Quantum over-barrier reflection effects manifested in photodetachment cross sections

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Abstract
The photodetachment of H− near a potential barrier is studied. A new formula is presented for the cross sections induced by a barrier. The new formula describes two quantum effects near the barrier tops. For energies near and above the barrier tops, the quantum over-barrier reflection effects are included and the induced oscillations in the cross sections are still prominent; for energies near and below the barrier tops, the quantum tunnelling across barriers is taken into account and consequently the oscillations are weakened. For energies far away from the barrier tops, the new formula agrees with the standard closed-orbit theory. We demonstrate that a potential barrier of various width and location can be realized by placing a negative ion near a metal surface and applying an electric field pointing to the surface. The width and location of the barrier can be systematically changed by varying the electric field strength and the distance between the negative ion and the surface. Results are also presented for estimating the sizes and locations of the aforementioned quantum effects in the cross sections.

1. Introduction
Photodetachment cross sections of a negative ion in an external electric field have been shown to exhibit oscillatory structures [1–5]. Similar oscillations occur in the presence of an interface [6]. Studies on the energy level shifts of Rydberg atoms and dynamics near a metal surface have also been reported [7–11]. Inspired by these developments, the study of photodetachment near a metal surface was proposed and the cross sections are shown to be oscillatory because of the image charge [12]. Recently, we demonstrated that the photodetachment cross sections near the metal surface can be systematically modified [13] by applying a weak electric field pointing away from the metal surface. However, if the applied electric field points towards the metal surface, then potential barriers are created near the negative ion. This new configuration implies interesting physics and has not been studied for negative-ion photodetachment before.

Because quantum tunnelling and quantum over-barrier reflection effects are ignored, the standard closed-orbit theory [14, 15] cannot appropriately describe the photodetachment cross sections for energies near the barrier tops. If the standard closed-orbit theory is applied to our present system, the photodetachment cross section would be oscillatory for energies below the barrier tops; for energies above the barrier tops, the oscillations would be absent. Therefore, there is a sudden change in the cross sections at the energies coinciding with the barrier tops. We note, in the atomic case, it has been shown that broader resonance states in the absorption spectra are associated with quasi-classical orbits undergoing quantum above-barrier reflection [16].

In this paper, we study the photodetachment of a hydrogen negative ion near a potential barrier created by a metal surface and an electric field. We will derive a modified formula for the cross sections, which requires the quantum reflection amplitude of the barrier as an input. The modified formula gives an oscillatory cross section for energy above the barrier top induced by quantum over-barrier reflection. It also gives a cross section with a weaker oscillation for energy...
slightly below the barrier top due to quantum tunnelling. The theoretical cross section is now smooth when the energy crosses the barrier-top energy. As expected, for energies far away from the barrier tops, the modified formula agrees with the formula based on standard closed-orbit theory.

For potential barriers created by a metal surface and an electric field, it is possible to vary the barrier width and location by varying the electric field strength \( F \) and the distance \( d \) between the negative ion and the metal surface. We will survey the parameter space \((F, d)\) and present a guide which can be used to estimate the location and the size of the energy window in which the quantum reflection and tunnelling effects are important. We find that the size of the window is determined by the electric field strength alone and is given accurately by \( 4F^3/\sqrt{\pi} \). Atomic units are used unless specified otherwise.

### 2. Hamiltonian and potential barriers

The configuration for creating a potential barrier is depicted schematically in figure 1 where a negative ion \( \mathbf{H}^- \) sits in front of a metal surface and an electric field \( F \) is applied pointing to the surface. We assume that the negative ion sits at the coordinate origin \((z = 0)\). The distance between the negative ion and the metal surface is denoted as \(d\). The metal surface cuts the \(z\)-axis at \(z = -d\).

Let \( p_r \) and \( p_z \) represent the momentum components of the electron in \(r\)- and \(z\)-directions in a cylindrical coordinate system, then the Hamiltonian governing the motion of the detached electron can be written using the image method [7, 12] as

\[
H = \frac{1}{2}(p_r^2 + p_z^2) - \frac{1}{4(d + z)} + \frac{1}{4d} - Fz, \tag{1}
\]

where the second term represents the image potential induced by the metal surface and the last term is the potential of the applied electric field. We have also added a constant term \(1/F\) so that the potential is set to zero at the origin. For the potential in equation (1) and a weak electric field satisfying \( F < \frac{1}{4d} \), a fatter barrier is created on the positive \(z\) side; when a strong electric field satisfying \( F > \frac{1}{4d} \) is applied, a thinner barrier is created lying between the surface and the negative ion. Because the quantum over-barrier reflection effects and quantum tunnelling effects are stronger for the thinner potential barriers, we will focus on the thinner barriers \(F > \frac{1}{4d}\) in the following discussions. It is easy to show that the peak position of the thinner potential barrier is located at \(z_{\text{max}} = \sqrt{7/4F} - d\) and the barrier-top energy is \(E_{\text{max}} = -\sqrt{F} + Fd + \frac{1}{4d}\). Furthermore, we find that the width of the barrier is determined by the electric field alone as will be discussed later. Thus, the width of the barrier can be varied by varying the electric field strength \( F \) and the location of the barrier in this system can be controlled by \(d\) and \(F\). A typical thin potential barrier is shown in figure 2.

The physical picture for the photodetachment of \(\mathbf{H}^-\) near the barrier top can be described as the following. When a photon is absorbed by the negative ion, the loosely bound \(s\)-state electron goes into an outgoing \(p\)-wave, propagating out from the negative ion in all directions. When the outgoing wave propagating initially towards the metal surface reaches the potential barrier, part of the wave is reflected by the potential barrier and the reflected wave then returns to the region of the negative ion. The interference between the returning wave and the outgoing wave induces oscillations in the cross sections.

If we assume that the metal surface absorbs the detached electron reaching the surface and the standard closed-orbit theory is applied for the present system [12], the photodetachment cross section for photon polarized in the \(z\)-direction would be given by the following formula:

\[
\sigma_{\text{standard}} = \frac{16\pi^2}{3c}\left(\frac{3}{2}\right)^{3/2} \sqrt{2B}E_0^{3/2} \left[\Theta(V_{\text{max}} - E) - \Theta(V_{\text{max}} - E)ight]^{1/2} B \cos[S_{z}], \tag{2}
\]

where \([5] B = 0.31552\) is related to the normalization of the initial bound state of the hydrogen negative ion; \(c\) is the speed of light in \(\text{au}\) and is approximately 137; \(E_0\) is the binding energy of \(\mathbf{H}^-\) and is approximately 0.7542 eV; \(E\) is the initial kinetic energy of the detached electron. The sum of \(E\) and \(E_0\) is equal to the photon energy. \(T_s\) and \(S_z\) are, respectively, the classical transit time and the action for the electron along the closed orbit which goes along the \(z\)-axis from the origin to the barrier and back to the origin for energy below the barrier top. \(\Theta(x)\) is the Heaviside step function:

\[
\Theta(x) = \begin{cases} 
0, & x < 0; \\
1, & x \geq 0.
\end{cases} \tag{3}
\]

However, the standard treatment leading to the formula in equation (2) ignores the quantum over-barrier reflection effects and quantum tunnelling effects and therefore is incorrect for energy close to the barrier top. According to equation (2), when the energy is above the barrier-top energy \(E_{\text{max}}\), the wave will not be reflected at all and consequently there is no oscillation.
(a) When the detached-electron energy is much lower than the barrier top, the reflection is negligible. When the detached-electron energy is much higher than the barrier top, the reflection is nearly complete; (b) in the grey region near the barrier top, quantum tunneling effects and quantum over-barrier reflection effects are significant, resulting in a partial reflection of the wave; (c) when the detached-electron energy is much higher than the barrier top, the reflection is negligible. 

Figure 2. Illustration of the reflected waves in the photodetachment near a potential barrier created with an electric field. The barrier reflection is nearly complete. (b) in the grey region near the barrier top, quantum tunneling effects and quantum over-barrier reflection effects are significant, resulting in a partial reflection of the wave; (c) when the detached-electron energy is much higher than the barrier top, the reflection is negligible.

3. Derivation of the modified formula

We start with a quite general expression for the photodetachment cross section [5, 17]:

$$\sigma = \frac{4\pi^2}{c(E+E_b)} \sum_n |\langle f_{E,n} | p_z | i \rangle|^2 \delta(E_f - E) \, df,$$  

where the laser polarization is assumed along the z-axis, $|i\rangle$ denotes the initial wavefunction, $|f_{E,n}\rangle$ represents the final wavefunction, and the summation includes all the involved final channels.

The initial bound-state wavefunction of $H^-$ is usually described as $\psi_i = Be^{-i\alpha z}$ [5], where $k_b = \sqrt{2E_b}$ and $B$ and $E_b$ are known constants. For this initial wavefunction and noting the potential in equation (1) depends on $z$ but not $x$ and $y$, the general expression in equation (4) can be turned into the following form with some manipulations given in the appendix:

$$\sigma = \frac{8\pi^3 B^2}{c(E+E_b)} \sum_{n=1}^2 \int_{-\infty}^E \left| \frac{\partial \langle z | E_z, n \rangle}{\partial z} \right|^2 \, dE_z,$$

where $\langle z | E_z, n \rangle$ is the energy-normalized coordinate-space wavefunction in the $z$-direction.

For the present problem with a potential barrier, there are two channels. The $n=1$ channel represents a wave with incoming and reflected waves on the left side of the barrier and transmitted wave on the right side of the barrier; the $n=2$ channel represents a wave with incoming and reflected waves on the right side of the barrier and a transmitted wave on the left side of the barrier.

The wavefunction for channel $n=1$ far away from the barrier and outside the surface can be written as

$$\langle z | E_z, n \rangle = 1 \frac{1}{\sqrt{2\pi k_c}} \exp \left( i \int_0^z k_c dz' \right) + \frac{\alpha}{\sqrt{2\pi k_c}} \exp \left( -i \int_0^z k_c dz' \right), -d < z \ll z_{max};$$

$$\frac{\eta}{\sqrt{2\pi k_c}} \exp \left( i \int_0^z k_c dz' \right), z \gg z_{max},$$

where $k_c = \sqrt{2(E_z - V(z))}$; $\alpha$ and $\eta$ are respectively the quantum reflection amplitude and the quantum transmission amplitude of the barrier with an incoming wave from the left.

For channel $n=2$, the wave far away from the barrier and outside the surface can be written as

$$\langle z | E_z, n = 2 \rangle = 1 \frac{1}{\sqrt{2\pi k_c}} \exp \left( -i \int_0^z k_c dz' \right) + \frac{\gamma}{\sqrt{2\pi k_c}} \exp \left( i \int_0^z k_c dz' \right), -d < z \ll z_{max};$$

$$\frac{\beta}{\sqrt{2\pi k_c}} \exp \left( -i \int_0^z k_c dz' \right), z \gg z_{max},$$

where $\gamma$ and $\beta$ are respectively the quantum reflection amplitude and the quantum transmission amplitude of the barrier with an incoming wave from the right. At the same scattering energy, $\eta$ and $\gamma$ satisfy the following relationship [18]:

$$|\eta|^2 + |\gamma|^2 = 1.$$
those displayed in figure 3. The phase energy and there is no stationary point in the interval \((0, E)\).

\[\phi(E) = \int_0^E \sqrt{2E - V(z)} \, dz \]

where \(\phi(E) = \phi_0 \gamma(E)\). Based on an analysis guided by closed-orbit theory \([14, 15]\), it is concluded that the contribution to the integral in equation (12) from the lower boundary should be neglected and when the approximation in equation (12) is used in equation (11), we obtain the modified photodetachment cross section near a barrier:

\[\sigma = \frac{16\sqrt{2\pi} B_3^{3/2}}{3c(E_b + E)^3} + \int_0^E \frac{8\pi^2 B_3^2 \sqrt{2E}}{c(E_b + E)^3} \cos[S_q(E)] \, dE.\]

where

\[S_q(E) = \phi(E) + \pi/2,\]

\[T_q(E) = \frac{\phi_0 d\phi(E)}{dE}.\]

By comparing with the numerical calculations using equation (5), we have verified that the approximate formula in equation (13) is accurate.

We note that for energy well below the barrier top, \(|\gamma|\) approaches unity; equation (13) then should go back to the standard formula equation (2). This implies, in the low energy limit, \(S_q\) should be equal to \(S_c = \frac{1}{2} \rho dq\) and \(T_q\) should be equal to the classical transit time \(T_c\). However, for energy above the barrier top, both \(T_c\) and \(S_c\) are not defined, while \(S_q\), \(T_q\) and \(|\gamma|\) are all well defined by quantum scattering theory. For energy below the barrier top, because \(|\gamma|\) is smaller than 1 due to quantum tunnelling across the potential barrier, the oscillation amplitude described by equation (13) is smaller compared to that in equation (2). For energy above the barrier top, the modified formula in equation (13) still describes an oscillation in the cross section associated with the quantum over-barrier reflection effects. The cross section described by equation (13) is smooth when the detached-electron energy crosses the barrier-top energy \(V_{\text{max}}\).

4. Numerical calculations

We now apply equation (13) to the Hamiltonian in equation (1). For \(d = 200a_0\) and \(F = 300 \text{ kV cm}^{-1}\), the potential barrier is illustrated in figure 2. An iterative numerical method was applied for solving the quantum scattering problem of the barrier \([19]\) with the boundary conditions given in equations (6) and (7) for \(z > -d\), assuming that the detached electron is absorbed when it reaches the surface. The calculated \(|\gamma|\) and \(S_q(E)\) are shown in figure 3 as functions of photon energy. The important energy window we are focusing on near the barrier top is marked grey in both figures 2 and 3. We observe, as the energy is increased crossing the energy window from below to above the barrier top, the amplitude \(\gamma\) displayed in figure 3(a) decreases and the phase \(S_q(E)\) shown in figure 3(b) increases smoothly. Figure 3(b) also demonstrates that the phase \(S_q(E)\) (dotted lines) and \(S_q(E)\) coincide for energy sufficiently below the barrier top.
Figure 4. Photodetachment cross sections (red curves) using the formula in equation (13) for the barrier in figure 2. For comparison, the results obtained by equation (2) are also shown as blue curves. The dotted lines are the cross sections of a free negative ion. The rectangular area in (a) corresponds to the grey energy window in figure 2, which is amplified in (b) for clarity.

Figure 5. Similar to figure 4 but now $d = 400a_0$ and $F = 300$ kV cm$^{-1}$.

5. Sketches of parameter space

For the Hamiltonian system generated by a metal surface and an electric field in equation (1), a potential barrier is always created between the metal surface and the negative ion for any pair of $(d, F)$ satisfying $F > \frac{1}{4d^2}$. The location and the width of the barrier of course depend on the values of $d$ and $F$ and they in turn control the location and size of the energy window in the cross sections related to the quantum over-barrier reflection effects and quantum tunnelling effects. In this section, we present some formulas and figures so one can quickly estimate the barrier scattering region in the cross sections for any given $d$ and $F$.

First, as we mentioned, the barrier top corresponds to a detached-electron energy $V_{\text{max}} = -\sqrt{F + Fd + \frac{1}{4d^2}}$. Therefore, the position of the barrier scattering region (such as the shaded energy range in figure 2 or the rectangular region in figure 4(a)) in the cross section is located at photon energy $E_c = E_b + V_{\text{max}}$. In figure 6(a), we show how $E_c$ depends on $d$ and $F$. In the figure, each solid line corresponds to the same value of $E_c$ given on the right in eV. The locations of the two calculations in the previous section are illustrated as circles.

Next, we estimate the energy width of the barrier scattering region or the rectangular region in figure 4 or figure 5. As illustrated in figure 3, the energy width corresponds to the reflection coefficient varying from almost 1 to almost zero or the transmission coefficient varying from almost zero to almost 1. To proceed, let us approximate the potential barrier
Figure 6. Estimating the barrier scattering region. (a) Barrier scattering window position in photon energy (eV) indicated on the right; (b) barrier scattering window width $\Delta E$ as a function of $F$ in equation (17); (c) number of oscillations within the barrier scattering window indicated on the right. The dotted line in (a) is the boundary of the region satisfying $F > 1/(4d^2)$.

near the barrier top by $-K(z - zm)^2/2 + V_{\text{max}}$. It is easy to show that $K = 4F^2/3$. The transmission coefficient for this parabolic potential can be written as [20]

$$T = \begin{cases} 
\frac{1}{1 + \exp(-2\pi|\mu - V_{\text{max}}|/\sqrt{K})}, & \text{if } E \geq V_{\text{max}}, \\
\frac{1}{1 + \exp(2\pi|E - V_{\text{max}}|/\sqrt{K})}, & \text{if } E \leq V_{\text{max}}.
\end{cases}$$

(16)

The energy width $\Delta E$ can be simply estimated as corresponding to the transmission coefficient in the range between $1/16$ ($\sim 0.0019$) and $1/16e$ ($\sim 0.9981$). Then, we have the result

$$\Delta E = 2\sqrt{K} = 4F^2.$$  

(17)

Figure 6(b) shows the energy width $\Delta E$ as a function of $F$.

The fact that the width $\Delta E$ depends only on the electric field can be understood in the following way. In fact, all the barriers corresponding to the same $F$ but different $d$ have the same shape. To see this, we use the new coordinate $z'$ = $z + d$ relative to the metal surface in the potential $V(z, d, F) = -\frac{1}{4d(z' + d)} + \frac{1}{2d} - Fz'$ in equation (1). We then have $V(z, d, F) = (-1/(4z_m') - Fz_m') + (Fd + 1/d)$. The shape of the potential $V(z, d, F)$ is determined by $(-1/(4z_m') - Fz_m')$ which is independent of $d$.

Finally, we estimate the number of oscillations in the barrier scattering window given in equation (17). The number of full oscillations in the energy window can be obtained from the phase change $\Delta \phi$ across the energy window divided by $2\pi$.

The phase change $\Delta \phi$ is approximately twice the semiclassical phase change from the barrier top to the lower edge of the energy window, $\Delta \phi = 2\int (V_{\text{max}} - S(E - \Delta E/2))$, where $S(E)$ is the integral along the closed orbit which goes out from the negative ion to the barrier and back to the negative ion. For the potential in equation (1), we have

$$S(E) = \frac{4\sqrt{2F}}{3} \left\{ \sqrt{z_m + d} \left[ (z_m + z_n + 2d)E(\mu, \lambda) \right] 
- (z_m - z_n)F(\mu, \lambda) \right\} - \left( z_m + 2z_n + 2d \right) \sqrt{\frac{z_m - d}{z_n - d}},$$

(18)

where $z_m$ and $z_n$ are the two turning points of the barrier. They are given by

$$z_m = \frac{1}{2F} \left[ -\left( Fd + E - \frac{1}{4d} \right) 
+ \sqrt{\left( Fd - (E - \frac{1}{4d}) \right)^2 - F} \right],$$

(19)

$$z_n = \frac{1}{2F} \left[ -\left( Fd + E - \frac{1}{4d} \right) 
- \sqrt{\left( Fd - (E - \frac{1}{4d}) \right)^2 - F} \right].$$

(20)

$F(\mu, \lambda)$ and $E(\mu, \lambda)$ are, respectively, the elliptic integrals of the first kind and the second kind\(^1\). The two parameters are defined as

$$\mu = \arcsin \sqrt{\frac{z_m}{z_n}}, \quad 0 < \mu \leq \frac{\pi}{2};$$

$$\lambda = \sqrt{\frac{z_n + d}{z_m + d}}, \quad 0 < \lambda \leq 1.$$  

(21)

In figure 6(c), we show the number of full oscillations as a function of $d$ and $F$. The estimated full oscillations for the two calculations in section 4 are respectively 2.05 and 3.30, which are consistent with the cross sections displayed in figures 4 and 5. Figures 6(a)–(c) together provide a quick and useful estimate of the location, the size and the number of oscillations for the barrier scattering energy window in the cross sections for any given $d$ and $F$.

6. Conclusions

We studied photodetachment cross sections for a negative ion near potential barriers. The potential barriers can be created with a metal surface and an electric field pointing to it. The image potential should be accurate when the distance from the metal surface is greater than about 10 au and if the kinetic energy of detached electron is less than the Fermi energy of the metal [22]. We have extended closed-orbit theory and derived a modified formula in equation (13). This new formula describes both quantum over-barrier reflection effects and quantum tunnelling effects. These effects have been excluded

\(^{1}\) We have used the integral formulas 3.141.36 of [21].
in the standard formula in equation (2). As illustrations, we have calculated cross sections for two different barriers and shown them in figures 4 and 5. The most significant features in the cross sections are the visible oscillations above the barrier tops. Such oscillations are direct manifestations of the quantum over-barrier reflection effect in the photodetachment cross sections and may be observable in experiments. One may imagine an experimental setup in which a beam of negative ions travels quickly through the interaction region from left to right as in figure 1, keeping the distances between the ions and the metal surface close to a certain value. While the ions are in the interaction region, a laser is applied and the resulting detached electron is measured.

For the combined electric field and metal surface system in equation (1), we have surveyed the parameter space (d, F) satisfying \( F > \frac{1}{k_e^2} \). We have estimated the location and the width of the barrier scattering region in the cross sections. The oscillations inside the barrier scattering region have been estimated as well. The results are comprehensively summarized in figure 6. For any pair of d and F, one can use the map given in figure 6 to find the barrier scattering window without performing complicated calculations. For future experimental reference, we note the electric field F alone determines the potential barrier shape and width.

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Appendix
In momentum space the initial-state wavefunction is [5]

\[
\langle p | i \rangle = 2^{1/2} \pi^{-1/2} \cdot \frac{B}{k_b^2 + p^2}
\]  
(A.1)

and the final-state wavefunctions are

\[
\langle p | f_{E_n} \rangle = \delta(p_i - p_f) \delta(p_f - p_z) \langle p_f | E_n \rangle.
\]  
(A.2)

The matrix element in equation (4) can therefore be written as

\[
\langle i | p_f | f_{E_n} \rangle = \frac{\sqrt{2} B}{\sqrt{\pi}} \int \frac{p_z \langle p_z | E_n \rangle}{k_b^2 + p_z^2 + p_e^2 + p_f^2} \; dp_z.
\]  
(A.3)

Recognizing the main contribution to the cross section comes from the energy shell E, the slowly varying denominator is evaluated at the final energy surface and then moved out of the integral as was done earlier [5]. The integral can then be simplified to

\[
\langle i | p_f | f_{E_n} \rangle = \frac{\sqrt{2} B \pi^{-1/2}}{k_b^2 + p_f^2} \int p_z \langle p_z | E_n \rangle \; dp_z.
\]  
(A.4)

Because

\[
\langle p_z | E_n \rangle = \int \langle p_z | z \rangle \langle z | E_n \rangle \; dz
\]  
(A.5)

and

\[
\langle p_z | z \rangle = \frac{1}{\sqrt{2\pi}} \exp(-i p_z z),
\]  
(A.6)

the integral in equation (A.4) can be written as

\[
\int p_z \langle p_z | E_n \rangle \; dp_z = i \cdot \sqrt{2\pi} \int \frac{\delta(z)}{dz} \cdot \langle z | E_n \rangle \; dz.
\]  
(A.7)

Thus, we have

\[
\langle i | p_f | f_{E_n} \rangle = -i \cdot \frac{2B}{k_b^2 + p^2} \left\{ \frac{\partial \langle z | E_n \rangle}{\partial z} \right\}_{z=0}.
\]  
(A.8)

When the result in equation (A.8) is used in equation (4), we obtain equation (5).

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