Radiative decay of autoionizing states in laser fields. I. General theory

G. S. Agarwal
School of Physics, University of Hyderabad, Hyderabad 500134, India

S. L. Haan* and J. Cooper
Joint Institute for Laboratory Astrophysics, University of Colorado and National Bureau of Standards, Boulder, Colorado 80309

(Received 27 October 1983)

A theory describing the radiative decay of an autoionizing state under strong pumping by a coherent field is developed. The theory systematically takes into account the radiative decay of the unperturbed continuum. The problem at hand corresponds to the case of strongly coupled bound states decaying to electron and photon continua with the two continua also weakly coupled to each other. A master equation describing the time evolution of the atomic system is derived, and its exact solution under arbitrary initial conditions is given. The effect of radiative decay on the Fano profiles and photoelectron spectra is analyzed in detail. The time development of the system is also examined. The radiative decay of the autoionizing state and the unperturbed continuum changes the spectra in a significant way. The characteristics of the spectra are correlated with the dressed states (with complex energies) of the system. The changes in the structure of the dressed states as a function of the system parameters such as the spontaneous-emission rate and laser intensity are discussed in detail.

I. INTRODUCTION

Fano, in his well-known paper,[1] considered the effect of configuration mixing on photoelectron spectra at energies near an autoionizing state and predicted asymmetric line profiles with asymmetry depending on the ratio of the certain matrix elements involving the autoionizing state, the bound state, and the continuum. Such profiles have been studied at length. Configuration mixing was also shown to be important in studies of the four-wave mixing[2–6] involving autoionizing states. Currently there is considerable interest in the study of the fluorescence[7–12] produced by such states—as such states lead to the laser action[13] which can be extended to the vacuum ultraviolet region, and in addition if the radiative decay of the autoionizing states is significant, then one has an optical method for studying the characteristics of the autoionizing states. One also must consider the excitation of the autoionizing states. The excitation process itself leads to several new features[14–23] particularly if the strength of the exciting laser is high. Note that any theory of the laser action in such a system should simultaneously treat all the coherent and incoherent processes.

Interference between different pathways of ionization in strong laser fields was noted theoretically by Beers and Armstrong[24] in 1975 in the context of multiphoton ionization, and in the same year Armstrong *et al.*[25] pointed out the similarity in formalism between multiphoton ionization and autoionization. In 1981 Lambropoulos and Zoller[14] studied autoionizing states in strong laser fields, and their results showed an interesting narrowing of the photoelectron spectrum near the Fano minimum. This narrowing was the primary topic of a letter by Rzążewski and Eberly,[15] who considered a very simple model system consisting of an initial state, autoionizing state, and continuum. They called the narrowing of the spectral line a "confluence of coherences." Subsequently, Agarwal and co-workers[8–10] showed how spontaneous radiative decay affects the coherence and can be used as a probe of the system.

Andryushin *et al.*[21] studied a model which included decay processes not included in the model of Rzążewski and Eberly, and showed that these new channels give a minimum width to the photoelectron spectral lines. They also considered the case of two autoionizing states. Coleman and Knight[16] have examined population trapping and laser-induced continuum structure, and Kim and Lambropoulos[19] have studied configuration mixing in multiphoton ionization and have shown how the autoionization formalism applies in some cases but not others.

Eberly, Rzążewski, and Agassi[17] have considered off-diagonal relaxation processes such as weak elastic collisions and finite laser bandwidths, and have shown how such processes affect the electron spectrum. Several groups have studied fluorescence from autoionizing states. Crance and Armstrong[23] considered one-photon decay processes for a slightly generalized system, and Lewenstein *et al.*[11] have examined the photon spectrum for the recycling case and, more recently, the time development of the photoemission spectra and photon yield. Recently, Agassi has also studied the photoelectron and photoemission spectra,[26] and, using complex dressed states similar to those discussed in this paper, has obtained several results equivalent to those detailed here.

We have carried out a detailed investigation of the decay of autoionizing states in the presence of a coherent pumping mechanism (which can be quite strong) and the results of our investigations are presented in the present series of two papers. The organization of this paper is as follows: In Sec. II we present our model and work out the dynamical equations describing the behavior of the atomic system. In Sec. III we show how these dynamical equations can be solved exactly. We give the solution in terms of certain auxiliary matrices which are chosen so that their dynamical equations are similar to the original dynamical equations, but with the significant difference that these dynamical equations are *factorizable.* As a first application of these solutions, we consider in Sec. IV the modification of Fano profiles due to radiative decay. The decay of the unperturbed continuum is shown to be very important in the existence of the Fano minimum in such profiles. The differences in the Fano profiles due to the radiative decay in two different channels are discussed. In Sec. V we examine the complex dressed states of the system as a function of various system parameters. Such dressed states with complex energies determine the structure of various photoelectron and photoemission spectra. The confluence of coherences discussed recently in the literature is shown to follow from the fact that in the absence of any radiative decay, one of the dressed states lies exactly at the position of Fano minimum for certain values of the field strength. In Sec. VI we calculate the photoelectron spectra in the long-time limit and show how the spectra are affected by spontaneous radiative decay. We conclude the main text of this paper in Sec. VII with a short discussion of the time development of the ground-state population. Finally, in the Appendix we discuss the connection between our master equation results and phenomenological decay equations. A study of the photons ejected by the system is deferred to the second paper.

II. BASIC MODEL AND DYNAMICAL EQUATIONS

In order to see the main features that emerge due to the radiative decay of autoionizing states, we consider the simplest possible situation, shown schematically in Fig. 1. We consider one autoionizing state $|a\rangle$, with energy E_a, interacting with the unperturbed continuum of states $|E)$. We assume that the state $|a\rangle$ is resonantly coupled to the state $|i\rangle$ by a laser field of frequency ω_l. The laser field also couples the states $|E)$ to $|i\rangle$. We also allow for radiative decay of $|a\rangle$ and $|E)$. In order to keep the analysis general, we allow the possibility of two radiative channels, i.e., we consider decay to both $|i\rangle$ and $|f\rangle$. This will allow us to treat the various special cases. We will further assume that the state $|f\rangle$ can decay incoherently at the rate ν to the state $|i\rangle$. All the energies of the atomic system are measured from the level $|i\rangle$. The total Hamiltonian of our atomic system and the radiation fields (both vacuum and the coherent driving fields) can be written as

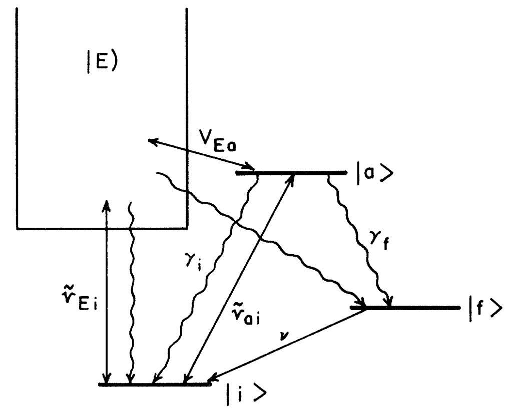

FIG. 1. Schematic diagram of the energy levels and interactions for the system of interest.

$$H=E_a|a\rangle\langle a|+\int E|E)(E|dE+E_f|f\rangle\langle f|+\int[V_{Ea}|E)\langle a|+\text{H.c.}]dE$$
$$+\left[\int[\tilde{v}_{Ei}|E)\langle i|e^{-i\omega_l t}+\text{H.c.}]dE+(\tilde{v}_{ai}|a\rangle\langle i|e^{-i\omega_l t}+\text{H.c.})\right]$$
$$+\sum_{k,s}\omega_k a_{ks}^{\dagger}a_{ks}-\int[|E)\langle i|\vec{d}_{Ei}\cdot\vec{E}_{\text{vac}}^{(+)}+\text{H.c.}]dE$$
$$-\int[|E)\langle f|\vec{d}_{Ef}\cdot\vec{E}_{\text{vac}}^{(+)}+\text{H.c.}]dE-[(\vec{d}_{ai}+\vec{d}_{af})\cdot\vec{E}_{\text{vac}}^{(+)}+\text{H.c.}]\ , \tag{2.1}$$

where $\vec{E}_{\text{vac}}^{(+)}$ is the positive-frequency part of the electric field operator associated with the vacuum field and $\vec{d}_{\alpha\beta}$ is the dipole matrix element. In writing (2.1), the rotating wave approximation has been made and $\hbar$ has been set to unity. Various terms in (2.1) have the following meaning—the first three terms represent the unperturbed Hamiltonian of the atom, the fourth term is responsible for the autoionization, the fifth term (enclosed in large parentheses) is the interaction with the coherent driving field, the sixth term is the free Hamiltonian of the radiation field, and the last three terms will account for the radiative decay.

Fano, in his classic work,[1] diagonalized the part responsible for autoionization. The new set of states will be denoted by $|E\rangle$ and we will refer to such states as Fano states. In what follows, we find it convenient to work with such states

$$|E\rangle=b(E,a)|a\rangle+\int dE'b(E,E')|E')\ , \tag{2.2}$$

where

$$b(E,a)=\frac{\sin\Delta}{\pi V_{Ea}}\ ,$$
$$b(E,E')=\frac{V_{E'a}}{\pi V_{Ea}}\frac{\sin\Delta}{(E-E')}-\cos\Delta\,\delta(E-E')\ , \tag{2.3}$$
$$\tan\Delta=\frac{-\pi\,|V_{Ea}|^2}{E-E_a-F(E)}$$

and where $F(E)$ is a small frequency shift due to the configuration interaction which we will ignore. The autoionization rate Γ is related to V_{Ea} by

$$\Gamma=2\pi\,|V_{Ea}|^2\ . \tag{2.4}$$

The Hamiltonian (2.1) can now be rewritten in terms of Fano states

$$\begin{aligned}H=&\int E\,|E\rangle\langle E\,|dE+E_f\,|f\rangle\langle f\,|+\sum_{k,s}\omega_{ks}a_{ks}^{\dagger}a_{ks}\\&+\int dE(v_{Ei}\,|E\rangle\langle i\,|e^{-i\omega_l t}+\text{H.c.})\\&-\int dE(\vec{\text{d}}_{Ei}\cdot\vec{\text{E}}_{\text{vac}}^{(+)}\,|E\rangle\langle i\,|+\text{H.c.})\\&-\int dE(\vec{\text{d}}_{Ef}\cdot\vec{\text{E}}_{\text{vac}}^{(+)}\,|E\rangle\langle f\,|+\text{H.c.})\ ,\end{aligned} \tag{2.5}$$

where now the matrix elements are in terms of Fano states:

$$v_{Ei}=\langle E\,|v\,|i\rangle\ . \tag{2.6}$$

The new matrix elements and the old matrix elements can be related by introducing Fano's asymmetry parameter q

$$\begin{aligned}q_i&\cong\frac{\langle a\,|\tilde{v}\,|i\rangle}{\pi\langle a\,|V\,|E\rangle\langle E\,|\tilde{v}\,|i\rangle}\ ,\\q_f&\cong\frac{\langle a\,|\tilde{v}\,|f\rangle}{\pi\langle a\,|V\,|E\rangle\langle E\,|\tilde{v}\,|f\rangle}\ .\end{aligned} \tag{2.7}$$

The details can be found in Fano's work. Here we quote the result:

$$v_{iE}\approx\tilde{v}_{ia}B_{Ea}\approx\tilde{v}_{iE}q_i\pi V_{Ea}B_{Ea}\ , \tag{2.8}$$

where

$$B_{Ea}=b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_i}\right]\ . \tag{2.9}$$

Similarly the matrix element for the other transition $|E\rangle\leftrightarrow|f\rangle$ will be

$$\begin{aligned}v_{fE}&\approx\tilde{v}_{fa}C_{Ea}\ ,\\C_{Ea}&=b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_f}\right]\ .\end{aligned} \tag{2.10}$$

The next major step is to eliminate the degrees of freedom associated with the vacuum of the radiation field. This can be done using the master equation techniques. The derivation of the master equation in the Born and Markov approximations (with regard to the interaction with the vacuum of the radiation field) is fairly standard[27] and we quote the result. Let ρ be the density matrix for the atomic system alone. On making the transformation to the rotating frame the dynamical equation of ρ is found to be

$$\begin{aligned}\frac{\partial\rho}{\partial t}=&-i[H_{\text{coh}},\rho]-\frac{\gamma_i}{2}(A_i^{\dagger}A_i\rho-2A_i\rho A_i^{\dagger}+\rho A_i^{\dagger}A_i)\\&-\frac{\gamma_f}{2}(A_f^{\dagger}A_f\rho-2A_f\rho A_f^{\dagger}+\rho A_f^{\dagger}A_f)\ ,\end{aligned} \tag{2.11}$$

where the operators A_i and A_f are given by

$$\begin{aligned}A_i&=\int dE\,|i\rangle\langle E\,|B_{Ea}\ ,\\A_f&=\int dE\,|f\rangle\langle E\,|C_{Ea}\end{aligned} \tag{2.12}$$

and the decay rates γ_i,γ_f are

$$\begin{aligned}\gamma_i&=\frac{4}{3}\,|d_{ai}|^2\frac{\omega_{ai}^3}{c^3}\ ,\\\gamma_f&=\frac{4}{3}\,|d_{af}|^2\frac{\omega_{af}^3}{c^3}\ .\end{aligned} \tag{2.13}$$

We have denoted the coherent part of the interactions by H_{coh},

$$\begin{aligned}H_{\text{coh}}=&\int(E-\omega_l)\,|E\rangle\langle E\,|dE\\&+\int(v_{Ei}\,|E\rangle\langle i\,|+\text{H.c.})dE\ .\end{aligned} \tag{2.14}$$

The incoherent transition from $|f\rangle$ to $|i\rangle$ can now be incorporated by modifying (2.11) to

$$\begin{aligned}\frac{\partial\rho}{\partial t}=&-i[H_{\text{coh}},\rho]-\frac{\gamma_i}{2}(A_i^{\dagger}A_i\rho-2A_i\rho A_i^{\dagger}+\rho A_i^{\dagger}A_i)\\&-\frac{\gamma_f}{2}(A_f^{\dagger}A_f\rho-2A_f\rho A_f^{\dagger}+\rho A_f^{\dagger}A_f)\\&-\frac{\nu}{2}(\,|f\rangle\langle f\,|\rho-|i\rangle\langle f\,|\rho\,|f\rangle\langle i\,|+\text{H.c.})\ .\end{aligned} \tag{2.15}$$

For the model system shown in Fig. 1, (2.15) is our basic dynamical equation characterizing the decay of the autoionizing states in the presence of a laser field, which could be of arbitrary intensity. Note that the radiative coupling between the unperturbed continuum $|E)$ and $|i\rangle$ and $|f\rangle$ has been completely taken into account in (2.15) since the seventh and eighth terms in Eq. (2.1) describe this decay. However, if we were to ignore such a decay, then one would still have the master equation (2.15) but with

$$\begin{aligned}A_i&\to\int dE\,|i\rangle\langle E\,|b_{Ea}\ ,\\A_f&\to\int dE\,|f\rangle\langle E\,|b_{Ea}\ .\end{aligned} \tag{2.16}$$

We will refer to the "decay" of the unperturbed continuum as "virtual recombination" since this decay does not imply the physical ejection and reabsorption of an electron.[28] In order to separate out the effects connected with this continuum interaction, we will take

$$A_i = \int dE \, | i \rangle \langle E | \, b_{Ea} \left[1 + \frac{2(E - E_a)}{\Gamma q_i'} \right] \tag{2.17}$$

or

$$B_{Ea} = b_{Ea} \left[1 + \frac{2(E - E_a)}{\Gamma q_i'} \right]$$

where appropriate.

The limits $q_i' \to q_i$ and $q_i' \to \infty$ correspond, respectively, to virtual recombination being included or ignored.

III. EXACT SOLUTION OF THE MASTER EQUATION UNDER ARBITRARY INITIAL CONDITIONS

In order to study the various features of the electrons and photons emitted from the autoionizing states, we need to know the solution of (2.15) under a variety of initial conditions. For this purpose we write (2.15) in terms of the various matrix elements of ρ,

$$\dot{\rho}_{ff} = \left[\frac{\gamma_f}{2} \int dE_1 \int dE \, C^*_{E_1 a} C_{Ea} \rho_{EE_1} + \text{c.c.} \right] - \nu \rho_{ff} \,, \tag{3.1}$$

$$\dot{\rho}_{ii} = \left[\left[-i \int v^*_{Ei} \rho_{Ei} dE + \frac{\gamma_i}{2} \int dE_1 \int dE \, B^*_{E_1 a} B_{Ea} \rho_{EE_1} \right] + \text{c.c.} \right] + \nu \rho_{ff} \,, \tag{3.2}$$

$$\dot{\rho}_{if} = -i \int v^*_{Ei} \rho_{Ef} dE - \frac{\nu}{2} \rho_{if} \,, \tag{3.3}$$

$$\dot{\rho}_{E_1 f} = -i \Delta_{E_1} \rho_{E_1 f} - i v_{E_1 i} \rho_{if} - \frac{\gamma_i}{2} \int dE \, B^*_{E_1 a} B_{Ea} \rho_{Ef} - \frac{\gamma_f}{2} \int dE \, C^*_{E_1 a} C_{Ea} \rho_{Ef} - \frac{\nu}{2} \rho_{E_1 f} \,, \tag{3.4}$$

$$\dot{\rho}_{E_1 i} = -i \Delta_{E_1} \rho_{E_1 i} - i \left[v_{E_1 i} \rho_{ii} - \int dE \, \rho_{E_1 E} v_{Ei} \right] - \frac{\gamma_i}{2} \int dE \, B^*_{E_1 a} B_{Ea} \rho_{Ei} - \frac{\gamma_f}{2} \int dE \, C^*_{E_1 a} C_{Ea} \rho_{Ei} \,, \tag{3.5}$$

$$\dot{\rho}_{E_1 E_2} = -i(E_1 - E_2) \rho_{E_1 E_2} - i v_{E_1 i} \rho_{iE_2} + i v^*_{E_2 i} \rho_{E_1 i} - \frac{\gamma_i}{2} \int dE \, B^*_{E_1 a} B_{Ea} \rho_{EE_2}$$
$$- \frac{\gamma_f}{2} \int dE \, C^*_{E_1 a} C_{Ea} \rho_{EE_2} - \frac{\gamma_i}{2} \int dE \, B_{E_2 a} B^*_{Ea} \rho_{E_1 E} - \frac{\gamma_f}{2} \int dE \, C_{E_2 a} C^*_{Ea} \rho_{E_1 E} \,, \tag{3.6}$$

where

$$\Delta_{E_1} = E_1 - \omega_l \,.$$

This set of equations is extremely complex and a direct solution is not immediately evident. However, a solution of (3.1)–(3.6) can be constructed in terms of certain auxiliary matrices. It should be remembered that the master equation (2.15) conserves the trace, i.e.,

$$\text{Tr}\rho(t) = 1 = \int \rho_{EE} dE + \rho_{ii} + \rho_{ff} \,. \tag{3.7}$$

In view of (3.7), one can see that the set of equations (3.1)–(3.6) reduces to two independent sets involving $\rho_{E_1 E_2}, \rho_{E_1 i}, \rho_{ii}$, and $\rho_{E_1 f}, \rho_{if}$. Let us denote the part of the density matrix with elements $\rho_{E_1 E_2}, \rho_{E_1 i}, \rho_{ii}, \rho_{iE_1}$ by Q, and we first consider the solution for Q.

A. Exact solution for $\rho_{E_1 E_2}, \rho_{E_1 i}, \rho_{ii}, \rho_{iE_1}$

An exact solution for Q can be constructed if we note that the equations for the elements of Q would be factorizable if the terms involving ρ_{ff} and ρ_{EE_1} in the equation for ρ_{ii} were absent. We now introduce an auxiliary matrix σ which is factorizable and which satisfies the equations for Q with the nonfactorizable terms neglected:

$$\sigma_{\alpha\beta}(t) = \psi_\alpha(t) \psi^*_\beta(t) \tag{3.8}$$

with the dynamical equations for the ψ's given by

$$\dot{\psi}_{E_1} = -i \Delta_{E_1} \psi_{E_1} - i v_{E_1 i} \psi_i - \frac{\gamma_i}{2} \int dE \, B^*_{E_1 a} B_{Ea} \psi_E$$
$$- \frac{\gamma_f}{2} \int dE \, C^*_{E_1 a} C_{Ea} \psi_E \,, \tag{3.9}$$

$$\dot{\psi}_i = -i \int v^*_{E_1 i} \psi_{E_1} dE_1 \,. \tag{3.10}$$

Thus formally one can show that

$$\frac{\partial Q}{\partial t} = LQ + \nu \rho_{ff}(t) \, | i \rangle \langle i | + g(t) \, | i \rangle \langle i | \,, \tag{3.11}$$

where L is defined by (3.9) and (3.10) through

$$\frac{\partial \sigma}{\partial t} = L\sigma \tag{3.12}$$

and

$$g(t) = \frac{\gamma_i}{2} \int dE_1 \int dE \, B^*_{E_1 a} B_{Ea} \rho_{EE_1}(t) + \text{c.c.} \tag{3.13}$$

In order to solve these equations we need the initial conditions. Assuming $\rho(0)$ to be a superposition of the states $| E \rangle$ and $| i \rangle$, i.e.,

$$\rho(0)=|\Phi\rangle\langle\Phi| ,\quad |\Phi\rangle=\int dE\, a_E|E\rangle+a_i|i\rangle , \tag{3.14}$$

we impose the conditions

$$\psi_E(0)=a_E,\ \ \psi_i(0)=a_i , \tag{3.15}$$

solve for σ, and denote such a solution by $\sigma^{(I)}$. We will denote the solution corresponding to the particular initial condition $a_E=0, a_i=1$ by σ. It is clear from (3.12) that the Laplace transform of σ is

$$\hat\sigma^{(I)}(z)=(z-L)^{-1}\sigma(0) ,\quad \hat\sigma(z)=(z-L)^{-1}|i\rangle\langle i| \tag{3.16}$$

whereas the Laplace transform of (3.11) is

$$\hat Q(z)=(z-L)^{-1}Q(0)+[\nu\hat\rho_{ff}(z)+\hat g(z)](z-L)^{-1}|i\rangle\langle i| . \tag{3.17}$$

On rewriting (3.17) in terms of σ's given by (3.16) we obtain

$$\hat Q(z)=\hat\sigma^{(I)}(z)+[\nu\hat\rho_{ff}(z)+\hat g(z)]\hat\sigma(z) . \tag{3.18}$$

The quantity appearing in square brackets is still unknown.

We can obtain another equation for $\hat g$ by using (3.18) in (3.13). Let us introduce

$$g_\sigma^{(I)}(t)=\gamma_i\int dE_1\int dE\, B^*_{E_1a}B_{Ea}\sigma^{(I)}_{EE_1}(t) ,\quad g_\sigma(t)=\gamma_i\int dE_1\int dE\, B^*_{E_1a}B_{Ea}\sigma_{EE_1}(t) . \tag{3.19}$$

Then

$$\hat g=\hat g^I_\sigma+(\nu\hat\rho_{ff}+\hat g)\hat g_\sigma . \tag{3.20}$$

A second relation follows from (3.7):

$$\frac{1}{z}-\hat\rho_{ff}=\mathrm{Tr}\hat Q(z)=\mathrm{Tr}\hat\sigma^{(I)}+(\upsilon\hat\rho_{ff}+\hat g)\mathrm{Tr}\hat\sigma . \tag{3.21}$$

Note that Eqs. (3.20) and (3.21) determine $\hat g$ and $\hat\rho_{ff}$ in terms of the known quantities. We have thus constructed a complete solution for Q provided σ is known:

$$\hat Q(z)=\hat\sigma^{(I)}(z)+\left[\frac{\nu/z+\hat g^{(I)}_\sigma(z)-\nu\mathrm{Tr}\hat\sigma^{(I)}(z)}{1+\nu\mathrm{Tr}\hat\sigma(z)-\hat g_\sigma(z)}\right]\hat\sigma(z) . \tag{3.22}$$

We defer the solution for σ to Sec. III C.

Let us consider some special cases of (3.22) which will be of interest later.

(i) $\gamma_f=0$. Then

$$\hat\rho_{ff}=0,\ \ \mathrm{Tr}\hat Q(z)=\frac{1}{z}$$

and

$$\hat Q(z)=\hat\sigma^{(I)}(z)+\left[\frac{1/z-\mathrm{Tr}\hat\sigma^{(I)}}{\mathrm{Tr}\hat\sigma}\right]\hat\sigma(z) . \tag{3.23}$$

If the atom is initially in state $|i\rangle$, then $\hat\sigma^{(I)}=\hat\sigma$ and

$$\hat Q(z)=\frac{\hat\sigma(z)}{z\,\mathrm{Tr}\hat\sigma(z)} . \tag{3.24}$$

(ii) $\gamma_i=0$. Then $\hat g=0$ and (3.22) yields

$$\hat Q(z)=\hat\sigma^{(I)}(z)+\left[\frac{\nu/z-\nu\mathrm{Tr}\hat\sigma^{(I)}(z)}{1+\nu\mathrm{Tr}\hat\sigma(z)}\right]\hat\sigma(z) \tag{3.25}$$

which in the limit $\nu\to 0$ reduces to

$$\hat Q(z)=\hat\sigma^{(I)}(z)\ \ (\nu=0,\gamma_i=0) . \tag{3.26}$$

B. Solution of (3.3) and (3.4)

We next consider the solution of (3.3) and (3.4). On defining

$$\rho_{E_1f}=e^{-(\nu/2)t}\Phi_{E_1},\ \ \rho_{if}=e^{-(\nu/2)t}\Phi_i , \tag{3.27}$$

we find the equations for Φ_i and Φ_E,

$$\dot\Phi_i=-i\int v^*_{E_1i}\Phi_{E_1}dE_1 , \tag{3.28}$$

$$\dot\Phi_{E_1}=-i\Delta_{E_1}\Phi_{E_1}-iv_{E_1i}\Phi_i-\frac{\gamma_i}{2}\int dE\, B^*_{E_1a}B_{Ea}\Phi_E-\frac{\gamma_f}{2}\int dE\, C^*_{E_1a}C_{Ea}\Phi_E , \tag{3.29}$$

which are identical in form to (3.9) and (3.10). Hence it is sufficient to know the solution of (3.9) and (3.10).

C. Solution of (3.9) and (3.10)

On taking the Laplace transforms of (3.9) and (3.10), we obtain an integral equation for ψ_E

$$\hat\psi_{E_1}+\int dE\sum_i K_i(E_1)L_i(E)\hat\psi_E=-iK_1(E_1)a_i+\frac{a_{E_1}}{z+i\Delta_{E_1}} , \tag{3.30}$$

where

$$K_1(E)=\frac{v_{Ei}}{z(z+i\Delta_E)},\ \ L_1(E)=v^*_{Ei} ,$$
$$K_2(E)=\left[\frac{\gamma_i}{2}\right]^{1/2}\frac{B^*_{Ea}}{z+i\Delta_E},\ \ L_2(E)=\left[\frac{\gamma_i}{2}\right]^{1/2}B_{Ea} ,$$
$$K_3(E)=\left[\frac{\gamma_f}{2}\right]^{1/2}\frac{C^*_{Ea}}{z+i\Delta_E},\ \ L_3(E)=\left[\frac{\gamma_f}{2}\right]^{1/2}C_{Ea} . \tag{3.31}$$

The integral equation (3.30) has a separable kernel and hence its solution can be obtained by matrix methods. Introducing the quantity

$$\hat\chi_i=\int dE\, L_i(E)\hat\psi_E , \tag{3.32}$$

multiplying (3.30) by $L_j(E_1)$ and integrating over E_1, we obtain

$$\hat\chi_j+\sum_j m_{ji}\hat\chi_i=-ia_im_{j1}+f_j , \tag{3.33}$$

$$m_{ji}=\int dE\, L_j(E)K_i(E)\ ,$$
$$f_j=\int dE\frac{L_j(E)a_E}{z+i\Delta_E}\ . \tag{3.34}$$

Hence the solution becomes

$$\hat{\chi}_i=\sum_j[(\mathbb{1}+\underline{m})^{-1}]_{ij}(-ia_i m_{j1}+f_j)\ , \tag{3.35}$$

$$\hat{\psi}_E\equiv -i\sum_i K_i(E)[(\mathbb{1}+\underline{m})^{-1}]_{i1}a_i$$
$$-\sum_{i,j}K_i(E)[(\mathbb{1}+\underline{m})^{-1}]_{ij}f_j+\frac{a_E}{z+i\Delta_E}\ . \tag{3.36}$$

For the initial conditions $a_i=1, a_E=0$, we obtain

$$\hat{\chi}_i(z)=i\{[\mathbb{1}+\underline{m}(z)]^{-1}\}_{i1}-i\delta_{i1} \tag{3.35$'$}$$

and

$$\hat{\psi}_E(z)=-i\sum_i K_i(E)\{[\mathbb{1}+\underline{m}(z)]^{-1}\}_{i1}\ . \tag{3.36$'$}$$

The matrix m can be computed using relations such as (2.8) and (2.10) and by assuming the flat structure of the initial continuum $|E)$. These calculations show that

$$m_{22}=\frac{\gamma_i}{\Gamma}\left[\frac{(1-i/q_i')^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_i'^2}\right],$$
$$m_{33}=\frac{\gamma_f}{\Gamma}\left[\frac{(1-i/q_f')^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_f'^2}\right],$$
$$m_{23}=m_{32}=\frac{(\gamma_i\gamma_f)^{1/2}}{\Gamma}\left[\frac{(1-i/q_i')(1-i/q_f')}{2z/\Gamma+1-i\alpha}+\frac{1}{q_i'q_f'}\right],$$
$$m_{11}=\frac{\Omega q_i^2}{z}\left[\frac{\Gamma}{2}\right]\left[\frac{(1-i/q_i)^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_i^2}\right],$$
$$\frac{V_{Ea}^*}{\tilde{v}_{Ei}^*}m_{12}=\frac{m_{21}zV_{Ea}}{\tilde{v}_{Ei}}=\left[\frac{\gamma_i}{2}\right]^{1/2}q_i\left[\frac{(1-i/q_i)(1-i/q_i')}{2z/\Gamma+1-i\alpha}+\frac{1}{q_iq_i'}\right],$$
$$\frac{V_{Ea}^*}{\tilde{v}_{Ei}^*}m_{13}=\frac{m_{31}zV_{Ea}}{\tilde{v}_{Ei}}=\left[\frac{\gamma_f}{2}\right]^{1/2}q_i\left[\frac{(1-i/q_i)(1-i/q_f')}{2z/\Gamma+1-i\alpha}+\frac{1}{q_iq_f'}\right], \tag{3.37}$$

where

$$\alpha=(2/\Gamma)(\omega_l-E_a),\ \ \Omega=(2\pi/\Gamma)\,|\tilde{v}_{Ei}|^2\ . \tag{3.38}$$

The determinant of the matrix $(\mathbb{1}+\underline{m})$ will be seen to determine the various features of the time-dependent and time-independent spectra and hence we list its value

$$\det(\mathbb{1}+\underline{m})=\left[\frac{\gamma_i}{\Gamma}+\frac{\Omega q_i^2\Gamma}{2z}\right]\left[\frac{(1-i/q_i)^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_i^2}+\frac{\gamma_f}{\Gamma}\frac{(1/q_i-1/q_f)^2}{2z/\Gamma+1-i\alpha}\right]+1+\frac{\gamma_f}{\Gamma}\left[\frac{(1-i/q_f)^2}{2z/\Gamma+1-i\alpha}+\frac{1}{q_f^2}\right], \tag{3.39}$$

where we have accounted for all recombination effects $q_i'=q_i, q_f'=q_f$. We will now use these exact solutions to discuss various features of the spectra of photons and photoelectrons.

IV. FANO PROFILES WITH RADIATION DAMPING

As a first application of our general results of Secs. II and III, we find how the usual Fano profiles change when the radiative decay of the autoionizing states is taken into account. In the absence of any radiative decay such profiles are given by

$$S_F(\epsilon)=(\epsilon+q)^2/(1+\epsilon^2),\ \ \epsilon=2(E-E_a)/\Gamma\ , \tag{4.1}$$

which for small values of q are highly asymmetric. In order to obtain such profiles from our analysis we evaluate $\lim_{t\to\infty}(d/dt)\rho_{EE}(t)$ to the lowest order in the laser field interaction, assuming that the atom was in the state $|i\rangle$ at $t=0$. From (3.35), we find $\hat{\psi}_E$ to lowest order in v_{Ei}:

$$\hat{\psi}_E(z)=-\frac{\tilde{v}_{Ei}i}{z(z+i\Delta_E)}\left[q_iB_{Ea}^*\pi V_{Ea}+z\left[\frac{\gamma_i}{2}\right]^{1/2}\frac{B_{Ea}^*D^{-1}(z)}{\tilde{v}_{Ei}}[m_{23}m_{31}-m_{21}(1+m_{33})]\right.$$
$$\left.+z\left[\frac{\gamma_f}{2}\right]^{1/2}C_{Ea}^*\frac{D^{-1}(z)}{\tilde{v}_{Ei}}[m_{32}m_{21}-(1+m_{22})m_{31}]\right], \tag{4.2}$$

$$D(z)=(1+m_{22})(1+m_{33})-m_{23}m_{32}\ . \tag{4.3}$$

[The second B_{Ea}^{*} and the C_{Ea}^{*} in (4.2) are related to the radiative decay, and thus involve q_i', q_f' as in Eq. (2.17).] Inverting (4.2) one finds that $\psi_E(t)$ has the form

$$\psi_E(t)=\frac{A}{i\Delta_E}+B\frac{e^{-i\Delta_E t}}{-i\Delta_E}+C(t)\ , \tag{4.4}$$

where the last term decays as $t\to\infty$. Thus

$$\lim_{t\to\infty}\frac{d}{dt}\,|\psi_E(t)|^2\to\lim_{t\to\infty}\left[B\frac{e^{-i\Delta_E t}A^*}{-i\Delta_E}+\text{c.c.}\right]$$

$$\to\pi\delta(\Delta_E)(BA^*+\text{c.c.})\ . \tag{4.5}$$

Expression (4.5) is still to be averaged over the density of final states. Note that while computing A and B we can now put $E=\omega_l$ and thus

$$\alpha\to\frac{2}{\Gamma}(\omega_l-E_a)=\frac{2}{\Gamma}(E-E_a)=\epsilon\ .$$

Note also that B is the residue of $z\hat{\psi}_E$ at $z=-i\Delta_E$ whereas A is the residue of $(z+i\Delta_E)\hat{\psi}_E$ at $z=0$. Because of $\delta(\Delta_E)$ in (4.5), these two residues are identical and hence

$$\lim_{t\to\infty}\frac{d}{dt}\,|\psi_E(t)|^2=2\pi\delta(\Delta_E)\,|A|^2\,|\tilde{v}_{Ei}|^2\ , \tag{4.6}$$

$$A=\lim_{z\to 0}\left[q_iB_{Ea}^{*}\pi V_{Ea}+z\left[\frac{\gamma_i}{2}\right]^{1/2}\frac{B_{Ea}^{*}D^{-1}(z)}{\tilde{v}_{Ei}}[m_{23}m_{31}-m_{21}(1+m_{33})]\right.$$
$$\left.+z\left[\frac{\gamma_f}{2}\right]^{1/2}C_{Ea}^{*}\frac{D^{-1}(z)}{\tilde{v}_{Ei}}[m_{32}m_{21}-(m_{22}+1)m_{31}]\right]\ , \tag{4.7}$$

where, as in (4.2), the second B_{Ea}^{*} and the C_{Ea}^{*} involve q_i' or q_f' in accord with Eq. (2.17). On simplification, we find

$$S_F(\epsilon)\equiv|A|^2=\frac{(q_i+\epsilon)^2+\left[\frac{\gamma_i}{\Gamma}\left[1-\frac{q_i}{q_i'}\right]+\frac{\gamma_f}{\Gamma}\left[1-\frac{q_i}{q_f'}\right]\right]^2}{\psi^2[(\epsilon-\Delta_a)^2+\eta^2]}\ , \tag{4.8}$$

where

$$\psi=1+\frac{\gamma_i}{\Gamma q_i'^2}+\frac{\gamma_f}{\Gamma q_f'^2}\ ,$$
$$\eta=\frac{1}{\psi}\left[1+\frac{\gamma_i}{\Gamma}+\frac{\gamma_f}{\Gamma}+\frac{\gamma_i\gamma_f}{\Gamma^2}\left[\frac{1}{q_i'}-\frac{1}{q_f'}\right]^2\right]\ , \tag{4.9}$$
$$\Delta_a=\frac{-2}{\psi}\left[\frac{\gamma_i}{\Gamma q_i'}+\frac{\gamma_f}{\Gamma q_f'}\right]\ .$$

The Δ_a and η are discussed in Sec. V, and are shown to represent the effective energy (relative to E_a) and half-width of the autoionizing state in units of $(\Gamma/2)$. Note that for $\gamma_i=\gamma_f=0$, we have the very simple result

$$|A|^2=\frac{(q_i+\epsilon)^2}{1+\epsilon^2}$$

which is just the classic result of Fano. If $\gamma_f=0$ but $\gamma_i\neq 0$, then $S_F(\epsilon)$ does not show a zero at $\epsilon=-q_i$ if virtual recombination is ignored ($q_i'\to\infty$), but the zero is restored when virtual recombination is included ($q_i'=q_i$). For nonzero γ_f, the Fano profiles do not show the zero at $\epsilon=-q_i$ when the virtual recombination is included unless $q_f=q_i$. We show a few modified Fano profiles in Figs. 2 and 3, which are self-explanatory.

V. COMPLEX DRESSED STATES OF THE SYSTEM

The time development of ψ_E can be obtained from the standard Laplace inversion

$$\psi_E(t)=\frac{1}{2\pi i}\int dz\, e^{zt}\hat{\psi}(z)\ , \tag{5.1}$$

where $\hat{\psi}$ is given in Eq. (3.35). The result depends on the pole at $z=-i\Delta_E$ and the zeros of $\det(1+m)$ [Eq. (3.38)]. This determinant is quadratic in z and its roots depend in a complicated manner on the field intensity, q's, and the spontaneous emission rates. Hence this section is devoted largely to a detailed study of the roots $z_\pm$ of (3.38).

It is clear that

$$\psi_E(t)=e^{-i\Delta_E t}\lim_{z\to -i\Delta_E+0^+}(z+i\Delta_E)\hat{\psi}_E(z)$$
$$+e^{z_+t}\lim_{z\to z_+}(z-z_+)\hat{\psi}_E(z)$$
$$+e^{z_-t}\lim_{z\to z_-}(z-z_-)\hat{\psi}_E(z)\ . \tag{5.2}$$

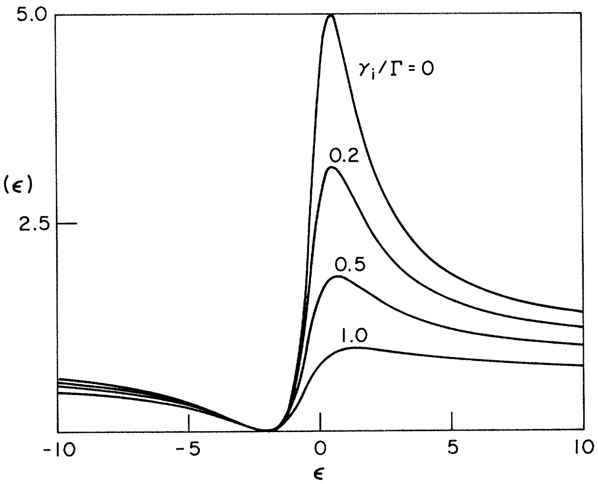

FIG. 2. Modified Fano profiles $S_F(\epsilon)$ for $q_i = q_i' = 2$, $\gamma_f = 0$, and $\gamma_i/\Gamma = 0$, 0.2, 0.5, and 1. The Fano minimum is retained at $\epsilon = -q_i$, but the maximum is washed out with increasing γ_i.

Thus $z_\pm$ represent complex dressed states of the system.

We will first examine the case when $\gamma_i = \gamma_f = 0$. In such a case (4.3) gives

$$D(z) = \Omega q_i^2 \left[(1 - i/q_i)^2 + \frac{2z/\Gamma + 1 - i\alpha}{q_i^2} \right] + \frac{2z}{\Gamma} \left[1 + \frac{2z}{\Gamma} - i\alpha \right] . \tag{5.3}$$

An analysis of (5.3) shows that if

$$\frac{2z_+}{\Gamma} = (i\alpha + iq_i), \quad \Omega = 1 + \frac{\alpha}{q_i} \tag{5.4}$$

then $D(z)$ vanishes. Thus for $\Omega = 1 + (\alpha/q_i)$, $(2z_+/\Gamma) = i(\alpha + q_i)$ is a root of (5.3). The other root is also easily found to be

$$\frac{2z_-}{\Gamma} = -iq_i - \left[2 + \frac{\alpha}{q_i} \right] . \tag{5.5}$$

Therefore, in the absence of any radiative decay, one of the roots of $D(z)$ lies on the imaginary axis, whereas the other root has always the finite width.

The root which lies on the imaginary axis represents a state which is stable against decay. This phenomonon was noted by Beers and Armstrong[24] in the context of multiphoton ionization, and was dubbed "confluence of coherences" by Rzążewski and Eberly[15] since the state lies directly at the Fano minimum. For Ω near but not equal to the confluence value $1 + (\alpha/q)$ the state decays only very slowly, and consequently the photoelectron spectrum would exhibit a very narrow spike at the energy of this state. This also leads to the so-called population trapping.[16]

Population cannot be trapped for nonzero γ, since spontaneous emission moves the root off the imaginary axis. For small values of γ, one can calculate this movement to lowest order in γ. Writing

$$D(\mathfrak{z}) = D_0(\mathfrak{z}) + \gamma_i D_i(\mathfrak{z}) + \gamma_f D_f(\mathfrak{z}) + \gamma_i \gamma_f D_{if}(\mathfrak{z}), \quad \mathfrak{z} = \frac{2z}{\Gamma} \tag{5.6}$$

and the root as

$$\mathfrak{z} = \mathfrak{z}_0 + \frac{\gamma_i}{\Gamma} \mathfrak{z}_i + \frac{\gamma_f}{\Gamma} \mathfrak{z}_f , \tag{5.7}$$

it follows that

$$\mathfrak{z}_i = -\frac{D_i(\mathfrak{z}_0)}{D_0'(\mathfrak{z}_0)}, \quad \mathfrak{z}_f = -\frac{D_f(\mathfrak{z}_0)}{D_0'(\mathfrak{z}_0)} . \tag{5.8}$$

Noting that $D_0' = \Omega + 1 - i\alpha + 2\mathfrak{z}$ and hence for the case (5.4), we get $D_0'(\mathfrak{z}_0) = [2 + (\alpha/q_i)][1 + (iq_i)]$. The correction terms $\mathfrak{z}_i$ and $\mathfrak{z}_f$ are then given by

$$\mathfrak{z}_i = -\left[1 + \frac{\alpha}{q_i} \right] \Big/ \left[2 + \frac{\alpha}{q_i} \right] = \mathfrak{z}_f . \tag{5.9}$$

The correction term $\mathfrak{z}_f$ agrees with the previously reported value[8] though in that work the virtual recombination was ignored. The above analysis shows that for $\Omega = 1 + (\alpha/q_i)$ the correction term (valid for small γ's) is independent of the virtual recombination effects.

The detailed structure of the zeros of $D(z)$ for several values of γ's, q's, Ω, and α is shown in Figs. 4 and 5. These figures show the behavior of $\epsilon_\pm$, which are related to $z_\pm$ by

$$z_\pm = -\frac{i\Gamma}{2}(\epsilon_\pm - \alpha) . \tag{5.10}$$

The $\epsilon_\pm$ are the solutions of the quadratic

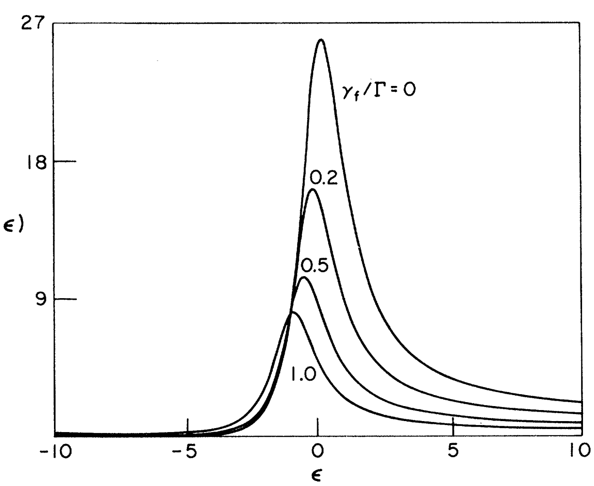

FIG. 3. Modifed Fano profiles $S_F(\epsilon)$ for $q_i = q_i' = 5$, $q_f = q_f' = 1$, $\gamma_i = 0$, and $\gamma_f/\Gamma = 0$, 0.2, 0.5, and 1. Now the Fano minimum at $\epsilon = -q_i$ is lost, but the curves remain asymmetric.

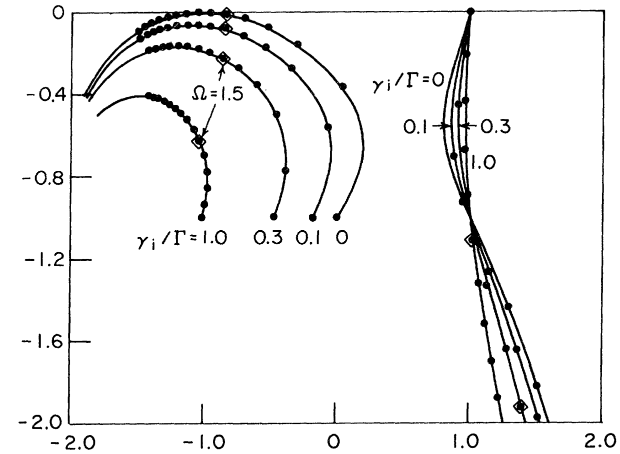

FIG. 4. Motion of $\epsilon_\pm$ when the laser intensity Ω is varied from 0 to 10 for $q_i=1$, $\alpha=1$, $\gamma_f=0$, and $\gamma_i/\Gamma=0$, 0.1, 0.3, and 1. The dots indicate increments of 0.3 in Ω; the $\Omega=1.5$ dots are indicated specially because we examine the spectra for this Ω in a later section. Identical curves would be obtained if we took $q_f=q_i$, $\gamma_i=0$, and varied γ_f.

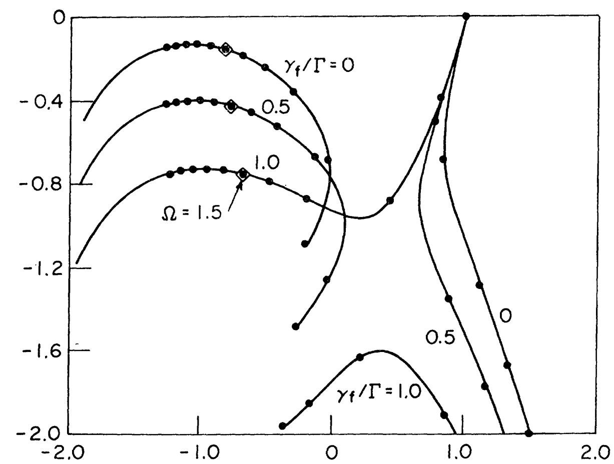

FIG. 5. Motion of $\epsilon_\pm$ with Ω (0 to 10) for $q_i=1$, $q_f=10$, $\alpha=1$, $\gamma_i/\Gamma=0.1$, and $\gamma_f/\Gamma=0$, 0.5, and 1. Again the dots indicate increments of 0.3, and the $\Omega=1.5$ dots are indicated. Note that the real parts of the poles move toward each other for small Ω and then repel for larger Ω. The $\gamma_f/\Gamma=1$ curve shows that at a particular laser intensity the real parts are equal, i.e., the two decaying states have the same energy.

$$\epsilon^2-[\alpha+\Delta_a-i(\eta+\Omega/\psi)]\epsilon+\alpha\Delta_a-\frac{\Omega}{\psi}q_i^2\left[1+\frac{\gamma_f}{\Gamma}\left[\frac{1}{q_i}-\frac{1}{q_f}\right]^2\right]-i\left[\alpha\eta-2\frac{\Omega}{\psi}q_i\right]=0\ , \tag{5.11}$$

where ψ, η, and Δ_a are defined in (4.9). The real parts of $\epsilon_\pm$ indicate the energies of the discrete states (relative to E_a) and the imaginary parts half the decay rates, both in units of $\Gamma/2$. They are related to $\det(\mathbb{1}+m)$ by

$$z\det(\mathbb{1}+\underline{m})=\frac{\Gamma/2}{1-i\epsilon}[-\psi(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)]\ , \tag{5.12}$$

where z and ϵ are related by

$$z=-\frac{i\Gamma}{2}(\epsilon-\alpha)\ . \tag{5.13}$$

In the limit of weak laser field ($\Omega\to0$), the states lie at $\alpha-i0$, where the laser is tuned, and at $\Delta_a-i\eta$. The effective shift Δ_a of the autoionizing state is a result of the "virtual recombination," and is analogous to the single decay channel shift discussed elsewhere.[9,29]

Figure 4 shows the motion of $\epsilon_\pm$ with increasing laser intensity for $\gamma_f=0$, $q_i=1$, $\alpha=1$, and $\gamma_i/\Gamma=0$, 0.1, 0.3, and 1. The "confluence" occurs when one of the poles for $\gamma_i=0$ touches the real axis at $\Omega=2$. The curves show how increasing γ_i shifts and broadens the states. Since γ_i/Γ is comparable to q_i, virtual recombination is important both in shifting the weak-field state and in keeping its width close to unity ($\eta\approx1$). Identical curves would be obtained if we took $q_f=q_i=1$, $\gamma_i=0$ and varied γ_f. Figure 5 shows the pole motion for $q_f=10$, $q_i=1$, $\alpha=1$, $\gamma_i=0.1$, and $\gamma_f=0$, 0.1, and 1. Since $\gamma_i\ll q_i$ and $\gamma_f\ll q_f$ in all the curves, ψ remains close to unity; consequently, there is only a small weak field shift of the autoionizing state, and increasing γ_f rapidly increases the width of the state.

VI. SPECTRA OF PHOTOELECTRONS IN LASER-INDUCED AUTOIONIZATION

Having discussed the dressed states of the system in Sec. V, we are now in a position to analyze the spectra of photoelectrons produced in strong laser-field–induced autoionization. The energy distribution of the photoelectrons is related to the density operator of the atomic system by

$$P_E(t)=\rho_{EE}(t)\ . \tag{6.1}$$

We will assume that the atom is in the state $|i\rangle$ at $t=0$. In such a case it follows from (3.22) that

$$\hat{P}_E(z)=\left[1+\frac{\nu}{z}\right]\hat{\sigma}_{EE}(z)\Big/(1+\nu\,\mathrm{Tr}\hat{\sigma}-\hat{g}_\sigma)\ . \tag{6.2}$$

In what follows we consider the spectra in the limit $t\to\infty$. Equation (6.2) then gives

$$P_E(\infty)\equiv P_E=\lim_{z\to0}\frac{(z+\nu)z\hat{\sigma}_{EE}(z)}{z+\nu z\,\mathrm{Tr}\hat{\sigma}(z)-z\hat{g}_\sigma(z)}\ , \tag{6.3}$$

which for nonzero ν is

$$P_E=\nu\sigma_{EE}(\infty)/[\nu\,\mathrm{Tr}\sigma(\infty)-g_\sigma(\infty)]\ . \tag{6.4}$$

We first note that if $\gamma_i=0$ but $\gamma_f\neq0$, then $g_\sigma(\infty)=0$ [Eq. (3.19)] and

$$P_E=\sigma_{EE}(\infty)/\mathrm{Tr}\sigma(\infty)\ , \tag{6.5}$$

i.e., the steady-state spectra are independent of ν. On the other hand, if $\gamma_f=0$, $\nu=0$ but $\gamma_i\neq 0$, then (3.24) shows that

$$P_E=\sigma_{EE}(\infty)/\mathrm{Tr}\sigma(\infty)\ . \tag{6.6}$$

Note that (6.5) and (6.6) are identical in form and will give the same P_E if $q_f=q_i$ and we interchange $\gamma_f\leftrightarrow\gamma_i$. This leads to the interesting conclusion that the photoelectron spectra are the same (for either $\gamma_f=0$ or $\gamma_i=0$, $\nu\neq 0$) no matter how the system is recycled.

In further discussion we drop the incoherent relaxation rate ν. Now spontaneous decay to $|f\rangle$ represents a sink and the $\gamma_i,q_i\leftrightarrow\gamma_f,q_f$ symmetry is lost. The steady-state spectra then become

$$P_E=\sigma_{EE}(\infty)/(1-\lim_{z\to 0}\hat{g}_\sigma)\ . \tag{6.7}$$

From the behavior of $\psi_E(t)$ in the limit $t\to\infty$, we have

$$\sigma_{EE}(\infty)=|\psi_E(\infty)|^2\ ,$$
$$\psi_E(\infty)=\lim_{z\to -i\Delta_E}(z+i\Delta_E)\hat{\psi}_E(z)\ . \tag{6.8}$$

The quantity $\hat{g}_\sigma$ [given by (3.20)] is expressible in terms of the solution of the integral equation (3.29) as follows:

$$g_\sigma(t)=\gamma_i\int dE_1 B^*_{E_1a}\psi^*_{E_1}(t)\int dE\, B_{Ea}\psi_E(t)$$
$$=2|\chi_2(t)|^2\ , \tag{6.9}$$

where we have used the definitions (3.30) and (3.31). Equation (3.35) gives $\hat{\chi}_2$,

$$\hat{\chi}_2=i[(\mathbb{1}+\underline{m})^{-1}]_{21} \tag{6.10}$$

which after some analysis can be written in terms of ϵ as

$$\hat{\chi}_2=\frac{(2\Omega\gamma_i)^{1/2}}{\Gamma}\frac{1}{\psi(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)}\times\left\{2+\frac{\epsilon}{q_i}+iq_i\left[1+\frac{\gamma_f}{\Gamma}\left[\frac{1}{q_i}-\frac{1}{q_f}\right]^2\right]\right\}\ . \tag{6.11}$$

We then can write

$$\chi_2(t)=\sum_{\alpha=1}^{2}\chi_2^{(\alpha)}e^{z_\alpha t}\ , \tag{6.12}$$

where

$$\chi_2^{(\alpha)}=(-1)^{\alpha+1}\left[\frac{\Omega\gamma_i}{2}\right]^{1/2}\times\frac{q_i\left[1+\frac{\gamma_f}{\Gamma}\left[\frac{1}{q_i}-\frac{1}{q_f}\right]^2\right]-i\left[2+\frac{\epsilon_\alpha}{q_i}\right]}{\psi(\epsilon_+-\epsilon_-)}\ ,$$
$$\epsilon_{1,2}=\epsilon_\pm\ . \tag{6.13}$$

On combining (6.7) and (6.9), we get

$$P_E=|\psi_E(\infty)|^2/[1-\hat{g}_\sigma(0)]\ , \tag{6.14}$$

$$\hat{g}_\sigma(0)=2\int_0^\infty dt\,|\chi_2(t)|^2$$
$$=2\sum_{\alpha,\beta}\frac{\chi_2^{(\alpha)}\chi_2^{(\beta)*}}{-z_\alpha-z^*_\beta}\ . \tag{6.15}$$

Note that the spectral properties of the electrons are all determined from ψ_E. The factor $[1-\hat{g}_\sigma(0)]^{-1}$ is a simple scaling factor (equal to one for $\gamma_i=0$) which represents the increased yield of photoelectrons in the recycling case: If the atom decays to $|i\rangle$ by photon emission it can be recycled, but no recycling is possible after a decay to $|f\rangle$ for $\nu=0$.

From (3.35) and (6.8) we find

$$\psi_E(\infty)=-i\lim_{z\to -i\Delta_E}\left[\frac{v_{Ei}}{z}[(\mathbb{1}+\underline{m})^{-1}]_{11}+\left[\frac{\gamma_i}{2}\right]^{1/2}B^*_{Ea}[(\mathbb{1}+\underline{m})^{-1}]_{12}+\left[\frac{\gamma_f}{2}\right]^{1/2}C^*_{Ea}[(\mathbb{1}+\underline{m})^{-1}]_{13}\right]\ , \tag{6.16}$$

where v_{Ei} [Eq. (2.8)] is proportional to B^*_{Ea} with B^*_{Ea} given by (2.9). It is interesting to note that if the initial and final states have the same q values, $q_i=q_f$, then $\psi_E(\infty)$ and hence P_E continues to show the Fano minimum at $\epsilon=-q_i=-q_f$ since $B_{Ea}=C_{Ea}=0$ at this point, even though we have taken into account the strong laser field and spontaneous emission effects. The situation obviously is different if $q_i\neq q_f$. Equation (6.16) can be simplified by writing explicit expressions for $(\mathbb{1}+\underline{m})^{-1}$, B_{Ea}, and C_{Ea}. In simplest form we find

$$\psi_E(\infty)=\left[\frac{2\Omega}{\pi\Gamma}\right]^{1/2}\frac{\epsilon+q_i+i(\gamma_f/\Gamma)(1-q_i/q_f)}{\psi(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)}\ . \tag{6.17}$$

The photoelectron spectra will have a doublet structure which is asymmetric as a function of ϵ. The width of the peaks can be obtained from our earlier figures. It is interesting to note that γ_i affects $\psi_E(\infty)$ only through $\epsilon_\pm$ and ψ. Equation (6.17) is identical in form to the expression presented elsewhere[9] for the case $\gamma_i=0$.

Figure 6 shows the photoelectron spectra in the long-time limit for $q_i=q_f=1$, $\alpha=1$, $\Omega=1.5$, $\gamma_i=0$, and $\gamma_f/\Gamma=0.1$, 0.3, and 1. The poles $\epsilon_\pm$ for this curve are shown in Fig. 4. The curves exhibit the Fano minimum since $q_i=q_f$. Note how increasing γ_f destroys the sharp feature of the spectrum. Since decay to $|f\rangle$ represents a sink, the total number of photoelectrons ejected (as determined by the areas under the curves) decreases with increasing γ_f. Figure 7 gives the spectra for the same parameters except that now $\gamma_f=0$ and $\gamma_i\neq 0$. The poles are the same as for Fig. 6, and the individual curves differ from those in Fig.6 by only a (γ-dependent) scaling factor. Now spontaneous decay does not act as a probability sink, but instead allows population to be transferred between the dressed states. The area under the curves

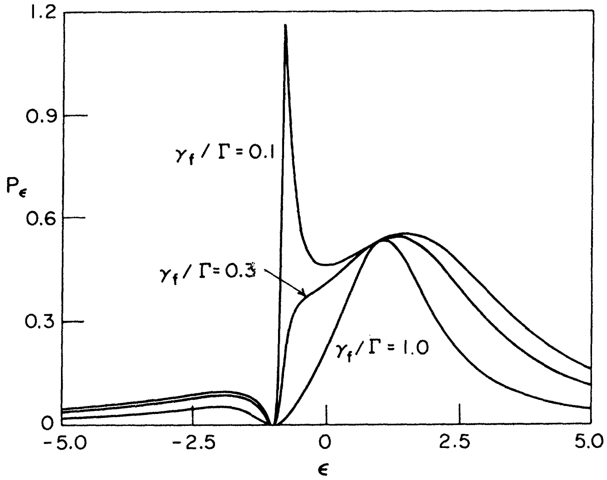

FIG. 6. Photoelectron spectra in limit $t\rightarrow\infty$ and in units of $1/\pi\Gamma$ for $q_i=q_f=1$, $\alpha=1$, $\Omega=1.5$, $\gamma_i=0$, and $\gamma_f/\Gamma=0.1$, 0.3, and 1.

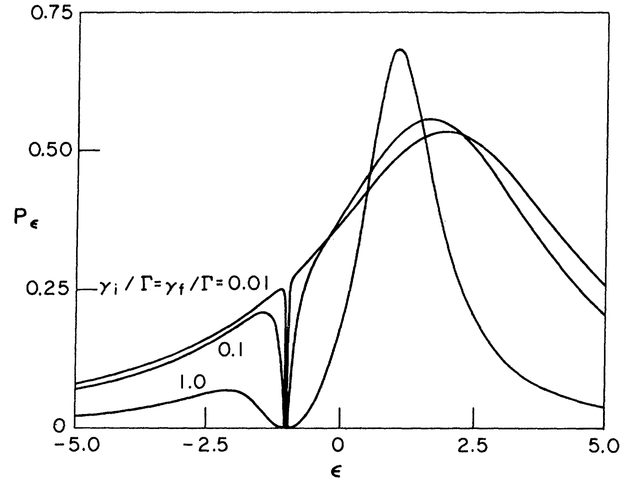

FIG. 8. Photoelectron spectra for $q_i=q_f=1$, $\alpha=1$, $\Omega=1+(\alpha/q_i)=2$ (directly at "confluence"), and $\gamma_i/\Gamma=\gamma_f/\Gamma=0.01$, 0.1, and 1.

remains constant since all the dressed-state population is eventually ejected as photoelectrons.

Figure 8 shows the spectrum when the laser is tuned to confluence ($\Omega=2$) for $\alpha=q_1=q_2=1$ for the cases $\gamma_i/\Gamma=\gamma_f/\Gamma=0.01$, 0.1, and 1. Figures 9 and 10 give the spectra for $q_i=1$, $q_f=10$, $\alpha=1$, $\Omega=1.5$, and various γ. In Fig. 9, γ_i/Γ is constant at 0.1 and $\gamma_f/\Gamma=0.02$, 0.1, and 0.5. Increasing γ_f washes out the Fano minimum, destroys the sharp feature, and decreases the area under the curve. In Fig. 10, γ_f/Γ is constant at 0.1 and γ_i is varied. The curves in Figs. 9 and 10 are not related by a simple scaling factor.

VII. TIME DEPENDENCE OF THE GROUND-STATE POPULATION

The ground-state population[30] $P_{ii}(t)$ can be obtained for the case $\gamma_f=0$ by transforming

$$\hat{p}_{ii}=\frac{\hat{\sigma}_{ii}(z)}{1-\hat{g}_\sigma(z)}\,, \qquad \hat{g}_\sigma(z)=2\int_0^\infty e^{-zt}\,|\chi_2(t)|^2dt\,. \tag{7.1}$$

Here σ_{ii} can be expressed in terms of χ,

$$\hat{\sigma}_{ii}=\int_0^\infty e^{-zt}dt\,|\psi_i(t)|^2\,, \qquad \hat{\psi}_i=\frac{1}{z}-\frac{i}{z}\chi_1\,. \tag{7.2}$$

On using (3.34), we find

$$\hat{\psi}_i=\frac{1}{z}[(\underline{1}+\underline{m})^{-1}]_{11} \tag{7.3}$$

which simplifies to

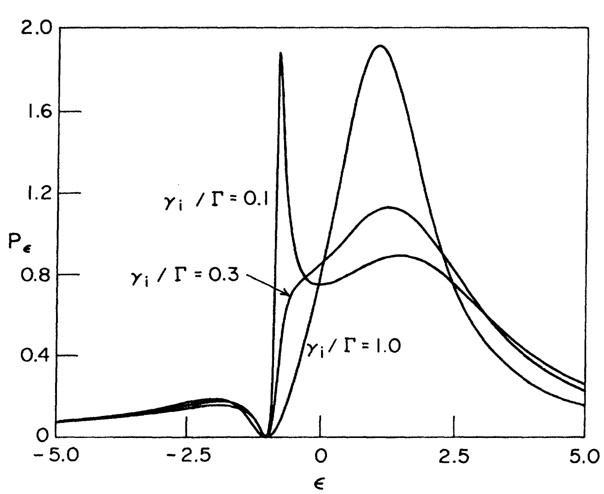

FIG. 7. Photoelectron spectra for the same conditions as Fig. 5 except now $\gamma_f=0$ and γ_i/Γ is varied.

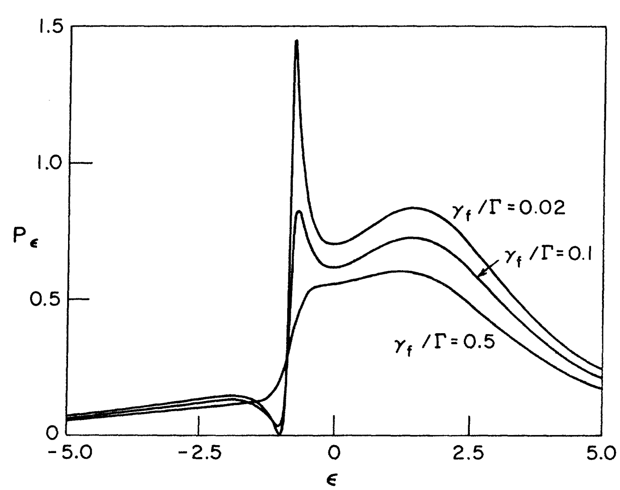

FIG. 9. Photoelectron spectra for $q_i=1$, $q_2=10$, $\alpha=1$, $\Omega=1.5$, $\gamma_i/\Gamma=0.1$, and $\gamma_f/\Gamma=0.02$, 0.1, and 0.5.

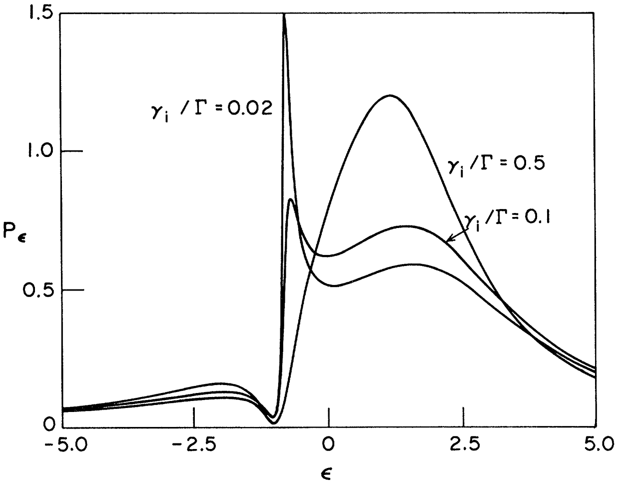

FIG. 10. Photoelectron spectra for the same conditions as Fig. 8 except now $\gamma_f/\Gamma=0.1$ and $\gamma_i/\Gamma=0.02$, 0.1, and 0.5.

$$\hat{\psi}_i=\frac{2i}{\Gamma}\frac{\epsilon-\Delta_a+i\eta}{(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)} . \tag{7.4}$$

(This is the general form for $\hat{\psi}_i$, and applies for all γ_i and γ_f.) The behavior of $P_{ii}(t)$ that results on using (7.1) is shown in Figs. 11 and 12. Figure 11 has $\alpha=q_i=1$, $\Omega=1.5$, and $\gamma_i/\Gamma=0$ and 0.1. One of the dressed states decays much more rapidly than the other, and there is little evidence of Rabi oscillations. Figure 12 has a larger q_i value, so that the discrete state–discrete state coupling is strong relative to the discrete state–initial continuum coupling. The upper curve is directly at confluence, and population is trapped in the atom[14–16] ($\alpha=0$, $\Omega=1$, $\gamma_i=0$) while the other curves have $\gamma_i=0.1$ and 1. Now the $|i\rangle$-$|a\rangle$ Rabi oscillations are clearly seen. In all curves but the one directly at confluence and $\gamma_i=0$, $P_{ii}(t)\to 0$ as $t\to\infty$.

ACKNOWLEDGMENTS

One of us (G.S.A.) is grateful to the National Science Foundation for an international travel grant that made this collaboration possible. This work was supported by National Science Foundation Grant No. PHY-82-00805.

APPENDIX: CONNECTION BETWEEN PHENOMENOLOGICAL DECAY EQUATIONS AND MASTER EQUATION RESULTS FOR $\gamma_i=0$, $\gamma_f\neq 0$, $q_f'\to\infty$

In many problems involving the interaction of an atomic system with external fields, one uses a wave-function description and one also puts in phenomenologically the rate constants corresponding to the radiative decays. This even has been done for the case of four-wave mixing involving autoionizing states by Crance and Armstrong.[4] One is obviously faced with the questions—what is the relationship between the master equation framework and the wave-function description, and what are the conditions under which such a phenomenological description may be valid? In this appendix, we examine this question.[31] Clearly one must have a situation for which $\gamma_i=0$, $\nu=0$. In such a case [result (3.26)] we indeed have a wave function description for

$$\rho_{\alpha\beta}(t)=\psi_\alpha(t)\psi_\beta^*(t) . \tag{A1}$$

However, the equations (3.9) and (3.10) for ψ do not have a very simple structure as far as the radiative decay terms are concerned. The complication arises due to the virtual recombination. Let us therefore make the further simplification and ignore the recombination, i.e., take $C_{Ea}\to b_{Ea}$. Then (3.9) becomes

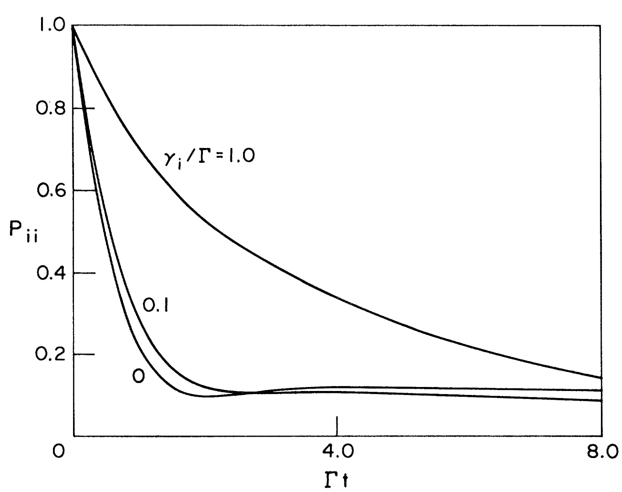

FIG. 11. Time development of the population of $|i\rangle$ for $q_i=1$, $\alpha=1$, $\Omega=1.5$, $\gamma_f=0$, and $\gamma_i/\Gamma=0$, 0.1, and 1.

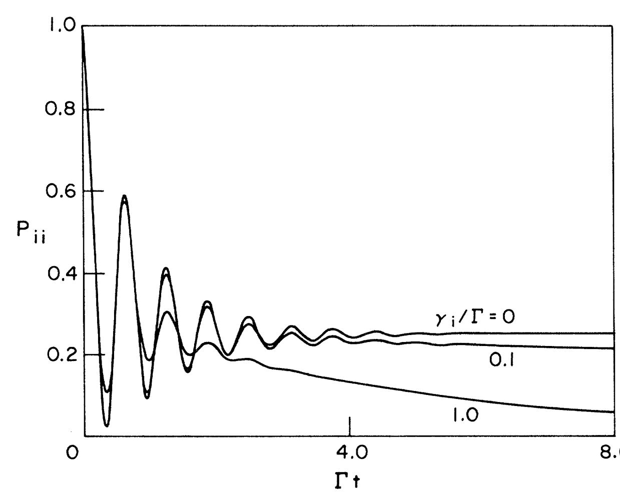

FIG. 12. Time development of the population of $|i\rangle$ for $q_i=10$, $\Omega=1$, $\gamma_f=0$, and $\gamma_i/\Gamma=0$, 0.1, and 1.

$$\dot{\psi}_{E_1} = -i\Delta_{E_1}\psi_{E_1} - iv_{E_1 i}\psi_i - \frac{\gamma_f}{2}\int dE\, b^*_{E_1 a} b_{Ea}\psi_E \ . \quad \text{(A2)}$$

If we now use (2.2) and the orthogonality of $|a\rangle$ and $|E'\rangle$, then (A2) leads to

$$\dot{\psi}_a = -\frac{\gamma_f}{2}\psi_a + \cdots \ , \qquad \dot{\widetilde{\psi}}_E = \cdots \ , \quad \text{(A3)}$$

where $\widetilde{\psi}_E = (E \mid \psi\rangle$ and the ellipses denote the contribution coming from the coherent components. Equations (A3) are just the equations which one would write on phenomenological grounds. We have thus established the connection between the master equation approach and phenomenological approach in a very special case—when the decay to a third level (from which the system does not return) is considered and when the virtual recombination effects are ignored. The description (A3) has the feature that the problem with the radiative decay is solved by changing E_a to $E_a - (i\gamma_f/2)$, i.e.,

$$\widetilde{\psi}_E \mid_{\gamma_f \neq 0} = \widetilde{\psi}_E \mid_{E_a \to E_a - i\gamma_f/2} \ . \quad \text{(A4)}$$

The final photoelectron spectrum is given by $|\psi_E|^2$ and so one still has to make a transformation from $\widetilde{\psi}_E$ to ψ_E using (2.2). However, it is known that the "steady-state behavior" can be directly obtained by using $\widetilde{\psi}_E$ since

$$\lim_{t\to\infty} |\psi_E|^2 = \lim_{t\to\infty} |\widetilde{\psi}_E|^2 \ .$$

Thus the steady-state spectral amplitudes obey the simple substitution rule $E_a \to E_a - (i\gamma_f/2)$. The explicit expression for the steady-state photoelectron spectra of Ref. 8 does indeed show that such a substitution rule holds since the conditions under which these results were derived are precisely the conditions $\gamma_i = 0$, $\nu = 0$, $q_f' = \infty$.

---

*Present address: Department of Physics, Calvin College, Grand Rapids, MI 49506.

[1]U. Fano, Phys. Rev. 124, 1866 (1961).

[2]J. A. Armstrong and J. J. Wynne, in *Nonlinear Spectroscopy*, edited by N. Bloembergen (North-Holland, Amsterdam, 1977), p. 152.

[3]L. Armstrong, Jr. and B. L. Beers, Phys. Rev. Lett. 34, 1290 (1975).

[4]M. Crance and L. Armstrong, Jr., J. Phys. B 15, 4637 (1982); see also, Yu. I. Geller and A. K. Popov, Pis'ma Zh. Tekh. Fiz. 7, 719 (1981) [Sov. Tech. Phys. Lett. 7, 307 (1981)].

[5]G. S. Agarwal and P. Anantha Lakshmi, Phys. Rev. A 28, 3430 (1983).

[6]G. Alber and P. Zoller, Phys. Rev. A 27, 1373 (1983).

[7]L. D. Van Woerkom, W. E. Cooke, and R. R. Freeman, Bull. Am. Phys. Soc. 28, 775 (1983).

[8]G. S. Agarwal, S. L. Haan, K. Burnett, and J. Cooper, Phys. Rev. Lett. 48, 1164 (1982); Phys. Rev. A 26, 2277 (1982).

[9]S. L. Haan and G. S. Agarwal, in *Spectral Line Shapes Volume 2,* Proceedings, Sixth International Conference, Boulder, Colorado, July 12—16, 1982, edited by K. Burnett (de Gruyter, Berlin, 1983), p. 1013.

[10]G. S. Agarwal, S. L. Haan, and J. Cooper, Phys. Rev. A 28, 1154 (1983).

[11]M. Lewenstein, J. W. Haus, and K. Rzążewski, Phys. Rev. Lett. 50, 417 (1983), and unpublished.

[12]L. Armstrong, Jr., C. E. Theodosiou, and M. J. Wall, Phys. Rev. A 18, 2538 (1978).

[13]J. Bokor, R. R. Freeman, and W. E. Cooke, Phys. Rev. Lett. 48, 1242 (1982).

[14]P. Lambropoulos and P. Zoller, Phys. Rev. A 24, 379 (1981); see also P. Lambropoulos, Appl. Opt. 19, 3926 (1980).

[15]K. Rzążewski and J. H. Eberly, Phys. Rev. Lett. 47, 408 (1981).

[16]P. E. Coleman, P. L. Knight, and K. Burnett, Opt. Commun. 42, 171 (1982); P. E. Coleman and P. L. Knight, J. Phys. B 15, L235 (1982); 15, 1957(E) (1982).

[17]J. H. Eberly, K. Rzążewski, and D. Agassi, Phys. Rev. Lett. 49, 693 (1982); K. Rzążewski and J. H. Eberly, Phys. Rev. A 27, 2026 (1983).

[18]G. S. Agarwal and D. Agassi, Phys. Rev. A 27, 2254 (1983).

[19]Y. S. Kim and P. Lambropoulos, Phys. Rev. Lett. 49, 1698 (1982).

[20]W. E. Cooke, T. F. Gallagher, S. A. Edelstein, and R. M. Hill, Phys. Rev. Lett. 40, 178 (1978).

[21]A. I. Andryushin, M. V. Fedorov, and A. E. Kazakov, J. Phys. B 15, 2851 (1982); A. I. Andryushin, A. E. Kazakov, and M. V. Fedorov, Zh. Eksp. Teor. Fiz. 82, 91 (1982) [Sov. Phys.—JETP 55, 53 (1982)].

[22]Yu. I. Geller and A. K. Popov, Zh. Eksp. Teor. Fiz. 78, 506 (1980) [Sov. Phys.—JETP 51, 255 (1980)].

[23]M. Crance and L. Armstrong, Jr., J. Phys. B 15, 3199 (1982).

[24]B. L. Beers and L. Armstrong, Jr., Phys. Rev. A 12, 2447 (1975).

[25]L. Armstrong, Jr., B. L. Beers, and S. Feneuille, Phys. Rev. A 12, 1903 (1975).

[26]D. Agassi, in *Proceedings of the NATO Advanced Study Institute of Quantum Electrodynamics and Quantum Optics, Boulder, 1983,* edited by A. O. Barut (Plenum, New York, in press); D. Agassi and J. H. Eberly (unpublished).

[27]G. S. Agarwal, *Quantum Optics,* Vol. 70 of *Springer Tracts in Modern Physics,* edited by G. Höhler (Springer, Berlin, 1974).

[28]This virtual recombination is discussed in more detail in Ref. 29. The "real recombination" problem is studied in P. C. W. Davies and M. J. Seaton, J. Phys. B 2, 757 (1969).

[29]S. L. Haan and J. Cooper, Phys. Rev. A 28, 3349 (1983).

[30]The time development of the ground-state population has recently been examined by D. Agassi, K. Rzążewski, and J. H. Eberly, Phys. Rev. A 28, 3648 (1983).

[31]The authors are grateful to P. Knight for pointing out that the photoelectron spectra of Ref. 8 can be obtained by using the substitution rule $E_a \to E_a - i\gamma_f/2$.