Photodetachment cross section of H^- in a gradient electric field

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Based on closed orbit theory, we derive a simple analytic formula for the photodetachment cross section of H^- in a gradient electric field. It is found that the detachment spectrum displays a sawtooth structure, in contrast to the smooth oscillation for the same but in a homogeneous electric field. [S1050-2947(99)07303-5]

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I. INTRODUCTION

In measurements of the photodetachment cross section of hydrogen ion H^- in a strong homogeneous static electric field, it has been observed that the detachment spectrum displays a “ripple” structure [1]. The structure can be identified as a smooth background upon which sinusoidal oscillations are superposed. Theoretical studies on the phenomenon have been carried out by a number of authors at the quantum and the semiclassical levels [2–9]. Photodetachments of H^- in other fields, such as in cross electric and magnetic fields [10] and in parallel electric and magnetic fields [11], have been also investigated theoretically.

To the best of our knowledge, no study on photodetachment of ions in an *inhomogeneous* electric field has been reported in the literature. However, there is a need to do the study. Actually, ions are set in an inhomogeneous field in some experiments. For example, the electric field in the Paul trap consists of a large gradient field and a small oscillating field [12]. In the zeroth-order approximation, the electric field in the Paul trap can be treated as a gradient field.

In the present work, we investigate the photodetachment cross section of H^- in a gradient electric field. Our study is based on the closed orbit theory [13], which emphasizes the role of closed orbits and their repetitions. We obtain an analytical expression for the cross section as the sum of the contributions of the closed orbits and their repetitions. We find that the cross section in a gradient field is quite different from that in a uniform field: the spectrum is an irregular sawtooth curve. We hope that our results will be useful in understanding the detachment process of ions in the Paul trap and other ion traps.

The paper is organized as follows. In Sec. II we give all closed orbits in closed form. In Sec. III the closed orbit theory applied to the detachment of H^- in a gradient electric field is presented and the formula for the cross section is obtained. Numerical results are given in Sec. IV and conclusions are drawn in Sec. V.

II. CLOSED ORBITS

To derive a formula for the photodetachment cross section of H^- by using the closed orbit theory, it is necessary to find all classical orbits of the active electron that start and end at the nucleus.

Let the applied electric field be in the z direction and given by

$$F = F_0 + \alpha z, \quad (1)$$

where F_0 is the constant “background” field and α is the field gradient in z . Without loss of generality, we assume both F_0 and α are positive. A hydrogen ion H^- can be regarded as a one-electron system, with the active electron loosely bound by a short-range, spherically symmetric potential $V_b(r)$, where r is the distance between the active electron and the origin where the nucleus is. In the cylindrical coordinates, (ρ, z, ϕ) , the Hamiltonian governing the electron motion is

$$H = \frac{1}{2}(p_\rho^2 + p_z^2) + (F_0 + \frac{1}{2}\alpha z)z + V_b(r). \quad (2)$$

Here (and throughout the article unless specified otherwise) the atomic units have been used. The ϕ motion has been separated because of the cylindrical symmetry of the system. The z component of the angular momentum is a constant of motion due to the symmetry, and has been set to zero.

When the electron is far away from the nucleus so that the short-range potential $V_b(r)$ is negligible, the classical motion of the electron is analytically solvable. Assuming that the electron is initially on the sphere of radius R centered at the nucleus and emanated with velocity k in the direction of θ , the solution of the equations of motion is

$$\rho(t) = R \sin \theta + (k \sin \theta)t,$$

$$z(t) = R \cos \theta + \frac{1}{\alpha} [F_0 \cos \sqrt{\alpha}t + k \sqrt{\alpha} \cos \theta \sin \sqrt{\alpha}t - F_0]. \quad (3)$$

Note that the ρ motion is a free motion. The z motion is a constant acceleration if $\alpha = 0$, i.e., $z(t) = R \cos \theta + k \cos \theta t - F_0 t^2$. When $\alpha \neq 0$, the z motion is that of a harmonic oscillator with angular frequency $\sqrt{\alpha}$, i.e.,

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$$z(t) = \frac{1}{\alpha} \sqrt{F_0^2 + k^2 \alpha \cos^2 \theta} \sin(\sqrt{\alpha} t + \varphi) - \frac{F_0}{\alpha}, \quad (4)$$

where $\varphi = \tan^{-1}[F_0/(k\sqrt{\alpha} \cos \theta)]$. In the following, we only consider the case for $\alpha \neq 0$ except in the end of this paper where the case for $\alpha = 0$ will be briefly considered as a special case.

It is obvious that only the trajectories emanating up ($\theta = 0$) or down ($\theta = \pi$) in the z direction can be drawn back to the origin by the electric field. Among all orbits closed at the nucleus, the following four are fundamental. (All other closed orbits take these fundamental orbits as parts.) (i) The electron goes up along the $+z$ direction, reaches its maximum and then returns to the origin. The orbit has the shortest returning time and therefore the most important contribution to the cross section. We call this orbit the up orbit. (ii) The electron completes the up orbit first and then, passing through the origin, continues to move down in the $-z$ direction and finally is drawn back to the origin by the field. (iii) The electron goes down along the $-z$ direction, reaches the lowest point where $F < 0$, and then turns back to the origin. Its contribution to the cross section is only less than that of the up orbit. We call this orbit the down orbit. (iv) The electron completes the same trajectory as the down orbit first, and then continues to go up at the origin and finally is back to the origin.

We label a close orbit by two indices (j, n) , where $j = 1, 2, 3, 4$ and $n = 0, 1, 2, \dots$. Here $n = 0$ means the orbit is a fundamental closed orbit ($j = 1, 2, 3$, and 4 for the fundamental closed orbits listed above, respectively). When $n > 0$, orbit (j, n) has two parts, the beginning part and the later part. The beginning part is always the j th fundamental closed orbit. The later part depends on whether the j th fundamental orbit is periodic or not: it is n repetitions of j th fundamental orbit itself if it is periodic (i.e., $j = 2$ or $j = 4$); it is n repetitions of the fourth fundamental orbit if $j = 1$, and of the second fundamental orbit if $j = 3$. For example, orbit $(j = 1, n = 2)$ is the combination of orbits $(j = 1, n = 0)$, $(j = 4, n = 0)$ and $(j = 4, n = 0)$.

From Eq. (4), it is easy to find that the returning times T_{jn} for orbits (j, n) , $j = 1, 2, 3, 4$, are

$$T_{1n} = \frac{1}{\sqrt{\alpha}} [2(n+1)\pi - 2\varphi_0 - \pi], \quad T_{2n} = \frac{1}{\sqrt{\alpha}} 2(n+1)\pi, \quad (5)$$

$$T_{3n} = \frac{1}{\sqrt{\alpha}} [2(n+1)\pi + 2\varphi_0 - \pi], \quad T_{4n} = T_{2n},$$

where $\varphi_0 = \tan^{-1}[F_0/(k\sqrt{\alpha})]$. The action along orbit (j, n) is also easy to find. It is given by

$$S_{jn} = \frac{1}{2\alpha} (F_0^2 + k^2 \alpha) \times \left[T_{jn} + \frac{1}{2\sqrt{\alpha}} [\sin 2(\sqrt{\alpha} T_{jn} + \varphi_0) - \sin 2\varphi_0] \right]. \quad (6)$$

The Maslov index, denoted by μ_{jn} for orbit (j, n) , is the number of the singular points (caustics and foci) along the trajectory. Counting these singular points, we have

$$\begin{aligned} \mu_{1n} &= \mu_{3n} = 2n + 1, \\ \mu_{2n} &= \mu_{4n} = 2(n + 1). \end{aligned} \quad (7)$$

III. PHOTODETACHMENT CROSS SECTION

The photodetachment cross section can be written as

$$\sigma(E) = -\frac{4e^2(E + E_b)}{c} \text{Im} \langle D\Psi_i | \hat{G}^+ | D\Psi_i \rangle. \quad (8)$$

where $E_b \approx 0.745$ eV is the binding energy, E is the energy of electron after absorbing a photon and overcoming the binding threshold, $\hat{G}^+$ is the outgoing Green's function, D is the dipole operator, and Ψ_i is the initial wave function. The physical meaning of Eq. (8) is as follows: the initial state $|\Psi_i\rangle$ is modified by the dipole operator associated with the laser field to become the "source wave function" $|D\Psi_i\rangle$; the Green's function $\hat{G}^+$ propagates these waves outward to become the outgoing waves $\hat{G}^+ |D\Psi_i\rangle$; and finally the waves $\hat{G}^+ |D\Psi_i\rangle$ overlap with the source wave $D\Psi_i$ to give the absorption spectrum.

The outgoing wave $\hat{G}^+ |D\Psi_i\rangle$ can be divided into two parts, $\hat{G}^+ |D\Psi_i\rangle = \hat{G}^{+(dir)} |D\Psi_i\rangle + \hat{G}^{+(ret)} |D\Psi_i\rangle$. The first part never goes far from the core and is called the direct wave. The second part is known as the returning wave that propagates outward into the external region first, then is turned around by the external field, and finally returns to the vicinity of the core. Accordingly, the cross section has two parts,

$$\sigma(E) = \sigma_0(E) + \sigma^{ret}(E). \quad (9)$$

The first part $\sigma_0(E) = -(4e^2 E_p / c) \text{Im} \langle D\Psi_i | \hat{G}^{+(dir)} | D\Psi_i \rangle$ is the contribution of the direct wave interfering with the source. It has been shown that $\sigma_0(E)$ is the field-free cross section [8],

$$\sigma_0(E) = \frac{16\sqrt{2}C^2\pi^2}{3c} \frac{E^{3/2}}{(E_b + E)^3}, \quad (10)$$

where $C = 0.31522$ is a constant related to the normalization of the initial bound state of H^- . It is understandable because there is no returned waves if there is no external field. The second part in Eq. (9) is the contribution of the returning wave,

$$\sigma^{ret}(E) = -\frac{4e^2 E_p}{c} \text{Im} \langle D\Psi_i | \Psi^{ret} \rangle, \quad (11)$$

where $\Psi^{ret} \equiv \hat{G}^{+(ret)} |D\Psi_i\rangle$ is the returning wave, which overlaps with the outgoing source wave to give the oscillation in the spectrum. In Eq. (11), the returning wave Ψ^{ret} will be given by the semiclassical approximation in the following subsection, while the initial wave Ψ_i will be approximated by

$$\Psi_i(r, \theta, \phi) = C \frac{e^{-k_b r}}{r}, \quad (12)$$

where $k_b = \sqrt{2E_b}$.

A. Semiclassical returning wave

To apply the semiclassical theory, consider a small sphere centered at the nucleus and of radius R , which may be any value between 5 and $10a_0$ (a_0 is the Bohr's radius). The outgoing wave on the small sphere, $\Psi^0(R, \theta, \phi)$, is the initial wave in the semiclassical theory. Outside the sphere, the wave propagates semiclassically and is given by

$$\Psi(\rho, z, \phi) = \sum_i \Psi^0(R, \theta, \phi) A_i e^{i[S_i - \mu_i \pi/2]}, \quad (13)$$

where S_i is the action along the i th trajectory, μ_i is the Maslov index characterizing the geometrical properties of the i th trajectory and its neighboring orbits, and A_i is the amplitude of the wave function given by

$$\begin{aligned} A_i(\rho, z, \phi) &= \left| \frac{\det J_i^{(2)}(\rho, z, 0)}{\det J_i^{(2)}(\rho, z, T_i)} \right|^{1/2} \left| \frac{\rho(0)}{\rho(T_i)} \right|^{1/2} \\ &= \left| \frac{R^2 k}{(R + kT_i) \det J_i^{(2)}(\rho, z, T_i)} \right|^{1/2}. \end{aligned} \quad (14)$$

Here T_i is the journey time of the i th orbit and $J_i^{(2)}(\rho, z, t)$ is the 2×2 Jacobian matrix in ρ and z . From the solution of the classical motion of the electron, Eq. (3), we have

$$\begin{aligned} \det J_i^{(2)}(\rho, z, T_i) &= \frac{R + kT_i}{\sqrt{\alpha}} \cos \theta (k \sqrt{\alpha} \cos \theta \cos \sqrt{\alpha} T_i - F_0 \sin \sqrt{\alpha} T_i) \\ &\quad + k \sin^2 \theta \left(R + \frac{k}{\sqrt{\alpha}} \sin \sqrt{\alpha} T_i \right). \end{aligned} \quad (15)$$

The ratio of determinants of Jacobian matrices in Eq. (14) measures the density of the family of trajectories near the i th orbit.

Not all outgoing waves return to the nucleus. Only the returning waves can overlap with the steadily-producing outgoing spherical wave and therefore contribute to the cross section. They are the ones associated to the closed orbits and their repetitions. Let us write the returning wave function associated with closed orbit (j, n) as

$$\Psi_{jn}^{ret} = A_{jn} e^{i[S_{jn} - \mu_{jn} \pi/2]}, \quad (16)$$

where $j = 1, 2, 3$, and 4, and $n = 0, 1, 2, \dots$ as discussed in the last section. All closed orbits lie on the z axis, so either $\theta = 0$ or $\theta = \pi$. After simple manipulation, we have

$$A_{jn} = \left| \frac{R^2 k}{(R + kT_{jn}) k^2 T_{jn}} \right|^{1/2}. \quad (17)$$

Note that the returning time T_{jn} , the action S_{jn} , and the Maslov index μ_{jn} for closed orbit (j, n) are given in the last section.

B. Incoming plane wave

Outside the small sphere, the wave function is approximated by the semiclassical wave given above. Inside the sphere, the electron motion is approximated, in the closed orbit theory, by a free motion, i.e., the wave function is approximately an incoming plane wave. On the sphere, these two wave functions should match. In other words, the returning semiclassical wave should be proportional to the incoming plane wave,

$$\Psi_{jn}^{ret} = N_{jn} e^{ikr \cos \theta_j^{ret}}, \quad (18)$$

where N_{jn} is the proportional constant and θ_j^{ret} is the returning angle of orbits (j, n) , $n = 0, 1, 2, \dots$. N_{jn} can be obtained by taking the limit of the ration $\Psi_{jn}^{ret}/\exp(-ikR \cos \theta_j^{ret})$ as the radius of the small sphere $R \rightarrow 0$. From Eq. (16), we therefore have

$$N_{jn} = \frac{1}{k^2 T_{jn}} \exp \left[i \left(S_{jn} - \mu_{jn} \frac{\pi}{2} \right) \right]. \quad (19)$$

C. Formula for cross section

Substitute Eq. (12) for the initial wave function and Eq. (18) for the returning wave function into Eq. (11), the cross section contributed by the returning waves can be written as

$$\begin{aligned} \sigma^{ret}(E) &= -\frac{4e^2 E_p}{c} \text{Im} \sum_{jn} \int C \frac{\exp(-k_b r)}{r} \\ &\quad \times DN_{jn} e^{-ikr \cos \theta_j^{ret}} r^2 dr \sin \theta d\theta d\phi, \end{aligned} \quad (20)$$

where $D = r \cos \theta$ for a laser linearly polarized in the field direction. Expanding the incoming wave as partial waves and using the integral formula $\int j_1(kr) e^{-k_b r} r^2 dr = 2k/(k_b^2 + k^2)^2$, Eq. (20) becomes

$$\sigma^{ret}(E) = -\frac{8\pi^2 k C^2}{c(E_b + E)^3} \sum_{jn} \frac{1}{T_{jn}} \sin \left(S_{jn} - \mu_{jn} \frac{\pi}{2} \right). \quad (21)$$

Finally, adding the field-free cross section in Eq. (10) to the cross section contributed by the returning waves in Eq. (21), the total photodetachment cross section is

$$\begin{aligned} \sigma(E) &= \frac{16\sqrt{2} C^2 \pi^2}{3c} \frac{E^{3/2}}{(E_b + E)^3} \\ &\quad - \frac{8\pi^2 k C^2}{c(E_b + E)^3} \sum_{j=1}^4 \sum_{n=0}^{\infty} \frac{1}{T_{jn}} \sin \left(S_{jn} - \mu_{jn} \frac{\pi}{2} \right), \end{aligned} \quad (22)$$

where T_{jn} , S_{jn} and μ_{jn} are given, for the case $\alpha \neq 0$, in Eqs. (5), (6), and (7), respectively.

FIG. 1. The dependence of the photodetachment cross section on the gradient of the electric field, α . The constant term of the field is fixed at $F_0 = 500$ Kv/cm and the gradient is (a) $\alpha = 0$ a.u., (b) $\alpha = 1 \times 10^{-7}$ a.u., and (c) $\alpha = 1 \times 10^{-6}$ a.u.

IV. NUMERICAL RESULTS

Using Eq. (22), the detachment cross section can be calculated for different values of background field F_0 and field gradient α . The results are shown in Figs. 1 and 2.

In Fig. 1, F_0 is fixed at 500 kV/cm and α varies. When $\alpha \rightarrow 0$, the present system should recover the case for a homogeneous electric field. Taking $\alpha = 10^{-12}$ in Eq. (22), the calculated spectrum is shown in Fig. 1(a). [One should not set α exactly equal to 0 in Eq. (22) since the equation is for $\alpha \neq 0$.] The spectrum is simply a superposition of a background and a sinusoidal oscillation. Actually, this spectrum is almost exactly the same as the one given by the following previously known formula for the detachment cross section of H^- in a homogeneous electric field (i.e., α equal to zero exactly) [8]

$$\sigma(E) = \frac{16\sqrt{2}C^2\pi^2}{3c} \frac{E^{3/2}}{(E_b + E)^3} + \frac{4\pi^2 C^2 F_0}{c(E + E_b)^3} \cos\left(\frac{4\sqrt{2}E^{3/2}}{3F_0}\right), \quad \text{for } \alpha = 0. \quad (23)$$

We note that this formula can be obtained by counting the contribution of the only one closed orbit, the up orbit. When $\alpha \neq 0$, the electron moving downward also returns and there

FIG. 2. The dependence of the photodetachment cross section on the constant term of the electric field F_0 . The gradient of the field is fixed at $\alpha = 1 \times 10^{-6}$ a.u. The constant term of the field is (a) $F_0 = 0$ Kv/cm, (b) $F_0 = 100$ Kv/cm, and (c) $F_0 = 250$ Kv/cm.

are three more closed orbits. They also contribute to the cross section and make the sawtooth spectrum, as shown in Fig. 1(b). Note that the amplitudes of the saw teeth are smaller than that of the smooth oscillation. Increasing α further, as shown in Fig. 1(c), the contributions of the down orbits become comparable to that of the up orbit and the amplitudes of the saw teeth become larger.

The influence of F_0 on the photodetachment cross section is shown in Fig. 2 for a fixed value of α . When $F_0 = 0$, the up orbit and the down orbit are symmetric about the ρ axis, and so are the $j=2$ and the $j=4$ fundamental orbits. Because of the symmetries, the double summation in Eq. (22) can be simplified to become a single summation, i.e.,

$$\sum_{j=1}^4 \sum_{n=0}^{\infty} \frac{1}{T_{jn}} \sin\left(S_{jn} - \mu_{jn} \frac{\pi}{2}\right) = \frac{2\sqrt{\alpha}}{\pi} \sum_{m=1}^{\infty} \frac{1}{m} \sin\left(m \frac{\pi}{\sqrt{\alpha}} E - \frac{m}{2} \pi\right), \quad \text{for } F_0 = 0. \quad (24)$$

This is the Fourier series of a sawtooth wave function. The cross section for $F_0 = 0$ is shown in Fig. 2(a) where the sawtooth structure can be seen clearly. Increasing F_0 , the up and the down orbits are no longer symmetric about the ρ axis, nor are the $j=2$ and the $j=4$ fundamental orbits. The double

summation in Eq. (22) cannot be expressed by a single summation as in the case for $F_0=0$. As a result of this complexity, the cross section is an irregular sawtooth curve, as shown in Fig. 2(b). The sawtooth structure becomes more irregular when F_0 is increased further, as shown in Fig. 2(c).

V. CONCLUSIONS

We derive an analytical formula for the photodetachment cross section of H^- in a gradient electric field in the framework of the closed orbit theory. The resulting spectrum is an irregular sawtooth curve in general, which is very different from that for a uniform electric field (the latter is a regular smooth oscillating curve).

The results have some possible applications. For example, the results may be used in determining the ion position in an ion trap experiment. Once a detachment spectrum would be measured after applying a laser on the trapped ion, one may compare the spectrum with the spectra calculated by using the formula derived in this article to determine the “effective” background electric field F_0 . From this effective F_0 , the position of the ion can be obtained since a move of the ion is equivalent to a change in F_0 .

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