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Nonperturbative calculation of partial differential rates for multiphoton ionization of a hydrogen atom in a strong laser field

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We show how the multiphoton-ionization matrix element for an atom in a laser field at intensities beyond the regime of perturbation theory can be calculated from the standard Floquet expansion of the electron wave function. We expand the harmonic components in a discrete complex Sturmian basis set, and we evaluate the resulting (divergent) sum for the matrix element by using the Padé method. We present results for partial rates and angular distributions for hydrogen.

We describe here a method for calculating the multiphoton-ionization matrix element, within the Floquet approximation,[1] for an atom exposed to an intense laser field. As a demonstration, we present estimates of partial rates for multiphoton ionization of hydrogen at intensities beyond the regime of perturbation theory. Total rates for multiphoton ionization of hydrogen (that is, widths of the initial energy level) were calculated previously by Chu and Cooper.[2] To our knowledge the results reported here are the first converged nonperturbative estimates of *partial* rates for ionization of hydrogen[3] within the Floquet formalism. We note that converged nonperturbative estimates of partial rates for multiphoton detachment of the negative hydrogen ion were previously calculated[4] within a one-electron model in which the (active) electron moves in a Yukawa potential. However, the method used in that application is not readily adaptable to the case of a long-range Coulomb potential.

We treat the radiation field as classical, monochromatic (frequency ω), and spatially homogeneous over atomic dimensions. We ignore the interval during which the field is turned on, and we suppose that the intensity reaches its peak value at $t=0$ and is constant thereafter.[5] For $t>0$ the electron wave vector satisfies the Schrödinger equation:

$$[i\hbar d/dt - H_a - V(\tau)]\,|\Psi(t)\rangle = 0\,, \tag{1}$$

where H_a is the atomic Hamiltonian and where $V(\tau)$, the interaction of the electrons with the radiation field, is for $t>0$ periodic in $\tau=\omega t$ with period 2π. Within the Floquet approximation[1] the solution to Eq. (1) can be expressed as

$$|F_i(t)\rangle = e^{-i\varepsilon_i t/\hbar}\,|\psi_i(\tau)\rangle\,, \tag{2}$$

where ε_i is the quasienergy and where $|\psi_i(\tau)\rangle$ is periodic in τ. The form (2) is an exact solution of Eq. (1) when $t>0$, but it does not develop continuously in time from the initial bound-state wave vector $|\Phi_i\rangle$ which represents the state i of the electrons before the field is turned on; $V(\tau)$ is not periodic during the interval that the field is turned on. The electrons should be described at $t>0$ by a normalizable wave packet whose energy spread is at least the induced width Γ_i of the initial energy level. The Floquet form (2) amounts to assuming that the active electrons have a well-defined total energy ε_i; therefore, wave-packet localization (and wave-packet spreading) are neglected.[6] However, ε_i has a negative imaginary part, $-\Gamma_i/2$, accounting for the decay of the initial state. Consequently, the coordinate space representation of $|\psi_i(\tau)\rangle$, which is just a sum of Gamow-Siegert functions corresponding to the resonance eigenvalues ε_i, explodes exponentially at large distances.

We express $V(\tau)$ as $V(\tau)=V_+e^{-i\tau}+V_-e^{i\tau}$ and expand $|\psi_i(\tau)\rangle$ in the harmonic series

$$|\psi_i(\tau)\rangle = \sum_n |\psi_{in}\rangle e^{-in\tau}\,, \tag{3}$$

where the harmonic components satisfy the equations

$$(\varepsilon_i + n\hbar\omega - H_a)\,|\psi_{in}\rangle = V_+\,|\psi_{i,n-1}\rangle + V_-\,|\psi_{i,n+1}\rangle\,. \tag{4}$$

We can write $\varepsilon_i = E_i + \Delta_i - i\Gamma_i/2$, where Δ_i is the shift from the unperturbed bound-state eigenvalue E_i. The eigenvalue ε_i is determined from the condition that the set of homogeneous linear equations possesses a solution which (in coordinate space) is regular at the origin and which at infinity is a superposition of spherical outgoing (albeit exploding) waves. We note that this boundary condition can be imposed in the velocity gauge but not in the length gauge.[7,8] [In the latter gauge the interaction $V(\tau)$ explodes at infinity.]

We work in the velocity gauge; for $\tau>0$ we have $V(\tau) = -(e/\mu c)\mathrm{Re}(e^{-i\omega t}\mathbf{A}_0\cdot\mathbf{p})$, where e and μ are the electron charge and mass, $\mathbf{p}$ is the total canonical linear momentum of the electrons, and $\mathbf{A}_0$ is the (constant) vector potential amplitude. [We drop the term in $|\mathbf{A}_0|^2$ from $V(\tau)$ since this can be absorbed into an irrelevant phase factor multiplying $|F_i(t)\rangle$. Consequently, Δ_i includes a downward ponderomotive shift.] The matrix element for N-photon ionization may be written as $M_N = M_N^{(+)} + M_N^{(-)}$, with[7]

$$M_N^{(\pm)} = \langle \Phi_{\mathbf{k}_f}^- |\, V_\pm\,|\psi_{i,N\mp1}\rangle\,, \tag{5}$$

where $\hbar^2\mathbf{k}_f^2/2\mu = E_{iN} = E_i + \Delta_i + N\hbar\omega$ and where $|\Phi_{\mathbf{k}_f}^-\rangle$ is the wave vector, satisfying out-asymptote boundary conditions, for an electron to scatter from the atomic potential in the absence of radiation and energy with momentum $\hbar\mathbf{k}_f$. Since $|\psi_{in}\rangle$ explodes exponentially in coordinate space, the partial matrix elements $M_N^{(\pm)}$ are divergent.

However, wave packet spreading is negligible during the ionization process only[6] if $\Gamma_i \ll \hbar^2 k_f^2/2\mu$, and so it is consistent with the Floquet approximation to take the limit $\Gamma_i \to 0$ in $M_N^{(\pm)}$. Note that even in the limit $\Gamma_i \to 0$ the $M_N^{(\pm)}$ are divergent because of phase cancellation in the integrands arising from *virtual* photon absorption and emission. Fortunately, and in accord with physical intuition, the divergences cancel[7,8] in the sum $M_N^{(+)}+M_N^{(-)}$ to yield a finite M_N. To take this limit in practice we cannot simply put $\Gamma_i=0$ in Eq. (4) because ε_i is an eigenvalue and consequently a solution does not exist when we put $\Gamma_i=0$, except in the weak-field limit. One way around this is to introduce an inhomogeneous term into Eq. (4) which is consistent with neglecting Γ_i; this has been discussed elsewhere.[7] An alternative procedure is to solve the homogeneous Eqs. (4) and analytically continue the integral defining M_N; this is what we do here, though with no pretense of mathematical rigor.

To understand better how we analytically continue the divergent integral defining M_N, consider the simpler integral

$$\int_0^\infty r\,dr \sin(Kr)e^{ikr} = \frac{-2ikK}{(k^2-K^2)^2}\,, \tag{6}$$

where K is real but k may be complex. If $\mathrm{Im}(k)<0$ the integrand explodes exponentially at infinity but the right-hand side of Eq. (6) is the analytic continuation of the integral into the lower half complex k plane. [The integral has poles at $k=\pm K$ because of the phase cancellation in the integrand between $\sin(Kr)$ and e^{ikr}. A similar phase cancellation occurs in $M_N^{(\pm)}$, as noted above.] We now illustrate how we would analytically continue this integral were we not able to evaluate it analytically. We begin by expanding $r\exp(ikr)$ in terms of a discrete set of orthonormal basis functions.[9] Here we choose the Sturmian basis functions[10]

$$S_{nl}^{\kappa}(r) = A_{nl}(-i\kappa r)^{l+1}e^{i\kappa r}{}_1F_1(l+1-n,2l+2,-2i\kappa r)\,, \tag{7}$$

where the $S_{nl}^{\kappa}(r)$ are orthogonal with respect to the weight function $1/r$ and A_{nl} is chosen so that

$$\int_0^\infty dr\, S_{ml}^{\kappa}(r)(1/r)S_{nl}^{\kappa}(r) = \delta_{mn}\,. \tag{8}$$

We have (with $A_{n0}=2n^{1/2}$)

$$re^{ikr} = \sum_{n=1}^{\infty} a_n(k)S_{n0}^{\kappa}(r)\,, \tag{9a}$$

$$a_n(k) = \int_0^\infty dr\, S_{n0}^{\kappa}(r)e^{ikr} \tag{9b}$$

$$= \frac{-2i\kappa n^{1/2}}{\kappa^2-k^2}\left(\frac{k-\kappa}{k+\kappa}\right)^n\,. \tag{9c}$$

Provided that $\mathrm{Im}(\kappa+k)>0$ the integrand of Eq. (9b) decays exponentially and the integral for $a_n(k)$ can be evaluated by numerical quadrature; of course, this is unnecessary in the present example, where we have the exact result. The expansion of $r\exp(ikr)$ converges, in the sense that $a_n(k)$ vanishes for $n\sim\infty$, provided that $|\arg(k)-\arg(\kappa)|<\pi/2$. This result appears to be quite general;[11] that is, any suitable function which behaves as r^{l+1} at the origin and has outgoing wavelike behavior $\exp(ikr)$ at infinity can be expanded in terms of basis functions $S_{nl}^{(\kappa)}(r)$ provided that the previous inequality holds. If we substitute the expansion of $r\exp(ikr)$ into the integrand of Eq. (6), and interchange sum and integral, we obtain

$$\int_0^\infty r\,dr \sin(Kr)e^{ikr} = \sum_{n=1}^{\infty} a_n(k)b_n(K)\,, \tag{10a}$$

$$b_n(K) = \int_0^\infty dr \sin(Kr)S_{n0}^{\kappa}(r)\,. \tag{10b}$$

The coefficients $b_n(K)$ are the coefficients of the expansion of $r\sin(Kr)$ in terms of the $S_n^{\kappa}(r)$. We know that this latter expansion does not converge since $\sin(Kr)$ is a standing wave and we cannot have both $|\arg(\pm K)-\arg(\kappa)|<\pi/2$. Indeed, we have $2ib_n(K)=a_n(K)-a_n(-K)$, and $b_n(K)$ grows exponentially as n increases. The sum on the right-hand side of (10a) converges only if $a_n(k)$ diminishes more rapidly than $b_n(K)$ explodes as n increases, a condition which is not satisfied for the interesting range of k. However, the analytic continuation of the sum is provided by the sequence of diagonal Padé approximates converging (here in a finite number of steps) towards the right-hand side of Eq. (6).

The integral of Eq. (6) is similar to the integral defining M_N in that $|\Phi_{\mathbf{k}_f}^-\rangle$ is a sum of standing waves, like $\sin(Kr)$, and the harmonic components $|\psi_{i,N\pm1}\rangle$ are (in the velocity gauge) sums of outgoing waves like $\exp(ikr)$. Each harmonic component has closed channel parts, $\arg(k)=\pi/2+\theta_m$ if $m<N_0$, and exploding open channel parts, $\arg(k)=-\theta_m$ if $m\geq N_0$, where $2\theta_m=\tan^{-1}(\Gamma_i/2|E_{im}|)$ and where N_0 is the smallest number of photons that the atom must absorb to ionize.[12] We solve Eq. (4) by expressing the harmonic components in terms of basis functions $S_{nl}^{\kappa}(r)Y_{lm}(\hat{x})$, whose coefficients are determined from the linear homogeneous equations[1,2] which result from projecting Eq. (4), using Eq. (8), onto our basis set. We must choose κ to be in the upper-right quadrant of the κ plane and such that its argument does not differ from either $\pi/2+\theta_m$ or $-\theta_m$ by more than $\pi/2$; see Fig. 1. We thereby obtain convergent expansions of the harmonic components, and we can evaluate the divergent sum representing M_N by using the Padé method.[13] Note that because of the strong cancellation (of divergences) between $M_N^{(+)}$ and $M_N^{(-)}$, discussed above, the Padé method must be applied to the sum of $M_N^{(\pm)}$ and not to these individual matrix elements.

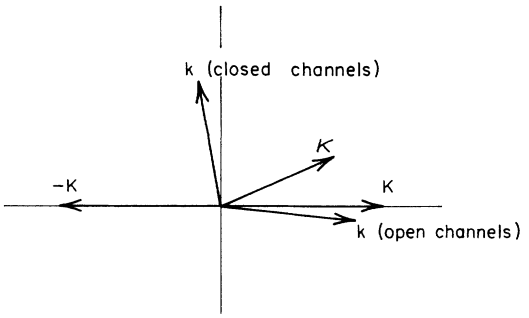

FIG. 1. Complex wave numbers k, K, and κ appearing in the integrand of ionization matrix element.

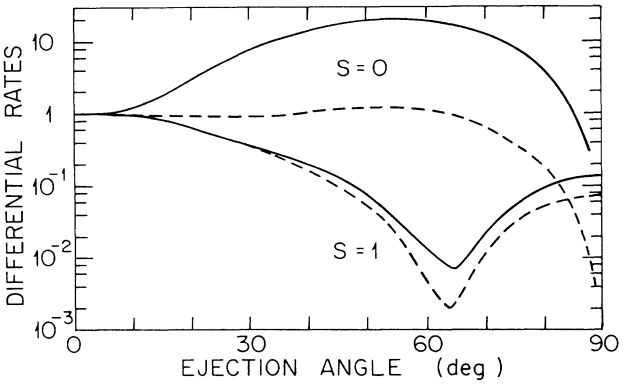

FIG. 2. Angular distributions, normalized to unity at zero angle, for (N_0+S)-photon ionization by linearly polarized light of wavelength 265 nm ($N_0=3$) and intensity 5×10^{13} W/cm^2. The solid and dashed curves are the results obtained from the present calculation and from perturbation theory (Ref. 15), respectively. Upper curves: $S=0$; lower curves: $S=1$. The angle of ejection is measured relative to the polarization axis.

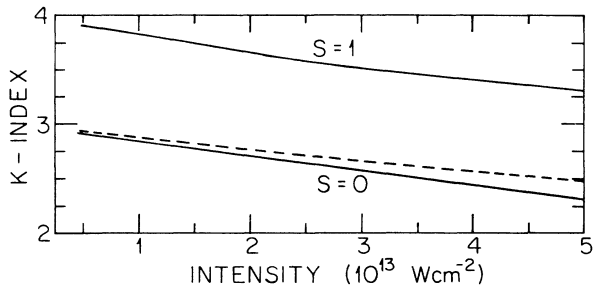

FIG. 3. K index of nonlinearity vs intensity for (N_0+S)-photon absorption by linearly polarized light of wavelength 265 nm ($N_0=3$). This index is defined as the derivative, with respect to the logarithm of the intensity, of the logarithm of the rate. Solid curves are for partial rates (integrated over angle) and dashed curve is for total rate (the width). In the limit of vanishing intensity the index becomes an integer (N_0+S for the partial rates, and N_0 for the total rate).

The partial differential rate for N-photon ionization to a group of states with density $\rho'(E_{iN})$ in the energy interval $(E_{iN}, E_{iN}+dE)$ is $\Gamma_{iN}=(2\pi/\hbar)\rho'(E_{iN})|M_N|^2$, provided that $|\psi_i(\tau)\rangle$ is normalized appropriately. The question of normalization has been discussed elsewhere[4,14] and we will postpone further discussion of this to a later paper. We note, however, that to the extent that $\Gamma_i \ll E_{iN_0}$, the partial rates, when integrated over angle, must sum to the total rate Γ_i. If this sum rule is significantly violated, the concept of a time-independent transition rate ceases to be meaningful. Hence the time-independent method used here is limited to intensities for which the sum rule is satisfied, i.e., $\Gamma_i \ll E_{iN_0}$. In the intensity region where a multiphoton ionization threshold is crossed ($E_{iN_0}=0$) time-dependent effects are important.[12]

In Figs. 2 and 3 we present some results which illustrate the departure from nonvanishing lowest-order perturbation theory at intensities not far below the lowest intensity (5.5×10^{13} W/cm^2) for which a multiphoton ionization threshold is crossed. We have normalized the results for the index of nonlinearity, in Fig. 3, so that the sum of partial rates equals with width Γ_i. In our calculations we retained harmonic components $|\psi_{in}\rangle$ with photon index n in the range $-3\le n\le 7$, each angular partial wave of $|\psi_{in}\rangle$ was expanded in 40 basis functions, with $l\le 7$, and we took $|\kappa|^2=0.4$ and $\arg(\kappa)=1.07$ rad. The results shown are stable with respect to changes in the ranges of the photon index and l, the number of basis functions, and the value of κ. Finally, we note that the present method is also useful in a weak-field perturbative treatment.

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[1]For a review of Floquet methods, see S.-I. Chu, Adv. At. Mol. Phys. **21**, 197 (1985).

[2]S.-I. Chu and J. Cooper, Phys. Rev. A **32**, 2769 (1985). See also A. Maquet, S.-I. Chu, and W. P. Reinhardt, *ibid.* **27**, 2946 (1983).

[3]We note, however, that quantum-defect theory has been used to obtain partial rates for multiphoton ionization from high Rydberg states in an intense field. See A. Giusti-Suzor and P. Zoller, Phys. Rev. A **36**, 5178 (1987).

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[5]The coupling of Floquet states due to temporal variations of the intensity can be important, particularly at frequencies near an intermediate resonance. The formalism presented below can be generalized to include this coupling.

[6]R. Shakeshaft, J. Opt. Soc. Am. B **4**, 705 (1987).

[7]R. M. Potvliege and R. Shakeshaft (unpublished).

[8]R. Shakeshaft, Z. Phys. D **8**, 47 (1988); M. Edwards and R. Shakeshaft, *ibid.* **8**, 51 (1988).

[9]Analytic continuation of an integral through basis set expansion of the integrand was discussed earlier by T. N. Rescigno and C. W. McCurdy, Phys. Rev. A **34**, 1882 (1986). For reviews of complex scaling, see B. R. Junker, Adv. At. Mol. Phys. **18**, 207 (1982); W. P. Reinhardt, Annu. Rev. Phys. Chem. **33**, 223 (1982).

[10]See, e.g., A. Maquet, Phys. Rev. A **15**, 1088 (1977).

[11]R. Shakeshaft, Phys. Rev. A **34**, 5119 (1986); **34**, 244 (1986).

[12]The relevant eigenvalue ε_i corresponds to a resonance pole of the resolvent operator for the atom plus field. There are also "shadow" poles, which are further from the physical energy axis. (See e.g., Ref. 6.) When an ionization threshold is crossed, the resonance pole interchanges roles with a shadow pole. This will be discussed in a future paper.

[13]The Padé method was first used to calculate the multiphoton ionization matrix element in perturbation theory by S. Klarsfeld and A. Maquet, Phys. Lett. **78A**, 40 (1980), and references therein.

[14]See also Rescigno and McCurdy, in Ref. 9, and references therein.

[15]Y. Gontier, N. K. Rahman, and M. Trahin, Phys. Rev. A **34**, 1112 (1986).