## Vector-field quantum model of degenerate four-wave mixing

Seng-Tiong Ho, Prem Kumar, and Jeffrey H. Shapiro
Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 29 January 1986)

A quantum theory of degenerate four-wave mixing is presented in which the atomic medium consists of stationary four-level atoms, each with three degenerate excited states, and the interacting light beams are allowed to be in different states of polarization. This vector-field theory differs from the scalar-field theory of Reid and Walls [Phys. Rev. A 31, 1622 (1985)] in that there are new atomic variables in the Langevin equations which are related to the induced coherence between the upper atomic states. It is found, for the assumed set of atomic levels, that this seemingly different mechanism of four-wave-mixing gain does not circumvent the degrading effect of spontaneous emission on squeezing obtainable via degenerate four-wave mixing. The theory is applied to both forward and backward degenerate four-wave mixing with nearly collinear geometry, and specialized to the case in which the polarization states of the two pump modes are mutually orthogonal. It is found that for both forward and backward configurations, the range of pump intensity for which squeezing can be achieved in the vector-field case is larger than that in the scalar-field case, and the maximum amount of squeezing obtainable at a particular pump detuning is comparable in both cases.

#### I. INTRODUCTION

A squeezed state is a nonclassical state of light having the property that its noise in one electric-field quadrature is less than that of a coherent state. Four-wave mixing (FWM) has been the most extensively studied scheme for its generation. Backward, forward, and intracavity beam configurations of both degenerate and nearly degenerate. FWM have been considered. Slusher et al. have recently reported observing such a state in homodyne detection of light emitted by a single-ended cavity containing the probe and conjugate beams of an intracavity nearly degenerate backward FWM experiment in an atomic beam of sodium.

The first proposal to generate a squeezed state via FWM was made by Yuen and Shapiro<sup>2</sup> who gave a simplified quantum description of the backward degenerate configuration. Describing only the probe beam (PB) and the phase-conjugate beam (PCB) modes by quantummechanical operators, they showed that an appropriate combination mode of the probe and PC beams at the output of the four-wave mixer was in an ideal squeezed state. Using the same approach, Yurke<sup>4</sup> proposed an intracavity beam configuration to enhance the FWM interaction, and Kumar and Shapiro<sup>3</sup> proposed a forward beam geometry, which showed favorable squeezed-state generation characteristics when propagation loss of the probe and PC beams was taken into account phenomenologically. A more realistic theory of degenerate FWM has recently been given by Reid and Walls, who have described the nonlinear medium and the light beams quantum mechanically. By modeling the medium as consisting of two-state atoms, they have shown that propagation loss and spontaneous emission severely limit the amount of squeezing obtainable via degenerate FWM.

The theory of Reid and Walls<sup>7</sup> assumes that all the four interacting modes are in the same state of polarization,

whereas in some experimental situations<sup>8</sup> it is desirable to have different polarization states for different beams. In this paper we present a theory of degenerate FWM in which the atomic medium is assumed to consist of four-level atoms and the interacting beams are allowed to be in different states of polarization.

The general approach we take here is similar to the scalar-field theory of Reid and Walls. In our model the electromagnetic field is treated as consisting of vector waves interacting with vector atomic dipoles. In Sec. II, we start with an atom-field Hamiltonian in which the medium is considered to consist of four-level atoms with three degenerate excited states and a ground state. This would be the case, for example, for the 555.6-nm transition of ytterbium. Each atom is assumed to be coupled to four field modes (two pump beam modes, a probe beam mode, and a PC beam mode) and a thermal field reservoir. We make the usual simplistic assumption that the thermal field reservoirs for different atoms are decoupled from each other, which amounts to neglecting the effect of superradiance. Following Reid and Walls, we first consider a microscopic volume of atoms interacting with the total field. This enables us to take care of wave coupling later with a spatial integration. An equation of motion is obtained for the reduced density operator of this system using the Markov approximation.

In Sec. III, the equation of motion for the reduced density operator is transformed to a c-number equation of motion for the associated distribution function using the usual technique of choosing an operator ordering. With some approximation, this c-number equation is reduced to a Fokker-Planck equation from which a corresponding set of Langevin equations are obtained. Here, our vector-field theory differs from the scalar-field theory of Reid and Walls in that there are new atomic variables in the Langevin equations which are related to the induced coherence between the upper atomic states. In a semiclas-

sical analysis, such an induced coherence (called Zeeman coherence) has been shown to contribute to the FWM gain in the vector-field case. <sup>10</sup> It is found that this seemingly different mechanism of FWM does not circumvent the effect of spontaneous emission with our choice of atomic levels. Nonetheless, the possibility of choosing different field polarization states provides an additional degree of freedom.

In Sec. IV, we obtain equations of motion for the PB and PCB modes by adiabatically eliminating the atomic variables in the Langevin equations. This step is algebraically so involved that the symbolic manipulations had to be done with the use of a computer. The final results, however, are relatively simple and the radiated noise pattern can be physically related to spontaneous emission by an atomic dipole. Coupling of modes arises when the field is considered to interact with the entire medium by an integration over the atomic volume which is done in Sec. V.

In Sec. VI, we consider forward and backward degenerate FWM with nearly collinear geometry and specialize to the case in which the polarization states of the two pump modes as well as the PB and PCB modes are mutually orthogonal. It has been found with a scalar-field theory<sup>7</sup> that propagation loss and spontaneous emission limit the amount of squeezing obtainable at a particular pump detuning and intensity. Here, we show that for both forward and backward degenerate FWM, the range of pump intensity for which squeezing can be achieved in the vector-field case is larger than that in the scalar-field case and the maximum amount of squeezing obtainable at a particular pump detuning is comparable in both cases. We conclude in Sec. VII with a discussion of the differences between the scalar-field and the vector-field results.

### II. ATOM-FIELD MODEL

Let us consider an ensemble of N atoms uniformly distributed in a volume  $V_m$  interacting with four external field modes of the same angular frequency  $\omega$ . Each atom is coupled to a separate thermal field reservoir. The atoms are assumed to be stationary in space and hence Doppler and collisional effects are ignored. Each atom is characterized by four states consisting of three degenerate excited states which are the eigenstates  $|l=1;m=0,\pm1\rangle$  of the total angular momentum operator, denoted separately as

$$|1\rangle, |0\rangle, |-1\rangle,$$
 (2.1a)

and a ground state  $| l = 0, m = 0 \rangle$ , abbreviated as

$$|g\rangle$$
. (2.1b)

In our treatment below, we will assume linearly polarized waves and so it is more convenient to use a set of excited states which are coupled to the linear  $(e_x, e_y, e_z)$  components of the field polarization instead of the circular components. This set of energy eigenstates, which can be obtained by a linear combination of the above states, is as follows:

$$|x\rangle = (|1\rangle + |-1\rangle)/2^{1/2},$$
 (2.2a)

$$|y\rangle = (|1\rangle - |-1\rangle)/2^{1/2},$$
 (2.2b)

$$|z\rangle = |0\rangle. \tag{2.2c}$$

Following the approach of Reid and Walls,<sup>7</sup> we first consider a microscopic volume  $\delta V$  at position r containing  $N_0$  atoms where  $\delta V$  has linear dimensions smaller than a wavelength. The Hamiltonian of this microscopic atomic volume interacting with the field can be expressed in terms of the following collective atomic operators:<sup>11</sup>

$$\widehat{V}_{l} = \sum_{i=1}^{N_0} |g\rangle_{ii} \langle l| , \qquad (2.3a)$$

$$\hat{n}_l = \sum_{i=1}^{N_0} |l\rangle_{ii} \langle l| , \qquad (2.3b)$$

$$\widehat{n}_{\mathbf{g}} = \sum_{i=1}^{N_0} |\mathbf{g}\rangle_{ii} \langle \mathbf{g}| , \qquad (2.3c)$$

$$\hat{n}_d = \left[ \sum_l \hat{n}_l \right] - \hat{n}_g , \qquad (2.3d)$$

where  $l \in \{x,y,z\}$ , i labels the states of the ith atom,  $\{\hat{V}_l\}$  are the collective atomic transition operators,  $\{\hat{n}_l\}$  and  $\hat{n}_g$  are the total occupation operators of the respective atomic levels, and  $\hat{n}_d$  is the total population inversion operator. Under electric-dipole and rotating-wave approximations, the Hamiltonian  $\hat{H}_r$  in the Schrödinger picture can be written as

$$\hat{H}_{r} = \hat{H}_{0} + \hat{H}_{Lr} + \hat{H}_{Rr} , \qquad (2.4)$$

where the free part is

$$\hat{H}_0 = \sum_{i=1}^4 \hbar \omega \hat{a}_j^{\dagger} \hat{a}_j + (\hbar \omega_0/2) \hat{n}_d , \qquad (2.5)$$

and the atom-field interaction term is

$$\hat{H}_{I,\mathbf{r}} = \sum_{l,i} i \hbar [C_{lj}^*(\mathbf{r}) \hat{a}_j^{\dagger} \hat{V}_l - \text{H.c.}], \qquad (2.6)$$

with H.c. denoting the Hermitian conjugate. The thermal field reservoir term consists of a free part and an interacting part

$$\hat{H}_{R,r} = \hat{R} + \hat{V}_r , \qquad (2.7)$$

with the former being

$$\hat{R} = \sum_{i,k} \hbar \omega_k \hat{b}_{ki}^{\dagger} \hat{b}_{ki} , \qquad (2.8a)$$

and the latter given by

$$\hat{V}_{\mathbf{r}} = \sum_{i,l,k} i \hbar \{ [C_{lk}^{\star}(\mathbf{r})\hat{b}_{ki}^{\dagger} | g \rangle_{ii} \langle l | ] - \text{H.c.} \} , \qquad (2.8b)$$

where  $\hat{b}_{ki}$  is the annihilation operator of the reservoir mode of frequency  $\omega_k$  that is coupled to the *i*th atom. Under the summation signs of the above equations, *j* denotes the sum over the four frequency-degenerate field modes representing the two pump, the probe, and the PC beams; k denotes the sum over the reservoir modes for each atom; l denotes the sum over the atomic excited

states  $l \in \{x,y,z\}$ ; and i denotes the sum over the number of atoms  $i \in \{1,\ldots,N_0\}$ . Also,  $\omega_0$  in the above equations is the atomic resonance frequency, and the coefficients are given by

$$C_{li}(\mathbf{r}) = g\mu_{li} \exp(i\mathbf{k}_i \cdot \mathbf{r}) , \qquad (2.9a)$$

$$g = (\omega_0^2 / 2\omega \hbar \epsilon_0 V)^{1/2} , \qquad (2.9b)$$

where V is the volume of quantization and  $\mu_{lj}$  is the component of the dipole  $\langle l | e \hat{\tau} | g \rangle$  coupled to the jth field mode. If the polarization vector of the jth mode is  $\mathbf{e}_j$ , then

$$\mu_{li} = \langle l \mid e \hat{\mathbf{r}} \mid g \rangle \cdot \mathbf{e}_i . \tag{2.10}$$

The equation of motion for  $\hat{a}_i$  is given by

$$\frac{\partial \hat{a}_{j}}{\partial t} = (1/i\hbar)[\hat{a}_{j}, \hat{H}_{r}] = -i\omega\hat{a}_{j} + \sum_{l} C_{lj}^{*}(\mathbf{r})\hat{V}_{l} . \qquad (2.11)$$

For the entire medium, the equation of motion for  $\hat{a}_j$  can be obtained by considering the total Hamiltonian as in Eq. (2.4) but with a sum over the atoms in the microscopic volume elements at different  $\mathbf{r}$ . The equation of motion for  $\hat{a}_j$  is then given in terms of an integration over the volume  $V_m$  of the medium as

$$\frac{\partial \hat{a}_j}{\partial t} = -i\omega \hat{a}_j + \int_{V_m} d^3 \mathbf{r} \sum_{l} C_{lj}^*(\mathbf{r}) \hat{V}_l(\mathbf{r}) , \qquad (2.12)$$

where  $\hat{V}_l(\mathbf{r})$  in the integrand is a component of the dipole-density operator<sup>12</sup> defined as

$$\hat{V}_{l}(\mathbf{r}) \equiv \hat{V}_{lr}/\delta V$$
,

for  $\hat{V}_{lr}$  a component of the dipole operator of the microscopic volume element  $\delta V$  at r.

With the adiabatic approximation as shown below,  $\hat{V}_{lr}$  can be solved in terms of  $\hat{a}_j$  without the use of Eq. (2.12). Equation (2.12) then allows us to solve for the correlations of the field annihilation operators once we obtain  $\hat{V}_{lr}$ . If we further assume that the thermal field reservoirs for different atoms are uncoupled, we can solve for  $\hat{V}_{lr}$  by just considering the atoms in the volume  $\delta V$  at r and neglect the presence of the other atoms in the medium. This assumption of uncoupled thermal reservoirs neglects the effect of superradiance.

Using  $\hat{H}_r$  given in Eq. (2.4), we can obtain the equation of motion for the density operator of the atom-field system consisting only of a single microscopic volume element. The reduced density operator is of interest; it is obtained by tracing the density operator over the reservoir modes which are assumed initially to be in thermal equilibrium. Using the Markov approximation, we employ the master equation given in Louisell<sup>9</sup> to derive the equation of motion for the reduced density operator  $\hat{\rho}$  in the interaction picture. After transforming away the free part of the pump modes this procedure yields

$$\frac{\partial \hat{\rho}}{\partial t} = (\delta \omega / 2i) [\hat{n}_d, \hat{\rho}] + (1/i\hbar) [\hat{H}_{I,r}, \hat{\rho}] + L(\hat{\rho}) . \quad (2.13)$$

In the above equation  $\delta\omega = \omega_0 - \omega$  and  $L(\hat{\rho})$  is a reservoir

term given by

$$L(\hat{\rho}) = \sum_{i,l} -\{([\hat{V}_{li}^{\dagger}, \hat{V}_{li}\hat{\rho}] + [\hat{\rho}\,\hat{V}_{li}^{\dagger}, \hat{V}_{li}])\gamma(n+1) + ([\hat{V}_{li}, \hat{V}_{li}^{\dagger}\hat{\rho}] + [\hat{\rho}\,\hat{V}_{li}, \hat{V}_{li}^{\dagger}])\gamma n\}, \qquad (2.14)$$

where  $\hat{V}_{li} = |g\rangle_{ii} \langle l|$ ,  $2\gamma$  is the spontaneous decay rate, and n is the average number of thermal photons at frequency  $\omega$ . At optical frequencies  $n \ll 1$  prevails, so we shall consider only the first term in Eq. (2.14). This neglects thermal-noise-induced atomic decay effects.

From Eqs. (2.13) and (2.14), we see that the interaction of the atoms with the field given by  $\hat{H}_{I,r}$  and  $L(\hat{\rho})$  decouples into x, y, and z components. This means, e.g., that if the atoms are excited by an x-polarized wave, there is no emission of waves with y or z polarizations, which need not be the case for atoms with more complicated atomic energy levels. As a result, there is no four-wave mixing gain if the two pump modes are copolarized in a direction orthogonal to those of the PB and PCB modes. Thus, in what follows we shall consider the case in which the two pump modes are orthogonally polarized.

# III. DERIVATION OF THE c-NUMBER FOKKER-PLANCK EQUATION

In order to derive the Fokker-Planck equation for the distribution function associated with the reduced density operator, let us assume for simplicity that all the beams have their polarization vectors lying in the x-y plane. This reduces our four-level atom problem to a three-level one. It turns out that by an argument of symmetry we can recover the solution for waves having arbitrary polarization vectors.

The equation of motion for the reduced density operator  $\hat{\rho}$  in Eq. (2.13) can be transformed into a c-number equation for the associated distribution function  $\rho_c$  by using the standard technique described in Louisell. This c-number transformation is not unique in that it depends on the choice of ordering of the atom and field operators. The operator ordering is arbitrary, but an appropriate choice minimizes the algebra in the calculation. The ordering we use is

$$\hat{Z}^{\dagger} \hat{V}_{y}^{\dagger} \hat{V}_{x}^{\dagger} \hat{n}_{x} \hat{n}_{y} \hat{n}_{g} \hat{V}_{x} \hat{V}_{y} \hat{Z} \hat{a}_{x}^{\dagger} \hat{a}_{y}^{\dagger} \hat{a}_{x} \hat{a}_{y},$$

where  $\hat{Z}$ , defined as

$$\widehat{Z} \equiv \sum_{i=1}^{N_0} |x\rangle_{ii} \langle y| , \qquad (3.1)$$

is an operator related to the Zeeman coherence between the  $|x\rangle$  and  $|y\rangle$  states. The introduction of this operator is necessary to obtain the c-number equation of motion for  $\rho_c$ . We note that our choice of operator ordering makes  $\rho_c$  real. This halves the effort in obtaining the equation of motion, as the second half of the solution can be obtained from the first by complex conjugation. After the c-number transformation, the corresponding c-number variables of the atom and field operators will be

denoted by the same symbols without carets, and with superscript pluses in lieu of daggers.

The equation of motion for  $\rho_c$  contains derivatives of infinite order in the  $n_x$ ,  $n_y$ , and  $n_g$  variables and third or-

der in some other atomic variables. In the limit of large  $N_0$  we can safely truncate by keeping only the first- and second-order derivatives. This reduces the equation of motion for  $\rho_c$  to the following Fokker-Planck equation:

$$\begin{split} \frac{\partial \rho_c}{\partial t} &= \sum_j \left[ -C_{xj}^* V_x \frac{\partial}{\partial a_j} - C_{xj}^* V_y \frac{\partial^2}{\partial a_j \partial Z} - C_{yj}^* V_y \frac{\partial}{\partial a_j} + C_{xj}^* a_j^+ (n_g - n_x) \frac{\partial}{\partial V_x^+} + C_{xj}^* a_j^+ V_x^+ \frac{\partial^2}{\partial V_x^+ \partial V_x^+} - C_{yj}^* a_j Z^+ \frac{\partial}{\partial V_x^+} \right. \\ &\quad - C_{yj}^* a_j^+ V_y \frac{\partial^2}{\partial V_x \partial V_x^+} - C_{yj}^* a_j^+ Z \frac{\partial^2}{\partial n_x \partial V_x^+} + C_{yj}^* a_j^+ Z \frac{\partial^2}{\partial n_y \partial V_x^+} \\ &\quad + (\gamma - i \delta \omega) V_x^+ \frac{\partial}{\partial V_x^+} - C_{xj}^* a_j^+ Z^+ \frac{\partial}{\partial V_y^+} + (n_g - n_y) C_{yj}^* a_j^+ \frac{\partial}{\partial V_y^+} \\ &\quad + C_{yj}^* a_j^+ V_y^+ \frac{\partial^2}{\partial V_y^+ \partial V_y^+} + C_{yj}^* a_j^+ V_x^+ \frac{\partial^2}{\partial V_x^+ \partial V_y^+} + (\gamma - i \delta \omega) V_y^+ \frac{\partial}{\partial V_y^+} \\ &\quad + C_{xj}^* a_j^+ V_x \frac{\partial}{\partial n_x} - C_{xj}^* a_j^+ V_x \frac{\partial}{\partial n_g} + C_{xj}^* a_j^+ V_x \frac{\partial^2}{\partial n_x \partial n_g} \\ &\quad - \frac{1}{2} C_{xj}^* a_j^+ V_x \frac{\partial^2}{\partial n_x \partial n_y} - \frac{1}{2} C_{xj}^* a_j^+ V_x \frac{\partial^2}{\partial n_g \partial n_g} + C_{yj}^* a_j^+ V_y \frac{\partial}{\partial n_y} - C_{yj}^* a_j^+ V_y \frac{\partial}{\partial n_g} + C_{yj}^* a_j^+ V_y \frac{\partial^2}{\partial n_y \partial n_g} \\ &\quad - \frac{1}{2} C_{yj}^* a_j^+ V_y \frac{\partial^2}{\partial n_y \partial n_y} - \frac{1}{2} C_{yj}^* a_j^+ V_y \frac{\partial^2}{\partial n_g \partial n_g} + \gamma n_x \frac{\partial}{\partial n_x} - \gamma n_x \frac{\partial}{\partial n_g} - \gamma n_x \frac{\partial^2}{\partial n_x \partial n_g} \\ &\quad + \frac{1}{2} \gamma n_x \frac{\partial^2}{\partial n_x \partial n_x} + \frac{1}{2} \gamma n_x \frac{\partial^2}{\partial n_g \partial n_g} + \gamma n_y \frac{\partial}{\partial n_y} - \gamma n_y \frac{\partial}{\partial n_g} - \gamma n_y \frac{\partial}{\partial n_g} - \gamma n_y \frac{\partial^2}{\partial n_x \partial n_g} \\ &\quad + \frac{1}{2} \gamma n_y \frac{\partial^2}{\partial n_y \partial n_y} + \frac{1}{2} \gamma n_y \frac{\partial^2}{\partial n_g \partial n_g} + C_{xj}^* a_j^+ V_y \frac{\partial^2}{\partial a_z} + C_{yj}^* a_j^+ V_x \frac{\partial}{\partial a_z} - C_{yj}^* a_j^+ V_x \frac{\partial^2}{\partial n_x \partial Z^+} \\ &\quad + C_{yj}^* a_j^+ V_x \frac{\partial^2}{\partial n_x \partial Z^+} + C_{yj}^* a_j^+ Z^+ \frac{\partial^2}{\partial V_y^+ \partial Z^+} - (n_g - n_x) C_{yj}^* a_j^+ V_x \frac{\partial^2}{\partial V_x^+ \partial Z^+} \\ &\quad + V_z \frac{\partial^2}{\partial n_x \partial Z^+} + V_z \frac{\partial^2}{\partial n_g \partial Z^+} + \gamma V_y^+ \frac{\partial^2}{\partial V_x^+ \partial Z^+} + \gamma Z^+ \frac{\partial^2}{\partial n_x \partial Z^+} + c.c. \left[ \rho_c \right], \end{split}$$

(3.3c)

where c.c. denotes complex conjugate. (In the above equation all the coefficients preceding the partial derivatives should instead follow them.)

Essentially by reading off the coefficients in this Fokker-Planck equation, as described in Louisell, we get the equivalent following set of Langevin equations:

$$\frac{\partial a_j}{\partial t} = C_{xj}^* V_x + C_{yj}^* V_y + f_{a_j} , \qquad (3.3a)$$

$$\frac{\partial a_j^+}{\partial t} = C_{xj} V_x^+ + C_{yj} V_y^+ + f_{a_i^+} , \qquad (3.3b)$$

$$\frac{\partial V_x}{\partial t} = -a_x(n_g - n_x) + a_y Z^+ - (\gamma + i\delta\omega)V_x + f_{V_x},$$

 $\frac{\partial V_y}{\partial t} = -a_y(n_g - n_y) + a_x Z - (\gamma + i\delta\omega)V_y + f_{V_y}, \quad (3.3d)$ 

$$\frac{\partial V_{x}^{+}}{\partial t} = -a_{x}^{+}(n_{g} - n_{x}) + a_{y}^{+} Z - (\gamma - i\delta\omega)V_{x}^{+} + f_{V_{x}^{+}},$$
(3.3e)

$$\frac{\partial V_{y}^{+}}{\partial t} = -a_{y}^{+}(n_{g} - n_{y}) + a_{x}^{+} Z^{+} - (\gamma - i\delta\omega)V_{y}^{+} + f_{V_{y}^{+}},$$
(3.3f)

$$\frac{\partial Z}{\partial t} = -a_x^+ V_y - a_y V_x^+ - 2\gamma Z + f_Z , \qquad (3.3g)$$

$$\frac{\partial Z^{+}}{\partial t} = -a_{x} V_{y}^{+} - a_{y}^{+} V_{x} - 2\gamma Z^{+} + f_{Z^{+}}, \qquad (3.3h)$$

$$\frac{\partial n_{x}}{\partial t} = -a_{x}^{+} V_{x} - a_{x} V_{x}^{+} - 2\gamma n_{x} + f_{n_{x}} , \qquad (3.3i)$$

$$\frac{\partial n_{y}}{\partial t} = -a_{y}^{+} V_{y} - a_{y} V_{y}^{+} - 2\gamma n_{y} + f_{n_{y}} , \qquad (3.3j)$$

where we have defined

$$a_{\mathbf{x}} \equiv \sum_{i} C_{\mathbf{x}j} a_{j} , \qquad (3.4a)$$

$$a_{y} \equiv \sum_{j} C_{yj} a_{j} , \qquad (3.4b)$$

and various Langevin forces have been denoted by subscripts on f. In Eqs. (3.3c)—(3.3j),  $n_g$  is tied to  $n_x$  and  $n_y$  by the completeness relation

$$n_{\mathbf{g}} = N_0 - n_{\mathbf{x}} - n_{\mathbf{y}} . {3.5}$$

The nonzero correlations between the various Langevin forces are

$$\langle f_{a_i}(t)f_Z(t')\rangle = -C_{xj}^* V_y \delta(t-t') , \qquad (3.6a)$$

$$\langle f_{a,+}(t)f_{Z,+}(t')\rangle = -C_{xj}V_{y}^{+}\delta(t-t'),$$
 (3.6b)

$$\langle f_{V+}(t)f_{V+}(t')\rangle = 2a_x^+ V_x^+ \delta(t-t')$$
, (3.6c)

$$\langle f_{V_x}(t)f_{V_x}(t')\rangle = 2a_x V_x \delta(t-t')$$
, (3.6d)

$$\langle f_{V_{\mathbf{v}}}(t)f_{V^{+}}(t')\rangle = (-a_{\mathbf{y}}^{+}V_{\mathbf{y}} - a_{\mathbf{y}}V_{\mathbf{y}}^{+})\delta(t-t')$$
, (3.6e)

$$\langle f_{V_y^+}(t)f_{V_y^+}(t')\rangle = 2a_y^+ V_y^+ \delta(t-t')$$
, (3.6f)

$$\langle f_{V_u}(t)f_{V_u}(t')\rangle = 2a_y V_y \delta(t-t')$$
, (3.6g)

$$\langle f_{V^{+}}(t)f_{V^{+}}(t')\rangle = a_{y}^{+}V_{x}^{+}\delta(t-t')$$
, (3.6h)

$$\langle f_{V_n}(t)f_{V_n}(t')\rangle = a_v V_x \delta(t-t')$$
, (3.6i)

$$\langle f_{n_x}(t)f_{n_x}(t')\rangle = (-a_x^+V_x - a_xV_x^+ + 2\gamma n_x)\delta(t-t') ,$$

(3.6i)

$$\langle f_{n_y}(t)f_{n_y}(t')\rangle = (-a_y^+ V_y - a_y V_y^+ + 2\gamma n_y)\delta(t - t'),$$
(3.6k)

$$\langle f_{V+}(t)f_{Z+}(t')\rangle = a_{y}^{+}Z^{+}\delta(t-t')$$
, (3.61)

$$\langle f_{V_u}(t)f_Z(t')\rangle = a_y Z\delta(t-t')$$
, (3.6m)

$$\langle f_{V_x^+}(t)f_{Z^+}(t')\rangle = [-a_y^+(n_g - n_x) + 2\gamma V_y^+]\delta(t - t')$$
,

(3.6n)

$$\langle f_{V_x}(t)f_Z(t')\rangle = [-a_y(n_g - n_x) + 2\gamma V_y]\delta(t - t'),$$
(3.60)

$$\langle f_{z+}(t)f_{z}(t')\rangle = 2\gamma n_{y}\delta(t-t')$$
, (3.6p)

$$\langle f_{n_x}(t)f_{z+}(t')\rangle = (-2\gamma Z^+ + a_y^+ V_x)\delta(t-t')$$
, (3.6q)

$$\langle f_{n_x}(t)f_Z(t')\rangle = (-2\gamma Z + a_y V_x^+)\delta(t - t'), \qquad (3.6r)$$

$$\langle f_{n_x}(t)f_{Z^+}(t')\rangle = (2\gamma Z^+ - a_y^+ V_x)\delta(t - t')$$
, (3.6s)

$$\langle f_{n_x}(t)f_Z(t')\rangle = (2\gamma Z - a_y V_x^+)\delta(t - t')$$
, (3.6t)

$$\langle f_{n_y}(t)f_{n_g}(t')\rangle\!=\!(-2\gamma n_y\!+\!a_y^{\phantom{\dagger}}V_y\!+\!a_y^{\phantom{\dagger}}V_y^{\phantom{\dagger}})\delta(t-t')\;,$$

(3.6u)

$$\langle f_{n_g}(t)f_{n_g}(t')\rangle = (2\gamma n_x + 2\gamma n_y - a_y^+ V_y - a_x^+ V_x - a_y V_y^+ - a_x V_x^+)\delta(t - t'),$$
 (3.6v)

$$\langle f_{V_{\tau}^+}(t)f_{n_x}(t')\rangle = -a_y^+ Z\delta(t-t')$$
, (3.6w)

$$\langle f_{V_{\nu}}(t)f_{n_{\nu}}(t')\rangle = -a_{\nu}Z^{+}\delta(t-t'), \qquad (3.6x)$$

$$\langle f_{V+}(t)f_{n_v}(t')\rangle = a_v^+ Z\delta(t-t')$$
, (3.6y)

$$\langle f_{V_x}(t)f_{n_y}(t')\rangle = a_y Z^+\delta(t-t')$$
, (3.6z)

$$\langle f_{n_x}(t)f_{n_x}(t')\rangle = (a_x^+V_x + a_xV_x^+ - 2\gamma n_x)\delta(t-t')$$
.

(3.6aa)

# IV. ADIABATIC ELIMINATION OF ATOMIC VARIABLES

The solution to the above Langevin equations can be obtained by first solving for the atomic polarizations  $V_x$  and  $V_y$  using the adiabatic approximation which is briefly reviewed here.

In adiabatic approximation, it is assumed that the fields  $\{a_j\}$  have a characteristic decay rate  $\gamma_R$  which is small compared to the decay rate  $\gamma$  of the atomic variables. This enables us to treat  $\{a_j\}$  as slowly varying in Eqs. (3.3c)—(3.3j). More importantly, it follows that the change in  $a_j$  as given by Eq. (3.3a) is sensitive only to driving frequencies that are small compared with  $\gamma_R$ . Hence, it is sufficient for us to solve for just the low-frequency part of  $V_x$  and  $V_y$  by looking at a coarse-grained time  $\delta t$  such that

$$1/\gamma_R > \delta t >> 1/\gamma . \tag{4.1}$$

Then in Eq. (3.3c), for example, we have

$$\frac{\partial V_x}{\partial t} < \frac{V_x}{\delta t} << \gamma V_x \tag{4.2}$$

so that we can set the time derivative equal to zero. More precisely, this approximation is equivalent to convolving both sides of Eq. (3.3c) with a unit-area pulse of width  $\delta t$ .

Similarly, after setting the time derivatives equal to zero in Eqs. (3.3d)—(3.3j), we solve for  $V_x$  and  $V_y$  in terms of  $\{a_j\}$  simply by algebraic manipulations. Because of the convolution mentioned above, we find that the noise parts of  $V_x$  and  $V_y$  have nonzero correlations for time differences of the order of  $\delta t$ , even though the Langevin forces are delta correlated. For coarse-grained time intervals, however, they can still be regarded as delta correlated.

The equation of motion for  $a_j$  that would be obtained in Sec. V from the solutions of  $V_x$  and  $V_y$  for our system is of the form

$$\frac{\partial a_j}{\partial t} = +i\gamma_I a_j - \gamma_R a_j + D_j + \Gamma_j , \qquad (4.3)$$

where  $\gamma_R, \gamma_I$  are real constants,  $D_j$  is a slowly varying drive term arising from coupling with the other modes, and  $\Gamma_j$  is a noise term contributed by the noise parts of  $V_x$  and  $V_y$ . The solution for  $a_j$  is in general of the form

 $A_j \exp(i\gamma_I't)$ , where  $A_j$  is a slowly varying amplitude and the exponential factor gives a frequency shift. In the degenerate case, the frequency shift comes from the  $i\gamma_I a_j$  term of Eq. (4.3) and hence  $\gamma_I' = \gamma_I$ . The magnitude of  $\gamma_I'$  could be larger than the atomic decay rate  $\gamma$  which would invalidate the assumption that  $a_j$  is slowly varying. This problem can, however, be avoided by making the transformation  $a_j \rightarrow a_j \exp(i\gamma_I't)$ , and similar transformations for all the other field and atomic-polarization variables before performing the adiabatic elimination. This transformation procedure, which then in the degenerate case eliminates the  $i\gamma_I a_j$  term in Eq. (4.3), changes  $\delta \omega$  in Eqs. (3.3c)–(3.3f) to  $\delta \omega - \gamma_I'$ . All the field and atomic-polarization variables below are to be taken as the transformed variables.

Because of its algebraic complexity, the adiabatic elimination is done on a computer with use of a symbolic manipulation program giving the following solutions for  $V_x$  and  $V_y$ :

$$V_{x} = -\frac{N_{0}a_{x}}{\gamma(1+i\delta)P} + G_{V_{x}}, \qquad (4.4a)$$

$$V_y = -\frac{N_0 a_y}{\nu (1 + i\delta) P} + G_{\nu_y}$$
, (4.4b)

where  $\delta$  is the normalized detuning defined as

$$\delta \equiv \frac{(\omega_0 - \omega_{op})}{\gamma} , \qquad (4.5a)$$

with

$$\omega_{op} = \omega - \gamma_I' , \qquad (4.5b)$$

P is a saturation factor given by

$$P = 1 + \frac{2(a_x a_x^+ + a_y a_y^+)}{\gamma^2 (1 + \delta^2)} , \qquad (4.5c)$$

and  $G_{V_x}$ ,  $G_{V_y}$  are the noise parts involving terms linear in  $f_{V_x}$ ,  $f_{V_y}$ ,  $f_{Z}$ ,  $f_{n_x}$ ,  $f_{n_y}$ ,  $f_{V_x^+}$ ,  $f_{V_y^+}$ , and  $f_{Z^+}$ . The correlation equations for  $G_{V_x}$  and  $G_{V_y}$  are complicated and are not presented here. However, we note that they are asymmetric in the x and y parameters. Using Eqs. (4.4a) and (4.4b) in Eq. (3.3a), we obtain the following stochastic equation of motion for  $a_i$ :

$$\frac{\partial a_{j}}{\partial t} = -i\gamma'_{I}a_{j} - \frac{N_{0}(C_{xj}^{*}a_{x} + C_{yj}^{*}a_{y})}{\gamma(1+i\delta)P} + G_{a_{jr}}, \qquad (4.6)$$

where  $G_{a_{ir}}$  is a Langevin force given by

$$G_{a_{jr}} = f_{a_j} + C_{xj}^* G_{V_x} + C_{yj}^* G_{V_y} . (4.7)$$

Below, we specialize to the case in which the PB and one of the pump modes have  $\mathbf{e}_x$  polarization, whereas the other pump and PCB modes have  $\mathbf{e}_y$  polarization. We also assume that the two pump modes are nondepleted having equal complex amplitudes  $\varepsilon$ . Let the PB and PCB modes be  $a_3$  and  $a_4$ , respectively, then  $a_x$  and  $a_y$  in Eq. (3.4) are

$$a_{\mathbf{x}} = C_{\mathbf{x}} \mathbf{1} \varepsilon + C_{\mathbf{x}} \mathbf{3} a_{\mathbf{3}} , \qquad (4.8a)$$

$$a_{\nu} = C_{\nu 2} \varepsilon + C_{\nu 4} a_4 , \qquad (4.8b)$$

where, because of the assumed polarization states of the fields, we have taken

$$C_{x2} = C_{x4} = C_{v1} = C_{v3} = 0$$
, (4.9)

and the nonzero C's of Eq. (2.9a) are given by

$$C_{\mathbf{x}1} = C \exp(i\mathbf{k}_1 \cdot \mathbf{r}) , \qquad (4.10a)$$

$$C_{x3} = C \exp(i\mathbf{k}_3 \cdot \mathbf{r}) , \qquad (4.10b)$$

$$C_{v2} = C \exp(i \mathbf{k}_2 \cdot \mathbf{r}) , \qquad (4.10c)$$

$$C_{v4} = C \exp(i \mathbf{k}_4 \cdot \mathbf{r}) , \qquad (4.10d)$$

$$C = g\mu_0 , \qquad (4.10e)$$

with

$$\langle x | e\hat{x} | g \rangle = \langle y | e\hat{y} | g \rangle \equiv \mu_0$$
. (4.10f)

Using Eq. (4.6), we get the following stochastic equations of motion for  $a_3$  and  $a_4$ :

$$\frac{\partial a_3}{\partial t} = -i\gamma'_1 a_3 + \left[ \frac{-N_0 C^* a_x}{\gamma (1+i\delta) P} + \Gamma_{a_{3r}} \right] \exp(-i\mathbf{k}_3 \cdot \mathbf{r}) ,$$
(4.11a)

$$\frac{\partial a_4}{\partial t} = -i\gamma'_1 a_4 + \left[ \frac{-N_0 C^* a_y}{\gamma (1+i\delta) P} + \Gamma_{a_{4r}} \right] \exp(-i\mathbf{k}_4 \cdot \mathbf{r}) ,$$
(4.11b)

where  $\Gamma_{a_{3r}} = G_{a_{3r}} \exp(i\mathbf{k}_3 \cdot \mathbf{r})$  and  $\Gamma_{a_{4r}} = G_{a_{4r}} \exp(i\mathbf{k}_4 \cdot \mathbf{r})$ . P can be expressed in terms of the total field intensity I and the detuned saturation intensity  $I_c$  by

$$P = (1 + I/I_s) , (4.12a)$$

where

$$I = (a_x a_x^+ + a_y a_y^+) / |C|^2, (4.12b)$$

$$I_s = I_{s0}(1+\delta^2)$$
, (4.12c)

and

$$I_{s0} = \gamma^2 / 2 |C|^2. \tag{4.12d}$$

The noise correlations for the Langevin forces are given by

$$\langle \Gamma_{a_{ir}}(t)\Gamma_{a_{jr}}(t')\rangle = D_{a_ia_j}\delta(t-t') , \qquad (4.13)$$

with

$$D_{a_3 a_3} = -\frac{a_x a_x (C^*)^2 N_0}{|C|^2 \gamma I_s P^3 (1 + \delta^2)^2} [(1 - i\delta)^3 + (I/I_{s0})^2/2],$$
(4.14a)

$$D_{a_3 a_3^+} = \frac{a_x a_x^+ N_0 (I/I_{s0})}{\gamma I_s P^3 (1 + \delta^2)^2} [2 + (I/I_{s0})/2], \qquad (4.14b)$$

$$D_{a_4 a_4} = -\frac{a_y a_y (C^*)^2 N_0}{\mid C\mid^2 \gamma I_s P^3 (1 + \delta^2)^2} [(1 - i\delta)^3 + (I/I_{s0})^2/2] \; ,$$

(4.14c)

$$D_{a_4 a_4^+} = \frac{a_y a_y^+ N_0}{\gamma I_s P^3 (1 + \delta^2)^2} \{ (I/I_{s0})[2 + (I/I_{s0})/2] \} ,$$
(4.14d)

$$D_{a_3 a_4} = \frac{-a_x a_y (C^*)^2 N_0}{|C|^2 \gamma I_s P^3 (1 + \delta^2)^2} [(1 - i\delta)^3 + (I/I_{s0})^2/2],$$
(4.14e)

and

$$D_{a_3a_4^+} = \frac{a_x a_y^+ N_0}{\gamma I_s P^3 (1 + \delta^2)^2} \{ (I/I_{s0}) [2 + (I/I_{s0})/2] \} .$$

(4.14f)

Unlike the correlations for  $G_{V_x}$  and  $G_{V_y}$ , the above correlations show x-y symmetry. The recovery of the x-y symmetry is due to the nonzero correlations of  $f_{a_j}$  and  $f_{a_j^+}$  with  $f_Z$  and  $f_{Z^+}$  as given in Eqs. (3.6a) and (3.6b), respectively, which are themselves asymmetric in the x-y parameters ( $f_{a_j}$  and  $f_{a_j^+}$  are uncorrelated with  $f_{Z^+}$  and  $f_{Z}$ , respectively). This recovery of symmetry is expected because the correlations of Eq. (4.13) can be physically related to spontaneous emission noise whereas the correlations of  $G_{V_x}$  and  $G_{V_y}$  in Eq. (4.4) have no direct physical meaning, i.e., they do not completely describe the correlations of  $V_x$  and  $V_y$  because of the noise terms present in  $a_x$  and  $a_y$ . We can generalize these correlations further to waves having arbitrary states of polarization, which is done in the Appendix.

## V. COUPLED-MODE EQUATIONS

The equations of motion for the field-annihilation operators interacting with the entire medium can be obtained by using the total Hamiltonian as mentioned in Sec. II. The c-number equivalent equation of motion for  $a_i$  (j=3,4) corresponding to the operator Eq. (2.12) is

$$\frac{\partial a_{j}}{\partial t} = -i\gamma'_{I}a_{j} 
+ \int_{V_{m}} (d^{3}\mathbf{r}/\delta V) \left[ \frac{-N_{0}(C_{xj}^{*}a_{x} + C_{yj}^{*}a_{y})}{[\gamma(1+i\delta)P]} \right] 
+ \Gamma_{a_{jr}} \exp(-i\mathbf{k}_{j} \cdot \mathbf{r}) .$$
(5.1)

To derive the coupled-mode equations, we expand 1/P in Eq. (5.1) to first order in the amplitudes of the PB and PCB fields  $a_3$  and  $a_4$ :

$$1/P = (1/P_0) \{ 1 - [a_3 \varepsilon^+ \exp(i \mathbf{k}_3 \cdot \mathbf{r} - i \mathbf{k}_1 \cdot \mathbf{r})$$

$$+ a_3^+ \varepsilon \exp(i \mathbf{k}_1 \cdot \mathbf{r} - i \mathbf{k}_3 \cdot \mathbf{r})$$

$$+ a_4 \varepsilon^+ \exp(i \mathbf{k}_4 \cdot \mathbf{r} - i \mathbf{k}_2 \cdot \mathbf{r})$$

$$+ a_4^+ \varepsilon \exp(i \mathbf{k}_2 \cdot \mathbf{r} - i \mathbf{k}_4 \cdot \mathbf{r})] / I_s P_0 \} ,$$

$$(5.2a)$$

$$P_0 = (1 + 2I_n/I_s) , (5.2b)$$

where  $I_p = |\epsilon|^2$ . The integration in Eq. (5.1) is to be done over the entire volume  $V_m$  of the medium. For simplicity, we take  $V_m$  to be equal to the volume of quantization V in Eq. (2.9b) which is equivalent to assuming an infinite medium because of the periodic boundary conditions imposed on the field. In the spatial integration, the exponential terms of Eq. (5.2a) make no contribution unless they satisfy the phase-matching condition

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{k}_4$$
 (5.3)

Unlike the scalar-field case, this integration is straightforward as  $P_0$  is not a function of  $\mathbf{r}$ . Using a similar procedure for the conjugate field mode  $a_j^+$ , we get the following coupled-mode equations for  $a_3$  and  $a_4^+$ :

$$\frac{\partial a_3}{\partial t} = i \left( \gamma_I - \gamma_I' \right) a_3 - \gamma_R a_3 + \chi a_4^+ + \Gamma_3(t) , \qquad (5.4a)$$

$$\frac{\partial a_4^+}{\partial t} = -i (\gamma_I - \gamma_I') a_4^+ - \gamma_R a_4^+ + \chi^* a_3 + \Gamma_4^+(t) , \quad (5.4b)$$

with

$$\gamma_I = 2C'(1 + I_n/I_s)\delta/(1 + \delta^2)P_0^2$$
, (5.5a)

$$\gamma_R = 2C'(1 + I_n/I_s)/(1 + \delta^2)P_0^2$$
, (5.5b)

$$\chi = 2C'(\varepsilon^2/I_c)/(1+i\delta)P_0^2$$

$$\equiv \chi_R + i\chi_I , \qquad (5.5c)$$

$$2C' = |C|^2 N/\gamma , \qquad (5.5d)$$

where N in Eq. (5.5d) is the total number of atoms in the medium. The coupled-mode equations for  $a_3^+$  and  $a_4$  can be obtained by interchanging subscripts 3 and 4. Following the discussion after Eq. (4.3),  $\gamma_I'$  should be equal to  $\gamma_I$  in Eqs. (5.4) so that the terms proportional to  $\gamma_I - \gamma_I'$  are zero. The equations of motion for the pump modes are similar to Eqs. (5.4) but with a different  $\gamma_I$  coefficient given by

$$\gamma_I(\text{pump}) = 2C'(1 + 2I_p/I_s)\delta/(1 + \delta^2)P_0^2$$
. (5.5e)

Due to this difference in  $\gamma_I$  coefficients it is impossible to make a transformation that removes the  $(\gamma_I - \gamma_I')$ -proportional terms not only from Eqs. (5.4) but also from the pump-mode equations. This problem can, however, be solved by considering modified FWM geometries<sup>13</sup> and in what follows we will assume that the  $(\gamma_I - \gamma_I')$ -proportional terms are zero. The integrated noise forces  $\Gamma_3$ ,  $\Gamma_4^+$  in Eqs. (5.4), and  $\Gamma_3^+$ ,  $\Gamma_4$  which appear in the equations for  $a_3^+$  and  $a_4$ , are given by

$$\Gamma_{j}(t) = \int_{V} (d^{3}\mathbf{r}/\delta V) \Gamma_{a_{jr}}(t) \exp(-i\mathbf{k}_{j} \cdot \mathbf{r}) , \qquad (5.6a)$$

$$\Gamma_j^+(t) = \int_V (d^3 \mathbf{r}/\delta V) \Gamma_{a_{ir}^+}(t) \exp(i\mathbf{k}_j \cdot \mathbf{r}) . \qquad (5.6b)$$

The noise forces  $\Gamma_{a_{jr}}$  and  $\Gamma_{a_{jr}^+}$  in Eqs. (5.6) at different r can be shown to be uncorrelated due to the assumption of uncoupled thermal reservoirs. Thus we have from Eq. (4.13)

$$\frac{\langle \Gamma_{a_{ir}}(t)\Gamma_{a_{jr'}}(t')\rangle}{\delta V \delta V} = (D_{a_i a_j}/\delta V)\delta(t-t')\delta(\mathbf{r}-\mathbf{r}'), \qquad (5.7a)$$

$$\frac{\langle \Gamma_{a_{ir}}(t)\Gamma_{a_{jr'}}(t')\rangle}{\delta V \delta V} = (D_{a_i a_j^+}/\delta V)\delta(t-t')\delta(\mathbf{r}-\mathbf{r}') . \quad (5.7b)$$

To determine the correlations for  $\Gamma_j$  and  $\Gamma_j^+$ , consider, for example,

$$\langle \Gamma_{i}(t)\Gamma_{j}(t')\rangle$$

$$= \int_{V} \int_{V} d^{3}\mathbf{r} d^{3}\mathbf{r}' \exp(-i\mathbf{k}_{i} \cdot \mathbf{r}) \exp(-i\mathbf{k}_{j} \cdot \mathbf{r}')$$

$$\times (D_{a_{i}a_{j}}/\delta V)\delta(t-t')\delta(\mathbf{r}-\mathbf{r}') , \qquad (5.8a)$$

and

$$\begin{split} \left\langle \, \Gamma_i(t) \Gamma_j^{\,+}(t') \, \right\rangle \\ &= \int_V \int_V d^3 \mathbf{r} \, d^3 \mathbf{r}' \mathrm{exp}(-i \mathbf{k}_i \cdot \mathbf{r}) \mathrm{exp}(i \mathbf{k}_j \cdot \mathbf{r}') \\ &\times (D_{a_i a_i^{\,+}} / \delta V) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}') \; . \end{split} \tag{5.8b}$$

Since the dominant term in  $D_{a_3a_4} \propto \exp(i\mathbf{k}_1\cdot\mathbf{r})\exp(i\mathbf{k}_2\cdot\mathbf{r})$ , we find that the anomalous correlator  $\langle \Gamma_3(t)\Gamma_4(t')\rangle$  is nonzero because of phase matching. The normal correlator  $\langle \Gamma_3(t)\Gamma_3^+(t')\rangle$  is nonzero too, but not because of phase matching as  $D_{a_3a_3^+}$  is independent of  $\mathbf{r}$ . We also find that  $\langle \Gamma_3(t)\Gamma_4^+(t')\rangle$  is zero due to phase mismatching. The nonzero correlations, therefore, are

$$\langle \Gamma_3(t)\Gamma_4(t')\rangle = R^*\delta(t-t')$$
, (5.9a)

$$\langle \Gamma_3^+(t)\Gamma_4^+(t')\rangle = R\delta(t-t')$$
, (5.9b)

$$\langle \Gamma_3(t)\Gamma_3^+(t')\rangle = \langle \Gamma_4(t)\Gamma_4^+(t')\rangle = \Lambda\delta(t-t'),$$
 (5.9c)

where

$$R^* = R_R - iR_I , \qquad (5.10a)$$

$$R_R = -\left[2C'(\epsilon^2/I_s)/(1+\delta^2)^2 P_0^3\right]$$

$$\times \left[(1-3\delta^2) + 2(I_p/I_s)^2(1+\delta^2)^2\right] , \qquad (5.10b)$$

$$R_I = -\left[2C'(\epsilon^2/I_s)/(1+\delta^2)^2 P_0^3\right](3\delta-\delta^3) , \qquad (5.10c)$$

$$\Lambda = \left[2(I_p/I_s)^2 2C'/(1+\delta^2) P_0^3\right]\left[2 + (I_p/I_s)(1+\delta^2)\right] . \qquad (5.10d)$$

The coupled-mode Eqs. (5.4) have been derived assuming that the fields are polarized only in the x-y plane, which is possible only if their k vectors are almost collinear. A more general analysis can, however, be performed for waves having arbitrary polarization states by means of the correlations given in the Appendix.

A preliminary calculation shows that in the general case, the polarization state of the PB or the PCB mode could undergo rotation as it propagates through the medium. This is because the component of the PB or the PCB mode with polarization parallel to the resultant polarization of the two pump modes sees a lower loss than the component with polarization which is perpendicular to that resultant. This loss asymmetry is due to saturation; the medium acts like a polarizer whose axis is determined by the resultant polarization of the two pump modes. In addition, for the case of forward FWM, the PB mode couples to the PCB mode, which in turn couples to the mode with polarization state orthogonal to the PB mode. This

mechanism also gives rise to polarization rotation of the PB and PCB modes. Because of these complications, we only consider nearly collinear cases in this paper.

#### VI. DEGENERATE FOUR-WAVE MIXING

In this section we apply the vector-field theory developed in Secs. II—V to the forward and backward degenerate FWM configurations for comparison with the scalar-field theory of Reid and Walls. As explained at the end of Sec. V, we restrict our attention to the geometry in which all the beams are nearly collinear.

## A. Forward degenerate FWM

As shown in Fig. 1, let us consider the case in which the two pump beams have mutually orthogonal states of polarization. For a probe beam copolarized with one of the pump beams, a PC beam with polarization vector orthogonal to that of the PB is generated via degenerate FWM in a direction which satisfies the phase-matching condition of Eq. (5.3).<sup>3,10</sup> Equation (5.4) describes the evolution of the coupled PB and PCB modes.

In our idealized theory, it is assumed that the medium is of infinite extent. All the modes, which are +z directed plane-waves, are assumed to be in coherent states at z=0 and observations are made at z=L. This provides an appropriate model for a medium of finite length L when the effect of reflection at the medium boundaries can be neglected.

In order to solve for spatial propagation, we make the  $t \rightarrow z/c$  transformation where c is the speed of light in the medium. Such a transformation, although not rigorous, has been used in previous works.<sup>3-5,7,8</sup> Further justification for this transformation can be provided by a multimode analysis of nondegenerate FWM.<sup>14</sup>

The propagation equations for the PB and PCB modes obtained in this way are

$$\frac{\partial a_3(z)}{\partial z} = -\alpha a_3(z) + \widetilde{\chi} a_4^+(z) + \Gamma_3(z) , \qquad (6.1a)$$

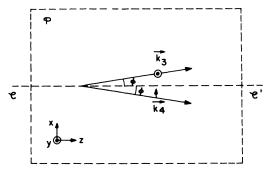


FIG. 1. Nearly collinear forward degenerate FWM geometry. PB and PCB wave vectors  $\mathbf{k}_3$  and  $\mathbf{k}_4$ , respectively, lie in plane  $\mathscr P$  and the pump-beam wave vectors are obtained by rotating plane  $\mathscr P$  along  $\mathscr C\mathscr C'$  by  $\pi/2$  rad. The pump beams are assumed orthogonally polarized with one polarization vector perpendicular to  $\mathscr P$ .

$$\frac{\partial a_4^+(z)}{\partial z} = -\alpha a_4^+(z) + \tilde{\chi}^* a_3(z) + \Gamma_4^+(z) , \qquad (6.1b)$$

where

$$\alpha = \gamma_R / c$$
, (6.2a)

$$\widetilde{\chi} = \chi/c = \widetilde{\chi}_R + i\widetilde{\chi}_I$$
, (6.2b)

$$\langle \Gamma_3(z)\Gamma_4(z')\rangle = \widetilde{R}^*\delta(z-z'),$$
 (6.2c)

$$\langle \Gamma_3^+(z)\Gamma_4^+(z')\rangle = \widetilde{R}\delta(z-z')$$
, (6.2d)

$$\langle \Gamma_3(z)\Gamma_3^+(z')\rangle = \langle \Gamma_4(z)\Gamma_4^+(z')\rangle = \widetilde{\Lambda}\delta(z-z')$$
, (6.2e)

$$\widetilde{R} = R/c = \widetilde{R}_R + i\widetilde{R}_I , \qquad (6.2f)$$

and

$$\widetilde{\Lambda} = \Lambda/c$$
 (6.2g)

We can solve for  $a_3(L)$  and  $a_4^+(L)$  using the standard method<sup>3</sup> to obtain

$$a_3(L) = T(L)a_3(0) + r(L)a_4^+(0) + G_3(L)$$
, (6.3a)

$$a_{4}^{+}(L) = T(L)a_{4}^{+}(0) + r^{*}(L)a_{3}(0) + G_{4}^{+}(L)$$
, (6.3b)

where

$$G_3(L) = \int_0^L [T(L-z')\Gamma_3(z') + r(L-z')\Gamma_4^+(z')]dz',$$
(6.4a)

$$G_4^+(L) = \int_0^L [T(L-z')\Gamma_4^+(z') + r^*(L-z')\Gamma_3(z')]dz', \qquad (6.4b)$$

$$T(z) = \exp(-\alpha z) \cosh(|\widetilde{\chi}|z)$$
, (6.4c)

$$r(z) = \frac{\widetilde{\chi}}{|\widetilde{\chi}|} \exp(-\alpha z) \sinh(|\widetilde{\chi}|z) . \tag{6.4d}$$

According to the ideal theory,<sup>3</sup> squeezed states are generated when the PB and PCB waves at the output are combined with a 50% beam splitter. Therefore, we consider the following combination mode:

$$\widehat{e} = [\widehat{a}_3(L) + \exp(i\theta)\widehat{a}_4(L)]/2^{1/2}$$

$$\equiv \widehat{X}_1(\theta) + i\widehat{X}_2(\theta) , \qquad (6.5)$$

where  $\hat{X}_1(\theta)$  and  $\hat{X}_2(\theta)$ , the two quadrature operators of the combination mode, can be measured via homodyne detection.<sup>15</sup> The fluctuation in the quadrature operator  $\hat{X}_1(\theta)$  as given by the variance

$$\begin{split} \delta \widehat{X}_{1}^{2}(\theta) &\equiv \langle [\widehat{X}_{1}(\theta) - \langle \widehat{X}_{1}(\theta) \rangle]^{2} \rangle \\ &= \frac{1}{4} + \frac{1}{4} [\langle a_{3}(L)a_{4}(L) \rangle \exp(i\theta) \\ &+ \langle a_{3}^{+}(L)a_{4}^{+}(L) \rangle \exp(-i\theta) \\ &+ \langle a_{3}^{+}(L)a_{3}(L) \rangle + \langle a_{4}^{+}(L)a_{4}(L) \rangle], \end{split}$$

$$(6.6)$$

is the key quantity of interest;  $\delta \hat{X}_{1}^{2}(\theta) < \frac{1}{4}$  is squeezing.

The calculations for the minimum noise in the quadrature operators of  $\hat{e}$  are identical with those in the scalar-

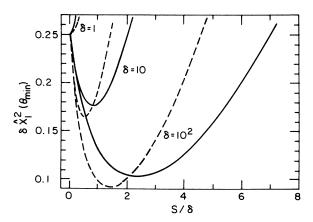


FIG 2. Plots of the minimum quadrature noise variance  $\delta \hat{X}_{1}^{2}(\theta_{min})$  for forward degenerate FWM as a function of the normalized pump intensity S divided by the normalized pump-frequency detuning  $\delta$  for various values of  $\delta$ . Dashed curves are for the scalar-field case of Reid and Walls (Ref. 7) and the solid lines are for our vector-field case.

field case<sup>7</sup> except that the expressions for  $\widetilde{R}$ ,  $\widetilde{\Lambda}$ ,  $\alpha$ , and  $\widetilde{\chi}$  are different. Let  $\theta = \theta_{\min}$  be the phase that gives the minimum value of the variance  $\delta \widehat{X}_{1}^{2}(\theta)$ , then as shown in Ref. 7

$$\delta \widehat{X}_{1}^{2}(\theta_{\min}) = \frac{1}{4} + \frac{1}{4} \left[ \widetilde{\Lambda} \mid \widetilde{\chi} \mid -(\widetilde{R}_{R}\widetilde{\chi}_{R} - \widetilde{R}_{I}\widetilde{\chi}_{I}) \right] / \left[ \mid \widetilde{\chi} \mid (\alpha + \mid \widetilde{\chi} \mid) \right].$$
(6.7)

In Fig. 2 we plot the minimum quadrature noise variance  $\delta \hat{X}_{1}^{2}(\theta_{\min})$  as a function of  $S/\delta$ , where  $S=I_{p}/I_{s0}$ , for various pump-frequency detunings  $\delta$ . For the purpose of comparison, the minimum quadrature noise variance at the same pump-frequency detuning for the scalar-field case is also plotted.

## B. Backward degenerate FWM

In backward degenerate FWM, the two pump beams counterpropagate as shown in Fig. 3. We again assume that the polarization vectors of the two pump beams are mutually orthogonal. Due to degenerate FWM, a PB copolarized with one of the pump beams generates a PC beam which counterpropagates to the PB with its polarization vector perpendicular to that of the PB.<sup>2,10</sup> The spatial propagation equation for the PB mode  $a_3(z)$  is obtained by making the  $t \rightarrow z/c$  transformation, whereas because of the counterpropagating beam geometry, the transformation for the PCB mode  $a_4(z)$  is  $t \rightarrow -z/c$ . Once again, apart from the differences in the expressions for  $\widetilde{R}$ ,  $\widetilde{\Lambda}$ ,  $\alpha$ , and  $\widetilde{\chi}$ , the calculations for the noise variance in the quadrature components of the combination mode are similar to those in the scalar-field case and we omit the details.

In Fig. 4 we plot the minimum quadrature noise variance  $\delta \hat{X}_{1}^{2}(\theta_{\min})$  as a function of S for both the vector- and

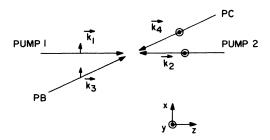


FIG. 3. Nearly collinear backward degenerate FWM geometry. Pump beams are assumed orthogonally polarized as indicated.

the scalar-field cases. We have assumed  $\delta = 100$  and  $\alpha_0 L \equiv 2C'L/c = 10^4$ , where  $\alpha_0$  is the on-resonance unsaturated loss coefficient.

#### VII. DISCUSSION

We have developed a theory of degenerate FWM in which the atomic medium consists of stationary four-level atoms. This allows the interacting beams to have different states of polarization. Our theory reduces to the scalar-field theory of Reid and Walls<sup>7</sup> when all the interacting beams are in the same state of polarization. As in the scalar-field theory, we find in our vector-field theory that the maximum amount of squeezing achievable via both forward and backward FWM is limited by propagation loss due to absorption in the medium and by spontaneous emission from the excited states of the four-level atoms. From Figs. 2 and 4 we see that for both forward and backward FWM, respectively, the range of pump intensity for which squeezing occurs in the vector-field case is larger than that in the scalar-field case, whereas the maximum amount of squeezing at a particular pump de-

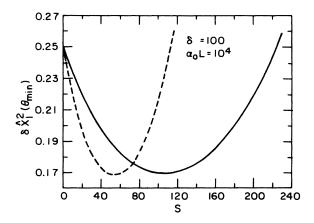


FIG. 4. Plots of minimum quadrature noise variance  $\delta \hat{X}_{i}^{2}(\theta_{\min})$  for backward degenerate FWM as a function of the normalized pump intensity S. Dashed curve is for the scalar-field case of Reid and Walls (Ref. 7) and the solid line is for our vector-field case.

tuning is comparable in both cases with the maximum occurring at a higher pump intensity in the former case.

To understand the origin of these differences we first note that significant squeezing occurs only when the atoms are weakly saturated, i.e., when  $I/I_s \ll 1$ . This is because in this limit, the contribution of spontaneous emission from the excited states as determined by the normal correlators of Eq. (6.2e) and as given by  $\Lambda$  in Eq. (5.10d) is negligible. Comparing the expressions for  $\chi$ , R, and  $\Lambda$  [Eqs. (5.5) and (5.10)] in this limit, we find that the first two are linearly proportional whereas the last depends quadratically on the pump intensities. Also, in both forward and backward configurations, the magnitudes of X and R are smaller by a factor of 2 in the vector-field case than those in the scalar-field case.<sup>7,10</sup> This factor-of-2 difference in  $\chi$  between the scalar-field case and the vector-field case as considered in this paper arises even when the light fields are treated classically. As shown in Ref. 10, the difference originates in the comparative strengths of various gratings leading to the nonlinear coupling in the two cases. This is not surprising in light of the fact that in the weak-saturation region, where squeezing occurs, quantum-field calculations of atomfield interaction reduce to the classical field results. Therefore, in the descending regions of the curves in Figs. 2 and 4, where  $\Lambda$  is negligible compared with R, the same amount of squeezing is achieved at twice the pump intensities in the former case. The ascending parts of the curves, however, are spontaneous-emission noise limited where due to the quadratic dependence of  $\Lambda$  on  $I/I_s$ , the contribution of spontaneous-emission noise at the same squeezing level is larger in the vector-field case. This explains why in the vector-field case the maximum amount of squeezing achievable is slightly lower than that in the scalar-field case.

#### **ACKNOWLEDGMENT**

This work was supported in part by the National Science Foundation under Grant No. ECS 8415580.

## APPENDIX

The correlations given by Eqs. (4.13) and (4.14) show x-y symmetry which is expected because they can be physically related to spontaneous emission noise. In general these correlations should also be symmetric with respect to the z coordinate because the medium is assumed isotropic in the absence of the applied light field. Let us define a new variable  $a_z$  similar to  $a_x$  and  $a_y$  of Eq. (3.4) as

$$a_z \equiv \sum_j C_{zj} a_j , \qquad (A1)$$

and redefine I of Eq. (4.12b) as

$$I \equiv (a_x a_x^+ + a_y a_y^+ + a_z a_z^+) / |C|^2. \tag{A2}$$

Motivated by the fact that in Eq. (4.14) only the x(y) component of the total field appears in  $D_{a_3a_3}(D_{a_4a_4})$ , we generalize the correlations of the Langevin forces  $\Gamma_{a_i}$  and  $\Gamma_{a_j}$  for the two modes  $a_i$  and  $a_j$  with arbitrary polariza-

tion states  $e_i$  and  $e_i$ , respectively, to

$$\left\langle \left. \Gamma_{a_{\rm jr}}(t) \Gamma_{a_{\rm jr}}(t') \right. \right\rangle = - \frac{(\left. \mathbf{A} \cdot \mathbf{e}_{i} \right.) (\left. \mathbf{A} \cdot \mathbf{e}_{j} \right.) (C^{*})^{2} N_{0}}{\left. \left. \right| C \left. \right|^{2} \gamma I_{s} P^{3} (1 + \delta^{2})^{2}}$$

$$\times [(1-i\delta)^3 + (I/I_{s0})^2/2]\delta(t-t')$$
,

(A3a)

$$\langle \Gamma_{a_{ir}}(t)\Gamma_{a_{jr}^+}(t')\rangle = \frac{(\mathbf{A} \cdot \mathbf{e}_i)(\mathbf{A}^+ \cdot \mathbf{e}_j)N_0}{\gamma I_s P^3 (1 + \delta^2)^2} (I/I_{s0})$$

$$\times [2 + (I/I_{s0})/2]\delta(t-t')$$
, (A3b)

where

$$\mathbf{A} \equiv a_{\mathbf{x}} \mathbf{e}_{\mathbf{x}} + a_{\mathbf{v}} \mathbf{e}_{\mathbf{v}} + a_{\mathbf{z}} \mathbf{e}_{\mathbf{z}} \ . \tag{A4}$$

The vectorial nature of Eq. (A3) can be interpreted geometrically by noting that the net induced atomic dipole moment vector **V** is parallel to the effective total light field vector **A**:

$$\mathbf{V} = V_x \mathbf{e}_x + V_y \mathbf{e}_y + V_z \mathbf{e}_z \propto (a_x \mathbf{e}_x + a_y \mathbf{e}_y + a_z \mathbf{e}_z) = \mathbf{A} .$$
(A5)

Thus the amount of spontaneous-emission noise into each mode is just the vectorial component of the dipole radiation in the direction of the polarization vector of that mode.

(Wiley, New York, 1973).

<sup>&</sup>lt;sup>1</sup>D. F. Walls, Nature 306, 141 (1983), and references therein.

<sup>&</sup>lt;sup>2</sup>H. P. Yuen and J. H. Shapiro, Opt. Lett. 4, 334 (1979).

<sup>&</sup>lt;sup>3</sup>P. Kumar and J. H. Shapiro, Phys. Rev. A 30, 1568 (1984).

<sup>&</sup>lt;sup>4</sup>B. Yurke, Phys. Rev. A 29, 408 (1984).

<sup>&</sup>lt;sup>5</sup>B. Yurke, Phys. Rev. A 32, 300 (1985).

<sup>&</sup>lt;sup>6</sup>R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **55**, 2409 (1985).

<sup>&</sup>lt;sup>7</sup>M. D. Reid and D. F. Walls, Phys. Rev. A 31, 1622 (1985).

<sup>&</sup>lt;sup>8</sup>R. S. Bondurant, P. Kumar, J. H. Shapiro, and M. Maeda, Phys. Rev. A 30, 343 (1984).

<sup>9</sup>W. H. Louisell, Quantum Statistical Properties of Radiation

<sup>&</sup>lt;sup>10</sup>J. F. Lam and R. L. Abrams, Phys. Rev. A 26, 1539 (1982).

<sup>&</sup>lt;sup>11</sup>We denote Hilbert state operators by a caret.

<sup>&</sup>lt;sup>12</sup>Note that the atomic operators for different atoms are different, which is reflected in the r dependence of the dipoledensity operator  $\hat{V}_l(\mathbf{r})$ .

<sup>&</sup>lt;sup>13</sup>P. Kumar (unpublished).

<sup>&</sup>lt;sup>14</sup>S.-T. Ho (unpublished).

<sup>&</sup>lt;sup>15</sup>H. P. Yuen and J. H. Shapiro, IEEE Trans. Inf. Theory IT-26, 78 (1980).