Resonant multiphoton ionization induced by pulsed excitation

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The object of this paper is to study the influence of the characteristics of the excitation pulse on resonant multiphoton ionization. A classical description of the field is introduced in the exactly solvable model initially developed by Beers and Armstrong. A detailed solution is given for square pulses. The case corresponding to a highly excited resonant level is particularly studied both analytically for square pulses and numerically for different amplitude-varying shapes.

In a recent paper, Beers and Armstrong[1] developed an exactly solvable model for describing resonant multiphoton ionization processes. They give the ionization probability as a function of time, light intensity, and a few atomic parameters. This result allows them to point out the influence of the characteristics of the excitation field (light intensity and duration) on the ionization cross section near a resonance. The recent development of experiments using picosecond pulses[2] shows the great interest in such a calculation. However, this model utilizes a quantum description of the driving field, and it is well known that this approach is not convenient for characterizing fields with an amplitude varying in time and thus for taking into account the influence of the excitation pulse shape.

We have previously adapted, in a particular case, the model proposed by Beers and Armstrong, by using a classical description of the driving field. An analytical result was given for square pulses. Numerical calculations have been performed for different pulse shapes. We have demonstrated the influence of the pulse shape on the characteristics of the resonant curves.[3]

The object of this paper is to show that, starting from the same basic assumption, one can use, in the general case, a classical approach for the driving field. This allows us not only to obtain the same results as Beers and Armstrong for a square pulse, but also to study the influence of the pulse shape by performing numerical calculations.

We first derive the basic equations (Sec. I) and give the exact solution for a square pulse, in the general case, without any restrictive assumption concerning the relative importance of the different processes appearing in the description of resonant multiphoton ionization (Sec. II). The result is rather complicated and could be useful only in very rare cases when the two steps of the resonant process have similar transition probabilities. We have analyzed, in greater detail, the case of a weak first step and a strong second step; the direct process is always assumed to be weak. As in our previous paper, we derive a simple result and discuss the analytical formula for a square pulse (Sec. III). We study the influence of the pulse shape by looking at the results of numerical integration of the basic equations (Sec. IV). Finally, we apply our result to the case of experiments on Cs.

I. BASIC EQUATIONS

Exactly as in the model introduced by Beers and Armstrong, we describe the multiphoton ionization near a resonance in the following way: (a) A nonresonant n-photon process is induced by an effective interaction H_n^E, which is obtained from perturbation theory to nth order; and (b) a two-step process occurs, where first there is a p-photon transition from the ground state i to the quasiresonant state φ (this first excitation is induced by an effective interaction H_p), then a $(n-p)$-photon transition from φ to a continuum state ψ_E occurs (this second excitation is induced by an effective interaction H_{n-p}^E). For reasons of simplicity, all the considered states are assumed to be nondegenerate and only one continuum is introduced in this model. Since we are essentially interested in the description of short light pulses, we have omitted the relaxation phenomena such as spontaneous emission or collisions which are characterized by times longer than the duration of the pulses of interest. Under these conditions, if one uses a classical description of the driving field, the Hamiltonian can be written $H = H_A + H_{AF}$ where H_A is a reduced atomic Hamiltonian and H_{AF} an effective interaction between the atom and the field. H_A can be written as

$$H_A = \hbar\Omega_i |i\rangle\langle i| + \hbar\Omega_\varphi |\varphi\rangle\langle\varphi| + \int_I^\infty E |\psi_E\rangle\langle\psi_E| dE \ , \tag{1}$$

where $\hbar\Omega_i$ and $\hbar\Omega_\varphi$ are the energies of the states i and φ, possibly light-shifted by nonresonant processes, and I is the ionization energy of the atom.

If $\omega/2\pi$, $\mathcal{E}(t)$, and $\varphi(t)$ are, respectively, the frequency, the amplitude, and the phase of the field (these last two quantities varying slowly with respect to ω), and if we keep only the energy-conserving terms in the Hamiltonian (rotating-wave approximation), H_{AF} reduces to

$$H_{AF} = |\varphi\rangle\langle\varphi|H_p|i\rangle\langle i| + \int_I^\infty |\psi_E\rangle\langle\psi_E|H^E_{n-p}|\varphi\rangle\langle\varphi|dE + \int_I^\infty |\psi_E\rangle\langle\psi_E|H^E_n|i\rangle\langle i|dE + \text{c.c.}, \quad (2)$$

$$\langle\varphi|H_p|i\rangle = \hbar K \exp\{ip[\omega t+\varphi(t)]\} = \hbar\kappa\mathcal{E}^p\exp\{ip[\omega t+\varphi(t)]\},$$

$$\langle\psi_E|H^E_{n-p}|\varphi\rangle = \hbar L_E \exp\{i(n-p)[\omega t+\varphi(t)]\} = \hbar\lambda\mathcal{E}^{n-p}\exp\{i(n-p)[\omega t+\varphi(t)]\},$$

$$\langle\psi_E|H^E_n|i\rangle = \hbar J_E\exp\{in[\omega t+\varphi(t)]\} = \hbar\gamma\mathcal{E}^n\exp\{in[\omega t+\varphi(t)]\}, \quad (3)$$

where κ, λ, and γ are atomic parameters which are assumed to be real. The wave function can be written as

$$|\Psi(t)\rangle = a_i\exp(-i\Omega_i t)|i\rangle + a_\varphi\exp[-i(\Omega_i+p\omega)t]|\varphi\rangle + \int_I^\infty dE\, a_E\exp[-i(\Omega_i+n\omega)t]|E\rangle. \quad (4)$$

The equations of motion for the wave-function components become

$$i\dot{a}_i = Ka_\varphi + \int_I^\infty dE\, J_E a_E, \quad (5a)$$

$$i(\dot{a}_\varphi - i\delta a_\varphi) = Ka_i + \int_I^\infty dE\, L_E a_E, \quad (5b)$$

$$i(\dot{a}_E - i\delta_E a_E) = J_E a_i + L_E a_\varphi, \quad (5c)$$

with

$$\delta = \Omega_i - \Omega_\varphi + p[\omega+\dot{\varphi}(t)],$$

$$\delta_E = \Omega_i - E/\hbar + n[\omega+\dot{\varphi}(t)].$$

The total number of produced ions is given by

$$N = P\int_I^\infty dE\,|a_E|^2, \quad (6)$$

where P is the number of atoms interacting with the field.

II. RESOLUTION OF THE EQUATIONS: GENERAL CASE

The only difficulty encountered in solving this system of equations comes from integration over energy. One can assume that J_E and L_E do not vary rapidly with E. The Fourier spectrum of a_i and a_φ has an extension of the order of K, J^2_E, or L^2_E, which are the characteristic frequencies of the atom-field interaction. One can consider J_E and L_E as constant over a range of variation of the order of K, J^2_E, or L^2_E. Moreover, if $n\omega+\Omega_i-I$ is much larger than the mean value of K, J_E, and L_E, the integration range can be extended to $-\infty$ without introducing significant errors. With these approximations, which are valid of course in most realistic cases, the solution corresponding to square-pulse excitation,

$$K=\bar{K},\quad J_{E_0}=\bar{J},\quad L_{E_0}=\bar{L},\quad \text{if } 0\le t\le T, \qquad K=J_{E_0}=L_{E_0}=0,\quad \text{if } t<0 \text{ and } t>T, \quad (7)$$

can be obtained by the following method:

In Ref. 3 we first resolved the equations and then integrated over energy. Here we integrate over energy first. We must calculate the two terms

$$\int_{-\infty}^{+\infty} L_E\,dE\int_{-\infty}^{t}[J_E a_i(t')+L_E a_\varphi(t')]\times\exp[i\delta_E(t-t')]\,dt'$$

and

$$\int_{-\infty}^{+\infty} J_E\,dE\int_{-\infty}^{t}[J_E a_i(t')+L_E a_\varphi(t')]\times\exp[i\delta_E(t-t')]\,dt'.$$

The Fourier transform of a term such as

$$\int_{-\infty}^{+\infty} L_E\,dE\int_{-\infty}^{t} L_E a_\varphi(t')\,dt'\exp[i\delta_E(t-t')] \quad (8a)$$

is

$$\left(L^2_E+\frac{i}{\pi}\mathcal{P}\int\frac{L^2_{E'}}{E-E'}dE'\right)\mathcal{A}_\varphi(E), \quad (8b)$$

where $\mathcal{A}_\varphi(E)$ is the Fourier transform of $a_\varphi(t)$ and $\mathcal{P}$ stands for the principal part. By looking at the extension of $\mathcal{A}_\varphi(E)$, one can replace E by $E_0=\hbar\Omega_i+n\hbar\omega$ in the term between brackets. Expression (8a) becomes

$$\left(L^2_{E_0}+\frac{i}{\pi}\mathcal{P}\int\frac{L^2_{E'}}{E_0-E'}dE'\right)a_\varphi(t). \quad (8c)$$

We then derive a new set of equations:

$$i\dot{a}_i = \Delta^J a_i + \tilde{K}a_\varphi - i(J^2_{E_0}a_i + L_{E_0}J_{E_0}a_\varphi),$$
$$i\dot{a}_\varphi = (\Delta^L-\delta)a_\varphi + \tilde{K}a_i - i(J_{E_0}L_{E_0}a_i + L^2_{E_0}a_\varphi), \quad (9)$$

where

$$\Delta^J = \frac{1}{\pi}\mathcal{P}\int\frac{J^2_{E'}}{E_0-E'}dE',$$

$$\Delta^L = \frac{1}{\pi}\mathcal{P}\int\frac{L^2_{E'}}{E_0-E'}dE',$$

$$\tilde{K} = K+\frac{1}{\pi}\mathcal{P}\int\frac{L_{E'}J_{E'}}{E_0-E'}dE',$$

which can be solved in a straightforward way. The total number of produced ions is given finally by

$$N=P\left\{1-\frac{1}{A^2}\left[\frac{\tilde{\delta}^2}{4}+\tilde{K}^2+\left(\frac{\bar{L}^2+\bar{J}^2}{2}\right)^2+\frac{A^2}{4}-\frac{\tilde{\delta}A}{2}\cos\varphi-\frac{\bar{L}^2-\bar{J}^2}{2}A\sin\varphi\right]\exp[-(\bar{L}^2+\bar{J}^2+A\sin\varphi)t]\right.$$
$$-\frac{1}{A^2}\left[\frac{\tilde{\delta}^2}{4}+\tilde{K}^2+\left(\frac{\bar{L}^2+\bar{J}^2}{2}\right)^2+\frac{A^2}{4}+\frac{\tilde{\delta}A}{2}\cos\varphi+\frac{\bar{L}^2-\bar{J}^2}{2}A\sin\varphi\right]\exp[-(\bar{L}^2+J^2-A\sin\varphi)t]$$
$$-\frac{1}{A^2}\left[\frac{\tilde{\delta}^2}{4}+\tilde{K}^2-\frac{A^2}{4}-\left(\frac{\bar{L}^2+\bar{J}^2}{2}\right)^2\right]2\cos(At\cos\varphi)\exp[-(\bar{L}^2+\bar{J}^2)t]$$
$$\left.-\frac{1}{A^2}\left[\tilde{\delta}A\sin\varphi-(\bar{L}^2-\bar{J}^2)A\cos\varphi\right]2\sin(At\cos\varphi)\exp[-(\bar{L}^2+\bar{J}^2)t]\right\}, \tag{10}$$

where

$$\frac{A^2}{4}=\left\{\left[\left(\frac{\bar{L}^2+\bar{J}^2}{2}\right)^2-\tilde{K}^2-\frac{\tilde{\delta}^2}{4}\right]^2+\left(\frac{\tilde{\delta}}{2}(J^2-L^2)+2\bar{K}\bar{J}\bar{L}\right)^2\right\}^{1/2},$$

$$\tan 2\varphi=-\frac{(\tilde{\delta}/2)(\bar{J}^2-\bar{L}^2)+2\tilde{K}\bar{J}\bar{L}}{\tilde{K}^2+(\tilde{\delta}^2/4)-[(\bar{L}^2+\bar{J}^2)/2]^2},$$

$$\tilde{\delta}=\delta-\Delta^L+\Delta^J.$$

This result was already obtained by Beers and Armstrong, who used a quantum approach for the driving field. Equation (10) is rather complicated, and only the study of some limiting cases is actually useful.

A first one is $\bar{K}\gg\bar{J},\bar{L}$. This condition is fulfilled if $n-p\gg p$ or if $n=2$. We have already studied this case in Ref. 3; this treatment is equivalent to the former one.

A second interesting case is $\bar{L}\gg\bar{K},\bar{J}$, which generally corresponds to $n-p\ll p,n$. The response to a continuous excitation has been studied by Armstrong, Beers, and Feneuille.[4]

III. RESOLUTION OF THE EQUATIONS IN THE CASE $L\gg K, J$

In this case, Eq. (10) can be simplified by making a first-order expansion in terms of the small quantities K/L^2, J^2/L^2, JL/L^2. However, a perturbative treatment of Eqs. (9) and (5c), using as zeroth-order solutions the ones obtained for $J^2=JL=K=0$, is equivalent to the expansion of the exact formula and in fact is simpler. This leads to the following result:

$$N=P\frac{2\bar{J}^2}{\tilde{\delta}^2+\bar{L}^4}\left[\left(\tilde{\delta}+\frac{\tilde{K}\bar{L}}{\bar{J}}\right)^2T+\frac{\bar{L}^2\bar{J}^2+\tilde{K}^2}{2\bar{J}^2}(1-e^{-2\bar{L}^2T})\right.$$
$$\left.+\frac{2\bar{L}^2(\tilde{\delta}+\tilde{K}\bar{L}/\bar{J})^2}{\tilde{\delta}^2+\bar{L}^4}(e^{-\bar{L}^2T}\cos\delta T-1)+\frac{2(\tilde{\delta}+\tilde{K}\bar{L}/\bar{J})(\bar{L}^4-\tilde{\delta}\tilde{K}\bar{L}/\bar{J})}{\tilde{\delta}^2+\bar{L}^4}e^{-\bar{L}^2T}\sin\tilde{\delta}T\right]. \tag{11}$$

It is possible to recognize in the term $2P\bar{J}^2[(\tilde{\delta}+\tilde{K}\bar{L}/\bar{J})^2/(\tilde{\delta}^2+\bar{L}^4)]T$ the result obtained by Armstrong, Beers, and Feneuille for the response to a continuous excitation. The resonant response is the sum of an even part (Lorentzian profile) and an odd part (dispersion type); the width of these two contributions is $\bar{L}^2$. The parameter $\tilde{K}/\bar{J}\bar{L}=q$ characterizes the relative contribution of the even and the odd parts and thus the asymmetry and the sharpness of the profile. The continuous regime is established after a period of the order of $\bar{L}^{-2}$.

The transient curve is characterized by $\bar{L}^2$, q, and a third dimensionless parameter $\theta=\bar{L}^2T$. When $\theta\gg1$, the continuous regime has been reached within the duration of the pulse. When $\theta\ll1$, the spectral width of the pulse, $\Delta=1/T$, is larger than $\bar{L}^2$, which is the frequency extension of the resonant phenomena, and therefore the resonant response becomes quite flat and has the excitation width. We study here the intermediate situation corresponding to θ of the order of one, that is to say, the transition from continuous monochromatic excitation to impact excitation. Figure 1 shows the evolution of the transient curves for $q=(10)^{1/2}$ with different values of θ. We have plotted N/N_D as a function of $y=\delta/q\bar{L}^2=\delta\bar{J}/\tilde{K}\bar{L}$, N_D being the intensity of the direct process. (One can remark that $\tilde{K}\bar{L}/\bar{J}$ is an atomic parameter; thus $\tilde{K}\bar{L}/\bar{J}$ does not depend strongly on the characteristics of the field pulse).

As can be seen in Eq. (11), the continuous regime is reached faster for the large values of δ than for the maximum of the curve.

It is particularly interesting to consider the

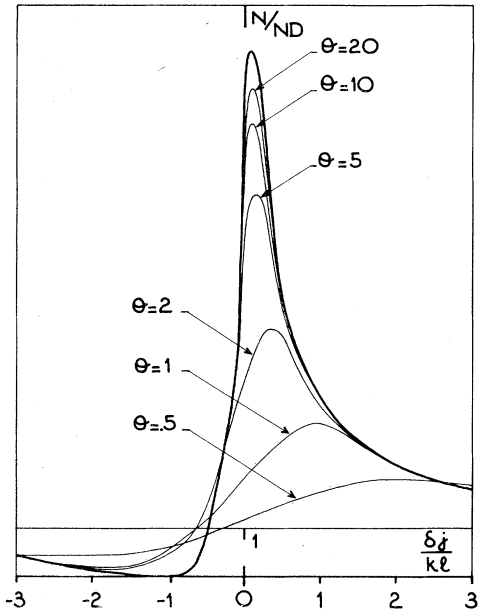

FIG. 1. Resonance curve induced by a square-pulse excitation: evolution with $\theta=\bar{L}^2T$. Fine curves, transients; bold curve, continuous.

cases where the resonant process is more important than the direct process. (Otherwise the resonance is too small to be detected.) This occurs when q^2 is larger than a few units. For the continuous regime, in this case, the wings of the curve depend essentially on the value of $\tilde{K}\bar{L}/\bar{J}$, which is quite independent of the field intensity, while the maximum is $1+q^2$, which strongly depends on the field intensity. The previous study shows that by using short excitation pulses and by ignoring transient effects, it is possible to measure a correct value of $\tilde{K}\bar{L}/\bar{J}$ by analyzing the wings of the curves, but in this case, one would certainly underestimate the value of q.

Another parameter which measures the effective order of nonlinearity is often introduced to characterize resonant multiphonon ionization processes: $k=\partial\log N/\partial\log I$ (I being the field intensity). For continuous excitation, N is given by

$$N=P\frac{\bar{J}^2}{\tilde{\delta}^2+\bar{L}^4}\left(\tilde{\delta}+\frac{\tilde{K}\bar{L}}{\bar{J}}\right)^2 .$$

Then

$$k(\delta)=n-2(n-p)\left(\frac{\bar{L}^4}{\tilde{\delta}^2+\bar{L}^4}-\frac{\tilde{\delta}\,\Delta^L}{\tilde{\delta}^2+\bar{L}^4}+\frac{\Delta^L}{\tilde{\delta}+\tilde{K}\bar{L}/\bar{J}}\right) .$$

In deriving the previous formula, we have neglected the contribution of nonresonant light shifts of i and φ which vary as I^2. Even with this approximation which could not be valid in some cases, the shape of $k(\delta)$ is very sensitive to the exact values of parameters such as Δ^L, which cannot be calculated *ab initio* with sufficient accuracy. On the other hand, one could imagine extracting these parameters from experimental shapes of k vs δ, but the number of these parameters is too high with respect to the precision of available experimental data, which still are very few. Nevertheless, $k(\delta)$ exhibits in any case a very rapid variation for the value of δ corresponding to a null value of ionization cross section. This result is in qualitative agreement with the experiments of four-photon ionization of Cs performed by the Saclay group.[2] In transient regime, the minimum value of N is always different from zero. The variation of N and accordingly the variation of k are expected to be smoother than in the continuous regime.

IV. NUMERICAL RESOLUTION

Equations (9) have been derived in the case of a square pulse, but they can be obtained also for a pulse with any shape. This is the main advantage of having introduced a semiclassical description of the field. When we consider a pulse of field with amplitude variation, L_E can be written $L_E f(t)$ and the expression (8a) becomes

$$f(t)\int_{-\infty}^{+\infty}L_E\,dE\int_{-\infty}^{t}L_E a_\varphi(t')\,dt' \times\exp[i\delta_E(t-t')]=f(t)\,g(t) . \tag{12a}$$

The Fourier transform of $g(t)$ is

$$\left(L_E^2+\frac{i}{\pi}\,\mathcal{P}\int\frac{L_{E'}^2}{E-E'}dE'\right)h(E) , \tag{12b}$$

where $h(E)$ is the Fourier transform of $a_\varphi(t)f(t)$. The extension of $h(E)$ is about the largest value between $K, L_E^2, J_E^2, 1/T$ (T being the width of the pulse). The description of the light pulse supposes that $1/T\ll\omega$; thus even if T is small, the extension of $h(E)$ is small enough that E can be replaced by E_0 in expression (12b). So expression (12a) can be written

$$\left(f^2(t)L_{E_0}^2+\frac{i}{\pi}\,\mathcal{P}\int\frac{L_{E'}^2 f(t)^2}{E_0-E'}\,dE'\right)a(t) ,$$

and we get again for a pulse with amplitude variation the system of equations (9). We have performed numerical resolutions of Eqs. (9) for different pulse shapes (carried out on the UNIVAC 1110 de l'Université de Paris-Sud). We have considered the case $p=6$, $n=7$. Although time-dependent phases appear in the general formalism, we have considered only field-amplitude variations. The different shapes are as follows:

Case 1:

$$L = 2.5(t/T)e^{-t^2/T^2}L_0 \quad \text{for } t/T \geq 0,$$

$$L = 0 \quad \text{for } t < 0;$$

Case 2:

$$L = (t/3T - 1)^2 \times 1.27 L_0 \quad \text{for } 0 \leq t/T \leq 3,$$

$$L = 0 \quad \text{for } t < 0 \text{ and } t/T > 3; \tag{13}$$

Case 3:

$$L = 1.13 \sin(2\pi t/3T)L_0 \quad \text{for } 0 < t/T < \tfrac{3}{2},$$

$$L = 0 \quad \text{for } t < 0 \text{ and } t/T > \tfrac{3}{2}.$$

These pulses correspond to the same half-maximum width of L and to the same value of

$$\langle L^2 \rangle = \frac{1}{T} \int_{-\infty}^{+\infty} L_E^2(t)\, dt.$$

In this particular case, $n - p = 1$. $\theta = \langle L^2 \rangle T$ is the energy of the pulse.

The resolution of the equations was done for different values of T and L_0. The results are very similar to those obtained with the square pulse. That is to say, when θ is short, the resonance disappears, and when θ increases, the transient curve tends towards the Fano continuous profile, faster for the large values of δ than for the maximum of the curve. As already noted in Sec. III, the continuous curve is characterized essentially by the value of $\tilde{K}\bar{L}/\bar{J}$ for the wings of the curve and the value of L^2 for the maximum. One can extract from the Fano curve, obtained as a limit of the transient curve, an effective value of $\tilde{K}\bar{L}/\bar{J}$ and L^2. The value of $(\tilde{K}\bar{L}/\bar{J})_{\text{eff}}$ deduced in this way is close to the atomic parameter $\bar{K}\bar{L}/\bar{J}$, and the wings of the curve appear to be independent of the shape of the pulse. On the contrary, the maximum of the curve depends on the shape of the pulse. This dependence can be easily estimated provided that the increasing part of the pulse shape does not suffer a too rapid variation. This can be achieved by calculating

$$\lim_{\theta \to \infty} (N/N_D) = \langle N/N_D \rangle;$$

the corresponding calculation is given in the Appendix. The most interesting result is obtained for $\tilde{\delta} = 0$. We show that

$$\left\langle \left(\frac{N}{N_D} \right)_{\delta=0} \right\rangle = \frac{\int_{-\infty}^{+\infty} (\tilde{K}^2/\bar{L}^2)\, dt}{\int_{-\infty}^{+\infty} \bar{J}^2\, dt},$$

while in the continuous regime

$$(N/N_D)_{\delta=0} = K^2/L^2J^2.$$

Therefore $\langle (N/N_D)_{\delta=0} \rangle$ is proportional to the parameter

$$\frac{\int_{-\infty}^{+\infty} \mathcal{E}^{4p-2n}(t)\, dt}{\int_{-\infty}^{+\infty} \mathcal{E}^{2n}(t)\, dt},$$

which depends only on the pulse characteristics. We have calculated the value of $\langle (N/N_D)_{\delta=0} \rangle$ for the

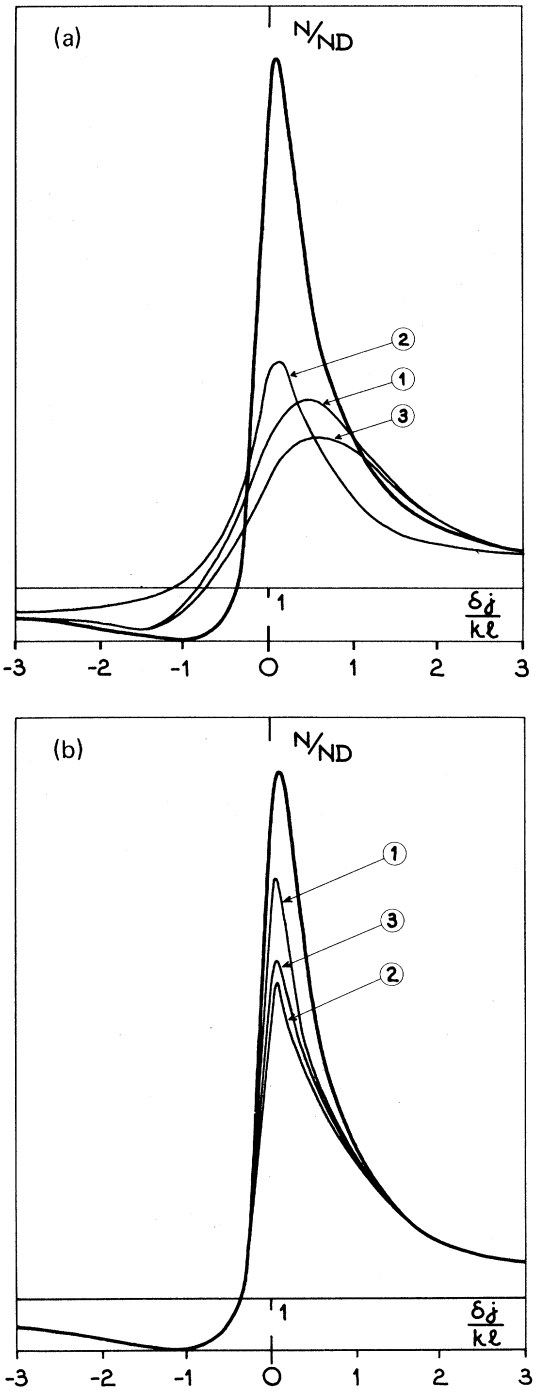

FIG. 2. Resonance curves obtained for different pulse shapes [see Eq. (13)], for (a) $\theta = 2$ and (b) $\theta = 10$.

different pulses considered in numerical calculations and compare them below with the result of numerical integration.

Case	$\langle (N/N_D)_{\delta=0} \rangle$	$(N/N_D)_{\delta=0}$
1	8.93	9
2	13.81	17
3	11.75	11

For cases 1 and 3, where the field increases slowly, the agreement is rather good, but for case 2 this prediction leads to too small a value of the resonance. This is not surprising since in the latter case the maximum of the field appears at the beginning of the pulse. These results are illustrated in Fig. 2. Figure 3 shows the evolution of the resonance for different values of θ.

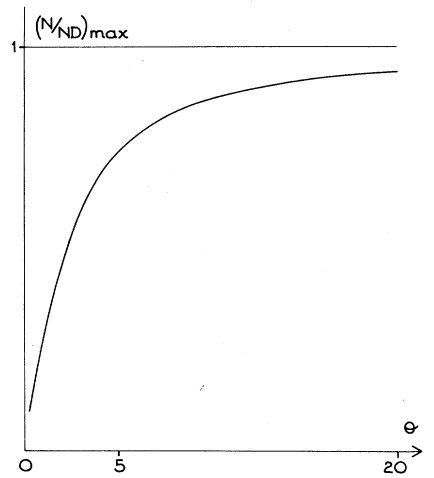

FIG. 3. Evolution with θ of the maximum of the resonant curves (excitation by a square pulse).

V. APPLICATION: CESIUM

For a complete comparison of our results with experimental curves, we would have to obtain reliable values of the atomic parameters k and l_E and know exactly the value of P; such parameters are not available in the literature. Thus we determine only those conditions in which the effect of the duration of the pulse becomes important. We consider the case of Ref. 5, where multiphoton resonant ionization in Cs is described in a sufficiently detailed way. However, we have no information about the pulse shape, and therefore only the influence of the pulse length can be discussed. In the first step, the resonant level $5p^6 6f$ (28 329.7 cm^{-1}) is reached by a three-photon process. The second step is a one-photon ionization. The resonance was observed for laser intensity of 10^8–10^9 W/cm^2. The ionization cross section from the $6f$ level calculated by Aymar[6] is 1.4×10^{-18} cm^2. Thus for a laser intensity of 10^9 W/cm^2, the value of L^2 is 3×10^9 sec^{-1}. In these experiments the duration of the pulse was 30 nsec, leading to the value $\theta = 2000$.

The resonance curve observed was very similar to the Fano curves obtained in a continuous regime. This fact is in agreement with our interpretation of the role of θ. We have noted in Sec. III that the effect of pulse duration becomes important when θ becomes smaller than 10. Thus one can think that these effects would appear in experiments using pulses of duration smaller than 3 nsec.

APPENDIX

Starting from Eq. (9) of Sec. II, we derive the density-matrix equations. Since $L_E^2 \ll J_E^2$, we suppose that $n_E = a_E a_E^*$ and $n_\varphi = a_\varphi a_\varphi^*$ are much smaller than $n_i = a_i a_i^*$, that is to say $n_i \simeq 1$. By introducing the new variable $\eta = \int_{-\infty}^{t} L_E^2\, dt$, we deduce

$$\frac{dn_E}{dt} = 2J_E^2 + 2L_E^2\left(e^{-2\eta}\int_0^\eta \frac{J_E J_E + iK}{L_E^2}\, e^{\eta'}\, d\eta' \int_0^{\eta'} e^{\eta''}\, \frac{J_E L_E - iK}{L_E^2}\, e^{i\delta(t''-t')} d\eta'' + \text{c.c.}\right)$$

$$- 2J_E L_E\left(e^{-\eta}\int_0^\eta \frac{J_E L_E - iK}{L_E^2}\, e^{\eta'} e^{i\delta(t'-t)} d\eta' + \text{c.c.}\right)$$

For large values of θ, η rapidly reaches large values, and we can consider that $e^{-\eta+\eta'}$, with $\eta' < \eta$, have values different from zero only when $\eta - \eta' \leq 1$. Since η varies very rapidly, the corresponding value of J_E/L_E varies slowly and the integral

$$e^{-\eta}\int_0^\eta \frac{J_E}{L_E}\, e^{\eta'} e^{i\delta(t'-t)} d\eta'$$

can be replaced by

$$\frac{J_E}{L_E}(\eta)\int_0^\eta e^{\eta'-\eta} e^{i\delta(t'-t)} d\eta' .$$

The same argument leads one to replace

$$e^{-2\eta}\int_0^\eta e^{\eta'}\frac{J_E}{L_E}\int_0^{\eta'}\frac{J_E}{L_E}\, e^{\eta''}\, d\eta''$$

by

$$\frac{J_E^2}{L_E^2}\int_0^{\eta} d\eta' \int_0^{\eta'} d\eta'' e^{\eta'+\eta''-2\eta} e^{i\delta(t''-t')}.$$

An integration by parts leads to

$$\int_0^{\eta} e^{\eta'-\eta} e^{i\delta(t'-t)} d\eta' = \frac{L_E^2}{L_E^2+i\delta}.$$

Under these approximations we obtain

$$n_E = \int_{-\infty}^{t} 2\,\frac{(J_E\delta + KL_E)^2}{\delta^2+L_E^4}\,dt'.$$

The direct process can be calculated by assuming that $k = L_E = 0$ in Eqs. (5) leading to

$$(n_E)_D = \int_{-\infty}^{t} 2J_E^2.$$

Then

$$\left\langle \frac{N}{N_D} \right\rangle = \int_{-\infty}^{+\infty} \frac{(J_E\delta + K_L L_E)^2}{\delta^2+L_E^4}\,dt \Big/ \int_{-\infty}^{+\infty} J_E^2\,dt.$$

The assumptions done to calculate the integrals on η are not valid for small values of η. If large values of the field amplitude appear when η is large, the contribution of the small values of η is negligible. Thus one can understand why the prediction is quite good for pulses where the maximum appears a long time after the beginning of the pulse. That is not true for the parabolic pulse (12b) where the maximum value is obtained just at the beginning of the pulse.

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