RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 30, NUMBER 3 SEPTEMBER 1984

Squeezed-state generation via forward degenerate four-wave mixing

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(Received 5 March 1984; revised manuscript received 9 July 1984)

Degenerate four-wave mixing has been suggested as a possible generation scheme for squeezed-state light. A recent analysis of the quantum effects of probe-conjugate loss in backward degenerate four-wave mixing has shown that such loss puts an absolute limit on the squeezing that can be obtained via this generation scheme. In this Rapid Communication we show that it is the counter-propagating beam geometry of backward degenerate four-wave mixing that makes it ill suited for squeezed-state generation. On the other hand, the nominally copropagating beam geometry of forward degenerate four-wave mixing is shown to alleviate the absolute probe-conjugate loss limit on squeezing.

I. INTRODUCTION

Degenerate four-wave mixing (DFWM) has been suggested by Yuen and Shapiro as a possible source for squeezed-state generation.[1] Their model was a simple extension of the classical description of DFWM given by Yariv and Pepper.[2] Quantizing only the probe and the signal beams while retaining classical descriptions for the pump beams and the nonlinear medium, they showed that a two-photon coherent state (TCS) (essentially a minimum uncertainty squeezed state) is obtained by a 50/50 combination of the phase conjugate reflected beam and the transmitted probe beam from backward DFWM. A recent analysis by Bondurant, Maeda, Kumar, and Shapiro has shown that probe-conjugate loss puts an absolute limit on the squeezing that can be obtained via backward DFWM.[3] Since then Reid and Walls have given a fully quantum-mechanical treatment of backward DFWM.[4] Their analysis neglected the spatial propagation effects and showed that pump-induced spontaneous emission limits the amount of squeezing achievable. In this Rapid Communication we show that the absolute limit on probe-conjugate loss is because the preceding work all addressed backward DFWM, which has a counterpropagating beam geometry. This geometry is ideal for correcting phase aberrations via conjugate wave generation, but is ill suited for squeezed-state generation because of the aforementioned probe-conjugate loss limit. We show that forward DFWM, which has a nominally copropagating nonplanar beam geometry, removes the absolute probe-conjugate loss limit. Such an interaction geometry has been applied recently in studies of pressure-induced four-wave mixing interactions.[5]

In Sec. II we start with a classical analysis of forward DFWM. It is well known that large nonlinearities are obtained when the operating frequency is chosen near an atomic or molecular resonance. Therefore, in Sec. III, we develop a semiclassical treatment of forward DFWM in an atomic medium consisting of an ensemble of stationary two-level atoms. In Sec. IV we quantize the electromagnetic fields and examine the squeezing behavior of the output beams.

II. CLASSICAL EQUATIONS

Consider the geometry shown in Fig. 1. Two weak waves of wave vectors $\vec{k}_1$ and $\vec{k}_2$ propagate at small angles $\pm\phi/2$ from the z direction, determining a plane $\mathcal{P}$. The pump waves of wave vectors $\vec{k}_3$ and $\vec{k}_4$ also nominally propagate along the z direction; $\vec{k}_3$ and $\vec{k}_4$ are obtained from $\vec{k}_1$ and $\vec{k}_2$ by rotating the plane $\mathcal{P}$ along the $\mathcal{C}\mathcal{C}'$ axis. With this choice of wave vectors we note that $\vec{k}_1+\vec{k}_2=\vec{k}_3+\vec{k}_4$, i.e., the phase-matching condition is satisfied.

The fields are taken to be copolarized[6] plane waves of angular frequency ω

$$E_j(\vec{r},t)=\tfrac{1}{2}A_j(r_j)\exp[i(\omega t-\vec{k}_j\cdot\vec{r})]+\text{c.c.} \quad (1)$$

where r_j denotes the distance measured along $\vec{k}_j$. Following Yariv and Pepper[2] we can derive the following equations for coupled modes 1 and 2:

$$\frac{dA_1}{dz}=-i\kappa^*A_2^*,\quad \frac{dA_2^*}{dz}=i\kappa A_1 \quad (2)$$

where κ is the nonlinear coupling constant given by (mks units)

$$\kappa^*=\omega\chi^{(3)}A_3A_4/2cn_0\cos\frac{\phi}{2} \quad (3)$$

$\chi^{(3)}$ is the third-order susceptibility of the nonlinear medium, c is the speed of light in vacuum, and n_0 is the background refractive index. Equation (2) has the following solution:

$$A_1(z)=\cosh(|\kappa|z)A_1(0)-i\frac{\kappa^*}{|\kappa|}\sinh(|\kappa|z)A_2^*(0) \quad (4a)$$

$$A_2^*(z)=\cosh(|\kappa|z)A_2^*(0)+i\frac{\kappa}{|\kappa|}\sinh(|\kappa|z)A_1(0) \quad (4b)$$

in terms of boundary conditions at $z=0$.

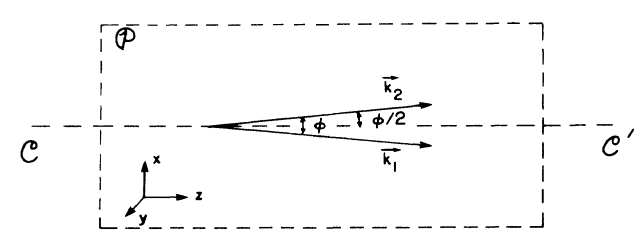

FIG. 1. Forward DFWM geometry.

III. COLLISIONLESS TWO-LEVEL MEDIUM

We now consider an ensemble of stationary two-level atoms forming the nonlinear medium. The atoms are characterized by a dipole moment, μ, and longitudinal and transverse relaxation times T_1 and T_2, respectively. Following Abrams and Lind[7] we obtain the following equations for the coupled modes 1 and 2:

$$\frac{dA_1}{dz}=-\gamma A_1-i\kappa^* A_2^*,\quad \frac{dA_2^*}{dz}=-\gamma A_2^*+i\kappa A_1\ , \tag{5}$$

where

$$\gamma=\frac{\alpha_0}{n}\frac{\omega}{\omega_0}\frac{(1+\delta^2)}{[1+\delta^2+(|A_3|^2+|A_4|^2)/A_s^2]^2}\frac{1}{\cos(\phi/2)}\ , \tag{6}$$

and

$$\kappa^*=\frac{\alpha_0}{n}\frac{\omega}{\omega_0}\frac{(\delta+i)}{[1+\delta^2+(|A_3|^2+|A_4|^2)/A_s^2]^2}\frac{2A_3A_4}{A_s^2\cos(\phi/2)}\ , \tag{7}$$

where $\delta=(\omega-\omega_0)T_2$ is the normalized detuning from line center, $A_s^2=\hbar^2/T_1T_2\mu^2$ is proportional to the line-center saturation intensity, ω_0 is the atomic transition frequency, $\alpha_0=\mu^2\Delta N_0 T_2\omega_0/2\epsilon_0 c\hbar$ is the line-center small signal-field attenuation coefficient, $\Delta N_0=(N_1-N_2)$ is the equilibrium population difference in the absence of the applied fields, n is the saturated refractive index given by

$$k^2=\frac{\omega^2}{c^2}\left[n_0^2-\frac{2\alpha_0 c}{\omega_0}\frac{\delta(1+\delta^2)}{[1+\delta^2+(|A_3|^2+|A_4|^2)/A_s^2]^2}\right]\equiv\frac{n^2\omega^2}{c^2}\ , \tag{8}$$

and k is the magnitude of the propagation vectors in the medium. We note that the pumps are nominally copropagating, so that no spatial averaging along the pump direction is required. Such averaging drastically reduces the DFWM reflectivity in the conventional counter-propagating pumps geometry.[7]

Equation (5) yields the following solution:

$$A_1(z)=e^{-\gamma z}\left[\cosh(|\kappa|z)A_1(0)-i\frac{\kappa^*}{|\kappa|}\sinh(|\kappa|z)A_2^*(0)\right]\ , \tag{9a}$$

$$A_2^*(z)=e^{-\gamma z}\left[i\frac{\kappa}{|\kappa|}\sinh(|\kappa|z)A_1(0)+\cosh(|\kappa|z)A_2^*(0)\right]\ , \tag{9b}$$

in terms of boundary conditions at $z=0$.

IV. SQUEEZED-STATE GENERATION

A. Lossless case

In giving a quantum treatment of backward DFWM, Yuen and Shapiro[1] replace the complex field amplitudes A_j and A_j^* with the photon annihilation and creation operators a_j and $a_j^\dagger$, respectively, for $j=1,2$. They assume that the pump fields A_3 and A_4 are strong and hence can be treated classically. They also describe the medium by a classical third-order susceptibility. Using their approach for our forward DFWM geometry we replace Eq. (2) with

$$\frac{da_1}{dz}=-i\kappa^* a_2^\dagger,\quad \frac{da_2^\dagger}{dz}=i\kappa a_1\ , \tag{10}$$

which has the following solution for a $z=0$ to $z=L$ interaction:

$$a_1(L)=\mu a_1(0)-i\nu a_2^\dagger(0)\ , \tag{11a}$$

$$a_2^\dagger(L)=\mu a_2^\dagger(0)+i\nu^* a_1(0)\ . \tag{11b}$$

Here $\mu=\cosh(|\kappa|L)$, $\nu=e^{-i\theta}\sinh(|\kappa|L)$, $\kappa=|\kappa|e^{i\theta}$, $a_1(0)$ and $a_2(0)$ are the input field operators at $z=0$. The outputs at $z=L$ are combined through a 50/50 beam splitter to generate two new modes described by annihilation operators c and d such that

$$c=[a_1(L)-ia_2(L)]/2^{1/2}\ , \tag{12a}$$

$$d=[a_1(L)+ia_2(L)]/2^{1/2}\ , \tag{12b}$$

in terms of which the solutions become

$$c=\mu c_{\text{in}}-\nu c_{\text{in}}^\dagger,\quad d=\mu d_{\text{in}}+\nu d_{\text{in}}^\dagger\ , \tag{13}$$

where

$$c_{\text{in}}=[a_1(0)-ia_2(0)]/2^{1/2}\ , \tag{14a}$$

$$d_{\text{in}}=[a_1(0)+ia_2(0)]/2^{1/2}\ , \tag{14b}$$

are annihilation operators describing field modes obtained by linear combination of the input modes to the four-wave mixer. Because $|\mu|^2-|\nu|^2=1$, it follows that c and d are in TCS if $a_1(0)$ and $a_2(0)$ are in coherent states (CS).[8]

B. DFWM in a lossy medium

It was shown in Sec. IV A that modes c and d are in TCS. Let us concentrate on the quadrature noise behavior of mode c. Let $c_1=(c+c^\dagger)/2$ and $c_2=(c-c^\dagger)/2i$ be the in-phase and out-of-phase quadratures of mode c, respectively. Then from Eq. (13) one can show that the quadrature variances are

$$\langle\Delta c_1^2\rangle\equiv\langle(c_1-\langle c_1\rangle)^2\rangle=|\mu-\nu|^2/4\ , \tag{15a}$$

$$\langle\Delta c_2^2\rangle\equiv\langle(c_2-\langle c_2\rangle)^2\rangle=|\mu+\nu|^2/4\ , \tag{15b}$$

when $a_1(0)$ and $a_2(0)$ are in CS.[8] Thus arbitrarily large squeezing is obtained in c_1 when $|\nu|$ is made arbitrarily large with $\mu\nu^*$ real and positive. Large values of $|\nu|$ have been shown to be obtainable in resonant media, such as described in Sec. III.[9] An inspection of Eqs. (6) and (7) together with the defining equations for μ and ν shows that a large value of $|\nu|$ is necessarily accompanied by a large value of γ, the loss per unit length in the medium. We follow the approach of Bondurant *et al.*[3] to analyze the effect of this probe-conjugate loss on the squeezing obtainable via forward DFWM in a resonant medium.

We note that Eq. (10) can be obtained from the effective interaction Hamiltonian

$$H_I=\hbar v(\kappa a_1 a_2+\kappa^* a_2^\dagger a_1^\dagger)\ , \tag{16}$$

using the Heisenberg equations of motion and then converting the temporal differential equations into spatial differential equations by the change of variable $z=vt$.

In order to account for probe-conjugate loss quantum mechanically we adjoin the system of Eq. (16) to two reservoirs of loss oscillators[10] described by annihilation operators b_l^s, for $l=1$ to ∞ and $s=1,2$. The total effective interaction Hamiltonian can therefore be written as

$$H_I^T=\hbar v(\kappa a_1 a_2+\kappa^* a_2^\dagger a_1^\dagger)+\hbar\sum_{s=1}^{2}\left(a_s\sum_{l=1}^{\infty}\kappa_l^* b_l^{s\dagger}+a_s^\dagger\sum_{l=1}^{\infty}\kappa_l b_l^s\right) , \quad (17)$$

where κ_l represents the coupling between the modes of interest, i.e., a_1 and a_2, and the loss oscillator modes. From Eq. (17), we obtain two coupled spatial differential equations for the slowly varying operators a_1 and a_2:

$$\frac{da_1}{dz}=-\gamma a_1-i\kappa^* a_2^\dagger+G_1(z) , \quad (18a)$$

$$\frac{da_2^\dagger}{dz}=-\gamma a_2^\dagger+i\kappa a_1+G_2^\dagger(z) , \quad (18b)$$

where γ is the loss per unit length and $G_s(z)$, $s=1,2$ are Langevin noise operators obeying

$$\gamma a_s(z)=v^{-2}\sum_l|\kappa_l|^2\int_0^z a_s(z')\exp[i(\omega_l-\omega)(z'-z)/v]\,dz'; \quad s=1,2 , \quad (19a)$$

and

$$G_s(z)=-\frac{i}{v}\sum_l\kappa_l b_l^s(0)\exp[i(\omega-\omega_l)z/v]; \quad s=1,2 , \quad (19b)$$

respectively. These noise operators, under the Wigner-Weisskopf approximation, obey the commutation rule

$$[G_s(z),G_{s'}^\dagger(z')]=2\gamma\delta_{ss'}\delta(z-z') \quad \text{for } s,s'=1,2 . \quad (20)$$

The set of Eqs. (18) can be integrated with the result

$$a_1(L)=e^{-\gamma L}[\mu a_1(0)-iv a_2^\dagger(0)]+\Gamma_1 , \quad (21a)$$

$$a_2^\dagger(L)=e^{-\gamma L}[iv^* a_1(0)+\mu a_2^\dagger(0)]+\Gamma_2^\dagger , \quad (21b)$$

where

$$\Gamma_1=\int_0^L e^{-\gamma(L-z')}\{\cosh[|\kappa|(L-z')]G_1(z')-ie^{-i\theta}\sinh[|\kappa|(L-z')]G_2^\dagger(z')\}\,dz' , \quad (22a)$$

$$\Gamma_2=\int_0^L e^{-\gamma(L-z')}\{-ie^{-i\theta}\sinh[|\kappa|(L-z')]G_1^\dagger(z')+\cosh[|\kappa|(L-z')]G_2(z')\}\,dz' . \quad (22b)$$

The loss per unit length, γ, appearing in Eq. (19a) is numerically the same as that in Eq. (6) for a medium consisting of stationary two level atoms. Operator equation (21) reduces to the classical equation (5) when expectation values are taken.

To calculate the effect of probe-conjugate loss on squeezing, we construct new modes as in Eq. (12) and evaluate the quadrature variances. As an example,

$$c=e^{-\gamma L}(\mu c_{\text{in}}-v c_{\text{in}}^\dagger)+(\Gamma_1-i\Gamma_2)/2^{1/2} , \quad (23)$$

and

$$\langle\Delta c_1^2\rangle=\frac{e^{-2\gamma L}}{4}(\mu-v)^2+\tfrac{1}{2}\langle(\Gamma_{11}+\Gamma_{22})^2\rangle , \quad (24)$$

where

$$\Gamma_{11}=(\Gamma_1+\Gamma_1^\dagger)/2,\ \ \Gamma_{22}=(\Gamma_2-\Gamma_2^\dagger)/2i ,$$

and we have chosen pump phases such that $\theta=0$. After substituting Eqs. (22a) and (22b) into Eq. (24) and evaluating the appropriate moments, we get

$$\langle\Delta c_1^2\rangle=\frac{\gamma(2N+1)}{4(\gamma+|\kappa|)}+\tfrac{1}{4}\exp[-2(\gamma+|\kappa|)L]\left(1-\frac{\gamma(2N+1)}{(\gamma+|\kappa|)}\right) , \quad (25a)$$

and

$$\langle\Delta c_2^2\rangle=\frac{\gamma(2N+1)}{4(\gamma-|\kappa|)}+\tfrac{1}{4}\exp[-2(\gamma-|\kappa|)L]\left(1-\frac{\gamma(2N+1)}{(\gamma-|\kappa|)}\right) , \quad (25b)$$

where N measures the initial excitation of the reservoir modes, i.e., $N=\langle b_l^{s\dagger}(0)b_l^s(0)\rangle$, and is assumed to be the same for all the modes.

Several cases of interest can now be considered.

(i) $\gamma=0$, i.e., the zero probe-conjugate loss limit, in which Eq. (25) reduces to Eq. (15) and ideal squeezing is obtained.

(ii) $\gamma\neq 0$, $\gamma<|\kappa|$, and $L\gg 1/(|\gamma-|\kappa||)$. In this limit $\langle\Delta c_2^2\rangle\to\infty$ and $\langle\Delta c_1^2\rangle\to\gamma(2N+1)/4(\gamma+|\kappa|)$, i.e., for a given N ideal squeezing can be obtained by making $|\kappa|\gg\gamma$. Though it should be noted that pump induced spontaneous emission noise will limit this squeezing as is the case in backward DFWM as shown in Ref. 4.

(iii) $\gamma>|\kappa|$ and $L\gg 1/(\gamma-|\kappa|)$. In this limit $\langle\Delta c_1^2\rangle\to\gamma(2N+1)/4(\gamma+|\kappa|)$ and $\langle\Delta c_2^2\rangle\to\gamma(2N+1)/4(\gamma-|\kappa|)$. For $N=0$, i.e., when the loss oscillators are initially unexcited, we get $\langle\Delta c_1^2\rangle=\gamma/4(\gamma+|\kappa|)\to\frac{1}{4}$ and $\langle\Delta c_2^2\rangle=\gamma/4(\gamma-|\kappa|)\to\frac{1}{4}$ for $\gamma\gg|\kappa|$. This result is expected here because loss totally dominates the nonlinear coupling and any quadrature noise asymmetry caused by the latter is swamped by the fluctuations introduced by the former. In the case of $\gamma\simeq|\kappa|$, a squeezing factor of 2 is still obtained in $\langle\Delta c_1^2\rangle$. The uncertainty product

$$\langle\Delta c_1^2\rangle\langle\Delta c_2^2\rangle=(2N+1)^2/16\left(1-\frac{|\kappa|^2}{\gamma^2}\right)>\tfrac{1}{16}$$

implying that a squeezed state which is not a minimum uncertainty state is generated. Also, since for our choice of θ, μv^* is real, this state is not a TCS either.[11]

V. DISCUSSION

The results of Sec. IV show that the DFWM beam geometry plays an important role in determining the squeezing that can be obtained in a realistic experiment. The nonlinear coupling introduces quantum noise asymmetry between the quadratures of the interacting modes. In DFWM this asymmetry is between the quadratures of two different modes and mode mixing at the output of the DFWM interaction is required to obtain new modes, whose two quadratures show this asymmetry. Probe-conjugate loss, on the other hand, introduces independent fluctuations into the two quadratures which are coupled via the nonlinear interaction, thus tending to equalize the observed output beam quadrature fluctuations. The latter is the result of case (iii) in Sec. IV, where loss dominates the nonlinear coupling.

In the counterpropagating geometry of backward DFWM, the interaction at any point couples forward and backward going waves. Because of loss, each of these waves has suffered the noise-symmetrizing effect noted above. It is the combination of the loss with the nonlocal nature (forward/backward wave coupling at all points in the interaction medium) that is responsible, we believe, for the severe loss limit on backward DFWM squeezed-state generation. On the other hand, in forward DFWM only forward going waves are coupled. Although loss injects a symmetric noise contribution at each point in the interaction medium, the nonlinear interaction from that point to the end of the interaction squeezes that noise contribution. Thus, with the forward interaction and gain coefficient in excess of the loss coefficient, the only fundamental limit on achievable forward DFWM squeezing will be due to pump induced spontaneous emission. Indeed, our view of the physics of this problem is supported by Yuen's loss analysis for DPA squeezed-state generation,[8] which shows that in that forward going three-wave interaction arbitrary squeezing is obtained for any γ and N so long as $|\kappa|$ can be made arbitrarily larger than γ.

In summary, forward DFWM appears more promising than backward DFWM as a squeezed-state generator. It is a phase-matched interaction with no fundamental limit on squeezing due to probe-conjugate loss. Moreover, we expect there will be differences in the limits on obtainable squeezing set by pump-induced spontaneous emission in forward and backward DFWM, because of the different physics of their spatial propagation characteristics, as described above. Furthermore, since all the beams are propagating in roughly the same direction, the interaction is not velocity selective. The participation of all velocity groups results in a very large nonlinear interaction. Both experimental and theoretical investigations of forward DFWM have recently been published.[12] Phase conjugation and sub-Doppler resolution due to strong saturation have been reported.

ACKNOWLEDGMENTS

The authors acknowledge helpful discussions with R. S. Bondurant and D. F. Walls. This work was supported by the Office of Naval Research Contract No. N00014-81-K-0662.

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