RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 36, NUMBER 1 JULY 1, 1987

Two-color ionization with an intense low-frequency field

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(Received 23 March 1987)

We formulate a gauge-invariant approximation for treating the ionization of a one-electron atom by two lasers, one intense and of low frequency, the other weak and of high frequency. We have applied the approximation to hydrogen and obtained an intensity-dependent structure in the photoelectron angular distributions.

In this paper we develop an approximation for treating the ionization of a one-electron atom by two lasers, one a weak laser of relatively high frequency ω_H, and the other an intense laser of low frequency ω_L. During the course of the analysis we clarify the important role played by the gauge of the radiation field. We have applied our approximation to the two-color ionization of hydrogen, and we present results which reveal intensity-dependent structure in the photoelectron angular distributions.

We consider an electron initially bound by a spherically symmetric potential W in a state represented by $|\phi_i(t)\rangle = |\phi_i\rangle\exp(-iE_it/\hbar)$. Denoting the interactions of the electron with the high- and low-frequency lasers by $V_H(t)$ and $V_L(t)$, respectively, the full Hamiltonian for the electron is

$$H(t)=T+W+V_H(t)+V_L(t) \ , \tag{1}$$

where $T\equiv\mathbf{p}^2/2\mu$ is the kinetic energy operator, with μ the electron mass. The evolution of the state vector $|\Psi(t)\rangle$ of the electron is governed by the time-development operator $U(t,t')$. Assuming that the lasers are turned on at time $t=0$ we have[1] $|\Psi(t)\rangle=U(t,0)|\phi_i\rangle$, where $U(t,t)=1$ and

$$i\hbar dU(t,t')/dt=H(t)U(t,t') \ . \tag{2}$$

We introduce $U_L(t,t')$, the time-development operator which would be appropriate if $V_H(t)$ were turned off:

$$i\hbar dU_L(t,t')/dt=[T+W+V_L(t)]U_L(t,t') \ . \tag{3}$$

Using the integral equation[2]

$$U(t,t')=U_L(t,t') - (i/\hbar)\int_{t'}^{t}dt''[U_L(t'',t)]^{\dagger}V_H(t'')U(t'',t') \ , \tag{4}$$

we have, putting $t'=0$ and $t=T\sim\infty$,

$$|\Psi(T)\rangle=U_L(T,0)|\phi_i\rangle - (i/\hbar)\int_0^T dt[U_L(t,T)]^{\dagger}V_H(t)|\Psi(t)\rangle \ . \tag{5}$$

At the time T the amplitude $A_{fi}(T)\equiv\langle\phi_f(T)|\Psi(T)\rangle$, for the electron to be found moving freely through the field in the continuum state characterized by the mean mechanical momentum $\hbar\mathbf{k}_f$ and represented by $|\phi_f(t)\rangle$, is, using Eq. (5),

$$A_{fi}(T)=\langle\chi_f(0)|\phi_i\rangle-(i/\hbar)\int_0^T dt\langle\chi_f(t)|V_H(t)|\Psi(t)\rangle \ , \tag{6}$$

where $|\chi_f(t)\rangle$ is the state vector for the electron to scatter from W in the presence of the low-frequency laser and emerge in the state represented by $|\phi_f(t)\rangle$. We have

$$|\chi_f(t)\rangle=\lim_{T\to\infty}U_L(t,T)|\phi_f(T)\rangle \ . \tag{7}$$

We describe the electric fields of the two lasers classically by the spatially independent vectors (assuming linear polarization)

$$\mathbf{F}_H(t)=\mathbf{F}_H\cos(\omega_Ht), \quad \mathbf{F}_L(t)=\mathbf{F}_L\cos(\omega_Lt).$$

We assume that $(F_L/\omega_L)\gg(F_H/\omega_H)$. Immediately after the atom ionizes, the electron emerges with a mechanical momentum which oscillates owing to the oscillation of the electron in the low-frequency field. The mean mechanical momentum $\hbar\mathbf{k}_f$ of the emergent electron differs from its instantaneous mechanical momentum by $\hbar\mathbf{k}(t)=(e/c)\mathbf{A}_L(t)$, where e is the electron charge and $\mathbf{A}_L(t)$ is the vector potential for the low-frequency field: $\mathbf{A}_L(t)=-(c/\omega_L)\mathbf{F}_L\sin(\omega_Lt)$.

In the Kroll-Watson low-frequency approximation[3,4] we replace $|\chi_f(t)\rangle$ by

$$|\chi^{\mathrm{KW}}(t)\rangle=e^{-i\xi(t)}|\psi^{-}_{\mathbf{K}(t)}\rangle \ , \tag{8a}$$

where $|\psi^{-}_{\mathbf{K}(t)}\rangle$ is the wave vector (satisfying outgoing-wave boundary conditions) for an electron to scatter from the potential W, in the *absence* of radiation, and emerge with momentum

$$\hbar\mathbf{K}(t)=\hbar[\mathbf{k}_f-\mathbf{k}(t)] \ , \tag{8b}$$

which becomes the instantaneous mechanical momentum in the presence of the field. The phase $\xi(t)$ in Eq. (8a) is defined as

$$\xi(t)=\frac{\hbar}{2\mu}\int_0^t dt'K^2(t') \ , \tag{8c}$$

with $K(t)=|\mathbf{K}(t)|$. It is important to note that in writing down Eq. (8a) we have assumed the length gauge for the low-frequency field; in this gauge we have

$$V_L(t)=-e\mathbf{x}\cdot\mathbf{F}_L(t) \ , \tag{9a}$$

where $\mathbf{x}$ is the electron coordinate. In the velocity gauge, the interaction has the form

$$V_L(t)=-(e/\mu c)\mathbf{p}\cdot\mathbf{A}_L(t)+e^2A_L^2(t)/2\mu c^2 \ . \tag{9b}$$

The wave function in the velocity gauge differs from the wave function in the length gauge by[5] the highly significant phase factor $\beta_L(t)=\exp[i\mathbf{k}(t)\cdot\mathbf{x}]$, assuming that

$V_H(t)$ is expressed in the length gauge.

We make the further approximation of replacing $|\Psi(t)\rangle$ on the right side of Eq. (6) by its unperturbed form $|\phi_i(t)\rangle$. As far as the weak high-frequency field is concerned, this approximation is justified. The coupling strength of the low-frequency field to the initial atomic state is, in the length gauge, $\alpha_1 \equiv (eaF_L/\Delta E_i)$, where a is the characteristic radius of the initial state, and ΔE_i is the distance of the nearest atomic energy level from the initial one. We therefore require $\alpha_1 \ll 1$. In the velocity gauge the coupling strength is $(evF_L/\omega_L \Delta E_i)$, where v is the characteristic speed of the electron in the initial state. This latter parameter is *not* small at low frequencies ω_L. Indeed, this parameter is effectively the exponent of $\beta_L(t)$. Hence if we use the velocity gauge we must approximate $|\Psi(t)\rangle$ by $\beta_L(t)|\phi_i(t)\rangle$, rather than by $|\phi_i(t)\rangle$; we must also multiply the right-hand side of Eq. (8a) by $\beta_L(t)$ since, as noted already, Eq. (8a) was presented in the length gauge.

Note that, in the length gauge, $|\chi_f^{\rm KW}(t)\rangle$ is an eigenvector of $(T+W)$ with positive eigenvalue $\hbar^2 K^2(t)/2\mu$ and is therefore orthogonal to $|\phi_i(t)\rangle$, which is also an eigenvector of $(T+W)$ but with negative eigenvalue E_i. This orthogonality is preserved in the velocity gauge since $\beta_L^*(t)\beta_L(t)=1$. A gratifying consequence is that, in either gauge, the approximate ionization amplitude $A_{fi}(T)$ is independent of any spatially independent function of t which we may add to the Hamiltonian $H(t)$. This is required on physical grounds, and indeed any such function can be transformed out of the Hamiltonian and absorbed as an insignificant phase factor into $|\Psi(t)\rangle$. One may recall an analogy in Wick's observation[6] that the integrated cross section for the excitation of an atom by a heavy particle should not change if we add to the Hamiltonian any function that depends only on the coordinate connecting the centers of mass of the two collision partners. We note further that $A_{fi}(T)$ is, up to a correction of order (ω_L/ω_H), independent of the gauge chosen for $V_H(t)$.[7]

Some remarks on the validity of the Kroll-Watson form of the low-frequency approximation to the scattering wave vector are in order. A necessary condition is that the phase of the low-frequency field does not change significantly during the time the electron scatters from W. Since the collision duration is roughly $\hbar/E_f$, with E_f the final mean electron energy, we require $\alpha_2 \equiv \hbar\omega_L/E_f \ll 1$. Numerical calculations[8] indicate that the Kroll-Watson approximation is remarkably accurate. However, the approximation breaks down even if $\alpha_2 \ll 1$ when E_f differs from the discrete energy of an atomic bound or resonance state by $n\hbar\omega_L$, with n an integer, since the electron can then be captured by W for a rather long time, during which the field phase changes. Therefore, we cannot apply our approximation to the two-color experiment recently performed by Muller, van Linden van den Heuvell, and van der Wiel.[9]

We now derive an expression for the ionization rate $\dot{P}_{fi}$ to a group of states f with density $\rho'(E_f)$ in the energy interval (E_f, E_f+dE). We have

$$\dot{P}_{fi} = \rho'(E_f)dE \frac{d}{dT}|A_{fi}(T)|^2 . \tag{10}$$

We simplify this expression using the length gauge. Note that we have assumed that one high-frequency photon is sufficient to ionize the atom. If N_0 high-frequency photons were required to ionize, we would approximate $|\Psi(t)\rangle$ through (N_0-1)th order in $V_H(t)$. We make the rotating-wave approximation and thereby replace $V_H(t)$ by $-(e/2)(\mathbf{x}\cdot\mathbf{F}_H)\exp(-i\omega_H t)$. Evaluating the integral of Eq. (8c), ignoring an insignificant constant, and defining $E_f = \hbar^2 k_f^2/2\mu$, we have

$$\xi(t) = (E_f + P)t/\hbar + \zeta(t), \tag{11a}$$

$$P = e^2 F_L^2/(4\mu\omega_L^2) , \tag{11b}$$

$$\zeta(t) = -(e\mathbf{k}_f\cdot\mathbf{F}_L/\mu\omega_L^2)\cos(\omega_L t) - (P/2\hbar\omega_L)\sin(2\omega_L t) . \tag{11c}$$

It follows that

$$\langle \chi^{\rm KW}(t)|V_H(t)|\phi_i(t)\rangle = e^{i(E_f+P-\hbar\omega_H-E_i)t/\hbar}M(t) , \tag{12}$$

where $M(t)$ is periodic in t with period $2\pi/\omega_L$ and is given as

$$M(t) = -e^{-i\zeta(t)}\langle \psi_{\bar{\mathbf{K}}(t)}|(e/2)(\mathbf{x}\cdot\mathbf{F}_H)|\phi_i\rangle . \tag{13}$$

We Fourier-decompose $M(t)$ as

$$M(t) = \sum_n M_n e^{-in\omega_L t} , \tag{14}$$

and substitute the right-hand side of Eq. (12) into the integrand of Eq. (6), noting that $\langle \chi_f(0)|\phi_i\rangle = 0$, to obtain[10] for $T\sim\infty$

$$A_{fi}(T) = -i\pi\sum_n \delta(E_f - E_{fn})M_n , \tag{15}$$

where $E_{fn} = n\hbar\omega_L + \hbar\omega_H + E_i - P$. The δ function expresses energy conservation: The amount of energy required to ionize the atom is $P-E_i$, where P is the ponderomotive shift in the continuum threshold. So, after absorbing one high-frequency photon and n low-frequency photons, the electron emerges, still in the laser fields, with energy E_{fn}. Note that E_f is the final mean energy of the electron relative to the *shifted* threshold. We have neglected the Stark shift of the initial atomic level. Note also that the levels E_{fn} are not lifetime broadened since we have approximated $|\Psi(t)\rangle$ through (N_0-1)th order in $V_H(t)$ and have therefore neglected corrections arising from the depopulation of the initial state. We have treated $V_L(t)$ to all orders, but we have assumed that the low-frequency laser does not depopulate the initial state; it is the high-frequency laser which causes ionization. Were we to include corrections beyond (N_0-1)th order in $V_H(t)$, the gauge-transform phase factor for $F_H(t)$ would be significant, and Eq. (6) would hold only if $V_H(t)$ were expressed in the velocity gauge; only in this gauge does $|\chi_f(t)\rangle$ represent at asymptotically large distances an electron unperturbed by $V_H(t)$, i.e., an electron whose canonical momentum is unchanged by $V_H(t)$. Following Kroll and Watson,[3] we can evaluate M_n approximately by noting that if $|e\mathbf{k}_f\cdot\mathbf{F}_L/\mu\omega_L^2| \gg 1$, the integrand of

$$M_n = \frac{\omega_L}{2\pi}\int_0^{2\pi/\omega_L} dt\, e^{in\omega_L t}M(t) \tag{16}$$

oscillates rapidly over the interval $2\pi/\omega_L$ because of the phase $\zeta(t)$. The main contribution to M_n comes from t near the point of stationary phase t_0. Assuming that

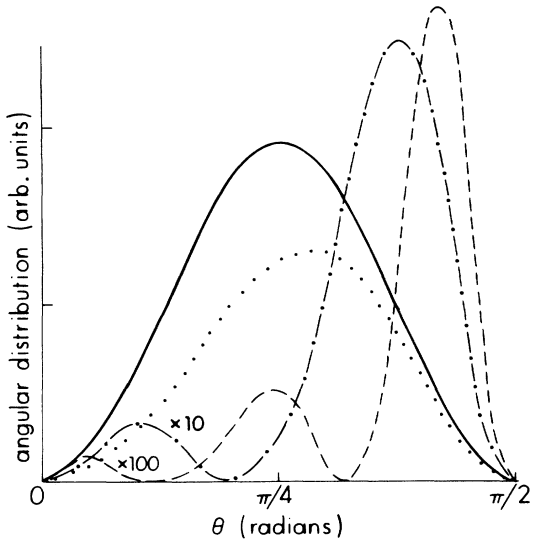

FIG. 1. Photoelectron angular distributions in the plane defined by the two laser polarizations, which are assumed to be perpendicular, with θ the angle between the photoelectron momentum and the CO_2 laser polarization. These curves are for the channel where one low-frequency photon is absorbed. In the limit where the CO_2 laser intensity I_L is weak, the photoelectron carries away two units of angular momentum and the angular distribution has the form $\sin^2\theta\cos^2\theta$, obtained when $V_L(t)$ is retained only in lowest order. Significant departures from this form are seen above about $I_L = 3\times10^7$ W/cm^2. The solid curve is the result obtained in lowest order in $V_L(t)$ for $I_L = 1.4\times10^8$ W/cm^2. The dotted curve is the "exact" result at the same intensity. The dot-dashed curve and the dashed curve are the "exact" results for $I_L = 9.0\times10^8$ and 2.3×10^9 W/cm^2, respectively, where the first peak of each of these curves is magnified by the factor indicated. Note that $d\dot{P}_{fn,i}/d\Omega$ nearly vanishes at the zeros of the Bessel function of Eq. (17) of the text. Note also that $\sum_n d\dot{P}_{fn,i}/d\Omega$ is approximately independent of I_L (and is approximately just the angular distribution for ionization by the high-frequency photon) so that $\dot{P}_{fn,i}$ diminishes as I_L increases —the ionization signal is shared between an increasing number of channels.

$\alpha_3 \equiv P/\hbar\omega_L$ is negligible, t_0 is given by $\mathbf{k}_f\cdot\mathbf{k}(t_0) = n\mu\omega_L/\hbar$. Putting $t = t_0$ in the more slowly varying part of the integrand we obtain

$$M_n = -J_n(e\mathbf{k}_f\cdot\mathbf{F}_L/\mu\omega_L^2)\langle\psi_{\bar{\mathbf{K}}(t_0)}|(e/2)(x\cdot\mathbf{F}_H)|\phi_i\rangle \ . \quad (17)$$

The factor multiplying the Bessel function is just the usual matrix element for an electron to be ejected with momentum $\hbar\mathbf{K}(t_0)$ by a single photon of energy $(\hbar^2/2\mu)K^2(t_0) - E_i = \hbar\omega_H$. If $|e\mathbf{k}_f\cdot\mathbf{F}_L/\mu\omega_L^2| \gg 1$, the electron can absorb a large amount of angular momentum through virtual absorption and emission, and the Bessel-function factor significantly effects the angular distribution even in the $n=0$ channel, where no real low-frequency photons are absorbed.

Observing that

$$dA_{fi}(T)/dT = -(i/\hbar)\sum_n \exp[i(E_f - E_{fn})T/\hbar]M_n \ ,$$

and using Eqs. (10) and (15), the rate for ionization to the channel where n low-frequency photons are absorbed is $\dot{P}_{fn,i} = (2\pi/\hbar)\rho'(E_{fn})|M_n|^2$. The total ionization rate is an incoherent sum of the partial ionization rates $\dot{P}_{fn,i}$; the sum is incoherent because interference terms arising from the absorption of different numbers of photons vanish when the signal is averaged over a macroscopically small volume.[3] We have applied our approximation to the calculation of the photoelectron angular distributions $d\dot{P}_{fn,i}/d\Omega$ for ionization of ground-state hydrogen by a CO_2 laser ($\hbar\omega_L \approx 4.4\times10^{-3}$ a.u.) and a high-frequency laser ($\hbar\omega_H \approx 0.65$ a.u.). The results, a sample of which is shown in Fig. 1, reveal interesting intensity-dependent structure in the angular distributions. Similar structure was reported earlier by Reiss[11] in the case of one-color ionization by a CO_2 laser. For very intense fields the angular distributions are rendered isotropic by ponderomotive scattering in the beam waist,[12] an effect we have ignored. Ponderomotive scattering is expected to be insignificant if $\alpha_4 = P/E_f \ll 1$. The parameters α_1, α_2, α_3, and α_4 are all small in the intensity region of Fig. 1.

Note added in proof. Other approximations for treating two-color ionization with an intense low-frequency field were recently brought to our attention. See E. Fiordilino and M. Mittleman, Phys. Rev. A **28**, 229 (1983), and references therein. However, these approximations are not gauge-invariant.

We thank Bill Cooke for helpful conversations. This work was supported by the National Science Foundation under Grant No. PHY-8512495.

[1]We assume that the lasers are turned on slow enough for the fields to be regarded as monochromatic, but fast enough so that the atom is not disturbed from its initial state during the turn-on period. We can assume that the turn-on time is large compared to an atomic orbital period, so that the atom responds adiabatically to the increasing intensity.

[2]See, e.g., L. Spruch, in *Lectures in Theoretical Physics,* edited by S. Geltman, K. T. Mahanthappa, and W. E. Britten (Gordon and Breach, New York, 1969), Vol. XIC, p. 77.

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

[4]L. Rosenberg, Adv. At. Mol. Phys. **18**, 1 (1982); H. Kruger and C. Jung, Phys. Rev. A **17**, 1706 (1978); M. H. Mittleman, *ibid.* **20**, 1965 (1979); R. Shakeshaft, *ibid.* **28**, 667 (1983).

[5]An interesting discussion on the gauge problem may be found in R. R. Schlicher, W. Becker, J. Bergou, and M. O. Scully, in *Quantum Electrodynamics and Quantum Optics,* edited by A. O. Barut (Plenum, New York, 1984), p. 405.

[6]See the footnote on p. 359 of J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).

[7]These considerations point to deficiencies in the type of approximation used, for example, in H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[8]R. Shakeshaft, Phys. Rev. A **29**, 383 (1984).

[9]H. G. Muller, H. B. van Linden van den Heuvell, and M. J. van der Wiel, J. Phys. B **19**, L733 (1986).

[10]We use $\int_0^\infty e^{i\Omega t}dt = iP(1/\Omega)+\pi\delta(\Omega)$ and neglect $P(1/\Omega)$, which vanishes when multiplied by the infinitesimally small increment dE.

[11]H. R. Reiss, Phys. Rev. A **22**, 1786 (1980).

[12]R. R. Freeman, T. J. McIlrath, P. H. Bucksbaum, and M. Bashkansky, Phys. Rev. Lett. **57**, 3156 (1986).