Closed-orbit theory for photodetachment of H^- in a static electric field

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Standard closed-orbit theory is applied to derive the photodetachment cross section of H^- in the presence of a static electric field. The result agrees with the one derived earlier using a quantum approach involving a momentum-space wave function and stationary-phase approximation. The advantage of the present derivation is the ability to separate the oscillation term and the smooth background term in the photodetachment cross section and to identify the two terms with different physical origins.

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More than a decade after Bryant and co-workers [1,2] observed the "ripple" structure in the photodetachment cross section of H^- in the presence of a static electric field of a few hundred kV/cm, in contrast with the smooth photodetachment cross section in the absence of an electric field, photodetachment of negative ions in a static electric field continues to attract theoretical and experimental attentions [3–8].

Rau and Wong provided a quantitative theory [9] for the observed ripple structure. They derived and expressed the photodetachment cross section in an electric field in terms of an integral involving Airy function. "Frame-transformation theory" was used in their derivation. The ripple structure was explained as an interference between a detached electron going "up hill" and "down hill" by Rau and Wong [9]. At about the same time, Du and Delos [10] presented a formula derived using a quantum approach involving a momentum-space wave function and stationary-phase approximation. By applying an asymptotic method, they were able to write the photodetachment cross section in an electric field as a sum of a smooth background term plus an oscillation term. This form is consistent with the general result of closed-orbit theory [11,12], the ripple structure was therefore interpreted as arising from the interference between the detached electron going out from the nucleus and the electron wave returning to the nucleus.

Closed-orbit theory not only provides a clear physical picture for the oscillations in the photodetachment or photoionization cross sections, it is also a quantitative tool being used to calculate and to analyze very complicated oscillations in the spectra for atoms in external fields [13]. It is therefore surprising to know that closed-orbit theory has not yet been applied to study the photodetachment cross section of H^- in the presence of a static electric field. This system is perhaps the simplest for closed-orbit theory because there is only one closed orbit. It is the purpose of this paper to fill in this existing gap. It will be shown that the result from closed-orbit theory is the same as the one derived earlier [10] using a quantum approach involving a momentum-space wave function and stationary-phase approximation. Furthermore, by going through the closed-orbit theory derivation, we are able to separate the oscillation term and the smooth background term and to identify each term with its physical origins. Atomic units will be used unless otherwise noted.

Assuming that the static electric field and the photon polarization are in the z direction. The photodetached electron wave function ψ_d satisfies the Schrödinger equation with a source term [14],

$$(E-H)\psi_d = z\psi_i, \tag{1}$$

where E is the energy of detached electron and ψ_i is the initial wave function of H^-. In the present study we follow Ref. [10] and take the one-electron approximation. The initial wave function in configuration space is given by $\psi_i(\boldsymbol{q}) = B(e^{-k_b r}/r)$, B is a "normalization" constant and is equal to 0.315 52, and k_b has a numerical value 0.235 $\underline{588}$ 3 and is related to the binding energy E_b of H^- by $k_b=\sqrt{2E_b}$. H is the Hamiltonian governing the motion of the detached electron in the combined atomic potential $V_p(r)$ and the static electric field; it can be written as $H=\boldsymbol{p}^2/2+V_p(r)+Fz$. Because the initial state is an S state, the detached electron carries one angular momentum right after being detached near the nucleus; it is a good approximation to neglect $V_p(r)$ here.

The physical solution of Eq. (1) requires that only an outgoing wave be present at large r. Once we have the detached electron wave function $\psi_d(\boldsymbol{q})$ satisfying the correct outgoing boundary condition, the oscillator-strength density can be calculated by using the formula [12]

$$Df(E,F) = -\frac{2(E_f-E_i)}{\pi}\mathrm{Im}\langle z\psi_i|\psi_d\rangle. \tag{2}$$

The oscillator-strength density is proportional to the photodetachment cross section.

We now construct the solution of Eq. (1) near the nucleus using closed-orbit theory [12]. First, the wave function ψ_d is separated into a direct part and a returning part, $\psi_d=(\psi_d)_{\mathrm{dir}}+(\psi_d)_{\mathrm{ret}}$. The direct part represents the detached electron wave initially going out from the nucleus after photodetachment and it satisfies the equation

$$\left(E-\frac{\boldsymbol{p}^2}{2}\right)(\psi_d)_{\mathrm{dir}} = z\psi_i, \tag{3}$$

which is obtained from Eq. (1) after dropping the static electric field term. The outgoing solution is [14]

$$(\psi_d)_{\rm dir}(q) = -\frac{4Bk^2 i}{(k_b^2+k^2)^2} h_1^{(1)}(kr)\cos(\theta), \tag{4}$$

where $k=\sqrt{2E}$ is the momentum of the detached electron and $h_1^{(1)}(kr)$ is the outgoing spherical Bessel function [15]. The overlap integral of the direct part with the source gives the smooth background [16]

$$Df_0 = -\frac{2(E_f-E_i)}{\pi}\mathrm{Im}\langle z\psi_i|(\psi_d)_{\rm dir}\rangle = \frac{8\sqrt{2}B^2E^{3/2}}{3(E_b+E)^3}. \tag{5}$$

If there is no static electric field, the detached electron will propagate away from the "source" region near the nucleus as a spherical wave and never return. The smooth background term will be the full cross section in this case. When there is a static electric field, most of the outgoing waves will not return except a pencil of waves propagating along the only closed orbit of the system. This wave, initially traveling in the z direction, is slowed down first by the electric field. Its propagation direction is then turned to the negative z direction, it is accelerated by the static electric field, and it eventually passes through the detached electron source region, where it interferes with the detached electron source (Fig. 4 of Ref. [10]). The phase difference between the returning wave and the direct wave $(\psi_d)_{\rm dir}$ near the nucleus determines whether the interference enhances or inhibits the total production of detached electrons. The returning wave function $(\psi_d)_{\rm ret}$ near the nucleus represents the electron wave coming back to the nucleus after traveling along the closed orbit. The phase and amplitude of this returning wave can be calculated by propagating and following the direct wave in Eq. (4) along the closed orbit until it comes back close to the nucleus. The general procedure is described elsewhere [12].

To obtain the returning wave function associated with the closed orbit, we draw a sphere of radius R large enough so that the asymptotic approximation $h_1^{(1)}(kr)=e^{i(kr-\pi)}/kr$ is valid. It also must be small enough so that the electric field potential term is much smaller than the initial kinetic energy term of the detached electron inside the sphere—that is, $zF \ll k^2/2$. The direct outgoing wave on the surface of this sphere is then

$$(\psi_d)_{\rm dir} = -i\frac{4Bk^2}{(k_b^2+k^2)^2}\cos(\theta)\frac{e^{i(kr-\pi)}}{kr}. \tag{6}$$

The phase and amplitude changes as it propagates out from the surface and along the closed orbit. When it comes back to a region (a few atomic units in size) near the nucleus, the returning wave can be approximated by a plane wave traveling in the negative z direction:

$$(\psi_d)_{\rm ret}(\boldsymbol{q}) = g e^{-ikz}, \tag{7}$$

where g is calculated according to the general method [12] as a product of initial outgoing wave in the z direction, an amplitude A, and a phase factor:

$$g = Ae^{i(S-\pi/2)}(\psi_d)_{\rm dir}(\theta=0,R), \tag{8}$$

where S is a phase integral $\int p dq$ along the closed orbit from the surface out and back to the origin $q=0$, $\pi/2$ is the phase correction at the turning point of the closed orbit, and A is an amplitude, which counts for the spreading of the wave as it propagates along the closed orbit and can be calculated by considering neighboring trajectories of the closed orbit. We have previously derived a formula for A applicable in this cylindric symmetric situation [14]:

$$A = \sqrt{\frac{R^2k}{(R+kt)^2|k-ft\cos(\theta_i)|}}, \tag{9}$$

where t is the time going from the surface out and back to the origin and θ_i is the outgoing direction of the closed orbit and equals zero here. In evaluating the expression for g in Eq. (8), we note that the result is independent of R as it must be. The result is

$$g = \frac{2BFi}{k(k_b^2+k^2)^2}e^{i(S_{\rm co}-\pi/2)}, \tag{10}$$

where $S_{\rm co}=4\sqrt{2}E^{3/2}/3F$ is the action integral around the closed orbit.

The overlap integral of the returning wave in Eq. (7) with the source gives the oscillation in the oscillator-strength density:

$$Df_1 = -\frac{2(E_f-E_i)}{\pi}\mathrm{Im}\langle z\psi_i|(\psi_d)_{\rm ret}\rangle = \frac{2FB^2}{(E_b+E)^3}\cos(S_{\rm co}). \tag{11}$$

Combining Eqs. (5) and (11) we obtained the total oscillator-strength density in an electric field:

$$Df(E,F) = \frac{8\sqrt{2}B^2E^{3/2}}{3(E_b+E)^3} + \frac{2FB^2}{(E_b+E)^3}\cos(S_{\rm co}). \tag{12}$$

Following Eq. (1) of Ref. [10], the photodetachment cross section is $\sigma(E,F)=(2\pi^2/c)Df(E,f)$. When the numerical values [17] $c=137.037$ and $B=0.315\,52$ are used, the result is exactly the same as Eq. (30) of Ref. [10] derived earlier using a quantum approach involving a momentum-space wave function and stationary-phase approximation.

The present derivation based on standard closed-orbit theory clearly separates the smooth background term and the oscillation term. We can identify the two terms with different physical origins. The smooth background term represents the intensity of the initial outgoing detached electron, and the oscillation term is the signature of the interference between the returning wave propagating along the only closed orbit in this system and the initial outgoing detached electron wave. The oscillation has a period $2\pi/T$ on the energy scale, where T is the classical closed-orbit time. The amplitude of the oscillation measures the wave spreading as it propagates along the closed orbit. Oscillations in the spectra are best analyzed by scaled energy spectroscopy [13].

[1] H. C. Bryant *et al.*, Phys. Rev. Lett. **58**, 2412 (1987).
[2] J. E. Stewart *et al.*, Phys. Rev. A **38**, 5628 (1988); H. C. Bryant, J. Chin. Chem. Soc. (Taipei) **48**, 333 (2001).
[3] N. D. Gibson, B. J. Davis, and D. J. Larson, Phys. Rev. A **47**, 1946 (1993).
[4] N. D. Gibson, B. J. Davis, and D. J. Larson, Phys. Rev. A **48**, 310 (1993).
[5] C. Rangan and A. R. P. Rau, Phys. Rev. A **61**, 033405 (2000).
[6] N. D. Gibson, M. D. Gasda, K. A. Moore, D. A. Zawistowski, and C. W. Walter, Phys. Rev. A **64**, 061403(R) (2001).
[7] I. I. Fabrikant, M. V. Frolov, N. L. Manakov, and A. F. Starace, Phys. Rev. A **64**, 037401 (2001).
[8] A. R. P. Rau and C. Rangan, Phys. Rev. A **64**, 037402 (2001).
[9] A. R. P. Rau and H. Wong, Phys. Rev. A **37**, 632 (1988).
[10] M. L. Du and J. B. Delos, Phys. Rev. A **38**, 5609 (1988).
[11] M. L. Du and J. B. Delos, Phys. Rev. Lett. **58**, 1731 (1987).
[12] M. L. Du and J. B. Delos, Phys. Rev. A **38**, 1896 (1988); **38**, 1913 (1988).
[13] A. Matzkin, P. A. Dando, and T. S. Monteiro, Phys. Rev. A **66**, 013410 (2002); D. Kleppner and J. B. Delos, Found. Phys. **31**, 593 (2001) and references therein; B. E. Granger and C. H. Greene, Phys. Rev. A **62**, 012511 (2000), and references therein.
[14] M. L. Du, Phys. Rev. A **40**, 4983 (1989).
[15] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1987).
[16] We have used the integral $\int_0^\infty j_1(kr)e^{-k_b r}r^2 dr = 2k/(k_b^2+k^2)^2$.
[17] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Plenum, New York, 1977).