Multiphoton ionization of hydrogen in ultrastrong laser fields

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The three previous calculations of this process yield differing results for the transition rate. We show the relations between them and difficulties with each of them. One difficulty is that the finite spatial extent of the laser field has been omitted. It is also found that a laser field, which is sufficiently intense to be labeled ultrastrong, makes the electron move relativistically so that it becomes necessary to use Volkov states to describe the electron in the laser field. The transition rate is obtained, with the use of a CO_2 laser as an example, and it is found that the transition rate rises as the laser intensity rises. This is a consequence of the use of relativistic kinematics and is not true nonrelativistically.

I. INTRODUCTION

When an atom is illuminated with a sufficiently intense electromagnetic wave (*a* laser) the motion of the electron is determined by that field more than by the interaction with the nucleus. The recognition of this fact has lead to several theoretical analyses[1–3] of the multiphoton ionization problem in ultrastrong laser fields which all yield different results for the transition probability per unit time. In Sec. II we provide a well-defined thought experiment which these theories should describe and show the flaws in each of the previous calculations. We show that (for a hydrogen atom example) the electron must be treated relativistically, and associated with this fact, we find that the dipole approximation for the laser is a poor one. These facts and the finite spatial extent of the laser field are incorporated into a calculation in Sec. III, where we find that the ionization rate, as a function of the laser intensity, is a rising function of the laser intensity and that this fact is a direct consequence of the use of relativistic kinematics of the electron.

II. ANALYSIS OF PREVIOUS WORK

The experiment that we have in mind here is the following: An atom is in its ground state in the absence of a laser field. Then in the rest frame of the atom the laser electric field amplitude E is adiabatically increased from zero to an ultrastrong plateau value (defined below) and held constant for a time T. It is then adiabatically decreased to zero and the probability of ionization is measured. Since the initial and final measurements on the atom are made in the absence of the field, there is no difficulty in defining the atomic state. The experiment is repeated with the same plateau value of the electric field but varying T and the variation of the curve of ionization probability versus T yields the ionization rate per unit time for ultrastrong fields. The experiment is then repeated for different plateau values in order to obtain the ionization rate as a function of field strength.

In order to reach the high plateau values of the laser intensity it must be increased adiabatically from zero to this value. A similar decrease occurs when the atom emerges from the laser. This implies that the atom is in the changing field for a finite length of time. It can be ionized during that time and if the ionization probability is essentially unity during that interval then the measurement which we are describing would be very difficult to perform.

We shall also see that the atom will have to overcome a potential energy which is of the order of the electron's rest energy in order penetrate into the laser beam. This means that the neutral atomic beam will have to have a very large kinetic energy in order to penetrate the laser beam. In more reasonable laser intensities thermal beams may not penetrate at all. For example, for a CO_2 laser of intensity 10^{12} W/cm^2 an atom will require of the order of 6 eV to penetrate into the interior of the laser beam.

The first applicable calculation by Keldysh[1] started from an exact expression for the S matrix ($\hbar=c=1$),

$$S_{\vec{q},0}=-i\langle\psi_{\vec{q}}^{(-)},H'\phi_0\rangle\ , \tag{2.1}$$

where ϕ_0 is the initial state of the atom. We deal only with hydrogen as the simplest example so that

$$\phi_0 = u_0(\vec{r})e^{-iW_0 t} . \tag{2.2}$$

The exact wave function, with incoming wave boundary conditions, satisfies

$$\left[i\frac{\partial}{\partial t} - H(t) \right] \psi_q^{(-)} = 0 \tag{2.3}$$

and

$$\lim_{t\to+\infty} \psi_q^{(-)} = u_q^{(-)}(\vec{r})e^{-iW_q t} , \tag{2.4}$$

where the spatial part of the initial and final wave functions both satisfy

$$(W_j - H_0)u_j(\vec{r}) = 0, \quad j = 0, \vec{q} . \tag{2.5}$$

The total Hamiltonian, $H(t)$, is written

$$H(t) = H_0 + H'(t) , \tag{2.6}$$

where H_0 is the atomic Hamiltonian in the absence of the laser and $H'(t)$ is the laser-atom interaction which is assumed to vanish when the laser field is zero. (In this, and all other calculations, the laser is treated as a classical- single-mode electromagnetic field. We shall only discuss the case of a linearly polarized wave.) Keldysh's approximation consisted of replacing the exact final wave function by one in which the electron-nucleus interaction is neglected on the grounds that its effect is small in the final continuum state compared to the interaction with the laser. Then the approximate S matrix becomes

$$S_{\vec{q},0} \cong -i\langle \chi_q^{(-)}, H'\phi_0 \rangle , \tag{2.7}$$

where

$$\left[i\frac{\partial}{\partial t} - T - H'(t) \right] \chi_q^{(-)} = 0 , \tag{2.8}$$

and where T is the kinetic energy operator of the electron. Keldysh worked in the dipole approximation in the r.E gauge and after some very lengthy calculations obtained a result for the total transition probability per unit time which, in the intense field limit, can be written as

$$w = \frac{\mathscr{R}_\infty}{\hbar} \sqrt{3\pi} 2^{-5/4} \left[\frac{E}{E_0} \right]^{1/2} . \tag{2.9}$$

Here E_0 is a convenient atomic unit for electric field intensity, $E_0 = e/2a_0^2 = 25.7 \times 10^8$ V/cm.

Pert's[3] calculation followed the same steps exactly except that he worked in the p.A gauge, also in dipole approximation. He obtained a transition matrix for absorption of l photons of the form

$$T_l = l\omega J_l(\vec{\alpha}\cdot\vec{q})\tilde{u}_0(q) , \tag{2.10a}$$

where $\vec{\alpha} = e\vec{E}/m\omega^2$, $\vec{q}$ is the momentum of the ionized electron, and $\tilde{u}_0$ is the Fourier transform of the initial bound state

$$\tilde{u}_0(q) = 8\sqrt{\pi} a_0^{-5/2}(q^2 + a_0^{-2})^{-2} . \tag{2.10b}$$

This leads to a total ionization rate which can be shown to behave as

$$w \sim E^{-1} \ln \left[\frac{E}{E_0} \right] , \tag{2.10c}$$

which is disturbingly different from (2.9). Clearly this gauge dependence is unacceptable. One reason for its appearance is the identification of the final (ionized) state in the *presence* of the laser. This is not a gauge invariant procedure.

Another calculation[2] started from an equivalent exact expression for the S matrix

$$S_{\vec{q},0} = -i\langle \phi_{\vec{q}}^{(-)}, H'\phi_0 \rangle - i\langle \phi_{\vec{q}}^{(-)}, H'G^{(+)}H'\phi_0 \rangle , \tag{2.11}$$

where $G^{(+)}$ is the full causal Green's function of the problem,

$$\left[i\frac{\partial}{\partial t} - H(t) \right] G^{(+)} = 1 . \tag{2.12}$$

The first term of (2.11) is a one-photon transition which can be discarded on the grounds of energy conservation. The Green's function in the second term is replaced by a simpler one, G_0^+, in which the electron-nucleus interaction is neglected compared to the interaction with the laser. Again the dipole approximation and the p.A gauge were used. An additional approximation used was the replacement of the function $\phi_q^{(-)}$, which is a continuum function of an electron in the field of the nucleus, by a plane wave. This will not affect the order of magnitude of the results but certainly will introduce factors of 2 or such.

The result for the T matrix was

$$T_l = l\omega J_l(\vec{\alpha}\cdot\vec{q})J_0(\vec{\alpha}\cdot\vec{q})\tilde{u}_0(q) , \tag{2.13a}$$

and for the total transition rate it was

$$w = \frac{4}{\pi} \frac{\mathscr{R}_\infty}{\hbar} \left[\frac{\hbar\omega}{\mathscr{R}_\infty} \right]^2 \left[\frac{E_0}{E} \right] . \tag{2.13b}$$

This is similar to but not quite the same as (2.10) and again the difference is disturbing.

If the first term of (2.11) is retained the approximations described above allow (2.11) to be written as

$$S_{\vec{q},0} \cong -i\langle \lambda_{\vec{q}}, (1+H'G_0^{(+)})H'\phi_0\rangle \ , \qquad (2.14)$$

where $\lambda_{\vec{q}}$ is a plane wave state. The form (2.7) is very similar to this and if the laser were a field of finite spatial extent we could use

$$\chi_{\vec{q}} = (1+G_0^{(+)}H')\lambda_{\vec{q}} \ , \qquad (2.15a)$$

which would make the forms (2.7) and (2.14) identical. However, all three calculations described above used a laser field which was infinite in its spatial extent in which case a careful examination of (2.15a) shows that it must be changed to the form

$$J_0(\vec{\alpha}\cdot\vec{q})\chi_{\vec{q}} = (1+G_0^{(+)}H')\lambda_{\vec{q}} \ . \qquad (2.15b)$$

The additional factor, J_0, arises from the definition of G_0^+ as

$$\left[i\frac{\partial}{\partial t} - T - H'(t) \right] G_0^{(+)} = 1 \ , \qquad (2.16)$$

where in the p.A gauge it can be written explicitly as

$$G_0^{(+)}(\vec{r}t,\vec{r}\,'t') = -i\Theta(t-t')\lim_{\eta\to 0+}\int \frac{d^3k}{(2\pi)^3} e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')} e^{-i(\epsilon_k - i\eta)(t-t')} \sum_{n,n'} J_n(\vec{\alpha}\cdot\vec{k}) J_{n'}(\vec{\alpha}\cdot\vec{k}) e^{-i\omega(nt-n't')} \ . \qquad (2.17)$$

Care must be taken in performing the integrals implied in (2.15) with the result (2.15b). The factor of J_0 in (2.15b) explains the discrepancy between (2.10a) and (2.13a) and since we shall work with a laser of finite extent in Sec. III we shall use the simpler form (2.7), of the completely equivalent forms of the S matrix.

The justification for the neglect of the electron-nucleus interaction compared to the electron-laser interaction in these calculations is the fact that the electric field which the electron experiences due to the laser is larger than some average value of the field due to the nucleus. The laser electric field is then larger than some field of the order of $E_0 = e/2a_0^2$. Then just treating the electron classically, its peak velocity in this field is

$$\frac{v}{c} \sim \frac{eE_0}{m\omega c} = \left[\frac{\mathcal{R}_\infty}{\hbar\omega} \right] \frac{e^2}{\hbar c} \sim 1 \ ,$$

where we have used a CO_2 laser ($\hbar\omega \simeq 0.1$ eV) as an example. Clearly the electron must be described relativistically. Moreover, the amplitude of the electron's oscillation times the laser wave number is exactly the same as v/c so the dipole approximation also becomes suspect.

The wave function of a Dirac electron in a traveling electromagnetic wave of infinite spatial extent was given many years ago by Volkov.[4] We have generalized his work to the situation of a traveling wave in a beam which has a finite extent in the directions perpendicular to the direction of propagation of the wave. This is described in the Appendix.

The Hamiltonian describing the atom in the field of the classical-single-mode electromagnetic field is a periodic function of time. This allows a simple proof[5] of the fact that the energy transfer to the particles is limited to integer multiples of ω, the frequency of the field. However, for a spatially inhomogeneous laser it is known both experimentally[6] and theoretically[7] that the time-averaged field intensity acts as a ponderomotive potential affecting the energy of the electron. This ponderomotive potential is

$$U_P^{\text{N.R.}}(r) = \frac{e^2E^2(r)}{4m\omega^2} \ , \qquad (2.18)$$

where $E(r)$ is the electric field amplitude and m is the electron mass. This apparent contradiction, in which, on the one hand, we know that only an integral number of photons is transferred and on the other hand we know that the electron emerges with a continuous distribution in energy, is easily resolved. The key is that the proton looses some of the energy. This may be seen in the following way. The hydrogen atom couples to the ponderomotive potential essentially through the electron since the coupling to the proton is m/M smaller. As the atom enters the laser beam it is slowed up by something like (2.18) until the ionization occurs at some point R_0. From that point on the proton is essentially decoupled from the field and so it leaves the laser having lost the energy $U_P^{\text{N.R}}(R_0)$.

The electron absorbs an integral number of photons, $l\omega$, and is expelled from the laser by the ponderomotive potential $U_P^{\mathrm{N.R}}(R_0)$ so that its energy upon leaving the laser is

$$E_q = W_0 + l\omega + U_P^{\mathrm{N.R}}(R_0) , \tag{2.19}$$

where W_0 is its original energy in the hydrogen atom.

The result described above, (2.18) and (2.19), is essentially nonrelativistic in that the ponderomotive potential in (2.18) is the time average of $e^2A^2(\vec{r},t)/2m$ which arises from the kinetic energy term of the Schrödinger equation. A more general result is obtained from the Dirac equation which we discuss now.

We describe the atom in the laser field in terms of center of mass and relative coordinates by

$$\left[i\frac{\partial}{\partial t} - T_R - [\vec{\alpha}\cdot\vec{\pi} + \beta m + V(r)] \right] \Psi = 0 , \tag{2.20}$$

where $\vec{R}$ is the center-of-mass coordinate and $\vec{r}$ is the position of the electron relative to the proton. Here $\vec{\alpha}$ and β are the usual Dirac matrices of the electron and

$$\vec{\pi} = \vec{p}_r + e\vec{A} , \tag{2.21}$$

where

$$\begin{aligned} \vec{A} &= \vec{a}(\vec{\rho})\cos\phi , \\ \phi &= \omega t - \vec{k}\cdot\left[\vec{R} + \frac{M}{M+m}\vec{r}\right] \\ &\simeq \omega t - \vec{k}\cdot(\vec{R}+\vec{r}) . \end{aligned} \tag{2.22}$$

Here $\vec{a}(\vec{\rho})$ is the (slowly varying) amplitude of the vector potential describing the laser and $\vec{\rho}$ is that part of $\vec{R}$ which is perpendicular to $\hat{k}$. We take $T_R = P_R^2/2(M+m)$ so that the internal motion of the atom is described relativisitically but the center of mass motion is described nonrelativistically. This is permissable since $M >> m$. The wave function of (2.20) can be decomposed into large and small components in a familiar way,

$$\Psi = \begin{bmatrix} U_1 \\ U_2 \end{bmatrix} , \tag{2.23}$$

and separated into a pair of equations

$$(E - m - V)U_1 = \vec{\sigma}\cdot\vec{\pi}U_2 , \tag{2.24a}$$

$$(E + m - V)U_2 = \vec{\sigma}\cdot\vec{\pi}U_1 , \tag{2.24b}$$

where we have used the short hand

$$E \equiv i\frac{\partial}{\partial t} - T_R . \tag{2.25}$$

We solve (2.24b) for U_2 and substitute back into (2.24a) to obtain an equation for U_1. Then $\vec{\pi}$ can be taken to commute with $(E+m-V)^{-1}$ since this involves the neglect of terms of order ω, and L^{-1} compared to m. (L is the cross-sectional size of the laser beam.) We must also drop a term resulting from the commutator $[p_r, V]$ but this can be shown to contribute a term of order $\alpha_F \sim (137)^{-1}$ compared to V itself. The resulting equation can then be written as

$$[(E-V)^2 - m^2]U_1 = [p_r^2 + \tfrac{1}{2}e^2a^2(\vec{\rho}) + \tfrac{1}{2}e^2a^2(\vec{\rho})\cos 2\phi + e\vec{A}\cdot\vec{p}_r + e\vec{\sigma}\cdot\vec{H}]U_1 , \tag{2.26}$$

where $\vec{H} = \vec{\nabla}\times\vec{A}$. The last three terms on the right-hand side oscillate with time and will be treated as perturbations and are dropped. The V^2 term on the left-hand side of (2.26) is an order α_F smaller than the term linear in V and so is also dropped and the remaining equation (having kept only first-order relativistic terms) is

$$(E^2 - 2EV - m^2)U_1 = [p_r^2 + 2m^2x^2(\rho)]U_1 , \tag{2.26a}$$

where we have defined

$$x(\vec{\rho}) = \frac{e}{2m} |\vec{a}(\vec{\rho})| . \tag{2.27}$$

The fact that $\vec{a}(\vec{\rho})$ is slowly varying can be exploited to give the approximate solution to (2.26a)

$$U_1 = u_0(r,\eta) \times \exp i \left[\int^{R} d\vec{R}'\cdot\vec{p}_i(\vec{R}') - t\left[W_0 + \frac{p_i^2}{2M} \right] \right] , \tag{2.28}$$

where the field free energy of hydrogen is

$$W_0 \simeq m - \mathscr{R}_\infty \tag{2.28a}$$

and the bound state wave function is

$$u_0 = \eta^{3/2}\pi^{-1/2}e^{-\eta r} , \tag{2.28b}$$

where

$$\eta=\frac{1}{a_0}(1-\tfrac{1}{2}\alpha_F^2)(1+2x^2)^{1/2}\ . \qquad (2.28c)$$

The center-of-mass momentum $\vec{p}_i(\vec{R})$ is the classical momentum[8] whose evolution is governed by the Hamiltonian

$$H_c(\vec{P},\vec{R})=\frac{P^2}{2M}+U_P(\vec{R})\ , \qquad (2.29)$$

where

$$U_P(\vec{R})=(m-\mathscr{R}_\infty)[(1+2x^2)^{1/2}-1]\ , \qquad (2.30)$$

and has the initial condition $\vec{p}_i(\vec{R})=\vec{p}_i$ at $t=-\infty$. The line integral in (2.28) is taken along the path of the classical motion. $U_P(\vec{R})$ is the relativistic generalization of the ponderomotive potential and becomes (2.18) in the weak-field limit. The largest error associated with this solution is the order of $[p_i(R)L]^{-1}$ and is sufficiently small to be negligible for macroscopic beams. Clearly our description of the initial state is that of a hydrogen atom which changes its size as it moves into the laser beam. Its "Bohr radius" is given by η^{-1}, (2.28c), which shows that the atom compresses under the influence of the laser.

The semiclassical motion of the center of mass, in (2.28), may be carried further to a completely classical description. Again this is justified when the spatial distribution of the laser is slowly varying on the scale of the proton's wavelength. In that case we again start from (2.20) and simply drop the kinetic energy operator T_R and treat $\vec{R}(t)$ as a prescribed function of t which is obtained from the classical Hamiltonian (2.29). The function U_1 in (2.28) is than replaced by

$$U_1=u_0(r,\eta)\times\exp\left[-i\left[\int_{-\infty}^{t}dt'U_P(\vec{R}(t'))+W_0t\right]\right]\ . \qquad (2.31)$$

In summary, our picture of the initial state is that of the atom moving into the relativistic generalization of the ponderomotive potential of the laser which is essentially the second term of H_C, (2.29), while the size of the atom is shrinking, as described by (2.28b) and (2.28c). The electron in the atom has acquired an effective mass given by

$$m_{\rm eff}=m(1+2x^2)^{1/2}\ .$$

We now form the Dirac spinor for this state by

$$\chi_i=C_i\begin{bmatrix}1\\ (E+m-V)^{-1}\vec{\sigma}\cdot\vec{\pi}\end{bmatrix}U_1\ . \qquad (2.32)$$

The normalization constant obtained from the condition $(\chi_i,\chi_i)=1$ is fairly complicated but it can be simplified by the use of the approximations outlined above to obtain

$$C_i=\frac{1}{\sqrt{2}}\frac{[1+(1+2x^2)^{1/2}]}{[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos2\phi_1)]^{1/2}}\ ,$$
$$\phi_1=\omega t-\vec{k}\cdot\vec{R}\ . \qquad (2.33)$$

Having obtained the "distorted wave" description of the initial state we may now write the exact S matrix as

$$S_{\vec{q},0}=-i\langle\psi_f^{(-)},H_i\chi_i\rangle\ , \qquad (2.34)$$

where the perturbation operator H_i is defined by

$$\left[H-i\frac{\partial}{\partial t}\right]\chi_i=H_i\chi_i\ , \qquad (2.35)$$

where H is the full Hamiltonian occurring in (2.20) (with $T_R=0$). The use of (2.26b) with $E=i(\partial/\partial t)$, the neglect of V compared to m and ω compared to m, results in the four spinor

$$H_i\chi_i=\frac{1}{\sqrt{2}}[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos2\phi_1)]^{-1/2}\begin{bmatrix}4x\cos\varphi\hat{a}\cdot\vec{p}+2mx^2\cos2\varphi\\ 0\end{bmatrix}U_1\ , \qquad (2.36)$$

where ϕ is given in (2.22), U_1 in (2.31), and ϕ_1 in (2.33).

III. RELATIVISTIC CALCULATION OF THE S MATRIX

The approximation which we shall use to calculate the S matrix, (2.34), is that of replacing the exact final state by one in which the electron-proton interaction is neglected. This is exactly the same approximation as has been made in all previous calculations except that we treat the electron relativistically and the laser beam as an electromagnetic wave which has a finite spatial extent in the dimension perpendicular to the direction of propagation. The proton motion is described classically and its coupling to the laser field in the final state is neglected since it is m/M smaller than it was in the initial state. The proton therefore moves with unaccelerated motion. The electron is described by a modified Volkov state, the modification resulting from the slowly varying nature of the laser amplitude. The details of this state are given in the Appendix but it is immediately clear from (2.36) that only the "large components" of the final state enter into our calculation. The electron coordinate in the final state must be transformed into the same coordinates as those in the initial state and then the S matrix takes the form

$$S_{\vec{q},0}=-\frac{i}{\sqrt{2}}\int dt\, d^3r C_q[A_{fi}^{\dagger}(\vec{q})+B_{fi}^{\dagger}(\vec{q})\cos\phi]e^{-i\int^{\vec{R}+\vec{r}} d\vec{x}\,'\cdot f(x')+iE_q t}e^{i[a_1\sin\phi+(b/2)\sin2\phi]}$$
$$\times\frac{(4x\cos\phi\hat{a}\cdot\vec{p}+2mx^2\cos2\phi)}{[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos2\phi_1)]^{1/2}}u_0(r,\eta)e^{-iW_0t-i\int_{-\infty}^{t}dt'U_P(R(t'))}, \tag{3.1}$$

where

$$a_1=\frac{2mx}{\omega\nu}\hat{a}\cdot\vec{f}(\vec{R}+\vec{r})\simeq\frac{2mx}{\omega\nu}\hat{a}\cdot\vec{f}(\vec{R}), \tag{3.2a}$$

$$b=\frac{m^2x^2}{\omega\nu}, \tag{3.2b}$$

$$\nu=E_q-\hat{k}\cdot\vec{f}(\vec{R}), \tag{3.2c}$$

and where A_{fi} and B_{fi} are the matrix elements of the Dirac matrices of the Volkov state between initial and final spin states. It is convenient to express ϕ_1 as $\phi_1=\phi+\vec{k}\cdot\vec{r}$ and then use ϕ as the integration variable instead of t. It is also useful to define the functions F_l^n by

$$\frac{e^{ia_1\sin\phi+i(b/2)\sin2\varphi}\cos^n\phi}{[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos2\varphi_1)]^{1/2}}=\sum_{l=-\infty}^{\infty}\frac{e^{-il\phi}F_l^n}{(1+2x^2)^{1/4}[1+(1+2x^2)^{1/2}]^{1/2}}, \tag{3.3}$$

which allows us to write

$$\frac{e^{ia_1\sin\phi+i(b/2)\sin2\phi}\cos^n\phi\cos2\phi}{[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos2\phi_1)]^{1/2}}=\frac{1}{2}\sum_{l=-\infty}^{\infty}\frac{e^{-il\phi}(F_{l-2}^n+F_{l+2}^n)}{(1+2x^2)^{1/2}[1+(1+2x^2)^{1/2}]^{1/2}}. \tag{3.3a}$$

It is also convenient to exploit the slowly varying nature of $\vec{f}(\vec{x})$, (A21), and U_p to expand the exponential factors

$$\begin{aligned}\int^{\vec{R}+\vec{r}}d\vec{x}\,'\cdot\vec{f}(\vec{x}\,')&\simeq\int^{\vec{R}}d\vec{x}\,'\cdot\vec{f}(\vec{x}\,')+\vec{f}(\vec{R})\cdot\vec{r}+\cdots,\\ \int_{-\infty}^{t}dt'U_P(\vec{R}(t'))&\simeq\int_{-\infty}^{(\phi/\omega)+\hat{k}\cdot\vec{R}}dt'U_P(\vec{R}(t'))+U_P(\vec{R})\hat{k}\cdot\vec{r}+\cdots.\end{aligned} \tag{3.4}$$

Then (3.1) can be rewritten as

$$S_{\vec{q},0}=-\frac{i}{\sqrt{2}}\sum_l\int\frac{d\varphi}{\omega}\int\frac{d^3rC_q\exp i\left[-\int^{\vec{R}}d\vec{x}\,'\cdot\vec{f}(\vec{x}\,')-\vec{f}(\vec{R})\cdot\vec{r}+(E_q-W_0)/\omega(\phi+\vec{k}\cdot\vec{r})-l\phi\right]}{(1+2x^2)^{1/4}[1+(1+2x^2)^{1/2}]^{1/2}}$$
$$\times\{A_{fi}^{\dagger}[4x\hat{a}\cdot\vec{p}F_l^{(1)}+mx^2(F_{l-2}^{(0)}+F_{l+2}^{(0)})]+B_{fi}^{\dagger}[4x\hat{a}\cdot\vec{p}F_l^{(2)}+mx^2(F_{l-2}^{(1)}+F_{l+2}^{(1)})]\}$$
$$\times u_0(r,\eta)\exp-i\left[\int_{-\infty}^{(\phi/\omega)+\hat{k}\cdot\vec{R}}dt'U_P(\vec{R}(t'))+\hat{k}\cdot\vec{r}U_P(\vec{R})\right]. \tag{3.5}$$

The function F_l^n are slowly varying functions of $\vec{k}\cdot\vec{r}$ and this dependence may be neglected since $(k/\eta) << 1$. In effect this replaces ϕ_1 by ϕ in (3.3) and (3.3a). A similar remark can be made for the last exponential factor of (3.5) and then the $\vec{r}$ integration can be performed as a Fourier transform of the initial state

$$\int d^3r\, e^{-i\vec{f}\cdot\vec{r}+i(E_q-W_0)/\omega\,\vec{k}\cdot\vec{r}-i\hat{k}\cdot\vec{r}U_P(\vec{R})}u_0(r,\eta)$$

$$=\tilde{u}_0\{\vec{f}-\hat{k}[E_q-W_0-U_P(\vec{R})],\eta\}$$

$$=8\sqrt{\pi}\eta^{5/2}\{[f-\hat{k}(E_q-W_0-U_p)]^2+\eta^2\}^{-2}\ , \tag{3.6}$$

and the momentum operator $\vec{p}$, can be replaced by $\vec{f}(\vec{R})$ since $\hat{a}\cdot\vec{k}=0$. The remaing ϕ integral can be performed as

$$\int_{-\infty}^{\infty}\frac{d\phi}{\omega}\exp i\left[\frac{\phi}{\omega}(E_q-W_0-l\omega)-\int_{-\infty}^{(\phi/\omega)+\hat{k}\cdot\vec{R}}dt'U_P(\vec{R}(t'))\right]\simeq 2\pi\delta(E_q-W_0-l\omega-U_P(\vec{R}_0))\ , \tag{3.7}$$

where we have dropped an irrelevant phase factor. In obtaining (3.7) we have again exploited the slowly varying nature of $\vec{a}(\vec{R})$ or $U_P(\vec{R})$ and have labeled as $\vec{R}_0$ the point in the laser beam at which the ionization takes place. The observed cross section will contain an ensemble average of this parameter in effect allowing for the ionization to occur at any point within the laser beam. This energy delta function taken with (2.30) is the relativistic generalization of (2.19).

We may then rewrite the S matrix as

$$S_{\vec{q},0}=-2\pi i\sum_l\delta(E_q-W_0-l\omega-U_P(\vec{R}_0))T_{\vec{q},0}(l)\ , \tag{3.8}$$

where the T matrix for the absorption of l photons is

$$T_{q,0}=\frac{1}{\sqrt{2}}C_q\{A_{fi}^{+}[4x\hat{a}\cdot\vec{f}F_l^{(1)}+mx^2(F_{l-2}^{(0)}+F_{l+2}^{(0)})]$$

$$+B_{fi}^{+}[4x\hat{a}\cdot\vec{f}F_l^{(2)}+mx^2(F_{l-2}^{(1)}+F_{l+2}^{(1)})]\}\frac{\tilde{u}_0(\vec{f}-\vec{k}l,\eta)}{(1+2x^2)^{1/4}[1+(1+2x^2)^{1/2}]^{1/2}}\ . \tag{3.9}$$

The function defined in (3.3) can be written [see below (3.5)]

$$F_l^n=\int_0^{2\pi}\frac{d\phi}{2\pi}\cos^n\varphi\, e^{i\Phi(l)}\frac{[1+2x^2+(1+2x^2)^{1/2}]^{1/2}}{[1+x^2+(1+2x^2)^{1/2}+x^2(1+\cos 2\phi)]^{1/2}}\ , \tag{3.10}$$

where

$$\Phi(l)=l\phi+a_1\sin\phi+\frac{b}{2}\sin 2\phi\ . \tag{3.10$'$}$$

We shall see that the parameters a_1, b, and l are all very large such that

$$\frac{a_1}{b}=\frac{2\hat{a}\cdot\vec{f}}{mx}\simeq\frac{\alpha_F}{x}<<1\ ,$$
$$\frac{l}{b}\simeq\frac{\mathcal{R}_\infty}{m}\simeq\alpha_F^2<<<1\ . \tag{3.11}$$

This allows a stationary phase evaluation of (3.10) and one finds that the stationary phase point, $\partial\Phi/\partial\phi=0$, yields four such points given by $\cos\phi=\lambda_{\pm}$ where

$$\lambda_{\pm}=-\frac{a_1}{4b}\pm\left[\frac{1}{2}-\frac{l}{2b}+\left[\frac{a_1}{4b}\right]^2\right]^{1/2}\ . \tag{3.12}$$

The use of (3.11) in (3.12) shows that the stationary phase points are approximately $\pm\pi/4$, $\pm 3\pi/4$, all of which satisfy $\cos 2\phi=0$. Thus the denominator in the integral of (3.10) can be removed from the integral and one obtains

$$F_l^{(n)} \simeq \int_0^{2\pi} \frac{d\phi}{2\pi} \cos^n\phi\, e^{i\Phi(l)} . \tag{3.10''}$$

The function for $n=0$, gives a "generalized Bessel" function which has been previously encountered in a similar context[9] and recently discussed extensively.[10] We shall not need the results of that discussion since the stationary phase evaluation can be used in our case. The T matrix can be simplified by the use of the recurrence relations

$$\begin{aligned} &4x\hat{a}\cdot\vec{\mathrm{f}}F_l^{(1)} + mx^2(F_{l+2}^{(0)} + F_{l-2}^{(0)}) = 2l\omega\nu/mF_l^{(0)} ,\\ &4x\hat{a}\cdot\vec{\mathrm{f}}F_l^{(2)} + mx^2(F_{l+2}^{(1)} + F_{l-2}^{(1)}) = 2l\omega\nu/mF_l^{(1)} + \frac{\omega\nu}{m}(F_{l+1}^{(0)} - F_{l-1}^{(0)}) , \end{aligned} \tag{3.13}$$

which results in

$$T_{\vec{\mathrm{q}}0} = \frac{C_q}{\sqrt{2}(1+2x^2)^{1/4}[1+(1+2x^2)^{1/2}]^{1/2}} \left[A_{fi}^{+} \frac{2l\omega\nu}{m} F_l^{(0)} + B_{fi}^{+}\left[\frac{2l\omega\nu}{m}F_l^{(1)} + \frac{\omega\nu}{m}(F_{l+1}^{(0)} - F_{l-1}^{(0)})\right]\right] \tilde{u}_0(\vec{\mathrm{f}} - l\vec{\mathrm{k}}, \eta) . \tag{3.14}$$

The stationary phase evaluation of these functions is straightforward and the result is

$$F_l^{(0)} = \left[\frac{\omega\nu}{\pi m^2 x^2}\right]^{1/2} \left[\cos\left[\Phi_+(l) + \frac{\pi}{4}\right] + \cos\left[\Phi_-(l) - \frac{\pi}{4}\right]\right] , \tag{3.15}$$

$$F_l^{(1)} = \left[\frac{\omega\nu}{2\pi m^2 x^2}\right]^{1/2} \left[\cos\left[\Phi_+(l) + \frac{\pi}{4}\right] - \cos\left[\Phi_-(l) - \frac{\pi}{4}\right]\right] . \tag{3.16}$$

Here $\Phi_\pm(l)$ are the result of evaluating $\Phi(l)$, (3.10a), at the stationary phase points $\cos\phi = \lambda\pm$. These are rather complicated functions of l, a, and b which we shall not need. It is, however, clear that the $F_l^{(1)}$ term in the coefficient B_{fi}^{+} in (3.14) will dominate the $F_{l\pm1}^{(0)}$ terms since the factor l will be very large.

The matrix terms are given (A16) and (A17) by

$$A_{fi}^{+}(q) = \left[E_q + m - \frac{m^2x^2}{\nu}\right]\delta_{fi} , \tag{3.17a}$$

$$B_{fi}^{+}(q) = \frac{mx}{\nu}[\vec{\mathrm{f}}\cdot\hat{a} + i\vec{\sigma}\cdot\hat{k}\,\vec{\mathrm{f}}\cdot\hat{a}\times\hat{k} + i\vec{\sigma}\cdot\hat{k}\times\hat{a}(\vec{\mathrm{f}}\cdot\hat{k} - E_q - m)]_{fi} , \tag{3.17b}$$

so that we can write

$$T_{\vec{\mathrm{q}},0}(l) = T_{\vec{\mathrm{q}},0}^{(0)}(l) + i\vec{\sigma}_{fi}\cdot\hat{k}\times\hat{a}T_{\vec{\mathrm{q}},0}^{(1)}(l) + i\vec{\sigma}_{fi}\cdot\hat{k}T_{\vec{\mathrm{q}},0}^{(2)}(l) , \tag{3.18}$$

where the matrix dependence has been made explicit. We may now obtain the spin-flip and nonspin-flip T matrices but these will depend upon the axis of quantization. For example, if we choose the $\hat{k}$ direction as that axis then the nonspin-flip T matrix is given by

$$T_{\vec{\mathrm{q}},0}^{(0)}(l) \pm iT_{\vec{\mathrm{q}},0}^{(2)}(l) \tag{3.19}$$

with different signs for two possible initial spin directions. The spin-flip T matrix is

$$\pm T_{\vec{\mathrm{q}},0}^{(1)}(l) . \tag{3.20}$$

Since these results are axis dependent we will not pursue this further but instead find the total squared T matrix. The spin-flip and nonspin-flip amplitudes add incoherently so the result, averaging over the initial spin and summing over the final spin directions, yields

$$\overline{|T_{\vec{q},0}(l)|^2}=2[|T^{(0)}_{\vec{q},0}(l)|^2+|T^{(1)}_{\vec{q},0}(l)|^2+|T^{(2)}_{\vec{q},0}(l)|^2], \tag{3.21}$$

which is independent of the spin quantization axis. This can be written

$$\overline{|T_{\vec{q},0}(l)|^2}=\frac{C_q^2|\tilde{u}_0(\vec{f}-l\vec{k},\eta)|^2}{(1+2x^2)^{1/2}[1+(1+2x^2)^{1/2}]}\Bigg[(2l\omega x)^2(F_l^{(1)})^2[f^2+(E_q+m)^2-2\hat{k}\cdot f(E_q+m)] + \left[\frac{2l\omega v}{m}\right]^2(F_l^{(0)})^2\left[E_q+m-\frac{m^2x^2}{v}\right]^2 + 8l^2\omega^2\frac{v}{m}x\hat{a}\cdot\vec{f}F_l^{(0)}F_l^{(1)}\left[E_q+m-\frac{m^2x^2}{v}\right]\Bigg]. \tag{3.22}$$

The transition rate for l photon ionization is then

$$w_l=2\pi\int\frac{d^3q}{(2\pi)^3}\delta(E_q-W_0-l\omega-U_P(R_0))\overline{|T_{q,0}(l)|^2}. \tag{3.23}$$

The functions $F_l^{(n)}$ obtained in (3.15) and (3.16) depend upon $\Phi_\pm(l)$, (3.10′), which are very rapidly varying functions of $\vec{q}$. So that terms such as

$$\cos^2[\Phi_\pm(l)\pm(\pi/4)]$$

average to $\frac{1}{2}$ and the term

$$\cos[\Phi_+(l)+(\pi/4)]\cos[\Phi_-(l)-(\pi/4)]$$

can be shown to average to zero. The result is then

$$w_l=2\pi\int\frac{d^3q}{(2\pi)^3}\delta(E_q-W_0-l\omega-U_P(\vec{R}_0))\frac{C_q^2|\tilde{u}_0(\vec{f}-l\vec{k},\eta)|^2}{(1+2x^2)^{1/2}[1+(1+2x^2)^{1/2}]}\left[\frac{2l^2\omega^3 v}{\pi m^2}\right] \times\left[f^2+(E_q+m)^2-2\hat{k}\cdot\vec{f}(E_q+m)+\frac{2v^2}{m^2x^2}\left[E_q+m-\frac{m^2x^2}{v}\right]^2\right]. \tag{3.24}$$

As we have shown in the Appendix, $\vec{f}(R_0)$ is the momentum of the electron resulting from an l photon ionization at $\vec{R}_0$, which evolves as a classical electron governed by the Hamiltonian

$$H(f,r)=\{p^2+m^2[1+2x^2(\vec{r})]\}^{1/2}, \tag{3.25}$$

which becomes $\vec{q}$ as it emerges from the laser beam. Energy convervation for this motion yields

$$E_q=(q^2+m^2)^{1/2} =\{f^2+m^2[1+2x^2(\vec{R}_0)]\}^{1/2}, \tag{3.26}$$

and the fact that the laser beam is assumed to be uniform in the $\hat{k}$ direction yields

$$q_{\parallel}=f_{\parallel}, \tag{3.27}$$

where the subscript indicates the component in the $\hat{k}$ direction. If we set $\vec{f}=0$ in (3.25) and use the value of E_q obtained from the energy delta function in (3.24) we obtain a minimum value of l given by

$$\omega l_{\min}=\mathscr{R}_\infty(1+2x^2)^{1/2}, \tag{3.27}$$

so that we may intepret $\mathscr{R}_\infty(1+2x^2)^{1/2}$ as the binding energy of the atom in the field of the laser. This is consistent with the remarks made below (2.31). The restriction $\vec{f}=0$ coupled with (3.25) shows that the minimum value of $q^2=2m^2x^2$, which is the result of the explusion of the electron from the laser by the ponderomotive potential.

The integral in (3.24) may conveniently be done in cylindrical coordinates in the $\vec{q}$ variable with $\hat{k}$ as the cylindrical axis. There is no remaining dependence upon the azimuthal angle of $\vec{q}$ in (3.24) so that integral can be done immediately. The energy delta function can be used to perform the $q_\perp$ integral and (3.25) and (3.26) can be used to relate $\vec{f}$ to $\vec{q}$. There is a maximum value of $q_{\parallel}$

implied by the delta function which is

$$q_{\parallel}^2 \le Q^2 = E^2(l) - m^2(1+2x^2) , \qquad (3.28)$$

where

$$E(l) = W_0 + l\omega + U_P(\vec{R}_0)$$
$$= W_0(1+2x^2)^{1/2} + l\omega . \qquad (3.29)$$

The final integral becomes

$$w_l = \frac{4}{\pi l^2 \omega a_0^5 m^4} \frac{(1+2x^2)^2}{x^2[1+(1+2x^2)^{1/2}]} \times \int_{-Q}^{Q} \frac{dq_{\parallel}}{[E(l)-q_{\parallel}]^2} \times \{[E(l)-q_{\parallel}][E(l)+m] - m^2x^2\} , \qquad (3.30)$$

where we have neglected $l\omega << m$ which is consistent with our evaluation of the functions F_l^n. For large values of l the stationary phase evaluation fails but more general techniques yield the result that F_l^n falls off very rapidly with increasing l. This is very similar to the behavior of the Bessel functions. The integral in (3.30) is easily performed and the result may be simplified by keeping only the dominant terms;

$$w_l = \frac{8}{\pi} \frac{m\alpha_F^5}{l^2\omega} \sqrt{2m} \frac{(1+2x^2)^{9/4}}{x^2[1+(1+2x^2)^{1/2}]} \times (l\omega - l_{\min}\omega)^{1/2} \left[1 + \frac{m}{E(l)} - \frac{m^2x^2}{E(l)^2} \right] . \qquad (3.31)$$

The total transition rate can be obtained by summing over all l, and although (3.31) grossly over estimates the high l contribution, the contribution in (3.31) from that region is still negligible. The result is

$$w = \sum_{l=l_{\min}}^{\infty} w_l \simeq 8 \frac{\mathscr{R}_\infty}{\hbar} \alpha_F^2 \frac{(1+2x^2)[1+(1+2x^2)^{1/2}]}{x^2} , \qquad (3.32)$$

which we see is a rising function of x, (2.27), or the field strength. It is proportional to the laser intensity to the $\frac{1}{2}$ power. The result arises essentially from the compression of the initial bound state in the laser field which is a relativistic effect. A nonrelativistic calculation would yield a value of w which decreases with increasing field strength.

In summary, we have shown that previous calculations of ionization by ultraintense laser fields all neglected the finite spatial extent of the laser beam and the relativistic behavior of the electron. The first omission gave the wrong energy distribution of the electrons in that the effect of the ponderomotive potential was omitted and the second gave the wrong behavior of the transition rate as a function of very high laser intensity, decreasing instead of increasing.

APPENDIX

Volkov[4] has obtained the wave function of a Dirac electron in an infinite traveling wave electromagnetic field. We must generalize his results to a beam which is finite in the direction perpendicular to the propagation direction. The vector potential describing the field is

$$\vec{A}(\vec{\xi},t) = \vec{a}(\vec{\rho})\cos\phi , \qquad (A1)$$

where

$$\phi = \omega t - \vec{k}\cdot\xi, \quad \vec{\nabla}\times\vec{a} = \vec{k}\times\vec{a} = 0 , \qquad (A2)$$

and $\vec{\rho}$ is the part of $\vec{\xi}$ perpendicular to $\vec{k}$. Our solution is obtained in the limit where $\vec{a}(\vec{\rho})$ is slowly varying on the scale of the laser wavelength and the electron wavelength.

The Dirac equation is

$$\left[i\frac{\partial}{\partial t} - (\vec{\alpha}\cdot\vec{\pi} + \beta m) \right] \psi = 0 , \qquad (A3)$$

where

$$\vec{\pi} = \vec{p}_\xi + eA .$$

We set

$$\psi = \left[i\frac{\partial}{\partial t} + \vec{\alpha}\cdot\vec{\pi} + \beta m \right] Z \qquad (A4)$$

and obtain

$$\left[\frac{\partial^2}{\partial t^2} + \pi^2 + m^2 + e\vec{\sigma}\cdot\vec{H} + ie\vec{\alpha}\cdot\vec{E} \right] Z = 0 , \qquad (A5)$$

where

$$\vec{H} = \vec{\nabla}\times\vec{A} \simeq \vec{k}\times\vec{a}\sin\phi ,$$
$$\vec{E} = -\frac{\partial\vec{A}}{\partial t} = \omega\vec{a}\sin\phi . \qquad (A6)$$

The ansatz

$$Z=C_q e^{i\left[\int^{\xi} d\vec{\xi}'\cdot\vec{f}(\xi')-E_q t-S(\xi,\phi)\right]} \tag{A7}$$

with the assumption

$$\left[S,\frac{\partial S}{\partial\varphi}\right]=0 \tag{A8}$$

results in

$$-2\omega(E_q-\hat{k}\cdot\vec{f})\frac{\partial S}{\partial\varphi}+e\vec{a}\cdot\vec{f}\cos\varphi+\frac{e^2a^2}{2}\cos2\varphi+e\sin\varphi(\vec{\sigma}\cdot\vec{k}x\vec{a}+i\omega\vec{\alpha}\cdot\vec{a})=0\ , \tag{A9}$$

where we have dropped terms of order ∇a and used

$$E_q^2=q^2+m^2=f^2(\xi)+m^2[1+2x^2(\xi)]\ . \tag{A10}$$

This may be integrated to give

$$S=\frac{e\vec{a}\cdot\vec{f}}{\omega\nu}\sin\varphi+\frac{e^2a^2}{8\omega\nu}\sin2\varphi-\frac{ea}{2\nu}\delta\cos\varphi\ , \tag{A11}$$

where

$$\nu=E_q-\hat{k}\cdot\vec{f} \tag{A12}$$

and

$$\delta=\vec{\sigma}\cdot\hat{k}\times\hat{a}+i\vec{\alpha}\cdot\hat{a}\ , \tag{A13}$$

which satisfies

$$\delta^2=0\ . \tag{A14}$$

The fact that δ is the only matrix occurring in S and that it is independent of ϕ means that (A8) is satisfied. S can be used to obtain Z, (A7), which can then be substituted back into (A4) to obtain

$$\psi=C_q(A_q+B_q\cos\phi)\exp i\left[\int^{\xi}d\vec{\xi}'\cdot\vec{f}(\xi')-E_qt-a_1\sin\varphi-\frac{b}{2}\sin2\varphi\right]\ , \tag{A15}$$

where

$$A_q=E_q+\beta m-\frac{mx^2}{\nu}(1+\vec{\alpha}\cdot\hat{k})+\vec{\alpha}\cdot\vec{f}(\xi)\ , \tag{A16}$$

$$B_q=\frac{mx}{\nu}[\ \vec{f}\cdot\hat{a}+i\vec{\sigma}\cdot\hat{k}\times\hat{a}(E_q+m\beta-\vec{f}\cdot\hat{k})+i\vec{\sigma}\cdot\hat{k}\vec{f}\cdot\hat{k}\times\hat{a}$$
$$+(i/3)\vec{\alpha}\cdot\vec{\sigma}\vec{f}\cdot\hat{k}\times\hat{a}-\vec{\alpha}\cdot\hat{a}(f\cdot\hat{k}-\beta m-E_q)+\vec{f}\cdot\hat{a}\vec{\alpha}\cdot\hat{k}]\ , \tag{A17}$$

and

$$a_1=\frac{2mx}{\omega\nu}\hat{a}\cdot\vec{f}\ , \tag{A18}$$

$$b=\frac{m^2x^2}{\omega\nu}\ , \tag{A19}$$

where we have used the definition

$$x=\frac{e\,|a(\xi)|}{2m}\ . \tag{A20}$$

The momentum $\vec{f}$ must be the classical momentum whose time development is governed by the Hamiltonian

$$H(\vec{p},\vec{\xi})=\{p^2+m^2[1+2x^2(\xi)]\}^{1/2} \tag{A21}$$

and becomes $\vec{q}$ for large t. This restriction on $\vec{f}$ has been discussed by Weinberg[8] in another context. We will not pursue the discussion here since the details of $\vec{f}$ other than the energy conservation relation implied by (A21) are not germane to our calculation.

Finally we must also obtain the normalization constant C_q. We do this by working in a finite volume of linear dimension D. Then if the cross section of the laser beam has a dimension L, the condition

$$(\psi_q,\psi_q)=1 \tag{A22}$$

yields

$$C_q^{-2}=2E_q(E_q+m)D^3\left[1+O\left[\frac{L}{D}\right]\right], \tag{A23}$$

and we shall assume that $L/D<<1$.

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