Laser-induced autoionization with pulsed excitation

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A theoretical study of laser-induced autoionization of an atomic system in interaction with a time-dependent excitation is presented. Interference effects between the two ionization channels of the atom are found to be responsible for coherent phenomena, such as the enhancement of the photon yield, for a pulsed excitation. An analysis of the pulse-shape effects shows that pumping mechanisms of the proper dressed state, occurring during the rise time of flat-top pulses, can result in an enhancement of the photon yield. An adiabatic approximation based on the use of time-dependent complex dressed states is shown to be adequate to describe the interaction with smooth pulses.

I. INTRODUCTION

Atomic bound states lying above the ionization threshold and imbedded in a continuum of free states have proved useful in the enhancement of multiphoton processes involving the continuum, such as generation of vacuum ultraviolet radiation[1,2] and multiphoton ionization.[3,4] Since these levels can be very energetic, they are also candidates for the production of extreme ultraviolet radiation.[5]

In recent years there has been considerable theoretical interest in the study of strong-field laser-induced autoionization (LIA) of an atomic system.[6–10] New coherent phenomena have been predicted as a result of the constant-field interference between the two ionization channels (autoionization and photoionization) of the atom. The most interesting of these phenomena are the appearance of the *confluence of coherences* in the electron spectrum,[7] the enhancement of photoemission spectra,[9] and the trapping of the atomic population.[6] The possibility of inducing structures in the continuum using an intense radiation field has also attracted theoretical[11,12] and experimental[13,14] attention. It is worth pointing out that the same kind of quantum-mechanical interferences encountered in LIA and laser-induced continuum structure (LICS) are exploited in the recent proposal of Harris of lasers without inversion.[15] In fact, there is a precedent to this case: Arkhipin and Heller[16] have shown that a discrete state which is imbedded in a continuum of free states exhibits a Fano profile in absorption but not in emission, suggesting that laser emission without population inversion could be possible.

In all the cited theoretical studies, the common assumption has been a sudden turn on of the laser field which then remains constant for all the interaction time. The case of pulsed excitation has received much less attention, even though the question of whether quantum-interference effects can play an important role with smooth laser pulses is of fundamental importance from an experimental viewpoint. In fact, the use of a quasimetastable core-excited level[17] as initial state and the possibility of probing autoionizing states with high-power, narrow-band dye-laser radiation[18,19] stimulates the study of intense-field LIA with pulsed excitation. To our knowledge, the only published paper on LIA dealing with smooth pulses is Ref. 10. However, in this work the very limit case of pulse duration equal to the autoionization lifetime has been considered.

In a very recent paper,[20] Buffa and Spong have presented an approach that appears suitable to the study of LIA with time-dependent excitation. Following this approach, the present work focuses on the pulse-shape effects on the enhancement of the photon yield in LIA. In view of an experimental investigation of the process, a major motivation for this work is to point out whether quantum-interference effects can play an important role with smooth laser pulse excitation.

Following a description of the physical model in Sec. II, we present in Sec. III an analytical expression for the absorption cross section valid in the weak-field regime. In Sec. IV we discuss the effects of the rise time of flat-top pulses on the photon yield, describing the results in a dressed-state framework. Finally, in Sec. V, we study the interaction with a smooth pulse, presenting an adiabatic approach based on the use of time-dependent complex dressed states.

II. THEORETICAL FRAMEWORK

Figure 1 shows a schematic diagram of our model. A laser pulse of electric field envelope $E(t)$ and central frequency ω_L couples the initial state $|1\rangle$ with an excited bound state $|2\rangle$ and with a continuum of free states $|\omega\rangle$ of an atomic system. Due to the Coulombic interaction with the continuum $|\omega\rangle$, state $|2\rangle$ can autoionize, emitting an electron of energy $\hbar\omega_e=\hbar\omega-\hbar\omega_{\rm th}$. Moreover, state $|2\rangle$ can radiate to a third atomic level $|3\rangle$ emitting a photon of energy $\hbar\omega_{\rm ph}$. According to Ref. 20, in the as-

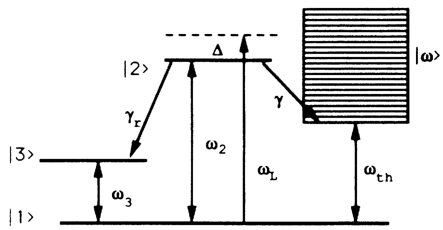

FIG. 1. Schematic diagram of the atomic model.

sumption of flat background, the electron and photon spectra can be expressed, respectively, as

$$P_e(\omega_e)=\frac{\gamma}{2\pi}\left|\int_{-\infty}^{+\infty}dt\{[\Omega(t)/q\gamma]\times a_1\exp[i(\omega-\omega_L)t]-a_2\exp[i(\omega-\omega_2)t]\}\right|^2 , \quad (1)$$

$$P_{\rm ph}(\omega_{\rm ph})=\frac{\gamma_r}{2\pi}\left|\int_{-\infty}^{+\infty}dt\{a_2\exp[i(\omega_{\rm ph}-\omega_2+\omega_3)t]\}\right|^2 . \quad (2)$$

Here a_1 and a_2 are the probability amplitudes of states $|1\rangle$ and $|2\rangle$, satisfying the coupled differential equations[20]

$$\frac{da_1}{dt}=[\Omega(t)/2q](1+iq)a_2\exp[i(\omega_L-\omega_2)t]-[\Omega^2(t)/2q^2\gamma]a_1 , \quad (3a)$$

$$\frac{da_2}{dt}=[\Omega(t)/2q](1+iq)a_1\exp[i(\omega_2-\omega_L)t]-[(\gamma+\gamma_r)/2]a_2 . \quad (3b)$$

$\hbar\omega_2$ and $\hbar\omega_3$ are the energies of the bound states $|2\rangle$ and $|3\rangle$, q is the Fano parameter,[21] γ and γ_r are, respectively, the autoionization and radiative decay rates of state $|2\rangle$ and $\Omega(t)=d_{12}E(t)/\hbar$ is the instantaneous Rabi frequency of the $|1\rangle-|2\rangle$ transition (d_{12}: electric dipole moment).

The electron and photon yields are then given by

$$P_e=\int_0^{+\infty}d\omega_e P_e(\omega_e)=\gamma\int_{-\infty}^{+\infty}dt|\chi(t)a_1-a_2\exp(i\Delta t)|^2 , \quad (4)$$

$$P_{\rm ph}=\int_0^{+\infty}d\omega_{\rm ph}P_{\rm ph}(\omega_{\rm ph})=\gamma_r\int_{-\infty}^{+\infty}dt|a_2|^2 , \quad (5)$$

where $\Delta=\omega_L-\omega_2$ and $\chi(t)=\Omega(t)/q\gamma$.

For a constant laser field, Eqs. (3) are analytically solvable[20] and (1) and (2) reproduce the spectra obtained by Agarwal *et al.*[8,9] by a master equation formalism. The main results can be summarized as follows.

(i) For $\gamma_r=0$, the electron spectrum shows a prominent feature, becoming sharper when approaching the confluence of coherences point[7] defined by

$$\Delta=(\chi^2-1)q\gamma/2 . \quad (6)$$

When (6) is satisfied, the atomic population is trapped in the bound states $|1\rangle$ and $|2\rangle$ at the values

$$|a_1|^2=1/[4(1+\Delta/q\gamma)^2] , \quad (7a)$$

$$|a_2|^2=(1+2\Delta/q\gamma)/[4(1+\Delta/q\gamma)^2] . \quad (7b)$$

(ii) For $\gamma_r\neq0$, even for relatively small values of the radiative decay rate, i.e., $\gamma_r/\gamma\ll1$, the photon yield, as a function of Ω and Δ, has a very sharp feature around the confluence of coherences point (6).[9]

A completely different situation is represented by a pulsed excitation. Equation (6), in general, can no longer be satisfied for all the interaction time, and the question arises whether a time-dependent interference between the two ionization channels of the atom can result in coherent phenomena such as, for instance, the enhancement of the photon yield.

III. TIME-DEPENDENT EXCITATION: WEAK-FIELD REGIME

Before proceeding with a numerical solution of (3) and (5) for various laser parameters and pulse shapes, we present an analytical expression for the absorption cross section for a weak, smooth laser pulse in the limit of negligible radiative damping γ_r.

For a pulsed excitation, the absorption cross section $\sigma(\Delta)$ can be defined as

$$\sigma(\Delta)=1-|a_1(+\infty)|^2 . \quad (8)$$

In a perturbative treatment in the laser field, we can develop a_1 as $a_1\approx1+a_1^{(1)}$, with $|a_1^{(1)}|^2\ll1$, to obtain

$$\sigma(\Delta)\approx-[a_1^{(1)}(+\infty)+{\rm c.c.}] , \quad (9)$$

with

$$a_1^{(1)}(+\infty)\approx(\gamma/2)\int_{-\infty}^{+\infty}dt[\chi(t)(1+iq)a_2\exp(i\Delta t)-\chi^2(t)], \quad (10)$$

and where

$$a_2\approx(\gamma/2)[\chi(t)(1+iq)/(\gamma/2-i\Delta)]\exp(-i\Delta t) \quad (11)$$

is the solution of (3b) in the hypothesis of a sufficiently smooth laser pulse (i.e., $d\Omega/dt\ll\gamma\Omega$). The combination of (9), (10), and (11) leads to

$$\sigma(\Delta)\approx\gamma(\Delta+q\gamma/2)^2/[\Delta^2+(\gamma/2)^2]\int_{-\infty}^{+\infty}dt\chi^2(t) , \quad (12)$$

which represents an asymmetric Fano profile,[21] showing that in the weak-field regime a time-dependent interaction can result in observable interference effects.

IV. STRONG-FIELD REGIME: PULSE-SHAPE EFFECTS

Equations (3) have been numerically integrated for different laser pulse shapes and parameters, using a Runge-Kutta algorithm. In Fig. 2(a) the photon yield versus the laser-pulse rise time τ is reported for a time-

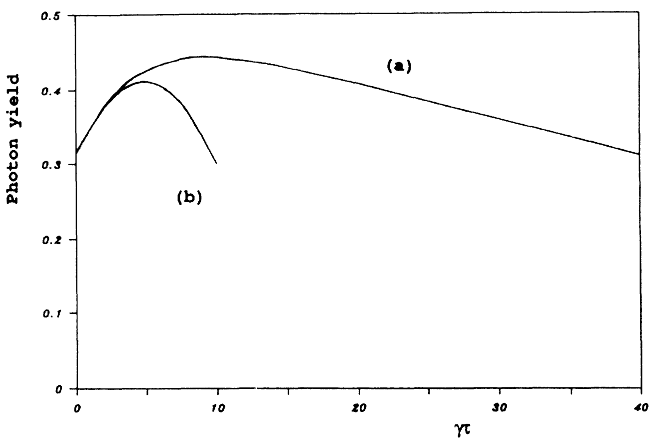

FIG. 2. Photon yield vs pulse rise time τ for the flat-top laser pulse (13). The data refer to the situation $q=1$, $\gamma_r/\gamma=0.01$, $\Delta=0$, $\gamma T=200$, and $\Omega_0=q\gamma$. (a): numerical calculation; (b) sudden approximation to the second order.

dependent Rabi frequency $\Omega(t)$ given by

$$\Omega(t)=\begin{cases}\Omega_0\sin^2[\pi(t+\tau)/2\tau, & t\in[-\tau,0]\\ \Omega_0, & t\in[0,T]\\ \Omega_0\sin^2[\pi(t-T-\tau)/2\tau], & t\in[T,T+\tau]\end{cases} \tag{13}$$

and equal to zero elsewhere. The data refer to the situation $q=1$, $\gamma_r/\gamma=0.01$, $\Delta=0$, $\gamma T=200$, and $\Omega_0=q\gamma$, satisfying the trapping condition (6). The enhancement of the photon yield with a laser pulse of rise time different from zero is at first glance surprising, since the trapping condition is never satisfied during the rise-time and fall-time intervals. However, a straightforward explanation is provided in the dressed-state basis in terms of a sudden approximation.

For a constant Rabi frequency Ω_0, in terms of the variables

$$\alpha=(\cos\delta)a_1-(\sin\delta)a_2\exp(i\Delta t) , \tag{14a}$$

$$\beta=(\sin\delta)a_1+(\cos\delta)a_2\exp(i\Delta t) , \tag{14b}$$

with $tg(2\delta)=\Omega_0/\Delta$, Eqs. (3) become

$$\frac{d\alpha}{dt}=[(i\lambda_\alpha-\gamma_\alpha-(\sin\delta)^2\gamma_r/2]\alpha+[W+(\cos\delta)^2\gamma_r/2]\beta , \tag{15a}$$

$$\frac{d\beta}{dt}=[(i\lambda_\beta-\gamma_\beta-(\cos\delta)^2\gamma_r/2]\beta+[W+(\sin\delta)^2\gamma_r/2]\alpha , \tag{15b}$$

with

$$\begin{aligned}\lambda_\alpha&=\frac{\Delta}{2}\{1-[1+(\Omega_0/\Delta)^2]^{1/2}\} ,\\ \lambda_\beta&=\frac{\Delta}{2}\{1+[1+(\Omega_0/\Delta)^2]^{1/2}\} ,\\ \gamma_\alpha&=(\chi\cos\delta+\sin\delta)^2\gamma/2 ,\\ \gamma_\beta&=(\cos\delta-\chi\sin\delta)^2\gamma/2 ,\\ W&=(\chi\cos\delta+\sin\delta)(\cos\delta-\chi\sin\delta)\gamma/2 .\end{aligned} \tag{16}$$

When (6) is satisfied then either $\gamma_\alpha=W=0$ and $\gamma_\beta=\gamma(1+\chi^2)/2$ (if $\Delta<0$) or $\gamma_\beta=W=0$ and $\gamma_\alpha=\gamma(1+\chi^2)/2$ (if $\Delta\geq 0$). For $\gamma t\gg 1$, this leads to the following solutions for (15):

$$\alpha\approx\alpha(0)\exp\{[i\lambda_\alpha-(\sin\delta)^2\gamma_r/2]t\} , \tag{17a}$$

$$\beta\approx 0 \tag{17b}$$

if $\Delta<0$, and

$$\alpha\approx 0 , \tag{18a}$$

$$\beta\approx\beta(0)\exp\{[i\lambda_\beta-(\cos\delta)^2\gamma_r/2]t\} \tag{18b}$$

if $\Delta\geq 0$. The corresponding expressions for the excited-state population are

$$|a_2|^2=(\sin\delta)^2|\alpha(0)|^2\exp[-(\sin\delta)^2\gamma_r t] \tag{19a}$$

if $\Delta<0$ and

$$|a_2|^2=(\cos\delta)^2|\beta(0)|^2\exp[-(\cos\delta)^2\gamma_r t] \tag{19b}$$

if $\Delta\geq 0$. Introducing (19) in (5), and neglecting the number of photons emitted during the rise time and fall time of (13), the expressions of the photon yield for a flat-top pulse become

$$P_{\rm ph}\approx|\cos\delta a_1(0)-\sin\delta a_2(0)|^2\{1-\exp[-(\sin\delta)^2\gamma_r T]\} \tag{20a}$$

for $\Delta<0$, and

$$P_{\rm ph}\approx|\sin\delta a_1(0)+\cos\delta a_2(0)|^2\{1-\exp[1-(\cos\delta)^2\gamma_r T]\} \tag{20b}$$

for $\Delta\geq 0$, showing that the photon yield is proportional to the population in one dressed state at time $t=0$.

The effect of the pulse rise time is a transfer of population from the initial state $|1\rangle$ to a dressed state. In a two-level system without losses, this transfer of population is equal to 0.5 for a resonant pulse with a sudden rise time, and equal to 1 for a pulse with an adiabatic rise-time. In our case, if the pumping mechanism exceeds the losses, due to the ionization of the atom, the total result is an enhancement of the photon yield, as shown in Fig. 2(a).

The quantities $a_1(0)$ and $a_2(0)$ that appear in (20) can be easily numerically computed from (3). However, for short rise times, they can be evaluated by using the sudden approximation.[22] For the risetime of the resonant pulse given by (13), the sudden approximation to the second order provides for the photon yield (20b) curve (b) in Fig. 2.

V. STRONG-FIELD SMOOTH PULSE EXCITATION

In Figs. 3(a) and 3(b) the photon yield (5) is reported as a function of the peak Rabi frequency Ω_0 and detuning Δ

for two different pulse shapes given by

$$\Omega_1(t)=\begin{cases}\Omega_0, & \gamma t\in[0,40]\\ 0 & \text{elsewhere}\ ,\end{cases} \tag{21}$$

$$\Omega_2(t)=\Omega_0\text{sech}(\gamma t/20)\ . \tag{22}$$

The two laser pulses have the same energy; i.e.,

$$\int_{-\infty}^{+\infty}dt\,\Omega_1^2(t)=\int_{-\infty}^{+\infty}dt\,\Omega_2^2(t)\ .$$

The data are the result of numerical calculations performed with $q=1$ and $\gamma_r/\gamma=0.01$. Similarly to the case of cw excitation, the sudden pulse (21) shows an enhancement of the photon yield for both positive and negative values of the detuning Δ, reflecting a constant-field trapping of population during the interaction time. Also for the smooth pulse (22) the photon yield shows an enhancement for appropriate values of Ω_0 and Δ, and its value can even exceed the value obtained with the sudden pulse excitation. This shows that laser-induced interference effects play a fundamental role also for a smooth pulse excitation. However, in this case, the *trappinglike* behavior occurs only for negative values of Δ. This result can be easily explained in terms of an adiabatic approximation based on the use of time-dependent *complex dressed states* (CDS).

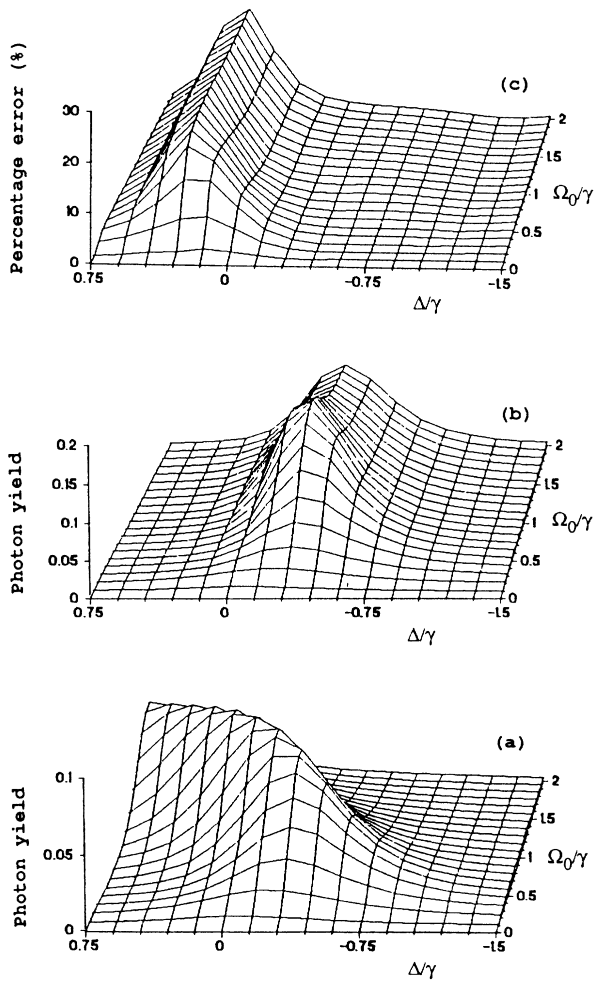

FIG. 3. Photon yield as a function of the peak Rabi frequency Ω_0 and detuning Δ for (a) the sudden laser pulse (21), (b) the smooth laser pulse (22), and (c) percentage error between numerical values (b) and data obtained with the adiabatic approximation (27b). The data refer to the situation $q=1$ and $\gamma_r/\gamma=0.01$.

Following Ref. 20, we introduce two new variables:

$$A=(\cos\theta)a_1-(\sin\theta)a_2\exp(i\Delta t)\ , \tag{23a}$$

$$B=(\sin\theta)a_1+(\cos\theta)a_2\exp(i\Delta t)\ , \tag{23b}$$

where the complex angle θ is defined by

$$\tan(2\theta)=\tilde{\Omega}/\tilde{\Delta}\ ,$$

with

$$\tilde{\Omega}=\Omega(1-i/q)\ ,$$

$$\tilde{\Delta}=\Delta+i[\gamma(1-\chi^2)+\gamma_r]/2\ .$$

The new variables (23) satisfy the following differential equations:

$$\frac{dA}{dt}=\lambda_A A-\left[\frac{d\theta}{dt}\right]B\ , \tag{24a}$$

$$\frac{dB}{dt}=\lambda_B B+\left[\frac{d\theta}{dt}\right]A\ , \tag{24b}$$

with

$$\lambda_A=i(\Delta-\Omega_R')/2-[(1+\chi^2)\gamma/2+\gamma_r/2-\Omega_I']/2$$
$$=i\omega_A-\gamma_A/2\ , \tag{25a}$$

$$\lambda_B=i(\Delta+\Omega_R')/2-[(1+\chi^2)\gamma/2+\gamma_r/2+\Omega_I']/2$$
$$=i\omega_B-\gamma_B/2\ , \tag{25b}$$

and where Ω_R' and Ω_I' are the real and imaginary parts, respectively, of the complex generalized Rabi frequency defined as

$$\Omega'=\tilde{\Delta}[1+(\tilde{\Omega}/\tilde{\Delta})^2]^{1/2}\ .$$

The adiabatic approximation consists in neglecting the terms in $d\theta/dt$ in (24) and then writing the solutions of (24) as

$$A(t)\approx A(t_i)\exp\left[\int_{t_i}^{t}\lambda_A dt'\right]\ , \tag{26a}$$

$$B(t)\approx B(t_i)\exp\left[\int_{t_i}^{t}\lambda_B dt'\right]\ , \tag{26b}$$

with t_i the initial time. The condition of validity for the adiabatic approximation is given by

$$\left|\frac{d\theta}{dt}\right|\ll|\Omega'|\ .$$

If the atom is initially in the ground state, both CDS's are populated with the sudden pulse (21), being $A(0)=\cos\theta(0)$ and $B(0)=\sin\theta(0)$, but only one CDS is populated with the smooth pulse (22), being $A(-\infty)=1$ and $B(-\infty)=0$. Combining (5), (23), (25), and (26), the following expressions for the photon yield are obtained, with the sudden pulse:

$$P_{\rm ph} \approx \gamma_r |\sin\theta\cos\theta|^2 \times \int_0^{+\infty} dt \,|\{\exp[(i\omega_B - \gamma_B/2)t] - \exp[(i\omega_A - \gamma_A/2)t]\}|^2 , \quad (27a)$$

and with the smooth pulse:

$$P_{\rm ph} \approx \gamma_r \int_{-\infty}^{+\infty} dt \left[|\sin\theta|^2 \exp\left(-\int_{-\infty}^{t} \gamma_A dt' \right) \right] . \quad (27b)$$

The time-dependent damping rates γ_A and γ_B of the CDS are strongly dependent on the instantaneous Rabi frequency and on the detuning. In general, one CDS is more stable for $\Delta > 0$ and the other one is more stable for $\Delta < 0$. Since with the sudden pulse (21) both CDS's are populated, an enhancement of the photon yield (27a) is obtained when either $\gamma_A/\gamma \ll 1$ or $\gamma_B/\gamma \ll 1$. On the contrary, since with the smooth pulse (22) only one CDS is populated, an enhancement of the photon yield is obtained only when $\gamma_A/\gamma \ll 1$, and this is possible only for $\Delta < 0$. In Fig. 3(c) the percentage error between data obtained using (27b) and the numerical values of Fig. 3(b) is reported, showing that the interaction can be considered adiabatic every time an enhancement of the photon yield is obtained.

VI. CONCLUSION

We have presented a theoretical study of laser-induced autoionization of an atomic system in interaction with a laser pulse, showing that interference effects between the two ionization channels of the atom play a fundamental role in the interaction dynamics in the case of time-dependent excitation. An analysis of the pulse-shape effects shows that pumping mechanisms of the proper dressed state, occurring during the rise time of flat-top laser pulses, can result in an enhancement of the photon yield. An adiabatic approximation, used in the framework of time-dependent complex dressed states, has proved to be adequate to describe the interaction with smooth pulses.

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