Absorption Spectroscopy beyond the Shot-Noise Limit

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A novel absorption spectrometer where an absorber is placed in one beam of a parametric oscillator is described. The sensitivity is not shot-noise limited and may be enhanced by the operation of the parametric oscillator close to threshold.

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Traditional optical-absorption measurements have a fundamental limit set by the shot noise due to photon-number fluctuations in a coherent light beam. The shot noise therefore sets a limit to the sensitivity that may be achieved with use of coherent light. Recent work on squeezed light fields with fluctuations reduced below the vacuum or shot-noise limit suggest ways of beating this limit.[1] An experiment using squeezed light to improve the precision of a measurement of phase modulation in an optical interferometer beyond the shot-noise limit has recently been reported.[2] In the experiment we are suggesting use is made of the quantum mechanical correlations existing between a pair of photons produced in a parametric oscillator.

The simultaneous creation of a pair of signal and idler photons in a parametric amplifier has been experimentally demonstrated.[3,4] Since the photons are correlated, measurements on the idler beam may be used to reduce the photon fluctuations in the signal and produce a nearly photon-number state.[5,6] A suggestion has been made[7] that this be used to enhance the sensitivity of absorption measurements.[8,9] That is, a twin-beam absorption spectrometer may be constructed from a parametric amplifier with an absorber placed in one beam. The correlation with its twin beam enables an absorption sensitivity to be achieved which is no longer shot-noise limited.

In this Letter we wish to suggest a different scheme which, though relying on the correlated beams, introduces some advantageous new features. We propose to use a parametric oscillator, where an absorbing element is placed inside the optical cavity. The parametric oscillator is tuned so that one beam (say the signal) is on resonance with the absorption line of the sample. The intensity fluctuations in the difference of the signal and idler photocurrents have been shown to be zero at zero frequency, rising to the shot-noise limit for high frequencies. The zero-frequency fluctuations correspond to an infinite time where the photon-number fluctuations of the signal and idler beams exactly balance. The presence of additional intracavity losses will reduce the correlation between the signal and idler beams. The increase in fluctuations in the difference current may be used as a measure of the absorption.

The quantum statistics of the parametric oscillator may be described by a set of stochastic differential equations for the amplitudes α_0, α_1, and α_2 of the pump, signal, and idler modes, respectively. In the generalized P representation, the equations are[10]

$$\begin{aligned}\dot{\alpha}_0 &= -\kappa_0\alpha_0+\epsilon-\chi\alpha_1\alpha_2,\\ \dot{\alpha}_1 &= -\kappa_1\alpha_1+\chi\alpha_0\alpha_2^\dagger+R_1(t),\\ \dot{\alpha}_2 &= -\kappa_2\alpha_2+\chi\alpha_0\alpha_1^\dagger+R_2(t),\end{aligned} \tag{1}$$

where κ_0, κ_1, and κ_2 are the sums of the absorption plus cavity losses of the three modes, χ is the optical nonlinearity, and ϵ is the amplitude of the pump. The noise terms have the following nonzero correlations:

$$\langle R_1(t)R_2(t')\rangle=\chi\langle\alpha_0\rangle\delta(t-t'). \tag{2}$$

These equations may be solved above the threshold by a linearization technique. The linearized equations for the intensities are

$$\begin{aligned}\Delta\dot{I}_0 &= -\kappa_0\Delta I_0-\Delta I_s,\\ \Delta\dot{I}_s &= -2\delta\Delta I_D+2\kappa\kappa_0(E-1)\Delta I_0+F_s(t),\\ \Delta\dot{I}_D &= -2\kappa\Delta I_D+2\delta\kappa_0(E-1)\Delta I_0+F_D(t),\end{aligned} \tag{3}$$

where

$$\Delta I_j=\alpha_j^\dagger\alpha_j-I_j^{ss},$$

$$\Delta I_s=\kappa_1\Delta I_1+\kappa_2\Delta I_2,$$

$$\Delta I_D=\kappa_1\Delta I_1-\kappa_2\Delta I_2,$$

and I_j^{ss} is the steady-state intensity. We have defined

$$\kappa=\tfrac{1}{2}(\kappa_1+\kappa_2),\quad \delta=\tfrac{1}{2}(\kappa_1-\kappa_2),$$

and the nonzero noise correlations are

$$\langle F_s(t)F_s(t')\rangle = -\langle F_D(t)F_D(t')\rangle = 4\kappa_1\kappa_2 C^2\delta(t-t'), \tag{4}$$

where

$$C = [(\kappa_0\kappa_1\kappa_2)^{1/2}/\chi](E-1)^{1/2}, \quad E = |\epsilon|/\epsilon_{\rm thr},$$

where $\epsilon_{\rm thr}$ is the threshold pump amplitude.

The effect of losses in the intracavity medium is introduced by our denoting the total losses (cavity plus internal) by κ_j and the cavity losses by γ_j ($\gamma_j < \kappa_j$).

We wish to calculate the spectrum of fluctuations in the intensity difference between the external signal and idler modes. This spectrum is defined by

$$S_D(\omega) = \int d\tau\, e^{-i\omega\tau}\langle \hat{I}_1(\tau) - \hat{I}_2(\tau), \hat{I}_1(0) - \hat{I}_2(0)\rangle_{\rm ss}, \tag{5}$$

where $\hat{I}_j(t)$ are the external (operator) intensities and we use the notation $\langle A,B\rangle = \langle AB\rangle - \langle A\rangle\langle B\rangle$. This leads to the following relation[11] between the two-time correlation functions of the external intensities $\hat{I}_j(t)$ and the internal c-number intensities $I_j(t)$ where $I_j(t) = \alpha_j^\dagger(t)\alpha_j(t)$:

$$\langle \hat{I}_j(\tau), \hat{I}_k(0)\rangle = 4\gamma_j\gamma_k\langle I_j(\tau), I_k(0)\rangle + 2\delta_{jk}\delta(\tau)\gamma_j\langle I_j(0)\rangle. \tag{6}$$

The intensity difference spectrum may be written in the form

$$S_D(\omega) = S_0 + 4\int d\tau\, e^{-i\omega\tau}\langle I_D(\tau), I_D(0)\rangle_{\rm ss}, \tag{7}$$

where

$$S_0 = 2\Gamma_1\kappa_1\langle I_1\rangle_{\rm ss} + 2\Gamma_2\kappa_2\langle I_2\rangle_{\rm ss},$$

$$I_D(\tau) = \Gamma_1\kappa_1 I_1(\tau) - \Gamma_2\kappa_2 I_2(\tau),$$

and

$$\Gamma_j = \gamma_j/\kappa_j.$$

With use of the linearized equations above threshold, this may be evaluated to give for the normalized spectrum

$$\bar{S}_D(\omega) = S_D(\omega)/S_0 = [\omega^2 + 4\kappa^2(1-\Gamma) + 4\delta^2\Gamma]/(\omega^2+4\kappa^2) + \Gamma Y(\omega)/|Z(\omega)|^2, \tag{8}$$

where

$$Y(\omega) = [4\delta^2\epsilon/(\omega^2+4\kappa^2)]\{\omega^2[4\kappa+\kappa_0(E+1)] - \epsilon\} + q4\delta\epsilon\omega^2 + q^2 4(\kappa^2-\delta^2)[4(\kappa^2-\delta^2)+\omega^2](\kappa_0^2+\omega^2),$$

$$|Z(\omega)|^2 = [\epsilon - \omega^2(2\kappa+\kappa_0)]^2 + \omega^2(2\kappa\kappa_0 E - \omega^2)^2,$$

with

$$\epsilon = 4(\kappa^2-\delta^2)\kappa_0(E-1), \quad q = \tfrac{1}{2}(\gamma_1/\kappa_1 - \gamma_2/\kappa_2)/\Gamma,$$

$$\Gamma = \tfrac{1}{2}(\gamma_1/\kappa_1 + \gamma_2/\kappa_2), \quad S_0 = 4C^2\Gamma.$$

The vacuum or shot-noise level is $\bar{S}_D(\omega) = 1$ and perfect noise suppression is $\bar{S}_D(\omega) = 0$.

Let us first consider the case where the dampings are symmetric between the signal and idler, that is, $\gamma_1 = \gamma_2 = \gamma$, $\kappa_1 = \kappa_2 = \kappa$. Then

$$\bar{S}_D(\omega) = [\omega^2 + 4\kappa^2(1-\Gamma)]/(\omega^2+4\kappa^2). \tag{9}$$

At zero frequency ($\omega = 0$),

$$S_D(0) = S_0(1-\Gamma) = S_0(1-\gamma/\kappa)$$

with $\gamma \le \kappa$. Thus the effect of the losses is to reduce the correlation between the modes and there is no longer perfect suppression of the noise (Fig. 1).

We now consider a cavity with equal cavity damping rates $\gamma_1 = \gamma_2$ where we introduce an absorber at the idler frequency so that $\kappa_1 \ne \kappa_2$. In this case at $\omega = 0$

$$\bar{S}_D(0) = 1 - \Gamma + q^2\Gamma/(E-1)^2. \tag{10}$$

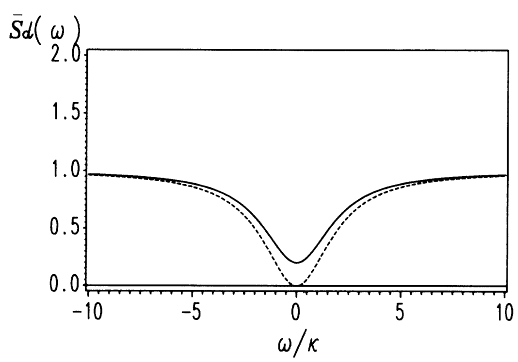

FIG. 1. Effect of intracavity absorptions. Plot of $\bar{S}_D(\omega)$: solid line, $\gamma_1 = \gamma_2 = \gamma$, $\kappa_1 = \kappa_2 = \kappa$, $\kappa_0/\kappa = 1$; dashed line, no intracavity absorption, $\gamma_j = \kappa_j$ and $\gamma_1 = \gamma_2$.

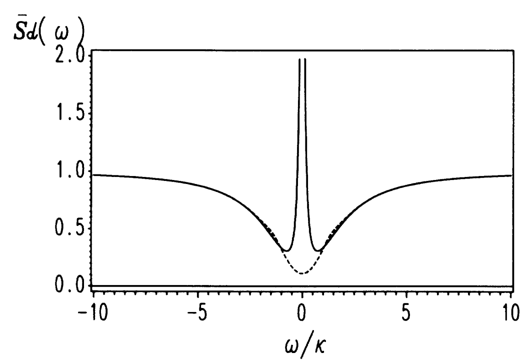

FIG. 2. Effect of asymmetrical intracavity absorption. Plot of $\bar{S}_D(\omega)$: $\kappa_0/\kappa = 1$, $\gamma_1/\kappa_1 = 1$, $\gamma_2/\kappa_2 = 0.8$, $\kappa_2/\kappa_1 = 1.25$, (solid line) $E = 1.05$, (dashed line) $E = 2$.

There is an extra noise term $q^2\Gamma/(E-1)^2$ which is sensitive to the difference $\gamma_1/\kappa_1 - \gamma_2/\kappa_2$. In Fig. 2 we show the intensity spectrum for the difference current in the presence of an intracavity absorber at the idler frequency. We see that from a noise background of zero for no absorption the presence of an absorber gives a marked increase in noise. Quantum noise reduced below the vacuum level in the difference current in a parametric oscillator has recently been observed experimentally by Heidmann *et al.*[12] At low frequencies, however, they observe an increase in the noise above the vacuum level. This may be explained by the slight difference in internal losses in the two beams and corresponds to the effect shown in Fig. 2. This sensitivity to absorption may be used to make an absorption spectrometer.

The level of absorption in the idler beam may be measured by insertion of a calibrated variable absorber in the signal beam. The absorption in the signal beam is then varied until a null in the noise is obtained. This indicates that the absorption in both arms is equal and we have a balanced twin-beam absorption spectrometer. The feature of the intracavity scheme is that the sensitivity may be enhanced by operation near threshold (compare Fig. 2). In the region of threshold there are critical fluctuations present which become manifest if there is an imbalance in the absorption in the two arms.

We have described a novel absorption spectrometer capable of achieving a sensitivity beyond the shot-noise limit. Such a spectrometer could be demonstrated with use of an optical parametric-oscillator of the type described by Wu *et al.*[13]

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[1] See J. Opt. Soc. Am. B **4** (1987) (special issue on squeezed states).

[2] M. Xiao, L. A. Wu, and H. J. Kimble, Phys. Rev. Lett. **59**, 278 (1987).

[3] D. Burnham and D. Weinberg, Phys. Rev. Lett. **25**, 84 (1970).

[4] S. Friberg, C. Hong, and L. Mandel, Phys. Rev. Lett. **54**, 2011 (1985).

[5] E. Jakeman and J. Walker, Opt. Commun. **55**, 219 (1985).

[6] R. Brown, E. Jakeman, R. Pike, J. Rarity, and P. Tapster, Europhys. Lett. **2**, 279 (1986).

[7] S. Reynaud, C. Fabre, and E. Giacobino, in Ref. 1.

[8] N. C. Wong and J. Hall, J. Opt. Soc. Am. B **2**, 1527 (1985).

[9] M. Gehrtz, G. Bjorklund, and E. Whittaker, J. Opt. Soc. Am. B **2**, 1510 (1985).

[10] P. D. Drummond, K. J. McNeil, and D. F. Walls, Opt. Acta **28**, 211 (1981).

[11] M. J. Collett and C. W. Gardiner, Phys. Rev. A **30**, 1386 (1984).

[12] A. Heidmann, R. J. Horowicz, S. Reynaud, E. Giacobiono, C. Fabre, and G. Camy, Phys. Rev. Lett. **59**, 2555 (1987).

[13] L. Wu, H. Kimble, J. Hall, and H. Wu, Phys. Rev. Lett. **57**, 2520 (1986).