Optical-double-resonance spectra and intensity-intensity correlations under intense fields with finite bandwidths: Some analytical results

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A formalism is presented to treat the effects of finite-bandwidth excitations 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. Under the conditions that one or both 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 ensembles. The quantum regression theorem is used to derive analytical expressions for the fluorescent spectra and the second-order intensity-correlation functions.

I. INTRODUCTION

The spectrum of the fluorescent radiation from a three-level atom undergoing stepwise transitions (optical double resonance) between the levels is known to exhibit many interesting characteristics depending upon the detuning from the atomic-transition frequencies and the strength of the two laser fields driving the atom.¹⁻⁶ It is known, for example, that when the atom is driven on resonance by strong fields, the spectrum of the radiation emitted by the upper as well as the lower transition is a Stark 'quintuplet.^{1,2} If, however, the driving fields are detune from the atomic-transition frequencies, the spectrum exhibits as many as seven Lorentzian peaks.^{1,3} If, on the other hand, 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. The upper spectrum, in this case, is the so called Autler-Townes doublet. $4-6$

The driving fields in the above studies are assumed to have zero bandwidth. A more realistic study should consider the effects of finite bandwidth of the driving laser fields. This bandwidth may arise, say, due to phase and/or amplitude fluctuations in the lasers. The effect of excitation bandwidths due to phase fluctuations on the Autler-Townes doublet has been studied theoretically by Agarwal and Narayana.⁷ They have shown that the asymmetry in the Autler-Townes doublet could switch due to the phase fluctuations. Some numerical results on the effects of the phase fluctuations on the fluorescent spectrum in the presence of two intense driving fields has been reported recently by D'Souza et al .⁸ 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 laser fields which are known to play a significant role in many situations. In the present paper, we consider the effects of both phase and amplitude fluctuations in the laser fields driving a three-level atom with stepwise excitation. We assume that the phase fluctuations follow a Wiener-Levy process⁹⁻¹² whereas the amplitude fluctuations are Levy process^{9–12} whereas the amplitude fluctuations are described by a nonwhite Gaussian process.^{13,14} Followin the treatment of Ref. 8, we derive an exact master equation for the atomic-density operator averaged over the distribution of phase fluctuations. Next, assuming one or both the fields driving the atom to be intense, we invoke the secular approximation to derive the master equation for the atomic-density operator averaged over nonwhite Gaussian amplitude fluctuations as well. We obtain an exact steady-state solution of the master equation derived in the high-field limit. We derive further analytic expressions for the spectrum of the fluorescent radiation from the upper as well as lower transitions which show explicitly the laser-bandwidth effects. If amplitude fluctuations are ignored, the analytic results are found to be in agreement with the numerical work of Ref. 8. We also obtain analytic expressions for the intensity-intensity correlation functions displaying the effects of driving-field fluctuations.

In Sec. II we present the basic formulation of the problem leading to the derivation of the master equation in the high-field limit and its steady-state solution. Section III is devoted to the derivation and discussion of the analytical expressions for the fluorescent spectra and the intensity-intensity correlation functions. Finally, a summary of the results is outlined in Sec. IV.

II. FORMULATION OF THE PROBLEM

A. Master equation

We consider a three-level atom interacting with two monochromatic applied laser fields which are in resonance with the atomic transition frequencies Ω_1 and Ω_2 . The levels denoted by $|1\rangle$, $|2\rangle$, and $|3\rangle$ are unequally spaced $(E_1>E_2>E_3)$ with $\Omega_1=(E_1-E_2)/\hbar$ and $\Omega_2 = (E_2 - E_3)/\hbar$. In the optical double resonance, we have the cascade configuration (Fig. 1), that is, the parity of $| 2 \rangle$ is opposite to that of levels $| 1 \rangle$ and $| 3 \rangle$ so that transitions from $|1\rangle$ to $|2\rangle$ and $|2\rangle$ to $|3\rangle$ are electric dipole allowed, while the transition from $|1\rangle$ to $|3\rangle$ is forbidden. The formulation developed in the following will, however, be applicable to treating the other configurations as well, with minor modifications. We start with the master equation¹⁵ for the reduced atomic-density operator ρ given by

$$
d\rho/dt = -i (d_1/2)[E_1^*(t)A_{12} + E_1(t)A_{21}, \rho]
$$

\n
$$
-i (d_2/2)[E_2^*(t)A_{23} + E_2(t)A_{32}, \rho]
$$

\n
$$
-\gamma_1(A_{11}\rho + \rho A_{11} - 2A_{21}\rho A_{12})
$$

\n
$$
-\gamma_2(A_{22}\rho + \rho A_{22} - 2A_{32}\rho A_{23}).
$$
\n(2.1)

The master equation (2.1) involves the usual rotating-wave and Markov approximations and is written in the frame rotating with respect to the laser frequencies. Further, $2\gamma_i$ are the Einstein A coefficients of the excited levels while d_1 and d_2 are transition matrix elements corresponding, respectively, to the transitions ¹ to 2 and 2 to 3. The operators $A_{mn} = |m\rangle\langle n|$ obey the usual commutation relations

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

We assume that the applied fields are described by

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

$$
E_j(t) = [E_j^{(0)} + E_j^{(1)}(t)] \exp[-i\phi_j(t)],
$$

\n
$$
\phi_j(0) = \phi_{j0} \quad (j = 1, 2)
$$
\n(2.3)

where the nonstochastic amplitude $E_j^{(0)}$ and the phase ϕ_{j0} are positive real numbers while the slowly varying time-dependent quantities $E_j(t)$ and ϕ_j are treated as stochastic variables. Now most of the theoretical treatments of noisy laser-atom interactions model the statistics of the fluctuations by means of suitable Gaussian distributions.^{$7-14$} This is inspired by the fact that Gaussian stochastic processes are simple to handle analytically. Also, in a realistic experimental situation, many different and independent sources of noise contribute and the resultant limiting distribution may indeed be Gaussian. 9 We assume here an extension of the standard model used to describe a single-mode noisy laser with small fluctuations.¹³ However, since two fields are acting on the same atom we may have to account for their mutual degree of incoherence.^{11} This cross-correlation also arises if the two fields are different modes of the same laser or if the second field is obtained from the first by splitting and frequency conversion. More specifically, we assume that the fluctuations in the phase and amplitude are statistically uncorrelated, described by independent Gaussian distributions. The statistics of phase fluctuations are described by the phase diffusion model in accordance with the wellknown Langevin equations for the phases: $9-12$

$$
d\phi_j/dt = \mu_j(t) , \qquad (2.4)
$$

where $\mu_i(t)$ is a Gaussian white noise with the properties (we use bold curly brackets to indicate an average)

(2.2)
$$
\{\mu_j(t)\} = 0, \qquad (2.5)
$$

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

Here $\gamma_{\rm cc}$ represents any cross-correlations that may be present between the lasers.

The amplitude fluctuations are assumed to be described by a Gaussian process with the mean and correlation given by^{13,1}

$$
\{E_j^{(1)}(t)\} = 0, \qquad (2.6)
$$

$$
\{E_j^{(1)}(t)E_k^{(1)}(t')\} = \begin{cases} \epsilon^2 \exp(-\gamma_{a_j} |t - t'|), & j = k \\ \epsilon_{12}^2 \exp(-\gamma_{aa} |t - t'|), & j \neq k \end{cases}
$$

where ϵ_{12} and γ_{aa} are the parameters accounting for the cross-correlation between the lasers.

In order to obtain the atomic observables averaged over phase and amplitude fluctuations, it is convenient to derive a master equation for the density operator averaged over the fluctuations. First we treat the phase fluctuations.

(2.9)

8. Phase-averaged density operator

We introduce the transformation

$$
W^{pq}(t) = \exp[-i(p\phi_1 + q\phi_2)]
$$

× $\exp{-i[(\phi_1 + \phi_2)A_{11} + \phi_2A_{22}]}$
× $\rho \exp{i[(\phi_1 + \phi_2)A_{11} + \phi_2A_{22}]}$ (2.7)

in (2.1) and obtain the equation obeyed by $W^{pq}(t)$:

$$
dW^{pq}(t)/dt = [L_0 - i\dot{\phi}_1(p + L_1) - i\dot{\phi}_2(q + L_2) + L_3]W^{pq}(t),
$$
 (2.8)

where

$$
L_0 W^{pq}(t) = -i\alpha_1 [A_{12} + A_{21}, W^{pq}] - i\alpha_2 [A_{23} + A_{32}, W^{pq}]
$$

$$
-\gamma_1 (A_{11} W^{pq} + W^{pq} A_{11} - 2A_{21} W^{pq} A_{12})
$$

$$
-\gamma_2 (A_{22} W^{pq} + W^{pq} A_{22} - 2A_{32} W^{pq} A_{23}),
$$

$$
L_1 W^{pq}(t) = [A_{11}, W^{pq}], \qquad (2.10)
$$

$$
L_2 W^{pq}(t) = [A_{11} + A_{22}, W^{pq}], \qquad (2.11)
$$

$$
L_3 W^{pq}(t) = -i [d_1 E_1^{(1)}(t)/2][A_{12} + A_{21}, W^{pq}]
$$

- $i [d_2 E_2^{(1)}(t)/2][A_{23} + A_{32}, W^{pq}],$ (2.12)

$$
\alpha_j = d_j E_j(0)/2 \quad (j = 1, 2) \tag{2.13}
$$

The next step is to obtain the master equation for $\{W^{pq}\},$ the transformed density operator averaged over the distributions of the phases $\phi_1(t)$ and $\phi_2(t)$. Since $\dot{\phi}_i = \mu_i(t)$ represent δ -correlated Gaussian processes, the theory of multiplicative stochastic processes¹⁶ yields an exact equation for the evolution of $\{W^{pq}\}\$:

$$
d\{W^{pq}(t)\} / dt
$$

= $[L_0 - \gamma_{c_1}(p + L_1)^2 - \gamma_{c_2}(q + L_2)^2$

$$
-2\gamma_{cc}(p + L_1)(q + L_2) + L_3]\{W^{pq}(t)\} .
$$
 (2.14)

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

$$
\{ \langle A_{kk} \rangle \} = \{ \operatorname{Tr} A_{kk} \rho \} = \operatorname{Tr} A_{kk} \{ W^{00} \} = \langle A_{kk} \rangle_{00} . \qquad (2.15)
$$

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

$$
\{ \langle A_{12} \rangle \} = \{ \operatorname{Tr}(A_{12}\rho) \} = \operatorname{Tr}(A_{12} \{ W^{10} \}) = \langle A_{12} \rangle_{10} ,
$$

$$
\{ \langle A_{23} \rangle \} = \{ \operatorname{Tr}(A_{23}\rho) \} = \operatorname{Tr}(A_{23} \{ W^{01} \}) = \langle A_{23} \rangle_{01} .
$$

(2.16)

The master equation (2.14) in the absence of amplitude fluctuations has been solved numerically in Ref. 8 to obtain the steady-state populations and the fluorescent spectra.

C. High-field limit and amplitude fluctuations

We note in (2.14) that the operator L_3 which contains the fluctuations in the field amplitudes does not commute with the remaining terms. Thus it is not immediately possible to derive an equation for ${W}^{pq}(t)$ _{amp}, the density operator averaged over both phase and amplitude fluctuations. However, considerable simplification results if we assume the fields to be strong. Indeed, in this high-field limit, the master equation for $\{W^{pq}(t)\}_{\text{amp}}$ may be easily derived from (2.14). For this purpose, it is convenient to introduce in (2.14) a set of new dressed operators B_{ij} relat ed to the operators A_{ij} as follows:

$$
B_{11} = \Gamma_1^2 A_{11} + \Gamma_2^2 A_{33} + \Gamma_1 \Gamma_2 (A_{13} + A_{31}),
$$

\n
$$
B_{22} = A_{22},
$$

\n
$$
B_{33} = \Gamma_2^2 A_{11} + \Gamma_1^2 A_{33} - \Gamma_1 \Gamma_2 (A_{13} + A_{31}),
$$

\n
$$
B_{12} = \Gamma_1 A_{12} + \Gamma_2 A_{32} = B_{21}^{\dagger},
$$

\n
$$
B_{13} = \Gamma_1 \Gamma_2 (A_{11} - A_{33}) - \Gamma_1^2 A_{13} + \Gamma_2^2 A_{31} = B_{31}^{\dagger},
$$

\n
$$
B_{23} = \Gamma_2 A_{21} - \Gamma_1 A_{23} = B_{32}^{\dagger},
$$

\n(2.17)

where

$$
\Gamma_j = \alpha_j / \Omega, \ \ \Omega = (\alpha_1^2 + \alpha_2^2)^{1/2} \ . \tag{2.18}
$$

Note that the transformation matrix is real and orthogonal and the new operators satisfy the same commutation relations as the old.

Next, we go over to the interaction representation by defining

$$
\{\widetilde{W}^{pq}\} = \exp[i\Omega(B_{12} + B_{21})t] \{W^{pq}\}\
$$

× $\exp[-i\Omega(B_{12} + B_{21})t]$ (2.19)

whereby the resulting master equation for $\{\widetilde{W}^{pq}(t)\}$ split into two parts: one containing no oscillatory terms and the other containing rapidly oscillating terms like $exp(\pm i \Omega t)$ and $exp(\pm 2i \Omega t)$. Making the secular approximation, that is, neglecting the oscillatory terms and reverting back to the Schrodinger picture, we arrive at the equation

$$
d\left\{W^{pq}\right\}/dt = \left\{\mathcal{L}_0 - i\left[\Gamma_1\eta_1(t) + \Gamma_2\eta_2(t)\right]\mathcal{L}_1\right\}\left\{W^{pq}\right\},\tag{2.20}
$$

where the operators \mathscr{L}_0 , \mathscr{L}_1 and the quantities η_j have the following meaning:

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$$
\mathcal{L}_{0}\{W^{pq}\} = -i\Omega[B_{12} + B_{21}, \{W^{pq}\}] - [(2c+g)/4][(B_{11} + B_{22})\{W^{pq}\} + \{W^{pq}\}(B_{11} + B_{22})]
$$

+ $[(c+g)/4][(B_{11} - B_{22})\{W^{pq}\}(B_{11} - B_{22})] + [(c-g)/4][B_{12}\{W^{pq}\}B_{12} + B_{21}\{W^{pq}\}B_{21}]$
+ $[(3c+g)/4][B_{12}\{W^{pq}\}B_{21} + B_{21}\{W^{pq}\}B_{12}] - a(p,q)\{W^{pq}\} - b(p,q)[B_{33}, \{W^{pq}\}]$
- $(e/4)(B_{33}\{W^{pq}\} + \{W^{pq}\}B_{33} - 2B_{33}\{W^{pq}\}B_{33})$
+ $f(B_{13}\{W^{pq}\}B_{13} + B_{31}\{W^{pq}\}B_{31} - B_{23}\{W^{pq}\}B_{23} - B_{32}\{W^{pq}\}B_{32})$
- $(a+f)(B_{33}\{W^{pq}\} + \{W^{pq}\}B_{33} - B_{13}\{W^{pq}\}B_{31} - B_{23}\{W^{pq}\}B_{32})$
- $(b+f)(B_{11}\{W^{pq}\} + \{W^{pq}\}B_{11} - B_{31}\{W^{pq}\}B_{13} - B_{32}\{W^{pq}\}B_{23})$, (2.21)

$$
\mathcal{L}_{1}\{W^{pq}\} = [B_{12} + B_{21}, \{W^{pq}\}],
$$
 (2.22)

$$
\eta_j(t) = [d_j E_j^{(1)}(t)]/2.
$$

The various constants occurring in (2.21) have the following definitions:

$$
a(p,q) = p^2 \gamma_{c_1} + q^2 \gamma_{c_2} + 2pq \gamma_{cc} , \qquad (2.24)
$$

$$
b(p,q) = (p\gamma_{c_1} + q\gamma_{cc})(2\Gamma_2^2 - \Gamma_1^2) - (q\gamma_{c_2} + p\gamma_{cc})(2\Gamma_1^2 - \Gamma_2^2) ,
$$
 (2.25)

$$
a = \gamma_1 \Gamma_2^2
$$
, $b = \gamma_2 \Gamma_1^2$, $c = \gamma_1 \Gamma_1^2 + \gamma_2 \Gamma_2^2$, (2.26)

$$
e = g + 4(\Gamma_2^2 - \Gamma_1^2)[\gamma_{c_1}\Gamma_2^2 - \gamma_{c_2}\Gamma_1^2 - \gamma_{cc}(\Gamma_2^2 - \Gamma_1^2)] ,
$$
\n(2.27)

$$
f = \Gamma_1^2 \Gamma_2^2 (\gamma_{c_1} + \gamma_{c_2} + 2\gamma_{cc}) , \qquad (2.28)
$$

$$
g = \gamma_{c_1} \Gamma_1^4 + \gamma_{c_2} \Gamma_2^4 - 2\gamma_{cc} \Gamma_1^2 \Gamma_2^2 . \qquad (2.29)
$$

Since the operator \mathscr{L}_1 multiplying the stochastic functions $\eta_1(t)$ and $\eta_2(t)$ commutes with \mathcal{L}_0 , it is now possible to apply the theory of multiplicative stochastic processes to obtain the following evolution equation for $\{W^{pq}(t)\}_{\text{amp}}$:

$$
d\left\{W^{pq}(t)\right\}_{amprm amp}/dt = \left[\mathcal{L}_0 - \eta^2(t)\mathcal{L}_1^2\right]\left\{W^{pq}(t)\right\}_{amprm amp}, \quad (2.30)
$$

where

$$
\eta^{2}(t) = d_{1}^{2} \epsilon_{1}^{2} \Gamma_{1}^{2} [1 - \exp(-\gamma_{a_{1}} t)]/4 \gamma_{a_{1}} + d_{2}^{2} \epsilon_{2}^{2} \Gamma_{2}^{2} [1 - \exp(-\gamma_{a_{2}} t)]/4 \gamma_{a_{2}} + d_{1} d_{2} \epsilon_{12}^{2} \Gamma_{1} \Gamma_{2} [1 - \exp(-\gamma_{aa} t)]/4 \gamma_{aa} .
$$
 (2.31)

The master equation (2.30) has two important properties. First, the equation describes a Markov process even though the evolution operator contains time-dependent coefficients. This property arises from the fact that the operators $\eta^2(t)\mathcal{L}_1^2$ and \mathcal{L}_0 commute for all t. The Markovian character of (2.30) in turn implies that the quantum regression theorem is applicable and it is possible to obtain two-time correlation functions from the one-time expectation values of the atomic operators. The second property of (2.30) is that it admits the steady-state solution

$$
\{W^{pq}\}_{amp;amp} = 0 \quad (p, q \neq 0) \tag{2.32a}
$$

$$
\{W^{00}\}_{\rm amp} = D^{-1} \exp(-\mu B_{33}), \qquad (2.32b)
$$

where

$$
\mu = \ln[(a+f)/(b+f)],
$$
\n(2.33)

$$
D = \text{Tr}[\exp(-\mu B_{33})]
$$

= 2 + \exp(-\mu) = (2a + b + 3f)/(a + f). (2.34)

The solution (2.32) is useful for obtaining the steady-state populations in the three levels. First note that

$$
\langle B_{11} \rangle_{00} = \text{Tr}(B_{11}W^{00}) = (a+f)/(2a+b+3f)
$$
, (2.35)

$$
\langle B_{22} \rangle_{00} = \langle B_{11} \rangle_{00} , \qquad (2.36)
$$

$$
\langle B_{33} \rangle_{00} = (b+f)/(2a+b+3f) , \qquad (2.37)
$$

so that

$$
\begin{aligned} \{ \langle A_{11} \rangle \}_{\text{amp}} &= \langle A_{11} \rangle_{00} = \Gamma_1^2 \langle B_{11} \rangle_{00} + \Gamma_2^2 \langle B_{33} \rangle_{00} \\ &+ \Gamma_1 \Gamma_2 (\langle B_{13} \rangle_{00} + \langle B_{31} \rangle_{00}) \\ &= (a \Gamma_1^2 + b \Gamma_2^2 + f) / (2a + b + 3f) \end{aligned} \tag{2.38}
$$

and similarly

$$
\{ \langle A_{22} \rangle \}_{\rm amp} = \langle B_{22} \rangle_{00} = (a + f)/(2a + b + 3f) , \qquad (2.39)
$$

$$
\{ \langle A_{33} \rangle \}_{\rm amp} = (a \Gamma_2^2 + b \Gamma_1^2 + f) / (2a + b + 3f) \ . \tag{2.40}
$$

It is clear that the steady-state populations are affected by the phase fluctuations alone. The fiuctuations in the field amplitudes have no effect on them at least in the highfield limit.

III. FLUORESCENT SPECTRA AND INTENSITY CORRELATIONS

A. One-time atomic operator averages

It is straightforward to obtain the equations for the evolution of one-time atomic expectation values averaged over the ensembles of the phase and amplitude fluctuations. For an atomic operator, we define the average as

$$
\langle O \rangle_{pq} = \text{Tr}(OW_{pq}) \tag{3.1}
$$

The equations for $\langle B_{ij} \rangle_{pq}$ are as follows:

$$
d \langle B_{11} \rangle_{pq} / dt = -i \Omega \psi_{pq}^- - \widetilde{\gamma} \langle B_{11} \rangle_{pq} + (a+f) \langle B_{33} \rangle_{pq}
$$

$$
- [(3c+g)/4 + 2\eta^2(t)] \chi_{pq}^- , \qquad (3.2)
$$

(2.23)

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$$
d \langle B_{22} \rangle_{pq} / dt = i \Omega \psi_{pq}^- - \widetilde{\gamma} \langle B_{22} \rangle_{pq} + (a + f) \langle B_{33} \rangle_{pq}
$$

$$
- [(3c + g) / 4 + 2\eta^2(t)] \chi_{pq}^- , \qquad (3.3)
$$

$$
d \langle B_{33} \rangle_{pq} / dt = -[2(a+f) + a(p,q)] \langle B_{33} \rangle_{pq} + (b+f) \chi_{pq}^{+} , \qquad (3.4)
$$

$$
d \langle B_{12} \rangle_{pq} / dt = -i \Omega \chi_{pq}^- - \widetilde{\gamma} \langle B_{12} \rangle_{pq}
$$

$$
+ [(c-g)/4] \langle B_{21} \rangle_{pq} - 2 \eta^2(t) \psi_{pq}^-, \quad (3.5)
$$

$$
d \langle B_{21} \rangle_{pq} / dt = i \Omega \chi_{pq}^- - \widetilde{\gamma} \langle B_{21} \rangle_{pq} + [(c - g) / 4] \langle B_{12} \rangle_{pq} + 2 \eta^2(t) \psi_{pq}^- , \quad (3.6)
$$

$$
d \langle B_{13} \rangle_{pq} / dt = i \Omega \langle B_{23} \rangle_{pq} - [\overline{\gamma} + b(p,q) - \eta^2(t)] \langle B_{13} \rangle_{pq}
$$

+ $f \langle B_{31} \rangle_{pq}$, (3.7)

$$
d \langle B_{31} \rangle_{pq} / dt = -i \Omega \langle B_{32} \rangle_{pq}
$$

$$
- [\overline{\gamma} - b (p,q) - \eta^2(t)] \langle B_{31} \rangle_{pq}
$$

$$
+ f \langle B_{13} \rangle_{pq} , \qquad (3.8)
$$

$$
d \langle B_{23} \rangle_{pq} / dt = i \Omega \langle B_{13} \rangle_{pq} - [\overline{\gamma} + b (p,q) - \eta^2(t)] \langle B_{23} \rangle_{pq}
$$

- $f \langle B_{32} \rangle_{pq}$, (3.9)

$$
d \langle B_{32} \rangle_{pq} / dt = -i \Omega \langle B_{31} \rangle_{pq} - [\overline{\gamma} - b (p, q) - \eta^2(t)] \langle B_{32} \rangle_{pq} - f \langle B_{23} \rangle_{pq} , \qquad (3.10)
$$

where we have used the notation

$$
\chi_{pq}^{\pm} = \langle B_{11} \rangle_{pq} \pm \langle B_{22} \rangle_{pq} , \qquad (3.11)
$$

$$
\psi_{pq}^{\pm} = \langle B_{12} \rangle_{pq} \pm \langle B_{21} \rangle_{pq} , \qquad (3.12)
$$

$$
\widetilde{\gamma}(p,q) = a(p,q) + b + f + [(5c + 3g)/4], \qquad (3.13)
$$

$$
\overline{\gamma}(p,q) = a(p,q) + a + [(b+c+3f)/2] + [(e+g)/4],
$$

 $\gamma_2(p,q) \!=\! \overline{\gamma}(p,q)+b\left(p,q\right)$, π) $\overline{x}(p, q) = b(p, q)$ (3.15)

$$
\gamma_3(p,q) = \gamma(p,q) - b(p,q) \; .
$$

Next, in addition to χ_{pq}^{\pm} and ψ_{pq}^{\pm} defined in (3.11) and (3.12) we need to introduce two pairs of new quantities

$$
\phi_{pq}^{\pm} = \langle B_{23} \rangle_{pq} \pm \langle B_{13} \rangle_{pq} , \qquad (3.16)
$$

$$
\zeta_{pq}^{\pm} = \langle B_{32} \rangle_{pq} \pm \langle B_{31} \rangle_{pq} . \tag{3.17}
$$

It is easy to see that the nine equations (3.2) - (3.10) split into a single equation for ψ_{pq}^{+} and four pairs of coupled equations:

$$
d\,\psi_{pq}^+ / dt = -\,\gamma_0(p,q)\psi_{pq}^+ \,, \tag{3.18}
$$

$$
dX_{pq}^+/dt = -\widetilde{\gamma}X_{pq}^+ + 2(a+f)\langle B_{33}\rangle_{pq} , \qquad (3.19a)
$$

$$
d \langle B_{33} \rangle_{pq} / dt = (b + f) \chi_{pq}^{+}
$$

- [2(a + f) + a (p,q)] $\langle B_{33} \rangle_{pq}$, (3.19b)

$$
d\chi_{pq}^-/dt = -[\gamma_1(p,q) + 4\eta^2(t)]\chi_{pq}^- - 2i\Omega\psi_{pq}^-, \qquad (3.20a)
$$

$$
d\psi_{pq}^-/dt = -2i\Omega\chi_{pq}^- - [\gamma_1(p,q) + 4\eta^2(t)]\psi_{pq}^-, \qquad (3.20b)
$$

$$
d\phi_{pq}^{+}/dt = [i\Omega - \gamma_2(p,q) - \eta(t)]\phi_{pq}^{+} - f\zeta_{pq}^{-} , \qquad (3.21a)
$$

$$
d\zeta_{pq}^-/dt = [i\Omega - \gamma_3(p,q) - \eta(t)]\zeta^- - f\phi_{pq}^+, \qquad (3.21b)
$$

$$
d\phi_{pq}^-/dt = -[i\Omega + \gamma_2(p,q) + \eta(t)]\phi_{pq}^- - f\zeta_{pq}^+, \qquad (3.22a)
$$

$$
d\phi_{pq}^{-}/dt = -[i\Omega + \gamma_{2}(p,q) + \eta(t)]\phi_{pq}^{-} - f\zeta_{pq}^{+} , \qquad (3.22a)
$$

$$
d\zeta_{pq}^{+}/dt = -[i\Omega + \gamma_{3}(p,q) + \eta(t)]\zeta_{pq}^{+} - f\phi_{pq}^{-}.
$$
 (3.22b)

For the computation of the fiuorescent spectra we need the Laplace transform of the above variables. Taking the Laplace transform and solving the resulting pairs of simultaneous algebraic equations we obtain

$$
\tilde{\psi}^+_{pq}(s) = \psi^+_{pq}(0)/(s+\gamma) \tag{3.23}
$$

(3.14)

 $\overline{}$

$$
\widetilde{\chi}^+_{pq}(s) = \frac{[s + a(p,q) + 2(a+f)]\chi^+_{pq}(0) + 2(a+f)(B_{33}(0))_{pq}}{[s + a(p,q)][s + a(p,q) + 2a + b + 3f]},
$$
\n(3.24)

$$
\langle \widetilde{B}_{33}(s) \rangle_{pq} = \frac{\left[s + a\left(p, q\right) + b + f\right] \langle B_{33}(0) \rangle_{pq} + (b + f) \chi_{pq}^{+}(0)}{\left[s + a\left(p, q\right)\right] \left[s + a\left(p, q\right) + 2a + b + 3f\right]} \tag{3.25}
$$

$$
\widetilde{\psi}_{pq}^{-}(s) = \sum_{\{\mathbf{k}\}} \frac{\{[s+\beta+\lambda+\gamma_1(p,q)]\psi_{pq}^{-}(0) - 2i\Omega\chi_{pq}^{-}(0)\}\Delta_1}{F_1(s+\beta+\lambda)},
$$
\n(3.26)

$$
\widetilde{\chi}_{pq}^{-}(s) = \sum_{\{\mathbf{k}\}} \frac{\{[s+\beta+\lambda+\gamma_{1}(p,q)]\chi_{pq}^{-}(0) - 2i\Omega\psi_{pq}^{-}(0)\}\Delta_{1}}{F_{1}(s+\beta+\lambda)},
$$
\n(3.27)

$$
\widetilde{\phi}_{pq}^{+}(s) = \sum_{\{k\}} \frac{\{[s + \beta/4 + \lambda + \gamma_{3}(p,q) - i\Omega]\phi_{pq}^{+}(0) - f\zeta_{pq}^{-}(0)\}\Delta_{2}}{F_{2}(s + \beta/4 + \lambda)},
$$
\n(3.28)\n
$$
\widetilde{\zeta}_{pq}^{-}(s) = \sum_{\{k\}} \frac{\{[s + \beta/4 + \lambda + \gamma_{2}(p,q) - i\Omega]\zeta_{pq}^{-}(0) - f\phi_{pq}^{+}(0)\}\Delta_{2}}{F_{2}(s + \beta/4 + \lambda)},
$$
\n(3.29)

$$
\widetilde{\zeta}_{pq}^{-}(s) = \sum_{\{\mathbf{k}\}} \frac{\{[s + \beta/4 + \lambda + \gamma_2(p,q) - i\Omega] \zeta_{pq}^{-}(0) - f\phi_{pq}^{+}(0)\} \Delta_2}{F_2(s + \beta/4 + \lambda)},
$$
\n(3.29)

$$
\widetilde{\phi}_{pq}^{-}(s) = \sum_{\{k\}} \frac{\{[s + \beta/4 + \lambda + \gamma_3(p,q) + i\Omega]\phi_{pq}^{-}(0) - f\zeta_{pq}^{+}(0)\}\Delta_2}{F_3(s + \beta/4 + \lambda)},
$$
\n(3.30)

(3.31)

$$
\widetilde{\zeta}_{pq}^{+}(s) = \sum_{\{\mathbf{k}\}} \frac{\{[s+\beta/4+\lambda+\gamma_{2}(p,q)+i\Omega\}_{pq}^{+}(0)-f\phi_{pq}^{-}(0)\}\Delta_{2}}{F_{3}(s+\beta/4+\lambda)}
$$

where $\tilde{f}(s)$ denotes the Laplace transform of $f(t)$, $\{k\}$ implies summation over k_1, k_2, k_3 ranging from 0 to ∞ , and the other notations used are as follows:

$$
\gamma_0(p,q) = \widetilde{\gamma}(p,q) + c + g \tag{3.32}
$$

$$
\gamma_1(p,q) = \widetilde{\gamma} + (3c+g)/2 \;,
$$

$$
\lambda \equiv \lambda(k_1, k_2, k_3) = k_1 \gamma_{a_1} + k_2 \gamma_{a_2} + k_3 \gamma_{aa} , \qquad (3.33)
$$

$$
\Delta_1(k_1, k_2, k_3) = \exp(\delta) \prod_{j=1}^3 (-\delta_j)^{k_j} / k_j!,
$$

$$
\Delta_2(k_1, k_2, k_3) = \Delta_1(k_1, k_2, k_3) / 2^{2(k_1 + k_2 + k_3)},
$$
 (3.34)

$$
\beta_j = d_j^2 \epsilon_j^2 \Gamma_j^2 / \gamma_{a_j} \quad (j = 1, 2) ,
$$
\n(3.35)

$$
\beta_3 = d_1 d_2 \epsilon_{12}^2 \Gamma_1 \Gamma_2 / \gamma_{aa} ,
$$

$$
\delta_j = \beta_j / \gamma_{a_j} \quad (j = 1, 2), \quad \delta_3 = \beta_3 / \gamma_{aa} \quad , \tag{3.36}
$$

$$
\beta = \beta_1 + \beta_2 + \beta_3, \quad \delta = \delta_1 + \delta_2 + \delta_3. \tag{3.37}
$$

The functions $F_1(s)$, $F_2(s)$, and $F_3(s)$ are defined as

 \rightarrow ²

$$
F_1(s) = [s + \gamma_1(p,q)]^2 + 4\Omega^2, \qquad (3.38)
$$

$$
F_2(s) = [s + \gamma_2(p,q) - i\Omega][s + \gamma_3(p,q) - i\Omega] - f^2,
$$

(3.39)

$$
F_3(s) = [s + \gamma_2(p,q) + i\Omega][s + \gamma_3(p,q) + i\Omega] - f^2.
$$

$$
F_3(s)=[s+\gamma_2(p,q)+i\Omega][s+\gamma_3(p,q)+i\Omega]-f^2.
$$

(3.40)

I

The definitions (3.11) and (3.12), and (3.16} and (3.17), then allow us to obtain the Laplace transform averages $\langle B_{ij}(s) \rangle$ of the atomic operators.

B. Fluorescent spectra in the steady state

The steady-state fiuorescent spectrum for emission (3.33) from the upper excited level $|1\rangle$ is defined as

$$
\Delta_1(k_1, k_2, k_3) = \exp(\delta) \prod_{j=1}^3 (-\delta_j)^{k_j} / k_j!,
$$
\n
$$
\Delta_1(k_1, k_2, k_3) = \exp(\delta) \prod_{j=1}^3 (-\delta_j)^{k_j} / k_j!,
$$
\n(3.34)\n
$$
\Delta_2(k_1 + k_2 + k_3)
$$
\n
$$
\Delta_3(k_1, k_2, k_3) = \text{Re}\left[\left\{\langle A_{12}(s)A_{21}(0)\rangle_{ss}\right\}_{\text{amp}}\big|_{s=i(\omega-\Omega_1)}\right].
$$
\n(3.41)

Now from Eqs. (2.16), (3.12), and (3.17) it follows that the phase- and amplitude-averaged expectation value of $A_{12}(s)$ is given by

$$
\delta_j = \beta_j / \gamma_{a_j} \quad (j = 1, 2), \quad \delta_3 = \beta_3 / \gamma_{aa} \quad (3.36)
$$
\n
$$
\beta = \beta_1 + \beta_2 + \beta_3, \quad \delta = \delta_1 + \delta_2 + \delta_3 \quad (3.37)
$$
\n
$$
\beta = \beta_1 + \beta_2 + \beta_3, \quad \delta = \delta_1 + \delta_2 + \delta_3 \quad (3.37)
$$
\n
$$
\beta = \beta_1 + \beta_2 + \beta_3, \quad \delta = \delta_1 + \delta_2 + \delta_3 \quad (3.38)
$$
\n
$$
\beta = \gamma_1 (s) + \gamma_2 (s), \quad \text{and} \quad F_3(s) \quad \text{are defined as}
$$
\n
$$
F_1(s) = [s + \gamma_1 (p, q)]^2 + 4\Omega^2, \quad (3.38)
$$
\n
$$
\beta = \gamma_1 (s) + \gamma_2 (s), \quad \text{and} \quad F_2(s) \quad (3.39)
$$
\n
$$
\beta = \gamma_1 (s) + \gamma_2 (s), \quad \text{and} \quad F_3(s) \quad (3.30)
$$
\n
$$
\beta = \gamma_1 (s) + \gamma_2 (s), \quad \text{and} \quad F_4(s) = \gamma_1 (s) + \gamma_2 (s), \quad (3.41)
$$

The right-hand side (rhs) of (3.42) is next expressed in terms of $\langle B_{ij}(0) \rangle_{10}$ using Eqs. (3.23)–(3.31) and the quantum regression theorem is applied to obtain $\{\langle \tilde{A}_{12}(s)A_{21}(0)\rangle\}_{amp{amp}}$. The analytical expression for the upper spectrum then reads as

$$
G_{1}(\omega) = \frac{\Gamma_{1}^{2}}{2} \langle B_{11} \rangle_{00} \frac{\gamma_{0}(1,0)}{\gamma_{0}^{2}(1,0) + (\omega - \Omega_{1})^{2}} + \frac{\Gamma_{2}^{2}}{4\bar{f}(1,0)} \langle B_{33} \rangle_{00} \sum_{\{k\}} \Delta_{2}(k) \left[\frac{f^{-(1,0)\gamma + (1,0)}}{\gamma + (1,0)^{2} + (\omega - \Omega_{1} - \Omega)^{2}} + \frac{f^{+(1,0)\gamma - (1,0)}}{\gamma - (1,0)^{2} + (\omega - \Omega_{2} - \Omega)^{2}} + (\Omega \to -\Omega) \right] + \frac{\Gamma_{1}^{2}}{4} \langle B_{11} \rangle_{00} \sum_{\{k\}} \Delta_{1}(k) \left[\frac{\gamma_{1}(1,0) + \beta + \lambda}{[\gamma_{1}(1,0) + \beta + \lambda]^{2} + (\omega - \Omega_{1} - 2\Omega)^{2}} + (\Omega \to -\Omega) \right],
$$
(3.43)

where $\langle B_{11} \rangle_{00}$ and $\langle B_{33} \rangle_{00}$ are defined in Eqs. (2.23) and $G_2(\omega) = \text{Re} \int_0^\infty \exp[-i(\omega - \Omega_2)\tau]$
(2.37), respectively, and the other notation is as follows:

$$
\bar{f}(p,q) = [f^2 + b^2(p,q)]^{1/2}, \qquad (3.44)
$$

$$
f^{\pm}(p,q) = \bar{f}(p,q) \pm b(p,q) , \qquad (3.45)
$$

$$
\gamma^{\pm}(p,q) = \gamma(p,q) \pm f(p,q) + \beta/4 + \lambda . \qquad (3.46)
$$

The steady-state averages $(B_{11})_{00}$ and $(B_{33})_{00}$ are given by (2.35) and (2.37) , respectively.

The steady-state spectrum for emission from the 1ower excited level $|2\rangle$ is similarly defined as

re
$$
\langle B_{11} \rangle_{00}
$$
 and $\langle B_{33} \rangle_{00}$ are defined in Eqs. (2.23) and
\n7), respectively, and the other notation is as follows:
\n
$$
\overline{f}(p,q) = [f^2 + b^2(p,q)]^{1/2},
$$
\n(3.44)
\n
$$
\overline{f}(p,q) = [f^2 + b^2(p,q)]^{1/2},
$$
\n(3.46)
\n
$$
\overline{f}(p,q) = [f^2 + b^2(p,q)]^{1/2},
$$
\n(3.47)

It is clear from Eqs. (2.16) , (3.12) , and (3.16) that

$$
\{\langle \widetilde{A}_{23}(s) \rangle\}_{amprm amp} = \langle \widetilde{A}_{23}(s) \rangle_{01}
$$

\n
$$
= \Gamma_2 \langle B_{21}(s) \rangle_{01} - \Gamma_1 \langle B_{23}(s) \rangle_{01}
$$

\n
$$
= (\Gamma_2/2) [\widetilde{\psi}^{\dagger}_{01}(s) + \widetilde{\psi}^{\dagger}_{01}(s)]
$$

\n
$$
- (\Gamma_1/2) [\widetilde{\phi}^{\dagger}_{01}(s) + \widetilde{\phi}^{\dagger}_{01}(s)] . \qquad (3.48)
$$

Inserting in (3.44) the solutions for $\tilde{\psi}_{01}^{\pm}(s)$ and $\tilde{\phi}_{01}^{\pm}(s)$ and applying the regression theorem we obtain $\{\langle \tilde{A}_{23}(s)A_{32}\rangle_{ss}\}_{amb}$ and hence the fluorescent spectrum for emission from the level $|2\rangle$ is

$$
G_{2}(\omega) = \frac{\Gamma_{2}^{2}}{2} \langle B_{22} \rangle_{00} \frac{\gamma_{0}(0,1)}{\gamma_{0}^{2}(0,1) + (\omega - \Omega_{2})^{2}} + \frac{\Gamma_{1}^{2}}{4f(0,1)} \langle B_{22} \rangle_{00} \sum_{\{k\}} \Delta_{2}(k) \left(\frac{f^{+}(0,1)\gamma^{+}(0,1)}{\gamma^{+}(1,0)^{2} + (\omega - \Omega_{2} - \Omega)^{2}} + \frac{f^{-}(1,0)\gamma^{-(1,0)}}{\gamma^{-(1,0)^{2} + (\omega - \Omega_{2} - \Omega)^{2}}} + (\Omega \to -\Omega) \right) + \frac{\Gamma_{2}^{2}}{4} \langle B_{22} \rangle_{00} \sum_{\{k\}} \Delta_{1}(k) \left(\frac{\gamma_{1}(0,1) + \beta + \lambda}{[\gamma_{1}(0,1) + \beta + \lambda]^{2} + (\omega - \Omega_{2} - 2\Omega)^{2}} + (\Omega \to -\Omega) \right), \tag{3.49}
$$

where $\langle B_{22} \rangle_{00}$ is defined in Eqs. (2.36) while the other quantities have the meaning given in Eqs. (3.44)–(3.46).

The expression (3.43) and (3.49) for the fluorescent spectra are somewhat complicated and it may be worthwhile to discuss at first some special cases. We first consider the case when fluctuations are absent so that the parameters $a(p,q)$, $b(p,q)$, e, f, and g, related to the phase fluctuations and the quantities β , λ , and γ related to the amplitude fluctuations are all zero. Consequently, the terms corresponding to $k_1 = k_2 = k_3 = 0$ alone survive in the sums on the rhs of Eqs. (3.43) and (3.49). Thus, the spectra, in absence of fluctuations, have the simple form

$$
G_i(\omega) = \frac{1}{2(2a+b)} \left[A_i \left(\frac{\gamma_0}{\gamma_0^2 + (\omega - \Omega_i)^2} + \frac{1}{2} \frac{\tilde{\gamma}}{\gamma^2 + (\omega - \Omega_i - 2\Omega)^2} + (\Omega \to -\Omega) \right) + B_i \left(\frac{\overline{\gamma}}{\gamma^2 + (\omega - \Omega_i - \Omega)^2} + (\Omega \to -\Omega) \right) \right]
$$
\n(3.50)

where $i = 1, 2$ and

$$
A_i = a\Gamma_i^2, \quad B_1 = b\Gamma_2^2, \quad B_2 = A_1 \tag{3.51}
$$

$$
\gamma_0 = b + c
$$
, $\tilde{\gamma} = b + 3c/2$, $\tilde{\gamma} = a + (b + c)/2$. (3.52)

It is clear from these expressions that, in general, the spectra are symmetric and have five peaks. The peaks correspond to the central Lorentzian of width γ_0 at the excitation frequency and two pairs of side bands located at $\omega = \Omega_i \pm \Omega$ and $\omega = \Omega_i \pm 2\Omega$ ($i = 1, 2$) with widths $\bar{\gamma}$ and $\tilde{\gamma}$, respectively. Although the positions (as measured from the respective excitation frequency) and the widths of the peaks are the same for both upper and lower spectra, the

heights of the peaks differ for the two spectra. For example, consider the extreme cases $\alpha_1 \gg \alpha_2$ and $\alpha_1 \ll \alpha_2$. In the former case, the lower spectrum $G_2(\omega)$ shows two prominent peaks at $\omega = \Omega_2 \pm \alpha_1$ and the three smaller ones at $\omega = \Omega_2$ and $\omega = \Omega_2 \pm 2\alpha_1$ are considerably suppressed; the upper spectrum $G_1(\omega)$ shows all five peaks at $\omega = \Omega_1$, $\omega = \Omega_1 \pm \alpha_1$, and $\omega = \Omega_1 \pm 2\alpha_1$ [Fig. 2(a)]. In the latter case, the upper spectrum shows five weak peaks as before but the lower spectrum shows prominent peaks at $\omega = \Omega_2$ and $\omega = \Omega_2 \pm 2\alpha_2$ while the peaks at $\omega = \Omega_2 \pm \alpha_2$ are suppressed [Fig. 2(b)]. These analytical results are in agreement with the dressed-atom treatment of Cohen-Tannoudji and Reynaud² and also with the numerical results of Whitley and

FIG. 2. Optical-double-resonance emission spectra in absence of fluctuations: (a) $\alpha_1 \gg \alpha_2$, (b) $\alpha_2 \gg \alpha_1$.

Stroud.¹ Thus, in absence of fluctuations, the present dressed operator formalism is found to be equivalent to other approaches and is expected to yield correct results even when fluctuations are considered.

Next, me ignore the amplitude fluctuations but retain

the phase fluctuations. Since the parameters β, λ, γ pertaining to amplitude fluctuations are all zero, the sums on the rhs of Eqs. (3.43) and (3.49) reduce to a single term corresponding to $k_1 = k_2 = k_3 = 0$. The analytical expressions for the spectra take the form

$$
G_i(Q) = F_i \left[\frac{\gamma_i^0}{(\gamma_i^0)^2 + (\omega - \Omega_i)^2} + \left[\frac{\gamma_i/2}{\gamma_i^2 + (\omega - \Omega_i - 2\Omega)^2} + (\Omega - \Omega) \right] \right]
$$

+
$$
\left[\left(\frac{G_i^+}{(\gamma_i^+)^2 + (\omega - \Omega_i - \Omega)^2} + \frac{G_i^-}{(\gamma_i^-)^2 + (\omega - \Omega_i - \Omega)^2} \right] + (\Omega - \Omega - \Omega) \right]
$$
(3.53)

with the following notation:

$$
F_i = (\Gamma_i^2/2) \langle B_{11} \rangle_{00} \quad (i = 1, 2) \tag{3.54}
$$

$$
G_1^{\pm} = (\Gamma_2^2 / 4 \bar{f}_1) f_1^{\mp} \gamma_1^{\pm} \langle B_{33} \rangle_{00} , \qquad (3.55)
$$

$$
G_2^{\pm} = (\Gamma_2^2 / 4 \bar{f}_2) f_2^{\pm} \gamma_2^{\pm} \langle B_{11} \rangle_{00} , \qquad (3.56)
$$

$$
\langle B_{11} \rangle_{00} = \langle B_{22} \rangle_{00} = (a+f)/(2a+b+3f) , \qquad (3.57)
$$

$$
\langle B_{33}\rangle_{00} = (b+f)/(2a+b+3f) ,
$$

$$
\overline{f}_i = (f^2 + b_i^2)^{1/2} f_i^{\pm} = \overline{f}_i \pm b_i
$$
\n
$$
b_1 = b(1,0), \quad b_2 = b(0,1) \tag{3.58}
$$

$$
\gamma_i^{\pm} = a + (b + c + 3f)/2 + (e + g)/4
$$

$$
+\gamma_{c_i} \pm (f^2 + b_i^2)^{1/2} \;, \tag{3.59}
$$

$$
\gamma_i^0 = b + c + f + g + \gamma_{c_i} \,, \tag{3.60}
$$

$$
\gamma_i = (3c + g)/2 + b + f + \gamma_{c_i} \tag{3.61}
$$

FIG. 3. Effect of phase fluctuations on the fluorescent spectra for the upper transition: curves $A - C$ correspond to $\gamma_{c_2} = 0$ and $\gamma_{c_1} = 0, 1, 2$, respectively. Curve D represents $\gamma_{c_1} = 0$, $\gamma_{c_2} = 2$; while E is for $\gamma_{c_1} = \gamma_{c_2} = 3$. In all these curves $\gamma_{cc} = 0$.

A notable feature of the analytical spectrum (3.53) in presence of phase fluctuations is that each of the near sidebands at $\omega = \Omega_i \pm \Omega$ arises from a superposition of two Lorentzians of unequal widths γ_i . In the absence of cross-correlations ($\gamma_{\rm cc}$ = 0) the major effect of phase fluctuations is to suppress the central peak as well as the remote sidebands as seen in Fig. 3 where the upper spectra are shown for several values of $\gamma_{c_1}, \gamma_{c_2}$. As the bandwidths $\gamma_{c_1}, \gamma_{c_2}$ are increased one expects that the Stark quintuplets reduce to Stark doublets in agreement with the completely numerical results of Ref. 8. On the other hand, for a fixed set of the bandwidth parameters γ_{c} and γ_{c_2} , an increase in the cross-correlation bandwidth γ_{cc} results in the suppression of the near sidebands with an enhancement of the central peak and the remote sidebands. This is clear from the curves $A - E$ in Fig. 4 where the upper spectra are shown for several values of γ_{cc} and for a given set of parameters γ_{c_i} .

Finally, we return to the expressions (3.43) and (3.49) for the fluorescent spectra from the upper and the lower excitation levels which contain the effects due to both

phase and amplitude fluctuations. The analytical spectra are symmetric with a central peak at $\omega = \Omega_i$ and two pairs of sidebands at $\omega = \Omega_i \pm \Omega$ and $\omega = \Omega_i \pm 2\Omega$. The contributions to the effective height and width of each of these sidebands come from a superposition of an infinite number of Lorentzians located at the sideband center with heights and widths depending on the parameter $\lambda(k_1, k_2, k_3)$ (with $k_1, k_2, k_3 = 0, 1, 2, ...$). It is also clear from the analytical expressions that phase fluctuations affect both the central peaks as well as the sidebands while amplitude fluctuations affect only the sidebands and have no effect whatsoever on the central peaks. In particular, for a given set of amplitude fluctuation parameters, an increase in any of the phase bandwidth parameters has the effect of decreasing the heights and increasing the widths of both the central and side peaks.

For a fixed set of phase bandwidth parameters $\gamma_{c_i}, \gamma_{cc}$ and a fixed set of amplitude fluctuation parameters β_i , the central peak is not affected by an increase in any of the amplitude bandwidth parameters γ_{a_i} or γ_{aa} whereas the side peaks show a slight increase in height and a reduction in width. This can be seen by comparing curves A and E

FIG. 4. Effect of phase cross-correlations on the fluorescent spectra for the upper transition: curves $A-D$ correspond to $\gamma_{c_1} = \gamma_{c_2} = 2$, and $\gamma_{\infty} = 0$, 0.5, 1, and 2, respectively.

FIG. 5. Effect of amplitude fluctuations on the fluorescent spectra for the upper transition: curves $A-D$ correspond to $\gamma_{a_1} = \gamma_{a_2} = \gamma_{aa} = 0.5$, and $(\epsilon_1^2, \epsilon_2^2, \epsilon_1^2)$ 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 parameters are $\gamma_{c_1} = \gamma_{c_2} = \gamma_{cc} = 0.5$.

in Fig. 5. Further, it is seen from the analytical expression and curves $A-D$ of Fig. 5 that given a set of phase parameters γ_{c_i} and γ_{cc} and amplitude bandwidth parameters γ_{a_i} and γ_{aa} an increase in any of the amplitude parameters β_i does not influence the central peak but leads to a reduction of height and a broadening of the side peaks.

C. Intensity-intensity correlation function

The nature of the fluorescent light emitted from the excited levels $|1\rangle$ and $|2\rangle$ may be understood from the second-order intensity correlation functions. We define these correlations in the steady state as follows:

$$
g_{11}^{(2)}(\tau) = \{ \langle A_{12}A_{12}(\tau)A_{21}(\tau)A_{21} \rangle \}_{amp;amp}/ |\{ \langle A_{12}A_{21} \rangle \}_{amp;amp}|^2
$$

$$
= \langle A_{12}A_{11}(\tau)A_{21} \rangle_{amp}/ |\langle A_{11} \rangle_{amp}/|^2 , \qquad (3.62)
$$

$$
g_{22}^{(2)}(\tau) = \{ \langle A_{23}A_{23}(\tau)A_{32}(\tau)A_{32} \rangle \}_{\text{amp}} / |\{ \langle A_{23}A_{32} \rangle \}_{\text{amp}}|^2
$$

= $\langle A_{23}A_{22}(\tau)A_{32} \rangle_{00} / |\langle A_{22} \rangle_{00}|^2$, (3.63)

and

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$$
g_{12}^{(2)}(\tau) = \{ \langle A_{12}A_{23}(\tau)A_{32}(\tau)A_{21} \rangle \}_{amp;amp}/ |\{ \langle A_{23}A_{32} \rangle \}_{amp;amp} \{ \langle A_{12}A_{21} \rangle \}_{amp;amp} |
$$

= $\langle A_{12}A_{22}(\tau)A_{21} \rangle_{00} / |\langle A_{22} \rangle_{00} \langle A_{11} \rangle_{00} |$. (3.64)

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, $g_{12}^{(2)}(\tau)$ is a mea photon emitted from the upper excitation level at time $t = 0$ and another photon emitted from the lower excitation level at a later time $t = \tau$.

In order to evaluate the correlation functions one first obtains the expectation values $\langle A_{11}(\tau)\rangle_{00}$ and $\langle A_{22}(\tau)\rangle_{00}$ and then applies the quantum regression theorem. The expectation values $\langle A_{11}(\tau) \rangle_{00}$ and $\langle A_{22}(\tau) \rangle_{00}$ may in turn be obtained by first expressing the A_{11} and A_{22} operators in terms of the operators $\langle B_{ij}$ Laplace transform of Eqs. (3.23) – (3.31) . Omitting these tedious but straightforward algebraic steps, we quote here the analytical results for the second-order correlation functions as

 \sim \sim \sim \sim \sim

$$
g_{11}^{(2)}(\tau) = 1 + \frac{(b+f)(\Gamma_1^2 - 2\Gamma_2^2)}{2[\Gamma_1^2(a+f) + \Gamma_2^2(b+f)]} \exp[-(2a+b+3f)\tau]
$$

$$
- \frac{\Gamma_1^2(2a+b+3f)}{2[\Gamma_1^2(a+f) + \Gamma_2^2(b+f)]} \exp\{-[\beta + \gamma_1(0,0)]\tau\} \sum_{\{k\}} \Delta_1 \exp(-\lambda \tau) \cos(2\Omega \tau) ,
$$
(3.65)

FIG. 6. Effect of phase fluctuations on the intensity-intensity correlation function for the upper transition: curves $A-D$ correspond to $\gamma_{cc} = 0$ and $(\gamma_{c_1}, \gamma_{c_2})$ are (0,0), (0.5,0.5), (1,1), and (2,2), respectively.

$$
g_{22}^{(2)} = 1 + \left[\frac{\Gamma_2^2(b+f)}{2(a+f)} - \Gamma_1^2 \right] \exp[-(2a+b+3f)\tau]
$$

$$
- \frac{\Gamma_2^2(2a+b+3f)}{2(a+f)} \exp\{-[\beta+\gamma_1(0,0)]\tau\} \sum_{\{k\}} \Delta_1 \exp(-\lambda \tau) \cos(2\Omega \tau) ,
$$

$$
g_{12}^{(2)}(\tau) = 1 + \frac{b+f}{2(a+f)} \exp[-(2a+b+3f)\tau]
$$

$$
+ \frac{2a+b+3f}{2(a+f)} \exp\{-[\beta+\gamma_1(0,0)]\tau\} \sum_{\{k\}} \Delta_1 \exp(-\lambda \tau) \cos(2\Omega \tau) .
$$
 (3.67)

It is interesting to note here that first two correlation functions vanish at $\tau=0$ and increase towards unity for long times τ . The second-order correlation function $g_{12}^{(2)}(\tau)$, however, does not vanish at $\tau=0$, which implies that there is a finite probability for simultaneous emission of two photons of frequency Ω_1 and frequency Ω_2 . It is clear from these expressions that in the absence of fluctuations both $g_{11}^{(2)}(\tau)$ and $g_{22}^{(2)}(\tau)$ show the expected oscillations through bunching and antibunching cycles decaying to their steady-state value of unity. The phase and/or amplitude fluctuations tend to reduce the amplitudes of

these fluctuations without changing the qualitative nature of these curves. This is indicated in Figs. 6 and 7 where $g_{11}^{(2)}(\tau)$ is plotted versus τ for some representative set of fluctuation parameters. We might mention here that the effects of cross-correlations $\gamma_{\rm cc}$ on the behavior of $g_{11}^{(2)}(\tau)$ and $g_{22}^{(2)}(\tau)$ cannot be distinguished from those of the self-correlations γ_{c_1} and γ_{c_2} . In order to look for the cross-correlation effects one has to rely mainly on the behavior of the fluorescent spectra. Also as is seen from curves $A - E$ of Fig. 7 the intensity-intensity correlations are relatively less sensitive to the variation in the band-

FIQ. 7. Effect of amplitude fluctuations on the intensity-intensity correlation function for the upper transition: other data as in Fig. 5.

width parameters γ_{a_1} , γ_{a_2} , and γ_{aa} as compared to the intensity parameters $\epsilon_1, \epsilon_2, \epsilon_{12}$ of the amplitude fluctuations.

IV. SUMMARY AND CONCLUSION

In this paper we have made a systematic study of the effects due to finite-bandwidth excitations on the optical double resonance. For this purpose, we assumed that finite bandwidth arises from phase and/or amplitude fluctuations. The phases follow the usual diffusion model while the amplitude fluctuations are described by colored Gaussian noise. Under these assumptions and when one or both of the driving fields are intense, we have derived an appropriate Markovian master equation for the atomic-density operator averaged over the both phase and amplitude ensembles. This master equation is further used to derive analytical expressions for the fluorescent spectra as well as second-order intensity correlation functions. These analytical results exhibit explicitly the effects due to phase and amplitude fluctuations. We found that the emission spectra are considerably affected by fluctuations in the exciting fields. In particular, the main effect of the phase diffusion on the spectra (which represent Stark quintuplets) is a decrease in the intensity and broadening of both the central and side peaks. On the other hand, the amplitude fluctuations affect only the sidebands. Cross-correlations between the lasers have significant effect on the spectra. In particular, crosscorrelations in phases tend to suppress the near sidebands but increase their intensity and reduce the width of the central peaks as well as the remote sidebands. The effect of fluctuations on the second-order intensity correlation functions is less significant. Fluctuations tend to reduce the amplitudes of the bunching and antibunching oscillations but do not change the basic character of these functions.

We have not considered the detuning effects in the present paper. However, with some modifications, our formalism is applicable to this case also. It is then possible to study analytically the excitation bandwidth effects not only on the fluorescent spectra but also on the Autler-Townes doublet. We hope to report the details of these interesting aspects in a subsequent paper.

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