

## Far-infrared spectroscopy of single quantum dots in high magnetic fields

O. Astafiev and V. Antonov\*

CREST, Japan Science and Technology Corporation (JST) Kawaguchi-shi, Saitama 332-0012, Japan

T. Kutsuwa and S. Komiyama

Department of Basic Science, University of Tokyo, Komaba 3-8-1, Meguro-ku, Tokyo 153-8902, Japan

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Excitation spectra of single GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As quantum dots (QD's) are studied by using a technique of far-infrared single-photon detection. The spectra consist of a single resonance line, the resonance frequency of which is by several percent larger than the cyclotron-resonance frequency but is substantially smaller than the magnetoplasma frequencies of mesa-etched QD's earlier found through the standard far-infrared transmission spectroscopy. The resonance frequency is found to be in substantial agreement with the characteristic frequency of the parabolic bare confinement potential of the QD's, and is ascribed to the excitation of the upper branch of the Kohn-mode collective plasma oscillations.

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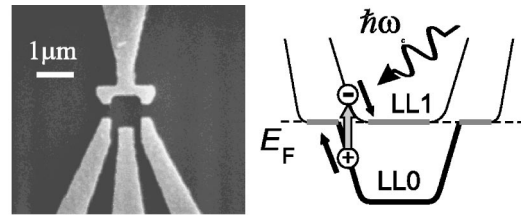
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Collective excitations of an electron system in semiconductor quantum dots (QD's) have been extensively studied through far-infrared (FIR) transmission experiments.<sup>1</sup> One restriction in those spectroscopic measurements is that the average spectra arising from a large number of QD's (arrays of QD's) have to be studied. This is because the radiation power absorbed by individual QD's is too small to be directly detected. It follows that some ambiguity might remain in physical parameters of the QD's and that fine spectral structures, if any, would be washed away after averaging. In general, therefore, it is desirable to carry out experiments on a single QD, for which physical parameters can be unambiguously determined. In this work, we demonstrate that the study of collective excitation is made in single QD's by exploiting a technique of single-FIR-photon detection.<sup>2,3</sup>

The number of FIR photons absorbed by a single QD placed in high magnetic fields can be counted as telegraph-like switches of tunnel conductance through the QD.<sup>2-5</sup> When one photon is absorbed via plasma-shifted cyclotron resonance (CR) in the QD, where the filling factor of Landau levels,  $\nu$ , lies in a range from 2 to 3, the excited electron (hole) in the higher (lower) Landau level rapidly moves to the inner-core (outer-ring) region of the QD as shown on the left panel of Fig. 1(a), where the outer ring and the inner core are compressible regions formed, respectively, by the lowest-orbital Landau level (LL1) and the first-excited orbital Landau level (LL2) (we ignore spin splitting). This induces an electrostatic polarization of the QD, which causes the tunnel conductance through the QD to switch on (off), when the QD is operated as a single-electron transistor<sup>6</sup> (SET).

The samples, similar to those used earlier,<sup>2,3</sup> are fabricated in a GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As single heterostructure crystal with a heterointerface-to-surface distance of 115 nm and a spacer layer thickness of 20 nm. A two-dimensional electron gas (2DEG) layer of a thickness about 10 nm is formed on the deeper side at the heterointerface. The electron mobility and density at 4.2 K are  $\mu = 85 \text{ m}^2/\text{Vs}$  and  $n_s = 2.6 \times 10^{15} \text{ m}^{-2}$ , respectively. The right panel of Fig. 1(a) is an image of scanning-electron microscope of the sample. When metal gates are negatively biased, the 2DEG layer is depleted

from the regions below the gates leaving a 2DEG island (QD) of 500 nm diameter (700 nm diameter in lithographic size) with about 300 electrons. To couple the QD to a long wavelength FIR radiation ( $\lambda \approx 0.17 - 0.22 \text{ mm}$ ), the metal gates extend to form a planar dipole antenna of a length of about 0.1 mm. The sample is placed in a mixing chamber of a dilution refrigerator at a base temperature of 50 mK, where the mixing chamber is located at the center of a superconducting solenoid.



(a)

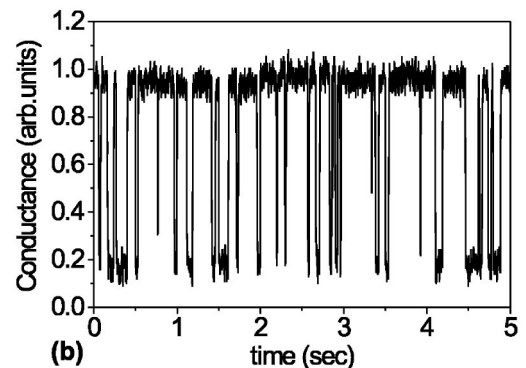


FIG. 1. (a) Left panel: Scanning-electron micrograph of the QD sample studied. Bright areas are metal gates on top of the GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As wafer. Right panel: Schematic representation of the electron excitation in the QD. (b) A typical time trace of the conductance in the regime of the single photon detection at a magnetic field of 3.85 T. Since the SET conductance is adjusted at a maximum in the dark, individual events of FIR photon absorption cause negative spikes in the conductance.

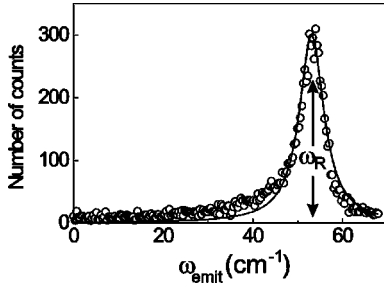


FIG. 2. An example of the excitation spectrum. The count of FIR photon absorption is displayed against the frequency  $\omega_{emit}$  of incident radiation calculated from  $B_{emit}$  applied to the emitter. The solid line shows a Lorentzian curve.

The gate bias voltages are so adjusted that the tunnel current through the QD (from the right- to the left-hand side reservoirs) exhibits Coulomb conductance oscillations. The charging energy in the SET regime is determined to be  $E_{ch} \approx 0.4$  meV. As a source of FIR radiation, we use a GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$  2DEG Hall bar in a magnetic field  $B_{emit}$ .<sup>8</sup> When a current (a few microamperes) is passed through the Hall bar, cyclotron resonance transition of non-equilibrium electrons yields cyclotron emission at a frequency,  $\omega_{emit} = eB_{emit}/m^*$  where  $m^* = 0.067m_0$  is the effective electron mass in GaAs. The radiation spectra of the Hall bar emitter, studied with a calibrated Hall-bar detector,<sup>8</sup> are well fitted to a Lorentzian curve with a full width at the half-maximum (FWHM) of about  $\Delta\omega_{emit} \approx 4.2$   $\text{cm}^{-1}$  at  $\omega_{emit} \approx 55$   $\text{cm}^{-1}$ .<sup>7</sup> The Hall bar is placed in an additional small superconducting solenoid (with a magnetic field  $B_{emit}$ ) at 80 cm distance from the QD sample, within the same cryostat as that of the QD sample. The emission, tuned by  $B_{emit}$ , is guided through a metal light pipe to the QD-sample. The FIR radiation power is roughly proportional to the electrical dissipated power. Unwanted mid- and/or near-infrared radiation is completely eliminated by filtering the radiation through silicon plates and black polyethylene films.

We choose a gate bias condition for the QD so that the SET conductance takes on maximum amplitude in the dark (the ground state). Photon-absorption events then give rise to telegraphlike conductance switches as shown in Fig. 1(b), where the time duration of switching off, or the dead time for counting, is given by the recombination lifetime  $\tau^*$  of the excited electron and the hole. We use weak power of incident FIR radiation, so that the average interval between successive events of photon absorption is much longer than  $\tau^*$ . In this condition, each event of photon absorption is counted without being hampered by dead time. Excitation spectra of the QD are studied if the conductance switches are counted as the radiation frequency  $\omega_{emit}$  or  $B_{emit}$  is scanned. (The electrical dissipated power of the emitter is kept unchanged during the sweep of  $B_{emit}$ .)

Figure 2 exemplifies one spectrum obtained when the magnetic field for the QD is fixed at  $B = 3.61$  T. The spectrum was taken by counting the conductance switches for 10 s at each point of  $\omega_{emit}$  ( $B_{emit}$ ), and by increasing  $\omega_{emit}$  with a step of  $\Delta\omega_{emit} \approx 0.33$   $\text{cm}^{-1}$ . The curve of the counts versus  $\omega_{emit}$  is featured by a prominent resonance peak with-

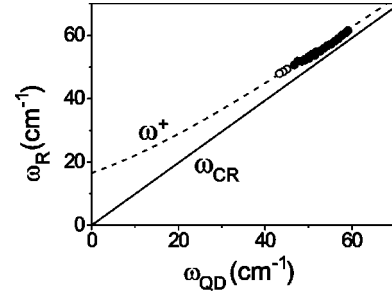


FIG. 3. Resonance frequency  $\omega_R$  as a function of the magnetic field  $B$  applied to the QD, expressed by the CR frequency  $\omega_{QD}$ . The data points indicated by solid circles are taken from the photon-counting method while those represented by open circles are from a standard modulation technique. The dashed line shows the dispersion curve  $\omega^+$  given by Eq. (1) with  $\omega_0 = 16$   $\text{cm}^{-1}$ . The solid line shows the pure CR,  $\omega_R = \omega_{QD}$ .

out appreciable background, indicating that the SET is free from dark switching and assuring that the curve correctly corresponds to the excitation spectrum. We find that the count is 300 photons per 10 s at the peak position, which corresponds to the absorbed radiation power of only  $3 \times 10^{-20}$  W. It should be stressed that this power level is far below the detectable limit of any conventional methods. The spectrum is found to be well fitted to the Lorentzian curve,  $1/\{[2(\omega_{emit} - \omega_R)/\Delta\omega]^2 + 1\}$ , indicated by a solid line with the resonance frequency  $\omega_R = 53.1$   $\text{cm}^{-1}$  and the linewidth  $\Delta\omega = 6.6$   $\text{cm}^{-1}$  (FWHM). Noting the linewidth of incident radiation,  $\Delta\omega_{emit} \approx 4.2$   $\text{cm}^{-1}$ , and deconvoluting two Lorentzians, the true linewidth is suggested to be not larger than  $\Delta\omega \approx 2.4$   $\text{cm}^{-1}$ , which is much narrower than that found in the averaged transmission spectra of QD arrays. We note that the value of  $\omega_R$  is higher than the bare CR frequency,  $\omega_{CR} = eB/m^* = 50.2$   $\text{cm}^{-1}$ , by 5.8%.

We found that the spectrum is substantially independent of the condition of gate bias voltages when the number of electrons in the QD is in the range 280–310. However, the resonance frequency of the spectrum is found to shift with varying the magnetic field  $B$  for the QD as shown in Fig. 3, where the values of  $\omega_R$  are plotted against  $B$ . Closed dots indicate the data studied through the photon-counting method described above, while open circles show results obtained through a standard modulation technique. (Photon-counting method is not applicable in the range of lower magnetic fields since  $\tau^*$  becomes shorter than the present instrumental time constant of about 1 ms.)

Figure 3 shows that values of  $\omega_{CR}$  are slightly higher than the CR frequency,  $\omega_{CR} = eB/m^*$ . They are well fitted to a solid curve drawn according to the upper branch of the relations

$$\omega^\pm = \sqrt{(\omega_{CR}/2)^2 + \omega_0^2} \pm \omega_{CR}/2 \quad (1)$$

expected for magnetoplasma resonance,<sup>9,1</sup> where the resonant-plasma frequency is determined to be  $\omega_0 = 16$   $\text{cm}^{-1}$  in Fig. 3. It may be interesting to consider what the value  $\omega_0 = 16$   $\text{cm}^{-1}$  implies for our QD.

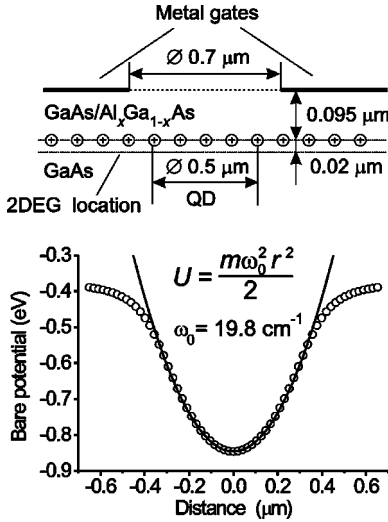


FIG. 4. Upper panel: Schematic representation of the heterostructure with metal gates on top of it. Lower panel: Open dots indicate calculated values of the bare potential induced by the positive background charge on the plane of the 2DEG. The horizontal scale is common for both panels.

In a case when a QD is defined by a cylindrical confining potential forming hard walls, a theory predicts<sup>10</sup>

$$\omega_0^2 = \frac{2\pi n_s e^2}{m^* \epsilon_{eff}} q_i, \quad (2)$$

where  $q_i$  is the  $i$ th wave vector, which can be assumed as  $q_i \approx \pi i/R$  for simplicity, and  $\epsilon_{eff} = \epsilon_{\text{GaAs}}/[1 + \exp(-2q_i h)]$  is an effective dielectric constant of the medium depending on the depth  $h$  of the 2DEG in the GaAs substrate. In existing experiments,<sup>11</sup> Eq. (2) is known to give a good approximation for relatively large QD's fabricated by mesa etching of GaAs/Al<sub>x</sub>Ga<sub>1-x</sub>As heterostructures. However, for our QD's of 0.7 μm diameter with  $h \approx 0.1 \mu\text{m}$  ( $\epsilon_{eff} \approx 9$ ), Eq. (2) yields the lowest plasma frequency ( $i=1$ ) of  $\omega_0 \approx 40 \text{ cm}^{-1}$ , which is by more than twice larger than the observed one. This strongly indicates that our metal-gate-shaped QD's are confined by a smooth potential.

To estimate the shape and the strength of a bare confining potential for our QD's, we consider a simplified structure depicted in the upper panel of Fig. 4, where a 2DEG layer is positioned at a depth,  $h_- = 115 \text{ nm}$ , below the surface of a GaAs substrate and neutralized by a remote layer of positive charges fixed to the wafer lattice at a depth,  $h_+ = 95 \text{ nm}$ , with a sheet density equal to the 2DEG density  $n_s$ . A metal gate is placed on top of the substrate leaving a 700-nm-diameter hole. (We neglect the surface charge because it is screened by positive charges induced through the band bending of GaAs.) A 2DEG island (QD) is formed by negative biasing the metal gate, resulting in electron depletion underneath it. We calculate a confinement potential formed by the bare positive background charge in presence of the grounded gate.

The potential can be found in the prolate spheroidal coordinates  $\xi, \eta, \theta$  related to the cylindrical coordinates  $r, z, \theta$

through a set of equations  $\xi = \sinh u$ ,  $\eta = \cos v$  and  $r = R \cosh u \sin v$ ,  $z = R \sinh u \cos v$ , where parameters  $u$  and  $v$  are defined as  $u > 0, 0 \leq v \leq \pi$  and the angle  $\theta (0 \leq \theta < 2\pi)$  is the same for both coordinates. A potential formed by a bounded conducting plane with a circular aperture situated in a uniform electric field  $E$  ( $E$  directed along  $z$  axis perpendicular to the plane in a half space  $z < 0$ ) is derived in an analytical form as  $\varphi = -Ez(\arctan 1/\xi - 1/\xi)/\pi$  at  $z > 0$  and  $\varphi = -Ez(\pi - \arctan 1/\xi + 1/\xi)/\pi$  at  $z < 0$ .<sup>12</sup> Here, the coordinates originate in a center of the aperture with  $z$  axis perpendicular to the interface. In our case, the electric field  $E = -4\pi n_s e / \epsilon_{\text{GaAs}}$  is induced by the positive charge sheet below the surface on the Fig. 4 and an effect of the GaAs-vacuum interface we will take into account as an additional electric field  $E' = 4\pi n'_s e / \epsilon_{\text{GaAs}}$  induced by a mirror charge with a sheet density  $n'_s = n_s(\epsilon - 1)/(\epsilon + 1)$  at a distance  $h_+$  above the GaAs surface. In the first order, the potential is parabolic and can be represented in terms of electron potential energy ( $U = e\varphi$ ) as  $U = [4/(1 + \beta^2)^2](e^2 n_s / \epsilon_{eff} R)(r^2/2) + U_0$ , where  $\beta = h_-/R$ ,  $\epsilon_{eff} = (\epsilon_{\text{GaAs}} + 1)/2$  and  $U_0$  is a constant potential energy, which can be easily derived but not affects the plasma oscillations. Remembering that a kinetic energy of the electron in the parabolic potential is  $U = m^* \omega_p^2 r^2/2$ , we find out square of the oscillation frequency as

$$\omega^2 = \frac{4}{(1 + \beta^2)^2} \frac{e^2 n_s}{\epsilon_{eff} m^* R}. \quad (3)$$

The lower panel of Fig. 4 demonstrates the exact bare confinement potential energy profile within our QD in the plane of 2DEG (open circles). This is very well approximated by a parabolic shape, with the characteristic frequency  $\omega_p \approx 19.8 \text{ cm}^{-1}$ , which is calculated from Eq. (3). In a pure parabolic potential, generalized Kohn theorem<sup>13,14</sup> predicts that a uniform alternating electric field excites only the center-of-mass motion at the resonance frequency  $\omega_p$ .

Thus, our estimate,  $\omega_p \approx 19.8 \text{ cm}^{-1}$ , reasonably accounts for the observed resonance frequency  $\omega_0 = 16 \text{ cm}^{-1}$ , and leads us to conclude that the confining potential of our QD's is well approximated by a parabolic one and that the observed resonance is due to a Kohn-mode plasma oscillation, in which electron-electron interaction does not affect the resonance frequency.<sup>15</sup> The discrepancy between the estimated and the observed values,  $\omega_p \approx 19.8 \text{ cm}^{-1}$ , and  $\omega_0 = 16 \text{ cm}^{-1}$ , might arise from our simplified model of metal gate structure.

Note that the collective center-of-mass motion of all the electrons, not a single-particle Landau level excitation, is excited in our QD. Nevertheless, it leads to a single-particle process [Fig. 1(a)]. Let us consider in which process this occurs. In the initial state of the excited collective center-of-mass motion, it is impossible to specify which particular electron is excited in the higher Landau level, because in quantum mechanics the excited state is a superposition of many different states in which different single electrons are excited. On the second stage, this many-body state will rapidly dephase through inelastic scattering processes into a mixed state in which one particular electron (hole) is excited

in the higher (lower) Landau level. In the experiments, the observed resonance linewidth,  $\Delta\omega < 2.4 \text{ cm}^{-1}$ , implies that the excited collective oscillation has a lifetime of  $\tau > 2/\Delta\omega \approx 4.5 \text{ ps}$ .<sup>16</sup> We suppose that this lifetime refers to the dephasing process discussed here. On the final stage, the excited electron (hole) releases its excess energy to the lattice through the electron-phonon interaction, whereby relaxing the slope of the confining potential. It is only this final stage that the electrical polarization develops in the QD, the process of which we suppose to be completed in the order of a few nanoseconds.

We confirmed in additional experiments that the lower branch excitation of Eq. (1),  $\omega^-$ , does not yield photoreponse. This is reasonable because it does not induce inter-Landau level transitions and consequently does not lead to the internal polarization of a QD.

In summary, FIR excitation spectra of electric-field-

induced single QD's at high magnetic fields have been successfully studied by applying a technique of single FIR photon counting. The spectra consist of a distinct single resonance line. The resonance frequency is slightly above the cyclotron-resonance frequency but is substantially smaller than the one found on mesa-etched QD's with similar dimensions. The resonance frequency is in substantial agreement with the estimated characteristic frequency of the parabolic bare confinement potential, and is identified as the upper branch of Kohn-mode plasma oscillation. The collective excitation of the Kohn mode leads to a single particle excitation that is relaxed to a state with an electric polarization of the QD.

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\*Present address: Department of Physics, Royal Holloway, University of London, Egham, Surrey TW20 0EX, England.

<sup>1</sup>D. Heitmann, and J.P. Kotthaus, *Phys. Today* **46** (6), 56 (1993).

<sup>2</sup>S. Komiyama, O. Astafiev, V. Antonov, T. Kutsuwa, and H. Hirai, *Nature (London)* **403**, 405 (2000).

<sup>3</sup>O. Astafiev, V. Antonov, T. Kutsuwa, and S. Komiyama, *Phys. Rev. B* **62**, 16 731 (2000).

<sup>4</sup>V. Antonov, O. Astafiev, T. Kutsuwa, H. Hirai, and S. Komiyama, *Physica E (Amsterdam)* **6**, 367 (2000).

<sup>5</sup>S. Komiyama, O. Astafiev, V. Antonov, H. Hirai, and T. Kutsuwa, *Physica E (Amsterdam)* **7**, 698 (2000).

<sup>6</sup>It has been found that the lifetime varies from 1 ms to 20 min when the last electron remains on the LL2.

<sup>7</sup>Hereafter, frequency values are presented in units of ( $\text{cm}^{-1}$ ), which corresponds to (Hz) in CGS system as  $\omega(\text{cm}^{-1}) = \omega(\text{Hz})/2\pi c$ .

<sup>8</sup>Y. Kawano, Y. Hisanaga, and S. Komiyama, *Phys. Rev. B* **59**, 12 537 (1999).

<sup>9</sup>V. Fock, *Z. Phys.* **47**, 446 (1928).

<sup>10</sup>Alexander L. Fetter, *Phys. Rev. B* **33**, 5221 (1986).

<sup>11</sup>See, for example, V. Gudmundsson, A. Brataas, P. Grambow, B. Meurer, T. Kurth, and D. Heitmann, *Phys. Rev. B* **51**, 17 744 (1995) and R. Krahne, M. Hochgrafe, Ch. Heyn, and D. Heitmann, *ibid.* **61**, 16 319 (2000).

<sup>12</sup>See, for example, L.D. Landau and E.M. Lifshitz, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon Press, Oxford, 1963), p. 29.

<sup>13</sup>Yu.A. Firsov and V.L. Gurevich, *Zh. Esp. Teor. Fiz.* **41**, 512 (1961) [*Sov. Phys. JETP* **14**, 367 (1962)].

<sup>14</sup>L. Brey, N.F. Johnson, and B.I. Halperin, *Phys. Rev. B* **40**, 10 647 (1986).

<sup>15</sup>R. Krahne, V. Gudmundsson, C. Heyn, and D. Heitmann, *Phys. Rev. B* **63**, 195303 (2001).

<sup>16</sup>We tentatively suppose that a possible dephasing scattering arises from time-dependent potential fluctuations, which may be caused either by charge fluctuations in the substrate or by electrical noise from metal gates.