Single-beam squeezed-state generation in sodium vapor and its self-focusing limitations

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We describe an experiment that generates squeezed states by means of forward four-wave mixing in sodium vapor with a single optical beam. The single-beam arrangement maximizes the pump–probe spatial overlap in the nonlinear medium. Self-focusing (or self-defocusing) is found to be the major limiting factor in achieving optimal squeezing.

Recently a number of experiments have demonstrated squeezing in sodium, a two-level atomic medium that has a large nonlinearity when it is excited near resonance. Squeezed states were first generated by Slusher *et al.*[1] in an intracavity backward four-wave mixing experiment with an atomic beam of sodium. Later, Maeda *et al.*[2] observed squeezing in sodium vapor using a single-pass, forward four-wave mixing configuration, which required angle phase matching of the four interacting beams. Orozco *et al.*[3] exploited the strong atom–cavity coupling in optical bistability and generated squeezing in a sodium atomic beam contained in a microscopic cavity that was pumped with a single optical beam.

We have carried out a series of single-beam squeezing experiments in Doppler-broadened sodium vapor in which all four interacting beams are collinear. This single-beam configuration ensures maximum spatial overlap of the four interacting beams, eliminates the need for careful beam alignment, and retains the advantage of a single-pass configuration in a vapor cell. Four-wave mixing quantum theories[4,5] have been reported, and a theory describing this single-beam configuration has been given by Ho *et al.*[6] that comprehensively takes into account the effects of loss, spontaneous emission, pump–probe phase mismatch, atomic collisions, Doppler broadening, and Gaussian-beam intensity variation. Despite theoretical predictions[4–6] that substantial squeezing should be achievable with four-wave mixing in two-level media, sodium-beam and sodium-vapor experiments to date have delivered inferred squeezing values of at most 60%. During the course of our measurements, we have identified self-focusing (by which we mean either self-focusing or self-defocusing) as the likely cause for the foregoing discrepancy between theory and experiment. The intense pump and probe Gaussian beams experience different, spatially varying nonlinear phase shifts. This differential self-focusing causes mode mismatch in the interaction region and thus imposes an upper limit on the effective interaction length.

In our experiments, a frequency-stabilized ring dye laser (300 kHz rms) tuned to the sodium D_2 line was passed through a vapor cell with a beam waist of ~80 μm at the center. The cell was a 47-cm-long, 2.5-cm-diameter stainless-steel pipe capped with two antireflection-coated windows and evacuated to ~1 mTorr. Sodium was placed in a 8-cm center section heat zone that was set at a temperature in the range of 150–250°C, with a stability of 1°C. Approximately 100 mTorr of helium buffer gas was used to prevent sodium from being deposited at the windows.

The single pump beam, of frequency Ω_p, provided for the two identical pump beams in the usual forward four-wave mixing geometry. In the interaction region, pairs of vacuum-state sidebands, with frequencies $\Omega_p \pm \omega_m$, were transformed into squeezed-vacuum sidebands by means of the nonlinear four-wave interaction.[5,6] The squeezed sidebands, however, were in quadrature with the mean field of the collinear transmitted pump and were therefore not observable using direct detection. The pump and the broadband squeezed sidebands, $\omega_m/2\pi \sim 50$–300 MHz, were separated with a 15-cm zigzag confocal filter cavity that was actively locked to the pump field in transmission. The reflected squeezed sidebands were then detected with the usual balanced homodyne detector,[7] with a local oscillator (LO) derived from the input pump beam. An ~3% reflected pump intensity was usually mixed with the reflected squeezed beam owing to imperfect cavity mode matching that was partially caused by self-focusing of the pump. We measured the residual pump intensity, which was usually 5–10 times smaller than the LO intensity, to obtain the correct detection shot noise. The output photocurrent from the balanced detector was fed into a rf spectrum analyzer that was used as a tuned receiver.

Figure 1 shows a typical trace of the photocurrent noise at frequency $\omega_m/2\pi = 212$ MHz as a function of the LO phase that was swept at 20π per second. The experimental parameters are as follows: the pump was 5-GHz blue detuned from the D_2 line, the cell

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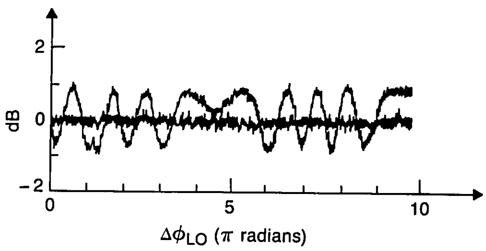

Fig. 1. Photocurrent noise levels showing the shot-noise level (the straight curve) and the squeezing and desqueezing (the wavy curve) as a function of the LO phase $\Delta\Phi_{LO}$. A break near $\Delta\Phi_{LO} = 5\pi$ was due to the piezoelectric return sweep.

temperature was 205°C, and the peak intensity averaged over the interaction path was $I_{peak} = 430\ W/cm^2$. Owing to the helium buffer gas, the transverse decay rate $\gamma/2\pi$ was assumed to be broadened from 5 to 5.8 MHz. In order to compare the experimental results with theory, we assumed the following parameters: line-center (zero detuning) saturation intensity $I_a = 18\ mW/cm^2$, line-center linear absorption coefficient $\alpha_a \simeq 2500/cm$, and pressure broadening coefficient of 8 MHz/Torr of helium owing to phase-changing collisions.

The straight line in Fig. 1 is the reference shot-noise level (±0.1 dB) caused by the LO plus the residual pump and was obtained when the vapor cell was not heated. The dips below the shot-noise level indicate squeezing, with a maximum observed squeezing of 0.8 ± 0.1 dB (~17%). When the LO phase was varied by $\pi/2$, the noise was enhanced with a maximum desqueezing of 1.0 ± 0.1 dB (~26%) above the shot-noise level. In the absence of propagation and detection losses, the inferred squeezing (desqueezing) was 32% (49%) for a system quantum efficiency of $\eta = 0.54 \pm 0.04$. η included the effects of detector quantum efficiency, losses, homodyne mode-matching effficiency, amplifier noise, and the residual pump intensity. With a larger beam waist of 130 μm and a longer interaction length of 15 cm, we have obtained an inferred squeezing of 57 ± 17%.

The single-beam theory by Ho *et al.*[6] assumes a uniform nonlinear phase shift $\delta\phi_p$, as experienced at the center of the pump Gaussian profile. In Fig. 2 we compare the experimental and theoretical values (for a given $\delta\phi_p$) for the inferred squeezing and desqueezing at different pump frequency detunings, with I_{peak} and ω_m the same as in Fig. 1. As the sodium density was increased by raising the cell temperature, the observed minimum and maximum variances of the resulting photocurrent noise were recorded, and the inferred squeezing and desqueezing were plotted. We monitored the onset of self-focusing by visually observing the far-field beam profile of the transmitted pump. As the temperature was increased and the squeezing reached its maximum value, the pump beam started to distort, i.e., self-focusing began. Beyond that temperature, the squeezing remained the same while the desqueezing continued to increase and the transmitted pump became more distorted. The crosses in Fig. 2 were obtained in a weak self-focusing regime, whereas the filled circles were taken at a higher temperature in a strong self-focusing regime. By temporarily unlocking the filter cavity, we measured the mode-matching efficiency between the transmitted pump and the LO, which was approximately 60–86% depending on the amount of self-focusing.

By self-focusing we mean that the center and the side of a Gaussian beam experience different amounts of nonlinear phase shift. Since the single-beam theory[6] assumes only a uniform nonlinear phase shift $\delta\phi_p$, we expect disagreement between theory and experiment when self-focusing is pronounced. In Fig. 2, the solid curves represent the theoretical squeezing and desqueezing for $\delta\phi_p = 0.25$, which is reasonable for the weak self-focusing case. We should point out that $\delta\phi_p = 0.25$ at different pump detunings corresponds to the same amount of phase shift but different line-center absorption coefficients α_a. Comparing the crosses with the solid curves, we see that in the region of minimal self-focusing, the theory agrees reasonably well with the experiment. We have made additional measurements (not shown) at different probe detunings ω_m and with different pressure-broadened linewidths in the weak self-focusing regime, with good agreement with theory.

The filled circles in Fig. 2 correspond to higher sodium densities (and therefore higher α_a), where strong self-focusing was encountered. The dashed curves are the theoretical values for squeezing and desqueezing with a larger α_a value so that $\delta\phi_p = 0.45$. In this case the theoretical fit, which uses a uniform $\delta\phi_p$, fails to account for the large increase in desqueezing with the same amount of squeezing, even if we adjust the various parameters within reasonable limits. As noted above, the actual mode-matching efficiency between the output pump and the LO was measured and taken into account for the data in Fig. 2. The disagreement between theory and experiment in this strong self-focusing regime indicates that self-focusing caused more than just the reduction of the mode-

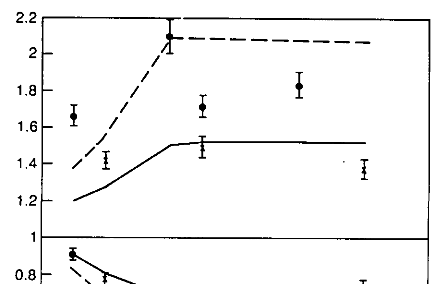

Fig. 2. Amount of inferred squeezing (lower trace) and desqueezing (upper trace) as function of pump detuning $\delta\omega_p/2\pi$, where the shot-noise level is unity. The crosses (filled circles) denote data pairs taken in the weak (strong) self-focusing regime. The solid curves are the theoretical predictions for $\delta\phi_p = 0.25$. The dashed curves are the theoretical prediction for $\delta\phi_p = 0.45$, which are to be compared with the data pairs with large desqueezing. The pump intensity was $I_{peak} = 430\ W/cm^2$, and the probe detuning was 212 MHz.

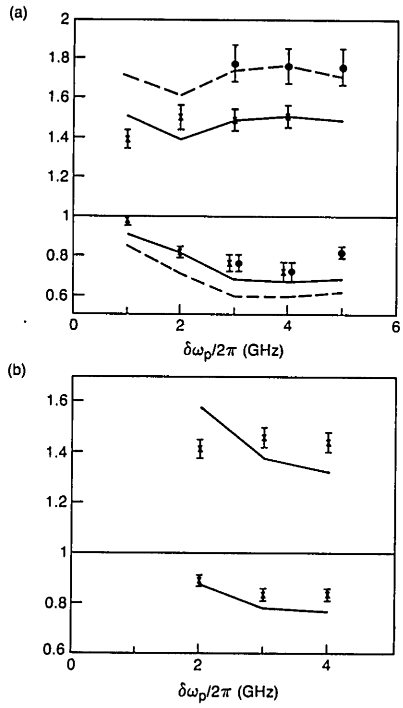

Fig. 3. Same as Fig. 2 except for the following differences: (a) $I_{\text{peak}} = 90$ W/cm^2, and the dashed curves are the theoretical prediction with $\delta\phi_p = 0.35$; (b) $I_{\text{peak}} = 17$ W/cm^2.

matching efficiency between the LO and the squeezed beam.

We believe that self-focusing causes an intrinsic mode mismatch between the strong pump beam and the weak probe beam in the interaction region. Theory[6] has shown that even at the degenerate frequency, the pump beam and the probe beam see a different nonlinear refractive index, thereby causing the two beams to self-focus or self-defocus differently. The overlap between the pump and probe beams is reduced as the beams propagate, owing to the difference in their beam curvatures. As a result, the effective interaction length is limited by the amount of self-focusing.

We have also taken data, with a procedure similar to that for Fig. 2, at lower pump intensities with $I_{\text{peak}} = 90$ W/cm^2 [Fig. 3(a)] and $I_{\text{peak}} = 17$ W/cm^2 [Fig. 3(b)] as a function of pump detuning. The solid curves in Figs. 3(a) and 3(b) are the theoretical predictions with $\delta\phi_p = 0.25$ that are appropriate for the weak self-focusing regime. The dashed curves in Fig. 3(a) are the theoretical predictions with $\delta\phi_p = 0.35$. Again, we see agreement between theory and those experimental data pairs, denoted by crosses, taken in the weak self-focusing region, and we see disagreement in the strong self-focusing regime. We also note that the maximum amounts of squeezing at $I_{\text{peak}} = 90$ W/cm^2 [Fig. 3(a)] and $I_{\text{peak}} = 430$ W/cm^2 (Fig. 2) were roughly the same, as they were both limited by self-focusing. At an even lower intensity $I_{\text{peak}} = 17$ W/cm^2 [Fig. 3(b)], other factors limited and reduced the maximum amount of squeezing. In Fig. 2 we did not take data beyond 10-GHz pump detuning because the highest obtainable vapor density at 250°C in our setup could not generate optimum squeezing outside this range. Similarly, in Figs. 3(a) and 3(b), where the pump intensities were lower, the maximum pump detunings were limited to 5 and 4 GHz, respectively.

We have made some numerical calculations that take into account the nonuniform nonlinear phase shift with some simplifying beam-propagation assumptions. Our preliminary research suggests that the maximum effective interaction length imposed by self-focusing for Gaussian beams will limit squeezing to approximately 75%, which is in agreement with an estimate given by Ho *et al.*[6] It also shows the rapid increase in desqueezing, for the same amount of squeezing, as observed experimentally. We note that the 75% squeezing limit can be achieved only with a larger beam waist than that in our experiments.

In conclusion, we have demonstrated a simple, single-beam configuration for generating squeezed states of light with a two-level atomic vapor. We showed that self-focusing played a major role in limiting the amount of squeezing, and in the regime where self-focusing was weak, there is good agreement between the theory and experiment for a wide range of experimental parameters. Our results indicate that the discrepancies between theory and previously reported experiments may be explained by the hitherto unexplored effect of self-focusing on four-wave mixing squeezed-state generation in atomic media.

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References

1. R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **55**, 2409 (1985); R. E. Slusher, B. Yurke, P. Grangier, A. Laporta, D. F. Walls, and M. Reid, J. Opt. Soc. Am. B **4**, 1453 (1987).
2. M. W. Maeda, P. Kumar, and J. H. Shapiro, Opt. Lett. **12**, 161 (1987); J. Opt. Soc. Am. B **4**, 1501 (1987).
3. L.A. Orozco, M. G. Raizen, M. Xiao, R. J. Brecha, and H. J. Kimble, J. Opt. Soc. Am. B **4**, 1490 (1987).
4. M. D. Reid and D. F. Walls, Phys. Rev. A **34**, 4929 (1986).
5. S. T. Ho, P. Kumar, and J. H. Shapiro, Phys. Rev. A **35**, 3982 (1987).
6. S. T. Ho, P. Kumar, and J. H. Shapiro, J. Opt. Soc. Am. B **8**, 37 (1991).
7. H. P. Yuen and V. W. Chan, Opt. Lett. **8**, 117 (1983); errata, Opt. Lett. **8**, 345 (1983).