Quantum-statistical theory of the growth and stabilization of fields in a two-photon medium with competing nonlinear processes

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(Received 5 October 1987)

We formulate a quantum-statistical theory of the generation of the fields in a two-photon medium in which several competing nonlinear processes such as four-wave mixing, two-photon absorption, and amplified spontaneous emission are taking place. We derive the equation for the density matrix of the generated fields. We give numerical results for the time-dependent solutions, which enable us to understand the growth and stabilization of the fields in such a medium. We show the generation of new types of the coherent states of the radiation field. Our analysis shows that the generated fields, both in transient and the steady-state domain, have very striking quantum properties leading to squeezing, sub-Poissonian statistics, and violations of Cauchy-Schwarz inequalities. Wherever possible we compare our results with the experiment of Malcuit et al. and the semiclassical theory of Boyd et al.

I. INTRODUCTION

The interaction of an intense field with a nonlinear medium results in several nonlinear processes which interfere with each other. This interference changes the character of the various signals generated either coherently or incoherently. Many examples of such interference effects have been discussed in the literature. For example, the five-photon ionization with three-photon resonance in Xe has been shown to be suppressed under certain conditions. This suppression has been shown to be due to the resonant generation of the radiation at the third harmonic. The generation of the third harmonic opens up a new channel for multiphoton ionization. It has been further shown that the interference effects can be controlled by the application of an additional laser resonant with a suitably chosen transition. Interference effects have also been studied for two-photon media. The interference effects control the growth of the fields in the medium and lead to a well-defined steady state of the system. Thus a sort of optical balance takes place in the medium due to the competition of several nonlinear processes. For example, the fields are known to grow in parametric and four-wave mixing processes. The growth of such fields can be controlled by the linear absorption in the medium. However, in a two-photon medium the linear absorption is insignificant. In such cases, the nonlinear absorption of the fields generated by a parametric process can lead to the stabilization of the fields. Obviously one needs to have a full quantum-mechanical theory of the optical balance so that not only can one study the initial quantum-mechanical growth of the fields at frequencies other than those applied, but also one can examine the statistical aspects of the generated fields.

In this paper we treat the case of a two-photon medium in which there is strong competition between four-wave mixing and two-photon absorption processes. Such a competition leads to drastic changes in the amplified spontaneous emission produced by a two-photon medium. One of us has previously treated some aspects of this problem. It was shown that the fields generated by such a medium had a very strong quantum character. While the previous paper gave results in the steady state, the present paper concentrates on the quantum dynamics. We also include the effects of off-resonant excitation. The dynamical study enables us to understand how the optical balance in nonlinear media occurs. A semiclassical study of the optical balance has already appeared.

The organization of the paper is as follows. In Sec. II we present the theoretical model. We derive the dynamical equation for the density matrix of the generated fields. In Sec. III we present numerical results for the dynamics. We discuss in detail the antibunching, sub-Poissonian, and squeezing properties of the generated fields. In Sec. IV we present a steady-state solution of the model. We demonstrate that the optical balance results in new types of the coherent states of the field.

II. QUANTUM THEORY OF THE COMPETING NONLINEAR PROCESSES IN A TWO-PHOTON MEDIUM

Consider the situation shown schematically in Fig. 1. A pump of frequency \( \omega_1 \), which is far detuned from the intermediate state, but which is close to the two-photon resonance, excites the gaseous system and creates the two-photon coherence. We assume that the pump is intense enough that its depletion can be ignored. However, the pump is not so intense that the saturation effects can
be ignored. These conditions conform to the experimental work\(^\text{1}\) of Malcuit et al. For the system shown in Fig. 1, the following processes are important.

1. Two-photon absorption (TPA) from the pump field described by the nonlinear susceptibility

\[
\chi_{TPA}^2 = \frac{N |\mu_{ba}|^2 |\mu_{cb}|^2}{\varepsilon_0^2 \Delta_1^2 \Delta_2 (\Delta_2 - i \Gamma_{ca})} .
\]

(2.1)

We keep our analysis parallel as much as possible to the semiclassical theory\(^2\) of Boyd et al. Here \(\mu_{ij}\) is the dipole matrix element and \(\Gamma_{ca}\) is the two-photon linewidth.

2. Generation of photons with frequencies \(\omega_2\) and \(\omega_3\) via four-wave mixing (FWM) characterized by the susceptibility

\[
\chi_{FWM}^2 = \frac{N |\mu_{ba}|^2 |\mu_{cb}|^2}{\varepsilon_0^2 \Delta_1 \Delta_2 \Delta_3 (\Delta_2 - i \Gamma_{ca})} , \quad \omega_2 + \omega_3 = 2 \omega_1 .
\]

(2.2)

3. Simultaneous absorption of the generated photons \(\omega_2\) and \(\omega_3\) described by the susceptibility

\[
\chi_{TPA}^d = \frac{N |\mu_{ba}|^2 |\mu_{cb}|^2}{\varepsilon_0^2 \Delta_1 \Delta_2 \Delta_3 (\Delta_2 - i \Gamma_{ca})} .
\]

(2.3)

Note that the single-photon absorption events are assumed to be unimportant.

4. Amplified spontaneous emission (ASE) from the excited state \(|c\rangle\). The ASE is proportional to the population in the state \(|c\rangle\).

All these four processes interfere with and determine the dynamical behavior of the system. The characteristics of the generated radiation can be understood systematically if we treat the generated fields quantum theoretically, since these fields grow from the vacuum. Besides, one would expect that such generated photons as \(\omega_2\) and \(\omega_3\) will be strongly correlated, since both photons are either created together or destroyed together. We will, in the special case, recover the results of the semiclassical theory of Boyd et al. Note that the semiclassical equations for the field envelopes \(\epsilon_2\) and \(\epsilon_3\) at the generated frequencies can be written in the form

\[
\frac{d\epsilon_2}{dz} = \frac{2\pi i \omega_2}{c} (\chi_{FWM}^2 \epsilon_2^2 + \chi_{TPA}^2 \epsilon_3^2) ,
\]

\[
\frac{d\epsilon_3}{dz} = \frac{2\pi i \omega_3}{c} (\chi_{FWM}^2 \epsilon_2^2 + \chi_{TPA}^d \epsilon_3^2) .
\]

(2.4)

(2.5)

We consider only the phase-matched case. Note that the refractive index of the medium at \(\omega_1\) (and \(\omega_2\)) is very close to 1 as it is far detuned from the single-photon resonance. Before we discuss the development of the quantum theory, we indicate the kind of structure that one might expect of the equations for the field operators. We express the positive frequency parts \(E_2^+\) and \(E_3^+\) of the field operators as

\[
E_2^{(+)} = -i \left[ \frac{2\pi \hbar \omega_2}{V} \right]^{1/2} be^{-i \epsilon_2^+ t} ,
\]

\[
E_3^{(+)} = -i \left[ \frac{2\pi \hbar \omega_3}{V} \right]^{1/2} ce^{-i \epsilon_3^+ t} ,
\]

(2.6)

where \(V\) is the quantization volume. Here \(b\) and \(c\) are boson annihilation operators with the commutation relations

\[
[b, c] = 0 , \quad [b, c^+] = 1 , \quad [c, c^+] = 1 , \quad [b, c^+] = 0 .
\]

(2.7)

Note that in the quantum theory one expects field envelopes to be replaced by the field operators and Eqs. (2.4) and (2.5) to be replaced by the quantum Langevin equations. For problems involving forward propagation and four-wave mixing one can use the correspondence rule \(i 
\to z/c\). Therefore in place of (2.4) and (2.5) we expect to get quantum Langevin equations

\[
\frac{d\epsilon_a}{dt} = -i G c^+ - \frac{1}{2} \kappa (1 + i \eta) c^+ \epsilon^+ c + F_b(t) ,
\]

\[
\frac{d\epsilon_c}{dt} = -i G b^+ - \frac{1}{2} \kappa (1 + i \eta) b^+ \epsilon^+ c + F_c(t) .
\]

(2.8)

The properties of the noise sources \(F_b\) and \(F_c\) are to be determined from the microscopic theory. The parameters \(G, \kappa,\) and \(\eta\) are related to the nonlinear susceptibilities (2.2) and (2.3): \(G\) is related to the four-wave mixing susceptibility

\[
G = 2\pi \sqrt{\omega_2 \omega_3 \epsilon_0^2 \chi_{FWM}^2} ,
\]

(2.9)

and \(\kappa\) and \(\eta\) are related to the two-photon absorption nonlinearity

\[
\kappa = \frac{8\pi^2 \hbar \omega_2 \omega_3}{V} \text{Im} \chi_{TPA}^2 , \quad \eta = -\Delta_2 / \Gamma_{ca} .
\]

(10)

Let \(\rho\) be the density matrix for the fields \(b\) and \(c\). It is obtained from the full density matrix by eliminating adiabatically\(^\text{3}\) the atomic degrees of freedom. The adiabatic elimination can be done, assuming that the response time of the medium is fast. Note that such an assumption is implicit in the semiclassical theory which is based on the steady-state response [Eqs. (2.2) and (2.3)] of the atomic system. In fact, the problem of two-photon absorption by

\[\text{FIG. 1. Schematic illustration of the two-photon medium pumped by a field } \epsilon(\omega_1), \text{ } \Delta \text{'s give the various detunings. The fields } \omega_2 \text{ and } \omega_3 \text{ are generated by four-wave mixing. These photons can be reabsorbed by a two-photon absorption process.}\]
an atomic system has been treated quantum theoretically.\textsuperscript{9} The density-matrix equation describing the absorption of two photons of frequencies $\omega_2$ and $\omega_3$ is\textsuperscript{10}
\[
\frac{\partial \rho}{\partial t} = -i\eta \frac{\hbar}{2} [b^\dagger b c^\dagger c, \rho] - \frac{1}{2} \eta [b^\dagger b^\dagger c \rho - 2 b c b^\dagger c^\dagger + \rho b^\dagger b c^\dagger c] ,
\] (2.11)
where $\eta$ and $\kappa$ are defined by Eq. (2.10). The four-wave mixing can be described in terms of an effective Hamiltonian\textsuperscript{11}
\[
H_{\text{eff}} = \hbar G b^\dagger c^\dagger + \text{H.c.} ,
\] (2.12)
where $G$ is defined by Eq. (2.9). On combining Eqs. (2.11) and (2.12) we obtain our basic equation, which describes the generation of the fields $\omega_2$ and $\omega_3$ by a pump wave of frequency $\omega_1$ propagating through a two-photon medium,
\[
\frac{\partial \rho}{\partial t} = -i [G b^\dagger c^\dagger + G^* b c, \rho] - i \frac{\hbar}{2} [b^\dagger b c^\dagger c, \rho] - \frac{1}{2} \eta [b^\dagger b^\dagger c \rho - 2 b c b^\dagger c^\dagger + \rho b^\dagger b c^\dagger c] .
\] (2.13)
Equation (2.13) is equivalent to the quantum Langevin equations (2.8). The properties of the noise operators $F_b(t)$ and $F_c(t)$ can be determined\textsuperscript{9} from (2.13), for example,
\[
\begin{align*}
\langle F_b(t) \rangle &= 0 , \\
\langle F_b(t) F_b^\dagger (t') \rangle &= 2 \kappa \langle \epsilon(t) \epsilon^\dagger (t') \rangle \delta(t - t') .
\end{align*}
\] (2.14)
We do not list other properties of $F$, as in the following we will work only with the density-matrix equation (2.13). Note that the mean value equations are given by
\[
\begin{align*}
\langle \dot{\epsilon} \rangle &= -i G \langle \epsilon^\dagger \rangle - \frac{1}{2} \kappa (1 + i \eta) \langle c b \rangle , \\
\langle \dot{c} \rangle &= -i G \langle b^\dagger \rangle - \frac{1}{2} \kappa (1 + i \eta) \langle b^\dagger c \rangle .
\end{align*}
\] (2.15)
In view of the nonlinearity of Eqs. (2.15), an infinite hierarchy of equations for the mean values is generated. This hierarchy can be closed by making some sort of decorrelation approximation. In what follows we avoid any decorrelation approximation, since the generated fields are expected to have strong correlations.

We next relate the properties of the amplified spontaneous emission\textsuperscript{12} to the properties of the generated fields. The ASE is proportional to the population of the uppermost state $|c\rangle$. The population of the state $|c\rangle$ can be obtained in terms of the rates of the absorption of two photons. As mentioned earlier, there are two channels for the nonlinear absorption: the atom can absorb either two photons from the pump or the two generated photons. The transition amplitudes for these two channels can interfere with each other. In order to obtain the total two-photon absorption rate we can introduce an effective two-photon Hamiltonian\textsuperscript{13}
\[
H_{\text{eff}}^{\text{TPA}} = \hbar \left[ M_p e^2(\omega_1) + M_G b c \right] c \langle \epsilon | + \text{H.c.} ,
\] (2.16)
where $M_p$ and $M_G$ are the effective two-photon matrix elements. These matrix elements are related to the susceptibilities $\chi_2^{\text{TPA}}$ and $\chi_3^{\text{TPA}} (|M_p|^2 \approx \text{Im} \chi_2^{\text{TPA}})$. Using the form of the nonlinear susceptibilities one can show that
\[
\frac{M_p e^2(\omega_1)}{M_G} = \Delta_3 e^2 V - 2 \pi \hbar \Delta_1 V / \omega_2 \omega_3 .
\] (2.17)
The population in the excited state will be determined essentially by the quantity
\[
\rho_{cc} \propto \langle [M_p e^2(\omega_1) + M_G b c^\dagger] [M_p e^2(\omega_1) + M_G b c] \rangle ,
\] (2.18)
which in turn will be determined by the solution of (2.13). Thus the complete solution of (2.13) will enable us to study not only the intensities of the generated fields but also their quantum-statistical properties and the characteristics of ASE.

III. GROWTH OF THE QUANTUM FIELDS AND APPROACH TO OPTICAL BALANCE

In this section we will obtain the time-dependent solution of (2.13). The time-dependent solution enables us to study how the fields grow starting from initial quantum noise and how the competing nonlinear processes lead to optical balance, i.e., a well-defined steady state. We note that Eq. (2.13) admits the conservation law
\[
\langle b^\dagger b - c^\dagger c \rho \rangle = 0 , \quad p = 1, 2, 3, \ldots
\] (3.1)
Since initially both fields $b$ and $c$ are in a vacuum state, the time-dependent solution of (2.13) has the form
\[
\rho = \sum_{m,n} \beta_{mn}(t) |m, m\rangle \langle n, n| .
\] (3.2)
Note that Eq. (3.2) has states with equal occupation number in the two modes. This is because the photons in the two modes are either created together or destroyed together. Since we start both the modes in the vacuum, it is clear that at any time the state must have equal occupation numbers in the two modes. The state (3.2) is also consistent with the conservation law (3.1). The coefficients $\beta_{mn}(t)$ satisfy
\[
\frac{\partial}{\partial T} \beta_{mn}(T) = -i \eta (m^2 - n^2) \beta_{mn} - [\beta_{mn}(m^2 + n^2) - 2(m + 1)n(n + 1) \beta_{m+1,n+1}] - \xi \beta_{m,n+1}(n + 1)
\]
\[+ \xi \beta_{m-1,n} + \xi^* \beta_{m,n-1} - \xi^* \beta_{m+1,n}(m + 1) = 0 ,
\] (3.3)
\[T = \frac{1}{2} \kappa t , \quad \xi = -2i G / \kappa .
\] (3.4)
Equation (3.3) cannot be solved analytically for the coefficients $\beta_{mn}$ except in the special case $\kappa \to 0$. In the limit of a thin sample the reabsorption of the generated photons can be ignored and then $\beta_{mn}$ are

$$\beta_{mn} = \gamma_m \gamma_n^* ,$$

$$\gamma_m(T) = \text{sech} \left( \frac{\xi T}{\xi} \right) \left( \frac{\xi}{\xi} \right)^m \tanh \left( \frac{\xi T}{\xi} \right).$$

For nonzero $\kappa$ we have solved Eq. (3.3) numerically and we show the results in Figs. 2 and 3 for the cases when the pump is tuned exactly to the two-photon resonance and when the pump is somewhat off the resonance $\eta \neq 0$.

We present time-dependent results for the mean intensities of the generated field and their quantum statistics. The following quantities are plotted in these figures.

(a) Mean number $N$ of photons in each mode,

$$N = \langle b^\dagger b \rangle = \langle c^\dagger c \rangle = \sum_{m=0}^{\infty} \beta_{mm} m.$$  

(b) ASE proportional to $A$,

$$A = \left( 1 + \frac{M_G^*}{M_p e^{2(\omega_1)}} b^\dagger c \right) \left( 1 + \frac{M_G}{M_p e^{2(\omega_1)}} bc \right)$$

$$= 1 + \langle b^\dagger c bc \rangle - \langle bc \rangle \frac{\langle b^\dagger c \rangle}{\varphi}.$$  

which can be expressed in terms of $\beta_{mn}$ as

$$A = 1 + \frac{1}{|\varphi|^2} \sum_{m=0}^{\infty} \beta_{mm} m^2 - 2 \text{Re} \frac{1}{\varphi} \sum_{m=0}^{\infty} \beta_{m,m-1} m.$$  

(c) The cross correlation $C$ between the two modes,

$$C = \langle b^\dagger bc^\dagger c \rangle - \langle b^\dagger b \rangle \langle c^\dagger c \rangle$$

$$= \sum_{m=0}^{\infty} \beta_{mm} m^2 - N^2.$$  

(d) The antibunching or the sub-Poissonian property of the fields,

$$G = \frac{\langle b^\dagger b^2 \rangle - \langle b^\dagger b \rangle^2}{\langle b^\dagger b \rangle^2} = \frac{\sum_{m=0}^{\infty} m^2 \beta_{mm} - N - N^2}{N^2}.$$  

(e) The parameter $S$,

$$S = N - \text{Re} \langle bc \rangle ;$$

which as we will see in Sec. IV, gives the squeezing property of the generated fields.

Using the definitions (2.9) and (2.10) it is seen that $\varphi$ [defined by (2.17)] is related to the parameter $\xi$,

$$\varphi = \xi / (1 + i \eta).$$  

Note further that semiclassically quantities like $C$ and $S$ would be zero. This is because in a decorrelated theory $C = |b|^2 |c|^2 - |b|^2 |c|^2 = 0$. Figures 2 and 3 show

FIG. 2. Evolution of the various quantum characteristics $N$, $A$, $C$, $G$, and $S$, defined by Eqs. (3.6)–(3.11), of the fields as a function of $T = \frac{1}{2} \kappa T = \frac{1}{2} \kappa \tau$. The curves are labeled by the quantities they represent. The scale for different curves is different and is marked on the left. The pump satisfies the two-photon resonance condition, i.e., $\eta = 0$. The parameter $\varphi$ is chosen to be equal to (a) 0.5, (b) 1.0, (c) 2.0, and (d) 5.0.
how important are the quantum features of the generated fields. The cross correlation $C$ between two modes remains quite significant over a range of parameters. Note that the parameter $A$ goes to zero in the limit of long times. Thus the ASE continuously gets less and less as the fields $b$ and $c$ grow. Eventually there is complete suppression of ASE, which is in conformity with the observation$^2$ of Malcuit et al. The complete suppression also occurs if the pump is somewhat detuned from the two-photon resonance. For moderate values of the parameter $\varphi$ the fields have considerable sub-Poissonian statistics. The amount of squeezing relative to the coherent state is given by $|2S|$. The figures show that the fields have a substantial amount of squeezing over a range of parameters. For example, for $\varphi=5$, $\eta=0$, the amount of squeezing is between 60% and 80% over a rather large range of $T$ values. Note that a given value of $T$ essentially corresponds to a given length of the sample.

**IV. GENERATION OF PAIR COHERENT STATES AS A RESULT OF OPTICAL BALANCE**

In this section we derive the steady-state solution of Eq. (3.3). The steady state arises because of the optical balance between two nonlinear processes, viz., coherent generation of the photons at $\omega_1$ and $\omega_2$ and the nonlinear absorption of the generated photons. On setting $\partial \beta_{mn}/\partial T=0$, we get a recursion relation for the coefficients $\beta_{mn}$. The recursion relation turns out to have the solution

$$\beta_{mn} = \frac{\varphi^m \varphi^n}{m! n!} \beta_{00}, \quad \varphi = -\frac{2iG}{\kappa (1+i\eta)} = -\frac{\zeta}{1+i\eta}. \quad (4.1)$$

The coefficient $\beta_{00}$ is determined from the normalization of $\rho$. On combining (4.1) and (3.2), the steady-state density matrix can be written in the form

$$\rho = \langle \varphi \rangle_p \langle \varphi |,$$  

$$\langle \varphi \rangle_p = N_0 \sum_{m=0}^{\infty} \frac{\varphi^m}{m!} |m,m\rangle, \quad N_0^2 \sum_{m=0}^{\infty} \frac{|\varphi|^2m}{(m!)^2} = 1. \quad (4.3)$$

It can be further proved that $\langle \varphi \rangle_p$ is an eigenstate of $bc$, 

$$bc |\varphi \rangle_p = \varphi |\varphi \rangle_p, \quad (4.4)$$

i.e., $|\varphi \rangle_p$ is an eigenstate of the operator which corresponds to the simultaneous annihilation of photons in the modes $b$ and $c$. Thus $|\varphi \rangle_p$ is a new type of coherent state associated with the pair annihilation operator. We would refer to such states as pair coherent states. These states are distinct from the usual oscillator coherent states, two-photon or SU(1,1) coherent states, etc. Such states have been encountered before in the literature in connection with the Abelian charge, etc. Our analysis shows how such states can be produced as a result of the optical balance arising from the competition of two nonlinear processes in a two-photon medium. The Sudarshan-Glauber distribution ($P$ function) does not exist for the pair coherent states. However, the $Q$ function for such states exists,
FIG. 4. Quantum-statistical properties of $N, G, C, S$, and $I$ [defined by Eq. (4.8)] of the generated fields in the steady-state limit. The scales for various curves are as shown.

\[
Q(z_b,z_c) = \langle z_b,z_c | \varphi \rangle_p \langle \varphi | z_b,z_c \rangle ,
\]
\[
= \left| I_0(2qz_b^*z_c^*)^{1/2} \right|^2 \left| I_0(2|\varphi|) \right|^2 \times \exp(-|z_b|^2-|z_c|^2). \tag{4.5}
\]

In (4.5), $|z_b,z_c\rangle$ is the usual two-mode coherent state. Thus the $Q$ function for pair coherent states is distinct from the Gaussian $Q$ function for two-photon coherent states. In Fig. 4 we show various quantum properties of the generated fields in the steady state. Note that ASE [Eq. (3.7)] vanishes in the steady state as the steady state is an eigenstate of $bc$ with eigenvalue $\varphi$. The semiclassical result for the field amplitude in the steady state is $\sqrt{\varphi}$. Hence $S$ [Eq. (4.11)] will also give the deviation in the mean number of photons from the semiclassical value. Figure 4 shows very significant quantum properties of the generated fields. In Fig. 4 we also plot $I$ defined by

\[
I \equiv \langle b^+b^2 \rangle \langle c^+c^2 \rangle^{1/2} / | \langle b^+bc^+c^+ \rangle | - 1 , \tag{4.7}
\]

\[
= \langle b^+b^2 \rangle / | \varphi |^2 - 1 , \tag{4.8}
\]

for the pair coherent state. Note that if the Cauchy-Schwarz inequality were to hold for our quantum system, then $I$ would be positive. Thus negative values of $I$ will arise for strictly quantum states of the field. It is seen from Fig. 4 that there is considerable violation of the Cauchy-Schwarz inequality for our two-photon system. Such violations of the Cauchy-Schwarz inequality can be studied by a suitably designed "two-photon" interference experiment.\(^{18}\)

The squeezing property of the generated fields can be studied by a heterodyne experiment.\(^{19}\) Let us write the total field in the form

\[
E(t) = e^{-i\omega t} \left( \epsilon_1 e^{i\Psi/2} + e^{i\epsilon_2}b + e^{-i\epsilon_3}c \right) ,
\]

where $\Psi$ is the phase shift. The instantaneous intensity can be written as

\[
I(t) = |\epsilon_1|^2 + e^{-i\epsilon_2}\epsilon_e e^{i\Psi/2} + b^+e^{i\Psi/2} + c^+e^{-i\Psi/2} + c.c. + O(b)^2 .
\]

Hence the cosine component of the intensity will be determined by the operator

\[
d = \frac{\left( e^{-i\Psi/2} + b^+e^{i\Psi/2} + H.c. \right)}{2\sqrt{2}} .
\]

For the pair coherent state the variance in $d$ will be

\[
\langle d^2 \rangle - \langle d \rangle^2 = \frac{1}{2} \left( N + \frac{1}{2} (\epsilon_e e^{-i\Psi} + \epsilon^*_e e^{i\Psi}) \right) ,
\]

\[
= \frac{1}{2} (N - \varphi) = \frac{1}{2} S ,
\]

if the phase of the local oscillator is chosen as $\pi$. From Fig. 4 we predict about 50% squeezing over a very wide range of parameters.

In conclusion we have shown how fields grow, starting from quantum noise and how these fields stabilize in a two-photon medium where several competing nonlinear processes occur. We have shown that the generated fields have very strong quantum properties such as squeezing, sub-Poissonian statistics, etc. The resonant two-photon excitations lead to the generation of new types of coherent states of the radiation field. Such pair coherent states are expected to be important in problems that involve simultaneous creation or destruction of two photons belonging to different modes of the radiation field.

ACKNOWLEDGMENTS

This work was completed while one of us (G.S.A.) was visiting the Institut für Theoretische Physik, Technische Universität, Vienna, and Professor Hittmair, Dr. Adam, and Dr. Seke for their kind hospitality at the Institute in Vienna. He is also indebted to the Department of Science and Technology, Government of India, and to Bundesministerium für Wissenschaft and Forschung (Grant No. GZ 70530/29-13/86), Austria, for partially supporting this work.

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7For a review of some of these properties see, for example, D. F. Walls, Nature (London) 306, 141 (1983); C. K. Hong and L.

8Master equation methods for such an adiabatic elimination can be used; see, for example, G. S. Agarwal, *Quantum Optics*, Vol. 70 of *Springer Tracts in Modern Physics*, edited by G. Hohler (Springer, Berlin, 1974).


10We deal only with the lowest-order nonlinearities described by $\chi^{(2)}$. This is sufficient for most experiments, including that of Ref. 2. Considerations of higher-order nonlinearities resulting, say, from $\chi^{(3)}(\omega_1, -\omega_1, \omega_2, -\omega_2, \omega_2)$ will lead to a much more complex master equation.


17This may be compared with the corresponding generation of the oscillator coherent state arising from the coherent pumping and absorption of a single-mode field propagating through the (one-photon) medium. This is described by the master equation

$$\frac{dp}{dt} = -\kappa(a^\dagger \rho - 2a \rho a^\dagger a - \rho a^\dagger a - ig \{a + a^\dagger, \rho\} ,$$

whose steady state solution is $\rho = \langle z | z \rangle$, where $|z\rangle$ is the usual oscillator coherent state with amplitude $-ig/\kappa$.
