Spectroscopy with Squeezed Light

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A frequency-tunable source of squeezed light is employed for spectroscopic measurements of atomic cesium. Relative to the usual quantum limit associated with vacuum-state fluctuations, enhanced sensitivity is demonstrated for the detection of Doppler-free resonances in saturation spectroscopy, with an improvement of 3.1 dB directly observed and 3.8 dB inferred after correcting for detector thermal noise.

Over the past several years the successful generation of a variety of manifestly quantum or nonclassical states of light has led to the achievement of measurement sensitivity beyond the quantum limit set by the vacuum fluctuations of the electromagnetic field [1–6]. Of particular importance both for these demonstrations and within the context of the theory of quantum measurement have been squeezed states of light, which are characterized by a phase-dependent distribution of quantum fluctuations with the variance of one quadrature component of the field dropping below that of the vacuum state [7]. Squeezed light has been employed for improvements in measurement precision beyond the vacuum-state (or shot-noise) limit in Mach-Zehnder [1] and polarization [2] interferometers and for the detection of directly encoded amplitude modulation [3].

In addition to these initial proof-of-principle demonstrations, another area of potentially broad import is that of spectroscopy with squeezed light, where it should likewise be possible to extend sensitivity beyond the usual quantum limit [8]. However, quite apart from issues of enhanced measurement capability, the interaction of squeezed light with atoms can lead to alterations of fundamental atomic radiative processes. Indeed, since the seminal work of Gardiner [9], diverse phenomena involving the atom-field interaction in the presence of the nonclassical fluctuations of a squeezed state have been analyzed theoretically, including resonance fluorescence [10], laser operation [11], and cavity quantum electrodynamics [12]. To explore the exciting possibilities suggested by these and other analyses, we have developed a source of frequency-tunable squeezed light suitable for spectroscopic applications over a broad range. In this Letter, we present results for operation around 855 nm and report observations of photocurrent noise reduction 5.0 dB below the vacuum-state level. With regard to the quantum limits to sensitivity in atomic spectroscopy, we utilize this source to achieve an improvement in sensitivity of 3.1 dB beyond the vacuum-state limit [13–15] for the detection of Doppler-free resonances in atomic cesium.

As depicted in broad outline in Fig. 1(a), our arrangement for spectroscopy with squeezed light is a conventional setup for frequency-modulated (FM) spectroscopy except that the probe beam $\hat{E}_1$ is in a squeezed state. As indicated in more detail in Fig. 1(b), the squeezed field is

generated by an optical parametric oscillator (OPO) operated below threshold to produce a squeezed vacuum state as its output through mirror M_1 [16]. The OPO consists of a four-mirror folded ring cavity containing a crystal of potassium niobate ($KNbO_3$) as the nonlinear element; the degenerate signal and idler beams at frequency ω are resonant, but the blue pump beam at 2ω is used in single pass. Since we require frequency tunability for the system, the primary source for all fields in the experiment is a tunable titanium-sapphire laser (50-kHz linewidth), with the pump field for the OPO obtained by harmonic conversion in an external cavity [17]. The laser, the doubling cavity, and the OPO are frequency locked relative to a reference interferometer [18]. By varying the length of the reference cavity, the frequency of the squeezed light can be tuned continuously over a range of 2 GHz with no observable reduction in the degree of squeezing.

An example of squeezed-state generation by our system is presented in Fig. 2, where we display the spectral density of photocurrent fluctuations $\Phi(\Omega, \theta)$ versus local os-
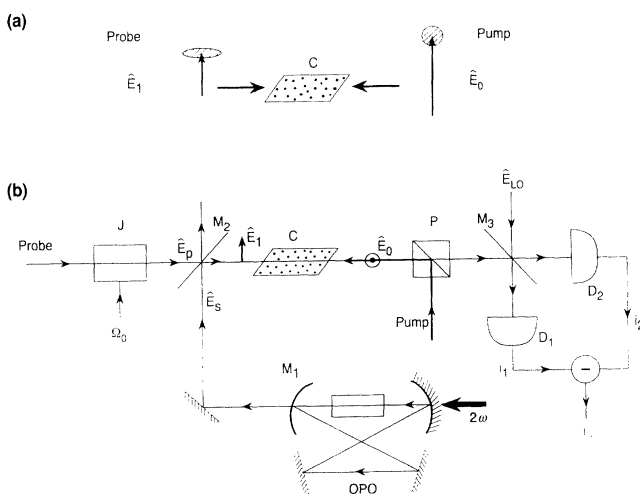

FIG. 1. (a) Saturation spectroscopy with squeezed light. The sample cell C is illuminated by the squeezed probe $\hat{E}_1$ and by a coherent pump $\hat{E}_0$, with the error circle for $\hat{E}_0$ representing vacuum-state fluctuations and the error ellipse for $\hat{E}_1$ indicating fluctuations for a squeezed state. (b) Experimental setup for FM spectroscopy with squeezed light.
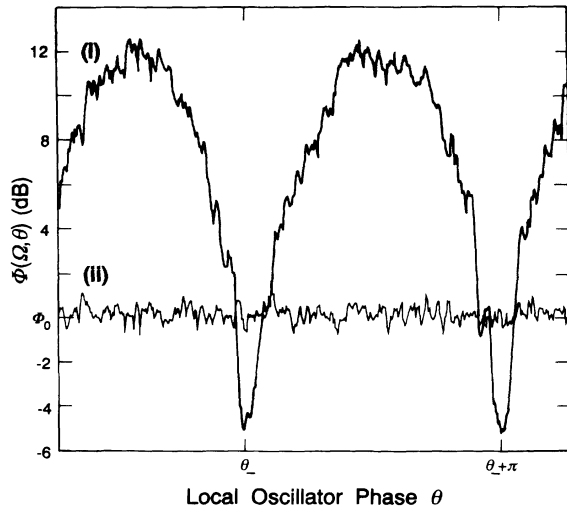
Local Oscillator Phase θ

FIG. 2. Spectral density of photocurrent fluctuations $\Phi(\Omega,\theta)$ vs local oscillator phase θ for (i) squeezed state input $\hat{E}_s$ and (ii) vacuum-state input to the balanced homodyne detector (D_1,D_2). Local oscillator power $P_{LO}=4.0$ mW; $\lambda=855$ nm; $\Omega/2\pi=2.7$ MHz; resolution bandwidth, 300 kHz; video bandwidth, 1 kHz; sweep time, 50 ms.

cillator phase θ for the difference photocurrent i_- in a balanced homodyne detector. With reference to Fig. 1(b), these measurements for the field $\hat{E}_s$ are taken without the cesium vapor cell C and polarizer P, with mirror M_2 as a high reflector in the absence of the probe beam $\hat{E}_p$, and with mirror M_3 as a 50-50 beam splitter. Relative to the "shot-noise" level indicated by Φ_0 and determined to within ± 0.2 dB [16], the photocurrent fluctuations produced by the squeezed field $\hat{E}_s$ exhibit a characteristic phase sensitivity with a directly observed noise level 5.0 ± 0.3 dB below Φ_0. When corrected for detector thermal noise, these data give a nonclassical noise reduction $\delta\Phi_- = -5.2\pm0.3$ dB below the vacuum-state limit.

A comparison of observations of i_- with theoretical expectation can be obtained by way of the spectrum of squeezing $S(\Omega,\theta)$ for an ideal OPO together with the measured losses for our system. In particular, denoting θ_- (θ_+) as the phase for minimum (maximum) fluctuations, we have that $\delta\Phi_- = \ln[1+\xi S(\Omega,\theta_-)]$, where $\xi \equiv \rho\zeta a\eta^2=0.81$ with $\rho=0.87$ as the cavity escape efficiency, $\zeta=0.965$ as the propagation efficiency from the OPO to the balanced detector, $\alpha=1.00\pm0.02$ as the quantum efficiency of the photodetectors, and $\eta=0.98$ as the homodyne efficiency for mode overlap of the squeezed field and the local oscillator. Although clearly the dominant loss mechanism is ρ, in fact the intracavity passive losses sum only to $L=3\times10^{-3}$ (exclusive of the transmission $T_1=0.105$ for the output mirror M_1), so that the passive escape efficiency $\rho_0 \equiv T_1/(T_1+L)=0.97$. However, in addition to the passive losses, we have unfortunately discovered a new loss mechanism which causes the linear absorption at ω to increase substantially in the presence

of the blue pump at 2ω. This blue-light-induced infrared absorption reduces the cavity escape efficiency to a value $\rho<\rho_0$; for example, for the data in Fig. 2, the total infrared loss for the $KNbO_3$ crystal of length 10 mm rises from 0.3% to 1.6% for a blue pump power of 0.17 W. Although the microscopic origin of this loss mechanism is unknown, we can nonetheless compare theory and experiment simply by employing the measured value of ξ together with the theoretical expression for $S(\Omega,\theta)$ [7,16]. Since the ratio r of blue pump power to threshold pump power (0.32 W) is $r \approx 0.5$, while $\Omega/2\pi=2.7$ MHz as compared to the cavity linewidth $\Gamma/2\pi=5.8$ MHz (HWHM), we find that $S(\Omega,\theta_-)=-0.90$ and hence predict a noise level $\delta\Phi_- = -5.7$ dB, which is in reasonable agreement with observation.

Turning from the characterization of our source of squeezed light to its actual implementation in atomic spectroscopy, we recall that the signal in FM saturation spectroscopy arises from imbalances in absorption and/or dispersion experienced by the probe field $\langle\hat{E}_1\rangle=A_1$ $\times\exp[-i(\omega t+\beta\sin\Omega_0 t)]$ (with $|A_1|^2\sim$photon/s), where for small modulation index β, only the Fourier components of the carrier at ω and the small FM sidebands at $\omega\pm\Omega_0$ need to be considered [14,15]. Assuming propagation through an optically thin sample with transmission factors $t(\omega)=T_0^{1/2}e^{i\phi_0}$ and $t_\pm(\omega\pm\Omega_0)$ $=T_0^{1/2}e^{-(\gamma_\pm-i\phi_\pm)}$, we find an rms signal upon photodetection given by $i_s(\Omega_0)=e\alpha T_0\beta|A_1|^2\Delta/\sqrt{2}$, where $\Delta^2\equiv(\gamma_+-\gamma_-)^2+(\phi_+-2\phi_0+\phi_-)^2$ expresses the atomic response for the fields at $(\omega,\omega\pm\Omega_0)$. For coherent-state spectroscopy the noise against which this signal must be detected is given by the shot-noise photocurrent $i_n^2(\Omega_0)$ $=2e^2\alpha T_0|A_1|^2B$, with B the detection bandwidth, leading to a signal-to-noise ratio $Q_c\equiv i_s^2/i_n^2=N\Delta^2/2$, where $N\equiv\alpha T_0\beta^2|A_1|^2/2B$. For $Q_c=1$, the minimum detectable atomic response is then $\Delta_c=2/\sqrt{N}$, which is the usual quantum limit set by the vacuum fluctuations of the field for detection of the modulus of the atomic response Δ (rather than of the individual rf quadratures for absorption and dispersion).

If we next consider the case of a squeezed probe field with $|A_1|^2\gg\int d\Omega\,S(\Omega,\theta_+)$ (i.e., small squeezed fluctuations compared to the mean field), then the principal effect associated with the presence of squeezing is to alter the noise current from i_n^2 to a value $i_s^2(\Omega_0,\theta)=i_n^2(\Omega_0)$ $\times[1+\xi'S(\Omega_0,\theta)]$, where $\xi'\equiv T_0\xi$ [8]. Since i_s is unchanged in this limit (with again $\beta\ll1$), the signal-to-noise ratio becomes $Q_{sq}(\theta)=Q_c/[1+\xi'S(\Omega_0,\theta)]$, with a corresponding minimum detectable response of Δ_{sq} $=\Delta_c[1+\xi'S(\Omega_0,\theta)]^{1/2}$. For efficient propagation and detection with $\xi\to1$ and for large squeezing with $S(\Omega_0,\theta_-)\to-1$, there is thus an enhancement in the signal-to-noise ratio $[Q_{sq}(\theta_-)/Q_c\sim(1-T_0)^{-1}\gg1]$ and a reduction in the minimum detectable atomic response $[\Delta_{sq}(\theta_-)/\Delta_c\sim(1-T_0)^{1/2}\ll1]$ [8]. On the other hand, there is a price to be paid for this enhanced sensitivity, since for a high degree of squeezing, $S(\Omega_0,\theta_+)\gg1$, and
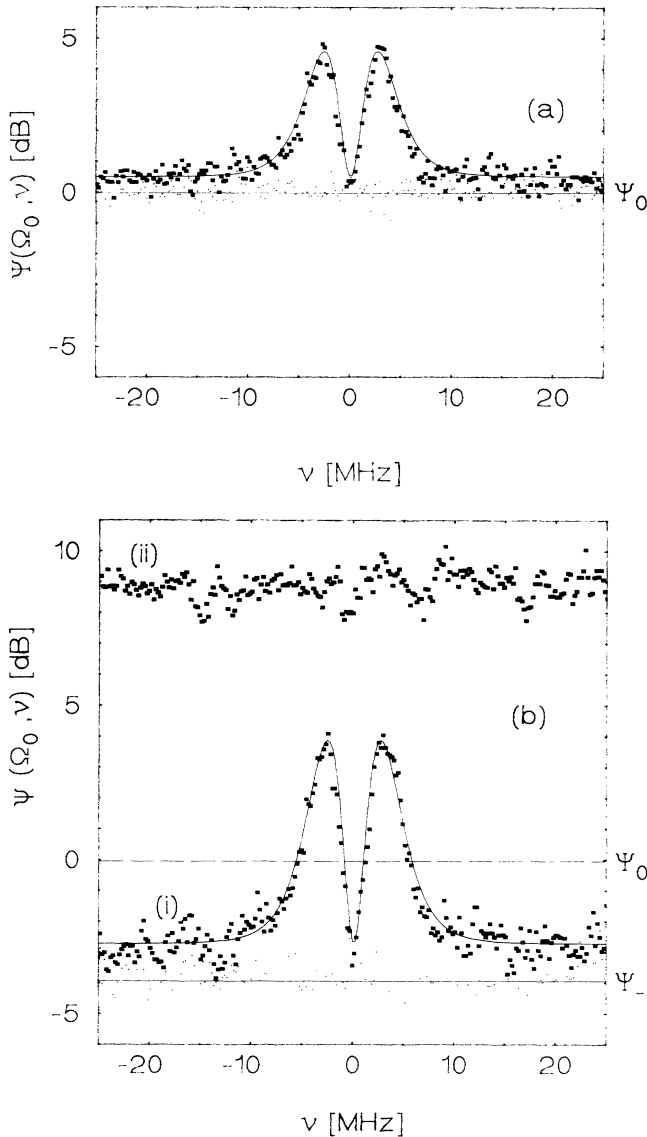
FIG. 3. Spectral density $\Psi(\Omega_0,v)$ for the probe photocurrent i_1 at Ω_0 vs laser frequency $v \equiv \omega/2\pi$ in the absence and presence of squeezed light. (a) Squeezed beam $\hat{E}_s$ is blocked to illustrate the usual vacuum-state limit for FM spectroscopy. The dots are taken with $\beta = 0$ (no modulation) and set the shot-noise level Ψ_0, while the squares are acquired with $\beta = 0.02$ and give the level of signal plus noise. (b) Squeezed beam $\hat{E}_s$ present demonstrating both an enhancement (trace i) and a degradation (trace ii) in sensitivity relative to the usual quantum limit. The dots are taken with $\beta = 0$ at the noise minimum Ψ_-, while the squares are taken with $\beta = 0.02$ with the phase offset between $(\hat{E}_s, \hat{E}_p)$ locked for minimum (i) and maximum (ii) photocurrent fluctuations. Other acquisition parameters are as follows: $\lambda = 852$ nm; $\Omega/2\pi = 2.7$ MHz; resolution bandwidth, 300 kHz; video bandwidth, 300 Hz; sweep time, 0.2 s; and residual amplitude modulation below Ψ_0 by 24 dB. In both (a) and (b) the thermal noise of the detector (which is 8.5 dB smaller than the "shot noise") has been subtracted to yield the points displayed. The full curves are from the theory of Ref. [14] as discussed in the text.

$Q_{sq}(\theta_+) \ll Q_v$.

Our experimental setup for atomic spectroscopy with squeezed light is as depicted in Fig. 1(b). A squeezed state for the probe beam $\hat{E}_1$ is created at M_2 by reflection of the squeezed-vacuum field $\hat{E}_s$ and transmission of the coherent probe $\hat{E}_p$, which has been filtered by an optical cavity of linewidth 0.5 MHz (HWHM) to reduce the excess technical noise of the Ti:Al$_2$O$_3$ laser and which has FM sidebands encoded by the modulator J. The decreased propagation efficiency for the squeezed field due to M_2 (reflectivity of 0.985) is of small consequence given the losses in the current experiment; likewise, the loss of probe power at M_2 is of little concern given the small power ($\sim 10^{-4}$ W) allowed by atomic saturation. Counterpropagating to $\hat{E}_1$ and frequency shifted by $\delta/2\pi = -40$ MHz is an orthogonally polarized pump beam $\hat{E}_0$ that creates narrow saturated resonances in the Cs velocity distribution which interact with the Fourier components of $\hat{E}_1$. The spectral line shapes $\Delta(\omega)$ for the transmitted probe beam are detected at D_1 by way of the current i_1, where now M_3 is a high reflector, $\hat{E}_{LO}$ is blocked, and D_2 is disconnected.

Focusing our attention on the particular hyperfine transition $6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F' = 5$ of the D_2 line of atomic Cs at 852 nm, we display in Fig. 3(a) a spectrum recorded with small absorption and with the squeezed field $\hat{E}_s$ blocked. This spectrum displays a characteristic "M" shape arising from the form of $\Delta(\omega)$ for a two-level transition [14,15] and represents the usual quantum limit for FM spectroscopy ($Q_v \sim 1$). By contrast, in Fig. 3(b) we present results obtained with a squeezed probe (i.e., with the squeezed output $\hat{E}_s$ of the OPO present). For the lower trace (i) the phase offset between $\hat{E}_p$ and $\hat{E}_s$ is servo controlled for minimum photocurrent noise (amplitude-squeezed probe), resulting in a drop in the noise floor and in a corresponding enhancement in signal-to-noise ratio [$Q_{sq}(\theta_-) > Q_v$]. Before subtraction of the thermal noise of the detector, the directly observed noise level lies 3.1 dB below the combined level of amplifier and shot noise; after subtraction as in Fig. 3, there is a reduction in noise of 3.8 dB below the vacuum level Ψ_0 which is determined to ± 0.1 dB. Note that in addition to enhanced sensitivity for the narrow Doppler-free signal, the broad contribution to the signal spectrum associated with the Doppler background is also more discernible. If we select instead a phase offset between $(\hat{E}_s, \hat{E}_p)$ for maximum photocurrent noise (phase-squeezed probe), then the upper trace (ii) in Fig. 3(b) shows that the spectroscopic signal is obliterated by the consequent storm tide of fluctuations from the "antisqueezed" quadrature [$Q_{sq}(\theta_+) < Q_v$]. A qualitative indication of the correspondence between the measurements and theory is provided by the full curves in Fig. 3, which are from Eq. (30) of Ref. [14] and which are drawn for parameters comparable to those in the experiment [namely, probe power $P_1 = 0.11$ mW (beam waist $w_1 = 1.5$ mm) and

pump power $P_0 = 1.5$ mW ($w_0 = 3.2$ mm) with effective saturation intensity $I_s \sim 1.7$ mW/cm^2]. Although this theory is certainly not quantitatively applicable to our data, its functional form nonetheless agrees reasonably well with the observed spectra. Note that because our reference cavity is not independently locked to the Cs resonance, the origins for the frequency axes in Fig. 3 are chosen to coincide with the line centers of the narrow spectral resonances.

There are a number of avenues by which these initial results can be improved, with the most straightforward being to increase the efficiency of the optical elements inserted for spectroscopy ($\xi' = 0.7$ for Fig. 3). Much more substantial improvements would result from the elimination of the blue-light-induced infrared absorption; indeed, without this process the passive escape efficiency ρ_0 would correspond to a loss-limited degree of squeezing 15.6 dB below the vacuum level for the output field of the OPO, while the product $\rho_0 \zeta a \eta^2$ of passive losses would imply a loss-limited noise level 10 dB below the shot-noise limit for the observed photocurrent. Apart from improved efficiencies, there is also a need to increase Γ to allow larger values of Ω_0 without suffering a degradation in noise performance due to the finite bandwidth of the squeezed field. While increases in β might likewise be desirable, our measurements with larger modulation index ($\beta = 0.10$) show smaller improvements in signal-to-noise ratio than in Fig. 3. This diminishing return for larger β is being investigated and is presumably associated with heterodyne noise arising from mixing of coherent FM sidebands with the fluctuations of the conjugate (noisy) quadrature.

In summary, we have described a frequency-tunable source of squeezed light for spectroscopic applications and have observed nonclassical reductions in photocurrent fluctuations 5.0 dB below the vacuum-state level. Improvements in measurement sensitivity of 3.1 dB beyond the usual quantum limit have been directly observed for FM spectroscopy of atomic cesium. Because of the potentially broad tunability of our source over the phase matching bandwidth of KNbO$_3$ (840–970 nm), enhanced detection sensitivity should be readily attainable for atoms and molecules other than Cs and could lead to improved capabilities beyond the shot-noise limit in a variety of spectroscopic investigations, including the detection of transient species and of overtone absorptions. However, apart from extensions of the quantum limits to precision measurement, our source should also find gainful employment in the exploration of a number of exciting problems in optical physics related to the nature of fundamental atomic radiative processes in the presence of squeezed light.

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