Quantum theory of nondegenerate multiwave mixing. II. Adiabatic elimination via slowly varying amplitude approximation

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In this paper, we continue our development of the quantum theory of nondegenerate multiwave mixing in an atomic medium [Phys. Rev. A 37, 2017 (1988)]. In our development, the atomic variables are eliminated using a frequency-domain approach employing a slowly varying amplitude approximation that is more rigorous than the usual adiabatic approximation. We then specialize to the case of four-wave interaction with two strong pump beams and obtain noise correlations of the atomic polarization that are applicable to nondegenerate four-wave mixing. The noise correlations include the effect of atomic collisions that is crucial to the atomic-vapor experiments. After making the usual rotating-wave approximation, the atomic-polarization equations give us a set of temporal coupled-mode equations for the c-number variables corresponding to the annihilation operators. We then further specialize to the single-beam case in which all the relevant modes of interest are collinear and obtain a paired set of coupled-mode equations. In order to apply the theory to experiments employing traveling-wave interaction geometries, in the following paper of this series we will present a formalism to treat the spatial propagation of a quantum field.

I. INTRODUCTION

In the first paper\(^1\) of this series on the quantum theory of nondegenerate multiwave mixing, hereafter referred to as I, we considered the explicit dynamics of a subset of the field quantization modes interacting with a system of stationary two-level atoms. Because we made the realistic assumption of leaving the remaining infinite set of field modes as a common thermal-field reservoir, the resulting Langevin equations contained extra decay terms that are due to collective spontaneous emission or superradiance. We further showed that these superradiance terms are negligible when the atomic medium is saturated by a pump mode and either (i) the number of atoms in a diffraction volume is small, or (ii) the atoms are pumped far off resonance. Neglecting the superradiance terms and using a general Fourier-expansion technique, we then obtained a solution for the c-number atomic polarization variable \(V(t)\) in terms of slowly varying modal amplitudes \(\{A_k(t)\}\). No adiabatic approximation was made up to this point.

Even though the focus of our attention has been on two-level atoms, the formalism presented in I is still too general to be applied directly to problems of interest. The purpose of this paper is to further develop the theory so that it can be applied directly to quantum-optics experiments employing traveling-wave interaction geometries.\(^2\) A summary of this development was recently reported in a Rapid Communication.\(^3\) In this paper, we express the atomic polarization \(V(t)\) in terms of slowly varying modal amplitudes instead of the Fourier amplitudes \(\{A_k(t)\}\). This is necessary for us to derive a set of coupled-mode equations for the traveling waves. The slowly varying assumption is similar to the usual adiabatic approximation, but is more rigorous.

We start, in Sec. II, by recapitulating the pertinent results of I.\(^1\) This is done for the sake of completeness and to reestablish the notation. The remainder of this paper can be divided into two parts. In part one, comprising Secs. III and IV, we solve for the c-number atomic-polarization–density variable \(V(t)\) in terms of the field-mode variables \(\{a_q(t)\}\) using a slowly varying amplitude approximation that is more rigorous than the usual adiabatic approximation.\(^4\) It correctly takes into account the frequency shift of the modes due to the medium refractive index in that all the coefficients are evaluated at the shifted mode frequencies. It also gives extra terms that are necessary to obtain a correct expression for the group velocity in the medium when spatial propagation is considered. This point will be discussed at length in the following paper of this series. The deterministic part of the solution for \(V(t)\), when expanded up to the third order, agrees with the expressions given by Boyd et al.\(^5\) for a much simpler case and the Langevin noise part

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gives the correct resonance fluorescence spectrum as first obtained by Mollow.\(^\text{5}\)

In part two, consisting of Secs. V and VI, using the rotating-wave approximation, we derive a set of general temporal coupled-mode equations for the slowly varying amplitudes of \(\{a_j(t)\}\) denoted by \(\{a_j(t)\}\). In Sec. V, we do so for the special case in which there are only two frequency-degenerate undepleted pump modes. In Sec. VI, we further specialize to the case in which the wave vectors of all the modes are collinear and the medium is of infinite extent. In this case the set of general temporal coupled-mode equations reduces to a paired set of coupled-mode equations.

II. RECAPITULATION OF PERTINENT RESULTS OF I

After neglecting the superradiant terms, the following c-number Langevin equations were obtained in Sec. VI of I (Ref. 1):

\[
\frac{\partial a_{j}}{\partial t} = -i \omega_{a} a_{j} + \sum_{i} C_{j}^{*}(r_{i}) V_{i}, \tag{2.1}
\]

\[
\frac{\partial V_{i}}{\partial t} = -i \omega_{a} V_{i} - a \mathbf{r}(r_{i},t)(n_{i} - n_{j}) - \gamma_{1} V_{i} + f_{V_{i}}, \tag{2.2}
\]

\[
\frac{\partial V_{i}^{+}}{\partial t} = i \omega_{a} V_{i}^{+} - a^{+}(r_{i},t)(n_{i} - n_{j}) - \gamma_{1} V_{i}^{+} + f_{V_{i}^{+}}, \tag{2.3}
\]

\[
\frac{\partial n_{i}}{\partial t} = -[a^{+}(r_{i},t)V_{i} + a(r_{i},t)V_{i}^{+}] - \gamma_{1} n_{i} + f_{n_{i}}, \tag{2.4}
\]

and \(n_{i} = 1 - n_{j}\). Here, \(\{a_{j} : 1 \leq j \leq q\}\) are the c-number variables associated with the annihilation operators \(\{a_{j}\}\) of the \(q\) x-polarized field modes whose explicit dynamics are of interest; \(V_{i}, V_{i}^{+}, n_{i}\), and \(n_{g}\) are the c-number variables associated with the atomic down-transition, up-transition, upper-level occupation, and ground-level occupation operators, respectively, for the \(i\)th atom; \(\gamma_{1}\) and \(\gamma_{1} = 2F \gamma_{1}, 0 \leq F \leq 1\), are the atomic transverse and longitudinal relaxation rates, respectively, with the collision factor \(F\) taking a unit value in the absence of atomic collisions; the coefficients \(C_{j}(r_{i})\) are given by

\[
C_{j}(r_{i}) = g_{j} \mu_{d} \exp(i \mathbf{k}_{j} \cdot \mathbf{r}_{i}), \tag{2.5}
\]

\[
g_{j} = (\omega_{a}^{2} / 2 \hbar \epsilon_{g}(\omega_{a}) V_{Q})^{1/2}, \tag{2.6}
\]

with \(\mu_{d}\) being the dipole matrix element defined via

\[
\mu_{d} = \langle x | \hat{r}_{i} | g \rangle \cdot \mathbf{e}_{x}, \tag{2.7}
\]

where \(|g\rangle\) is the ground state and \(|x\rangle\) is the upper state of the \(i\)th atom that is coupled to the \(x\) polarization of the field; \(\hat{r}_{i}\) is the position-vector operator of the electron in

the \(i\)th atom with \(e\) the electron charge \((e < 0)\); \(\omega_{a}\) is the resonance angular frequency of the stationary two-level atoms and \(\omega_{j} = c |\mathbf{k}_{j}|\), where \(\mathbf{k}_{j}\) \((1 \leq j \leq q)\) is the free-space wave vector of the \(j\)th field mode defined via periodic boundary conditions over the quantization volume \(V_{Q}\); the multimode field variables \(a(r_{i},t)\) and \(a^{+}(r_{i},t)\) are defined via

\[
a(r_{i},t) = \sum_{j} C_{j}(r_{i}) a_{j}(t), \tag{2.8}
\]

\[
a^{+}(r_{i},t) = \sum_{j} C_{j}^{*}(r_{i}) a_{j}^{+}(t), \tag{2.9}
\]

and the Langevin noise forces \(f_{V_{i}}, f_{V_{i}^{+}}, \) and \(f_{n_{i}}\) have the following nonzero correlations:

\[
\langle f_{V_{i}^{+}}(t) f_{V_{i}^{+}}(t') \rangle = 2 \gamma_{1} (1 - F) n_{i}(t) \delta(t - t'), \tag{2.10}
\]

\[
\langle f_{V_{i}}(t) f_{V_{i}}(t') \rangle = a(r_{i},t)V_{i}(t) \delta(t - t'), \tag{2.11}
\]

\[
\langle f_{V_{i}^{+}}(t) f_{V_{i}^{+}}(t') \rangle = a^{+}(r_{i},t)V_{i}^{+}(t) \delta(t - t'), \tag{2.12}
\]

\[
\langle f_{n_{i}}(t) f_{n_{i}}(t') \rangle = \langle -[a^{+}(r_{i},t)V_{i}(t) + a(r_{i},t)V_{i}^{+}(t)]
\]
\[
+ \gamma_{1} n_{i}(t) \delta(t - t') \rangle, \tag{2.13}
\]

\[
\langle f_{n_{j}}(t) f_{n_{j}}(t') \rangle = -\langle f_{n_{i}}(t) f_{n_{i}}(t') \rangle
\]
\[
= \langle f_{n_{i}}(t) f_{n_{i}}^{+}(t') \rangle. \tag{2.14}
\]

We note that Eq. (2.1) has been obtained with the near-resonant approximation, which replaces \(\partial V_{i} / \partial t\) by \(-i \omega_{a} V_{i}\), and the rotating-wave approximation, which neglects \(\partial V_{i}^{+} / \partial t\). The expression for Eq. (2.1) without the near-resonant approximation has \((i / \omega_{a}) \partial V_{i}(t) / \partial t\) in place of \(V_{i}(t)\). The set of Langevin equations (2.1)–(2.4) were solved in Sec. VI of I (Ref. 1) by Fourier expanding \(a(r_{i},t)\) and \(a^{+}(r_{i},t)\) in terms of an infinite number of Fourier coefficients \(A_{n_{i}}(r_{i})\) and \(A_{n_{i}}^{+}(r_{i})\), respectively, as

\[
a(r_{i},t) = \sum_{m} A_{m}(r_{i}) \exp(-i \nu_{m} t), \tag{2.15}
\]

\[
a^{+}(r_{i},t) = \sum_{m} A_{m}^{+}(r_{i}) \exp(i \nu_{m} t), \tag{2.16}
\]

where \(\nu_{m} = 2 \pi m / T\) with \(T\) being an arbitrary period of interest. The atomic variables were also Fourier transformed to enable us to obtain a series solution for \(V_{i}(t)\), which was written in a compact form as follows:

\[
V_{i}(t) = \frac{(2 \gamma_{1} F) A_{m}(r_{i}) \exp(-i \nu_{m} t)}{D^{-}(\nu_{m} + \nu_{m} + \omega) \Pi_{m}^{-}(\nu_{m}, t) + \Gamma V_{i}(t), \tag{2.17}
\]

where \(\Pi_{m}(\omega, t)\) is defined recursively as

\[
\Pi_{m}(\omega, t) = 2 \gamma_{1} F - i(\omega - \nu_{m}) + \frac{2 A_{m}^{+}(r_{i}) A_{m}(r_{i}) \exp[i(\nu_{m} - \nu_{m}) t] \Pi_{m}(\omega, t) D^{-}(\omega)}{D^{-}(\nu_{m} + \omega, \omega, t) \Pi_{m}^{-}(\nu_{m}, t) + \Pi_{m}^{+}(\nu_{m}, \omega, t)} \times \frac{1}{D^{-}(\nu_{m} + \omega, \omega, t) + D^{+}(\nu_{m} + \omega, \omega, t)} \tag{2.18}
\]
with
\[ D^±(ω) \equiv -i(ω ± ω_d) + γ_1. \tag{2.19} \]

In Eq. (2.17), \( Γ_V(t) \) is a Langevin noise term, which can be related to its Fourier transform \( Γ_V(ω) \) by
\[ Γ_V(t) = \int_{-∞}^{∞} dω \ Γ_V(ω) \exp(-iωt), \tag{2.20} \]
where \( Γ_V(ω) \) was obtained in Sec. VI of I (Ref. 1) up to the third order in the field amplitudes \( \{ A_m(τ) \} \), and is given by
\[ Γ_V(ω) = \frac{2A_m(τ) A^*_m(τ) f_{τ+m}}{D^+_0 \pi - D^-_m} - \frac{2A_m(τ) A^*_m(τ) f_{τ-m}}{D^+_0 \pi - D^-_m} + \frac{2A_m(τ) f_{τ-m}}{D^+_0 \pi - D^-_m} + \frac{f_0}{D^+_0}. \tag{2.21} \]

Here, the subscript \( i \) on the Langevin forces has been dropped and the following definitions have been employed:
\[ D^+_0 ≡ D^+(ω ± v_1), \tag{2.22} \]
\[ D^-_0 ≡ D^±(ω), \tag{2.23} \]
\[ π(ω) ≡ \left[ −iω + 2F_1 + 2A_k^+ A_k \left( \frac{1}{D_k^+} + \frac{1}{D_k^-} \right) \right], \tag{2.24} \]
\[ π_0 ≡ π(ω), \tag{2.25} \]
\[ π_0 ± π(ω ± v_k), \tag{2.26} \]
\[ X_0 ≡ X(ω), \tag{2.27} \]
\[ X_{±±m} ≡ X(ω ± v_m), \tag{2.28} \]
for \( X \in \{ f_{τ+m}, f_{τ-m}, f_0 \} \).

### III. SLOWLY VARYING AMPLITUDE APPROXIMATION

The compact solution for the polarization \( V_i(t) \) of the \( i \)th atom given by Eqs. (2.17) and (2.18) is of the following form:
\[ V_i(t) = \sum Y(v_0, v_m, r_i, t) A_m(τ) \exp(-iv_mτ) + Γ_V(τ), \tag{3.1} \]
where \( Y(v, v_m, r_i, t) \) is a function of \( \{ A_m(τ) \} \) through the recursive function \( Π_m \). In arriving at this compact form of \( V_i(t) \), no further approximation beyond the convergence assumption of the series expansion has been made.\(^1\)

However, to solve for \( a_j(t) \) using Eq. (2.1), we need to further express \( V_i(t) \) in terms of \( \{ a_j(t) \} \). This cannot be done, in general, without further approximation and is the subject of this section.

Let us assume that \( a_j(t) \) can be written in the form
\[ a_j(t) = α_j(t) \exp[-i(ω_j - Δω_j)t], \tag{3.2} \]
where \( α_j(t) \) is slowly varying with changes occurring over time scales of the order of \( γ_1^{-1} \), \( ω_j \) is the free-field mode frequency, and \( Δω_j \) is an anticipated frequency shift due to interaction with the medium. Our \textit{a priori} inclusion of \( Δω_j \) in Eq. (3.2) is crucial to obtaining the correct result when the dielectric constant of the medium is large. Without such inclusion, the atomic polarization would be evaluated at the wrong, unshifted frequency. Equations (2.8) and (2.9) then can be written as
\[ a(τ+iτ) = \sum C_j(τ_i) α_j(t) \exp(-iω_j^t), \tag{3.3} \]
\[ a^+(τ+iτ) = \sum C_j^+(τ_i) α_j^+(t) \exp(iω_j^t), \tag{3.4} \]
where
\[ ω_j^t ≡ ω_j - Δω_j. \tag{3.5} \]

However, \( a(τ+iτ) \) and \( a^+(τ+iτ) \) can also be expressed in terms of the Fourier amplitudes \( \{ A_m(τ), A_m^*(τ) \} \) via Eqs. (2.15) and (2.16), respectively, which in the continuum limit become
\[ a(τ+iτ) = \int_{-∞}^{∞} dv A(v, τ) \exp(-ivτ), \tag{3.6} \]
\[ a^+(τ+iτ) = \int_{-∞}^{∞} dv A^+(v, τ) \exp(ivτ), \tag{3.7} \]
where \( A(v, τ) \) with \( δv = v_{m+1} - v_m \). Using the usual procedure of converting a product in the frequency domain to a convolution in the time domain, the deterministic part of \( V_i(t) \) in Eq. (3.1) can be written as
\[ V_i(t) = \int_{-∞}^{∞} dt' \langle 1/2πτ \rangle \tilde{Y}(t', τ) a(τ, t-t') \tag{3.8} \]
where \( \tilde{Y}(t', τ) \) is the inverse Fourier transform defined via
\[ \tilde{Y}(t', τ) \equiv \int_{-∞}^{∞} dv \ Y(v, τ) \exp(-ivt'). \tag{3.9} \]

We see from Eqs. (2.17) and (2.19) that the magnitude of \( Y(v, τ) \) has a peak at \( v ≈ ω_0 \) of width \( Δv ≈ γ_1^{-1} \) due to the presence of \( D^-(v) \) in the denominator. This means that \( Y(t', τ) \) must be a decaying sinusoid in \( t' \) with a decay time \( ≈ γ_1^{-1} \). It is thus sufficient to approximate \( a(τ, t-t') \) in Eq. (3.8) with an expansion around \( t' = 0 \) up to \( t' = γ_1^{-1} \). Moreover, in order for the expansion to be convergent, we assume that \( γ_1^{-1} ||\partial α_j(t)/∂t|| ≪ α_j(t) \). This assumption implies that the pump beam cannot have a linewidth broader than \( γ_1 \). When \( α_j(t) \) is sufficiently slowly varying, a good approximation is obtained by expanding the slowly varying amplitude \( α_j(t-t') \) in Eq. (3.3) up to the first order as
\[ α_j(t-t') \frac{∂α_j(t)}{∂t}, \tag{3.10} \]
giving
\[ a(τ, t-t') \approx \sum_j C_j(τ_i) α_j(t) \exp[-iω_j^t(t-t')] \]
\[ -t' \sum_j C_j(τ_i) \frac{∂α_j(t)}{∂t} \exp[-iω_j^t(t-t')], \tag{3.11} \]
which upon substituting into Eq. (3.8) yields

\[ V_j(t) = \sum_j Y(\omega_j^*, r_j, t) C_j(r_j) \alpha_j(t) \exp(-i\omega_j^* t) \]

\[ + \sum_j i \frac{\partial Y(\omega_j^*, r_j, t)}{\partial \omega_j^*} C_j(r_j) \frac{\partial \alpha_j(t)}{\partial t} \exp(-i\omega_j^* t) , \]

\[ \text{(3.12)} \]

where

\[ \frac{\partial \alpha_j(t)}{\partial t} \equiv \frac{\partial \alpha_j(t')}{\partial t'} \Bigg|_{t'=t} , \]

\[ \text{(3.13)} \]

and

\[ \frac{\partial Y(\omega_j^*, r_j, t)}{\partial \omega_j^*} \equiv \frac{\partial Y(\omega_j, r_j, t)}{\partial \omega} \bigg|_{\omega = \omega_j^*} \]

\[ \text{(3.14)} \]

Equation (3.12) can be abbreviated as

\[ V_j(t) = \sum_j C_j(r_j) \exp(-i\omega_j^* t) V_j(t) Y(j, r_j, t) , \]

\[ \text{(3.15)} \]

where \( D_j(t) \) is a differential operator defined by

\[ D_j(t) \equiv \alpha_j(t) + i \frac{\partial \alpha_j(t)}{\partial t} \frac{\partial}{\partial \omega_j^*} \]

\[ \text{(3.16)} \]

We thus arrive at the following simple result: the slowly varying amplitude approximation allows us to replace \( \sum_m A_m(r) \exp(-i\nu_m t) \) in Eq. (3.1) by \( \sum_j C_j(r) \exp(-i\omega_j^* t) D_j(t) \). As noted earlier \( Y(\nu_m, r_j, t) \) in Eq. (3.1) also depends upon sums like \( \sum_m A_m(r) \exp(-i\nu_m t) \) via the recursive function \( \Pi_m \) of Eq. (2.18). These sums can be approximated in a similar way giving the following result for \( V_j(t) \):

\[ V_j(t) = - \sum_j C_j(r_j) \exp(-i\omega_j^* t) \]

\[ \times D_j(t) \left[ \frac{2\gamma F}{D(-\omega_j^*) \Pi_j(\omega_j^*, r_j, t)} \right] + \Gamma V_j(t) , \]

\[ \text{(3.17)} \]

where the differential operator \( \Pi_j(\omega, r_j, t) \) is, once again, defined recursively as

\[ \Pi_j(\omega, r_j, t) = 2\gamma F - i(\omega - \omega_j^*) + 2 \sum_{l,n} C_l^*(r_j) \exp(i\omega_j^* t) D_l^*(t) C_n(r_j) \exp(-i\omega_n^* t) D_n(t) \]

\[ \times \frac{\Pi_j(\omega, r_j, t) D^{-}(\omega)}{D^{-}(\omega_j^* + \omega_n^* + \omega) \Pi_j(\omega_j^* - \omega_j^* - \omega_n^* + \omega, r_j, t)} \]

\[ \times \left[ \frac{1}{D^{-}(\omega_j^* + \omega - \omega_j^* + \omega)} + \frac{1}{D^{+}(-\omega_j^* + \omega - \omega_j^*)} \right] , \]

\[ \text{(3.18)} \]

with

\[ D_j(t) \equiv \alpha_j(t) - i \frac{\partial \alpha_j(t)}{\partial t} \frac{\partial}{\partial \omega_j^*} \]

\[ \text{(3.19)} \]

Note that \( D_j(t) \) is like the “Hermitian conjugate” of \( D_j(t) \). Unlike Eqs. (2.17) and (2.18), the summing indices in Eqs. (3.17) and (3.18) now range from 1 to \( q \) denoting sums over the \( q \) field modes \( \{ a_j(t); 1 \leq j \leq q \} \).

For later convenience, let us define a polarization density \( V(r, t) \) by

\[ V(r, t) \equiv \sum_{r_i \in \delta v} \frac{V_{r_i}(t)}{\delta v} , \]

\[ \text{(3.20)} \]

where \( V_{r_i}(t) \equiv V(t) \), and the sum is taken over all atoms contained in a microscopic volume element \( \delta v \) at \( r \) whose linear dimensions are assumed smaller than a wavelength. If we further assume that the \( N_0 \) atoms within \( \delta v \) are homogeneously distributed and similarly excited, then the polarization density can be written as

\[ V(r, t) = \frac{N_0}{\delta v} V(t) = \rho V(t) , \]

\[ \text{(3.21)} \]

where \( \rho \) is the atomic number density. Equivalently, \( V(t) \) can be taken as an average by setting

\[ V(t) = \frac{\sum_{i=1}^{N_0} V_{r_i}(t)}{N_0} , \]

\[ \text{(3.22)} \]

Reexpanding \( V(t) \) of Eq. (3.17) using the recursive relation (3.18), we obtain the following expression for \( V(t) \), which is correct up to the third order in \( D_j(t) \):
\begin{align}
V_r(t) &= -\sum_j \sum_n \exp[-i\omega_{jn} t] D_j(t) \left[ \frac{\mu_d g_d}{\gamma_1 (1+i\delta_{aj})} \right] \\
&+ \sum_j \sum_n \sum_t \sum_{i, \gamma} \exp[-i(\omega_{jn}-\omega_{ij}+\omega_{ni}^*) t] D_j(t) D_i^*(t) D_n^*(t) \left[ \frac{\mu_d g_d}{\gamma_r [1+i(\delta_{nj}+\delta_{in})]} \right] S_n F_{in} I_{in} + \Gamma V_r(t). \tag{3.23}
\end{align}

Here, the coupling coefficient \(g_j\) and the dipole matrix element \(\mu_d\) are as defined in Eqs. (2.6) and (2.7), respectively,
\begin{align}
\delta_{aj} &= (\omega_0 - \omega_{aj}) / \gamma_1, \tag{3.24} \\
\delta_{ij} &= (\omega_0 - \omega_{ij}) / \gamma_1, \tag{3.25} \\
I_{in} &= \frac{\gamma_r [1+i(\delta_{nj}+\delta_{in})]}{4 g_n^2 |\mu_d|^2}, \tag{3.26} \\
S &= 1 + \sum_{n, \omega_{nl}^* = \omega_j^*} \sum_{l} \frac{\alpha_l^* \alpha_m \exp[-i(\mathbf{k}_l - \mathbf{k}_m) \cdot \mathbf{r}]}{I_l}, \tag{3.27} \\
I_l &= \frac{\gamma_r [1+i(\delta_{nl}+\delta_{aj})]}{4 g_l^2 |\mu_d|^2}, \tag{3.28} \\
S_{in} &= 1 + \sum_{m, \omega_{ml}^* = \omega_j^*} \sum_{j} \frac{\alpha_j^* \alpha_m \exp[-i(\mathbf{k}_j - \mathbf{k}_m) \cdot \mathbf{r}]}{I_{in}}, \tag{3.29} \\
I_{inj} &= \frac{\gamma_r [1+i(\delta_{nl}+\delta_{aj})]}{4 g_{nl}^2 |\mu_d|^2} F_{inj} \left[ 1 + i(\delta_{nl}+\delta_{aj}) \right] \times \left[ 1 - i(\delta_{nl}+\delta_{aj}) \right] \frac{1+i\delta_{in}/2}{1+i\delta_{in}}. \tag{3.30} \\
F_{in} &= \frac{1+i\delta_{in}/2F}{1+i\delta_{in}/2}, \tag{3.31} \\
\Gamma V_r(t) &= \sum_{i=1}^{N_0} \frac{\Gamma V_r(t)}{N_0}, \tag{3.32}
\end{align}

and \(F \equiv \gamma_r / 2 \gamma_1^*.

In Eqs. (3.27) and (3.29), the summing indices \(n\) and \(m\), respectively, denote sums over equal frequency (and not equal wave vector) field modes whereas the remaining summing indices \((l\) and \(j\)\) in the above equations range from 1 to \(q\). As defined by Eq. (3.27) is the usual saturation factor and \(S_{in}\) of Eq. (3.29) can be regarded as a generalized saturation factor when more than one modes of different frequencies are present. \(I_{inj}\) of Eq. (3.30) represents the corresponding generalized normalized saturation intensity. The generalizations are apparent if we note that \(I_{nlj}^* |\omega_n - \omega_j| = I_{nl}^* |\omega_n - \omega_l| = I_{nl},\) and \(S_{nl} |\omega_n - \omega_l| = S.

Similarly, the effect of collisions is expressed by the generalized collision factor \(F_{in}\) of Eq. (3.31), which attains a unit value in the absence of collisions.

We show in the Appendix that the deterministic part of Eq. (3.23) reduces to the expression obtained by Boyd et al.\(^{3}\) for the simple case in which the total field consists of a pump mode in the form of a monochromatic plane wave and two weak sideband modes, also in the form of monochromatic plane waves.

One may wonder what the difference is between Eq. (3.23) and that obtained by the usual adiabatic elimination procedure. Besides the extra care that we have taken to evaluate \(V_r(t)\) at the correct, shifted frequencies \(\omega_j^*\) of the modes, the main difference from the usual adiabatic elimination procedure is the appearance of higher-order derivative terms in \(D_j(t)\). These terms are crucial for obtaining the correct group velocity of propagation for the deterministic as well as the fluctuating part of the quantum field. This point will be elaborated upon in detail in the following paper of this series.

**IV. NOISE CORRELATIONS**

In principle, the correlation of the noise appearing in the solution for \(V_r(t)\) [cf. Eq. (2.17)] can be found using Eqs. (2.10)–(2.14), (2.20), and (2.21). However, a general solution is algebraically quite complicated. Therefore, we specialize to the case in which there are only two dominant pump-field modes in the medium that are at the same frequency \(\Omega_p = \omega_p^*\), and treat them as classical fields. Using the technique developed in the preceding section, we can likewise express the correlation of the Langevin noise of Eq. (2.21) in terms of a sum of the slowly varying amplitudes \(\alpha_j(t)\). However, while evaluating the expressions for the correlation of the Langevin noise, we retain the contribution of only the two strong pump modes. Even for this case, the correlation expressions are so complicated that we have to resort to a symbolic manipulation program. The extra complication arises because of our inclusion of the atomic collisions.\(^{3}\) Our results agree with those obtained by Reid and Walls.\(^{7}\)

Let us denote the wave vectors of the two pump modes by \(\mathbf{k}_{r1}\) and \(\mathbf{k}_{r2}\) and their slowly varying amplitudes by \(\alpha_{r1}(t)\) and \(\alpha_{r2}(t)\), respectively. Let us further define \(\Gamma V_r(\omega)\) and \(\Gamma V_r^+(\omega)\) as follows:
\begin{align}
\Gamma V_r(t) &= \int_{-\infty}^{\infty} \langle \bar{V}_r(t) \rangle \exp[-i(\omega + \Omega_p) t] d\omega, \tag{4.1} \\
\Gamma V_r^+(t) &= \int_{-\infty}^{\infty} \langle \bar{V}_r^+(t) \rangle \exp[-i(\omega - \Omega_p) t] d\omega, \tag{4.2}
\end{align}

so that \(\Gamma V_r(\omega) = \Gamma V_r(\omega + \Omega_p)\) and \(\Gamma V_r^+(\omega) = \Gamma V_r^+(\omega - \Omega_p)\).

From Eqs. (2.21) and (2.10)–(2.14), for this special case, we obtain the following correlations with use of a symbolic manipulation program:
\begin{align}
\langle \bar{V}_{r1}(\omega) \bar{V}_{r1}(\omega') \rangle &= \frac{D V_{r1} V_{r1}^+(d)}{2\pi} \delta(\omega + \omega') \delta_{n_1}, \tag{4.3} \\
\langle \bar{V}_{r2}(\omega) \bar{V}_{r2}(\omega') \rangle &= \frac{D V_{r2} V_{r2}^+(d)}{2\pi} \delta(\omega + \omega') \delta_{n_2}, \tag{4.4} \\
\langle \bar{V}_{r1}(\omega) \bar{V}_{r2}(\omega') \rangle &= \frac{D V_{r1} V_{r2}^+(d)}{2\pi} \delta(\omega + \omega') \delta_{n_1}, \tag{4.5}
\end{align}

where \(d = \omega / \gamma_1\).
\[ D_{V_1^+V_1^-}(d) = \frac{\beta^2 F}{2\gamma_1 D(d)} \left[(1-F)[d^4+2\Delta_p d^3+d^2(4F^2+1-\beta^2 F+\Delta_p^2)-d\Delta_p\beta^2 F-8F^2-2\beta^2 F^2] + 4F^2+\Delta_p^2(4F^2+2\beta^2 F^2) + d^2\beta^2 F^2+4\beta^2 F^2 + \beta^4 F^2 / 2 \right], \]  
(4.6)

\[ D_{V_1^+V_1^+}(d) = -\frac{\beta^2 F}{2\gamma_1 D(d)} \frac{A_p^2}{A_p^+ A_p^-} \left[(1-F)(-d^2\beta^2 F+2\Delta_p^2\beta^2 F^2)+F d^4+d^2[4F^3+F-3(\Delta_p^2+\beta^2 F)F] + 4F^3+\beta^2 F^2 / 2-12\Delta_p^2 F^3+i \Delta_p \left[(1-F)(-2\beta^2 F^2)+d^4 F+d^2[4F^3+3F-(\Delta_p^2+\beta^2 F)F] + 12F^3-4\Delta_p^2 F^3 \right) \right], \]  
(4.7)

\[ D_{V_1^+V_1^-}(d) = D_{V_1^+V_1^-}(d), \]  
(4.8)

\[ D(d) = (F+\Delta_p^2 F+\beta^2 F^2 / 2) + d^4 + d^4[2+4\beta^2 F-2\Delta_p^2] + d^2[1+8F^2+2\beta^2 F(2F-1)+\beta^4 F^2+\Delta_p^4-\Delta_p^2(8F^2-2\beta^2 F-2)] + 4F^2+4\beta^2 F^2 + \beta^4 F^2 + \Delta_p^2(8F^2+4\beta^2 F^2) + 4\Delta_p^4 F^2 \].  
(4.9)

\[ \Delta_p \equiv (\Omega_p-\omega_2) / \gamma_1 \]  

is the normalized pump-frequency detuning from the atomic resonance, \( \beta^2 \equiv 4 A_p^+ A_p^- / \gamma_1^2 F = 8 A_p^+ A_p^- / \gamma_1 \gamma_2 \), and

\[ A_p \equiv g_{\rho_1} |\mu_d| \langle \alpha_{p_1}(t) \exp(-i\mathbf{k}_{\rho_1} \cdot \mathbf{r}_1) + g_{\rho_2} |\mu_d| \langle \alpha_{p_2}(t) \exp(-i\mathbf{k}_{\rho_2} \cdot \mathbf{r}_1) \rangle \].

In the special situation where the two pump beams are collinear and are of equal amplitudes, i.e., \( \mathbf{k}_{p_1} = \mathbf{k}_{p_2} \) and \( \alpha_{p_1} = \alpha_{p_2} \), \( \beta^2 \) can be simply related to the pump intensity \( I_p \) and the line-center saturation intensity \( I_{sa} \) via

\[ \beta^2 = 2I_p / I_{sa}(\Omega_p / \omega_2)^2, \] where

\[ I_p = 8\epsilon_p e_0(\Omega_p / \omega_2)^2 |\alpha_{p_1}|^2 \] and

\[ I_{sa} = \epsilon_0 e_0(\gamma_1 / 2 / \theta / |\mu_d|)^2. \] Here, \( \beta \) is known as the Rabi frequency. Note that in arriving at the above relation, we have used the following relation between \( \alpha_{p_1} \) and the net electric-field amplitude of the pump beams

\[ \epsilon_p \] [for the exponential mode \( \exp(-i\mathbf{k}_{p_1} \cdot \mathbf{r}) \)],

\[ \epsilon_p = \hbar g_{\rho_1} 2\alpha_{p_1} \Omega_p / \omega_2. \] We also note that in the presence of collisions (i.e., \( F < 1 \)), \( D_{V_1^+V_1^-}(d) \) is not an even function of \( d \), whereas both \( D_{V_1^+V_1^-}(d) \) and \( D_{V_1^+V_1^-}(d) \) remain even. Furthermore, we show in the Appendix that our expression for \( D_{V_1^+V_1^-}(d) \) [Eq. (4.6)] gives the correct resonance-fluorescence spectrum as first obtained by Mollow in the collisionless limit.

The noise correlations obtained in this section, Eqs. (4.3)-(4.9), are crucial for the application to squeezed-state generation using nondegenerate four-wave mixing in atomic vapors, wherein the effect of atomic collisions cannot be neglected. In the next two sections, we illustrate how the results obtained so far lead to the coupled-mode equations for the slowly varying amplitudes \( \{\alpha_m(t)\} \) when a sum over the large number of atoms comprising the medium is performed.

**V. TEMPORAL COUPLED-MODE EQUATIONS**

Using Eqs. (2.1) and (3.1), the following equation for the slowly varying amplitude \( \alpha_m(t) \) is easily obtained:

\[ \frac{\partial}{\partial t} \alpha_m(t) = -i\Delta \omega_m \alpha_m(t) + \int_{V_m} d\mathbf{r} \rho_\mathbf{a} C_m^*(\mathbf{r}) V_\mathbf{a}(t) \exp(i\omega_m t) \].  
(5.1)

Here, we have assumed that the atoms are uniformly distributed so that Rayleigh scattering could be ignored. Also, using Eq. (3.21) we have replaced \( \Sigma_m \) appearing in Eq. (2.1) by the integral \( \int_{V_m} d\mathbf{r} \rho_\mathbf{a} \). With \( V_\mathbf{a}(t) \) of Eq. (3.23), the above equation gives a set of \( q \) coupled-mode equations for \( \{\alpha_m(t)\} \).

As in Sec. IV, in the following we restrict ourselves to the case where there are only two frequency-degenerate undepleted pump modes with wave vectors \( \mathbf{k}_{p_1} \) and \( \mathbf{k}_{p_2} \), respectively. We further assume that these pump modes have the same approximately constant amplitude \( \alpha_{p_1}(t) \approx \alpha_{p_2}(t) \approx \alpha_{p_0} \), which is much larger than the amplitudes \( \{\alpha_m(t) : m \neq p_1, p_2\} \) of the remaining \( (q-2) \) modes. With these assumptions, Eqs. (5.1) and (3.23) yield the following temporal coupled-mode equation for \( \alpha_m(t) \) up to the first order in \( \alpha_m / \alpha_{p_0} \):
\[
\frac{\partial}{\partial t} \alpha_m(t) = -i \Delta \omega_m \alpha_m(t) \\
+ \int_{V_m} d\mathbf{r}_p \rho_p \\
\times \left[ -\sum_{n \neq p_1, p_2} \exp[i(\omega_m - \omega_n^*) \Delta N \left\{ \frac{\mu_d}{\gamma} \left| g_m g_p \exp[i(\mathbf{k}_n - \mathbf{k}_m) \cdot \mathbf{r}] \right|}{\gamma_1 (1 + i \delta_{an}) S} \right] \\
- \exp[i(\omega_m - \omega_p^*) \Delta N \left\{ \frac{\mu_d}{\gamma} \left| g_m g_p \left[ \exp[i(\mathbf{k}_{p_1} - \mathbf{k}_m) \cdot \mathbf{r}] + \exp[i(\mathbf{k}_{p_2} - \mathbf{k}_m) \cdot \mathbf{r}] \right] \right|}{\gamma_1 (1 + i \delta_{ap}) S} \right] \\
+ \sum_{n \neq p_1, p_2} \exp[i(\omega_m - \omega_n^*) \Delta N \left\{ \frac{\mu_d}{\gamma} \left| g_m g_p \exp[-i(\mathbf{k}_m - \mathbf{k}_n) \cdot \mathbf{r}] \right|}{\gamma_1 (1 + i \delta_{an}) S} \right] \\
\times \exp[i(\omega_m - \omega_n^*) \Delta N \left\{ \frac{\mu_d}{\gamma} \left| g_m g_p \exp[-i(\mathbf{k}_m - \mathbf{k}_n) \cdot \mathbf{r}] \right|}{\gamma_1 (1 + i \delta_{an}) S} \right] \\
\times \exp[i(\omega_m - \omega_n^*) \Delta N \left\{ \frac{\mu_d}{\gamma} \left| g_m g_p \exp[-i(\mathbf{k}_m - \mathbf{k}_n) \cdot \mathbf{r}] \right|}{\gamma_1 (1 + i \delta_{an}) S} \right] \\
+ g_m \mu_d^* \Gamma_{\mathbf{r}_p}(t) \exp[-i\mathbf{k}_m \cdot \mathbf{r}] \exp(i\omega_m t) \right], \tag{5.2}
\]

where \(D_n\) and \(D_n^*\) are given by Eqs. (3.16) and (3.19), respectively, and \(D_p = D_{p_1} = D_{p_2} = \alpha_p\) due to the constant pump-amplitude assumption. The fourth and fifth terms of the above equation are obtained from the second term of Eq. (3.23) by setting \(l, j\) equal to \(p_1, p_2\) (or \(p_2, p_1\)), and \(n, j\) equal to \(p_1, p_2\) (or \(p_2, p_1\)), respectively. \(S, S_{np}, F_{np},\) and \(I_{np}\) obtained from Eqs. (3.26)--(3.31), are approximately given by

\[
S = 1 + \frac{\alpha_{p_1}^* \alpha_{p_1} \left\{ \exp[i(\mathbf{k}_{p_1} - \mathbf{k}_{p_2}) \cdot \mathbf{r}] + \exp[-i(\mathbf{k}_{p_1} - \mathbf{k}_{p_2}) \cdot \mathbf{r}] + 2 \right\]}{I_p}, \tag{5.3}
\]

\[
S_{np} = 1 + \frac{\alpha_{p_1}^* \alpha_{p_1} \left\{ \exp[i(\mathbf{k}_{p_1} - \mathbf{k}_{p_2}) \cdot \mathbf{r}] + \exp[-i(\mathbf{k}_{p_1} - \mathbf{k}_{p_2}) \cdot \mathbf{r}] + 2 \right\]}{I_{np}}, \tag{5.4}
\]

\[
I_p = \frac{\gamma P_0^2}{\gamma_1^2 (1 + 2 \delta_{an}) / 4 g_p^2 |\mu_d|^2}, \tag{5.5}
\]

\[
I_{np} = \frac{\gamma P_0^2}{4 g_p^2 |\mu_d|^2} \left[ F_{np} \left( 1 + i \delta_{an} \right) \left( 1 - i \delta_{an} / 2 \right) \frac{1 + i \delta_{np}/2}{1 + i \delta_{np}} \right], \tag{5.6}
\]

\[
F_{np} = \left( 1 + i \delta_{np} / 2 \right) / \left( 1 + i \delta_{np} / 2 \right) = F_{np}^* \tag{5.7}
\]

\[
I_{np} = \frac{\gamma P_0^2}{4 g_p^2 |\mu_d|^2} \left( 1 - i \delta_{an} \right) \left( 1 + i \delta_{an} \right) \frac{1 + i \delta_{np}/2}{1 + i \delta_{np}} \right], \tag{5.8}
\]

Equation (5.2) is greatly simplified if we take the medium volume \(V_m\) to be equal to the quantization volume \(V_Q\). In this case, the spatial integral in Eq. (5.2), which extends over an infinite volume due to the periodic boundary conditions, can be approximately evaluated by retaining only the phase-matching terms to give the following coupled-mode equation:
\[
\frac{\partial}{\partial t}\alpha_m(t) = -i\Delta \omega_m \alpha_m(t) + \int_{r_Q} \left[ D_m \left( \sum_{j} \rho_d |\mu_d|^2 g_{mj}^2 \gamma_j (1 + i\delta_{\omega_m} S) \right) + D_m \left( \sum_{j} \rho_d |\mu_d|^2 g_{mj}^2 \gamma_j (1 + i\delta_{\omega_m} S) \Sigma_{pm} F_{pm} \right) \right. \\
\left. + \exp[i(\omega_m - \omega_j) t] \right] \left[ \frac{\rho_d |\mu_d|^2 g_{mj}^2 \gamma_j (1 + i\delta_{\omega_m} S) \Sigma_{pm} F_{pm} \right] \left. + \exp[i(\omega_m + \omega_j - 2\omega_p) t] \right] \left[ \frac{\rho_d |\mu_d|^2 g_{mj}^2 \gamma_j (1 + i\delta_{\omega_m} S) \Sigma_{pm} F_{pm} \right] \right)
\]
\[+ \Gamma_m(t), \tag{5.9} \]

where
\[
\Gamma_m(t) = g_m \mu_d^2 \int_{r_Q} d\tau \rho_d \Gamma_r(t) \times \exp(-i\kappa_m \cdot \tau) \exp(i\omega_m t). \tag{5.10} \]

Note that \( S \) and \( \Sigma_{pm} \) in the denominators of the terms in Eq. (5.9) are \( r \) dependent. This is why our retaining of the phase-matching terms in Eq. (5.9) is only approximate, except in the special case when \( \kappa_{p_1} \equiv \kappa_{p_2} \). The spatial integrals can be evaluated analytically as is done by Reid and Walls in the case of degenerate four-wave mixing.

In the fifth term of Eq. (5.9),
\[
\delta \omega_m = \omega_m^+ + \omega_m^* - 2\omega_p, \tag{5.11} \]
where \( \omega^\pm \) is the shifted frequency of mode \( n \) [cf. Eqs. (3.2) and (3.5)] picked by the phase-matching condition
\[
\kappa_m + \kappa_n = \kappa_{p_1} + \kappa_{p_2}. \tag{5.12} \]

The sixth term of Eq. (5.9) represents three-wave mixing with \( \kappa_m + \kappa_n = 2\kappa_{p_1} \) or \( \kappa_m + \kappa_n = 2\kappa_{p_2} \) as the phase-matching condition. Similarly, the fifth term gives rise to four-wave mixing with Eq. (5.12) as the phase-matching condition. Both the fifth and sixth terms lead to coupling with the conjugate modes \( [\alpha^\dagger] \) appearing in the differential operators \( [D^\dagger] \) [cf. Eq. (3.19)].

The fourth term of Eq. (5.9) is also a four-wave mixing term with \( \kappa_n - \kappa_j = \pm (\kappa_{p_1} - \kappa_{p_2}) \) as the phase-matching condition. This term, however, does not lead to coupling with the conjugate modes. When \( \kappa_{p_1} = \kappa_{p_2} \), it couples a mode with itself giving rise to extra nonlinear gain (loss) and frequency shift.

In the following, we will use \( \alpha_m(t) \), instead of \( \alpha_n(t) \), to denote the image mode to \( \alpha_m(t) \), i.e., the mode whose conjugate \( \alpha_m^* \) couples to \( \alpha_m \) via the nonlinear terms of Eq. (5.9). So whenever the subscript \( m \) appears, it is to be understood as \( n \) with the wave vector of the corresponding mode determined by Eq. (5.12).

The result obtained in this section, Eq. (5.9), can involve many weak probe beams and is somewhat general. In the following section, we specialize to the case of spatially degenerate forward four-wave mixing, which would be important for application to squeezed-state generation experiments using such a configuration. The results of one such application have been published.

\section*{VI. SINGLE-BEAM INFINITE MEDIUM CASE}

In this section, we derive the temporal coupled-mode equations for a single pump beam of wave vector \( \kappa_p \) and amplitude \( \alpha_{p1}(t) \sim 2a_{p1} \) propagating through an infinite medium. The equation of motion for a copropagating nondegenerate mode with frequency \( \omega^\pm \), which is different from the pump-mode frequency \( \omega_p \), can be obtained from Eq. (5.9). In this case, the third term of Eq. (5.9) is identical to the fourth term and the fifth term is identical to the sixth term. Adopting the image mode notation defined at the end of Sec. V, we obtain the following coupled-mode equations:

\[
\frac{\partial \alpha_m}{\partial t} = -i\Delta \omega_m \alpha_m + \gamma_m \alpha_m + i \frac{\partial \alpha_m}{\partial \omega_m^\dagger} \frac{\partial \gamma_m^*}{\partial \omega_m^\dagger} + X_m \exp(i\omega_m t) \alpha_m^\dagger + i \exp(i\omega_m t) \frac{\partial \alpha_m^\dagger}{\partial t} \frac{\partial X_m}{\partial \omega_m^\dagger} + \Gamma_m, \tag{6.1} \]

\[
\frac{\partial \alpha_m^\dagger}{\partial t} = i\Delta \omega_m^\dagger \alpha_m^\dagger + \gamma_m^\dagger \alpha_m^\dagger + i \frac{\partial \alpha_m^\dagger}{\partial \omega_m^\dagger} \frac{\partial \gamma_m^*}{\partial \omega_m^\dagger} + X_m^* \exp(-i\omega_m^* t) \alpha_m + i \exp(-i\omega_m^* t) \frac{\partial \alpha_m}{\partial t} \frac{\partial X_m^*}{\partial \omega_m^*} + \Gamma^*_m, \tag{6.2} \]

where
\[
\gamma_m = - \frac{\rho_d V g_m^2 |\mu_d|^2}{\gamma_j (1 + i\delta_{\omega_m}) S} \left( 1 - \frac{4g_m^2 |\mu_d|^2 \alpha_{p1}^2}{\Sigma_{pm} F_{pm} \Sigma_{pm} S} \right), \tag{6.3} \]
\[ X_m = \frac{4p_a V Q g_m g_m' g_p |\alpha_p|^2}{\gamma'_(1 + i(\Delta \omega_p + \Delta \omega_p')) SS_{mp} F_{mp} I_{mp} g_p g_m} , \]  
\[ k_m + k_m' = 2k_p , \]  
\[ \delta \omega'_m = \omega'_m + \omega'_m - 2\omega'_p , \]  
\[ S = 1 + \frac{4\alpha_p^* \alpha_p}{I_p} , \]  
\[ S_{mp} = 1 + \frac{4\alpha_p^* \alpha_p}{I_{mp}} , \]  
\[ = S^*_{pm} , \]  
and the remaining symbols are as defined in Eqs. (5.4)–(5.8).

To determine \( \omega_p \), we need the equation of motion for the pump-mode amplitude \( \alpha_p(t) \). Noting that, for \( m = p = p_1 = p_2 \), only the second term of the spatial integral in Eq. (5.2) contributes, in which case we obtain the following equation for \( \alpha_p(t) \):

\[ \frac{\partial \alpha_p(t)}{\partial t} = (-i \Delta \omega_p + \gamma_p) \alpha_p(t) , \]  
where

\[ \gamma_p = -\frac{\rho_o V Q g_p^2 |\mu_d|^2}{\gamma'_(1 + i \delta \omega_p) S} . \]  

Defining \( \gamma_p' = \gamma_p + i \gamma_p \), we have

\[ \gamma_p' = \frac{-\rho_o V Q g_p^2 |\mu_d|^2}{\gamma'_(1 + \delta \omega_p) S} , \]  

and

\[ \gamma_p' = \frac{-\rho_o V Q g_p^2 |\mu_d|^2 \Delta \omega_m}{\gamma'_(1 + \delta \omega_p) S} . \]  

Clearly, the loss coefficient \( \gamma'_p \) must be small if our assumption of a constant \( \alpha_p(t) \approx 2\alpha_p \) is to be valid. In this case, the solution for \( \alpha_p(t) \) is given by

\[ \alpha_p(t) = \exp[ -i(\Delta \omega_p - \gamma_p t)] \alpha_p(0) . \]  

Hence, in order for \( \alpha_p(t) \) to be truly slowly varying so that \( \alpha_p(t) \approx 2\alpha_p \), we must set \( \Delta \omega_p = \gamma_p t \). Then, from defining Eqs. (3.2)–(3.5), we determine \( \omega'_p = \gamma_p' \) and \( k_p/c \). It is clear that the refractive index seen by the pump beam is given by \( n_p = 1 + \gamma_p'/k_p/c \).

Following a similar argument for the other modes, we also have \( \Delta \omega_m = \text{Im}(\gamma_m) \) with \( \gamma_m \) given by Eq. (6.3). We note from Eqs. (6.3) and (6.12) that even when \( \omega'_m = \omega'_p \), \( \Delta \omega_m \) and \( \Delta \omega_p \) are not equal. That is, the pump-mode and the copropagating nondegenerate modes always see different refractive indices so that the frequency mismatch \( \Delta \omega_m \) in Eq. (6.6) is, in general, not zero. Physically, this is due to an additional index seen by the probe beams because of coherent scattering of the pump beam off the population grating formed by the beating between the pump and the probe beams.

Note that if we had derived Eq. (6.1) using the more exact expression for Eq. (2.1), which involves \((i/\omega_p)\partial V / \partial t\), instead of \( V / \partial t \) as mentioned after Eq. (2.14), then we would have obtained \((\partial \alpha_m / \partial t)(\partial V'_m / \partial \omega_m) + (\partial \gamma_m / \partial \omega_m)\partial \gamma_m / \partial \omega_m \) in Eq. (6.1). Also, the coefficient \( \gamma_m \) in Eq. (6.3) would have been multiplied by the factor \( \omega_m / \omega_p \). Similar modifications would also apply to the coefficient \( X_m \) and the \( \partial \alpha_m / \partial t \) term in Eq. (6.1). The physical significance of the \( \partial \alpha_m / \partial t \) on the right-hand side of Eq. (6.1) will become clear in the following paper of this series, in which we rigorously treat the simple case of a lossless dielectric medium without making the rotating-wave (near-resonance) approximation, and also approximately deal with the more complex case of a slightly lossy medium. There, we will show that the rotating-wave approximation is valid only in the limit where \( |\gamma_m| \ll \omega_m \) and that the \( \partial \alpha_m / \partial t \) on the right-hand side of Eq. (6.1) is necessary for obtaining the correct group velocity.

The temporal coupled-mode equations [i.e., Eqs. (5.9), (6.1), and (6.2)] would be directly applicable to cavity experiments where the quantum state in the cavity is known at time \( t = 0 \), and one is interested in the correlations \( \langle \alpha_m(t)\alpha_m(t) \rangle \) and \( \langle \alpha_m(t)\alpha_m(t) \rangle \) at a later time \( t \) inside the cavity. In a traveling-wave experiment, however, one has a spatial boundary-value problem and one must deal with the issue of spatial propagation. In the following paper of this series, we will present a consistent formalism to treat with rigor, the spatial propagation of a quantum field. This formalism will allow us to convert the temporal coupled-mode equations, Eqs. (6.1) and (6.2), into spatial coupled-mode equations.

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**APPENDIX**

In this appendix, we show that the expressions derived in this paper, although quite general, do in fact reduce to those derived in earlier works, in particular the works of Boyd et al., and Mollow.

In the paper of Boyd et al., \( E_1 \) corresponds to our pump field \( A_p \). \( E_1 \) corresponds to our pump field \( \alpha_m \), \( \rho_{ab} \) equals our \( V/1/T \), \( \rho_{ab} \) equals our \( N_1 \), \( \rho_{ab} \) equals our \( n_1 \), \( 1/T \) equals our \( \gamma'_m \), \( 1/T \) equals our \( \gamma'_m \), \( \mu_{ab} \) equals our \( \mu_{ab} \), \( \omega_m \) equals our \( \Omega_m \), \( \Omega_1 \) equals our \( \Omega_m \), \( \omega_{ab} \) equals our \( \omega_{ab} \). With these identifications, it can be shown that their expression for \( \rho_{ab} \langle \omega \rangle \) [Eq. (5a) in their paper] is identical to the \( (j = p) \) term of the first term of Eq. (3.23), i.e.,

\[ \rho_{ab}(\Omega_p) = -\frac{A_p \mu_d g_p}{\gamma'_(1 + \delta \omega_p) S} , \]  

where we have let \( D_j(t\omega) = \alpha_p = A_p \). The expression for \( \rho_{ab}(\omega) \) in their paper [Eq. (5b)] can be obtained by addi-
ing the \((j = m)\) term of the first term and the
\((j = l = p, n = m)\) term of the second term of Eq. (3.23),
\(i.e.,\)
\[
\rho_{ab}(\Omega_m) = \frac{A_p^+ A_p \alpha_m \mu_d g_p}{\gamma_\parallel (1 + i \delta_{ap}) S}
\]
\[+ \frac{A_p^+ A_p \alpha_m \mu_d g_p}{\gamma_\parallel (1 + i \delta_{ap}) SS_{pm} F_{pm} I_{pm}}.\]  
\(\text{ (A2)}\)

Finally, the expression for \(\rho_{ba}(2\omega_1 - \omega_2)\) in their paper
[Eq. (5c)] can be shown to be identical to the
\((j = n = p, l = m)\) term of the second term of Eq. (3.23),
\(i.e.,\)
\[
\rho_{ba}(2\Omega_p - \Omega_m) = \frac{A_p^+ A_p \alpha_m \mu_d g_p}{\gamma_\parallel [1 + i (\delta_{ap} + \delta_{mp})] SS_{mp} F_{mp} I_{mp}}.\]  
\(\text{ (A3)}\)

To show that Eq. (3.23) also gives the correct resonance
fluorescence spectrum, we need to calculate the correlation

\[
\langle V_r^+(t) V_r(t + \tau) \rangle = |V_\infty|^2 \exp(-i\Omega_p \tau)
\]
\[+ \langle \Gamma_r^+(t) \Gamma_r(t + \tau) \rangle,\]  
\(\text{ (A4)}\)

where
\[
V_\infty = -\frac{A_p \mu_d g_p}{\gamma_\parallel (1 + i \delta_{ap}) S},\]  
\(\text{ (A5)}\)

and evaluate the Fourier transform
\[
\tilde{g}(\omega) = \int_{-\infty}^{\infty} \langle V_r^+(t) V_r(t + \tau) \rangle \exp(i\omega \tau) d\tau
\]
\[= 2\pi |V_\infty|^2 \delta(\Omega_p - \omega)
\]
\[+ \int_{-\infty}^{\infty} \langle \Gamma_r^+(t) \Gamma_r(t + \tau) \rangle \exp(i\omega \tau) d\tau.\]  
\(\text{ (A6)}\)

Simple manipulation shows that the expression for \(V_\infty\)
agrees with Eq. (3.17) and that for \(\tilde{g}(\omega)\) agrees with Eq.
(4.15) of Mollow's paper,\(^6\) provided we make the
identifications \(\gamma_\parallel = \kappa / 2, (\Omega_p - \omega_a) = \Delta\omega,\) and \(\gamma_\parallel^2 \beta^2 = \Omega^2.\)

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