Manipulation of the Line Shape and Final Products of Autoionization through the Phase of the Electric Fields

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We show that the autoionization line shape can be modified by the choice of two laser intensities and the relative phase. Under well specified conditions, proper choice of the phase can lead to selective cancellation of the transition to the discrete or the continuum part of the state. Application of the idea to a multichannel problem in the rare gases also provides significant effects.

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Recent work [1-6] on the possibility of controlling the final state of photoabsorption through the phases of electromagnetic fields attests to the relevance of this problem from a basic as well as a practical point of view. A key motivation behind such investigations is the control of the relative amount of final products when the transition in question leads to more than one channel. A scheme of particular significance in this context is the excitation through the simultaneous presence of a one- and a three-photon transition, because of the simplicity with which one can change the phase of the third harmonic relative to that of the pump.

An autoionizing state (AIS) represents a prototype of channel interactions. Even in the simplest case of an isolated AIS, we have the interaction of a discrete state with a continuum. In the more general case, we have the interaction of two or more channels with the possibility of final state products corresponding to different ionic states. It is our purpose in this paper to demonstrate through general formal arguments, as well as through specific calculations, corresponding to a multichannel situation in the rare gases, that the combination of a one- with a three-photon transition with a controllable relative phase can lead to profound alteration of the line shape, which entails alteration of the relative contribution of the participating channels.

We begin with the simplest case of an isolated AIS $|1\rangle$, to which the single-photon transition from an initial state $|0\rangle$ is describable in terms of a Rabi frequency $\Omega=\varepsilon_1 D_{10}$, a dipole transition $\varepsilon_1 D_{c0}$ directly into the continuum, an autoionizing width Γ_1, and an asymmetry parameter q. The two states can also be coupled through a three-photon transition characterized by a three-photon Rabi frequency $\Omega^{(3)}=\varepsilon_3^3 D_{10}^{(3)}$, a three-photon transition $\varepsilon_3^3 D_{c0}^{(3)}$ directly into the continuum, and another asymmetry parameter $q^{(3)}$ (in general different from q). The quantities D_{ij} and $D_{ij}^{(3)}$ indicate electric dipole, and effective three-photon dipole, matrix elements, respectively, with all the necessary coefficients so as to yield the Rabi frequencies defined above, assuming that the total externally imposed ac electric field has the form $E(t)=(\varepsilon_1 e^{i\omega_1 t}+\varepsilon_3 e^{i(\omega_3 t+\phi)})$ +c.c., where $\omega_1=3\omega_3$ and ϕ is a fixed but controllable relative phase. An overall absolute phase can of course be factored out and has no effect on the transition.

With the above expression for the field, we derive the following set of density matrix equations describing the complete evolution of the system in time:

$$\frac{\partial}{\partial t}\sigma_{00}=-\gamma\sigma_{00}+2\,\mathrm{Im}\left[\left\{\Omega^{(3)}\left(1-\frac{i}{q^{(3)}}\right)+e^{i\phi}\Omega\left(1-\frac{i}{q}\right)\right\}\sigma_{10}\right], \tag{1}$$

$$\frac{\partial}{\partial t}\sigma_{11}=-\Gamma_1\sigma_{11}-2\,\mathrm{Im}\left[\left\{\Omega^{(3)}\left(1+\frac{i}{q^{(3)}}\right)+e^{i\phi}\Omega\left(1+\frac{i}{q}\right)\right\}\sigma_{10}\right], \tag{2}$$

$$\left[\frac{\partial}{\partial t}-i\delta+\frac{1}{2}(\gamma+\Gamma_1)\right]\sigma_{10}=-i\left[\Omega^{(3)}\left(1-\frac{i}{q^{(3)}}\right)+e^{-i\phi}\Omega\left(1-\frac{i}{q}\right)\right]\sigma_{00}+i\left[\Omega^{(3)}\left(1+\frac{i}{q^{(3)}}\right)-e^{-i\phi}\Omega\left(1+\frac{i}{q}\right)\right]\sigma_{11}, \tag{3}$$

where γ is the effective ionization width of $|0\rangle$ caused by the combined fields, due to the direct transition into the continuum, and is given by $\gamma=|\Omega^{(3)}/q^{(3)}(\Gamma_1/2)^{1/2}+e^{i\phi}\Omega/q(\Gamma_1/2)^{1/2}|^2+\gamma_{\mathrm{incoherent}}^{(3)}$, with $\gamma_{\mathrm{incoherent}}^{(3)}$ being the ionization width due to the possible presence of an additional channel of ionization directly into a continuum not coupled to the discrete state. This may or may not be important, depending on the particular state and atom. The detuning δ from resonance is defined as $\delta=\omega_1-\hbar^{-1}(E_1-E_0)$. The derivation of the above equations is based on the Refs. [7-9].

(I) Weak field limit.— In this limit, corresponding to $\Omega,\Omega^{(3)}\ll\Gamma_1$ and obtained under the conditions $\sigma_{00}(t)\simeq 1$, $\sigma_{11}(t)\simeq 0$, and $\partial\sigma_{10}(t)/\partial t=0$, the ionization P is given by a single rate, which can be written in the two equivalent forms:

$$P=-\dot{\sigma}_{11}(t)=\frac{2}{\epsilon^2+1}\left|\frac{\Omega^{(3)}}{q^{(3)}(\Gamma_1/2)^{1/2}}(q^{(3)}+\epsilon)+e^{i\phi}\frac{\Omega}{q(\Gamma_1/2)^{1/2}}(q+\epsilon)\right|^2=\frac{2}{\epsilon^2+1}|\varepsilon_3^3D_{c0}^{(3)}(q^{(3)}+\epsilon)+e^{i\phi}\varepsilon_1D_{c0}(q+\epsilon)|^2, \tag{4}$$

where ϵ is the normalized detuning defined by $\epsilon=\delta/(\Gamma_1/2)$. For $D_{c0}^{(3)}=0$ this equation reduces to the simple case of a single AIS excited by a weak single-photon transition as given by Fano [7,10].

On the basis of the above expressions, it is to be expected that the effect of ϕ on the line shape will depend on the relative magnitudes of the Rabi frequencies and the q parameters. We can always assume that the field amplitudes are chosen so as to make either the two Rabi frequencies or the two transitions into the continuum equal. More generally, we can control the ratio of the single-photon to the three-photon transition through the magnitude of the field amplitudes. That leaves q and $q^{(3)}$ as the two parameters which are fixed by the choice of the particular atom and state. After exploration of the effect of these parameters on the line shape, we illustrate some representative cases in Figs. 1(a)–1(d).

An extreme case corresponds to $q=-q^{(3)}$. Then, if we arrange the field amplitudes so that $|\Omega|=|\Omega^{(3)}|$, as shown in Fig. 1(a), a change of ϕ from π to 0 causes a peak in the photoabsorption line shape to become a dip (window) and vice versa. In the absence of an incoherent channel of ionization [as has been assumed in the calculation leading to Fig. 1(a)] the minimum of the dip is exactly zero, which implies complete stabilization of the AIS against autoionization. If, on the other hand, q and $q^{(3)}$ have the same sign (not necessarily the same magnitudes), a change of the phase from 0 to π does not cause a dramatic alteration of the line shape. An example is shown in Fig. 1(b). Note that in this particular figure, since we have set $q=q^{(3)}$, transition amplitudes cancel not only at the minimum but for all detunings when $\phi=\pi$.

A rather interesting scenario emerges through a careful examination of Eq. (4). We first note that for $\varepsilon_3^3D_{c0}^{(3)}=\varepsilon_1D_{c0}$, it reduces to

$$P\propto\frac{1}{\epsilon^2+1}|(q^{(3)}+\epsilon)+e^{i\phi}(q+\epsilon)|^2. \tag{5}$$

If we choose now $\phi=\pi$, we obtain $P\propto|q^{(3)}-q|^2/(\epsilon^2+1)$, which implies a completely symmetric Lorentzian line shape, irrespective of the values of q and $q^{(3)}$. The physical phenomenon underlying this result is the complete cancellation of the direct transition into the continuum. Partial cancellation is obtained as ϕ tends from 0 to π, as illustrated in Fig. 1(c).

Along similar lines of reasoning, we can obtain the conditions under which we can cancel the transition to the discrete part leaving only the transition directly into the continuum. This is achieved by adjusting the field strengths so that $\varepsilon_3^3D_{c0}^{(3)}q^{(3)}=\pm\varepsilon_1D_{c0}q$ and choosing $\phi=\pi$ or 0, respectively. We obtain

$$P\propto\frac{\epsilon^2}{\epsilon^2+1}|\varepsilon_3^3D_{c0}^{(3)}\mp\varepsilon_1D_{c0}|^2=\frac{\epsilon^2}{\epsilon^2+1}|\varepsilon_1D_{c0}|^2\left(1-\frac{q}{q^{(3)}}\right)^3, \tag{6}$$

which indicates a flat line shape with a window at $\epsilon=0$ whose depth depends on the value of the ratio $q/q^{(3)}$. The case $q=q^{(3)}$ is quite special in that ionization is then turned off completely for all ϵ [see also Fig. 1(b)], as is also the case with Eq. (5). An example illustrating the

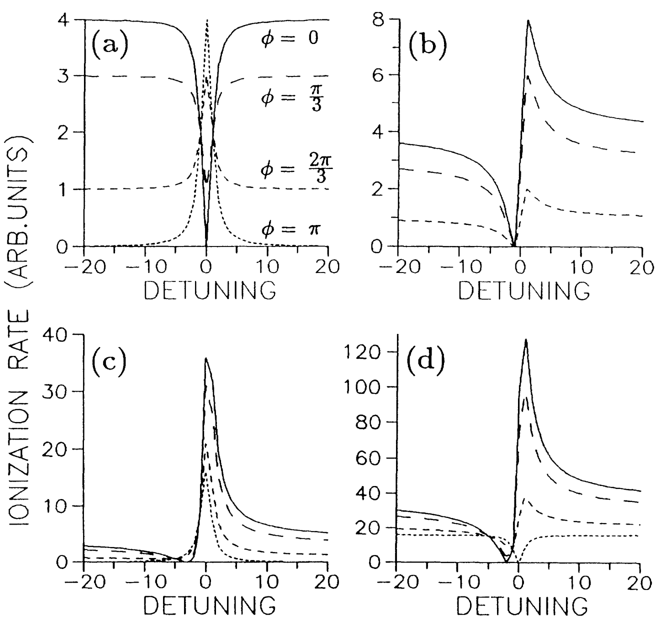

FIG. 1. (a),(b) Change of the autoionization line shape in a weak field as a function of the dimensionless detuning. (a) $\varepsilon_3^3D_{c0}^{(3)}=1$, $q^{(3)}=1$, $\varepsilon_1D_{c0}=1$, $q=-1$. (b) $\varepsilon_3^3D_{c0}^{(3)}=1$, $q^{(3)}=1$, $\varepsilon_1D_{c0}=1$, $q=1$. (c) Cancellation of the continuum part of an autoionizing state. $\varepsilon_3^3D_{c0}^{(3)}=1$, $q^{(3)}=5$, $\varepsilon_1D_{c0}=1$, $q=1$. (d) Cancellation of the bound part of an autoionizing state. $\varepsilon_3^3D_{c0}^{(3)}=1$, $q^{(3)}=5$, $\varepsilon_1D_{c0}=5$, $q=1$. For all of the figures, the same line type corresponds to the same relative phase given in (a). Note that no ionization occurs at any detunings for $\phi=\pi$ in (b).

transition from $\phi=0$ to π is given in Fig. 1(d), for $q=1$ and $q^{(3)}=5$.

(2) Moderate and high intensity region.— In this intensity regime, which corresponds to $\Omega, \Omega^{(3)} \gtrsim \Gamma_1$, the time-dependent behavior of the system must be obtained through the solution of the denisty matrix equations (1)-(3). Since related experiments must be envisioned in terms of pulsed beams, the pulse duration T_L is an important quantity, because if $T_L\Gamma_1 \ll 1$ the spectrum is flat. If, on the other hand, $T_L\gamma \gg 1$, the system is ionized completely during the pulse (saturation) and the structure of the line shape is obliterated, irrespective of the value of ϕ. We present here as an illustration two cases of moderate field excitation ($\Omega^{(3)}=\Omega=\Gamma_1/5$) and $\Gamma_1 T_L=10$ which corresponds to a pulse duration sufficiently long for the line shape to be fully developed in the weak field limit (for detailed discussion of the effect of $\Gamma_1 T$ see Ref. [8]). Thus the line shapes shown in Figs. 2(a) and 2(b) represent the effect of the moderate intensity combined with that of the phase. As a general comment for the case of moderate to high intensity, we note that the structure of Eqs. (1)-(3) suggests that the sensitivity to ϕ is maximum when $\Omega^{(3)}/q^{(3)} \simeq \Omega/q$.

(3) The effect of incoherent ionization.— An obvious example of such a channel is the $l=3$ continuum reached via the three-photon ionization from an ns^2 ground state. This continuum does not interact with an $l=1$ autoionizing state, and each contribution appears as an incoherent and separate ionization background. If such channels are stronger than that leading to the three-photon resonance, the resonance structure itself would be very small compared with the incoherent background. The question is whether the interference windows due to the phase ϕ are below such a background, even if most of the resonance profile is above. This has to be decided case by case. It would depend on experimental resolution. However, selective calculations that we have performed with various $\gamma^{(3)}_{\mathrm{incoherent}}$'s indicate that this is not expected to represent a general difficulty.

(4) Application to a multichannel problem.— Having

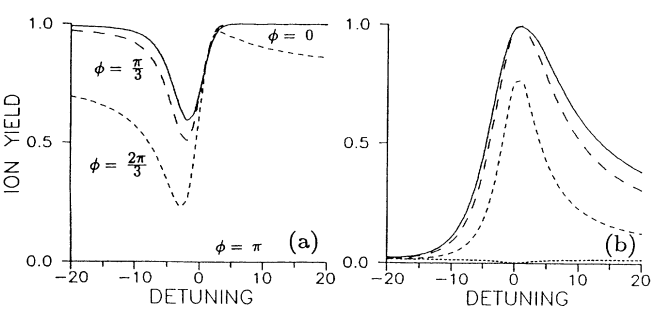

FIG. 2. Change of the line shape of an autoionizing state at the moderate intensity. $\Omega^{(3)}=\Omega=\Gamma_1/5$ and $\Gamma_1 T=10$. (a) $q^{(3)}=q=1$. (b) $q^{(3)}=10$, $q=5$. Note that ionization is completely suppressed for $\phi=\pi$ in (a).

established the basic ideas and quantitative relations in the previous formal context, we close this paper with an example of quantitative calculations in a realistic and much more complex context involving five coupled channels. We have chosen the excitation (by single- and three-photon transitions) of the autoionizing states of Xe. Autoionization here comes from the fine-structure coupling in the ionic core and not from the more typical configuration interaction found, for example, in atoms with two valence electrons. Between the two ionic fine-structure states (corresponding to two ionization thresholds of the atom) we have a series of AIS converging to the upper threshold $P_{1/2}$. Here we cannot employ the simpler formalism discussed above. We must instead derive expressions for the combined "single- plus three-photon" amplitude including the phase ϕ, in terms of the complete five channel wave functions. For this, we need to also perform the double summation over intermediate states necessary in the three-photon amplitude. We do this through the formalism of multichannel quantum defect theory (MQDT) as extended in previous work [11] to describe three-photon processes.

Formal expressions [11] for this problem are too lengthy to be presented here. It should be noted at this point, however, that formally it is far from evident how much of an effect, if any, the variation of ϕ would produce in such a multichannel process. It is only the intuition gained through the previous formalism that leads to the expectation of similar phenomena since the underlying fundamental physics is quite similar. We do in any case expect the effect to depend on the relative magnitude of the single- and three-photon transitions, as controlled by the amplitudes of the two fields.

Our expectations are indeed fulfilled as demonstrated by the results presented in Figs. 3(a)-3(c). The photon energy range of the figures spans two AIS, one fairly broad and one quite narrow. As a point of reference, we fix the single-photon intensity at 10 W/cm^2 and study the change of the line shape as the three-photon intensity is varied from 10^9 to 10^{10} W/cm^2. We have also plotted on each figure the line shape of the incoherent sum of ionization produced by the two beams separately. In all cases reported on these figures, the incoherent sum lies between the results for $\phi=0$ and $\phi=\pi$, indicating that the coherent effect of the two beams involves more than mere alteration of the line shape of the process. In Fig. 3(c), for example, the amount of ionization over most of the energy range is significantly enhanced (by more than a factor of 2) for $\phi=\pi$ and decreased by about as much for $\phi=0$. The most dramatic effect in Fig. 3(c), however, is the complete suppression of the autoionizing structure for $\phi=0$. And this suppression extends over the energy range of two of the three AIS included in that figure. We have thus a more complicated version of cancellation of the resonance discussed earlier, except that here the context is too complex to simply identify the effect as the

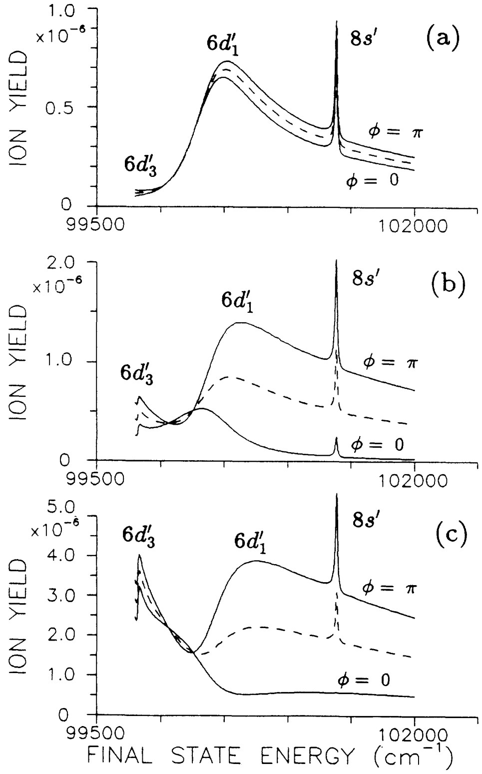

FIG. 3. Change of the autoionization line shape in the weak field limit for Xe. $I_1=10$ W/cm^2 [fixed through (a) to (c)]. 5 ns square pulse. (a) $I_3=10^9$ W/cm^2. (b) $I_3=5\times10^9$ W/cm^2. (c) $I_3=10^{10}$ W/cm^2. Dashed line corresponds to the incoherent ionization by the two fields. $8s'$, $6d_1'$, and $6d_3'$ in the figures correspond to the autoionizing states $[P_{1/2}]8s_{1/2}(J=1)$, $[P_{1/2}]6d_{3/2}(J=1)$, and $[P_{1/2}]6d_{5/2}(J=3)$, respectively.

cancellation of a discrete part. The distinction between discrete and continuum parts is not meaningful here.

Through the simplest model of autoionization as well as a multichannel problem, we have demonstrated that the line shape and final products of autoionization can be manipulated to a very significant degree through the relative phase of two external light beams. These effects provide an interesting vehicle for the study of intra-atomic interferences, and the control of final products. They can also be related to questions of lasing without inversion through AIS. Clearly the externally manipulated phase of the field sets up an interference which mirrors the phase with which discrete and continuum parts of the wave function combine in the bare atom to create the structure in the continuum. In the multichannel problem, it is the phases of various channels that enter in the interference. Restrictions on space allow us to briefly mention here one additional result. Through the manipulation of the phase for ionization above both thresholds, we have obtained effects as large as a factor of 5 on the ratio of the two final ionic states ($P_{3/2}$ and $P_{1/2}$) in Xe. Finally, the scheme employed in our analysis is readily implementable in a broad wavelength range widely available through present day lasers and their third harmonic.

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