Laser-induced autoionizinglike behavior, population trapping, and stimulated Raman processes in real atoms

Bo-nian Dai and P. Lambropoulos
University of Crete and Research Center of Crete, Iraklion, Crete, Greece
and Physics Department, University of Southern California, Los Angeles, California 90089-0484
(Received 22 September 1986)

We present a unified theory of laser-induced autoionizinglike behavior in single-photon and multiphoton processes. We incorporate processes assumed unimportant in previous theories and on the basis of realistic calculations we interpret existing experimental results.

Beginning with the work of Heller and Popov,[1] the last ten years have seen a number of papers[2-7] devoted to what is referred to as autoionizinglike behavior, when two bound atomic states of a one-electron atom are coupled to the same continuum state by two independent lasers. Let $|g\rangle$ be a ground and $|a\rangle$ an excited state below threshold, with respective energies E_g and E_a. One laser of frequency ω_1 is chosen so that $E_g+\hbar\omega_1$ is above the ionization threshold, while the frequency ω_2 of a second laser is such that $E_a+\hbar\omega_2 \cong E_g+\hbar\omega_1$ as in Fig. 1(a). The atom, being initially in state $|g\rangle$, will simply ionize if only laser ω_1 is turned on. With both lasers on, a number of interference effects may be manifested as the frequencies and intensities of the two sources are varied. One much discussed[3,4] such effect, for example, may lead to trapping atomic population in the excited state $|a\rangle$. A related quantity is the line shape of ionization[1] (or equivalently absorption[2] of the radiation at ω_1) as ω_1 is tuned around the resonance value $\hbar\omega_1 = E_a - E_g + \hbar\omega_2$ with ω_2 held fixed. An asymmetric line shape has been predicted[1,2] (hence the characterization autoionizinglike) and its implication on other processes, such as harmonic generation, has been contemplated.[5,6]

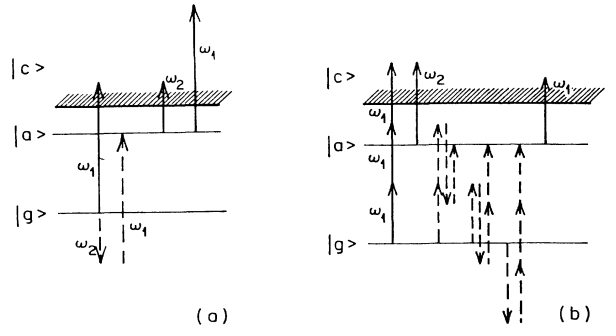

FIG. 1. (a) Schematic representation of processes coupling two discrete states to each other and to the same continuum. Two separate lasers with frequencies ω_1 and ω_2 are assumed, as in the experiment of Ref. 2. (b) Same as in (a) except that it takes three photons to couple one of the states with the continuum as in the experiment of Ref. 7.

In recent experiments, however, Feldman *et al.*[7] found no such asymmetry and pointed out that Raman processes made the dominant contribution. As shown below, this is to be expected under the conditions of their experiment.

In this paper we present a theory and the results of realistic calculations that (a) demonstrate that certain important aspects assumed unimportant in previous theories introduce qualitative changes and drastically modify expectations on population trapping, (b) unify apparently different processes, (c) interpret existing experimental results, and (d) demonstrate that theoretical modeling of these processes with free parameters not derived from atomic calculations can lead to quite misleading conclusions and expectations.

The simplest autoionizinglike process as described above is depicted in Fig. 1(a). The state $|g\rangle$ and $|a\rangle$ are coupled to the same continuum $|c\rangle$ by separate lasers. Denoting by D the dipole interaction between atom and radiation, the respective coupling matrix elements are D_{cg} and D_{ca}. Thus states $|g\rangle$ and $|a\rangle$ are indirectly coupled to each other via the continuum. There is, however, another coupling between $|g\rangle$ and $|a\rangle$ through the nonresonant Raman process also shown in Fig. 1(a) by dashed lines. This process, which has been left out of most previous theories,[1-6] will be shown to be of crucial importance. Although Alber and Zoller[8] have noted its significance under conditions of near resonance with an atomic state, it has generally been neglected in most previous theories.[1-6] As we show below, it can be of crucial importance even when it is nonresonant. The formal analogy[9] with autoionization is now complete. Two discrete states are coupled to each other by radiation while each of them is coupled to the continuum. If instead of D_{ca} we had a matrix element V_{ca} of configuration interaction, which is independent of radiation, we would have proper autoionization found in atoms with more than one electron. The difference between the two is quite significant, since V_{ca} cannot be controlled while D_{ca} is proportional to the strength of the laser field.

We formulate the theory in terms of the resolvent

operator[9] $G(z)=(z-H)^{-1}$, where $H=H^A+H^R+D=H^0+D$ is the total Hamiltonian consisting of a sum of the free atomic H^A and radiation H^R parts coupled through the interaction D. A quantized H^R with two discrete modes and a representation in photon-number states $|n\rangle$ are assumed and sufficient for our purposes. At $t=0$ the system is in state $|\tilde{g}\rangle=|g\rangle|n_1+1,n_2-1\rangle$ whose energy is $\tilde{E}_g=E_g+(n_1+1)\hbar\omega_1+(n_2-1)\hbar\omega_2$. Other states entering the problem are $|\tilde{a}\rangle=|a\rangle|n_1,n_2\rangle$, $|\tilde{c}_1\rangle=|c\rangle|n_1-1,n_2\rangle$, and $|\tilde{c}_2\rangle=|c\rangle|n_1,n_2-1\rangle$ with respective energies $\tilde{E}_a=E_a+n_1\hbar\omega_1+n_2\hbar\omega_2$, $\tilde{E}_{c_1}=E_c+(n_1-1)\hbar\omega_1+n_2\hbar\omega_2$, and $\tilde{E}_{c_2}=E_c+n_1\hbar\omega_1+(n_2-1)\hbar\omega_2$. The matrix elements $G_{\tilde{g}\tilde{g}}$, $G_{\tilde{a}\tilde{g}}$, $G_{\tilde{c}_1\tilde{g}}$, and $G_{\tilde{c}_2\tilde{g}}$ of $G(z)$ must be considered explicitly. Through the standard algebraic procedure,[9] we eliminate $G_{\tilde{c}_1\tilde{g}}$ and $G_{\tilde{c}_2\tilde{g}}$ obtaining the equations

$$(z+\frac{i}{2}\gamma_g^{(1)})G_{\tilde{g}\tilde{g}}-\tilde{D}_{\tilde{g}\tilde{a}}^{(2)}(1-i/q)G_{\tilde{a}\tilde{g}}=1\ , \tag{1a}$$

$$-\tilde{D}_{\tilde{a}\tilde{g}}^{(2)}(1-i/q)G_{\tilde{g}\tilde{g}}+(z+\delta+\frac{i}{2}\gamma_a^{(1)}+\frac{i}{2}\gamma_a^{(2)})G_{\tilde{a}\tilde{g}}=0\ , \tag{1b}$$

where

$$\tilde{D}_{\tilde{g}\tilde{a}}^{(2)}=D_{\tilde{g}\tilde{a}}^{(2)}+\mathrm{P}\int d\tilde{E}_c\frac{D_{\tilde{g}\tilde{c}}D_{\tilde{c}\tilde{a}}}{\tilde{E}_g-\tilde{E}_c}\ ,$$

$$D_{\tilde{g}\tilde{a}}^{(2)}=\sum_l\frac{D_{\tilde{g}\tilde{l}}D_{\tilde{l}\tilde{a}}}{\tilde{E}_g-\tilde{E}_l}\ ,$$

$$\gamma_g^{(1)}=\pi|D_{\tilde{c}_2\tilde{g}}|^2_{\tilde{E}_{c_2}=\tilde{E}_g}\ ,$$

$$\gamma_a^{(j)}=\pi|D_{\tilde{c}_j\tilde{a}}|^2E_{cj}=\tilde{E}_g\quad(j=1,2)$$

and $\delta=\hbar\omega_1+E_g+S_g-\hbar\omega_2-E_a-S_a$. Both states $|g\rangle$ and $|a\rangle$ are Stark shifted by the amounts S_g and S_a which are linear in the intensity of all radiation present. These shifts are included in all calculations reported in this paper but, in order to keep the equations as simple as possible, are not shown explicitly. All dipole matrix elements are proportional to the amplitude of the electric field or the square root of the photon number. The two-photon matrix element $D_{\tilde{g}\tilde{a}}^{(2)}$ represents the Raman process that couples $|g\rangle$ with $|a\rangle$. The principal-value integral

$$\mathrm{P}\int d\tilde{E}_c\frac{D_{\tilde{g}c}D_{\tilde{c}a}}{\tilde{E}_g-\tilde{E}_c}$$

represents the absorption of ω_1 followed by the emission of ω_2. It refers to atomic continuum states. It also implicitly includes the sum over bound states. What appears in Eqs. (1a) and (1b), however, is an effective two-photon matrix element $\tilde{D}_{\tilde{g}\tilde{a}}^{(2)}$ modified by the couplings to the continuum, very much as in autoionization. The quantities γ represent ionization widths (rates) of the respective atomic states and are proportional to the appropriate laser intensity. State $|a\rangle$, being higher than $|g\rangle$, can always be ionized by both lasers; a fact that has been noted by Feldman *et al.*[7] and will be shown to play a critical role in population trapping. Its total ionization width is therefore the sum of two widths $\gamma_a^{(1)}$ and $\gamma_a^{(2)}$

From the solution of Eqs. (1a) and (1b) we obtain $G_{\tilde{g}\tilde{g}}(z)$ and $G_{\tilde{a}\tilde{g}}(z)$, which give the amplitudes $U_g(t)$ and $U_a(t)$ through the inverse Laplace transform $\int G(z)e^{izt}dz$ on the appropriate contour[9,11] of the complex z plane. For times $t>0$, the probability of ionization (total ionization up to that time) is given by $P_{\mathrm{ion}}(t)=1-|U_g(t)|^2-|U_a(t)|^2$, while the total absorption of radiation from laser ω_1 is given by $P_{\mathrm{abs}}(t)=1-|U_g(t)|^2$. In general, these probabilities can be nontrivial functions of time, especially for higher laser intensities, say, above 10^6 W/cm^2. For not so high intensity, $P(t)$ increases linearly with t, the constant of proportionality (dP/dt) being the transition probability per unit time. The profile of dP/dt as a function of ω_1 can in that case be written as $(q+\epsilon)^2/(1+\epsilon^2)$, where $\epsilon=2\delta/(\gamma_a^{(1)}+\gamma_a^{(2)})$ is the detuning in units of the half-width of $|a\rangle$ (which plays here the role of the autoionization width) and q is a shape parameter given by

$$q=\frac{D_{\tilde{g}\tilde{a}}^{(2)}+\mathrm{P}\int d\tilde{E}_c(D_{\tilde{g}\tilde{c}}D_{\tilde{c}\tilde{a}}/\tilde{E}_g-\tilde{E}_c)}{\pi(D_{\tilde{g}\tilde{c}}D_{\tilde{c}\tilde{a}})_{\tilde{E}_c=\tilde{E}_g}}\ . \tag{2}$$

The tildes simply remind us that $|\tilde{c}_1\rangle$ and $|\tilde{c}_2\rangle$ differ only in the photon states. As a consequence, $D_{\tilde{g}\tilde{c}}=\mathcal{E}_1 r_{gc}$ and $D_{c\tilde{a}}\simeq\mathcal{E}_2 r_{ca}$, where $\mathcal{E}_j$ is the respective laser fields and r_{gc}, r_{ca} the corresponding atomic dipole matrix elements. Thus the principal-value integral in the numerator as well as the product in the denominator are proportional to $\mathcal{E}_1\mathcal{E}_2$. Similarly, $D_{\tilde{g}\tilde{a}}^{(2)}$ is proportional to $\mathcal{E}_1\mathcal{E}_2$, because the Raman process it represents involves the stimulated emission of a photon ω_2 followed by the absorption of a photon ω_1. The quantity q of Eq. (2) is therefore an atomic parameter independent of the fields, and cannot be changed by varying the laser intensities.

The presence of $D_{\tilde{g}\tilde{a}}^{(2)}$ has also been assumed unimportant in much of the literature on the subject.[1-6] Perhaps the best way to demonstrate here its importance is to apply our results to the interpretation of experimental data reported by Heller *et al.*[2] In that experiment, $|g\rangle$ was the $6s$ ground state of Cs and $|a\rangle$ the $8s$ state; the photon frequencies were $\omega_1\cong 33\,715$ cm^{-1} and $\omega_2=9\,397.83$ cm^{-1}, while the intensities of both lasers were quoted as $I_1\simeq I_2\simeq 10^8$ W/cm^2. After having calculated all of the atomic parameters entering the problem (using quantum-defect theory with a Green's-function technique[10] for the infinite summation involved in $D_{\tilde{g}\tilde{a}}^{(2)}$), we have calculated absorption of ω_1 as well as ionization, as a function of ω_1 around $\hbar\omega_1=\hbar\omega_2+E_{8s}-E_{6s}$ with ω_2 kept fixed at the above value. The line profile for two different values of the intensity I_2 under 5-ns pulse duration (square shape) is shown in Fig. 2(a). Both profiles correspond to $q=-4.3$ as obtained in our calculations. Obviously the change of the shape is not due to

a change of q; which brings us to another point whose significance has often been overlooked in the literature. The change is due to the time dependence[9] (time saturation) of the whole process combined with power broadening and Rabi oscillations. In their analysis of the data, Heller *et al.*[2] obtain a value of $q=6$ by fitting to the usual autoionization profile $(q+\epsilon)^2/(1+\epsilon^2)$. We assume they mean the absolute value of q, because if it were positive the asymmetry of the line would be the reverse of that obtained in the experiment.[2] The difference between their 6 and our 4.3 can be attributed to their fitting to an expression which can be only approximately valid owing to the above-mentioned time effects. The authors of the experiment appear to have relied on a rough theoretical estimate[1] of their q based on an expression for q without $D_{g\bar{a}}^{(2)}$. That such an expression leads to results which disagree with the experiment is also shown in our Fig. 2(a) where we plot the profile obtained by setting $D_{g\bar{a}}^{(2)}=0$. It is qualitatively different from what was seen in the experiment and the resulting value of q is $+0.22$. We have thus a dramatic demonstration of the importance of the nonresonant Raman process.

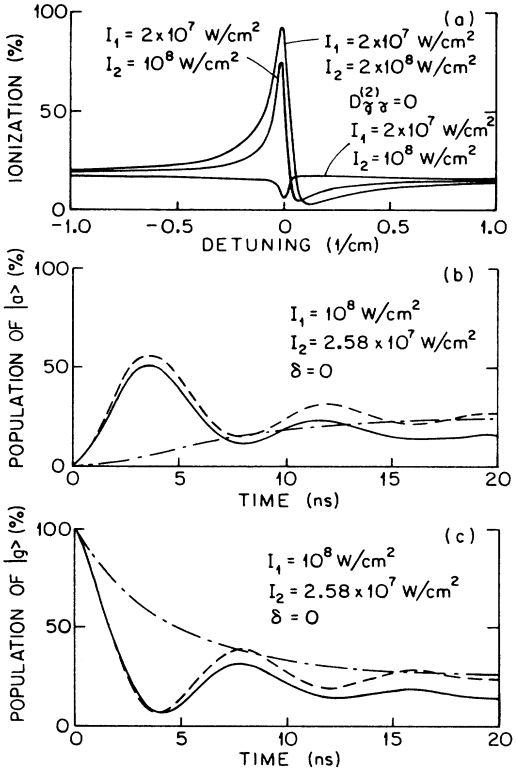

FIG. 2. (a) Ionization line shape for various combinations of laser intensities with and without the Raman process indicated. The states $6s$ and $8s$ of Cs are coupled to the p continuum by two lasers of intensities I_1 and I_2 and frequencies $\omega_1=33\,715$ cm^{-1}, $\omega_2=9\,397.83$ cm^{-1}, as in the experiment of Ref. 2. The pulse duration is 5 ns. (b) Population of state $|a\rangle=8s$ as a function of pulse duration for $\delta=0$ and the indicated intensities. ——, calculation includes all decays for $|a\rangle$ and $|g\rangle$, i.e., $\gamma_a^{(1)}\neq0,\gamma_a^{(2)}\neq0,D_{g\bar{a}}^{(2)}\neq0$; — — —, calculation with $\gamma_a^{(1)}=0$ but with the Raman included; —·—·—, calculation with $\gamma_a^{(1)}$ and $D_{g\bar{a}}^{(2)}$ set equal to zero. (c) Population of state $|g\rangle=6s$ as a function of pulse duration for $\delta=0$ and the indicated intensities. The various lines correspond to the conditions of (b) above.

What about population trapping in state $|a\rangle$? Since both fields lead to ionization of $|a\rangle$, while only the transition induced by ω_2 interferes with that of $|g\rangle+\omega_1\rightarrow|c_2\rangle$, state $|a\rangle$ decays constantly into the continuum because of the transition $|a\rangle+\omega_1\rightarrow|c_1\rangle$. And since $|g\rangle$ is coupled to $|a\rangle$, it also decays. As a result, there can be no long-term trapping. How much trapping is possible and for how long depends on the laser intensities, the atomic parameters, and where ω_1 is tuned. For the Cs experiment,[2] for example, our calculations show that, if ω_1 is tuned to $\delta=0$, the maximum of $|U_a|^2$ is about 0.55 for a pulse of 3.5 ns [Fig. 2(b)]. For pulses longer than 15 ns the population trapping is much smaller [Figs. 2(b) and 2(c)]. Two competing factors enter in attempting to achieve large population trapping in $|a\rangle$. Large intensities I_1 and I_2 help by bringing population into $|a\rangle$, but large I_1 also removes population from $|a\rangle$ into the continuum. Clearly, an optimum requires a choice of energy $\tilde{E}_g=E_g+\hbar\omega_1$ such that $E_a+\hbar\omega_1$ falls into a minimum of the ionization cross section of $|a\rangle$. A not unlikely complication may arise, however, if $\tilde{E}_g$ is so high that the $\hbar\omega_2$ necessary for resonance can also cause ionization of $|g\rangle$ (see also related discussion below). In any case, it is irrelevant to predict[3,4] population trapping by calculating $\lim_{t\rightarrow\infty}|U_a(t)|^2$. Whatever trapping occurs will be done at some optimum time which will in fact be short, in the sense $\gamma_a^{(1)}t<1$.

We turn now to another type of process[5–7] and experiment where it takes three ω_1 photons to ionize $|g\rangle$, while a second laser ω_2 is such that $E_g+3\hbar\omega_1=E_a+\hbar\omega_2$, as depicted in Fig. 1(b). Again, there exists a Raman process involving the absorption of three ω_1 and the emission of one ω_2 in all possible orders, as shown in Fig. 1(b) by dashed lines. In other words, there are three distinct Feynman diagrams contributing to the Raman coupling. The calculation is now much more complex, but formally similar to the previous case. Assuming ω_1 to be such that there are neither one-photon nor two-photon resonances with real atomic intermediate states (which has been the case in the experiments[5–7]) an extremely lengthy derivation, which need not be reproduced here, leads to a set of equations similar to Eqs. (1a) and (1b) with $D_{g\bar{a}}^{(2)}$ replaced by a four-photon Raman matrix element $D_{g\bar{a}}^{(4)}$, $D_{g\bar{c}}$ replaced by a three-photon ionization matrix element $D_{g\bar{c}}^{(3)}$, while $D_{\bar{c}\bar{a}}$ remains a single-photon matrix element as before. Now $\gamma_g^{(1)}$ is a three-photon ionization width, $\gamma_a^{(2)}$ remains a single-photon ionization width, while $\gamma_a^{(1)}$ may be a single- or two-photon ionization width, depending on the choice of $|g\rangle$, $|a\rangle$, ω_1, and ω_2. An additional width of $|g\rangle$, namely $\gamma_g^{(2)}$, representing its multiphoton ionization by ω_2, as well as combinations of ω_1 with ω_2, must now be included if both lasers are to be allowed the same inten-

sity. In principle, it is always present, but it was 6–7 orders of magnitude smaller than $\gamma_g^{(1)}$ in the previous case since the latter was a single-photon process.

A shape parameter $q_{(3)}$ can again be written for the line profile of either ionization or absorption as a function of ω_1 and ω_2 fixed. It is given by

$$q_{(3)}=\frac{D^{(4)}_{\tilde{g}\,\tilde{a}}+\mathrm{P}\int d\tilde{E}_c(D^{(3)}_{\tilde{g}\,\tilde{c}}D_{\tilde{c}\,\tilde{a}}/\tilde{E}_g-\tilde{E}_c)}{\pi(D^{(3)}_{\tilde{g}\,\tilde{c}}D_{\tilde{c}\,\tilde{a}})_{\tilde{E}_c=\tilde{E}_g}} . \tag{3}$$

It can be easily verified [with the aid of Fig. 1(b)] that all terms in the numerator and denominator of Eq. (3) are proportional to $\mathcal{E}_1^3\mathcal{E}_2$. As a result, $q_{(3)}$ is again an atomic parameter independent of the laser intensities. Experiments of this type have been reported[5–7] for Na where $|g\rangle=|3s\rangle$, with $|a\rangle=|4d\rangle$ in one experiment[7] and $|a\rangle=|5s\rangle$ in the other.[6] No autoionizinglike behavior, i.e., no asymmetry was observed in either and the matter has remained a bit of a mystery. The explanation follows directly from our results. First of all, the term $D^{(4)}_{\tilde{a}\,\tilde{g}}$ had been neglected. Including this term in a realistic calculation, we find q to be very large in both cases, being equal to 1.8×10^4 in one case[5] and 2.4×10^3 in the other.[7] A completely symmetric profile is thus to be expected and that is exactly what was observed by Feldman *et al.*[7] who report the absence of autoionizinglike behavior, recognizing at the same time that Raman processes have contributed to ionization. This is surprising only if the Raman contribution is viewed as separate from or insignificant compared to the autoionizinglike coupling. If we leave them out of our calculation, the resulting $q_{(3)}=5.1$ predicts a very asymmetric line shape. As we have shown above, $D^{(4)}_{\tilde{a}\,\tilde{g}}$ is just one of the couplings that connect the states under consideration and can not be regarded as a separate process. Thus what is commonly referred to as autoionizinglike behavior simply corresponds to the case in which the Raman coupling does not dominate the principal value integral. This leads to a reduced value of q which entails a more or less asymmetric profile; but the physics remains the same. The three diagrams do not contribute equally to the coupling of $|g\rangle$ with $|a\rangle$. Depending on proximity to near-resonances with intermediate atomic states one or two may dominate. All three, however, are inseparable parts of a coherent process.

This work was supported in part by the National Science Foundation, under Grant No. PHY-83-06263, and in part by U. S. Department of Energy Grant No. DE-FG03-87 ER60504.

[1]Yu. I. Heller and A. K. Popov, Opt. Commun. **18**, 449 (1976); Zh. Eksp. Teor. Fiz. **78**, 506 (1980) [Sov. Phys.—JETP **51**, 255 (1980)].

[2]Yu. I. Heller, V. F. Lukinykh, A. K. Popov, and V. V. Slabko, Phys. Lett. **82A**, 4 (1981)].

[3]P. E. Coleman and P. L. Knight, J. Phys. B **15**, L235 (1982).

[4]P. E. Coleman, P. L. Knight, and K. Burnett, Opt. Commun. **42**, 171 (1982); P. L. Knight, M. A. Lauder, P. M. Radmore, and B. J. Dalton, Acta Phys. Austriaca **56**, 103 (1984), and references therein.

[5]S. S. Dimov, L. I. Pavlov, K. V. Stamenov, and G. B. Altshuller, Opt. Quantum Electron. **15**, 305 (1983).

[6]S. S. Dimov, L. I. Pavlov, K. V. Stamenov, Yu. I. Heller, and A. K. Popov, Appl. Phys. B **30**, 35 (1983).

[7]D. Feldman, G. Otto, D. Petring, and K. H. Welge, J. Phys. B **19**, 269 (1986).

[8]G. Alber and P. Zoller, Phys. Rev. A **27**, 1713 (1983).

[9]P. Lambropoulos and P. Zoller, Phys. Rev. A **24**, 379 (1981).

[10]P. Lambropoulos, Adv. At. Mol. Phys. **12**, 87 (1976).

[11]M. L. Goldberger and K. M. Watson, *Collision Theory* (Wiley, New York, 1964), Chap. 8.