Cavity-field-assisted atomic relaxation and suppression of resonance fluorescence at high intensities

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Within the framework of the cavity QED model we consider a cavity-field-assisted dissipation process via a Raman-type coupling of the atom-cavity system (under off-resonance conditions) to a reservoir. We show that at higher applied field strength the Mollow triplet gets significantly suppressed due to this cavity-field-dependent atomic relaxation.

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I. INTRODUCTION

The interaction of a two-level atom with a damped driven quantized cavity has been studied in detail both theoretically and experimentally during the past few years [1–9]. The significant physical aspect that has emerged from these studies is that the light emitted by the atom confined in a cavity can be reabsorbed by the atom. The effect of this back reaction and the related aspects have been the subject of dynamical suppression of spontaneous emission [6], population inversion in the steady state [8], broadening of the Mollow sidebands in resonance fluorescence [9], vacuum field Rabi splitting [2–5], and several other studies. In general, the effects observed due to the presence of a cavity get enhanced when the coupling between the atom and the field mode exceeds the damping rate. The enhancement is due to the multiple exchange of photons between the cavity mode and the atom before the attainment of the stationary state [1].

However, instead of coherent interaction processes, the cavity may also assist the dissipation processes in a different way. For example, if the atom and the cavity mode are detuned from each other, then (in addition to the linear interaction with the heat baths for the atom and the field) a cavity-assisted Raman-type coupling with the heat bath may be considered and described by an interaction of the form $\sigma_+ a\Gamma_r + \sigma_- a^\dagger \Gamma_r^\dagger$, where σ_+ (σ_-) and $a^\dagger$ (a) are the creation (annihilation) operator for the atom and the field mode, respectively. The reservoir $\Gamma_r(\Gamma_r^\dagger)$ describes the loss of energy from the combined atom-cavity system via the Raman-type process. The purpose of the present paper is to study the cavity-field-assisted dissipation process and the subsequent modification of the resonance fluorescence spectrum [10]. We show that the atomic relaxation may depend on the strength of the cavity field and that, under off-resonance conditions for the cavity and the field, the Mollow triplet gets suppressed at higher intensities due to this cavity-field-dependent damping. Also the quenching of peak intensities is much stronger in the presence of this coupling compared to that under simple off-resonance conditions.

To put the present nonlinear damping scheme in a proper perspective, it is better to recall in this context some of the earlier studies. Since the demonstration by Machida and Yamamoto [11] that the intensity fluctuations in the output of a semiconductor laser can be reduced below the shot-noise limit by feedback, the possibility of reducing intensity noise by introducing intracavity nonlinear damping has been explored by a number of workers [12–14]. Very recently, Wiseman and Milburn [14] have introduced a general class of nonlinear interactions with a reservoir of the type $(a^\dagger)^P a^Q \Gamma_r$ (and its Hermitian conjugate) where P,Q are the integers defined as $P=(N-M)/2$ and $Q=(N+M)/2$. For various values of M and N, different loss mechanisms, such as linear loss ($M=N=1$), two-photon loss ($M=N=2$), Raman loss ($M<N$), etc. have been realized. The dissipation through a cavity-assisted Raman-type interaction [H_5 term in Eq. (1)], with the reservoir as considered in the present paper, is a simple variant among these nonlinear damping mechanisms. This dissipation term is expected to be important when the cavity field is relatively high and is detuned from the atomic resonance, the overall energy being conserved through the reservoir Γ_r.

It is also pertinent to mention at this point the several other treatments of the dissipative processes associated with different kinds of reservoirs for different interactions. For example, phase relaxation due to additional nonlinear energy-conserving interaction with the heat bath was considered by Schubert and Ponath [15]. Tanimura and Kubo [16] have considered the reservoirs with colored noise. Ekert and Knight [17] have treated the reservoirs with a phase-dependent distribution of noise. Squeezed vacuum reservoirs [18] have also been found to be important in modifying the resonance fluorescence spectrum. The effect of reservoir when the system is a nonlinear oscillator [19] has also been treated very recently. In the present paper, however, we consider a typical situation whereby the strength of the cavity field may enhance the atomic relaxation and lead to subsequent modification of the resonance fluorescence spectrum.

II. THE MODEL AND THE CAVITY-FIELD-ASSISTED DAMPING

The dynamical model of a single two-level atom coupled to a cavity can be described by the following model Hamiltonian in the electric dipole and rotating-wave approximations:

$$
\begin{aligned}
H &= \sum_{\mu=0}^{5} H_\mu \, , \\
H_0 &= (\hbar\omega_0/2)\sigma_z + \hbar\omega a^\dagger a \, , \\
H_1 &= \hbar k (a\sigma_+ + a^\dagger\sigma_-) \, , \\
H_2 &= \hbar[a^\dagger E(t) + aE^*(t)] \, , \\
H_3 &= \Gamma\sigma_+ + \Gamma^\dagger\sigma_- \, , \\
H_4 &= \Gamma_c a^\dagger + \Gamma_c^\dagger a \, , \\
H_5 &= \Gamma_r a\sigma_+ + \Gamma_r^\dagger a^\dagger \sigma_- \, .
\end{aligned} \tag{1}
$$

$a^\dagger, a$ are the creation and annihilation operators describing the cavity mode at frequency ω. $\sigma_+, \sigma_-, \sigma_z$ are the pseudospin operators describing the two-level atom with atomic resonance frequency ω_0 such that the atom-cavity detuning is given by $\Delta = \omega_0 - \omega$. We have allowed, in our Hamiltonian, for the situation where the cavity mode is resonantly driven by an external coherent input field $E(t)$ of frequency ω. The atomic reservoir terms Γ and $\Gamma^\dagger$ describe the energy loss from the atom via spontaneous emission and other processes. The loss of energy of the cavity field mode due to dissipation through the mirrors and otherwise is described by the field reservoir terms Γ_c and $\Gamma_c^\dagger$. The H_5 term in the Hamiltonian (1) denotes the cavity-assisted damping via a Raman-type coupling. [For a microscopic realization of this term, we discuss a convenient three-state (an effective two-state) model in the Appendix.] Since the cavity field is detuned from the atomic resonance ($\Delta \neq 0$), this nonlinear coupling term conserves the overall energy through the reservoir term Γ_r.

Projecting out the bosonic reservoir variables in the usual way under Born and Markov approximations [20], one can derive the appropriate master equation for the atom-cavity field system as follows:

$$
\frac{d\rho}{dt} = -\frac{i}{\hbar}[H_0 + H_1 + H_2, \rho] + L_a\rho + L_f\rho + L_{af}\rho \, . \tag{2}
$$

Here

$$
L_f\rho = (1+\bar{n})\frac{\gamma_f}{2}[2a\rho a^\dagger - \rho a^\dagger a - a^\dagger a \rho] + \bar{n}\frac{\gamma_f}{2}[2a^\dagger\rho a - aa^\dagger\rho - \rho a a^\dagger] \, , \tag{3}
$$

$$
L_a\rho = (1+N)\frac{\gamma_a}{2}[2\sigma_-\rho\sigma_+ - \rho\sigma_+\sigma_- - \sigma_+\sigma_-\rho] + N\frac{\gamma_a}{2}[2\sigma_+\rho\sigma_- - \sigma_-\sigma_+\rho - \rho\sigma_-\sigma_+] \tag{4}
$$

denote the damping operator for the cavity mode and the atom, respectively, corresponding to the terms H_3 and H_4 of the Hamiltonian (1). The cavity and the atom are damped at the rate $2\gamma_f$ and $2\gamma_a$, respectively, due to coupling at finite-temperature reservoirs, $\bar{n}$ and N being the mean number of photons in the respective cases. L_{af} is the Liouvillian corresponding to the Raman-type dissipative term H_5, and describes the loss of energy at the rate $2\gamma_r$ from the combined atom-field system. This is given by

$$
L_{af}\rho = (1+N_r)\frac{\gamma_r}{2}[2\sigma_- a^\dagger \rho \sigma_+ a - \sigma_+\sigma_- a a^\dagger \rho - \rho\sigma_+\sigma_- a a^\dagger] + N_r\frac{\gamma_r}{2}[2\sigma_+ a \rho \sigma_- a^\dagger - \sigma_-\sigma_+ a^\dagger a \rho - \rho\sigma_-\sigma_+ a^\dagger a] \, , \tag{5}
$$

where N_r is the mean number of thermal quanta and is given by $N_r = 1/(e^{\hbar\Delta/kT} - 1)$.

The nature of dissipation as expressed through the Liouvillian (5) can be made more explicit if we write down the mean-field dissipative dynamical equations involving the polarization and the population inversion variables for the atom as follows:

$$
\begin{aligned}
\langle \dot\sigma_+ \rangle &= -(\gamma_a/2)\langle\sigma_+\rangle - (\gamma_r/2)(1+\langle a^\dagger a\rangle)\langle\sigma_+\rangle \, , \\
\langle \dot\sigma_- \rangle &= -(\gamma_a/2)\langle\sigma_-\rangle - (\gamma_r/2)(1+\langle a^\dagger a\rangle)\langle\sigma_-\rangle \, , \\
\langle \dot\sigma_z \rangle &= -\gamma_a(1+\langle\sigma_z\rangle) - \gamma_r(1+\langle a^\dagger a\rangle)(1+\langle\sigma_z\rangle) \, .
\end{aligned}
$$

It is immediately apparent that the relaxation of the atomic system is dependent on the strength of the cavity field. What follows next is to show that this cavity-assisted dissipation leads to an interesting modification of the resonance fluorescence spectrum.

III. THE MODIFICATION OF THE RESONANCE FLUORESCENCE SPECTRUM

To study the master equation (2), we adopt the treatment of operator ordering followed in Refs. [21–25]. To summarize, we choose an ordering for atomic and field operators by defining the characteristic function

$$
X(\xi) = \mathrm{Tr}(O\rho) \, , \tag{6}
$$

where

$$
O(\xi) = e^{i\xi^* a^\dagger} e^{i\xi a} e^{i\xi_+\sigma_+} e^{i\xi_z\sigma_z} e^{i\xi_-\sigma_-} \, , \tag{7}
$$

and

$$
\boldsymbol{\xi} = [\xi^*, \xi, \xi_+, \xi_z, \xi_-]^t \, .
$$

We then define the generalized positive P-representation [15] by

$$
X(\boldsymbol{\xi}) = \int_{-\infty}^{+\infty} d^2\boldsymbol{\alpha} \exp(i\boldsymbol{\xi}\cdot\boldsymbol{\alpha}) P(\boldsymbol{\alpha}) \, , \tag{8}
$$

and $\boldsymbol{\alpha}$ is the column vector composed of the c-numbers α_i; $\boldsymbol{\alpha} = [\alpha^*, \alpha, \alpha_+, \alpha_z, \alpha_-]$, and establishes a correspondence between the c-numbers and the operators as follows:

$$
\begin{aligned}
&a \leftrightarrow \alpha \, , \quad \sigma_+ \leftrightarrow \alpha_+ \, , \\
&a^\dagger \leftrightarrow \alpha^* \, , \quad \sigma_- \leftrightarrow \alpha_- \, , \\
&\sigma_z \leftrightarrow \alpha_z \, .
\end{aligned}
$$

Using the standard operator-disentanglement technique and the definition of $P(\boldsymbol{\alpha})$, a partial differential equation is deduced for $P(\boldsymbol{\alpha})$. This has derivatives of all orders present for α_z as exponentials of derivatives, but can be approximated as a Fokker-Planck equation for $P(\alpha)$ by keeping up to second derivatives. Having obtained the Fokker-Planck equation, which is the c-number equivalent of the master equation, one can immediately write down the Langevin equations as follows:

$$
\begin{aligned}
\dot{\alpha} &= \left[-i\omega\alpha - \gamma_f\alpha - ik\alpha_- + \frac{\gamma_r}{2}(1+\alpha_z)\alpha - iE(t) \right] + G_\alpha(t) , \\
\dot{\alpha}^* &= \left[i\omega\alpha^* - \gamma_f\alpha^* + ik\alpha_+ + \frac{\gamma_r}{2}(1+\alpha_z)\alpha^* + iE(t) \right] + G_{\alpha^*}(t) , \\
\dot{\alpha}_+ &= [i\omega_0\alpha_+ - ik\alpha^*\alpha_z - (\gamma_a/2)\alpha_+ - (\gamma_r/2)(1+\alpha^*\alpha)\alpha_+] + G_{\alpha_+}(t) , \\
\dot{\alpha}_- &= [-i\omega_0\alpha_- + ik\alpha\alpha_z - (\gamma_a/2)\alpha_- - (\gamma_r/2)(1+\alpha^*\alpha)\alpha_-] + G_{\alpha_-}(t) , \\
\dot{\alpha}_z &= [-2ik(\alpha\alpha_+ - \alpha^*\alpha_-) - \gamma_a(1+\alpha_z) - \gamma_r(1+\alpha^*\alpha)(1+\alpha_z)] + G_{\alpha_z}(t) .
\end{aligned}
\tag{9}
$$

Here the G_{α_i}'s are the independent Langevin forces with zero reservoir averages as follows: $\langle G_{\alpha_i} \rangle_R = 0$. The nonzero noise correlation of the random forces are given by

$$\langle G_{\alpha_+}(t) G_{\alpha_+}(t') \rangle_R = 2D_{\alpha_i\alpha_j}\delta(t-t') ,$$

where

$$D_{\alpha_+\alpha_+} = -ik\alpha^*\alpha_+ , \quad D_{\alpha_-\alpha_-} = ik\alpha\alpha_- , \quad D_{\alpha^*\alpha} = \gamma_r(1+\alpha_z),$$

$$D_{\alpha_z\alpha_z} = -2ik(\alpha\alpha_+ - \alpha^*\alpha_-) + \gamma_a(1+\alpha_z) + \gamma_r(1+\alpha^*\alpha)(1+\alpha_z) ,$$

$$D_{\alpha\alpha_z} = -\gamma_r(1+\alpha_z)\alpha, \quad D_{\alpha^*\alpha_z} = -\gamma_r(1+\alpha_z)\alpha^* ,$$

$$D_{\alpha\alpha_-} = -\frac{\gamma_r}{2}\alpha_-\alpha, \quad D_{\alpha^*\alpha_+} = -\frac{\gamma_r}{2}\alpha_+\alpha^* .$$

The terms involving γ_r in Eqs. (9) are the essential content of the present work. It is evident that the relaxation of the atom is manifestly dependent on the intensity of the cavity field, since both the polarization and population inversion variables decay at rates that are functions of the cavity field.

To eliminate the fast-time dependence, we now invoke the slowly varying envelope approximation:

$$
\begin{aligned}
&\alpha = \beta e^{-i\omega t} , \quad \alpha^* = \beta^* e^{i\omega t} , \quad \alpha_- = \beta_- e^{-i\omega_0 t} , \\
&\alpha_+ = \beta_+ e^{i\omega_0 t} , \quad \alpha_z = \beta_z .
\end{aligned}
\tag{10}
$$

We also assume the driving field $E(t) = E_0 e^{-i\omega t}$ to be at exact resonance with the cavity field mode. The c-number stochastic differential equations for the slowly varying amplitudes β's are obtained by substituting Eqs. (10) in (9). The resulting equations are

$$
\begin{aligned}
\dot{\beta} &= \left[-\gamma_f\beta - ik\beta_- - iE_0 + \frac{\gamma_r}{4}(1+\beta_z)\beta \right] + G_\beta(t) , \\
\dot{\beta}^* &= \left[-\gamma_f\beta^* + ik\beta_+ + iE_0 + \frac{\gamma_r}{4}(1+\beta_z)\beta^* \right] + G_{\beta^*}(t) , \\
\dot{\beta}_+ &= \left[i\Delta\beta_+ - ik\beta^*\beta_z - (\gamma_a/2)\beta_+ - \frac{\gamma_r}{2}(1+\beta^*\beta)\beta_+ \right] + G_{\beta_+}(t) , \\
\dot{\beta}_- &= \left[-i\Delta\beta_- + ik\beta\beta_z - (\gamma_a/2)\beta_- - \frac{\gamma_r}{2}(1+\beta^*\beta)\beta_- \right] + G_{\beta_-}(t) , \\
\dot{\beta}_z &= [-2ik(\beta\beta_+ - \beta^*\beta_-) - \gamma_a(1+\beta_z) - \gamma_r(1+\beta^*\beta)(1+\beta_z)] + G_{\beta_z}(t) ,
\end{aligned}
\tag{11}
$$

where $G_{\alpha_i}(t) = G_{\beta_i}(t)e^{i\omega_i t}$ and $\Delta = \omega_0 - \omega$.

The mean-field solutions are obtained from Eqs. (11) by neglecting the quantum fluctuations and making $\dot{\beta}_i = 0$ $(i = 1,5)$. The result is a set of five algebraic equations whose solutions give the average of the atomic and field quantities in the steady state. Particularly, for the steady-state cavity field β^3, we have the following equation of state:

$$
\begin{aligned}
&\{\tfrac{1}{4}[\gamma_a + \gamma_r(1+|\beta^e|^2)]^2 + \Delta^2 + 2k^2|\beta^e|^2\}[(-\gamma_r + 4\gamma_f)\beta^e + 4iE_0] + \{\tfrac{1}{4}[\gamma_a + \gamma_r(1+|\beta^e|^2)]^2 + \Delta^2\} \\
&\qquad \times \left[\gamma_r + \frac{4k^2}{i\Delta + \frac{\gamma_a}{2} + \frac{\gamma_r}{2}(1+|\beta^e|^2)} \right] \beta^e = 0 .
\end{aligned}
\tag{12}
$$

Numerical solution of the equation yields the values of the slowly varying amplitudes β^{e}'s in the steady state.

Having obtained the mean-field solution for our problem, we now go to the next order in perturbation to find the fluctuation around the mean. Thus we let $\beta_i=\beta_i^e+\delta\beta_i$, where $\delta\beta_i$ are the fluctuations around the mean values β_i^e. The result is the Ito-stochastic differential equation of the form [20]

$$d\delta\boldsymbol{\beta}(t)=-\underline{A}\,\delta\boldsymbol{\beta}(t)dt+\underline{B}\,d\mathbf{W}(t)\ , \tag{13}$$

where $\underline{A}$ and $\underline{B}$ are the drift and diffusion matrices, respectively, expressed in terms of the steady-state values. Here $d\mathbf{W}$ defines the vectorial Wiener processes, and $\delta\boldsymbol{\beta}$ represents the fluctuation vector. The Ito-stochastic differential equation (13) describes a multivariate Ornstein-Uhlenbeck process that is analytically solvable. For more details, we refer to Ref. [26].

Next we calculate the power spectrum [26] for the emitted light in the usual way:

$$\begin{aligned}S_{\delta\beta_+\delta\beta_-}(\nu-\omega_0)&=\int_{-\infty}^{+\infty}\exp[-i(\nu-\omega_0)t]\\&\quad\times\langle\delta\beta_+(t)\delta\beta_-(0)\rangle dt\\&=\{[\underline{A}+i(\nu-\omega_0)\underline{I}]^{-1}\underline{BB}^t\\&\quad\times\underline{A}^t-i(\nu-\omega_0)\underline{I}]^{-1}\}_{\delta\beta_+\delta\beta_-}\ .\end{aligned}$$

Explicit calculation yields the spectra that are given in Figs. 1–4. In order to compare with the standard result with nonzero detuning but for $\gamma_r=0$, we first reproduce the Mollow triplet in Fig. 1. As the external field strength increases, the spectrum splits into a triplet, and the incoherent peaks get more separated from the coherent one, accompanied by an enhancement of peak heights. In Figs. 2 and 3, we plot the spectra in the presence of cavity-assisted damping, i.e., $\gamma_r\neq0$ for different external field intensities. It is evident that, with the increase of field intensities, peak heights first increase up to a critical value and then start depleting. This suppression of peak intensity at higher applied field strength is a direct result of the cavity-field-assisted damping. This is accompanied by the usual broadening of the spectra. Lastly, in Fig. 4, we show a schematic variation of spec-

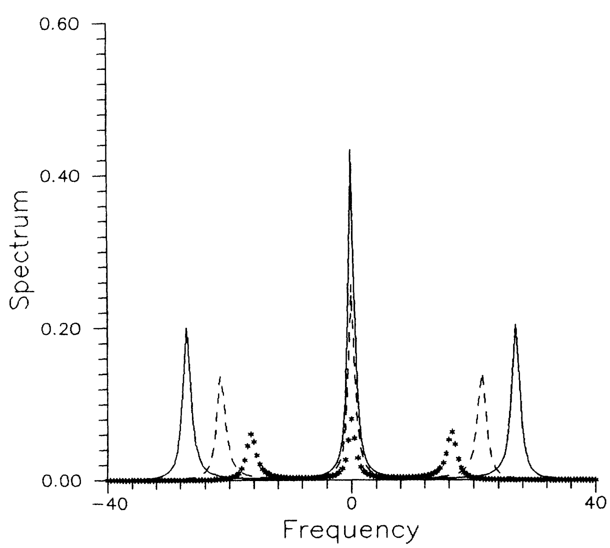

FIG. 1. Power spectrum $S(\nu-\omega_0)\gamma_a$ is plotted against the frequency $(\nu-\omega_0)/\gamma_a$, with $\gamma_f/\gamma_a=8.0$, $\gamma_r/\gamma_a=0.0$, $k/\gamma_a=5.0$, and $\Delta/\gamma_a=10.0$ for three different values of field strengths $E_0/\gamma_a=10,15,20$, on the curves of asterisks, dashed lines, and solid lines, respectively. (Both scales are arbitrary.)

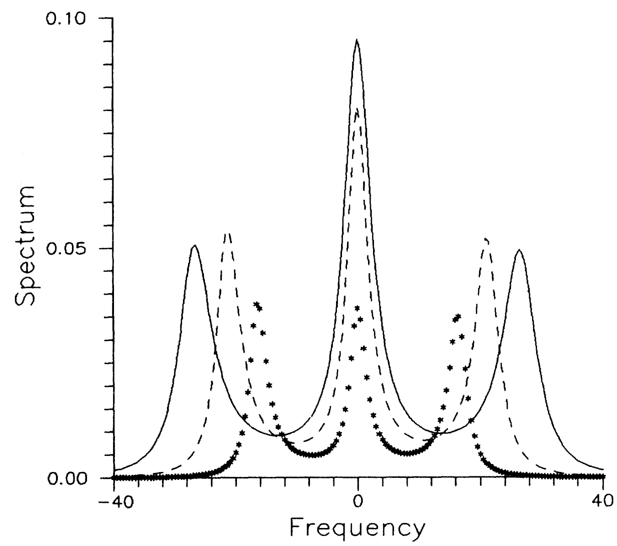

FIG. 2. Power spectrum $S(\nu-\omega_0)\gamma_a$ is plotted against the frequency $(\nu-\omega_0)/\gamma_a$, with $\gamma_f/\gamma_a=8.0$, $k/\gamma_a=5.0$, and $\Delta/\gamma_a=10.0$, but $\gamma_r/\gamma_a=0.5$, for three different values of field strengths $E_0/\gamma_a=10,15,20$, on the curves of asterisks, dashed lines, and solids lines, respectively. (Both scales are arbitrary.)

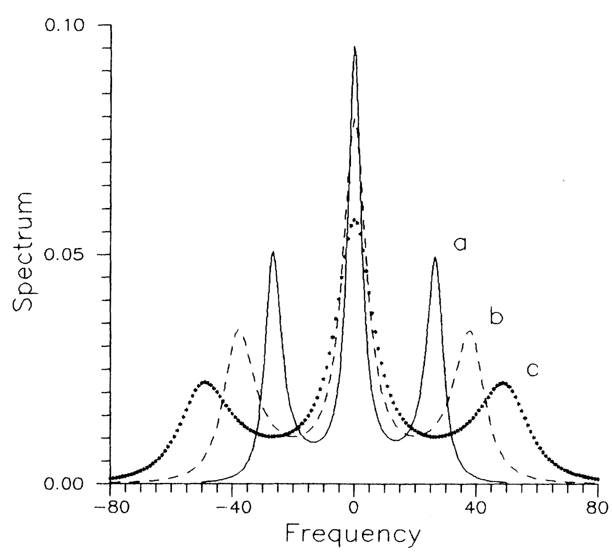

FIG. 3. Power spectrum $S(\nu-\omega_0)\gamma_a$ is plotted against the frequency $(\nu-\omega_0)/\gamma_a$, with $\gamma_f/\gamma_a=8.0$, $k/\gamma_a=5.0$, and $\Delta/\gamma_a=10.0$, $\gamma_r/\gamma_a=0.5$, for three different values of field strengths $E_0/\gamma_a=20$ (a), 30 (b), 40 (c). (Both scales are arbitrary.)

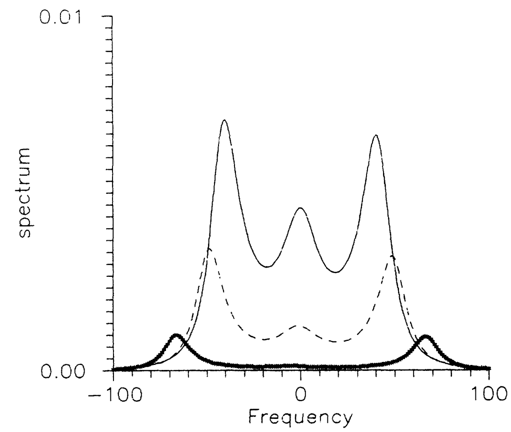

FIG. 4. Power spectrum $S(\nu-\omega_0)\gamma_a$ vs frequency $(\nu-\omega_0)/\gamma_a$ is plotted, with $\gamma_f/\gamma_a=8.0$, $k/\gamma_a=5.0$, and $E_0/\gamma_1=10.0$, $\gamma_r/\gamma_a=0.5$, for three different values of atom-cavity detuning $\Delta/\gamma_a=30,40,60$, on the curves of solid lines, dashed lines, and asterisks, respectively. (Both scales are arbitrary.)

tra as a function of the detuning of the atom and the cavity field. It is evident that, at large detuning, the triplet practically reduces to a doublet, with almost complete suppression of the coherent peak.

IV. CONCLUSION

It is well known that, in cavity QED problems, the cavity enhances significantly the effects due to the coherent interaction processes because of the increased multiple exchange of photons between the cavity and the atom. We have shown, however, that under off-resonance conditions, the cavity may assist the dissipation processes via a Raman-type nonlinear coupling to a reservoir. This results in damping of the atomic system, which is dependent on the strength of the cavity field. We have also shown that this intensity-dependent damping leads to an interesting modification of the resonance fluorescence spectrum, which is reflected in the suppression of the Mollow triplet at the higher field strength with nonzero detuning. It may also be noted that the intensity-dependent relaxation process was also found to be important in the recent past for two-level atoms interacting with a superintense field [27]. We hope that the cavity-assisted relaxation as considered in the present paper may also play a fundamental role in similar issues in other cavity QED problems.

ACKNOWLEDGMENT

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APPENDIX

In this appendix, we show how the H_5 term in Eq. (1) can be realized in terms of a microscopic model interaction scheme.

We consider a model three-state system [28] (similar to that used in the Raman-coupled model) where the excited state $|2\rangle$ gets deexcited, by a detuned cavity mode with frequency ω and one or more reservoir modes ω_j, to state $|1\rangle$ via the state $|3\rangle$, in a manner shown in Fig. 5. The relevant Hamiltonian is given by

$$\bar{H}=\bar{H}_0+\bar{H}_I\ ,$$
$$\bar{H}_0=E_1\sigma_{11}+E_2\sigma_{22}+E_3\sigma_{33}+\hbar\omega a^\dagger a+\sum_j \hbar\omega_j\Gamma_j^\dagger\Gamma_j\ , \tag{A1}$$
$$\bar{H}_I=\hbar g_{23}(a\sigma_{23}+a^\dagger\sigma_{32})+\hbar\sum_j g_{13}^j(\Gamma_j^\dagger\sigma_{13}+\Gamma_j\sigma_{31})\ ,$$

where σ_{ii} $(i=1,2,3)$ are the level population operators, Γ_j $(\Gamma_j^\dagger)$ are the usual harmonic-oscillator heat bath operators, and g_{ij}'s are the usual coupling constants.

The Heisenberg equations for transition operators σ_{13} and σ_{23} are

$$\dot{\sigma}_{13}=-i(\omega_0-\omega')\sigma_{13}+ig_{23}a^\dagger\sigma_{12} -i\sum_j g_{13}^j(\sigma_{33}-\sigma_{11})\Gamma_j\ , \tag{A2}$$

$$\dot{\sigma}_{23}=-i\omega'\sigma_{23}+i\sum_j g_{13}^j\Gamma_j\sigma_{21}-ig_{23}a^\dagger(\sigma_{33}-\sigma_{22})\ , \tag{A3}$$

where $\hbar\omega'=E_2-E_3$. For simplicity, we may take g_{ij}'s to be real.

Now introducing the slow variables with tildes,

$$\sigma_{13}=\tilde{\sigma}_{13}e^{-i\omega_j t},\ \sigma_{23}=\tilde{\sigma}_{23}e^{-i\omega t},\ a^\dagger=\tilde{a}^\dagger e^{-i\omega t}\ ,$$
$$\Gamma_j=\tilde{\Gamma}_j e^{-i\omega_j t},\ \sigma_{ii}=\tilde{\sigma}_{ii}\ \text{ for } i=1,2,3\ , \tag{A4}$$

and also introducing

$$\sigma_{21}=\sigma_{23}\sigma_{31}=\tilde{\sigma}_{23}\tilde{\sigma}_{31}e^{-i(\omega-\omega_j)t}=\tilde{\sigma}_{21}e^{-i(\omega-\omega_j)t}\ , \tag{A5}$$

and

$$\omega_0-\omega'=\delta \text{ and } \omega'-\omega=\delta'\ ,$$

we obtain

$$\dot{\tilde{\sigma}}_{13}=-i(\delta-\omega_j)\tilde{\sigma}_{13}+ig_{23}\tilde{a}^\dagger\tilde{\sigma}_{12} -i\sum_j g_{13}^j(\tilde{\sigma}_{33}-\tilde{\sigma}_{11})\tilde{\Gamma}_j\ , \tag{A6}$$

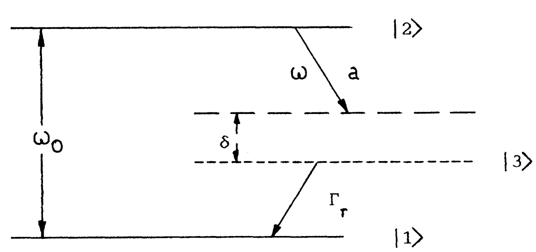

FIG. 5. Energy-level diagram of three-level system. The cavity mode with frequency ω is interacting with the dipole moment of transition $2\rightarrow3$. The quasistationary state $|3\rangle$ is relaxed through reservoir modes.

$$\dot{\tilde{\sigma}}_{23}=-i\delta'\tilde{\sigma}_{23}+i\sum_j g_{13}^j\tilde{\Gamma}_j\tilde{\sigma}_{21}-ig_{23}\tilde{a}^\dagger(\tilde{\sigma}_{33}-\tilde{\sigma}_{22}) . \quad (A7)$$

Now, if the state $|3\rangle$ is quasistationary, one can find $\tilde{\sigma}_{13},\tilde{\sigma}_{23}$ adiabatically as

$$\sigma_{13}=\frac{1}{\delta-\omega_j}\left[g_{23}a^\dagger\sigma_{12}+\sum_j g_{13}^j(\sigma_{33}-\sigma_{11})\Gamma_j\right] , \quad (A8)$$

$$\sigma_{23}=\frac{1}{\delta}'\left[\sum_j g_{13}^j\Gamma_j\sigma_{21}-g_{23}a^\dagger(\sigma_{33}-\sigma_{22})\right] . \quad (A9)$$

Now the terms diagonal in the atomic variables will lead to Stark shifts, which we shall assume to be small. By dropping these terms, we have

$$\sigma_{13}\approx(g_{23}a^\dagger\sigma_{12})/(\delta-\omega_j) ,$$
$$\sigma_{23}\approx\left[\sum_j g_{13}^j\Gamma_j\sigma_{21}\right]\Big/\delta' . \quad (A10)$$

Upon inserting these terms in $\bar{H}_I$ in Eq. (A1), we have the effective interaction

$$H_I^{\text{eff}}=\hbar\sum_j g_{23}g_{13}^j\frac{\delta+\delta'-\omega_j}{(\delta-\omega_j)\delta'}(a\Gamma_j\sigma_++\sigma_-a^\dagger\Gamma_j^\dagger) , \quad (A11)$$

where we have defined $\sigma_{21}=\sigma_+$ and $\sigma_{12}=\sigma_-$ for the effective two-level atom.

If one makes the choice $\delta=\delta'$, then we may write $\delta=\Delta/2$, where $\Delta=\omega_0-\omega$, the atom-field detuning. Putting

$$\lambda_j=g_{23}g_{13}^j\frac{\delta+\delta'-\omega_j}{(\delta-\omega_j)\delta'} , \quad \Gamma_r=\hbar\sum_j\lambda_j\Gamma_j ;$$

we obtain

$$H_I^{\text{eff}}=\hbar\sum_j\lambda_j(a\Gamma_j\sigma_++\sigma_-a^\dagger\Gamma_j^\dagger)=\Gamma_ra\sigma_++\Gamma_r^\dagger a^\dagger\sigma_-\equiv H_5$$

$\bar{H}_0$ includes $E_{33}\sigma_{33}$, a constant shift term that does not affect the dynamics and need not be considered further. This H_5 term, together with $H_{1,2,3,4}$ for linear terms and the H_0 term [of Eq. (1)] for the effective two-level system and single-mode field, comprise the full Hamiltonian in Eq. (1).

It is thus immediately apparent that the order of magnitude of the dissipative coupling constant λ_j in the H_5 term is $\sim O(g^2/\delta)$, while for the linear dissipative term (H_3/H_4) it is $\sim O(g)$. Therefore the cavity field strength should be of the order of $\sim\delta/g$ for the nonlinear dissipative term H_5 to be important. It should be emphasized that nonzero detuning δ (or Δ) is an important quantity in this context. Specializing the model for Rydberg atoms, where the difference frequency $\Delta\sim10^{15}n^{-3}$ sec^{-1} and the coupling constant $g\sim(2\pi\hbar\omega/V)^{1/2}d/\hbar\sim10^4n^{1/2}V^{-1/2}$ sec^{-1}, where n is the principal quantum number and V is the cavity volume, one can obtain a reasonable range of cavity field strength for the experimental realization of the scheme.

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[1] See, for example, S. Haroche and J. M. Raimond, in *Advances in Atomic and Molecular Physics,* edited by D. Bates and B. Bederson (Academic, New York, 1985), Vol. 20, p. 347.
[2] J. J. Sanchez-Mondragon, N. B. Narozhny, and J. H. Eberly, Phys. Rev. Lett. **51**, 550 (1983).
[3] G. S. Agarwal, Phys. Rev. Lett. **51**, 1732 (1984).
[4] G. S. Agarwal, R. K. Bullough, and G. P. Hildred, Opt. Commun. **59**, 23 (1986).
[5] D. Meschede, H. Walther, and G. Miller, Phys. Rev. Lett. **54**, 551 (1985); G. Rempe, H. Walther, and N. Klein, Phys. Rev. Lett. **58**, 353 (1987).
[6] Y. Zhu, A. Lezana, T. Mossberg, and M. Lewenstein, Phys. Rev. Lett. **61**, 1946 (1988).
[7] M. J. Raizen, R. J. Thompson, R. J. Brecha, H. J. Kimble, and H. J. Carmichael, Phys. Rev. Lett. **63**, 240 (1989).
[8] C. M. Savage, Phys. Rev. Lett. **60**, 1829 (1988).
[9] C. M. Savage, Phys. Rev. Lett. **63**, 1376 (1989).
[10] B. R. Mollow, Phys. Rev. A **5**, 1522 (1972); **8**, 1949 (1973); C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 365 (1977).
[11] S. Machida and Y. Yamamoto, Opt. Commun. **57**, 290 (1988).
[12] H. Ritsch, Quantum Opt. **2**, 189 (1990).
[13] D. F. Walls, M. J. Collet, and A. S. Lane, Phys. Rev. A **42**, 4366 (1990).
[14] H. M. Wiseman and G. J. Milburn, Phys. Rev. A **44**, 7815 (1991).
[15] M. Schubert and H. E. Ponath, Ann. Phys. Paris **34**, 456 (1977).
[16] Y. Tanimura and R. Kubo, J. Phys. Soc. Jpn. **58**, 101 (1989).
[17] A. K. Ekert and P. L. Knight, Phys. Rev. A **42**, 487 (1990).
[18] C. W. Gardiner and M. J. Collet, Phys. Rev. A **31**, 3761 (1985).
[19] G. Gangopadhyay and D. S. Ray, J. Chem. Phys. (to be published).
[20] See, for example, W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).
[21] P. D. Drummond and D. F. Walls, Phys. Rev. A **23**, 2563 (1981).
[22] M. D. Reid and D. F. Walls, Phys. Rev. A **28**, 332 (1983).
[23] F. A. M. de Oliveira and P. L. Knight, Phys. Rev. A **39**, 3417 (1989).
[24] G. Gangopadhyay and D. S. Ray, Phys. Rev. A **43**, 6424 (1991).
[25] G. Gangopadhyay and D. S. Ray, Phys. Rev. A **45**, 1843 (1992).
[26] C. W. Gardiner, *Handbook of Stochastic Methods* (Springer-Verlag, Berlin, 1983).
[27] G. Gangopadhyay and D. S. Ray, Phys. Rev. A **44**, 2206 (1991).
[28] C. C. Gerry and J. H. Eberly, Phys. Rev. A **42**, 6805 (1990); S. J. D. Phoenix and P. L. Knight, J. Opt. Soc. Am. B **7**, 116 (1990); see also Ref. [25].