Quantum theory of multiwave mixing. VIII. Squeezed states

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We apply our quantum theory of multiwave mixing to the generation of squeezed states of light via three- and four-wave mixing in cavities and in propagation. We compare the cavity predictions to the experimental results of Slusher *et al.* [Phys. Rev. Lett. **55**, 2409 (1985)], finding reasonably good agreement. The squeezing found there is due to the fact that the number operators are driven by resonance fluorescence, which nearly vanishes for the large detunings chosen, while the combination tone operators are driven by relatively large source terms. We give a physical discussion as to why the spectral quantities outside the cavity are given by those inside multiplied by the cavity linewidth. We have found analytic formulas for the variances for arbitrary propagation distances, detunings, and pump intensities. We derive these formulas both from the density operator and from the Langevin methods. To obtain significant squeezing, the propagation distances must be large compared with the resonant Beer's law length.

I. INTRODUCTION

Since the early days of quantum mechanics, the quantization of the electromagnetic field has been a subject of interest among both theorists and experimentalists. Although it is well known that such a description is necessary to account completely for such phenomena as spontaneous emission and the Lamb shift, only comparatively recently has attention focused on the purely quantum properties of the radiation, independent of a medium. This was first realized with the prediction of photon antibunching in resonance fluorescence by Carmichael and Walls,[1] Cohen-Tannoudji,[2] and Kimble and Mandel[3] and its subsequent verification in the experiments of Mandel *et al.*[4]

More recently another purely quantum property of light has received a large amount of interest—that of squeezed states of light.[5] A squeezed state of the electromagnetic field occurs when the quantum fluctuations in some quadrature phase of the electric field are reduced below the average minimum variance permitted by the uncertainty principle. Reduction of the noise in one quadrature must be simultaneously accompanied by an increase of noise in the orthogonal quadrature since the product of canonically conjugate variances must satisfy the uncertainty minimum. Due to the dependence of squeezing on the phase of the electric field, squeezed states have been predicted to occur in phase-sensitive processes, such as parametric amplification,[6] second-harmonic generation,[7] and four-wave mixing.[8-10] The first successful generation of squeezed states has been reported by Slusher *et al.*[11] using nondegenerate four-wave mixing. Recently three other groups have also succeeded in producing squeezed states. The work of Shelby *et al.*[12] using three-wave mixing in an optical fiber, Maeda *et al.*[13] by means of nondegenerate forward four-wave mixing with a sodium vapor cell, and Kimble *et al.*[14] employing parametric amplification in a nonlinear, nonresonant crystal, who have reported observing squeezing up to 60%. The significant work of these four experimental groups has convincingly demonstrated the reality of squeezing.

This eighth paper in our series on the quantum theory of multiwave mixing applies the theory to the generation of squeezed states by four-wave mixing inside and outside of cavities. Preliminary results were presented in two letters.[15,16] Our theory considers the interaction between one or two strong classical waves and one or two weak quantum waves in two-level media. In the first three papers[17-19] we presented two separate derivations of the theory including averages over inhomogeneous broadening, spatial hole burning, and Gaussian beams. We then applied our theory in the next three papers to the problem of resonance fluorescence in a cavity,[20] the two-photon two-level model,[21] and the effects of quantum noise on modulation spectroscopy.[22] Some of the main results from this work showed how coupling between the vacuum modes alters the spontaneous emission spectrum from a two-level system in a cavity, derived the first analytic expression for the resonance fluorescence spectrum from a two-photon two-level media interacting with a strong pump, including the effects of the dynamic Stark shifts, and finally presented a quantum limit to frequency-modulated spectroscopy. The seventh paper[23] in the series demonstrated how the theory could be converted into the equivalent quantum Langevin picture. This is useful in that it connects our results to other methods.

Squeezing is a quantum property of the radiation and a proper theoretical description must include quantization of the fields being squeezed. The theory we have developed is thus appropriate to study squeezed states generated by three- and four-wave mixing. Because our theory provides a uniform description of resonance fluorescence, saturation spectroscopy, and the semiclassical and quantum theory of three- and four-wave mixing, it is clear that the squeezing we consider has an intricate dependence upon all of these seemingly disparate phenomena. In this paper we show how these features of quantum multiwave mixing affect squeezing.

Reid and Walls have recently generalized their quantum theory of degenerate four-wave mixing to include nondegeneracy.[24,25] Although their approach is quite different from ours, they have shown that their final expressions are in complete agreement with ours, and with the conclusions of paper VII in our series.[23] Some of the results of this paper have also been presented in their work, but such redundancy serves to reenforce the foundations of the theory.

Section II summarizes the basic multiwave mixing equations. Section III applies the theory to squeezing inside and outside of cavities and includes a derivation of the spectral formulas commonly used. Section IV specializes to the fourth-order approximation, which agrees well with the experiments of Slusher *et al.*[11] Section V treats squeezed states generated in propagation.

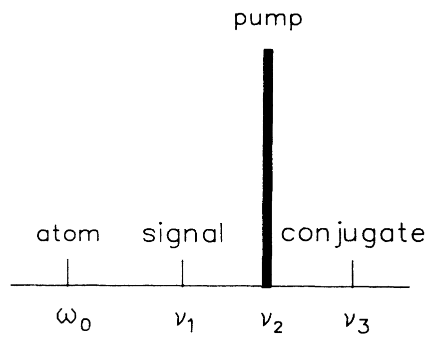

FIG. 1. Spectrum of the modes used in this paper relative to the atomic resonance.

II. SUMMARY OF BASIC EQUATIONS

This section summarizes the theory developed in Refs. 17 and 18 that forms the basis for this paper. We consider three modes of the electromagnetic field with frequencies ν_1, ν_2, and ν_3 that are arbitrarily detuned (consistent with the rotating-wave approximation) from the atomic resonance ω. ν_1 and ν_3 are placed symmetrically on opposite sides of ν_2, that is, $\nu_3=\nu_2+(\nu_2-\nu_1)$ as depicted in Fig. 1. In radians per second our Hamiltonian in the frame rotating at ν_2 is

$$\mathscr{H}=(\omega-\nu_2)\sigma_z-\Delta(a_1^\dagger a_1-a_3^\dagger a_3) +[\mathscr{V}_2U_2\sigma^\dagger+g\sigma^\dagger(a_1U_1+a_3U_3)+\text{H.c.}] \ . \tag{1}$$

In this expression a_j is the annihilation operator for the jth field mode, $U_j=U_j(\mathbf{r})$ is the corresponding spatial mode factor, $\Delta=\nu_2-\nu_1$, g is the atom-field coupling constant, and $\mathscr{V}_2=-\wp\mathscr{E}_2/2\hbar$, where $\wp$ is atomic dipole matrix element and $\mathscr{E}_2$ is the amplitude of mode 2. The rotating-wave approximation has been made and this Hamiltonian is in an interaction picture rotating at the strong-field frequency ν_2. The atomic spin-flip and probability-difference matrices $\sigma^\dagger$ and σ_z are given by

$$\sigma_z=\frac{1}{2}\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix},\quad \sigma^\dagger=\begin{bmatrix}0 & 1\\ 0 & 0\end{bmatrix} \ . \tag{2}$$

We treat mode 2 classically and allow it to be arbitrarily intense. Modes 1 and 3 are quantum fields treated only to second order in amplitude, and cannot by themselves saturate the atomic response. This is an important assumption and limits the applicability of the theory. We define an atom-field density operator $\rho_{a\text{-}f}$ and obtain its time dependence from the standard density operator equation of motion. In this work we consider the simple decay scheme where the upper level 2 decays to the lower level 1. This gives a term

$$\frac{d}{dt}\begin{bmatrix}\rho_{22} & \rho_{21}\\ \rho_{12} & \rho_{11}\end{bmatrix}=\begin{bmatrix}-\Gamma\rho_{22} & -\gamma\rho_{21}\\ -\gamma\rho_{12} & \Gamma\rho_{22}\end{bmatrix}\equiv\Gamma(\rho) \ . \tag{3}$$

The equation of motion for the atom-field density operator is then

$$\dot{\rho}_{a\text{-}f}=-i[\mathscr{H},\rho_{a\text{-}f}]+\Gamma(\rho_{a\text{-}f}) \ . \tag{4}$$

The upper-to-lower-level decay is described by the decay constant Γ $(=1/T_1)$, and the dipole decay described by γ $(=1/T_2)$. For pure spontaneous decay, $\gamma=\Gamma/2$. We calculate the reduced electric field density operator ρ describing the time dependence of the two quantized fields by taking the trace of $\rho_{a\text{-}f}$ over the atomic states. We assume all field amplitudes to vary little during atomic decay times. This allows us to solve the atomic equations of motion in steady state and then to obtain the slowly varying field density operator equation of motion

$$\begin{aligned}\dot{\rho}=&-A_1(\rho a_1a_1^\dagger-a_1^\dagger\rho a_1)-B_1(a_1^\dagger a_1\rho-a_1\rho a_1^\dagger)\\ &+C_1(a_1^\dagger a_3^\dagger\rho-a_3^\dagger\rho a_1^\dagger)+D_1(\rho a_3^\dagger a_1^\dagger-a_1^\dagger\rho a_3^\dagger)\\ &+[(1\leftrightarrow 3)+(\text{adjoint})] \ ,\end{aligned} \tag{5}$$

where $1\leftrightarrow 3$ represents the same previous terms with 1 interchanged with 3, and the coefficients A_1, B_1, C_1, and D_1 are given by

$$A_1=\frac{Ng^2\mathscr{D}_1}{1+I_2\mathscr{L}_2}\left[\frac{I_2\mathscr{L}_2}{2}-\frac{I_2\frac{\gamma}{2}\mathscr{F}\left[\frac{I_2\mathscr{L}_2}{2}\mathscr{D}_1-\mathscr{D}_2^*(1+\Gamma/i\Delta)/2\right]}{1+I_2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right] , \tag{6}$$

$$B_1=\frac{Ng^2\mathscr{D}_1}{1+I_2\mathscr{L}_2}\left[1+\frac{I_2\mathscr{L}_2}{2}-\frac{I_2\frac{\gamma}{2}\mathscr{F}[(1+I_2\mathscr{L}_2/2)\mathscr{D}_1+\mathscr{D}_2^*(1-\Gamma/i\Delta)/2]}{1+I_2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}\right], \tag{7}$$

$$C_1=-\frac{Ng^2\mathscr{D}_1}{1+I_2\mathscr{L}_2}U_1^*U_3^*\frac{2T_1\mathscr{V}_2^2\mathscr{F}\left[\frac{I_2\mathscr{L}_2}{2}\mathscr{D}_3^*-\mathscr{D}_2(1+\Gamma/i\Delta)/2\right]}{1+I_2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}, \tag{8}$$

$$D_1=-\frac{Ng^2\mathscr{D}_1}{1+I_2\mathscr{L}_2}U_1^*U_3^*\frac{2T_1\mathscr{V}_2^2\mathscr{F}[(1+I_2\mathscr{L}_2/2)\mathscr{D}_3^*+\mathscr{D}_2(1-\Gamma/i\Delta)/2]}{1+I_2\mathscr{F}\frac{\gamma}{2}(\mathscr{D}_1+\mathscr{D}_3^*)}, \tag{9}$$

where following the notation of Ref. 17, the complex Lorentzian denominators $\mathscr{D}_n$ are given by

$$\mathscr{D}_n=\frac{1}{\gamma+i(\omega-\nu_n)},$$

the dimensionless Lorentzian $\mathscr{L}_2$ is

$$\mathscr{L}_2=\frac{\gamma^2}{\gamma^2+(\omega-\nu_2)^2},$$

the dimensionless intensity I_2 is

$$I_2=4|\mathscr{V}_2|^2T_1T_2,$$

the dimensionless "population pulsation" term $\mathscr{F}$ is

$$\mathscr{F}=\frac{\Gamma}{\Gamma+i\Delta},$$

and N is the total number of interacting atoms.

The above equations apply both to propagation problems and to situations where the medium is placed inside an optical cavity. In the latter case it is necessary to include a term accounting for cavity losses. Following the analogy from laser theory we designate this as ν/Q. As pointed out by Reid and Walls,[25] the amount of squeezing is highly dependent on the detuning of the field modes from the cavity modes. This is especially important when the pump field ν_2 is highly detuned from the atomic resonance ω since then dispersion dominates absorption. We define $\Delta\Omega=\Omega_2-\nu_2$ to represent this detuning, where Ω_2 is the passive-cavity mode frequency near ν_2. To include these new terms in our theory we replace the B_1 coefficient in Eq. (5) by $B_1+\nu/2Q+i\Delta\Omega$. As we show in Sec. III, this recovers the correct semiclassical equations of motion for the field amplitudes.

III. SQUEEZING INSIDE AND OUTSIDE OF OPTICAL CAVITIES

The equation of motion (5) for the field operator ρ and the expressions for the coefficients A_1 and D_1 are the fundamental equations of our theory. We may use Eq. (5) to obtain the equation of motion for any operator of the quantized fields 1 and 3. This was also done in Ref. 17. For example, the expectation value of the annihilation operator $\langle a_1\rangle$ corresponds to the classical Fourier amplitude for the electric field of mode 1, $\mathscr{E}_1$, and its equation of motion is given by

$$\begin{aligned}\frac{d}{dt}\langle a_1\rangle=\frac{d}{dt}\mathscr{E}_1=\langle a_1\dot\rho\rangle&=\sum_n\langle n\,|\,a_1\dot\rho\,|\,n\rangle\\&=(A_1-B_1-\nu/2Q-i\Delta\Omega)\mathscr{E}_1\\&\quad+(C_1-D_1)\mathscr{E}_3^*,\end{aligned} \tag{10}$$

where $\mathscr{E}_3^*=\langle a_3^\dagger\rangle$. Equation (10) is the semiclassical coupled-mode equation of motion for the field amplitude $\mathscr{E}_1$ and is valid inside a laser cavity. We thus see that the quantity B_1-A_1 is the semiclassical complex absorption coefficient of a weak probe wave in the presence of a strong field, and that the C_1-D_1 term multiplying $\mathscr{E}_3^*$ is the mode-coupling coefficient of phase conjugation and modulation spectroscopy. Each of these coefficients can be derived purely semiclassically.[26]

Of interest for the calculation of squeezing are the equations of motion for the number operator $a_1^\dagger a_1$ and the combination tone operator a_1a_3. These can be obtained in the same manner as Eq. (10):

$$\begin{aligned}\frac{d}{dt}\langle a_1^\dagger a_1\rangle=\langle a_1^\dagger a_1\dot\rho\rangle&=\sum_n\langle n\,|\,a_1^\dagger a_1\dot\rho\,|\,n\rangle\\&=(A_1+A_1^*-B_1-B_1^*-\nu/Q)\langle a_1^\dagger a_1\rangle\\&\quad+(C_1-D_1)\langle a_3^\dagger a_1^\dagger\rangle\\&\quad+(C_1^*-D_1^*)\langle a_3a_1\rangle+A_1+A_1^*,\end{aligned} \tag{11}$$

$$\begin{aligned}\frac{d}{dt}\langle a_1a_3\rangle&=\langle a_3a_1\dot\rho\rangle\\&=\sum_n\langle n\,|\,a_3a_1\dot\rho\,|\,n\rangle\\&=(A_1+A_3-B_1-B_3-\nu/Q-2i\Delta\Omega)\langle a_3a_1\rangle\\&\quad+(C_1-D_1)\langle a_3^\dagger a_3\rangle\\&\quad+(C_3-D_3)\langle a_1^\dagger a_1\rangle+C_1+C_3.\end{aligned} \tag{12}$$

The equations of motion for $\langle a_3^\dagger a_3\rangle$ and $\langle a_1^\dagger a_3^\dagger\rangle$ are given by interchanging the subscripts 1 and 3 in Eq. (11) and taking the complex conjugate of Eq. (12). In free space, no build up of photon number occurs, and $d\langle n_1\rangle/dt = A_1 + A_1^*$. Thus we interpret $A_1 + A_1^*$ as the spectrum of resonance fluorescence and the expression in Eq. (5) plus its complex conjugate is identical to the expression first derived by Mollow.[27] Similarly the inhomogeneous term of Eq. (12), $C_1 + C_3$, is the source contribution for the quantum combination tone $\langle a_3 a_1\rangle$. As we show, this quantity is responsible for squeezing.

The squeezed light results from a linear combination of the sideband amplitudes a_1 and a_3. Figure 2 depicts a simplified diagram of the experimental configuration of Slusher *et al.*[11] The pump laser at frequency ν_2 is reflected off a mirror to form a standing wave through which the atomic beam is passed. The vacuum cavity is servo-locked to the sidemode frequencies ν_1 and ν_3 and encloses the interaction region. Quantum noise builds up in the cavity that, due to the four-wave mixing process, develops correlations between the sidemodes. To detect the squeezing a homodyne detection scheme[11,28] may be used wherein the sidemode fields exiting the cavity are mixed with a local oscillator whose phase is at an angle θ with respect to the pump field ν_2. This homodyne detection permits the direct measurement of the variance for any relative phase shift θ. We average the quantum coefficients A_1 to D_3 over the spatial hole burning caused by the pump field since this is believed to more closely correspond to this experiment. Analytic expressions for these averages are given in Ref. 19. The total amplitude d of the squeezed field is thus given by

$$d = 2^{-1/2}(a_1 e^{i\theta} + a_3^\dagger e^{-i\theta}) . \quad (13)$$

From this operator we define two Hermitian operators d_1 and d_2 as

$$d_1 = \tfrac{1}{2}(d + d^\dagger) \text{ and } d_2 = \frac{i}{2}(d - d^\dagger) . \quad (14)$$

The variance of an operator is defined to be the expectation value of its square minus the square of its expectation value, so the variance Δd_1^2 is

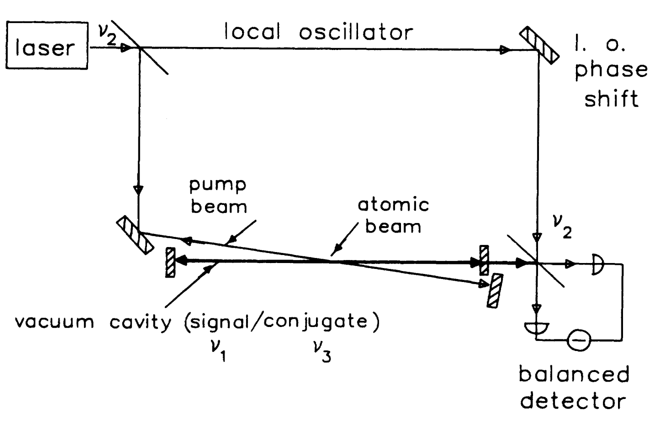

FIG. 2. Diagram for the experiment of Ref. 11.

$$\Delta d_1^2 = \langle d_1^2\rangle - \langle d_1\rangle^2 . \quad (15)$$

Substituting Eqs. (13) and (14) into Eq. (15) and noting that $\langle a_1\rangle = \langle a_3\rangle = \langle a_1^\dagger a_3\rangle = 0$ since we consider the sidebands to arise from the vacuum, we have

$$\Delta d_1^2 = \tfrac{1}{4} + \tfrac{1}{4}[\langle a_1^\dagger a_1\rangle + \langle a_3^\dagger a_3\rangle + (\langle a_1 a_3\rangle e^{2i\theta} + \text{c.c.})] , \quad (16)$$

where we have made use of the commutation relations $[a_1, a_1^\dagger] = [a_3, a_3^\dagger] = 1$. Squeezing occurs whenever Δd_1^2 drops below $\frac{1}{4}$. The expectation values $\langle a_1^\dagger a_1\rangle$ and $\langle a_3^\dagger a_3\rangle$ are in fact the average number of photons in modes 1 and 3, respectively, and consequently are never negative. Thus for squeezing, the quantity in parentheses in Eq. (16) must be negative. Maximum squeezing occurs when $\langle a_1 a_3\rangle e^{2i\theta} = -|\langle a_1 a_3\rangle|$, that is

$$\Delta d_1^2 = \tfrac{1}{4} + \tfrac{1}{4}(\langle a_1^\dagger a_1\rangle + \langle a_3^\dagger a_3\rangle - 2|\langle a_1 a_3\rangle|) . \quad (17)$$

Classically, the correlations between the amplitudes factor and $\langle a_1^\dagger a_1\rangle = \langle a_1^\dagger\rangle\langle a_1\rangle = |\langle a_1\rangle|^2$ and so on. Substituting into Eq. (17), we find in this limit

$$\Delta d_1^2 - \tfrac{1}{4} = \tfrac{1}{4}(|\langle a_1\rangle| - |\langle a_3\rangle|)^2 \geq 0 , \quad (18)$$

and there is no squeezing when this factorization is valid. From Eq. (17), squeezing occurs if $2|\langle a_1 a_3\rangle| > \langle a_1^\dagger a_1\rangle + \langle a_3^\dagger a_3\rangle$. Physically, this means that the sideband fields are more correlated with each other than with themselves, and thus we seek a range of parameters to enhance this coupling.

To calculate the variance we may use the quantum coupled mode equations (11) and (12) to solve for $\langle a_1^\dagger a_1\rangle$, etc., and substitute this into Eq. (17). For the medium inside a cavity a steady state occurs and one sets the time derivatives in Eqs. (11) and (12) equal to zero. The resulting equations may then be solved algebraically. We employed this procedure in our recent work on this subject.[15] The steady-state solutions of Eqs. (11) and (12) are, from Eqs. (9) and (10) of Ref. 15,

$$\begin{aligned}\langle a_1^\dagger a_1\rangle D = {} & A_1[\alpha_3|\alpha_1+\alpha_3|^2 - (\alpha_1+\alpha_3)\chi_1^*\chi_3 + \text{c.c.}] \\ & + A_3(\alpha_1+\alpha_3+\text{c.c.})|\chi_1|^2 \\ & + \chi_1^*(C_1+C_3)[(\alpha_3+\alpha_3^*)(\alpha_1^*+\alpha_3^*) - \chi_1^*\chi_3 \\ & \qquad + \chi_1\chi_3^*] + \text{c.c.} ,\end{aligned} \quad (19)$$

$$\begin{aligned}\langle a_1 a_3\rangle D = {} & -\chi_3[\chi_1^*\chi_3 - \chi_1\chi_3^* \\ & \quad -(\alpha_3+\alpha_3^*)(\alpha_1^*+\alpha_3^*)](A_1+A_1^*) \\ & + (\alpha_1+\alpha_1^*)[\alpha_1^*(\alpha_3+\alpha_3^*)(C_1+C_3) \\ & \qquad -\chi_3(\chi_1^* C_1 + \chi_1^* C_3 + \text{c.c.})] \\ & + (1\leftrightarrow 3) ,\end{aligned} \quad (20)$$

where the denominator D is

$$\begin{aligned}D = {} & (\alpha_1+\alpha_1^*)(\alpha_3+\alpha_3^*)|\alpha_1+\alpha_3|^2 + (\chi_1^*\chi_3 - \chi_1\chi_3^*)^2 \\ & -[(\alpha_1+\alpha_1^*)\chi_1^*\chi_3(\alpha_1+\alpha_3) + (1\leftrightarrow 3) + \text{c.c.}] ,\end{aligned} \quad (21)$$

where the absorption coefficients ($n = 1$ or 3)

$$\alpha_n = B_n - A_n + \nu/2Q + i\Delta\Omega \ , \tag{22}$$

the coupling coefficients

$$\chi_n = C_n - D_n \ , \tag{23}$$

and where $\langle a_3^\dagger a_3 \rangle$ is given by Eq. (19) with 1 interchanged with 3.

It is well known from laser and optical bistability instability theories that due to sidemode gain the steady-state solutions of Eqs. (19)–(21) may not be stable.[29] The stability of these solutions may be found by calculating the eigenvalues of the semiclassical drift matrix A [defined by Eq. (26) below]; the solution is stable if the real parts of each eigenvalue are positive. Another method to check the stability (used by Reid and Walls[25]) is the Hurwitz criteria, which is discussed by Haken.[30] In the figures we present, the solutions have been shown to be stable.

As noted in a series of papers by Collett and Walls[31] and Reid and Walls,[10,25] a detector outside the cavity sees a non-δ-function spectrum around the cavity mode frequencies due to the time-varying fluctuations of the modes about their steady-state values. Steady-state values like Eqs. (19) and (20) are the inverse Fourier transforms of the corresponding spectral quantities for the time $t=0$; that is, they are integrals over the spectra quantities. In contrast, a narrow-band detector might measure peak values larger than these average values. Furthermore, as noted by Collett and Gardiner,[32] in passing from inside a cavity to outside, a spectral quantity must be multiplied by the cavity linewidth. References 10, 25, and 31 use these facts, but give no derivation of the spectral formulas they use and Ref. 32 gives a very complicated derivation about the cavity-linewidth factor. To clarify these important results and to define our notation, we give such a derivation. The spectral formulas result from a generalization of the classical derivation given by Lax,[33] and the cavity linewidth factor follows heuristically from reservoir theory considerations.

Specifically, we define a vector $\alpha^T = (a_1, a_1^\dagger, a_3, a_3^\dagger)$ and from this we obtain the matrix

$$\boldsymbol{\alpha} \times \boldsymbol{\alpha}^T \equiv \boldsymbol{\alpha\alpha} \ , \tag{24}$$

where × denotes the direct product. Using formulas like Eq. (10), we find the equation of motion for $\langle \alpha(t) \rangle$ is given by

$$\frac{d}{dt}\langle \boldsymbol{\alpha}(t) \rangle = -\mathbf{A}\langle \boldsymbol{\alpha}(t) \rangle \ , \tag{25}$$

where the drift matrix $\mathbf{A}$ is

$$\mathbf{A} = \begin{bmatrix} \alpha_1 & 0 & 0 & -\chi_1 \\ 0 & \alpha_1^* & -\chi_1^* & 0 \\ 0 & -\chi_3^* & \alpha_3 & 0 \\ -\chi_3 & 0 & 0 & \alpha_3^* \end{bmatrix} . \tag{26}$$

To obtain the low-frequency spectra of the various mode correlations, we need to calculate the Fourier transform of the corresponding two-time correlations. We thus define the spectral matrix

$$\mathscr{S}(\delta) = \int_{-\infty}^{\infty} e^{-i\delta t} \langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle dt \ , \tag{27}$$

where $\boldsymbol{\alpha}(0)$ means that $\boldsymbol{\alpha}$ is evaluated at a time sufficient for steady state to occur. We determine the equation of motion for $\langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle$ with the help of the quantum regression theorem. According to this theorem, a two-time average like $\langle a_1^\dagger(t) a_1(0) \rangle$ obeys the same equation of motion as the single time average like $\langle a_1^\dagger \rangle$. For the general two-time average matrix, the quantum regression theorem is

$$\frac{d}{dt}\langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle = -\mathbf{A}\langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle \ . \tag{28}$$

The formal solution to Eq. (28), for $t>0$, is

$$\langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle = e^{-\mathbf{A}t} \langle \boldsymbol{\alpha}(0)\boldsymbol{\alpha}(0) \rangle \ , \tag{29}$$

where $e^{-\mathbf{A}t}$ is defined from its power series. Similarly

$$\langle \boldsymbol{\alpha}(0)\boldsymbol{\alpha}(t) \rangle = \langle \boldsymbol{\alpha}(0)\boldsymbol{\alpha}(0) \rangle e^{-\mathbf{A}^T t} \ . \tag{30}$$

Breaking Eq. (27) into two time domains $0 \to \infty$ and $-\infty \to 0$, and using stationarity in the latter, we find

$$\mathscr{S}(\delta) = \int_0^{\infty} e^{-i\delta t} \langle \boldsymbol{\alpha}(t)\boldsymbol{\alpha}(0) \rangle dt + \int_{-\infty}^{0} e^{-i\delta t} \langle \boldsymbol{\alpha}(0)\boldsymbol{\alpha}(-t) \rangle dt \ . \tag{31}$$

Substituting Eqs. (29) and (30) into Eq. (31) and changing $-t \to t$ in the second integral yields

$$\begin{aligned} \mathscr{S}(\delta) &= \int_0^{\infty} e^{-i\delta t} e^{-\mathbf{A}t} \langle \boldsymbol{\alpha\alpha} \rangle dt + \int_0^{\infty} e^{i\delta t} \langle \boldsymbol{\alpha\alpha} \rangle e^{-\mathbf{A}^T t} dt \\ &= (\mathbf{A} + i\delta)^{-1} \langle \boldsymbol{\alpha\alpha} \rangle + \langle \boldsymbol{\alpha\alpha} \rangle (\mathbf{A}^T - i\delta)^{-1} \ , \end{aligned} \tag{32}$$

where $\langle \boldsymbol{\alpha\alpha} \rangle \equiv \langle \boldsymbol{\alpha}(0)\boldsymbol{\alpha}(0) \rangle$ and where $\pm i\delta$ is multiplied by the identity matrix. Equation (32) was first derived by Lax[33] in his famous papers on classical and quantum noise. $\langle \boldsymbol{\alpha\alpha} \rangle$ is given by the steady-state solution of the quantum coupled mode equations and is presented in Eqs. (19)–(21) above. We could determine $\mathscr{S}(\delta)$ by making the matrix $\langle \boldsymbol{\alpha\alpha} \rangle$ from these solutions and carrying out the matrix multiplications in Eq. (32), but here we use an alternative, and this leads to the expression used in Refs. 25 and 31.

The equation of motion for $\langle \boldsymbol{\alpha\alpha} \rangle$ describing its transition to steady-state may be found by expressing Eqs. (11) and (12) in matrix form. We find

$$\frac{d}{dt}\langle \boldsymbol{\alpha\alpha} \rangle = -\mathbf{A}\langle \boldsymbol{\alpha\alpha} \rangle - \langle \boldsymbol{\alpha\alpha} \rangle A^T + \mathbf{D} \ , \tag{33}$$

where $\mathbf{D}$ is the diffusion matrix

$$\mathbf{D} = \begin{bmatrix} 0 & A_1 + A_1^* & C_1 + C_3 & 0 \\ A_1 + A_1^* & 0 & 0 & C_1^* + C_3^* \\ C_1 + C_3 & 0 & 0 & A_3 + A_3^* \\ 0 & C_1^* + C_3^* & A_3 + A_3^* & 0 \end{bmatrix} . \tag{34}$$

Equation (33) is a matrix form of the generalized Einstein relation[33] that we used in paper VII (Ref. 23) in our series to calculate the diffusion coefficients for the quantum Langevin equations from our theory. In steady state this becomes

$$\mathbf{D}=\mathbf{A}\langle \alpha\alpha \rangle + \langle \alpha\alpha \rangle \mathbf{A}^T$$
$$=(\mathbf{A}+i\delta)\langle \alpha\alpha \rangle + \langle \alpha\alpha \rangle (\mathbf{A}^T - i\delta) . \quad (35)$$

Multiplying on the left by $(\mathbf{A}+i\delta)^{-1}$ and on the right by $(\mathbf{A}^T-i\delta)^{-1}$,

$$(\mathbf{A}+i\delta)^{-1}\mathbf{D}(\mathbf{A}-i\delta)^{-1}=\langle \alpha\alpha \rangle (\mathbf{A}^T-i\delta)^{-1} + (\mathbf{A}+i\delta)^{-1}\langle \alpha\alpha \rangle . \quad (36)$$

The right-hand side (rhs) of Eq. (36) is precisely the same as the rhs of Eq. (32), so

$$\mathscr{S}(\delta)=(\mathbf{A}+i\delta)^{-1}\mathbf{D}(\mathbf{A}^T-i\delta)^{-1} , \quad (37)$$

which with δ replaced by ω is identical to the expression in Refs. 25 and 31. We note that the derivation of Eq. (37) follows from the equation of motion of the reduced density operator ρ, Eq. (5), and does not require the use of Langevin equations.

Equation (37) yields the spectral density for the components $\langle a_1^\dagger a_1 \rangle$, $\langle a_1 a_3 \rangle$, etc., inside the cavity. It is still necessary to calculate the corresponding spectral density outside the cavity. To do this we must multiply by the density of states $\mathscr{D}(\Omega_2)$ outside the cavity. Furthermore, the amount passed is proportional to the square of coupling constant g between a field mode inside the cavity and those outside. Hence we expect that the spectral quantity outside is given by

$$\mathscr{S}_{\text{out}}(\delta)=2\pi g^2 \mathscr{D}(\Omega_2)\mathscr{S}(\delta) .$$

Louisell and Walker[34] showed that the damping constant of a simple harmonic oscillator coupled to a bath of simple harmonic oscillators with density of states $\mathscr{D}(\Omega_2)$ is given by $2\pi g^2\mathscr{D}(\Omega_2)$. In the present case, this damping constant is the cavity linewidth ν/Q, i.e.,

$$\mathscr{S}_{\text{out}}(\delta)=\frac{\nu}{Q}\mathscr{S}(\delta) .$$

It is straightforward to calculate the components of $\mathscr{S}(\delta)$ directly from Eqs. (27), (34), and (37). We find

$$\mathscr{S}_{12}=\frac{(\alpha_3-i\delta)(\alpha_3^*+i\delta)A_1+|\chi_1|^2A_3+(\alpha_3^*+i\delta)\chi_1^*(C_1+C_3)+\text{c.c.}}{|(\alpha_1+i\delta)(\alpha_3^*+i\delta)-\chi_1\chi_3^*|^2} , \quad (38)$$

$$\mathscr{S}_{13}=\frac{(\alpha_3^*+i\delta)\chi_3(A_1+A_1^*)+(\alpha_1^*-i\delta)\chi_1(A_3+A_3^*)+(\alpha_1^*-i\delta)(\alpha_3^*+i\delta)(C_1+C_3)+\chi_1\chi_3}{|(\alpha_1+i\delta)(\alpha_3^*+i\delta)-\chi_1\chi_3^*|^2} , \quad (39)$$

and

$$\mathscr{S}_{34}=\mathscr{S}_{12}(1\leftrightarrow 3) . \quad (40)$$

Equations (38)–(40) are identical to those obtained by Reid and Walls,[25] Eq. (34b), if we let $\delta=0$. As discussed in Ref. 25, the best squeezing occurs at the sidemode cavity resonance $\delta=0$, and in that limit, from the expression for the variance from Eq. (17),

$$\Delta d_1^2=\frac{1}{4}+\frac{1}{4}\frac{\nu}{Q}(\mathscr{S}_{12}+\mathscr{S}_{34}-2|\mathscr{S}_{13}|) . \quad (41)$$

IV. FOURTH-ORDER EXPRESSIONS; COMPARISON TO EXPERIMENT

By substituting Eqs. (38)–(40) into Eq. (41) and letting $\delta=0$, we calculate the squeezing variance Δd_1^2 as a function of the pump intensity I_2, the detuning of the pump from the atomic resonance $\omega-\nu_2$, the detuning of the sidebands from the pump Δ, the atomic lifetimes T_1 and T_2, the cavity detuning $\Delta\Omega$ and linewidth ν/Q, and the cooperativity parameter

$$C=\frac{Ng^2}{\gamma(\nu/Q)} . \quad (42)$$

In this section we investigate the dependence of the amount of squeezing on these parameters. In the following discussion, however, we assume pure radiative decay, $T_2=2T_1$, and that the cavity detuning is adjusted to compensate for the nonlinear dispersion at the pump frequency, i.e.,

$$\Delta\Omega=\frac{\nu}{2Q}\frac{2C\mathscr{L}_2}{1+I_2\mathscr{L}_2}\frac{\Delta_2}{\gamma} . \quad (43)$$

The expressions for the quantum coefficients A_1 through D_3 are complex and are not easily interpreted. In the experiment of Slusher *et al.*,[11] although the pump field intensity is quite high (1803 times the saturation intensity), the pump is sufficiently detuned from the atomic resonance (303 times the homogeneous linewidth) so that the experiment is in a low saturation regime. This means that Eqs. (6)–(9) may be expanded in a power series in the intensity I_2, and only the lowest order need be retained. For the A_1 coefficient, we have

$$A_1=\frac{Ng^2\mathscr{D}_1I_2}{4}\left[2\mathscr{L}_2+\frac{\gamma\Gamma\mathscr{D}_2^*}{i\Delta}\right] . \quad (44)$$

Thus the more important quantity $A_1+A_1^*$ is

$$A_1+A_1^*=\frac{Ng^2I_2\mathscr{L}_2}{4}\left[\frac{2\mathscr{L}_1}{\gamma}(1-\Gamma/2\gamma)+\frac{\pi\Gamma}{\gamma}\delta(\Delta)\right] , \quad (45)$$

where $\mathscr{L}_1$ is the same as $\mathscr{L}_2$ with ν_1 replacing ν_2. The δ function in Eq. (45) is the well-known elastic, or Rayleigh, contribution to the resonance fluorescence spectrum. We see that to this order, for pure radiative decay ($\Gamma=2\gamma$), the inelastic part of $A_1+A_1^*$ is zero. The C_1 coefficient can also be expanded, and to lowest order in I_2 Eq. (8) becomes

$$C_1=-\frac{Ng^2\mathcal{D}_1\mathcal{D}_2\gamma\Gamma I_2}{4i\Delta}, \tag{46}$$

and thus C_1+C_3 is

$$C_1+C_3=-\frac{Ng^2I_2}{2}\left[\gamma\Gamma\mathcal{D}_1\mathcal{D}_2\mathcal{D}_3+\frac{\pi\Gamma(\Delta_2^2-\gamma^2)}{\gamma^3}\delta(\Delta)\right]. \tag{47}$$

In this limit the semiclassical quantities B_1-A_1 and $\mathcal{X}_1=C_1-D_1$ are given by

$$B_1-A_1=Ng^2\mathcal{D}_1\{1-I_2[\mathcal{L}_2+\gamma\mathcal{F}(\mathcal{D}_1+\mathcal{D}_3^*)/2]\}, \tag{48}$$

and

$$C_1-D_1=Ng^2\mathcal{D}_1\mathcal{D}_2\mathcal{D}_3^*I_2\frac{\gamma\Gamma}{2}\frac{2\gamma+i\Delta}{\Gamma+i\Delta}. \tag{49}$$

Note that unlike all of the other expressions, the absorption coefficient contribution B_1-A_1 does not vanish for I_2 equal to zero. Hence the constant $Ng^2\mathcal{D}_1$ is the leading term.

For the parameters of the Slusher experiment, it is possible to simplify these expressions even further. For that case, Δ_1, Δ_2, Δ_3, and Δ are much larger than γ or Γ, so the complex Lorentzians $\mathcal{D}_n$ and population pulsation factor $\mathcal{F}$ become pure imaginary and

$$C_1+C_3=C_1-D_1=-i\frac{Ng^2I_2\gamma\Gamma}{2\Delta_1\Delta_2\Delta_3}, \tag{50}$$

and

$$B_1-A_1=-iNg^2/\Delta_1. \tag{51}$$

In this limit α_1 and α_3 of Eq. (22) are given by

$$\alpha_1=\frac{\nu}{2Q}\left[1+i2C\gamma\left[\frac{1}{\Delta_2}-\frac{1}{\Delta_1}\right]\right] \tag{52}$$

and

$$\alpha_3=\frac{\nu}{2Q}\left[1+i2C\gamma\left[\frac{1}{\Delta_2}-\frac{1}{\Delta_3}\right]\right], \tag{53}$$

where we have chosen $\Delta\Omega$ to be given by Eq. (43) for small I_2. Although Eqs. (50)–(53) for C_1+C_3, $\mathcal{X}_1$, and A_1-B_1 are considerably simpler than their complete expressions, the expression for the variance given by Eq. (41) is still rather complicated. Let us consider the various magnitudes of these terms for the parameters of Slusher's experiment. The intensity I_2 in units of the resonant saturation intensity is 1800, the detuning from atomic resonance is -300γ, and the pump-probe frequency difference Δ is 84γ. Thus $\Delta_1=-216\gamma$ and $\Delta_3=-384\gamma$. The cavity cooperativity $2C$ is 1500. Using these numbers we find, in units of $\nu/2Q$, that $|\alpha_1|=1$, $|\alpha_3|=3.2$, and $|\mathcal{X}_1|=|\mathcal{X}_3|=|C_1+C_3|=6\times10^{-5}$. Thus absorption coefficients dominate the expressions for the $\mathcal{S}$ matrix elements and the variance, Eqs. (38)–(41), and so this simplifies to

$$\Delta d_1^2=\frac{1}{4}-\frac{1}{2}\frac{\nu}{Q}\left|\frac{C_1+C_3}{\alpha_1\alpha_3^*}\right|, \tag{54}$$

where we have dropped the terms $\mathcal{S}_{12}$ and $\mathcal{S}_{34}$ from Eq. (54). Because $\mathcal{S}_{12}$ and $\mathcal{S}_{34}$ are real and positive, Eq. (54) tends to overestimate the squeezing. Inserting Eqs. (50), (52), and (53) into Eq. (54), we obtain

$$\Delta d_1^2=\frac{1}{4}-2C\frac{\nu}{Q}\frac{I_2\Gamma\gamma}{|\Delta_1\Delta_2\Delta_3|}\times\left|\left[1+i\left[\frac{2C\gamma\Delta}{\Delta_1\Delta_2}\right]\right]\left[1-i\left[\frac{2C\gamma\Delta}{\Delta_2\Delta_3}\right]\right]\right|^{-1/2}. \tag{55}$$

We now examine our theoretical predictions for the variance given by Eq. (41) using the complete expressions of Eqs. (38)–(40) for the parameters of Slusher's experiment. On the same graph we also plot the corresponding curve from Eq. (55). In the recent work by Reid and Walls,[25] they present numerous figures such as this, and we refer the reader to that paper for more examples. We are mainly concerned here with our predictions for Slusher's experiment, and we attempt not to duplicate too many of their curves.

Figure 3 depicts the squeezing variance Δd_1^2 versus the pump-signal frequency difference $(\nu_2-\nu_1)T_2$ for $I_2=1800$, $C=750$, and $\Delta_2=-300T_2^{-1}$. The solid curve is the full expression from Eq. (41) and the dotted curve is the approximate expression from Eq. (55). The vertical line shows where Slusher's experiment was performed at $\Delta=84T_2^{-1}$. We see from Fig. 3 that we predict a variance of about 0.18 for this experiment, yielding squeezing of about 21%. This agrees reasonably well with the reported

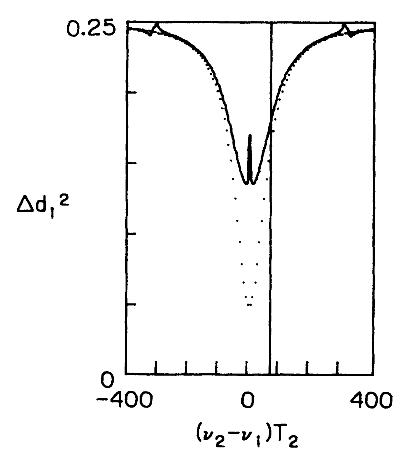

FIG. 3. Δd_1^2 vs $(\nu_2-\nu_1)T_2=\Delta T_2$ from Eq. (39) (solid curve) and Eq. (52) (dotted curve). $I_2=1800$, $\Delta_2=-300T_2^{-1}$, and $C=750$. In this figure and in Fig. 4 the vertical line shows the value used in the experiment of Ref. 11.

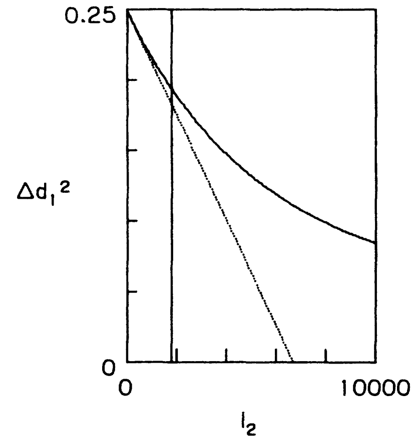

FIG. 4. Δd_1^2 vs I_2 from Eq. (39) (solid curve) and Eq. (52) (dotted curve). Other parameters the same as in Fig. 3.

observation of 10%, since we neglect the detrimental effects of phase jitter in the pump beam.[11] We predict improved squeezing for smaller Δ, but that in the region around the degenerate limit where $\Delta=0$, the variance again increases. We also see that the approximate dotted curve agrees well with the exact expression for Δ greater than $50T_2^{-1}$, except perhaps around the detuned Rabi sidebands. For $\Delta=84T_2^{-1}$, we obtain $\Delta d_1^2=0.17$, giving an error of about 10%. For $\Delta<50T_2^{-1}$, the intensity terms $\mathscr{S}_{12}$ and $\mathscr{S}_{34}$ become more relevant, and the approximations leading to Eq. (55) break down.

In Fig. 4 we plot Δd_1^2 versus the pump intensity I_2. From Eq. (55), the dotted curve is simply a straight line. In agreement with Eq. (55), the variance decreases with increasing pump intensity, although not as rapidly as a linear relationship. The physical reason for this is that the sidebands are coupled to each other by means of the pump field and the nonlinear two-level medium, and increasing the pump field increases this coupling, as seen in the expressions for χ_1 and C_1+C_3. For higher intensities, our third-order relationships break down and again $\mathscr{S}_{12}$ and $\mathscr{S}_{34}$ become important in Eq. (41).

Although Eq. (55) is rather complicated, it still allows for some interpretations of the parameter dependence of the squeezing. For example, in Fig. 5 we plot Δd_1^2 versus the detuning $(\omega-\nu_2)T_2=\Delta_2 T_2$ for pump-signal frequency differences Δ of 0, $40T_2^{-1}$, and $84T_2^{-1}$. We see detuning is always necessary for squeezing and that in general optimum squeezing is obtained at a specific detuning depending on the value of Δ. For $\Delta=84T_2^{-1}$, this optimum value occurs at approximately $\pm 300T_2^{-1}$, indicating Slusher's detuning is at a good position for his experimental parameters. This is consistent with Eq. (55).

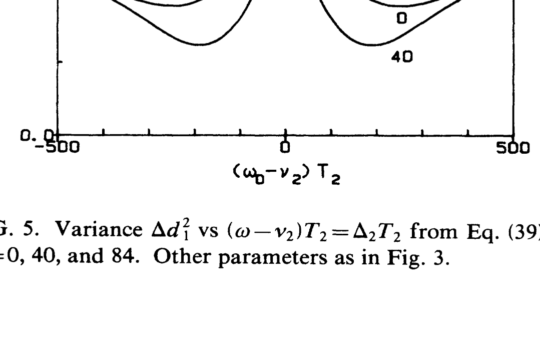

FIG. 5. Variance Δd_1^2 vs $(\omega-\nu_2)T_2=\Delta_2 T_2$ from Eq. (39) for $\Delta T_2=0$, 40, and 84. Other parameters as in Fig. 3.

V. SQUEEZING VIA PROPAGATION

Sections III and IV treated squeezing in optical cavities. In this section we apply our theory to study the generation of squeezed states by three-wave mixing propagating in a medium without a cavity. This applies to the experiment of Bondurant *et al.*[35] and the recently reported achievement of squeezing by Maeda *et al.*[13] Other authors have treated this problem in the degenerate case.[8–10,36] Following these papers, we obtain the propagation equations of motion by letting $z=ct$ in Eqs. (11) had (12) and drop the cavity-dependent terms ν/Q and $\Delta\Omega$. An alternative approach to this problem has been presented by Heidmann and Reynaud,[37] who applied the Fresnel-Huygens principle to the propagation of multimode fields in an optically thin medium.

We have recently presented the solution to the quantum coupled mode equations, Eqs. (11) and (12).[16] Because our solution is valid for nongenerate, detuned fields and for arbitrary propagation lengths, it is algebraically complex. Appendix A gives a detailed derivation of the solution to the differential equations of Eqs. (11) and (12). Appendix B solves these equations from the corresponding quantum Langevin equation picture, providing a check on our solutions and yielding additional insight into the problem.

Instead of using the cavity formalism developed for Eq. (41), we now use Eq. (17) to calculate the variance. We insert Eqs. (A15) and (A16), along with the solution for $\langle a_3^\dagger a_3\rangle=n_3$, into Eq. (17). Just as in the cavity problem, the pump field induces a nonlinear dispersion in the medium reducing the squeezing. In this case we may compensate for this effect by adjusting the phase mismatch of the wave vectors $\Delta\mathbf{K}\cdot\mathbf{r}=(2\mathbf{K}_2-\mathbf{K}_1-\mathbf{K}_3)\cdot\mathbf{r}$ to cancel out this dispersion. This corresponds to adjusting $\Delta\Omega$ in the cavity problem. We thus let

$$|\Delta\mathbf{k}|=\mathrm{Im}(\alpha_{\mathrm{pump}})=-\frac{\alpha_0\mathscr{L}_2}{1+I_2\mathscr{L}_2}\frac{\Delta_2}{\gamma}, \tag{56}$$

where for propagation we set $\alpha_0=cNg^2/\gamma$. Examples of this solution have been presented in Ref. 16. Here we give additional examples.

In the experiment of Maeda *et al.*,[13] two nearly collinear pump beams are directed into a sodium vapor cell. The pump beams are detuned from the atomic resonance by 5 GHz, and so they lie somewhat outside the Doppler width of about 2 GHz. Each pump beam has an intensity of about 1000 in units of the resonant saturation intensity. Initially a probe field is sent through the medium, generating a conjugate field, and the direction of the probe field is adjusted to maximize the signal of the conjugate

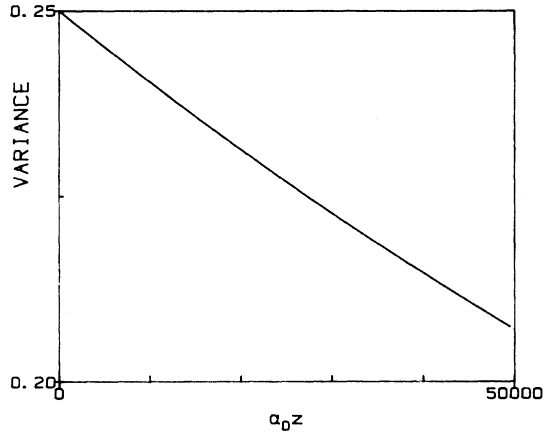

FIG. 6. Variance Δd_1^2 vs $\alpha_0 z$ for $\Delta = 11T_2^{-1}$, $\Delta_2 = -1000$ T_2^{-1}, and $I_2 = 2000$.

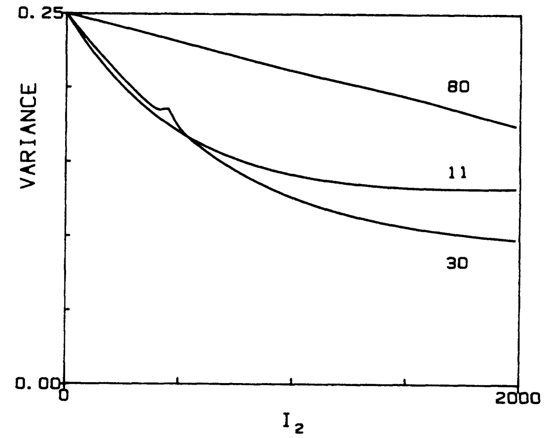

FIG. 8. Variance Δd_1^2 vs I_2 for $\Delta = 11T_2^{-1}$, $30T_2^{-1}$, and $80T_2^{-1}$ and $\alpha_0 z = 10^5$. Other parameters the same as Fig. 6.

field, i.e., until Eq. (56) is satisfied. The probe field is then blocked; the signals now incident on the balanced detector arise from the quantum multiwave mixing processes in the medium. The detectors measure noise about 55 MHz from the pump field, making $\Delta = 11T_2^{-1}$ in our notation.

Figure 6 plots the variance of Eq. (17) versus $\alpha_0 z$ for the above parameters. We see that the variance rapidly drops below the vacuum level of 0.25 with increasing distance. The experiment of Ref. 13 reports squeezing of about 3%. Allowing for detector losses, our theory should predict about 9% to be in agreement with this. This occurs at a propagation distance of about $20\,000\alpha_0 z$. We note that significantly better squeezing may be achieved if the propagation distance can be lengthened. In Fig. 7 we show the variance versus $\alpha_0 z$ for the same parameters as Fig. 6 for the pump-probe detunings $\Delta = 11T_2^{-1}$, $30T_2^{-1}$, and $80T_2^{-1}$. We note that in the first two curves the variance drops to about 0.1 before increasing again. The oscillatory behavior in the $\Delta = 80T_2^{-1}$ curve arises from the sinusoidal functions in the analytic solution, indicating that a larger propagation distance is

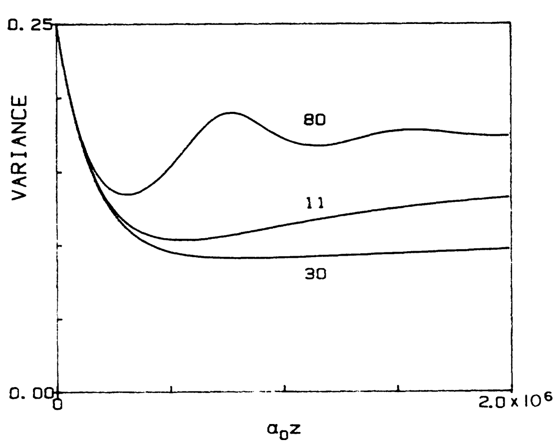

FIG. 7. Variance Δd_1^2 vs $\alpha_0 z$ for $\Delta = 11T_2^{-1}$, $30T_2^{-1}$, and $80T_2^{-1}$. The other parameters are the same as Fig. 6.

required in this case to achieve a steady-state.

Finally, Fig. 8 shows the variance versus the pump intensity I_2 for the same pump-probe detunings. The propagation distance $\alpha_0 z$ is 1×10^5 and the other parameters are the same as in Fig. 7. We note the similarity of these curves plotted versus the pump intensity to those of Fig. 7 which are plotted versus the propagation distance. In each case we see that the coupling between the sidemodes, the source of squeezing, is best for long interaction distances and high pump intensities. As in the previous curves for the medium in a cavity, there is an optimum intensity for each pump-probe detuning.

VI. CONCLUSIONS

In conclusion we have applied our quantum theory of multiwave mixing to the generation of squeezed states of light via three- and four-wave mixing in cavities and in propagation. We have specifically compared the cavity predictions to the experimental results of Slusher *et al.*[11] finding reasonably good agreement. Our cavity discussion uses the spectral matrix methods of Reid and Walls,[25] and we give derivations of the spectral formulas they use based on the classical noise methods developed by Lax.[33] In particular, we note that the reason the spectral quantities outside the cavity are given by those inside multiplied by the cavity linewidth is that this linewidth is, in fact, the product of the outside density of states factor and the square of the coupling constant between modes inside and outside. One expects the inside spectral quantities to be multiplied by the outside density of states in passing outside. This process enhances the squeezing predicted by steady-state variances, which amount to averages of the spectral variances over the cavity linewidth.

We have found analytic formulas for the variances for arbitrary propagation distances, detunings, and pump intensities. We derive these formulas both from the density operator and from the Langevin methods. To obtain significant squeezing, the propagation distances must be very large compared to the resonant Beer's law length.

In analyzing the Slusher *et al.*[11] experiments, we have found that the squeezing is accurately predicted by approximating our mixing formulas to lowest order in the pump intensity. This results from choosing sufficiently large detunings from line center. A second consequence of these large detunings is that the multimode coefficients A_1, B_1, C_1, and D_1 are nearly pure imaginary since they represent the response of driven anharmonic oscillators off resonance. This causes the source for the sidemode number operators, namely the resonance fluorescence $A_1+A_1^*$, to be small, since it is the real part of a nearly pure imaginary number. In contrast, the source terms for the combination tone operator is the complex quantity C_1+C_3, which can have appreciable magnitude. This leads to squeezing, since the combination tone expectation values have larger magnitudes than those for the number operators. On the other hand, the next paper[38] in this series studies the corresponding squeezing generated in two-photon media and finds that even in the presence of small two-photon resonance fluorescence, the number operators can have substantial values due to coupling with the combination tone terms.

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APPENDIX A: PROPAGATION SOLUTION FROM THE DENSITY OPERATOR EQUATIONS

In this appendix we obtain the solution to the quantum coupled mode equations using the equations found from the reduced density operator equation of motion. In Appendix B we show how this solution may also be obtained by using Langevin equations. The semiclassical coupled mode equations are

$$\frac{d\mathscr{E}_1}{dz}=-\alpha_1\mathscr{E}_1+\chi_1\mathscr{E}_3^* , \tag{A1}$$

$$\frac{d\mathscr{E}_3}{dz}=-\alpha_3\mathscr{E}_3+\chi_3\mathscr{E}_1^* , \tag{A2}$$

where, as in Eqs. (19)–(21) with $\nu/Q=0$, $\alpha_1=B_1-A_1$ and $\chi_1=C_1-D_1$. Equations (A1) and (A2) may be easily solved in any number of ways. For example, by taking the Laplace transform of Eqs. (A1) and (A2) and solving for the transform of $\mathscr{E}_3(z)$, we have

$$E_3(s)=\frac{(s+\alpha_1^*)\mathscr{E}_{30}+\chi_3\mathscr{E}_{10}^*}{(s-s_1)(s-s_2)} , \tag{A3}$$

where $E_3(s)$ is the Laplace transform of $\mathscr{E}_3(z)$, s is the transform parameter, $\mathscr{E}_{10}^*$ and $\mathscr{E}_{30}$ are the initial conditions of $\mathscr{E}_1^*$ and $\mathscr{E}_3$, respectively, and where

$$s_{1,2}=-\tfrac{1}{2}(\alpha_3+\alpha_1^*)\pm w=-a\pm w , \tag{A4}$$

and

$$w=\left[\frac{(\alpha_3-\alpha_1^*)^2}{4}+\chi_1^*\chi_3\right]^{1/2} . \tag{A5}$$

Taking the inverse Laplace transform of Eq. (A3) and the similar equation for $E_1^*(s)$ yields

$$\mathscr{E}_1^*(z)=F\mathscr{E}_1^*(0)+\chi_1^*G\mathscr{E}_3(0) , \tag{A6}$$

$$\mathscr{E}_3(z)=H\mathscr{E}_3(0)+\chi_3G\mathscr{E}_1^*(0) , \tag{A7}$$

where

$$F=\frac{1}{w}\left[w\cosh(wz)+\frac{\alpha_3-\alpha_1^*}{2}\sinh(wz)\right]e^{-az} ,$$

$$G=\frac{1}{w}\sinh(wz)e^{-az} .$$

The H term is identical to F except that α_1^* and α_3 are interchanged.

The corresponding quantum-mechanical coupled mode equations are, from Eqs. (11) and (12),

$$\frac{dn_1}{dz}\equiv\frac{d\langle a_1^\dagger a_1\rangle}{dz}=-\alpha_1 n_1+\chi_1 m^*+A_1+\text{c.c.} , \tag{A8}$$

$$\frac{dm}{dz}\equiv\frac{d\langle a_1 a_3\rangle}{dz}=-a_1 m+\chi_1 n_3+C_1+(1\leftrightarrow 3) . \tag{A9}$$

$1\leftrightarrow 3$ represents the same previous terms with 1 interchanged with 3. The equations of motion for $n_3\equiv\langle a_3^\dagger a_3\rangle$ and $m^*\equiv\langle a_1^\dagger a_3^\dagger\rangle$ are found by replacing the subscript 1 with 3 in Eq. (A8) and taking the complex conjugate of Eq. (A9). It is possible to derive the homogeneous form (without the source terms $A_1+A_1^*$ and C_1+C_3) of Eqs. (A8) and (A9) from the semiclassical Eqs. (A1) and (A2). Multiplying both sides of Eq. (A1) by $\mathscr{E}_1^*$ and adding the complex conjugate, we obtain

$$\frac{d}{dz}(\mathscr{E}_1\mathscr{E}_1^*)=-\alpha_1\mathscr{E}_1\mathscr{E}_1^*+\chi_1\mathscr{E}_1^*\mathscr{E}_3^*+\text{c.c.} \tag{A10}$$

Similarly, multiplying Eq. (A1) by $\mathscr{E}_3$ and Eq. (A2) by $\mathscr{E}_1$ and adding, we have

$$\frac{d}{dz}(\mathscr{E}_1\mathscr{E}_3)=-\alpha_1\mathscr{E}_1\mathscr{E}_3+\chi_1\mathscr{E}_3\mathscr{E}_3^*+(1\leftrightarrow 3) . \tag{A11}$$

Equations (A10) and (A11) are identical to the homogeneous form of Eqs. (A8) and (A9) if we identify $\mathscr{E}_1\mathscr{E}_1^*$ with n_1 and $\mathscr{E}_1\mathscr{E}_3$ with m. Hence the solutions to the homogeneous Eqs. (A8) and (A9) are the appropriate products of $\mathscr{E}_1(z)$ and $\mathscr{E}_3(z)$ as given by Eqs. (A4) and (A5), provided we use for the initial conditions for n_1 and m, $\mathscr{E}_1^*(0)\mathscr{E}_1(0)$ and $\mathscr{E}_1(0)\mathscr{E}_3(0)$, respectively.

We use these homogeneous solutions to solve the inhomogeneous Eqs. (A8) and (A9). If we denote the Laplace transform of n_1 by $\mathscr{N}_1$ and m by $\mathscr{M}$ and take the transform of Eq. (A8), we obtain

$$s\mathscr{N}_1-n_{10}=-\alpha_1\mathscr{N}_1+\chi_1\mathscr{M}^*+a_1/s+\text{c.c.} \tag{A12}$$

Note that by grouping terms with $\mathscr{N}_1(s)$ and $\mathscr{M}(s)$ on the left-hand side (lhs), we are left with $n_{10}+(A_1+A_1^*)/s$ on the right-hand side. The lhs of the equation is identical with the Laplace transform of the homogeneous equations. Thus the solution including the inhomogeneous source terms is the same except that we replace n_{10} by $n_{10}+(A_1+A_1^*)/s$, m_0 by $m_0+(C_1+C_3)/s$, n_{30} by $n_{30}+(A_3+A_3^*)/s$, and m_0^* by $m_0^*+(C_1^*+C_3^*)/s$.

The homogeneous solution of n_1 is

$$n_{1h}=(F\mathscr{E}_{10}+\mathcal{X}_1^*G\mathscr{E}_{30})(F^*\mathscr{E}_{10}^*+\mathcal{X}_1G^*\mathscr{E}_{30}^*)$$
$$=|F|^2n_{10}+|\mathcal{X}_1G|^2n_{30}+(\mathcal{X}_1FG^*m^*+\text{c.c.})\ . \tag{A13}$$

Substituting the expressions for F and G into Eq. (A13) yields

$$n_{1h}e^{(a+a^*)z}=n_{10}\cosh(wz)\cosh(w^*z)+\frac{\sinh(wz)\sinh(w^*z)}{4|w|^2}\{|\alpha_3-\alpha_1^*|^2n_{10}+4|\mathcal{X}_1|^2n_{30}+[2\mathcal{X}_1(\alpha_3-\alpha_1^*)m^*+\text{c.c.}]\}$$
$$+\left\{\frac{\cosh(wz)\sinh(w^*z)}{2w^*}[(\alpha_3^*-\alpha_1)n_{10}+2\mathcal{X}_1m^*]+\text{c.c.}\right\}\ . \tag{A14}$$

Because the Laplace transform of e^{cz} is $1/(s-c)$, we expand the hyperbolic functions into their exponential form and find the Laplace transform of n_{1h} to be

$$\mathscr{N}_{1h}=\frac{n_{10}}{4}\left[\frac{1}{s-(w+w^*-b)}+\frac{1}{s-(w-w^*-b)}+\frac{1}{s-(w^*-w-b)}+\frac{1}{s+w+w^*+b}\right]$$
$$+\{|\alpha_3-\alpha_1^*|^2n_{10}+4|\mathcal{X}_1|^2n_{30}+[2\mathcal{X}_1(\alpha_3-\alpha_1^*)m^*+\text{c.c.}]\}$$
$$\times\frac{1}{4}\left[\frac{1}{s-(w+w^*-b)}-\frac{1}{s-(w-w^*-b)}-\frac{1}{s-(w^*-w-b)}+\frac{1}{s+w+w^*+b}\right]$$
$$+\left[[(\alpha_3^*-\alpha_1)n_{10}+2\mathcal{X}_1m^*]\right.$$
$$\left.\times\frac{1}{4}\left[\frac{1}{s-(w+w^*-b)}-\frac{1}{s-(w-w^*-b)}+\frac{1}{s-(w^*-w-b)}-\frac{1}{s+w+w^*+b}\right]+\text{c.c.}\right]\ , \tag{A15}$$

where $b=a+a^*$. We follow the above prescription and replace n_{10} by $n_{10}+(A_1+A_1^*)/s$, etc. This is then the Laplace transform of $\mathscr{N}_1(s)$. We see that the effect of the inhomogeneous source terms is to multiply the Laplace transform of Eq. (A15) by $1/s$. Since the inverse transform of $1/s(s-c)$ is $(e^{cz}-1)/c$, the solution for $n_1(z)$ is (setting the initial conditions n_{10}, n_{30}, and m_0 equal to zero because that solution is already known)

$$n_1=\frac{A_1+A_1^*}{4}\left[\frac{e^{(w+w^*-b)z}-1}{w+w^*-b}+\frac{e^{(w-w^*-b)z}-1}{w-w^*-b}+\frac{e^{(w^*-w-b)z}-1}{w^*-w-b}-\frac{e^{-(w+w^*+b)z}-1}{w+w^*+b}\right]$$
$$+\{|\alpha_3-\alpha_1^*|^2(A_1+A_1^*)+4|\mathcal{X}_1|^2(A_3+A_3^*)+[2\mathcal{X}_1(\alpha_3-\alpha_1^*)(C_1^*+C_3^*)+\text{c.c}]\}$$
$$\times\frac{1}{16|w|^2}\left[\frac{e^{(w+w^*-b)z}-1}{w+w^*-b}-\frac{e^{(w-w^*-b)z}-1}{w-w^*-b}-\frac{e^{(w^*-w-b)z}-1}{w^*-w-b}-\frac{e^{-(w+w^*+b)z}-1}{w+w^*+b}\right]$$
$$+\left[[(\alpha_3^*-\alpha_1)(A_1+A_1^*)+2\mathcal{X}_1(C_1^*+C_3^*)]\right.$$
$$\left.\times\frac{1}{8w^*}\left[\frac{e^{(w+w^*-b)z}-1}{w+w^*-b}-\frac{e^{(w-w^*-b)z}-1}{w-w^*-b}+\frac{e^{(w^*-w-b)z}-1}{w^*-w-b}+\frac{e^{-(w+w^*+b)z}-1}{w+w^*+b}\right]+\text{c.c.}\right]\ . \tag{A16}$$

Equation (A9) for $m\equiv\langle a_1a_3\rangle$ may be solved in a similar fashion. Equation (A16) may be written in a more compact form by recombining the exponentials into hyperbolic and sinusoidal functions. After this algebraic simplification, we find

$$n_1=(A_{1r}+N_1)q_1+(A_{1r}-N_1)q_2+N_{2r}q_3-N_{2i}q_4\ , \tag{A17}$$

$$m=(C+M_1)q_1+(C-M_1)q_2$$
$$+(M_{13}+M_{31})q_3+i(M_{13}-M_{31})q_4\ , \tag{A18}$$

where

$$C=(C_1+C_3)/2\ ,$$

$$N_1=(4|w|^2)^{-1}\{|\alpha_3-\alpha_1^*|^2A_{1r}+4|\mathcal{X}_1|^2A_{3r}$$
$$+[2\mathcal{X}_1^*(\alpha_3^*-\alpha_1)C+\text{c.c.}]\}\ ,$$

$$N_2 = w^{-1}[(\alpha_3-\alpha_1^*)A_{1r}+2\chi_1^*C] = N_{2r}+iN_{2i} ,$$

$$M_1 = (4|w|^2)^{-1}[-|\alpha_3-\alpha_1^*|^2C+4\chi_1\chi_3C^* + 2\chi_3(\alpha_3^*-\alpha_1)A_{1r}+2\chi_1(\alpha_1^*-\alpha_3)A_{3r}] ,$$

$$M_{13} = (2w)^{-1}[-(\alpha_3-\alpha_1^*)C+2\chi_3A_{1r}] ,$$

$$M_{31} = M_{13} \text{ with } 1\leftrightarrow 3 ,$$

$$q_1 = \{b-[u\sinh(uz)+b\cosh(uz)]e^{-bz}\}/(b^2-u^2) ,$$

$$q_2 = \{b+[v\sin(vz)-b\cos(vz)]e^{-bz}\}/(b^2+v^2) ,$$

$$q_3 = \{u-[b\sinh(uz)+u\cosh(uz)]e^{-bz}\}/(b^2-u^2) ,$$

$$q_4 = \{v-[b\sin(vz)+v\cos(vz)]e^{-bz}\}/(b^2+v^2) ,$$

and where $A_{1r}=(A_1+A_1^*)/2$ and $u+iv=2w$. To obtain the solution for the mode 3 quantities, interchange the subscripts 1 and 3 everywhere. Note that $w(1\leftrightarrow 3)=w^*$.

APPENDIX B: PROPAGATION SOLUTION FROM LANGEVIN EQUATIONS

In this Appendix we present another method to solve the quantum coupled mode equations by beginning from the appropriate Langevin equations. The Langevin equations in this case are given by Eqs. (A1) and (A2) with the appropriate noise operators $F_1(z)$ and $F_3(z)$:

$$\frac{da_1}{dz} = -\alpha_1a_1+\chi_1a_3^\dagger+F_1(z) , \quad \text{(B1)}$$

$$\frac{da_3}{dz} = -\alpha_3a_3+\chi_3a_1^\dagger+F_3(z) . \quad \text{(B2)}$$

The noise operators obey the usual conditions under the Markovian assumption that

$$\langle F_1(z)\rangle = \langle F_3(z)\rangle = 0 , \quad \text{(B3)}$$

$$\langle F_1(z)F_1^\dagger(z')\rangle = D_{a_1^\dagger a_1}\delta(z-z') , \quad \text{(B4)}$$

$$\langle F_1(z)F_3(z')\rangle = D_{a_1a_3}\delta(z-z') . \quad \text{(B5)}$$

In Ref. 23 we showed by using the generalized Einstein relation and Eq. (5) that the diffusion coefficient of Eqs. (B4) and (B5) are simply the inhomogeneous source terms of our quantum coupled mode equations, so

$$D_{a_1^\dagger a_1} = A_1+A_1^* \text{ and } D_{a_1a_3} = C_1+C_3 . \quad \text{(B6)}$$

Defining the vector $\boldsymbol{\alpha}^T=(a_1,a_1^\dagger,a_3,a_3^\dagger)$ as before, we may write Eqs. (B1) and (B2) as

$$\frac{d\boldsymbol{\alpha}}{dz} = -A\boldsymbol{\alpha}+\mathbf{F}(z) , \quad \text{(B7)}$$

where $\mathbf{F}^T=(F_1,F_1^\dagger,F_3,F_3^\dagger)$ and the matrix A is the same as in Eq. (25) with $\nu/Q=\Delta\Omega=0$.

We solve Eq. (B7) by first determining the eigenvectors of A. As one can see from Eq. (25), the 4×4 matrix for A may be thought of as two separate 2×2 matrices, and the secular equation becomes

$$[(\alpha_1-\lambda)(\alpha_3^*-\lambda)-\chi_1\chi_3^*][(\alpha_1^*-\lambda)(\alpha_3-\lambda)-\chi_1^*\chi_3] = 0 . \quad \text{(B8)}$$

Each quadratic equation may be readily factored and the four eigenvalues are for $\lambda_{1,2}=a^*\pm w^*$ and $\lambda_{3,4}=a\pm w$ where a and w are given by Eqs. (A4) and (A5). Defining the eigenvector for λ_1 as $\mathbf{U}_1^T=(U_{11},U_{12},U_{13},U_{14})$, the components may be found from

$$\mathbf{U}_1^T(A-I\lambda_1) = 0 . \quad \text{(B9)}$$

A possible solution of Eq. (B9) is $U_{11}=1$, $U_{14}=(\alpha_1-\lambda_1)/\chi_3^*$, and $U_{12}=U_{13}=0$. In a similar manner, we may insert the other eigenvalues into Eq. (B9) to obtain the other eigenvectors. Our results are

$$\mathbf{U}_1=\begin{pmatrix}1\\0\\0\\ \dfrac{\alpha_1-\lambda_1}{\chi_3^*}\end{pmatrix}, \quad \mathbf{U}_2=\begin{pmatrix}1\\0\\0\\ \dfrac{\alpha_1-\lambda_2}{\chi_3^*}\end{pmatrix}, \quad \mathbf{U}_3=\begin{pmatrix}0\\1\\ \dfrac{\alpha_1^*-\lambda_3}{\chi_3}\\0\end{pmatrix}, \quad \mathbf{U}_4=\begin{pmatrix}0\\1\\ \dfrac{\alpha_1^*-\lambda_4}{\chi_3}\\0\end{pmatrix} . \quad \text{(B10)}$$

It is now possible to solve the quantum Langevin equations. We define the new variables β_i and noise operators $G_i(t)$ as

$$\beta_i = \boldsymbol{\alpha}^T\mathbf{U}_i, \quad G_i = \mathbf{F}^T(\mathbf{z})\mathbf{U}_i , \quad \text{(B11)}$$

where $\mathbf{U}$ is the matrix formed by the row vectors U_i, $i=1,4$. For example, the equation of motion for β_i is

$$\frac{d\beta_i}{dz} = -\lambda_i\beta_i+G_i(z) , \quad \text{(B12)}$$

Using the integrating factor $\exp(-\lambda_iz)$ the solution to Eq. (B12) is

$$\beta_i(z) = \beta_i(0)e^{-\lambda_iz}+\int_0^z e^{-\lambda_i(z-z')}G_i(z')dz' . \quad \text{(B13)}$$

In terms of the β_i variables, the annihilation operators a_1 and a_3 are

$$a_1 = \frac{(\beta_1-\beta_2)(\alpha_1-\alpha_3^*)+2w^*(\beta_1+\beta_2)}{4w^*} , \quad \text{(B14)}$$

and

$$a_3 = \frac{\chi_3(\beta_2^\dagger-\beta_1^\dagger)}{2w} . \quad \text{(B15)}$$

From Eq. (B14) the expression for $\langle a_1^\dagger a_1\rangle$ is given by

$$\langle a_1^\dagger a_1\rangle = \left\langle \left[\frac{\alpha_1^*-\alpha_3}{4w}(\beta_1^\dagger+\beta_2^\dagger)+\tfrac{1}{2}(\beta_1^\dagger+\beta_2^\dagger)\right]\left[\frac{\alpha_1-\alpha_3^*}{4w^*}(\beta_1-\beta_2)+\tfrac{1}{2}(\beta_1+\beta_2)\right]\right\rangle . \quad \text{(B16)}$$

We see from Eq. (B16) we need expressions for $\langle \beta_1^\dagger \beta_1 \rangle$, $\langle \beta_1^\dagger \beta_2 \rangle$, $\langle \beta_2^\dagger \beta_1 \rangle$, and $\langle \beta_2^\dagger \beta_2 \rangle$. We outline here the calculation for $\langle \beta_1^\dagger \beta_1 \rangle$; the others are similar. The initial condition term in Eq. (B13) reproduces the semiclassical results, so we drop that term. We then have

$$\langle \beta_1^\dagger \beta_1 \rangle = \left\langle \int_0^z dz' G_1^\dagger(z') e^{-\lambda_1^*(z-z')} \int_0^z dz'' G_1(z'') e^{-\lambda_1(z-z'')} \right\rangle . \tag{B17}$$

From Eq. (B11), we obtain

$$\begin{aligned}\langle G_1^\dagger(z')G_1(z'')\rangle &= \langle F_1^\dagger(z')F_1(z'')\rangle + \frac{\alpha_1-\lambda_1}{\chi_3^*}\langle F_1^\dagger(z')F_3^\dagger(z'')\rangle + \frac{\alpha_1^*-\lambda_1^*}{\chi_3^*}\langle F_1(z')F_3(z'')\rangle + \left|\frac{\alpha_1-\lambda_1}{\chi_3}\right|^2 \langle F_3^\dagger(z')F_3(z'')\rangle \\ &= \left[(A_1+A_1^*) + \frac{\alpha_1-\lambda_1}{\chi_3^*}(C_1^*+C_3^*) + \frac{\alpha_1^*-\lambda_1^*}{\chi_3}(C_1+C_3) + \left|\frac{\alpha_1-\lambda_1}{\chi_3}\right|^2 (A_3+A_3^*)\right]\delta(z'-z'') ,\end{aligned} \tag{B18}$$

where we have used Eqs. (B4)—(B6). Carrying out the integration in Eq. (B17) and substituting Eq. (B18), we have

$$\langle \beta_1^\dagger \beta_1 \rangle = \left[A_1 + \left|\frac{\alpha_1-\alpha_3^*-2w^*}{2\chi_3^*}\right|^2 A_3 + \frac{\alpha_1^*-\alpha_3-2w}{2\chi_3}(C_1+C_3) + \text{c.c.}\right] \frac{1-e^{-(b+w+w^*)z}}{b+w+w^*} , \tag{B19}$$

where $b=a+a^*$. In a similar manner we solve for the other bilinear products of the eigenvectors, and

$$\langle \beta_2^\dagger \beta_2 \rangle = \left[A_1 + \left|\frac{\alpha_1-\alpha_3^*+2w^*}{2\chi_3^*}\right|^2 A_3 + \frac{\alpha_1^*-\alpha_3+2w}{2\chi_3}(C_1+C_3) + \text{c.c.}\right] \frac{1-e^{-(b-w-w^*)z}}{b-w-w^*} , \tag{B20}$$

$$\begin{aligned}\langle \beta_1^\dagger \beta_2 \rangle = \Bigg[& A_1 + A_1^* + \frac{(\alpha_1-\alpha_3^*+2w^*)(\alpha_1^*-\alpha_3-2w)}{4|\chi_3|^2}(A_3+A_3^*) + \frac{\alpha_1^*-\alpha_3-2w}{2\chi_3}(C_1+C_3) \\ & + \frac{\alpha_1-\alpha_3^*+2w^*}{2\chi_3^*}(C_1^*+C_3^*)\Bigg] \frac{1-e^{-(b+w-w^*)z}}{b+w-w^*} ,\end{aligned} \tag{B21}$$

and

$$\langle \beta_2^\dagger \beta_1 \rangle = \langle \beta_1^\dagger \beta_2 \rangle^* . \tag{B22}$$

It is a straightforward although tedious task to substitute Eqs. (B19)—(B22) into (B16) to obtain the expression for $\langle a_1^\dagger a_1 \rangle \equiv n_1$. When this is done, we recover the previous solution for n_1 in Appendix A, Eq. (A17) and those following. From the Langevin approach adopted here, we see that the functions $q_1(z)$ and $q_3(z)$ following (A18) result from combinations of $\langle \beta_1^\dagger \beta_1 \rangle$ and $\langle \beta_2^\dagger \beta_2 \rangle$, while the functions $q_2(z)$ and $q_4(z)$ arise from $\langle \beta_1^\dagger \beta_2 \rangle$ and $\langle \beta_2^\dagger \beta_1 \rangle$.

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[1]H. Carmichael and D. F. Walls, J. Phys. B 9, 1199 (1976).
[2]C. Cohen-Tannoudji, in *Frontiers in Laser Spectroscopy,* edited by R. Balian, S. Harouch, and S. Lieberman (North Holland, Amsterdam, 1977).
[3]H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).
[4]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. A **18**, 201 (1978).
[5]For a review, see D. F. Walls, Nature **306**, 141 (1983).
[6]G. J. Milburn and D. F. Walls, Opt. Commun. **39**, 410 (1981).
[7]M. J. Collett and D. F. Walls, Phys. Rev. A **32**, 396 (1985).
[8]H. P. Yuen and J. H. Shapiro, Opt. Lett. **4**, 334 (1979).
[9]P. Kumar and J. H. Shapiro, Phys. Rev. A **30**, 1568 (1984).
[10]M. D. Reid and D. F. Walls, Phys. Rev. A **31**, 1622 (1985).
[11]R. E. Slusher, L. W. Hollberg, B. Yurke, J. C. Mertz, and J. F. Valley, Phys. Rev. Lett. **55**, 2409 (1985).
[12]R. M. Shelby, M. D. Levenson, S. H. Perlmutter, R. V. DeVoe, and D. F. Walls, Phys. Rev. Lett. **57**, 691 (1986).
[13]M. Maeda, P. Kumar, and J. H. Shapiro, XIV International Quantum Electronics Conference Technical Digest (unpublished).
[14]H. J. Kimble, XIV International Quantum Electronics Conference, Technical Digest, San Francisco, 1986 (unpublished).
[15]D. A. Holm, M. Sargent III, and B. A. Capron, Opt. Lett. **11**, 443 (1986).
[16]D. A. Holm, S. An, and M. Sargent III, Optics Commun. **60**, 328 (1986).
[17]M. Sargent III, D. A. Holm, and M. S. Zubairy, Phys. Rev. A **31**, 3112 (1985).
[18]S. Stenholm, D. A. Holm, and M. Sargent III, Phys. Rev. A **31**, 3124 (1986).
[19]D. A. Holm, M. Sargent III, and L. M. Hoffer, Phys. Rev. A **32**, 963 (1986).
[20]D. A. Holm, M. Sargent III, and S. Stenholm, J. Opt. Soc. Am. **B2**, 1456 (1985).
[21]D. A. Holm and M. Sargent III, Phys. Rev. A **33**, 1073 (1986).
[22]D. A. Holm and M. Sargent III, J. Opt. Soc. Am. **B3**, 732 (1986).
[23]D. A. Holm and M. Sargent III, Phys. Rev. A **33**, 4001 (1986).
[24]M. D. Reid and D. F. Walls, Phys. Rev. A **33**, 4465 (1986).
[25]M. D. Reid and D. F. Walls, Phys. Rev. A **34**, 4929 (1986).

[26]T. Fu and M. Sargent, Opt. Lett. **4**, 366 (1979).
[27]B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
[28]B. L. Schumaker, Opt. Lett. **9**, 189 (1984).
[29]N. B. Abraham, L. A. Lugiato, and L. M. Narducci, J. Opt. Soc. **B2**, 7 (1985).
[30]H. Haken, *Synergetics* (Springer-Verlag, New York, 1983), p. 123.
[31]M. J. Collett and D. F. Walls, Phys. Rev. A **32**, 2887 (1985).
[32]M. J. Collett and C. W. Gardiner, Phys. Rev. A **30**, 1386 (1984).
[33]M. Lax, Rev. Mod. Phys. **38**, 541 (1966).
[34]W. H. Louisell and L. R. Walker, Phys. Rev. **137**, B204 (1965).
[35]R. S. Bondurant, P. Kumar, J. H. Shapiro, and M. Maeda, Phys. Rev. A **30**, 343 (1984).
[36]S. Ya. Kilin, Opt. Commun. **53**, 409 (1985).
[37]A. Heidmann and S. Reynaud, J. Phys. (Paris) **46**, 1937 (1985).
[38]B. A. Capron, D. A. Holm, and M. Sargent III, Phys. Rev. A (to be published).