PHYSICAL REVIEW A VOLUME 45, NUMBER 3 1 FEBRUARY 1992

Spectral modification of the Stokes line of a Raman-coupled three-level system in a cavity

Gautam Gangopadhyay and Deb Shankar Ray

Department of Physical Chemistry, Indian Association for the Cultivation of Science, Jadavpur, Calcutta 700032, India

(Received 11 June 1991)

We have investigated the spectral modification of the Raman Stokes line in the cavity version of Raman scattering where a three-level system in the Λ configuration interacts with an externally driven pump mode and a Stokes mode in a cavity. Anti-Stokes modes are eliminated by the cavity-resonance condition. At very low intensities the spectrum consists of a single-peak structure (Raman Stokes) that splits into a doublet when the external pump field is sufficiently intense so that the effective Rabi frequency exceeds the rate of spontaneous emission. At very high intensity, however, a triplet structure appears where the strong sidebands are symmetrically separated from the central weak band.

PACS number(s): 42.50.Hz, 42.60.Da, 32.80.Bx

I. INTRODUCTION

The spectral modification of radiation characteristics of quantum-optical systems when confined in a cavity has become one of the major issues of cavity QED today [1–7]. The model that lies at the heart of these developments is the Jaynes-Cummings model [2,3,7]. Recently a two-mode variant of this model describing the dynamics of a Raman-coupled [8] system has been proposed [9–12]. The model describes a three-level atomic system in the Λ configuration where the excited state is considered to be far off resonance and is adiabatically eliminated [11]. The interaction term of this effective two-level system thus consists of products of the creation operator of the pump mode and the annihilation operator of the Stokes mode. The model is exactly solvable and admits of a number of nonclassical effects studied recently by Gerry and Eberly [11] and Gou [12].

The purpose of the present paper is to investigate the spectral modification of the Raman Stokes line in this cavity version of Raman scattering where the pump and the Stokes cavity modes are interacting with this effective two-level system in a three-level Λ configuration. To make the problem more realistic we have introduced the loss due to radiation damping for the material system and the cavity losses for the two modes and also in addition consider the pump mode to be externally driven. At very low intensities the spectrum consists of a single-peak structure (Raman Stokes) which splits into a doublet when the external pump field is sufficiently intense so that the effective Rabi frequency exceeds the rate of spontaneous emission. The doublet structure of the Raman Stokes line may be interpreted as the usual manifestation of vacuum field Rabi splitting in a cavity [2]. At very high intensity, however, a triplet structure appears where the two strong sidebands are symmetrically separated from the central weak band.

One pertinent point regarding the present model needs attention. It is well known from the earlier works of Mollow and others [8] that the explanation of the spectral modification of Raman scattering in an intense field is based on a three-level model system. The present model proposed by Gerry and Eberly is a variant of the same in the sense that with an appropriate adiabatic elimination of the third level due to large detuning it behaves like the effective two-level system which is describable in terms of an exactly solvable two-mode Jaynes-Cummings-type Hamiltonian. One object here is to explore the spectral modification due to the effect of the cavity based on the latter model modified by introducing the driving and relaxation terms for the cavity modes and atomic system.

II. THE MODIFICATION OF THE RAMAN STOKES SCATTERING IN THE CAVITY

In Fig. 1 we describe the three-level atomic system in the Λ configuration as considered in Ref. [11]. The Hamiltonian describing this system is given by $H = H_0 + H_1$, where

$$H_0 = E_1\sigma_{11} + E_2\sigma_{22} + E_3\sigma_{33} + \hbar\omega_1 a_1^\dagger a_1 + \hbar\omega_2 a_2^\dagger a_2 + \hbar[E(t)a_1^\dagger + E^*(t)a_1]$$

and

$$H_I = \hbar g_{12}(a_1\sigma_{21} + a_1^\dagger\sigma_{12}) + \hbar g_{23}(a_2\sigma_{23} + a_2^\dagger\sigma_{32}) .$$

Here a_1 and a_2 are the operators for the pump and the Stokes cavity mode, respectively, and σ_{ii} and σ_{ij} are the level occupation number operator and transition operator for the levels i and j, respectively. $E(t)$ is the external field which drives the cavity pump mode. The cavity is tuned in such a way that $E_3 - E_1 = \hbar\omega_1 - \hbar\omega_2$ and there is one detuning parameter Δ, defined by $\hbar\Delta = E_2 - E_1 - \hbar\omega_1 = E_2 - E_3 - \hbar\omega_2$. Assuming $\hbar\Delta \gg E_3 - E_1$ one can adiabatically eliminate the second level as described in Ref. [11]. The resulting effective interaction [8] Hamiltonian is given by

$$H_{I,\text{eff}} = \hbar\lambda(\sigma_+ a_1 a_2^\dagger + \sigma_- a_1^\dagger a_2),$$

where $\sigma_+ = \sigma_{31}$, $\sigma_- = \sigma_{13}$, and λ is the effective coupling constant such that $\lambda = 2g_{12}g_{23}/\Delta$. Since the second level

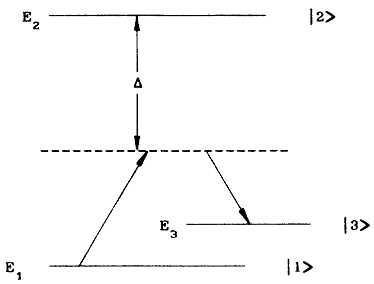

FIG. 1. Energy levels of the three-level atom in the Λ configuration. The detuning Δ is large compared to E_3-E_1.

is far off resonance we also assume that $\sigma_{22}=0$. This implies that $\sigma_{11}+\sigma_{33}=1$. We then obtain the total effective Hamiltonian as follows:

$$H=\hbar\omega_0\sigma_0/2+\hbar\omega_1 a_1^\dagger a_1+\hbar\omega_2 a_2^\dagger a_2 +\hbar[E(t)a_1^\dagger+E^*(t)a_1]+(E_1+E_3)I +\hbar\lambda(\sigma_+ a_1 a_2^\dagger+\sigma_- a_1^\dagger a_2) .$$

In a standard cavity QED problem one is essentially concerned with a number of competing processes, e.g., the fundamental coherent interaction between the material system with cavity field modes, the spontaneous decay of the system, and the loss of photons from the cavity field modes. The thermally induced processes are negligible in the case of optical transition at low temperatures. In the present problem of Raman scattering in a cavity we need be concerned with all these processes within the quantum-statistical scheme for an analysis of spectral characteristics of scattered radiation. For simplicity we omit all other relaxation processes and neglect the wave-mixing effects from the present analysis.

We now introduce the loss terms by coupling this atom-field system with the bosonic heat baths. The master equation [13] for the reduced density operator (ρ) can be obtained in the rotating-wave, Born-Markov approximations as follows:

$$\frac{\partial\rho}{\partial t}=(i\hbar)^{-1}[H_{AF}+H_{I,\mathrm{eff}},\rho]+L_f(\rho)+L_a(\rho) , \quad (1)$$

where L_f and L_a are the Liouville operators representing the decay of the cavity field modes and the atom, respectively. These are given by

$$L_f(\rho)=\sum_{i=1}^{2}\gamma_{f_i}(2a_i\rho a_i^\dagger-\rho a_i^\dagger a_i-a_i^\dagger a_i\rho), \quad (2)$$

where for simplicity the decay rates of two fields γ_{f_1} and γ_{f_2} are assumed to be equal ($\gamma_{f_1}=\gamma_{f_2}=\gamma_f$) and

$$L_a(\rho)=(\gamma_a/2)(2\sigma_-\rho\sigma_+-\rho\sigma_+\sigma_--\sigma_+\sigma_-\rho), \quad (3)$$

where γ_a is the atomic decay rate.

Next we choose an ordering for the atomic and field operators [14,15], by defining the normal order characteristic function as follows:

$$O(\xi)=e^{i\xi_1^* a_1^\dagger}e^{i\xi_1 a_1}e^{i\xi_2^* a_2^\dagger}e^{i\xi_2 a_2}e^{i\xi_+\sigma_+}e^{i\xi_0\sigma_0}e^{i\xi_-\sigma_-}$$

with

$$X(\xi)=\mathrm{Tr}(O\rho) \quad \text{where } \xi=(\xi_1^*,\xi_1,\xi_2^*,\xi_2,\xi_-,\xi_+,\xi_0)^t .$$

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

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

and $\boldsymbol{\alpha}$ is the column vector composed by the c numbers α_i; $\boldsymbol{\alpha}=(\alpha_1^*,\alpha_1,\alpha_2^*,\alpha_2,\alpha_-,\alpha_+,\alpha_0)$, and establish a correspondence between the c numbers and the operators as follows:

$$a_1\leftrightarrow\alpha_1,\quad \sigma_+\leftrightarrow\alpha_+,\quad a_1^\dagger\leftrightarrow\alpha_1^*,\quad \sigma_-\leftrightarrow\alpha_- ,$$
$$a_2\leftrightarrow\alpha_2,\quad \sigma_0\leftrightarrow\alpha_0,\quad a_2^\dagger\leftrightarrow\alpha_2^* .$$

Using the standard operator disentanglement technique and definition of $P(\boldsymbol{\alpha})$ a partial differential equation is deduced for $P(\boldsymbol{\alpha})$. This has derivatives of all orders present for α_0 as exponentials of derivatives, but can be approximated as a Fokker-Planck equation for $P(\boldsymbol{\alpha})$ 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:

$$\begin{aligned}
\dot\alpha_1&=[-i\omega_1\alpha_1-\gamma_f\alpha_1-i\lambda\alpha_2\alpha_--E(t)]+G_{\alpha_1}(t) ,\\
\dot\alpha_2&=(-i\omega_2\alpha_2-\gamma_f\alpha_2-i\lambda\alpha_1\alpha_+)+G_{\alpha_2}(t) ,\\
\dot\alpha_+&=[i\omega_0\alpha_+-2i\lambda\alpha_1^*\alpha_2\alpha_0-(\gamma_a/2)\alpha_+]+G_{\alpha_+}(t) ,\\
\dot\alpha_-&=[-i\omega_0\alpha_-+2i\lambda\alpha_2^*\alpha_1\alpha_0-(\gamma_a/2)\alpha_-]+G_{\alpha_-}(t) ,\\
\dot\alpha_0&=[-i\lambda\alpha_1\alpha_2^*\alpha_++i\lambda\alpha_1^*\alpha_2\alpha_--\gamma_a(\tfrac{1}{2}+\alpha_0)]+G_{\alpha_0}(t) .
\end{aligned} \quad (4)$$

Here 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 correlations of the random forces are given by

$$\begin{aligned}
\langle G_{\alpha_+}(t)G_{\alpha_+}(t')\rangle_R&=-i\lambda\alpha_1^*\alpha_2\alpha_+\delta(t-t') ,\\
\langle G_{\alpha_-}(t)G_{\alpha_-}(t')\rangle_R&=i\lambda\alpha_1\alpha_2^*\alpha_-\delta(t-t') ,\\
\langle G_{\alpha_0}(t)G_{\alpha_0}(t')\rangle_R&=[-(i\lambda/2)(\alpha_1\alpha_2^*\alpha_+-\alpha_1^*\alpha_2\alpha_-)\\
&\qquad+2\gamma_a(\tfrac{1}{2}+\alpha_0)]\delta(t-t') ,\\
\langle G_{\alpha_2}(t)G_{\alpha_0}(t')\rangle_R&=-i\lambda\alpha_1\alpha_+\delta(t-t') ,\\
\langle G_{\alpha_2^*}(t)G_{\alpha_0}(t')\rangle_R&=i\lambda\alpha_1^*\alpha_-\delta(t-t') ,\\
\langle G_{\alpha_2}(t)G_{\alpha_-}(t')\rangle_R&=2i\lambda\alpha_1\alpha_0\delta(t-t') ,\\
\langle G_{\alpha_2^*}(t)G_{\alpha_+}(t')\rangle_R&=-2i\lambda\alpha_1^*\alpha_+\delta(t-t') .
\end{aligned}$$

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

$$\alpha_1=\beta_1 e^{-i\omega_1 t},\quad \alpha_1^*=\beta_1^* e^{i\omega_1 t},\quad \alpha_2=\beta_2 e^{-i\omega_2 t} ,$$
$$\alpha_2^*=\beta_2^* e^{i\omega_2 t},\quad \alpha_-=\beta_- e^{-i\omega_0 t},\ \text{and}\ \alpha_+=\beta_+ e^{i\omega_0 t} .$$

We also assume the driving field $E(t)=Ee^{-i\omega_1 t}$ to be at exact resonance with the cavity pump mode. Now making use of the exact two photon resonance, $\omega_1=\omega_2+\omega_0$ we obtain the following Langevin equations:

$$\dot{\beta}_1=(-\gamma_f\beta_1-i\lambda\beta_2\beta_- -E)+G_{\beta_1}(t)\ ,$$
$$\dot{\beta}_2=(-\gamma_f\beta_2-i\lambda\beta_1\beta_+)+G_{\beta_2}(t)\ ,$$
$$\dot{\beta}_+=[-2i\lambda\beta_1^*\beta_2\beta_0-(\gamma_a/2)\beta_+]+G_{\beta_+}(t)\ , \qquad (5)$$
$$\dot{\beta}_-=[2i\lambda\beta_2^*\beta_1\beta_0-(\gamma_a/2)\beta_-]+G_{\beta_-}(t)\ ,$$
$$\dot{\beta}_0=[-i\lambda\beta_1\beta_2^*\beta_+ +i\lambda\beta_1^*\beta_2\beta_- -\gamma_a(\tfrac{1}{2}+\beta_0)]+G_{\beta_0}(t),$$

where $G_{\alpha_i}(t)=G_{\beta_i}(t)e^{i\omega_i t}$, $i=\{1,2,+,-,0\}$.

To obtain the mean-field solutions from the above equations we disregard the fluctuation terms. We also assume that the pump field is so strong that it is not modified by any feedback from the other cavity mode. To find the fluctuations around these mean values of the variables we go further to the next order in the perturbation. 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 [16]

$$d\delta\boldsymbol{\beta}(t)=-\underline{A}\,\delta\boldsymbol{\beta}(t)dt+\underline{B}d\mathbf{W}(t), \qquad (6)$$

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 a vector. The Ito stochastic differential equation (6) describes a multivariate Ornstein-Uhlenbeck process which is analytically solvable. For more details we refer to Ref. [16].

Next we calculate the power spectrum [16] for the Stokes emission in the usual way.

$$\begin{aligned} S_{\delta\beta_+\delta\beta_-}(\nu-\omega_2)&=\int_{-\infty}^{+\infty}\exp[-i(\nu-\omega_2)] \\ &\qquad\times\langle\delta\beta_+(t)\delta\beta_-(0)\rangle dt \\ &=\{[\underline{A}+i(\nu-\omega_2)\underline{I}]^{-1}\underline{B}\,\underline{B}^t \\ &\qquad\times[\underline{A}^t-i(\nu-\omega_2)\underline{I}]^{-1}\}_{\delta\beta_+\delta\beta_-}\ . \end{aligned}$$

The explicit calculation yields the spectra which are given in Figs. 2 and 3. For a weak field we find a single-peak Raman Stokes line centered at $\nu=\omega_2$. As the external field strength increases the spectrum splits into a doublet. The doublet structure is characteristic of vacuum field Rabi splitting in a cavity where a single quantum of energy transferred back and forth between the atom and the cavity. At a very high intensity, however, a triplet structure appears where the two strong sidebands are symmetrically separated from the central weak band.

III. CONCLUSION

We have calculated the power spectra of the radiation scattered from the Stokes mode of a three-level system interacting with an externally driven pump mode and a Stokes mode in a cavity. Anti-Stokes modes are eliminated by cavity-resonance condition. The model, which is a cavity version of Raman scattering, is similar to the Jaynes-Cummings model but with the single-mode operators in the interaction term replaced by the product of the creation operator of one mode and the annihilation operator of the other mode which describes a Raman coupling scheme. Notwithstanding the nonlinear nature of the interaction term in the present model as well as the nonlinearity intrinsic to the model system such as the Morse oscillator as considered in our earlier paper [14],

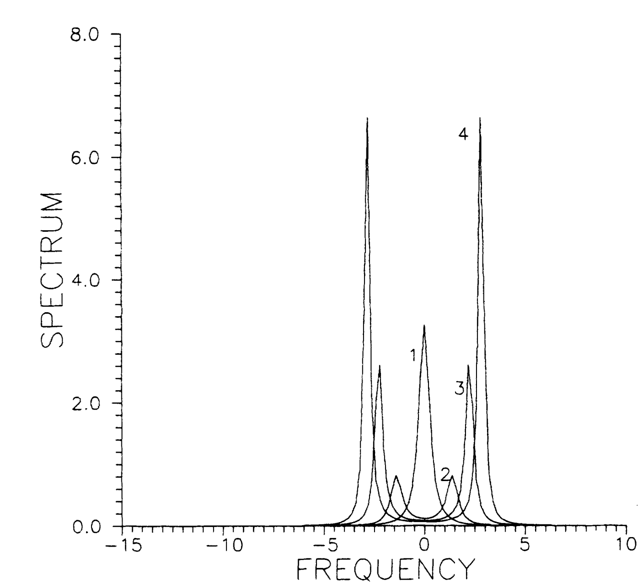

FIG. 2. The power spectrum $S(\nu-\omega_2)\gamma_a$ as a function of frequency $(\nu-\omega_2)/\gamma_a$ for $\gamma_f/\gamma_a=2.0$, $\lambda/\gamma_a=5.0$, and E/γ_a equal to 0.45 (curve 1), 0.8 (curve 2), 1.2 (curve 3), and 1.5 (curve 4) (both scales arbitrary).

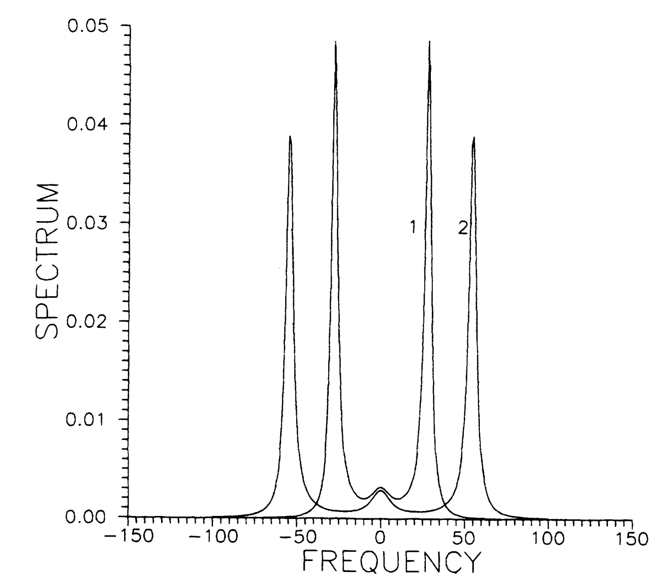

FIG. 3. The power spectrum $S(\nu-\omega_2)\gamma_a$ as a function of frequency $(\nu-\omega_2)/\gamma_a$ for $\gamma_f/\gamma_a=7.0$, $\lambda/\gamma_a=10.0$, and E/γ_a equal to 50 (curve 1) and 100 (curve 2) (both scales arbitrary).

spectral features are almost the same. This leads us to suspect that the system-field mode coupled oscillator model is generic at the fundamental cavity QED level. However, the detailed effect of nonlinearity in the study of resonance fluorescence is still out of the scope of the present treatment because of the fact that we are almost always restricted within a linearized scheme in calculating the spectra by the quantum-statistical method as it is currently developed.

ACKNOWLEDGMENT

Partial financial support from the Council of Scientific and Industrial Research under Grant No. 5/137/88 EMR II is thankfully acknowledged.

[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. Narozhky, 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, *ibid.* **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] B. R. Mollow, Phys. Rev. A **5**, 1522 (1972); **8**, 1949 (1973); C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 365 (1977); J. Ducuing, J. Chim. Phys. **64**, 209 (1967).

[9] P. L. Knight, Phys. Scr. **T12**, 51 (1986).

[10] S. J. D. Phoenix and P. L. Knight, J. Opt. Soc. Am. B **7**, 116 (1990).

[11] C. C. Gerry and J. H. Eberly, Phys. Rev. A. **42**, 6805 (1990).

[12] S. C. Gou, Phys. Rev. A **40**, 5116 (1989).

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

[14] G. Gangopadhyay and D. S. Ray, Phys. Rev. A **43**, 6424 (1991).

[15] P. D. Drummond and C. W. Gardiner, J. Phys. A **13**, 2353 (1980).

[16] C. W. Gardiner, *Handbook of Stochastic Methods* (Springer-Verlag, Berlin, 1983).