Quantum effects in four-wave mixing in a cavity

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The four-wave-mixing signal at $2\omega_1-\omega_2$ from a collective system of N two-level atoms dissipating through spontaneous emission and dephasing and driven by weak external fields of frequencies ω_1 and ω_2 in a cavity of arbitrary Q is found by evaluating exactly the susceptibility $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$. The exact results show that the customarily employed secular approximation for treating the problem of a strongly coupled atom-cavity system is hopelessly inadequate for describing the process of four-wave mixing. [S1050-2947(98)04805-7]

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In a recent paper [1] we addressed the issue of true signatures of the nonlinear and quantum effects in the interaction of a two-level atom with a single mode field in a cavity. As has been discussed in detail in that reference, the characteristic features of the field quantization and the two-level nature of the atom are contained in the structure of the spectrum of the second and higher excited manifolds of the dressed states. We showed there that the method of four-wave mixing provides a useful means of probing the manifolds up to the second one. The signatures of the quantum and the nonlinear effects are provided by the positions of the resonances in the third-order susceptibility $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$ as a function of ω_2 where ω_1 and ω_2 are the frequencies constituting the bichromatic field used as a probe.

The evaluation of the susceptibility in that paper is, however, restricted to the case when each atom interacts independently with the fields and is based on the secular approximation. That approximation (see [1–4] and the references therein) is applicable when the atom-cavity coupling is much stronger than the dissipations. We subsequently carried out an exact evaluation of that susceptibility for N atoms interacting collectively with the fields and found, rather surprisingly, that the predictions based on the secular approximation are in significant qualitative and quantitative disagreement with the exact results for all N even when that approximation is expected to hold. In this paper we present the results of the exact calculation of $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$ along with the predictions of the secular approximation for an arbitrary number N of atoms and highlight the differences between the two.

Following Ref. [1] we consider a system of N two-level atoms, each of transition frequency ω_0 interacting collectively with a cavity mode of frequency $\omega_c=\omega_0$. The quantized cavity mode is described by the operators $a,a^\dagger$, which obey the bosonic commutation relations whereas the atomic system is described by the spin operators $S_\pm,S_z$ that obey the angular momentum commutation relations. The atom-cavity field interaction is governed by the Hamiltonian ($\hbar=1$)

$$H_0=\omega_0 S_z+\omega_0 a^\dagger a+g(S_+a+a^\dagger S_-), \tag{1}$$

where g is the atom-field coupling constant. The interaction of atoms with the external weak bichromatic probe having frequencies ω_1 and ω_2 is described by the Hamiltonian

$$H_e(t)=d[\epsilon_1\exp(-i\omega_1 t)+\epsilon_2\exp(-i\omega_2 t)]S_+ + \text{H.c.}, \tag{2}$$

where ϵ_1 and ϵ_2 are the field amplitudes and d is the atomic dipole moment. The atoms and the field also dissipate energy governed by the Liouvillian

$$L\rho=\kappa(2a\rho a^\dagger-a^\dagger a\rho-\rho a^\dagger a)+\gamma(2S_-\rho S_+-S_+S_-\rho-\rho S_+S_-)+\gamma_c(2S_z\rho S_z-S_z^2\rho-\rho S_z^2), \tag{3}$$

where 2κ is the rate of the loss of photons, 2γ is the rate of the atomic radiative decay and γ_c describes the losses due to atomic dephasing. The dephasing may arise, for example, by atomic collisions. The dynamics of the system is then governed by the equation

$$\frac{d\rho}{dt}=-i[H_0+H_e,\rho]+L\rho \tag{4}$$

for the density matrix ρ. Here we are interested in the characteristics of the four-wave-mixing signal at the frequency $\Omega\equiv 2\omega_1-\omega_2$. That signal is determined by the third-order susceptibility $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)=d^*\ \mathrm{Tr}\,[\rho^{(3)}S_-]$ where $\rho^{(3)}$ is the relevant density matrix in the third order of perturbation in H_e [1].

We evaluate $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$ by following Ref. [1]. To that end it is convenient to work in the basis of the dressed states, i.e., the eigenstates of H_0. The lowest state of H_0 is, of course, $|-N/2,0\rangle$. The first excited manifold consists of the states $|\psi_0^\pm\rangle$, which correspond to the eigenvalues $\lambda_0^\pm=\omega_0(-N/2+1)\pm g\sqrt{N}$. The explicit expression for $|\psi_0^\pm\rangle$ is given in Ref. [1]. The second excited manifold of the dressed states is three (two) dimensional for $N>1$ ($N=1$). That space for $N>1$ is spanned by the eigenstates [4]

$$|\psi_i\rangle=\sum_{j=1}^{3} a_{ij}|-N/2+3-j,j-1\rangle \tag{5}$$

of H_0 where the explicit form of the a_{ij} is given in Ref. [4]. The corresponding eigenvalues are $E_\pm=(-N/2+2)\omega_0\pm g\sqrt{4N-2}$ and $E_0=(-N/2+2)\omega_0$. The eigenstates $|\psi_1^\pm\rangle$ of H_0 for the second excited manifold for $N=1$ correspond

to the eigenvalues $E_{\pm}=(-N/2+2)\omega_0 \pm g\sqrt{2}$ [1]. Next one determines the action of the field and the collective atomic operators on the dressed states and uses it to find how the dressed states transform under the action of the damping operator L. It can be shown that the operator L transforms each of the vectors

$$(\Psi_1,\Psi_2)\equiv(|\psi_0^+\rangle\langle -N/2,0|,|\psi_0^-\rangle\langle -N/2,0|);$$

$$(X_1;X_2;X_3;X_4)\equiv(|\psi_0^+\rangle\langle\psi_0^+|-|-N/2,0\rangle\langle -N/2,0|;|\psi_0^-\rangle \times\langle\psi_0^-|-|-N/2,0\rangle\langle -N/2,0|;|\psi_0^+\rangle \times\langle\psi_0^-|;|\psi_0^-\rangle\langle\psi_0^+|);$$

$$(Y_1;Y_2;Y_3)\equiv(|\psi_1\rangle\langle 0,-N/2|;|\psi_2\rangle\langle 0,-N/2|;|\psi_3\rangle \times\langle 0,-N/2|);$$

$$(Z_1,Z_2,Z_3,Z_4,Z_5,Z_6),$$

where $Z_i=|\psi_i\rangle\langle\psi_0^+|$; $Z_{i+3}=|\psi_i\rangle\langle\psi_0^-|$; $i=1,2,3$ onto itself. The susceptibility $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$ can now be evaluated by following Ref. [1].

As discussed in Ref. [1] , the resonances in $\chi^{(3)}(\omega_1,\omega_1,-\omega_2)$ as a function of ω_2 can provide information about the dressed-state spectrum if the dissipative losses are small compared with the atom-cavity coupling, i.e., if $\kappa,N\gamma,\gamma_c \ll \sqrt{N}g$. Under those conditions, the secular approximation [1–4] is expected to hold. The explicit expression for the susceptibility in the secular approximation for $N=1$ is given in Ref. [1]. On generalizing the arguments of Ref. [1] for $N=1$ to $N>1$, it follows that the resonances in the susceptibility in an N-atom system are expected to occur at (a) $\omega_2=\omega_0\pm\sqrt{N}g$; (b) $\omega_2=2\omega_1-\omega_0\pm\sqrt{N}g$; (c) $\omega_2=2\omega_1-\omega_0\pm g(\sqrt{4N-2}-\sqrt{N})$; (d) $\omega_2=2\omega_1-\omega_0 \pm g(\sqrt{4N-2}+\sqrt{N})$; (e) $\omega_2=\omega_1$; (f) $\omega_2=\omega_1\pm 2g\sqrt{N}$. The resonances (a) and (b) correspond to transitions between the first excited doublet and the ground state in the case of $N=1$. In the case of a multiatom system, contributions to those resonances arise also from transitions from the state $|\psi_2\rangle$ corresponding to the eigenvalue $(-N/2+2)\omega_0$ in the second excited manifold to the first excited one. Note that in a single atom system there is no transition from the second manifold to the first that has the same frequency as a transition from the first manifold to the ground state. The resonances (c) and (d) correspond to transitions between the first and the second excited manifolds. The resonances (e) and (f) do not correspond to any allowed transition. As shown in Ref. [1], the resonances (e) and (f) disappear for $N=1$ if $\gamma_c=0$. Those are thus like the collision-induced Bloembergen resonances [5]. On generalizing the arguments of Ref. [1] for $N=1$ to $N>1$ it follows that the resonance (f) is due to atomic dephasing induced processes between the pair of dressed states $|\psi_0^{\pm}\rangle$, which are separated by $2\sqrt{N}g$ and the ground state. Those types of resonances have also been reported by Agarwal [6] in the case of second-order response of N two-level atoms in a cavity to a modulated field in the secular approximation.

Thus the resonances (a), (b), (e), and (f) reveal the nature of the dressed-state spectrum up to the first excited state. As has already been pointed out earlier, that part of the dressed-state spectrum does not carry any signature either of the nonlinearity of the atom-field interaction or of the field quantization. The signature of those effects is contained in the nature of the spectrum of the higher excited states, which is revealed by the resonances (c) and (d). We, therefore, refer to resonances (c) and (d) as ''quantum resonances.''

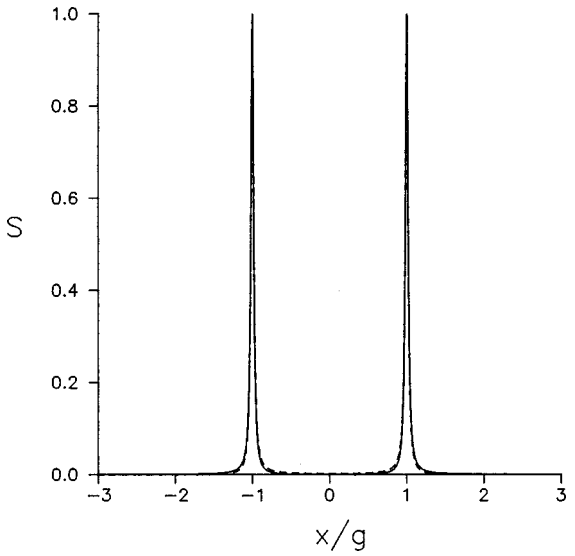

FIG. 1. $S=|\chi^{(3)}(X)|/|\chi^{(3)}(g)|$ as a function of $X/g\equiv(\omega_2-\omega_0)/g$ for $N=1$, $\omega_1=\omega_0$; $\gamma=0.01g$, $\kappa=0.03g$, and $\gamma_c=0$. The solid curve is the result of exact calculation whereas the dashed one is obtained by making the secular approximation with $|\chi^{(3)}(g)|_{\text{solid}}/|\chi^{(3)}(g)|_{\text{dashed}}=0.0018$.

Let us now present the results of numerical evaluation of $\chi^{(3)}$ by considering first the case of a single atom system. We present in Figs. 1–4 the plots of $|\chi^{(3)}|$ as a function of $\omega_2-\omega_0$ for $N=1$, $\omega_1=\omega_0$, $\gamma=0.01g$, $\kappa=0.03g$ and for different rates of the atomic dephasing. The curves are drawn after normalizing the maximum peak height in each case to one. The solid curves are the results of evaluation of $\chi^{(3)}$ without making the secular approximation whereas the dashed curves are the predictions of the secular

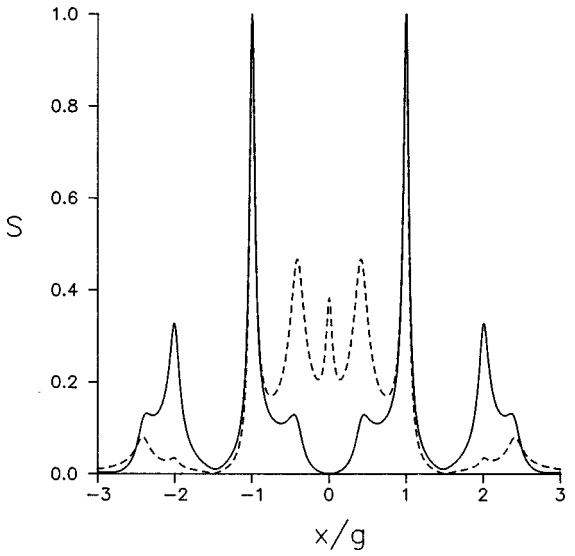

FIG. 2. The same as Fig. 1 but for $\gamma_c=0.04g$ with $|\chi^{(3)}(g)|_{\text{solid}}/|\chi^{(3)}(g)|_{\text{dashed}}=0.0078$.

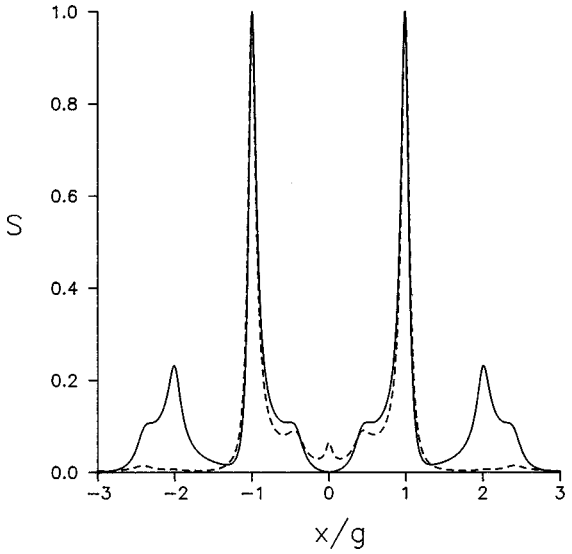

FIG. 3. The same as Fig. 1 but for $\gamma_c = 0.08g$ with $|\chi^{(3)}(g)|_{\text{solid}}/|\chi^{(3)}(g)|_{\text{dashed}} = 0.0018$.

approximation. Note first from the figure captions that the scaling factors for the dashed and the solid curves differ by orders of magnitude. Figure 1 for $\gamma_c = 0$ exhibits resonances only at $\omega_2 - \omega_0 = \pm g$ for the exact as well as the secular approximation calculations even though both of them predict other resonances. We have not been able to observe any other resonance for any other value of the damping rates as long as $\gamma_c = 0$. Those other resonances seem to be suppressed by the observed ones in the absence of the atomic dephasing. The quantum as well as the Bloembergen type resonances are exhibited in Figs. 2–4 for $\gamma_c \neq 0$. Note, however, the significantly large differences between the predictions of the exact and the approximate results. Note in particular that the exact calculation does not exhibit any resonance at $\omega_1 = \omega_2$ whereas that resonance is exhibited in the secular approximation. We have not been able to observe that resonance by exact calculations even for many other values of the damping rates. The differences in the predictions of the exact and the approximate calculations as regards the weight of the quantum resonances at $\omega_2 - \omega_0 = \pm g(\sqrt{2} \pm 1)$ as well as the Bloembergen type resonances at $\omega_2 - \omega_0 = \pm 2g$ are also clearly seen in Figs. 2–4. Thus the quantum resonances seem to be observable only in the presence of atomic dephasing even in the exact description.

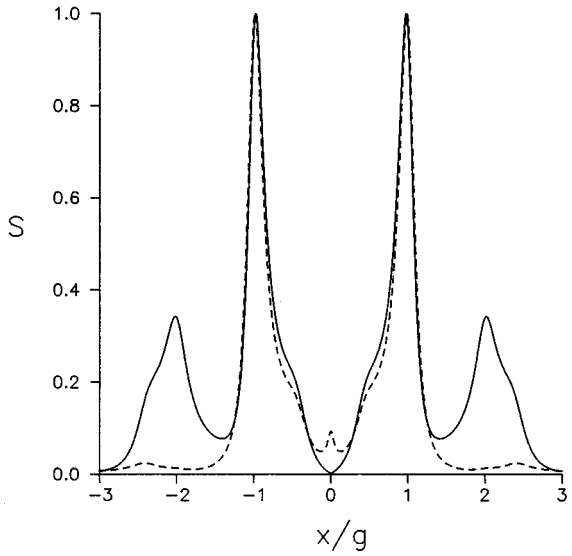

FIG. 4. The same as Fig. 1 but for $\gamma_c = 0.2g$ with $|\chi^{(3)}(g)|_{\text{solid}}/|\chi^{(3)}(g)|_{\text{dashed}} = 0.0018$.

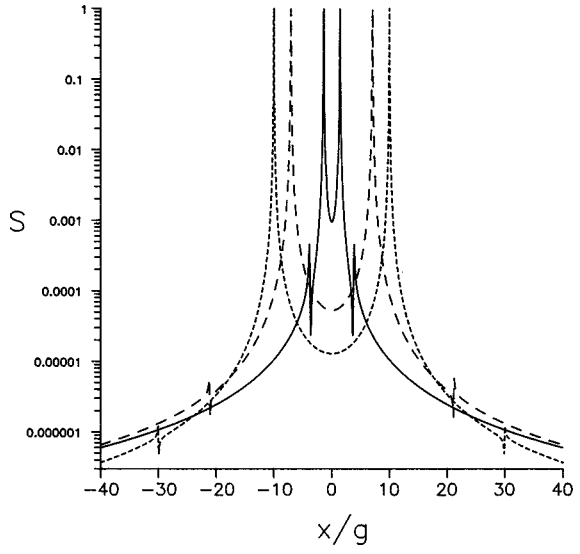

FIG. 5. $|\chi^{(3)}(X)|/|\chi^{(3)}(g\sqrt{N}|)$ on a logarithmic scale as a function of $X/g \equiv (\omega_2 - \omega_0)/g$ for $\omega_1 = \omega_0$, $\kappa = 0.01$, $N\gamma = 0.02$, $\gamma_c = 0.01$ for $N = 2$ (solid), 50 (long dashed), 100 (small dashes).

The disagreement between the exact results and those obtained in the secular approximation seems surprising. However, a close examination of the expression for the susceptibility (not given here because of lack of space) reveals the origin of the stated discrepency. We find that any term contributing to a resonance is multiplied by a sum of terms that are off the given resonance. That sum of terms contains diagonal as well as nondiagonal elements of the inverse of the matrices representing the action of $L + iz$, where z is linear combination of frequencies, on the vectors introduced towards the end of the text following Eq. (5). Since the off-resonance contribution of the diagonal elements is of the same order of magnitude as that of the off-diagonal ones, it follows that the secular approximation, which is based on ignoring the off-diagonal elements, is unjustified in this case. The secular approximation holds good, however, for the first-order perturbation as in that case there are no ''product of sums'' type of terms.

Next, for $N > 1$, we find that $\chi^{(3)}$ remains close to zero for $\gamma_c = 0$ if it is evaluated without making the secular approximation but is finite and exhibits resonances at $\omega_0 \pm \sqrt{N}g$ in the secular approximation for all N. Varada *et al.* [4], working in the secular approximation, have also reported similar resonances for large N. That result of the secular approximation is evidently misleading because the nonlinear susceptibility is expected to approach zero in the limit of large N. That is because in that limit the eigenvalues of the second manifold, given after Eq. (5), approach $(-N/2+2)\omega_0 \pm 2g\sqrt{N}, (-N/2+2)\omega_0$, which, together with the eigenvalues of the lower eigenstates define the energy levels of a harmonic oscillator. For a general proof of the linearity of the spectrum of an N-atom system interacting with a single-mode field see Scharf [7]. Since, as is well known, a har-

monic oscillator cannot lead to any nonlinear wave mixing, it follows that $\chi^{(3)}$ should approach zero for large N. Our exact calculations thus confirm that the conclusions of Varada *et al.* [4] are the artifact of the secular approximation.

The susceptibility is found to be finite for smaller N exhibiting some of the expected resonances if $\gamma_c \neq 0$ as is shown by the plots in Fig. 5 for a given set of rates of damping and different values of the number N of atoms.

We are, however, unable to explain why the susceptibility is finite for $N=1$ but vanishingly small, not only for large N but for all $N>1$ in the absence of the atomic dephasing, i.e., for $\gamma_c=0$. A crucial difference between $N=1$ and $N>1$ is the presence of a third level in the second excited manifold for $N>1$. The transitions involving that level appear to be playing a crucial role in suppressing the wave mixing perhaps by way of causing destructive interference between two channels of transition. However, we leave that as an open question.

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