Absorption spectroscopy of strongly perturbed bound-continuum transitions

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Absorption properties of a system consisting of two autoionizing states coupled strongly either by internal interactions or by external fields are investigated. The autoionizing states may themselves correspond to those induced by laser fields as in resonant multiphoton ionization. The analysis is based on the density matrices and incoherent processes like spontaneous emission and radiative recombination are included. Optical susceptibilities exist in a number of special cases for autoionizing transitions. Exact results for such optical susceptibilities are obtained. Explicit numerical results for susceptibilities, corresponding traditionally to strong-field inverse Raman effect, are given with a view to recent work on *dc*-field-coupled autoionizing states and on laser-induced autoionization. The behavior of both absorptive and dispersive parts is discussed for a range of parameters.

I. INTRODUCTION

The physics of bound-continuum transitions has been studied in a number of ways. In particular, optical absorption studies[1,2] have played a key role in the understanding of the autoionizing transitions. The susceptibility $\chi_{\alpha\beta}(\omega)$, as is well known, yields information on both line positions and linewidths. According to the linear response theory, χ can be written formally in terms of the eigenfunctions $|\psi_i\rangle$ and the eigenvalues E_i of the system's Hamiltonian

$$\chi_{\alpha\beta}(\omega)=\sum_{\substack{i,g\\(i\neq g)}}\frac{\langle\psi_g|\mathbf{d}_\alpha^*|\psi_i\rangle\langle\psi_i|\mathbf{d}_\beta|\psi_g\rangle}{(E_i-E_g-\hbar\omega)}\,, \tag{1.1}$$

where $|\psi_g\rangle$ ($|\psi_i\rangle$) is the ground state (initial state) of the system. Counter rotating terms have been ignored from (1.1). The summation in (1.1) is over all the discrete and continuum states of the system. The susceptibility can be evaluated from the knowledge of exact eigenfunctions and eigenvalues. Shore[3] has developed a scattering matrix approach for the calculation of $\chi_{\alpha\beta}$. For a single autoionizing state $|a\rangle$ lying in a structureless continuum $|E)$, evaluation of (1.1) leads to

$$\chi(\omega)\approx|(E|d|\psi_g\rangle|^2q^2\pi i\left[\frac{\left[1-\frac{i}{q}\right]^2}{1-ix}+\frac{1}{q^2}\right], \tag{1.2}$$

$$x=\frac{2}{\Gamma}(E_g-E_a+\hbar\omega)\,. \tag{1.3}$$

Here Γ is the rate of autoionization and q is Fano's asymmetry parameter. For simplicity we have ignored the vectorial aspects of the dipole matrix elements. From (1.2) one has[2]

$$\mathrm{Im}\chi(\omega)\propto\frac{(q+x)^2}{1+x^2}\,. \tag{1.4}$$

Optical absorption profiles which are proportional to $\mathrm{Im}\chi$ coincide with the Fano profiles[1] which give the photoelectron spectrum. This is expected since in the absence of any other relaxation effects, the total rate of absorption of energy from the field must be equal to the rate of production of photoelectrons of energy equal to the incident photon energy. The real part of χ gives the dispersive properties[3] of the bound-continuum transitions. Results like (1.2) have been extensively used in the study of autoionizing transitions. However, in view of the activity over the last decade, generalizations of (1.2) are warranted in several directions as detailed below. Some generalizations, such as to the case of several outgoing channels[4] in the vicinity of an autoionizing resonance, exist. Spontaneous emission opens up additional channels. Line profiles are also complicated by the fact that electrons in the vicinity of the ion can recombine to yield a neutral atom and a photon. Furthermore, the autoionizing state can decay via spontaneous emission. It was seen earlier[5,6] that the radiative recombination process depends on, among other things, the asymmetry parameter q. Smaller values of q lead to considerable radiative recombination. It is not *a priori* clear if the radiative decay can be accounted for by replacing the energy E_a by the complex energy. Calculations show that the radiative recombination makes the situation quite complex as we now have a bound state interacting with two continua (electron and photon) which themselves are also interacting. It may be borne in mind that now $\mathrm{Im}\chi$ and the photoelectron spectrum will be different as part of the absorbed energy is used in creating the spontaneously emitted photon.

Recently a number of experiments[7–10] have reported autoionization when some of the autoionizing states are strongly mixed either by a dc electric field or by the spin-orbit interaction. The important outcome of these experiments is essentially the inhibition of the autoionization, i.e., pronounced narrowing of the autoionization line shapes under certain conditions on the field strengths, spin-orbit interaction, etc. A theoretical study of these results[11] will require the calculation of the absorption profiles for a system of states which are strongly coupled and which decay by autoionization.

The susceptibility for these situations can be calculated from the knowledge of the eigenfunctions and eigenvalues of the system's Hamiltonian in presence of a dc electric field. We will present simplified models where such susceptibilities can be calculated in closed form.

Another class of physical phenomena concerns multiphoton ionization in strong fields. Here an autoionizinglike structure can be induced by a resonant laser[12–15] field. Such laser-induced structures can considerably modify the efficiency of a nonlinear optical process such as harmonic generation.[16] The influence of such structures can be investigated in terms of the susceptibilities. For example, experimental results[17] on the rotation of plane of polarization of light passing through cesium beam can be understood either in terms of a third-order susceptibility or equivalently a first-order susceptibility[14,17] of an autoionizinglike structure. A study of resonant two-photon ionization in strong fields using the autoionizationlike formulation will require the knowledge of intensity-dependent χ. The spontaneous emission could be quite important for laser-induced continuum structures.

One can also consider optical mixing of an autoionizing state and a bound state by a strong field of frequency ω. The strength of such a mixing can be varied by both amplitude and frequency of the external field. Such a mixing has been the subject of many investigations.[18–22] Here we discuss how such mixing of states can be probed through an absorption experiment. The probing is to be done by using a transition starting from a bound state which is different from the one which is strongly mixed by the external field. For these problems optical susceptibilities[23] will depend on the strength of the mixing field in a nonperturbative manner.

In this paper we calculate intensity-dependent optical susceptibilities for a model system. The model is general enough to describe many of the situations mentioned above. In Sec. II we discuss the model and the dynamical equations describing the model. Calculations are done in the density matrix framework as we account for both radiative decay and saturation effects. Exact solutions for various elements of the density matrix are given. In Sec. III general expressions for the susceptibilities are given. Various limiting cases corresponding to weak fields are discussed in Sec. IV. We also comment on the form of the Raman susceptibility. In Sec. V we give details of the susceptibility relevant for probing the behavior of strongly coupled bound-continuum transitions. Numerical results for a range of parameters are presented. In the traditional language of nonlinear optics, Sec. V essentially calculates the susceptibility for the inverse Raman effect.

II. MODEL FOR INTENSITY-DEPENDENT SUSCEPTIBILITIES

For a system interacting with external fields, the Hamiltonian becomes time dependent and in general it is not possible to think in terms of the eigenvalues and eigenfunctions of a time-dependent Hamiltonian. There are, however, situations in which, by suitable canonical transformations, the strong-field part of the Hamiltonian can be made time independent. Hence we consider the specific model schematically shown in Fig. 1. The model is general enough[11] to handle the very many situations mentioned in the Introduction. The state $|a\rangle$ is the autoionizing state or it can be a laser-induced structure as in multiphoton-ionization problems. We take the state $|f\rangle$ as the state from which the physical phenomena are probed, i.e., for most problems we will assume that the initial population is in state $|f\rangle$. The state $|i\rangle$ is a bound state outside the continuum which is strongly coupled by the laser with frequency ω_1 and amplitude ε_1. The coupling of the state $|i\rangle$ with the structured continuum can be thought of as producing another autoionizing structure in the continuum and thus the same may also be looked at from the point of view of probing the two strongly coupled autoionizing states.

The Hamiltonian for the model system of Fig. 1 can be written as

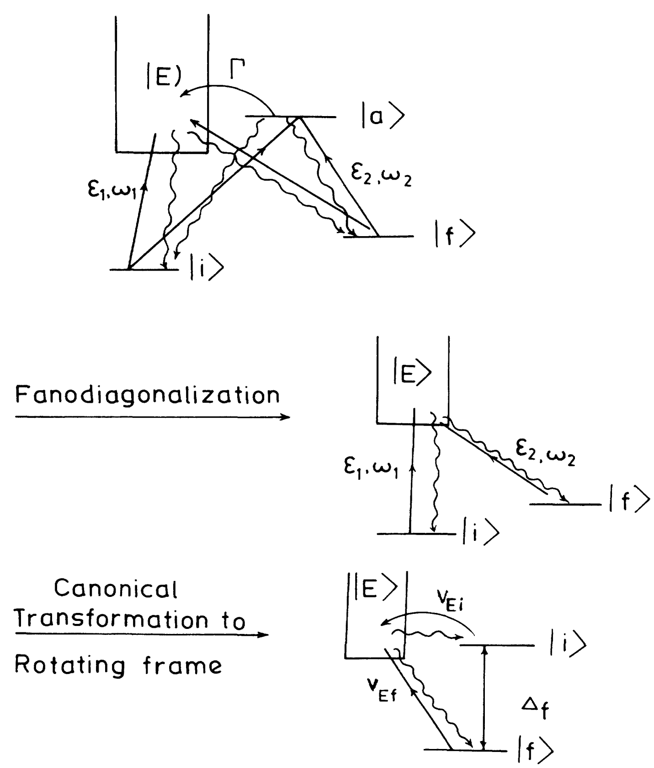

FIG. 1. Schematic diagram of the model system. $|E)$ is the unperturbed flat continuum, $|E\rangle$ is the structured continuum, and ε_i,ω_i gives the amplitude and the frequency of the ith laser. After canonical transformation, the state $|i\rangle$ appears as another autoionizing state. The wavy lines represent the radiation decay of the structured continuum $|E\rangle$.

$$H=E_a|a\rangle\langle a|+\int E|E)(E|dE+E_f|f\rangle\langle f|+E_i|i\rangle\langle i|+\int[V_{Ea}|E)\langle a|+\text{H.c.}]dE$$
$$+\int[\tilde{v}_{Ei}|E)\langle i|e^{-i\omega_1 t}+\text{H.c.}]dE+(\tilde{v}_{ai}|a\rangle\langle i|e^{-i\omega_1 t}+\text{H.c.})+(\tilde{v}_{af}|a\rangle\langle f|e^{-i\omega_2 t}+\text{H.c.})$$
$$+\left[\int\tilde{v}_{Ef}|E)\langle f|e^{-i\omega_2 t}dE+\text{H.c.}\right]+H_R';\ \hbar\equiv1\ . \tag{2.1}$$

The interaction V_{Ea} is the configuration interaction and $\tilde{v}$ is the interaction with the applied fields with amplitudes $\varepsilon_1,\varepsilon_2$ and frequencies ω_1,ω_2. The field ε_2 will be a probing field and need not be laser field. It can, for example, be the synchrotron radiation[2,4] in the vacuum-uv region. The part H_R' represents the radiative decay of the autoionizing states and the radiative recombination. Without H_R' terms some aspects of the above Hamiltonian have been previously considered. For example, Deng and Eberly[24] investigated the effects of the laser strengths on the photoelectron spectra. Lami and Rahman[25] used a similar Hamiltonian in studies of predissociation of molecules. Previous studies[24,25] essentially concentrated on the population distributions, whereas in the present work we examine the macroscopic coherences or the induced polarization in the system. This leads to the possibility of the system giving rise to coherent generation of radiation. Our work also accounts for, in a systematic manner, the radiative effects. It may be added that it is the same dipole matrix element which governs the strength of the laser transition and the radiative decay and thus the radiative decay should be important at least for problems involving excitations by weak fields.

Optical susceptibilities can be evaluated from the knowledge of the off-diagonal density matrix elements like $\rho_{ai},\rho_{af},\rho_{Ef}$, etc. For the model (2.1), the density matrix elements ρ_{af}, etc., can be calculated to all orders in the strength of the fields at ω_1 and ω_2. For obtaining the density matrix elements, we will use the methods used by Agarwal *et al.*[22] (Papers I and II) for the case $\tilde{v}_{af}=0$. We work in a representation in which configuration interaction is diagonal, i.e., we work with Fano states $|E\rangle$. On removing the fast time dependence from the Hamiltonian and treating the radiative-decay terms in the usual manner,[26] the density matrix equation becomes

$$\frac{\partial\rho}{\partial t}=-i[H_{\text{coh}},\rho]-\frac{\gamma_i}{2}(A_i^\dagger A_i\rho-2A_i\rho A_i^\dagger+\rho A_i^\dagger A_i)$$
$$-\frac{\gamma_f}{2}(A_f^\dagger A_f\rho-2A_f\rho A_f^\dagger+\rho A_f^\dagger A_f)\ . \tag{2.2}$$

Here H_{coh} represents the coherent interactions with laser fields,

$$H_{\text{coh}}=\int\Delta_E|E\rangle\langle E|dE+\int(v_{Ei}|E\rangle\langle i|+\text{H.c.})dE$$
$$+\int(v_{Ef}|E\rangle\langle f|+\text{H.c.})dE+\Delta_f|f\rangle\langle f|\ , \tag{2.3}$$

where in terms of Fano parameters q_i, q_f, and the autoionization rate one has

$$\Delta_f=E_f-(\omega_1-\omega_2),\quad E_i=0,\quad \Delta_E=E-\omega_1\ ,$$
$$v_{iE}\approx\tilde{v}_{ia}B_{Ea},\quad v_{fE}\approx\tilde{v}_{fa}C_{Ea}\ ,$$
$$B_{Ea}=b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_i}\right],$$
$$C_{Ea}=b(E,a)\left[1+\frac{2(E-E_a)}{\Gamma q_f}\right]. \tag{2.4}$$

Here $b(E,a)$ represents the overlap between Fano state and the autoionizing level

$$b(E,a)=\langle a|E\rangle\ . \tag{2.5}$$

The incoherent terms in (2.2) correspond to the radiative-decay processes

$$\gamma_j=\tfrac{4}{3}d_{aj}^2\frac{\omega_{aj}^3}{c^3},\quad j=i,f\ . \tag{2.6}$$

The operators A's are essentially dipole moment operators connecting the Fano states and the bound states $|i\rangle$, $|f\rangle$, i.e.,

$$A_i=\int dE\,|i\rangle\langle E|B_{Ea}\ ,$$
$$A_f=\int dE\,|f\rangle\langle E|C_{Ea}\ . \tag{2.7}$$

Thus all the effects of the radiative coupling between the unperturbed continuum $|E\rangle$ and the autoionizing state $|a\rangle$ and the states $|i\rangle$ and $|f\rangle$ enter through the operators A_i and A_f.

The solution of (2.2) will be obtained by following the same method as in paper I. The method developed in I relies on the fact that in the absence of radiative-decay effects, one can work with the wave functions and this simplifies the problem considerably. We thus introduce auxiliary matrix σ defined by

$$\sigma_{\alpha\beta}=\psi_\alpha\psi_\beta^*\ , \tag{2.8}$$

$$\dot{\psi}_i=-i\int dE\,v_{Ei}^*\psi_E\ ,$$
$$\dot{\psi}_f=-i\int dE\,v_{Ef}^*\psi_E-i\Delta_f\psi_f\ ,$$
$$\dot{\psi}_{E_1}=-i\Delta_{E_1}\psi_{E_1}-iv_{E_1i}\psi_i-iv_{E_1f}\psi_f$$
$$-\frac{\gamma_i}{2}\int dE B_{E_1a}^*B_{Ea}\psi_E$$
$$-\frac{\gamma_f}{2}\int dE C_{E_1a}^*C_{Ea}\psi_E\ . \tag{2.9}$$

Using (2.8) and (2.9), the equation for σ can be written as

$$\dot{\sigma}=L\sigma\ , \tag{2.10}$$

where the form of L can be written down from (2.9).

In terms of the operator L and the operators $G^{(i)}$ and

$G^{(f)}$ defined by

$$G^{(i)}=\frac{\gamma_i}{2}\int dE_1\int dE\, B^*_{E_1a}B_{Ea}\,|E_1\rangle\langle E\,|+\text{H.c.}\;,$$
$$G^{(f)}=\frac{\gamma_f}{2}\int dE_1\int dE C^*_{E_1a}C_{Ea}\,|E_1\rangle\langle E\,|+\text{H.c.}\;, \tag{2.11}$$

we can write the basic equation (2.2) as

$$\frac{\partial\rho}{\partial t}=L\rho+|\,i\,\rangle\langle i\,|\,I^{(i)}(t)+|\,f\,\rangle\langle f\,|\,I^{(f)}(t)\;, \tag{2.12}$$

$$I^{(i)}(t)=\text{Tr}\{\rho(t)G^{(i)}\},\;\; I^{(f)}(t)=\text{Tr}\{\rho(t)G^{(f)}\}\;. \tag{2.13}$$

Let $\sigma^{(i)}(\sigma^{(f)})$ be the solution of (2.9) subject to the initial condition $\psi_i=1$, $\psi_f=\psi_E=0$ ($\psi_f=1$, $\psi_i=\psi_E=0$) and let $\rho(0)$ be the incoherent superposition of the states $|\,i\,\rangle$ and $|\,f\,\rangle$,

$$\rho(0)=p^{(i)}\,|\,i\,\rangle\langle i\,|+p^{(f)}\,|\,f\,\rangle\langle f\,|\;. \tag{2.14}$$

The solution of (2.12) can be written as

$$\begin{aligned}\rho(t)=&\sigma^{(i)}(t)p^{(i)}+\sigma^{(f)}(t)p^{(f)}\\&+\int_0^t d\tau[\sigma^{(i)}(t-\tau)I^{(i)}(\tau)\\&\qquad+\sigma^{(f)}(t-\tau)I^{(f)}(\tau)]\;.\end{aligned} \tag{2.15}$$

The unknown terms from the right-hand side of (2.15) can be eliminated as follows. We introduce matrices $T^{\alpha\beta}$ defined by

$$T^{\alpha\beta}(t)=\text{Tr}[\sigma^{(\alpha)}(t)G^{(\beta)}]\;. \tag{2.16}$$

Note that the first (second) superscript α (β) refers to the initial conditions (the state to which spontaneous emission takes place). From (2.15) we obtain equations for the Laplace transforms (denoted by carets) of various functions,

$$\begin{bmatrix}1-\hat{T}^{ii} & -\hat{T}^{fi}\\ -\hat{T}^{if} & 1-\hat{T}^{ff}\end{bmatrix}\begin{bmatrix}\hat{I}^{(i)}\\ \hat{I}^{f}\end{bmatrix}=\begin{bmatrix}\hat{T}^{ii} & \hat{T}^{fi}\\ \hat{T}^{if} & \hat{T}^{ff}\end{bmatrix}\begin{bmatrix}p^{(i)}\\ p^{(f)}\end{bmatrix}. \tag{2.17}$$

Thus the calculation of $\rho_{\alpha\beta}(t)$ involves the following steps: (1) solution of (2.9) and then the construction of the matrices $\sigma^{(i)}(t),\sigma^{(f)}(t)$; (2) calculation of $T^{\alpha\beta}$ [Eq. (2.16)]; (3) calculation of $I^{(i)}$, $I^{(f)}$ [Eq. (2.17)]; (4) calculation of the Laplace transform of ρ obtained from (2.15), i.e., from

$$\hat{\rho}(z)=\hat{\sigma}^{(i)}(p^{(i)}+\hat{I}^{(i)})+\hat{\sigma}^{f}(p^{(f)}+\hat{I}^{(f)})\;. \tag{2.18}$$

For a steady-state result it is sufficient to know the Laplace transforms. For example the probability of finding an electron with energy E is given by

$$\begin{aligned}p(E)=&\lim_{t\to\infty}\rho_{EE}(t)\\=&\lim_{z\to 0}z\hat{\rho}_{EE}(z)\\=&[p^{(i)}+\hat{I}^{(i)}(0)]\sigma^{(i)}_{EE}(\infty)\\&+[p^{(f)}+\hat{I}^{(f)}(0)]\sigma^{(f)}_{EE}(\infty)\;.\end{aligned} \tag{2.19}$$

Here $\sigma_{EE}(\infty)$ is the steady-state value of $\sigma_{EE}(t)$ and $\hat{I}^{(i)}(0)$, $\hat{I}^{(f)}(0)$ are assumed to exist. Off-diagonal matrix elements are similarly obtained. In Appendix A we present the solution of Eqs. (2.9). Solutions for T matrices are given in the Appendix B.

It should be borne in mind that the coherent interaction (2.3) also describes the autoionization produced by two strongly coupled autoionizing states. As discussed in Ref. 11, the matrix elements $\tilde{v}_{ai}$ gives the strong interaction between two autoionizing states. Thus this alternate physical situation should be kept in view while dealing with results like (2.18) and those in subsequent sections.

III. INDUCED POLARIZATION AND INTENSITY-DEPENDENT OPTICAL SUSCEPTIBILITIES

The total induced polarization P for our model system is given by

$$\begin{aligned}\mathbf{P}(t)=&\text{Tr}[\rho(t)\mathbf{d}]\\=&\int \mathbf{d}^*_{Ei}\rho_{Ei}(t)e^{-i\omega_1 t}dE\\&+\int \mathbf{d}^*_{Ef}\rho_{Ef}(t)e^{-i\omega_2 t}dE+\text{c.c.}\end{aligned} \tag{3.1}$$

The matrix elements of ρ are to be obtained from the solution of the dynamical equation (2.2). In Eq. (3.1) the first term gives the response at ω_1 whereas the second term yields the response at ω_2. The steady-state response can be obtained if

$$\lim_{t\to\infty}\int \mathbf{d}_{iE}\rho_{Ei}(t)dE,\;\;\lim_{t\to\infty}\int \mathbf{d}_{fE}\rho_{Ef}(t)dE\;,$$

exist, otherwise one can calculate the transient response. The dipole matrix elements can be expressed in terms of the functions B_{Ea} and C_{Ea} given by (2.4)

$$\begin{aligned}v_{iE}&=-\mathbf{d}_{iE}\cdot\boldsymbol{\varepsilon}_1=\tilde{v}_{ia}B_{Ea}\;,\\ v_{fE}&=-\mathbf{d}_{fE}\cdot\boldsymbol{\varepsilon}_2=\tilde{v}_{fa}C_{Ea}\;.\end{aligned} \tag{3.2}$$

Ignoring vectorial properties of the matrix elements, we can define the transient susceptibilities $\chi^{(i)}(t,\omega_1),\chi^{(f)}(t,\omega_2)$ by

$$\begin{aligned}\chi^{(i)}(t,\omega_1)&=-\int v_{iE}\rho_{Ei}(t)dE/\varepsilon_1^2\;,\\ \chi^{(f)}(t,\omega_2)&=-\int v_{fE}\rho_{Ef}(t)dE/\varepsilon_2^2\;.\end{aligned} \tag{3.3}$$

It should be borne in mind that ω_1 (ω_2) is the field acting on the transition $|\,i\,\rangle\leftrightarrow|\,E\,\rangle(\,|\,f\,\rangle\leftrightarrow|\,E\,\rangle)$. These susceptibilities can be obtained in terms of ψ's of Appendix A. From (2.18) we have

$$\begin{aligned}\int\hat{\rho}_{Ei}(z)v_{iE}dE=&(p^{(i)}+\hat{I}^{(i)})\int\hat{\sigma}^{(i)}_{Ei}(z)v_{iE}dE\\&+(p^{(f)}+\hat{I}^{(f)})\int\hat{\sigma}^{(f)}_{Ei}(z)v_{iE}dE\;,\end{aligned} \tag{3.4}$$

which, on using the relation

$$\begin{aligned}\int\hat{\sigma}_{Ei}(z)v_{iE}dE=&\int_0^\infty e^{-zt}dt\int v_{iE}\sigma_{Ei}(t)dE\\=&\int_0^\infty e^{-zt}dt\int v_{iE}\psi_E(t)\psi_i^*(t)dE\\=&\int_0^\infty e^{-zt}dt\,\Phi_1(t)\psi_i^*(t)\;,\end{aligned} \tag{3.5}$$

simplifies to

$$\int \hat{\rho}_{Ei}(z) v_{iE} dE = (p^{(i)} + \hat{I}^{(i)}) \int_0^\infty e^{-zt} dt\, \Phi_1^{(i)}(t) \psi_i^{*(i)}(t) + (p^{(f)} + \hat{I}^{(f)}) \int_0^\infty e^{-zt} dt\, \Phi_1^{(f)}(t) \psi_i^{*(f)}(t) . \tag{3.6}$$

On substituting (3.6) in (3.3) we get the expression for the transient susceptibility

$$\hat{\chi}^{(i)}(z,\omega_1) = -\int_0^\infty dt \frac{e^{-zt}}{\varepsilon_1^2} \{ [p^{(i)} + \hat{I}^{(i)}(z)] \Phi_1^{(i)}(t) \psi_i^{(i)*}(t) + [p^{(f)} + \hat{I}^{(f)}(z)] \Phi_1^{(f)}(t) \psi_i^{(f)*}(t) \} . \tag{3.7}$$

Similarly one can derive

$$\hat{\chi}^{(f)}(z,\omega_2) = -\int_0^\infty dt \frac{e^{-zt}}{\varepsilon_2^2} \{ [p^{(i)} + \hat{I}^{(i)}(z)] \Phi_2^{(i)}(t) \psi_f^{(i)*}(t) + [p^{(f)} + \hat{I}^{(f)}(z)] \Phi_2^{(f)}(t) \psi_f^{(f)*}(t) \} . \tag{3.8}$$

Transient susceptibilities can be explicitly evaluated by using the solutions given in Appendix A. No approximation on the strength of either the field ε_1 or ε_2 needs to be made.

The steady-state response denoted by $\chi^{(i)}(\omega_1)$, $\chi^{(f)}(\omega_2)$ can be obtained from (3.7) and (3.8) in the usual manner. The existence of the steady-state response depends on the structure of the roots of $\det(1+m)$. Since the time dependence of ψ's and Φ's is governed by the complex zeros of $\det(1+m)$, we will see that nonzero steady-state susceptibilities do exist in a number of cases of interest treated in Secs. IV and V. If the fields ε_1 and ε_2 pumping the two transitions are quite strong, then the system is expected to ionize completely in the long-time limit $t \to \infty$. In such a case the system is unlikely to show any coherence, i.e., $\rho_{Ei}(t) \to 0$ as $t \to \infty$. We will therefore consider the situation when the field acting, say, on the transition $|i\rangle \leftrightarrow |E\rangle$ is of arbitrary magnitude but the field ε_2 is weak. In other words, the behavior of the strongly coupled system of states $|i\rangle$, $|E\rangle$ is probed by a weak field. This is indeed the way in which most experiments are typically done. For example, recent experiments[7–10] probe strongly coupled autoionizing states (coupled either by dc field or by spin-orbit interaction) by another weak field. Thus one essentially needs to know $\chi^{(f)}(\omega_2)$ to all orders in the field ε_1 but to zero order in ε_2. It should be remembered that the field ε_1 may be either an external field or an internal field.

IV. WEAK-FIELD SUSCEPTIBILITIES

In this section we examine the form of the susceptibility in the limit of weak fields. We present results for the linear susceptibility and Raman susceptibility. These susceptibilities enable us to interpret the elements of the matrix m [Eq. (A10)] in a transparent way.

A. Linear susceptibility $\chi^{(i)}(\omega_1)$ and the element m_{11}

Consider the case when the field ε_2 is absent and initially the system is in state $|i\rangle$, ($p^{(f)}=0$, $p^{(i)}=1$). We calculate $\chi^{(i)}$ by ignoring the radiative decay of the state $|E\rangle$. Since $v_{Ef}=0$ and since the radiative decay is ignored the matrix m becomes scalar with $m_{11} \neq 0$. From (A8) and (A9) we have

$$\hat{\Phi}_1^{(i)} = -\frac{i m_{11}}{1+m_{11}}, \quad \hat{\psi}_i^{(i)} = z^{-1} \left[1 - \frac{m_{11}}{1+m_{11}} \right] . \tag{4.1}$$

To obtain susceptibilities, these are to be used in (3.7), i.e., in the equation

$$\hat{\chi}^{(i)}(z,\omega_1) = -\int_0^\infty dt \frac{e^{-zt}}{\varepsilon_1^2} \Phi_1^{(i)}(t) \psi_i^{(i)*}(t) . \tag{4.2}$$

The linear susceptibility is obtained by evaluating $\Phi_1(t)\psi_i^*(t)$ to second order in ε_1. From (A10) we see that $m_{11} \sim O(\Omega_i)$, i.e., $O(\varepsilon_1^2)$ and hence we can substitute

$$\hat{\Phi}_1^{(i)} \sim -i m_{11}, \quad \psi_i(t) \sim 1 \tag{4.3}$$

to get

$$\hat{\chi}^{(i)}(z,\omega_1) = \frac{i m_{11}}{\varepsilon_1^2} \tag{4.4}$$

$$= \frac{2i\, |\mathbf{d}_{ia} \cdot \boldsymbol{\varepsilon}_1|^2}{\varepsilon_1^2 \Gamma z} \left[\frac{\left[1 - \frac{i}{q_i} \right]^2}{\frac{2z}{\Gamma} + 1 - i\alpha} + \frac{1}{q_i^2} \right] , \tag{4.5}$$

where (A10) has been used. Thus the element m_{11} gives the transient linear susceptibility associated with the transition $|i\rangle \leftrightarrow |E\rangle$. A number of linear and nonlinear absorption experiments can be interpreted in terms of the steady-state susceptibility which has the simpler form[27]

$$\chi^{(i)}(\omega_1) = \frac{2i\, |\mathbf{d}_{ia} \cdot \boldsymbol{\varepsilon}_1|^2}{\varepsilon_1^2 \Gamma} \left[\frac{\left[1 - \frac{i}{q_i} \right]^2}{1 - i\alpha} + \frac{1}{q_i^2} \right] , \tag{4.6}$$

which leads to the well-known absorption profile.

Similarly the element m_{22} of the matrix m is related to the transient susceptibility at ω_2 associated with the transition $|f\rangle \leftrightarrow |E\rangle$,

$$\hat{\chi}^{(f)}(z,\omega_2) = \left. \frac{i m_{22}}{\varepsilon_2^2} \right|_{z = z - i\Delta_f} . \tag{4.7}$$

Note that the usual two-photon ionization to a flat continuum can also be studied[12] in terms of the susceptibility $\chi^{(i)}$ if we identify the intermediate state with the autoionizing state $|a\rangle$. Here Γ is to be identified with the rate of ionization of the intermediate state.

B. Raman susceptibility and the element m_{21}

The Raman susceptibility can be obtained by calculating $\hat{\chi}^{(f)}(z,\omega_2)$ to second order in ε_1 and to zero order in ε_2

assuming that initially $p^{(i)}=1$, $p^{(f)}=0$. Under these conditions (3.8) reduces to

$$\hat{\chi}^{(f)}(z,\omega_2)=-\int_0^\infty dt\frac{e^{-zt}}{\varepsilon_2^2}\Phi_2^{(i)}(t)\psi_f^{(i)*}(t)\ , \tag{4.8}$$

where the product $\Phi_2^{(i)}\psi_f^{(i)*}$ is to be evaluated to order $\Omega_i\Omega_f$. From (A8) and (A9), ψ's and Φ's are known to be

$$\begin{aligned}\hat{\Phi}_2^{(i)}&=-i\left[\frac{m}{1+m}\right]_{21}\\&=\frac{-im_{21}}{(1+m_{11})(1+m_{22})-m_{12}m_{21}}\ ,\\\hat{\psi}_f^{(i)}&=(z+i\Delta_f)^{-1}(1+m)_{21}^{-1}\ .\end{aligned} \tag{4.9}$$

We also see that $m_{12}\sim O(\sqrt{\Omega_i\Omega_f})$, $m_{11}\sim O(\Omega_i)$, $m_{22}\sim O(\Omega_f)$. Therefore, for the calculation of the Raman susceptibility it is sufficient to use

$$\hat{\Phi}_2^{(i)}\simeq -im_{21},\quad \hat{\psi}_f^{(i)}\simeq-(z+i\Delta_f)^{-1}m_{21}\ .$$

Thus m_{21} is connected directly with the probability amplitude of finding the system in the state $|f\rangle$ given that it was in the state $|i\rangle$ at $t=0$. Similarly m_{12} is connected with the probability amplitude of finding the system in the state $|i\rangle$ given that it was in the state $|f\rangle$ at $t=0$.

Elements such as m_{33},m_{44} are connected with the radiative decay of the Fano state and this is shown in Appendix C.

C. Effect of radiative decay processes on linear susceptibility $\chi^{(i)}(\omega_1)$

We next consider how the susceptibility (4.6) is modified due to spontaneous emission from excited states.[28] We need to compute $\Phi_1(t)\psi_i^*(t)$ to second order in ε_1, zero order in ε_2 but keeping the elements m_{33}, etc., nonzero as $\gamma_i\neq0$, $\gamma_f\neq0$. In the limit of weak fields (i.e., to order ε_1^2), recycling effects do not contribute ($I^{(i)}=I^{(f)}=0$). Using (4.2) and the results from Appendix A, calculations show that

$$\chi^{(i)}(\omega_1)=\frac{i\Gamma}{2}\frac{q_i^2\Omega_i}{\varepsilon_1^2}\frac{\left[\frac{\gamma_f}{\Gamma}\left[\frac{1}{q_i}-\frac{1}{q_f}\right]^2+\left[1-\frac{i}{q_i}\right]^2+\frac{1}{q_i^2}(1-i\alpha)\right]}{\psi(\eta-i\alpha+i\Delta_a)}\ , \tag{4.10}$$

where

$$\alpha=\frac{2}{\Gamma}(\omega_1-E_a)\ ,$$

$$\psi=1+\frac{\gamma_i}{\Gamma q_i^2}+\frac{\gamma_f}{\Gamma q_f^2},\quad \Delta_a=-\frac{2}{\psi}\left[\frac{\gamma_i}{\Gamma q_i}+\frac{\gamma_f}{\Gamma q_f}\right]\ , \tag{4.11}$$

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

The expression for $\chi^{(f)}(\omega_1)$ is obtained from (4.10) by interchanging the indices i and f and by replacing α by $(2/\Gamma)(\omega_2+E_f-E_a)$.

In the limit of large $q\to\infty$ (radiative recombination now negligible), one has a symmetric profile with a width $(\Gamma+\gamma_i+\gamma_f)$. For finite q the lines are asymmetric. Note also the asymmetry of the line with respect to the decay in two channels, i.e., with respect to γ_i and γ_f. Figures 2 and 3 show the dependence of the imaginary and real parts of $\chi^{(i)}$ on the radiative-decay processes. The structure of the absorption profiles is similar to Fano profiles with radiation damping [as computed in I, Eq. (4.8)] though they are not identical because γ_i's$\neq0$. The imaginary part of (4.10) can be expressed in a more instructive form,

$$\mathrm{Im}\chi^{(i)}(\omega_1)=\frac{\Gamma}{2}\frac{\Omega_i}{\varepsilon_1^2\psi}\left[1+\frac{\bar{\alpha}P+Q\eta}{\eta^2+\bar{\alpha}^2}\right]\ , \tag{4.12}$$

where

$$\begin{aligned}P&=2q_i+\Delta_a\ ,\\Q&=q_i^2-\eta+\frac{\gamma_f}{\Gamma}\left[1-\frac{q_i}{q_f}\right]^2\ ,\\\bar{\alpha}&=\alpha-\Delta_a\ .\end{aligned} \tag{4.13}$$

The absorption profile has the same form as that discussed by Shore.[3] Parameters of the profile are now explicitly given in terms of the system parameters. Similar considerations yield the modifications in the Raman susceptibility due to the nonvanishing of γ_i and γ_f.

V. EFFECT OF STRONG COUPLING BETWEEN TWO BOUND STATES $|i\rangle$ AND $|a\rangle$ ON OPTICAL ABSORPTION

We now consider the intensity-dependent effects in optical absorption. We will calculate the susceptibility $\chi^{(f)}(\omega_2)$ which will depend on all powers of the electric field at ω_1, i.e., $\chi^{(f)}(\omega_2)$ will give the linear response of the system consisting of two strongly coupled bound states which decay via either autoionization or field-induced ionization. In addition we assume that the state can decay via spontaneous emission. For this purpose one has to compute quantities such as ψ_i and Φ_j to second order in ε_2, i.e., to order Ω_f but to all orders in Ω_i. The calculations are rather clumsy and hence we will present the results for various quantities needed in the evaluation of $\chi^{(f)}(\omega_2)$. Introducing the parameters $\mu^{(i)}$, $\mu^{(f)}$ defined by

$$\mu^{(i)}=q_i\left[\frac{\Gamma\Omega_i}{\gamma_i}\right]^{1/2},\quad \mu^{(f)}=q_f\left[\frac{\Gamma\Omega_f}{\gamma_f}\right]^{1/2} \tag{5.1}$$

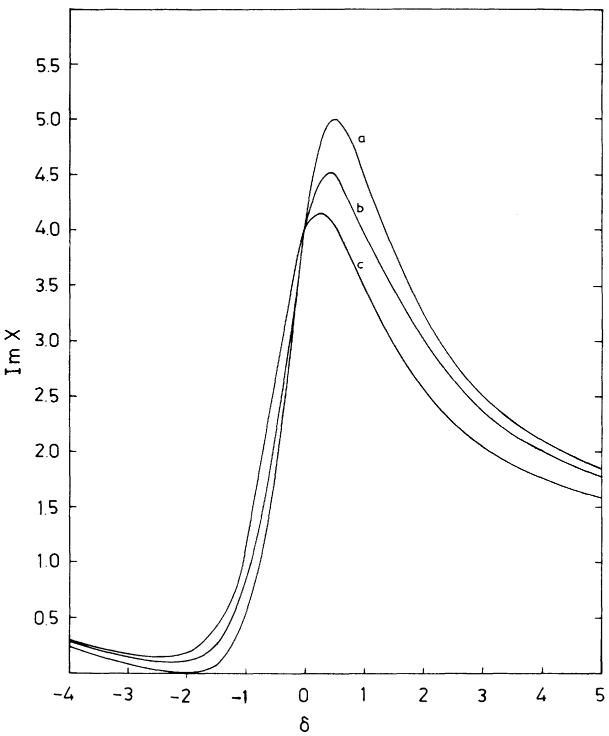

FIG. 2. Absorption profiles proportional to $\mathrm{Im}\chi^{(f)}(\omega_2)$ as a function of detuning $\delta=(2/\Gamma)(E_f+\omega_2-E_a)$ for $q_i=1$, $q_f=2$, $\alpha=1$ and for different values of the radiative decay parameters (a) $\gamma_i/\Gamma=\gamma_f/\Gamma=0$, (b) $\gamma_i/\Gamma=0.1$, $\gamma_f/\Gamma=0$, and (c) $\gamma_i/\Gamma=0.1$, $\gamma_f/\Gamma=0.1$.

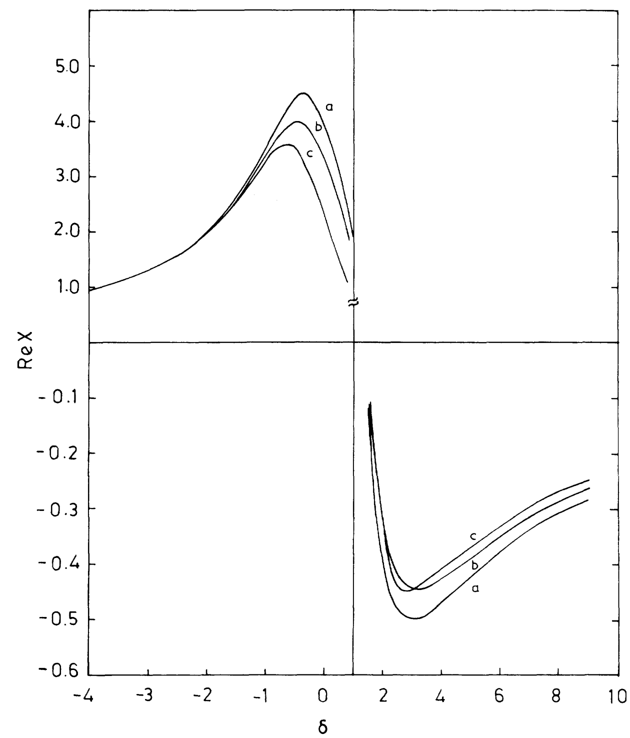

FIG. 3. Effects of radiative decay on the dispersion characteristics ($\mathrm{Re}\chi^{(f)}$) of autoionizing states for same parameters as in Fig. 2.

we find that

$$\det(1+m)^{-1}=D^{-1}(z)-\frac{D_1\Gamma}{2D^2}\frac{\mu^{(f)}\mu^{(f)}}{(z+i\Delta_f)} . \tag{5.2}$$

Here D's are defined in terms of the matrix m,

$$D(z)=(1+m_{44}) +(m_{33}+m_{33}m_{44}-m_{34}m_{43})\left[1+\frac{\mu^{(i)^2}\Gamma}{2z}\right], \tag{5.3}$$

$$D_1=(m_{33}m_{44}-m_{34}m_{43})\left[1+\frac{\mu^{(i)}\mu^{(i)}\Gamma}{2z}\right]+m_{44} .$$

On simplification $D(z)$ reduces to

$$D^{-1}(z)=\frac{z\Gamma}{2}\left[\frac{2z}{\Gamma}+1-i\alpha\right]/\psi(z-z_+)(z-z_-) , \tag{5.4}$$

where ψ has been defined earlier [Eq. (4.11)] and $z_\pm$ are the roots of the equation

$$\varepsilon^2-\left[\alpha+\Delta_a-i\left[\eta+\frac{\Omega_i}{\psi}\right]\right]\varepsilon+\alpha\Delta_a-\frac{\Omega_i}{\psi}q_i^2\left[1+\frac{\gamma_f}{\Gamma}\left[\frac{1}{q_i}-\frac{1}{q_f}\right]^2\right]-i\left[\alpha\eta-\frac{2\Omega_i}{\psi}q_i\right]=0 ,$$
$$z_\pm=-\frac{i\Gamma}{2}(\varepsilon_\pm-\alpha) . \tag{5.5}$$

These roots are known from I and were shown earlier to determine the structure of the spontaneous-emission spectrum.[21] Various ψ's and Φ's are found to be

$$\hat{\psi}_f^{(i)}(z)=\frac{\mu^{(i)}\mu^{(f)}m_{43}\Gamma}{2z(z+i\Delta_f)D(z)}+O((\mu^{(f)})^3) , \tag{5.6}$$

$$\hat{\Phi}_2^{(i)}(z)=\frac{i\mu^{(i)}\mu^{(f)}m_{43}\Gamma}{2zD(z)}+O((\mu^{(f)})^3) , \tag{5.7}$$

$$\hat{\psi}_f^{(f)}(z)=\frac{1}{(z+i\Delta_f)}\left[1-\frac{D_1\mu_f^2\Gamma}{2D(z+i\Delta_f)}\right] +O((\mu^{(f)})^4) , \tag{5.8}$$

$$\hat{\Phi}_2^{(f)}(z)=-\frac{i\mu^{(f)^2}\Gamma D_1}{2D(z+i\Delta_f)}+O((\mu^{(f)})^4) . \tag{5.9}$$

On using (5.4) and (A10) we find that $\Phi_2^{(i)}(t)$ has the structure

$$\Phi_2^{(i)}(t)=Ae^{z_+t}+Be^{z_-t} . \tag{5.10}$$

The roots $z_\pm$ are known from I where it was shown that $\mathrm{Re}z_\pm<0$ as long as $\gamma_i,\gamma_f\neq 0$. For a realistic system γ's will never be zero though they may be negligibly small and hence

$$\lim_{t\to\infty}\Phi_2^{(i)}(t)=0 . \tag{5.11}$$

It may be added that the two limits $\gamma\to 0$, $t\to\infty$ do not commute. Physical ordering of limits obviously should correspond to $\lim_{\gamma\to 0}\lim_{t\to\infty}$. The functions $\Phi_2^{(f)}$ and $\psi_f^{(f)}(t)$ have oscillating contributions in addition to the decaying contributions and hence in the steady state only the oscillating terms will contribute. Since $\Phi_2^{(f)}(t)$ is already of order $\mu^{(f)^2}$, it is therefore sufficient to take

$$\psi^{(f)}(t)=e^{-i\Delta_f t} . \tag{5.12}$$

Using (5.9) and (5.12) we obtain the result

$$\lim_{t\to\infty}\psi_f^{(f)*}(t)\Phi_2^{(f)}(t)=-\frac{i\mu^{(f)^2}\Gamma D_1(-i\Delta_f)}{2D(-i\Delta_f)}+O((\mu^{(f)})^4) . \tag{5.13}$$

The steady-state susceptibility can now be obtained by using (5.11) and (5.13) in (3.8),

$$\chi^{(f)}(\omega_2)=[p^{(f)}+\hat{I}^{(f)}(0)]\frac{i\mu^{(f)}\mu^{(f)}\Gamma D_1(-i\Delta_f)}{2D(-i\Delta_f)\varepsilon_2^2} . \tag{5.14}$$

The function $\hat{I}^{(f)}(0)$ now needs to be calculated to zero order in Ω_f. Equation (2.17) gives the function $\hat{I}^{(f)}$. Setting $\Omega_f=0$ leads to (cf. Appendix B)

$$\hat{T}^{ff}=0,\quad \hat{T}^{fi}=0 \tag{5.15}$$

and hence

$$\hat{I}^{(f)}=p^{(i)}\hat{T}^{if}/(1-\hat{T}^{ii}) . \tag{5.16}$$

The nonvanishing elements $\hat{T}^{if},\hat{T}^{ii}$ are obtained from (B2) and (B3),

$$\hat{T}^{if}(0)=2\left[\frac{P_+P_+^*}{z_++z_+^*}+\frac{P_+^*P_-}{z_+^*+z_-}+\frac{P_-^*P_+}{z_++z_-^*}+\frac{P_-P_-^*}{z_-+z_-^*}\right] , \tag{5.17}$$

$$\hat{T}^{ii}(0)=2\left[\frac{Q_+Q_+^*}{z_++z_+^*}+\frac{Q_+^*Q_-}{z_+^*+z_-}+\frac{Q_-^*Q_+}{z_++z_-^*}+\frac{Q_-Q_-^*}{z_-+z_-^*}\right] , \tag{5.18}$$

$$P_\pm=\frac{2}{\Gamma}\lim_{z\to z_\pm}(z-z_\pm)P ,\qquad Q_\pm=\frac{2}{\Gamma}\lim_{z\to z_\pm}(z-z_\pm)Q . \tag{5.19}$$

Thus $P_\pm$ ($Q_\pm$) are the residues of the function $P(Q)$ at the poles $z_\pm$,

$$P=\left[\frac{\Gamma}{2}\right]^{1/2}\frac{m_{43}\mu^{(i)}}{zD(z)} , \tag{5.20}$$

$$Q=\left[\frac{\Gamma}{2}\right]^{1/2}\frac{\mu^{(i)}(m_{33}m_{44}-m_{34}m_{43}+m_{33})}{D(z)} . \tag{5.21}$$

A. Probing of laser-induced continuum structure—optical mixing of a bound state and an autoionizing state

Our final expression for the intensity-dependent susceptibility is given by (5.14) with $\hat{I}^{(f)}$ determined by Eqs. (5.16)–(5.21). This will now be used for specific applications. The frequency dependence of $\chi^{(f)}(\omega_2)$ is determined by the roots of $D(-i\Delta_f)$ which depend on the coupling between $|i\rangle$ and $|E\rangle$, i.e., on the strength of the field one and the frequency ω_1. The susceptibility $\chi^{(f)}(\omega_2)$ acquires a doublet structure, the resolution of the doublet depends on E_1, ω_1, and the q values of the transitions.

From (5.14) we find that $\chi^{(f)}(\omega_2)=0$ if $p^{(f)}=0$, $p^{(i)}=1$, and if spontaneous emission is negligible. The Raman susceptibility is zero if the radiative decay of states is negligible and if the transition $|i\rangle\leftrightarrow|E\rangle$ is strongly pumped. The susceptibility $\chi^{(f)}(\omega_2)$ is nonzero for $p^{(f)}=1$, $p^{(i)}=0$ even if $\gamma_i\sim\gamma_f\sim 0$. In the conventional language this situation corresponds to inverse Raman effect.

In the limit $\gamma_i=\gamma_f=0$, we can obtain a much more transparent expression for the intensity-dependent susceptibility $\chi^{(f)}(\omega_2)$. On simplification Eq. (5.14) leads to

$$\chi^{(f)}(\omega_2)=\frac{i\Gamma\Omega_f}{2\varepsilon_2^2}\frac{\left[(\delta-\alpha)(\delta+2q_f+iq_f^2)-q_i^2\Omega_i\left[\frac{q_f}{q_i}-1\right]^2\right]}{(\delta-\varepsilon_+)(\delta-\varepsilon_-)} , \tag{5.22}$$

where

$$\delta=\frac{2}{\Gamma}(E_f+\omega_2-E_a) . \tag{5.23}$$

We now present numerical results for the dependence of $\chi^{(f)}$ on the intensity Ω_i and the radiative-decay parameters. The typical dependence on these parameters is displayed in Figs. 4–8. Figures 4 and 5 give the behavior of the imaginary and real parts of $\chi^{(f)}$ when the radiative decay is negligible. The real (imaginary) part of χ is typically dispersive (absorptive) in nature. Figures 4 and 5 show the very narrow resonance in optical absorption cor-

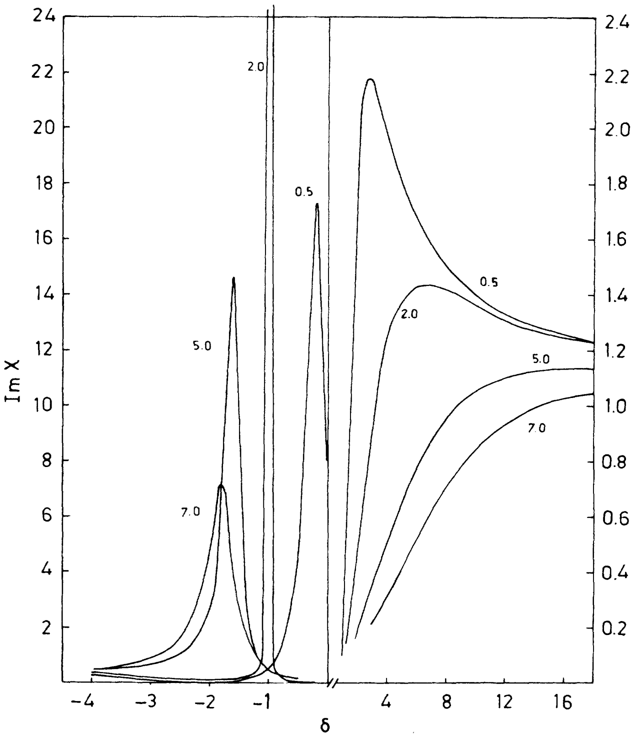

FIG. 4. Absorption profiles (proportional to $\mathrm{Im}\chi^{(f)}$) for two strongly coupled autoionizing states for $\gamma_i/\Gamma=\gamma_f/\Gamma=0.001$. Different curves are labeled by the values of Ω_i. The scale on the right corresponds to the curves on the right side of the central line. The behavior in the region where there is a minimum is not shown though the minimum values are nonzero.

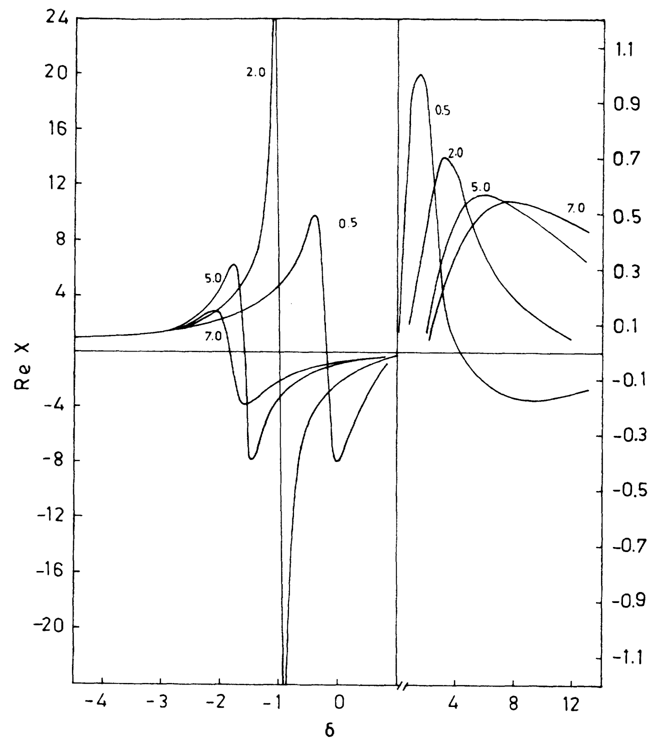

FIG. 5. Dispersive properties of the susceptibility $\chi^{(f)}(\omega_2)$ for two strongly coupled bound states for same parameters as in Fig. 4.

responding to system parameters such that

$$\Omega_i=1+\frac{\alpha}{q_i} \ . \tag{5.24}$$

This spike is well known[18,19] in related studies on the spectrum of photoelectrons obtained by strong field-induced autoionization. Figure 6 shows the sensitiveness of the optical absorption on the tuning of the field. For the problem of two strongly coupled autoionizing levels, Figs. 4 and 6 show how the absorption can differ depending on the relative location of the two autoionizing levels. Note the presence of two interference minima in Fig. 6. Deng and Eberly[24] have discussed the presence of two minima in the photoelectron profiles when both the transitions are strongly driven. Figures 7 and 8 give the behavior of absorption profiles and dispersion profiles for nonzero γ values.

B. Two strongly coupled autoionizing states either by dc field or by internal interactions

Recent experiments[7–10] on the inhibition[29] of autoionization are essentially connected with a real root of the polynomial (5.5) for certain values of the coupling between the two autoionizing states and the relative separation between the two states. We now obtain explicitly the result for the situation discussed in Ref. 7. The state $|i\rangle$ is a real autoionizing state. We assume that its autoionization is negligible, i.e., we take the limit $\tilde{v}_{Ei}\to 0$. The parameter α now becomes the relative separation between two autoionizing states. The dc field connects the states $|i\rangle$ and $|a\rangle$. Let Ω_0 be the strength of such interaction, i.e.,

$$\Omega_0=\frac{4\tilde{v}_{ai}^2}{\Gamma^2} \ . \tag{5.25}$$

We can obtain the intensity-dependent susceptibility for the situation of Ref. 7 by taking the limits[11]

$$\Omega_i\to 0,\ \ q_i\to\infty,\ \ \Omega_i q_i^2=\text{const}=\Omega_0 \tag{5.26}$$

in the expression (5.22). The relevant susceptibility is then found to be

$$\chi^{(f)}(\omega_2)=\frac{i\Gamma\Omega_f}{2\varepsilon_2^2}\frac{[(\delta-\alpha)(\delta+2q_f+iq_f^2)-\Omega_0]}{(\delta-\varepsilon_+)(\delta-\varepsilon_-)} \ , \tag{5.27}$$

where $\varepsilon_\pm$ are now given by the solution of

$$\varepsilon^2-[\alpha-i]\varepsilon-\Omega_0-i\alpha=0 \ . \tag{5.28}$$

We can now study the behavior of $\chi^{(f)}(\omega_2)$ as a function of dc field strength Ω_0. We show some typical results in Figs. 9 and 10. The general behavior of $\mathrm{Im}\chi^{(f)}$ is in agreement with the observations reported in Ref. 7. The imaginary part of the susceptibility (5.27) is identical to the expression for the photoelectron profiles as calculated

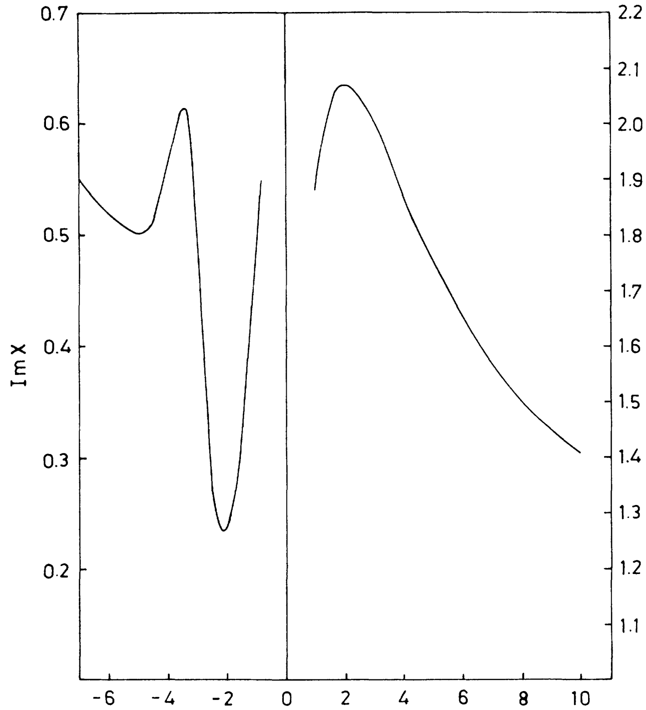

FIG. 6. Same as in Fig. 4, but the strong field is tuned differently, $\alpha=-3$. Other parameters are chosen to be $\Omega_i=2$, $q_i=1$, $q_f=2$. For brevity we show the behavior in different regions. The two curves join continuously.

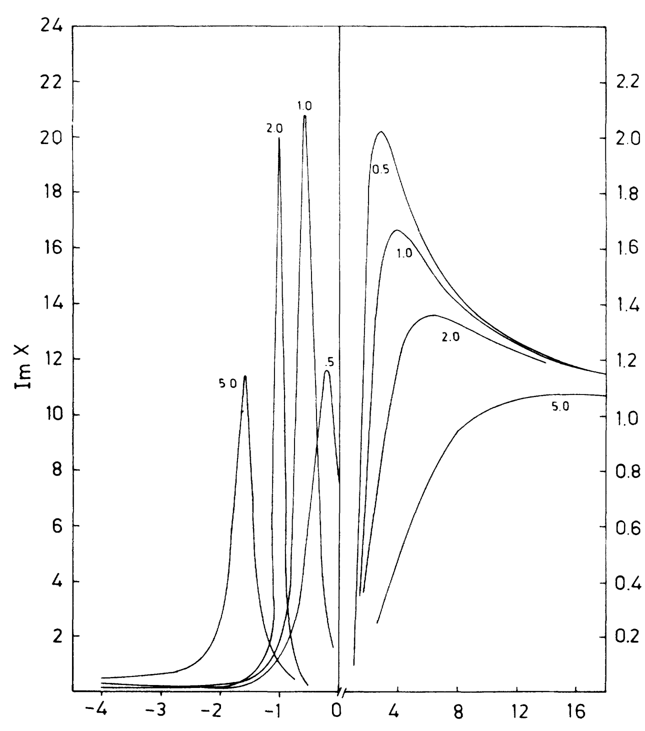

FIG. 7. Absorption profiles for the same values of the parameters as in Fig. 4, except now $\gamma_1/\Gamma=\gamma_f/\Gamma=0.05$. The actual values for the curve marked $\Omega=2.0$ are twice that shown.

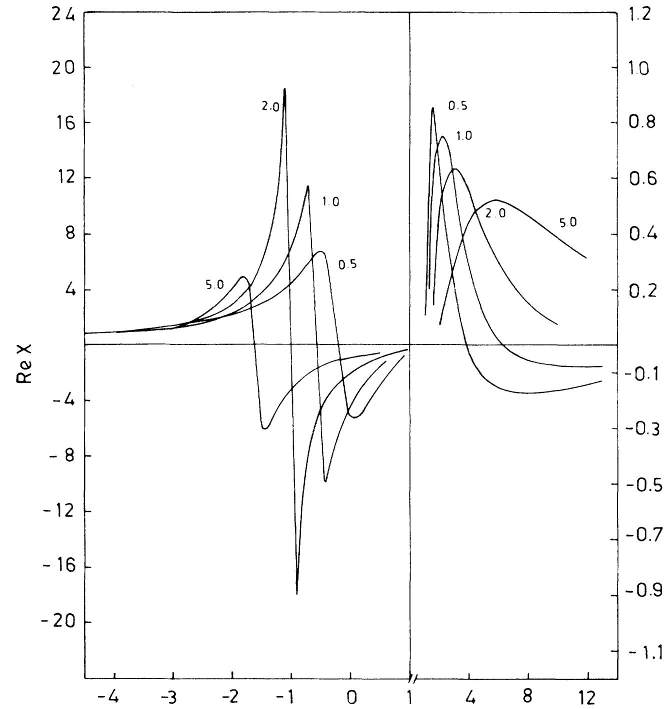

FIG. 8. Dispersion characteristics of the intensity-dependent susceptibility $\chi^{(f)}(\omega_2)$, for $\gamma_i/\Gamma=\gamma_f/\Gamma=0.05$. The other parameters are the same as in Fig. 5.

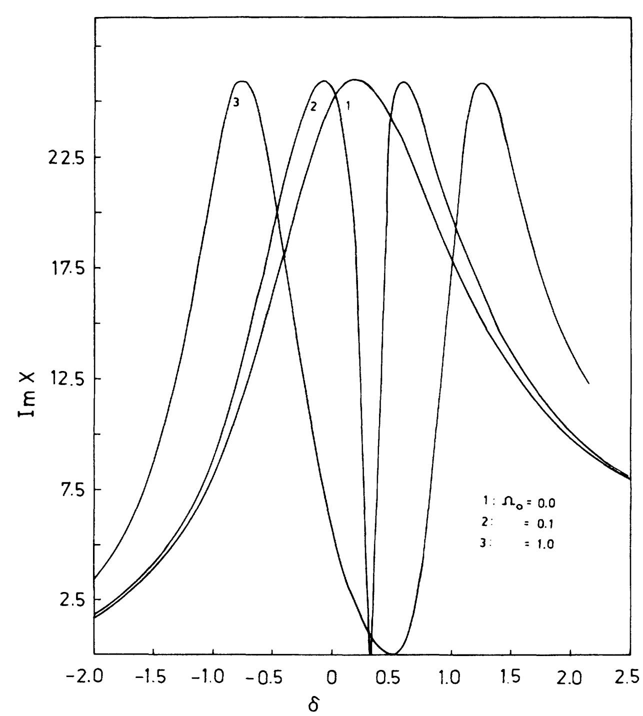

FIG. 9. Imaginary part of the dc-field-dependent susceptibility $\chi^{(f)}$ [Eq. (5.27)] for two coupled autoionizing states. Curves are marked by the values of Ω_0. Other parameters are taken to be $q_f=5$, $\alpha=0.03$.

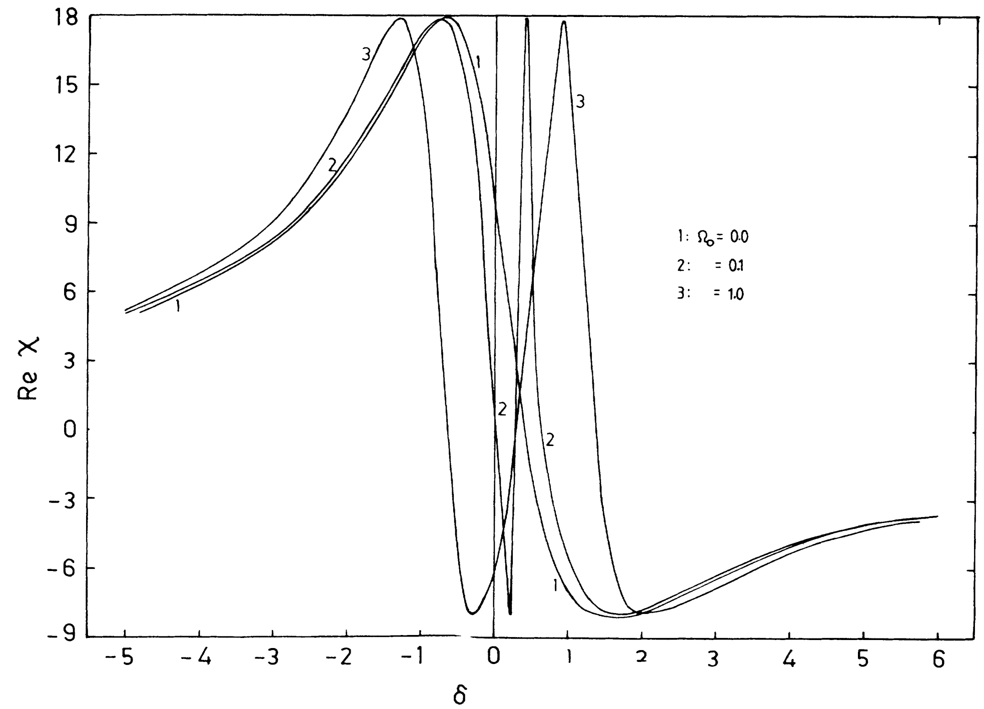

FIG. 10. Dispersive properties of the dc-field-dependent susceptibility for the same parameters as in Fig. 9.

by Agarwal *et al.* [Ref. 11, Eq. (8) with $\gamma_f=0$]. This is expected since, in the absence of the radiative decay, any absorbed energy must result in photoionization.

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APPENDIX A: EXPLICIT CALCULATION OF THE AUXILIARY MATRICES σ

In this appendix we present the solution of Eqs. (2.9) so that the matrix σ can be constructed. On taking Laplace transforms of Eqs. (2.9) and on formally solving for ψ_i, ψ_f assuming that $\psi_E(0)=0$, we get the equations

$$\hat{\psi}_i=z^{-1}\psi_i(0)-iz^{-1}\int dE\, v_{Ei}^*\hat{\psi}_E \;, \tag{A1}$$

$$\hat{\psi}_f=(z+i\Delta_f)^{-1}\psi_f(0)-i(z+i\Delta_f)^{-1}\int dE\, v_{Ef}^*\hat{\psi}_E \;, \tag{A2}$$

$$\hat{\psi}_{E_1}+\sum_{i=1}^{4}\int dE\, K_i(E_1)L_i(E)\hat{\psi}_E = -iK_1(E_1)\psi_i(0)-iK_2(E_1)\psi_f(0) \;. \tag{A3}$$

Here the functions K and L are defined by

$$\begin{aligned}
K_1(E)&=v_{Ei}[z(z+i\Delta_E)]^{-1} \;,\\
L_1(E)&=v_{Ei}^*=v_{iE} \;,\\
K_2(E)&=v_{Ef}(z+i\Delta_E)^{-1}(z+i\Delta_f)^{-1} \;,\\
L_2(E)&=v_{Ef}^* \;,\\
K_3(E)&=\left[\frac{\gamma_i}{2}\right]^{1/2}(z+i\Delta_E)^{-1}B_{Ea}^* \;,\\
L_3(E)&=\left[\frac{\gamma_i}{2}\right]^{1/2}B_{Ea} \;,\\
K_4(E)&=\left[\frac{\gamma_f}{2}\right]^{1/2}C_{Ea}^*(z+i\Delta_E)^{-1} \;,\\
L_4(E)&=\left[\frac{\gamma_f}{2}\right]^{1/2}C_{Ea} \;.
\end{aligned} \tag{A4}$$

The integral equation (A3) can be solved analytically as it involves a separable kernel. In terms of 4×4 matrix m defined by

$$m_{ji}=\int L_j(E)K_i(E)dE \;, \tag{A5}$$

one finds that ψ_E is given by

$$\hat{\psi}_E=-i\sum_j K_j(E)[(1+m)_{j1}^{-1}\psi_i(0)+(1+m)_{j2}^{-1}\psi_f(0)] \;. \tag{A6}$$

As discussed in Sec. III the calculations of the polarization also involve the quantities Φ defined by

$$\hat{\Phi}_i = \int L_i(E)\hat{\psi}_E dE \ . \tag{A7}$$

Using (A3) one finds the solution for Φ's,

$$\hat{\Phi}_j = -i\left[\frac{m}{1+m}\right]_{j1}\psi_i(0) - i\left[\frac{m}{1+m}\right]_{j2}\psi_f(0) \ . \tag{A8}$$

We can rewrite $\hat{\psi}_i, \hat{\psi}_f$ in terms of $\hat{\Phi}$'s as

$$\begin{aligned}\hat{\psi}_i &= z^{-1}[\psi_i(0) - i\hat{\Phi}_1] \ , \\ \hat{\psi}_f &= (z+i\Delta_f)^{-1}[\psi_f(0) - i\hat{\Phi}_2] \ .\end{aligned} \tag{A9}$$

The structure of the m matrix depends on the energy dependence of the matrix elements v_{Ei}, i.e., it depends on the structure of the continuum. For a flat continuum v_{Ei} can be taken to be energy independent. However, for our system the continuum described by Fano states $|E\rangle$ is not flat, though the original continuum [described by $|E)$] is flat. Equation (2.4) gives the resonant structure of the matrix elements. We assume that $\tilde{v}_{Ei}$ are approximately energy independent. Calculations then lead to the following results for the various elements of m:

$$\begin{aligned}
m_{11} &= \frac{2}{\gamma_i z} m_{33}\,|\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1|^2 \ , \\
m_{22} &= \frac{2}{\gamma_f(z+i\Delta_f)} m_{44}\,|\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2|^2 \ , \\
m_{13} &= \left[\frac{2}{\gamma_i}\right]^{1/2}(\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1)m_{33} = m_{31}z \ , \\
m_{24} &= \left[\frac{2}{\gamma_f}\right]^{1/2}(\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2)m_{44} = m_{42}(z+i\Delta_f) \ , \\
m_{14} &= \left[\frac{2}{\gamma_i}\right]^{1/2}(\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1)m_{34} = m_{41}z\frac{m_{34}}{m_{43}} \ , \\
m_{23} &= \left[\frac{2}{\gamma_f}\right]^{1/2}(\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2)m_{43} = m_{32}(z+i\Delta_f)\frac{m_{43}}{m_{34}} \ , \\
m_{12} &= (\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1)(\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2)\frac{2}{\sqrt{\gamma_i\gamma_f}}\frac{m_{34}}{(z+i\Delta_f)} \ , \\
m_{21} &= (\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1)(\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2)\frac{2}{z\sqrt{\gamma_i\gamma_f}} m_{43} \ , \\
m_{33} &= \frac{\gamma_i}{\Gamma}\left[\frac{\left[1-\frac{i}{q_i}\right]^2}{\frac{2z}{\Gamma}+1-i\alpha} + \frac{1}{q_i^2}\right] , \\
m_{44} &= \frac{\gamma_f}{\Gamma}\left[\frac{\left[1-\frac{i}{q_f}\right]^2}{\frac{2z}{\Gamma}+1-i\alpha} + \frac{1}{q_f^2}\right] , \\
m_{34} &= m_{43} = \frac{\sqrt{\gamma_i\gamma_f}}{\Gamma}\left[\frac{\left[1-\frac{i}{q_i}\right]\left[1-\frac{i}{q_f}\right]}{\frac{2z}{\Gamma}+1-i\alpha} + \frac{1}{q_iq_f}\right] ,
\end{aligned} \tag{A10}$$

$$\alpha = \frac{2}{\Gamma}(\omega_1 - E_a), \ \ \Omega_i = \frac{2\pi}{\Gamma}|\tilde{v}_{\varepsilon i}|^2, \ \ \Omega_f = \frac{2\pi}{\Gamma}|\tilde{v}_{\varepsilon f}|^2 \ , \tag{A11}$$

$$|\mathbf{d}_{ia}\cdot\boldsymbol{\varepsilon}_1|^2 = \left[\frac{\Gamma q_i}{2}\right]^2\Omega_i, \ \ |\mathbf{d}_{fa}\cdot\boldsymbol{\varepsilon}_2|^2 = \left[\frac{\Gamma q_f}{2}\right]^2\Omega_f \ . \tag{A12}$$

The parameter $\Omega_i(\Omega_f)$ is a measure of the strength of the transition from the state $|i\rangle$ ($|f\rangle$ to the continuum of states $|E)$ caused by a field with amplitude ε_1 (ε_2). It is the rate of ionization (measured in units of Γ) of the level $|i\rangle$ due to the applied field with amplitude ε_i. In Sec. IV and Appendix C we show how various element of m correspond to the elementary processes among the unperturbed states of the system. Note that the elements of m are already in terms of the matrix elements of various interactions between the Fano continuum $|E\rangle$ and the states $|i\rangle$ and $|f\rangle$. More explicitly, one has the relations of the form

$$\begin{aligned}\lim_{z\to 0} zm_{11}(z) &= \lim_{z\to 0}\int\frac{|v_{Ei}|^2 dE}{(z+i\Delta_E)} \\ &= |v_{E_a i}|^2 - iP\int\frac{|v_{Ei}|^2 dE}{\Delta_E} \ .\end{aligned} \tag{A13}$$

APPENDIX B: CONSTRUCTION OF T MATRICES

We now use solutions from Appendix A to construct T matrices as defined by (2.16). Let $\psi^{(i)}$ ($\psi^{(f)}$) be the solution for the functions ψ corresponding to the initial condition $\psi_i(0)=1$, $\psi_f(0)=0$ [$\psi_f(0)=1$, $\psi_i(0)=0$]. Thus the superscript on functions such as ψ and Φ will indicate the initial condition. From (2.8) one then has

$$\begin{aligned}\sigma^{(i)}_{\alpha\beta}(t) &= \psi^{(i)}_\alpha(t)\psi^{(i)*}_\beta(t), \sigma^{f}_{\alpha\beta}(t) \\ &= \psi^{(f)}_\alpha(t)\psi^{(f)*}_\beta(t) \ .\end{aligned} \tag{B1}$$

The T matrices then acquire a factorized form, for example,

$$\begin{aligned}T^{ii}(t) &= \frac{\gamma_i}{2}\int dE_1\int dE B^*_{E_1 a}B_{Ea}\sigma^{(i)}_{EE_1}(t) + \text{c.c.} \\ &= \frac{\gamma_i}{2}\int dE_1 B^*_{E_1 a}\psi^{(i)*}_{E_1}(t)\int dE\, B_{Ea}\psi^{(i)}_E(t) + \text{c.c.} \\ &= \left|\left[\frac{\gamma_i}{2}\right]^{1/2}\int dE\, B_{Ea}\psi^{(i)}_E(t)\right|^2 + \text{c.c.} \ ,\end{aligned}$$

which on using (A4) becomes

$$\begin{aligned}T^{ii}(t) &= 2\left|\int dE\, L_3(E)\psi^{(i)}_E(t)\right|^2 \\ &= 2|\Phi^{(i)}_3(t)|^2 \ ,\end{aligned} \tag{B2}$$

where (A7) has been used. Similarly other T matrices are obtained,

$$\begin{aligned}&T^{fi}(t) = 2|\Phi^{(f)}_3(t)|^2, \ \ T^{if}(t) = 2|\Phi^{(i)}_4(t)|^2 \ , \\ &T^{ff}(t) = 2|\Phi^{(f)}_4(t)|^2 \ .\end{aligned} \tag{B3}$$

For completeness sake we record $\Phi_\beta^{(\alpha)}$'s, ψ_j's as obtained from Eqs. (A8) and (A9),

$$\hat{\Phi}_\alpha^{(i)} = -i\left[\frac{m}{1+m}\right]_{\alpha 1}, \quad \hat{\Phi}_\alpha^{(f)} = -i\left[\frac{m}{1+m}\right]_{\alpha 2}, \tag{B4}$$

$$\hat{\psi}_i^{(i)} = z^{-1}(1+m)_{11}^{-1}, \quad \hat{\psi}_f^{(f)} = (z+i\Delta_f)^{-1}(1+m)_{22}^{-1},$$
$$\hat{\psi}_f^{(i)} = (z+i\Delta_f)^{-1}(1+m)_{21}^{-1}, \quad \hat{\psi}_i^{(f)} = z^{-1}(1+m)_{12}^{-1}. \tag{B5}$$

APPENDIX C: BUILD UP OF POPULATION IN THE GROUND STATE AND THE ELEMENT m_{33}

In order to see the physical meaning of the matrix elements m_{33}, etc., let us consider the case of only the radiative decay of the Fano state in the absence of any exciting field, i.e., we set $\psi_i(0)=\psi_f(0)=0$, $\psi_E(0)\neq 0$, $\Omega_i=\Omega_f=0$. For simplicity we also set $\gamma_f=0$. Then using (2.2) we find that the population in the state $|i\rangle$ builds up according to

$$\dot{\rho}_{ii} = \frac{\gamma_i}{2}\int dE_1 \int dE_2 B_{E_1a}^* B_{E_2a}\rho_{E_2E_1} + \text{c.c.}, \tag{C1}$$

where $\rho_{E_1E_2}$ is to be obtained from the solution of the equation

$$\dot{\rho}_{E_1E_2} = -i(E_1-E_2)\rho_{E_1E_2} - \frac{\gamma_i}{2}\int dE\, B_{E_1a}^* B_{Ea}\rho_{EE_2} - \frac{\gamma_i}{2}\int dE\, B_{E_2a} B_{Ea}^*\rho_{E_1E}. \tag{C2}$$

Equation (C2) admits a solution of the form

$$\rho_{E_1E_2}(t) = \psi_{E_1}(t)\psi_{E_2}^*(t), \tag{C3}$$

where

$$\dot{\psi}_{E_1} = -iE_1\psi_{E_1} - \frac{\gamma_i}{2}\int B_{E_1a}^* B_{Ea}\psi_E dE. \tag{C4}$$

We next introduce the function $f(t)$ by

$$f(t) = \int dE\, B_{Ea}\psi_E(t). \tag{C5}$$

Using (C4) we find the Laplace transform of the function $f(t)$,

$$\hat{f}(z) = \left[1+\frac{\gamma_i}{2}\int dE_1 \frac{|B_{E_1}a|^2}{z+iE_1}\right]^{-1}\int dE \frac{\psi_E(0)B_{Ea}}{(z+iE)}. \tag{C6}$$

The rate of change of the population in the ground state can be expressed in terms of $f(t)$,

$$\dot{\rho}_{ii} = \gamma_i |f(t)|^2. \tag{C7}$$

From Eq. (A5) we also have

$$m_{33} = \frac{\gamma_i}{2}\int dE_1 \frac{|B_{E_1}a|^2}{z+i\Delta_E}. \tag{C8}$$

From (C6) and (C8) it is evident that the time dependence of $f(t)$ is determined from the zeros of $1+m_{33}$, with m_{33} computed with $\omega_1=0$, i.e., from

$$\frac{2z}{\Gamma} = -\frac{2}{\Gamma}E_a - \left[1+\frac{\gamma_i}{\Gamma q_i^2}\right]^{-1}\left[1+\frac{\gamma_i}{\Gamma}\left[1-\frac{2i}{q_i}\right]\right]. \tag{C9}$$

This thus establishes the importance of m_{33} in the problem of the radiative decay of the Fano state $|E\rangle$.

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