Radiative decay of autoionizing states in laser fields. II. Photoemission spectra

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The fluorescence produced by an autoionizing state under strong pumping by a coherent laser field is considered. It is shown how the spectrum of emitted photons and the total number of photons are related to the two-time and single-time atomic expectation values, and explicit expressions are derived for them. Numerical results for the time development of the number of photons ejected are presented for finite times and in the long-time limit. The photon spectra are shown to exhibit ac Stark splitting of the states involved. The spectrum is a doublet for transitions to a stable final state and a triplet for transitions connected back to the initial, strongly driven state. The decay of an autoionizing state in the absence of a laser driving field is considered, and the square of the Fano q value is interpreted as the ratio of two decay rates.

I. INTRODUCTION

In paper I of this series,[1] we developed a general theory to describe the radiative decay of autoionizing states in the presence of strong laser fields.[2] The general theory was then applied in studying the dependence of the photoelectron spectra on spontaneous emission and laser intensity. In the present paper we analyze the characteristics of the fluorescence produced by the autoionizing states, and show how studies of the fluorescence[3–6] can provide important information on the characteristics of the autoionizing states.

In Sec. II we show how the spectrum of the emitted photons and the total number of photons are related to the two-time and single-time atomic expectation values. We also show that in the steady state the dipole moment is zero, and hence the emitted radiation has no coherent component at the driving frequency of the laser. In Sec. III we calculate the total number of photons emitted in each of the transitions $|E\rangle \rightarrow |i\rangle$ (back to the state from which the system is being pumped by the laser field) and $|E\rangle \rightarrow |f\rangle$ (to a third level which can act as a sink for the atomic population). It is shown how the "confluence of coherences" is manifested in the number of photons emitted. The time development of the photon number is studied, and is shown to reflect the Rabi oscillations within the atom for large values of Fano's q parameter.

In Sec. IV we calculate the spectrum of photons ejected in the $|E\rangle \rightarrow |f\rangle$ transition. The spectrum is shown to exhibit a doublet, reflecting the ac Stark splitting of the autoionizing state. For small γ_i, the spectrum to $|f\rangle$ can serve as a probe of the coherences within the system. We show how recycling (nonzero γ_i) affects the Raman spectrum. In Sec. V we calculate the spectrum of photons ejected in the $|E\rangle \rightarrow |i\rangle$ transition. This spectrum exhibits a triplet reminiscent of the Mollow triplet,[7] but with no coherent (δ-function) component. The triplet shows the ac Stark splitting of both the autoionizing state and the initial state. It is shown how nonzero γ_f affects the spectrum.

In the Appendix we consider the decay of autoionizing states in the absence of a laser driving field, as studied by Armstrong, Theodosiou, and Wall.[5] We study the branching ratios of the two decay channels, and discuss the importance of the "virtual recombination" or coupling between the photon and electron channels. Our results allow for an interesting interpretation[8] of the Fano q value as the ratio of two decay rates.

II. PHOTOEMISSION SPECTRA—RELATION TO ATOMIC CORRELATIONS

In this section we first introduce the definition of photoemission spectra in terms of the correlation function of the atomic operators. This can be done by obtaining the Heisenberg equation of motion for $a_{ks}(t)$, and then formally integrating such an equation. Having obtained $a_{ks}(t)$, the mean number of photons in any mode can be calculated. This procedure is fairly standard.[9] Using the Hamiltonian Eq. I(2.5) [i.e., Eq. (2.5) from paper I], we obtain for the mean number of photons

$$N_{ks,i}(t)=|\vec{d}_{ai}\cdot\vec{u}_{ks}|^2\int_0^t dt_1\int_0^t dt_2\langle A_i^\dagger(t-t_1)A_i(t-t_2)\rangle e^{i(\omega_{ks}-\omega_l)(t_1-t_2)}, \tag{2.1}$$

$$N_{ks,f}(t)=|\vec{d}_{af}\cdot\vec{u}_{ks}|^2\int_0^t dt_1\int_0^t dt_2\langle A_f^\dagger(t-t_1)A_f(t-t_2)\rangle e^{i(\omega_{ks}-\omega_l+\omega/E_f)(t_1-t_2)}, \tag{2.2}$$

where the suffixes i and f represent the two channels open for spontaneous emission, and where the operators A_i and A_f are defined by I(2.12):

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

The total number of photons are given by

$$N_i(t)=\sum_{k,s}N_{k,si}(t)$$
$$=\gamma_i\int_0^t d\tau\langle A_i^\dagger(\tau)A_i(\tau)\rangle\ , \qquad (2.3)$$

$$N_f(t)=\gamma_f\int_0^t d\tau\langle A_f^\dagger(\tau)A_f(\tau)\rangle\ . \qquad (2.4)$$

On summing (2.1) and (2.2) over all the directions, we can obtain the number of photons at a given frequency

$$N_i(\delta,t)=\frac{\gamma_i}{2\pi}\int_0^t dt_1\int_0^t dt_2\langle A_i^\dagger(t_1)A_i(t_2)\rangle \times e^{-i\delta(t_1-t_2)}\ ,\quad \delta=\omega-\omega_l \qquad (2.5)$$

$$N_f(\delta_f,t)=\frac{\gamma_f}{2\pi}\int_0^t dt_1\int_0^t dt_2\langle A_f^\dagger(t_1)A_f(t_2)\rangle \times e^{-i\delta_f(t_1-t_2)}\ ,\quad \delta_f=\delta+E_f\ . \qquad (2.6)$$

The time integrals in (2.3)—(2.6) can be removed by using Laplace transforms and the structure of the two-time correlation functions. On using the quantum regression theorem, the two-time correlations can be written in the form

$$\langle A_i^\dagger(t_1)A_i(t_2)\rangle=\sum_\alpha F_i^{(\alpha)}(t_1-t_2)G_i^{(\alpha)}(t_2)\ . \qquad (2.7)$$

A simple analysis leads to the Laplace transforms of (2.3)—(2.7):

$$\hat N_i(z)=\frac{\gamma_i}{z}\mathscr{L}\langle A_i^\dagger A_i\rangle\ ,$$
$$\hat N_f(z)=\frac{\gamma_f}{z}\mathscr{L}\langle A_f^\dagger A_f\rangle \qquad (2.8)$$

(where carets and $\mathscr{L}$ denote Laplace transforms),

$$\hat N_i(\delta,z)=\frac{\gamma_i}{\pi}\mathrm{Re}\left[\frac{1}{z}\sum_\alpha F_i^{(\alpha)}(z+i\delta)G_i^{(\alpha)}(z)\right]\ , \qquad (2.9)$$

$$\hat N_f(\delta_f,z)=\frac{\gamma_f}{\pi}\mathrm{Re}\left[\frac{1}{z}\sum_\alpha F_f^{(\alpha)}(z+i\delta_f)G_f^{(\alpha)}(z)\right]\ . \qquad (2.10)$$

The use of Laplace transforms is especially attractive, as then the expressions for steady-state quantities follow,

$$N_i=\lim_{t\to\infty}N_i(t)=\lim_{z\to 0}\gamma_i\mathscr{L}\langle A_i^\dagger A_i\rangle\ , \qquad (2.11)$$

$$N_f=\lim_{z\to 0}\gamma_f\mathscr{L}\langle A_f^\dagger A_f\rangle\ , \qquad (2.12)$$

$$N_i(\delta)=\frac{\gamma_i}{\pi}\mathrm{Re}\left[\sum_\alpha F_i^{(\alpha)}(0+i\delta)G_i^{(\alpha)}(0)\right]\ , \qquad (2.13)$$

$$N_f(\delta_f)=\frac{\gamma_f}{\pi}\mathrm{Re}\left[\sum_\alpha F_f^{(\alpha)}(0+i\delta_f)G_f^{(\alpha)}(0)\right]\ , \qquad (2.14)$$

where we have assumed that the limit as $z\to 0^+$ of all quantities like $F^{(\alpha)}$ exist. It will be seen in Secs. III—V that all these limits indeed do exist, implying that $dN_i(\delta)/dt=dN_f(\delta_f)/dt=0$. Thus the spectra discussed in Secs. IV and V are different from conventional spectra as the conventional spectra deal with the rate of change of photons in a given frequency interval. In order to have nonvanishing values of the conventional spectra for the autoionization system, it would be necessary to continuously pump the ground state.

Another point worth noticing about autoionizing states is that in the steady state $\langle A_i\rangle=\langle A_f\rangle=0$. To see this consider, for example, $\langle A_i\rangle$:

$$\langle\hat A_i\rangle=\int dE\,B_{Ea}\hat\rho_{Ei}(z)$$
$$=\int dE\,B_{Ea}\frac{\hat\sigma_{Ei}(z)}{1-\hat g_\sigma(z)}\equiv\left[\frac{2}{\gamma_i}\right]^{1/2}D(z) \qquad (2.15)$$

and hence in the steady state

$$\langle A_i(\infty)\rangle=\int dE\,B_{Ea}\lim_{z\to 0}z\hat\sigma_{Ei}(z)/[1-\hat g_\sigma(0)]$$
$$=[1-\hat g_\sigma(0)]^{-1}\lim_{t\to\infty}\int dE\,B_{Ea}\psi_E(t)\psi_i^*(t)$$
$$=[1-\hat g_\sigma(0)]^{-1}\left[\frac{2}{\gamma_i}\right]^{1/2}\lim_{t\to\infty}\psi_i^*(t)\chi_2(t)\ , \qquad (2.16)$$

which is zero since both ψ_i and χ_2 decay with time. In other words, the induced dipole moment is zero, which implies that there will be no coherent scattering [i.e., no $\delta(\omega-\omega_l)$-like contribution].

III. TOTAL NUMBER OF PHOTONS EMITTED IN THE DECAY OF AUTOIONIZING STATES

In this section we study the features of the total number of photons that can be emitted in each channel of spontaneous emission. It will be seen that these quantities carry very interesting information regarding the features of the autoionizing states. From the definition I(2.12) and the orthogonality of states, we find that (2.8) can be reduced to

$$\hat N_i(z)=\frac{\gamma_i}{z}\int dE_1\int dE_2B_{E_1a}^*B_{E_2a}\hat\rho_{E_2E_1}(z)\ ,$$
$$\hat N_f(z)=\frac{\gamma_f}{z}\int dE_1\int dE_2C_{E_1a}^*C_{E_2a}\hat\rho_{E_2E_1}(z)\ , \qquad (3.1)$$

which on using the solution I(3.22) with $\nu=0$ and the initial condition $\rho(t=0)=|i\rangle\langle i|$ reduces to

$$\hat N_i(z)=\frac{\gamma_i}{z}\int dE_1\int dE_2B_{E_1a}^*B_{E_2a}\frac{\hat\sigma_{E_2E_1}(z)}{1-\hat g_\sigma(z)}\ ,$$
$$\hat N_f(z)=\frac{\gamma_f}{z}\int dE_1\int dE_2C_{E_1a}^*C_{E_2a}\frac{\hat\sigma_{E_2E_1}(z)}{1-\hat g_\sigma(z)}\ . \qquad (3.2)$$

On recalling $\sigma_{E_2E_1}(t)=\psi_{E_2}(t)\psi^*_{E_1}(t)$ and using I(3.32), we can reduce the double integrals in (3.2) to

$$\hat{N}_i(z)=\frac{2}{z}\frac{\int_0^\infty e^{-zt}|\chi_2(t)|^2dt}{1-\hat{g}_\sigma}\;,$$
$$\hat{N}_f(z)=\frac{2}{z}\frac{\int_0^\infty e^{-zt}|\chi_3(t)|^2}{1-\hat{g}_\sigma}\;. \quad (3.3)$$

The expressions for the χ's are

$$\hat{\chi}_i=-i[\underline{m}(\mathbb{1}+\underline{m})^{-1}]_{i1}\;. \quad (3.4)$$

Thus

$$\hat{\chi}_2=i[(\mathbb{1}+\underline{m})^{-1}]_{21}\;,$$
$$\hat{\chi}_3=i[(\mathbb{1}+\underline{m})^{-1}]_{31}\;,$$

with the matrix $\underline{m}$ defined by I(3.37). An explicit expression for $\hat{\chi}_2$ is given in Eq. I(6.11). $\hat{\chi}_3(z)$ can be obtained by writing

$$\hat{\chi}_3(z)=i[(\mathbb{1}+\underline{m})^{-1}]_{31}$$
$$=\frac{i}{z\det(\mathbb{1}+\underline{m})}[m_{12}m_{23}-(1+m_{22})m_{13}] \quad (3.5)$$

and noting from I(3.37) that

$$m_{12}(z)m_{23}(z)=m_{22}(z)m_{13}(z)\;. \quad (3.6)$$

It follows that

$$\hat{\chi}_3(z)=\frac{-im_{13}(z)}{z\det[\mathbb{1}+\underline{m}(z)]}\;, \quad (3.7)$$

and in terms of ϵ

$$\hat{\chi}_3=\sqrt{2\Omega\gamma_f}\frac{1+(\epsilon+q_i)/q_f+iq_i}{\psi(\epsilon-\epsilon_+)(\epsilon-\epsilon_-)}\;. \quad (3.8)$$

The expressions in the steady state are even simpler:

$$N_i=\frac{2\int_0^\infty|\chi_2(t)|^2dt}{1-2\int_0^\infty|\chi_2(t)|^2dt}=\frac{g_\sigma(0)}{1-g_\sigma(0)}\;, \quad (3.9)$$

$$N_f=\frac{2\int_0^\infty|\chi_3(t)|^2dt}{1-2\int_0^\infty|\chi_2(t)|^2dt}=\frac{2\int_0^\infty|\chi_3(t)|^2}{1-g_\sigma(0)}\;. \quad (3.10)$$

It is instructive to note that for $q_f=q_i$, $\hat{\chi}_2$ and $\hat{\chi}_3$ are identical under a $\gamma_i \leftrightarrow \gamma_f$ interchange. If $\gamma_i=0$ so that no recycling can occur, then the probability of the system's ejecting a photon is

$$P_{\rm ph}=2\int_0^\infty|\chi_3(t)|^2dt\;.$$

However, if $\gamma_f=0$ but $\gamma_i\neq0$, so that the system can recycle, then the expected number of photons is increased to

$$N_i=\frac{P_{\rm ph}}{1-P_{\rm ph}}\;,$$

where $P_{\rm ph}$ can be interpreted as the "probability per cycle of ejecting a photon."

We have evaluated the quantities N_i, N_f for various values of the system parameters and the results are shown in Figs. 1–4. Figure 1 gives N_i as a function of field strength Ω for $\gamma_f=0$ and the laser tuned close to the Fano minimum ($q_i=1$, $\alpha=-0.9$). The dashed curves show the corresponding results when the "virtual recombination" is ignored and show that this continuum-continuum coupling can only be neglected when $\gamma_i \ll \Gamma$. The "confluence of coherences" is shown by the large increase in N_i near $\Omega=0.10$ for small γ_i. Near confluence the electron channel is nearly closed and the system ejects many photons before ejecting an electron. However, for

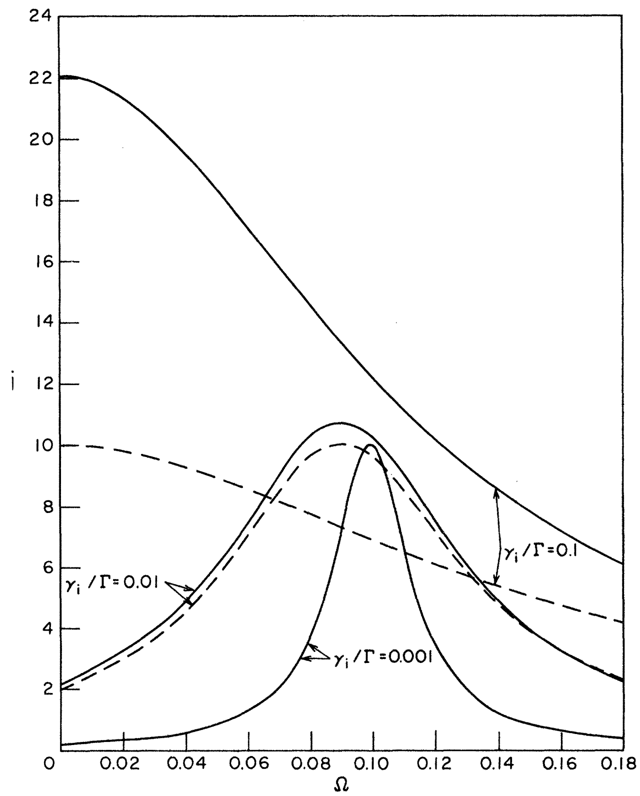

FIG. 1. Number of photons N_i vs Ω for $\gamma_f=0$, $q_i=1$, $\alpha=-0.9$, and for various values of γ_i. The dashed curves neglect the virtual recombination.

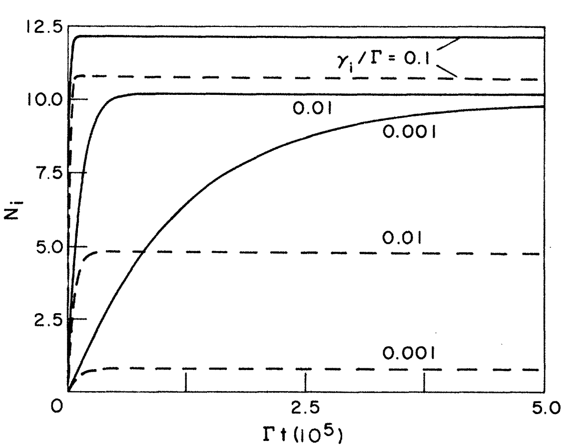

FIG. 2. Time development of N_i for $q_i=1$, $\alpha=-0.9$, $\Omega=0.1$, and various γ_i. The solid curves have $\gamma_f=0$, the dashed curves $\gamma_f/\Gamma=0.001$.

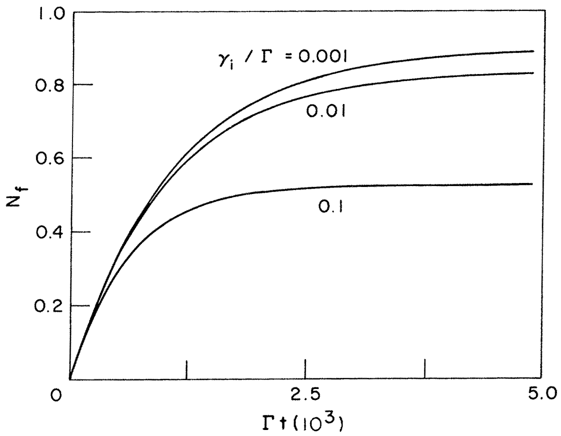

FIG. 3. Time development of N_f for the conditions of the dashed curves of Fig. 2 except $\gamma_f/\Gamma=0.01$.

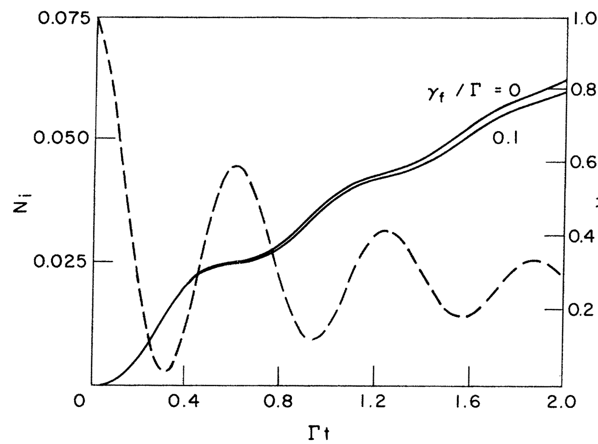

FIG. 4. Time development of N_i for $q_i=10$, $\alpha=0$, $\Omega=1.0$, $\gamma_i/\Gamma=0.1$, and $\gamma_f/\Gamma=0$ and 0.1. The dashed curve gives the population of state $|i\rangle P_{ii}$ for $\gamma_f=0$. The scale at the left-hand side of the graph is for N_i, and the scale at the right for P_{ii}.

small γ_i, a long time will be required for these photons to be emitted, and other sources of incoherence could be important. Figure 2 shows the time development of N_i for $\Omega=1+(\alpha/q)=0.10$ for the conditions of Fig. 1 (with virtual recombination included), and shows the very long-time scales involved. The dashed curves show the time development for $\gamma_f/\Gamma=0.001$. Figure 3 depicts the time development of N_f for the conditions of the dashed curves of Fig. 2 except $\gamma_f/\Gamma=0.01$.

Finally, in Fig. 4 we superpose the time development of N_i near confluence for large q_i with the population of $|i\rangle$, P_{ii}, from paper I. The Rabi oscillations between $|i\rangle$ and $|a\rangle$ are seen to be reflected by the staircase form of $N_i(t)$.

IV. SPECTRAL CHARACTERISTICS OF THE PHOTONS EMITTED IN TRANSITION $|E\rangle \rightarrow |f\rangle$

We first consider the spectra of photons emitted in the transition $|E\rangle \rightarrow |f\rangle$, which we will refer to as the Raman transition. The corresponding spectra may be called Raman spectra. Note that the pump field on the transition $|i\rangle \leftrightarrow |E\rangle$ can be of arbitrary intensity. To find the spectra we need the correlation function $\langle A_f^\dagger(t_1) A_f(t_2)\rangle$, which can be evaluated by using the solution of Eqs. I(3.27)–I(3.29) and the quantum regression theorem.

Using the definition of the operator $A_f^\dagger$, we have

$$\langle A_f^\dagger(t_1)\rangle = \int dE\, C_{Ea}^* \rho_{fE}(t_1)$$

$$= \int dE\, C_{Ea}^* \Phi_E^* \quad (\nu=0)\,, \tag{4.1}$$

where I(3.27) has been used. As mentioned in I, the time dependence of Φ_E can be obtained from the solution for the ψ_E's. Thus Eq. I(3.35) can be used:

$$\left[\frac{\gamma_f}{2}\right]^{1/2} \int dE\, C_{Ea} \hat{\Phi}_E \equiv -i\{[(\mathbb{1}+\underline{m})\underline{m}]^{-1}\}_{31}\Phi_i(0) + \sum_j [(\mathbb{1}+\underline{m})^{-1}]_{3j} \int dE \frac{L_j(E)\Phi_E(0)}{z+i\Delta_E}\,, \tag{4.2}$$

where the L's are defined by I(3.31). Equation (4.2) written in operator language implies that

$$\left[\frac{\gamma_f}{2}\right]^{1/2} \mathscr{L}\langle A_f^\dagger\rangle = +i\{[(\mathbb{1}+\underline{m})\underline{m}]^{-1}\}_{31}^* \langle\, |i\rangle\langle f|\,\rangle + \sum_j [(\mathbb{1}+\underline{m})^{-1}]_{3j}^* \int dE \frac{L_j^*(E)}{z-i\Delta_E} \langle\, |E\rangle\langle f|\,\rangle\,, \tag{4.3}$$

where complex conjugation has also been done [note z remains z as it is the Laplace variable: The convention we will use here is that, for example, $\hat{\psi}^*(z)$ represents the Laplace transform of $\psi^*(t)$, and hence z is not changed in taking the complex conjugate]. The regression theorem can now be applied:

$$\begin{aligned} \langle\, |i\rangle\langle f| A_f\rangle &= \int dE_1 C_{E_1 a} \langle\, |i\rangle\langle E_1|\,\rangle = \int dE_1 C_{E_1 a} \rho_{E_1 i}\,, \\ \langle\, |E\rangle\langle f| A_f\rangle &= \int dE_1 C_{E_1 a} \rho_{E_1 E}\,. \end{aligned} \tag{4.4}$$

On combining (4.3) and (4.4) we obtain

$$\frac{\gamma_f}{2}\int_0^\infty \int_0^\infty \langle A_f^\dagger(t_1)A_f(t_2)\rangle e^{-z(t_1-t_2)}d(t_1-t_2)dt_2 e^{-z't_2}$$
$$\equiv f(z,z')=i\{[(\mathbb{1}+\underline{m})\underline{m}]^{-1}\}^*_{31}\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\hat{\rho}_{E_1i}(z')+\sum_j[(\mathbb{1}+\underline{m})^{-1}]^*_{3j}\int dE\frac{L_j^*(E)}{(z-i\Delta_E)}\left[\frac{\gamma_f}{2}\right]^{1/2}$$
$$\times\int dE_1 C_{E_1a}\hat{\rho}_{E_1E}(z')\,, \quad (4.5)$$

which on using I(3.22) reduces to

$$f(z,z')=-i[(\mathbb{1}+\underline{m}^*)^{-1}]_{31}[1-\hat{g}_\sigma(z')]^{-1}\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\hat{\sigma}_{E_1i}(z')$$
$$+\sum_j[(\mathbb{1}+\underline{m}^*)^{-1}]_{3j}\int dE\frac{L_j^*(E)}{z-i\Delta_E}\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\hat{\sigma}_{E_1E}(z')[1-\hat{g}_\sigma(z')]^{-1}\,. \quad (4.6)$$

The quantities that appear in (4.6) can be simplified by using the factorization property of $\hat{\sigma}$ in the time domain:

$$\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\sigma_{E_1i}(t)=\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\psi_{E_1}(t)\psi_i^*(t)$$
$$=\chi_3(t)\psi_i^*(t)\,,$$
$$\left[\frac{\gamma_f}{2}\right]^{1/2}\int dE_1 C_{E_1a}\sigma_{E_1E}(t)=\chi_3(t)\psi_E^*(t)\,. \quad (4.7)$$

Though now we have the complete form for $f(z,z')$, in what follows we concentrate only on the steady-state spectra and hence we can let $z'\to 0$:

$$f(z,0)=-i[(\mathbb{1}+\underline{m}^*)^{-1}]_{31}[1-\hat{g}_\sigma(0)]^{-1}\int_0^\infty dt\,\chi_3(t)\psi_i^*(t)$$
$$+\sum_j[(\mathbb{1}+\underline{m}^*)^{-1}]_{3j}[1-\hat{g}_\sigma(0)]^{-1}\int dE\frac{L_j^*(E)}{z-i\Delta_E}\int_0^\infty dt\,\chi_3(t)\psi_E^*(t)\,. \quad (4.8)$$

Let us denote the second term in (4.8) by $[1-\hat{g}_\sigma(0)]^{-1}\bar{f}(z,0)$. Thus

$$\bar{f}^*(z,0)=\sum_j[(\mathbb{1}+\underline{m})^{-1}]_{3j}\int dE\frac{L_j(E)}{z+i\Delta_E}$$
$$\times\sum_\alpha \chi_3^{(\alpha)*}\hat{\psi}_E(-z_\alpha^*)\,, \quad (4.9)$$

where we have written $\chi_3(t)$ as

$$\chi_3(t)=\sum_{\alpha=1}^{2}\chi_3^{(\alpha)}e^{z_\alpha t}\,. \quad (4.10)$$

To do the energy integral in (4.9), consider, for any $\mathfrak{z}$,

$$K(z,\mathfrak{z})=\int dE\frac{L_j(E)}{z+i\Delta_E}\hat{\psi}_E(\mathfrak{z})\,,$$

which on using I(3.36) reduces to

$$K(z,\mathfrak{z})=-i\int dE\frac{L_j(E)}{z+i\Delta_E}$$
$$\times\sum_i K_i(E,\mathfrak{z})\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{i1}\,, \quad (4.11)$$

remembering that K_i and m depend on the Laplace variable. On using the definition of the m matrices, relation (4.11) can be simplified to

$$K(z,\mathfrak{z})=-\frac{i}{\mathfrak{z}-z}\left[\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{11}\left[\frac{z}{\mathfrak{z}}m_{j1}(z)-m_{j1}(\mathfrak{z})\right]+\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{21}[m_{j2}(z)-m_{j2}(\mathfrak{z})]\right.$$
$$\left.+\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{31}[m_{j3}(z)-m_{j3}(\mathfrak{z})]\right]\,. \quad (4.12)$$

On substituting (4.12) in (4.9), we find that

$$\bar{f}^*(z,0)=\sum_\alpha \chi_3^{(\alpha)*}[\hat{h}_{31}(z,\mathfrak{z})]_{\mathfrak{z}=-z_\alpha^*}\,, \quad (4.13)$$

where

$$\hat{h}_{31}(z,\mathfrak{z}) = -\frac{i}{\mathfrak{z}-z}\left[\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{31} - \{[1+m(z)]^{-1}\}_{31} - \left[\frac{z-\mathfrak{z}}{\mathfrak{z}}\right]\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{11}\{[\mathbb{1}+\underline{m}(z)]^{-1}\}_{31}\right] . \quad (4.14a)$$

Using Eqs. (3.35), $\hat{h}$ can be rewritten as

$$\hat{h}_{31}(z,\mathfrak{z}) = \frac{\hat{\chi}_3(\mathfrak{z})-\hat{\chi}_3(z)}{z-\mathfrak{z}} - \hat{\psi}_i(\mathfrak{z})\hat{\chi}_3(z) . \quad (4.14b)$$

The spectrum of the photons emitted in the transition $|E\rangle \rightarrow |f\rangle$ is obtained by substituting (4.5) in (2.14):

$$N_f(\delta_f) = \frac{2}{\pi}\text{Re}[f(i\delta_f,0)] , \quad \delta_f = \omega - \omega_l + E_f , \quad (4.15)$$

$$f(z,0) = [1-g_\sigma(0)]^{-1}\sum_\alpha \chi_3^{(\alpha)}(-i\{[\mathbb{1}+\underline{m}^*(z)]^{-1}\}_{31}\hat{\psi}_i^*(-z_\alpha) + \hat{h}_{31}^*(z,-z_\alpha)) . \quad (4.16)$$

($\hat{h}_{31}$ is a function of two Laplace transform variables and neither is changed in taking the complex conjugate.) Combining (4.14b) and (4.16) one obtains

$$f(z,0) = \frac{1}{1-g_\sigma(0)} \times \sum_\alpha \frac{\chi_3^{(\alpha)}}{z+z_\alpha}[\hat{\chi}_3^*(-z_\alpha) - \hat{\chi}_3^*(z)] . \quad (4.17)$$

On using (4.10) this simplifies to

$$f(z,0) = \frac{1}{1-g_\sigma(0)}\sum_{\alpha,\beta}\frac{\chi_3^{(\alpha)}\chi_3^{(\beta)*}}{(-z_\alpha - z_\beta^*)(z-z_\beta^*)} . \quad (4.18)$$

Finally, a very simple expression for $N_f(\delta_f)$ can be obtained by adding (4.17) and its complex conjugate. On simplifying one finds that

$$N_f(\delta_f) = \frac{1}{\pi[1-g_\sigma(0)]}|\hat{\chi}_3^*(i\delta_f)|^2 = \frac{1}{\pi[1-g_\sigma(0)]}|\hat{\chi}_3(-i\delta_f)|^2 \quad (4.19)$$

which can be written explicitly as

$$N_f(\delta_f) = \frac{1}{1-g_\sigma(0)} \times \left|\left[\frac{2\Omega\gamma_f}{\pi\Gamma^2}\right]^{1/2}\frac{q_i - i[1+(\delta_0+q_i)/q_f]}{\psi(\delta_0-\epsilon_+)(\delta_0-\epsilon_-)}\right|^2 , \quad (4.20)$$

$$-i\delta_f = -\tfrac{1}{2}i\Gamma(\delta_0-\alpha)$$

or

$$\delta_0 = \frac{\omega - E_a + E_f}{\Gamma/2} . \quad (4.21)$$

The modulus squared in (4.20) is identical in form to the result[6] when $\gamma_i = 0$. Thus γ_i affects the spectrum only by altering the values of ψ and $\epsilon_\pm$ and through the scaling factor $[1-g_\sigma(0)]^{-1}$.

It is clear from (4.20) that the Raman spectrum is an asymmetric doublet. Figure 5 shows graphs of $N_f(\delta_f)$ for $\gamma_i = 0$, $q_i = 1$, $\alpha = 1$, and various Ω and γ_f. Since we are close to confluence, we show only one component of the doublet. The solid curves have $q_f = 1$ and the dashed curves show the limit $q_f \rightarrow \infty$ (equivalent to neglecting the virtual recombination).[10] Figure 6 shows how increasing γ_i smooths the sharp feature of the spectra.

V. SPECTRAL CHARACTERISTICS OF THE PHOTONS EMITTED IN THE TRANSITION $|E\rangle \rightarrow |i\rangle$

This section is devoted to the study of the characteristics of the photons emitted in the transition $|E\rangle \rightarrow |i\rangle$.

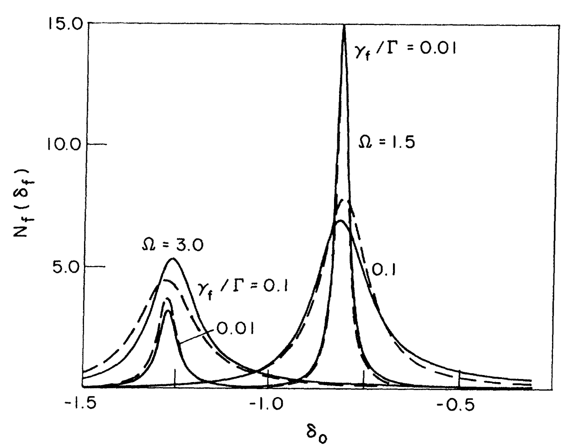

FIG. 5. Raman spectrum in units of $1/\pi\Gamma$ graphed vs $\delta_0 = (2/\Gamma)\delta_f + \alpha$ for $\alpha = q_i = 1$, $\gamma_i = 0$, $q_f = 1$ (solid curves), and $q_f \rightarrow \infty$ (dashed curves). Confluence occurs at $\Omega = 2.0$ and the Fano minimum at $\delta_0 = -1$.

These spectral features are expected to be different from those discussed in Sec. IV because the $|E\rangle \leftrightarrow |i\rangle$ transition is strongly driven. To obtain the spectra, we calculate the two-time correlation function $\langle A_i^\dagger(t_1)A_i(t_2)\rangle$ directly using the equations of motion for such two-time correlations. Equations I(3.1)—I(3.6) can be written as expectation value equations since $\rho_{\alpha\beta}=\langle\,|\beta\rangle\langle\alpha|\,\rangle$. Defining a number of two-time correlations by

$$\Gamma_{E_2E_1}(\tau,t)=\langle(\,|E_2\rangle\langle E_1|\,)_{t+\tau}A_i(t)\rangle\ ,$$
$$\Gamma_{iE_1}(\tau,t)=\langle(\,|i\rangle\langle E_1|\,)_{t+\tau}A_i(t)\rangle\ , \tag{5.1}$$
$$\Gamma_{ii}(\tau,t)=\langle(\,|i\rangle\langle i|\,)_{t+\tau}A_i(t)\rangle\ ,$$

we obtain equations of motion for these using I(3.1)—I(3.6) for $\nu=0$ and the quantum regression theorem:

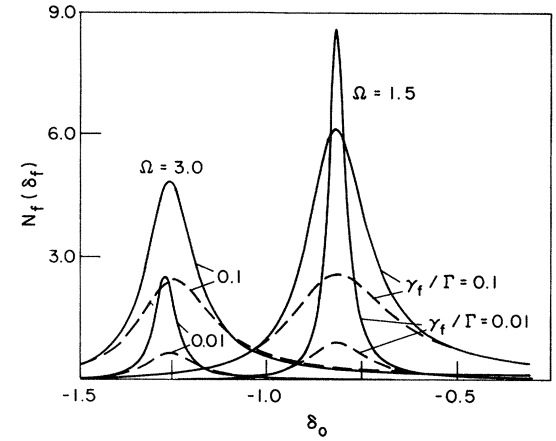

FIG. 6. Spectra to $|f\rangle$ for the same conditions as the solid curves in Fig. 5 except for nonzero γ_i. $\gamma_i/\Gamma=0.01$ for the solid curves, 0.1 for dashed curves.

$$\frac{\partial}{\partial\tau}\Gamma_{E_2E_1}=-i(E_1-E_2)\Gamma_{E_2E_1}-iv_{E_1i}\Gamma_{E_2i}+iv_{E_2i}^*\Gamma_{iE_1}-\frac{\gamma_i}{2}\int dE\,B_{E_1a}^*B_{Ea}\Gamma_{E_2E}$$
$$-\frac{\gamma_f}{2}\int dE\,C_{E_1a}^*C_{Ea}\Gamma_{E_2E}-\frac{\gamma_i}{2}\int dE\,B_{E_2a}B_{Ea}^*\Gamma_{EE_1}-\frac{\gamma_f}{2}\int dE\,C_{E_2a}C_{Ea}^*\Gamma_{EE_1}\ , \tag{5.2}$$

$$\frac{\partial}{\partial\tau}\Gamma_{iE_1}=-i\Delta_{E_1}\Gamma_{iE_1}-i\left[v_{E_1i}\Gamma_{ii}-\int dE\,v_{Ei}\Gamma_{EE_1}\right]$$
$$-\frac{\gamma_i}{2}\int dE\,B_{E_1a}^*B_{Ea}\Gamma_{iE}-\frac{\gamma_f}{2}\int dE\,C_{E_1a}^*C_{Ea}\Gamma_{iE}\ , \tag{5.3}$$

$$\frac{\partial}{\partial\tau}\Gamma_{ii}=-i\int dE\,v_{Ei}^*\Gamma_{iE}+i\int dE\,v_{Ei}\Gamma_{Ei}+\frac{\gamma_i}{2}\int dE_1\int dE\,B_{E_1a}^*B_{Ea}\Gamma_{E_1E}$$
$$+\frac{\gamma_i}{2}\int dE_1\int dE\,B_{E_1a}B_{Ea}^*\Gamma_{EE_1}\ . \tag{5.4}$$

The initial conditions are

$$\Gamma_{E_2E_1}(0,t)=\Gamma_{E_2E_1}(t)=\langle\,|E_2\rangle\langle E_1|A_i(t)\rangle=0\ ,$$
$$\Gamma_{iE_1}(t)=\langle\,|i\rangle\langle E_1|A_i(t)\rangle=0\ ,$$
$$\Gamma_{ii}(t)=\langle\,|i\rangle\langle i|A_i(t)\rangle=\langle A_i(t)\rangle=\int dE\,B_{Ea}\rho_{Ei}(t)\ , \tag{5.5}$$
$$\Gamma_{E_1i}(t)=\langle\,|E_1\rangle\langle i|A_i(t)\rangle=\int dE\,B_{Ea}\langle\,|E_1\rangle\langle E|\,\rangle=\int dE\,B_{Ea}\rho_{EE_1}(t)\ .$$

The initial conditions involve the expectation values at one time, the solutions to which are given in paper I, Sec. III. We now introduce a set of auxiliary quantities $R_{E_2E_1}, R_{iE_1}, R_{E_1i}$ such that these satisfy (5.2) and (5.3) with $\Gamma\to R$ and

$$\frac{\partial}{\partial\tau}R_{ii}=-i\int dE\,v_{Ei}^*R_{iE}+i\int dE\,v_{Ei}R_{Ei}\ . \tag{5.6}$$

For the time being we leave the initial conditions on R quite arbitrary. We have constructed the R matrix such that the equations are factorizable:

$$R_{\alpha\beta}(t)=U_\alpha^*(t)U_\beta(t) \tag{5.7}$$

with

$$\dot U_i=-i\int dE_1 v_{E_1i}^*U_{E_1}\ ,$$
$$\dot U_{E_1}=-i\Delta_{E_1}U_{E_1}-iv_{E_1i}U_i-\frac{\gamma_i}{2}\int dE\,B_{E_1a}^*B_{Ea}U_E-\frac{\gamma_f}{2}\int dE\,C_{E_1a}^*C_{Ea}U_E\ , \tag{5.8}$$

which are identical in form to I(3.9) and I(3.10). The functions U are thus known in terms of matrices $\underline{m}$:

$$\hat U_E=-i\sum_i K_i(E)[(\mathbb{1}+\underline{m})^{-1}]_{i1}U_i(0)-\sum_{i,j}K_i(E)[(\mathbb{1}+\underline{m})^{-1}]_{ij}\int dE'\frac{L_j(E')U_{E'}(0)}{z+i\Delta_{E'}}+\frac{U_E(0)}{z+i\Delta_E}\ , \tag{5.9}$$

$$\hat{V}_i \equiv \int dE\, L_i(E)\hat{U}_E = \sum_j [(\mathbb{1}+\underline{m})^{-1}]_{ij} \left[-iU_i(0)m_{j1} + \int dE_1 \frac{L_j(E_1)U_{E_1}(0)}{z+i\Delta_{E_1}} \right] , \quad (5.10)$$

$$\hat{U}_i = \frac{U_i(0)}{z} - \frac{i\hat{V}_1}{z} . \quad (5.11)$$

We now use (5.9)–(5.11) in (5.7) and replace quantities like $U^*_\alpha(0)U_\beta(0)$ by $R_{\alpha\beta}(0)$. Formally we will have

$$\dot{R} = LR , \quad R(t) = e^{Lt}R(0) . \quad (5.12)$$

We are now in a position to construct the matrix elements of e^{Lt}. It turns out that we only need R_{ii} and $\int dE_1 R_{E_1 i} B^*_{E_1 a}$:

$$R_{ii}(t) = U_i^*(t)U_i(t) ,$$
$$\int dE_1 R_{E_1 i} B^*_{E_1 a} = \left[\frac{2}{\gamma_i} \right]^{1/2} V_2^*(t)U_i(t) . \quad (5.13)$$

If we now write

$$\hat{V}_2 = \hat{f}_{2i} U_i(0) + \int dE_1 U_{E_1}(0)\hat{f}_{2E_1} ,$$
$$\hat{U}_i = \hat{f}_{ii} U_i(0) + \int dE_1 U_{E_1}(0)\hat{f}_{iE_1} , \quad (5.14)$$

then the matrix elements of the operator e^{Lt} follow:

$$(e^{Lt})_{ii,ii} = f^*_{ii}(t) f_{ii}(t) ,$$
$$(e^{Lt})_{ii,iE_1} = f^*_{ii}(t) f_{iE_1}(t) ,$$
$$(e^{Lt})_{ii,E_1 i} = f_{ii}(t) f^*_{iE_1}(t) ,$$
$$(e^{Lt})_{ii,E_1E_2} = f^*_{iE_1}(t) f_{iE_2}(t) ,$$
$$\int dE_1 B^*_{E_1 a}(e^{Lt})_{E_1 i,ii} = \left[\frac{2}{\gamma_i} \right]^{1/2} f^*_{2i}(t) f_{ii}(t) ,$$
$$\int dE_1 B^*_{E_1 a}(e^{Lt})_{E_1 i,iE_1'} = \left[\frac{2}{\gamma_i} \right]^{1/2} f^*_{2i}(t) f_{iE_1'}(t) ,$$
$$\int dE_1 B^*_{E_1 a}(e^{Lt})_{E_1 i,E_1' i} = \left[\frac{2}{\gamma_i} \right]^{1/2} f^*_{2E_1'}(t) f_{ii}(t) ,$$
$$\int dE_1 B^*_{E_1 a}(e^{Lt})_{E_1 i,E_1'E_2'} = \left[\frac{2}{\gamma_i} \right]^{1/2} f^*_{2E_1'}(t) f_{iE_2'}(t) . \quad (5.15)$$

It should be remembered that $f_{2i}(t)=\chi_2(t)$, $f_{ii}(t)=\psi_i(t)$ if the equations for ψ's are solved subject to the initial condition $\psi_E(0)=0$, $\psi_i(0)=1$.

Having obtained the operator e^{Lt}, we are now in a position to solve Eqs. (5.2)–(5.4) which can be written as

$$\dot{\Gamma} = L\Gamma + |i\rangle\langle i|\, g(\tau,t) , \quad (5.16)$$

$$g(\tau,t) = \frac{\gamma_i}{2} \int dE_1 \int dE\, B^*_{E_1 a} B_{Ea} \Gamma_{E_1 E} + \frac{\gamma_i}{2} \int dE_1 \int dE\, B_{E_1 a} B^*_{Ea} \Gamma_{EE_1} . \quad (5.17)$$

On taking the Laplace transform with respect to τ, the solution of (5.16) is

$$\hat{\Gamma}(z,t) = (z-L)^{-1}\Gamma(t) + \hat{g}(z,t)(z-L)^{-1}|i\rangle\langle i| . \quad (5.18)$$

As noted above, the construction of the R matrix gives us the operator $(z-I)^{-1}$. The last term in (5.18) is just the R matrix under the initial condition $U_E=0$, $U_i=1$. Let us call this solution $\underline{S}$:

$$\hat{S}(z) = (z-L)^{-1}|i\rangle\langle i| \implies$$
$$S_{\alpha\beta}(t) = U^*_\alpha(t)U_\beta(t) , \quad U_E(0)=0 , \quad U_i(0)=1 . \quad (5.19)$$

In terms of $\hat{S}$ and $\hat{T}$ matrices (which are now known)

$$\hat{T} = (z-L)^{-1}\Gamma(t) , \quad (5.20)$$

we have

$$\hat{\Gamma}(z,t) = \hat{T}(z,t) + \hat{g}(z,t)\hat{S}(z) . \quad (5.21)$$

The unknown $\hat{g}$ can be eliminated by introducing

$$\hat{g}_T(z,t) = \frac{\gamma_i}{2} \int dE_1 \int dE [B^*_{E_1 a} B_{Ea} \hat{T}_{E_1 E}(z,t) + B_{E_1 a} B^*_{Ea} \hat{T}_{EE_1}(z,t)] ,$$
$$\hat{g}_S(z) = \frac{\gamma_i}{2} \int dE_1 \int dE [B^*_{E_1 a} B_{Ea} \hat{S}_{E_1 E}(z) + B_{E_1 a} B^*_{Ea} \hat{S}_{EE_1}(z)] , \quad (5.22)$$

with the final result:

$$\hat{\Gamma}(z,t) = \hat{T}(z,t) + \frac{\hat{g}_T(z,t)}{1-\hat{g}_S(z)} \hat{S}(z) . \quad (5.23)$$

It should be remembered that the matrix $\hat{S}$ does not depend on t. Moreover, the way the matrix $\hat{S}$ has been constructed, it is related to $\hat{\sigma}$ [Eq. I(3.16)] by

$$S_{\alpha\beta}(t) = \sigma_{\beta\alpha}(t) . \quad (5.24)$$

For the calculation of the spectrum we need $\langle A_i^\dagger(t+\tau)A_i(t)\rangle$

$$\langle A_i^\dagger(t+\tau)A_i(t)\rangle = \int dE\, B^*_{Ea} \langle (|E\rangle\langle i|)_{t+\tau} A_i(t)\rangle = \int dE\, B^*_{Ea} \Gamma_{Ei}(\tau,t) \quad (5.25)$$

and hence

$$\int_0^\infty \langle A_i^\dagger(t+\tau)A_i(t)\rangle e^{-z\tau}d\tau \equiv \hat{F}(z,t)$$

$$= \int dE\, B_{Ea}^* \hat{T}_{Ei}(z,t) + \hat{g}_T(z,t)\frac{\int dE\, B_{Ea}^* \hat{S}_{Ei}(z)}{1-\hat{g}_S(z)} \tag{5.26}$$

$$= \int dE\, B_{Ea}^* \hat{T}_{Ei}(z,t) + \left[\frac{2}{\gamma_i}\right]^{1/2} \hat{g}_T(z,t)\hat{D}^*(z) , \tag{5.27}$$

$$\hat{D}^*(z) \equiv \frac{\int_0^\infty e^{-zt}\chi_2^*(t)\psi_i(t)dt}{1-2\int_0^\infty e^{-zt}|\chi_2(t)|^2 dt} , \tag{5.28}$$

where (5.24) and the definition of χ [Eq. I(3.32)] have been used. We will now simplify the other terms in (5.26). We take the matrix element of (5.20) and use (5.5):

$$\hat{T}_{\alpha\beta}(z,t) = (z-L)^{-1}_{\alpha\beta,ii}\int dE\, B_{Ea}\rho_{Ei}(t) + \int dE_2 (z-L)^{-1}_{\alpha\beta,E_2 i}\int dE\, B_{Ea}\rho_{EE_2}(t) . \tag{5.29}$$

Thus the first term in (5.27) reduces to

$$\hat{X}(z,t) \equiv \int dE_1 B_{E_1 a}^* \hat{T}_{E_1 i}(z,t)$$

$$\equiv \int dE_1 \int dE\, B_{E_1 a}^* B_{Ea}\left[(z-L)^{-1}_{E_1 i,ii}\rho_{Ei}(t) + \int dE_2 (z-L)^{-1}_{E_1 i,E_2 i}\rho_{EE_2}(t)\right] . \tag{5.30}$$

Using the matrix elements given by (5.15) and the definition of $D(t)$ [Eq. (2.15)] (5.30) can be reduced to

$$\hat{X}(z,t) \equiv \left[\frac{2}{\gamma_i}\right]\left[\int_0^\infty d\tau\, e^{-z\tau}\chi_2^*(\tau)\psi_i(\tau)\right]D(t)$$

$$+\int\int\int dE\, dE_1 dE_2 B_{E_1 a}^* B_{Ea}\rho_{EE_2}(t)(z-L)^{-1}_{E_1 i,E_2 i}$$

$$= \frac{2}{\gamma_i}\mathscr{L}(\chi_2^*\psi_i)D(t) + \left[\frac{2}{\gamma_i}\right]^{1/2}\int dE_2 \int dE\, \mathscr{L}(f_{2E_2}^* f_{ii})\rho_{EE_2}(t)B_{Ea} . \tag{5.31}$$

We next simplify $\hat{g}_T$ using (5.29) in (5.22),

$$\left[\frac{2}{\gamma_i}\right]^{1/2}\hat{g}_T(z,t) = (2)\frac{\gamma_i}{2}\int dE_1 \int dE_2 B_{E_1 a}^* B_{E_2 a}(z-L)^{-1}_{E_1 E_2 ii}\left[\frac{2}{\gamma_i}\right]^{1/2}\int dE\, B_{Ea}\rho_{Ei}(t)$$

$$+2\frac{\gamma_i}{2}\int dE_1 \int dE_2 B_{E_1 a}^* B_{E_2 a}\int dE_3 (z-L)^{-1}_{E_1 E_2,E_3 i}\int dE\, B_{Ea}\left[\frac{2}{\gamma_i}\right]^{1/2}\rho_{EE_3}(t) ,$$

which on using the matrix elements given by (5.15) can be simplified to

$$\left[\frac{2}{\gamma_i}\right]^{1/2}\hat{g}_T(z,t) = 2\mathscr{L}(\chi_2^*\chi_2)D(t)(2/\gamma_i) + 2\int\int dE\, dE_3 \mathscr{L}(f_{2E_3}^* f_{2i})\left[\frac{2}{\gamma_i}\right]^{1/2} B_{Ea}\rho_{EE_3}(t) . \tag{5.32}$$

On substituting (5.31) and (5.32) in (5.25) we obtain the dipole-dipole correlation function $(\gamma_i/2)\langle A_i^\dagger(t+\tau)A_i(t)\rangle$,

$$\frac{\gamma_i}{2}\hat{F}(z,t) = \mathscr{L}(\chi_2^*\psi_i)D(t) + 2\mathscr{L}(\chi_2^*\chi_2)D(t)\hat{D}^*(z) + \left[\frac{\gamma_i}{2}\right]^{1/2}\int dE\, B_{Ea}\int dE_2 \rho_{EE_2}(t)(f_{2E_2}^* f_{ii})$$

$$+2\left[\frac{\gamma_i}{2}\right]^{1/2}\int dE\, B_{Ea}\int dE_2 \rho_{EE_2}(t)\mathscr{L}(f_{2E_2}^* f_{2i})\hat{D}^*(z) . \tag{5.33}$$

The steady-state photon spectra can be obtained by combining (5.33), (2.9), and (2.13). The result is

$$N_i(\delta) = \frac{2}{\pi}\mathrm{Re}[S^{(1)}(z)+S^{(2)}(z)]_{z=i\delta} , \tag{5.34}$$

where

$$S^{(1)}(z) = \mathscr{L}(\chi_2^*\psi_i)\hat{D}(0) + 2\mathscr{L}(|\chi_2|^2)\hat{D}(0)\hat{D}^*(z) , \tag{5.35}$$

$$S^{(2)}(z) = \int_0^\infty dt\, e^{-zt}[\psi_i(t) + 2\hat{D}^*(z)\chi_2(t)] \times W(t)[1-\hat{g}_\sigma(0)]^{-1} , \tag{5.36}$$

and where the function $W(\tau)$ is given by

$$W(\tau)=\left[\frac{\gamma_i}{2}\right]^{1/2}\int dE\, B_{Ea}\int dE_2\hat{\sigma}_{EE_2}(0)f^*_{2E_2}(\tau)\ . \qquad (5.37)$$

In deriving (5.35) and (5.36) we have also used $\chi_2=V_2=f_{2i}$, $\psi_i=f_{ii}$. Note further that $\hat{\sigma}_{EE_2}(0)=\mathcal{L}(\psi_E\psi^*_{E_2})$ and hence

$$\left[\frac{\gamma_i}{2}\right]^{1/2}\int dE\, B_{Ea}\hat{\sigma}_{EE_2}=\mathcal{L}(\chi_2\psi^*_{E_2})\ . \qquad (5.38)$$

In view of (5.38) and using I(6.12), (5.37) reduces to

$$\hat{W}(z)=\sum_\alpha \chi_2^\alpha\int dE_2\psi^*_{E_2}(-z_\alpha)\hat{f}^*_{2E_2}(z)\ . \qquad (5.39)$$

The energy integral that appears in (5.39) is similar to that appearing in (4.8) and hence can be similarly reduced to

$$W(z)=\sum_\alpha \chi_2^{(\alpha)}\hat{h}^*_{21}(z,-z_\alpha)\ , \qquad (5.40)$$

$$\hat{h}_{21}(z,\mathfrak{z})=-\frac{i}{\mathfrak{z}-z}\left[\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{21}-\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{21} -\left[\frac{z-\mathfrak{z}}{\mathfrak{z}}\right]\{[\mathbb{1}+\underline{m}(\mathfrak{z})]^{-1}\}_{11}\{[\mathbb{1}+\underline{m}(z)]^{-1}\}_{21}\right]\ , \qquad (5.41)$$

$$\hat{h}_{21}(z,-z_\alpha)=\frac{\hat{\chi}_2(-z_\alpha)-\hat{\chi}_2(z)}{z+z_\alpha}-\hat{\psi}_i(-z_\alpha)\hat{\chi}_2(z)\ . \qquad (5.42)$$

$S^{(1)}(z)$ can be simplified further by noting from (5.28) that

$$\hat{D}^*(z)=\frac{C(z)}{1-g_\sigma(z)}\ , \qquad (5.43)$$

$$\hat{D}(0)=[\lim_{z\to 0}\hat{D}^*(z)]^*=\frac{C(0)^*}{1-g_\sigma(0)}\ , \qquad (5.44)$$

$$C(z)=\mathcal{L}(\chi_2^*\psi_i)=\sum_{i,j}\frac{\psi_i^{(i)}\chi_2^{(j)*}}{z-z_i-z_j^*}\ , \qquad (5.45)$$

$$g_\sigma(z)=2\,|\,\hat{\chi}_2(z)\,|^2=2\sum_{i,j}\frac{\chi_2^{(i)}\chi_2^{(j)*}}{z-z_i-z_j^*}\ . \qquad (5.46)$$

One then obtains

$$S^{(1)}(z)=\frac{C(z)}{1-g_\sigma(z)}\frac{C(0)^*}{1-g_\sigma(0)} =\hat{D}^*(z)\hat{D}(0)\ . \qquad (5.47)$$

To simplify $S^{(2)}(z)$ we return to (5.36), write $\chi_2(t)$ as in Eq. I(6.12), and expand $\psi_i(t)$ similarly. It follows that

$$S^{(2)}(z)=\frac{1}{1-\hat{g}_\sigma(0)} \times\sum_i[\psi_i^{(i)}+2\hat{D}^*(z)\chi_2^{(i)}]\hat{W}(z-z_i)\ . \qquad (5.48)$$

The terms in (5.42) can be expanded in summations to give

$$\hat{W}(z-z_i)=\hat{W}_1(z-z_i)+\hat{W}_2(z-z_i)\ ,$$

$$\hat{W}_1(z-z_i)=\sum_{\alpha,\beta}\frac{\chi_2^{(\alpha)}\chi_2^{(\beta)*}}{(z-z_i-z^*_\beta)(-z_\alpha-z^*_\beta)}\ , \qquad (5.49)$$

$$\hat{W}_2(z-z_i)=\sum_{\alpha,\beta}\frac{\chi_2^{(\alpha)}\chi_2^{(\beta)*}}{z-z_i-z^*_\beta}\left[\sum_k\frac{\psi_i^{(k)*}}{z_\alpha+z_k^*}\right]\ .$$

When (5.48) and (5.49) are combined explicitly, the terms involving $\hat{W}_2$ simplify nicely. The summations over i and β can be separated from the sums over α and k to give

$$S^{(2)}(z)=\frac{1}{1-\hat{g}_\sigma(0)}\sum_i[\psi_i^{(i)}+2\hat{D}^*(z)\chi_2^{(i)}]\hat{W}_1(z-z_i) +\left[\frac{-1}{1-\hat{g}_\sigma(0)}[C(z)+\hat{D}^*(z)\hat{g}_\sigma(z)]C(0)^*\right]\ . \qquad (5.50)$$

Next using expression (5.43) for $\hat{D}^*(z)$ the expression in large parentheses in (5.50) is

$$-\frac{1}{1-g_\sigma(0)}\left[\frac{C(z)}{1-\hat{g}_\sigma(z)}\right]C(0)^*=-S^{(1)}(z)\ . \qquad (5.51)$$

Thus we obtain

$$S^{(1)}(z)+S^{(2)}(z)=\frac{1}{1-\hat{g}_\sigma(0)} \times\sum_i[\psi_i^{(i)}+2\hat{D}^*(z)\chi_2^{(i)}]\hat{W}_1(z-z_i)\ . \qquad (5.52)$$

An explicit expression for $\chi_2^{(i)}$ is given in I(6.13), and $\hat{\psi}_i$ is given in I(7.4). It follows that

$$\psi_i^{(k)}=(-1)^{k+1}\frac{\epsilon_k-\Delta_a+i\eta}{\epsilon_+-\epsilon_-}\ . \qquad (5.53)$$

To examine the structure of the spectrum, note that $C(z)$ [Eq. (5.45)] and $\hat{W}_2$ [Eq. (5.49)] both have poles at $z=z_i+z_j^*$, $i,j=1,2$ and therefore will exhibit a triplet structure when we take $z=i\delta=i(\omega-\omega_l)$. The peaks will be located at

$$\delta = \mathrm{Re}[-i(z_i + z_j^*)] = \begin{cases} -\frac{1}{2}\Gamma\,\mathrm{Re}(\epsilon_i - \epsilon_j^*)\,, & i,j = 1,2 \\ 0\,, \quad \pm\frac{1}{2}\Gamma\,\mathrm{Re}(\epsilon_+ - \epsilon_-)\,. \end{cases} \tag{5.54}$$

The two peaks at $\delta = 0$ have widths $-\Gamma\,\mathrm{Im}(\epsilon_\pm)$ and the side peaks have width $-(\Gamma/2)\mathrm{Im}(\epsilon_+ + \epsilon_-)$. Notice, however, that the structure of the photon spectrum is complicated slightly by the factor $[1 - g_\sigma(z)]^{-1}$ appearing in $\hat{D}^*(z)$. When the spectrum is determined, this factor leads to a fourth-order polynomial in the denominator. The roots of this polynomial are expected to correspond to the four possible transitions between two effective dressed states of the problem. Two of these transitions are expected to have the same (laser) energy, but different widths (both these widths will be nonzero, since there is no coherent component in the present problem).

Numerical results for the steady state (long-time limit) photon spectra are shown in Figs. 7 and 8. Figure 7 shows the spectrum for large q_i but fairly low laser intensity, and clearly shows the triplet nature of the spectrum. The figure also shows the effects of nonzero γ_f. Figure 8 gives a series of curves with changing field strength[11,12] for $\alpha = q_i = 1$, $\gamma_i/\Gamma = 0.01$, and $\gamma_f/\Gamma = 0$, 0.01, and 0.1. The poles $\epsilon_\pm$ for these parameters can be interpolated from Fig. 4 of paper I. For small γ_f, the confluence is clearly shown by the narrow spike at the laser frequency ($\delta = 0$) and the increased area under the curve.

VI. CONCLUSIONS

In this series of papers we have shown in detail how the method of master equations can be applied to the problem of laser-induced autoionization. In the first paper we presented the general theory and applied it in studying the spectrum of ejected electrons. In this second paper, we have examined the properties of the ejected photons, both in the Raman transition to $|f\rangle$ and in the transition back to $|i\rangle$, the state from which the atom was excited by the laser. We have shown how the "confluence of coherences" is manifested by the number and frequency distribution of ejected photons.

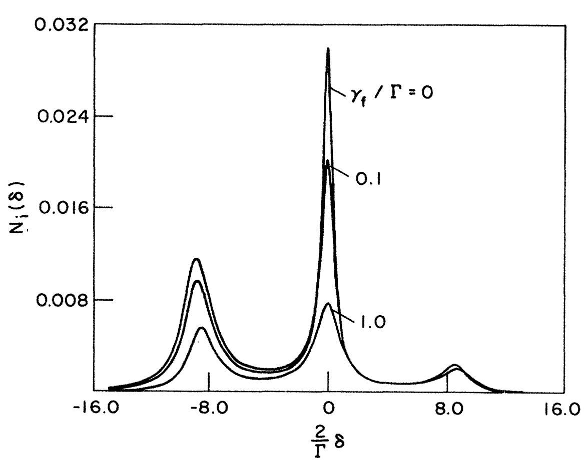

FIG. 7. Spectra to $|i\rangle$ in units of $1/\pi\Gamma$ vs $(2/\Gamma)\delta = (2/\Gamma)(\omega - \omega_l)$ for $q_i = 10$, $\alpha = 0$, $\Omega = 0.2$, $\gamma_i/\Gamma = 0.01$ and for $q_f = 1$, $\gamma_f/\Gamma = 0$, 0.1, and 1. Increasing γ_f diminishes the left and central peaks, but slightly enhances the right peak.

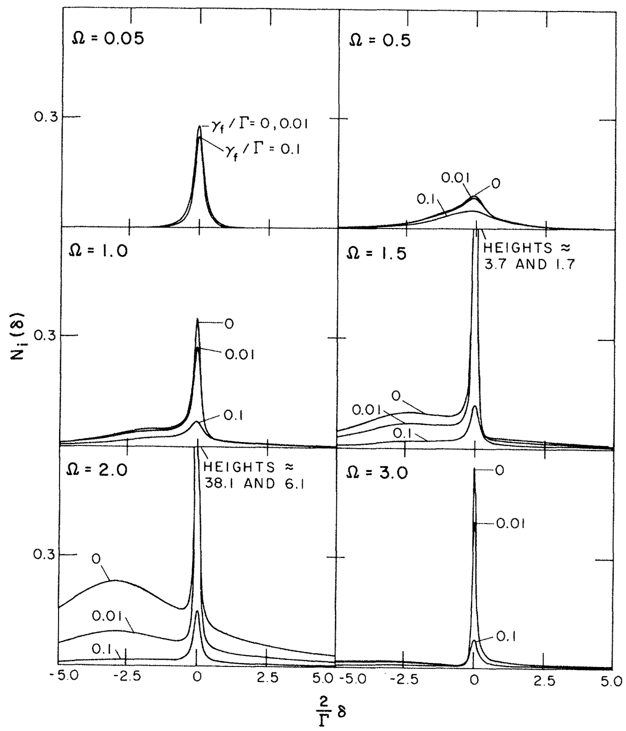

FIG. 8. Spectra to $|i\rangle$ for $\alpha = q_i = 1$, $\gamma_i/\Gamma = 0.01$, $\gamma_f/\Gamma = 0$, 0.01, and 0.1, and various Ω. The confluence occurs at $\Omega = 2.0$.

We have also shown that the photon spectrum for the Raman transition is a doublet, reflecting the ac Stark splitting of the autoionizing state, and that the spectrum for the transition back to $|i\rangle$ is essentially a triplet (since it contains the superposition of two peaks around the laser frequency) reflecting the Stark splitting of both the initial and final state. In addition, we have shown how the presence of two decay channels can affect the spectra and number of photons ejected in the individual channels.

Finally, it is worth noting again that the decay of a single atom in LIA is a transient process, and in the long-time limit the rate of photon emission is zero. Thus our spectrum differs from the usual photon spectrum, since the latter studies the *rate* of photon emission in the long-time limit. It is interesting to consider what would happen if the state $|i\rangle$ were continuously pumped at some rate λ, so that there was always some population in $|i\rangle$. Intuitively, one would expect a coherent component to appear since under these circumstances the system has a nonzero mean dipole moment. This question will be examined elsewhere.

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APPENDIX: BRANCHING RATIOS AND SPECTRA FROM AUTOIONIZING STATES IN THE ABSENCE OF DRIVING FIELDS

In this appendix we show how some of the usual results on branching ratios, etc., follow from our master equation. We will assume that the atom was initially in the excited state $|a\rangle$ and that no coherent field is present. We also neglect the state $|f\rangle$. The initial conditions to be used are

$$\psi_i=0\ ,\quad \psi_E=b^*_{Ea}\ . \tag{A1}$$

For simplicity, we will assume only one channel of spontaneous emission by putting $\gamma_f=0$. Equation I(3.6) is directly soluble as

$$\rho_{E_1E_2}(t)=\psi_{E_1}(t)\psi^*_{E_2}(t)\ , \tag{A2}$$

$$\hat{\psi}_{E_1}+\int dE\,K_2(E_1)L_2(E)\hat{\psi}_E=\frac{b^*_{E_1a}}{z+i\Delta_{E_1}} \tag{A3}$$

with the result

$$\hat{\psi}_E=-K_2(E)(1+m_{22})^{-1}f_2+\frac{b^*_{Ea}}{z+i\Delta_E}\ ,$$

$$\int \hat{\psi}_E L_2(E)dE=f_2/(1+m_{22})\ , \tag{A4}$$

$$f_2=\int dE\frac{L_2(E)b^*_{Ea}}{z+i\Delta_E}$$

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

Hence in the steady state

$$p_E=\lim_{t\to\infty}\rho_{EE}(t)=|\psi_E|^2\ ,$$

$$\psi_E=b^*_{Ea}-B^*_{Ea}\frac{f_2}{1+m_{22}}\ ,$$

which on simplification leads to

$$p_E=\frac{\Gamma}{2\pi\psi^2}\left[1+\frac{\gamma_i^2}{\Gamma^2q_i^2}\right]\left[\left[\frac{\Gamma+\gamma_i}{2\psi}\right]^2+\frac{\Gamma^2}{4}(E-\Delta_a)^2\right]^{-1}, \tag{A5}$$

where Δ_a,ψ have been previously defined

$$\psi=1+\frac{\gamma_i}{\Gamma q_i^2}\ ,\quad \Delta_a=-\frac{2\gamma_i}{q_i\psi\Gamma}\ . \tag{A6}$$

The total number of electrons emitted will be

$$p=\int dE\,p_E=\left[\frac{\Gamma}{\Gamma+\gamma_i}\right]\frac{1}{\psi}\left[1+\frac{\gamma_i^2}{\Gamma^2q_i^2}\right]. \tag{A7}$$

We next consider the photon spectrum, which can be obtained from I(3.5) and the regression theorem:

$$\frac{d}{d\tau}\langle(|E_1\rangle\langle i|)_{t+\tau}A_i(t)\rangle=i\Delta_{E_1}\langle(|E_1\rangle\langle i|)_{t+\tau}A_i(t)\rangle-\frac{\gamma_i}{2}\int dE\,B_{E_1a}B^*_{Ea}\langle(|E\rangle\langle i|)_{t+\tau}A_i(t)\rangle\ , \tag{A8}$$

the initial condition being

$$\langle(|E_1\rangle\langle i|)_tA_i(t)\rangle=\int dE_2B_{E_2a}\rho_{E_2E_1}(t)\ . \tag{A9}$$

The solutions of these equations are trivial [cf. (A4)] and we quote the result for the photon spectrum that follows from (2.13), (A8), and (A9):

$$N_i(\delta')=\frac{1}{\psi^2}\left[\frac{\gamma_i}{2\pi}\right]\frac{1+\frac{1}{q_i^2}}{\frac{(\Gamma+\gamma_i)^2}{4\psi^2}+\frac{\Gamma^2}{4}(\delta'-\Delta_a)^2}\ ,$$

$$\delta'=\frac{2(\omega-E_a)}{\Gamma}\ . \tag{A10}$$

The total number of photons will be

$$N_i=\int N_i(\delta')d\omega=\frac{\gamma_i}{\Gamma+\gamma_i}\frac{1}{\psi}\left[1+\frac{1}{q_i^2}\right]. \tag{A11}$$

We can now introduce the branching ratios $\tilde{\gamma},\tilde{\Gamma}$ by defining

$$N_i=\frac{\tilde{\gamma}}{\tilde{\gamma}+\tilde{\Gamma}}\ ,\quad N_p=\frac{\tilde{\Gamma}}{\tilde{\Gamma}+\tilde{\gamma}}\ , \tag{A12}$$

$$\tilde{\gamma}=\frac{\gamma}{\psi^2}\left[1+\frac{1}{q_i^2}\right],\quad \tilde{\Gamma}=\frac{\Gamma}{\psi^2}\left[1+\frac{\gamma_i^2}{\Gamma^2q_i^2}\right]. \tag{A13}$$

The rate $\tilde{\gamma}$ also turns out to have the interpretation as it should

$$\lim_{t\to 0}\frac{\partial\rho_{ii}}{\partial t}=\tilde{\gamma}\ . \tag{A14}$$

The branching ratios $\tilde{\gamma}$ and $\tilde{\Gamma}$ are in agreement with Armstrong *et al.*[5] The characteristic shifts in the peak positions of the photon and electron spectra are to be noticed. They were discussed in paper I, and such shifts depend on q values. If we had ignored the decay of the unperturbed continuum, or "virtual recombination," then $\Delta_a\to 0,\psi\to 1$ and the spectra have a very simple form. For small values of q and moderate values of γ, the decay of the unperturbed continuum cannot be ignored.

The analysis for the branching ratios or the effective decay rates helps us to reinterpret Fano's asymmetry parameter q. For this purpose we recall the definitions of q

$$q^2=\frac{2|V_{ia}|^2}{\pi\Gamma|V_{iE}|^2}=\frac{\gamma\beta}{\Gamma|V_{iE}|^2}\ , \tag{A15}$$

where β is a numerical parameter. Using (A15) we can rewrite $\tilde{\gamma}$ as

$$\tilde{\gamma}=\frac{\gamma\left[1+\frac{\Gamma}{\gamma\beta}\mid V_{iE}\mid^2\right]}{\left[1+\frac{\gamma}{\Gamma q^2}\right]^2}=\frac{\left[\gamma+\frac{\Gamma}{\beta}\mid V_{iE}\mid^2\right]}{\left[1+\frac{\mid V_{iE}\mid^2}{\beta}\right]^2}\,. \quad \text{(A16)}$$

We now examine (A16) in two limits—(a) When $\Gamma\rightarrow 0$, i.e., the state $\mid a\rangle$ does not autoionize, then

$$\tilde{\gamma}_1=\lim_{\Gamma\rightarrow 0}\tilde{\gamma}=\gamma\left[1+\frac{\mid V_{iE}\mid^2}{\beta}\right]^{-2}. \quad \text{(A17)}$$

(b) When $\gamma\rightarrow 0$, i.e., the state $\mid a\rangle$ does not decay by spontaneous emission, then

$$\tilde{\gamma}_2=\lim_{\gamma\rightarrow 0}\tilde{\gamma}=\frac{\Gamma}{\beta}\mid V_{iE}\mid^2\left[1+\frac{\mid V_{iE}\mid^2}{\beta}\right]^{-2}. \quad \text{(A18)}$$

It is evident from (A17) and (A18) that

$$q^2=\tilde{\gamma}_1/\tilde{\gamma}_2\,. \quad \text{(A19)}$$

Therefore, the square of Fano's q parameter is equal to the total rate of radiative decay without autoionization of $\mid a\rangle$ divided by the total rate of radiative decay with autoionization but with no radiative decay of $\mid a\rangle$.

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[10]This figure is a corrected version of Fig. 6 of Ref. 6.

[11]The results obtained in this paper for $\gamma_f=0$ agree with those obtained by Lewenstein *et al.* (Ref. 4) using a different approach. Our analysis is based on the quantum regression theorem, whereas Lewenstein *et al.* work directly with Heisenberg equations of motion. One can show that the structure of Eqs. (2.38)—(2.40) with $\gamma_f=0$ of Haus *et al.* is identical to our Eqs. (5.2)—(5.5).

[12]Some of our results for $\gamma_f=0$ have been described in a previous paper: G. S. Agarwal, S. L. Haan, and J. Cooper, Phys. Rev. A 28, 1154 (1983).