Oscillations of electron flux in photodetachment of H^- in an electric field

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Outgoing electron waves produced in the photodetachment of H^- are propagated to large distances in a homogeneous electric field, and the electron flux is then computed. It is shown that the interference of waves propagating along two distinctive paths from the region of the bound state of H^- to the same point induces oscillations in the electron flux distribution. Simple analytic expressions are presented for the flux distribution.

I. INTRODUCTION

Experiments[1] and theories[1–5] have now shown that the total cross section of photodetachment of H^- in the presence of a homogeneous electric field is oscillatory when the polarization of photons is parallel to the applied electric field. The oscillations can be attributed[5] to the interference of electron waves reflected by the potential barrier of the electric field and the source of the waves localized in the regions of the bound state of H^-, similar to the oscillations of the absorption spectra of atoms in a magnetic field.[6]

The photodetachment cross section is proportional to the integrated outgoing electron flux across a large enclosure in which the bound H^- sits. Therefore it is clear that the oscillations in the total cross section reflect the oscillations in the microscopic electron flux distribution, whose quantitative description is the subject of the present paper.

In an earlier treatment of a similar problem, Fabrikant[2] used the exact known time-dependent propagator in a uniform field to obtain the formula for the electron flux distribution, ignoring the effects of the polarized field of the atom on the electron. However, since the propagator in a combined central field and a uniform field is not known, at present it is not clear how to apply this approach to the general case.

Our alternative description, following the physical picture in Ref. 5, takes the polarized central field into account from the start. When a laser is applied to a negative ion, like H^-, in an electric field, the ion may absorb a photon. When it does, the electron goes into an outgoing wave of the polarized field. This wave then propagates away from the atom. Sufficiently far from the atom, the wave propagates according to semiclassical mechanics, and it is correlated with classical trajectories. The wave fronts are perpendicular to the trajectories, and the waves propagate along the trajectories. Eventually the waves initially propagating in the electric field direction are turned back by the electric field, which then propagate to large distances down field to interfere with the waves initially propagating in that direction, giving rise to a two-term interference pattern in the electron flux distribution.

To solve the problem, we draw a sphere of radius R satisfying certain conditions, which will be specified later. Inside the sphere, the electric field is small and can be neglected. The outgoing electron wave in the polarized central field is then computed. On the surface of the sphere, asymptotic forms of the outgoing wave are used. The asymptotic form of the outgoing wave is propagated out semiclassically in the combined polarized field and the electric field until far away from the atom where the electron flux is evaluated. The final result is shown to be independent of the joining radius R, which can be taken to be zero in the calculations.

The paper is organized as follows. In Sec. II we describe the details of the initial outgoing electron wave, its semiclassical propagation, and the formulas for the electron flux distribution. In Sec. III we discuss the characteristics of the derived electron flux distribution and its possible experimental observation.

II. THEORY

The H^- will be considered as effectively a one-electron system. Let the initial bound state of H^- be $\phi_i(r)$, the binding energy of the electron E_b, and the photon energy $E_{ph}=E+E_b$; the steady outgoing electron wave produced in the photodetachment in the presence of a homogeneous electric field pointing in the z direction satisfies the inhomogeneous Schrödinger equation,

$$(E-H)\psi_F^{\dagger}=D\phi_i \ , \tag{1a}$$

where D is the dipole operator, z or $(x+iy)/\sqrt{2}$, for linear or circular polarized photons; the Hamiltonian

$$H=-\tfrac{1}{2}\nabla^2+V_p(r)+Fz \ , \tag{1b}$$

where $V_p(r)$ is the polarized central field of the H atom. The physical solution $\psi_F^{\dagger}$ is required to be purely outgoing at large distances.

Imagine a surface Γ (for example, the surface of a sphere) enclosing the source region, a generalized differential cross section $d\sigma(\mathbf{q})/ds$ may be defined on the surface from the electron flux crossing the surface,

$$\frac{d\sigma(\mathbf{q})}{ds}=\frac{2\pi E_{ph}}{c}\mathbf{j}\cdot\mathbf{n} \tag{2a}$$

and

$$\mathbf{j}=\frac{i}{2}(\psi_F^{\dagger}\cdot\nabla_F\psi^{\dagger *}-\psi_F^{\dagger *}\nabla\psi_F^{\dagger}) , \tag{2b}$$

where $\mathbf{q}$ is the coordinate on the surface Γ, $\mathbf{n}$ is the exterior norm vector at $\mathbf{q}$, and ds is the differential area on the surface. The total cross section may then be obtained by integrating the differential cross section over the surface

$$\sigma(E,F)=\int_{\Gamma}\frac{d\sigma(\mathbf{q})}{ds}ds . \tag{2c}$$

It can be shown that the total cross section defined in (2c) is equivalent to the conventional definition involving a summation over the dipole matrix elements[5] with the help of the spectral representation of the Green's function[7] and the Gauss theorem.

A. The outgoing wave in the absence of the electric field

In the vanishing limit of the electric field F the outgoing wave $\psi_{F=0}^{\dagger}(\mathbf{r})$ satisfies

$$[E+\tfrac{1}{2}\nabla^2-V_p(r)]\psi_{F=0}^{\dagger}(\mathbf{r})=D\phi_i . \tag{3}$$

Since ϕ_i is an S state for H^-, it is readily shown that for r outside the source region,[8]

$$\psi_{F=0}^{\dagger}(\mathbf{r})=\left[\sum_{m=-1}^{1}C_m(k)Y_{1m}(\theta,\varphi)\right]h_1^{(1)}(kr) \tag{4a}$$

with

$$C_m(k)\equiv -2ki\int j_1(kr)Y_{1m}^{*}(\theta,\varphi)D(\mathbf{r})\phi_i(r)dV , \tag{4b}$$

where in Eq. (4), $j_1(kr)$ and $h_1^{(1)}(kr)$ are the regular and outgoing spherical Bessel functions.[9] In obtaining (4a) and (4b), the small phase shift of the p wave caused by $V_p(r)$ is neglected. This is a good approximation for H^-. In general, however, the regular and outgoing Bessel functions should be replaced by the corresponding regular and outgoing functions in the central field $V_p(r)$.

Using $\phi_i=B\exp(-k_b r)/r$ with $B=0.315\,52$ and $K_b=0.234\,488\,3$ as previously used,[5,10] and writing the dipole operator D in the form

$$D=\frac{a^+}{\sqrt{2}}(x+iy)+\frac{a^-}{\sqrt{2}}(x-iy)+a^0z$$
$$=r\left[\frac{a^+}{\sqrt{2}}(\sin\theta)e^{i\varphi}+\frac{a^-}{\sqrt{2}}(\sin\theta)e^{-i\varphi}+a^0(\cos\theta)\right] , \tag{5}$$

then one arrives at[11]

$$\psi_{F=0}^{\dagger}(\mathbf{r})=-\frac{4k^2Bi}{(k_b^2+k^2)^2}\times\left[\frac{a^+}{\sqrt{2}}(\sin\theta)e^{i\varphi}+\frac{a^-}{\sqrt{2}}(\sin\theta)e^{-i\varphi}+a^0(\cos\theta)\right]h_1^{(1)}(kr) . \tag{6}$$

At large r, using the asymptotic form

$$h_1^{(1)}(kr)\sim\frac{1}{kr}[\exp i(kr-\pi)],\quad r\to\infty \tag{7}$$

Eq. (6) becomes

$$\psi_{F=0}^{\dagger}(r,\theta,\varphi)=U(k,\theta,\varphi)\frac{\exp(ikr)}{kr} , \tag{8a}$$

where

$$U(k,\theta,\varphi)\frac{4k^2Bi}{(k_b^2+k^2)^2}\left[\frac{a^+}{\sqrt{2}}(\sin\theta)e^{i\varphi}+\frac{a^-}{\sqrt{2}}(\sin\theta)e^{-i\varphi}+a^0(\cos\theta)\right] . \tag{8b}$$

$\psi_{F=0}^{\dagger}$ in Eqs. (8) represents the outgoing electron wave produced in the detachment of H^- in the absence of the electric field. Choosing the surface Γ as the surface of a sphere centered at the origin and using $\psi_{F=0}^{\dagger}$ in Eqs. (2), the correct zero-field differential and total cross section may be obtained, which of course agree with the results computed directly from the dipole matrix elements.[5,10]

B. The outgoing wave in the presence of the electric field

In the presence of the electric field, the asymptotic form of the zero-field outgoing wave in (8) still represents the outgoing wave in the region

$$\frac{1}{k}\ll r\ll\frac{k^2}{2F} . \tag{9}$$

Such a region exists provided the photon energy and the electric field strength F satisfies $1/k\ll k^2/2F$, which excludes the photon energy very close to the threshold.

As the electron wave in (8) propagates out to large r, the electric field becomes more and more important. Fortunately, the wave function away from the nuclei is semiclassical in nature and semiclassical methods may be used to propagate the wave function in (8) by computing the trajectories of an electron in an electric field.

Details of the semiclassical method have been described elsewhere.[12] Here we follow the procedure given in Ref. 6, which takes the cylindrical symmetry into consideration, thereby simplifying the formulas.

We first draw the surface of a sphere of radius R centered at the origin. R may take any value in the range of Eq. (9). The final result for the electron flux will be shown to be independent of the value of R.

Let the polar angle of the initial velocity perpendicular to the surface of the sphere be θ; then the trajectory going out from the sphere at time zero may be written in cylindrical coordinates as

$$\rho(t)=R(\sin\theta)+k(\sin\theta)t , \tag{10a}$$

$$z(t)=R(\cos\theta)+k(\cos\theta)t-Ft^2/2 . \tag{10b}$$

Figure 1 shows the trajectories described by (10) with varying θ. The whole space is divided into classically allowed and forbidden regions by the caustic surface given by

$$\rho_{\max}=\left[\frac{k^4}{F^2}-\frac{2kz}{F}\right]^{1/2} \tag{10c}$$

for $z<0$. It is easy to show that for any given point in the classically allowed region, (ρ,z,φ), there are two dis-

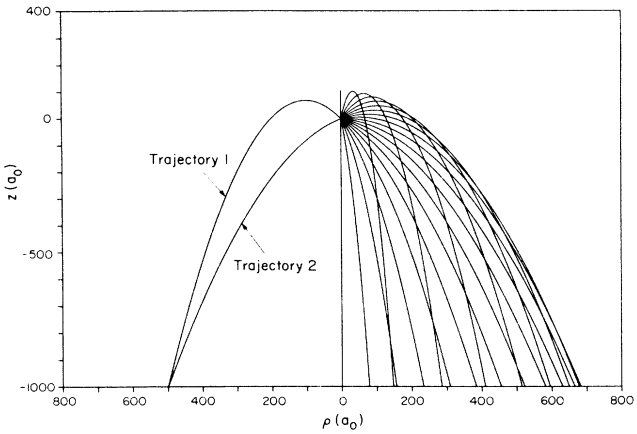

FIG. 1. Outgoing electron trajectories in the photodetachment of H^- in a homogeneous electric field F in the z direction. Trajectory 1 and 2 cross at the point $\rho=500.0a_0$, $z=-1000.0a_0$.

tinctive trajectories given by (10) with the initial angle θ_1 and θ_2 arriving at the point. In the limit of zero R, the two angles may be shown to satisfy

$$\cot\theta_1=\frac{k^2}{\rho F}\left[1+\left[1-\frac{F^2\rho^2}{k^4}-\frac{2Fz}{k^2}\right]^{1/2}\right], \tag{11a}$$

$$\cot\theta_2=\frac{k^2}{\rho F}\left[1-\left[1-\frac{F^2\rho^2}{k^4}-\frac{2Fz}{k^2}\right]^{1/2}\right]. \tag{11b}$$

Equations (11) are valid in the region $z<0$ and for $\rho\le\rho_{\max}$, θ_1 is the velocity angle of the trajectory that initially goes in the same direction of the electric field while θ_2 is the velocity angle of the trajectory that initially goes in the opposite direction of the electric field. In the limit of zero ρ, θ_1 and θ_2 approach 0 and π, respectively. Once the initial angles of the two trajectories are determined, the time duration along the trajectories t_1 and t_2 from the surface of the sphere to the same final point (ρ,z,φ) can be found from Eq. (10a).

The wave function at (ρ,z,φ) can be written as a sum of the contributions from the two trajectories,[6,12]

$$\psi_F^\dagger(\rho,z,\varphi)=\sum_{i=1}^{2}\psi_{F=0}^\dagger(R,\theta_i,\varphi)A_i(\rho,z,\varphi)\times\exp\left[i\left[S_i(\rho,z,\varphi)-\mu_i\frac{\pi}{2}\right]\right], \tag{12a}$$

where the amplitude

$$A_i(\rho,z,\varphi)=\left[\frac{R^2k}{(R+kt_i)^2|k-Ft_i\cos\theta_i|}\right]^{1/2}, \tag{12b}$$

and the phase accumulated along the trajectory

$$S_i(\rho,z,\varphi)=\int_0^t \mathbf{p}\,d\mathbf{q}=k^2t_i-k(\cos\theta_i)Ft_i^2+F^2t_i^3/3\;. \tag{12c}$$

μ_i is the Maslow index of the ith trajectory. In the present case, because the caustic is a fold, μ_i takes only two possible values, 0 or 1, corresponding to the positive or negative sign of $(k-Ft_i\cos\theta_i)$.

Now using $\psi_{F=0}^\dagger$ of Eq. (8) in Eq. (12) and the right inequality in Eq. (9), one can show that the different choice of R results in a negligible error in $\psi_F^\dagger$. The consistence of the formulation demands $\psi_F^\dagger$ to be independent of R. Because of this fact, R may be taken as zero.

C. Oscillations in electron flux

In the presence of a homogeneous electric field, there are two distinctive trajectories along which the electron waves produced in the photodetachment in the bound-state region of H^- propagate to arrive at the same point (ρ,z,φ) in the classically allowed region. The resulting wave function, given quantitatively in Eqs. (12), is therefore a sum of two terms. When inserting Eq. (12) in Eq. (2), the differential cross section is seen to exhibit a typical two-term interference pattern. The situation is very similar to the double-slit experiment discussed in the beginning of most quantum mechanics textbooks.[13]

Now consider a plane surface perpendicular to the electric field that intersects the z axis at $z(<0)$. The differential cross section defined on this plane may be readily evaluated from Eqs. (12) and (2) as

$$\frac{d^2\sigma(\rho,z,\varphi)}{\rho\,d\rho\,d\varphi}=-\frac{2\pi(E_b+E)}{c}\left[|\psi_1|^2v_{1z}+|\psi_2|^2v_{2z}+(v_{1z}+v_{2z})\mathrm{Re}(\psi_1\psi_2^*)\cos\left[S_1-S_2-\frac{\mu_1\pi}{2}+\frac{\mu_2\pi}{2}\right]\right], \tag{13a}$$

where

$$\psi_i=\frac{4k^{3/2}B}{(k_b^2+k^2)^2}\,\frac{[(a^+/\sqrt{2})\sin\theta_ie^{i\varphi}+(a^-/\sqrt{2})\sin\theta_ie^{-i\varphi}+a^0\cos\theta_i]}{(R+kt_i)|k-Ft_i\cos\theta_i|^{1/2}},\quad i=1,2\;. \tag{13b}$$

v_{1z} and v_{2z} are the z component of the electron velocities of trajectories 1 and 2, respectively, at (ρ,z,φ). Note that v_{1z} and v_{2z} are negative, ensuring that the differential cross section in (13) is positive.

III. RESULTS AND DISCUSSION

For $z=-1.0\times10^3a_0$, $F=500$ kV/cm, and $D=z$ $(a^+=a^-=0,\ a^0=1)$, we plotted in Fig. 2 the differential

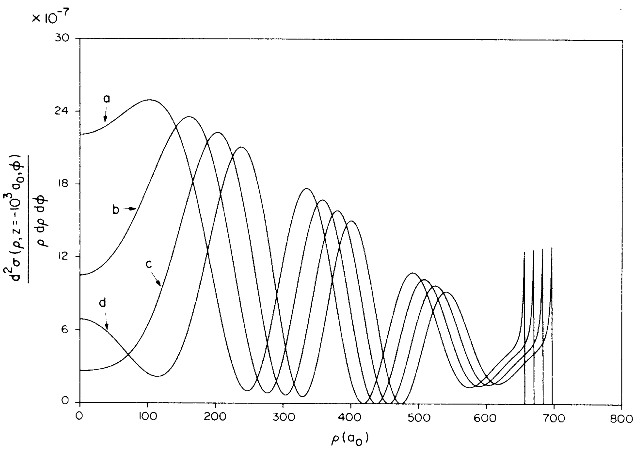

FIG. 2. Differential cross section of photodetachment of H^- defined on the (ρ,φ) plane at $z=-10^3 a_0$ and photon energies (a) $E_b+0.26$ eV, (b) $E_b+0.27$ eV, (c) $E_b+0.28$ eV, and (d) $E_b+0.29$ eV for linear polarization.

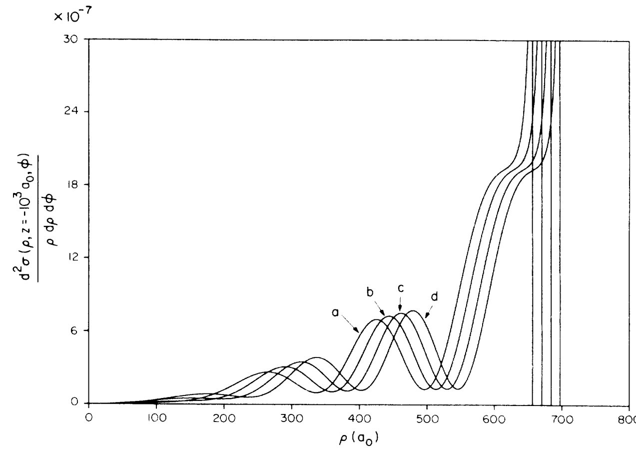

FIG. 3. Same as in Fig. 2 except the photons are circularly polarized $D=(x+iy)/\sqrt{2}$.

cross section in (13) as a function of distance from the z axis, ρ, for four different photon energies. There are three or four oscillations in the differential cross sections. As the photon energy is changed, the amplitude near the z axis ($\rho \sim 0$) varies. On the other hand, the oscillation patterns at large ρ remain similar in shape, but are shifted slightly for different photon energies. The electron flux at small ρ and large ρ may, therefore, be identified with the weak oscillations and the strong background in the total cross section.[2,5]

The semiclassical approximation for the electron flux in Eq. (13) breaks down at the right edge of Fig. 2. Very close to $\rho_{\max}$, the two trajectories approach each other and the quantity $|k-Ft_i\cos\theta_i|$ approaches zero. The problem is very similar to the WKB approximation of the eigenfunction in one dimension, and it can be fixed by using an Airy function, which is joined smoothly on to the valid semiclassical approximation at smaller ρ. As a test of accuracy, we have compared the numerically integrated differential cross section derived here with the total cross section derived earlier[5] using different methods, and found the difference is, in general, a few percent.

In Fig. 3 we plotted the differential cross section as in Fig. 2 except now $D=(x+iy)/\sqrt{2}$ ($a^+=1$, $a^-=a^0=0$). One observes that unlike Fig. 2 for the linear photon polarization, the electron flux for circular polarization near the z axis remains small at different photon energies, resulting in a nonoscillatory total cross section.[1,5]

It should be pointed out that the minima in the electron flux shown in Figs. 2 and 3, in general, are not zero. There are a few physical reasons for this fact. First, the amplitude of the initial radial outgoing electron wave is angular dependent; second, the waves propagating along the two paths spread by different amount; and third, the two final velocities are not the same. From Eq. (13) it is clear that the minima are not zero. However, as z goes to $-\infty$, the minima approach zero.

Earlier, in a general discussion of the photodetachment of a negative ion in a homogeneous electric field, Fabrikant[2] considered particularly the propagation of electrons from an isotropic point source and found formulas for the resulting two-term interference flux. Based on this result, Demkov. Kondratovich, and Ostrovskii[14] subsequently pointed out that the scale of the interference pattern can be large, and under favorable conditions it could be observed in a direct experiment. Such observation might provide more accurate information about the negative ion.

The direct consequences of the above simplified but unrealistic assumptions about the electron source are that the minima in the electron flux are always zero and that the height of the peak at larger ρ is higher, both of which are inconsistent with our findings. By using the exactly known time-dependent propagator in an electric field, Fabrikant[2] was able to remove some of the simplified assumptions. The interference electron flux formulas obtained then involve quantities defined as three-dimensional integrals. It is not known at present how to extend the time-dependent Green's function approach to the case where the polarized field is strong and cannot be neglected, since the analytic form of the time-dependent propagator in a combined central field plus an electric field is not known.

Our present approach takes the final interaction into account in a straightforward way by dividing the whole space into two regions. In other applications, the effects of the long-range Coulomb field were included.[6]

In summary, we have analyzed the electron flux produced in the photodetachment of H^- in a homogeneous electric field. It has been shown that outgoing electron waves propagating along the distinctive trajectories from the region of the bound state of H^- to the same point interfere to produce oscillations in the electron flux distribution described quantitatively by Eq. (13). A direct observation of the large scale oscillations may be possible.

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