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Effects of detuning and fluctuations on fluorescence radiation from a strongly driven three-level atom: Some analytical results

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We generalize our earlier formalism to treat the effects of both the finite-bandwidth excitations and finite arbitrary detunings, on the fluorescent spectra and second-order intensity correlation functions in optical double resonance. It is assumed that the bandwidth arises from the phase and/or amplitude fluctuations in the fields driving a three-level atom in cascade configuration. Under the conditions that one or both of these fields are intense, secular approximation and the theory of multiplicative stochastic processes are invoked to derive a Markovian master equation for the atomic-density operator averaged over both phase and amplitude. The quantum regression theorem is used to derive analytical expressions for the fluorescent spectra and the second-order intensity correlation functions which explicitly display the effects due to detunings and fluctuations.

I. INTRODUCTION

The interaction of a three-level atom with one or more strong laser fields (double resonance) continues to draw considerable attention both theoretically[1–4] and experimentally.[5,6] The spectrum of the fluorescent radiation from a three-level atom undergoing stepwise resonant transition is a Stark quintuplet for both the upper as well as the lower transitions. If, however, the driving fields are detuned from the atomic-transition frequencies, the spectrum exhibits as many as seven Lorentzian peaks.[1,3] On the other hand, if the lower transition is driven by a strong field whereas the upper transition is probed by a weak field, then the spectrum from the lower transition is found to be the Stark triplet which is characteristic of a strongly driven two-level system.[7] The upper spectrum, in this case, is the so-called Autler-Townes doublet.[4–6,8]

There are two other interesting aspects connected with the fluorescence from a three-level atom. First is the quantum nature of the fluorescent light which has been discussed extensively in literature.[9] The second which has evoked much recent discussion is related to the fact that fluorescence from two separate transitions in a three-level atom is affected considerably if the atom undergoes a quantum jump between two levels.[10] It has even been suggested that such coupled transitions may be used to detect experimentally[11] a weak transition. Theoretical understanding of these aspects is obtained through the intensity-intensity correlation functions.

An important problem both in its own right as well as having a direct bearing on experiments is to study how finite bandwidths of the driving laser fields affects the fluorescence from the atom. The bandwidths may arise, say, due to phase and/or amplitude fluctuations in the lasers. Further, the fluctuations of the two lasers may not be statistically independent, giving rise to cross correlations. The effects of excitation bandwidths due to phase fluctuations on the Autler-Townes doublet were studied theoretically at first by Georges and Lambropoulos[12] and subsequently by Agarwal and Narayana.[13] It was shown[12,13] that the asymmetry in the Autler-Townes doublet could switch due to phase fluctuations. Cross correlations have also been shown to modify the Autler-Townes doublet.[14,15] The effects of cross correlations on population trapping and other phenomena in three-level systems have been discussed theoretically by Dalton and Knight.[16] Numerical results on the effects of the phase fluctuations on the fluorescent spectrum in the presence of two intense driving fields has been reported by D'Souza *et al.*[17] They have shown that with an increase in the bandwidths of phase fluctuations, the central peak and the remote sidebands in the fluorescent spectrum begin to disappear and the Stark quintuplet tends to reduce to a Stark doublet.

These studies, however, have not taken into account the effects due to fluctuations in the amplitude of the driving fields which are known to play a significant role in many situations. In this context some analytic results under the conditions that one or both of the fields are intense were presented very recently by Lawande *et al.*[18] However, the formulation of this paper was restricted to the case where both the excitation fields were in exact resonance with the respective atomic transitions and some subtle effects due to detunings could not be dis-

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cussed. In this paper, we remove this restriction and present a more general formalism which is valid for arbitrary detunings between the excitation fields and the atomic transitions. The basic assumptions of Ref. 18, viz., that the phases follow a Wiener-Levy process and that the amplitude fluctuations are described by a colored Gaussian process[12–24] are retained in the present paper. This assumption for the phase fluctuations leads to an exact master equation averaged over the distribution of phase fluctuations. The subsequent treatment of amplitude fluctuations requires one or both the excitation fields to be intense so that a secular approximation can be invoked. Within this approximation, we derive the master equation for the atomic density operator averaged over the amplitude fluctuations as well. This master equation in the high-field limit is shown to possess an exact steady-state solution. We subsequently use the master equation to derive analytic expressions for the spectrum of the fluorescent radiation from the upper as well as lower excitation levels which show explicitly the effects due to detunings and fluctuations. The analytic results for phase fluctuations are found to be in agreement with the numerical work of Ref. 17. We also obtain analytic expressions for the intensity-intensity correlation functions which also display the detunings and fluctuation effects.

We present in Sec. II the basic formulation of the problem leading to the derivation of the master equation in the high-field limit and its steady-state solution. The derivation and discussion of the analytic expressions for the fluorescent spectra and the intensity-intensity correlation functions is relegated to Sec. III. A summary of the general results is outlined in Sec. IV. Finally, the special case where both excitation fields are in resonance with the atomic transitions is briefly treated in Appendix A.

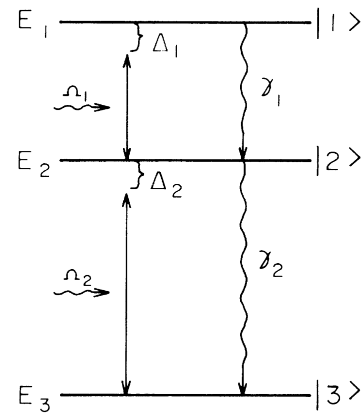

FIG. 1. Schematic diagram of a three-level atom interacting with two monochromatic fields.

II. FORMULATION OF THE PROBLEM

A. Master equation

We consider a three-level atom with an upper excited state $|1\rangle$, an intermediate excited state $|2\rangle$, and the ground state $|3\rangle$ of energies $\hbar\omega_1$, $\hbar\omega_2$, and $\hbar\omega_3$ in simultaneous interaction with two single-mode cw laser fields (Fig. 1). The first laser driving the atomic transition $|3\rangle$ to $|2\rangle$ has a frequency Ω_2 which is detuned from the atomic-transition frequency $\omega_{23}=\omega_2-\omega_3$ by an amount Δ_2. The second laser of frequency Ω_1 drives the atomic transition $|2\rangle$ to $|1\rangle$ and is detuned from the atomic transition frequency $\omega_{12}=\omega_1-\omega_2$ by an amount Δ_1.

It is customary to describe the system by means of the following master equation[25] for the reduced atomic density operator ρ:

$$d\rho/dt=-i(d_1/2)[E_1^*(t)A_{12}+E_1(t)A_{21},\rho]-i(d_2/2)[E_2^*(t)A_{23}+E_2(t)A_{32},\rho]$$
$$-i(\Delta_1+\Delta_2)[A_{11},\rho]-i\Delta_2[A_{22},\rho]-\gamma_1(A_{11}\rho+\rho A_{11}-2A_{21}\rho A_{12})-\gamma_2(A_{22}\rho+\rho A_{22}-2A_{32}\rho A_{23}) . \quad (2.1)$$

The master equation (2.1) involves the usual electric dipole and rotating-wave approximations. Further, the Born and Markov approximations with respect to the interaction with the continuum modes of the radiation field are inherent in the derivation. Lastly, the equation is written in the frame rotating with respect to the laser frequencies. The coefficients d_1 and d_2 represent the dipole matrix elements corresponding, respectively, to the transitions $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$. Also, $2\gamma_1$ and $2\gamma_2$ are radiative spontaneous transition probabilities per unit time for the atom to make a transition from the level $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$ respectively. The operators $A_{mn}=|m\rangle\langle n|$ obey the usual commutation relations

$$[A_{mn},A_{pq}]=A_{mq}\delta_{np}-A_{pn}\delta_{qm} . \quad (2.2)$$

The time dependence of the applied fields $E_1(t)$ and $E_2(t)$ arises from the stochastic nature of their amplitude and phases. We may write

$$E_j(t)=[E_j^{(0)}+E_j^{(1)}(t)]\exp[-i\phi_j(t)], \quad \phi_j(0)=\phi_{j0} \quad (j=1,2) . \quad (2.3)$$

where the nonstochastic amplitude $E_j^{(0)}$ and the phases ϕ_{j0} are positive real numbers while the slowly varying time-dependent quantities $E_j^{(1)}(t)$ and $\phi_j(t)$ are treated as stochastic variables. Now most of the papers dealing with stochastic theory of atoms interacting with noisy lasers assume that the noise can be adequately described by Gaussian stochastic processes.[12–24] The physical basis for such an assumption arises from the fact that in real laser systems many independent fluctuation mechanisms contribute to the laser bandwidth and superposition of all these can, within central-limit theorem, be described by a Gaussian stochastic process.[19] Moreover, Gaussian stochastic description is analytically simple and tractable. In the present work we therefore adopt an extension of the standard model used to describe a single-mode noisy laser with small fluctuations.[23] However, additional complications arise when two lasers interact simultaneously with the atom as in the present problem. In this case we have to take into account not only the individual correlations in each laser but also the cross correlations between the lasers.[21] These cross correlations may also arise naturally if the two fields are different modes of the same laser or if the second field is produced by splitting and frequency conversion from the first laser beam.

We assume that the fluctuations in phase and amplitude are statistically uncorrelated, described by independent Gaussian distributions. The statistics of phase fluctuations are described by the Wiener-Levy phase-diffusion model described by the Langevin equations[12–22]

$$d\phi_j(t)/dt=\mu_j(t)\ , \tag{2.4}$$

where $\mu_j(t)$ is a Gaussian white noise with the properties

$$\langle \mu_j(t)\rangle =0\ , \tag{2.5a}$$

$$\langle \mu_j(t)\mu_k(t')\rangle =2\delta(t-t')\times\begin{cases}\gamma_{c_j} & j=k\\ \gamma_{cc} & j\neq k\ .\end{cases} \tag{2.5b}$$

Here γ_{cc} represents any cross correlations that may be present between the lasers. Note that this model implies purely Lorentzian line shapes. It is known that the real laser linewidth may be more nearly Gaussian than Lorentzian, particularly in the wings. A model of non-Lorentzian laser line shapes is provided if we assume $\phi_j(t)$ to be an Ornstein-Uhlenbeck process.[19] Incidently, this model has been employed in Ref. 15 to discuss the cross-correlation effects on the optical double-resonance spectra. The formulation of the present paper requires a nontrivial extension to treat this case and needs a separate discussion. The amplitude fluctuations are assumed to be described by a Gaussian colored noise with the mean and correlations given by[23,24]

$$\langle E_j^{(1)}(t)\rangle =0\ , \tag{2.6a}$$

$$\langle E_j^{(1)}(t)E_k^{(1)}(t')\rangle =\begin{cases}\epsilon_j^2\exp[\gamma_{a_j}\,|\,t-t'\,|\,] & j=k\\ \epsilon_{12}^2\exp[\gamma_{aa}\,|\,t-t'\,|\,] & j\neq k\ ,\end{cases} \tag{2.6b}$$

where ϵ_j^2 and γ_{a_j} are the intensities and the bandwidths for the autocorrelations of the two fields while ϵ_{12}^2 and γ_{aa} are similar parameters accounting for the cross correlation between the two fields.

B. Density operator averaged over phase fluctuations

An advantage of the Wiener-Levy model of phase fluctuations is that it is possible to derive from (2.1) a master equation for the density operator averaged over the ensemble of phase fluctuations. For this purpose we introduce the transformation[17,18]

$$\begin{aligned}W^{pq}(t)=&\exp[-i(p\phi_1+q\phi_2)]\\&\times\exp\{-i[(\phi_1+\phi_2)A_{11}+\phi_2A_{22}]\}\\&\times\rho\exp\{i[(\phi_1+\phi_2)A_{11}+\phi_2A_{22}]\}\ ,\end{aligned} \tag{2.7}$$

in (2.1). This results in the following equation:

$$\begin{aligned}dW^{pq}(t)/dt=\{&L_0-i\dot\phi_1(p+L_1)-i\dot\phi_2(q+L_2)\\&-i(d_1/2)E_1^{(1)}(t)L_3\\&-i(d_2/2)E_2^{(1)}(t)L_4\}W^{pq}(t)\ ,\end{aligned} \tag{2.8}$$

where

$$\begin{aligned}L_oW^{pq}=&-i[H_0,W^{pq}]\\&-\gamma_1(A_{11}W^{pq}+W^{pq}A_{11}-2A_{21}W^{pq}A_{12})\\&-\gamma_2(A_{22}W^{pq}+W^{pq}A_{22}-2A_{32}W^{pq}A_{23})\ ,\end{aligned} \tag{2.9}$$

$$H_0=\alpha_1(A_{12}+A_{21})+\alpha_2(A_{23}+A_{32})+\Delta A_{11}+\Delta_2A_{22}\ , \tag{2.10}$$

$$L_1W^{pq}=[A_{11},W^{pq}]\ , \tag{2.11}$$

$$L_2W^{pq}=[A_{11}+A_{22},W^{pq}]\ , \tag{2.12}$$

$$L_3W^{pq}=[A_{12}+A_{21},W^{pq}]\ , \tag{2.13}$$

$$L_4W^{pq}=[A_{23}+A_{32},W^{pq}]\ , \tag{2.14}$$

$$\alpha_j=d_jE_j^{(0)},\quad \Delta=\Delta_1+\Delta_2\ . \tag{2.15}$$

The next step is to obtain the master equation for $\chi^{pq}=\langle W^{pq}\rangle$, the transformed density operator averaged over the distributions of the phases $\phi_1(t)$ and $\phi_2(t)$. Since $\dot\phi_j=\mu_j(t)$ represent δ-correlated Gaussian processes, it is possible to apply immediately the theory of multiplicative stochastic processes[26] to obtain an exact equation for the evolution of χ^{pq},

$$\begin{aligned}d\chi^{pq}(t)/dt=[&L_0-\gamma_{c_1}(p+L_1)^2-\gamma_{c_2}(q+L_2)^2\\&-2\gamma_{cc}(p+L_1)(q+L_2)\\&-i(d_1/2)E_1^{(1)}(t)L_3\\&-i(d_2/2)E_2^{(1)}(t)L_4]\chi^{pq}\ .\end{aligned} \tag{2.16}$$

The density operator χ^{pq} may be used directly to compute the one-time expectation values of the atomic operators averaged over the ensembles of the phase fluctuations. In particular, the phase-averaged expectation value of the operator A_{kk} connecting the diagonal states is given by

$$\langle \overline{A}_{kk}\rangle =\overline{\mathrm{Tr}(A_{kk}\rho)}=\mathrm{Tr}(A_{kk}\chi^{00})\ . \tag{2.17}$$

On the other hand, the phase-averaged expectation values of the operators connecting the off-diagonal states are given by

$$\langle \overline{A_{12}}\rangle =\overline{\mathrm{Tr}(A_{12}\rho)}=\mathrm{Tr}(A_{12}\chi^{10})\ , \tag{2.18}$$

$$\langle \overline{A_{23}} \rangle = \overline{\mathrm{Tr}(A_{23}\rho)} = \mathrm{Tr}(A_{23}\chi^{01}) \ . \tag{2.19}$$

Thus, in principle, within the diffusion model the phase fluctuations can be treated exactly. In fact, the master equation (2.16) in the absence of amplitude fluctuations has been solved numerically in Ref. 17 to study the effects due to phase fluctuations on the steady-state populations, fluorescent spectra, and intensity-intensity correlation functions.

C. Strong-field limit and amplitude fluctuations

It is clear from (2.16) that the operators L_3 and L_4 containing the fluctuations in the field amplitudes do not commute with the remaining operators in the master equation. It is therefore not immediately possible to derive an equation for $\overline{\chi}^{pq}$, the density operator averaged over both phase and amplitude fluctuations. A considerable simplification arises if we assume the fields to be strong. We shall show below that in this limit, a master equation for $\overline{\chi}^{pq}$ can be derived.

We first diagonalize the Hamiltonian H_0 [Eq. (2.10)] of the driven atomic system. Denoting by $|\psi_i\rangle$ the eigenstates of H_0 corresponding to the eigenvalue $-p_i$, we express the atomic states $|i\rangle$ by

$$|1\rangle = \sum_{i=1}^{3} a_i |\psi_i\rangle \ , \tag{2.20a}$$

$$|2\rangle = \sum_{i=1}^{3} b_i |\psi_i\rangle \ , \tag{2.20b}$$

$$|3\rangle = \sum_{i=1}^{3} c_i |\psi_i\rangle \ , \tag{2.20c}$$

where a_i, b_i, and c_i have the explicit expressions

$$\begin{aligned} a_i &= \alpha_1\alpha_2 N_i \ , \\ b_i &= -\alpha_2(p_i+\Delta)N_i \ , \\ c_i &= [(p_i+\Delta)(p_i+\Delta_2)-\alpha_1^2]N_i \ , \end{aligned} \tag{2.21}$$

where the constants N_i are defined as

$$N_i = \{\alpha_1^2\alpha_2^2+\alpha_2^2(p_i+\Delta)^2+[(p_i+\Delta)(p_i+\Delta_2)-\alpha_1^2]^2\}^{-1/2} \ , \tag{2.22}$$

and p_i are the roots of the cubic equation

$$p^3+p^2(\Delta+\Delta_2)-p(\alpha_1^2+\alpha_2^2-\Delta_2\Delta)-\alpha_2^2\Delta=0 \ . \tag{2.23}$$

The cubic equation can be solved readily whenever $\Delta_1=\Delta_2=0$ or when $\Delta_1=-\Delta_2$. In these cases the coefficients a_i,b_i,c_i can be written down explicitly. Thus, under resonance conditions ($\Delta_1=\Delta_2=0$) we have

$$p_1=0, \quad p_{2,3}=\pm\Omega, \quad \Omega=(\alpha_1^2+\alpha_2^2)^{1/2} \ , \tag{2.24}$$

$$a_1=\alpha_2/\Omega, \quad a_2=a_3=\alpha_1/(\sqrt{2}\Omega) \ , \tag{2.25a}$$

$$b_1=0, \quad b_2=-b_3=-1/\sqrt{2} \ , \tag{2.25b}$$

$$c_1=-\alpha_1/\Omega, \quad c_2=c_3=\alpha_2/(\sqrt{2}\Omega) \ . \tag{2.25c}$$

On the other hand, under exact optical double-resonance conditions ($\Delta=\Delta_1+\Delta_2=0$), we have

$$\begin{aligned} p_1 &= 0 \ , \\ p_{2,3} &= (\Delta_1\pm\Gamma)/2 \ , \\ \Gamma &= (4\Omega^2+\Delta_1^2)^{1/2} \ , \end{aligned} \tag{2.26}$$

$$\begin{aligned} a_1 &= \alpha_2/\Omega \ , \\ a_2 &= \alpha_1[2/\Gamma(\Gamma+\Delta_1)]^{1/2} \ , \\ a_3 &= \alpha_1[2/\Gamma(\Gamma-\Delta_1)]^{1/2} \ , \end{aligned} \tag{2.27a}$$

$$\begin{aligned} b_1 &= 0 \ , \\ b_2 &= -[(\Gamma+\Delta_1)/2\Gamma]^{1/2} \ , \\ b_3 &= [(\Gamma-\Delta_1)/2\Gamma]^{1/2} \ , \end{aligned} \tag{2.27b}$$

$$\begin{aligned} c_1 &= -\alpha_1/\Omega \ , \\ c_2 &= \alpha_2[2/\Gamma(\Gamma+\Delta_1)]^{1/2} \ , \\ c_3 &= \alpha_2[2/\Gamma(\Gamma-\Delta_1)]^{1/2} \ . \end{aligned} \tag{2.27c}$$

In other cases, we have to obtain the roots of the cubic equation numerically to obtain the coefficients a_i, b_i, and c_i.

We may now express the original operators A_{ij} appearing in the master equation (2.16) in terms of the new dressed operators $B_{ij}=|\psi_i\rangle\langle\psi_j|$ by means of the relations

$$A_{11} = \sum_{k,l=1}^{3} a_k a_l B_{kl} \ , \tag{2.28a}$$

$$A_{12} = \sum_{k,l=1}^{3} a_k b_l B_{kl} = A_{21}^{\dagger} \ , \tag{2.28b}$$

$$A_{13} = \sum_{k,l=1}^{3} a_k c_l B_{kl} = A_{31}^{\dagger} \ , \tag{2.28c}$$

$$A_{22} = \sum_{k,l=1}^{3} b_k b_l B_{kl} \tag{2.28d}$$

$$A_{23} = \sum_{k,l=1}^{3} b_k c_l B_{kl} = A_{32}^{\dagger} \ , \tag{2.28e}$$

$$A_{33} = \sum_{k,l=1}^{3} c_k c_l B_{kl} \ . \tag{2.28f}$$

The reciprocal relations between B_{ij} and A_{ij} can be easily written down by noting that the transformation matrix involving the products of a_i, b_i, and c_i is real orthogonal. The new operators B_{ij} obviously satisfy the same commutation relations as the old. Also, in terms of the new operators the Hamiltonian H_0 takes the simple form

$$H_0 = -\sum_{k=1}^{3} p_k B_{kk} \tag{2.29}$$

and that under the action of the Hamiltonian H_0, B_{ij} evolve with time as

$$B_{ij}(t) = B_{ij}(0)\exp[-i(p_i-p_j)t] \ . \tag{2.30}$$

Next, we go over to the interaction representation by defining

$$\tilde{\chi}^{pq} = \exp\left[-it\sum_k p_k B_{kk}\right]\chi^{pq}\exp\left[it\sum_k p_k B_{kk}\right], \quad (2.31)$$

whereby the resulting master equation for $\tilde{\chi}^{pq}$ splits into two parts, viz., the one containing no oscillatory terms and the other containing rapidly oscillating terms such as $\exp[\pm i(p_i - p_j)t]$, $\exp[\pm 2i(p_i - p_j)t]$, and $\exp[\pm i(2p_i - p_j - p_k)t]$ $(i,j,k=1,2,3;\ i\neq j)$. Making the secular approximation, that is, neglecting the oscillatory terms and finally reverting back to the Schrödinger picture, we arrive at the master equation

$$d\chi^{pq}/dt = [\mathcal{L}_0 - i(d_1 E_1^{(1)})\mathcal{L}_1 - i(d_2 E_2^{(1)})\mathcal{L}_2]\chi^{pq}, \quad (2.32)$$

where the operators $\mathcal{L}_0$, $\mathcal{L}_1$, and $\mathcal{L}_2$ have the following structure:

$$\mathcal{L}_0\chi^{pq} = i\sum_k p_k[B_{kk},\chi^{pq}] - a(p,q)\chi^{pq} - \sum_k g_k(p,q)B_{kk}\chi^{pq} - \sum_k g_k(-p,-q)B_{kk}\chi^{pq}$$
$$+2\sum_{k,l} e_{kl}B_{kk}\chi^{pq}B_{ll} + 2\sum_{\substack{k,l\\(k\neq l)}} f_{kl}B_{kl}\chi^{pq}B_{lk} + 2\sum_{\substack{k,l\\(k\neq l\neq 1)}} g_{kl}(B_{1k}\chi^{pq}B_{1l} + B_{k1}\chi^{pq}B_{l1})\delta_{\Delta_1,0}\delta_{\Delta_2,0}, \quad (2.33)$$

$$\mathcal{L}_1\chi^{pq} = \sum_k a_k b_k [B_{kk},\chi^{pq}], \quad (2.34)$$

$$\mathcal{L}_2\chi^{pq} = \sum_k b_k c_k [B_{kk},\chi^{pq}], \quad (2.35)$$

where

$$a(p,q) = p^2\gamma_{c_1} + 2pq\gamma_{cc} + q^2\gamma_{c_2}, \quad (2.36)$$

$$g_k(p,q) = a_k^2[\gamma_1 + (2p+1)\gamma_{c_1} + (2q+1)\gamma_{c_2} + 2(p+q+1)\gamma_{cc}] + b_k^2[\gamma_2 + (2q+1)\gamma_{c_2} + p\gamma_{cc}], \quad (2.37)$$

$$e_{kl} = b_k b_l(\gamma_1 a_k a_l + \gamma_2 c_k c_l) + a_k^2 a_l^2\gamma_{c_1} + \gamma_{c_2}(1-c_k^2)(1-c_l^2) + \gamma_{cc}(2a_k^2 a_l^2 + a_l^2 b_k^2 + a_k^2 b_l^2) = e_{lk}, \quad (2.38)$$

$$f_{kl} = b_k^2 a_l^2\gamma_1 + c_k^2 b_l^2\gamma_2 + a_k^2 a_l^2\gamma_{c_1} + c_k^2 c_l^2\gamma_{c_2} - 2a_k c_k a_l c_l\gamma_{cc}, \quad k\neq l \quad (2.39)$$

$$g_{kl} = 2a_1^2(\gamma_{c_1} + \gamma_{c_2} + \gamma_{cc})a_k a_l, \quad k\neq l\neq 1. \quad (2.40)$$

We might mention here that if there are some relations between the roots p_1, p_2, p_3 of the cubic equation additional terms may appear in the master equation. An obvious example of this is the case of resonant excitations where $\Delta_1 = \Delta_2 = 0$. Here, $2p_1 - p_2 - p_3 = 0$ and terms which are otherwise oscillatory now become independent of time and have to be included in the master equation. We have explicitly written these terms in the master equation above. Relations of this type might also occur for specific choice of the Rabi frequencies α_1 and α_2 and detunings Δ_1 and Δ_2. It is easy, however, to evaluate and take into account these additional terms. As we shall see subsequently, these terms do not affect the steady-state behavior of the atomic operator averages. In particular, they do not affect the equations of motion of the averages of the diagonal operators B_{ii}. Physical consequences of this include that intensity-intensity correlations are not affected but the fluorescent spectra are affected, as we shall see in the particular case of resonant excitations ($\Delta_1 = \Delta_2 = 0$). Moreover, note that in the absence of phase fluctuations, these terms do not contribute at all. Lastly, it may be emphasized that the equation is valid under the conditions that $|\Omega_{ij}| = |(p_i - p_j)| \gg \gamma_k, \gamma_c, \gamma_a, \epsilon_k$ for $i,j = 1-3$ and $k = 1,2$.

Note that the operators $\mathcal{L}_1$ and $\mathcal{L}_2$ multiplying the stochastic variables related to the amplitude fluctuations commute with each other and also with $\mathcal{L}_0$. We can therefore invoke the theory of multiplicative stochastic processes to arrive at the following evolution equation for the phase- and amplitude-averaged density operator $\bar{\chi}^{pq}$,

$$d\bar{\chi}^{pq}/dt = \{\mathcal{L}_0 - \epsilon_1^2(t)\mathcal{L}_1^2 - \epsilon_2^2(t)\mathcal{L}_2^2 - \epsilon_{12}^2\mathcal{L}_1\mathcal{L}_2\}\chi^{pq}, \quad (2.41)$$

where

$$\epsilon_j^2(t) = d_j^2\epsilon_j^2[1 - \exp(-\gamma_{a_j}t)]/\gamma_{a_j}, \quad j = 1,2 \quad (2.42a)$$

$$\epsilon_{12}^2(t) = 2d_1 d_2\epsilon_{12}^2[1 - \exp(-\gamma_{aa}t)]/\gamma_{aa}. \quad (2.42b)$$

Recalling the definitions of the operators $\mathcal{L}_0$, $\mathcal{L}_1$, and $\mathcal{L}_2$ and after some simplifications, we rewrite the master equation for $\bar{\chi}^{pq}$ in the following form:

$$
\begin{aligned}
d\bar{X}^{pq}/dt = & \, i \sum_k p_k[B_{kk},\bar{X}^{pq}] - a(p,q)\bar{X}^{pq} - \sum_k [g_k(p,q)+\zeta_k(t)]B_{kk}\bar{X}^{pq} \\
& - \sum_k [g_k(-p,-q)+\zeta_k(t)]\bar{X}^{pq}B_{kk} + 2\sum_{k,l}[e_{kl}+\xi_{kl}(t)]B_{kk}\bar{X}^{pq}B_{ll} \\
& + 2\sum_{\substack{k,l \\ (k\neq l)}} f_{kl}B_{kl}\bar{X}^{pq}B_{lk} + 2\sum_{\substack{k,l \\ (k\neq l\neq 1)}} g_{kl}[B_{1k}\bar{X}^{pq}B_{1l}+B_{k1}\bar{X}^{pq}B_{l1}]\delta_{\Delta_1,0}\delta_{\Delta_2,0} ,
\end{aligned} \tag{2.43}
$$

where the new time-dependent quantities $\zeta_k(t)$ and $\xi_{kl}(t)$ are defined as

$$\zeta_k(t) = a_k^2 b_k^2 \epsilon_1^2(t) + b_k^2 c_k^2 \epsilon_2^2(t) + a_k b_k^2 c_k \epsilon_{12}^2(t) , \tag{2.44}$$

$$
\begin{aligned}
\xi_{kl}(t) = & \, 2a_k b_k a_l b_l \epsilon_1^2(t) + 2b_k c_k b_l c_l \epsilon_2^2(t) \\
& + b_k b_l (a_k c_l + a_l c_k)\epsilon_{12}^2(t) = \xi_{lk}(t) .
\end{aligned} \tag{2.45}
$$

The master equation (2.43) has two remarkable properties. First, the equation describes a Markov process despite the fact that it contains time-dependent coefficients. The Markovian character essentially arises from the fact that the operator $[\epsilon_1^2(t)\mathcal{L}_1^2+\epsilon_2^2(t)\mathcal{L}_2^2+\epsilon_{12}^2(t)\mathcal{L}_1\mathcal{L}_2]$ commutes with $\mathcal{L}_0$ for all t. The Markovian property of (2.43) implies further that the quantum regression theorem is applicable. It will therefore be possible to derive two-time correlation functions from the one-time expectation values of the atomic operators. The second property of (2.43) is that it has a steady-state solution

$$
\begin{aligned}
& \bar{X}_{SS}^{pq} = 0 \quad (p,q\neq 0) , \\
& \bar{X}_{SS}^{00} = D^{-1}\exp[-(\mu_1 B_{11}+\mu_2 B_{22})] ,
\end{aligned} \tag{2.46}
$$

where the subscript SS denotes the steady state and μ_1 and μ_2 have the following expressions:

$$\mu_1 = \ln[(f_{11}f_{22}-f_{12}f_{21})/(f_{13}f_{22}+f_{12}f_{23})] , \tag{2.47}$$

$$\mu_2 = \ln[(f_{11}f_{22}-f_{12}f_{21})/(f_{11}f_{23}+f_{13}f_{21})] . \tag{2.48}$$

The quantities f_{ij} $(i\neq j)$ are defined in (2.39) while the constants f_{kk} $(k=1,2,3)$ are given by

$$
\begin{aligned}
f_{kk} = & \, \gamma_1 a_k^2(1-b_k^2) + \gamma_2 b_k^2(1-c_k^2) + \gamma_{c_1} a_k^2(1-a_k^2) \\
& + \gamma_{c_2}(1-c_k^2)c_k^2 + 2\gamma_{cc} a_k^2 c_k^2 .
\end{aligned} \tag{2.49}
$$

The normalization factor D is given by

$$
\begin{aligned}
D & = \mathrm{Tr}\{\exp[-(\mu_1 B_{11}+\mu_2 B_{22})]\} \\
& = 1 + \exp(-\mu_1) + \exp(-\mu_2) \\
& = [(f_{11}+f_{13})(f_{22}+f_{23}) \\
& \quad -(f_{12}-f_{13})(f_{21}-f_{23})]/(f_{11}f_{22}-f_{12}f_{21}) .
\end{aligned} \tag{2.50}
$$

We note here that under resonant conditions $\Delta_1=\Delta_2=0$, further simplification arises (cf. Appendix A).

III. FLUORESCENT SPECTRA AND INTENSITY CORRELATIONS

A. One-time atomic operator averages

The master equation (2.43) can be used directly to obtain the evolution of one-time atomic expectation values averaged over the ensembles of the phase and amplitude fluctuations. For an atomic operator $\hat{O}$, we define the average as

$$\langle \hat{O} \rangle_{pq} = \mathrm{Tr}(O\bar{X}^{pq}) . \tag{3.1}$$

The general equation of motion for $\langle B_{kl}\rangle_{pq}$ reads as

$$
\begin{aligned}
\frac{d}{dt}\langle B_{ij}\rangle_{pq} = & -i(p_i-p_j)\langle B_{ij}\rangle_{pq} - [g_j(p,q)+g_i(-p,-q)-2e_{ij}]\langle B_{ij}\rangle_{pq} - [\xi_{ij}(t)-\zeta_i(t)-\zeta_j(t)]\langle B_{ij}\rangle_{pq} \\
& + 2\sum_{\substack{k,l \\ (k\neq l)}} f_{kl}\langle B_{ll}\rangle_{pq}\delta_{ik}\delta_{jk} - a(p,q)\langle B_{ij}\rangle_{pq} + \left[\left[\sum_{\substack{k,i \\ (k\neq i\neq 1)}} g_{ki}\langle B_{1k}\rangle_{pq} \right]\delta_{j1} + \left[\sum_{\substack{l,j \\ (l\neq j\neq 1)}} g_{jl}\langle B_{l1}\rangle_{pq} \right]\delta_{i1} \right]\delta_{\Delta_1,0}\delta_{\Delta_2,0} .
\end{aligned} \tag{3.2}
$$

In particular, when $i=j$ we have the three coupled equations

$$
\begin{aligned}
d\langle B_{11}\rangle_{pq}/dt = & -[2f_{11}+a(p,q)]\langle B_{11}\rangle_{pq} \\
& + 2f_{12}\langle B_{22}\rangle_{pq} + 2f_{13}\langle B_{33}\rangle_{pq} ,
\end{aligned} \tag{3.3a}
$$

$$
\begin{aligned}
d\langle B_{22}\rangle_{pq}/dt = & \, 2f_{21}\langle B_{11}\rangle_{pq} - [2f_{22}+a(p,q)]\langle B_{22}\rangle_{pq} \\
& + 2f_{23}\langle B_{33}\rangle_{pq} ,
\end{aligned} \tag{3.3b}
$$

$$
\begin{aligned}
d\langle B_{33}\rangle_{pq}/dt = & \, 2f_{31}\langle B_{11}\rangle_{pq} + 2f_{32}\langle B_{22}\rangle_{pq} \\
& - [2f_{33}+a(p,q)]\langle B_{33}\rangle_{pq} ,
\end{aligned} \tag{3.3c}
$$

and for $i\neq j$ we obtain the six equations

$$
\begin{aligned}
\frac{d}{dt}\langle B_{ij}\rangle_{pq} = & -[i\Omega_{ij} + \Gamma_{ij}(p,q) + \xi_{ij}(t) \\
& - \zeta_i(t) - \zeta_j(t)]\langle B_{ij}\rangle_{pq} \\
& + \left[\left[\sum_{\substack{k \\ (k\neq i\neq 1)}} g_{ki}\langle B_{1k}\rangle_{pq} \right]\delta_{j1} \right. \\
& \left. + \sum_{\substack{l \\ (l\neq j\neq 1)}} (g_{jl}\langle B_{l1}\rangle_{pq})\delta_{i1} \right]\delta_{\Delta_1,0}\delta_{\Delta_2,0} ,
\end{aligned} \tag{3.4}
$$

where f_{kk}, f_{ij} $(i \neq j)$ are defined as in (2.49) and (2.39), respectively, and

$$\Omega_{ij} = p_i - p_j, \quad i \neq j \tag{3.5}$$

$$\Gamma_{ij}(p,q) = [g_j(p,q) + g_i(-p,-q) + a(p,q) - 2e_{ij}] = \Gamma_{ji}(-p,-q) . \tag{3.6}$$

Note that the evolution of the diagonal operators $\langle B_{ij}(t)\rangle_{pq}$ does not depend on the amplitude fluctuations at least in the strong-field limit. This is indeed a consequence of our approximate treatment of the amplitude fluctuations, viz., that the stochastic averaging over the ensemble of amplitude fluctuation was carried out only after invoking the secular approximation. Also, as mentioned before, the additional terms in the master equation do not contribute to the evolution of $\langle B_{ii}(t)\rangle_{pq}$. The three coupled equations can be solved readily and the solution can be written in the compact form

$$\langle B_{ii}\rangle_{pq} = \Bigg[\frac{g_i}{\nu_1\nu_2}\langle B(0)\rangle_{pq} + \frac{1}{\nu_1(\nu_1-\nu_2)}\Bigg[(\nu_1^2 - 2h_i\nu_1 + g_i)\langle B_{ii}(0)\rangle_{pq} + \sum_{\substack{j \\ (j\neq i)}} (-2f_{ij}\nu_1 + g_i)\langle B_{jj}(0)\rangle_{pq}\Bigg]\exp(-\nu_1 t) + \frac{1}{\nu_2(\nu_2-\nu_1)}\Bigg[(\nu_2^2 - 2h_i\nu_2 + g_i)\langle B_{ii}(0)\rangle_{pq} + \sum_{\substack{j \\ (j\neq i)}} (-2f_{ij}\nu_2 + g_i)\langle B_{jj}(0)\rangle_{pq}\Bigg]\exp(-\nu_2 t)\Bigg]\exp[-a(p,q)t] , \tag{3.7}$$

where

$$\langle B(0)\rangle_{pq} = \sum_{i=1}^{3} \langle B_{ii}(0)\rangle_{pq} , \tag{3.8}$$

$$h_1 = f_{13} + f_{22} + f_{23} , \tag{3.9a}$$

$$h_2 = f_{11} + f_{13} + f_{23} , \tag{3.9b}$$

$$h_3 = f_{11} + f_{22} , \tag{3.9c}$$

$$g_1 = 4(f_{13}f_{22} + f_{23}f_{12}) , \tag{3.10a}$$

$$g_2 = 4(f_{11}f_{23} + f_{13}f_{21}) , \tag{3.10b}$$

$$g_3 = 4(f_{11}f_{22} - f_{12}f_{21}) , \tag{3.10c}$$

and ν_1, ν_2 are the two roots of the quadratic equation

$$\nu^2 - (h_1 + h_2 + h_3)\nu + (g_1 + g_2 + g_3) = 0 . \tag{3.11}$$

In the case when Δ_1 and Δ_2 are not simultaneously zero, the equations for $\langle B_{ij}(t)\rangle_{pq}$ are not coupled and can be solved individually. The solution for the off-diagonal operator averages therefore reads as

$$\langle B_{ij}(t)\rangle_{pq} = \langle B_{ij}(0)\rangle_{pq} \exp\left[-\beta_{ij}t + \sum_{k=1}^{3} \delta_{ij}^{(k)}(t)\chi_k(t)\right] \times \exp\{-[i\Omega_{ij} + \Gamma_{ij}(p,q)]t\} , \tag{3.12}$$

where the coefficients $\beta_{ij}, \delta_{ij}^{(k)}$ and the time-dependent functions $\chi_k(t)$ have the following meaning:

$$\beta_{ij} = \sum_{k=1}^{3} \beta_{ij}^{(k)} , \tag{3.13}$$

$$\delta_{ij}^{(k)} = \beta_{ij}^{(k)}/\gamma_{a_k} \quad (k=1,2) , \tag{3.14a}$$

$$\delta_{ij}^{(3)} = \beta_{ij}^{(3)}/\gamma_{aa} , \tag{3.14b}$$

$$\beta_{ij}^{(1)} = (a_ib_i - a_jb_j)^2 d_1^2\epsilon_1^2/\gamma_{a_1} , \tag{3.15a}$$

$$\beta_{ij}^{(2)} = (b_ic_i - b_jc_j)^2 d_2^2\epsilon_2^2/\gamma_{a_2} , \tag{3.15b}$$

$$\beta_{ij}^{(3)} = (a_ib_i - a_jb_j)(b_ic_i - b_jc_j)d_1d_2\epsilon_{12}^2/\gamma_{aa} , \tag{3.15c}$$

$$\chi_k(t) = [1 - \exp(-\gamma_{a_k}t)] \quad (k=1,2) , \tag{3.16a}$$

$$\chi_3(t) = [1 - \exp(-\gamma_{aa}t)] . \tag{3.16b}$$

For future use, it is more convenient to express the above solution in an alternative form obtained by expanding the $\chi_k(t)$ in a series. Thus

$$\langle B_{ij}(t)\rangle_{pq} = \langle B_{ij}(0)\rangle_{pq} \exp(\delta_{ij}) \times \sum_{\{k\}} \Delta_{ij}(\{k\}) \exp(-[i\Omega_{ij} + \Gamma_{ij}(p,q) + \lambda(\{k\}) + \beta_{ij}]t) , \tag{3.17}$$

where the symbol $\{k\}$ implies that the summation in (3.17) is over triplet of integers k_1, k_2, k_3 each ranging from zero to infinity. The other notation is as follows:

$$\Delta_{ij}(\{k\}) = \prod_{l=1}^{3} \frac{(-\delta_{ij}^{(l)})^{k_j}}{k_j!} , \tag{3.18}$$

$$\delta_{ij} = \sum_{l=1}^{3} \delta_{ij}^{(l)}, \quad \lambda \equiv \lambda(\{k\}) = k_1\gamma_{a_1} + k_2\gamma_{a_2} + k_3\gamma_{aa} . \tag{3.19}$$

It is clear that the amplitude fluctuations affect the evolution of the off-diagonal atomic-operator averages considerably.

In the special case of resonant excitations, $\Delta_1 = \Delta_2 = 0$, there is a coupling between the expectation values $\langle B_{12}(t)\rangle_{pq}$ and $\langle B_{31}(t)\rangle_{pq}$ and also between $\langle B_{21}(t)\rangle_{pq}$ and $\langle B_{13}(t)\rangle_{pq}$. The equations for $\langle B_{23}(t)\rangle_{pq}$ and $\langle B_{32}(t)\rangle_{pq}$ are, however, uncoupled. The coupling between $\langle B_{12}(t)\rangle_{pq}$ and $\langle B_{31}(t)\rangle_{pq}$ is to be expected because under resonant conditions, $B_{12}(t)$ and $B_{31}(t)$ have the same behavior in time according to (2.30). The explicit solutions of these equations of motion for the atomic-operator averages are displayed in Appendix A.

B. Fluorescent spectra in the steady state

We now use the one-time atomic-operator averages to compute the steady-state spectra of spontaneous emission from the upper and lower excited levels. We denote the positive and negative frequency parts of the atomic spontaneous transitions from the level $|1\rangle$ to $|2\rangle$ and from level $|2\rangle$ to $|3\rangle$ by $E_1^{(+)}$ and $E_2^{(+)}$, respectively. The spectra are then related to the Fourier transform of the two-time correlation function $\langle \overline{\langle E_i^{-}(t)E_i^{*}(t+\tau)\rangle}\rangle_a$ where $i=1,2$; the bar denotes the average over the phase and the second angular bracket $\langle\ \rangle_a$ stands for averaging over the amplitude fluctuations. As shown by Agarwal[25] and others, if the measurements are carried out in a manner such that the incident laser fields do not contribute, the operators $E_i^{(+)}$ are related to the atomic operators by

$$E_1^{(+)}(\mathbf{r},t)\to \mathbf{k}_1 A_{21}(t)\ ,$$

$$E_2^{(+)}(\mathbf{r},t)\to \mathbf{k}_2 A_{31}(t)\ ,$$

where $\mathbf{k}_1$ and $\mathbf{k}_2$ are factors which depend on the spatial dimension $\mathbf{r}$ and the dipole moments d_1 and d_2. With this we can define the steady-state spectra as

$$G_{1,2}(\omega)=\mathrm{Re}\int_0^\infty \exp[-i(\omega-\Omega_{1,2})\tau]G_{1,2}^{(1)}(\tau)d\tau\ , \qquad (3.20)$$

where

$$G_1^{(1)}(\tau)=\langle\langle \overline{A_{12}(\tau)A_{21}}\rangle_{\mathrm{SS}}\rangle_a\ , \qquad (3.21)$$

$$G_2^{(1)}(\tau)=\langle\langle \overline{A_{23}(\tau)A_{32}}\rangle_{\mathrm{SS}}\rangle_a\ . \qquad (3.22)$$

We note that

$$\langle\langle \overline{A_{12}(\tau)}\rangle\rangle_a=\langle A_{12}(\tau)\rangle_{10} =\sum_{i,j} a_i b_j\langle B_{ij}(\tau)\rangle_{10}\ , \qquad (3.23)$$

$$\langle\langle \overline{A_{23}(\tau)}\rangle\rangle_a=\sum_{i,j} b_i c_j\langle B_{ij}(\tau)\rangle_{01}\ , \qquad (3.24)$$

where the solutions for the expectation values $\langle B_{ij}(t)\rangle_{10}$ and $\langle B_{ij}(t)\rangle_{01}$ can be written down from Eq. (3.7) and (3.17). Expressing A_{21} also in terms of B_{ij} and applying the quantum regression theorem, we can compute all expectation values of the type $\langle B_{ij}(t)B_{kl}\rangle_{10}$ and $\langle B_{ij}(t)B_{kl}\rangle_{01}$ and obtain $G_{1,2}(\omega)$. Explicit expressions for these spectra read as

$$G_j(\omega)=\frac{A_j}{(\omega-\Omega_j)^2+\gamma_{c_j}^2}+\frac{B_j(\nu_1)}{(\omega-\Omega_j)^2+(\gamma_{c_j}+\nu_1)^2}+\frac{B_j(\nu_2)}{(\omega-\Omega_j)^2+(\gamma_{c_j}+\nu_2)^2} + \sum_{\substack{k,l\\(k\neq l)}}^{3} D_{kl}^{(j)}\langle B_{kk}\rangle_{00}\sum_{\{k\}}\frac{[\Gamma_{kl}^{(j)}+\lambda(\{k\})+\beta_{kl}]\Delta_{kl}(\{k\})}{(\omega-\Omega_j+\Omega_{kl})^2+[\Gamma_{kl}^{(j)}+\lambda(\{k\})+\beta_{kl}]^2}\ , \qquad (3.25)$$

where $j=1,2$ corresponds to the spectra of emission from the upper and lower excited levels, respectively, and the other notations are as follows:

$$A_1=\gamma_{c_1}\left[\sum_i a_i b_i\langle B_{ii}\rangle_{00}\right]^2\ , \qquad (3.26)$$

$$\langle B_{ii}\rangle_{00}=g_i/\nu_1\nu_2\ , \qquad (3.27)$$

$$B_1(\nu)=\frac{\gamma_{c_j}+\nu}{\nu^2-\nu_1\nu_2}\sum_i a_i b_i Q_{ab}^{(i)}(\nu)\langle B_{ii}\rangle_{00}\ , \qquad (3.28)$$

$$Q_{ab}^{(i)}(\nu)=a_i b_i\nu^2-2\nu\Big(a_i b_i h_i+\sum_{\substack{i,j\\(j\neq i)}} a_j b_j f_{ji}\Big)+\sum_i a_i b_i g_i\ , \qquad (3.29)$$

$$D_{kl}^{(1)}=a_k^2 b_l^2,\quad D_{kl}^{(2)}=b_k^2 c_l^2\ , \qquad (3.30)$$

$$\Gamma_{kl}^{(1)}=\Gamma_{kl}(1,0),\quad \Gamma_{kl}^{(2)}=\Gamma_{kl}(0,1)\ . \qquad (3.31)$$

$A_2, B_2(\nu)$ are to be obtained from A_1 and $B_1(\nu)$ by replacing therein a_i by b_i and b_i by c_i. The other quantities ν_i, h_i, f_{ji}, and g_i have the same meaning as before.

The expression for the fluorescent spectra under resonance conditions $\Delta_1=\Delta_2=0$ is derived in Appendix A [cf. Eq. (A35)].

The first term in the expression (3.25) for the spectra represents the coherent peak at the applied laser frequency Ω_j. The width of this peak clearly depends only on the phase bandwidth parameter γ_{c_j} while its height depends on all parameters γ_{c_1}, γ_{c_2}, and γ_{cc}. The remaining terms in the expression (3.25) show the nature of the incoherent spectra. In order to highlight the effects due to detunings, let us momentarily ignore the laser-bandwidth effects due to fluctuations by setting all the parameters related to the phase and amplitude fluctuations as zeros. This has an obvious consequence that terms corresponding to $k_1=k_2=k_3=0$ alone survive in the summation over $\{k\}$ on the right-hand side of Eq. (3.25). The incoherent spectra, in the absence of fluctuations, are in general asymmetric around the center $\omega=\Omega_j$ and consist of as many as seven peaks. These peaks correspond to the central Lorentzian located at the excitation frequency and three pairs of side bands located at $\omega=\Omega_j\pm\Omega_{12}$, $\omega=\Omega_j\pm\Omega_{23}$ and $\omega=\Omega_j\pm\Omega_{31}$ $(j=1,2)$. However, the resolution of these peaks depend on the eigenvalues λ_i of the cubic equation (2.23) which in turn depends on the values of α_i and Δ_i and one may have spectra varying from five to seven peaks. Note that the central peak at $\omega=\Omega_j$ is superposition of two Lorentzians of unequal widths and heights. This is in contrast with the resonance case where, as is clear from Eq. (A35) in Appendix A, the spectra are symmetric around the center ($\omega=\Omega_j$) and have five peaks (instead of seven), located at $\omega=\Omega_j$, $\omega=\Omega_j\pm\Omega$, and $\omega=\Omega_j\pm2\Omega$ where $\Omega=(\alpha_1^2+\alpha_2^2)^{1/2}$. Note also that the central peak is a single Lorentzian and not a superposition of two Lorentzians. We may also remark here that under the optical double-resonance conditions $\Delta_1=-\Delta_2=\Delta$, the spectra show five peaks located at $\omega=\Omega_j$, $\omega=\Omega_j+(\Delta_j/2-\Gamma)$, $\omega=\Omega_1+(\Gamma+\Delta_j/2)$ and $\omega=\Omega_j\pm2\Gamma$, and $\Gamma=[\Omega^2+(\Delta/2)^2]^{1/2}$. The important feature here is the asymmetry of the location of the near side bands. This together with the asymmetric heights and widths of the side peaks and the two-component nature of the central peak distinguishes this case from the case where Δ_1 and Δ_2 are both zero.

We discuss next the effects due to phase fluctuations by ignoring the contributions due to amplitude fluctuations in Eq. (3.25). We may first consider the case when the two driving fields have opposite detunings ($\Delta_1=-\Delta_2$). In this case, in the absence of phase fluctuations $\gamma_{c_1}=\gamma_{c_2}=\gamma_{cc}=0$, both the upper and lower spectra show five distinct peaks with asymmetry around the applied field frequency. Figure 2 shows the essential effects due to the phase parameters γ_{c_1} and γ_{c_2} on the two spectra when the cross correlations between the driving fields are absent ($\gamma_{cc}=0$). The major effect of phase fluctuations γ_{c_1} and γ_{c_2} is to suppress the central peak as well as the remote sidebands. The upper spectrum responds more to the variation in γ_{c_1} rather than γ_{c_2} (curves D and E in Fig. 2) and vice versa for the lower spectrum. As the bandwidth $\gamma_{c_1},\gamma_{c_2}$ are increased we expect that the Stark quintuplets reduce to the Stark doublet in complete agreement with the purely numerical results of Ref. 17.

On the other hand, for a given set of the parameters γ_{c_1} and γ_{c_2}, an increase in the cross-correlation bandwidth γ_{cc} leads to a suppression of the near side bands with a slight enhancement of the central peak and the remote side bands. This is reflected in curves A–E in Fig. 3 where the spectra are shown for several values of γ_{cc} and a fixed set of parameters γ_{c_j}. These results are similar to those in the resonance case. In the latter case, however, there is another important feature shown by the analytical spectra in the presence of phase fluctuations, namely, that each of the near side bands at $\omega=\Omega_j\pm\Omega$ arises from a superposition of two Lorentzians of unequal widths [cf. Eq. (A35) in Appendix A].

In the general case of unequal detunings ($\Delta_1\neq\Delta_2$), the effects of phase fluctuations on the individual peaks of the spectrum can be more subtle. As an example of this we consider a case in which one of the driving fields is in exact resonance with the corresponding atomic transition while the other field has a finite detuning ($\Delta_1=0$, $\Delta_2=-20$). In the absence of fluctuations, the upper spectrum [curve A in Fig. 4(a)] shows seven distinct peaks while the lower spectrum [curve A in Fig. 4(b)]

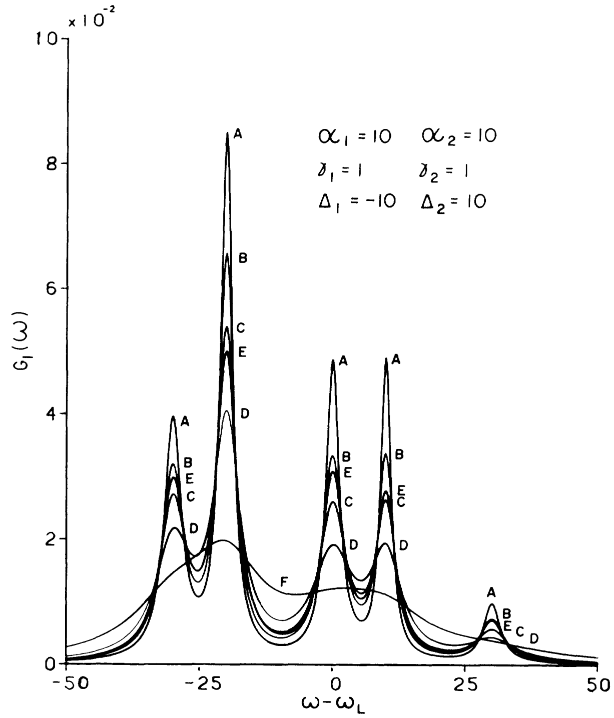

FIG. 2. Effect of phase fluctuations on the upper fluorescent spectra when the driving fields have equal and opposite detunings. Curves A–D correspond to $\gamma_{c_2}=0$ and $\gamma_{c_1}=0$, 0.5, 1, 2, respectively. Curve E represents $\gamma_{c_1}=0$ and $\gamma_{c_2}=2$, while F is for $\gamma_{c_1}=\gamma_{c_2}=5$. In all these curves $\gamma_{cc}=0$.

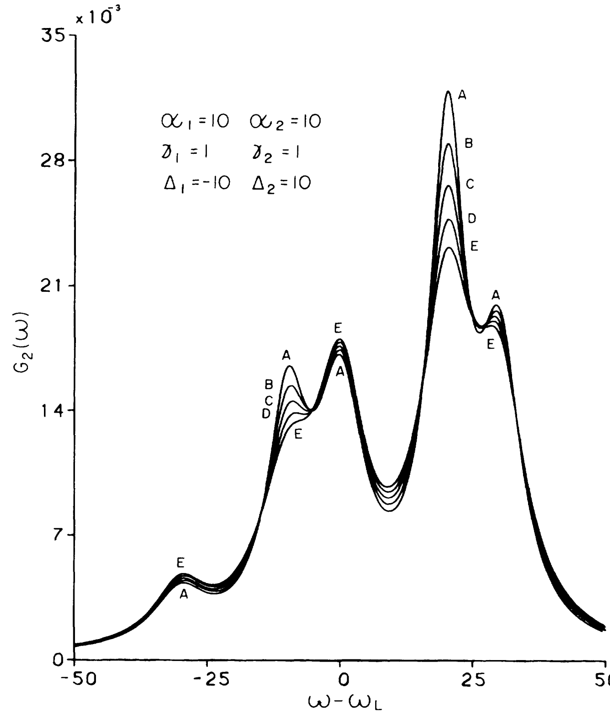

FIG. 3. Effect of phase cross correlations on the lower fluorescent spectra when the driving fields have equal and opposite detunings. Curves A–E correspond to $\gamma_{c_1}=\gamma_{c_2}=2$ and $\gamma_{cc}=0$, 0.5, 1, 1.5, and 2, respectively.

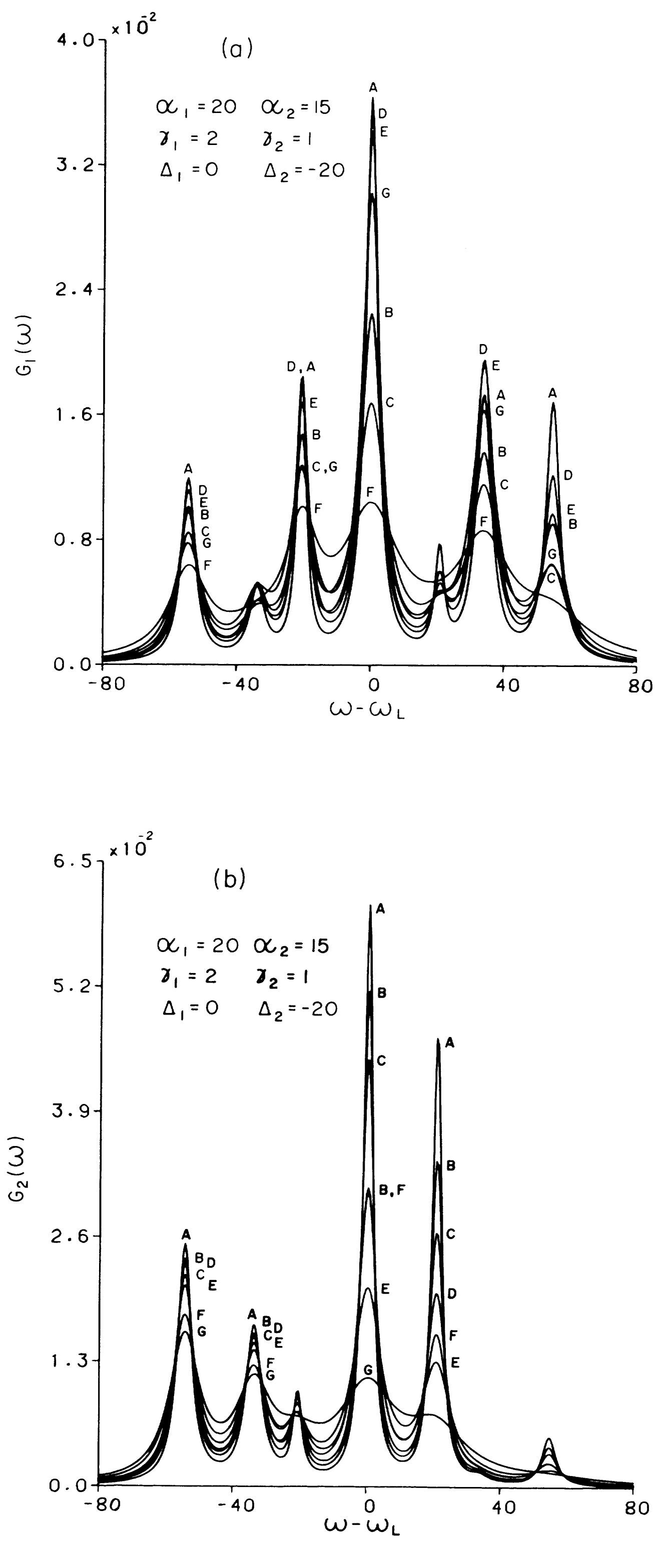

FIG. 4. Effect of phase fluctuations on the fluorescent spectra when the upper field is in exact resonance and lower field is off resonance. (a) Upper spectrum and (b) lower spectrum. Curves $A-G$ correspond to $\gamma_{cc}=0$ and $(\gamma_{c_1},\gamma_{c_2})$ are (0,0), (1,0), (2,0), (0,1), (0,2), (5,0), and (0,5), respectively.

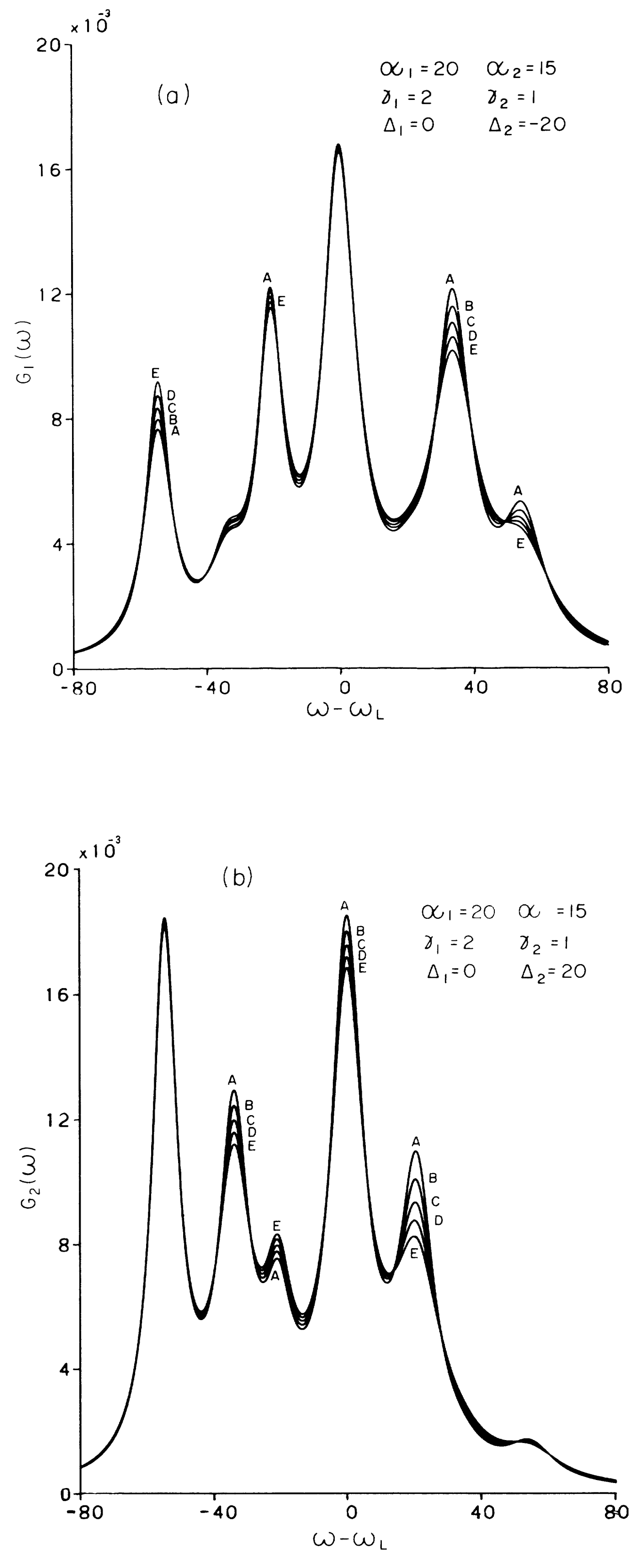

FIG. 5. Effect of phase cross correlations on the fluorescent spectra when the upper field is in exact resonance and lower field is off resonance. (a) Upper spectra and (b) lower spectra. Other data as in Fig. 3.

shows only six resolved peaks. The curves $B-F$ in Fig. 4 show the variation of the spectra for several values of the parameters γ_{c_1} and γ_{c_2} with $\gamma_{cc}=0$. The overall effect of increasing γ_{c_1} and/or γ_{c_2} is the suppression of the central and side peaks, as is to be expected. The upper spectrum is more sensitive to the changes in γ_{c_1} values rather than γ_{c_2} and vice versa for the lower spectrum. Note, however, that for intermediate values of γ_{c_2} (with γ_{c_1} fixed) the second side band peaks have a tendency to get enhanced before getting suppressed or smoothened out for very large values of γ_{c_2}. Similar effects are also seen in Fig. 5 where the effects of cross correlations γ_{cc} on the upper and lower spectra are shown for a fixed set of γ_{c_j} values. In Fig. 5(a) for the upper spectrum one sees the enhancement of the two remote side peaks on the left and suppression of all other peaks in the upper spectrum with increasing values of γ_{cc}. On the other hand, the lower spectrum shown in Fig. 5(b) shows the suppression of all peaks except the first peak at the left as γ_{cc} is increased.

Finally, we consider the full expression (3.25) for the fluorescent spectra from the upper and lower excited levels which contain the effects due to both phase and amplitude fluctuations. The analytical spectra are asymmetric with a central peak at $\omega=\Omega_j$ and three pairs of side bands at $\omega=\Omega_j\pm\Omega_{12}$, $\omega=\Omega_j\pm\Omega_{23}$, and $\omega=\Omega_j\pm\Omega_{31}$. The contributions to the effective height and width of each of these side bands come from a superposition of an infinite number of Lorentzians located at the side-band center with heights and widths depending on the parameters $\lambda(\{k\})$ (with $k_1,k_2,k_3=0,1,2,\ldots$). It is also clear from the analytical expression (3.35) that unlike the phase fluctuations which affect both the central peaks as well as the side bands the amplitude fluctuations do not show any effect on the central peak at all and affect only the side bands. This can be attributed to the approximations made in our formulation. We have carried out the stochastic averaging over the ensembles of phases exactly in our formulation. On the other hand, the averaging over the ensembles of amplitude fluctuations could be carried out only after taking a recourse to the high-field approximation. The general effect of the amplitude fluctuations is as shown in Fig. 6 for a typical set of detuning parameters. For a fixed set of phase-bandwidth parameters $\gamma_{c_1},\gamma_{c_2},\gamma_{cc}$ and a fixed set of the amplitude fluctuation parameters $d_j^2\epsilon_j^2$ and $d_1d_2\epsilon_{12}^2$, the side peaks show a slight increase in the height and reduction in width with an increase in any of the amplitude-bandwidth parameters γ_{a_1} or γ_{aa} [curve E in Figs. 6(a) and (b)]. On the other hand, it is seen from the analytical expression (3.25) and the curves $A-D$ of Fig. 6 that given a set of phase parameters γ_{c_1}, γ_{c_2}, and γ_{cc} and amplitude-bandwidth parameters γ_{a_1}, γ_{a_2}, and γ_{aa} an increase in any of the amplitude-fluctuation parameters $d_j\epsilon_j/\gamma_{a_1}$ or $d_1d_2\epsilon_{12}/\gamma_{aa}$ results in the reduction of height and a broadening of the side bands.

C. Intensity-intensity correlation functions

The nature of the fluorescent light emitted from the excited levels $|1\rangle$ and $|2\rangle$ may be known from the second-order intensity correlation functions. The quantum-mechanical first and second-order correlation functions are defined by

$$\Gamma_{ij}(t)=\langle\langle \overline{E_i^-(r,t)E_j^+(r,t)}\rangle\rangle_a , \quad (3.32)$$

$$\Gamma_{ij}(t,t+\tau)=\langle\langle \overline{E_i^-(r,t)E_j^-(t,t+\tau)E_j^+(r,t+\tau)E_i^-(r,t)}\rangle\rangle_a . \quad (3.33)$$

We are interested, in particular, in the normalized intensity-intensity correlations in the steady state defined by

$$g_{ij}(\tau)=\lim_{t\to\infty}\frac{\Gamma_{ij}(t,t+\tau)}{\Gamma_{ii}(t)\Gamma_{jj}(t)} . \quad (3.34)$$

More explicitly, this expression results in four kinds of second-order coherence functions

$$\begin{aligned} g_{11}^{(2)}(\tau)&=\langle\langle \overline{A_{12}A_{12}(\tau)A_{21}(\tau)A_{21}}\rangle_{SS}\rangle_a / |\langle\langle \overline{A_{12}A_{21}}\rangle\rangle_a|^2 \\ &=\langle\langle A_{12}A_{11}(\tau)A_{21}\rangle\rangle_{00}/|\langle A_{11}\rangle_{00}|^2 , \end{aligned} \quad (3.35)$$

$$\begin{aligned} g_{22}^{(2)}(\tau)&=\langle\langle \overline{A_{23}A_{23}(\tau)A_{32}(\tau)A_{32}}\rangle_{SS}\rangle_a / |\langle\langle \overline{A_{23}A_{32}}\rangle\rangle_a|^2 \\ &=\langle A_{23}A_{22}(\tau)A_{32}\rangle_{00}/|\langle A_{22}\rangle_{00}|^2 , \end{aligned} \quad (3.36)$$

$$\begin{aligned} g_{12}^{(2)}(\tau)&=\langle\langle \overline{A_{12}A_{23}(\tau)A_{32}(\tau)A_{21}}\rangle_{SS}\rangle_a / |\langle\langle \overline{A_{23}A_{32}}\rangle\rangle_a\langle\langle \overline{A_{23}A_{32}}\rangle\langle_a| \\ &=\langle A_{12}A_{22}(\tau)A_{21}\rangle_{00}/|\langle A_{22}\rangle_{00}\langle A_{11}\rangle_{00}| , \end{aligned} \quad (3.37)$$

$$\begin{aligned} g_{21}^{(2)}(\tau)&=\langle\langle \overline{A_{23}A_{12}(\tau)A_{21}(\tau)A_{32}}\rangle_{SS}\rangle_a / |\langle\langle \overline{A_{23}A_{32}}\rangle\rangle_a\langle\langle \overline{A_{23}A_{32}}\rangle\rangle_a| \\ &=\langle A_{23}A_{11}(\tau)A_{32}\rangle_{00}/|\langle A_{22}\rangle_{00}\langle A_{11}\rangle_{00}| . \end{aligned} \quad (3.38)$$

The quantity $g_{11}^{(2)}(\tau)$ ($[g_{22}^{(2)}(\tau)]$ is a measure of the correlations between the photons emitted at time $t=0$ and at time $t=\tau$ from the upper (lower) excitation level. On the other hand, $g_{12}^{(2)}(\tau)$ $[g_{21}^{(2)}(\tau)]$ is a measure of the probability of detecting a phonon emitted from the upper (lower) excitation level at time $t=0$ and another photon emitted from the lower (upper) excitation level at a later time $t=\tau$.

In order to obtain these correlation functions we have to obtain first the expectation values $\langle A_{11}(\tau)\rangle_{00}$ and $\langle A_{22}(\tau)\rangle_{00}$ and then apply the quantum regression theorem. The expectation values $\langle A_{11}(\tau)\rangle_{00}$ and $\langle A_{22}(\tau)\rangle_{00}$ are in turn obtained by first expressing the A_{11} and A_{22} operators in terms of the operators B_{ij} according to Eqs. (2.28) and subsequently employing the solutions for the expectation values $\langle B_{ij}(\tau)\rangle$. The resulting expression for $g_{ij}^{(2)}(\tau)$ may be written in the following compact form:

$$\begin{aligned} g_{ij}^{(2)}(\tau)=1&+\frac{\nu_2}{\nu_1-\nu_2}P_{ij}(\nu_1)\exp(-\nu_1\tau)+\frac{\nu_1}{\nu_2-\nu_1}P_{ij}(\nu_2)\exp(-\nu_2\tau) \\ &+R_{ij}\exp\{-[\beta_{12}+\Gamma_{12}(0,0)\tau]\}\sum_{\{k\}}\Delta_{12}(\{k\})\exp[-\lambda(\{k\})\tau]\cos(\Omega_{12}\tau) \\ &+S_{ij}\exp\{-[\beta_{23}+\Gamma_{23}(0,0)]\tau\}\sum_{\{k\}}\Delta_{23}(\{k\})\exp[-\lambda(\{k\})\tau]\cos(\Omega_{23}\tau) \\ &+T_{ij}\exp\{-[\beta_{31}+\Gamma_{31}(0,0)]\tau\}\sum_{\{k\}}\Delta_{31}(\{k\})\exp[-\lambda(\{k\})\tau]\cos(\Omega_{31}\tau)\ . \end{aligned} \tag{3.39}$$

The coefficients $P_{ij}(\nu)$, R_{ij}, S_{ij}, and T_{ij} are to be defined as follows. We first define the four related quantities

$$P(x,y;\nu)=\frac{\left[\sum_k x_k^2y_k^2\right]\nu^2-2\nu\left[\sum_k x_k^2y_k^2h_k+\sum_{k\neq l}x_k^2y_k^2f_{kl}\right]+\sum_k x_k^2g_k}{\sum_k x_k^2g_k}\ , \tag{3.40}$$

$$R(x,y)=2\nu_1\nu_2(x_1x_2y_1y_2)\exp(\delta_{12})\Big/\sum_k x_k^2g_k\ , \tag{3.41}$$

$$S(x,y)=2\nu_1\nu_2(x_2x_3y_2y_3)\exp(\delta_{23})\Big/\sum_k x_k^2g_k\ , \tag{3.42}$$

$$T(x,y)=2\nu_1\nu_2(x_1x_3y_1y_3)\exp(\delta_{13})\Big/\sum_k x_k^2g_k\ . \tag{3.43}$$

With this definition $P_{11}(\nu)$, R_{11}, S_{11}, T_{11} are obtained from the above expressions by letting $x_i=a_i$ and $y_i=b_i$; $P_{22}(\nu)$, R_{22}, S_{22}, T_{22} are obtained from the corresponding expressions by letting $x_i=b_i$ and $y_i=c_i$; $P_{12}(\nu)$, R_{12}, S_{12}, T_{12} are similarly obtained by letting $x_i=y_i=b_i$. Finally $P_{21}(\nu)$, R_{21}, S_{21}, T_{21} are obtained by replacing x_i by a_i and y_i by c_i.

It is clear from these expressions that the correlation functions $g_{11}^{(2)}(\tau)$, $g_{22}^{(2)}(\tau)$, and $g_{21}^{(2)}(\tau)$ vanish at $\tau=0$ and tend towards unity for long times τ. On the other hand, the function $g_{12}^{(2)}(\tau)$ does not vanish at $\tau=0$, implying thereby that there is finite probability for simultaneous emission of two photons of frequency Ω_1 and frequency Ω_2. In the absence of fluctuations all $g_{ij}^{(2)}(\tau)$ show the expected oscillatory behavior through bunching and antibunching cycles decaying to their steady-state value of unity. The phase and/or amplitude fluctuations do not change their qualitative behavior but merely tend to reduce the amplitude of oscillations. This is shown in Figs. 7 and 8 where $g_{ij}^{(2)}(\tau)$ are plotted versus τ for some typical set of fluctuation parameters. Note that in Figs. 7 and 8 the curves $A-E$ for different fluctuation parameters are distinguished by labeling only a typical peak. The behavior at other peaks follow the same pattern. We may remark here that the effects of phase (amplitude) cross correlations between the lasers γ_{cc} (γ_{aa}) on the behavior of $g_{ij}^{(2)}(\tau)$ are not distinguishable from those of the self-correlations γ_{c_1} (γ_{a_1}) and γ_{c_2} (γ_{a_2}). This is in contrast with the case of fluorescent spectra discussed previously where such effects are clearly discernible. Also, in the case of amplitude fluctuations, the curves $A-E$ Of Fig. 8 indicate that the intensity-intensity correlations are relatively less sensitive to the variation in the bandwidth parameters γ_{a_1}, γ_{a_2}, and γ_{aa} as com-

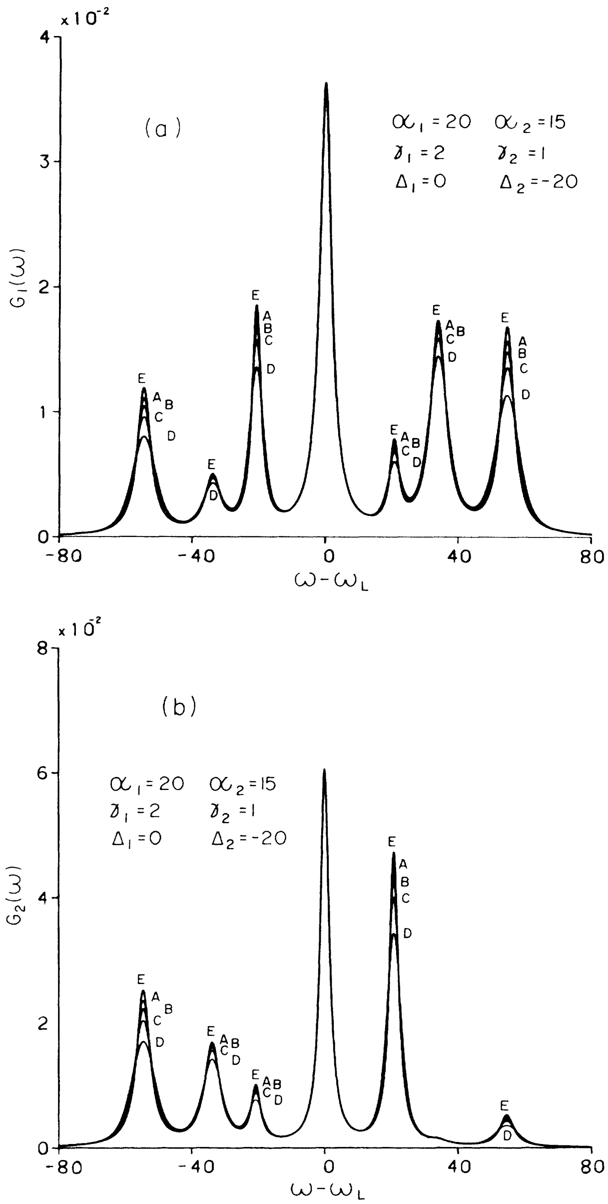

FIG. 6. Effect of amplitude fluctuations on the fluorescent spectra for (a) upper transition and (b) lower transition. Curves A–D correspond to $\gamma_{a_1} = \gamma_{a_2} = \gamma_{aa} = 0.5$ and $(\epsilon_1^2, \epsilon_2^2, \epsilon_{12}^2)$ are (0.5,0.5,0.5), (1,1,1), (2,2,2), and (3,3,3) while E is for $\gamma_{a_1} = \gamma_{a_2} = \gamma_{aa} = 3$ and $\epsilon_1^2 = \epsilon_2^2 = \epsilon_{12}^2 = 0.5$. In all these curves the phase fluctuations are absent, i.e., $\gamma_{c_1} = \gamma_{c_2} = \gamma_{cc} = 0$.

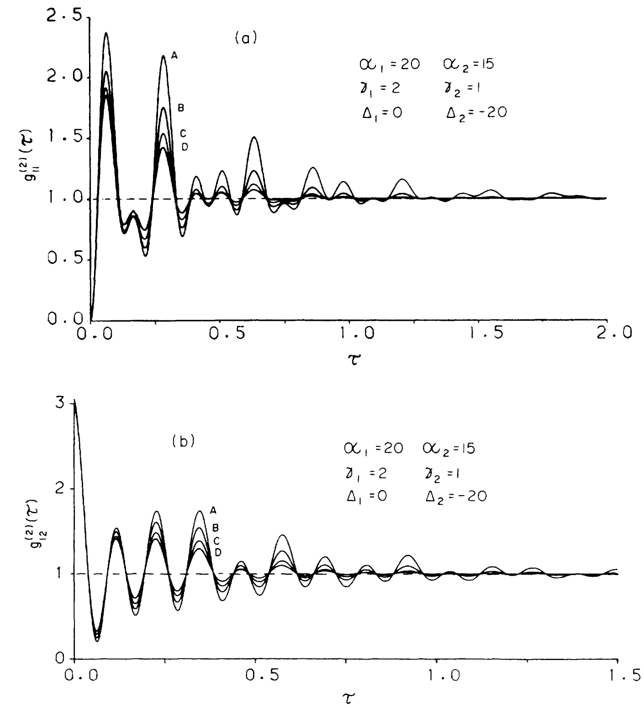

FIG. 7. Effect of phase fluctuations on the intensity-intensity correlation functions. (a) $g_{11}^{(2)}(\tau)$, (b) $g_{12}^{(2)}(\tau)$. Curves A–C correspond to $\gamma_{cc} = 0$ and $\gamma_{c_1} = \gamma_{c_2} = 0, 1, 2$, while curve D is for $\gamma_{c_1} = \gamma_{c_2} = \gamma_{cc} = 2$. For simplicity only one peak is labeled. The behavior at other peaks is similar.

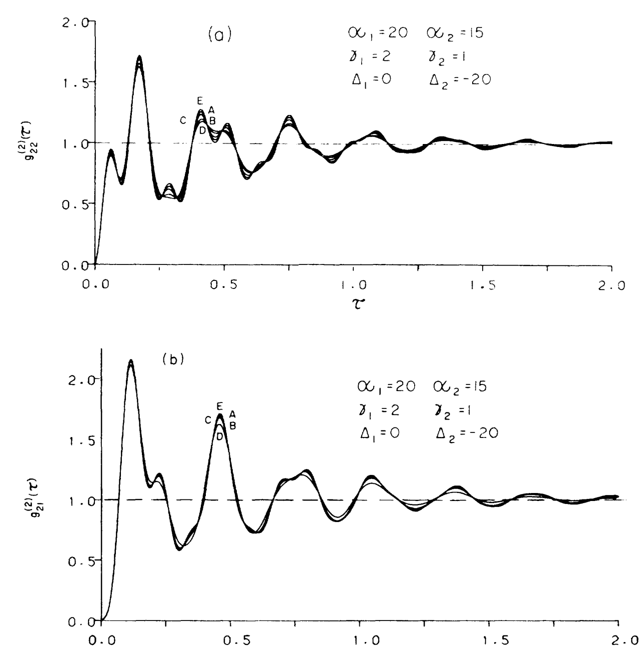

FIG. 8. Effect of amplitude fluctuations on the intensity-intensity correlation functions. (a) $g_{22}^{(2)}(\tau)$, (b) $g_{21}^{(2)}(\tau)$. Other data as in Fig. 6.

pared to the intensity parameters $\epsilon_1, \epsilon_2, \epsilon_{12}$. This is similar to what one observes in the case of the fluorescent spectra.

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APPENDIX A: SPECIAL CASE OF RESONANT EXCITATIONS $\Delta_1=\Delta_2=0$

When both the driving fields are in resonance with the corresponding atomic transitions, we have $\Delta_1=\Delta_2=0$. In this case the roots p_i and the coefficients a_i, b_i, c_i are explicitly determined by (2.21). This results in considerable simplifications. First, the coefficients f_{ij} take the form

$$f_{12}=f_{13}=\gamma_2\Gamma_1^2/2+f_0/2\ , \tag{A1}$$

$$f_{11}=2f_{21}=2f_{31}=\gamma_1\Gamma_2^2+f_0\ , \tag{A2}$$

$$f_{22}=f_{33}=\gamma_1\Gamma_1^2/4+\gamma_2(1-\Gamma_2^2/2)/2+\gamma_{c_1}\Gamma_1^4/4 +\gamma_{c_2}\Gamma_2^4/4-\Gamma_1^2\Gamma_2^2\gamma_{cc}/2+f_0/2\ , \tag{A3}$$

$$f_{23}=f_{32}=\gamma_1\Gamma_1^2/4+\gamma_2\Gamma_2^2/4+\gamma_{c_1}\Gamma_1^4/4 +\gamma_{c_2}\Gamma_2^4/4-\Gamma_1^2\Gamma_2^2\gamma_{cc}/2\ , \tag{A4}$$

$$f_0=\Gamma_1^2\Gamma_2^2(\gamma_{c_1}+\gamma_{c_2}+2\gamma_{cc})\ , \tag{A5}$$

$$g_1=2f_{12}\nu_2, g_2=g_3=2f_{21}\nu_2\ , \tag{A6}$$

$$\nu_1=2\gamma_1\Gamma_2^2+\gamma_2\Gamma_1^2+3\Gamma_1^2\Gamma_2^2(\gamma_{c_1}+\gamma_{c_2}+2\gamma_{cc})\ , \tag{A7}$$

$$\nu_2=\gamma_1\Gamma_1^2+\gamma_2+\gamma_{c_1}\Gamma_1^2(1+\Gamma_2^2)+\gamma_{c_2}\Gamma_2^2(1+\Gamma_1^2)\ , \tag{A8}$$

where

$$\Gamma_1=\alpha_1/\Omega,\quad \Gamma_2=\alpha_2/\Omega,\quad \Omega=(\alpha_1^2+\alpha_2^2)^{1/2}\ .$$

The solution for $\langle B_{ii}(t)\rangle_{pq}$ given by Eq. (3.7) in the text then simplifies to the following form:

$$\langle B_{11}(t)\rangle_{pq}=\frac{2f_{12}}{\nu_1}B(0)\exp[-a(p,q)t]+\frac{2}{\nu_1}[2f_{21}\langle B_{11}(0)\rangle_{pq}-f_{12}B^+(0)]\exp\{-[\nu_1+a(p,q)t]\}\ , \tag{A9}$$

$$\langle B_{22}(t)\rangle_{pq}=\frac{2f_{21}}{\nu_1}B(0)\exp[-a(p,q)t]+\frac{1}{\nu_1}[-2f_{21}\langle B_{11}(0)\rangle_{pq}+f_{12}B^+(0)]\exp\{-[\nu_1+a(p,q)t]\} +\tfrac{1}{2}B^-(0)\exp\{-[\nu_2+a(p,q)t]\}\ , \tag{A10}$$

$$\langle B_{33}(t)\rangle_{pq}=\frac{2f_{21}}{\nu_1}B(0)\exp[-a(p,q)t]+\frac{1}{\nu_1}[-2f_{21}\langle B_{11}(0)\rangle_{pq}+f_{12}B^+(0)]\exp\{-[\nu_1+a(p,q)t]\} -\tfrac{1}{2}B^-(0)\exp\{-[\nu_2+a(p,q)t]\}\ , \tag{A11}$$

where we have used the notation

$$B(0)=\langle B_{11}(0)\rangle_{pq}+\langle B_{22}(0)\rangle_{pq}+\langle B_{33}(0)\rangle_{pq}\ ,\quad B^{\pm}(0)=\langle B_{22}(0)\rangle_{pq}\pm\langle B_{33}(0)\rangle_{pq}\ . \tag{A12}$$

Further, when $\Delta_1=\Delta_2=0$ we have $b_1=0$ which implies that

$$\zeta_1(t)=\zeta_{12}(t)=\zeta_{13}(t)=\zeta_{31}(t)=\zeta_{21}(t)=0\ , \tag{A13}$$

$$\Gamma_{12}(p,q)=\Gamma_{13}(p,q)=\gamma_2(p,q)=\gamma(p,q)-b(p,q)\ , \tag{A14}$$

$$\Gamma_{21}(p,q)=\Gamma_{31}(p,q)=\gamma_3(p,q)=\gamma_2(-p,-q) =\gamma(p,q)+b(p,q)\ , \tag{A15}$$

$$\Gamma_{23}(p,q)=\Gamma_{32}(p,q)=\gamma_1(p,q)\ , \tag{A16}$$

where

$$\gamma(p,q)=a(p,q)+(\Gamma_2^4+\Gamma_1^2/2)\gamma_1+\gamma_2/2+(\Gamma_2^4+\Gamma_1^2/2)\gamma_{c_1} +(\Gamma_1^4+\Gamma_2^2/2)\gamma_{c_2}+2\gamma_{cc}(\Gamma_1^4+\Gamma_2^4)\ , \tag{A17}$$

$$b(p,q)=(p\gamma_{c_1}+q\gamma_{c_2})(2\Gamma_2^2-\Gamma_1^2) +(q\gamma_{c_2}+p\gamma_{cc})(\Gamma_2^2-2\Gamma_1^2)\ , \tag{A18}$$

$$\gamma_1(p,q)=3\gamma_1\Gamma_1^2/2+(1+\Gamma_2^2/2)\gamma_2+\Gamma_1^4\gamma_{c_1}/4+\Gamma_2^4\gamma_{c_2}/4 +\Gamma_1^2\Gamma_2^2(\gamma_{c_1}+\gamma_{c_2}+\gamma_{cc})+a(p,q)\ . \tag{A19}$$

The six equations for $\langle B_{ij}(t)\rangle_{pq}$ $(i\neq j)$ following from the general equation (3.4) in the text split into two pairs of coupled equations and two single equations,

$$d\langle B_{12}(t)\rangle_{pq}/dt=[i\Omega-\gamma_2(p,q)-\eta(t)]\langle B_{12}(t)\rangle_{pq} +f_0\langle B_{31}(t)\rangle_{pq}\ , \tag{A20}$$

$$d\langle B_{31}(t)\rangle_{pq}/dt=[i\Omega-\gamma_3(p,q)-\eta(t)]\langle B_{31}(t)\rangle_{pq} +f_0\langle B_{12}(t)\rangle_{pq}\ , \tag{A21}$$

$$d\langle B_{21}(t)\rangle_{pq}/dt=[-i\Omega-\gamma_3(p,q)-\eta(t)]\langle B_{21}(t)\rangle_{pq} +f_0\langle B_{13}(t)\rangle_{pq}\ , \tag{A22}$$

$$d\langle B_{13}(t)\rangle_{pq}/dt=[-i\Omega-\gamma_2(p,q)-\eta(t)]\langle B_{13}(t)\rangle_{pq} +f_0\langle B_{21}(t)\rangle_{pq}\ , \tag{A23}$$

$$d\langle B_{23}(t)\rangle_{pq}/dt=[-2i\Omega-\gamma_1(p,q)-4\eta(t)]\langle B_{23}(t)\rangle_{pq}\ , \tag{A24}$$

$$d\langle B_{32}(t)\rangle_{pq}/dt=[2i\Omega-\gamma_1(p,q)-4\eta(t)]\langle B_{32}(t)\rangle_{pq}\ , \tag{A25}$$

where the new function $\eta(t)$ is defined as

$$\eta(t)=\sum_{i=1}^{3}\beta_i\chi_i(t)\ , \tag{A26}$$

$$\beta_i=\Gamma_i^2 d_i^2\epsilon_i^2/\gamma_{a_i}\ (i=1,2)\ ,\quad \beta_3=2\Gamma_1\Gamma_2\epsilon_{12}^2/\gamma_{aa}\ , \tag{A27}$$

and $\chi_i(t)$ is defined in the text [Eq. (3.16)]. Taking the Laplace transform and solving the resulting pairs of simultaneous algebraic equations, we obtain

$$\langle\tilde{B}_{12}(s)\rangle_{pq}=\sum_{\{k\}}\frac{[(s+\beta/4+\lambda+\gamma_3(p,q)-i\Omega)\langle B_{12}(0)\rangle_{pq}+f_0\langle B_{31}(0)\rangle_{pq}]\Delta_1}{F_-(s+\beta/4+\lambda)}\ , \tag{A28}$$

$$\langle\tilde{B}_{31}(s)\rangle_{pq}=\sum_{\{k\}}\frac{\{[s+\beta/4+\lambda+\gamma_2(p,q)-i\Omega]\langle B_{21}(0)\rangle_{pq}+f_0\langle B_{21}(0)\rangle_{pq}\}\Delta_1}{F_-(s+\beta/4+\lambda)}\ , \tag{A29}$$

$$\langle\tilde{B}_{21}(s)\rangle_{pq}=\sum_{\{k\}}\frac{\{[s+\beta/4+\lambda+\gamma_2(p,q)+i\Omega]\langle B_{21}(0)\rangle_{pq}+f_0\langle B_{13}(0)\rangle_{pq}\}\Delta_1}{F_+(s+\beta/4+\lambda)}\ , \tag{A30}$$

$$\langle\tilde{B}_{13}(s)\rangle_{pq}=\sum_{\{k\}}\frac{\{[s+\beta/4+\lambda+\gamma_2(p,q)+i\Omega]\langle B_{13}(0)\rangle_{pq}+f_0\langle B_{21}(0)\rangle_{pq}\}\Delta_1}{F_+(s+\beta/4+\lambda)}\ , \tag{A31}$$

$$\langle\tilde{B}_{23}(s)\rangle_{pq}=\sum_{\{k\}}\frac{\Delta_2\langle B_{23}(0)\rangle_{pq}}{s+\beta+\lambda+\gamma_1(p,q)-2i\Omega}\ , \tag{A32}$$

$$\langle\tilde{B}_{32}(s)\rangle_{pq}=\sum_{\{k\}}\frac{\Delta_2\langle B_{32}(0)\rangle_{pq}}{s+\beta+\lambda+\gamma_1(p,q)+2i\Omega}\ , \tag{A33}$$

where $\langle\tilde{B}_{ij}(s)\rangle_{pq}$ is the Laplace transform of $\langle B_{ij}(t)\rangle_{pq}$, $\{k\}$ implies summation over integers k_1,k_2,k_3 ranging from zero to infinity, and the other notations used are as follows:

$$\begin{aligned}
&\beta=\sum_i\beta_i,\quad \delta=\sum_i\delta_i\ ,\\
&\delta_i=\beta_i/\gamma_{a_i}\quad (i=1,2),\quad \delta_3=\beta_3/\gamma_{aa}\ ,\\
&\Delta_1=\exp(\delta/4)\prod_{j=1}^{3}(-\delta_j/2)^{k_j}/k_j!\ ,\\
&\Delta_2=\exp(\delta)\prod_{j=1}^{3}(-\delta_j/2)^{k_j}/k_j!\ ,\\
&F_\pm=[s+\gamma_2(p,q)\pm i\Omega][s+\gamma_3(p,q)-i\Omega]-f_0^2\ .
\end{aligned} \tag{A34}$$

These Laplace transformed averages $\langle B_{ij}(s)\rangle$ are used to compute the fluorescent spectra under resonant conditions. The expressions for these spectra read as

$$\begin{aligned}
G_i(\omega)=&\frac{A_i}{(\gamma_i^0)^2+(\omega-\Omega_i)^2}+\sum_{\{k\}}\Delta_1\{k\}\left[\left[\frac{B_i^+}{(\gamma_i^+)^2+(\omega-\Omega_i-\Omega)^2}+\frac{B_i^-}{(\gamma_i^-)^2+(\omega-\Omega_i-\Omega)^2}\right]+(\Omega\to-\Omega)\right]\\
&+\sum_{\{k\}}\Delta_2\{k\}\left[\left[\frac{C_i}{(\gamma_i^1)^2+(\omega-\Omega_i-2\Omega)^2}\right]+(\Omega\to-\Omega)\right]\ ,
\end{aligned} \tag{A35}$$

where $i=1,2$ stand for the upper and lower spectrum, respectively, and

$$A_i=\gamma_i^0\Gamma_i^2\langle B_{22}\rangle_{00}/2\ , \tag{A36}$$

$$\gamma_i^0=\nu_2+\gamma_{c_i}\ , \tag{A37}$$

$$B_1^\pm=\frac{\Gamma_2^2\gamma_1^\pm f_1^\mp}{4q_1}\langle B_{11}\rangle_{00},\quad B_2^\pm=\frac{\Gamma_1^2\gamma_2^\pm f_2^\mp}{4q_2}\langle B_{22}\rangle_{00}\ , \tag{A38}$$

$$q_i=(f_0^2+r_i^2)^{1/2},\quad r_1=b(1,0),\quad r_2=b(0,1)\ , \tag{A39}$$

$$\gamma_i^\pm=\gamma_i\pm f_i^\pm+\beta/4+\lambda\ , \tag{A40}$$

$$f_i^{\pm}=q_i\pm r_i\ , \tag{A41}$$

$$\gamma_1=\gamma_1(1,0),\quad \gamma_2=\gamma_1(0,1)\ , \tag{A42}$$

$$C_i=\gamma_i^1\Gamma_i^2\langle B_{22}\rangle_{00}/4\ , \tag{A43}$$

$$\gamma_1^1=\gamma_1(1,0)+\beta+\lambda\ , \tag{A44}$$

$$\gamma_2^1=\gamma_1(0,1)+\beta+\lambda\ . \tag{A45}$$

In a similar manner, using the definitions in the text, the expressions for the intensity-intensity correlations can be derived as

$$g_{11}^{(2)}(\tau)=1+\frac{(\Gamma_1^2-2\Gamma_2^2)f_{12}}{2(\Gamma_2^2f_{12}+\Gamma_1^2f_{21})}\exp(-\nu_1\tau)-\frac{\nu_1\Gamma_1^2\exp(-[\beta+\gamma_1(0,0)]\tau)}{4(\Gamma_2^2f_{12}+\Gamma_1^2f_{21})}\sum_{\{k\}}\Delta_2(\{k\})\exp[-\lambda(\{k\})\tau]\cos(2\Omega\tau)\ , \tag{A46}$$

$$g_{22}^{(2)}(\tau)=1+\frac{(\Gamma_2^2f_{12}-2\Gamma_1^2f_{21})}{2f_{21}}\exp(-\nu_1\tau)-\frac{v_1\Gamma_2^2}{4f_{21}}\exp\{-[\beta+\gamma_1(0,0)]\tau\}\sum_{\{k\}}\Delta_2(\{k\})\exp[-\lambda(\{k\})\tau]\cos(2\Omega\tau)\ , \tag{A47}$$

$$g_{12}^{(2)}(\tau)=1+\frac{f_{12}}{2f_{21}}\exp(-\nu_1\tau)+\frac{\nu_1}{4f_{21}}\exp(-[\beta+\gamma_1(0,0)]\tau)\sum_{\{k\}}\Delta_2(\{k\})\exp[-\lambda(\{k\})\tau]\cos(2\Omega\tau)\ , \tag{A48}$$

$$\begin{aligned}g_{21}^{(2)}(\tau)=1&+\frac{[3\nu_1\Gamma_1^2\Gamma_2^2/2-2(\Gamma_2^2f_{12}+\Gamma_1^2f_{21})]}{2(\Gamma_2^2f_{12}+\Gamma_1^2f_{21})}\exp(-\nu_1\tau)\\&+\frac{\nu_1\Gamma_1^2\Gamma_2^2\exp(-[\beta+\gamma_1(0,0)]\tau)}{4(\Gamma_2^2f_{12}+\Gamma_1^2f_{21})}\sum_{\{k\}}\Delta_2(\{k\})\exp[-\lambda(\{k\})\tau]\cos(2\Omega\tau)\\&-\frac{\nu_1\Gamma_1^2\Gamma_2^2\exp(-[\beta/4+\gamma(0,0)]\tau)}{\Gamma_2^2f_{12}+\Gamma_1^2f_{21}}\sum_{\{k\}}\Delta_1(\{k\})\exp[-\lambda(\{k\})\tau]\cos(\Omega\tau)\ .\end{aligned} \tag{A49}$$

Note that these expressions can also be obtained from the corresponding expressions in the text by setting $b_1=0$ and subsequently using the simplifying relations (A1)–(A5) valid for $\Delta_1=\Delta_2=0$. This is because the extra terms present in the master equation do not affect the time behavior of the expectation values of the diagonal atomic operators.

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