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Multiphoton absorption by alkali-metal atoms above the ionization threshold

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We describe a powerful method for computing cross sections for multiphoton ionization of atoms. The method is especially suited to absorption above threshold.

Multiphoton absorption above the ionization threshold is currently a topic under active investigation.[1] Most theoretical works (e.g., Refs. 2–6) have focused on hydrogen as the atomic target, with the various numerical methods relying on the analytic nature of the electron wave functions. An exception is the approach of Aymar and Crance[7] which has been successfully applied to alkali-metal atoms, a case of experimental interest. In this Rapid Communication we describe a powerful method for rather quickly obtaining reasonable estimates of cross sections for multiphoton ionization of atoms, with no restriction placed, in principle, on the number of photons absorbed above the ionization threshold. We report results for four-photon ionization of Cs, with two photons absorbed above threshold.

We treat the electron-laser interaction as a perturbation, and use the dipole approximation. The radial Green's function, $g_l^+(E)$, for the electron in the atomic field has outgoing wave character, while the radial wave function, $|u_{KL}\rangle$, of the emergent photoelectron consists of both outgoing and ingoing waves, $|u_{KL}^+\rangle$ and $|u_{KL}^-\rangle$, respectively. The transition matrix element for N-photon absorption can be decomposed into matrix elements of the form $M^{(N)}=M^{+(N)}+M^{-(N)}$, where

$$M^{\pm(N)}=(u_{KL}^{\pm}|rg_{l_{N-1}}^{+}(E_{N-1})\cdots rg_{l_1}^{+}(E_1)r|u_{nl}) \quad , \tag{1}$$

where $(a|b)=\int_0^\infty dr\, a(r)b(r)$, r is the radial coordinate operator, l_i is an intermediate angular momentum quantum number, E_i is an intermediate energy, and $|u_{nl}\rangle$ represents the initial state of the electron. These matrix elements are N-dimensional integrals over the radial coordinates $r_1, r_2, \ldots, r_N$. When some intermediate energies lie above threshold, that is, when $P<N$ where P is the minimum number of photons required to ionize the atom, these integrals are not formally convergent because of the behavior of the integrand for $r_i\sim\infty$. However, the integrand of $M^{+(N)}$ consists of functions that either decrease exponentially or behave as outgoing waves for $r_i\sim\infty$. Consequently, we can rotate the integration contour in $M^{+(N)}$ into the upper half complex r plane, that is, we let $r_i\rightarrow|r_i|e^{i\theta}$; provided that we choose $0<\theta<\pi/2$ the entire integrand decays exponentially, thereby permitting direct numerical integration. (As a bonus, this transformation produces an integrand which is less oscillatory, and which can therefore be integrated rapidly and with high precision.) The integrand of $M^{-(N)}$ contains functions that, for $P<N$, have both outgoing and ingoing wave character, precluding a rotation of the integration contour. However, we have shown previously[8] how this problem can be circumvented. Using the relationship

$$g_l^+(E)-g_l^-(E)=-2\pi i\delta(E-H_l) \quad , \tag{2}$$

where H_l is the radial Hamiltonian for the electron in the atomic field, we can express $M^{-(N)}$ in terms of $M^{+(N)}$. It follows[8] that

$$M^{(N)}=2\,\mathrm{Re}(M^{+(N)})-2\pi i\sum_{i=1}^{N-P}J_i^{(N)}M^{(N-i)} \quad , \tag{3a}$$

$$J_i^{(N)}=(u_{KL}^{+}|rg_{l_{N-1}}^{+}(E_{N-1})\cdots g_{l_{N-i+1}}^{+}(E_{N-i+1}) \times r|u_{k_{N-i},l_{N-i}})^* \quad , \tag{3b}$$

$$J_1^{(N)}=(u_{KL}^{+}|r|u_{k_{N-1},l_{N-1}})^* \quad , \tag{3c}$$

where $\hbar k_i=(2mE_i)^{1/2}$ with m the electron mass. The $|u_{kl}\rangle$ are normalized on the energy scale. Note that for $i=P+1,\ldots,N$ we have $k_i>k_{i-1}$. We see that the integrands of the integrals $[J_i^{(N)}]^*$ consist of outgoing waves only, and so once again we can rotate the integration contour into the upper half plane. Equation (3a) provides a recurrence relation for the required matrix elements $M^{(N)}$.

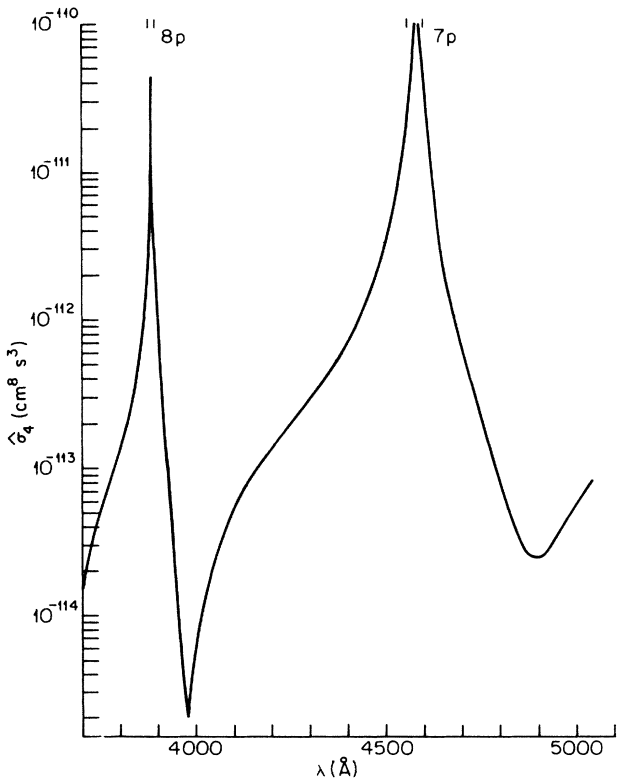

FIG. 1. Integrated generalized cross section (in $\mathrm{cm}^8\ \mathrm{sec}^3$) vs wavelength (in Å) for four-photon ionization of Cs by circularly polarized light.

The integrals which must be evaluated all have the following general form:

$$I_n(r)=\int_0^r dr_n f_n(r_n)\int_0^{r_n} dr_{n-1} \times f_{n-1}(r_{n-1})\cdots\int_0^{r_2} dr_1 f_1(r_1) \ . \tag{4}$$

These integrals satisfy a system of first-order coupled differential equations:

$$\frac{dI_n}{dr}=f_n(r)I_{n-1}(r), \quad I_n(0)=0 \ . \tag{5}$$

These differential equations can be integrated extremely rapidly using the Lawson form[9] of the Runge-Kutta class of algorithms. The Lawson method has a wide region of stability in the complex plane (most algorithms have a narrow region of stability around the real axis). Consequently, a large step size gives a meaningful result even when r is complex. The Lawson method was advocated previously and used with considerable success in work on the proton-hydrogen atom scattering problem.[10]

In our calculations the initial and final continuum electron wave functions were approximated by Coulomb waves, modified according to quantum-defect theory,[11] with the irregular part regularized[12] at the origin through multiplication of the cutoff factor $(1-\exp\{-10r/[a_0L(L+1)]\})^{2L+1}$. The radial Green's function was approximated by a Coulomb Green's function, also modified[13] as in quantum-defect theory; it can be expressed in coordinate space as

$$g_l(r,r';E)=-\frac{im}{\hbar^2 k}\Gamma(l+1-i\gamma)W_{i\gamma,l+1/2}(-2ikr_>) \times\left[\frac{1}{(2l+1)!}M_{i\gamma,l+1/2}(-2ikr_<)+\frac{2(-1)^l e^{-\pi\gamma}}{\Gamma(l+1+i\gamma)[1+i\cot\delta_l(E)]}W_{i\gamma,l+1/2}(-2ikr_<)\right] , \tag{6}$$

where $\hbar k=\sqrt{2mE}$ and $\gamma=Z/(a_0k)$, with Z the atomic number of the core, and where $\delta_l(E)$ is the phase shift due to the non-Coulomb part of the interaction; $\delta_l(E)$ is related to the quantum defect $\mu_l(E)$ by the relation[11,13]

$$\cot\delta_l(E)=(1-e^{-2\pi\gamma})\left[\cot\pi\mu_l(E)+\frac{iH(-E)}{1-e^{2\pi\gamma}}\right] , \tag{7}$$

where $H(E)$ is the Heaviside step function: $H(E<0)=0$, $H(E\geq 0)=1$. Note that Eq. (6) is valid for both positive and negative energies. The phase shift was obtained by interpolating between (when $E<0$) or extrapolating from (when $E>0$) the measured values of the quantum defects. The irregular Whittaker function in large parentheses on the right of Eq. (6) was multiplied by the same cut-off factor as mentioned above (but with L replaced by l). For large values of the argument the (regular and irregular) Whittaker functions were evaluated using the well-known asymptotic expansions; for smaller values of the argument, where the asymptotic expansions break down, the Whittaker functions were evaluated by using the numerical method described by Seaton.[14] The rotation angle for all integrations was taken to be $\pi/4$.

We first tested the method for four-photon ionization of hydrogen, with $\delta_l(E)=0$. By using only 50 points on the integration mesh we obtained agreement with the exact results to at least three significant figures. It was unnecessary to integrate beyond $|r|=40$ a.u., even in the case of above threshold absorption, when continuum-continuum matrix elements must be evaluated.

We also applied the method to Cs. For three- and four-photon ionization of the ground state with 5280-Å circularly polarized light we obtained good agreement with the previously published values of Aymar and Crance,[7] the relative difference being 45% or less. Again few integration points were needed (about 70, the slightly greater number being due to the greater extension and oscillation of the Cs $6s$ wave function). In Fig. 1 we show results for the integrated generalized cross section for four-photon ionization of ground-state Cs by circularly polarized light at wavelengths spanning part of the range between the thresholds for one- and two-photon ionization. The structure in the cross section is similar to that[15] for two-photon ionization at the same wavelengths. More detailed results for both Cs and Rb atoms, including angular distributions and comparisons with experimental data, will be given in a longer forthcoming paper.

The method has a wide range of applicability. It is straightforward to adapt it to the differential equation approach of Aymar and Crance, or to incorporate the random-phase approximation, for example.

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