Free-Free Transitions in Intense High-Frequency Laser Fields

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A nonperturbative theory is developed for free-free transitions of electrons colliding with atoms in intense high-frequency laser fields. The energy of the incoming electrons is assumed to be smaller than that of the photons. Rather simple expressions are derived for the multiphoton absorption amplitudes. Large deviations from the laser-free case are shown to appear in elastic scattering. The theory applies at already existing laser frequencies but extends beyond, to the extreme ultraviolet range.

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Studies of multiphoton free-free transitions of electron-atom systems in intense laser fields have been done predominantly at low frequencies, stimulated by the theory of Kroll and Watson.[1,2] However, lasers of increasingly high frequencies have become available, some of them operated at high intensities.[3] No theory exists so far for this case. We shall present here a method for treating free-free transitions specifically adapted for the high-frequency, high-intensity regime. The method is nonperturbative with respect to the coupling of the electron to the radiation field.

A fully realistic description of the target atom, taking into account its internal degrees of freedom, is prohibitively difficult. We shall represent the atom by a potential model, the merits and limitations of which are well understood.[4] Besides, it is a logical starting point towards a more complete solution of the problem. The potential will be taken to be of the central self-consistent type: Coulomb-like at the origin [$V(r) \simeq -Z/r$], short range or ionic [$V(r) \simeq -Z'/r$] at large distances, but unspecified otherwise.

The laser field will be represented by a monochromatic plane wave, linearly polarized, in the dipole approximation. The plane-wave assumption is not critical, as the extension to a laser pulse can subsequently be made.[5] Linear polarization is assumed in view of simplifying the algebra, and the dipole approximation is justified in the frequency range we are interested in (from the visible to the extreme ultraviolet). Consequently, we take the electrodynamic potentials of the wave in the form $\vec{A} = \vec{a}\cos\omega t$ (with $\vec{a}$ real) and $\varphi = 0$.[6]

Application of the space translation transformation (Kramers and Henneberger[7]) to the Schrödinger equation gives[8]

$$[\tfrac{1}{2}\vec{p}^{\,2} + V(\vec{r} + \vec{\alpha}(t))]\psi = i(\partial\psi/\partial t), \tag{1}$$

where[9]

$$\vec{\alpha}(t) = -c^{-1}\int_0^t \vec{A}(t')dt' = \vec{\alpha}_0 \sin\omega t,$$
$$\vec{\alpha}_0 = -\vec{a}/\omega c. \tag{2}$$

This should be solved by imposing the boundary conditions of our problem: an incoming current of particles of energy $E = p^2/2$, and radially outgoing currents of scattered particles of energies and momenta

$$E_n = E + n\omega,\ E_n = p_n{}^2/2,\ n = 0, \pm 1, \pm 2, \ldots . \tag{3}$$

Equation (1) has periodic time-dependent coefficients. As usual, we seek a quasiperiodic solution of the form

$$\psi(\vec{r}, t) = e^{-iEt}\sum_{n=-\infty}^{+\infty} \psi_n(\vec{r})e^{-in\omega t}. \tag{4}$$

Then, we Fourier analyze the potential:

$$V(\vec{r} + \vec{\alpha}(t)) = \sum_{n=-\infty}^{+\infty} V_n(\vec{\alpha}_0; \vec{r})e^{-in\omega t}. \tag{5}$$

By some algebraic manipulations the coefficients can be written as

$$V_n(\vec{\alpha}_0; \vec{r}) = (i^n/\pi)\int_{-1}^{+1} V(\vec{r} + \vec{\alpha}_0 u)T_n(u)(1-u^2)^{-1/2}du, \tag{6}$$

where $T_n(u)$ are Chebyshev polynomials.

Insertion of Eqs. (4) and (5) into Eq. (1) leads to a system of coupled differential equations for the components $\psi_n(\vec{r})$, which we write

$$[\tfrac{1}{2}\vec{p}^{\,2} + V_0 - (E + n\omega)]\psi_n = -\sum_{\substack{m=-\infty \\ (m\neq n)}}^{+\infty} V_{n-m}\psi_m. \tag{7}$$

The boundary conditions require that our solutions $\psi_n(\vec{\alpha}_0, \omega; \vec{r})$ behave asymptotically as follows:

$$\psi_0(\vec{\alpha}_0, \omega; \vec{r}) \to \exp\{i[\vec{p}\vec{r} + \gamma_0 \ln(pr - \vec{p}\vec{r})]\} + f_0(\vec{\alpha}_0, \omega; \hat{r})\exp[i(pr - \gamma_0 \ln 2pr)]/r, \tag{8}$$

$$\psi_n(\vec{\alpha}_0, \omega; \vec{r}) \to f_n(\vec{\alpha}_0, \omega; \hat{r})\exp[i(p_n r - \gamma_n \ln 2p_n r)]/r \quad (n \neq 0), \tag{9}$$

with $\gamma_n = -Z'/p_n$ (for a short-range potential $Z' = 0$). Equation (8) contains the elastic scattering amplitude $f_0(\vec{\alpha}_0, \omega; \hat{r})$, and Eq. (9) that for absorption/emission $f_n(\vec{\alpha}_0, \omega; \hat{r})$. The associated scattering cross sections are[10]

$$d\sigma_n/d\Omega = (p_n/p)|f_n(\vec{\alpha}_0, \omega; \hat{r})|^2 \quad (n = 0, \pm 1, \pm 2, \ldots). \tag{10}$$

We shall now describe a method for handling the system Eq. (7). The left-hand side contains the Hamiltonian

$$H = \tfrac{1}{2}P^2 + V_0(\vec{\alpha}_0; \vec{r}). \tag{11}$$

By use of the Green's operator $G(\Omega)$ associated to it, where Ω is the energy parameter, Eq. (7) may be formally solved as

$$\psi_n = \psi_{\vec{p}}^{(+)}\delta_{n0} - G^{(+)}(E_n)\sum_{\substack{m \\ (m \neq n)}} V_{n-m}\psi_m . \tag{12}$$

Here $\psi_{\vec{p}}^{(+)}$ is the ($\vec{\alpha}_0$-dependent) solution of the equation[11]

$$H\psi_{\vec{p}} = E\psi_{\vec{p}}, \tag{13}$$

satisfying the boundary condition Eq. (8) with an amplitude $f_0^{(0)}(\vec{\alpha}_0; \vec{r})$. It then follows from Eq. (12) that the ψ_n satisfy the boundary condition required by Eqs. (8) and (9) with the following expression for the scattering amplitudes:

$$f_n(\vec{\alpha}_0, \omega; \hat{r}) = f_0^{(0)}(\vec{\alpha}_0; \hat{r})\delta_{n0} - (1-\delta_{n0})\frac{1}{2\pi}\langle \psi_{\vec{p}_n}^{(-)}| V_n |\psi_{\vec{p}}^{(+)}\rangle + \frac{1}{2\pi}\sum_{\substack{m \\ (m\neq n)}}\sum_{\substack{m' \\ (m'\neq m)}}\langle \psi_{\vec{p}_n}^{(-)}| V_{n-m}G^{+}(E_m)V_{m-m'}|\psi_{m'}\rangle . \tag{14}$$

Besides $\psi_{\vec{p}}^{(+)}$, Eq. (14) also contains $\psi_{\vec{p}_n}^{(-)}$, which is an incoming-wave solution of Eq. (13), as well as the unknown set of components $\psi_{m'}(\boldsymbol{r})$ satisfying Eq. (12).

By repeated insertion of Eq. (12) into Eq. (14) an expansion can be derived for f_n. Obviously, the iteration will have practical significance only if the successive terms decrease sufficiently rapidly. Since this will not be true in general, it is important to establish the conditions under which the first nonvanishing term of Eq. (14) represents a good approximation. For (a) $\omega \gg |E_0(\boldsymbol{\alpha}_0)|$, where $E_0(\boldsymbol{\alpha}_0)$ is the ground-state energy of the Hamiltonian Eq. (11)[12]; (b) $\alpha_0^2\omega \gg 1$; (c) $\omega \gg E$, it was possible to extract the *exact* form of the dominant contribution to the last term of Eq. (14) (denoted below by $f^{(1)}$) for an *arbitrary* potential of the type discussed before. In the case of elastic scattering we find

$$\begin{pmatrix} \mathrm{Re}f_0^{(1)}(\hat{r}) \\ \mathrm{Im}f_0^{(1)}(\hat{r}) \end{pmatrix} = \frac{Z^2}{6\boldsymbol{\alpha}_0\omega^2}\left[\psi_{\vec{p}_0}^{(-)*}(\vec{\alpha}_0)\psi_{\vec{p}}^{(+)}(\vec{\alpha}_0) + \psi_{\vec{p}_0}^{(-)*}(-\vec{\alpha}_0)\psi_{\vec{p}}^{(+)}(-\vec{\alpha}_0)\right]\begin{pmatrix} -(\ln\alpha_0^2\omega)^2 + O(\ln\alpha_0^2\omega) \\ \pi\ln\alpha_0^2\omega + O((\alpha_0^2\omega)^0) \end{pmatrix}, \tag{15}$$

where $\vec{p}_0 = p\hat{r}$ is the *final* momentum [see Eq. (3) for $n = 0$], and the corrective terms O also depend on α_0, E, θ. Thus at given α_0,[13] and sufficiently high ω [obeying conditions (a), (b), and (c) and the dipole-approximation assumption] it is possible to satisfy the inequality (d) $|f_0^{(1)}(\boldsymbol{\alpha}_0, \omega; E, \theta)| \ll |f_0^{(0)}(\boldsymbol{\alpha}_0; E, \theta)|$. Whereas this holds in general over wide ranges of parameter values, it should nevertheless be checked for each case separately because $f_0^{(0)}$ may become exceptionally small for certain angles (e.g., see Fig. 2). Conditions (a)–(d) together ensure the dominance of the first term in Eq. (14). However, this may hold under wider conditions than we were able to prove.

Thus, the elastic amplitude f_0 reduces, to lowest order (in the sense discussed above), to $f_0^{(0)}$, which is that calculated from the time-independent Schrödinger equation Eq. (13). This shows that in the high-frequency, high-intensity regime the incoming electron feels only the static distorted potential $V_0(\vec{\alpha}_0; \vec{r})$, the "dressed" potential associated to $V(r)$. From Eq. (7) it follows that $V_0(\vec{\alpha}_0; \vec{r})$ is simply the potential created by a linear "charge" distribution of density $\pi^{-1}(1-u^2)^{-1/2}$ extending from $-\vec{\alpha}_0(u=1)$ to $\vec{\alpha}(u=-1)$, the potential generated by the unit of "charge" being $V(r)$. This behavior appears natural due to the rapid oscillations of the center of force in Eq. (1).[11]

For the absorption amplitude ($n > 0$) we get to lowest order from Eq. (14)

$$f_n(\vec{\alpha}_0, \omega; \hat{r}) = -\frac{1}{2\pi}\langle \psi_{\vec{p}_n}^{(-)}| V_n |\psi_{\vec{p}}^{(+)}\rangle . \tag{16}$$

Thus, the Fourier component V_n acts in our regime as a transition operator between scattering states $\psi_{\vec{q}}^{(+)}$ of the dressed potential V_0. The amplitude f_n depends on ω via the final momentum $\vec{p}_n$ [see Eq. (3)]. Because ω was assumed to be large at given α_0,[13] all f_n will be small with respect to f_0. (This contrasts with the low-frequency case where many f_n may be larger than f_0.) Note that the condition (c) above precludes free-free emission ($n < 0$).

Since the original potential $V(r)$ is spherically symmetric, $V_0(\vec{\alpha}_0;\vec{r})$ and all $V_n(\vec{\alpha}_0;\vec{r})$ have axial symmetry around $\vec{\alpha}_0$ (we have assumed linear polarization). For example, in the case of pure Coulomb potential $V(r)=-Z/r$, Eq. (6) yields

$$V_0(\vec{\alpha}_0;\vec{r})=-(2Z/\pi)(r_+r_-)^{-1/2}K(2^{-1/2}(1-\hat{r}_+\hat{r}_-)^{1/2}), \tag{17}$$

where $\vec{r}_\pm=\vec{r}\pm\vec{\alpha}_0$ and K is a complete elliptic integral of the first kind. V_0 has a logarithmic singularity along the distribution of charges, and $r_\pm^{-1/2}$ singularities at its end points (all weaker than the original Coulomb singularity). The dressed Coulomb potential Eq. (17) is represented graphically in Fig. 1.

The axial symmetry of the elastic scattering problem Eq. (13) makes it resemble the electron–diatomic-molecule and the proton–deformed-nucleus scattering (both in the static approximation with the internal degrees of freedom neglected). The powerful computer programs developed for these cases can be adapted to our needs.[14] However, for the time being, one may get an idea of the magnitude of the effects involved by considering a simplified problem, in which the dressed potential $V_0(\vec{\alpha}_0;\vec{r})$ is simulated by its spherical average $\bar{V}_0(r)$.[15]

We shall illustrate the simplified version on the elastic scattering from a pure *Coulomb potential.* In this case, from Eq. (17),

$$\bar{V}_0(r)=\begin{cases}-(Z/\pi\alpha_0\rho)\left[2\arcsin\rho-\rho\ln\dfrac{1-[1-\rho^2]^{1/2}}{1+[1-\rho^2]^{1/2}}\right], & \rho\le 1,\\ -(Z/\alpha_0\rho), & \rho\ge 1,\end{cases} \tag{18}$$

where $\rho=r/\alpha_0$. Since $\bar{V}_0(r)$ can be regarded as a Coulomb potential modified by a short-range distortion, the scattering amplitude can be calculated by the two-potential formalism.[16] The total, modified amplitude $\bar{f}_0(\hat{r})$ is then the sum of the Coulomb amplitude $f^c(\hat{r})$ and of a short-range contribution $f'(\hat{r})$, which can be obtained from a phase-shift calculation. In Fig. 2 we give the ra-

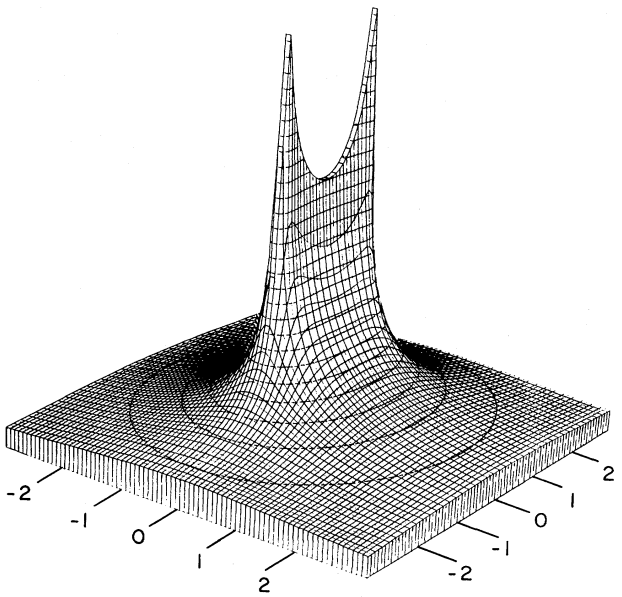

FIG. 1. Values of the dressed Coulomb potential V_0 in a plane passing through the symmetry axis defined by $\vec{\alpha}_0$. In this plane, taken as the horizontal base of the figure, distances are measured in units of α_0. Along the vertical we represent $(-\alpha_0V_0/Z)$ in atomic units, up to the value 10. The saddle of the figure reflects the rise in V_0 near the line of singularities (extending from $-\vec{\alpha}_0$ to $\vec{\alpha}_0$) and their increasing strength towards the end points. At radial distances larger than α_0 in the horizontal plane, the distortion of the Coulomb potential fades away (the level lines become circular).

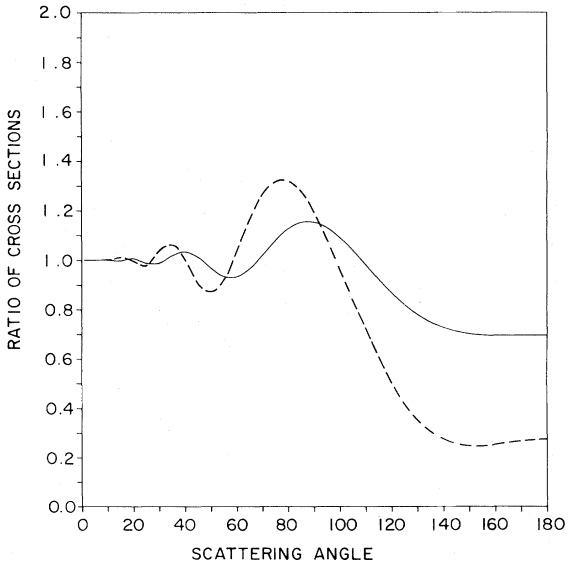

FIG. 2. Ratio of differential cross section for elastic scattering from the *averaged* dressed Coulomb potential Eq. (18), to the Rutherford cross section. Nuclear charge $Z=1$; electron energy $E=0.05$ Ry; $\alpha_0=1$, solid line, and $\alpha_0=3$, dashed line [for the definition of α_0 see Eq. (2) and Ref. 9].

tio of the modified to the original (Rutherford) cross sections for two cases within the validity of our theory, and accessible experimentally.[9] It is apparent that the modified cross section (for $\bar{V}_0$) can differ considerably from that of the laser-free case.[17]

We conclude that electron-atom free-free transitions in the intense, high-frequency laser fields now becoming available for experiments reveal new features, contrasting with those known at low frequencies.[1,2] A detailed numerical investigation is in progress (see Ref. 14). We are applying similar methods to multiphoton ionization.

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[1]N. M. Kroll and K. M. Watson, Phys. Rev. A 8, 804 (1973).

[2]For a background of the problem, see M. J. Mittleman, *Theory of Laser-Atom Interactions* (Plenum, New York, 1982); L. Rosenberg, Adv. At. Mol. Phys. 18, 1 (1982); M. Gavrila and M. van der Wiel, Comments At. Mol. Phys. 8, 1 (1978).

[3]T. S. Luk, H. Pummer, K. Boyer, M. Shahidi, H. Egger, and C. K. Rhodes, Phys. Rev. Lett. 51, 110 (1983).

[4]For the case of radiation-free scattering, see Ch. Joachain, *Quantum Collisions Theory* (North-Holland, Amsterdam, 1975), Chap. 20. With the laser on, the extra difficulties stemming from the decay of the atom by multiphoton ionization are attenuated in our regime, since in the limit of high frequencies the ionization is quenched.

[5]H. Krüger and Ch. Jung, Phys. Rev. A 17, 1706 (1978).

[6]Note that our premises are the same as those of Kroll and Watson, Ref. 1.

[7]H. A. Kramers, *Collected Scientific Papers* (North-Holland, Amsterdam, 1956), p. 262; W. C. Henneberger, Phys. Rev. Lett. 21, 838 (1968).

[8]We are using atomic units throughout.

[9]α_0 can be expressed in atomic units as $\alpha_0 = (I/I_0)^{1/2}\omega^{-2}$, where I is the time-averaged beam intensity and $I_0 = 3.51 \times 10^{16}$ W/cm^2. The values of α_0 of present interest lie somewhere between 0 and 100. For $\omega = 0.23$ a.u. and $I \simeq 10^{15}$ W/cm^2, which correspond to the conditions of Ref. 3 (under somewhat stronger focusing), $\alpha_0 \simeq 3.1$.

[10]For a laser pulse with slowly varying amplitude on the atomic scale, Eq. (10) should be time averaged appropriately; see Ref. 5.

[11]The energy eigenvalue equation Eqs. (11) and (13) was considered before for bound states in a high-frequency laser field by J. I. Gersten and M. H. Mittleman, J. Phys. B 9, 2561 (1976).

[12]It follows from Eq. (6) that by *increasing* α_0, V_0 becomes shallower, and therefore $|E_0(\alpha_0)|$ *decreases* in general from its unperturbed value at $\alpha_0 = 0$. This trend is confirmed by the calculation of Gersten and Mittleman, Ref. 11, Fig. 1.

[13]The low-frequency result of Kroll and Watson is derived under the same constraint (see Ref. 1, and also Mittleman, Ref. 2, Sec. 6.4).

[14]A computation along these lines for several model potentials is now being carried out in collaboration with Dr. J. van de Ree.

[15]This should give the right order of magnitude for elastic scattering, especially when the de Broglie wavelength of the electron is larger than the extension of the "charge" distribution ($p\alpha_0 \lesssim 1$). On the other hand, interesting features related to the dependence on the direction of the polarization vector will be washed out.

[16]See the book of Joachain, Ref. 2, Chap. 17.

[17]The ratio is equal to 1 for vanishing θ because f^c tends to infinity, whereas f' stays finite.