Subnatural linewidth averaging for coupled atomic and cavity-mode oscillators

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We calculate the spontaneous-emission spectrum and the spectrum of weakly driven fluorescence for a two-level atom coupled to a resonant-cavity mode. For strong atom-cavity coupling the spectra split into two peaks that can have subnatural linewidths. If the cavity linewidth is negligible, the spontaneous-emission spectrum has half the radiative linewidth of the atom; the spectrum of weakly driven fluorescence shows an additional 36% squeezing-induced narrowing. These effects can be observed using coupled-field and collective-polarization oscillators excited in a cavity containing N two-level atoms.

The spontaneous-emission rate for an atom in free space can change when the atom radiates inside an electromagnetic cavity. The emission rate is reduced if the cavity subtends a large solid angle at the atom and has no resonant modes into which the atom can emit.[1] It is increased when the atom couples strongly to a resonant mode of the cavity.[2] These effects are explained by perturbation theory; the altered emission rate is obtained from Fermi's golden rule using a density of states modified to account for the cavity boundary conditions. However, when the coupling between the atom and the cavity mode is so strong that a photon emitted into the cavity is likely to be reabsorbed before it escapes, perturbation theory cannot be used. Previous authors have studied atomic decay under these conditions assuming that spontaneous emission to modes other than the privileged cavity mode is negligible. Haroche and Raymond[3] show that an initially excited atom undergoes single-quantum Rabi oscillations which decay at a rate determined by the cavity Q. Sanchez-Mondragon *et al.*[4] derive a double-peaked "spontaneous-emission" spectrum by convolving the single-quantum Rabi oscillation in a lossless cavity against an exponential detector response function. This is not, however, a spontaneous-emission spectrum in the usual sense of irreversible decay into a reservoir of vacuum modes; in particular, linewidths are assigned by the detector response function and are not radiative in origin.

In this paper we consider the interaction between a two-level atom and a resonant cavity mode that subtends a *small* solid angle at the atom so that the spontaneous-emission rate γ into free-space is not negligible compared to the photon decay rate 2κ from the cavity. We derive the spontaneous emission spectrum and spectrum of weakly driven fluorescence measured by observing the light emitted (scattered) by the atom into free space. Our derivation places no restriction on the coupling strength g between the atom and the cavity mode. For $\kappa \gg g \gg \gamma/2$, we recover the increased linewidth associated with cavity-enhanced spontaneous emission. In the strong-coupling limit, $g \gg \kappa, \gamma/2$, the spontaneous-emission spectrum and the spectrum of weakly driven fluorescence are doublets, similar to those obtained by Sanchez-Mondragon *et al.*[4] However, our spectra have meaningful radiative linewidths. For $\kappa \ll \gamma/2$ they have linewidths equal to one-half and one-third the free-space radiative linewidth of the atom, respectively.

The master equation describing the resonant interaction between a two-level atom and a single cavity mode, including both atomic and cavity loss, is given by

$$\dot{\rho}=(1/i\hbar)[\hat{H},\rho]+(\gamma/2)(2\hat{\sigma}_-\rho\hat{\sigma}_+-\hat{\sigma}_+\hat{\sigma}_-\rho-\rho\hat{\sigma}_+\hat{\sigma}_-)+\kappa(2\hat{a}\rho\hat{a}^\dagger-\hat{a}^\dagger\hat{a}\rho-\rho\hat{a}^\dagger\hat{a}) , \tag{1}$$

where $\hat{H}=i\hbar g(\hat{\sigma}_-\hat{a}^\dagger-\hat{\sigma}_+\hat{a})$ and ρ is the reduced density operator in the interaction picture; $\hat{a}^\dagger$ and $\hat{a}$ are creation and annihilation operators for the cavity mode, and $\hat{\sigma}_-$, $\hat{\sigma}_+$, and $\hat{\sigma}_z$ are Pauli pseudospin operators for the atom. Equation (1) describes a composite atom-cavity-mode system that radiates via two distinct channels: by the coupling of the atom to free-space modes (the term proportional to $\gamma/2$), and by loss through the cavity mirrors (the term proportional to κ). We will calculate spectra for the light emitted by the atom into free space.

To describe spontaneous emission for arbitrary values of g, κ, and $\gamma/2$, we solve Eq. (1) in the three-state basis $|+\rangle|0\rangle, |-\rangle|1\rangle, |-\rangle|0\rangle$, where $|+\rangle$ and $|-\rangle$ are the upper and lower states of the atom and $|1\rangle$ and $|0\rangle$ are the one-photon and zero-photon Fock states of the field. In this basis ρ has eight independent matrix elements. The equations of motion for these matrix elements can be written as two sets of coupled equations for operator expectation values,

$$\langle \dot{\hat{a}} \rangle = g\langle \hat{\sigma}_- \rangle - \kappa \langle \hat{a} \rangle , \tag{2a}$$

$$\langle \dot{\hat{\sigma}}_- \rangle = -g\langle \hat{a} \rangle - (\gamma/2)\langle \hat{\sigma}_- \rangle , \tag{2b}$$

and

$$\frac{d}{dt}\langle \hat{a}^\dagger \hat{a}\rangle = g(\langle \hat{a}^\dagger \hat{\sigma}_-\rangle + \text{c.c.}) - 2\kappa\langle \hat{a}^\dagger \hat{a}\rangle , \tag{3a}$$

$$\frac{d}{dt}\langle \hat{\sigma}_+ \hat{\sigma}_-\rangle = -g(\langle \hat{a}^\dagger \hat{\sigma}_-\rangle + \text{c.c.}) - \gamma\langle \hat{\sigma}_+ \hat{\sigma}_-\rangle , \tag{3b}$$

$$\frac{d}{dt}\langle \hat{a}^\dagger \hat{\sigma}_-\rangle = -g(\langle \hat{a}^\dagger \hat{a}\rangle - \langle \hat{\sigma}_+ \hat{\sigma}_-\rangle) - (\kappa + \gamma/2)\langle \hat{a}^\dagger \hat{\sigma}_-\rangle . \tag{3c}$$

The spontaneous-emission spectrum measured by an ideal detection system (with negligible bandwidth) is given by[5]

$$2\pi S(\omega) = \left[\int_0^\infty dt\, C_s(t,t)\right]^{-1} \times \int_0^\infty dt \int_0^\infty dt' e^{-i(\omega-\omega_0)(t-t')} C_s(t,t') , \tag{4}$$

where $C_s(t,t') = \langle \hat{\sigma}_+(t)\hat{\sigma}_-(t')\rangle$ and the term in square brackets normalizes the integral of $S(\omega)$ to unity. The double time integral on the right-hand side of Eq. (4) is proportional to the probability for the detector—tuned to the frequency ω—to record a photon in an infinite observation time beginning at $t=0$.

According to the quantum regression theorem the correlation functions $C_s(t,t')$ and $C_a(t,t') = \langle \hat{\sigma}_+(t)\hat{a}(t')\rangle$ obey the same equations as $\langle \hat{\sigma}_-\rangle$ and $\langle \hat{a}\rangle$. Thus, for $t' \geq t$,

$$\dot{C}_a = gC_s - \kappa C_a, \quad \dot{C}_s = -gC_a - (\gamma/2)C_s , \tag{5}$$

where the dot denotes differentiation with respect to t'. The initial conditions $C_a(t,t) = \langle (\hat{\sigma}_+ \hat{a})(t)\rangle$ and $C_s(t,t) = \langle (\hat{\sigma}_+ \hat{\sigma}_-)(t)\rangle$ needed to solve Eqs. (5) are given by the solution to Eqs. (3). We assume that the atom-cavity-mode system is prepared in the state $|+\rangle|0\rangle$ (more generally it might be prepared in an arbitrary single-quantum state). Equations (3) are then solved with $\langle (\hat{a}^\dagger \hat{a})(0)\rangle = \langle (\hat{a}^\dagger \hat{\sigma}_-)(0)\rangle = 0$ and $\langle (\hat{\sigma}_+ \hat{\sigma}_-)(0)\rangle = 1$, and from Eqs. (4) and (5) we obtain the spontaneous-emission spectrum

$$2\pi S(\omega) = \left[\frac{1}{2}\frac{\kappa(\kappa+\gamma/2)+g^2}{(\kappa+\gamma/2)(\kappa\gamma/2+g^2)}|\lambda_+ - \lambda_-|^2\right]^{-1} \times \left|\frac{\lambda_+ + \kappa}{\lambda_+ - i(\omega-\omega_0)} - \frac{\lambda_- + \kappa}{\lambda_- - i(\omega-\omega_0)}\right|^2 , \tag{6}$$

where $\lambda_\pm = -\frac{1}{2}(\kappa+\gamma/2) \pm [(\kappa-\gamma/2)^2 - g^2]^{1/2}$.

For $\kappa \gg g \gg \gamma/2$, Eq. (6) gives

$$2\pi S(\omega) = \frac{\gamma + 2g^2/\kappa}{\frac{1}{4}(\gamma+2g^2/\kappa)^2 + (\omega-\omega_0)^2} , \tag{7a}$$

which shows the increased linewidth associated with cavity-enhanced spontaneous emission. We are interested in the strong-coupling limit, $g \gg \kappa, \gamma/2$. In this limit,

$$2\pi S(\omega) = \frac{\frac{1}{2}(\kappa+\gamma/2)}{\frac{1}{4}(\kappa+\gamma/2)^2 + (\omega-\omega_0-g)^2} + \frac{\frac{1}{2}(\kappa+\gamma/2)}{\frac{1}{4}(\kappa+\gamma/2)^2 + (\omega-\omega_0+g)^2} \tag{7b}$$

The spontaneous-emission spectrum is a doublet (Fig. 1)

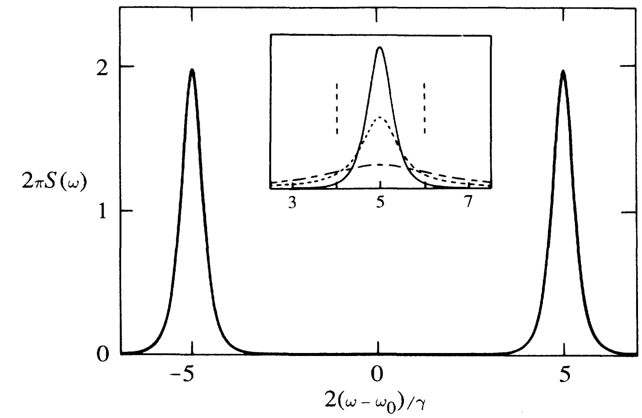

FIG. 1. (a) The spontaneous-emission spectrum $2\pi S(\omega)$ for $\kappa \ll \gamma/2$ and $2g/\gamma = 5$. The inset compares the spectral sidebands in free-space resonance fluorescence (Ref. 6), ----, the spontaneous-emission spectrum $2\pi S(\omega)$, — — —, and the fluorescent spectrum $2\pi T(\omega)$, ——. The vertical dashed lines show the full free-space radiative width γ.

with the averaged linewidth (half-width) $\frac{1}{2}(\kappa+\gamma/2)$. This linewidth is less than the free-space radiative width $\gamma/2$ whenever $\kappa < \gamma/2$. When $\kappa \ll \gamma/2$ the linewidth is one-half the free-space radiative width.

Before we discuss the origin of the subnatural linewidth we show that the same averaged linewidth appears in the spectrum of weakly driven fluorescence. We may study intracavity resonance fluorescence in two distinct configurations—with either the atom or the cavity mode driven by a coherent field. We consider the second alternative; to describe the coherent driving of the cavity mode we add the term $i\hbar\mathcal{E}(\hat{a}^\dagger - \hat{a})$ to the Hamiltonian in Eq. (1), where $\mathcal{E}$ is the (real) amplitude of the driving field. Then, if $g \ll \kappa, \gamma/2$, the driven cavity fills with the coherent state $|\mathcal{E}/\kappa\rangle$. For $\bar{n} = (\mathcal{E}/\kappa)^2 \gg 1$ the variation of the Rabi frequency across the Poisson photon-number distribution for this state is $2(\bar{n}+\bar{n}^{1/2})^{1/2}g - 2(\bar{n}-\bar{n}^{1/2})^{1/2}g \approx 2g$. The theory of free-space resonance fluorescence assumes that this variation is negligible. This theory is a weak-coupling theory—strictly, it takes the limit $g/\gamma \to 0, \bar{n} = (\mathcal{E}/\kappa)^2 \to \infty$, with $g\sqrt{\bar{n}}/\gamma$ constant. For intracavity resonance fluorescence in the strong-coupling limit, the variation of the Rabi frequency across the photon-number distribution of the field is not negligible and we can expect to see substantial departures from the results of the standard weak-coupling theory.

A calculation of fluorescent spectra for arbitrary excitation strengths only seems feasible numerically. However, we can calculate spectra for weakly driven fluorescence analytically. This calculation is still considerably more complicated than the calculation of the spontaneous-emission spectrum, since we must include more basis states than the three used to derive Eq. (6). We have calculated spectra for arbitrary values of g, κ and $\gamma/2$. Here we only present results for the strong-coupling limit where linewidth averaging is seen. For $g \gg \kappa, \gamma/2$, the inelastic part of the fluorescent spectrum is given by

$$2\pi T(\omega)=\frac{\frac{1}{4}(\kappa+\gamma/2)^3}{[\frac{1}{4}(\kappa+\gamma/2)^2+(\omega-\omega_0-g)^2]^2}+\frac{\frac{1}{4}(\kappa+\gamma/2)^3}{[\frac{1}{4}(\kappa+\gamma/2)^2+(\omega-\omega_0+g)^2]^2}\ . \quad (8)$$

On comparing Eq. (8) with Eq. (7b) a notable new feature is present; the peaks in the fluorescent spectrum are squared Lorentzians, which show an additional 36% narrowing relative to the Lorentzians in Eq. (7b). For $\kappa \ll \gamma/2$, this narrowing, together with linewidth averaging, results in spectral peaks that have approximately one-third the free-space radiative width (Fig. 1). The square appears because the fluorescence is squeezed. More precisely, scattering from the atomic dipole contributes two Lorentzian doublets that are added together to obtain the full inelastic spectrum—one doublet is contributed by the scattering from each of the two quadrature phase amplitudes of the induced atomic dipole. The quantum fluctuations *in phase* with the mean induced atomic dipole are squeezed; therefore the Lorentzian doublet they contribute has negative weight. The resulting subtraction of Lorentzians in the full spectrum gives rise to the squares in Eq. (8). A similar effect occurs for weakly driven fluorescence in free space, but without line splitting and linewidth averaging.[6,7]

Now let us discuss the origin of the linewidth average that appears in Eqs. (7b) and (8). We offer two complimentary views of the underlying physics. The first is provided by an alternative derivation of Eq. (7b) using the coupled system eigenstates [eigenstates of $\hat{H}'=\hbar\omega_0(\hat{\sigma}_z+\hat{a}^\dagger\hat{a})+\hat{H}$]

$$|l\rangle=(1/\sqrt{2})(|+\rangle|0\rangle-i|-\rangle|1\rangle)\ , \quad (9a)$$

$$|u\rangle=(1/\sqrt{2})(|+\rangle|0\rangle+i|-\rangle|1\rangle)\ . \quad (9b)$$

The eigenvalues $\hbar(\omega_0-g)$ and $\hbar(\omega_0+g)$ associated with these states identify the central frequencies of the Lorentzians in Eq. (7b). If we adopt the three-state basis $|l\rangle, |u\rangle, |-\rangle|0\rangle$, the atomic dipole operator takes the form $\hat{\sigma}_-=(1/\sqrt{2})(\hat{l}_-+\hat{u}_-)$, and the cavity-mode annihilation operator takes the form $\hat{a}=(-i/\sqrt{2})(\hat{l}_--\hat{u}_-)$, where $\hat{l}_-=|-\rangle|0\rangle\langle l|$ and $\hat{u}_-=|-\rangle|0\rangle\langle u|$. In the strong-coupling limit Eq. (1) can then be written as

$$\dot{\tilde{\rho}}=\tfrac{1}{2}(\kappa+\gamma/2)(2\hat{l}_-\tilde{\rho}\hat{l}_+-\hat{l}_+\hat{l}_-\tilde{\rho}-\tilde{\rho}\hat{l}_+\hat{l}_-)+\tfrac{1}{2}(\kappa+\gamma/2)(2\hat{u}_-\tilde{\rho}\hat{u}_+-\hat{u}_+\hat{u}_-\tilde{\rho}-\tilde{\rho}\hat{u}_+\hat{u}_-)\ , \quad (10)$$

where $\tilde{\rho}=\exp(-i\hat{H}t/\hbar)\rho\exp(i\hat{H}t/\hbar)$, and $\hat{l}_+$ and $\hat{u}_+$ are the Hermitian conjugates of $\hat{l}_-$ and $\hat{u}_-$, respectively; we have dropped the cross terms $\hat{l}_+\hat{u}_-$ and $\hat{u}_+\hat{l}_-$ which oscillate as $\exp(2igt)$ and $\exp(-2igt)$. The initial state $|+\rangle|0\rangle$ is a superposition of the eigenstates $|l\rangle$ and $|u\rangle$, and Eq. (10) describes an independent decay at the rate $\kappa+\gamma/2$ from each of these states. Thus, for the initial state $|+\rangle|0\rangle$, Eq. (10) leads immediately to the spontaneous-emission spectrum given by Eq. (7b).

Note that the state $|-\rangle|0\rangle$ reached by the spontaneous transitions from $|l\rangle$ and $|u\rangle$ is not split by the atom-cavity-mode coupling. For this reason the process described here differs from the spontaneous-emission cascade down the ladder of dressed states used to explain the strong-field resonance fluorescence spectrum for an atom in free space.[8] Rabi sidebands in free-space resonance fluorescence do not have subnatural linewidths. We obtain subnatural linewidths because the doublets in Eqs. (7b) and (8) result from *single-quantum* Rabi splitting. We will return to the comparison with free-space resonance fluorescence shortly.

A second explanation of linewidth averaging follows from the coupled equations 2(a) and 2(b). Formally, these equations describe the decay of coupled harmonic-oscillator amplitudes. If we prepare the atom-cavity-mode system in the superposition state $c|-\rangle|0\rangle+(\alpha_0/c)|-\rangle|1\rangle+(\beta_0/c)|+\rangle|0\rangle$ (where c, α_0, and β_0 are real constants), Eqs. (2a) and (2b) describe the decay of the mean-field and polarization amplitudes. In the strong-coupling limit the mean "energy" $E=\langle\hat{a}\rangle^2+\langle\hat{\sigma}_-\rangle^2$ oscillates between the polarization and the field as it decays. For $\kappa=0$ and initial conditions $\langle\hat{a}\rangle=\alpha_0=0, \langle\hat{\sigma}_-\rangle=\beta_0$,

$$E=\beta_0^2\exp[-(\gamma/2)t][1-(\gamma/4g)\sin(2gt)]\ . \quad (11)$$

E decays at the averaged rate $\frac{1}{2}(2\kappa+\gamma)=\gamma/2$. The reason for the average is revealed by the decay rate $-\dot{E}/E=(\gamma/2)[1+\cos(2gt)]$. This oscillates between a maximum value of γ and a minimum value of 0 ($=2\kappa$). When $-\dot{E}/E=\gamma$ the energy resides entirely in the *damped* polarization oscillator, and when $-\dot{E}/E=0$ the energy resides entirely in the *undamped* field oscillator. Thus, the decay rate is averaged as the energy oscillates between the polarization and the field.

With the replacements $\langle\hat{\sigma}_-\rangle\to\langle\hat{J}_-\rangle/\sqrt{N}$ and $g\to\sqrt{N}g$, Eqs. (2) describe the decay of coupled-field and collective-polarization amplitudes in a cavity containing N two-level atoms.[9,10] The subnatural linewidths we find for a single atom should therefore also occur in a system of N atoms. Indeed, using Ref. 9 we have calculated the inelastic spectrum for the light transmitted by a coherently driven cavity containing N two-level atoms. In the limit of weak excitation and strong coupling this spectrum has the same form as Eq. (8) (with $\sqrt{N}g$ in the place of g). The strong-coupling requirement, $\sqrt{N}g \gg \kappa, \gamma/2$, for N atoms, is less restrictive than for a single atom. It can be met in the optical domain (together with $\kappa \ll \gamma/2$) using a high finesse cavity ($\sim 10^4$) and a few hundred atoms. Under these conditions we have recently observed subnatural linewidth averaging.[11]

Linewidth averaging for coupled oscillator amplitudes is a common phenomenon; mathematically, it follows from the eigenvalues $\lambda_\pm$ appearing in Eq. (6) whose form is generic to any 2×2 real antisymmetric matrix. However, we know of no other situation in which this averaging leads to subnatural radiative linewidths. In particular, although linewidth averaging occurs in the strong-driving-field limit of free-space resonance fluorescence, in this context it does not lead to subnatural linewidths. (Stroud's one-photon approximation gives a spectrum with subnatural linewidths;[12] but the peak widths and heights obtained in the one-photon approximation are incorrect.)

To emphasize the difference between the linewidths we obtain and those in free-space resonance fluorescence, we recover the standard theory of free-space resonance fluorescence from our model in the weak-coupling, high excitation limit ($g/\gamma \to 0, \bar{n} \to \infty, g\sqrt{\bar{n}}/\gamma$ constant). In this limit we may trace over the quantum state of the field and write Bloch equations for an atom driven by a classical field amplitude. Let $\hat{\sigma}_\nu = \langle \hat{\sigma}_\nu \rangle_{ss} + \hat{s}_\nu$, where $\langle \hat{\sigma}_\nu \rangle_{ss}$, $\nu = x,y,z$, are the steady-state Bloch vector components. Then the central peak of the Mollow triplet is produced by the fluctuations $\hat{s}_y$. The Rabi sidebands are produced by the fluctuations $\hat{s}_x$, which satisfy coupled equations

$$\langle \dot{\hat{s}}_x \rangle = g\sqrt{\bar{n}}\,\langle \hat{s}_z \rangle - (\gamma/2)\langle \hat{s}_x \rangle \;, \qquad (12a)$$

$$\langle \dot{\hat{s}}_z \rangle = -4g\sqrt{\bar{n}}\,\langle \hat{s}_x \rangle - \gamma \langle \hat{s}_z \rangle \;. \qquad (12b)$$

Equations (12a) and (12b) have the same form as Eqs. (2). Formally they also describe coupled oscillator amplitudes. But they describe an oscillation between the atomic polarization and inversion, not between the polarization and the field. Indeed, the field does not even enter the equations as a dynamical variable. This is because in the weak-coupling, high excitation limit the oscillatory exchange of a single quantum of energy with the field has negligible effect on the atomic dynamics; therefore a *constant* field amplitude can be used in the Bloch equations. The width of the Rabi sidebands in free-space resonance fluorescence is certainly determined by an averaging of decay rates. But the polarization decay rate $\gamma/2$ is averaged against the inversion decay rate γ, rather than against κ. This gives a linewidth $\frac{1}{2}(\gamma+\gamma/2) = 3\gamma/4$ (Fig. 1). There is no possibility for obtaining subnatural linewidths, as there is in the strongly coupled system realized using an optical cavity.

In conclusion, we have shown that the interaction of an atom and a resonant cavity mode can produce subnatural linewidths by linewidth averaging. The same linewidth averaging occurs when a collective atomic polarization couples to a resonant cavity mode. In this system subnatural linewidths have recently been observed.

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