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

Seng-Tiong Ho
AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974

Prem Kumar
Department of Electrical Engineering and Computer Science, Robert R. McCormick School of Engineering and Applied Science, Technological Institute, Northwestern University, Evanston, Illinois 60208

Jeffrey H. Shapiro
Department of Electrical Engineering and Computer Science and Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139
(Received 3 August 1989; revised manuscript received 25 October 1990)

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 $V_i(t)$ in terms of the multimode-field Fourier amplitudes $\{A_k(\mathbf{r}_i)\}$. 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 $V_i(t)$ in terms of slowly varying modal amplitudes instead of the Fourier amplitudes $\{A_k(\mathbf{r}_i)\}$. 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 $V(\mathbf{r},t)$ in terms of the q field-mode variables $\{a_j(t)\}$ 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 $V(\mathbf{r},t)$, 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

gives the correct resonance fluorescence spectrum as first obtained by Mollow.[6]

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 $\{\alpha_j(t)\}$. 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 of interest 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):

$$\frac{\partial a_j}{\partial t}=-i\omega_j a_j+\sum_i C_j^*(\mathbf{r}_i)V_i \ , \tag{2.1}$$

$$\frac{\partial V_i}{\partial t}=-i\omega_a V_i-a(\mathbf{r}_i,t)(n_{gi}-n_i)-\gamma_\perp V_i+f_{V_i} \ , \tag{2.2}$$

$$\frac{\partial V_i^+}{\partial t}=i\omega_a V_i^+-a^+(\mathbf{r}_i,t)(n_{gi}-n_i)-\gamma_\perp V_i^++f_{V_i^+} \ , \tag{2.3}$$

$$\frac{\partial n_i}{\partial t}=-[a^+(\mathbf{r}_i,t)V_i+a(\mathbf{r}_i,t)V_i^+]-\gamma_\parallel n_i+f_{n_i} \ , \tag{2.4}$$

and $n_{gi}=1-n_i$. Here, $\{a_j: 1\le j\le q\}$ are the c-number variables associated with the annihilation operators $\{\hat{a}_j\}$ of the q x-polarized field modes whose explicit dynamics are of interest; V_i, V_i^+, n_i, and n_{gi} are the c-number variables associated with the atomic down-transition, up-transition, upper-level occupation, and ground-level occupation operators, respectively, for the ith atom; $\gamma_\perp$ and $\gamma_\parallel=2F\gamma_\perp$, $0\le F\le 1$, 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 $C_j(\mathbf{r}_i)$ are given by

$$C_j(\mathbf{r}_i)=g_j\mu_d\exp(i\mathbf{k}_j\cdot\mathbf{r}_i) \ , \tag{2.5}$$

$$g_j=(\omega_a^2/2\hbar\epsilon_0\omega_j V_Q)^{1/2} \ , \tag{2.6}$$

with μ_d being the dipole matrix element defined via

$$\mu_d\equiv e_i\langle x|\hat{\mathbf{r}}_i|g\rangle_i\cdot\mathbf{e}_x \ , \tag{2.7}$$

where $|g\rangle_i$ is the ground state and $|x\rangle_i$ is the upper state of the ith atom that is coupled to the x polarization of the field; $\hat{\mathbf{r}}_i$ is the position-vector operator of the electron in the ith atom with e the electron charge ($e<0$); ω_a is the resonance angular frequency of the stationary two-level atoms and $\omega_j\equiv c|\mathbf{k}_j|$, where $\mathbf{k}_j$ $(1\le j\le q)$ is the free-space wave vector of the jth field mode defined via periodic boundary conditions over the quantization volume V_Q; the multimode field variables $a(\mathbf{r}_i,t)$ and $a^+(\mathbf{r}_i,t)$ are defined via

$$a(\mathbf{r}_i,t)\equiv\sum_j C_j(\mathbf{r}_i)a_j(t) \ , \tag{2.8}$$

$$a^+(\mathbf{r}_i,t)\equiv\sum_j C_j^*(\mathbf{r}_i)a_j^+(t) \ , \tag{2.9}$$

and the Langevin noise forces f_{V_i}, $f_{V_i^+}$, and f_{n_i} have the following nonzero correlations:

$$\langle f_{V_i^+}(t)f_{V_i}(t')\rangle=2\gamma_\perp(1-F)n_i(t)\delta(t-t') \ , \tag{2.10}$$

$$\langle f_{V_i}(t)f_{V_i}(t')\rangle=a(\mathbf{r}_i,t)V_i(t)\delta(t-t') \ , \tag{2.11}$$

$$\langle f_{V_i^+}(t)f_{V_i^+}(t')\rangle=a^+(\mathbf{r}_i,t)V_i^+(t)\delta(t-t') \ , \tag{2.12}$$

$$\langle f_{n_i}(t)f_{n_i}(t')\rangle=\{-[a^+(\mathbf{r}_i,t)V_i(t)+a(\mathbf{r}_i,t)V_i^+(t)]+\gamma_\parallel n_i(t)\}\delta(t-t') \ , \tag{2.13}$$

$$\langle f_{n_{gi}}(t)f_{n_{gi}}(t')\rangle=-\langle f_{n_{gi}}(t)f_{n_i}(t')\rangle=\langle f_{n_i}(t)f_{n_i}(t')\rangle \ . \tag{2.14}$$

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

$$a(\mathbf{r}_i,t)\equiv\sum_m A_m(\mathbf{r}_i)\exp(-i\nu_m t) \ , \tag{2.15}$$

$$a^+(\mathbf{r}_i,t)\equiv\sum_m A_m^+(\mathbf{r}_i)\exp(i\nu_m t) \ , \tag{2.16}$$

where $\nu_m\equiv 2\pi m/T$ 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:

$$V_i(t)=-\frac{(2\gamma_\perp F)A_m(\mathbf{r}_i)\exp(-i\nu_m t)}{D^-(\nu_m)\Pi_m(\nu_m,t)}+\Gamma_{V_i}(t) \ , \tag{2.17}$$

where $\Pi_m(\omega,t)$ is defined recursively as

$$\Pi_m(\omega,t)\equiv 2\gamma_\perp F-i(\omega-\nu_m)+\frac{2A_l^+(\mathbf{r}_i)A_n(\mathbf{r}_i)\exp[i(\nu_l-\nu_n)t]\Pi_m(\omega,t)D^-(\omega)}{D^-(-\nu_l+\nu_m+\omega)\Pi_m(-\nu_l+\nu_n+\omega,t)}\times\left[\frac{1}{D^-(\nu_n+\omega-\nu_m)}+\frac{1}{D^+(-\nu_l+\omega-\nu_m)}\right] \tag{2.18}$$

with

$$D^{\pm}(\omega)\equiv -i(\omega\pm\omega_a)+\gamma_\perp \ . \tag{2.19}$$

In Eq. (2.17), $\Gamma_{V_i}(t)$ is a Langevin noise term, which can be related to its Fourier transform $\Gamma_{V_i}(\omega)$ by

$$\Gamma_{V_i}(t)=\int_{-\infty}^{\infty} d\omega\, \Gamma_{V_i}(\omega)\exp(-i\omega t) \ , \tag{2.20}$$

where $\Gamma_{V_i}(\omega)$ was obtained in Sec. VI of I (Ref. 1) up to the third order in the field amplitudes $\{A_m(\mathbf{r}_i)\}$, and is given by

$$\begin{aligned}\Gamma_{V_i}(\omega)=&-\frac{2A_m(\mathbf{r}_i)A_l^{+}(\mathbf{r}_i)f_{V_{l-m}}}{D_0^{-}\pi_{-m}D_{l-m}^{-}}\\&-\frac{2A_m(\mathbf{r}_i)}{D_0^{-}}\frac{A_l(\mathbf{r}_i)f_{V^{+}_{-l-m}}}{\pi_{-m}D^{+}_{-l-m}}\\&+\frac{2A_m(\mathbf{r}_i)}{D_0^{-}}\frac{f_{n_{-m}}}{\pi_{-m}}+\frac{f_{V_0}}{D_0^{-}} \ .\end{aligned} \tag{2.21}$$

Here, the subscript i on the Langevin forces has been dropped and the following definitions have been employed:

$$D^{\pm}_{\pm l}\equiv D^{\pm}(\omega\pm\nu_l) \ , \tag{2.22}$$

$$D_0^{\pm}\equiv D^{\pm}(\omega) \ , \tag{2.23}$$

$$\pi(\omega)\equiv\left[-i\omega+2F\gamma_\perp+2A_k^{+}A_k\left[\frac{1}{D_k^{-}}+\frac{1}{D^{+}_{-k}}\right]\right] \ , \tag{2.24}$$

$$\pi_0\equiv\pi(\omega) \ , \tag{2.25}$$

$$\pi_{\pm k}\equiv\pi(\omega\pm\nu_k) \ , \tag{2.26}$$

$$X_0\equiv X(\omega) \ , \tag{2.27}$$

$$X_{\pm l\pm m}\equiv X(\omega\pm\nu_l\pm\nu_m) \ , \tag{2.28}$$

for $X\in\{f_{V_i},f_{V_i}^{\dagger},f_{n_i}\}$.

III. SLOWLY VARYING AMPLITUDE APPROXIMATION

The compact solution for the polarization $V_i(t)$ of the ith atom given by Eqs. (2.17) and (2.18) is of the following form:

$$V_i(t)=\sum_m Y(\nu_m,\mathbf{r}_i,t)A_m(\mathbf{r}_i)\exp(-i\nu_m t)+\Gamma_{V_i}(t) \ , \tag{3.1}$$

where $Y(\nu_m,\mathbf{r}_i,t)$ is a function of $\{A_m(\mathbf{r}_i)\}$ 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

$$a_j(t)=\alpha_j(t)\exp[-i(\omega_j-\Delta\omega_j)t] \ , \tag{3.2}$$

where $\alpha_j(t)$ is slowly varying with changes occurring over time scales of the order of $\gamma_\perp^{-1}$, ω_j is the free-field mode frequency, and $\Delta\omega_j$ is an anticipated frequency shift due to interaction with the medium. Our *a priori* inclusion of $\Delta\omega_j$ 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) can then be written as

$$a(\mathbf{r}_i,t)=\sum_j C_j(\mathbf{r}_i)\alpha_j(t)\exp(-i\omega_j^s t) \ , \tag{3.3}$$

$$a^{+}(\mathbf{r}_i,t)=\sum_j C_j^{*}(\mathbf{r}_i)\alpha_j^{+}(t)\exp(i\omega_j^s t) \ , \tag{3.4}$$

where

$$\omega_j^s\equiv\omega_j-\Delta\omega_j \ . \tag{3.5}$$

However, $a(\mathbf{r}_i,t)$ and $a^{+}(\mathbf{r}_i,t)$ can also be expressed in terms of the Fourier amplitudes $\{A_m(\mathbf{r}_i),A_m^{+}(\mathbf{r}_i)\}$ via Eqs. (2.15) and (2.16), respectively, which in the continuum limit become

$$a(\mathbf{r}_i,t)=\int_{-\infty}^{\infty} d\nu\, A(\nu,\mathbf{r}_i)\exp(-i\nu t) \ , \tag{3.6}$$

$$a^{+}(\mathbf{r}_i,t)=\int_{-\infty}^{\infty} d\nu\, A^{+}(\nu,\mathbf{r}_i)\exp(i\nu t) \ , \tag{3.7}$$

where $A(\nu,\mathbf{r}_i)_{\nu=\nu_m}=A_m(\mathbf{r}_i)/\delta\nu$ with $\delta\nu=\nu_{m+1}-\nu_m$. 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

$$V_i(t)=\int_{-\infty}^{\infty} dt'(1/2\pi)\tilde{Y}(t',\mathbf{r}_i,t)a(\mathbf{r}_i,t-t') \ , \tag{3.8}$$

where $\tilde{Y}(t',\mathbf{r}_i,t)$ is the inverse Fourier transform defined via

$$\tilde{Y}(t',\mathbf{r}_i,t)\equiv\int_{-\infty}^{\infty} d\nu\, Y(\nu,\mathbf{r}_i,t)\exp(-i\nu t') \ . \tag{3.9}$$

We see from Eqs. (2.17) and (2.19) that the magnitude of $Y(\nu,\mathbf{r}_i,t)$ has a peak at $\nu\simeq\omega_a$ of width $\Delta\nu\simeq\gamma_\perp$ due to the presence of $D^{-}(\nu)$ in the denominator. This means that $\tilde{Y}(t',\mathbf{r}_i,t)$ must be a decaying sinusoid in t' with a decay time $\simeq\gamma_\perp^{-1}$. It is thus sufficient to approximate $a(\mathbf{r}_i,t-t')$ in Eq. (3.8) with an expansion around $t'=0$ up to $t'\simeq\gamma_\perp^{-1}$. Moreover, in order for the expansion to be convergent, we assume that $\gamma_\perp^{-1}[\partial\alpha_j(t)/\partial t]\ll\alpha_j(t)$. This assumption implies that the pump beam cannot have a linewidth broader than $\gamma_\perp$. When $\alpha_j(t)$ is sufficiently slowly varying, a good approximation is obtained by expanding the slowly varying amplitude $\alpha_j(t-t')$ in Eq. (3.3) up to the first order as

$$\alpha_j(t)-t'\frac{\partial\alpha_j(t)}{\partial t} \ , \tag{3.10}$$

giving

$$\begin{aligned}a(\mathbf{r}_i,t-t')\simeq&\sum_j C_j(\mathbf{r}_i)\alpha_j(t)\exp[-i\omega_j^s(t-t')]\\&-t'\sum_j C_j(\mathbf{r}_i)\frac{\partial\alpha_j(t)}{\partial t}\exp[-i\omega_j^s(t-t')] \ ,\end{aligned} \tag{3.11}$$

which upon substituting into Eq. (3.8) yields

$$V_i(t)=\sum_j Y(\omega_j^s,\mathbf{r}_i,t)C_j(\mathbf{r}_i)\alpha_j(t)\exp(-i\omega_j^s t) + \sum_j i\frac{\partial Y(\omega_j^s,\mathbf{r}_i,t)}{\partial\omega_j^s}C_j(\mathbf{r}_i)\frac{\partial\alpha_j(t)}{\partial t}\exp(-i\omega_j^s t)\ , \tag{3.12}$$

where

$$\frac{\partial\alpha_j(t)}{\partial t}\equiv\left.\frac{\partial\alpha_j(t')}{\partial t'}\right|_{t'=t}\ , \tag{3.13}$$

and

$$\frac{\partial Y(\omega_j^s,\mathbf{r}_i,t)}{\partial\omega_j^s}\equiv\left.\frac{\partial Y(\omega,\mathbf{r}_i,t)}{\partial\omega}\right|_{\omega=\omega_j^s}\ . \tag{3.14}$$

Equation (3.12) can be abbreviated as

$$V_i(t)=\sum_j C_j(\mathbf{r}_i)\exp(-i\omega_j^s t)\mathcal{D}_j(t)Y(\omega_j^s,\mathbf{r}_i,t)\ , \tag{3.15}$$

where $\mathcal{D}_j(t)$ is a differential operator defined by

$$\mathcal{D}_j(t)\equiv\left[\alpha_j(t)+i\frac{\partial\alpha_j(t)}{\partial t}\frac{\partial}{\partial\omega_j^s}\right]\ . \tag{3.16}$$

We thus arrive at the following simple result: the slowly varying amplitude approximation allows us to replace $\sum_m A_m(\mathbf{r})\exp(-i\nu_m t)$ in Eq. (3.1) by $\sum_j C_j(\mathbf{r})\exp(-i\omega_j^s t)\mathcal{D}_j(t)$. As noted earlier $Y(\nu_m,\mathbf{r}_i,t)$ in Eq. (3.1) also depends upon sums like $\sum_m A_m(\mathbf{r})\exp(-i\nu_m t)$ via the recursive function Π_m of Eq. (2.18). These sums can be approximated in a similar way giving the following result for $V_i(t)$:

$$V_i(t)=-\sum_j C_j(\mathbf{r}_i)\exp(-i\omega_j^s t)\times\mathcal{D}_j(t)\left[\frac{2\gamma_\perp F}{D^-(\omega_j^s)\Pi_j(\omega_j^s,\mathbf{r}_i,t)}\right]+\Gamma_{V_i}(t)\ , \tag{3.17}$$

where the differential operator $\Pi_j(\omega,\mathbf{r}_i,t)$ is, once again, defined recursively as

$$\Pi_j(\omega,\mathbf{r}_i,t)=2\gamma_\perp F-i(\omega-\omega_j^s)+2\sum_{l,n}C_l^*(\mathbf{r}_i)\exp(i\omega_l^s t)\mathcal{D}_l^+(t)C_n(\mathbf{r}_i)\exp(-i\omega_n^s t)\mathcal{D}_n(t) \times\frac{\Pi_j(\omega,\mathbf{r}_i,t)D^-(\omega)}{D^-(-\omega_l^s+\omega_n^s+\omega)\Pi_j(-\omega_l^s+\omega_n^s+\omega,\mathbf{r}_i,t)} \times\left[\frac{1}{D^-(\omega_n^s+\omega-\omega_j^s)}+\frac{1}{D^+(-\omega_l^s+\omega-\omega_j^s)}\right]\ , \tag{3.18}$$

with

$$\mathcal{D}_l^+(t)\equiv\alpha_l^+(t)-i\frac{\partial\alpha_l^+(t)}{\partial t}\frac{\partial}{\partial\omega_l^s}\ . \tag{3.19}$$

Note that $\mathcal{D}_l^+(t)$ is like the "Hermitian conjugate" of $\mathcal{D}_l(t)$. 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 $\{a_j(t):1\le j\le q\}$.

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

$$V(\mathbf{r},t)\equiv\sum_{\mathbf{r}_i\ (\in\delta v)}\frac{V_{\mathbf{r}_i}(t)}{\delta v}\ , \tag{3.20}$$

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

$$V(\mathbf{r},t)=\frac{N_0}{\delta v}V_{\mathbf{r}}(t)=\rho_a V_{\mathbf{r}}(t)\ , \tag{3.21}$$

where ρ_a is the atomic number density. Equivalently, $V_{\mathbf{r}}(t)$ can be taken as an average by setting

$$V_{\mathbf{r}}(t)=\sum_{i=1}^{N_0}\frac{V_{\mathbf{r}_i}(t)}{N_0}\ . \tag{3.22}$$

Reexpanding $V_i(t)$ of Eq. (3.17) using the recursive relation (3.18), we obtain the following expression for $V_{\mathbf{r}}(t)$, which is correct up to the third order in $\mathcal{D}_j(t)$:

$$V_r(t)=-\sum_j \exp(-i\omega_j^s t)\mathcal{D}_j(t)\left[\frac{\mu_d g_j \exp(i\mathbf{k}_j\cdot\mathbf{r})}{\gamma_\perp(1+i\delta_{aj})S}\right] + \sum_j \sum_{n\,(\neq l)} \sum_l \exp[-i(\omega_j^s-\omega_l^s+\omega_n^s)t]\mathcal{D}_j(t)\mathcal{D}_l^+(t)\mathcal{D}_n(t)\frac{\mu_d g_j \exp[i(\mathbf{k}_j+\mathbf{k}_n-\mathbf{k}_l)\cdot\mathbf{r}]}{\gamma_\perp[1+i(\delta_{aj}+\delta_{ln})]SS_{ln}F_{ln}I_{ln}} + \Gamma_{V_r}(t)\ . \tag{3.23}$$

Here, the coupling coefficient g_j and the dipole matrix element μ_d are as defined in Eqs. (2.6) and (2.7), respectively,

$$\delta_{aj}\equiv(\omega_a-\omega_j^s)/\gamma_\perp\ , \tag{3.24}$$

$$\delta_{lj}\equiv(\omega_l^s-\omega_j^s)/\gamma_\perp\ , \tag{3.25}$$

$$I_{ln}\equiv\frac{\gamma_\parallel\gamma_\perp(1-i\delta_{al})(1+i\delta_{an})}{4g_l g_n|\mu_d|^2}\ , \tag{3.26}$$

$$S\equiv 1+\sum_{n:\omega_n^s=\omega_l^s}\sum_l \frac{\alpha_l^+\alpha_n\exp[-i(\mathbf{k}_l-\mathbf{k}_n)\cdot\mathbf{r}]}{I_l}\ , \tag{3.27}$$

$$I_l\equiv\frac{\gamma_\parallel\gamma_\perp(1+\delta_{al}^2)}{4g_l^2|\mu_d|^2}\ , \tag{3.28}$$

$$S_{ln}\equiv 1+\sum_{m:\omega_m^s=\omega_j^s}\sum_j \frac{\alpha_j^+\alpha_m\exp[-i(\mathbf{k}_j-\mathbf{k}_m)\cdot\mathbf{r}]}{I_{lnj}}\ , \tag{3.29}$$

$$I_{lnj}\equiv\frac{\gamma_\parallel\gamma_\perp}{4g_j^2|\mu_d|^2}F_{ln}[1+i(\delta_{ln}+\delta_{aj})] \times[1-i(\delta_{nl}+\delta_{aj})]\frac{1+i\delta_{ln}/2}{1+i\delta_{ln}}\ , \tag{3.30}$$

$$F_{ln}\equiv\frac{1+i\delta_{ln}/2F}{1+i\delta_{ln}/2}\ , \tag{3.31}$$

$$\Gamma_{V_r}(t)\equiv\sum_{i=1}^{N_0}\frac{\Gamma_{V_i}(t)}{N_0}\ , \tag{3.32}$$

and $F\equiv\gamma_\parallel/2\gamma_\perp$.

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. S as defined by Eq. (3.27) is the usual saturation factor and S_{ln} of Eq. (3.29) can be regarded as a generalized saturation factor when more than one modes of different frequencies are present. I_{lnj} of Eq. (3.30) represents the corresponding generalized normalized saturation intensity. The generalizations become apparent if we note that $I_{njl}|_{\omega_n^s=\omega_j^s}=I_{nl}|_{\omega_n^s=\omega_l^s}=I_l$, and $S_{nl}|_{\omega_n^s=\omega_l^s}=S$. Similarly, the effect of collisions is expressed by the generalized collision factor F_{ln} 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.*[5] 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 ω_j^s of the modes, the main difference from the usual adiabatic elimination procedure is the appearance of higher-order derivative terms in $\mathcal{D}_j(t)$. 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 $V_i(t)$ [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 $\Omega_p\equiv\omega_p^s$, 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 $\mathbf{k}_{p_1}$ and $\mathbf{k}_{p_2}$ and their slowly varying amplitudes by $\alpha_{p_1}(t)$ and $\alpha_{p_2}(t)$, respectively. Let us further define $\tilde{\Gamma}_{V_i}(\omega)$ and $\tilde{\Gamma}_{V_i^+}(\omega)$ as follows:

$$\Gamma_{V_i}(t)\equiv\int_{-\infty}^{\infty}\tilde{\Gamma}_{V_i}(\omega)\exp[-i(\omega+\Omega_p)t]d\omega\ , \tag{4.1}$$

$$\Gamma_{V_i^+}(t)\equiv\int_{-\infty}^{\infty}\tilde{\Gamma}_{V_i^+}(\omega)\exp[-i(\omega-\Omega_p)t]d\omega\ , \tag{4.2}$$

so that $\tilde{\Gamma}_{V_i}(\omega)=\Gamma_{V_i}(\omega+\Omega_p)$ and $\tilde{\Gamma}_{V_i^+}(\omega)=\Gamma_{V_i^+}(\omega-\Omega_p)$. 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:

$$\langle\tilde{\Gamma}_{V_i^+}(\omega)\tilde{\Gamma}_{V_{i'}}(\omega')\rangle=\frac{D_{V_i^+V_i}(d)}{2\pi}\delta(\omega+\omega')\delta_{ii'}\ , \tag{4.3}$$

$$\langle\tilde{\Gamma}_{V_i}(\omega)\tilde{\Gamma}_{V_{i'}}(\omega')\rangle=\frac{D_{V_iV_i}(d)}{2\pi}\delta(\omega+\omega')\delta_{ii'}\ , \tag{4.4}$$

$$\langle\tilde{\Gamma}_{V_i^+}(\omega)\tilde{\Gamma}_{V_{i'}^+}(\omega')\rangle=\frac{D_{V_i^+V_i^+}(d)}{2\pi}\delta(\omega+\omega')\delta_{ii'}\ , \tag{4.5}$$

where $d\equiv\omega/\gamma_\perp$,

$$D_{V_i^+V_i}(d)=\frac{\beta^2F}{2\gamma_\perp D(d)}\{(1-F)[d^4+2\Delta_p d^3+d^2(4F^2+1-\beta^2F+\Delta_p^2)-d\Delta_p(\beta^2F-8F^2-2\beta^2F^2)$$
$$+4F^2+\Delta_p^2(4F^2+2\beta^2F^2)]+d^2\beta^2F^2+4\beta^2F^2+\beta^4F^2/2\}\ , \tag{4.6}$$

$$D_{V_iV_i}(d)=-\frac{\beta^2F}{2\gamma_\perp D(d)}\frac{A_p^2}{A_p^+A_p}((1-F)(-d^2\beta^2F+2\Delta_p^2\beta^2F^2)+Fd^4+d^2[4F^3+F-3(\Delta_p^2+\beta^2F)F]$$
$$+4F^3+\beta^4F^2/2-12\Delta_p^2F^3+i\Delta_p\{(1-F)(-2\beta^2F^2)+d^4F+d^2[4F^3+3F-(\Delta_p^2+\beta^2F)F]$$
$$+12F^3-4\Delta_p^2F^3\})\ , \tag{4.7}$$

$$D_{V_i^+V_i^+}(d)=D^*_{V_iV_i}(d)\ , \tag{4.8}$$

$$D(d)=(F+\Delta_p^2F+\beta^2F/2)\{d^6+d^4(2+4F^2-2\beta^2F-2\Delta_p^2)$$
$$+d^2[1+8F^2+2\beta^2F(2F-1)+\beta^4F^2+\Delta_p^4-\Delta_p^2(8F^2-2\beta^2F-2)]$$
$$+4F^2+4\beta^2F^2+\beta^4F^2+\Delta_p^2(8F^2+4\beta^2F^2)+4\Delta_p^4F^2\}\ , \tag{4.9}$$

$\Delta_p\equiv(\Omega_p-\omega_a)/\gamma_\perp$ is the normalized pump-frequency detuning from the atomic resonance, $\beta^2\equiv 4A_p^+A_p/\gamma_\perp^2F=8A_p^+A_p/\gamma_\perp\gamma_\parallel$, and

$$A_p\equiv g_{p_1}|\mu_d|\alpha_{p_1}(t)\exp(-i\mathbf{k}_{p_1}\cdot\mathbf{r}_i)+g_{p_2}|\mu_d|\alpha_{p_2}(t)\exp(-i\mathbf{k}_{p_2}\cdot\mathbf{r}_i)\ .$$

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 $\alpha_{p_1}=\alpha_{p_2}$, β^2 can be simply related to the pump intensity I_p and the line-center saturation intensity $I_{\rm sa}$ via $\beta^2=2I_p/I_{\rm sa}(\Omega_p/\omega_a)^2$, where $I_p=8\epsilon_0c(\Omega_p/\omega_a)^2|\alpha_{p_1}|^2$ and $I_{\rm sa}=\epsilon_0c(\gamma_\perp\gamma_\parallel/2)(\hbar/|\mu_d|)^2$. Here, β is known as the Rabi frequency. Note that in arriving at the above relation, we have used the following relation between α_{p_1} and the net electric-field amplitude of the pump beams $\mathscr{E}_p$ [for the exponential mode $\exp(-i\mathbf{k}_{p_1}\cdot\mathbf{r})$]: $\mathscr{E}_p=\hbar g_{p_1}2\alpha_{p_1}\Omega_p/\omega_a$. We also note that in the presence of collisions (i.e., $F<1$), $D_{V_i^+V_i}(d)$ is not an even function of d, whereas both $D_{V_iV_i}(d)$ and $D_{V_i^+V_i^+}(d)$ remain even. Furthermore, we show in the Appendix that our expression for $D_{V_i^+V_i}(d)$ [Eq. (4.6)] gives the correct resonance-fluorescence spectrum as first obtained by Mollow in the collisionless limit.[6]

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:

$$\frac{\partial}{\partial t}\alpha_m(t)=-i\Delta\omega_m\alpha_m(t)+\int_{V_M}d\mathbf{r}\,\rho_aC_m^*(\mathbf{r})V_{\rm r}(t)\exp(i\omega_m^st)\ . \tag{5.1}$$

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 $\sum_i$ appearing in Eq. (2.1) by the integral $\int_{V_M}d\mathbf{r}\,\rho_a$. With $V_{\rm r}(t)$ 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 $\alpha_{p_1}(t)=\alpha_{p_2}(t)\simeq\alpha_{p_1}=$const, 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 α_m/α_{p_1}:

$$\frac{\partial}{\partial t}\alpha_m(t) = -i\Delta\omega_m\alpha_m(t)$$
$$+\int_{V_M} d\mathbf{r}\,\rho_a$$
$$\times\Bigg[-\sum_{n\,(\neq p_1,p_2)}\exp[i(\omega_m^s-\omega_n^s)t]\mathcal{D}_n\left[\frac{|\mu_d|^2 g_m g_n\exp[i(\mathbf{k}_n-\mathbf{k}_m)\cdot\mathbf{r}]}{\gamma_\perp(1+i\delta_{an})S}\right]$$
$$-\exp[i(\omega_m^s-\omega_p^s)t]D_p\left[\frac{|\mu_d|^2 g_m g_p\{\exp[i(\mathbf{k}_{p_1}-\mathbf{k}_m)\cdot\mathbf{r}]+\exp[i(\mathbf{k}_{p_2}-\mathbf{k}_m)\cdot\mathbf{r}]\}}{\gamma_\perp(1+i\delta_{ap})S}\right]$$
$$+\sum_{n\,(\neq p_1,p_2)}\exp[i(\omega_m^s-\omega_n^s)t]D_p^+ D_p\mathcal{D}_n$$
$$\times\left[\frac{|\mu_d|^2 g_m g_p\exp[-i(\mathbf{k}_m-\mathbf{k}_n)\cdot\mathbf{r}]\{\exp[i(\mathbf{k}_{p_1}-\mathbf{k}_{p_2})\cdot\mathbf{r}]+\exp[-i(\mathbf{k}_{p_1}-\mathbf{k}_{p_2})\cdot\mathbf{r}]+2\}}{\gamma_\perp(1+i\delta_{an})SS_{pn}F_{pn}I_{pn}}\right]$$
$$+\sum_{n\,(\neq p_1,p_2)}\exp[i(\omega_m^s+\omega_n^s-2\omega_p^s)t]D_pD_p\mathcal{D}_n^+|\mu_d|^2 g_m g_p\exp[-i(\mathbf{k}_m+\mathbf{k}_n)\cdot\mathbf{r}]$$
$$\times\left[\frac{\exp(2i\mathbf{k}_{p_1}\cdot\mathbf{r})+\exp(2i\mathbf{k}_{p_2}\cdot\mathbf{r})+2\exp[i(\mathbf{k}_{p_1}+\mathbf{k}_{p_2})\cdot\mathbf{r}]}{\gamma_\perp[1+i(\delta_{ap}+\delta_{np})]SS_{np}F_{np}I_{np}}\right]$$
$$+g_m\mu_d^*\Gamma_{V_r}(t)\exp(-i\mathbf{k}_m\cdot\mathbf{r})\exp(i\omega_m^s t)\Bigg], \tag{5.2}$$

where $\mathcal{D}_n$ and $\mathcal{D}_n^+$ are given by Eqs. (3.16) and (3.19), respectively, and $D_p=\mathcal{D}_{p_1}=\mathcal{D}_{p_2}=\alpha_{p_1}$ 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

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

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

$$I_p=\gamma_\perp\gamma_\parallel(1+\delta_{ap}^2)/4g_p^2|\mu_d|^2, \tag{5.5}$$

$$I_{npp}=\frac{\gamma_\parallel\gamma_\perp}{4g_p^2|\mu_d|^2}F_{np}[1+i(\delta_{np}+\delta_{ap})](1-i\delta_{an})\left[\frac{1+i\delta_{np}/2}{1+i\delta_{np}}\right], \tag{5.6}$$

$$F_{np}=(1+i\delta_{np}/2F)/(1+i\delta_{np}/2)$$
$$=F_{pn}^*, \tag{5.7}$$

$$I_{np}=\frac{\gamma_\parallel\gamma_\perp(1-i\delta_{an})(1+i\delta_{ap})}{4g_n g_p|\mu_d|^2}$$
$$=I_{pn}^*. \tag{5.8}$$

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:

$$\frac{\partial}{\partial t}\alpha_m(t) = -i\Delta\omega_m\alpha_m(t) - \int_{V_Q} d\mathbf{r}\left[\mathcal{D}_m\left[\frac{\rho_a|\mu_d|^2 g_m^2}{\gamma_\perp(1+i\delta_{am})S}\right] + \mathcal{D}_m\left[\frac{2\rho_a|\mu_d|^2 g_m g_p \alpha_{p_1}^+\alpha_{p_1}}{\gamma_\perp(1+i\delta_{am})SS_{pm}F_{pm}I_{pm}}\right] + \sum_{\{j:\mathbf{k}_m-\mathbf{k}_j=\pm(\mathbf{k}_{p_1}-\mathbf{k}_{p_2})\}} \exp[i(\omega_m^s-\omega_j^s)]\mathcal{D}_j\left[\frac{\rho_a|\mu_d|^2 g_m g_p \alpha_{p_1}^+\alpha_{p_1}}{\gamma_\perp(1+i\delta_{aj})SS_{pj}F_{pj}I_{pj}}\right] + \exp(i\delta\omega_m^s t)\mathcal{D}_n^+\left[\frac{2\rho_a|\mu_d|^2 g_m g_p \alpha_{p_1}^2}{\gamma_\perp[1+i(\delta_{ap}+\delta_{np})]SS_{np}F_{np}I_{np}}\right] + \sum_{\{j:\mathbf{k}_m+\mathbf{k}_j=2\mathbf{k}_{p_1}\text{ or }2\mathbf{k}_{p_2}\}} \exp[i(\omega_m^s+\omega_j^s-2\omega_p^s)t]\mathcal{D}_j^+\left[\frac{\rho_a|\mu_d|^2 g_m g_p \alpha_{p_1}^2}{\gamma_\perp[1+i(\delta_{ap}+\delta_{jp})]SS_{jp}F_{jp}I_{jp}}\right]\right] + \Gamma_m(t) , \tag{5.9}$$

where

$$\Gamma_m(t) = g_m\mu_d^* \int_{V_Q} d\mathbf{r}\,\rho_a\Gamma_{V_r}(t) \times \exp(-i\mathbf{k}_m\cdot\mathbf{r})\exp(i\omega_m^s t) . \tag{5.10}$$

Note that S and S_{pn} in the denominators of the terms in Eq. (5.9) are $\mathbf{r}$ dependent. This is why our retaining of the phase-matching terms in Eq. (5.9) is only approximate, except in the special case when $\mathbf{k}_{p_1}=\mathbf{k}_{p_2}$. The spatial integrals can be evaluated analytically as is done by Reid and Walls[8] in the case of degenerate four-wave mixing.

In the fifth term of Eq. (5.9),

$$\delta\omega_m^s \equiv \omega_m^s + \omega_n^s - 2\omega_p^s , \tag{5.11}$$

where ω_n^s is the shifted frequency of mode n [cf. Eqs. (3.2) and (3.5)] picked by the phase-matching condition

$$\mathbf{k}_m + \mathbf{k}_n = \mathbf{k}_{p_1} + \mathbf{k}_{p_2} . \tag{5.12}$$

The sixth term of Eq. (5.9) represents three-wave mixing with $\mathbf{k}_m+\mathbf{k}_j=2\mathbf{k}_{p_1}$ or $\mathbf{k}_m+\mathbf{k}_j=2\mathbf{k}_{p_2}$ 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 $\{\alpha^+\}$ appearing in the differential operators $\{\mathcal{D}^+\}$ [cf. Eq. (3.19)].

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

In the following, we will use $\alpha_{\tilde{m}}(t)$, instead of $\alpha_n(t)$, to denote the image mode to $\alpha_m(t)$, i.e., the mode whose conjugate α_n^+ couples to α_m via the nonlinear terms of Eq. (5.9). So whenever the subscript $\tilde{m}$ 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.[2] The results of one such application have been published.[9]

VI. SINGLE-BEAM INFINITE MEDIUM CASE

In this section, we derive the temporal coupled-mode equations for a single pump beam of wave vector $\mathbf{k}_p$ and amplitude $\alpha_p(t)\simeq 2\alpha_{p_1}$ propagating through an infinite medium. The equation of motion for a copropagating nondegenerate mode with frequency ω_m^s, which is different from the pump-mode frequency ω_p^s, 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:

$$\frac{\partial\alpha_m}{\partial t} = -i\Delta\omega_m\alpha_m + \gamma_m\alpha_m + i\frac{\partial\alpha_m}{\partial t}\frac{\partial\gamma_m}{\partial\omega_m^s} + X_m\exp(i\delta\omega_m^s t)\alpha_{\tilde{m}}^+ + i\exp(i\delta\omega_m^s t)\frac{\partial\alpha_{\tilde{m}}^+}{\partial t}\frac{\partial X_m}{\partial\omega_m^s} + \Gamma_m , \tag{6.1}$$

$$\frac{\partial\alpha_{\tilde{m}}^+}{\partial t} = i\Delta\omega_{\tilde{m}}\alpha_{\tilde{m}}^+ + \gamma_{\tilde{m}}^*\alpha_{\tilde{m}}^+ - i\frac{\partial\alpha_{\tilde{m}}^+}{\partial t}\frac{\partial\gamma_{\tilde{m}}^*}{\partial\omega_{\tilde{m}}^s} + X_{\tilde{m}}^*\exp(-i\delta\omega_m^s t)\alpha_m - i\exp(-i\delta\omega_m^s t)\frac{\partial\alpha_m}{\partial t}\frac{\partial X_{\tilde{m}}^*}{\partial\omega_{\tilde{m}}^s} + \Gamma_{\tilde{m}}^+ , \tag{6.2}$$

where

$$\gamma_m = -\frac{\rho_a V_Q g_m^2|\mu_d|^2}{\gamma_\perp(1+i\delta_{am})\tilde{S}}\left[1-\frac{4g_p^2\alpha_{p_1}^+\alpha_{p_1}}{\tilde{S}_{pm}F_{pm}I_{pm}g_p g_m}\right] , \tag{6.3}$$

$$X_m = \frac{4\rho_a V_Q g_m g_{\bar{m}} g_p^2 |\mu_d|^2 \alpha_{p_1}^2}{\gamma_\perp [1 + i(\delta_{ap} + \delta_{\bar{m}p})] \tilde{S} \tilde{S}_{\bar{m}p} F_{\bar{m}p} I_{\bar{m}p} g_p g_{\bar{m}}} , \tag{6.4}$$

$$\mathbf{k}_m + \mathbf{k}_{\bar{m}} = 2\mathbf{k}_p , \tag{6.5}$$

$$\delta\omega_m^s = \omega_m^s + \omega_{\bar{m}}^s - 2\omega_p^s , \tag{6.6}$$

$$\tilde{S} = 1 + \frac{4\alpha_{p_1}^+ \alpha_{p_1}}{I_p} , \tag{6.7}$$

$$\tilde{S}_{mp} = 1 + \frac{4\alpha_{p_1}^+ \alpha_{p_1}}{I_{mpp}} = \tilde{S}_{pm}^* , \tag{6.8}$$

and the remaining symbols are as defined in Eqs. (5.4)–(5.8).

To determine ω_p^s, 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 $\alpha_p(t)$:

$$\frac{\partial \alpha_p(t)}{\partial t} = (-i\Delta\omega_p + \gamma_p)\alpha_p(t) , \tag{6.9}$$

where

$$\gamma_p = -\frac{\rho_a V_Q g_p^2 |\mu_d|^2}{\gamma_\perp (1 + i\delta_{ap}) \tilde{S}} . \tag{6.10}$$

Defining $\gamma_p \equiv \gamma_{Rp} + i\gamma_{Ip}$, we have

$$\gamma_{Rp} = -\frac{\rho_a V_Q g_p^2 |\mu_d|^2}{\gamma_\perp (1 + \delta_{ap}^2) \tilde{S}} , \tag{6.11}$$

and

$$\gamma_{Ip} = \frac{\rho_a V_Q g_p^2 |\mu_d|^2 \delta_{ap}}{\gamma_\perp (1 + \delta_{ap}^2) \tilde{S}} . \tag{6.12}$$

Clearly, the loss coefficient γ_{Rp} must be small if our assumption of a constant $\alpha_p(t) \simeq 2\alpha_{p_1}$ is to be valid. In this case, the solution for $\alpha_p(t)$ is given by

$$\alpha_p(t) \simeq \exp[-i(\Delta\omega_p - \gamma_{Ip})t]\alpha_p(0) . \tag{6.13}$$

Hence in order for $\alpha_p(t)$ to be truly slowly varying so that $\alpha_p(t) \simeq 2\alpha_{p_1}$, we must set $\Delta\omega_p = \gamma_{Ip}$. Then, from defining Eqs. (3.2)–(3.5), we determine $\omega_p^s = \gamma_{Ip} + |\mathbf{k}_p| c$. This implies that the refractive index seen by the pump beam is given by $n_p = 1 + \gamma_{Ip}/|\mathbf{k}_p| c$.

Following a similar argument for the other modes, we also have $\Delta\omega_m = \mathrm{Im}(\gamma_m)$ with γ_m given by Eq. (6.3). We note from Eqs. (6.3) and (6.12) that even when $\omega_m^s \simeq \omega_p^s$, $\Delta\omega_m$ 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 $\delta\omega_m^s$ 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 $(i/\omega_a)(\partial V_i/\partial t)$, instead of V_i as mentioned after Eq. (2.14), then we would have obtained $(\partial\alpha_m/\partial t)[i(\partial\gamma_m/\partial\omega_m^s) + i(\gamma_m/\omega_m^s)]$ instead of $(\partial\alpha_m/\partial t) i(\partial\gamma_m/\partial\omega_m^s)$ in Eq. (6.1). Also the coefficient γ_m in Eq. (6.3) would have been multiplied by the factor ω_m^s/ω_a. Similar modifications would also apply to the coefficient X_m and the $\partial\alpha_{\bar{m}}^+/\partial t$ term in Eq. (6.1). The physical significance of the $\partial\alpha_m/\partial t$ term 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 $|\gamma_m| \ll \omega_m^s$ and that the $\partial\alpha_m/\partial t$ term 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.

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation under Grant No. ECS-8718970 and No. EET-8715275.

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.*[5] and Mollow.[6]

In the paper of Boyd *et al.*,[5] E_1 corresponds to our pump field A_p, E_3 corresponds to our probe field α_m, ρ_{ab} equals our V_i, ρ_{bb} equals our n_i, ρ_{aa} equals our n_{gi}, $1/T_1$ equals our $\gamma_\parallel$, $1/T_2$ equals our $\gamma_\perp$, μ_{ab} equals our μ_d, ω_1 equals our Ω_p, ω_3 equals our $\Omega_m \equiv \omega_m^s$, and ω_{ba} equals our ω_a. With these identifications, it can be shown that their expression for $\rho_{ba}(\omega_1)$ [Eq. (5a) in their paper] is identical to the $(j = p)$ term of the first term of Eq. (3.23), i.e.,

$$\rho_{ba}(\Omega_p) = -\frac{A_p \mu_d g_p}{\gamma_\perp (1 + i\delta_{ap}) S} , \tag{A1}$$

where we have let $\mathcal{D}_j(t)|_{\omega_j^s = \Omega_p} = A_p$. The expression for $\rho_{ba}(\omega_3)$ in their paper [Eq. (5b)] can be obtained by add-

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.,

$$\rho_{ba}(\Omega_m)=-\frac{\alpha_m\mu_d g_m}{\gamma_\perp(1+i\delta_{am})S}+\frac{A_p^+A_p\alpha_m\mu_d g_p}{\gamma_\perp(1+i\delta_{am})SS_{pm}F_{pm}I_{pm}}\ . \tag{A2}$$

Finally, the expression for $\rho_{ba}(2\omega_1-\omega_3)$ 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.,

$$\rho_{ba}(2\Omega_p-\Omega_m)=\frac{A_p^2\alpha_m^+\mu_d g_p}{\gamma_\perp[1+i(\delta_{ap}+\delta_{mp})]SS_{mp}F_{mp}I_{mp}}\ . \tag{A3}$$

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

$$\langle V_r^+(t)V_r(t+\tau)\rangle=|V_\infty|^2\exp(-i\Omega_p\tau)+\langle\Gamma_{V_r^+}(t)\Gamma_{V_r}(t+\tau)\rangle\ , \tag{A4}$$

where

$$V_\infty=-\frac{A_p\mu_d g_p}{\gamma_\perp(1+i\delta_{ap})S}\ , \tag{A5}$$

and evaluate the Fourier transform

$$\begin{aligned}\tilde g(\omega)&\equiv\int_{-\infty}^{\infty}\langle V_r^+(t)V_r(t+\tau)\rangle\exp(i\omega\tau)d\tau\\&=2\pi|V_\infty|^2\delta(\Omega_p-\omega)+\int_{-\infty}^{\infty}\langle\Gamma_{V_r^+}(t)\Gamma_{V_r}(t+\tau)\rangle\exp(i\omega\tau)d\tau\ .\end{aligned} \tag{A6}$$

Simple manipulation shows that the expression for V_∞ 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 $\gamma_\perp=\kappa/2$, $(\Omega_p-\omega_a)=\Delta\omega$, and $\gamma_\perp^2\beta^2=\Omega^2$.

[1]S. -T. Ho, P. Kumar, and J. H. Shapiro, Phys. Rev. A **37**, 2017 (1988).

[2]S. -T. Ho, N. C. Wong, J. H. Shapiro, and P. Kumar, *Technical Digest Series* (Optical Society of America, Washington, D.C., 1988), Vol. 11, p. 84; M. W. Maeda, P. Kumar, and J. H. Shapiro, J. Opt. Soc. Am. B **4**, 1501 (1987); Opt. Lett. **12**, 161 (1987); Phys. Rev. A **32**, 3803 (1985).

[3]S. -T. Ho, P. Kumar, and J. H. Shapiro, Phys. Rev. A **35**, 3982 (1987); J. Opt. Soc. Am. B **3**, P64 (1986).

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

[5]R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A **24**, 411 (1981); T. Fu and M. Sargent III, Opt. Lett. **4**, 366 (1979).

[6]B. R. Mollow, Phys. Rev. **188**, 1969 (1969); H. J. Kimble and L. Mandel, Phys. Rev. A **13**, 2123 (1976).

[7]M. D. Reid and D. F. Walls, Phys. Rev. A **34**, 4929 (1986).

[8]M. D. Reid and D. F. Walls, Phys. Rev. A **31**, 1622 (1985).

[9]S. -T. Ho, P. Kumar, and J. H. Shapiro, J. Opt. Soc. Am. B **8**, 37 (1991).