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Two-frequency above-threshold ionization with smooth pulses

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A model of atomic ionization by smooth pulses is solved with bound-continuum and energy-dependent continuum-continuum dipole coupling. The rotating-wave approximation for the continuum-continuum transitions is avoided. In contrast with the flat continuum result, our ionization rate is independent of the continuum-continuum coupling. We also derive the photoelectron energy spectra and show that counterrotating terms make important contributions. Relation to above-threshold ionization experiments with two frequencies is discussed.

Our understanding of the photoelectric effect, as first explained by Einstein in 1905, has been revolutionized by the development of lasers to produce intense coherent radiation. These light sources made multiphoton ionization of matter possible, even when the photon energy was much less than the ionization potential.[1] However, more striking and surprising is the experimental finding of electrons with kinetic energies much larger than the photon energy;[2] this phenomenon caused by laser-induced electron transitions between the continua is called above-threshold ionization (ATI).

The experimental ATI photoelectron energy spectra exhibit several peaks that are separated by single-photon energy, $\hbar\omega_L$.[2] As the laser intensity is increased, the lower energy peaks are suppressed and the maximum of the spectrum occurs at higher electron energies; while all the previous mentioned results could be explained within the confines of perturbation theory, most researchers agree that this last result, peak suppression, could not be so explained and this finding challenged and stimulated the theoretical developments.

Nonperturbative models, to explain ATI, fall into three categories: (i) the ponderomotive potential models, which account for the electron energy change due to the electron's motion in a laser field gradient;[3] (ii) models that assume the Volkov-type final states, which retain the electromagnetic field nonperturbatively in the electron wave function;[4] and (iii) the essential states models, which identify those continuum states that get populated during the evolution.[5] A number of papers performing numerical integrations of the Schrödinger equation describing the electron in a (typically one-dimensional) potential subject to a time-dependent electric field[6] could also be added to this list.

In this Rapid Communication we solve a model which is closely related to the essential states models, but we avoid some of the simplifications made in the past. An attempt at avoiding the rotating-wave approximation was made by incorporating several transitions between the same continua.[5(e)] Our model of ATI is schematically depicted in Fig. 1. There are two colored lasers; the one with frequency ω_{L_0} promotes the electron from a bound state to continuum, the second laser has a frequency $\omega_{L_1} \ll \omega_{L_0}$ and has large enough intensity to induce multiple transitions in the continua. The continuum-continuum transitions are permitted to both higher and lower electron energies. Ideally, the ionization laser's energy would be large enough to promote the electron directly to the continuum and avoid the complications due to coupling between bound states and ponderomotive shifts of the threshold.

A variation of this scheme was experimentally realized in a study of ATI by Muller, van den Heuvell, and van der Wiel.[7] The first laser emitted light with ω_{L_0} in the ultra-

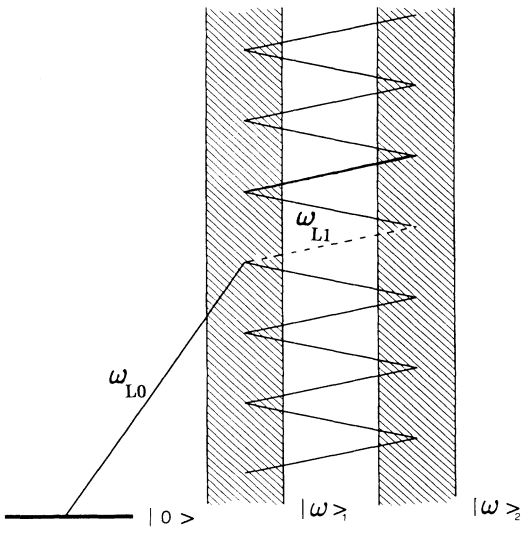

FIG. 1. A schematic illustration of the model. The bound state is coupled to one continuum. The two continua are coupled by multiple transitions. The dashed line indicates the only allowed transition in the rotating-wave approximation.

violet, whereas, the second emitted light in the infrared. The ultraviolet light was responsible for the multiphoton ionization of the atoms and induced continuum-continuum transitions, while the more intense laser in the infrared was responsible for repopulation between the continua and the observed multiple peaks in the photoelectron spectra. Two-frequency experiments provide the opportunity to separate out the ionization process from the continuum-continuum transitions.

Counter-rotating terms are retained in our analysis; this is important here because these terms also find resonance in the continua and permit multiple transitions between two continua as in Fig. 1. Use of the rotating-wave approximation would result in a single transition between the two continua, as shown by the dashed line in Fig. 1. The wave function is

$$|\Psi(t)\rangle = a(t)|0\rangle + \int d\omega\, C_1(\omega,t)|\omega\rangle_1 + \int d\omega\, C_2(\omega,t)|\omega\rangle_2 .$$

The energy ω is relative to the energy of the ground state, E_0, plus the ionizing photon energy, $\hbar\omega_{L_0}$. The amplitudes are coupled together by dipole matrix elements giving the following equations of motion:

$$\frac{da(t)}{dt} = -iV_i(t)\int d\omega\, C_1(\omega,t) , \tag{1}$$

$$\frac{dC_1(\omega,t)}{dt} = -i\omega C_1(\omega,t) - iV_i(t)a(t) - iV_1(t)\cos(\omega_{L_1}t)\int d\omega' D(\omega-\omega')C_2(\omega',t) , \tag{2}$$

and

$$\frac{dC_2(\omega,t)}{dt} = -i\omega C_2(\omega,t) - iV_1(t)\cos(\omega_{L_1}t)\int d\omega' D(\omega-\omega')C_1(\omega',t) . \tag{3}$$

The coefficient $V_i(t)$ is the matrix element coupling the bound state with a continuum state; its time dependence describes the pulse envelope. For the bound-continuum transition the rotating-wave approximation has been used and is well justified because the counterrotating terms, unlike the continuum-continuum transitions, are far from resonance. The laser frequency, ω_{L_0}, and the energy of the bound state can be scaled out of the problem. As discussed above we choose $E_0+\hbar\omega_{L_0}=0$. The energy-dependent matrix element, $D(\omega-\omega')$, coupling the two continua depends on the difference between the electron energies in each of the continua. This form is motivated by continuum-continuum matrix element calculations for hydrogenlike atoms; they have sharp maxima as the energy difference $\hbar(\omega-\omega')$ tends to zero.[8] The electric field amplitude coupling the continua $V_1(t)\cos(\omega_{L_1}t)$ has a carrier frequency ω_{L_1} and an envelope function $V_1(t)$. Models used in previous publications have either assumed that the matrix element is constant or that the energy dependence is separable into a product;[5] also, earlier publications on similar analytic models were not solved for smooth laser pulses.

The analytic solution of the equations of motion for the amplitudes is accomplished by taking the Fourier transform of the energy index ω in Eqs. (2) and (3) and using the method of characteristics to solve the resulting partial differential equations. The atom is initially in the bound state $a(0)=1$ and all other initial amplitudes vanish. The solution for the bound-state amplitude is remarkably simple; the ionizing pulse envelope is $V_i(t)=V_0 f_0(t)$, where the maximum value of $f_0(t)$, in unity, then the solution is

$$a(t) = \exp\left[-\frac{R}{2}\int_{-\infty}^{t} dt' f_0^2(t')\right] , \tag{4}$$

where the ionization rate is

$$R = 2\pi V_0^2 . \tag{5}$$

For square pulse envelope function $f_0(t)$, Eq. (4) is identical to the Weisskopf-Wigner result, which is the result expected from the Fermi's "golden rule" rate of the coupling between a bound state and a continuum. In the models of Ref. 5 the ionization rate is reduced by saturation effects of the continuum-continuum transitions; by contrast our rate is unaffected by these transitions.

The two probability amplitudes for the continua are given by

$$C_1(\omega,t) = -ie^{-i\omega t}\int_{-\infty}^{t} dk\, e^{+i\omega k} V_i(k)a(k)\cos\left[\int_k^t du\, \tilde{D}(u-k)V_1(u)\cos(\omega_{L_1}u)\right] , \tag{6}$$

and

$$C_2(\omega,t) = -e^{-i\omega t}\int_{-\infty}^{t} dk\, e^{+i\omega k} V_i(k)a(k)\sin\left[\int_k^t du\, \tilde{D}(u-k)V_1(u)\cos(\omega_{L_1}u)\right] . \tag{7}$$

The function $\tilde{D}(u-k)$ is the Fourier transform of $D(\omega)$. We illustrated these results by defining the following functions. For the continuum-continuum matrix element we choose a form that has the correct singularity derived near $\omega=0$

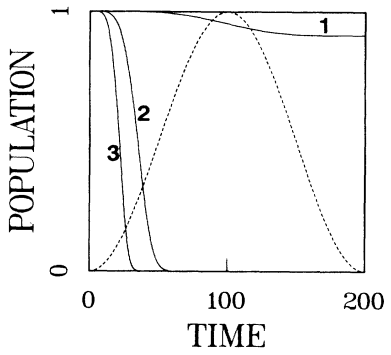

FIG. 2. The ground-state population vs time for three values of the ionization rate $R/2=10^{-4}$, 10^{-1}, and 1. The curves are labeled 1, 2, and 3, respectively; the time is in units of $2\pi/\omega_{L_1}$. Also shown by a dashed line is the pulse shape and duration.

for hydrogenic atoms[9]

$$D(\omega)=\frac{1}{\sqrt{2\pi}}\left[\frac{1}{(\omega+i\epsilon)^2}+\frac{1}{(\omega-i\epsilon)^2}\right]. \tag{8}$$

The parameter ϵ is made to vanish after all integrations are taken. The Fourier transform of this function is used in the evaluation of Eqs. (6) and (7). For the envelope functions we choose a squared cosine shape

$$f_0(t)=\begin{cases}\cos^2[(\pi/T_0)t], & -\frac{1}{2}T_0<t<\frac{1}{2}T_0,\\ 0 \text{ otherwise},\end{cases} \tag{9}$$

and

$$V_1(t)=\begin{cases}A\cos^2[\pi(t+t_0)/T_1], & -\frac{1}{2}T_1<(t+t_0)<\frac{1}{2}T_1,\\ 0 \text{ otherwise}.\end{cases} \tag{10}$$

The width of the pulse envelopes is chosen larger than the oscillation period $2\pi/\omega_{L_1}$. The two pulses may have different widths and one pulse can be time shifted with respect to the other by a time t_0.

Figure 2 shows the population of the ground state for three values of the rate R. We choose $T_0=400\pi/\omega_L$. At the smallest ionization rate $R/2=10^{-4}$, the ground-state population is not depleted and the laser pulse redistributing electrons among the continua acts over the entire ionization process. As the rate R is increased, the population is completely depleted by a smaller portion of the exciting pulse envelope. The rate is changed over four orders of magnitude.

In Fig. 3 we plot the long-time photoelectron spectra defined by

$$S(\omega)=\lim_{t\to\infty}[|C_1(\omega,t)|^2+|C_2(\omega,t)|^2]. \tag{11}$$

The value of A in Eq. (10) is taken as $5\omega_{L_1}$ and we have overlap of the ionization and redistribution pulses, $T_1=T_0$ and $t_0=0$. The three curves corresponding to the ionization rates used in Fig. 2 show how the photoelectron spectra are reshaped by the ionization rate. The peak amplitudes are strongly affected by the rate R.

The peaks at even indexed frequencies, 0, $\pm 2\omega_{L_1}$, $\pm 4\omega_{L_1}$, etc. are contributed from the population of the first continuum. The second continuum contributes the remaining peaks. This result is contrasted with other models using the rotating-wave approximation,[5] which only allow single transitions between continua (the dashed line in Fig. 1). The number of peaks in their spectra is determined by the number of continua in their model.

Our model also exhibits peak suppression. That is, the peak at $\omega=0$ is not the strongest for larger values of A. The value in Fig. 3 has been chosen to exemplify this phenomenon. For small R the maximum occurs at $\omega=\pm 7\omega_{L_1}$. This maximum changes to smaller values as R is increased. Physically, this phenomenon can be attributed to the fast ionization of the atom, as demonstrated in Fig. 2, which does not allow the electron to experience the entire continuum-continuum coupling pulse. The energy redistribution occurs mainly over the time that the electron is being ionized.

A two-color experiment, designed after that of Muller, van den Heuvell, and van der Wiel,[7] with a strong laser at frequency ω_{L_1} could distinguish between the results of our model and the earlier essential states treatment. We do not predict saturation of the ionization rate and we find that continuum-continuum transitions to higher and lower electron energies are symmetric, as long as the continuum edge is not intervening to prevent transitions to lower energies.

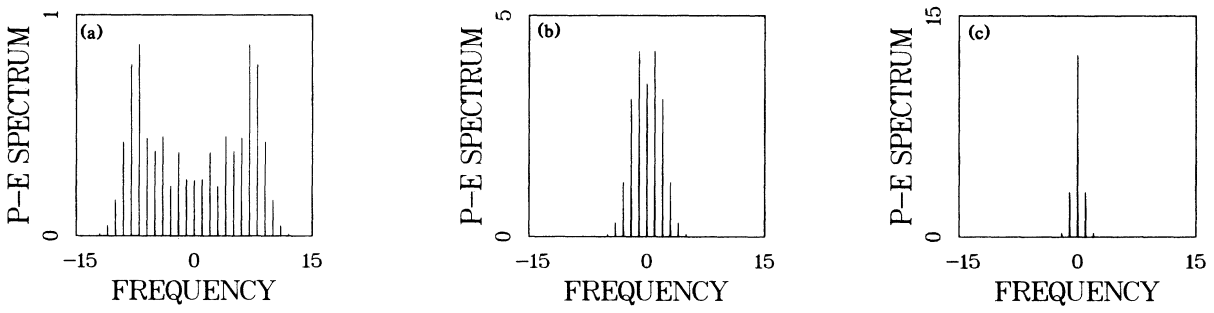

FIG. 3. The long-time photoelectron spectrum as a function of R for the values used in Fig. 2, labels (a)–(c) begin with the smallest value of R and progress to the largest value. The frequency is scaled to ω_{L_1}.

Our model differs from previous essential states treatments of ATI in three respects. (i) We use an energy-dependent continuum-continuum transition matrix element that does not have a separable energy dependence. (ii) We do not resort to the rotating-wave approximation for continuum-continuum transitions. Analytical results are given for the entire time evolution of the amplitudes, Eqs. (6) and (7), and the photoelectron spectra at long times exhibit the characteristic signatures of ATI, multiple peaks, and peak suppression. (iii) We use smooth pulses to excite the electron between the states.

We remark that two-color ATI has been discussed within the context of strong-field approximations and the S-matrix approach by Leone *et al.*[10] They do not give the time evolution of the wave-function amplitudes or treat smooth pulse solutions, nor can they give the widths of the photoelectron spectra. Nevertheless, some of the photoelectron spectra obtained from their paper resemble our own; their total cross-section calculations exhibit many peaks with peak suppression at higher intensities. When the peaks are shifted far from the threshold, including the ponderomotive shift of the threshold, then the spectra are symmetric.

Finally, the reader should recognize that experimental spectra are statistical averages over many repetitions of the experiment. Further complications occur because the laser intensity fluctuates, from shot to shot, and the position of the atom in the laser field also fluctuates; hence the comparison of our results with experiment should be done after a statistical average over field amplitude A and the rates R have been performed. This will smooth the multipeak structure that we have presented in this Rapid Communication.

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