Phase-dependent fluorescence linewidth narrowing in a three-level atom damped by a finite-bandwidth squeezed vacuum

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We examine subnatural phase-dependent linewidths in the fluorescence spectrum of a three-level atom damped by a narrow-bandwidth squeezed vacuum in a cavity. Using the dressed-atom model approach of a strongly driven three-level cascade system, we derive the master equation of the system from which we obtain simple analytical expressions for the fluorescence spectrum. We show that the phase effects depend on the bandwidths of the squeezed vacuum and the cavity relative to the Rabi frequency of the driving fields. When the squeezing bandwidth is much larger than the Rabi frequency, the spectrum consists of five lines with only the central and outer sidebands dependent on the phase. For a squeezing bandwidth much smaller than the Rabi frequency the number of lines in the spectrum and their phase properties depend on the frequency at which the squeezing and cavity modes are centered. When the squeezing and cavity modes are centered on the inner Rabi sidebands, the spectrum exhibits five lines that are completely independent of the squeezing phase with only the inner Rabi sidebands dependent on the squeezing correlations. Matching the squeezing and cavity modes to the outer Rabi sidebands leads to the disappearance of the inner Rabi sidebands and a strong phase dependence of the central line and the outer Rabi sidebands. We find that in this case the system behaves as an individual two-level system that reveals exactly the noise distribution in the input squeezed vacuum.
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I. INTRODUCTION

The radiative properties of atoms coupled to a squeezed vacuum field have been the subject of intense investigations in recent years [1]. Most well known are the effects of squeezing phase-dependent inhibition of spontaneous emission [2], the violation of the Boltzmann distribution of the populations of the atomic states [3,4], and modifications of multiphoton processes [3–6]. Such modifications have recently been confirmed experimentally in the first nonclassical spectroscopic experiment [7]. In the experiment the linear intensity dependence of the two-photon transition rate in a cascade three-level atom has been observed. This dependence is in contrast with the quadratic intensity dependence produced by classical light sources.

The most intriguing squeezing-induced effect is the phase-dependent inhibited spontaneous emission, first predicted by Gardiner [2]. He showed theoretically that the inhibition of the atomic dipole moment could occur in spontaneous emission from a two-level atom damped by a broadband squeezed vacuum field. With the addition of a coherent driving field this modification can lead to phase-dependent subnatural linewidths in the fluorescence and absorption spectra [8–12]. These subnatural linewidths lead to a partial suppression of spontaneous emission at certain frequencies; of course the total amount of emission is unchanged. Further work has demonstrated subnatural phase-dependent linewidths in the resonance fluorescence from three-level atoms [13–16]. Such effects might be more easily observed in three-level atoms since the driving field frequency can be distinguished more easily from the fluorescence field. However, no significant narrowing of the spectral lines was predicted for three-level atoms, making the phase-dependent effects even in three-level atoms difficult to observe experimentally. Although the phase-dependent narrowing of the spectral lines has not yet been observed, it appears that this observation is not very far off [17]. The major difficulty in any attempt to observe the phase-dependent spectral narrowing is that the present sources of the squeezed vacuum field generate a beam that can couple only to a small fraction of the modes surrounding the atom. It has been suggested [18,19] that the best way to overcome this difficulty is to place the atom inside an optical cavity. Inside the cavity the mode structure of the vacuum field is dramatically altered, allowing for an effective squeezed-vacuum-atom coupling to be achieved with an incoming squeezed-light beam propagating under a small solid angle.

Apart from this difficulty there is another important aspect of the interaction that must be considered in any attempt to observe the phase dependence of the spectral linewidth narrowing. The effect appears when another ''reference phase'' field is added to the system. The introduction of a coherent driving field leads to the dependence of the spectral linewidths on the relative phase of the squeezed vacuum and the driving field. As pointed out by Carmichael *et al.* [8,9] the phase dependence in the resonance fluorescence from a two-level atom is most evident for strong driving fields rather than for weak fields. However, for a strong driving field the system is no longer a two-level system. A two-level atom driven by a strong laser field is represented by a system of an infinite set of the energy states, called dressed states [20,21]. The dressed states of the system group into doublets; the neighboring doublets are separated by the frequency of the driving field, whereas dressed states in each doublet are separated by the Rabi frequency of the driving field. When the dressed system is coupled to a vacuum field the fluorescence will reveal the multilevel structure of the system. As pointed

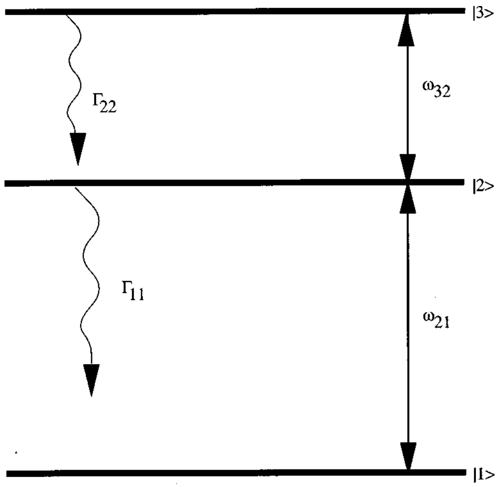

FIG. 1. Energy-level scheme for the three-level cascade configuration.

out above, the strongest phase dependence of the spectral linewidths has been predicted in a two-level system. The multifrequency transitions between the dressed states diminish the phase-dependent spectral narrowing. For example, for a two-level atom driven by a strong laser field only the central component of the fluorescence spectrum can be significantly narrowed [8–12]. For a three-level atom driven by two strong laser fields only a small narrowing can be observed in the central component of the spectrum [13–16].

Recently, Parkins *et al.* [22] have proposed a system that one might consider as a realization of a two-level dressed system coupled to a squeezed vacuum inside an optical cavity. In the model a two-level atom is located in a cavity driven by a squeezed vacuum field. They have shown that in the strong coupling regime, in which the dipole coupling strength between the atom and the cavity mode is much larger than the bandwidth of the squeezed vacuum, the system reveals the essential features of a two-level (single-frequency) system. Since there is no coherent driving field the features, however, are not sensitive to the squeezing phase. Significantly narrower fluorescence spectra have, however, been predicted by Swain [23] in two-level atom cases with a broadband squeezed vacuum with large photon numbers in the regime where the Rabi frequency and atomic linewidth are comparable. In the three-level atom case such effects are also seen [13–15] when the two transitions are coupled to two independent squeezed vacuum fields.

In this paper we present a modified version of the above model. We consider a three-level atom in a cascade configuration driven by two coherent laser fields inside an optical cavity. We examine the situation of how the cavity, operating in a weak coupling regime, driven by a finite-bandwidth squeezed vacuum, not only recovers the essential features of a two-level system, but also realizes a two-level system with phase dependent linewidths. Our proposal is based on the observation by Lewenstein *et al.* [24] that by driving an atom with a strong laser field inside an optical cavity one can dynamically change the transition rates between the dressed states, which can lead to the suppression of the spontaneous emission between some of the dressed states.

In our model we use the master equation technique based on dressed-atom states, first applied to narrow-bandwidth

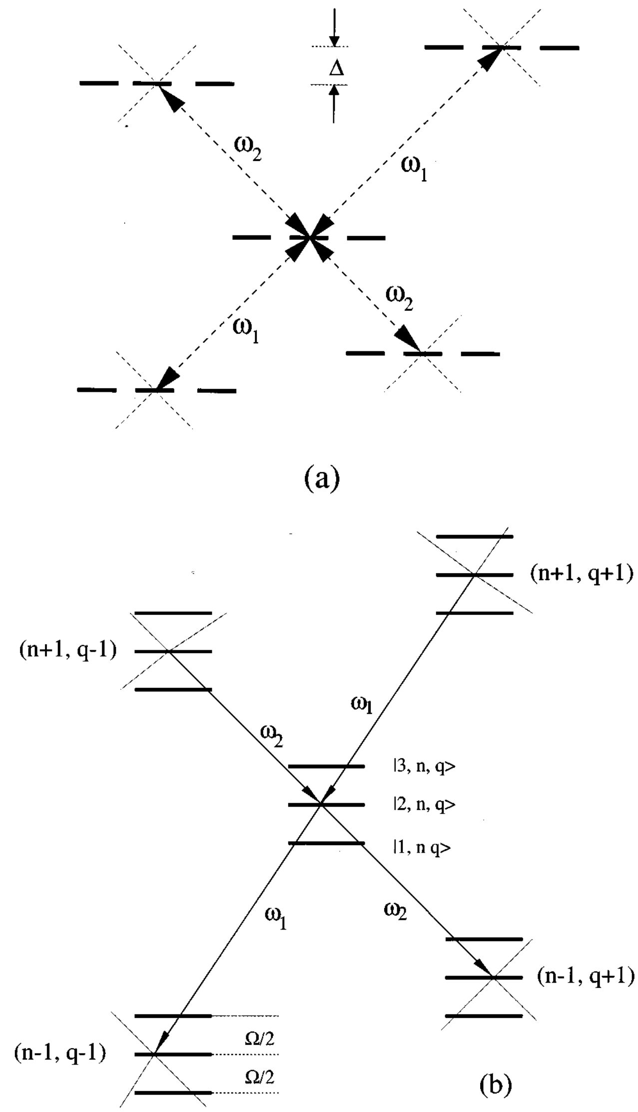

FIG. 2. Energy-level diagrams of (a) the undressed Hamiltonian, and (b) the dressed system. The manifold (n,q) is separated from the manifolds $(n\mp1,q\pm1)$ by the frequency ω_2 and from the manifolds $(n\pm1,q\pm1)$ by the frequency ω_1; Δ is the frequency difference $\omega_1-\omega_2$.

squeezed vacuum problems by Yeoman and Barnett [25], to calculate analytically the fluorescence spectrum of a three-level cascade system driven by two coherent laser fields and coupled to a finite-bandwidth squeezed vacuum inside an optical cavity. This approach has also recently been applied to calculate probe absorption spectra for driven three-level systems in narrow-bandwidth squeezed vacuum fields [26]. We assume that the squeezed vacuum is the output of a nondegenerate parametric amplifier operating below threshold. The nondegenerate parametric amplifier has proven to be the most successful source of a squeezed vacuum field suitable for nonclassical atomic spectroscopy [27,28]. In the derivation of the master equation, we first ''dress'' the atom with the laser fields and next couple the resulting system to the narrow-bandwidth squeezed vacuum and the cavity modes. The advantage of working in the dressed-atom picture is manifest in the simple analytical expressions for the spectral linewidths and the fluorescence spectrum [25,27,28].

This paper is organized as follows. In Sec. II we present the model and derive the equations of motion for the density matrix elements. In Sec. III we discuss the intensities and widths of the spectral features for free space and cavity situations. In Sec. IV, we present the fluorescence spectrum. We summarize our results in Sec. V. A detailed derivation of the master equation of the system is presented in the Appendix.

II. MODEL AND METHOD

We consider a three-level atom in the cascade configuration (Fig. 1) driven by two strong single-mode laser fields of frequencies ω_1 and ω_2, each of them coupled to one of the two atomic transitions. The laser fields are on resonance. The combined atom + driving fields system can be represented by the infinite number of energy states [29]. The states, called dressed states of the system, group into manifolds composed of triplets (Fig. 2),

$$|1,n,q\rangle=\frac{1}{\sqrt{2}\Omega}(i\Omega_1|1,n_1+1,n_2+1\rangle+\Omega|2,n_1,n_2+1\rangle - i\Omega_2|3,n_1,n_2\rangle),$$

$$|2,n,q\rangle=\frac{1}{\Omega}(\Omega_2|1,n_1+1,n_2+1\rangle+\Omega_1|3,n_1,n_2\rangle), \quad (1)$$

$$|3,n,q\rangle=\frac{1}{\sqrt{2}\Omega}(-i\Omega_1|1,n_1+1,n_2+1\rangle+\Omega|2,n_1,n_2+1\rangle + i\Omega_2|3,n_1,n_2\rangle),$$

with energies

$$E_{1nq}=E_{nq}-\tfrac{1}{2}\hbar\Omega,$$
$$E_{2nq}=E_{nq}, \quad (2)$$
$$E_{3nq}=E_{nq}+\tfrac{1}{2}\hbar\Omega,$$

where Ω_1 and Ω_2 are the Rabi frequencies of the driving fields, $\Omega=\sqrt{\Omega_1^2+\Omega_2^2}$, $q=n_1-n_2$, $n=n_1+n_2$, with n_1 (n_2) the number of photons in the laser mode of frequency ω_1 (ω_2) and E_{nq} is the energy of the nth manifold [see Eq. (A15)]. We assume that the dressed system is coupled to all other modes of the electromagnetic field, which are initially in the vacuum state, and that a part of the vacuum modes, those with frequencies around the laser frequency ω_1, are in a squeezed vacuum state.

The time evolution of the dressed system in a squeezed vacuum field centered around the frequency ω_1 is given by the master equation, derived in the Appendix, as

$$\begin{aligned}\frac{\partial\rho_I(t)}{\partial t}=&-\frac{1}{2}\sum_{i=1}^{2}\sum_{l=-2}^{2}\Gamma_{ii}\Bigg(\tilde{N}\left(\omega_i\pm\frac{l}{2}\Omega\right)\left\{\left[S^-\left(\omega_i\pm\frac{l}{2}\Omega\right),S^+\left(\omega_i\pm\frac{l}{2}\Omega\right)\rho_I(t)\right]+\left[\rho_I(t)S^-\left(\omega_i\pm\frac{l}{2}\Omega\right),S^+\left(\omega_i\pm\frac{l}{2}\Omega\right)\right]\right\}\\&+\left[\tilde{N}\left(\omega_i\pm\frac{l}{2}\Omega\right)+\left|D\left(\omega_i\pm\frac{l}{2}\Omega\right)\right|^2\right]\left\{\left[S^+\left(\omega_i\pm\frac{l}{2}\Omega\right),S^-\left(\omega_i\pm\frac{l}{2}\Omega\right)\rho_I(t)\right]\right.\\&\left.+\left[\rho_I(t)S^+\left(\omega_i\pm\frac{l}{2}\Omega\right),S^-\left(\omega_i\pm\frac{l}{2}\Omega\right)\right]\right\}\Bigg)+\frac{1}{2}\sum_{l=-2}^{2}\Gamma_{11}\Bigg(\tilde{M}\left(\omega_1\pm\frac{l}{2}\Omega\right)\left\{\left[S^+\left(\omega_1\mp\frac{l}{2}\Omega\right),S^+\left(\omega_1\pm\frac{l}{2}\Omega\right)\rho_I(t)\right]\right.\\&\left.+\left[\rho_I(t)S^+\left(\omega_1\mp\frac{l}{2}\Omega\right),S^+\left(\omega_1\pm\frac{l}{2}\Omega\right)\right]\right\}+\tilde{M}^*\left(\omega_1\pm\frac{l}{2}\Omega\right)\left\{\left[S^-\left(\omega_1\mp\frac{l}{2}\Omega\right),S^-\left(\omega_1\pm\frac{l}{2}\Omega\right)\rho_I(t)\right]\right.\\&\left.+\left[\rho_I(t)S^-\left(\omega_1\mp\frac{l}{2}\Omega\right),S^-\left(\omega_1\pm\frac{l}{2}\Omega\right)\right]\right\}\Bigg),\end{aligned} \quad (3)$$

where $S^+[\omega_i\pm(l/2)\Omega]$ and $S^-[\omega_i\pm(l/2)\Omega]=\{S^+[\omega_i\pm(l/2)\Omega]\}^*$ ($i=1,2$; $l=0,1,2$) are the raising and lowering operators for the transitions between the dressed states separated by the frequency $\omega_i\pm(l/2)\Omega$, Γ_{11} and Γ_{22} are the spontaneous decay rates respectively of the $|2\rangle\rightarrow|1\rangle$ and $|3\rangle\rightarrow|2\rangle$ transitions. The frequency-dependent parameters

$$\tilde{N}(\omega)=|D(\omega)|^2N(\omega),$$
$$|\tilde{M}(\omega)|=|D(\omega)|^2|M(\omega)| \quad (4)$$

are the effective squeezing parameters modified by the mode function of the reservoir. Expressions for N and M are given in Eq. (A5). ρ_I is the atom-laser modes density operator in the interaction picture. It is seen from Eq. (3) that any phase dependence is associated with the decay rate Γ_{11} of the $|2\rangle\rightarrow|1\rangle$ transition, which is a consequence of the choice of squeezed vacuum carrier frequency $\omega_s=\omega_1$. Moreover, the evolution of the density matrix depends on the vacuum mode density $|D(\omega)|^2$ and squeezing at different dressed-state transition frequencies. In free space the mode function is slowly varying compared to the transition frequencies of the atom and thus $|D(\omega)|^2=1$ after evaluation of Eq. (A32). In a cavity situation the mode function strongly depends on the frequency [30,31]. In this case $|D(\omega)|^2$ can be identified as the Airy function of the cavity. The explicit form of $|D(\omega)|^2$ depends on the type of cavity. For the so-called one-mode cavity the function $|D(\omega)|^2$ can be approximated by a Lorentzian peak centered on the cavity frequency ω_c. For a two-mode cavity, $|D(\omega)|^2$ can be approximated by two Lorentzians centered on $\omega_c \pm \delta_c$, where δ_c is the displacement of the cavity peaks from the central frequency.

The master equation (3) allows us to calculate the populations of the dressed states and coherences. Taking the diagonal matrix elements of each side of Eq. (3) for dressed-atom states $|i,n,q\rangle$ and summing over n,q, we obtain the following equations of motion for the populations of the dressed states:

$$\begin{aligned}\dot{\rho}_{11}=&-\frac{\Gamma}{8}\{2[\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)+|D(\omega_1-\tfrac{1}{2}\Omega)|^2]\rho_{11}\\&+[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1-\Omega)|^2\\&-2|\tilde{M}(\omega_1+\Omega)|\cos\varphi_s]\rho_{11}+|D(\omega_2-\Omega)|^2\rho_{11}\\&-2\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)\rho_{22}-2|D(\omega_2+\tfrac{1}{2}\Omega)|^2\rho_{22}\\&-[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1+\Omega)|^2\\&-2|\tilde{M}(\omega_1-\Omega)|\cos\varphi_s]\rho_{33}-|D(\omega_2+\Omega)|^2\rho_{33}\},\end{aligned}$$

$$\begin{aligned}\dot{\rho}_{22}=&-\frac{\Gamma}{8}\{-2[\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)+|D(\omega_1-\tfrac{1}{2}\Omega)|^2]\rho_{11}\\&+2[\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)]\rho_{22}\\&+2[|D(\omega_2+\tfrac{1}{2}\Omega)|^2+|D(\omega_2-\tfrac{1}{2}\Omega)|^2]\rho_{22}\\&-2[\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+|D(\omega_1+\tfrac{1}{2}\Omega)|^2]\rho_{33}\},\end{aligned}$$

$$\begin{aligned}\dot{\rho}_{33}=&-\frac{\Gamma}{8}\{-[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1-\Omega)|^2\\&-2|\tilde{M}(\omega_1+\Omega)|\cos\varphi_s]\rho_{11}-|D(\omega_2-\Omega)|^2\rho_{11}\\&-2[\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)]\rho_{22}-2|D(\omega_2-\tfrac{1}{2}\Omega)|^2\rho_{22}\\&+2[\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+|D(\omega_1+\tfrac{1}{2}\Omega)|^2]\rho_{33}\\&+[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1+\Omega)|^2\\&-2|\tilde{M}(\omega_1-\Omega)|\cos\varphi_s]\rho_{33}+|D(\omega_2-\Omega)|^2\rho_{33}\},\end{aligned}\tag{5}$$

where $\rho_{ii}=\Sigma_{nq}\rho_{inq,inq}$. Taking the matrix elements of each side of Eq. (3) between dressed-atom states $\langle i,n,q|$ and $|j,n-1,q+1\rangle$ and summing over n,q, we obtain the following equations of motion for the coherences between the dressed states:

$$\begin{aligned}\dot{\rho}_{12}=&-\frac{\Gamma}{8}\{\tfrac{1}{2}[2\tilde{N}(\omega_1)+|D(\omega_1)|^2+2|\tilde{M}(\omega_1)|\cos\varphi_s]+[2\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)+\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+|D(\omega_1-\tfrac{1}{2}\Omega)|^2]\\&+\tfrac{1}{2}[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1-\Omega)|^2-2|\tilde{M}(\omega_1+\Omega)|\cos\varphi_s]+\tfrac{1}{2}[|D(\omega_2)|^2+2|D(\omega_2-\tfrac{1}{2}\Omega)|^2\\&+2|D(\omega_2+\tfrac{1}{2}\Omega)|^2+|D(\omega_2-\Omega)|^2]\}\rho_{12}-\tfrac{1}{4}\tilde{M}(\omega_1-\tfrac{1}{2}\Omega)\Gamma\rho_{23},\end{aligned}$$

$$\begin{aligned}\dot{\rho}_{23}=&-\frac{\Gamma}{8}\{\tfrac{1}{2}[2\tilde{N}(\omega_1)+|D(\omega_1)|^2+2|\tilde{M}(\omega_1)|\cos\varphi_s]+[2\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)+|D(\omega_1+\tfrac{1}{2}\Omega)|^2]\\&+\tfrac{1}{2}[\tilde{N}(\omega_1+\Omega)+\tilde{N}(\omega_1-\Omega)+|D(\omega_1+\Omega)|^2-2|\tilde{M}(\omega_1+\Omega)|\cos\varphi_s]+\tfrac{1}{2}[|D(\omega_2)|^2+2|D(\omega_2-\tfrac{1}{2}\Omega)|^2\\&+2|D(\omega_2+\tfrac{1}{2}\Omega)|^2+|D(\omega_2+\Omega)|^2]\}\rho_{23}-\tfrac{1}{4}\tilde{M}^*(\omega_1-\tfrac{1}{2}\Omega)\Gamma\rho_{12},\end{aligned}$$

$$\begin{aligned}\dot{\rho}_{13}=&-\frac{\Gamma}{8}(2[2\tilde{N}(\omega_1)+|D(\omega_1)|^2+2|\tilde{M}(\omega_1)|\cos\varphi_s]+[\tilde{N}(\omega_1-\tfrac{1}{2}\Omega)+\tilde{N}(\omega_1+\tfrac{1}{2}\Omega)+|D(\omega_1-\tfrac{1}{2}\Omega)|^2+|D(\omega_1+\tfrac{1}{2}\Omega)|^2]\\&+\{\tfrac{1}{2}[2\tilde{N}(\omega_1-\Omega)]+2\tilde{N}(\omega_1+\Omega)+|D(\omega_1-\Omega)|^2+|D(\omega_1+\Omega)|^2-4|\tilde{M}(\omega_1+\Omega)|\cos\varphi_s\}\\&+2\{|D(\omega_2)|^2+\tfrac{1}{4}[|D(\omega_2+\Omega)|^2]+|D(\omega_2-\Omega)|^2]\})\rho_{13},\end{aligned}\tag{6}$$

where $\rho_{ij}=\Sigma_{nq}\rho_{inq,jn-1q+1}$ and the remaining equations of motion for the coherences ρ_{ii} $(i=1,2,3)$ obey the same equations of motion as the populations of the dressed states (3). For simplicity, we have assumed equal decay rates, $\Gamma_{11}=\Gamma_{22}=\Gamma$, and equal Rabi frequencies, $\Omega_1=\Omega_2$ in Eqs. (5) and (6). The coherences $\rho_{12}(\rho_{21})$ and $\rho_{23}(\rho_{32})$ correspond to the spectral lines at frequencies $\omega_1+\frac{1}{2}\Omega(\omega_1-\frac{1}{2}\Omega)$, the coherences $\rho_{13}(\rho_{31})$ correspond to the lines at $\omega_1+\Omega(\omega_1-\Omega)$, whereas the coherences ρ_{ii} $(i=1,2,3)$ correspond to the central component of the spectrum. In general, the spectrum is composed of five lines [29], the central and two pairs of sidebands located at $\omega_1\pm\frac{1}{2}\Omega$ and $\omega_1\pm\Omega$.

It is interesting to note from Eq. (5) that the time evolution of the populations of the dressed states depends on the Airy function of the cavity centered only at the dressed-state resonances corresponding to the inner and outer Rabi sidebands.

Therefore, in order to observe any fluorescence from the system, the cavity linewidth should be, at least, comparable to the inner Rabi sidebands frequency, or one should use a two-mode cavity with the modes centered on either the inner or outer Rabi sidebands. This suggests that the dynamics of the system coupled to the cavity modes could be completely different from that in free space, where $|D(\omega)|^2=1$ for all relevant ω. This also suggests that a two-mode rather than one-mode squeezed vacuum would be most useful in any attempt to observe the squeezing effects on the fluorescence spectrum. Therefore, we consider, as an input squeezed vacuum, the output of a nondegenerate parametric amplifier for which the squeezing parameters are [32]

$$N(\omega_i)=\frac{b_y^2-b_x^2}{8}\left\{\left[\frac{1}{(\omega_s-\omega_i-\delta_s)^2+b_x^2}-\frac{1}{(\omega_s-\omega_i-\delta_s)^2+b_y^2}\right]+\left[\frac{1}{(\omega_s-\omega_i+\delta_s)^2+b_x^2}-\frac{1}{(\omega_s-\omega_i+\delta_s)^2+b_y^2}\right]\right\},$$

$$|M(\omega_i)|=\frac{b_y^2-b_x^2}{8}\left\{\left[\frac{1}{(\omega_s-\omega_i-\delta_s)^2+b_x^2}+\frac{1}{(\omega_s-\omega_i-\delta_s)^2+b_y^2}\right]+\left[\frac{1}{(\omega_s-\omega_i+\delta_s)^2+b_x^2}+\frac{1}{(\omega_s-\omega_i+\delta_s)^2+b_y^2}\right]\right\}, \quad (7)$$

where ω_s is the carrier frequency of the squeezed vacuum and δ_s is the displacement from the carrier frequency at which the vacuum is maximally squeezed. The squeezing parameters are composed of two Lorentzians, with bandwidths given by

$$b_x=\frac{\gamma}{2}-|\varepsilon|,$$
$$b_y=\frac{\gamma}{2}+|\varepsilon|, \quad (8)$$

where γ is the cavity damping rate and $|\varepsilon|$ is the effective pump intensity. The maximum squeezing occurs at the threshold for parametric oscillation, i.e., as $|\varepsilon|\to\gamma/2$. We assume that the bandwidths of the nondegenerate parametric amplifier can be much smaller than the Rabi frequencies of the driving fields. This ensures that if the squeezed vacuum is coupled to a particular frequency ω_i, there is not a significant contribution at another frequency removed from it by the order of a Rabi frequency.

III. INTENSITIES AND WIDTHS OF THE SPECTRAL FEATURES

According to the dressed-atom model of Cohen-Tannoudji and Reynaud [21,29] the fluorescence spectrum is associated with transitions between dressed states of two neighboring manifolds and the frequencies of the spectral lines are given by the transition frequencies Eq. (A17). The intensities of the spectral lines are proportional to the total number of transitions between the corresponding dressed states. Thus, the stationary intensities $G[\omega_1\pm(l/2)\Omega]$ of the spectral lines are given by the product of the upper state population ρ_{ii} and the transition rate γ_{ij}, i.e., $G(\omega_k)=\tau\gamma_{ij}\rho_{ii}(\infty)$, where τ is an experimental term that depends on the atom–laser-field interaction time and is assumed large compared to Γ^{-1}.

The transition rates are defined by Fermi's golden rule as

$$\gamma_{ij}=\Gamma_{11}|\langle i,n,q|S_1^+|j,n-1,q-1\rangle|^2+\Gamma_{22}|\langle i,n,q|S_2^+|j,n-1,q-1\rangle|^2. \quad (9)$$

Using Eq. (A14), we find that the transition rates between the dressed states $|i,n,q\rangle$ and $|j,n-1,q+1\rangle$ are

$$\gamma_{11}=\gamma_{12}=\gamma_{13}=\gamma_{21}=\gamma_{23}=\gamma_{31}=\gamma_{32}=\gamma_{33}=\frac{\Gamma}{4},$$
$$\gamma_{22}=0. \quad (10)$$

In order to find the stationary populations of the dressed states and the linewidths of the spectral features, we have to solve respectively the system of three coupled equations (5) and the system of six coupled equations (6).

A. Free-space situation

First, assume that the dressed-atom system is coupled to the vacuum modes in free space. In this case $|D[\omega_1\pm(l/2)\Omega]|^2=1$, and it is easy to find from Eq. (3) that the dressed states are equally populated with

$$\rho_{11}(\infty)=\rho_{22}(\infty)=\rho_{33}(\infty)=\tfrac{1}{3}. \quad (11)$$

Interestingly, the stationary populations are independent of squeezing parameters and are the same for a broadband as well as a narrow-band squeezed vacuum. From Eqs. (10) and (11), we find that the spectrum is composed of five lines with the intensities

$$G_{\rm inel}(\omega_1)=\frac{\Gamma\tau}{6},$$
$$G(\omega_1\pm\tfrac{1}{2}\Omega)=\frac{\Gamma\tau}{6}, \quad (12)$$
$$G(\omega_1\pm\Omega)=\frac{\Gamma\tau}{12}.$$

The intensity of the inelastic central spectral line has been determined by subtracting from the total weight of the central peak the weight of the coherent component, which is given by

$$G_{\rm el}(\omega_2)=\tau[d_{11}\rho_{11}(\infty)+d_{22}\rho_{22}(\infty)+d_{33}\rho_{33}(\infty)]^2, \quad (13)$$

where $d_{ji}=\langle j,n-1,q+1|\mu|i,n,q\rangle$ are the transition dipole moments between the dressed states.

The linewidths of the spectral features, however, depend on the squeezing parameters and in particular on the bandwidth of the squeezed vacuum. In order to determine the widths of the spectral lines, we follow the approach of Cohen-Tannoudji and Reynaud [21], that the linewidths of the spectral components are given by the eigenvalues of the coupled equations of motion for the coherences ρ_{ii} and ρ_{ij}. For the central component of the spectrum, we find that the eigenvalues of Eq. (5) for a broadband squeezed vacuum, where $N[\omega_i \pm (l/2)\,\Omega] = N$, $|M[\omega_i \pm (l/2)\,\Omega]| = |M|$, and the free-space situation where $|D[\omega_i \pm (l/2)\,\Omega]|^2 = 1$, are

$$b_1 = 0,$$
$$b_2 = \tfrac{3}{4}(N+1)\Gamma, \tag{14}$$
$$b_3 = \tfrac{1}{4}(3N+3-2|M|\cos\varphi_s)\Gamma.$$

The eigenvalue b_1 corresponds to the elastic component of the spectrum, while the eigenvalues b_2 and b_3 correspond to the inelastic components at the frequency ω_1.

The linewidth of the inner sidebands at $\omega_1 \pm \frac{1}{2}\Omega$ is given by the damping rate of the coherences ρ_{12} and ρ_{23},

$$\eta_b = \tfrac{5}{8}(N+1 \pm \tfrac{1}{4}|M|)\Gamma, \tag{15}$$

and similarly, the linewidth of the outer sidebands at $\omega_1 \pm \Omega$ is given by the damping rate of the coherences ρ_{13} and ρ_{31},

$$\beta_b = (N+1+\tfrac{1}{4}|M|\cos\varphi_s)\Gamma. \tag{16}$$

It is seen from Eqs. (14)–(16) that only the central spectral peak and the outer Rabi sideband are phase dependent, with the inner Rabi sideband dependent only on the degree of squeezing $|M|$. It is worth noting the phase difference between the outer Rabi sidebands and the central spectral component. The two lines are out of phase by π, resulting in only one of the two being narrowed for a particular choice of phase, while the other is broadened.

If the squeezed vacuum has a finite bandwidth, such that the squeezing is confined to only specific modes, the linewidths of the spectral features and their phase properties are significantly different from the above for the broadband case. For narrow squeezed vacuum modes centered around $\omega_1 \pm \frac{1}{2}\Omega$, $N(\omega_1 \pm \frac{1}{2}\Omega) = N$, $|M(\omega_1 \pm \frac{1}{2}\Omega)| = |M|$, and the other squeezing parameters are zero. In this case the stationary populations of the dressed states are given by Eq. (11), and the widths of the spectral lines are

$$n_1 = 0,$$
$$n_2 = \tfrac{3}{4}(N+1)\Gamma, \tag{17}$$
$$n_3 = \tfrac{1}{4}(N+1)\Gamma$$

for the central component,

$$\eta_n = \tfrac{1}{8}(3N+5)\Gamma \pm \tfrac{1}{4}|M|\Gamma \tag{18}$$

for the inner Rabi sidebands, and

$$\beta_n = \tfrac{1}{4}(N+4)\Gamma \tag{19}$$

for the outer Rabi sidebands.

Interestingly, the spectral linewidths are independent of the squeezing phase and all are broadened due to the presence of thermal photons at the frequencies $\omega_1 \pm \frac{1}{2}\Omega$. However, for squeezing frequencies centered around $\omega_1 \pm \Omega$, i.e., $N(\omega_1 \pm \Omega) = N$, $|M(\omega_1 \pm \Omega)| = |M|$, $|D[\omega_i \pm (l/2)\,\Omega]|^2 = 1$ and the other squeezing parameters equal to zero, we find that the width of the central spectral peak is given by

$$f_1 = 0,$$
$$f_2 = \tfrac{3}{4}\Gamma, \tag{20}$$
$$f_3 = \tfrac{1}{4}(2N+3-2|M|\cos\varphi_s)\Gamma,$$

where, as before, the eigenvalue f_1 corresponds to the elastic component of the spectrum, while the eigenvalues f_2 and f_3 correspond to the inelastic components at the frequency ω_1. Similarly, for the sideband at $\omega_1 \pm \frac{1}{2}\Omega$ we find that the linewidth is given by

$$\eta_f = \tfrac{1}{8}(N+5-|M|\cos\varphi_s)\Gamma, \tag{21}$$

and for the outer Rabi sideband at $\omega_2 \pm \Omega$ the linewidth is

$$\beta_f = \tfrac{1}{4}(N+4-|M|\cos\varphi_s)\Gamma. \tag{22}$$

In this situation, *all* spectral lines are phase dependent and, in contrast to the broadband case, the spectral lines can be narrowed below the ordinary vacuum level. It happens for all values of N and $\varphi_s = 0$. The situation of the atom being damped by an ordinary vacuum can be obtained by setting $N = |M| = 0$ in Eqs. (14)–(16), which results in linewidths for the spectral components equal to those determined in [33,29]. It can be seen that in such a situation, the linewidths are not phase dependent.

B. Cavity situation

When the dressed system is located inside an optical cavity, the spectral intensities and linewidths differ significantly from those derived previously for the free-space situation. Firstly, we consider a two-mode narrow-bandwidth cavity and a two-mode narrow-bandwidth squeezed vacuum, both centered on $\omega_1 \pm \frac{1}{2}\Omega$. From Eqs. (5) and (6), we find that in this case the populations and coherences, similar to those in free space, are independent of the squeezing phase. The stationary populations of the dressed states are dependent on the thermal fluctuations N, and are given by

$$\rho_{11} = \rho_{33} = \frac{N}{3N+1}, \qquad \rho_{22} = \frac{N+1}{3N+1}, \tag{23}$$

where $N = \widetilde{N}(\omega_1 \pm \frac{1}{2}\Omega)$.

Similarly, we find from Eq. (6) that the linewidths of the central component and the outer Rabi sidebands depend only on N and are given by

$$c_1 = 0,$$
$$c_2 = \tfrac{1}{4}(N+1)\Gamma, \tag{24}$$
$$c_3 = \tfrac{1}{4}(3N+1)\Gamma$$

for the central component and

$$\beta_c = \tfrac{1}{4}(N+1)\Gamma \tag{25}$$

for the outer Rabi sidebands. The inner Rabi sidebands, however, are composed of two lines of the same frequency but different linewidth, dependent on the squeezing correlations $|M|$ as

$$\eta_c = \tfrac{1}{8}(3N+1\pm 2|M|)\Gamma. \tag{26}$$

When the two-mode cavity and the two-mode squeezed vacuum are centered on the outer Rabi sidebands at $\omega_1 \pm \Omega$, the stationary populations are independent of the squeezing parameters and are given by

$$\rho_{11} = \rho_{33} = \tfrac{1}{2}, \qquad \rho_{22} = 0. \tag{27}$$

Then we find that the intensities of the spectral lines are

$$G_{\text{inel}}(\omega_1) = \frac{\Gamma\tau}{4},$$
$$G(\omega_1 \pm \tfrac{1}{2}\Omega) = 0, \tag{28}$$
$$G(\omega_1 \pm \Omega) = \frac{\Gamma\tau}{8}.$$

In this case, the modified environment results in the spectrum effectively being reduced to three lines. With the cavity modes maximized at the frequency $\omega_1 \pm \Omega$ we see from Eqs. (6) and (27) that the population ρ_{22} decouples from the remaining equations of motion, leaving the population distributed between the states $|3,n,q\rangle$ and $|1,n,q\rangle$ only. Thus, in this situation the dressed system reduces to that of a driven two-level atom and we would only expect to observe three spectral lines in the fluorescent spectra: the central peak and the outer Rabi sidebands at $\omega_1 \pm \Omega$.

The width of the spectral lines is given by the eigenvalues of the equations of motion. For the central peak

$$c_1 = 0, \qquad c_2 = \tfrac{1}{2}(N + \tfrac{1}{2} - |M|\cos\varphi_s)\Gamma \tag{29}$$

and for the outer Rabi sideband

$$\beta_c = \tfrac{1}{4}(N + \tfrac{1}{2} - |M|\cos\varphi_s)\Gamma. \tag{30}$$

Clearly, incorporating into the system a two-mode cavity centered on the outer Rabi sidebands can lead to narrowing of all the spectral lines to zero for $\varphi_s = 0$ and $N \gg 1$. The latter condition leads to the approximation $|M| \approx N + \tfrac{1}{2}$, which can be observed from the inequality directly following Eq. (A5); thus the linewidths approach zero in the limit of

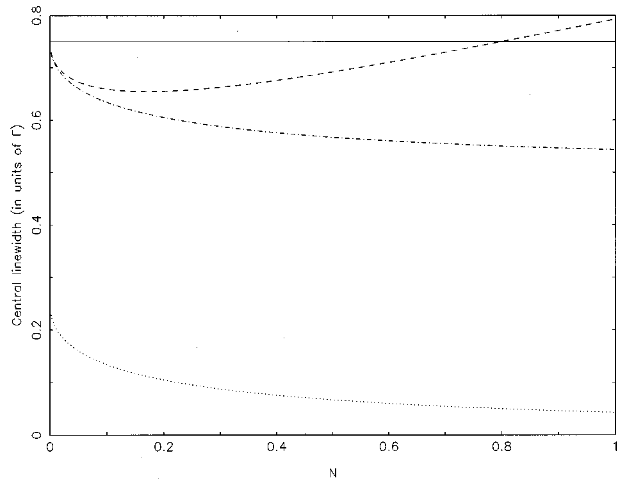

FIG. 3. Linewidth of the central spectral component (in units of Γ) as a function of N for the three different squeezed reservoirs: (a) broadband squeezed vacuum (---), (b) finite-bandwidth squeezed vacuum with $N(\omega_1 \pm \Omega) = N$ and $|M(\omega_1 \pm \Omega)| = |M|$ (-·-) in free space, and (c) finite-bandwidth squeezed vacuum in a cavity with $|D(\omega_1 \pm \Omega)|^2$ and $|D(\omega)|^2 = 0$ for all other ω (···). The solid line represents the normal vacuum linewidth (in units of Γ).

large squeezing when $\varphi_s = 0$. It is interesting to note from Eqs. (29) and (30) that the linewidths of the spectral features reveal exactly the noise distribution in the input squeezed vacuum. It is easy to show from Eq. (A5) that the fluctuations in the quadrature components of the input squeezed vacuum are

$$F = \langle (\Delta a_\varphi)^2 \rangle = \tfrac{1}{2}(N + \tfrac{1}{2} - |M|\cos\varphi_s). \tag{31}$$

Therefore, the potential for narrowing of the spectral lines is not impaired in the presence of a two-mode cavity centered on the outer Rabi sidebands. We may conclude that in this

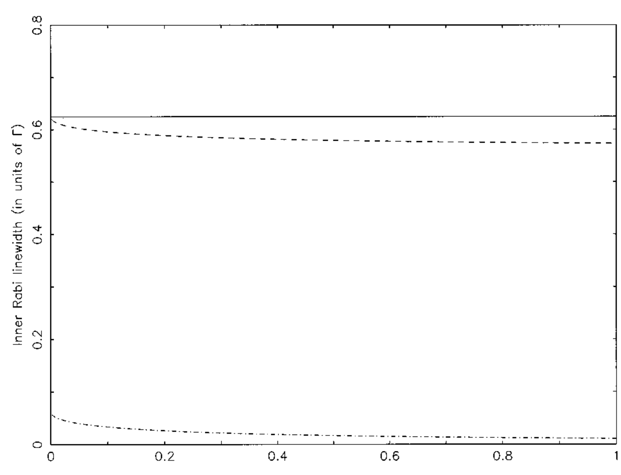

FIG. 4. Linewidth of the inner Rabi sideband (in units of Γ) as a function of N for the two different squeezed reservoirs: (a) finite bandwidth squeezed vacuum with $N(\omega_1 \pm \Omega) = N$ and $|M(\omega_1 \pm \Omega)| = |M|$ (---) in free space and (b) in a cavity with $|D(\omega_1 \pm \Omega)|^2$ and $|D(\omega)|^2 = 0$ for all other ω (-·-). The solid line represents the normal vacuum linewidth (in units of Γ).

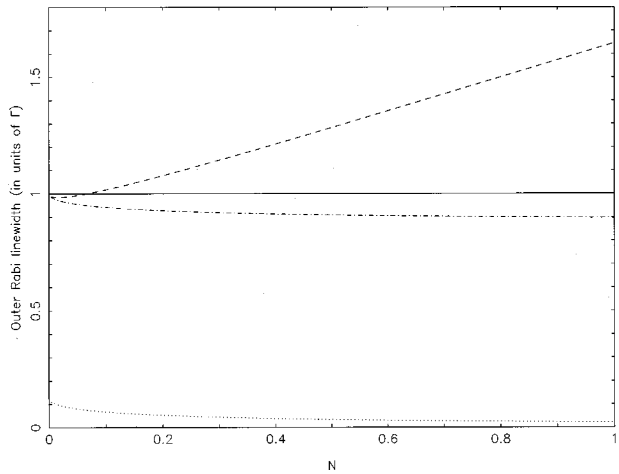

FIG. 5. Linewidth of the outer Rabi sideband (in units of Γ) as a function of N for the three different squeezed reservoirs: (a) broadband squeezed vacuum (---), (b) finite bandwidth squeezed vacuum with $N(\omega_1 \pm \Omega) = N$ and $|M(\omega_1 \pm \Omega)| = |M|$ (-·-) in free space, and (c) in a cavity with $|D(\omega_1 \pm \Omega)|^2$ and $|D(\omega)|^2 = 0$ for all other ω (···). The solid line represents the normal vacuum linewidth (in units of Γ).

case each of the transitions individually couples to the input squeezed vacuum and acts as a single two-level system with phase-dependent noise.

To conclude our discussion of the spectral linewidths in the free space and cavity situations, we plot in Figs. 3–5 the linewidth of the spectral lines as a function of N and three different squeezed vacua. Figure 3 shows the width of the central spectral line. In the case of the system interacting with a broadband squeezed vacuum, line narrowing of $\approx 13\%$ below the normal vacuum width is possible for small values of N. However, when N approaches unity the linewidth becomes broader than the normal vacuum width. For the finite bandwidth squeezed vacuum case it can be seen that in the limit of large squeezing, i.e., as $N \to \infty$, a reduction of $\approx 33\%$ below the normal vacuum linewidth is possible. Finally, for the atom in a cavity, the reduction of spontaneous emission outside the finite frequency intervals $\omega \pm \Omega$ results in less noise present at the fluorescent frequencies, thus allowing further reduction of the linewidths. Indeed, it can be seen that in the limit of large squeezing the linewidth can be reduced by 100%.

The width of the spectral Rabi sidebands at $\omega \pm \frac{1}{2}\Omega$ is plotted in Fig. 4 for the finite-bandwidth squeezed vacuum as there was no phase dependence for a broadband squeezed vacuum, only a dependence on the *degree* of squeezing $|M|$, and the spectral line is not present when the atom is coupled to a squeezed vacuum in a cavity. We see from Fig. 4 that in the limit of large squeezing a linewidth reduction of up to 10% is possible.

Finally, for the Rabi sidebands at $\omega \pm \Omega$ we see from Fig. 5 that when the atom is damped by a broadband squeezed

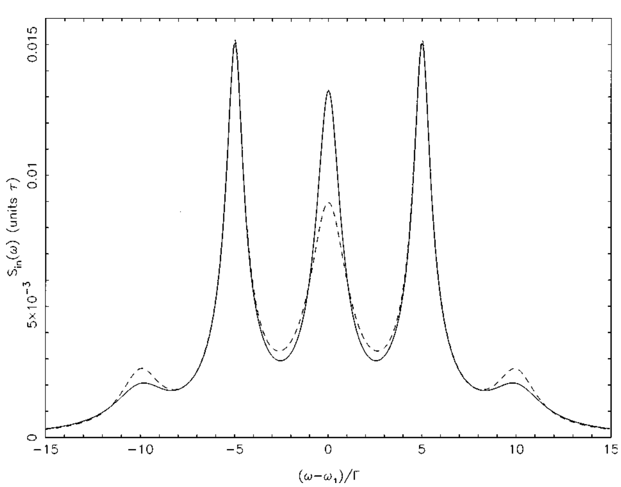

FIG. 6. Resonance fluorescence spectra (in units of τ) vs the frequency detuning $\omega - \omega_1$ (in units of Γ) for the three-level atom damped by a broadband squeezed vacuum with $\Omega = 10\Gamma$, $N = 0.5$, $|M| = [N(N+1)]^{1/2}$, and $\varphi_s = 0$ (—), π (---).

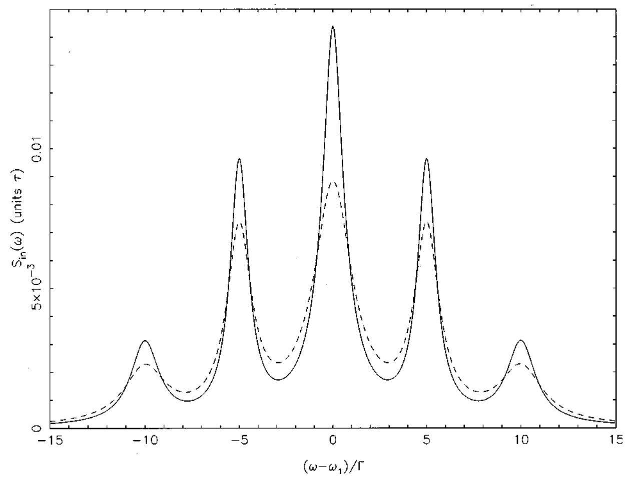

FIG. 7. Resonance fluorescence spectra (in units of τ) vs the frequency detuning $\omega - \omega_1$ (in units of Γ) for the three-level atom damped by a finite bandwidth squeezed vacuum in free space, where $N(\omega_1 \pm \Omega) = N$, $|M(\omega_1 \pm \Omega)| = |M|$, and $|D(\omega)|^2 = 1$, with $N = 0.5$, $\Omega = 10\Gamma$, $|M| = [N(N+1)]^{1/2}$, and $\varphi_s = 0$ (—), π (---).

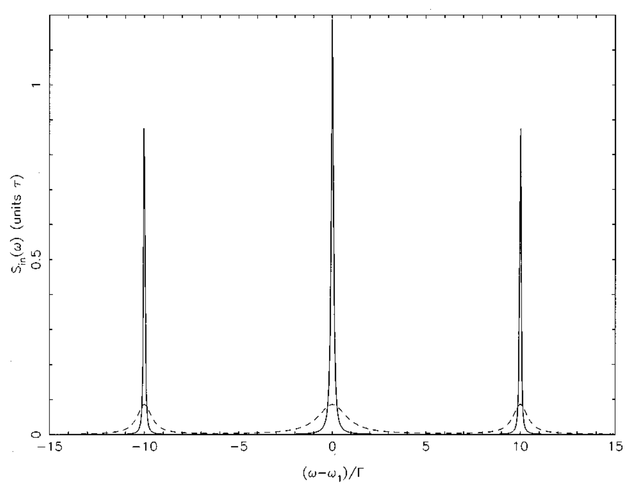

FIG. 8. Resonance fluorescence spectra (in units of τ) vs the frequency detuning $\omega - \omega_1$ (in units of Γ) for the three-level atom damped by a finite bandwidth squeezed vacuum in a cavity both centered on the outer Rabi sideband where $N(\omega_1 \pm \Omega) = N$, $|M(\omega_1 \pm \Omega)| = |M|$, $|D(\omega_1 \pm \Omega)|^2 = 1$, and $|D(\omega)|^2 = 0$ for all other ω, with $N = 0.5$, $\Omega = 10\Gamma$, $|M| = [N(N+1)]^{1/2}$, and $\varphi_s = 0$ (—), π (---).

vacuum only a limited amount of line reduction is possible for small N. This reduction is lost when N approaches 0.1, being replaced with a linearly increasing linewidth. If the atom is coupled by a narrow-bandwith squeezed vacuum maximized at $\omega_1 \pm \Omega$ we see that in the limit of large squeezing it is possible to narrow the linewidth by 12%. However, if the atom is then placed in a cavity, it is possible to reduce the linewidth by 100% below the normal vacuum linewidth due to the suppression of spontaneous emission.

IV. FLUORESCENCE SPECTRUM

Having available the intensities of the spectral lines and their widths, we can write down the entire spectrum of the fluorescent field emitted at the $|2\rangle \leftrightarrow |1\rangle$ transition as

$$S(\omega) = G_{\text{inel}}(\omega_1) \frac{\lambda/\pi}{(\omega-\omega_1)^2+\lambda^2} + G(\omega_1 - \tfrac{1}{2}\Omega) \frac{\eta/\pi}{(\omega-\omega_1+\tfrac{1}{2}\Omega)^2+\eta^2} + G(\omega_1 + \tfrac{1}{2}\Omega) \frac{\eta/\pi}{(\omega-\omega_1-\tfrac{1}{2}\Omega)^2+\eta^2} + G(\omega_1 - \Omega) \frac{\beta/\pi}{(\omega-\omega_1+\Omega)^2+\beta^2} + G(\omega_1 + \Omega) \frac{\beta/\pi}{(\omega-\omega_1-\Omega)^2+\beta^2}, \tag{32}$$

where λ refers to the linewidth of the central peak, and η and β are the linewidths of the inner and outer Rabi sidebands, respectively. Substituting the linewidths into Eq. (32) along with the intensities of the spectral features calculated earlier and assuming that $\Gamma_{11} = \Gamma_{22}$ and $\Omega = 10\Gamma$ we are able to plot the fluorescent spectra for the atom damped by a variety of reservoirs in order to emphasize the dependency of the spectra on squeezing phase and the squeezing bandwidth. In Fig. 6 we plot the fluorescent spectrum for an atom damped by a broadband squeezed vacuum with two different phases $\varphi_s = 0$ and π. It is apparent that for the different values of φ_s only the outer Rabi sideband and the central peak are affected.

The fluorescent spectrum of the system damped in free space by a finite bandwidth squeezed vacuum maximized at the frequencies $\omega_1 \pm \Omega$ is shown in Fig. 7. Here all lines in the fluorescent spectrum are phase dependent.

Placing the system in a two-mode cavity with the modes centered on the outer sidebands results in the loss of the inner Rabi sidebands from the fluorescent spectrum. This is shown in Fig. 8, and indicates that inside the cavity the system effectively behaves as a two-level system. In this case all spectral lines are very narrow and phase dependent. The linewidths can even be reduced to zero in the limit of large squeezing.

V. SUMMARY

In this paper we have examined the phase dependence of the fluorescence spectrum of a three-level atom driven by two resonant laser fields and damped by a finite-bandwidth squeezed vacuum. We have assumed that the bandwidth of the squeezing is much smaller than the Rabi frequency of the driving fields, but much larger than the natural atomic linewidth, in order to ensure that the behavior of the reduced density operator for the system is Markovian. We derived the master equation in the dressed-atom basis, which removes the fast-time-scale Rabi oscillations from the interaction picture. The system now evolves on a much longer time scale, allowing us to consider finite-bandwidth reservoir effects using the Markoff approximation.

We specifically examined the Cascade system where the squeezed vacuum carrier frequency was equal to the dressed-atom frequency, $\omega_s = \omega_1$. Assuming the source of the squeezed vacuum is a nondegenerate parametric amplifier (NDPA) operating below the threshold, we have found that all spectral lines show a dependence on the squeezing phase and can be significantly narrowed below the ordinary vacuum level. The phase dependence and the narrowing strongly depend on the frequency at which the squeezing and cavity modes are centered. When the system interacts with a narrow-bandwidth squeezed vacuum in free space and the squeezed modes are centered on the inner Rabi sidebands the spectrum exhibits five lines that are completely independent of the squeezing phase. Matching the squeezing modes to the outer Rabi sidebands results in all the spectral lines dependent on the phase with the possibility of a 33% reduction of the spectral linewidths below the vacuum level.

Placing the system in a cavity results in the further narrowing of the spectral lines due to the modification of the density of modes interacting with the atom. Along with avoiding the experimentally difficult situation of squeezing all modes coupled to the atom, the cavity modifies the atomic spontaneous rates, which can even reduce the three-level atom dynamics to that characteristic of a two-level atom. As a consequence, the linewidths of the peaks in the fluorescent spectra are reduced compared to the free-space situation. Therefore, when the cavity modes are in a squeezed vacuum state, a further reduction of the spectral lines is possible, resulting in the linewidths being reduced to zero, with the appropriate choice of phase and in the limit of large squeezing.

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF THE MASTER EQUATION (3)

The time evolution of a system interacting with a reservoir is given by the equation of motion for the reduced density operator ρ. In the interaction picture, and after the Born approximation, the master equation is given by [34]

$$\frac{\partial \rho_I(t)}{\partial t} = -\frac{1}{\hbar^2}\int_0^t \text{Tr}_R[V_I(t),[V_I(t'),\rho_R(0)\rho_I(t')]]dt', \tag{A1}$$

where $V(t)$ is the interaction Hamiltonian for the system and reservoir, which in the rotating wave approximation is given by

$$V_I(t) = -\frac{i}{2}\hbar \sum_\lambda [\Omega_\lambda^{(1)} S_1^+(t) a_\lambda(t) - (\Omega_\lambda^{(1)})^* a_\lambda^\dagger(t) S_1^-(t) + \Omega_\lambda^{(2)} S_2^+(t) a_\lambda(t) - (\Omega_\lambda^{(2)})^* a_\lambda^\dagger(t) S_2^-(t)]. \quad \text{(A2)}$$

We specifically assume the system is a three-level atom in the cascade configuration (Fig. 1) where $S_i^+(t)$ $[S_i^-(t)]$ is the raising [lowering] operator of the ith transition ($i=1,2$). The vacuum Rabi frequencies $\Omega_\lambda^{(1)}$ and $\Omega_\lambda^{(2)}$ are given by

$$\Omega_\lambda^{(1)} = (\vec{\mu}_{21} \cdot \vec{e}_\lambda) \left(\frac{2\omega_\lambda}{\hbar \epsilon_0 V} \right)^{1/2} D(\omega_\lambda) \quad \text{(A3)}$$

and

$$\Omega_\lambda^{(2)} = (\vec{\mu}_{32} \cdot \vec{e}_\lambda) \left(\frac{2\omega_\lambda}{\hbar \epsilon_0 V} \right)^{1/2} D(\omega_\lambda), \quad \text{(A4)}$$

where $\vec{e}_\lambda$ is the unit polarization vector, $\vec{\mu}_{21}$ and $\vec{\mu}_{32}$ are the matrix elements for the transition dipole moments, respectively, for the $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |2\rangle$ transitions. The frequency-dependent parameter $D(\omega_\lambda)$ represents the mode function at the position of the atom [30,31]. The system is coupled to a multimode reservoir, which in a squeezed vacuum state is characterized by the following correlation functions of the field operators:

$$\begin{aligned}
\langle a_\lambda a_\mu^\dagger \rangle &= N(\omega_\lambda) + 1, \quad \omega_\lambda = \omega_\mu, \\
\langle a_\lambda^\dagger a_\mu \rangle &= N(\omega_\lambda), \quad \omega_\lambda = \omega_\mu, \\
\langle a_\lambda a_\mu \rangle &= M(\omega_\lambda), \quad \omega_\lambda + \omega_\mu = 2\omega_s, \\
\langle a_\lambda^\dagger a_\mu^\dagger \rangle &= M^*(\omega_\lambda), \quad \omega_\lambda + \omega_\mu = 2\omega_s.
\end{aligned} \quad \text{(A5)}$$

In Eq. (A5) the parameters $N(\omega_\lambda)$ and $M(\omega_\lambda)$ characterize squeezing such that $|M(\omega_\lambda)|^2 \leqslant N(\omega_\lambda)[N(2\omega_s - \omega_\lambda) + 1]$, where equality holds for a minimum uncertainty state, and ω_s is the carrier frequency of the squeezed vacuum field. The complex parameter $M(\omega_\lambda) = M(2\omega_s - \omega_\lambda) = |M(\omega_\lambda)| \exp(i\varphi_s)$, where $|M(\omega_\lambda)|$ is the degree of squeezing and φ_s is the phase of the squeezed vacuum, results from the correlations between the field mode at frequency ω_λ, and the mode at frequency $2\omega_s - \omega_\lambda$. The parameter $N(\omega_\lambda)$ is proportional to the number of photons in the field modes. Nonzero M implies that the reservoir density operator $\rho_R(0)$ does not commute with the reservoir Hamiltonian; thus the squeezed vacuum is not a reservoir stationary state.

The coupling $V_I(t)$ can be written as a linear combination of the products of the system S_a and reservoir R_a operators,

$$V_I = \sum_a S_a(t) R_a(t), \quad \text{(A6)}$$

where $S_a = S_j^\pm$ and $R_a = \pm \frac{1}{2} \Sigma_\lambda \Omega_\lambda^{(j)} a_\lambda^\pm \exp(\pm i\omega_\lambda t)$ ($j=1,2$). Rewriting Eq. (A1) in terms of Eq. (A6), expanding the double commutator, and obeying the cyclic properties of the trace, we find that

$$\begin{aligned}
i\hbar \frac{\partial \rho_I(t)}{\partial t} = {} & \sum_a \langle R_a(t) \rangle [S_a(t), \rho_I(0)] \\
& + \frac{1}{i\hbar} \sum_{ab} \int_0^t dt \langle R_a(t) R_b(t-\tau) \rangle \\
& \times [S_a(t), S_b(t-\tau) \rho_I(t-\tau)] \\
& + \frac{1}{i\hbar} \sum_{ab} \int_0^t dt \langle R_b(t-\tau) R_a(t) \rangle \\
& \times [S_b(t-\tau) \rho_I(t-\tau), S_a(t)],
\end{aligned} \quad \text{(A7)}$$

where we have made the substitution $t' = t - \tau$. In Eq. (A7),

$$\begin{aligned}
\langle R_a(t) \rangle &= \mathrm{Tr}_R[\rho_R(0) R_a(t)], \\
\langle R_a(t_1) R_b(t_2) \rangle &= \mathrm{Tr}_R[\rho_R(0) R_a(t_1) R_b(t_2)]
\end{aligned} \quad \text{(A8)}$$

are the first- and second-order reservoir correlation functions. Generally, the first-order correlation function depends on time and the second-order correlation function depends on the time difference. Typically, the correlation functions in Eq. (A8) decay to zero within a very short correlation time τ_c, that is,

$$\begin{aligned}
\langle R_a(t) \rangle &\to 0, \quad t \gg \tau_c \\
\langle R_a(t_1) R_b(t_2) \rangle &\to 0, \quad |t_1 - t_2| \gg \tau_c.
\end{aligned} \quad \text{(A9)}$$

If the system evolves on a much longer time scale than the reservoir correlation time τ_c then the reservoir can be considered Markovian. Previous Markovian master equations for a system interacting with a squeezed vacuum [2,8,9] have relied on the squeezed vacuum being δ correlated on the time scales of the natural atomic lifetime and the Rabi oscillations induced by the driving fields, which translates in the frequency domain to making the bandwidth of the squeezed vacuum, Γ_S, much larger than the natural atomic linewidth and the Rabi frequency, that is, $\Gamma_S \gg \Omega, \Gamma$, where Γ is the natural atomic bandwidth.

The main idea here is to first ''dress'' the atom by the driving fields, and next couple the remaining dressed-atom system to the frequency-dependent reservoir. In the dressed-atom basis, the ''system'' evolves on the much longer time scale, Γ^{-1} as we have effectively removed the short time scale Rabi oscillations from the interaction picture system density operator. Thus, deriving the master equation in the dressed-atom basis allows us to consider finite-bandwidth effects, as we only have to assume that, $\Gamma_S \gg \Gamma$, i.e., the reservoir bandwidth is much greater than the natural atomic bandwidth.

The dressed-atom states are the eigenstates of the Hamiltonian [29]

$$H_S = H_{AF} + H_{IN}, \tag{A10}$$

where

$$H_{AF} = \hbar[\omega_1|2\rangle\langle 2| + (\omega_1+\omega_2)|3\rangle\langle 3|] + \hbar\omega_1 b_1^\dagger b_1 + \hbar\omega_2 b_2^\dagger b_2 \tag{A11}$$

is the unperturbed Hamiltonian of the cascade three-level atom plus driving laser fields of frequencies ω_1 and ω_2, and

$$H_{IN} = \tfrac{1}{2} i\hbar g_1(S_1^+ b_1 - b_1^\dagger S_1^-) + \tfrac{1}{2} i\hbar g_2(S_2^+ b_2 - b_2^\dagger S_2^-) \tag{A12}$$

is the interaction Hamiltonian between the atom and the driving fields. In Eq. (A11) and Eq. (A12) $b_1(b_2)$ and $b_1^\dagger(b_2^\dagger)$ are, respectively, the annihilation and creation operators for the driving field of frequency $\omega_1(\omega_2)$ and $g_1(g_2)$ are the coupling constants between the atom and the quantized driving field. We assume that the driving fields are single-mode laser fields in the coherent states $|\alpha_1\rangle$ and $|\alpha_2\rangle$. The dressed-atom states of the coupled system will be designated $|i,n,q\rangle$, where for convenience we write $n=n_1+n_2$ as the total number of photons in the laser fields and $q=n_1-n_2$ as the photon number difference. The states of the uncoupled atom+driving fields system are given by $|i,n_1,n_2\rangle = |i\rangle \otimes |n_1\rangle \otimes |n_2\rangle$. We assume that the mean photon number $\overline{n_i} = |\alpha_i|^2$ of the driving fields is much larger than the width of the photon number Δn; then the laser field fluctuations are not significant at frequencies removed from the driving frequency.

For the cascade configuration, the undressed states (eigenstates of H_{AF}) form manifolds composed of threefold nearly degenerate states $|1,n_1+1,n_2+1\rangle$, $|2,n_1,n_2+1\rangle$, and $|3,n_1,n_2\rangle$ of the energy

$$E_{nq} = \hbar[(n_1+1)\omega_1 + (n_2+1)\omega_2]. \tag{A13}$$

The inclusion of the interaction H_{IN} lifts the degeneracy resulting in an energy level scheme composed of triplets (Fig. 2),

$$|1,n,q\rangle = \frac{1}{\sqrt{2}\Omega}(i\Omega_1|1,n_1+1,n_2+1\rangle + \Omega|2,n_1,n_2+1\rangle - i\Omega_2|3,n_1,n_2\rangle),$$

$$|2,n,q\rangle = \frac{1}{\Omega}(\Omega_2|1,n_1+1,n_2+1\rangle + \Omega_1|3,n_1,n_2\rangle), \tag{A14}$$

$$|3,n,q\rangle = \frac{1}{\sqrt{2}\Omega}(-i\Omega_1|1,n_1+1,n_2+1\rangle + \Omega|2,n_1,n_2+1\rangle + i\Omega_2|3,n_1,n_2\rangle),$$

with energies

$$E_{1nq} = E_{nq} - \tfrac{1}{2}\hbar\Omega,$$
$$E_{2nq} = E_{nq}, \tag{A15}$$
$$E_{3nq} = E_{nq} + \tfrac{1}{2}\hbar\Omega,$$

where $\Omega = \sqrt{\Omega_1^2+\Omega_2^2}$, with $\Omega_1 = |\Omega_1|\exp(i\varphi_1) = g_1\sqrt{\overline{n_1}}$, and $\Omega_2 = |\Omega_2|\exp(i\varphi_2) = g_2\sqrt{\overline{n_2}}$. We have assumed the driving fields to be sufficiently intense that the variation of the n photon Rabi frequencies with n_1 and n_2 has been neglected and the photon numbers replaced by the average photon numbers $\overline{n_1}$ and $\overline{n_2}$ in the laser modes. We set the phase of the lasers, φ_1 and φ_2, equal to zero for convenience. From the energy-level diagram in Fig. 2, it is apparent that the possibility of fluorescence exists at the frequencies,

$$\omega_{ij} = \hbar^{-1}(E_{inq} - E_{jnq}), \tag{A16}$$

given by

$$\omega_{11} = \omega_{22} = \omega_{33} = \omega_2,$$
$$\omega_{21} = \omega_{32} = \omega_2 + \frac{\Omega}{2},$$
$$\omega_{12} = \omega_{23} = \omega_2 - \frac{\Omega}{2}, \tag{A17}$$
$$\omega_{31} = \omega_2 + \Omega,$$
$$\omega_{13} = \omega_2 - \Omega.$$

These transition frequencies between the dressed states indicate that up to five lines can be observed in the fluorescence spectrum.

The transformation in Eq. (A14) is unitary and can be easily inverted to give the undressed atomic states in terms of the dressed states. This allows us to express the original atomic operators as a linear combination of their dressed-state counterparts in the interaction picture; thus

$$S_1^+(t)=[S_1^-(t)]^*=\exp[-i(H_S+H_R)t/\hbar]|2,n_1,n_2\rangle\langle 1,n_1+1,n_2|\exp[i(H_S+H_R)t/\hbar]$$
$$=\left[S^+(\omega_1)+S^+\left(\omega_1-\frac{\Omega}{2}\right)\exp\left(-i\frac{\Omega}{2}t\right)+S^+\left(\omega_1+\frac{\Omega}{2}\right)\exp\left(i\frac{\Omega}{2}t\right)\right.$$
$$\left.+S^+(\omega_1-\Omega)\exp(-i\Omega t)+S^+(\omega_1+\Omega)\exp(i\Omega t)\right]\exp(i\omega_1 t), \tag{A18}$$

and

$$S_2^+(t)=[S_2^-(t)]^*=\exp[-i(H_S+H_R)t/\hbar]|2,n_1,n_2\rangle\langle 3,n_1,n_2+1|\exp[i(H_S+H_R)t/\hbar]$$
$$=\left[S^+(\omega_2)+S^+\left(\omega_2-\frac{\Omega}{2}\right)\exp\left(-i\frac{\Omega}{2}t\right)+S^+\left(\omega_2+\frac{\Omega}{2}\right)\exp\left(i\frac{\Omega}{2}t\right)\right.$$
$$\left.+S^+(\omega_2-\Omega)\exp(-i\Omega t)+S^+(\omega_2+\Omega)\exp(i\Omega t)\right]\exp(i\omega_2 t), \tag{A19}$$

where $S_i^+[\omega_i\pm(l/2)\Omega]$ are the raising operators for the transitions between the dressed states separated by the frequency $\omega_i\pm(l/2)\Omega$. We now substitute Eqs. (A18) and (A19) into the master equation and make the Markoff approximation.

As the system now evolves on a time scale of the order Γ^{-1}, provided that $\Gamma\tau_c\ll 1$ we can replace $\rho_I(t-\tau)$ with $\rho_I(t)$ in Eq. (A7) as we assume that over the time scale in which the second-order reservoir correlation functions are nonzero, $\rho_I(t-\tau)$ would have hardly changed from $\rho_I(t)$, thus the system can be considered to be Markovian. The time dependence of the system operators is given by Eqs. (A18) and (A19). For $t\gg\tau_c$, we may ignore the first term in Eq. (A7) as a result of Eq. (A8) and then the master equation becomes

$$i\hbar\frac{\partial\rho_I(t)}{\partial t}=-\sum_{ab}\omega_{ab}^+[S_a(t),S_b(t)\rho_I(t)]$$
$$-\sum_{ab}\omega_{ab}^-[\rho_I(t)S_b(t),S_a(t)], \tag{A20}$$

where ω_{ab}^+ and ω_{ab}^- are the reservoir spectral densities

$$\omega_{ab}^+=\frac{1}{\hbar^2}\int_0^t d\tau\,\exp[-i(\omega_b-i\epsilon)\tau]\langle R_a(t)R_b(t-\tau)\rangle, \tag{A21}$$
$$\omega_{ab}^-=\frac{1}{\hbar^2}\int_0^t d\tau\,\exp[-i(\omega_b-i\epsilon)\tau]\langle R_b(t-\tau)R_a(t)\rangle.$$

The reservoir spectral densities contain the mathematically convenient factor $\exp(-\epsilon\tau)$, where we assume that $\epsilon\rightarrow 0_+$ for $t\gg\tau_c$.

In order to evaluate the reservoir spectral densities, of which there would be a total of 400, we begin by making the rotating-wave approximation [where we assume that any terms oscillating at frequencies other than $2\omega_1$, $2\omega_2$, and $(\omega_1+\omega_2)$ are ignored] and take into account the complex conjugate relationships. With this the number of spectral densities may be reduced to 80. Defining

$$S_i=S^+\left(\omega_1+\frac{l}{2}\Omega\right)\exp\left[i\left(\omega_1+\frac{l}{2}\Omega\right)t\right], \tag{A22}$$

where $i\in[1,2,3,4,5]$ and $l\in[0,-1,1,-2,2]$. The sets, of which i and l are members are one to one and onto mappings. Similarly, we label

$$S_j=(S_i^+)^*, \tag{A23}$$

where $j\in[6,7,8,9,10]$ and maps one to one and onto the set of which l is a member. For the second transition frequency ω_2,

$$S_k=S^+\left(\omega_2+\frac{l}{2}\Omega\right)\exp\left[i\left(\omega_2+\frac{l}{2}\Omega\right)t\right],$$
$$S_p=(S_k^+)^*, \tag{A24}$$

where $k\in[11,12,13,14,15]$, $p\in[16,17,18,19,20]$ and the mapping onto l is the same as before.

The corresponding reservoir operators may be written in the form

$$R_{1,2,3,4,5}=-\frac{1}{2}i\hbar\sum_\lambda\Omega_\lambda^{(1)}a_\lambda\exp(-i\omega_\lambda t),$$
$$R_{6,7,8,9,10}=\frac{1}{2}i\hbar\sum_\lambda\Omega_\lambda^{(1)*}a_\lambda^\dagger\exp(i\omega_\lambda t),$$
$$R_{11,12,13,14,15}=-\frac{1}{2}i\hbar\sum_\lambda\Omega_\lambda^{(2)}a_\lambda\exp(-i\omega_\lambda t),$$
$$R_{16,17,18,19,20}=\frac{1}{2}i\hbar\sum_\lambda\Omega_\lambda^{(2)*}a_\lambda^\dagger\exp(i\omega_\lambda t). \tag{A25}$$

It would be time consuming to list all the reservoir spectral densities, therefore we settle for an example, such as

$$\omega_{11}^{+}(t)=\frac{1}{\hbar^2}\int_0^t d\tau\,\exp[-i(\omega_1-i\epsilon)\tau]\langle R_1(t)R_1(t-\tau)\rangle,$$

$$=-\frac{1}{4}\sum_{\lambda\mu}\Omega_\lambda^{(1)}\Omega_\mu^{(1)}\exp[-i(\omega_\mu+\omega_\lambda)t]$$

$$\times\int_0^t d\tau\,\exp[-i(\omega_1-\omega_\mu-i\epsilon)\tau]\langle a_\lambda a_\mu\rangle. \quad \text{(A26)}$$

To evaluate the integral in Eq. (A26) we need to make a statement concerning the type of reservoir interacting with the system. From Eq. (A5) we see that Eq. (A26) becomes

$$\omega_{11}^{+}(t)=-\frac{1}{4}\sum_{\lambda\mu}\Omega_\lambda^{(1)}\Omega_\mu^{(1)}\exp[-i(\omega_\mu+\omega_\lambda)t]$$

$$\times\int_0^t d\tau\,\exp[-i(\omega_1-\omega_\mu-i\epsilon)\tau]M(\omega_\lambda). \quad \text{(A27)}$$

The master equation is evaluated at times $t\gg\tau_c$, and choosing ϵ sufficiently small that $\exp(-\epsilon\tau_c)\ll 1$, the time integral in Eq. (A27) may be evaluated to give

$$\int_0^t d\tau\,\exp[-i(\Delta-i\epsilon)\tau]=\frac{-i}{\Delta-i\epsilon}, \quad \text{(A28)}$$

which in the limit $\epsilon\to 0_+$ reduces to

$$\pi\delta(\Delta)-i\mathrm{P}\frac{1}{\Delta}, \quad \text{(A29)}$$

where P is the principal Cauchy value and in this case $\Delta=\omega_1-\omega_\mu$. The principal Cauchy values lead to frequency shifts of the atomic spectral lines as a result of the coupling to the large number of modes of the reservoir. These shifts are of no concern to us at this time, therefore we will ignore them. Due to the large number of reservoir modes, we may assume that they are closely spaced, and then the summation in Eq. (A27) may be approximated to a good degree by an integral over frequency space, that is,

$$\sum_\lambda\to\int d\omega_\lambda\rho(\omega_\lambda), \quad \text{(A30)}$$

where $\rho(\omega_\lambda)$ is the density of the reservoir modes in frequency space. Thus Eq. (A27) becomes

$$\omega_{11}^{+}(t)=-\frac{1}{4}\int d\omega_\mu\rho(\omega_\mu)\Omega_{2\omega_s-\omega_\mu}^{(1)}\Omega_{\omega_\mu}^{(1)}$$

$$\times\exp(-i2\omega_s t)M(\omega_\mu)\pi\delta(\omega_1-\omega_\mu)$$

$$=-\frac{1}{2}M(\omega_1)\Gamma_{11}\exp(-2i\omega_s t), \quad \text{(A31)}$$

where we have defined

$$\Gamma_{11}=\frac{\pi}{2}[\Omega_{\omega_1}^{(1)}\Omega_{\omega_1}^{(1)}\rho(\omega_1)]=|\Gamma_{11}||D(\omega_1)|^2, \quad \text{(A32)}$$

where $|\Gamma_{11}|=(\pi/2)|\hat{\vec{\mu}}_{21}\cdot\hat{\vec{e}}_\lambda|^2(2\omega_1/\hbar\epsilon_0 V)$. All other reservoir spectral densities follow a similar derivation.

With the reservoir spectral densities, and making the rotating-wave approximation, in which we may ignore all terms oscillating at frequencies $2\omega_i$ and $\omega_i+\omega_j$, we obtain the master equation (3).

[1] A. S. Parkins, in *Modern Nonlinear Optics, Part 2*, edited by M. Evans and S. Kielich (Wiley, New York, 1993), p. 607.
[2] C. W. Gardiner, Phys. Rev. Lett. **56**, 1917 (1986).
[3] Z. Ficek and P. D. Drummond, Phys. Rev. A **43**, 6247 (1991); **43**, 6258 (1991).
[4] V. Buzek, P. L. Knight, and J. K. Kudryavtsev, Phys. Rev. A **44**, 1931 (1991).
[5] J. Gea-Banacloche, Phys. Rev. Lett. **62**, 1603 (1989).
[6] J. Javanainen and P. L. Gould, Phys. Rev. A **41**, 5088 (1990).
[7] N. P. Georgiades, E. S. Polzik, K. Edamatsu, H. J. Kimble, and A. S. Parkins, Phys. Rev. Lett. **75**, 3426 (1995).
[8] H. J. Carmichael, A. S. Lane, and D. F. Walls, Phys. Rev. Lett. **58**, 2539 (1987).
[9] H. J. Carmichael, A. S. Lane, and D. F. Walls, J. Mod. Opt. **34**, 821 (1987).
[10] H. Ritsch and P. Zoller, Opt. Commun. **64**, 523 (1987).
[11] P. Zhou and S. Swain, Quantum Semiclassic. Opt. **8**, 959 (1996).
[12] S. An and M. Sargent III, Phys. Rev. A **39**, 3998 (1989).
[13] B. N. Jagatap, Q. V. Lawande, and S. V. Lawande, Phys. Rev. A **43**, 535 (1991).
[14] S. Smart and S. Swain, Quantum Opt. **4**, 281 (1992).
[15] S. Smart and S. Swain, J. Mod. Opt. **40**, 1939 (1993).
[16] M. R. Ferguson, Z. Ficek, and B. J. Dalton, Phys. Rev. A **54**, 2379 (1996).
[17] H. J. Kimble, O. Carnal, N. P. Georgiades, H. Mubachi, E. S. Polzik, R. J. Thompson, and Q. A. Turchette, in *Atomic Physics 14*, edited by D. J. Wineland, C. E. Wieman, and S. J. Smith (AIP, New York, 1995).
[18] A. S. Parkins and C. W. Gardiner, Phys. Rev. A **40**, 3796 (1989).
[19] J. I. Cirac and L. L. Sanchez-Soto, Phys. Rev. A **44**, 1948 (1991).
[20] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).
[21] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1992).
[22] A. S. Parkins, P. Zoller, and H. J. Carmichael, Phys. Rev. A **48**, 758 (1993).
[23] S. Swain, Phys. Rev. Lett. **73**, 1493 (1994).
[24] M. Lewenstein, T. W. Mossberg, and R. J. Glauber, Phys. Rev. Lett. **59**, 775 (1987).
[25] G. Yeoman and S. M. Barnett, J. Mod. Opt. **43**, 2037 (1996).
[26] M. Bosticky, Z. Ficek, and B. J. Dalton, Phys. Rev. A **53**, 4439 (1996).
[27] J. Opt. Soc. Am. B **4** (10), (1987), special issue on squeezed

states of the electromagnetic field, edited by H. J. Kimble and D. F. Walls.

[28] J. Mod. Opt. **34** (6/7) (1987), special issue on squeezed light, edited by R. Loudon and P. L. Knight.

[29] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 2311 (1977).

[30] M. Born and E. Wolf, *Principles of Optics* (Macmillan, New York, 1964).

[31] B. J. Dalton, E. S. Guerra, and P. L. Knight, Phys. Rev. A **54**, 2292 (1996).

[32] P. D. Drummond and M. D. Reid, Phys. Rev. A **41**, 3930 (1990).

[33] R. M. Whitley and C. R. Stroud, Jr., Phys. Rev. A **14**, 1498 (1976).

[34] W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).