Multiphoton transitions in a strong field: Inclusion of the photon momentum

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Taking into account the photon momentum (i.e., without making the dipole approximation) we derive some different expressions for the nonrelativistic amplitude for a one-electron system to undergo a transition in a classical monochromatic field. As an application, we use the Keldysh approximation, generalized beyond the dipole approximation, to examine the effect of the photon momentum on the angular distribution for multiphoton detachment of a model H^- ion by a strong low-frequency field. We find that when the field is very strong the neglect of the *transverse* Doppler shift (but inclusion of the nonrelativistic Doppler shift) leads to a spuriously large rate for multiphoton detachment, even though the relativistic correction to the rate is very small.

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I. INTRODUCTION

In treating multiphoton ionization of an atomic system by a laser field it is usually legitimate to neglect the photon momentum, i.e., to make the dipole approximation. However, in the presence of very strong fields the photoelectron can absorb many photons, and even though the momentum of a single photon may be negligible, the net photon momentum may be significant. Inclusion of the photon momentum leads not only to a (nonrelativistic) Doppler shift in the frequency seen by the ejected photoelectron, but also to a change in the angular distribution.

In this paper we derive expressions for the *non*relativistic amplitude for a one-electron system to undergo a multiphoton transition, without making the dipole approximation. We treat the field as a classical, purely homogeneous and monochromatic plane wave—we ignore the entrance of the system into the field, and its subsequent exit (processes which eventually must be treated). We reformulate a general variational principle, originally derived by Rosenberg [1], which yields an estimate of the amplitude with an error that is of second order in the errors of the trial state vectors. As input to this variational principle, we consider different choices of trial state vectors. For example, we generalize, beyond the dipole approximation, an expression for the amplitude for multiphoton ionization by a nonperturbative field which has proved to be very useful for calculating (within the dipole approximation) the photoelectron energy spectrum of atomic hydrogen [2].

We also generalize the Keldysh approximation [3] to the amplitude for multiphoton detachment by a strong low-frequency field, and we apply this to the calculation of the angular distribution for multiphoton detachment of a model H^- ion. For sufficiently high intensities the angular distribution may be significantly shifted by the inclusion of the photon momentum; this can happen in the microwave region of frequencies (as demonstrated in Sec. IV) but not in the infrared frequency range since in the latter frequency range the intensity at which the photon momentum becomes significant is so high that the ions would be completely depleted by photodetachment at lower intensities on the rising edge of the pulse. The relevant parameter δ, which is a measure of the shift of the angular distribution, is defined by Eq. (86) in Sec. IV; this parameter scales with the intensity I and frequency ω as $I^{3/4}/\omega^2$. Recently, three experiments on multiphoton detachment of negative ions by strong laser light have been carried out: Angular distributions for the F^- ion have been measured by Blondel *et al.* [4], and total photodetachment rates for Cl^- and for Au^-, respectively, have been measured by Davidson *et al.* [5] and by Stapelfeldt *et al.* [6]. However, infrared light (1064-nm wavelength) was used in all three experiments and, as remarked above, in this regime the dipole approximation may be used without hesitation. We note that within the nonrelativistic framework the total (integrated) photodetachment rate should be unaffected by the inclusion of the photon momentum provided that the initial state is symmetric with respect to an inversion through the origin. (In a reference frame in which the atomic system is at rest, the total rate, integrated over angles, should be insensitive to a reversal in the direction of propagation of the light. Therefore the correction introduced by the inclusion of the photon momentum cannot depend on the sign of the speed of light c; this correction is quadratic in $1/c$, that is, relativistic.) However, we find that when the field is very strong the neglect of the *transverse* Doppler shift, but inclusion of the nonrelativistic Doppler shift, leads to a spuriously large rate for multiphoton detachment even though the photoelectron may be moving with a final speed v such that $(v/c)^2 \ll 1$, i.e., such that the difference between the relativistic and nonrelativistic rates is a very small (relativistic) correction. The reason for this peculiar discrepancy lies in the sensitivity of the rate to a small change in the frequency, as explained further in Sec. IV.

We leave open, at present, the question of whether the inclusion of the photon momentum can adversely effect the stabilization of an atom in the *high*-frequency, high-intensity, limit [7].

Corrections to the dipole approximation have already

been developed within the context of potential scattering in a laser field. Furthermore, the low-frequency limit of the scattering amplitude has been studied without making the dipole approximation, both treating the laser field quantum mechanically [8] and classically [9], the latter treatment being closest to the work presented in the present paper.

In the next section we present the formal development, in Sec. III we consider particular choices for the trial state vectors, and in Sec. IV we present the results of our application to H^-.

II. FORMAL DEVELOPMENT

A. Motion of free electron

We first examine the motion of a free electron, of charge e and mass μ, that is originally moving with velocity $\mathbf{v}$ and is overtaken by a plane-wave pulse which propagates along the z axis. The vector potential of the field of the pulse is $\mathbf{A}(\nu)$ where $\nu = t - (z/c)$, with t the time. We work within the Coulomb gauge, so that $\nabla_{\mathbf{r}} \cdot \mathbf{A}(\nu) = 0$ and hence $\mathbf{A}(\nu) \cdot \hat{\mathbf{z}} = 0$, where $\hat{\mathbf{z}}$ is a unit vector along the z axis. (We use a caret to denote a unit vector.) Neglecting relativistic corrections throughout this paper, we may solve Newton's equation $\mu d^2\mathbf{r}/dt^2 = -(e/c)\partial\mathbf{A}(\nu)/\partial t$ to give the classical position $\mathbf{r}$ of the electron: We have $\mathbf{r} = \mathbf{v}t + \bar{\boldsymbol{\alpha}}_{\mathbf{k}}(\nu)$, where $\mathbf{v} \equiv \hbar\mathbf{k}/\mu$ and where [10]

$$\bar{\boldsymbol{\alpha}}_{\mathbf{k}}(\nu) = \left(1 + \frac{v_z}{c}\right) \int_{-\infty}^{\nu} d\nu' \left(-\frac{e}{\mu c}\mathbf{A}(\nu') + \frac{e^2|\mathbf{A}(\nu')|^2}{2\mu^2 c^3}\hat{\mathbf{z}}\right), \tag{1}$$

where $v_z = \hat{\mathbf{z}} \cdot \mathbf{v}$. If $\Phi(t)$ is the scalar potential, which we take to be spatially constant, the interaction of the electron with the field is

$$V(t) = -\frac{e}{\mu c}\mathbf{A}(\nu) \cdot \mathbf{p} + \frac{e^2|\mathbf{A}(\nu')|^2}{2\mu c^2} + e\Phi(t'), \tag{2}$$

with $\mathbf{p}$ the canonical momentum operator. The change in the classical action of the electron due to the field is $\bar{S}_{\mathbf{k}} \equiv -\int^t dt'\ V(t')$, where, since the component of the canonical momentum perpendicular to $\hat{\mathbf{z}}$ commutes with $V(t)$ and is therefore a constant of the motion, we may replace $\mathbf{p}$ by $\hbar\mathbf{k}$ in $V(t')$. Performing the integral over t', using $z'/c = (v_z/c)t'$ (the perturbation to the rectilinear motion along the z axis is a correction of order $1/c$), and changing variables from t' to $\nu' = t' - z'/c = (1 - v_z/c)t'$, we obtain

$$\bar{S}_{\mathbf{k}} = -\hbar\mathbf{k}\cdot\bar{\boldsymbol{\alpha}}_{\mathbf{k}}(\nu) - \int_{-\infty}^{\nu} d\nu' \frac{e^2|\mathbf{A}(\nu')|^2}{2\mu c^2} - \int_{-\infty}^{t} dt'\, e\Phi(t'). \tag{3}$$

We now consider a pulse of extremely long duration, with frequency ω and an infinitesimal bandwidth. The vector potential is

$$\mathbf{A}(\nu) \equiv \mathrm{Re}(\mathbf{A}_0 e^{-i\omega\nu}), \tag{4}$$

where, ignoring the rise and fall of the pulse, $\mathbf{A}_0$ is constant. The polarization plane is the xy plane, and we can write

$$\mathbf{A}_0 = A_0[-i\cos(\zeta/2)\hat{\mathbf{x}} + \sin(\zeta/2)\hat{\mathbf{y}}], \tag{5}$$

where $A_0 = |\mathbf{A}_0|$ and where ζ is the ellipticity parameter ($|\zeta| \le \pi/2$). An electron which is at rest before the pulse arrives has, in the presence of the field, a cycle-average energy of $P = (e^2/4\mu c^2)A_0^2$. This "ponderomotive energy" transfer to the electron arises from the stimulated scattering of photons along the z axis during the rising edge of the pulse. Thus photons are scattered between occupied modes of slightly different frequencies within the very narrow bandwidth, resulting in a change in the net energy of the photons by P when the electron is initially at rest. Since the energy-momentum ratio for a photon is c, the energy transfer must be accompanied by a momentum transfer of $(P/c)\hat{\mathbf{z}}$. We incorporate the momentum transfer $(P/c)\hat{\mathbf{z}}$ into the total drift momentum $\hbar\mathbf{k}$ of the electron. Hence, the classical position of the electron is $\mathbf{r} = \mathbf{v}t + \boldsymbol{\alpha}_{\mathbf{k}}(\nu)$, where now $\mathbf{v} \equiv \hbar\mathbf{k}/\mu$ is the drift velocity of the electron inside the field and where

$$\boldsymbol{\alpha}_{\mathbf{k}}(\nu) = \bar{\boldsymbol{\alpha}}_{\mathbf{k}}(\nu) - (P/\mu c)t\hat{\mathbf{z}}. \tag{6}$$

We may reexpress $\boldsymbol{\alpha}_{\mathbf{k}}(\nu)$ as

$$\boldsymbol{\alpha}_{\mathbf{k}}(\nu) = (e/\mu c\omega_0)\mathrm{Im}(\mathbf{A}_0 e^{-i\omega\nu}) + (P/2\mu c\omega)\cos(\zeta)\sin(2\omega\nu)\hat{\mathbf{z}}, \tag{7}$$

with ω_0 the Doppler-shifted frequency in the frame that drifts with the electron:

$$\omega_0 = (1 - v_z/c)\omega. \tag{8}$$

Note that the energy transferred to a moving electron on the rising edge of the pulse is $(\omega_0/\omega)P$ since, in the rest frame of the electron, the photons that scatter from the electron have frequency ω_0. We are free to choose the scalar potential, subject to $\nabla_{\mathbf{r}}^2\Phi(t) = 0$, and it is convenient to set

$$\Phi(t) = -P/e, \tag{9}$$

so that $e\Phi$ cancels with P; this amounts simply to a shift in the zero reference energy, against which the electron's energy is just the drift energy $E \equiv |\hbar\mathbf{k}|^2/(2\mu)$.

In the absence of the field, the motion of an electron moving with momentum $\hbar\mathbf{k}$ is represented by $e^{-iEt/\hbar}|\mathbf{k}\rangle$, where $\langle\mathbf{r}|\mathbf{k}\rangle = (2\pi)^{-3/2}e^{i\mathbf{k}\cdot\mathbf{r}}$. The inclusion of the momentum transfer $(P/c)\hat{\mathbf{z}}$ into $\hbar\mathbf{k}$ results in the addition of the term $(P/c\hbar)(v_z t - z)$ to the phase $-Et/\hbar + \mathbf{k}\cdot\mathbf{r}$ of the free-particle wave function. We therefore introduce the effective change $S_{\mathbf{k}}(\nu)$ in the action due to the field

$$S_{\mathbf{k}}(\nu) = (P/c)(v_z t - z) + \bar{S}_{\mathbf{k}} \tag{10a}$$

$$= -\hbar\mathbf{k}\cdot\boldsymbol{\alpha}_{\mathbf{k}}(\nu) - (P/2\omega)\cos(\zeta)\sin(2\omega\nu) \tag{10b}$$

$$= \hbar\rho\sin(\omega\nu - \chi) - \hbar\beta\sin(2\omega\nu), \tag{10c}$$

where the dimensionless parameters ρ, χ, and β are defined by

$$\rho e^{i\chi} \equiv (e/\mu c\omega_0)\mathbf{k}\cdot\mathbf{A}_0, \tag{10d}$$

$$\beta \equiv (P/2\hbar\omega_0)\cos(\zeta), \tag{10e}$$

with ρ and β real and positive. In the presence of the field, an electron moving with *total* drift momentum $\hbar\mathbf{k}$ is represented by $e^{(i/\hbar)[-Et+S_{\mathbf{k}}(\nu)]}|\mathbf{k}\rangle$, which we can express as $e^{-iEt/\hbar}|\mathcal{F}_{0\mathbf{k}}(t)\rangle$, where

$$|\mathcal{F}_{0\mathbf{k}}(t)\rangle = e^{iS_{\mathbf{k}}(\nu)/\hbar}|\mathbf{k}\rangle. \tag{11}$$

An equivalent state vector of a free electron moving inside a monochroamtic plane-wave field has been derived by Rosenberg [8] and Ehlotzky [11]. We can replace the exponential factor on the right-hand side of Eq. (11) by the operator

$$\mathcal{O}(t) \equiv e^{(i/\hbar)S_{\mathbf{p}/\hbar}(\nu)}, \tag{12}$$

where z (in $\nu = t - z/c$) is now an operator. Note that we may commute $\mathbf{p}$ with ν in $S_{\mathbf{p}/\hbar}(\nu)$ since it is only the commutator of $p_z \equiv \hat{\mathbf{z}}\cdot\mathbf{p}$ with ν that does not vanish, and from Eq. (7) and (10b) we see that p_z appears divided by μc, so that the commutator is a relativistic correction. We have $|\mathcal{F}_{0\mathbf{k}}(t)\rangle = \mathcal{O}(t)|\mathbf{k}\rangle$, and the operator $\mathcal{O}(t)$ transforms the state vector of a free electron in the absence of the field into the state vector of the electron in the presence of the field; within the dipole approximation, $\mathcal{O}(t)$ is just the operator which accomplishes the transformation to the Kramers-Henneberger frame [12].

A free electron moving through the field can virtually absorb N photons, each having momentum $\hbar\mathbf{K}$, with $\mathbf{K} \equiv (\omega/c)\hat{\mathbf{z}}$. Thus $|\mathcal{F}_{0\mathbf{k}}(t)\rangle$, which is periodic in t, has the harmonic expansion

$$|\mathcal{F}_{0\mathbf{k}}(t)\rangle = \sum_N e^{-iN\omega t}|\mathcal{F}_{0\mathbf{k},N}\rangle, \tag{13a}$$

where, expanding $\exp[iS_{\mathbf{k}}(\nu)/\hbar]$ in the Fourier series $\sum_N \mathcal{J}_N(\rho,\chi,\beta)e^{-iN\omega\nu}$, we obtain

$$|\mathcal{F}_{0\mathbf{k},N}\rangle = \mathcal{J}_N(\rho,\chi,\beta)|\mathbf{k}+N\mathbf{K}\rangle, \tag{13b}$$

$$\mathcal{J}_N(\rho,\chi,\beta) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} d\nu\, e^{iN\omega\nu + iS_{\mathbf{k}}(\nu)/\hbar}. \tag{13c}$$

The coefficient $\mathcal{J}_N(\rho,\chi,\beta)$ is often called the generalized Bessel function, and some of its properties are summarized, for example, in Refs. [8, 13].

B. Boundary conditions

Suppose that an electron, traveling in a uniform monochromatic field, is incident with drift momentum $\hbar\mathbf{k}_i$ on a potential W which is *short range*, i.e., which falls off faster than $1/r$ with increasing distance r. The state vector of the electron may be expressed as $e^{-iE_it/\hbar}|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle$, where $E_i = |\hbar\mathbf{k}_i|^2/(2\mu)$; the "Floquet state vector" $|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle$ is periodic in t (the Floquet ansatz) and it therefore has the harmonic expansion

$$|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle = \sum_N e^{-iN\omega t}|\mathcal{F}^+_{\mathbf{k}_i,N}\rangle. \tag{14}$$

The Hamiltonian of the electron is $H(t) \equiv H_a + V(t)$, where H_a is the atomic Hamiltonian, consisting of the kinetic-energy operator plus W, and where $V(t)$ is the electron-field interaction, defined by Eq. (2) above. Defining

$$\mathcal{H}(t) \equiv H(t) - i\hbar\frac{d}{dt}, \tag{15}$$

the time-dependent Schrödinger equations for the state vectors $|\mathcal{F}_{0\mathbf{k}}(t)\rangle$ and $|\mathcal{F}^+_{\mathbf{k}}(t)\rangle$ are

$$\mathcal{H}(t)|\mathcal{F}_{0\mathbf{k}}(t)\rangle = (W+E)|\mathcal{F}_{0\mathbf{k}}(t)\rangle, \tag{16}$$

$$\mathcal{H}(t)|\mathcal{F}^+_{\mathbf{k}}(t)\rangle = E|\mathcal{F}^+_{\mathbf{k}}(t)\rangle. \tag{17}$$

The harmonic components $|\mathcal{F}^+_{\mathbf{k}_i,N}\rangle$ satisfy a set of coupled equations, which we can write down after first making a harmonic expansion of $V(t)$,

$$V(t) = {\sum_{M=-2}^{2}}' V_M e^{-iM\omega t}, \tag{18a}$$

where the prime on $\sum'$ signifies that the $M = 0$ (nonoscillatory) term is absent due to our addition of the scalar potential $-P/e$, and where $V_{-M} = V_M^\dagger$ with

$$V_1 = (e/2\mu c)e^{iKz}\mathbf{A}\cdot\mathbf{p}, \tag{18b}$$

$$V_2 = -(P/2)\cos(\zeta)e^{2iKz}. \tag{18c}$$

Substitution of the harmonic expansion Eq. (14) into Eq. (17), and use of Eq. (18a), gives the coupled equations

$$(E + N\hbar\omega - H_a)|\mathcal{F}^+_{\mathbf{k},N}\rangle = {\sum_{M=-2}^{2}}' V_M|\mathcal{F}^+_{\mathbf{k},N-M}\rangle. \tag{19}$$

We must supplement these equations with appropriate boundary conditions, which we now discuss.

Upon scattering from W the electron may absorb N real photons, and emerge with drift energy $E_f = E_i + N\hbar\omega$ and drift momentum $\mathbf{k}_f = k_N(E_i)\hat{\mathbf{r}}$, where $\hat{\mathbf{r}}$ points along the direction of observation, and where

$$k_N(E) = \sqrt{(2\mu/\hbar^2)(E+N\hbar\omega)}. \tag{20}$$

Since the electron is free from W at asymptotically large distances, and since $\mathcal{O}(t)^\dagger$ transforms the motion of a free electron moving in the field into the motion in the absence of the field, we have, for $r \sim \infty$,

$$\langle\mathbf{r}|\mathcal{O}(t)^\dagger|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle \sim (2\pi)^{-3/2}\sum_N e^{-iN\omega t}[\delta_{N0}e^{i\mathbf{k}_i\cdot\mathbf{r}} + f_N(\mathbf{k}_f,\mathbf{k}_i)e^{ik_N(E_i)r}/r], \tag{21}$$

where $f_N(\mathbf{k}_f,\mathbf{k}_i)$ is the amplitude for the electron to absorb N real photons and emerge with drift momentum $\hbar\mathbf{k}_f$.

From Eqs. (10b), (12), and (21) we can immediately write down the asymptotic form of $\langle \mathbf{r}|\mathcal{F}_{\mathbf{k}_i}(t)\rangle$; we have, for $r \sim \infty$,

$$\langle \mathbf{r}|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle \sim (2\pi)^{-3/2}\sum_N e^{-iN\omega t}[\delta_{N0}e^{iS_{\mathbf{k}_i}(\nu)/\hbar}e^{i\mathbf{k}_i\cdot\mathbf{r}} + f_N(\mathbf{k}_f,\mathbf{k}_i)e^{iS_{\mathbf{k}_f}(\nu)/\hbar}e^{ik_N(E_i)r}/r]. \tag{22}$$

It follows from Eqs. (14) and (22) that

$$\langle \mathbf{r}|\mathcal{F}^+_{\mathbf{k}_i,N}\rangle \sim (2\pi)^{-3/2}\left(\mathcal{J}_N(\rho_i,\chi_i,\beta_i)e^{iN\mathbf{K}\cdot\mathbf{r}}e^{i\mathbf{k}_i\cdot\mathbf{r}} + \sum_M f_{MN}(\mathbf{k}_f,\mathbf{k}_i)e^{i(N-M)\mathbf{K}\cdot\mathbf{r}}e^{ik_M(E_i)r}/r\right), \tag{23}$$

where, using subscripts i and f to indicate the values of the parameters ρ, χ, and β when $\mathbf{k}$ is either $\mathbf{k}_i$ or $\mathbf{k}_f$ in Eqs. (10d) and (10e) (recall β depends on $\mathbf{k}$ through the Doppler shift),

$$f_{MN}(\mathbf{k}_f,\mathbf{k}_i) = \mathcal{J}_{N-M}(\rho_f,\chi_f,\beta_f)f_M(\mathbf{k}_f,\mathbf{k}_i). \tag{24}$$

We may interpret $f_{MN}(\mathbf{k}_f,\mathbf{k}_i)$ as the amplitude for the electron to absorb N photons, of which only M are real photons. The amplitude $f_M(\mathbf{k}_f,\mathbf{k}_i)$, which is the amplitude for absorption of M real photons, is a coherent sum of the $f_{MN}(\mathbf{k}_f,\mathbf{k}_i)$. Using the sum rule [8]

$$\sum_M \mathcal{J}^*_M(\rho,\chi,\beta)\mathcal{J}_{M+N}(\rho',\chi',\beta') = \mathcal{J}_N(\rho'-\rho,\chi'-\chi,\beta'-\beta), \tag{25}$$

we have, from Eq. (24),

$$f_N(\mathbf{k}_f,\mathbf{k}_i) = \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)f_{N,N+M}(\mathbf{k}_f,\mathbf{k}_i). \tag{26}$$

The differential cross section for the electron to absorb N photons and emerge into a solid angle $d\Omega_{\mathbf{k}_f}$ along $\mathbf{k}_f$ is

$$\frac{d\sigma_N}{d\Omega_{\mathbf{k}_f}} = \frac{k_f}{k_i}|f_N(\mathbf{k}_f,\mathbf{k}_i)|^2, \tag{27}$$

with $E_f = E_i + N\hbar\omega$. The superscript + attached to $|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle$ signifies boundary conditions such that the electron enters the collision with a well-defined drift momentum $\hbar\mathbf{k}_i$ and emerges in a state represented by a superposition of outgoing scattered waves. We could specify boundary conditions for which the electron emerges with a well-defined drift momentum $\hbar\mathbf{k}_f$, while it enters the collision in a state represented by a superposition of ingoing waves; as usual, we indicate such boundary conditions by a superscript $-$, the appropriate Floquet state vector being $|\mathcal{F}^-_{\mathbf{k}_f}(t)\rangle$.

C. Scattering amplitude

The scattering amplitude may be written as

$$f_N(\mathbf{k}_f,\mathbf{k}_i) = -(2\pi)^2(\mu/\hbar^2)T_N(\mathbf{k}_f,\mathbf{k}_i), \tag{28}$$

where the N-photon T-matrix is [8]

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle \mathcal{F}_{0\mathbf{k}_f}(t)|W|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle \tag{29}$$

$$= \sum_M \langle \mathcal{F}_{0\mathbf{k}_f,M}|W|\mathcal{F}^+_{\mathbf{k}_i,M+N}\rangle. \tag{30}$$

Using Eq. (16) we can replace $W|\mathcal{F}_{0\mathbf{k}_f}(t)\rangle$ by $[\mathcal{H}(t) - E_f]|\mathcal{F}_{0\mathbf{k}_f}(t)\rangle$, and therefore we can rewrite Eq. (29) as

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle \mathcal{F}_{0\mathbf{k}_f}(t)|\left[\mathcal{H}(t)^\dagger - E_f\right]|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle, \tag{31}$$

where here and below it is to be understood that $\langle\phi|\mathcal{H}(t)^\dagger|\psi\rangle = [\langle\psi|\mathcal{H}(t)|\phi\rangle]^*$, with the time derivative not acting beyond the ket. Integrating the term in the time derivative by parts on the right-hand side of Eq. (31), noting that the surface term vanishes due to the periodicity of the integrand, and using Eq. (17), we arrive at

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle \mathcal{F}_{0\mathbf{k}_f}(t)|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle. \tag{32}$$

Note that $H(t)$ is not Hermitian when acting between the ket and bra in the matrix element on the right-hand side of Eq. (32); if it were, this matrix element would vanish. The non-Hermitian part of $H(t)$ is the kinetic-energy operator. The potential W is Hermitian, and so is $V(t)$ even though $V(t)$ contains the momentum operator. [In contrast to the kinetic energy operator, which is isotropic, $V(t)$ includes only one component of the momentum operator, the component along the polarization axis, and as a result $V(t)$ is Hermitian. This can be seen explicitly by evaluating the surface term—it vanishes on the surface at infinity.]

Since we cannot, in general, calculate the Floquet state vectors $|\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle$ and $|\mathcal{F}^-_{\mathbf{k}_f}(t)\rangle$ exactly, we must approxi-

mate them by trial Floquet state vectors, $|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle$ and $|\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)\rangle$, respectively; we assume that these trial approximations are periodic and satisfy the correct asymptotic boundary conditions in space and time. We now derive a *variational* estimate of $T_N(\mathbf{k}_f,\mathbf{k}_i)$, that is, an estimate whose error is of second order in the errors of the trial vectors. We denote the error in $|\mathcal{F}^{\pm}_{\mathbf{k},\mathrm{tr}}(t)\rangle$ by $|\delta\mathcal{F}^{\pm}_{\mathbf{k},\mathrm{tr}}(t)\rangle \equiv |\mathcal{F}^{\pm}_{\mathbf{k}}(t)\rangle - |\mathcal{F}^{\pm}_{\mathbf{k},\mathrm{tr}}(t)\rangle$. We rewrite Eq. (32) as

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}_{0\mathbf{k}_f}(t)|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle + T_N^{(1)}(\mathbf{k}_f,\mathbf{k}_i), \tag{33}$$

$$T_N^{(1)}(\mathbf{k}_f,\mathbf{k}_i) \equiv \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}_{0\mathbf{k}_f}(t)|\left[H(t)^\dagger - H(t)\right]|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle. \tag{34}$$

The superscript 1 on the remainder $T_N^{(1)}(\mathbf{k}_f,\mathbf{k}_i)$ serves to indicate that this remainder is of *first* order in the error in the initial trial Floquet state vector. We use the following identities:

$$[\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)| - \langle\mathcal{F}_{0\mathbf{k}_f}(t)|][H(t)^\dagger - H(t)]|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle = 0, \tag{35}$$

$$[\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)| - \langle\mathcal{F}_{0\mathbf{k}_f}(t)|][H(t)^\dagger - H(t)][|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle - |\mathcal{F}_{0\mathbf{k}_i}(t)\rangle] = 0. \tag{36}$$

Equations (35) and (36) may be proved by using Green's theorem to convert the volume integrals to surface integrals, noting that the surface integrals vanish since the surface integrands oscillate (infinitely rapidly for $r \sim \infty$). We first use Eq. (35) to replace the bra $\langle\mathcal{F}_{0\mathbf{k}_f}(t)|$ in Eq. (34) by $\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|$. It follows that

$$T_N^{(1)}(\mathbf{k}_f,\mathbf{k}_i) = -\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[H(t)^\dagger - H(t)\right]|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle \tag{37}$$

$$= -\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\{\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_f\right]^\dagger|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle - \langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_i\right)]|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle\}. \tag{38}$$

The equivalence of Eqs. (37) and (38) follows after integrating by parts the time derivative in $\mathcal{H}(t)^\dagger$ in the first term on the right-hand side of Eq. (38). This term is

$$T_{N,\mathrm{err}}(\mathbf{k}_f,\mathbf{k}_i) \equiv -\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_f\right]^\dagger|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle$$

$$= -\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\delta\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_f\right]^\dagger|\delta\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle, \tag{39}$$

where in the second step we used $[\mathcal{H}(t) - E_f]|\mathcal{F}^{-}_{\mathbf{k}_f}(t)\rangle = 0$. We see that $T_{N,\mathrm{err}}(\mathbf{k}_f,\mathbf{k}_i)$ is of *second* order in the errors in the trial Floquet state vectors. Combining Eqs. (33), (38), and (39) yields

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = T_{N,\mathrm{var}}(\mathbf{k}_f,\mathbf{k}_i) + T_{N,\mathrm{err}}(\mathbf{k}_f,\mathbf{k}_i), \tag{40}$$

$$T_{N,\mathrm{var}}(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\{\langle\mathcal{F}_{0\mathbf{k}_f}(t)|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle + \langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_i\right]|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle\}. \tag{41}$$

Since $T_{N,\mathrm{err}}(\mathbf{k}_f,\mathbf{k}_i)$ is of second-order smallness, $T_{N,\mathrm{var}}(\mathbf{k}_f,\mathbf{k}_i)$ is a variational estimate (stationary with respect to variations of the trial state vectors about the exact ones). We can obtain an alternative expression for the variational estimate by first integrating by parts the time derivative in $\mathcal{H}(t)$ in the second term in square brackets on the right-hand side of Eq. (41); in doing so we encounter

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}[\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)| - \langle\mathcal{F}_{0\mathbf{k}_f}(t)|]|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}^{+}_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle$$

$$= \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}[\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)| - \langle\mathcal{F}_{0\mathbf{k}_f}(t)|]|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle$$

$$= \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}^{-}_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle, \tag{42}$$

where in the second step we used Eq. (36) and where in the third step we used

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}_{0\mathbf{k}_f}(t)|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle$$

$$= \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle\mathcal{F}_{0\mathbf{k}_f}(t)|\{[\mathcal{H}(t) - W - E_f]^\dagger - [\mathcal{H}(t) - W - E_i]\}|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle = 0, \quad (43)$$

recalling Eq. (16). We thereby arrive at

$$T_{N,\mathrm{var}}(\mathbf{k}_f,\mathbf{k}_i) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\{\langle\mathcal{F}^-_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[H(t) - H(t)^\dagger\right]|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle + \langle\mathcal{F}^-_{\mathbf{k}_f,\mathrm{tr}}(t)|\left[\mathcal{H}(t) - E_f\right]^\dagger|\mathcal{F}^+_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle\}. \quad (44)$$

It can be shown that the variational principle embodied by Eqs. (40), (41), and (44) is equivalent to the variational principle derived and used by Rosenberg and coworkers [14].

III. APPROXIMATIONS

A. Simplified approximation to the inelastic T matrix

We now derive a useful approximation to the T matrix for inelastic scattering or for ionization (i.e., $N \neq 0$). We insert into Eq. (44) the following trial vector:

$$|\mathcal{F}^-_{\mathbf{k}_f,\mathrm{tr}}(t)\rangle = \sum_M e^{-iM\omega t}\mathcal{J}_M(\rho_f,\chi_f,\beta_f)|\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\rangle, \quad (45)$$

where $|\Phi^-_{\mathbf{k}}\rangle$ represents an electron which emerges with momentum $\hbar\mathbf{k}$ after scattering from the potential W *in the absence of the field.* (While we continue to assume for the moment that W is short range, we can, in fact, allow W to become long range in the final expressions developed below.) The trial vector given in Eq. (45) is a generalization of a form that has often been used within the dipole approximation—see, e.g., Ref. [15]—and a form not too dissimilar from this (the difference is in the replacement of $|\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\rangle$ by $|\Phi^-_{\mathbf{k}_f}\rangle$) has also been used in going beyond the dipole approximation [1]. The trial vector of Eq. (45) satisfies the correct boundary condition, since $\langle\mathbf{r}|\mathcal{F}^-_{\mathbf{k}_f,\mathrm{tr}}(t)\rangle$ approaches $\langle\mathbf{r}|\mathcal{F}_{0\mathbf{k}_f}(t)\rangle$ for large r. Provided that $N \neq 0$, so that $|\mathbf{k}_i| \neq |\mathbf{k}_f|$, the scalar product

$$\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}|\left[H(t)^\dagger - H(t)\right]|\mathcal{F}_{0\mathbf{k}_i}(t)\rangle$$

vanishes, since it may be converted into a surface integral whose integrand oscillates. Hence, putting $|\mathcal{F}^+_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle = |\mathcal{F}^+_{\mathbf{k}_i}(t)\rangle$ in Eq. (44) gives, for $N \neq 0$,

$$\begin{aligned}
T_N(\mathbf{k}_f,\mathbf{k}_i) &= \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\left[\left\langle\mathcal{F}^+_{\mathbf{k}_i}(t)\middle|\left(V(t) - i\hbar\frac{d}{dt} + M\frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)e^{-iM\omega t}\middle|\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\right\rangle\right]^* \\
&= \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{i(N+M)\omega t}\left\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\middle|\left(N\hbar\omega + V(t)^\dagger - i\hbar\frac{d}{dt} + M\frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)\middle|\mathcal{F}^+_{\mathbf{k}_i}(t)\right\rangle \\
&= \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{i(N+M)\omega t}\left\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\middle|\left(E_i + N\hbar\omega - H_a + M\frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)\middle|\mathcal{F}^+_{\mathbf{k}_i}(t)\right\rangle \\
&= \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)\left\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\middle|\left(E_i + N\hbar\omega - H_a + M\frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)\middle|\mathcal{F}^+_{\mathbf{k}_i,M+N}\right\rangle, \quad (46)
\end{aligned}$$

where in arriving at the second step we integrated by parts the term in the time derivative and where in the third step we used the Hermiticity of $V(t)$ and Eq. (17). It follows that

$$T_N(\mathbf{k}_f,\mathbf{k}_i) = \sum_M \mathcal{J}^*_M(\rho_f,\chi_f,\beta_f)T_{N,N+M}(\mathbf{k}_f,\mathbf{k}_i), \quad (47)$$

$$T_{N,N+M}(\mathbf{k}_f,\mathbf{k}_i) = \left\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\middle|\left(E_i + N\hbar\omega - H_a + M\frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)\middle|\mathcal{F}^+_{\mathbf{k}_i,M+N}\right\rangle \quad (48)$$

$$= -M\left\langle\Phi^-_{\mathbf{k}_f+M\mathbf{K}}\middle|\left(1 - \frac{\hbar^2}{\mu}\mathbf{k}_f\cdot\mathbf{K}\right)\middle|\mathcal{F}^+_{\mathbf{k}_i,M+N}\right\rangle + \sum_{L=-2}^{2}{}'\langle\Phi^-_{\mathbf{k}_f}|V_L|\mathcal{F}^+_{\mathbf{k},M+N-L}\rangle, \quad (49)$$

where in arriving at the last step we used Eq. (19). Since the T matrix differs from the scattering amplitude only by a constant factor, comparison of Eq. (47) with Eqs. (24) and (26) leads to the identification

$$T_{NM}(\mathbf{k}_f,\mathbf{k}_i)=\mathcal{J}_{M-N}(\rho_f,\chi_f,\beta_f)T_N(\mathbf{k}_f,\mathbf{k}_i). \tag{50}$$

Now from Eq. (49) we have

$$T_{NN}(\mathbf{k}_f,\mathbf{k}_i)=\sum_{L=-2}^{2}{}'\langle\Phi^-_{\mathbf{k}_f}|V_L|\mathcal{F}^+_{\mathbf{k}_i,N-L}\rangle, \tag{51}$$

and therefore using Eq. (50) we arrive at

$$T_N(\mathbf{k}_f,\mathbf{k}_i)=\left(\sum_{L=-2}^{2}{}'\langle\Phi^-_{\mathbf{k}_f}|V_L|\mathcal{F}^+_{\mathbf{k}_i,N-L}\rangle\right)\Big/\mathcal{J}_0(\rho_f,\chi_f,\beta_f),\quad N\neq 0. \tag{52}$$

The expression given by Eqs. (47) and (49), and also the expression given by Eq. (52), hold *even when W has a Coulomb tail*, since the effect of the Coulomb tail is included in $|\Phi^-_{\mathbf{k}_f}\rangle$. Both of these expressions would be exact if $|\mathcal{F}^+_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle$ were chosen to be the exact $|\mathcal{F}^+_{\mathbf{k}_i}\rangle$. In practice, of course, the infinite set of coupled equations (19) must be truncated, and hence the harmonic components $|\mathcal{F}^+_{\mathbf{k}_i,M}\rangle$ appearing in Eqs. (49) and (52) are inexact. Nevertheless, since we started from a variational expression, the error in the expression given by Eqs. (47) and (49) is of second order. However, in arriving at the expression given by Eq. (52), we supplemented the variational principle by Eq. (50), and the latter identity has a first-order error when the harmonic components $|\mathcal{F}^+_{\mathbf{k}_i,M}\rangle$ are inexact, so that presumably the error in Eq. (52) is of first order. A different derivation of these results, within the dipole approximation, was given earlier by Potvliege and Shakeshaft [16] and these expressions were applied to the calculation of the photoelectron energy and angular distributions of atomic hydrogen in a strong field [2]. In the dipole approximation we put $\mathbf{K}=\mathbf{0}$, and in this approximation the harmonic components $V_{\pm 2}$ of $V(t)$ are constants and therefore have no physical effect and may be transformed away; therefore the sum over L in Eqs. (49) and (52) collapses from four to two terms.

We note that several different calculations of the amplitude for the scattering of an electron from a Coulomb potential in the presence of a monochromatic field have been carried out within the dipole approximation [17]. These calculations were performed in the Kramers-Henneberger frame, based on an expression for the T matrix which involves the matrix elements of the (space-translated) atomic potential W rather than matrix elements of the interaction of the electron with the radiation field.

B. Generalized Keldysh approximation

In this section we generalize, beyond the dipole approximation, the Keldysh approximation to the amplitude for ionization by low-frequency light. To formulate the Keldysh approximation we must first consider a transformation to another gauge.

In the Coulomb gauge we have $\nabla_{\mathbf{r}}\cdot\mathbf{A}=0$, but the vector and scalar potentials, $\mathbf{A}$ and Φ, are not uniquely fixed by this requirement. We have been working in a particular subgauge, the "velocity" gauge, in which Φ is spatially constant; we set $\Phi=-P/e$. Another widely used subgauge of the Coulomb gauge is the "length" gauge in which the vector potential $\mathbf{A}'$ is a small quantity (the square of $e\mathbf{A}'/c$ is a relativistic correction) but in which the scalar potential Φ' is spatially dependent. We have $\mathbf{A}'=\mathbf{A}+\nabla_{\mathbf{r}}\Lambda$ and $\Phi'=\Phi-(1/c)\partial\Lambda/\partial t$, where

$$\Lambda(t)=-\mathbf{A}(\nu)\cdot\mathbf{r}-(cP/e)t. \tag{53}$$

Thus, in the length gauge the vector and scalar potentials are, introducing the electric-field vector $\mathbf{F}\equiv-(1/c)\partial\mathbf{A}/\partial t=\partial\mathbf{A}/\partial z$ and recalling that $\Phi=-P/e$,

$$\mathbf{A}'=-(\mathbf{F}\cdot\mathbf{r})\hat{\mathbf{K}}, \tag{54}$$

$$\Phi'=-\mathbf{F}\cdot\mathbf{r}. \tag{55}$$

Hence, in the length gauge the interaction of an electron with a monochromatic plane-wave field is, defining the electric-field amplitude $\mathbf{F}_0$ by $\mathbf{F}\equiv\mathrm{Re}\mathbf{F}_0e^{-i\omega\nu}$ and recalling that the square of $e\mathbf{A}'/c$ is a relativistic correction,

$$V'(t)=Ue^{-i\omega\nu}+U^\dagger e^{i\omega\nu}, \tag{56}$$

$$U=-(e/2)(\mathbf{F}_0\cdot\mathbf{r})\left(1+\frac{\hat{\mathbf{K}}\cdot\mathbf{p}}{\mu c}\right). \tag{57}$$

In the velocity gauge the field is coupled predominantly to the electric current of the electron, while in the length gauge the field is coupled predominantly to the electric dipole of the electron. For a low-frequency field, the characteristic value of $(\mathbf{F}_0\cdot\mathbf{r})$, for a tightly *bound* electron, is considerably smaller than the characteristic value of $(\mathbf{A}_0\cdot\mathbf{p})/(\mu c)$. Provided that $|e\mathbf{F}_0|$ is small compared to the electric-field force exerted by the atomic nucleus, $(\mathbf{F}_0\cdot\mathbf{r})$ may be regarded as a weak perturbation, and hence, provided that we use the length gauge, the initial bound state can be represented well by $e^{-iE_i^{(0)}t/\hbar}|\Phi_i\rangle$, where $|\Phi_i\rangle$ and $E_i^{(0)}$ are the initial *un*perturbed bound-state vector and eigenvalue. However, we are using the velocity gauge; the equivalent state vector in this gauge is obtained by multiplying $e^{-iE_i^{(0)}t/\hbar}|\Phi_i\rangle$ by the unitary operator $e^{-i(e/\hbar c)\Lambda(t)}$ [18]. Therefore, in the spirit of the Keldysh approximation [3], we insert into Eq. (44) the initial-state trial vector

$$|\mathcal{F}^+_{\mathbf{k}_i,\mathrm{tr}}(t)\rangle=e^{-i\mathbf{k}_A(\nu)\cdot\mathbf{r}}|\Phi_i\rangle, \tag{58}$$

where $\hbar\mathbf{k}_A(\nu)\equiv-(e/c)\mathbf{A}(\nu)$ and where we have extracted $-(cP/e)t$ from $\Lambda(t)$ since this term contributes

an energy shift $-P$ to the initial bound-state energy to give a perturbed bound-state energy of $E_i = E_i^{(0)} - P$. Note that to obtain a nonzero amplitude for ionization we require that at least $N_{\min}$ photons be absorbed, where $N_{\min}$ is the smallest value of N for which $E_i + N\hbar\omega > 0$. Inserting $|\mathcal{F}^-_{\mathbf{k}_f,\mathrm{tr}}(t)\rangle = |\mathcal{F}_{0\mathbf{k}_f}(t)\rangle$ into Eq. (44), and writing $M_{i,N}(\mathbf{k}_f)$ in place of $T_N(\mathbf{k}_f, \mathbf{k}_i)$, with it understood that $N \geq N_{\min}$, we obtain

$$M_{i,N}(\mathbf{k}_f) = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle \mathcal{F}_{0\mathbf{k}_f}(t)|We^{-i\mathbf{k}_A(\nu)\cdot\mathbf{r}}|\Phi_i\rangle \tag{59}$$

$$= \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\omega t}\langle \mathbf{k}_f|e^{-iS_{\mathbf{k}_f}(\nu)/\hbar - i\mathbf{k}_A(\nu)\cdot\mathbf{r}}W|\Phi_i\rangle. \tag{60}$$

The differential rate for the electron to absorb N photons and emerge with drift momentum $\hbar\mathbf{k}_f$ into a solid angle $d\Omega_{\mathbf{k}_f}$ is

$$\frac{d\Gamma_N}{d\Omega_{\mathbf{k}_f}} = \frac{2\pi}{\hbar}\frac{\mu k_f}{\hbar^2}|M_{i,N}(\mathbf{k}_f)|^2, \tag{61}$$

with $E_f = E_i + N\hbar\omega$.

Assuming that $\hbar\omega$ is small compared to $|E_i^{(0)} - P|$, the minimum number $N_{\min}$ of photons that the atom must absorb to ionize is large compared to unity. Therefore $N \gg 1$ and $\exp[iN\omega t - iS_{\mathbf{k}_f}(\nu)/\hbar]$ oscillates rapidly over the range of t integration. Hence the main contribution to the integral over t comes from the point of stationary phase. This point, which is given by the equation $N\hbar\omega - \partial S_{\mathbf{k}_f}(\nu)/\partial t = 0$, depends weakly on z. However, to gain further insight we temporarily neglect the z dependence and thereby we obtain a constant point of stationary phase t_s, where

$$N\hbar\omega_t - \frac{dS_{\mathbf{k}_f}(t)}{dt} = 0, \quad t = t_s. \tag{62}$$

Noting that

$$\frac{dS_{\mathbf{k}_f}(\nu)}{dt} = E_f + P - \frac{\hbar^2}{2\mu}|\mathbf{k}_f + \mathbf{k}_A(\nu)|^2, \tag{63}$$

and using $E_f = E_i^{(0)} - P + N\hbar\omega$, we can rewrite Eq. (62) as

$$\frac{\hbar^2}{2\mu}|\mathbf{k}_f + \mathbf{k}_A(\nu)|^2 - E_i^{(0)} = 0, \quad t = t_s. \tag{64}$$

The two terms on the left-hand side of Eq. (64) are positive for real t. Hence the point t_s is not in fact a point of stationary phase but rather a saddle point in the complex t plane. (Keldysh, within the dipole approximation, evaluated the integral over t using the method of steepest descent.) We now proceed to turn the expression of Eq. (60) for $M_{i,N}(\mathbf{k}_f)$ into a form more suitable for computation.

Since the component of $\mathbf{r}$ along $\mathbf{k}_A(\nu)$ is independent of ν we may write

$$e^{iS_{\mathbf{k}}(\nu)/\hbar + i\mathbf{k}_A(\nu)\cdot\mathbf{r}} = \sum_N e^{-iN\omega\nu}\frac{\omega}{2\pi}\int_0^{2\pi/\omega} d\nu'\, e^{iN\omega\nu' + iS_{\mathbf{k}}(\nu')/\hbar + i\mathbf{k}_A(\nu')\cdot\mathbf{r}}, \tag{65}$$

and hence we obtain

$$e^{iS_{\mathbf{k}}(\nu)/\hbar + i\mathbf{k}_A(\nu)\cdot\mathbf{r}}|\mathbf{k}_f\rangle = \sum_N e^{-iN\omega t}|\mathcal{F}'_{0\mathbf{k},N}\rangle, \tag{66}$$

$$|\mathcal{F}'_{0\mathbf{k},N}\rangle = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} d\nu\, e^{iN\omega\nu + iS_{\mathbf{k}}(\nu)/\hbar}|\mathbf{k} + N\mathbf{K} + \mathbf{k}_A(\nu)\rangle \tag{67}$$

$$= \sum_M \mathcal{J}_{N-M}(\rho, \chi, \beta)\frac{\omega}{2\pi}\int_0^{2\pi/\omega} d\nu\, e^{iM\omega\nu}|\mathbf{k} + N\mathbf{K} + \mathbf{k}_A(\nu)\rangle. \tag{68}$$

Using Eqs. (66) and (68) in Eq. (60), and integrating over t, yields

$$M_{i,N}(\mathbf{k}_f) = \sum_M \mathcal{J}^*_{-N-M}(\rho_f, \chi_f, \beta_f)\frac{\omega}{2\pi}\int_0^{2\pi/\omega} d\nu\, e^{-iM\omega\nu}\langle \mathbf{k}_f - N\mathbf{K} + \mathbf{k}_A(\nu)|W|\Phi_i\rangle. \tag{69}$$

Equation (69) is a convenient expression for computation of $M_{i,N}(\mathbf{k}_f)$ since the number of terms which contribute to the sum over M is limited by the fact that the integral over ν decreases rapidly with increasing M due to the integrand being oscillatory. Note that since the matrix element $\langle \mathbf{k}|W|\Phi_i\rangle$ decreases rapidly with increasing $|\mathbf{k}|$, the photoelectron energy distribution will peak at the values of E_f for which

$$\mathbf{k}_f \approx N\mathbf{K} - \mathbf{k}_A(\nu), \tag{70}$$

with $\nu = (1 - v_z/c)t_s$.

IV. MULTIPHOTON DETACHMENT OF H^-

In this section we examine the angular distribution for the multiphoton detachment of a model H^- ion. We treat H^- as a one-electron system, with the electron moving in a spherical potential $W(r)$. We consider two forms for $W(r)$.

A. Zero-range potential

A very simple model, first analyzed by Berson [19] and Manakov and Rapoport [20], is obtained by choosing

$$W(r) = \frac{2\pi}{\kappa}\delta^3(\mathbf{r})\frac{\partial}{\partial r}r. \tag{71}$$

This potential supports one bound state, whose energy is $\frac{-\kappa^2}{2}$ and whose wave function is

$$\langle \mathbf{r}|\Phi_i\rangle = \sqrt{\frac{\kappa}{2\pi}}\ \frac{e^{-\kappa r}}{r}. \tag{72}$$

We may adjust κ so that the binding energy is equal to the electron affinity of H^-, and the model has been successfully applied to a study of multiphoton detachment of H^- by Becker, McIver, and Confer [21]. Since the form of W given by Eq. (71) is nonlocal, we use Eq. (59), which becomes

$$M_{i,N}(\mathbf{k}_f) = -\sqrt{2\pi\kappa}\ \mathcal{J}^*_{-N}(\rho_f, \chi_f, \beta_f). \tag{73}$$

Hence, in this zero-range-potential model, the matrix element for multiphoton detachment differs from the dipole-approximation result only by the Doppler shift, which is present in ρ_f and β_f. We see that the sum over M on the right-hand side of Eq. (69) collapses to a single term, but this is not an enormous simplification since, as already observed, the number of terms in the sum which contribute significantly is relatively small.

B. Yukawa potential

Rather than continue our analysis with the zero-range potential, we pursue a one-electron model which may be somewhat more realistic; we choose the finite-range potential

$$W(r) = -ae^{-br}/r, \tag{74}$$

where $b = 1$ a.u. and $a = -1.1$ a.u. This potential also supports only one bound state, its binding energy is 0.027 565, very close to the electron affinity of H^-, and the bound-state probability distribution is close to the exact two-electron distribution integrated over one of the electron coordinates. We determined the bound-state wave function by solving the Schrödinger equation on a basis set (composed of radial Sturmian functions).

We have calculated rates for the multiphoton detachment of an electron bound to this potential when the light is *circularly* polarized (i.e., $\zeta = \pi/2$). We evaluated $M_{i,N}(\mathbf{k}_f)$ using Eq. (69); noting that $\zeta = \pi/2$ we have $\beta_f = 0$ and therefore $\mathcal{J}_N(\rho_f, \chi_f, \beta_f) = e^{iN\chi_f}J_N(\rho_f)$, where $J_N(\rho)$ is the standard Bessel function and where $\chi_f = \pi/2 \pm \phi$ with ϕ the azimuthal angle of $\mathbf{k}_f$ relative to the x axis. The photodetachment angular distribution is azimuthally symmetric, and therefore without loss in generality we may take $\chi_f = 0$. If θ is the angle which $\mathbf{k}_f$ makes with the z axis (the direction of propagation of the laser field) we have

$$\rho_f = |(eA_0/\mu c\omega_0)|k_f \sin(\theta), \tag{75}$$

where $|(e/c)A_0| = \sqrt{2\mu P}$. Since (for circular polarization) $|\mathbf{A}(\nu)| = A_0$, we see from Eq. (70) that the photoelectron energy distribution peaks [22, 23] at $E_f \approx P$, which corresponds to $N\hbar\omega \approx 2P$ since $E_i \approx -P$. For this value of E_f we have $\rho_f \approx N$ if $\mathbf{k}_f$ lies in the polarization plane, i.e., if $\theta = \pi/2$. The precise value of ρ_f depends on the angle $\pi/2 - \theta$ which $\mathbf{k}_f$ makes with the polarization plane; from Eq. (75) we have

$$\rho_f = \rho_0(1 - u^2)^{1/2}(1 - vu/c)^{-1}, \tag{76}$$

where $u = \cos(\theta)$, where $v = \hbar k_f/\mu$ (the final speed of the photoelectron), and where

$$\rho_0 \equiv \sqrt{2\mu P}\ k_f/(\mu\omega), \tag{77}$$

i.e., ρ_0 is the value of ρ_f when $u = 0$ (noting $\omega_0 = \omega$ when $u = 0$). If we define N_0 by $P \equiv N_0\hbar\omega$, and if we write $N = 2N_0 + \Delta N$ with $\Delta N \ll N$, we have

$$\rho_0 = \left[[N^2 - (\Delta N)^2]\left(1 + \frac{2E_i^{(0)}}{(N + \Delta N)\hbar\omega}\right)\right]^{1/2} \tag{78}$$

$$\approx N - |E_i^{(0)}|/(\hbar\omega), \tag{79}$$

where we have neglected terms of order $\Delta N/N$. It follows that for $|u| \ll 1$ we have

$$\rho_f \approx (1 + vu/c)\left(N - \frac{|E_i^{(0)}|}{\hbar\omega} - \frac{Nu^2}{2}\right). \tag{80}$$

To obtain some idea as to the shape of the angular distribution, we observe from Eq. (69) that the angular distribution will be determined largely by the form of $J_N(\rho_f)$. Using the Debye approximation

$$J_N(N + N^{1/3}z) \approx (2/N)^{1/3}\mathrm{Ai}(-2^{1/3}z), \tag{81}$$

and the asymptotic form of the Airy function, we have

$$J_N(N + N^{1/3}z)$$

$$\approx (2/N)^{1/3}e^{-(2/3)(-2^{1/3}z)^{3/2}}/[2\sqrt{2}(-2^{1/3}z)^{1/4}], \tag{82}$$

and if we neglect the Doppler shift we have, from Eq. (80), that $\rho_f \approx N + N^{1/3}z$ with

$$z = -|E_i^{(0)}|/(N^{1/3}\hbar\omega) - N^{2/3}u^2/2. \tag{83}$$

Hence, if $u^2 \ll 2|E_i^{(0)}|/(N\hbar\omega)$ we have

$$-z^{3/2} \approx \left(\frac{|E_i^{(0)}|}{N^{1/3}\hbar\omega}\right)^{3/2} + \frac{3}{4}\left(\frac{N|E_i^{(0)}|}{\hbar\omega}\right)^{1/2} u^2. \tag{84}$$

Therefore we expect the angular distribution to be a Gaussian in $u = \cos(\theta)$, peaked at $u = 0$ (when the Doppler shift is neglected) with a full width at half maximum (FWHM) of

$$\Delta u \equiv 2(\hbar\omega/2N|E_i^{(0)}|)^{1/4}\sqrt{\ln(2)}. \tag{85}$$

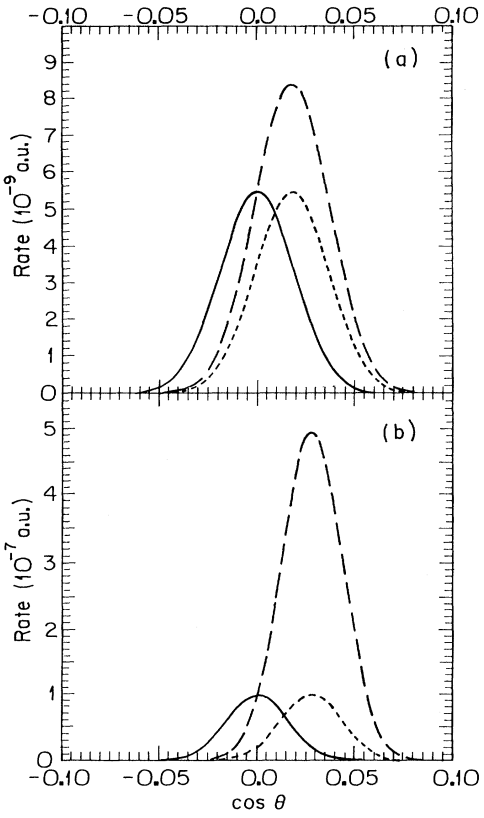

FIG. 1. Angular distribution for N-photon electron detachment of H^- (within our Yukawa potential model) by a circularly polarized laser field of frequency $\omega = 0.000\,44$ a.u. The angle θ is measured with respect to the direction of propagation of the laser beam. The solid line (which is symmetric with respect to reflections in the polarization plane) corresponds to the dipole approximation. The two dashed lines correspond to inclusion of the photon momentum. The long dashed line results from using the nonrelativistic Doppler shift, the short one from using the relativistic Doppler shift. (a) $I = 0.82 \times 10^{11}$ W/cm^2 ($P = 3.0$ a.u.) and $N = 13\,760$. (b) $I = 2.0 \times 10^{11}$ W/cm^2 ($P = 7.3589$ a.u.) and $N = 33\,576$. In both cases (a) and (b) the number N of absorbed photons is $N = 2(P + |E_i^{(0)}|)/\hbar\omega$.

We now describe our results.

In Fig. 1 we show the angular distribution $d\Gamma_N/d\Omega_{\mathbf{k}_f}$ for $N = 2(P + |E_i^{(0)}|)/(\hbar\omega)$ (so $E_f = P + |E_i^{(0)}|$), for a fixed frequency of $\omega = 0.000\,44$ a.u. (which is one-tenth of the CO_2 laser frequency) and for two different intensities corresponding to N [that is, $2(P + |E_i^{(0)}|)/(\hbar\omega)$] equal to 13 760 and 33 576. We show the angular distribution obtained (i) within the dipole approximation (solid line), (ii) by taking into account the photon momentum, but within the nonrelativistic framework, so the Doppler-shifted frequency is ω_0 (long dashed line), and (iii) by taking into account the photon momentum, but using the relativistic Doppler shift (short dashed line). The relativistic Doppler-shifted frequency is $\gamma\omega_0$, where $\gamma \equiv (1 - v^2/c^2)^{-1/2}$, and in computing the short dashed line we modified ρ_f by dividing the right-hand side of Eq. (75) by γ. Within the dipole approximation the shape of the angular distribution is a symmetric peak centered at $\theta = \pi/2$; the photoelectron is preferentially ejected in the plane in which the electric-field force lies, i.e., the polarization plane. Indeed, as expected from the analysis of the preceding paragraph, the peak is approximately a Gaussian and its FWHM is given accurately (to within an error of a few percent) by Eq. (85). When the photon momentum is included the photoelectron is no longer preferentially ejected in the polarization plane; due to the absorption of momentum $N\hbar\mathbf{K}$ from the radiation field, the electron receives a kick perpendicular to this plane and the peak of the angular distribution shifts by a small angle θ_K, where $\sin(\theta_K) \approx \theta_K \approx (NK)/k_f$. This effect is purely kinematical and, since θ_K is of order v/c, is nonrelativistic in origin. It is well known that a similar effect occurs when an atom is photoionized by a single very-high-frequency photon; in fact, if the photon energy is comparable to the rest energy of the electron (the relativistic regime) the shift in the angular distribution is very pronounced [24]. The shift in the angular distribution in the case of multiphoton detachment by a low-frequency field has also been noted by Reiss [25], who carried out a calculation within the relativistic framework. Although the shift in the angular distribution is very small in the present case, its effect is amplified due to the FWHM of the the angular distribution being so small. The relevant parameter is the ratio of the shift to the FWHM; this parameter is

$$\delta \equiv \theta_K/\Delta u \tag{86}$$

$$\approx \frac{N^{3/4}}{2\sqrt{\ln(2)}}\left(\frac{2\hbar\omega|E_i^{(0)}|}{\mu^2c^4}\right)^{1/4}. \tag{87}$$

This last estimate of δ agrees with the results of Fig. 1 to within 5%.

Since $\theta_K^2 \ll 1$ in the present case, it is reasonable to assume that relativistic corrections are negligible. Indeed, the shift θ_K in the peak is the same whether the nonrelativistic or relativistic Doppler shift is used. Surprisingly, however, the size of the peak is considerably larger when the nonrelativistic Doppler shift is used. This significant increase in the size of the peak must be spurious since

corrections to the N-photon rate *integrated* over θ are relativistic, i.e., of order $(v/c)^2$, and are therefore very small. In fact, when the relativistic Doppler shift is used, that is, when the transverse Doppler shift is included, the size of the peak, and therefore the integrated N-photon rate, are the same as when the photon momentum is neglected altogether; in other words, the only consequence of including the photon momentum when the relativistic Doppler shift is used is a small shift in the angular distribution.

The reason for the discrepancy which arises when the nonrelativistic Doppler shift is used may be seen as follows. From Eq. (69) we know that the variation of the photodetachment matrix element $M_{i,N}(\mathbf{k}_f)$ with respect to I and ω will be determined largely by $J_N(\rho_f)$. [Indeed, in the zero-range potential model we have $|M_{i,N}(\mathbf{k}_f)| = |\sqrt{2\pi\kappa}J_N(\rho_f)|$.] Assuming that $N \gg 1$, the Bessel function $J_N(x)$ is extremely small when (with x positive) $x \ll N$, and it reaches a first maximum for x close to (slightly larger than) N. As x approaches N from below, $J_N(x)$ rises very rapidly, and, after reaching the first maximum, subsequently oscillates. In Fig. 2 we show $J_N(x)$ vs x for $N = 33\,576$. Now, as noted above, $\rho_f \approx N$ when (as in the present case) $E_f \approx P$. Using Eq. (76) to differentiate ρ_f with respect to u shows that the maximum value of ρ_f is $\gamma\rho_0$. Hence, as u increases from zero, ρ_f first rises from ρ_0 to $\gamma\rho_0$, and although this change is very small the increase in $J_N(\rho_f)$ is considerable, as shown in Fig. 1 for $N = 33\,576$. [As u increases further, ρ_f decreases, and $J_N(\rho_f)$ rapidly drops to zero.] It is this significant increase in $J_N(\rho_f)$, as ρ_f changes from ρ_0 to $\gamma\rho_0$, which is responsible for the significant change in the size of the peak in the angular distribution when the nonrelativistic Doppler shift is used.

However, the difference between ρ_0 and $\gamma\rho_0$ is a relativistic correction, and should be dropped since we are working within the nonrelativistic framework. Alternatively, we can use the relativistic Doppler shift, which amounts to dividing the nonrelativistic ρ_f by γ, so the maximum in the relativistic ρ_f is ρ_0, the same as when the Doppler shift is neglected altogether. Thus, when we use the relativistic Doppler shift we obtain the same angular distribution as when the Doppler shift is neglected, except for a small but physically significant shift.

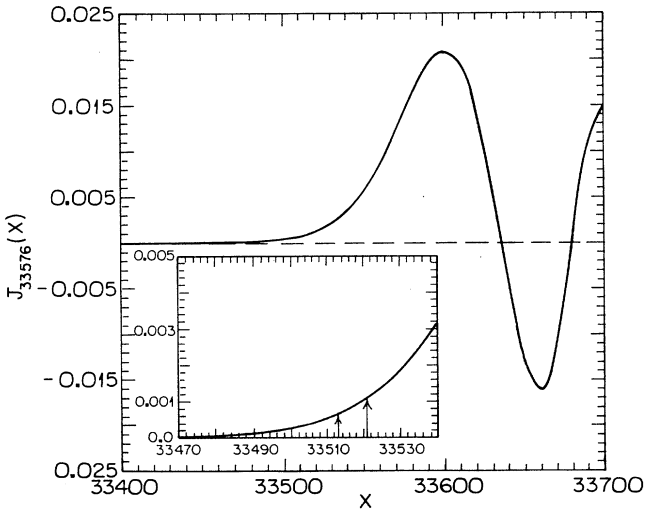

FIG. 2. Standard regular Bessel function $J_N(x)$ for $N = 33\,576$. The arrows indicate the maximum value of ρ_f when the relativistic (left arrow) or the nonrelativistic (right arrow) Doppler shift is considered.

If we increase the intensity, we increase the value of N_0 [$= P/(\hbar\omega)$] and therefore we increase the value of N ($\approx 2N_0$) for which the photodetachment rate is maximum. Accordingly, we increase δ, and the dipole approximation becomes less and less reliable. However, the photodetachment rate also increases as the intensity does, and eventually we reach the point where photodetachment occurs within a few cycles or less, and our analysis no longer applies — we must take into account the depletion of ions and also the temporal variation of the intensity on the rising edge of the pulse. We end this paper by estimating the intensity for which photodetachment occurs within about a cycle. The total rate for photodetachment, integrated over the angle and summed over N, is $\Gamma/\hbar$, where

$$\Gamma = \int d\Omega_{\mathbf{k}_f} \sum_{N \geq N_{\min}} \frac{d\Gamma_N}{d\Omega_{\mathbf{k}_f}}. \tag{88}$$

We use the tunneling formula [26] to estimate Γ

$$\Gamma = CFe^{-F_0/F}, \tag{89}$$

where $F_0 = (2/3)(\sqrt{\mu}/e\hbar)|2E_i^{(0)}|^{3/2}$ and where we obtained the value of C by fitting to our calculated results (although this value could be derived analytically [26]). Putting $C = 2.89$ a.u. gives an accurate estimate of Γ over a wide range of frequencies and intensities. If $\omega = 0.000\,44$ a.u. we find, using the tunneling formula, that $\Gamma = \hbar\omega/(2\pi)$ when $I = 2.71 \times 10^{11}$ W/cm^2, and at this intensity $\delta = 0.813$. However, if $\omega = 0.0044$ a.u. we find that $\Gamma = \hbar\omega/(2\pi)$ when $I = 7.65 \times 10^{11}$ W/cm^2 and, at this intensity, δ is only 0.019. Thus we conclude that at frequencies above the CO_2 laser frequency *but still well below the unperturbed atomic orbital frequency*, δ is small and consequently corrections to the dipole approximation may be ignored. It remains to investigate the adequacy of the dipole approximation at frequencies above the unperturbed atomic orbital frequency when atoms are ionized by strong light. In the latter process, the applied field strength can considerably exceed the atomic field strength before the atom is ionized, and consequently the initial bound state may be significantly perturbed, as a result of which the analysis is rather more complicated than in the case of multiphoton detachment by a low-frequency field.

ACKNOWLEDGMENT

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