Dynamic evolution of above-threshold ionization spectra

Guan-hua Yao
Shanghai Institute of Optics and Fine Mechanics, Academia Sinica, P.O. Box 8211, Shanghai, People's Republic of China

Zhi-zhan Xu and Wei Yu
Center of Theoretical Physcis, China Center of Advanced Science and Technology (World Laboratory), Beijing, People's Republic of China
and Shanghai Institute of Optics and Fine Mechanics, Academia Sinica, P.O. Box 8211, Shanghai, People's Republic of China

(Received 8 March 1989)

Above-threshold ionization (ATI) by a smooth laser pulse is investigated in terms of the essential-state approach. Time evolution of ATI spectra is demonstrated. It is shown that the size of the electron peaks grows with the interaction time, and the peaks' bell-shaped envelope in the spectra moves to higher electron energies during the rise of the laser pulse.

I. INTRODUCTION

The recent discovery[1-5] of many novel features in above-threshold ionization (ATI) has invoked a great amount of theoretical interest. A zero-rangle potential model was first studied by Muller *et al.*[6] and Muller and Tip,[7] and the simultaneous influence of a ponderomotive effect and an AC Stark shift[6-8] was stressed. In addition, Crance has proposed a simple statistical model[9] and she has also studied the space-charge effect.[10]

Another line of thought is represented in a series of papers by Bialynicka-Birula,[11] Edwards *et al.*,[12] and Deng and Eberly[13,14] who used the so-called "essential-state" approach (ESA) and pointed to the importance of saturation in continuum-continuum transitions. This approach has been widely used and extended to calculate angular momentum distribution of photoelectrons[15] and to include the diagonal couplings between different continuum states.[16] In these studies, the nonperturbative description in terms of ESA proves to be successful in explaining many common features of ATI, such as peak switching of ATI spectra, significant suppression of lowest-order peaks with increasing laser intensity, and roughly the same order of nonlinearity, etc. However, as all these authors neglected the temporal shape of the laser pulse, they could not offer an adequate account of the dynamical evolution of ATI electron spectra, such as the formation of the bell-shaped envelope characterizing peak switching in ATI spectra.

In this paper, we extend previous work of the ESA by considering ATI by a smoothly switched-on laser pulse and investigate how ATI spectrum evolves in time. Our results indicate that the peaks' envelope in ATI spectra move to higher electron energies during the rise of the pulse, while the size of these peaks grows with the interaction time. These results underscore the importance of accounting for the temporal laser shape and are helpful to the understanding of the dynamic behavior of ATI.

II. MODEL DESCRIPTION

We consider a model atom irradiated by a smooth laser pulse which is described by $I(t)$. As in standard ESA treatments,[11-14] the model space of the atom plus field system consists of essential states: the ground state $|0\rangle$ and a set of continuum bands $|\omega_l\rangle$ $(l=1,2,\ldots)$. The ground state is connected to the first continuum $|\omega_1\rangle$ via N-photon absorption; the dipole moment is described by an effective matrix element D_{0,ω_1}. In addition, transition matrix elements between two successive continua are denoted by $D_{\omega_l,\omega_{l+1}}$. Note that all these matrix elements are proportional to the square root of the laser intensity and thus are time dependent.

Under this model, the equations of motion governing time evolution of the state probabilities are given by

$$i\dot{u}_0 = \int d\omega_1 D_{0,\omega_1}(t) u_{\omega_1}(t) , \tag{1a}$$

$$i\dot{u}_{\omega_1} - \Delta(\omega_1) u_{\omega_1} = D_{\omega_1,0}(t) u_0(t) + \int d\omega_2 D_{\omega_1,\omega_2}(t) u_{\omega_2}(t) , \tag{1b}$$

$$i\dot{u}_{\omega_l} - \Delta(\omega_l) u_{\omega_l} = \int d\omega_{l-1} D_{\omega_l,\omega_{l-1}}(t) u_{\omega_{l-1}}(t) + \int d\omega_{l+1} D_{\omega_l,\omega_{l+1}}(t) u_{\omega_{l+1}}(t) , \quad l=2,3,\ldots, \tag{1c}$$

where

$$\Delta(\omega_l) = \omega_l - (N+l-1)\omega_0 , \tag{2}$$

are energies of the continuum states $|\omega_l\rangle$, and ω_0 is the laser frequency. Though Eqs. (1) are of the same form as those given by Deng and Eberly,[14] there is an essential difference between theirs and ours. In Eqs. (1), all dipole couplings are time dependent, so that it is generally formidable to derive their exact solutions. However, as will

be seen hereafter, they are soluble if the continua are assumed to be flat, i.e., $D_{\omega_l,\omega_{l+1}} \simeq D_{l,l+1}$ and $D_{0,\omega_1} \simeq D_{01}$ are assumed.[11-14]

To obtain these solutions, it is helpful to introduce a new set of quantities:

$$K_l(t) = \int d\omega_l u_{\omega_l}(t) \tag{3}$$

and to rewrite Eqs. (1) under the assumption of flat continua as

$$i\dot{u}_0 = D_{01}(t)K_1(t) , \tag{4a}$$

$$i\dot{u}_{\omega_1} - \Delta(\omega_1)u_{\omega_1} = D_{10}(t)u_0(t) + D_{12}(t)K_2(t) , \tag{4b}$$

$$i\dot{u}_{\omega_l} - \Delta(\omega_l)u_{\omega_l} = D_{l,l-1}(t)K_{l-1}(t) + D_{l,l+1}(t)K_{l+1}(t) , \quad l = 2,3,\ldots . \tag{4c}$$

It is worth stating that the terms on the right-hand side of the above equations are independent of continuum energies ω_l, whereas those on the left-hand side rely explicitly on ω_l. Thus it is reasonable to define the right-hand-side terms as $G_l(t)$. The state amplitude $u_{\omega_l}(t)$ satisfies

$$i\dot{u}_{\omega_l} - \Delta(\omega_l)u_{\omega_l} = G_l(t) , \tag{5}$$

which can be solved formally. According to the definition of $K_l(t)$, we have

$$K_l(t) = -\int_{-\infty}^{t} G_l(t) \left[\int d\omega_l e^{-i\Delta(\omega_l)(t-\tau)} \right] d\tau , \tag{6}$$

where the integration over ω_l runs from $\omega_l = 0$ to infinity. Ignoring threshold effect, we obtain from Eq. (6)

$$K_l(t) = -i\pi G_l(t) . \tag{7}$$

This is an important result which directly stems from the assumption of flat continua and which greatly facilitates the solution of the problem.

Taking advantage of this relation, we lead to an infinite number of algebraic equations determining $K_l(t)$,

$$iK_1(t) = \pi D_{10}(t)u_0(t) + \pi D_{12}(t)K_2(t) , \tag{8a}$$

$$iK_l(t) = \pi D_{l,l-1}(t)K_{l-1}(t) + \pi D_{l,l+1}(t)K_{l+1}(t) \quad (l = 2,3,\ldots) . \tag{8b}$$

For a sufficient large m, the $|\omega_{m+1}\rangle$ continuum has a neglibible effect on the lower continua, so that we may cut off the infinite set of equations by taking $D_{m,m+1}(t) = 0$. The analytical expressions of $K_l(t)$ are then

$$K_l(t) = (-1)^l \left[\prod_{q=1}^{l} \pi D_{q-1,q}(t)R_q(t) \right] u_0(t) , \tag{9}$$

were R_q satisfies recursion relation

$$R_q(t) = \frac{1}{1 + Z_{q+1}(t)R_{q+1}(t)} , \qquad Z_q(t) = \pi^2 |D_{q,q-1}(t)|^2 , \tag{10}$$

with $R_{m+1}(t) = 0$. They are able to be expressed as a finite-continued fraction (see, e.g., Ref. 14). On substituting $K_1(t)$ into eq. (4a), the ground-state population reads

$$P_0(t) = |u_0(t)|^2 = \exp\left[-2\pi \int_{-\infty}^{t} d\tau |D_{10}(\tau)|^2 R_2(\tau) \right] . \tag{11}$$

It is straightforward to derive probability amplitudes of continuum states,

$$u_{\omega_l}(t) = (-i)^l/\pi \int_{-\infty}^{t} S_l(\tau)P_0(\tau)e^{-i\Delta(\omega_l)(t-\tau)} d\tau , \tag{12}$$

and the total population of the lth peak,

$$P_l(t) = \int_0^{\infty} |u_{\omega_l}(t)|^2 d\omega_l = \frac{2}{\pi} \int_{-\infty}^{t} S_l^2(\tau)P_0(\tau)d\tau , \tag{13}$$

where the threshold effect is also ignored, and $S_l(t)$ is defined as the part inside the large parentheses of Eq. (9).

Equations (11) and (13) are two of the main results of the present work. They determine the dynamic behavior of the system and can be applied to the ATI of atomic or molecular systems with arbitrary continuum-continuum

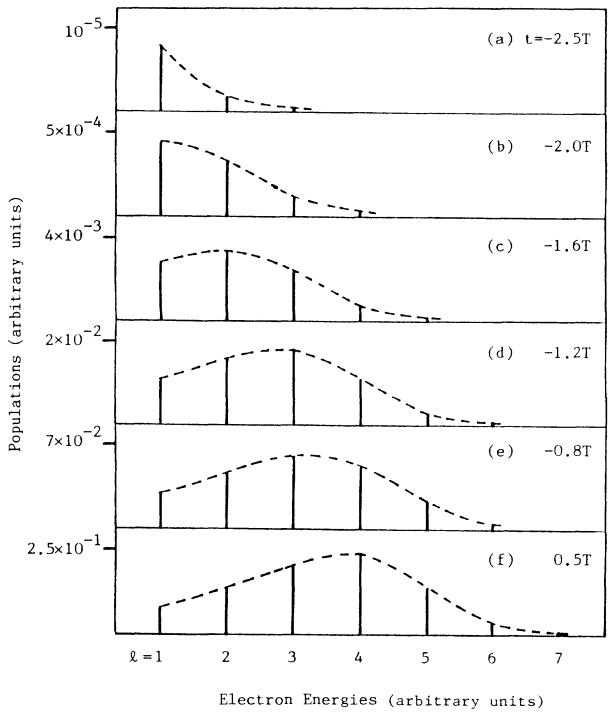

FIG. 1. Time evolution of ATI photoelectron spectra, showing how the peaks' bell-shaped envelope is switched from low continuum states to higher ones. The temporal shape of the laser pulse is given in Eq. (16). t is the interaction time. Note that the vertical lines are total populations for some given continuum states, and the scales in these boxes are increased from the top to the bottom. Parameters are $N = 2$, $T = 20$, $\beta = 2$, and $I_0 = 80$.

transition-dipole moments $D_{l,l+1}$ by a laser pulse with arbitrary pulse shape and laser intensity.

If we ignore the temporal effect of the pulse shape and describe $I(t)$ as a square pulse,[14] then $R_l(t)=R_l$ and $S_l(t)=S_l$ are constant during the interaction time. In this simplest case,

$$P_0(t)=e^{-2\pi D_{10}^2 R_1 t} \tag{14}$$

is simply an exponential decay function, and

$$P_l(t)=R_1\prod_{q=2}^{l} Z_q R_q^2(1-e^{-2\pi D_{10}^2 R_1 t}) , \tag{15}$$

It turns out from Eq. (15) that all the continua exhibit the same time behavior. Obviously it is an unrealistic result which leads to the conclusion that peak switching, if it happens, is formed at the very moment when the laser pulse is switched on.

On the other hand, as is apparent in Eq. (13), different continua will have different time behaviors as long as $S_l(t)$ in the equation is a function of time, i.e., when the pulse shape is taken into account. Therefore, the detailed temporal shape of the laser pulse is essential to the understanding of the dynamics of ATI.

III. RESULTS AND DISCUSSIONS

Although Eqs. (11) and (13) are applicable to arbitrary pulse shape, we will deal with, in this paper, a Gaussian laser pulse described by

$$I(t)=I_0 e^{-(t/T)^2} , \tag{16}$$

where I_0 is the peak intensity of the pulse and T is the characteristic time of the pulse which relates to the pulse width τ_0 by $\tau_0=2T\ln 2$. In addition, in view of the fact that matrix elements $D_{l,l+1}$ decrease slowly as the index l increases[17] we choose them to be a decreasing function according to the simple empirical rule[14]

$$D_{l,l+1}/D_{l+1,l+2}=\beta \quad (l=1,2,\cdots) . \tag{17}$$

Displayed in Fig. 1 are the photoelectron energy distributions for different interaction times. The vertical lines are areas of consecutive electron peaks and the dashed lines are envelopes of the electron distributions. The maximum of $Z_{12}(t)$ is defined as the peak laser intensity I_0 and is chosen in this figure to be 80 to ensure that continuum-continuum saturation giving rise to peak switching is set up in the end. We aim to understand how such peak switching is formed in time. At the beginning (the uppermost box) when the instantaneous laser intensity is small, a decrement of peak areas with l is observed, which is consistent with perturbation prediction. However, as the instantaneous intensity is increased during the rise of the pulse, it is seen that the peak with maximum peak area is switched from the first continuum to the second, and then from the second to the third, and so on. Hence the bell-shaped structure of the photoelectron spectrum, which is a striking feature of ATI, is formed during the rising time of the laser pulse, and higher-lying continuum states successively become most populated.

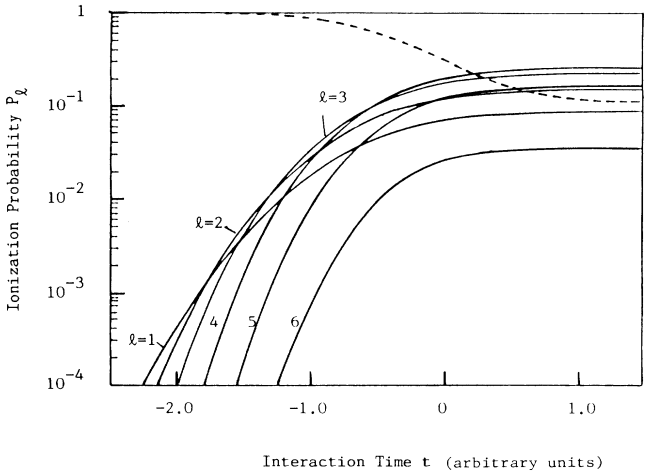

FIG. 2. Detailed time behavior of the photoelectron energy distribution. The dashed curve is the ground-state population and the parameters are the same as those in Fig. 1.

In addition, note increasing scales from top to bottom in these boxes; the populations of all continuum bands increase with the interaction time. Thus the whole envelope itself grows with the interaction time as it is switched to higher electron energies.

A detailed dynamical evolution of ionization probability is shown in Fig. 2. The parameters are the same as in Fig. 1. The dashed line is the time behavior of the ground-state population and the solid curves are populations of different continuum states corresponding to the absorption of different numbers of excess photons. The crossing points in these curves have the evident meanings of critical time when the l_1th peak area starts to exceed the l_2th peak. Therefore it is very clear to see how the population of higher-order ionization states gradually exceeds those of lower-order ones and finally forms a stationary distribution. In addition, it is also interesting to observe that dynamic behavior of various continua are different. For instance, with increasing interaction time, the ionization to higher continuum states becomes faster as compared to lower ones. Finally, the time behavior of the ATI for larger values of N is easy to study, although here we have only discussed the case of $N=2$. It can be shown that the qualitative results will be the same except that the population of higher continuum states will exceed lower ones at an earlier time.

The time evolution of ATI spectra given above may be understood as follows. At the beginning when the laser intensity is so weak that all continuum-continuum transition channels are not "open," the lowest-order perturbation description is valid and the first continuum is most populated. This is the case of $t=-2.5T$ in Fig. 1, where a fast decrement of the size for consecutive peaks is visible. By increasing the interaction time during the rising of the pulse, the resulting instantaneous laser intensity is increased. When it is strong enough to make $Z_{12}(t)$ approach to unity, the coherent coupling channel between the first and the second continuum states is

open[14] and the second continuum is then quickly populated. When the laser intensity is further increased, so that even $Z_{23}(t)$, which is equal to $Z_{12}(t)/\beta^2$, is greater than unity, the corresponding continuum states are strongly coupled and the transition channel is open. Meanwhile, the $\omega_1-\omega_2$ transition may be saturated so that more population current is accumulated in the third continuum, as argued by Deng and Eberly.[14] In this way, more channels of coherent transition between two successive continuum levels are open when the laser intensity is increased during the rising of the laser pulse, and the most populated continuum is changed from low energy to high-electron energy.

It is necessary to point out that, in the limiting case when the laser intensity is very close slowly time dependent (adiabatic), the picture given above regarding the formation of the bell-shaped envelope may be deduced directly from the results of Deng and Eberly.[14] Under the circumstance, the decay rates are described by the square pulse results,[11-14] but they vary with time through the laser intensity. The relative instantaneous ionization rates then change with the interaction time and the peak switching is formed during the rise of the pulse. In spite of the above equivalence in the adiabatic limit, however, our results are not restricted to adiabatic approximation, as the only assumption we have made is that the pulse varies in time much more slowly than the laser frequency, i.e., $(dI/dt)/I \ll \omega_0$, which is often the case for optical frequency. As a consequence, the present results are more generally applicable than the adiabatic results. Finally, the diagonal continuum-continuum couplings may be included in the present work to deal with the regime of higher field strengths, but the qualitative results will be unchanged. We will return to this problem in future research.

In conclusion, we have extended previous theories of ESA by considering the temporal effects of the laser pulse in ATI. Neglecting the detailed continuum structure, analytical expressions for ground-state population and the photoelectron spectra are available for arbitrary pulse shape and continuum-continuum couplings. In particular, we have taken Gaussian pulse and decreasing coupling coefficients as an example to discuss the time evolution of ATI spectra. It has been found that different continua have different time behavior, as expected, and with the size of the whole electron distribution growing, the center of the peaks' envelope in ATI spectra is switched successively from the lowest peaks to a certain higher one during the rise of the pulse.

ACKNOWLEDGMENTS

The authors are gratefully indebted to Dr. Z. Deng for helpful discussions.

[1]P. Kruit, J. Kimman, H. G. Muller, and M. J. van der Wiel, Phys. Rev. A **28**, 248 (1983).

[2]L. A. Lompre, A. Huillier, G. Mainfray, and C. Manus, J. Opt. Soc. Am. B **2**, 1906 (1985); L. A. Lompre, G. Mainfray, C. Manus, and J. Kupersztych, J. Phys. B **20**, 1009 (1987).

[3]F. Yergeau, G. Petite, and P. Agostini, J. Phys. B **19**, L663 (1986); F. Yergeau, S. L. Chin, and P. Lavigne, *ibid.* **20**, 723 (1987).

[4]R. R. Freeman, T. J. McIlrath, P. H. Bucksbaum, and M. Bashkansky, Phys. Rev. Lett. **57**, 3156 (1986); P. H. Bucksbaum, R. R. Freeman, M. Bashkansky, and T. J. McIlrath, J. Opt. Soc. Am. B **4**, 760 (1987); R. R. Freeman, P. H. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. E. Geusic, Phys. Rev. Lett. **59**, 1092 (1987); H. G. Muller, H. B. van der Heuvell, P. Agostini, G. Petite, A. Antonetti, M. Franco, and A. Migus, Phys. Rev. Lett. **60**, 565 (1988).

[5]M. Bashkansky, P. H. Bucksbaum, and D. W. Schumacher, Phys. Rev. Lett. **59**, 274 (1987); **60**, 2458 (1988).

[6]H. G. Muller, A. Tip, and M. J. van der Wiel, J. Phys. B **16**, L679 (1983).

[7]H. G. Muller and A. Tip, Phys. Rev. A **30**, 3039 (1984).

[8]S. I. Chu and J. Cooper, Phys. Rev. A **32**, 2769 (1985); A. Szoke, J. Phys. B **18**, L427 (1985); L. Pan, L. Armstrong, Jr., and J. H. Eberly, J. Opt. Soc. Am. B **3**, 1319 (1986).

[9]M. Crance, J. Phys. B **17**, L355 (1984); **17**, 3503 (1984); **17**, 4323 (1984); **17**, 4333 (1984).

[10]M. Crance, J. Phys. B **19**, L267 (1986); **19**, L671 (1986).

[11]Z. Bialynicka-Birula, J. Phys. B **17**, 3091 (1984).

[12]M. Edwards, L. Pan, and L. Armstrong, Jr., J. Phys. B **17**, L515 (1984); **18**, 1927 (1985).

[13]Z. Deng and J. H. Eberly, Phys. Rev. Lett. **53**, 1810 (1984); J. Phys. B **18**, L287 (1985).

[14]Z. Deng and J. H. Eberly, J. Opt. Soc. Am. B **2**, 486 (1985).

[15]K. Rzazewski and R. Grobe, Phys. Rev. A **33**, 1855 (1986).

[16]See, for example, H. Lewenstein, J. Mostowski, and M. Trippenbach, J. Phys. B **18**, L461 (1985); J. Grochmalicki, J. R. Kuklinski, and M. Lewenstein, J. Phys. B **19**, 3649 (1986); A. Dulcic, Phys. Rev. A **35**, 1673 (1987); M. Trippenbach, K. Rzazewski, and R. Grobe, *ibid.* **37**, 4194 (1988).

[17]Y. Gontier and M. Trahin, J. Phys. B **13**, 4383 (1980); M. Aymar and M. Crance, J. Phys. B **14**, 3585 (1980).