

Tunable two-mode and single-mode squeezing in resonance fluorescence, including phase-diffusion effects

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The quadrature squeezing spectra produced in the resonance fluorescence of a two-level atom are investigated, including the effects of a finite laser bandwidth due to phase diffusion. For resonant excitation at low intensities, the fluorescence field exhibits narrow bandwidth squeezing in the out-of-phase quadrature, centered at the laser frequency. Otherwise, squeezing occurs only in the in-phase quadrature. Specifically, for slightly off-resonance and weak excitation, the squeezing still centers at the laser frequency, with a finite bandwidth of the order of γ . More importantly, for far-off-resonance and strong excitation, the resonance fluorescence exhibits two-mode squeezing around the Rabi sideband frequencies. Thus we have a source of two-mode squeezing that is very easily frequency tuned simply by adjusting the generalized Rabi frequency. The presence of a laser linewidth substantially reduces the degree of squeezing obtainable. The fluorescence shows no squeezing at all when the laser linewidth is greater than the atomic spontaneous emission linewidth. [S1050-2947(99)08401-2]

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I. INTRODUCTION

Squeezing of the radiation field, one of the most fundamental effects in the quantum theory of radiation, has been extensively investigated both theoretically and experimentally [1]. Apart from the basic interest in manipulating quantum fluctuations, the study of squeezed light was directly motivated by potential applications of such sources, for example, in the fields of telecommunications, high-precision measurement, and atomic spectroscopy [1–3]. The frequency tunability of the squeezed light source is essential for its applications. This has been demonstrated in experiments by Polzik *et al.* [3], who reported enhanced sensitivity for the detection of Doppler-free resonances in saturation spectroscopy, a linear dependence of the two-photon excitation rate, and the observation of quantum correlation at ultrahigh frequencies, using a frequency tunable source of squeezed light generated by an optical parametric oscillator operating below threshold.

Both theoretical and experimental studies have shown that the resonance fluorescence of a driven atom can serve as a source of nonclassical light. For example, Carmichael and Walls, and Kimble and Mandel [4] predicted that the resonance fluorescence from a single two-level atom driven by a coherent laser field of low intensity would exhibit photon antibunching. The prediction has been confirmed in many laboratories [5,6]. The sub-Poissonian statistics of the fluorescent photons emitted in a short time interval by a single atom was also investigated experimentally [7]. Squeezing in resonance fluorescence, in terms of the total variances and fluctuation spectra of the phase quadratures, was reported as well. Walls and Zoller, and Loudon [8] showed that the total

quantum fluctuations in the phase quadratures of the resonance fluorescence of a driven two-level atom can be squeezed below the shot-noise limit. It has been suggested that these effects may be observed by employing the system of a single, trapped, laser-cooled two-level ion, as in the recent experiments of Hoffges *et al.* [6,9]. The phase-quadrature noise spectra of the resonance fluorescence of a two-level atom with *resonant* laser excitation were also studied in both the standard vacuum and the squeezed vacuum [10]. Squeezing in the out-of-phase quadrature, centered at the laser frequency and with a finite bandwidth, is predicted for low excitation intensities ($\Omega < \gamma$). It is this squeezing that results in the spectral line narrowing of the resonance fluorescence [11]. Very recently, Zhao *et al.* [12] have measured the noise spectra in different phase quadratures of the resonance fluorescence of a coherently driven two-level atom with a *long* lifetime and the squeezing is confirmed. Their observations have also shown that the phase-dependent noise spectra for off-resonance excitation, which exhibit direct manifestations of the time ordering, are particularly interesting. These nonclassical properties, squeezing, photon antibunching, and sub-Poissonian statistics, are also demonstrated in resonance fluorescence in a bichromatic field [13] and in three-level atom systems [14].

In this paper we study the phase-quadrature noise spectra in the resonance fluorescence from a two-level atom and show that the squeezing features vary significantly with the frequency and intensity of the driving field. While the properties of the squeezing spectra produced by resonant excitation have been known for a long time [8], those for off-resonance excitation have not been reported. We show here that these have interesting properties. For weak excitation, the squeezing in the resonance fluorescence occurs at the laser frequency, while the resonance fluorescence with far off-resonance and strong excitation exhibits two-mode narrow bandwidth squeezing at the Rabi sideband frequencies, which are tunable by varying the frequency and intensity of

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the driving field. Under the latter conditions, this system provides a valuable source of easily tunable, two-mode squeezed light with potential spectroscopic applications [3]. We also take account of the degradation of squeezing due to diffusion of the phase of the exciting field. The presence of a finite laser bandwidth strongly reduces the strength of the squeezing.

We consider a single two-level atom with transition frequency ω_A driven by a laser field with amplitude \mathcal{E} , frequency ω_L , and fluctuating phase $\phi(t)$. The master equation for the atomic density matrix operator ρ , in a frame rotating at the frequency ω_L , is

$$\dot{\rho} = -i[H_{AL}, \rho] + \mathcal{L}\rho, \quad (1)$$

where

$$H_{AL} = \frac{\Delta}{2}\sigma_z + \frac{\Omega}{2}[e^{-i\phi(t)}\sigma_+ + e^{i\phi(t)}\sigma_-], \quad (2)$$

$$\mathcal{L}\rho = \gamma(2\sigma_-\rho\sigma_+ - \sigma_+\sigma_-\rho - \rho\sigma_+\sigma_-), \quad (3)$$

where H_{AL} is the Hamiltonian of the coherently driven atom, $\mathcal{L}\rho$ describes the atomic spontaneous decay with the rate γ , σ_{\pm} and σ_z are the atomic upper (lower) transition and population inversion operators, respectively, $\Omega = 2|\mu_{01}\mathcal{E}|/\hbar$ is the driving Rabi frequency, and $\Delta = \omega_A - \omega_L$ is the detuning between the atomic transition and the driving laser. The fluctuating phase $\phi(t)$ of the laser may be described as

$$\phi(t) = \phi_0 + \Phi(t), \quad (4)$$

where ϕ_0 is a constant corresponding to the average value of the fluctuating phase, which is assumed to be zero for simplicity, and $\Phi(t)$ is the random part, satisfying $\langle \Phi(t) \rangle = 0$, which results in a stochastic frequency $\vartheta(t) = \dot{\Phi}(t)$, assumed to be a Gaussian random process with the properties [15–17]

$$\langle \vartheta(t) \rangle = 0, \quad \langle \vartheta(t) \vartheta(t') \rangle = L\kappa e^{-\kappa|t-t'|}, \quad (5)$$

where L is the strength of the frequency fluctuations and physically describes the effective bandwidth of the laser

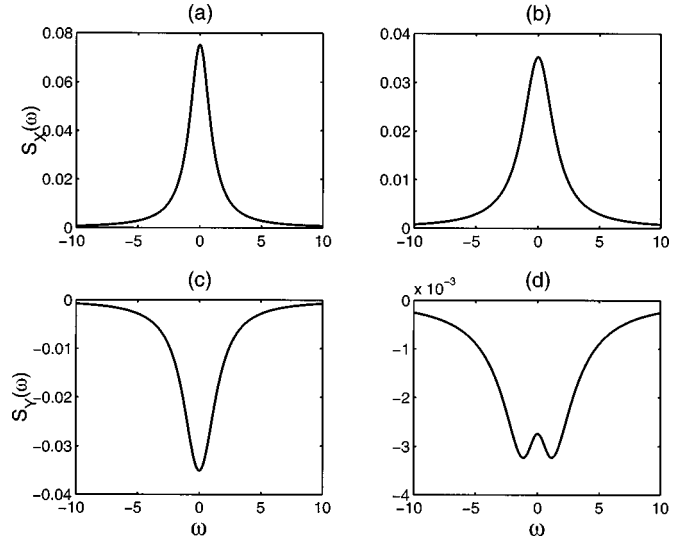


FIG. 1. Normally ordered noise spectra of the phase quadratures of resonance fluorescence for $\gamma=1$, $\Omega=0.595$, $\Delta=0$, and (a) and (c) $L=0$ and (b) and (d) $L=0.5$. (a) and (b) represent the X-quadrature noise spectrum $S_X(\omega)$, while (c) and (d) show the Y-quadrature noise spectrum $S_Y(\omega)$. [All parameters are scaled by $\gamma(=1)$ throughout these figures.]

beam due to the phase diffusion, while κ^{-1} is the time of the frequency (phase) correlation. In this paper we are interested in very short correlation times, i.e., $\kappa \gg L$, so that the correlation function (5) reduces to a δ function [15–18]

$$\langle \vartheta(t) \vartheta(t') \rangle = 2L\delta(t-t'). \quad (6)$$

This is the situation most appropriate for describing the radiation from a diode laser, which has a very stable amplitude and very large phase diffusions when operated far above threshold [18]. As shown by Osman and Swain [19], the effect of averaging over the stochastic phase may be taken into account simply by modifying the decay constants in the optical Bloch equations, which take the form [15,16]

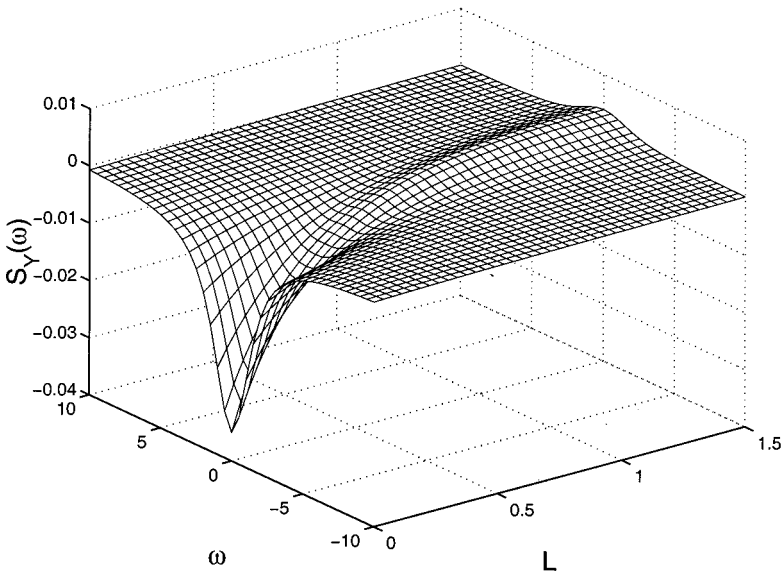


FIG. 2. Three-dimensional noise spectrum of the Y quadrature $S_Y(\omega)$ as a function of the laser linewidth L for $\gamma=1$, $\Omega=0.595$, and $\Delta=0$.

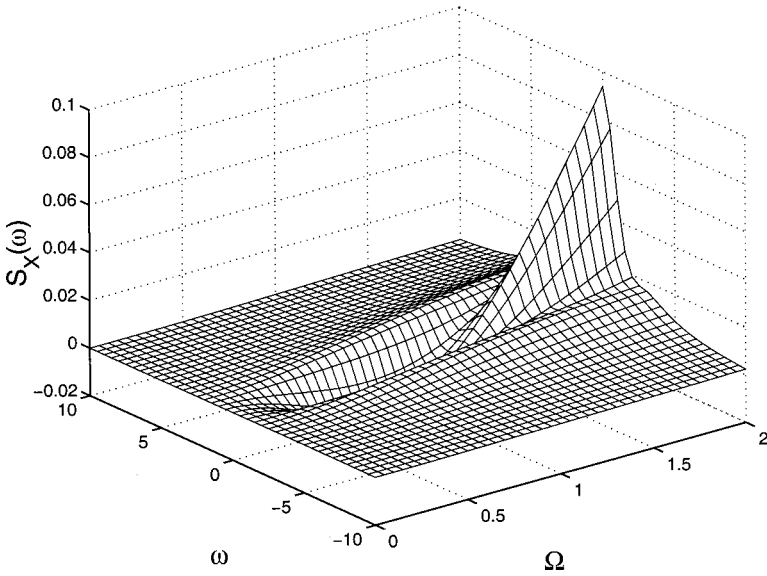


FIG. 3. Three-dimensional noise spectrum of the X quadrature $S_X(\omega)$ as a function of the Rabi frequency Ω for $\gamma=1$, $\Delta=1$, and $L=0$.

$$\begin{aligned} \langle \dot{\sigma}_+ \rangle &= -\Gamma \langle \sigma_+ \rangle - i \frac{\Omega}{2} \langle \sigma_z \rangle, \\ \langle \dot{\sigma}_- \rangle &= -\Gamma^* \langle \sigma_- \rangle + i \frac{\Omega}{2} \langle \sigma_z \rangle, \end{aligned} \quad (7)$$

$$\langle \dot{\sigma}_z \rangle = -2\gamma \langle \sigma_z \rangle + i\Omega (\langle \sigma_- \rangle - \langle \sigma_+ \rangle) - 2\gamma,$$

where $\Gamma = \gamma + L - i\Delta$.

This model has been much studied in resonance fluorescence [15–17]. Spectral broadening, sideband suppression, and asymmetry of the Mollow spectrum are predicted. Here, however, we are particularly interested in this system as a source of tunable two-mode squeezed light, which we show can be produced by large, off-resonance excitation. It is also demonstrated that it is important to take account of the finite laser linewidth (due to phase diffusion), as even quite a small laser linewidth is found to seriously degrade the squeezing obtainable. We concentrate on the squeezing spectra for off-resonance excitation, which have not been investigated previously.

II. SQUEEZING SPECTRUM

The normally ordered noise spectrum of the fluorescence field in the steady state is usually defined as [10]

$$S_{X(Y)}(\omega) = \int_{-\infty}^{\infty} d\tau \lim_{t \rightarrow \infty} \langle : \delta E_{X(Y)}(\mathbf{r}, t + \tau) \delta E_{X(Y)}(\mathbf{r}, t) : \rangle e^{i\omega\tau}, \quad (8)$$

where $\delta A = A - \langle A \rangle$ and $E_{X(Y)}(\mathbf{r}, t)$ is the slowly varying in-phase (out-of-phase) quadrature operator of the fluorescent radiation field. Using the relation between the fluorescence field and the atomic polarization operators in the far radiation zone, we may express the noise spectrum in the form [10,11]

$$\begin{aligned} S_{X(Y)}(\omega) &= \text{Re} \int_0^{\infty} d\tau \lim_{t \rightarrow \infty} \cos(\omega\tau) \\ &\quad \times [\langle \delta\sigma_+(t+\tau) \delta\sigma_-(t) \rangle \pm \langle \delta\sigma_-(t+\tau) \delta\sigma_-(t) \rangle] \\ &= \frac{1}{2} \text{Re} [\mathcal{D}_+(-i\omega) + \mathcal{D}_+(i\omega) \\ &\quad \pm \mathcal{D}_-(-i\omega) \pm \mathcal{D}_-(i\omega)], \end{aligned} \quad (9)$$

where $\mathcal{D}_{\pm}(z)$ is the Laplace transform of the two-time correlation function $\lim_{t \rightarrow \infty} \langle \delta\sigma_{\pm}(t+\tau) \delta\sigma_{\pm}(t) \rangle$. When $S_{X(Y)}(\omega) < 0$, the fluorescence light shows the quadrature squeezing. From the Bloch equation (7) one can evaluate the $\mathcal{D}_{\pm}(z)$ by invoking the quantum regression theorem to obtain

$$\mathcal{D}_+(z) = \frac{[2(\Gamma^* + z)(2\gamma + z) + \Omega^2]\chi_1 + \Omega^2\chi_2 - i\Omega(\Gamma^* + z)\chi_3}{2(\Gamma^* + z)(\Gamma + z)(2\gamma + z) + \Omega^2(\Gamma + \Gamma^* + 2z)}, \quad (10)$$

$$\mathcal{D}_-(z) = \frac{\Omega^2\chi_1 + [2(\Gamma + z)(2\gamma + z) + \Omega^2]\chi_2 + i\Omega(\Gamma + z)\chi_3}{2(\Gamma^* + z)(\Gamma + z)(2\gamma + z) + \Omega^2(\Gamma + \Gamma^* + 2z)},$$

with

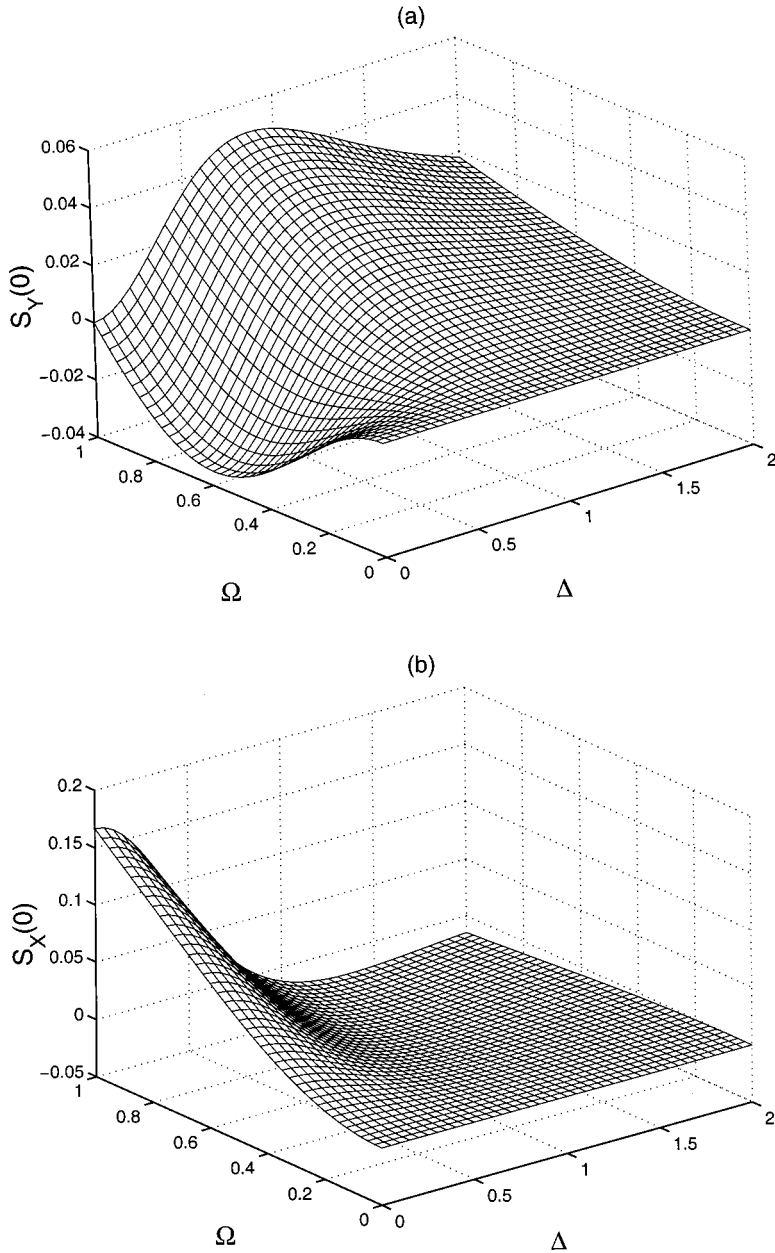


FIG. 4. Three-dimensional plots of the noise spectra at line center as a function of the Rabi frequency Ω and detuning Δ with $\gamma=1$ and $L=0$ for (a) the $S_Y(\omega=0)$ component and (b) the $S_X(\omega=0)$ component.

$$\chi_1 = \frac{1}{2}(1 + \langle \sigma_z \rangle_s) - |\langle \sigma_- \rangle_s|^2, \quad (11)$$

$$\chi_2 = -\langle \sigma_- \rangle_s^2, \quad \chi_3 = -(1 + \langle \sigma_z \rangle_s)\langle \sigma_- \rangle_s,$$

where $\langle \sigma_- \rangle_s$ and $\langle \sigma_z \rangle_s$ are the steady-state solutions of the Bloch equation (7).

We first consider the noise spectra of the phase quadratures of the resonance fluorescence from the atom with resonant excitation of a laser at low intensities, as shown in Fig. 1, where $\gamma=1$, $\Delta=0$, $\Omega=0.595$, and $L=0$ [Figs. 1(a) and 1(c)] and $L=0.5$ [Figs. 1(b) and 1(d)], respectively. For resonantly monochromatic excitation ($\Delta=0$ and $L=0$), our predictions are the same as those of Collett *et al.* [10], that is, the squeezing occurs only for low laser intensities and only in the Y quadrature, at the cost of the noise in the X quadrature. Specifically, the squeezing takes place at the laser (atomic transition) frequency and has a finite bandwidth. (Note that the parameter γ in the present paper is defined as

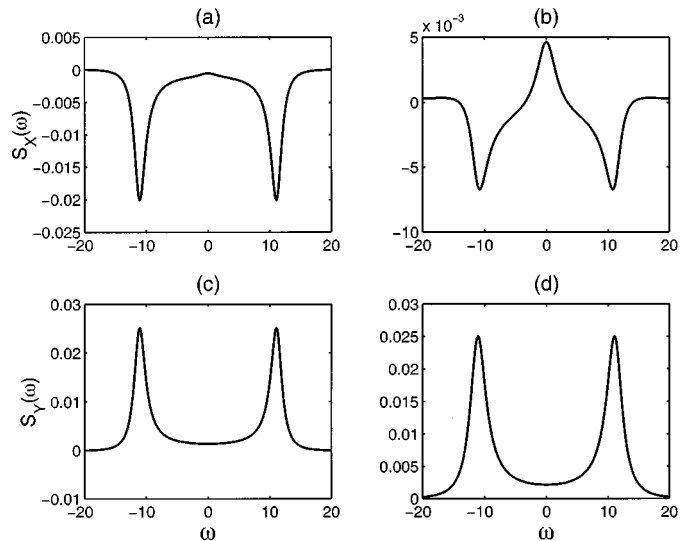


FIG. 5. Same as Fig. 1, but with $\Omega=5$ and $\Delta=10$.

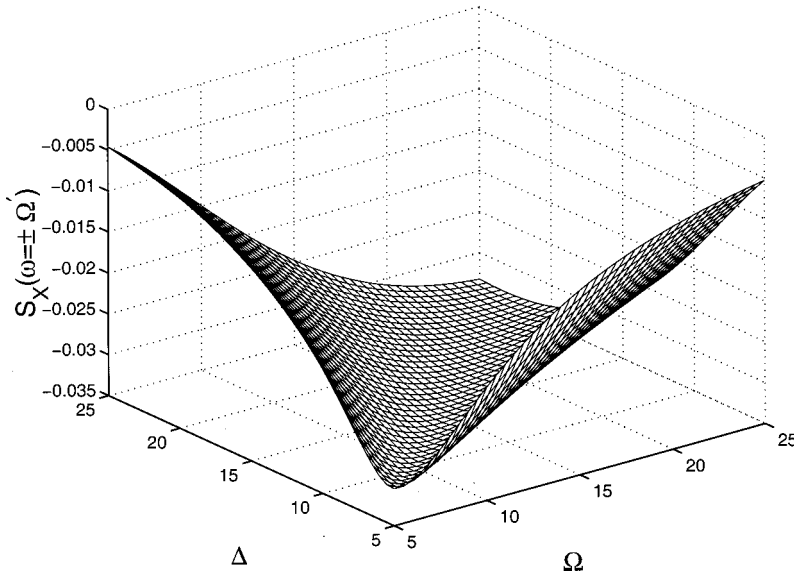


FIG. 6. Value of the X-quadrature noise spectrum at the Rabi sideband frequencies $S_X(\omega = \pm \Omega')$ against the frequency and intensity of the laser for $\gamma=1$, $L=0$, and $\Omega = \pm \Omega'$.

twice the one in [10].) If the laser linewidth is taken into account, the strength of the squeezing, however, is greatly reduced, as displayed in Fig. 1(d). When the laser linewidth is larger than the atomic natural linewidth, there will be no squeezing occurring in the resonance fluorescence. This may be seen from the expression for the Y-quadrature noise spectrum at the laser frequency, which, for $\Delta=0$, is of the form

$$S_Y(\omega=0) = \frac{2\gamma\Omega^2[\Omega^2 + \gamma(L - \gamma)]}{(\Omega^2 + 2\gamma L + 2\gamma^2)^3}. \quad (12)$$

When $L \geq \gamma$, $S_Y(\omega=0) > 0$ and no squeezing is exhibited. We present a three-dimensional graph in Fig. 2, which clearly demonstrates the reduction of the degree of the squeezing of the Y-quadrature noise spectrum due to the presence of the laser linewidth. Note the very high rate of increase of $S_Y(0)$ with L . One also finds from Eq. (12) that maximal squeezing occurs at Rabi frequency $\Omega^2 = \gamma L + 3\gamma^2 - \gamma\sqrt{3L^2 + 6\gamma L + 7\gamma^2}$. When $L=0$, the maximal squeezing [$S_Y(\omega=0) = -0.03516$] takes place at $\Omega = 0.595$.

We may also obtain squeezing in the X quadrature at low driving intensities by slightly detuning the laser frequency from the atomic transition frequency; see, for example, Fig. 3, where $L=0$ and $\Delta=1$. This is at the cost of the Y-quadrature noise. This figure exhibits a similar spectral profile to that of the Y quadrature squeezing spectrum with resonant excitation: squeezing around the laser frequency, with a finite bandwidth, and the reduced squeezing in the presence of the laser linewidth. (The latter feature is not illustrated graphically here.)

In Fig. 4 we show how the squeezing switches from the Y quadrature to the X quadrature as the detuning increases. We present only the squeezing at line center so that we may also display the dependence on the Rabi frequency. The critical value of the detuning at which the switch occurs is seen to be $\Delta \approx 0.5\gamma$.

For weak excitation, we may thus conclude that the resonance fluorescence may exhibit finite bandwidth squeezing around the laser frequency either in the Y quadrature or in the

X quadrature, depending on the laser-atom detuning. The phase diffusion of the laser substantially reduces the degree of the squeezing.

Next, we show that for strong, off-resonance excitation, the resonance fluorescence exhibits a two-mode, finite bandwidth, X-quadrature squeezing at the Rabi sideband frequencies. The central frequencies of the squeezing are thus tunable by varying the generalized Rabi frequency $\Omega' = \sqrt{\Omega^2 + \Delta^2}$. From the experimental point of view, the off-resonance, strong-field resonance fluorescence may serve as a frequency-tunable squeezed-light source for exploiting atomic spectroscopy with nonclassical light [3]. This is the main result of our paper.

Figure 5 presents both noise spectra for $\gamma=1, \Omega=5, \Delta=10$, and different laser linewidths $L=0$ and 0.5 . Evidently, the squeezing occurs only in the X quadrature, centered at the Rabi sideband frequencies $\pm \Omega'$. Correspondingly, the Y-quadrature noise spectrum has two positive peaks at the Rabi sidebands. It is also demonstrated in Fig. 4(b) that the presence of the laser linewidth reduces the strength of the squeezing.

We plot the value of the X-quadrature noise spectrum at the Rabi sideband frequencies as a function of the frequency and intensity of the laser in Fig. 6. It is apparent that the maximum squeezing takes place around $\Omega \approx \Delta$.

The dependence of the X-quadrature noise spectrum on the laser bandwidth is shown in Fig. 7, where $\gamma=1, \Omega=10$, and $\Delta=10$. Again, this figure clearly demonstrates that the presence of the laser linewidth significantly decreases the degree of the squeezing. For excitation by a broadband laser, the atomic fluorescence exhibits no squeezing at all.

For high Rabi frequencies $\Omega' \gg \gamma, L$, it is convenient to work in the basis of the semiclassical dressed states $|\pm\rangle$, defined by the eigenvalue equation $H_A|\pm\rangle = \pm(\Omega'/2)|\pm\rangle$, which are associated with the bare atomic states $|0\rangle, |1\rangle$ through the expressions

$$|+\rangle = s|0\rangle + c|1\rangle, \quad |-\rangle = c|0\rangle - s|1\rangle, \quad (13)$$

where

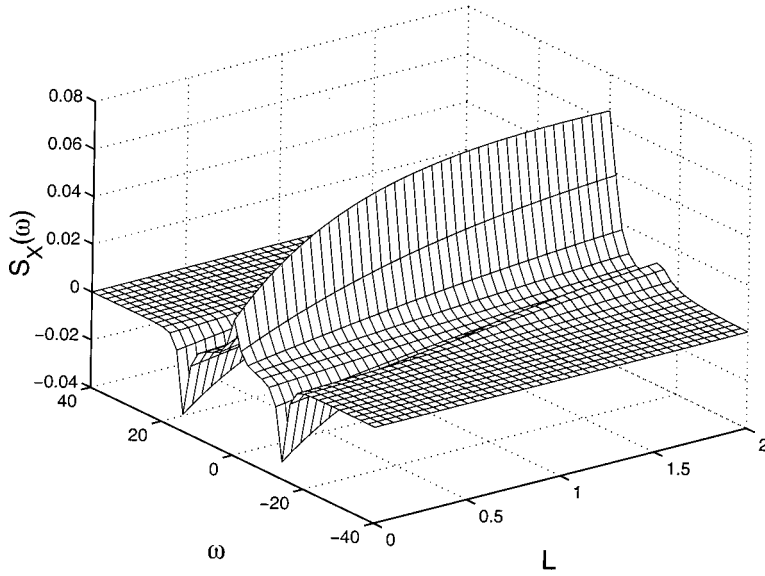


FIG. 7. Three-dimensional noise spectrum of the X quadrature $S_X(\omega)$ as a function of the laser linewidth L for $\gamma=1$, $\Omega=10$, and $\Delta=10$.

$$c = \sqrt{\frac{\Omega' + \Delta}{2\Omega'}}, \quad s = \sqrt{\frac{\Omega' - \Delta}{2\Omega'}}, \quad \Omega' = \sqrt{\Omega^2 + \Delta^2}. \quad (14)$$

Under the secular approximation [20], the quadrature noise spectra take the simple forms

$$S_X(\omega) = \frac{8c^2s^2\Gamma_{\parallel}\mathcal{P}_-\mathcal{P}_+}{\Gamma_{\parallel}^2 + \omega^2} + \frac{(c^2 - s^2)\Gamma_{\perp}}{2} \times \left[\frac{c^2\mathcal{P}_+ - s^2\mathcal{P}_-}{\Gamma_{\perp}^2 + (\omega + \Omega')^2} + \frac{c^2\mathcal{P}_+ - s^2\mathcal{P}_-}{\Gamma_{\perp}^2 + (\omega - \Omega')^2} \right], \quad (15)$$

$$S_Y(\omega) = \frac{\Gamma_{\perp}}{2} \left[\frac{c^2\mathcal{P}_+ + s^2\mathcal{P}_-}{\Gamma_{\perp}^2 + (\omega + \Omega')^2} + \frac{c^2\mathcal{P}_+ + s^2\mathcal{P}_-}{\Gamma_{\perp}^2 + (\omega - \Omega')^2} \right],$$

where

$$\mathcal{P}_+ = \frac{\gamma s^4 + Lc^2s^2}{\gamma(c^4 + s^4) + 2Lc^2s^2}, \quad (16)$$

$$\mathcal{P}_- = \frac{\gamma c^4 + Lc^2s^2}{\gamma(c^4 + s^4) + 2Lc^2s^2};$$

$$\Gamma_{\perp} = \gamma(1 + 2c^2s^2) + L(1 - 2c^2s^2), \quad (17)$$

$$\Gamma_{\parallel} = 2\gamma(1 - 2c^2s^2) + L4c^2s^2.$$

The \mathcal{P}_{\pm} are the steady-state populations in the dressed states $|\pm\rangle$ and $\Gamma_{\perp}, \Gamma_{\parallel}$ indicate the decay rates of the dressed-state polarization and the dressed-state population inversion, respectively.

Equation (15) clearly shows that the noise spectrum of the in-phase quadrature $S_X(\omega)$ has the same resonance structure as the resonance fluorescence spectrum, i.e., the central resonance at frequency ω_L with width $2\Gamma_{\parallel}$ and the side resonances at frequencies $\omega_L \pm \Omega'$ with width $2\Gamma_{\perp}$, whereas the resonances in the noise spectrum of the out-of-phase quadrature

$S_Y(\omega)$ occur only at the Rabi sidebands $\omega_L \pm \Omega'$. Both noise spectra $S_{X(Y)}(\omega)$ are symmetric around the laser frequency. It is not difficult to see that when the atom is resonantly driven by the laser field ($\Delta=0$), no squeezing occurs in either phase quadrature, while for a nonzero laser-atom detuning ($\Delta \neq 0$), the noise in the X quadrature may be squeezed around the Rabi sidebands. Therefore, the fluorescent field is a nonclassical two-mode squeezed field with the squeezing frequencies tunable via adjusting the generalized Rabi frequency Ω' and with a finite bandwidth $2\Gamma_{\perp}$. This is a remarkable departure from the behavior of a resonantly driven atom with low Rabi frequencies, where the squeezing occurs in the Y-quadrature component around the laser (atomic transition) frequency $\omega_L (= \omega_A)$ [10,11].

From Eq. (15) one obtains the value of $S_X(\omega)$ at the frequencies $\omega_L \pm \Omega'$ to be

$$S_X(\pm\Omega') = \frac{c^2s^2(c^2 - s^2)(L - \gamma)}{\Gamma_{\parallel}\Gamma_{\perp}}. \quad (18)$$

Obviously, $S_X(\pm\Omega')=0$ for $\Delta=0$, that is, there is no squeezing in the resonance fluorescence. However, $S_X(\pm\Omega') < 0$ if $\Delta \neq 0$ and $\gamma > L$, i.e., the resonance fluorescence shows the X-quadrature squeezing at the Rabi sideband frequencies. When the laser linewidth is nonzero, the degree of the squeezing is reduced; see, for example, Fig. 6.

III. SUMMARY

We have considered the noise spectra of the phase quadratures in the resonance fluorescence of a driven two-level atom, including the effect of finite laser linewidths. Squeezing occurs at the laser frequency for low excitation intensities. When the driving laser is resonant with the atom, the squeezing is in the out-of-phase quadrature, while it is in the in-phase quadrature when the laser is slightly detuned from the atomic transition frequency. However, for strong, off-resonance excitation, the noise spectrum of the in-phase quadrature exhibits two-mode squeezing at the Rabi sideband frequencies, at the cost of increased fluctuations in the out-of-phase quadrature. The squeezing has a finite band-

width of $2\Gamma_{\perp}$ and is frequency tunable (via adjusting the generalized Rabi frequency Ω'). It thus represents a potential source of such nonclassical light.

A nonzero laser linewidth substantially reduces the degree of the squeezing and the resonance fluorescence displays no squeezing at all when the laser bandwidth is greater than the atomic natural linewidth. Thus, in order to use this system as a practical source, it is essential to use highly phase-stabilized lasers.

We should point out that the system of an atom driven by a laser field with a finite bandwidth has some similarities to

that of an atom driven by a monochromatic laser, but undergoing phase-changing collisions. We may therefore conclude that dephasing collisions also decrease the strength of the squeezing. In other words, one has to use a monochromatic laser and to be free of atomic dephasing collisions in order to produce significant squeezing in resonance fluorescence [6].

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