

Interference effects in photodetachment and photoionization of atoms in a homogeneous electric field

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An analysis is made of the processes of photoionization and photodetachment of atoms in a homogeneous electric field without allowance for the interaction between an electron and the atomic core in the final state. The quasiclassical and semiclassical approximations are used to obtain the expressions for the total cross section of a given process and for the angular distribution of electrons. It is shown that the oscillatory energy dependences of the cross sections are due to interference between classical trajectories of electron motion in a homogeneous electric field.

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1. INTRODUCTION

Experimental studies of the photoionization of rubidium atoms in a static electric field^{1,2} have revealed oscillations in the energy dependence of the photoionization cross section at photon energies both above and below the threshold in the absence of a static field. Interpretation of these results¹⁻⁴ and a quantum-mechanical calculation⁵ are in agreement with the hypothesis of the existence of quasidecrete levels in a field which is a superposition of the Coulomb and homogeneous fields. However, oscillations of the cross sections need not be associated with the Coulomb field and quasidecrete levels. Several theoretical investigations of the influence of electric fields on the absorption of light in solids (see, for example, Refs. 6 and 7) have yielded oscillatory dependences of the absorption coefficient on the frequency of light without allowance for the Coulomb interaction. In his book,⁸ Ansel'm explains these oscillations by interference between the reflected and incident waves.

We shall consider this effect in greater detail. We shall show that, irrespective of the interaction between an electron and the atomic core in the final state, the photo-process cross section exhibits oscillations resulting from the interference between classical trajectories of motion in a homogeneous field.

2. OSCILLATORY STRUCTURE OF THE TOTAL CROSS SECTIONS

We shall consider the photoprocess accompanied by a transition of an electron to a state of energy $E = \hbar^2 k^2 / 2\mu$, where $E = 0$ corresponds to the photoprocess threshold in zero static field. We shall consider only the case when $E > 0$, although photoionization or photodetachment in an external field are also possible when $E \leq 0$. According to the secular form of perturbation theory, the differential cross section of the photoprocess is (see, for example, Ref. 9)

$$d\sigma = \frac{8\pi^2 e^2 k}{c\omega \hbar^2 \mu} \left| \int \psi^*(\mathbf{r}) V(\mathbf{r}) \varphi_{\mathbf{k}}(\mathbf{r}) d\mathbf{r} \right|^2 d\Omega_{\mathbf{k}}, \quad (1)$$

where $V = -i\hbar \mathbf{e}_{\text{ph}} \nabla e^{-i\mathbf{x}\cdot\mathbf{r}}$ is the transition operator (\mathbf{e}_{ph} is the polarization vector of a linearly polarized photon and \mathbf{x} is its wave vector); $\omega = \kappa c$ is the photon frequency; e and μ are the charge and mass of an electron;

$\psi(\mathbf{r})$ is the wave function of the initial state; $\varphi_{\mathbf{k}}(\mathbf{r})$ is the wave function of the final state normalized to $\delta(\mathbf{k} - \mathbf{k}')$.

It should be noted that Eq. (1) does not give the complete angular distribution of photoelectrons, because the direction of motion of an electron escaping from an atom changes under the influence of the static field. The angular distribution will be derived in Sec. 4.

Our main approximation will be to neglect the interaction between an electron and an atom in the final state, which corresponds to the usual Born approximation in the absence of a static field. If the force \mathbf{F} exerted on an electron by a static homogeneous field is directed along the z axis, the wave function $\varphi_{\mathbf{k}}$ can be expressed in the form

$$\varphi_{\mathbf{k}}(r) = \left(\frac{\hbar^2}{2\mu F} \right)^{1/4} \frac{k_x^{3/4}}{2\pi^{3/4}} \exp[i(k_x x + k_y y)] \Phi(-\xi),$$

$$\xi = (z + \hbar^2 k_z^2 / 2\mu F) (2\mu F / \hbar^2)^{1/4},$$

where Φ is the Airy function defined as in Ref. 10.

The next approximation consists of the conditions

$$\beta = \frac{2}{3} \frac{\mu^2 v^3}{F \hbar} \gg 1, \quad v = \frac{\hbar k}{\mu},$$

which correspond to quasiclassical motion and are satisfied in moderate strong fields. It then follows from the asymptotic nature of Φ that

$$\varphi_{\mathbf{k}}(r) = \frac{1}{2\pi^{3/4}} \exp[i(k_x x + k_y y)] \sin \left(k_z z + \frac{\beta}{2} |\cos \theta_{\mathbf{k}}|^2 + \frac{\pi}{4} \right). \quad (2)$$

We obtain from Eq. (1)

$$d\sigma = \frac{e^2 k}{2\pi c \hbar^2 \mu \omega} \left[|M_{\mathbf{k}}|^2 + |M_{-\mathbf{k}}|^2 + 2 \text{Im}(M_{\mathbf{k}} M_{-\mathbf{k}}^* \exp(i\beta |\cos \theta_{\mathbf{k}}|^2)) \right] d\Omega_{\mathbf{k}}, \quad (3)$$

$$M_{\mathbf{k}} = \int \psi^*(\mathbf{r}) V(\mathbf{r}) e^{i\mathbf{k}\cdot\mathbf{r}} d\mathbf{r},$$

where \mathbf{k}' is a vector with components k_x , k_y , and $-k_z$.

We shall also assume that the photoprocess probability is much higher than the ionization probability (or the probability of dissociation of a negative ion) in a static field and that the influence of the static field on the initial state can be ignored. Then, the first two terms in Eq. (3) yield the photoprocess cross section in the absence of a field and the interference term gives rise to a correction due to the action of the field on the electron in the final state. We shall estimate the contribution of this correction to the total cross section by represent-

ing the initial-state function in the form

$$\psi(r) = R_l(r) Y_{lm}(\Omega), \quad (4)$$

where l and m are the orbital momentum and its projection for an electron in an atom.

When the photon polarization is parallel to the field (π polarization), the dipole approximation gives the following expression for M_k

$$\left. \begin{aligned} M_k &= -4\pi i^l \omega \mu [Y_{l-1, m}^*(\Omega_k) T_{l, l-1} d_{l, m} - Y_{l+1, m}^*(\Omega_k) T_{l, l+1} d_{l+1, m}], \\ M_k &= (-1)^{l+m} M_k; \\ T_{l, l'} &= \int r R_l(r) j_{l'}(kr) r^2 dr, \quad d_{l, m} = \left[\frac{(l+m)(l-m)}{(2l-1)(2l+1)} \right]^{1/2}, \end{aligned} \right\} (5)$$

where j_l is a spherical Bessel function.

If the photon polarization is perpendicular to the field (σ polarization), assumed to be specifically directed along the x axis, the corresponding expression becomes

$$\left. \begin{aligned} M_k &= -2\pi i^l \omega \mu \{ T_{l, l-1} [g_{l, -m} Y_{l-1, m-1}^*(\Omega_k) - g_{l, m} Y_{l-1, m+1}^*(\Omega_k)] \\ &+ T_{l, l+1} [g_{l+1, m+1} Y_{l+1, m-1}^*(\Omega_k) - g_{l+1, -m+1} Y_{l+1, m+1}^*(\Omega_k)] \}, \\ M_k &= (-1)^{l+m} M_k, \quad g_{l, m} = \left[\frac{(l+m)(l+m-1)}{(2l+1)(2l-1)} \right]^{1/2}. \end{aligned} \right\} (6)$$

We can similarly obtain M_k for any other polarization.

In view of the large value of β , the main contribution in the integration of the interference term of Eq. (3) with respect to θ_k is made by the stationary point $\theta_k = \pi/2$ and the boundary points $\theta_k = 0, \pi$. In the case of the σ polarization it follows from Eq. (5) that $|M_k|$ differs from zero for $\theta_k = \pi/2$ if $l+m$ is odd and it vanishes in accordance with the law $\cos^2 \theta_k$ if $l+m$ is even. Therefore, the contribution of σ which does not oscillate with respect to β is proportional to $\beta^{-1/3}$ or β^{-1} for odd and even values of $l+m$, respectively. For $\theta_k = 0$ or π , the value of $|M_k|^2$ varies in accordance with the law $(\sin \theta_k)^{2m}$. Therefore, the contribution to β which does oscillate in respect of β is proportional to $\cos \beta / \beta^{m+1}$ or $\sin \beta / \beta^{m+1}$ for even and odd m , respectively ($m \geq 0$).

A similar analysis in the case of the π polarization shows that the nonoscillatory contribution to σ is proportional to $\beta^{-1/3}$ or β^{-1} for even or odd $l+m$, respectively, and the oscillatory contribution to σ is proportional to $\sin \beta / \beta^{l+m-1}$ or $\cos \beta / \beta^{l+m-1}$ for even or odd m , respectively ($m \geq 0$).

We shall now give the final formulas for the π and σ polarizations in the $l = m = 0$ case:

$$\sigma_\pi = \sigma_0 \left[1 + \frac{\cos \beta - 1}{\beta} + O(\beta^{-2}) \right], \quad (7)$$

$$\sigma_\sigma = \sigma_0 \left[1 + \frac{\Gamma(1/2)}{4\beta^{1/2}} - \frac{\sin \beta}{\beta^2} + O(\beta^{-3}) \right], \quad (8)$$

where

$$\sigma_0 = 8\pi e^2 \omega \mu k T_0 / 3ch^2.$$

It follows from Eq. (7) that the positions of the oscillation peaks are given by

$$E_n = (0.06663Fn)^{2/3}, \quad n=0, 1, \dots,$$

where F is in volts per centimeter and the energy in reciprocal centimeters. Similarly, the values of E_n' at which $\cos \beta$ passes through its average value are given

by

$$E_n' = [0.03332F(n+1/2)]^{2/3}.$$

The separations between the oscillations peaks $(\Delta E)_n$ and the widths of the peaks Γ_n can be described by

$$\begin{aligned} (\Delta E)_n &= E_n - E_{n-1}, \\ \Gamma_n &= E_{2n}' - E_{2n-1}', \quad n=1, 2, \dots \end{aligned}$$

We shall now give the distances between the oscillation peaks $(\Delta E)_n$ in reciprocal centimeters (for $n = 1, 2$, and 3 , respectively) and compare them with the experimental results (the last value) in two fields:

$F=1016$ V/cm:	16.6	9.8	8.2	7
$F=6146$ V/cm:	55.2	32.4	27.2	28

The widths of the oscillation peaks Γ_n in reciprocal centimeters (for $n = 1, 2$, and 3 , respectively) compared with the experimental results (last column) are as follows:

$F=1016$ V/cm:	5.6	4.4	3.8	4
$F=6146$ V/cm:	18.5	14.6	12.8	16

We can see that the theory is valid beginning from the second maximum (i.e., for $\beta \geq 2\pi$). Since the experimental data are available only for first three oscillations, it is not possible to study the reduction in $(\Delta E)_n$ and Γ_n on increase in E . The absence of oscillations of the experimental cross sections in the σ polarization case can be explained, on the basis of Eq. (8) by the fact that the oscillatory component is the second order of smallness in respect of β^{-1} and cannot be detected experimentally.

3. SEMICLASSICAL APPROACH

We shall elucidate the physical meaning of the results obtained by considering the following classical problem. We shall assume that a source of electrons of intensity I is located at the origin of a coordinate system. For simplicity, we shall postulate that the emission of electrons is isotropic. We shall consider a plane P perpendicular to the z axis located at a distance z from the source. In terms of cylindrical coordinates the z component of the current density in this plane P is

$$j_z = \frac{I}{2^l \pi^{l/2} t(r)} \left\{ \frac{1+2z/l+t(r)}{[1+z/l+t(r)]^{3/2}} + \frac{1+2z/l-t(r)}{[1+z/l-t(r)]^{3/2}} \right\}, \quad (9)$$

$$t(r) = 1+2z/l - \rho^2/l^2, \quad l = \mu v^2/F,$$

where v is the initial electron velocity.

Two curves in Eq. (9) represent the contributions of two trajectories to the current density at the point (z, ρ, φ) . These trajectories correspond to different angles of flight of an electron, θ_1 and θ_2 , relative to the z axis, where

$$\operatorname{ctg} \theta_{1,2} = l[1 \pm t(r)]/\rho.$$

The influence of the interference terms resulting on transition to a quantum-mechanical description can be estimated from semiclassical considerations in the same way as was done by Ford and Wheeler¹¹ in the case of collisions between heavy particles.

The wave function in the plane P can be represented in the form

$$f = \varphi_1 e^{iS_1/\hbar} + \varphi_2 e^{iS_2/\hbar}, \quad (10)$$

where S_i is the reduced action corresponding to the motion along the j -th trajectory and φ_i is a slowly varying real function. If we assume that S_i is large compared with \hbar , the current density becomes

$$j_z = \frac{1}{\mu} \left[\varphi_1^2 \frac{dS_1}{dz} + \varphi_2^2 \frac{dS_2}{dz} + \varphi_1 \varphi_2 \left(\frac{dS_1}{dz} + \frac{dS_2}{dz} \right) \cos \frac{S_1 - S_2}{\hbar} \right]. \quad (11)$$

Calculating S_i for the motion of a particle in a homogeneous field, we obtain

$$S_{1,2} = \frac{2^{3/2}}{3} \mu v l [1 + z/l \pm t(r)]^{3/2} [2 + 2z/l \mp t(r)]. \quad (12)$$

Comparing Eqs. (11) and (12) with Eq. (9), we find that the interference contribution to the current density in the classically allowed region is

$$j_{\text{int}} = \frac{I}{2^{3/2} \pi l (z^2 + \rho^2)^{3/2} t(r)} \left\{ \frac{1 + 2z/l + t(r)}{[1 + z/l + t(r)]^{3/2}} + \frac{1 + 2z/l - t(r)}{[1 + z/l - t(r)]^{3/2}} \right\} \cos \frac{S_1 - S_2}{\hbar}. \quad (13)$$

Here, j_{int} represents the interference pattern in the plane P and is an oscillatory function of the variables z , ρ , and v .

We shall now consider how the interference part of the total current depends on v :

$$I_{\text{int}} = 2\pi \int_0^{l(1+2z/l)^{1/2}} j_{\text{int}}(\rho) \rho d\rho + I' = 2\pi l^2 \int_0^{(1+2z/l)^{1/2}} j_{\text{int}}(t) t dt + I', \quad (14)$$

where I' is the contribution of the classically forbidden region. In view of the quasiclassical nature of the motion, the integral can be found by the stationary phase method. Then, $\Delta S = S_1 - S_2$ considered as a function t has just one stationary point $t = 0$. However, $\Delta S(0) = 0$ so that the point $t = 0$ makes no oscillatory contribution to I . In the classically forbidden region, the value of j_{int} decays exponentially and the main contribution to I' is again given by the point $t = 0$. In the next approximation, we have to allow for the boundary point $t = (1 + 2z/l)^{1/2}$, which corresponds to $\rho = 0$. Since for $\rho = 0$, we have

$$\Delta S = \frac{2}{3} \frac{\mu^2 v^2}{F \hbar} = \beta,$$

integration in the vicinity of the point $\rho = 0$ gives

$$I_{\text{osc}} = I \sin \beta / 3\beta. \quad (15)$$

The oscillatory contribution to I is of the same form as in Eq. (7). The difference in the numerical coefficient is due to lack of allowance for the anisotropy in the angular distribution of electrons emitted from the source. It should also be noted that, for the same reason, the phase of the oscillatory factor acquires an additional term π and the inaccuracy of the semiclassical analysis gives an extra phase $\pi/2$. This follows from the results of a quantum-mechanical treatment (see the end of Sec. 4). The extra phase $\pi/2$ appears because of inaccuracy of the calculation of the quasiclassical wave function. In the derivation of Eq. (10) we have assumed that the functions $\varphi_{1,2}$ are real, whereas in fact they should be multiplied by phase factors deduced from the boundary conditions for quasiclassical solutions of the Schrödinger equation.

We thus find that, in contrast to the case considered by Ford and Wheeler, the oscillatory (in respect of the energy) contribution to the total energy is not zero and

is found by integration near $\rho = 0$. This is due to the fact that the total probability of electron emission should depend not on the whole interference pattern in the P plane, but only on the interference inside the source, which corresponds to the trajectory with $\rho = 0$. This becomes even more obvious when the one-dimensional problem is considered. Interference between the trajectories directed along and against the field yields the following expression for the current:

$$I = I_0 (1 + \cos \beta), \quad (16)$$

where I_0 is the current corresponding to $F = 0$. It is clear from Eq. (16) that interference in the one-dimensional case is much more effective.

The trajectories which are at the limit of the classically allowed region interfere at any distance (no matter how close to the origin). Their contribution is represented by the nonoscillatory terms in Eqs. (7) and (8).

The conclusions reached above allow us also to interpret the dependence of the oscillatory contribution made by the polarization to the cross section. Favorable conditions for interference, resulting in oscillations, occur when photoelectrons escape from an atom mainly along and against the field. According to Eqs. (5) and (6), this is true only for $m = 0$ in the case of the π polarization and for $m = \pm 1$ in the case of the σ polarization.

4. ANGULAR DISTRIBUTION OF PHOTOELECTRONS

We shall justify the semiclassical approach and find the angular distribution of photoelectrons by considering a stationary variant of perturbation theory. The stationary part of a photoelectron function satisfies a system of coupled equations which can be decoupled, in the first order of perturbation theory in respect of the electromagnetic interaction, so that the wave function of the final state is described by

$$\left(\frac{\hbar^2}{2\mu} \Delta + E + Fz \right) f = - \frac{e}{\mu c} \left(\frac{2\pi c^2 n_s \hbar}{\omega \tau} \right)^{1/2} V \psi, \quad (17)$$

where n_s is the number of photons with a given polarization and wave vector κ ; τ is the normalization volume; V is the transition operator introduced above.

After calculation of the current density in the plane P introduced in Sec. 3, we find that the differential cross section of the photoprocess is

$$d\sigma_d(r) = \frac{2\pi e^2 \hbar^2}{c \mu^2 \omega} \text{Im} \left[u^*(r) \frac{du(r)}{dz} \right] \rho d\rho d\varphi, \\ u(r) = \int G(r, r') V(r') \psi(r') dr',$$

where $G(r, r')$ is the Green function of the left-hand side of Eq. (17). The total cross sections is found by integration in the plane P and is independent of z because of conservation of the total current.

The function $G(r, r')$ can be expressed in terms of the integral of the time-dependent Green function of an electron in a homogeneous electric field. Using the well-known expression¹² for the latter, we obtain

$$G(r, r') = \frac{i}{\hbar} \int_0^\infty \left(\frac{\mu}{2\pi i \hbar \tau} \right)^{3/2} \exp \left[\frac{i}{\hbar} S_i(r, r', \tau) \right] d\tau, \quad (18)$$

where

$$S_{\tau}(r, r') = \frac{\mu(r-r')^2}{2\tau} + \frac{1}{2} F\tau(z+z') - \frac{F^2}{24\mu} \tau^3 + E\tau.$$

As before, we shall assume that the motion is quasi-classical and that the reduced action S is large. Then, the integral (18) is found by the stationary phase method:

$$G(r, r') = \frac{\mu}{2\pi\hbar^2} \sum_{n=1}^2 \frac{\exp[iS_{\tau_n}(r, r')/\hbar]}{[1 + (z+z')/l - \varepsilon_n S(r, r')]^{1/2} \varepsilon_n}, \quad (19)$$

where $\varepsilon_{1,2} = \pm 1$,

$$[s(r, r')]^2 = [(2z+l)(2z'+l) - (x-x')^2 - (y-y')^2]/l^2,$$

and the stationary points are found from

$$\tau_{1,2} = \frac{2\mu l}{F} \left[1 + \frac{z+z'}{l} \pm s(r, r') \right].$$

If we assume that the dimensions of an atom are small compared with ρ , z , and l , we obtain

$$d\sigma_n(r) = \frac{e^2 v}{2^{1/2} \pi \mu \omega l(r)} \left\{ \sum_{n=1}^2 |P_n(r)|^2 \frac{1+2z/l - \varepsilon_n t(r)}{[1+z/l - \varepsilon_n t(r)]^{1/2}} + \frac{l}{(\rho^2+z^2)^{1/2}} \operatorname{Re}[P_1(r)P_2^*(r)] \sum_{n=1}^2 \frac{1+2z/l - \varepsilon_n t(r)}{[1+z/l - \varepsilon_n t(r)]^{1/2}} \right\}, \quad (20)$$

$$P_n(r) = \frac{1}{\varepsilon_n^{1/2}} \int \exp[iS_{\tau_n}(r, r')/\hbar] V(r') \psi(r') dr'. \quad (21)$$

The expression (20) has the same structure as the semiclassical formulas (9) and (13). A complete agreement is obtained by assuming that $V(r)\psi(r) = \text{const} \cdot \delta(r)$, which corresponds to an isotropic structure-free source, and if the extra phase $\pi/2$ arising from the inaccuracy of the semiclassical treatment is subtracted from the cosine term in Eq. (13). Since r and r' cannot be separated in the expression for $S_{\tau_n}(r, r')$, the results of integration in Eq. (21) depend on the relationship between r , r' , and l . In general, only numerical integration can be carried out.

5. ALLOWANCE FOR THE INTERACTION IN THE FINAL STATE

Allowance for the interaction in the final state complicates greatly the quantum-mechanical treatment. It is simpler to use the semiclassical approach. We shall show how to estimate the distance between oscillation peaks of the cross sections using the ideas put forward in Secs. 3 and 4. Let the interaction between an electron and the atom in the final state be described by the potential $U(z, \rho)$. Let us consider the trajectories of a photoelectron directed parallel and antiparallel to the z axis. We have seen earlier that these are the trajectories that determine the oscillatory structure of the cross sections.

The difference of the actions is

$$\Delta S = 2 \int_{z_0}^0 [2\mu(E+Fz-U(z, 0))]^{1/2} dz,$$

where z_0 is a negative root of the equation

$$E + Fz - U(z, 0) = 0.$$

The oscillatory contribution to the cross section is proportional to $\cos(\Delta S/\hbar)$. The phase in the cosine term should be modified allowing for: 1) the inaccuracy of the semiclassical approach; 2) the three-dimensional

nature of the real problem; 3) the structure of an atom. It follows from the above discussion that in the $U = 0$ case the sum of the corrections to the phase vanishes (or becomes 2π). We shall assume without proof that the same result is also obtained for $U \neq 0$. Then, the positions of the interference maxima is given by

$$\int_{z_0}^0 [2\mu(E+Fz-U(z, 0))]^{1/2} dz = \pi n \hbar, \quad (22)$$

which (apart from the term $\pi\hbar/4$, which is small compared with ΔS) is identical with the Bohr-Sommerfeld quantization rule for the potential U' , defined by

$$U' = \begin{cases} -Fz + U(z, 0), & z < 0 \\ \infty, & z > 0. \end{cases}$$

These are the quantization conditions used by Freeman *et al.*^{1,2} for the Coulomb case $U(z, 0) = -e^2/|z|$. The positions of the oscillation peaks obtained by them from these conditions are in good agreement with the experimental results, confirming the validity of the semiclassical approach.

However, the approach adopted above has a different physical meaning compared with that employed by Freeman *et al.*^{1,2} Freeman *et al.* in fact imposed the condition that the quasiclassical function vanishes at $z = 0$ and Eq. (2) implies the requirement of an interference maximum at $z = 0$. This is the origin of the difference $\pi\hbar/4$. However, in view of the inaccuracy of the semiclassical approach at low values of n it is not possible to check which of these treatments is in better agreement with the experimental results.

We shall simply point out that our approach is more general because it makes it possible to predict oscillation peaks and calculate the distances between them for any type of interaction U , whereas the results of Freeman *et al.*^{1,2} are obtained only for $U = -e^2/r$ using a special analysis of the classical motion along quasi-periodic trajectories and the potential $-e^2/r - Fz$. Our results are not related to the existence of quasidiscrete levels in a field $U(r) - Fz$. For example, if $U(r)$ is identical with the zero-radius potential, there is one quasisdiscrete level studied by Demkov and Drukarev,¹³ but the number of oscillations of the photodetachment cross section is infinitely large.

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