-\underline{k}_{\ell'}).\underline{r}}(\hat{e}_{\ell\sigma\mu}.\hat{e}_{\ell'\sigma'\nu'})\}\langle a^{\dagger}_{\ell\sigma}(t)a_{\ell'\sigma'}(t')\rangle$$

$$= \sum_{\ell,\sigma} \sum_{\ell',\sigma'} \{\ \}\langle a^{\dagger}_{\ell\sigma}(0)a_{\ell'\sigma'}(0)\rangle e^{i\omega_{\ell}t} e^{-i\omega_{\ell'}t'} \quad (3.B.9)$$

$$= \sum_{\ell,\sigma} \sum_{\ell',\sigma'} \{\ \}\langle n_{\ell\sigma}(0)\rangle \delta_{\ell\ell'}\delta_{\sigma\sigma'} e^{i\omega_{\ell}t} e^{-i\omega_{\ell'}t'}$$

I.E. $\langle a^{\dagger}_{\ell\sigma}(0)a_{\ell'\sigma'}(0)\rangle = \mathrm{Tr}\{\rho(t)a^{\dagger}_{\ell\sigma}(0)a_{\ell'\sigma'}(0)\} = \langle n_{\ell\sigma}\rangle \delta_{\ell\ell'}\delta_{\sigma\sigma'}$

since for any stationary field represented by a density operator ρ

there will exist some particular choice of mode functions such that the matrix reduces to diagonal form.

$$\therefore\ G^{(1)}_{\mu\nu}(\underline{r},t;\underline{r}'t') = \frac{2\pi\hbar}{V}\sum_{\ell,\sigma}\omega_\ell\langle n_{\ell\sigma}\rangle(\hat{e}_{\ell\sigma\mu}\cdot\hat{e}_{\ell\sigma\nu})e^{i\omega_\ell(t-t')} \tag{3.B.10}$$

$$\therefore\ \sum_\mu G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r}'t') = (4\pi)^{\frac{1}{2}}\sum_{\ell,\sigma}\frac{\hbar\omega_\ell}{V}\langle n_{\ell\sigma}\rangle e^{i\omega_\ell(t-t')}$$

since $\sum_\mu(\hat{e}_{\ell\sigma\mu}\cdot\hat{e}_{\ell\sigma\mu}) = 1$ (3.B.11)

$= (4\pi)$ x (Glaubers' equation (10.14))

Now, if the field is incoherent, and the volume $V \to \infty$

$$G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r}'t') = (4\pi)^{\frac{1}{2}}\frac{\hbar c}{(2\pi)^3}\sum_{\sigma=1}^{2}\int_{o}^{\infty}dkk^3\int_{o}^{4\pi}d\Omega_{\hat{k}}\langle n_{\ell\sigma}\rangle e^{ick(t-t')}$$

$$= (4\pi)^{\frac{1}{2}}\frac{\hbar c}{(2\pi)^3}\sum_{\sigma=1}^{2}\int_{o}^{\infty}d\underline{k}k\langle n_{\ell\sigma}\rangle e^{ick(t-t')} \tag{3.B.12}$$

where $d\underline{k} = k^2dkd\Omega_{\hat{k}}$

$\therefore$

$$G^{(1)}_{\mu\mu}(\underline{r},t;\underline{r}'t') = (4\pi)^{\frac{1}{2}}\frac{\hbar}{(2\pi c)^3}\sum_{\sigma=1}^{2}\int_{o}^{\infty}d\omega\omega^3\int_{o}^{4\pi}d\Omega_{\hat{k}}\langle n_{\ell\sigma}\rangle e^{i\omega(t-t')}$$

$$= \frac{1}{2\pi}\int_{o}^{\infty}d\omega\frac{\hbar}{(2\pi)}\int_{o}^{4\pi}d\Omega_{\hat{k}}\sum_{\sigma=1}^{2}\frac{\omega^3}{c^3}\langle n_{\ell\sigma}\rangle e^{i\omega(t-t')} \tag{3.B.13}$$

$$\therefore \quad G^{(1)}_{\mu\mu}(\underline{r}t, \underline{r}t') = \frac{1}{2\pi}\int_0^\infty d\omega I(\omega) e^{i\omega(t-t')}$$

$$\text{or} \quad G^{(1)}_{\mu\mu}(\underline{r}t', \underline{r}t) = \frac{1}{2\pi}\int_0^\infty d\omega I(\omega) e^{-i\omega(t-t')} \qquad (3.B.14)$$

$$\text{where} \quad I(\omega) = \frac{\hbar}{2\pi}\int_0^\pi d\Omega_{\hat{k}} \sum_{\sigma=1}^{2} \frac{\omega^3}{c^3} \langle n_{\ell\sigma}\rangle .$$

N.B. In Mollow and Miller's paper[1] eq. (4.3) an identical equation for $G^{(1)}(t',t)$ is given for stationary fields where $G^{(1)}(t',t) \equiv \langle \varepsilon^*(t')\varepsilon(t)\rangle$, but they do not identify the field intensity $I(\omega)$. Also in Mollow's paper[9] eq. (2.14) gives

$$G^{(1)}_{jj}(\underline{r}t', \underline{r}t) \equiv \langle E^{(-)}_j(t')E^{(+)}_j(t)\rangle$$

Mollow goes on to consider the case where the atomic system is in equilibrium with the driving field so that the atomic correlation function (rather the quantum mechanical analogue of a correlation function) is given by $\langle A^\dagger(t')A(t)\rangle = g(t-t')$ for the 2-level atom.

$I(\omega)$ is $W(\omega)$ of Glauber's paper[48] where $\sum_\mu$ is ignored, since $\sum_\mu(\hat{e}_\mu.\hat{e}_\mu)^2 = 1$, and is the energy density per unit frequency interval.

$$\text{N.B.} \quad G^{(1)}_{jj}(\underline{r}0, \underline{r}\tau) = \frac{1}{2\pi}\int_0^\infty I(\omega)e^{-i\omega\tau}d\omega \qquad (3.B.15)$$

According to Mollow[9] the power spectrum of the scattered field is

$$I(\underline{r},\nu) = \int_{-\infty}^{\infty} d\tau e^{i\nu\tau} \sum_j G^{(1)}_{jj}(\underline{r}0, \underline{r}\tau)$$

$$\text{i.e.} \quad I(\underline{r},\nu) = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\tau e^{i(\nu-\omega)\tau}\int_0^\infty d\omega I(\underline{r}\omega) \text{ where } \sum_j \text{ is ignored again} \qquad (3.B.16)$$

so that eq. (2.18) of Mollow's paper[9] is identified.

He also writes

$$I(\underline{r},\nu) = |Q|^2 \int_{-\infty}^{\infty} d\tau e^{i\nu\tau} g(\tau) = |Q|^2 \tilde{g}(\nu) \tag{3.B.17}$$

where he has approximated $\underline{E}^{(+)}(t)$ as in eq. (2.11) and assumed the atomic system to be in equilibrium with the driving field so that $G^{(1)}$ only depends on $(t-t')$. If we let $I(\omega) = 0$ for $\omega < 0$ we can extend the integral over ω from $-\infty$ to $+\infty$ so that

$$I(\underline{r},\nu) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau e^{i(\nu-\omega)} \int_{-\infty}^{\infty} d\omega I(\underline{r},\omega) \tag{3.B.18}$$

Using our units we can identify $|Q|^2 g(\tau)$ by the equation

$$|Q|^2 g(\tau) = \frac{1}{2\pi} \frac{\hbar}{c^3} \left[\frac{1}{2\pi} \int_{o}^{\infty} d\omega\omega^3 e^{-i\omega\tau} \right] \sum_{\sigma=1}^{2} \int_{o}^{4\pi} \langle n_{\ell\sigma} \rangle d\Omega_{\hat{k}} \tag{3.B.19}$$

N.B. There is a factor of $1/2\pi$ difference between our notation and that of Mollow.

Now

$$\sum_{j} G_{jj}^{(1)}(\underline{r}0,\underline{r}\tau) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega I(\omega) e^{-i\omega\tau} \tag{3.B.20}$$

$$\left.\begin{aligned} \therefore \quad \int_{-\infty}^{\infty} \sum_{j} G_{jj}^{(1)}(\underline{r}0,\underline{r}\tau) e^{i\nu\tau} d\tau &= \frac{1}{2\pi} \int_{-\infty}^{\infty} d\tau e^{i(\nu-\omega)\tau} \int_{-\infty}^{\infty} d\omega I(\underline{r},\omega) \\ &= \int_{-\infty}^{\infty} d\omega W(\omega) \delta(\nu-\omega) \\ &= W(\nu) \end{aligned}\right\} \tag{3.B.21}$$

So
$$I(\underline{r}\omega) = \int_{-\infty}^{\infty} \sum_{\mu} G_{\mu\mu}^{(1)}(\underline{r}0,\underline{r}\tau) e^{i\omega\tau} d\tau \tag{3.B.22}$$

is the <u>energy spectrum for the quanta present</u> and is the Fourier transform of the 1st. order time-dependent correlation function.

Also

$$I(\underline{r},\nu) = \frac{\hbar}{2\pi}\frac{\nu^3}{C^3}\sum_{=1}^{2}\int_{o}^{4\pi}\langle n_{\ell\sigma}\rangle d\Omega_{\hat{k}} \tag{3.B.23}$$

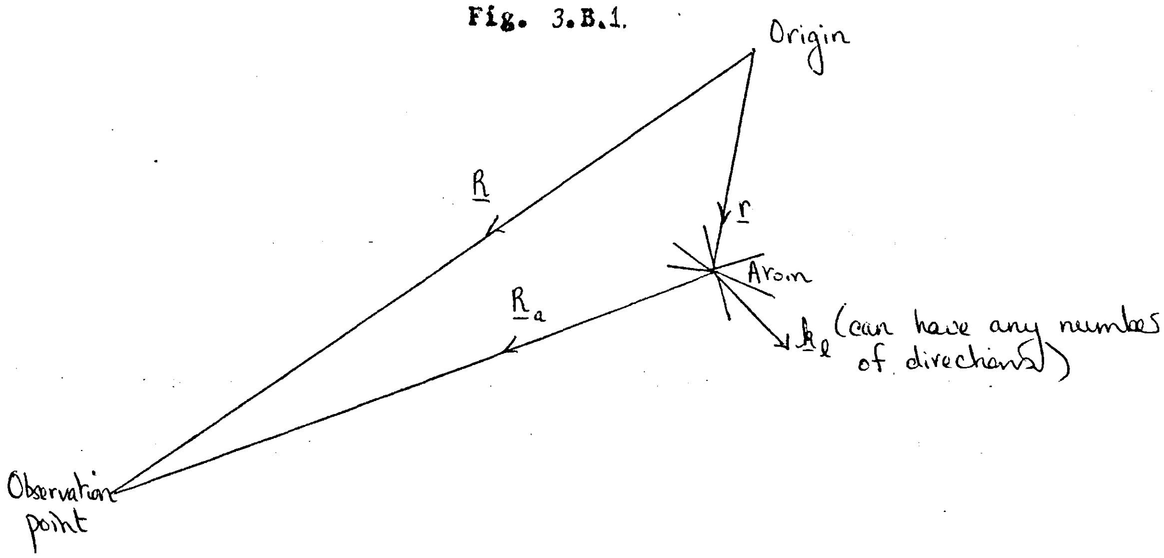
is the energy of power spectrum of the scattered field.

Now we wish to find the spectral properties of the FAR field. So following the reasoning of Lehmberg [2] we let $\underline{R}$ be an observation point (referred to as arbitrary origin within the ensemble) and assume that $|\underline{R}-\underline{r}| \equiv R_a$ satisfies

$$\left.\begin{aligned} &\frac{\omega_{j'}}{C} R_a \gg 1 \\ &\frac{\omega_{j''}}{C} R_a \gg 1 \\ \text{i.e.}\quad & R_a \gg k_{j'} \\ & R_a \gg k_{j''} \end{aligned}\right\} \tag{3.B.24}$$

(R_a is very large compared with the distance traversed by photons of energy $\hbar\omega_{j'}$ and $\hbar\omega_{j''}$ in a second.)

Fig. 3.B.1.



The quantities of interest can be obtained from the correlation functions

$$\left.\begin{aligned} f_{\hat{R}_j}(t,t') &= \frac{R^2c}{2\pi\omega_j\hbar} \langle \underline{E}^{(+)\dagger}(\underline{R},t).\underline{E}^{(+)}(\underline{R},t')\rangle \\ &= \frac{R^2c}{2\pi\omega_j\hbar} \sum_j G^{(1)}_{jj}(\underline{R},t;\underline{R},t') \\ \text{and} \quad f(t,t') &= \int_0^{4\pi} d\Omega_{\hat{R}}\, f_{\hat{R}_j}(t,t') \end{aligned}\right\} \qquad (3.B.25)$$

(where $\omega_j = \omega_{j'}$ or $\omega_{j''}$ depending in which transitions we are interested in),

where the average is taken over initial states of the atomic system, and $\underline{E}^{(+)}(\underline{R},t)$ is given by

$$\underline{E}^{(+)}(\underline{R},t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma} a_{\ell\sigma}(t) e^{i\underline{k}_\ell\cdot\underline{R}} \qquad (3.B.26)$$

as in Lehmberg's appendix.[2]

If we substitute our expression for $a_{\ell\sigma}(t)$ for $r \neq 0$ into $\underline{E}^{\dagger}(\underline{R},t)$ evaluated at $t = t_R = t + \frac{R}{C}$ we obtain

$$\left.\begin{aligned} \underline{E}^{(+)}(\underline{R},t_R) &= \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma} a_{\ell\sigma}(0) e^{-i\omega_\ell t_R}\, e^{i\underline{k}_\ell\cdot(\underline{R}-\underline{r})} \\ &+ i\sum_\ell \sqrt{\frac{2\pi\omega_\ell}{V}} \sum_\sigma \left[\hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot\underline{p}_{j'})\right] \int_0^{t_R} dt'\, e^{-i\omega_\ell(t_R-t')} P_{ij'}(t') e^{i\underline{k}\cdot(\underline{R}-\underline{r})} \\ &+ i\sum_\ell \sqrt{\frac{2\pi\omega_\ell}{V}} \sum_\sigma \left[\hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot\underline{p}_{j''})\right] \int_0^{t_R} dt''\, e^{-i\omega_\ell(t_R-t'')} P_{ij''}(t'') e^{i\underline{k}_\ell\cdot(\underline{R}-\underline{r})} \end{aligned}\right\} \qquad (3.B.27)$$

If we now assume the time for a light signal to cross the ensemble $\ll \Delta t$, the time for appreciable (secular) changes to occur in the atomic levels

$$\left.\begin{aligned} &\text{then} \quad P_{ij'}(t') = P_{ij'}(t_R)e^{-i\omega_{j'}(t'-t_R)} \\ &\text{and} \quad P_{ij''}(t'') = P_{ij''}(t_R)e^{-i\omega_{j''}(t''-t_R)} \end{aligned}\right\} \qquad (3.B.28)$$

We now also need to know $\underline{\sum_{\sigma=1}^{2} \hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot p_j)}$ where $j = j'$ or j''.

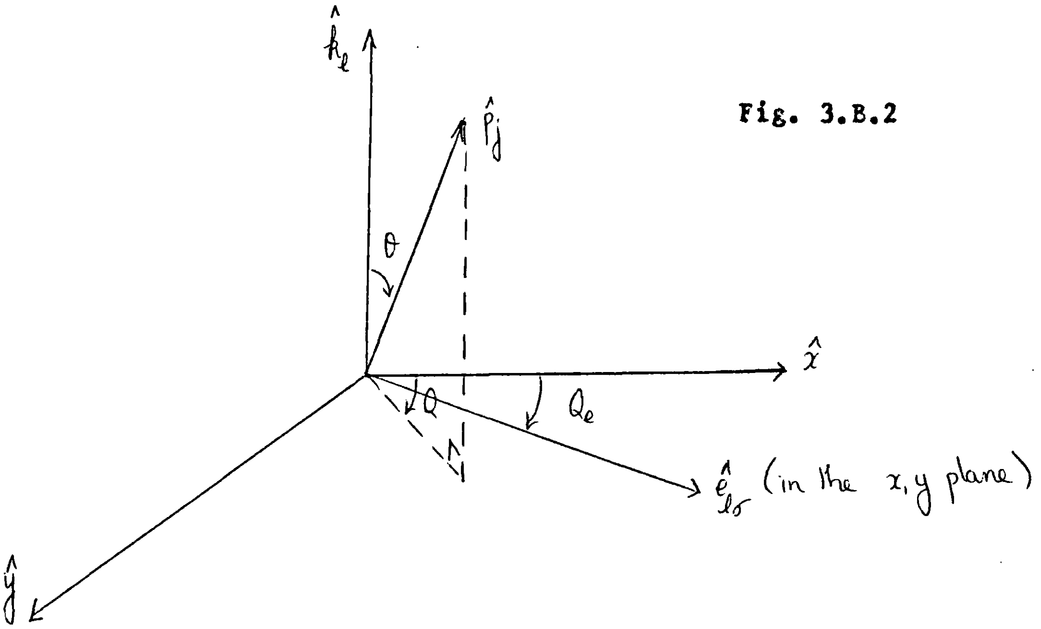


Fig. 3.B.2

Using the above axes as before we can write

$$\left.\begin{aligned} \hat{e}_{\ell\sigma} &= \hat{x}\cos Q_e + \hat{y}\sin Q_e \\ \hat{p}_j &= \hat{x}\sin\theta\cos Q_p + \hat{y}\sin\theta\sin Q_p + \hat{k}\cos\theta \end{aligned}\right\} \qquad (3.B.29)$$

$$\therefore \quad \hat{e}_{\ell\sigma}\cdot\hat{p}_j = \sin\theta.\cos(Q_e - Q_p) \qquad (3.B.30)$$

$$\hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot\hat{p}_j) = \sin\theta.\cos(Q_e - Q_p)\left[\cos Q_e\,\hat{x} + \sin Q_e\,\hat{y}\right] \qquad (3.B.31)$$

Considering the adjoint figure, as before we can write

$$\hat{p}_j = \hat{e}_{\ell_1}\sin\theta\cos\alpha + \hat{e}_{\ell_2}\sin\theta\sin\alpha + \hat{k}_\ell\cos\theta \qquad (3.B.32)$$

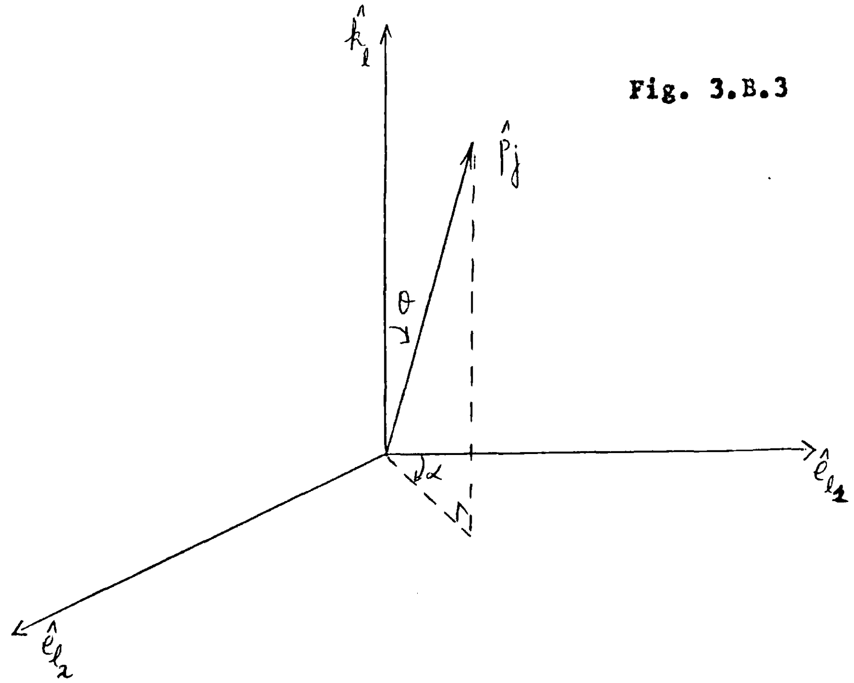


Fig. 3.B.3

$$\left.\begin{aligned}
\sum_{\sigma=1}^{2} \hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot\hat{p}_j) &= \hat{e}_{\ell_1}(\hat{e}_{\ell_1}\cdot\hat{p}_j) + \hat{e}_{\ell_2}(\hat{e}_{\ell_2}\cdot\hat{p}_j) \\
&= \hat{e}_{\ell_1}\Big[\sin\theta\cos\alpha\Big] + \hat{e}_{\ell_2}\Big[\sin\theta\sin\alpha\Big] \\
&= \hat{p}_j - \hat{k}_\ell\cos\theta \\
\therefore \quad \sum_{\sigma=1}^{2} \hat{e}_{\ell\sigma}(\hat{e}_{\ell\sigma}\cdot\hat{p}_j) &= \hat{p}_j - \hat{k}_\ell(\hat{k}_\ell\cdot\hat{p}_j)
\end{aligned}\right\} \quad (3.B.33)$$

Hence

$$\left.\begin{aligned}
\underline{E}^{(+)}(\underline{R},t_R) &= \underline{E}^{(+)(0)}(\underline{R},t_R) + \frac{i2\pi}{V}\sum_{\ell}\omega_\ell\Big[\underline{p}_{j'} - \hat{k}_\ell(\hat{k}_\ell\cdot\hat{\underline{p}}_{j'})\Big]\int_0^{t_R} dt' \\
&\quad \times e^{+i(\omega_\ell-\omega_{j'})(t'-t_R)}P_{ij'}(t)e^{i\underline{k}_\ell\cdot\underline{R}} \\
&\quad + \frac{i2\pi}{V}\sum_{\ell}\omega_\ell\Big[\underline{p}_{j''} - \hat{k}_\ell(\hat{k}_\ell\cdot\underline{p}_{j''})\Big]\int_0^{t_R} dt'' \\
&\quad \times e^{i(\omega_\ell-\omega_{j''})(t''-t_R)}P_{ij''}(t)e^{i\underline{k}_\ell\cdot\underline{R}} \\
\text{where} \qquad & \\
\underline{E}^{(+)(0)}(\underline{R},t_R) &= \sum_{\ell,\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\,\hat{e}_{\ell\sigma}\hat{a}_{\ell\sigma}(0)e^{-i\omega_\ell t_R}e^{i\underline{k}_\ell\cdot\underline{R}}
\end{aligned}\right\} \quad (3.B.34)$$

Considering $V \to \infty$, we obtain

$$\left.\begin{aligned}
&\underline{E}^{(+)}(\underline{R},t_R) = \underline{E}^{(+)(0)}(\underline{R},t_R) \\
&+ \frac{i2\pi}{(2\pi C)^3} \int_0^\infty d\omega\omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \left[\underline{p}_{j'} - \hat{k}(\hat{k}.\underline{p}_{j'})\right] \int_0^{t_R} dt' e^{i(\omega-\omega_{j'})(t'-t_R)} \\
&\times P_{ij'}(t) e^{i(\omega/c)\hat{k}.(\underline{R}-\underline{r})} \\
&+ \frac{i2\pi}{(2\pi c)^3} \int_0^\infty d\omega\omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \left[\underline{p}_{j''} - \hat{k}(\hat{k}.\underline{p}_{j''})\right] \int_0^{t_R} dt'' e^{i(\omega-\omega_{j''})(t''-t_R)} P_{ij''}(t) \\
&\times e^{i\omega/c\ \hat{k}.(\underline{R}-\underline{r})} \qquad \text{where } \underline{k} = k\ \hat{k} = \frac{\omega}{c}\hat{k}
\end{aligned}\right\} \quad (3.B.35)$$

$$\left.\begin{aligned}
\text{I.E.} \quad &\underline{E}^{\dagger}(\underline{R},t_R) = \underline{E}^{(+)(0)}(\underline{R},t_R) \\
&+ \frac{i}{4\pi^2 c^3} \sum_{j=j'j''} \int_0^\infty d\omega\omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \left[\underline{p}_j - \hat{k}(\hat{k}.\underline{p}_j)\right] \int_0^{t_R} dt' e^{i(\omega-\omega_j)(t'-t_R)} \\
&\qquad \times P_{ij}(t) e^{i\omega/c\ \hat{k}.(\underline{R}-\underline{r})}
\end{aligned}\right\} \quad (3.B.36)$$

Thus we see that the field has several properties:

(i) The largest contributions to the field come from $\omega \simeq \omega_j$, which is physically reasonable.

(ii) Since $\frac{\omega}{c} R \sim \frac{\omega_j}{c} R$ and $\frac{\omega_j}{c} R \gg 1$ $\quad e^{i\omega/c\ \hat{k}.\underline{R}}$ $\quad e^{i\omega/c\ \hat{k}.\underline{R}}$ will oscillate rapidly over a range of $\hat{R}_a.\hat{k}$ in which $\underline{p}_j - \hat{k}(\hat{k}.\underline{p}_j)$ remains essentially constant. As $\cos^{-1}(\hat{k}.\hat{R})$ increases $e^{i\omega/c\ R\hat{k}.\hat{R}} \to 1$ and for $\hat{k}.\hat{R} = \cos 90^\circ = 0$ this is an identity.

(iii) Whatever the direction of $\hat{k}$ the relative change in $\left[\underline{p}_j - \hat{k}(\hat{k}.\underline{p}_j)\right]$ is very small indeed and so has little effect. Consider the maximum variation $\hat{k} = \pm \hat{R}_a$, since the important contribution comes from these directions around $\hat{k} = \pm \hat{R}_a$ where the phase $\pm \frac{\omega}{c} R_a$ is stationary. Substituting this in the slowly varying function

$$\left[\underline{p}_j - \hat{k}(\hat{k}.\underline{p}_j)\right]$$

we see that for both directions of $\hat{k}$

$$\left[\underline{p}_j - \hat{k}(\hat{k}.\underline{p}_j)\right] \to \left[\underline{p}_j - \hat{R}_a(\hat{R}_a.\underline{p}_j)\right]$$

and this is almost constant, contributing mainly around $\hat{k} = \pm \hat{R}$, and so can be taken out of the integral over $\Omega_{\hat{k}}$ leaving only $e^{i\omega/c\ \hat{k}.\underline{R}}$ to be evaluated over $\Omega_{\hat{k}}$

$$\left.\begin{aligned} \int d\Omega_{\hat{k}} e^{i\omega/c\ \hat{k}.\underline{R}_a} &= \int_0^{2\pi} dQ' \int_0^{\pi} dQ' \sin\theta' e^{i\omega/c\ R_a \cos\theta'} \qquad \text{where } \cos\theta' = \hat{k}.\hat{R} \\ &= \frac{-i2\pi c}{\omega R}\left(e^{i\omega R_a/c} - e^{-i\omega R_a/c}\right) \end{aligned}\right\} \quad (3.B.37)$$

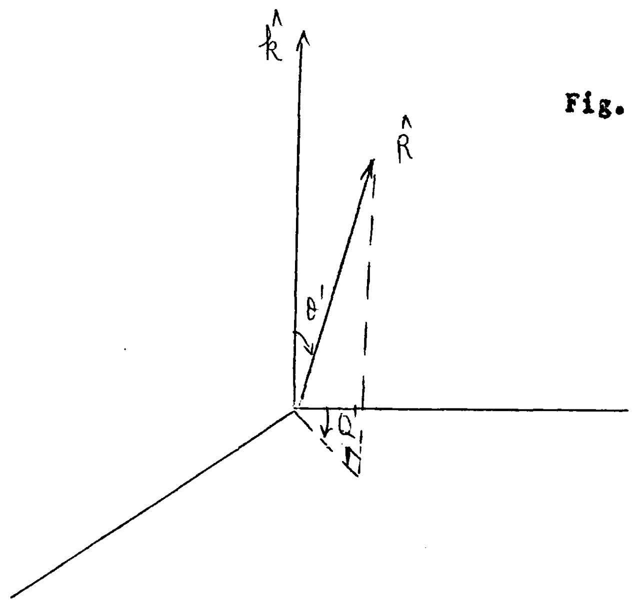


Fig. 3.B.4

Thus, substituting $t_R = t + \frac{R}{C}$ also, we obtain

$$\left.\begin{aligned}
&\underline{E}^{(+)}(\underline{R},t_R) = \underline{E}^{(+)}(\underline{R},t_R) \\
&+ \left[\frac{1}{2\pi C^2}\right] \sum_{j=j'j''} \frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a} \int_0^\infty d\omega\omega^2(e^{i\omega/c(R_a-R)}-e^{-i\omega/c(R_a+R)}) \\
&\qquad \int_0^{t_R} dt' e^{i(\omega-\omega_j)(t'-t)} P_{ij} \times e^{+i\omega_j R/C}
\end{aligned}\right\} \quad (3.B.38)$$

There are two cases of interest

(a) when the initial radiation is absent entirely, or

(b) confined to a narrow beam.

In case (b), if $\underline{R}$ lies outside any such beam, and if $R \gg r$ (so that $R_a - R \triangleq -\underline{R}.\underline{r}$) then the term $e^{i\omega(R_a-R)/C} \triangleq e^{-i\omega r/c}$ and is a moderate term but $e^{-i\omega(R_a+R)/c} \triangleq e^{-i\omega 2R/c}$ is an extremely rapidly oscillating, since R is very large compared with r, and so is neglected.

Hence

$$\left.\begin{aligned}
\underline{E}^{(+)}(\underline{R},t_R) &= \underline{E}^{(+)(0)}(\underline{R},t_R) + \frac{1}{2\pi c^2} \sum_{j=j'j''} \frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a} \int_0^\infty d\omega\omega^2 e^{i\omega(R_a-R)/c} \\
&\quad \times \int_0^{t_R} dt' e^{i(\omega-\omega_j)(t'-t)} e^{i\omega_j R/c} P_{ij}(t)
\end{aligned}\right\} \quad (3.B.39)$$

[N.B. If $\underline{r} = 0$, i.e. the atom is at the origin, then $R = R_a$.]

Integrating over t' we obtain, on using $R_a - R \triangleq r$,

$$\left.\begin{aligned}
&\underline{E}^{(+)}(\underline{R},t_R) = \underline{E}^{(+)(0)}(\underline{R},t_R) \\
&+ \frac{1}{2\pi c^2} \sum_{j=j'j''} \frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a} \int_0^\infty d\omega\omega^2 e^{-i\omega_j r/c} P_{ij}(t) \\
&\times \left[\frac{e^{i(\omega-\omega_j)(R-r)/c} - e^{-i(\omega-\omega_j)(R+r/c)}}{i(\omega-\omega_j)}\right]
\end{aligned}\right\} \quad (3.B.80)$$

where the 2nd term in [ ] is a H.F. term and can be neglected and we can approximate [ ] by $2\pi\delta(\omega-\omega_j)$. Hence

$$\left.\begin{aligned}
&\underline{E}^{(+)}(\underline{R},t_R) = \underline{E}^{(+)(0)}(\underline{R},t_R)\\
&\quad + \sum_{j=j'j''} \frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a}\frac{\omega_j^2}{c^2}P_{ij}(t)e^{-i\omega_j/c\,r} \quad \text{where } r = R-R_a,
\end{aligned}\right\}(3.B.41)$$

which is identical in form to eq. (36) of Lehmberg[2] when $\hbar = 1$. We have already assumed $\omega_j R_{a/c} \gg 1$ and since $|R_a-R| \leq r$ max conditions $\omega_j t \gg 1$ and $ct \gg r$ max lead to $\omega_j\left[t-(R-R_a)/c\right] \gg 1$.

[N.B. H.F. terms in the Hamiltonian result in expressions of the form $\int_0^\infty d\omega f(\omega)\delta(\omega+\omega_j)$ which are identically zero since $\omega = -\omega_j$ does not lie within the range of the integral over ω and so has no effect.]

Substituting in the expression for $f_{\hat{R}_j}(t,t')$ eq. (3.B.25) for $j = j'$ (i.e. transitions between j' and i) we obtain 9 terms, as follows:

$$f_{R_{j'}}(t_R,t_{R'}) = \frac{R^2c}{2\pi\omega_{j'}\hbar}\langle\underline{E}^{(+)\dagger}(\underline{R},t_R)\cdot\underline{E}^{(+)}(\underline{R},t_{R'})\rangle \tag{3.B.42}$$

(which is proportional to $\langle a_{\ell\sigma}(t_R)a_{\ell\sigma}(t_{R'})\rangle$, as we see from eq. (3.B.26).)

$$\left.\begin{aligned}
&= \frac{R^2c}{2\pi\omega_{j'}\hbar}\Bigg[\langle\underline{E}^{(+)(0)\dagger}(\underline{R},t_R)\cdot\underline{E}^{(+)(0)}(\underline{R},t_{R'})\rangle\\
&\quad + \sum_{j=j'j''}\frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]^2}{R_a^2}\left(\frac{\omega_j}{c}\right)^4\langle P_{ij}^\dagger(t_R)P_{ij}(t_{R'})\rangle\\
&+ \frac{\left[\underline{p}_{j'}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_{j'})\right]\left[\underline{p}_{j''}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a^2}\frac{\omega_{j'}^2\omega_{j''}^2}{c^4}\{e^{i(\omega_{j'}-\omega_{j''})r/c}\\
&\times\langle P_{ij'}^\dagger(t_R)P_{ij''}(t_{R'})\rangle + e^{-i(\omega_{j'}-\omega_{j''})R/c}\langle P_{ij''}^\dagger(t_R)P_{ij'}(t_{R'})\rangle\}\\
&+ \sum_{j=j'j''}\frac{\left[\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)\right]}{R_a}\frac{\omega_j^2}{c^2}\{e^{-i\omega_j r/c}\langle\underline{E}^{(+)(0)\dagger}(\underline{R},t_R)P_{ij}(t_{R'})\rangle\\
&+ e^{i\omega_j r/c}\langle P_{ij}^\dagger(t_R)\underline{E}^{(+)(0)}(\underline{R},t_{R''})\rangle\}\Bigg]
\end{aligned}\right\}(3.B.43)$$

Now when R is sufficiently large the interference between the incident and scattered radiation is negligible, hence the last two terms in $\sum_j$ can be ignored, so that

$$f_{\hat{R}_{j'}}(t_R,t_{R'}) = f^{(0)}_{\hat{R}_{j'}}(t_R,t_{R'}) + L_{\hat{R}_{j'}}(t_R,t_{R'}) \tag{3.B.44}$$

which can be shown to be the same as a factor of $(R^2c/2\pi\hbar\omega_{j'})$ times the following expression:

$$\begin{aligned}\sum_\mu G^{(1)}_{\mu\mu}(\underline{R},t_R;\underline{R},t_{R'}) &= \sum_\mu G^{(1)(0)}_{\mu\mu}(\underline{R},t_R;\underline{R},t_{R'}) \\ &+ \sum_{j=j',j''} Q^2_j(\hat{R})\langle P^\dagger_{ij}(t_R)P_{ij}(t_{R'})\rangle + \underline{Q}_{j'}\cdot\underline{Q}_{j''}\langle P^\dagger_{ij'}(t_R)P_{ij''}(t_{R'})\rangle \\ &+ \underline{Q}_{j''}\cdot\underline{Q}_{j'}\langle P^\dagger_{ij''}(t_R)P_{ij'}(t_{R'})\rangle\end{aligned} \tag{3.B.45}$$

where $$f^{(0)}_{\hat{R}_{j'}}(t_R,t_{R'}) = \frac{R^2c}{2\pi\omega_{j'}\hbar}\langle\underline{E}^{(+)(0)\dagger}(\underline{R},t_R)\cdot\underline{E}^{(+)(0)}(\underline{R},t_{R'})\rangle \tag{3.B.46}$$

and
$$\begin{aligned}L_{\hat{R}_{j'}}(t_R,t_{R'}) = \frac{R^2c}{2\pi\omega_{j'}\hbar}\Bigg|&\sum_{j=j',j''}\frac{|\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)|^2}{R_a^2}\left(\frac{\omega_j}{c}\right)^4 \\ &\times\langle P^\dagger_{ij}(t_R)P_{ij}(t_{R'})\rangle \\ &+\frac{|\underline{p}_{j'}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)||\underline{p}_{j''}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_{j''})|}{R_a^2}\frac{\omega_{j'}{}^2\omega_{j''}{}^2}{c^4}\{e^{i(\omega_{j'}-\omega_{j''})r/c} \\ &\times\langle P^\dagger_{ij'}(t_R)P_{ij''}(t_R)\rangle + e^{-i(\omega_{j'}-\omega_{j''})r/c}\langle P^\dagger_{ij''}(t_R)P_{ij}(t_{R'})\rangle\}\Bigg|\end{aligned} \tag{3.B.47}$$

Now
$$\begin{aligned}|\underline{p}_j-\hat{R}_a(\hat{R}_a\cdot\underline{p}_j)|^2 &= p_j{}^2(\hat{R}_a\cdot\underline{p}_j)^2 - 2(\underline{p}_j\cdot\hat{R}_a)^2 \\ &= p_j{}^2|1-(\hat{R}_a\cdot\hat{p}_j)^2|\end{aligned} \tag{3.B.48}$$

and
$$|\underline{p}_{j'}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_{j'})||\underline{p}_{j''}-\hat{R}_a(\hat{R}_a\cdot\underline{p}_{j''})| = p_{j'}p_{j''}|\hat{p}_{j'}\cdot\hat{p}_{j''}-(\hat{R}_a\cdot\hat{p}_{j'})(\hat{R}_a\cdot\hat{p}_{j''})|$$

so that

$$L_{\hat{R}_{j'}}(t_R,t_{R'}) = \frac{R^2 c}{2\pi\omega_{j'}\hbar}\left[\sum_{j=j',j''} p_j^2 \frac{\left[1-(\hat{R}_a\cdot\hat{p}_j)^2\right]}{R_a^2}\left(\frac{\omega_j}{c}\right)^4 \langle P_{ij}^\dagger(t_R)P_{ij}(t_{R'})\rangle\right.$$

$$+\, p_{j'}p_{j''}\left[\hat{p}_{j'}\cdot\hat{p}_{j''}-(\hat{R}_a\cdot\hat{p}_{j'})(\hat{R}_a\cdot\hat{p}_{j''})\right]\frac{\omega_{j'}^2\omega_{j''}^2}{c^4}\{e^{i(\omega_{j'}-\omega_{j''})r/c}$$

$$\left.\langle P_{ij'}^\dagger(t_R)P_{ij''}(t_{R'})\rangle + e^{-i(\omega_{j'}-\omega_{j''})r/c}\langle P_{ij''}^\dagger(t_R)P_{ij'}(t_{R'})\rangle\}\right]$$

$$= \frac{R^2}{2\pi\hbar}\left[p_{j'}^2\frac{\left[1-(\hat{R}_a\cdot\hat{p}_{j'})^2\right]}{R_a^2}\frac{\omega_{j'}^3}{c^3}\langle P_{ij'}^\dagger(t_R)P_{ij'}(t_{R'})\rangle\right.$$

$$+\, p_{j''}^2\frac{\left[1-(\hat{R}_a\cdot\hat{p}_{j''})^2\right]\omega_{j''}^3}{R_a^2\, c^3}\langle P_{ij''}^\dagger(t_R)P_{ij''}(t_{R'})\rangle\frac{\omega_{j''}}{\omega_{j'}}$$

$$+\, p_{j'}p_{j''}\frac{\left[(\hat{p}_{j'}\cdot\hat{p}_{j''})-(\hat{R}_a\cdot\hat{p}_{j'})(\hat{R}_a\cdot\hat{p}_{j''})\right]}{R_a^2}\frac{\omega_{j''}^3}{c^3}\cdot\frac{\omega_{j'}}{\omega_{j''}}\{e^{i(\omega_{j'}-\omega_{j''})r/c}$$

$$\left.\langle P_{ij'}^\dagger(t_R)P_{ij''}(t_{R'})\rangle + e^{-i(\omega_{j'}-\omega_{j''})r/c}\langle P_{ij''}^\dagger(t_R)P_{ij'}(t_{R'})\rangle\}\right] \tag{3.B.49}$$

$$= \frac{3}{8\pi}\gamma_{j'}\frac{R^2}{R_a^2}\left[1-(\hat{R}_a\cdot\hat{p}_{j'})^2\right]\langle P_{ij'}^\dagger(t_R)P_{ij'}(t_{R'})\rangle$$

$$+\,\frac{3}{8\pi}\gamma_{j''}\frac{R^2}{R_a^2}\left[1-(\hat{R}_a\cdot\hat{p}_{j''})^2\right]\langle P_{ij''}^\dagger(t_R)P_{ij''}(t_{R'})\rangle\frac{\omega_{j''}}{\omega_{j'}}$$

$$+\,\frac{1}{2\pi\hbar}p_{j'}p_{j''}\left[\hat{p}_{j'}\cdot\hat{p}_{j''}-(\hat{R}_a\cdot\hat{p}_{j'})(\hat{R}_a\cdot\hat{p}_{j''})\right]\frac{R^2}{R_a^2}\frac{\omega_{j''}^3}{c^3}\frac{\omega_{j'}}{\omega_{j''}}$$

$$\{e^{i(\omega_{j'}-\omega_{j''})r/c}\langle P_{ij'}^\dagger(t_R)P_{ij''}(t_{R'})\rangle + e^{-i(\omega_{j'}-\omega_{j''})r/c}$$

$$\langle P_{ij''}^\dagger(t_R)P_{ij'}(t_{R'})\rangle\}$$

where $\gamma_{j'}$, $\gamma_{j''}$, are defined by eq. (3.A.246).

Writing,

$$W^{(1)}_{\hat{R}_j} = \frac{3\gamma_j}{8\pi}\left[1-(\hat{R}_a\cdot\hat{p}_{j'})^2\right] \tag{3.B.50}$$

we obtain

$$\left.\begin{aligned} L_{\hat{R}}(t_R,t'_R) &= W^{(1)}_{\hat{R}_{j'}}\frac{R^2}{R_a^2}\langle P^\dagger_{ij'}(t_R)P_{ij'}(t'_R)\rangle \\ &+ W^{(1)}_{R_{j''}}\frac{R^2}{R_a^2}\langle P^\dagger_{ij''}(t_R)P_{ij'}(t'_R)\rangle\frac{\omega_{j''}}{\omega_{j'}} + W^{(2)} \end{aligned}\right\} \tag{3.B.51}$$

where

$$\left.\begin{aligned} W^{(2)} &= \frac{1}{2\pi\hbar}\, p_{j'}p_{j''}\left[\hat{p}_{j'}\cdot\hat{p}_{j''}-(\hat{R}_a\cdot\hat{p}_{j'})(\hat{R}_a\cdot\hat{p}_{j''})\right]\frac{R^2}{R_a^2}\frac{\omega_{j''}{}^3}{c^3}\frac{\omega_{j'}}{\omega_{j''}} \\ &\times \{e^{i(\omega_{j'}-\omega_{j''})r/c}\langle P^\dagger_{ij'}(t_R)P_{ij''}(t'_R)\rangle + e^{-i(\omega_{j'}-\omega_{j''})r/c} \\ &\qquad\times\langle P_{ij''}(t_R)P_{ij'}(t'_R)\rangle\} \end{aligned}\right\} \tag{3.B.52}$$

and, if the atom is at the origin, $R = R_a$ and

$$\left.\begin{aligned} f_{\hat{R}_{j'}}(t_R,t'_R) &= \frac{R^2 c}{2\ {}_{j'}\hbar}\ \underline{E}^{(+)(0)}(\underline{R},t_R)\cdot\underline{E}^{(+)(0)}(\underline{R},t'_R) \\ &+ \left[W^{(1)}_{\hat{R}_{j'}}\langle P^\dagger_{ij'}(t_R)P_{ij'}(t'_R)\rangle + W^{(1)}_{\hat{R}_{j''}}\langle P^\dagger_{ij''}(t_R)P_{ij'}(t'_R)\rangle\frac{\omega_{j''}}{\omega_{j'}}\right] + W^{(2)} \end{aligned}\right\} \tag{3.B.53}$$

Now

$$\left.\begin{aligned} f_{j'}(t_R,t'_R) &= \oint d\Omega_{\hat{R}} f_{\hat{R}_{j'}}(t_R,t'_R) = \oint d\Omega_{\hat{R}} f^{(0)}_{\hat{R}_{j'}}(t_R,t'_R) \\ &+ \oint d\Omega_{\hat{R}} L_{\hat{R}_{j'}}(t_R,t'_R) = f^{(0)}_{j'}(t_R,t'_R) + L_{j'}(t_R,t'_R) \end{aligned}\right\} \tag{3.B.54}$$

where

$$f^{(0)}_{j'}(t_R,t'_R) = \frac{R^2c}{(2\pi c)^3\omega_{j'}} \sum_{\sigma=1}^{2} \oint d\Omega_{\hat{R}} \oint d\Omega_{\hat{R}} \int_0^\infty d\omega\omega^3 e^{i\omega(t_R-t'_R)} \times \langle a^\dagger_{\ell\sigma}(0)a_{\ell\sigma}(0)\rangle$$

and

$$L_{j'}(t_R,t'_R) = \gamma_{j'} \langle P^\dagger_{ij'}(t_R)P_{ij'}(t'_R)\rangle + \gamma_{j''} \frac{\omega_{j''}}{\omega_{j'}} \langle P^\dagger_{ij''}(t_R)P_{ij''}(t'_R)\rangle + \oint d\Omega_{\hat{R}}\, W^{(2)} \qquad (3.B.54)$$

since $\oint d\Omega_{\hat{R}}|1-(\hat{R}.\hat{p}_j)^2| = \frac{8\pi}{3}$ for $j = j',j''$ (see Chapter VII for detailed explanation)

N.B. $\langle a^\dagger_{\ell\sigma}(0)a_{\ell\sigma}(0)\rangle = n_{\ell\sigma}(0)$ = no. of photons in mode $\ell\sigma$ at $t = 0$ so that if initially there is no radiation $n_{\ell\sigma}(0) = 0$ and so this term is irrelevant. We can see that it has nothing to do with the radiation damping since it exists even when g's are zero. $f^{(0)}_{j'}(t_R,t'_R)$ is simply the incident beam's characteristic and shows whether it is coherent, incoherent, chaotic, etc., i.e. it depends on the photon distribution of the incident beam. So that, for no radiation present initially, the energy spectrum of the scattered field for transitions from level j is:

$$L_{j'}(t_R,t'_R) = \gamma_{j'} \langle P^\dagger_{ij'}(t_R)P_{ij'}(t'_R)\rangle + \gamma_{j''} \frac{\omega_{j''}}{\omega_{j'}} \langle P^\dagger_{ij''}(t_R)P_{ij''}(t'_R)\rangle \qquad (3.B.55)$$
$$+\oint d\Omega_{\hat{R}}\, W^{(2)}$$

and this term arises from the interaction.

N.B. $\oint d\Omega_{\hat{R}} W^{(2)} = \frac{\omega_{j'}\gamma}{\omega_{j''}}\{\langle P^\dagger_{ij'}(t_R)P_{ij''}(t'_R)\rangle + \langle P^\dagger_{ij''}(t_R)P_{ij'}(t'_R)\rangle$ as shown in Chapter VII eq. (7.B.74) except that the factor outside the brackets is different since in the latter case the $j'' \rightarrow i$ transition is considered and not the $j' \rightarrow i$ transition as here.

In Chapters V, VI and VII we shall be evaluating 2-time atomic correlation functions using the Markoff approximation as in Mollow's paper[9]. Here we shall avoid this by instead solving for states such $P_{ij'}(t'_R)|rad.\rangle$, $P_{ij''}(t_{R'})|rad.\rangle$ as suggested by Lehmberg[2] p. 887 at the end of Section III where he gives this method as an alternative to approximating terms $\langle P^\dagger_{ij}(t_R)P_{ij}(t_{R'})\rangle$ etc. by using the fluctuation-regression theorem.[51] This we do in order to find $\langle a^\dagger_{\ell\sigma}(t_R)a_{\ell\sigma}(t_{R'})\rangle$, which we see from eq. (3.B.32) is proportional to $f_{\hat{R}_{j'}}(t_R,t_{R'})$, for retarded times t_R and $t_{R'} \gg \gamma^{-1}$. We can compare its value from eq. (3.A.8) with eq. (3.B.43).

Since $|rad.\rangle = |0\rangle$ initially we shall now derive equations for $P_{ij'}(t)|0\rangle$ and $P_{ij''}(t)|0\rangle$ from (3.A.13) and (3.A.14) by multiplying those eqs. on the right of the vacuum state $|0\rangle$ for all $\ell\sigma$ photons. Then, since

$$\underline{E}_+^{(0)}(t)|0\rangle \sim a_{\ell\sigma}(0)|0\rangle = 0,$$

and there are no terms involving $\underline{E}_+^{(0)\dagger}(t)$,

we obtain two coupled linear differential equations:

$$\dot{P}_{ij''}|0\rangle = -(\tfrac{1}{2}\gamma_{j''}+i\omega_{j''_o})P_{ij''}|0\rangle - \tfrac{1}{2}\Gamma_{j'_o}P_{ij'}|0\rangle \tag{3.B.56}$$

$$\dot{P}_{ij'}|0\rangle = -(\tfrac{1}{2}\gamma_{j'}+i\omega_{j'_o})P_{ij'}|0\rangle - \tfrac{1}{2}\Gamma_{j''_o}P_{ij''}|0\rangle \tag{3.B.57}$$

Let $\quad A = P_{ij''}(t)|0\rangle \quad$ and $\quad B = P_{ij'}(t)|0\rangle \qquad$ (3.B.58)

then

$$\dot{A} = -(\tfrac{1}{2}\gamma_{j''}+i\omega_{j''_o})A - \tfrac{1}{2}\Gamma_{j'_o}B \tag{3.B.59}$$

$$\dot{B} = -(\tfrac{1}{2}\gamma_{j'}+i\omega_{j'_o})B - \tfrac{1}{2}\Gamma_{j''_o}A \tag{3.B.60}$$

Taking Laplace transforms for initial time t' = 0 we obtain simultaneous equations

$$(s+\tfrac{1}{2}\gamma_{j''}+i\omega_{j''_o})L(A) = -\tfrac{1}{2}\Gamma_{j'_o}L(B) + A(0) \tag{3.B.61}$$

$$(s+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'_o})L(B) = -\tfrac{1}{2}\Gamma_{j''_o}L(A) + B(0) \tag{3.B.62}$$

Solving for L(A) and L(B) we obtain

$$L(A) = \frac{(s+\frac{1}{2}\gamma_{j'}+i\omega_{j'_o})}{Z}A(0) - \tfrac{1}{2}\Gamma_{j'_o}\frac{1}{Z}B(0) \tag{3.B.63}$$

$$L(B) = \frac{(s+\frac{1}{2}\gamma_{j''}+i\omega_{j''_o})}{Z}B(0) - \tfrac{1}{2}\Gamma_{j''_o}\frac{1}{Z}A(0) \tag{3.B.64}$$

where

$$\left.\begin{aligned} Z &= s^2 + bs + c = (s-s_1)(s-s_2) \\ b &= \tfrac{1}{2}(\gamma_{j'}+\gamma_{j''}) + i(\omega_{j'_o}+\omega_{j''_o}) \\ c &= (\tfrac{1}{2}\gamma_{j'}+i\omega_{j'_o})(\tfrac{1}{2}\gamma_{j''}+i\omega_{j''_o}) - \tfrac{1}{4}\Gamma_{j'o}\Gamma_{j''o} \end{aligned}\right\} \tag{3.B.65}$$

If we now separate the terms in eqs. (3.B.53) and (3.B.54) into partial fractions and take the inverse transforms we obtain:

$$\left.\begin{aligned} P_{ij''}(t)|0> = {} & \frac{1}{(s_1-s_2)}\left[(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{s_1t} - (s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{s_2t}\right] \\ & \frac{-\frac{1}{2}\Gamma_{j'o}}{(s_1-s_2)}\left[e^{s_1t} - e^{s_2t}\right]P_{ij'}(0)|0> \end{aligned}\right\} \tag{3.B.66}$$

$$\left.\begin{aligned} P_{ij'}(t)|0> = {} & \frac{1}{(s_1-s_2)}\left[(s_1+\tfrac{1}{2}\gamma_{j''}+i\omega_{j''o})e^{s_1t} - (s_2+\tfrac{1}{2}\gamma_{j''}+i\omega_{j''o})e^{s_2t}\right] \\ & \times P_{ij'}(0)|0> \frac{-\frac{1}{2}\Gamma_{j''o}}{(s_1-s_2)}\left[e^{s_1t} - e^{s_2t}\right]P_{ij''}(0)|0> \end{aligned}\right\} \tag{3.B.67}$$

We shall find values s_1 and s_2 later.

Now what we are interested in calculating is the correlation function:

$$\left.\begin{aligned} f_{\hat{R}_{j''}}(t_R,t_{R'}) &= \frac{R^2 c}{2\pi\omega_{j''}\hbar} \langle \underline{E}^{(+)\dagger}(\underline{R},t_R).\underline{E}^{(+)}(\underline{R},t_{R'})\rangle \\ &= \frac{R^2 c}{\omega_{j''}} \frac{1}{V} \sum_{\ell,\sigma} \omega_\ell \langle a^\dagger_{\ell\sigma}(t_R) a_{\ell\sigma}(t_{R'})\rangle \end{aligned}\right\} \tag{3.B.68}$$

and this is determined by $\langle a^\dagger_{\ell\sigma}(t_R)a_{\ell\sigma}(t_{R'})\rangle$

where averaging is performed over initial atomic and radiation states. Assuming that there is no radiation present initially and the atom is in state $|j''\rangle$ this can be written:

$$\langle j''| \ {}_{rad}\langle 0|a^\dagger_{\ell\sigma}(t_R)a_{\ell\sigma}(t_{R'})|0\rangle_{rad} \ |j''\rangle \tag{3.B.69}$$

If we put $t_R = t_{R'}$ then the expression

$$\underline{f_{\hat{R}_{j''}}(t_R,t_R)} = \frac{R^2 c}{\omega_{j''}} \frac{1}{V} \sum_{\ell,\sigma} \omega_\ell \langle j''| \ {}_{rad}\langle 0|a^\dagger_{\ell\sigma}(t_R)a_{\ell\sigma}(t_R)|0\rangle_{rad} \ |j''\rangle \tag{3.B.70}$$

gives the average photon emission rate into solid angle $d\Omega_{\hat{R}}$ for spontaneous emission. This is the <u>actual intensity that would be measured by an ordinary photo detector</u> at point $\underline{R}$ at time t_R or by a similar device.[44] (N.B. it contains a summation over all possible modes.)

We now calculate

$$\langle j''| \ {}_{rad}\langle 0|a^\dagger_{\ell\sigma}(t_R)a_{\ell\sigma}(t_{R'})|0\rangle_{rad} \ |j''\rangle$$

from the equations for $P_{ij''}(t)\,|0\rangle_{rad}|j''\rangle$ and $P_{ij'}(t)|0\rangle_{rad}|j'\rangle$ given below:

$$
\left.\begin{aligned}
&P_{ij''}(t)|0\rangle_{rad}|j''\rangle = |i,t\rangle\langle j'',t|j'',0\rangle|0\rangle_{rad} \\
&= \frac{1}{(s_1-s_2)}\left[(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{s_1 t} - (s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{s_2 t}\right] \quad |i,0\rangle
\end{aligned}\right\} \quad (3.B.71)
$$

$$
\left.\begin{aligned}
&P_{ij'}(t)|0\rangle_{rad}|j''\rangle = |i,t\rangle\langle j',t|j'',0\rangle|0\rangle_{rad} \\
&= \frac{-\frac{1}{2}\Gamma_{j''o}}{(s_1-s_2)}\left[e^{s_1 t} - e^{s_2 t}\right] |i,0\rangle
\end{aligned}\right\} \quad (3.B.72)
$$

Applying operators $|0\rangle_{rad}$ and $|j''\rangle$ to the RHS of eq. (3.A.8) we obtain:

$$
\left.\begin{aligned}
&a_{\ell\sigma}(t_R)|0\rangle_{rad}|j''\rangle = ig_{\ell\sigma j'}\int_0^{t_R} dt' e^{-i\omega_\ell(t_R-t')}P_{ij'}(t')|0\rangle_{rad}|j''\rangle \\
&+ ig_{\ell\sigma j''}\int_0^{t_R} dt'' e^{-i\omega_\ell(t_R-t'')}P_{ij''}(t'')|0\rangle_{rad}|j''\rangle
\end{aligned}\right\} \quad (3.B.73)
$$

and substituting for $P_{ij'}(t')|0\rangle_{rad}|j''\rangle$ and $P_{ij''}(t'')|0\rangle_{rad}|j''\rangle$ we obtain

$$
\left.\begin{aligned}
&a_{\ell\sigma}(t_R)|0\rangle_{rad}|j''\rangle = -ig_{\ell\sigma j'}e^{-i\omega_\ell t_R}\int_0^{t_R} dt' \frac{\frac{1}{2}\Gamma_{j''o}}{s_1-s_2} \times \\
&\qquad \left[e^{(s_1+i\omega_\ell)t'} - e^{(s_2+i\omega_\ell)t'}\right] |i,0\rangle \\
&+ ig_{\ell\sigma j''}e^{-i\omega_\ell t_R}\int_0^{t_R} dt'' \frac{1}{(s_1-s_2)}\left[(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{(s_1+i\omega_\ell)t''}\right. \\
&\qquad \left. - (s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{(s_2+i\omega_\ell)t''}\right] \quad |i,0\rangle \\
&= \frac{-ig_{\ell\sigma j'}\frac{1}{2}\Gamma_{j''o}e^{-i\omega_\ell t_R}}{(s_1-s_2)}\int_0^{t_R} dt'(e^{(s_1+i\omega_\ell)t'} - e^{(s_2+i\omega_\ell)t'})|i,0\rangle \\
&+ \frac{ig_{\ell\sigma j''}e^{-i\omega_\ell t_R}}{(s_1-s_2)}\int_0^{t_R} dt''\left[(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'p})e^{(s_1+i\omega_\ell)t''} - (s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})\times\right. \\
&\qquad \left. e^{(s_2+i\omega_\ell)t''}\right] |i,0\rangle
\end{aligned}\right\} \quad (3.B.74)
$$

Before integrating we shall find s_1 and s_2

N.B.
$$s_{1,2} = \frac{-b \pm\sqrt{b^2-4c}}{2}$$
$$= -\tfrac{1}{2}\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''}) + i(\omega_{j'o}+\omega_{j''o})\right] \pm \tfrac{1}{2}\sqrt{p+iq}$$

where $p = \frac{1}{4}(\gamma_{j'}+\gamma_{j''})^2 - (\omega_{j'o}+\omega_{j''o})^2 - (\gamma_{j'}\gamma_{j''}-\Gamma_{j'o}\Gamma_{j''o})$
$+ 4\omega_{j'o}\omega_{j''o}$

$$q = (\gamma_{j'}+\gamma_{j''})(\omega_{j'o}+\omega_{j''o}) - 2(\gamma_{j'}\omega_{j''}+\gamma_{j''}\omega_{j'o}) \tag{3.B.75}$$

Now let
$$\sqrt{p+iq} = x + iy$$
∴ on squaring both sides of the equation
$$p + iq = (x^2-y^2) + i2xy$$
and on equating real and imaginary parts
$$p = (x^2-y^2)$$
$$q = 2xy$$
solving the last 2 equations we obtain
$$x = \sqrt{\frac{p+\sqrt{p^2+q^2}}{2}} \quad \text{and} \quad y = \sqrt{\frac{-p+\sqrt{p^2+q^2}}{2}} \tag{3.B.76}$$

so that
$$s_{1,2} = -\tfrac{1}{2}\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''}) \mp x\right] - i\tfrac{1}{2}\left[(\omega_{j'o}+\omega_{j''o}) \mp y\right] \tag{3.B.77}$$

and these roots are referred to as $-s^{\pm}$ in the paper.[4]

$$s_1 + s_2 = -\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''}) - i(\omega_{j'o}+\omega_{j''o}) = -(s_+ + s_-)$$
$s_1 - s_2 = x + iy = -s_+ + s_- = m$ where x and y are given above
$$-s_1 = s_+ = \tfrac{1}{2}\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''})-x\right] + i\tfrac{1}{2}\left[(\omega_{j'o}+\omega_{j''o})-y\right]$$
$$-s_2 = s_- = \tfrac{1}{2}\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''})+x\right] + i\tfrac{1}{2}\left[(\omega_{j'o}+\omega_{j''o})+y\right] \tag{3.B.78}$$

If we consider times $t_R \gg \gamma_{j'}^{-1}$ and $\gamma_{j''}^{-1}$ then $e^{-1/4(\gamma_{j'}+\gamma_{j''})t_R} \to 0$

and thus $e^{s_1 t_R}$ and $e^{s_2 t_R} \to 0$

$$\left.\begin{aligned}&\text{and } \int_0^{t_R} dt' \left[e^{(s_1+i\omega_\ell)t'} - e^{(s_2+i\omega_\ell)t'}\right]\\ &= \left[\frac{e^{(s_1+i\omega_\ell)t_R} - 1}{s_1+i\omega_\ell} - \frac{e^{(s_2+i\omega_\ell)t_R} - 1}{s_2+i\omega_\ell}\right] \doteq - \left[\frac{1}{(s_1+i\omega_\ell)} - \frac{1}{s_2+i\omega_\ell}\right]\end{aligned}\right\} \quad (3.B.79)$$

$$\left.\begin{aligned}&\text{and } \int_0^{t_R} dt'' \left[(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})e^{(s_1+i\omega_\ell)t''} + (s_2+\tfrac{1}{2}\gamma_{j''}+i\omega_{j''o})e^{(s_2+i\omega_\ell)t''}\right]\\ &\doteq - \left[\frac{(s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})}{(s_1+i\omega_\ell)} - \frac{(s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})}{s_2+i\omega_\ell}\right]\end{aligned}\right\} (3.B.80)$$

Thus eq. (3.B.64) becomes

$$a_{\ell\sigma}(t_R \gg \gamma_j^{-1})|0\rangle_{rad}\,|j''\rangle = \frac{+ig_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o}e^{-i\omega_\ell t_R}}{m}\left[\frac{1}{s_1+i\omega_\ell} - \frac{1}{s_2+i\omega_\ell}\right] |i,0\rangle$$

$$- \frac{ig_{\ell\sigma j''}e^{-i\omega_\ell t_R}}{m}\left[\frac{s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o}}{s_1+i\omega_\ell} - \frac{s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o}}{s_2+i\omega_\ell}\right] |i,0\rangle$$

$$= \frac{ie^{-i\omega_\ell t_R}}{m(s_1+i\omega_\ell)(s_2+i\omega_\ell)}\Big[g_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o}((s_2+i\omega_\ell)-(s_2+i\omega_\ell))$$

$$- g_{\ell\sigma j''}((s_1+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})(s_2+i\omega_\ell)-(s_2+\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})(s_1+i\omega_\ell))\Big] |i,0\rangle$$

$$= \frac{ie^{-i\omega_\ell t_R}}{m(s_1+i\omega_\ell)(s_2+i\omega_\ell)}\Big[-g_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o}m$$

$$- g_{\ell\sigma j''}((\tfrac{1}{2}\gamma_{j'}+i\omega_{j'o})(-m) + i\omega_\ell(m))\Big] |i,0\rangle$$

$$\left.\begin{aligned}&\text{i.e. } a_{\ell\sigma}(t_R \gg \gamma_j^{-1})|0\rangle_{rad}\,|j''\rangle = \frac{-ie^{-i\omega_\ell t_R}}{(s_1+i\omega_\ell)(s_2+i\omega_\ell)}\\ &\left[g_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o} - g_{\ell\sigma j''}(\tfrac{1}{2}\gamma_{j'}+i(\omega_{j'o}-\omega_\ell))\right] |i,0\rangle\end{aligned}\right\} (3.B.81)$$

Hence, multiplying the last equation by its hermitian conjugate

$$ {}_{\text{rad}}\langle 0|\langle j''|a^{\dagger}_{\ell\sigma}(t_R \gg \gamma^{-1})a_{\ell\sigma}(t'_R \gg \gamma^{-1})|j''\rangle|0\rangle_{\text{rad}} = |b_{\ell\sigma}|^2 $$

$$ = \frac{1}{(s_1+i\omega_\ell)(s_2+i\omega_\ell)(s_1^*-i\omega_\ell)(s_2^*-i\omega_\ell)}\Big[\{g_{\ell\sigma j'}\tfrac{1}{2}\Gamma^*_{j''o} - g_{\ell\sigma j''}(\tfrac{1}{2}\gamma_{j'}-i(\omega_{j'o}-\omega_\ell))\} $$

$$ \times\{g_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o} - g_{\ell\sigma j''}(\tfrac{1}{2}\gamma_{j'}+i(\omega_{j'o}-\omega_\ell))\}\Big] $$

and since $\langle i,0|0,i\rangle = 1$

$$ \left. |b_{\ell\sigma}|^2 = \frac{\{g_{\ell\sigma j'}\tfrac{1}{2}\Gamma^*_{j''o}-g_{\ell\sigma j''}(\tfrac{1}{2}\gamma_{j'}+i(\omega_\ell-\omega_{j'o}))\}\times\{g_{\ell\sigma j'}\tfrac{1}{2}\Gamma_{j''o} - g_{\ell\sigma j''}(\tfrac{1}{2}\gamma_{j'}-i(\omega_\ell-\omega_{j'o}))\}}{\{1/4\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''})-x\right]^2 + \left[\omega_\ell-\tfrac{1}{2}(\omega_{j'o}+\omega_{j''o}) + \tfrac{1}{2}y\right]^2\}\times\{1/4\left[\tfrac{1}{2}(\gamma_{j'}+\gamma_{j''})+x\right]^2 + \left[\omega_\ell-\tfrac{1}{2}(\omega_{j'o}+\omega_{j''o})-\tfrac{1}{2}y\right]^2\}} \right\} \quad (3.B.82) $$

Now, if we neglect all frequency shifts, then

$$ \omega_{j'o} = \omega_{j'},\ \omega_{j''o} = \omega_{j''},\ \tfrac{1}{2}\Gamma_{j'o} = \tfrac{1}{2}\Gamma^*_{j'o} = \tfrac{1}{2}\Gamma_{j'},\ \tfrac{1}{2}\Gamma_{j''o} = \tfrac{1}{2}\Gamma^*_{j''o} = \tfrac{1}{2}\Gamma_{j''}. \quad (3.B.83) $$

We now also assume $g_{\ell\sigma j'} = g_{\ell\sigma j''}$ (3.B.84)

i.e. for the region of overlap $(\hat{e}_{\ell\sigma}\cdot \underline{p}_{j'}) = (\hat{e}_{\ell\sigma}\cdot \underline{p}_{j''})$, since $\underline{p}_{j'} \hat{=} \underline{p}_{j''}$.

We shall finally assume that all decay constants are practically equal,

i.e. $\gamma_{j'} = \gamma_{j''} = \Gamma_{j'} = \Gamma_{j''} = \gamma$, (3.B.85)

since $\gamma_{j'} = \frac{4}{3}\frac{p_{j'}^2}{\hbar}\frac{\omega_{j'}^3}{c^3}$, $\gamma_{j''} = \frac{4}{3}\frac{p_{j''}^2}{\hbar}\frac{\omega_{j''}^3}{c^3}$,

$$ \Gamma_{j'} = \frac{4}{3}\frac{\underline{p}_{j'}\cdot\underline{p}_{j''}}{\hbar}\frac{\omega_{j'}^3}{c^3},\quad \Gamma_{j''} = \frac{4}{3}\frac{\underline{p}_{j'}\cdot\underline{p}_{j''}}{\hbar}\frac{\omega_{j''}^3}{c^3} $$

and $\quad \underline{p}_{j'}^{\ 2} \doteq p_{j''}^{\ 2}$ ($\doteq \underline{p}_{j'} \cdot \underline{p}_{j''}$ for $\hat{p}_{j'} \cdot \hat{p}_{j''}$ small).

so that $\quad \dfrac{\gamma_{j''}}{\gamma_{j'}} \doteq \dfrac{\omega_{j''}^{\ 3}}{\omega_{j'}^{\ 3}} \doteq \dfrac{\gamma_{j''}}{\Gamma_{j'}} \doteq \dfrac{\Gamma_{j''}}{\gamma_{j'}}$

and if $\quad \dfrac{\omega_{j''}^{\ 3}}{\omega_{j'}^{\ 3}} \doteq 1 \quad$ then our assumption of equal decay constants is correct. (N.B. For Na, d_1 and d_2 $\dfrac{\omega_{j''}^{3}}{\omega_{j'}^{3}} = (\dfrac{5896}{5890})^3 = 1.003$ and even if the separation is around $1\overset{\circ}{A}$ $\dfrac{\omega_{j''}^{3}}{\omega_{j'}^{3}} \sim 1.005$.)

(In Chapter VII we assume that $\Gamma_{j''} \doteq \Gamma_{j'} = \Gamma$, i.e. $\omega_{j''}^{\ 3} \doteq \omega_{j'}^{\ 3}$ and point out that we could leave the dipole matrix elements $\underline{p}_{j'}$ and $\underline{p}_{j''}$ arbitrary so that $\gamma_{j'} \neq \gamma_{j''} \neq \Gamma$.) With these approximations

$$\left.\begin{aligned}
|b_{\ell\sigma}|^2 &= \frac{g_{\ell\sigma}^2(\omega_\ell-\omega_{j'})^2}{\left[\frac{1}{4}(\gamma-x)^2+(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''})+\frac{1}{2}y)^2\right]\left[\frac{1}{4}(\gamma+x)^2+(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''})-\frac{1}{2}y)^2\right]} \\
&\text{where } x=\sqrt{\frac{p+\sqrt{p^2+q^2}}{2}}\ , \quad y=\sqrt{\frac{-p+\sqrt{p^2+q^2}}{2}}\ ; \\
&\text{and } \ p = +\left[\gamma^2-(\omega_{j'}-\omega_{j''})^2\right],\ q=0. \\
&\text{I.E. } x=\sqrt{p}\ , \quad y=0.
\end{aligned}\right\} \qquad (3.B.86)$$

Thus

$$\left.\begin{aligned}
|b_{\ell\sigma}|^2 &= \frac{g_{\ell\sigma}^2(\omega_\ell-\omega_{j'})^2}{\left|\frac{1}{16}(\gamma^2-x^2)^2+(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^4+\frac{1}{2}(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^2(\gamma^2+x^2)\right|} \\
&\text{where } \quad \gamma^2-x^2 = \gamma^2-p = (\omega_{j'}-\omega_{j''})^2 \\
&\text{and } \quad \gamma^2+x^2 = \gamma^2+p = 2\gamma^2-(\omega_{j'}-\omega_{j''})^2.
\end{aligned}\right\} \qquad (3.B.87)$$

Therefore

$$|b_{\ell\sigma}|^2 = \frac{g_{\ell\sigma}^2(\omega_\ell-\omega_{j'})^2}{\left[\frac{1}{16}(\omega_{j'}-\omega_{j''})^4+(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^4+\frac{1}{2}(2\gamma^2-(\omega_{j'}-\omega_{j''})2)(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^2\right]}$$

$$= \frac{g_{\ell\sigma}^2(\omega_\ell-\omega_{j'})^2}{\gamma^2(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^2-\frac{1}{2}(\omega_{j'}-\omega_{j''})^2(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^2+(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^4+\frac{1}{16}(\omega_{j'}-\omega_{j''})^4}$$

$$|b_{\ell\sigma}|^2 = \frac{g_{\ell\sigma}^2(\omega_\ell-\omega_{j'})^2}{\left[\gamma^2(\omega_\ell-\frac{1}{2}(\omega_{j'}+\omega_{j''}))^2+(\omega_\ell-\omega_{j'})^2(\omega_\ell-\omega_{j''})^2\right]} \tag{3.B.88}$$

This equation for the spectral profile of the atomic decay is the same as that derived by Morozov and Shorygin[8] using the Heitler-Ma method with terms higher than quadratic in the coupling eonstant ignored. Comparing the above equation with their eq. (15) given below under the same approximations,

$$|b_{i\sigma}(\infty)|^2 = \frac{|V_{i\sigma}{}^j|^2}{\hbar^2}\left[\frac{(\omega_\sigma-\omega_{j'})^2}{\gamma^2(\omega_\sigma-\frac{1}{2}(\omega_{j'}+\omega_{j''}))+(\omega_\sigma-\omega_{j'})^2(\omega_\sigma-\omega_{j''})^2}\right]$$

where $\quad E_{i\sigma} = \hbar\omega_\sigma \quad (= \hbar\omega_\ell$ in our notation)

$$E_{j'} = \hbar\omega_{j'}$$

$$E_{j''} = \hbar\omega_{j''} \tag{3.B.89}$$

we see that $\quad g_{\ell\sigma}^2 \to \dfrac{|V_\ell{}^j|^2}{\hbar^2}$

i.e. $$\frac{2\pi\hbar\omega_\ell}{V}(\hat{e}_{\ell\sigma}\cdot p_j)^2 \to |V_\ell{}^j|^2 \tag{3.B.90}$$

where $V_\ell{}^j$ is the interaction Hamiltonian, which in the Heitler-Ma method is treated completely generally without regard to the interaction mechanism.

Thus, as in Morozov and Shorygin's paper, we see that the spontaneous emission line shape is altered by the consideration of the exchange of virtual photons between overlapping levels $E_{j'}$ and $E_{j''}$. If this exchange is ignored and also the probability amplitude of level j' then we obtain a dispersion curve with maximum at $\omega_\ell = \omega_{j''}$ and of half-width $\gamma_{j''}$:

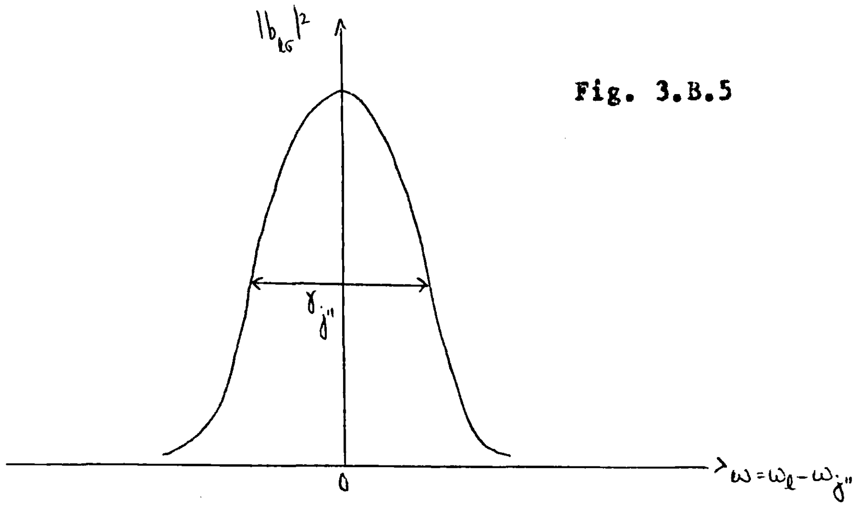


Fig. 3.B.5

In the language of Morozov and Shorygin's paper when we allow for the possibility that the state $|j'\rangle$ can be reached by absorption of a photon emitted virtually during the transition of the molecule from the level $E_{j''}$ to the level E_i we see that the line shape is no longer Lorentzian given by the formula (3.B.78). As in Morozov and Shorygin's paper the contours of the emission line for $\gamma = \omega' = 2\Delta$ and $\gamma = 2\omega' = 4\Delta$, where $\omega' = \omega_{j''} - \omega_{j'} = \epsilon_{j''} - \epsilon_{j'}$, as given earlier, can be obtained. We plot $y = \dfrac{|b_{\ell\sigma}|^2}{(g_{\ell\sigma}/\Delta)^2}$ along the ordinate and $X = \dfrac{\omega_\ell - \omega_{j''}}{\Delta}$ along the absissa so that the equation in terms of these values is

$$y = \left[\frac{(X+2)^2}{(\frac{\gamma}{\Delta})^2(X+1)^2 + X^2(X+2)^2}\right] \tag{3.B.91}$$

(3.B.92)

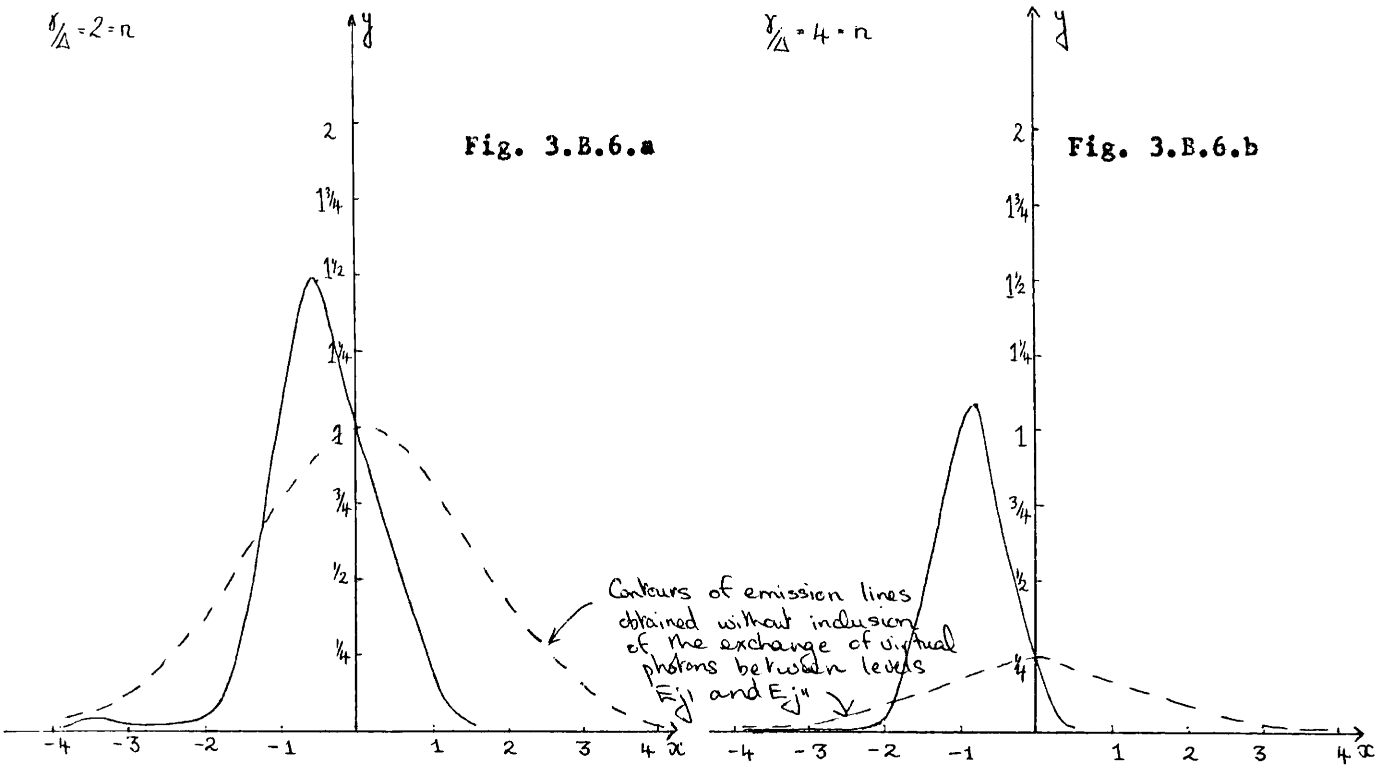


Fig. 3.B.6.a

Fig. 3.B.6.b

It would appear , from the following reasoning, that the ordinates of Morozov and Shorygin's curves are not correct. The positions of the turning points are given by $dy/dx = 0$

i.e. $(x+2)\left[X^4+6X^3+12X^2+(8+n^2)X+n^2\right] = 0$ where $n = \frac{\gamma}{\Delta}$ (3.B.93)

$x = -2$ is the position of a minimum.

To solve the quartic equation (see ref. [52] p. 42) we first reduce it to standard form by substituting $x = y - \frac{3}{2}$. The new equation is then

$$\left.\begin{aligned} & y^4 + py^2 + qy + r = 0 \\ & \quad \text{where} \quad p = -\frac{3}{2} \\ & \qquad\qquad\quad q = (n^2-1) \\ & \qquad\qquad\quad r = -\frac{1}{16}(8n^2+3) \end{aligned}\right\} \quad (3.B.94)$$

We may rewrite this by resolving it into 2 quadratic factors, which, since there is no y^3 term, must be of the form

$$\left.\begin{aligned} & (y^2+ay+b)(y^2-ay+c) = 0 \\ & \text{with solutions} \quad y_{1,2} = \frac{-a \pm\sqrt{a^2-4b}}{2} \quad y_{3,4} = \frac{a \pm\sqrt{a^2-4c}}{2} \end{aligned}\right\} \quad (3.B.95)$$

Comparing coefficients in these 2 expressions, we have

$$\left.\begin{aligned} p &= c + b - a^2 \\ q &= ac - ab \\ r &= bc \end{aligned}\right\} \qquad (3.B.96)$$

The first 2 of these equations give

$$\left.\begin{aligned} & b + c = p + a^2 \;, \quad b - c = -q/a \\ \text{so} \qquad & b = \tfrac{1}{2}(p+a^2-q/a) \;, \quad c = \tfrac{1}{2}(p+a^2+q/a) \end{aligned}\right\} \qquad (3.B.97)$$

The product of these is r, so

$$4r = (p+a^2)^2 - q^2/a^2 \qquad (3.B.98)$$

this being a bicubic in a^2:

$$a^2 + 2pa^4 + (p^2-4r)a^2 - q^2 = 0 \qquad (3.B.99)$$

One value, at least, of a^2 from this equation must be positive, so a real value of a results. The values of b and c then follow, all being real.

The cubic equation is

$$(a^2)^3 + 2p(a^2)^2 + (p^2-4r)(a^2) - q^2 = 0 \qquad (3.B.100)$$

Let $a^2 = \ell$

$$\therefore \qquad \ell^3 + 2p\ell^2 + (p^2-4r)\ell - q^2 = 0 \qquad (3.B.101)$$

Reducing this to standard form by the substitution $\ell = z - 2p/3$ we obtain

$$\left.\begin{aligned} & z^3 - (\tfrac{1}{3}\,p^2 + 4r)z - \tfrac{1}{27}(2p^3 - 72pr + 27q^2) = 0 \\ \text{i.e.} \quad & z^3 + 2n^2z - n^2(n^2 - 4) = 0 \end{aligned}\right\} \qquad (3.B.102)$$

Solving this, as in Appendix III, we obtain

$$\left.\begin{aligned}\alpha &= \left[\tfrac{1}{2}\left[n^2(n^2-4) + n^2\sqrt{n^4-\tfrac{184}{27}n^2+16}\right]\right]^{1/3}\\ \beta &= -\tfrac{2}{3}n^2\alpha^{-1}\end{aligned}\right\} \quad (3.B.103)$$

So, when $n = 2$,

$$\alpha = 2\sqrt{\tfrac{2}{3}} \quad \text{and} \quad \beta = -2\sqrt{\tfrac{2}{3}} \quad \therefore \alpha + \beta = 0 \qquad (3.B.104)$$

and when $n = 4$

$$\alpha \triangleq 6.650 \; ; \; \beta = -1.603 \quad \therefore \quad \alpha + \beta = 5.047 \qquad (3.B.105)$$

The real root of the cubic in ℓ when $n = 2$ is

$$\left.\begin{aligned}\ell &= \alpha + \beta - \tfrac{2p}{3}\\ &= \tfrac{-2p}{3}\\ &= 1 \;\therefore\; a_2 = 1\end{aligned}\right\} \quad (3.B.106)$$

and when $n = 4$ $\ell = 5.047+1 = 6.047 \;\therefore\; a_4 = \sqrt{6.047} = 2.459$ (3.B.107)

$$\left.\begin{aligned}b_2 &= \tfrac{1}{2}(p+a_2^2-q/a_2) = -7/4\\ c_2 &= \tfrac{1}{2}(p+a_2^2+q/a_2) = 5/4\end{aligned}\right\} \quad (3.B.108)$$

$\therefore\; y = \dfrac{-1 \pm 2\sqrt{2}}{2}$ for $n = 2$ are the real roots and $\underline{x} = y-3/2 = \underline{-2 \pm\sqrt{2}}$ (3.B.109)

$b_4 = -1.653$

$c_4 = 6.2$

$\therefore\; y = -3.0085, -5495$ are the real roots and $\underline{x = -0.9505, -4.5085}$ (3.B.110)

Thus for $n = 2$ maxima occur at $x = -2 \pm\sqrt{2} = -0.59$ and -3.4

and for $n = 4$ maxima occur at $x = -0.95$ and -4.5

When $n = 2$ the maximumof values are $y = 1.5, 0.04$

and when $n = 4$ the maximumof values are $y = 1.1, 0.02$

respectively.

By comparing the two figures the change in the line contours becomes increasingly apparent as $\frac{\gamma}{\Delta}$ increases and conversely for $\gamma << \Delta$ can be neglected since then there is no overlapping of line widths.

The new lines intersect the Lorentzian ones at $\omega_\ell = \omega_{j''}$ and approach zero at $\omega_\ell = \omega_{j''} - 2\Delta$ i.e. at $x = \pm 2$ since (1) the photon $\omega_\ell = \omega_{j''}$ cannot be virtual and does not participate in the exchange so that the intensity at $\omega_\ell = \omega_{j''}$ coincides with the intensity due to radiation from the single level $E_{j''}$; (2) the virtual photon $\omega_\ell = \omega_{j'}$ is real with regard to absorption when a transition to level j' occurs.

When the molecule is initially in state $E_{j'}$ the contour emission line is the mirror reflection relative to the plane perpendicular to the axis x, and passing through $X = \frac{\omega_o}{\Delta} -1$ of the contour obtained for $E_{j''}$.

Our calculation has avoided both the Markoff approximation, use of the fluctuation regression theorem and approximation to any order in coupling constants, g, which is necessary in perturbation theory where coupling is considered to be weak, and so our methods can be used when perturbation theory becomes invalid as e.g. in the presence of very intense radiation fields. Our main approximation, as in Lehmberg's papers [1], [2] is that no appreciable changes occur in the atomic states during times on the order of atomic periods.

CHAPTER IV

A MULTI-LEVEL ATOM WITH NON-OVERLAPPING LEVELS

A.. Heisenberg equations of motion

We have previously considered the cases of (i) the 2-level atom in Chapter II, and (ii) the 3-lèvel atom with exċited levels closely spaced, in Chapter III. We shall now go on to consider a multi-level atom with a total of ℓ levels. It has $(\ell-1)$ excited states $|2\rangle$, $|3\rangle$, ..., $|\ell\rangle$, with energies $\hbar\varepsilon_2$, $\hbar\varepsilon_3$, ..., $\hbar\varepsilon_\ell$, coupled to the ground state, $|1\rangle$, by a quantised multimode e.m. field. We shall allow for all possible transitions starting at a higher level y and ending in a lower one z.

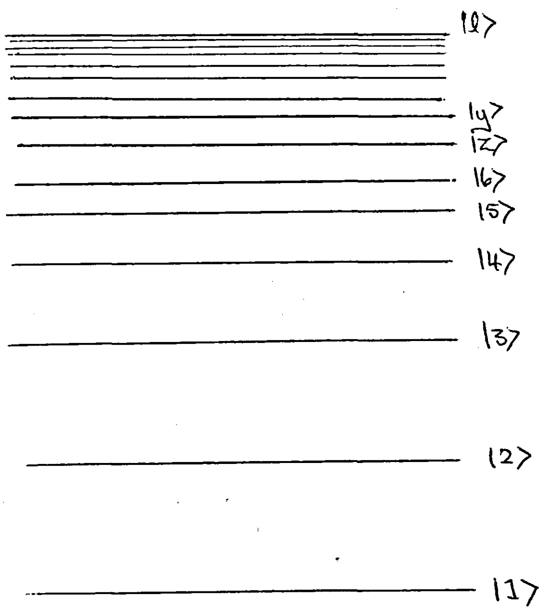


Fig. 4.A.1

The Hamiltonian, in the dipole approximation is then

$$H = \hbar \sum_{x=1}^{\ell} \epsilon_x P_{xx} + \frac{1}{8\pi} \int (|\underline{E}|^2 + |\underline{B}|^2) d^3r - \underline{E} \cdot \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} \underline{p}_{yz} (P_{zy} + P_{zy}^{\dagger}) \qquad (4.A.1)$$

and substituting for $\underline{E}$ and $\underline{B}$ from (1.B.58) and (1.B.59)

$$H = \hbar \sum_{x=1}^{\ell} txPxx + \hbar \sum_{\ell\sigma} \omega_\ell a^\dagger_{\ell\alpha} a_{\ell\sigma}$$

$$- \hbar \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} \sum_{\ell\sigma} g_{\ell\sigma yz}(s_{zy} a_{\ell\sigma} e^{i\underline{k}_\ell \cdot \underline{r}} + a^\dagger_{\ell\sigma} s_{zy} e^{-i\underline{k}_\ell \cdot \underline{r}}) \tag{4.A.2}$$

where $S_{zy}(t) = P_{zy}(t) + P^\dagger_{zy}(t)$

Also $g_{\ell\sigma yz} = \sqrt{\dfrac{2\pi\omega_{\ell yz}}{\hbar V}}\, \hat{e}_{\ell\sigma} \cdot \underline{p}_{yz}$ where all other symbols have their usual meaning. N.B. $\underline{p}_{yz} = \langle y|e\underline{x}|z\rangle$ and all $\underline{p}$'s are not necessarily in the same direction, as seen also in Chapter III.

Omitting HF terms $P_{zy} a_{\ell\sigma}$ and $a^\dagger_{\ell\sigma} P^\dagger_{zy}$ the Hamiltonian becomes, in the RWA:

$$H = \hbar \sum_{x=1}^{\ell} \epsilon_x P_{xx} + \hbar \sum_{\ell\sigma} \omega_\ell a^\dagger_{\ell\sigma} a_{\ell\sigma}$$

$$- \hbar \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} \sum_{\ell\sigma} g_{\ell\sigma yz}(P^\dagger_{zy} a_{\ell\sigma} e^{i\underline{k}_\ell \cdot \underline{r}} + a^\dagger_{\ell\sigma} P_{zy} e^{-i\underline{k}_\ell \cdot \underline{r}}) \tag{4.A.3}$$

N.B. Levelsy can decay to lower level z, emitting a photon into a broad band of closely spaced modes characterised by frequencies $\omega_{\ell yz}$. Similarly level y' can decay to level z' emitting photon band of frequencies $\omega_{\ell yz}$ etc. For simplicity we assume, as in Lehmberg[1] that all these bands do NOT overlap, i.e. $\omega_{\ell yz} \neq \omega_{\ell y'z'} \neq \omega_{\ell y''z''}$ etc. for all y's and z's. In Chapter VII we shall reintroduce overlapping terms for the 3-level atom, as in Chapter III. Since no atom is likely to have all levels overlapping it is better to introduce specific overlapping as and when required.

Assuming the atom lies at the origin of co-ordinates so that $\underline{r} = 0$ in the Hamiltonian, we can obtain Heisenberg equations of motion for $a_{\ell\sigma}(t)$, $P_{mn}(t)$, $P^{\dagger}_{mn}(t)$ (where $m < n$) and $P_{mm}(t)$ as given below:

$$\dot{a}_{\ell\sigma} = \frac{i}{\hbar}\left[H, a_{\ell\sigma}\right] = -i\omega_{\ell} a_{\ell\sigma} + i \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} g_{\ell\sigma,yz}\, P_{zy} \tag{4.A.4}$$

$$\dot{P}_{mn} = \frac{i}{\hbar}\left[H, P_{mn}\right]$$

$$= i\epsilon_{mn} P_{mn} - i \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} P_{yn} a_{\ell\sigma} + i \sum_{z=1}^{n-1} \sum_{\ell\sigma} g_{\ell\sigma nz} P_{mz} a_{\ell\sigma} \tag{4.A.5}$$

$$- i \sum_{z=1}^{m-1} \sum_{\ell\sigma} g_{\ell\sigma mz} a^{\dagger}_{\ell\sigma} P_{zn} + i \sum_{y=n+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma yn} a^{\dagger}_{\ell\sigma} P_{my}$$

$$= \dot{P}^{\dagger}_{nm}$$

where care has been taken to ensure that z remains $<y$ and m remains $<n$. $\epsilon_{mn} = \epsilon_m - \epsilon_n$.

N.B. The equation for $\dot{P}^{\dagger}_{mn}$ is simply the hermitian conjugate of (4.A.5) according to the definition of $P_{mn}(t)$ given in eq. (1.B.109a).

$$\dot{P}_{mm} = \frac{i}{\hbar}\left[H, P_{mm}\right]$$

$$= - i \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} P_{ym} a_{\ell\sigma} + i \sum_{z=1}^{m-1} \sum_{\ell\sigma} g_{\ell\sigma mz} P_{mz} a_{\ell\sigma} \tag{4.A.6}$$

$$- i \sum_{z=1}^{m-1} \sum_{\ell\sigma} g_{\ell\sigma mz} a^{\dagger}_{\ell\sigma} P_{zm} + i \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} a^{\dagger}_{\ell\sigma} P_{my}$$

Equation (4.A.4) has the formal solution

$$a_{\ell\sigma}(t) = a_{\ell\sigma}(0)e^{-i\omega_\ell t} + i \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} g_{\ell\sigma yz} \int_0^t dt' e^{-i\omega_\ell (t-t')} P_{zy}(t') \quad (4.A.7)$$

Now let us consider

$$\text{(i)} \quad \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} a_{\ell\sigma} \quad \text{and} \quad (4.A.8)$$

$$\text{(ii)} \quad \sum_{z=1}^{n-1} \sum_{\ell\sigma} g_{\ell\sigma nz} a_{\ell\sigma} \quad (4.A.9)$$

Substituting (4.A.7) in (4.A.8),

$$\left.\begin{aligned} \text{(i)} \quad \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} a_{\ell\sigma}(t) &= \sum_{y=m+1}^{\ell} \sum_{\ell\sigma} g_{\ell\sigma ym} a_{\ell\sigma}(0) e^{-i\omega_\ell t} \\ &+ \sum_{y=m+1}^{\ell} \sum_{y'=z'+1}^{\ell} \sum_{z'=1}^{\ell-1} \sum_{\ell\sigma} g_{\ell\sigma ym} g_{\ell\sigma y'z'} \int_0^t dt' e^{-i\omega_\ell (t-t')} P_{z'y'}(t') \end{aligned}\right\} (4.A.10)$$

∴ when $V \to \infty$, as in Chapter II

$$\left.\begin{aligned} &\sum_{y=m+1}^{\ell} \Big\{ \sum_{\ell\sigma} g_{\ell\sigma ym} a_{\ell\sigma}(t) - \frac{1}{\hbar} \underline{E}^{(0)}(t) . \underline{p}_{ym} \Big\} \\ &= \sum_{y'=z'+1}^{\ell} \sum_{z'=1}^{\ell-1} \sum_{y=m+1}^{\ell} \frac{i p_{ym} p_{y'z'}}{4\pi^2 C^3 \hbar} \int_0^\infty d\omega \omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} . \underline{p}_{ym})(\hat{e}_{\ell\sigma} . \underline{p}_{y'z'}) \\ &\times \int_0^t dt' e^{-i\omega(t-t')} P_{z'y'}(t') \end{aligned}\right\} (4.A.11)$$

where $\underline{E}_{+}^{(0)}(t) = \sum_{\ell,\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\, \hat{e}_{\ell\sigma} a_{\ell\sigma}(0) e^{-i\omega_\ell t}$ from (2.A.7)

N.B. In the term on the R.H.S. we have integrated over frequencies ω_ℓ common to photon transitions between levels y and m and y' and z' and vice versa. But, since we are ignoring overlapping of atomic levels, levels y and y' and levels m and z' must be the same so that

$$\sum_{y=m+1}^{\ell} \sum_{\ell\sigma} \left\{ g_{\ell\sigma ym} a_{\ell\sigma}(t) - \frac{1}{\hbar} E_{+}^{(0)}(t) \cdot p_{ym} \right\}$$

$$= \sum_{y'=z'+1}^{\ell} \sum_{z'=1}^{\ell-1} \sum_{y=m+1}^{\ell} \delta_{yy'} \delta_{mz'} \frac{i p_{ym} p_{y'z'}}{4\pi^2 c^3 \hbar} \int_0^\infty d\omega \omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \sum_{\sigma=1}^{2}$$

$$(\hat{e}_{\ell\sigma} \cdot p_{ym})(\hat{e}_{\ell\sigma} \cdot p_{y'z'}) \int_0^t dt' e^{-i\omega(t-t')} P_{z'y'}(t')$$

$$= i \sum_{y=m+1}^{\ell} \frac{p^2_{ym}}{4\pi^2 c^3 \hbar} \int_0^\infty d\omega \omega^3 \int_0^{4\pi} d\Omega_{\hat{k}} \sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot p_{ym})^2 \int_0^t dt' e^{-i\omega(t-t')} P_{my}(t')$$

N.B. $\sum_{\sigma=1}^{2} (\hat{e}_{\ell\sigma} \cdot p_{ym})^2 = 1 - (\hat{p} \cdot \hat{k})^2$ from (2.A.10)

and $\int_0^{4\pi} d\Omega_{\hat{k}} (1 - (\hat{p} \cdot \hat{k})^2) = \frac{8\pi}{3}$ from (2.A.13)

$$\therefore \sum_{y=m+1}^{\ell} \left(\sum_{\ell\sigma} g_{\ell\sigma ym} a_{\ell\sigma}(t) - \frac{1}{\hbar} E_{+}^{(0)}(t) \cdot p_{ym} \right) =$$

$$\frac{i2}{3\pi c^3 \hbar} \sum_{y=m+1} p^2_{ym} \int_0^\infty d\omega \omega^3 \int_0^t dt' e^{-i\omega(t-t')} P_{my}(t')$$

Proceeding, as in Chapters II and III, to take $P_{my}(t') = P_{my}(t) e^{-i\omega_{ym}(t'-t)}$,

and $\omega_{ym} t \gg 1$ for all y, we obtain

$$\sum_{y=m+1}^{\ell} \left(\sum_{\ell,\sigma} g_{\ell\sigma ym} a_{\ell\sigma}(t) - \frac{1}{\hbar} E_{+}^{(0)}(t) \cdot p_{ym} \right) = i \sum_{y=m+1} \left(\tfrac{1}{2}\gamma_{ym} - i\Omega_{ym} \right) P_{my}(t) \qquad (4.A.12)$$

Substituting (4.A.7) in (4.A.9), and proceeding as in (i), we obtain:-

$$\text{(ii)}\quad \sum_{z=1}^{n-1}\sum_{l,\sigma} g_{l\sigma nz}\, a_{l\sigma}(t) - \frac{1}{\hbar}\,\underline{E}_+^{(0)}(t)\cdot\underline{p}_{nz} = i\sum_{z=1}^{n-1}\left(\tfrac{1}{2}\gamma_{nz} - i\Omega_{nz-}\right)P_{zn}(t) \qquad (4.A.13)$$

where the spontaneous decay rate OR decay constant

$$\gamma_{mn} = \frac{4}{3\hbar} P_{mn}^2 \frac{\epsilon_{mn}^3}{c^3} \quad \text{and the frequency shift } \Omega_{mn-} = \frac{\gamma_{mn}}{\epsilon_{mn}^3}\, P\int_0^\infty \frac{d\omega}{2\pi}\frac{\omega^3}{\omega-\epsilon_{mn}} \qquad (4.A.14)$$

Rewriting eqs. (4.A.12) and (4.A.13) in terms of q_{ym} and q_{nz}

$$\left.\begin{aligned} \text{where}\quad q_{ym} &= \frac{1}{\hbar}\,\underline{E}_+^{(0)}(t)\cdot\underline{p}_{ym} = \sum_{l\sigma} g_{l\sigma ym}\, a_{l\sigma}(0)e^{-i\omega_l t} \\ q_{nz} &= \frac{1}{\hbar}\,\underline{E}_+^{(0)}(t)\cdot\underline{p}_{nz} = \sum_{l\sigma} g_{l\sigma nz}\, a_{l\sigma}(0)e^{-i\omega_l t} \end{aligned}\right\} \qquad (4.A.15)$$

we obtain

$$\left.\begin{aligned} \sum_{y=m+1}^{l}\sum_{l\sigma} g_{l\sigma ym}\, a_{l\sigma}(t) &= \sum_{y=m+1}^{l} q_{ym} + i\sum_{y=m+1}^{l}\left(\tfrac{1}{2}\gamma_{ym} - i\Omega_{ym-}\right)P_{my}(t) \\ \sum_{z=1}^{n-1}\sum_{l\sigma} g_{l\sigma nz}\, a_{l\sigma}(t) &= \sum_{z=1}^{n-1} q_{nz} + i\sum_{z=1}^{n-1}\left(\tfrac{1}{2}\gamma_{nz} - i\Omega_{nz-}\right)P_{zn}(t) \end{aligned}\right\} \qquad (4.A.16)$$

Similarly

$$\left.\begin{aligned} \sum_{z=1}^{m-1}\sum_{l\sigma} g_{l\sigma mz}\, a^+_{l\sigma}(t) &= \sum_{z=1}^{m-1} q^+_{mz} - i\sum_{z=1}^{m-1}\left(\tfrac{1}{2}\gamma_{mz} + i\Omega_{mz-}\right)P^+_{zm}(t) \\ \sum_{y=n+1}^{l}\sum_{l\sigma} g_{l\sigma yn}\, a^+_{l\sigma}(t) &= \sum_{y=n+1}^{l} q^+_{yn} - i\sum_{y=n+1}^{l}\left(\tfrac{1}{2}\gamma_{yn} + i\Omega_{yn-}\right)P^+_{ny}(t) \end{aligned}\right\} \qquad (4.A.17)$$

$$\left.\begin{aligned} \text{where}\quad q^+_{mz} &= \frac{1}{\hbar}\,\underline{E}_+^{(0)+}(t)\cdot\underline{p}_{mz} = \sum_{l\sigma} g_{l\sigma mz}\, a^+_{l\sigma}(0)e^{+i\ \ t} \\ q^+_{yn} &= \frac{1}{\hbar}\,\underline{E}_+^{(0)+}(t)\cdot\underline{p}_{yn} = \sum_{l\sigma} g_{l\sigma yn}\, a^+_{l\sigma}(0)e^{+i\ \ t} \end{aligned}\right\} \qquad (4.A.18)$$

Substituting eqs. (4.A.16) and (4.A.17) into eqs. (4.A.5) and (4.A.6) we obtain:-

$$
\left.
\begin{aligned}
\dot{P}_{nm} = &- \sum_{i=1}^{n-1} \sum_{j=1}^{m-1} \tfrac{1}{2}(\gamma_{ni} + \gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni-} + \Omega_{mj-}) \; P_{nm} \\
&- i \sum_{\substack{r=m+1 \\ (r>n)}}^{l} P^{+}_{nr}\, q_{rm} \quad + i \sum_{\substack{i=1 \\ (i<m)}}^{n-1} P^{+}_{im}\, q_{ni} \\
&- i \sum_{j=1}^{n-1} q^{+}_{mj}\, P_{jn} \quad + i \sum_{k=m+1}^{l} q^{+}_{km}\, P_{nk}
\end{aligned}
\right\} \qquad (4.A.19)
$$

$$
\left.
\begin{aligned}
\dot{P}_{mm} = &- \sum_{j=1}^{m-1} \left\{ \gamma_{mj}\, P_{mm} - i((P^{+}_{jm}\, q_{mj})_{j\,m} - q^{+}_{mj}\, P_{jm}) \right\} \\
&+ \sum_{r=m+1}^{l} \left\{ \gamma_{rm}\, P_{rr} - i((P^{+}_{mr}\, q_{rm})_{r\,m} - q^{+}_{rm}\, P_{mr}) \right\}
\end{aligned}
\right\} \qquad (4.A.20)
$$

Note the limitations on certain terms to make them non H-F (see Appendix IV). We shall ignore these extra limitations in Chapters V, VI and VII since we find they are fulfilled automatically.

It is of interest to note that Mollow (53), in his paper, derives equations of motion for diagonal and off-diagonal matrix elements of the atomic density operator, i.e. $\rho^{(s)}_{mm}(t)$ and $\rho^{(s)}_{nm}(t)$ for an atom with a number of levels. In his case he introduces a classical external field to couple states $|m\rangle$ and $|n\rangle$. If we also do this we can derive similar equations for comparison with those of Mollow eqs. (2.4a) and (2.4b). In order to incorporate classical driving terms in the Hamiltonian we add on a term

$$
H_{I.D}(t) = \hbar \sum_{y=z+1}^{l} \sum_{z=1}^{l-1} \left\{ P^{+}_{zy} \lambda_{yz} \mathcal{E}_D(t) + \lambda_{zy} \mathcal{E}^{*}_D(t) P_{zy} \right.
$$

$$
\text{where } \lambda_{yz} = \lambda_{zy} = \frac{\hat{e}_{OD} \cdot \mu_{yz}}{\hbar} = \frac{\hat{e}_{OD} \cdot \mu_{zy}}{\hbar}
$$

(cf. Appendix II for 2-level atom case)

or $$-\hbar \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} \lambda_{yz} \left\{ P^{+}_{zy}\mathcal{E}_D(t) + \mathcal{E}^{*}_D(t) P_{zy} \right\} \tag{4.A.21}$$

where the classical field is given, in terms of the positive and negative frequency parts $_D(t)$ and $_D(t)$, as

$$E_D(t) = (\mathcal{E}_D(t) + \mathcal{E}^{*}_D(t))\,\hat{e}_{OD} \tag{4.A.22}$$

and couples all pairs of levels.

$H_{I.D.}(t)$ introduces extra terms $\dot{P}_{mn_D}(t)$ and $\dot{P}_{mm_D}(t)$ into the expressions for $\dot{P}_{mn}(t)$ and $\dot{P}_{mm}(t)$ given by:-

$$\begin{aligned}
\dot{P}_{mn_D}(t) &= \frac{i}{\hbar}\left[H_{I.D.}, P_{mn}\right] \\
&= -i \sum_{y=z+1}^{\ell} \sum_{z=1}^{\ell-1} (P_{yn}\delta_{zm} - P_{mz}\delta_{yn})\mathcal{E}_D(t)\lambda_{yz} + \\
&\qquad \lambda_{zy}\mathcal{E}^{*}_D(t)\,(P_{zn}\delta_{ym} - P_{my}\delta_{zn}) \\
&= -i\sum_{y=m+1}^{\ell}\lambda_{ym}P_{yn}\mathcal{E}_D(t) + i\sum_{z=1}^{n-1}\lambda_{nz}P_{mz}\mathcal{E}_D(t) \\
&\qquad - i\sum_{z=1}^{m-1}\mathcal{E}^{*}_D(t)P_{zn}\lambda_{mz} + i\sum_{y=n+1}^{\ell}\mathcal{E}_D P_{my}\lambda_{ny}
\end{aligned} \tag{4.A.23}$$

$$\begin{aligned}
\dot{P}_{mm_D}(t) &= -i\sum_{y=z+1}^{\ell}\sum_{z=1}^{\ell-1}\Big\{ (P_{ym}\delta_{zm} - P_{mz}\delta_{ym})\lambda_{yz}\mathcal{E}_D(t) \\
&\qquad + \lambda_{zy}\mathcal{E}^{*}_D(t)(P_{zm}\delta_{ym} - P_{my}\delta_{zm})\Big\} \\
&= -i\sum_{y=m+1}^{\ell}\lambda_{ym}P_{ym}\mathcal{E}_D + i\sum_{z=1}^{m-1}\lambda_{mz}P_{mz}\mathcal{E}_D - i\sum_{z=1}^{m-1}\mathcal{E}_D P_{zm}\lambda_{mz} \\
&\qquad + i\sum_{y=m+1}^{\ell}\mathcal{E}^{*}_D P_{my}\lambda_{ym}
\end{aligned} \tag{4.A.24}$$

Hence (4.A.19) and (4.A.20) become:-

$$\dot{P}_{mn} = -\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\Big\{\tfrac{1}{2}(\gamma_{ni}+\gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni-} + \Omega_{mj-}\Big)\ P_{mn}$$

$$- \sum_{r=m+1}^{\ell} P^{+}_{nr}(q_{rm} + \lambda_{rm}\mathcal{E}_D(t)) + i\sum_{i=1}^{n-1} P^{+}_{im}(q_{ni} + \lambda_{ni}\mathcal{E}_D(t)) \qquad (4.A.25)$$

$$- \sum_{j=1}^{m-1}(q^{+}_{mj} + \lambda_{jm}\mathcal{E}^{\alpha}_D(t))P_{jn} + i \sum_{k=n+1}(q^{+}_{km} + \lambda_{nk}\mathcal{E}^{\alpha}_D(t))P_{mk}$$

and

$$\dot{P}_{mn} = -\sum_{j=1}^{m-1}\Big\{\gamma_{mj}\ P_{mm} - i(P^{+}_{jm}(q_{mj} + \lambda_{mj}\mathcal{E}_D(t)) - (q^{+}_{mj} + \lambda_{jm}\mathcal{E}^{\alpha}_D(t))P_{jm})$$

$$(r.A.26)$$

$$+ \sum_{r=m+1}^{\ell}\Big\{\gamma_{rm}\ P_{rr} - i(P^{+}_{mr}(q_{rm} + \lambda_{rm}\mathcal{E}_D(t)) - (q^{+}_{rm} + \lambda_{mr}\mathcal{E}^{\alpha}_D(t))P_{mr})$$

We may now take the initial radiation state to be a vacuum state and multiply the above two equations from left and right by ${}_{rad}\langle 0|$ and $|0\rangle_{rad}$. If we let ${}_{rad}\langle 0|\,P_{mn}\,|0\rangle_{rad} = X_{mn}$ and ${}_{rad}\langle 0\,|P_{mm}|\,0\rangle_{rad} = Y_m$

then

$$\dot{X}_{mn}(t) = -\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\ \tfrac{1}{2}(\gamma_{ni} + \gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni-} + \Omega_{mj-})\ X_{mn}$$

$$- i\sum_{r=m+1}^{\ell} X_{rn}\lambda_{rm}\mathcal{E}_D(t) + i\sum_{i=1}^{n-1} X_{mi}\lambda_{ni}\mathcal{E}_D(t) \qquad (4.A.27)$$

$$-\sum_{j=1}^{m-1}\lambda_{jm}\mathcal{E}_D(t)X_{jn} + i\sum_{k=n+1}^{\ell}\lambda_{nk}\mathcal{E}^{\alpha}_D(t)X_{mk}$$

$$\dot{Y}_m(t) = -\sum_{j=1}^{m-1}\Big\{\gamma_{mj}Y_m - i(X_{mj}\lambda_{mj}\mathcal{E}_D(t) - \lambda_{jm}\mathcal{E}^{\alpha}_D(t)X_{jm})$$

$$(4.A.28)$$

$$+ \sum_{r=m+1}^{\ell}\Big\{\gamma_{rm}\ Y_r - i(X_{rm}\lambda_{rm}\mathcal{E}_D(t) - \lambda_{mr}\mathcal{E}^{\alpha}_D(t)X_{mr})$$

From these equations we can obtain

$$\rho^{(s)}_{nm}(t) = \mathrm{Tr}\,\rho(t)P_{mn}(0) = \sum_s \langle s\,|\,X_{mn}(t)\ {}^{(s)}(0)\,|\,s\rangle$$

and $\rho^{(s)}_{mm}(t) = T_r\{\rho(t)\, P_{mm}(0)\} = \sum_s \langle s|Y_m(t)\rho^{(s)}(0)|s\rangle$ as follows:-

$$\left.\begin{aligned}
\dot{\rho}^{(s)}_{nm}(t) &= -\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\left\{\tfrac{1}{2}(\gamma_{ni}+\gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni} + \Omega_{mj})\right\}\rho^{(s)}_{nm}(t)\\
&\quad -i\sum_{r=m+1}^{\ell}\rho^{(s)}_{nr}(t)\lambda_{rm}\mathcal{E}_D(t) + i\sum_{i=1}^{n-1}\rho^{(s)}_{im}(t)\lambda_{ni}\mathcal{E}_D(t)\\
&\quad -\sum_{j=1}^{m-1}\lambda_{jm}\mathcal{E}^*_D(t)\rho^{(s)}_{nj}(t) + i\sum_{k=n+1}^{\ell}\lambda_{nk}\mathcal{E}^*_D(t)\rho^{(s)}_{km}(t)
\end{aligned}\right\}\quad(4.A.29)$$

$$\left.\begin{aligned}
{}^{(s)}_{mm}(t) &= -\sum_{j=1}^{m-1}\left\{\gamma_{mj}\rho^{(s)}_{mm}(t) - i(\rho^{(s)}_{jm}(t)\lambda_{mj}\mathcal{E}_D(t) - \lambda_{jm}\mathcal{E}^*_D(t)\rho^{(s)}_{mj}(t)\right\}\\
&\quad + \sum_{r=m+1}^{\ell}\left\{\gamma_{rm}\rho^{(s)}_{rr}(t) - i(\rho^{(s)}_{mr}(t)\lambda_{rm}\mathcal{E}_D(t) - \lambda_{mr}\mathcal{E}^*_D(t)\rho^{(s)}_{rm}(t))\right\}
\end{aligned}\right\}\quad(4.A.30)$$

Rewriting equations (2.4a) and (2.4b) of Mollow[53] in our notation by means of the following transformations:-

$$\bar{n}_j(t) \longrightarrow \rho^{(s)}_{mm}(t)$$

$$\alpha_{jk}(t) \longrightarrow \rho^{(s)}_{mn}(t) = \rho^{(s)+}_{nm}(t)$$

$$\sqrt{2}\lambda_{jk} \longrightarrow \lambda_{mn} = \frac{\mathbf{p}_{mn}\cdot e_{OD}}{\hbar}$$

$$\omega_{jk} \longrightarrow \epsilon_{mn} = \frac{(E_m - E_n)}{\hbar}$$

$$K_j = \sum_k K_{kj} \longrightarrow \gamma_m = \sum_n \gamma_{nm} \quad \text{where } n > m.$$

$$K'_{jk} = \tfrac{1}{2}(K_j + K_k) = \tfrac{1}{2}\sum_{\ell}\sum_{m}(K_{\ell j} + K_{mk}) \longrightarrow \gamma'_{mn} = \tfrac{1}{2}(\gamma_m + \gamma_n) = \tfrac{1}{2}\sum_j\sum_i(\gamma_{jm}+\gamma_{in})$$

$$\mathcal{E}(t) \longrightarrow \mathcal{E}_D(t)$$

We obtain:-

$$\dot{\rho}^{(s)}_{mm}(t) = -\sum_n \{ \gamma_{nm}\rho^{(s)}_{mm}(t) - i\,(\rho^{(s)}_{nm}(t)\lambda_{mn}\mathcal{E}_D(t) - \lambda_{nm}\mathcal{E}^*_D(t)\rho^{(s)}_{mn}(t))$$

$$+ i \sum_n \{ \gamma_{mn}\rho^{(s)}_{nn}(t) - i\,(\rho^{(s)}_{mn}(t)\lambda_{nm}\mathcal{E}_D(t) - \lambda_{mn}\mathcal{E}^*_D(t)\rho^{(s)}_{nm}(t))$$

and

$$\dot{\rho}^{(s)}_{mn}(t) = -\sum_i \sum_j \{ \tfrac{1}{2}(\gamma_{in} + \gamma_{jm}) + i\,\epsilon_{mn} \} \rho^{(s)}_{mn}(t)$$

$$- i \sum_{x(x=m,n)} \rho^{(s)}_{mx}(t)\lambda_{xn}\mathcal{E}_D(t) + i \sum_{x(x=m,n)} \rho^{(s)}_{xn}(t)\lambda_{mx}\mathcal{E}_D(t) \tag{4.A.31}$$

$$- \sum_{x(x=m,n)} \lambda_{xn}\mathcal{E}_D(t)\rho^{(s)}_{mx}(t) + i \sum_{x(x=m,n)} \lambda_{mx}\mathcal{E}^*_D(t)\rho^{(s)}_{xn}(t)$$

$$+ i\lambda_{mn}\,(\mathcal{E}_D(t) + \mathcal{E}^*_D(t))\,(\rho_{nn}(t) - \rho_{mm}(t))$$

so that

$$\left.\begin{aligned}
\dot{\rho}^{(s)}_{nm}(t) = &-\sum_i \sum_j (\tfrac{1}{2}(\gamma_{in} + \gamma_{jm}) + i\,\epsilon_{nm})\rho^{(s)}_{nm}(t) \\
&- i \sum_{x(x=m,n)} \lambda_{xm}\mathcal{E}^*_D(t)\rho^{(s)}_{nx}(t) + i \sum_{x(x=m,n)} \lambda_{nx}\mathcal{E}^*_D(t)\rho^{(s)}_{xm}(t) \\
&- i \sum_{x(x=m,n)} \rho^{(s)}_{nx}(t)\lambda_{xm}\mathcal{E}_D(t) + i \sum_{x(x=m,n)} \rho^{(s)}_{xm}(t)\lambda_{nx}\mathcal{E}_D(t) \\
&+ i\lambda_{nm}\,(\mathcal{E}_D(t) + \mathcal{E}^*_D(t))\,(\rho_{mm}(t) - \rho_{nn}(t))
\end{aligned}\right\} \tag{4.A.32}$$

So that the main differences are:-

1) The lack of specific limits to summations in Hollow's equations.

2) Indices on γ's are reversed.

3) Frequency shifts Ω are ignored in Hollows case.

4) Expression for $\rho^{(s)}_{rm}(t)$ appears to contain an extra term

$$i\,\lambda_{nm}(\mathcal{E}_D + \mathcal{E}^*_D)(\rho_{mm}(t) - \rho_{nn}(t))$$

but by redefining the $\mathcal{E}$'s we can write equation (4.A.32) as

$$\begin{aligned}
\dot{\rho}^{(s)}_{nm}(t) = &- \sum_i \sum_j (\tfrac{1}{2}(\gamma_{in} + \gamma_{jm}) + i\,\epsilon_{nm})\rho^{(s)}_{nm}(t) \\
&- i \sum_{x(x=m)} \lambda_{xm}\mathcal{E}^*_D(t)\rho^{(s)}_{nx}(t) + i \sum_{x(x=n)} \lambda_{nx}\mathcal{E}^*_D(t)\rho^{(s)}_{xm}(t) \\
&- i \sum_{x(x=m)} \rho^{(s)}_{nx}(t)\,\lambda_{xm}\mathcal{E}_D(t) + i \sum_{x(x=n)} \rho^{(s)}_{xm}(t)\,\lambda_{nx}\mathcal{E}_D(t)
\end{aligned} \quad (4.A.33)$$

which is equivalent to (4.A.29).

B. The Power Spectrum

In Chapter III we were able to calculate the spectral profile for spontaneous emission from

$$_{rad}\langle 0| \langle j''| a^{+}_{\ell\sigma}(t_R >> \gamma^{-1})\, a_{\ell\sigma}(t_R >> \gamma^{-1})|j''\rangle |0\rangle_{rad}$$

by finding equations for $P_{jj'}(t)|0\rangle_{rad}|j''\rangle$ and $P_{jj''}(t)|0\rangle_{rad}|j''\rangle$.
It is obvious that this method is of no use in the present case since multiplying equations (4.A.19) and (4.A.20) from the RHS by $|0\rangle_{rad}$ still leaves terms of the form $q^{+}_{mj} P_{jn}|0\rangle_{rad}$, $q^{+}_{kn} P_{mk}|0\rangle_{rad}$, which cause difficulty. Similarly when classical driving terms are introduced, as in equations (4.A.25) and (4.A.26) we obtain expressions containing terms $(q^{+}_{mj} + \lambda_{jm}\mathcal{E}^{*}_{D}(t))P_{jn}|0\rangle_{rad}$ and $(q^{+}_{kn} + \lambda_{nk}\mathcal{E}^{*}_{D}(t))P_{mk}|0\rangle_{rad}$. The alternative is to solve the equations (4.A.29) and (4.A.30) for the reduced density operator components and then, as in Mollow's paper[9], resort to the Markoff approximation in order to calculate the 1st-order field correlation function which, we have seen, is determined by the 2-time atomic correlation function for the transition under consideration (see expression for $L_{jj'}(t_R, t'_R)$ given in Chapter III, equation (3.B.45)). If we keep the initial time t' arbitrary we will be able to calculate the overall effect of absorptions and emissions between the required levels since then we will not be able to specify the initial atomic state. The specific way in which we calculate the spectral profiles in various cases will be outlined in the following three chapters for the specific cases of the K atom, and H atom and the 3-level atom, where allowance is made for the possibility of overlapping upper levels in the latter case.

In fact, following the arguments of the previous chapter,

$$\underline{E}_{+}(\underline{R}, t_R) = \underline{E}^{(0)}_{+}(\underline{R}, t_R) + \sum_{y=z+1}^{\ell}\sum_{z=1}^{\ell-1} k^2_{yz} \frac{\{\underline{p}_{yz} - (\hat{R}_a \cdot \underline{p}_{yz})\hat{R}_a\}}{R_a} P_{zy}(t) e^{-ik_{yz}r} \quad (4.B.1)$$

where if r = 0 (atom at origin), $R_a = R$ and so

$$\underline{E}_+(\underline{R},t_R) = \underline{E}_+^{(0)}(\underline{R},t_R) + \sum_{y=z+1}^{l}\sum_{z=1}^{l-1} k_{yz}^2 \frac{\{\underline{p}_{yz} - (\hat{R}\cdot\underline{p}_{yz})\hat{R}\}}{R} P_{zy}(t) \tag{4.B.2}$$

$$f_{\hat{R}y'z'}(t_R,t'_R) = \frac{R^2 c}{2\pi\epsilon_{y'z'}\hbar} \langle|\underline{E}_+^+(R,t_R).\underline{E}_+(\underline{R},t'_R)|\rangle \tag{4.B.3}$$

and so, ignoring interference terms,

$$f_{\hat{R}y'z'}(t_R,t'_R) = f_{\hat{R}y'z'}^{(0)}(t_R,t'_R) + L_{\hat{R}}(t_R,t'_R) \tag{4.B.4}$$

where $$f_{\hat{R}y'z'}^{(0)}(t_R,t'_R) = \frac{R^2 c}{2\pi\epsilon_{y'z'}\hbar} \langle|\underline{E}_+^{(0)+}(\underline{R},t_R)\cdot\underline{E}_+(\underline{R},t'_R)|\rangle \tag{4.B.5}$$

and
$$\left.\begin{aligned} L_{\hat{R}y'z'}(t_R,t'_R) = \frac{R^2 c}{2\pi\epsilon_{y'z'}\hbar} \sum_{y_1=z_1+1}^{l}\sum_{z_1=1}^{l-1}\sum_{y_2=z_2+1}^{l}\sum_{z_2=1}^{l-1} k_{y_1z_1}^2 k_{y_2z_2}^2 \times \\ \langle|P_{z_1y_1}^+(t_R)P_{z_2y_2}(t'_R)|\rangle \left\{\frac{\underline{p}_{y_1z_1}-(\hat{R}\cdot\underline{p}_{y_1z_1})\hat{R}}{R}\right\}\left\{\frac{\underline{p}_{y_2z_2}-(\hat{R}\cdot\underline{p}_{y_2z_2})\hat{R}}{R}\right\}\end{aligned}\right\} \tag{4.B.6}$$

If we consider only transitions between the pair of levels y' and z' and ignore all others then (see ref. 124 eq. (3.5) where the emission spectrum only is considered)

$$L_{\hat{R}y'z'}(t_R,t'_R) = \gamma_{y'z'} \langle|P_{z'y'}(t_R)\,P_{z'y'}(t'_R)|\rangle \tag{4.B.7}$$

N.B. The effect of other transitions is taken into account, since the equations of motion to be solved are coupled equations. Only when there are overlapping levels is it necessary to take into account more than one 2-time correlation function since in such cases the additional 2-time correlation functions produce intensity distributions for the same frequency range (see Chapters III and VII).

Hence the intensity distribution is given by

$$\left.\begin{aligned} f_{y'z'}(t_R, t'_R) = \frac{R^2 c}{(2\pi c)^3 \epsilon_{y'z'}} \sum_{\sigma=1}^{2} \oint d\Omega_{\hat{R}} \oint d\Omega_{\hat{k}} \int_0^\infty d\omega\, \omega^3 e^{i\omega(t_R - t_R')} \\ \times \langle | a^+_{\ell\sigma}(0)\, a_{\ell\sigma}(0) | \rangle \\ + \gamma_{y'z'} \langle | P_{z'y'}(t_R)\ P_{z'y'}(t'_R) | \rangle \end{aligned}\right\} \quad (4.B.8)$$

The first term, as explained earlier, only gives the energy spectrum for the INCIDENT field. To find the energy or POWER SPECTRUM for the scattered field we must calculate

$$I_{y'z'} = \gamma_{y'z'} \langle | P^+_{z'y'}(t_R)\ P_{z'y'}(t'_R) | \rangle \quad (4.B.9)$$

i.e. basically we need to know the 2-time atomic correlation function

$$\langle | P^+_{z'y'}(t_R)\ P_{z'y'}(t'_R) | \rangle \quad (4.B.10)$$

C. Mollow's method for evaluation the power spectrum for the scattered light

We will now outline the general method for evaluating the 2-time atomic correlation function $\langle | P^+_{z'y'}(t_R)\ P_{z'y'}(t'_R) | \rangle$, based on Mollow's method, explained in ref. (9), which avoids use of the fluctuation regression theorem. This function can be expressed in terms of the Schrodinger density operator at time t_R i.e. $\rho(t'_R)$ and the time-independent Schrodinger operators $P^+_{z'y'}(0)$ and $P_{z'y'}(0)$ as follows

$$\langle | P^+_{z'y'}(t'_R) P_{z'y'}(t'_R) | \rangle = \mathrm{Tr}\left\{ \rho(0)\, P^+_{z'y'}(t'_R)\, P_{z'y'}(t''_R) \right\}$$

N.B.

$$\rho_s(t) = U(t,t')\, \rho_H(t')\, U^{-1}(t,t')$$
$$A_H(t) = U^{-1}(t,t')\, A_s(t')\, U(t,t')$$

ignoring subscript R, which indicates retarded time, we can write

$$\begin{aligned}
\langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle &= \mathrm{Tr}\{\rho(0)\, U^{-1}(t'',0)\, P^{+}_{z'y'}(0)\, \underline{U(t',0)\, U^{-1}(t'',0)} \times \\
&\qquad P_{z'y'}(0)\, U(t'',0)\} \\
&= \mathrm{Tr}\{\rho(0)\, U^{-1}(t',0)\, P^{+}_{z'y'}(0)\, \underline{U(t',t'')}\, P_{z'y'}(0) \times \\
&\qquad U(t'',0)\} \\
&= \mathrm{Tr}\{U^{-1}(t',0)[\rho(t')\, \underline{U(t',0)\, U^{-1}(t',0)}\, P^{+}_{z'y'}(0)\, U(t',t'') \times \\
&\qquad P_{z'y'}(0)\, U(t'',0)]\} \\
&= \mathrm{Tr}\{[\rho(t')\, P^{+}_{z'y'}(0)\, U(t',t'')\, P_{z'y'}(0)\, \underline{U(t'',0)]\, U^{-1}(t',0)}\} \\
&= \mathrm{Tr}\{\rho(t')\, P^{+}_{z'y'}(0)\, \underline{\underline{U(t',t'')}}\, P_{z'y'}(0)\, \underline{U(t'',t')}\} \\
&= \mathrm{Tr}\{\underline{\rho(t')\, P^{+}_{z'y'}(0)}\, \underline{\underline{U^{-1}(t'',t')}}\, P_{z'y'}(0)\, U(t'',t')\}
\end{aligned}$$

where $U(t'',t'$ is the time-development operator

According to the Markoff approximation, explained in Chapter I, we can factorise $\rho(t')$ as

$$\rho(t') = |i_{ph}\rangle\langle i_{ph}|\, \rho^{(s)}(t')$$

Hence

$$\langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle \simeq \mathrm{Tr}\{|i_{ph}\rangle\langle i_{ph}|\, \rho^{(s)}(t')\, P^{+}_{z'y'}(0)\, U^{-1}(t'',t')\, P_{z'y'}(0) \times U(t'',t')\} \qquad (4.C.1)$$

We can now compare this with the mean value of the atomic operator $P_{z'y'}$ at a given time t". Now the expectation value of $P_{z'y'}(t'')$ is

$$\left.\begin{aligned}
\langle P_{z'y'}(t'')\rangle = \rho_{y'z'}(t'') &= \mathrm{Tr}\{\rho(t'')\, P_{z'y'}(0)\} \\
&= \mathrm{Tr}\{U(t'',t')[\rho(t')\, U(t'',t')^{-1}\, P_{z'y'}(0)]\} \\
&= \mathrm{Tr}\{\underline{\underline{\rho(t')}}\, U(t'',t')^{-1}\, P_{z'y'}(0)\, U(t'',t')\} \quad \text{since } \mathrm{Tr}(AB) = \mathrm{Tr}(BA) \\
&= \mathrm{Tr}\{|i_{ph}\rangle\langle i_{ph}|\, \underline{\rho^{(s)}(t')}\, U(t'',t')^{-1}\, P_{z'y'}(0) \times \\
&\qquad U(t'',t')\} \quad \text{under Markoff approximation.}
\end{aligned}\right\} \quad (4.C.2)$$

Comparing RHS's of equations (4.C.1) and (4.C.2) shows that we can obtain the former from the latter by making the substitution

$$\rho^{(s)}(t') \longrightarrow \rho^{(s)}(t')\, P^{+}_{z'y'}(0) \tag{4.C.3}$$

so that then $\langle P_{z'y'}(t'')\rangle \longrightarrow \langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle$
(Without the Markoff approximation the required substitution is for the full density operator, $\rho(t') \longrightarrow \rho(t')\, P^{+}_{z'y'}(0)$).

Thus, in order to evaluate the 2-time correlation function $\langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle$, we must first evaluate the singe-time mean-value $\rho_{y'z'}(t'') \cdot \langle P_{z'y'}(t'')\rangle$, the only difference being that the non-hermitian operator $\rho^{(s)}(t')\, P^{+}_{z'y'}(0)$ must be used in place of the density operator $\rho^{(s)}(t')$.

Since we may write

$$U^{-1}(t'',t')\, P_{z'y'}(0)\, U(t'',t') = P_{z'y'}(t''-t') = P_{z'y'}(\tau)$$

a function of the time-difference only, then, from (4.C.1), we have

$$\begin{aligned}
\langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle &\doteq \mathrm{Tr}\left\{ |iph\rangle\langle iph|\, \rho^{(s)}(t')\, P^{+}_{z'y'}(0)\, P_{z'y'}(t''-t')\right\} \\
&= \sum_{s,R} \langle s,R|iph\rangle\langle iph|\, \rho^{(s)}(t')\, P^{+}_{z'y'}(0)\, P_{z'y'}(t''-t')\,|R,s\rangle \\
&= \sum_{s,R} \langle s,iph|\, \delta_{R,iph}\, \rho^{(s)}(t')\, P^{+}_{z'y'}(0)\, P_{z'y'}(t''-t')\,|R,s\rangle \\
&= \sum_{s} \langle s,iph|\, \rho^{(s)}(t')\, P^{+}_{z'y'}(0)\, P_{z'y'}(t''-t')\,|iph,s\rangle \\
&= \langle iph|\langle 1|\, \rho^{(s)}(t')\,|y'\rangle\langle z'|\, P_{z'y'}(t''-t')\,|1\rangle|iph\rangle \\
&\quad + \langle iph|\langle 2|\, \rho^{(s)}(t')\,|y'\rangle\langle z'|\, P_{z'y'}(t''-t')\,|2\rangle|iph\rangle \\
&\quad + \langle iph|\langle \ell|\, \rho^{(s)}(t')\,|y'\rangle\langle z'|\, P_{z'y'}(t''-t')\,|\ell\rangle|iph\rangle
\end{aligned}$$

$$\therefore \langle P^{+}_{z'y'}(t')\, P_{z'y'}(t'')\rangle \doteq \sum_{s=1}^{\ell} \rho^{(s)}_{sy'}(t')\, \langle iph|\langle z'|\, P_{z'y'}(t''-t')\,|s\rangle|iph\rangle \tag{4.C.4}$$

But

$$\rho_{y'z'}(t) = \langle P_{z'y'}(t) \rangle = \mathrm{Tr}\{|iph\rangle\langle iph|\,\rho^{(s)}(t')\,P^{+}_{z'y'}(0)\,P_{z'y'}(t''-t')\}$$

$$= \sum_{s,R} \langle s,R|iph\rangle\langle iph|\,\rho^{(s)}(t')\,P^{+}_{z'y'}(0)\,P_{z'y'}(t''-t')|R,s\rangle$$

$$= \sum_{s,R} \langle s,iph|\,\delta_{R,iph}\,\rho^{(s)}(t')\,P^{+}_{z'y'}(0)\,P_{z'y'}(t''-t')|R,s\rangle$$

$$= \sum_{s} \langle s,iph|\,\rho^{(s)}(t')\,P^{+}_{z'y'}(0)\,P_{z'y'}(t''-t')|iph,s\rangle$$

where $s = 1, 2, 3, \ldots, \ell$

$$= \langle iph|\langle 1|\,\rho^{(s)}(t')\,|y'\rangle\langle z'|\,P_{z'y'}(t''-t')\,|1\rangle|iph\rangle$$
$$+ \langle iph|\langle 2|\,\rho^{(s)}(t')\,|y'\rangle\langle z'|\,P_{z'y'}(t''-t')\,|2\rangle|iph\rangle$$
$$\vdots \qquad\qquad\qquad\qquad \vdots$$
$$+ \langle iph|\langle \ell|\,\rho^{(s)}(t')\,|y'\rangle\langle z'|\,P_{z'y'}(t''-t')\,|\ell\rangle|iph\rangle$$

$$\boxed{\rho_{y'z'}(t) \underset{M.A.}{\hat{=}} \rho^{(s)}_{y'z'}(t)} \quad \text{since } \langle iph|iph\rangle = 1 \qquad (4.C.5)$$

i.e. under the Markoff approximation there is no difference between the matrix elements of the full density operator and the reduced atomic density operator. This is convenient since it is easier to calculate the reduced density operator matrix elements $\rho^{(s)}_{y'z'}(t)$.

If, on solving the equations of motion for the reduced density matrix elements we find that $\rho^{(s)}_{y'z'}(t'')$ is of the following form:-

$$\rho^{(s)}_{y'z'}(t'') = \rho^{(s)}_{y'z'}(t'+\tau) = \sum_{y''=1}^{\ell}\sum_{z''=1}^{\ell} U_{y'z',y''z''}(\tau,t')\,\rho^{(s)}_{y''z''}(t') \qquad (4.C.6)$$

where $t'' = t' + \tau$ and $\tau > 0$

(τ is the time difference and t' the initial time), which is the case when <u>all</u> the reduced density matrix equations are interdependent (generally only a few are).

Making the substitution, given in eq. (4.C.3), in the R.H.S. of eq. (4.C.6)

i.e. $\rho^{(s)}_{y''z''}(t') = \langle y''|\rho^{(s)}(t')|z''\rangle \longrightarrow \langle y''|\rho^{(s)}(t')\,P^{+}_{z'y'}(0)\,|z''\rangle$

$$\left.\begin{aligned} &= \langle y''|\rho^{(s)}(t')\,|y'\rangle\langle z'|z''\rangle \\ &= \langle y''|\rho^{(s)}(t')\,|y'\rangle\,\delta_{z'z''} \end{aligned}\right\} \quad (4.C.7)$$

we obtain the 2-time atomic correlation function:-

$$\langle P^{+}_{z'y'}(t')\,P_{z'y'}(t'')\rangle \doteq \frac{1}{M.A.}\,\rho_{y'z'}(t'')$$
$$\left(\text{when } \rho^{(s)}(t') \longrightarrow \rho^{(s)}(t')\,P^{+}_{z'y'}(0)\right)$$

OR $\rho^{(s)}_{y'z'}(t'')$ with the same substitution (4.C.3),

$$\frac{1}{M.A.}\sum_{y''=1}^{\ell}\sum_{z''=1}^{\ell} u_{y'z',y''z''}(\tau,t')\,\rho^{(s)}_{y''y'}(t')\,\delta_{z'z''}$$

i.e.
$$\boxed{\langle P^{+}_{z'y'}(t')\,P_{z'y'}(t'')\rangle \doteq \frac{1}{M.A.}\sum_{y''=1}^{\ell} u_{y'z',y''z''}(\tau,t')\,\rho^{(s)}_{y''y'}(t')} \quad (4.C.8)$$

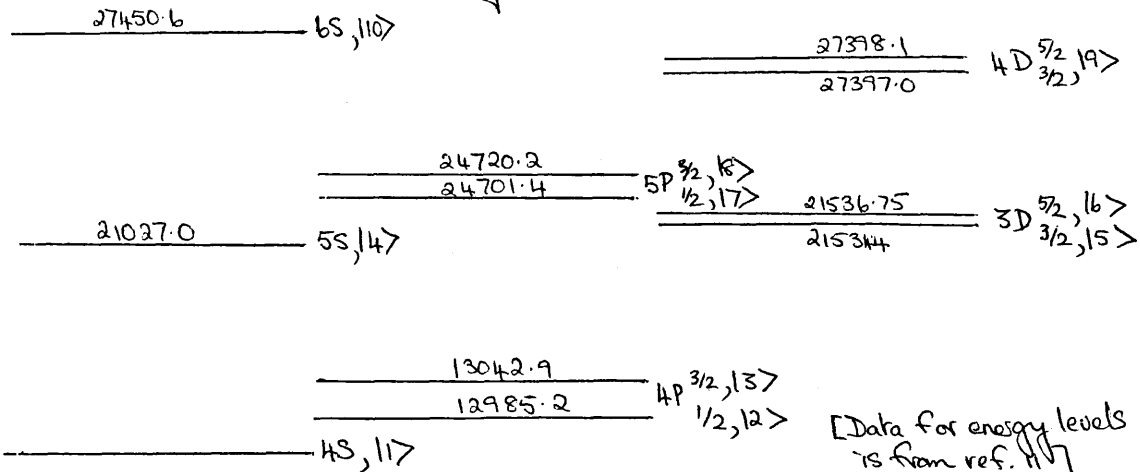
and this is the expression we shall be implementing in the following three chapters.

CHAPTER V

THE POTASSIUM ATOM DRIVEN BY TWO EXTERNAL FIELDS COUPLING LEVELS 4S AND $4D_{3/2}$ AND LEVELS $4P_{3/2}$ AND 6S RESPECTIVELY

In this Chapter we shall calculate the power spectra for certain transitions in the potassium atom. The model of the atom we shall use is as sketched below:

Fig. 5.1



[Data for energy levels is from ref. (11)]

It is composed of 10 of the energy levels, i.e. levels up to and including level 6S, but ignoring the degeneracy of levels 4S (ground state), 5S, 4D (unresolved experimentally[11]) and 6S. We also ignore the fact that level $4P_{3/2}$ has substates $|m| = 3/2$ and $|m| = 1/2$.

It is known that the dipole matrix element connecting levels $6S_{1/2}$ and $4P_{3/2}$ is much stronger than that connecting levels $4D_{3/2}$ and $4P_{1/2}$.

We shall number the levels as indicated in the diagram and could consider a general 10-level atom but since there is a wealth of literature (10)-(31) on transitions in this atom we shall particularise our equations for comparison with experimental results. From the literature we see that level 5S does not take part in any transitions but we include it all the same.

A. Equations of motion

From the equations (4.A.19) and (4.A.20) derived in Chapter IV for the transition operators for the multilevel-level atom we can deduce equations for the 10-level atom:-

$$\left.\begin{aligned}\dot{P}_{mn} = -i\sum_{i=1}^{n-1}&\left\{\tfrac{1}{2}(\gamma_{ni}+\gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni} + \Omega_{mj})\right\}P_{mn}\\ &-i\sum_{r=m+1}^{10} P^{+}_{nr}\,q_{rm} \qquad +i\sum_{i=1}^{n-1} P^{+}_{im}\,q_{ni}\\ &-i\sum_{j=1}^{m-1} q^{+}_{mj}\,P_{jn} \qquad +i\sum_{k=n+1}^{10} q^{+}_{kn}\,P_{mk}\end{aligned}\right\} \qquad (5.A.1)$$

$$\left.\begin{aligned}\dot{P}_{mm} = -\sum_{j=1}^{m-1}&\left\{\gamma_{mj}P_{mm} - i\left(P^{+}_{jm}\,q_{mj} - q^{+}_{mj}\,P_{jm}\right)\right\}\\ +\sum_{r=m+1}^{10}&\left\{\gamma_{rm}P_{rr} - i\left(P^{+}_{mr}\,q_{rm} - q^{+}_{rm}\,P_{mr}\right)\right\}\end{aligned}\right\} \qquad (5.A.2)$$

where the Hamiltonian is given by

$$H = \hbar\sum_{x=1}^{10}\epsilon_x P_{xx} + \hbar\sum_{l\sigma}\omega_l a^{+}_{l\sigma}a_{l\sigma} - \hbar\sum_{y=z+1}^{10}\sum_{z=1}^{9} g_{l\sigma,yz}\times\left(P^{+}_{zy}a_{l\sigma} + a^{+}_{l\sigma}P_{zy}\right) \qquad (5.A.3)$$

We shall be interested in transitions between

a) state $5P_{3/2}$ ($|3\rangle$) and 4S ($|1\rangle$), ground state, and

b) state 6S ($|10\rangle$) and $5P_{3/2}$ ($|3\rangle$)

when two driving fields of arbitrary strength are present. One field E_S couples levels 4S and $4P_{3/2}$, i.e. levels 1 and 3, and the other E_L levels $4P_{3/2}$ and 6S, i.e. levels 3 and 10. The former is supplied by SRS radiation and the latter by ruby laser radiation, since it is known that such radiations are in approximate resonance with the corresponding atomic transitions. The tuning conditions are varied experimentally according to the effect one wants to observe. Field E_L, of frequency ω_L, is supplied by giant pulse ruby laser radiation but its intensity is reduced by 10X in order to inhibit other multiphoton processes in which we are not interested[10],[11]. Field E_S, of frequency ω_S, is supplied by stimulated Raman radiation (SRS) of FNB which is 1345 cm^{-1} down shifted with respect to ω_L[10]. Other molecular liquids can be used as level $4P_{3/2}$ (13) is populated even for incident radiation 30 cm^{-1} off resonance[11] though Yatsiu, Rokni and Barak cite NB (nitrobenzene) for enhanced 2-photon emission and BMN, β-methyl napthalene, for 3 photon Raman effect. According to refs. (10) and (11) $\omega_S = 1/\hbar(E_3 - E_1)$ for a ruby rod at 34.5°C i.e. exact resonance occurs at this temperature.

According ref. (10) resonant absorption of ω_S excites $|m| = \frac{1}{2}$ substate of $4P_{3/2}$ (13) exclusively and thus auxillary Stokes radiation, ω_S, "aligns" excited $4P_{3/2}$ state since other substates of $4P_{3/2}$ are unpopulated and this results in 3 types of resonant Raman scattering of from this initial state $13>$ of which the transition terminating in $5P_{3/2}$ ($18>$) is the one we consider. According to ref. (10) and (3) when transition from $13>$ to $18>$ is by resonant Raman scattering I.R. emission at 3.66 μ is observed, these transitions then result in new atomic levels being filled up which then start to participate in second generation stimulated transitions of which we consider that from level $5P_{3/2}$ ($18>$) to $4S$ ($11>$). The $6S_{1/2}$ state serves as a near resonant intermediate state in the above transitions. In fact when the aforementioned two fields are present four different near-resonant multiphoton processes have been observed in potassium vapour. They are

i) stimulated 2-photon Raman effect

ii) 4-photon parametric coupling

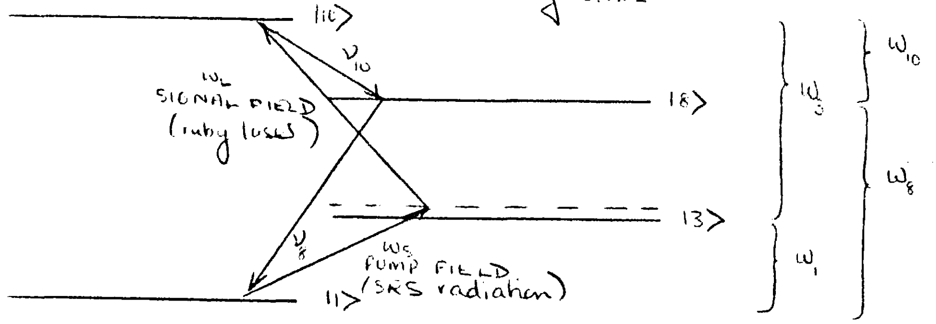
iii) enhanced 2-photon emission

(iv) 3-photon Raman scattering.

What we will calculate is the overall effect of all possible transitions between certain levels by assuming the initial state to be unknown.

The main 4 levels with which we are concerned are illustrated below although we shall take into account all 10 levels in our calculations.

Fig. 5.A.1



We shall define frequencies

$$\omega_3 = \epsilon_{10} - \epsilon_3$$

$$\omega_1 = \epsilon_3 - \epsilon_1$$

$$\omega_{10} = \epsilon_{10} - \epsilon_8$$

$$\omega_8 = \epsilon_8 - \epsilon_1$$

Since we are only considering dipole transitions it is necessary that the change in the orbital momentum quantum number l be such that

$$\Delta l = \pm 1$$

Thus a dipole moment only exists between levels

S and P

and P and D

and NOT between levels

$$\left.\begin{array}{l}\text{S and S}\\ \text{P and P}\\ \text{D and D}\end{array}\right\}\Delta l = 0$$

and S and D $\}\Delta l = \pm 2$, since for level S, $l = 0$; P, $l = 1$; D, $l = 2$.

Hence the dipole matrix elements are as follows

$$r_{SP} = r_{PS} = r_{PD} = r_{DP} \quad \text{are} \neq 0$$

and $$r_{SS} = r_{PP} = r_{DD} = r_{SD} = 0$$

Now since q's, γ's and Ω's all depend on r's through the following relations:-

$$q_{ij} = \sum_{l\sigma} \left(\frac{2\pi\omega_l}{\hbar V}{}_{ij}\right)^{1/2} \hat{e}_{l\sigma} \cdot \mathbf{p}_{ij}\, a_{l\sigma}(0)\, e^{-i\omega_l t}$$

$$\gamma_{ij} = 4/3\hbar \;\; \epsilon_{ij}^{3}/c^{3} \;\; p_{ij}^{2}$$

$$\delta_{ij} = \gamma_{ij} 1/\epsilon_{ij}^{3} \;\; P\int_0^\infty \frac{d\omega}{2\pi} \frac{\omega^3}{\omega - \epsilon_{ij}}$$

we know that zero components of x are given by the following equations, where x = q, ,

$$\left.\begin{array}{l}
x_{1,4} = x_{1,5} = x_{1,6} = x_{1,9} = x_{1,10} = 0 \\
x_{2,3} = x_{2,7} = x_{2,8} = 0 \\
x_{3,7} = x_{3,8} = 0 \\
x_{4,5} = x_{4,6} = x_{4,9} = x_{4,10} = 0 \\
x_{5,6} = x_{5,9} = x_{5,10} = 0 \\
x_{6,9} = x_{6,10} = 0 \\
x_{7,8} = 0 \\
x_{9,10} = 0
\end{array}\right\} \qquad (5.A.4)$$

These equations are also true for reversed subscripts and in the case of q for the hermitian conjugates, since p's are real.

B. Power Spectra

From equations (5.A.1) and (5.A.2) it is possible to derive 100 Heisenberg equations of motion for the transition operators but, in order to calculate the power spectra or spectral densities of the scattered light for (a) transitions between states $|8>$ and $|1>$ and (b) between states $|10>$ and $|8>$, we have seen that it is necessary to use only 3 of these equations. These are the equations for transition operators P_{38}, P_{18} and $P_{10,8}$ which are given below, substitutions (5.A.4) having already been performed.

$$\left.\begin{aligned}\dot{P}_{10,8} = & -\left[\tfrac{1}{2}(\gamma_8+\gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\right] P_{10,8} \\ & + i\left(P_{10,1}\, q_{81} + P_{10,4}\, q_{84} + P_{10,5}\, q_{85} + P_{10,6}\, q_{86}\right) \\ & - i\left(q^{+}_{10,2}\, P_{28} + q^{+}_{10,3}\, P_{38} + q^{+}_{10,7}\, P_{78} + q^{+}_{10,8}\, P_{88}\right) \\ & + i\left(q^{+}_{98}\, P_{10,9} + q^{+}_{10,8}\, P_{10,10}\right)\end{aligned}\right\} \quad (5.B.1)$$

$$\left.\begin{aligned}\dot{P}_{3,8} = & -\left[\tfrac{1}{2}(\gamma_8+\gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\right] P_{38} \\ & - i\left(P_{48}\, q_{43} + P_{58}\, q_{53} + P_{68}\, q_{63} + P_{98}\, q_{93} + P_{10,8}\, q_{10,3}\right) \\ & + i\left(P_{31}\, q_{81} + P_{34}\, q_{84} + P_{35}\, q_{85} + P_{36}\, q_{86}\right) \\ & - i\left(q^{+}_{31}\, P_{18}\right) \\ & + i\left(q^{+}_{98}\, P_{39} + q^{+}_{10,8}\, P_{3,10}\right)\end{aligned}\right\} \quad (5.B.2)$$

$$\left.\begin{aligned}\dot{P}_{1,8} = & -\left[\tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\right] P_{18} \\ & - i\left(P_{28}\, q_{21} + P_{38}\, q_{31} + P_{78}\, q_{71} + P_{88}\, q_{81}\right) \\ & + i\left(P_{11}\, q_{81} + P_{14}\, q_{84} + P_{15}\, q_{85} + P_{16}\, q_{86}\right) \\ & + \left(q^{+}_{98}\, P_{19} + q^{+}_{10,8}\, P_{1,10}\right)\end{aligned}\right\} \quad (5.B.3)$$

where
$$\left.\begin{aligned} & \gamma_8 = \gamma_{81} + \gamma_{84} + \gamma_{85} + \gamma_{86} \qquad \Omega_8 = \Omega_{81} + \Omega_{84} + \Omega_{85} + \Omega_{86} \\ & \gamma_{10} = \gamma_{10,2} + \gamma_{10,3} + \gamma_{10,7} + \gamma_{10,8} \qquad \Omega_{10} = \Omega_{10,2} + \Omega_{10,3} + \Omega_{10,7} + \Omega_{10,8} \\ & \epsilon_{8,10} = -\omega_{10} \\ & \epsilon_{8,3} = \omega_8 - \omega_1 \\ & \epsilon_{8,1} = \omega_8 \end{aligned}\right\} \quad (5.B.4)$$

Levels 1 and 3, 3 and 10 are connected by monochromatic radiation of arbitrary intensity and this can be considered in two ways:- either (i) by considering driving fields as additional classical terms

in the Hamiltonian and thus considering the initial photon state to be a zero photon state as already explained. This has been done in Mollow's papers(7) & (9) and is reasonable for intense fields which can be treated classically or (ii) by including driving fields in the rotation and having the initial photon state as $|\alpha_{10}, \alpha_{3}\rangle$ using Glauber's notation (48)(49),(50).

We shall see that both methods give equivalent results although the second is preferable as it is completely quantum mechanical and does not require the addition of any extra terms.

Method (i) (based on Mollow's formalism; driving field introduced classically)

In Chapter IV, Section A, the driving field was considered to couple all levels and an additional term (4.A.21) was added to the Hamiltonian. We are now concerned with only two classical fields $\underline{E}_S(t)$ and $\underline{E}_L(t)$ coupling levels 1 and 3 and 3 and 10 respectively where

$$\left.\begin{aligned} \underline{E}_S(t) &= \hat{e}_{oS}\left\{\mathcal{E}_S(t) + \mathcal{E}_S^*(t)\right\} \\ \underline{E}_L(t) &= \hat{e}_{oL}\left\{\mathcal{E}_L(t) + \mathcal{E}_L^*(t)\right\} \end{aligned}\right\} \quad (5.B.5)$$

Hence the additional term, instead of being

$$H_{1D}(t) = -\hbar \sum_{y=z+1}^{10} \sum_{z=1}^{9} \left\{ P_{yz}(t)\, \lambda_{yz}\, \mathcal{E}_D(t) + \lambda_{zy}\, \mathcal{E}_D^*(t)\, P_{zy}(t) \right\} \quad (5.B.6)$$

where $\lambda_{yz} = \dfrac{\underline{p}_{yz}\cdot \hat{e}_{oD}}{\hbar} = \lambda_{zy}$

and $\underline{E}_D(t) = \hat{e}_{oD}\left\{\mathcal{E}_D(t) + \mathcal{E}_D^*(t)\right\}$

becomes

$$\left.\begin{aligned} H_{1D}(t) = &-\hbar\left\{P_{31}\lambda_{31}\mathcal{E}_S(t) + \lambda_{13}\mathcal{E}_S^*(t)\, P_{13}(t)\right\} \\ &-\hbar\left\{P_{10,3}\lambda_{10,3}\mathcal{E}_L(t) + \lambda_{3,10}\mathcal{E}_L^*(t)\, P_{3,10}(t)\right\} \end{aligned}\right\} \quad (5.B.7)$$

$$\left.\begin{aligned} \text{where} \quad \lambda_{31} &= \frac{p_{31}\cdot\hat{e}_{os}}{\hbar} = \lambda_{13} \\ \text{and} \quad \lambda_{10,3} &= \frac{p_{10,3}\cdot\hat{e}_{oL}}{\hbar} = \lambda_{3,10} \end{aligned}\right\} \qquad (5.B.7)$$

N.B. Order of indices on λ's is not important as stated elsewhere since the same applies to p's indices since p's are assumed real and hence λ's are real for real $\hat{e}_{os}$ and $\hat{e}_{oL}$.

We shall assume each field oscillates harmonically so that

$$\begin{aligned} \mathcal{E}_s(t) &= \mathcal{E}_{os}\, e^{-i\omega_s t} \\ \mathcal{E}_L(t) &= \mathcal{E}_{oL}\, e^{-i\omega_L t} \end{aligned} \qquad (5.B.8)$$

and that the frequencies of oscillation and nearly coincide with the resonant frequencies for the corresponding transitions

$$\begin{aligned} \text{i.e.} \quad \omega_s &\doteq \epsilon_{3,1} \\ \text{and} \quad \omega_L &\doteq \epsilon_{10,3} \end{aligned} \qquad (5.B.9)$$

but neither are near to the resonant frequency for transitions between any other pairs of levels.

The extra terms which $H_{I.D.}(t)$ introduces into $\dot{P}_{mn}(t)$ and $\dot{P}_{mm}(t)$ are

$$\text{(5.B.10)}\left\{\begin{aligned} \dot{P}_{mn_D}(t) = &-i\{(P_{3n}\delta_{m1} - P_{m1}\delta_{n3})\lambda_{31}\mathcal{E}_s(t) + \lambda_{13}\mathcal{E}_s^*(t)(P_{1n}\delta_{3m} - P_{m3}\delta_{n1})\} \\ &-i\{(P_{10,n}\delta_{3m} - P_{n3}\delta_{n10})\lambda_{10,3}\mathcal{E}_L(t) + \lambda_{3,10}\mathcal{E}_L^*(t)(P_{3n}\delta_{10,m} - P_{m,10}\delta_{n,3})\} \end{aligned}\right.$$

$$\text{(5.B.11)}\left\{\begin{aligned} \dot{P}_{mm_D}(t) = &-i\{(P_{3m}\delta_{m1} - P_{m1}\delta_{m3})\lambda_{31}\mathcal{E}_s(t) + \lambda_{13}\mathcal{E}_s^*(t)(P_{1m}\delta_{3m} - P_{m3}\delta_{m1})\} \\ &-i\{(P_{10,m}\delta_{m3} - P_{m3}\delta_{m,10})\lambda_{10,3}\mathcal{E}_L(t) + \lambda_{3,10}\mathcal{E}_L^*(t)(P_{3m}\delta_{10,m} - P_{m,10}\delta_{m,3})\} \end{aligned}\right.$$

Hence equations (5.B.1), (5.B.2), (5.B.3) become:

$$\left.\begin{aligned}\dot{P}_{3,8} = & -\left\{\tfrac{1}{2}(\gamma_8+\gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\right\} P_{38} \\ & -i\left\{P_{48}\, q_{43} + P_{58}\, q_{53} + P_{68}\, q_{63} + P_{98}\, q_{93} + P_{10,8}\left(q_{10,3} + \lambda_{10,3}\,\mathcal{E}_L(t)\right)\right\} \\ & +i\left\{P_{31}\, q_{81} + P_{34}\, q_{84} + P_{35}\, q_{85} + P_{36}\, q_{86}\right\} \\ & -i\left(q_{31}^{+} + \lambda_{13}\,\mathcal{E}_S^{-}(t)\right) P_{18} \\ & +i\left\{q_{98}^{+}\, P_{39} + q_{10,8}^{+}\, P_{3,10}\right\}\end{aligned}\right\} \quad (5.B.12)$$

$$\begin{aligned}\dot{P}_{1,8} = & -\left\{\tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\right\} P_{18} \\ & -i\left\{P_{28}\, q_{21} + P_{38}\left(q_{31} + \lambda_{31}\,\mathcal{E}_S(t)\right) + P_{78}\, q_{71} + P_{88}\, q_{81}\right\} \\ & +i\left\{P_{11}\, q_{81} + P_{14}\, q_{84} + P_{15}\, q_{85} + P_{16}\, q_{86}\right\} \\ & +i\left\{q_{98}^{+}\, P_{19} + q_{10,8}^{+}\, P_{1,10}\right\}\end{aligned} \qquad (5.B.13)$$

$$\left.\begin{aligned}\dot{P}_{10,8} = & -\left\{\tfrac{1}{2}(\gamma_8+\gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\right\} P_{10,8} \\ & +i\left\{P_{10,1}\, q_{81} + P_{10,4}\, q_{84} + P_{10,5}\, q_{85} + P_{10,6}\, q_{86}\right\} \\ & -i\left\{q_{10,2}^{+}\, P_{28} + \left(q_{10,3}^{+} + \lambda_{3,10}\,\mathcal{E}_L^{-}(t)\right) P_{38} + q_{10,7}^{+}\, P_{78} + q_{10,8}^{+}\, P_{88}\right\} \\ & +i\left\{q_{98}^{+}\, P_{10,9} + q_{10,8}^{+}\, P_{10,10}\right\}\end{aligned}\right\} \quad (5.B.14)$$

Multiplying equations (5.B.12) - (5.B.14) from left and right by ${}_{rad}\langle 0|$ and $|0\rangle_{rad}$ respectively and calling

$$\left.\begin{aligned} X &= {}_{rad}\langle 0|\, P_{38}\, |0\rangle_{rad} \\ Y &= {}_{rad}\langle 0|\, P_{18}\, |0\rangle_{rad} \\ Z &= {}_{rad}\langle 0|\, P_{10,8}\, |0\rangle_{rad} \end{aligned}\right\} \quad (5.B.15)$$

we obtain the following equations

$$\left.\begin{aligned}\dot{X}(t) = & -\left\{ \tfrac{1}{2}(\gamma_8 + \gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\right\} X(t) \\ & - i\lambda_{10,3}\mathcal{E}_L(t) Z(t) \\ & - i\lambda_{13}\mathcal{E}_s^*(t) Y(t)\end{aligned}\right\} \quad (5.B.16)$$

$$\left.\begin{aligned}\dot{Y}(t) = & -\left\{ \tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\right\} Y(t) \\ & - i\lambda_{31}\mathcal{E}_s(t) X(t)\end{aligned}\right\} \quad (5.B.17)$$

$$\left.\begin{aligned}\dot{Z}(t) = & -\left\{ \tfrac{1}{2}(\gamma_8 + \gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\right\} Z(t) \\ & - i\lambda_{3,10}\mathcal{E}_L^*(t) X(t)\end{aligned}\right\} \quad (5.B.18)$$

In order to obtain equations for the matrix elements of the reduced density operators we recall that

$$\left.\begin{aligned}\rho^{(s)}_{8,3}(t) &= \sum_s \langle s | X(t)\rho^{(s)}(0) | s\rangle \\ \rho^{(s)}_{8,1}(t) &= \sum_s \langle s | Y(t)\rho^{(s)}(0) | s\rangle \\ \rho^{(s)}_{8,10}(t) &= \sum_s \langle s | Z(t)\rho^{(s)}(0) | s\rangle\end{aligned}\right\} \quad (5.B.19)$$

Hence multiplying equations (5.B.16) - (18) from the R.H.S. by $\rho^{(s)}(0)$ and taking Tr_s we obtain

$$\left.\begin{aligned}\dot{\rho}^{(s)}_{8,3}(t) = & -\left\{ \tfrac{1}{2}(\gamma_8 + \gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\right\} \rho^{(s)}_{8,3}(t) \\ & - i\lambda_{10,3}\mathcal{E}_L(t)\rho^{(s)}_{8,10}(t) \\ & - i\lambda_{13}\mathcal{E}_s^*(t)\rho^{(s)}_{8,1}(t)\end{aligned}\right\} \quad (5.B.20)$$

$$\left.\begin{aligned}\dot{\rho}^{(s)}_{8,1}(t) = & -\left\{ \tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\right\} \rho^{(s)}_{8,1}(t) \\ & - i\lambda_{31}\mathcal{E}_s(t)\rho^{(s)}_{8,3}(t)\end{aligned}\right\} \quad (5.B.21)$$

$$\left.\begin{aligned}\dot{\rho}^{(5)}_{8,10}(t) = &-\left\{\tfrac{1}{2}(\gamma_8+\gamma_{10}) + i\,(\epsilon_{8,10}-\Omega_8+\Omega_{10})\right\}\rho^{(5)}_{8,10}(t)\\ &- i\,\lambda_{3,10}\,\mathcal{E}_L^*(t)\,\rho^{(5)}_{8,3}(t)\end{aligned}\right\}\quad (5.B.22)$$

Method (ii) (based on Glauber's formalism; driving field introduced in a completely quantum mechanical way)

We shall now take into account the monochromatic radiation fields coupling levels 1 and 3, 3 and 10 by assuming that the driving fields are already included in the notation so that all we have to do is to consider the initial photon state $|i_{ph}\rangle = |\alpha_{10},\alpha_3\rangle$ (5.B.23) instead of a zero photon state as in Method (i).

The reasoning behind this method depends on the R.W.A. We will recall that in neglecting certain hermitian terms in the interaction Hamiltonian we assume $g_{l\sigma,yz} \ll \epsilon_{yz} \doteq \omega_l$ (5.B.24) so that ω_l is not completely general. In fact we have been assigned extra subscripts to it for this reason, i.e. $\omega_l \longrightarrow \omega_{lyz}$ in the R.W.A. Hence each pair of terms in the interaction Hamiltonian is summed over all l but bearing in mind the ω_l has only a small variation in each case, e.g.

$$\left.\begin{aligned}H_I = &-\hbar\sum_{l,\sigma} g_{l\sigma,21}(P_{12}^+ a_{l\sigma} + a_{l\sigma}^+ P_{12}) \quad \text{where } \omega_l \doteq \epsilon_{21}\\ &-\hbar\sum_{l,\sigma} g_{l\sigma,31}(P_{13}^+ a_{l\sigma} + a_{l\sigma}^+ P_{13}) \quad \text{where } \omega_l \doteq \epsilon_{31}\\ &\quad \cdots\cdots\end{aligned}\right\}\quad (5.B.25)$$

We could thus assign subscripts to the boson operators to indicate this so that

$$\left.\begin{aligned}H_I = &-\hbar\sum_{l,\sigma} g_{l\sigma,21}(P_{12}^+ a_{l\sigma,21} + a_{l\sigma,21}^+ P_{12})\\ &-\hbar\sum_{l,\sigma} g_{l\sigma,31}(P_{13}^+ a_{l\sigma,31} + a_{l\sigma,31}^+ P_{13})\\ &\quad \cdots\end{aligned}\right\}\quad (5.B.26)$$

We shall be using Glauber's formalism(48) which says (see eq. (2.21)) that individual coherent states, $|\alpha_{k\sigma}\rangle$, of the field obey the relations

$$a_{k\sigma}|\alpha_{k\sigma}\rangle_{k\sigma} = \alpha_{k\sigma}|\alpha_{k\sigma}\rangle_{k\sigma}$$

where operators and states are evaluated at the same time and $\alpha_{k\sigma}$ is complex. In fact we shall consider only a single mode of each of the two fields. $\alpha_{k\sigma}$ is the number of photons in mode $k\sigma$ and so for the zero photon state or vacuum state $\alpha_{k\sigma} = 0$ and the corresponding coherent state is the unique ground state of the field, i.e. the state $|\alpha_{k\sigma}\rangle$ for $\alpha_{k\sigma} = 0$.

We shall assume that initially $|\alpha_{k\sigma}\rangle = |\alpha_{10},\alpha_{3}\rangle$ where $|\alpha_{10}\rangle$ is the state of the laser field initially and $|\alpha_{3}\rangle$ is the initial state of the SRS field. In order to find the value of terms

$$q_{nm}|1_{ph}\rangle$$

we expand q_{nm} and $|1_{ph}\rangle$ so that

$$q_{nm}|1_{ph}\rangle = \sum_{k\sigma} g_{k\sigma,nm}\, e^{-i\omega_k t}\, a_{k\sigma,nm}(0)|\alpha_{10},\alpha_{3}\rangle \qquad (5.B.27)$$

where $n > m$ and $|\alpha_{10},\alpha_{3}\rangle$ is the initial photon state and so is measured at time $t' = 0$ as is $a_{k\sigma}(0)$.

The value of $a_{k\sigma,nm}(0)|\alpha_{10},\alpha_{3}\rangle$ thus needs to be known and it is obvious that for certain n,m it is zero, viz. $a_{k\sigma,nm}|\alpha_{10},\alpha_{3}\rangle$ for $n > m$ is zero for all values of n,m except when

$$n=10,\ m=3 \quad \text{and} \quad n=3,\ m=1 \qquad (5.B.28)$$

In fact

$$\left.\begin{aligned} q_{10,3}|1_{ph}\rangle &= \sum_{k\sigma} g_{k\sigma,10,3}\, e^{-i\omega_k t}\, a_{k\sigma,10,3}(0)|\alpha_{10},\alpha_{3}\rangle \\ &= g_{\omega_L,10,3}\, e^{-i\omega_L t}\, \alpha_{10}\,|\alpha_{10},\alpha_{3}\rangle \\ &= \sqrt{\frac{2\pi\omega_L}{\hbar V}}\,(\hat{e}_{0L}\cdot \mathbf{f}_{10,3})\, e^{-i\omega_L t}\, \alpha_{10}\,|\alpha_{10},\alpha_{3}\rangle \end{aligned}\right\} \qquad (5.B.29)$$

$\langle 1_{ph}|q_{10,3}|1_{ph}\rangle = g_{\omega_L,10,3}\, e^{-i\omega_L t}\, \alpha_{10}$ since $\langle \alpha_{10},\alpha_{3}|\alpha_{3},\alpha_{10}\rangle = 1$!

Similarly

$$\begin{aligned}
\langle i_{ph}| q^{+}_{10,3} | i_{ph}\rangle &= g_{\omega_L,10,3}\, e^{i\omega_L t}\, \alpha^{*}_{10} \\
\langle i_{ph}| q_{3,1} | i_{ph}\rangle &= g_{\omega_s,3,1}\, e^{-i\omega_s t}\, \alpha_{3} \\
\langle i_{ph}| q^{+}_{3,1} | i_{ph}\rangle &= g_{\omega_s,3,1}\, e^{i\omega_s t}\, \alpha^{*}_{3}
\end{aligned} \qquad (5.B.30)$$

In this method, we let

$$\begin{aligned}
&\tfrac{1}{2}G_L = g_{\omega_L,10,3}\,|\alpha_{10}|, \quad \tfrac{1}{2}G'_L = g_{\omega_L,10,3}\,\alpha_{10}, \quad \tfrac{1}{2}G'^{*}_L = g_{\omega_L,10,3}\,\alpha^{*}_{10} \\
&\tfrac{1}{2}G_s = g_{\omega_s,3,1}\,|\alpha_3|, \quad \tfrac{1}{2}G'_s = g_{\omega_s,3,1}\,\alpha_3, \quad \tfrac{1}{2}G'^{*}_s = g_{\omega_s,3,1}\,\alpha^{*}_3
\end{aligned} \qquad (5.B.31)$$

$$\begin{aligned}
\therefore \langle i_{ph}| q_{10,3} | i_{ph}\rangle &= \tfrac{1}{2}G'_L\, e^{-i\omega_L t}, \quad \langle i_{ph}| q^{+}_{10,3} | i_{ph}\rangle = \tfrac{1}{2}G'^{*}_L\, e^{i\omega_L t} \\
\langle i_{ph}| q_{3,1} | i_{ph}\rangle &= \tfrac{1}{2}G'_s\, e^{-i\omega_s t}, \quad \langle i_{ph}| q^{+}_{3,1} | i_{ph}\rangle = \tfrac{1}{2}G'^{*}_s\, e^{i\omega_s t}
\end{aligned} \qquad (5.B.32)$$

I.E. now

$$\begin{aligned}
\tfrac{1}{4}G_L^2 = g^2_{\omega_L,10,3}|\alpha_{10}|^2 \text{ and } \tfrac{1}{4}G_s^2 &= g^2_{\omega_s,3,1}|\alpha_3|^2 \\
= \left(\frac{2\pi\omega_L}{\hbar V}\right)(\hat{e}_{oL}\cdot p_{10,3})^2|\alpha_{10}|^2 &= \left(\frac{2\pi\omega_s}{\hbar V}\right)(\hat{e}_{os}\cdot p_{31})^2|\alpha_3|^2
\end{aligned} \qquad (5.B.33)$$

instead of

$$\begin{aligned}
\tfrac{1}{4}G_{10}^2 = (\lambda_{10,3}\,\mathcal{E}_{oL})^2 \quad &\text{and} \quad \tfrac{1}{4}G_3^2 = (\lambda_{31}\,\mathcal{E}_{os})^2 \\
= \left(\frac{p_{10,3}\cdot\hat{e}_{oL}\,\mathcal{E}_{oL}}{\hbar}\right)^2 \quad &\quad = \left(\frac{p_{31}\cdot\hat{e}_{os}\,\mathcal{E}_{os}}{\hbar}\right)^2
\end{aligned} \qquad (5.B.34)$$

Multiplying equations (5.B.1)-(5) from left and right by $\langle i_{ph}|$ and $|i_{ph}\rangle$ respectively we obtain:-

$$\begin{aligned}
\langle i_{ph}|\dot{P}_{10,8}|i_{ph}\rangle = &-\left\{\tfrac{1}{2}(\gamma_8+\gamma_{10})+i(\epsilon_{8,10}-\Omega_8+\Omega_{10})\right\}\langle i_{ph}|P_{10,8}|i_{ph}\rangle \\
&- i\langle i_{ph}|q^{+}_{10,3}\,P_{38}\,|i_{ph}\rangle
\end{aligned} \qquad (5.B.35)$$

$$\begin{aligned}
\langle i_{ph}|\dot{P}_{38}|i_{ph}\rangle = &-\left\{\tfrac{1}{2}(\gamma_8+\gamma_{31})+i(\epsilon_{8,3}-\Omega_8+\Omega_{31})\right\}\langle i_{ph}|P_{3,8}|i_{ph}\rangle \\
&- i\langle i_{ph}|P_{10,8}\,q_{10,3}\,|i_{ph}\rangle \\
&- i\langle i_{ph}|q^{+}_{31}\,P_{18}\,|i_{ph}\rangle
\end{aligned} \qquad (5.B.36)$$

$$\begin{aligned}
\langle i_{ph}|\dot{P}_{18}|i_{ph}\rangle = &-\left\{\tfrac{1}{2}\gamma_8+i(\epsilon_{81}-\Omega_8)\right\}\langle i_{ph}|P_{18}|i_{ph}\rangle \\
&- i\langle i_{ph}|P_{38}\,q_{31}\,|i_{ph}\rangle
\end{aligned} \qquad (5.B.37)$$

Let $X_a = \langle i_{ph} | P_{38} | i_{ph} \rangle$

$y_a = \langle i_{ph} | P_{18} | i_{ph} \rangle$

$Z_a = \langle i_{ph} | P_{10,8} | i_{ph} \rangle$

hence

$$\left.\begin{aligned} \dot{X}_a &= -\{ \tfrac{1}{2}(\gamma_8 + \gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\} X_a \\ &\quad - i\tfrac{1}{2} G_L' e^{-i\omega_L t} Z_a \\ &\quad - i\tfrac{1}{2} G_s'^{*} e^{i\omega_s t} y_a \end{aligned}\right\} \qquad (5.B.38)$$

$$\left.\begin{aligned} \dot{Y}_a &= -\{ \tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\} y_a \\ &\quad - i\tfrac{1}{2} G_s' e^{-i\omega_s t} X_a \end{aligned}\right\} \qquad (5.B.39)$$

$$\left.\begin{aligned} \dot{Z}_a &= -\{ \tfrac{1}{2}(\gamma_8 + \gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\} Z_a \\ &\quad - i\tfrac{1}{2} G_L'^{*} e^{i\omega_L t} X_a \end{aligned}\right\} \qquad (5.B.40)$$

Hence, recalling that

$$\rho_{83}^{(s)}(t) = \sum_s \langle s | X_a(t) \rho^{(s)}(0) | s \rangle \qquad (5.B.41)$$

we obtain equations for the reduced density matrix operators

$$\left.\begin{aligned} \dot{\rho}_{83}^{(s)}(t) &= -\{ \tfrac{1}{2}(\gamma_8 + \gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\} \rho_{83}^{(s)}(t) \\ &\quad - i\tfrac{1}{2} G_L' e^{-i\omega_L t} \rho_{8,10}^{(s)}(t) \\ &\quad - i\tfrac{1}{2} G_s'^{*} e^{i\omega_s t} \rho_{81}^{(s)}(t) \end{aligned}\right\} \qquad (5.B.42)$$

$$\left.\begin{aligned} \dot{\rho}_{81}^{(s)}(t) &= -\{ \tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\} \rho_{81}^{(s)}(t) \\ &\quad - i\tfrac{1}{2} G_s' e^{-i\omega_s t} \rho_{83}^{(s)}(t) \end{aligned}\right\} \qquad (5.B.43)$$

$$\left.\begin{aligned} \dot{\rho}_{8,10}^{(s)}(t) &= -\{ \tfrac{1}{2}(\gamma_8 + \gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\} \rho_{8,10}^{(s)}(t) \\ &\quad - i\tfrac{1}{2} G_L'^{*} e^{i\omega_L t} \rho_{83}^{(s)}(t) \end{aligned}\right\} \qquad (5.B.44)$$

Comparing these equations with equations (5.B.20-22) of method (i), when $\mathcal{E}_s(t)$ and $\mathcal{E}_L(t)$ are written as $\mathcal{E}_{os} e^{-i\omega_s t}$, $\mathcal{E}_{oL} e^{-i\omega_L t}$ respectively, we see that the equations are equivalent if

$$\left.\begin{array}{llllll}
\tfrac{1}{2}G'_L \longrightarrow \lambda_{10,3}\mathcal{E}_{0L} & \text{i.e.} & g_{\omega_L,10,3}\,\alpha_{10} \longrightarrow \lambda_{10,3}\mathcal{E}_{0L} & \text{i.e.} & \alpha_{10} \longrightarrow \sqrt{\dfrac{V}{2\pi\hbar\omega_L}}\,\mathcal{E}_{0L} \\
\tfrac{1}{2}G'^{*}_S \longrightarrow \lambda_{13}\mathcal{E}_{0S} & & g_{\omega_S,31}\,\alpha^{*}_3 \longrightarrow \lambda_{13}\mathcal{E}_{0S} & & \alpha^{*}_3 \longrightarrow \sqrt{\dfrac{V}{2\pi\hbar\omega_S}}\,\mathcal{E}_{0S} \\
\tfrac{1}{2}G'_S \longrightarrow \lambda_{31}\mathcal{E}_{0S} & & g_{\omega_S,31}\,\alpha_3 \longrightarrow \lambda_{31}\mathcal{E}_{0S} & & \alpha_3 \longrightarrow \sqrt{\dfrac{V}{2\pi\hbar\omega_S}}\,\mathcal{E}_{0S} \\
\tfrac{1}{2}G'^{*}_L \longrightarrow \lambda_{3,10}\mathcal{E}_{0L} & & g_{\omega_L,10,3}\,\alpha^{*}_{10} \longrightarrow \lambda_{3,10}\mathcal{E}_{0L} & & \alpha^{*}_{10} \longrightarrow \sqrt{\dfrac{V}{2\pi\hbar\omega_L}}\,\mathcal{E}_{0L}
\end{array}\right\} \quad (5.B.45)$$

i.e. $\alpha_{10}, \alpha^{*}_{10} \propto (\mathcal{E}_{0L}/\sqrt{\omega_L})$ and $\alpha_3, \alpha^{*}_3 \propto (\mathcal{E}_{0S}/\sqrt{\omega_S})$ i.e. α_3 and α_{10} are REAL in method (1).

Bearing these transformations in mind we can use either method and obtain results in the same form.

According to equation (4.C.8) for transitions

a) between levels 8 and 1, we need to know

$$\langle P^{+}_{18}(t)\,P_{18}(t')\rangle = \sum_{y''=1}^{10} U_{81,y''1}(\tau,t')\,\rho^{(s)}_{y''8}(t') \qquad (5.B.46)$$

and for transitions

b) between levels 10 and 8, we need to know

$$\langle P^{+}_{8,10}(t)\,P_{8,10}(t')\rangle = \sum_{y''=1}^{10} U_{10,8;y''8}(\tau,t')\,\rho^{(s)}_{y'',10}(t') \qquad (5.B.47)$$

So for case (a) we need to solve equations (5.B.20)-(22) for $\rho^{(s)}_{81}(t)$ in order to find $U_{81,11}$, $U_{81,21}$ . . . . . $U_{81,10,1}$ and for case (b) we need to solve for $\rho^{(s)}_{10,8}(t)$ in order to find functions $U_{10,8;18}$, $U_{108;28}$, . . . $U_{10,8;10,8}$ where all U's depend on τ and t'. In fact it is obvious that there will only be three U's in each case since equations (5.B.20)-(22) are independent. They can be solved by taking Laplace transforms, assuming the initial time to be t'.

First, on substituting for $\mathcal{E}_s(t)$ and $\mathcal{E}_L(t)$ explicitly, we obtain

$$\left.\begin{aligned}
\dot{\rho}^{(s)}_{83}(t) = &-\left\{\tfrac{1}{2}(\gamma_8+\gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\right\}\rho^{(s)}_{83}(t) \\
&- i\lambda_{10,3}\mathcal{E}_{0L}\,e^{-i\omega_L t}\,\rho^{(s)}_{8,10}(t) \\
&- i\lambda_{13}\,\mathcal{E}_{0S}\,e^{i\omega_S t}\,\rho^{(s)}_{81}(t)
\end{aligned}\right\} \quad (5.B.48)$$

$$\left.\begin{aligned}\dot{\rho}^{(s)}_{81}(t) = & -\{\tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\}\rho^{(s)}_{81}(t) \\ & - i\lambda_{31}\mathcal{E}_{0S}e^{-i\omega_S t}\rho^{(s)}_{83}(t)\end{aligned}\right\} \quad (5.B.49)$$

$$\left.\begin{aligned}\dot{\rho}^{(s)}_{8,10}(t) = & -\{\tfrac{1}{2}(\gamma_8+\gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\}\rho^{(s)}_{8,10}(t) \\ & - i\lambda_{3,10}\mathcal{E}_{0L}e^{i\omega_L t}\rho^{(s)}_{83}(t)\end{aligned}\right\} \quad (5.B.50)$$

and in order to have only three distinct functions of t so that we may solve the three equations easily we multiply equation (5.B.49) by $e^{i\omega_S t}$ and (5.B.50) by $e^{-i\omega_L t}$.

Then

$$\left.\begin{aligned}\dot{\rho}^{(s)}_{83}(t) = & -\{\tfrac{1}{2}(\gamma_8+\gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\}\rho^{(s)}_{83}(t) \\ & - i\lambda_{103}\mathcal{E}_{0L}e^{-i\omega_L t}\rho^{(s)}_{8,10}(t) \\ & - i\lambda_{13}\mathcal{E}_{0S}e^{i\omega_S t}\rho^{(s)}_{81}(t)\end{aligned}\right\} \quad (5.B.51)$$

$$\left.\begin{aligned}e^{i\omega_S t}\dot{\rho}^{(s)}_{81}(t) = & -\{\tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \Omega_8)\}\rho^{(s)}_{81}(t) \\ & - i\lambda_{31}\mathcal{E}_{0S}\rho^{(s)}_{83}(t)\end{aligned}\right\} \quad (5.B.52)$$

$$\left.\begin{aligned}e^{-i\omega_L t}\dot{\rho}^{(s)}_{8,10}(t) = & -\{\tfrac{1}{2}(\gamma_8+\gamma_{10}) + i(\epsilon_{8,10} - \Omega_8 + \Omega_{10})\}e^{-i\omega_L t}\rho^{(s)}_{8,10}(t) \\ & - i\lambda_{3,10}\mathcal{E}_{0L}\rho^{(s)}_{83}(t)\end{aligned}\right\} \quad (5.B.53)$$

Then letting the three distinct functions of time be represented by x(t), y(t), z(t), where

$$\left.\begin{aligned}x(t) &= \rho^{(s)}_{83}(t) \\ y(t) &= e^{i\omega_S t}\rho^{(s)}_{81}(t) \quad \therefore \dot{y}(t) = i\omega_S y(t) + e^{i\omega_S t}\dot{\rho}^{(s)}_{81}(t) \\ z(t) &= e^{-i\omega_L t}\rho^{(s)}_{8,10}(t) \qquad \dot{z}(t) = -i\omega_L z(t) + e^{-i\omega_L t}\dot{\rho}^{(s)}_{8,10}(t)\end{aligned}\right\} \quad (5.B.54)$$

we have, on substitution

$$\left.\begin{aligned}\dot{x}(t) = & -\{\tfrac{1}{2}(\gamma_8+\gamma_{31}) + i(\epsilon_{83} - \Omega_8 + \Omega_{31})\}x(t) \\ & - i\lambda_{10,3}\mathcal{E}_{0L}z(t) \\ & - i\lambda_{13}\mathcal{E}_{0S}y(t)\end{aligned}\right\} \quad (5.B.55)$$

$$\dot{y}(t) = -\left\{ \tfrac{1}{2}\gamma_8 + i(\epsilon_{81} - \omega_s - \Omega_8) \right\} y(t) - i\lambda_{31}\mathcal{E}_{os}\, x(t) \tag{5.B.56}$$

$$\dot{z}(t) = -\left\{ \tfrac{1}{2}(\gamma_8 + \gamma_{10}) + i(\epsilon_{10} + \omega_L - \Omega_8 + \Omega_{10}) \right\} z(t) - i\lambda_{3,10}\mathcal{E}_{oL}\, x(t) \tag{5.B.57}$$

Taking Laplace transforms of equations (5.B.55-57) we obtain

$$\left\{ s + \tfrac{1}{2}\gamma' + i(\omega_8' - \omega_1') \right\} \hat{x}(s) = -i\lambda_{10,3}\mathcal{E}_{oL}\hat{z}(s) - i\lambda_{13}\mathcal{E}_{os}\hat{y}(s) + e^{-st'}x(t') \tag{5.B.58}$$

$$\left\{ s + \tfrac{1}{2}\gamma_8 + i(\omega_8' - \omega_s) \right\} \hat{y}(s) = -i\lambda_{31}\mathcal{E}_{os}\hat{x}(s) + e^{-st'}y(t') \tag{5.B.59}$$

$$\left\{ s + \tfrac{1}{2}\gamma'' - i(\omega_{10}' - \omega_L) \right\} \hat{z}(s) = -i\lambda_{3,10}\mathcal{E}_{oL}\hat{x}(s) + e^{-st'}z(t') \tag{5.B.60}$$

where

$$\left.\begin{aligned}
\gamma' &= \gamma_8 + \gamma_{31} \\
\gamma'' &= \gamma_8 + \gamma_{10} \\
\epsilon_{81} - \Omega_8 &= \omega_8 - \Omega_8 = \omega_8' \\
\epsilon_{31} - \Omega_{31} &= \omega_1 - \Omega_{31} = \omega_1' \\
\epsilon_{10,8} - (\Omega_{10} - \Omega_8) &= \omega_{10} - (\Omega_{10} - \Omega_8) = \omega_{10}' \\
\epsilon_{83} - (\Omega_8 - \Omega_{31}) &= \left\{ (\epsilon_{81} - \epsilon_{31}) - (\Omega_8 - \Omega_{31}) \right\} = \left\{ (\omega_8 - \omega_1) - (\Omega_8 - \Omega_{31}) \right\} = \omega_8' - \omega_1'
\end{aligned}\right\} \tag{5.B.61}$$

i.e. we have absorbed the frequency shifts into the definitions of ω''s.

(a) Spectral correlation function for transitions between levels $5P_{3/2}$ (8) and $4S$ (1)

In order to solve equations (5.B.58-60) for $\hat{y}(s) = \mathcal{L}\left(e^{i\omega_s t}\rho_{81}^{(s)}(t)\right)$ we substitute for $\hat{z}(s)$ from (5.B.60) in (5.B.58) and substitute the resulting value for $\hat{x}(s)$ in (5.B.59). Hence we obtain:

$$\left.\begin{aligned}
\hat{y}(s) = &-(\lambda_{31}\mathcal{E}_{os})(\lambda_{10,3}\mathcal{E}_{oL}) \frac{1}{F(s)} e^{-st'}z(t') \\
&- i\lambda_{31}\mathcal{E}_{os}\frac{f_2(s)}{F(s)} e^{-st'}x(t') \\
&+ \left\{ \frac{f_1(s)f_2(s) + \tfrac{1}{4}G_{10}^2}{F(s)} \right\} e^{-st'}y(t')
\end{aligned}\right\} \tag{5.B.62}$$

where
$$\left.\begin{aligned} f_1(s) &= s + \tfrac{1}{2}\gamma' + i(\omega_8' - \omega_1') \\ f_2(s) &= s + \tfrac{1}{2}\gamma'' - i(\omega_{10}' - \omega_L) \\ f_3(s) &= s + \tfrac{1}{2}\gamma_8 + i(\omega_8' - \omega_S) \\ F(s) &= f_3(s)\{f_1(s)f_2(s) + \tfrac{1}{4}G_{10}^2\} + \tfrac{1}{4}G_3^2 f_2(s) \\ G_{10} &= 2\lambda_{10,3}\mathcal{E}_{0L} = 2\lambda_{3,10}\mathcal{E}_{0L} = 2\lambda_{10}\mathcal{E}_{0L} \\ \text{and}\quad G_3 &= 2\lambda_{31}\mathcal{E}_{0S} = 2\lambda_{13}\mathcal{E}_{0S} = 2\lambda_3\mathcal{E}_{0S} \end{aligned}\right\} \quad (5.B.63)$$

Now
$$\left.\begin{aligned} \mathcal{L}(e^{ixt}A(t)) &= \int_{t'} dt\, e^{-st} e^{ixt} A(t) \\ &= \int_{t'}^{\infty} dt\, e^{-(s-ix)t} A(t) \\ &= \hat{A}(s-ix) \end{aligned}\right\} \quad (5.B.64)$$

$$\left.\begin{aligned} \therefore \hat{\rho}^{(s)}_{81}(s) &= -(\lambda_{31}\mathcal{E}_{0S})(\lambda_{10,3}\mathcal{E}_{0L})\frac{1}{F(s+i\omega_S)} e^{-(s+i\omega_S)t'} e^{-i\omega_L t'} \rho^{(s)}_{8,10}(t') \\ &- i\lambda_{31}\mathcal{E}_{0S}\frac{f_2(s+i\omega_S)}{F(s+i\omega_S)} e^{-(s+i\omega_S)t'} \rho^{(s)}_{83}(t') \\ &+ \left\{\frac{f_1(s+i\omega_S)f_2(s+i\omega_S) + \tfrac{1}{4}G_{10}^2}{F(s+i\omega_S)}\right\} e^{-(s+i\omega_S)t'} e^{i\omega_S t'} \rho^{(s)}_{81}(t') \end{aligned}\right\} \quad (5.B.65)$$

Hence
$$\hat{\rho}^{(s)}_{81}(s) = \hat{U}_{81,8,10}(s)\rho^{(s)}_{8,10}(t') + \hat{U}_{81,83}(s)\rho^{(s)}_{83}(t') + \hat{U}_{81,81}(s)\rho^{(s)}_{81}(t')$$

where
$$\left.\begin{aligned} \hat{U}_{81,8,10}(s) &= -(\lambda_3\mathcal{E}_{0S})(\lambda_{10}\mathcal{E}_{0L})\frac{1}{F(s+i\omega_S)} e^{-(s+i(\omega_S+\omega_L))t'} \\ \hat{U}_{81,83}(s) &= -i\lambda_{31}\mathcal{E}_{0S}\frac{f_2(s+i\omega_S)}{F(s+i\omega_S)} e^{-(s+i\omega_S)t'} \\ \hat{U}_{81,81}(s) &= \left\{\frac{f_1(s+i\omega_S)f_2(s+i\omega_S) + \tfrac{1}{4}G_{10}^2}{F(s+i\omega_S)}\right\} e^{-st'} \end{aligned}\right\} \quad (6.B.66)$$

Now the quantity we are interested in is the 2-time atomic correlation function

$g_8(t',t'') = \langle P^+_{18}(t')P_{18}(t'')\rangle$ OR $g_8(\tau,t') = \langle P^+_{18}(t')P_{18}(t'+\tau)\rangle$ where $t'' = t' + \tau$

$$\left.\begin{aligned} &\doteq \sum_{y''=1}^{10} \hat{U}_{81,y''1}(\tau,t')\rho^{(s)}_{y''8}(t') \quad \text{under Markoff approximation} \\ &= U_{81,81}(\tau,t')\rho^{(s)}_{88}(t') \quad \text{in the present case since} \end{aligned}\right\} \quad (5.B.67)$$

there will be only one U of the form $U_{81,y''1}$ when $y''=8$

Let

$$\hat{U}_{81,\ldots}(s) = \hat{\psi}_{81,\ldots}(s)\,\epsilon^{-st'}\,\epsilon^{-i(\ldots)t'} \quad [illegible] \tag{5.B.68a}$$

$$\hat{U}_{81,\ldots}(s) = \hat{\psi}_{81,\ldots}(s)\,\epsilon^{-st'}\,\epsilon^{\ldots t'} \quad [illegible] \tag{5.B.68b}$$

$$\hat{U}_{81,\ldots}(s) = \hat{\psi}_{81,\ldots}(s)\,\epsilon^{-st'} \quad [illegible] \tag{5.B.68c}$$

Now if we take the inverse Laplace transform of (5.B.68a) we obtain

$$U_{81,81}(t) = \frac{1}{2\pi i}\oint ds\,\epsilon^{st}\,\hat{U}_{81,81}(s)$$

and if we let $t = t' + \tau$

$$\left.\begin{aligned} U_{81,81}(t'+\tau) &= \frac{1}{2\pi i}\oint ds\,\hat{\psi}_{81,81}(s)\,\epsilon^{-st'}\,\epsilon^{s(t'+\tau)} \\ &= \frac{1}{2\pi i}\oint ds\,\hat{\psi}_{81,81}(s)\,\epsilon^{s\tau} \\ &= \psi_{81,81}(\tau) \end{aligned}\right\} \tag{5.B.69a}$$

Similarly

$$U_{81,83}(t'+\tau) = \psi_{81,83}(\tau)\,\epsilon^{-i\omega_3 t'} \tag{5.B.69b}$$

and $U_{81,8,10}(t'+\tau) = \psi_{81,8,10}(\tau)\,\epsilon^{-i(\omega_\ldots+\omega_\ldots)t'}$ (5.B.69c)

Now $U_{81,81}(\tau,t') = U_{81,81}(t'+\tau)$ since t' and τ always occur as $t' + \tau$

$$\therefore U_{81,81}(\tau,t') = \psi_{81,81}(\tau)$$

$$\therefore \boxed{g_6(\tau,t') = \psi_{81,81}(\tau)\,\rho^{(s)}_{88}(t')} \tag{5.B.70}$$

Thus the atomic correlation function depends on t' only through the term $\rho^{(s)}_{88}(t')$ and not through any factor in $\psi_{81,81}(\tau)$

If we assume the atom is in equilibrium with the field so that $\rho^{(s)}_{8,10}(t')$, $\rho^{(s)}_{83}(t')$, $\rho^{(s)}_{81}(t')$ are given by their asymptotic expressions $\rho^{(s)}_{8,10}(t\to\infty) = \rho^{(s)}_{8,10\infty}$, $\rho^{(s)}_{83}(t\to\infty) = \rho^{(s)}_{83\infty}$, $\rho^{(s)}_{81}(t\to\infty) = \rho^{(s)}_{81\infty}$ with the latter evaluated at time t'

i.e.

$$\left.\begin{aligned} \rho^{(s)}_{8,10}(t') &= \left\{\rho^{(s)}_{8,10\infty}\right\}_{t=t'} \\ \rho^{(s)}_{83}(t') &= \left\{\rho^{(s)}_{83\infty}\right\}_{t=t'} \\ \rho^{(s)}_{\ldots}(t') &= \left\{\rho^{(s)}_{\ldots}\right\}_{t=t'} \end{aligned}\right\} \tag{5.B.71}$$

and similarly for all other initial values of the reduced density matrices.

The atomic correlation function in equation (5.B.70) is then independent of the initial time t' and is given by the relation

$$g_{8}(\tau) = \psi_{s1,s1}(\tau)\{\rho^{(\cdot)}_{\cdot 8_-}\}_{t\,t'} \tag{5.B.72}$$

Taking the Laplace transform of $g_8(\tau)$, we obtain

$$\left.\begin{aligned}\hat{g}_{8}(s) &= \int_0^\infty d\tau\, g(\tau)\epsilon^{-s\tau} = \hat{\psi}_{s1,s1}(s)\{\rho^{(s)}_{s8_-}\}_{t\,t'}\\ &= \left\{\frac{f_1(s+i\omega_s)f_2(s+i\omega_s) + \tfrac{1}{4}G_{10}^2}{F(s+i\omega_s)}\right\}\{\rho^{(s)}_{s8_-}\}_{t\,t'}\end{aligned}\right\} \tag{5.B.73}$$

We now wish to evaluate the spectral correlation function $\tilde{g}_8(\nu)$, defined by the Fourier transform of the atomic correlation function

$$\tilde{g}_8(\nu) = \int_{-\infty}^{\infty} d\tau\, g_8(\tau)\epsilon^{i\nu\tau} \tag{5.B.74}$$

where ν is the scattered frequency.

<u>$\tilde{g}_8(\nu)$, when multiplied by a simple factor $|\varphi(\underline{r})|^2$</u>, as in equation (2.19) of Mollow's paper[9], and equation (3.B.17) of this thesis, <u>gives the energy or power spectrum of the scattered field at any point in space</u>.

i.e. $$I(\nu,\underline{r}) = |\varphi(\underline{r})|^2\,\tilde{g}(\nu) \tag{5.B.75}$$

where in Mollow's units $|\varphi(\underline{r})|^2 = \frac{1}{8\pi^2 r^6}\frac{\omega_0^4}{c^4}\{(\underline{p}\times\underline{r})\times\underline{r}\}^2$ for the 2-level atom of energy separation $\hbar\omega_0$ (see eq. (2.12) of ref. 9), where in the previous chapter we have shown, using Glauber's formalism, that

$$I(\nu,\underline{r}) = \frac{\hbar}{2\pi}\frac{\nu^3}{c^3}\sum_{\sigma=1}^{2}\int_0^{4\pi}\langle n_{\varrho\sigma}\rangle\, d\Omega_{\hat{k}} \tag{5.B.76}$$

where $\langle n_{\varrho\sigma}\rangle = \langle a^+_{\varrho\sigma}\, a_{\varrho\sigma}\rangle$ is the function evaluated in the previous chapter.

In Mollow's paper[9] he explains how the coherent and incoherent parts of the spectrum can be separated off. For the present, since we don't yet know the roots of $F(s+i\omega_s)$, we shall ignore this and just calculate the total spectral correlation function $\tilde{g}_8(\nu)$.

$$\tilde{g}_8(\nu) = \int_{-\infty}^{\infty} d\tau \, g_8(\tau) e^{i\nu\tau}$$

$$= \left\{ \int_{-\infty}^{\infty} d\tau \, e^{i\nu\tau} \psi_{81,51}(\tau) \right\} \{\rho^{(1)}_{88_\infty}\}_{t t'}$$

$$= \{\rho^{(1)}_{88_\infty}\}_{t t'} \left\{ \int_0^{\infty} d\tau \, \psi_{81,51}(\tau) e^{i\nu\tau} + \int_{-\infty}^{0} d\tau \, \psi_{81,51}(-\tau) e^{-i\nu\tau} \right\}$$

$$= \rho^{(1)}_{88} \left\{ \int_0^{\infty} d\tau \, \psi_{81,51}(\tau) e^{i\nu\tau} - \int_{\infty}^{0} d\tau \, \psi_{81,51}(-\tau) e^{-i\nu\tau} \right\}$$

where $\rho^{(1)}_{88} = (\rho^{(1)}_{88_\infty})_{t t'}$ (5.B.77)

$$= \rho^{(1)}_{88} \left\{ \int_0^{\infty} d\tau \, \psi_{81,51}(\tau) e^{i\nu\tau} + \int_0^{\infty} d\tau \, \psi^*_{81,51}(\tau) e^{-i\nu\tau} \right\}$$

since $\psi_{81,51}(-\tau) = \psi^*_{81,51}(\tau)$

$$= \rho^{(1)}_{88} \, 2 \operatorname{Re} \int_0^{\infty} d\tau \, e^{i\nu\tau} \psi_{81,51}(\tau)$$

$$= 2\rho^{(1)}_{88} \operatorname{Re} \hat{\psi}_{81,51}(-i\nu)$$

if $\rho^{(1)}_{88}$ is REAL as it should be since it represents the equilibrium probability of finding the atom in excited states $|8\rangle$.

where $$\hat{\psi}_{81,51}(s) = \int_0^{\infty} d\tau \, \psi_{81,51}(\tau) e^{-s\tau} = \left\{ \frac{f_1(s+i\omega_s) f_2(s+i\omega_s) + \tfrac{1}{4} G_{10}^2}{F(s+i\omega_s)} \right\}$$

$$\therefore \; g_8(\nu) = \frac{2\rho^{(1)}_{88}}{|F(-i(\nu-\omega_s))|^2} \operatorname{Re} \left\{ F(-i(\nu-\omega_s))^* \left[f_1(-i(\nu-\omega_s)) f_2(-i(\nu-\omega_s)) + \tfrac{1}{4} G_{10}^2 \right] \right\} \quad \text{(5.B.78a)}$$

where
$$f_1(-i(\nu-\omega_s)) = \tfrac{1}{2}\gamma' - i(\nu' + \Delta_{51})$$
$$f_2(-i(\nu-\omega_s)) = \tfrac{1}{2}\gamma'' - i(\nu' + \Delta_{51} + \Delta_{10,5})$$
$$f_3(-i(\nu-\omega_s)) = \tfrac{1}{2}\gamma_8 - i\nu'$$
$$F(-i(\nu-\omega_s)) = f_3(-i(\nu-\omega_s))\{ f_1(-i(\nu-\omega_s)) f_2(-i(\nu-\omega_s)) + \tfrac{1}{4} G_{10}^2 \} + \tfrac{1}{4} G_5^2 f_2(-i(\nu-\omega_s))$$
$$\nu' = \nu - \omega_8' = \nu - (\epsilon_{81} - \Omega_8) \quad \text{(5.B.78b)}$$
$$\Delta_{51} = \omega_1' - \omega_s = (\epsilon_{51} - \Omega_{51}) - \omega_s$$
$$\Delta_{10,5} = \omega_3' - \omega_L = (\epsilon_{10,5} - (\Omega_{10} - \Omega_{51})) - \omega_L$$

Thus

$$\tilde{g}_8(\nu) = \frac{2(\rho^{(\,)}_{aa})_{t\,t'}}{|F(-i(\nu-\omega_5))|^2} \Big\{ \tfrac12\gamma_8\Big[\{\tfrac14\gamma'\gamma'' - (\nu'+\Delta_{31})(\nu'+\Delta_{31}+\Delta_{10,3}) + \tfrac14 G_{10}^2\}^2 + \{\tfrac12\gamma'(\nu'+\Delta_{31}+\Delta_{10,3}) + \tfrac12\gamma''(\nu'+\Delta_{31})\}^2 + \tfrac14 G_3^2\big[\tfrac12\gamma''\{\tfrac14\gamma'\gamma'' + \tfrac14 G_{10}^2\} + \tfrac12\gamma'\{\nu'+\Delta_{31}+\Delta_{10,3}\}^2\big]\Big]\Big\} \tag{5.B.79}$$

on expanding the numerator and taking the real part.

Now we will consider the cubic equation

$$F(s) = s^3 + as^2 + bs + c$$

where $a = \tfrac12(\gamma'+\gamma''+\gamma_8) + i(3\omega'+\Delta_{31}-\Delta_{10,3})$

$b = \{\tfrac14(\gamma'\gamma''+\gamma_8(\gamma'+\gamma'')) - \omega'(\omega'-\Delta_{10,3}) - (\omega'+\Delta_{31})(2\omega'-\Delta_{10,3}) + \tfrac14(G_{10}^2+G_3^2)\}$
$+ i\tfrac12\{\gamma'(\omega'-\Delta_{10,3}) + \gamma''\omega' + \gamma_8(2\omega'-\Delta_{10,3}) + (\gamma'+\gamma'')(\omega'+\Delta_{31})\}$

$c = \tfrac12\{\gamma_8[\tfrac14\gamma'\gamma'' - \omega'(\omega'-\Delta_{10,3}) + \tfrac14 G_{10}^2] + \gamma''\tfrac14 G_3^2 - (\omega'+\Delta_{31})(\gamma'(\omega'-\Delta_{10,3}) + \gamma''\omega')\}$
$+ i\{(\omega'+\Delta_{31})[\tfrac14\gamma'\gamma'' - \omega'(\omega'-\Delta_{10,3}) + \tfrac14 G_{10}^2] + \tfrac14\gamma_8[\gamma'(\omega'-\Delta_{10,3}) + \gamma''\omega']$
$+ (\omega'-\Delta_{10,3})\tfrac14 G_3^2\}$ (5.B.80)

and $\omega' = \omega_8' - \omega_4'$

The solutions, as shown in Appendix III, are

$$s_1 = x_1 + ix_2 = (\alpha+\beta) - \tfrac13 a$$

$$s_2 = x_3 + ix_4 = -\{[\tfrac12(\alpha+\beta) + \tfrac13 a] - i\tfrac{\sqrt3}{2}(\alpha-\beta)\}$$
$$= -\tfrac12\{(\alpha+\beta) + \sqrt3\, i(\alpha-\beta)\} - \tfrac13\{\tfrac12(\gamma'+\gamma''+\gamma_8) + i(3\omega'+\Delta_{31}-\Delta_{10,3})\}$$

$$s_3 = x_5 + ix_6 = -\{[\tfrac12(\alpha+\beta) + \tfrac13 a] + i\tfrac{\sqrt3}{2}(\alpha-\beta)\}$$
$$= -\tfrac12\{(\alpha+\beta) - \sqrt3\, i(\alpha-\beta)\} - \tfrac13\{\tfrac12(\gamma'+\gamma''+\gamma_8) + i(3\omega'+\Delta_{31}-\Delta_{10,3})\} \tag{5.B.81}$$

where $\alpha = \left\{\dfrac{-G+\sqrt{G^2+4H^3}}{2}\right\}^{1/3} = \left\{\dfrac{-G+A}{2}\right\}^{1/3}$

$\beta = \left\{\dfrac{-G-\sqrt{G^2+4H^3}}{2}\right\}^{1/3} = -\left\{\dfrac{G+A}{2}\right\}^{1/3}$

and $G = \tfrac{2}{27}a^3 - \tfrac13 ab + c$

$H = \tfrac13(-\tfrac13 a^2 + b)$

also $A = \sqrt{G^2+4H^3}$

It can be shown that

$$\left.\begin{aligned}
G &= \tfrac{1}{216}\left\{27\Gamma G_5^2 + 2g^3 - 9gh\right\} \\
H &= -\tfrac{1}{36}\left\{g^2 - 3h\right\} \\
\text{and}\quad A &= \tfrac{\sqrt{3}}{72}\left\{27\Gamma^2 G_5^4 + 4\Gamma G_5^2 g^3 - 18\Gamma G_5^2 gh - g^2h^2 + 4h^3\right\}^{1/2} \\
\text{where}\quad i\Gamma_{31} &= \tfrac{1}{2}\gamma_{31} - i\Delta_{31} \\
i\Gamma &= \tfrac{1}{2}\gamma_{10} - i\left(\Delta_{10,3} + \Delta_{31}\right) \\
g &= \Gamma + \Gamma_{31} \\
\text{and}\quad h &= G_5^2 + G_{10}^2 + \Gamma\Gamma_{31}
\end{aligned}\right\} \qquad (5.B.82)$$

Thus, using this notation, we can write

$$S_1 = x_1 + ix_2 = (\alpha+\beta) - \tfrac{1}{2}\gamma_8 - \tfrac{1}{6}(\Gamma+\Gamma_{31}) - i(\omega_8' - \omega_5)$$

$$S_2 \quad x_3 + ix_4 = -\tfrac{1}{2}\left\{(\alpha+\beta) + i\sqrt{3}(\alpha-\beta)\right\} - \tfrac{1}{2}\gamma_8 - \tfrac{1}{6}(\Gamma+\Gamma_{31}) - i(\omega_8' - \omega_5)$$

$$S_3 \quad x_5 + ix_6 = -\tfrac{1}{2}\left\{(\alpha+\beta) - i\sqrt{3}(\alpha-\beta)\right\} - \tfrac{1}{2}\gamma_8 - \tfrac{1}{6}(\Gamma+\Gamma_{31}) - i(\omega_8' - \omega_5)$$

and

$$\bar{g}_0(\nu) = \frac{2\left(\rho_{88}^{(5)}\right)_{t=t'}\left\{\tfrac{1}{2}\gamma_8\left[\left\{\tfrac{1}{4}\gamma'\gamma'' - (\nu'+\Delta_{31})(\nu'+\Delta_{31}+\Delta_{10,3}) + \tfrac{1}{4}G_{10}^2\right\}^2 + \left\{\tfrac{1}{2}\gamma'(\nu'+\Delta_{31}+\Delta_{10,3}) + \tfrac{1}{2}\gamma''(\nu'+\Delta_{31})\right\}^2\right] + \tfrac{1}{4}G_5^2\left[\tfrac{1}{2}\gamma''\left\{\tfrac{1}{4}\gamma'\gamma'' + \tfrac{1}{4}G_{10}^2\right\} + \tfrac{1}{2}\gamma'\left\{\nu'+\Delta_{31}+\Delta_{10,3}\right\}^2\right]\right\}}{\left\{x_1^2 + (\nu-(\omega_5-x_2))^2\right\}\left\{x_3^2 + (\nu-(\omega_5-x_4))^2\right\}\left\{x_5^2 + (\nu-(\omega_5-x_6))^2\right\}}$$

Thus it is obvious that the spectral profile contains three peaks situated at

$$\nu_1 = \omega_5 - x_2$$

$$\nu_2 = \omega_5 - x_4$$

$$\nu_3 = \omega_5 - x_6$$

where x_2, x_4 and x_6 are determined by the roots of the cubic equation $F(s)$. The $\tfrac{1}{2}$-widths of the peaks are x_1, x_3, x_5.

Since it is not possible to solve the cubic equation exactly, unless numerical values are assigned to all the symbols, we shall consider various approximations:-

(i) $\underline{\Delta_{3,1} = \Delta_{10,3} = 0}$

i.e. both fields, E_S and E_L, are in EXACT resonance with the corresponding atomic transitions

$$\tfrac{1}{2}\Gamma_{31} = \tfrac{1}{2}\gamma_{31}$$

and $\tfrac{1}{2}\Gamma_{10} = \tfrac{1}{2}\gamma_{10}$

(ii) $\underline{\gamma_{31} >> \gamma_{10}}$ where $\gamma_{10} = \gamma_{10,2} + \gamma_{10,3} + \gamma_{10,7} + \gamma_{10,8}$

[N.B. It is usually true that lower energy levels have larger time widths.]

We will ignore 1st and higher order terms in $(\gamma_{10}/\gamma_{31})$.

(iii) $\underline{G^2 >> \gamma_{31}}$ where $G^2 = G_3^2 + G_{10}^2$.

We will ignore 1st and higher order terms in $(\gamma_{31}/G)^2$.

i.e. both fields are strong in comparison with the line width for level 3.

With approximation (i)

$$\tilde{\mathcal{E}}_S(\nu) = \frac{2(\rho^{(-)}_{88x})_{t\,t'}\left\{\tfrac{1}{2}\gamma_8\left[\left\{\tfrac{1}{4}\gamma'\gamma'' - \nu'^2 + \tfrac{1}{4}G_{10}^2\right\}^2 + \tfrac{1}{4}(\gamma'+\gamma'')^2\nu'^2\right] + \tfrac{1}{4}G_3^2\left[\tfrac{1}{2}\gamma'\left\{\tfrac{1}{4}\gamma'\gamma'' + \tfrac{1}{4}G_{10}^2\right\} + \tfrac{1}{2}\gamma'\nu'^2\right]\right\}}{|F(-i(\nu-\omega_S))|^2}$$

where $G = \tfrac{1}{216}\left\{27\gamma_{10}G_3^2 + 2(\gamma_{10}+\gamma_{31})^3 - 9(\gamma_{10}+\gamma_{31})(G_3^2+G_{10}^2+\gamma_{10}\gamma_{31})\right\}$ and is real

$$= \tfrac{1}{216}\left\{27\gamma_{10}G_3^2 + 2\gamma_{10}^3 + 2\gamma_{31}^3 + 6\gamma_{10}\gamma_{31}^2 + 6\gamma_{10}^2\gamma_{31} - 9(\gamma_{10}+\gamma_{31})G^2 - 9(\gamma_{10}+\gamma_{31})\gamma_{10}\gamma_{31}\right\} \quad (5.B.83)$$

$$H = -\tfrac{1}{36}\left\{(\gamma_{10}+\gamma_{31})^2 - 3(G_3^2+G_{10}^2+\gamma_{10}\gamma_{31})\right\} \text{ and is also real}$$

$$= -\tfrac{1}{36}\left\{\gamma_{10}^2+\gamma_{31}^2 - \gamma_{10}\gamma_{31} - 3G^2\right\}$$

$$A = \tfrac{\sqrt{3}}{72}\left\{27\gamma_{10}^2G_3^4 + 4\gamma_{10}G_3^2(\gamma_{10}+\gamma_{31})^3 - 18\gamma_{10}G_3^2(\gamma_{10}+\gamma_{31})(G_3^2+G_{10}^2+\gamma_{10}\gamma_{31}) - (\gamma_{10}+\gamma_{31})^2(G_3^2+G_{10}^2+\gamma_{31}\gamma_{10})^2 + 4(G_3^2+G_{10}^2+\gamma_{10}\gamma_{31})^3\right\}^{1/2}$$

With approximation (ii)

$$H = -\tfrac{1}{36}\gamma_{31}^2\left\{1 - \gamma_{10}/\gamma_{31} - 3G^2/\gamma_{31}^2\right\} \quad \text{to 1st order in } \gamma_{10}/\gamma_{31}$$

$$= -\tfrac{1}{36}\gamma_{31}^2\left\{1 - 3G^2/\gamma_{31}^2\right\} \quad \text{to zeroeth order}$$

$$G = \tfrac{1}{216}\gamma_{31}^3\left\{2 + 9\gamma_{10}/\gamma_{31}\left(2G_3^2/\gamma_{31}^2 - G_{10}^2/\gamma_{31}^2\right) - 3\gamma_{10}/\gamma_{31} - 9G^2/\gamma_{31}^2\right\} \quad \text{to 1st order} \quad (5.B.84)$$

$$= \tfrac{1}{108}\gamma_{31}^3\left\{1 - \tfrac{9}{2}G^2/\gamma_{31}^2\right\} \quad \text{to zeroeth order}$$

and $A = \tfrac{1}{72}\gamma_{31}^3\,G^2/\gamma_{31}^2\sqrt{12G^2/\gamma_{31}^2 - 3}$ to zeroeth order

With approximation (iii)

$$
\begin{aligned}
H &= 1/12\, G^2 \\
G &= -1/24\, G^2 \gamma_{31} \\
\therefore A &= \sqrt{3}/36\, G^3 \\
\therefore \alpha &= \sqrt{3}/6\, G \\
\text{and} \quad \beta &= -\sqrt{3}/6\, G \qquad (5.B.85) \\
\alpha+\beta &= 0 \\
\alpha-\beta &= \sqrt{3}/3\, G \\
\therefore S_1 &= x_1 + i x_2 = -1/3 \left\{ 1/2 (\gamma' + \gamma'' + \gamma_8) + i(3\omega' + \Delta_{31} - \Delta_{10,3}) \right\} \\
S_2 &= x_3 + i x_4 = -1/6 \left\{ (\gamma' + \gamma'' + \gamma_8) + i\left(2(3\omega' + \Delta_{31} - \Delta_{10,3}\right) + 3G \right\} \\
S_3 &= x_5 + i x_6 = -1/6 \left\{ (\gamma' + \gamma'' + \gamma_8) + i\left(2(3\omega' + \Delta_{31} - \Delta_{10,3}\right) - 3G \right\}
\end{aligned}
$$

and hence when $\Delta_{31} = \Delta_{10,3} = 0$ the three peaks occur at

$$
\left.
\begin{aligned}
\nu_1 &= \omega_5 - x_2 = \omega_5 + 1/3(3\omega') = \omega_5 + \omega' = \omega_5 + \omega_8' - \omega_1' \\
\nu_2 &= \omega_5 - x_4 = \omega_5 + 1/6\{2(3\omega') + 3G\} = \omega_5 + \omega' + 1/2\, G = \omega_5 + \omega_8' - \omega_1' + 1/2\sqrt{G_3^2 + G_{10}^2} \\
\nu_3 &= \omega_5 - x_6 = \omega_5 + 1/6\{2(3\omega') - 3G\} = \omega_5 + \omega' - 1/2\, G = \omega_5 + \omega_8' - \omega_1' - 1/2\sqrt{G_3^2 + G_{10}^2} \\
&\text{i.e. at} \\
\nu_1' &= \omega_5 - \omega_1' \\
\nu_2' &= \omega_5 - \omega_1' + 1/2\sqrt{G_3^2 + G_{10}^2} \\
\nu_3' &= \omega_5 - \omega_1' - 1/2\sqrt{G_3^2 + G_{10}^2}
\end{aligned}
\right\} \quad (5.B.86)
$$

but at resonance $\omega_5 = \omega_1'$

$$
\begin{aligned}
\therefore \nu_1' = \nu_1 - \omega_8' &= 0 \\
\nu_2' = \nu_2 - \omega_8' &= 1/2\sqrt{G_3^2 + G_{10}^2} \\
\nu_3' = \nu_3 - \omega_8' &= 1/2\sqrt{G_3^2 + G_{10}^2}
\end{aligned}
$$

The half widths are

$$
\left.
\begin{aligned}
|x_1| = |x_3| = |x_5| &= 1/6(\gamma' + \gamma'' + \gamma_8) \\
&= 1/6(3\gamma_8 + \gamma_{31} + \gamma_{10}) \\
&= 1/6(3\gamma_8 + \gamma_{31}) \quad \text{since } \gamma_{10} << \gamma_{31}
\end{aligned}
\right\} \quad (5.B.87)
$$

and the spectral correlation function is:-

$$\frac{\tilde{g}_8(\nu')}{(\rho^{(\cdot)})_{t,t'}} = \frac{\begin{aligned}&\Big[\gamma_8/\gamma_{31}\Big\{\Big[\tfrac14(\gamma/\gamma_{31})^2 + \tfrac14\,\gamma/\gamma_{31} + \tfrac14\,\gamma/\gamma_{31} + \tfrac14\,\gamma\gamma/\gamma_{31}^2 - (\nu'/\gamma_{31})^2 + \tfrac14(\epsilon_{10}/\gamma_{31})^2\Big]^2 \\ &\quad + \tfrac14(1+\gamma/\gamma_{31})^2(\nu'/\gamma_{31})^2\Big\} \\ &\quad + [\text{illegible}]\Big\{(\gamma/\gamma_{31} + \gamma/\gamma_{31})\Big[\tfrac14(\gamma/\gamma_{31})^2 + \tfrac14\,\gamma/\gamma_{31} + \tfrac14\,\gamma\gamma/\gamma_{31}^2 + \tfrac14\,\gamma/\gamma_{31} + \tfrac14(\epsilon/\gamma_{31})^2\Big] \\ &\quad + (1+\gamma/\gamma_{31})(\nu'/\gamma_{31})^2\Big\}\Big]\gamma_{31}^5\end{aligned}}{\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + \nu'^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'-\tfrac12 G)^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'+\tfrac12 G)^2\}} \tag{5.B.88}$$

and if $(\epsilon/\gamma_{31})^2 >> (\epsilon/\gamma_{31}),\ (\gamma/\gamma_{31})\ (\gamma/\gamma_{31}),\ (\gamma\gamma/\gamma_{31}^2)$

i.e. $\epsilon_{10} >> \gamma_{31},\ \sqrt{\gamma_8\gamma_{31}},\ \sqrt{\gamma_8\gamma_{31}},\ \sqrt{\gamma_8\gamma_{10}}$

then the numerator is modified so that

$$\frac{\tilde{g}_8(\nu')}{(\rho^{(\cdot)})_{t,t'}} = \frac{\begin{aligned}&\Big[\gamma_8/\gamma_{31}\Big\{\Big[\tfrac14(\epsilon_{10}/\gamma_{31})^2 - (\nu'/\gamma_{31})^2\Big]^2 + \tfrac14(1+\gamma_8/\gamma_{31})^2(\nu'/\gamma_{31})^2\Big\} \\ &\quad + \tfrac14(\epsilon/\gamma_{31})^2\Big\{(\gamma_8/\gamma_{31} + \gamma_{10}/\gamma_{31})\tfrac14(\epsilon_{10}/\gamma_{31})^2 + (1+\gamma_8/\gamma_{31})(\nu'/\gamma_{31})^2\Big\}\Big]\gamma_{31}^5\end{aligned}}{\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + \nu'^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'-\tfrac12 G)^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'+\tfrac12 G)^2\}}$$

$$= \frac{\begin{aligned}&\Big[\gamma_8/\gamma_{31}\Big\{\Big[\tfrac1{16}(\epsilon_{10}/\gamma_{31})^4 + (\nu'/\gamma_{31})^4\Big] + \Big[\tfrac14(1+\gamma_8/\gamma_{31})^2 - \tfrac12(\epsilon_{10}/\gamma_{31})^2\Big](\nu'/\gamma_{31})^2\Big\} \\ &\quad + \tfrac14(\epsilon_3/\gamma_{31})^2\Big\{(\gamma_8/\gamma_{31} + \gamma_{10}/\gamma_{31})\tfrac14(\epsilon_{10}/\gamma_{31})^2 + (1+\gamma_8/\gamma_{31})(\nu'/\gamma_{31})^2\Big\}\Big]\gamma_{31}^5\end{aligned}}{\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + \nu'^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'-\tfrac12 G)^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'+\tfrac12 G)^2\}}$$

and for $\tfrac12(\epsilon_{10}/\gamma_{31})^2 >> \tfrac14,\ \tfrac14(\gamma_8/\gamma_{31})^2,\ \tfrac12(\gamma_8/\gamma_{31})$

i.e. $\epsilon_{10} >> \sqrt{2}/2\,\gamma_{31},\ \sqrt{2}/2\,\gamma_8,\ \sqrt{\gamma_8\gamma_{31}}$

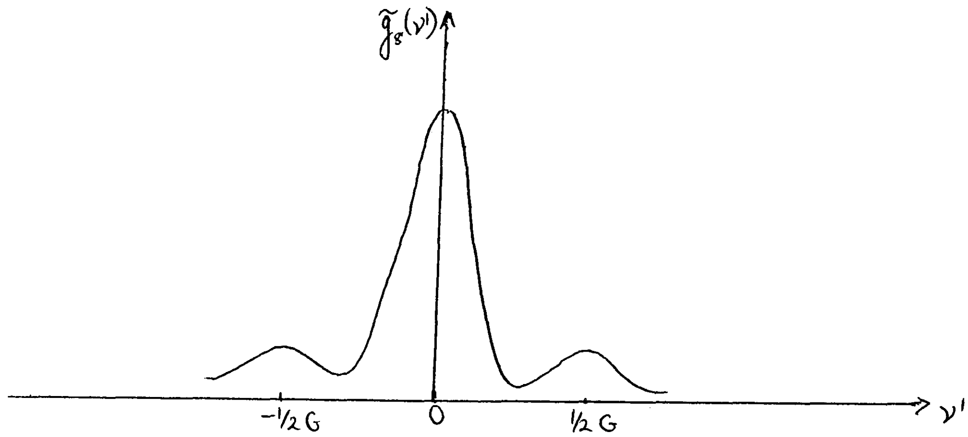
$$\boxed{\frac{\tilde{g}_8(\nu')}{(\rho^{(\cdot)})_{t,t'}} = \frac{\Big[\gamma_8\{\tfrac1{16}\epsilon_{10}^4 + \nu'^4 - \tfrac12 G_{10}^2\nu'^2\} + \tfrac14 G_3^2\{(\gamma_8+\gamma_{10})\tfrac14 G_{10}^2 + (\gamma_8+\gamma_{31})\nu'^2\}\Big]}{\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + \nu'^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'-\tfrac12 G)^2\}\{\tfrac1{36}(3\gamma_8+\gamma_{31})^2 + (\nu'+\tfrac12 G)^2\}}} \tag{5.B.89}$$

We can show that the central peak at $\nu' = 0$ is higher than the two side peaks at $\nu' = \pm\tfrac12 G$, which are of equal height. The central peak is of height

$$\tilde{g}_8(\nu'=\nu_1') = \frac{\tfrac14 G_{10}^2\{\gamma_8\tfrac14 G_{10}^2 + (\gamma_8+\gamma_{10})\tfrac14 G_3^2\}}{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)^2\left\{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)^2 + \tfrac14 G^2\right\}^2} \tag{5.B.90a}$$

and the two side peaks are of height

$$\tilde{g}_8(\nu'=\nu_2') = \frac{\tfrac14 G_3^2\{(2\gamma_8+\gamma_{31})\tfrac14 G_3^2 + (2\gamma_8+\gamma_{31}+\gamma_{10})\tfrac14 G_{10}^2\}}{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)^2\left\{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right) + \tfrac14 G^2\right\}\left\{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right) + G^2\right\}}\,(\rho^{(\cdot)}_{88_x})_{t\cdot t'} \tag{5.B.90b}$$



We will now consider some different approximations.

(iv) If we had assumed approximation (i) and then

$$G_3^2, G_{10}^2 \gg \gamma_8^2, \gamma_8\gamma_{31}, \gamma_{10}\gamma_{31}, \gamma_8\gamma_{10},$$

without assuming approximation (ii), i.e. $\gamma_{31} \gg \gamma_{10}$, then we would have obtained a similar result but the peak widths would be wider, viz. $\left(\frac{3\gamma_8+\gamma_{10}+\gamma_{31}}{6}\right)$ instead of $\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)$

(v) $\Delta_{31} = \Delta_{10,3} = 0$

$G_{10}^2 \gg G_3^2$ (laser field STRONGER than SRS field, which is usually the case experimentally - in fact E_L is generally $\sim 2\,MW$. and $E_S \sim 300\,KW$. for β-methyl naphthalene)

$G_{10}^2 \gg \gamma_8^2, \gamma_8\gamma_{31}, \gamma_{10}\gamma_{31}, \gamma_8\gamma_{10}$

then

$$\tilde{\varepsilon}_8(\nu') = \frac{\left\{\gamma_8(\nu'^2-\tfrac14 G_{10}^2)^2 + \tfrac14 G_3^2\,\gamma'(\tfrac14 G_{10}^2\,\gamma''/\gamma' + \nu'^2\right\}\left(\rho^{(s)}_{88\infty}\right)_{t=t'}}{\left\{\left(\frac{3\gamma_8+\gamma_{31}+\gamma_{10}}{6}\right)^2+\nu'^2\right\}\left\{\left(\frac{3\gamma_8+\gamma_{31}+\gamma_{10}}{6}\right)^2+(\nu'-\tfrac12 G_{10})^2\right\}\left\{\left(\frac{3\gamma_8+\gamma_{31}+\gamma_{10}}{6}\right)^2+(\nu'+\tfrac12 G_{10})^2\right\}} \quad (5.B.91)$$

which is the same as case (iv) except that now side peaks occur at $\nu' = \pm\tfrac12 G_{10}$ not at $\nu' = \pm\tfrac12\sqrt{G_3^2+G_{10}^2}$, i.e. they are now nearer to the central peak.

(vi) $\Delta_{31} = \Delta_{10,3} = 0$

$\gamma_{31} \gg \gamma_{10}$

$\gamma_{31}^2 \gg G^2$ i.e. both fields WEAK then

$$[illegible] \qquad (5.B.92)$$

i.e. all three peaks are coincident at $\nu' = 0$ and there is no level shift or splitting.

The height of this peak is

$$[illegible] \qquad (5.B.93)$$

Thus we see that the splitting up of the spectral profile occurs only when one or both of the fields are STRONG and so it must be a truly non-linear effect.

We shall consider one further case:

(vii) $\underline{\Delta_{..} = \Delta_{..} = 0}$

$\underline{\gamma_{..} \gg \gamma_{..}}$

$\underline{\gamma_{..} \gg G_3^2}$ i.e. SRS field WEAK compared to line width of level 3

$\underline{\gamma_{..} \ll G_{10}^2}$ i.e. laser field STRONG compared to line width of level 3

This is also close to the experimental situation. The spectral correlation function at $\nu = \nu'$ is now

$$J^{(\nu)} = \frac{\left\{\tfrac12\gamma_2\left[[illegible](\gamma_5+\gamma_{10}) - \nu'^2 + \tfrac14 G_{10}^2\right]^2 + \left\{\tfrac12(2\gamma_5+\gamma_{51}+\gamma_{10})\right\}^2\nu'^2\right] + \tfrac12 G\left[[illegible](\gamma_5+\gamma_{10})\left\{(\gamma_5+\gamma_{51})(\gamma_5+\gamma_{10}) + G_{10}^2\right\} - [illegible](\gamma_5+\gamma_{51})\nu'^2\right]\Big\}\left(\rho^{(0)}_{..}\right)}{\left\{\left(\frac{3\gamma_5+\gamma_{51}}{6}\right)^2 + \nu'^2\right\}\left\{\left(\frac{3\gamma_5+\gamma_{51}}{6}\right)^2 + \left(\nu' - \tfrac12 G_{10}\right)^2\right\}\left\{\left(\frac{3\gamma_5+\gamma_{51}}{6}\right)^2 + \left(\nu' + \tfrac12 G_{10}\right)^2\right\}} \qquad (5.B.94)$$

This is a similar situation to case (v) except that different assumptions have been made about the sizes of the line widths. Here the side peaks occur in the same places $\nu' \pm \tfrac12 G_{10}$ but their widths are narrower being $\left(\frac{3\gamma_5+\gamma_{51}}{6}\right)$ not $\left(\frac{3\gamma_5+\gamma_{51}+\gamma_{10}}{6}\right)$

We shall discuss these results after calculating the other spectral correlation function for transitions between levels 10 and 8.

(b) Spectral correlation function for transitions between levels 6S (10) and $5P_{3/2}$ (8)

We now need to find $\rho^{(\,)}(t)$ [illegible] by solving the hermitian conjugate equations of (5.B.51-53).

i.e. $\dot{\rho}^{(\,)}_{[illegible]}(t) = -\{\tfrac{1}{2}(\gamma + \gamma) - i(\omega - \omega + \omega_{5'})\}\rho^{(\,)}_{5'}(t)$ [illegible]

$+ i\lambda$ [illegible] $\rho(t)$

$+ i\lambda$ [illegible] $\rho(t)$ (5.B.95)

$\dot{\rho}^{(\,)}(t) = -\{$ [illegible] $\}\rho^{(\,)}(t)$

$+ i\lambda\mathcal{E}$ [illegible] $\rho(t)$ (5.B.96)

$\dot{\rho}^{(\,)}(t) = -\{\tfrac{1}{2}(\gamma + \gamma_{10}) - i(\omega - \omega + \omega_{10})\}\rho^{(\,)}_{10}(t)$

$+ \lambda\mathcal{E}$ [illegible] $\rho(t)$ (5.B.97)

Multiplying (5.B.96) by [illegible] and (5.B.97) by $e^{i\omega_L t}$, we obtain equations which may be solved in a way analogous to the method used in Sec. (b). Thus

$$\hat{\rho}_{10,8}(s) = \hat{U}_{10,8,10,8}(s)\rho_{10,8}(t') - \hat{U}_{10,8,1,8}(s)\rho_{1,8}(t') + \hat{U}_{10,8,3,8}(s)\rho_{38}(t') \qquad (5.B.98)$$

where

$$U_{10,8,10,8}(s) = \frac{\{g_1(s+i\omega_L)\,g_2(s+i\omega_L) + \tfrac{1}{4}\mathcal{E}_3^2\}}{G(s+i\omega_L)}\,e^{-st'} = \hat{\psi}_{10,8,10,8}(s)\,e^{-st'}$$

$$U_{10,8,1,8}(s) = -\frac{(\lambda_{51}\mathcal{E}_{cs})(\lambda_{10,3}\mathcal{E}_{cL})}{G(s+i\omega_L)}\,e^{-(s+i(\omega_L+\omega_s))t'} = \hat{\psi}_{10,8,1,8}(s)\,e^{-(s+i(\omega_L+\omega_s))t'} \qquad (5.B.99)$$

$$U_{10,8,3,8}(s) = \frac{(i\lambda_{10,3}\mathcal{E}_{cL})\,g_3(s)}{G(s+i\omega_L)}\,e^{-(s+i\omega_L)t'} = \hat{\psi}_{10,8,3,8}(s)\,e^{-(s+i\omega_L)t'}$$

$$g_1(s) = (s + \tfrac{1}{2}\gamma' - i(\omega_8' - \omega_1'))$$

$$g_2(s) = (s + \tfrac{1}{2}\gamma'' + i(\omega_{10}' - \omega_L))$$

$$g_3(s) = (s + \tfrac{1}{2}\gamma_8 - i(\omega_8' - \omega_s)) \qquad (5.B.100)$$

$$G(s) = \{g_1(s)g_2(s) + \tfrac{1}{4}\mathcal{E}_{11}^2\}\,g_3(s) + \tfrac{1}{4}\mathcal{E}_3^2\,g_2(s)$$

The relevant 2-time atomic correlation function, in this case, according to equation (5.B.50) is

$$g^{(1,1)} = \langle P^{\dagger}(t') P(t') \rangle \; g_{10}(\tau,t') \; \langle P^{\dagger}(t') P(t'+\tau) \rangle$$

$$\triangleq \sum_{j} U_{[illegible]}(\tau,t')\rho^{(1)}_{j}(t') \text{ under Markoff approximation} \qquad (5.B.101)$$

$$= U_{[illegible]}(\tau,t')\,[illegible] + U_{[illegible]}(\tau,t')\rho^{(1)}(t') + U_{[illegible]}(\tau,t')\rho^{(1)}(t')$$

As in (5.B.69)

$$U \; [illegible] \; \Psi \; [illegible]$$

$$U \; [illegible] \; \Psi \; [illegible](\tau)\, e^{[illegible]t'} \qquad (5.B.102)$$

$$U \; [illegible](t'+\tau) \; \Psi \; [illegible](\tau)\, e^{[illegible]t'}$$

Hence, since $U_{[illegible]}(\tau,t') \; U_{[illegible]}(t'+\tau)$, we have

$$g^{(1,1)} \; \Psi_{[illegible]}(\tau)\rho^{(1)}(t') + \Psi_{[illegible]}(\tau)\, e^{[illegible]t'}\rho^{(1)}(t') + \Psi_{[illegible]}(\tau)\, e^{[illegible]t'}\rho^{(1)}(t') \qquad (5.B.103)$$

Assuming the atom to be in equilibrium with the field, we write

$$\rho^{(1)}(t') = \left(\rho^{(1)}\right)_{[illegible]}$$

$$\rho^{(1)}(t') = \left(\rho^{(1)}\right)_{[illegible]} \qquad (5.B.104)$$

$$\rho^{(1)}(t') = \left(\rho^{(1)}\right)_{[illegible]}$$

Since we shall find that the latter two functions can be written in the following form:-

$$\rho^{(1)}(t') = \overline{\rho^{(1)}}\, e^{i(\omega [illegible])t'}$$

$$\rho^{(1)}(t') = \overline{\rho^{(1)}}\, e^{i\omega_1 t'} \qquad (5.B.105)$$

$g_{10}(\tau,t')$ is independent of t' and can be written

$$g_{10}(\tau) = \Psi_{[illegible]}(\tau)\overline{\rho^{(1)}} + \Psi_{[illegible]}(\tau)\overline{\rho^{(1)}} + \Psi_{[illegible]}(\tau)\overline{\rho^{(1)}} \qquad (5.B.106)$$

where $\overline{\rho}$ [illegible] $(\rho$ [illegible]$)$

As in case (a) equation (5.B.77) we can deduce the <u>total</u> spectral correlation function to be:-

$$
\begin{aligned}
\mathcal{J}(\nu) = \; & \overline{\rho} \,\mathrm{Re}\big(\hat{\psi}\,[\text{illegible}]\,(-i\nu)\big) \\
& + \,\mathrm{Re}\big(\hat{\psi}\,[\text{illegible}]\,(-i\nu)\,\overline{\rho_{1,10}}\big) \\
& + \,\mathrm{Re}\big(\hat{\psi}\,[\text{illegible}]\,(-i\nu)\,\overline{\rho}\,[\text{illegible}]\big)
\end{aligned}
\qquad (5.B.107)
$$

where $\overline{\rho}$ [illegible] and $\overline{\rho}$ [illegible] will be complex.

$$
\begin{aligned}
\hat{\psi}\,[\text{illegible}](-i\nu) &= \left\{ \frac{g\,(-i(\nu-\omega_L))\,g_2(-i(\nu-\omega_L)) + \tfrac{1}{4}G_3^2}{G(-i(\nu-\omega_L))} \right\} = \frac{1}{|G(-i(\nu-\omega_L))|^2} \{G^*(-i(\nu-\omega_L)) \times [\text{illegible}]\} \\
\hat{\psi}\,[\text{illegible}](-i\nu) &= -\left\{ \frac{(\lambda\,\varepsilon\,[\text{illegible}])(\lambda\,\varepsilon\,[\text{illegible}])}{G(-i(\nu-\omega_L))} \right\} = -\frac{(\lambda\,\varepsilon\,[\text{illegible}])(\lambda\,\varepsilon\,[\text{illegible}])\,G^*(-i(\nu-\omega_L))}{|G(-i(\nu-\omega_L))|^2} \qquad (5.B.108) \\
\hat{\psi}\,[\text{illegible}](-i\nu) &= \left\{ \frac{-\lambda\,\varepsilon\,[\text{illegible}]\,g_2(-i(\nu-\omega_L))}{G(-i(\nu-\omega_L))} \right\} = \frac{\lambda\,\varepsilon\,[\text{illegible}]}{|G(-i(\nu-\omega_L))|^2} \{G^*(-i(\nu-\omega_L))\,g_2(-i(\nu-\omega_L))\}
\end{aligned}
$$

$$
\begin{aligned}
\text{where}\quad \varepsilon_1(-i(\nu-\omega_L)) &= \tfrac{1}{2}\gamma' - i(\nu'' + \Delta_{\omega 3}) \\
\varepsilon_2(-i(\nu-\omega_L)) &= \tfrac{1}{2}\gamma' - i\nu'' \\
\varepsilon_3(-i(\nu-\omega_L)) &= \tfrac{1}{2}\gamma_1 - i(\nu'' + \Delta_{\omega 3} + \Delta_{31}) \\
G(-i(\nu-\omega_L)) &= \{g_1(-i(\nu-\omega_L))\,g_2(-i(\nu-\omega_L)) + \tfrac{1}{4}G_{10}^2\}\,g_3(-i(\nu-\omega_L)) + \tfrac{1}{4}G_3^2 \times g_2(-i(\nu-\omega_L)) \\
\nu'' &= \nu - \omega_{10}'
\end{aligned}
\qquad (5.B.109)
$$

Let $G(s) = (s-(a-ib))(s-(c-id))(s-(e-if))$

then $|G(-i(\nu-\omega_L))|^2 = (a^2 + (\nu-(\omega_L+b))^2)(c^2+(\nu-(\omega_L+d))^2)(e^2+(\nu-(\omega_L+f))^2)$

so that, since this will be the only factor in the denominator dependent on ν, we shall have three peaks at $\nu_1 = \omega_L + b$, $\nu_2 = \omega_L + d$, $\nu_3 = \omega_L + f$ of half widths a, c and e, where a, b, c, d, e, f are determined by the cube roots of $G(s)$.

We shall now endeavour to find values for the values of $\rho_{1,10}^{(s)}(t)$, $\rho_{3,10}^{(s)}(t)$ as $t \to \infty$ and evaluate them at t = t' so that the spectral correlation function can be determined more precisely. This demands the solution of 16 equations for $\rho_{nn}^{(1)}(t)$ for n = 1 $\longrightarrow$ 10, and $\rho_{10,5}^{(1)}$, $\rho_{5,10}^{(1)}$, $\rho_{10,3}^{(1)}$, $\rho_{3,10}^{(1)}$, $\rho_{13}^{(1)}$, $\rho_{31}^{(1)}$. In fact we shall determine the values of these two off diagonal matrix elements in terms of the diagonal matrix elements $\rho_{11}^{(1)}$, $\rho_{33}^{(1)}$ and $\rho_{10,10}^{(1)}$ since otherwise we must solve all 16 equations. These diagonal elements are real and so enable simplification of equation (5.B.107).

Letting:-

$a = \rho_{11}^{(1)}$, $b = \rho_{22}^{(1)}$, $c = \rho_{33}^{(1)}$, $d = \rho_{44}^{(1)}$, $e = \rho_{55}^{(1)}$, $f = \rho_{66}^{(1)}$,
$g = \rho_{77}^{(1)}$, $h = \rho_{88}^{(1)}$, $i = \rho_{99}^{(1)}$, $j = \rho_{10,10}^{(1)}$, $k = \rho_{5,10}^{(1)} e^{-i\omega_2 t}$,
$l = \rho_{10,5}^{(1)} e^{i(\omega_1+\omega_2)t}$, $m = \rho_{10,3}^{(1)} e^{i\omega_2 t}$, $n = \rho_{10,1}^{(1)} e^{i(\omega_1+\omega_2)t}$, $o = \rho_{13}^{(1)} e^{-i\omega_1 t}$,
$p = \rho_{31}^{(1)} e^{i\omega_1 t}$.

$$\gamma_{11} = \gamma_{12} + \gamma_{13} \tag{5.B.110}$$

$$\gamma_5 = \gamma_{52} + \gamma_{53}$$

$$\gamma_6 = \gamma_{62} + \gamma_{63}$$

$$\gamma_7 = \gamma_{71} + \gamma_{74} + \gamma_{75} + \gamma_{76}$$

$$\gamma_9 = \gamma_{92} + \gamma_{93} + \gamma_{97} + \gamma_{98}$$

and taking the Laplace transforms of the resulting 16 equations, we obtain

$$s\hat{a}(s) = \gamma_{21}\hat{b}(s) + \gamma_{31}\hat{c}(s) + \gamma_{71}\hat{g}(s) + \gamma_{81}\hat{h}(s) - i\lambda_{31}\mathcal{E}_{cs}\hat{o}(s) + i\lambda_{13}\mathcal{E}_{cs}\hat{p}(s) + e^{-st'}a(t') \tag{5.B.111}$$

$$(s+\gamma_2)\hat{b}(s) = \gamma_{42}\hat{d}(s) + \gamma_{52}\hat{e}(s) + \gamma_{62}\hat{f}(s) + \gamma_{92}\hat{i}(s) + \gamma_{10,2}\hat{j}(s) + e^{-st'}b(t') \tag{5.B.112}$$

$$(s+\gamma_3)\hat{c}(s) = \gamma_{43}\hat{d}(s) + \gamma_{53}\hat{e}(s) + \gamma_{63}\hat{f}(s) + \gamma_{93}\hat{i}(s) + \gamma_{10,3}\hat{j}(s) + i\lambda_{31}\mathcal{E}_{cs}\hat{o}(s) - i\lambda_{13}\mathcal{E}_{cs}\hat{p}(s) - i\lambda_{10,3}\mathcal{E}_{oL}\hat{k}(s) + i\lambda_{3,10}\mathcal{E}_{oL}\hat{m}(s) + e^{-st'}c(t') \tag{5.B.113}$$

$$(s+\gamma_4)\hat{d}(s) = \gamma_{74}\hat{g}(s) + \gamma_{84}\hat{h}(s) + e^{-st'}d(t') \tag{5.B.114}$$

$$(s+\gamma_5)\hat{e}(s) = \gamma_{75}\hat{g}(s) + \gamma_{85}\hat{h}(s) + e^{-st'}e(t') \tag{5.B.115}$$

$$(s+\gamma_6)\hat{f}(s) = \gamma_{76}\hat{g}(s) + \gamma_{86}\hat{h}(s) + e^{-st'}f(t') \tag{5.B.116}$$

$$(s+\gamma_7)\hat{g}(s) = \gamma_{97}\hat{i}(s) + \gamma_{10,7}\hat{j}(s) + e^{-st'}g(t') \tag{5.B.117}$$

$$(s+\gamma_{1})\hat{h}(s) = \gamma_{s1}\hat{i}(s) + \gamma_{1}\hat{j}(s) + e^{-st'}h(t') \tag{5.B.118}$$

$$(s+\gamma)\hat{i}(s) = e^{-st'}i(t') \tag{5.B.119}$$

$$(s+\gamma_{1})\hat{j}(s) = i\lambda_{3,10}\varepsilon_{cL}\hat{k}(s) - i\lambda_{10,3}\varepsilon_{cL}\hat{m}(s) + e^{-st'}j(t') \tag{5.B.120}$$

$$(s+\tfrac{1}{2}(\gamma_{31}+\gamma_{10})-i\Delta_{10,3})\hat{k}(s) = i\lambda_{31}\varepsilon_{cs}\hat{l}(s) - i\lambda_{3,10}\varepsilon_{cL}\hat{c}(s) + i\lambda_{3,10}\varepsilon_{cL}\hat{j}(s) + e^{-st'}k(t') \tag{5.B.121}$$

$$(s+\tfrac{1}{2}\gamma_{10}-i(\Delta_{31}+\Delta_{10,3}))\hat{l}(s) = -i\lambda_{3,10}\varepsilon_{cL}\hat{o}(s) + i\lambda_{13}\varepsilon_{cs}\hat{k}(s) + e^{-st'}l(t') \tag{5.B.122}$$

$$(s+\tfrac{1}{2}(\gamma_{31}+\gamma_{10})+i\Delta_{10,3})\hat{m}(s) = -i\lambda_{10,3}\varepsilon_{cL}\hat{j}(s) - i\lambda_{13}\varepsilon_{cs}\hat{n}(s) + i\lambda_{10,3}\varepsilon_{cL}\hat{c}(s) + e^{-st'}m(t') \tag{5.B.123}$$

$$(s+\tfrac{1}{2}\gamma_{10}+i(\Delta_{31}+\Delta_{10,3}))\hat{n}(s) = -i\lambda_{31}\varepsilon_{cs}\hat{m}(s) + i\lambda_{10,3}\varepsilon_{cL}\hat{p}(s) + e^{-st'}n(t') \tag{5.B.124}$$

$$(s+\tfrac{1}{2}\gamma_{31}-i\Delta_{31})\hat{o}(s) = -i\lambda_{10,3}\varepsilon_{cL}\hat{l}(s) - i\lambda_{13}\varepsilon_{cs}\hat{a}(s) + i\lambda_{13}\varepsilon_{cs}\hat{c}(s) + e^{-st'}o(t') \tag{5.B.125}$$

$$(s+\tfrac{1}{2}\gamma_{31}+i\Delta_{31})\hat{p}(s) = -i\lambda_{31}\varepsilon_{cs}\hat{c}(s) + i\lambda_{31}\varepsilon_{cs}\hat{a}(s) + i\lambda_{3,10}\varepsilon_{cL}\hat{n}(s) + e^{-st'}p(t') \tag{5.B.126}$$

From these equations it is obvious that it is possible to solve for $\hat{k}(s)$, $\hat{l}(s)$, $\hat{o}(s)$ in terms of $\hat{c}(s)$, $\hat{j}(s)$, $\hat{a}(s)$.

i.e. $\hat{\rho}^{(s)}_{3,10}(s+i\omega_L)$, $\hat{\rho}^{(s)}_{1,10}(s+i(\omega_s+\omega_L))$, $\hat{\rho}^{(s)}_{1,3}(s+i\omega_s)$ in terms of $\hat{\rho}^{(s)}_{33}(s)$, $\hat{\rho}^{(s)}_{10,10}(s)$, $\hat{\rho}^{(s)}_{11}(s)$, $\hat{h}(s)$ and $\hat{l}(s)$ being the most important to us.

The three equations we need are (5.B.121), (122), (125):-

$$(s+\tfrac{1}{2}(\gamma_{31}+\gamma_{10})-i\Delta_{10,3})\hat{k}(s) = i\lambda_{31}\varepsilon_{0s}\hat{l}(s) - i\lambda_{3,10}\varepsilon_{0L}\hat{c}(s) + i\lambda_{3,10}\varepsilon_{0L}\hat{j}(s) + e^{-st'}k(t') \tag{5.B.121}$$

$$(s+\tfrac{1}{2}\gamma_{10}-i(\Delta_{31}+\Delta_{10,3}))\hat{l}(s) = -i\lambda_{3,10}\varepsilon_{0L}\hat{o}(s) + i\lambda_{13}\varepsilon_{0s}\hat{k}(s) + e^{-st'}l(t') \tag{5.B.122}$$

$$(s+\tfrac{1}{2}\gamma_{31}-i\Delta_{31})\hat{o}(s) = -i\lambda_{10,3}\varepsilon_{0L}\hat{l}(s) - i\lambda_{13}\varepsilon_{0s}\hat{a}(s) + i\lambda_{13}\varepsilon_{0s}\hat{c}(s) + e^{-st'}o(t') \tag{5.B.125}$$

We obtain $\hat{o}(s)$ in terms of $\hat{l}(s)$, $\hat{a}(s)$, $\hat{c}(s)$ from (5.B.125) and substitute for it in equation (5.B.122) to obtain $\hat{l}(s)$ in terms of $\hat{k}(s)$, $\hat{a}(s)$, $\hat{c}(s)$ and then we substitute this in equation (5.B.121) to obtain $\hat{k}(s)$ in terms of $\hat{a}(s)$, $\hat{c}(s)$, $\hat{j}(s)$ which is:-

$$\hat{\rho}^{(s)}_{[illegible]}(s) = \left\{\frac{i\lambda\,\mathcal{E}\,\tfrac{1}{4}G^{2}}{H(s)}\right\} f(s) - i\lambda_{3,10}\mathcal{E}_{oL}\left\{\frac{h_2(s)h_3(s)+\tfrac{1}{4}(G_{10}^{2}-G_{3}^{2})}{H(s)}\right\} g(s)$$

$$+\ i\lambda\,\mathcal{E}\left\{\frac{h_2(s)h_3(s)+\tfrac{1}{4}G_{10}^{2}}{H(s)}\right\} d(s)$$

$$+\ \frac{(\lambda\,\mathcal{E})(\lambda\,\mathcal{E})}{H(s)}\, e^{-st'} c(t') + \frac{i\lambda_{31}\mathcal{E}_{os}\,h_3(s)}{H(s)}\, e^{-st'} f(t')$$

$$+\left\{\frac{h_2(s)h_3(s)+\tfrac{1}{4}G_{10}^{2}}{H(s)}\right\} e^{-st'} h(t') \qquad (5.B.127)$$

where $h_1(s) = s + \tfrac{1}{2}(\gamma_{31}+\gamma_{10}) - i\Delta_{10,3}$

$h_2(s) = s + \tfrac{1}{2}\gamma_{10} - i(\Delta_{31} + \Delta_{10,3})$

$h_3(s) = s + \tfrac{1}{2}\gamma_{31} - i\Delta_{31}$

$H(s) = h_1(s)\{h_2(s)h_3(s) + \tfrac{1}{4}G_{10}^{2}\} + \tfrac{1}{4}G_3^{2}h_3(s)$

$$\hat{\rho}^{(s)}_{3,10}(s)_{\text{eq value}} = \frac{i\lambda_{3,10}\mathcal{E}_{oL}\tfrac{1}{4}G_3^{2}}{H(s-i\omega_L)}\frac{\rho^{(s)}_{11}(\infty)}{(s-i\omega_L)} - i\lambda_{3,10}\mathcal{E}_{oL}\left\{\frac{h_2(s-i\omega_L)h_3(s-i\omega_L)+\tfrac{1}{4}(G_{10}^{2}-G_3^{2})}{H(s-i\omega_L)}\right\} \times\frac{\rho^{(s)}_{33}(\infty)}{(s-i\omega_L)}$$

$$+\ i\lambda_{3,10}\mathcal{E}_{oL}\left\{\frac{h_2(s-i\omega_L)h_3(s-i\omega_L)+\tfrac{1}{4}G_{10}^{2}}{H(s-i\omega_L)}\right\}\frac{\rho^{(s)}_{10,10}(\infty)}{(s-i\omega_L)}$$

$$+\ \frac{(\lambda_{31}\mathcal{E}_{os})(\lambda_{3,10}\mathcal{E}_{oL})}{H(s-i\omega_L)}\, e^{-(s-i(\omega_L-\omega_S))t'}\rho^{(s)}_{13}(t') + \frac{i\lambda_{31}\mathcal{E}_{os}\,h_3(s-i\omega_L)}{H(s-i\omega_L)}\, e^{-(s+i\omega_S)t'}\rho^{(s)}_{1,10}(t') \qquad (5.B.128)$$

$$+\left\{\frac{h_2(s-i\omega_L)h_3(s-i\omega_L)+\tfrac{1}{4}G_{10}^{2}}{H(s-i\omega_L)}\right\} e^{-st'}\rho^{(s)}_{3,10}(t')$$

where we have let $t \to \infty$ in order to find the equilibrium value and assumed that $\rho^{(s)}_{11}(t)$, $\rho^{(s)}_{33}(t)$, $\rho^{(s)}_{10,10}(t)$ are time independent in the limit $t \to \infty$ since the level populations become steady so that

$$\hat{\rho}^{(s)}_{11}(s-i\omega_L) = \frac{1}{s-i\omega_L}\rho^{(s)}_{11}(\infty) \text{ etc.}$$

If we assume also that the real parts of the cube roots of $H(s)$ are negative,

i.e. if $H(s-i\omega_L) = (s-y_1)(s-y_2)(s-y_3)$, where $y_1 = l'+im'$, $y_2 = n'+io'$, $y_3 = p'+iq'$, and l', n' and p' are negative, then the last three terms go out on taking the inverse Laplace transform since

$$\mathcal{L}^{-1}\left\{\frac{1}{H(s-i\omega_L)}\right\} = \mathcal{L}^{-1}\left\{\frac{A}{s-(\ell'+im')} + \frac{B}{s-(n'+ic')} + \frac{C}{s-(p'+iq')}\right\}$$
$$= A e^{(\ell'+im')t} + B e^{(n'+ic')t} + C e^{(p'+iq')t}$$

and if $\ell' = -|x|$, $n' = -|y|$, $p' = -|z|$ all three terms $\longrightarrow 0$ as $t \longrightarrow \infty$.

Hence

$$\left(\rho_{3,10}(t\to\infty)\right)_{t=t'} = e^{i\omega_L t'}\frac{(-i\lambda_{\cdot\,\cdot}\mathcal{E}_{\cdot L})}{y_1 y_2 y_3}\left\{\tfrac{1}{4}G_3^2\rho_{11}^{(\cdot)}(\infty) - \left[h_1(0)h_3(0) + \tfrac{1}{4}(G_{10}^2 - G_3^2)\right]\rho_{33}^{(0)}(\infty) + \left[h_2(0)h_3(0) + \tfrac{1}{4}G_{10}^2\right]\rho_{10,10}^{(0)}(\infty)\right\} \qquad (5.B.129)$$

since $H(0) = -y_1 y_2 y_3 = H(s-i\omega_L=0)$
$= h_1(0)\{h_2(0)h_3(0) + \tfrac{1}{4}G_{10}^2\} + \tfrac{1}{4}G_3^2 h_3(0)$

From equation (5.B.122) we can now obtain $\hat{\ell}(s)$ and hence $\hat{\rho}_{1,10}(s+i(\omega_3+\omega_L))$ and following the argument just given

$$\left(\rho_{1,10}(t\to\infty)\right)_{t=t'} = e^{i(\omega_3+\omega_L)t'}\frac{(-\tfrac{1}{4}G_3^2)}{y_1 y_2 y_3}\left(\frac{\lambda_{3,10}\mathcal{E}_{0L}}{\lambda_{31}\mathcal{E}_{0S}}\right)\left\{h_1(0)\rho_{11}^{(0)}(\infty) + (h_1(0)+h_2(0))\rho_{33}^{(0)}(\infty) - h_3(0)\rho_{10,10}^{(0)}(\infty)\right\} \qquad (5.B.130)$$

where
$h_1(0) = \tfrac{1}{2}(\gamma_{31}+\gamma_{10}) + i(\omega_L - \Delta_{10,3}) = h_1(s=i\omega_L)$
$h_2(0) = \tfrac{1}{2}\gamma_{10} + i(\omega_L - \Delta_{10,3} - \Delta_{31}) = h_2(s=i\omega_L)$
$h_3(0) = \tfrac{1}{2}\gamma_{31} + i(\omega_L - \Delta_{31}) = h_3(s=i\omega_L)$

Thus the form assumed in equation (5.B.105) was correct and on substituting in (5.B.107)

$$\mathcal{E}_{10}(\nu) = \frac{2\,\overline{\rho_{10,10}}}{|G(-i(\nu-\omega_L))|^2}\,\mathrm{Re}\left\{G^*(-i(\nu-\omega_L))\left[g_1(-i(\nu-\omega_L))g_3(-i(\nu-\omega_L)) + \tfrac{1}{4}G_3^2\right]\right\}$$
$$- \frac{2\,\tfrac{1}{4}G_{10}^2\,\tfrac{1}{4}G_3^2}{|G(-i(\nu-\omega_L))|^2\,|H(s=i\omega_L)|^2}\,\mathrm{Re}\left\{G^*(-i(\nu-\omega_L))H^*(s=i\omega_L)\left[h_1(0)\overline{\rho_{11}} + (h_1(0)+h_2(0))\overline{\rho_{33}} - h_3(0)\overline{\rho_{10,10}}\right]\right\}$$
$$- \frac{2\,\tfrac{1}{4}G_{10}^2}{|G(-i(\nu-\omega_L))|^2\,|H(s=i\omega_L)|^2}\,\mathrm{Re}\left\{G^*(-i(\nu-\omega_L))g_3(-i(\nu-\omega_L))H^*(-i(\nu-\omega_L)) \times\left[\tfrac{1}{4}G_3^2\overline{\rho_{11}} - (h_2(0)h_3(0) + \tfrac{1}{4}(G_{10}^2-G_3^2))\overline{\rho_{33}} + (h_2(0)h_3(0) + \tfrac{1}{4}G_{10}^2)\overline{\rho_{10,10}}\right]\right\}$$

where
$$\bar{\rho}_{11} = (\rho_{11}^{(1)}(t \to \infty))_{\nu,\nu'}$$
$$\bar{\rho}_{33} = (\rho_{33}^{(1)}(t \to \infty))_{\nu,\nu'}$$
$$\bar{\rho}_{10,3} = (\rho_{10,3}^{(1)}(t \to \infty))_{\nu,\nu'}$$

and
$$\begin{aligned} G^*(-i(\nu-\omega_L)) = \{ & \tfrac{1}{2}\gamma_x [\tfrac{1}{4}\gamma'\gamma'' - \nu''(\nu'' + \Delta_{10,3}) + \tfrac{1}{4}G_{10}^2] \\ & - \tfrac{1}{2}(\nu'' + \Delta_{10,3} + \Delta_{31})[\gamma'\nu'' + \gamma''(\nu'' + \Delta_{10,3})] \\ & + \tfrac{1}{4}G_3^2 \tfrac{1}{2}\gamma''\} \\ + i\{ & (\nu'' + \Delta_{10,3} + \Delta_{31})[\tfrac{1}{4}\gamma'\gamma'' - \nu''(\nu'' + \Delta_{10,3}) + \tfrac{1}{4}G_{10}^2] \\ & + \tfrac{1}{4}\gamma_x [\gamma'\nu'' + \gamma''(\nu'' + \Delta_{10,3})] \\ & + \tfrac{1}{4}G_3^2 \nu''\} \end{aligned}$$
$$\varepsilon_1(-i(\nu-\omega_L)) = \tfrac{1}{2}\gamma' - i(\nu'' + \Delta_{10,3})$$
$$\varepsilon_3(-i(\nu-\omega_L)) = \tfrac{1}{2}\gamma_x - i(\nu'' + \Delta_{10,3} + \Delta_{31}) \qquad (5.D.131)$$

We will now consider various approximations:

(i) $\underline{\Delta_{31} = \Delta_{10,3} = 0}$

$$h_1(0) = \tfrac{1}{2}(\gamma_{31} + \gamma_{10}) + i\omega_L$$
$$h_2(0) = \tfrac{1}{2}\gamma_{10} + i\omega_L$$
$$h_3(0) = \tfrac{1}{2}\gamma_{31} + i\omega_L$$

$$\begin{aligned} H(0) &= \{\tfrac{1}{2}(\gamma_{31}+\gamma_{10})[\tfrac{1}{4}\gamma_{31}\gamma_{10} + \tfrac{1}{4}G_{10}^2 - 2\omega_L^2] + \tfrac{1}{2}\gamma_{31}\tfrac{1}{4}G_3^2\} \\ &\quad + i\omega_L\{\tfrac{1}{4}(\gamma_{31}+\gamma_{10})^2 + \tfrac{1}{4}\gamma_{10}\gamma_{31} + \tfrac{1}{4}(G_{10}^2 + G_3^2) - \omega_L^2\} \\ &= A + iB \end{aligned}$$

$$\varepsilon_1(-i(\nu-\omega_L)) = \tfrac{1}{2}\gamma' - i\nu''$$
$$\varepsilon_3(-i(\nu-\omega_L)) = \tfrac{1}{2}\gamma_x - i\nu''$$

$$\begin{aligned} G^*(-i(\nu-\omega_L)) &= \{\tfrac{1}{2}\gamma_x[\tfrac{1}{4}\gamma'\gamma'' - \nu''^2 + \tfrac{1}{4}G_{10}^2] - \tfrac{1}{2}\nu''^2(\gamma'+\gamma'') + \tfrac{1}{4}G_3^2\tfrac{1}{2}\gamma''\} \\ &\quad + i\{[\tfrac{1}{4}\gamma'\gamma'' - \nu''^2 + \tfrac{1}{4}G_{10}^2] + [\tfrac{1}{4}\gamma_x(\gamma'+\gamma'')] + \tfrac{1}{4}G_3^2]\}\nu'' \\ &= C + iD \end{aligned} \qquad (5.D.132a)$$

$$\tilde{\varepsilon}_{10}(0) = \frac{2\bar{\Gamma}_{10,0}}{(C^2+D^2)}\Bigg[\{C(\tfrac{1}{4}\gamma_{[illegible]} \cdots + \tfrac{1}{4}G_{[illegible]}) + D\omega_L\tfrac{1}{2}(\gamma' + \gamma_{[illegible]})\}$$

$$- \frac{\tfrac{1}{4}[illegible]\;\tfrac{1}{4}G_3^2}{(A^2+B^2)}\{(CA+DB)\tfrac{1}{2}\gamma_{31} + \omega_L(CB-DA)\}$$

$$- \frac{\tfrac{1}{4}G_{10}^2}{(A^2+B^2)}\{[\tfrac{1}{2}\gamma_8((CA+DB) - \nu''(CB-DA)]$$

$$\times[\tfrac{1}{4}\gamma_{10}\gamma_{31} - \omega_L^2 + \tfrac{1}{4}G_{10}^2]$$

$$+\omega_L\tfrac{1}{2}(\gamma_{10}+\gamma_{31})[(CA+DB)\nu'' + (CB-DA)\tfrac{1}{2}\gamma_8]\}\Bigg] \qquad \textbf{(5.B.132b)}$$

$$- \frac{2\,\tfrac{1}{4}G_{10}^2\,\tfrac{1}{4}G_3^2}{(C^2+D^2)(A^2+B^2)}\bar{\Gamma}_{11}\Bigg[\{(CA+DB)\tfrac{1}{2}(\gamma_{31}+\gamma_{10}) + \omega_L(CB-DA)\}$$

$$+\{\tfrac{1}{2}\gamma_8((CA+DB) - \nu''(CB-DA)\}\Bigg]$$

$$- \frac{2\,\tfrac{1}{4}G_{10}^2}{(C^2+D^2)(A^2+B^2)}\bar{\Gamma}_{88}\Bigg[\tfrac{1}{4}G_3^2\{(CA+DB)\tfrac{1}{2}(2\gamma_{10}+\gamma_{31}) + 2\omega_L(CB-DA)\}$$

$$+\{[\tfrac{1}{2}\gamma_8((CA+DB) - \nu''(CB-DA)]$$

$$\times[\tfrac{1}{4}\gamma_{10}\gamma_{31} - \omega_L^2 + \tfrac{1}{4}(G_{10}^2 - G_3^2)]$$

$$+\omega_L\tfrac{1}{2}(\gamma_{10}+\gamma_{31})[(CA+DB)\nu'' + (CB-DA)\tfrac{1}{2}\gamma_8]\}\Bigg]$$

Now we have yet to solve $G(s) = [g_1(s)g_2(s) + \tfrac{1}{4}G_{10}^2]\,g_3(s) + \tfrac{1}{4}G_3^2 g_2(s)$

where
$$\left.\begin{aligned} g_1(s) &= s + \tfrac{1}{2}\gamma' - i\omega' \\ g_2(s) &= s + \tfrac{1}{2}\gamma' - i(\omega' - \Delta_{10,3}) \\ g_3(s) &= s + \tfrac{1}{2}\gamma_8 - i(\omega' + \Delta_{31}) \\ \omega' &= \omega_8' - \omega_1' \end{aligned}\right\} \qquad \textbf{(5.B.133)}$$

This is simply the complex conjugate of $F(s)$, hence the roots are

$$\left.\begin{aligned} s_1 &= a - ib = x_1 - ix_2 = (\alpha+\beta)^* - \tfrac{1}{3}\{\tfrac{1}{2}(\gamma'+\gamma''+\gamma_8) - i(3\omega' + \Delta_{31} - \Delta_{10,3})\} \\ s_2 &= c - id = x_3 - ix_4 = -\tfrac{1}{2}\{(\alpha+\beta)^* - i\sqrt{3}(\alpha-\beta)^*\} - \tfrac{1}{3}\{\tfrac{1}{2}(\gamma'+\gamma''+\gamma_8) - i(3\omega' + \Delta_{31} - \Delta_{10,3})\} \\ s_3 &= e - if = x_5 - ix_6 = -\tfrac{1}{2}\{(\alpha+\beta)^* + i\sqrt{3}(\alpha-\beta)^*\} - \tfrac{1}{3}\{\tfrac{1}{2}(\gamma'+\gamma''+\gamma_8) - i(3\omega' + \Delta_{31} - \Delta_{10,3})\} \end{aligned}\right\} \qquad \textbf{(5.B.134)}$$

So if we use the same approximations

(i) $\Delta_{31} = \Delta_{10,3} = 0$

(ii) $\gamma_{31} >> \gamma_{10}$

(iii) $G^2 >> \gamma_{31}^2$

then
$$\left.\begin{aligned} s_1' &= a - ib = -\tfrac{1}{3}\{\tfrac{1}{2}(\gamma'+\gamma''+\gamma_8) - i(3\omega' + \Delta_{31} - \Delta_{10,3})\} \\ s_2' &= c - id = -\tfrac{1}{6}\{(\gamma'+\gamma''+\gamma_8) - i[2(3\omega' + \Delta_{31} - \Delta_{10,3}) + 3G]\} \\ s_3' &= e - if = -\tfrac{1}{6}\{(\gamma'+\gamma''+\gamma_8) - i[2(3\omega' + \Delta_{31} - \Delta_{10,3}) - 3G]\} \end{aligned}\right\} \qquad \textbf{(5.B.135)}$$

and hence when $\Delta_{31} = \Delta_{10,3} = 0$, the three peaks occur at

$$\nu_1 = \omega_L + \omega' \qquad = \omega_L - \omega_8' + \omega_1' \qquad = \omega_3' - \omega_8' + \omega_1'$$

$$\nu_2 = \omega_L + \omega' + \tfrac{1}{2}G = \omega_L - \omega_8' + \omega_1' - \tfrac{1}{2}G = \omega_3' - \omega_8' + \omega_1' - \tfrac{1}{2}G \;.$$

$$\nu_3 = \omega_L + \omega' - \tfrac{1}{2}G = \omega_L - \omega_8' + \omega_1' + \tfrac{1}{2}G = \omega_3' - \omega_8' + \omega_1' + \tfrac{1}{2}G$$

i.e. at

$$\left.\begin{aligned} \nu_1'' &= 0 \\ \nu_2'' &= -\tfrac{1}{2}G \\ \nu_3'' &= \tfrac{1}{2}G \end{aligned}\right\} \qquad (5.B.136)$$

by half widths $\tfrac{1}{6}(\gamma' + \gamma'' + \gamma_8)$.

Thus we see that the shape of the spectral profile is essentially the same as that for transitions between levels 8 and 1, though we cannot tell the relative heights of the peaks unless we find specific values for $\bar{\rho}_{11}$, $\bar{\rho}_{33}$, $\bar{\rho}_{10,10}$ and $\bar{\rho}_{88}$.

C. Discussion

In all the approximations we have considered we have taken as our first assumption that both fields are in EXACT resonance with the corresponding atomic transitions. In this case we should have absorption followed by emission and NOT scattering since the latter depends on NEAR resonance conditions. Nevertheless, since values of $\Delta_{3,1}$ and $\Delta_{10,3}$ are generally very small, though usually $\Delta_{10,3} > \Delta_{31}$, we can assume that neglecting them is a reasonable assumption and so our theory could also account for scattering. The type of process that occurs will also depend on the spread of frequencies in the incident beam, as explained by Heitler[40]. If the spread of frequencies is large, i.e. a continuous spectrum, then absorption and emission occur as two independent processes. If, on the other hand, the spread is < the natural line width then scattering occurs (cf. P.201ff Heitler). In fact we assume the quantised part of the em field which is responsible for the radiative decay to have a continuous spectrum and replace $\sum_{\rho}$ by an integral. This further supports the idea that what we are considering is absorption followed by subsequent emission. On the other hand we consider the driving fields either (i) to be classical monochromatic fields or (ii) to be single modes of the quantised field. In either case each field has a single frequency and so their spectral profiles are infinitely sharp and hence very narrow in comparison with the natural emission lines of the levels, notably those of levels 10 and 8 with which we are concerned. In fact, such high levels have fairly narrow line widths, as we have assumed under the various approximations, but they do have finite ones. Furthermore, in order for our theory to account for dispersion and Ramaneffect, the latter of which occurs in the K-atom and results when the final state of the atom $\neq$ the initial state and incoherent scattering occurs, we should have included the term in the Hamiltonian $\sim A^2$, as pointed out by Heitler(40) (P. 190).

There are a variety of multiphoton processes which can occur in the K-atom depending on the tuning conditions for ω_L and ω_s. We shall point out those involving levels, 1, 3, 10 and 8, namely processes (i) - (iv) mentioned in Sec. A.

(i) Stimulated 2-photon Raman effect (ref. 13)

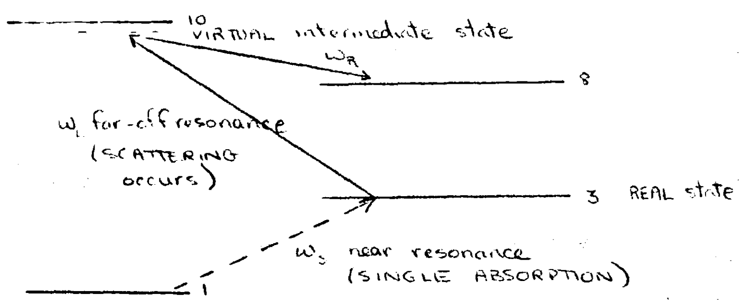


Fig. 5.C.1

This is one of the methods by which level $5P_{3/2}$ (8) can become populated. Frequency, ω_L, of laser emission is Raman scattered after absorption of ω_s, level $4P_{3/2}$ (3) being the initial state and $5P_{3/2}$ or $5P_{1/2}$ (8 or 7) the final atomic state. According to ref. (13) this results in an intense stimulated electronic Raman emission line at 2720.6 cm^{-1} or 3.6μ, i.e. I.R. for all four Raman liquids used. In ref. 11 they point out that the atomic coefficient determining the strength of the coupling between levels 10 and 8 is LARGER for scattering of 2720 cm^{-1} than for 10 to 8 emission but that the 4-photon process with strong 2720 cm^{-1} radiation is entirely absent. This is given as strong evidence in favour of mechanism (iv) 3 photon Raman scattering.

N.B. since level 10 itself does not become populated in this case, we can consider $\bar{\rho}_{10,10} = 0$ in equation (5.B.132b).

(ii) 4 photon parametric coupling (ref. 17)(more correctly termed 4-frequency parametric coupling (ref. 10))

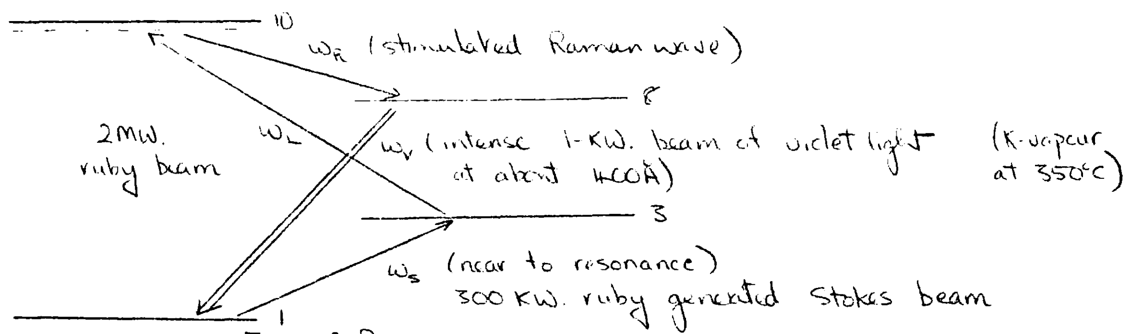


Fig. 6.C.2

In this case the frequency of each photon is close to resonance with successive energy intervals of potassium (resonance 4-photon process). Lumpkin (in ref. 17) observed an intense coherent beam of violet light when he irradiated K vapour at about 350°C with simultaneous laser pulses and he attributed this to 4-wave parametric interaction, in which two of the waves are the applied laser fields and the third an I.R. wave generated in the vapour by stimulated Raman emission (ref. 13). The parametrically generated violet beam is a doublet centred on $5P_{3/2} - 4S_{\frac{1}{2}}$ (8-1) K emission line and typically had about 20% more intensity in the high frequency component. The measured peak power in the doublet was about 1 kW. He says that as the doublet components usually split off by unequal amounts from the doublet centre this suggests wavefunction modulations by the strong applied laser fields, since the separation varies only when the amount of resonant Stokes power incident on the K-cell is varied. His theory, based on wavefunction modulation, also predicts ω_R to have a doublet structure though this was not observed by Rokni & Yatsiv (ref. 13). The strength of the polarisation at ω_V depends on the Raman material used and is strongest in NB and FNB and relatively weak in NM (ref. 12). Parametric emission at ω_P is not observed for ω_S of NM but appears strongly with ω_S of FNB and weaker, but easily observable, for ω_S of NB.

(iii) Enhanced 2-photon emission (ref. 16)

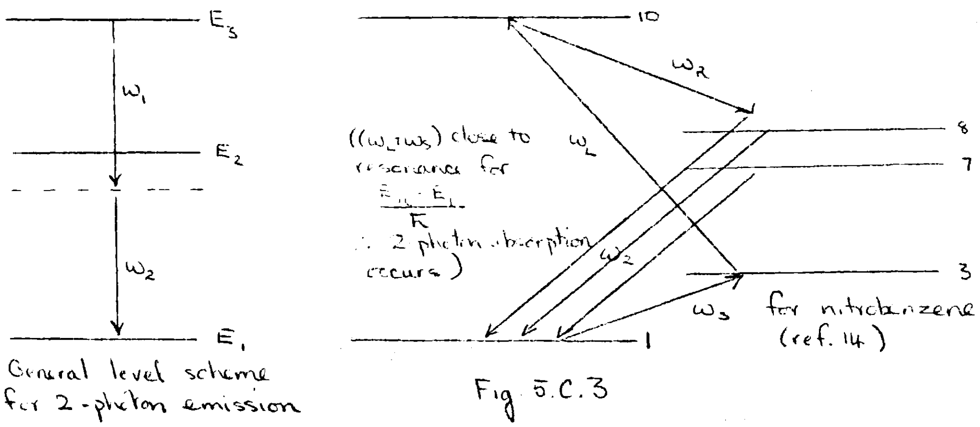


Fig. 5.C.3

State $|10\rangle$ is excited by simultaneous absorption of 2 photons: one at the ruby laser frequency ω_L and the other at the Stokes shifted frequency of stimulated Raman emission in nitrobenzene ω_S (ref. 14). (N.B. in ref. 13 Rokni & Yatsiv observed two additional strong emission lines at 2730.4 and 2749.2 cm^{-1} when using nitrobenzene owing to double quantum absorption to state 10 as well as at ω_R = 2720.6 cm^{-1}.) Under the conditions of (ref. 15) 2-photon excitation of state $|10\rangle$ and atomic Raman radiation in K take place at the same time.

Emission at the complementary frequency ω_2 is 10 cm^{-1} above the 8 - 1 resonance line in K and enhanced BLUE emission at ω_2 = 24730 cm^{-1} satisfies the equation

$$\hbar(\omega_1+\omega_2) = E_{10} - E_1 \quad \text{i.e.} \quad \hbar(\omega_R+\omega_2) = E_{10} - E_1$$

to within one wave number. The doublet structure observed by Lumpkin (ref. 17) in lines for transitions 8 → 1 (and 7 → 1) was attributed by him to wavefunction modulation but Yatsiv, Rokni and Barak (ref. 16) did not observe this structure when the K-vapour pressure was sufficiently low. The condition for 2-photon excitation of level 10 is not satisfield for 1-bromonaphthalene, 1-chloronaphthalene or 1-methyl-naphthalene (ref. 13).

(iv) <u>3-photon Raman scattering</u> (ref. 11)

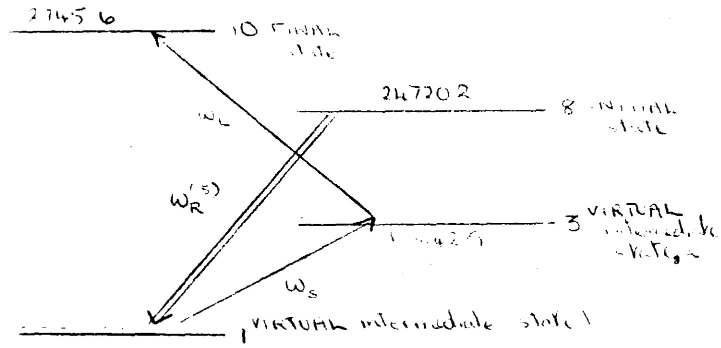


Fig. 5.C.4

3-photon Raman effect between states $|8\rangle$ and $|10\rangle$ of potassium.

NB requires near resonance conditions in two of the 3 photon processes and these are satisfied for ω_L & ω_s.

One of the three ways in which a 3-photon transition can take place between states of different parities is when we have the absorption of two photons and the <u>simultaneous</u> emission of a third by a process similar to 2-photon Raman scattering. It has been observed in the scattering of ruby (ω_L) and a stimulated molecular Stokes radiation (ω_s) in potassium atoms (see above scheme). The SRS radiation is due to β -methyl-naphthalene with ω_s = 13015 cm^{-1}. A blue line at

$\omega_R^{(3)}$ = 24681 cm^{-1} is emitted in potassium satisfying

$$\omega_R^{(3)} = \omega_L + \omega_s - 1/\hbar\,(E_{10} - E_8)$$

This can either be explained by 4-photon parametric coupling (ii) or (iv) 3-photon Raman-type scattering in which and are absorbed, a photon of frequency $\omega_R^{(3)}$ is simultaneously emitted and a K-atom already in state $|8\rangle$ is excited to higher state $|10\rangle$. Yatsiv, Rokni and Barak (ref. 11) put forward evidence, given at the beginning of this section, for the latter-type process even though both processes satisfy the same equation and conform with the same parity requirements. They finally mention that diverse multiphoton processes are observed in free atoms and many more will be found when new techniques for high-power lasers yield a richer choice of frequencies.

Before discussing our results it is necessary also to remember that experimentally both ruby and SRS radiations are linearly polarised in the same plane. Barak and Yatsiv (ref. 10) point out that parametric couplings give rise to emissions polarised in both p and s directions (i.e. // and $\perp$ directions). We will recall that in our analysis we have summed over all possible polarsiations (ξ_σ) of the incident beam.

In ref. 12 it is pointed out that assignment of an observed emission line is often equivocal and may be due to more than one multiphoton process.

When K-vapour is irradiated by the ruby frequency ω_L and by the SRS frequency ω_S of nitrobenzene (NB) resonance conditions are satisfied for 2-photon absorption,

$$\hbar(\omega_L+\omega_S) \doteq E_{10} - E_1$$

and in this case the frequency ω_λ = 24730 cm^{-1}, for emission close to the 10 $\rightarrow$ 8 violet resonance doublet in K, assigned to an enhanced 2-photon emission from level 10 and primed by electronic SRS infra-red emission at ω_R = 2720 cm^{-1}, coincides with $\omega_P = \omega_L + \omega_S - \omega_R$ the frequency of the 4th wave in a 4-photon parametric cycle. But this does not hold when other values of ω_S, not satisfying the 2-photon resonance condition, are used. ω_λ and ω_P shift by equal amounts, $\Delta\omega$ in opposite directions on altering ω_L by $\Delta\omega$. Also, whereas radiation should be collimated and collinear with the incident radiations, since it satisfies the momentum conservation rule $k_L + k_S - k_R - k_P = 0$, ω_λ radiation should not be collimated, since it is a semi-spontaneous process and is therefore expected to be emitted in all directions consistent with the radiation pattern of a classical dipole. In fact, ω_λ is often collimated and collinear with the incident radiations at ω_L and ω_S.

Since $$\omega_2 = \frac{E_{10} - E_1}{\hbar} - \omega_R$$

this emission must be STIMULATED and not enhanced 2 photon emission although in certain circumstances the radiation is uncollimated and truly semi-spontaneous or enhanced. Another difference between radiations and ω_P is that the spectral width of ω_P, $\Delta\omega_P$, is BROAD due to the broadness of SRS radiation ω_S, whereas $\Delta\omega_2$ is SMALL (1 cm^{-1}) matching the width of ω_L.

The main experimental results are that an intense IR line is seen at ω_R = 2720.6 cm^{-1} due to stimulated electronic Raman emission from level 10 ⟶ 8 and an intense 1 kW coherent beam of blue or violet light at ω_B = 24681 cm^{-1} for ω_S in BMW (ω_S = 13015 cm^{-1}) due to 3-photon Raman scattering or ω_B = 24730 cm^{-1} for ω_S in NB (ω_S = 13054 cm^{-1}) which has a doublet structure and is centred around the 8 - 1 K emission line (the high frequency component is 20% more intense but the components are usually split by equal amounts from the doublet centre) and is caused by stimulated 2-photon emission following on from 2-photon excitation of level 10.

In ref. 28 the authors find that for ω_L = 14399 cm^{-1} and ω_S = 13054 cm^{-1} (in NB) the 10 ⟶ 8 line (λ = 4044Å) has a narrow "dip" with centre coinciding with the transition frequency and that its total width, including both components increased with increasing laser power:-

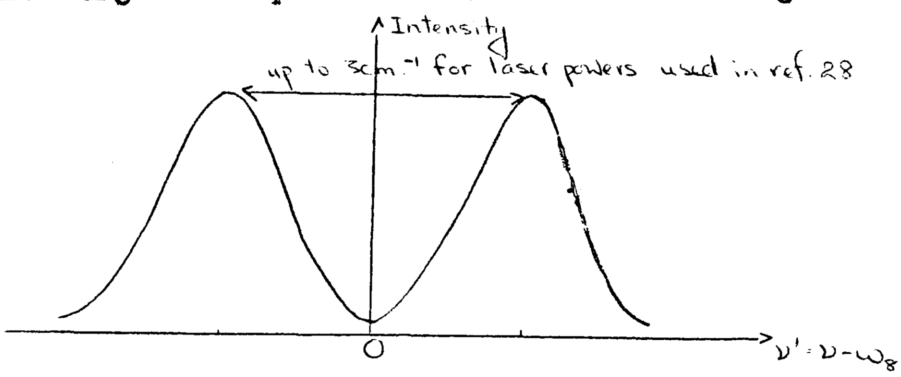


Fig 5.C.5

We have a central peak also since we include the coherent part in our final expression and this corresponds to resonance emission from state $|8\rangle$. In Chapter VII we point out that the central peak results from the fact that we consider an open system, under stationary conditions. In both approximations (v) and (vii) we find the outer peak separation increases for increasing laser field since it is given by G_{10} where G_{10} is related to the laser power and is generally larger than G_3 (in the ratio 20:3, i.e. 2 MW to 300 kW).

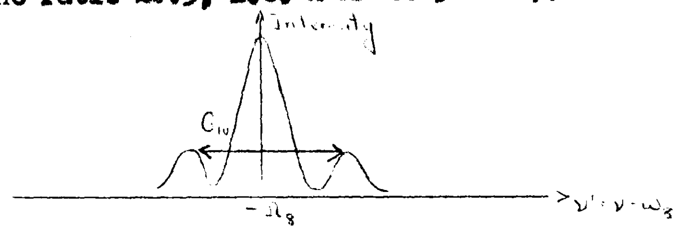


Fig 5C.6

According to ref. 12 the 2-photon emissions are stimulated and their intensities are strikingly comparable with that of the resonance emission from the state $|8\rangle$. Our calculations show the ratio of the intensity of resonance emission: 2-photon emissions for case (v) to be

$$\frac{\left\{\gamma_8 \tfrac{1}{4} G_{10}^2 + \gamma'' \tfrac{1}{4} G_3^2\right\}}{\left\{\left(\frac{3\gamma_8 + \gamma_{31} + \gamma_{10}}{6}\right)^2 + \tfrac{1}{4} G_{10}^2\right\}} : \frac{\tfrac{1}{4} G_3^2 (\gamma'' + \gamma')}{\left\{\left(\frac{3\gamma_8 + \gamma_{31} + \gamma_{10}}{6}\right)^2 + G_{10}^2\right\}}$$

and for case (vii)

$$\frac{\tfrac{1}{2}\gamma_8 \left\{\left[\tfrac{1}{4}(\gamma_8+\gamma_{31})(\gamma_8+\gamma_{10}) + \tfrac{1}{4}G_{10}^2\right]^2\right\} + \tfrac{1}{8} G_3^2 \left\{\tfrac{1}{4}(\gamma_8+\gamma_{10})\left[(\gamma_8+\gamma_{31})(\gamma_8+\gamma_{10}) + G_{10}^2\right]\right\}}{\left\{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)^2 + \tfrac{1}{4}G_{10}^2\right\}}$$

$$: \frac{\tfrac{1}{2}\gamma_8 \left\{\left[\tfrac{1}{4}(\gamma_8+\gamma_{31})(\gamma_8+\gamma_{10})\right]^2 + \left[\tfrac{1}{2}(2\gamma_8+\gamma_{31}+\gamma_{10})\right]^2 + \tfrac{1}{4}G_{10}^2\right\} + \tfrac{1}{4}G_3^2\left\{\tfrac{1}{4}(\gamma_8+\gamma_{10})\left[(\gamma_8+\gamma_{31})(\gamma_8+\gamma_{10}) + G_{10}^2\right] + (\gamma_8+\gamma_{31})\tfrac{1}{4}G_{10}^2\right\}}{\left\{\left(\frac{3\gamma_8+\gamma_{31}}{6}\right)^2 + G_{10}^2\right\}}$$

Thus, in both cases, the ratio shows the intensities to be comparable.

Furthermore in ref. 28 it is stated that observation showed the resonant-transition lines to have NO doublet structure but to have an appreciable width (0.5 - 1.0 cm^{-1}) and to be shifted (by 0.7 - 1.7 cm^{-1}) to the RED side relative to the transition frequency.

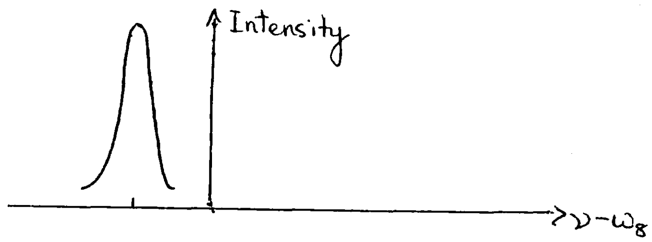


Fig. 5.C.7

We have found also that the resonant-transition lines have no doublet structure and in the case of the 8 ⟶ 1 and the 10 ⟶ 8 transitions the shifts are Ω_8 and $\Omega_8 - \Omega_{10}$ respectively and reduce the frequency of the transition line, i.e. it is shifted to the RED side. The width of the peak in each case for approximations (v) and (vii) is $\Delta_5 = \left(\frac{3\gamma_8 + \gamma_{31} + \gamma_{10}}{6}\right)$ or $\Delta_7 = \left(\frac{3\gamma_8 + \gamma_{31}}{6}\right)$ respectively, the same as that for the side peaks.

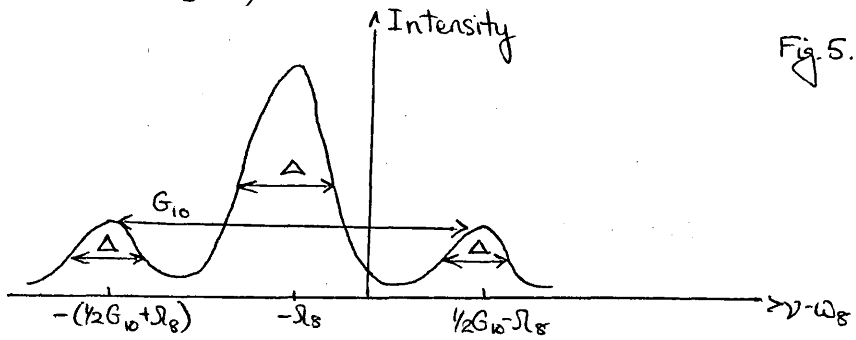


Fig. 5.C.8

The authors in ref. 28 also investigated the absorption band but that is of no interest here. In order to interpret their findings, they assume that the wave function of the atom in the field of the ruby laser is given by [54]

$$\psi = \psi_m \{ A_1 e^{-i(\omega_m + \epsilon_1)t} + A_2 e^{-i(\omega_m + \epsilon_2)t} \} + \psi_n \{ B_1 e^{-i(\omega_n + \epsilon_1)t} + B_2 e^{-i(\omega_n + \epsilon_2)t} \}$$

where

$$\epsilon_{1,2} = \frac{\Omega}{2} \pm \sqrt{\left(\frac{\Omega}{2}\right)^2 + G^2}$$

$$\Omega = \omega - \omega_{mn}$$

$$G = \left| \frac{P_{mn} E}{2\hbar} \right|$$

[i.e. in our case:-

$G_{10} = \frac{2P_{10,1}\hat{e}_L\mathcal{E}_{0L}}{\hbar}$

$G_3 = \frac{2P_{31}\hat{e}_S\mathcal{E}_{0S}}{\hbar}$

The splitting is $\pm\frac{1}{2}G$ where G is G_3 or G_{10} or $\sqrt{G_3^{\,2} + G_{10}^{\,2}}$ depending on which transitions are considered and what approximations are used.]

I.E. one can interpret ψ as the splitting of the nondegenerate states of the atom (diag. below), due to the external field (see refs. 33, 55-57).

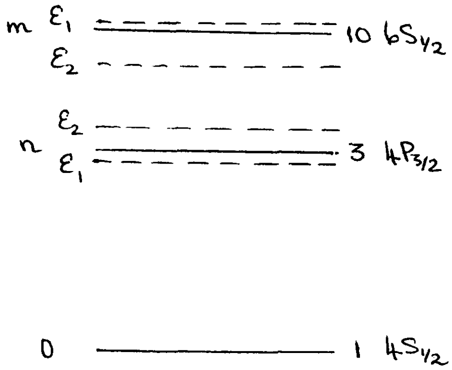


Fig 5C9

They also say that the symmetrical broadening of the 4044Å ($1 \rightarrow 8$ or $4S_{\frac{1}{2}} \rightarrow 5P_{3/2}$ line) line suggests a NEW TYPE OF PERTURBATION of the $5P_{3/2}$ state (state $|8\rangle$). Apparently the cascade population $10 \rightarrow 8$ of the level 8 occurs with the aid of two fields that differ in frequency by an amount $\sim\mathcal{E}_1$ and this should lead to an effect analogous to phase modulation, which is characterised by a broad and symmetrical spectrum. Their general conclusion is thus that the observed effects are connected with the splitting of the atomic levels in the external field. (N.B. frequency shifts Ω_{mn} have nothing to do with the external field and occur in spontaneous emission also).

The authors of ref. 29 also investigate the fine structure of the potassium emission spectrum and $\lambda = 4044$Å ($5P_{3/2} - 4S_{\frac{1}{2}}$ transition ($8 \rightarrow 1$)). We may note that similar conclusions can also be drawn about the $\lambda = 4047$Å ($5P_{\frac{1}{2}} - 4S_{\frac{1}{2}}$ transition ($7 \rightarrow 1$)). They find, as do the authors of ref. 28, a strong dependence on the field intensity (and also on the vapour pressure in which we are not interested here). They also assign the case of the structure to the field splitting of energy levels and this splitting of the ato mic levels in the e.m. field they point out is one of the basic effects of **nonlinear** spectroscopy. Golubev et al. consider the situation where the potassium atom is exposed to a field whose spectrum contains **several** monochromatic lines splitting the line into many components, rather than into TWO, as in a monochromatic field, which is what we consider. They find that the structure of the 8 ⟶1 line is fairly complex and strongly dependent on the laser field power (and vapour pressure). At (low pressures and) WEAK powers the centre of markedly assymetric lines has a single sharp dip but as the powers the laser and SRS radiation increase, the dip widens and then splits successively into 2, 3 or more components:

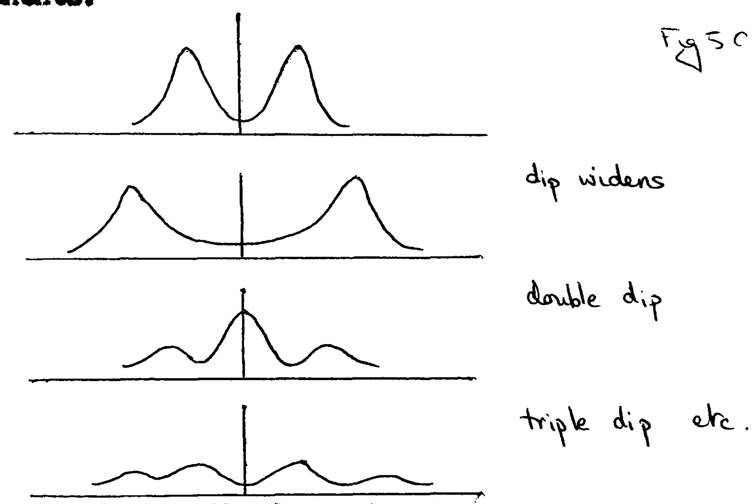


Fig 5C.10

When the laser power is 100 MW the line structure becomes quite unsharp and inhomogeneous along the line height.

Golubev et al. attribute these changes to field splitting of level $5P_{3/2}$ (8) into "quasi-energy sublevels". Apparently, both the laser (ω_L) and the infra red (ω_2 & ω_3) emissions play a significant role in this effect.

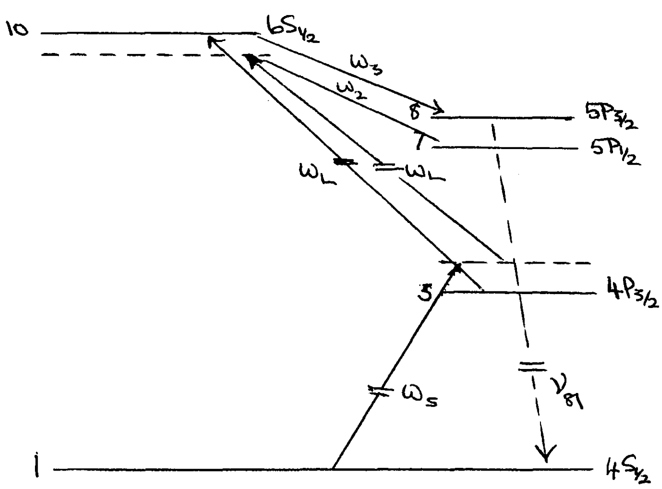


Fig 5C11

They show that the laser field (ω_L) and infrared lines (ω_2, ω_3) are the most intense components of the emission spectrum interacting with K atoms. If only these three dields are taken into account then, according to perturbation theory, the following three processes are possible that can lead to absorption of emission in the vicinity of the frequency $\omega = \epsilon_{10,1}$:

i) 1-quantum absorption of photon $\hbar\omega$, $4S_{\frac{1}{2}} - 5P_{3/2}$ (1 - 8);

ii) 2-quantum absorption of photon $\hbar\omega$ and $\hbar\omega_3$, $4S_{\frac{1}{2}} - 5P_{3/2} - 6S_{\frac{1}{2}}$ (1-8-10);

iii) 3-quantum process (of the Raman scattering type) with absorption of photons $\hbar\omega$ and $\hbar\omega_2$ and emission of a laser-frequency photon $\hbar\omega_L$, $4S_{\frac{1}{2}} - 5P_{3/2} - 6S_{\frac{1}{2}} - 4P_{3/2}$ (1-8-10-3).

Thus the absorption line for the $4S_{\frac{1}{2}} - 5P_{3/2}$ transition is TRIPLY degenerate in terms of the "3-strong-fields model" (ω_L , ω_3 , ω_2). If the intensities are high enough the above processes can no longer be considered independent, the degeneracy is removed, and the line split into three components.

Golubev et al. calculate, by considering only Hamiltonians V_s and V_{sl} perturbatively, that the frequencies of the absorption maxima are given by the following formulas, in our notation, with an accuracy to terms containing squared field amplitudes,

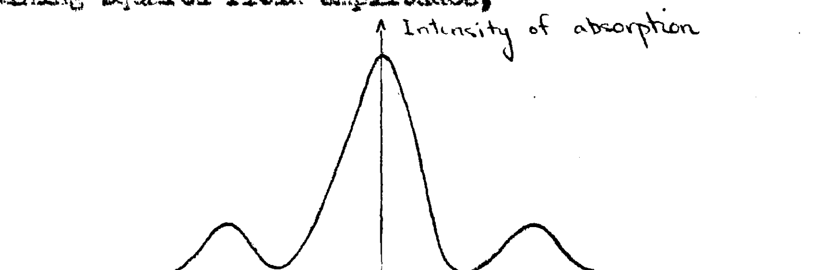


Fig 5.C.12

$$\left.\begin{aligned} \omega^{(1)} &\approx \epsilon_{8,1} + \Omega_L - \Omega_2 \\ \Omega_L &= \omega_L - \epsilon_{10,3} \\ \Omega_2 &= \omega_2 - \epsilon_{10,8} \end{aligned}\right\} \qquad \left.\begin{aligned} \omega^{(2,3)} &\approx \epsilon_{8,1} - \tfrac{1}{2}\Omega_3 \pm G_3 - G_L^2/2\Omega_L \\ \Omega_3 &= \omega_3 - \epsilon_{10,8} \\ G_3 &= \frac{|d_{10,8} E_3|}{2\hbar} \\ G_L &= \frac{|d_{10,3} E_L|}{2\hbar} \end{aligned}\right\}$$

[N.B. In order to convert to our notation

$$\Omega_L \longrightarrow -\Delta_{10,3} - \Omega_{10} + \Omega_{31} \qquad \text{where} \qquad \Delta_{10,3} = \omega_3' - \omega_L \qquad \omega_3' = \omega_3 - (\Omega_{10} - \Omega_{31})$$

$$\Omega_2 \longrightarrow -\Delta_{10,8} - \Omega_{10} + \Omega_8 \qquad \Delta_{10,8} = \omega_{10}' - \omega_2 \qquad \omega_{10}' = \omega_{10} - (\Omega_{10} - \Omega_8)$$

$$\Omega_3 \longrightarrow -\Delta_{10,8} - \Omega_{10} + \Omega_8 \qquad \Delta_{10,8} = \omega_{10}' - \omega_3$$

$$G_L = \frac{|d_{10,3} E_L|}{2\hbar} \longrightarrow \frac{\rho_{10,3} \cdot \hat{e}_{0L} \mathcal{E}_{0L}}{\hbar}$$ owing to the use of different units

$$G_3 = \frac{|d_{10,8} E_3|}{2\hbar} \longrightarrow \frac{\rho_{10,8} \cdot \hat{e}_{03} \mathcal{E}_{03}}{\hbar}$$ " "]

The frequency of the photon generated in Raman scattering has a field shift, i.e.

$$\Omega_2 = \Omega_L + G_L^2/\Omega_L \tag{50}$$

A field shift of line ω_3 is also expected so that

$$\Omega_3 = -G_L^2/\Omega_L$$
$$\therefore \omega^{(1)} \approx \epsilon_{81}$$
$$\omega^{(2,3)} \approx \epsilon_{81} \pm G_3$$

For sufficiently WEAK fields only a single absorption line is absorbed experimentally whereas for ω_3 field sufficiently intense 3 components should be visible. The central line coincided with the location of ϵ_{81} within an accuracy to 0.1 - 0.3 cm^{-1} and the side components were mostly symmetric as required in the last formula for $\omega^{(2,3)}$ though there are some deviations from symmetry reaching 1.5 - 2 cm^{-1}.

The spectral density of the work of field V_{01} is proportional to their expression

$$|V_{81}(\omega)|^2 \; \mathrm{Re}\left\{ \sum_m \sum_{\ell=1}^{3} \frac{C_{3\ell}^{(m)} \; D_{\ell 3}^{(m)}}{[\mu_\ell'' + i(\omega - \epsilon_{81} + \mu_\ell' + m\Omega)]} \right\}$$

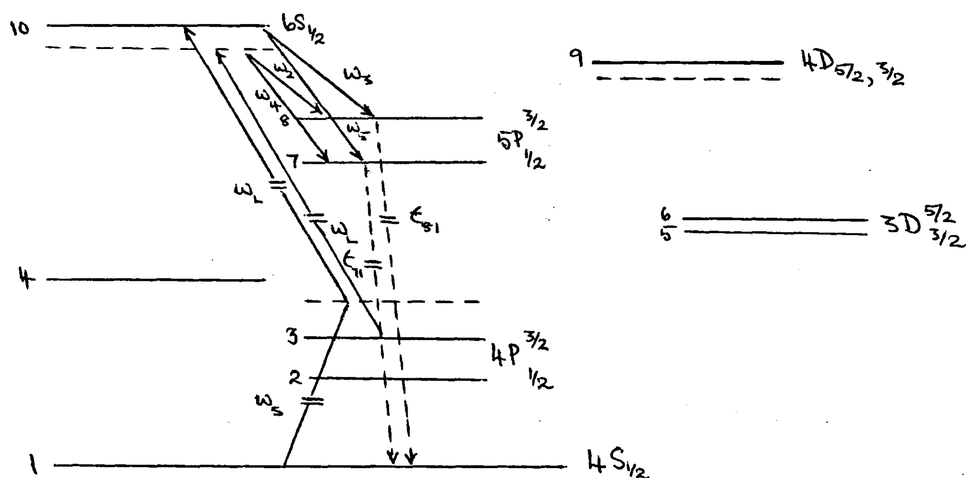
which shows that the absorption spectrum represents a set of equidistant triplets (index m) separately by $|\Omega_2 - \Omega_3|$, triplet m = 0 determining the structure of line 4044Å. In fact, the structure of the absorption coefficient in the short-wave region should simulate that of the $\lambda = 4044$Å line itself. This is true for (pressures sufficiently high and) powers of laser and SR fields sufficiently high, when in fact the components are approximately equally separated.

The fact that the number of observed components of the $\lambda = 4044$ Å absorption line becomes > 3, under certain conditions, is explained by considering not only the three emission frequencies ω_L (laser and ω_2 and ω_3 (resonating with 10 ⟶ 8 transition) but also their analogues for

$6S_{\frac{1}{2}} \longrightarrow 5P_{\frac{1}{2}}$ transition (10 ⟶7) (ω_+ and ω_-)

and emission due to transitions from level $5P_{3/2,\frac{1}{2}}$ to lower levels $5S_{\frac{1}{2}}$ and $3D_{5/2,3/2}$ (8,7 ⟶ 4, 6, 5). In fact if there are five monochromatic fields ω_L, ω_2, ω_3, ω_+ and ω_- the $4S_{\frac{1}{2}} - 5P_{3/2}$ (1 - 8) absorption line can be split off into eight components but analysis of this case would be much more complicated.

Fig.5.C.13



5-strong-fields-model (ω_L, ω_2, ω_3, ω_+, ω_-)

(solid arrows - "strong fields"

dashed arrows - K emission lines in violet spectral range)

Golubev has considered how the violet lines are generated and give three processes:-

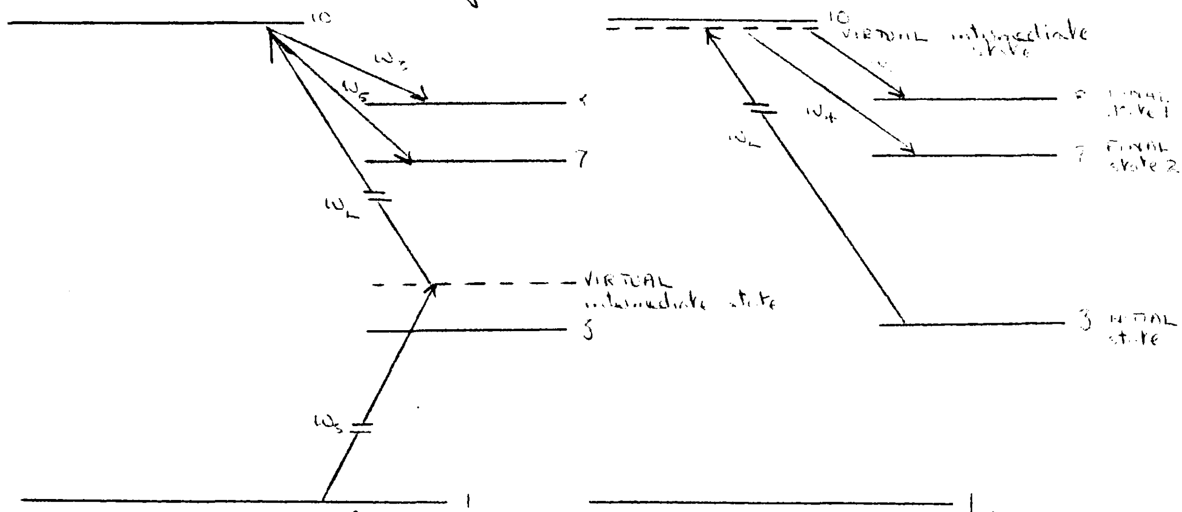
1) 2-quantum absorption of laser emission and SRS (ω_L, ω_s) causing population of level $6S_{\frac{1}{2}}$ and population inversion at transitions $6S_{\frac{1}{2}} - 5P_{3/2,\frac{1}{2}}$ and hence emission and amplification at frequencies

ω_5 and ω_5 .

Hence term 5P is populated and lines $\lambda = 4044/47\,\mathrm{\AA}$ at transition $5P_{3/2,\frac{1}{2}}$ $4S_{\frac{1}{2}}$ is generated.

2) Levels $5P_{3/2,\frac{1}{2}}$ are populated by Raman scattering (ω_2, ω_4) of laser emission ω_L in which $4P_{3/2}$ is the initial and $5P_{3/2,\frac{1}{2}}$ the final atomic state.

Fig. 5.C-14



2-quantum absorption of (ω_5, ω_L) & emission of (ω_5, ω_5) (2 separate processes)

Raman scattering (ω_2, ω_4) of ω_L (1 process)

3) 4-photon scattering ("light-by-light scattering") $\omega_5 + \omega_L \longrightarrow \omega_3 + \omega$

This explains the fact that violet emission ($8 \rightarrow 1$ and $7 \rightarrow 1$) is sharply directed and orientated exclusively in the same direction as ω_5 and ω_L . This process is particularly effective when the following condition for wave vectors is satisfied (see [59] for example)

$$\underline{k}_5 + \underline{k}_L = \underline{k}_3 + \underline{k}$$

This explains the singular direction of the emission and so this process should be significant at least in the initial generation stages of infra-red and violet emissions. On the other hand, the cascade mechanism probably amplifies the light resulting from the 4-photon scattering. In fact, Goluber et al. conclude that the role of this process is only reliable in relation to the broad "short-wave" line ω_{g5} , whereas its role in the remaining regions of the spectrum requires further experimental research.

Nevertheless the field origin of absorption line splitting at the

$4S_{\frac{1}{2}} - 5P_{3/2,\frac{1}{2}}$ transition is well established. Also they note that in the perturbation theory approach processes involving different numbers of photons are regarded as different processes whereas this distinction becomes groundless for sufficiently high emission intensity. E.g. in the case of the $\lambda = 4044$ Å absorption line 1-, 2- and 3-photon processes are "mixed-up" in the intense resonance field. Thus it is better instead to talk of a SINGLE process of violet absorption in which the line structure is interpreted as a result of field splitting of atomic levels and "this approach is in full accord with the spirit of nonlinear spectroscopy"(60). Golubev et al regard their data as an experimental verification of one of nonlinear spectroscopy's main theses.

The other conclusion of our calculation, regarding the structure of the $6S_{\frac{1}{2}} - 5P_{3/2}$ line (10 - 8 line), remains to be discussed with reference to experimental data. The doublet structure which we find (plus of course the resonance peak at $\nu = \omega'_{10}$) agrees with Lumpkin's conclusion(17) that the infra-red Raman emission (3.63 μ) should have doublet structure, though he points out that this has not been observed(13). Lumpkin's analysis is criticized by Golubev et al.(28) who claim that Lumpkin's model is inadequate since it assumes the nitrobenzene SRS radiation to be monochromatic and STRONG and all remaining fields to be WEAK. Lumpkin concludes that the splitting of the resonance level is real if the SRS spectral width is smaller than the splitting whereas in reality the SRS spectrum is BROAD, as noted earlier. Golubev et al. therefore say that Lumpkin's conclusions are invalid though his theory does in fact account for the doublet structure of the violet lines.

They are, in fact, two possible frequencies for the emission in the 10 – 8 transition, namely ω_3 and ω_2, but which occurs depends on the resonance conditions for ω_s and ω_L. For ω_3 to be emitted we require:

1) $\omega_s + \omega_s = \omega_s' + \omega_{10}' = \epsilon_{10,1} - \Omega_{10} \quad \therefore \Delta_{10,3} \neq 0 \ \& \ \Delta_{31} \neq 0$
$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \text{or } \Delta_{10,3} = 0 \ \& \ \Delta_{31} = 0$

whereas for ω_2 to be emitted, we require

2) $\omega_s = \omega_1' = \omega_1 - \Omega_{31} \quad \therefore \Delta_{31} = 0$
$\quad\ \omega_L \simeq \omega_3' = \epsilon_{10,3} - (\Omega_{10} - \Omega_{31}) \quad \therefore \Delta_{10,3} = 0$

In case (1)

$$\bar{\rho}_{33} = 0$$

and in case (2)

$$\bar{\rho}_{10,10} = 0$$

Since we are considering the overall effect of both processes and Δ's are negligible we can assume that we should obtain a spectral profile:

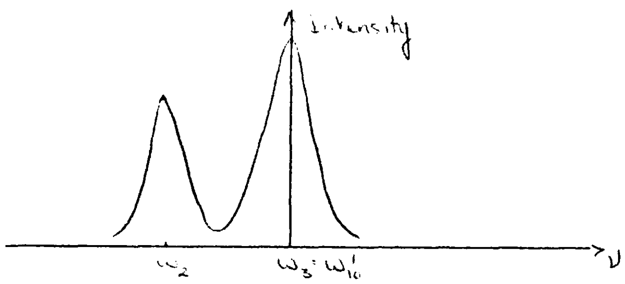


Fig 5C 15

According to ref. 23 the value of ω_2 is determined by $\mathcal{E}_2$, i.e.

$$\omega_2 = \omega_{10} - \mathcal{E}_2$$
$$= \omega_{10} - \left\{ \Omega/2 - \sqrt{(\Omega/2)^2 + G^2} \right\}$$

where $\Omega = \omega - \omega_{10}$

and $G = \left| \frac{p_{10,8} E}{2\hbar} \right|$

If ω_L has a spread of values of the order of the field splitting of level 10 another emission line would be expected on the high frequency side of ω_2. The criterion for this to occur would be

$$\Delta_{10,3} \sim \mathcal{E}_1 - \mathcal{E}_2$$

which for moderate powers of laser and SRS fields $G \ll |\Omega|$ means

$$\Delta_{10,3} \sim 2G^2/|\Omega|$$

which is very small indeed. The origin of the two outer components of the spectrum can thus be assigned to the field splitting of level 10 and the central component to the resonant transition between levels 10 and 8 when the 2-quantum absorption condition is satisfied. The separation between the outer peaks is the same as for transitions between levels 8 and 1 and so should also be observable unless their intensities are very low. We can therefore only conclude that the laser field is for all practical purposes completely monochromatic. The spectral profile for transitions between levels 8 and 1 must therefore be governed by the spectral width of the SRS radiation which is known to be BROAD and so the latter can result in > 1 component for the resulting $8 - 1$ spectrum.

CHAPTER VI

THE HYDROGEN ATOM DRIVEN BY AN EXTERNAL FIELD COUPLING LEVELS $2S_{\frac{1}{2}}$ AND 3P

For the purpose of our calculations concerning the hydrogen atom we shall take into account the following levels: $1S_{\frac{1}{2}}$ (ground level), $2P_{\frac{1}{2}}$, $2S_{\frac{1}{2}}$ (metastable), $2P_{3/2}$ and 3P, i.e. levels up to and including excited state 3P. We shall refer to these levels by numbers 1 - 5 as shown below

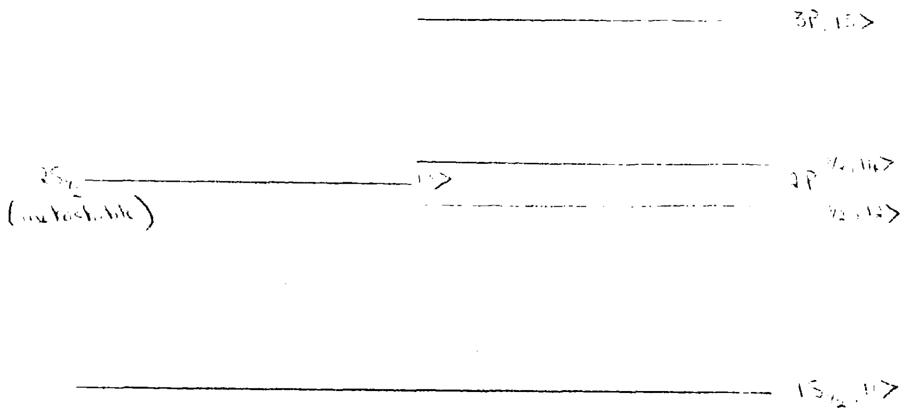


Fig. 1

A. (i) Equations of Motion

The equations of motion for a general 5-level atom are, according to equations (4.A.19), (4.A.20) of Chapter IV,

$$\left.\begin{aligned}\dot{P}_{mn} = & -\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\left\{\tfrac{1}{2}(\gamma_{ni}+\gamma_{mj}) + i(\epsilon_{mn} - \lambda_{ni} + \lambda_{mj})\right\}P_{mn} \\ & - i\sum_{l=n+1}^{5} P_{ml}\, q_{ln} \qquad + i\sum_{l=1}^{n-1} P_{ml}\, q_{nl} \\ & - i\sum_{j=1}^{m-1} q^{+}_{mj} P_{jn} \qquad + i\sum_{k=m+1}^{5} q^{+}_{km} P_{nk}\end{aligned}\right\} \qquad (6.A.1)$$

$$\left.\begin{aligned}\dot{P}_{mm} = &-\sum_{i=1}^{m-1}\left\{\gamma_{mi} P_{mm} - i\left(P_{mi} q_{mi} - q^{+}_{mi} P_{im}\right)\right\}\\ &+\sum_{j=m+1}^{5}\left\{\gamma_{jm} P_{jj} - i\left(P_{jm} q_{jm} - q^{+}_{jm} P_{mj}\right)\right\}\end{aligned}\right\} \quad (6.A.2)$$

where the Hamiltonian is given by:-

$$H = \hbar\sum_{x=1}^{5}\epsilon_x P_{xx} + \hbar\sum_{l\sigma}\omega_l a^{+}_{l\sigma} a_{l\sigma} - \hbar\sum_{y=z+1}^{5}\sum_{z=1}^{4}\sum_{l,\sigma} g_{l\sigma,yz}\left(P^{+}_{zy} a_{l\sigma} + a^{+}_{l\sigma} P_{zy}\right) \quad (6.A.3)$$

Since we are always concerned with dipole transitions we find that in the present case

$$\left.\begin{aligned}x_{13} &= 0\\ x_{24} = x_{25} &= 0\\ x_{45} &= 0\end{aligned}\right\} \quad (6.A.4)$$

where $x = q, \gamma, \Omega$ and the equalities are true for reversed subscripts and, in the case of q, for the hermitian conjugate also.

We shall be interested in transitions between

a) states 3P ($|5\rangle$) and $1S_{\frac{1}{2}}$ ($|1\rangle$)

and b) states 3P ($|5\rangle$) and $2S_{\frac{1}{2}}$ ($|3\rangle$)

when a strong driving field E_L couples levels 3P and $2S_{\frac{1}{2}}$, i.e. levels 5 and 3, the field being supplied by ruby laser radiation which is known to be in approximate resonance with the atomic transition $3P - 2S_{\frac{1}{2}}$ (5-3).

A(ii) Review of relevant papers

We shall compare our results with those of Zernik[32] and Rautian and Sobelman[33]. Zernik's concern is the optical quenching of metastable hydrogen by means of a high-frequency e.m. wave and the theories he uses are the quantum electrodynamical perturbation theory and a strong signal theory. If the atom is isolated, and in the $2S_{\frac{1}{2}}$ state, then it is metastable (mean lifetime $\frac{1}{7}$ sec.) since it can decay only by 2-photon emission[61 - 63]. It is well known that a transition to the ground state can be easily induced by a d.c. electric field [64].

When the 2S state is quenched by a WEAK d.c. field what occurs can be explained as in Zernik's paper: In the presence of the field there are two stationary states and the wave functions of each of these are a linear superposition of those for the 2S and $2P_{\frac{1}{2}}$ states ($|3\rangle$ and $|2\rangle$)

i.e.
$$\left.\begin{aligned} \psi_A &= A_3\psi_3 + A_2\psi_2 \\ \psi_B &= B_3\psi_3 + B_2\psi_2 \end{aligned}\right\} \qquad (6.A.5)$$

The decay probability comes from the admixture of the $2P_{\frac{1}{2}}$ state (2). For fields $\ll$ 475 V/cm. the 2 states are ALMOST PURE 2S and $2P_{\frac{1}{2}}$. But for fields $>$475 V/cm. a saturation condition is approached where the MIXING is COMPLETE and both states decay at a rate given by the average of the decay rates of the pure states

i.e. $\frac{1}{2}(\gamma_{21}+\gamma_{32})$.

Since light from a ruby laser contains photons of energy 0.0657 $me^4/\hbar^2$, just slightly less than the energy difference between 3P and 2S states 0.0695 $me^4/\hbar^2$, the most likely process by which optical quenching of the 2S state can occur is by means of a VIRTUAL transition via the 3P state

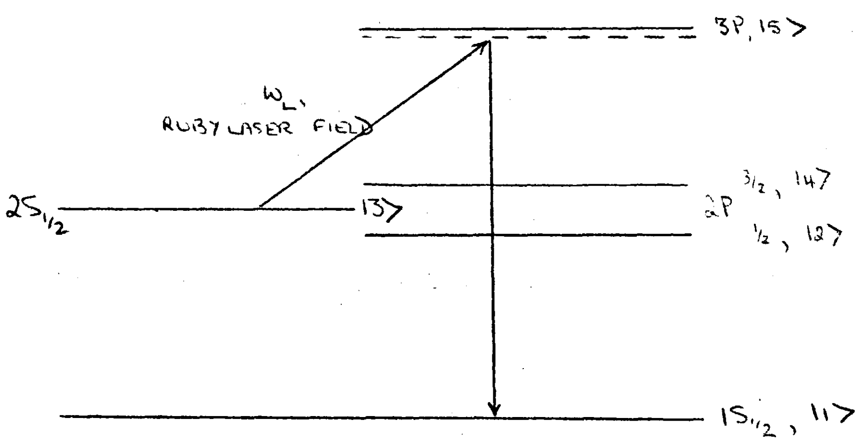


Fig. 6.A.1

If elementary perturbation theory is valid (see criterion given later) this process is a linear and not a non-linear one. The energy of the emitted photon corresponds to $\lambda = 1035\ \mathring{A}$, as determined by the conservation of energy.

At HIGH intensities saturation effects occur and Zernik gives as a reason the fact that the 2S and 3P states become strongly coupled and when this coupling is very strong both states are expected to decay to the ground state at the rate $\frac{1}{2}(\gamma_{21} + \gamma_{32})$ implying that when the intensity is high enough the cross section will start to decrease and will tend eventually to vary inversely with the incident intensity so as to maintain a constant rate

$$\text{i.e.}\quad \sigma \propto \frac{1}{I_0} \tag{6.A.6}$$

Quenching by other states, such as 4P, is possible and this modifies the effect just described.

Zernik calculates the differential cross sections, using perturbation theory, for two types of 2nd order transitions from state $2S_{\frac{1}{2}}$ to $1S_{\frac{1}{2}}$ via an intermediate state and two types of intermediate states in each case. Then he goes on to use the strong signal theory and assumes that as the laser beam is very intense the 2S-3P transition can be treated by semiclassical radiation theory and spontaneous decay from 3P to 2S may be neglected (i.e. $\gamma_{53} = 0$). We shall not neglect this entirely but will show that it is small. He neglects also the antiresonant terms, as do we. He notes finally that the strong-signal theory provides the damping corrections to the perturbation theory. Also he deduces the criterion for the validity of perturbation theory to be

$$|V|^2 << \hbar^2 \left| \tfrac{1}{2}\{(\gamma_{53} + \gamma_{51}) - \gamma_{32}\} + i\Omega \right|^2 \tag{6.A.7}$$

where $V = \langle 5|e\,\underline{E}_L\,\underline{r}|3\rangle = -\underline{E}_L\cdot\underline{D}_{53}$ is the interaction Hamiltonian, and $\Omega = \epsilon_{53} - \omega_L$ where ω_L is the frequency of the laser field. This is the condition also for which higher order terms in the perturbation theory treatment of the 2S to 3P transition are small. In his experiment $\hbar\Omega = 0.0038\,(me^4/\hbar^2) \doteq 0.1$ eV, thus making the saturation effect observable when $|V| \gtrsim 0.1$ eV, i.e. when $E \doteq 10^7$ V/cm. For field strengths $E < 10^7$ V/cm. perturbation theory is valid. Strong signal theory is necessary for higher field strengths.

In Rautian and Sobel'man's paper(33) they treat the effect of a STRONG monochromatic e.m. field of requency close to one of the characteristic frequencies of a system on the spectral composition of the radiation. They consider an atom with 3 non-degenerate levels which could correspond to the dominant energy levels under consideration, i.e.

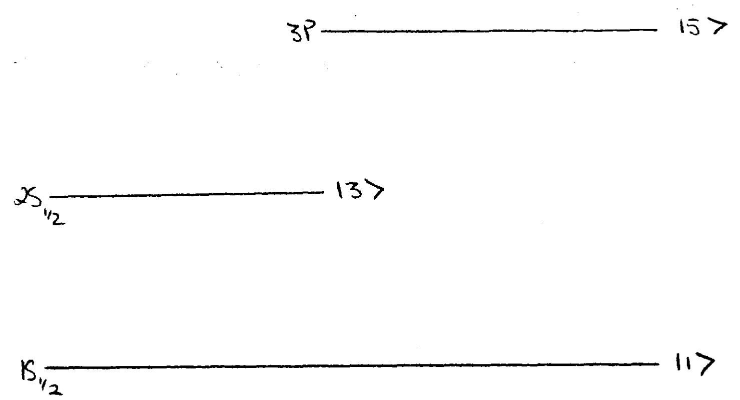


Fig. 6.A.2

They point out that inverted population of levels E5, E3 is possible if the probability of decay of level 3

$$2\gamma_3 = 2\gamma_{32} \gg 2\gamma_{53} \qquad (6.A.8)$$

(the probability of the spontaneous transition 5 → 3) and so they consider this situation, though they do not restrict the decay probability $2\gamma_5$ of level 5. We do implicitly assume $\gamma_{51} \gg \gamma_{32}$

at one stage and in fact γ_{32} is not large. Rautian and Sobelman's calculation cannot truly be compared with that concerning the hydrogen atom. In their case, in contrast with Zernik's, the atom is initially in the level corresponding to our level 5 (and not in level 3 as in Zernik's paper) and the radiation field contains photons n_L, n_μ with frequencies ω_L, ω_μ there being two fields: one strong e.m. field of frequency $\omega_L \sim \epsilon_{53}$ and one weak field of continuous spectrum which is necessary for the calculation of induced emission and absorption at ω_L, ω_μ. They use perturbation theory to find out the characteristics of the radiation in the transition of the atom from an excited state. They discover that for strong fields, i.e. N_L large, the damping curve becomes complex:

$$|a(5,n_L,n_\mu)|^2 = |A_1 e^{-\Gamma_1 t} + A_2 e^{-\Gamma_2 t}|^2 \quad \text{(the aperiodic case)} \qquad (6.A.9)$$

because states 5 and 3 become MIXED, owing to interaction with the field. For still stronger fields

$$|a(5,n_L,n_\mu)|^2 = A\cos^2(\delta t + \psi)e^{-(\gamma_3+\gamma_5)t}$$
$$\text{where } \delta^2 = G^2 - \frac{(\gamma_3-\gamma_5)^2}{4} \qquad (6.A.10)$$

i.e. the time-dependence of this function (the modulus squared of the probability amplitude) represents damped oscillations. They go on to calculate the probability W_L of induced emission of a photon of frequency ω_L and the integrated probability W_μ of spontaneous emission $5 \longrightarrow 3$.

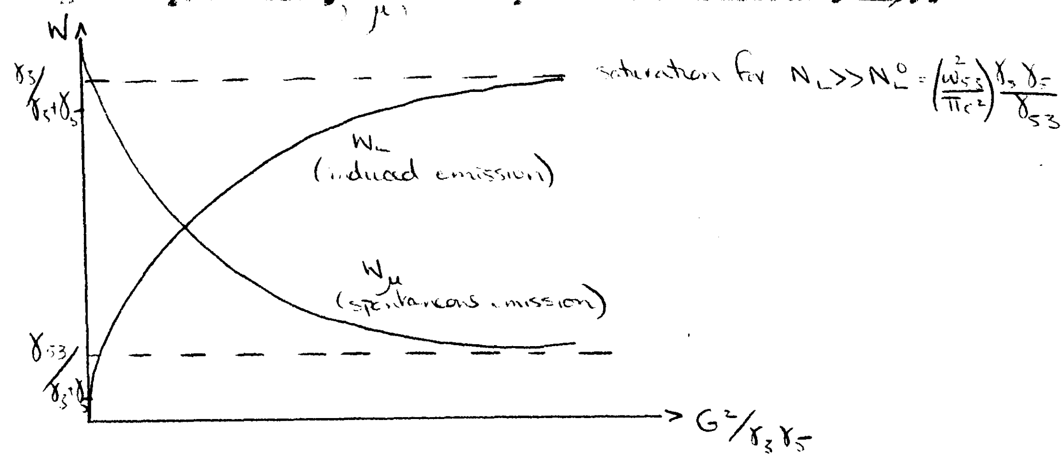


Fig. 6.A.3

In the case when oscillations are damped $G^2 > (\gamma_3 - \gamma_5)^2/4$ the line shape becomes split into 3 components of equal widths, $\Delta = (\gamma_3 + \gamma_5)$, separated by distances $2\delta_1 = \sqrt{G^2 - (\gamma_3 - \gamma_5)^2/4}$

where $G^2 = (\pi c^2 \gamma_L N_L)/\omega^2_{53}$ and N_L is the total number of photons of frequency ν_L in unit volume

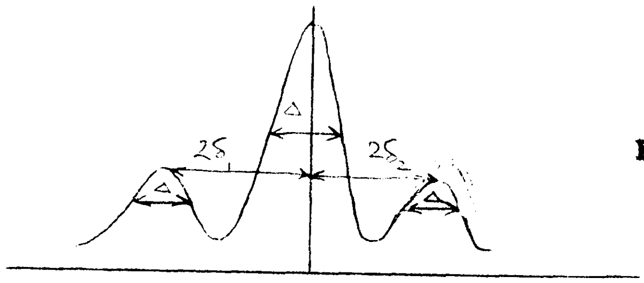
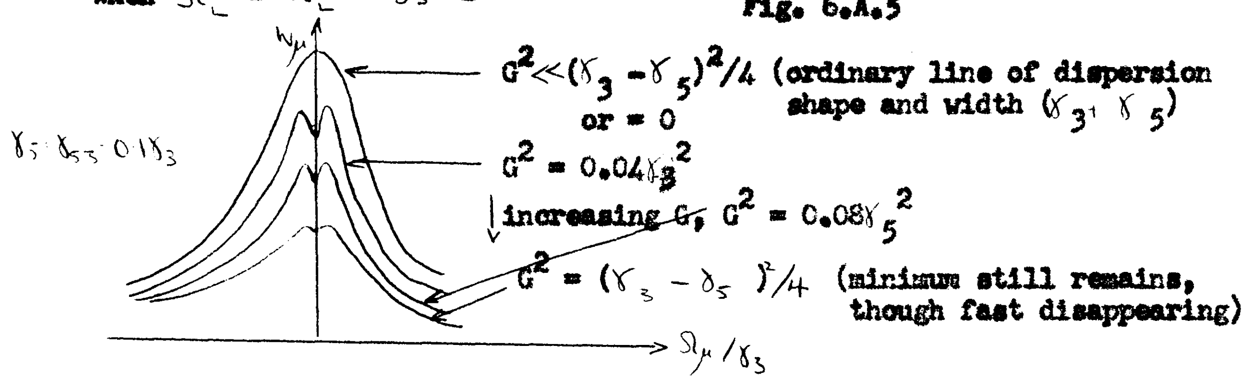


Fig. 6.A.4

In the "aperiodic case" there are no oscillations, but the amplitudes of the different states fall off with different damping constants Γ_1 and Γ_2 leading to a change in the shapes of the emission and absorption lines.

When $\Omega_L = \omega_L - \epsilon_{53} = 0$

Fig. 6.A.5



Probability of spontaneous emission (in units $\gamma_{53}/\pi\gamma_5(\gamma_3 + \gamma_5)$)

W_μ vs Ω_μ/γ_3

When external field increased still further the emission line consists of the 3 components spoken of earlier, the splitting being detectable for $G^2 \gtrsim \gamma_3\gamma_5 \left\{ \frac{3\gamma_3}{4\gamma_5} + \frac{1}{2} + \frac{5\gamma_5}{4\gamma_3} \right\}$

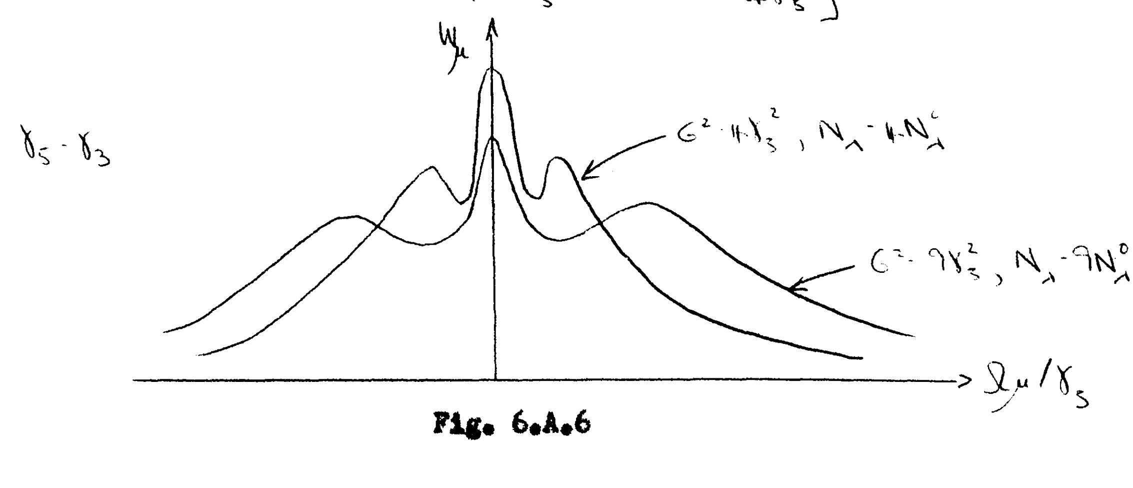


Fig. 6.A.6

Splitting is appreciable only for comparatively strong saturation i.e. for $G^2 >> \gamma_3\gamma_5$

With departure from resonance ($\Delta \neq 0$) the line becomes extremely asymmetrical, the maximum of the ~~one~~ side components approaching ω_{53} and its magnitude rising steeply, whilst the other two terms become smaller and the positions of the maxima go further from ω_{53}.

We shall in fact be considering the case when the atom is initially in level 5, as in Rautian and Sobelman's paper(33) so that our calculations should rightly be compared with theirs, though theirs are not specifically for the hydrogen atom. (N.B. In Rautian and Sobelman's paper they consider that the material system is coupled to many modes of the radiation field, but that initially only one mode of the field is in a high-energy eigenstate. They obtained a solvable set of equations for the atom field probability amplitudes by truncating the infinite set to correspond to a SMALL number of multiphoton processes. Newstein (ref. 65) thinks that this procedure is only valid for __small__ initial field energies).

Before proceeding further we shall define frequencies

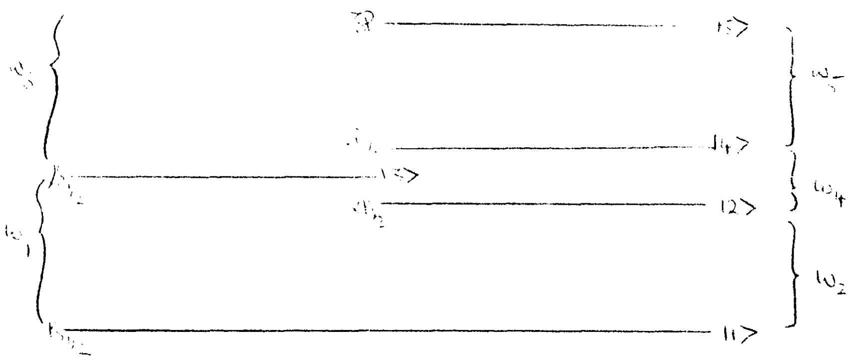


Fig. 6.A.7

$$\omega_5 = \epsilon_5 - \epsilon_4$$
$$\omega_4 = \epsilon_4 - \epsilon_2$$
$$\omega_2 = \epsilon_2 - \epsilon_1$$
$$\omega_3 = \epsilon_5 - \epsilon_3$$
$$\omega_1 = \epsilon_3 - \epsilon_1$$

B. Power Spectra (Emission spectra only)

In order to calculate the spectral densities of the emitted light for case a) it proves necessary to consider only two of the twenty-five equations for the transition operators. These are:

$\dot{P}_{15} =$ [illegible] (6.B.1)

$\dot{P}_{13} =$ [illegible] (6.B.2)

The monochromatic driving field coupling levels 5 and 3 will now be considered in the two ways explained in the last chapter.

Method (i)

In this case

[illegible] (6.B.3)

where [illegible]

hence [illegible] (6.B.4)

[illegible] (6.B.5)

and thus the new equations of motion are:

$$\left.\begin{aligned}\dot{P}_{15} = &-\{\tfrac{1}{2}\gamma_{51} + i(\epsilon_{51} - \Omega_{5})\}P_{15}\\ &-i(P_{25}q_{21} + P_{45}q_{41} + P_{55}q_{51})\\ &+i(P_{11}q_{51} + P_{13}(q_{53} + \lambda_{53}\mathcal{E}_L(t)))\end{aligned}\right\} \quad (6.B.6)$$

$$\left.\begin{aligned}\dot{P}_{13} = &-\{\tfrac{1}{2}\gamma_{32} + i(\epsilon_{31} - \Omega_{32})\}P_{13}\\ &-i(P_{23}q_{21} + P_{43}q_{41} + P_{53}q_{51})\\ &+iP_{12}q_{32}\\ &+i(q^{+}_{43}P_{41} + (q^{+}_{53} + \lambda_{35}\mathcal{E}^{*}_L(t))P_{15}\end{aligned}\right\} \quad (6.B.7)$$

where $\gamma_5 = \gamma_{51} + \gamma_{53}$

$\Omega_5 = \Omega_{51} + \Omega_{53}$

Multiplying these two equations from right and left by ${}_{rad}\langle 0|$ **and** $|0\rangle_{rad}$ **respectively and letting**

$$\left.\begin{aligned}{}_{rad}\langle 0|P_{15}(t)|0\rangle_{rad} &= X(t)\\ {}_{rad}\langle 0|P_{13}(t)|0\rangle_{rad} &= Y(t)\end{aligned}\right\} \quad (6.B.8)$$

we obtain

$$\left.\begin{aligned}\dot{X} = &-\{\tfrac{1}{2}\gamma_5 + i(\epsilon_{51} - \Omega_5)\}X(t)\\ &+i\lambda_{53}\mathcal{E}_L(t)Y(t)\\ \dot{Y} = &-\{\tfrac{1}{2}\gamma_{32} + i(\epsilon_{31} - \Omega_{32})\}Y(t)\\ &+i\lambda_{35}\mathcal{E}^{*}_L(t)X(t)\end{aligned}\right\} \quad (6.B.9)$$

Since

$$\left.\begin{aligned}\rho^{(s)}_{51}(t) &= \sum_s \langle s|X(t)\rho^{(s)}(0)|s\rangle\\ \rho^{(s)}_{51}(t) &= \sum_s \langle s|Y(t)\rho^{(s)}(0)|s\rangle\end{aligned}\right\} \quad (6.B.10)$$

we obtain the following two equations for the reduced density matrix elements

$$\left.\begin{aligned}\dot{\rho}^{(s)}(t) = &-\{\tfrac{1}{2}\gamma_5 + i(\epsilon_{51} - \Omega_5)\}\rho^{(s)}_{51}(t)\\ &+i\lambda_{53}\mathcal{E}_L(t)\rho^{(s)}_{31}(t)\end{aligned}\right\} \quad (6.B.11)$$

$$\left.\begin{aligned}\dot{\rho}_{21}(t) &= -\{\tfrac{1}{2}\gamma_{2} + i(\epsilon_{21} - \Omega_{2})\}\rho_{21}^{(1)}(t) \\ &\quad + i\lambda_{25}\mathcal{E}_L^{*}(t)\rho_{51}^{(1)}(t)\end{aligned}\right\} \qquad (6.B.12)$$

Method (ii)

In this case the initial photon state is $|\alpha_5\rangle$

$$q_{53}|i_{ph}\rangle = \sqrt{\frac{2\pi\omega_L}{\hbar V}}\,(\hat{e}_{0L}\cdot p_{53})\,e^{-i\omega_L t}\,\alpha_5|\alpha_5\rangle \qquad (6.B.13)$$

whereas $\quad q_{mn}|i_{ph}\rangle = 0 \quad$ for all other m and n where $m > n$

$$\left.\begin{aligned}\langle i_{ph}|q_{53}|i_{ph}\rangle &= g_{\omega_L,53}\,e^{-i\omega_L t}\,\alpha_5 \\ &= \tfrac{1}{2}G_L^{1}\,e^{-i\omega_L t}\end{aligned}\right\} \qquad (6.B.14a)$$

$$\left.\begin{aligned}\langle i_{ph}|q_{53}^{+}|i_{ph}\rangle &= g_{\omega_L,53}\,e^{i\omega_L t}\,\alpha_5^{*} \\ &= \tfrac{1}{2}G_L^{1*}\,e^{i\omega_L t}\end{aligned}\right\} \qquad (6.B.14b)$$

$$\left.\begin{aligned}\tfrac{1}{4}G_L^{2} &= g^{2}_{\omega_L,53}\,|\alpha_5|^2 \\ &= \left(\frac{2\pi\omega_L}{\hbar V}\right)(\hat{e}_{0L}\cdot p_{53})^2\,|\alpha_5|^2 \\ \text{as opposed to} \quad \tfrac{1}{4}G_5^{2} &= (\lambda_{53}\mathcal{E}_{0L})^2 \\ &= \left(\frac{p_{53}\cdot\hat{e}_{0L}\mathcal{E}_{0L}}{\hbar}\right)^2 \quad \text{of method (i)}\end{aligned}\right\} \qquad (6.B.15)$$

Hence multiplying the two transition operator equations from right and left by $\langle i_{ph}|$ and $|i_{ph}\rangle$ respectively we obtain

$$\left.\begin{aligned}\langle i_{ph}|\dot{P}_{15}|i_{ph}\rangle &= -\{\tfrac{1}{2}\gamma_5 + (\epsilon_{51} - \Omega_5)\}\langle i_{ph}|P_{15}|i_{ph}\rangle \\ &\quad + i\langle i_{ph}|P_{13}q_{53}|i_{ph}\rangle\end{aligned}\right\} \qquad (6.B.16)$$

$$\left.\begin{aligned}\langle i_{ph}|\dot{P}_{13}|i_{ph}\rangle &= -\{\tfrac{1}{2}\gamma_{32} + i(\epsilon_{31} - \Omega_{32})\}\langle i_{ph}|P_{13}|i_{ph}\rangle \\ &\quad + i\langle i_{ph}|q_{53}^{+}P_{15}|i_{ph}\rangle\end{aligned}\right\} \qquad (6.B.17)$$

and hence, since $$\rho^{(s)}_{mn}(t) = \sum_{s} \langle s| \langle ph| P_{mn} |ph\rangle \rho^{(s)}(0) |s\rangle \tag{6.B.18}$$

we find that

$$\left.\begin{aligned} \dot{\rho}^{(s)}_{51}(t) &= -\{ \tfrac{1}{2}\gamma_5 + i(\epsilon_{51} - \Omega_5)\} \rho^{(s)}_{51}(t) \\ &\quad + i\, g_{\omega_L, 53}\, e^{-i\omega_L t} \rho^{(s)}_{31}(t) \end{aligned}\right\} \tag{6.B.19}$$

$$\left.\begin{aligned} \dot{\rho}^{(s)}_{31}(t) &= -\{ \tfrac{1}{2}\gamma_{32} + i(\epsilon_{31} - \Omega_{32})\} \rho^{(s)}_{31}(t) \\ &\quad + i\, g_{\omega_L, 53}\, e^{i\omega_L t} \rho^{(s)}_{51}(t) \end{aligned}\right\} \tag{6.B.20}$$

Thus on comparing these equations with those of method (i), after assuming $\mathcal{E}_L(t)$ oscillates harmonically at ω_L such that

$\mathcal{E}_L(t) = \mathcal{E}_{0L} e^{-i\omega_L t}$ where $\omega_L \doteq \epsilon_{53}$ we find that

$$\left.\begin{aligned} \tfrac{1}{2} G_L' &\longrightarrow \lambda_{53}\mathcal{E}_{0L} \qquad \text{i.e.} \quad \alpha_5 \longrightarrow \sqrt{\frac{V}{2\pi\hbar\omega_L}}\,\mathcal{E}_{0L} \\ \tfrac{1}{2} G_L'^{*} &\longrightarrow \lambda_{35}\mathcal{E}_{0L} \qquad \phantom{\text{i.e.}} \quad \alpha_5^{*} \longrightarrow \sqrt{\frac{V}{2\pi\hbar\omega_L}}\,\mathcal{E}_{0L} \end{aligned}\right\} \tag{6.B.21}$$

Using the notation of method (i) we can rewrite the reduced density matrix equations

$$\left.\begin{aligned} \dot{\rho}^{(s)}_{51}(t) &= -(\tfrac{1}{2}\gamma_5 + i\epsilon_{51}')\rho^{(s)}_{51}(t) + i\lambda_{53}\mathcal{E}_{0L} e^{-i\omega_L t} \rho^{(s)}_{31}(t) \\ \text{where} \quad \epsilon_{51}' &= \epsilon_{51} - \Omega_5 \\ &= \omega_1' + \omega_3' \end{aligned}\right\} \tag{6.B.22a}$$

$$\left.\begin{aligned} \dot{\rho}^{(s)}_{31}(t) &= -(\tfrac{1}{2}\gamma_{32} + i\epsilon_{31}')\rho^{(s)}_{31}(t) + i\lambda_{35}\mathcal{E}_{0L} e^{i\omega_L t} \rho^{(s)}_{51}(t) \\ \text{where} \quad \epsilon_{31}' &= \epsilon_{31} - \Omega_{32} \\ &= \omega_1' \end{aligned}\right\} \tag{6.B.22b}$$

[According to Heitler[44], P. 338 - 9, in hydrogen-like atoms, for the P-levels which all have allowed transitions to the ground state (e.g. level 3P i.e. level 5) the displacement is very much smaller than the level width,

i.e. $\hbar(\Delta_{1} + \Delta_{53}) << \frac{1}{2}\gamma_{5}\left(= \frac{1}{2}(\gamma_{51} + \gamma_{53})\right)$

For the 2S-state, which is metastable (i.e. $2S_{\frac{1}{2}}$ or level 3) and has negligible width, the displacement is much larger than the width

i.e. $\Delta_{32} >> \frac{1}{2}\gamma_{32}$

The same is true for the ground state, of course, which is quite sharp. For the higher S-levels (which we do not consider), although allowed transitions to lower P-states occur, the transition probabilities as well as the level shift decrease rapidly with the main quantum number n and the level shift remains in general much bigger than the width.

The relative values of the level shifts and widths of the 2S and 2P levels of hydrogen are given by him in Table IX for the three hydrogen-levels with n = 2.]

Multiplying the first equation by $e^{i\omega_L t}$ and letting

$$\left.\begin{array}{l} x(t) = \rho_{51}^{(s)}(t) \quad \text{so that} \quad \dot{x}(t) - i\omega_L \rho_{51}^{(s)}(t) = e^{i\omega_L t}\dot{\rho}_{51}^{(s)}(t) \\ y(t) = \rho_{31}^{(s)}(t) \end{array}\right\} \quad (6.B.23)$$

$$\dot{x} - \omega_L x = -(\tfrac{1}{2}\gamma_5 + i\epsilon_{51}')x + i\lambda\mathcal{E}_{0L}y \tag{6.B.24}$$

$$\dot{y} = -(\tfrac{1}{2}\gamma_{32} + i\epsilon_{31}')y + i\lambda\mathcal{E}_{0L}x \tag{6.B.25}$$

If we now assume the initial time t' to be zero and take Laplace transforms of these two equations we obtain

$$(s + \tfrac{1}{2}\gamma_5 + i(\omega_1 + \omega_3' - \omega_L))\hat{x}(s) = i\lambda\mathcal{E}_{0L}\hat{y}(s) + x(0) \tag{6.B.26}$$

$$(s + \tfrac{1}{2}\gamma_{32} + i\omega_1')\hat{y}(s) = i\lambda\mathcal{E}_{0L}\hat{x}(s) + y(0) \tag{6.B.27}$$

(a) Spectral correlation function for transitions between levels 3F(5) and 1S₁(1)

In this case we need to solve for $\hat{\rho}_{51}^{(s)}(s)$ i.e. for $\hat{x}(s)$ $\hat{\rho}_{51}^{(s)}(s - i\omega_L)$ which we find is

$$\hat{x}(s) = \hat{\rho}_{51}^{(s)}(s - i\omega_L) = \frac{i\lambda\mathcal{E}_{0L}}{F(s)}\rho_{31}(0) + \frac{f_2(s)}{F(s)}\rho_{51}(0) \tag{6.B.28}$$

and dropping the superscript

$$\therefore \hat{\rho}_{51}^{(s)}(s) = \frac{i\lambda\mathcal{E}_{0L}}{F(s + i\omega_L)}\rho_{31}(0) + \frac{f_2(s + i\omega_L)}{F(s + i\omega_L)}\rho_{51}(0) \tag{6.B.29}$$

where

$$f_1(s) = s + \tfrac{1}{2}\gamma_5 + i(\omega_1' + \Delta_{53})$$

$$f_2(s) = s + \tfrac{1}{2}\gamma_{32} + i\omega_1'$$

$$F(s) = f_1(s)f_2(s) + \tfrac{1}{4}G^2 = (s - s_1)(s - s_2)$$

$$\therefore f_1(s + i\omega_L) = s + \tfrac{1}{2}\gamma_5 + i(\omega_1' + \omega_3')$$

$$f_2(s + i\omega_L) = s + \tfrac{1}{2}\gamma_{32} + i(\omega_1' + \omega_3' - \Delta_{53})$$

where $\Delta_{53} = \omega_5' - \omega_L$

$$\tfrac{1}{4}G^2 = \tfrac{1}{4}G_s^2$$

$$s_{1,2} = -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\left(2(\omega_L+\omega_3') - \Delta_{53}\right)\right\} \pm \tfrac{1}{2}\sqrt{\left\{\tfrac{1}{2}(\gamma_5-\gamma_{32}) + i\Delta_{53}\right\}^2 - G^2}$$

In other words

$$\hat{\rho}_{51}(s) = \hat{U}_{51,31}(s)\rho_{31}(0) + \hat{U}_{51,51}(s)\rho_{51}(0) \qquad (6.B.30)$$

where
$$\hat{U}_{51,31}(s) = \frac{i\lambda\hat{\varepsilon}_L}{F(s+i\omega_L)}$$

$$\hat{U}_{51,51}(s) = \frac{f_2(s+i\omega_L)}{F(s+i\omega_L)}$$

The quantity we now have to calculate is the 2-time atomic correlation function

$$g_{51}(\tau,t') = \langle P_{15}^{+}(t')P_{15}(t'+\tau)\rangle$$

$$\simeq \sum_{y''=1}^{5} U_{51,y''1}(\tau,t')\rho^{(s)}(t')_{y''5} \quad \text{under Markoff approximation} \qquad (6.B.31)$$

$$= U_{51,51}(\tau,t')\rho_{55}^{(s)}(t') + U_{51,31}(\tau,t')\rho_{35}^{(s)}(t')$$

$$= U_{51,51}(\tau)\rho_{55}^{(s)}(0) + U_{51,31}(\tau)\rho_{35}^{(s)}(0) \quad \text{for initial time } t' = 0$$

Taking the Laplace transform, we obtain

$$\hat{g}_{51}(s) = \hat{U}_{51,51}(s)\rho_{55}^{(s)}(0) + \hat{U}_{51,31}(s)\rho_{35}^{(s)}(0) \qquad (6.B.32)$$

We can show, by a method analogous to that used in Chapter V that the spectral correlation function

$$\left.\begin{aligned}\hat{g}_{51}(\nu) &= \int_{-\infty}^{\infty} d\tau\, e^{i\nu\tau}\, g_{51}(\tau) \\ &= 2\,\mathrm{Re}\left\{ \hat{U}_{51,51}(-i\nu)\rho_{55}^{(1)}(0) + \hat{U}_{51,31}(-i\nu)\rho_{35}^{(1)}(0) \right\}\end{aligned}\right\} \quad (6.B.33)$$

but we now assume the atom to be initially in level 5, having been excited to this state by the field long ago,

$$\therefore\ \rho_{55}^{(1)}(0) = 1 \quad , \rho_{35}^{(1)}(0) = 0 \qquad (6.B.34)$$

$$\left.\begin{aligned}\therefore\ \hat{g}_{51}(\nu) &= 2\,\mathrm{Re}\ \hat{U}_{51,51}(-i\nu) \\ &= 2\,\mathrm{Re}\left\{ \frac{f_2(-i(\nu-\omega_L))}{F(-i(\nu-\omega_L))} \right\} \\ &= \frac{2}{|F(-i(\nu-\omega_L))|^2}\ \mathrm{Re}\left\{ F^*(-i(\nu-\omega_L))\, f_2(-i(\nu-\omega_L)) \right\}\end{aligned}\right\} \quad (6.B.35)$$

Let $\quad s_1 = a+ib \quad , \quad s_2 = c+id$

where $\quad s_{1,2} = -\tfrac{1}{2}\left\{ \tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\,(2(\omega_1'+\omega_3') - \Delta_{53}) \right\} \pm \tfrac{1}{2}\sqrt{x+iy}$

$$= -\tfrac{1}{2}\left\{ \tfrac{1}{2}(\gamma_5+\gamma_{32}) \mp A \right\} - \tfrac{1}{2}\, i \left\{ 2(\omega_1'+\omega_3') - \Delta_{53} \mp B \right\}$$

and $\quad x = \tfrac{1}{4}(\gamma_5-\gamma_{32})^2 - (\Delta_{53}^2 + G_5^2)$

$\quad y = (\gamma_5-\gamma_{32})\,\Delta_{53}$

$$\sqrt{x+iy} = A+iB \qquad (6.B.36)$$

where $\quad A = \sqrt{\dfrac{x+\sqrt{x^2+y^2}}{2}}$

$\quad B = \sqrt{\dfrac{-x+\sqrt{x^2+y^2}}{2}}$

Thus $\quad a = -\tfrac{1}{2}\left\{ \tfrac{1}{2}(\gamma_5+\gamma_{32}) - A \right\} \quad , \quad b = -\tfrac{1}{2}\left\{ 2(\omega_1'+\omega_3') - \Delta_{53} - B \right\}$

$\quad c = -\tfrac{1}{2}\left\{ \tfrac{1}{2}(\gamma_5+\gamma_{32}) + A \right\} \quad , \quad d = -\tfrac{1}{2}\left\{ 2(\omega_1'+\omega_3') - \Delta_{53} + B \right\}$

In terms of these values the spectral correlation function is

$$\tilde{g}_{51}(\nu) = \frac{2\{\tfrac{1}{2}\gamma_{32}[\tfrac{1}{4}\gamma_5\gamma_{32} + \tfrac{1}{4}G^2 - (\nu-\omega_1'-\omega_3')(\nu-\omega_1'-\omega_3'+\Delta_{53})] - \tfrac{1}{2}(\nu-\omega_1'-\omega_3'+\Delta_{53})[\gamma_5(\nu-\omega_1'-\omega_3'+\Delta_{53}) + \gamma_{32}(\nu-\omega_1'-\omega_3')]\}}{\{a^2 + (\nu-\omega_- + b)^2\}\{c^2 + (\nu-\omega_- + d)^2\}} \tag{6.B.37}$$

or
$$\tilde{g}_{51}(\nu') = \frac{2\{\tfrac{1}{2}\gamma_{32}[\tfrac{1}{4}\gamma_5\gamma_{32} + \tfrac{1}{4}G^2 - \nu'(\nu'+\Delta_{53})] - \tfrac{1}{2}(\nu'+\Delta_{53})[\gamma_5(\nu'+\Delta_{53}) + \gamma_{32}\nu']\}}{\{a^2 + (\nu' + (\omega_1'+\omega_3'+b))^2\}\{c^2 + (\nu' + (\omega_1'+\omega_3'+d))^2\}}$$

where $\nu' = \nu - (\omega_1' + \omega_3')$

Hence the spectral profile has two peaks at

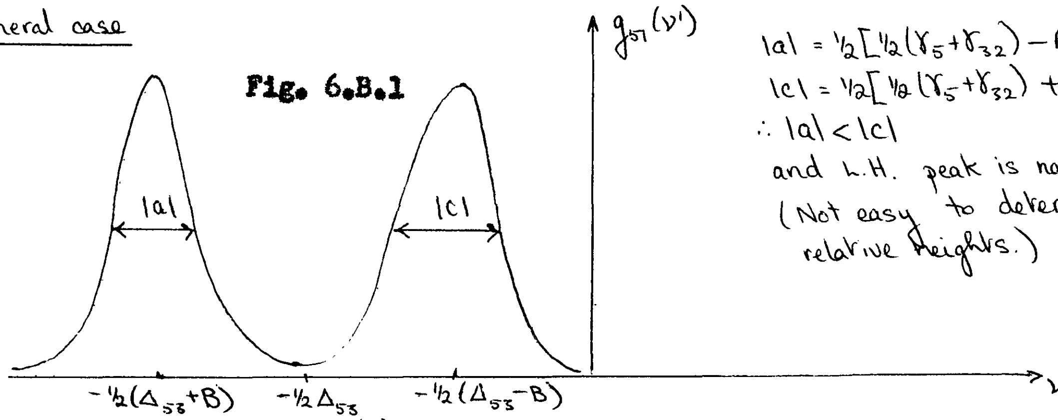
$$\left.\begin{aligned} \nu' &= -(\omega_1'+\omega_3'+b) = -\tfrac{1}{2}(\Delta_{53}+B) \\ \nu' &= -(\omega_1'+\omega_3'+d) = -\tfrac{1}{2}(\Delta_{53}-B) \end{aligned}\right\} \tag{6.B.38}$$

of widths $|a|$ and $|c|$.

Thus we can see that for __resonance,__ $\Delta_{53} = 0$, $B = 0$ and so there is only one peak at $\nu' = 0$ of width $\frac{1}{4}(\gamma_5 + \gamma_{32})$ and this corresponds to Rautian and Sobelman's case of $\Omega_L = 0$ and $G = 0$ although they obtain a splitting for larger G.

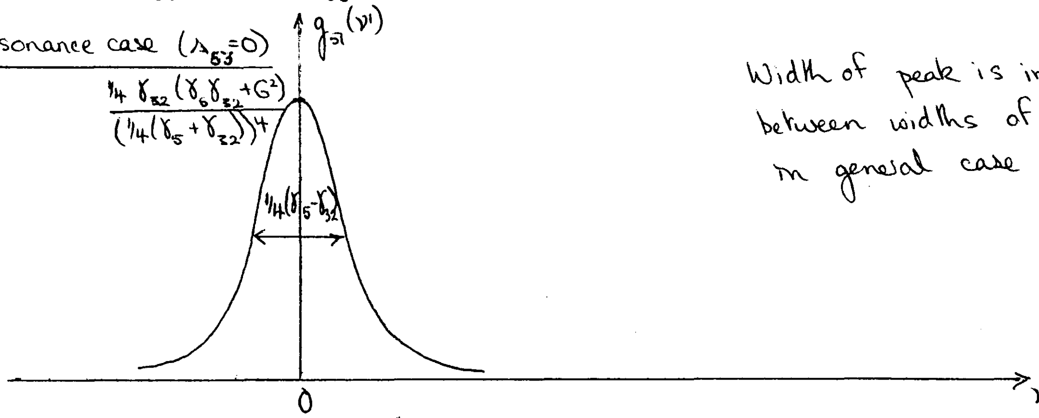
General case

Fig. 6.B.1



$|a| = \frac{1}{2}[\frac{1}{2}(\gamma_5+\gamma_{32}) - A]$
$|c| = \frac{1}{2}[\frac{1}{2}(\gamma_5+\gamma_{32}) + A]$
$\therefore |a| < |c|$
and L.H. peak is narrower
(Not easy to determine relative heights.)

Resonance case ($\Delta_{53}=0$)



Width of peak is intermediate between widths of 2 peaks in general case.

Fig. 6.B.2

We shall next consider $G^2 << |\tfrac{1}{2}(\gamma_5 - \gamma_{32}) + i\ldots$

i.e. the <u>small perturbation limit</u>, mentioned earlier. In this case

$$s_{1,2} = -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\left(2(\omega_1+\omega_3') - \Delta_{53}\right)\right\} \pm \tfrac{1}{2}\left\{1 - \frac{G^2}{\left[\tfrac{1}{2}(\gamma_5-\gamma_{32}) + i\Delta_{53}\right]^2}\right\}^{1/2}$$

and to first order in $\dfrac{G^2}{|\tfrac{1}{2}(\gamma_5-\gamma_{32}) + i\Delta_{53}|^2}$

$$s_{1,2} \simeq -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\left(2(\omega_1+\omega_3') - \Delta_{53}\right)\right\} \pm \tfrac{1}{2}\left\{1 - \tfrac{1}{2}\frac{G^2}{\left[\tfrac{1}{2}(\gamma_5-\gamma_{32}) + i\Delta_{53}\right]^2}\right\}$$

$$= -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\left(2(\omega_1+\omega_3') - \Delta_{53}\right)\right\} \pm \tfrac{1}{2}\left\{1 - \tfrac{1}{2}\frac{G^2}{\left[\tfrac{1}{4}(\gamma_5-\gamma_{32})^2 + \Delta_{53}^2\right]}\right\}$$

$$\therefore\ s_{1,2} = -\tfrac{1}{2}\left[\tfrac{1}{2}(\gamma_5+\gamma_{32}) \mp \left\{\frac{\tfrac{1}{2}G^2\,\tfrac{1}{2}(\gamma_5-\gamma_{32})}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\}\right] - \tfrac{1}{2}i\left[2(\omega_1+\omega_3') - \Delta_{53} \mp \frac{\tfrac{1}{2}G^2\Delta_{53}}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right]$$

$$\left.\begin{aligned} s_1 = a+ib &= -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) - 1 + \frac{\tfrac{1}{2}G^2\,\tfrac{1}{2}(\gamma_5-\gamma_{32})}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\} \\ &\quad -\tfrac{1}{2}i\left\{2(\omega_1+\omega_3') - \Delta_{53} - \frac{\tfrac{1}{2}G^2\Delta_{53}}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\} \\ s_2 = c+id &= -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) - 1 - \frac{\tfrac{1}{2}G^2\,\tfrac{1}{2}(\gamma_5-\gamma_{32})}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\} \\ &\quad -\tfrac{1}{2}i\left\{2(\omega_1+\omega_3') - \Delta_{53} + \frac{\tfrac{1}{2}G^2\Delta_{53}}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\} \end{aligned}\right\} \quad (6.B.39)$$

s.t. peaks occur at

$$\nu_1' = -(\omega_1+\omega_3'+b) = -\tfrac{1}{2}\Delta_{53}\left(1 + \frac{\tfrac{1}{2}G^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right)$$

$$\nu_2' = -(\omega_1+\omega_3'+d) = -\tfrac{1}{2}\Delta_{53}\left(1 - \frac{\tfrac{1}{2}G^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right)$$

and have widths

$$\Delta_1 = \tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) - 1 + \frac{\tfrac{1}{2}G^2\,\tfrac{1}{2}(\gamma_5-\gamma_{32})}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\}$$

$$\Delta_2 = \tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) - 1 - \frac{\tfrac{1}{2}G^2\,\tfrac{1}{2}(\gamma_5-\gamma_{32})}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2+\Delta_{53}^2}\right\} \quad (6.B.40)$$

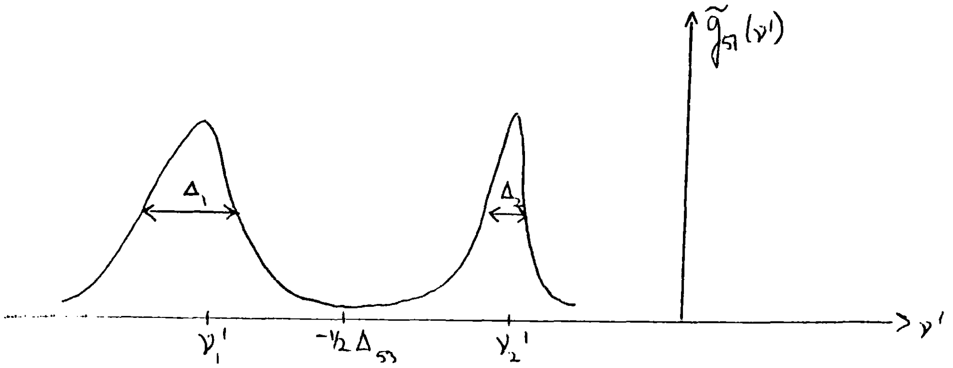


Fig. 6.B.3

right hand peak narrower (opposite to general case for field of arbitrary magnitude)

If $G^2 >> |\frac{1}{2}(\gamma_5 - \gamma_{32}) + i\Delta_{53}|^2$ i.e. <u>the strong field limit</u>, then to zeroeth order in $\dfrac{|\frac{1}{2}(\gamma_5-\gamma_{32}) + i\Delta_{53}|^2}{G^2}$ we have

$$S_{1,2} \triangleq -\tfrac{1}{2}\left\{\tfrac{1}{2}(\gamma_5+\gamma_{32}) + i\,(2(\omega_1'+\omega_3') - \Delta_{53})\right\} \pm \tfrac{1}{2}iG$$

$$= -\tfrac{1}{4}(\gamma_5+\gamma_{32}) - \tfrac{1}{2}i\left\{2(\omega_1'+\omega_3') - \Delta_{53} \mp G\right\}$$

$$\left.\begin{aligned} \therefore\; S_1 &= a+ib = -\tfrac{1}{4}(\gamma_5+\gamma_{32}) - \tfrac{1}{2}i\,(2(\omega_1'+\omega_3') - \Delta_{53} - G) \\ S_2 &= c+id = -\tfrac{1}{4}(\gamma_5+\gamma_{32}) - \tfrac{1}{2}i\,(2(\omega_1'+\omega_3') - \Delta_{53} + G) \end{aligned}\right\} \quad (6.B.41)$$

s.t. 2 peaks occur at

$$\left.\begin{aligned} \nu_1' &= -(\omega_1'+\omega_3'+b) = -\tfrac{1}{2}(\Delta_{53}+G) \\ \nu_2' &= -(\omega_1'+\omega_3'+d) = -\tfrac{1}{2}(\Delta_{53}-G) \end{aligned}\right\} \quad (6.B.42)$$

and both have the same width $|a| = |c| = \frac{1}{4}(\gamma_5 + \gamma_{32})$

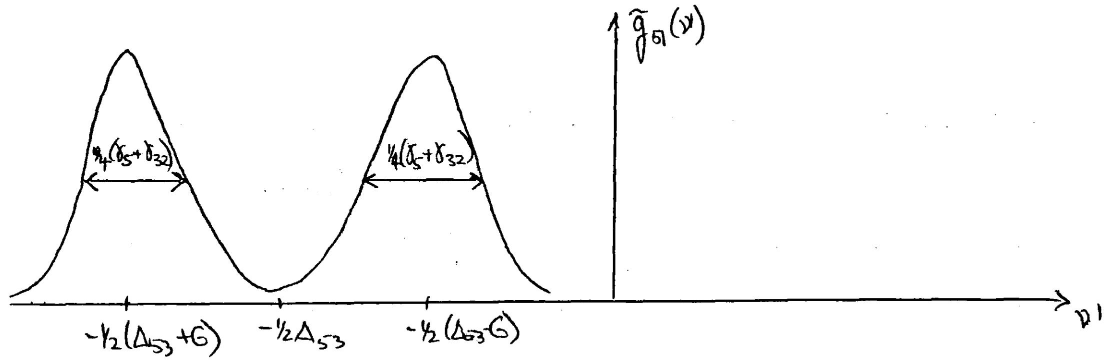


Fig. 6.B.4

When the driving field vanishes identically, i.e. $G_5 = 0$ then we

$$\left.\begin{aligned} S_{1,2} &= -\tfrac{1}{2}\{\tfrac{1}{2}(\gamma_5+\gamma_{32})+i(2(\omega_1'+\omega_3')-\Delta_{53})\} \pm \tfrac{1}{2}\{\tfrac{1}{2}(\gamma_5-\gamma_{32})+i\Delta_{53}\} \\ S_1 &= a+ib = -\tfrac{1}{2}\gamma_{32}-i(\omega_1'+\omega_3'-\Delta_{53}) \\ S_2 &= c+id = -\tfrac{1}{2}\gamma_5-i(\omega_1'+\omega_3') \end{aligned}\right\} \qquad (6.B.43)$$

s.t. 2 peaks occur at

$$\nu_1' = -(\omega_1'+\omega_3'+b) = -\Delta_{53}$$

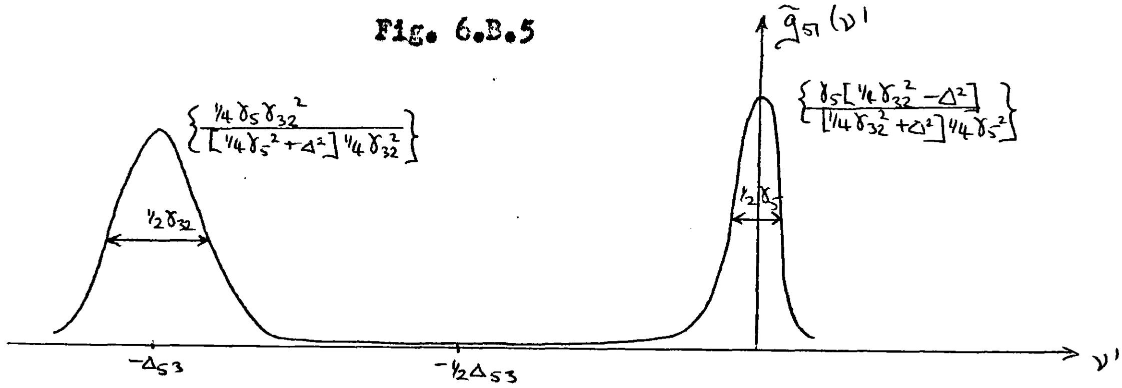
$$\nu_2' = -(\omega_1'+\omega_3'+d) = 0 \qquad (6.B.44)$$

and have widths $\Delta_1 = |a| = \tfrac{1}{2}\gamma_{32}$

$$\Delta_2 = |b| = \tfrac{1}{2}\gamma_5$$

i.e. their spontaneous decay widths as expected

Fig. 6.B.5



The heights of the two components, in each case, are not identical but their relative values will depend on the relative magnitudes of the decay constants, field strength and the nearness to resonance.

(b) Spectral correlation function for transitions between levels 3P (5) and $2S_{\frac{1}{2}}$ (3)

Here we need to solve for $\hat{\rho}_{53}^{(5)}(s)$ and thus we need to consider five equations as follows, where we have dropped the superscript indicating the density matrices to be reduced ones since in the Markoff approximation, which we shall again be using, this distinction is not important:

$$\dot{\rho}_{35} = -\{\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i\omega_3'\}\rho_{35} - i\lambda\mathcal{E}_{0L}\rho_{33}e^{i\omega_L t} + i\lambda\mathcal{E}_{0L}\rho_{55}e^{i\omega_L t} \tag{6.B.45}$$

$$\dot{\rho}_{33} = -\gamma_{32}\rho_{33} + \gamma_{43}\rho_{44} + \gamma_{53}\rho_{55} - i\lambda\mathcal{E}_{0L}\rho_{35}e^{-i\omega_L t} + i\lambda\mathcal{E}_{0L}\rho_{53}e^{i\omega_L t} \tag{6.B.46}$$

$$\dot{\rho}_{55} = -\gamma_5\rho_{55} + i\lambda\mathcal{E}_{0L}e^{-i\omega_L t}\rho_{35} - i\lambda\mathcal{E}_{0L}e^{i\omega_L t}\rho_{53} \tag{6.B.47}$$

$$\dot{\rho}_{53} = -\{\tfrac{1}{2}(\gamma_{32}+\gamma_5) + i\omega_3'\}\rho_{53} - i\lambda\mathcal{E}_{0L}e^{-i\omega_L t}\rho_{55} + i\lambda\mathcal{E}_{0L}e^{-i\omega_L t}\rho_{33} \tag{6.B.48}$$

$$\dot{\rho}_{44} = -\gamma_4\rho_{44} \tag{6.B.49}$$

where $\gamma_4 = \gamma_{41} + \gamma_{43}$

Multiplying the first equation by $e^{-i\omega_L t}$ and the fourth by $e^{i\omega_L t}$ and letting

$$\left.\begin{array}{lll} \rho_{35}e^{-i\omega_L t} = x & & \therefore\ \dot{x} + i\omega_L x = e^{-i\omega_L t}\dot{\rho}_{35} \\ \rho_{33} = y & & \\ \rho_{55} = z & & \\ \rho_{53}e^{i\omega_L t} = l & & \therefore\ \dot{l} - i\omega_L l = e^{i\omega_L t}\dot{\rho}_{53} \\ \rho_{44} = m & & \end{array}\right\} \tag{6.B.50}$$

we obtain

$$\dot{x} = -\{\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i(\omega_3'-\omega_L)\}x - i\lambda\mathcal{E}_{0L}y + i\lambda\mathcal{E}_{0L}z \tag{6.B.51}$$

$$\dot{y} = -\gamma_{32}y + \gamma_{43}m + \gamma_{53}z - i\lambda\mathcal{E}_{0L}x + i\lambda\mathcal{E}_{0L}l \tag{6.B.52}$$

$$\dot{z} = -\gamma_5 z + i\lambda\mathcal{E}_{0L}x - i\lambda\mathcal{E}_{0L}l \tag{6.B.53}$$

$$\dot{l} = -\{\tfrac{1}{2}(\gamma_{32}+\gamma_5) + i(\omega_3'-\omega_L)\}l - i\lambda\mathcal{E}_{0L}z + i\lambda\mathcal{E}_{0L}y \tag{6.B.54}$$

$$\dot{m} = -\gamma_4 m \tag{6.B.55}$$

Taking Laplace transforms and assuming the initial time t' = 0 we obtain

$$\left\{ s+\tfrac{1}{2}(\gamma_{32}+\gamma_5)-i(\omega_2'-\omega_L)\right\}\hat{x}(s) = -i\lambda\mathcal{E}_{oL}\,\hat{y}(s) + i\lambda\mathcal{E}_{oL}\,\hat{z}(s) + x(0) \qquad (6.B.56)$$

$$\left.\begin{aligned}(s+\gamma_{32})\hat{y}(s) &= \gamma_{43}\,\hat{m}(s) + \gamma_{53}\,\hat{z}(s) - i\lambda\mathcal{E}_{oL}\,\hat{x}(s)\\ &\quad + i\lambda\mathcal{E}_{oL}\,\hat{\ell}(s) + y(0)\end{aligned}\right\} \qquad (6.B.57)$$

$$(s+\gamma_5)\hat{z}(s) = i\lambda\mathcal{E}_{oL}\,\hat{x}(s) - i\lambda\mathcal{E}_{oL}\,\hat{\ell}(s) + z(0) \qquad (6.B.58)$$

$$\left\{ s+\tfrac{1}{2}(\gamma_{32}+\gamma_5)+i(\omega_3'-\omega_L)\right\}\hat{\ell}(s) = -i\lambda\mathcal{E}_{oL}\,\hat{z}(s) + i\lambda\mathcal{E}_{oL}\,\hat{y}(s) + \ell(0) \qquad (6.B.59)$$

$$(s+\gamma_4)\,\hat{m}(s) = m(0) \qquad (6.B.60)$$

These five equations can be solved for $\hat{\ell}(s)$ as follows. We first obtain an expression for $\hat{m}(s)$ from (6.B.60), then one for $\hat{z}(s)$ from (6.B.58), which we substitute, together with $\hat{m}(s)$ in (6.B.57) to obtain $\hat{y}(s)$. Then from (6.B.56) we obtain $\hat{x}(s)$ in terms of $\hat{\ell}(s)$ which we substitute in (6.B.59) to find $\hat{\ell}(s)$ which is given by the following equation:-

$$\begin{aligned}\hat{\ell}(s) = \hat{\rho}_{53}(s-i\omega_L) = {} & \frac{(i\lambda\mathcal{E}_{oL})\gamma_{43}(s+\gamma_5)}{H(s)\,(s+\gamma_4)}\left\{G(s) - 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)\right\}\rho_{44}(0)\\ & - \frac{(i\lambda\mathcal{E}_{oL})(s+\gamma_{32}+\gamma_{53})}{H(s)}\left\{G(s) - 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)\right\}\rho_{55}(0)\\ & + \frac{(i\lambda\mathcal{E}_{oL})(s+\gamma_5)}{H(s)}\left\{G(s) - 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)\right\}\rho_{33}(0) \qquad (6.B.61a)\\ & + \frac{2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)(s+\gamma_{32})(s+\gamma_5)}{H(s)}\rho_{35}(0)\\ & + \frac{(s+\gamma_{32})(s+\gamma_5)G(s)}{H(s)}\rho_{53}(0)\end{aligned}$$

where

$$\begin{aligned}H(s) &= f_3'(s)(s+\gamma_{32})(s+\gamma_5)G(s) + 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)G(s) - \left(2\,\tfrac{1}{4}G^2\right)^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)^2\\ &= G(s)\left\{f_3'(s)\,(s+\gamma_{32})(s+\gamma_5) + 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)\right\} - \left(2\,\tfrac{1}{4}G^2\right)^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right)^2\\ G(s) &= f_3(s)(s+\gamma_5)(s+\gamma_{32}) + 2\,\tfrac{1}{4}G^2\left(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})\right) \qquad (6.B.61b)\\ f_3(s) &= s+\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i\Delta_{53}\\ f_3'(s) &= s+\tfrac{1}{2}(\gamma_{32}+\gamma_5) + i\Delta_{53}\end{aligned}$$

$$\hat{\rho}_{53}(s) = \frac{(i\lambda\mathcal{E}_{0L})\gamma_{43}(s+\gamma_5+i\omega_L)\{G(s+i\omega_L)-\tfrac{1}{2}G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})+i\omega_L)\}}{H(s+i\omega_L).(s+\gamma_4+i\omega_L)}\rho_{44}(0)$$

$$-\frac{(i\lambda\mathcal{E}_{0L})(s+\gamma_{32}-\gamma_{55}+i\omega_L)\{G(s+i\omega_L)-\tfrac{1}{2}G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})+i\omega_L)\}}{H(s+i\omega_L)}\rho_{55}(0)$$

$$+\frac{(i\lambda\mathcal{E}_{0L})(s+\gamma_5+i\omega_L)\{G(s+i\omega_L)-\tfrac{1}{2}G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})+i\omega_L)\}}{H(s+i\omega_L)}\rho_{33}(0)$$

$$+\frac{\tfrac{1}{2}G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51})+i\omega_L)(s+\gamma_{32}+i\omega_L)(s+\gamma_5+i\omega_L)}{H(s+i\omega_L)}\rho_{35}(0) \qquad (6.B.62)$$

$$+\frac{(s+\gamma_{32}+i\omega_L)(s+\gamma_5+i\omega_L)}{H(s+i\omega_L)}\rho_{53}(0)$$

$$= \hat{u}_{53,44}(s)\rho_{44}(0) + \hat{u}_{53,55}(s)\rho_{55}(0) + \hat{u}_{53,33}(s)\rho_{33}(0)$$

$$+ \hat{u}_{53,35}(s)\rho_{35}(0) + \hat{u}_{53,53}(s)\rho_{53}(0)$$

The relevant 2-time correlation function, in this case, is

$$g_{53}(\tau,t') = \langle P_{35}^{+}(t')P_{35}(t'+\tau)\rangle$$

$$\doteq \sum_{y''=1}^{5} u_{53;y''3}(\tau,t')\rho_{y''5}(t') \qquad \text{under Markoff approximation} \qquad (6.B.63)$$

i.e. $\quad g_{53}(\tau) = u_{53,35}(\tau)\rho_{35}(0) + u_{53,53}(\tau)\rho_{55}(0)$ for initial time $t'=0$

Taking the Laplace transform

$$\hat{g}_{53}(s) = \hat{u}_{53,35}(s)\rho_{35}(0) + \hat{u}_{53,53}(s)\rho_{55}(0)$$

Hence, as before, the spectral correlation function is given by

$$\hat{g}_{53}(\nu) = \int_{-\infty}^{\infty} d\tau\, e^{i\nu\tau}\, g_{53}(\tau)$$

$$= 2\,\mathrm{Re}\{\hat{u}_{53,35}(-i\nu)\rho_{35}(0) + \hat{u}_{53,53}(-i\nu)\rho_{55}(0)\} \qquad (6.B.64)$$

and as the atom is initially in level 5

$$\rho_{55}(0) = 1 \;;\; \rho_{35}(0) = 0 \qquad (6.B.65)$$

so $\tilde{g}_{53}(\nu) = 2\,\mathrm{Re}\{\hat{U}_{53,53}(-i\nu)\}$

$$= 2\,\mathrm{Re}\left\{\frac{(\gamma_{32}-i(\nu-\omega_L))(\gamma_5-i(\nu-\omega_L))}{H(-i(\nu-\omega_L))}\right\}$$

$$\therefore\ \tilde{g}_{53}(\nu) = \frac{2}{|H(-i(\nu-\omega_L))|^2}\,\mathrm{R}\left\{H(-i(\nu-\omega_L))^*(\gamma_{32}-i(\nu-\omega_L))(\gamma_5-i(\nu-\omega_L))\right\} \quad (6.B.66)$$

At first sight it would appear that the spectrum has six peaks since H is a 6th order equation but we will consider $H(s)$ more carefully.

$$H(s) = G(s)\{(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5)+i\Delta_{53})(s+\gamma_{32})(s+\gamma_5)+\tfrac{1}{2}G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51}))\}$$
$$-(\tfrac{1}{2}G^2)^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51}))^2$$

and can be factorised so that,

$$\left.\begin{aligned} H(s) &= (s+\gamma_{32})(s+\gamma_5)\{(s+\gamma_{32})(s+\gamma_5)[(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))^2+\Delta_{53}^2] \\ &\quad +G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{51}))\} \\ &= (s+\gamma_{32})(s+\gamma_5)J(s) \end{aligned}\right\} \quad (6.B.67)$$

$$\left.\begin{aligned} \text{Hence}\quad \hat{U}_{53,53}(-i\nu) &= \frac{1}{J(-i(\nu-\omega_L))} \\ \therefore\ \tilde{g}_{53}(\nu) &= \frac{2}{|J(-i(\nu-\omega_L))|^2}\,\mathrm{Re}\{J(-i(\nu-\omega_L))\}^* \end{aligned}\right\} \quad (6.B.68)$$

$$\left.\begin{aligned} J(-i(\nu-\omega_L)) &= \{[\gamma_{32}\gamma_5-(\nu-\omega_L)^2][\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-(\nu-\omega_L)^2+\Delta_{53}^2] \\ &\quad -(\nu-\omega_L)^2(\gamma_{32}+\gamma_5)^2+G^2[\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-(\nu-\omega_L)^2]\} \\ &\quad - i(\nu-\omega_L)(\gamma_{32}+\gamma_5)\{\gamma_{32}\gamma_5+\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-2(\nu-\omega_L)^2+\Delta_{53}^2+G^2\} \end{aligned}\right\} \quad (6.B.69)$$

$$\boxed{\begin{aligned} \therefore\ \tilde{g}_{53}(\nu) = \frac{2}{|J(-i(\nu-\omega_L))|^2}\{&[\gamma_{32}\gamma_5-(\nu-\omega_L)^2][\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-(\nu-\omega_L)^2+\Delta_{53}^2] \\ &-(\nu-\omega_L)^2(\gamma_{32}+\gamma_5)^2+G^2[\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-(\nu-\omega_L)^2]\} \end{aligned}} \quad (6.B.70)$$

or

$$\boxed{\begin{aligned} \tilde{g}_{53}(\nu'') = \frac{2}{|J(-i(\nu''+\Delta_{53}))|^2}\{&[\gamma_{32}\gamma_5-(\nu''+\Delta_{53})][\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2-(\nu''+\Delta_{53})^2+\Delta_{53}^2] \\ &-(\nu''+\Delta_{53})^2(\gamma_{32}+\gamma_5)^2+G^2[\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2 \\ &-(\nu''+\Delta_{53})^2]\} \end{aligned}} \quad (6.B.71)$$

where $\nu'' = \nu-\omega_3' = \nu-(\Delta_{53}+\omega_L)$

i.e. the spectral profile has four peaks and to find this we must discover the roots of the quartic equation $J(s)$.

$$J(s) = (s+\gamma_{32})(s+\gamma_5)\{(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))^2+\Delta_{53}^2\}+G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))(s+\tfrac{1}{2}(\gamma_{57}+\gamma_5)) \tag{6.B.72}$$

In the case of resonance, $\Delta_{53} = 0$, we can thus see that $J(s)$ is given by

$$\left.\begin{aligned} J(s) &= (s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))\{(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))(s+\gamma_{32})(s+\gamma_5)+G^2(s+\tfrac{1}{2}(\gamma_{32}+\gamma_{57}))\} \\ &= (s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))\,K(s) \end{aligned}\right\} \tag{6.B.73}$$

i.e. we need only find the cube roots of $K(s)$

First we shall consider the non-resonance case:-

$$\left.\begin{aligned} J(s) &= s^4+as^3+bs^2+cs+d \\ \text{where}\quad a &= 2(\gamma_{32}+\gamma_5) \\ b &= \tfrac{5}{4}(\gamma_{32}+\gamma_5)^2+\gamma_{32}\gamma_5+G^2+\Delta^2 \\ c &= (\gamma_{32}+\gamma_5)\{\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2+\Delta^2+\gamma_{32}\gamma_5\}+\tfrac{1}{2}(2\gamma_{32}+\gamma_5+\gamma_{57})G^2 \\ d &= \gamma_{32}\gamma_5\{\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2+\Delta^2\}+G^2\tfrac{1}{4}(\gamma_{32}+\gamma_5)(\gamma_{32}+\gamma_{57}) \end{aligned}\right\} \tag{6.B.74}$$

First we must reduce $J(s)$ to standard form by substituting

$$\left.\begin{aligned} y &= s+\tfrac{1}{4}a \quad \text{then} \\ J(y) &= y^4+py^2+qy+r \\ \text{where}\quad p &= -\tfrac{3}{8}a^2+b \\ q &= \tfrac{1}{8}a^3-\tfrac{1}{2}ab+c \\ r &= -\tfrac{3}{4\cdot 64}a^4+\tfrac{1}{16}a^2b-\tfrac{1}{4}ac+d \end{aligned}\right\} \tag{6.B.75}$$

Then, proceeding as in Chapter III we write

$$J(y) = (y^2+ly+m)(y^2-ly+n) \tag{6.B.76}$$

$$\left.\begin{aligned} p &= n+m-l^2 \\ q &= ln-lm = l(n-m) \\ r &= mn \end{aligned}\right\} \tag{6.B.77}$$

$$\left.\begin{aligned} m + n &= p + l^2 \\ m - n &= -q/l \end{aligned}\right\} \quad (6.B.78)$$

hence

$$\left.\begin{aligned} m &= \tfrac{1}{2}(p + l^2 - q/l) \\ n &= \tfrac{1}{2}(p + l^2 + q/l) \end{aligned}\right\} \quad (6.B.79)$$

$$\left.\begin{aligned} mn &= r \\ \therefore 4r &= (p + l^2)^2 - q^2/l^2 \end{aligned}\right\} \quad (6.B.80)$$

$$\therefore (l^2)^3 + 2p(l^2)^2 + (p^2 - 4r)(l^2) - q^2 = 0 \quad (6.B.81)$$

and this can be solved for l^2 and using the positive value, a real value of l can be found and hence m and n which will also be real. The four roots of J(y) are then

$$y_{1,2} = \frac{-l \pm \sqrt{l^2 - 4m}}{2} \qquad y_{3,4} = \frac{l \pm \sqrt{l^2 - 4n}}{2} \quad (6.B.82)$$

(real for $l^2 \geqslant 4m$) (real for $l^2 \geqslant 4n$)

and thus the four roots of J(s) are

$$\begin{aligned} s_{1,2} &= y_{1,2} - \tfrac{1}{4}a & s_{3,4} &= y_{3,4} - \tfrac{1}{4}a \\ &= \left\{\frac{-l \pm \sqrt{l^2 - 4m} - \tfrac{1}{2}a}{2}\right\} & &= \left\{\frac{l \pm \sqrt{l^2 - 4n} - \tfrac{1}{2}a}{2}\right\} \end{aligned} \quad (6.B.83)$$

Let $J(s) = (s - s_1)(s - s_2)(s - s_3)(s - s_4)$

$$\therefore J(-i(\nu - \omega_L)) = (s_1 + i(\nu - \omega_L))(s_2 + i(\nu - \omega_L))(s_3 + i(\nu - \omega_L))(s_4 + i(\nu - \omega_L)) \quad (6.B.84)$$

and assuming the four roots are complex

$$\left.\begin{aligned} J(-i(\nu - \omega_L)) &= (z_{R_1} + i(\nu - \omega_L + z_{I_1}))(z_{R_2} + i(\nu - \omega_L + z_{I_2}))(z_{R_3} + i(\nu - \omega_L + z_{I_3})) \\ &\quad \times (z_{R_4} + i(\nu - \omega_L + z_{I_4})) \\ J(-i(\nu'' + \Delta_{53})) &= (z_{R_1} + i(\nu'' + \Delta_{53} + z_{I_1}))(z_{R_2} + i(\nu'' + \Delta_{53} + z_{I_2}))\ldots\ldots \end{aligned}\right\} \quad (6.B.85)$$

so that the four peaks are at

$$\nu'' = -(\Delta_{53} + z_{I_1}),\ -(\Delta_{53} + z_{I_2}),\ -(\Delta_{53} + z_{I_3}),\ -(\Delta_{53} + z_{I_4})$$

of widths

$$|z_{R_1}|,\ |z_{R_2}|,\ |z_{R_3}|,\ |z_{R_4}|$$

We shall not proceed any further with this solution but concentrate on the resonance case when

$$J(s) = (s + (\gamma_{32} + \gamma_5))K(s) \tag{6.B.86}$$

where $K(s) = s^3 + as^2 + bs + c = (s-s_1)(s-s_2)(s-s_3)$

and
$$a = 3/2(\gamma_{32} + \gamma_5)$$
$$b = 1/2(\gamma_{32} + \gamma_5)^2 + \gamma_{32}\gamma_5 + G^2$$
$$c = 1/2(\gamma_{32} + \gamma_5)\gamma_{32}\gamma_5 + 1/2(\gamma_{32} + \gamma_5)G^2 \tag{6.B.87}$$

If we let $y = s + 1/3\,a$, then
$$J(y) = y^3 + 3Hy + G$$
$$H = 1/3(-1/3\,a^2 + b)$$
$$G = 2/27\,a^3 - 1/3\,ab + c \tag{6.B.88}$$

Finally, on proceeding as in Appendix III, we obtain the three roots

$$s_1 = u + iv = (\alpha + \beta) - 1/3\,a$$
$$s_2 = w + ix = -\{[1/2(\alpha+\beta) + 1/3\,a] - i\,\sqrt{3}/2\,(\alpha - \beta)\}$$
$$s_3 = y + iz = -\{[1/2(\alpha+\beta) + 1/3\,a] + i\,\sqrt{3}/2\,(\alpha - \beta)\}$$

where
$$\alpha = \left\{\frac{-G + \sqrt{G^2 + 4H^3}}{2}\right\}^{1/3}$$
$$\beta = \left\{\frac{-G - \sqrt{G^2 + 4H^3}}{2}\right\}^{1/3} \tag{6.B.89}$$

$$G^2 + 4H^3 = (1/2\,G_5^2)^2\gamma_{53}^2 + 4/27\{G_5^2 - 1/4(\gamma_{32} - \gamma_5)^2\}^3 = (1/2\,G_5^2)^2\gamma_{53}^2 + 4/27\{G_5^2 - 1/4(\gamma_5 - \gamma_{32})^2\}^3 \tag{6.B.90}$$

$$G = -1/2\,G_5^2\,\gamma_{53} \tag{6.B.91}$$

If $G^2 << 1/4(\gamma_5 - \gamma_{32})^2$, i.e. the small perturbation limit, then we can expand

$$G^2 + 4H^3 = (G_5^2)^2\,1/4\,\gamma_{53}^2 + 4/27\{-1/4(\gamma_5 - \gamma_{32})^2[1 - \frac{G_5^2}{1/4(\gamma_5 - \gamma_{32})^2}]\}^3$$

using the Binomial theorem:-

$$G^2+4H^3 \doteq (G_5)^2\, \tfrac{1}{4}\gamma_{53}^2 \;-\; \tfrac{1}{27}\,\tfrac{1}{16}\,(\gamma_5-\gamma_{32})^6\left\{1-3\,\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\right\}$$

to 1st order in $\dfrac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}$

$$\doteq -\frac{(\gamma_5-\gamma_{32})^6}{16.27}\left\{1-3\,\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})}\right\}, \text{ if } \frac{\gamma_{53}^2}{(\gamma_5-\gamma_{32})^2}$$ is small enough not to make the 3rd term large

$$(G^2+4H^3)^{1/2} \doteq \frac{i}{12\sqrt{3}}(\gamma_5-\gamma_{32})^3\left\{1-\frac{3}{2}\,\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\right\} \tag{6.B.92}$$

$$\therefore \alpha = \left\{\frac{\tfrac{1}{2}G_5^2\gamma_{53} + \tfrac{i}{12\sqrt{3}}(\gamma_5-\gamma_{32})^3\left[1-\tfrac{3}{2}\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\right]}{2}\right\}^{1/3}$$

$$\beta = \left\{\frac{\tfrac{1}{2}G_5^2\gamma_{53} - \tfrac{i}{12\sqrt{3}}(\gamma_5-\gamma_{32})^3\left[1-\tfrac{3}{2}\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\right]}{2}\right\} \tag{6.B.93}$$

$$\therefore \alpha = \left\{\frac{\tfrac{i}{12\sqrt{3}}(\gamma_5-\gamma_{32})^3\left\{1-\tfrac{3}{2}\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2} + \frac{6\sqrt{3}}{i(\gamma_5-\gamma_{32})^3}G_5^2\gamma_{53}\right.}{2}\right\}^{1/3}$$

$$\alpha \doteq \frac{1}{2\sqrt{3}}(\gamma_5-\gamma_{32})(i)^{1/3}\left\{1-\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\,\frac{3}{2}\left(1+\sqrt{3}\,i\left(\frac{\gamma_{53}}{\gamma_5-\gamma_{32}}\right)\right)\right\}^{1/3}$$

Assuming $\left\{1+\sqrt{3}\,i\left(\dfrac{\gamma_{53}}{\gamma_5-\gamma_{32}}\right)\right\}^2$ small enough not to make $\left\{\dfrac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\right\}^2$ large then

$$\alpha \doteq \frac{1}{2\sqrt{3}}(\gamma_5-\gamma_{32})(i)^{1/3}\left\{1-\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\,\frac{1}{2}\left(1+\sqrt{3}\,i\,\frac{\gamma_{53}}{\gamma_5-\gamma_{32}}\right)\right\}$$

$$\beta \doteq \frac{-1}{2\sqrt{3}}(\gamma_5-\gamma_{32})(i)^{1/3}\left\{1-\frac{G_5^2}{\tfrac{1}{4}(\gamma_5-\gamma_{32})^2}\,\frac{1}{2}\left(1-\sqrt{3}\,i\,\frac{\gamma_{53}}{\gamma_5-\gamma_{32}}\right)\right\}$$

taking $(-i)^{1/3} = -1$

Now $(i)^{1/3}$ can be evaluated by de Moivre's theorem

$$(i)^{1/3} = (\cos \pi/2 + i \sin \pi/2)^{1/3}$$
$$= \cos(\pi/6 + 2/3\, r\pi) + i \sin(\pi/6 + 2/3\, r\pi) \qquad \text{where } r = 0,1,2$$

$$(i)^{1/3} = \tfrac{1}{2}\sqrt{3} + \tfrac{1}{2} i \quad \text{for } r = 0$$
$$(i)^{1/3} = -\tfrac{1}{2}\sqrt{3} + \tfrac{1}{2} i \quad \text{for } r = 1$$
$$(i)^{1/3} = -i \quad \text{for } r = 2$$

Considering the simplest value $(i)^{1/3} = -i$

$$\alpha = \frac{-i}{2\sqrt{3}} (\gamma_5 - \gamma_{32}) \left\{ 1 - \frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \left(1 + \sqrt{3} i \frac{\gamma_{52}}{\gamma_5 - \gamma_{32}} \right) \right\} \tag{6.B.94}$$

$$\beta = \frac{i}{2\sqrt{3}} (\gamma_5 - \gamma_{32}) \left\{ 1 - \frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \left(1 - \sqrt{3} i \frac{\gamma_{53}}{\gamma_5 - \gamma_{32}} \right) \right\} \tag{6.B.95}$$

$$\alpha + \beta = - \frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \frac{1}{2} \left\{ \gamma_5 - i \frac{1}{\sqrt{3}} (\gamma_5 - \gamma_{32}) \right\} \tag{6.B.96}$$

$$\alpha - \beta = -\frac{i}{\sqrt{3}} (\gamma_5 - \gamma_{32}) \left\{ 1 - \frac{1}{2} \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) \right\} \tag{6.B.97}$$

$$s_1 = u + iv = -\frac{1}{2} \left\{ (\gamma_{32} + \gamma_5) + \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) \gamma_5 \right\} + i \left\{ \frac{1}{2\sqrt{3}} (\gamma_5 - \gamma_{32}) \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) \right\} \tag{6.B.98}$$

$$s_2 = w + ix = -\left\{ 1 - \frac{1}{4} \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) \right\} \gamma_{32} - i \left\{ \frac{1}{4\sqrt{3}} \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) (\gamma_5 - \gamma_{32}) \right\} \tag{6.B.99}$$

$$s_3 = y + iz = -\left\{ \gamma_5 - \frac{1}{4} \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) (2\gamma_5 - \gamma_{32}) \right\} - i \left\{ \frac{1}{4\sqrt{3}} \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \right) (\gamma_5 - \gamma_{32}) \right\} \tag{6.B.100}$$

i.e. $X = Z$ (6.B.101)

$$J(s) = (s + (\gamma_{32} + \gamma_5))(s + (u+iv))(s + (w+ix))(s + (y+iz)) \tag{6.B.102}$$

$$J\left(-i(\nu''+\Delta_{53})\right) = \left((\gamma_{32}+\gamma_5) - i(\nu''+\Delta_{53})\right)\left(u + i(\nu''+\Delta_{53}+V)\right) \times \left(w + i(\nu''+\Delta_{53}+X)\right)\left(y + i(\nu''+\Delta_{53}+X)\right) \tag{6.B.103}$$

and hence 2 peaks are coincident at $\nu'' = -(\Delta_{53}+X) = -X$

at resonance and the other 2 occur at $\nu'' = -\Delta_{53} = 0$ and $\nu'' = -(\Delta_{53}+V) = -V$

I.E. 2 peaks are at

$$\nu_1'' = \frac{1}{4\sqrt{3}} \frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} (\gamma_5-\gamma_{32}) \tag{6.B.104}$$

and the 2 others are at

$$\nu_2'' = 0$$

$$\nu_3'' = \frac{-1}{2\sqrt{3}} (\gamma_5-\gamma_{32}) \frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2} \tag{6.B.105}$$

The 2 coincident peaks have widths $|w|$ and $|y|$

i.e. $\gamma_{32}\left(1 - \frac{1}{4}\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2}\right)$ and $\left(\gamma_5 - \frac{1}{4}\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2}(2\gamma_5-\gamma_{32})\right)$ (6.B.106)

and so the total width is

$$\Delta_1 = (\gamma_{32}+\gamma_5) - \left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2}\right)\frac{\gamma_5}{2} \tag{6.B.107}$$

The other 2 peaks have widths

$$\Delta_2 = \gamma_{32}+\gamma_5 \quad \text{and} \quad \Delta_3 = |u| = \frac{1}{2}\left\{(\gamma_{32}+\gamma_5) + \gamma_5\left(\frac{G_5^2}{1/4(\gamma_5-\gamma_{32})^2}\right)\right\} \tag{6.B.108}$$

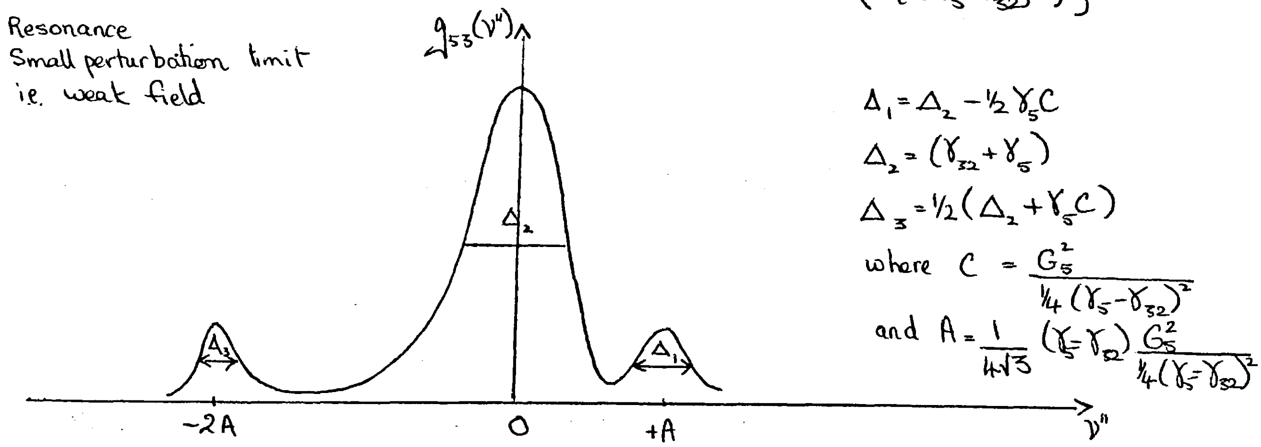


Fig. 6.B.6

I.E. side peaks areasymmetrically spaced, the left-hand one being both further from the central peak and narrower in width than the right-hand one. The central peak is the widest.

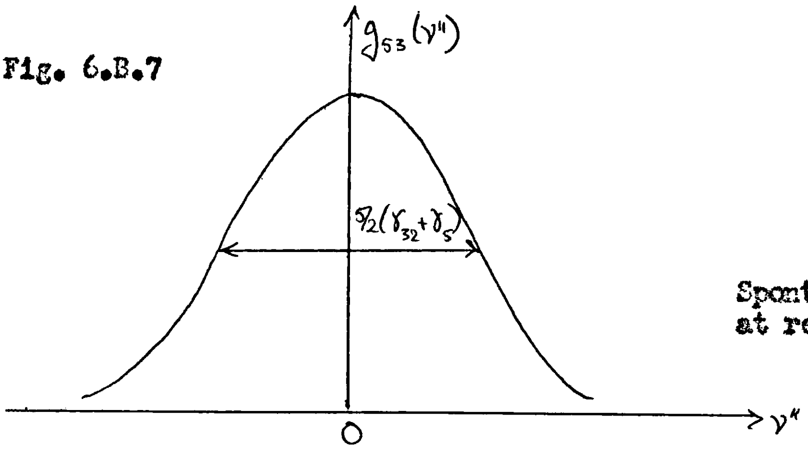
When $G_5 = 0$ (spontaneous emission) all peaks coincide at $\nu'' = 0$ and

$$\left.\begin{aligned} S_1 &= -\tfrac{1}{2}(\gamma_{32}+\gamma_5) \\ S_2 &= -\gamma_{32} \\ S_3 &= -\gamma_5 \\ \therefore\ J(s) &= (s+(\gamma_{32}+\gamma_5))(s+\tfrac{1}{2}(\gamma_{32}+\gamma_5))(s+\gamma_{32})(s+\gamma_5) \\ J(-i\nu'') &= ((\gamma_{32}+\gamma_5)-i\nu'')(\tfrac{1}{2}(\gamma_{32}+\gamma_5)-i\nu'')(\gamma_{32}-i\nu'')(\gamma_5-i\nu'') \\ \tilde{g}_{53}(\nu'') &= \frac{2\left\{(\gamma_{32}\gamma_5-\nu''^2)(\tfrac{1}{4}(\gamma_{32}\gamma_5)^2-\nu''^2)-\nu''^2(\gamma_{32}+\gamma_5)^2\right\}}{\left\{(\gamma_{32}+\gamma_5)^2+\nu''^2\right\}\left\{\tfrac{1}{4}(\gamma_{32}+\gamma_5)^2+\nu''^2\right\}\left\{\gamma_{32}^2+\nu''^2\right\}\left\{\gamma_5^2+\nu''^2\right\}} \end{aligned}\right\} \quad (6.B.109)$$

The width of the central peak is now

$$(\gamma_{32}+\gamma_5)+\tfrac{1}{2}(\gamma_{32}+\gamma_5)+(\gamma_{32}+\gamma_5)$$
$$=\tfrac{5}{2}(\gamma_{32}+\gamma_5)=\tfrac{5}{2}\Delta_2 \qquad (6.B.110)$$

Fig. 6.B.7



Spontaneous emission at resonance

According to Zernik this emission is negligible. In fact

$$\tilde{g}_{53}(0)=\frac{2}{\gamma_{32}\gamma_5(\gamma_{32}+\gamma_5)^2} \qquad (6.B.111)$$

which is in reality very small, as was to be expected.

If $G_5^2 >> \frac{1}{4}(\gamma_5-\gamma_{32})^2$ i.e. the strong field limit, then to zeroeth order in $\frac{1}{4}(\gamma_5-\gamma_{32})^2/G_5^2$ we have

$$G^2 + 4H^3 \doteq (G_5^2)^2\, \tfrac{1}{4}\gamma_{53}^2 + \tfrac{4}{27}(G_5^2)^3$$

$$= \tfrac{4}{27}(G_5^2)^3\left\{1 + \tfrac{27}{16}\gamma_{53}^2/G_5^2\right\}$$

$$(G^2+4H^3)^{\frac{1}{2}} \doteq \tfrac{2}{3\sqrt{3}}G_5^3\left\{1 + \tfrac{27}{32}\gamma_{53}^2/G_5^2\right\} \quad \text{to 2nd order in } \gamma_{53}/G_5 \qquad (6.B.112)$$

$$\alpha = \left\{\frac{\tfrac{1}{2}\gamma_{53}G_5^2 + \tfrac{2}{3\sqrt{3}}G_5^3\left(1+\tfrac{27}{32}\gamma_{53}^2/G_5^2\right)}{2}\right\}^{1/3} = \left\{\frac{\tfrac{2}{3\sqrt{3}}G_5^3\left(1+\tfrac{3\sqrt{3}}{4}\gamma_{53}/G_5 + \tfrac{27}{32}\gamma_{53}^2/G_5^2\right)}{2}\right\}^{1/3} \qquad (6.B.113)$$

$$\beta = \left\{\frac{\tfrac{1}{2}\gamma_{53}G_5^2 - \tfrac{2}{3\sqrt{3}}G_5^3\left(1+\tfrac{27}{32}\gamma_{53}^2/G_5^2\right)}{2}\right\}^{1/3} = -\left\{\frac{\tfrac{2}{3\sqrt{3}}G_5^3\left(1-\tfrac{3\sqrt{3}}{4}\gamma_{53}/G_5 + \tfrac{27}{32}\gamma_{53}^2/G_5^2\right)}{2}\right\}^{1/3}$$

where we have taken $(-1)^{1/3} = -1$ i.e. the cube root of (-1) to be real.

According to Zernik, for very intense fields spontaneous decay from 3P to 2S may be neglected showing that γ_{53} is very small in comparison with G_5 so that we shall consider (γ_{53}/G_5) to 1st order only.

$$\left.\begin{aligned} \therefore\ \alpha &\doteq \left\{\tfrac{1}{3\sqrt{3}}G_5^3\left(1+\tfrac{3\sqrt{3}}{4}\gamma_{53}/G_5\right)\right\}^{1/3} \doteq \tfrac{1}{\sqrt{3}}G_5\left\{1+\tfrac{\sqrt{3}}{4}\gamma_{53}/G_5\right\} \\ \beta &\doteq -\left\{\tfrac{1}{3\sqrt{3}}G_5^3\left(1-\tfrac{3\sqrt{3}}{4}\gamma_{53}/G_5\right)\right\}^{1/3} \doteq -\tfrac{1}{\sqrt{3}}G_5\left\{1-\tfrac{\sqrt{3}}{4}\gamma_{53}/G_5\right\} \end{aligned}\right\} \quad (6.B.114)$$

$$\left.\begin{aligned} \alpha+\beta &= \tfrac{1}{2}\gamma_{53} \\ \alpha-\beta &= \tfrac{2}{\sqrt{3}}G_5 \end{aligned}\right\} \quad (6.B.115)$$

[If we were to have kept (γ_{53}/G_5) to 2nd order, then

$$\alpha \doteq \tfrac{1}{\sqrt{3}}G_5\left\{1 + \tfrac{\sqrt{3}}{4}\gamma_{53}/G_5 + \tfrac{3}{32}\gamma_{53}^2/G_5^2\right\}$$

$$\beta \doteq -\tfrac{1}{\sqrt{3}}G_5\left\{1 - \tfrac{\sqrt{3}}{4}\gamma_{53}/G_5 + \tfrac{3}{32}\gamma_{53}^2/G_5^2\right\}$$

$$\alpha + \beta = \tfrac{1}{2}\gamma_{53} \tag{6.B.116}$$

$$\alpha - \beta = \tfrac{2}{\sqrt{3}} G_5 \left\{ 1 + \tfrac{3}{32} \, \gamma_{53}^2 / G_5^2 \right\} \tag{6.B.117}$$

$$\left.\begin{aligned}
\therefore S_1 = u + iv &= \tfrac{1}{2}\gamma_{53} - \tfrac{1}{2}(\gamma_{32} - \gamma_5) = -\tfrac{1}{2}(\gamma_{32} + \gamma_{51}) \\
S_2 = w + ix &= -\left\{ \left[\tfrac{1}{4}\gamma_{53} + \tfrac{1}{2}(\gamma_{32} + \gamma_5) \right] - iG_5 \right\} = -\left\{ \tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) - iG_5 \right\} \\
S_3 = y + iz &= -\left\{ \left[\tfrac{1}{4}\gamma_{53} + \tfrac{1}{2}(\gamma_{32} + \gamma_5) \right] + iG_5 \right\} = -\left\{ \tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) + iG_5 \right\} \\
\therefore u &= -\tfrac{1}{2}(\gamma_{32} + \gamma_{51}) \\
w &= y = -\tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) \\
x &= -z = G_5
\end{aligned}\right\} \tag{6.B.118}$$

Now

$$\left.\begin{aligned}
J(s) &= (s + (\gamma_{32} + \gamma_5))\, K(s) \\
&= (s + (\gamma_{32} + \gamma_5))(s - s_1)(s - s_2)(s - s_3)
\end{aligned}\right\} \tag{6.B.119}$$

$$J(-i(\nu - \omega_L)) = -\left\{ (\gamma_{32} + \gamma_5) - i(\nu - \omega_L) \right\}\left\{ s_1 + i(\nu - \omega_L) \right\}\left\{ s_2 + i(\nu - \omega_L) \right\}\left\{ s_3 + i(\nu - \omega_L) \right\}$$

$$\begin{aligned}
= &-\left\{ (\gamma_{32} + \gamma_5) - i(\nu - \omega_L) \right\}\left\{ -\tfrac{1}{2}(\gamma_{32} + \gamma_{51}) + i(\nu - \omega_L) \right\} \\
&\times \left\{ -\tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) + i(\nu - \omega_L + G_5) \right\}\left\{ -\tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) + i(\nu - \omega_L - G_5) \right\}
\end{aligned}$$

$$\left.\begin{aligned}
\therefore J(-i(\nu'' + \Delta_{53})) = &-\left\{ (\gamma_{32} + \gamma_5) - i(\nu'' + \Delta_{53}) \right\}\left\{ -\tfrac{1}{2}(\gamma_{32} + \gamma_{51}) + i(\nu'' + \Delta_{53}) \right\} \\
&\times \left\{ -\tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) + i(\nu'' + \Delta_{53} + G_5) \right\} \\
&\times \left\{ -\tfrac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5)) + i(\nu'' + \Delta_{53} - G_5) \right\}
\end{aligned}\right\} \tag{6.B.120}$$

Thus the 2 of the peaks are coincident at $\nu'' = -\Delta_{53}$ of widths $(\gamma_{32} + \gamma_5)$ and $\tfrac{1}{2}(\gamma_{32} + \gamma_{51})$ and the remaining are at

$\nu'' = -(\Delta_{53} - G_5)$, $\nu'' = -(\Delta_{53} + G_5)$ each of width

$\Delta = \frac{1}{4}(\gamma_{53} + 2(\gamma_{32} + \gamma_5))$ where of course $\Delta_{53} = 0$ as we are

considering resonance

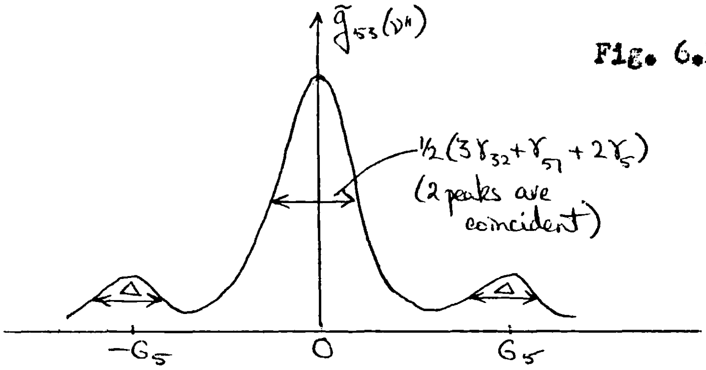


Fig. 6.B.8

Resonance

$G_5^2 >> \frac{1}{4}(\gamma_5 \gamma_{52})^2$

$G_5^2 >> \gamma_{53}^2$

i.e. strong field case

The expression for $\tilde{g}_{53}(\nu'')$ is

$$\tilde{g}_{53}(\nu'') = \frac{2\{[\gamma_{32}\gamma_5 - \nu''^2][\frac{1}{4}(\gamma_{32}+\gamma_5)^2 - \nu''^2] - \nu''^2(\gamma_{32}+\gamma_5)^2 + G_5^2[\frac{1}{4}(\gamma_{32}+\gamma_5)^2 - \nu''^2]\}}{[\{(\gamma_{32}+\gamma_5)^2 + \nu''^2\}\{\frac{1}{4}(\gamma_{32}+\gamma_{51})^2 + \nu''^2\}\{\frac{1}{16}(\gamma_{53}+2(\gamma_{32}+\gamma_5))^2 + (\nu''+G_5)^2\} \times \{\frac{1}{16}(\gamma_{53}+2(\gamma_{32}+\gamma_5))^2 + (\nu''-G_5)^2\}]} \quad (6.B.121)$$

$$= \frac{2\,\frac{1}{4}(\gamma_{32}+\gamma_5)^2\{(\gamma_{32}\gamma_5 - \nu''^2)(1 - \frac{\nu''^2}{\frac{1}{4}(\gamma_{32}+\gamma_5)^2}) - 4\nu''^2 + G_5^2(1 - \frac{\nu''^2}{\frac{1}{4}(\gamma_{32}+\gamma_5)^2})\}}{|J(-i\nu'')|^2}$$

N.B. Rautian and Sobelman also obtain three peaks in the case $G^2 > \frac{(\gamma_3-\gamma_5)^2}{4}$ but in their case all peaks have equal width. They are, however, equally spaced about the central peak as ours are also. They say that for discernable splitting, comparatively strong saturation is required, i.e. $G^2 >> \gamma_3\gamma_5$.

C. Discussion

The triplet nature of the 5 $\rightarrow$ 3, i.e. 3P to 2S, decay for resonance and for strong, or even not so strong, fields is owing to the field splitting of level 5 (3P) explained in the previous chapter. The separation of the side peaks is small for weak fields so that the effect is not noticeable and probably only the central peak is seen. In fact, this transition is only important for strong fields and the spontaneous decay has been shown to be negligible. For non-resonance we have seen that a 4th peak is discernable though it may be possible that in the limit of a strong field this may reduce to 3 peaks.

In the case of the 5 $\to$ 1, i.e. 3P $\to$ 1S decay, when resonance conditions apply a single line is seen and the splitting is only noticeable for the non-resonance case. In the latter case 2 peaks are always seen, regardless of the field strength, and these are always symmetric about $\nu' = -\frac{1}{2}\Delta_{53}$. Both peaks are generally shifted towards low frequency except in the case of spontaneous decay when no field is applied when, naturally, the peak of width $\frac{1}{2}\gamma_5$, corresponding to the resonant transition 5 $\to$ 1, occurs at $\nu' = 0$. The splitting is field dependent and is largest for strong fields. The absence of a 3rd peak is owing to the fact that we consider the non-resonance situation and the 2 peaks owe their origin to the field splitting of level 5 (3P).

The spectral profiles calculated in this chapter represent emission only since in both cases the atom was assumed to be in state $|5>$ at time t' = 0 (the initial time). Hence only single transitions can be considered and in this method one cannot assume the atom to be initially in level 3 without obtaining zero spectral profiles.

We have, in fact, not assumed any conditions with respect to the magnitudes of the relative decay rates, only their magnitudes with respect to the size of the field so that we have removed the restrictions of Zernik's paper that $\gamma_{53} = 0$ and Rautian and Sobelman's paper, that $\gamma_{32} >> \gamma_{53}$, though in the case of large fields in Section (b) we assumed $(\gamma_{53}/G_5)^2$ negligible and for weak fields $(\gamma_{53}/(\gamma_5 - \gamma_{32}))^2$ to be small.

D. Addendum

If $t' = 0$ equations (6.B.26) and (6.B.27) become

$$\left.\begin{aligned} f_1(s)\,\hat{x}(s) &= i\lambda\mathcal{E}_{0L}\,\hat{y}(s) + x(t')e^{-st'} \\ f_2(s)\,\hat{y}(s) &= i\lambda\mathcal{E}_{0L}\,\hat{x}(s) + y(t')e^{-st'} \end{aligned}\right\} \quad (6.D.1)$$

where $\hat{x}(s) = \hat{\rho}_{51}(s - i\omega_L)$

$\hat{y}(s) = \hat{\rho}_{31}(s)$

Hence

$$\left.\begin{aligned} \hat{x}(s) &= \frac{i\lambda\mathcal{E}_{0L}}{F(s)}\,e^{-st'}\,y(t') + \frac{f_2(s)}{F(s)}\,x(t')\,e^{-st'} \\ \text{i.e.}\quad \hat{\rho}_{51}(s-i\omega_L) &= \frac{i\lambda\mathcal{E}_{0L}}{F(s)}\,e^{-st'}\rho_{31}(t') + \frac{f_2(s)}{F(s)}\,e^{(s-i\omega_L)t'}\rho_{57}(t') \end{aligned}\right\} \quad (6.D.2)$$

Hence

$$\tilde{g}_{57}(\nu) = 2\,\mathrm{Re}\left\{\hat{\psi}_{57,57}(-i\nu)\bar{\rho}_{55} + \hat{\psi}_{57,31}(-i\nu)\bar{\rho}_{35}\right\}$$

$$= 2\bar{\rho}_{55}\,\mathrm{Re}\,\hat{\psi}_{57,57}(-i\nu) + 2\,\mathrm{Re}\left(\hat{\psi}_{57,31}(-i\nu)\bar{\rho}_{35}\right)$$

where $\bar{\rho}_{55} = (\rho_{55\infty})_{t=t'}$ and is real

$\bar{\rho}_{35} = (\rho_{35\infty})_{t=t'}\,e^{-i\omega_L t'}$ and is complex

$$\hat{\psi}_{51,57}(-i\nu) = \frac{f_2(-i(\nu-\omega_L))}{F(-i(\nu-\omega_L))}$$

$$\hat{\psi}_{51,31}(-i\nu) = \frac{i\lambda\mathcal{E}_{0L}}{F(-i(\nu-\omega_L))} \quad (6.D.3)$$

In order to find $\tilde{g}_{57}(\nu)$ explicitly therefore values of $\bar{\rho}_{55}$ and $\bar{\rho}_{35}$ must be found and these are also required for the calculation of $\tilde{g}_{53}(\nu)$ since

$$\left.\begin{aligned}\tilde{g}_{53}(\;) &= 2\,\mathrm{Re}\left\{\hat{\psi}_{53,33}(-i\nu)\bar{\rho}_{35} + \hat{\psi}_{53,53}(-i\nu)\bar{\rho}_{55}\right\} \\ &= 2\,\mathrm{Re}\left(\hat{\psi}_{53,33}(-i\nu)\bar{\rho}_{35}\right) + 2\bar{\rho}_{55}\,\mathrm{Re}\,\hat{\psi}_{53,53}(-i\nu)\end{aligned}\right\} \quad (6.D.4)$$

where $\hat{\psi}_{53,33}(-i\nu) = \dfrac{i\lambda\mathcal{E}_{oL}\left(\gamma_5 - i(\nu-\omega_L)\right)}{W(-i(\nu-\omega_L))}\left\{C(-i(\nu-\omega_L)) - \tfrac{1}{2}C_5^{\,i}\left(\tfrac{1}{2}(\gamma_{32}+\gamma_{5}) - i(\nu-\omega_L)\right)\right\}$

and $\hat{\psi}_{53,53}(-i\nu) = \dfrac{\left(\gamma_{32} - i(\nu-\omega_L)\right)\left(\gamma_5 - i(\nu-\omega_L)\right)}{W(-i(\nu-\omega_L))}$

From equation (6.B.56)

$$\left.\begin{aligned}\hat{x}(s) &= -\frac{i\lambda\mathcal{E}_{oL}}{f_3(s)}\,\hat{y}(s) + \frac{i\lambda\mathcal{E}_{oL}}{f_3(s)}\,\hat{z}(s) + \frac{x(t')}{f_3(s)}\,e^{-st'}\end{aligned}\right\} \quad (6.D.5)$$

where $f_3(s) = s + \tfrac{1}{2}(\gamma_{32}+\gamma_5) - i\Delta_{53}$

$$\therefore \hat{\rho}_{35}(s+i\omega_L) = -\frac{i\lambda\mathcal{E}_{oL}}{f_3(s)}\,\hat{\rho}_{33}(s) + \frac{i\lambda\mathcal{E}_{oL}}{f_3(s)}\,\hat{\rho}_{55}(s) + \frac{1}{f_3(s)}\,e^{-st'}\rho_{35}(t')e^{-i\omega_L t'} \quad (6.D.6)$$

$$\therefore \hat{\rho}_{35}(s)_{equil.} = \frac{-i\lambda\mathcal{E}_{oL}}{f_3(s-i\omega_L)}\,\frac{\rho_{33}(\infty)}{s-i\omega_L} + \frac{i\lambda\mathcal{E}_{oL}}{f_3(s-i\omega_L)}\,\frac{\rho_{55}(\infty)}{s-i\omega_L} + \frac{1}{f_3(s-i\omega_L)}\,e^{-st'}\rho_{35}(t') \quad (6.D.7)$$

$$\left.\begin{aligned}\therefore \left(\rho_{35}(\infty)\right)_{t=t'} &= e^{i\omega_L t'}\left\{\frac{-i\lambda\mathcal{E}_{oL}}{f_3(s-i\omega_L)}\,\rho_{33}(\infty) + \frac{i\lambda\mathcal{E}_{oL}}{f_3(s-i\omega_L)}\,\rho_{55}(\infty)\right\} \\ &= \frac{e^{i\omega_L t'}\,i\lambda\mathcal{E}_{oL}}{\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i(2\Delta-\omega_3')}\left(\rho_{55}(\infty) - \rho_{33}(\infty)\right) \\ &= e^{i\omega_L t'}\,\bar{\rho}_{35}\end{aligned}\right\} \quad (6.D.8)$$

Hence

$$\hat{g}_{51}(\nu) = 2\bar{\rho}_{55}\,\mathrm{Re}\,\hat{\psi}_{51,51}(-i\nu) + 2\,\mathrm{Re}\left\{\frac{i\lambda\mathcal{E}_{oL}}{\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i(2\Delta-\omega_3')}\left(\bar{\rho}_{55}-\bar{\rho}_{33}\right)\hat{\psi}_{51,31}(-i\nu)\right\} \quad (6.D.9)$$

and

$$\hat{g}_{53}(\nu) = 2\bar{\rho}_{55}\,\mathrm{Re}\,\hat{\psi}_{53,53}(-i\nu) + 2\,\mathrm{Re}\left\{\frac{i\lambda\mathcal{E}_{oL}}{\tfrac{1}{2}(\gamma_{32}+\gamma_5) - i(2\Delta-\omega_3')}\left(\bar{\rho}_{55}-\bar{\rho}_{33}\right)\hat{\psi}_{53,33}(-i\nu)\right\} \quad (6.D.10)$$

give the spectral profiles for the scattered light for the relevant transitions and would give the overall effect of all possible transitions as in the previous chapter.

CHAPTER VII

AN ATOM WITH TWO EXCITED STATES (ALLOWANCE BEING MADE FOR THE POSSIBILITY OF THEIR OVERLAPPING) DRIVEN BY AN EXTERNAL FIELD COUPLING THE UPPERMOST EXCITED STATE TO THE GROUND STATE

We shall consider a 3-level atom with ground level 1, and two excited levels, 2 and 3, as indicated below

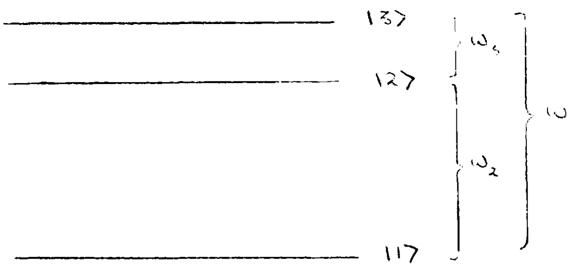


Fig 1

where we define the frequency separations as follows:

$$\omega_s = \epsilon_3 - \epsilon_2 = \epsilon_{32}$$

$$\omega_2 = \epsilon_2 - \epsilon_1 = \epsilon_{21}$$

$$\omega = \epsilon_3 - \epsilon_1 = \epsilon_{31}$$

A. Equations of motion

Since we wish now to consider the case when levels 3 and 2 may be overlapping it is necessary to re-derive the Heisenberg equations of motion since those of Chapter IV refer only to the case when there is no overlapping.

We recall that for a 3-level atom, the Hamiltonian in the dipole and rotating-wave approximations for an atom at the origin of coordinates is, according to equation (4.A.3)

$$H = \hbar \sum_{x=1}^{3} \epsilon_x P_{xx} + \hbar \sum_{ls} \omega_l a_{ls}^{+} a_{ls} - \hbar \sum_{y=z+1}^{3} \sum_{z=1}^{2} g_{ls,yz} \left(P_{zy}^{+} a_{ls} + a_{ls}^{+} P_{zy} \right) \qquad (7.A.1)$$

or, more fundamentally, before RWA is assumed, according to equation (4.A.1)

$$H = \hbar \sum_{x=1}^{3} \epsilon_x P_{xx} + \frac{1}{8\pi} \int \left(|\underline{E}|^2 + |\underline{B}|^2 \right) d^3 r - \underline{E} \cdot \sum_{y=z+1}^{3} \sum_{z=1}^{2} \underline{p} \left(P_{zy} + P_{zy}^{+} \right) \qquad (7.A.2)$$

Hence, from equation (4.A.4)

$$\dot{a}_{l\sigma}(t) = -i\omega_l a_{l\sigma}(t) + i\sum_{y=\ldots}^{3}\sum_{z} g_{l\sigma,yz} P_{zy}(t) \qquad (7.A.3)$$

with formal solution, according to equation (4.A.7)

$$a_{l\sigma}(t) = a_{l\sigma}(0)\, e^{-i\omega_l t} + i\sum_{y=\ldots}^{3}\sum_{z} g_{l\sigma,yz}\int_0^t dt'\, e^{-i\omega_l (t-t')} P_{zy}(t') \qquad (7.A.4)$$

The other equations of motion are, from equations (4.A.5), (4.A.6),

for $m < n$

$$\begin{aligned}\dot{P}_{mn}(t) = {} & i\epsilon_{mn} P_{mn}(t) - i\sum_{y=m+1}^{3}\sum_{l\sigma} g_{l\sigma,yn} P_{yn}(t)\, a_{l\sigma}(t) + i\sum_{z=1}^{n-1}\sum_{l\sigma} g_{l\sigma,nz} P_{mz}(t)\, a_{l\sigma}(t) \\ & - i\sum_{z=1}^{m-1}\sum_{l\sigma} g_{l\sigma,mz}\, a^{+}_{l\sigma}(t)\, P_{zn}(t) + i\sum_{y=m+1}^{3}\sum_{l\sigma} g_{l\sigma,yz}\, a^{+}_{l\sigma}(t)\, P_{ny}(t) \\ = {} & P^{+}_{nm}(t)\end{aligned} \qquad (7.A.5)$$

and

$$\begin{aligned}\dot{P}_{mm}(t) = {} & -i\sum_{y=m+1}^{3}\sum_{l\sigma} g_{l\sigma,ym} P^{+}_{my}(t)\, a_{l\sigma}(t) + i\sum_{z=1}^{m-1}\sum_{l\sigma} g_{l\sigma,mz} P^{+}_{zm}(t)\, a_{l\sigma}(t) \\ & + i\sum_{y=m+1}^{3}\sum_{l\sigma} g_{l\sigma,ym}\, a^{+}_{l\sigma}(t)\, P_{my}(t) - i\sum_{z=1}^{m-1}\sum_{l\sigma} g_{l\sigma,mz}\, a^{+}_{l\sigma}(t)\, P_{zm}(t)\end{aligned} \qquad (7.A.6)$$

Thus the 9 transition operator equations of motion are:-

$$\dot{P}_{11}(t) = -i\sum_{y=2}^{3} P^{+}_{1y}(t)\left\{\sum_{l\sigma} g_{l\sigma,y1}\, a_{l\sigma}(t)\right\} + i\sum_{y=2}^{3}\left\{\sum_{l\sigma} g_{l\sigma,y1}\, a^{+}_{l\sigma}(t)\right\} P_{1y}(t) \qquad (7.A.7)$$

$$\begin{aligned}\dot{P}_{22}(t) = {} & -i P^{+}_{23}(t)\left\{\sum_{l\sigma} g_{l\sigma,32}\, a_{l\sigma}(t)\right\} + i P^{+}_{12}(t)\left\{\sum_{l\sigma} g_{l\sigma,21}\, a_{l\sigma}(t)\right\} \\ & + i\left\{\sum_{l\sigma} g_{l\sigma,32}\, a^{+}_{l\sigma}(t)\right\} P_{23}(t) - i\left\{\sum_{l\sigma} g_{l\sigma,21}\, a^{+}_{l\sigma}(t)\right\} P_{12}(t)\end{aligned} \qquad (7.A.8)$$

$$\dot{P}_{33}(t) = i\sum_{z=1}^{2} P^{+}_{z3}(t)\left\{\sum_{l\sigma} g_{l\sigma,3z}\, a_{l\sigma}(t)\right\} - i\sum_{z=1}^{2}\left\{\sum_{l\sigma} g_{l\sigma,3z}\, a^{+}_{l\sigma}(t)\right\} P_{z3}(t) \qquad (7.A.9)$$

$$\begin{aligned}\dot{P}_{12}(t) = {} & i\omega_2 P_{12}(t) - i\sum_{y=2}^{3} P_{y2}(t)\left\{\sum_{l\sigma} g_{l\sigma,y1}\, a_{l\sigma}(t)\right\} + i P_{11}(t)\left\{\sum_{l\sigma} g_{l\sigma,21}\, a_{l\sigma}(t)\right\} \\ & + i\left\{\sum_{l\sigma} g_{l\sigma,32}\, a^{+}_{l\sigma}(t)\right\} P_{13}(t)\end{aligned} \qquad (7.A.10)$$

$$\dot{P}_{13}(t) = -i\omega P_{13}(t) - i\sum_{y=2} P_{y3}(t)\Big\{\sum_{k\sigma} g_{k\sigma,y1}\, a_{k\sigma}(t)\Big\} + i\sum_{z=1} P_{1z}(t)\Big\{\sum_{k\sigma} g_{k\sigma,3z}\, a_{k\sigma}(t)\Big\} \tag{7.A.11}$$

$$\left.\begin{aligned}\dot{P}_{23}(t) = &-i\omega_3 P_{23}(t) - i P_{33}(t)\Big\{\sum_{k\sigma} g_{k\sigma,32}\, a_{k\sigma}(t)\Big\} + i\sum_{z=1} P_{2z}(t)\Big\{\sum_{k\sigma} g_{k\sigma,3z}\, a_{k\sigma}(t)\Big\} \\ &- i\Big\{\sum_{k\sigma} g_{k\sigma,21}\, a^{+}_{k\sigma}(t)\Big\} P_{13}(t)\end{aligned}\right\} \tag{7.A.12}$$

$$\dot{P}_{21}(t) = \dot{P}_{12}^{+}(t) \tag{7.A.13}$$

$$\dot{P}_{31}(t) = \dot{P}_{13}^{+}(t) \tag{7.A.14}$$

$$\dot{P}_{32}(t) = \dot{P}_{23}^{+}(t) \tag{7.A.15}$$

Considering equation (7.A.7) and substituting for $a_{k\sigma}(t)$, $a^{+}_{k\sigma}(t)$

$$\left.\begin{aligned}\dot{P}_{11}(t) = &-i\sum_{y=2}^{3} P^{+}_{1y}(t)\Big\{\sum_{k\sigma} g_{k\sigma,y1}\, a_{k\sigma}(0)e^{-i\omega_k t} + i\sum_{k\sigma} g_{k\sigma,y1}\sum_{y'=z'+1}^{3}\sum_{z'=1} g_{k\sigma,y'z'} \\ &\times\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{z'y'}(t')\Big\} \\ &+ i\sum_{y=2}^{3}\Big\{\sum_{k\sigma} g_{k\sigma,y1}\, a^{+}_{k\sigma}(0)e^{i\omega_k t} - i\sum_{k\sigma} g_{k\sigma,y1}\sum_{y'=z'+1}\sum_{z'=1} g_{k\sigma,y'z'} \\ &\times\int_0^t dt'\, e^{i\omega_k(t-t')} P_{y'z'}(t')\Big\} P_{1y}(t)\end{aligned}\right\} \tag{7.A.16}$$

Let $V \longrightarrow \infty$

$$\left.\begin{aligned}\dot{P}_{11}(t) = &-i\sum_{y=2}^{3} P^{+}_{1y}(t)\Big\{\tfrac{1}{\hbar}\, \underline{E}_{+}^{(1)}(t)\cdot \underline{P}_{y1} + i\sum_{y'=z'+1}^{3}\sum_{z'=1}^{2}\sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,y'z'} \\ &\times\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{z'y'}(t')\Big\} \\ &+ i\sum_{y=2}^{3}\Big\{\tfrac{1}{\hbar}\, \underline{E}_{+}^{(1)+}(t)\cdot \underline{P}_{y1} - i\sum_{y'=z'+1}^{3}\sum_{z'=1}^{2}\sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,y'z'}\int_0^t dt' \\ &\times e^{i\omega_k(t-t')} P_{y'z'}(t')\Big\} P_{1y}(t)\end{aligned}\right\} \tag{7.A.17}$$

$$\left.\begin{aligned}\text{Now }&\sum_{y'=z'+1}^{3}\sum_{z'=1}^{2}\sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,y'z'}\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{z'y'}(t') \\ = &\sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,21}\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{12}(t') \\ &+ \sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,31}\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{13}(t') \\ &+ \sum_{\substack{k\sigma \\ V\to\infty}} g_{k\sigma,y1}\, g_{k\sigma,32}\int_0^t dt'\, e^{-i\omega_k(t-t')} P_{23}(t')\end{aligned}\right\} \tag{7.A.18}$$

Since we wish to include the possibility of levels 3 and 2 overlapping we shall include integration over frequencies common to photon transitions between levels 3 and 1 and 2 and 1 and vice versa, as in Chapter III.

Now

$$\sum_j P_j^{+}(t) \sum_{\lambda,\, V\to\infty} g_{\lambda,j}\, g_{\lambda,j'} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{j'}(t')$$

$$\left.\begin{aligned}
&= \sum_{\substack{\lambda \\ V\to\infty}} g^2_{\lambda,21} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{12}^{+}(t)\, P_{12}(t') \\
&+ \sum_{\substack{\lambda \\ V\to\infty}} g_{\lambda,21}\, g_{\lambda,31} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{12}^{+}(t)\, P_{13}(t') \\
&+ \sum_{\substack{\lambda \\ V\to\infty}} g_{\lambda,21}\, g_{\lambda,32} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{12}^{+}(t)\, P_{23}(t') \\
&+ \sum_{\substack{\lambda \\ V\to\infty}} g_{\lambda,31}\, g_{\lambda,21} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{13}^{+}(t)\, P_{12}(t') \\
&+ \sum_{\substack{\lambda \\ V\to\infty}} g^2_{\lambda,31} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{13}^{+}(t)\, P_{13}(t') \\
&+ \sum_{\substack{\lambda \\ V\to\infty}} g_{\lambda,31}\, g_{\lambda,32} \int_0^t dt'\, e^{-i\omega_\lambda(t-t')}\, P_{13}^{+}(t)\, P_{23}(t')
\end{aligned}\right\} \qquad (7.A.19)$$

Since there are no frequencies common to photon transitions between levels 3 and 2 and 2 and 1 or 3 and 2 and 3 and 1, the third and sixth terms go out on taking the limit V

$$\left.\begin{aligned}
\text{LHS} = \;& \frac{2}{3\pi c^3 \hbar}\, p_{21}^2 \int_0^\infty d\omega\, \omega^3 \int_0^t dt'\, e^{-i(\omega-\omega_{21})(t-t')}\, P_{22}(t) \\
+ \;& \frac{2}{3\pi c^3 \hbar}\, (p_{31}\cdot p_{21}) \int_0^\infty d\omega'\, \omega'^3 \int_0^t dt'\, e^{-i(\omega'-\omega)(t-t')}\, P_{23}(t) \\
+ \;& \frac{2}{3\pi c^3 \hbar}\, (p_{31}\cdot p_{21}) \int_0^\infty d\omega'\, \omega'^3 \int_0^t dt'\, e^{-i(\omega-\omega_2)(t-t')}\, P_{32}(t) \\
+ \;& \frac{2}{3\pi c^3 \hbar}\, p_{31}^2 \int_0^\infty d\omega'\, \omega'^3 \int_0^t dt'\, e^{-i(\omega'-\omega)(t-t')}\, P_{33}(t)
\end{aligned}\right\} \qquad (7.A.20)$$

since $P_{mn}(t') \simeq P_{mn}(t)\, e^{i\epsilon_{mn}(t-t')}$ where $n > m$ also, as shown in earlier chapters.

$$\left.\begin{aligned}
\therefore\ \text{LHS} = \;& (\tfrac{1}{2}\gamma_{21} - i\Omega_{21})\, P_{22}(t) + (\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31})\, P_{23}(t) + (\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21})\, P_{32}(t) \\
& + (\tfrac{1}{2}\gamma_{31} - i\Omega_{31})\, P_{33}(t)
\end{aligned}\right\} \qquad (7.A.21)$$

Hence using this and its h.c. in (7.A.17), we obtain

$$\dot{P}_{11}(t) = -\frac{i}{\hbar}\sum_{j}\left\{P^{+}_{j}(t)E^{(+)}(t)\cdot p_{j1} + E^{(+)*}(t)\cdot p_{1j}\,P_{j1}(t)\right\}$$
$$+ \left(\tfrac{1}{2}\gamma_{21} - i\Omega_{21}\right)P_{22}(t) + \left(\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\right)P_{23}(t) + \left(\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\right)P_{32}(t) + \left(\tfrac{1}{2}\gamma_{31} - i\Omega_{31}\right)P_{33}(t)$$
$$+ \left(\tfrac{1}{2}\gamma_{21} + i\Omega_{21}\right)P_{22}(t) + \left(\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\right)P_{32}(t) + \left(\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\right)P_{23}(t) + \left(\tfrac{1}{2}\gamma_{31} + i\Omega_{31}\right)P_{33}(t) \qquad (7.A.22)$$

i.e.
$$\dot{P}_{11}(t) = -i\sum_{j=2}^{3}\left\{P_{j1}(t)\,q_{1j}(t) + q^{+}_{j1}(t)P_{1j}(t)\right\}$$
$$+ \gamma_{21}P_{22}(t) + \gamma_{31}P_{33}(t) + \left\{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31}-\Omega'_{21})\right\}P_{23}(t)$$
$$+ \left\{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31}-\Omega'_{21})\right\}P_{32}(t) \qquad (7.A.23)$$

Using a similar procedure for the remaining nine equations and assuming that no dipole transitions occur between levels 3 and 2 (e.g. as in the case when levels 3 and 2 have the same ℓ value. This situation occurs when they have resulted from the Zeeman splitting of an excited level into two components as the result of the application of a magnetic field) so that

$$\gamma_{32} = q_{32} = q^{+}_{32} = \Omega_{32} = 0$$, and the same for reversed subscripts,

we obtain:-

$$\dot{P}_{11}(t) = -iP_{21}(t)\,q_{21}(t) + iq^{+}_{21}(t)P_{12}(t) - iP_{31}(t)\,q_{31}(t) + iq^{+}_{31}(t)P_{13}(t)$$
$$+ \gamma_{21}P_{22}(t) + \gamma_{31}P_{33}(t) + \left\{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31}-\Omega'_{21})\right\}P_{23}(t)$$
$$+ \left\{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) + i(\Omega'_{31}-\Omega'_{21})\right\}P_{32}(t) \qquad (7.A.24)$$

$$\dot{P}_{22}(t) = iP_{21}(t)\,q_{21}(t) - iq^{+}_{21}(t)P_{12}(t)$$
$$- \gamma_{21}P_{22}(t) - \left\{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\right\}P_{23}(t) - \left\{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\right\}P_{32}(t) \qquad (7.A.25)$$

$$\dot{P}_{33}(t) = i\,P_{31}(t)\,q_{31}(t) - iq^{+}_{31}(t)P_{13}(t)$$
$$- \gamma_{31}P_{33}(t) - \left\{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\right\}P_{23}(t) - \left\{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\right\}P_{32}(t) \qquad (7.A.26)$$

$$\dot{P}_{12}(t) = i\left(P_{11}(t) - P_{22}(t)\right)q_{21}(t) - iP_{32}(t)\,q_{31}(t)$$
$$- \left\{\tfrac{1}{2}\gamma_{21} + i(\omega_{2} - \Omega_{21})\right\}P_{12}(t) - \left\{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\right\}P_{13}(t) \qquad (7.A.27)$$

$$\dot{P}_{31}(t) = -iq^{+}_{31}(t)\left(P_{11}(t) - P_{33}(t)\right) + iq^{+}_{21}(t)P_{32}(t)$$
$$- \left\{\tfrac{1}{2}\gamma_{31} - i(\omega - \Omega_{31})\right\}P_{31}(t) - \left\{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\right\}P_{21}(t) \qquad (7.A.28)$$

$$\dot{P}_{23}(t) = iP_{21}(t)\,q_{31}(t) - iq^{+}_{21}(t)\,P_{13}(t) - \left\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) + i(\omega_3 - \Omega_{31} + \Omega_{21})\right\}P_{23}(t) - \left\{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\right\}P_{33}(t) - \left\{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\right\}P_{22}(t) \qquad \text{(7.A.29)}$$

$$\dot{P}_{21}(t) = -iq^{+}_{21}(t)\left(P_{11}(t) - P_{22}(t)\right) + iq^{+}_{31}(t)\,P_{23}(t) - \left\{\tfrac{1}{2}\gamma_{21} - i(\omega_2 - \Omega_{21})\right\}P_{21}(t) - \left\{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\right\}P_{31}(t) \qquad \text{(7.A.30)}$$

$$\dot{P}_{13}(t) = i\left(P_{11}(t) - P_{33}(t)\right)q_{31}(t) - iP_{23}(t)\,q_{21}(t) - \left\{\tfrac{1}{2}\gamma_{31} + i(\omega - \Omega_{31})\right\}P_{13}(t) - \left\{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\right\}P_{12}(t) \qquad \text{(7.A.31)}$$

$$\dot{P}_{32}(t) = iP_{31}(t)\,q_{21}(t) - iq^{+}_{31}(t)\,P_{12}(t) - \left\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) - i(\omega_3 + \Omega_{21} - \Omega_{31})\right\}P_{32}(t) - \left\{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\right\}P_{33}(t) - \left\{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\right\}P_{22}(t) \qquad \text{(7.A.32)}$$

where

$$\gamma_{31} = \tfrac{4}{3\hbar}\,\mathbf{p}_{31}^2\,\omega^3/c^3 \qquad \Omega_{31} = \gamma_{31}/\omega^3\, P\!\int_0^\infty \frac{d\omega'}{2\pi}\,\frac{\omega'^3}{\omega' - \omega}$$

$$\gamma_{21} = \tfrac{4}{3\hbar}\,\mathbf{p}_{21}^2\,\omega_2^3/c^3 \qquad \Omega_{21} = \gamma_{21}/\omega_2^3\, P\!\int_0^\infty \frac{d\omega'}{2\pi}\,\frac{\omega'^3}{\omega' - \omega_2}$$

$$\Gamma_{31} = \tfrac{4}{3\hbar}\,\mathbf{p}_{21}\cdot\mathbf{p}_{31}\,\omega^3/c^3 \qquad \Omega'_{31} = \Gamma/\omega^3\, P\!\int_0^\infty \frac{d\omega'}{2\pi}\,\frac{\omega'^3}{\omega' - \omega} \qquad \text{(7.A.33)}$$

$$\Gamma_{21} = \tfrac{4}{3\hbar}\,\mathbf{p}_{21}\cdot\mathbf{p}_{31}\,\omega_2^3/c^3 \qquad \Omega'_{21} = \Gamma_{21}/\omega_2^3\, P\!\int_0^\infty \frac{d\omega'}{2\pi}\,\frac{\omega'^3}{\omega' - \omega_2}$$

$$q_{mn} = \tfrac{1}{\hbar}\,E^{(0)}(t)\cdot\mathbf{p}_{mn}$$

$$E^{(0)} = \sum_{\ell,\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\;\hat{e}_{\ell\sigma}\,a_{\ell\sigma}(0)\,e^{-i\omega_\ell t}$$

N.B. P indicates principal part.

[The transitions 2 ⟶ 1 and 3 ⟶ 1 are σ components of a Zeeman triplet when $\Delta m = \pm 1$ and π components when $\Delta m = 0$.]

Method (i)

We shall now consider the case when a driving field of arbitrary strength, $E_D(t)$, couples levels 3 and 1. Then

$$H_{1D}(t) = -\hbar\left\{P_{31}(t)\,\lambda_{31}\,\mathcal{E}_D(t) + \lambda_{13}\,\mathcal{E}_D^{-}(t)\,P_{13}(t)\right\}$$

$$\text{where} \quad \lambda_{31} = \frac{\mathbf{p}_{31}\cdot\hat{\epsilon}_{0D}}{\hbar} = \frac{\mathbf{p}_{13}\cdot\hat{\epsilon}_{0D}}{\hbar} = \lambda_{13}$$

$$E_D(t) = \hat{\epsilon}_{0D}\left\{\mathcal{E}_D(t) + \mathcal{E}_D^{-}(t)\right\}$$

and hence

$$\dot{P}_{mn_D}(t) = -i\left(P_{3n}\delta_{1m} - P_{m1}\delta_{n3}\right)\lambda_{31}\mathcal{E}_D(t) + \lambda_{13}\mathcal{E}_D^*(t)\left(P_{1n}\delta_{3m} - P_{m3}\delta_{n1}\right)$$

$$\dot{P}_{mm_D}(t) = -i\left(P_{3m}\delta_{1m} - P_{m1}\delta_{m3}\right)\lambda_{31}\mathcal{E}_D(t) + \lambda_{13}\mathcal{E}_D^*(t)\left(P_{1m}\delta_{3m} - P_{m3}\delta_{m1}\right)$$

Thus the new equations of motion are

$$\begin{aligned}\dot{P}_{11}(t) = &-iP_{21}(t)q_{21}(t) + iq_{21}^+(t)P_{12}(t) - iP_{31}(t)\{q_{31}(t) + \lambda_{31}\mathcal{E}_D(t)\} + i\{q_{31}^+(t) + \lambda_{13}\mathcal{E}_D^*(t)\}P_{13}(t) \\ &+ \gamma_{21}P_{22}(t) + \gamma_{31}P_{33}(t) + \{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31} - \Omega'_{21})\}P_{23}(t) \\ &+ \{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) + i(\Omega'_{31} - \Omega'_{21})\}P_{32}(t)\end{aligned} \qquad (7.A.34)$$

$$\left.\begin{aligned}\dot{P}_{22}(t) = &\ iP_{21}(t)q_{21}(t) - iq_{21}^+(t)P_{12}(t) \\ &- \gamma_{21}P_{22}(t) - \{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\}P_{23}(t) - \{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\}P_{32}(t)\end{aligned}\right\} \quad (7.A.35)$$

$$\left.\begin{aligned}\dot{P}_{33}(t) = &\ iP_{31}(t)\{q_{31}(t) + \lambda_{31}\mathcal{E}_D(t)\} - i\{q_{31}^+(t) + \lambda_{13}\mathcal{E}_D^*(t)\}P_{13}(t) \\ &- \gamma_{31}P_{33}(t) - \{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\}P_{23}(t) - \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}P_{32}(t)\end{aligned}\right\} \quad (7.A.36)$$

$$\left.\begin{aligned}\dot{P}_{12}(t) = &\ i\left(P_{11}(t) - P_{22}(t)\right)q_{21}(t) - iP_{32}(t)\{q_{31}(t) + \lambda_{31}\mathcal{E}_D(t)\} \\ &- \{\tfrac{1}{2}\gamma_{21} + i(\omega_2 - \Omega_{21})\}P_{12}(t) - \{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\}P_{13}(t)\end{aligned}\right\} \quad (7.A.37)$$

$$\left.\begin{aligned}\dot{P}_{31}(t) = &-i\{q_{31}^+(t) + \lambda_{13}\mathcal{E}_D^*(t)\}\left(P_{11}(t) - P_{33}(t)\right) + iq_{21}^+(t)P_{32}(t) \\ &- \{\tfrac{1}{2}\gamma_{31} - i(\omega - \Omega_{31})\}P_{31}(t) - \{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\}P_{21}(t)\end{aligned}\right\} \quad (7.A.38)$$

$$\left.\begin{aligned}\dot{P}_{23}(t) = &\ iP_{21}(t)\{q_{31} + \lambda_{31}\mathcal{E}_D(t)\} - iq_{21}^+(t)P_{13}(t) \\ &- \{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) + i(\omega_3 - \Omega_{31} + \Omega_{21})\}P_{23}(t) - \{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\}P_{33}(t) \\ &- \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}P_{22}(t)\end{aligned}\right\} \quad (7.A.39)$$

$$\left.\begin{aligned}\dot{P}_{21}(t) = &-iq_{21}^+(t)\left(P_{11}(t) - P_{22}(t)\right) + i\left(q_{31}^+(t) + \lambda_{13}\mathcal{E}_D^*(t)\right)P_{23}(t) \\ &- \{\tfrac{1}{2}\gamma_{21} - i(\omega_2 - \Omega_{21})\}P_{21}(t) - \{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\}P_{31}(t)\end{aligned}\right\} \quad (7.A.40)$$

$$\left.\begin{aligned}\dot{P}_{13}(t) = &\ i\left(P_{11}(t) - P_{33}(t)\right)\{q_{31}(t) + \lambda_{31}\mathcal{E}_D(t)\} - iP_{23}(t)q_{21}(t) \\ &- \{\tfrac{1}{2}\gamma_{31} + i(\omega - \Omega_{31})\}P_{13}(t) - \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}P_{12}(t)\end{aligned}\right\} \quad (7.A.41)$$

$$\left.\begin{aligned}\dot{P}_{32}(t) = &\ iP_{31}(t)q_{21}(t) - i\{q_{31}^+(t) + \lambda_{13}\mathcal{E}_D^*(t)\}P_{12}(t) \\ &- \{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) - i(\omega_3 + \Omega_{21} - \Omega_{31})\}P_{32}(t) - \{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\}P_{33}(t) \\ &- \{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\}P_{22}(t)\end{aligned}\right\} \quad (7.A.42)$$

N.B. $\dot{f}_{11}(t) + \dot{f}_{22}(t) + \dot{f}_{33}(t) = 0$ (7.A.42a)

Multiplying equations (7.A.34) to (7.A.42) from left and right by [illegible] and [illegible] respectively and them multipling on the right by $\rho(0)$ and taking Tr_s we obtain the reduced density matrix equations:

$$\dot\rho^{(s)}_{11}(t) = -i\rho^{(s)}_{13}(t)\lambda_{31}\mathcal{E}_D(t) + i\lambda_{13}\mathcal{E}_D^*(t)\rho^{(s)}_{31}(t) + \gamma_{21}\rho^{(s)}_{22}(t) + \gamma_{31}\rho^{(s)}_{33}(t) + \{\tfrac12(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31}-\Omega'_{21})\}\rho^{(s)}_{32}(t) + \{\tfrac12(\Gamma_{31}+\Gamma_{21}) + i(\Omega'_{31}-\Omega'_{21})\}\rho^{(s)}_{23}(t) \qquad (7.A.43)$$

$$\dot\rho^{(s)}_{22}(t) = -\gamma_{21}\rho^{(s)}_{22}(t) - \{\tfrac12\Gamma_{31} - i\Omega'_{31}\}\rho^{(s)}_{32}(t) - \{\tfrac12\Gamma_{31} + i\Omega'_{31}\}\rho^{(s)}_{23}(t) \qquad (7.A.44)$$

$$\dot\rho^{(s)}_{33}(t) = i\rho^{(s)}_{13}(t)\lambda_{31}\mathcal{E}_D(t) - i\lambda_{13}\mathcal{E}_D^*(t)\rho^{(s)}_{31}(t) - \gamma_{31}\rho^{(s)}_{33}(t) - \{\tfrac12\Gamma_{21} + i\Omega'_{21}\}\rho^{(s)}_{32}(t) - \{\tfrac12\Gamma_{21} - i\Omega'_{21}\}\rho^{(s)}_{23}(t) \qquad (7.A.45)$$

$$\dot\rho^{(s)}_{21}(t) = -i\rho^{(s)}_{23}(t)\lambda_{31}\mathcal{E}_D(t) - \{\tfrac12\gamma_{21} + i(\omega_2 - \Omega_{21})\}\rho^{(s)}_{21}(t) - \{\tfrac12\Gamma_{31} - i\Omega'_{31}\}\rho^{(s)}_{31}(t) \qquad (7.A.46)$$

$$\dot\rho^{(s)}_{13}(t) = -i\lambda_{13}\mathcal{E}_D^*(t)(\rho^{(s)}_{11}(t) - \rho^{(s)}_{33}(t)) - \{\tfrac12\gamma_{31} - i(\omega - \Omega_{31})\}\rho^{(s)}_{13}(t) - \{\tfrac12\Gamma_{21} + i\Omega'_{21}\}\rho^{(s)}_{12}(t) \qquad (7.A.47)$$

$$\dot\rho^{(s)}_{32}(t) = i\rho^{(s)}_{12}(t)\lambda_{31}\mathcal{E}_D(t) - \{\tfrac12(\gamma_{21}+\gamma_{31}) + i(\omega_3 - \Omega_{31} + \Omega_{21})\}\rho^{(s)}_{32}(t) - \{\tfrac12\Gamma_{31} + i\Omega'_{31}\}\rho^{(s)}_{33}(t) - \{\tfrac12\Gamma_{21} - i\Omega'_{21}\}\rho^{(s)}_{22}(t) \qquad (7.A.48)$$

$$\dot\rho^{(s)}_{12}(t) = i\lambda_{13}\mathcal{E}_D^*(t)\rho^{(s)}_{32}(t) - \{\tfrac12\gamma_{21} - i(\omega_2 - \Omega_{21})\}\rho^{(s)}_{12}(t) - \{\tfrac12\Gamma_{31} + i\Omega'_{31}\}\rho^{(s)}_{13}(t) \qquad (7.A.49)$$

$$\dot\rho^{(s)}_{31}(t) = i(\rho^{(s)}_{11}(t) - \rho^{(s)}_{33}(t))\lambda_{31}\mathcal{E}_D(t) - \{\tfrac12\gamma_{31} + i(\omega - \Omega_{31})\}\rho^{(s)}_{31}(t) - \{\tfrac12\Gamma_{21} - i\Omega'_{21}\}\rho^{(s)}_{21}(t) \qquad (7.A.50)$$

$$\dot\rho^{(s)}_{23}(t) = -i\lambda_{13}\mathcal{E}_D^*(t)\rho^{(s)}_{21}(t) - \{\tfrac12(\gamma_{21}+\gamma_{31}) - i(\omega_3 + \Omega_{21} - \Omega_{31})\}\rho^{(s)}_{23}(t) - \{\tfrac12\Gamma_{31} - i\Omega'_{31}\}\rho^{(s)}_{33}(t) - \{\tfrac12\Gamma_{21} + i\Omega'_{21}\}\rho^{(s)}_{22}(t) \qquad (7.A.51)$$

Method (ii)

The initial photon state is $|\alpha_3\rangle$

$$\left.\begin{aligned} \therefore q_{31}|i_{ph}\rangle &= \sqrt{\frac{2\pi\omega_D}{\hbar V}}\,(\hat{e}_{oD}\cdot p_{31})\,e^{-i\omega_D t}\,\alpha_3|\alpha_3\rangle \\ \text{and } q_{mn}|i_{ph}\rangle &= 0 \quad \text{for all other m and n where } m > n \end{aligned}\right\} \quad (7.A.52)$$

$$\left.\begin{aligned} \langle i_{ph}|q_{31}|i_{ph}\rangle &= g_{\omega_D,31}\,e^{-i\omega_D t}\,\alpha_3 \\ &= \tfrac{1}{2}G_D'\,e^{-i\omega_D t} \end{aligned}\right\} \quad (7.A.53)$$

$$\left.\begin{aligned} \langle i_{ph}|q_{31}^{+}|i_{ph}\rangle &= g_{\omega_D,31}\,e^{i\omega_D t}\,\alpha_3^{*} \\ &= \tfrac{1}{2}G_D'^{*}\,e^{i\omega_D t} \end{aligned}\right\} \quad (7.A.54)$$

$$\left.\begin{aligned} \tfrac{1}{4}G_D^2 &= g^2_{\omega_D,31}\,|\alpha_3|^2 \\ &= \left(\frac{2\pi\omega_D}{\hbar}\right)(\hat{e}_{oD}\cdot p_{31})^2\,|\alpha_3|^2 \\ \text{(as opposed to } \tfrac{1}{4}G_3^2 &= (\lambda_{31}\mathcal{E}_{oD})^2 \\ &= \left(\frac{p_{31}\cdot\hat{e}_{oD}\,\mathcal{E}_{oL}}{\hbar}\right)^2 \quad \text{in method (i))} \end{aligned}\right\} \quad (7.A.55)$$

Thus on multiplying the transition operator equations (7.A.34)-(42) by $\langle i_{ph}|$ and $|i_{ph}\rangle$ on left and right respectively and then multipling on the right by $\rho^{(s)}(0)$ and taking the Tr_s we obtain reduced density matrix equations, similar to those of method (i), i.e. equations (7.A.43) - (51), when in the former equations we assume oscillates harmonically at ω_D such that $\mathcal{E}_D(t) = \mathcal{E}_{oD}e^{-i\omega_D t}$ where $\omega_D \triangleq \omega$. (N.B. the assumption of oscillation at one frequency in method (i) is equivalent to the assumption in method (ii) that only one mode of the e.m. field interacts with the atom.) For complete correspondence we require

$$\left.\begin{aligned} \tfrac{1}{2}G_D' &\longrightarrow \lambda_{31}\mathcal{E}_{oD} \quad \text{i.e. } \alpha_3 \longrightarrow \sqrt{\frac{V}{2\pi\hbar\omega_D}}\,\mathcal{E}_{oD} \\ \tfrac{1}{2}G_D'^{*} &\longrightarrow \lambda_{13}\mathcal{E}_{oD} \quad \text{i.e. } \alpha_3^{*} \longrightarrow \sqrt{\frac{V}{2\pi\hbar\omega_D}}\,\mathcal{E}_{oD} \end{aligned}\right\} \quad (7.A.56)$$

i.e. α_3 becomes real for real dipole matrix elements.

B. Power Spectrum

In equations (7.A.43) - (51) we shall now let

$$\left.\begin{aligned} \omega_2 - \Omega_{21} &= \omega_2' \\ \omega - \Omega_{31} &= \omega' \\ \omega_3 - \Omega_{31} + \Omega_{21} &= \omega_3' \\ \tfrac{1}{2}\Gamma_{31} + i\Omega_{31}' &= \tfrac{1}{2}\Gamma_{31}' \\ \tfrac{1}{2}\Gamma_{31} - i\Omega_{31}' &= \tfrac{1}{2}\Gamma_{31}'^{*} \\ \tfrac{1}{2}\Gamma_{21} + i\Omega_{21}' &= \tfrac{1}{2}\Gamma_{21}' \\ \tfrac{1}{2}\Gamma_{21} - i\Omega_{21}' &= \tfrac{1}{2}\Gamma_{21}'^{*} \end{aligned}\right\} \qquad (7.B.1)$$

and, in order to simplify the equations still further, we shall ignore frequency shifts Ω_{21} , Ω_{31} and terms Ω_{21}' , Ω_{31}' so that instead of ω_2' , ω' , ω_3' , $\tfrac{1}{2}\Gamma_{31}'$, $\tfrac{1}{2}\Gamma_{31}'^{*}$, $\tfrac{1}{2}\Gamma_{21}'$, $\tfrac{1}{2}\Gamma_{21}'^{*}$ we shall write ω_2 , ω , ω_3 , $\tfrac{1}{2}\Gamma_{31}$, $\tfrac{1}{2}\Gamma_{21}$. We do this since we are not interested in these small frequency shifts but rather the effects of the driving field and separation of levels 3 and 2 on the resulting spectral profile for transitions between levels 3 and 1.

On making the above simplifications we can write equations (7.A.43)-(51) as follows, where we neglect the superscripts on ρ 's, indicating their reduced nature, for convenience:-

$$\left.\begin{aligned} \dot{\rho}_{11}(t) = &- i\lambda_{31}\mathcal{E}_{0D}\left(e^{-i\omega_D t}\rho_{13}(t)\right) + i\lambda_{13}\mathcal{E}_{0D}\left(e^{i\omega_D t}\rho_{31}(t)\right) \\ &+ \gamma_{21}\rho_{22}(t) + \gamma_{31}\rho_{33}(t) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})\rho_{32}(t) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})\rho_{23}(t) \end{aligned}\right\} \qquad (7.B.2)$$

$$\dot{\rho}_{22}(t) = -\gamma_{21}\rho_{22}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{32}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{23}(t) \qquad (7.B.3)$$

$$\left.\begin{aligned} \dot{\rho}_{33}(t) = &\; i\lambda_{31}\mathcal{E}_{0D}\left(e^{-i\omega_D t}\rho_{13}(t)\right) - i\lambda_{13}\mathcal{E}_{0D}\left(e^{i\omega_D t}\rho_{31}(t)\right) \\ &- \gamma_{31}\rho_{33}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{32}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{23}(t) \end{aligned}\right\} \qquad (7.B.4)$$

$$\left.\begin{aligned} \dot{\rho}_{21}(t) = &- i\lambda_{31}\mathcal{E}_{0D}\left(e^{-i\omega_D t}\rho_{23}(t)\right) \\ &- \left(\tfrac{1}{2}\gamma_{21} + i\omega_2\right)\rho_{21}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{31}(t) \end{aligned}\right\} \qquad (7.B.5)$$

$$\dot{\rho}_{13}(t) = -i\lambda_{13}\mathcal{E}_{0D}(e^{i\omega_D t}\rho_{11}(t)) + i\lambda_{13}\mathcal{E}_{0D}(e^{i\omega_D t}\rho_{33}(t)) - (\tfrac{1}{2}\gamma_{31} - i\omega)\rho_{13}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{12}(t) \tag{7.B.6}$$

$$\dot{\rho}_{32}(t) = i\lambda_{31}\mathcal{E}_{0D}(e^{-i\omega_D t}\rho_{12}(t)) - \{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) + i\omega_3\}\rho_{32}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{33}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{22}(t) \tag{7.B.7}$$

$$\dot{\rho}_{12}(t) = i\lambda_{13}\mathcal{E}_{0D}(e^{i\omega_D t}\rho_{32}(t)) - (\tfrac{1}{2}\gamma_{21} - i\omega_2)\rho_{12}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{13}(t) \tag{7.B.8}$$

$$\dot{\rho}_{31}(t) = i\lambda_{31}\mathcal{E}_{0D}e^{-i\omega_D t}\rho_{11}(t) - i\lambda_{31}\mathcal{E}_{0D}e^{-i\omega_D t}\rho_{33}(t) - (\tfrac{1}{2}\gamma_{31} + i\omega)\rho_{31}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{21}(t) \tag{7.B.9}$$

$$\dot{\rho}_{23}(t) = -i\lambda_{13}\mathcal{E}_{0D}e^{i\omega_D t}\rho_{21}(t) - \{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) - i\omega_3\}\rho_{23}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{33}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{22}(t) \tag{7.B.10}$$

__a) Total spectral correlation function for transitions between the uppermost level, 3, and ground level, 1__

In order to calculate the spectral correlation function for transitions between levels 3 and 1 we need to solve equations (7.B.2)-(10) for $\rho_{31}(t)$. Thus, in order to obtain nine distinct functions of time, and to avoid having to deduce $\hat{\rho}_{31}(s)$ from $\hat{\rho}_{31}(s-i\omega_D)$ eg. we multiply equations (7.B.2), (3), (4), (7), (10) by $e^{-i\omega_D t}$ and (7.B.6), (8) by $e^{-i2\omega_D t}$. Hence we obtain the following equations where $\lambda_{31} = \lambda_{13} = \lambda$ and $\mathcal{E}_{0D} = \mathcal{E}_0$:-

$$e^{-i\omega_D t}\dot{\rho}_{11}(t) = -i\lambda\mathcal{E}_0(e^{-i2\omega_D t}\rho_{13}(t)) + i\lambda\mathcal{E}_0\rho_{31}(t) + \gamma_{21}(e^{-i\omega_D t}\rho_{22}(t)) + \gamma_{31}(e^{-i\omega_D t}\rho_{33}(t)) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(e^{-i\omega_D t}\rho_{32}(t)) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(e^{-i\omega_D t}\rho_{23}(t)) \tag{7.B.11}$$

$$e^{-i\omega_D t}\dot{\rho}_{22}(t) = -\gamma_{21}(e^{-i\omega_D t}\rho_{22}(t)) - \tfrac{1}{2}\Gamma_{31}(e^{-i\omega_D t}\rho_{32}(t)) - \tfrac{1}{2}\Gamma_{31}(e^{-i\omega_D t}\rho_{23}(t)) \tag{7.B.12}$$

$$e^{-i\omega_D t}\dot{\rho}_{33}(t) = i\lambda\mathcal{E}_0(e^{-i2\omega_D t}\rho_{13}(t)) - i\lambda\mathcal{E}_0\rho_{31}(t) - \gamma_{31}(e^{-i\omega_D t}\rho_{33}(t)) - \tfrac{1}{2}\Gamma_{21}(e^{-i\omega_D t}\rho_{32}(t)) - \tfrac{1}{2}\Gamma_{21}(e^{-i\omega_D t}\rho_{23}(t)) \tag{7.B.13}$$

$$\left.\begin{aligned}\dot{\rho}_{21}(t) &= -i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{23}(t)\right)\\ &\quad -\left(\tfrac{1}{2}\gamma_{21}+i\omega_2\right)\rho_{21}(t) - \tfrac{1}{2}\Gamma_{31}\rho_{21}(t)\end{aligned}\right\} \quad (7.B.14)$$

$$\left.\begin{aligned}e^{-i2\omega_3 t}\dot{\rho}_{13}(t) &= -i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{11}(t)\right) + i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{33}(t)\right)\\ &\quad -\left(\tfrac{1}{2}\gamma_{31}-i\omega\right)\left(e^{-i2\omega_3 t}\rho_{13}(t)\right) - \tfrac{1}{2}\Gamma_{21}\left(e^{-i2\omega_3 t}\rho_{12}(t)\right)\end{aligned}\right\} \quad (7.B.15)$$

$$\left.\begin{aligned}e^{-i\omega_3 t}\dot{\rho}_{32}(t) &= i\lambda\mathcal{E}_0\left(e^{-i2\omega_3 t}\rho_{12}(t)\right)\\ &\quad -\left\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31})+i\omega_3\right\}\left(e^{-i\omega_3 t}\rho_{32}(t)\right) - \tfrac{1}{2}\Gamma_{31}\left(e^{-i\omega_3 t}\rho_{33}(t)\right)\\ &\quad - \tfrac{1}{2}\Gamma_{21}\left(e^{-i\omega_3 t}\rho_{22}(t)\right)\end{aligned}\right\} \quad (7.B.16)$$

$$\left.\begin{aligned}e^{-i2\omega_3 t}\dot{\rho}_{12}(t) &= i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{32}(t)\right)\\ &\quad -\left(\tfrac{1}{2}\gamma_{21}-i\omega_2\right)\left(e^{-i2\omega_3 t}\rho_{12}(t)\right) - \tfrac{1}{2}\Gamma_{31}\left(e^{-i2\omega_3 t}\rho_{13}(t)\right)\end{aligned}\right\} \quad (7.B.17)$$

$$\left.\begin{aligned}\dot{\rho}_{31}(t) &= i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{11}(t)\right) - i\lambda\mathcal{E}_0\left(e^{-i\omega_3 t}\rho_{33}(t)\right)\\ &\quad -\left(\tfrac{1}{2}\gamma_{31}+i\omega\right)\rho_{31}(t) - \tfrac{1}{2}\Gamma_{21}\rho_{21}(t)\end{aligned}\right\} \quad (7.B.18)$$

$$\left.\begin{aligned}e^{-i\omega_3 t}\dot{\rho}_{23}(t) &= -i\lambda\mathcal{E}_0\rho_{21}(t)\\ &\quad -\left\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31})-i\omega_3\right\}\left(e^{-i\omega_3 t}\rho_{23}(t)\right) - \tfrac{1}{2}\Gamma_{31}\left(e^{-i\omega_3 t}\rho_{33}(t)\right)\\ &\quad - \tfrac{1}{2}\Gamma_{21}\left(e^{-i\omega_3 t}\rho_{22}(t)\right)\end{aligned}\right\} \quad (7.B.19)$$

Let

$$\left.\begin{aligned}
a(t) &= \rho_{11}(t)e^{-i\omega_3 t} & \therefore\ \dot{\rho}_{11}e^{-i\omega_3 t} &= \dot{a}(t)+i\omega_3 a(t)\\
b(t) &= \rho_{22}(t)e^{-i\omega_3 t} & \dot{\rho}_{22}e^{-i\omega_3 t} &= \dot{b}(t)+i\omega_3 b(t)\\
c(t) &= \rho_{33}(t)e^{-i\omega_3 t} & \dot{\rho}_{33}e^{-i\omega_3 t} &= \dot{c}(t)+i\omega_3 c(t)\\
d(t) &= \rho_{13}(t)e^{-i2\omega_3 t} & \dot{\rho}_{13}e^{-i2\omega_3 t} &= \dot{d}(t)+i2\omega_3 d(t)\\
e(t) &= \rho_{32}(t)e^{-i\omega_3 t} & \dot{\rho}_{32}e^{-i\omega_3 t} &= \dot{e}(t)+i\omega_3 e(t)\\
f(t) &= \rho_{12}(t)e^{-i2\omega_3 t} & \dot{\rho}_{12}e^{-i2\omega_3 t} &= \dot{f}(t)+i2\omega_3 f(t)\\
g(t) &= \rho_{23}(t)e^{-i\omega_3 t} & \dot{\rho}_{23}e^{-i\omega_3 t} &= \dot{g}(t)+i\omega_3 g(t)
\end{aligned}\right\} \quad (7.B.20)$$

Then, using equation (7.B.20), we can rewrite equations (7.B.11)-(19) as follows:-

$$\left.\begin{aligned}\dot{a}+i\omega_3 a &= -i\lambda\mathcal{E}_0 d + i\lambda\mathcal{E}_0\rho_{31}\\ &\quad +\gamma_{21}b + \gamma_{31}c + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})e + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})g\end{aligned}\right\} \quad (7.B.21)$$

$$\dot{b} + i\omega_3 b = -\gamma_{21} b - \tfrac12\Gamma_{31} e - \tfrac12\Gamma_{31} g \tag{7.B.22}$$

$$\begin{aligned}\dot{c} + i\omega_3 c &= i\lambda\mathcal{E}_0 d - i\lambda\mathcal{E}_0 \rho_{31} \\ &\quad - \gamma_{31} c - \tfrac12\Gamma_{21} e - \tfrac12\Gamma_{21} g\end{aligned} \tag{7.B.23}$$

$$\begin{aligned}\dot\rho_{21} &= - i\lambda\mathcal{E}_0 g \\ &\quad - (\tfrac12\gamma_{21} + i\omega_2)\rho_{21} - \tfrac12\Gamma_{31}\rho_{31}\end{aligned} \tag{7.B.24}$$

$$\begin{aligned}\dot d + i2\omega_3 d &= - i\lambda\mathcal{E}_0 a + i\lambda\mathcal{E}_0 c \\ &\quad - (\tfrac12\gamma_{31} - i\omega) d - \tfrac12\Gamma_{21} f\end{aligned} \tag{7.B.25}$$

$$\begin{aligned}\dot e + i\omega_3 e &= i\lambda\mathcal{E}_0 f \\ &\quad - \{\tfrac12(\gamma_{21}+\gamma_{31}) + i\omega_3\} e - \tfrac12\Gamma_{31} c - \tfrac12\Gamma_{21} b\end{aligned} \tag{7.B.26}$$

$$\begin{aligned}\dot f + i2\omega_3 f &= i\lambda\mathcal{E}_0 e \\ &\quad - (\tfrac12\gamma_{21} - i\omega_2) f - \tfrac12\Gamma_{31} d\end{aligned} \tag{7.B.27}$$

$$\begin{aligned}\dot\rho_{31} &= i\lambda\mathcal{E}_0 a - i\lambda\mathcal{E}_0 c \\ &\quad - (\tfrac12\gamma_{31} + i\omega)\rho_{31} - \tfrac12\Gamma_{21}\rho_{21}\end{aligned} \tag{7.B.28}$$

$$\begin{aligned}\dot g + i\omega_3 g &= - i\lambda\mathcal{E}_0 \rho_{21} \\ &\quad - \{\tfrac12(\gamma_{21}+\gamma_{31}) - i\omega_3\} g - \tfrac12\Gamma_{31} c - \tfrac12\Gamma_{21} b\end{aligned} \tag{7.B.29}$$

Taking Laplace transforms, for initial time t' = 0, we obtain:-

$$\begin{aligned}(s + i\omega_3)\hat a(s) &= - i\lambda\mathcal{E}_0 \hat d(s) + i\lambda\mathcal{E}_0\hat\rho_{31}(s) + \gamma_{21}\hat b(s) + \gamma_{31}\hat c(s) + \tfrac12(\Gamma_{31}+\Gamma_{21})\hat e(s) \\ &\quad + \tfrac12(\Gamma_{31}+\Gamma_{21})\hat g(s) + e^{-st'}a(t')\end{aligned} \tag{7.B.30}$$

$$(s+\gamma_{21}+i\omega_3)\hat b(s) = -\tfrac12\Gamma_{31}\hat e(s) - \tfrac12\Gamma_{31}\hat g(s) + e^{-st'}b(t') \tag{7.B.31}$$

$$(s+\gamma_{31}+i\omega_3)\hat c(s) = i\lambda\mathcal{E}_0\hat d(s) - i\lambda\mathcal{E}_0\hat\rho_{31}(s) - \tfrac12\Gamma_{21}\hat e(s) - \tfrac12\Gamma_{21}\hat g(s) + e^{-st'}c(t') \tag{7.B.32}$$

$$(s+\tfrac12\gamma_{21}+i\omega_2)\hat\rho_{21}(s) = - i\lambda\mathcal{E}_0\hat g(s) - \tfrac12\Gamma_{31}\hat\rho_{31}(s) + e^{-st'}\rho_{21}(t') \tag{7.B.33}$$

$$(s+\tfrac12\gamma_{31} - i(\omega-2\omega_3))\hat d(s) = - i\lambda\mathcal{E}_0\hat a(s) + i\lambda\mathcal{E}_0\hat c(s) - \tfrac12\Gamma_{21}\hat f(s) + e^{-st'}d(t') \tag{7.B.34}$$

$$(s+\tfrac12(\gamma_{21}+\gamma_{31}) + i(\omega_3+\omega_3))\hat e(s) = i\lambda\mathcal{E}_0\hat f(s) - \tfrac12\Gamma_{31}\hat c(s) - \tfrac12\Gamma_{21}\hat b(s) + e^{-st'}e(t') \tag{7.B.35}$$

$$\left(s+\tfrac{1}{2}\gamma_{21}+i(\omega_2-\Delta\omega)\right)\hat{f}(s) = i\lambda\mathcal{E}_0\hat{c}(s) - \tfrac{1}{2}\Gamma_{31}\hat{d}(s) + e^{-st'}f(t') \tag{7.B.36}$$

$$\left(s+\tfrac{1}{2}\gamma_{31}+i\omega\right)\hat{\rho}_{31}(s) = i\lambda\mathcal{E}_0\hat{a}(s) - i\lambda\mathcal{E}_0\hat{c}(s) - \tfrac{1}{2}\Gamma_{21}\hat{\rho}_{21}(s) + e^{-st'}\rho_{31}(t') \tag{7.B.37}$$

$$\left(s+\tfrac{1}{2}(\gamma_{21}+\gamma_{31})+i(\omega_2-\Delta\omega)\right)\hat{g}(s) = -i\lambda\mathcal{E}_0\hat{\rho}_{21}(s) - \tfrac{1}{2}\Gamma_{31}\hat{c}(s) - \tfrac{1}{2}\Gamma_{21}\hat{b}(s) + e^{-st'}g(t') \tag{7.B.38}$$

First we shall solve equations (7.B.33), (37), (38) for $\hat{\rho}_{21}(s)$, $\hat{\rho}_{31}(s)$ and $\hat{g}(s)$ in terms of $\hat{a}(s)$, $\hat{b}(s)$, $\hat{c}(s)$. In this way we obtain:

$$\left.\begin{aligned}
\hat{g}(s) = &- \tfrac{1}{2}\Gamma_{31}\,\tfrac{1}{4}G^2\,\frac{1}{H_1}\,\hat{a}(s) \\
&- \tfrac{1}{2}\Gamma_{21}\,\frac{F_1}{H_1}\,\hat{b}(s) \\
&- \tfrac{1}{2}\Gamma_{31}\,\frac{(F_1-\tfrac{1}{4}G^2)}{H_1}\,\hat{c}(s) \\
&+ \tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\frac{1}{H_1}\,e^{-st'}\rho_{31}(t') \\
&- (i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i\omega)}{H_1}\,e^{-st'}\rho_{21}(t') \\
&+ \frac{F_1}{H_1}\,e^{-st'}g(t')
\end{aligned}\right\} \tag{7.B.39}$$

$$\left.\begin{aligned}
\hat{\rho}_{21}(s) = &- \tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\frac{(s+\gamma+i(\omega_2-\Delta\omega))}{H_1}\,\hat{a}(s) \\
&+ \tfrac{1}{2}\Gamma_{21}(i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i\omega)}{H_1}\,\hat{b}(s) \\
&+ \tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\left\{\frac{(s+\tfrac{1}{2}\gamma_{31}+i\omega)+(s+\gamma+i(\omega_2-\Delta\omega))}{H_1}\right\}\hat{c}(s) \\
&- \tfrac{1}{2}\Gamma_{31}\frac{(s+\gamma+i(\omega_2-\Delta\omega))}{H_1}\,e^{-st'}\rho_{31}(t') \\
&+ \frac{(s+\tfrac{1}{2}\gamma_{31}+i\omega)(s+\gamma+i(\omega_2-\Delta\omega))}{H_1}\,e^{-st'}\rho_{21}(t') \\
&- (i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i\omega)}{H_1}\,e^{-st'}g(t')
\end{aligned}\right\} \tag{7.B.40}$$

$$\left.\begin{aligned}
\hat{\rho}_{31}(s) = &\,(i\lambda\mathcal{E}_0)\frac{B_1}{H_1}\,\hat{a}(s) \\
&- \tfrac{1}{4}\Gamma_{21}^2(i\lambda\mathcal{E}_0)\frac{1}{H_1}\,\hat{b}(s) \\
&- (i\lambda\mathcal{E}_0)\left(\frac{B_1+\tfrac{1}{4}\Gamma^2}{H_1}\right)\hat{c}(s) \\
&+ \frac{B_1}{H_1}\,e^{-st'}\rho_{31}(t') \\
&- \tfrac{1}{2}\Gamma_{21}\frac{(s+\gamma+i(\omega_2-\Delta\omega))}{H_1}\,e^{-st'}\rho_{21}(t') \\
&+ \tfrac{1}{2}\Gamma_{21}(i\lambda\mathcal{E}_0)\frac{1}{H_1}\,e^{-st'}g(t')
\end{aligned}\right\} \tag{7.B.41}$$

where

$$H_2(s) = (s+\gamma+i(\omega_3-2\Delta\omega))F_2 + \tfrac{1}{4}G^2(s+\tfrac{1}{2}\gamma_{31}+i\omega)$$

$$F_2(s) = (s+\tfrac{1}{2}\gamma_{21}+i\omega_2)(s+\tfrac{1}{2}\gamma_{31}+i\omega) - \tfrac{1}{4}\Gamma^2 \qquad \textbf{(7.B.42)}$$

$$B_2(s) = (s+\gamma+i(\omega_2-\Delta\omega))(s+\tfrac{1}{2}\gamma_{21}+i\omega_2) + \tfrac{1}{4}G^2$$

$$\gamma = \tfrac{1}{2}(\gamma_{21}+\gamma_{31})$$

$$\Gamma = \Gamma_{21}\Gamma_{31} \qquad \textbf{(7.B.43)}$$

$$\Delta\omega = \omega - \omega_0$$

$$(\lambda\mathcal{E}_0)^2 = \tfrac{1}{4}G^2$$

Next we solve equations (7.B.34), (35), (36) for $\hat{d}(s)$, $\hat{e}(s)$, $\hat{f}(s)$ in terms of $\hat{a}(s)$, $\hat{b}(s)$, $\hat{c}(s)$:-

$$\begin{aligned}\hat{e}(s) = &-\tfrac{1}{4}G^2\,\tfrac{1}{2}\Gamma_{31}\frac{1}{H_2}\hat{a}(s)\\ &-\tfrac{1}{2}\Gamma_{21}\frac{F_2}{H_2}\hat{b}(s)\\ &-\tfrac{1}{2}\Gamma_{31}\frac{(F_2-\tfrac{1}{4}G^2)}{H_2}\hat{c}(s)\\ &-\tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\frac{1}{H_2}e^{-st'}d(t')\\ &+(i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))}{H_2}e^{-st'}f(t')\\ &+\frac{F_2}{H_2}e^{-st'}e(t')\end{aligned}$$

$$\begin{aligned}\hat{d}(s) = &-(i\lambda\mathcal{E}_0)\frac{B_2}{H_2}\hat{a}(s)\\ &+(i\lambda\mathcal{E}_0)\tfrac{1}{4}\Gamma_{21}^2\frac{1}{H_2}\hat{b}(s)\\ &+(i\lambda\mathcal{E}_0)\frac{(B_2+\tfrac{1}{4}\Gamma^2)}{H_2}\hat{c}(s)\\ &+\frac{B_2}{H_2}e^{-st'}d(t')\\ &-\tfrac{1}{2}\Gamma_{21}\frac{(s+\gamma+i(\omega+\omega_3-\Delta\omega))}{H_2}e^{-st'}f(t')\\ &-\tfrac{1}{2}\Gamma_{21}(i\lambda\mathcal{E}_0)\frac{1}{H_2}e^{-st'}e(t')\end{aligned}$$

(7.B.44) **(7.B.45)**

$$\begin{aligned}\hat{f}(s) = &\tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\frac{(s+\gamma+i(\omega+\omega_3-\Delta\omega))}{H_2}\hat{a}(s)\\ &-\tfrac{1}{2}\Gamma_{21}(i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))}{H_2}\hat{b}(s)\\ &-\tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)\frac{\{(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))+(s+\gamma+i(\omega+\omega_3-\Delta\omega))\}}{H_2}\hat{c}(s)\\ &-\tfrac{1}{2}\Gamma_{31}\frac{(s+\gamma+i(\omega+\omega_3-\Delta\omega))}{H_2}e^{-st'}d(t')\\ &+\frac{(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))(s+\gamma+i(\omega+\omega_3-\Delta\omega))}{H_2}e^{-st'}f(t')\\ &+(i\lambda\mathcal{E}_0)\frac{(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))}{H_2}e^{-st'}e(t')\end{aligned} \qquad \textbf{(7.B.46)}$$

where $H_1(s) = (s+\beta+i(\omega+\omega_3-\Delta\omega))F_2 + \tfrac{1}{4}G^2(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))$

$$F_2(s) = (s+\tfrac{1}{2}\gamma_{21}+i(\omega+\omega_2+2\Delta\omega))(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega)) - \tfrac{1}{4}\Gamma^2 \qquad (7.B.47)$$

$$F_1(s) = (s+\beta+i(\omega+\omega_3-\Delta\omega))(s+\tfrac{1}{2}\gamma_{21}+i(\omega+\omega_2+2\Delta\omega)) + \tfrac{1}{4}G^2$$

Now we can solve equations (7.B.30), (7.B.31), (7.B.32) for $\hat{a}(s)$, $\hat{b}(s)$, $\hat{c}(s)$ by substituting equations (7.B.39) - (46). These three equations can then be reduced to two by eliminating $\hat{b}(s)$ as follows:-

Since, from equation (7.A.42a)

$$\dot{P}_{11} + \dot{P}_{22} + \dot{P}_{33} = 0$$

we therefore know that

$$\dot{\rho}_{11}^{(s)} + \dot{\rho}_{22}^{(s)} + \dot{\rho}_{33}^{(s)} = 0$$

and on taking Laplace transforms

$$s\{\hat{\rho}_{11}^{(s)}(s) + \hat{\rho}_{22}^{(s)}(s) + \hat{\rho}_{33}^{(s)}(s)\} = e^{-st'}\{\rho_{11}^{(s)}(t') + \rho_{22}^{(s)}(t') + \rho_{33}^{(s)}(t')\}$$

i.e. $(s+i(\omega-\Delta\omega))\{\hat{a}(s)+\hat{b}(s)+\hat{c}(s)\} = e^{-st'}\{a(t')+b(t')+c(t')\}$

$$\therefore \hat{b}(s) = -\hat{a}(s) - \hat{c}(s) + \frac{e^{-st'}}{s+i(\omega-\Delta\omega)} \times (a(t') + b(t') + c(t')) \qquad (7.B.48)$$

The resulting two equations may be written as follows:-

$$a_1\hat{a}(s) + a_3\hat{c}(s) = a_0 \qquad (7.B.49)$$

$$b_1\hat{a}(s) + b_3\hat{c}(s) = b_0 \qquad (7.B.50)$$

where

$$\begin{aligned}
a_1(s) &= -(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac{1}{4}\Gamma_{31}^2\,\tfrac{1}{4}G^2(H_1+H_2) + \tfrac{1}{4}\Gamma^2(F_2H_1+F_1H_2) \\
a_3(s) &= -(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 + \tfrac{1}{4}\Gamma_{31}^2\,\tfrac{1}{4}G^2(H_1+H_2) + \tfrac{1}{4}(\Gamma^2-\Gamma_{31}^2)(F_2H_1+F_1H_2) \\
a_0(s) &= -e^{-st'}a(t')\frac{1}{(s+i(\omega-\Delta\omega))}\{(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac{1}{4}\Gamma^2(F_2H_1+F_1H_2)\} \\
&\quad - e^{-st'}b(t')\frac{1}{(s+i(\omega-\Delta\omega))}\}\qquad \gamma_{21}H_1H_2 - \tfrac{1}{4}\Gamma^2(F_2H_1+F_1H_2)\}
\end{aligned} \qquad (7.B.51)$$

$$\begin{aligned}
&- e^{-st'}\, c(t')\frac{1}{(s+i(\omega-\Delta\omega))}\left\{(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac{1}{4}\Gamma^2(F_2H_1+F_1H_2)\right\}\\
&- e^{-st'}\, d(t')\left\{-\tfrac{1}{4}\Gamma_{31}^2(i\lambda\mathcal{E}_0)H_1\right\}\\
&- e^{-st'}\, f(t')\left\{\tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))H_1\right\}\\
&- e^{-st'}\, e(t')\left\{\tfrac{1}{2}\Gamma_{31}F_2H_1\right\}\\
&- e^{-st'}\, \rho_{31}(t')\left\{\tfrac{1}{4}\Gamma_{31}^2(i\lambda\mathcal{E}_0)H_2\right\}\\
&- e^{-st'}\, \rho_{21}(t')\left\{-\tfrac{1}{2}\Gamma_{31}(i\lambda\mathcal{E}_0)(s+\tfrac{1}{2}\gamma_{31}+i\omega)H_2\right\}\\
&- e^{-st'}\, g(t')\left\{\tfrac{1}{2}\Gamma_{31}F_1H_2\right\}
\end{aligned} \tag{7.B.51}$$

$$\begin{aligned}
b_1(s) = {}& (s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 + \tfrac{1}{4}G^2\,\tfrac{1}{4}(\Gamma_{31}^2+\Gamma_{21}^2+\Gamma^2)(H_1+H_2) + \tfrac{1}{4}G^2(B_2H_1+B_1H_2)\\
&- \tfrac{1}{4}(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\\
b_3(s) = {}& (\gamma_{21}-\gamma_{31})H_1H_2 + \tfrac{1}{4}G^2\,\tfrac{1}{4}(\Gamma_{21}^2-\Gamma_{31}^2-2\Gamma^2)(H_1+H_2) - \tfrac{1}{4}G^2(B_2H_1+B_1H_2)\\
&- \tfrac{1}{4}(\Gamma_{21}^2-\Gamma_{31}^2)(F_2H_1+F_1H_2)\\
b_0(s) = {}& e^{-st'}a(t')\frac{1}{(s+i(\omega-\Delta\omega))}\Big\{(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 + \tfrac{1}{4}G^2\,\tfrac{1}{4}\Gamma_{21}^2(H_1+H_2) - \tfrac{1}{4}(\Gamma^2+\Gamma_{21}^2)\\
&\qquad \times(F_2H_1+F_1H_2)\Big\}\\
&+ e^{-st'}b(t')\frac{1}{(s+i(\omega-\Delta\omega))}\Big\{\gamma_{21}H_1H_2 + \tfrac{1}{4}G^2\,\tfrac{1}{4}\Gamma_{21}^2(H_1+H_2) - \tfrac{1}{4}(\Gamma^2+\Gamma_{21}^2)\\
&\qquad \times(F_2H_1+F_1H_2)\Big\}\\
&+ e^{-st'}c(t')\frac{1}{(s+i(\omega-\Delta\omega))}\Big\{\gamma_{21}H_1H_2 + \tfrac{1}{4}G^2\,\tfrac{1}{4}\Gamma_{21}^2(H_1+H_2) - \tfrac{1}{4}(\Gamma^2+\Gamma_{21}^2)\\
&\qquad \times(F_2H_1+F_1H_2)\Big\}\\
&+ e^{-st'}d(t')\left\{-(i\lambda\mathcal{E}_0)H_1\left[B_2+\tfrac{1}{4}(\Gamma_{31}^2+\Gamma^2)\right]\right\}\\
&+ e^{-st'}f(t')\left\{(i\lambda\mathcal{E}_0)H_1\left[\tfrac{1}{2}\Gamma_{21}(s+\gamma+i(\omega+\omega_3-\Delta\omega))+\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(s+\tfrac{1}{2}\gamma_{31}+i(\omega-2\Delta\omega))\right]\right\}\\
&+ e^{-st'}e(t')\left\{H_1\left[-\tfrac{1}{4}G^2\,\tfrac{1}{2}\Gamma_{21}+\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})F_2\right]\right\}\\
&+ e^{-st'}\rho_{31}(t')\left\{(i\lambda\mathcal{E}_0)\left[B_1+\tfrac{1}{4}(\Gamma_{31}^2+\Gamma^2)\right]H_2\right\}\\
&+ e^{-st'}\rho_{21}(t')\left\{-(i\lambda\mathcal{E}_0)H_2\left[\tfrac{1}{2}\Gamma_{21}(s+\gamma+i(\omega_2-\Delta\omega))+\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(s+\tfrac{1}{2}\gamma_{31}+i\omega)\right]\right\}\\
&+ e^{-st'}g(t')\left\{H_2\left[-\tfrac{1}{4}G^2\,\tfrac{1}{2}\Gamma_{21}+\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})F_1\right]\right\}
\end{aligned} \tag{7.B.52}$$

From equations (7.B.49) and (50)

$$\hat{a}(s) = \left[\frac{a_3b_0 - a_0b_3}{a_3b_1 - a_1b_3}\right] = \left[\frac{a_3b_0 - a_0b_3}{Z}\right] \tag{7.B.53}$$

$$\hat{c}(s) = \left[\frac{a_2 b_1 - a_1 b_0}{a_2 b_1 - a_1 b_3}\right] = \left[\frac{a_2 b_1 - a_1 b_0}{Z}\right] \tag{7.B.54}$$

and, of course, from (7.B.43), on substituting (7.B.53) and (54), we obtain

$$\hat{b}(s) = \left[\frac{(a_3 - a_1) b_0 - (b_3 - b_1) a_0}{Z}\right] + \frac{e^{-st'}}{(s + i(\omega - \Delta\omega))}\left[a(t') + b(t') + c(t')\right] \tag{7.B.55}$$

where $$Z(s) = a_2(s) b_1(s) - a_1(s) b_2(s) = H_1(s) H_2(s) T(s + i(\omega - \Delta\omega)) \tag{7.B.56}$$

and

$$
\begin{aligned}
T(s + i(\omega - \Delta\omega)) = - \{ & H_1 H_2 (s + \gamma_{31} + i(\omega - \Delta\omega))(s + \gamma_{21} + i(\omega - \Delta\omega)) \\
& + (H_1 + H_2)\tfrac{1}{4}G^2 \left[\tfrac{1}{4}\Gamma^2 (s + \gamma_{21} + i(\omega - \Delta\omega)) \right. \\
& \qquad \left. + \tfrac{1}{4}\Gamma_{31}^2 (s + \gamma_{31} + i(\omega - \Delta\omega))\right] \\
& + 2(B_2 H_1 + B_1 H_2)(s + \gamma_{21} + i(\omega - \Delta\omega))\tfrac{1}{4}G^2 \\
& - (\tfrac{1}{4}G^2)^2 \tfrac{1}{8}\Gamma_{31}^2 (2\Gamma_{21}^2 - \Gamma^2) \\
& - \tfrac{1}{4}G^2 (F_1 B_2 + F_2 B_1)\tfrac{1}{4}(2\Gamma^2 - \Gamma_{31}^2) \\
& - (F_2 H_1 + F_1 H_2)\tfrac{1}{2}\Gamma^2 (s + \gamma + i(\omega - \Delta\omega)) \\
& - \tfrac{1}{4}G^2 \tfrac{1}{4}(2\Gamma^2 - \Gamma_{31}^2)\left[H_1 (s + \tfrac{1}{2}\gamma_{21} + i(\omega + \omega_3 + 2\Delta\omega)) \right. \\
& \qquad \left. + H_2 (s + \tfrac{1}{2}\gamma_{21} + i\omega_2)\right]
\end{aligned} \tag{5.B.57}
$$

We have used the fact that

$$B_1(s) + \tfrac{1}{16}\Gamma^2 G^2 = H_1(s).(s + \tfrac{1}{2}\gamma_{21} + i\omega_2) \tag{7.B.58a}$$

and $$B_2(s) + \tfrac{1}{16}\Gamma^2 G^2 = H_2(s).(s + \tfrac{1}{2}\gamma_{21} + i(\omega + \omega_3 + 2\omega))$$

On substituting the values for $\hat{a}(s)$, $\hat{b}(s)$ and $\hat{c}(s)$ from equations (7.B.53) - (55) in equation (7.B.41) for $\hat{\rho}_{31}(s)$, we obtain:

$$
\begin{aligned}
\hat{\rho}_{31}(s) = \ & \hat{\psi}_{x11}(s) e^{-(s + i(\omega - \Delta\omega))t} \rho_{11}(t') \\
& + \hat{\psi}_{x22}(s) e^{-(s + i(\omega - \Delta\omega))t} \rho_{22}(t') \\
& + \hat{\psi}_{x33}(s) e^{-(s + i(\omega - \Delta\omega))t} \rho_{33}(t') \\
& + \hat{\psi}_{x13}(s) e^{-(s + i2(\omega - \Delta\omega))t} \rho_{13}(t') \\
& + \hat{\psi}_{x12}(s) e^{-(s + i2(\omega - \Delta\omega))t} \rho_{12}(t')
\end{aligned} \tag{7.B.59a}
$$

$$
\left.\begin{aligned}
&+\hat{\psi}_{\alpha 32}(s)\, e^{-(s+i(\omega-\Delta\omega))t'} \rho_{32}(t') && + \hat{u}_{\alpha 32}(s)\rho_{32}(t') \\
&+\hat{\psi}_{\alpha 31}(s)\, e^{-st'} \rho_{31}(t') \Bigg\} && + \hat{u}_{\alpha 31}(s)\rho_{31}(t') \\
&+\hat{\psi}_{\alpha 21}(s)\, e^{-st'} \rho_{21}(t') && + \hat{u}_{\alpha 21}(s)\rho_{21}(t') \\
&+\hat{\psi}_{\alpha 23}(s)\, e^{-(s+i(\omega-\Delta\omega))t'} \rho_{23}(t') \Big) && + \hat{u}_{\alpha 23}(s)\rho_{23}(t')
\end{aligned}\right\} \quad \text{(7.B.59a) cont'd}
$$

where α **= 31, and**

$$
\left.\begin{aligned}
\hat{\psi}_{\alpha 11}(s) = \frac{1}{(s+i(\omega-\Delta\omega))H_1}\, \frac{i\lambda\mathcal{E}_0}{Z} \Big\{ &\Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 + \tfrac14\Gamma_{21}^2\,\tfrac14 G^2(H_1+H_2) \\
&\quad - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))a_1 + (B_1+\tfrac14\Gamma_{21}^2)a_3\Big] \\
&+\Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))b_1 + (B_1+\tfrac14\Gamma_{21}^2)b_3\Big] \\
&-\tfrac14\Gamma_{21}^2 Z\Big\}
\\[2ex]
\hat{\psi}_{\alpha 22}(s) = \frac{1}{(s+i(\omega-\Delta\omega))H_1}\, \frac{i\lambda\mathcal{E}_0}{Z} \Big\{ &\Big[\gamma_{21}H_1H_2 + \tfrac14\Gamma_{21}^2\,\tfrac14 G^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))a_1 + (B_1+\tfrac14\Gamma_{21}^2)a_3\Big] \\
&+\Big[\gamma_{21}H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))b_1 + (B_1+\tfrac14\Gamma_{21}^2)b_3\Big] \\
&-\tfrac14\Gamma_{21}^2 Z\Big\}
\\[2ex]
\hat{\psi}_{\alpha 33}(s) = \frac{1}{(s+i(\omega-\Delta\omega))H_1}\, \frac{i\lambda\mathcal{E}_0}{Z} \Big\{ &\Big[\gamma_{21}H_1H_2 + \tfrac14\Gamma_{21}^2\,\tfrac14 G^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))a_1 + (B_1+\tfrac14\Gamma_{21}^2)a_3\Big] \\
&+\Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big] \\
&\times\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))b_1 + (B_1+\tfrac14\Gamma_{21}^2)b_3\Big] \\
&-\tfrac14\Gamma_{21}^2 Z\Big\}
\end{aligned}\right\} \quad \text{(7.B.59b)}
$$

$$
\begin{aligned}
\hat{\psi}_{\alpha 13}(s) = \frac{\tfrac14 G^2}{Z}\Big\{ &\Big[B_2 + \tfrac14(\Gamma_{31}^2+\Gamma^2)\Big]\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))a_1 + (B_1+\tfrac14\Gamma_{21}^2)a_3\Big] \\
&+\tfrac14\Gamma_{31}^2\Big[(B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2))b_1 + (B_1+\tfrac14\Gamma_{21}^2)b_3\Big]\Big\}
\end{aligned}
$$

$$\hat{\psi}_{\alpha\ldots}(\ ) = -\frac{\ldots G^{2}}{Z}\Big\{\big[\tfrac{1}{2}\Gamma_{21}(s+\gamma+i(\omega_{2}-\Delta\omega)) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(s+\tfrac{1}{2}\gamma_{31}+i(\omega-\Delta\omega))\big]$$
$$\times\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))a_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})a_{3}\big]$$
$$+\tfrac{1}{2}\Gamma_{31}(s+\tfrac{1}{2}\gamma_{31}+i(\omega-\Delta\omega))\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))b_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})b_{3}\big]\Big\}$$

$$\hat{\psi}_{\alpha\ldots}(s) = \frac{i\lambda\varepsilon_{1}}{Z}\Big\{\big[\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})F_{1} - \tfrac{1}{2}\Gamma_{21}\tfrac{1}{4}G^{2}\big]\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))a_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})a_{3}\big]$$
$$+\tfrac{1}{2}\Gamma_{31}F_{2}\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))b_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})b_{3}\big]\Big\}$$

$$\hat{\psi}_{\alpha 11}(\ ) = \frac{1}{H_{1}Z}\Big\{-\tfrac{1}{4}G^{2}H_{2}\big\{\big[\ldots\tfrac{1}{4}(\Gamma_{21}^{2}+\Gamma^{2})\big]\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))a_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})a_{3}\big]$$
$$+\tfrac{1}{4}\Gamma_{31}\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))b_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})b_{3}\big]\big\}$$
$$+B_{1}Z\Big\}$$

$$\hat{\psi}_{\alpha 21}(s) = \frac{1}{H_{1}Z}\Big\{\tfrac{1}{4}G^{2}H_{2}\big\{\big[\tfrac{1}{2}\Gamma_{21}(s+\gamma+i(\omega_{2}-\Delta\omega)) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(s+\tfrac{1}{2}\gamma_{31}+i\omega)\big]$$
$$\times\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))a_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})a_{3}\big]$$
$$+\tfrac{1}{2}\Gamma_{31}(s+\tfrac{1}{2}\gamma_{31}+i\omega)\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))b_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})b_{3}\big]\big\}$$
$$-\tfrac{1}{2}\Gamma_{21}(s+\gamma+i(\omega_{2}-\Delta\omega))Z\Big\}$$

$$\hat{\psi}_{\alpha 23}(s) = \frac{i\lambda\varepsilon}{H_{1}Z}\Big\{H_{2}\big\{\big[\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})F_{1} - \tfrac{1}{2}\Gamma_{21}\tfrac{1}{4}G^{2}\big]\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))a_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})a_{3}\big]$$
$$+\tfrac{1}{2}\Gamma_{31}F_{1}\big[(B_{1}+\tfrac{1}{4}(\Gamma^{2}-\Gamma_{21}^{2}))b_{1} + (B_{1}+\tfrac{1}{4}\Gamma_{21}^{2})b_{3}\big]\big\}$$
$$+\tfrac{1}{2}\Gamma_{21}Z\Big\}$$

(7.B.59b) cont'd

$$\hat{\rho}_{21}(s) = \hat{\psi}_{\beta 11}(s)\,e^{-(s+i(\omega-\Delta\omega))t'}\rho_{11}(t') = \hat{U}_{\beta 11}(s)\rho_{11}(t')$$
$$+\hat{\psi}_{\beta 22}(s)\,e^{-(s+i(\omega-\Delta\omega))t'}\rho_{22}(t') \quad + \hat{U}_{\beta 22}(s)\rho_{22}(t')$$
$$+\hat{\psi}_{\beta 33}(s)\,e^{-(s+i(\omega-\Delta\omega))t'}\rho_{33}(t') \quad + \hat{U}_{\beta 33}(s)\rho_{33}(t')$$
$$+\hat{\psi}_{\beta 13}(s)\,e^{-(s+i2\ldots)t'}\rho_{13}(t') \quad + \hat{U}_{\beta 13}(s)\rho_{13}(t')$$
$$+\hat{\psi}_{\beta 12}(s)\,e^{-(s+i2(\omega-\Delta\omega))t'}\rho_{12}(t') \quad + \hat{U}_{\beta 12}(s)\rho_{12}(t')$$
$$+\hat{\psi}_{\beta 32}(s)\,e^{-(s+i(\omega-\Delta\omega))t'}\rho_{32}(t') \quad + \hat{U}_{\beta 32}(s)\rho_{32}(t')$$

(7.B.60a)

$$
\begin{aligned}
&+\hat{\psi}_{\beta 31}(s)\,e^{-\cdots t}\,\rho_{31}(t) && +\hat{U}_{\beta 31}(s)\,\rho_{31}(t)\\
&+\hat{\psi}_{\beta 21}(s)\,e^{-\cdots t}\,\rho_{21}(t) && +\hat{U}_{\beta 21}(s)\,\rho_{21}(t)\\
&+\hat{\psi}_{\beta 23}(s)\,e^{-(\cdots+i(\omega-\Delta\omega))t}\,\rho_{23}(t) && +\hat{U}_{\beta 23}(s)\,\rho_{23}(t)\Big\}
\end{aligned}
\qquad \text{(7.B.60a) cont'd}
$$

where $\beta = 21$

and

$$
\begin{aligned}
\hat{\psi}_{\beta 11}(s) = {} & \frac{1}{(s+i(\omega-\Delta\omega))}\,\frac{(-i\lambda\mathcal{E}_0)}{H_1 Z}\Big\{\Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 + \tfrac14 G^2\,\tfrac14\Gamma_{21}^2(H_1+H_2)\\
& \qquad - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\Big]\\
& + \Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\Big]\\
& - \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)Z\Big\}\\[1ex]
\hat{\psi}_{\beta 22}(s) = {} & \frac{1}{(s+i(\omega-\Delta\omega))}\,\frac{(-i\lambda\mathcal{E}_0)}{H_1 Z}\Big\{\Big[\gamma_{21}H_1H_2 + \tfrac14 G^2\,\tfrac14\Gamma_{21}^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\Big]\\
& + \Big[\gamma_{21}H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\Big]\\
& - \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)Z\Big\}\\[1ex]
\hat{\psi}_{\beta 33}(s) = {} & \frac{1}{(s+i(\omega-\Delta\omega))}\,\frac{(-i\lambda\mathcal{E}_0)}{H_1 Z}\Big\{\Big[\gamma_{21}H_1H_2 + \tfrac14 G^2\,\tfrac14\Gamma_{21}^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\Big]\\
& + \Big[(s+\gamma_{21}+i(\omega-\Delta\omega))H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big]\\
& \times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\Big]\\
& - \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)Z\Big\}\\[1ex]
\hat{\psi}_{\beta 13}(s) = {} & -\frac{\tfrac14 G^2}{Z}\Big\{\Big[B_2 + \tfrac14(\Gamma_{31}^2+\Gamma^2)\Big]\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\Big]\\
& + \Big[\tfrac14\Gamma_{31}^2\Big]\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1\\
& \quad + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\Big]\Big\}
\end{aligned}
\qquad \text{(7.B.60b)}
$$

$$\hat{\psi}_{\beta[illegible]}(s) = \frac{[illegible]}{Z}\Big\{\big[[illegible](s+\gamma+i(\omega_2-\Delta\omega))\big] \times\big[[illegible](s+\tfrac12\gamma_{31}+i\omega)+\tfrac12\Gamma_{[illegible]}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1 + [illegible](s+\gamma+i(\omega_2-\Delta\omega))+\tfrac12\Gamma_{[illegible]}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\big] + \big[[illegible](s+\gamma+i(\omega_2-\Delta\omega))\big]\big[\{\tfrac12(\Gamma_{[illegible]})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1 + \{[illegible](s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\big]\Big\}$$

$$\hat{\psi}_{\beta32}(s) = \frac{(-i\lambda\epsilon_0)}{Z}\Big\{\big[\tfrac12(\Gamma_{31}+\Gamma_{21})F_1 - \tfrac14 G^2 [illegible]\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\big] + \big[[illegible]\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\big]\Big\}$$

$$\hat{\psi}_{\beta31}(s) = \frac{1}{H_1 Z}\Big\{\tfrac14 G^2\{\big[H_2 F_1 + \tfrac14(\Gamma_{31}^2+\Gamma_{21}^2)\big]\}\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\big] + \big[\tfrac14\Gamma_{31}H_2\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\big]\} - \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))ZH_1\Big\}$$

(7.B.60b) cont'd

$$\hat{\psi}_{\beta21}(s) = \frac{1}{H_1 Z}\Big\{-\tfrac14 G^2 H_2\{\big[\tfrac12(\Gamma_{31}+\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{21}(s+\gamma+i(\omega_2-\Delta\omega))\big] \times\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\big] + \big[\tfrac12\Gamma_{31}(s+\tfrac12\gamma_{31}+i\omega)\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\big]\} + (s+\tfrac12\gamma_{31}+i\omega)(s+\gamma+i(\omega_2-\Delta\omega))Z\Big\}$$

$$\hat{\psi}_{\beta23}(s) = \frac{1}{H_1 Z}(-i\lambda\epsilon_0)\Big\{H_2\{\big[\tfrac12(\Gamma_{31}+\Gamma_{21})F_1 - \tfrac12\Gamma_{21}\tfrac14 G^2\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}a_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\big] + \big[\tfrac12\Gamma_{31}F_1\big]\big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega))\}b_1 + \{\tfrac12\Gamma_{31}(s+\gamma+i(\omega_2-\Delta\omega)) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\big]\} + (s+\tfrac12\gamma_{31}+i\omega)Z\Big\}$$

Taking the inverse Laplace transforms of equations (7.B.59a) and (7.B.60a), we obtain:-

$$\left.\begin{aligned}
\rho_{31}(t')\,\rho_{31}(t'+\tau) = {} & \Psi_{\alpha 11}(\tau)\,e^{\text{[illegible]}}\rho_{11}(t') + \Psi_{\alpha 22}(\tau)\,e^{-i(\omega-\omega)t'}\rho_{22}(t') \\
& + \Psi_{\alpha 33}(\tau)\,e^{\text{[illegible]}}\rho_{33}(t') + \Psi_{\alpha 13}(\tau)\,e^{-i2(\omega-2\omega)t'}\rho_{13}(t') \\
& + \Psi_{\alpha 12}(\tau)\,e^{\text{[illegible]}}\rho_{12}(t') + \Psi_{\alpha 32}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{32}(t') \\
& + \Psi_{\alpha 31}(\tau)\,\rho_{31}(t') + \Psi_{\alpha 21}(\tau)\,\rho_{21}(t') \\
& + \Psi_{\alpha 23}(\tau)\,e^{\text{[illegible]}}\rho_{23}(t')
\end{aligned}\right\} \qquad (7.B.61)$$

and

$$\left.\begin{aligned}
\rho_{21}(t')\,\rho_{21}(t'+\tau) = {} & \Psi_{\beta 11}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{11}(t') + \Psi_{\beta 22}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{22}(t') \\
& + \Psi_{\beta 33}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{33}(t') + \Psi_{\beta 13}(\tau)\,e^{-i2(\omega-2\omega)t'}\rho_{13}(t') \\
& + \Psi_{\beta 12}(\tau)\,e^{-i2(\omega-2\omega)t'}\rho_{12}(t') + \Psi_{\beta 32}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{32}(t') \\
& + \Psi_{\beta 31}(\tau)\,\rho_{31}(t') + \Psi_{\beta 21}(\tau)\,\rho_{21}(t') \\
& + \Psi_{\beta 23}(\tau)\,e^{-i(\omega-2\omega)t'}\rho_{23}(t')
\end{aligned}\right\} \qquad (7.B.62)$$

We shall now proceed to find an expression for the spectral profile after the manner of Chapter III. We recall that equation (3.B.41) gives, for $\underline{r} = 0$,

$$\underline{E}^{(+)}(\underline{R}, t_R) = \underline{E}^{(+)(0)}(\underline{R}, t_R) + \sum_{i=2,3} \frac{[\underline{p}_{i1} - \hat{R}(\hat{R}\cdot\underline{p}_i)]}{R}\,\frac{\omega_{i1}^2}{c^2}\,P_{1i}(t) \qquad (7.B.63)$$

where $t_R = t + R/c$.

I.E.

$$\underline{E}^{(+)}(\underline{R}, t_R) = \underline{E}^{(+)(0)}(\underline{R}, t_R) + \underline{\psi}_2\,P_{12}(t_R - R/c) + \underline{\psi}_3\,P_{13}(t_R - R/c) \qquad (7.B.64)$$

where $\underline{\psi}_2(\hat{R}) = \dfrac{\omega_2^2}{c^2}\,\dfrac{[\underline{p}_{21} - \hat{R}(\hat{R}\cdot\underline{p}_{21})]}{R}$; $\underline{\psi}_3(\hat{R}) = \dfrac{\omega_3^2}{c^2}\,\dfrac{[\underline{p}_{31} - \hat{R}(\hat{R}\cdot\underline{p}_{31})]}{R}$

The 1st order field correlation function is:-

$$\begin{aligned}
G^{(1)}_{jk}(\underline{R}', t'_R; \underline{R}, t_R) &= \langle E_j(\underline{R}', t'_R)\,.\,E_k(\underline{R}, t_R)\rangle \\
&= \langle \{E_j^{(+)(0)+}(\underline{R}', t'_R) + \psi_{2j}^*(\hat{R}')\,P_{12}^+(t') + \psi_{3j}^*(\hat{R}')\,P_{13}(t')\} \\
&\qquad \times \{E_k^{(+)(0)}(\underline{R}, t_R) + \psi_{2k}(\hat{R})\,P_{12}(t) + \psi_{3k}(\hat{R})\,P_{13}(t)\}\rangle \\
&= \langle E_j^{(+)(0)+}(\underline{R}', t'_R)\,.\,E_k^{(+)(0)}(\underline{R}, t_R) \\
&\quad + \psi_{2j}^*(\hat{R})\,.\,\psi_{2k}(\hat{R})\,P_{12}^+(t')\,P_{12}(t) \\
&\quad + \psi_{3j}^*(\hat{R})\,\psi_{3k}(\hat{R})\,P_{13}^+(t')\,P_{13}(t) \\
&\quad + E_j^{(+)(0)+}(\underline{R}', t'_R)\,.\,\psi_{2k}(\hat{R})\,P_{12}(t) + E_j^{(+)(0)+}(\underline{R}', t'_R)\,.\,\psi_{3k}(\hat{R})\,P_{13}(t) \\
&\quad + \psi_{2k}^*(\hat{R}')\,P_{12}^+(t')\,.\,E_k^{(+)(0)}(\underline{R}, t_R) + \psi_{3j}^*(\hat{R}')\,P_{13}(t')\,E_k^{(+)(0)}(\underline{R}, t_R) \\
&\quad + \psi_{2j}^*(\hat{R}')\,P_{12}^+(t')\,\psi_{3k}(\hat{R})\,P_{13}(t) + \psi_{3j}^*(\hat{R}')\,P_{13}^+(t')\,.\,\psi_{2k}(\hat{R})\,P_{12}(t)\rangle
\end{aligned} \qquad (7.B.65)$$

The 1st term in this expression gives the correlation function for the incident beam. The 4th - 7th terms are cross terms and represent interference between the incident and scattered beams of radiation. They are negligible for R, the observation distance, sufficiently large.

Hence the correlation function for the scattered field is, for $\underline{R}' = \underline{R}$:-

$$G_{jk}^{(1)\,\cdots\,\mathrm{II}}(\underline{R},t_R',\underline{R},t_R) = \Big\langle \underline{\psi}_{j}^{*}(\hat{R})\,\underline{\psi}_{2k}(\hat{R})\,P_{12}^{+}(t')\,P_{12}(t) + \underline{\psi}_{j}^{*}(\hat{R})\,\underline{\psi}_{3k}(\hat{R})\,P_{13}^{+}(t')\,P_{13}(t) + \underline{\psi}_{j}^{*}(\hat{R})\,\underline{\psi}_{3k}(\hat{R})\,P_{12}^{+}(t')\,P_{13}(t) + \underline{\psi}_{j}^{*}(\hat{R})\,\underline{\psi}_{2k}(\hat{R})\,P_{13}^{+}(t')\,P_{12}(t)\Big\rangle \quad (7.B.66)$$

But we recall that ψ's are all real, as shown in Lehmberg's paper[2], hence we can write

$$\left.\begin{aligned} G_{jj}^{(1)\,\cdots\,\mathrm{II}}(\underline{R},t_R',\underline{R},t_R) &= \Big\langle \underline{\psi}_{2}\cdot\underline{\psi}_{2}\,P_{12}^{+}(t')\,P_{12}(t) + \underline{\psi}_{3}\cdot\underline{\psi}_{3}\,P_{13}^{+}(t')\,P_{13}(t) \\ &\quad + \underline{\psi}_{2}\cdot\underline{\psi}_{3}\,P_{12}^{+}(t')\,P_{13}(t) + \underline{\psi}_{3}\cdot\underline{\psi}_{2}\,P_{13}^{+}(t')\,P_{12}(t)\Big\rangle \\ &= L_{\hat{R}3}(t_R',t_R) \end{aligned}\right\} \quad (7.B.67)$$

(N.B. This equation could have been obtained from the general expression in Chapter IV equation (4.B.6), remembering that in the present case $\underline{R}_{23} = \underline{R}_{32} = 0$, so that instead of nine terms there are only four.)

Let us now define

$$G_{\cdots\mathrm{II}}^{(1)}(t_R',t_R) = \oint G_{jj}^{(1)\,\cdots\,\mathrm{II}}(\underline{R},t_R',\underline{R},t_R)\,\frac{R^2c}{2\pi\hbar\omega}\,d\Omega_{\hat{R}} = \oint L_{\hat{R}3}(t_R',t_R)\,d\Omega_{\hat{R}} \quad (7.B.68)$$

$$\left.\begin{aligned} &\text{N.B. } \sum_{j} G_{jj}^{(1)}(\underline{R},t_R',\underline{R},t_R)\,\frac{R^2c}{2\pi\hbar\omega} = f_{\hat{R}3}(t_R',t_R) = f_{\hat{R}}^{(1)}(t_R',t_R) + L_{\hat{R}3}(t_R',t) \\ &\text{and } G^{(1)}(t_R',t_R) = f(t_R',t_R) \end{aligned}\right\} \quad (7.B.69)$$

where $f_{\hat{R}}(t_R',t_R)$ and $f(t_R',t_R)$ are defined by equation (3.B.25) and $f_{\hat{R}3}(t_R',t_R)$ and $L_{\hat{R}3}(t_R',t_R)$ by equations (3.B.46) and (3.B.47). (N.B. 3 is replaced by j' not j" since in this chapter we are interested in transitions between levels 3 and 1, not levels 2 and 1 as in Chapter III).

$G_{jj}^{(1)\,\cdots\,\mathrm{II}}(\underline{R},t_R',\underline{R},t_R)$ has the dimensions of $\underline{E}^2$, whereas $G_{\cdots\mathrm{II}}^{(1)}(t_R',t_R)$ has the dimensions of $R^2c/\hbar\omega\,\underline{E}^2 \longrightarrow \left(\frac{R^2c}{\hbar\omega}\right)\Big/\left(\frac{m^2R^2}{e^2t^4}\right)$

since $[eE] = \dfrac{mR}{t^2}$

$$\longrightarrow (m^2c^4)\frac{c}{e^2\hbar\omega} \quad \textbf{since} \quad [c] = \frac{R}{t}$$

$$\longrightarrow (\hbar\omega)\frac{c}{(\hbar c)(\hbar\omega)} \quad \textbf{since} \quad [mc^2] = \hbar\omega \text{ and } [e^2] = \hbar c$$

$$\longrightarrow \omega$$

$$\longrightarrow t^{-1}$$

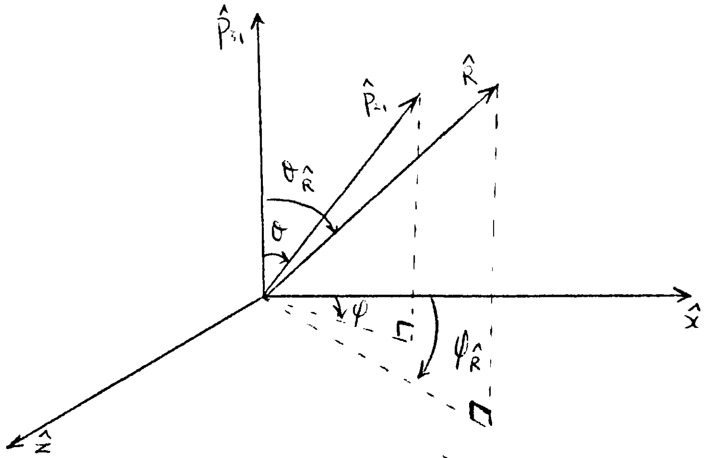
and when $t'_R = t_R$, $G^{(1)}_{\text{rad}}(t_R, t_R)$ represents the total radiation <u>rate</u> (see equation (35) of ref. 2).

Now,

$$\left.\begin{aligned} G^{(1)}_{\text{rad}}(t'_R, t_R) &= \oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega}\Big\{\Big\langle \boldsymbol{\varphi}_3\cdot\boldsymbol{\varphi}_3\, P^{+}_{13}(t')P_{13}(t) + \boldsymbol{\varphi}_2\cdot\boldsymbol{\varphi}_3\, P^{+}_{12}(t')P_{13}(t) \\ &\qquad + \boldsymbol{\varphi}_2\cdot\boldsymbol{\varphi}_2\, P^{+}_{12}(t')P_{12}(t) + \boldsymbol{\varphi}_3\cdot\boldsymbol{\varphi}_2\, P^{+}_{13}(t')P_{12}(t)\Big\rangle\Big\} \\ &= \oint d\Omega_{\hat{R}}\, L_{\hat{R}3}(t'_R, t_R) \end{aligned}\right\} \quad (7.B.70)$$

where $d\Omega_{\hat{R}} = \sin\theta_{\hat{R}}\, d\theta_{\hat{R}}\, d\varphi_{\hat{R}}$, as shown in the following diagram:-

Fig 7B.1



$$\left.\begin{aligned} \hat{P}_{21} &= (\sin\varphi\sin\theta, \cos\theta, \cos\varphi\sin\theta) \\ \hat{R} &= (\sin\varphi_{\hat{R}}\sin\theta_{\hat{R}}, \cos\theta_{\hat{R}}, \cos\varphi_{\hat{R}}\sin\theta_{\hat{R}}) \\ \hat{P}_{31} &= (0, 1, 0) \end{aligned}\right\} \quad (7.B.71)$$

N.B.

$$\boldsymbol{\varphi}_2(\hat{R}) = \frac{\omega_2^2}{c^2}\frac{[\mathbf{p}_{21} - \hat{R}(\hat{R}\cdot\mathbf{p}_{21})]}{R}$$

$$\boldsymbol{\varphi}_3(\hat{R}) = \frac{\omega^2}{c^2}\frac{[\mathbf{p}_{31} - \hat{R}(\hat{R}\cdot\mathbf{p}_{31})]}{R}$$

$$\therefore \boldsymbol{\varphi}_3\cdot\boldsymbol{\varphi}_3 = \frac{\omega^4}{R^2c^4}\, p_{31}^2[1 - (\hat{R}\cdot\hat{p}_{31})^2]$$

$$= \frac{\omega^4}{R^2c^4}\, p_{31}^2 \sin^2\theta_{\hat{R}}$$

$$= \frac{3\hbar\gamma_{31}\omega}{4cR^2}\sin^2\theta_{\hat{R}}$$

(see definition of γ_{31} given by eq. (3.A.24b))

Hence

$$\oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{L}_3 \cdot \mathcal{L}_3 = \frac{R^2 c}{2\pi\hbar\omega}\left(\frac{3\hbar\gamma_{21}\omega}{4cR^2}\right)\int_0^{2\pi} d\phi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \sin\theta_{\hat{R}} \sin\theta_{\hat{R}}$$

$$= \left(\frac{3\gamma_{21}}{8\pi}\right) 2\pi \int_0^{\pi} (1-\cos^2\theta_{\hat{R}})\sin\theta_{\hat{R}}\, d\theta_{\hat{R}}$$

$$= \frac{3\gamma_{21}}{4}\int_{-1}^{1}(1-x^2)\,dx$$

$$\boxed{\oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{L}_3 \cdot \mathcal{L}_3 = \gamma_{21}} \tag{7.B.72}$$

Also,

$$\mathcal{L}_2 \cdot \mathcal{L}_2 = \frac{\omega_2^4}{R^2c^4} p_{21}^2 \left[1-(\hat{p}_{21}\cdot\hat{R})^2\right]$$

$$\therefore \oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{L}_2 \cdot \mathcal{L}_2 = \frac{R^2 c}{2\pi\hbar\omega}\left(\frac{3\hbar\gamma_{21}\omega_2}{4cR^2}\right)\int_0^{2\pi} d\phi_{\hat{R}}\int_0^{\pi} d\theta_{\hat{R}}\left[1-(\hat{p}_{21}\cdot\hat{R})^2\right]\sin\theta_{\hat{R}}$$

$$= \frac{3\gamma_{21}}{8\pi}\frac{\omega_2}{\omega} 2\pi\int_0^{\pi} d\theta_{\hat{R}}\left[1-(\hat{p}_{21}\cdot\hat{R})^2\right]\sin\theta_{\hat{R}}$$

(see definition of γ_{21} in eq. (3.A.24b)

where

$$\int_0^{2\pi} d\phi_{\hat{R}}\int_0^{\pi} d\theta_{\hat{R}}\left[1-(\hat{p}_{21}\cdot\hat{R})^2\right]\sin\theta_{\hat{R}} = \int_0^{2\pi} d\phi_{\hat{R}}\int_0^{\pi} d\theta_{\hat{R}} \sin\theta_{\hat{R}}\Big[1-\cos^2\theta\cos^2\theta_{\hat{R}}$$
$$-\sin^2\theta\cos^2\phi\sin^2\theta_{\hat{R}}\underline{\cos^2\phi_{\hat{R}}}$$
$$-\sin^2\theta\sin^2\theta_{\hat{R}}\sin^2\phi\,\underline{\sin^2\phi_{\hat{R}}}$$
$$-2\sin\theta\cos\theta\cos\phi\cos\theta_{\hat{R}}\sin\theta_{\hat{R}}\times\underline{\cos\phi_{\hat{R}}}$$
$$-2\cos\theta\sin\theta\sin\phi\cos\theta_{\hat{R}}\sin\theta_{\hat{R}}\times\underline{\sin\phi_{\hat{R}}}$$
$$-2\sin^2\theta\sin\phi\cos\phi\sin^2\theta_{\hat{R}}\times\underline{\sin\phi_{\hat{R}}\cos\phi_{\hat{R}}}\Big]$$

Now,

$$\int_0^{2\pi}\cos^2\phi_{\hat{R}}\,d\phi_{\hat{R}} = \pi$$

$$\int_0^{2\pi}\sin^2\phi_{\hat{R}}\,d\phi_{\hat{R}} = \pi$$

$$\int_0^{2\pi}\cos\phi_{\hat{R}}\,d\phi_{\hat{R}} = 0$$

$$\int_0^{2\pi}\sin\phi_{\hat{R}}\,d\phi_{\hat{R}} = 0$$

$$\int_0^{2\pi}\sin\phi_{\hat{R}}\cos\phi_{\hat{R}}\,d\phi_{\hat{R}} = 0$$

$$\therefore \int_0^{2\pi} d\phi_{\hat{R}}\int_0^{\pi} d\theta_{\hat{R}}\left[1-(\hat{p}_{21}\cdot\hat{R})^2\right] = \pi\int_0^{\pi} d\theta_{\hat{R}}\sin\theta_{\hat{R}}\left[2-2\cos^2\theta\cos^2\theta_{\hat{R}}-\sin^2\theta\sin^2\theta_{\hat{R}}\right]$$

Now

$$\int_0^{\pi} \sin\theta_{\hat{R}}\, d\theta_{\hat{R}} = 2$$

$$\int_0^{\pi} \cos^2\theta_{\hat{R}} \sin\theta_{\hat{R}}\, d\theta_{\hat{R}} = 2/3$$

$$\int_0^{\pi} \sin^2\theta \sin\theta_{\hat{R}}\, d\theta_{\hat{R}} = 4/3$$

$$\therefore \int_0^{2\pi} d\varphi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \left[1 - (\hat{p}_{21} \cdot \hat{R})^2\right] \sin\theta_{\hat{R}} = \pi\left[4 - 4/3\cos^2\theta - 4/3\sin^2\theta\right]$$

$$= 8\pi/3$$

$$\therefore \int d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{E}_2 \cdot \mathcal{E}_2 = \frac{3\gamma_{21}}{8\pi} \frac{\omega_2}{\omega} \frac{8\pi}{3}$$

$$\boxed{\oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{E}_2 \cdot \mathcal{E}_2 = \frac{\omega_2}{\omega} \gamma_{21}} \qquad (7.B.73)$$

Next we must consider:-

$$\oint d\Omega_{\hat{R}} \frac{R^2 c}{2\pi\hbar\omega} \mathcal{E}_2 \cdot \mathcal{E}_3 = \frac{R^2 c}{2\pi\hbar\omega} \frac{\omega_2^2 \omega^2}{R^2 c^4} p_{21} p_{31} \int_0^{2\pi} d\varphi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \left[\hat{p}_{21} - \hat{R}(\hat{R} \cdot \hat{p}_{21})\right] \times \left[\hat{p}_{31} - \hat{R}(\hat{R} \cdot \hat{p}_{31})\right]$$

$$= \frac{\omega_2^2 \omega}{2\pi\hbar c^3} p_{21} p_{31} \int_0^{2\pi} d\varphi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \left[\hat{p}_{21} \cdot \hat{p}_{31} - (\hat{p}_{21} \cdot \hat{R})(\hat{p}_{31} \cdot \hat{R})\right]$$

where $\hat{p}_{21} \cdot \hat{R} = \sin\varphi \sin\theta \sin\varphi_{\hat{R}} \sin\theta_{\hat{R}} + \cos\theta \cos\theta_{\hat{R}} + \cos\varphi \sin\theta \cos\varphi_{\hat{R}} \sin\theta_{\hat{R}}$

$\hat{p}_{21} \cdot \hat{p}_{31} = \cos\theta$

$\hat{p}_{31} \cdot \hat{R} = \cos\theta_{\hat{R}}$

$$\therefore \int_0^{2\pi} d\varphi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \sin\theta_{\hat{R}} = \int_0^{2\pi} d\varphi_{\hat{R}} \int_0^{\pi} d\theta_{\hat{R}} \sin\theta_{\hat{R}} \left[\cos\theta - \sin\varphi \sin\theta \underline{\sin\varphi_{\hat{R}}} \sin\theta_{\hat{R}} \cos\theta_{\hat{R}} - \cos\theta \cos^2\theta_{\hat{R}} - \cos\varphi \sin\theta \underline{\cos\varphi_{\hat{R}}} \sin\theta_{\hat{R}} \cos\theta_{\hat{R}}\right]$$

$$= 2\pi \int_0^{\pi} d\theta_{\hat{R}} \sin\theta_{\hat{R}} \left[\cos\theta - \cos\theta \cos^2\theta_{\hat{R}}\right]$$

$$= 2\pi \cos\theta \int_0^{\pi} d\theta_{\hat{R}} \left[\sin\theta_{\hat{R}} - \sin\theta_{\hat{R}} \cos^2\theta_{\hat{R}}\right]$$

$$= 2\pi \cos\theta \left[2 - 2/3\right]$$

$$= 8\pi/3 \cos\theta$$

$$= \frac{\omega_2^2 \omega}{2\pi\hbar c^3} p_{21} p_{31} \frac{8\pi}{3} \cos\theta$$

$$= \frac{\omega_2^2}{\omega^2} \frac{4}{3} \frac{\omega^3}{\hbar c^3} p_{21} p_{31} \qquad = \frac{\omega}{\omega_2} \frac{4}{3} \frac{\omega_2^3}{c^3} \mathbf{p}_{21} \cdot \mathbf{p}_{31}$$

$$\boxed{\oint d\Omega_k \frac{k^2 c}{2\pi\hbar\omega} \mathscr{g}_2 \cdot \mathscr{g}_3 = \frac{\omega_2^2}{\omega^2} \Gamma_{31} = \frac{\omega}{\omega_2} \Gamma_{21}} \tag{7.B.74}$$

(see definitions of Γ_{31} and Γ_{21} given in equation (3.A.24b).

Hence,

$$\left.\begin{aligned} G^{(1)}_{n}(t_R', t_R) &= \gamma_{31} \langle P_{13}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega_2}{\omega} \gamma_{21} \langle P_{12}^+(t') P_{12}(t'+\tau)\rangle \\ &+ \frac{\omega_2^2}{\omega^2} \Gamma_{31} \langle P_{12}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega}{\omega_2} \Gamma_{21} \langle P_{13}^+(t') P_{12}(t'+\tau)\rangle \\ &= \oint d\Omega_k L_{n}(t_R', t_R) \end{aligned}\right\} \tag{7.B.75a}$$

$$\left.\begin{aligned} \frac{1}{\gamma_{31}} G^{(1)}_{n}(t_R, t_R)\ g(\tau, t') &= \langle P_{13}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega_2}{\omega} \frac{\gamma_{21}}{\gamma_{31}} \langle P_{12}^+(t') P_{12}(t'+\tau)\rangle \\ &+ \frac{\omega_2^2}{\omega^2} \frac{\Gamma_{31}}{\gamma_{31}} \langle P_{12}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} \langle P_{13}^+(t') P_{12}(t'+\tau)\rangle \end{aligned}\right\} \tag{7.B.75b}$$

where $t = t' + \tau$.

Now, since $\frac{\omega_2^2}{\omega^2} \frac{\Gamma_{31}}{\gamma_{31}} = \frac{\omega_2^2}{\omega^2} \frac{\mathscr{P}_{21} \cdot \mathscr{P}_{31}}{P_{31}^2}$

and $\frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} = \frac{\omega}{\omega_2} \frac{\mathscr{P}_{21} \cdot \mathscr{P}_{31}}{P_{31}^2} \frac{\omega_2^3}{\omega^3} = \frac{\omega_2^2}{\omega^2} \frac{\mathscr{P}_{21} \cdot \mathscr{P}_{31}}{P_{31}^2}$,

we can write

$$\left.\begin{aligned} g(\tau, t') &= \langle P_{13}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega_2}{\omega} \frac{\gamma_{21}}{\gamma_{31}} \langle P_{12}^+(t') P_{12}(t'+\tau)\rangle \\ &+ \frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} \langle P_{12}^+(t') P_{13}(t'+\tau)\rangle + \frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} \langle P_{13}^+(t') P_{12}(t'+\tau)\rangle \end{aligned}\right\} \tag{7.B.75c}$$

Let

$$\left.\begin{aligned} g_1(\tau, t') &= \langle P_{13}^+(t')\ P_{13}(t'+\tau)\rangle \\ g_2(\tau, t') &= \langle P_{12}^+(t')\ P_{13}(t'+\tau)\rangle \\ g_3(\tau, t') &= \langle P_{12}^+(t')\ P_{12}(t'+\tau)\rangle \\ g_4(\tau, t') &= \langle P_{13}^+(t')\ P_{12}(t'+\tau)\rangle \\ g(\tau, t') &= g_1(\tau, t') + \frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} g_2(\tau, t') + \frac{\omega_2}{\omega} \frac{\gamma_{21}}{\gamma_{31}} g_3(\tau, t') + \frac{\omega}{\omega_2} \frac{\Gamma_{21}}{\gamma_{31}} g_4(\tau, t') \end{aligned}\right\} \tag{7.B.76}$$

Under the Markoff approximation, we know that

$$\langle P_{13}^+(t') P_{13}(t'+\tau)\rangle \triangleq \sum_{i=1}^{3} U_{31,i1}(\tau, t') \rho_{i3}^{(s)}(t') \tag{7.B.77}$$

and $\langle P_{12}^{+}(t')P_{12}(t'+\tau)\rangle \doteq \sum_{i=1}^{3} U_{21,i1}(\tau,t')\rho_{i2}^{(s)}(t')$ (7.B.78)

(see Chapter IV equation (4.C.8)).

Similarly,

$$\langle P_{12}^{+}(t')P_{13}(t'+\tau)\rangle \doteq_{MA} Tr\{|i_{ph}\rangle\langle i_{ph}|\rho^{(s)}(t')P_{12}^{+}(0)U^{-1}(t'+\tau,t')P_{13}(0)U(t'+\tau,t')\} \quad (7.B.79)$$

and

$$\langle P_{13}(t'+\tau)\rangle \rho_{31}(t'+\tau) \doteq_{MA} Tr\{|i_{ph}\rangle\langle i_{ph}|\rho^{(s)}(t')P_{12}^{+}(0)U^{-1}(t'+\tau,t')P_{13}(0)U(t'+\tau,t')\} \quad (7.B.80)$$

so that the former equation can be obtained from the latter by the substitution

$$\rho^{(s)}(t') \longrightarrow \rho^{(s)}(t')P_{12}^{+}(0) \quad (7.B.81)$$

Let:-

$$\rho_{31}(t'+\tau) = \sum_{i=1}^{3}\sum_{j=1}^{3} U_{31,ij}(\tau,t')\rho_{ij}^{(s)}(t') \quad (7.B.82)$$

Now

$$\left.\begin{aligned}\rho_{ij}^{(s)}(t') &\longrightarrow \langle i|\rho^{(s)}(t')P_{12}^{+}(0)|j\rangle \\ &= \delta_{j1}\rho_{i2}^{(s)}(t')\end{aligned}\right\} \quad (7.B.83)$$

$$\therefore \langle P_{12}^{+}(t')P_{13}(t'+\tau)\rangle \doteq_{MA} \sum_{i=1}^{3} U_{31,i1}(\tau,t')\rho_{i2}^{(s)}(t') \quad (7.B.84)$$

On the other hand:-

$$\langle P_{13}^{+}(t')P_{12}(t'+\tau)\rangle \doteq_{MA} Tr\{|i_{ph}\rangle\langle i_{ph}|\rho^{(s)}(t')P_{13}^{+}(0)U^{-1}(t'+\tau,t')P_{12}(0)U(t'+\tau,t')\} \quad (7.B.85)$$

and

$$\langle P_{12}(t'+\tau)\rangle = \rho_{21}(t'+\tau) \doteq_{MA} Tr\{|i_{ph}\rangle\langle i_{ph}|\rho^{(s)}(t')U^{-1}(t'+\tau,t')P_{12}(0)U(t'+\tau,t')\} \quad (7.B.86)$$

so for $\rho_{21}(t'+\tau) \longrightarrow \langle P_{13}^{+}(t')P_{12}(t'+\tau)\rangle$

we require that $\rho^{(s)}(t') \longrightarrow \rho^{(s)}(t')P_{13}^{+}(0)$ (7.B.87)

Thus if $\rho_{21}(t'+\tau) = \sum_{i=1}^{3}\sum_{j=1}^{3} U_{21,ij}(\tau,t')\rho_{ij}^{(s)}(t')$ (7.B.88)

then $\rho_{ij}^{(s)}(t') \longrightarrow \langle i|\rho^{(s)}(t')P_{13}^{+}(0)|j\rangle = \delta_{j1}\rho_{i3}^{(s)}(t')$ (7.B.89)

$$\therefore \langle P_{13}^{+}(t')P_{12}(t'+\tau)\rangle \doteq_{MA} \sum_{i=1}^{3} U_{21,i1}(\tau,t')\rho_{i3}^{(s)}(t') \quad (7.B.90)$$

Hence

$$\left.\begin{aligned} g_1(\tau,t') &= \langle P_{12}^{+}(t')P_{13}(t'+\tau)\rangle \\ &\stackrel{\wedge}{=}_{MA} \sum_{i=1}^{3} U_{31,i1}(\tau,t')\rho_{i3}^{(s)}(t') \\ &= U_{31,11}(\tau,t')\rho_{13}^{(s)}(t') + U_{31,21}(\tau,t')\rho_{23}^{(s)}(t') + U_{31,31}(\tau,t')\rho_{33}^{(s)}(t') \\ &= \psi_{\alpha,11}(\tau)e^{-i(\omega-\Delta\omega)t'}\rho_{13}^{(s)}(t') + \psi_{\alpha,21}(\tau)\rho_{23}^{(s)}(t') + \psi_{\alpha,31}(\tau)\rho_{33}^{(s)}(t') \end{aligned}\right\} \tag{7.B.91}$$

$$\left.\begin{aligned} g_2(\tau,t') &= \langle P_{12}^{+}(t')P_{13}(t'+\tau)\rangle \\ &\stackrel{\wedge}{=}_{MA} \sum_{i=1}^{3} U_{31,i1}(\tau,t')\rho_{i2}^{(s)}(t') \\ &= U_{31,11}(\tau,t')\rho_{12}^{(s)}(t') + U_{31,21}(\tau,t')\rho_{22}^{(s)}(t') + U_{31,31}(\tau,t')\rho_{32}^{(s)}(t') \\ &= \psi_{\alpha,11}(\tau)e^{-i(\omega-\Delta\omega)t'}\rho_{12}^{(s)}(t') + \psi_{\alpha,21}(\tau)\rho_{22}^{(s)}(t') + \psi_{\alpha,31}(\tau)\rho_{32}^{(s)}(t') \end{aligned}\right\} \tag{7.B.92}$$

$$\left.\begin{aligned} g_3(\tau,t') &= \langle P_{12}^{+}(t')P_{12}(t'+\tau)\rangle \\ &\stackrel{\wedge}{=}_{MA} \sum_{i=1}^{3} U_{21,i1}(\tau,t')\rho_{i2}^{(s)}(t') \\ &= U_{21,11}(\tau,t')\rho_{12}^{(s)}(t') + U_{21,21}(\tau,t')\rho_{22}^{(s)}(t') + U_{21,31}(\tau,t')\rho_{32}^{(s)}(t') \\ &= \psi_{\beta,11}(\tau)e^{-i(\omega-\Delta\omega)t'}\rho_{12}^{(s)}(t') + \psi_{\beta,21}(\tau)\rho_{22}^{(s)}(t') + \psi_{\beta,31}(\tau)\rho_{32}^{(s)}(t') \end{aligned}\right\} \tag{7.B.93}$$

$$\left.\begin{aligned} g_4(\tau,t') &= \langle P_{13}^{+}(t')P_{12}(t'+\tau)\rangle \\ &\stackrel{\wedge}{=}_{MA} \sum_{i=1}^{3} U_{21,i1}(\tau,t')\rho_{i3}^{(s)}(t') \\ &= U_{21,11}(\tau,t')\rho_{13}^{(s)}(t') + U_{21,21}(\tau,t')\rho_{23}^{(s)}(t') + U_{21,31}(\tau,t')\rho_{33}^{(s)}(t') \\ &= \psi_{\beta 11}(\tau)e^{-i(\omega-\Delta\omega)t'}\rho_{13}^{(s)}(t') + \psi_{\beta 21}(\tau)\rho_{23}^{(s)}(t') + \psi_{\beta 31}(\tau)\rho_{33}^{(s)}(t') \end{aligned}\right\} \tag{7.B.94}$$

$$\left.\begin{aligned} g(\tau,t') &= g_1(\tau,t') + \omega/\omega_2\, \Gamma_{21}/\gamma_{31}\, g_2(\tau,t') + \omega_2/\omega\, \gamma_{21}/\gamma_{31}\, g_3(\tau,t') + \omega/\omega_2\, \Gamma_{21}/\gamma_{31}\, g_4(\tau,t') \\ &= \Big[\psi_{\alpha 11}(\tau)e^{-i(\omega-\Delta\omega)t'}\rho_{13}^{(s)}(t') + \psi_{\alpha 21}(\tau)\rho_{23}^{(s)}(t') + \psi_{\alpha 31}(\tau)\rho_{33}^{(s)}(t')\Big] \end{aligned}\right\} \tag{7.B.95}$$

$$
\left.
\begin{aligned}
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\psi_{\alpha 11}(\tau)e^{-i(\omega-\omega_0)t'}\rho_{12}^{(1)}(t') + \psi_{\alpha 21}(\tau)\rho_{22}^{(1)}(t') + \psi_{\alpha 31}(\tau)\rho_{32}^{(1)}(t')\right]\\
&+\frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left[\psi_{\beta 11}(\tau)e^{-i(\omega-\omega_0)t'}\rho_{12}^{(1)}(t') + \psi_{\beta 21}(\tau)\rho_{22}^{(1)}(t') + \psi_{\beta 31}(\tau)\rho_{32}^{(1)}(t')\right]\\
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\psi_{\beta 11}(\tau)e^{-i(\omega-\omega_0)t'}\rho_{13}^{(1)}(t') + \psi_{\beta 21}(\tau)\rho_{23}^{(1)}(t') + \psi_{\beta 31}(\tau)\rho_{33}^{(1)}(t')\right]
\end{aligned}
\right\}
\quad \text{(7.B.95) cont'd}
$$

Assuming the atom to be in equilibrium with the field, we write

$$\rho_{ij}(t') = (\rho_{ij}(t\to\infty))_{t=t'}$$

Let

$$
\left.
\begin{aligned}
&(\rho_{13\alpha})_{t,t'} = e^{i(\omega-\omega_0)t'}\bar{\rho}_{13}\\
&(\rho_{12})_{t,t'} = e^{i(\omega-\omega_0)t'}\bar{\rho}_{12}\\
&(\rho_{23})_{t,t'} = \bar{\rho}_{23} \qquad , \quad (\rho_{22\alpha})_{t,t'} = \bar{\rho}_{22}\\
&(\rho_{33\alpha})_{t,t'} = \bar{\rho}_{33} \qquad , \quad (\rho_{32\alpha})_{t,t'} = \bar{\rho}_{32}
\end{aligned}
\right\}
\quad \text{(7.B.96)}
$$

where $\bar{\rho}_{ij}$'s are independent of t', as we are about to show.

Hence

$$
\left.
\begin{aligned}
g(\tau) = &\left[\psi_{\alpha 11}(\tau)\bar{\rho}_{13} + \psi_{\alpha 21}(\tau)\bar{\rho}_{23} + \psi_{\alpha 31}(\tau)\bar{\rho}_{33}\right]\\
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\psi_{\alpha 11}(\tau)\bar{\rho}_{12} + \psi_{\alpha 21}(\tau)\bar{\rho}_{22} + \psi_{\alpha 31}(\tau)\bar{\rho}_{32}\right]\\
&+\frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left[\psi_{\beta 11}(\tau)\bar{\rho}_{12} + \psi_{\beta 21}(\tau)\bar{\rho}_{22} + \psi_{\beta 31}(\tau)\bar{\rho}_{32}\right]\\
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\psi_{\beta 11}(\tau)\bar{\rho}_{13} + \psi_{\beta 21}(\tau)\bar{\rho}_{23} + \psi_{\beta 31}(\tau)\bar{\rho}_{33}\right]
\end{aligned}
\right\}
\quad \text{(7.B.97)}
$$

Then the total spectral correlation function is

$$
\left.
\begin{aligned}
\tilde{g}(\nu) = 2\,\mathrm{Re}\Big\{ &\left[\hat{\psi}_{\alpha 11}(-\nu)\bar{\rho}_{13} + \hat{\psi}_{\alpha 21}(-\nu)\bar{\rho}_{23} + \hat{\psi}_{\alpha 31}(-\nu)\bar{\rho}_{33}\right]\\
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\hat{\psi}_{\alpha 11}(-\nu)\bar{\rho}_{12} + \hat{\psi}_{\alpha 21}(-\nu)\bar{\rho}_{22} + \hat{\psi}_{\alpha 31}(-\nu)\bar{\rho}_{32}\right]\\
&+\frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left[\hat{\psi}_{\beta 11}(-\nu)\bar{\rho}_{12} + \hat{\psi}_{\beta 21}(-\nu)\bar{\rho}_{22} + \hat{\psi}_{\beta 31}(-\nu)\bar{\rho}_{32}\right]\\
&+\frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left[\hat{\psi}_{\beta 11}(-\nu)\bar{\rho}_{13} + \hat{\psi}_{\beta 21}(-\nu)\bar{\rho}_{23} + \hat{\psi}_{\beta 31}(-\nu)\bar{\rho}_{33}\right]
\end{aligned}
\right\}
\quad \text{(7.B.98a)}
$$

(N.B. $\bar{\rho}_{33}$ and $\bar{\rho}_{22}$ are real since they represent state populations).

We can rearrange terms so that:-

$$\begin{aligned} j(\nu) = 2\mathrm{Re}\Big\{ & \hat{\psi}_{\alpha 31}(-\nu)\left[\bar{\rho}_{13} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{12}\right] + \hat{\psi}_{\alpha 21}(-\nu)\left[\bar{\rho}_{13} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{22}\right] \\ & + \hat{\psi}_{\alpha 31}(-\nu)\left[\bar{\rho}_{33} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{32}\right] \\ & + \tfrac{\omega}{\omega}\tfrac{\Gamma_{21}}{\gamma_{21}}\Big[\hat{\psi}_{\beta 21}(-\nu)\left[\bar{\rho}_{12} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{13}\right] + \hat{\psi}_{\beta 21}(-\nu)\left[\bar{\rho}_{22} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{23}\right] \\ & + \hat{\psi}_{\beta 31}(-\nu)\left[\bar{\rho}_{32} + \tfrac{\omega^2}{\omega_2^2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{33}\right]\Big]\Big\} \end{aligned} \tag{7.B.98b}$$

We shall now proceed to find $\bar{\rho}_{13}$, $\bar{\rho}_{23}$, $\bar{\rho}_{33}$. At equilibrium, the diagonal density matrix elements are constant

i.e.

$$\left.\begin{aligned} \rho_{11}(t\to\infty) &= \bar{\rho}_{11} \\ \rho_{22}(t\to\infty) &= \bar{\rho}_{22} \\ \rho_{33}(t\to\infty) &= \bar{\rho}_{33} \end{aligned}\right\} \tag{7.B.99a}$$

but the following diagonal elements are harmonically varying

$$\left.\begin{aligned} \rho_{31}(t\to\infty) &= \bar{\rho}_{31}\, e^{-i(\omega-\Delta\omega)t} \\ \rho_{13}(t\to\infty) &= \bar{\rho}_{13}\, e^{+i(\omega-\Delta\omega)t} \\ \rho_{21}(t\to\infty) &= \bar{\rho}_{21}\, e^{-i(\omega-\Delta\omega)t} \\ \rho_{12}(t\to\infty) &= \bar{\rho}_{12}\, e^{+i(\omega-\Delta\omega)t} \end{aligned}\right\} \tag{7.B.99b}$$

whereas $\rho_{32}(t\to\infty) = \bar{\rho}_{32}$

and $\rho_{23}(t\to\infty) = \bar{\rho}_{23}$ (7.B.99c)

are constant since the levels 3 and 2 are not connected by a dipole moment (see ref 65).

Hence at equilibrium

$$\left.\begin{aligned} \dot{\rho}_{11}(t\to\infty) &= 0 \\ \dot{\rho}_{22}(t\to\infty) &= 0 \\ \dot{\rho}_{33}(t\to\infty) &= 0 \\ \dot{\rho}_{32}(t\to\infty) &= 0 \\ \dot{\rho}_{23}(t\to\infty) &= 0 \\ \dot{\rho}_{31}(t\to\infty) &= -i(\omega-\Delta\omega)\bar{\rho}_{31}\, e^{-i(\omega-\Delta\omega)t} \\ \dot{\rho}_{13}(t\to\infty) &= +i(\omega-\Delta\omega)\bar{\rho}_{13}\, e^{+i(\omega-\Delta\omega)t} \\ \dot{\rho}_{21}(t\to\infty) &= -i(\omega-\Delta\omega)\bar{\rho}_{21}\, e^{-i(\omega-\Delta\omega)t} \\ \dot{\rho}_{12}(t\to\infty) &= +i(\omega-\Delta\omega)\bar{\rho}_{12}\, e^{+i(\omega-\Delta\omega)t} \end{aligned}\right\} \tag{7.B.100}$$

so that the equations of motion (7.A.43) - (51) at equilibrium reduce to:

i.e.

$$\left.\begin{aligned}0 = -i\lambda\mathcal{E}_0\bar{\rho}_{13} + i\lambda\mathcal{E}_0\bar{\rho}_{31} + \gamma_{21}\bar{\rho}_{22} + \gamma_{31}\bar{\rho}_{33} &+ \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})\bar{\rho}_{32}\\ &+ \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})\bar{\rho}_{23}\end{aligned}\right\} \quad (7.B.101)$$

$$\gamma_{21}\bar{\rho}_{22} = -\tfrac{1}{2}\Gamma_{31}\bar{\rho}_{32} - \tfrac{1}{2}\Gamma_{31}\bar{\rho}_{23} \quad (7.B.102)$$

$$\gamma_{21}\bar{\rho}_{33} = i\lambda\mathcal{E}_0\bar{\rho}_{13} - i\lambda\mathcal{E}_0\bar{\rho}_{31} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{32} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{23} \quad (7.B.103)$$

$$\{\tfrac{1}{2}\gamma_{21} - i(\omega_3-\Delta\omega)\}\bar{\rho}_{21} = i\lambda\mathcal{E}_0\bar{\rho}_{23} - \tfrac{1}{2}\Gamma_{31}\bar{\rho}_{31} \quad (7.B.104)$$

$$(\tfrac{1}{2}\gamma_{31} - i\Delta\omega)\bar{\rho}_{13} = -i\lambda\mathcal{E}_0\bar{\rho}_{11} + i\lambda\mathcal{E}_0\bar{\rho}_{33} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{12} \quad (7.B.105)$$

$$\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) + i\omega_3\}\bar{\rho}_{32} = i\lambda\mathcal{E}_0\bar{\rho}_{12} - \tfrac{1}{2}\Gamma_{31}\bar{\rho}_{33} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{22} \quad (7.B.106)$$

$$\{\tfrac{1}{2}\gamma_{21} + i(\omega_3-\Delta\omega)\}\bar{\rho}_{12} = i\lambda\mathcal{E}_0\bar{\rho}_{32} - \tfrac{1}{2}\Gamma_{31}\bar{\rho}_{13} \quad (7.B.107)$$

$$(\tfrac{1}{2}\gamma_{31} + i\Delta\omega)\bar{\rho}_{31} = i\lambda\mathcal{E}_0\bar{\rho}_{11} - i\lambda\mathcal{E}_0\bar{\rho}_{33} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{21} \quad (7.B.108)$$

$$\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) - i\omega_3\}\bar{\rho}_{23} = -i\lambda\mathcal{E}_0\bar{\rho}_{21} - \tfrac{1}{2}\Gamma_{31}\bar{\rho}_{33} - \tfrac{1}{2}\Gamma_{21}\bar{\rho}_{22} \quad (7.B.109)$$

As in the general case, we shall solve equations (7.B.104), (7.B.108), (7.B.109) for $\bar{\rho}_{21}$, $\bar{\rho}_{31}$, and $\bar{\rho}_{23}$ in terms of $\bar{\rho}_{11}$, $\bar{\rho}_{22}$ and $\bar{\rho}_{33}$. Hence

$$\bar{\rho}_{23} = -\tfrac{1}{2}\Gamma_{31}\tfrac{1}{4}G^2\frac{1}{H_0}\bar{\rho}_{11} - \tfrac{1}{2}\Gamma_{21}\frac{F_0}{H_0}\bar{\rho}_{22} - \tfrac{1}{2}\Gamma_{31}\frac{(F_0-\tfrac{1}{4}G^2)}{H_0}\bar{\rho}_{33} \quad (7.B.110)$$

$$\left.\begin{aligned}\bar{\rho}_{21} = -(i\lambda\mathcal{E}_0)\tfrac{1}{2}\Gamma_{31}\frac{(\gamma - i\omega_3)}{H_0}\bar{\rho}_{11} &+ (i\lambda\mathcal{E}_0)\tfrac{1}{2}\Gamma_{21}\frac{(\tfrac{1}{2}\gamma_{31}+i\Delta\omega)}{H_0}\bar{\rho}_{22}\\ &+ (i\lambda\mathcal{E}_0)\tfrac{1}{2}\Gamma_{31}\frac{(\gamma+\tfrac{1}{2}\gamma_{31} - i(\omega_3-\Delta\omega))}{H_0}\bar{\rho}_{33}\end{aligned}\right\} \quad (7.B.111)$$

$$\bar{\rho}_{31} = (i\lambda\mathcal{E}_0)\frac{B_0}{H_0}\bar{\rho}_{11} - (i\lambda\mathcal{E}_0)\tfrac{1}{4}\Gamma_{21}^2\frac{1}{H_0}\bar{\rho}_{22} - (i\lambda\mathcal{E}_0)\frac{(B_0+\tfrac{1}{4}\Gamma^2)}{H_0}\bar{\rho}_{33} \quad (7.B.112)$$

On solving equations (7.B.106), (107), (105) for , and we obtain the h.c.'s of the above three equations, and, on substituting these six equations in (7.B.101) - (103) we find that:

$$0 = \bar{\rho}_{11}\left[\frac{-\tfrac{1}{2}(\Gamma_{21}+\Gamma_{31})\tfrac{1}{2}\Gamma_{31}\tfrac{1}{4}G^2(H_0+H_0^*)}{|H_0|^2} - \frac{\tfrac{1}{4}G^2(B_0^*H_0+B_0H_0^*)}{|H_0|^2}\right] \quad (7.B.113)$$

$$+\bar{\rho}_{22}\left[\gamma_{31}-\frac{\tfrac{1}{2}\Gamma_{21}}{|H_0|^2}\left\{\tfrac{1}{2}(\Gamma_{21}+\Gamma_{31})(F_0H_0^*+F_0^*H_0)-\tfrac{1}{2}\Gamma_{21}\tfrac{1}{4}G^2(H_0+H_0^*)\right\}\right]$$

$$+\bar{\rho}_{33}\left[\gamma_{31}-\frac{\tfrac{1}{2}(\Gamma_{21}+\Gamma_{31})\tfrac{1}{2}\Gamma_{31}}{|H_0|^2}\left\{(F_0H_0^*+F_0^*H_0)-\tfrac{1}{4}G^2(H_0+H_0^*)\right\}\right.$$

$$+\frac{\tfrac{1}{4}G^2(B_0^*H_0+B_0H_0^*)}{|H_0|^2}$$

$$\left.+\frac{\tfrac{1}{4}G^2\tfrac{1}{4}\Gamma^2(H_0+H_0^*)}{|H_0|^2}\right] \qquad \text{(7.B.113) cont'd}$$

$$0=\bar{\rho}_{11}\left[\frac{\tfrac{1}{4}\Gamma_{31}^2\tfrac{1}{4}G^2(H_0+H_0^*)}{|H_0|^2}\right]+\bar{\rho}_{22}\left[-\gamma_{21}+\frac{\tfrac{1}{4}\Gamma^2(F_0H_0^*+F_0^*H_0)}{|H_0|^2}\right]+\bar{\rho}_{33}\left[\frac{\tfrac{1}{4}\Gamma_{31}^2}{|H_0|^2}\left\{(F_0H_0^*+F_0^*H_0)-\tfrac{1}{4}G^2(H_0+H_0^*)\right\}\right] \qquad \text{(7.B.114)}$$

$$0=\bar{\rho}_{11}\left[\frac{\tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*)}{|H_0|^2}+\frac{\tfrac{1}{4}G^2(B_0^*H_0+B_0H_0^*)}{|H_0|^2}\right]$$

$$+\bar{\rho}_{22}\left[\frac{\tfrac{1}{4}\Gamma_{31}^2(F_0H_0^*+F_0^*H_0)}{|H_0|^2}-\frac{\tfrac{1}{4}G^2\tfrac{1}{4}\Gamma_{31}^2(H_0+H_0^*)}{|H_0|^2}\right]$$

$$+\bar{\rho}_{33}\left[-\gamma_{31}+\frac{\tfrac{1}{4}\Gamma^2}{|H_0|^2}\left\{(F_0H_0^*+F_0^*H_0)-\tfrac{1}{4}G^2(H_0+H_0^*)\right\}\right.$$

$$-\frac{\tfrac{1}{4}G^2(B_0^*H_0+B_0H_0^*)}{|H_0|^2}$$

$$\left.-\frac{\tfrac{1}{4}G^2\tfrac{1}{4}\Gamma^2(H_0+H_0^*)}{|H_0|^2}\right] \qquad \text{(7.B.115)}$$

where

$$H_0=(\gamma-i\omega_3)F_0+\tfrac{1}{4}G^2(\tfrac{1}{2}\gamma_{21}+i\Delta\omega)$$

$$F_0=(\tfrac{1}{2}\gamma_{21}+i\Delta\omega)(\tfrac{1}{2}\gamma_{21}-i(\omega_3-\Delta\omega))-\tfrac{1}{4}\Gamma^2 \qquad \text{(7.B.116)}$$

$$B_0=(\gamma-i\omega_3)(\tfrac{1}{2}\gamma_{21}-i(\omega_3-\Delta\omega))+\tfrac{1}{4}G^2$$

We can see that

$$\bar{\rho}_{11}+\bar{\rho}_{22}+\bar{\rho}_{33}=1$$

$$\therefore\ \bar{\rho}_{22}=1-\bar{\rho}_{11}-\bar{\rho}_{33} \qquad \text{(7.B.117)}$$

Thus we can write two simultaneous equations for and , viz.

$$a_{10}\bar{\rho}_{11}+a_{30}\bar{\rho}_{33}=a_{00} \qquad \text{(7.B.118)}$$

$$b_{10}\bar{\rho}_{11}+b_{30}\bar{\rho}_{33}=b_{00} \qquad \text{(7.B.119)}$$

where

$$a_{10}=-\tfrac{1}{4}\Gamma_{31}^2\tfrac{1}{4}G^2(H_0+H_0^*)-\gamma_{21}|H_0|^2+\tfrac{1}{4}\Gamma^2(F_0H_0^*+F_0^*H_0)$$

$$a_{30}=\tfrac{1}{4}\Gamma_{31}^2\tfrac{1}{4}G^2(H_0+H_0^*)-\gamma_{21}|H_0|^2+\tfrac{1}{4}(\Gamma^2-\Gamma_{31}^2)(F_0H_0^*+F_0^*H_0) \qquad \text{(7.B.120)}$$

$$a_{00}=-\gamma_{21}|H_0|^2+\tfrac{1}{4}\Gamma^2(F_0H_0^*+F_0^*H_0)$$

$$b_{10} = [illegible] + \tfrac{1}{4}G^2\,\tfrac{1}{4}(\Gamma^2 + \Gamma_{21}^2 + \Gamma_{31}^2)(H_0 + H_0^*) + \tfrac{1}{4}G^2(B_0^* H_0 + B_0 H_0^*)$$

$$- \tfrac{1}{4}\Gamma_{21}(\Gamma_{21} + \Gamma_{31})(F_0^* H_0 + F_0 H_0^*)$$

$$b_{20} = \tfrac{1}{4}G^2\,\tfrac{1}{4}(-2\Gamma^2 - \Gamma_{21}^2 + \Gamma_{31}^2)(H_0 + H_0^*) - \tfrac{1}{4}G^2(B_0^* H_0 + B_0 H_0^*) + (\gamma_{21} - \gamma_{31})|H_0|^2 \qquad \text{(7.B.120) cont'd}$$

$$+ \tfrac{1}{4}\Gamma_{31}(\Gamma_{21} - \Gamma_{31})(F_0^* H_0 + F_0 H_0^*)$$

$$b_{30} = [illegible] + \tfrac{1}{4}\Gamma_{31}\,\tfrac{1}{4}G^2(H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21}(\Gamma_{21} + \Gamma_{31})(F_0 H_0^* + F_0^* H_0)$$

Solving (7.B.118) and (7.B.119) we obtain

$$\left.\begin{aligned} \bar{\rho}_{11} &= \frac{A}{X} \\ \bar{\rho}_{33} &= \frac{B}{X} \\ \bar{\rho}_{22} &= 1 - \left(\frac{A+B}{X}\right) \end{aligned}\right\} \qquad (7.B.121)$$

where $X = a_{30}b_{10} - a_{10}b_{30}$, $A = a_{30}b_{00} - a_{00}b_{30}$, $B = a_{00}b_{10} - a_{10}b_{00}$

Now if we substitute $\Delta\omega = 0$, i.e. we assume that the driving field is in exact resonance with the corresponding atomic transition, then

$$X = a_{30}b_{10} - a_{10}b_{30} = |H_0|^2 T_0 \qquad (7.B.122)$$

where

$$\begin{aligned} T_0 &= \left(T(s+i(\omega-\Delta\omega)) = 0\right)_{\Delta\omega=0} \\ &= -\Big[\gamma_{21}\gamma_{31}|H_0|^2 + \tfrac{1}{4}\Gamma_{31}(3\gamma_{31}\Gamma_{21} + \gamma_{31}\Gamma_{31})\tfrac{1}{4}G^2(H_0 + H_0^*) \\ &\quad + 2\gamma_{21}\tfrac{1}{4}G^2(B_0^* H_0 + B_0 H_0^*) - \tfrac{1}{4}\Gamma^2(\gamma_{21} + \gamma_{31})(F_0^* H_0 + F_0 H_0^*) \\ &\quad - \tfrac{1}{4}\Gamma_{31}(2\Gamma_{21} - \Gamma_{31})\tfrac{1}{4}G^2\Big\{\tfrac{1}{2}\Gamma^2 + (F_0^* H_0 + F_0 H_0^*) \\ &\qquad + (\tfrac{1}{2}\gamma_{21} + i\omega_s)H_0 + (\tfrac{1}{2}\gamma_{21} - i\omega_s)H_0^*\Big\}\Big] \end{aligned} \qquad (7.B.123)$$

(N.B. we have used the fact that

$$B_0 F_0 + \tfrac{1}{16}\Gamma^2 G^2 = (\tfrac{1}{2}\gamma_{21} - i\omega_s)H_0 \qquad (7.B.124a)$$

and

$$B_0^* F_0^* + \tfrac{1}{16}\Gamma^2 G^2 = (\tfrac{1}{2}\gamma_{21} + i\omega_s)H_0^* \qquad (7.B.124b)$$

Hence we have

$$\bar{\rho}_{11} = \left[\frac{a_{30}b_{00} - a_{00}b_{30}}{|H_0|^2 T_0}\right] \qquad (7.B.125)$$

$$\bar{\rho}_{22} = \left[\frac{|H_0|^2 T_0 - \{(a_{30} - a_{10})b_{00} - (b_{30} - b_{10})a_{00}\}}{|H_0|^2 T_0}\right] \qquad (7.B.126)$$

$$\bar{\rho}_{21} = \left[\frac{a_{10} b_{10} - a_{10} b_{10}}{|H_0| T_0} \right] \tag{7.B.127}$$

and substituting in equations (7.B.110), (7.B.111) and (7.B.112) we obtain:-

$$\bar{\rho}_{21} = \frac{-1}{H_0 |H_0|^2 T_0} \Big[a_{10} \{ -\tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 b_{30} + \tfrac{1}{2}\Gamma_{21} F_0 (b_{30} - b_{10}) + \tfrac{1}{2}\Gamma_{31} (\Gamma - \tfrac{1}{4} G^2) b_{10} \} + b_{10} \{ \tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 a_{30} + \tfrac{1}{2}\Gamma_{21} F_0 (a_{30} - a_{10}) - \tfrac{1}{2}\Gamma_{31} (\Gamma - \tfrac{1}{4} G^2)^* a_{10} \} + \tfrac{1}{2}\Gamma_{21} F_0 |H_0|^2 T_0 \Big] \tag{7.B.128}$$

$$\bar{\rho}_{31} = \frac{-(i\lambda\mathcal{E}_0)}{H_0 |H_0|^2 T_0} \Big[a_{10} \{ -\tfrac{1}{2}\Gamma_{31} (\gamma - i\omega_3) b_{30} - \tfrac{1}{2}\Gamma_{21} \tfrac{1}{2}\gamma_{31} (b_{30} - b_{10}) - \tfrac{1}{2}\Gamma_{31} (\gamma + \tfrac{1}{2}\gamma_{31} - i\omega_3) b_{10} \} + b_{10} \{ \tfrac{1}{2}\Gamma_{31} (\gamma - i\omega_3) a_{30} + \tfrac{1}{2}\Gamma_{21} \tfrac{1}{2}\gamma_{31} (a_{30} - a_{10}) + \tfrac{1}{2}\Gamma_{31} (\gamma + \tfrac{1}{2}\gamma_{31} - i\omega_3) a_{10} \} - \tfrac{1}{2}\Gamma_{21} \tfrac{1}{2}\gamma_{31} |H_0|^2 T_0 \Big] \tag{7.B.129}$$

$$\bar{\rho}_{31} = \frac{(i\lambda\mathcal{E}_0)}{H_0 |H_0|^2 T_0} \Big[a_{10} \{ -B_0 b_{30} - \tfrac{1}{4}\Gamma_{21}^2 (b_{30} - b_{10}) - (B_0 + \tfrac{1}{4}\Gamma^2) b_{10} \} + b_{10} \{ B_0 a_{30} + \tfrac{1}{4}\Gamma_{21}^2 (a_{30} - a_{10}) + (B_0 + \tfrac{1}{4}\Gamma^2) a_{10} \} - \tfrac{1}{4}\Gamma_{21} |H_0|^2 T_0 \Big] \tag{7.B.130}$$

On substituting for a_{00} and b_{00} we obtain the following equations:-

$$\bar{\rho}_{11} = \left[\frac{[\gamma_{21} |H_0|^2 - \tfrac{1}{4}\Gamma^2 (F_0 H_0^* + F_0^* H_0)] b_{30} + [\gamma_{21} |H_0|^2 + \tfrac{1}{4}\Gamma_{21}^2 \tfrac{1}{4} G^2 (H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21} (\Gamma_{21} + \Gamma_{31})(F_0 H_0^* + F_0^* H_0)] a_{30}}{|H_0|^2 T_0} \right] \tag{7.B.131}$$

$$\bar{\rho}_{22} = \frac{\left[[\gamma_{21} |H_0|^2 - \tfrac{1}{4}\Gamma^2 (F_0 H_0^* + F_0^* H_0)](b_{10} - b_{30}) + [\gamma_{21} |H_0|^2 + \tfrac{1}{4}\Gamma_{21}^2 \tfrac{1}{4} G^2 (H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21} (\Gamma_{21} + \Gamma_{31})(F_0 H_0^* + F_0^* H_0)](a_{10} - a_{30}) + |H_0|^2 T_0 \right]}{|H_0|^2 T_0} \tag{7.B.132}$$

$$\bar{\rho}_{33} = \frac{[\gamma_{21} |H_0|^2 - \tfrac{1}{4}\Gamma^2 (F_0 H_0^* + F_0^* H_0)] b_{10} + [\gamma_{21} |H_0|^2 + \tfrac{1}{4}\Gamma_{21}^2 \tfrac{1}{4} G^2 (H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21} (\Gamma_{21} + \Gamma_{31})(F_0 H_0^* + F_0^* H_0)] a_{30}}{|H_0|^2 T_0} \tag{7.B.133}$$

$$\bar{\rho}_{23} = \frac{-1}{H_0 |H_0|^2 T_0} \Big[[\gamma_{21} |H_0|^2 - \tfrac{1}{4}\Gamma^2 (F_0 H_0^* + F_0^* H_0)][\{ \tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 + \tfrac{1}{2}(\Gamma_{21} - \Gamma_{31}) F_0 \} b_{10} + \{ \tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 - \tfrac{1}{2}\Gamma_{21} F_0 \} b_{30}] + [\gamma_{21} |H_0|^2 + \tfrac{1}{4}\Gamma_{21}^2 \tfrac{1}{4} G^2 (H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21} (\Gamma_{21} + \Gamma_{31})(F_0 H_0^* + F_0^* H_0)][\{ \tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 + \tfrac{1}{2}(\Gamma_{21} - \Gamma_{31}) F_0 \} a_{10} + \{ \tfrac{1}{2}\Gamma_{31} \tfrac{1}{4} G^2 - \tfrac{1}{2}\Gamma_{21} F_0 \} a_{30}] + \tfrac{1}{2}\Gamma_{21} F_0 |H_0|^2 T_0 \Big] \tag{7.B.134}$$

$$\bar{\rho}_{21} = \frac{-(i\lambda\mathcal{E}_0)}{H_0|H_0|^2T_0}\Big[\big[\gamma_{21}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[\{-\tfrac{1}{2}\Gamma_{21}\tfrac{1}{2}\gamma_{31} + \tfrac{1}{2}\Gamma_{31}(\gamma + \tfrac{1}{2}\gamma_{31} - i\omega_3)\}b_{10}$$
$$+\{\tfrac{1}{2}\Gamma_{21}\tfrac{1}{2}\gamma_{31} + \tfrac{1}{2}\Gamma_{31}(\gamma - i\omega_3)\}b_{30}\big] \qquad \text{(7.B.135)}$$
$$+\big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma_{21}\tfrac{1}{4}C^2(H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21}(\Gamma_{21} + \Gamma_{31})(F_0H_0^* + F_0^*H_0)\big]$$
$$\times\big[\{-\tfrac{1}{2}\Gamma_{21}\tfrac{1}{2}\gamma_{31} + \tfrac{1}{2}\Gamma_{31}(\gamma + \tfrac{1}{2}\gamma_{31} - i\omega_3)\}a_{10} + \{\tfrac{1}{2}\Gamma_{31}(\gamma - i\omega_3) + \tfrac{1}{2}\Gamma_{21}\tfrac{1}{2}\gamma_{31}\}a_{30}\big]$$
$$-\tfrac{1}{2}\Gamma_{21}\tfrac{1}{2}\gamma_{31}|H_0|^2T_0\Big]$$

$$\left.\begin{aligned}\bar{\rho}_{31} = \frac{(i\lambda\mathcal{E}_0)}{H_0|H_0|^2T_0}\Big[&\big[\gamma_{21}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[\{B_0 + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\}b_{10} + \{B_0 + \tfrac{1}{4}\Gamma_{21}^2\}b_{30}\big]\\ &+\big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma_{21}\tfrac{1}{4}C^2(H_0 + H_0^*) - \tfrac{1}{4}\Gamma_{21}(\Gamma_{21} + \Gamma_{31})(F_0H_0^* + F_0^*H_0)\big]\\ &\times\big[\{B_0 + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\}a_{10} + \{B_0 + \tfrac{1}{4}\Gamma_{21}^2\}a_{30}\big]\\ &-\tfrac{1}{4}\Gamma_{21}^2|H_0|^2T_0\Big]\end{aligned}\right\} \qquad \text{(7.B.136)}$$

We may now note that:-

$$\bar{\rho}_{31} = \underset{s+i\omega\to 0}{Lt.}\left[(s+i\omega)\hat{\psi}_{\alpha 11}(s)\right] = \underset{s+i\omega\to 0}{Lt.}\left[(s+i\omega)\,\hat{U}_{\alpha 11}(s,t')\,e^{i\omega t'}\right] \qquad \text{(7.B.137)}$$

$$\bar{\rho}_{21} = \underset{s+i\omega\to 0}{Lt.}\left[(s+i\omega)\hat{\psi}_{\beta 11}(s)\right] = \underset{s+i\omega\to 0}{Lt.}\left[(s+i\omega)\,\hat{U}_{\beta 11}(s,t')\,e^{i\omega t'}\right] \qquad \text{(7.B.138)}$$

Now, we recall that according to equation (7.B.98b)

$$\bar{g}(\nu) = 2Re\Big[\hat{\psi}_{\alpha 11}(-\nu)\big[\bar{\rho}_{13} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{12}\big] + \hat{\psi}_{\alpha 21}(-\nu)\big[\bar{\rho}_{23} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{22}\big] + \hat{\psi}_{\alpha 31}(-\nu)\big[\bar{\rho}_{33} + \tfrac{\omega}{\omega_2}\tfrac{\Gamma_{21}}{\gamma_{31}}\bar{\rho}_{32}\big]$$
$$+\tfrac{\omega_2}{\omega}\tfrac{\gamma_{21}}{\gamma_{31}}\Big\{\hat{\psi}_{\beta 11}(-\nu)\big[\bar{\rho}_{12} + \tfrac{\omega^2}{\omega_2^2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{13}\big] + \hat{\psi}_{\beta 21}(-\nu)\big[\bar{\rho}_{22} + \tfrac{\omega^2}{\omega_2^2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{23}\big]$$
$$+\hat{\psi}_{\beta 31}(-\nu)\big[\bar{\rho}_{32} + \tfrac{\omega^2}{\omega_2^2}\tfrac{\Gamma_{21}}{\gamma_{21}}\bar{\rho}_{33}\big]\Big\}\Big]$$

so that all the quantities on the RHS are now known. If we now evaluate all the $\hat{\psi}$'s given in equations (7.B.59b) and (7.B.60b) at $(-i\nu)$, and substitute $\Omega = \nu - \omega$ in the final expressions, we obtain, for $\Delta\omega = 0$:-

$$\hat{\psi}_{\alpha 11}(-i\nu) = \frac{1}{-i\Omega}\,\frac{i\lambda\mathcal{E}_0}{H_1'Z'}\Big[\big[(\gamma_{21} - i\Omega)H_1'H_2' + \tfrac{1}{4}\Gamma_{21}^2\tfrac{1}{4}C^2(H_1' + H_2') - \tfrac{1}{4}(\Gamma^2 + \Gamma_{21}^2)(F_2'H_1' + F_1'H_2')\big]$$
$$\times\big[\{B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\}a_1' + \{B_1' + \tfrac{1}{4}\Gamma_{21}^2\}a_3'\big]$$
$$+\big[(\gamma_{21} - i\Omega)H_1'H_2' - \tfrac{1}{4}\Gamma^2(F_2'H_1' + F_1'H_2')\big] \qquad \text{(7.B.139a)}$$
$$\times\big[\{B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\}b_1' + \{B_1' + \tfrac{1}{4}\Gamma_{21}^2\}b_3'\big]$$
$$-\tfrac{1}{4}\Gamma_{21}^2Z'\Big]$$

$$\hat{\psi}_{\rho 21}(-i\nu) = \frac{1}{H_1' Z'}\Big[\tfrac{1}{4}G^2 H_2' \Big\{ \big[\tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}(\Gamma_{31}+\Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) \big]$$
$$\times \big[\{ B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\} a_1' + \{ B_1' + \tfrac{1}{4}\Gamma_{21}^2 \} a_3' \big]$$
$$+ \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega)\big[\{ B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\} b_1' + \{ B_1' + \tfrac{1}{4}\Gamma_{21}^2 \} b_3' \big] \Big\}$$
$$- \tfrac{1}{2}\Gamma_{21}(\gamma - i(\Omega+\omega_3)) Z' \Big] \tag{7.B.139b}$$

$$\hat{\psi}_{\rho 31}(-i\nu) = \frac{1}{H_1' Z'}\Big[-\tfrac{1}{4}G^2 H_2' \Big\{ \big[B_1' + \tfrac{1}{4}(\Gamma_{31}^2 + \Gamma^2) \big]\big[\{ B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\} a_1' + \{ B_1' + \tfrac{1}{4}\Gamma_{21}^2 \} a_3' \big]$$
$$+ \tfrac{1}{4}\Gamma_{31}^2 \big[\{ B_1' + \tfrac{1}{4}(\Gamma^2 - \Gamma_{21}^2)\} b_1' + \{ B_1' + \tfrac{1}{4}\Gamma_{21}^2 \} b_3' \big] \Big\}$$
$$+ B_1' Z' \Big] \tag{7.B.139c}$$

$$\hat{\psi}_{\beta 11}(-i\nu) = \frac{1}{-i\Omega}\,\frac{(-i\lambda\Gamma_0)}{H_1' Z'}\Big[\big[(\gamma_{21} - i\Omega) H_1 H_2' + \tfrac{1}{4}\Gamma_{21}\tfrac{1}{4}G^2 (H_1 + H_2) - \tfrac{1}{4}(\Gamma^2 + \Gamma_{21}^2)(F_2' H_1 + F_1' H_2') \big]$$
$$\times \big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) \} a_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} a_3' \big]$$
$$+ \big[(\gamma_{21} - i\Omega) H_1' H_2' - \tfrac{1}{4}\Gamma^2 (F_2' H_1' + F_1' H_2') \big]$$
$$\times \big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{21}(\gamma - i(\Omega+\omega_3)) \} b_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} b_3' \big]$$
$$- \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) Z' \Big] \tag{7.B.140a}$$

$$\hat{\psi}_{\beta 21}(-i\nu) = \frac{1}{H_1 Z'}\Big[-\tfrac{1}{4}G^2 H_2' \Big\{ \big[\tfrac{1}{2}(\Gamma_{31} + \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{21}(\gamma - i(\Omega+\omega_3)) \big]$$
$$\times \big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) \} a_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} a_3' \big]$$
$$+ \big[\tfrac{1}{2}\Gamma_{31}(\tfrac{1}{2}\gamma_{31} - i\Omega) \big]\big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) \} b_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} b_3' \big] \Big\}$$
$$+ (\tfrac{1}{2}\gamma_{31} - i\Omega)(\gamma - i(\Omega+\omega_3)) Z' \Big] \tag{7.B.140b}$$

$$\hat{\psi}_{\beta 31}(-i\nu) = \frac{1}{H_1' Z'}\Big[\tfrac{1}{4}G^2 \Big\{ \big[H_2' \{ B_1' + \tfrac{1}{4}(\Gamma_{31}^2 + \Gamma_{21}^2) \} \big]\big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) \} a_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} a_3' \big]$$
$$+ \big[\tfrac{1}{4}\Gamma_{31}^2 H_2' \big]\big[\{ \tfrac{1}{2}(\Gamma_{31} - \Gamma_{21})(\tfrac{1}{2}\gamma_{31} - i\Omega) + \tfrac{1}{2}\Gamma_{21}(\gamma - i(\Omega+\omega_3)) \} b_1'$$
$$+ \{ \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) + \tfrac{1}{2}\Gamma_{21}(\tfrac{1}{2}\gamma_{31} - i\Omega) \} b_3' \big]$$
$$- \tfrac{1}{2}\Gamma_{31}(\gamma - i(\Omega+\omega_3)) Z' \Big] \tag{7.B.140c}$$

where

$$\left.\begin{aligned}
H_1' &= (\gamma - i(\Omega+\omega_3))F_1' + \tfrac{1}{4}G^2(\tfrac{1}{2}\gamma_{31} - i\Omega) &&= H_1(s = -i\nu) \\
F_1' &= (\tfrac{1}{2}\gamma_{21} - i(\Omega+\omega_3))(\tfrac{1}{2}\gamma_{31} - i\Omega) - \tfrac{1}{4}\Gamma^2 &&= F_1(s = -i\nu) \\
B_1' &= (\gamma - i(\Omega+\omega_3))(\tfrac{1}{2}\gamma_{21} - i(\Omega+\omega_3)) + \tfrac{1}{4}G^2 &&= B_1(s = -i\nu) \\
H_2' &= (\gamma - i(\Omega-\omega_3))F_2' + \tfrac{1}{4}G^2(\tfrac{1}{2}\gamma_{31} - i\Omega) &&= H_2(s = -i\nu)
\end{aligned}\right\} \tag{7.B.141}$$

$$F_1' = (\tfrac{1}{2}\gamma_{31} - i(\Omega + \omega_3))(\tfrac{1}{2}\gamma_{21} - i\Omega)\ [\text{illegible}] \qquad F_2(s = -i\nu)$$

$$B_1' = (\gamma\ [\text{illegible}]\ \omega_3))(\ [\text{illegible}]\ (\Omega + \omega_3)) - \tfrac{1}{4}G^2 \qquad B_2(s = -i\nu)$$

$$[\text{illegible}] = -(\gamma_{21} - i\Omega)H_1'H_2' - \tfrac{1}{4}\Gamma_{31}^2\,\tfrac{1}{4}G^2(H_1' + H_2') + \tfrac{1}{4}\Gamma^2(F_2'H_1' + F_1'H_2')\ [\text{illegible}]$$

$$[\text{illegible}] = -(\gamma_{21} - i\Omega)H_1'H_2' + \tfrac{1}{4}\Gamma_{31}^2\,\tfrac{1}{4}G^2(H_1' + H_2') + \tfrac{1}{4}(\Gamma^2 - \Gamma_{31}^2)(F_2'H_1' + F_1'H_2')\,[\text{illegible}] \qquad \text{(7.B.141) cont'd}$$

$$[\text{illegible}] = (\gamma_{21} - i\Omega)H_1'H_2' + \tfrac{1}{4}(\ [\text{illegible}]\ + \Gamma^2)\tfrac{1}{4}G^2(H_1' + H_2') + \tfrac{1}{4}G^2(B_2'H_1' + B_1'H_2')$$
$$- \tfrac{1}{4}(\Gamma^2 + \Gamma_{31}^2)(F_2'H_1' + F_1'H_2')\ [\text{illegible}]$$

$$[\text{illegible}] = [\text{illegible}]\ H_1'H_2'\ \ \tfrac{1}{4}(\Gamma_{31}^2 - [\text{illegible}]\ 2\Gamma^2)\tfrac{1}{4}G^2(H_1' + H_2') - \tfrac{1}{4}G^2(B_2'H_1' + B_1'H_2')$$
$$- \tfrac{1}{4}(\Gamma_{31}^2 - \Gamma_{21}^2)(F_2'H_1' + F_1'H_2')\ [\text{illegible}]$$

$$Z' = [\text{illegible}]\ - H_1H_1'\,T(-i\nu) = H_1H_1'T' - Z(s = -i\nu)$$

i.e. primed quantities are evaluated at $s = -i\nu$,

and $$T' = T(-i\nu) = -\Big[H_1'H_2'(\gamma_{21} - i\Omega)(\gamma_{21} - i\Omega) + (H_1' + H_2')\tfrac{1}{4}G^2\{\tfrac{1}{4}\Gamma^2(\gamma_{21} - i\Omega)$$
$$+ \tfrac{1}{4}\Gamma_{31}^2(\gamma_{31} - i\Omega)\}$$
$$+ 2(B_2'H_1' + B_1'H_2')(\gamma_{21} - i\Omega)\tfrac{1}{4}G^2 - (\tfrac{1}{4}G^2)^2\tfrac{1}{8}\Gamma_{31}^2(2\Gamma_{21}^2 - \Gamma^2) \qquad \text{(7.B.142a)}$$
$$- \tfrac{1}{4}G^2(F_1'B_2' + F_2'B_1')\tfrac{1}{4}(2\Gamma^2 - \Gamma_{31}^2)$$
$$- (F_2'H_1' + F_1'H_2')\tfrac{1}{2}\Gamma^2(\gamma - i\Omega)$$
$$- \tfrac{1}{4}G^2\tfrac{1}{4}(2\Gamma^2 - \Gamma_{31}^2)\{H_1'(\tfrac{1}{2}\gamma_{21} - i(\Omega - \omega_3)) + H_2'(\tfrac{1}{2}\gamma_{21} - i(\Omega + \omega_3))\}\Big]$$

or

$$= -\Big[(\gamma_{21} - i\Omega)^2H_1'H_2' + \Gamma^2\tfrac{1}{4}G^2(\gamma_{21} - i\Omega)(H_1' + H_2') + 2\tfrac{1}{4}G^2(\gamma_{21} - i\Omega)(B_2'H_1' + B_1'H_2')$$
$$- \tfrac{1}{2}\Gamma^2(\gamma_{21} - i\Omega)(F_2'H_1' + F_1'H_2')$$
$$- \tfrac{1}{8}\Gamma^4(\tfrac{1}{4}G^2)^2 \qquad \text{(7.B.142b)}$$
$$- \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(B_1'F_2' + B_2'F_1')$$
$$- \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2\{(\tfrac{1}{2}\gamma_{21} - i(\Omega - \omega_3))H_1' + (\tfrac{1}{2}\gamma_{21} - i(\Omega + \omega_3))H_2'\}$$
$$- (\gamma_{21} - \gamma_{31})\{\tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_1' + H_2') + (\gamma_{21} - i\Omega)H_1'H_2' - \tfrac{1}{4}\Gamma^2(H_1'F_2' + H_2'F_1')\}\Big]$$

b) Spectral correlation function split into coherent and incoherent parts

We are now interested in separating the expression for the total spectral correlation function, $\hat{g}(\nu)$, into coherent and incoherent parts, as in Mollow's paper[9].

In order to find the coherent part it is necessary to find the asymptotic form of $g(\tau)$ in the limit $\tau \to \infty$, which originates from the poles of on the imaginary axis of the s plane. $\hat{g}(s)$ is, of course, the Laplace transform of $g(\tau)$, so that, since $g(\tau)$ is given by equation (7.B.97) as

$$\begin{aligned} g(\tau) = {}& \psi_{x11}(\tau)\left[\bar\rho_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{12}\right] + \psi_{x21}(\tau)\left[\bar\rho_{23} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{22}\right] + \psi_{x31}(\tau)\left[\bar\rho_{33} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{32}\right] \\ & + \omega/\omega\,\gamma_{21}/\gamma_{31}\Big\{\psi_{\rho11}(\tau)\left[\bar\rho_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{13}\right] + \psi_{\rho21}(\tau)\left[\bar\rho_{22} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{23}\right] \\ & \qquad + \psi_{\rho31}(\tau)\left[\bar\rho_{32} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{33}\right]\Big\} \end{aligned}$$

$$\begin{aligned} \hat g(s) = {}& \int \ldots g(\tau) \\ = {}& \hat\psi_{x11}(s)\left[\bar\rho_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{12}\right] + \hat\psi_{x21}(s)\left[\bar\rho_{23} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{22}\right] + \hat\psi_{x31}(s)\left[\bar\rho_{33} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{32}\right] \\ & + \omega/\omega\,\gamma_{21}/\gamma_{31}\Big\{\hat\psi_{\rho11}(s)\left[\bar\rho_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{13}\right] + \hat\psi_{\rho21}(s)\left[\bar\rho_{22} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{23}\right] \\ & \qquad + \hat\psi_{\rho31}(s)\left[\bar\rho_{32} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{33}\right]\Big\} \end{aligned} \tag{7.B.143}$$

where the expressions for the $\hat\psi(s)$'s are given by equations (7.B.59b) and (7.B.60b) where $\Delta\omega$ now equals zero. If we assume that the real parts of the roots of T are all negative then the only contribution to $g(\tau\to\infty)$ comes from the pole at $s=-i\omega$ on the RHS of the expressions for $\hat\psi_{x11}(s)$ and $\hat\psi_{\rho11}(s)$. The residue of the pole is, by virtue of equations (7.B.59b), (7.B.60b) and (7.B.143),:-

$$\begin{aligned} \lim_{s\to -i\omega}\left[(s+i\omega)\,\hat g(s)\right] = {}& \lim_{s\to -i\omega}\Big[(s+i\omega)\Big\{\hat\psi_{x11}(s)\left[\bar\rho_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{12}\right] \\ & \qquad + \omega/\omega\,\gamma_{21}/\gamma_{31}\,\hat\psi_{\rho11}(s)\left[\bar\rho_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{13}\right]\Big\}\Big] \\ = {}& \lim_{s\to -i\omega}\Big[\frac{i\lambda\mathcal{E}_0}{H_1 Z}\Big\{\left[(s+\gamma_{21}+i\omega)H_1H_2 + 1/4\,\Gamma_{21}^2\,1/4\,G^2(H_1+H_2) - 1/4(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\right] \\ & \times\left[\{B_1 + 1/4(\Gamma^2-\Gamma_{21}^2)\}a_1 + \{B_1 + 1/4\,\Gamma_{21}^2\}a_3\right] \\ & + \left[(s+\gamma_{21}+i\omega)H_1H_2 - 1/4\,\Gamma^2(F_2H_1+F_1H_2)\right] \\ & \times\left[\{B_1 + 1/4(\Gamma^2-\Gamma_{21}^2)\}b_1 + \{B_1 + 1/4\,\Gamma_{21}^2\}b_3\right] \\ & - 1/4\,\Gamma_{21}^2\,Z\Big\}\left\{\bar\rho_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{12}\right\} \\ & + \omega/\omega\,\gamma_{21}/\gamma_{31}\,\frac{(-i\lambda\mathcal{E}_0)}{H_1 Z}\Big\{\big[(s+\gamma_{21}+i\omega)H_1H_2 + 1/4\,\Gamma_{21}^2\,1/4\,G^2(H_1+H_2) \\ & \qquad - 1/4(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\big] \\ & \times\big[\{1/2(\Gamma_{31}-\Gamma_{21})(s+1/2\gamma_{21}+i\omega) + 1/2\,\Gamma_{31}(s+\gamma+i\omega_2)\}a_1 \\ & \qquad + \{1/2\,\Gamma_{31}(s+\gamma+i\omega_2) + 1/2\,\Gamma_{21}(s+1/2\gamma_{31}+i\omega)\}a_3\big] \\ & + \left[(s+\gamma_{21}+i\omega)H_1H_2 - 1/4\,\Gamma^2(F_2H_1+F_1H_2)\right] \\ & \times\big[\{1/2(\Gamma_{31}-\Gamma_{21})(s+1/2\gamma_{21}+i\omega) + 1/2\,\Gamma_{31}(s+\gamma+i\omega_2)\}b_1 \\ & \qquad + \{1/2\,\Gamma_{31}(s+\gamma+i\omega_2) + 1/2\,\Gamma_{21}(s+1/2\gamma_{31}+i\omega)\}b_3\big] \\ & - 1/2\,\Gamma_{21}(s+1/2\gamma_{21}+i\omega)Z\Big\}\left\{\bar\rho_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar\rho_{13}\right\}\Big] \end{aligned}$$

i.e.

$$\begin{aligned} \lim_{s\to -i\omega}\left[(s+i\omega)\,\hat g(s)\right] = {}& \frac{i\lambda\mathcal{E}_0}{H_1'' Z''}\Big[\Big\{\left[\gamma_{21}H_1''H_2'' + 1/4\,\Gamma_{21}^2\,1/4\,G^2(H_1''+H_2'') - 1/4(\Gamma^2+\Gamma_{21}^2)(F_2''H_1''+F_1''H_2'')\right] \\ & \times\left[\{B_1'' + 1/4(\Gamma^2-\Gamma_{21}^2)\}a_1'' + \{B_1'' + 1/4\,\Gamma_{21}^2\}a_3''\right] \\ & + \left[\gamma_{21}H_1''H_2'' - 1/4\,\Gamma^2(F_2''H_1''+F_1''H_2'')\right] \\ & \times\left[\{B_1'' + 1/4(\Gamma^2-\Gamma_{21}^2)\}b_1'' + \{B_1'' + 1/4\,\Gamma_{21}^2\}b_3''\right] \\ & - 1/4\,\Gamma_{21}^2\,Z''\Big\}\left\{\bar\rho_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar\rho_{12}\right\} \end{aligned} \tag{7.B.144}$$

$$- [illegible] \Big\{ \big[[illegible] (H_1'' + H_2'') - \tfrac14 [illegible] (F_2'' H_1'' + F_1'' H_2'') \big] \tag{7.B.144 cont'd}$$
$$[illegible] (\gamma - i\omega_3) [illegible] + \tfrac12 \Gamma_{21} \tfrac12 \gamma_{31} \} a_5'' \big]$$
$$- \big[[illegible] - \tfrac14 \Gamma^2 (F_2'' H_1'' + F_1'' H_2'') \big]$$
$$[illegible] (\gamma - i\omega_3) \} b_1''$$
$$[illegible] (\gamma - i\omega_3) + \tfrac12 \Gamma_{21} \tfrac12 \gamma_{31} \} b_3'' \big]$$
$$- [illegible] \beta_{12} + [illegible] \beta_{13} \} \Big]$$

where

$$
\begin{aligned}
H_1'' &= H_1(s = -i\omega) = H_1'(\Omega = 0) = (\gamma - i\omega_3) F_1'' + \tfrac14 G^2 \tfrac12 \gamma_{31} &&= H_0 \\
F_1'' &= F_1(s = -i\omega) = F_1'(\Omega = 0) = (\tfrac12 \gamma_{21} - i\omega_3) \tfrac12 \gamma_{31} - \tfrac14 \Gamma^2 &&= F_0 \\
B_1'' &= B_1(s = -i\omega) = B_1'(\Omega = 0) = (\gamma - i\omega_3)(\tfrac12 \gamma_{21} - i\omega_3) + \tfrac14 G^2 &&= B_0 \\
H_2'' &= H_2(s = -i\omega) = H_2'(\Omega = 0) = (\gamma + i\omega_3) F_2'' + \tfrac14 G^2 \tfrac12 \gamma_{31} &&= H_0^* \\
F_2'' &= F_2(s = -i\omega) = F_2'(\Omega = 0) = (\tfrac12 \gamma_{21} + i\omega_3) \tfrac12 \gamma_{31} - \tfrac14 \Gamma^2 &&= F_0^* \\
B_2'' &= B_2(s = -i\omega) = B_2'(\Omega = 0) = (\gamma + i\omega_3)(\tfrac12 \gamma_{21} + i\omega_3) + \tfrac14 G^2 &&= B_0^*
\end{aligned}
\tag{7.B.145a}
$$

from equations (7.B.42) and (7.B.47) at $\Delta\omega = 0$, (7.B.141) and (7.B.116),

and $Z'' = Z(s = -i\omega) = Z'(\Omega = 0) = H_1'' H_2'' T'' = H_2 H_1^{*} T_0$

where
$$
\begin{aligned}
T'' = T(s = -i\omega) = T'(\Omega = 0) = -\Big[& H_1'' H_2'' \gamma_{31} \gamma_{21} + (H_1'' + H_2'') [illegible] \\
& + 2 (B_2'' H_1'' + B_1'' H_2'') \gamma_{21} \tfrac14 G^2 \\
& - (\tfrac14 G^2)^2 \tfrac18 \Gamma_{31}^2 (2\Gamma_{21}^2 - \Gamma^2) \\
& - \tfrac14 G^2 (F_1'' B_2'' + F_2'' B_1'') \tfrac14 (2\Gamma^2 - \Gamma_{31}^2) \\
& - (F_2'' H_1'' + F_1'' H_2'') \tfrac12 \Gamma^2 \gamma \\
& - \tfrac14 (2\Gamma^2 - \Gamma_{31}^2) \tfrac14 G^2 \{ H_1'' (\tfrac12 \gamma_{21} + i\omega_3) \\
& \quad + H_2'' (\tfrac12 \gamma_{21} - i\omega_3) \} \Big] = T_0
\end{aligned}
\tag{7.B.145b}
$$

from equations (7.B.56) at $\Delta\omega = 0$, (7.B.142a) and (7.B.123).

Also

$$
\begin{aligned}
a_1'' &= a_1(s = -i\omega) = a_1'(\Omega = 0) = -\gamma_{21} H_1'' H_2'' - \tfrac14 \Gamma_{31}^2 \tfrac14 G^2 (H_1'' + H_2'') + \tfrac14 \Gamma^2 (F_2'' H_1'' + F_1'' H_2'') \\
&= a_{10} \\
a_5'' &= a_5(s = -i\omega) = a_5'(\Omega = 0) = -\gamma_{21} H_1'' H_2'' + \tfrac14 \Gamma_{31}^2 \tfrac14 G^2 (H_1'' + H_2'') + \tfrac14 (\Gamma^2 - \Gamma_{31}^2)(F_2'' H_1'' + F_1'' H_2'') \\
&= a_{50} \\
b_1'' &= b_1(s = -i\omega) = b_1'(\Omega = 0) = \gamma_{21} H_1'' H_2'' + \tfrac14 (\Gamma_{31}^2 + \Gamma_{21}^2 + \Gamma^2) \tfrac14 G^2 (H_1'' + H_2'') \\
&\quad + \tfrac14 G^2 (B_2'' H_1'' + B_1'' H_2'') - \tfrac14 (\Gamma^2 + \Gamma_{21}^2)(F_2'' H_1'' + F_1'' H_2'') \\
&= b_{10} \\
b_3'' &= b_3(s = -i\omega) = b_3'(\Omega = 0) = (\gamma_{21} - \gamma_{31}) H_1'' H_2'' + \tfrac14 (\Gamma_{21}^2 - \Gamma_{31}^2 - 2\Gamma^2) \tfrac14 G^2 (H_1'' + H_2'') \\
&\quad - \tfrac14 G^2 (B_2'' H_1'' + B_1'' H_2'') - \tfrac14 (\Gamma_{21}^2 - \Gamma_{31}^2)(F_2'' H_1'' + F_1'' H_2'') \\
&= b_{30}
\end{aligned}
\tag{7.B.145c}
$$

i.e. doubly primed quantities are equal to the corresponding unprimed quantities at $s = -i\omega$ or the primed quantities at $\eta = 0$ BUT are only equal to the corresponding quantities with zero subscripts when $\Delta\omega = 0$, so that

$$\lim_{s\to -i\omega}\left[(s+i\omega)\hat{g}(s)\right] = \bar{\rho}_{31}\left(\bar{\rho}_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar{\rho}_{12}\right) + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,\bar{\rho}_{21}\left(\bar{\rho}_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar{\rho}_{13}\right)$$
$$= |\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right) \quad (7.B.146)$$

We can now see that the contributicn of this term to the correlation function $g(\tau)$ is the coherent, harmonically varying expression

$$g_{coh}(\tau) = \left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\}e^{-i\omega\tau}$$
$$\text{as } g_{coh}(\tau) = \frac{1}{2\pi i}\oint ds\, \hat{g}_{coh}(s)\,e^{s\tau} \quad (7.B.147)$$

N.B. we can also write this as

$$g_{coh}(\tau) = \left\{\rho_{13_\infty}^*(t')\rho_{13_\infty}(t'+\tau) + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,\rho_{12_\infty}^*(t')\rho_{12_\infty}(t'+\tau)\right.$$
$$\left. + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\rho_{13_\infty}^*(t')\rho_{12_\infty}(t'+\tau) + \rho_{12_\infty}^*(t')\rho_{13_\infty}(t'+\tau)\right)\right\} \quad (7.B.148)$$

The Laplace transform of the remaining, incoherent part of the atomic correlation function is

$$\hat{g}_{inch}(s) = \hat{g}_{TOT}(s) - \frac{1}{(s+i\omega)}\left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\} \quad (7.B.149)$$

$\therefore \hat{g}_{TOT}(\nu) = \int_{-\infty}^{\infty} d\tau\, e^{i\nu\tau} g(\tau)$, the Fourier transform of the correlation function $g(\tau)$

$$= 2\pi\delta(\nu)\left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\} + \hat{g}_{inch}(\nu) \quad (7.B.150)$$

$$\boxed{\hat{g}_{TOT}(\nu) = 2\pi\delta(\nu)\left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\} + 2\,\mathrm{Re}\left[\hat{g}_{inch}(-i\nu)\right]}$$

since $g(\nu) = 2\,\mathrm{Re}\,\hat{g}(-i\nu)$ as shown earlier.

We can write $\hat{g}_{inch}(s)$ from (7.B.149) more explicitly as

$$\hat{g}_{inch}(s) = \hat{\psi}_{x11}(s)\left(\bar{\rho}_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar{\rho}_{12}\right) + \hat{\psi}_{x21}(s)\left(\bar{\rho}_{23} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar{\rho}_{22}\right)$$
$$+ \hat{\psi}_{x31}(s)\left(\bar{\rho}_{33} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\,\bar{\rho}_{32}\right)$$
$$+ \omega_2/\omega\,\gamma_{21}/\gamma_{31}\left\{\hat{\psi}_{\beta 11}(s)\left(\bar{\rho}_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar{\rho}_{13}\right) + \hat{\psi}_{\beta 21}(s)\left(\bar{\rho}_{22} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar{\rho}_{23}\right)\right.$$
$$\left. + \hat{\psi}_{\beta 31}(s)\left(\bar{\rho}_{32} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{21}\,\bar{\rho}_{33}\right)\right\}$$
$$- \frac{1}{(s+i\omega)}\left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}\,|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\} \quad (7.B.151)$$

We shall now let

$$\hat{\psi}_{\alpha 11}(s) = \frac{1}{(s+i\omega)T}\,\hat{\psi}_{\alpha 11N}(s)$$

where

$$\hat{\psi}_{\alpha 11N}(s) = \frac{i\lambda\mathcal{E}_0}{H_1 H_2}\Big[\Big[(s+\gamma_{21}+i\omega)H_1H_2 + \tfrac14\Gamma_{21}^2\,\tfrac14 G^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big]$$
$$\times\Big[\{B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2)\}a_1 + \{B_1+\tfrac14\Gamma_{21}^2\}a_3\Big]$$
$$+\Big[(s+\gamma_{21}+i\omega)H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big]$$
$$\times\Big[\{B_1+\tfrac14(\Gamma^2-\Gamma_{21}^2)\}b_1 + \{B_1+\tfrac14\Gamma_{21}^2\}b_3\Big]$$
$$-\tfrac14\Gamma_{21}^2 H_1H_2T\Big] \qquad \text{(7.B.152)}$$

and

$$\hat{\psi}_{\beta 11}(s) = \frac{1}{(s+i\omega)T}\,\hat{\psi}_{\beta 11N}(s)$$

where

$$\hat{\psi}_{\beta 11N}(s) = \frac{(-i\lambda\mathcal{E}_0)}{H_1^2 H_2}\Big[\Big[(s+\gamma_{21}+i\omega)H_1H_2 + \tfrac14\Gamma_{21}^2\,\tfrac14 G^2(H_1+H_2) - \tfrac14(\Gamma^2+\Gamma_{21}^2)(F_2H_1+F_1H_2)\Big]$$
$$\times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i\omega_2)\}a_1$$
$$+\{\tfrac12\Gamma_{31}(s+\gamma+i\omega_2) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}a_3\Big]$$
$$+\Big[(s+\gamma_{21}+i\omega)H_1H_2 - \tfrac14\Gamma^2(F_2H_1+F_1H_2)\Big]$$
$$\times\Big[\{\tfrac12(\Gamma_{31}-\Gamma_{21})(s+\tfrac12\gamma_{31}+i\omega) + \tfrac12\Gamma_{31}(s+\gamma+i\omega_2)\}b_1$$
$$+\{\tfrac12\Gamma_{31}(s+\gamma+i\omega_2) + \tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)\}b_3\Big]$$
$$-\tfrac12\Gamma_{21}(s+\tfrac12\gamma_{31}+i\omega)H_1H_2T\Big] \qquad \text{(7.B.153)}$$

Now let us separate $\hat{\psi}_{\alpha 11N}(s)$, given by equation (7.B.152) into two terms, so that

$$\hat{\psi}_{\alpha 11N}(s) = (s+i\omega)T\hat{\psi}_{\alpha 11}(s) = (s+i\omega)\,\hat{\psi}_{\alpha 11N}(\text{rest}) + T_0\bar{\rho}_{13}^{\,*} \qquad \text{(7.B.154a)}$$

Similarly

$$\hat{\psi}_{\beta 11N}(s) = (s+i\omega)T\hat{\psi}_{\beta 11}(s) = (s+i\omega)\,\hat{\psi}_{\beta 11N}(\text{rest}) + T_0\bar{\rho}_{12}^{\,*} \qquad \text{(7.B.155a)}$$

Then

$$\hat{\psi}_{\alpha 11}(s) = \frac{\hat{\psi}_{\alpha 11N}(\text{rest.})}{T} + \frac{T_0}{(s+i\omega)T}\bar{\rho}_{13}^{\,*} \qquad \text{(7.B.154b)}$$

and

$$\hat{\psi}_{\beta 11}(s) = \frac{\hat{\psi}_{\beta 11N}(\text{rest.})}{T} + \frac{T_0}{(s+i\omega)T}\bar{\rho}_{12}^{\,*} \qquad \text{(7.B.155b)}$$

(N.B. We already know that

$$\lim_{s\to -i\omega}\hat{\psi}_{\alpha 11N}(s) = \bar{\rho}_{31}T_0 = \bar{\rho}_{13}^{\,*}T_0 \qquad \text{(see eq. (7.B.137))}$$

and

$$\lim_{s\to -i\omega}\hat{\psi}_{\beta 11N}(s) = \bar{\rho}_{21}T_0 = \bar{\rho}_{12}^{\,*}T_0 \qquad \text{(see eq. (7.B.138))}$$

so that equations (7.B.154a) and (7.B.155a) are valid.

Similarly, we can also separate T, given by equation (7.B.57) with now equal to zero, so that

$$T = ([illegible]) T([illegible]) + T_0 \qquad (7.B.156)$$

Using these substitutions in equation (7.B.150)

$$\begin{aligned}\hat{g}[illegible] = {} & \frac{\hat{\psi}[illegible]}{T}\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}_{12}\right) + \hat{\psi}[illegible]\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}_{22}\right) \\ & + \hat{\psi}[illegible]\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}_{32}\right) \\ & + \frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left\{\frac{[illegible]}{T}\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}_{13}\right) + \hat{\psi}[illegible]\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}[illegible]\right) \right. \\ & \left. + \hat{\psi}[illegible]\left(\bar{\rho}[illegible] + [illegible]/\gamma_{31}\bar{\rho}[illegible]\right)\right\} \\ & - \frac{T([illegible])}{T}\left\{|\bar{\rho}_{13}|^2 + [illegible]/\gamma_{31}|\bar{\rho}_{12}|^2 + [illegible]/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\}\end{aligned} \qquad (7.B.157)$$

Making three further substitutions

$$\left.\begin{aligned} V_{\alpha 11}(\text{rest}) &= \frac{\hat{\psi}_{\alpha 11}(\text{rest})}{T} \\ V_{\beta 11}(\text{rest}) &= \frac{\hat{\psi}_{\beta 11}(\text{rest})}{T} \\ J(\text{rest}) &= \frac{T(\text{rest})}{T}\end{aligned}\right\} \qquad (7.B.158)$$

we obtain:-

$$\begin{aligned}\hat{g}[illegible] = {} & V_{\alpha 11}(\text{rest})\left(\bar{\rho}_{13} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{12}\right) + \hat{\psi}_{\alpha 21}(s)\left(\bar{\rho}_{23} + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{22}\right) \\ & + \hat{\psi}_{\alpha 31}(s)\left(\bar{\rho}[illegible] + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{32}\right) \\ & + \frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left\{V_{\beta 11}(\text{rest})\left(\bar{\rho}_{12} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{13}\right) + \hat{\psi}_{\beta 21}(s)\left(\bar{\rho}_{22} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{23}\right)\right. \\ & \left. + \hat{\psi}_{\beta 31}(s)\left(\bar{\rho}_{32} + \omega^2/\omega_2^2\,\Gamma_{21}/\gamma_{31}\bar{\rho}_{33}\right)\right\} \\ & - J(\text{rest})\left\{|\bar{\rho}_{13}|^2 + \omega_2/\omega\,\gamma_{21}/\gamma_{31}|\bar{\rho}_{12}|^2 + \omega/\omega_2\,\Gamma_{21}/\gamma_{31}\left(\bar{\rho}_{13}^*\bar{\rho}_{12} + \bar{\rho}_{12}^*\bar{\rho}_{13}\right)\right\}\end{aligned}$$

i.e.

$$\left.\begin{aligned}\hat{g}_{\text{mech}}(s) = {} & \left[V_{\alpha 11}(\text{rest}) - J(\text{rest})\bar{\rho}_{13}^*\right]\bar{\rho}_{13} + \hat{\psi}_{\alpha 21}(s)\bar{\rho}_{23} + \hat{\psi}_{\alpha 31}(s)\bar{\rho}_{33} \\ & + \frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left\{\left[V_{\beta 11}(\text{rest}) - J(\text{rest})\bar{\rho}_{12}^*\right]\bar{\rho}_{12} + \hat{\psi}_{\beta 21}(s)\bar{\rho}_{22} + \hat{\psi}_{\beta 31}(s)\bar{\rho}_{32}\right\} \\ & + \frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left\{\left[V_{\alpha 11}(\text{rest}) - J(\text{rest})\bar{\rho}_{13}^*\right]\bar{\rho}_{12} + \hat{\psi}_{\alpha 21}(s)\bar{\rho}_{22} + \hat{\psi}_{\alpha 31}(s)\bar{\rho}_{32}\right\} \\ & + \frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left\{\left[V_{\beta 11}(\text{rest}) - J(\text{rest})\bar{\rho}_{12}^*\right]\bar{\rho}_{13} + \hat{\psi}_{\beta 21}(s)\bar{\rho}_{23} + \hat{\psi}_{\beta 31}(s)\bar{\rho}_{33}\right\}\end{aligned}\right\} \qquad (7.B.159)$$

$$\therefore \boxed{\begin{aligned}\hat{g}_{\text{mech}}(s) = {} & \left[V_{\alpha 11}(\text{rest at } s=-i\nu) - J(\text{rest at } s=-i\nu)\bar{\rho}_{13}^*\right] + \hat{\psi}_{\alpha 21}(-i\nu)\bar{\rho}_{23} + \hat{\psi}_{\alpha 31}(-i\nu)\bar{\rho}[illegible] \\ & + \frac{\omega_2}{\omega}\frac{\gamma_{21}}{\gamma_{31}}\left\{\left[V_{\beta 11}(\text{rest at } s=-i\nu) - J(\text{rest at } s=-i\nu)\bar{\rho}_{12}^*\right] + \hat{\psi}_{\beta 21}(-i\nu)\bar{\rho}_{22} + \hat{\psi}_{\beta 31}(-i\nu)\bar{\rho}_{32}\right\} \\ & + \frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left\{\left[V_{\alpha 11}(\text{rest at } s=-i\nu) - J(\text{rest at } s=-i\nu)\bar{\rho}_{13}^*\right] + \hat{\psi}_{\alpha 21}(-i\nu)\bar{\rho}_{22} + \hat{\psi}_{\alpha 31}(-i\nu)\bar{\rho}[illegible]\right\} \\ & + \frac{\omega}{\omega_2}\frac{\Gamma_{21}}{\gamma_{31}}\left\{\left[V_{\beta 11}(\text{rest at } s=-i\nu) - J(\text{rest at } s=-i\nu)\bar{\rho}[illegible]\right] + \hat{\psi}_{\beta 21}(-i\nu)\bar{\rho}_{23} + \hat{\psi}_{\beta 31}(-i\nu)\bar{\rho}[illegible]\right\}\end{aligned}} \qquad (7.B.160)$$

We can now see that once this expression is determined the value of the total spectral correlation function $\tilde{g}_{coh}(\nu)$, given by (7.B.98b), is known.

We may now write down the result in the form:-

$$\tilde{g}(\nu) = \tilde{g}_{coh}(\nu) + \tilde{g}_{incoh}(\nu) \tag{7.B.161a}$$

where

$$\tilde{g}_{coh}(\nu) = \tilde{g}_{coh_1}(\nu) + \omega/\omega_2 \, \Gamma_{21}/\gamma_{31} \, \tilde{g}_{coh_2}(\nu) + \omega_2/\omega \, \gamma_{21}/\gamma_{31} \, \tilde{g}_{coh_3}(\nu) + \omega/\omega_2 \, \Gamma_{21}/\gamma_{31} \, \tilde{g}_{coh_4}(\nu)$$

and

$$\begin{aligned} \tilde{g}_{coh_1}(\nu) &= 2\pi \, \delta(\Omega) \, |\bar{\rho}_{13}|^2 \\ \tilde{g}_{coh_2}(\nu) &= 2\pi \, \delta(\Omega) \, \bar{\rho}_{13}^{*} \bar{\rho}_{12} \\ \tilde{g}_{coh_3}(\nu) &= 2\pi \, \delta(\Omega) \, |\bar{\rho}_{12}|^2 \\ \tilde{g}_{coh_4}(\nu) &= 2\pi \, \delta(\Omega) \, \bar{\rho}_{12}^{*} \bar{\rho}_{13} \end{aligned} \tag{7.B.161b}$$

and

$$\begin{aligned} \tilde{g}_{incoh}(\nu) &= 2 \operatorname{Re} \hat{g}_{incoh}(-\nu) \\ &= 2 \operatorname{Re}\Big[\hat{g}_{incoh_1}(-\nu) + \omega/\omega_2 \, \Gamma_{21}/\gamma_{31} \, \hat{g}_{incoh_2}(-\nu) + \omega_2/\omega \, \gamma_{21}/\gamma_{31} \, \hat{g}_{incoh_3}(-\nu) + \omega/\omega_2 \, \Gamma_{21}/\gamma_{31} \, \hat{g}_{incoh_4}(-\nu) \Big] \\ \hat{g}_{incoh_1}(-\nu) &= \left[V_{\alpha 11}(rest(-\nu)) - J(rest(-\nu)) \bar{\rho}_{13}^{\,0} \right] + \hat{\psi}_{\alpha 21}(-\nu) \bar{\rho}_{23} + \hat{\psi}_{\alpha 31}(-\nu) \bar{\rho}_{33} \\ \hat{g}_{incoh_2}(-\nu) &= \left[V_{\alpha 11}(rest(-\nu)) - J(rest(-\nu)) \bar{\rho}_{13}^{\,0} \right] + \hat{\psi}_{\alpha 21}(-\nu) \bar{\rho}_{22} + \hat{\psi}_{\alpha 31}(-\nu) \bar{\rho}_{32} \\ \hat{g}_{incoh_3}(-\nu) &= \left[V_{\beta 11}(rest(-\nu)) - J(rest(-\nu)) \bar{\rho}_{12}^{\,*} \right] + \hat{\psi}_{\beta 21}(-\nu) \bar{\rho}_{22} + \hat{\psi}_{\beta 31}(-\nu) \bar{\rho}_{32} \\ \hat{g}_{incoh_4}(-\nu) &= \left[V_{\beta 11}(rest(-\nu)) - J(rest(-\nu)) \bar{\rho}_{12}^{\,*} \right] + \hat{\psi}_{\beta 21}(-\nu) \bar{\rho}_{23} + \hat{\psi}_{\beta 31}(-\nu) \bar{\rho}_{33} \end{aligned} \tag{7.B.161c}$$

Before we proceed with any further evaluations, we shall made certain approximations. We recall that (see equation (7.A.33)):-

$$\gamma_{31} = 4/3\hbar \, P_{31}^2 \, \omega^3/c^3 \qquad \gamma_{21} = 4/3\hbar \, P_{21}^2 \, \omega_2^3/c^3$$

$$\Gamma_{31} = 4/3\hbar \, P_{21} \cdot P_{31} \, \omega^3/c^3 \qquad \Gamma_{21} = 4/3\hbar \, P_{21} \cdot P_{31} \, \omega_2^3/c^3$$

Hence,

$$\Gamma_{21}/\Gamma_{31} = \left(\omega_2/\omega\right)^3$$

and in the case of the Na D lines:-

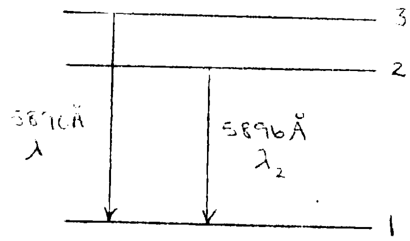


Fig 7 B.2

$$\left(\frac{\omega_2}{\omega}\right)^3_{Na} = \left(\frac{5890}{5896}\right)^3 = 0.9972$$

and
$$\left(\frac{\omega}{\omega_2}\right)^3_{Na} = \left(\frac{5896}{5890}\right)^3 = 1.002$$

Also in the case of the K lines:-

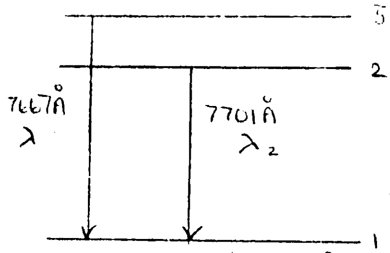


Fig 7 B.3

$$\left(\frac{\omega_2}{\omega}\right)^3_{K} = \left(\frac{7667}{7701}\right)^3 = 0.9863$$

$$\left(\frac{\omega}{\omega_2}\right)^3_{K} = \left(\frac{7701}{7667}\right)^3 = 1.014$$

Thus, for such differences between ω and ω_2, i.e. for $\lambda_{s2} \sim$ 4–6 Å, we can consider

$$\Gamma_{31} \simeq \Gamma_{21} \; (= \Gamma)$$

and we can also take (ω/ω_2) and (ω_2/ω) in the expression for $\tilde{g}(\nu)$ to be unity.

Let

$$\gamma_{21}/\gamma_{31} = d_2 \simeq p_{21}^2/p_{31}^2$$

and
$$\Gamma/\gamma_{31} = d_1 \simeq \mathbf{p}_{21}\cdot\mathbf{p}_{31}/p_{31}^2 = p_{21}/p_{31}\cos\theta$$

where $\cos\theta = \hat{p}_{21}\cdot\hat{p}_{31}$ and $\omega_2^3 \simeq \omega^3$

Then

$$d_2 = \phi d_1^2, \text{ where } \phi = 1/\cos^2\theta \simeq \gamma_{21}\gamma_{31}/\Gamma^2$$

If we assume that $p_{31}^2 \simeq p_{21}^2 \simeq \mathbf{p}_{31}\cdot\mathbf{p}_{21}$, i.e. $\hat{p}_{31}\cdot\hat{p}_{21} \simeq 1$, as in Chapter III, then $\gamma_{31} \simeq \gamma_{21} \simeq \Gamma(=\gamma)$, i.e. $d_1 = d_2 = 1$. This is a reasonable assumption, but we shall keep d_1 and d_2 unequal for the present.

Now, evaluating equations (7.B.158) at $s = -i\nu$ and using (7.B.154a),

$$V_{\alpha 11}(rest(-\nu)) = \frac{\hat{\psi}_{\alpha 11N}(rest(-\nu))}{T'} = \frac{\hat{\psi}_{\alpha 11N}(-\nu) - T_i\bar{\beta}_{13}^{*}}{(-i\Omega)T'}$$

and $\hat{\psi}_{\alpha 11N}(-\nu) = (-i\Omega)T'\hat{\psi}_{\alpha 11}(-\nu)$

and this can be shown to be the sum of two terms, one containing a factor of $(-i\Omega)$, as required, viz.

$$\hat{\psi}_{\alpha 11N}(-\nu) = (-i\Omega)T' V_{\alpha 11}(rest(-\nu)) + T_i\bar{\beta}_{13}^{*}$$

but for the present we shall use the expression for $\hat{\psi}_{\alpha 11N}(-\nu)$ already found (see equation (7.B.152)).

Now the intensity spectrum is represented by $\gamma_{31}\tilde{g}_{TOT}(\nu) = \tilde{I}(\nu)$ where

$$\begin{aligned}\gamma_{31}\tilde{g}_{TOT}(\nu) = \tilde{I}(\nu) &= \gamma_{31}\tilde{g}_{coh}(\nu) + \gamma_{31}\tilde{g}_{incoh}(\nu) \\ &= \gamma_{31}\tilde{g}_{coh}(\nu) + \gamma_{31}2Re\left[\hat{g}_{incoh}(-\nu)\right]\end{aligned} \tag{7.B.162a}$$

where

$$\begin{aligned}\tilde{g}_{coh}(\nu) &= \tilde{g}_{coh_1}(\nu) + d_1\tilde{g}_{coh_2}(\nu) + d_2\tilde{g}_{coh_3}(\nu) + d_1\tilde{g}_{coh_4}(\nu) \\ \tilde{g}_{coh_1}(\nu) &= 2\pi\,\delta(\Omega)\,|\bar{\beta}_{13}|^2 \\ \tilde{g}_{coh_2}(\nu) &= 2\pi\,\delta(\Omega)\,\bar{\beta}_{13}^{*}\bar{\beta}_{12} \\ \tilde{g}_{coh_3}(\nu) &= 2\pi\,\delta(\Omega)\,|\bar{\beta}_{12}|^2 \\ \tilde{g}_{coh_4}(\nu) &= 2\pi\,\delta(\Omega)\,\bar{\beta}_{12}^{*}\bar{\beta}_{13}\end{aligned} \tag{7.B.162b}$$

and

$$\begin{aligned}\hat{g}_{incoh}(-\nu) &= \hat{g}_{incoh_1}(-\nu) + d_1\hat{g}_{incoh_2}(-\nu) + d_2\hat{g}_{incoh_3}(-\nu) + d_1\hat{g}_{incoh_4}(-\nu) \\ \hat{g}_{incoh_1}(-\nu) &= \left[V_{\alpha 11}(rest(-\nu)) - J(rest(-\nu))\bar{\beta}_{13}^{*}\right]\bar{\beta}_{13} + \hat{\psi}_{\alpha 21}(-\nu)\bar{\beta}_{23} + \hat{\psi}_{\alpha 31}(-\nu)\bar{\beta}_{33} \\ \hat{g}_{incoh_2}(-\nu) &= \left[V_{\alpha 11}(rest(-\nu)) - J(rest(-\nu))\bar{\beta}_{13}^{*}\right]\bar{\beta}_{12} + \hat{\psi}_{\alpha 21}(-\nu)\bar{\beta}_{22} + \hat{\psi}_{\alpha 31}(-\nu)\bar{\beta}_{32} \\ \hat{g}_{incoh_3}(-\nu) &= \left[V_{\beta 11}(rest(-\nu)) - J(rest(-\nu))\bar{\beta}_{12}^{*}\right]\bar{\beta}_{12} + \hat{\psi}_{\beta 21}(-\nu)\bar{\beta}_{22} + \hat{\psi}_{\beta 31}(-\nu)\bar{\beta}_{32} \\ \hat{g}_{incoh_4}(-\nu) &= \left[V_{\beta 11}(rest(-\nu)) - J(rest(-\nu))\bar{\beta}_{12}^{*}\right]\bar{\beta}_{13} + \hat{\psi}_{\beta 21}(-\nu)\bar{\beta}_{23} + \hat{\psi}_{\beta 31}(-\nu)\bar{\beta}_{33}\end{aligned} \tag{7.B.162c}$$

N.B. $V_{\alpha 1}(rest(-\nu)) = \dfrac{\hat{\psi}_{\alpha 11N}(rest(-\nu))}{T'} = \dfrac{\hat{\psi}_{\alpha 11N}(-\nu) - T_i\bar{\beta}_{13}^{*}}{(-i\Omega)T'}$

$$V_{\beta 31}(\mathrm{cut}(i\nu)) = \frac{\hat{\psi}_{\beta 31}(\mathrm{cut}(i\nu))}{T'} - \frac{\hat{\psi}_{\beta 31}(i\nu) - T_1\,\bar{\rho}_{32}^{*}}{(i\nu)T'}$$

$$J(\mathrm{cut}(i\nu)) = \frac{T(\mathrm{cut}(i\nu))}{T'} - \frac{T'-T_1}{(i\nu)T'} \qquad \text{(7.B.162c) cont'd}$$

$\hat{\psi}_{\alpha 11}(i\nu)$, $\hat{\psi}_{\alpha 21}(i\nu)$, $\hat{\psi}_{\alpha 31}(i\nu)$, $\hat{\psi}_{\beta 11}(i\nu)$, $\hat{\psi}_{\beta 21}(i\nu)$, $\hat{\psi}_{\beta 31}(i\nu)$

are obtained from equations (7.B.152) and (7.B.153) where $s = i\nu$ and $\Gamma_{31} = \Gamma_{21} = \Gamma$.

We can rewrite the equilibrium density matrices given in equations (7.B.131)-(136), on letting $\Gamma_{31} = \Gamma_{21} = \Gamma$, as:-

$$\bar{\rho}_{11} = \frac{1}{|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]b_{30} + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]a_{30}\Big\}$$

$$\bar{\rho}_{22} = \frac{1}{|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big](b_{10} - b_{30}) + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big](a_{10} - a_{30}) + |H_0|^2 T_0\Big\}$$

$$\bar{\rho}_{33} = \frac{-1}{|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]b_{10} + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]a_{10}\Big\}$$

$$\bar{\rho}_{23} = \frac{-\tfrac{1}{2}\Gamma}{H_0|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[\tfrac{1}{4}G^2 b_{10} + (\tfrac{1}{4}G^2 - F_0)b_{30}\big] + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[\tfrac{1}{4}G^2 a_{10} + (\tfrac{1}{4}G^2 - F_0)a_{30}\big] + F_0|H_0|^2 T_0\Big\}$$

$$\text{(7.B.163)}$$

$$\bar{\rho}_{32} = \bar{\rho}_{23}^{*}$$

$$\bar{\rho}_{21} = \frac{-(i\lambda\mathcal{E}_0)\tfrac{1}{2}\Gamma}{H_0|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[(\gamma - i\Omega_3)b_{10} + (\tfrac{1}{2}\gamma_{31} + \gamma - i\Omega_3)b_{30}\big] + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big] \times \big[(\gamma - i\Omega_3)a_{10} + (\gamma + \tfrac{1}{2}\gamma_{31} - i\Omega_3)a_{30}\big] - \tfrac{1}{2}\gamma_{31}|H_0|^2 T_0\Big\}$$

$$\bar{\rho}_{12} = \bar{\rho}_{21}^{*}$$

$$\bar{\rho}_{31} = \frac{(i\lambda\mathcal{E}_0)}{H_0|H_0|^2 T_0}\Big\{\big[\gamma_{31}|H_0|^2 - \tfrac{1}{4}\Gamma^2(F_0H_0^* + F_0^*H_0)\big]\big[B_0 b_{10} + (B_0 + \tfrac{1}{4}\Gamma^2)b_{30}\big] + \big[\gamma_{21}|H_0|^2 + \tfrac{1}{4}\Gamma^2\tfrac{1}{4}G^2(H_0+H_0^*) - \tfrac{1}{2}\Gamma^2(F_0H_0^* + F_0^*H_0)\big] \times \big[B_0 a_{10} + (B_0 + \tfrac{1}{4}\Gamma^2)a_{30}\big] - \tfrac{1}{4}\Gamma^2|H_0|^2 T_0\Big\}$$

$$\bar{\rho}_{13} = \bar{\rho}_{31}^{*}$$

where $\Omega = \nu - \omega$

and $\gamma = \tfrac{1}{2}(\gamma_{21} + \gamma_{31})$

We shall now made the following substitutions:-

$$a^2 = \frac{\lambda C}{\gamma_{31}} = \frac{(\lambda C_{\cdot})}{\gamma_{31}^2} = \left(\frac{\hat{e}_{CD}\cdot \underline{p}_{31}\, \mathcal{E}_{CD}}{\hbar \gamma_{31}}\right)^2$$

$$b = \frac{\eta}{\gamma_{31}} = \frac{(\nu - \omega)}{\gamma_{31}} = \frac{(\nu - \epsilon_{31})}{\gamma_{31}}$$

$$c = \frac{\omega_{3}}{\gamma_{31}} = \frac{\epsilon_{32}}{\gamma_{31}}$$

where $\underline{p}_{31} = \underline{p}_{13}$ and is real, so that a is also real (7.B.164)

and

$$\frac{\gamma_{21}}{\gamma_{31}} = d_2 \simeq \frac{p_{21}^2}{p_{31}^2} \qquad \text{since} \quad \omega_2^3 \simeq \omega^3$$

$$\frac{\Gamma}{\gamma_{31}} = d_1 \simeq \frac{\underline{p}_{21}\cdot \underline{p}_{31}}{p_{31}^2} = \frac{p_{21}}{p_{31}} \cos\theta \qquad \text{where} \quad \cos\theta = \hat{p}_{21}\cdot\hat{p}_{31}$$

so that $\quad d_2 = \phi\, d_1^2 \quad$ where $\quad \phi = \frac{1}{\cos^2\theta} \simeq \frac{\gamma_{21}\gamma_{31}}{\Gamma^2}$ (7.B.165)

then

$$P = \gamma_{31}^{-2} B_1' = (\tfrac{1}{2}(1+d_2) - i(b+c))(\tfrac{1}{2}d_2 - i(b+c)) + a^2$$

$$Q = \gamma_{31}^{-2} F_1' = (\tfrac{1}{2}d_2 - i(b+c))(\tfrac{1}{2} - ib) - \tfrac{1}{4}d_1^2$$

$$\begin{aligned} R = \gamma_{31}^{-3} H_1' &= (\tfrac{1}{2}(1+d_2) - i(b+c))Q + a^2(\tfrac{1}{2} - ib) \\ &= (\tfrac{1}{2} - ib)P - \tfrac{1}{4}d_1^2(\tfrac{1}{2}(1+d_2) - i(b+c)) \\ &= (\tfrac{1}{2}(1+d_2) - i(b+c))(\tfrac{1}{2}d_2 - i(b+c))(\tfrac{1}{2} - ib) - \tfrac{1}{4}d_1^2(\tfrac{1}{2}(1+d_2) - i(b+c)) \\ &\qquad + a^2(\tfrac{1}{2} - ib) \end{aligned}$$

$$L = \gamma_{31}^{-2} B_2' = (\tfrac{1}{2}(1+d_2) - i(b-c))(\tfrac{1}{2}d_2 - i(b-c)) + a^2$$

$$M = \gamma_{31}^{-2} F_2' = (\tfrac{1}{2}d_2 - i(b-c))(\tfrac{1}{2} - ib) - \tfrac{1}{4}d_1^2$$

$$\begin{aligned} N = \gamma_{31}^{-3} H_2' &= (\tfrac{1}{2}(1+d_2) - i(b-c))M + a^2(\tfrac{1}{2} - ib) \\ &= (\tfrac{1}{2} - ib)L - \tfrac{1}{4}d_1^2(\tfrac{1}{2}(1+d_2) - i(b-c)) \\ &= (\tfrac{1}{2}(1+d_2) - i(b-c))(\tfrac{1}{2}d_2 - i(b-c))(\tfrac{1}{2} - ib) - \tfrac{1}{4}d_1^2(\tfrac{1}{2}(1+d_2) - i(b-c)) \\ &\qquad + a^2(\tfrac{1}{2} - ib) \end{aligned}$$

(7.B.166)

We shall also let,

$$X_1' = \hat{\psi}_{\alpha 11 N}(\text{rest}(-\nu)) = \frac{\hat{\psi}_{\alpha 11 N}(-\nu) - T_0\, \bar{\rho}_{13}^{\,\alpha}}{(-i\Omega)} = \gamma_{31}^{7} X_1 \qquad (7.B.167)$$

$$\left.\begin{aligned}
X_2' &= \hat{\psi}_{x21}(-\nu)\,T' && = \gamma_{31}^{7}\,X_2 \\
X_3' &= \hat{\psi}_{x31}(-\nu)\,T' && = \gamma_{31}^{7}\,X_3 \\
X_4' &= T\,(\mathrm{rest}(-\nu)) && = \frac{T'-T_0}{-i\Omega} = \gamma_{31}^{7}\,X_4 \\
X_9' &= \hat{\psi}_{\beta 11N}(\mathrm{rest}(-\nu)) && = \frac{\hat{\psi}_{\beta 11N}(-\nu) - T_0\,\bar{\beta}_{12}'}{-i\Omega} = \gamma_{31}^{7}\,X_9 \\
X_{10}' &= \hat{\psi}_{\beta 21}(-\nu)\,T' && = \gamma_{31}^{7}\,X_{10} \\
X_{11}' &= \hat{\psi}_{\beta 31}(-\nu)\,T' && = \gamma_{31}^{7}\,X_{11} \\
M_1' &= \hat{\psi}_{x11N}(-\nu) && = \gamma_{31}^{8}\,M_1 \\
M_2' &= \hat{\psi}_{\beta 11N}(-\nu) && = \gamma_{31}^{8}\,M_2
\end{aligned}\right\}\quad \begin{matrix}\text{(7.B.167)}\\ \text{cont'd}\end{matrix}$$

$$\left.\begin{aligned}
Y_1' &= B_0 = \gamma_{31}^{2}\,Y_1 = \gamma_{31}^{-2}\,P\,(b=0) \\
Y_2' &= F_0 = \gamma_{31}^{2}\,Y_2 = \gamma_{31}^{-2}\,Q\,(b=0) \\
Y_3' &= H_0 = \gamma_{31}^{3}\,Y_3 = \gamma_{31}^{-3}\,R\,(b=0)
\end{aligned}\right\}\quad \text{(7.B.168)}$$

$$\left.\begin{aligned}
T' &= \gamma_{31}^{8}\,T \\
T_0 &= \gamma_{31}^{8}\,T_0
\end{aligned}\right\}\quad \text{(7.B.169)}$$

$$\left.\begin{aligned}
a_1' &= \gamma_{31}^{7}\,A_1' \\
a_3' &= \gamma_{31}^{7}\,A_3' \\
b_1' &= \gamma_{31}^{7}\,B_1' \\
b_3' &= \gamma_{31}^{7}\,B_3'
\end{aligned}\right\}\quad \text{(7.B.170)}$$

$$\left.\begin{aligned}
a_{10} &= \gamma_{31}^{7}\,A_{10} = \gamma_{31}^{7}\,A_1'(b=0) \\
a_{30} &= \gamma_{31}^{7}\,A_{30} = \gamma_{31}^{7}\,A_3'(b=0) \\
b_{10} &= \gamma_{31}^{7}\,B_{10} = \gamma_{31}^{7}\,B_1'(b=0) \\
b_{30} &= \gamma_{31}^{7}\,B_{30} = \gamma_{31}^{1}\,B_3'(b=0)
\end{aligned}\right\}\quad \text{(7.B.171)}$$

$$
\left.\begin{aligned}
P_1^* &= Y_3|Y_3|^2 T_0 \bar{\beta}_{13}^* = Y_3|Y_3|^2 T_0 \left(\frac{M_1(b=0)}{T_0}\right) = Y_3|Y_3|^2 M_1(b=0) \\
P_2 &= Y_3|Y_3|^2 T_0 \bar{\beta}_{23} \\
P_3 &= Y_3|Y_3|^2 T_0 \bar{\beta}_{33} \\
P_4^* &= Y_3|Y_3|^2 T_0 \bar{\beta}_{21} = Y_3|Y_3|^2 T_0 \left(\frac{M_2(b=0)}{T_0}\right) = Y_3|Y_3|^2 M_2(b=0) \\
P_5 &= Y_3|Y_3|^2 T_0 \bar{\beta}_{22}
\end{aligned}\right\} \quad (7.B.172)
$$

$$
\left.\begin{aligned}
P_{11} &= PA_1' + (P + \tfrac{1}{4}d_1^2)A_3' \\
P_{12} &= PB_1' + (P + \tfrac{1}{4}d_1^2)B_3' \\
P_{13} &= (\tfrac{1}{2}(1+d_2) - i(b+c))A_1' + (\tfrac{1}{2}(2+d_2) - i(2b+c))A_3' \\
P_{14} &= (\tfrac{1}{2}(1+d_2) - i(b+c))B_1' + (\tfrac{1}{2}(2+d_2) - i(2b+c))B_3' \\
P_{15} &= (d_2 - ib)RN + \tfrac{1}{4}d_1 a^2(R+N) - \tfrac{1}{2}d_1^2(RM + NQ) \\
P_{16} &= (d_2 - ib)RN - \tfrac{1}{4}d_1^2(RM + NQ)
\end{aligned}\right\} \quad (7.B.173)
$$

$$
\left.\begin{aligned}
P_{110} &= P_{11}(b=0) = Y_1 A_{10} + (Y_1 + \tfrac{1}{4}d_1^2)A_{30} \\
P_{120} &= P_{12}(b=0) = Y_1 B_{10} + (Y_1 + \tfrac{1}{4}d_1^2)B_{30} \\
P_{130} &= P_{13}(b=0) = (\tfrac{1}{2}(1+d_2) - ic)A_{10} + (\tfrac{1}{2}(2+d_2) - ic)A_{30} \\
P_{140} &= P_{14}(b=0) = (\tfrac{1}{2}(1+d_2) - ic)B_{10} + (\tfrac{1}{2}(2+d_2) - ic)B_{30} \\
P_{150} &= P_{15}(b=0) = d_2|Y_3|^2 + \tfrac{1}{4}d_1 a^2(Y_3 + Y_3^*) - \tfrac{1}{2}d_1^2(Y_2 Y_3^* + Y_2^* Y_3) \\
P_{160} &= P_{16}(b=0) = d_2|Y_3|^2 - \tfrac{1}{4}d_1^2(Y_2 Y_3^* + Y_2^* Y_3)
\end{aligned}\right\} \quad (7.B.174)
$$

Hence

$$
\left.\begin{aligned}
X_1 &= \frac{M_1 - T_0\bar{\beta}_{13}^*}{-ib} = \frac{M_1 - M_1(b=0)}{-ib} = \frac{M_1 - P_1^*/Y_3|Y_3|^2}{-ib} = \frac{Y_3|Y_3|^2 M_1 - P_1^*}{Y_3|Y_3|^2(-ib)} \\
X_9 &= \frac{M_2 - T_0\bar{\beta}_{12}^*}{-ib} = \frac{M_2 - M_2(b=0)}{-ib} = \frac{M_2 - P_4^*/Y_3|Y_3|^2}{-ib} = \frac{Y_3|Y_3|^2 M_2 - P_4^*}{Y_3|Y_3|^2(-ib)}
\end{aligned}\right\} \quad (7.B.175)
$$

$$
X_4 = \frac{T - T_0}{-ib}
$$

$$
\left.\begin{aligned}
M_1 &= \frac{ia}{RN}\left[P_{15}P_{11} + P_{16}P_{12} - \tfrac{1}{4}d_1^2 RNT\right] \\
M_2 &= -\frac{ia\,\tfrac{1}{2}d_1}{RN}\left[P_{15}P_{13} + P_{16}P_{14} - (\tfrac{1}{2} - ib)RNT\right] \\
X_2 &= \frac{\tfrac{1}{2}d_1}{R}\left[a^2\left\{\left(\tfrac{1}{2}(3+d_2) - i(3b+c)\right)P_{11} + (\tfrac{1}{2} - ib)P_{12}\right\} + \left(\tfrac{1}{2}(1+d_2) - i(b+c)\right)RT\right] \\
X_3 &= \frac{1}{R}\left[-a^2\left\{(P + \tfrac{1}{2}d_1^2)P_{11} + \tfrac{1}{4}d_1^2 P_{12}\right\} + PRT\right] \\
X_{10} &= -\frac{1}{R^2}\Big[a^2\,\tfrac{1}{4}d_1^2\left\{\left(\tfrac{1}{2}(3+d_2) - i(3b+c)\right)P_{13} + (\tfrac{1}{2} - ib)P_{14}\right\} \\
&\qquad - (\tfrac{1}{2} - ib)\left(\tfrac{1}{2}(1+d_2) - i(b+c)\right)RT\Big] \\
X_{11} &= \frac{\tfrac{1}{2}d_1}{R}\left[a^2\left\{(P + \tfrac{1}{2}d_1^2)P_{13} + \tfrac{1}{4}d_1^2 P_{14}\right\} - \left(\tfrac{1}{2}(1+d_2) - i(b+c)\right)RT\right]
\end{aligned}\right\} \text{(7.B.175) cont'd}
$$

$$
\left.\begin{aligned}
T &= \frac{A_3' B_1' - A_1' B_3'}{RN} \\
T &= \frac{A_{30}B_{10} - A_{10}B_{30}}{|Y_3|^2}
\end{aligned}\right\} \text{(7.B.176)}
$$

$$
\left.\begin{aligned}
Y_1 &= \left(\tfrac{1}{2}(1+d_2) - ic\right)\left(\tfrac{1}{2}d_2 - ic\right) + a^2 \\
Y_2 &= \tfrac{1}{2}\left(\tfrac{1}{2}d_2 - ic\right) - \tfrac{1}{4}d_1^2 \\
Y_3 &= \tfrac{1}{2}
\end{aligned}\right\} \text{(7.B.177)}
$$

$$
\left.\begin{aligned}
A_1' &= -(d_2 - ib)RN - \tfrac{1}{4}d_1^2 a^2(R+N) + \tfrac{1}{4}d_1^2(RM + QN) \\
A_3' &= -(d_2 - ib)RN + \tfrac{1}{4}d_1^2 a^2(R+N) \\
B_1' &= (d_2 - ib)RN + \tfrac{3}{4}d_1^2 a^2(R+N) + a^2(LR + PN) - \tfrac{1}{2}d_1^2(RM + QN) \\
B_3' &= (d_2 - 1)RN - \tfrac{1}{2}d_1^2 a^2(R+N) - a^2(LR + PN)
\end{aligned}\right\} \text{(7.B.178)}
$$

$$
\left.\begin{aligned}
A_{10} &= -d_2|Y_3|^2 - \tfrac{1}{4}d_1^2 a^2(Y_3 + Y_3^*) + \tfrac{1}{4}d_1^2(Y_2 Y_3^* + Y_2^* Y_3) \\
A_{30} &= -d_2|Y_3|^2 + \tfrac{1}{4}d_1^2 a^2(Y_3 + Y_3^*) \\
B_{10} &= d_2|Y_3|^2 + \tfrac{3}{4}d_1^2 a^2(Y_3 + Y_3^*) + a^2(Y_1 Y_3^* + Y_1^* Y_3) \\
&\qquad - \tfrac{1}{2}d_1^2(Y_2 Y_3^* + Y_2^* Y_3) \\
B_{30} &= (d_2 - 1)|Y_3|^2 - \tfrac{1}{2}d_1^2 a^2(Y_3 + Y_3^*) - a^2(Y_1 Y_3^* + Y_1^* Y_3)
\end{aligned}\right\} \text{(7.B.179)}

$$\left.\begin{aligned}
P_1^* &= ia\left[P_{140}P_{130} + P_{150}P_{110} - \tfrac{1}{4}d_1^2|Y_3|^2T_0\right] \\
P_2 &= -\tfrac{1}{2}d_1\left[P_{140}\{a^2B_{10} + (a^2-Y_2)B_{30}\} + P_{150}\{a^2A_{10} + (a^2-Y_2)A_{30}\} + Y_2|Y_3|^2T_0\right] \\
P_3 &= -Y_3\left[P_{140}B_{10} + P_{150}A_{10}\right] \\
P_4^* &= -ia\tfrac{1}{2}d_1\left[P_{140}P_{140} + P_{150}P_{150} - \tfrac{1}{2}|Y_3|^2T_0\right] \\
P_5 &= Y_3\left[P_{140}(B_{10}-B_{30}) + P_{150}(A_{10}-A_{30}) + |Y_3|^2T_0\right]
\end{aligned}\right\} \quad (7.B.180)$$

We can further factorise the expressions for P_1, P_2, P_3, P_4, P_5, T, T, M_1, M_2, X_1, X_9, X_4, X_2, X_3, X_{10}, X_{11} given in equations (7.B.177), (7.B.178), (7.B.182) so that we obtain the following expressions:

$$\left.\begin{aligned}
S_1^* &= \frac{P_1^*}{Y_3|Y_3|^2} = ia\Big[Y_1\{-d_2(Y_3^* + \tfrac{1}{4}d_1^2a^2) + \tfrac{1}{4}d_1^2(d_2+1)Y_2^*\} + \tfrac{1}{4}d_1^2\{(d_2+1)[(\tfrac{1}{2}d_2-ic)Y_3^* + \tfrac{1}{4}d_1^2a^2] \\
&\qquad + a^2d_2Y_1^*\}\Big] \\
S_2 &= \frac{P_2}{Y_3|Y_3|^2} = -\tfrac{1}{2}d_1\Big[a^2\{-d_2Y_3^*(1+\tfrac{1}{2}d_2-ic) - \tfrac{1}{4}d_1^2(a^2d_2 - (d_2+1)Y_2^*\} \\
&\qquad - Y_2a^2\{\tfrac{1}{4}d_1^2(d_2+1) + d_2Y_1^*\}\Big] \\
S_3 &= \frac{P_3}{Y_3|Y_3|^2} = -a^2\Big[Y_3\{d_2Y_1^* + \tfrac{1}{4}d_1^2(\tfrac{1}{2}d_2-ic)\} + Y_3^*\{d_2Y_1 + \tfrac{1}{4}d_1^2(\tfrac{1}{2}d_2+ic) \\
&\qquad - \tfrac{1}{4}d_1^2\{d_2(Y_1Y_2^* + Y_1^*Y_2) + \tfrac{1}{2}d_1^2a^2\}\Big] \\
S_4^* &= \frac{P_4^*}{Y_3|Y_3|^2} = -ia\tfrac{1}{2}d_1\Big[\{\tfrac{1}{2}(1+d_2)-ic\}\{-\tfrac{1}{4}d_1^2a^2d_2 - d_2Y_3^* + \tfrac{1}{4}d_1^2(1+d_2)Y_2^*\} \\
&\qquad + \tfrac{1}{2}a^2\{\tfrac{1}{4}d_1^2(1+d_2) + d_2Y_1^*\} \\
&\qquad + Y_3^*\{\tfrac{1}{4}d_1^2(1+d_2) + a^2d_2\}\Big] \\
S_5 &= \frac{P_5}{Y_3|Y_3|^2} = -\tfrac{1}{4}d_1^2a^2\Big[(Y_3+Y_3^*) + \tfrac{1}{2}d_1^2a^2 + (Y_1Y_2^* + Y_1^*Y_2) + (\tfrac{1}{2}d_2+ic)Y_3 \\
&\qquad + (\tfrac{1}{2}d_2-ic)Y_3^*\Big]
\end{aligned}\right\} \quad (7.B.181)$$

$$\left.\begin{aligned}
T &= -\Big[d_2|Y_3|^2 + \tfrac{1}{4}d_1^2a^2(3d_2+1)(Y_3+Y_3^*) + 2a^2d_2(Y_1Y_3^* + Y_1^*Y_3) \\
&\qquad - \tfrac{1}{4}d_1^2(d_2+1)(Y_2Y_3^* + Y_2^*Y_3) - \tfrac{1}{8}d_1^4a^4 - \tfrac{1}{4}d_1^2a^2(Y_1Y_2^* + Y_1^*Y_2) \\
&\qquad - \tfrac{1}{4}d_1^2a^2\{(\tfrac{1}{2}d_2+ic)Y_3 + (\tfrac{1}{2}d_2-ic)Y_3^*\}\Big] \\
T &= -\Big[(d_2-ib)(1-ib)RN + \tfrac{1}{4}d_1^2a^2(3d_2+1-4ib)(R+N) + 2a^2(d_2-ib)(LR+PN) \\
&\qquad - \tfrac{1}{2}d_1^2(\tfrac{1}{2}(1+d_2)-ib)(RM+QN) - \tfrac{1}{8}d_1^4a^4 - \tfrac{1}{4}d_1^2a^2(PM+QL) \\
&\qquad - \tfrac{1}{4}d_1^2a^2\{(\tfrac{1}{2}d_2-i(b-c))R + (\tfrac{1}{2}d_2-i(b+c))N\}\Big]
\end{aligned}\right\} \quad (7.B.182)$$

$$
\left.\begin{aligned}
X_1 = ia\Big\{ & -\Big[(Y_1 - ibP')\{(1-ib)[(1-ib)N' + Y_2^*] + Y_2^* + \tfrac14 d_1^2 a^2\} \\
& \qquad + P' d_2 (Y_2^* + \tfrac14 d_1^2 a^2)\Big] \\
& + \tfrac12 d_1^2 \Big[(Y_1 - ibP')\{(\tfrac12(1+d_2)-ib)M' + Y_2^*\} + P'\tfrac12(1+d_2)Y_2^* \\
& \qquad + (\tfrac12(1+d_2)-ib)\{(\tfrac12 d_2 - i(b+c))N' + Y_3^*\} \\
& \qquad + (\tfrac12 d_2 - ic)Y_3^* + \tfrac14 d_1^2 a^2\Big] \\
& + \tfrac12 a^2 \big[(d_2 - ib)L' + Y_1^*\big]\Big\}
\end{aligned}\right\} \quad \textbf{(7.B.183a)}
$$

$$
\left.\begin{aligned}
X_3 = -ia\tfrac12 d_1\Big\{ & a\big[(\tfrac12 + d_2 - ib)(Y_1^* - ibL') + \tfrac12 d_2 L'\big] \\
& + \tfrac14 d_1^2\big[2(1+d_2 - i(b+c))(Y_2^* - ibM') + (1+d_2)(\tfrac12(1+d_2) - ic)M'\big] \\
& - \big[(Y_3^* + ibN')\{(\tfrac12(1+3d_2) - i(b+c))(1-ib) + d_2(\tfrac12(1+d_2) - ic)\}\big] \\
& + N'\big[d_2(\tfrac12(1+d_2) - ic)\big] \\
& + \tfrac12 d_1^2\big[Y_3^* + (1+d_2 - ib)N'\big] \\
& + a^2\big[Y_3^* + (d_2 - ib)N'\big] \\
& + \tfrac14 d_1^2 a^2\big[\tfrac12(3-d_2) + i(b+c)\big]\Big\}
\end{aligned}\right\} \quad \textbf{(7.B.183b)}
$$

$$
\left.\begin{aligned}
X_4 = -\Big\{ & (Y_3^* R' + Y_3 N')\big[d_2 - ib(d_2 + 1 - ib)\big] + |Y_3|^2(d_2 + 1 - ib) - ibR'N'(2d_2 + 1 - ib) \\
& + \tfrac14 d_1^2 a^2\big[(R' + N')(3d_2 + 1 - 4ib) + 4(Y_3 + Y_3^*)\big] \\
& + 2a^2\big[(Y_1^* R' + Y_1 N' + Y_3 L' + Y_3^* P')(d_2 - ib) + Y_1^* Y_3 + Y_1 Y_3^* \\
& \qquad - ib(L'R' + P'N')(d_2 - ib)\big] \\
& - \tfrac12 d_1^2\big[(Y_3 M' + Y_3^* Q' + Y_2 N' + Y_2^* R')(\tfrac12(1+d_2) - ib) + Y_3 Y_2^* + Y_3^* Y_2 \\
& \qquad - ib(R'M' + Q'N')(\tfrac12(1+d_2) - ib)\big] \\
& - \tfrac14 d_1^2 a^2\big[Y_1 M' + Y_1^* Q' + Y_2 L' + Y_2^* P' - ib(P'M' + Q'L') \\
& \qquad + R'(\tfrac12 d_2 - i(b-c)) + N'(\tfrac12 d_2 - i(b+c)) + Y_3 + Y_3^*\big]\Big\}
\end{aligned}\right\} \quad \textbf{(7.B.183c)}
$$

$$
\left.\begin{aligned}
X_2 = \tfrac12 d_1\Big\{ & \big[\tfrac12(1+d_2) - i(b+c)\big]\big[(d_2 - ib)(1-ib)N + \tfrac14 d_1^2 a^2(3d_2 + 1 - 4ib) + 2a^2(d_2 - ib)L \\
& \qquad - \tfrac12 d_1^2\{\tfrac12(1+d_2) - ic\}M - \tfrac14 d_1^2 a^2(\tfrac12 d_2 - i(b-c))\big] \\
& + \tfrac14 d_1^2 a^2(a^2 - L) \\
& - (2d_2 + 1 - 3ib)a^2 \\
& - \tfrac12 d_1^2(1 + d_2 - ib)\Big\}
\end{aligned}\right\} \quad \textbf{(7.B.183d)}
$$

$$
\left.\begin{aligned}
X_3 = -P\{&(1_2-ib)(1-ib)N - \tfrac{1}{4}d_1^2a^2(3d_2+1-4ib) + 2a^2(d_2-ib)L - \tfrac{1}{2}d_1^2(\tfrac{1}{2}(1+d_2)-ib)M \\
&- \tfrac{1}{4}d_1^2a^2(\tfrac{1}{2}d_2-i(b-c))\} \\
&+ (\tfrac{1}{2}d_2-i(b+c))\}\tfrac{1}{4}d_1^2a^2L + 2(\tfrac{1}{2}(1+d_2)-ib)\}
\end{aligned}\right\} \quad (7.B.183e)
$$

$$
\left.\begin{aligned}
X_{10} = -\{&(\tfrac{1}{2}-ib)(\tfrac{1}{2}(1+d_2)-i(b+c))[(d_2-ib)(1-ib)N + \tfrac{1}{4}d_1^2a^2(3d_2+1-4ib) \\
&\qquad + 2a^2(d_2-ib)L - \tfrac{1}{2}d_1^2(\tfrac{1}{2}(1+d_2)-ib)M \\
&\qquad - \tfrac{1}{4}d_1^2a^2(\tfrac{1}{2}d_2-i(b-c))] \\
&+ N[2a^2(d_2-ib)(\tfrac{1}{2}(1+d_2)-i(b+c)) - \tfrac{1}{2}d_1^2(\tfrac{1}{2}-ib)(\tfrac{1}{2}(1+d_2)-ib) - \tfrac{1}{4}d_1^2a^2] \\
&+ \tfrac{1}{4}d_1^2a^2[(\tfrac{1}{2}(1+d_2)-i(b+c))M - (\tfrac{1}{2}-ib)L]\}
\end{aligned}\right\} \quad (7.B.183f)
$$

$$
\left.\begin{aligned}
X_{11} = \tfrac{1}{2}d_1\{&[\tfrac{1}{2}(1+d_2)-i(b+c)][(1_2-ib)(1-ib)N + \tfrac{1}{4}d_1^2a^2(3d_2+1-4ib) \\
&\qquad + 2a^2(d_2-ib)L - \tfrac{1}{2}d_1^2(\tfrac{1}{2}(1+d_2)-ib)M \\
&\qquad - \tfrac{1}{4}d_1^2a^2(\tfrac{1}{2}d_2-i(b-c))] \\
&\quad - [a^2(d_2-ib) + \tfrac{1}{2}d_1^2(\tfrac{1}{2}(1+d_2)-ib)] \\
&\quad + \tfrac{1}{4}d_1^2a^2(L-a^2)\}
\end{aligned}\right\} \quad (7.B.183g)
$$

where

$$
\left.\begin{aligned}
P &= Y_1 - ibP', \quad P' = \tfrac{1}{2}(1+2d_2) - i2(b+c) \\
Q &= Y_2 - ibQ', \quad Q' = \tfrac{1}{2}(1+d_2) - i(2b+c) \\
R &= Y_3 - ibR', \quad R' = \tfrac{1}{2}\{\tfrac{1}{2}(1+2d_2)-i2(b+c)\} + \{\tfrac{1}{2}(1+d_2)-i(b+c)\}\{\tfrac{1}{2}d_2-i(b+c)\} \\
&\qquad\qquad - \tfrac{1}{4}d_1^2 + a^2 \\
L &= Y_1^* - ibL', \quad L' = \tfrac{1}{2}(1+2d_2) - i2(b-c) \\
M &= Y_2^* - ibM', \quad M' = \tfrac{1}{2}(1+d_2) - i(2b-c) \\
N &= Y_3^* - ibN', \quad N' = \tfrac{1}{2}\{\tfrac{1}{2}(1+2d_2)-i2(b-c)\} + \{\tfrac{1}{2}(1+d_2)-i(b-c)\}\{\tfrac{1}{2}d_2-i(b-c)\} \\
&\qquad\qquad - \tfrac{1}{4}d_1^2 + a^2
\end{aligned}\right\} \quad (7.B.184)
$$

Hence we can write the intensity spectrum as:-

$$
\left.\begin{aligned}
I(b) &= \widehat{I}_{coh}(b) + \widehat{I}_{incoh}(b) \\
\text{where} \quad \widehat{I}_{coh}(b) &= \sum_{i=1}^{4} \widehat{I}_{coh_i}(b) \\
\text{and} \quad \widehat{I}_{incoh}(b) &= \frac{2\pi\,\delta(b)\,|S_1|^2}{T_0^2} \\
\widehat{I}_{incoh_1}(b) &= \frac{2\pi\,\delta(b)\,S_1^*S_4}{T_0^2}
\end{aligned}\right\} \quad (7.B.185)
$$

$$\tilde{I}_{coh_3}(b) = \frac{2\pi\,\delta(b)\,|S_4|^2}{T_0^{\,2}}$$

$$\tilde{I}_{coh_4}(b) = \frac{2\pi\,\delta(b)\,S_4^{*}S_1}{T_0^{\,2}}$$

$$\tilde{I}_{incoh}(b) = \sum_{i=1}^{4} \tilde{I}_{incoh_i}(b)$$

$$\tilde{I}_{incoh_1}(b) = \frac{2}{|T|^2 T_0^2}\,\mathrm{Re}\Big[\,T^{*}\big\{T_0\left(X_1S_1 + X_2S_2 + X_3S_3\right) - X_4\,|S_1|^2\big\}\Big]$$

$$\tilde{I}_{incoh_2}(b) = \frac{2}{|T|^2 T_0^2}\,\mathrm{Re}\Big[T^{*}d_1\big\{T_0\left(X_1S_4 + X_2S_5 + X_3S_2^{*}\right) - X_4\,S_1^{*}S_4\big\}\Big]$$

$$\tilde{I}_{incoh_3}(b) = \frac{2}{|T|^2 T_0^2}\,\mathrm{Re}\Big[T^{*}d_2\big\{T_0\left(X_9S_4 + X_{10}S_5 + X_{11}S_2^{*}\right) - X_4\,|S_4|^2\big\}\Big]$$

$$\tilde{I}_{incoh_4}(b) = \frac{2}{|T|^2 T_0^2}\,\mathrm{Re}\Big[T^{*}d_1\big\{T_0\left(X_9S_1 + X_{10}S_2 + X_{11}S_3\right) - X_4\,S_4^{*}S_1\big\}\Big]$$

(7.B.125) cont'd

Graphs for $\tilde{I}(b)$ against b can now be plotted for various values of a, c, d_1, d_2 and some typical computer generated ones are shown in section E for $d_1 = d_2 = 1$, a = 3 and various values of c. These, and others for which data only are given in section C, are analysed in the following section C, part (c).

C. Analysis of spectral profiles shown in Section E for $d_1 = d_2 = 1$

(a) Predictions

We shall first of all consider what effect the parameters a and c might have on the spectria profile $\tilde{I}(b)$.

In the case of a, we should expect noticeable non-linear effects for a large since a is related to the amplitude of the driving field, $\mathcal{E}_0$, by the expression a = $\mathcal{E}_0\lambda/\gamma$ and so when a is large the driving field is strong. When $\mathcal{E}_0$ is of the magnitude to be found in laser radiation the field-splitting of atomic level 3, a non-linear effect, should be significant. We deduce this from the fact that such field-dependent splitting was found in the cases of potassium and hydrogen (see Chapters V and VI). Our assumption that the field has one mode, or one frequency, is particularly valid for such strong fields.

On the other hand, in the case of c, we should expect the asymmetry in the magnitude of the intensity of the peaks to be noticeable (see ref. 8). c is related to the separation of the atomic levels 2 and 3, ω_3, by the expression $c = \omega_3/\gamma$ and so when c is small, ω_3 is small compared with the line-width γ. When ω_3 is in the range of 0 to 1 the overlapping of the line-widths of levels 3 and 2 varies from a maximum of γ to 0:-

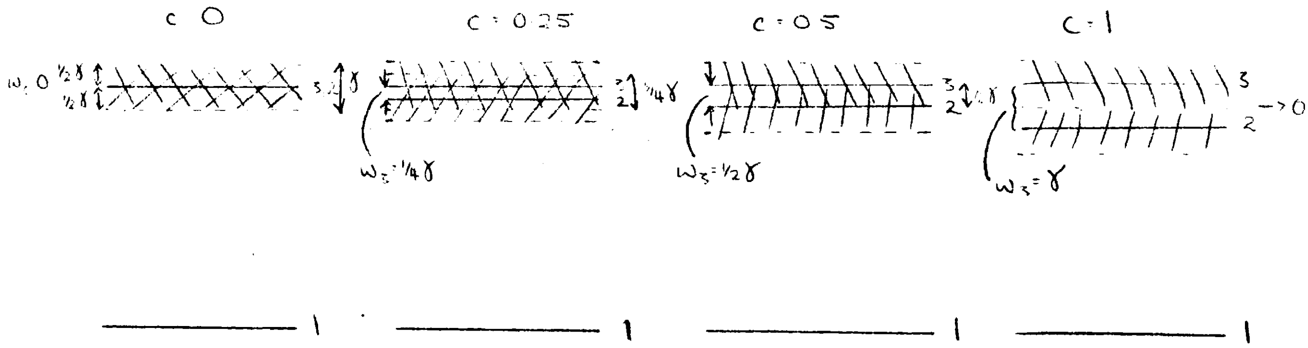


Fig 7.C.1

In Chapter III we saw how the profiles for spontaneous emission became asymmetric when levels 3 and 2 overlapped. Graphs for c = 1 and c = ½ are given in Figs. 3.B.6a and 6b respectively of Chapter III.

Since in the present case we have profiles dependent on a and c the effects of field splitting and asymmetry should both be present. The field splitting should be large for a large and the asymmetry large for c small. Splitting would only disappear for a = 0 but we should expect it be unresolvable for a sufficiently small. Similarly, when c is larger than 1, one would expect the asymmetry due to overlapping of line widths to be negligible, although, if c is not too large, then level 2 might still be close enough to level 3 to effect the profile in some way. The expected results might be further complicated by the position or the split levels of level 3 relative to level 2. Now we should expect the split levels to occur at b = ±2a (see ref. 9) so that the magnitude of a relative to c is also important. E.g. if a = 1 then split levels should occur at $b_{1,2} = \pm 2$ and if c = 1 also then

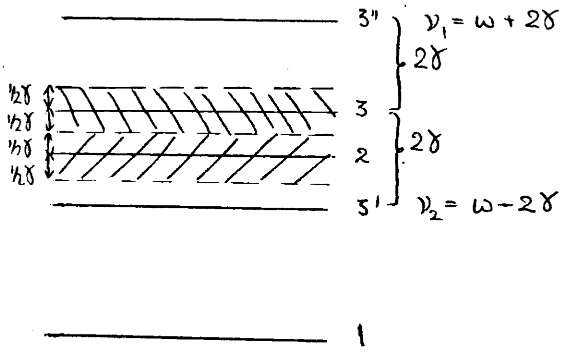


Fig. 7.C.2

If $\underline{a = \frac{1}{2}}$, then $\underline{b_{1,2} = \pm 1}$ and if $c = \frac{1}{2}$ also then

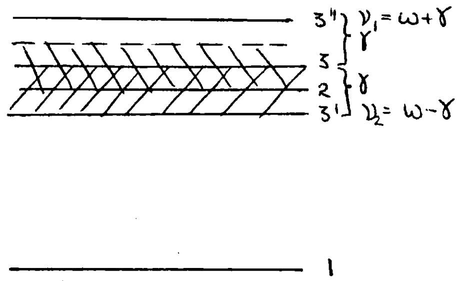


Fig. 7.C.3

If $a = \frac{1}{4}$, then $b_{1,2} = \pm\frac{1}{2}$ and, if $c = \frac{1}{4}$ also

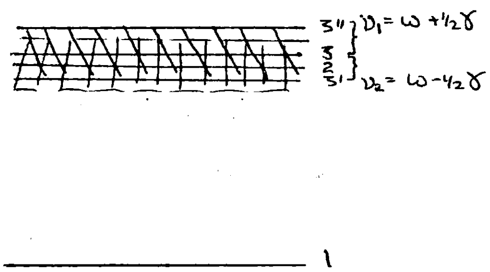


Fig. 7.C.4

In this case the split levels occur at the extremities of the half width of level 3 and will not be resolvable, though they might appear as shoulders on the resonance profile if their intensity is sufficiently large, viz.

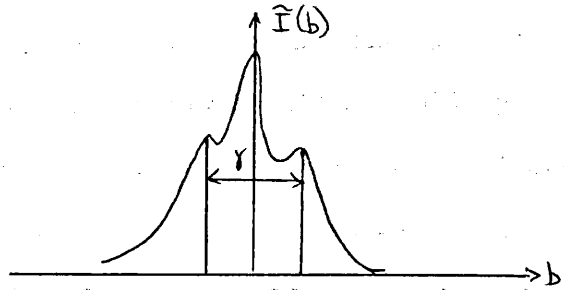


Fig. 7.C.5

Since $c < 1$ these shoulders are shown to be asymmetric in intensity. Suitable values for a and c would be $a \geq 0.25$, $c = 0 \rightarrow 1$, for the effects of overlapping and $c = 1$ upwards for the effect of level 2 when not overlapping level 3. When $c \gg 1$ then the atom would be effectively a 2-level atom and the results would be only due to the field. When $a < \frac{1}{4}$ one would not expect anything different from the normal Lorentzian profile. When a is very large one would expect the profiles of the split levels to be independent of c.

Newstein has pointed out that the size of the driving field is critical. In ref. 65 where he considers the effect of collisional relaxation he says that if the driving field is not sufficiently large to alter the state of the atomic system in a relaxation time, then the spectral distribution of the spontaneous emission will be unaffected (see also ref. 85). In ref. 66 he says that the effect would not be significant in such a case. We can see, in the above analysis, that unless $a > \frac{1}{4}$ the profile would be Lorentzian.

Newstein[66] considers the spectrum for spontaneous emission from an ensemble of 2-level atoms interacting with a relaxation mechanism and the effect of the application of a classical driving field. He finds that the single Lorentzian line-shape characteristic of the power spectrum of spontaneous emission for the undriven case is split into components by the driving field and that their splitting is associated with the establishment of definite phase relations between the corresponding components of the field spectrum. The effects become significant when the strength of the field is sufficient to appreciably alter the state of the material system in a relaxation time. He says that: "The splitting of the power spectrum can be associated with the sinusoidal modulation of the population of the upper level of the material system,* between relaxation collisions, due to the coupling to the driving field. The establishment of phase relations between the components of the field spectrum can be associated with response characteristics of the driven material system. Senitzky et al.[67] have shown that the linear response to an additional small signal depends on the phase of the small signal relative to the driving field. The susceptibility of the medium to a small signal phased for frequency modulation relative to a resonant driving field has a Lorentzian line shape centred at the central atomic frequency. The susceptibility to a small signal phased for amplitude modulation consists of the sums of the two Lorentzians, symmetrically displaced relative to the central frequency. These three peaks in the

* cf. note on oscillation at the Rabi frequency in Chapter II.

susceptibility and the associated phase relations correspond to the same features of the spontaneous emission spectrum". The three peaks of Newstein's[(65)] Fig. 1 resemble those of Mollow[(9)] except that Newstein's curves are for $\Omega = 4K$, when the shape is similar to Mollow's for $\Omega = 3K$ and $5K$ except that the side peaks occur at ± 4 instead of at ± 3 and ± 5; for $\Omega = 2K$, when the side peaks merge with the central one, as they do for $\Omega = K$ in Mollow's case, and for $\Omega = 0$ when there is no driving field.

Apanasevich[(34)] does not obtain 3 peaks in his susceptibility curves (Fig. 2), owing to the fact that they only represent linear susceptibility resulting from a weak field, i.e. a field insufficiently strong to alter the state of the atomic system in a relaxation time. His anomalies are due to the nearness of the upper levels and the fact that the weak field can couple both upper levels to the ground level since it has a frequency spread.

We can however deduce some results from those of Mollow[(9)], since, if we compare our equations (7.B.2), (7.B.4), (7.B.6), (7.B.9) for $\dot{\rho}_{11}$, $\dot{\rho}_{33}$, $\dot{\rho}_{13}$ and $\dot{\rho}_{31}$, for the case when $\gamma_{21} = \gamma_{31} = \Gamma_{21} = \Gamma_{31} = \gamma$, with those of Mollow for the driven 2 level situation, we see that the equations are equivalent, bearing in mind that

$$\begin{aligned}
\bar{n} &\longrightarrow \rho_{33} \\
\bar{m} &\longrightarrow \rho_{11} \\
\alpha &\longrightarrow \rho_{31} \\
\alpha^* &\longrightarrow \rho_{13} \\
K &\longrightarrow \gamma \\
\tfrac{1}{4}\Omega^2 &\longrightarrow \tfrac{1}{4}G^2
\end{aligned}$$

In Mollow's case he finds that, for very intense fields, $\Omega \gg |\Delta\omega|$, K, the peaks of the power spectrum occur at $\nu = \omega_0$ and $\nu = \omega_0 \pm \Omega$, i.e. if $\Omega = nK$, then the side-peaks are near to $\nu = \omega_0 \pm nK$. Therefore we should also expect peaks at $\nu = \omega \pm G$, i.e. at $b = \pm 2a$, for very intense fields.

(b) Review of other relevant references (for the purpose of comparing results)

The first reference of interest is the paper by Morozov and Shcherygin (ref. 8) which has already been discussed, both in Chapter III and in part (c) of the present section. They find that the central peak in profile for spontaneous emission from a 3-level atom initially in level 3 becomes shifted to the left and increased in height owing to the influence of the exchange of virtual photons between overlapping intermediate levels. This change becomes increasingly apparent for decreasing c and is negligible for c >> 1. In our case we should expect the split levels of level 3 to be affected by the magnitude of c and not the central resonance peak since we are not considering the undriven atom.

In ref. 68 a molecule with 2 intermediate levels is considered when it is coupled to a quantised radiation field only and allowance is also made for virtual photon exchange between overlapping intermediate levels. They calculate the intensity and shape of the absorbed and secondary radiation lines using the Heitler-Ma method[47] as do the authors of ref. 8, and point out that it provides a more rigorous and complete solution than does that of Weisskopf and Wigner[41]. In their figs. 2-5 they show how consideration of the extra intermediate level results in changes in intensity profile. It is interesting to note that when considering one intermediate level only they find that consideration of the transformation of light by the Heitler-Ma method, which we saw in Chapter III, gives the same results as our method, for $\tau_j << t << \tau_i$, is fully equivalent mathematically to the consideration of the processes in the non-closed radiation field-molecule system suggested by Apanasevich (refs. 69 & 70) who claims his model accurately reflects real experimental conditions.

In ref. 71, Morozov calculates the influence of incomplete intermixing of degenerate states on the line-shape of spontaneous emission. The degeneracy of the 2 excited levels is eliminated by a perturbation of energy, $\hbar\omega$, e.g. a constant electric field, having matrix elements $W_{12} = W_{21} \neq 0$ and $W_{11} = W_{22} = 0$. In fact in his calculation he assumes that one of the excited levels cannot decay to the ground state by a dipole transition, but we do not. He also considers the initial state of the atom to be known, whereas we do not assume this. Also the field in his case is a constant electric field and the effect considered is the Stark effect. In our case the field is time-dependent. For these three reasons his results cannot throw any light on our calculations.

In a later paper (ref. 75) Morozov considers the splitting of the resonance scattering line under STRONG monochromatic radiation for 2-level particles. He points out that this has already been investigated theoretically by Apanasevich(76,77), who, in ref. 76, solves the equation for the density matrix of the transformation under stationary conditions (open or non-closed system) and then in ref. 77 studies the shape of the scattering line of the 2-level particles as a function of the irradiation intensity on the basis of his solution. In contrast, Morozov(75) examines this question by solving the Schrodinger equation in the energy representation of a closed system of radiation field and the 2-level particle and obtains a line shape qualitatively different from the shape of refs. 76 and 77.

Morozov(75) considers the following initial conditions; at $t = 0$ the particle is in the ground state with energy E_0 and the radiation field contains s_λ photons of each type λ whose frequency distribution is defined by a symmetrical curve $I_0(\omega_\lambda)$ centred at $\omega_\lambda = \omega$ with halfwidth $\Delta\omega$ *. The energy of the excited state is E_1 and $E_1 - E_0 = \hbar\omega_0$

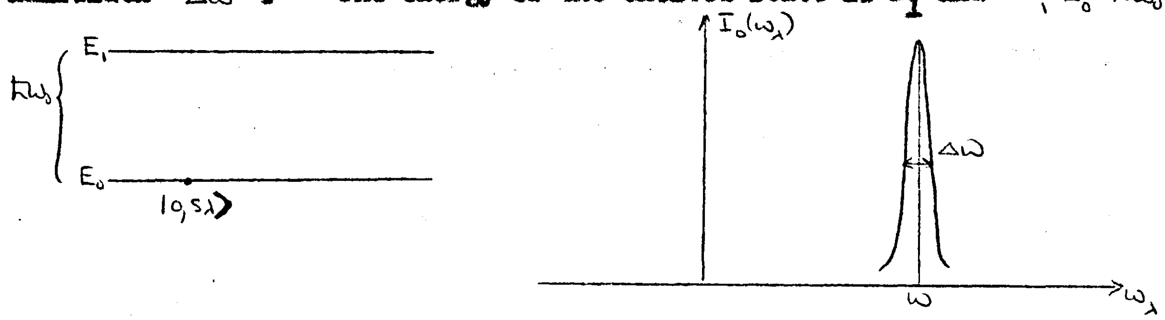


Fig. 7.C.6

He finds that the energy distribution density of scattered photons, the shape of the scattering line, which is expressed through the probability amplitude $b_{0\sigma}(\infty)$ (symbolically as in ref. 68) in the following way (see refs. 40 and 68):

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* We do not assume a specific initial state and we assume $I_0(\omega_\lambda)$ to be of negligible width.

$$I(\omega_\sigma) = \hbar\omega_\sigma \rho_{\omega_\sigma} |b_{0\sigma}(\infty)|^2$$

is given by

$$I(\omega_\sigma) = \frac{\hbar^{-1}\gamma_\sigma \omega_\sigma |(M_\lambda)_{10}|^2}{(\omega_\sigma-\omega_0)^2+(\gamma/2)^2}\int_{-\infty}^{\infty}\frac{I_0(\omega_\lambda)\,d\omega_\lambda}{[\omega_\sigma-\omega_\lambda-\frac{1}{2}\,\mathrm{Im}\,\Gamma(E_{0\sigma})]^2+[\frac{1}{2}\,\mathrm{Re}\,\Gamma(E_{0\sigma})]^2}$$

where $\hbar A$ is the interaction energy of the particle with the field and

$$(\hbar A)^2 = \sum_\lambda |V'_{0\lambda}|^2 = 2\pi |(M_\lambda)_{10}|^2 \int_{-\infty}^{\infty} I_0(\omega_\lambda)\,d\omega_\lambda = 2\pi|(M_\lambda)_{10}|^2 I_0$$

and $\sum_\sigma \gamma_\sigma = \gamma$ is the usual radiation damping of the excited state

and $\Gamma(E_{0\sigma}) = \frac{2i}{\hbar}\sum_\lambda \frac{|V'_{0\lambda}|^2}{E_{0\sigma}-E_1+i\hbar/2\,\gamma} = \frac{2iA^2}{\omega_\sigma-\omega_0+i\gamma/2}$

Thus when the interaction is WEAK $A\gamma^{-1} \ll \Delta\omega \ll \gamma$ or $A \lll \gamma$ and the radiation field has a narrow line distribution, this equation for $I(\omega_\sigma)$ reduces to equation 25 of ref. 68 except that the factor $\gamma_\sigma\gamma^{-1}$ is omitted there.

For a laser field, where number s_λ exceeds unity by many orders of magnitude, and therefore for $\Delta\omega \ll \gamma$ one has $I_0 \gtrsim \gamma\hbar$ and accordingly $A \gtrsim \gamma$ (strong interaction) the shape of the resonance scattering line is:

$$I(\omega_\sigma) = \hbar\omega_\sigma \frac{\gamma_\sigma}{2\pi} A^2 \left[\frac{(\omega_\sigma-\omega_0)^2+(\gamma/2)^2}{\{(\omega_\sigma-\omega_0)[(\omega_\sigma-\omega_0)^2+(\gamma/2)^2]-A^2(\omega_\sigma-\omega_0)\}^2+A^4(\gamma/2)^2}\right]$$

The shape of the resonance scattering line, according to this formula, for different values of A $(A=\gamma, 2\gamma)$ and ω $(\omega=\omega_0, \omega_0-\gamma/2, \omega_0-\gamma)$ is shown in the figure below:

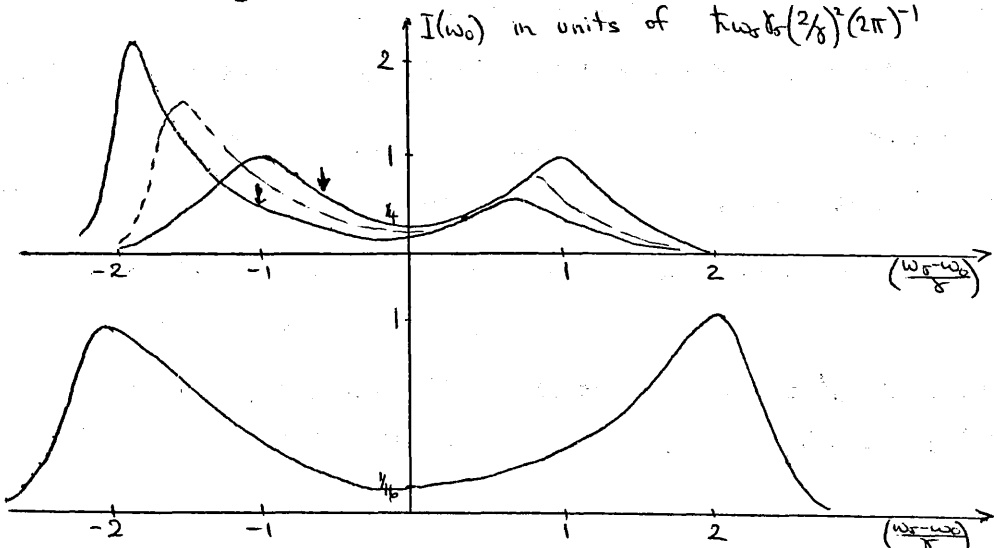


Fig. 7.C.7a $\boxed{A=\gamma}$

Fig. 7.C.7b $\boxed{A=2\gamma}$

(Arrows indicate excitation frequencies ω and simultaneously the corresponding shapes of the scattering line.)

From the figure it is clear that for strong interaction $A \geqslant \gamma$ the resonance scattering line is split into 2 components with the component located close to the exciting frequency having the higher intensity. At strict resonance, $\omega = \omega_0$, the intensities of the components and their distance from the exciting frequencies become equal but as the field becomes further from resonance so the components become less symmetrical, the intensity of the left hand component increasing and both components being shifted leftwards so that the distance of the left hand peak from the exciting frequency is increased and the left hand peak decreased. The distance between the maxima of the split components is roughly equal to 2A, as in Mollow's paper (ref. 9), i.e. the frequency of particle transition between levels, under the influence of a perturbation that depends on time as cos $\omega_0 t$. Autler and Townes (ref. 55) have predicted and experimentally observed analogous splitting of an absorption line corresponding to a transition between states, one of which is exposed to strong resonance excitation. We have seen also in ref. 33 a theoretical study of how the absorption line in a 2-level system is split under monochromatic resonance irradiation.

In ref. 77 the splitting of the scattering line has a different character, e.g. for $\omega = \omega_0$ (resonance) and $A \doteq 2\gamma$ the line has 3 weakly defined components (a central one and two lateral ones) with roughly equal intensities at the maxima. Three components were also found by Mollow[9] though he found the lateral ones to possess lower intensities. Morozov points out the discrepancy to be due to the nonequivalence of investigations (with respect to line shape) of a stationary transformation of radiation and a transformation in a closed system for $A \geqslant \gamma$, although as shown in ref. 69 these investigations are equivalent for weak interactions $A \lll \gamma$.

Ref. [75] thus predicts that, in our resonance situation, we should find the side peaks to be equally spaced about $\omega_\sigma - \omega_0 = 0$ and to be separated by a distance of 2A (or 4A in our notation). Also that, since our calculation is carried out for an open or non-closed system, i.e. under stationary conditions, we should expect a central peak also as in refs. 77 and 9. Modifications will no doubt appear because we consider an intermediate level.*

In ref. 73 Morozov considers the intensities and shapes of the components of the Stark splitting in e.m. resonance fields which are alternating. He treats the e.m. field quantum mechanically and points out that such a study, which ignores the widths of the lines due to collisions, is relevant when the widths of the levels is set primarily by the interaction with the radiation field as is the situation for gases in a good vacuum, particularly for the upper, optical transition levels. This is the situation we also consider in all our calculations. Morozov is particularly interested in the situation where $\hbar A >> \gamma$ i.e. the field is strong and uses the approximation that takes into account transformation of only one photon of the strong field into each transformed photon of the weak field. In the exact resonance situation the values obtained for the intensity and positions of the components agree well with the experimental results for $A \gtrsim \gamma$ but not for $A >> \gamma$ because, so he suggests, when $A >> \gamma$ one must also consider those processes in which a single transformed photon of the weak field is accompanied by more than one transformed photon of the strong field. This would be a higher order approximation, in quantum language. Although in this paper Morozov considers a 4-level atom his conclusions concerning the shape of the absorption line coincide with those of the previous paper [71], i.e. when $\gamma_2 = \gamma_{3\to1} = \gamma$ and $A \geqslant \gamma$ the line has 2 components and for exact resonance, $\omega_4 = \omega_{32}$, their intensities are equal, the separation of their maxima being again 2A etc. as shown in his fig. 2 for $A = 2\gamma$. Morozov's limitation to consideration of amplitudes of statesdiffering from the initial state by only one photon results from having to know the initial state of the atom and consider each transition seperately instead of

* In fact our results for the case of an intermediate overlapping level are similar to those for the 2-level atom off resonance and as c decreases we get similar results to those shown in ref. 75 for the case where the atom goes increasingly off-resonance.

considering overall effects, as we do, by considering the initial time to be arbitrary.

The last paper of Morozov we shall look at, in connection with our results, is ref. 79 on the theory of the line shape of resonance scattering of strong radiation, $A \gtrsim \gamma$, by a 2-level particle. In ref. 75, each photon is considered to be scattered independently by the particle so that the line shape can be represented as a superposition of the distributions obtained. But if one supposes that conversion of photons of a strong field is a single process of the type, e.g.

$$0\, s_\lambda \to 1, (s-1)_\lambda \to 0, (s-1)_\lambda\, \sigma_1 \to 1, (s-2)_\lambda\, \sigma_1 \to 0\, (s-2)_\lambda\, \sigma_1 \sigma_2 \to \quad \to 0\, \sigma_1 \sigma_2 \ldots \sigma_s$$

where λ and σ are photon indices and $\lambda \neq \sigma$

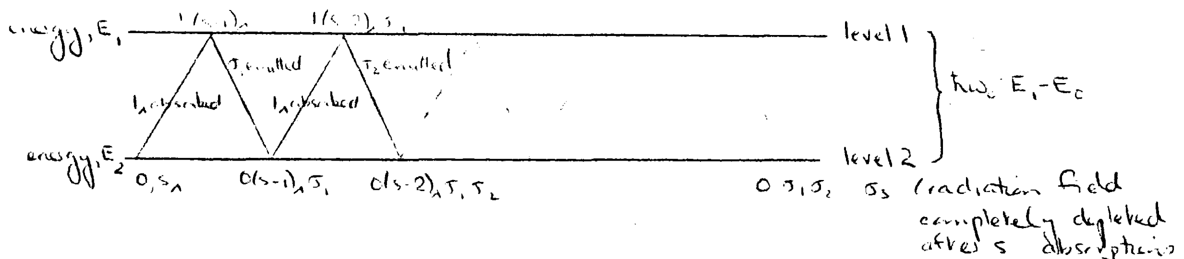
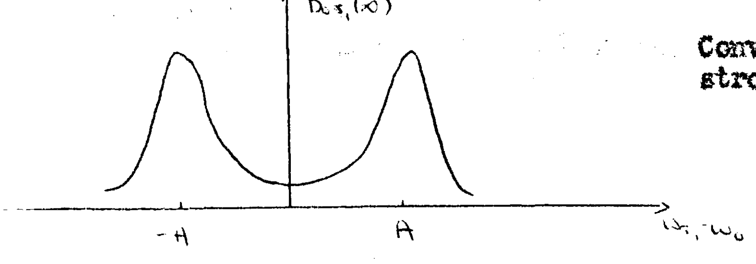


Fig. 7C.8

then a different analysis is required to find the line shape of the scattered radiation. Morozov studies the frequency distribution of photons scattered as a result of transitions of this type since he considers that such processes are of vital importance in the scattering of strong radiation.

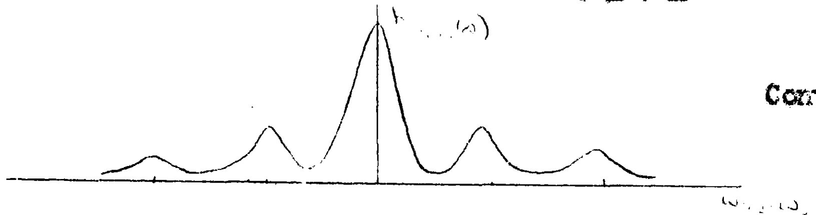
He considers a single mode field with $\omega_\lambda = \omega_0$ (strict resonance) and calculates the probability amplitude $b_{0\sigma_1\sigma_2 \ldots \sigma_s}(\omega)$, which is the solution of the Schrodinger equation (in the energy representation) at t = ∞ assuming the atom is in the ground state at t = 0 and the radiation field contains s photons of type λ. He uses the Heitler-Ma method of ref. 40 to find $b_{0\sigma_1\sigma_2 \ldots \sigma_s}(\omega)$ for $\omega \gg \gamma$ and $A_s \gtrsim \gamma$ and concludes that for conversion of 2 photons of a strong field, instead of having 2 components of equal intensities at distances A and -A from ω_0 as in ref. 75:-



Conversion of 1 photon of a strong field

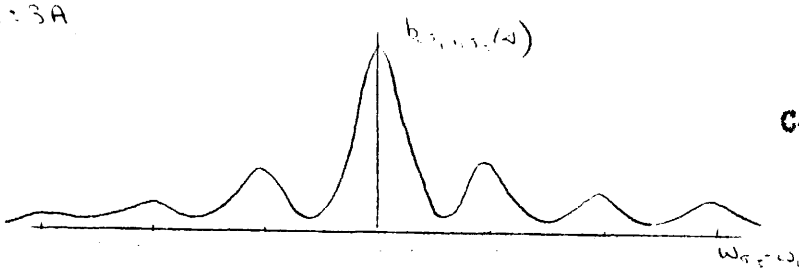
Fig 7.C.9

we have a distribution containing broadened components (approximately twice that of ref. 40) maxima at $\omega_{s_2} - \omega_0 = 0, \pm A, \pm 2A$.



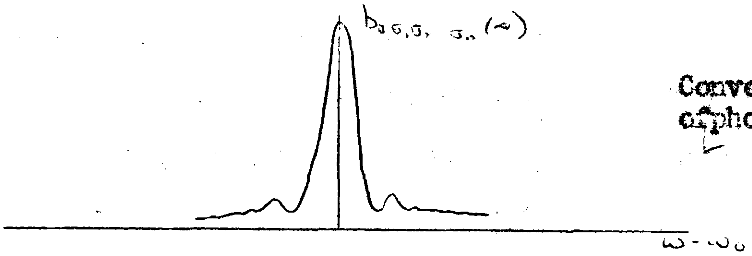
Conversion of 2 photons
Fig 7C10

For conversion of 3 photons there will be components at ω_0, $\omega_0 \pm A$, $\omega_0 \pm 2A$, $\omega_0 \pm 3A$



Conversion of 3 photons
Fig 7C11

As the number of converted photons increases so the components merge and generate a continuous curve. The intensity of the centre of the line increases more rapidly than in the tails. Thus after a large number of conversions by a single process, the frequency distribution of scattered resonance will be a narrow line at $\omega = \omega_0$.



Conversion of a large number of photons
Fig 7C12

(N.B. Although our results should show the overall effects of conversion of a large number of photons since the initial state is arbitrary our results ∴ indicate that our calculation effectively considers only the conversion of ONE photon of the strong field.)

As we have already mentioned Mollow[9] calculates the power spectrum of light scattered by 2-level systems. When the incident field is weak so that $\Omega \ll K$ then, when the field is off-resonance, the integrated spectral intensity of the incoherent part of the scattered field is only a very small fraction $\left[\frac{\frac{1}{4}\Omega^2}{(\frac{1}{4}K^2 + (\Delta\omega)^2)}\right]$ of the intensity of the coherently scattered field and when $|\Delta\omega| \gg K$ the incoherent part of the power spectrum is sharply peaked at the 2 displaced frequencies $\omega-\Delta\omega=\omega_0$ and $\omega+\Delta\omega=\omega_0+2\Delta\omega$

When the field is intense enough so that $\Omega >> K$ and the field is off resonance then the function $\bar{g}(\nu)$ may be approximated in the domain in which it is appreciable by a superposition of Lorentzian functions at each of its maxima at

$$\nu = \omega$$
$$\nu = \omega + \Omega'$$
$$\nu = \omega - \Omega'$$

where
$$\Omega' = \left[\Omega^2 + (\Delta\omega)^2\right]^{1/2}$$

When the field is very intense so that $\Omega >> |\Delta\omega|, K$ then maxima occur at $\nu = \omega \pm \Omega$ and $\nu = \Omega$ and the 2 side peaks have maxima of one-third of the maximum at $\nu = \Omega$, the integrated spectral intensity being one-half of that at the central frequency. The intensity of the coherent classically scattered light in this limit is inversely proportional to the incident field intensity, and is only a very small fraction of the total scattering intensity. We expect that in our resonant case $|\Delta\omega| = 0$, we should find similar results only modified by the existence of level 2.

In ref. 80, Mollow calculates the power spectrum of the radiation emitted when the driven 2-level system is collision damped and the collisions are assumed to be strong, i.e. to instantaneously thermalise the state of the atom. He considers the cases of low and high excitation of the atomic system In his calculations he assumes the incident field to oscillate at a fixed frequency ω, near to the atomic resonance frequency ω_0 and to have arbitrarily great intensity. He also assumes that the collision rate, K, (previously used to designate the spontaneous emission rate) is very much greater than all other relaxation rates, e.g. the radiative decay rate, the effect of which was analysed in ref. 9. He finds that for weak driving fields the results differ markedly from the case of radiative relaxation. In the collision case, the profile contains both a coherent monochromatic spectral component oscillating at ω and incoherent components oscillating within an interval κ of ω_0. The incoherent components are appreciable even at low temperatures and equal the intensity of the coherent component in the zero-temp. limit. For strong driving fields, on the other hand, the solutions for the radiative and collisional cases resemble each other quite strongly. In both cases there are three components, one centred at ω and one at each of the displaced frequencies $\omega \pm \Omega$ where Ω is the frequency of the field-induced atomic transitions. In his calculation he considers that the atom is driven only by a classical electric field and includes no coupling to an e.m. radiation field in the Hamiltonian as we do when considering radiative relaxation.

In ref. 53, Mollow considers the effect of a driving or pump field on the emission and absorption line-shape functions separately, for collisional and general relaxation when the field is applied to the atom at a frequency near resonance for atomic transitions between a particular pair of states. He considers transitions between pairs of states, only one of which is a member of the resonantly coupled pair, as we do in the chapters on potassium and hydrogen and shows that the driving field affects these transitions also. The absorption line-shape function is defined as the rate of absorption from a weak signal field applied in addition to the pump field. He finds that, in the limit of high pump-field intensity, both absorption and emission spectra are doubly peaked at frequencies differing from the usual resonance frequency by $\pm\frac{1}{2}\Omega$ (- Ω is the frequency of the pump-field-induced oscillations in the populations of the two strongly coupled states). For high and vanishing pump-field intensity, the absorption and emission spectra are represented by essentially the same function but for intermediate intensities the two functions have quite different forms and there is no simple proportionality between them. The difference between the emission and absorption spectra, which emerges directly from the basic method of evaluating the associated correlation functions in the Markoff approximation, is due to the presence of the off-diagonal states which are strongly coupled by the pump field. Although we do not calculate emission and absorption line-shape functions separately, it is interesting to note the effect of the field's intensity on them.

Mollow(53) also draws attention to the fact that Lehmberg has criticized the use of the familiar form of atomic relaxation theory for the case of coupling to "soft" (low frequency) photon or phonon modes when relaxation occurs in the presence of very strong driving fields and has proposed a more complicated theory in ref. 81. On the other hand, Mollow does claim that the equations of motion for the elements of the 2 x 2 density submatrix elements referring to the pair of strongly coupled states $|0\rangle$ and $|1\rangle$ remain valid even when, e.g. the coupling between the states $|0\rangle$ and $|1\rangle$, but not between either of these states and the other weakly coupled states of the atom, is described by means of more general forms of relaxation theory.

In ref. 82, an extension of ref. 80, Mollow goes on to consider the absorption spectrum corresponding to transitions from one of the resonantly coupled states to the other. In this paper, the signal field also has a frequency near to the atomic resonance frequency for the transition in question, whereas in ref. 80 the signal frequency is well separated from the pump frequency and so can induce transitions between pairs of states of which only one is a member of the pair resonantly coupled by the pump field. In fact, in all, he considers both stimulated emission and absorption, near resonance, for driven systems. He finds that, even though population inversion does not occur, stimulated emission rather than absorption, is indicated by the negative values of the signal-field absorption line-shape function. This amplification of the signal is most pronounced at high pump intensities exactly on resonance and he shows it to occur primarily at the expense of the pump field, which suffers an increased rate of attentuation. Mollow's results can be extended to more general pumping mechanisms.

In the present chapter, we are considering similar transitions to those of ref. 82 and so for high pump field intensities, i.e. A large, we should expect the profiles to indicate stimulated emission although we do not explicitly consider any extra signal field. In fact, at the end of his paper, Mollow points out that the signal field plays no direct role in his more general treatment of the problem, which gives the same result so he assumes that the results should apply wherever the two coupled levels are driven by a suitable pumping mechanism.

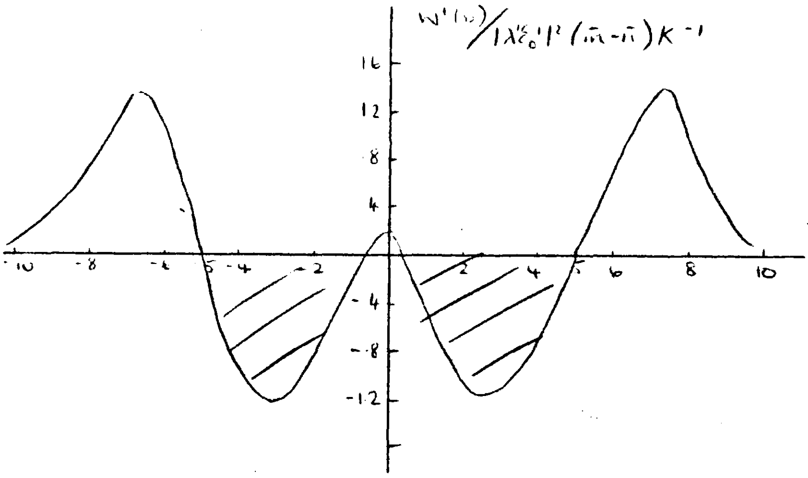


Fig. 7C.13

Signal-field absorption line-shape function for an atom driven exactly on resonance by a pump field for which = 5K. The negative values of the absorption function (shaded area) represent stimulated emission, i.e. amplification of the signal field.

ν - signal-field frequency, ω - pump-field frequency

κ - mean collision rate or spontaneous emission rate depending on whether considering the case of the strong collision model of atomic relaxation [83] or (zero-temp.) radiative relaxation [9].

N.B. For $|\Delta\nu| > \Omega$, i.e. in the case above, for $(\nu-\omega)/\kappa > 5$, the signal field is attenuated instead of the pump field and the rate of attentuation of the pump field is correspondingly reduced.

The absorption line-shape function of ref. 82 is quite different from that for the emission spectrum evaluated in refs. 9 and 80 and the difference is more pronounced when the pump field is intense since then the former line shape takes on appreciable negative values, within a wide range of signal-field frequencies.

The possibility of obtaining stimulated emission without inversion has also been suggested by Sharma in ref. 84. He shows that in a 2 photon emission at frequencies ω and ω', amplification at frequency can be achieved without population inversion if (i) $\omega<\omega'$ and N', the number of black body photons at frequency ω', is much smaller than unity. He points out that if the squares of the dipole matrix elements are approximately equal, $d_{13}^2 \approx d_{23}^2$, then stimulated inversion is possible. In fact his situation does not correspond to ours as we do not allow levels 3 and 2 to be radiatively connected nor do we forbid single photon transitions between levels 1 and 2.

In ref. 87 the authors consider spontaneous emission of atoms in an external e.m. field which is considered to be either monochromatic or to have a large Doppler broadening. The superposition of the external field leads to a deformation of the line contour, which can be interpreted as the consequence of (i) a charge in the velocity distribution of the atoms, and (ii) of interference effects arising in the mixing of stationary states of the isolated atom by the external field. Under certain conditions, the second factor completely counterbalances the effect of the first.

In ref. 33 Rautian and Sobelman considered the atom to be fixed and the external field to be monochromatic, whereas in ref. 86 a number of specific cases were analysed in which account was taken of the motion of the atoms.

Kotkin, Rautian and Feoktistov[87] point out that normally Doppler broadening >> radiation broadening and almost completely masks it but that when an external e.m. field is applied, a relatively sharp structure appears on the Doppler-broadened line, the width of which is determined by the radiative decay. The changes produced in the spectrum of spontaneous emission of a quantum system by the external field can be interpreted as the result of mixing of the stationary states of the isolated atom in the external field. Since unusual interference phenomena appear, discussions based on the picture of "splitting" of the levels of an isolated atom can lead to a seriously wrong conclusion. This applies for both adjacent transitions considered in ref. 87 as well as to the spontaneous emission by the transition m -> n (i.e. between levels coupled by the field considered in refs. 33 and 86.

Similar interference effects occur, to a known extent, in these cases when, by selection of processes of excitation the states of the atom at the initial instant of time and "prepared" in the form of a linear combination of stationary states. N. R. and F. are interested in the case distinguished by the fact that mixing of states takes place after the excitation of the system while at the initial instant the atom is in some stationary state. In ref. 87 collisional relaxation has been ignored.

In ref. 88 Rautian and Feoktistov go on to take into account collisions, in the impact approximation, when considering nonlinear effects in the spontaneous emission. They find that changes in the spontaneous emission spectrum are due to (1) changes in the atom velocity distribution, (2) the characteristics of the collision process, and (3) interference effects due to the presence of an external field that intermixes the stationary states of the isolated atoms. They point out that in refs. 33, 86, 87, where the atom is at rest, the external monochromatic field leads to line splitting if the energy of interaction between the atom and the field exceeds the level width, i.e. in our case we must have $\underline{A > \Gamma}$ as pointed out in part (a). Also when account is taken of the atoms' motion, and of the corresponding Doppler broadening, a relatively sharp line structure also appears (with radiation width), not masked by the thermal motion of the atoms.

The effects of refs 33, 86, 87 have been experimentally observed[89,90] in gas systems but, since the line structure due to the external field depends appreciably on gas pressure also, it appears that it is very sensitive to atom collisions and these must be taken into account in the theory as do the authors of ref. 88. This is not necessary for gases at low pressure.

In ref. 91, Dodd and Series give a theory of the phenomenon of the strong modulation of fluorescent light in a double resonance experiment. In such an experiment free atoms are subjected simultaneously to optical and radio-frequency radiations, both of which are near to resonant frequencies of the atoms. They consider the case of mercury vapour situated in a uniform magnetic field, when optical radiation at 2537 Å is used to excite the atoms from the ground state, $^1S_0$, to the state m = 0 of the level $3P_1$, from which transitions to m = ±1 were induced by a radio-frequency field at the Larmor frequency. We do not consider the presence of such a r.f. field but rather assume that in the 3-level atom in question the optical radiation excites the atom to the uppermost level, which could be the m = $\pm\frac{1}{2}$ level of a Zeeman doublet, and that it then decays from this level. Hence there is no need for a r.f. field as in the case where there is a Zeeman triplet.

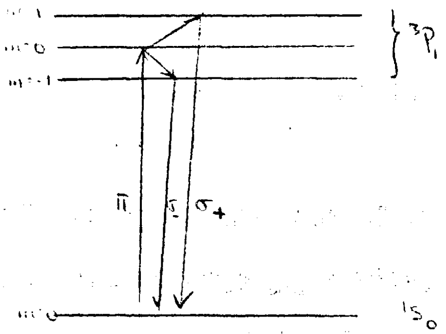


Fig 7 C.14

N.B. There is a change in the polarisation of the light resulting from r.f. transitions $\Delta m = 0$ & $-1, +1$. For observations made in a direction perpendicular to the field, the lines with $\Delta m = 0$ are polarised parallel to the field (π- components) and the lines with $\Delta m = \pm 1$ are polarised perpendicular to the field (σ- components).

The transitions are detected by the changes which they bring about in the polarisation and spatial distribution of the fluorescent light.

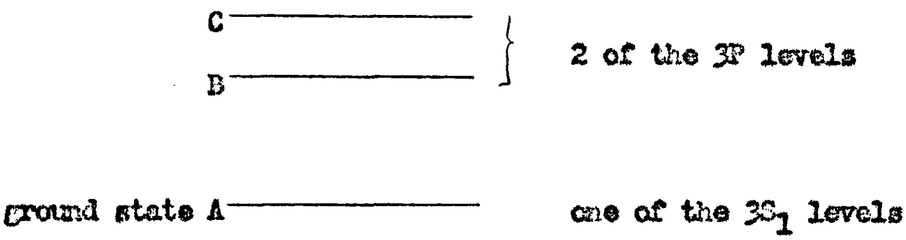
Dodd and Series carry out calculations with particular reference to the transition ($6^3P_1 - 6^1S_0$), λ 2537 Å in mercury, where the radio-frequency mixing is taking place between the Zeeman states of the upper level. In this case the 3 excited states are equally spaced in energy and are damped at the same rate and there is only one ground state. In ref. 92, Series proposes a type of experiment which would allow the location of crossing points between energy levels of different parity, and thus provide an alternative method for measuring the Lamb shift. In the previous paper[91] the states concerned were members of a Zeeman multiplet and so were of the same parity and decayed at the same rate. In the present paper they are of different parity and decay at different rates as e.g. the $2^2S_{1/2}$ and $2^2P_{1/2}$ states in hydrogen. Whether they are of different or the same parity, the application of oscillating fields is capable of inducing transitions between the states of interest, thus causing changes in the emitted radiation. When the states are of the same parity the transitions are magnetic dipole and the effect of these transitions is primarily to alter the spatial distribution of the radiation whereas the spectral distribution is changed very little. When the states are of different parity the transitions are electric dipole and because of the change in parity the atom must decay to an entirely different term and then the spectral distribution changes profoundly.

Series[92] analyses the case of an excited pair of energy levels belonging to states of opposite parity, having in mind any pair of intersecting s and p levels in hydrogen-like atoms. In our case transitions can occur from both excited levels to the lower level and not only from one of the excited levels. Hence the results of this paper are not really relevant to the present chapter nor are they relevant to Chapter VI since the driving field in that case couples different levels, i.e. levels $2S_{\frac{1}{2}}$ and 3P and not $2S_{\frac{1}{2}}$ or $2P_{\frac{1}{2}}$ and $1S_{\frac{1}{2}}$, as here. We are also not concerned with the possibility of level crossing.

In ref. 93 the authors are concerned with the interference phenomena that can occur in the resonance fluorescence of an atom in which 2 of the excited Zeeman sublevels cross. They consider the particular case of the helium atom in which 2 of the 3P Zeeman sublevels cross. This leads to the idea of a new spectroscopic method which promises to yield very precise measurements of some atomic fine structure intervals. If the levels are all distinct (i.e. separations greater than their natural widths), then each contributes separately to the resonance scattering. However, when two levels are degenerate, their contributions to the scattering may interfere thus giving rise to a change in scattered light. In our case this would be when $c \leq 1$.

Colegrove et al[93] consider a system having one ground state A and 2 excited states B and C:

Fig 7C.15



In order to calculate the resonance fluorescence of this system, it is necessary to first calculate the rate at which photons of polarisation e_1 are absorbed and photons of polarisation e_2 are re-emitted. When levels B and C are well resolved i.e. their separation > their natural line widths (in our case 1) then the rate is given by

$$R \sim |\langle A|e_2 . r|B\rangle\langle B|e_1 . r|A\rangle|^2 + |\langle A|e_2 . r|C\rangle\langle C|e_1 . r|A\rangle|^2$$

whereas when B and C have the same energy i.e. are completely unresolved or crossed (in our case = 0)

$$R \sim |\{\langle A|e_2 . r|B\rangle\langle B|e_1 . r|A\rangle + \langle A|e_2 . r|C\rangle\langle C|e_1 . r|A\rangle\}|^2$$

In order that these two equations should yield different results both terms in each expression must be nonvanishing, i.e. each of levels B and C must be capable of sharing photons of polarisation e_1 and e_2, as when the line-widths of levels B and C overlap. In fact we must have

$$\langle A|e_2 . r|B\rangle\langle B|e_1 . r|A\rangle\langle A|e_2 . r|C\rangle\langle C|e_1 . r|A\rangle \neq 0$$

otherwise the interference effect vanishes. For unpolarised light one finds that the 2 degenerate levels must differ in m by 0 or 2 in order for there to be an interference term. When the incident light is linearly polarised one can observe the crossover of levels differing in m by 1 provided the direction of polarisation is not parallel or perpendicular to the magnetic field. Levels differing in m by 3 or more do not interfere. In fact, if one sums over the directions of e_2 the polarisation of the re-emitted light, then it can be shown that the total resonance scattering rate is not affected by the crossover of the 2 levels but the angular distribution is changed.

In ref. 94 a detailed analysis of the effects briefly described in ref. 93 is given.

We have not considered the possibility of sublevel crossing in the present chapter but rather that of overlapping of line-widths. At the crossing point levels behave as though they are of the same energy as in the case when c = C.

The 2 rate formulas of refs. 93 and 94 are sufficient for calculation of many experimental parameters e.g. magnitude of the interference effect, its directional sensitivity, polarisation conditions for optimum sensitivity etc. (for a more general formula for the angular dependence of the interference effects derived by a detailed analysis of the interference effects to be expected in atomic hydrogen one must see a publication of M.E. Rose and R. Carovillano referred to in ref. 94). The expressions for R in refs. 93 and 94 do not give any information about the interference other than at its extremes, i.e. at complete crossing and at complete separation of the excited states. A more detailed treatment of the resonance fluorescence process that yields information about the line shape as well as the equations for R in the appropriate limits is required for the in-between region. We have endeavoured to study this region, i.e. where $0 < C < 1$ for a driving field of arbitrary strength indicated by A.

In ref. 97 Breit has derived an expression for the resonance fluorescence under pulse excitation for atoms exhibiting partial or complete degeneracy (crossing) in the excited states. The Breit formula is applicable only to weak fields. For conventional sources Franken[(94)] points out that the light beam consists of much less than one photon per mode in the appropriate frequency interval and must be considered as a weak field whereas in all strong beams of resonance radiation the number of photons per mode in the appropriate range is $\sim 10^4$. Thus the rate of absorption of resonance radiation per atom in such strong beams is some 4 orders of magnitude less than the rate of spontaneous emission. The derivation of Breit's formula in Appendix II of ref. 94 does treat the radiation field classically but none the less gives the correct weak-field result.

In ref. 94 the rate at which photons of polarisation $\underline{f}$ are absorbed and those of polarisation $\underline{g}$ are re-emitted in the resonance fluorescence process is:

$$R(\underline{f},\underline{g}) = c \sum_{\mu\mu' m m'} \frac{f_{\mu m} f_{m\mu'} g_{\mu' m'} g_{m'\mu}}{1 - 2\pi i \tau \nu(\mu,\mu')}$$

where $f_{\mu m} = \langle \mu | \underline{f} . \underline{r} | m \rangle$, etc.;

τ is the mean lifetime of each excited state,

$\nu(\mu,\mu') = (E_\mu - E_{\mu'})/\hbar$;

c is a parameter proportional to the intensity of the lamp, geometrical factors etc.

m, m' etc. are the ground state levels

μ, μ' etc. are the group of excited states which may exhibit partial or complete degeneracy (crossings)

When the excited states are completely resolved, $2\pi\tau\nu(\mu,\mu') >> 1$ or in our case $>>$ 1 for all values of $\mu \neq \mu'$ and the above general equation reduces to

$$R(\underline{f},\underline{g}) = R_0 = c \sum_{\mu m m'} |f_{\mu m}|^2 |g_{\mu m'}|^2$$

which is the resonance fluorescence rate without interference terms as shown in ref. 93. When $2\pi\tau\nu(\mu,\mu') \lesssim 1$ i.e. in our case $c \lesssim$ 1 then 2 of the excited states are close enough together for interference effects to occur.

If the system contains only one ground state a and 2 excited states b and c:

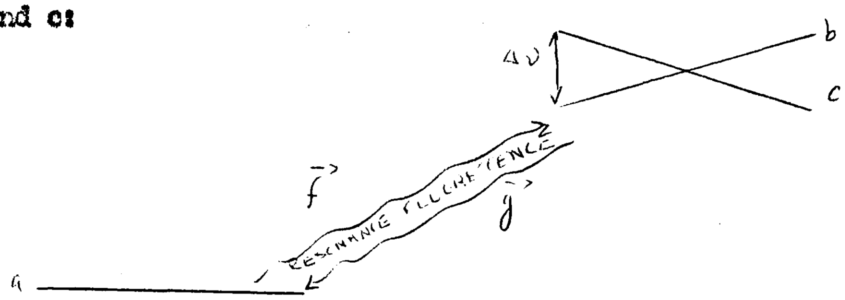


Fig 7.C.16

then when b and c are well resolved

$$R(f,g) = R_0 = |f_{ab}|^2 |f_{ba}|^2 + |g_{ac}|^2 |g_{ca}|^2$$

and when they are "close"

$$R(f,g) = R_0 + \frac{A}{1-2\pi i\tau\nu(b,c)} + \frac{A^*}{1+2\pi i\tau\nu(b,c)} \equiv R + S$$

where $A = f_{bc} f_{ac} g_{ca} g_{ab}$

and S is also given by

$$S = \frac{A+A^*}{1+4\pi^2\tau^2\nu^2(b,c)} + \frac{(A-A^*)\,2\pi i\tau\nu(b,c)}{1+4\pi^2\tau^2\nu^2(b,c)}$$

When the matrix product A is real then S is the well-known Lorentz line-shape with full half-width which is twice the width of each excited state.

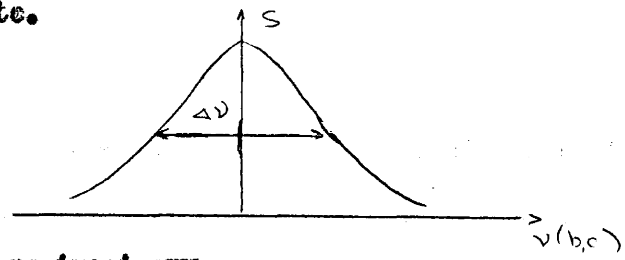
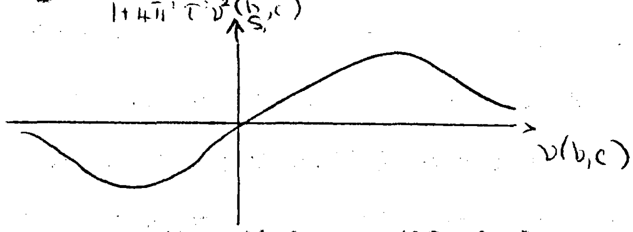


Fig 7.C.17

When A is pure imaginary

Fig. 7.C.18

$$S = \frac{4\pi i A\tau\nu(b,c)}{1+4\pi^2\tau^2\nu^2(b,c)}$$



If A is complex, then it is possible to have a mixture of these 2 pure forms. The conditions for which A is real, imaginary, or complex depends on the direction and polarisation of the incoming and outgoing beams of light. In general all three cases can be realised experimentally.

Wangness also considers a situation similar to our case. In Ref. 95 he investigates the effect of externally produced Stark splitting upon fine-structure-level probabilities in order to describe periodic intensity variations previously found in hydrogen lines. Coupled differential equations for the probability amplitudes of mixed levels are solved exactly in terms of initial amplitudes; effects due to spontaneous transition probabilities and Lamb shift are included. In general, the time-dependent line intensity is a combination of exponential decays and sinusoidal oscillations as in Chapter II. The frequencies of oscillations are discussed in detail, and the effects of different initial conditions upon the intensities are considered for several special cases.

In this paper Wangness considers levels 0, 1, 2 where levels 1 and 2 can decay to lower level 0 with spontaneous transitionsprobabilities per unit time A_1 and A_2. Levels 1 and 2 have energies E_1 and $E_2 = E_1 + \hbar\delta$. $\hbar\delta$ is assumed to be so small that the separate spectral lines cannot be resolved ($c \lesssim$ 1 in our case). The observed intensity I of the line will then be proportional to the sum of the products of the A's and the a verage probabilities of occupations of the levels, i.e.

$$I = A_1 \langle |C_1|^2 \rangle_{av.} + A_2 \langle |C_2|^2 \rangle_{av.}$$

where C_1 and C_2 are the probability amplitudes of the 2 levels and the constant multiplicative factor has been omitted.

Wangness also assumes that in the presence of a small external electric field F, the perturbation potential $V = eFz$ $(e > 0)$ has a non-vanishing matrix element $V_{12} = V_{21} = \hbar v$ connecting these 2 levels. The resulting equations for C_1 and C_2 are given by his equations (9)-(13). In this chapter the perturbing potential connects levels 0 and 2, not 1 and 2.

To illustrate the effect of mixing produced by the field one can consider the extreme case in which one of the initial c's is zero, e.g. $C_{20} = 0$ so that

$$|C_2(t)|^2 = |C_{10}|^2 \left[\frac{v^2}{\omega_r^2 + \omega_i^2} \right] (\sinh^2 \omega_i t + \sin^2 \omega_r t) \exp\left[-\tfrac{1}{2}(A_1 + A_2)t\right]$$

which vanishes in the absence of the field so that $v = 0$. ω_r and ω are defined by equations (11), (12), (13). The oscillating term has an exponential decay factor equal to the average of those associated with the 2 states, while the $\sin \omega t$ term gives rise to 3 terms with exponential decay factors of $\frac{1}{2}(A_1 + A_2)$ and $\frac{1}{2}(A_1 + A_2) \pm 2\omega_r$, although the last 2 terms will not appear if $A_1 = A_2$ so that $\omega_r = 0$.

The problem discussed by Series in ref. 92 is similar although less general.

Stroud[98] considers the effect of an "applied field" on the line shape of spontaneous emission by a "2-level atom" interacting with a highly excited field mode. Despite the fact that he uses quantum electrodynamics, without time-dependent perturbation theory, he obtains spectral profiles similar to those of Mollow[9] (see Fig. 3 where the fluorescent spectrum is given for various applied fields when the Lamb shift is neglected). See Chapter VIII, Sec. 9 for a comment on Stroud's method.

(e) Analysis of profiles obtained

We shall now proceed to analyse the shapes of the spectral profiles. If we first of all tabulate the positions and heights of the peaks for various values of A and C we can see more clearly what is happening.

A = 1	1st L.H. peak		2nd L.H. peak		Central	1st R.H. peak		2nd R.H. peak	
C	Position	Height	Position	Height	peak	Position	Height	Position	Height
0	-1.4	.771			0.752			1.4	.771
0.01	-1.4	.769			0.752			1.4	.772
0.1	-1.6	.759			0.743			1.4	.761
0.5	-1.6	.501			.530			1.6	.573
0.75	-1.8	.162			0.282			1.8	.215
1.0									
1.25	-2.0	.298			0.238			1.8	.0877
1.5	-2.0	.476			0.753			1.8	.230
2.0	-2.2	.455			1.11			1.6	.389
3.0	-1.6	.441			1.27			2.0	.496
								5.2	.00580
5.0	-1.8	.456			1.33			1.8	.390
10.0	-1.8	.428			1.36			1.8	.400
100.0	-1.8	.414			1.36			1.8	.411

A = 2	1st L.H. peak		2nd L.H. peak		Central	1st R.H. peak		2nd R.H. peak	
C	Position	Height	Position	Height	peak	Position	Height	Position	Height
0	-3.6	.435			1.06			3.6	.435
0.01	-3.6	.435			1.06			3.6	.434
0.1	-3.6	.437			1.06			3.6	.433
0.5	-3.6	.456			1.04			3.6	.433
0.75	-3.6	.477			1.00			3.6	.437
1.0	-3.6	.498			0.931			3.6	.439
1.25	-3.6	.476			0.801			3.6	.411
1.5	-3.8	.377			0.563			3.8	.320
2.0									
3.0	-4.2	.437			0.967			4.0	.305
5.0	-4.0	.309			1.09			3.8	.344
10.0	-3.8	.367			1.11			3.8	.348
100.0	-3.8	.356			1.12			3.8	.354
A = 3									
0	-5.8	.371			1.06				.371
0.01	-5.8	.371			1.06				.371
0.1	-5.8	.372			1.06				.371
0.5	-5.8	.375			1.05				.370
0.75	-6.0	.379			1.05	2.8	.0654		.370
1.0	-5.6	.388			1.04	2.6	.0700		.371
1.25	-5.6	.400			1.03	2.2	.0737		.374
1.5	-5.6	.416			1.01				.380
2.0	-5.6	.457			.912				.389
2.5	-5.8	.370			.556				.304
2.8	-6.0	.105			.143				.0856
2.9	-6.0	.0297	-0.4	.0131	.0408				.0225
3.0									
3.1	-5.8	.0504			.0430				.0256
3.2	-6.0	.153			.169				.0851
3.3	-6.0	.256			.339				.149
3.5	-6.2	.374			.642				.244
4.0	-6.2	.432			.923				.313
4.5	-6.2	.384			.985				.321
5.0	-6.2	.355			1.01				.327
	→ -6.0								
6.0	-6.0	.340			1.03				.331
7.0	-6.0	.326	-3.4	.0851	1.04				.333
8.0	-6.0	.294			1.04				.336
10.0	-5.8	.338			1.05				.343
100.0	-6.0	.341			1.06				.340

<u>A = 100</u>

Side peaks are consistently at B = ±200 and are of height .333, the central peak being of height 1.00. At C = 100 there is no spectral profile.

If we first of all look at the results for the case when A is large, viz. A = 100, we see that the split levels of level 3 are so far from level 2, for most values of C, that the latter has no effect on them except when $C \sim A$ i.e. C is also large.

In the case of A = 3, the side peaks are more or less symmetrically spaced about B = ±2A, as predicted and this was also found for the case when A = 100. However, their intensities vary with C, so that, although at C = 0, 0.01 both are of the same intensity, as C increases the L.H. peak dominates over the R.H. one until at C = 7 the L.H. one dominates and continues to do so until at C = 100 they are both practically equal again. The central peak dominates throughout except at C = 3.1, when the L.H. peak dominates. At A = C = 3 no spectral profile is visible, as was the case when A = C = 100. For C = .75, 1, 1.25 a second R.H. peak is visible, though of very low intensity and for C = 2.9, 7.0 a second L.H. peak is visible, also of low intensity. We may also note that L.H. and R.H. peaks do not steadily increase or decrease but rather waver up and down, although the overall dominance of L.H. over R.H. becomes increasingly apparent as C   Although both fall off near C = A and the change over of dominance becomes apparent for C > A until both are again equal at C = 100. The central peak height decreases as $C \to A$ and increases as $C \to 100$.

When C = 0, $\omega_3 = 0$ and we have the 2-level or degenerate situation and for this reason we find that the profiles resemble those of Mollow[9] in that they have side-peaks of equal height, situated at $\sim \pm 2A$ (compare Mollow's curves for $\Omega = 3K, 5K$ with that for A = 3, C = 0).

When $C \gg 1$ and $\gg A$, i.e. $\omega_3 \gg \gamma$ and $\gg \lambda\mathcal{E}_0$, and so one would not expect level 2 to affect the profile for transitions between levels 3 and 1. It only has an effect when $A \sim C$. In fact, for $C \gg 1$, the profile is, for this reason, similar to that for $C = 0$.

Had we considered the profile for transitions between levels 2 and 1 for the same A we should have found that the R.H. gradually becomes dominant as $C \longrightarrow A$.

Now for the case when $A = 2$ the L.H. peak again becomes increasingly dominant as $C \longrightarrow A$ though with the usual fall off around $C = A$ and for $C > A$ the L.H. peak remains dominant at $C = 3,10$ but the R.H. dominates at $C = 5$ only and at $C = 100$ both are practically equal. Thus there is a smaller region, i.e. around $C = 5$, where the R.H. peak dominates. The central peak follows the same pattern as before and is always dominant. No profile is again obtained for $A = C$ (=2).

In the case of $A = 1$ the R.H. peak dominates as $C \longrightarrow A$, though all peaks fall off near $C = A$.

For $C > A$ the L.H. peak becomes increasingly dominant as level 2 approaches the low frequency split-level of level 3 and reinforces it, except for $C = 3$ when R.H. peak dominates. The rest of the features are the same as previously described.

The deviation from the curves found by Mollow in ref. 9 is due solely to the introduction of terms involving Γ_{31} and Γ_{21}, i.e. decay constants $\propto$ $P_{31} \cdot P_{21}$, which may be termed "cross-type" decay constants. We can see this by putting $\Gamma_{31} = \Gamma_{21} = 0$ in equations (7.B.2)-(10) and also ignoring level 2, since it is now not assumed to be close to level 3. We then recover Mollow's equations (ref. 9) and obtain his profiles.

For certain values of A and C there is a substantial difference between our curves and those of Mollow(9). In these cases we assume that the contribution of the "cross-type" decay constants is significant. It does not appear as though anyone except Morozov, and Apanasevich(34), who have both considered only WEAK fields, has yet evaluated the contribution of these cross-type terms in the case when the field can have arbitrary magnitude. We have found that their contributions are particularly significant when $C \sim A$, when level 2 lies between level 3 and level 3', the low-frequency split level of level 3':-

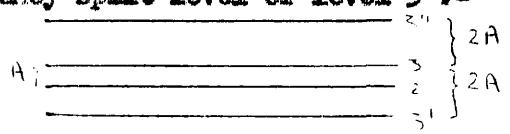


Fig 7 C 19

The phenomenon of no emission when $C = A$ requires verification by experiment as do the other features of the profiles.

D. Some notes on the assumption that $\Gamma_{31} \doteq \Gamma_{21} (=\Gamma)$ and its validity for the range of values of a and c chosen for plotting the intensity profiles $I(\nu)$

The splittings discovered by Mollow[9], i.e. X in the diagram below, are not very big. In fact, for the case of resonance, the greatest value of X, plotted there for $\Omega = 5K$ is at $(\nu - \omega_0)/K = 5$, i.e. in our notation, $X \sim 5$ for $A = 5$.

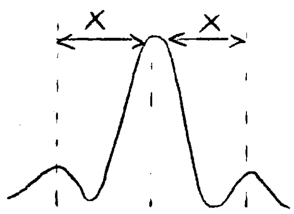


Fig 7.D. 1

Even if $\gamma \sim 0.01$ Å then $X \sim 0.05$ Å for $\Omega = 5K$. If $\Omega >> K$, i.e. in our notation, a large, then, e.g. if $\Omega = 1{,}000$ K then $X \sim 1{,}000\ \gamma$ and for $\gamma \sim 0.01$ Å, $X = 10$ Å. So for such intense fields X would not be such as to make Γ_{31} and Γ_{21} very different from each other.

N.B. $\Gamma_{21} \propto \omega_2^3$

and $\Gamma_{31} \propto \omega^3$

thus, if level 3 is split, then

$$\Gamma_{31} \propto (\omega \pm X)^3$$

but $$\frac{\Gamma_{31}}{\Gamma_{21}} = \frac{(\omega \pm X)^3}{\omega_2^3} \sim 1$$ even for very intense fields e.g. for sodium d_1 and d_2 lines

$$\frac{(\omega \pm X)^3}{\omega_2^3} = \frac{(5896 \pm 10)^3}{5890} = \frac{5886}{5890} \doteq .9979$$

$$\text{or } \frac{5906}{5890} \doteq 1.008$$

It is thus valid to assume $\Gamma_{31} \doteq \Gamma_{21} (= \Gamma)$ for a even larger than the largest value we have used, namely a = 100.

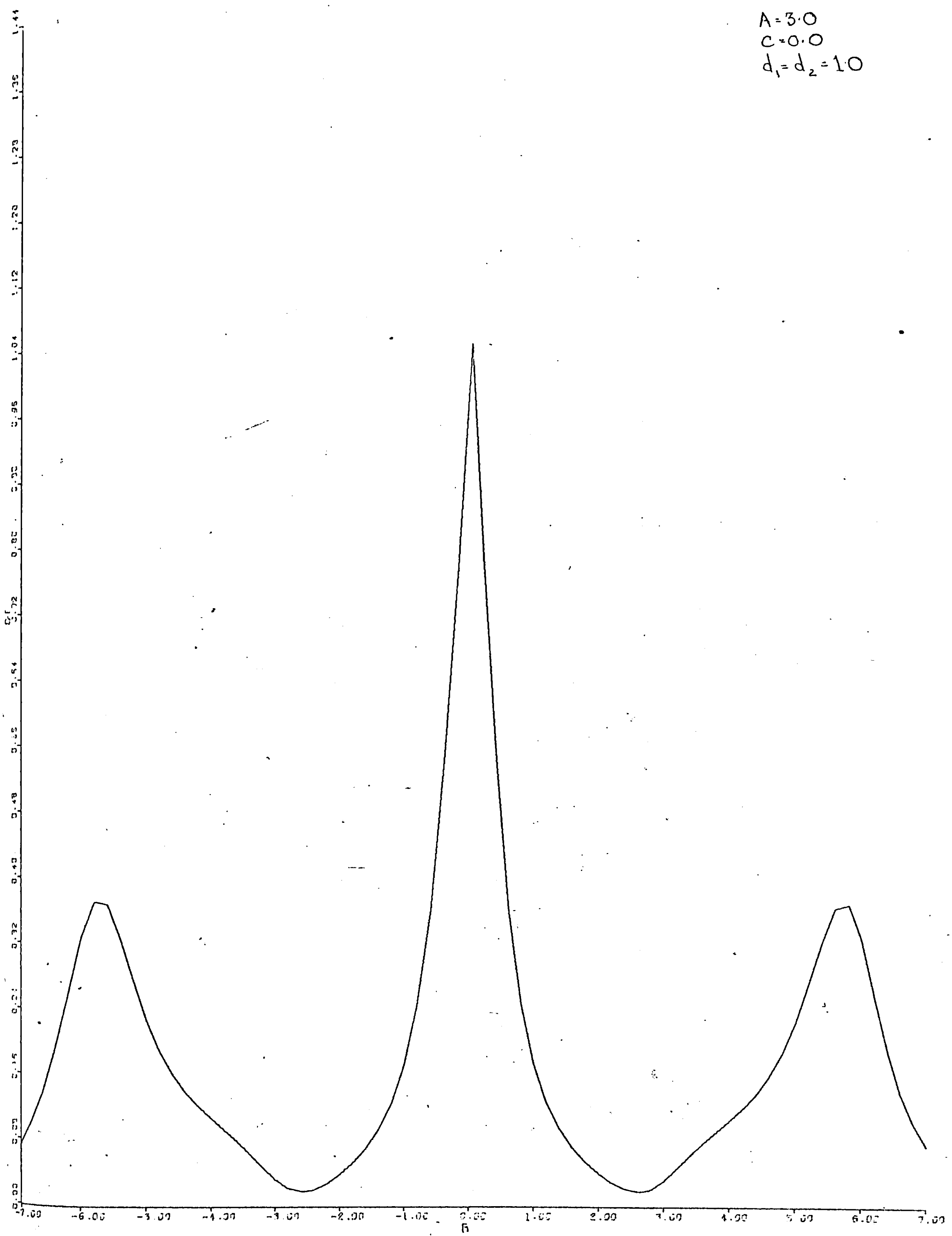
We shall now investigate whether the approximation is true when γ_3/γ = C is large, since then it would seem that ω_2 and ω are not so close in value, although in fact it is not ω_s which is large but ω/γ. When C = 100, ω_s = 100 γ, then, depending on the size of γ, ω^3 will either remain $\sim \omega_2^3$ or no longer be of this order. In general γ would be quite small enough to render ω_s a not very sizeable quantity so that ω^3/ω_2^3 would still be $\sim$ 1, even for C = 100. E.g. if γ = 0.01 Å and C = 100 $\omega_s = 100 \times 10^{-8} = 10^{-6}$. Thus we see that considering C large does not in fact mean a large absolute separation of levels 2 and 3 but rather a large separation compared with the overlapping case, when C 1, i.e. $\omega_s \sim \gamma$. When ω_s = 10 γ or > 10 γ i.e. C = 10 or >10 then levels 2 and 3 are in not overlapping in the normal sense of the word but from the spectral profiles it can be seen that level 2 still has an important effect.

Our assumption $\Gamma_{31} \doteq \Gamma_{21} (= \Gamma)$ is thus valid for large values of a and c. It follows that it would be valid for a small since then X is small, and for c small since then $\omega \doteq \omega_2$.

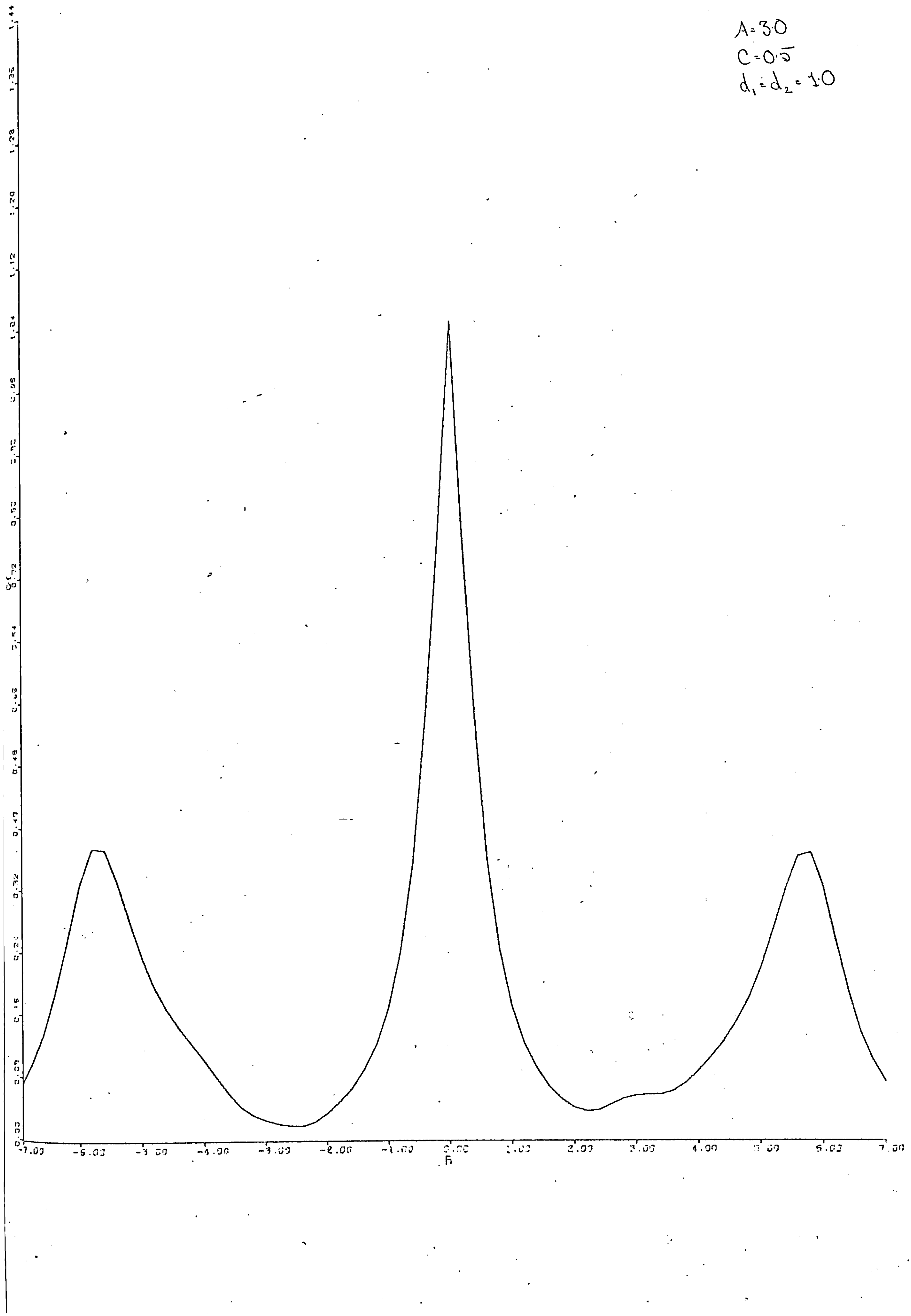
When c = 0, levels 2 and 3 are completely degenerate, i.e. they may have different magnetic quantum numbers, ℓ, but the same principal quantum numbers, n, and the same energy. The field then couples levels 2 and 1 as well, as in Apanasevich's paper[34] But since in this case levels 2 and 3 are indistinguishable, introducing an extra dipole moment into the calculations would be meaningless and in fact our equations

(7.A.43)-(51) are still valid for $\theta = 0$ although of course $\rho_{23} = \rho_{32} = 0$ and $\rho_{22} = 0$ so that we need only equations for $\dot{\rho}_{11}^{(5)}$, $\dot{\rho}_{33}^{(5)}$, $\dot{\rho}_{13}^{(5)}$ and $\dot{\rho}_{31}^{(5)}$, as in the 2-level atom.

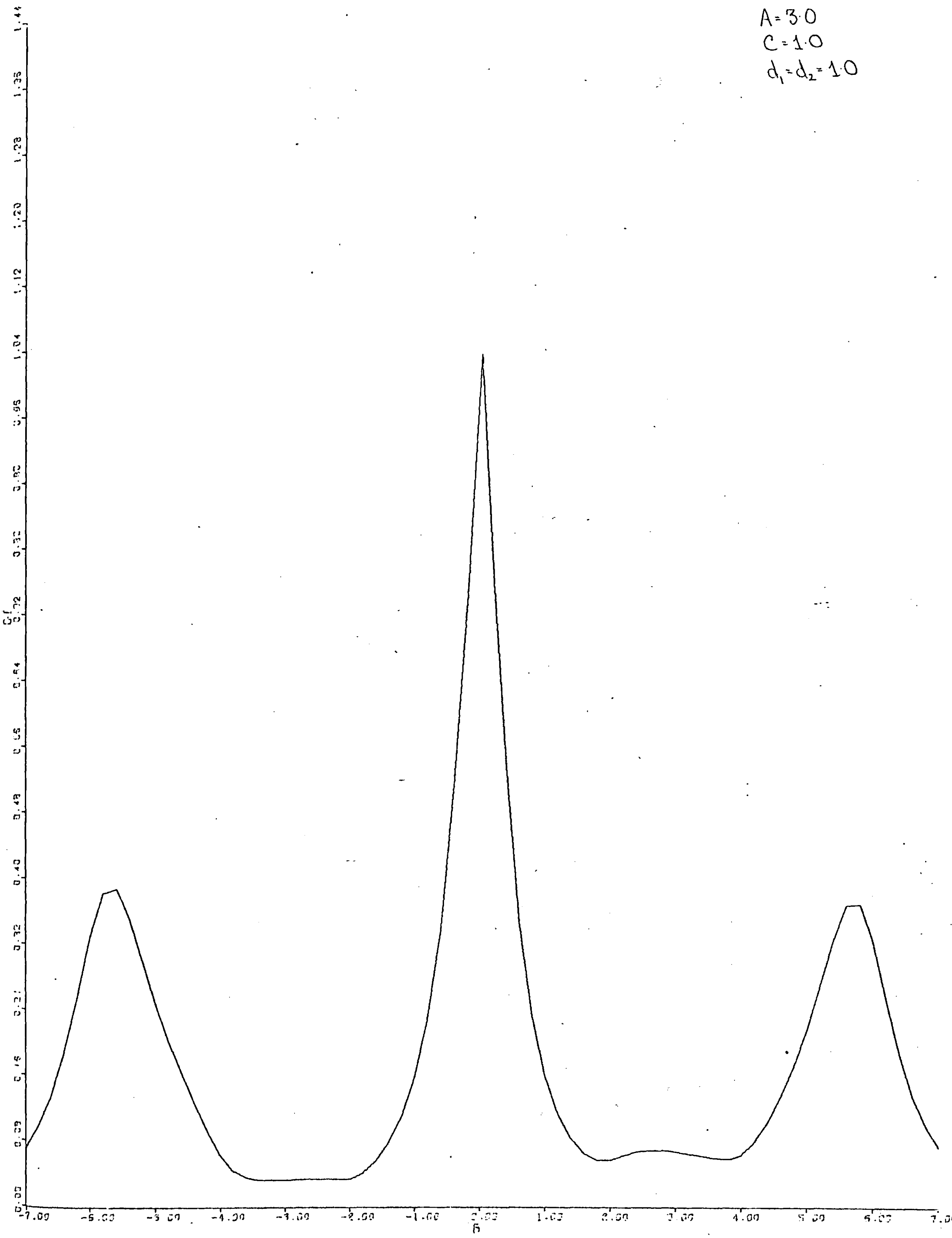
E. Graphs of spectral profiles for $d_1 = d_2 = 1$, $\Lambda = 3$ (i.e. field of moderate strength) and various values of c (i.e. graphs of $\widetilde{I}(b)$ versus b, or GI versus B)



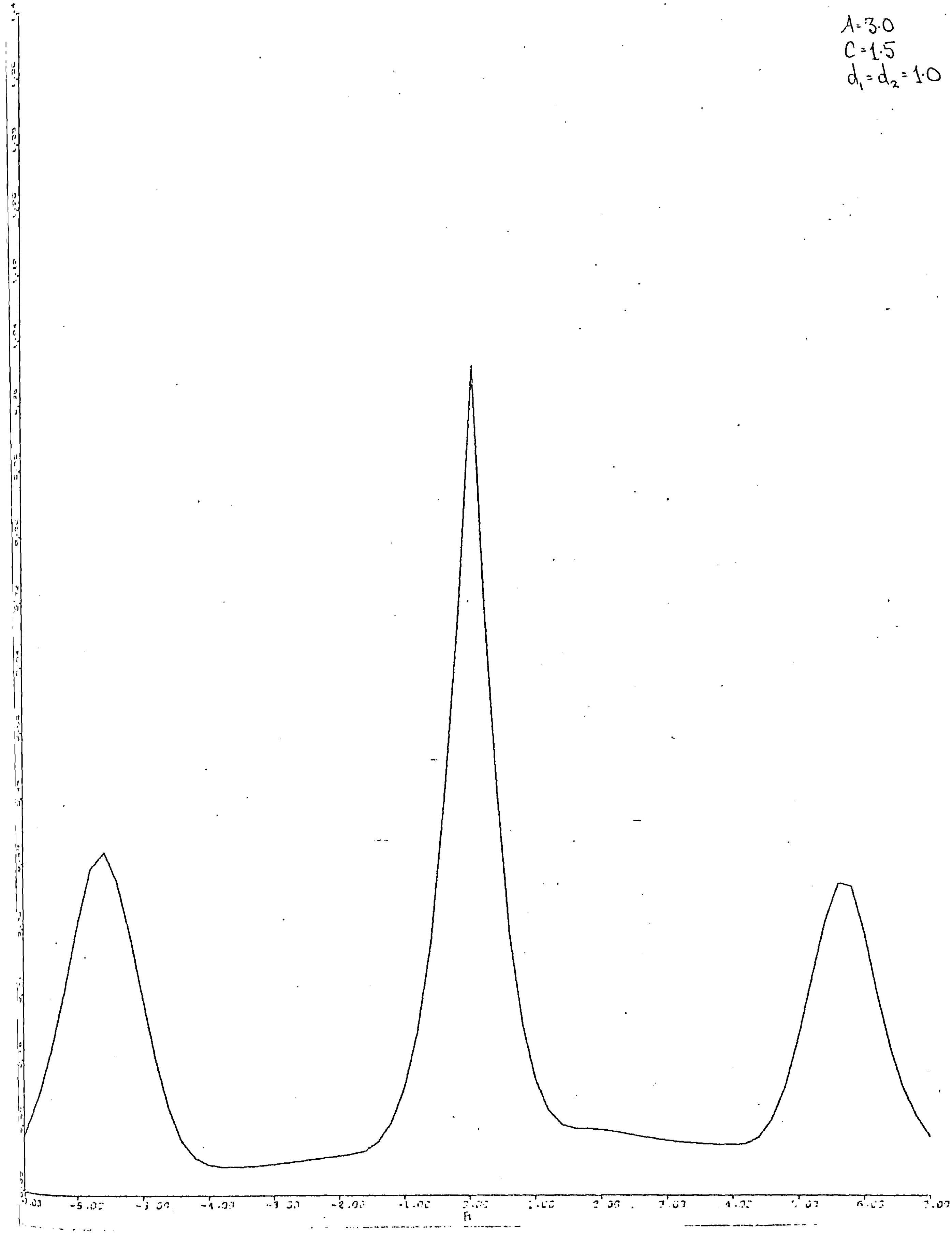
A = 3·0
C = 0·0
$d_1 = d_2 = 1·0$
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
0.00
1.00
2.00
3.00
4.00
5.00
6.00
7.00
B

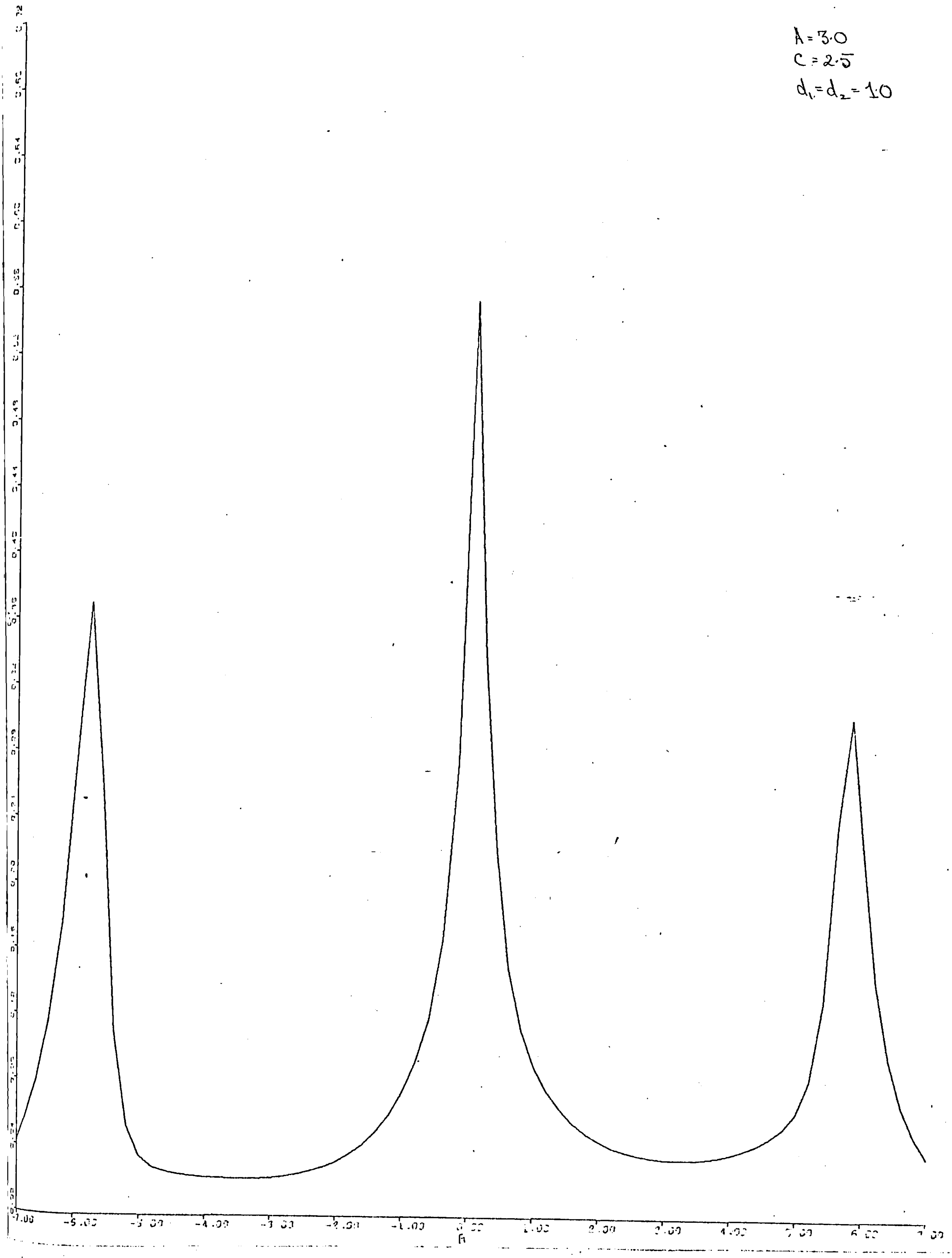


A=3·0
C=0·5
$d_1 = d_2 = 1·0$
G1
h
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
0.00
1.00
2.00
3.00
4.00
5.00
6.00
7.00

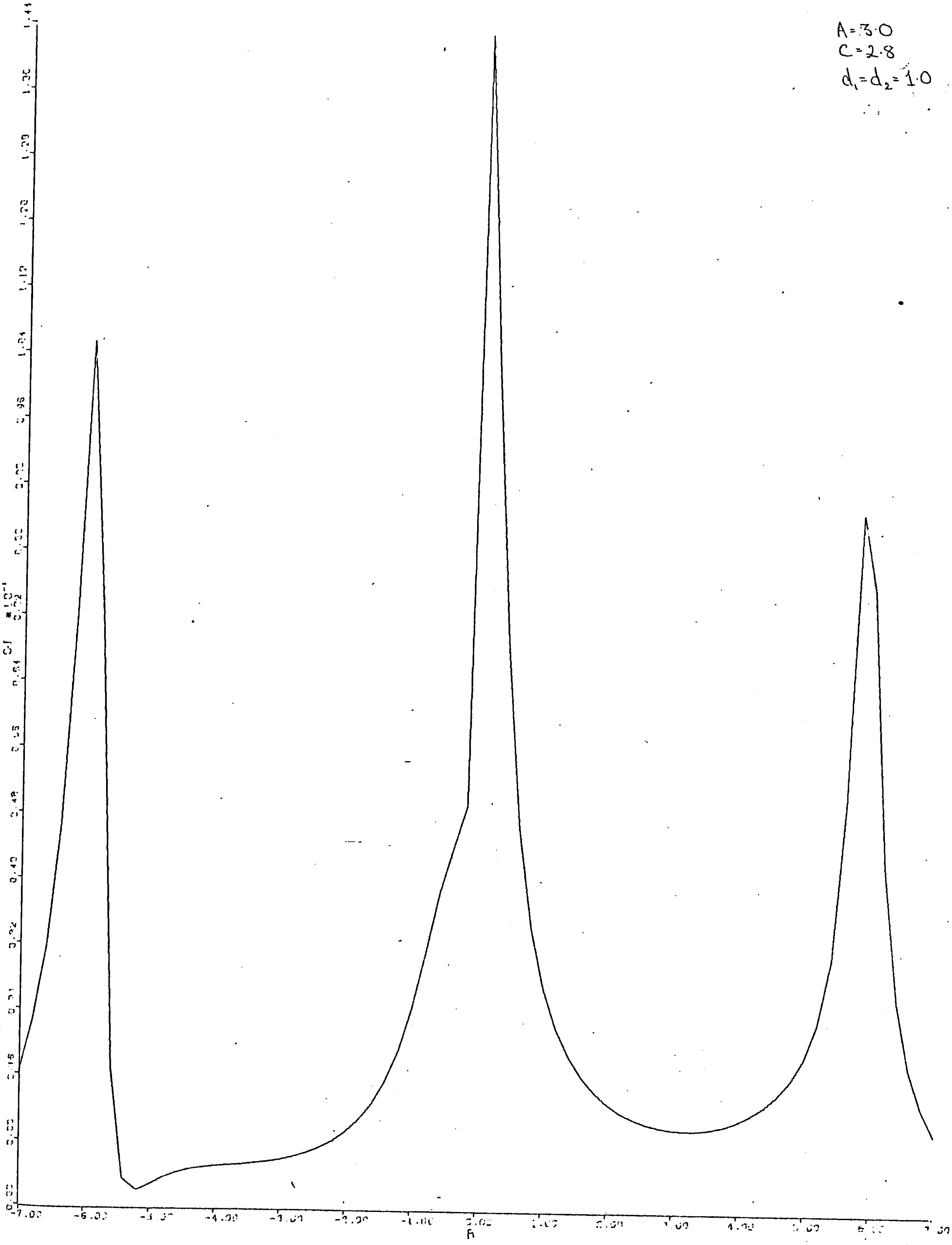


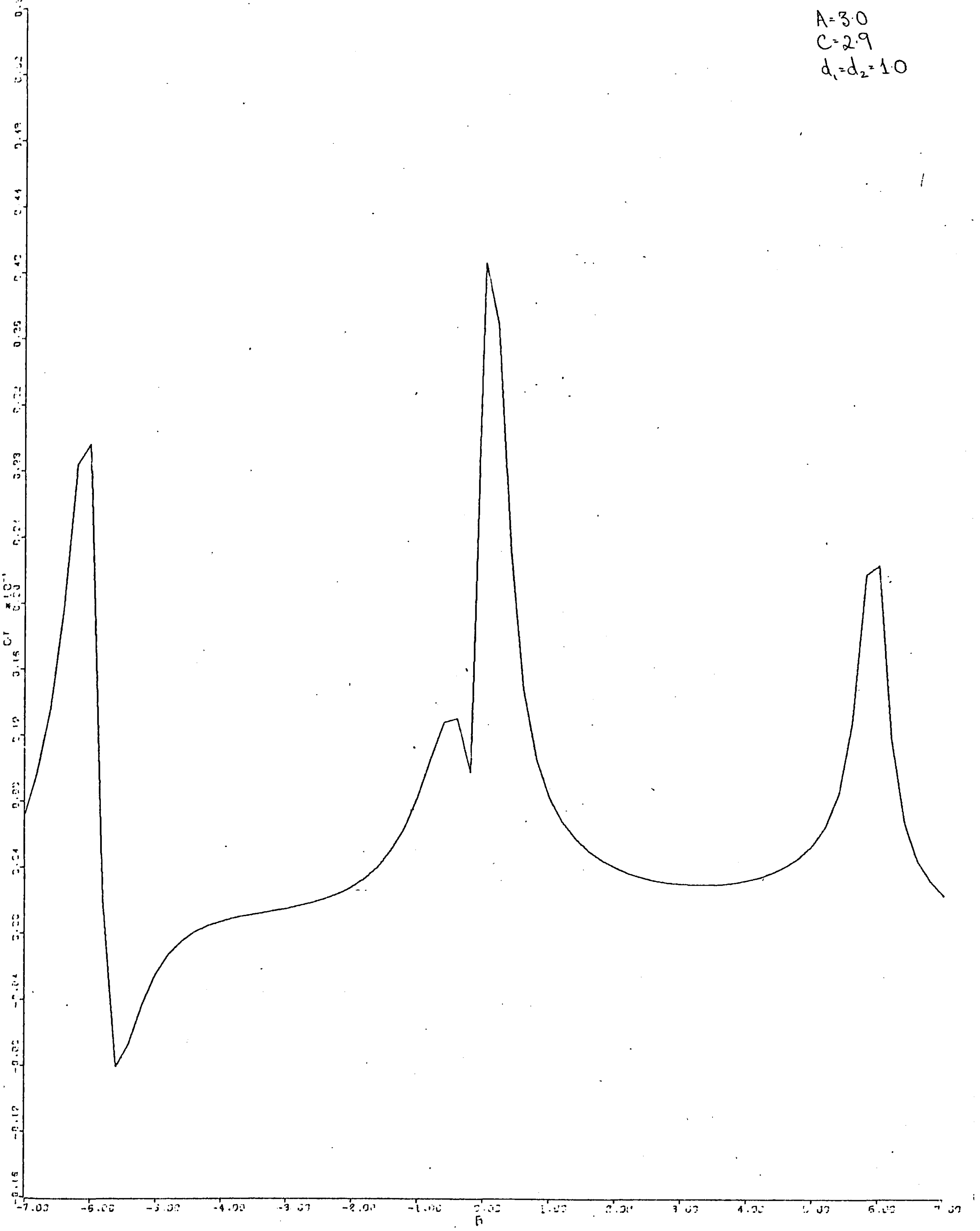
A = 3·0
C = 1·0
$d_1 = d_2 = 1·0$

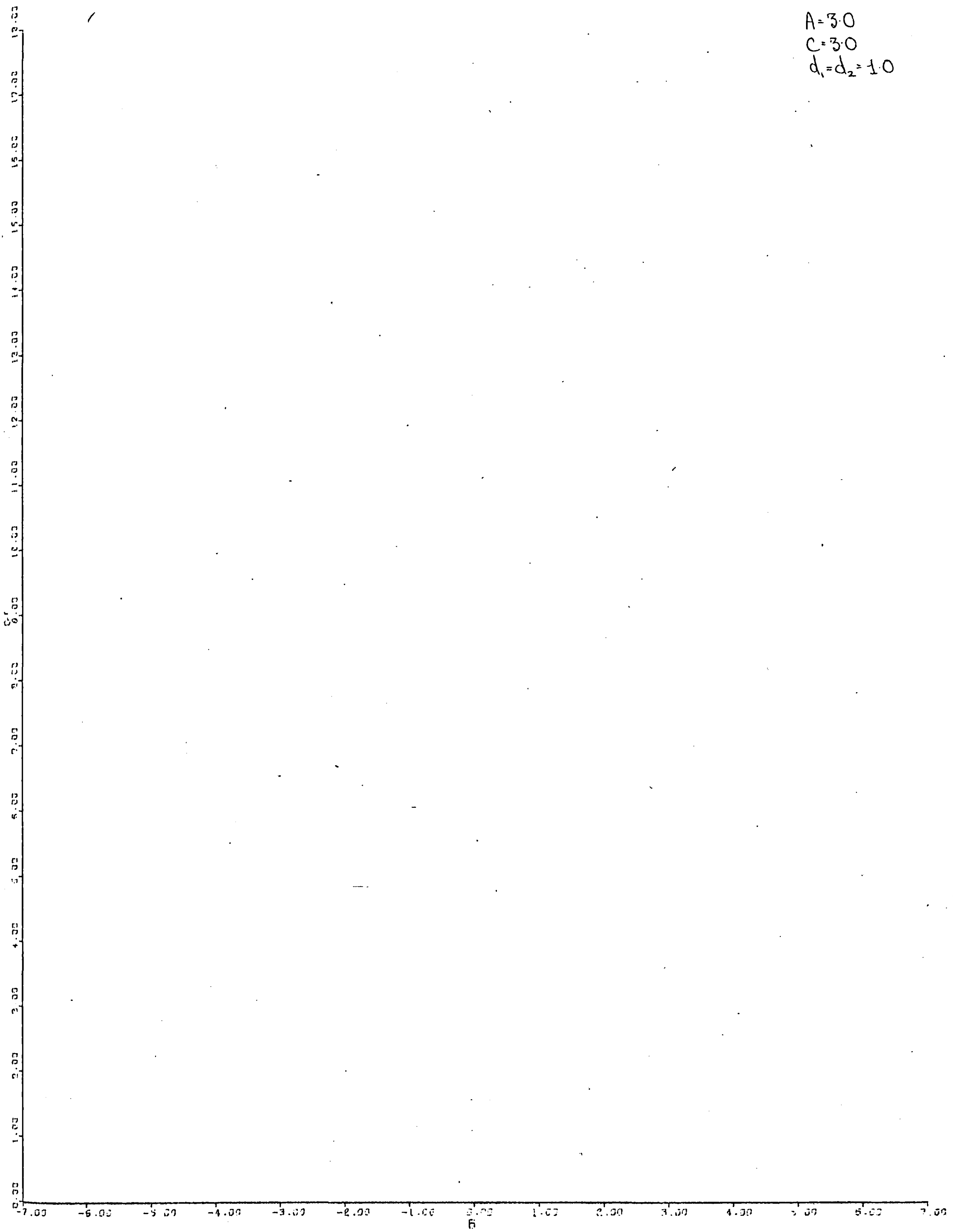

A=3·0
C=1·5
$d_1 = d_2 = 1·0$
-5.00
-3.00
-4.00
-2.00
-1.00
1.00
2.00
4.00
6.00
7.00

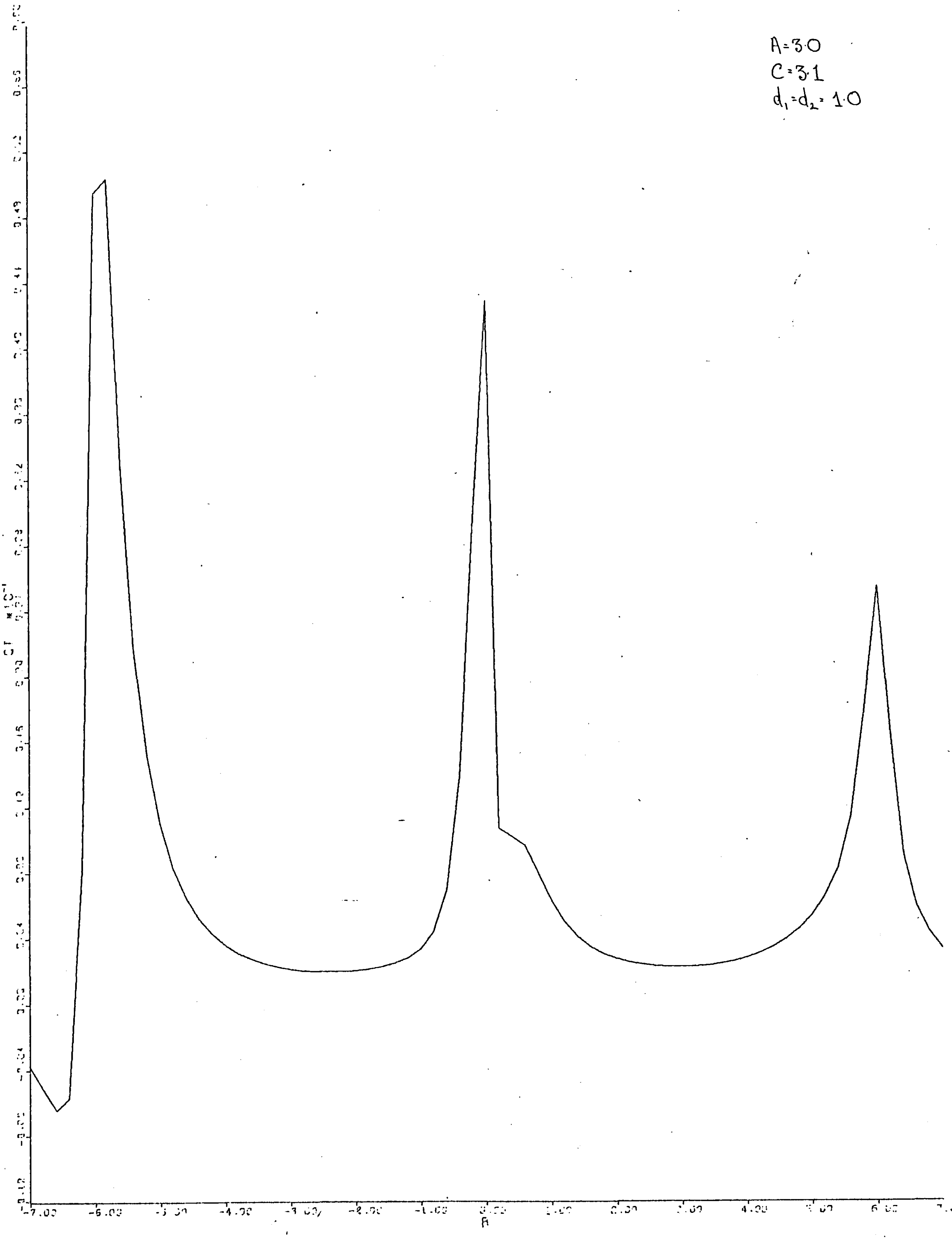


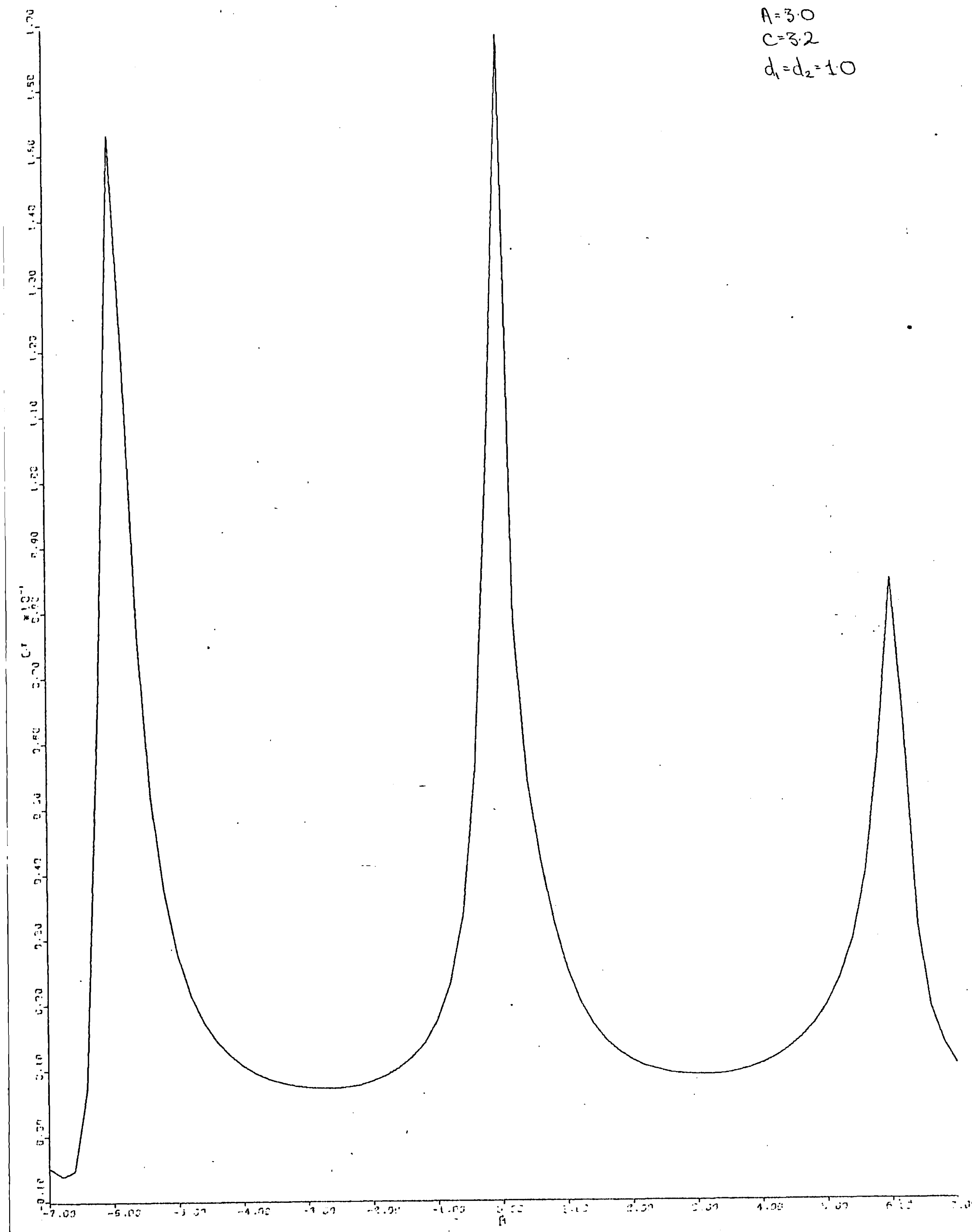
A = 3.0
C = 2.5
$d_1 = d_2 = 1.0$

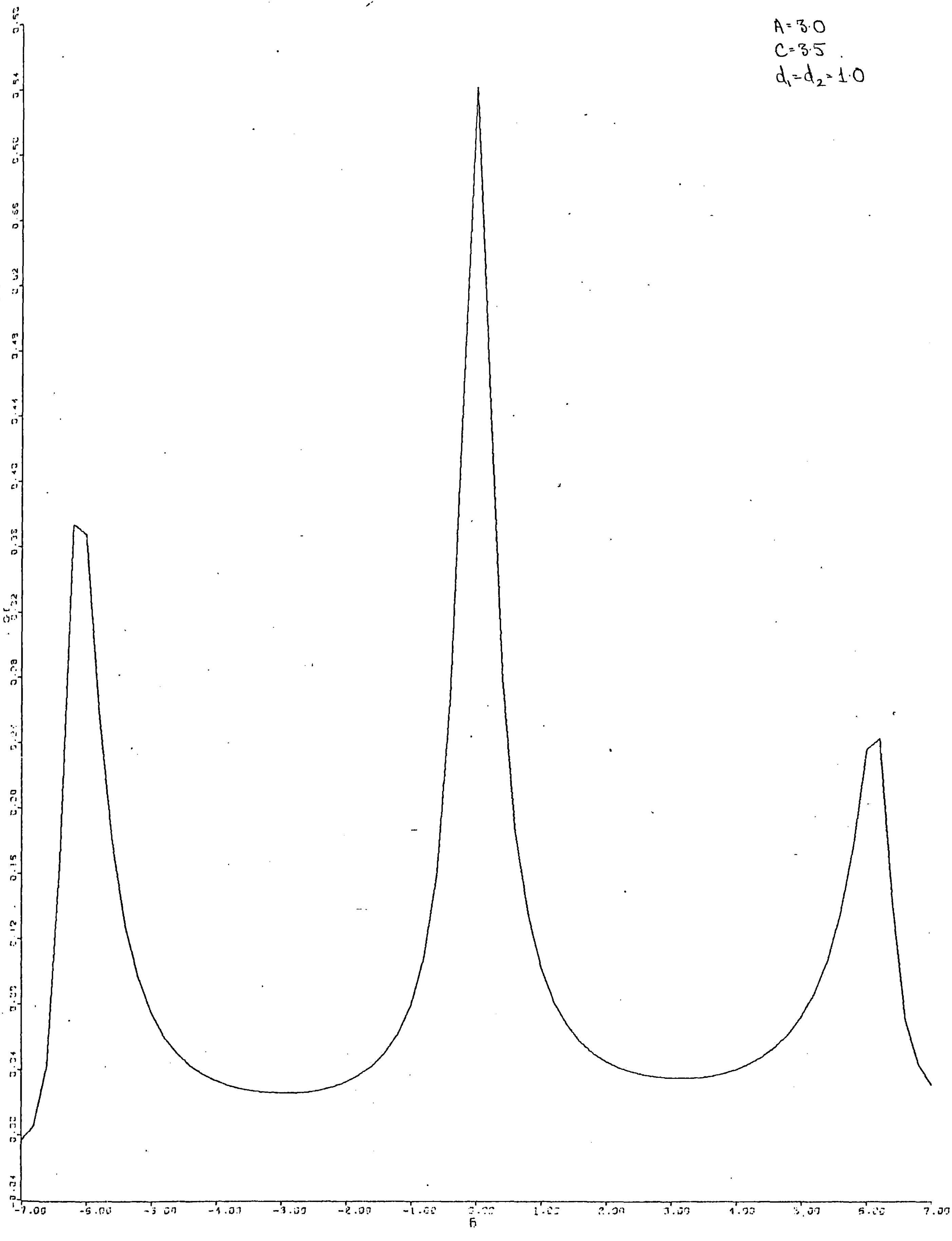

A=3.0
C=2.8
$d_1=d_2=1.0$
Cf $*10^{-1}$
h

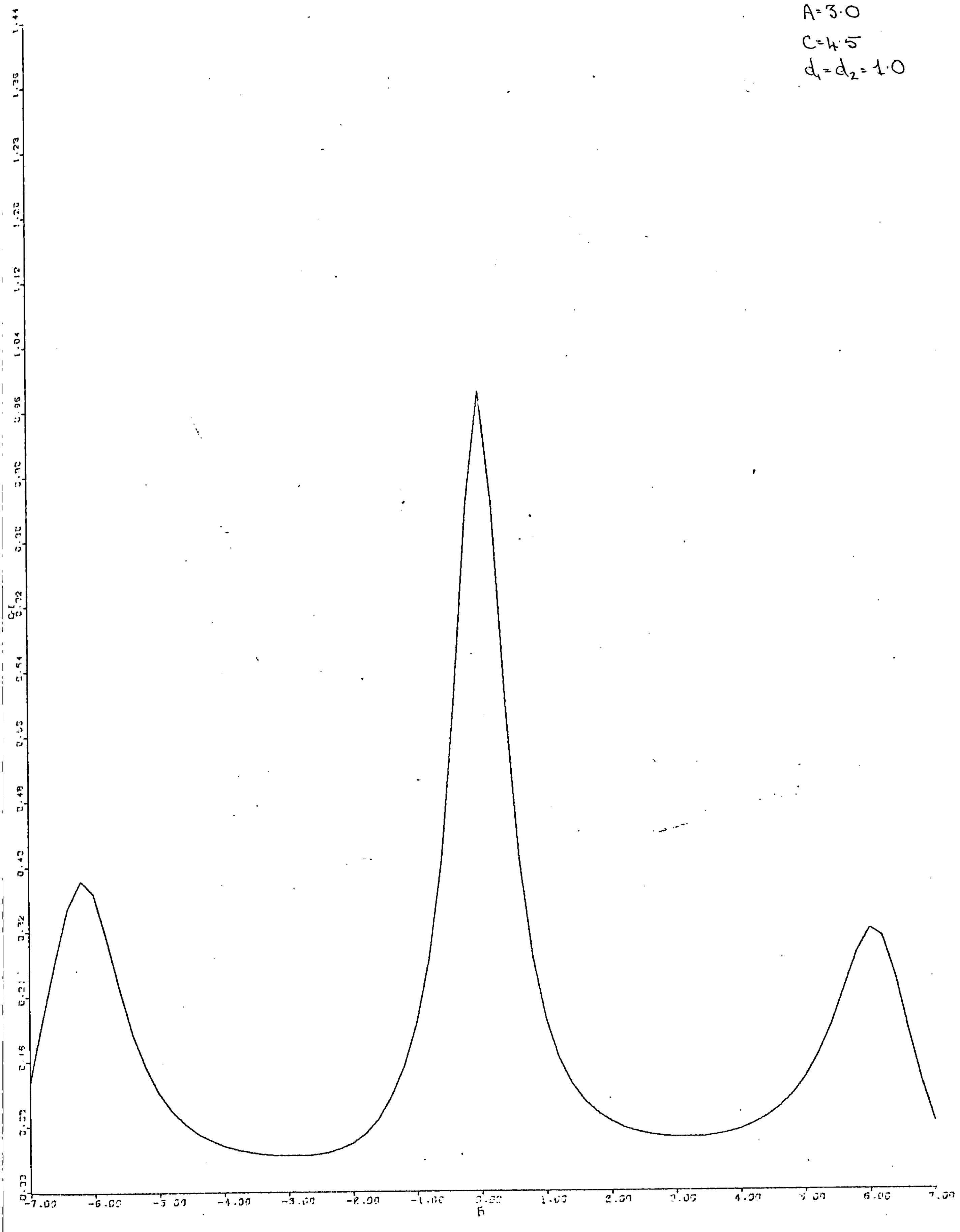

A=3·0
C=2·9
d_1=d_2=1·0
-7.00
-6.00
-4.00
-2.00
-1.00
1.00
4.00
6.00

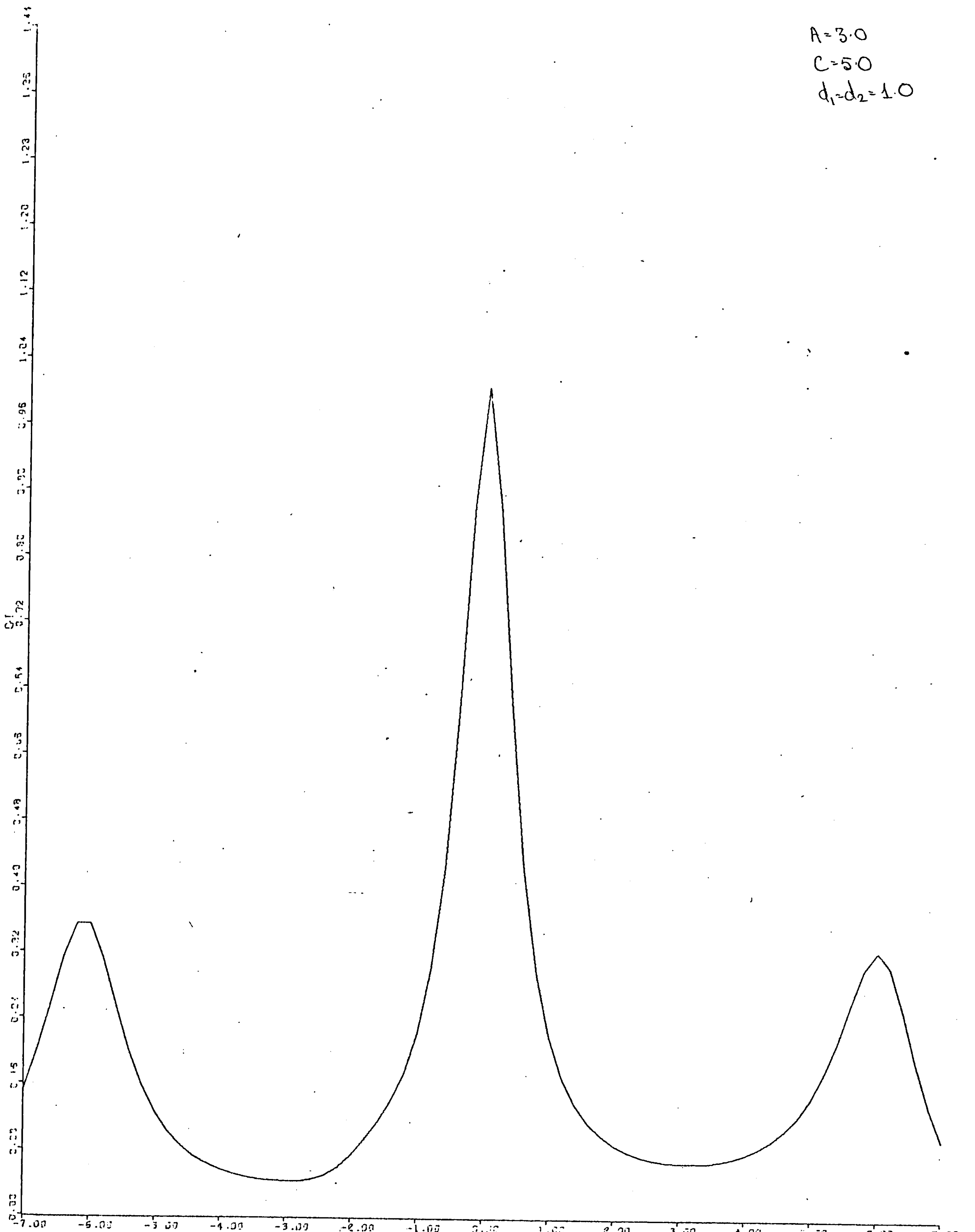

A=3.0
C=3.0
$d_1=d_2=1.0$
-7.00 -6.00 -5.00 -4.00 -3.00 -2.00 -1.00 0.00 1.00 2.00 3.00 4.00 5.00 6.00 7.00
B
0.00 1.00 2.00 3.00 4.00 5.00 6.00 7.00 8.00 9.00 10.00 11.00 12.00 13.00 14.00 15.00 16.00 17.00 18.00


A=3.0
C=3.1
d_1=d_2= 1.0
-7.00
-6.00
-4.00
-1.00
4.00
A

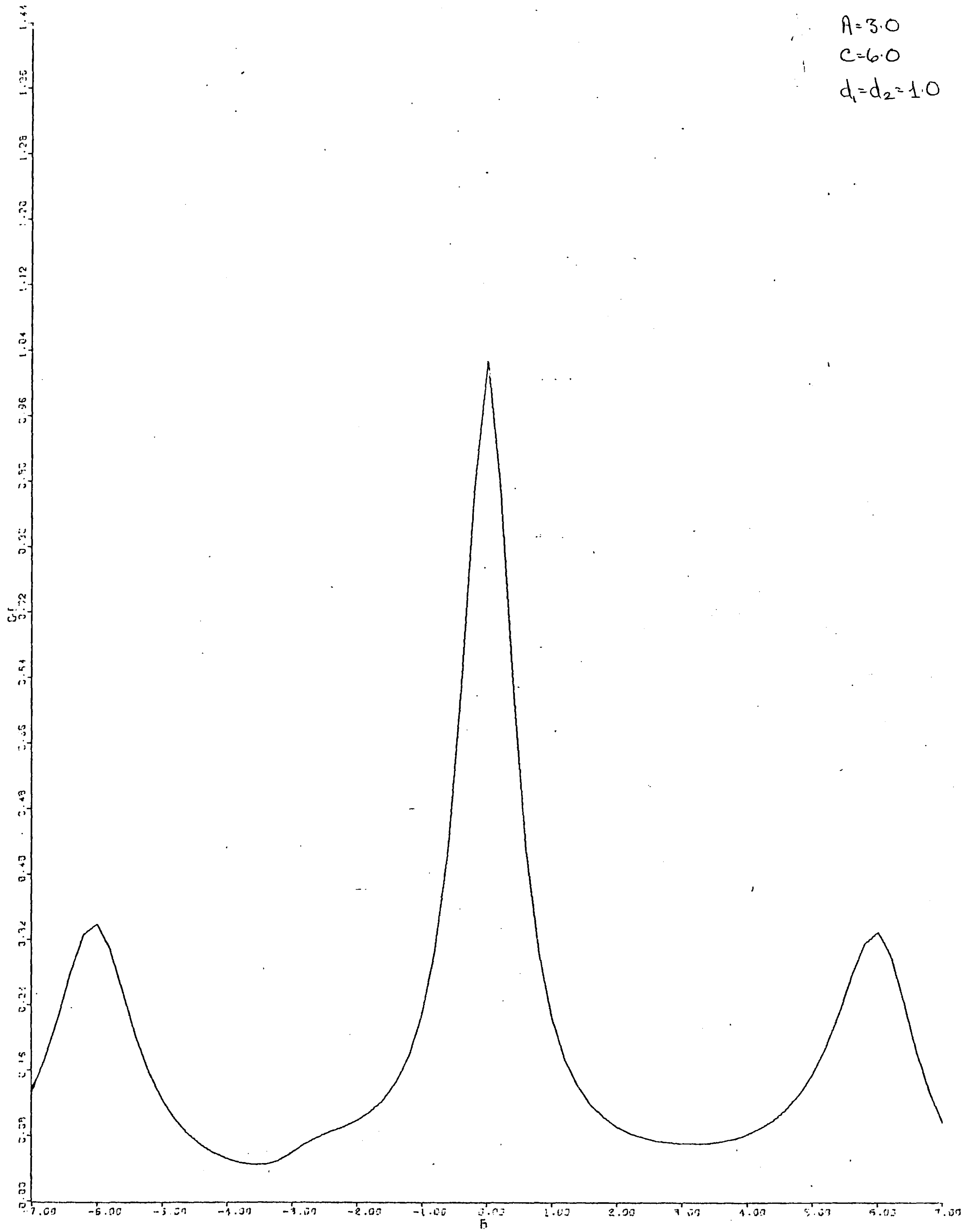

A=3·0
C=3·2
$d_1=d_2=1{\cdot}0$


A=3·0
C=3·5
d1=d2=1·0
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
0.00
1.00
2.00
3.00
4.00
5.00
6.00
7.00
б

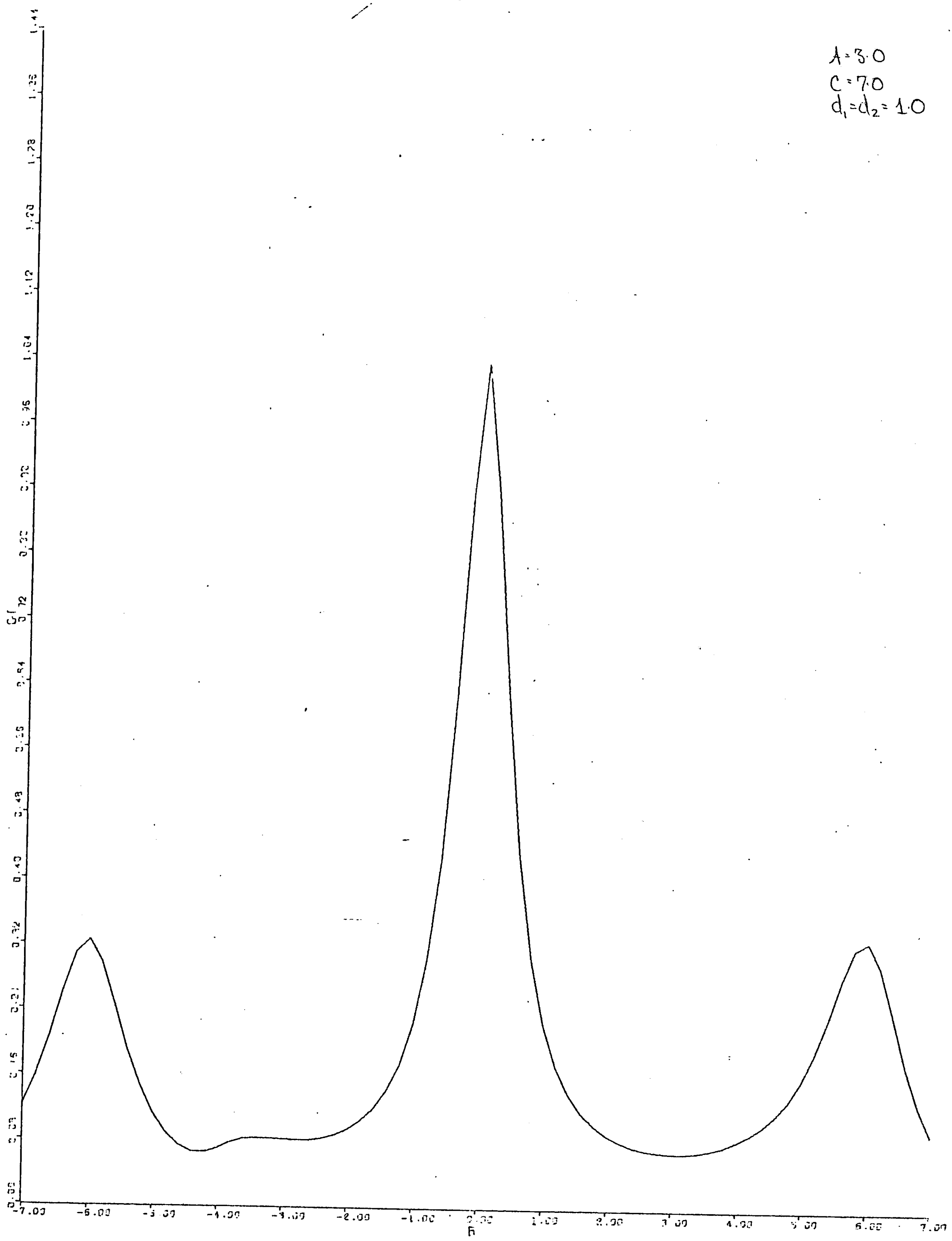

A=3·0
C=4·5
$d_1=d_2=1·0$
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
0.00
1.00
2.00
3.00
4.00
5.00
6.00
7.00
b

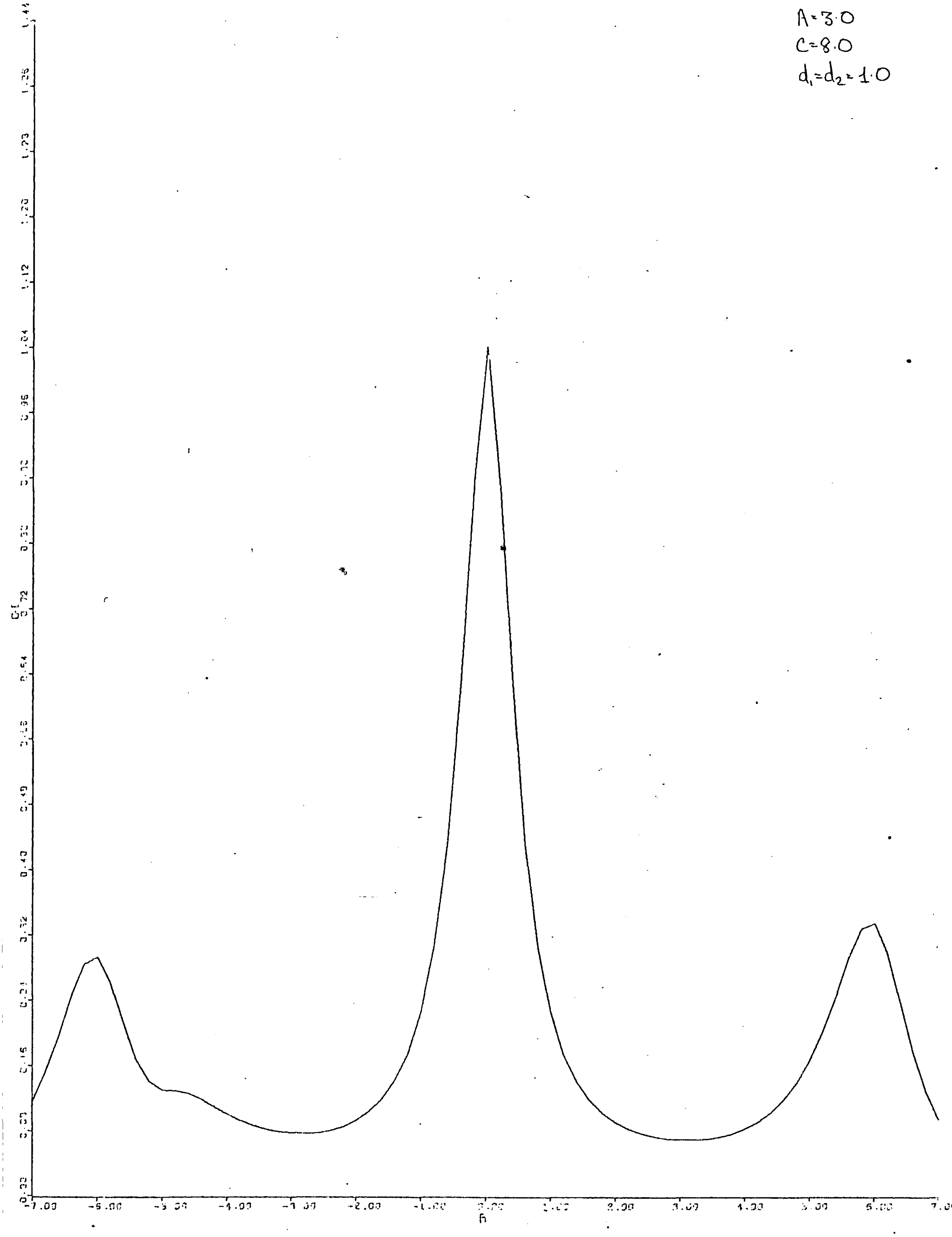


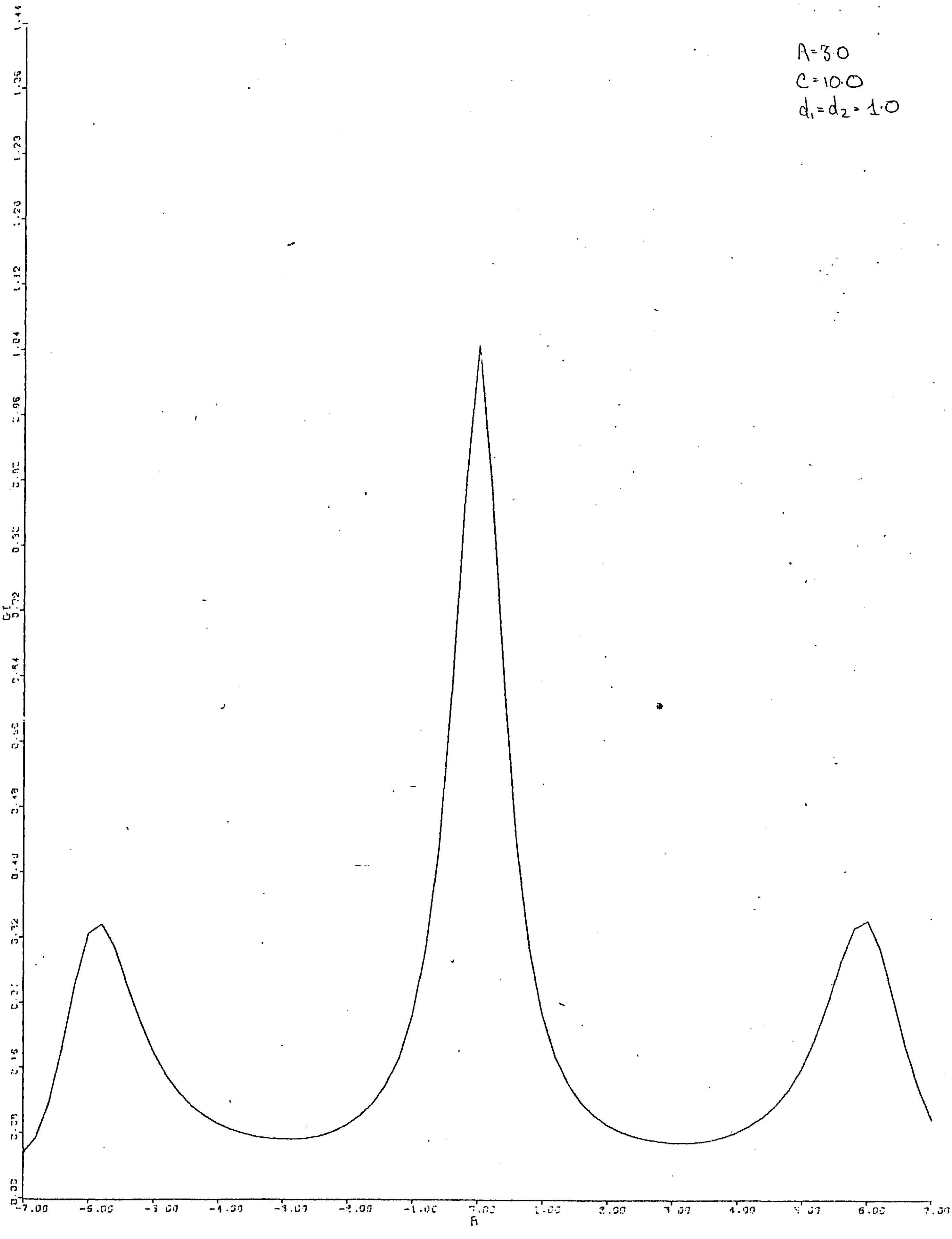
A=3.0
C=5.0
$d_1=d_2=1.0$
CI
A
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
1.00
2.00
3.00
4.00
7.00
0.00
0.32
0.40
0.48
0.72
0.80
0.96
1.04
1.12
1.20
1.23
1.41

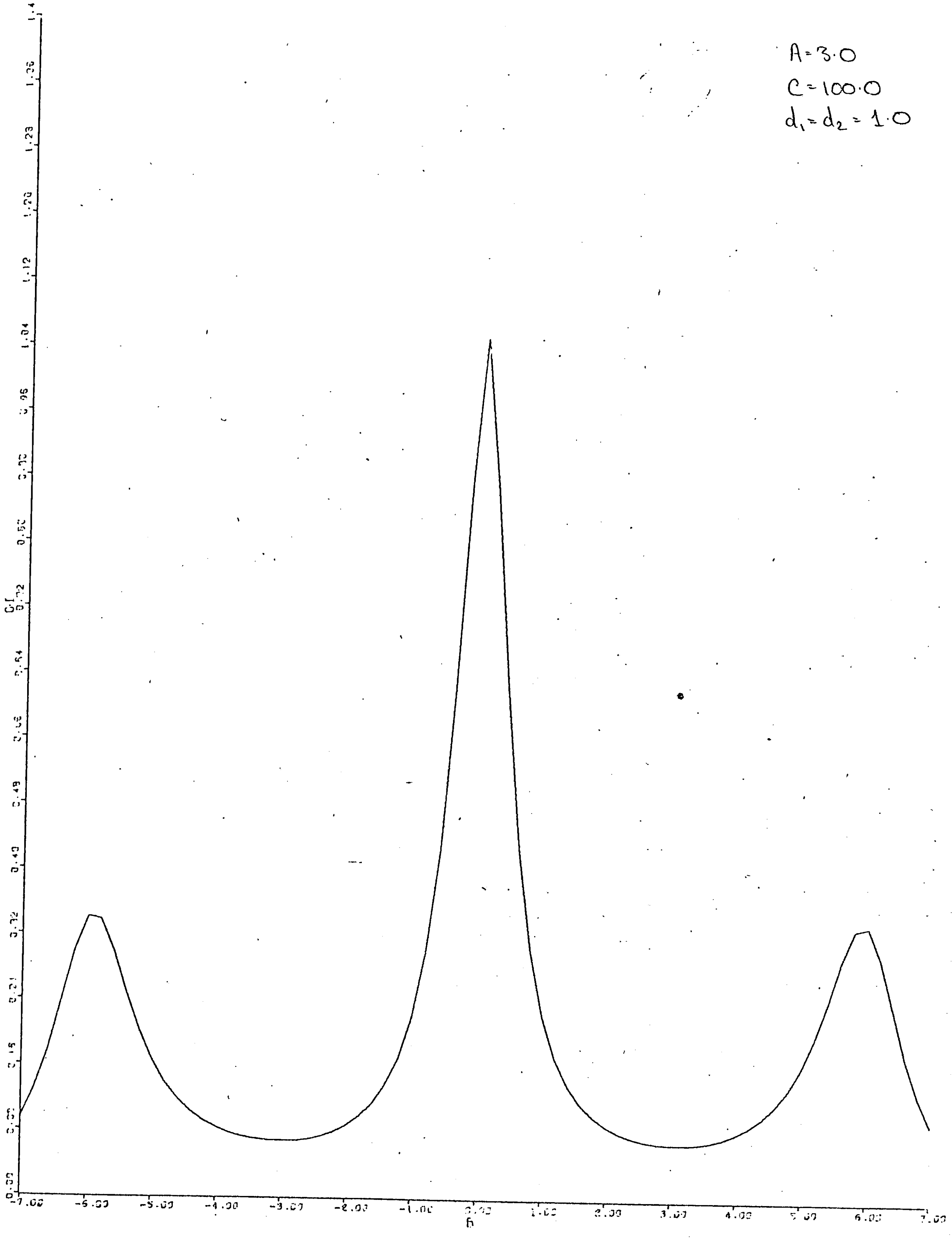


A=3.0
C=6.0
d1=d2=1.0


A = 3.0
C = 7.0
$d_1 = d_2 = 1.0$
Gf
R


A=3.0
C=8.0
d1=d2=1.0
h


A=3.0
C=10.0
$d_1=d_2=1.0$
h


A = 3.0
C = 100.0
$d_1 = d_2 = 1.0$
G.I.
h
-7.00
-6.00
-5.00
-4.00
-3.00
-2.00
-1.00
0.00
1.00
2.00
3.00
4.00
5.00
6.00
7.00

CHAPTER VIII

DISCUSSION

8.1 Criterion for assuming no cooperative effects occur

In all our calculations we have considered single atoms, so that we have implicitly assumed that the effects produced in such isolated atoms typify these effects produced on all the atoms of the ensemble as a whole. Our calculations are thus of particular relevance to atomic vapours (cf. Chapters V and VI on potassium vapour and hydrogen gas) where cooperative effects are unimportant since the coupling between atoms is negligible and so also is effective potential $V_{AA}(r)$ between the atoms.

According to Landau and Lifshitz (ref. 99, p. 387) scattering will take place independently at each molecule (or atom) provided that we consider the limiting case of a rarified gas where the mean free path of the molecules (or atoms), $\ell >> \lambda$, the wavelength of the light radiation. This is the restriction used in ref.1, namely that the atom is restricted to a region small in comparison to the average wavelength $2\pi c/\epsilon$. In this case Landau & Lifshitz point out that we are justified in discussing the scattering microscopically, using quantum mechanics.

In ref. 2, Part II, the case where a pair of atoms A_1 and A_2 is separated by a distance $r_{12} \sim \lambda$ is considered. The atoms then exhibit superradiant behaviour. When both the atoms are initially inverted Lehmberg finds that the intensity pattern $R(\theta, t)$ develops lobes in different directions at different times, so that the spatial distribution of the photons at time $t \rightarrow \infty$ is the same as in the independent-atom case.

The fact that we have ignored the line widths due to collisions and so considered only situations where the widths of the levels is set primarily by the interaction with the radiation field means that our calculations are, for this reason also, suited to a gas in a good vacuum, particularly for the upper optical transition (see ref. 75).

S.2 Dipole approximation

Throughout our calculations we consider only dipole transitions. This is obvious from our choice of the interaction Hamiltonian V_{SR}, where $V_{SR} = -\underline{E}(\underline{r},t).\underline{p}$ classically, or $V_{SR} = -\underline{E}(\underline{r},t).\underline{p}(A + A^{+})$ quantum mechanically (see eq. (1.B.96)) instead of the multipole expansion for a classical field given in Fiutak's paper (ref. 100 eq. (38)). Fiutak shows that V_{SR} can be written as the dipole interaction

$$V'_{SR} = -\underline{p}.\underline{E}(\underline{r},t)$$

[N.B. $V_{AL} = -\underline{E}(\underline{r},t).\underline{p}$ (S.1)

according to eq. (1.B.96), when the atomic operators are not considered, and this is equivalent to, but not identical with,

$$V_{AR} = V_{AR}^{(1)} + V_{AR}^{(2)} = -e/m\, \underline{A}.\underline{p} + e^2/2m\, \underline{A}^2 \tag{S.2}$$

(see equations (1.B.79) and (1.B.80)) as shown by Fiutak [100] since, if the original Hamiltonian is

$$H = 1/2m\, \underline{p}^2 + V(\underline{r}) - e/m\, \underline{p}.\underline{A}(\underline{r},t) + e^2/2m\, \underline{A}(\underline{r},t)^2 \tag{S.3}$$

then the corresponding Lagrangian is

$$L = 1/2\, m\dot{\underline{r}}^2 - V(\underline{r}) + e\dot{\underline{r}}.\underline{A}(\underline{r},t) \tag{S.4}$$

Since adding a total time derivative will not affect the equations of motion of the system, the system is equally well described by

$$L' = 1/2\, m\dot{\underline{r}}^2 - V(\underline{r}) - e\dot{\underline{r}}.\frac{d\underline{A}(\underline{r},t)}{dt} \tag{S.5}$$

which is equivalent, but not identical with equation (S.4).

If we construct the Hamiltonian belonging to Lagrangian (S.5) we obtain

$$H' = 1/2m\, \underline{p}^2 + V(\underline{r}) + e\dot{\underline{r}}.\frac{d\underline{A}(\underline{r},t)}{dt}$$

which is therefore equivalent to the original Hamiltonian (S.3), since

$$e\dot{\underline{r}} = \underline{p}$$

and $\frac{dA(r,t)}{dt} = -E(r,t)$

$$\therefore H' = \tfrac{1}{2m} p^2 + V(r) - p \cdot E(r,t)$$

According to ref. 65, even though the magnitude of the field is not required to be small, the quadratic term $A^2(t)$ (which would otherwise appear) may be dropped in the dipole approximation.]

$$V'_{SR} = - p \cdot E(t) \tag{8.6}$$

when the wavelengths of the e.m. waves, $\lambda >>$ the dimensions of the system. Then $A(r,t)$ can be replaced by $A(t)$ evaluated e.g. at the nucleus (see also ref. 65, eq. (1)) and $\frac{dA(r,t)}{dt} = \frac{\partial(A(t))}{\partial t} = -E(t)$ where $E(t)$ is the external field at the nucleus.

For a system interacting with a classical field this form of the interaction Hamiltonian (eq. (8.6)) is equivalent to t he original form, viz.

$$V_{SR} = -e/m\, p \cdot A(r,t) + e^2/2m\, A(r,t)^2$$

where the notation of equations (1.B.79) and (1.B.80) is used, if the total Hamiltonian, $H = H_a + V_{SR}$ is given by equation (1.B.77).

When higher-order multipole interactions are considered Fiutak shows that the total Hamiltonian for systems interacting with an external e.m. field, which is classical and time-dependent, can be written, as in eq. (38), as

$$H' = \mathcal{H}(\Pi'_s, r_s) + e\,\phi(R,t) - \sum_{s=1}^{n} e_s \sum_{k=0}^{\infty} \frac{1}{(k+1)!} (\xi_s \cdot \nabla)^k E(R,t)\,\xi_s \tag{8.7a}$$

where the momenta Π_s are

$$\left.\begin{aligned} \Pi_s = {} & p_s + e_s \sum_{k=0}^{\infty} \frac{(k+1)}{(k+2)!} (\xi \cdot \nabla)^k \xi_s \times B(R,t) - a_s e\, A(R,t) \\ & + a_s \sum_{r=1}^{n} e_r \sum_{k=0}^{\infty} \frac{1}{(k+2)!} (\xi_r \wedge \nabla)^k \xi_r \times B(R,t) \end{aligned}\right\} \tag{8.7b}$$

$r_s = r_1$ ...... $, r_n$ are the positions of the n charged particles or systems,

$\mathcal{H}(\Pi_s, r_s)$ is the Hamiltonian of the ensemble in the absence of interaction with the external field

$\underline{R}$ is the centre of the multipole expansion

ξ_s are coordinates relative to this centre

$$\underline{R} = \sum_{s}^{n} q_s x_s \qquad \xi = x - R \tag{8.8}$$

Π_s are kinetic momenta, which are expressed entirely in terms of the magnetic field strength.

$e = \sum_s e_s$ = total charge of the ensemble.

$\phi(\underline{R},t)$ is the scalar potential.

N.B. dashes indicate a canonical transformation from the original equation.

This equation for H shows that the interaction of the ensemble with the electric component of the field is independent of the dynamics of the systems. It has the familiar form of a sum of electric dipole, electric quadrupole, etc. interactions. In particular, in the long wavelength limit, specified earlier, and for vanishing total charge, $e = 0$, the interaction with the electric field is given by

$$-\underline{P}.\underline{E}(t)$$

where $\underline{P} = \sum_s p_s = \sum_s e_s x_s$ is the total electric dipole moment of the ensemble. The interaction with the magnetic component of the field, on the other hand, depends on the form of the system Hamiltonian $\mathcal{H}$. The non-relativistic Hamiltonian of an atom contains, in 1st approximation from (38) the familiar magnetic dipole interaction term. We ignore this also since it is only important for shell energies $\sim mc^2$.

Although he shows that for a classical field the multipole form of the Hamiltonian is exactly equivalent to the original Hamiltonian, of eq. (7) $H = \mathcal{H}(\Pi_s, x_s) + \sum_{s=1}^{n} e\phi(x_s, t)$ in the case of a quantised field he shows that the multipole form of the transformed Hamiltonian is equivalent to the original Hamiltonian only for 1st-order radiation processes.

Hence we see that if we consider the e.m. field classically our calculations, using the dipole approximation, are valid for the long wavelength limit when the sum of all the charges of all the individual systems is negligible. In considering the e.m. field quantum mechanically we are also limited to 1st-order radiation processes. Thus in assuming the form of $V_{<R}$ given in (1.B.96) and considering the e.m. field quantum mechanically we have 4 limitations:

1) λ of the e.m. waves $>>$ dimensions of the system, i.e. we ignore the dependence of $\underline{E}$ on posiion $\underline{r}$,
2) total charge of all the individual systems is negligible,
3) only 1st-order radiation processes can be considered, i.e. we cannot consider Compton effect and dispersion,
4) shell energies must not be $\sim mc^2$, i.e. we cannot consider heavy atoms.

N.B. In his quantum mechanical derivation Fiutak assumes that relativistic effects are negligible, i.e. characteristic frequencies of the system frequencies of relevant virtual photons. In this case momenta of the ensemble are replaced by transformed momenta in the multipole form (see $\underline{\Pi}'_{<}$) and the following term representing the interaction of the system with the electric component of the field, is added

$$e\phi(R,t) - \sum_s e_s \int_0^1 du\, \xi_s \cdot \underline{E}(\underline{R}+u\xi_s,t)$$

A discussion of quadrupole and magnetic dipole radiation is also given by Heitler[43] where he shows that even if an electric dipole is forbidden there may be electric quadrupole or magnetic dipole transitions even though the ratio of the intensity for such transitions to the intensity for allowed electric dipole transitions is $\sim (a/\lambda)^2$ and so is <u>unimportant</u> for emitted wavelengths <u>long</u> in comparison with the dimensions of the atom.

8.3 R.W.A.

We have used the R.W.A. (which assumes that frequencies, ω_ν, close to the atomic resonance frequency, ω_0, are the mominant ones) throughout, although we have shown in Appendix I, for the 2-level atom, that if it is not assumed, we obtain some extra high frequency terms, which are negligible, and, <u>anti-resonant frequency shift modifications</u> which we can neglect as they are not our main concern. In fact when the coupling becomes stronger, i.e. when the dipole moments are large, R.W.A. breaks down (cf. Louisell[35]) but it would anyway only give rise to a very small correction (see ref. 100). This is also pointed out in the papers we shall be quoting where several approximations are compared.

Various papers, e.g. refs. 101, 102, 103, discuss the importance of the R.W.A. in spontaneous emission theory. Knight and Allen show that the Wigner-Weisskopf theory of spontaneous exponential decay of a single excited atom contains an explicit form of the R.W.A. However, they say that R.W.A. should not be used to describe cooperative superradiant level shift and that care should be taken over the choice of the interaction Hamiltonian, V_{SR}. They point out that the R.W.A. has a significant effect, both on (a) the spontaneous emission from a single excited atom, and on (b) the cooperative spontaneous emission level of radiatively coupled atoms.

K. and A. say that use of R.W.A. to neglect overlapping diagrams (b) for the single 2-level atom is NOT a serious limitation, since there is NO anti-resonant vacuum contribution to the dominant 2nd-order diagram. On the other hand, R.W.A. must not be used in calculating the change in the radiative level shift owing to external fields since there is an antiresonant contribution to the 2nd-order perturbation term, which alters the frequency dependence of the energy shift, but we shall not be concerned with this. They say this alteration makes possible extensive cancellation of otherwise misleading terms although we did not find this (see Appendix I).

In the rest of K. and A.'s paper they show the effects of not using the R.W.A. on the simplest cooperative system of 2 atoms one being in the excited state, cooperation deriving from an exchange of a photon between the atoms. We ignore this possibility of exchange.

For superradiant systems, in the Dicke limit, large frequency shifts are expected and it is possible that they may alter the essential dynamics of cooperative emission. It has been suggested by Friedberg[104] that the strong dispersive effect of dipole interactions between closely spaced atoms might destroy cooperation responsible for superradiance. These dipole interactions seem to some extent to be equivalent to the frequency shifts. In LOW density systems, which we consider neither objection applies since there will be no dipole interaction between widely spaced atoms and retardation and anti-resonant terms should be included.

We consider atoms which are coupled ONLY by the radiation from their transitions and ignore all cooperative effects discussed by K. and A., including superradiance. The only point of relevance is then that anti-resonant terms enable extensive cancellation of otherwise misleading terms but in our case we haven't found this to be so and have thus neglected anti-resonant terms.

Agarwal [102] has also questioned the validity of the R.W.A. in quantum optics. He considers spontaneous emission from (i) N identical harmonic oscillators and (ii) N identical 2 level atoms and finds that when R.W.A. is not used the radiation rate depends on the initial dipole moment phase.

Agarwal is concerned with discovering whether the assumption that R.W.A. is a good approximation, provided the interaction between the radiation field and matter is weak. For case (i) if each oscillator at t = 0 was excited to a coherent state $|z_0\rangle$ having FINITE dipole moment then the radiation rate at t = 0 may be very different from that obtain in R.W.A. $I^r(0)$ depending on the phase φ of the initial coherent amplitude. But if the system is excited to a state with ZERO dipole moment then $I(0)$ becomes identical with $I^r(0)$. In the limit of weak coupling $N\gamma_0 << \omega_0$ for the system initially having a FINITE dipole moment. $I(t)$ is the same as $I^r(t)$ provided that rapidly oscillating terms are ignored. Anyway it is reasonable to ignore them since they cannot be observed by available photodetectors and so an averaging operation over a few optical periods is always performed. Agarwal expects that this result will also hold good for 2-level atoms emitting spontaneously as opposed to harmonic oscillators. On the other hand, the correlation function $\langle a_j^\dagger(t)a_k(0)\rangle$

oscillates at the optical frequency ω . If the system's initial state has ZERO dipole moment then for weak-coupling $I(t) = I'(t)$.

For case (ii) if the system is excited to a state with ZERO dipole moment $I(0) = I'(0)$. Whereas if the system's initial state has a FINITE dipole moment then $I(0)$ has a strong dependence on the dipole moment phase ϕ and we see that for $\theta = \pi/2$ the emission is not necessarily superradiant unlike the result for R.W.A. When we consider ONE 2-level atom only $I_1(t) = I_N(0)$ but the dipole moment differs from that obtained by making R.W.A.

We are not concerned with radiation rates but rather with spectral densities. The initial state is not known at all in Chapters V and VII and in the other chapters is not specified explicitly with regard to its dipole moment.

Finally we shall consider Walls and Cardiner's paper [103] in which he shows that the 3 approximations used in spontaneous emission theory are clearly related. They are

1) Wigner-Weisskopf (W-W)

2) R.W.A.

and 3) ladder approximation.

They consider a 2-level atom and use a Hamiltonian in the dipole approximation, like our eq. (B.8.21), which they say excludes the A^2 term, which makes a negligible contribution to a single photon emission process.

They consider the W-W approximation is two parts.

(i) W-WI probability amplitudes don't change significantly from their initial values for small values of t.

(ii) W-WII introducing irreversible behaviour by replacing the dependent summation in their eq. (6) by a complex constant. This is explained by von Foerster[43], who also shows it to be equivalent to the assumption of a Markoffian approximation. W-WII yields the familiar result of exponential decay which is well validated by experiment.

They show that W-WI is equivalent to the R.W.A. Knight and Allen [105] also point out that W-WI approximation and the ladder approximation bear close resemblance to the Bethe-Salpeter equation or the "overlapping" self energy terms discussed by Salam [106]. Walls and Gardiner point out that one aspect of these papers is confusing, namely that they start with V_{SR} in the R.W.A. and then obtain the same probability equations and exact solution as do W. and G. using W-WI. The analogy to the ladder approximation in fact comes in making the R.W.A. which ensures that only contributions from processes with a ONE photon intermediate state are included. Since all these approximations allow summation over a restricted class of physical processes to all orders, they are superior to perturbative techniques.

When R.W.A. is made H_0 and V_{SR} commute and so a subset of total eigenvalues of H_0 may be taken as a basis for the complete system and the use of this subset corresponds to making the ladder approximation.

When an atom emits spontaneously into a SINGLE field mode Walls [107] has found that R.W.A. is equivalent to ignoring a frequency shift $\delta\omega \cdot \kappa^2/2\omega$ and is therefore valid for $g \ll \omega$ as pointed out also by Louisell [35]. Agarwal [102] derived a master equation for spontaneous emission without making R.W.A. and found an identical expression for the radiation rate but a different expression for the atomic dipole moment. This difference is negligible for $g \ll \omega$. These two corrections are thus very small and W. and G. point out that maybe the approximations made in deriving the master equations are perhaps of same order of magnitude as those involved in making the R.W.A.

In ref. 120 R.W.A. and exact solutions are compared (for parametric amplifier and frequency converter etc.) and the validity of R.W.A. is established. It is likely that its validity could be established also for the multilevel atom of Chapter IV (see Appendix IV for equations of motion when R.W.A. is not assumed).

8.4 Broad spectrum limit

In assuming the loss oscillators to have a broad spectrum of frequencies, ω_γ, we satisfy one of the criteria necessary for the Markoff approximation to be valid so that it can be used when desired. Since the frequencies, ω_γ, are also closely spaced, $\sum_\gamma$ can be replaced by integrals where desired. Lehmberg [1] also assumes the emitted photons decay into broad bands of closely spaced modes. He considers more than one such band, each one being associated with a different transition, whereas we only ever consider one such band characterised by a frequency ω_γ or ω_ℓ.

8.5 Atom at origin

The atom is assumed to be at the origin of coordinates, e.g. at the nucleus of the atom. Fiutak [100] has pointed that this is a long wavelength assumption. We use it because the position of the atom is of no relevance when the only coupling between atoms we consider is the radiation from their transitions.

8.6 Mixing of states

In reference 7, the mixing of states, as a result of the damping mechanism, is assumed and this is given as a reason for the necessity of using the density operator technique.

Newstein [65] criticizes Bergmann's approach [108] to the problem of spontaneous emission from a 2-level system with incident beams of radiation which are initially either (a) in a coherent state or (b) in a n-photon state. Bergmann uses two approximations; (i) he retains only diagonal elements of the field time-development operator (in order to avoid P.Th.) and (ii) he restricts his equations of motion to material 2-level systems which cannot develop into mixed states. Newstein says that it is necessary to consider the effect of mixed states and for large fields the main features of the spontaneously radiated power spectrum are simple related to the time development of these states.

[N.B. Newstein's paper [65] is also of interest because it criticizes two other approaches to the problem of spontaneous emission in the presence of a driving field. Firstly, he says that the technique of treating the interaction Hamiltonian V_{SR} as a perturbation of the full Hamiltonian H is not valid here since it relies on the fact that the frequency associated with the interaction energy of the atomic currents with the e.m. field is $<<$ the resonance frequency. Secondly, he says that the procedure adopted by Rautian and Sobelman [33] for the solution is only valid for relatively small initial field energies. R. and S. consider that the material system is coupled to MANY modes of the radiation field R but that initially only ONE mode of the field is in a high-energy eigenstate. They obtain a solvable FINITE set of equations for the atom field probability amplitudes by truncating the infinite system to a small number of multiphoton processes.]

8.7 Ordering of Operators

We pointed out in the introduction that normal ordering enables the solutions of problems involving non-conservative systems without cumbersome iterated solution. In ref. 109, the authors show that the ordering of commuting atomic and field operators results in attributing a different origin to the radiative line shifts and widths in spontaneous emission, i.e. in fact whether it should be attributed to vacuum field fluctuations or to quantum electrodynamic radiation reaction.
They show that when normal ordering is used the radiative correction can be interpreted as entirely due to radiation reaction effect but when symmetric ordering is used the radiative correction can be interpreted as entirely the effect of vacuum field fluctuations. If in obtaining the average of the atom energy operator we use normal ordering then the decay would be considered entirely due to radiation reaction effect, but there is no ordering which would attribute it entirely to a vacuum fluctuation effect. They come to the conclusion at the end of the paper that as there is some arbitrariness over the interpretation of physical processes and as this

is equivalent to the ordering of commuting operators it must imply that the interpretations are closely related, although not fully equivalent, as the net process cannot be exclusively described in terms of vacuum fluctuations. On the other hand spontaneous emission can be entirely attributed to radiation reaction. Hence if one does not use normal ordering one must remember to attribute radiative corrections to field fluctuations as well as to radiation reaction.

8.8 Markoff approximation (M.A.)

In von Foester's paper he deals with the equations of motion for the dynamical operators of the system (written in the Heisenberg picture). He uses the R.W.A. and calculates equation of motion for $A(t)$ and $\tau(t) = A^+A - AA^+$ and points out that these cannot be solved exactly and that this is desirable since if they were then they would describe entirely reversible behaviour whereas damped behaviour is quite irreversible and can appear only as an approximate solution. He uses an approximation which makes the interaction between the atom and the radiation field a Markoffian process; the field at time t depends on the values of parameters only at t and not at earlier times. He does this by using an approximate expression which results in the field becoming so complex that it immediately loses all memory of its previous state, i.e. instead of writing

$$\sum_q g_q^2 \exp[-i\omega_q(t-t')]$$

we write

$$\Gamma(t-t')$$

where

$$\left.\begin{matrix}\Gamma(t-t') \\ \Gamma(t-t')\eta(t-t')\end{matrix}\right\} \begin{matrix} = \sum_q g_q^2 \exp[-i\omega_q(t-t')]\,\eta(t-t') \\ = [A_0(t'), A_0(t')]\,\eta(t-t')\end{matrix}$$

where $A_0(t) = \sum_q g_q a_q(0) \exp(-i\omega_q t)$

and $\eta(t)$ is the Heaviside unit step function.

$\Gamma(t)$ has a broad and fairly flat spectrum:
[This is equivalent to our assumption of closely spaced modes which enables $\sum_q g_q^2$ to be replaced by $\int_0^\infty d\omega_q\, \rho(\omega_q)\, g^2(\omega_q)$, where $g^2(\omega) = (\overline{g^2})_{\omega=\omega_q}$. The effective bandwidth of $\rho(\omega_q)\, g^2(\omega_q)$

is $\Delta\omega_q \sim \pi/\delta t$, where δt is the autocorrelation time. The most important contributions to the integral are those for which $t - t' \leq 2\pi/\Delta\omega_q$. $\Delta\omega_q \sim \omega_0$, $\therefore \delta t \sim 2\pi/\omega_0$, i.e. only a few cycles duration. We assume anyway that

$\delta t <<$ the time for appreciable secular changes in the atomic states and so

$$P_q(t') \simeq P_q\, e^{-i\omega_0(t-t')}$$

in the lowest approximation, to a high degree of accuracy, so that

$$\int_0^t dt' \sum_q g_q^2\, e^{-i\omega_q(t-t')} A(t') = (\tfrac{1}{2}\gamma - i\Omega) A(t) \quad \text{for times } t >> \omega_0^{-1}$$

This is an equivalent expression arrived at without the direct assumption of a Markoff process.]

ω_q does not vary rapidly with q, so that $\Gamma(t)$ is quite sharply peaked near t = 0 and very small elsewhere. If it varies much more rapidly than the slowly varying operator A(t), then

$$\int_0^t dt'\, \Gamma(t-t')\, A(t') \simeq A(t) \int_0^t dt'\, \Gamma(t-t')$$

$$= (\gamma + i\nu) A(t)$$

where $$\int_0^t dt'\, \Gamma(t-t') = \gamma + i\nu$$

which thus removes dependence of the field on earlier times t' so that

$$A(t) = \sum_q g_q a_q(t) = \sum_q g_q a_q(0) \exp(-i\omega_q t) - i(\gamma + i\nu) A(t)$$

The approximation for the above integral is shown by von Foerster to be equivalent to the W-W approximation and is discussed in detail. He argues that ν, the frequency shift, vanishes to a very good approximation since he assumes $g(\omega)^2 \rho(\omega)$ is nearly constant in the region near $\omega = 0$.

According to Louisell p. 184 of 636: "if we assume a system to be Markoffian we mean that its future behaviour is determined by the present and not the past and this assumption is valid since damping destroys knowledge of the past. Mathematically, we therefore replace

$$\rho^{(s)}(t') = \mathrm{Tr}_R\{\rho(t')\}$$

by its present value $\rho^{(s)}(t)$.

The Markoff approximation has been done crudely and instead one should take a coarse grained average on a time scale t where

system damping time $\gg$ t $\gg$ reservoir correlation time.

t should be long enough to contain many cycles of the undamped system motion. Then we may let the upper limit of the integration over t' go into infinity and derive the master equation describing the statistical behaviour of the system when coupled to a reservoir under the M.A. after assuming V_{SR} can be written

$$V_{SR} = \sum_i F_i Q_i$$

where F_i are reservoir operators,

and Q_i are system operators. [In fact Louisell and Marburger [110] eq. 34, p. 351 $\rho_I(t')$ is replaced by $\rho_I(t)$ and in addition assume that $\rho_I(t)$ has the form

$$\rho_I(t) = \mathrm{Tr}_R\{\rho(t)\}\rho^{(R)}(0) + \Delta\rho_I$$

where $\Delta\rho_I$ is at most of order V_{SR} since if $V_{SR} = 0$, $\rho_I(t) = \rho^{(s)}(t)\rho^{(R)}(0)$.

I indicates the interaction picture

$$\rho_I^{(s)}(t) = e^{i/\hbar H_s t}\rho^{(s)}(t)e^{-i/\hbar H_s t}$$

$$= \mathrm{Tr}_R\,\rho_I(t)$$

$$\rho_I(t) = e^{i/\hbar H_0 t}\rho(t)e^{-i/\hbar H_0 t}$$

They then go on to ignore all quantities of order higher than second in V_{SR}. Their method contains only 2 basic assumptions

1) memory of the system is destroyed by its interaction with the reservoir,

2) this interaction is sufficiently weak that its effects need only be considered to 2nd order in P.T. This we don't assume.]

When we use the Markoff approximation in Chapters V, VI, VII we use the former procedure not the latter.

According to Haake p. 508[36] when the adibatic of Markoff approximation is used with reference to the laser there are three objections to its use:

(1) The "proper" laser system (active atoms + field) can be treated as undergoing a Markoff process BUT the field alone as a subsystem cannot. This is a familiar argument in the theory of stochastic processes. We keep to this limitation as we apply the M.A. to the TOTAL DENSITY operator for the atomic system and reservoir (or field) though in our case this is not a laser system.

(2) Since we may intuitively reason that active atoms produce indirect interaction between PHOTONS of the lasing field mode he reasons that an exact elimination of atomic variables in laser theory must lead to a retarded photon interaction in order for the results to be physical.

[In his paper[112] Haake points out that Pauli type 1st order differential equations may be obtained under the following conditions:

(1) Initially there are no correlations between S and R. (We assume this).

(2) Internal correlations in the bath R are characterised by correlation times the relaxation times of the system S arising from its interaction with R. This amounts to assuming that the energy levels of R are closely spaced, which we do, and that the properly defined "strength function"-density of eigenfrequencies of the degrees of freedom of R, $-\rho(\omega_q)$, is a smoothly varying function of frequency, especially near eigenfrequencies of S.

(3) S is observed at times t >> internal bath correlation times and, if R is finite, << POINCARÉ recurrence times for R.

(4) Interaction between S and R is sufficiently weak to allow for a simplification of the integral kernal leaving the latter correct up to 2nd order in the interaction (Born approximation B.A.). This derivation of the Pauli equations avoids 2nd order perturbation treatment and the repeated random phase approximation.

Typical non-Markoffian effects in S may be treated from an exact master equation if assumptions (2) and (3) are not made.

We see therefore that in fact though in Chapters II and III a Markoffian process is not assumed the assumptions made are equivalent to that assumption.

In his paper [112] he goes on to discuss and justify the B.A. since it is the major approximation in this method for turning exact master equations into eventually solvable ones. He finds that the B.A. is valid

(1) if the "strength function" $\rho(\omega_q)$ does not degenerate to one or more extremely sharp and high lines. One must not assume a white spectrum. (N.B. This is only assumed if one uses the method which involves the Markoff approximation.)

(2) that temperature of R is not too low,

(3) contributions to the Dyson series stemming from "overlapping" 2nd order and from all higher order thermal equilibrium bath correlations are negligibly small compared with those from non-overlapping 2nd order correlations. (N.B. This does not amount to suppressing internal bath correlations.)]

We consider an e.m. field in which there is no retarded photon interaction and only consider interaction between the atom and the field.

(3) The essential condition for the elimination of heat baths in the Markoff approximation is that the heat bath should have a WHITE or at least an extremely broad band energy spectrum. Although this is just the opposite condition sought after in a laser it is the condition sought after in the e.m. field we choose to act as a damping reservoir. The damping reservoir in our case is considered as a heat bath H in thermal equilibrium with an ensemble of harmonic oscillators ε which together are equivalent to the PHONONS. These phonons spread into a broad band of frequencies ω_q about the original frequency ω_p when the atom is coupled in the solid and in $\sum_q$ q varies from $1 \longrightarrow 10^{23}$ over this band. In our case therefore this objection to the use of the M.A. does not apply.

Finally we would like to point out that the reservoir (namely the quantised e.m. field) is thus assumed to stay in its initial pure state by virtue of using the M.A. If the field is very strong, i.e. contains a large number of photons depletion is only very slight in comparison to the total number of photons and so this assumption is valid for a strong field, N, such as we choose ($N \approx 10^{23}$ the number of degrees of freedom of the bath).

Further in Sasada's paper [111] which criticizes Lehmberg's method and produces a better one he considers only the time evolution of the intensity and not its frequency dependence, as we do. He is also mainly concerned with the non-Markoffian effect on superradiance from an assembly of harmonic oscillators, whereas we ignore superradiance entirely. He points out that the non-Markoffian effect retards a correlated motion which is responsible for superradiance, of the atomic system induced by the photon-atom coupling, and it consequently modifies a feature of superradiance. In fact it modifies the shape of superradiant pulses and the photon statistics.

8.9 Why t' is chosen to be arbitrary in Chapters V and VII.

In Chapters V and VII we are concerned with averaged effects, i.e. instead of considering a particular process of emission or absorption we consider the averaged effect of all possible multiphoton processes. For this reason we do not assign the value of zero to the initial time t' since this would imply that we know the initial state of the atom, i.e. the starting point of the process, whereas, in fact, since we are considering that there are several photons in the vicinity of the atom, we therefore cannot tell whether a particular photon is being emitted or absorbed at a particular instant in time, and also we are not able to ascertain the initial state of the atom. This uncertainty is as a result of the fact that there is a continuous series of processes. It has also been given earlier as a reason why there is a need for the statistical approach implementing density matrices. The resulting spectral profiles calculated when t'   0 do not represent just one process of emission (as in Chapter VI) or absorption but the overall effect of all possible processes when several photons are in the vicinity of the atom. The spectral profiles represent the power spectrum of the scattered field as in Mollows paper[9]. In Mollow's paper[52] he calculates emission and absorption spectra separately by considering the state of the atom is known at initial time $t' = 0$.

In contrast to this approach, if we were to have attempted to use the time-dependent perturbation theory to solve the problems, then we would have had to have known in which state the atom was at the initial time. We would then have to have considered each multiphoton process separately. In Mollow's paper[9], on the other hand, we consider the initial atomic state to be arbitrary and then, in order to find the initial density matrices, we first of all evaluate their equilibrium values attained at $t \rightarrow \infty$ and then evaluate these at $t = t'$ the initial time. In this way dependence on t' appears only as the harmonic factor $e^{-i\omega t'}$ in certain terms where ω is the frequency of the driving field. These factors cancel out when the spectrum is finally calculated so that the value of t' is never required. In this way the overall effect of several multiphoton processes is calculated.

It is owing to the fact that the state of the atom at any time is uncertain that the wave functions become mixed as a result of the interaction. New wave functions have to be defined which are a mixture of the old ones. This mixing is considered both by Newstein[67] and by several Russian authors[86], but is erroneously ignored by Bergmann[103] who is criticized by Newstein on this point. This mixing as a result of damping has been pointed out several times as being mentioned by Mollow and Miller[7]. They make the point also on p. 473 that in weak fields the atomic state remains pure and one can just add a phenomenological damping term to the equation of the time derivative of the excited state.

Stroud[98] has developed an alternative approach which is quantum electrodynamical (Q.E.D.) in nature but does not involve the use of time-dependent perturbation theory. Its disadvantage is that it is a very large method since he first of all considers the splitting of each level and then goes on to consider all possible transitions between the resulting split levels, considering each multiphoton process separately. His method also has the disadvantage of only being able to take into account the process of spontaneous emission and not overall processes.

Chang & Stehle [115] also use (Q.E.D.) theory for the purpose of doing some calculations on multiple-quantum transitions in magnetic resonance. They find that their results agree better with the experimental working of Kusch [116] than do those obtained by Salwen [116,117] using semiclassical theory. They therefore conclude that semiclassical theory is inferior to their own work in interpreting this and other experiments but Pegg & Series [118] assert that the papers of Chang & Stehle do not constitute a proper comparison with semiclassical theory because Salwen's calculations and other results cited by C. & S. are admittedly only approximate solutions to the semiclassical equations of motion, and have been superseded by more recent work [119] in this field. Also Pegg and Series draw attention to the fact that C. & S. do not specify their e.m. field in sufficient detail.

Pegg and Series say that in two particular situations an exact semi-classical solution can be obtained:

1) in the "rotating-wave" situation and (2) when the field consists of a linearly oscillating component and a parallel static component which may be zero. Chang and Stehle do not specify the polarisation of the fields they discuss but it appears they deal with linearly oscillating fields (which exclude case (1)) and that interactions having diagonal matrix elements (case (2) are of no interest to them. For this reason their strictures against the exact semiclassical solutions are misplaced.

Another point made by Pegg and Series is that semi-classical and Q.E.D. treatments do not only agree in the low-frequency low-field region, i.e. "in those systems where transitions involve the emission or absorption of a single photon" as stated by Chang & Stehle but also for strong fields where the photon number $n \gg 1$ according to the correspondence principle.

The main difference (apart from virtual processes ignored by C. & S.) between semiclassical and quantised field theories lies in the treatment of spontaneous emission [120]. The interaction matrix elements which determine the evolution of the atomic state vector under stimulated emission and absorption are identical in the two theories; hence the predictions of the theories must coincide for those situations in which spontaneous emission plays no significant role.

N. R. and F.[86] show that the external field changes the spectrum of spontaneous emission of a quantum system significantly and interprets them as the result of mixing of stationary states of the isolated atom in the external field. He points out that any discussion based on the "splitting" of levels of an isolated atom can lead to a seriously wrong conclusion since unusual interference phenomena appear. They are interested in the case when mixing of states takes place after the excitation of the system, while at the initial instant the atom is in some stationary state.

In Stroud's case[98], if there are say 15 photons in the vicinity of the atom, he considers what happens to one at a time but in Lehmberg's[1,2] and Mollow's[7,9] papers they consider the interference caused when the next photon arrives within 10^{-8} sec. so that the states become mixed. The perturbation approach is valid only for times between 10^{-15} sec. and 10^{-8} sec. and not $> 10^{-8}$ sec. where 10^{-8} sec. is the approximate atomic lifetime. For longer times other methods are necessary as pointed also in the introduction Sec. B.

8.10 Limitations on Lehmberg's method.

In Lehmberg's 1st paper[1] atoms are confined to a region SMALL in comparison to the transition wavelength i.e.

but the formalism of the 2nd paper(2), which we use, removes this restriction. It also takes into account the frequency shifts due to the e.m. coupling between the atoms and . These we ignore since we are only interested in rarified gases in which the e.m. coupling between atoms is negligibly small. The only limitation on the size of the ensemble of systems in (2) is expressed by condition (13),

i.e.

or the time required for a signal to cross the system being small in comparison with the time t required for appreciable (secular) changes in the atomic levels. Since we only consider a single atom in our case is replaced by R the distance of the atom from the observation point.

For a system driven by a sufficiently strong resonant pulse is determined by the field amplitude and pulsewidth and the problem can be adequately treated by semi-classical theory.

Lehmberg's analysis precludes application to macroscopic lasing materials but the formalism can still be applied to multi-atom systems extending over many wavelengths and still be capable to developing pornounced directional effects.

Von Foerster's paper also uses a method of approach similar to Lehmberg's and provides a backing for it.

8.11 The two ways of considering the driving field in Chapters V, VI, VII.

In Chapters V, VI, VII we show that whether we use Glauber's notation(48,49,50) or introduce driving fields as additional classical terms makes no difference to the form of the resulting equations although Glauber's approach is more consistent as it is truly quantum mechanical. In the latter the driving fields can be considered within the formalism by making initial photon states non-zero. According to von Foerster(43) strong fields, such as the laser fields we consider, should be represented classically. When we use the quantum mechanical approach and consider ONE mode in each case the results take the same form as when we consider the field classically. This is because in the beginning the strength of the field is not stipulated and is left as a variable parameter in the resulting formula.

According to Newstein [68] when a material system S interacts with a relaxation mechanism R and the e.m. field E, the interaction of S with E is describable as the sum of two terms;

1) the first term gives the effect of a prescribed classical field, and
2) the second gives the effect of a WEAK quantum field that is responsible for spontaneous emission. This interaction is treated perturbatively. The driving field is treated classically in his paper. In fact when the incident field is of arbitrary value it can be treated non-perturbatively.

APPENDICES

APPENDIX I

Heisenberg equations of motion, derived from the Hamiltonian in which R.W.A. is not assumed, for the 2-level atom (cf. ref 2)

If we use the interaction Hamiltonian as given in equation (1.B.103), i.e.

$$H = \hbar\omega_0 A^+A + \sum_{\ell\sigma} \hbar\omega_\ell a^+_{\ell\sigma} a_{\ell\sigma} - \sum_{\ell,\sigma} \hbar g_{\ell\sigma} \left(S a_{\ell\sigma} e^{ik_\ell \cdot r} + a^+_{\ell\sigma} S e^{-ik_\ell \cdot r} \right) \tag{I.1}$$

instead of eq. (1.B.105), obtained under R.W.A., then we can show that the equations for $a_{\ell\sigma}(t)$, A(t) and Q(t) will be modified by the underlined terms so that we have:-

$$\dot{a}_{\ell\sigma}(t) = -i\omega_\ell a_{\ell\sigma} + i g_{\ell\sigma} e^{-ik_\ell \cdot r} \left(A(t) + \underline{A^+(t)} \right) \tag{I.2}$$

$$\left.\begin{aligned} \dot{A}(t) = & -i\omega_0 A(t) - i2(Q(t) - 1/2) \sum_{\ell,\sigma} g_{\ell\sigma} e^{ik_\ell \cdot r} a_{\ell\sigma}(t) - i\sum_{\ell,\sigma} g_{\ell\sigma} e^{-ik_\ell \cdot r} a^+_{\ell\sigma} \\ & - i \sum_{\ell,\sigma} g_{\ell\sigma} e^{-ik_\ell \cdot r} \underline{a^+_{\ell\sigma}\, 2(Q(t) - 1/2)} \end{aligned}\right\} \tag{II.3}$$

$$\dot{Q}(t) = -i(A(t) + A^+(t)) \sum_{\ell\sigma} g_{\ell\sigma} e^{ik_\ell \cdot r} a_{\ell\sigma}(t) - i\sum_{\ell,\sigma} g_{\ell\sigma} e^{-ik_\ell \cdot r} \underline{a^+_{\ell\sigma}}(t) (A(t) \underline{- A^+(t)}) \tag{II.4}$$

where there is no external driving field, as opposed to equations (2.2), (2.4), (2.5) of Chapter II when $\lambda' = 0$ and $r \to 0$ and $a_{\ell\sigma} e^{-i\varepsilon_\ell} \longrightarrow -a_{\ell\sigma}$, $a^+_{\ell\sigma} e^{i\varepsilon_\ell} \longrightarrow -a_{\ell\sigma}$. The terms underlined are high frequency (HF) ones since they originate from the high frequency terms in the Hamiltonian. (N.B. $A \sim e^{-i\omega_0 t}$, $A^+ \sim e^{i\omega_0 t}$ $\therefore$ $Q = A^+A \sim e^0 = 1$; $a_{\ell\sigma} \sim e^{-i\omega_\ell t}$, $a^+_{\ell\sigma} \sim e^{i\omega_\ell t}$ $\therefore$ $a^+_{\ell\sigma} Q \sim e^{i\omega_\ell t}$ and $a^+_{\ell\sigma} A^+(t) \sim e^{i(\omega_0 + \omega_\ell)t}$ where $\omega_\ell \sim \omega_0$ and hence A^+, $a_{\ell\sigma} Q$, $a^+_{\ell\sigma} A^+$ are all HF terms.)

When we go through the same procedure as in Chapter II, we finally obtain the following two equations:-

$$\left.\begin{aligned} \dot{A}(t) = & -i2(Q(t) - 1/2) q(r) - i2q^+(r)(Q(t) - 1/2) \\ & - (1/2\gamma + i(\omega_0 - \Omega)) A(t) + (1/2\gamma + i\Omega) A^+(t) \end{aligned}\right\} \tag{II.5}$$

$$\left.\begin{aligned}\dot{Q}(t) = & -i\left(\underline{A(t)} - A^{+}(t)\right)\underline{q(\underline{r})} - i\underline{q^{+}(\underline{r})}\left(A(t) + \underline{A^{+}(t)}\right) \\ & - \gamma Q(t)\end{aligned}\right\} \quad \text{(II.6)}$$

where $\Omega = \Omega_{-} - \Omega_{+}$ (II.7)

replaces Ω_{-}

and underlined terms result from HF terms in the Hamiltonian, viz. $q^{+}(\underline{r})Q(t) \sim e^{i\omega_{e}t}$, $A^{+}(t) \sim e^{i\omega_{e}t}$, $A(t)\,q(\underline{r}) \sim e^{-i(\omega_{k}+\omega_{e})t}$

$q^{+}(\underline{r})\,A^{+}(t) \sim e^{i(\omega_{k}+\omega_{e})t}$

(N.B. there is no γ_{+} corresponding to Ω_{+} since $\int_{0}^{\infty} d\omega\,\omega^{3}\,\delta(\omega+\omega_{e}) = 0$ and $\omega = -\omega_{e}$ does not lie within the limits of integration.)

If we neglect the high frequency terms at this stage, since they will have only a very small effect, we obtain equations

$$\dot{A}(t) = i\,2\left(Q(t) - \tfrac{1}{2}\right)q(\underline{r}) - \left(\tfrac{1}{2}\gamma + i(\omega_{k} - \Omega)\right)A(t) \quad \text{(II.8)}$$

which is the same as equation (23) of Lehmberg (ref. 2) when $\alpha = \beta$ and $\sigma = Q$, i.e. our A.

$$\dot{Q}(t) = iA^{+}(t)\,q(\underline{r}) - iq^{+}(\underline{r})\,A(t) - \gamma Q(t) \quad \text{(II.9)}$$

which is the same as equation (23) of Lehmberg if   is replaced by Q throughout.

Thus, this more rigorous treatment only results in negligible HF terms and a frequency shift modification which can be neglected as it is not our main concern. We are therefore justified, on this basis, in using the R.W.A., from the start, in Chapter II.

APPENDIX II

Comparison of the notation in Chapter II with that of method (i) in Chapters V, VI and VII where the external field is treated classically

In Chapters V, VI, VII we consider the external perturbations to be time dependent and, in method (i), based on Mollow's treatment[7,9] of the perturbation, let $E_D(t)$ be the signal generator. This feeds the signal continuously into the cavity. In this case the Hamiltonian for the interaction between the system and the signal is

$$H_{ID}(t) = V_{SE}(t) = \hbar\lambda\{A^+(t)\mathcal{E}_D(t) + \mathcal{E}_D^*(t)A(t)\} \tag{II.1}$$

where λ is a c-number coupling parameter (according to ref. (35), p. 272-3). In Mollow and Miller's paper[9], the classical external perturbation E can be presumed to be a classical (c-number) electric field $E_D(t)$, with positive and negative frequency parts $\mathcal{E}_D(t)$ and $\mathcal{E}_D^*(t)$ respectively and polarisation specified by unit vector $\hat{e}_{oD}$ where

$$E_D(t) = \{\mathcal{E}_D(t) + \mathcal{E}_D^*(t)\}\hat{e}_{oD} \tag{II.2}$$

Then the coupling between the atom, S, and field, F, is taken to be

$$\left.\begin{aligned} V_{SE} &= -E_D(t).\{p\,A^+(t) + p^*A(t)\} \qquad \text{(N.B. in our calculations we take } p \text{ to be real)} \\ &= -\hat{e}_{oD}.p\{\mathcal{E}_D(t)+\mathcal{E}_D^*(t)\}\{A^+(t)+A(t)\} \quad \text{for } p \text{ real} \\ &= -\hbar\lambda\{A^+(t)\mathcal{E}_D(t) + \mathcal{E}_D^*(t)A(t) + \mathcal{E}_D(t)A(t) + A^+(t)\mathcal{E}_D^*(t)\} \end{aligned}\right\} \tag{II.3}$$

$$\text{where } \lambda = \frac{\hat{e}_{oD}.p}{\hbar} \text{ and is real} \tag{II.4}$$

Now $A(t) = A(0)e^{-i\omega_0 t}$ (cf. (1.B.83))

and $A^+(t) = A^+(0)e^{i\omega_0 t}$,

and also the driving field is assumed to be harmonically varying at frequency $\omega_D \sim \omega_0$, the resonant frequency, so that

$$\left.\begin{aligned} \mathcal{E}_D(t) &= \mathcal{E}_{oD}\,e^{-i\omega_D t} \\ \mathcal{E}_D^*(t) &= \mathcal{E}_{oD}\,e^{i\omega_D t} \qquad \text{where } \mathcal{E}_{oD} \text{ is assumed to be real} \end{aligned}\right\} \tag{II.5}$$

$$\left.\begin{aligned} \therefore V_{SE} &= -\hbar\lambda\{A^{+}(0)\mathcal{E}_{oD}e^{i(\omega_0-\omega_D)t} + \mathcal{E}_{oD}^{*}A(0)e^{-i(\omega_0-\omega_D)t} \\ &\qquad + \mathcal{E}_{oD}A(0)e^{-i(\omega_0+\omega_D)t} + A^{+}(0)\mathcal{E}_{oD}^{*}e^{i(\omega_0+\omega_D)t}\} \\ &\doteq -\hbar\lambda\{A^{+}\mathcal{E}_D + \mathcal{E}_D^{*}A\} \\ \text{or } &-\hbar\lambda\{A^{+}(0)\mathcal{E}_D'(t) + \mathcal{E}_D'^{*}(t)A(0)\}, \\ &\qquad \text{where } \mathcal{E}_D'(t) = \mathcal{E}_D(t)e^{i\omega_0 t} \end{aligned}\right\} \quad \text{(II.6)}$$

since terms $\mathcal{E}_D A$ and $A^{+}\mathcal{E}_D^{*}$ are rapidly oscillating at a frequency $\sim 2\omega_0$ and so can be ignored in comparison with the terms $A^{+}\mathcal{E}_D$ and $\mathcal{E}_D^{*}A$ oscillating at $\omega_0-\omega_D\sim 0$ and which are therefore practically d.c. (cf. discussion of R.W.A. after eq. (1.B.92). Thus the expression in ref. (35) i.e. eq. (II.1) assumes the R.W.A. and is valid for weak coupling.

We shall consider V_{SE} for a classical time independent perturbation to be given by

$$V_{SE} = \hbar\lambda' A^{+}e^{-i\varphi} + \hbar\lambda'^{*}Ae^{i\varphi} \quad \text{(II.7)}$$

where we have introduced an arbitrary phase ϕ which will be seen to have no effect on the results, so that comparing terms we see that

$$-\lambda\mathcal{E}_D(t) \longrightarrow \lambda' e^{-i\varphi}$$

$$-\lambda\mathcal{E}_D^{*}(t) \longrightarrow \lambda'^{*}e^{i\varphi}$$

When $\omega_D = 0$, as in Chapter II, we have

$$\left.\begin{aligned} \lambda' &= -\lambda\mathcal{E}_{oD}e^{i\varphi} = -\left\{\frac{\hat{e}_{oD}\cdot p\ \mathcal{E}_{oD}}{\hbar}\right\}e^{i\varphi} \\ \lambda'^{*} &= -\lambda\mathcal{E}_{oD}e^{-i\varphi} = -\left\{\frac{\hat{e}_{oD}\cdot p^{*}\mathcal{E}_{oD}}{\hbar}\right\}e^{-i\varphi} \end{aligned}\right\} \quad \text{(II.8)}$$

so that we can identify c-number coupling parameter λ' as above, when the perturbation is an electric field.

N.B. $$|\lambda'|^2 = (\lambda\mathcal{E}_{oD})^2 \text{ or } \tfrac{1}{4}\bar{G}^2 \quad \text{(II.9)}$$

in the notation used in Chapters V, VI, VII and is related to the strength of the perturbation.

In Mollow's paper[9] he expresses the total electric field as the sum of the classical expression (2.2) and a quantum mechanical field expanded in a region of volume V, as in Newstein's paper discussed in Chapter VIII[68]. I.E. using eq. (1.B.97) for the quantum mechanical part we obtain

$$\left.\begin{aligned} E^{+}(\underline{r},t) &= \mathcal{E}_D(r,t)\,\hat{e}_{cD} + \sum_{\ell} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\;\hat{e}_{\ell\sigma}\,a_{\ell\sigma}\,e^{ik_\ell r} \\ E^{(-)}(\underline{r},t) &= \mathcal{E}_D^{*}(r,t)\,\hat{e}_{cD} + \sum_{\ell\sigma} \sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\;\hat{e}_{\ell\sigma}\,a^{+}_{\ell\sigma}\,e^{-ik_\ell r} \end{aligned}\right\} \qquad (II.10)$$

where, in the resonant approximation, we may express the electric field as the sum of positive and negative frequency parts as in (II.2), viz.

$$\underline{E}(r,t) = \{\underline{E}^{(+)}(r,t) + \underline{E}^{(-)}(r,t)\} \qquad (II.11)$$

Then the interacting Hamiltonian is

$$H_I(t) = -\underline{d}(t)\cdot\underline{E}(0,t) \qquad (II.12)$$

when the atom is at the origin of coordinates.

is the dipole moment operator for the atom expressed in terms of the dipole matrix element

$$\underline{p} = \langle j|\underline{d}|i\rangle$$

as
$$\underline{d}(t) = \underline{p}A^{+}(t) + \underline{p}\,A(t)$$

N.B. we write $\underline{p} = \langle j|e\underline{r}|i\rangle$ and assume it to be real so that

$\underline{d}(t) = \underline{p}(A^{+}(t) + A(t))$

$$\left.\begin{aligned} H_I(t) = -A^{+}(t)\Big\{&\mathcal{E}_D(t)\,\underline{p}\cdot\hat{e}_{cD} + \sum_{\ell\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\;\underline{p}\cdot\hat{e}_{\ell\sigma}\,a_{\ell\sigma} \\ &+ \mathcal{E}_D^{*}(t)\,\underline{p}\cdot\hat{e}_{cD} + \sum_{\ell\sigma}\sqrt{\frac{2\pi\hbar\omega_\ell}{V}}\;\underline{p}\cdot\hat{e}_{\ell\sigma}\,\hat{a}_{\ell\sigma}\Big\} \\ &+ \text{H.C.} \end{aligned}\right\} \qquad (II.13)$$

$$\left.\begin{aligned} \text{i.e. } H_I(t) &= -A^{+}\Big\{(\mathcal{E}_D(t)+\mathcal{E}_D^{*}(t))\hbar\lambda + \sum_{\ell\sigma}\hbar g_{\ell\sigma}(a_{\ell\sigma}+a^{+}_{\ell\sigma})\Big\} + \text{H.C.} \\ &= -A^{+}\Big\{\mathcal{E}_D(t)\hbar\lambda + \sum_{\ell\sigma}\hbar g_{\ell\sigma}a_{\ell\sigma}\Big\} + \text{H.C.} \quad \text{in the resonant approximation} \end{aligned}\right\} \qquad (II.14)$$

$$= -\hbar\lambda\{A^{+}\mathcal{E}_{D}(t) + \mathcal{E}_{D}^{*}(t)A\} - \sum_{\ell\sigma}\hbar g_{\ell\sigma}(A^{+}a_{\ell\sigma} + a_{\ell\sigma}^{+}A)$$

(N.B. $g_{\ell\sigma} = \sqrt{\frac{2\pi\omega_{\ell}}{\hbar V}}\,\hat{e}_{\ell\sigma}\cdot\mathcal{P}$) (II.14) cont'd

$$= V_{SR} + V_{SE}$$

N.B. In Chapter II we introduce a phase factor into the expression for V_{SR} as well as into that for V_{SE} but these are seen to have no effect on the results we derive.

APPENDIX III (ref. 121)

Algebraic solution of the cubic equation

This appendix will be devoted to a derivation of the solutions of the general cubic equation

$$s^3 + as^2 + bs + c = 0 \qquad \text{(III.1)}$$

In order to reduce this equation to its standard form we remove the term by making the change of variable

$$s = y - 1/3a$$

The resulting equation is

$$y^3 + (-1/3a^2 + b)y + (2/27a^3 - 1/3ab + c) = 0$$

or

$$y^3 + 3Hy + G = 0 \qquad \text{(III.2)}$$

where $H = 1/3(-1/3a^2 + b)$

and $G = 2/27a^3 - 1/3ab + c$

In order to solv eq. (III.2), we assume that

$$y = u + v \qquad \text{(III.3)}$$

so that

$$y^3 = (u^3 + v^3) + 3uv(u + v)$$

Substituting in this last equation for $u + v$, from (III.3), we obtain the equation

$$y^3 - 3uvy - (u^3 + v^3) = 0 \qquad \text{(III.4)}$$

A comparison of (III.4) with (III.2) shows that

$$3uv = -3H$$

and $u^3 + v^3 = -G$

$$\left.\begin{array}{ll} \text{or} & u^3v^3 = -H^3 \\ \text{and} & u^3 + v^3 = -G \end{array}\right\} \qquad \text{(III.5)}$$

If v^3 is eliminated by substituting from the second of equations (III.5) into the first, then the quadratic equation in A^3 appears,

$$(u^3)^2 + Gu^3 - H^3 = 0$$

whose roots are

$$u^3 = \frac{-G \pm \sqrt{G^2 + 4H^3}}{2}$$

The solution for v^3 yields precisely the same values. However, in order to satisfy eq. (III.5), we choose

$$\left.\begin{aligned} u^3 &= \frac{-G + \sqrt{G^2 + 4H^3}}{2} \\ v^3 &= \frac{-G - \sqrt{G^2 + 4H^3}}{2} \end{aligned}\right\} \quad \text{(III.6)}$$

i.e. the positive square root term for u^3 and the negative for v^3. The opposite choice for the values of u^3 and v^3 simply interchanges their roles in what follows.

If the values of y and to be determined from eq. (III.6), it is necessary to find the cube roots of u^3 and v^3. Now we know that if $s^3 - d^3 = 0$ then the solutions for s are given by d, ω d and ω^2d, where $\omega = -\frac{1}{2} + \sqrt{3}/2\, i$ and $\omega = -\frac{1}{2} - \sqrt{3}/2\, i$ are the complex roots of unity. Hence, if one cube root of u^3 be denoted by α and one cube root of v^3 by β, the cube roots of u^3 are

α, $\omega\alpha$ and $\omega^2\alpha$, i.e. the roots of $u^3 - \alpha^3 = 0$,

whereas those of are

β, $\omega\beta$ and $\omega^2\beta$, i.e. the roots of $v^3 - \beta^3 = 0$.

It would appear that there are 9 choices for y, but values must be paired so that $uv = -H$. The only pairs that satisfy this condition are α and β, $\omega^2\alpha$ and $\omega\beta$, and $\omega\alpha$ and $\omega^2\beta$. Hence, the values of y are

$$\left.\begin{aligned} y_1 &= \alpha + \beta \\ y_2 &= \omega^2\alpha + \omega\beta \\ y_3 &= \omega\alpha + \omega^2\beta \end{aligned}\right\} \quad \text{(III.7)}$$

where $$\alpha = \left[\frac{-G + \sqrt{G^2 + 4H^3}}{2}\right]^{1/3}$$

and $$\beta = \left[\frac{-G - \sqrt{G^2 + 4H^3}}{2}\right]^{1/3}$$ (III.7) cont'd

The solutions of eq. (III.1) can be obtained from the values given in (III.7) by recalling that $s = y - {}^1/_3 a$. Thus

$$\left.\begin{aligned} s_1 &= y_1 - \tfrac{1}{3}a = (\alpha+\beta) - \tfrac{1}{3}a \\ s_2 &= y_2 - \tfrac{1}{3}a = (\omega^2\alpha + \omega\beta) - \tfrac{1}{3}a = -\tfrac{1}{2}\{(\alpha+\beta) + i\sqrt{3}(\alpha-\beta)\} - \tfrac{1}{3}a \\ s_3 &= y_3 - \tfrac{1}{3}a = (\omega\alpha + \omega^2\beta) - \tfrac{1}{3}a = -\tfrac{1}{2}\{(\alpha+\beta) - i\sqrt{3}(\alpha-\beta)\} - \tfrac{1}{3}a \end{aligned}\right\} \quad \text{(III.8)}$$

It is thus obvious that if <u>α, β, and a are real</u> s_1 is real and s_2 and s_3 are complex conjugates of each other so that the three roots can be written

$$\left.\begin{aligned} s_1 &= x_1 = (\alpha+\beta) - \tfrac{1}{3}a \\ s_2 &= x_2 - ix_3 = -\{[\tfrac{1}{2}(\alpha+\beta) + \tfrac{1}{3}a] - i\tfrac{\sqrt{3}}{2}(\alpha-\beta)\} \\ s_3 &= x_2 + ix_3 = -\{[\tfrac{1}{2}(\alpha+\beta) + \tfrac{1}{3}a] + i\tfrac{\sqrt{3}}{2}(\alpha-\beta)\} \end{aligned}\right\} \quad \text{(III.9)}$$

N.B. $$\left.\begin{aligned} \alpha &= \left[\frac{-(\tfrac{2}{27}a^3 - \tfrac{1}{3}ab + c) + \sqrt{(\tfrac{2}{27}a^3 - \tfrac{1}{3}ab + c)^2 + 4(\tfrac{1}{3}(\tfrac{1}{3}a^2 + b))^3}}{2}\right]^{1/3} \\ \beta &= \left[\frac{-(\tfrac{2}{27}a^3 - \tfrac{1}{3}ab + c) - \sqrt{(\tfrac{2}{27}a^3\ \tfrac{1}{3}ab + c)^2 + 4(\tfrac{1}{3}(-\tfrac{1}{3}a^2 + b))^3}}{2}\right]^{1/3} \end{aligned}\right\} \quad \text{(III.10)}$$

in terms of the known quantities a, b, and c.

Exact values of α and β cannot be found and in the text we use various approximations in order to find α and β and hence s_1, s_2 and s_3.

APPENDIX IV

The effect of H.F. terms in the Hamiltonian on the equations of motion for the multilevel-level atom

Assuming the Hamiltonian to be given by eq. (4.A.2) when H.F. terms are included i.e.

$$H = \hbar \sum_{x=1}^{l} \epsilon_x P_{xx} + \hbar \sum_{l\sigma} \omega_l a_{l\sigma}^{+} a_{l\sigma} - \hbar \sum \sum \sum g_{l\sigma,yz} \left\{ e^{i k_l \cdot r} (P_{zy} + \underbrace{P_{zy}^{+}}_{H.F.}) a_{l\sigma} + e^{-i k_l \cdot r} a_{l\sigma}^{+} (P_{zy} + \underbrace{P_{zy}^{+}}_{H.F.}) \right\} \quad \text{(IV.1)}$$

we obtain the following equations of motion

$$\dot{a}_{l\sigma} = -i\omega_l a_{l\sigma} + i \sum_{y=z+1}^{l} \sum_{z=1}^{l-1} g_{l\sigma,yz} e^{-i k_l \cdot r} (P_{zy} + P_{zy}^{+}) \quad \text{(IV.2)}$$

with formal solution

$$a_{l\sigma}(t) = a_{l\sigma}(0) e^{-i\omega_l t} + i e^{-i k_l \cdot r} \int_0^t dt' \sum_{y=z+1}^{l} \sum_{z=1}^{l-1} g_{l\sigma,yz} (P_{zy}(t') + P_{zy}^{+}(t')) e^{-i\omega_l (t-t')} \quad \text{(IV.3)}$$

$$\dot{P}_{mn}(t) = i\epsilon_{mn} P_{mn} - i \sum_{y=z+1}^{l} \sum_{z=1}^{l-1} \sum_{l\sigma} g_{l\sigma,yz} \left\{ e^{i k_l \cdot r} [S_{zy}, P_{mn}]_t a_{l\sigma} - e^{-i k_l \cdot r} a_{l\sigma}^{+} [P_{mn}, S_{zy}]_t \right\} \quad \text{(IV.4)}$$

$$\dot{P}_{mm}(t) = -i \sum_{y=z+1}^{l} \sum_{z=1}^{l-1} \sum_{l\sigma} g_{l\sigma,yz} \left\{ e^{i k_l \cdot r} [S_{zy}, P_{mm}]_t a_{l\sigma} - e^{-i k_l \cdot r} a_{l\sigma}^{+} [P_{mm}, S_{zy}]_t \right\} \quad \text{(IV.5)}$$

where $[S_{zy}, P_{mn}]_t = [S_{zy}(t), P_{mn}(t)]$ (IV.6)

[N.B. $m < n$]

Hence

$$\left.\begin{aligned} \dot{P}_{mn}(t) = i\epsilon_{mn} P_{mn} &- i \sum_{l\sigma} e^{i k_l \cdot r} \Big\{ \sum_{z=1}^{m-1} g_{l\sigma,mz} \underline{P_{zn}} - \sum_{y=n+1}^{l} g_{l\sigma,yn} \underline{P_{my}} \\ &+ \sum_{y=m+1}^{l} g_{l\sigma,ym} P_{yn} - \sum_{z=1}^{n-1} g_{l\sigma,nz} P_{mz} \Big\} a_{l\sigma} \\ &+ i \sum_{l\sigma} e^{-i k_l \cdot r} a_{l\sigma}^{+} \Big\{ \sum_{y=n+1}^{l} g_{l\sigma,yn} P_{my} - \sum_{z=1}^{m-1} g_{l\sigma,mz} P_{zn} \\ &+ \sum_{z=1}^{n-1} g_{l\sigma,nz} \underline{P_{mz}} - \sum_{y=m+1}^{l} g_{l\sigma,ym} \underline{P_{yn}} \Big\} \end{aligned}\right\} \quad \text{(IV.7)}$$

N.B. Underlined terms result from HF terms in the Hamiltonian.

It is now necessary to derive new expressions for

$\sum_{z=1}^{m-1}\sum_{k\sigma} g_{k\sigma,mz}\, a_{k\sigma}(t)\, e^{ik\cdot r}$ etc. using the new expression for $a_{k\sigma}(t)$.

$$\sum_{z=1}^{m-1}\sum_{k\sigma} g_{k\sigma,mz}\, a_{k\sigma}(t)\, e^{ik\cdot r} = \sum_{z=1}^{m-1}\sum_{k\sigma} g_{k\sigma,mz}\, a_{k\sigma}(0)\, e^{-i\omega_k t}\, e^{ik\cdot r}$$
$$+ i\sum_{z}^{m-1}\sum_{y=m+1}^{\ell}\sum_{z=1}^{\ell}\sum_{k\sigma} g_{k\sigma,yz}\int^{t} dt'\left(P_{zy}(t') + P^{+}_{zy}(t')\right) \times e^{-i\omega_k(t-t')}$$

and proceeding, as in Chapter IV, to let $V \longrightarrow \infty$, ignore overlapping terms, and to assume $\epsilon_{mz}t \gg 1$, we obtain,

$$\left.\begin{aligned}
&\sum_{z=1}^{m-1}\sum_{k\sigma} g_{k\sigma,mz}\, a_{k\sigma}(t) = \sum_{z=1}^{m-1}\Big\{ q_{mz}(\underline{r}) + i\left(\tfrac{1}{2}\gamma_{mz} - i\Omega_{mz_-}\right)P_{zm}(t) \\
&\qquad\qquad + i\left(i\Omega_{mz_+}\right)P^{+}_{zm}(t)\Big\} \\
&\qquad \text{where } \Omega_{mz_+} = \frac{\gamma_{mz}}{\epsilon^{3}_{mz}}\int_0^{\infty}\frac{d\omega}{2\pi}\,\frac{\omega^{3}}{\omega+\epsilon_{mz}} \text{ as in (2.A.15)}
\end{aligned}\right\}\quad\text{(IV.8)}$$

Similarly,

$$\left.\begin{aligned}
&\sum_{y=m+1}^{\ell}\sum_{k\sigma} g_{k\sigma,yn}\, a_{k\sigma}(t) = \sum_{y=m+1}^{\ell}\Big\{ q_{yn}(\underline{r}) + i\left(\tfrac{1}{2}\gamma_{yn} - i\Omega_{yn_-}\right)P_{ny}(t) \\
&\qquad\qquad + i\left(i\Omega_{yn_+}\right)P^{+}_{ny}(t)\Big\}
\end{aligned}\right\}\quad\text{(IV.9)}$$

etc.

Substituting in eq. for $\dot{P}_{mn}(t)$, we obtain:-

$$\left.\begin{aligned}
\dot{P}_{mn}(t) = i\epsilon_{mn}P_{mn} - i\Big\{ &\sum_{z=1}^{m-1}\underline{P_{zn}}\; q_{mz}(\underline{r}) \\
&- \sum_{y=m+1}^{\ell}\left(\underline{P_{my}}\; q_{yn}(\underline{r}) - \Omega_{yn_+}\underline{P_{mn}}\right) \\
&+ \sum_{y=m+1}^{\ell} P_{yn}\, q_{ym}(\underline{r}) - \Omega_{mm_+}P_{mm} \\
&- \sum_{z}^{n-1}\left(P_{mz}\, q_{nz}(\underline{r}) + i\left(\tfrac{1}{2}\gamma_{nz} - i\Omega_{nz_-}\right)P_{mn}\right)\Big\} \\
+ i\Big\{ &\sum_{y=m+1}^{\ell} q^{+}_{yn}(\underline{r})P_{my} \\
&- \sum_{z=1}^{m-1}\left(q^{+}_{mz}(\underline{r})P_{zn} - i\left(\tfrac{1}{2}\gamma_{mz} + i\Omega_{mz_-}\right)P_{mn}\right) \\
&+ \sum_{z=1}^{n-1} q^{+}_{nz}(\underline{r})\,\underline{P_{mz}} - i\left(\tfrac{1}{2}\gamma_{nn} + i\Omega_{nn_-}\right)\underline{P_{mn}} \\
&- \sum_{y=m+1}^{\ell}\left(q^{+}_{ym}(\underline{r})\underline{P_{yn}} + i\left(i\Omega_{ym_+}\right)P_{mn}\right)\Big\}
\end{aligned}\right\}\quad\text{(IV.10)}$$

Collecting terms, we obtain

$$
\left.
\begin{aligned}
\dot P_{mn}(t) = -i\epsilon_{nm} P_{mn} &- i\sum_{z=1}^{m-1}\left\{ \underline{P_{zn}}\, q_{mz}(r) + q^{+}_{mz}(r) P_{zn} - i(\tfrac12\gamma_{mz} + i\Omega_{mz-}) P_{mn} \right\} \\
&+ i\sum_{y=n+1}^{l}\left\{ \underline{P_{my}}\, q_{yn}(r) - i\Omega_{yn+} \underline{P_{mn}} + q^{+}_{yn}(r) P_{my} \right\} \\
&- i\sum_{y=n+1}^{l}\left\{ P_{yn}\, q_{ym}(r) - \Omega_{nm+} P_{mn} + \underline{\underline{q^{+}_{ym}(r) P_{yn}}} + i(i\Omega_{ym+}) \underline{P_{mn}} \right\} \\
&+ i\sum_{z=1}^{n-1}\left\{ P_{mz}\, q_{nz}(r) + i(\tfrac12\gamma_{nz} - i\Omega_{nz-}) P_{mn} + \underline{\underline{q^{+}_{nz}(r) P_{mz}}} - i(\tfrac12\gamma_{nm} - i\Omega_{nm-}) \underline{P_{mn}} \right\}
\end{aligned}
\right\} \quad \text{(IV.11)}
$$

where underlined terms result from H.F. terms in the Hamiltonian. This equation can also be written as:-

$$
\left.
\begin{aligned}
\dot P_{mn}(t) = &-\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\left\{ \tfrac12(\gamma_{ni} + \gamma_{mj}) + i\left(\epsilon_{nm} - \Omega_{ni-} + \Omega_{mj-} - \sum_{y=n+1}^{l} \underline{\Omega_{ym+}} + \sum_{y=n+1}^{l} \underline{\Omega_{yn+}}\right)\right\} P_{mn} \\
&+ \left(\underline{\tfrac12\gamma_{nm} - i\Omega_{nm-}} + \Omega_{nm+}\right) P_{mn} \\
&- i\sum_{r=m+1}^{l} P^{+}_{nr}\, q_{rm}(r) - i\sum_{r=n+1}^{l} \underline{q^{+}_{rn}(r) P_{rn}} + i\sum_{i=1}^{n-1} P^{+}_{im}\, q_{ni}(r) + i\sum_{i=1}^{n-1} \underline{q^{+}_{ni}(r) P_{mi}} \\
&- i\sum_{j=1}^{m-1} q^{+}_{mj}(r) P_{jn} \qquad\qquad + i\sum_{k=n+1}^{l} q^{+}_{kn}(r) P_{mk} \\
&- i\sum_{z=1}^{m-1} \underline{P_{zn}\, q_{mz}(r)} + i\sum_{y=n+1}^{l} \underline{P_{my}\, q_{yn}(r)}
\end{aligned}
\right\} \quad \text{(IV.12)}
$$

When we ignore terms originating from H.F. terms in the Hamiltonian we obtain

$$
\left.
\begin{aligned}
\dot P_{mn}(t) = &-\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\left\{ \tfrac12(\gamma_{ni} + \gamma_{mj}) + i(\epsilon_{nm} - \Omega_{ni-} + \Omega_{mj-})\right\} P_{mn} \\
&- i\sum_{r=n+1}^{l} \left(P^{+}_{nr}\, q_{rm}(r)\right)_{\substack{\text{H.F. if } r<n \\ \text{N.B. } r>m<n}} \qquad + i\sum_{i=1}^{n-1}\left(P^{+}_{im}\, q_{ni}(r)\right)_{\substack{\text{H.F. if } m<i \\ \text{N.B. } i<n>m}} \\
&- i\sum_{j=1}^{m-1}\left(q^{+}_{mj}(r) P_{jn}\right)_{\substack{j\ngtr n \\ \text{since } j<m<n \\ \text{i.e. } j<<n \\ \therefore\ \text{not H.F. term}}} \qquad + i\sum_{k=n+1}^{l}\left(q^{+}_{kn}(r) P_{mk}\right)_{\substack{m\ngtr k \\ \text{since } k>n>m \\ \text{i.e. } m<<k \\ \text{not H.F. term}}} \\
&+ i\sum_{y=n+1}^{l} \Omega_{nm+} P_{nm}
\end{aligned}
\right\} \quad \text{(IV.13)}
$$

though, in fact $P_{nm}(t) \sim e^{i\epsilon_{nm}t}$ is also a H.F. term.

In fact two of the terms, the terms which are doubly underlined, originating from H.F. terms in the Hamiltonian are in fact not necessarily H.F., viz.

$$\sum_{y=m+1}^{\ell} q^{+}_{ym}(\underline{r})\, P_{yn}$$ is only H.F. if $y > n$, where $n > m$, whereas y can be $= n$, $> n$ or $< n$. (IV.14)

$$\sum_{z=1}^{n-1} q^{+}_{nz}(\underline{r})\, P_{mz}$$ is only H.F. if $m > z$, where $m < n$, whereas m can be $= z$, $> z$ or $< z$. (IV.15)

so, in fact, we should use the following equation:-

$$\begin{aligned}\dot{P}_{mn}(t) = &-\sum_{i=1}^{n-1}\sum_{j=1}^{m-1}\left\{\tfrac{1}{2}(\gamma_{ni}+\gamma_{mj})+i(\epsilon_{nm}-\Omega_{ni\_}+\Omega_{mj+})\right\}P_{mn}\\ &-i\sum_{r=m+1}(P_{nr}\,q_{rm}(\underline{r}))-i\sum_{\substack{r=m+1\\ r<n}}^{\ell}(q^{+}_{rn}(\underline{r})P_{rn})+i\sum_{i=1}^{n-1}(P^{+}_{in}\,q_{in}(\underline{r}))\\ &\qquad\qquad+i\sum_{\substack{\ell=1\\ \ell<m}}^{n-1}(q^{+}_{n\ell}(\underline{r})\,P_{m\ell})\\ &-i\sum_{j=1}^{m-1}q^{+}_{mj}(\underline{r})\,P_{jn}\qquad\qquad+i\sum_{k=n+1}^{\ell}q^{+}_{kn}(\underline{r})\,P_{mk}\end{aligned} \quad \text{(IV.16)}$$

and from this

$$\begin{aligned}\dot{P}_{mm}(t) = &-\sum_{i=1}^{m-1}\left\{\gamma_{mi}-i\left(\underset{i<m}{P_{mi}}\,q_{mi}(\underline{r})-q^{+}_{mi}(\underline{r})P_{im}+\underset{i>m}{q^{+}_{mi}(\underline{r})P_{mi}}\right)\right\}\\ &+\sum_{j=m+1}^{\ell}\left\{\gamma_{jm}-i\left(\underset{j>m}{P_{jm}}\,q_{jm}(\underline{r})-q^{+}_{jm}(\underline{r})P_{mj}+\underset{j<m}{q^{+}_{jm}(\underline{r})P_{jm}}\right)\right\}\end{aligned} \quad \text{(IV.17)}$$

I.E. we have two extra terms in each equation and also two restrictions on the existing summations.

APPENDIX V

Some notes on Apanasevich's paper (34)

In Apanasevich's paper he considers a 3-level atomic system with closely spaced (or coincident) excited levels, and the affect on it on WEAK radiation. There are thus no non-linear effects and the susceptibility he calculates is the linear one. Since the field is weak it can be considered to possess a spread of frequencies such that levels 2 and 3 are both covered by it since they are very close. He thus not only includes $\lambda_{31} = \lambda_{13}$ but also $\lambda_{21} = \lambda_{12}$. Now in our case we wish to be able to consider the possibility of the field becoming very strong. Mathematically it would be impossible to consider any spread in the frequency of the incident field for this case. In our method (ii) we consider one mode of the incident field and in method (i), based on Mollow's analysis, the incident field has just ONE frequency with no spread. Stroud(98) also considers just ONE mode to be strongly excited and no more. But even this single frequency intense incident field is not quantum mechanically manageable and that is why Mollow considers the incident single frequency field to be classical. In Stroud's method he imposes the restriction that in the process under consideration only one photon has been consumed from this single frequency field and so on. (This is also assumed by Morozov in his paper on Stark splitting(122)). In this way Stroud is able to treat the single frequency strong incident field quantum mechanically. Physically, it is reasonable to consider just one frequency in the intense incident field since all intense coherent fields obtained from lasers have a very small spread. Thus when we are dealing with such a single frequency intense incident field it is obviously not possible for it to couple levels 1 and 2 at the same time as levels 1 and 3. In fact if we have a physical situation where the spread of the incident field is smaller than the distance between the central frequencies of the close-lying levels 2 and 3 then we can approximate the incident field with a single frequency field which couples only levels 1 and 3 and not 1 and 2, i.e. $\lambda_{12} = \lambda_{21} = 0$ in this case.

It is interesting to see what the <u>exact</u> equations corresponding to equations (2a) and (b) of Apanasevich would be. In fact, they are

$$\dot{\rho}_{21} = -\{\tfrac{1}{2}\gamma_{21} + i(\omega - \Omega_{21})\}\rho_{21} - \underline{i\lambda_{31}\mathcal{E}_D(t)\rho_{23}} + \underline{i\lambda_{21}\mathcal{E}_D(t)(\rho_{11}-\rho_{22})} - \{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\}\rho_{31} \tag{V.1}$$

$$\dot{\rho}_{31} = -\{\tfrac{1}{2}\gamma_{31} + i(\omega - \Omega_{31})\}\rho_{31} - \underline{i\lambda_{21}\mathcal{E}_D(t)\rho_{32}} + i\lambda_{31}\mathcal{E}_D(t)(\rho_{11}-\rho_{33}) - \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}\rho_{21} \tag{V.2}$$

in our notation, where the following transformations are necessary for conversion to Apanasevich's notation:-

$$\left.\begin{aligned}
&\tfrac{1}{2}\gamma_{21} \longrightarrow \gamma_{1}, \quad \omega - \Omega_{21} \longrightarrow \omega_{21}, \quad \tfrac{1}{2}\Gamma_{31} - i\Omega'_{31} \longrightarrow \beta_{21} \\
&\tfrac{1}{2}\gamma_{31} \longrightarrow \gamma_{31}, \quad \omega - \Omega_{31} \longrightarrow \omega_{31}, \quad \tfrac{1}{2}\Gamma_{21} - i\Omega'_{21} \longrightarrow \beta_{31} \\
&\lambda_{21}\mathcal{E}_D(t) = \frac{\underline{p}_{21}\cdot\hat{e}_{0D}}{\hbar}\mathcal{E}_{0D}e^{-i\omega_D t} \longrightarrow \frac{d_{21}}{\hbar}E\,e^{-i\omega t} \qquad \text{i.e. } \underline{p}_{21}\cdot\hat{e}_{0D} \to d_{21},\ \mathcal{E}_{0D} \to E,\ \omega_D \to \omega \\
&\lambda_{31}\mathcal{E}_D(t) = \frac{\underline{p}_{31}\cdot\hat{e}_{0D}}{\hbar}\mathcal{E}_{0D}e^{-i\omega_D t} \longrightarrow \frac{d_{31}}{\hbar}E\,e^{-i\omega t} \qquad \text{i.e. } \underline{p}_{31}\cdot\hat{e}_{0D} \to d_{31},\ \mathcal{E}_{0D} \to E,\ \omega_D \to \omega
\end{aligned}\right\} \tag{V.3}$$

where ω_{21} and ω_{31} include frequency shifts.

Thus we see that the two straight underlined terms have been neglected in the linear approximation. If these two terms were included it would then be necessary to solve nine equations in all where the remaining seven equations are:-

$$\left.\begin{aligned}
\dot{\rho}_{11} = {} & \{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) - i(\Omega'_{31}-\Omega'_{21})\}\rho_{32} + \{\tfrac{1}{2}(\Gamma_{31}+\Gamma_{21}) + i(\Omega'_{31}-\Omega'_{21})\}\rho_{23} + \gamma_{21}\rho_{22} + \gamma_{31}\rho_{33} \\
& - i\lambda_{21}\mathcal{E}_D(t)\rho_{12} + i\lambda_{12}\mathcal{E}_D^*(t)\rho_{21} - i\lambda_{31}\mathcal{E}_D(t)\rho_{13} + i\lambda_{13}\mathcal{E}_D^*(t)\rho_{31}
\end{aligned}\right\} \tag{V.4}$$

$$\dot{\rho}_{22} = -\{\tfrac{1}{2}\Gamma_{31} - i\Omega'_{31}\}\rho_{32} - \{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\}\rho_{23} + i\lambda_{21}\mathcal{E}_D(t)\rho_{12} - i\lambda_{12}\mathcal{E}_D^*(t)\rho_{21} \tag{V.5}$$

$$\left.\begin{aligned}
\dot{\rho}_{33} = {} & -\{\tfrac{1}{2}\Gamma_{21} + i\Omega'_{21}\}\rho_{32} - \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}\rho_{23} - \gamma_{31}\rho_{33} \\
& + i\lambda_{31}\mathcal{E}_D(t)\rho_{13} - i\lambda_{13}\mathcal{E}_D^*(t)\rho_{31}
\end{aligned}\right\} \tag{V.6}$$

$$\dot{\rho}_{12} = \dot{\rho}_{21}^{+} \tag{V.7}$$

$$\dot{\rho}_{13} = \dot{\rho}_{31}^{+} \tag{V.8}$$

$$\left.\begin{aligned}\dot{\rho}_{32} = -\{\tfrac{1}{2}(\gamma_{21}+\gamma_{31}) + i(\omega_3 - \Omega_{31} + \Omega_{21})\}\rho_{32} - \{\tfrac{1}{2}\Gamma_{31} + i\Omega'_{31}\}\rho_{33} - \{\tfrac{1}{2}\Gamma_{21} - i\Omega'_{21}\}\rho_{22} \\ + i\lambda_{31}\mathcal{E}_D(t)\rho_{12} - \underline{i\lambda_{12}\mathcal{E}_D^*(t)\rho_{31}}\end{aligned}\right\} \quad \text{(V.9)}$$

$$\dot{\rho}_{23} = \dot{\rho}_{32}^{+} \quad \text{(V.10)}$$

where the wavy underlined terms are those extra ones due to the fact that a weak field couples levels 2 and 1 also. This extra coupling modifies the Hamiltonian by addition of the term:-

$$\left.\begin{aligned} & H_{ID_2}(t) = -\hbar\{P_{21}(t)\lambda_{21}\mathcal{E}_D(t) + \mathcal{E}_D^*(t)\lambda_{12}P_{12}(t)\} \\ & \quad \text{where } \lambda_{21} = \frac{P_{21}\hat{e}_D}{\hbar} \\ & \quad \text{and } \mathcal{E}_D(t) = [illegible] \\ & \qquad \mathcal{E}_D^*(t) = [illegible] \end{aligned}\right\} \quad \text{(V.11)}$$

and spread of $\omega_D \sim$ of the distance between the central frequencies of the close-lying levels 2 and 3.

In Apanasevich's paper

$$\left.\begin{aligned} & \beta_{21} = \gamma_0^* + i\Delta^* \text{ and } \beta_{31} = \gamma_0 + i\Delta \text{ where } \gamma_0 \text{ is complex} \\ & \text{as opposed to } \tfrac{1}{2}\Gamma_{31} - i\Omega'_{31} \text{ and } \tfrac{1}{2}\Gamma_{21} - i\Omega'_{21} \text{ where } \Gamma_{31} \text{ and } \Gamma_{21} \text{ are} \\ & \text{real in our case.}\end{aligned}\right\} \quad \text{(V.12)}$$

Apanasevich considers the two extreme cases: (i) when levels 2 and 3 are far apart and (ii) when they are near. He finds that in case (i) the effect of relaxation coupling on the position and width of different components becomes weak and they are determined by the parameters of the different transitions. In case (ii) on the other hand, the relaxation coupling is found to change the contours of different components and to displace them. It also leads to a significant redistribution of intensities.

The effects of the weak field in Chapter VII (i.e. A = .1, .01) cannot really be compared with the effects found by Apanasevich because as already pointed out, we have not considered the field to have a spread such that it can couple levels 2 and 1, as well as levels 3 and 1. Also we used the same equations of motion to describe the situation when A is large or small, i.e. we did not assume the equations of motion to be linearly approximated as in Apanasevich's equations (2), when A was small, but included the straight-underlined terms of (V.1) and (II.2) for A small also. I.E. we have used exact equations of motion, in this respect, throughout.

The effects of strong radiation have been considered by Apanasevich et al.[123,124] elsewhere and they underline the well-known fact that strong radiation can lead to establishment of a definite coupling between different levels.

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ON MODULATED DECAY IN A TWO-LEVEL QUANTUM SYSTEM

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The radiative decay in a two level system with the excited state coupled by an external perturbation is investigated. The results obtained differ from those of Keller and Robiscoe who claim to have discoverd a new type of modulation factor.

Keller and Robiscoe [1] have in a recent paper carried out Weisskopf-Wigner type calculations for a system of two quantum levels which are simultaneously coupled by a quantized radiation interaction, describing the radiative decay, and by a classical external perturbation. They claim to have discovered a new type of modulation factor. Further, on the basis of their calculations, they speculate that this type of modulation will be found as well for a three level quantum system* in which the same external perturbation which couples the upper two levels also couples the lower two levels. Their theory has been worked out for a time scale very much larger than the atomic life time.

* A detailed analysis of the radiative decay of an atom with two excited states coupled by an external perturbation has recently appeared [2]. The coupling of the lower two levels with the external perturbation is not considered here.

Here we treat essentially the same problem as Keller and Robiscoe by using the transition operator technique as described by Lehmberg [3]. But we obtain solutions for times very much larger than the inverse of the atomic resonance frequency; these times may or may not be very much larger than the atomic life time. The more complete solutions lead to entirely different conclusions about the effect of the external perturbation on the radiative decay of the system. Besides, the method followed enables us to eveluate the expressions for the state populations in a very simple and direct way. Unlike the derivations made by Keller and Robiscoe, classical external perturbation need not necessarily be small compared to the atomic level separation.

We treat a two-level atom coupled to a bath of oscillators. These are assumed to be closely spaced in frequency such that their frequencies ω_k overlap the atomic resonance frequency ω_0.

The atom is coupled also to a time independent classical external perturbation. Their Hamiltonian may be written as

$$H = \tfrac{1}{2}\hbar\omega_0 A^+A + \tfrac{1}{2}\hbar\sum_k \omega_k a_k^+ a_k + \hbar A^+ \sum_k g_k a_k \exp(\mathrm{i}\theta_k)$$

$$+ \hbar\lambda A^+ \exp(-\mathrm{i}\phi) + \text{h.c.} \quad , \tag{1}$$

where g_k and λ are c-number coupling parameters, θ_k and ϕ denote arbitrary phases, and A^+, A are the atomic and a_k^+, a_k are the radiation (raising and lowering) operators.

Deriving Heisenberg equations of motion for $A(t)$, $Q(t) = A^+(t)A(t)$ and $a_k(t)$ using eq. (1) and the relations $[A,A^+]_+ = 1$, $[a_k, a_m^+] = \delta_{km}$, we follow a procedure identical to that which led from equations (2.6 - 2.11) to equation (2.19) in ref. [3], and get

$$\dot{A}(t) = -(\mathrm{i}\omega + \tfrac{1}{2}\gamma)A(t) + [2Q(t) - 1]\, B(t), \tag{2a}$$

$$\dot{Q}(t) = -\tfrac{1}{2}\gamma Q(t) - A^+(t)B(t) + \text{h.c.} \quad , \tag{2b}$$

where $B(t) = \mathrm{i}\sum_k g_k a_k(0)\exp[\mathrm{i}(\theta_k - \omega_k t)] + \mathrm{i}\lambda\exp(-\mathrm{i}\phi)$, $\omega = \omega_0 - \Omega$, Ω and γ denote the familiar frequency shift and the decay constant, respectively. We may note that these results are valid for $t \gg \omega_0^{-1}$.

Let us now write $P(t) = \mathrm{Tr}[\rho(t)Q]$, and $\sigma(t) = \mathrm{Tr}[\rho(t)A]$, where ρ is the full density operator for the joint system of bath oscillators and atom. Here, $P(t)$ is the probability of finding the atom in its excited state at time t. It is easily seen that for no radiation present initially, eqs. (2) reduce to coupled linear differential equations for $P(t)$, $\sigma(t)$ and $\sigma^*(t)$. These equations can be solved exactly. Their solutions for $P(0) = 1$, $\sigma(0) = 0$ yield

$$P(t) = \mu + (1 - 3\mu)\exp[-\gamma t(1 - 2\mu)]$$

$$+ 2\mu\cos(\omega t + \beta)\exp[-\tfrac{1}{2}\gamma t(1 + 2\mu)] \, , \tag{3}$$

where $\mu = |\lambda|^2/\omega^2$ and $\beta = 2\gamma/\omega$. Note that eq. (3) does not involve arbitrary phases θ_k and ϕ. In writing the expression for $P(t)$ given above μ and β have been treated as small compared to unity and their powers higher than the first have been neglected.

We may check that eq. (3) gives for $\lambda \to 0$ the exponential decay solution and for $\gamma \to 0$ the quantum oscillation solution as it should [1].

The expression (3) differs from eq. (43) of ref. [1] in describing two important features of the decaying process: (i) For $\gamma t \ggg 1$, eq. (3) gives a steady state solution $P(t) = \mu$ which is in contrast to the quantum oscillation solution described in fig. 3 of ref. [1]. (ii) The third term in eq. (3) decays at nearly half the rate for the second term. This is due to the mixing of the diagonal (P) and the off-diagonal(σ) matrix elements caused by coupling with the external perturbation. It is easily seen that $\sigma(t)$ decays at half the rate for $P(t)$ when there is no external perturbation present. Similar terms do not occur in eq. (43) [1].

Finally, it is worth remarking that the characteristic features of the decaying process described by eq. (3) and noted in (i) and (ii) are similar to those derived for the three level problem by Fontana and Lynch *.

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* eg. cf. the expression $(1 - P(t))$ with eq. 24) of ref. [2].

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Erratum
In eq. (3) ω should be replaced by (1+2μ)ω

ON RADIATIVE DECAY OF AN ATOM WITH TWO CLOSE-LYING EXCITED STATES

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The line shape of spontaneous emission of an atom with closely spaced energy levels is determined using transition operator techniques.

Mollow and Miller [1] have shown in detail how the effect of spontaneous emission of an atom can be described by considering the coupling of the atom to a "bath" of harmonic oscillators representing the modes of the electromagnetic field *. They discuss the case of a two-level atom. Their method of solution is based on a Markoff approximation. Assuming the coupling between the atom and the bath to be sufficiently weak they consider its effect up to second order in perturbation theory. Lehmberg [3] has shown that these approximations are unnecessary for obtaining the required equations of motion. His derivations use an approximation which may be taken to mean that no appreciable secular change occurs in the atomic states during times of the order of an atomic period. The equations of motion for transition operators derived by him pass in to the usual ones for reduced density matrix operators when initial states are specified. In this paper we use Lehmberg's method to calculate the radiation from overlapping energy levels by considering the simplest case of spontaneous emission from a three level atom.

We take an atom (lying at the origin of coordinates) which has two excited states $|j''\rangle$ and $|j'\rangle$ coupled to a ground state $|i\rangle$ by a quantized multimode electromagnetic field. We can write the Hamiltonian, in the dipole approximation, when direct transitions between $|j''\rangle$ and $|j'\rangle$ are neglected, as [3]

$$H = \hbar \sum_{\alpha=i,j} \epsilon_\alpha P_{\alpha,\alpha} + \hbar\sum_q \omega_q a_q^\dagger a_q - \hbar \sum_{q,j} g_{jq}(P_{j,i} a_q + a_q^\dagger P_{i,j}) \tag{1}$$

where $P_{\alpha,\beta} = |\alpha\rangle\langle\beta|$ is the atomic transition operator $(P^\dagger_{\alpha,\beta} = P_{\beta,\alpha})$, and a_q and $a_q^\dagger$ are the usual photon (annihilation and creation) operators. We shall denote the energy separation between states $|j''\rangle$ and $|i\rangle$, $|j'\rangle$ and $|i\rangle$, $|j''\rangle$ and $|j'\rangle$ by $\epsilon_{j''} - \epsilon_i \equiv \omega_{j''}$, $\epsilon_{j'} - \epsilon_i \equiv \omega_{j'}$, and $\epsilon_{j''} - \epsilon_{j'} \equiv \omega' = \omega_{j''} - \omega_{j'}$, respectively. In writing (1) we have omitted high frequency terms like $P_{i,j} a_q$ and $a_q^\dagger P_{j,i}$. Inclusion of such terms results mainly in modifying the frequency shifts in which we are not interested here. The coupling constants g_{jq} are given by $g_{jq} = K_q(\boldsymbol{e}_q \cdot \boldsymbol{p}_j)$, $K_q = (2\pi\omega_q/\hbar V)^{1/2}$, where $\boldsymbol{p}_j = \langle j|e\boldsymbol{x}|i\rangle$ are the dipole matrix elements, $\boldsymbol{e}_q$ is the unit polarization vector and V is the normalization volume.

The Hamiltonian (1) gives the following Heisenberg equations of motion

$$\dot{a}_q = -\mathrm{i}\,\omega_q a_q + \mathrm{i}\sum_j g_{jq} P_{i,j}, \tag{2a}$$

$$\dot{P}_{i,j''} = -\mathrm{i}\omega_{j''} P_{i,j''} - \mathrm{i}\, P_{j',j''}\sum_q g_{j'q} a_q - \mathrm{i}(P_{j'',j''} - P_{i,i})\sum_q g_{j''q} a_q \,. \tag{3a}$$

Equation (2a) has the formal solution

$$a_q(t) = \exp(-\mathrm{i}\,\omega_q t)\, a_q(0) + \mathrm{i}\sum_j g_{jq} \int_0^t \mathrm{d}t' \exp\{-\mathrm{i}\omega_q(t-t')\} P_{i,j}(t') \,. \tag{2b}$$

* Radiation damping model described in ref. [1] has also been used by Mollow [2] to obtain power spectrum of light scattered by a two-level atom driven near resonance by a monochromatic classical electric field.

Using eq. (2b) in (3a) and following the arguments put forward by Lehmberg for carrying out the summations and integrations, we obtain after neglecting frequency shifts:

$$P_{i,j''}(t) = -\sigma_{j''} P_{i,j''}(t) - \tfrac{1}{2}\Gamma_{j'} P_{i,j'}(t) - \mathrm{i}\sum_q [P_{j',j''}(t) g_{j'q} + \{P_{j'',j''}(t) - P_{i,i}(t)\} g_{j''q}] a_q(0) \exp(-\mathrm{i}\omega_q t), \quad (3b)$$

where $\sigma_j = \frac{1}{2}\gamma_j + \mathrm{i}\omega_j$, $\gamma_j = (4p_j^2 \omega_j^3/3\hbar C^3)$ and $\Gamma_j = (4p_{j'} \, p_{j''}\omega_j^3/3\hbar C^3)$.

Equations of motion for $P_{i,j'}(t)$ corresponding to those given by eq. (3) can be obtained by interchanging the indices j'' and j'. It may be emphasized that in deriving Γ_j we have integrated over those frequencies which are common to photon transitions between levels j'' to i and i to j' or vice versa. If levels j'' and j' are far apart these cross-terms, and hence Γ_j, can be neglected.

If equations of motion for $P_{i,j}$ (eq. (3b)) are multiplied on the right by the vacuum state $|0\rangle$ for all q photons they reduce to two coupled linear differential equations. Their solution for the case when the atom is initially in the state $|j''\rangle$ (with no radiation present) is given by

$$P_{i,j''}|0,j''\rangle = (1/2m)[(m+\sigma_{j''}-\sigma_{j'})\exp(-s_+t) + (m-\sigma_{j''}+\sigma_{j'})\exp(-s_-t)]|0,i\rangle, \quad (4a)$$

$$P_{i,j'}|0,j''\rangle = (\Gamma_{j''}/2m)(\exp(-s_+t) - \exp(-s_-t)|0,i\rangle \quad (4b)$$

where $s_\pm = \frac{1}{2}(\sigma_{j''}+\sigma_{j'} \pm m)$, $m = [(\sigma_{j''}-\sigma_{j'})^2 + \Gamma_{j''}\Gamma_{j'}]^{1/2}$.

We can now evaluate $b_q \equiv a_q(t \gg \gamma^{-1})|0,j''\rangle$ at the place of observation by using eq. (4) in (2b). If we put $g_{j''q} = g_{j'q} = g_q$, $\gamma_j = \Gamma_j = \gamma$, the line shape of spontaneous emission of the atom initially in state $|j''\rangle$ is given by

$$|b_q|^2 = \frac{4|g_q|^2(\omega_{j'}-\omega_q)^2}{4(\omega_{j''}-\omega_q)^2(\omega_{j'}-\omega_q)^2 + \gamma^2(\omega_{j''}+\omega_{j'}-2\omega_q)^2} \quad (5)$$

We see that eq. (5) is the same as that derived by Morozov and Shorygin [4] using the Heitler-Ma method. The proximity of level j' to that of j'' causes the line shape of the atom which is initially in level j'' to change over from the Lorentzian to that given above. It may also be noted that for the simple case considered here the spectral profile of the atomic decay, which is essentially the Fourier-transform of a two-time atomic correlation function, has been obtained without the use of the fluctuation-regression theorem. Besides, the Markoff approximation has not been used and the calculations are not limited to any specific order in coupling constants. The method of evaluation for the effect of overlapping given here should prove useful in describing situations where the calculations based on perturbation theory become invalid, for example, in the presence of a very intense external radiation field.

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