Quantum theory of nonlinear mixing in multimode fields: General theory

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A general quantum theory of nonlinear mixing in multimode fields is formulated. The theory is valid for arbitrary media and thus includes various diverse cases such as those corresponding to nonlinear mixing in multilevel systems, optical fibers, etc. Thus Zeeman coherence effects are automatically included. It is applicable to both degenerate and nondegenerate mixing experiments. The theory is in terms of two distinct sets of correlation functions of the polarization operators. One set of correlations is related to the nonlinear susceptibilities. The other set has no counterpart in semiclassical theory and is related to the quantum-mechanical fluctuations of the polarization operator. The general structure of the evolution of the density matrix of the generated fields is discussed. The quantum statistics of the generated fields can be studied in terms of the Wigner distribution function which is explicitly evaluated. Higher-order squeezing characteristics of the field are discussed. Conditions under which the quantization of the semiclassical equations is adequate are examined. The application of the quantum theory to nonlinear mixing in two-photon media is also presented. The theory is also capable of accounting for the interparticle correlations.

I. INTRODUCTION

The classical theory of four-wave mixing and, more generally nonlinear mixing is well understood.[1] The third-order nonlinear susceptibilities $\chi^{(3)}(\omega_2,\omega_2,-\omega_1)$, $\chi^{(3)}(\omega_1,-\omega_1,\omega_1)$, and $\chi^{(3)}(\omega_2,-\omega_2,\omega_2)$ and linear susceptibilities $\chi^{(1)}(\omega_1)$ and $\chi^{(1)}(\omega_2)$ determine the structure of the generated fields. For resonant systems it may be necessary to include saturation effects by calculating the analogous intensity-dependent susceptibilities.[2-6] The nature of the medium enters through these susceptibilities and the knowledge of susceptibilities is sufficient for studying the characteristics of the generated waves. However, for studies of the statistics of the generated fields one needs to have a quantum theory of nonlinear mixing. The quantum theory is interesting not only in its own right but is also needed in studies on the fundamental characteristics of the radiation fields such as squeezing.[7-10] Several groups have already made important contributions to the quantum theory of four-wave mixing.[11-13] Our objective is different here and we present a theory with the following specific questions in mind: How does one formulate a quantum theory which is general enough to include four-wave mixing in a variety of resonant and nonresonant media? Is the quantization of semiclassical equations of nonlinear mixing adequate? Can the quantum theory be completely formulated in terms of the nonlinear susceptibilities or does one need other characteristics, hitherto unknown in the semiclassical theory? One would expect that quantum fluctuations associated with the medium will enter in any quantum theory. This is because the semiclassical theory is a mean-field theory and it uses a mean-field characterization of the material medium in terms of the susceptibilities.

The purpose of this paper is to present a general formulation of the quantum theory of nonlinear mixing. Our theory is in terms of the nonlinear susceptibilities and a set of correlation functions of the polarization operators, and it includes both the cases of degenerate and nondegenerate mixings. Such correlation functions can be calculated for specific systems. We present the general structure of the density matrix of the generated field. This is independent of the nature of the medium in which nonlinear mixing takes place. Thus questions on the photon statistics of the generated field can be answered.

The organization of this paper is as follows. In Sec. II we formulate the general theory of nonlinear mixing in multimode fields. No approximation is made regarding the strength of the pump field, whereas both the probe and the generated fields are assumed to be weak. The weak fields are treated perturbatively [cf. Eq. (2.14)]. In Sec. III we obtain the dynamical equation describing nonlinear mixing for the case when each of the fields is a single-mode field—these results translate easily to the case of nonlinear mixing with traveling waves. In Sec. IV we discuss the physical meaning of various coefficients in the density-matrix equation. In Sec. V we present the complete solution for the density matrix of the generated fields. This is done via the Wigner distribution function,[14] which can be used to discuss statistical aspects of the generated fields. If initially the generated and probe fields are in coherent states, then at time t the Wigner function will be Gaussian in two complex field amplitudes. Such Gaussian Wigner functions are shown to be especially useful in the context of higher-order squeezing[15] of the fields. In Sec. VI we show how the coefficients in the basic density-matrix equation can be obtained for a class of important systems: (i) two-level transitions, (ii) transitions in V systems, and (iii) optical fibers. In

Sec. VII we present the quantum theory of nonlinear mixing in two-photon media.[16] In a second paper of this series of papers we will present numerical results on the quantum theory of nonlinear mixing in three-level systems. We will discuss the quantum theory of optical phase conjugation in a third paper. Optical phase conjugation typically involves standing waves and thus a complete quantum theory of phase conjugation has to use a multimode description of the type presented in Sec. II of this paper.

II. QUANTUM STATISTICAL THEORY OF NONLINEAR MIXING

In this section we develop a very general quantum theory of nonlinear mixing. In our theory the role of the different types of nonlinear susceptibilities can be clearly seen. In addition, the quantum theory will involve certain strictly quantum-mechanical correlation functions which will not have any classical counterpart. We will treat the pump field as a prescribed classical field and its depletion due to the interaction with the medium will be ignored. The probe field and the field generated in the process of nonlinear mixing will be treated quantum mechanically. The fields interact with a number of atoms distributed over the cell volume. We assume that the time scales associated with the atomic dynamics are much *smaller* than those associated with the field dynamics. Our approach will consist of finding the dynamical equation for the density matrix of the probe and generated fields.

In dipole approximation, the interaction of the atoms and the fields can be written as

$$H_1 = -\int \mathbf{P}(\mathbf{r})\cdot\mathbf{E}_p(\mathbf{r},t)d^3r - \int \mathbf{P}(\mathbf{r})\cdot\mathbf{E}(\mathbf{r},t)d^3r \ . \tag{2.1}$$

Here, $\mathbf{P}(\mathbf{r})$ is the polarization operator associated with the medium. The pump field $\mathbf{E}_p(\mathbf{r},t)$, which is treated semiclassically, is taken to have the form

$$\mathbf{E}_p(\mathbf{r},t) = \varepsilon_p(\mathbf{r})e^{-i\omega t} + \text{c.c.} \tag{2.2}$$

The polarization operator can be expressed, in terms of the dipole operator $\mathbf{d}(i)$ associated with an atom located at $\mathbf{R}^{(i)}$, as

$$\mathbf{P}(\mathbf{r}) = \sum_i \delta(\mathbf{r}-\mathbf{R}^{(i)})\mathbf{d}^{(i)} \ . \tag{2.3}$$

The probe and generated fields are represented by the operator $\mathbf{E}(\mathbf{r},t)$. If the probe field has the frequency ω_s, then the generated field will have frequency $2\omega-\omega_s$. In such a case the field $\mathbf{E}(\mathbf{r},t)$ will have frequency components $\omega_s, 2\omega-\omega_s$. We are interested in resonant situations and therefore we would derive results valid to all orders in $\mathbf{E}_p$. For resonant situations we can make the rotating-wave approximation. We introduce positive and negative frequency parts of various fields and the polarization operator. The rotating-wave approximation simplifies (2.1) to

$$H_1 = -\int d^3r\, \mathbf{P}^-(\mathbf{r})\cdot[\mathbf{E}_p^+(\mathbf{r},t)+\mathbf{E}^+(\mathbf{r},t)] + \text{H.c.} \tag{2.4}$$

Note that $\mathbf{E}_p^+$ has a time dependence $e^{-i\omega t}$, whereas in the interaction picture the operator $\mathbf{P}^-(\mathbf{r})$ has the time dependence

$$\mathbf{P}^-(\mathbf{r},t) = \sum_{i,\alpha,\beta}{}' \delta(\mathbf{r}-\mathbf{R}^{(i)})\mathbf{d}_{\alpha\beta}^{(i)}|\alpha\rangle\langle\beta|e^{+i\omega_{\alpha\beta}t} \ . \tag{2.5}$$

The eigenstates of the atom are represented by $|\alpha\rangle$ and $\omega_{\alpha\beta}$ gives the energy separation between the levels $|\alpha\rangle$ and $|\beta\rangle$. The summation in (2.5) is over all values of α and β such that $\omega_{\alpha\beta}>0$.

The density matrix ρ of the coupled atom-field system obeys the equation

$$\frac{\partial\rho}{\partial t} = -\frac{i}{\hbar}[H_{0A}+H_{0F}+H_1,\rho] + L_A\rho \ , \tag{2.6}$$

where H_{0F} (H_{0A}) is the unperturbed Hamiltonian of the field (atoms). The Liouville operator L_A represents the relaxation operator for the atoms. It should be borne in mind that L_A is a function of atomic operators only. The losses in the system due to spontaneous emission and collisions are contained in L_A. Since we are dealing with resonant situations, it is possible to make canonical transformation so that the transformed density matrix satisfies the equation

$$\frac{\partial\tilde{\rho}}{\partial t} = -\frac{i}{\hbar}[\tilde{H}_A+\tilde{H}_{AF},\tilde{\rho}] + L_A\tilde{\rho} \ , \tag{2.7}$$

where $\tilde{H}_{AF}$ represents the interaction of the medium with the quantized probe and generated fields

$$\tilde{H}_{AF} = -\int d^3r\, \mathbf{P}^-(\mathbf{r})e^{i\omega t}\cdot\mathbf{E}^+(\mathbf{r},t) + \text{H.c.} \tag{2.8}$$

Here, $\tilde{H}_A$ includes the contribution from the interaction with the pump field. In the absence of the interaction with the probe and generated fields, the atomic dynamics is given by the equation for the atomic density matrix ρ_A

$$\frac{\partial}{\partial t}\tilde{\rho}_A = -\frac{i}{\hbar}[\tilde{H}_A,\tilde{\rho}_A] + L_A\tilde{\rho}_A \ . \tag{2.9}$$

Solution of Eq. (2.9) yields the known results[17,18] on the behavior of the atomic system interacting with a pump field. In what follows we assume that the solution of Eq. (2.9) is known. Let $\tilde{\rho}_A^{(0)}$ be the steady-state solution of (2.9).

We will now derive the dynamical equation for the field density matrix ρ_F defined by

$$\tilde{\rho}_F = \text{Tr}_A\tilde{\rho} \ . \tag{2.10}$$

In what follows we drop the tildes from various quantities. To obtain the reduced equation for ρ_F we use the projection operator techniques.[19] Let $\mathscr{P}$ be the projection operator defined by

$$\mathscr{P}\cdots = \rho_A^{(0)}\text{Tr}_A\cdots \ . \tag{2.11}$$

We next write (2.7) as

$$\frac{\partial\rho}{\partial t} = \mathscr{L}_A\rho + \mathscr{L}_{AF}(t)\rho \tag{2.12}$$

which can be further transformed to

$$\frac{\partial\rho'}{\partial t} = \mathscr{L}'_{AF}(t)\rho', \quad \rho' = e^{-\mathscr{L}_A t}\rho \ ,$$
$$\mathscr{L}'_{AF}(t) = e^{-\mathscr{L}_A t}\mathscr{L}_{AF}(t)e^{\mathscr{L}_A t} \ . \tag{2.13}$$

Using (2.11) and the standard projection-operator techniques we can obtain a closed equation for ρ_F. We also make the following approximations: (i) The probe and the generated fields are so weak that it is sufficient to retain terms up to second order only in $\mathscr{L}_{AF}$, and (ii) the time scales of interest are much slower than the time scales associated with the atomic dynamics, so the Markovian approximation can be made. The resulting equation for ρ_F is then found to be

$$\frac{\partial \rho_F}{\partial t}=[\mathrm{Tr}_A \mathscr{L}'_{AF}(t)\rho_A^{(0)}]\rho_F+\lim_{t\to\infty}\int_0^t d\tau\{\mathrm{Tr}_A \mathscr{L}'_{AF}(t)\mathscr{L}'_{AF}(t-\tau)\rho_A^{(0)} -[\mathrm{Tr}_A \mathscr{L}'_{AF}(t)\rho_A^{(0)}][\mathrm{Tr}_A \mathscr{L}'_{AF}(t-\tau)\rho_A^{(0)}]\}\rho_F(t)\ , \tag{2.14}$$

which, on using the properties of trace and the steady-state nature of $\rho_A^{(0)}$ (i.e., $\mathscr{L}_A\rho_A^{(0)}=0$), reduces to

$$\frac{\partial \rho_F}{\partial t}=\mathrm{Tr}_A \mathscr{L}_{AF}(t)\rho_A^{(0)}\rho_F + \lim_{t\to\infty}\int_0^t d\tau[\mathrm{Tr}_A \mathscr{L}_{AF}(t)e^{\mathscr{L}_A\tau}\mathscr{L}_{AF}(t-\tau)\rho_A^{(0)}\rho_F(t)-\mathrm{Tr}_A \mathscr{L}_{AF}(t)\rho_A^{(0)}\mathrm{Tr}_A \mathscr{L}_{AF}(t-\tau)\rho_A^{(0)}\rho_F(t)]\ . \tag{2.15}$$

All the trace operations in (2.15) can be carried out if we introduce the correlation functions for the atomic polarization operators defined by

$$C_{\alpha\beta}(\tau)=\lim_{t\to\infty}\langle G_\alpha(t+\tau)G_\beta(t)\rangle\ , \tag{2.16}$$

where G_α is one of the atomic polarization operators. The correlation function $C_{\alpha\beta}$ can be calculated in terms of $e^{\mathscr{L}_A\tau}$ and $\rho_A^{(0)}$ by using

$$C_{\alpha\beta}(\tau)=\mathrm{Tr}_A(G_\alpha e^{\mathscr{L}_A\tau}G_\beta\rho_A^{(0)})\ . \tag{2.17}$$

Using (2.8) and the properties of the trace, one can show that

$$\mathrm{Tr}_A \mathscr{L}_{AF}(t)\rho_A^{(0)}\rho_F=\frac{i}{\hbar}\int d^3r[\langle \mathbf{P}^-(\mathbf{r})\rangle\cdot e^{i\omega t}\mathbf{E}^+(\mathbf{r},t)+\langle \mathbf{P}^+(\mathbf{r})\rangle e^{-i\omega t}\cdot\mathbf{E}^-(\mathbf{r},t),\rho_F(t)]\ . \tag{2.18}$$

It is important to note that expectation values like $\langle P^{\mp}\rangle$ are nonzero as the atoms are driven by a coherent pump field of frequency ω. In fact, $\langle \mathbf{P}^+(\mathbf{r})\rangle$ gives the atomic system's response at frequency ω. Such a response depends on all orders of the applied pump field. In view of the structure of Eqs. (2.15), (2.17), and (2.18), we also introduce the correlation function

$$A_{\alpha\beta}(\tau)=\lim_{t\to\infty}[\langle G_\alpha(t+\tau)G_\beta(t)\rangle-\langle G_\alpha(t+\tau)\rangle\langle G_\beta(t)\rangle]\ . \tag{2.19}$$

Detailed calculations using Eqs. (2.8) and (2.15)–(2.19) then lead to the following density-matrix equation for the field

$$\begin{aligned}\frac{\partial \rho_F}{\partial t}=&\left[\frac{i}{\hbar}\int d^3r\langle \mathbf{P}^-(\mathbf{r})\rangle\cdot e^{i\omega t}[\mathbf{E}^+(\mathbf{r},t),\rho_F]+\mathrm{H.c.}\right]\\ &-\frac{1}{\hbar^2}\Big[\int d^3r_1\int d^3r_2\int_0^\infty d\tau[e^{-i\omega t}\mathbf{E}_i^-(\mathbf{r}_1,t),E_j^-(\mathbf{r}_2,t-\tau)e^{-i\omega(t-\tau)}\rho_F A_{ij}^{++}(\mathbf{r}_1,\mathbf{r}_2,\tau)\\ &-\rho_F E_j^-(\mathbf{r}_2,t-\tau)e^{-i\omega(t-\tau)}A_{ji}^{++}(\mathbf{r}_2,\mathbf{r}_1,-\tau)+E_j^+(\mathbf{r}_2,t-\tau)\rho_F e^{i\omega(t-\tau)}\\ &\times A_{ij}^{+-}(\mathbf{r}_1,\mathbf{r}_2,\tau)-\rho_F E_j^+(\mathbf{r}_2,t-\tau)A_{ji}^{-+}(\mathbf{r}_2,\mathbf{r}_1,-\tau)e^{i\omega(t-\tau)}]\\ &+(\omega\to-\omega \text{ and } +\leftrightarrow-)\Big]\ ,\end{aligned} \tag{2.20}$$

where the last term in the square brackets represents the same previous terms with the appropriate variable and superscript changes made. Here the summation over the repeated indices i and j is implied. The atomic correlation functions appearing in Eq. (2.20) are defined by (2.19), i.e., by

$$A_{ij}^{\pm\mp}(\mathbf{r}_1,\mathbf{r}_2,\tau)=\lim_{t\to\infty}[\langle P_i^{\pm}(\mathbf{r}_1,\tau+t)P_j^{\mp}(\mathbf{r}_2,t)\rangle-\langle P_i^{\pm}(\mathbf{r}_1,t+\tau)\rangle\langle P_j^{\mp}(\mathbf{r}_2,t)\rangle]\ . \tag{2.21}$$

The density-matrix equation (2.20) describes the propagation of a multimode field through a nonlinear medium.

The nonlinear medium itself is interacting with a strong pump field. In order to extract the equation relevant for four-wave mixing, we make use of the coherent nature of the radiation generated in four-wave mixing. We thus have to make use of the phase-matching conditions. If the pump and probe fields have, respectively, wave vectors $\mathbf{k}$, and $\mathbf{k}_s$, then the coherent signal is generated in the direction $2\mathbf{k}-\mathbf{k}_s \equiv \mathbf{k}_c$. Thus to obtain the dynamical equation describing the generation of coherent radiation we carry out spatial averaging, i.e., we drop all the rapidly varying spatial terms from Eq. (2.20). The final equation obtained by spatial averaging can be written down only in special cases, since one needs to know the $\mathbf{r}$ dependence of various atomic correlation functions, which in turn depends on the interparticle distribution function and on the traveling or standing-wave nature of the pump field.

III. FORWARD FOUR-WAVE MIXING

We now use the general theory of Sec. II to obtain the dynamical equations for the fields produced in the configuration involving forward four-wave mixing. The pump wave is a traveling wave with wave vector $\mathbf{k}$, i.e., $\varepsilon_p(\mathbf{r})=\varepsilon_p e^{i\mathbf{k}\cdot\mathbf{r}}$. We further assume that the different atoms are uncorrelated, i.e.,

$$A_{ij}^{\pm\pm}(\mathbf{r}_1,\mathbf{r}_2,\tau)=\delta(\mathbf{r}_1-\mathbf{r}_2)A_{ij}^{\pm\pm}(\mathbf{r}_1,\mathbf{r}_1,\tau) , \quad (3.1)$$

otherwise the density-matrix equation (2.20) will involve the interparticle correlation $g(r,r')$. Moreover, since the pump wave is a traveling wave, the density-matrix equations for the atomic system show that[20]

$$A_{ij}^{\pm\pm}(\mathbf{r},\mathbf{r},\tau)=e^{\pm 2i\mathbf{k}\cdot\mathbf{r}}A_{ij}^{\pm\pm}(0,0,\tau) ,$$
$$A_{ij}^{\pm\mp}(\mathbf{r},\mathbf{r},\tau)=A_{ij}^{\pm\mp}(0,0,\tau) , \quad (3.2)$$
$$\langle \mathbf{P}^{-}(\mathbf{r})\rangle=\langle \mathbf{P}^{-}(0)\rangle e^{-i\mathbf{k}\cdot\mathbf{r}} .$$

Let $\mathbf{k}_s$ be the wave vector of the probe and let $\mathbf{k}_c$ be the wave vector of the generated radiation. We write the electric field operator as

$$\mathbf{E}^{(+)}(\mathbf{r},t)=\beta_s\hat{\boldsymbol{\epsilon}}_s a e^{i\mathbf{k}_s\cdot\mathbf{r}-i\omega_s t}+\hat{\boldsymbol{\epsilon}}_c b\beta_c e^{i\mathbf{k}_c\cdot\mathbf{r}-i\omega_c t} , \quad (3.3)$$

where $\hat{\boldsymbol{\epsilon}}_s$ and $\hat{\boldsymbol{\epsilon}}_c$ are, respectively, the unit polarization vectors associated with the probe and generated fields; a and b are the photon annihilation operators for the two modes, $\beta\sim -i\sqrt{2\pi\hbar\omega/V}$ and V is the quantization volume. On substituting (3.3) and using the structure (3.1), (3.2) of the atomic correlation functions, and the phase matching condition

$$2\mathbf{k}=\mathbf{k}_c+\mathbf{k}_s , \quad (3.4)$$

the basic equation (2.20) reduces to

$$\frac{\partial\rho_F}{\partial t}=\left[\frac{\partial\rho_F}{\partial t}\right]_{\mathrm{FWM}}+\left[\frac{\partial\rho_F}{\partial t}\right]_{\mathrm{NLA}} , \quad (3.5)$$

where

$$\left[\frac{\partial\rho_F}{\partial t}\right]_{\mathrm{FWM}}=\frac{N}{\hbar^2}\left[-\beta_s^*\beta_c^*[a^\dagger,b^\dagger\rho_F]\int_0^\infty d\tau\, e^{i(\omega-\omega_c)\tau}\varepsilon_{si}^*\varepsilon_{cj}^*A_{ij}^{++}(\tau)\right.$$
$$-\beta_s^*\beta_c^*[b^\dagger,a^\dagger\rho_F]\int_0^\infty d\tau\, e^{i(\omega-\omega_s)\tau}\varepsilon_{sj}^*\varepsilon_{ci}^*A_{ij}^{++}(\tau)$$
$$+\beta_s^*\beta_c^*[a^\dagger,\rho_F b^\dagger]\int_0^\infty d\tau\, e^{i(\omega-\omega_c)\tau}\varepsilon_{si}^*\varepsilon_{cj}^*A_{ji}^{++}(-\tau)$$
$$\left.+\beta_s^*\beta_c^*[b^\dagger,\rho_F a^\dagger]\int_0^\infty d\tau\, e^{i(\omega-\omega_s)\tau}\varepsilon_{sj}^*\varepsilon_{ci}^*A_{ji}^{++}(-\tau)\right]e^{-it(2\omega-\omega_s-\omega_c)}+\mathrm{H.c.} , \quad (3.6)$$

$$\left[\frac{\partial\rho_F}{\partial t}\right]_{\mathrm{NLA}}=-\frac{N}{\hbar^2}\left[|\beta_s|^2[a^\dagger,a\rho_F]\int_0^\infty d\tau\, e^{-i(\omega-\omega_s)\tau}\varepsilon_{si}^*\varepsilon_{sj}A_{ij}^{+-}(\tau)\right.$$
$$-|\beta_c|^2[b^\dagger,b\rho_F]\int_0^\infty d\tau\, e^{-i(\omega-\omega_c)\tau}\varepsilon_{ci}^*\varepsilon_{cj}A_{ij}^{+-}(\tau)$$
$$+|\beta_s|^2[a^\dagger,\rho_F a]\int_0^\infty d\tau\, e^{-i(\omega-\omega_s)\tau}\varepsilon_{si}^*\varepsilon_{sj}A_{ji}^{-+}(-\tau)$$
$$\left.+|\beta_c|^2[b^\dagger,\rho_F b]\int_0^\infty d\tau\, e^{-i(\omega-\omega_c)\tau}\varepsilon_{ci}^*\varepsilon_{cj}A_{ji}^{-+}(-\tau)+\mathrm{H.c.}\right] , \quad (3.7)$$

where N is the total number of atoms in the sample volume. In deriving (3.6) and (3.7) we have used the property

$$A_{ij}^{++}(-\tau)=[A_{ji}^{--}(\tau)]^* \quad (3.8)$$

of the atomic correlation functions. In the density-matrix equation (3.5) the terms $(\partial\rho_F/\partial t)_{\mathrm{FWM}}$ describe the four-wave-mixing process involving two photons of the pump and one photon from each of the modes a and b. The terms $(\partial\rho_F/\partial t)_{\mathrm{NLA}}$ describe the changes in the characteristics of the field modes a and b due to the linear absorption and emission processes in the presence of a pump wave. Note that these two processes in the semiclassical theory will be described by the four-wave-mixing susceptibilities $\chi^{(3)}(\omega,\omega,-\omega_s)$, $\chi^{(3)}(\omega,\omega,-\omega_c)$, and the saturated absorption susceptibilities. The latter susceptibilities in the simplest situations are related to $\chi^{(3)}(\omega,-\omega,\omega_s)$, $\chi^{(3)}(\omega,-\omega,\omega_c)$. We will see that the quantum theory not only involves such nonlinear susceptibilities but also a set of other characteristics of the atomic system that have no classical counterpart. It should be borne in mind that the steady-state traveling-wave situation is obtained by replacing t by z/c.

In order to understand the various terms in the basic equation (3.5), we transform it in such a way that the symmetrized correlation functions and the correlation functions involving the commutator of the atomic operators at different times appear. Therefore, instead of Eq. (2.19), we introduce the correlation functions

$$C_{\alpha\beta}(\tau)=\lim_{t\to\infty}\langle[G_\alpha(t+\tau),G_\beta(t)]\rangle=-C_{\beta\alpha}(-\tau)\ , \tag{3.9}$$

$$Q_{\alpha\beta}(\tau)=\lim_{t\to\infty}\langle\{G_\alpha(t+\tau)-\langle G_\alpha(t+\tau)\rangle,G_\beta(t)-\langle G_\beta(t)\rangle\}\rangle=Q_{\beta\alpha}(-\tau) \tag{3.10}$$

so that

$$A_{\alpha\beta}(\tau)=[C_{\alpha\beta}(\tau)+Q_{\alpha\beta}(\tau)]/2\ , \tag{3.11}$$

$$A_{\beta\alpha}(-\tau)=[Q_{\alpha\beta}(\tau)-C_{\alpha\beta}(\tau)]/2\ . \tag{3.12}$$

Let us also introduce the notation

$$P^+_{s,c}=\mathbf{P}^+\cdot\boldsymbol{\varepsilon}^*_{s,c},\quad P^-_{s,c}=(P^+_{s,c})^*,$$
$$C^{++}_{s,c}(\tau)=\langle[P^+_s(\tau),P^+_c(0)]\rangle \tag{3.13}$$

and let $\hat{A}_{\alpha\beta}(z)$ be the Laplace transform of $A_{\alpha\beta}(\tau)$

$$\hat{A}_{\alpha\beta}(z)=\int_0^\infty d\tau\, e^{-z\tau}A_{\alpha\beta}(\tau)\ . \tag{3.14}$$

Straightforward but long calculations then show that the Eq. (3.5) can be written as

$$\begin{aligned}\frac{\partial\rho_F}{\partial t}=&-\frac{\beta_s^*\beta_c^*N}{2\hbar^2}(\hat{Q}^{++}_{sc}(-i\nu_c)[a^\dagger,[b^\dagger,\rho_F]]+\hat{C}^{++}_{sc}(-i\nu_c)[a^\dagger,\{b^+,\rho_F\}]+\hat{Q}^{++}_{cs}(-i\nu_s)[b^\dagger,[a^\dagger,\rho_F]]\\&+\hat{C}^{++}_{cs}(-i\nu_s)[b^\dagger,\{a^\dagger,\rho_F\}])e^{-it(2\omega-\omega_s-\omega_c)}\\&-\frac{|\beta_s|^2N}{2\hbar^2}(\hat{Q}^{+-}_{ss}(i\nu_s)[a^\dagger,[a,\rho_F]]+\hat{C}^{+-}_{ss}(i\nu_s)[a^\dagger,\{a,\rho_F\}])\\&-\frac{|\beta_c|^2N}{2\hbar^2}(\hat{Q}^{+-}_{cc}(i\nu_c)[b^\dagger,[b,\rho_F]]+\hat{C}^{+-}_{cc}(i\nu_c)[b^\dagger,\{b,\rho_F\}])+\text{H.c.}\ ,\end{aligned} \tag{3.15}$$

where we have also introduced the detunings of the probe and the generated fields by

$$\nu_c=\omega-\omega_c,\quad \nu_s=\omega-\omega_s\ . \tag{3.16}$$

We show in the next section that the correlation functions C^{++}_{sc}, etc., have a simple interpretation in terms of the nonlinear susceptibilities. The functions Q appear only in the quantum theory and are connected with the fluctuation properties of the atomic system. The functions Q do not appear in the mean values of the field amplitudes. For example, one can show that the mean-value equation for b is

$$\begin{aligned}\langle\dot{b}\rangle=&-\frac{\beta_s^*\beta_c^*N}{\hbar^2}\hat{C}^{++}_{cs}(-i\nu_s)\langle a^\dagger\rangle e^{-it(2\omega-\omega_s-\omega_c)}\\&-\frac{|\beta_c|^2N}{\hbar^2}\hat{C}^{+-}_{cc}(i\nu_c)\langle b\rangle\ .\end{aligned} \tag{3.17}$$

If we compare (3.17) with the corresponding classical equation we see immediately that $\hat{C}^{++}_{cs}(-i\nu_c)$ is related to the four-wave-mixing susceptibility. The function $\hat{C}^{+-}_{cc}(i\nu_c)$ is related to the net absorption and dispersion in the medium at the frequency of the generated wave.

We conclude this section by examining the consequences of dropping the Q terms from the master equation (3.15). This would enable us to compare the results of our theory with those obtained by the quantization of the semiclassical equations of nonlinear mixing. We also ignore the terms involving the correlation functions C^{+-}_{cc} and C^{+-}_{ss}, i.e., we assume that the absorption in the medium can be neglected. On setting $2\omega=\omega_s+\omega_c$, we then obtain

$$\begin{aligned}\frac{\partial\rho_F}{\partial t}=&-\frac{\beta_s^*\beta_c^*N}{2\hbar^2}\{[a^\dagger b^\dagger,\rho][\hat{C}^{++}_{sc}(-i\nu_c)+\hat{C}^{++}_{cs}(-i\nu_s)]\\&+(a^\dagger\rho_F b^\dagger-b^\dagger\rho_F a^\dagger)\\&\times[\hat{C}^{++}_{sc}(-i\nu_c)-\hat{C}^{++}_{cs}(-i\nu_s)]\}\\&+\text{H.c.}\ ,\end{aligned} \tag{3.18}$$

which in the case of degenerate four-wave mixing further reduces to

$$\frac{\partial\rho_F}{\partial t}=-\frac{i}{\hbar}[\kappa a^\dagger b^\dagger,\rho_F]+\text{H.c.}\ , \tag{3.19}$$

$$\kappa=-\frac{\beta_s^*\beta_c^*N}{\hbar}i\hat{C}^{++}_{sc}(0)\ . \tag{3.20}$$

Our analysis thus shows that an effective Hamiltonian

$$H_{\text{eff}}=\kappa a^\dagger b^\dagger+\kappa^* ab \tag{3.21}$$

can be used to describe degenerate four-wave mixing in a nonabsorbing medium, provided we drop all the terms connected with the fluctuations in the medium. Our analysis further shows that an effective Hamiltonian description can be used for the case of nondegenerate four-wave mixing, provided the dispersion of the susceptibilities $\chi^{(3)}(\omega,\omega,-\omega')$ is not important, i.e., $C_{sc}^{++}(-i\nu_c)=C_{cs}^{++}(-i\nu_s)$. Since, in general, both dispersion and absorption effects are important, an effective Hamiltonian description for four-wave mixing is inadequate.

IV. PHYSICAL INTERPRETATION OF THE ATOMIC CORRELATION FUNCTIONS C AND Q

In order to understand the meaning of the quantities C and Q which appear in the quantum theory, we examine the dynamical equation (2.7) with the assumption that the field $\mathbf{E}^+(\mathbf{r},t)$ is a given field with wave vector $\mathbf{q}$ and frequency Ω, i.e.,

$$\mathbf{E}^+(\mathbf{r},t)=\boldsymbol{\varepsilon}e^{i\mathbf{q}\cdot\mathbf{r}-i\Omega t}+\text{H.c.} \tag{4.1}$$

Here ε is a given prescribed quantity. Thus instead of (2.7) we now have

$$\frac{\partial\tilde{\rho}}{\partial t}=-\frac{i}{\hbar}[\tilde{H}_A+\tilde{H}_{AF},\tilde{\rho}]+L_A\tilde{\rho}\ , \tag{4.2}$$

where $\tilde{\rho}$ is the atomic density matrix and

$$\tilde{H}_{AF}=-\int d^3r\ \mathbf{P}^-(\mathbf{r})e^{i\omega t-i\Omega t+i\mathbf{q}\cdot\mathbf{r}}\cdot\boldsymbol{\varepsilon}+\text{H.c.} \tag{4.3}$$

We next investigate the linear response of the system to the applied field ε. Note that before the application of the field ε, the atomic system is in a state $\rho_A^{(0)}$ such that

$$L_A\rho_A^{(0)}-\frac{i}{\hbar}[\tilde{H}_A,\rho_A^{(0)}]=0\ . \tag{4.4}$$

Using (4.2) one can show that the linear response of the dynamical variable G at time t can be written as

$$\begin{aligned}\text{Tr}[\tilde{\rho}(t)G]=&\,e^{i(\omega-\Omega)t}\left[\frac{i}{\hbar}\right]\int d^3r\int_0^\infty d\tau\langle[G(\tau),\mathbf{P}^-(\mathbf{r})\cdot\boldsymbol{\varepsilon}]\rangle e^{i\mathbf{q}\cdot\mathbf{r}-i(\omega-\Omega)\tau}\\&+e^{-i(\omega-\Omega)t}\left[\frac{i}{\hbar}\right]\int d^3r\int_0^\infty d\tau\langle[G(\tau),\mathbf{P}^+(\mathbf{r})\cdot\boldsymbol{\varepsilon}^*]\rangle e^{-i\mathbf{q}\cdot\mathbf{r}+i(\omega-\Omega)\tau}\ .\end{aligned} \tag{4.5}$$

The induced polarization is given by

$$\mathbf{P}(\mathbf{r},t)=e^{-i\omega t}\text{Tr}[\tilde{\rho}\mathbf{P}^+(\mathbf{r})]+\text{H.c.}\ , \tag{4.6}$$

which on using the result (4.5) becomes

$$\begin{aligned}\mathbf{P}(\mathbf{r},t)=&\,e^{-i\Omega t}\left[\frac{i}{\hbar}\right]\int_0^\infty d\tau\int d^3r_1\langle[\mathbf{P}^+(\mathbf{r},\tau),\mathbf{P}^-(\mathbf{r}_1)\cdot\boldsymbol{\varepsilon}]\rangle e^{i\mathbf{q}\cdot\mathbf{r}_1}e^{-i(\omega-\Omega)\tau}\\&+e^{-i(2\omega-\Omega)t}\left[\frac{i}{\hbar}\right]\int_0^\infty d\tau\int d^3r_1\langle[\mathbf{P}^+(\mathbf{r},\tau),\mathbf{P}^+(\mathbf{r}_1)\cdot\boldsymbol{\varepsilon}^*]\rangle e^{-i\mathbf{q}\cdot\mathbf{r}_1}e^{i(\omega-\Omega)\tau}+\text{H.c.}\end{aligned} \tag{4.7}$$

If the different atoms are uncorrelated than (4.7) can be simplified by using Eq. (3.2) and the definition (3.9) with the result

$$\begin{aligned}\mathbf{P}_i(\mathbf{r},t)=&\,e^{-i\Omega t+i\mathbf{q}\cdot\mathbf{r}}\left[\frac{i}{\hbar}\right]\hat{C}_{ij}^{+-}(i(\omega-\Omega))n\varepsilon_j\\&+e^{-i(2\omega-\Omega)t+i(2\mathbf{k}-\mathbf{q})\cdot\mathbf{r}}n\left[\frac{i}{\hbar}\right]\\&\times\hat{C}_{ij}^{++}(-i(\omega-\Omega))\varepsilon_j^*+\text{H.c.}\ ,\end{aligned} \tag{4.8}$$

where n is the density of atoms. Note that the first term in (4.8) gives the response of the system at the applied frequency Ω. Thus if $\chi_{ij}(\Omega)$ is the susceptibility of the system, then we can identify

$$\chi_{ij}(\Omega)=\frac{in}{\hbar}\hat{C}_{ij}^{+-}(i(\omega-\Omega))\ . \tag{4.9}$$

It should be borne in mind that $\chi_{ij}(\Omega)$ depends on all the powers of the pump field of frequency ω. If the pump field is zero, then $\chi_{ij}(\Omega)$ reduces to the usual linear susceptibility of the system. In the language of conventional nonlinear optical susceptibilities, the intensity-dependent χ has the expansion

$$\begin{aligned}\chi_{ij}(\Omega)=&\,\chi_{ij}^{(1)}(\Omega)\\&+\sum_{n=1}^{\infty}\chi_{i\alpha_1\beta_1\cdots\alpha_n\beta_nj}^{(2n+1)}(\Omega,\omega,-\omega,\ldots,\omega,-\omega)\\&\times\prod_n\varepsilon_{\alpha_n}(\omega)\varepsilon_{\beta_n}^*(\omega)\ .\end{aligned} \tag{4.10}$$

The second term in (4.8) gives the induced polarization at the frequency $2\omega-\Omega$. Such an induced polarization has the form of a plane wave in the direction $2\mathbf{k}-\mathbf{q}$. Thus this term corresponds to four-wave mixing. If the correlation function $\hat{C}_{ij}^{++}(-i(\omega-\Omega))$ is computed to second order in the pump field ε_p, then we can obviously identify

$$(\varepsilon_p)_\alpha(\varepsilon_p)_\beta\chi_{i\alpha\beta j}^{(3)}(\omega,\omega,-\Omega)=\frac{i}{\hbar}\hat{C}_{ij}^{++}(-i(\omega-\Omega))n\ . \tag{4.11}$$

In the more general case the four-wave-mixing susceptibility depends on all the powers of the pump field and hence can be written in a form analogous to (4.10):

$$\frac{i}{\hbar}n\hat{C}_{ij}^{++}(-i(\omega-\Omega))=\sum_{n=0}^{\infty}\chi^{(2n+3)}_{i\alpha\beta\alpha_1\beta_1\cdots\alpha_n\beta_n j}(\omega,\omega,-\Omega;\omega,-\omega,\ldots\omega,-\omega)\varepsilon_\alpha(\omega)\varepsilon_\beta(\omega)\prod_n\epsilon_{\alpha_n}(\omega)\epsilon^*_{\beta_n}(\omega)\ . \tag{4.12}$$

We have thus established the relation of all the atomic correlation functions involving the commutator of the atomic operators to the intensity-dependent susceptibilities of the atomic system.

The quantities Q are more difficult to interpret. However, it is well known that the inelastic scattering from a quantum-mechanical system driven by an external field is determined by the correlation functions of the form $\langle P^{(-)}(\mathbf{r},\tau)P^+(\mathbf{r},0)\rangle - \langle P^-(\mathbf{r},\tau)\rangle\langle P^+(\mathbf{r},0)\rangle$. Atomic correlations of the form $\langle P^{(-)}(\mathbf{r},\tau)P^-(\mathbf{r},0)\rangle$ determine the anomalous correlation functions[20] of the field radiated by an atomic system. Thus the quantum-mechanical character of the field radiated by a system driven by a field of frequency ω is determined in terms of the atomic correlation functions Q. This can also be appreciated by looking at the short-time solution of (3.15) for an initial vacuum state of the field

$$\begin{aligned}\frac{\partial\rho_F}{\partial t}=&-\frac{\beta_s^*\beta_c^*N}{2\hbar^2}[\hat{Q}_{sc}^{++}(-i\nu_c)+\hat{Q}_{cs}^{++}(-i\nu_s)+\hat{C}_{sc}^{++}(-i\nu_c)+\hat{C}_{cs}^{++}(-i\nu_s)]\,|1,1\rangle\langle 0,0|\\ &+\frac{|\beta_s|^2N}{2\hbar^2}[\hat{Q}_{ss}^{+-}(i\nu_s)-\hat{C}_{ss}^{+-}(i\nu s)](\,|1,0\rangle\langle 1,0|-|0,0\rangle\langle 0,0|\,)\\ &+\frac{|\beta_c|^2N}{2\hbar^2}[\hat{Q}_{cc}^{+-}(i\nu_c)-\hat{C}_{cc}^{+-}(i\nu_c)](\,|0,1\rangle\langle 0,1|-|0,0\rangle\langle 0,0|\,)+\text{H.c.}\end{aligned} \tag{4.13}$$

Thus for short times, the probability that a photon of the mode a grows from vacuum is related to the real part of $\hat{Q}_{ss}^{+-}(i\nu_s)-\hat{C}_{ss}^{+-}(i\nu_s)$. The off-diagonal elements of the field depend on the correlations like $\hat{Q}_{sc}^{++}(-i\nu_c)$. Equation (4.13) leads to nonvanishing of the expectation values like $\langle ab\rangle$. Such nonvanishing expectation values in turn lead to the simultaneous production of photons in modes a and b.

V. SOLUTION OF THE MASTER EQUATION (3.15)

In this section we present the general structure of the solution of (3.15). It proves convenient to work with the Wigner distribution function $\phi(z_a,z_b)$ associated with the density matrix ρ_F. Formally the Wigner distribution function[14] is defined by

$$\phi(z_a,z_b)=\frac{1}{\pi^4}\text{Tr}\left\{\rho_F\int\int d^2\alpha\, d^2\beta\exp\{-[\alpha(z_a^*-a^\dagger)-\alpha^*(z_a-a)+\beta(z_b^*-b^\dagger)-\beta^*(z_b-b)]\}\right\}\ , \tag{5.1}$$

where $\int d^2\alpha$ stands for the integration over the whole complex α plane. The basic equation (3.15) can be transformed into an equation for ϕ by using the following rules of mapping[21] associated with Weyl ordering:

$$\begin{aligned}&[a^\dagger,\rho]\to-\frac{\partial\phi}{\partial z_a},\ [a,\rho]\to\frac{\partial\phi}{\partial z_a^*}\ ,\\ &\{a,\rho\}\to 2z_a\phi,\ \{a^\dagger,\rho\}\to 2z_a^*\phi\ ,\\ &[a^\dagger,[b^\dagger,\rho]]\to\frac{\partial^2\phi}{\partial z_a\partial z_b},\ [a^\dagger,\{a,\rho\}]\to-\frac{\partial}{\partial z_a}(2z_a\phi)\ldots\ .\end{aligned} \tag{5.2}$$

The equation for ϕ is then found to be

$$\begin{aligned}\frac{\partial\phi}{\partial t}=&-\frac{\beta_s^*\beta_c^*N}{2\hbar^2}\left[[\hat{Q}_{sc}^{++}(-i\nu_c)+\hat{Q}_{cs}^{++}(-i\nu_s)]\frac{\partial^2\phi}{\partial z_a\partial z_b}\right.\\ &\left.-\hat{C}_{sc}^{++}(-i\nu_c)\frac{\partial}{\partial z_a}(2z_b^*\phi)-\hat{C}_{cs}^{++}(-i\nu_s)\frac{\partial}{\partial z_b}(2z_a^*\phi)\right]e^{-it(2\omega-\omega_s-\omega_c)}\\ &+\frac{|\beta_s|^2N}{2\hbar^2}\hat{Q}_{ss}^{+-}(i\nu_s)\frac{\partial^2\phi}{\partial z_a\partial z_a^*}+\frac{|\beta_c|^2N}{2\hbar^2}\hat{Q}_{cc}^{+-}(i\nu_c)\frac{\partial^2\phi}{\partial z_b\partial z_b^*}\\ &+\frac{|\beta_s|^2N}{2\hbar^2}\hat{C}_{ss}^{+-}(i\nu_s)\frac{\partial}{\partial z_a}(2z_a\phi)+\frac{|\beta_c|^2N}{2\hbar^2}\hat{C}_{cc}^{+-}(i\nu_c)\frac{\partial}{\partial z_b}(2z_b\phi)+\text{c.c.}\end{aligned} \tag{5.3}$$

The equation for the Wigner distribution function has the form of a linearized Fokker-Planck equation[22] in four variables. The diffusion terms in this Fokker-Planck equation arise from the nonvanishing of the symmetrized atomic correlation functions Q. The drift terms are related to the nonlinear susceptibilities. It should be borne in mind that $\hat{C}_{sc}^{++}(-i\nu_c)\neq\hat{C}_{cs}^{++}(-i\nu_s)$ for the case of nondegenerate four-wave mixing. Equation (5.3) clearly shows that the quantization of the semiclassical equations is not adequate, for if one had used the semiclassical equations and quantized them, then all the second derivative terms in (5.3) would have been missed. It should be remembered that the Wigner function enables one to calculate the symmetrized expectation values; for example,

$$\left\langle\frac{a^\dagger a+aa^\dagger}{2}\right\rangle=\int\int d^2z_a d^2z_b\,|z_a|^2\phi(z_a,z_b)\ . \tag{5.4}$$

If $\phi_0(z_a,z_b)$ is the Wigner function at time $t=0$, then the general solution of (5.3) can be written as

$$\phi(z_a,z_b,t)=\int d^2z_a'\int d^2z_b'\phi_0(z_a',z_b')K(z_a,z_b,t;z_a',z_b')\ , \tag{5.5}$$

where K is the conditional distribution function associated with the Fokker-Planck equation (5.3). It is the solution of (5.3) subject to the initial condition $\delta^{(2)}(z_a-z_a')\delta^{(2)}(z_b-z_b')$. The conditional distribution associated with a linearized Fokker-Planck equation is well known. It is Gaussian in four real variables. It is centered at the solution of the equations

$$\begin{aligned}\dot{Z}_a&=-\beta_s^*\beta_c^*\frac{N}{\hbar^2}\hat{C}_{sc}^{++}(-i\nu_c)Z_b^*-\frac{|\beta_s|^2N}{\hbar^2}\hat{C}_{ss}^{+-}(i\nu_s)Z_a\ ,\\ \dot{Z}_b&=-\frac{\beta_s^*\beta_c^*N}{\hbar^2}\hat{C}_{cs}^{++}(-i\nu_s)Z_a^*-\frac{|\beta_c|^2N}{\hbar^2}\hat{C}_{cc}^{+-}(i\nu_c)Z_b\ ,\end{aligned} \tag{5.6}$$

subject to the initial values Z_a',Z_b'. If initially each mode is in a coherent state $|z_a\rangle|z_b\rangle$, then

$$\phi_0(Z_a,Z_b)=\frac{4}{\pi^2}\exp(-2|Z_a-z_a|^2-2|Z_b-z_b|^2)\ . \tag{5.7}$$

Using (5.7) and the Gaussian nature of the conditional distribution, it follows that $\phi(Z_a,Z_b,t)$ is also Gaussian. Since any Gaussian distribution is completely characterized by the mean values of the variables Z and the fluctuations in them, it is sufficient to solve mean value equations like (5.6) and the equations for quantities like $\langle Z_aZ_b\rangle$. Let ψ ($\psi^\dagger$) be the column (row) matrix with components Z_a and Z_b^* (Z_a^* and Z_b). Then from (5.3) it follows that

$$\frac{d}{dt}\langle\psi\rangle=A\langle\psi\rangle,\quad A=-\frac{N}{\hbar^2}\begin{bmatrix}|\beta_s|^2\hat{C}_{ss}^{+-}(i\nu_s) & \beta_s^*\beta_c^*\hat{C}_{sc}^{++}(-i\nu_c)\\ \beta_s\beta_c(\hat{C}_{cs}^{++})^*(-i\nu_s) & |\beta_c|^2[\hat{C}_{cc}^{+-}(i\nu_c)]^*\end{bmatrix} \tag{5.8}$$

$$\frac{d}{dt}\langle\psi\psi^\dagger\rangle=A\langle\psi\psi^\dagger\rangle+\langle\psi\psi^\dagger\rangle A^\dagger+2D\ ,$$

$$2D=\frac{N}{2\hbar^2}\begin{bmatrix}|\beta_s|^2[\hat{Q}_{ss}^{+-}(i\nu_s)+\text{c.c.}] & -\beta_s^*\beta_c^*[\hat{Q}_{sc}^{++}(-i\nu_c)+\hat{Q}_{cs}^{++}(-i\nu_s)]\\ -\beta_s\beta_c\{\hat{Q}_{sc}^{++}(-i\nu_c)+[\hat{Q}_{cs}^{++}(-i\nu_s)]\}^* & |\beta_c|^2[\hat{Q}_{cc}^{+-}(i\nu_c)+\text{c.c.}]\end{bmatrix}\ . \tag{5.9}$$

These are to be solved subject to the initial conditions obtained from equations like

$$\langle\psi_1\rangle=\langle Z_a\rangle=\langle a\rangle,\quad\langle\psi_1\psi_1^*\rangle=\langle Z_aZ_a^*\rangle=\left\langle\frac{aa^\dagger+a^\dagger a}{2}\right\rangle\ ,$$

$$\langle\psi\rangle=\begin{bmatrix}z_a\\ z_b^*\end{bmatrix},\quad\langle\psi\psi^\dagger\rangle_0=\begin{bmatrix}|z_a|^2+\frac{1}{2} & z_az_b\\ z_b^*z_b^* & |z_b|^2+\frac{1}{2}\end{bmatrix}\ . \tag{5.10}$$

Note that the solution of (5.9) can also be written as

$$\langle\psi\psi^\dagger\rangle=e^{At}\langle\psi\psi^\dagger\rangle_0(e^{At})^\dagger+2\int_0^t d\tau\, e^{A\tau}D(e^{A\tau})^\dagger \tag{5.11}$$

$$\equiv\begin{bmatrix}\langle a^\dagger a\rangle+\frac{1}{2} & \langle ab\rangle\\ \langle a^\dagger b^\dagger\rangle & \langle b^\dagger b\rangle+\frac{1}{2}\end{bmatrix}\ . \tag{5.12}$$

Thus the Gaussian property of ϕ and the matrix e^{At} and (5.11) completely characterize the time development of the density matrix. Explicit results can be obtained by considering specific examples of the media which determine the form of the correlation functions Q and C. Numerical results on the photon statistics will be presented in a future paper.

If we define $\delta\psi$ as the fluctuation of ψ from its mean value

$$\delta\psi=\psi-\langle\psi\rangle\ , \tag{5.13}$$

then it follows from (5.10) and (5.11) that

$$\langle\delta\psi\,\delta\psi^\dagger\rangle=\tfrac{1}{2}e^{At}(e^{At})^\dagger+2\int_0^t d\tau\, e^{A\tau}D(e^{A\tau})^\dagger\ . \tag{5.14}$$

Equation (5.14) shows that the field fluctuations at time t are determined by (i) the growth of the initial quantum fluctuations of the field, and (ii) the quantum-mechanical fluctuations associated with the medium (D terms).

Finally, note that in the context of squeezing one needs to know the fluctuations in the components like $(a+a^\dagger)$. These can be directly obtained from the Wigner function. For example let A be the operator

$$A=\mu a+\nu b,\quad A^{\dagger}=\mu^{*}a^{\dagger}+\nu^{*}b^{\dagger}\,,$$
$$[A,A^{\dagger}]=1,\quad |\mu|^{2}+|\nu|^{2}=1\,. \tag{5.15}$$

The moments of A can be written in terms of the Wigner function as

$$\langle (A+A^{\dagger})^{n}\rangle=\int\int d^{2}Z_{a}d^{2}Z_{b}\phi(Z_{a},Z_{b})\times(\mu Z_{a}+\nu Z_{b}+\text{c.c.})^{n} \tag{5.16}$$

and hence

$$\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{n}\rangle=\int\int d^{2}Z_{a}d^{2}Z_{b}\phi(Z_{a},Z_{b})(\mu Z_{a}+\nu Z_{b}-\mu\langle Z_{a}\rangle-\nu\langle Z_{b}\rangle+\text{c.c.})^{n}\,. \tag{5.17}$$

Using our earlier result that ϕ at time t is Gaussian if initially the field modes were in coherent states, we can reduce (5.17) by using the moment theorem for Gaussian distributions. We then obtain

$$\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2n}\rangle=\frac{2n!}{2^{n}n!}\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2}\rangle^{n}\,. \tag{5.18}$$

The result (5.18) can be used in the considerations of higher-order squeezing. If a field is in a coherent state, then the $2n$th-order moment also obeys the property (5.18)

$$\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2n}\rangle_{\text{coh}}=\frac{2n!}{2^{n}n!}\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2}\rangle^{n}_{\text{coh}}\,. \tag{5.19}$$

Hence, if

$$\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2}\rangle<\langle (A+A^{+}-\langle A+A^{\dagger}\rangle)^{2}\rangle_{\text{coh}}\,, \tag{5.20}$$

then

$$\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2n}\rangle<\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2n}\rangle_{\text{coh}}\,. \tag{5.21}$$

It follows from the above that if the variable $(A+A^{\dagger})$ is squeezed to second order, then it remains squeezed to all even orders. We have thus proved a general result in the quantum theory of four-wave mixing. The second-order squeezing can be studied using the result (5.14) and the easily proved relation

$$\begin{aligned}\langle (A+A^{\dagger}-\langle A+A^{\dagger}\rangle)^{2}\rangle&=2|\mu|^{2}\langle\delta\psi_{1}\delta\psi_{1}^{*}\rangle\\&+2|\nu|^{2}\langle\delta\psi_{2}\delta\psi_{2}^{*}\rangle\\&+2\mu\nu\langle\delta\psi_{1}\delta\psi_{2}^{*}\rangle\\&+2\mu^{*}\nu^{*}\langle\delta\psi_{2}\delta\psi_{1}^{*}\rangle\,.\end{aligned} \tag{5.22}$$

VI. SPECIFIC EXAMPLES

The general theory given in Secs. II and III is valid for a wide class of systems in which nonlinear mixing can be studied. Different systems will lead to different coefficients in the dynamical equation (3.15) as the correlation functions C and Q will depend on the nature of the medium, the nature of the optical transitions, and the characteristics of the pump field. These correlation functions can be explicitly evaluated in special cases and we now consider several important ones.

A. Two-level optical transitions

Let us consider the quantum theory of four-wave mixing in a medium of two-level atoms with frequency separation ω_0. This is the case most extensively studied[2,11–13] both theoretically and experimentally. The polarization operator, for an atom located at the position $\mathbf{R}$, can be written as

$$\mathbf{P}(\mathbf{r})=\mathbf{d}S^{+}\delta(\mathbf{r}-\mathbf{R})+\text{H.c.}\,, \tag{6.1}$$

where S^{+}, its adjoint S^{-}, and $S^{z}=\frac{1}{2}[S^{+},S^{-}]$ satisfy the commutation relations for spin-$\frac{1}{2}$ systems. We do the calculations in a frame rotating with the frequency ω of the pump field. The dynamical equation (2.9) for the atomic system can be written in the matrix form as

$$\frac{\partial\psi}{\partial t}=A\psi+I,\quad \psi_{1}=\langle S^{+}\rangle e^{i\mathbf{k}\cdot\mathbf{R}-i\omega t},\quad \psi_{2}=\psi_{1}^{*}\,,$$
$$\psi_{3}=\langle S^{z}\rangle,\quad I_{1}=I_{2}=0,\quad I_{3}=\eta/2T_{1}\,, \tag{6.2}$$
$$A=\begin{bmatrix}-\frac{1}{T_{2}}+i\Delta & 0 & 2ig^{*}\\ 0 & -\frac{1}{T_{2}}-i\Delta & -2ig\\ ig & -ig^{*} & -\frac{1}{T_{1}}\end{bmatrix},$$

$$\Delta=\omega_{0}-\omega,\quad g=\frac{\mathbf{d}\cdot\boldsymbol{\varepsilon}}{\hbar}\,. \tag{6.3}$$

Here T_1 and T_2 are the longitudinal and transverse relaxation times and η gives the equilibrium value of the atomic inversion in the absence of the pump. By using the quantum regression theorem all atomic correlation functions can be computed in terms of the solution of (6.2). We give results for various correlation functions in terms of the steady-state solution

$$\psi=-A^{-1}I \tag{6.4}$$

and the elements of the matrix U defined by

$$U(z)=(z-A)^{-1}\,. \tag{6.5}$$

Our calculations show that

$$\hat{Q}_{sc}^{++}(z)=(\boldsymbol{\epsilon}_s^*\cdot\mathbf{d})^*(\boldsymbol{\epsilon}_c\cdot\mathbf{d})^*\sum_l U_{2l}(z)(1-2\psi_1\psi_2,-2\psi_2\psi_2,-2\psi_2\psi_3)_l\,, \tag{6.6}$$

$$\hat{C}_{sc}^{++}(z)=(\boldsymbol{\epsilon}_s\cdot\mathbf{d})^*(\boldsymbol{\epsilon}_c\cdot\mathbf{d})^*\sum_l U_{2l}(z)(2\psi_3,0,-\psi_2)_l\,, \tag{6.7}$$

$$\hat{Q}_{ss}^{+-}(z)=|\boldsymbol{\epsilon}_s\cdot\mathbf{d}|^2\sum_l U_{2l}(z)(-2\psi_1\psi_1,1-2\psi_1\psi_2,-2\psi_1\psi_3)_l\,, \tag{6.8}$$

$$\hat{C}_{ss}^{+-}(z)=|\boldsymbol{\epsilon}_s\cdot\mathbf{d}|^2\sum_l U_{2l}(z)(0,-2\psi_3,\psi_1)_l\,. \tag{6.9}$$

Thus all the coefficients needed for the quantum theory of four-wave mixing in a two-level medium can be explicitly obtained.

For many practical purposes[8] it is sufficient to do a third-order calculation in the applied (pump and probe) fields. Hence, we give explicit results for the atomic correlation functions to second order in pump fields:

$$\begin{bmatrix}\hat{Q}_{sc}^{++}(z)\\ \hat{C}_{sc}^{++}(z)\end{bmatrix}=\frac{2(\boldsymbol{\epsilon}_s\cdot\mathbf{d})^*(\boldsymbol{\epsilon}_c\cdot\mathbf{d})^*g^2}{(z+i\Delta+1/T_2)}\left[\frac{\eta^2}{(1/T_2+i\Delta)^2}\begin{bmatrix}1\\0\end{bmatrix}+\frac{1}{(z+1/T_1)(z+1/T_2-i\Delta)}\begin{bmatrix}1\\ \eta\end{bmatrix}+\frac{1}{(z+1/T_1)(1/T_2+i\Delta)}\begin{bmatrix}\eta^2\\ \eta\end{bmatrix}\right], \tag{6.10}$$

$$\begin{bmatrix}\hat{Q}_{ss}^{+-}(z)\\ \hat{C}_{ss}^{+-}(z)\end{bmatrix}=\frac{|\boldsymbol{\epsilon}_s\cdot\mathbf{d}|^2}{(z+1/T_2+i\Delta)}\left\{\begin{bmatrix}1-\dfrac{2\eta^2|g|^2}{(1/T_2)^2+\Delta^2}\\ -\eta\left[1-\dfrac{4|g|^2T_1}{T_2[\Delta^2+(1/T_2)^2]}\right]\end{bmatrix}-\frac{2|g|^2}{(z+1/T_1)}\left[(z+1/T_2+i\Delta)^{-1}\begin{bmatrix}1\\-\eta\end{bmatrix}+(1/T_2-i\Delta)^{-1}\begin{bmatrix}\eta^2\\-\eta\end{bmatrix}\right]\right\}. \tag{6.11}$$

These expressions simplify considerably for degenerate four-wave mixing in a medium which only has radiative relaxation ($\eta=-1$, $1/T_1=2/T_2$)

$$\begin{bmatrix}\hat{Q}_{sc}^{++}(0)\\ \hat{C}_{sc}^{++}(0)\end{bmatrix}=\frac{2g^2(\boldsymbol{\epsilon}_s\cdot\mathbf{d})^*(\boldsymbol{\epsilon}_c\cdot\mathbf{d})^*}{[(1/T_2)^2+\Delta^2](1/T_2+i\Delta)}\times\begin{bmatrix}\dfrac{2}{T_2(i\Delta+1/T_2)}\\ -1\end{bmatrix}, \tag{6.12}$$

$$\begin{bmatrix}\hat{Q}_{ss}^{+-}(0)\\ \hat{C}_{ss}^{+-}(0)\end{bmatrix}=\frac{|\boldsymbol{\epsilon}_s\cdot\mathbf{d}|^2}{(1/T_2+i\Delta)}\left[1-\frac{4|g|^2}{(1/T_2)^2+\Delta^2}\right]\begin{bmatrix}1\\1\end{bmatrix}. \tag{6.13}$$

Thus for large detunings $\Delta T_2\gg1$, the fluctuation Q_{sc}^{++} can be ignored. Moreover, real $\hat{Q}_{ss}^{+-}(0)\sim O(1/\Delta T_2)^2$ and hence can also be ignored. In such a case the diffusion [Eq. (5.9)] is negligible and the field fluctuations are given by the first term in the solution (5.14).

B. Three-level optical transitions—V system

There exists considerable work on four-wave mixing in a medium in which optical transitions can be modeled as three-level transitions.[3,4,6] The semiclassical theory is in terms of the nonlinear susceptibilities, whereas the quantum theory requires the additional knowledge[23] of the correlation functions $Q^{\pm\pm}$, etc. In the following we treat the important case of the V system.[4] The Λ system is very similar. The ladder system is discussed in Sec. VII in connection with two-photon media.

Let us consider the transitions $|j=0,m=0\rangle\leftrightarrow|j=1,m=\pm1\rangle$ in a three-level system due to the interaction with a linearly polarized pump field. The linearly polarized pump field can be decomposed into right and left circularly polarized components

$$\mathbf{E}(t)=(\tfrac{1}{2}\boldsymbol{\epsilon}_R E_R+\tfrac{1}{2}\boldsymbol{\epsilon}_L E_L)e^{i\mathbf{k}\cdot\mathbf{R}-i\omega t}+\text{c.c.} \tag{6.14}$$

We further assume that the signal wave is a right circularly polarized wave so that $\boldsymbol{\epsilon}_s=\boldsymbol{\epsilon}_R$. In such a case the generated wave is left circularly polarized, i.e., $\boldsymbol{\epsilon}_c=\boldsymbol{\epsilon}_L$. We have taken the strengths of the left and right circularly polarized components to be different so that the case of light with arbitrary polarization can be handled. The dipole moment operator for the atomic system can be expressed as

$$\mathbf{d}=d_{13}\boldsymbol{\epsilon}_R|1\rangle\langle3|+d_{23}\boldsymbol{\epsilon}_L|2\rangle\langle3|+\text{H.c.}\,, \tag{6.15}$$

where the states are labeled as $|1\rangle$, $|2\rangle$, and $|3\rangle$ i.e., $|1\rangle\equiv|j=1,m=1\rangle$, $|2\rangle\equiv|j=1,m=-1\rangle$, $|3\rangle\equiv|j=0,m=0\rangle$. Let E_1 and E_2 be the energies of the states $|1\rangle$ and $|2\rangle$ relative to the state $|3\rangle$. The density-matrix equation for the three-level system interacting with the pump field (6.14) can be written in the matrix from as[4]

$$\dot{\psi}=A\psi+I,\quad \psi_1=\rho_{11},\quad \psi_2=\rho_{12},\quad \psi_3=\rho_{21},\quad \psi_4=\rho_{13}e^{i(\omega t-\mathbf{k}\cdot\mathbf{R})},\quad \psi_5=\psi_4^*,\quad \psi_6=\rho_{22},\psi_7=\rho_{23}e^{i(\omega t-\mathbf{k}\cdot\mathbf{R})},\quad \psi_8=\psi_7^*\ ,$$

$$A=\begin{bmatrix} -2\gamma_1 & 0 & 0 & i\,|g_L| & -i\,|g_L| & 0 & 0 & 0 \\ 0 & -(\Gamma_{12}+2i\delta) & 0 & i\,|g_R| & 0 & 0 & 0 & -i\,|g_L| \\ 0 & 0 & -(\Gamma_{12}-2i\delta) & 0 & -i\,|g_R| & 0 & i\,|g_L| & 0 \\ 2i\,|g_L| & i\,|g_R| & 0 & -(\Gamma_{13}+i\delta+i\Delta) & 0 & i\,|g_L| & 0 & 0 \\ -2i\,|g_L| & 0 & -i\,|g_R| & 0 & -(\Gamma_{13}-i\delta-i\Delta) & -i\,|g_L| & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -2\gamma_2 & i\,|g_R| & -i\,|g_R| \\ i\,|g_R| & 0 & i\,|g_L| & 0 & 0 & 2i\,|g_R| & -(\Gamma_{23}-i\delta+i\Delta) & 0 \\ -i\,|g_R| & -i\,|g_L| & 0 & 0 & 0 & -2i\,|g_R| & 0 & -(\Gamma_{23}+i\delta-i\Delta) \end{bmatrix},$$

$$I=\begin{bmatrix} 0 \\ 0 \\ 0 \\ -i\,|g_L| \\ +i\,|g_L| \\ 0 \\ -i\,|g_R| \\ +i\,|g_R| \end{bmatrix},\quad \delta=(E_1-E_2)/2\hbar,\quad \Delta=\omega_0-\omega,\quad \omega_0=\frac{E_1+E_2}{2\hbar},\quad g_R=-\frac{\mathbf{d}_{23}\cdot\mathbf{E}_R}{\hbar},\quad g_L=-\frac{\mathbf{d}_{13}\cdot\mathbf{E}_2}{\hbar}\ . \tag{6.16}$$

In these equations Γ_{ij} is the relaxation rate of the off-diagonal element ρ_{ij}. These relaxation rates include contributions from phase-changing collisions. The rate of radiative decay of the state $|1\rangle$ ($|2\rangle$) is taken as $2\gamma_1$ ($2\gamma_2$). Note that 2δ gives the Zeeman splitting between the levels $|1\rangle$ and $|2\rangle$ and ω_0 gives the position of levels $|1\rangle$ and $|2\rangle$ in the absence of the magnetic field. The polarization operator for the V system is

$$\mathbf{P}^{(+)}=d_{13}^*\boldsymbol{\varepsilon}_R^*\,|3\rangle\langle 1|+d_{23}^*\boldsymbol{\varepsilon}_L^*\,|3\rangle\langle 2| \tag{6.17}$$

and hence

$$P_s^{(+)}=\mathbf{P}^{(+)}\cdot\boldsymbol{\varepsilon}_R^*=d_{23}^*\,|3\rangle\langle 2|\ ,$$
$$P_c^{(+)}=\mathbf{P}^{+}\cdot\boldsymbol{\varepsilon}_L^*=d_{13}^*\,|3\rangle\langle 1|\ . \tag{6.18}$$

All the correlation functions relevant for the quantum theory of four-wave mixing in a V system can be calculated using Eq. (6.16) and the quantum regression theorem. We quote the results. Let ψ be the steady-state solution of (6.16), i.e.,

$$\psi=-A^{-1}I\ , \tag{6.19}$$

and let $U(z)$ be the matrix (6.5) with A now given by (6.16). Then our calculations lead to the results

$$\begin{bmatrix} \hat{Q}_{sc}^{++}(z) \\ \hat{C}_{sc}^{++}(z) \end{bmatrix}=d_{23}^*d_{13}^*\sum_l U_{7l}(z)[(\pm\psi_4,0,\pm\psi_7,0,\psi_1\pm(1-\psi_1-\psi_6),0,0,\psi_2)_l-2q\psi_l\psi_4]\ , \tag{6.20}$$

$$\begin{bmatrix} \hat{Q}_{ss}^{+-}(z) \\ \hat{C}_{ss}^{+-}(z) \end{bmatrix}=d_{23}^*d_{23}\sum_l U_{7l}(z)[(0,0,\psi_5,\pm\psi_2,0,\psi_8,(1-\psi_1-\psi_6)\pm\psi_6,0)_l-2q\psi_l\psi_8]\ , \tag{6.21}$$

$$\begin{bmatrix} \hat{Q}_{cs}^{++}(z) \\ \hat{C}_{cs}^{++}(z) \end{bmatrix}=d_{23}^*d_{13}^*\sum_l U_{4l}(z)[(0,\pm\psi_4,0,0,\psi_3,\pm\psi_7,0,\psi_6\pm(1-\psi_1-\psi_6))_l-2q\psi_l\psi_7]\ , \tag{6.22}$$

$$\begin{bmatrix} \hat{Q}_{cc}^{+-}(z) \\ \hat{C}_{cc}^{+-}(z) \end{bmatrix}=|d_{13}|^2\sum_l U_{4l}(z)[(\psi_5,\psi_8,0,1-\psi_1-\psi_6\pm\psi_1,0,0,\pm\psi_3,0)_l-2q\psi_l\psi_5]\ . \tag{6.23}$$

Here the parameter q is to be set zero (one) for the correlation functions C (Q). Moreover, in Eqs. (6.20)–(6.23) the upper (lower) signs are to be taken in calculating the functions Q (C).

With the knowledge of the correlation functions (6.20)–(6.23), the quantum theory of four-wave mixing in V systems is complete. The correlation functions for the quantum theory of four-wave mixing in the Λ system can be evaluated similarly. Numerical results in these two cases will be discussed in a future paper.

C. Four-wave mixing in an optical fiber

Recently, there has been considerable interest in the production of squeezed states of the radiation field using four-wave mixing in an optical fiber.[10,12] The study of squeezing requires the quantum theory of four-wave mixing in an optical fiber. We now show how the general theory of Secs. II and III can be applied to four-wave mixing in optical fibers. We need to have a quantum theoretical description of the optical fiber. These can be modeled in terms of a set of anharmonic oscillators. The equation (2.9) for the optical fiber will have the form

$$\frac{\partial\rho}{\partial t}=-i[\Delta c^{\dagger}c+\chi(c^{\dagger})^2c^2-c^{\dagger}g-cg^*,\rho]-\kappa(c^{\dagger}c\rho-2c\rho c^{\dagger}+\rho c^{\dagger}c)\,, \tag{6.24}$$

$$\Delta=\omega_0-\omega,\quad g=\frac{d\cdot\varepsilon}{\hbar}e^{i\mathbf{k}\cdot\mathbf{R}}\,.$$

The operators c and $c^{\dagger}$ are the annihilation and creation operators for the anharmonic oscillator. The anharmonicity is represented by χ. The nonlinear response of the fiber arises from the nonvanishing of χ. The coupling with the pump field is represented by g. The terms involving κ describe the linear losses in the fiber.

The steady-state response can be computed to various orders in χ; for example, the mean oscillator motion will be

$$\langle c\rangle=\frac{ig}{(\kappa+i\Delta)}+\frac{2g\,|g|^2\chi}{(\kappa^2+\Delta^2)(\kappa+i\Delta)^2}+O(\chi^2)\,. \tag{6.25}$$

The second term in (6.25) yields the third-order susceptibility of the anharmonic oscillator driven by a pump of frequency ω. The correlation functions of c operators can be calculated to various orders in χ. For this purpose we need the time-dependent solution of (6.24) in the absence of the nonlinearity. Such a solution can be obtained by using coherent-state techniques and is well discussed in the literature.[19,24] If the oscillator at time $t=0$ is in a coherent state $|z_0\rangle$, then the time-dependent solution of (6.24), for the case $\chi=0$, is

$$\rho(t)=|z(t)\rangle\langle z(t)|\,, \tag{6.26}$$

where $|z(t)\rangle$ is a coherent state with amplitude $z(t)$ given by

$$z(t)=z_0e^{-i\Delta t-\kappa t}+\frac{ig}{(\kappa+i\Delta)}(1-e^{-i\Delta t-\kappa t})\,. \tag{6.27}$$

In steady state one has

$$\rho=\left|\frac{ig}{\kappa+i\Delta}\right\rangle\left\langle\frac{ig}{\kappa+i\Delta}\right|\,. \tag{6.28}$$

Various steady-state correlation functions of the oscillator system can be obtained using (6.27) and the Markovian property of the system. We list several of these which are needed in the computations of the correlation functions C and Q:

$$\langle c^{\dagger}(\tau)c^2(\tau)c(0)\rangle=\frac{|g|^2}{(\kappa^2+\Delta^2)}\left[\frac{ig}{\kappa+i\Delta}\right]^2, \tag{6.29}$$

$$\langle c(0)c^{\dagger}(\tau)c^2(\tau)\rangle=\frac{|g|^2}{\kappa^2+\Delta^2}\left[\frac{ig}{\kappa+i\Delta}\right]^2+\left[\frac{ig}{\kappa+i\Delta}\right]^2(e^{i\Delta\tau-\kappa\tau})\,, \tag{6.30}$$

$$\langle c^{\dagger}(0)c^{\dagger}(\tau)c^2(\tau)\rangle=\frac{|g|^4}{(\kappa^2+\Delta^2)^2}\,, \tag{6.31}$$

$$\langle c^{\dagger}(\tau)c^2(\tau)c^{\dagger}(0)\rangle=\frac{|g|^4}{(\kappa^2+\Delta^2)^2}+\frac{2|g|^2}{(\kappa^2+\Delta^2)}e^{-i\Delta\tau-\kappa\tau}\,. \tag{6.32}$$

We now calculate the correlation functions for the anharmonic oscillator. From (6.24) we can show that

$$\left[\frac{d}{d\tau}+i\Delta+\kappa\right]\langle[c(\tau)-\langle c(\tau)\rangle,c(0)-\langle c(0)\rangle]_{\pm}\rangle=-2i\chi\langle[c^{\dagger}(\tau)c^2(\tau)-\langle c^{\dagger}(\tau)c^2(\tau)\rangle,c(0)-\langle c(0)\rangle]_{\pm}\rangle \tag{6.33}$$

and hence to first order in χ,

$$\hat{L}\langle[c(\tau)-\langle c(\tau)\rangle,c(0)-\langle c(0)\rangle]_{\pm}\rangle=(z+i\Delta+\kappa)^{-1}\{\langle[c(0)-\langle c(0)\rangle,c(0)-\langle c(0)\rangle]_{\pm}\rangle-2i\chi\hat{L}\langle[c^{\dagger}(\tau)c^2(\tau)-\langle c^{\dagger}(\tau)c^2(\tau)\rangle,c(0)-\langle c(0)\rangle]_{\pm}\rangle_0\}\,, \tag{6.34}$$

where $\hat{L}f$ denotes the Laplace transform of $f(\tau)$. The suffixes 0 and 1 indicate the orders of χ. Using (6.24) we find that

$$\langle c^2\rangle-\langle c\rangle^2=i\chi(2\langle c\rangle^3\langle c^{\dagger}\rangle-2\langle c\rangle\langle c^{\dagger}\rangle-1)\,. \tag{6.35}$$

Using Eqs. (6.29)–(6.32) and (6.35), we obtain the correlation functions of the oscillator operators

$$\hat{L}\langle[c(\tau),c(0)]\rangle=2i\chi(z+i\Delta+\kappa)^{-1}(z-i\Delta+\kappa)^{-1}\left[\frac{ig}{\kappa+i\Delta}\right]^2+O(\chi^2)=\hat{C}_{sc}^{++}(z)/(\mathbf{d}\cdot\boldsymbol{\varepsilon}_s^*)(\mathbf{d}\cdot\boldsymbol{\varepsilon}_c^*)\,, \tag{6.36}$$

$$\hat{L}\langle[c(\tau)-\langle c(\tau)\rangle,c(0)-\langle c(0)\rangle]_+\rangle=-2i\chi(z+i\Delta+\kappa)^{-1}(z-i\Delta+\kappa)^{-1}\left[\frac{ig}{\kappa+i\Delta}\right]^2+2i\chi(z+i\Delta+\kappa)^{-1}\left\{\frac{2|g|^2}{\kappa^2+\Delta^2}\left[\left[\frac{ig}{\kappa+i\Delta}\right]^2-1\right]-1\right\}+O(\chi^2)$$

$$\equiv\hat{Q}_{sc}^{++}(z)/(\mathbf{d}\cdot\boldsymbol{\varepsilon}_s^*)(\mathbf{d}\cdot\boldsymbol{\varepsilon}_c^*)\ , \tag{6.37}$$

$$\hat{C}_{ss}^{+-}(z)=|\mathbf{d}\cdot\boldsymbol{\varepsilon}_s^*|^2(z+i\Delta+\kappa)^{-1}\left[(-2i\chi)(z+i\Delta+\kappa)^{-1}\frac{2|g|^2}{(\kappa^2+\Delta^2)}+1\right]+O(\chi^2)\ , \tag{6.38}$$

$$\hat{Q}_{ss}^{+-}(z)=\hat{C}_{ss}^{+-}\ . \tag{6.39}$$

Using the explicit results (6.36)—(6.39), we have a complete description of the quantum theory of four-wave mixing in optical fibers. From the foregoing examples it is also clear how the quantum theory of four-wave mixing can be discussed for other types of media.

VII. QUANTUM THEORY OF NONLINEAR MIXING IN TWO PHOTON MEDIA

As a final application of the general theory of Secs. II and III, we consider four-wave mixing in two-photon media. Results of semiclassical calculations for such media can be found in Ref. 16. Let us consider the optical transitions in a ladder system consisting of the states labeled as $|1\rangle$ (upper most state), $|2\rangle$ (intermediate state), and $|3\rangle$ (ground state). A pump field ω interacts with three levels such that $2\omega\sim\omega_{13}$. We leave the intensity of the pump quite arbitrary. If the intermediate-state detunings are large then one has the case of a two-photon medium; otherwise, one in general has the case of stepwise transitions. We leave the intermediate-state detunings quite arbitrary so that by taking appropriate limits we will recover the cases of two-photon media and stepwise transitions.

The polarization operator for the three-level system under consideration can be written as

$$\mathbf{P}^+=\mathbf{d}_{32}A_{32}+\mathbf{d}_{21}A_{21},\quad A_{ij}=|i\rangle\langle j|\ . \tag{7.1}$$

The density-matrix equations can be cast into a matrix form the standard way[25] and we quote the result

$$\dot{\psi}=A\psi+I,\quad \psi_1=\rho_{11},\quad \psi_2=\rho_{12}e^{i\omega t},\quad \psi_3=\rho_{13}e^{2i\omega t},\quad \psi_4=\psi_2^*,\quad \psi_5=\rho_{22},\quad \psi_6=\rho_{23}e^{i\omega t}\ ,$$

$$\psi_7=\psi_3^*,\quad \psi_8=\psi_6^*,\quad I_6=I_8^*=ig_2,\quad g_1=\frac{\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}}{\hbar},\quad g_2=\frac{\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}}{\hbar}\ ,$$

$$A=\begin{bmatrix}
-2\gamma_1 & -ig_i^* & 0 & ig_1 & 0 & 0 & 0 & 0\\
-ig_1 & -\Gamma_{12}-i\Delta_1 & -ig_2^* & 0 & ig_1 & 0 & 0 & 0\\
0 & -ig_2 & -\Gamma_{13}-i\Delta_1-i\Delta_2 & 0 & 0 & ig_1 & 0 & 0\\
ig_1^* & 0 & 0 & -\Gamma_{12}+i\Delta_1 & -ig_1^* & 0 & ig_2 & 0\\
2\gamma_1 & ig_1^* & 0 & -ig_1 & -2\gamma_2 & -ig_2^* & 0 & ig_2\\
-ig_2 & 0 & ig_1^* & 0 & -2ig_2 & -\Gamma_{23}-i\Delta_2 & 0 & 0\\
0 & 0 & 0 & ig_2^* & 0 & 0 & -\Gamma_{31}+i\Delta+i\Delta_2 & -ig_1^*\\
ig_2^* & 0 & 0 & 0 & 2ig_2^* & 0 & -ig_1 & -\Gamma_{23}+i\Delta_2
\end{bmatrix},$$

$$\Delta_1=\omega_{12}-\omega,\quad \Delta_2=\omega_{23}-\omega\ . \tag{7.2}$$

Here, Γ_{ij} are the relaxation rates of the off-diagonal elements and these include contributions of collisions; $2\gamma_1$ and $2\gamma_2$ are the radiative decay rates of the states $|1\rangle$ and $|2\rangle$. The steady-state solution of (7.2) is

$$\psi=-A^{-1}I\ . \tag{7.3}$$

The atomic correlation functions relevant for the quantum theory can be calculated in terms of the steady-state solution (7.3) and the elements of the matrix $U(z)\equiv(z-A)^{-1}$. Our calculations lead to the following results for the correlation functions C and Q.

$$\begin{bmatrix} \hat{C}_{sc}^{++}(z) \\ \hat{Q}_{sc}^{++}(z) \end{bmatrix} = \sum_l [\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_s^* U_{6l}(z)+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_s^* U_{2l}(z)]\{(\mp\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*\psi_2,\mp\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*\psi_3,0,\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*(\psi_1\mp\psi_5),$$

$$\mp\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*\psi_6+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*\psi_2,\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*\psi_3,\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*\psi_4\mp\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*\psi_8,$$

$$\times\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*[\psi_5\mp(1-\psi_1-\psi_5)])_l-2q(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*\psi_6+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*\psi_2)\psi_l\}\ , \quad (7.4)$$

$$\begin{bmatrix} \hat{C}_{ss}^{+-}(z) \\ \hat{Q}_{ss}^{+-}(z) \end{bmatrix} = \sum_l [\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_s^* U_{6l}(z)+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_s^* U_{2l}(z)]$$

$$\times[(\mp\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s\psi_4,\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s(\psi_5\mp\psi_1),\mp\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_s\psi_2+\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s\psi_6,\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_s\psi_7,\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_s\psi_8$$

$$\mp\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s\psi_4,\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_s(1-\psi_1-\psi_5\mp\psi_5),0,-\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s\psi_7)_l-2q(\mathbf{d}_{23}\cdot\boldsymbol{\varepsilon}_s\psi_8+\mathbf{d}_{12}\cdot\boldsymbol{\varepsilon}_s\psi_4)\psi_l]\ . \quad (7.5)$$

In Eqs. (7.4) and (7.5) upper (lower) signs are to be taken for the correlation functions C's (Q's). The parameter q is to be set zero (one) for obtaining correlation functions C (Q).

We conclude this section by examining the results for two-photon media, assuming that the intermediate-state detunings Δ_1 and Δ_2 are large. We further assume that the saturation effects are unimportant and therefore we calculate the correlation functions C and Q to second order in the pump field. From Eq. (7.2) one can show that

$$\rho_{13}=-\frac{g_1g_2}{i\Delta_2[i(\Delta_1+\Delta_2)+\Gamma_{13}]}+O(g^3)\ ,$$
$$\langle \mathbf{P}^+\cdot\boldsymbol{\varepsilon}^*\rangle=-(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}^*)g_2/\Delta_2+O(g^2)\ . \quad (7.6)$$

From the operator algebra equal time correlation functions can be obtained:

$$\langle[A_{21}(0),\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}^*A_{32}(0)+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}^*A_{21}(0)]_\pm\rangle =\pm\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}^*\rho_{13}\ , \quad (7.7)$$

$$\langle[A_{32}(0),\mathbf{P}^+\cdot\boldsymbol{\varepsilon}^*]_\pm\rangle=+\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}^*\rho_{13}\ . \quad (7.8)$$

Using Eqs. (7.2)—(7.5), detailed calculations show that

$$\hat{L}\langle[A_{21}(\tau),(\mathbf{P}^+\cdot\boldsymbol{\varepsilon}^*)]_\mp\rangle=\pm\frac{\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}^*g_1g_2}{i\Delta_2[i(\Delta_1+\Delta_2)+\Gamma_{13}]}\frac{1}{(z+i\Delta_1)}+O(g^3), \quad (7.9)$$

$$\hat{L}\langle[A_{32}(\tau),(\mathbf{P}^+\cdot\boldsymbol{\varepsilon}^*)]_\mp\rangle=-\frac{\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}^*g_1g_2}{i\Delta_2[i(\Delta_1+\Delta_2)+\Gamma_{13}]}\frac{1}{(z+i\Delta_2)}+O(g^3)+(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}^*)\begin{bmatrix}\frac{2g_2}{\Delta_2}\end{bmatrix}z^{-1}(+ig_2)\frac{1}{(z+i\Delta_2)}\ . \quad (7.10)$$

From Eqs. (7.6), (7.9), and (7.10) we find the following results for the correlation functions $\hat{C}$ and $\hat{Q}$:

$$\hat{Q}_{cs}^{++}(z)=\left[\frac{(\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*)(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_s)}{(z+i\Delta_1)}+\frac{(\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_s^*)(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_c^*)}{(z+i\Delta_2)}\right]\frac{g_1g_2}{i\Delta_2[\Gamma_{13}+i(\Delta_1+\Delta_2)]}+\text{nonresonant contributions}\ , \quad (7.11)$$

$$\hat{C}_{cs}^{++}(z)=\frac{g_1g_2}{i\Delta_2[\Gamma_{13}+i(\Delta_1+\Delta_2)]}\left[\frac{(\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_c^*)(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_s^*)}{(z+i\Delta_1)}-\frac{(\mathbf{d}_{21}\cdot\boldsymbol{\varepsilon}_s^*)(\mathbf{d}_{32}\cdot\boldsymbol{\varepsilon}_s^*)}{(z+i\Delta_2)}\right]+\text{nonresonant contributions}\ , \quad (7.12)$$

which can be used in the quantum theory of four-wave mixing in two-photon media. For the degenerate case one has $z=0$. It is interesting to observe that at exact two-photon resonance, $\Delta_1+\Delta_2=0$, the fluctuation term Q^{++} is not important if the dipole matrix elements are nearly equal.

In conclusion we have developed a general quantum theory of nonlinear mixing in arbitrary media. Central to the theory are the two time correlation functions involving the commutators and anticommutators of the polarization operators.[26] Such correlations can be calculated from the knowledge of the microscopic dynamics of the medium pumped by an external field. Use of the Wigner distribution function enables us to obtain general results on the quantum statistics of the generated fields. Other detailed applications of the present theory will be treated in future papers.

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[26]If the system were in thermal equilibrium then the fluctuation dissipation theorem [cf. R. Kubo, Rep. Prog. Phys. **29**, 255 (1966)] relates the Fourier transform of the correlation functions C and Q. However, we are dealing with systems which are far from thermal equilibrium and C and Q are distinct quantities and no relation between these two exists in general.