Threshold Detachment of Negative Ions by Electron Impact

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The description of threshold fragmentation under long range repulsive forces is presented. The dominant energy dependence near threshold is isolated by decomposing the cross section into a product of a background part and a barrier penetration probability resulting from the repulsive Coulomb interaction. This tunneling probability contains the dominant energy variation and it can be calculated analytically based on the same principles as Wannier's description [Phys. Rev. **90**, 817 (1953)] for threshold ionization under attractive forces. Good agreement is found with the available experimental cross sections on detachment by electron impact from D^-, O^-, and B^-. [S0031-9007(99)08480-X]

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Storage ring based experiments on threshold detachment from the deuteron (D^-) and the oxygen (O^-) negative ions by electron impact [1–3], and recently also from B^- [4], have stimulated the theoretical interest in the mechanism and the quantitative description of this process [5–9]. It is a fundamental question how threshold detachment proceeds since for very low energies the impacting electron does not even reach the atom because it is repelled by the loosely bound electron. Early theoretical work on this problem tried to describe the process by asymptotic properties of the wave function for the two electrons in the continuum after the collision [10], following the spirit of Wigner's treatment for two-body break up [11]. However, the predicted cross section agrees poorly with the experimental results.

Some recent theoretical treatments, following another idea of the early days [12], emphasize the importance of tunneling contributions, by either treating the impacting electron as a constant perturbing electric field [2] or merging a quantum and a classical description [5]. Astonishingly good agreement with the experiment, even at low energies near threshold, comes from a coupled channel calculation in the impact-parameter formalism where a classical trajectory is used for the relative motion of target and projectile electron and the electron to be detached is described quantum mechanically [7]. These results, at least the shape of the cross section, depend little on the polarization potential used, as Lin *et al.* emphasize [7]. Results of similar accuracy have been reported using a lowest order distorted-wave scheme, however, in contrast to [7], with a sensitive dependence on the polarization potential [8].

Without a full calculation of all electrons, one cannot avoid to use parameters in one or another way, either directly in the simpler models [1,2] or indirectly in the more involved calculations modeling polarization potentials for the loosely bound electron [5–9].

The theoretical work so far remains inconclusive concerning a dominant mechanism of threshold detachment, and the reason for the seemingly contradicting findings concerning the robustness of the results with respect to changes in the polarization potential is unknown.

A successful description of near threshold detachment focusing on threshold properties should naturally depend very little on details of the polarization since the long range repulsion between target and projectile electron dominates. Moreover, such an approach should uncover a mechanism for threshold detachment and thereby clarify the issue of robustness with respect to different polarization potentials.

In the following we will show that threshold detachment by electrons can be described with the same technique which has led to the successful (and purely classical) description of threshold ionization under long range *attractive* Coulomb forces, pioneered by Wannier [13].

However, in order to learn how to deal with repulsive Coulomb forces, one must go back to a semiclassical formulation of threshold ionization and analyze the reason why Wannier's classical treatment was appropriate. Semiclassically, one may write the scattering amplitude in the form [14]

$$f = \sum_j \sqrt{\mathcal{P}_j} \exp[i\Phi_j(E)/\hbar - i\nu_j\pi/2], \qquad (1)$$

where the sum runs over all scattering orbits j which contribute with the weight $\sqrt{\mathcal{P}_j}$. The phase contains the Maslov index ν_j [15] and the action Φ_j along the orbit which may be expressed as

$$\Phi_j(E) = \phi_j(E)E^{-1/2}, \qquad (2)$$

where $\phi_j(E \to 0) = \text{const}$ [14]. This special form is a consequence of the homogeneous Coulomb interaction. It is crucial for the justification of the classical treatment since $E \to 0$, i.e., approaching threshold, and $\hbar \to 0$ have the same effect in Eq. (1). If Φ_j is real, which is the case for all classically allowed trajectories, one arrives by stationary phase approximation (for $E \to 0$ or $\hbar \to 0$) at the result

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$$\sigma = \sum_j \mathcal{P}_j = \sigma_{\mathrm{CL}}\,, \tag{3}$$

which sums all individual contributions $\mathcal{P}_j$ of the trajectories to the classical cross section σ_{CL}.

Looking for the dominant energy dependence of $\sigma(E \to 0)$ we decompose the cross section into

$$\sigma(E) = \sigma_B(E)P(E)\,, \tag{4}$$

where $\sigma_B(E)$ is a smooth background cross section with $\sigma_B(E \to 0) = \text{const}$. Wannier showed that the dominant energy dependence $P(E) = P_*$ is contained in a single fixed point orbit $j = *$ [13]. Formally, this orbit represents an outgoing trajectory with fixed angle $\theta^* = \pi$ between the two electrons and symmetric distances $r_1 = r_2$ of electron 1 and 2 from the core. It is convenient to use hyperspherical coordinates with an overall radius $r^2 = r_1^2 + r_2^2$ of the system and the hyperangle defined by $\tan\alpha = r_1/r_2$. The orbit represents a classical fixed point because the classical equations of motion with the full Hamiltonian do not change (α, θ) in time, i.e., $\alpha(t) = \alpha^* = \pi/4$ and $\theta(t) = \theta^* = \pi$. The potential energy of the two electrons interacting with a core of charge Z can be written in the form of a Coulomb potential with an angular dependent charge, $V = C(\alpha, \theta)/r$. For $Z > 1/4$ the potential at the fixed point is with $C_* = C(\alpha^*, \theta^*) < 0$ attractive. Hence the relevant threshold orbit at $E \geq 0$ is classically allowed with a real action Φ_*. Then, as sketched above, the semiclassical scattering amplitude leads for $E \to 0$ to the classical cross section with a dominant energy variation of the form

$$P_{\mathrm{CL}}(E) = (E/E_0)^{\beta}; \qquad \beta = \sqrt{\frac{100Z - 9}{32Z - 4}} - \frac{1}{4}\,, \tag{5}$$

as derived by Wannier [13].

On the other hand, for a fixed point charge $C_* > 0$ the Coulomb interaction is repulsive. Then, the relevant threshold orbit is classically forbidden and represents a tunneling trajectory with imaginary action $\Phi_* = i\Gamma_*$. In this case the semiclassical cross section does not reduce to the classical one in the limit $E \to 0$. Rather, its major energy dependence results from a tunneling mechanism which produces a Gamow factor

$$P(E) = \exp[-2\Gamma_*(E)/\hbar]\,. \tag{6}$$

Clearly, the threshold cross section is through Eq. (6) $\hbar$ dependent. Nevertheless, the important dynamical quantities, namely, the tunneling action Γ_*, are still given classically, as will be shown next.

In the traditional description of classically allowed threshold fragmentation of charged particles the initial configuration is unimportant—the energy dependence of the cross section is completely determined by the stability of the final fragment configuration. This stability enters P_* of the escape orbit. That only the fixed point orbit is relevant close to threshold is justified by the fact that all available energy (which approaches zero for $E \to 0$ and $r \to \infty$) must be put into the radial degree of freedom r in order to fragment the system. Hence, the system evolves asymptotically in a frozen configuration where neither its geometrical shape ($\theta = \theta^*$) nor the relative interparticle distances $r_1/r_2 = \tan\alpha^*$ change. Moreover, due to the Coulomb scaling properties, any partial wave with angular momentum L reduces in scaled coordinates to an S wave since the scaled angular momentum reads $\tilde{L} = L\sqrt{E}$ [14]. Therefore, only the S wave has to be considered which remains also valid in the case of a repulsive Coulomb force. Finally, for two escaping electrons, the fixed point configuration $\theta^* = \pi$ and $\alpha^* = \pi/4$ remains the same for all charges of the core including the limit $Z = 0$ which applies to the neutral atom for our problem of electron detachment. Hence, the radial motion on the fixed point manifold is governed by the Hamiltonian (atomic units are used unless otherwise stated)

$$H_* = \frac{P_r^2}{2} + \frac{C(\alpha^*, \theta^*)}{r}\,, \tag{7}$$

where the effective charge $C_* = 2^{-1/2}$ results from the evaluation of the electron-electron repulsion $V = |\vec{r}_1 - \vec{r}_2|^{-1}$ at the fixed point.

For each energy $E = H_*$ we can calculate the tunneling action $\Gamma_*(E)$ entering Eq. (6) from the imaginary momentum $p = (-P_r^2)^{1/2}$ of Eq. (7),

$$\Gamma_* = \int_{r_i}^{r_t} p\, dr\,. \tag{8}$$

The integration limits are the outer turning point r_t where the orbit becomes classically allowed, $p(r_t) = 0$, and a starting point r_i; see Fig. 1. In contrast to threshold fragmentation under attractive Coulomb forces tunneling threshold fragmentation depends on the initial configuration, at least as far as the value of r_i in Eq. (8) is

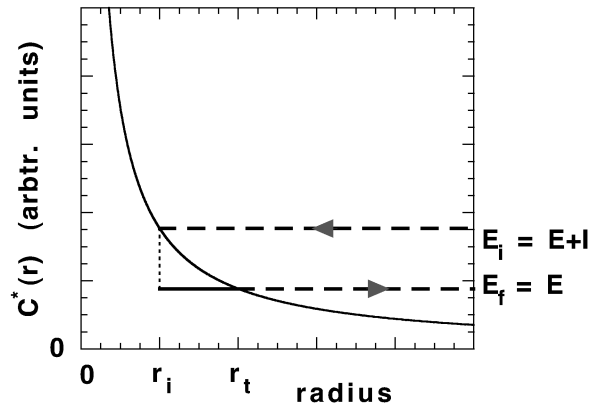

FIG. 1. Sketch of tunneling threshold dynamics on the fixed point manifold with potential $C_*(r)$ from Eq. (7). The classically allowed incoming and outgoing trajectories on the respective energies E_i and E_f are shown (dashed lines), as well as the tunneling part (solid thick line) which determines the threshold fragmentation probability.

concerned, which will influence shape and magnitude of $P(E)$ in Eq. (6).

In a very crude approximation one could put $r_i = 0$ arguing that the electronic momentum transfer requires the recoil to be absorbed by the nucleus and its position is where the outgoing electrons should start. However, in the light of the (small) tunneling probability which determines threshold detachment according to Eq. (6) close to $E = 0$ this is certainly too crude. For small excess energy the projectile electron impacts roughly with the binding energy I which is of the order of 1 eV. Repelled by the loosely bound electron the projectile will never reach $r_i \approx 0$ at this low impact energy. More realistically, one can approximate r_i by the classical turning point of the incoming electron, as it appears on the fixed point manifold whose dynamics is specified by Eq. (7). Hence, to determine this turning point of the *incoming* electron we put $P_r = 0$ in Eq. (7) at the incoming electron energy of $E_i = E + I$ to yield

$$r_i = C_*/(E + I)\,. \tag{9}$$

The initial momentum of the *outgoing* electron pair $p(r_i) = \sqrt{2I}$ follows from the Hamiltonian Eq. (7) on the final energy surface $E_f = E$. The situation is sketched in Fig. 1. Using Eqs. (8) and (9) the threshold detachment probability Eq. (6) reads in dimensionless units explicitly

$$P(E) = \exp\left[-4C\alpha\sqrt{\frac{m_e c^2}{2E}}\left(\arctan\sqrt{\frac{I}{E}} - \frac{\sqrt{IE}}{I + E}\right)\right], \tag{10}$$

where $\alpha = 1/137$ is the fine structure constant, $m_e c^2 = 511$ keV is the rest mass of the electron, and $C = C_*$ is the repelling charge of the two electrons on the fixed point manifold in units of e; see Eq. (7). One can cast Eq. (10) into a more familiar form of atomic units by noting that $m_e c^2/\alpha^2 = e^2/a_0 = 27.2116$ eV is just the atomic energy unit. Clearly, the tunneling mechanism breaks the scaling invariance of $P(E)$ for different systems characterized by different ionization potentials I since $P(E)$ does not depend only on E/I but also on $m_0 c^2/E$. This is one of the major differences compared to Wannier's classical result [Eq. (5)] for threshold ionization under attractive Coulomb forces.

Different $P(E)$ are shown in Fig. 2 with solid lines corresponding to detachment from the ions B^-, D^-, and O^-, respectively. The "experimental" tunneling probabilities are extracted by fitting the experimental cross sections (Fig. 3) to Eq. (4) with

$$\sigma_B(E) = \sigma_0/(b_0 + E/I)\,, \tag{11}$$

where σ_0, b_0 are fitting parameters. The $\sigma_B(E)$ obtained in this way are shown in Fig. 4 for completeness and exhibit the expected monotonically decreasing behavior.

As a final support for the analytical $P(E)$ from Eq. (10) we have fitted the experimental cross sections with σ_0, b_0 and I as free parameters. The result for I was

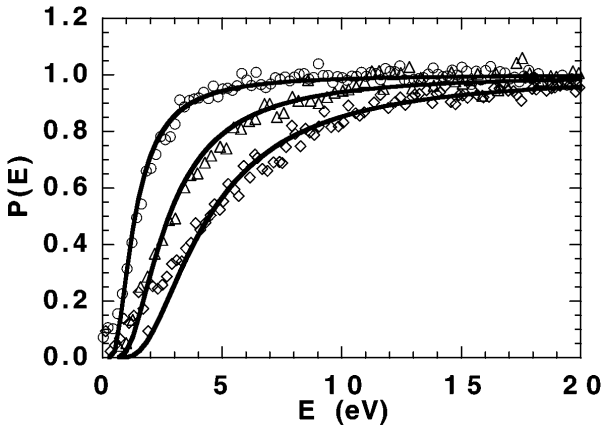

FIG. 2. Experimental detachment probabilities, obtained by dividing the cross section by $\sigma_B(E)$ from Eq. (11). The coding of the data is as in Fig. 3. Theoretical $P(E)$ from Eq. (6).

0.297 ± 0.008, 0.79 ± 0.03, and 1.58 ± 0.04 eV, which is close to the accurate values of 0.28, 0.75, and 1.46 eV for B^-, D^-, and O^-, respectively.

The present description differs from various published tunneling models approximating in one or another way the actual electron motion by tunneling. In the present treatment, only the dominant energy dependence of the cross section is derived from a fixed point orbit which represents a tunneling trajectory. However, this trajectory does not correspond to a true, physical two electron orbit. Rather, it is a stationary point solution for $\hbar \to 0$, in complete analogy to Wannier's solution for the classically allowed case of attractive forces. This stationary point calculated in the limit $E \to 0$ does not depend at all on the polarization potential. Only the binding energy of the target electron enters $P(E)$ through r_i as defined in Eq. (9) from the turning point of the incoming trajectory. It is exactly this element which is similarly contained in the calculation of Ref. [7]. Hence, this impact parameter

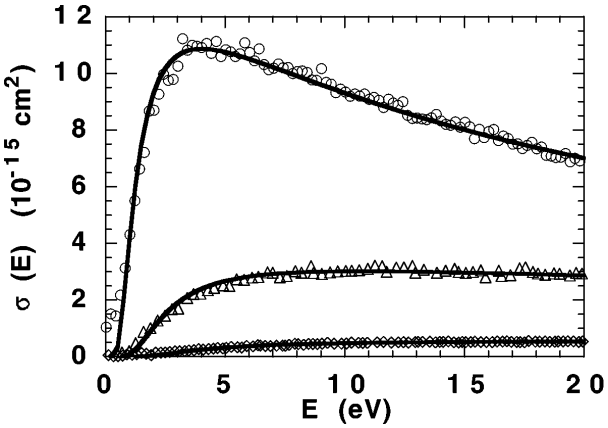

FIG. 3. Detachment cross section by electron impact as a function of excess energy for B^- (circles) from 4, O^- (triangles), and D^- (diamonds) from 1. The solid lines are the cross sections from Eq. (4).

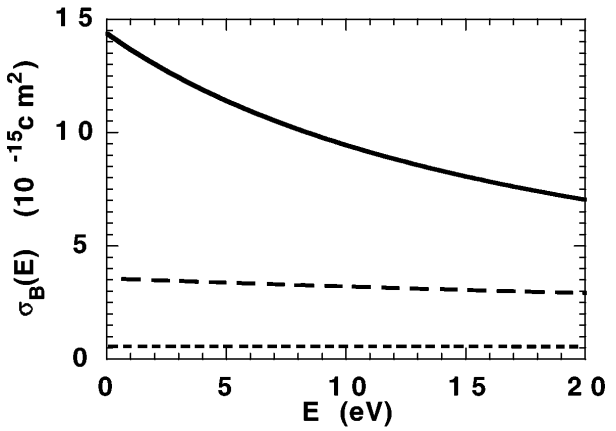

FIG. 4. Background cross sections $\sigma_B(E)$ for B^- (solid line), D^- (dashed line), and O^- (dotted line); see text.

calculation captures an essential feature of the threshold detachment dynamics making the whole calculation robust against details of the polarization potential. These details will influence on the other hand the background cross section $\sigma_B(E)$ much more strongly. The distorted wave calculation [8] by nature approximates the threshold region from an expansion of the high energy limit which is much more sensitive on details of the (shorter range) polarization potential.

In summary, separating the rapidly changing detachment probability $P(E)$ from the background cross section $\sigma_B(E)$ we have shown that threshold fragmentation under asymptotic repulsive Coulomb forces can be treated on the same footing as the well established threshold ionization under attractive Coulomb forces. In contrast to the classical result for attractive forces, threshold detachment of negative ions by electrons can be interpreted to proceed via quantum mechanical tunneling of the outgoing electron pair. This implies a breaking of the scale invariance of $P(E)$ with respect to energy since $P(E)$ depends on $m_e c^2/E$ irrespectively of the target properties while $P_{\mathrm{CL}}(E)$ [Eq. (5)] under attractive Coulomb forces is scale invariant. Yet, $P(E)$ for threshold detachment can be described semiclassically due to the dominant (repulsive) Coulomb interaction which ensures through its scaling properties that $E \to 0$ also means $\hbar \to 0$ [see Eq. (2)]. The same scaling properties also reduce the dominant energy dependence of all partial waves to that of $L = 0$. Therefore, $P(E)$ can be determined from the S wave only, as has been done in the present work.

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