

Quantum theory of multiwave mixing. VII. Connection to quantum Langevin theory

David A. Holm and Murray Sargent III

Optical Sciences Center, University of Arizona, Tucson, Arizona 85721

(Received 3 February 1986)

We transform our quantum theory of multiwave mixing, which is based on a reduced density operator, to the corresponding nondegenerate quantum-noise Langevin equations. We derive the diffusion coefficients from the drift coefficients using the generalized Einstein relation and illustrate the resulting quantum Langevin theory of multiwave mixing by specializing to the Reid and Walls degenerate theory of squeezed states of light.

I. INTRODUCTION

In the field of theoretical quantum optics, much research has been performed in the Heisenberg picture with the help of quantum-noise operators. This includes the early work on this subject by Lax,¹ Haken and co-workers,² the resonance fluorescence description by Cohen-Tannoudji,³ the quantum theory of optical bistability by Bonifacio and Lugiato⁴ and Drummond and Walls,⁵ and more recently, the investigation of squeezed states of light by Reid and Walls.^{6,7} In these papers Langevin equations were calculated (often from an appropriate Fokker-Planck equation) and the required quantities resulted from analysis of their drift and diffusion terms.

An alternate approach derives the desired equations of motion from a combined atom-field density operator traced over atomic and field states of lesser interest to obtain a Schrödinger-picture reduced-density operator for the field mode(s) of primary interest. This was the method used in the Scully-Lamb quantum theory of the laser,⁸ in Stenholm's analysis of the interaction of radiation with matter,⁹ in the quantum three-level work of Singh and Zubairy,¹⁰ and in our quantum theory of multiwave mixing.¹¹⁻¹⁷

In general, the Heisenberg-picture quantum Langevin approach and the Schrödinger-picture reduced-density operator approach are equivalent, but provide different insights and ease with which problems can be solved. The present paper builds a bridge over which to carry new results from one approach to the other as well as to provide thoroughly independent checks of results found on either side of this bridge. Specifically this paper connects the two viewpoints by calculating the Langevin drift coefficients directly from the reduced-density operator equation of motion and by calculating the diffusion coefficients using the generalized Einstein relation.^{1,18,19} It uses the bridge to obtain a quantum Langevin theory of multiwave mixing equivalent to our reduced-density operator version.

Although this procedure has been known since the early days of quantum optics and most of it even found its way into a textbook,¹⁸ it is surprisingly not well known in the field and is especially useful in relating the application of our quantum multiwave mixing theory to squeezing to the work of Reid and Walls.^{6,7} Since both of these theoretical

treatments of four-wave mixing consider weak quantum-mechanical sidemode fields and classical pump fields interacting with two-level media, one would expect a relationship to exist. We show this to be the case. Reid and Walls omit many steps in their derivations, particularly of the all important quantum-mechanical diffusion coefficients. Our derivations¹²⁻¹⁷ are substantially more detailed and with the present paper give a totally independent check on their work. This has enabled us to provide new physical insight into their calculations, such as the relationship to saturation spectroscopy and resonance fluorescence. It also generalizes their results to nondegenerate four-wave mixing, of major importance to the theory of squeezed states, and it identifies some elastic scattering terms missing in their degenerate treatment.

In Sec. II we summarize the fundamental results of papers I and II (Refs. 12 and 13) for the case of upper-to-ground lower-level decay. Although this summary has also appeared in other papers in this series, including it here allows for an immediate understanding of our assumptions and results. Section III reviews how quantum Langevin equations can be obtained from the reduced-density operator equation of motion and how the diffusion coefficients can be calculated from the drift coefficients via the generalized Einstein relation. Section IV applies this formalism to our quantum theory of multiwave mixing, yielding the nondegenerate multiwave diffusion coefficients and reducing them to those published by Reid and Walls⁶ by subtracting out the elastic scattering contributions. Section V presents a simple example of how solutions to the differential equations derived by each method do, in fact, result in the same answer, and discusses the implications for theories concerning squeezed states.

II. SUMMARY OF BASIC EQUATIONS

In this section we summarize the theory developed in Refs. 12 and 13 that forms the basis for this paper. Our Hamiltonian (in rad/sec) is

$$\mathcal{H} = (\omega - \nu_2)\sigma_z + \sum_{j=1}^3 [(v_j - \nu_2)a_j^\dagger a_j + (g a_j U_j \sigma^\dagger + \text{H.c.})] . \quad (1)$$

In this expression a_j is the annihilation operator for the j th field mode, $U_j = U_j(\mathbf{r})$ is the corresponding spatial mode factor, σ and σ_z are the atomic spin-flip and probability-difference operators, ω and ν_j are the atomic and field frequencies (see Fig. 1), g is the atom-field coupling constant. We take mode 2 to be arbitrarily intense, and treat it classically and undepleted. 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. The rotating-wave approximation has been made and the Hamiltonian is in an interaction picture rotating at the strong field frequency ν_2 . We define an atom-field density operator ρ_{a-f} and obtain its time dependence from the standard density operator equation of motion

$$\dot{\rho}_{a-f} = -i[H, \rho_{a-f}] + \dots, \quad (2)$$

where the ellipsis represents relaxation processes.

We calculate the reduced electric field density operator ρ that describes the time dependence of the two quantized fields by taking the trace of ρ_{a-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

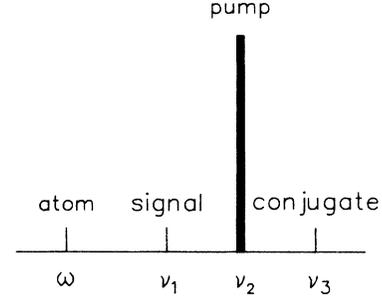


FIG. 1. Mode spectrum showing weak quantum sidemodes at frequencies ν_1 and ν_3 symmetrically placed about the arbitrarily intense pump frequency ν_2 . The atomic resonance frequency ω is near, but not necessarily equal to, the pump frequency.

$$\begin{aligned} \dot{\rho} = & -A_1(\rho a_1 a_1^\dagger - a_1^\dagger \rho a_1) - (B_1 + \nu/2Q_1)(a_1^\dagger a_1 \rho - a_1 \rho a_1^\dagger) \\ & + C_1(a_1^\dagger a_3^\dagger \rho - a_3^\dagger \rho a_1^\dagger) + D_1(\rho a_3^\dagger a_1^\dagger - a_1^\dagger \rho a_3^\dagger) \\ & + [1 \leftrightarrow 3] + \text{H.c.}, \end{aligned} \quad (3)$$

where $[1 \leftrightarrow 3]$ represents the same terms with 1 and 3 interchanged, ν/Q_1 is the cavity loss rate for cavity configurations, the coefficients A_1 , B_1 , C_1 , and D_1 are given by

$$A_1 = \frac{Ng^2 \mathcal{D}_1}{1 + I_2 \mathcal{L}_2} \left[\frac{I_2 \mathcal{L}_2}{2} - \frac{I_2 \mathcal{Y} \mathcal{F}}{2} \frac{\left[\frac{I_2 \mathcal{L}_2}{2} \mathcal{D}_1 - \mathcal{D}_2^*(1 + \Gamma/i\Delta)/2 \right]}{1 + I_2 \mathcal{F} \frac{\mathcal{Y}}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} \right], \quad (4)$$

$$B_1 = \frac{Ng^2 \mathcal{D}_1}{1 + I_2 \mathcal{L}_2} \left[1 + \frac{I_2 \mathcal{L}_2}{2} - \frac{I_2 \mathcal{Y} \mathcal{F} [(1 + I_2 \mathcal{L}_2/2) \mathcal{D}_1 + \mathcal{D}_2^*(1 - \Gamma/i\Delta)/2]}{1 + I_2 \mathcal{F} \frac{\mathcal{Y}}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} \right], \quad (5)$$

$$C_1 = -\frac{Ng^2 \mathcal{D}_1}{1 + I_2 \mathcal{L}_2} U_1^* U_3^* \frac{2T_1 \mathcal{V}_2^2 \mathcal{F} \left[\frac{I_2 \mathcal{L}_2}{2} \mathcal{D}_3^* - \mathcal{D}_2(1 + \Gamma/i\Delta)/2 \right]}{1 + I_2 \mathcal{F} \frac{\mathcal{Y}}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)}, \quad (6)$$

$$D_1 = -\frac{Ng^2 \mathcal{D}_1}{1 + I_2 \mathcal{L}_2} U_1^* U_3^* \frac{2T_1 \mathcal{V}_2^2 \mathcal{F} [(1 + I_2 \mathcal{L}_2/2) \mathcal{D}_3^* + \mathcal{D}_2(1 - \Gamma/i\Delta)/2]}{1 + I_2 \mathcal{F} \frac{\mathcal{Y}}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)}, \quad (7)$$

where following the notation of Ref. 12, the complex Lorentzian denominators \mathcal{D}_n are given by

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

the dimensionless Lorentzian \mathcal{L}_2 is

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

the dimensionless intensity I_2 is

$$I_2 = 4 |\mathcal{V}_2|^2 T_1 T_2,$$

the dimensionless "population pulsation" term \mathcal{F} is

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

and where $\mathcal{V}_2 = \mu \mathcal{E}_2 U_2 / 2\hbar$, μ is the electric-dipole matrix element, N is the total number of interacting atoms,

and $\Delta = \nu_2 - \nu_1$ is the beat frequency between modes one and two. These coefficients assume the only relaxation processes are upper-to-lower-level decay described by the decay constant $\Gamma (=1/T_1)$, and the dipole decay described by $\gamma (=1/T_2)$. This is the usual experimental situation in laser spectroscopy. For pure spontaneous decay, γ is equal to $\Gamma/2$. For more general coefficients, see Refs. 12 and 16.

The equation of motion (3) for the field reduced-density operator ρ and the expressions for the coefficients A_1 through D_1 are the fundamental equations of our theory. We may use Eq. (3) to obtain the equation of motion for the expectation value of any operator of the quantized fields 1 and 3. This was also done in Ref. 12. For example, the annihilation operator $\langle a_1 \rangle$ corresponds to the classical Fourier amplitude for the electric field of mode 1, \mathcal{E}_1 , and its equation of motion is given by

$$\begin{aligned} \frac{d}{dt} \langle a_1 \rangle &= \frac{d}{dt} \mathcal{E}_1 = \langle a_1 \dot{\rho} \rangle = \sum_n \langle n | a_1 \dot{\rho} | n \rangle \\ &= (A_1 - B_1 - \nu/2Q_1) \mathcal{E}_1 \\ &\quad + (C_1 - D_1) \mathcal{E}_3^*, \end{aligned} \quad (8)$$

where $\mathcal{E}_3^* = \langle a_3^\dagger \rangle$. Equation (8) is the semiclassical coupled-mode equation of motion for the field amplitude \mathcal{E}_1 . 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 $C_1 - D_1$ is the mode-coupling coefficient in phase conjugation and modulation spectroscopy. Each of these coefficients can be derived purely semiclassically.^{20,21}

Also of interest, especially for the calculation of squeezing, are the equations of motion for the number operator $a_1^\dagger a_1$ and combination tone operator $a_1 a_3$. These can be obtained in the same manner as for Eq. (8):

$$\begin{aligned} \frac{d}{dt} \langle a_1^\dagger a_1 \rangle &= \langle a_1^\dagger a_1 \dot{\rho} \rangle = \sum_n \langle n | a_1^\dagger a_1 \dot{\rho} | n \rangle \\ &= (A_1 - B_1 - \nu/2Q_1) \langle a_1^\dagger a_1 \rangle \\ &\quad + (C_1 - D_1) \langle a_3^\dagger a_1^\dagger \rangle + A_1 + \text{c.c.}, \end{aligned} \quad (9)$$

$$\begin{aligned} \frac{d}{dt} \langle a_1 a_3 \rangle &= \langle a_1 a_3 \dot{\rho} \rangle = \sum_n \langle n | a_1 a_3 \dot{\rho} | n \rangle \\ &= (A_1 - B_1 - \nu/2Q_1) \langle a_1 a_3 \rangle \\ &\quad + (C_1 - D_1) \langle a_3^\dagger a_3 \rangle + C_1 \\ &\quad + [1 \leftrightarrow 3]. \end{aligned} \quad (10)$$

In free space, no build up of photon number occurs, and $d \langle n_1 \rangle / dt = A_1 + A_1^*$. Thus, we interpret $A_1 + A_1^*$ given by Eq. (4) as the spectrum of resonance fluorescence and

it is identical to the expression first derived by Mollow.²² Similarly, the inhomogeneous term of Eq. (10), $C_1 + C_3$, is the quantum source contribution for the "combination tone"²³ $\langle a_1 a_3 \rangle$ and is responsible for squeezing.²⁴

III. QUANTUM LANGEVIN EQUATIONS AND THE GENERALIZED EINSTEIN RELATION

This section shows how to derive the quantum Langevin equations of motion for operators which the reduced density depends on. For the sidemode reduced-density operator of Sec. II, this means we can find equations of motion for the sidemode annihilation operators a_1 and a_3 as well as their adjoints and the various bilinear products of these operators. It is no longer possible to obtain Langevin equations for the atomic operators, since we have traced the atom-field density operator over the atomic coordinates in deriving the sidemode reduced-density operator.

The Langevin equation for an operator is immediately obtained by deriving the equation of motion for the expectation value of the operator as described in Sec. II and then removing the expectation value angle brackets and adding a noise operator with zero expectation value. This recipe does not tell us what the noise operator is, but all we typically need to know about the noise operator is its two-time correlation function. We assume the noise process is Markovian (δ correlated in time) with appropriate diffusion coefficients. The generalized Einstein relation^{1,18,19} reviewed in this section shows how these diffusion coefficients can be derived from the drift coefficients in the Langevin equations.

We start with the double sidemode example pertinent to multiwave mixing and then consider a more general context. Removing the expectation value angle brackets from Eq. (8), we have the Langevin equation for a_1 ,

$$\dot{a}_1 = (A_1 - B_1 - \nu/2Q_1) a_1 + (C_1 - D_1) a_3^\dagger + F_1, \quad (11)$$

and similarly

$$\dot{a}_3^\dagger = (A_3^* - B_3^* - \nu/2Q_3) a_3^\dagger + (C_3^* - D_3^*) a_1 + F_3^\dagger. \quad (12)$$

Here $F_1(t)$ and $F_3(t)$ are random rapidly varying noise operators with vanishing mean,

$$\langle F_1(t) \rangle = \langle F_3(t) \rangle = 0, \quad (13)$$

but δ -correlated two-time averages

$$\langle F_1^\dagger(t) F_1(t') \rangle = 2 \langle D_{a_1^\dagger a_1} \rangle \delta(t - t'), \quad (14)$$

$$\langle F_3(t) F_1(t') \rangle = 2 \langle D_{a_1 a_3} \rangle \delta(t - t'). \quad (15)$$

Here $\langle D_{a_1^\dagger a_1} \rangle$ and $\langle D_{a_1 a_3} \rangle$ are two of four important diffusion coefficients that describe the field operator correlations (the other two, $\langle D_{a_3^\dagger a_3} \rangle$ and $\langle D_{a_3^\dagger a_1^\dagger} \rangle$, are given by interchanging 1 and 3, and taking the complex conjugate, respectively). By using the delta functions in Eqs. (14) and (15), we have made the Markovian approximation. To find their values, we review the generalized Einstein relation.

Equations (11) and (12) are each of the general operator form¹⁸

$$\dot{A}_\mu = D_\mu(t) + F_\mu(t), \quad (16)$$

where A_μ is the quantum operator, D_μ is its drift term, and F_μ is the noise operator. From the identity

$$A_\mu(t) = A_\mu(t - \Delta t) + \int_{t-\Delta t}^t dt' \dot{A}_\mu(t'), \quad (17)$$

we have

$$\begin{aligned} \langle A_\mu(t) F_\nu(t) \rangle &= \langle A_\mu(t - \Delta t) F_\nu(t) \rangle \\ &+ \int_{t-\Delta t}^t dt' \langle [D_\mu(t') + F_\mu(t')] F_\nu(t) \rangle. \end{aligned} \quad (18)$$

Because the operator $A_\mu(t')$ at time t' cannot be affected by a fluctuation at a later time t , the first term on the right-hand side of Eq. (18) is zero. Similarly the correlation $\langle D_\mu(t') F_\nu(t) \rangle$ is zero except at the point $t'=t$, but the interval of integration is zero (set of measure zero). All that remains is

$$\begin{aligned} \langle A_\mu(t) F_\nu(t) \rangle &= \int_{t-\Delta t}^t dt' \langle F_\mu(t') F_\nu(t) \rangle \\ &= \frac{1}{2} \int_{-\infty}^{\infty} dt' \langle F_\mu(t') F_\nu(t) \rangle, \end{aligned} \quad (19)$$

where in the last step we have assumed the noise to be stationary. Substituting the equivalent of Eqs. (14) and (15) into (19) we have

$$\langle A_\mu(t) F_\nu(t) \rangle = \langle D_{\mu\nu} \rangle. \quad (20)$$

In an analogous manner we find

$$\langle F_\mu(t) A_\nu(t) \rangle = \langle D_{\mu\nu} \rangle. \quad (21)$$

We now may use Eqs. (20) and (21) to determine the equation of motion for the expectation value of $\langle A_\mu A_\nu \rangle$. From Eq. (16)

$$\begin{aligned} \frac{d}{dt} \langle A_\mu A_\nu \rangle &= \langle \dot{A}_\mu A_\nu \rangle + \langle A_\mu \dot{A}_\nu \rangle \\ &= \langle D_\mu A_\nu \rangle + \langle F_\mu A_\nu \rangle + \langle A_\mu D_\nu \rangle + \langle A_\mu F_\nu \rangle. \end{aligned}$$

Substituting Eqs. (20) and (21) and rearranging gives

$$2 \langle D_{\mu\nu} \rangle = \frac{d}{dt} \langle A_\mu A_\nu \rangle - \langle A_\mu D_\nu \rangle - \langle D_\mu A_\nu \rangle. \quad (22)$$

Equation (22) is called the "generalized Einstein relation." It shows that the diffusion coefficients $\langle D_{\mu\nu} \rangle$ are directly related to drift coefficients D_μ and D_ν and thus compromises a quantum fluctuation-dissipation theorem. This very useful equation makes it possible to calculate the diffusion coefficients immediately from the drift coefficients, provided one can independently calculate the equation of motion for $\langle A_\mu A_\nu \rangle$.

IV. APPLICATIONS TO THE QUANTUM THEORY OF MULTIWAVE MIXING

It is straightforward to apply Eq. (22) to Eqs. (9) and (10) to obtain the diffusion coefficients for the operators $a_1^\dagger a_1$ and $a_1 a_3$. From Eq. (11) we see that the drift term for a_1 is

$$D_{a_1} = (A_1 - B_1) a_1 + (C_1 - D_1) a_3^\dagger. \quad (23)$$

The drift term for a_1^\dagger is the adjoint of Eq. (23). Substituting these into Eq. (22) and using Eq. (9), we find

$$2 \langle D_{a_1^\dagger a_1} \rangle = A_1 + A_1^\dagger. \quad (24)$$

This remarkable result reveals that the diffusion coefficient (24) for the operator $a_1^\dagger a_1$ is simply the resonance fluorescence spectrum.²² Figure 2 graphs this well-known spectrum both for centrally tuned pump as well as a slightly detuned pump.

To find the diffusion coefficient for the combination-tone operator $a_1 a_3$, we note the operators a_3 and a_1 commute, and hence that the Einstein relation must be symmetric under an interchange of the indices 1 and 3. Thus,

$$2 \langle D_{a_1 a_3} \rangle = \frac{d}{dt} \langle a_1 a_3 \rangle - \langle D_{a_1 a_3} \rangle - \langle D_{a_3 a_1} \rangle.$$

With the help of Eq. (10), we obtain

$$2 \langle D_{a_1 a_3} \rangle = C_1 + C_3. \quad (25)$$

Once again the diffusion coefficient is the inhomogeneous term of an equation of motion (10), that is, it is the quantum source term for the combination tone operator $a_1 a_3$.

The coefficient $C_1 + C_3$ represents a quantum mechanical quantity new with our quantum theory of multiwave mixing and as shown in Ref. 24 it is responsible for squeezing. Since $C_3(\Delta) = C_1(-\Delta)$, $C_1 + C_3$ is a symmetric function of Δ , but unlike $A_1 + A_1^\dagger$ it is only real for a centrally tuned pump ($\nu_2 = \omega$). Figure 3 plots the real part of $C_1 + C_3$ vs ΔT_2 for the atomic detunings $\Delta_2 = \omega - \nu_2$ of 0, $4T_2^{-1}$, and $8T_2^{-1}$. $I_2 = 50$ and $T_2 = 2T_1$, the case of pure radiative decay. We note its similarity to the three peaked spectrum of resonance fluorescence,²² except that the central peak is inverted. Just as in the case of resonance fluorescence, the sideband peaks move outward with detuning and the central dip rapidly disap-

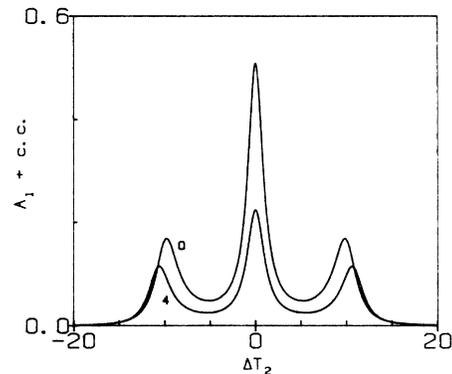


FIG. 2. Resonance fluorescence spectrum $A_1 + A_1^\dagger$ given by Eq. (3) vs the pump-sidemode beat frequency $\Delta = \nu_2 - \nu_1$ with pump both on and off resonance (elastic delta function at $\Delta = 0$ exists, but is not shown). Equation (23) shows that this spectrum is also the diffusion coefficient for $\langle F_1^\dagger F_1 \rangle$. The two curves are for the pump detunings $\nu_2 - \omega = 0, 4/T_2$, with pump intensity $I_2 = 50$, and $T_2 = 2T_1$ (pure radiative decay).

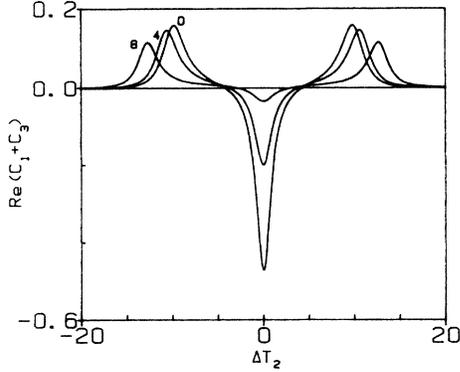


FIG. 3. Real part of combination-tone source term $C_1 + C_3$ vs ΔT_2 for pump detunings $\nu_2 - \omega = 0, 4T_2^{-1}$, and $8T_2^{-1}$, pump intensity $I_2 = 50$, and $T_2 = 2T_1$.

pears. The imaginary part of $C_1 + C_3$ is zero when $\nu_2 = \omega$, since in that case $C_3 = C_1^*$, but when the pump field at frequency ν_2 is detuned from atomic resonance, it is nonzero and can be much larger than the real part if $\Delta_2 \gg T_2^{-1}$. Figure 4 depicts the imaginary part of $C_1 + C_3$ vs ΔT_2 for the detunings of $4T_2^{-1}$ and $8T_2^{-1}$. The other parameters are the same as in Fig. 3. Note the dispersive character of the Rabi sidebands, just as in the case of a complex absorption coefficient.

Having obtained the diffusion coefficients of Eqs. (24) and (25), we may now compare them to the ones published by Drummond and Walls⁵ (DW) and by Reid and Walls.^{6,7} In each of their papers, their expressions for the diffusion coefficients are valid only in the degenerate limit, i.e., $\nu_1 = \nu_2$ or $\Delta = 0$. From Eqs. (24) and (23) in Ref. 5, the coefficients are

$$\Lambda(I_2) = \frac{Ng^2 I_2 \mathcal{L}_2^3}{\gamma(1 + I_2 \mathcal{L}_2)^3} \left[(1 + \delta^2)(1 - f) + I_2 [2 + \delta^2(1 - f)] + \frac{I_2^2}{2} \right] \quad (\text{DW}) \quad (26)$$

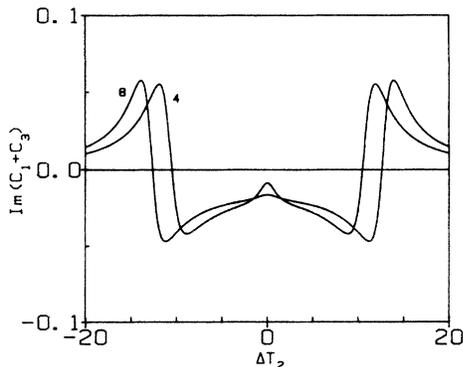


FIG. 4. Imaginary part of $C_1 + C_3$ vs ΔT_2 for $\nu_2 - \omega = 4T_2^{-1}$ and $8T_2^{-1}$, $I_2 = 50$, and $T_2 = 2T_1$.

for $2\langle D_{a_1 a_1} \rangle$ and

$$d(I_2) = \frac{Ng^2 I_2 \mathcal{L}_2^3}{\gamma(1 + I_2 \mathcal{L}_2)^3} \left[(1 - i\delta)^3 f + i\delta I_2 (1 - f)(1 - i\delta) + \frac{I_2^2}{2} \right] \quad (\text{DW}) \quad (27)$$

for $-2\langle D_{a_1 a_3} \rangle$ and where $\delta = (\omega - \nu_2)/\gamma$ and $f = \Gamma/2\gamma$. In transforming their notation to ours, $X \rightarrow I_2$, $(1 + \delta^2)^{-1} \rightarrow \mathcal{L}_2$, and $2Ck' \rightarrow Ng^2/\gamma$. Equations (26) and (27) certainly do not at first glance appear to be equivalent to the equations for A_1 and C_1 , Eqs. (4) and (6). Indeed, evaluating either of them at $\Delta = 0$ immediately leads to difficulties due to the presence of the $\Gamma/i\Delta$ term in each of them. In resonance fluorescence it is precisely that quantity that gives rise to the elastic portion of the spectrum, also called the Rayleigh peak, since $i/\Delta + \text{c.c.} = 2\pi\delta(\Delta)$, where $\delta(\Delta)$ is δ function. In Eqs. (26) and (27) there is no evidence of a delta function, and we are therefore led to assume that they represent only the inelastic portions of the spectrum. We now show that this is the case.

As first shown by Mollow,²² the separation of the spectrum into the elastic and inelastic portions is somewhat complicated, because the $\Gamma/i\Delta$ term also contributes to the inelastic spectrum when $\Delta \neq 0$. The elastic part of A_1 is found by letting $\Delta = 0$ in the $\Gamma/i\Delta$ term. This yields

$$A_{1\text{el}} = \frac{Ng^2}{4\gamma} \frac{I_2 \mathcal{L}_2}{(1 + I_2 \mathcal{L}_2)^2} \frac{\Gamma}{i\Delta} \quad (28)$$

The inelastic spectrum is defined to be

$$A_{1\text{inel}} = A_1 - A_{1\text{el}} \quad (29)$$

Substituting Eqs. (4) and (28) into (29) gives

$$A_{1\text{inel}} = \frac{Ng^2 \mathcal{D}_1}{1 + I_2 \mathcal{L}_2} \frac{\frac{I_2 \mathcal{L}_2}{2} + I_2 \frac{\gamma}{2} \mathcal{F}}{1 + I_2 \mathcal{F} \frac{\gamma}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} \left[\frac{I_2 \mathcal{L}_2}{2} \mathcal{D}_3^* + \mathcal{D}_2^*/2 \right] + \frac{Ng^2}{1 + I_2 \mathcal{L}_2} \frac{\gamma I_2}{4} \frac{\Gamma}{i\Delta} \times \left[\frac{\mathcal{F} \mathcal{D}_1 \mathcal{D}_2^*}{1 + I_2 \mathcal{F} \frac{\gamma}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} - \frac{\frac{\mathcal{L}_2}{\gamma^2}}{1 + I_2 \mathcal{L}_2} \right] \quad (30)$$

We now wish to evaluate $A_{1\text{inel}}$ at $\Delta = 0$. However, the second term in Eq. (30) becomes indeterminate when $\Delta = 0$. To evaluate it we use l'Hospital's rule and differentiate the numerator and denominator with respect to Δ . Doing this and evaluating at $\Delta = 0$, we find

$$A_{1\text{inel}}|_{\Delta=0} = \frac{Ng^2 I_2}{(1+I_2 \mathcal{L}_2)^3} \left[\frac{\mathcal{D}_2 \mathcal{L}_2}{2} (1-\Gamma/2\gamma) + \frac{I_2 \mathcal{L}_2^2}{2\gamma} \left[1 + \gamma \mathcal{D}_2 + \frac{i\Delta_2 \mathcal{D}_2^* \Gamma}{2\gamma} \right] + \frac{I_2^2 \mathcal{L}_2^3}{4\gamma} \right], \quad (31)$$

where $\Delta_2 = \omega - \nu_2$. Upon taking the complex conjugate of Eq. (31) and adding, we obtain Drummond and Walls's⁵ result of Eq. (26).

In a similar manner, we relate the "inelastic" part of $C_1 + C_3$ to the diffusion coefficient $2\langle D_{a_1 a_3} \rangle$. From Eq. (6), the elastic portion of C_1 is given by

$$C_{1\text{el}} = \frac{Ng^2 I_2 \gamma \mathcal{D}_2^2}{4(1+I_2 \mathcal{L}_2)^2} \frac{\Gamma}{i\Delta}. \quad (32)$$

Note that unlike $A_{1\text{el}}$, $C_{1\text{el}}$ is a complex number divided by $i\Delta$, implying that the imaginary part of that number diverges when $\Delta=0$ (only the real part gives the δ function). However, recall that we also must add C_3 to C_1 , and because $C_3(\Delta) = C_1(-\Delta)$, that diverging term vanishes. C_1 by itself is not a meaningful quantity; only when it is added to C_3 or $-D_1$ does it acquire physical significance. $C_{1\text{inel}}$ is defined like $A_{1\text{inel}}$, so

$$C_{1\text{inel}} = C_1 - C_{1\text{el}}. \quad (33)$$

Substituting Eqs. (6) and (32) into (33), we find

$$C_{1\text{inel}} = -\frac{Ng^2 \mathcal{D}_1}{1+I_2 \mathcal{L}_2} \frac{I_2 \frac{\gamma}{2} \mathcal{F} \left[\frac{I_2 \mathcal{L}_2}{2} \mathcal{D}_3^* - \mathcal{D}_2/2 \right]}{1+I_2 \mathcal{F} \frac{\gamma}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} + \frac{Ng^2}{1+I_2 \mathcal{L}_2} \frac{\gamma I_2 \mathcal{D}_2}{4} \frac{\Gamma}{i\Delta} \times \left[\frac{\mathcal{F} \mathcal{D}_1}{1+I_2 \mathcal{F} \frac{\gamma}{2} (\mathcal{D}_1 + \mathcal{D}_3^*)} - \frac{\mathcal{D}_2}{1+I_2 \mathcal{L}_2} \right]. \quad (34)$$

The second term is again indeterminate when $\Delta=0$. Applying l'Hospital's rule and evaluating at $\Delta=0$, we eventually find

$$C_{1\text{inel}}|_{\Delta=0} = -\frac{Ng^2 \gamma \mathcal{D}_2 I_2}{4(1+I_2 \mathcal{L}_2)^3} \left[\Gamma \mathcal{D}_2^2 + I_2 \mathcal{L}_2^2 \frac{i\Delta_2}{\gamma^2} (1-\Gamma/2\gamma) + I_2^2 \mathcal{L}_2^2 \mathcal{D}_2^* \right]. \quad (35)$$

$C_3(\Delta=0) = C_1(\Delta=0)$, so multiplying Eq. (35) times two and inserting the appropriate expressions for \mathcal{D}_2 and \mathcal{D}_2^* recovers Drummond and Walls's⁵ result of Eq. (27).

Reid and Walls⁶ considered squeezing in degenerate four-wave mixing. In order to account for the spatial hole burning (SHB) of the two counterpropagating pump beams, they integrated their diffusion coefficients over the intensity distribution

$$I_2 \rightarrow 4I_2 \cos^2(\mathbf{K}_2 \cdot \mathbf{r}) = 2I_2 [1 + \cos(2K_2 z)]$$

as

$$\langle D_{a_1 a_3} \rangle_{\text{SHB}} = \frac{1}{2\pi} \int_0^{2\pi} \langle D_{a_1 a_3} \rangle d(2K_2 z). \quad (36)$$

Substituting Eqs. (26) and (27) into Eq. (36), and assuming $f=1$ (pure radiative decay), we recover the expressions in Eq. (3) of Ref. 6. In our third paper in this series,¹⁴ we performed the same type of average for all of the coefficients A_1 , B_1 , C_1 , and D_1 , as well as averages over inhomogeneous broadening and Gaussian transverse profiles.

We have thus shown that the diffusion coefficients of Drummond and Walls⁵ and of Reid and Walls⁷ are, in fact, the inelastic parts of the resonance fluorescence spectrum and the $C_1 + C_3$ coefficient. Our theory reveals that the diffusion coefficients should contain elastic contributions as well. It is well known that for either low pump intensities or high detunings, i.e., for $I_2 \mathcal{L}_2 \ll 1$, the elastic part of the spectrum dominates the emission.^{3,22} Specifically, $I_{\text{el}}/I_{\text{tot}} = 1/(1+I_2 \mathcal{L}_2)$. Since this is the parameter range Reid and Walls consider for optimum squeezing, these contributions should be included in the degenerate theory. The effects of the elastic component is planned to be considered in the next paper in this series.

V. COMPARISON BETWEEN METHODS OF SOLUTIONS

Equations (9) and (10) are linear, coupled, ordinary differential equations, and they may be solved by the usual techniques, such as the Laplace transform. Equations (11) and (12), on the other hand, are linear, coupled, stochastic differential equations, due to the noise operators $F_1(t)$ and $F_3(t)$. Their solution require the introduction of a new type of calculus, the Ito calculus. A good, recent discussion of Ito and other types of calculus is given by Gardiner.²⁵ In this section we show that solutions obtained by this method and our reduced density operator method produce the same results. We use a simple one dimensional problem, but the two-dimensional coupled-mode problem is a straightforward extension.

Consider the simple stochastic differential equation

$$\frac{da}{dt} = -\alpha a + F(t), \quad (37)$$

where $a(t)$ is an operator, α a complex number independent of t , and $F(t)$ the noise operator satisfying

$$\langle F^\dagger(t)F(t') \rangle = A\delta(t-t'). \quad (38)$$

The general solution to Eq. (37), from the Ito calculus [using the standard integration factor $\exp(\alpha t)$], is given in Ref. 25, p. 105 as

$$a(t) = a(0)e^{-\alpha t} + \int_0^t dt' e^{-\alpha(t-t')} F(t'). \quad (39)$$

From Eq. (39) we may calculate $\langle a^\dagger(t)a(t) \rangle$ as

$$\begin{aligned} \langle a^\dagger(t)a(t) \rangle &= \langle a^\dagger(0)a(0) \rangle e^{-(\alpha+\alpha^*)t} + \int_0^t dt'' e^{-\alpha(t-t'')} e^{-\alpha^*t'} \langle a^\dagger(0)F(t'') \rangle \\ &+ \int_0^t dt' e^{-\alpha^*(t-t')} e^{-\alpha t} \langle F^\dagger(t')a(0) \rangle + \int_0^t \int_0^t dt' dt'' e^{-\alpha^*(t-t')} e^{-\alpha(t-t'')} \langle F^\dagger(t')F(t'') \rangle. \end{aligned} \quad (40)$$

The second and third terms of Eq. (40) are zero because, once again, a fluctuation at time t' or t'' cannot be correlated to the operator at the future time t . Inserting Eq. (38) into the last term of (40) and integrating, we obtain

$$\begin{aligned} \langle a^\dagger(t)a(t) \rangle &= \langle a^\dagger(0)a(0) \rangle e^{-(\alpha+\alpha^*)t} \\ &+ \frac{A}{\alpha+\alpha^*} [1 - e^{-(\alpha+\alpha^*)t}] \end{aligned} \quad (41)$$

as our solution.

Alternatively, we can use a reduced-density operator to obtain the differential equation for $\langle a^\dagger a \rangle$ directly

$$\frac{d\langle a^\dagger a \rangle}{dt} = -(\alpha+\alpha^*)\langle a^\dagger a \rangle + A. \quad (42)$$

In fact, this follows from Eq. (9) by dropping the a_3 contributions, where $\alpha = B_1 + \nu/2Q_1 - A_1$. Equation (42) can be easily solved to yield Eq. (41). We have thus illustrated the equivalence between the two techniques. The solutions of the stochastic differential equations of Langevin theory can also be obtained by solving the equations of motion derived from Eq. (3).

In conclusion, we have demonstrated that the Langevin diffusion coefficients are identical to the inhomogeneous source terms of our quantum coupled-mode equations (10) and (11). In particular, the diffusion coefficient labeled Λ

by Reid and Walls⁶ is, in fact, the inelastic portion of the resonance fluorescence spectrum $A_1 + A_1^*$ evaluated at $\nu_1 = \nu_2$. Thus, the drift and diffusion coefficients for non-degenerate four-wave mixing have already been published in the earlier papers of this series, although they were not named as such. The present analysis gives another demonstration that our quantum theory of multiwave mixing is fully applicable to the study of squeezed states, and with its automatic nondegeneracy and inclusion of the elastic contributions, represents a significant generalization. The diffusion coefficients derived here can be substituted into Reid and Walls⁶ to obtain nondegenerate squeezing. The completely independent derivations of these coefficients by our method and by that of Reid and Walls helps to confirm the validity of the results. The next three papers in this series will treat the quantum theory of four-wave mixing in detail uniformly using the reduced density operator approach for one- and two-photon two-level media.

ACKNOWLEDGMENTS

Work supported in part by the U.S. Office of Naval Research, in part by the U.S. Army Research Office, and in part by the U.S. Air Force Office of Scientific Research.

¹M. Lax, in *Brandeis University Summer Institute Lectures, 1966*, edited by M. Chretien, E. P. Gross, and S. Deser (Gordon and Breach, New York, 1968), Vol. II; Phys. Rev. **145**, 110 (1966).

²See, for example, H. Haken, *Laser Theory* (Springer, Berlin, 1970).

³C. Cohen-Tannoudji, in *Frontiers in Laser Spectroscopy*, edited by R. Balian, S. Haroche, and S. Lieberman (North-Holland, Amsterdam, 1977).

⁴R. Bonifacio and L. A. Lugiato, Phys. Rev. A **18**, 1129 (1978).

⁵P. D. Drummond and D. F. Walls, Phys. Rev. A **23**, 2563 (1981).

⁶M. Reid and D. F. Walls, Phys. Rev. A **31**, 1622 (1985). Note: Reid and Walls have presented nondegenerate numerical results, [Proceedings of the MIT Workshop on Squeezed States, 1985, edited by P. Kumar and J. M. Shapiro (unpublished)], but have not yet published the corresponding formulas.

⁷M. Reid and D. F. Walls, Phys. Rev. A **32**, 396 (1985).

⁸M. O. Scully and W. E. Lamb, Jr., Phys. Rev. **159**, 208 (1967).

⁹S. Stenholm, Phys. Rep. **C6**, 1 (1973).

¹⁰S. Singh and M. S. Zubairy, Phys. Rev. A **21**, 281 (1980).

¹¹M. Sargent III, M. S. Zubairy, and F. DeMartini, Opt. Lett. **8**, 76 (1983).

¹²M. Sargent III, D. A. Holm, and M. S. Zubairy, Phys. Rev. A **31**, 3112 (1985). (This is the first in the series entitled "Quantum theory of multiwave mixing," which we abbreviate

QTMWM I.)

¹³S. Stenholm, D. A. Holm, and M. Sargent III, Phys. Rev. A **31**, 3124 (1985). (QTMWM II.)

¹⁴D. A. Holm, M. Sargent III, and L. M. Hoffer, Phys. Rev. A **32**, 963 (1985). (QTMWM III.)

¹⁵D. A. Holm, M. Sargent III, and S. Stenholm, J. Opt. Soc. Am. B **2**, 1456 (1985). (QTMWM IV.)

¹⁶D. A. Holm and M. Sargent III, Phys. Rev. A **33**, 1073 (1986). (QTMWM V.)

¹⁷D. A. Holm and M. Sargent III, J. Opt. Soc. Am. B (QTMWM VI.)

¹⁸M. Sargent II, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Mass., 1974), Chaps. 19 and 20.

¹⁹M. O. Scully, Phys. Rev. Lett. **55**, 2802 (1985).

²⁰M. Sargent III, Phys. Rep. **43C**, 223 (1978).

²¹T. Fu and M. Sargent III, Opt. Lett. **4**, 366 (1979).

²²B. R. Mollow, Phys. Rev. **188**, 1969 (1969).

²³We use the term "combination tone" since the operator $a_1 a_3$ corresponds to the semiclassical combination tone of W. E. Lamb, Jr., Phys. Rev. **134**, A1429 (1964).

²⁴D. A. Holm, M. Sargent III, and B. A. Capron, Opt. Lett. (to be published).

²⁵C. W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences* (Springer, New York, 1983).