Resonance fluorescence spectrum in a weak squeezed field with an arbitrary bandwidth

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We analyze the linewidth narrowing in the fluorescence spectrum of a two-level atom driven by a squeezed vacuum field of a finite bandwidth. It is found that the fluorescence spectrum in a low-intensity squeezed field can exhibit a $(\omega-\omega_0)^{-6}$ frequency dependence in the wings. We show that this fast fall-off behavior is intimately related to the properties of a narrow-bandwidth squeezed field and does not extend into the region of broadband excitation. We apply the linear response model and find that the narrowing results from a convolution of the atom response with the spectrum of the incident field. On the experimental side, we emphasize that the linewidth narrowing is not sensitive to the solid angle of the squeezed modes coupled to the atom. We also compare the fluorescence spectrum with the quadrature-noise spectrum and find that the fluorescence spectrum for an off-resonance excitation does not reveal the noise spectrum. We show that this difference arises from the competing three-photon scattering processes. [S1050-2947(98)04308-X]

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The interaction of squeezed light with atoms has attracted considerable attention in recent years [1]. Many unusual and interesting effects have been predicted exhibiting the basic differences which occur when atoms interact with squeezed light rather than with the ordinary vacuum. The most significant of these predictions was the line narrowing in the fluorescence and absorption spectra [2–4]. This effect has been predicted by Gardiner [2] in the context of spontaneous emission. He showed that, compared to the normal atomic vacuum-decay rate, in a squeezed vacuum the two quadratures of the atomic polarization are damped at different rates, one at an enhanced rate and the other at the reduced rate. This prediction, however, has been derived under the assumption that the squeezed field appears as an infinite-bandwidth (Markovian) reservoir to the atom.

The assumption that the squeezed vacuum has an infinite bandwidth is, of course, an approximation. The squeezed light generated in the present experiments has a finite rather than an infinite bandwidth and, in fact, exhibits bandwidths only of the order of typical atomic linewidths [5]. Recognizing this, several authors have studied the influence of the finite bandwidth on the squeezing effects without applying the Markov approximation [6,7]. It has been demonstrated via numerical simulation that subnatural linewidths predicted for a strong infinite-bandwidth squeezed field can be observed for any bandwidth of the squeezed field, but are diminished and ultimately lost for bandwidths comparable to the atomic linewidth.

In this Brief Report we show that the spectral narrowing with a low-intensity squeezed field exhibits significant quantitative differences in behavior in comparison with the narrowing by a strong squeezed field. Previous theoretical studies of the fluorescence and absorption spectra [3,4] have shown that a large narrowing is possible for a strong broadband squeezed field and the potential of the narrowing decreases with the decreasing bandwidth of the squeezed field [6,7]. We show that a low-intensity squeezed field of bandwidths comparable to the atomic linewidth narrows the spectral line below any bandwidth involved in the process and this effect does not extend into the region of broadband excitation. We explain the origin of the line narrowing as arising from the convolution of the atomic response with the spectrum of the incident field, which is characteristic of the linear response model. With respect to experimental tests of the line narrowing, we find that the feature is not sensitive to the solid angle of the squeezed modes coupled to the atom. The coupling affects only the intensity of the fluorescence field. We further calculate the quadrature-noise spectrum of the fluorescence field and find that for an off-resonance excitation the fluorescence spectrum does not reveal the quadrature-noise spectrum.

We consider a single two-level atom with the excited state $|2\rangle$ and the ground state $|1\rangle$, separated by the transition frequency ω_0, and coupled by the electric transition dipole moment $\vec{d}$. We assume that the atom is driven by a squeezed vacuum field of the carrier frequency ω_L and is also coupled to the remaining modes of the three-dimensional electromagnetic field, which are initially in the vacuum state.

The dynamics of the driven two-level atom is described by the time evolution of the slowly varying part $\hat{b}(t)$ of the atomic lowering operator $\hat{\sigma}^-=|1\rangle\langle 2|$, which in the low-intensity limit of the driving field obeys the equation [8–11]

$$\hat{b}(t)=\hat{b}(0)e^{(-\beta+i\Delta)t}-\alpha\int_0^t dt_1\hat{A}_s^+(t_1)e^{(\beta-i\Delta)(t_1-t)}, \quad (1)$$

where

$$\hat{b}(t)=\hat{\sigma}^-(t)e^{i\omega_L t}, \quad \hat{A}_s^+(t)=\hat{A}_{\rm free}^+(t)e^{i\omega_L t} \quad (2)$$

are the slowly varying parts of the atomic lowering operator and the positive frequency part of the vector potential operator for the driving field, respectively, β is one-half the natu-

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ral decay rate of the atom, $\Delta=\omega_L-\omega_0$ is the detuning of the field frequency from the atomic resonance, and $\alpha=|\vec{d}|\omega_0/\hbar$.

The low-intensity model considered here is known as the linear response model [10–12] as the evolution of the atomic dipole moment depends linearly on the amplitude of the incident field. Raymer and Cooper [10] have shown that in this model the steady-state fluorescence spectrum, which is given by the Fourier transform of the two-time correlation function $\langle\hat{b}^+(t)\hat{b}(t+\tau)\rangle$, can be written as a simple convolution

$$S_f(\omega)=u(r)\lim_{t\to\infty}\int_{-\infty}^{\infty}d\omega' S_A(\omega,\omega')N(\omega'), \tag{3}$$

where $u(r)$ is a constant containing geometrical factors [9],

$$S_A(\omega,\omega')=\int_{-\infty}^{\infty}d\tau\, e^{i\omega\tau}\int_0^t dt_1 e^{(\beta+i\Delta)(t_1-t)} \times\int_0^{t+\tau}dt_2 e^{(\beta-i\Delta)(t_2-t-\tau)}e^{i\omega'(t_1-t_2)} \tag{4}$$

is the spectrum of the atom response, and

$$N(\omega')=\frac{\alpha^2}{2\pi}\int_{-\infty}^{\infty}d\tau\, e^{i(\omega'-\omega_L)\tau}\langle\hat{A}_s^+(t)\hat{A}_s(t+\tau)\rangle \tag{5}$$

is the intensity spectrum of the driving field.

The field-correlation function, which appears in Eq. (5), depends on the statistics of the driving field. Our source of the driving field is taken to be a degenerate parametric oscillator (DPO) operating below threshold [13]. The intensity spectrum of the output of the DPO is given by

$$N(\omega')=\frac{1}{2}\beta\eta\left[\frac{(N+M)\mu^2}{\mu^2+(\omega'-\omega_L)^2}+\frac{(N-M)\lambda^2}{\lambda^2+(\omega'-\omega_L)^2}\right], \tag{6}$$

where $\mu=\frac{1}{2}\gamma_c-\epsilon$, $\lambda=\frac{1}{2}\gamma_c+\epsilon$, γ_c is the damping constant of the DPO, ϵ is its amplification parameter (proportional to the amplitude of the classical pump field of the DPO cavity), and

$$N-M=-\frac{\lambda^2-\mu^2}{2\lambda^2},\quad N+M=\frac{\lambda^2-\mu^2}{2\mu^2}, \tag{7}$$

with $M=\sqrt{N(N+1)}$. The parameters μ and λ determine the bandwidth of the squeezed field. In the broadband case both μ and λ are much larger than any decay rates occurring in the atomic dynamics. In the narrow-bandwidth case both μ and λ are smaller than or comparable to the atomic linewidth. The factor η, important from the experimental point of view, determines the matching of the input squeezed modes to the vacuum modes coupled to the atom. For perfect matching, when all modes coupled to the atom are squeezed, $\eta=1$, whereas $\eta<1$ for imperfect matching.

We note here that, in contrast to a strong driving squeezed field, the fluorescence spectrum (3) is not sensitive to the two-photon correlations $M(\tau)=\langle\hat{A}(t)\hat{A}(t+\tau)\rangle$ characteristic of the squeezed vacuum field [13]. Despite this the fluorescence spectrum is sensitive to squeezing as the intensity spectrum (6) exhibits the $(\omega-\omega_0)^{-4}$ frequency dependence in the wings, which is intimately related to the properties of the DPO as a source of squeezed light. The $(\omega-\omega_0)^{-4}$ dependence in the wings is due to the negative weight of the second term in Eq. (6).

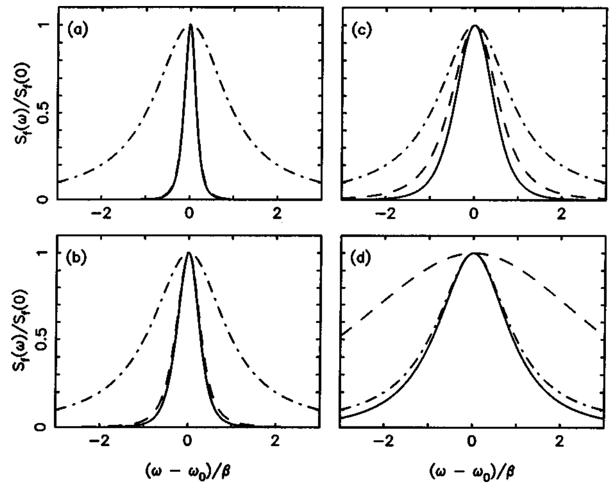

FIG. 1. Fluorescence spectrum, normalized to one at $\omega-\omega_0=0$, for $\Delta=0$, $N=0.125$, $M=0.375$, and different γ_c: (a) $\gamma_c=\frac{1}{2}\beta$, (b) $\gamma_c=\beta$, (c) $\gamma_c=2\beta$, (d) $\gamma_c=10\beta$. The solid line is the normalized fluorescence spectrum $S_f(\omega)/S_f(0)$, the dashed line is the normalized spectrum $N(\omega)/N(0)$ of the incident field, and the dashed-dotted line is the Lorentzian $\beta^2[\beta^2+(\omega-\omega_0)^2]^{-1}$.

Performing the integrals in Eq. (4), we find that the fluorescence spectrum (3) can be written as

$$S_f(\omega)=4u(r)\frac{\beta}{\beta^2+(\nu+\Delta)^2}N(\nu), \tag{8}$$

where $\nu=\omega-\omega_L$.

The result (8) is valid for arbitrary detunings Δ and bandwidths of the incident squeezed field and shows that at low intensities the fluorescence spectrum is the product of the atom response (Lorentzian of the bandwidth β) with the spectrum of the incident field. However, the result (8) is not valid for more intense saturating driving fields, as the response of the atom is then nonlinear and depends not only on the intensity spectrum $N(\omega)$ but also on the spectrum of the two-photon correlations $M(\tau)$.

The spectrum (8) contains two terms, a Lorentzian of the width β, centered on ω_0, corresponding to inelastic scattering of the driving field and a spectral line, centered on ω_L, corresponding to elastic scattering of the driving field. In the limit of narrow-band excitation ($\mu,\lambda\ll\beta$) the coherent scattering dominates over the incoherent, and then the spectral line reduces to that of the driving field. For a broadband excitation ($\mu,\lambda\gg\beta$) the coherent scattering is negligible compared with the incoherent scattering and then the spectral width tends to the atomic linewidth β. For bandwidths comparable to the atomic linewidth ($\mu,\lambda\approx\beta$) the product of the coherent and the incoherent fields can lead to narrowing of the spectral line below both: the atomic and the incident squeezed field linewidths. This is shown in Fig. 1, where we plot the fluorescence spectrum for $\Delta=0$, $N=0.125$, $M=0.375$ (50% squeezing), and different bandwidths γ_c of the DPO cavity. In order to see the linewidth narrowing we compare the fluorescence spectrum with the Lorentzian of the bandwidth β and with the spectrum $N(\nu)$ of the incident field. It is evident from Fig. 1 that for the cavity bandwidths comparable to the atomic linewidth the fluorescence spectrum is narrower than any bandwidth involved in the process.

The convolution (3) is characteristic of the linear response model [10,11] and can lead to narrowing of the spectral line independent of the shape and statistics of the driving field. For a Lorentzian shape of the driving field the convolution can produce a narrow spectrum which falls off as $(\omega-\omega_0)^{-4}$ in the wings. The $(\omega-\omega_0)^{-4}$ dependence in the wings has also been found in the fluorescence spectrum of a two-level atom driven by a weak coherent laser field [14]. In the case considered here, the spectrum falls off as $(\omega-\omega_0)^{-6}$ in the wings. This strong fall-off behavior arises from the $(\omega-\omega_0)^{-4}$ dependence in the wings of the spectrum of the incident squeezed field.

It is also interesting to consider the noise properties of the *emitted* fluorescence [15] as one could expect that, in the low-intensity limit, the fluorescence spectrum should reveal the quadrature-noise spectrum. However, in terms of the quadrature components of the emitted field the fluorescence spectrum can be written as

$$S_f(\omega)=S_0(\omega)+S_{\pi/2}(\omega)+S_a(\omega), \tag{9}$$

where $S_0(\omega)$ and $S_{\pi/2}(\omega)$ are the in-phase and out-of-phase components of the quadrature-noise spectrum [15,16], respectively, and

$$S_a(\omega)=-\tfrac{1}{2}\beta u(r)\int_0^\infty d\tau\,\sin(\omega-\omega_L)\tau \times \mathrm{Im}[\langle \hat{b}^+(t),\hat{b}(t+\tau)\rangle] \tag{10}$$

is the asymmetric contribution to the spectrum. From Eqs. (1) and (10), we find that

$$\begin{aligned} S_a(\omega)=-4\beta^2\eta u(r)\Bigg\{&\frac{\Delta(N-M)\lambda^2\nu}{[(\beta+\lambda)^2+\Delta^2][(\beta-\lambda)^2+\Delta^2]}\\ &\times\left(\frac{1}{\lambda^2+\nu^2}-\frac{(2\beta^2-\lambda^2)-2\Delta^2+\nu^2}{[\beta^2+(\nu+\Delta)^2][\beta^2+(\nu-\Delta)^2]}\right)\\ &+\frac{\Delta(N+M)\mu^2\nu}{[(\beta+\mu)^2+\Delta^2][(\beta-\mu)^2+\Delta^2]}\\ &\times\left(\frac{1}{\mu^2+\nu^2}-\frac{(2\beta^2-\mu^2)-2\Delta^2+\nu^2}{[\beta^2+(\nu+\Delta)^2][\beta^2+(\nu-\Delta)^2]}\right)\Bigg\}. \end{aligned} \tag{11}$$

We see from Eq. (11) that the asymmetric part vanishes for $\Delta=0$. In this case the fluorescence spectrum is symmetric and reveals the noise spectrum of the emitted field. Otherwise, for $\Delta\neq 0$, the fluorescence spectrum is asymmetric and does not reveal the noise spectrum.

This interesting effect is shown in more detail in Fig. 2, where we plot the fluorescence, quadrature noise, and asymmetric spectra for $N=0.125$, $M=0.375$, $\Delta=5\beta$, and $\gamma_c=4\beta$. The graphs show that there are two peaks in the fluorescence spectrum. However, the noise spectrum exhibits three peaks, the central peak at the frequency ω_L, corresponding to the elastic scattering of the driving field, and two sidebands; one at $\nu=-\Delta$ $(\omega=\omega_0)$ and the other at $\nu=\Delta$ $(\omega=2\omega_L-\omega_0)$. The peak at $\nu=\Delta$ is due to a three-photon incoherent scattering of the noise of the input squeezed field. This peak is absent in the fluorescence spectrum. The "missing" peak is in fact contained in the asymmetric part of the fluorescence spectrum.

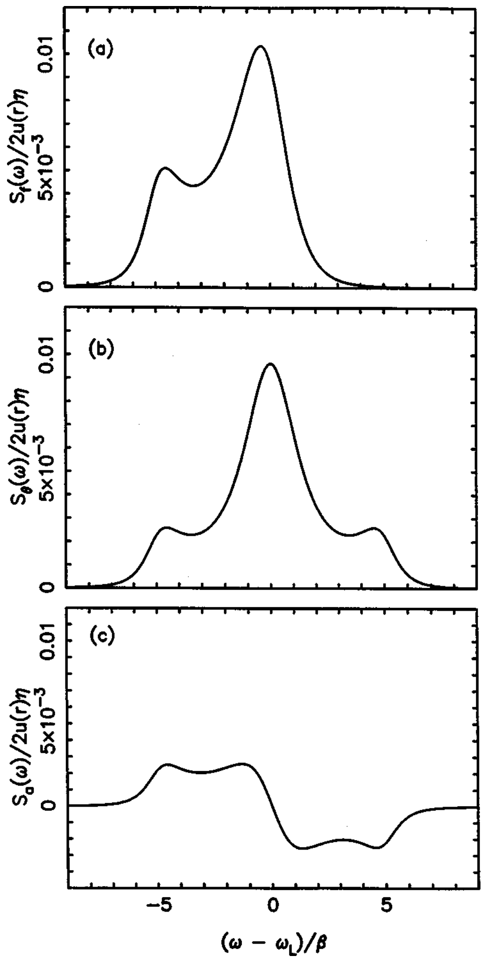

FIG. 2. (a) Fluorescence spectrum, (b) the quadrature-noise spectrum, and (c) the asymmetric contribution, for $\Delta=5\beta$, $N=0.125$, $M=0.375$, and $\gamma_c=4\beta$.

The asymmetric part of the spectrum shows a negative peak at $\nu=\Delta$ whose amplitude is exactly equal to the corresponding positive peak in the quadrature-noise spectrum. The negative peak in the asymmetric part of the fluorescence spectrum can be interpreted as arising from a three-photon coherent scattering. In this process two photons of the frequency ω_L are absorbed and one photon of the frequency $2\omega_L-\omega_0$ is emitted into the incident field.

In summary, we have calculated the fluorescence spectrum of a two-level atom driven by a weak squeezed field of an arbitrary bandwidth. We have shown that the spectrum can be narrower than any bandwidth involved in the problem, and have found that in this case the spectrum exhibits a $(\omega-\omega_0)^{-6}$ dependence in the wings which is attributed to squeezing in the incident field.

We have also calculated the quadrature-noise spectrum of the fluorescence field and have shown that for an off-resonance excitation the spectrum can exhibit three peaks.

The central peak, located at the frequency ω_L, and two sidebands; one located at the atomic transition frequency ω_0, and the other located at the frequency $2\omega_L-\omega_0$ corresponding to the three-photon incoherent scattering process. The fluorescence spectrum, however, does not exhibit the presence of the three-photon scattering. We have shown that the asymmetric part of the spectrum exhibits a negative peak at the frequency $2\omega_L-\omega_0$, which exactly compensates the positive peak of the quadrature-noise spectrum.

Finally, we would like to point out that the system considered here is realistic experimentally. It is valid for an arbitrary bandwidth of the squeezed field, and does not require squeezing of all modes to which the atom is coupled. Moreover, this model is valid for weak squeezed fields, which are now available in experiments [5].

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