Coherent nonlinear optical spectroscopy using photon-number squeezed light

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Using coherent nonlinear optical spectroscopy below the band edge of a GaAs/$Al_{0.3}Ga_{1-0.3}As$ multiple quantum well, photon-number squeezed light generated by a quantum-well laser is used to resolve the exciton resonances and the Urbach tail with detection sensitivity below the semiclassical shot-noise limit. The effect of polarization-dependent noise in the nonclassical probe field is also investigated. [S1050-2947(96)51009-7]

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With recent advances in nonclassical optics, noise reduction limits thought to be rigid from the semiclassical theory have been exceeded. In work by Caves [1], squeezed states of light were predicted to enhance the sensitivity of interferometric measurements beyond established semiclassical limits. The development of quadrature squeezed light sources that shortly followed led to a series of critical experiments that confirmed many benefits of nonclassical fields [2]. Recently, atom-photon interactions involving quadrature squeezed light demonstrated the predicted [3] sensitivity enhancement in frequency modulation (FM) spectroscopy [4]. In addition to improved noise properties, the use of quadrature squeezed light was demonstrated to lead to fundamental alterations in the atom-photon interaction [5]. The application of photon-number squeezed light has been predicted to result in nonclassical sensitivity enhancements for problems in spectroscopy, communications, and biomedical sensing [6]. Recently sub-shot-noise amplitude modulation [7,8] and frequency modulation [9] measurements were performed using photon-number squeezed light.

In this paper, coherent nonlinear spectroscopy is demonstrated in which a photon-number squeezed probe field and an amplitude-modulated coherent-state pump field are used to probe the differential transmission of the exciton resonances and Urbach tail in a GaAs/$Al_{0.3}Ga_{1-0.3}As$ multiple quantum well (MQW). The use of a photon-number squeezed probe field enables the measurement of spectroscopic signatures that are below the semiclassical shot-noise limit (SNL). Starting with a 1.9-dB squeezed probe field and a 30% attenuation in the sample, the spectroscopic sensitivity is enhanced by 1.0 dB (20%) beyond the sensitivity that can be achieved with a classical field, under identical experimental conditions. The impact of correlated noise in the nonclassical probe field on the spectroscopic sensitivity is evaluated, and the cancellation of polarization-dependent fluctuations results in sub-shot-noise sensitivity from a measurement using a polarized field that exhibits super-Poissonian amplitude fluctuations.

The true quantum limit (i.e., below the SNL) to low-noise spectroscopy (ideally determined by sample losses) can be most closely approached by interacting the full available intensity of the nonclassical probe field with the sample. The noise associated with such an experiment is described by the usual formula for attenuation of a photon-number squeezed field: $\langle(\Delta n')^2\rangle=[t\sigma+(1-t)]t\langle n\rangle$, where $1-\sigma$ is the fraction below the corresponding SNL ($\langle n\rangle$) of noise power in the field before the sample, $0\leqslant t\leqslant 1$ is the transmission coefficient of the sample and optical losses after the sample, and $\langle(\Delta n')^2\rangle$ is the photon-number variance at the detector. From this equation, it is clear that even with ideal optics and squeezing (for a number state $\sigma=0$) the noise level will be limited by the sample transmission to $(1-t)t\langle n\rangle=(1-t)\mathcal{S}$, which is 5.2 dB below the SNL ($\mathcal{S}$) for this experiment. This limitation is due to vacuum field fluctuations that enter through background losses in the sample.

The basic setup for the spectroscopy experiment is shown in Fig. 1(A). The nonclassical probe field is generated by a quantum-well laser injection-locked at 820 nm (12 193 cm^{-1}) and held at 124 K in a cryostat. Details of squeezed-light generation are given elsewhere [10]. One of two tunable dye lasers serves as an injection-locking source for the quantum-well laser, although use of a semiconductor laser for injection-locking is also possible [8]. The other dye laser provides a coherent-state pump field that is amplitude modulated at 3.6 MHz with an acousto-optic modulator (AOM). Pump intensity modulation depths measured directly before

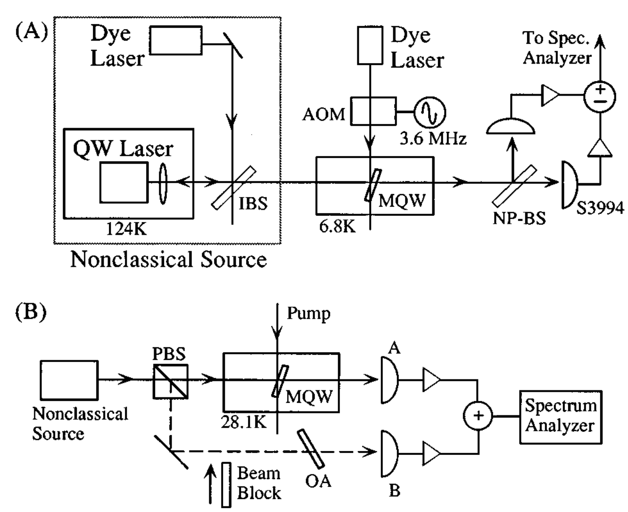

FIG. 1. (A) The setup used in polarization-insensitive spectroscopy measurements: QW, quantum well; IBS, injection-locking beam splitter; AOM, acousto-optic modulator; NP-BS, nonpolarizing beam splitter; MQW, multiple quantum well. (B) The setup used in polarization-dependent measurements: PBS, polarizing beam splitter; OA, optical attenuator.

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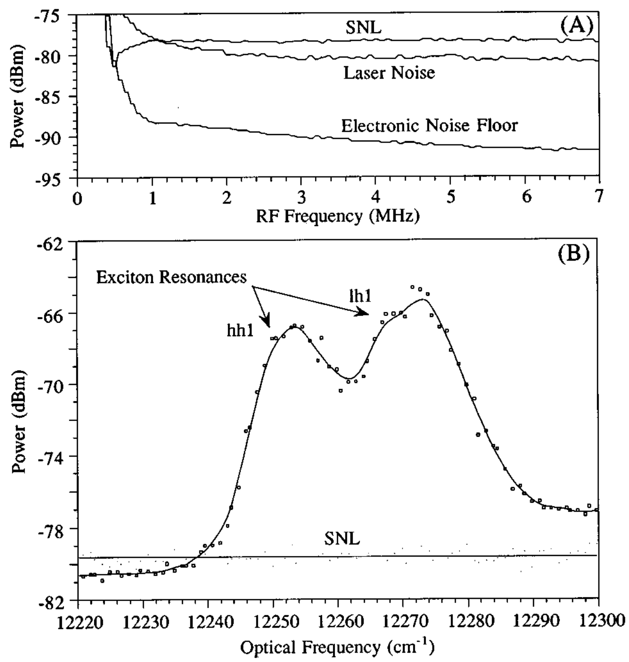

FIG. 2. (A) Amplitude noise spectrum with the sample removed; at the signal frequency, 3.6 MHz, the field is 1.9 dB squeezed. (B) Coherent nonlinear optical spectrum of a GaAs/$Al_xGa_{1-x}As$ multiple quantum well at 6.8 K. Probe power at 3.6 MHz is displayed as a function of the pump frequency. The low-frequency Urbach tail is observed to a power level 1.0 dB below the measured SNL. Spectrum analyzer: RBW 300 kHz, VF 300 Hz; five-scan average.

the sample are as large as 5% (limited by the AOM frequency response). The probe field output from the sample is detected in a balanced homodyne detector [10]. However, the noise reduction benefits of the squeezed probe field can be realized with direct detection in a single photodetector. In order to demonstrate sub-shot-noise detection, the balanced homodyne detector is used to accurately measure the signal strength and laser noise power relative to the SNL.

The GaAs/$Al_{0.3}Ga_{1-0.3}As$ MQW sample was grown by molecular-beam epitaxy and consists of 30 periods of 200-Å GaAs quantum wells separated by 200-Å barriers of $Al_{0.3}Ga_{0.7}As$. The n^+ GaAs substrate is removed by etching. The sample is mounted on an antireflection coated glass disk in a cryostat at 6.8 K. To avoid Fabry-Pérot modulation effects and optical feedback to the laser, the sample is angled with respect to both the pump and the probe fields.

The photon-number squeezing spectrum measured with the sample removed from the cryostat is shown in Fig. 2(A). The laser noise trace, which is 1.9 dB (2.1 dB corrected to the cryostat) squeezed at 3.6 MHz, is the measured amplitude noise from the sum photocurrent of the balanced homodyne detector. The SNL trace is measured on the difference photocurrent and is identical to the spectrum of a red-filtered halogen lamp to within 0.1 dB.

The photon-number squeezed field described above is used to probe the nonlinear response of the MQW sample. The probe field overlaps the classical pump field at a weak focus on the sample [Fig. 1(A)]. The optical frequency dependence of the nonlinear response in the MQW is shown in Fig. 2(B). The spectrum is recorded by tuning the pump wavelength through the sample resonances (note: the probe can also be tuned over the 10-GHz locking bandwidth). In order to observe the entire spectrum at a single reference level on the spectrum analyzer, the modulation depth on the pump was reduced to 0.3% for the run shown in Fig. 2(B). The solid curves are drawn as an aid to the eye. The SNL trace is the corresponding balanced homodyne detector difference photocurrent noise power. The two peaks in Fig. 2(B) are, in increasing wave number, the 1*s* heavy hole (hh1) and light hole (lh1) exciton resonances, respectively. The creation of exciton population by the pump modulates the probe transmission through such mechanisms as phase-space filling, excitation-induced dephasing, and induced absorption [11]. In separate measurements, induced absorption was found to dominate at the excitation densities present in this experiment.

The nonclassical sensitivity of this experiment is apparent in the Urbach tail at energies below the hh1 resonance. At frequencies less than 12 238 cm^{-1}, the power at the signal frequency is below the SNL. The minimum detectable amplitude is 1.0 dB below the SNL, which is the level of the quantum background noise limiting the measurement sensitivity. The range of frequencies over which the Urbach tail may be observed is consequently increased beyond the range that would be obtained if a classical (shot-noise-limited) probe field were used in this experiment.

Supporting measurements are required to confirm that the sub-shot-noise intensity of the probe is in fact the limiting noise source. Stray pump scatter or amplitude noise converted from the pump can potentially limit the experimental sensitivity. Measurements of the noise at 3.6 MHz when the probe field is blocked (before the sample) show that noise due to pump scatter is unobservable below the amplifier thermal noise floor (which is 10 dB below the probe field intensity noise). When the spectroscopic signal is present, the noise level at the signal frequency, measured by an electrical homodyne technique described previously [8], is equal to the intensity noise of the probe field (for 300-kHz RBW). In addition, the amplitude noise on the probe was measured with the pump modulation turned off and the pump wavelength tuned to various points in the spectrum. The presence of the pump was not observed to influence the amplitude noise on the probe, which may be important due to nonlinear processes.

One characteristic of nonclassical fields that is not considered in the experiment up to this point and that can be important in many practical applications is the presence of strong, often quantum, noise correlations. For example, in multimode semiconductor lasers, correlations have been observed between different longitudinal modes, such that while one or more individual modes may exhibit amplitude fluctuations in excess of the SNL, the sum over the fluctuations on all of the modes is below the SNL [12,13]. Even when a semiconductor laser is operating highly single mode ($>10^3$ side-mode suppression) [14], such anticorrelations in the residual side modes can become unbalanced after propagating through a material with frequency-dependent loss, as is often the case in spectroscopy. Thus, the measurement process would in this case not only introduce vacuum fluctuations, but also couple noise from other optical parameters (i.e., the mode partition) into the photon-number fluctuations. The net result can be background noise on the measurement that is larger than the SNL.

In this experiment, injection locking is used to suppress noise due to longitudinal mode effects; however, the low-temperature laser operation with polarization-sensitive injection locking can result in strong noise correlations between orthogonally polarized fields in the laser output. Due to a combination of birefringence-induced polarization mixing and intrinsic polarization coupling in the laser gain medium, amplitude fluctuations on the two polarization axes can be anticorrelated [10]. Similar to the case for the longitudinal modes, the polarization-dependent fluctuations are balanced between the two orthogonally polarized output fields. If a polarizer is introduced that disrupts this balance, polarization-dependent fluctuations will couple into the photon-number fluctuations, which can lead to even super-Poissonian statistics. Polarization-sensitive losses in the optics (e.g., optics tilted to avoid Fabry-Pérot fringes) or sample will also reduce the measurement sensitivity. More importantly, polarization dependence on the material nonlinearities (e.g., $\chi^{(3)}$) under measurement can influence such noise correlations.

To examine this aspect of spectroscopy with photon-number squeezed light, measurements are performed with and without a polarizer in the probe field. Without using a polarizer the quantum-well laser naturally has a polarization extinction ratio of 170:1. The spectrum in Fig. 2(B) is recorded using the quantum-well laser output without polarization analysis. Because the sample is tilted to avoid Fabry-Pérot effects, the sample transmission is made polarization dependent, and as a result the squeezing is less than would be expected from the 30% sample losses alone. Given 2.1-dB squeezing in the incident field and 7% losses after the sample, one would expect that the measurement sensitivity would be 1.2 dB rather than the observed 1.0 dB below the SNL. This difference can be accounted for by the coupling of polarization-dependent noise into the amplitude noise spectrum due to the polarization dependence of the sample transmission. The magnitude of this effect depends on the angle at which the sample is tilted. Because the sample index of refraction is strongly frequency dependent near the optical resonances, the strength of this effect also depends on the probe frequency.

Sub-shot-noise spectroscopy is also performed using polarization-selective techniques. When a polarizer is placed in the field (extinction ratio $>500:1$), the spectrum of trace *A* in Fig. 3 showing the Urbach tail below the hh1 resonance (MQW at 28.1 K) is measured. Because the polarizer couples polarization-dependent noise into the amplitude noise, the sensitivity of this measurement is, in fact, 0.5 dB above the SNL. It is important to realize, however, that the orthogonally polarized field, which is discarded by the polarizer, holds highly correlated information about the photon-number fluctuations on the field used in the measurement. If a polarizing beam splitter is used, then the photon-number fluctuations on the orthogonally polarized field may be measured. This information can then be recombined with the photocurrent from the field that passed through the sample and used to remove the background noise imposed by the polarization-dependent fluctuations.

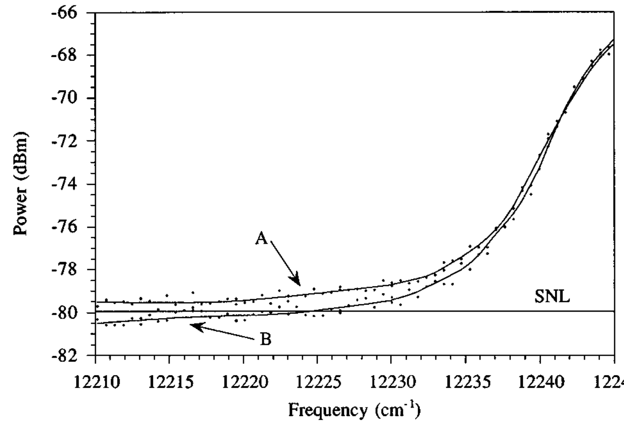

FIG. 3. Effect of polarization-dependent probe field fluctuations that are correlated below the SNL. Trace *A*: spectroscopy with a polarized field shows sensitivity 0.5 dB above the SNL. Trace *B*: when fluctuations of the orthogonally polarized field (previously discarded) are detected and recombined with the probe field noise, the sensitivity is reduced to 0.5 dB below the SNL.

The polarization-dependent noise is reduced in this way using the setup in Fig. 1(B), in which the beam splitter, PBS, is polarizing. The experimental conditions in this measurement are similar to those described above. When the beam block is removed and the attenuation in the weak orthogonally polarized field that does not pass through the sample is adjusted to match the loss in the sample (tilting the OA beam splitter), the polarization fluctuations cancel, and the sub-shot-noise spectrum shown in Fig. 3 (trace *B*) is observed. Although the spectroscopic measurement in this case is performed with a super-Poissonian field, the sensitivity is nonclassical and sub-Poissonian. Of course, if the probe field is not required to be highly polarized, then it is still better to work without a polarizer, as shown by the data in Fig. 2(B). Limitations to this technique include increased sensitivity to detector saturation, noise introduced through nonideal detector quantum efficiencies, and amplifier thermal noise.

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