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Quantum electrodynamical approach to multiphoton ionisation in the high-intensity H field

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Abstract. We solve the Dirac equation for an electron interacting with a quantised and elliptically polarised electromagnetic field. We use the solution to obtain a relativistic S-matrix amplitude for multiphoton ionisation in the high-intensity limit. Its non-relativistic limit is also derived and is used to construct a multiphoton transition-rate formula which is compared with previous results.

1. Introduction

The development of high-power lasers has made it possible to achieve light intensities which are of the order of one atomic unit (Rhodes 1987). At these field strengths relativistic effects become important (Mittleman 1982, Krstic and Mittleman 1982). It is thus of interest to study multiphoton ionisation from the point of view of quantum electrodynamics (QED).

In recent work Filipowicz (1985) obtained the solution of the Dirac equation for an electron interacting with a quantised electromagnetic plane wave. He also discussed the non-quantum limit of this solution. The wave was assumed to be circularly polarised. The wavefunction was given in a form which suggests that a generalisation to elliptically polarised light is feasible. This generalisation is carried out in the present work and is used to obtain a relativistic S-matrix amplitude for multiphoton ionisation in the high-intensity limit. The corresponding classical limits are derived in both the relativistic and non-relativistic approximations. Our procedure leads to a unique identification of an index in the classical expressions that gives the number of absorbed photons.

A classical non-relativistic transition-rate formula for elliptically polarised light has recently been reported by Bashkansky *et al* (1987) who have successfully used it for an analysis of angular distributions in above-threshold ionisation (ATI). This form of the cross section originates from the work of Keldysh (1965), Faisal (1973) and Reiss (1980). In essence it is based on the approximation of using, for the final state, the non-relativistic Volkov solution (Reiss 1980) representing a free electron in the electromagnetic field. The effect of the ionic field is thus ignored. It is shown that our non-relativistic classical S-matrix amplitude leads to the same transition-rate formula as theirs (Bashkansky *et al* 1987). Its quantum electrodynamical origin is thus established. Consequently, systematic improvements should be feasible since the availability

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of a complete set of quantised solutions of the Dirac equation for an arbitrary electromagnetic field makes it possible to consider perturbation corrections involving the ionic field.

The solutions obtained in this work are also useful for the analysis of a number of other physical processes involving electrons and positrons in external fields, such as the bremsstrahlung processes (Bergou 1980, Bergou and Varró 1980).

2. The Volkov solution in a quantised and elliptically polarised field

In the following we use 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 4-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 4-potential of the electromagnetic field is given by

$$A_{\mu}(kx)=g[\varepsilon_{\mu}a\exp(-\mathrm{i}kx)+\varepsilon_{\mu}^{*}a^{+}\exp(\mathrm{i}kx)] \tag{1}$$

where

$$\varepsilon^{*}\varepsilon=-1 \qquad \varepsilon\varepsilon=\varepsilon^{*}\varepsilon^{*}=-\cos\xi. \tag{2}$$

These relations follow from the representation $\varepsilon=(0,\boldsymbol{\varepsilon})$, where $\boldsymbol{\varepsilon}=\boldsymbol{\varepsilon}_x\cos(\xi/2)+\mathrm{i}\boldsymbol{\varepsilon}_y\sin(\xi/2)$. It is understood that the wave propagates in the direction $\boldsymbol{\varepsilon}_z$ of $k=(\omega,\boldsymbol{k})$.

Consequently the potential (1) corresponds to a situation in which the polarisation is obtained by means of half- and quarter-wave retardation plates inserted into the laser beam. The angle $\xi/2$ is the angle between the plane of oscillation after the half-wave plate and the optical axis of the quarter-wave plate. Thus $\xi=\pi/2$ corresponds to circular polarisation and $\xi=0$ to linear polarisation.

The Dirac equation to be solved is

$$[\mathrm{i}\partial\!\!\!/-e\not{A}(kx)-m]\psi(x)=0 \tag{3}$$

where $e<0$ for an electron and where relativistic units, with $\hbar=1$ and $c=1$, are used. Following Filipowicz (1985) we introduce the transformation

$$\psi(x)=\exp(\mathrm{i}kxN_a)\phi(x) \tag{4}$$

where $N_a=\frac{1}{2}(a^{+}a+aa^{+})$ is the occupation number operator. It transforms equation (3) into

$$(\mathrm{i}\partial\!\!\!/-k\!\!\!/N_a-e\not{A}-m)\phi(x)=0 \tag{5}$$

where $\not{A}=g(\not{\varepsilon}a+\not{\varepsilon}^{*}a^{+})$ is now coordinate independent. The solution of equation (5) has the form

$$\phi(x)=\exp(-\mathrm{i}px)\phi \tag{6}$$

where ϕ is a coordinate-independent bispinor satisfying the equation

$$(p\!\!\!/-k\!\!\!/N_a-e\not{A}-m)\phi=0. \tag{7}$$

We introduce the projection operators

$$\mathcal{P}=(p\!\!\!/+m)k\!\!\!//2kp \qquad \mathcal{Q}=k\!\!\!/(p\!\!\!/-m)/2kp \tag{8}$$

and express ϕ in terms of $\mathcal{P}\phi$. It follows from equation (7) and definitions (8) that

$$\phi=[1+e(k\!\!\!/A/2kp)]\mathcal{P}\phi. \tag{9}$$

The advantage of using this relationship lies in the fact that it is easier to solve for $\mathscr{P}\phi$ than ϕ. Multiplying equation (7) from the left by $(\not{p}+m)/2kp$, using equation (9) and properties of the γ matrices yields

$$(2kpN_a + 2eAp - e^2A^2 + e^2g^2\mathscr{P}\mathscr{S} - p^2 + m^2)\mathscr{P}\phi = 0 \tag{10}$$

where $A = g(\varepsilon^* a^+ + \varepsilon a)$ and $\mathscr{S} = \frac{1}{2}[\not{\varepsilon}^*, \not{\varepsilon}]$ is a polarisation-dependent spin operator. Since $\mathscr{P}\mathscr{S}\mathscr{P} = \mathscr{P}\mathscr{S}$ and $\mathscr{S}^2 = s^2$ with $s = \pm\sin\xi$, we have the eigenvalue equation

$$\mathscr{P}\mathscr{S}\mathscr{P}(\mathscr{S}+s)v = s\mathscr{P}(\mathscr{S}+s)v \tag{11}$$

where v is an arbitrary bispinor. For circular polarisation $s = \pm 1$ and for linear polarisation $s = 0$. Filipowicz (1985) only considered the $s = \pm 1$ case and used eigenvectors of $\mathscr{S}$ rather than $\mathscr{P}\mathscr{S}$ to classify the solutions of equation (3) which lead to some unnecessary conditions regarding the choice of v.

We shall use the ansatz

$$\mathscr{P}\phi = \mathscr{P}(\mathscr{S}+s)v|\rho\rangle \tag{12}$$

where $|\rho\rangle$ is a state which depends solely on the field operators and has to be determined. It follows from equations (10) and (11) that

$$(2kpN_a + 2eAp - e^2A^2 + e^2g^2 s - p^2 + m^2)|\rho\rangle = 0. \tag{13}$$

Since $\varepsilon\varepsilon = \varepsilon^*\varepsilon^* = -\cos\xi$, the A^2 term does not reduce to a bilinear form, proportional to N_a. Hence equation (13) cannot be diagonalised directly using a shift operator as in the circularly polarised case (Filipowicz 1985).

We define two operators c and c^+ as the linear combinations

$$\begin{aligned} c &= a\cosh\chi - a^+\sinh\chi \\ c^+ &= -a\sinh\chi + a^+\cosh\chi \end{aligned} \tag{14}$$

where χ will be determined such that it eliminates the quadratic term $a^2 + a^{+2}$ in equation (13). The operators (14) which obey the commutation relation $[c, c^+] = 1$ have an occupation number representation of eigenkets, $|n_c\rangle$, which form a complete orthonormal set. Thus it can be proved that

$$N_c|n_c\rangle = (n_c + \tfrac{1}{2})|n_c\rangle \tag{15}$$

where $N_c = \frac{1}{2}(c^+c + cc^+)$ is a Hermitian operator and where $n_c = 0, 1, 2, 3, \ldots$. The states $|n_c\rangle$ $(n_c \geqslant 1)$ are generated from the 'vacuum' state with $(-1)!! = 1$

$$|0_c\rangle = (\cosh\chi)^{-1/2} \sum_{n_a=0}^{\infty} (\tanh\chi)^{n_a} \left(\frac{(2n_a-1)!!}{(2n_a)!!}\right)^{1/2} |2n_a\rangle \tag{16}$$

for which $c|0_c\rangle = 0$ and $\langle 0_c|0_c\rangle = 1$, by

$$|n_c\rangle = \frac{(c^+)^{n_c}}{(n_c!)^{1/2}}|0_c\rangle. \tag{17}$$

It also follows from the proof which is given in appendix 1 that $c|n_c\rangle = \sqrt{n_c}|n_c - 1\rangle$ and $c^+|n_c - 1\rangle = \sqrt{n_c}|n_c\rangle$, in accordance with equations (15) and (17). Consequently, the N_c and N_a representations are completely analogous except that the 'vacuum' states are different. Furthermore, it shows that they are connected by a unitary transformation, V, such that

$$\begin{pmatrix} c^+ \\ c \end{pmatrix} = V\begin{pmatrix} a^+ \\ a \end{pmatrix} V^+ = \mathscr{L}\begin{pmatrix} a^+ \\ a \end{pmatrix}$$

where $\mathscr{L}$ is the transformation (14).

By using the inverse

$$a = c\cosh\chi + c^+\sinh\chi$$
$$a^+ = c\sinh\chi + c^+\cosh\chi \tag{18}$$

of transformation (14) and by putting

$$\tanh 2\chi = -e^2g^2\cos\xi/(kp + e^2g^2) \tag{19}$$

we have

$$[C(c^+c + cc^+) + 2egp(\varepsilon_c c + \varepsilon_c^* c^+) + e^2g^2s - p^2 + m^2]|\rho\rangle = 0 \tag{20}$$

where $C = [(kp + e^2g^2)^2 - e^4g^4\cos^2\xi]^{1/2}$ and where ε_c and ε_c^* are obtained from ε and ε^* be replacing c by ε and c^+ by ε^* in the transformation (18).

The linear form $\varepsilon_c c + \varepsilon_c^* c^+$ in equation (20) can now be eliminated by using the shift operator

$$D_p = \exp(-\delta c^+ + \delta^* c) \tag{21}$$

in analogy with the work of Filipowicz (1985). The choice $\delta = -egp\varepsilon_c^*/C$ leads to the equation

$$[p^2 - 2CN_c + 2e^2g^2C^{-1}(p\varepsilon_c)(p\varepsilon_c^*) - e^2g^2s - m^2]D_p|\rho\rangle = 0 \tag{22}$$

which is the equation of the 'harmonic oscillator' in the N_c representation. If we set $|\rho\rangle = D_p^+|n_c\rangle$ in equation (22) we find

$$p^2 = m^2 + 2C_{n_cs}kp \tag{23}$$

where

$$C_{n_cs} = C_{n_cs}(kp, p\varepsilon_c, p\varepsilon_c^*) = (kp)^{-1}[C(n_c + \tfrac{1}{2}) + \tfrac{1}{2}e^2g^2s - e^2g^2C^{-1}(p\varepsilon_c)(p\varepsilon_c^*)]. \tag{24}$$

Before writing the complete solution of the Dirac equation (3) we shall introduce the 4-vector $P = p - C_{n_cs}k$ which, according to equation (23), is on the mass shell, i.e. $P^2 - m^2 = 0$. Since $kp = kP$, $\varepsilon p = \varepsilon P$, and $\varepsilon^* p = \varepsilon^* P$ we may replace p by P in equation (24). Consequently, by combining equations (4), (6), (9), (12), (23) and (24) the solution of the Dirac equation can be given in the following explicit form:

$$\psi_{Pn_cs} = \exp[-iPx - iC_{n_cs}(kP, P\varepsilon_c, P\varepsilon_c^*)kx + ikxN_a][1 + e(k\boldsymbol{A}/2kP)]D_P^+\mathcal{P}(\boldsymbol{S} + s)v|n_c\rangle \tag{25}$$

where the shift operator D_P and the porjection operator $\mathcal{P}$ now have p replaced by P everywhere. This replacement can also be carried out in $|n_c\rangle$ according to equation (19).

By using the identity $A_\mu(kx) = \exp(i\omega tN_a)A_\mu(-\boldsymbol{k}\cdot\boldsymbol{r})\exp(-i\omega tN_a)$ and the transformation $\psi(x) = \exp(i\omega tN_a)\psi_0(x)$ the Dirac equation (3) can be cast in a form in which the vector potential is time independent. The solution $\psi_0(x)$ is given by equation (25) except that kxN_a is replaced by $-(\boldsymbol{k}\cdot\boldsymbol{r})N_a$. From the remaining time dependence of the exponential in equation (25) one finds that the eigenenergy of the corresponding stationary solution is given by $\mathcal{E} = E + C_{n_cs}\omega$, where $E = (\boldsymbol{P}^2 + m^2)^{1/2}$ is the kinetic energy of the electron and where C_{n_cs} is given by equation (24). In the $g \to 0$ limit $C_{n_cs}\omega$ reduces to the free photon energy $(n_c + \frac{1}{2})\omega$. Thus the shifted energy $Z\omega = C_{n_cs}\omega - (n_c + \frac{1}{2})\omega$ is the quantum-mechanical analogue of the ponderomotive potential energy.

Since the eigenkets $|n_c\rangle$ form a complete set in the photon Hilbert space and the transformation from the N_a representation to the N_c representation is unitary, the solutions (25) are orthogonal. The explicit proof follows very much the same lines as the proof of Filipowicz (1985) for the case of circularly polarised light. In fact equation (25) reduces to the Filipowicz equation, provided we identify his eigenfunction of the spin operator $\boldsymbol{S}$ as $(\boldsymbol{S}+s)|v\rangle$.

3. *S*-matrix element for multiphoton ionisation

In this section we shall use the wavefunction (25) to obtain a lowest-order S-matrix element for multiphoton ionisation in the high-intensity limit. In terms of non-relativistic time-dependent scattering theory, the *model* to be considered (Mittleman 1982) is the following. An atom is put into a monochromatic EM field of any polarisation. The field strength is raised adiabatically in the time interval $(-\infty, 0)$ to a constant value which persists for a long time τ. During that time, an electron interacts with the field and becomes ionised with a certain probability. After a time τ the field is switched off adiabatically in the time interval $(\tau, +\infty)$. This is a uniquely defined situation which allows the calculation of the ionisation probability per unit time from the S-matrix element

$$S_{fi} = -\int_{-\infty}^{+\infty} \langle \psi_f^-(t)|\, V_{\rm ph}(t)|\phi_i(t)\rangle \, {\rm d}t \tag{26}$$

where $V_{\rm ph}(t)$ is the time-dependent potential, representing the interaction between the electron and the field. In equation (26), $\phi_i(t) = \phi_i \exp(-{\rm i}E_i t)$ is the stationary initial-state wavefunction of a bound electron. Thus we have $[-(1/2m)\nabla^2 + V_{\rm c}]\phi_i = E_i\phi_i$, where $V_{\rm c}$ is an effective potential energy describing the interaction between the electron and the ionic core. The scattering wavefunction ψ_f^- is given by

$$\psi_f^-(t) = \phi_f^-(t) + \lim_{\nu\to 0} \frac{V_{\rm c}\phi_f^-(t)}{E_f - H(t) - {\rm i}\nu} \tag{27}$$

where $\phi_f^-(t)$ is the scattering solution of the time-dependent Schrödinger equation ${\rm i}\partial\phi_f(t)/\partial t = [-(1/2m)\nabla^2 + V_{\rm ph}(t)]\phi_f(t)$ satisfying the ingoing-wave boundary condition. In equation (27), $H = H(t) = [-(1/2m)\nabla^2 + V_{\rm c} + V_{\rm ph}(t)]$ is the full Hamiltonian in which $V_{\rm ph}(\pm\infty) = 0$ due to adiabatic switching. Thus, E_f is the kinetic energy of the outgoing electron in the absence of the EM field. Equation (27) describes the scattering of the electron by the ion core when the electron is removed under the action of the field. The underlying assumption of equations (26) and (27) is the *independence* of $V_{\rm c}$ on the field and thus on the time.

Usually it is assumed (Keldysh 1965, Faisal 1973, Reiss 1980, Krstic and Mittleman 1982) that $\psi_f^-(t)$ can be replaced by the non-relativistic 'Volkov' solution $\phi_f(t)$ of the time-dependent Schrödinger equation with the potential $V_{\rm ph}(t)$ in the S-matrix element (26). This seems to be a reasonable approximation for high field strengths but it has never been thoroughly tested. Since $V_{\rm c} \to -Z_{\rm c}e^2 r^{-1}$ the validity of this approximation surely depends on $Z_{\rm c}$ and it may only be applicable to $Z_{\rm c} = 0$ (Reiss 1980) because of the long-range nature of the Coulomb potential. In order to explore this question it is useful to formulate the model based on equations (26) and (27) in terms of QED. This also proves the interpretation in the photon picture of the non-relativistic transition

rate formula (Reiss 1980, Bashkansky *et al* 1987). Furthermore, the magnetic field is automatically included.

Within QED a scattering event is usually described by terms of the lowest possible order in the expansion of the S matrix. This will obviously not work in the present case, where the absorption of, say, 500 photons (Chin *et al* 1987) must be considered. It is, however, possible to formulate the scattering theory within QED in a manner which is analogous to the general non-relativistic treatment, described in many textbooks (e.g. Wu and Ohmura 1962, Taylor 1983). The treatment is based on the use of the Stückelberg-Feynman propagator (Greiner *et al* 1985). The application of this technique to our case indicates that the S-matrix element which corresponds to equation (26), but with $\psi_f^-(t)$ replaced by $\phi_f(t)$, is given by

$$S_{fi} = -ie\langle\psi_{Pn_cs}|\gamma^0\gamma A(kx)|\psi_i, l\rangle \tag{28}$$

where ψ_{Pn_cs} is given by equation (25) and where $|\psi_i, l\rangle$ is the direct product of the initial-state 4-spinor ψ_i and the photon state $|l\rangle$. The photons are assumed to be in the elliptically polarised state, defined by the 4-vector k in equation (1) and by the polarisation vector (2). Since $\gamma^0\gamma A$ is a Hermitian operator we have

$$S_{fi}^* = ie\int d^4x\,\psi_i^+\langle l|\gamma^0\gamma A(kx)|\psi_{Pn_cs}\rangle \tag{29}$$

where $X = -|e|\langle l|\gamma^0\gamma A(kx)|\psi_{Pn_cs}\rangle$ can be evaluated using boson-operator techniques (Louisell 1973). The result is

$$\begin{aligned} X = -\exp(-iPx + i(l+\tfrac{1}{2} - C_{n_cs})kx]\sum_{m=0}^{\infty}\,&[|e|g[(l+1)^{1/2}(\gamma^0\mathcal{E})D_{l+1,m}^+ + l^{1/2}(\gamma^0\mathcal{E}^*)D_{l-1,m}^+] \\ &+ (e^2g^2/2kp)(\gamma^0 k)][[\mathcal{E}^*\mathcal{E}l + \mathcal{E}\mathcal{E}^*(l+1)]D_{l,m}^+ \\ &- \cos\xi\{[(l+1)(l+2)]^{1/2}D_{l+2,m}^+ + [l(l-1)]^{1/2}D_{l-2,m}^+\}]\langle m|n_c\rangle]\mathcal{P}(\mathcal{S}+s)v \end{aligned} \tag{30}$$

where we have introduced the complete set of photon states $|m\rangle$ in the N_a representation. The matrix element $D_{l',m}^+$ is given by

$$D_{l',m}^+ = \langle l'|\exp(\delta_a a^+ - \delta_a^* a)|m\rangle \tag{31}$$

where

$$\delta_a = |e|gP(\varepsilon^*\cosh 2\chi + \varepsilon\sinh 2\chi)/C \tag{32}$$

according to equations (14), (18) and (21).

The integration over time in equation (29) results in the Dirac delta function $2\pi\delta[E_i - E + (l+\frac{1}{2})\omega - C_{n_cs}\omega]$ which expresses energy conservation in terms of the initial-state energy $E_i + (l+\frac{1}{2})\omega$ and the final-state energy $\mathcal{E} = E + C_{n_cs}\omega$. The factor $l+\frac{1}{2} - C_{n_cs}$, which must be positive, determines the dependence of the kinetic energy E on n_c for the two possible values of s. For example, for circular polarisation the Dirac delta function yields the condition

$$E = q\omega - Z\omega + E_i \tag{33}$$

where $q = l - n$ is the number of photons absorbed and where $Z\omega$ is the quantum-mechanical ponderomotive potential energy. The factor Z is given, according to equation (24), by

$$Z = (e^2g^2/kP)[n + \tfrac{1}{2} + \tfrac{1}{2}s - (P\varepsilon)(P\varepsilon^*)/(kP + e^2g^2)]. \tag{34}$$

For intense fields, l is large and hence the electromagnetic field should be described in terms of a classical vector potential. Using the phase representation of the photon field (Mittleman 1982) the transformation from the quantum-mechanical to the classical field is accomplished by putting $g = \lambda/\sqrt{l}$ in equation (1) and by letting l go to infinity. Thus $\lambda\omega$ is the classical field strength. In the following we shall show that in the classical limit and for any polarisation the amplitude (28) reduces to a relativistic amplitude in which $j = l - n$, with both l and n large, has the precise meaning of j (transverse) photons absorbed.

The derivation is based on two relations which we prove in appendix 2. The first one is

$$\lim_{\substack{l\to\infty \\ m\to\infty}} D^{+}_{l,m} = \sum_{q=-\infty}^{+\infty} (-1)^q \delta_{q,l-m} J_q(\zeta) \exp(-\mathrm{i}q\phi_\xi) \tag{35}$$

where

$$\zeta = \frac{2|e|\lambda}{kP} |\boldsymbol{P}\cdot\boldsymbol{\varepsilon}| \tag{36}$$

and

$$\phi_\xi = \tan^{-1}[(P_y/P_x)\tan(\xi/2)]. \tag{37}$$

The angle ξ is defined in equation (2), and P_y and P_x are the components of $P = (E, \boldsymbol{P})$ in the directions of $\boldsymbol{\varepsilon}_y$ and $\boldsymbol{\varepsilon}_x$, respectively. The second relation is

$$\lim_{\substack{m\to\infty \\ n\to\infty}} \langle m|n_c\rangle = \sum_{t=-\infty}^{+\infty} J_t(\eta)\delta_{2t,n-m} \tag{38}$$

where

$$\eta = (Z/2)\cos\xi. \tag{39}$$

The quantity $Z = e^2\lambda^2/kP$ is obtained from equation (24) by letting n go to infinity and it is thus the classical ponderomotive potential energy per unit frequency. As indicated by equations (35) and (38), finite contributions to the scattering amplitude (28) come from regions in the photon space in which the limit $l\to\infty$ implies that the photon numbers m and n both go to infinity in such a way that $l - m = q$ and $n - m = 2t$ stay finite. As shown by equation (30), each term in the amplitude (28) is either proportional to $g\sqrt{l'}$ or $g^2 l'$, where l' is a large integer. Hence the amplitude (28) becomes proportional to the classical field strength. By taking the high-photon limit in the exponential of the integrand (30) and by using equations (35) and (38) we obtain

$$\begin{aligned} S_{fi} = 2\pi\mathrm{i}\chi_s^{+}|e|\lambda\,(&\mathcal{J}_{j-1}\exp[\mathrm{i}(j-1)\phi_\xi]\gamma^0\boldsymbol{\varepsilon} + \mathcal{J}_{j+1}\exp[\mathrm{i}(j+1)\phi_\xi]\gamma^0\boldsymbol{\varepsilon}^* \\ &-(Z/|e|\lambda)[\![\mathcal{J}_j\exp(\mathrm{i}j\phi_\xi) + (\cos\xi/2)\{\mathcal{J}_{j+2}\exp[\mathrm{i}(j+2)\phi_\xi] \\ &+\mathcal{J}_{j-2}\exp[\mathrm{i}(j-2)\phi_\xi]\}]\!]\gamma^0 k)\Phi_i(\boldsymbol{P} - j\boldsymbol{k} + Z\boldsymbol{k})\delta(E_i - E + j\omega - Z\omega) \end{aligned} \tag{40}$$

where $j = l - n$ is the difference between the number of initial and final photons. The function $\mathcal{J}_j$, which depends on ζ, η and ϕ_ξ, is given by

$$\mathcal{J}_j = \mathcal{J}_j(\zeta, \eta, \phi_\xi) = \sum_{m=-\infty}^{+\infty} J_{-j-2m}(\zeta)J_m(\eta)\exp(2\mathrm{i}m\phi_\xi). \tag{41}$$

For $\phi_\xi = 0$ it reduces to the generalised Bessel function $J_{-j}(\zeta, \eta)$ (Reiss 1980). In equation (40), $\Phi_i(\boldsymbol{p})$ is the Fourier transform of the initial-state 4-spinor $\psi_i(\boldsymbol{r})$ and is given by

$$\Phi_i(\boldsymbol{p}) = \int \mathrm{d}^3\boldsymbol{r}\, \psi_i(\boldsymbol{r}) \exp(-\mathrm{i}\boldsymbol{p}\cdot\boldsymbol{r}). \tag{42}$$

We have used the abbreviation χ_s for the eigenvector (11) of the spin operator $\mathscr{P}\mathscr{S}$. Equation (40) shows that in this model the multiphoton absorption does not only occur as a consequence of absorption of j photons but also as a result of virtual processes in which one or two photons are either re-emitted or re-absorbed. The two-photon processes do not occur in the circularly polarised case. This interpretation also reveals one of the shortcomings of the lowest-order approximation (28) since the inclusion of the interaction between the ion core and the electron would introduce higher-order virtual processes along with modifications of the j, $j \pm 1$ and $j \pm 2$ terms in equation (40). Systematic improvements may be easier within the full photon picture than within the classical framework.

Equation (40) is our final result. Further developments for particular cases should be based on the general expression

$$\mathrm{d}W/\mathrm{d}\Omega = 2\pi\, \mathrm{Tr}(S\rho_i \bar{S}\rho_f) \tag{43}$$

which gives the probability, as a function of time τ, that an electron is emitted in a given direction as a result of multiphoton absorption of j photons. In equation (43), ρ_i is a density matrix which describes the orientation of the target atoms. The density matrix ρ_f describes the efficiency with which the detector detects the components of the spin in a given direction. The use of the scattering matrix elements (40) in equation (43) implies that we still consider the model pulse shape depicted in the beginning of this section. For random initial orientation and for a polarisation-insenstive detector, the probability (43) reduces to

$$\frac{\mathrm{d}W}{\mathrm{d}\Omega} = \frac{1}{2j_i+1} \sum_{m_j s} S_{m_j s} \bar{S}_{m_j s} \tag{44}$$

which contains an average over the initial magnetic quantum numbers m_j and a summation over the quantum numbers $s = \pm\sin\xi$. The probability per unit time is given by

$$\frac{\mathrm{d}w}{\mathrm{d}\Omega} = \lim_{\tau\to\infty} \frac{\mathrm{d}W}{\mathrm{d}\Omega} \tag{45}$$

in which the relation $2\pi\delta^2(E_i - E + j\omega - Z\omega) = \delta(E_i - E + j\omega - Z\omega)\tau$ must be used. It is in principle possible to write a general closed-form expression for the rate (45) using equation (40).

4. The non-relativaitic limit

In this section we derive the non-relativistic transition rate formula of Bashkansky *et al* (1987) from our equation (40). The derivation serves as a consistency check of our approach. It is based on the observations that $\gamma^0 \not{\varepsilon} = -\boldsymbol{\alpha}\cdot\boldsymbol{\varepsilon}$, $\gamma^0 \not{K} = \omega - \boldsymbol{\alpha}\cdot\boldsymbol{k}$ and

that in the non-relativistic limit and the long-wave approximation we have $\boldsymbol{\alpha}\cdot\boldsymbol{\varepsilon}\to(\boldsymbol{\varepsilon}\cdot\boldsymbol{P})/m$, $kP\to m\omega$, and $\boldsymbol{P}-j\boldsymbol{k}+Z\boldsymbol{k}\to\boldsymbol{P}$. According to equation (40), we then have

$$S_{fi}^{\mathrm{nr}}=-2\pi\mathrm{i}\frac{|e|\lambda}{m}\exp(\mathrm{i}j\phi_\xi)\Big(\big[\mathscr{J}_{j-1}\exp(-\mathrm{i}\phi_\xi)(\boldsymbol{P}\cdot\boldsymbol{\varepsilon})+\mathscr{J}_{j+1}\exp(\mathrm{i}\phi_\xi)(\boldsymbol{P}\cdot\boldsymbol{\varepsilon}^*)\big]$$
$$+\frac{Zm\omega}{\lambda|e|}\{\mathscr{J}_j+(\cos\xi/2)[\mathscr{J}_{j-2}\exp(-2\mathrm{i}\phi_\xi)+\mathscr{J}_{j+2}\exp(2\mathrm{i}\phi_\xi)]\}\Big)$$
$$\times\phi_i(\boldsymbol{P})\delta(E-E_i-j\omega+Z\omega) \tag{46}$$

where $E=\boldsymbol{P}^2/2m$, $Z=e^2\lambda^2/m\omega$, and where the function $\mathscr{J}_j=\mathscr{J}_j(\zeta,\eta,\phi_\xi)$, defined by equation (41), now depends on $\zeta=2|e|\lambda|\boldsymbol{P}\cdot\boldsymbol{\varepsilon}|/m\omega$ and $\eta=(Z/2)\cos\xi$. The momentum vector P is defined on the mass shell and it is the one which must be used in the limiting procedure. In the matrix multiplication of χ_s^+ with $\Phi_i(\boldsymbol{P})$ we keep only the large component which is $\phi_i(\boldsymbol{P})$. Since $\boldsymbol{P}\cdot\boldsymbol{\varepsilon}=|\boldsymbol{P}\cdot\boldsymbol{\varepsilon}|\exp(\mathrm{i}\phi_\xi)$ and since it can easily be proved that

$$2j\mathscr{J}_j+\zeta(\mathscr{J}_{j-1}+\mathscr{J}_{j+1})+2\eta[\mathscr{J}_{j-2}\exp(-2\mathrm{i}\phi_\xi)+\mathscr{J}_{j+2}\exp(2\mathrm{i}\phi_\xi)]=0 \tag{47}$$

the non-relativistic S-matrix element (46) reduces to

$$S_{fi}^{\mathrm{nr}}=2\pi\mathrm{i}\omega(j-Z)\mathscr{J}_j(\zeta,\eta,\phi_\xi)\exp(\mathrm{i}j\phi_\xi)\phi_i(\boldsymbol{P})\delta(E-E_i-j\omega+Z\omega) \tag{48}$$

for the absorption of j photons.

The rate corresponding to equation (48) is obtained by using the procedure associated with equation (45) and by taking the proper normalisation of the solution (25) into account. We have

$$\frac{\mathrm{d}w}{\mathrm{d}\Omega}=\lim_{\tau\to\infty}\int_0^\infty|S_{fi}^{\mathrm{nr}}|^2\frac{\boldsymbol{P}^2\,\mathrm{d}|\boldsymbol{P}|}{(2\pi)^3}$$
$$=\frac{(2m^3\omega^5)^{1/2}}{(2\pi)^2}(j-Z)^2(j-Z-E_{\mathrm{B}}/\omega)^{1/2}|\phi_i(\boldsymbol{P})|^2|\mathscr{J}_j(\zeta,\eta,\phi_\xi)|^2. \tag{49}$$

The integration over $|\boldsymbol{P}|$ is accomplished by using the relation (Reiss 1980)

$$\delta(\boldsymbol{P}^2/2m+E_{\mathrm{B}}-j\omega+Z\omega)$$
$$=(m/2\omega)^{1/2}(j-Z-E_{\mathrm{B}}/\omega)^{-1/2}\delta(|\boldsymbol{P}|-(2m\omega)^{1/2}(j-Z-E_{\mathrm{B}}/\omega)^{1/2}) \tag{50}$$

where $E_{\mathrm{B}}=-E_i$ is the positive ionisation (binding) energy of the electron in the ground state of the unperturbed atom. It should be noted that the length of the $\boldsymbol{P}$ vector in the Fourier transform $\phi_i(\boldsymbol{P})$ must fulfil the energy conservation relation $E=\boldsymbol{P}^2/2m=(j-Z)\omega-E_{\mathrm{B}}>0$. This relation also shows that $j>Z+E_{\mathrm{B}}/\omega$ (Reiss 1980).

The rate expression (49) is identical with the one reported by Bashkansky *et al* (1987). To see this we note that $\mathscr{J}_{-j}^*(\zeta,\eta,\phi_\xi)=(-1)^j\mathscr{J}_j(\zeta,-\eta,\phi_\xi)$ according to equation (41) and that

$$\zeta=\frac{2|e|\lambda}{m\omega}|\boldsymbol{P}\cdot\boldsymbol{\varepsilon}|=2\sin\theta[Z(j-Z-E_B/\omega)(1+\cos\xi\cos2\phi)]^{1/2} \tag{51}$$

where θ and ϕ are the polar and azimuthal angles of $\boldsymbol{P}$, respectively. In their expression, Bashkansky *et al* (1987) use $\mathscr{J}_{-j}$ rather than $\mathscr{J}_j$ and erroneously write $Z=e^2A^2/2m\omega$ instead of $e^2\lambda^2/m\omega$ ($A^2=2\lambda^2$ holds only for circular polarisation).

5. Discussion

The QED solutions (25) may be reduced to the classical solutions

$$\psi_P(x) = (1 + e\not{k}\not{A}/2kP) \exp\left[i\left(-Px - \int_{(kx)_0}^{(kx)} (kP)^{-1}(ePA - \tfrac{1}{2}e^2A^2)\, d(kx) \right) \right] \mathcal{P}u \tag{52}$$

using the same technique that was used to obtain the S-matrix element (40) from its QED version (28). The solutions can also be obtained directly by solving the Dirac equation for the external field potential

$$A = \lambda[\varepsilon \exp(-ikx) + \varepsilon^* \exp(ikx)] \tag{53}$$

where the 4-vectors ε and ε^* fulfil equations (2). In equation (52), P is on the mass shell and consequently $\mathcal{P}u$, where u can be any bispinor, is a plane-wave bispinor. This follows from the definition of $\mathcal{P}$ given by equation (8) and from the fact that $P^2 - m^2 = 0$.

The result given above has some relevance to a comment made by Filipowicz (1985) on his classical solution. This author has $(2kp)^{-1}(\not{P} - e\not{A} + m)\not{k}u'$ instead of $(1 + e\not{k}\not{A}/2kP)\mathcal{P}u$ in equation (52) and claims that, since u' is an arbitrary bispinor, his is a more general solution than the Volkov solution (Berestetskii *et al* 1971) where u' is the plane-wave bispinor. However, using the definition (8) of $\mathcal{P}$ and the relation $\not{k}\mathcal{P} = \not{K}$, it can be seen that both forms are identical provided $u' = u$. This means that, since $\mathcal{P}u$ is a plane-wave bispinor for any u, the Filipowicz solutions and our classical solutions are exactly the same as the Volkov solution.

It is important to realise that our QED procedure for multiphoton ionisation, even with systematic improvements, is equivalent to the model problem defined by equations (26) and (27) in the non-relativistic limit. Nevertheless, one may learn a great deal by using the basis set (25) to obtain systematic improvements of the S-matrix element (28). First, there is the problem of including the effect of the Coulomb field. It is rewarding that the Coulomb matrix element can be worked out exactly using the basis set (28). Second, there is the question of the gauge dependence of the matrix element (28). Although the classical Volkov solution is invariant under gauge transformations of the type

$$A'_\mu = A_\mu + \partial_\mu \eta = 0 \qquad \Box \eta = 0 \tag{54}$$

where $\eta = \eta(kx)$ (Reiss 1979), the matrix element (28) is in general not so. The reason for this feature is that the initial and final states in the matrix element (28) are not solutions of the same Dirac equation. As far as we know, the gauge-dependence problem has only been studied within the classical approximation (Antunes Neto and Davidovich 1984) where the A^2 term introduces additional complications (Kupersztych 1979). Third, there is the question of relativistic effects. For example, according to equation (24), the ponderomotive potential is actually angle dependent in the relativistic limit. This means that the minimum number of photons required for ionisation may vary as a function of the direction of emission of the photoelectron. It seems that the relativistic matrix element (40) must be worked out in particular cases to study this and other effects, such as spin polarisation.

Whether the study of the model problem discussed here is relevant to the understanding of multiphoton ionisation of many-electron atoms by strong laser pulses is another question. The use of the rate equation (49) in one form or another indicates that it correctly predicts, at least qualitatively, a number of experimental features. These

features include the suppression of low-energy peaks in ATI spectra (Bucksbaum *et al* 1986) including the general shape of the envelope of the peak structure for both circularly and linearly polarised light (Reiss 1980, 1987a, b). Equation (49) has also been found to yield angular distributions which agree quite well with experimental findings (Bashkansky *et al* 1987). The model based on equation (49) predicts an intensity-dependent shift of the ATI peaks due to the ponderomotive potential energy. This shift is absent in ATI spectra for above-picosecond laser pulses. This can, however, be explained by the fact that the model discussed in the present paper more or less corresponds to a situation in which the electron stays in the pulse all the time as realised in subpicosecond experiments (Freeman *et al* 1987, Muller *et al* 1988). The above-picosecond experiments, on the other hand, correspond to a situation in which the electron rapidly leaves the pulse before the pulse decays (Becker *et al* 1986, Agostini *et al* 1987). There also are some indications that equation (49) predicts absolute rates which are too small by as much as orders of magnitude when a Coulomb field is present (Bashkansky *et al* 1987, Shakeshaft and Potvliege 1987). Consequently, it is important to study the connection between the present approach and more realistic but elaborate theories (Crance 1987, Shakeshaft and Potviliege 1987, Shakeshaft and Tang 1987).

In conclusion, we have obtained a complete orthogonal set of solutions of the Dirac equation for an electron interacting with a quantised, elliptically polarised, electromagnetic field. We have used the wavefunctions for a derivation of a relativistic S-matrix amplitude which describes the one-electron ionisation of an atom that has absorbed a given arbitrarily large number of photons. By considering the non-relativistic limit of the S-matrix element, we have obtained the transition rate formula of Bashkansky *et al* (1987) rigorously in terms of the number of absorbed photons. It is suggested that the present QED result can be improved by taking the Coulomb field into account in the final scattering state.

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Appendix

Starting from the transformation (14) we wish to define a complete set $\{|n_c\rangle\}$ ($n = 0, 1, 2 \ldots$) of photon states. We require that

$$\begin{aligned} c|n_c\rangle &= \sqrt{n_c}|n_c - 1\rangle \\ c^+|n_c - 1\rangle &= \sqrt{n_c}|n_c\rangle \end{aligned} \tag{A1.1}$$

with $|-1\rangle = 0$. By substituting the transformation (14) into equations (A1.1) and by using the closure relation $\Sigma|n\rangle\langle n| = 1$ in the N_a representation, we find that the 'vacuum'

state $|0_c\rangle$ must fulfil the recursion relation

$$\cosh\chi\sqrt{n+1}\langle n+1|0_c\rangle-\sinh\chi\sqrt{n}\langle n-1|0_c\rangle=0 \tag{A1.2}$$

with $\langle 1|0_c\rangle=0$. Consequently, we have $\langle n'|0_c\rangle=0$ if n' is odd. If n' is even, we find from equation (A1.2) that

$$\langle 2n|0_c\rangle=\tanh^n\chi\left(\frac{(2n-1)!!}{(2n)!!}\right)^{1/2}t \tag{A1.3}$$

where $t=\langle 0|0_c\rangle$, $(-1)!!\equiv 1$ and $n=0,1,2\ldots$. Hence

$$|0_c\rangle=t\sum_{n=0}^{\infty}\tanh^n\chi\left(\frac{(2n-1)!!}{2n!!}\right)^{1/2}|2n\rangle \tag{A1.4}$$

where t is determined by the normalisation condition $\langle 0_c|0_c\rangle=1$. Using the expansion of $(1-\tanh^2\chi)^{-1/2}$ in powers of $\tanh\chi$, it can easily be seen that $t=(\cosh\chi)^{-1/2}$, which proves equation (16). Once we have found $|0_c\rangle$ we can generate a set of states

$$|n_c\rangle=\frac{(c^+)^{n_c}}{\sqrt{n_c!}}|0_c\rangle. \tag{A1.5}$$

These states, given by equation (17), are eigenstates of the Hermitian 'number' operator $N_c=\frac{1}{2}(c^+c+cc^+)=c^+c+\frac{1}{2}$, as can be seen from the following argument. Since $c|0_c\rangle=0$ according to the definition (14) and equation (A1.4), we have $N_c|0_c\rangle=\frac{1}{2}|0_c\rangle$. From the assumption $N_c|n_c-1\rangle=(n_c-\frac{1}{2})|n_c\rangle$ and from equation (A1.5), it follows that

$$N_c|n_c\rangle=(n_c+\tfrac{1}{2})|n_c\rangle. \tag{A1.6}$$

Hence, by induction, the states (A1.5) fulfil the eigenvalue equation (A1.6), where the operator N_c is Hermitian by virtue of the definitions (14) of c and c^+. It follows that the set $\{|n_c\rangle\}$ is complete. It is easily checked by induction that the states (A1.5) also fulfil the starting relations (A1.1). Hence, it also follows that $\langle n_c|n_c\rangle=1$.

Appendix 2

The shift operator (21) can be written in the N_a representation by using the definition (14) and the inverse relation (18) to express ε_c and ε_c^* in terms of ε and ε^*. The result is

$$D_p^+=\exp(\delta_a a^+-\delta_a^* a) \tag{A2.1}$$

where δ_a is given by equation (32) which we repeat here:

$$\delta_a=-egp(\varepsilon^*\cosh 2\chi+\varepsilon\sinh 2\chi)/C. \tag{A2.2}$$

The factor C has been given in connection with equation (20). We use the Baker-Hausdorff theorem in equation (A2.1) according to which

$$e^{A+B}=e^B e^A e^{[A,B]/2} \tag{A2.3}$$

for two non-commuting operators A and B which satisfy $[A,[A,B]]=[B,[A,B]]=0$. The result is

$$D_p^+=\exp(-\tfrac{1}{2}|\delta_a|^2)\sum_{q=-\infty}^{+\infty}(\delta_a a^+)^q\sum_{k=0}^{\infty}\frac{(\delta_a a^+)^k(-\delta_a^* a)^k}{(k+q)!\,k!}. \tag{A2.4}$$

Consequently, $D^{+}_{l',m}$ in equation (30) becomes

$$D^{+}_{l',m} = \exp(-\tfrac{1}{2}|\delta_a|^2) \sum_{q=-\infty}^{+\infty} \delta_a^q [l'(l'-1)\ldots(l'-q+1)]^{1/2} \times \sum_{k=0}^{\infty} \frac{(-\delta_a \delta_a^*)^k m(m-1)\ldots(m-k+1)}{(k+q)!\,k!} \delta_{l'-m,q}. \tag{A2.5}$$

Now the classical limit is taken by keeping $q = l' - m$ finite while $l' \to \infty$ and $m \to \infty$ simultaneously. We use the relation $g\sqrt{m} \to \lambda$ and observe that $\delta_a \sqrt{m} \to -e\lambda P\varepsilon^*/kP$. The same limit prevails for $\delta_a \sqrt{l'}$ in equation (A2.5), where the square root can be replaced by $l'^{q/2}$. Using the definitions (36) and (37), we have

$$\lim_{\substack{l' \to \infty \\ m \to \infty}} D^{+}_{l'm} = \sum_{q=-\infty}^{+\infty} (-1)^q \delta_{l'-m,q} J_q(\zeta) \exp(-\mathrm{i}q\phi_\xi) \tag{A2.6}$$

where we have used the definition

$$J_q(\zeta) = (\zeta/2)^q \sum_{k=0}^{\infty} \frac{(\zeta/2)^{2k}}{(k+q)!\,k!} (-1)^k \tag{A2.7}$$

of a Bessel function of the first kind for integer q. This proves equation (35).

The proof of equation (38) is rather lengthy. It is based on the idea that

$$J_t = \lim_{\substack{m \to \infty \\ n \to \infty \\ n-m=2t}} \langle m|n_c\rangle \tag{A2.8}$$

must fulfil the same recursion relations as the Bessel function $J_t = J_t(\eta)$, namely

$$2tJ_t = \eta(J_{t+1} + J_{t-1}) \tag{A2.9}$$

and

$$\mathrm{d}J_t/\mathrm{d}\eta = \tfrac{1}{2}(J_{t-1} - J_{t+1}) \tag{A2.10}$$

with $J_0(0) = 1$. If this can be proved then we know that equation (A2.8) defines a Bessel function.

We start from equation (A1.6) and, by using the transformation (14), express the operator N_c in terms of $N_a = \frac{1}{2}(a^+a + aa^+)$ and $M_a = \frac{1}{2}(a^2 + a^{+2})$. The result is

$$\sum_{m=0}^{\infty} (\cosh 2\chi N_a - \sinh 2\chi M_a)|m\rangle\langle m|n_c\rangle = (n + \tfrac{1}{2})|n_c\rangle \tag{A2.11}$$

which gives

$$\sum_{m=0}^{\infty} [\![(m \cosh 2\chi - n - \tfrac{1}{2})\langle m|n_c\rangle - \tfrac{1}{2}\sinh 2\chi\{[(m-1)^{1/2} m^{1/2}\langle m-2|n_c\rangle] + [(m+1)(m+2)]^{1/2}\langle m+2|n_c\rangle\}]\!] = 0 \tag{A2.12}$$

where

$$\chi = -\tfrac{1}{2}\tanh^{-1}\left(\frac{e^2 g^2 \cos\xi}{kP + e^2 g^2}\right) \tag{A2.13}$$

according to equation (19). We introduce the difference $2t = n - m$ and examine equation (A2.12) when both m and n become large but t stays finite. We need the limit

$$\eta = -\lim_{n\to\infty} n\chi = \tfrac{1}{2}Z\cos\xi \tag{A2.14}$$

where $Z = e^2\lambda^2/kP$ is the ponderomotive potential energy per unit frequency in accordance with equations (33), (34) and (39). The limit follows from equation (A2.13) by using $g\sqrt{n} \to \lambda$. Since $\cosh 2\chi \to 1$ and $\sinh 2\chi \to 2\chi$ in that limit we have

$$2t\langle m|n_c\rangle + \chi m(\langle m-2|n_c\rangle + \langle m+2|n_c\rangle) = 0 \tag{A2.15}$$

for large m and n according to equation (A2.12). The use of equation (A2.14) and of the definition of t in equation (A2.15) shows that the quantity (A2.8) fulfils the first recursion relation (A2.9).

In order to prove the second recursion relation (A2.10) we need the derivative

$$\frac{\mathrm{d}}{\mathrm{d}\chi}\langle m|n_c\rangle = \frac{1}{\cosh^2\chi}\langle m|Q/\sqrt{n!}|0_c\rangle \tag{A2.16}$$

where

$$Q = n\frac{\mathrm{d}c^+}{\mathrm{d}\chi}(c^+)^{n-1} + \tfrac{1}{2}(n-1)n(c^+)^{n-2} + \tfrac{1}{2}c^{+n}a^{+2}. \tag{A2.17}$$

The first two terms in equation (A2.17) represent $\mathrm{d}c^{+n}/\mathrm{d}\chi$ and the third one is $\mathrm{d}|0_c\rangle/\mathrm{d}\chi$. In taking the classical limit of equation (A2.16) we observe that

$$\lim_{n\to\infty}\frac{1}{n}\frac{\mathrm{d}}{\mathrm{d}\chi} = -\frac{\mathrm{d}}{\mathrm{d}\eta} \tag{A2.18}$$

$$\lim_{n\to\infty}\frac{\mathrm{d}c^+}{\mathrm{d}\chi} = -a \tag{A2.19}$$

and that c^+ and a^+ commute for large n. Hence for large m and n but for fixed $2t = n - m$ equation (A2.16) becomes

$$-\frac{\mathrm{d}}{\mathrm{d}\eta}\langle m|n_c\rangle = -\langle m+1|(n-1)_c\rangle + \tfrac{1}{2}[\langle m|(n-2)_c\rangle + \langle m|(n+2)_c\rangle] \tag{A2.20}$$

which is identical with the second recursion relation (A2.10) according to the definition (A2.8).

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