Strong-field multiphoton ionization of hydrogen:
Nondipolar asymmetry and ponderomotive scattering

Yousef I. Salamin

Physics Department, Birzeit University, P.O. Box 14, Birzeit, West Bank, via Israel

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We present an extension of the strong-field theory of multiphoton ionization due to Reiss aimed at retaining the retardation effects. The time-reversed S-matrix approach and a recently proposed strong-field solution to the Schrödinger equation are used in order to obtain analytic expressions for the ionization rates without making the dipole approximation. Photoionization of the hydrogen ground state is used as an example to demonstrate that deviation from the dipole approximation is substantial for high photon orders or, equivalently, highly energetic photoelectrons. As a by-product, an expression is also obtained for the ponderomotive scattering angle of the photoelectron, measured relative to the field propagation direction, in terms of its directed kinetic and binding energies. [S1050-2947(97)07111-4]

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I. INTRODUCTION

Multiphoton ionization (MPI) of neutral atoms has been the subject of intense theoretical investigation employing various techniques to date [1], mostly treating the radiation field in the dipole approximation (DA). In the DA the expression $\exp(i\mathbf{k}\cdot\mathbf{r}) = 1 + i\mathbf{k}\cdot\mathbf{r} + \cdots$ is replaced by unity when the radiation wavelength is large compared to a dimension of length pertinent to the system. Recent experiments [2], however, have demonstrated that, in describing the radiation field, one may have to go beyond just the first term in the series expansion of the exponential, especially when intensities comparable to 1 a.u., $I_0 = 3.51 \times 10^{16}$ W/cm^2, are considered. ($\mathbf{k}$ is the radiation field propagation vector and $\mathbf{r}$ is the position vector of the electron that interacts with that field.) This *nondipolar asymmetry* has been alluded to by many authors [3–5], motivated by the introduction of ever higher (laser) field intensities for use in the related experiments.

Following ionization, a photoelectron is expected to move in the direction of the polarization vector of the field. However, as has been shown recently in experiments [6–8], a photoelectron gets scattered away from that direction due to acquired longitudinal momentum, a nonzero component of the electron momentum parallel to the field propagation direction. This has been theoretically predicted some years ago [9–12] and termed *ponderomotive scattering*. Ponderomotive scattering has also been the subject of several recent investigations [6–8,13].

In this paper, we employ a strong-field solution to the time-dependent Schrödinger equation suggested recently [14,15] to address these two issues. The formalism leading to closed-form analytic expressions for the n-photon differential transition rates will be developed in Sec. II. Expressions corresponding to linearly and circularly polarized fields will be derived separately. This formalism will be applied to the case of MPI of ground-state hydrogen in Sec. III. The nondipolar asymmetry will be estimated by the magnitude of deviation from unity of the quantity $R_n(\theta)$, henceforth referred to as the the *asymmetry factor*, defined by

$$\frac{dW_n}{d\Omega} = R_n(\theta)\frac{dW_n^{DA}}{d\Omega}, \tag{1}$$

where θ gives the direction of motion of the photoelectron relative to $\mathbf{k}$, the laser propagation vector, z is an intensity parameter to be defined below, $dW_n^{DA}/d\Omega$ is the n-photon (partial) differential transition rate per unit solid angle when the calculation is done in the dipole approximation, and $dW_n/d\Omega$ is the same quantity calculated without making the dipole approximation.

Moreover, an exact expression will be derived for the angle relative to the direction of $\mathbf{k}$ at which the photoelectron may be detected (the ponderomotive scattering angle). The angle in question may be obtained from

$$\cos\Theta = \frac{p_\parallel}{p}, \tag{2}$$

where $p_\parallel$ is the component of the total electron momentum $\boldsymbol{p}$ parallel to $\mathbf{k}$. Specific results will be presented and discussed in Sec. IV. Finally, a brief summary will be given in Sec. V.

II. THEORY

Most of the details presented in this section may be found elsewhere [16]. They are reproduced here in order to make this paper as self-contained as possible. We start by considering a one-electron system, a negative ion, or a hydrogen-like atom, interacting with a radiation (laser) field. Let H_0 be the noninteracting (field-free) Hamiltonian of the system and denote by H_I the effect of the externally applied field. Provided the field is strong enough, the electron may absorb many photons from it, using part of the absorbed energy to free itself from the binding effect of the nucleus and the remainder as kinetic energy of oscillation and translation in the field.

Following Reiss [17–20], the strong-field multiphoton transition rates may be calculated from the *time-reversed S matrix*

$$(S-1)_{fi} = -\frac{i}{\hbar}\int dt\langle\Psi_f|H_I|\Phi_i\rangle. \tag{3}$$

In Eq. (3), Ψ is a fully interacting electron state, a solution to the Schrödinger equation

$$\left(i\hbar \frac{\partial}{\partial t} - H_0 - H_I \right) \Psi = 0, \tag{4}$$

while Φ is a noninteracting state satisfying the equation

$$\left(i\hbar \frac{\partial}{\partial t} - H_0 \right) \Phi = 0. \tag{5}$$

Equation (5) will be assumed to possess exact solutions, a set of known stationary states of which one may typically be written as

$$\Phi(\mathbf{r}, t) = \phi(\mathbf{r}) e^{-iEt/\hbar}, \tag{6}$$

where E is the state eigenenergy. Analytic solutions for Eq. (4) do not exist for most practical situations. Many numerical techniques instead have been designed [1] in order to obtain solutions for this equation, with varying degrees of success. It is well known that the numerical methods are computationally very demanding, even for the simplest of systems, and that perturbation theory breaks down when field intensities comparable to 1 a.u. are used.

In the present paper, the following solution to Eq. (4) involving an interaction term H_I of arbitrary strength will be used. The solution, recently proposed by Frasca [14], reads

$$\Psi(t) = T_0(t) \left\{ 1 + \left(-\frac{i}{\hbar} \right) \int_{-\infty}^{t} H_0'(t_1) dt_1 \right.$$
$$+ \cdots + \left(-\frac{i}{\hbar} \right)^k \int_{-\infty}^{t} H_0'(t_1) dt_1 \int_{-\infty}^{t_1} H_0'(t_2) dt_2 \cdots$$
$$\left. \times \int_{-\infty}^{t_{k-1}} H_0'(t_k) dt_k + \cdots \right\} \Psi^{(0)}, \tag{7}$$

where

$$T_0(t) = \exp \left[-\frac{i}{\hbar} \int_{-\infty}^{t} H_I(t') dt' \right] \tag{8}$$

and

$$H_0'(t) = T_0(t)^{-1} H_0 T_0(t). \tag{9}$$

Equation (7) looks deceptively like a Dyson series [21]; the difference is that in Eq. (7), in place of the interaction piece of the Hamiltonian, we have the field-free Hamiltonian transformed according to Eq. (9). In other words, the field-free Hamiltonian H_0 and the interaction term H_I play roles here that are the opposite of their familiar Dyson-series roles. Unfortunately, like the Dyson series, the series expressed by Eq. (7) cannot be summed except under specialized conditions. Furthermore, it has been shown that corrections to the zeroth-order term contain secular as well as oscillatory terms that may render the wave function unnormalizable over long times. The good news, though, is that one does not always need to consider many terms beyond zeroth order in order to arrive at meaningful and reliable results, as will be shown shortly.

Let m and e be the mass and charge of the electron and denote by $\mathbf{A}$ the vector potential of the radiation (laser) field. The interaction piece of the system's Hamiltonian may be written as

$$H_I = -\frac{e}{mc} \mathbf{A} \cdot \hat{\mathbf{P}} + \frac{e^2}{2mc^2} \mathbf{A}^2, \tag{10}$$

where c is the speed of light and $\hat{\mathbf{P}}$ is the linear momentum operator of the electron. Within the spirit of the time-reversed S-matrix approach to strong-field processes [20], we may now take for the initial state

$$\Phi_i = \phi_i(\mathbf{r}) e^{-iE_i t/\hbar}, \tag{11}$$

i.e., one of the stationary states of the bound electron, assumed to be known exactly. On the other hand, the final state will be taken as the zeroth-order term in Eq. (7), namely,

$$\Psi_f \approx T_0 \Psi^{(0)} = T_0(t) [\phi_f(\mathbf{r}) e^{-iE_f t/\hbar}]. \tag{12}$$

This choice is motivated by the fact that, especially when very intense fields are considered, the final state of the electron is dominated by the radiation field and is almost free from the influence of the binding potential. In the following two subsections, transition matrix elements to describe the interaction of the single electron with linearly and circularly polarized laser fields of arbitrary strength will be obtained according to Eq. (3) and employing the recipe expressed in Eqs. (11) and (12).

A. Linear polarization

A linearly polarized single-mode field of constant amplitude a, propagation vector $\mathbf{k}$, and frequency ω may be modeled by the (classical) vector potential

$$\mathbf{A} = \hat{\boldsymbol{\epsilon}} a \cos(\mathbf{k} \cdot \mathbf{r} - \omega t), \tag{13}$$

where $\hat{\boldsymbol{\epsilon}}$ is a polarization unit vector, perpendicular to the field propagation direction (transverse gauge). We now calculate the various terms making up the S matrix in Eq. (3), beginning with

$$H_I \Phi_i = \hbar \omega \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s})$$
$$\times [-\zeta \cos\eta + z \cos 2\eta + z] e^{-i(E_i t - \mathbf{s} \cdot \mathbf{r})/\hbar}, \tag{14}$$

where

$$\zeta = \frac{ea}{mc\hbar\omega} \hat{\boldsymbol{\epsilon}} \cdot \mathbf{s}, \quad z = \frac{(ea)^2}{4mc^2\hbar\omega}, \quad \eta = \mathbf{k} \cdot \mathbf{r} - \omega t. \tag{15}$$

In order to facilitate action of the momentum operator, the momentum representation of the stationary-state wave function has been introduced via the definition

$$\phi(\mathbf{r}) = \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}(\mathbf{s}) e^{i\mathbf{s} \cdot \mathbf{r}/\hbar}. \tag{16}$$

Note that, in arriving at Eq. (14), use has been made of the fact that, for any function $f(\eta)$,

$$\hat{\boldsymbol{\epsilon}} \cdot \hat{\mathbf{P}}(f e^{i\mathbf{s} \cdot \mathbf{r}/\hbar}) = \hat{\boldsymbol{\epsilon}} \cdot \mathbf{s}(f e^{i\mathbf{s} \cdot \mathbf{r}/\hbar}), \qquad (17)$$

due to the transversality condition $\hat{\boldsymbol{\epsilon}} \cdot \mathbf{k} = 0$. The remaining term in the S matrix will now be calculated. With the help of Eqs. (16) and (17), the zeroth-order term in the fully interacting state (7) becomes

$$\Psi_f = \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f(\mathbf{s}') e^{-i(E_f t - \mathbf{s}' \cdot \mathbf{r} - z\hbar\eta)/\hbar}$$

$$\times e^{i[-\zeta' \sin\eta + (z/2)\sin 2\eta]}, \qquad (18)$$

where ζ' differs from ζ in the obvious dependence upon the linear momentum of the electron. We are now in a position to evaluate the transition S-matrix element. Using Eqs. (14) and (18) in Eq. (3) leads to

$$(S-1)_{if}^{(lin)} = -i\omega \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f^*(\mathbf{s}')$$

$$\times \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s})$$

$$\times \int d^3 r \ e^{i(\mathbf{s} - \mathbf{s}' - z\hbar\mathbf{k}) \cdot \mathbf{r}/\hbar} \int dt \ e^{i(E_f - E_i + z\hbar\omega)t/\hbar}$$

$$\times [-\zeta\cos\eta + z\cos 2\eta + z]$$

$$\times e^{i[\zeta' \sin\eta - (z/2)\sin 2\eta]}, \qquad (19)$$

where the superscript (lin) stands for *linear polarization* and Eq. (17) has been used again. Next we proceed to evaluate the integrals over t, $\mathbf{r}$, and $\mathbf{s}$, in that order for convenience. The time integration may easily be done after the following transformations have been made. First, the trigonometric functions in the expression between square brackets are expressed in exponential form. This is followed by replacing the exponential factor on the far right of Eq. (19) by a generalized Bessel function expansion [17], according to

$$e^{i[u \sin\phi + v\sin 2\phi]} = \sum_{n=-\infty}^{\infty} e^{in\phi} J_n(u,v). \qquad (20)$$

After this has been done, Eq. (19) becomes

$$(S-1)_{if}^{(lin)} = -i\omega \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f^*(\mathbf{s}')$$

$$\times \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s})$$

$$\times \int d^3 r \ e^{i(\mathbf{s} - \mathbf{s}' - z\hbar\mathbf{k}) \cdot \mathbf{r}/\hbar} \sum_{n=-\infty}^{\infty} J_n\left(\zeta', -\frac{z}{2}\right)$$

$$\times \left[-\frac{\zeta}{2}(e^{i(n+1)\mathbf{k} \cdot \mathbf{r}} I_1 + e^{i(n-1)\mathbf{k} \cdot \mathbf{r}} I_{-1}) \right.$$

$$\left. + \frac{z}{2}(e^{i(n+2)\mathbf{k} \cdot \mathbf{r}} I_2 + e^{i(n-2)\mathbf{k} \cdot \mathbf{r}} I_{-2}) + z e^{in\mathbf{k} \cdot \mathbf{r}} I_0 \right], \qquad (21)$$

where typically

$$I_\ell = \int_{-\infty}^{\infty} dt e^{i[(E_f - E_i)/\hbar - (n + \ell - z)\omega]t}$$

$$= 2\pi\delta\left(\frac{E_f - E_i}{\hbar} - (n + \ell - z)\omega \right) \qquad (22)$$

for $\ell = 1$, -1, 2, -2, and 0, respectively. The following steps consist of changing the summation index n to $n - \ell$ in the respective terms of Eq. (21) and pulling out the common δ function and the resulting exponential factor in $\mathbf{k} \cdot \mathbf{r}$. After this has been done, the S-matrix element takes the form

$$(S-1)_{if}^{(lin)} = -i\omega \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f^*(\mathbf{s}')$$

$$\times \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s}) \sum_{n=-\infty}^{\infty}$$

$$\times \int d^3 r \ e^{i[\mathbf{s} - \mathbf{s}' + (n-z)\hbar\mathbf{k}] \cdot \mathbf{r}/\hbar}$$

$$\times \left\{ -\frac{\zeta}{2}(J_{n-1} + J_{n+1}) \right.$$

$$\left. + \frac{z}{2}(J_{n-2} + J_{n+2}) + z J_n \right\}$$

$$\times 2\pi\delta\left(\frac{E_f - E_i}{\hbar} - (n-z)\omega \right). \qquad (23)$$

The arguments of the generalized Bessel functions in Eq. (23) have been suppressed, but are understood to be $(\zeta', -z/2)$. Now is a good point to carry out the integration over the spatial coordinates. This results in the momentum-conserving δ function

$$\int_{-\infty}^{\infty} d^3 r \ e^{i[\mathbf{s} - \mathbf{s}' + (n-z)\hbar\mathbf{k}] \cdot \mathbf{r}/\hbar}$$

$$= (2\pi\hbar)^3 \delta^{(3)}(\mathbf{s} - \mathbf{s}' + (n-z)\hbar\mathbf{k}). \qquad (24)$$

Equation (24) may next be used to calculate the integration over $\mathbf{s}'$, resulting in the replacement of $\mathbf{s}'$ by $\mathbf{s} + (n-z)\hbar\mathbf{k}$ everywhere. In particular, in place of ζ we will have

$$\zeta' = \frac{ea}{mc\hbar\omega} \hat{\boldsymbol{\epsilon}} \cdot [\mathbf{s} + (n-z)\hbar\mathbf{k}] = \frac{ea}{mc\hbar\omega} \hat{\boldsymbol{\epsilon}} \cdot \mathbf{s} = \zeta \quad (25)$$

due to the transversality condition. The recurrence relation [17]

$$2n J_n(u,v) = u\{J_{n-1}(u,v) + J_{n+1}(u,v)\}$$

$$+ 2v\{J_{n-2}(u,v) + J_{n+2}(u,v)\} \qquad (26)$$

will next be used to simplify the quantity between curly brackets in Eq. (23). After this series of steps has been carried out, the desired matrix element takes the form

$$(S-1)_{if}^{(lin)} = i\omega \sum_{n=-\infty}^{\infty} (n-z) \left\langle \widetilde{\phi}_f(\mathbf{s} + (n-z)\hbar\mathbf{k}) \right|$$

$$\times J_n\left(\zeta, -\frac{z}{2}\right)\bigg|\widetilde{\phi}_i(\mathbf{s})\bigg\rangle$$

$$\times 2\pi\delta\left(\frac{E_f - E_i}{\hbar} - (n-z)\omega\right). \quad (27)$$

The remaining integration is understood to be over $\mathbf{s}$. Equation (27) may then be used to calculate the transition probability per unit time, as usual [21–23], from

$$w^{(lin)} = \lim_{t\to\infty} \frac{|(S-1)_{if}|^2}{t}$$

$$= \omega^2 \sum_{n=-\infty}^{\infty} (n-z)^2 \bigg|\bigg\langle \widetilde{\phi}_f(\mathbf{s}+(n-z)\hbar\mathbf{k})\bigg| J_n\left(\zeta, -\frac{z}{2}\right)$$

$$\times \bigg|\widetilde{\phi}_i(\mathbf{s})\bigg\rangle\bigg|^2 2\pi\delta\left(\frac{E_f - E_i}{\hbar} - (n-z)\omega\right). \quad (28)$$

For a particular value of n, the energy-conserving δ function in Eq. (28) effectively says that the transition $i\to f$ is accompanied by the absorption of n photons of energy $n\hbar\omega$, of which $z\hbar\omega$ goes into the classical oscillatory motion of the electron and the rest $(n-z)\hbar\omega$ goes into kinetic energy of oscillation and translation in the field [3]. On the other hand, Eq. (16) tells us that the momentum of the initial state is $\mathbf{p}_i = \mathbf{s}$. Hence the final-state momentum, due to the momentum-conserving δ function of Eq. (24), is

$$\mathbf{p}_f = \mathbf{s}' = \mathbf{p}_i + (n-z)\hbar\mathbf{k}. \quad (29)$$

This equation clearly shows that the electron absorbs momentum in the amount $(n-z)\hbar\mathbf{k}$ from the radiation field. The absorbed momentum is all in the forward direction and is important for high photon orders (large n). Thus the photoelectron will be ejected at some angle θ_d relative to the polarization direction of the field [20]. This issue will be discussed further below.

B. Circular polarization

We consider next the case of a (left) circularly polarized, plane-wave, monomode radiation field of polarization vectors $\hat{\boldsymbol{\epsilon}}_x$ and $\hat{\boldsymbol{\epsilon}}_y$ along the x and y directions. This case is well known to involve only ordinary rather than generalized Bessel functions [3]. Using notation similar to what has been adopted in Eq. (13) above, the vector potential of such a field may be written as

$$\mathbf{A} = \frac{a}{\sqrt{2}}[\hat{\boldsymbol{\epsilon}}_x \sin(\mathbf{k}\cdot\mathbf{r} - \omega t) - \hat{\boldsymbol{\epsilon}}_y \cos(\mathbf{k}\cdot\mathbf{r} - \omega t)]. \quad (30)$$

The steps leading to an expression for the transition probability per unit time similar to the one given in Eq. (28) above, for the linear polarization case, are more or less the same in the present case. We shall therefore skip most of the details and give below only the most important intermediate results. For example,

$$H_I = -\hbar\omega\left[\frac{\beta}{\sqrt{2}}(\hat{P}_x\sin\eta - \hat{P}_y\cos\eta) - z\right], \quad (31)$$

where $\hat{P}_x$ and $\hat{P}_y$ are the x and y components, respectively, of the electron's linear momentum operator and $\beta = ea/mc\hbar\omega$. Writing $\mathbf{s}$ in spherical polar coordinates, $\mathbf{s} = (s, \theta_s, \phi_s)$, in H_I, and making use of Eqs. (16) and (17), the S matrix for the present case becomes

$$(S-1)_{if}^{cir} = i\omega \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f^*(\mathbf{s}') \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s})$$

$$\times \int d^3 r\, e^{i(\mathbf{s}-\mathbf{s}'-z\hbar\mathbf{k})\cdot\mathbf{r}/\hbar}$$

$$\times \int dt\, e^{i(E_f - E_i + z\hbar\omega)t/\hbar}$$

$$\times \left[\frac{\beta}{\sqrt{2}} s\,\sin\theta_s\sin(\eta - \phi_s) - z\right]$$

$$\times e^{-i[(\beta/\sqrt{2})s'\sin\theta_{s'}\cos(\eta - \phi_{s'})]}. \quad (32)$$

The exponential term on the far right of Eq. (32) will next be expanded in terms of the ordinary Bessel functions using the Fourier-Bessel series

$$e^{-iu\cos\xi} = \sum_{n=-\infty}^{\infty} (-i)^n J_n(u)e^{in\xi}. \quad (33)$$

When $\sin(\eta - \phi_s)$ is also written in terms of exponentials, one arrives at an equation similar to Eq. (21), which reads

$$(S-1)_{if}^{(cir)} = i\omega \int \frac{d^3 s'}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_f^*(\mathbf{s}') \int \frac{d^3 s}{(2\pi\hbar)^{3/2}} \widetilde{\phi}_i(\mathbf{s})$$

$$\times \int d^3 r\, e^{i(\mathbf{s}-\mathbf{s}'-z\hbar\mathbf{k})\cdot\mathbf{r}/\hbar}$$

$$\times \sum_{n=-\infty}^{\infty} J_n\left(\frac{\beta}{\sqrt{2}}s'\sin\theta_{s'}\right)$$

$$\times \left[\frac{\beta}{2\sqrt{2}}s\,\sin\theta_s[(-i)^{n+1}e^{i(n+1)(\mathbf{k}\cdot\mathbf{r} - \phi_{s'})}\right.$$

$$\times e^{i(\phi_{s'} - \phi_s)}I_1 - (-i)^{n+1}$$

$$\times e^{i(n-1)(\mathbf{k}\cdot\mathbf{r} - \phi_{s'})}e^{-i(\phi_{s'} - \phi_s)}I_{-1}]$$

$$\left. + (-i)^n z e^{in(\mathbf{k}\cdot\mathbf{r} - \phi_{s'})}I_0\right], \quad (34)$$

with

$$I_q = \int_{-\infty}^{\infty} dt\, e^{i[(E_f - E_i)/\hbar - (n+q-z)\omega]t}$$

$$= 2\pi\delta\left(\frac{E_f - E_i}{\hbar} - (n+q-z)\omega\right) \quad (35)$$

for $q = 1, -1$, and 0, respectively. Next, we let $n\to n-q$ in Eq. (34), after which one should be able to evaluate the integration over $\mathbf{r}$ and get the linear momentum-conserving δ

function $(2\pi\hbar)^3\delta^{(3)}(\mathbf{s}-\mathbf{s}'+(n-z)\hbar\mathbf{k})$. Then the integration over $\mathbf{s}'$ is carried out, where by definition the δ function is nonzero only if

$$\mathbf{s}'=\mathbf{s}+(n-z)\hbar\mathbf{k}. \tag{36}$$

Recall that in Eq. (30) the direction of the field propagation has been taken along $+z$, i.e., $\mathbf{k}$ has only a z component. Thus the x and y components of Eq. (36) yield

$$s'\sin\theta_{s'}\cos\phi_{s'}=s\,\sin\theta_s\cos\phi_s \tag{37}$$

and

$$s'\sin\theta_{s'}\sin\phi_{s'}=s\,\sin\theta_s\sin\phi_s, \tag{38}$$

respectively. When Eqs. (37) and (38) are solved simultaneously, $\phi_{s'}=\phi_s$ and $s'\sin\theta_{s'}=s\sin\theta_s$ result. We then use these results in Eq. (34) and employ the well-known Bessel function recurrence relation

$$\frac{2n}{u}J_n(u)=J_{n-1}(u)+J_{n+1}(u) \tag{39}$$

to write the S-matrix element in terms of a sum containing only J_n. The end result is

$$(S-1)_{if}^{(cir)}=i\omega\sum_{n=-\infty}^{\infty}(-i)^n(n-z)\left\langle\widetilde{\phi}_f(\mathbf{s}+(n-z)\hbar\mathbf{k})\right|$$

$$\times e^{-in\phi_s}J_n\left(\frac{\beta}{\sqrt{2}}s\,\sin\theta_s\right)\left|\widetilde{\phi}_i(\mathbf{s})\right\rangle$$

$$\times 2\pi\delta\left(\frac{E_f-E_i}{\hbar}-(n-z)\omega\right). \tag{40}$$

The corresponding transition probability per unit time will finally be given by

$$w^{(cir)}=\omega^2\sum_{n=-\infty}^{\infty}(n-z)^2\left|\left\langle\widetilde{\phi}_f(\mathbf{s}+(n-z)\hbar\mathbf{k})\right.\right.$$

$$\times\left.\left.e^{-in\phi_s}J_n\left(\frac{\beta}{\sqrt{2}}s\,\sin\theta_s\right)\right|\widetilde{\phi}_i(\mathbf{s})\right\rangle\Bigg|^2$$

$$\times 2\pi\delta\left(\frac{E_f-E_i}{\hbar}-(n-z)\omega\right). \tag{41}$$

Note that the expressions obtained in this work for the transition probabilities per unit time [Eqs. (28) and (41)] may be compared with the corresponding ones obtained in the dipole approximation by Reiss [3,11]. One can arrive at the dipole limits of Eqs. (28) and (41) simply by letting $\mathbf{k}\to\mathbf{0}$. Agreement is complete.

III. MULTIPHOTON IONIZATION

A. General

In a typical photoionization process, the initial state of the electron is a bound state, assumed to be known exactly, whose binding energy is given by $E_i=-E_B$. A description of the final state, however, in terms of a plane wave seems to be quite acceptable [22,24–27]. Thus we take $\phi_f(\mathbf{r})=(2\pi\hbar)^{-3/2}\exp(i\mathbf{p}\cdot\mathbf{r}/\hbar)$, where $\mathbf{p}$ is the (constant) momentum of the ionized electron, with the corresponding energy $E_f=p^2/2m$. The Fourier transform of this wave function is the three-dimensional δ function $\widetilde{\phi}_f(\mathbf{s})=\delta^{(3)}(\mathbf{p}-\mathbf{s})$. When this is used in Eq. (28) above and after the remaining integration over $\mathbf{s}$ has been carried out,

$$w^{(lin)}=\omega^2\sum_{n=-\infty}^{\infty}(n-z)^2|\widetilde{\phi}_i(\mathbf{p}-(n-z)\hbar\mathbf{k})|^2$$

$$\times\left|J_n\left(\zeta,-\frac{z}{2}\right)\right|^2 2\pi\delta\left(\frac{p^2/2m+E_B}{\hbar}-(n-z)\omega\right). \tag{42}$$

results. This expression too has the correct limit [12] when the dipole approximation is made in the manner described above. For the case of circular polarization, one obtains the following transition rate without much difficulty:

$$w^{(cir)}=\omega^2\sum_{n=-\infty}^{\infty}(n-z)^2|\widetilde{\phi}_i(\mathbf{p}-(n-z)\hbar\mathbf{k})|^2 J_n^2\left(\frac{\beta}{\sqrt{2}}p\,\sin\theta\right)$$

$$\times 2\pi\delta\left(\frac{p^2/2m+E_B}{\hbar}-(n-z)\omega\right), \tag{43}$$

where θ is the angle between the direction of electron emission and the laser propagation direction. Note that Eqs. (42) and (43) are almost structurally the same; the only difference between them is in the kind of Bessel function involved. We now transform the δ function in both expressions according to

$$\delta\left(\frac{p^2/2m+E_B}{\hbar}-(n-z)\omega\right)=\frac{m\hbar}{\bar{p}}\delta(p-\bar{p}), \tag{44}$$

where

$$\bar{p}=\sqrt{2m\hbar\omega(n-z-\epsilon_B)} \tag{45}$$

and $\epsilon_B=E_B/\hbar\omega$ gives the number of photons the electron must absorb in order to break loose of the binding potential. Note that in Eq. (44) another δ function, $\delta(p+\bar{p})$, has been dropped on account of the fact that the magnitude of the free-particle momentum cannot be negative. Furthermore, Eq. (45) replaces the lower limit on the sum in Eqs. (42) and (43) by a *threshold* value. According to Eq. (45), the threshold photon order is an integer n_0 given by

$$n_0\geq z+\epsilon_B. \tag{46}$$

Recall that z may be written as the ratio of the average classical energy of oscillation of the (free) electron in the radiation field, *the ponderomotive potential energy* $U_p=(ea)^2/4mc^2$, to the photon energy $\hbar\omega$. In other words, z is the number of photons absorbed whose energy goes into energy of classical oscillation in the field. This leaves the electron with a directed kinetic energy in the magnitude

$$K=\hbar\omega(n-z-\epsilon_B). \tag{47}$$

Recall also that the final electron momentum, whose magnitude is given by Eq. (45), has a forward component given by

$$p_\parallel = (n-z)\hbar \mathbf{k}. \tag{48}$$

This value for the forward momentum may be read off of Eq. (29) assuming, of course, that all the initial momentum is transverse. With this assumption in mind, Eq. (2) now yields

$$\cos\Theta = \frac{(n-z)\hbar k}{\sqrt{2m\hbar\omega(n-z-\epsilon_B)}} = \sqrt{\frac{K}{2mc^2}} + \frac{E_B}{\sqrt{2mc^2 K}} \tag{49}$$

for the angle relative to the field propagation direction at which the photoelectron emerges. In arriving at the second line in Eq. (49) use has been made of Eq. (47). Note that, apart from the dependence upon the binding energy E_B, Eq. (49) gives Θ as a function of the electron kinetic energy K. The term involving the binding energy, whose presence limits its applicability of the result only to electrons produced by photodetachment or photoionization of a specific system, may be dropped for situations in which $K \gg E_B$. The resulting approximate expression is then applicable to electrons produced by any means from any atomic, ionic, or molecular system [6–8].

Finally, the total transition rate (to all possible final states) may be obtained by integrating the transition rates given by Eqs. (42) and (43) over the phase space available to the free particle. For this purpose, we use

$$W = \int w \, \frac{d^3p}{(2\pi\hbar)^3}, \tag{50}$$

where $(2\pi\hbar)^3$ is the volume in phase space of a unit quantum-mechanical cell. Thus the total transition rate per unit solid angle becomes

$$\frac{dW}{d\Omega} = \frac{(2m^3\omega^5)^{1/2}}{(2\pi)^2\hbar^{3/2}} \sum_{n=n_0}^{\infty} (n-z)^2(n-z-\epsilon_B)^{1/2}|\widetilde{\phi}_i|^2 J_n^2. \tag{51}$$

In this expression, J_n stands for $J_n(\zeta, -z/2)$ in the case of linear polarization and for $J_n((\beta/\sqrt{2}\bar{p}\sin\theta)$ in the circular polarization case. It should also be borne in mind that the dependence on the momentum, in the momentum-space wave function as well as in the arguments of the Bessel functions, ought to be through $\mathbf{p} - (n-z)\hbar\mathbf{k}$, with p replaced by $\bar{p}$ everywhere in accordance with the δ function of Eq. (44). See the example below.

B. Photoionization of ground-state hydrogen

We now move on to some applications. The example we wish to take is photoionization of the hydrogen $1S$ state. Not only is this example relatively simple to handle, but it has also been studied before in the dipole approximation and relativistically [3,11,12,17]. The binding energy of the hydrogen ground-state electron is $E_B = \hbar^2/2ma_0^2$, where a_0 is the Bohr radius. The ground-state wave function is

$$\phi_i(\mathbf{r}) = \frac{1}{a_0^{3/2}\sqrt{\pi}} e^{-r/a_0}. \tag{52}$$

For photoionization studies employing the formalism developed above, one needs the Fourier transform of the bound-state wave function. Direct calculation gives

$$\widetilde{\phi}_i(s) = \frac{8\sqrt{\pi a_0^3}}{(2\pi\hbar)^{3/2}} \frac{1}{[1+(sa_0/\hbar)^2]^2}. \tag{53}$$

We now calculate the photoionization rate of the hydrogen $1S$ state by circularly polarized light. Inserting expression (17) into Eq. (51) and after some simplification, we get

$$\frac{dW}{d\Omega} = \frac{8}{(2\pi\hbar)^3} \frac{\omega}{\pi} \epsilon_B^{5/2} \sum_{n=n_0}^{\infty} \frac{(n-z-\epsilon_B)^{1/2} J_n^2(\sqrt{4z(n-z-\epsilon_B)}\sin\theta)}{\left[1 - \frac{\hbar k}{m\hbar\omega}\sqrt{2m\hbar\omega(n-z-\epsilon_B)}\cos\theta + \frac{\hbar^2 k^2}{2m\hbar\omega}(n-z)\right]^4}. \tag{54}$$

Note at this point that, apart from the factor $(2\pi\hbar)^{-3}$ upfront, Eq. (54) reduces, in the dipole approximation ($k \to 0$), exactly to the expression derived by Reiss [17] for the same system. The absence of the factor $(2\pi\hbar)^{-3}$ from the result of Reiss may be traced back to the way the Fourier transform has been defined.

The computing power available to the author at the present time is not adequate for calculations involving the total transition rate on the basis of Eq. (54). Thus we will not be able to check the ascertion made by Reiss [11], among other things, that the retardation effects are of the same order of magnitude as the *other* relativistic corrections. Some meaningful conclusions can still be arrived at by studying the n-photon (partial) rates of ionization. For example, we are now in a position to obtain the following expression for the nondipolar asymmetry factor as defined in Eq. (1):

$$R_n(\theta) = \left[1 - \frac{\hbar k}{m\hbar\omega}\sqrt{2m\hbar\omega(n-z-\epsilon_B)}\cos\theta \right.$$
$$\left. + \frac{\hbar^2 k^2}{2m\hbar\omega}(n-z)\right]^{-4} \tag{55}$$

$$= \left[1 - \frac{\hbar k}{m\hbar\omega}\sqrt{2m\hbar\omega(n-n_0)}\cos\theta \right.$$
$$\left. + \frac{\hbar^2 k^2}{2m\hbar\omega}(n-n_0+\epsilon_B)\right]^{-4} \tag{56}$$

$$= \left[1 - \sqrt{\frac{2K}{mc^2}}\cos\theta + \left(\frac{K+E_B}{2mc^2}\right)\right]^{-4}. \tag{57}$$
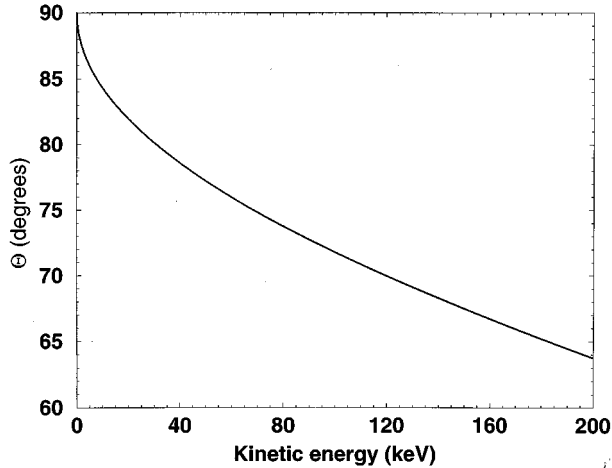
FIG. 1. Ponderomotive scattering angle Θ of an electron produced by photoionization of the hydrogen $1S$ state vs its directed kinetic energy K.

Exactly the same expressions describe the asymmetry factor in the linear polarization case. Finally, the relative error resulting from adopting the dipole approximation in a calculation of the n-photon rate of ionization may be written as

$$\Delta = 1 - R(\theta)^{-1}. \tag{58}$$

The main results obtained in this section will be the subject of discussion in the following section.

IV. RESULTS AND DISCUSSION

In the recent experiments by Moore *et al.* [6] and Meyerhofer *et al.* [7], which reported observation of the phenomenon of ponderomotive scattering, the electrons were produced *almost at rest* by photoionization. Various expressions have been reported in the past for the ponderomotive scattering angle Θ in terms of the directed kinetic energy of the electron, some of them based on quantum-mechanical arguments [10,11], while others were derived classically [6,7,13]. The experimental results of Meyerhofer *et al.* are consistent with these expressions. Equation (49), derived for the same quantity in this paper, involves a feature not found in the previous expressions, namely, the dependence upon the binding energy E_B. This dependence, however, is very weak for situations such that $E_B \ll K$. We plot the scattering angle against the kinetic energy of the photoelectron in Fig. 1 for electrons produced by photoionization of the hydrogen ground-state electron. The curve agrees quite well with the earlier results [6–8,13].

The other issue we set out to investigate in this paper concerns the need to go beyond the dipole approximation in calculating the MPI rates when high-intensity fields are employed. Note first that the intensity parameter z may be eliminated from the expression for the asymmetry factor in favor of the quantity $n - n_0$, the number of photons whose absorbed energy goes into directed kinetic energy of the photoelectron [see Eq. (47)]. This has been done in Eqs. (56) and (57).

One can still push the last point even further by eliminating the $\cos\theta$ term in Eq. (57) in favor of the directed kinetic
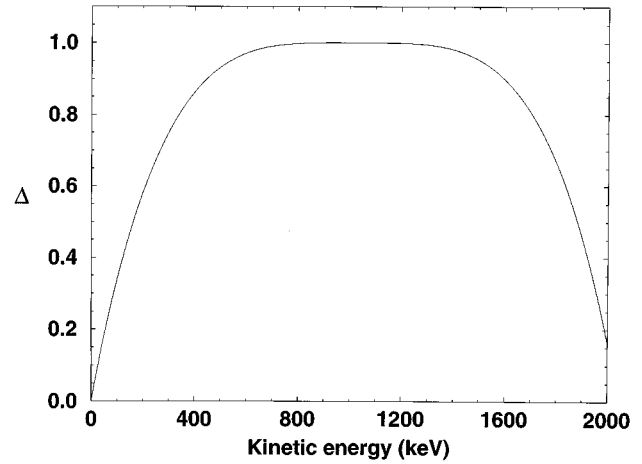

FIG. 2. Relative error Δ made when the dipole approximation is employed in calculating the n-photon (partial) rate of ionization of the hydrogen ground-state electron vs the photoelectron escape kinetic energy K for photoelectron kinetic energies of up to 2 MeV.

energy K, employing Eq. (49) with θ replaced by Θ. This leads to the following expression for Δ:

$$\Delta = 1 - \left[1 - \frac{K + E_B}{2mc^2} \right]^4. \tag{59}$$

One easily finds that Δ has a maximum value corresponding to the photoelectron kinetic energy $K = 2mc^2 - E_B \approx 1$ MeV. For electrons moving with this kinetic energy, $\Delta = 1$ and hence the dipole approximation is completely unreliable. On the other hand, for low-energy electrons ($K \approx 0$), the dipole approximation results in a minimum of error, $\Delta \approx 0$. In Fig. 2, Δ is shown against the directed kinetic energy of the photoelectron for kinetic energies of up to 2 MeV.

Since Eq. (49) has been used in arriving at Eq. (59), they and Figs. 1 and 2 may best be read together. Equation (49) and Fig. 1 basically tell us where to look for a photoelectron with a particular kinetic energy. Then Eq. (59) and Fig. 2 give the error in the value of the n-photon rate of ionization calculated on the basis of the dipole approximation. Recall that n may be calculated from K and a knowledge of ϵ_B and n_0 [see Eq. (47)].

V. SUMMARY

In this paper, general expressions for the multiphoton detachment and multiphoton ionization rates have been derived for one-electron systems based upon a recently proposed strong-field solution to the Schrödinger equation, without resorting to the usual dipole approximation. It has been shown that, especially in the presence of superintense fields that involve absorption of extremely large numbers of photons, the dipole approximation results in substantial errors only when differential multiphoton ionization rates of high photon orders are calculated. In such situations involving fast-moving photoelectrons, the dipole approximation must clearly be abandoned. On the other hand, the main contribution to the total rates comes from photons of order near the

threshold number n_0 and for electrons detected in the transverse direction. For such circumstances, the dipole approximation is quite adequate.

As a by-product of the analysis presented in this paper, we have derived an expression giving the ponderomotive scattering angle of the photoelectron in terms of its directed kinetic energy. Unlike previous derivations, the formula derived here involves the binding energy of the electron in the parent system (the neutral atom). As such, our expression should be suitable for analysis of the data of experimental situations involving electrons produced via ionization [6–8].

In order to be able to make an estimate of the effects of retardation on the magnitude of a typical total ionization rate, better computing power than is currently available to the present author is needed. This particular issue, among others, will be taken up in the future.

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