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Guo, D. S. and Drake, Gordon W. F.. (1992). Stationary solutions for an electron in an intense laser field. I. Single-mode case. *Journal of Physics A: Mathematical and General*, 25 (11), 3383-3397.
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Stationary solutions for an electron in an intense laser field:

I. Single-mode case

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Received 24 December 1991

Abstract. The Schrödinger equation for an electron and a single-mode photon field with interactions is solved by a direct method. A unique feature of these solutions is the inclusion of retardation effects in the photon field. Some interesting physical questions arising from the solutions are discussed. The Keldysh–Faisal–Reiss formula for the transition rate of multiphoton ionization modified by the inclusion of retardation effects is simplified by averaging the degenerate initial states. The result shows that the retardation effects can be calculated in terms of the radial part of the momentum wavefunction of the initial state. The physical significance of the inclusion is analysed in the near-threshold case of multiphoton ionization. Our result shows that in the near-threshold case, retardation effects depend exponentially on the orbital angular momentum of the initial state. The effect vanishes for s-states, but is significant for states with high orbital angular momentum.

1. Introduction

The interaction of atoms with very strong electromagnetic fields has recently become accessible to experimental study with increasingly powerful lasers. Observations of multiphoton ionization under field intensities in excess of 1 TW cm^{-2} lead to phenomena that cannot be explained in terms of lowest-order perturbation theory (Bucksbaum *et al* 1987, Gallagher 1988). Theoretical approaches to multiphoton ionization which use non-perturbative techniques have therefore been developed (Keldysh 1964, Faisal 1973, Reiss 1980, Chu and Cooper 1985, Becker *et al* 1986, Shakeshaft and Potvlige 1987, Javanainen and Eberly 1989, Parker and Stroud 1989), many of which make use of the classical Volkov state, i.e., the solution of the time-dependent Schrödinger equation for an unbound electron in an oscillating electromagnetic field (Volkov 1935, Faisal 1987). A widely used simple model originally initiated by Keldysh and combined with later improvements is now often called Keldysh–Faisal–Reiss (KFR) theory (Keldysh 1964, Faisal 1973, Reiss 1980). Due to its simplicity, KFR theory is frequently used by experimentalists in data analysis (Bashkansky *et al* 1987, Freeman and Bucksbaum 1991). Many theoretical studies have been devoted to questions of validity (Antunes, Neto and Davidovich 1984, Milonni 1988) and origins of KFR theory (Guo and Åberg 1988, Mu 1990), or to constructing improved theories (Guo *et al* 1989, Guo and Drake 1992). KFR theory still requires further discussion before an evaluation can be made of its contributions. The non-relativistic solutions for an electron in a classical and a quantized EM field have been applied to many situations

(Keldysh 1964, Faisal 1973, Reiss 1980, Bialynicki-Birula and Bialynicki-Birula 1976, Bialynicki-Birula 1980, Faisal 1987, Rosenberg 1982, Ehloltzky 1985).

A quantized field approach in cases of strong laser fields is necessary in two respects. First, a quantized field approach regards the complete system of interacting atoms and photons as an isolated system, for which the total energy from the initial state, via the intermediate states, to the final states is well-defined, in contrast to the semiclassical approach which regards the field as a time-dependent external field. Thus, in the former approach, the evolution of the energy distribution among the electrons and the photons due to interactions and energy transfers is well determined, while in the latter the energy for the interacting system is not conserved. Second, high intensity radiation fields are commonly thought of as classical fields, but this does not mean that there are no quantum effects in the classical field regime. Above-threshold ionization peaks spaced by one photon energy show quantum effects in strong radiation fields (Agostini *et al* 1979). It is of considerable mathematical and physical interest to study the strong field limit of quantum field theories to establish the connection with the semiclassical pictures and perhaps uncover new phenomena in the process. A recent success of this approach (Guo and Drake, 1992) is an explanation for the peak splittings in the angular distribution of photoelectrons in strong laser fields observed by Bucksbaum *et al* (1988).

In our search for a quantum electrodynamical approach to multiphoton ionization by high intensity light, we have solved the Dirac equation exactly for a relativistic electron interacting with a quantized and elliptically polarized single-mode electromagnetic field (Guo and Åberg 1988). Taking the non-relativistic and large-photon-number limits from the relativistic solution, we obtained a simple form of the solutions (Guo *et al* 1989) which gives a proper justification for the non-relativistic solution of the Schrödinger equation in the large-photon-number limit. The quantized-field version of the solutions enabled us to treat multiphoton ionization as a genuine scattering process in an isolated system that consists of photons and an atom (Guo *et al* 1989). A different type of scattering theory has been developed by Mu (1990), who applied the same set of solutions in the H^- detachment case and good agreement with experimental measurements was achieved by Mu *et al* (1990).

In a recent paper (Guo 1990) we developed a Lie algebra method by which we obtained solutions for a non-relativistic electron in a single-mode and a multimode quantized radiation field. The solutions obtained in the present work introduce the following three features. (1) The field has an arbitrary elliptical polarization. (2) The present solutions are for the large-photon-number limit, but in the quantized-field version, thus making it possible to describe absorption and emission processes with definite transferred-photon numbers. They also enable us to treat the electron and photons as an isolated system, so that the wavefunctions for the electron and photons are energy eigenfunctions of the Hamiltonian. (3) The present solutions are non-relativistic for the electron, but there is no long wavelength approximation for the photons; i.e. retardation is included in the photon vector potential, hence in the photon part of the wavefunction. This feature is particularly advantageous for treating strong radiation fields, in contrast to earlier non-relativistic semiclassical approaches that are mostly in the dipole approximation where the light-cone directions are deformed, limiting their range of applications. The Bucksbaum *et al* experiment (1988) strongly shows that there could be large momentum transfers in multiphoton ionization processes. Thus, keeping the momentum terms due to the photon field and the interaction in the general formulation may be necessary. As in the energies terms,

all momentum terms in the quantum electrodynamical approach are well-defined for the electrons, the fields, and the interaction. Thus, treating a non-relativistic electron by keeping retardation effects is possible, and intrinsically consistent. Actually it turns out to be an elegant feature of this theory.

The paper is organized as follows. In section 2, we solve the Schrödinger equation for a non-relativistic electron interacting with a single-mode quantized elliptically polarized EM plane wave by a straightforward method. Then we use a limiting process to obtain the solution in the large-photon-number limit. In section 3, we briefly discuss some interesting questions arising from these solutions, such as interaction energy, ponderomotive four-momentum, and the conversion between ponderomotive energy and the photons. In section 4, we give a complete treatment of the multi-photon ionization rate in KFR theory, modified by the inclusion of retardation effects. Even though the formula with retardation appeared formally in our earlier work, it has never been carried through explicitly to a calculable form. By averaging the degenerate initial states, we obtain a simplified rate formula, which shows that the effect is determined by the radial part of the momentum wavefunction of the initial state. Then, we analyse the significance of the effect in near-threshold multiphoton ionizations and provide a numerical example by treating the outermost shell of xenon gas. Our analysis shows that the numerical result is largely independent of the model of the atom.

2. Electron in a single-mode quantized radiation field

In this section we employ a straightforward method to obtain solutions of the Schrödinger equation for a non-relativistic electron interacting with a single-mode photon field. A number of transformations are employed which are now familiar in quantum optics. Then we take the large-photon-number limit to obtain a simple form of the solutions for strong radiation fields. In the present paper, we use units with $\hbar = c = 1$, and $e = -|e|$.

The Hamiltonian for a non-relativistic electron in a single-mode quantized radiation field can be obtained from the minimum-coupling principle as

$$H = \frac{(-i\nabla)^2}{2m_e} - \frac{e}{2m_e} [(-i\nabla) \cdot \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) + \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) \cdot (-i\nabla)] + \frac{e^2 \mathbf{A}^2(-\mathbf{k} \cdot \mathbf{r})}{2m_e} + \omega N_a \quad (1)$$

where

$$\mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) = g(\epsilon e^{i\mathbf{k} \cdot \mathbf{r}} a + \epsilon^* e^{-i\mathbf{k} \cdot \mathbf{r}} a^\dagger) \quad (2)$$

and $g = (2V_\gamma \omega)^{-1/2}$, V_γ being the normalization volume of the photon field. N_a is the photon number operator:

$$N_a = \frac{1}{2}(aa^\dagger + a^\dagger a). \quad (3)$$

The polarization vectors ϵ and ϵ^* are defined by

$$\begin{aligned} \epsilon &= [\epsilon_x \cos(\xi/2) + i\epsilon_y \sin(\xi/2)]e^{i\Theta/2} \\ \epsilon^* &= [\epsilon_x \cos(\xi/2) - i\epsilon_y \sin(\xi/2)]e^{-i\Theta/2} \end{aligned} \quad (4)$$

and satisfy

$$\begin{aligned}\epsilon \cdot \epsilon^* &= 1 \\ \epsilon \cdot \epsilon &= \cos \xi e^{i\Theta} \\ \epsilon^* \cdot \epsilon^* &= \cos \xi e^{-i\Theta}.\end{aligned}\tag{5}$$

The angle ξ determines the degree of polarization, such that $\xi = \pi/2$ corresponds to circular polarization and $\xi = 0$, to linear polarization. The phase angle Θ is introduced to characterize the initial phase value of the simple harmonic oscillator in our work (Guo 1990). With this phase, a full squeezed light transformation (Loudon and Knight 1987) can be fulfilled in the solving process. In multimode cases, the relative value of this phase for each mode will be important.

The Schrödinger equation to be solved is the eigenvalue equation

$$H\Psi(\mathbf{r}) = \mathcal{E}\Psi(\mathbf{r}).\tag{6}$$

To remove the coordinate dependence of the $A(-\mathbf{k} \cdot \mathbf{r})$ field, we apply a canonical transformation (Lee *et al* 1953, Girardeau 1960, Filipowicz 1985)

$$\Psi(\mathbf{r}) = e^{-i\mathbf{k} \cdot \mathbf{r} N_a} \phi(\mathbf{r}).\tag{7}$$

Equation (6) then becomes

$$\left\{ \frac{1}{2m_e} [(-i\nabla) - \mathbf{k} N_a]^2 - \frac{e}{2m_e} [(-i\nabla) \cdot \mathbf{A} + \mathbf{A} \cdot (-i\nabla)] + \frac{e^2 \mathbf{A}^2}{2m_e} + \omega N_a \right\} \phi(\mathbf{r}) = \mathcal{E} \phi(\mathbf{r})\tag{8}$$

where $\mathbf{k} \cdot \mathbf{A} = 0$ by transversality. Here, $\mathbf{A}$ is coordinate-independent and defined as

$$\mathbf{A} = e^{i\mathbf{k} \cdot \mathbf{r} N_a} \mathbf{A}(-\mathbf{k} \cdot \mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r} N_a} = g(\epsilon a + \epsilon^* a^\dagger).\tag{9}$$

The coordinate-independent equation is obtained by setting

$$\phi(\mathbf{r}) = e^{i\mathbf{p} \cdot \mathbf{r}} \phi\tag{10}$$

i.e.

$$\left\{ \frac{1}{2m_e} (\mathbf{p} - \mathbf{k} N_a)^2 - \frac{e}{2m_e} [\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p}] + \frac{e^2 \mathbf{A}^2}{2m_e} + \omega N_a \right\} \phi = \mathcal{E} \phi.\tag{11}$$

The $(\mathbf{k} N_a)^2$ term is an operator which gives an equation whose solutions are not known. In weak radiation fields with a background-photon number n , the term can be neglected because we have $\omega N_a \approx \omega n \ll m_e$ and $\mathbf{k} N_a \approx \mathbf{k} n \ll \mathbf{p}$. For strong radiation fields, however, the term cannot be neglected. The difficulty can be overcome in the following way, which allows one to include most of the contribution of the $\mathbf{k} N_a$ terms, while the remaining contributions are small relativistic corrections of order v/c for the electron.

We postulate that there is a real number κ , such that the operator $(\kappa \mathbf{k} - N_a \mathbf{k})$ vanishes in the non-relativistic limit. This will allow us to include retardation effects

to all orders for the photons, while neglecting relativistic corrections of order v/c for the electron. A full justification is given at the end of this section. Using this postulate, we now replace kN_a in (11) by κk , with κ to be determined later. We further define a vector P through the relation

$$p = P + \kappa k. \quad (12)$$

Then (11) is reduced to the simpler form

$$\left(\frac{P^2}{2m_e} - \frac{eP \cdot A}{m_e} + \frac{e^2 A^2}{2m_e} + \omega N_a \right) \phi = \mathcal{E} \phi \quad (13)$$

which is a solvable equation in quantum optics.

To remove the terms which are quadratic in the creation and annihilation operators in equation (13), we apply the full 'squeezed-light' transformation (Loudon and Knight 1987, Guo 1990)

$$\begin{aligned} c &= \cosh \chi a - \sinh \chi e^{-i\Theta} a^\dagger \\ c^\dagger &= -\sinh \chi e^{i\Theta} a + \cosh \chi a^\dagger \end{aligned} \quad (14)$$

with the inverse

$$\begin{aligned} a &= \cosh \chi c + \sinh \chi e^{-i\Theta} c^\dagger \\ a^\dagger &= \sinh \chi e^{i\Theta} c + \cosh \chi c^\dagger \end{aligned} \quad (15)$$

and follow the same steps as in the relativistic case (Guo and Åberg 1988) to obtain the exact non-relativistic wavefunction. By finding

$$\phi = D^\dagger |n\rangle_c \quad (16)$$

we finally have

$$\Psi(r) = V_e^{-1/2} \exp[i(-kN_a + P + \kappa k) \cdot r] D^\dagger |n\rangle_c \quad (17)$$

where V_e is the normalization volume for this wavefunction. We have

$$\begin{aligned} |n\rangle_c &= \frac{c^{\dagger n}}{\sqrt{n!}} |0\rangle_c \\ |0\rangle_c &= (\cosh \chi)^{-1/2} \sum_{s=0}^{\infty} (\tanh \chi)^s \left(\frac{(2s-1)!!}{(2s)!!} \right)^{1/2} e^{-is\Theta} |2s\rangle \\ \chi &= -\frac{1}{2} \tanh^{-1} \left(\frac{e^2 g^2 \cos \xi}{m_e \omega + e^2 g^2} \right). \end{aligned} \quad (18)$$

The shift operator D , which is introduced to eliminate the linear terms of the creation and annihilation operator in equation (13) (Filipowicz 1985, Guo and Åberg 1988), is given by

$$\begin{aligned} D &= \exp(-\delta c^\dagger + \delta^* c) \\ \delta &= egP \cdot \epsilon_c^* / C \\ C &= [(m_e \omega + e^2 g^2)^2 - e^4 g^4 \cos^2 \xi]^{1/2}. \end{aligned} \quad (19)$$

The operator D is a coherent light transformation (Loudon and Knight 1987), which shifts the creation and annihilation operator

$$\begin{aligned} DcD^\dagger &= c + \delta \\ Dc^\dagger D^\dagger &= c^\dagger + \delta^*. \end{aligned} \quad (20)$$

The rotated polarization vectors are defined by

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

The energy eigenvalues of the wavefunction (17) are found to be

$$\mathcal{E} = \frac{\mathbf{P}^2}{2m_e} + \frac{C(n + \frac{1}{2})}{m_e} - \frac{e^2 g^2 (\mathbf{P} \cdot \epsilon_c)(\mathbf{P} \cdot \epsilon_c^*)}{m_e C}. \quad (22)$$

If we define

$$E = \frac{\mathbf{P}^2}{2m_e} \quad (23)$$

we see that (23) is just the on-mass-shell condition for a non-relativistic electron with four-momentum $(E + m_e, \mathbf{P})$. Thus we have

$$\mathcal{E} + m_e = (E + m_e) + \kappa' \omega \quad (24)$$

where the constant κ' is defined by

$$\kappa' = \frac{C(n + \frac{1}{2})}{m_e \omega} - \frac{e^2 g^2 (\mathbf{P} \cdot \epsilon_c)(\mathbf{P} \cdot \epsilon_c^*)}{C m_e \omega}. \quad (25)$$

The electron is described non-relativistically, its velocity being much less than that of light, but retardation is included for the photons. Comparing (24) and (12) and using the fact that $(\mathcal{E} + m_e, \mathbf{p})$, $(E + m_e, \mathbf{P})$ and $(\omega, \mathbf{k})$ are Lorentz four-vectors, it follows that

$$\kappa = \kappa' = \frac{C(n + \frac{1}{2})}{m_e \omega} - \frac{e^2 g^2 (\mathbf{P} \cdot \epsilon_c)(\mathbf{P} \cdot \epsilon_c^*)}{C m_e \omega}. \quad (26)$$

We can define the important parameter z

$$z = \kappa - (n + \frac{1}{2}) \quad (27)$$

with the interpretation that $z\omega$ is the interaction energy.

If the radiation field is strong, the photon number becomes very large and the field takes on classical characteristics. As in the earlier work (Guo and Åberg 1988), we let

$$\begin{aligned} g\sqrt{n} &\rightarrow \Lambda \\ n &\rightarrow \infty \\ g &\rightarrow 0 \end{aligned} \quad (28)$$

where Λ is the amplitude of the classical field. The present formalism remains valid for weak fields if the photon normalization volume tends to infinity, because we have $g = (2V_\gamma\omega)^{-1/2}$ and the classical amplitude Λ of the field is finite, not infinitesimal. We will call the limit (28) the large-photon-number limit.

We need to evaluate the matrix element $\langle l|D^\dagger|n\rangle_c$ in the large-photon-number limit, $|l\rangle$ being a state of l free photons in the N_a representation. Applying the intermediate-states method, we insert $\sum_m |m\rangle\langle m|$, which is the identity operator, in the matrix element:

$$\langle l|D^\dagger|n\rangle_c = \sum_m \langle l|D^\dagger|m\rangle\langle m|n\rangle_c. \quad (29)$$

Following the same procedure as in the earlier work (Guo and Åberg 1988), we find the limiting form when $m \rightarrow \infty, l \rightarrow \infty, g \rightarrow 0$,

$$\langle l|D^\dagger|m\rangle \rightarrow \sum_{q=-\infty}^{\infty} J_{-q}(\zeta) e^{-iq(\phi_\xi + \frac{\theta}{2})} \delta_{q, l-m}, \quad (30)$$

where

$$\begin{aligned} \phi_\xi &= \tan^{-1}[(P_y/P_x) \tan(\xi/2)] \\ \zeta &= \frac{2|e|\Lambda}{m_e\omega} |\mathbf{P} \cdot \boldsymbol{\epsilon}| \end{aligned} \quad (31)$$

and

$$\begin{aligned} \langle m|n\rangle_c &\rightarrow \sum_{t=-\infty}^{\infty} J_t(\eta) e^{it\Theta} \delta_{2t, n-m} \\ \eta &= \frac{1}{2} z \cos \xi. \end{aligned} \quad (32)$$

Combining (30) and (32), we have

$$\langle l|D^\dagger|n\rangle_c \rightarrow \sum_j \mathcal{J}_j(\zeta, \eta, \phi_\xi)^* e^{-ij(\phi_\xi + \frac{\theta}{2})} \delta_{j, l-n}. \quad (33)$$

The generalized Bessel function $\mathcal{J}_j(\zeta, \eta, \phi_\xi)$ is defined (Guo and Åberg 1988) as

$$\mathcal{J}_j(\zeta, \eta, \phi_\xi) = \sum_{m=-\infty}^{\infty} J_{-j-2m}(\zeta) J_m(\eta) \exp(2im\phi_\xi). \quad (34)$$

Thus one has the limiting form

$$\phi = \sum_l |l\rangle\langle l|D^\dagger|n\rangle_c \rightarrow \sum_{j=-n}^{\infty} \mathcal{J}_j(\zeta, \eta, \phi_\xi)^* e^{-ij(\phi_\xi + \frac{\theta}{2})} |n+j\rangle. \quad (35)$$

For the wavefunction describing the non-relativistic electron in the large-photon-number limit we then have

$$\Psi_{P_n}(\mathbf{r}) = V_e^{-1/2} \sum_{j=-n}^{\infty} e^{i[\mathbf{P} + (z-j)\mathbf{k}] \cdot \mathbf{r}} |n+j\rangle \mathcal{J}_j(\zeta, \eta, \phi_\xi)^* e^{-ij(\phi_\xi + \frac{\theta}{2})} \quad (36)$$

with the energy eigenvalue

$$\mathcal{E} = P^2/2m_e + (n + \frac{1}{2})\omega + z\omega. \quad (37)$$

Combining (26), (27), and the limiting process (28), we see that z in (37) is given by

$$z = \frac{e^2 \Lambda^2}{m_e \omega}. \quad (38)$$

We now justify the ansatz leading to the solution (36). The principal justification is that (36) agrees exactly with the non-relativistic large-photon-number limit of the corresponding relativistic solution (Guo *et al* 1989). Within the present context, it can also be justified as follows. We have replaced $p - N_a k$ by P . From (12) and (27), we have

$$p = P + (n + \frac{1}{2})k + zk. \quad (39)$$

By comparing (17) and (36) we know that N_a , when it acts on $|n + j\rangle$, produces $(n + j + \frac{1}{2})k$ in each term of the sum. Thus, with (39), we have

$$p - N_a k \approx P + (z - j)k. \quad (40)$$

Now $z = (eg)^2 n / (m_e \omega)$ is finite in the limit $n \rightarrow \infty$ and $g \rightarrow 0$ and hence satisfies $z \ll n$. On the other hand, we can take $j \ll n$, even though j in principle runs from $-n$ to $+\infty$. The physical reason is that j represents the number of transferred photons, while n is the number of background photons; the mathematical reason is that j is the order index of the generalized Bessel functions, whence those terms which make non-negligible contributions must satisfy $j \ll n$. For example, in Xe $5p_{3/2}$ multiphoton ionization, the terms with $11 \leq j \leq 19$ contribute importantly, and the others are negligible. Hence we have the inequality

$$|(z - j)\omega| \ll m_e. \quad (41)$$

Comparing (11), (13), (22), and (40), we see that the quantity neglected by using the present ansatz is

$$\begin{aligned} & [P + (z - j)k]^2 / (2m_e) - P^2 / (2m_e) \\ & \approx (P_z / m_e)(z - j)\omega + [(z - j)\omega / (2m_e)](z - j)\omega \end{aligned} \quad (42)$$

which is of higher order relativistically than the energy $(z - j)\omega$ which we kept in the derivation.

3. Physical significance of the solutions

In this section, we briefly discuss some interesting features of these solutions.

3.1. Interaction energy

From (22), the energy levels of the system of an electron and many photons in the single-mode case including the interactions can be rewritten as

$$\mathcal{E} = \frac{P^2}{2m_e} + (n + \frac{1}{2})\omega + \left[\frac{C(n + \frac{1}{2})}{m_e} - (n + \frac{1}{2})\omega \right] - \frac{e^2 g^2 (P \cdot \epsilon_c)(P \cdot \epsilon_c^*)}{m_e C} \quad (43)$$

which contains four terms. The first term is the kinetic energy of the electron. The second term is the free energy and the zero-point energy of the photons. The third term, which originates from the A^2 term, has an asymptotic form $(e^2 \Lambda^2 / m_e \omega) \omega$ in the large-photon-number limit. This term is the ponderomotive potential energy, which is a repulsive energy and depends on the background-photon number. It is thus much larger than the attractive energy in strong-field cases. Thus we can say photon-pair emission and absorption produce the repulsive energy. The fourth term, which originates from the $P \cdot A$ term of the Hamiltonian and does not depend on the background-photon number n , is an attractive energy. Thus we can say that the electron has a natural attractive tendency with the photon cloud emitted by itself if there are no background photons. We can also say that single-photon emission and absorption produce the attractive energy.

3.2. Ponderomotive four-momentum

As shown in our earlier work, we once again see the existence of the ponderomotive momentum, which together with its counterpart the well-known ponderomotive energy forms the ponderomotive four-momentum. The ponderomotive four-momentum, which is from the interaction of two kinds of particles, forms only a part of the four-momentum of the system. An interacting system of an electron and a single-mode photon field has a definite four-momentum which can be called the total four-momentum (Guo and Åberg 1991). A peculiar property of the total four-momentum is that it can be decomposed into a sum of an on-mass-shell four-vector and a null four-vector (Berestetskii *et al* 1982, Filipowicz 1985, Guo and Åberg 1988). Here, an on-mass-shell four-vector means a four vector with a norm of m_e^2 , while a null four-vector means a four-vector with a vanishing norm, or a light-like vector. The relations can be written algebraically as

$$\begin{aligned} p &= P + \kappa k \\ P^2 &= m_e^2 \\ k^2 &= 0 \end{aligned} \quad (44)$$

where κ is a real number. Since the four-vector κk is along the light cone direction, it possesses the properties of a photon field. By subtracting the four-momentum of the free photon fields from the κk , what is left is the ponderomotive four-momentum (Guo and Åberg 1988, Guo 1990)

$$\kappa k - (n + \frac{1}{2})k = zk, \quad (45)$$

which is a consequence of equations (37) and (39). In semiclassical treatments, the ponderomotive energy is considered as a part of the energy of an electron, or as a property of the electron, while the ponderomotive momentum has never been addressed. In our treatment, one can see that the ponderomotive four-momentum is a property of both the electron and the field, but its null vector character gives it a light-like quality.

3.3. Conversions between ponderomotive potential energy and photons

By mapping these solutions onto a plane wave on the same energy shell, (Guo *et al* 1989) we have found that ponderomotive potential energy converts into photons when the photoelectron leaves the radiation field. A treatment by using a semiclassical theory supports our conclusion (Unnikrishnan 1990). The conversion between ponderomotive potential energy and photons in multimode cases is an interesting topic for further investigation. In a very recent paper (Guo and Drake 1992b), we extended the scattering theory developed for the case of single-mode multiphoton ionization by Guo *et al* (1989) to the standing wave case. The theory correctly predicts the angular distribution peak splitting observed by Bucksbaum *et al* (1988). The conversion between the ponderomotive potential energy and photons is a natural feature of the theory which affects the splittings.

4. Keldysh-Faisal-Reiss theory with the inclusion of retardation effects

As a simple application, we can use the single-mode solutions as the final state for a photoelectron to calculate the transition rate in multiphoton ionization processes according to KFR theory. This is just one possible use of the quantum field solutions for the final electron states, and is not an essential part of the quantum treatment itself.

The first non-perturbative treatment for multiphoton ionization in the single-mode case was made by Keldysh (1964), by assuming the final state of the electron to be a non-relativistic, time-dependent, single-mode, and semiclassical Volkov state. This means that the photoelectron is moving in a classical time-dependent electromagnetic wave, while the initial state is an atomic bound state and the interaction is in the dipole approximation. Later this treatment was improved and made more rigorous by Faisal (1973) and Reiss (1980). Reiss employed the radiation gauge for the interaction with the long wavelength approximation for the A field, and with the inclusion of the A^2 term. Bashkansky *et al* (1987) generalized Reiss's formulae for circularly and linearly polarized cases to the elliptically polarized case. Guo and Åberg (1988) reproduced the formulae of Reiss and Bashkansky *et al* by a non-perturbative quantum electrodynamical approach, but without the long wavelength approximation. A recent discussion of the gauge dependence of KFR theory has been given by Milonni and Ackerhalt (1989). They show that one must be careful in the interpretation of the A^2 term within the context of KFR theory, but gauge dependence does not have practical consequences in the low-frequency regime of current experiments. A full quantum treatment which includes the atomic binding potential along with the terms in (1) would of course be gauge invariant.

Despite this and other shortcomings discussed by Milonni and Ackerhalt (1989), it is of interest to show that the KFR results can be obtained from our stationary solutions, including the modifications due to retardation as follows. The transition matrix element in the elliptically polarized case without the long wavelength approximation is

$$T_{fi} = \langle \Psi_{P_n}(\mathbf{r}) | V | \Phi_i(\mathbf{r}), l \rangle$$

$$= \omega(z - j) \mathcal{J}_j(\zeta, \eta, \phi_\xi) e^{ij(\phi_\epsilon + \frac{\pi}{2})} \Phi(\mathbf{P} + z\mathbf{k} - j\mathbf{k}) \quad (46)$$

where V contains the interaction terms in (1) and $\Phi_i(\mathbf{r})$ is the initial atomic bound state wavefunction with its representation $\Phi(\mathbf{P} + z\mathbf{k} - j\mathbf{k})$ in momentum space, and $j = l - n$ is the absorbed photon number, i.e. the photon number difference between the initial free-photon number l and the final background photon number n . The definitions of the generalized Bessel function and its argument are in equations (30) to (32).

By imposing energy conservation between the initial and the final states

$$\frac{P^2}{2m_e} = j\omega - z\omega - E_b \quad (47)$$

the transition rate for multiphoton ionization in the single-mode radiation field case is derived as (Reiss 1980, Bashkansky *et al* 1987, Guo and Åberg 1988)

$$\begin{aligned} \frac{dw}{d\Omega} &= \int_0^\infty (2\pi)^{-3} P^2 d|P| 2\pi |T_{fi}|_{av}^2 \delta\left(\frac{P^2}{2m_e} - j\omega + z\omega + E_b\right) \\ &= \frac{(2m_e^3 \omega^5)^{\frac{1}{2}}}{(2\pi)^2} (j-z)^2 (j-z-E_b/\omega)^{\frac{1}{2}} |\Phi(\mathbf{P} + z\mathbf{k} - j\mathbf{k})|_{av}^2 \\ &\quad \times |\mathcal{J}_j(\zeta, \eta, \phi_\xi)|^2 \end{aligned} \quad (48)$$

by noticing

$$\begin{aligned} \delta\left(\frac{P^2}{2m_e} - j\omega + z\omega + E_b\right) &= (m_e/2\omega)^{\frac{1}{2}} (j-z-E_b/\omega)^{-\frac{1}{2}} \delta[|P| \\ &\quad - (2m_e\omega)^{\frac{1}{2}} (j-z-E_b/\omega)^{\frac{1}{2}}]. \end{aligned} \quad (49)$$

The subscript *av* on the squares of the transition matrix element and the momentum wavefunction means the average value among the initial states with the same principal quantum number n and orbital angular quantum number l , but different magnetic quantum numbers. Equation (48) can be further simplified. The initial wavefunction $\Phi_i(\mathbf{r})$ can be written as

$$\Phi_{nlm}(\mathbf{r}) = \frac{1}{r} P_{nl}(r) Y_{lm}(\theta_1, \phi_1) \quad (50)$$

where r, θ_1 and ϕ_1 are spherical coordinates for $\mathbf{r}$. With the partial wave expansion

$$e^{-i\mathbf{P}' \cdot \mathbf{r}} = 4\pi \sum_{l'=0}^{\infty} (-i)^{l'} j_{l'}(|\mathbf{P}'|r) \sum_{m'=-l'}^{l'} Y_{l'm'}^*(\theta_1, \phi_1) Y_{l'm'}(\theta_2, \phi_2) \quad (51)$$

where θ_2 and ϕ_2 are the spherical angles for $\mathbf{P}'$, and $\mathbf{P}' \equiv \mathbf{P} - (j-z)\mathbf{k}$, the Fourier transform of the initial wavefunction can be written explicitly as

$$\Phi(\mathbf{P}') = 4\pi (-i)^l Y_{lm}(\theta_2, \phi_2) \Phi_{nl}(|\mathbf{P}'|) \quad (52)$$

where

$$\Phi_{nl}(|\mathbf{P}'|) = \int_0^\infty r dr P_{nl}(r) j_l(|\mathbf{P}'|r) \quad (53)$$

can be regarded as the Fourier transform of the radial wavefunction, and $j_l(|\mathbf{P}'|r)$ is a spherical Bessel function. By using the identity

$$\sum_{m=-l}^l Y_{lm}(\theta_2, \phi_2) Y_{lm}^*(\theta_2, \phi_2) = \frac{2l+1}{4\pi} \quad (54)$$

one can eliminate the dependence on the angles θ_2 and ϕ_2 . Thus we find

$$|\Phi(\mathbf{P}')|_{\text{av}}^2 = 4\pi |\Phi_{nl}(|\mathbf{P}'|)|^2. \quad (55)$$

Retardation effects have not been treated before in multiphoton processes. Equation (48) together with (55) provide a simple way to include them. The retardation momentum could be important in the following cases. Theoretically, it is important in the balance of the four-momentum in a scattering process. The scattering theory of multiphoton ionization developed by Guo *et al* (1989) predicts that when the photoelectron leaves the radiation field ponderomotive potential energy will turn into photons. This prediction is based on the four-momentum balance. In near threshold ionization cases, the retardation momentum could be comparable to the kinetic momentum of the photoelectron, which can be proven as follows. The condition

$$|(j-z)\mathbf{k}| \approx |\mathbf{P}| = \sqrt{2m_e[(j-z)\omega - E_b]} \quad (56)$$

leads to

$$[(j-z)\omega]^2 - 2m_e(j-z)\omega + 2m_e E_b \approx 0 \quad (57)$$

which has two roots

$$\begin{aligned} (j-z)\omega &\approx 2m_e - E_b \\ (j-z)\omega &\approx E_b. \end{aligned} \quad (58)$$

The first root corresponds to an extremely deeply bound case which is out of the energy regime we are discussing. The approximate equality of the second root combining with the energy conservation (47) gives

$$\frac{P^2}{2m_e} \ll E_b \quad (59)$$

which is the near-threshold condition.

To show the significance of retardation effects in near-threshold multiphoton ionization processes, let us consider a hydrogenic model atom with the momentum wavefunctions

$$\Phi_{nl}(|\mathbf{P}|) = i^l \left[\frac{(n-l-1)!}{(n+l)!} \right]^{1/2} n^2 2^{2(l+1)} l! \frac{x^{l/2}}{(x+1)^{l+2}} C_{n-l-1}^{l+1} \left(\frac{x-1}{x+1} \right) \quad (60)$$

where

$$x \equiv P^2 a_0^2 n^2 / Z^2 = \frac{P^2}{2m_e} / E_b \quad (61)$$

and a_0 stands for the Bohr radius, Z the nuclear charge, and the $C_N^\nu(x)$ are the Gegenbauer polynomials. The expression (60) can be simplified under the condition (59) to

$$\Phi_{nl}(|P|) = i^l \left[\frac{(n-l-1)!}{(n+l)!} \right]^{1/2} n^2 2^{2(l+1)} l! (-1)^{n-l-1} \binom{n+l}{2l+1} \times (2m_e E_b)^{-l/2} |P|^l \{1 + O[(l+2)x]\}. \quad (62)$$

Thus the ratio of differential transition rates expressed by (48) obeys the following simple rule

$$\frac{dw(P')}{dw(P)} \simeq \left(\frac{|P'|}{|P|} \right)^{2l}. \quad (63)$$

This rule shows that there is no significant retardation effect for an s state, but the effect grows rapidly with l . In (63) the lack of dependence on the principal quantum number n follows from the fact that the radial wavefunction is proportional to $|P|^l$ for small $|P|$, independent of n .

To illustrate the effect on a p state and test the model-independence of the rule, an artificial atom with parameters close to those for the photoionization of the outermost $5p_{3/2}$ shell of xenon was chosen as a numerical example. The threshold energy of the atom is assumed to be 0.4392 au, while the light is circularly polarized with wavelength 1064 nm and intensity $8 \times 10^{12} \text{ W cm}^{-2}$. The radial wavefunction for the $5p_{3/2}$ electron is obtained by taking the large component of the same orbital generated by fitting all energy levels of a relativistic electron to agree with that of xenon in a Tietz (1954) model potential. The calculated v^2/c^2 of the photoelectron is 8.22×10^{-8} , which meets the near-threshold condition. When the scattering angle is $5\pi/12$, we have $dw(P')/dw(P) = 0.9647$ with $P'^2/P^2 = 0.9644$; when the scattering angle is $2\pi/3$, $dw(P')/dw(P) = 1.0876$ with $P'^2/P^2 = 1.0884$. The results agree to within 0.1% with the rule derived from hydrogenic wavefunctions. The artificial example shown here is not too unrealistic, because the threshold energy can be varied by another low frequency weaker laser beam and the laser frequency may also be changed by a tunable laser to meet the near-threshold ionization condition. The principle point is to illustrate the model-independence of the scaling law expressed by (63).

It is also instructive to show the necessity of the non-perturbative treatment by comparison of KFR theory with lowest order perturbation theory. In the circularly polarized case, where $\xi = \pi/2$, the generalized Bessel function reduces to an ordinary Bessel function $J_{-j}(\zeta)$. The sign difference of j in Reiss's original work is due to a difference in sign convention of the electron charge. Since $J_j(\zeta) = (-1)^j J_{-j}(\zeta)$, the final result is the same. The Bessel function has its power expansion

$$J_j(\zeta) = (\zeta/2)^j \sum_{k=0}^{\infty} \frac{(\zeta/2)^{2k}}{(k+j)!k!} (-1)^k \quad (64)$$

whose leading term $(\zeta/2)^j/j!$ gives the lowest order contribution if one applies perturbation theory to treat the final electron in an external radiation field. Here $\zeta = (2|e|\Lambda/m_e\omega)|P \cdot e|$ as defined in (31). The rate ratio Q of the lowest order

perturbation result to the non-perturbative result for the case of an above-threshold ionization peak is

$$Q = \left| j! \sum_{k=0}^{\infty} \frac{(\zeta/2)^{2k}}{(k+j)!k!} (-1)^k \right|^{-2}. \quad (65)$$

The following are numerical examples to show the importance of the higher order terms in equations (64) and (65). Consider 1064 nm light, with intensity $4 \times 10^{12} \text{ W cm}^{-2}$ to ionize an xenon atom with the outermost shell $5p_{3/2}$. For the $j = 15$ peak, with a scattering angle of $\theta = \pi/2$, ζ has value 2.52 and Q is 1.22. When the intensity increases to $2 \times 10^{13} \text{ W cm}^{-2}$, ζ increases to 4.60 and Q becomes 1.94. In this intensity region, it becomes necessary to use the full Bessel function.

5. Discussion

This paper establishes a clean connection between a quantum field picture of multiphoton ionization and the semi-classical picture represented by KFR theory. The derivation is simpler and more direct than the Lie algebra approach used previously (Guo 1990) and the result coincides with the non-relativistic limit of the solution to the Dirac equation obtained by Guo *et al* (1989). The latter provides the justification for the operator replacement $N_a k \rightarrow \kappa k$ leading to (13). The final results contain a number of generalizations and clarifications of the earlier work, as outlined in the Introduction.

A motivation for studying the semi-classical limit of quantum field theories is that it provides a rigorous foundation for the form of the final results that might otherwise be obtained by physical arguments. Since the interacting system of atoms and photons has a well-defined total energy and momentum at every step, the quantum field results provide a precise prescription for formulating the relevant energy-momentum conservation laws as the electron ionizes, acquires ponderomotive energy-momentum, and then ultimately leaves the radiation field. Time-dependent semi-classical theories are not always so well defined. The prescription for the proper way to include retardation effects when ponderomotive energy is important is one example worked out in this paper. In a future paper, it will be shown that the theory provides a natural explanation for the peak-splittings in the angular distributions of photoelectrons recently observed by Bucksbaum *et al* (1988). The theory is also capable of describing the partial decay of ponderomotive energy into photons as the electron leaves the radiation field. (Freeman *et al* 1987).

A limitation of the present single-mode theory is that the scattering formalism on which it is based assumes adiabatic switching for the photon field. It therefore does not provide a good description of interactions with very short pulses. However, some of these time-dependent features can be incorporated into a multimode extension of the theory and will be discussed in future work.

Acknowledgments

D S Guo would like to thank Bernd Crasemann for his advice, support, and Teijo Åberg for stimulative discussions. This work has been supported in part by Natural

Science and Engineering Research Council of Canada and National Science Foundation grant PHY-8908124. G W F Drake thanks the Killam Foundation for support.

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