Over-the-barrier electron detachment in the hydrogen negative ion

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Abstract

The electron detachment from the hydrogen negative ion in strong fields is studied using the two-electron and different single-electron models within the quasistatic approximation. Special attention is payed to over-the-barrier regime where the Stark saddle is suppressed below the lowest energy level. It is demonstrated that the single-electron description of the lowest state of the ion, that is a good approximation for weak fields, fails in this and partially in the tunnelling regime. The exact lowest state energies and detachment rates for the ion at different strengths of the applied field are determined by solving the eigenvalue problem of the full two-electron Hamiltonian. A simple formula for the rate, which is valid in both regimes, is determined by fitting the exact data to the expression estimated using single-electron descriptions.

Keywords: hydrogen negative ion, strong field, potential barrier, tunneling, electron detachment

(Some figures may appear in colour only in the online journal)

1. Introduction

Although it is one of the simplest systems in atomic physics, the hydrogen negative ion (H⁻) is even today a subject of extensive experimental and theoretical studies, including those dealing with the electron photodetachment (among very recent publications see [1–5]). The first studies concerning H⁻ began in the early years of quantum mechanics with the focus on the ground state of the free ion. The existence of H⁻ as a bound system had been proposed theoretically by Bethe in 1929 [6] (see a historical review of H⁻ in [7] and an overview in the context of negative ions in [8]). Earlier predictions based on simple perturbational or variational methods (using, for example, the variational wave function \( \psi \sim \exp \left(-a(r_1 + r_2)\right) \)) had failed, even though these methods were well suited to predict the most of properties of other members of the two-electron isoelectronic sequence such as He, Li⁺⁺, Be⁺⁺, etc. This is not surprising since the interaction between electrons in the hydrogen negative ion, unlike to helium atom and two-electron positive ions, is comparable in magnitude to that between the nucleus and electrons. As a consequence H⁻ is a weakly bound system which has only one bound state—the ground state. Its binding energy is \( E_0 = 0.75420 \) eV (0.027716 a.u.) [9, 10]. (The reference value for our calculation will be, however, the value \( E_0 = 0.027751 \) a.u. obtained in the nonrelativistic approach for infinite nuclear mass [11].) A very weak binding and the absence of a long-range Coulomb attraction for the separated electron (the atomic residue is the neutral hydrogen atom) results in the fact that this two-electron system has no singly excited states.

The wave function and probability amplitude of a weakly bound system such as H⁻ can extend beyond the range of the binding potential itself. As it was recognized by Chandrasekhar more than 70 years ago [12] (see also [7]), the ground state wave function of H⁻ exhibits a specific radial correlation between the electrons such that one electron is bound much closer to the nucleus than the other which is weakly held at a distance of 4–5 Bohr radii from the nucleus. In contrast to the wave function with equivalent electrons \( \psi \sim \exp \left(-a(r_1 + r_2)\right) \) (see figure 1(c)), the Chandrasekhar’s wave function of the form \( \psi \sim \exp(-ar_1 - br_2) + \exp(-ar_2 - br_1) \) with the parameters \( a = 1.03925, \ b = 0.28309 \) (see figure 1(a)) provides the stability of H⁻ [12]. Such a configuration suggests a very useful one-electron picture where the outer electron is weakly

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perturbation treatment was done by Keldysh [17]. Subsequently, the idea was developed by Perelomov, Popov and Terentev (the PPT theory) [18] and later reconsidered by Faisal [19] and Reiss [20]. Finally, Gribakin and Kuchiev [21] demonstrated that this approach produces very accurate quantitative results for the photodetachment of negative ions. During the last two decades, intense lasers have made it possible to observe effects of multiphoton absorption by atoms and ions [7, 8, 22, 23].

At larger intensities, however, another mechanism for the electron detachment arises—the quantum-mechanical tunnelling. A strong field distorts the potential of atomic residue forming a potential barrier (Stark saddle) through which the electron can tunnel. Finally, at a sufficiently strong field the barrier is suppressed below the energy of the bound state. This regime can be referred to as over-the-barrier detachment (OBD). The transition from the multiphoton to the tunnelling regime is governed by the Keldysh parameter \( \gamma = \omega (2mE_B)^{1/2}/eF \) [17], where \( F \) is the peak value of the electric component of electromagnetic field. This parameter characterizes the degree of adiabaticity of the motion through or over the barrier: If \( \gamma \gg 1 \) (low-intensity/short-wavelength limit) multiphoton processes dominate, whereas for \( \gamma \ll 1 \) (high-intensity/long-wavelength limit) the tunnelling or OBD mechanism does.

A good approximation in the later case (\( \gamma \ll 1 \)) is the quasistatic approximation. It assumes that the electric field changes slowly enough that a static detachment rate can be used at each instantaneous value of the field. Then the detachment rate for an alternating field can be obtained by averaging the static rates over the field period. The static rates can be determined by the PPT theory (or its extensions) in the limit \( \omega \rightarrow 0 \). For neutral atoms the most commonly used rate formula is the Ammosov–Delone–Krainov (ADK) semiclassical formula [24]. It accurately predicts tunnelling rates in experiments with atomic ionization in strong fields [25, 26] and shows good agreement with available results of \( ab \text{ initio} \) calculations (for H and He see [27–29]). Even for atoms with low ionization potentials like alkali metals, using a correction which accounts for the Stark shift, the ADK rates agree well with numerical results [30].

For the field strengths belonging to the barrier-suppression region, however, the PPT (or ADK) theory significantly overestimates the exact ionization rates. For this reason different modifications of the rate formula which extend its applicability to this region are proposed (for the hydrogen atom see [31, 32]). A formula which significantly improves the rates at stronger fields and can be applied to a wide class of atoms and molecules is the empirical formula proposed by Tong and Lin [33]. This formula, however, does not provide an improvement in the rates when the atomic residue is neutral, that is the case in the \( H^- \) detachment. Hence, one of the goals in this paper is to determine exact static rates for this ion and find an adequate empirical formula.

In the next section we consider the full two-electron model for the hydrogen negative ion in the static electric field, investigate the change of form of the lowest-state-wave-function when the field changes from zero to a value in OBD.
domain and present the results of \textit{ab initio} calculations of the lowest state energies and widths (detachment rates) for different field strengths. In section 3 we consider the single-electron description of the same problem using several approaches, present the corresponding results and compare them with the two-electron results and mutually. Finally, in section 4 we give conclusions and summary.

2. Two-electron description

The Hamiltonian describing the full dynamics of two electrons of $H^-$ in a static electric field $F$ reads (in atomic units)

$$H = -\frac{1}{2} (\Delta_1 + \Delta_2) - \frac{1}{r_1} - \frac{1}{r_2} + \frac{1}{r_{12}} - F (z_1 + z_2), \quad (1)$$

where $r_1$ and $\frac{1}{r_1}$ are the position and the kinetic energy operator of the $i$th electron, respectively, and $r_{12} = |r_1 - r_2|$. Due to presence of the barrier all eigenstates of (1) have the resonant character when $F \neq 0$, including the lowest which is an exact bound state for $F = 0$. The width $\Gamma$ of the lowest state determines the electron detachment rate $\Gamma (F) = \Gamma (\tilde{F}) / h$ (hereafter we set $\hbar = 1$ and use atomic units). The eigenstates of (1) are calculated numerically using the complex rotation (scaling) method [34–36]. The calculations are performed in the basis whose elements are the symmetrized products of Sturmian functions [37] for each electron. Before applying to the hydrogen negative ion, the method has been tested by calculating the ionization rates for the helium atom in the static electric field. The excellent agreement with similar \textit{ab initio} calculations of other authors [27–29] is obtained.

Figure 1 in parts (b) and (d) shows the $\lambda_1 = \lambda_2 = \lambda_2 = 0$ cuts of the lowest state wave function of $H^-$ at $F = 0$ (i.e. the $H^-$ ground state wave function) and at the field of strength $F = 0.03$ a.u., respectively, obtained using the above mentioned two-electron approach. By comparing parts (a) and (b) of the same figure, one can see that the Chandrasekhar’s wave function is indeed a good approximation for the ground state of $H^-$. A small difference is due to the lack of angular correlations in the approximate wave function. The outgoing waves of the wave function shown in figure 1(d), representing the (single-electron) escape channels for the first and for the second electron, clearly demonstrate the resonant character of this state.

The lowest state energies and widths of $H^-$ at different strengths of the applied electric field, obtained using the same model and method, are presented in table 1 and in figure 2 together with the results obtained using a single-electron approach (that will be discussed later). Figure 2(a) shows that for weak fields the lowest state energy $E (F)$ decreases by increasing the field strength according to the Stark shift expansion formula

$$\Delta E = E (F) - E (0) = -\frac{1}{2} \alpha F^2 - \frac{1}{4} \gamma F^4 - \cdots \quad (2)$$

Here $E (0) = -0.5 + E_0 = -0.52775$ a.u. is the ground state energy of the free ion, whereas $\alpha = 206$ and $\gamma = 8.03 \times 10^7$ are the corresponding values for the dipole polarizability and the second dipole hyperpolarizability [38, 39]. At stronger fields ($F > 0.01$), however, $E (F)$ decreases almost linearly. The width $\Gamma (F)$, on the other hand, grows rapidly with $F$ (figure 2(b)). These values are consistent with the \textit{ab initio} calculations done by Themelis and Nicolaides [40] using a different method (see figure 1 in this reference).

| $F$ | $-E$ | $\Gamma$ |  | $-E$ | $\Gamma$ |
|-----|------|---------|---|------|---------|
| 0   | 0.52775 | 0   | 0.52775 | 0   |
| 0.001 | 0.52785 | –  | 0.52782 | –  |
| 0.002 | 0.52819 | –  | 0.52806 | 7.330 \times 10^{-5} |
| 0.003 | 0.52881 | 4.310 \times 10^{-4} | 0.52846 | 4.511 \times 10^{-4} |
| 0.004 | 0.52944 | 1.247 \times 10^{-3} | 0.52878 | 1.091 \times 10^{-3} |
| 0.005 | 0.53003 | 2.475 \times 10^{-3} | 0.52931 | 1.913 \times 10^{-3} |
| 0.006 | 0.53067 | 3.845 \times 10^{-3} | 0.52974 | 2.933 \times 10^{-3} |
| 0.007 | 0.53128 | 5.369 \times 10^{-3} | 0.53041 | 4.122 \times 10^{-3} |
| 0.008 | 0.53187 | 7.022 \times 10^{-3} | 0.53053 | 5.335 \times 10^{-3} |
| 0.009 | 0.53248 | 8.789 \times 10^{-3} | 0.53088 | 7.034 \times 10^{-3} |
| 0.010 | 0.53311 | 0.01066 | 0.53121 | 8.440 \times 10^{-3} |
| 0.011 | 0.53361 | 0.01258 | 0.53153 | 9.872 \times 10^{-3} |
| 0.012 | 0.53412 | 0.01451 | 0.53182 | 0.01132 |
| 0.013 | 0.53468 | 0.01654 | 0.53214 | 0.01284 |
| 0.014 | 0.53522 | 0.01861 | 0.53240 | 0.01443 |
| 0.015 | 0.53580 | 0.02078 | 0.53265 | 0.01595 |
| 0.016 | 0.53631 | 0.02291 | 0.53289 | 0.01750 |
| 0.017 | 0.53684 | 0.02505 | 0.53313 | 0.01894 |
| 0.018 | 0.53734 | 0.02730 | 0.53336 | 0.02065 |
| 0.019 | 0.53790 | 0.02956 | 0.53360 | 0.02227 |
| 0.020 | 0.53846 | 0.03186 | 0.53376 | 0.02394 |
| 0.021 | 0.53895 | 0.03414 | 0.53399 | 0.02562 |
| 0.022 | 0.53948 | 0.03647 | 0.53422 | 0.02720 |
| 0.023 | 0.53999 | 0.03883 | 0.53441 | 0.02883 |
| 0.024 | 0.54052 | 0.04122 | 0.53454 | 0.03066 |
| 0.025 | 0.54109 | 0.04362 | 0.53473 | 0.03229 |
| 0.026 | 0.54160 | 0.04606 | 0.53492 | 0.03395 |
| 0.027 | 0.54211 | 0.04848 | 0.53512 | 0.03563 |
| 0.028 | 0.54265 | 0.05097 | 0.53525 | 0.03733 |
| 0.029 | 0.54319 | 0.05349 | 0.53538 | 0.03900 |
| 0.030 | 0.54367 | 0.05599 | 0.53556 | 0.04062 |

\( \gamma = 8.03 \times 10^7 \) are the corresponding values for the dipole polarizability and the second dipole hyperpolarizability.

3. Single-electron description

As mentioned in the introduction, the configuration of the ground state of $H^-$ suggests a one-electron description where the outer (loosely bound) electron moves in a short-range potential $V (r)$ describing the attraction by the neutral atomic residue. Then, in the presence of a (quasi)static electric field $F$ the outer electron may be considered as moving in the total potential $V_{\text{tot}} = V (r) - Fr_z$. $V (r)$ is usually calibrated to give
the value \(-E_B\) for the lowest energy level \(\epsilon(F)\) at \(F = 0\). When \(F \neq 0\) the total potential has a potential barrier that explains the resonant character of states. The saddle point of the barrier is located at the \(z\)-axis. Its position \(r_0 = (0, 0, z_{sp})\) and height \(V_{sp} = V_{ao}(r_0; F)\) depend on the field strength \(F\) and can be determined from the condition \((\partial V_{ao}/\partial z)_{z_{sp}} = 0\).

The field strength \(F_S\) that separates the tunnelling and OBD regimes is defined by the condition \(\epsilon(F_S) = V_{sp}(F_S)\). Note that these values may vary by changing the model for \(V(r)\).

### 3.1. The PPT theory

The first among the single-electron approaches we consider here is the PPT theory [18]. It is based on the assumption that most atoms are nearly hydrogenic, the difference being a small quantum defect that changes the quantum numbers to noninteger effective values. In the case of negative ions, however, the atomic residue is neutral \((Z = 0)\) and the effective principal quantum number \(n^* = Z/\kappa\), where \(\kappa = (2E_B)^{1/2}\), is equal to zero. Then the static-field tunnelling rate formula (see equation (4) in [18]) for negative ions in the ground state reduces to

\[
\Gamma = C_{PPT}^2 \frac{F^2}{\kappa} \exp\left(-\frac{2\kappa^3}{3F}\right).
\]

It should be mentioned that in the general case the PPT and ADK static rate formulae coincide, whereas the ADK theory [24] provides also an explicit expression for \(C_{ADK}\). This expression, however, is not useful in the case when \(n^* = 0\). The coefficient in the pre-exponential factor determined from Hartree–Fock calculations has for \(H^-\) the value \(C_{ADK} = 1.15\) [41]. Figure 2(b) shows that, although \(H^-\) does not belong to the class of hydrogenic atoms, the PPT rate formula exhibits a qualitative agreement with the results obtained by solving the full two-electron problem.

Among new theoretical approaches in the study of tunnel ionization we refer to the so-called weak-field asymptotic theory (WFAT) [42] which is applicable to different atomic and molecular systems. The theory is recently extended to the first-order terms in the asymptotic expansion in the field and a simple analytical formula giving the tunnelling rate for two-electron atoms is obtained [43]. In the case of \(H^-\) the formula reduces to \(w = w_{PPT}(F)(1 - 136.1F)\), where \(w_{PPT}(F)\) is the rate given by equation (3) with \(C_{PPT} = 1.15\). This correction significantly improves the PPT rates in the tunnelling domain (see figure 2(b)), however, the formula diverges in OBD area.

### 3.2. The ZRP model

A better single-electron description of the detachment process in OBD regime is achieved by applying models with a loosely bound electron in a short-range potential. As mentioned above, the simplest short-range potential that can be used to describe the dynamics of a weakly bound electron in negative ions is the ZRP

\[
V(r) = -a\delta(r) \quad (a > 0).
\]

This potential supports only one bound state whose wave function has the form \(\psi_{ZRP}(r) \sim \exp\left(-\kappa r/r\right), \) where \(\kappa \equiv (2E_B)^{1/2} = a\) [14].

The eigenvalue problem of the single-electron Hamiltonian with \(V_{tot} = -a\delta(r) - F_z\) admits for weak fields a solution in a closed analytical form [14]. The position and width of the lowest (and here the only) energy level, determined from the real and imaginary parts of the associated complex eigenenergy, are

\[
\epsilon = -E_B - \frac{F^2}{32E_B}.
\]
\[ \Gamma = \frac{F}{2\kappa} \exp \left( -\frac{2\kappa^3}{3F} \right). \]  

The polarizability \( (\alpha = 1/(16E_B^2) \approx 81.5) \) and the decay rate \( (\mu = \Gamma) \) given by the ZRP approximation are minimal among all systems with potentials \( V(R) \leq 0 \) at a fixed value of the binding energy \( E_B \) [14]. For the ground state in the Coulomb potential, for instance, one has \( \alpha \equiv \alpha^2 = 9/8 \), whereas for the extreme case of a ZRP it is \( \alpha = 1/16 \). This difference may be explained by the change of form of the ground state wave function in the transition from a long-range Coulomb potential (when \( \psi \sim \exp(-\kappa r) \)) to a short (zero) range potential. The ZRP model has a state with the most compressed wave function (\( \psi_{\text{ZRP}} \)) and the polarizability in the external electric field will be minimal. The decay rate \( \Gamma \) exhibits even stronger dependence on the potential form. The rate formula for the hydrogenic atoms in the ground state (then \( E_0 = Z^2/2, \kappa = Z \) and \( n' = 1 \)) reads \( w_C = (4\pi^2/F) \exp(-2\kappa^3/3F) \). Thus \( w_{\text{ZRP}}/w_C = F^2/8\kappa^6 \) and for weak fields one has \( w_{\text{ZRP}} \ll w_C \).

The exponential factor in the tunnelling rate formula in principle remains the same for any form of the attractive potential \( V(r) \) describing the interaction between the outgoing electron and the atomic residue. This follows from the fact that the tunnelling probability is essentially determined by the outer side of the barrier where \( V_{\text{tot}} \approx -Fz \). The pre-exponential factor, on the other hand, may vary by several orders of magnitude. Note, however, that a difference between the detachment rate formulae for \( \text{H}^- \) obtained from the ZRP model and the PPT theory is only in the constant in the pre-exponential factor (that is 0.5 and 1.32, respectively, see equations (3) and (6)). In addition, the rates obtained using the two-electron model also fit well to equation (3). Hence, the optimal value for \( C_n \) can be estimated from these data. For this purpose we express the rate in terms of the variable \( \xi = (F/\kappa) \exp(-2\kappa^3/3F) \). Then the rate formula (3) reduces to the linear dependence \( w = C_n^2 \xi \) (see the inset in figure 2(b)). The linear fit of the two-electron data for \( F < F_3 \) (tunnelling) and for \( F > F_3 \) (OBD) gives \( C_n^2 = 0.65 \) and 0.58, respectively.

### 3.3. Short-range model-potential with the polarization term

Finally, we consider the single-electron model for \( \text{H}^- \) where the loosely bound electron moves in an effective potential which is the sum of a short-range potential and the polarization term. A widely used potential of this type is the Cohen–Fiorentini (CF) potential [44]

\[ V = \left( 1 + \frac{1}{r} \right) e^{-2r} - \frac{\alpha_H}{2r^4} e^{-\alpha_H/2r^2}. \]

where \( \alpha_H = 9/2 \) is the polarizability of the hydrogen atom. The parameter \( \nu_0 = 1.6 \) is chosen by the condition that the potential (7) has a single bound state with the correct binding energy. The lowest state energies \( (E(F) = \epsilon(F) = 0.5 \text{ a.u.}) \) and widths of \( \text{H}^- \) in the static electric field, obtained using the CF potential, are shown in table 1 and figure 2. The calculations were performed using the complex rotation method [34, 36] and the Sturmian basis [37].

At low values of \( F \) the energies obtained by the CF model approximately agree with the two-electron (accurate) results (see figure 2(a)). At stronger fields, however, the difference between these results increases, particularly in the OBD area. The value of \( F \) that separates the tunnelling and OBD regimes obtained using the potential (7) is \( F_3 = 0.0056 \pm 0.0001 \). For \( F > 2F_3 \) the accurate Stark shift \( \Delta E_{2R} \) is approximately two times larger than \( \Delta E_1 \) obtained using the single-electron (CF) model (the uncertainty in \( F_3 \) is due to this difference). The rates determined using the CF model agree with the two-electron results approximately for \( F < F_3/2 \), see figure 2(b). Otherwise the single-electron calculations underestimate the two-electron results (for about 30% in OBD regime).

### 4. Conclusions

Deviations of the energies and detachment rates obtained using the single-electron description of the hydrogen negative ion in strong fields from those obtained using the full two-electron description indicate that the single-electron picture in principle is not valid at stronger fields. At the field strengths \( F \sim F_3 \) the potential barrier is suppressed enough that the lowest state cannot be treated as bound even approximately. In this case the Chandrasekhar’s concept of outer electron is not adequate because a significant part of the corresponding probability amplitude lies at the outer side of barrier (\( |z| \) or \( |z| > |z_{\text{eq}}| \)). In other words the ‘outer’ electron becomes the ‘outgoing’ electron. Simultaneously, the form of the two-electron wave function in the inner region \( (r_1, r_2 < |z_{\text{eq}}| \) becomes more similar to that for equivalent electrons (see figures 1(c) and (d)), that explains the failure of single-electron approach (particularly for energies). The ratio between the Stark shifts determined numerically using the two-electron and the one-electron model, \( \Delta E_{2R}/\Delta E_1 \approx 2 \) for \( F \gg F_3 \), may be explained by the fact that in the states of this form the shift \( \Delta E_{2R} \) includes the contributions of both electrons.

Regarding the detachment rate formulae, it is found that if we relax the constant in the pre-exponential factor of equation (3), the rates obtained using the two-electron model fit well to this formula although it is derived in the single-electron approach. This can be explained by a weak sensitivity of the tunnelling rate on the form of potential describing the interaction between the outgoing electron and the atomic residue. As mentioned at the end of section 3.2, the optimal values for this constant are \( C_n^2 = 0.65 \) for \( F < F_3 \) (tunnelling) and \( C_n^2 = 0.58 \), for \( F > F_3 \) (OBD). With these values of \( C_n^2 \) formula (3) can be used to calculate the photodetachment probability in the quasistatic approximation (\( \gamma \ll 1 \))

\[ P_{\text{det}} = 1 - \exp \left[ -\int w(|F(t)|) dt \right], \]  

where \( F(t) \) is the electric component of the laser field (beam/pulse). In this case the most optimal value for \( C_n \) is that giving the best static rates for the field strengths around the peak value of \( F(t) \).
In summary, the single-electron description of the lowest state of $H^-$, that is a good approximation in the field-free and low-field cases, fails in OBD and partially in the tunnelling regime. We determined the accurate lowest state energies and detachment rates for $H^-$ at different strengths of the applied (quasi)static field by solving the eigenvalue problem of the full two-electron Hamiltonian. The PPT and ZRP theories lead to the same rate formula, but with different values of the constant in the pre-exponential factor. Optimal values of the constant in the tunnelling and OBD domains are obtained by fitting the numerical results determined using the two-electron model to this expression.

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