

Scattering theory of multiphoton ionization in strong fields

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Formal time-independent scattering theory is applied to multiphoton ionization of atoms in intense electromagnetic fields. The quantized-field version of the Volkov solution makes this approach possible. With the electron-photon interaction in a monochromatic photon field, it is found that, in the nonrelativistic and large-photon-number limits, the final scattering state exists only in the special case in which the ponderomotive potential per unit photon energy is an integer; otherwise the final state vanishes. In the integer case the corresponding wave function reduces to a single Volkov function, multiplied by an overlap factor. A simple interpretation of this result is given, and some other consequences of this work are discussed.

I. INTRODUCTION

Multiphoton ionization (MPI) of atoms in strong electromagnetic fields has become of growing interest as experiments with increasingly powerful lasers and low-pressure gas targets are performed.¹⁻³ Particularly intriguing is the observation of above-threshold ionization (ATI), where the photoelectron spectrum exhibits a series of peaks with spacing equal to the photon energy.^{4,5}

Lowest-order perturbation theory fails to explain such features as the shape of the envelope of the ATI photoelectron peaks or the dependence of their strength on the intensity of the laser light, typically exceeding 1 TW cm⁻². Several theoretical approaches to MPI with non-perturbative aspects have consequently been developed.⁶ Among existing schemes, a number of models make use of the Volkov⁷⁻⁹ or Volkov-Coulomb¹⁰ wave function. By Volkov state we mean the solution of the time-dependent Schrödinger equation for an unbound electron in an oscillating electromagnetic field, obtained in an appropriate gauge.¹¹ These models have their precedents in the work of Keldysh, Faisal, and Reiss¹² (KFR), who took the final state of the electron in MPI to be a Volkov state. The consistency of the KFR approach has, however, been questioned by Antunes Neto and Davidovich and Milonni.¹³ These authors argue that the KFR amplitude is effectively canceled in the conventional perturbation expansion of the transition amplitude in which the atomic binding potential is treated as a perturbation.¹⁴

The models cited above^{7-10,12} are based on the usual semiclassical time-dependent description of the laser field and on scattering-theoretical arguments. The photons and electrons are thus not treated as particles of a truly closed system to which formal scattering theory¹⁵ is applicable. The link between these two approaches has been provided by Mollow,¹⁶ who showed that, in the electric-dipole approximation, the assumption of a coherent state in the remote past in connection with the quantum-mechanical photon-electron interaction operator is equivalent to addition of the classical field operator to the quantum-mechanical one, in association with a replace-

ment of the coherent state by the vacuum state. Neglect of the quantum-mechanical operator leads to the semiclassical approximation, and so does the assumption of a very large but fixed number of photons in the initial state.¹¹ Restoration of time independence now requires a unitary transformation to a noninertial frame, as to a rotating frame, in the circularly polarized case.¹ Subtle questions consequently remain regarding the applicability of formal scattering theory to MPI in the semiclassical approximation, not unrelated to the proposed inconsistency¹³ of the KFR approach.

A set of exact solutions of the Dirac equation for an electron in a quantized, elliptically polarized, monochromatic electromagnetic field has recently been found.¹⁷ This quantized-field version of the Volkov solution enables one to treat MPI as a genuine scattering process in an isolated system that consists of photons and an atom. Energy is conserved throughout the interaction, resulting in a free electron which has kinetic energy equal to the energy of the absorbed photons minus its initial binding energy. Formal time-independent scattering theory¹⁵ thus is applicable.

Here we outline the treatment of MPI in terms of scattering theory and within the framework of quantum electrodynamics (QED). We assume that the electron-photon interaction takes place in a single-mode quantized photon field with a fixed number of photons in the initial state. This assumption *per se* does not provide a realistic description of strong laser fields used in current experiments.^{4,5} Our approach may be considered, however, as the first step towards a more complete theory based on a multimode field, in which the pulsed-laser interaction and the photon statistics are described in terms of time-dependent density matrices. Secondly, we make a consistent application of scattering theory to the entire MPI process in the nonrelativistic and large-photon-number limits which correspond to the single-mode semiclassical approximation of the vector potential.¹¹ Our theory thus incorporates the escape of the electron from the laser field as a part of the scattering process, also in this approximation, in contrast to previous work in which the

escape is either ignored or treated separately from MPI. Although the latter procedure may account realistically for the decay of the laser pulse, it on the other hand involves the approximation of treating MPI as a two-step process, in contrast to the present scattering approach. It is hence of interest to compare, in the limit of a vanishing Coulomb potential, the genuine scattering amplitude with the KFR amplitude which follows from the scattering theory and QED in the same way, provided that the final scattering state is replaced by the quantized Volkov state.¹⁷

In this work we thus examine the form of the scattering amplitude in which the final scattering wave function arises from the scattering of the electron by the electromagnetic potential. As a result we show that, in general, the final scattering state does not reduce to a quantized Volkov state by examining the case in which the ponderomotive energy is an integer multiple of the photon energy. In the nonrelativistic and large-photon-number limits, however, the final state reduces to a single but modified Volkov state if the ponderomotive energy is an integer multiple of the photon energy. Otherwise, the final state vanishes. The scattering amplitude consequently only exists in the integer case, where it becomes a product of the original KFR amplitude and the overlap between the final plane-wave state and the Volkov state. A simple interpretation of this result is presented. Suggestions are also made as to how the restrictions on the ponderomotive potential may be removed in a more realistic nonrelativistic theory.

II. THEORY

A. Quantum-electrodynamical Volkov solution

In the following we use relativistic units, $c = \hbar = 1$, and the metric tensor $g_{\mu\nu}$ with $g_{00} = 1$ and $g_{11} = g_{22} = g_{33} = -1$ and $g_{\mu\nu} = 0$ ($\mu \neq \nu$). The scalar product of two four-vectors is defined as $ab = g_{\mu\nu} a^\mu b^\nu$, and the γa scalar product is denoted by \not{a} ; γ stands for 4×4 Dirac matrices.

The Hamiltonian of a single atomic electron in a quantized monochromatic electromagnetic radiation field is

$$H = H_\gamma + H_e + U + V, \quad (1)$$

where

$$H_\gamma = \frac{1}{2} \omega (a^\dagger a + a a^\dagger) \quad (2)$$

is the free-photon energy operator, and

$$H_e = \gamma^0 [\gamma \cdot (-i\partial) + m_e] \quad (3)$$

is the free-electron energy operator, and where

$$U = U(\mathbf{r}) \quad (4)$$

is the atomic Coulomb potential due to the nucleus and the other electrons in the atom. The electron-photon interaction is

$$V = -e \gamma^0 \gamma \cdot \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}). \quad (5)$$

In Eq. (3), m_e is the electron rest mass; the electron

charge is denoted by $e = -|e|$.

The photon field is assumed to be elliptically polarized, described by

$$\mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) = g(\epsilon a e^{ik \cdot \mathbf{r}} + \epsilon^* a^\dagger e^{-ik \cdot \mathbf{r}}), \quad (6)$$

where we have

$$g \equiv (2V_\gamma \omega)^{-1/2} \quad (7)$$

and the polarization vectors satisfy

$$\begin{aligned} \epsilon \cdot \epsilon &= \epsilon^* \cdot \epsilon^* = \cos \xi, \\ \epsilon \cdot \epsilon^* &= 1. \end{aligned} \quad (8)$$

Here, V_γ is the normalization volume of the photon field and ω is the photon frequency. The angle ξ monitors the degree of polarization, such that $\xi = \pi/2$ corresponds to circular polarization and $\xi = 0$, to linear polarization.

The eigenstates of $H_V = H_\gamma + H_e + V$ can be found exactly.¹⁷ The solution of

$$H_V \Psi_{\mathbf{P}sn} = (E + C_{ns} \omega) \Psi_{\mathbf{P}sn}, \quad (9)$$

where $(E, \mathbf{P}) \equiv P$ is the electron four-momentum on the mass shell, is

$$\begin{aligned} \Psi_{\mathbf{P}sn} &= V_e^{-1/2} \exp[i(\mathbf{P} \cdot \mathbf{r} + C_{ns} \mathbf{k} \cdot \mathbf{r} - \mathbf{k} \cdot \mathbf{r} N_a)] \\ &\times \left[1 + e \frac{\not{K} \not{A}}{2kP} \right] D_P^\dagger |n\rangle_c \chi_s, \end{aligned} \quad (10)$$

where V_e is the normalization volume of the electron, k stands for (ω, \mathbf{k}) , and

$$\begin{aligned} C_{ns} &= (kP)^{-1} [C(n + \frac{1}{2}) + \frac{1}{2} e^2 g^2 s \\ &\quad - e^2 g^2 C^{-1}(P \epsilon_c)(P \epsilon_c^*)], \end{aligned} \quad (11)$$

with

$$C = [(kP + e^2 g^2)^2 - e^4 g^4 \cos^2 \xi]^{1/2}. \quad (12)$$

In Eq. (10), the coordinate-independent photon field operator

$$A = g(\epsilon a + \epsilon^* a^\dagger) \quad (13)$$

involves the polarization four-vectors $\epsilon = (0, \boldsymbol{\epsilon})$ and $\epsilon^* = (0, \boldsymbol{\epsilon}^*)$, respectively. According to Eqs. (8), they satisfy the relations

$$\begin{aligned} \epsilon^* \epsilon &= -1, \\ \epsilon \epsilon &= \epsilon^* \epsilon^* = -\cos \xi. \end{aligned} \quad (14)$$

The photon number operator is denoted by

$$N_a = \frac{1}{2} (a^\dagger a + a a^\dagger), \quad (15)$$

and

$$N_c = \frac{1}{2} (c^\dagger c + c c^\dagger) \quad (16)$$

is a hyperbolically rotated photon number operator. The rotation is defined by a Bogoliubov transformation, introduced to cancel the quadratic term in solving Eq. (9).¹⁷

$$\begin{aligned} c &= a \cosh \chi - a^\dagger \sinh \chi, \\ c^\dagger &= -a \sinh \chi + a^\dagger \cosh \chi, \end{aligned} \quad (17)$$

where

$$\chi = -\frac{1}{2} \tanh^{-1} \frac{e^2 g^2 \cos \xi}{kP + e^2 g^2}. \quad (18)$$

The eigenstate of N_c is

$$|n\rangle_c = \frac{(c^\dagger)^n}{(n!)^{1/2}} |0\rangle_c \quad (19)$$

and the rotated vacuum state is

$$\begin{aligned} |0\rangle_c &= (\cosh \chi)^{-1/2} \sum_{m=0}^{\infty} (\tanh \chi)^m \left[\frac{(2m-1)!!}{(2m)!!} \right]^{1/2} \\ &\quad \times |2m\rangle. \end{aligned} \quad (20)$$

with $(-1)!! \equiv 1$. The rotated polarization vectors in Eq. (11) are thus given by

$$\begin{aligned} \epsilon_c &= \epsilon \cosh \chi + \epsilon^* \sinh \chi, \\ \epsilon_c^* &= \epsilon \sinh \chi + \epsilon^* \cosh \chi. \end{aligned} \quad (21)$$

In the N_a representation the shift operator is

$$D_p^\dagger = \exp(\delta_a a^\dagger - \delta_a^* a), \quad (22)$$

where the parameter δ_a is defined as

$$\delta_a = -egP[\epsilon^* \cosh(2\chi) + \epsilon \sinh(2\chi)]/C. \quad (23)$$

In Eq. (10), the shift operator acts on the photon eigenstate (19) which can by virtue of Eq. (17) be transformed into the N_a representation.

We use a projection operator in the spinor space,¹⁷

$$\mathcal{P} = (\mathbf{P} + m_e) \mathbf{k} / 2kP \quad (24)$$

and a polarization-dependent spin operator

$$\mathcal{S} = \frac{1}{2} [\boldsymbol{\epsilon}^*, \boldsymbol{\epsilon}] \quad (25)$$

with eigenvalues

$$s = \pm \sin \xi \quad (26)$$

for the description of the bispinor

$$\chi_s = \mathcal{P}(\mathcal{S} + s)v. \quad (27)$$

Here, v is an arbitrary but properly normalized bispinor.

It can be shown¹⁸ that if $\mathbf{p} = \mathbf{P} + C_{ns} \mathbf{k}$ is subject to a box boundary condition, then the wave functions (10) satisfy the orthogonality relation

$$\langle \bar{\Psi}_{\mathbf{P}'s'n'} | \gamma^0 | \Psi_{\mathbf{P}sn} \rangle = J^0 \delta_{\mathbf{P}'\mathbf{P}} \delta_{s's} \delta_{n'n}, \quad (28)$$

where we have

$$J^0 = m_e^{-1} [E + C_{ns} \omega - \delta_a \delta_a^* \omega - (n + \frac{1}{2}) \omega \cosh(2\chi)]. \quad (29)$$

The bispinors (10) consequently form a complete set with respect to the momentum, photon, and spinor space.

B. Transition matrix element

Using formal scattering theory,¹⁵ the transition matrix element for MPI can be written

$$T_{fi} = \langle \Phi_f, m | V | \Psi_i^{(+)} \rangle, \quad (30)$$

where i and f refer to the initial and final state, respectively. Here, $|\Psi_i^{(+)}\rangle$ is a scattering solution that satisfies

$$|\Psi_i^{(+)}\rangle = |\Phi_i, l\rangle + \frac{1}{\mathcal{E} - K - V - U + i\epsilon} V |\Phi_i, l\rangle, \quad (31)$$

where $|\Phi_f, m\rangle \equiv \Phi_f \otimes |m\rangle$, $|\Phi_i, l\rangle \equiv \Phi_i \otimes |l\rangle$. In the direct products Φ_i and Φ_f are atomic bound and continuum states, respectively, which pertain to the Coulomb potential U , while $|l\rangle$ and $|m\rangle$ are free photon number states. The “kinetic energy” operator $K = H_\gamma + H_e$ is the sum of the free-photon-energy operator (2) and the free-electron-energy operator (3). Thus we have

$$(K + U - \mathcal{E}) |\Phi_i, l\rangle = 0, \quad (32)$$

$$(K + U - \mathcal{E}) |\Phi_f, m\rangle = 0,$$

which indicates energy conservation for the entire system from beginning to end.

After some algebraic manipulation, we find

$$T_{fi} = \langle \Psi_f^- | V | \Phi_i, l \rangle + \langle \Psi_f^- | UG^+ V | \Phi_i, l \rangle, \quad (33)$$

where

$$\Psi_f^- = (1 + G_v^- V) |\phi_f, m\rangle. \quad (34)$$

We have $|\phi_f, m\rangle \equiv \phi_f \otimes |m\rangle$, ϕ_f is the final electron plane wave, and

$$\begin{aligned} G^+ &= (\mathcal{E} - H + i\epsilon)^{-1}, \\ G_v^\pm &= (\mathcal{E} - K - V \pm i\epsilon)^{-1}. \end{aligned} \quad (35)$$

By expansion, we have

$$G^+ = G_v^+ + G_v^+ UG^+ \quad (36)$$

and G_v^\pm can be expressed according to Eqs. (9), (10), (28), and (29) in calculable form as

$$G_v^\pm = \sum_{ns} V_e (2\pi)^{-3} \int d^3\mathbf{P} \frac{1}{J^0} \frac{|\Psi_{\mathbf{P}sn}\rangle \langle \Psi_{\mathbf{P}sn}|}{\mathcal{E} - E - C_{ns} \omega \pm i\epsilon}. \quad (37)$$

Now we can express the final scattering state (34) as

$$\Psi_f^- = \Psi_f^0 + i\pi \sum_{\mu \in (\mathbf{P}, s, n)} \frac{1}{J^0} |\Psi_\mu\rangle \langle \Psi_\mu | V | \phi_f, m \rangle \delta(\mathcal{E}_\mu - \mathcal{E}), \quad (38)$$

where

$$\Psi_f^0 = |\phi_f, m\rangle + \mathbf{P} \frac{1}{\mathcal{E} - K - V} V |\phi_f, m\rangle \quad (39)$$

and the Ψ_μ are the relativistic quantized-field Volkov solutions (10) in shorthand notation. Here, \mathbf{P} means the principal value.

Using Eq. (36) and the Green's operator (37), the transition matrix element can be calculated order by order according to the power expansion of the Coulomb potential

U. In lowest order, the transition matrix element is then

$$T_{fi} = \langle \Psi_f^- | V | \Phi_i, l \rangle, \quad (40)$$

where Ψ_f^- is given by Eq. (38).

C. Nature of the final scattering state

The question that has remained unanswered since Keldysh's pioneering work¹² is whether or not Ψ_f^- is a single Volkov solution. The following three theorems, proved in the Appendix, lead to the answer.

Let $z\omega$ be the ponderomotive energy of Ψ_{Psn} , so that

$$z = C_{ns} - (n + \frac{1}{2}). \quad (41)$$

From this definition it follows that $z\omega$ reduces in the non-relativistic semiclassical limit to the usual expression for the time-averaged energy of the quiver motion of an electron in an oscillating electric field.¹⁷

The first two theorems pertain to the Volkov solutions (10) for the quantized field.

Theorem 1. If Ψ_{Psn} and a plane wave $|\phi_{P's}, m\rangle \equiv \phi_{P's} \otimes |m\rangle$ have the same energy eigenvalue, then

$$\langle \Psi_{Psn} | \phi_{P's}, m \rangle = 0 \quad (42)$$

whenever z is not an integer equal to $j = m - n$.

Theorem 2. If Ψ_{Psn} and a plane wave $|\phi_{P's}, m\rangle$ have the same energy eigenvalue, then

$$\langle \Psi_{Psn} | V | \phi_{P's}, m \rangle = 0 \quad (43)$$

whenever z is not an integer equal to $j = m - n$.

The key point in proving these theorems (Appendix) is to note that the matrix elements in Eqs. (42) and (43) vanish unless we have four-momentum conservation:

$$(E + (n + \frac{1}{2})\omega + z\omega, \mathbf{P} + (n + \frac{1}{2})\mathbf{k} + z\mathbf{k}) \\ = (E' + (m + \frac{1}{2})\omega, \mathbf{P}' + (m + \frac{1}{2})\mathbf{k}), \quad (44)$$

where $(P) = (E, \mathbf{P})$ and $(P') = (E', \mathbf{P}')$ are both on the electron mass shell. Hence $P = P' + (j - z)k$, where $P^2 = P'^2 = m_e^2$ and $k^2 = 0$. Since $kP' > 0$, Eqs. (42) and (43) must be valid except when

$$z = j = m - n. \quad (45)$$

The third theorem is more general. It applies to a complete set $\{\Psi_\nu\}$ of an interacting system and a wave function ϕ_μ of a noninteracting system in which the indices $\nu = \mu$ indicate the same energies.

Theorem 3. For a complete set $\{\Psi_\nu\}$ and ϕ_μ satisfying

$$(\mathcal{E}_\nu - K - V)\Psi_\nu = 0, \quad (46)$$

$$(\mathcal{E}_\mu - K)\phi_\mu = 0,$$

respectively, the Ψ_ν obey the orthogonality relation

$$\langle \Psi_\lambda | \Psi_\nu \rangle = \delta_{\lambda\nu} \quad (47)$$

whether or not $\mathcal{E}_\lambda = \mathcal{E}_\nu$. The necessary and sufficient conditions for a $\Psi_\mu \in \{\Psi_\nu\}$ to satisfy

$$N\Psi_\mu = \phi_\mu + P \frac{1}{\mathcal{E}_\mu - K - V} V \phi_\mu, \quad (48)$$

where N is a constant, are (i)

$$\langle \Psi_{\mu'} | \phi_\mu \rangle = N\delta_{\mu\mu'}, \quad (49)$$

for the case $\mathcal{E}_{\mu'} = \mathcal{E}_\mu$ and (ii)

$$\vec{K} = \vec{K}^\dagger \quad (50)$$

between $\{\Psi_\nu^\dagger\}$ and ϕ_μ . Here, the notation \vec{K} is intended to emphasize that K acts upon the quantities to its right, the linear space of kets, and \vec{K}^\dagger , that the adjoint operator K^\dagger acts on the quantities to its left, the linear space of bras.¹⁹ Condition (50) is thus equivalent to the Hermiticity condition.

When Ψ_{Psn} and $|\phi_f, m\rangle$ have the same energy eigenvalue and $z = m - n$, the fact that $\langle \Psi_{Psn} | V | \phi_f, m \rangle$ does not vanish shows that the final scattering-state wave function (38) is a true scattering solution incorporating the δ -function part. Thus it cannot be proportional to a single relativistic quantized-field Volkov solution. The non-Hermiticity of K between $\{\Psi_{Psn}^\dagger\}$ and $|\phi_f, m\rangle$, apparent from Eqs. (42) and (43), and Theorem 3 implies that its standing-wave part (39) also is not proportional to that kind of Volkov solution.

In the large-photon-number and nonrelativistic limits, however, the situation is quite different. Using formulas¹⁷ which relate $\langle l | D_p^\dagger | m \rangle$ and $\langle m | n \rangle_c$ to Bessel functions, it can be shown that the quantum-electrodynamical Volkov solution (10) in this case reduces to

$$\Psi_{Pn} = V e^{-1/2} e^{i(\mathbf{P} + z\mathbf{k})\cdot\mathbf{r}} \sum_{j=-n}^{\infty} \mathcal{J}_j^* e^{-ij\phi_\xi} e^{-ij\mathbf{k}\cdot\mathbf{r}} |n+j\rangle, \quad (51)$$

where the \mathcal{J}_j are elliptically polarized Bessel functions, defined in terms of ordinary Bessel functions as¹⁷

$$\mathcal{J}_j = \mathcal{J}_j(\xi, \eta, \phi_\xi) = \sum_{m=-\infty}^{\infty} J_{-j-2m}(\xi) J_m(\eta) \exp(2im\phi_\xi), \quad (52)$$

where

$$\xi = \frac{2|e|\Lambda}{kP} |\mathbf{P}\cdot\boldsymbol{\epsilon}| \rightarrow \frac{2|e|\Lambda}{m_e\omega} |\mathbf{P}\cdot\boldsymbol{\epsilon}|, \\ \eta = (z/2)\cos\xi, \\ \phi_\xi = \tan^{-1}[(P_y/P_x)\tan(\xi/2)]. \quad (53)$$

We further have

$$\Lambda = g\sqrt{n} = (n/2V_\gamma\omega)^{1/2}, \quad (54)$$

which gives the maximum strength of the vector potential in the semiclassical approximation.¹¹ It follows from Eqs. (10) and (11) that the ponderomotive (potential) energy divided by the photon energy [Eq. (41)] reduces in the limit of large n to

$$z = \frac{e^2\Lambda^2}{kP} \rightarrow \frac{e^2\Lambda^2}{m_e\omega}. \quad (55)$$

The Volkov solutions (51) form an orthonormal set,¹⁸ and satisfy the nonrelativistic counterpart of the Schrödinger equation (9), where $H_e + V$ is now obtained by substituting the canonical momentum $\hat{\mathbf{p}}_\xi = \hat{\mathbf{p}} - e \mathbf{A}$ in the nonrelativistic kinetic energy operator $\hat{\mathbf{p}}_\xi^2/2m_e$. Thus we have

$$V = -(e/2m_e)[\hat{\mathbf{p}} \cdot \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) + \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) \cdot \hat{\mathbf{p}}] \\ + (e^2/2m_e) \mathbf{A}^2(-\mathbf{k} \cdot \mathbf{r}), \quad (56)$$

where $\mathbf{A}(-\mathbf{k} \cdot \mathbf{r})$ is given by Eq. (6) with ga and ga^\dagger replaced by Λ .

A direct calculation using the wave functions (51) and interaction (56) shows that we have

$$\langle \Psi_{\mathbf{P}_n} | V | \phi_f, m \rangle = \omega(z-j) \mathcal{J}_j e^{ij\phi_\xi} \frac{(2\pi)^3}{V_e} \\ \times \delta(\mathbf{P}_f - \mathbf{P} + j\mathbf{k} - z\mathbf{k}), \quad (57)$$

where $j = m - n$. A similar calculation gives

$$\langle \Psi_{\mathbf{P}_n} | \phi_f, m \rangle = \mathcal{J}_j e^{ij\phi_\xi} \frac{(2\pi)^3}{V_e} \delta(\mathbf{P}_f - \mathbf{P} + j\mathbf{k} - z\mathbf{k}). \quad (58)$$

The results which also follow from Eqs. (A1) and (A3) by taking the appropriate limits are valid whether or not $|\Psi_{\mathbf{P},n}\rangle$ and $|\phi_f, m\rangle$ correspond to equal eigenenergies.

The energy eigenvalue of the free state $|\phi_f, m\rangle$ is

$$\mathcal{E} = E_f + (m + \frac{1}{2})\omega, \quad (59)$$

while the energy eigenvalue of the nonrelativistic Volkov state (51) is

$$\mathcal{E}_\mu = E + (n + \frac{1}{2})\omega + z\omega. \quad (60)$$

The relationship between $E_f = \mathbf{P}_f^2/2m_e$ and $E = \mathbf{P}^2/2m_e$ is determined through the δ function in Eqs. (57) or (58), whence we have

$$E = E_f + (j-z)\omega(\mathbf{P}_f/m_e) + (j-z)\omega[(j-z)\omega/2m_e]. \quad (61)$$

Omitting quantities which are of the order of $\mathbf{P}_f/m_e c$ or higher (in ordinary units), we find the energy difference

$$\mathcal{E}_\mu - \mathcal{E} = (z-j)\omega. \quad (62)$$

Combining this result with Eqs. (57) and (58), we obtain the relation which indicates the Hermiticity of $K = \hat{\mathbf{p}}^2/(2m_e) + H_\gamma$ between the two kinds of wave functions:

$$\langle \Psi_{\mathbf{P}_n} | V | \phi_f, m \rangle = (\mathcal{E}_\mu - \mathcal{E}) \langle \Psi_{\mathbf{P}_n} | \phi_f, m \rangle. \quad (63)$$

This result, together with the property $(\mathcal{E}_\mu - \mathcal{E})\delta(\mathcal{E}_\mu - \mathcal{E}) = 0$, shows that the δ -function part of the final scattering state (38) vanishes in the large-photon-number and nonrelativistic limits. Consequently, in these limits,

$$\Psi_f^- = \Psi_f^0 \quad (64)$$

always holds, whether or not \mathcal{E}_μ equals \mathcal{E} , i.e., z equals j .

Since $J^0 \rightarrow 1$ according to Eq. (29), we have

$$\Psi_f^- = |\phi_f, m\rangle + \sum_{\substack{\mu \in (\mathbf{P}, n) \\ \mathcal{E}_\mu \neq \mathcal{E}}} \frac{|\Psi_\mu\rangle \langle \Psi_\mu | V | \phi_f, m \rangle}{\mathcal{E} - \mathcal{E}_\mu} \\ = |\phi_f, m\rangle - \sum_{\substack{\mu \in (\mathbf{P}, n) \\ \mathcal{E}_\mu \neq \mathcal{E}}} |\Psi_\mu\rangle \langle \Psi_\mu | \phi_f, m \rangle \\ = \sum_{\mathcal{E}_\mu = \mathcal{E}} |\Psi_\mu\rangle \langle \Psi_\mu | \phi_f, m \rangle. \quad (65)$$

According to Eq. (62), the condition of $\mathcal{E}_\mu = \mathcal{E}$ requires that $z = j = m - n$ is an integer. In that case only one value of n appears in the third sum of Eq. (65), uniquely determined by $n + z(n) = m$. In view of Eq. (57), the final-state scattering wave function is thus given by

$$\Psi_f^- = \Psi_{\mathbf{P}_f, n} \mathcal{J}_z e^{iz\phi_\xi}, \quad (66)$$

where $\Psi_{\mathbf{P}_f, n}$ is the nonrelativistic classical time-independent Volkov solution (51). The arguments in \mathcal{J}_z are given by Eq. (53), where $\mathbf{P} = \mathbf{P}_f$.

If z is not an integer, we have $\mathcal{E}_\mu \neq \mathcal{E}$, and the last sum in Eq. (65) is over an empty set. Consequently in this case the final state must vanish:

$$\Psi_f^- = 0. \quad (67)$$

This result can also be found through explicit evaluation, which shows that the first expression in Eq. (65) becomes

$$\Psi_f^0 = V_e^{-1/2} e^{i\mathbf{P} \cdot \mathbf{r}} |m\rangle \\ - V_e^{-1/2} e^{i\mathbf{P} \cdot \mathbf{r}} \sum_{j, j'} \mathcal{J}_j^* \mathcal{J}_{j-j'} e^{-ij' \mathbf{k} \cdot \mathbf{r}} e^{-ij' \phi_\xi} |m + j'\rangle. \quad (68)$$

From the algebraic identity

$$\sum_{j'=-\infty}^{\infty} \mathcal{J}_j^* \mathcal{J}_{j-j'} = \delta_{j0} \quad (69)$$

Eq. (67) follows.

As pointed out in the Appendix, Theorems 1–3 still hold in the large-photon-number nonrelativistic limits. This fact provides an alternative proof of Eqs. (66) and (67). The intermediate result (64) follows from Theorem 2 and Eq. (57), whereas Theorems 1 and 3 combined with Eq. (58) immediately lead to the final result. The results of Eqs. (66) and (67) correspond to the $N \neq 0$ and $N = 0$ cases, respectively, of Theorem 3.

III. DISCUSSION

The analysis of the quantized Volkov solutions (10) and of the final scattering-state wave function (38) requires further detailed considerations, part of which are in progress.¹⁸ We thus limit the present discussion to the nonrelativistic solution (51) and the corresponding final scattering state (66).

As indicated by Eq. (56), we do not make any use of the dipole approximation. The wave function (51) conse-

quently is more general than the usual zeroth-order solution¹¹ of the Floquet equation for radiative potential scattering in the dressed-oscillator representation. If it is used as a final state in the lowest-order transition matrix element (40), and if no constraint is put on z it leads to a transition-rate formula for elliptically polarized light which has been derived previously from the KFR approach⁹ and also from quantum electrodynamics¹⁷ by taking the large-photon-number and nonrelativistic limits.

If we use the final scattering state (66) in the lowest-order transition matrix element (40) in combination with the constraint $z = m - n$, we can construct a differential transition-rate formula for ionization of electrons which have absorbed $q = l - m$ photons. The result is

$$\frac{dw}{d\Omega} = (2m_e^3 \omega^5)^{1/2} (2\pi)^{-2} |\Phi_i(\mathbf{P} - q\mathbf{k})|^2 q^2 (q - E_B/\omega)^{1/2} \times |\mathcal{J}_{q+z}|^2 |\mathcal{J}_z|^2, \quad (70)$$

where $\Phi_i(\mathbf{P} - q\mathbf{k})$ is the Fourier transform of the initial-state wave function, corresponding to the binding energy E_B . The angular and polarization dependences of the rate (70) are determined by $\mathbf{P} - q\mathbf{k}$ in Φ_i and by the arguments (53) of the two elliptically polarized Bessel functions \mathcal{J}_i . Equation (70) deviates from the original KFR formula^{9,17} in two respects: $l - n - z$ is replaced by q and the resulting rate is multiplied by the modulation factor $|\mathcal{J}_z|^2$.

According to Eqs. (58), (65), and (66), the rate (70) can also be obtained in the large-photon-number and nonrelativistic limits from the transition matrix element

$$T = \langle \phi_{f,m} | \Psi_{P,n} \rangle \langle \Psi_{P,n} | V | \Phi_i, l \rangle,$$

provided we set $z = m - n$ in $\Psi_{P,n}$. Otherwise T vanishes. This result has a simple physical interpretation. The interaction element containing V represents the ionization of the electron *into* the field by absorption of $j = l - n$ photons. The overlap factor thus represents the probability of finding the electron in the free-electron plane-wave state $\phi_{f,m}$ when the electron “suddenly” leaves the field. Thereby it stops to jitter by converting its ponderomotive energy $z\omega$ into that of $m - n$ emitted photons. The net effect is the absorption of $q = j - z = l - m$ photons, such that the electron enters the detector with kinetic energy $E_f = q\omega - E_B$. This interpretation is consistent with the formal scattering approach in Sec. II which incorporates energy conservation for the complete system of photons and electrons throughout the entire MPI process, with the field turned off in the remote past and future.

Until now we have considered the condition $z = m - n$ to be literally true. In reality, this condition should only be interpreted in an average sense, i.e., $z_{av} = (m - n)_{av}$, where the average depends on the kind of photon statistics and mode configurations that are used to character-

ize the field. In this model, the integer orders of the Bessel functions which occur in the transition-rate formula (70) are replaced by real numbers. Furthermore, a realistic description of the laser pulse may require the introduction of time- and position-dependent density matrices and characterization of its temporal and spatial behavior.

In order to examine whether the MPI process must be treated as a single-step process, it might be of interest to compute the rate (70) for some test cases, and to compare the results with the KFR rate.²⁰ The simplest choice of z would be to take it equal to the integer which is nearest to the ponderomotive energy divided by the frequency, at various representative intensities. The crucial point is whether Eq. (70) leads to a suppression of low-order ATI peaks⁵ at high intensities. Whereas KFR and related models⁸ attribute this suppression to the constraint $q \geq z + E_B/\omega$, it can in the present formulation only come from the overlap factor $|\mathcal{J}_z|^2$ in combination with the factor $(q - E_B/\omega)^{1/2}$, which vanishes for $q\omega = E_B$.

IV. CONCLUSIONS

The application of formal time-independent scattering theory to the MPI process in a quantized single-mode electromagnetic field leads to the result that in the nonrelativistic and large-photon-number limits the final scattering state based on the photon-electron interaction vanishes unless the ponderomotive energy per unit frequency is an integer. If this condition is fulfilled, the final-scattering-state wave function becomes proportional to a single Volkov solution. These results hold whether the Coulomb field is included or not. A transition-rate formula has been derived in the limit of vanishing Coulomb field. It modifies the results of the Keldysh-Faisal-Reiss approach in accordance with energy conservation during the entire multiphoton ionization process. Whether the present approach leads to improved agreement between theory and experiment in the long-pulse regime²¹ remains to be seen.

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APPENDIX

1. Proof of Theorem 1

The plane wave has the form $|\phi_{P's', m}\rangle = V_e^{-1/2} e^{i\mathbf{P}' \cdot \mathbf{r}} \chi_{s'}^m$. Direct calculation leads to

$$\langle \Psi_{Psn} | \phi_{P's', m} \rangle = \frac{(2\pi)^3}{V_e} \left\langle n \left| c D_P \chi_s^\dagger \left[1 + e \frac{A\mathbf{k}}{2kP} \right] \gamma^0 \chi_{s'}^m \right| m \right\rangle \delta(\mathbf{P}' - \mathbf{P} + j\mathbf{k} - z\mathbf{k}), \quad (A1)$$

where $j = m - n$.

In addition to energy conservation,

$$E' + j\omega = E + z\omega, \quad (\text{A2})$$

we also have momentum conservation:

$$\mathbf{P}' + j\mathbf{k} = \mathbf{P} + z\mathbf{k}. \quad (\text{A3})$$

The four-momentum conservation reads

$$P' + (j - z)k = P. \quad (\text{A4})$$

We square both sides of Eq. (A4) and after cancellation have

$$(E' - P'_z)\omega(j - z) = 0. \quad (\text{A5})$$

If $j \neq z$, Eq. (A5) requires $E' = P'_z c$ (in ordinary units), which is clearly incompatible with $|P'|c < E'$ for a particle with $v < c$. The argument in the δ function of Eq. (A1) therefore cannot vanish simultaneously with $\mathcal{E}_\mu - \mathcal{E} = 0$.

2. Proof of Theorem 2

We introduce an interaction picture that can be called the photon momentum picture, by shifting the wave function by a factor $\exp(iN_a \mathbf{k} \cdot \mathbf{r})$. This transformation leads to

$$\begin{aligned} \Psi_{\mathbf{P}sn} &\rightarrow e^{iN_a \mathbf{k} \cdot \mathbf{r}} \Psi_{\mathbf{P}sn} = V_e^{-1/2} \exp\{i[\mathbf{P} \cdot \mathbf{r} + (n + \frac{1}{2} + z)\mathbf{k} \cdot \mathbf{r}]\} \left[1 + e^{-\frac{kA}{2kP}}\right] D_P^\dagger |n\rangle_c \chi_s, \\ |\phi_{\mathbf{P}'s'}, m\rangle &\rightarrow e^{iN_a \mathbf{k} \cdot \mathbf{r}} |\phi_{\mathbf{P}'s'}, m\rangle = e^{i[m + (1/2)]\mathbf{k} \cdot \mathbf{r}} |\phi_{\mathbf{P}'s'}, m\rangle, \\ V &\rightarrow e^{iN_a \mathbf{k} \cdot \mathbf{r}} V e^{-iN_a \mathbf{k} \cdot \mathbf{r}} = -e\gamma^0 \boldsymbol{\gamma} \cdot \mathbf{A}, \end{aligned} \quad (\text{A6})$$

where $\mathbf{A} = g(\boldsymbol{\epsilon}a + \boldsymbol{\epsilon}^* a^\dagger)$, independently of the spatial coordinates. In this picture it is easily seen that the following relation holds:

$$\langle \Psi_{\mathbf{P}sn} | V | \phi_{\mathbf{P}'s'}, m \rangle = \frac{(2\pi)^3}{V_e} \left\langle n \left| D_P \chi_s^\dagger \left[1 + e^{-\frac{Ak}{2kP}}\right] (-e\boldsymbol{\gamma} \cdot \mathbf{A}) \chi'_s \right| m \right\rangle \delta(\mathbf{P}' - \mathbf{P} + j\mathbf{k} - z\mathbf{k}). \quad (\text{A7})$$

The same reasoning as in the proof of Theorem 1 leads to the same conclusion here.

3. Proof of Theorem 3

(1) *Necessary condition.* Let

$$N\Psi_\mu = \phi_\mu + P \frac{1}{\mathcal{E}_\mu - K - V} V\phi_{\mu'};$$

then we have

$$N|\Psi_\mu\rangle = |\phi_\mu\rangle + \sum_{\substack{\nu \\ \mathcal{E}_\nu \neq \mathcal{E}_\mu}} \frac{|\Psi_\nu\rangle \langle \Psi_\nu | V | \phi_\mu \rangle}{\mathcal{E}_\mu - \mathcal{E}_\nu}. \quad (\text{A8})$$

Operating on Eq. (A8) with $\langle \Psi_{\mu'} |$, which has the same energy eigenvalue $\mathcal{E}_{\mu'} = \mathcal{E}_\mu$ we find condition (i) [Eq. (49)]:

$$\langle \Psi_{\mu'} | \phi_\mu \rangle = N\delta_{\mu'\mu} \quad (\mathcal{E}_{\mu'} = \mathcal{E}_\mu). \quad (\text{A9})$$

We apply $(\mathcal{E}_\mu - K - V)$ to both sides of Eq. (A8) again, and obtain

$$-V|\phi_\mu\rangle + \sum_{\substack{\nu \\ \mathcal{E}_\nu \neq \mathcal{E}_\mu}} |\Psi_\nu\rangle \langle \Psi_\nu | V | \phi_\mu \rangle = 0. \quad (\text{A10})$$

By the completeness relation $\sum_\nu |\Psi_\nu\rangle \langle \Psi_\nu| = 1$, Eq. (A10) is equivalent to

$$\sum_{\substack{\nu \\ \mathcal{E}_\nu \neq \mathcal{E}_\mu}} |\Psi_\nu\rangle \langle \Psi_\nu | V | \phi_\mu \rangle = 0. \quad (\text{A11})$$

The $\{\Psi_\nu\}$ are linearly independent, whence we have

$$\langle \Psi_\nu | V | \phi_\mu \rangle = 0 \quad (\mathcal{E}_\nu = \mathcal{E}_\mu). \quad (\text{A12})$$

On the other hand, by using condition (i) just proven, $N\Psi_\mu$ can always be expressed as

$$N|\Psi_\mu\rangle = \sum_{\substack{\mu' \\ \mathcal{E}_{\mu'} = \mathcal{E}_\mu}} |\Psi_{\mu'}\rangle \langle \Psi_{\mu'} | \phi_\mu \rangle = 0. \quad (\text{A13})$$

By using the completeness relation, we have

$$N|\Psi_\mu\rangle = |\phi_\mu\rangle - \sum_{\substack{\nu \\ \mathcal{E}_\nu \neq \mathcal{E}_\mu}} |\Psi_\nu\rangle \langle \Psi_\nu | \phi_\mu \rangle. \quad (\text{A14})$$

Comparing with Eq. (A8), we see

$$(\mathcal{E}_\nu - \mathcal{E}_\mu) \langle \Psi_\nu | \phi_\mu \rangle = \langle \Psi_\nu | V | \phi_\mu \rangle \quad (\mathcal{E}_\nu \neq \mathcal{E}_\mu). \quad (\text{A15})$$

Combining this result with the $\mathcal{E}_\nu = \mathcal{E}_\mu$ case of Eq. (A12), we have, in general,

$$(\mathcal{E}_\nu - \mathcal{E}_\mu) \langle \Psi_\nu | \phi_\mu \rangle = \langle \Psi_\nu | V | \phi_\mu \rangle. \quad (\text{A16})$$

This result is equivalent to

$$\langle \Psi_\nu | (K - K^\dagger) | \phi_\mu \rangle = 0, \quad (\text{A17})$$

whence condition (ii) [Eq. (50)] is proven.

(2) *Sufficient condition.* From condition (ii) [Eq. (A17)] we have

$$\begin{aligned} \langle \Psi_\nu | \phi_\mu \rangle (\mathcal{E}_\nu - \mathcal{E}_\mu) &= \langle \Psi_\nu | [K^\dagger + V] - K | \phi_\mu \rangle \\ &= \langle \Psi_\nu | V | \phi_\mu \rangle. \end{aligned} \quad (\text{A18})$$

For $\mathcal{E}_\nu \neq \mathcal{E}_\mu$ this equation leads to

$$\frac{\langle \Psi_\nu | V | \phi_\mu \rangle}{\mathcal{E}_\nu - \mathcal{E}_\mu} = \langle \Psi_\nu | \phi_\mu \rangle \quad (\mathcal{E}_\nu \neq \mathcal{E}_\mu). \quad (\text{A19})$$

Condition (i) yields

$$\begin{aligned} N | \Psi_\mu \rangle &= \sum_{\mathcal{E}_\nu = \mathcal{E}_\mu} | \Psi_\nu \rangle \langle \Psi_\nu | \phi_\mu \rangle \\ &= | \phi_\mu \rangle - \sum_{\mathcal{E}_\nu \neq \mathcal{E}_\mu} | \Psi_\nu \rangle \langle \Psi_\nu | \phi_\mu \rangle. \end{aligned} \quad (\text{A20})$$

Combining the two preceding equations, we have

$$N | \Psi_\mu \rangle = | \phi_\mu \rangle + \sum_{\mathcal{E}_\nu \neq \mathcal{E}_\mu} \frac{| \Psi_\nu \rangle \langle \Psi_\nu | V | \phi_\mu \rangle}{\mathcal{E}_\mu - \mathcal{E}_\nu} \quad (\text{A21})$$

or

$$N \Psi_\mu = \phi_\mu + P \frac{1}{\mathcal{E}_\mu - K - V} V \phi_\mu, \quad (\text{A22})$$

which completes the proof.

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