# Strong-field autoionization induced by smooth laser pulses, including high-order ionization processes

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Strong-field autoionization induced by smooth laser pulses is studied to take into account the high-order ionization processes. A second-order differential equation of the amplitude of the ground state is derived. For some special laser pulses, which include all the exponential ones, analytical results may be obtained. As an example, the explicit results of ground-state population and the photoelectron spectrum are given for an exponentially switched-on cw pulse. The multipeak structure in the photoelectron spectrum is more obviously manifested in the high spectrum resulted from the high-order ionization processes.

## I. INTRODUCTION

Recently there has been some interest in the study of the interaction of atoms with time-dependent laser fields. Many methods have been developed for two-level atoms [1]. As is known, atoms with autoionization induced by smooth laser pulses are a rather complicated system. In fact, it was in 1983 when Rzążewski presented the first results dealing with this system and showed that the photoelectron spectrum (PES) exhibited a multipeak structure with the number of peaks closely related to the laser-pulse area [2]. From that time on, the above system has been studied for some particular laser-pulse envelopes and also the fluorescence energy spectrum has been calculated [2–5]. The multipeak structure is discovered to be essential to the atoms subjected to a smooth laser pulse [2–6].

On the other hand, strong-field autoionization has been studied extensively for time-independent cw laser irradiation, which switches on suddenly and remains unchanged during the entire interaction time, with many physical mechanisms taken into account [7–15]. Specially among them are the high-order ionization processes, such as the photoionization of the autoionizing state (AIS). The photoionization of the AIS refers to the transition from the AIS to the higher states of the continuum and the rate of this quasi-bound-continuum transition, proportional to the laser intensity, can be expected of the same order of magnitudes as the direct photoionization of the bound state, usually the ground state, which is a bound-continuum transition [4]. And it becomes more and more important under strong laser fields and drastically affects the autoionization processes in the cw cases [9,14,15]. And still more, during the last two years, the interest in autoionization has been enforced because it bears close relation to the laser action without inversion [16].

This paper presents the first attempt to tackle with, simultaneously, a smooth laser pulse and the photoionization of the AIS. After describing the model atom and using the Schrödinger time-dependent equation we are able to obtain a second-order differential equation for the ground-state amplitude. This equation can be solved numerically for arbitrary laser pulses. And analytical results may be obtained for some special pulses, especially with various exponential ones included. For an exponential switched-on cw pulse, for example, the explicit results of the ground-state population and the long-time PES are given.

## II. THE MODEL AND THE EQUATIONS

Our model atom is shown in Fig. 1. It consists of a bound state (ground state) $|0\rangle$, an AIS $|a\rangle$, and two continua $|\omega_1\rangle$ and $|\omega_2\rangle$. $|0\rangle$ to $|a\rangle$ and $|a\rangle$ to $|\omega_2\rangle$ are coupled by a strong laser pulse with an envelope $E(t)=Ef(t)$ where $E$ is its typical field strength and $f(t)$ its dimensionless shape. The configuration interaction couples $|a\rangle$ to $|\omega_1\rangle$, while the latter cannot be reached directly (because of, for example, selection rules) from the ground state, i.e., the coupling between $|0\rangle$ and $|\omega_1\rangle$ is not taken into account, which corresponds to the simplest, symmetric profile of the autoionization resonance under weak laser fields (Fano asymmetric parameter related with $|0\rangle$ is infinite [16]). Also neglected is the continuum-continuum coupling between $|\omega_1\rangle$ and $|\omega_2\rangle$ (Fano asymmetric parameter related with $|\omega_2\rangle$ is infinite too [14,17]) which is a well-established approximation under the usual strong fields in the autoionization study [18], because in this case the above continuum-continuum coupling is far below its saturation [19].

The Hamiltonian of the above system under the rotating-wave approximation reads ($\hbar=1$) as

$$H=E_a|a\rangle\langle a|+\int d\omega_1\omega_1|\omega_1\rangle\langle\omega_1|+\int d\omega_2\omega_2|\omega_2\rangle\langle\omega_2| + \left[D_{0a}f(t)e^{i\omega_L t}|0\rangle\langle a|+\int d\omega_2 D_{a2}f(t)e^{i\omega_L t}|a\rangle\langle\omega_2|+\text{H.c.}\right]+\left[\int d\omega_1 V_{a1}|a\rangle\langle\omega_1|+\text{H.c.}\right], \quad (1)$$

 

where, for convenience, we have chosen the ground-state energy $E_0=0$, and integrals extend over the entire real axis, for we neglect the ionization threshold as done by Rzążewski and co-workers [2,3].

Introducing the following form of the state vector

$$|\Psi(t)\rangle=u_0(t)|0\rangle+\left[u_a(t)|a\rangle+\int d\omega_1 u_1(\omega_1,t)|\omega_1\rangle\right]e^{-i\omega_L t}+\int d\omega_2 u_2(\omega_2,t)e^{-2i\omega_L t}|\omega_2\rangle\ , \tag{2}$$

we obtain from the time-dependent Schrödinger equation a set of integro-differential $c$-number equations:

$$i\dot{u}_0=D_{0a}fu_a\ , \tag{3}$$

$$i\dot{u}_a=(\omega_a-\omega_L)u_a+D_{0a}f^*u_0+\int d\omega_1 V_{a1}u_1+\int d\omega_2 D_{a2}fu_2\ , \tag{4}$$

$$i\dot{u}_1=(\omega_1-\omega_L)u_1+V_{1a}u_a\ , \tag{5}$$

$$i\dot{u}_2=(\omega_2-2\omega_L)u_2+D_{2a}f^*u_a\ . \tag{6}$$

Assuming that the atom was in the ground state before the pulse arrived so that

$$\lim_{t\to-\infty}[u_0(t),u_a(t),u_1(\omega_1,t),u_2(\omega_2,t)]=(1,0,0,0) \tag{7}$$

and

$$\lim_{t\to-\infty}f(t)=0\ , \tag{8}$$

we can formally solve Eqs. (5) and (6):

$$u_1(\omega_1,t)=-i\int_{-\infty}^{t}d\tau\, e^{-i(\omega_1-\omega_L)(t-\tau)}V_{1a}u_a(\tau)\ , \tag{9}$$

$$u_2(\omega_2,t)=-i\int_{-\infty}^{t}d\tau\, e^{-i(\omega_2-2\omega_L)(t-\tau)}D_{2a}f^*(\tau)u_a(\tau)\ . \tag{10}$$

Recalling that the matrix elements $V_{1a}$ and $D_{2a}$ are slowly varying functions of $\omega_1$ and $\omega_2$ over the range of the resonance [8], we consider them as constants evaluated, respectively, at $\omega_1=\omega_a$ and $\omega_2=\omega_a+\omega_L$. This results in

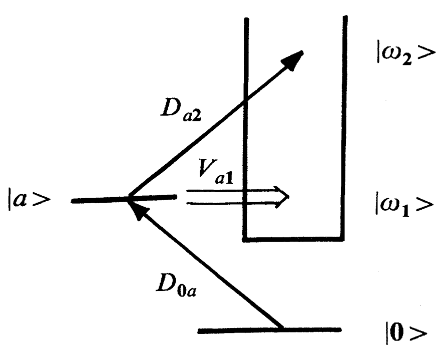


FIG. 1. Schematic representation of the model space and couplings in the unperturbed representation. The states $|0\rangle$ and $|a\rangle$ denote the initial and the autoionizing state, whereas $|\omega_1\rangle$ and $|\omega_2\rangle$ represent the first and second (single-electron) continua, respectively. The one-way arrow denotes the laser-stimulated transition, and the two-way arrow indicates the configuration interaction between the autoionizing state and the first continuum.

$$\int d\omega_1 u_1(\omega_1,t)=-i\pi V_{1a}u_a(t), \tag{11}$$

$$\int d\omega_2 u_2(\omega_2,t)=-i\pi D_{2a}f^*(t)u_a(t)\ . \tag{12}$$

Inserting the above two equations into (4) we arrive at

$$i\dot{u}_a=(\Delta-i\gamma_0-i\gamma_2 f^*f)u_a+D_{a0}f^*u_0\ , \tag{13}$$

where $\Delta=\omega_a-\omega_L$ is the laser detuning and $\gamma_0=\pi|V_{a1}|^2$ and $\gamma_2=\pi|D_{a2}|^2$ are the half-maximum widths, respectively, of the autoionization resonance and of the photoionization of the AIS. Using (2) and after some manipulations we can eliminate from Eq. (13) the amplitude $u_a(t)$:

$$\ddot{u}_0+[-\dot{f}/f+i\Delta+\gamma_0+\gamma_2 f^*f]\dot{u}_0+D_{a0}D_{0a}f^*fu_0=0\ . \tag{14}$$

The above equation, similar to those discussed by several other authors [2–5], is essential in strong-field autoionization under smooth laser pulses for the model concerned [20]. In the case of $\gamma_2=0$, it can be reduced by the one discussed by Rzążewski [2]. And after the solution to (14) being got, the other amplitudes can be obtained from (2), (9), and (10):

$$u_a(t)=i\dot{u}_0(t)/D_{0a}f(t)\ , \tag{15a}$$

$$u_1(\omega_1,t)=\frac{V_{1a}}{D_{0a}}\int_{-\infty}^{t}d\tau\, e^{-i(\omega_1-\omega_L)(t-\tau)}\frac{\dot{u}_0(\tau)}{f(\tau)}\ , \tag{15b}$$

and

$$u_2(\omega_2,t)=\frac{D_{2a}}{D_{0a}}\int_{-\infty}^{t}d\tau\, e^{-i(\omega_2+2\omega_L)(t-\tau)}\frac{f^*(\tau)}{f(\tau)}\dot{u}_0(\tau)\ , \tag{15c}$$

In laser-induced autoionization as in all ionizations induced by laser, the PES is an important physical quantity, which is now composed of two parts:

$$W_1(\omega,t)=|u_1(\omega,t)|^2\ ,\quad W_2(\omega,t)=|u_2(\omega,t)|^2\ . \tag{16}$$

For an arbitrary laser pulse we must solve Eq. (14) numerically. For some special laser pulses, however, analytical results may be obtained. Important among these special ones are those composed of exponentially time-dependent fields.

## III. RESULTS AND DISCUSSIONS

As an example, we present here the results for an exponentially switched-on cw pulse:

$$f(t)=\begin{cases} e^{\gamma t} & \text{for } t\le 0 \\ 1 & \text{otherwise.} \end{cases} \tag{17}$$

The population of the ground state and the long-time PES may be expressed explicitly in terms of the confluent hypergeometric functions:

$$P_0(t)=\begin{cases} |{}_1F_1(\alpha;\beta;z)|^2 & \text{for } t\le 0 \\ Ae^{iE_1 t}+Be^{iE_2 t} & \text{for } t\ge 0\ , \end{cases} \tag{18a}$$

$$W_1(\omega,\infty)=\frac{|D_{0a}|^2}{\pi\gamma_0}|\theta(\omega)(I_1-I_1')|^2, \tag{18b}$$

$$W_2(\omega,\infty)=\frac{\gamma_2}{\pi|D_{0a}|^2}|I_2-I_2'|^2\ , \tag{18c}$$

where

$$\alpha=|D_{a0}|^2/2\gamma\gamma_2,\quad \beta=(\gamma_0+i\Delta)/2\gamma\ ,$$
$$z_0=-\gamma_2/2\gamma,\quad z=z_0e^{2\gamma t},\quad \theta(\omega)=\gamma_0/(\omega-\omega_a+i\gamma_0)\ ,$$
$$I_1={}_2F_2(\alpha,\chi_1;\beta,\chi_1+1;z_0)/2\gamma\chi_1+{}_2F_2(\alpha+1,\chi_1+1;\beta+1,\chi_1+2;z_0)/\beta(1+\chi_1),$$
$$I_1'=A(1+iE_1A)/iE_1'+B(1+iE_2B)/iE_2'\ ,\quad I_2=\alpha z_0\,{}_2F_2(\alpha+1,\chi_2;\beta+1,\chi_2+1;z_0)/\beta\chi_2\ ,$$
$$I_2'=AE_1/E_1'+BE_2/E_2',\quad \chi_k=[k\gamma+i(\omega-k\omega_L)]/2\gamma,\quad E_k'=E_k+\omega-k\omega_L\quad \text{for } k=1,2\ , \tag{19}$$

with $E_1$ and $E_2$ to be the roots satisfying

$$E^2+[\Delta-i(\gamma_0+\gamma_2)]-|D_{0a}|^2=0, \tag{20}$$

while $A$ and $B$ are determined by the continuity at $t=0$:

$$\begin{aligned} A+B&=u_0(t=0)\ ,\\ AE_1+BE_2&=-i\dot{u}_0(t=0). \end{aligned} \tag{21}$$

In the case of $\gamma_2=0$, the above results are reduced to those obtained by Rzążewski and co-workers [3]. They can be used to discuss the influences of the photoionization of the AIS on strong-field autoionization.

In the following calculations we have scaled the peak intensity (or, more accurately, the peak value of the oscillating electric field, which is proportional to $D_{0a}$) and the width of the photoionization of the AIS:

$$\eta=|D_{0a}|/\gamma_0,\quad \xi=\gamma_2\gamma_0/|D_{0a}|^2\ . \tag{22}$$

Therefore $\xi$ is a parameter determined by both the real structure of the atom and the characteristics of the AIS. Also scaled is the PES:

$$P_1=\gamma_0W_1,\quad P_2=\gamma_0W_2\ , \tag{23}$$

and we will hereafter refer to $P_1$ and $P_2$ as the low and the high PES, respectively.

We plot the population $P_0(t)$ of the ground state in Fig. 2, with the coefficients to be $\xi=0.01$, $\gamma=\gamma_0$, and $\Delta=0$. When the laser peak intensity is high, oscillations occur in $P_0(t)$. The higher the peak intensity is, the more the oscillation number in the time range $t\le 0$ is. The oscillations are similar to the so-called Rabi oscillation [11], a characteristic strong-field effect in autoionization, for time-independent cw laser irradiation (corresponding to $\gamma=\infty$ in our model, i.e., neglecting the effects of the exponentially growing part of the laser pulse). If the peak intensity is low, the oscillations are quenched and $P_0(t)$ exhibits a decaying behavior.

In Fig. 3 is shown the long-time low PES $P_1(\omega_1)$ with the same coefficients as in Fig. 2. Different curves correspond to the different laser peak intensities, related to those in Fig. 2. For the sake of clarity, the curve of $\eta=31.4$ is not drawn in Fig. 3, because it is too close to the curve of $\eta=15.7$ to be distinguished. Similar to the results obtained by Rzążewski and co-workers [3] for the case of infinite Fano parameter (corresponding to the case

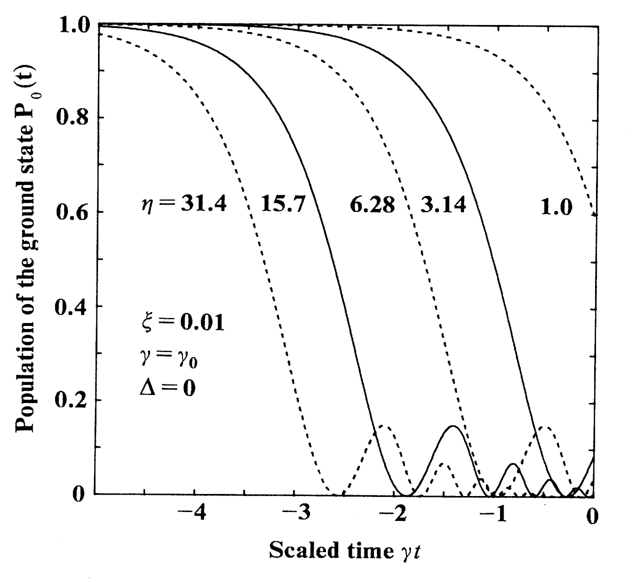


FIG. 2. The ground-state population as a function of scaled time $\gamma t$. Both the population and the scaled time are dimensionless. The coefficients $(\xi,\gamma,\Delta)$ and the laser peak intensities (scaled as $\eta$) are indicated in the figure. $\xi$, $\gamma$, $\Delta$, and $\eta$ are defined in the text.

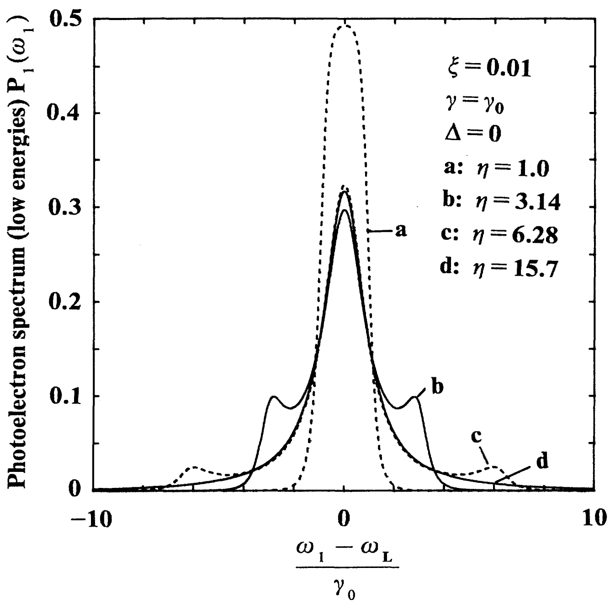


FIG. 3. The long-time low photoelectron spectrum for the same laser peak intensities (except $\eta=31.4$) and the same coefficients as in Fig. 2, where the photoelectron energies are scaled by the dimensionless quantity $(\omega_1-\omega_L)/\gamma_0$. The curve corresponding to $\eta=31.4$ is not drawn in the figure, for it is very close to the curve corresponding to $\eta=15.7$.

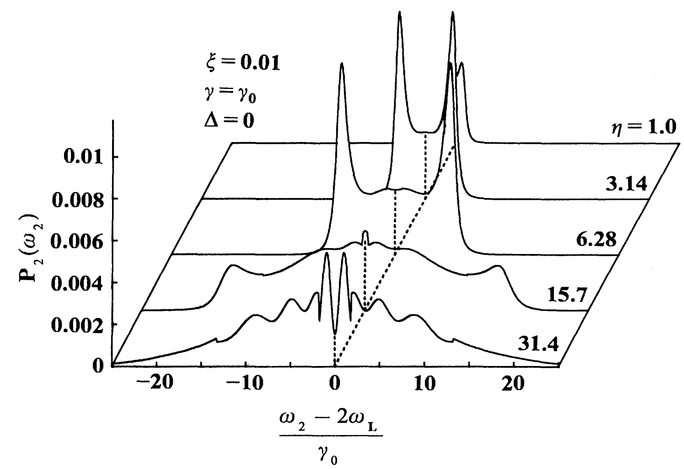


FIG. 4. The long-time high photoelectron spectrum for the same laser peak intensities and the same coefficients as in Fig. 2, where the photoelectron energies are scaled by the dimensionless quantity $(\omega_2-2\omega_L)/\gamma_0$. Evident multipeak splitting is shown.

$\xi=0$ here), $P_1(\omega_1)$ does not show evident multipeak splitting (see also the last paragraph in this section). This may be mostly because the laser pulse here is composed of an exponentially growing part and a flat-top cw part. In the case of strong-field autoionization induced by such a smooth laser pulse as, say, the hyperbolic secant one, evident multipeak splitting will occur in $P_1(\omega_1)$, which we will discuss elsewhere.

The long-time high PES $P_2(\omega_2)$ is shown in Fig. 4, with the same peak-intensities and the same coefficients as in Fig. 2. The structure of $P_2(\omega_2)$ is more complex than that of $P_1(\omega_1)$ (see Fig. 3), which is a common phenomenon in strong-field autoionization with both the time dependence of the laser irradiation and the high order ionization processes such as the photoionization of the AIS considered here. If the time dependence of the laser pulse were not taken into account, one would get the same structure of $P_2(\omega_2)$ as that of $P_1(\omega_1)$, because we consider here only the case of infinite Fano coefficients [14]. And if the high-order ionization were neglected, $P_2(\omega_2)$ would always be zeroness.

Clearly shown in Fig. 4 is the evident multipeak splitting under high peak intensities. And the number of peaks increases with the peak intensity. Recalling that the laser pulses in our calculations are exponentially growing cw ones, we can get the so-called pulse area [2–4] in $t\leq 0$: $A=2\eta\gamma_0/\gamma$. Therefore the number of peaks increases with the pulse area $A$. One can compare the results here with the corresponding ones obtained by Rzążewski *et al.* in strong-field autoionization [2–4] (i.e., the case $\xi=0$) and those obtained by Rogus and Lewenstein in multiphoton resonant ionization [21] by smooth laser pulses. Comparing Figs. 4 and 3 we can see, because the high-order ionization processes are taken into account, that the multipeak splitting is more evident in the high spectrum than in the low spectrum. This may be understood from the following two facts. First, although the photoionization of the AIS is a considerably large relaxation, there still exist Rabi oscillations in the ground-state population (Fig. 2). Second, the high spectrum in our model results from the photoionization of the AIS, which is produced by the time-dependent laser excitation of the AIS. From the previous study on the strong-field autoionization, Rabi oscillation and the Fourier transform of the AIS amplitude $u_a$ [see (4)] are closely related with the multipeak structure in PES [2–6]. However, a physically simple explanation of this structure still remains an open problem. A much more detailed study of the multipeak structure in the strong-field autoionization, with the high-order ionization processes included, is required.

It is shown that in Figs. 3 and 4 that there are two kinds of peaks in the long-time PES. One kind is the nearly elastic part and is located at small photoelectron energies, mainly formed in the time range $t\leq 0$. And it is in this range that the multipeak structure is produced. Obviously this structure can only be related with the time-dependence of the laser field. The other kind is located near the energies of the dressed states in strong-field autoionization induced by a time-independent cw laser irradiation [11,15], and is formed in the flat-top part of the laser irradiation in $t>0$. The laser field in this part tends to smear the multipeak structure in the photoelectron spectrum and produce the photoelectrons in the dressed states. When the peak intensity is low, the two kinds of peaks can hardly be distinguished from each other. When the peak intensity is high, they separate from each other. With the growing peak intensity for the same coefficients, more and more atoms will be ionized in $t \leq 0$, and less atoms will remain in the bound states $|0\rangle$ and $|a\rangle$ at the time $t=0$ (see Fig. 2). The higher the peak intensity is, the less importance the second kind of peaks is of. For example, in the cases $\eta=15.7$ and $\eta=31.4$, the second kind of peaks in Fig. 3 (located outside the figure) is too low to be shown clearly.

## IV. CONCLUSIONS

In summary, we have presented the first results, to our knowledge, of strong-field autoionization induced by smooth laser pulses with the photoionization of an autoionizing state included. The equations given are suitable for numerical study for arbitrary laser pulses. Analytical results, however, may be obtained for some special pulses which include the important pulses composed of exponentially time-dependent fields. For the sake of illustration, the results of the population of the ground state and the photoelectron spectrum are discussed for an exponentially switched-on cw pulse. We have compared our results with the previous ones and find that more evident multipeak splitting is shown in the high photoelectron spectrum resulted from the high-order ionization processes.

Generally speaking, strong-field autoionization induced by a smooth laser pulse is difficult to derive analytical expressions. And it becomes far more complicated when we introduce further the photoionization of the AIS. This paper, as a first attempt to tackle this latter problem, studies the model with both the Fano parameters related, respectively, to $|0\rangle$ and $|\omega_2\rangle$ to be infinite. Strong-field autoionization involving the photoionization of the AIS induced by pulses such as hyperbolic secant ones, with general asymmetric profiles of the autoionization resonances, remains to be investigated in future.

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