Quantum theory of Stokes–anti-Stokes scattering in a degenerate system in a cavity and vacuum-field Raman splittings

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(Received 15 October 1990)

The quantum theory of Stokes–anti-Stokes processes in a degenerate system in a cavity is developed. The Raman transition is pumped by a coherent field. Effects of many atoms, cavity losses, and thermal photons are included in the theory. An exact solution for the model is obtained. An explicit expression for the reduced density matrix of the cavity field is given. The spectrum of the cavity field is obtained and is interpreted in terms of the eigenstates of the effective Hamiltonian describing Stokes–anti-Stokes processes. The spectrum develops sidebands due to strong Raman coupling.

I. INTRODUCTION

The classical theory of the Stokes–anti-Stokes scattering and the coupling of Stokes and anti-Stokes fields is well understood.[1] However, very little has been done in quantum theory because of the tremendous complexity of the Hamiltonian simultaneously describing both Stokes and anti-Stokes processes. The situation is even more complex in a cavity because of the losses from the mirrors. In the special case when one considers a highly degenerate situation, an exact solution can be obtained.[2–4] The degenerate situation corresponds to (a) the initial and the final energy levels of the atom being degenerate, and (b) the same field mode interacting on both pump and the Raman transitions. If the field is initially in a Fock state $|n\rangle$, then the Rabi frequency of oscillations of the atom making two-photon Raman transitions is proportional to n. As a consequence, the atomic population and the photon number, etc., exhibit periodic behavior even in a field having arbitrary photon-number distribution. Schoendorff and Risken[3] generalized the work of Knight[2] to account for the losses from the cavity mirrors and for the presence of thermal photons in the cavity. They calculated the analytical expressions for various quasiprobability distributions like the P function, the Q function, and the Wigner function. It is rather remarkable that the degenerate Raman problem is exactly soluble even for cavities with finite Q and at finite temperatures.[5] The model so far considered has to be generalized in an important way—one has to include the pumping of the Raman transitions by an external field. Besides we will show that in order to see some of the quantum effects predicted in this paper, one has to include many atoms in the cavity.

Thus in this paper we consider the coupling of Stokes and anti-Stokes fields in a degenerate system consisting of *many* atoms. The Raman transition is being *pumped* by an external coherent field. The cavity losses and the effects of finite temperature are also included. We present the basic mathematical equations for the model system in Sec. II. In Sec. III we discuss the dressed states of the model. We also discuss on the basis of dressed states the kind of spectrum of the cavity photons one expects. In Sec. IV we present an exact solution for the density matrix of the field. The time evolution of the cavity field and its statistics can be obtained from the solutions of this section. In Sec. V we show how the strong Raman coupling between many atoms and the field can lead to new spectral features which may be termed as vacuum-field Raman splittings of the spectra.

II. MODEL

We consider a system of atoms undergoing Raman transitions between two degenerate states $|1\rangle$ and $|2\rangle$ on interaction with a single-mode quantized field inside a lossy cavity. The Raman transitions take place through the intermediate states $|j\rangle$. By eliminating the virtual levels $|j\rangle$ from the equations of motion, the Raman coupling between the levels $|1\rangle$ and $|2\rangle$ is described by the effective Hamiltonian[2]

$$H_R = \hbar g a^\dagger a (S^+ + S^-) = 2\hbar g a^\dagger a S_x \ , \tag{2.1}$$

where $a^\dagger$ (a) is the creation (annihilation) operator for the cavity field and $S^+ = |1\rangle\langle 2|$, $S^- = |2\rangle\langle 1|$ are the atomic dipole operators. The term $a^\dagger a S^+$ in Eq. (2.1) is responsible for the Stokes transition whereas $a^\dagger a S^-$ gives rise to the anti-Stokes transition in the scattered radiation. These lines, however, coincide in frequency as the Raman coupled states $|1\rangle$ and $|2\rangle$ are degenerate in the model considered here. Equation (2.1) is for a single atom, where the operators $\mathbf{S}$ correspond to spin value equal to half. We will account for the presence of *many* atoms (N) in the cavity. The Hamiltonian is still given by (2.1) but now $\mathbf{S}$ will correspond to angular momentum value $N/2$. Since it is the properties of the field which are of interest in the study of Raman scattering, it is, therefore, desirable to excite the cavity field mode. This can be done by coupling the mode to an external pump. The coupling of the cavity field mode with the external pump is described by the Hamiltonian

$$H_p = \hbar \exp(-i\omega_0 t) G a^\dagger + \hbar \exp(i\omega_0 t) G^* a \ , \tag{2.2}$$

where the pump frequency ω_0 is assumed to be equal to the frequency of the mode driving the Raman transitions. Under the action of the interactions (2.1) and (2.2) the density matrix ρ of the system, in the frame rotating with the frequency ω_0 of the field, evolves as

$$\frac{d\rho}{dt} = -i[H,\rho]/\hbar \ , \tag{2.3}$$

where

$$H = H_R + \hbar G a^\dagger + \hbar G^* a \ . \tag{2.4}$$

Equation (2.3), however, ignores the cavity losses and also the interaction of the atoms with thermal photons which are always present in many experimental situations. The effects of the thermal photons are particularly significant in the experiments involving Rydberg atoms even at very low temperatures.[6,7] The density-matrix equation including the effects of thermal photons and cavity damping is given by

$$\frac{d\rho}{dt} = -i[H,\rho]/\hbar + L_f \rho \ , \tag{2.5}$$

where

$$L_f \rho = \kappa(\bar{n}+1)(2a\rho a^\dagger - a^\dagger a \rho - \rho a^\dagger a) + \kappa \bar{n}(2a^\dagger \rho a - a a^\dagger \rho - \rho a a^\dagger) \tag{2.6}$$

describes the irreversible cavity losses at a rate κ in the presence of thermal photons with average number $\bar{n}$. These losses are expected to broaden the otherwise sharp Raman lines.

Our work is based on the exact analytic solution of the master equation (2.5). Note that special cases of (2.7) have been discussed earlier.[2,3] For a *single atom* Knight[2] introduced the model Hamiltonian (2.1) and obtained the time-dependent populations in the states $|1\rangle$ and $|2\rangle$. He found periodic behavior. Again for a *single* atom and in the *absence of any pumping* Schoendorff and Risken[3] obtained an expression for the density matrix in the presence of cavity losses and thermal photons. In this paper we study the statistical and spectral properties of the field for $N \neq 1$, $G \neq 0$.

III. THE DRESSED STATES

One can understand the dynamical features by examining the eigenstates of H. In order to solve the eigenvalue problem we first introduce the eigenstates of S_x. Let the eigenstate of S_x be denoted by $|\psi_m\rangle$. Here m takes values from $-N/2$ to $+N/2$ in steps of unity. The eigenstates of S_x can be related to the eigenstates $|m\rangle$ of S_z and S^2 via the rotation operator

$$|\psi_m\rangle = \exp\left[-\frac{i\pi}{2} S_y\right] |m\rangle \ . \tag{3.1}$$

Let us now write the eigenstates of H in the form

$$H|\psi_m\rangle|\phi_{nm}\rangle = \hbar\omega_{mn}|\psi_m\rangle|\phi_{nm}\rangle \ , \tag{3.2}$$

where $|\phi_{nm}\rangle$ is the photon part of the wave function. On using (2.4) and (3.1) in (3.2) we get the eigenvalue equation for $|\phi_{nm}\rangle$

$$(2gma^\dagger a + Ga^\dagger + G^* a)|\phi_{n,m}\rangle = \omega_{nm}|\phi_{nm}\rangle \ . \tag{3.3}$$

The eigenvalue equation can be written in the form

$$\left[b_m^\dagger b_m - \frac{|G^2|}{4g^2 m^2}\right]|\phi_{nm}\rangle = \frac{\omega_{nm}}{2gm}|\phi_{nm}\rangle \ , \tag{3.4}$$

where

$$b_m = a + \frac{G}{2mg} = D^\dagger\left[\frac{G}{2mg}\right] a D\left[\frac{G}{2mg}\right] \ , \tag{3.5}$$

and where $D(\alpha)$ is the displacement operator[8] $\exp(a^\dagger \alpha - a\alpha^*)$. Note that b operators obey the same algebra as a operators. Thus the eigenvalues and eigenfunctions are

$$\begin{aligned} \omega_{nm} &= 2gm\left[n - \frac{|G|^2}{4g^2 m^2}\right] \ , \\ |\phi_{nm}\rangle &= D^\dagger\left[\frac{G}{2mg}\right]|n\rangle \ , \end{aligned} \tag{3.6}$$

where $|n\rangle$ is the Fock state of the operator $a^\dagger a$. Thus to summarize, the eigenstates $|\chi_{mn}\rangle$ of H are

$$|\chi_{mn}\rangle = \left[\exp\left[-\frac{i\pi}{2}S_y\right]|m\rangle\right] \times \left[\exp\left[-\frac{a^\dagger G}{2mg} + \frac{aG^*}{2mg}\right]|n\rangle\right] \tag{3.7}$$

with eigenvalues

$$\omega_{nm} = 2mg\left[n - \frac{|G|^2}{4g^2 m^2}\right] \ . \tag{3.8}$$

The ground state of the combined atom-field system is $\chi_{N/2,0}$. Note that for a single atom, $m = \pm\frac{1}{2}$.

We next examine the expression for the spectrum of the cavity field. Note that the field operator a has *no* matrix element connecting states with *two different m values.* Using (3.7) one finds that

$$\begin{aligned} \langle \chi_{mn}|a|\chi_{mn'}\rangle &= \left\langle n \left| D\left[\frac{G}{2mg}\right] a D^\dagger\left[\frac{G}{2mg}\right] \right| n' \right\rangle \\ &= \left\langle n \left| \left[a - \frac{G}{2mg}\right] \right| n' \right\rangle \\ &= \sqrt{n'}\delta_{n,n'-1} - \frac{G}{2mg}\delta_{nn'} \ . \end{aligned} \tag{3.9}$$

Thus in the rotating frame the operator a will have a time dependence determined by

$$-\omega_{nm} + \omega_{n'm} = \begin{cases} 2gm(-n+n') = 2mg & \text{if } n' = n+1 \\ 0 & \text{if } n = n' \end{cases} \tag{3.10}$$

Thus the cavity field is expected to have frequency components

$$\Omega \equiv \omega_0, \quad \omega_0 \pm 2g|m| \ . \tag{3.11}$$

Therefore for a large number of atoms, the spectrum of the cavity field will show additional sidebands at $\pm 2g|m|$, $m=-N/2$ to $+N/2$.

IV. EXACT SOLUTION OF THE MASTER EQUATION (2.5)

In this section we derive the exact solution of (2.5). We define a set of field density matrices by

$$\rho^{(m)} = \langle \psi_m|\rho|\psi_m\rangle / p^{(m)} \ , \tag{4.1}$$

$$p^{(m)} = \mathrm{Tr}_f \langle \psi_m|\rho|\psi_m\rangle \neq 1 \ .$$

The density matrix $\rho^{(f)}$ of the field alone is given by

$$\rho^{(f)} = \sum_{m=-N/2}^{+N/2} p^{(m)}\rho^{(m)}, \quad \mathrm{Tr}_f \rho^{(f)} = 1 \ . \tag{4.2}$$

It can be shown from (2.5) that $p^{(m)}$ is a constant of motion. On combining (2.5) and (4.1) we obtain the equation for $\rho^{(m)}$

$$\dot{\rho}^{(m)} = -2gmi[a^\dagger a, \rho^{(m)}] + L_f \rho^{(m)} - i[Ga^\dagger + G^* a, \rho^{(m)}] \ . \tag{4.3}$$

Equation (4.3) is to be solved subject to the initial condition determined by (4.1). The solution of equations like (4.3) is well known. This equation is the same as the equation for a damped, driven harmonic oscillator[9] interacting with a heat bath. The driving field is *not* on resonance with the oscillator frequency. The detuning parameter is 2 mg.

The steady-state solution of (4.3) is

$$\rho^{(m)} = N_m \exp\left[-\beta\left[a^\dagger - \frac{iG^*}{\kappa - 2igm}\right]\left[a + \frac{iG}{\kappa + 2igm}\right]\right] , \tag{4.4}$$

where

$$(e^\beta - 1)^{-1} = \bar{n} \ , \tag{4.5}$$

and where N_m is determined by the normalization condition

$$\mathrm{Tr}_f \rho^{(m)} = 1 \ . \tag{4.6}$$

Thus the complete density matrix for the field in the steady state is

$$\rho^{(f)} = \sum_{m=-N/2}^{+N/2} p^{(m)} N_m \exp\left[-\beta\left[a^\dagger - \frac{iG^*}{\kappa - 2igm}\right] \times \left[a + \frac{iG}{\kappa + 2igm}\right]\right] . \tag{4.7}$$

Hence the field in the steady state is the incoherent superposition of the density matrices which themselves represent the superposition of thermal and coherent fields. Thus the mean value of any physical observable can be expressed as

$$\langle G\rangle = \sum_{m=-N/2}^{+N/2} p^{(m)} G^{(m)} \ , \tag{4.8}$$

where $G^{(m)}$ represents the mean value with respect to the density matrix (4.4). The field in the steady state exhibits no nonclassical features since each individual density matrix (4.4) represents superposition of thermal and coherent fields.[10]

The time-dependent solutions of (4.3) can also be obtained. For this purpose it is convenient to work with the Glauber-Sudarshan P function defined by

$$\rho^{(m)} = \int P^{(m)}(\alpha)|\alpha\rangle\langle\alpha| d^2\alpha \ , \tag{4.9}$$

where $|\alpha\rangle$ is a coherent state. The function $P^{(m)}$ is found to obey the equation

$$\frac{\partial P^{(m)}}{\partial t} = \frac{\partial}{\partial \alpha}[(\kappa + iG + 2img)\alpha p^{(m)}] + \kappa\bar{n}\frac{\partial^2}{\partial\alpha\,\partial\alpha^*}P^{(m)} + \text{c.c.} \tag{4.10}$$

The equation for $P^{(m)}$ has the same structure as the linearized Fokker-Planck equation whose solution is well known.[11]

V. SPECTRUM OF THE CAVITY FIELD

In this section we calculate the spectrum of the field in the cavity. We show how the spectrum can develop sidebands even if the external field is zero. Such sidebands can be termed as vacuum-field Raman splittings like the vacuum-field Rabi splittings which are well known[12] in the context of the Jaynes-Cummings model. Note (4.2) and (4.3) show that

$$\begin{aligned}\langle a^\dagger(t+\tau)\rangle &= \mathrm{Tr}[\rho^{(f)}(t+\tau)a^\dagger] \\ &= \sum_m p^m \mathrm{Tr}[\rho^{(m)}(t+\tau)a^\dagger] \\ &= \sum_m p^m \left[\exp(-\kappa\tau + 2igm\tau)\mathrm{Tr}[\rho^{(m)}(t)a^\dagger] + iG\left[\frac{1-\exp[-\tau(\kappa - 2igm)]}{\kappa - 2igm}\right]\mathrm{Tr}[\rho^{(m)}(t)]\right] .\end{aligned} \tag{5.1}$$

The two-time correlation function $\langle a^\dagger(t+\tau)a(t)\rangle$ can be obtained using (5.1) and the equation regression theorem. Thus in the steady state

$$\lim_{t\to\infty}\langle a^\dagger(t+\tau)a(t)\rangle=\sum_m p^{(m)}\left[\exp(-\kappa\tau+2igm\tau)\mathrm{Tr}(\rho^{(m)}a^\dagger a)+iG^*\left[\frac{1-\exp[-\tau(\kappa-2igm)]}{\kappa-2igm}\right]\mathrm{Tr}(\rho^{(m)}a)\right]. \quad (5.2)$$

The expectation values appearing in (5.2) can be obtained from (4.4):

$$\mathrm{Tr}(\rho^{(m)}a)=-\frac{iG}{\kappa+2igm},$$
$$\mathrm{Tr}(\rho^{(m)}a^\dagger a)=\left|\frac{G}{\kappa+2igm}\right|^2+\bar{n}. \quad (5.3)$$

On substituting (5.3) in (5.2) we get

$$\lim_{t\to\infty}\langle a^\dagger(t+\tau)a(t)\rangle=\bar{n}\sum_m p^{(m)}e^{-\kappa\tau+2igm\tau}+\sum_m p^{(m)}\left|\frac{G}{\kappa+2igm}\right|^2. \quad (5.4)$$

Note that the spectrum of radiation in the absence of any atoms is obtained from

$$\lim_{t\to\infty}\langle a^\dagger(t+\tau)a(t)\rangle=\bar{n}e^{-\kappa\tau}. \quad (5.5)$$

The spectrum of the cavity field thus consists of a coherent component at ω_0 and a number of incoherent components at the frequencies

$$\Omega_m=\omega_0\pm 2g|m|, \quad (5.6)$$

with half-width κ. The weight factor of the incoherent component at Ω_m is determined by $p^{(m)}$ which in turn is determined by the initial conditions. At time $t=0$, the density matrix of the system will be factorizable in terms of the atomic and field parts. Thus in terms of the atomic part $\rho^{(A)}$ one will get

$$p^{(m)}=\langle\psi_m|\rho^{(A)}|\psi_m\rangle. \quad (5.7)$$

If at time $t=0$, the atomic population is equally distributed in two states, then $\rho^{(A)}=|0\rangle\langle 0|$ and hence

$$p^{(m)}=|\langle 0|e^{-i\pi/2S_y}|m\rangle|^2 \quad (5.8)$$

which on using the properties of the rotation matrices[13] reduces to

$$p^{(m)}=\frac{\left[\frac{N}{2}+m\right]!\left[\frac{N}{2}-m\right]!\frac{N}{2}!\frac{N}{2}!}{2^N}\times\left|\frac{\sum_{p=m}^{N/2-m}(-1)^p}{\left[\frac{N}{2}-p\right]!p!(p-m)!\left[\frac{N}{2}+m-p\right]!}\right|^2. \quad (5.9)$$

Note further that $p^{(m)}=0$ if $N/2+m$ is odd. Thus the sidebands, corresponding to m values such that $N/2+m$ is odd, disappear. The sidebands will be resolved if $g>\kappa$. Generally the Raman matrix element g is rather small and hence it may be difficult to observe such sidebands unless the number of atoms is large[14] when at least the last couple of sidebands should be observable. The origin of these sidebands can be traced back to the matrix elements of the field operators among the dressed states [Eqs. (3.9)–(3.11)].

Thus, in conclusion, we have shown how the exact solutions for the quantum dynamics of Stokes-anti-Stokes transitions in a degenerate system of many atoms in a cavity with finite Q and thermal photons can be obtained. The pumping of the Raman transition is included. We have further shown how the strong interaction with the cavity mode leads to vacuum field Raman splittings in the spectrum of the cavity field.

ACKNOWLEDGMENTS

G.S.A. is grateful to the Department of Science and Technology (Government of India) for supporting this collaboration.

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[14]Note that the Raman matrix element g will be equal to g_0^2/Δ, where g_0 is the single-photon Rabi frequency and Δ is the detuning from the intermediate state. For Rydberg atoms g_0 can be of the order 10^4-10^5 Hz and if $\Delta \sim 100$ MHz, $\kappa \lesssim 10^2$ Hz (corresponding to say $\omega_0 \sim 100$ GHz, $Q \sim 10^9$) then one should be able to see the sidebands with $N \sim 10^3$. For optical transitions a similar situation can be obtained by a suitable choice of parameters.