# Scattering-theoretical approaches to multiphoton ionization in strong fields

T. Åberg
*Laboratory of Physics, Helsinki University of Technology, 02150 Espoo, Finland*

D.-S. Guo,* J. Ruscheinski, and Bernd Crasemann
*Department of Physics, University of Oregon, Eugene, Oregon 97403*
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The nonrelativistic version of the authors' theory of multiphoton ionization [Phys. Rev. **40**, 4997 (1989)] is compared with other forms of scattering-theoretical approaches that employ different boundary conditions. These methods, which are expressed in terms of formal time-independent scattering theory, are shown to be equivalent in the large-photon-number limit to use of the semiclassical time-dependent $S$-matrix theory. In the authors' treatment it is assumed that the photoelectron has escaped from both the electromagnetic and atomic fields; a perturbation expansion in the photon-electron interaction is obtained that correctly accounts for the atomic final-state interactions of the photoelectron. The case of multiphoton detachment of $I^-$ is treated, for which the influence of the final-state atomic potential on the transition rate can be ignored. It is shown both analytically and numerically for the Nd–yttrium aluminum garnet wavelength that the transition rates obtained for this case from the various approaches are distinctly different. The present approach is shown to be consistent with an abrupt switching off of a spatially unlimited monomode field, in contrast with Keldysh-type approaches, which correspond to a long switch-off time if the *ad hoc* assumption of ponderomotive acceleration is made. Above-threshold-ionization spectra calculated from the present approach exhibit interference effects and are redshifted with respect to what is expected for slow switch-off. It is demonstrated, using the semiclassical time-dependent approach, that the corresponding final off-field scattering-state wave function only exists if the ponderomotive potential per unit photon energy is an integer, in accordance with our earlier time-independent analysis. The origin of this curious feature is made clear.

## I. INTRODUCTION

Most theoretical work on laser-atom interactions has been based on the time-dependent Schrödinger equation in which the vector potential is described in terms of a classical time-varying electromagnetic field [1–5]. There is good reason for this approach, because it is well known that the laser field can be accurately described as a classical electromagnetic field. Furthermore, the temporal characteristics of the laser pulse can be included in a time-dependent formalism.

Fundamentally, however, the laser-atom interaction should be treated within the framework of quantum electrodynamics. This requirement becomes increasingly acute as laser intensities reach higher values, even exceeding one atomic unit ($3.5\times10^4$ TW cm$^{-2}$). Even though the solutions of the Dirac equation for an electron interacting with a quantized, elliptically polarized electromagnetic field are known [6], this does not help very much unless a proper scattering-theoretical approach is also established. It has been argued that such a treatment can only be carried out within the framework of the formal field-theoretical theory of scattering if it is possible by means of the density-matrix technique to take into account the actual scattering conditions that involve the spatial and temporal characteristics of the laser pulse [7]. It is thus important to examine the question to what extent formal scattering theory is applicable, in the nonrelativistic limit, to processes such as multiphoton ionization. It is in the nonrelativistic regime that multiphoton ionization by laser light has been studied most thoroughly [8–10].

In Sec. II of this paper we reexamine the correspondence [1,5,11,12] between the quantum-mechanical and semiclassical descriptions of a system in which an electron is exposed to an atomic potential, short or long range, and to the vector potential of a monochromatic, elliptically polarized electromagnetic field. We present a completely general derivation of the semiclassical Schrödinger equation from the quantum-mechanical equation by projecting on a coherent state and taking the large-photon-number limit. The derivation of this "many-photon correspondence principle" does not depend on the boundary conditions imposed on the wave functions and thus indicates that there is a one-to-one correspondence between the quantum-mechanical time-independent and the semiclassical time-dependent scattering solutions. In the present work, this result is needed to demonstrate the correspondence between various quantum-mechanical and semiclassical scattering models of multiphoton ionization.

In Sec. III we examine and compare three different transition matrix elements for multiphoton ionization. The matrix elements are obtained by means of formal scattering theory [13,14] and are shown to correspond to three different boundary conditions in the semiclassical

 

framework: (1) the atomic interaction and the photon-electron interaction are both required to vanish in the remote future, (2) only the atomic interaction, or (3) only the photon-electron interaction is required to vanish in the remote future. These three possibilities correspond to entirely different physical pictures, namely, multiphoton ionization considered as (1) a breakup reaction like ordinary photoionization, (2) a pickup reaction in which the electron is captured by the electromagnetic field, or (3) a single-potential excitation process. It is shown explicitly that in case (1) the semiclassical final-state scattering wave function has the same properties as its quantum-mechanical counterpart. In particular, the final-state scattering wave function only exists when the ponderomotive potential energy is an integer times the photon energy. A simple interpretation of this result is outlined; it is argued that this curious property of the final-state wave function may lead to observable consequences when the switch-off time of the electromagnetic field is very short. A numerical demonstration of the effect is provided in Sec. IV, where calculations of above-threshold-ionization spectra of $I^-$ are presented for both short and long switch-off times. We use natural units, $\hbar=c=1$, unless otherwise stated.

## II. THE MANY-PHOTON CORRESPONDENCE PRINCIPLE

We present a completely general derivation of the semiclassical time-dependent Schrödinger equation which is often used to describe laser-atom interactions and in which the laser field is represented by a classical electromagnetic field. Previous work, which has been reviewed by Mittleman [1], is based either on the use of the phase representation [11] or on the correspondence between the coherent state and the classical electromagnetic field [12]. The equivalence of the semiclassical and quantum-mechanical resolvent equations in the large-photon-number limit has been demonstrated by Faisal [5]. We utilize the coherent-state description in the large-photon-number limit, but do not require that the field is initially in a coherent state [1,12].

Consider the time-dependent Schrödinger equation

$$(H_\gamma+H_e+U+V)\psi=i\frac{\partial\psi}{\partial t}\ , \tag{1}$$

where we assume for simplicity that

$$H_\gamma=(\omega/2)(a^\dagger a+aa^\dagger) \tag{2}$$

represents a single-mode field. Generalizations to spatially limited and multimode fields are possible but will not be discussed here. In Eq. (1), $H_e=(1/2m_e)\nabla^2$ is the kinetic-energy operator, and $U=U(\mathbf{r})$ is the potential energy of the electron. These operators could also be specified in a many-electron form. The photon-electron interaction is described in the Coulomb gauge by

$$V=\frac{e}{m_e}\mathbf{A}(-\mathbf{k}\cdot\mathbf{r})\cdot i\nabla+\frac{e^2}{2m_e}\mathbf{A}^2(-\mathbf{k}\cdot\mathbf{r})\ , \tag{3}$$

where we have $e=-|e|$ and the photon field is described by the vector potential

$$\mathbf{A}(-\mathbf{k}\cdot\mathbf{r})=g(\boldsymbol{\epsilon}e^{i\mathbf{k}\cdot\mathbf{r}+i\theta/2}a+\boldsymbol{\epsilon}^*e^{-i\mathbf{k}\cdot\mathbf{r}-i\theta/2}a^\dagger)\ . \tag{4}$$

In Eq. (4), the quantity $g=(2V_\gamma\omega)^{-1/2}$ contains an arbitrary normalization volume $V_\gamma$ of the photon field, and $\theta$ represents an arbitrary phase factor.

The polarization vector $\boldsymbol{\epsilon}$ is given by $\boldsymbol{\epsilon}=\boldsymbol{\epsilon}_x\cos\xi/2+i\boldsymbol{\epsilon}_y\sin\xi/2$, so that $\xi=0$ corresponds to linear polarization and $\xi=\pm\pi/2$, to circular polarization. The unit vectors $\boldsymbol{\epsilon}_x$ and $\boldsymbol{\epsilon}_y$ are perpendicular to each other and to the propagation vector $\mathbf{k}=k\boldsymbol{\epsilon}_z$. We thus have

$$\boldsymbol{\epsilon}\cdot\boldsymbol{\epsilon}=\boldsymbol{\epsilon}^*\cdot\boldsymbol{\epsilon}^*=\cos\xi\ ,\quad \boldsymbol{\epsilon}\cdot\boldsymbol{\epsilon}^*=1\ . \tag{5}$$

The eigenfunctions $\Psi$ and the corresponding eigenenergies $\mathcal{E}$ of the time-independent Hamiltonian $H=H_\gamma+H_e+U+V$ can in principle be obtained by substituting $\psi=\Psi\exp(-i\mathcal{E}t)$ into Eq. (1). We introduce the unitary operator $U_t=\exp(iH_\gamma t)$ and transform Eq. (1) into an equation for $\psi_t=U_t\psi$ by multiplying it from the left and using the relation $U_t^\dagger U_t=1$. The result is

$$H_t\psi_t=i\frac{\partial\psi_t}{\partial t}\ , \tag{6}$$

where $H_t=H_e+U+V_t$. The interaction

$$V_t=\frac{e}{m_e}\mathbf{A}(kx)\cdot i\nabla+\frac{e^2}{2m_e}\mathbf{A}(kx)^2 \tag{7}$$

now depends on the time, since $-\mathbf{k}\cdot\mathbf{r}$ in the vector potential has become $kx=\omega t-\mathbf{k}\cdot\mathbf{r}$.

The next step involves the projection of Eq. (6) onto the coherent state $|\alpha\rangle=\mathcal{D}^\dagger|0\rangle$, where

$$\mathcal{D}=e^{\alpha^*a-\alpha a^\dagger} \tag{8}$$

is the unitary shift operator [2]. By introducing a complete set of photon states $|n\rangle$ ($n=0,1,2,\ldots$), we have

$$\sum_n\langle 0|\mathcal{D}H_t\mathcal{D}^\dagger|n\rangle\langle n|\mathcal{D}\psi_t\rangle=i\frac{\partial}{\partial t}\langle 0|\mathcal{D}\psi_t\rangle\ , \tag{9}$$

which is exact. It is straightforward to calculate the matrix elements $\langle 0|\mathcal{D}H_t\mathcal{D}^\dagger|n\rangle$ using known properties of the shift operator [2], that is,

$$\mathcal{D}a\mathcal{D}^\dagger=a+\alpha\ ,\quad \mathcal{D}a^\dagger\mathcal{D}^\dagger=a^\dagger+\alpha^*\ . \tag{10}$$

Clearly, $n$ is limited to $n\le 2$. From Eqs. (5) and (10), we have

$$\langle 0|\mathcal{D}H_t\mathcal{D}^\dagger|0\rangle=\langle\alpha|H_t|\alpha\rangle=H_e+U+V_{\rm cl}+\frac{e^2}{2m_e}g^2\ , \tag{11}$$

where the "classical" interaction operator $V_{\rm cl}$ is obtained by replacing $a$ by $\alpha$ and $a^\dagger$ by $\alpha^*$ in the corresponding quantum-mechanical operator (7). The two other nonvanishing terms are

$$\langle 0|\mathcal{D}H_t\mathcal{D}^\dagger|1\rangle=\frac{eg}{m_e}e^{-i\gamma}(\boldsymbol{\epsilon}\cdot i\nabla)+\frac{e^2g^2}{m_e}(\alpha\cos\xi e^{-2i\gamma}+\alpha^*) \tag{12a}$$

and

$$\langle 0|\mathcal{D}H_t\mathcal{D}^\dagger|2\rangle=\frac{e^2g^2\cos\xi}{\sqrt{2}m_e}e^{-2i\gamma}\ , \tag{12b}$$

where we have used the abbreviation $\gamma=kx-\theta/2$.

If it is assumed that the parameter $\alpha$ in $a|\alpha\rangle=\alpha|\alpha\rangle$ is real, then it can be shown with the aid of the relation $|\alpha\rangle=\mathcal{D}^\dagger|0\rangle$ and Eqs. (10) that

$$\langle 1|\mathcal{D}\psi_t\rangle=\langle 0|\mathcal{D}(a-\alpha)|\psi_t\rangle \tag{13a}$$

and

$$\langle 2|\mathcal{D}\psi_t\rangle=\left\langle 0\left|\mathcal{D}\frac{(a-\alpha)^2}{\sqrt{2}}\right|\psi_t\right\rangle\ . \tag{13b}$$

We use the notation $\Lambda=g\alpha$ and, in accordance with Eq. (11), define the semiclassical Hamiltonian

$$H_{\rm cl}=H_e+U+V_{\rm cl}=\frac{1}{2m_e}[i\nabla+e\,\mathbf{A}_{\rm cl}(kx)]^2+U\ , \tag{14}$$

where

$$\mathbf{A}_{\rm cl}(kx)=\Lambda(\boldsymbol{\epsilon}e^{-i\gamma}+\boldsymbol{\epsilon}^*e^{i\gamma}) \tag{15}$$

is the classical vector potential with $\gamma=kx-\theta/2$. According to Eqs. (11)–(13), the semiclassical time-dependent Schrödinger equation

$$H_{\rm cl}\langle\alpha|\psi_t\rangle_\infty=i\frac{\partial\langle\alpha|\psi_t\rangle_\infty}{\partial t} \tag{16}$$

is obtained from Eq. (9) by taking the limit $g\to 0$ ($\alpha\to\infty$, $\Lambda=$const). We wish to find the solutions $\langle\alpha|\psi_t\rangle_\infty$ which correspond to the stationary solutions

$$\psi=e^{-i\mathcal{E}t}\sum_{j=-N}^{\infty}c_j(\mathbf{r},\mathbf{k},\boldsymbol{\epsilon})|N+j\rangle \tag{17a}$$

of Eq. (1), where we have expanded $\Psi$ in terms of the photon-number states $|N+j\rangle$. With the notation $\mathcal{E}_e=\mathcal{E}-(N+\frac{1}{2})\omega$, we have

$$\langle\alpha|\psi_t\rangle=e^{-i\mathcal{E}_et}\sum_{j=-N}^{\infty}c_j(\mathbf{r},\mathbf{k},\boldsymbol{\epsilon})\langle\alpha|N+j\rangle e^{ij\omega t}\ . \tag{17b}$$

We assume that, for a given average intensity $I=N\omega/V_\gamma=2Ng^2\omega^2$, $N$ is a large number of photons, such that the average number $j$ of transferred photons is much smaller than $N$. Since classically $I=2\Lambda^2\omega^2=2\alpha^2g^2\omega^2$, we set $\alpha=\sqrt{N}$. Consequently, the limit $\alpha\to\infty$ ($g\to 0$, $I$ const) is obtained in Eq. (17b) in such a way that the coefficients $\langle\alpha|N+j\rangle\to(2\pi N)^{-1/4}$ become independent of $j$ according to Stirling's formula. In this limit, we have $\langle\alpha|H_\gamma|\alpha\rangle=\langle\Psi|H_\gamma|\Psi\rangle=N$, and

$$\langle\alpha|\psi_t\rangle_\infty=e^{-i\mathcal{E}_et}\sum_{j=-\infty}^{+\infty}c_j(\mathbf{r},\mathbf{k},\boldsymbol{\epsilon})e^{ij\omega t}\ , \tag{18}$$

where the $c_j$ are the same time-independent expansion coefficients as in the stationary solutions (17a).

Our derivation of the correspondence between the wave functions (17a) and (18) is completely general, in the sense that it is valid for any boundary conditions that are imposed on $\psi=\Psi e^{-i\mathcal{E}t}$. The coefficients $c_j$ are uniquely determined by Eq. (1) or Eq. (16) once the boundary conditions have been established. In the application of formal scattering theory [13,14] to collisions in the electromagnetic field, these conditions are specified in terms of (fully or partly) noninteracting states, corresponding in the semiclassical picture to the requirement that the exchange of $U$ and $V$ must take place in time intervals which are used to *define* the switch-on and -off of the potentials in the beginning and end of the collision. This requirement is fully justified by the wave-packet formalism [13,14].

An analytical example of the correspondence principle is provided by the solution of Eq. (16) in the limit of a vanishing atomic potential $U$. The vector potential (15) is assumed to be switched on and off adiabatically. These "Volkov" solutions [15] correspond to

$$c_j^V(\mathbf{r};\mathbf{k},\boldsymbol{\epsilon})=(2\pi)^{-3/2}e^{i[\mathbf{P}+(Z-j)\mathbf{k}]\cdot\mathbf{r}-ij(\phi_\xi+\theta/2)}\mathcal{J}_j^*(\zeta,\eta,\phi_\xi) \tag{19}$$

in Eqs. (17a) and (18). The eigenenergies $\mathcal{E}_e$ are given by

$$\mathcal{E}_e=E_{\rm drift}+\Delta\ , \tag{20}$$

where $E_{\rm drift}=\mathbf{P}^2/2m_e$ is the drift energy of the electron in the electromagnetic field and

$$\Delta=Z\omega=\frac{e^2\Lambda^2}{m_e} \tag{21}$$

is its ponderomotive potential or quiver energy. The generalized Bessel function $\mathcal{J}_i$ in Eq. (19) depends on

$$\zeta=\frac{2|e|\Lambda}{m_e\omega}|\mathbf{P}\cdot\boldsymbol{\epsilon}|\ , \tag{22a}$$

$$\eta=(Z/2)\cos\xi\ , \tag{22b}$$

and the angle $\phi_\xi$ in

$$\mathbf{P}\cdot\boldsymbol{\epsilon}=|\mathbf{P}\cdot\boldsymbol{\epsilon}|e^{i\phi_\xi}\ . \tag{22c}$$

It is defined by

$$\mathcal{J}_j(\zeta,\eta,\phi_\xi)=\sum_{m=-\infty}^{+\infty}J_{-j-2m}(\zeta)J_m(\eta)e^{2im\phi_\xi}\ , \tag{23}$$

where the $J_\nu$ are ordinary Bessel functions.

The coefficients (19) have also been obtained from the exact quantum-electrodynamical solution [6] by taking the large-photon-number and nonrelativistic limits [7], and from the time-independent, nonrelativistic Schrödinger equation by replacing the number operator $N_a$ in the field momentum operator $\mathbf{k}(N_a+1/2)$ by a real number [16].

Kelsey and Rosenberg [17] have previously noticed a similar correspondence between approximate semiclassical and quantum-mechanical Volkov solutions. These authors' solutions do not incorporate the ponderomotive momentum $Z\mathbf{k}$, which may have important consequences. First, the construction of Green's functions in scattering wave functions involves an integration over *all* kinematical momenta $\mathbf{P}$ [13], and thus also incorporates

absolute values of $\mathbf{P}$ that are equal to or smaller than $Zk$. Second, one can easily imagine scattering situations within the nonrelativistic regime in which $P \simeq Zk$. As an example, consider an electron with 1 Ry energy, scattering or drifting in a long $CO_2$ laser pulse of intensity $I = 10^{-2}$ a.u. and wavelength $\lambda = 10.6\ \mu$m. Yet, its classical peak velocity of oscillation is only $\simeq 0.3c$.

## III. FORMAL TIME-INDEPENDENT SCATTERING THEORY OF MULTIPHOTON IONIZATION

Multiphoton ionization (MPI) involves scattering from two potentials, $U$ and $V$, that describe the atom (ion)-electron and photon-electron interactions, respectively. Initially, the electron is bound by $U$ but does not interact with the electromagnetic field. In the semiclassical approach based on Eq. (16), this situation corresponds to the boundary condition that $U$ is on but $V$ is off in the remote past. In formal scattering theory [13,14], this boundary condition is introduced by defining the corresponding noninteracting atomic-electron and free-photon states.

The interaction is assumed to take place in the presence of the time-independent potentials $U$ and $V$. After the interaction, the wave function of the electron may be subject to three different boundary conditions, which correspond to entirely different physical pictures of MPI and in the semiclassical approach are as follows: (1) $U$ and $V$ off, (2) $U$ off but $V$ on, and (3) $U$ on but $V$ off. In the formal scattering theory these boundary conditions are again established by defining the corresponding noninteracting electron and photon states. According to the many-photon correspondence principle of Sec. II, the semiclassical approach and formal scattering theory should lead to the same ionization rates, provided the photon-electron interaction takes place in the presence of a large number of photons, i.e., the "laser approximation" [5] is valid. Then there is a one-to-one correspondence between the transition matrix elements, i.e., they look exactly the same in both representations. In the following, we examine the ionization process under all three conditions indicated above.

*Model (1): MPI as a breakup process.* This case, which implies *U off, V off after the interaction,* has been considered by Gersten and Mittleman [18] in terms of the semiclassical approach and by Guo, Åberg, and Crasemann [7] in the framework of formal scattering theory. We review the results of Ref. [7] and show explicitly that they remain valid within the semiclassical approximation.

The derivation of the exact transition matrix element

$$T_{fi}^{(1)} = \langle \Phi_f^-, m | V | \Psi_i^+ \rangle + \langle \Phi_f, m | U | \Phi_i, l \rangle \tag{24}$$

is analogous to the treatment of a "breakup" process, such as $\pi^+ + d \rightarrow 2p$, in which case $U$ would describe the nucleon-nucleon and $V$, the pion-nucleon interaction [13,14]. It also gives the correct transition matrix element for ordinary photoionization [13], as shown below by Eq. (35). In Eq. (24), the direct products $|\Phi_i, l\rangle = \Phi_i \otimes |l\rangle$ and $|\Phi_f, m\rangle = \Phi_f \otimes |m\rangle$ involve the initial- and final-state wave functions of the electron and the corresponding photon-number states. Since $m \neq l$, the second amplitude vanishes. The scattering wave function $\Psi_i^+$ is

$$\Psi_i^+ = (1 + G^+ V) | \Phi_i, l \rangle , \tag{25}$$

where $G^+ = (\mathcal{E} - K - U - V + i\varepsilon)^{-1}$ is the full Green's operator and where we have used the notation $K = H_\gamma + H_e$. The final-state scattering wave function

$$|\Phi_f^-, m\rangle = (1 + G_U^- U) | \Phi_f, m \rangle , \tag{26}$$

where $G_U^- = (\mathcal{E} - K - U - i\varepsilon)^{-1}$ accounts for the scattering of the electron by the potential $U$. If $U$ is a short-range atomic potential, $\Phi_f$ is a plane wave. If $U$ is a long-range ionic potential, a Coulomb-distorted plane wave must be used [19], such that the off-shell wave function of form (26) becomes in the on-shell limit equal to the incoming-wave-normalized wave function [20] for an electron that moves in a potential with a Coulomb tail. We introduce the off-field scattering wave function

$$\psi_f^- = (1 + G_V^- V) | \Phi_f, m \rangle , \tag{27}$$

where $\Phi_f$ is usually taken to be a plane wave without distortion, and where $G_V^- = (\mathcal{E} - K - V - i\varepsilon)^{-1}$. The next step in the derivation is based on the assumption that the $\Phi_f$'s in Eqs. (26) and (27) are identical, but not necessarily plane waves. We have [7]

$$T_{fi}^{(1)} = \langle \psi_f^- | V | \Phi_i, l \rangle + \langle \psi_f^- | U G^+ V | \Phi_i, l \rangle \tag{28}$$

and note that so far there are no constraints on the potentials $U$ and $V$, other than their time independence during the combined atomic (ionic) and photon-electron interactions.

An examination [7] of the wave function (27) shows that, if $\Phi_f$ is a plane wave and there is no spatial restriction of the vector potential $\mathbf{A}(-\mathbf{k}\cdot\mathbf{r})$, then $\psi_f^-$ reduces in the large-photon-number limit to

$$\psi_f^- = \Psi_{\mathbf{P}_f, n} \mathcal{J}_Z e^{iZ(\phi_\xi + \theta/2)} \tag{29}$$

if and only if $Z$ is an integer, equal to $m - n$. Otherwise, $\psi_f^-$ approximately vanishes. The Volkov state $\Psi_{\mathbf{P}_f, n}$ is given by Eq. (17a) with the coefficients (19) and with $N = n = m - Z$; $\mathbf{P} = \mathbf{P}_f$ is the momentum of the electron in the field-free region. In the Appendix we show that the corresponding semiclassical scattering wave function exhibits the same behavior, in accordance with the many-photon correspondence principle. Hence we can trace the origin of this curious behavior and decide whether it is an artifact or not; this is done in Sec. IV.

*Model (2): MPI as a pickup process.* This case, which implies *U off, V on after the interaction,* has been treated by Mu [21] from the point of view of formal scattering theory. The result, which originates from work by Keldysh [22], has apparently been considered earlier by Reiss [23,24], who employed the semiclassical time-dependent scattering approach. The derivation is analogous to the evaluation of the cross section for a "pickup" reaction such as $n + p \rightarrow d$ [13], in which the proton is initially bound by $U$. The interaction between neutron and pro-

ton is described by $V$, both initially and in the "picked-up" deuteron state. There is also a close relationship between the distorted-wave theory of electron capture [25] and model (2) [26].

The exact transition matrix element [13,21] can be written

$$\begin{aligned} T_{fi}^{(2)} &= \langle \psi_V | U | \psi_i^+ \rangle \\ &= \langle \psi_V^- | V | \Phi_i, l \rangle \\ &= \langle \psi_V | V | \Phi_i, l \rangle + \langle \psi_V | U G^+ V | \Phi_i, l \rangle , \end{aligned} \tag{30}$$

where $\Psi_i^+$ is given by Eq. (25) and we have

$$\psi_V^- = (1 + G^- U)\psi_V . \tag{31}$$

Here, $\psi_V$ satisfies the Schrödinger equation

$$(K + V)\psi_V = \mathcal{E}\psi_V . \tag{32}$$

This means that $\psi_V$ should be taken to be the Volkov solution [Eqs. (17a) and (19)] in the large-photon-number limit. The equality of the two forms in Eq. (30) is based on the fact that $\langle K\psi_V | \Phi_i, l \rangle = \langle \psi_V | K \Phi_i, l \rangle$.

The difference between this MPI model and model (1) becomes apparent if one compares Eqs. (28) and (30): The scattering wave function $\psi_f^-$ is replaced by the Volkov solution $\psi_V$ in the "pickup" model and, in accordance with Eq. (29), there is no reason why it should lead to the same transition rate as model (1), even for the case in which $U$ is weak compared with $V$.

*Model (3): MPI as a single-potential excitation process.* This case, which implies *U on, V off after the interaction,* is equivalent to scattering from a single potential, which is $V$. According to formal scattering theory [13,14], we have

$$T_{fi}^{(3)} = \langle \Phi_f, m | V | \Psi_i^+ \rangle = \langle \chi_f^- | V | \Phi_i, l \rangle , \tag{33}$$

where $\Psi_i^+$ is given by Eq. (25). The final-state scattering wave function $\chi_f^- = (1 + G^- V)|\Phi_f, m\rangle$ describes the scattering from $V$ in the presence of $U$ and should not be confused with any of the final-state scattering wave functions in models (1) and (2).

The transition matrix elements $T_{fi}^{(\nu)}$ ($\nu = 1,2,3$) lead to distinctly different transition rates. It is instructive to compare the transition amplitudes in the weak- and strong-field cases.

*(i) Weak-field case.* From the form

$$T_{fi}^{(1)} = \langle \Phi_f^-, m | V(1 + G^+ V) | \Phi_i, l \rangle$$

and the expansion

$$\begin{aligned} G^+ &= G_U^+ + G_U^+ V G^+ \\ &= G_U^+ + G_U^+ V G_U^+ + G_U^+ V G_U^+ V G_U^+ + \cdots \end{aligned} \tag{34}$$

we obtain

$$T_{fi}^{(1)} = \left\langle \Phi_f^-, m \left| \left[ \sum_{\nu=0}^{\infty} (V G_U^+)^\nu \right] V \right| \Phi_i, l \right\rangle . \tag{35}$$

The representation

$$G_U^+ = \sum_{\nu,\mu} \frac{|\Phi_\nu, \mu\rangle\langle\Phi_\nu, \mu|}{\mathcal{E} - E_\nu - \omega(\mu + 1/2) + i\varepsilon} , \tag{36}$$

where $|\Phi_\nu, \mu\rangle$ are eigenstates of $K + U$, immediately leads to the lowest-order nonresonant transition-rate formula [8] for the absorption of $j = l - m$ photons. The final-state wave function $|\Phi_f^-, m\rangle$ is the correct wave function, normalized to the incoming wave, for scattering by $U$ [20]; thus $T_{fi}^{(1)}$ can be used for calculations of angular distributions or for studies of final-state channel interaction in the many-electron case at any field strength. This becomes apparent when one realizes that the $\nu = 0$ term in Eq. (35) gives the correct transition matrix element [20] for ordinary photoionization. In $T_{fi}^{(3)}$, $|\Phi_f^-, m\rangle$ is replaced by one of the standing-wave eigenfunctions of $K + U$, which indicates that this transition matrix element can only be used to obtain total lowest-order MPI rates. Hence model (3) will not be considered further here.

The expansion (34) of the propagator $G^+$ in $T_{fi}^{(2)}$ does not lead to the perturbation expansion (35). This fact may limit the validity of model (2) for a universal description of MPI.

*(ii) Strong-field case.* If the potential is short range, it may be argued that the second amplitude in Eq. (28) can be neglected; there is numerical evidence that this is possible [27]. Then we have $T_{fi}^{(1)} \simeq \langle \psi_f^{(-)} | V | \Phi_i, l \rangle$. According to Eq. (30), the same procedure leads to $T_{fi}^{(2)} \simeq \langle \psi_f | V | \Phi_i, l \rangle$, which is equivalent to the strong-field approximation in the semiclassical approach [21–24]. Equation (29) shows that we have $T_{fi}^{(1)} \neq T_{fi}^{(2)}$, in general. The corresponding rates are compared in detail in Sec. IV, with MPI of $I^-$ as an example.

## IV. STRONG-FIELD MULTIPHOTON DETACHMENT RATE FOR NEGATIVE IONS

### A. Photoelectron energy

Before considering the energetics predicted by the scattering models discussed in Sec. III, we briefly summarize the current view regarding nonresonant photoionization by a realistic laser pulse [28,29]. Let the electron originally be bound to an atom with binding energy $E_B$. The electron is released from the atom during a laser pulse under absorption of $j = j_{\min} + s$ photons, where we have $j_{\min}\omega \geq E_B$ but $(j_{\min} - 1)\omega < E_B$, and $s \geq 0$. If the pulse has a duration $\tau$, energy conservation requires

$$j\omega = -E + \kappa\Delta + E_B + \Delta_i - \Delta_g , \tag{37}$$

where $E = E_{\text{obs}}$ is the energy of the photoelectron as *observed* in a detector, and $\Delta$ is the ponderomotive potential energy (21). Equation (21) is usually expressed in terms of the time-averaged peak intensity $I$ of the field, i.e., in Système International (SI) units,

$$\Delta = \frac{e^2 I}{2c\epsilon_0 m_e \omega^2} . \tag{38}$$

In Eq. (37), $\Delta_g$ and $\Delta_i$ are the ac Stark shifts of the ground and ionized states, respectively. We shall neglect

the small difference $\Delta_i - \Delta_g$ in the following. The factor $\kappa$, which lies between 0 and 1, describes how much of $\Delta$ is converted back to the electromagnetic field when the pulse vanishes and the electron leaves the pulse; this depends on the pulse duration $\Delta\tau = \tau_{on} + \tau_{off}$, particularly on the switch-off time $\tau_{off}$. The factor $\kappa$ also depends on the spatial extension of the pulse, on the peak intensity $I$, and on how many times the electromagnetic field oscillates during $\Delta\tau$, i.e., on $\omega$. It should be noted that *inside* the field the energy of the electron consists, in accordance with Eq. (20), of both the potential-energy part $\Delta$ and a kinetic-energy part $E_{drift}$ [28]. As shown in Fig. 1, there are two limiting cases.

(i) $\tau_{off} = 0$. When the pulse vanishes suddenly, the electron has no time to escape. It stops jittering, and $\Delta$ vanishes together with the field; we have $\kappa = 1$ and

$$E = E_{drift} = j\omega - \Delta - E_B \ . \tag{39}$$

(ii) $\tau_{off} = \infty$. When the pulse is turned off slowly, the electron has time to escape from the field. It slowly stops jittering while increasing its velocity, since it is repelled by the field. Hence we have $\kappa = 0$ and

$$E = E_{drift} + \Delta = j\omega - E_B \ . \tag{40}$$

We now use the scattering-theoretical description of models (1) and (2) of Sec. III. The total energy $\mathcal{E}$ is given in model (1) by

$$\mathcal{E} = \mathcal{E}_i = \mathcal{E}_f = -E_B + (l + 1/2)\omega = E + (m + 1/2)\omega \ , \tag{41}$$

where $E = \mathbf{P}_f^2 / 2m_e$ is the energy of the electron outside the field.

It follows from the form (29) of the final-state scattering wave function $\psi_f^-$ that $\mathcal{E}$ is also given by $\mathcal{E} = E + (n + 1/2)\omega + \Delta$, where $\Delta = (m - n)\omega$. We have

$$E = (l - m)\omega - E_B = (l - n)\omega - E_B - \Delta \ , \tag{42}$$

where $j = l - n$ is the number of photons which is absorbed by the atom *inside* the electromagnetic field. Model (1) thus predicts that the electron enters the detector with a kinetic energy $E$ that corresponds to $\tau_{off} = 0$ [cf. Eqs. (39) and (42)].

In order to see why model (1) corresponds to a sudden pulse switch-off we substitute the wave function (29) in the transition matrix element (28) and notice, according to Eqs. (17a), (19), and (30), that

$$T_{fi}^{(1)} = \int \langle \Phi_f, m | \Psi_{\mathbf{P},n} \rangle T_{fi}^{(2)}(\mathbf{P}, n) d\mathbf{P} \ , \tag{43}$$

where $\langle \Phi_f, m | \Psi_{\mathbf{P},n} \rangle = \mathcal{J}_Z^* e^{-iZ(\phi_\xi + \theta/2)} \delta(\mathbf{P}_f - \mathbf{P})$ is the probability amplitude per unit momentum that the electron finds itself in the free-electron and free-photon state $|\Phi_f, m\rangle$ if the photon-electron interaction is suddenly removed. In Eq. (43), $T_{fi}^{(2)}$ must be evaluated using $\psi_V = \Psi_{\mathbf{P},n}$, and we have $Z = m - n$. According to Eq. (A12), the use of the semiclassical scattering wave function (A10) also leads to the result (43), which shows that it follows from the sudden approximation if it is assumed that the pulse is suddenly switched off after an adiabatic rise.

Since the energy of a monochromatic field can only be changed by changing the number of photons, $Z$ must, in accordance with Fig. 1, be an integer in $\Delta = Z\omega$. The electron thus responds to the sudden drop in field intensity by the opposing effect of converting its ponderomotive potential energy entirely into free photons. As shown in the Appendix, the constraint $Z$ = integer also prevents the semiclassical off-field scattering wave function from vanishing and reduces it to the Volkov solution $\psi_{\mathbf{P}}$ multiplied by the overlap matrix element $\langle \psi_{\mathbf{P}}^Z(t) | \phi_f(t) \rangle$. According to Eqs. (A4) and (A5), this result is essentially a consequence of the Bessel-function expansion (18) of the Volkov solution (19), which leads to the proportionality relation (A9). This expansion is consistent with an adiabatic switch-on of a plane-wave electromagnetic field of infinite extent. When this spatially unlimited field is switched off, however, the electron would never become free in a finite time, and that is why the scattering wave function leads to the solution (A12) or (29). It is thus clear that a spatial confinement of the electromagnetic field in the scattering wave function (27) or its semiclassical counterpart (A1) is required for a consistent treatment of the case (37) in which only a fraction of the ponderomotive potential energy is converted into an average number of photons. This conclusion agrees with a similar suggestion by Kristić and Mittleman [30]. Nevertheless, we can use model (1) as a simple approximation to examine what happens to the photoelectron at a sharp cutoff of a not-too-short laser pulse.

Model (2) does not incorporate the escape phase of the MPI process, and thus does not imply any predictions regarding the observed photoelectron energy $E$. Instead, this model predicts that the drift energy $E_{drift}$ is given by Eq. (39). This is often taken as an explanation for the suppression of the lowest-order above-threshold-ionization (ATI) peaks, because in Eq. (39) a large $\Delta$ value prohibits the existence of $j_{min}$, and possibly, of higher $j$ values [31]. Model (2) is often used, in conjunction with the *ad hoc* hypothesis that the observed photoelectron energy is given by Eq. (40) rather than Eq. (39). In this form, the model is only applicable to the long-pulse regime [28].

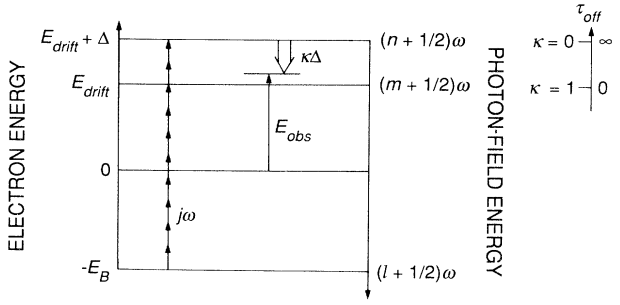


FIG. 1. Energy-balance diagram for the switching-off of a monomode electromagnetic field. The quantities shown are defined in the text in association with Eqs. (37) and (38).

### B. Strong-field multiphoton rates with application to electron detachment of $I^-$

In this subsection, we use both models (1) and (2) to simulate the behavior of ATI for $\tau_{\rm off}=0$ and $\tau_{\rm off}=\infty$ in the case of a short-range potential $U$. By setting $U=0$ in Eq. (28), it follows that the strong-field transition matrix element is

$$T_{fi}^{(1)} \simeq \langle \psi_f^- | V | \Phi_i, l \rangle \ , \tag{44}$$

where $\psi_f^-$ is the scattering wave function (29). It follows from the third expression in Eq. (30) that the corresponding transition matrix element in model (2) is given by

$$T_{fi}^{(2)} \simeq \langle \psi_V | V | \Phi_i, l \rangle \ , \tag{45}$$

where $\psi_V$ is the Volkov wave function. The amplitudes (44) and (45) are related by Eq. (43), as are the exact amplitudes. These amplitudes do not, however, provide a good description of MPI if the potential is long range.

The differential transition-rate formulas that correspond to Eqs. (44) and (45) have previously been worked out for the general case of elliptical polarization [6,7,32]. We have

$$\frac{dw^{(1)}}{d\Omega} = |\mathcal{J}_Z(\zeta,\eta,\phi_\xi)|^2 \frac{dw^{(2)}}{d\Omega} \ , \tag{46}$$

where $\mathcal{J}_Z$ is the generalized Bessel function (23) for integer $Z=\Delta/\omega$, and where we have

$$\frac{dw^{(2)}}{d\Omega} = (2m_e^3\omega^5)^{1/2}(2\pi)^{-2}|\Phi_i(\mathbf{P}-j\mathbf{k}+Z\mathbf{k})|^2 \times (j-Z)^2(j-Z-E_B/\omega)^{1/2}|\mathcal{J}_j(\zeta,\eta,\phi_\xi)|^2 \ . \tag{47}$$

Here, $\Phi_i(\mathbf{P}-j\mathbf{k}+Z\mathbf{k})$ is the Fourier transform of the initial-state wave function, corresponding to the binding energy $E_B$. The transition rates (46) and (47) pertain to a situation in which the electron is emitted into the solid angle $d\Omega$ with momentum $\mathbf{P}$ and kinetic energy $E=\mathbf{P}^2/2m_e$. The number of photons that are absorbed inside the electromagnetic field is denoted by $j$; it is constrained by Eq. (39) to be larger than or equal to $(\Delta+E_B)/\omega$. Even though the differential MPI rate (46) exists only for integer $Z$, an obvious and possible generalization is to allow for noninteger $Z$ values as well. This would mimic a monomode electromagnetic field of finite extent or a multimode field [7]. According to Sec. IV A, the rate (46) is only applicable to a situation in which the laser pulse is switched off swiftly; if applied to MPI by short pulses, there is the question of whether the condition of adiabatic switch-on is fulfilled. Nevertheless, a numerical simulation for $I^-$ shows that interference effects induced by the overlap factor $|\mathcal{J}_Z|^2$ are so dramatic that they may be observable in MPI of negative ions by short pulses.

In model (2) there is no constraint on $Z$. Equation (47) is applicable when the electron is ejected with kinetic energy $E=j\omega-E_B$ in accordance with Eq. (40), which is valid only for slow switch-off. We adopt this additional assumption in comparing the rates (46) and (47). Model (1) thus predicts that a photoionization peak observed at energy $E_2$ when produced by a slowly decaying pulse will be shifted to $E_1=E_2-\Delta$ when produced by a rapidly decaying pulse.

The photoelectron angular distributions predicted by Eqs. (46) or (47) depend on the polarization angle $\xi$. We express

$$\mathbf{P}=(P\sin\theta\cos\phi, P\sin\theta\sin\phi, P\cos\theta)$$

in a coordinate system in which the unit vectors $\epsilon_x$, $\epsilon_y$, and $\epsilon_z$ are as defined in Sec. II. Substitution of $\pi-\phi$ for $\phi$ changes $\phi_\xi$ to $\pi-\phi_\xi$, according to Eq. (22a). Since we have

$$\mathcal{J}_j(\zeta,\eta,\pi-\phi_\xi)=\mathcal{J}_j^*(\zeta,\eta,\phi_\xi) \ ,$$

the distributions predicted by Eqs. (46) and (47) are symmetrical in the $(\epsilon_x,\epsilon_y)$ plane, provided that the atoms are not aligned. Then we can sum over magnetic quantum numbers in the Fourier transforms of the initial-state wave functions, and the rates become proportional to the absolute square of

$$\Phi_i(|\mathbf{P}'|)=\int_0^\infty r^2 dr\, R_{nl}(r) j_l(|\mathbf{P}-j\mathbf{k}+Z\mathbf{k}|r) \ , \tag{48}$$

where $j_l$ is the spherical Bessel function of the first kind and $R_{nl}(r)$ is the radial wave function for principal quantum number $n$ and angular momentum quantum number $l$. For elliptical polarization, the symmetry with respect to $\phi$ is broken if the second amplitude in Eq. (28) or the corresponding amplitude in the third expression of Eq. (30) is included [27]. In the case of a long-range potential, the effect is pronounced [29].

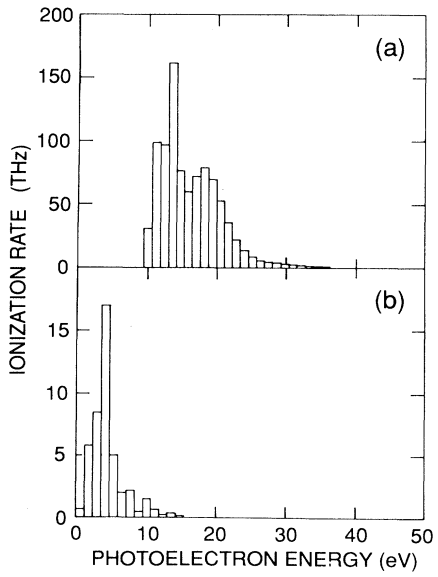


FIG. 2. ATI spectra of $I^-$ ionized by 1.064-$\mu$m 88-TW cm$^{-2}$ linearly polarized ($\xi=0°$) electromagnetic radiation: (a) $\tau_{\rm off}=\infty$, (b) $\tau_{\rm off}=0$. The rates were obtained by integrating over the solid angle in Eqs. (46) and (47).

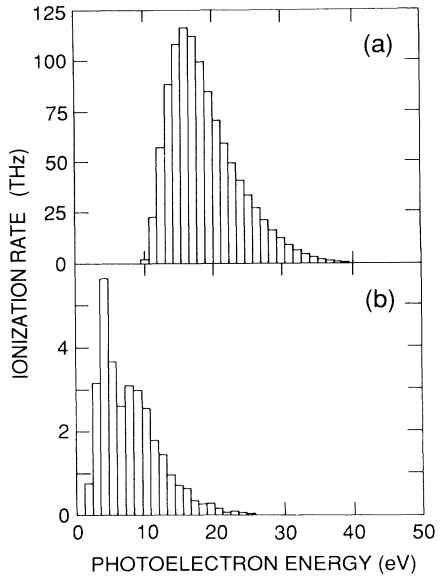


FIG. 3. ATI spectra of $I^-$ ionized by 1.064-$\mu$m 88-TW cm$^{-2}$ elliptically polarized ($\xi=50°$) electromagnetic radiation: (a) $\tau_{off}=\infty$, (b) $\tau_{off}=0$. The rates were calculated by integrating over the solid angle in Eqs. (46) and (47).

We have carried out computations of MPI rates for $I^-$ by means of a specially designed code for the calculation of the generalized Bessel functions (23) [33]. In the Fourier transform (48) we used the measured $I^-$ electron affinity [34] of 3.06 eV as the binding energy, and the $5p$ Hartree-Fock wave function. The Nd–yttrium aluminum garnet wavelength of $\lambda=1.064$ $\mu$m and intensity $I=88$ TW cm$^{-2}$ were chosen. Then we have $\Delta\cong 8\hbar\omega$, with $\hbar\omega=1.16$ eV. The calculations were carried out for different polarization states by changing $\xi$ in steps of 10° from $\xi=0°$ (linear polarization) to $\xi=90°$ (circular polarization). In Figs. 2–4, the results for $\xi=0°$, 50°, and 90° are presented.

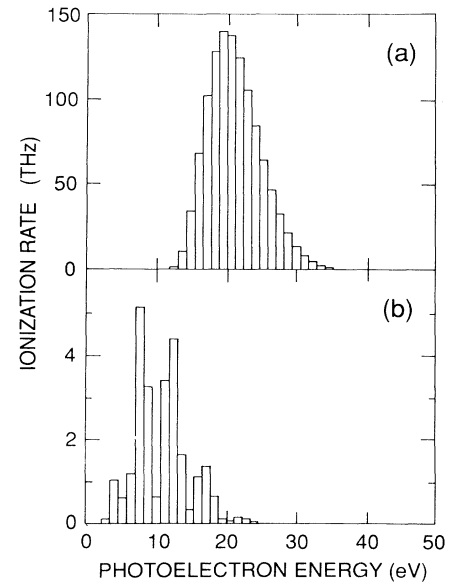


FIG. 4. ATI spectra of $I^-$ ionized by 1.064-$\mu$m 88-TW cm$^{-2}$ circularly polarized ($\xi=90°$) electromagnetic radiation: (a) $\tau_{off}=\infty$, (b) $\tau_{off}=0$. The rates were calculated by integrating over the solid angle in Eqs. (46) and (47).

The total intensities are generally lower by a factor of 10 for model (1) in comparison with model (2). The interference effects induced in the ATI spectra by the overlap factor $|\mathcal{J}_Z|^2$ are very large. The higher-order photoelectron peaks are almost wiped out by this factor if the light is linearly polarized (Fig. 2). The trend continues up to $\xi=30°$. With increasing $\xi$, the maximum of the ATI photoelectron distribution is pushed towards higher energies, and the overall shape of the distributions becomes somewhat more similar for the two models. As Fig. 3 shows, however, model (1) predicts a narrower and more asymmetric shape. As circular polarization is approached, the differences again become dramatic (Fig. 4): the model-(1) spectrum exhibits several minima, in contrast to the almost Gaussian shape of the model-(2) spectrum. It remains to be seen whether any of these effects can be observed in real experiments with pulsed high-powered lasers.

## V. CONCLUSIONS

With the aid of the many-photon correspondence principle we have shown, for an electron that interacts with a monochromatic plane-wave electromagnetic field, that in the large-photon-number limit formal scattering theory leads to exactly the same results as semiclassical time-dependent scattering theory. This conclusion holds regardless of whether the electromagnetic field has infinite extent or is spatially limited. In the semiclassical theory we have assumed that the field is switched on adiabatically; the treatment of more complex time behavior of the vector potential may require the use of time-dependent scattering theory. An alternative may be to combine formal scattering theory with the density-matrix technique; this approach has yet to be explored.

We have shown that, depending on the final-state boundary conditions, three different transition matrix elements exist for multiphoton ionization. These matrix elements correspond to entirely different interpretations of multiphoton ionization. Only the breakup case, in which it is assumed that both the atomic and electromagnetic interactions cease to have any effect at the end of the reaction, admits a consistent approach with regard to final-state interactions, as demonstrated by deriving the perturbation expansion with respect to the photon-electron interaction.

We have examined the final-state scattering wave function for the case in which both interactions are switched off at the end. It was possible to show explicitly that the semiclassical off-field scattering wave function exhibits the same behavior as the corresponding quantum-mechanical wave function in the high-photon-number limit. This proof confirms the curious fact that the final-state scattering wave function exists only if the pondero-

motive potential energy is an integer multiple of the photon energy. This strange feature can be traced to the somewhat unrealistic assumption that the electromagnetic field is monochromatic and of infinite extent, yet it leads to a simple physical interpretation of the final-state interaction between the electron and the field, as follows. The final-state wave function in this case is seen to correspond to an instantaneous switch-off of the field. When this takes place, the photoelectron opposes the decrease in photon number by converting its ponderomotive potential energy into free photons. Consequently, the above-threshold-ionization spectrum is red shifted with respect to the slow-switch-off spectrum, and it exhibits interference effects. Calculations of the above-threshold-ionization spectrum of $I^-$ explicitly illustrate this predicted behavior. It appears that the differences between the slow- and fast-switch-off spectra may have observable consequences.

## ACKNOWLEDGMENTS

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## APPENDIX: THE SEMICLASSICAL OFF-FIELD SCATTERING WAVE FUNCTION

According to the many-photon correspondence principle, the semiclassical wave function that corresponds to the scattering wave function (27) is

$$\psi_f^-(t)=|\phi_f(t)\rangle+\int dt_1\,\mathcal{G}_V^-(t,t_1)V(t_1)|\phi_f(t_1)\rangle\ , \tag{A1}$$

where $V(t_1)$ is the semiclassical photon-electron interaction, and

$$\begin{aligned}\mathcal{G}_V^-(t,t_1)&=\mathcal{G}_V^{\dagger+}(t_1,t)\\&=i\Theta(t_1-t)\int d\mathbf{P}|\psi_{\mathbf{P}}(t)\rangle\langle\psi_{\mathbf{P}}(t_1)|\end{aligned} \tag{A2}$$

is the time-advanced propagator, such that

$$\Theta(t_1-t)=\begin{cases}1\ , & t_1>t\\0\ , & t_1<t\ .\end{cases} \tag{A3}$$

We let $|\phi_f(t)\rangle$ be a plane wave with momentum $\mathbf{P}_f$ and energy $E_f=\mathbf{P}_f^2/2m_e$, and assume that a vector potential (15) with constant $\Lambda$ is used in the wave equation (A1). Then the Volkov solutions $\Psi_{\mathbf{P}}(t)$ have the form (18) with the coefficents (19) in the propagator (A2), and the matrix elements

$$V_{\mathbf{P}\mathbf{P}_f}(t_1)=\sum_{j=-\infty}^{+\infty}\langle\Psi_{\mathbf{P}}^j(t_1)|V(t_1)|\phi_f(t_1)\rangle \tag{A4}$$

can be calculated explicitly. We have

$$\begin{aligned}V_{\mathbf{P}\mathbf{P}_f}^j(t_1)&=\langle\Psi_{\mathbf{P}}^j(t_1)|V(t_1)|\phi_f(t_1)\rangle\\&=R_j(\omega,\mathcal{Z})\mathcal{J}_j(\xi,\eta,\phi_\xi)e^{ij(\phi_\xi+\theta/2)}\\&\quad\times\delta(\mathbf{P}_f-\mathbf{P}+j\mathbf{k}-\mathcal{Z}\mathbf{k})e^{iE_jt_1}\ ,\end{aligned} \tag{A5}$$

where

$$R_j=R_j(\omega,\mathcal{Z})=\omega(\mathcal{Z}-j) \tag{A6}$$

and

$$E_j=\mathbf{P}^2/2m_e-\mathbf{P}_f^2/2m_e+R_j\ . \tag{A7}$$

In the derivation of Eq. (A5) we have made use of the identity [7]

$$\frac{\xi}{2}(\mathcal{J}_{j-1}+\mathcal{J}_{j+1})+\eta(\mathcal{J}_{j-2}e^{-2i\phi_\xi}+\mathcal{J}_{j+2}e^{2i\phi_\xi})+j\mathcal{J}_j=0\ . \tag{A8}$$

We also need the matrix elements $\langle\psi_{\mathbf{P}}^j(t)|\phi_f(t)\rangle$, which can be shown to be related to the interaction matrix element (A5) by

$$V_{\mathbf{P}\mathbf{P}_f}^j(t)=R_j\langle\Psi_{\mathbf{P}}^j(t)|\phi_f(t)\rangle\ . \tag{A9}$$

If we now expand $|\phi_f(t)\rangle$ in terms of the Volkov solutions $\psi_{\mathbf{P}}(t)$, we can combine the plane-wave and propagator parts in (A1) into a single expansion. The result is

$$\psi_f^-(t)=\sum_{j=-\infty}^{+\infty}\int\left[1+i\pi\delta(E_j)R_j-\mathbf{P}\frac{R_j}{E_j}\right]|\psi_{\mathbf{P}}(t)\rangle\langle\psi_{\mathbf{P}}^j(t)|\phi_f(t)\rangle d\mathbf{P}\ , \tag{A10}$$

where we have used

$$i\int\Theta(t_1-t)e^{iE_jt_1}dt_1=\left[i\pi\delta(E_j)-\mathbf{P}\frac{1}{E_j}\right]e^{iE_jt}\ . \tag{A11}$$

Here, P means that one should take the principal part when integrating over $d\mathbf{P}=dP_xdP_ydP_z$. Since the overlap integral $\langle\psi_{\mathbf{P}}^j(t)|\phi_f(t)\rangle$ is proportional to

$$\begin{aligned}&\delta(\mathbf{P}_f-\mathbf{P}+j\mathbf{k}-\mathcal{Z}\mathbf{k})\\&\quad=\delta(P_{fx}-P_x)\delta(P_{fy}-P_y)\delta(P_{fz}-P_z-R_j)\ ,\end{aligned}$$

the integration over $d\mathbf{P}$ reduces to a one-dimensional integral over $P_z=P$. We distinguish between two cases.

(i) $R_j\neq0$. The energy factor

$$E_j=-\frac{PR_j}{m_e}-\frac{R_j^2}{2m_e}+R_j\simeq R_j\neq0$$

is almost independent of $P$, since $P\ll m_ec$ and $\omega(\mathcal{Z}-j)\ll m_ec^2$ (in SI units). The first and third approximate terms in the large parentheses in Eq. (A10) cancel and the second term vanishes. The scattering wave function $\psi_j^-(t)$ only exists up to the order of $P/m_ec$, and thus vanishes in the nonrelativistic limit.

(ii) We assume that there is an integer $j'$ such that $Z=j'$. Consequently, we have $R_{j'}=0$, and the second and third terms in the large parentheses in Eq. (A10) vanish for $j=j'$. This can be seen, either from the fact that $R_j$ appears as a factor of the $j$-dependent time integral (A11) in $\psi_f^-(t)$, or one may argue that the corresponding terms in the large parentheses of Eq. (A10) vanish. For an explicit proof, we recognize that in the second term in the large parentheses of Eq. (A10), $R_j=0$ plays the role of a well-behaved function in the definition of $\delta(E_j)=\delta(P-P_f)$, and that in the third term, $R_j$ is multiplied by

$$\mathrm{P}\int \frac{\delta(P-P_f)dP}{P^2-P_f^2}=0 .$$

This integral vanishes because $(P^2-P_f^2)^{-1}$ is an odd function of $P$, whence the principal-value interval $(P_f-\epsilon,P_f+\epsilon)$ does not contain $P_f$ for any $\epsilon$. We thus have

$$\psi_f^-(t)=\int \langle \psi_{\mathbf{P}}^Z(t)|\phi_f(t)\rangle |\psi_{\mathbf{P}}(t)\rangle d\mathbf{P} = \psi_{\mathbf{P}_f}(t)\mathcal{J}_Z(\zeta,\eta,\phi_\xi)e^{iZ(\phi_\xi+\theta/2)} , \tag{A12}$$

since the $j\neq Z$ terms do not contribute to the wave function in the nonrelativistic limit according to paragraph (i). The result (A12) is the semiclassical analog to $\psi_f^-$ in Eq. (29), in accordance with the many-electron correspondence principle.

*Present address: Department of Physics, University of Windsor, Windsor, Ontario, Canada N9B 3P4.


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