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I. Introduction

It is now well known that nonlinear optical effects arise as a result of nonlinear response of a medium to intense light fields. The description of these effects is often semi-classical and non statistical - the light fields are treated as classical waves with no fluctuations either in amplitudes or phases. Such a description neglects the contribution of spontaneous emission to the stimulated scattering. The questions of how the statistical properties of the light fields are disturbed and how the nonlinear optical effects depend on the statistical properties of light fields are also left unanswered. In fact, as one would expect, statistical treatment of nonlinear effects should be more important since they always manifest stronger fluctuations than linear effects. Thus, a complete description of a nonlinear optical effect requires the application of quantum statistics.[1]

In this review, we shall not concern ourselves too much about how the statistical properties of the intense pump fields are changed in a nonlinear optical process. We shall always assume that the depletion of power in the pump fields is negligible. Consequently, the perturbance in the statistical properties of the pump fields can also be neglected. We are more interested in how the average rate of a nonlinear optical process is effected by statistical fluctuations in the pump fields and in the material properties. Above all, it is interesting to find the statistical properties of the fields generated or amplified in the nonlinear process as a function of the statistical properties of the pump fields and of the medium. Conversely, measurements on the nonlinearly generated fields should yield information about the statistical properties of the pump field and/or of the medium.

In the following sections, four important problems of nonlinear optics will be discussed, namely, multiphoton emission and absorption, incoherent scattering, sum and difference frequency generation, and parametric amplification and oscillation. Each field component is assumed to be a single mode. Extension of the calculations to multimode problems is straightforward, and will be discussed briefly in Sec. VI. We shall assume for all cases except incoherent scattering that the light fields are contained in a cavity. However, as one would expect from the corresponding classical description, a cavity problem of coherent scattering can be converted to a steady-state travelling wave problem by simply replacing t by $-z\epsilon^{1/2}/c$ where $\hat{z}$ is the direction of propagation. That this is also true for our quantum description will be illustrated in the final section.

II. Multiphoton Absorption and Emission, Raman Transitions

Let us begin by assuming as the unperturbed system the fields in a cavity filled with linear, isotropic, non-absorbing medium with a linear dielectric constant $\epsilon_k(\underset{\sim}{r})$ at frequency ω_k. From the Dirac quantization process,[2] we can write the vector potential in the usual form

$$\underset{\sim}{A}(\underset{\sim}{r},t) = c \sum_k (2\pi\hbar/\omega_k)^{1/3}\{a_k \underset{\sim}{u}_k(\underset{\sim}{r})\exp(i\omega_k t) + a_k^\dagger \underset{\sim}{u}_k^*(\underset{\sim}{r}) \exp(-i\omega_k t)\} \quad (1)$$

where $a_k^\dagger$ and a_k are the creation and the annihilation operators for the

k^{th} mode. The spatial function $\underset{\sim}{u}_k(\underset{\sim}{r})$ is an eigenfunction of the equation

$$[\nabla^2 + \omega_k{}^2 \epsilon_k(\underset{\sim}{r})/c^2]\, \underset{\sim}{u}_k(\underset{\sim}{r}) = 0 \tag{2}$$

and obeys the orthonormality condition

$$\int (\epsilon_k \epsilon_\ell)^{\frac{1}{2}}\, \underset{\sim}{u}_k(\underset{\sim}{r}) \cdot \underset{\sim}{u}_\ell{}^*(\underset{\sim}{r}) d^3r = \delta_{k\ell} \; . \tag{3}$$

The Hamiltonian for this unperturbed system is simply

$$\mathcal{H}_o = \sum_k \hbar\omega_k (a_k{}^\dagger a_k + \frac{1}{2}) + \mathcal{H}_{matter} \; . \tag{4}$$

The Hamiltonian describing absorption or emission of photons in the medium is then taken as a perturbation.

Consider first the case of single-photon transitions between two states $|\psi_1>$ and $|\psi_2>$ with a frequency separation $\omega_{21} \approx \omega_k$. This can be described by a perturbing Hamiltonian

$$\mathcal{H}_1 = \sum_i \{\xi\, c_{2i}^\dagger c_{1i}\, \underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i) + \xi^* c_{2i}\, c_{1i}^\dagger\, \underset{\sim}{E}_k^{(+)}(\underset{\sim}{r}_i)\, \} \tag{5}$$

where c_{1i}, c_{2i}, $c_{1i}^\dagger$, and $c_{2i}^\dagger$ are creation and annihilation operators for electronic states 1 and 2 respectively at the i^{th} atom, ξ is the electric-dipole matrix element between the two states, and

$$\underset{\sim}{E}_k^{(+)}(\underset{\sim}{r}_i) = [\underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i)]^\dagger = i(2\pi\hbar\omega_k)^{\frac{1}{2}}\, \underset{\sim}{u}_k{}^*\, (\underset{\sim}{r}_i) a_k^\dagger \; . \tag{6}$$

We are interested in the change of statistical properties of the fields, which is most easily described by the density matrix formalism. In the interaction representation, the density matrix ρ obeys the equation of motion

$$i\hbar\, \partial\rho/\partial t = [\mathcal{H}_1^*, \rho] \tag{7}$$

where

$$\begin{aligned} \mathcal{H}_1^* &= \exp(i\mathcal{H}_0 t/\hbar)\mathcal{H}_1 \exp(-i\mathcal{H}_0 t/\hbar) \\ &= \sum_i (\xi\, c_{2i}^\dagger c_{1i} \underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i) \exp[i(\omega_k - \omega_{21})t] + \text{Adjoint}). \end{aligned} \tag{8}$$

By using a procedure often used in the relaxation calculation for magnetic resonance,[3] Eq. (7) can be reduced to give an equation of motion for the density matrix ρ_F of the fields alone. First, the equation is expanded through iteration to yield[3]

$$\begin{aligned} \partial\rho(t + \Delta t)/\partial t &= (1/i\hbar)[\mathcal{H}_1^*(t + \Delta t), \rho(t)] \\ &\quad -(1/\hbar^2)\int_t^{t+\Delta t} [\mathcal{H}_1^*(t + \Delta t), [\mathcal{H}_1^*(t'), \rho(t)]]dt' + \cdots \end{aligned} \tag{9}$$

Then, an irreversible approximation on the density matrix is used by assuming that the thermal equilibrium of the matter system is hardly disturbed by interaction with the photon fields, so that we can write $\rho(t) = \rho_F(t) \prod_i \rho_{Ai}(0)$, where $\rho_{Ai}(0)$ is the density matrix for the i^{th} atom at thermal equilibrium with diagonal matrix elements ρ_{1A}^o and

ρ_{2A}^{o} indicating the thermal populations in the two states $|\psi_1>$ and $|\psi_2>$. The off-diagonal elements of $\rho_A(0)$ always vanish. Equation (9) can now be reduced by taking trace over the matter system. With the help of Eq. (8), straightforward calculation will lead to

$$\partial\rho_F(t)/\partial t = -\beta[(a_k^\dagger a_k\rho_F - 2a_k \rho_F a_k^\dagger + \rho_F a_k^\dagger a_k)\rho_{1A}^{o} + (a_k a_k^\dagger \rho_F - 2a_k^\dagger \rho_F a_k + \rho_F a_k a_k^\dagger)\rho_{2A}^{o}] + \cdots \quad (10)$$

with

$$\beta = \sum_i 2\pi^2\omega_k|\xi|^2|\underset{\sim}{u}_k(\underset{\sim}{r}_i)|^2 g(\omega_k)/\hbar$$

$$= [2\pi^2\omega_k|\xi|^2 g(\omega_k)/\hbar] \int_V d^3r\, |\underset{\sim}{u}_k(\underset{\sim}{r})|^2 N(\underset{\sim}{r})$$

$$g(\omega_k) = (1/2\pi) \int_{-\Delta t}^{\Delta t} \exp[-i(\omega_k - \omega_{21})t]dt$$

where $N(\underset{\sim}{r})$ is the atomic density at $\underset{\sim}{r}$. If $\Delta t < \hbar/|\mathcal{H}_1|$, higher-order terms in Eq. (9) or (10) can be neglected. If, in addition, $\Delta t \gg$ (1/linewidth), then the limits of integration in $g(\omega_k)$ can be approximated by $-\infty$ to $+\infty$, and hence $g(\omega_k)$ becomes a δ-function, or more generally a lineshape function centered at ω_{21}. These restrictions on Δt are in fact exactly the same as those required for ordinary time-dependent perturbation calculation.[4]

Equation (10) governs the change of statistical properties of the photon system in the single-photon absorption $(\rho_{1A}^{o} > \rho_{2A}^{o})$ or emission $(\rho_{1A}^{o} < \rho_{2A}^{o})$ process. This is actually the same equation derived by

Scully and Lamb[5] for a laser amplifier or oscillator except that the higher-order nonlinear terms have been neglected. The derivation can of course be extended to include the nonlinear terms without much difficulty. The solution of Eq. (10) can be obtained analytically for individual density matrix elements in the number representation, if either ρ^o_{1A} or ρ^o_{2A} vanishes. In the absorption process, it is easy to show that at zero temperature ($\rho^o_{2A} = 0$), the statistical properties of a photon system, which can be described by the P-representation,[6] remain basically unchanged.[1] At finite temperature, however, they are disturbed by spontaneous emission in the absorbing medium.

A similar calculation can be applied to the case of two-photon absorption or emission. Here, the perturbing Hamiltonian for transitions between the states $|\psi_1>$ and $|\psi_2>$ is

$$\mathcal{H}_1 = \sum_i \{\xi^{(2)} c^\dagger_{2i} c_{1i} \underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i)\underset{\sim}{E}_\ell^{(-)}(\underset{\sim}{r}_i) + \text{Adjoint}\} \tag{11}$$

where $\xi^{(2)}$ is the matrix element for the two-photon transitions. Using exactly the same procedure as in the single-photon transition case, one would obtain

$$\begin{aligned}\partial\rho_F/\partial t = -\beta^{(2)} [&(a^\dagger_k a^\dagger_\ell a_k a_\ell \rho_F - 2a_k a_\ell \rho_F a^\dagger_k a^\dagger_\ell + \rho_F a^\dagger_k a^\dagger_\ell a_k a_\ell)\rho^o_{1A} \\ &+(a_k a_\ell a^\dagger_k a^\dagger_\ell \rho_F - 2a^\dagger_k a^\dagger_\ell \rho_F - 2a^\dagger_k a^\dagger_\ell \rho_F a_k a_\ell + \rho_F a_k a_\ell a^\dagger_k a^\dagger_\ell)\rho^o_{2A}]\end{aligned} \tag{12}$$

with

$$\beta^{(2)} = 4\pi^3\omega_k\omega_\ell \, |\xi^{(2)}|^2 g(\omega_k + \omega_\ell) \int_V d^3r \, N(r)|u_k(r)|^2|u_\ell(r)|^2.$$

The above equation of motion governs the change of statistical properties of the photon fields in two-photon absorption or emission. In particular, from Eq. (12), we find for $k \neq \ell$, [7]

$$\partial < a_k^\dagger a_k >/\partial t = \partial < a_\ell^\dagger a_\ell >/\partial t$$
$$= -2\beta^{(2)}[(\rho_{1A}^o - \rho_{2A}^o) < a_k^\dagger a_k a_\ell^\dagger a_\ell > - \rho_{2A}^o < a_k^\dagger a_k + a_\ell^\dagger a_\ell + 1>] \qquad (13)$$

and for $k = \ell$

$$\partial < a_k^\dagger a_k >/\partial t = -4\beta^{(2)}[(\rho_{1A}^o - \rho_{2A}^o) < a_k^\dagger a_k^\dagger a_k a_k > - \rho_{2A}^o(2< a_k^\dagger a_k > + 1)]. \qquad (14)$$

Equations (13) and (14) show that the rate of photon absorption or emission depends on the second-order correlation function of the fields, and hence on the initial statistical properties of the fields. For $k = \ell$, even the initial two-photon transition rate is higher for chaotic than for coherent fields, since[6].

$$< a^\dagger a^\dagger a \; a >_{chaotic} = 2(< a^\dagger a >)^2$$
$$< a^\dagger a^\dagger a \; a >_{coherent} = (< a^\dagger a >)^2 \quad . \qquad (15)$$

Physically, a chaotic field has more fluctuations. Nonlinear response always amplifies the fluctuations, and yields a higher average value.

Lambropoulous et al. have also discussed the case of two-photon absorption and emission with $k = \ell$. Although in a different form, they find the same equation as our Eq. (12). Their derivation is however much more complicated and less transparent.

Analytical solution of $\rho_F(t)$ for Eq. (12) can be obtained for $\rho^o_{1A} = 0$ or $\rho^o_{2A} = 0$ in the number representation.[8] Consider two-photon emission with $\rho^o_{1A} = 0$. Equation (12) gives

$$\partial\rho_{n_k n_\ell;n'_k n'_\ell}/\partial t = C_n \rho_{n_k n_\ell;n'_k n'_\ell} + D_n\, \rho_{(n-1)_k (n-1)_\ell;(n'-1)_k (n'-1)_\ell}$$

$$C_n = -\beta^{(2)}\rho^o_{2A}[(n_k+1)(n_\ell+1) + (n'_k+1)(n'_\ell+1)]$$

$$D_n = 2\beta^{(2)}\rho^o_{2A}[n_k n_\ell n'_k n'_\ell]^{\frac{1}{2}} . \tag{16}$$

From the above equation, we obtain the Laplace transform of $\rho_{n_k n_\ell;n'_k n'_\ell}$,

$$\begin{aligned}\varphi_{n_k n_\ell;n'_k n'_\ell}(s) &= [\rho_{n_k n_\ell;n'_k n'_\ell}(0) + D_n \varphi_{(n-1)_k(n-1)_\ell;(n'-1)_k(n'-1)_\ell}]/(s-C_n) \\ &= \sum_{i=0}^{M} \rho_{(n-i)_k(n-i)_\ell;(n'-i)_k(n'-i)_\ell}(0) \prod_{q=0}^{i-1} D_{n-q} \Big/ \prod_{q=0}^{i} (s-C_{n-q})\end{aligned} \tag{17}$$

where M denotes the smallest integer among the set $(n_k, n_\ell, n'_k, n'_\ell)$. The inverse transform yields

$$\rho_{n_k n_\ell;n'_k n'_\ell}(t) = \sum_{i=0}^{M}\sum_{r=0}^{i} [\prod_{q=0}^{i=1} D_{n-q} \Big/ \prod_{\substack{q=0\\ q\neq r}}^{i} (C_{n-r} - C_{n-q})] \times \rho_{(n-i)_k(n-i)_\ell;(n'-i)_k(n'-i)_\ell}(0) \exp(C_{n-r}t) \tag{18}$$

which fully describes the statistical properties of a two-photon amplifier or oscillator. Because of limited space, we shall not go into detailed analysis of Eq. (18) here except a few remarks. 1) If initially, ρ has only diagonal elements $(n_k = n_k', n_\ell = n_\ell')$, then it remains so at any time t. In fact, the off-diagonal elements of ρ, even if they exist, would vanish eventually through diffusion processes as in the case of a laser oscillator.[5,6] 2) If initially, $\rho_{n_k n_\ell; n_k n_\ell}(0) = \rho_{n_k n_k}(0) \rho_{n_\ell n_\ell}(0)$ has a distribution peaked at $n_k = n_{k_0}$ and $n_\ell = n_{\ell_0}$, then as time progresses, the peak will move to larger values of n_k and n_ℓ, as one would expect from a two-photon amplifier or oscillator. Statistical properties of the fields also change with time. 3) Since higher-order terms have been neglected in the calculation, the solution does not show saturation of amplification or stabilization of oscillation.

The two-photon oscillator is of great practical importance, since there is potential possibility of its becoming a tunable oscillator. It is interesting to note that if initially E_k is a strong coherent field, such that $E_k^{(\pm)}$ in Eq. (11) can be treated as constant c numbers, then the Hamiltonian $\mathcal{H}_1$ reduces to the form for single-photon transitions, and the E_ℓ field created in the two-photon emission process has the same statistical properties as those of the field created in a laser oscillator. This is of course a practically realizable case.

The calculation can be extended to the case of n-photon absorption or emission. The perturbing Hamiltonian then becomes

$$\mathcal{H}_1 = \sum_i \{\xi^{(n)} c_{2i}^\dagger c_{1i} \prod_{k=1}^{n} \underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i) + \text{Adjoint}\}. \tag{19}$$

The corresponding equation of motion for ρ_F is

$$\partial\rho_F/\partial t = -\beta^{(n)} \Big[\Big(\prod_{k=1}^{n} a_k^\dagger \prod_{k=1}^{n} a_k \rho_F - 2 \prod_{k=1}^{n} a_k \rho_F \prod_{k=1}^{n} a_k^\dagger + \rho_F \prod_{k=1}^{n} a_k^\dagger \prod_{k=1}^{n} a_k\Big)\rho_{1A}^{o}$$
$$+ \Big(\prod_{k=1}^{n} a_k \prod_{k=1}^{n} a_k^\dagger \rho_F - 2 \prod_{k=1}^{n} a_k^\dagger \rho_F \prod_{k=1}^{n} a_k + \rho_F \prod_{k=1}^{n} a_k \prod_{k=1}^{n} a_k^\dagger\Big)\rho_{2A}^{o}\Big] \tag{20}$$

with

$$\beta^{(n)} = (2\pi/\hbar^2)\Big(\prod_{k=1}^{n} 2\pi\hbar\omega_k\Big)|\xi^{(n)}|^2\, g\Big(\sum_{k=1}^{n} \omega_k\Big)\int_V d^3r N(\underset{\sim}{r}) \prod_{k=1}^{n} |\underset{\sim}{u}_k(\underset{\sim}{r})|^2 .$$

The calculation requires only slight modification if in an n-photon transition process, photons with $k = 1, \cdots, m$ are absorbed, but photons with $k = m + 1, \cdots, n$ are emitted. The perturbing Hamiltonian becomes

$$\mathcal{H}_1 = \sum_i \{\xi^{(n)} c_{2i}^\dagger c_{1i} \prod_{k=1}^{m} \underset{\sim}{E}_k^{(-)}(\underset{\sim}{r}_i) \prod_{k=m+1}^{n} \underset{\sim}{E}_k^{(+)}(\underset{\sim}{r}_i) + \text{Adjoint}\} \tag{21}$$

and the equation of motion for ρ_F must be modified accordingly. A case of practical interest is the Raman transitions, in which one photon at ω_ℓ is absorbed and another photon at ω_s emitted as the atomic energy system makes transition from $|\psi_1\rangle$ to $|\psi_2\rangle$. The equation of motion for ρ_F in this case is

$$\partial\rho/\partial t = -\beta^{(2)}[(a_\ell^\dagger a_s a_\ell a_s^\dagger \rho_F - 2a_\ell a_s^\dagger \rho_F a_\ell^\dagger a_s + \rho_F a_\ell^\dagger a_s a_\ell a_s^\dagger)\rho_{1A}^{o}$$
$$+ (a_\ell a_s^\dagger a_\ell^\dagger a_s \rho_F - 2a_\ell^\dagger a_s \rho_F a_\ell a_s^\dagger + \rho_F a_\ell a_s^\dagger a_\ell^\dagger a_s)\rho_{2A}^{o}] \tag{22}$$

from which we obtain the rate of Stokes generation

$$\partial < a_s^\dagger a_s > /\partial t = -\partial < a_\ell^\dagger a_\ell > / \partial t$$

$$= 2\beta^{(2)} [(\rho_{1A}^o - \rho_{2A}^o) < a_\ell^\dagger a_\ell a_s^\dagger a_s >$$

$$+ < a_\ell^\dagger a_\ell > \rho_{1A}^o - < a_s^\dagger a_s > \rho_{2A}^o] \qquad (23)$$

as was given by Hellwarth.[9] Usually, E_ℓ is a strong field, such that a_ℓ and $a_\ell^\dagger$ in Eq. (22) can be treated as constant c numbers but subject to statistics. The solution of Eq. (23) then becomes

$$< a_s^\dagger a_s > (t) = \mathrm{Tr}_\ell \, \rho_\ell(0)\{[< a_s^\dagger a_s > (0) + A/B] \exp(Bt) - A/B) \qquad (24)$$

where

$$A = 2\beta^{(2)} a_\ell^\dagger a_\ell \rho_{1A}^o$$

$$B = 2\beta^{(2)}[a_\ell^\dagger a_\ell(\rho_{1A}^o - \rho_{1A}^o) - \rho_{2A}^o] \approx 2\beta^{(2)} a_\ell^\dagger a_\ell(\rho_{1A}^o - \rho_{2A}^o).$$

It is clear from Eq. (24) that a chaotic pump field would yield a more intense Stokes field than a coherent pump field, since[6]

$$\mathrm{Tr}_\ell \rho_\ell(0) \exp[2\beta^{(2)} a_\ell^\dagger a_\ell(\rho_{1A}^o - \rho_{1A}^o)] = \sum_{n=o}^{\infty} [2\beta^{(2)}(\rho_{1A}^o - \rho_{2A}^o)]^n <(a_\ell^\dagger a_\ell)^n>/n!$$

$$<(a_\ell^\dagger a_\ell)^n>_{chaotic} = n! < a_\ell^\dagger a_\ell >$$

$$< (a_\ell^\dagger a_\ell)^n>_{coherent} = < a_\ell^\dagger a_\ell >^n \qquad (25)$$

In many cases, one may also find $\rho_{2A}^{o} \approx 0$. Then, analytical solution for $\rho_F(t)$ can be obtained from Eq. (22) using the same procedure as in the two-photon emission case. With a_ℓ and $a_\ell^\dagger$ treated as constant c numbers with a statistical distribution, the density matrix elements for the growing Stokes field at time t are given by

$$\rho_{n_s,n_s'}(t) = \sum_{i=o}^{M} \sum_{r=o}^{i} \left[\prod_{q=o}^{i-1} D_{n-q} \Big/ \prod_{\substack{q=o \\ q\neq r}}^{i} (c_{n-r} - c_{n-q}) \right]$$

$$\times \rho_{(n-i)_s,(n'-i)_s}(0) \exp(c_{n-r}\, t) \qquad (26)$$

with

$$c_n = -(n_s + n_s' + 2)\beta^{(2)} a_\ell^\dagger a_\ell \rho_{1A}^{o}$$

$$D_n = (n_s\, n_s')^{\frac{1}{2}}\, 2\beta^{(2)}\, a_\ell^\dagger a_\ell \rho_{1A}^{o} .$$

If in addition, the pump field E_ℓ is also coherent, then a_ℓ and $a_\ell^\dagger$ can be replaced by constants α_ℓ and α_ℓ^*. Equation (22) for the Stokes field now becomes the same as for a laser amplifier except that ρ_{1A}^{o} and ρ_{2A}^{o} are interchanged. Therefore, the statistical properties of the Stokes field generated in stimulated Raman transitions would also be the same as those of a laser amplifier. Saturation of Stokes amplification or oscillation has not been taken into account in the above discussion.

III. Incoherent Scattering

Linear, incoherent Rayleigh and Brillouin scattering belongs to the class of nonlinear optics in the sense that an excitational wave in the medium now plays the role of a light wave. The quantum description of incoherent scattering has close resemblance to the classical description.[10]

Let us consider only scattering by density fluctuations. The perturbing Hamiltonian in this case is

$$\mathcal{H}_1 = -\sum_{i,k} [\, \underset{\sim}{E}_k^{(+)}(\underset{\sim}{r}_i) \cdot \underset{\approx}{p} \cdot \underset{\sim}{E}_{k_o}^{(-)}(\underset{\sim}{r}_i) + \text{Adjoint}] \tag{27}$$

where $\underset{\approx}{p}$ is the polarizability, k_o the pump mode, and k the scattered modes. If we take into account the propagation effect by assuming running modes for the fields with

$$\underset{\sim}{u}_k(\underset{\sim}{r}) = \hat{e}_k(1/L^3\ \epsilon_k)^{\frac{1}{2}} \exp(i\underset{\sim}{k}\cdot\underset{\sim}{r}),$$

then Eq. (26) becomes

$$\mathcal{H}_1 = \sum_k [\, a_k^\dagger(t)\ a_{k_o}(t)\ f_k^* + \text{Adjoint}] \tag{28}$$

where in the Heisenberg representation,

$$f_k^* = -\sum_i (2\pi\hbar\omega_k)^{\frac{1}{2}}\ \omega_{k_o}^{\frac{1}{2}} /\epsilon_k L^3)\ \hat{e}_k \cdot \underset{\approx}{p}^* \cdot \hat{e}_{k_o} \exp[i(\underset{\sim}{k}-\underset{\sim}{k}_o) \cdot r_i].$$

The corresponding equation of motion is

$$da_k(t)/dt = i\omega_k a_k(t) - (i/\hbar)f_k^{\,*} a_{k_o}(t) \; . \qquad (29)$$

Usually, the pump field is of relatively high intensity, and is hardly disturbed by incoherent scattering. We can therefore treat a_{k_o} and $a_{k_o}^{\dagger}$ as c numbers; but subject to statistical distribution. The problem then becomes essentially classical. In fact, it reduces to the one discussed by Glauber on radiation by a prescribed current distribution.[6] The solution of Eq. (29) would lead to the classical expression for scattered radiation. In our notations, at a point r sufficiently far away from the scattering region, the scattered field is given by[1]

$$E^{(-)}_{sc.}(\underset{\sim}{r},t) = a_{k_o} \underset{\sim}{F}(\underset{\sim}{r},t) \exp(i\underset{\sim}{k}_o\cdot\underset{\sim}{R})\int_V d^3r' \; N(\underset{\sim}{r}',t)\exp\left[i(\underset{\sim}{k}_o-\underset{\sim}{k})\cdot\underset{\sim}{r}'\right] \qquad (30)$$

$$\underset{\sim}{F}(\underset{\sim}{r},t) = (\underset{\sim}{k} \times i\underset{\approx}{p} \cdot \hat{e}_{k_o}) \times (\underset{\sim}{k}/|\underset{\sim}{r}-\underset{\sim}{R}|)(2\pi\hbar\omega_o/\epsilon_{k_o} L^3)^{1/2} \exp(i\underset{\sim}{k}\cdot\underset{\sim}{r}-i\omega_o t)$$

where $\underset{\sim}{R}$ is the center of the scattering volume V. From Eq. (30), we obtain the first-order correlation function

$$\langle \underset{\sim}{E}^{(+)}_{sc.}(\underset{\sim}{r}_1,t_1)\, \underset{\sim}{E}^{(-)}_{sc.}(\underset{\sim}{r}_2,t_2)\rangle = \underset{\sim}{F}(\underset{\sim}{r}_1)\cdot\underset{\sim}{F}^*(\underset{\sim}{r}_2) < a_{k_o}^{\dagger} a_{k_o} > \exp\left[ik\cdot(\underset{\sim}{r}_1-\underset{\sim}{r}_2)-i\omega_o(t_1-t_2)\right]$$

$$\times \int_V d^3r d^3r' \langle N(\underset{\sim}{r},t_1)N(\underset{\sim}{r}',t_2)\rangle \exp[i(\underset{\sim}{k}_o-\underset{\sim}{k})\cdot(\underset{\sim}{r}-\underset{\sim}{r}')] . \qquad (31)$$

Note that the Fourier transform of $< \underset{\sim}{E}^{(+)}_{sc.}(\underset{\sim}{r},t_1)\, \underset{\sim}{E}^{(-)}_{sc.}(\underset{\sim}{r},t_2) >$ gives the power spectral density of the scattered radiation, and the Fourier transform of $< E^{(+)}_{sc.}(r_1,t)\, E^{(-)}_{sc.}(r_2,t)>$ gives the power density distribution of the scattered radiation in the wave-vector space. From Eq. (31), it is clear that the average scattering intensity is proportional to the average number of photons in the pump mode, and is independent of the coherent property of the pump field. Higher-order correlation functions of $\underset{\sim}{E}_{sc}$ can be obtained from Eq. (30), and hence the statistical properties of the scattered radiation are described completely. It is seen that the n^{th} order correlation functions of E_{sc} is related to the n^{th} order correlation functions of a_{k_o} and $N(\underset{\sim}{r},t)$. Therefore, if the statistical properties of the pump field are known, then measurements of correlation functions of E_{sc} (for example, from photon counting measurements[6]) would yield information about the statistical properties of the density variation. Thus, for example, if the thermal density fluctuations are elastic with a Gaussian distribution, the n^{th} order correlation functions of $N(\underset{\sim}{r},t)$ can be written as[6]

$$G_N^{(n)}(\underset{\sim}{r}_1,t_1 \ldots r_{2n},t_{2n}) = \sum_p \prod_{j=1}^{n} G_N^{(1)}(\underset{\sim}{r}_j,t_j; \underset{\sim}{r}_{p(n+j)},t_{p(n+j)})$$

where the sum $\sum_p$ is taken over the $n!$ possible ways of permuting the set of coordinates $\underset{\sim}{r}_{n+1}, t_{n+1}; \ldots \underset{\sim}{r}_{2n}, t_{2n}$. If the pump field is in a coherent state, so that a_{k_o} can be replaced by the parameter α_{k_o}, then it is readily shown that the n^{th} order correlation function of $\underset{\sim}{E}_{sc}$ can also be written as

$$G_{E_{sc}}^{(n)}(\underset{\sim}{r}_1,t_1 \ldots \underset{\sim}{r}_{2n}, t_{2n}) = \sum_p \prod_{j=1}^{n} G_{E_{sc}}^{(1)}(\underset{\sim}{r}_j,t_j; r_{p(n+j)}\ t_{p(n+j)})$$

which indicates that the scattered radiation is also chaotic with a Gaussian distribution. Crosignani, Di Porto, et al.[11] have discussed the possibility of investigating statistical fluctuations in liquids and in plasmas from the photon statistics of the scattered light.

The integral in Eq. (31) is sometimes decomposed in two parts.[12]

$$\int_V d^3r d^3r' < N(\underset{\sim}{r},t_1)N(\underset{\sim}{r}',t_2) > \exp[i(\underset{\sim}{k} - \underset{\sim}{k}_o)\cdot(\underset{\sim}{r} - \underset{\sim}{r}')]$$

$$= \int_V d^3r < N(\underset{\sim}{r},t_1)N(\underset{\sim}{r},t_2)> + \int_{\substack{V \\ r\neq r'}} d^3r d^3r' <N(\underset{\sim}{r},t_1)N(\underset{\sim}{r}',t_2)> \exp[i(\underset{\sim}{k}-\underset{\sim}{k}_o)\cdot(\underset{\sim}{r}-\underset{\sim}{r}')] \qquad (32)$$

The second part in the above equation shows explicit dependence on correlation of density fluctuations at two space-time points. Higher order correlation functions can be expressed in a similar way.

The density fluctuations are usually Fourier decomposed into

$$N(\underset{\sim}{r},t) = \sum_q N_q(\underset{\sim}{r},t)\exp(i\underset{\sim}{q}\cdot\underset{\sim}{r} - i\omega_q t). \qquad (33)$$

Then, it is clear from Eq. (30) that the scattered radiation with wave vector $\underset{\sim}{k}$ comes essentially from the Fourier components $N_q(\underset{\sim}{r},t)$ with $\underset{\sim}{q} = \underset{\sim}{k}_o - \underset{\sim}{k}$. In crystals, density fluctuations can be quantized in the form of acoustic phonons. Accordingly, N_q and $N_q^\dagger$ become operators proportional to the annihilation and the creation operators of phonons respectively.

For incoherent scattering, explicit expression of the density matrix for the scattered radiation can also be obtained, following Glauber's calculation for the radiation by a prescribed current distribution.[6] We find

$$\rho_k(t) = D(\alpha_k)|0><0|D^{-1}(\alpha_k) \tag{34}$$

$$D(\alpha_k) = \exp[\alpha_k(a_{k_o}, N, t)\, a_k^\dagger - \text{Adjoint}]$$

or in the coherent-state representation,

$$\rho_k(t) = |\alpha_k><\alpha_k|$$

where $\alpha_k(a_{k_o}, N, t) = (-i/\hbar)\int_o^t dt(2\pi\hbar\omega_{k_o}^{1/2}\omega_k^{1/2}/\epsilon_{k_o} L^3)\hat{e}_k \cdot \underset{\approx}{p} \cdot \hat{e}_{k_o}\, a_{k_o}$

$$\int_V d^3\underset{\sim}{r}\, N(r,t) \exp[i(\underset{\sim}{k}-\underset{\sim}{k}_o)\cdot\underset{\sim}{r}]$$

with the statistical distribution of a_k and $N(\underset{\sim}{r}, t)$ taken into account.

Eq. (33) would yield the same expressions for the correlation functions of $\underset{\sim}{E}_{sc.}$ as obtained from Eq. (30). Thus,

$$< E^{(+)}_{sc.}(r_1,t_1)\cdots E^{(+)}_{sc.}(r_n,t_n),\ E^{(-)}_{sc.}(r_{n+1},\ t_{n+1}) \cdots E^{(-)}_{sc.}\ (r_{2n},\ t_{2n}) >$$

is proportional to

$$< (a^{\dagger}_{k_o})^n (a_{k_o})^n > \int_V \left(\prod_{i=1}^{2n} d^3 r_i\right) < \prod_{i=1}^{2n} N(\underset{\sim}{r}_i, t_i) > \exp[i(\underset{\sim}{k}_o - \underset{\sim}{k})\cdot \sum_{i=n+1}^{2n} \underset{\sim}{r}_i - \sum_{i=1}^{n} \underset{\sim}{r}_i)].$$

Clearly, the statistical properties of the scattered radiation are determined by those of incident radiation and density variations.

The calculation can be extended to the case of incoherent nonlinear scattering, recently reported by Terhune, et al.[13] Here, two photons in the pump modes $\underset{\sim}{k}_o$ and $\underset{\sim}{k}'_o$ are scattered by density variations into a single photon in the scattered mode $\underset{\sim}{k}$. The perturbing Hamiltonian is

$$\mathcal{H}_1 = -\sum_{i,k} [\ \underset{\sim}{E}^{(+)}_k (r_i) \cdot \underset{\approx}{p}^{(2)} : \underset{\sim}{E}^{(-)}_{k_o}\ (\underset{\sim}{r}_i)\ \underset{\sim}{E}^{(-)}_{k'_o} (\underset{\sim}{r}_i) + \text{Adjoint}]. \tag{35}$$

We find the same equations as Eqs. (30) - (34), except that $-i(2\pi\hbar\omega_{k_o}/\epsilon_{k_o} L^3)^{\frac{1}{2}}\ a_{k_o}\ \exp(i\underset{\sim}{k}_o \cdot \underset{\sim}{r})$ is now replaced by $-(2\pi\hbar/L^3)(\omega_{k_o}\omega_{k'_o}/\epsilon_{k_o}\epsilon_{k'_o})^{\frac{1}{2}}\ a_{k_o} a_{k'_o}\ \exp[i(\underset{\sim}{k}_o + \underset{\sim}{k}'_o)\cdot r]$, $\hat{e}_{k_o}$ by $\hat{e}_{k_o}\hat{e}_{k'_o}$, and $\underset{\approx}{p}$ by $\underset{\approx}{p}^{(2)}$. Thus, the statistical properties of the nonlinearly scattered radiation are fully described, and again, they depend on those of incident radiation and density variations in the medium. In particular, the intensity of the scattered radiation $< E^{(+)}_{sc.}(r,t)\; E^{(-)}_{sc.}(r,t) >$ is proportional to $< a^{\dagger}_{k_o} a^{\dagger}_{k_o}{}' a_{k_o} a_{k_o}{}' > \int_V d^3r\, d^3r' < N(\underset{\sim}{r},t) N(\underset{\sim}{r}',t) > \exp[i(\underset{\sim}{k}-\underset{\sim}{k}_o-\underset{\sim}{k}_o')\cdot(\underset{\sim}{r}-\underset{\sim}{r}')]$. Then, if $k_o = k_o'$, even the scattering intensity depends on the statistics of the pump modes, and is two times higher for chaotic than for coherent fields.

IV. Sum-Frequency Generation

The sum-frequency generation is closely related to incoherent nonlinear scattering discussed in the previous section. In fact, the same Hamiltonian applies to both cases. The difference is that here the density $N(\underset{\sim}{r},t)$ is constant throughout the medium. Therefore, the scattered radiation is non-vanishing only in the phase matched direction given by $\underset{\sim}{k} = \underset{\sim}{k}_o + \underset{\sim}{k}_o'$, and is called the coherent scattering. The calculation is however essentially the smae as the incoherent scattering case. Again, if the pump fields are very intense and hardly disturbed by the sum-frequency generation, then a_{k_o} and $a_{k_o'}$ can be treated as constant c numbers but subject to statistics, and the problem reduces to that of radiation by a prescribed current distribution.[6]

Thus, in the Heisenberg representation, the equation of motion for a_k is[14]

$$da_k/dt = -i\omega_k a_k(t) - (i/\hbar) f_k \, a_{k_o}(t) a_{k_o'}(t) \tag{36}$$

$$f_k = -NV(8\pi^3\hbar^3\omega_o\omega_o'\omega_k/\epsilon_{k_o}\epsilon_{k_o'}\epsilon_k L^9)^{-\frac{1}{2}} \hat{e}_k \cdot \underset{\approx}{p}^{(2)} : \hat{e}_{k_o}\hat{e}_{k_o'}$$

from which we obtain

$$a_k(t) = [a_k(0) - (i/\hbar) f_k a_{k_o} a_{k_o'} t] \exp[-i(\omega_o + \omega_o')t]. \tag{37}$$

The density matrix for the sum-frequency field is

$$\rho_k(t) = D(\alpha_k) \, \rho_k(0) \, D^{-1}(\alpha_k) \tag{38}$$

$$D(\alpha_k) = \exp[\, \alpha_k(a_{k_o}, a_{k_o'}, t) \, a_k^\dagger - \text{Adjoint}]$$

$$\alpha_k(a_{k_o}, a_{k_o'}, t) = (i/\hbar) \int_o^t dt \, f_k \, a_{k_o} a_{k_o'}$$

$$< (a_k^\dagger)^m (a_k)^n > = < (\alpha_k^*)^m (\alpha_k)^n >_{a_{k_o}, a_{k_o'}} \, .$$

From either Eq. (37) or (38), correlation fun ctions of a_k can be obtained in terms of correlation functions of a_{k_o} and $a_{k_o'}$. Therefore, the statistical properties of the sum-frequency field depend on the statistical properties of the pump fields. It is easily seen that for coherent pump fields, if $\rho_k(0) = |0><0|$, the n^{th}-order correlation function of the sum-frequency field is proportional to the $2n^{th}$-order correlation function of the pump fields. Therefore, measurements of the statistics of the sum-frequency or more conveniently, the second-harmonic output would yield direct information about the statistics of the pump fields. Beran, et al.[15] have proposed a practical means using the combined experimental arrangement of second-harmonic generation in nonlinear crystals and Young's double slit interference to measure the second-order correlation functions of the pump fields.

The sum-frequency output has the average photon number given by

$$< a_k^\dagger a_k > (t) = < a_k^\dagger a_k > (0) + (i/\hbar)[f_k^* < a_{k_o}^\dagger a_{k_o'}^\dagger a_k >(0) - f_k < a_k^\dagger a_{k_o} a_{k_o'} > (0)]$$

$$+ (2|f_k|^2 t^2/\hbar^2) < a_{k_o}^\dagger a_{k_o'}^\dagger a_{k_o} a_{k_o'} > (0) \qquad (39)$$

which shows explicitly that for $< a_k(0)> = 0$, the average sum-frequency output, or the rate of sum-frequency generation $d< a_k^\dagger a_k >(t)/dt$, is proportional to $< a_{k_o}^\dagger a_{k_o'}^\dagger a_{k_o} a_{k_o'} > (0)$. Therefore, in the second-harmonic generation where $k_o = k_o'$, the second-harmonic output depends on the initial statistical properties of the pump field, and is two times larger for a chaotic than for a coherent pump field.

In the above discussion, reaction of the sum-frequency generation on the pump fields have been neglected. When this is taken into account, the sum-frequency generation would depend on higher-order correlation function of the initial pump fields, and the calculation becomes more involved. Ducuing and Armstrong[16] have discussed the statistics of second-harmonic generation with appreciable depletion of pump power in the classical limit.

V. Parametric Amplification and Oscillation

Parametric conversion has recently received much attention. It is important because it leads to the realization of tunable optical oscillator.[17] More generally, it also describes stimulated Raman and Brillouin processes in which elementary boson excitations in the medium play the role of one of the photon modes.[18] Physically, parametric conversion is simply the inverse process of sum-frequency generation. Here however, the sum-frequency field is the pump mode and the others are the generated signal and idler modes.

The same perturbing Hamiltonian in Eq. (35) also describes the parametric process. In notations familiar for parametric amplification, it reads

$$\mathcal{H}_1 = -\sum_i \left[\underset{\sim}{E}_p^{(+)}(\underset{\sim}{r}_i) \cdot \underset{\approx}{P}^{(2)} : \underset{\sim}{E}_s^{(-)}(\underset{\sim}{r}_i)\underset{\sim}{E}_I^{(-)}(\underset{\sim}{r}_i) + \text{Adjoint}\right] \qquad (40)$$

where the subscripts p, s, and I denote pump, signal, and idler modes respectively. The Heisenberg equations of motion for a_s and $a_I^\dagger$ are

$$da_s/dt = -\omega_s a_s(t) - i\kappa a_p(t)\, a_I^\dagger(t)$$

$$da_I^\dagger/dt = i\omega_I a_I^\dagger(t) + i\kappa^* a_p^\dagger(t) a_s^\dagger(t)$$

$$\kappa = -NV(8\pi^3\hbar^3\omega_p\omega_s\omega_I/\epsilon_p\epsilon_s\epsilon_I L^9)^{\frac{1}{2}}\, \hat{e}_p \cdot \underset{\approx}{P}^{(2)*} : \hat{e}_s\, \hat{e}_I \quad . \qquad (41)$$

Again, we assume a pump field of high intensity unperturbed by the parametric process, so that a_k and $a_k^\dagger$ can be treated as constant c numbers but subject to statistical variation. The solution of Eq. (41) can be easily found.[19]

$$a_s(t) = \{a_s(0) \cosh[|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t] + [i\kappa a_p/|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}}] a_I(0) \sinh[|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t]\} \exp(-i\omega_s t)$$

$$a_I(t) = \{ a_I(0) \cosh[|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t] + [i\kappa a_p/|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}}] a_s(0) \sinh[|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t] \exp(-i\omega_I t) . \quad (42)$$

From the above expressions, correlation functions of all orders of a_k and a_I can be obtained. Note that we have the invariant condition $d(a_s^\dagger a_s)/dt = d(a_I^\dagger a_I)/dt$. The same is true in sum-frequency generation. The statistical properties of the generated signal and idler modes are thus fully described and shown to be dependent on the initial statistical properties of the pump field. In particular, for $< a_s a_I > (0) = 0$, we find the average number of photons in the signal out,

$$< a_s^\dagger a_s > (t) = \frac{1}{2} [< a_s^\dagger a_s > (0) - < a_I^\dagger a_I > (0) - 1] + \frac{1}{2} [< a_s^\dagger a_s > (0) + < a_I^\dagger a_I > (0) + 1] < \cosh[2|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t] > \quad (43)$$

where $< \cos h[2|\kappa|(a_p^\dagger a_p)^{\frac{1}{2}} t] >$ is apparently much larger for a chaotic than for a coherent pump field. Statistical properties of a parametric amplifier or oscillator have been discussed by Gordon et al.[19] They have, however, neglected the statistics of the pump mode. Their results therefore apply only to a coherent pump field.

For the present case, the density matrix for the generated fields is most conveniently obtained through the use of characteristic function.[19,20] Let us consider the density matrices for the signal and for the idler fields separately, $\rho_s(t) = Tr_I \rho_{s,I}(t)$ and $\rho_I(t) = Tr_s \rho_{s,I}(t)$. The corresponding characteristic functions are defined as[20]

$$\chi_s(\gamma,t) = Tr_{s,I}\{ \rho_{s,I}(t) \exp[\gamma a_s^\dagger(0)] \exp[-\gamma^* a_s(0)]\}$$
$$\chi_I(\gamma,t) = Tr_{s,I}\{ \rho_{s,I}(t) \exp[\gamma a_I^\dagger(0)] \exp[-\gamma^* a_I(0)]\} \tag{44}$$

which can be rearranged to give

$$\chi_s(\gamma,t) = Tr_{s,I} \{\rho_s(0)\, \rho_I(0) \exp[\gamma a_s^\dagger(t)] \exp[-\gamma^* a_s(t)]\}$$
$$\chi_I(\gamma,t) = Tr_{s,I} \{\rho_s(0)\, \rho_I(0) \exp[\gamma a_I^\dagger(t)] \exp[-\gamma^* a_I(t)]\}. \tag{45}$$

Explicit expressions of the characteristic functions can now be obtained by substituting the expressions of $a_s(t)$ and $a_I(t)$ of Eq. (42) and the initial distribution $\rho_s(0)$ and $\rho_I(0)$ into Eq. (45). The density matrices $\rho_s(t)$ and $\rho_I(t)$ are uniquely determined by the characteristic functions $\chi_s(r,t)$ and $\chi_I(\gamma,t)$.[21] If the Fourier transforms of $\chi_s(\gamma,t)$

and $\chi_I(\gamma,t)$ exist, then $\rho_s(t)$ and $\rho_I(t)$ can assume a P-representation.[20]

$$\rho_s(t) = \int d^2\alpha_s P_s(\alpha_s, a_p, t)|\alpha_s><\alpha_s|$$

$$P_s(\alpha_s, a_p, t) = (1/\pi^2)\int d^2\gamma\, \chi(\gamma, a_p, t)\exp(\alpha_s\gamma^* - \alpha_s^*\gamma) \tag{46}$$

with a similar expression for $\rho_I(t)$. If the pump field also has a P-representation, $\rho_p(t) = \int d^2\alpha_p P_p(\alpha_p)|\alpha_p><\alpha_p|$, we would find

$$P_s(\alpha_s,t) = (1/\pi^2)\int d^2\alpha_p d^2\gamma P_p(\alpha_p)\, \chi(\gamma, \alpha_p, t)\exp(\alpha_s\gamma^* - \alpha_s^*\gamma). \tag{47}$$

Gordon et al.[19] first used this method to find the density matrices for the signal output from a parametric oscillator. Mollow and Glauber[22] have given explicit expressions of the density matrices corresponding to various input distribution $\rho_s(0)$ and $\rho_I(0)$. They have also shown that a P-representation necessarily exists for the signal output after a critical time is reached, and that a non-negative P-function, which resembles a classical distribution, shows up at somewhat later times. The statistics of the pump field, however, have not been included in their treatment. The joint density matrix $\rho_{s,I}(t)$ can also be defined uniquely by a joint characteristic function $\chi_{s,I}(\eta,\zeta, t)$, or a corresponding Wigner distribution function as discussed by Mollow and Glauber in great details.[22]

By neglecting the depletion of pump power, we have not taken into account saturation of parametric amplification here. The general calculation considering the reaction of parametric conversion on the pump field would be extremely difficult.

VI. Multimode Problems

In all previous sections, we assumed that each field component, specified by, say, k and ω_k, consists of a single mode, as indicated explicitly in Eq. (6). The calculations can however be extended to the multimode case, even though it becomes somewhat more complex. Let us assume that there is a set of spatial modes for each field component of frequency ω_k. We have

$$\underset{\sim}{E}_k^{(+)}(\underset{\sim}{r}) = i(2\pi\hbar\omega_k)^{\frac{1}{2}} \sum_\lambda \underset{\sim}{u}^*_{k_\lambda}(r) a^\dagger_{k_\lambda} \quad . \tag{48}$$

The calculations in previous sections should then be modified accordingly.

Thus, for example, in the case of two-photon absorption or emission with $k = \ell$, we find[1]

$$\partial < \sum_\lambda a^\dagger_{k_\lambda} a_{k_\lambda} >/\partial t = 4\gamma \int_V d^3 r N(r) [(\rho^o_{2A} - \rho^o_{1A}) < E_k^{(+)} E_k^{(+)} E_k^{(-)} E_k^{(-)} > (\underset{\sim}{r}, t)$$

$$+ \rho^o_{2A} (2 < E_k^{(+)} E_k^{(-)} > (\underset{\sim}{r}, t) + 1)]$$

$$\gamma = [|\zeta^{(2)}|^2 g(\omega_k + \omega_\ell)/2\hbar^2] \quad . \tag{49}$$

In the case of parametric oscillation, we have approximately

$$< E_s^{(+)} E_s^{(-)} > (r,t) = < E_s^{(+)} E_s^{(-)} > (\underset{\sim}{r}, 0) < \cosh^2[|\kappa'|(E_p^{(+)} E_p^{(-)})^{1/2} t] >$$

$$+ < E_I^{(-)} E_I^{(+)} > (\underset{\sim}{r}, 0) < \sinh^2[|\kappa'|(E_p^{(+)} E_p^{(-)})^{1/2} t] >$$

$$\kappa' = (2\pi\hbar\omega_p/\epsilon_p L^3)^{-1/2} \kappa \tag{50}$$

assuming $< E_s^{(+)} E_I^{(+)} > (0) = 0$. In all cases, the magnitudes of correlation functions depend on both the mode structure and the statistics of the fields. For stationary fields with large number of modes, we have[6]

$$< (E_k^{(+)})^n (E_k^{(-)})^n > = \int d^2 \mathcal{E}_k W(\mathcal{E}_k) |\mathcal{E}_k|^{2n}$$

$$= n! < E_k^{(+)} E_k^{(-)} >^n \qquad (51)$$

where

$$W(\mathcal{E}_k) = \exp [- |\mathcal{E}_k|^2 / < E_k^{(+)} E_k^{(-)} >] / \pi < E_k^{(+)} E_k^{(-)} > .$$

Equation (51) actually holds for arbitrary chaotic fields independent of the number of modes, as followed directly from Eq. (25). This is expected since physically a stationary field composed of many uncorrelated modes is equivalent to a chaotic field. Thus, for chaotic fields, the rate of a nonlinear optical process is unchanged in going from the single-mode to the multimode limit. For coherent fields, however, the rate increases as a result of its dependence on higher-order correlation functions and Eq. (51). For coherent fields with many correlated modes, the rate increase could be much higher. The latter case may actually happen in nonlinear optical experiments using laser beams, and is possibly the cause of the observed anomalous gain in stimulated Raman scattering in non-self-focusing materials.

VII. Traveling Wave Problems

In many experiments, we are interested in the change of statistical properties of the beam as it propagates through a nonlinear medium. For steady-state propagation, the statistical properties of the beam should be functions of position only, independent of time. Therefore, a proper description of such problems requires existance of a local statistical average which changes with position. This can be achieved by using localized operators and a localized density matrix.

Let $\hat{z}$ be the direction of propagation. Since the field amplitudes depend only on z, the vector potential for a plane wave can be written as

$$A(z,t) = c \sum_k (\hbar/2\omega_k \epsilon_k L^3)^{\frac{1}{2}} \{\psi_k(z)\exp(-i\omega_k t) + \psi_k^\dagger(z)\exp(i\omega_k t)\}$$

$$\psi_k(z) = b_k(z) \exp(ikz) \tag{52}$$

$$[b_k(z), b_{k'}^\dagger(z)] = \delta_{kk'} .$$

Here, the localized annihilation and creation operators b and $b^\dagger$ are defined under the assumption that $< (b_k^\dagger)^m (b_k)^n >$ does not vary appreciably in a distance d large compared with the wavelength,[23] and that $k \cong 2\pi n/d$, where n is an integer for free fields, b_k is independent of z. Then, the field in a volume of length d, centered at z, can be quantized with a corresponding localized photon number operator.[24]

$$\hat{n}(z) = (\mathcal{A} d / L^3) \sum_k \tilde{b}_k^\dagger(z)\, b_k(z) \tag{53}$$

where $\mathcal{A}$ is the cross-sectional area of the beam. We now define the localized Hamiltonian and momentum operators as

$$\mathcal{H}(z_o) = (L^3/d) \int_{z_o - d/2}^{z_o + d/2} H(z)\, dz \tag{54}$$

$$\underset{\sim}{\mathcal{P}}(z_o) = \hat{z}\, \mathcal{H}(z_o)/c\epsilon^{-\frac{1}{2}}$$

where $H(z)$ is the Hamiltonian density at z. Note that $\mathcal{H}(z_o)$ has the same form as that of a cavity except that a and $a^\dagger$ are replaced by $b(z_o)$ and $b^\dagger(z_o)$. The momentum operator plays the role of a translation operator.

$$dE^{(\pm)}(z)/dz = (1/i\hbar)[\mathcal{P}(z), E^{(\pm)}(z)]. \tag{55}$$

Accordingly, the unitary translation operator is

$$U(z, z_o) = \exp[(i/\hbar) \int_{z_o}^{z} \mathcal{P}(z) dz]_+ \tag{56}$$

where the space-ordered product $\{\ \}_+$ has a similar definition as the time-ordered product. Fields at different spatial points are then connected by the unitary transformation

$$E^{(\pm)}(z,t) = U^{-1}(z,z_o)E^{(\pm)}(z_o,t)U(z,z_o). \quad (57)$$

Thus, field at arbitrary point can be found in terms of the field at the boundary. In fact, the operator equations (53) yields the same field amplitude equation as in the classical description. For example, in sum frequency generation, Eq. (55) gives

$$dE_k^{(-)}(z)/dz = ikE_k^{(-)}(z) + [i2\pi\omega_k/c\epsilon_k^{\frac{1}{2}}(z)]N(z)\hat{e}_k\cdot \underset{\approx}{p}^{(2)}\cdot\hat{e}_{k_o}\hat{e}_{k'_o}E_{k_o}^{(-)}(z)E_{k'_o}^{(-)}(z) \quad (58)$$

We also define a localized density matrix $\rho(z)$ as describing an ensemble of photon systems which has the statistical properties of fields at z. The density matrices at different spatial points are also connected by the unitary transformation

$$\rho(z) = U(z,z_o)\rho(z_o)\,U^{-1}(z,z_o). \quad (59)$$

The equation of motion for $\rho(z)$ is

$$\partial\rho(z)/\partial z = (1/i\hbar)[\rho(z), \mathcal{P}(z)]\;. \quad (60)$$

Correlation functions of fields are given by

$$< E^{(+)}(z_1,t_1)\ldots E^{(+)}(z_n,t_n)E^{(-)}(z_{n+1},t_{n+1})\ldots E^{(-)}(z_{2n},t_{2n}) >$$

$$= \mathrm{Tr}\{\rho(0)E^{(+)}(z_1,t_1)\ldots E^{(+)}(z_n,t_n)E^{(-)}(z_{n+1},t_{n+1})\ldots E^{(-)}(z_{2n},t_{2n})\}$$

$$\overset{\uparrow}{=} \mathrm{Tr}\,\{\rho(z)E^{(+)}(0,t_1)\ldots E^{(+)}(0,t_n)E^{(-)}(0,t_{n+1})\ldots E^{(-)}(0,t_{2n})\}$$

for $z = \ldots = z_{2n}$. (61)

With the help of these localized operators, the calculations now become exactly the same as the calculations for cavity problems with t replaced by $z\epsilon^{1/2}/c$ as one would expect from the classical wave description. We can also imagine a thin slab as a cavity, in which the photon fields are quantized, propagating in a medium. The fields in the slab interact with the medium for a time t, while the slab travels for a distance $z = ct/\sqrt{\epsilon}$. The statistical properties of the fields in the slab at z can therefore be obtained from the results of calculation for a cavity in which the fields interact with the medium for a time $t = z\epsilon^{1/2}/c$. In the above discussion, we have deliberately avoided the question of reflection and transmission at the boundaries of the medium. To describe the propagation problems fully, a quantum statistical treatment of reflection and transmission would be important. A more rigorous treatment of the travelling wave problems is to treat each photon as a wave packet and construct creation and annihilation operators for wave packets. The technique has been developed in quantum theory of transport in solid state physics,[25] but the calculation in practice becomes much more difficult.

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