Threshold Law For Positron Impact Ionization of Atoms

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We demonstrate that recent experiments for positron impact ionization of He and H_2 can be interpreted by extending Wannier theory to higher energies. Anharmonicities in the expansion of the three-particle potential around the Wannier configuration give rise to corrections in the threshold behavior of the breakup cross section. These corrections are taken into account perturbatively by employing the hidden crossing theory. The resulting threshold law is $\sigma(E) \propto E^{2.640} \exp[-0.73\sqrt{E}]$. The actual energy range for which the Wannier law is valid is found to be smaller for positron impact ionization than for electron impact ionization. [S0031-9007(97)03161-X]

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The three-body breakup of Coulomb systems near threshold is of fundamental importance in atomic physics. The dependence of the threshold cross section on the excess energy probes the highly correlated dynamics of the three-body Coulomb system [1]. Recently new detailed measurements of the cross section for ionization of neutral atoms and molecules by positron impact in the near threshold region have been reported [2]. These experiments offer an exciting new opportunity to enlarge our understanding of the dynamics of three-body Coulomb systems immediately above the breakup threshold.

Wannier's theory [3] of breakup has been used to explain the threshold behavior for electron impact ionization of atoms. He predicted a threshold law as a function of the excess energy E of the form

$$\sigma(E) \propto E^{\zeta}. \tag{1}$$

The Wannier exponent has the value $\zeta = 1.127$ for electron impact ionization. Klar [4] subsequently showed that Wannier's theory for positron impact ionization leads to a power law with an exponent $\zeta = 2.651$. The recent experimental data [2], however, were best fitted to a power law with an exponent which ranges from $\zeta = 1.71$ for a H_2 target to $\zeta = 2.27$ for a He target. In any case the power law exponent is significantly smaller than predicted by Wannier theory. We demonstrate in this Letter how these results can be reconciled by showing that it is essential to incorporate fully the coupling between different modes of the three-body motion to account for the effects manifest in the experimental data. The result will be a modified threshold law giving the usual Wannier law in the zero energy limit. We also find the energy range over which the Wannier power law is valid. (See also [5].)

The threshold behavior of the cross section for positron impact ionization is a much more sensitive test for three-particle correlations than electron impact ionization. For electron impact ionization, were the interaction between the electrons in the final channel "turned off," the threshold law would be a linear function of the energy as opposed to having the exponent $\zeta = 1.127$. The Wannier configuration for positron impact ionization is a three-particle arrangement where the electron is situated on a line between the two positive charges, but not at equal distances from them [4]. The turning off of the final state interaction between the electron and the positron results in a much more dramatic change of the form of the threshold law. It would have the form $\sigma(E) \propto \int_0^E \exp[-\pi(2/x)^{1/2}]\,dx$ [6]. This form can be understood as the positron tunneling in the repulsive Coulomb potential of the final channel from near the nucleus to larger distances. Because of the dynamical screening for motion around the Wannier configuration such a tunneling effect is missing in Wannier theory. An alternative picture was given by Temkin's dipole theory [7], which is, however, only valid in a very limited energy region above threshold and is not yet experimentally accessible. Recent calculations of time-dependent wave functions support the original Wannier picture [8–10] of a double escape wave function confined to the vicinity of the classical Wannier configuration on the ridge of the three-particle potential.

Our approach is based on an analysis of the motion around the Wannier ridge. We confine our analysis to total angular momentum $L = 0$ because the *functional* behavior of the double ionization cross section as a function of the energy is the same for all partial waves in the zero energy limit [11]. In the body-fixed plane defined by the nucleus, the positron, and the electron the full three-dimensional Hamiltonian can be written in atomic units as [4]

$$H = -\frac{1}{2R^5}\frac{\partial}{\partial R}R^5\frac{\partial}{\partial R} + \frac{1}{R^2}h(R,\beta,\gamma), \tag{2}$$

$$h = \frac{1}{2}\Lambda^2(\beta,\gamma) + RC(\beta,\gamma). \tag{3}$$

R is the usual hyperradius and the mock angles $0 \le \beta \le \pi/4$ and $0 \le \gamma \le 2\pi$ are related to the moments of

inertia of the system. Expressions for the grand angular momentum $\Lambda^2(\beta,\gamma)$ and the effective charge $C(\beta,\gamma)$ as a function of the angles are given in [4]. We will refer to $h(R,\beta,\gamma)$ as the adiabatic Hamiltonian for reasons which will become clear later on.

For the moment it is important to notice that the effective charge has a saddle point at [4]

$$\beta = 0, \qquad \gamma = \arccos\frac{3Z - \sqrt{Z(Z+4)}}{2Z-1}. \quad (4)$$

$\beta = 0$ corresponds to a collinear configuration of the three particles while the value of γ selects a configuration where the electron is situated between the positive particles. The equilibrium position is stable with respect to β but it is unstable with respect to γ. It defines the Wannier ridge.

Our strategy is to construct a local solution of the Schrödinger equation around the Wannier saddle which takes the anharmonicities of the effective charge into account. We introduce the new coordinates $x = (\gamma - \gamma_0)/2$ and $y = \beta$ and expand the adiabatic Hamiltonian in these coordinates which gives the four terms $h = h_0 + h_1 + h_2 - RC_0$ with

$$h_0 = -\frac{1}{2}\left[\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{1}{y}\frac{\partial}{\partial y}\right] - RC_{x2}x^2 + RC_{y2}y^2, \quad (5)$$

$$h_1 = RC_{x3}x^3 - RC_{xy2}xy^2, \quad (6)$$

$$h_2 = \frac{8y}{3}\frac{\partial}{\partial y} - 2y^2\frac{\partial^2}{\partial x^2} - RC_{x4}x^4 + RC_{x2y2}x^2y^2 - RC_{y4}y^4. \quad (7)$$

Numerical values of the expansion coefficients are recorded in Table I for the charge $Z = 1$ of the residual ion.

We now expand the three-particle wave function around the saddle into adiabatic channels:

$$\Psi(R;x,y) = \frac{1}{R^{5/2}}\sum_\mu F_\mu(R)\varphi_\mu(R;x,y). \quad (8)$$

Neglecting the nonadiabatic couplings results in the Schrödinger equation

$$[h_0 + h_1 + h_2]\varphi_\mu(R;x,y) = R^2\varepsilon_\mu(R)\varphi_\mu(R;x,y) \quad (9)$$

for the adiabatic channel functions. Had we taken only the first term h_0 into account we would have ended up with second order Wannier theory and a power law for the cross section: The motion in x and y decouples and the adiabatic channels are products of a quasidiscretized one-dimensional antiharmonic oscillator in x and a two-dimensional harmonic oscillator in y. The adiabatic energies associated with these wave functions are

$$E_{nm} = R^2\varepsilon^{(0)}_{nm}(R) = -i(n+1/2)\sqrt{2RC_{x2}} + (2m+1)\sqrt{2RC_{y2}}. \quad (10)$$

The wave functions for the antiharmonic oscillator were chosen with outgoing wave boundary conditions which correspond to the picture of particles falling off the saddle. The minus sign in front of the imaginary part of the adiabatic energy reflects this fact. The transformation from adiabatic to diabatic channels was performed explicitly in second order Wannier theory in an elegant way [12]. The diabatic theory leads to the correct value of the Wannier exponent ζ in Eq. (1).

At higher energies it is important to include anharmonic corrections in x and y in the adiabatic Hamiltonian as has been demonstrated for electron impact ionization [13]. In this case the construction of diabatic channel wave functions, which takes the coupling between the angular motion and the motion in the hyperradius into account, is possible although rather tedious. This direct way, however, is not practicable for positron impact ionization because of the occurrence of the cubic terms in the adiabatic Hamiltonian. A central task of this Letter is to demonstrate that the higher order corrections to the simple Wannier law (1) can be calculated also for positron impact ionization, but a different approach is needed.

The general framework involves the hidden crossing theory applied to ionization processes [14–18]. Higher order corrections to the Wannier law are then calculated within a perturbative approach. The central idea of the hidden crossing theory stems from the observation that the adiabatic energies $\varepsilon_\mu(R)$ typically show avoided crossings at real positive values of the hyperradius. Asymptotically they correspond to the excitation channels. Double ionization can be achieved via a path in the *complex R* plane where promotion into the double continuum on the single valued sheet of $\varepsilon(R)$ is possible. The transition probability for a path which starts at a positive real value R_0 on the potential curve of the initial state into the double continuum is given by

$$P_{\rm asy}(E) = \exp\left\{-2\,\mathrm{Im}\int_{R_0}^{\infty}\sqrt{2[E + C_0/R - \varepsilon(R)]}\,dR\right\}. \quad (11)$$

An analogous expression occurs in the diabatic theory where it is interpreted as the survival probability on the saddle as the two escaping particles travel from R_0 to infinite hyperradius [12].

The absolute value of the double escape cross section for total angular momentum zero is given by

$$\sigma(E) = \frac{\pi}{k_i^2}P_{\rm inner}(E)P_{\rm asy}(E) \times \int dx \int y\,dy|\varphi_{\rm asy}(R_W;x,y)|^2, \quad (12)$$

TABLE I. Expansion parameters of the adiabatic Hamiltonian $h(R,\beta,\gamma)$ in Eqs. (3) and (5)–(7) around the Wannier saddle.

C_0	3.3302	C_{x3}	18.206	C_{x4}	197.16
C_{x2}	27.821	C_{xy2}	27.309	C_{x2y2}	570.62
C_{y2}	11.413			C_{y4}	47.669

where k_i is the asymptotic momentum of the particle in the incoming channel. The factor $P_{\text{inner}}(E)$ contributes to transitions taking place within the reaction zone at $R < R_0$. Since at small interparticle separations the Coulomb potential dominates, this factor is only weakly dependent on the energy E and is not needed to determine the functional dependence of the threshold law as a function of the energy. The third factor stems from the integration of the asymptotic wave function in the angular coordinates x and y at the Wannier radius R_W. This radius characterizes the transition from the Coulomb zone to the asymptotic free zone and therefore scales as $R_W \sim 1/E$.

The analytic continuation of the solutions of the adiabatic Schrödinger equation to complex values of R in the hidden crossing theory requires that the dual of the wave function is defined as the wave function itself $\langle\varphi|\beta,\gamma\rangle = \langle\beta,\gamma|\varphi\rangle$ instead of taking its complex conjugate [17]. Expectation values of operators are understood to be taken with the modified scalar product. With this prescription at hand, corrections to the adiabatic energy (10) are readily calculated perturbatively. We first expand the transition probability (11) as

$$P_{\text{asy}}(E) \approx \exp\left\{2\,\text{Im}\int_{R_0}^{\infty}\left[\frac{\varepsilon(R)}{K_0(R)} + \frac{\varepsilon^2(R)}{2K_0^3(R)}\right]dR\right\}, \tag{13}$$

where the zero order momentum $K_0(R) = \sqrt{2(E + C_0/R)}$ has been introduced. The only imaginary contributions from first order perturbation theory arise from the cross terms involving both coordinates x and y in h_2 and give

$$\begin{aligned}\text{Im}\,\Delta\varepsilon^{(1)} &= \text{Im}\,\langle 00|h_2|00\rangle/R^2 \\ &= \frac{1}{R^2\sqrt{C_{y2}}}\left[\frac{C_{x2y2}}{4\sqrt{C_{x2}}} - \sqrt{C_{x2}}\right].\end{aligned} \tag{14}$$

The product states of the one-dimensional antiharmonic oscillator and the two-dimensional harmonic oscillator are denoted $|nm\rangle$. Contibutions from h_1 arise in second order perturbation theory, namely,

$$\text{Im}\,\Delta\varepsilon^{(2)} = \frac{1}{R^2}\,\text{Im}\sum_{n,m}\frac{|\langle 00|h_1|nm\rangle|^2}{E_{00} - E_{nm}}. \tag{15}$$

There are two contributions to the imaginary part. The first one comes from terms involving products of matrix elements of x^3 and xy^2. It gives

$$\text{Im}\,\Delta\varepsilon_a^{(2)} = -\frac{3C_{x3}C_{xy2}}{8R^2C_{x2}\sqrt{C_{x2}C_{y2}}}. \tag{16}$$

The second one involves the square of the matrix elements of xy^2. It gives

$$\text{Im}\,\Delta\varepsilon_b^{(2)} = \frac{C_{xy2}^2}{4R^2C_{x2}\sqrt{C_{x2}C_{y2}}}\left[1 + \frac{4C_{y2}}{C_{x2}}\right]^{-1}. \tag{17}$$

The adiabatic energy up to terms of order $1/R^2$ is then

$$\varepsilon(R) = \varepsilon_{0,0}^{(0)} + D/R^2, \qquad D = 6.2107. \tag{18}$$

We are now able to determine the higher order corrections to theWannier law (1) by inserting (18) into the expression (13) for the transition probability. The integrations can be performed analytically; the full result will be given elsewhere. If only the lowest order terms in E are retained the threshold behavior of the ionization cross section (12) is

$$\sigma(E) = \frac{\text{const}}{E + I}\,E^{\zeta_{\text{ad}}}\exp\left[-\frac{2D'}{C_0}\sqrt{2E}\right], \tag{19}$$

with $D' = D - \sqrt{C_{x2}C_{y2}}/C_0$ and $\zeta_{\text{ad}} = \sqrt{C_{x2}/C_0} - 1/4$. I is the ionization energy of the target. The numerical values are $D' = 0.86$, $2\sqrt{2}\,D'/C_0 = 0.730$, and $\zeta_{\text{ad}} = 2.640$. The adiabatic threshold exponent ζ_{ad} departs from the exact Wannier value by a relative error of only 0.4%. This is remarkable compared to the case of electron impact ionization where the relative error is 2%. The small difference between the adiabatic exponent and the Wannier exponent gives us confidence that the errors introduced by the adiabatic approximation are also small for the perturbative calculation of the exponential correction term. The threshold law can thus be written as

$$\sigma(E) \propto E^{2.640}\exp[-0.73\sqrt{E}\,]. \tag{20}$$

We do not consider terms linear in the energy in the argument of the exponential function because they also include terms in ER_0/C_0, which are analytic in E and would lead us to specify the boundary R_0 of the reaction zone. Moreover such terms also occur if higher orders than $1/R^2$ are included into the adiabatic energy. To be consistent it is therefore appropriate to compare experimental results with the first order nonanalytic corrections in $\sqrt{E}$ to the Wannier law.

We can now determine the range of validity of the power law using Eq. (20). At an excess energy of $E = 0.57$ eV the exponential factor in (20) already has the value 0.9. The ionization cross section measured in [2] was fitted to a power law in the excess energy range between 3 and 10 eV. The exponent thus obtained was $\zeta = 2.27$ for a He target. However, it is seen from the above that in this energy range the influence of the anharmonic corrections cannot be neglected. Figure 1 shows the experimental data together with the Wannier law, the power law fit to the experiment, and the threshold behavior (20). The latter was normalized to the power law fit at 4 eV. It is seen that for $E > 3$ eV the power law fit is almost indistinguishable from the modified threshold law (20). We are thus led to conclude that the experimental data imitate a power law behavior in the energy range under consideration where the actual functional dependence on E is the more complicated one given by (20).

It is interesting to compare the range of validity of the power law behavior with the case of electron impact ionization. An analysis analogous to the one outlined

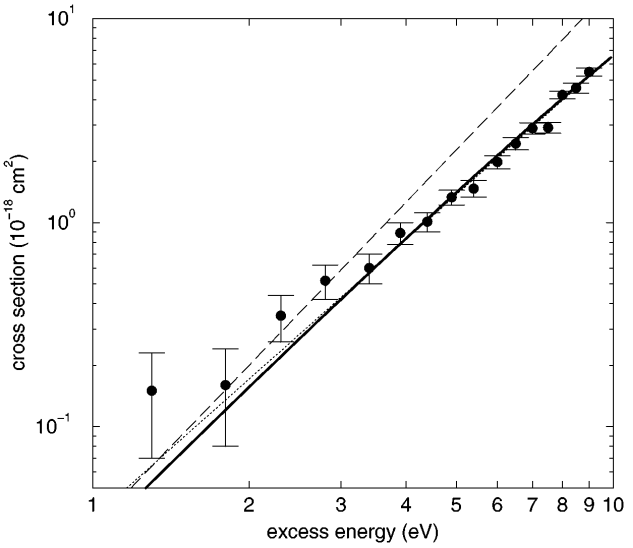

FIG. 1. Cross section for positron impact ionization of helium as a function of excess energy. Filled circles: experimental data Ref. [2], dashed line: Wannier power law (1) with $\zeta = 2.651$, solid line: Wannier theory including anharmonic corrections Eq. (20), dotted line: power law fit with exponent $\zeta = 2.27$ of Ref. [2].

here gives $C_0 = 3/\sqrt{2}$, $D = 7/(8\sqrt{11})$, and $D' = D - \sqrt{11}/6$. The resulting factor in the exponential term then has the value $2\sqrt{2}D'/C_0 = -0.385$. Had we used D instead of D' its value would be 0.352. This corresponds to the analysis in Ref. [13], which neglects the term quadratic in the asymptotic energy in (13). The diabatic value is $D = 0.329$ which also confirms the close agreement with the hidden crossing theory. In any case it is seen that the departure from the power law behavior comes into play at higher energies for electron impact ionization compared to the positron case.

We have shown that it is important to incorporate *both* the bending and stretching motions around the Wannier configuration. The presence of the cross terms in x and y proves to be essential to account for the anharmonic effects in the behavior of the cross section at energies close to threshold. In this respect we differ from the result of Ref. [11] which incorporates the full three-particle potential but restricts the configuration to a collinear one, thus fixing $y = 0$. They observed that the power law behavior holds up to at least 3 eV. The recent experimental data give strong support that at least above 3 eV anharmonic corrections due to the coupling of both degrees of freedom become quite pronounced for positron impact ionization.

Two further points should be noted: First, the modified threshold law (20) does not fit the experimental results at energies less than 3 eV. We checked that incorporating higher order terms depending on R_0 did not improve the situation. Because of the large experimental error bars and an energy spread of 0.5 eV of the incoming positron beam in the experiment [2] there is however a large uncertainty in the experimental data at lower energies. Clearly more experimental work which is able to probe our proposed modified threshold law at energies <3 eV would be highly desirable. Second, the fit of the experimental data to a power law is not completely independent of the target. The value 2.27 for helium changes to 1.71 for H_2 as the target. However for the latter it cannot be excluded that effects of the reaction zone arising from the molecular nature of the target play a genuine role. The general feature of an effective exponent which is less than the Wannier value however remains unchanged in accordance with our quantitative anharmonic theory.

In conclusion, we have shown that recent experimental results for positron impact ionization can be explained if anharmonic corrections to the Wannier law are taken into account. Measurement of the ionization cross section provides a very sensitive test for the Wannier theory due to the large threshold exponent compared to the electron impact case but—as experiment and theory both indicate—at the price of a less extended energy range in which the Wannier law is valid.

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