Quantum Statistics of Nonlinear Optics*

Y. R. SHEN†
Physics Department, University of California, Berkeley, California
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Nonlinear interaction of light with matter is described from the quantum-statistical point of view. The cases of two-photon absorption, Raman transition, sum-frequency generation, parametric amplification, and incoherent scattering are discussed. It is shown that the nonlinear optical effects depend strongly on the statistical properties of the light fields. The rate of nonlinear absorption, generation, and amplification is higher for chaotic than for coherent, and also higher for multimode than for single-mode pump fields. Measurements of the statistics of the output fields may yield information about the statistics of the input fields and the properties of the medium.

I. INTRODUCTION

THE quantum statistical properties of light from various sources have recently been extensively investigated.[1,2] However, the question whether interaction of light with matter would change statistical properties of light fields has seldom been raised. The purpose of this paper is to extend the quantum-statistical description to the case of light fields after interacting with a medium. Emphasis is on the effect of nonlinear interaction of light with the medium.

It is usually assumed in the literature that statistical properties of a light beam remain unchanged in traversing a medium if the response of the medium to the light fields is linear. This assumption is certainly a valid one for a nonabsorbing medium, since the linear interaction of light with the medium cannot disturb the probability distribution of photons in their number states (if the disturbance due to incoherent scattering can be neglected. See Sec. III). Only their spatial distribution is changed through the interaction. Let the vector potential be written in the usual form[3]

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

where $a^\dagger$ and a are the creation and the annihilation operators, respectively, for the kth mode. (The subindices indicating the polarization of the fields are being omitted.) The spatial function $\mathbf{u}_k(\mathbf{r})$ is a normalized eigenfunction of the differential equation

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

where $\epsilon_k(\mathbf{r})$ is the linear dielectric constant at frequency ω_k.[4] From Sturm-Louivelle theory, the orthonormality condition gives

$$\int(\epsilon_k\epsilon_l)^{1/2}\mathbf{u}_k(\mathbf{r})\mathbf{u}_l^*(\mathbf{r})d^3r=\delta_{kl}. \quad (3)$$

Then, the Hamiltonian of the fields in the presence of the linear nonabsorbing medium reduces to the familiar form

$$\mathcal{H}=\sum_k \hbar\omega_k(a_k^\dagger a_k+\tfrac{1}{2}). \quad (4)$$

Thus, the photon statistics of the fields is not changed except that the spatial distribution, described by $\mathbf{u}_k(\mathbf{r})$, is now different from the vacuum case.

This is not quite true if the medium is lossy. An obvious example is the case where originally there are exactly n_k photons present in such a medium. After the absorption has been switched on for a finite length of time, the photon system has finite probabilities in the occupation number states $|n_k\rangle$, $|(n-1)_k\rangle$, $|(n-2)_k\rangle$, etc. The statistical properties of the photon system have clearly been changed. Assume that the medium has an electric-dipole transition between atomic states $|\psi_2\rangle$ and $|\psi_1\rangle$ with frequency separation ω_{21}, which coincides with the photon frequency of the kth mode. The single-photon absorption can be described by the interaction Hamiltonian

$$\mathcal{H}_{\rm int}=\sum_i \{\xi c_{2i}^\dagger c_{1i}\mathbf{E}_k^{(-)}(\mathbf{r}_i)+\xi^* c_{2i}c_{1i}^\dagger\mathbf{E}_k^{(+)}(\mathbf{r}_i)\}. \quad (5)$$

Here, c_{1i}, c_{2i}, $c_{1i}^\dagger$, and $c_{2i}^\dagger$ are creation and annihilation operators for the ith atom in states 1 and 2, respectively. ξ is the electric-dipole matrix element for the transition. The positive-frequency part of the electric field at the ith atom is given by

$$\mathbf{E}_k^{(+)}(\mathbf{r}_i)=\mathbf{E}_k^{(-)}(\mathbf{r}_i)^\dagger=i(2\pi\hbar\omega_k)^{1/2}\mathbf{u}_k^*(r_i)a_k^\dagger. \quad (6)$$

In the interaction representation, the equation of

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† Alfred P. Sloan Research Fellow.

[1] R. Glauber, *Quantum Optics and Electronics*, edited by C. DeWitt *et al.* (Gordon and Breach Science Publishers, Inc., New York, 1965).

[2] See, for example, Abstracts on Second Rochester Conference on Coherence and Quantum Optics, Rochester (unpublished).

[3] See, for example, W. Heitler, *Quantum Theory of Radiation* (Clarendon Press, Oxford, England, 1954), p. 54.

[4] In general the coherent linear response of a medium to the fields can be described completely by a generalized linear dielectric tensor $\boldsymbol{\varepsilon}_k(\mathbf{r})$; see Y. R. Shen, Phys. Rev. **133**, A511 (1964). In this paper, we shall assume that $\boldsymbol{\varepsilon}_k(\mathbf{r})$ is a scalar, and that all fields are linearly polarized.

motion for the density matrix ρ of the composite system is

$$i\hbar\partial\rho/\partial t=[\mathcal{H}_{\rm int}(t),\rho(t)]. \quad (7)$$

Iteration of ρ for small increment of t in the above equation gives[5]

$$\partial\rho(t_0+t)/\partial t=(1/i\hbar)[\mathcal{H}_{\rm int}(t_0+t),\rho(t_0)] +(-1/\hbar^2)\int_{t_0}^{t_0+t}[\mathcal{H}_{\rm int}(t_0+t), \times[\mathcal{H}_{\rm int}(t'),\rho(t_0)]]dt'+\cdots. \quad (8)$$

We now assume that the thermal equilibrium of the atomic system is not disturbed by photon fields. The density matrix can then be written as $\rho(t)=\rho_F(t)\times\prod_i\rho_{Ai}(0)$, where ρ_F and ρ_{Ai} are density matrix operators for the photon system and for the ith atom, respectively. This is known as the irreversible approximation.[6] We have, with the same approximation as used in the ordinary time-dependent perturbation calculation,[5,7] namely, $\hbar/|\mathcal{H}_{\rm int}|>t\gg 1/(\text{linewidth})$,

$$\begin{aligned}\partial\rho_F/\partial t&={\rm Tr}_A(\partial\rho/\partial t)\\&=-\beta[(a_k^\dagger a_k\rho_F-2a_k\rho_F a_k^\dagger+\rho_F a_k^\dagger a_k)\rho_{1A}{}^0\\&\quad+(a_k a_k^\dagger\rho_F-2a_k^\dagger\rho_F a_k+\rho_F a_k a_k^\dagger)\rho_{2A}{}^0], \quad (9)\end{aligned}$$

where

$$\begin{aligned}\beta&=\sum_i \omega_k|\xi|^2|\mathbf{u}_k(\mathbf{r}_i)|^2 g(\omega_k)/4\hbar\\&\approx[\pi\omega_k|\xi|^2 g(\omega_k)/\hbar]\int_V d^3r|\mathbf{u}_k(\mathbf{r})|^2N(\mathbf{r}).\end{aligned}$$

Here $g(\omega_k)$ is the line shape function, $N(\mathbf{r})$ is the density of atoms at the position $\mathbf{r}$, and $\rho_{1A}{}^0$ and $\rho_{2A}{}^0$ are the thermal populations for the two atomic states. The integration extends over the volume of the medium. The constant β is related to the absorption coefficient. If $\rho_F(t)$ is known, statistical properties of the fields, such as temporal and spatial coherence, can readily be determined. From Eq. (9), one obtains

$$\begin{aligned}\partial\langle a_k\rangle/\partial t&=-\beta(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k\rangle,\\ \partial\langle a_k^\dagger a_k\rangle/\partial t&=-2\beta(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k^\dagger a_k\rangle+2\beta\rho_{2A}{}^0. \quad (10)\end{aligned}$$

The last term in the above equation corresponds to spontaneous emission.

Equation (9) governs the change of statistical properties of the photon system in the single-photon absorption process. In particular, at zero temperature, if initially the photon system is in a coherent state,[1] $\rho_F(t_0)=|\alpha_k\rangle\langle\alpha_k|$, then it is easily shown that

$$\begin{aligned}\rho_F(t_0+\Delta t)&=\{\sum_{n_k}[1-\beta\Delta t(n_k-|\alpha_k|^2)](\alpha_k^{2n_k}/n_k!)^{1/2}\\&\quad\times\exp(-\tfrac{1}{2}|\alpha_k|^2)|n_k\rangle\}\sum_{m_k}\langle m_k|\\&\quad\times[1-\beta\Delta t(m_k-|\alpha_k|^2)(\alpha_k^{2m_k}/m_k!)^{1/2}\\&\quad\times\exp(-\tfrac{1}{2}|\alpha_k|^2)\}\underset{\Delta t\to 0}{\longrightarrow}|\alpha_k\exp(-\beta\Delta t)\rangle\\&\quad\times\langle\alpha_k\exp(-\beta\Delta t)|\,;\end{aligned}$$

$$\rho_F(t_0+t)=|\alpha_k\exp(-\beta t)\rangle\langle\alpha_k\exp(-\beta t)|. \quad (11)$$

This shows that a coherent photon system remains coherent although the field amplitude decreases exponentially with time. More generally, if the initial photon field can be described by the P representation,[1]

$$\rho_F(t_0)=\int d^2\alpha_k P(\alpha_k)|\alpha_k\rangle\langle\alpha_k|,$$

one would get

$$\rho_F(t_0+t)=\int d^2\alpha_k P(\alpha_k)|\alpha_k\exp(-\beta t)\rangle \times\langle\alpha_k\exp(-\beta t)|. \quad (12)$$

The statistical properties of the fields are being changed in a rather trivial way, since it is simply a translation of the distribution $P(\alpha_k)$ in the α_k space.

No such simple solution exists if the equilibrium temperature of the atomic system is finite, since the spontaneous emission now comes into play. Consequently, the coherent properties of a beam will be disturbed in passing through the absorbing medium. The disturbance is, of course, small if the spontaneous emission process can be neglected in comparison with either the stimulated absorption or emission. This is certainly true for light beams in a medium at room temperature.

The same approach can be applied to the case of multiphoton transitions. Again, since the photon distribution can be disturbed by the transitions, statistical properties of the photon system are changed. The case of two-photon transitions, which includes Raman transitions, will be discussed in Sec. II. In general, even if the medium is not lossy, statistical properties of light are changed by nonlinear interaction of light with a medium, although the disturbance might be small for weak interaction. The nonlinear interaction couples different photon modes and leads to energy transfer between the modes. Photons in some modes may be annihilated, while those in other modes created, and hence the photon distribution is disturbed. Oftentimes the rate of energy transfer between the modes depends on the statistical properties of the light fields, usually higher for chaotic than for coherent sources.

For investigation of properties of a medium, incoherent scattering has long been a useful tool. Statistics

[5] C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row Publishers Inc., New York, 1963), p. 127.
[6] F. Bloch, Phys. Rev. **102**, 104 (1956).
[7] See, for example, L. I. Shiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), p. 189.

is particularly important in this case for analyzing the results of experiments. In Sec. III, linear and nonlinear incoherent scattering are discussed. As is expected, the scattered radiation depends on the statistical nature of both the incident beam and the fluctuations in the medium. For nonlinear optics, one is perhaps more interested in coherent scattering. We shall discuss in Secs. IV and V two important cases, sum-frequency generation and parametric amplification, respectively. In all cases, there are one or more pump fields present. We shall not concern ourselves too much about how the statistical properties of the pump modes change. Instead, we are interested in finding the statistical properties of the generated modes, and the rate of generation as a function of the statistical nature of the pump modes. Conversely, from the statistical properties of the generated modes or the rate of generation, one could obtain some information about the statistical nature of the pump modes.

It must be noted that in our discussion of single-photon absorption, we have assumed a bounded system for the photon fields. This type of treatment is most conveniently applied to the case of a cavity; photons are neither coming into nor going out of the cavity. In principle, the same treatment can be applied to problems of light propagation in a medium. In practice, it is indeed successful in dealing with incoherent scattering (see Sec. III), but for coherent scattering, it becomes extremely difficult. Rigorously, the latter case should perhaps be treated by the method of many-body transport theory.[8] However, imagine an infinite medium and a box of finite volume in which the photon fields are quantized.[9] This box of photons interacts with the medium for a time t, as its center modes in the z direction from z_1 to z_1+ct, where c is the light velocity in the medium. The resultant change of statistical properties of fields in the box can now be calculated using the cavity treatment. A more general treatment of the propagation problems is given in the Appendix. For steady-state propagation, it is shown that the results are essentially the same as in the cavity case with t replaced by $-z/c$, as one would expect.

II. TWO-PHOTON ABSORPTION AND RAMAN TRANSITIONS

The calculation for two-photon absorption is essentially similar to that for single-photon absorption, except that the mathematics becomes more complicated. Here, an atom makes transition from the state $|\psi_1\rangle$ to the state $|\psi_2\rangle$ by absorbing one photon in the kth mode and another in the lth mode. The interaction Hamiltonian is

$$\mathcal{H}_{\text{int}}=\sum_i \{\eta c_{2i}^\dagger c_{1i}\mathbf{E}_k^{(-)}(\mathbf{r}_i)\mathbf{E}_l^{(-)}(\mathbf{r}_i)+\text{adjoint}\}, \quad (13)$$

where η is the matrix element for the two-photon transitions.[10,11] Using the same procedure as in the case of single-photon absorption, one can find that the density matrix ρ_F for the photon system obeys the equation

$$\partial\rho_F/\partial t=-\beta^{(2)}[(a_k^\dagger a_l^\dagger a_k a_l\rho_F-2a_k a_l\rho_F a_k^\dagger a_l^\dagger +\rho_F a_k^\dagger a_l^\dagger a_k a_l)\rho_{1A}{}^0+(a_k a_l a_k^\dagger a_l^\dagger \rho_F-2a_k^\dagger a_l^\dagger \rho_F a_k a_l +\rho_F a_k a_l a_k^\dagger a_l^\dagger)\rho_{2A}{}^0], \quad (14)$$

with

$$\beta^{(2)}\cong[2\pi^2\omega_k\omega_l|\eta|^2 g(\omega_k+\omega_l)]\int_V d^3r \times N(r)|\mathbf{u}_k(\mathbf{r})|^2|\mathbf{u}_l(\mathbf{r})|^2.$$

The above equation governs the change of statistical properties of the photon system in the two-photon absorption process. The solution of Eq. (14) is difficult. However, it is clear that if the absorption is large, the statistical properties of the fields will be appreciably disturbed. A coherent beam will no longer be coherent after interacting with the medium.

From Eq. (14), we obtain

$$\partial\langle a_k\rangle/\partial t=-\beta^{(2)}(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k a_l^\dagger a_l\rangle +\beta^{(2)}\rho_{2A}{}^0\langle a_k\rangle, \quad (15a)$$

$$\partial\langle a_k^\dagger a_k\rangle/\partial t=\partial\langle a_l^\dagger a_l\rangle/\partial t, =-2\beta^{(2)}(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k^\dagger a_k a_l^\dagger a_l\rangle +2\beta^{(2)}\rho_{2A}{}^0\langle(a_k^\dagger a_k+a_l^\dagger a_l+1)\rangle. \quad (15b)$$

In Eq. (15) the last term, which is proportional to the population $\rho_{2A}{}^0$ in the excited state, arises because $a^\dagger$ and a do not commute. It can be regarded as the spontaneous emission term in the two-photon absorption process. Assume that the two photon modes are independent initially. Then, as long as the photon distribution is not appreciably disturbed by the absorption, we can write

$$\langle a_k^\dagger a_k a_l^\dagger a_l\rangle\cong\langle a_k^\dagger a_k\rangle\langle a_l^\dagger a_l\rangle=\langle n_k\rangle\langle n_l\rangle.$$

The average rate of two-photon absorption depends on the average numbers of photons in the kth and the lth modes. However, if $k=l$, one would find

$$\partial\langle a_k\rangle/\partial t=-2\beta^{(2)}(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k^\dagger a_k a_k\rangle +4\beta^{(2)}\rho_{2A}{}^0\langle a_k\rangle,$$

$$\partial\langle a_k^\dagger a_k\rangle/\partial t=-4\beta^{(2)}(\rho_{1A}{}^0-\rho_{2A}{}^0)\langle a_k^\dagger a_k^\dagger a_k a_k\rangle +4\beta^{(2)}\rho_{2A}{}^0(2\langle a_k^\dagger a_k\rangle+1). \quad (16)$$

Here the absorption rate with $\rho_{2A}{}^0=0$ is twice as much as that of Eq. (15b) with $k=l$, since two photons in the same mode are being absorbed simultaneously. With the spontaneous-emission term being neglected, the average absorption rate is now proportional to the second-order correlation function $\langle a_k^\dagger a_k^\dagger a_k a_k\rangle$, and therefore depends

[8] See, for example, D. Ter Haar, Rept. Progr. Phys. 24, 304 (1961).

[9] The length of the box can be taken as the product of the light velocity and the response time of the photon detector.

[10] M. Göppert-Mayer, Ann. Physik 9, 273 (1931).

[11] P. Lambropoulos, C. Kikuchi, and R. K. Osborn, Phys. Rev. 144, 1081 (1966).

on the statistical nature of the fields.[11] It is two times higher for chaotic than for coherent sources, since[1]

$$\langle a^\dagger a^\dagger aa\rangle_{\text{chaotic}}=2(\langle a^\dagger a\rangle)^2,$$
$$\langle a^\dagger a^\dagger aa\rangle_{\text{coherent}}=(\langle a^\dagger a\rangle)^2. \quad (17)$$

Physically, a chaotic source has more irregularities in its intensity distribution than a coherent source. In a nonlinear response proportional to higher-order correlation functions of $a^\dagger$ and a, the peaks in the irregularities are weighted more strongly than the valleys. Consequently, the average nonlinear response from a source of more irregularities appears to be greater. It must be noted that if the absorption is appreciable, then $\langle a_k^\dagger a_k a_l^\dagger a_l\rangle(t)$ in Eq. (15) also depends on higher-order correlation functions of the initial field, as is seen by iteration on Eq. (15). A similar discussion can be given to the case where the fields contain many modes.

Assume that at each frequency there is a set of spatial modes, and for simplicity the fields consist of only two frequencies, ω_k and ω_l. The electric field at the position $\mathbf{r}$ is now given by

$$\mathbf{E}(\mathbf{r})=\mathbf{E}_k(\mathbf{r})+\mathbf{E}_l(\mathbf{r}),$$
$$\mathbf{E}_k^{(+)}(\mathbf{r})=\mathbf{E}_k^{(-)}(\mathbf{r})^\dagger=i(2\pi\hbar\omega_k)^{1/2}\sum_\lambda \mathbf{u}_{k\lambda}^*(\mathbf{r})a_{k\lambda}^\dagger. \quad (18)$$

By carrying out similar calculations as in the single-mode case, one would find at zero temperature

$$\partial\langle\sum_\lambda a_{k\lambda}^\dagger a_{k\lambda}\rangle/\partial t=2\gamma\int_V d^3r$$
$$\times N(r)\langle E_k^{(+)}E_k^{(+)}E_l^{(-)}E_l^{(-)}\rangle,$$
$$\gamma=[|\eta|^2 g(\omega_k+\omega_l)/2\hbar^2], \quad (19)$$

assuming, for simplicity, that all fields are polarized in the same direction. If $\omega_k=\omega_l$, Eq. (19) becomes

$$\partial\langle\sum_\lambda a_{k\lambda}^\dagger a_{k\lambda}\rangle/\partial t=-4\gamma\int_V d^3r$$
$$\times N(r)\langle E_k^{(+)}E_k^{(+)}E_k^{(-)}E_k^{(-)}\rangle. \quad (20)$$

Using Glauber's P representation and the quasiprobability distribution for the field amplitude $\mathcal{E}_k$,[1] we can write for $t=0$

$$\langle E_k^{(+)}E_k^{(+)}E_k^{(-)}E_k^{(-)}\rangle$$
$$=\int d^2\mathcal{E}_k W(\mathcal{E}_k)|\mathcal{E}_k(\alpha,\mathbf{r})|^4. \quad (20\text{a})$$

Then, if $N(\mathbf{r})=$constant, and $W(\mathcal{E}_k)\geq 0$, one would have a higher initial absorption rate in the multimode case than in the single-mode case since

$$\int_V d^3r|\mathcal{E}_k(\alpha,r)|^4/V>\left[\int_V d^3r|\mathcal{E}_k(\alpha,r)|^2/V\right]^2.$$

The discussion on two-photon absorptions can be applied with slight modification to Raman transitions between localized states. Here, instead of two photons being absorbed in a transition, one photon is now emitted, while the other is absorbed. Thus, for Raman transitions, the interaction Hamiltonian in Eq. (13) should be changed into the form

$$\mathcal{H}_{\text{int}}=\sum_i\{\eta_R c_{2i}^\dagger c_{1i}\mathbf{E}_k^{(-)}(\mathbf{r}_i)\mathbf{E}_s^{(+)}(\mathbf{r}_i)+\text{adjoint}\}. \quad (21)$$

In the single-mode case, the density matrix for the photon system becomes

$$\partial\rho_F/\partial t=-\beta_R[(a_k^\dagger a_s a_k a_s^\dagger\rho_F-2a_k a_s^\dagger\rho_F a_k^\dagger a_s$$
$$+\rho_F a_k^\dagger a_s a_k a_s^\dagger)\rho_{1A}^0+(a_k a_s^\dagger a_k^\dagger a_s\rho_F-2a_k^\dagger a_s\rho_F a_k a_s^\dagger$$
$$+\rho_F a_k a_s^\dagger a_k^\dagger a_s)\rho_{2A}^0], \quad (22)$$

where β_R has the same form as in Eq. (14). From the above equation, we find the average rate of Stokes photon generation or the pump photon absorption[12];

$$\partial\langle a_s^\dagger a_s\rangle/\partial t=-\partial\langle a_k^\dagger a_k\rangle/\partial t$$
$$=2\beta_R(\rho_{1A}^0-\rho_{2A}^0)\langle a_k^\dagger a_k a_s^\dagger a_s\rangle$$
$$+2\beta_R[\langle a_k^\dagger a_k\rangle\rho_{1A}^0-\langle a_s^\dagger a_s\rangle\rho_{2A}^0]. \quad (23)$$

The first term in Eq. (23) corresponds to stimulated Stokes emission, whereas the last term corresponds to spontaneous emission. The latter appears as a noise source and is responsible for the self-generation of the Stokes field. If the pump field is of high intensity and is not depleted appreciably in the Stokes generation, we can treat a_k and $a_k^\dagger$ as c numbers in the approximation and $\rho_F(t)=\rho_k(0)\rho_s(t)$, where ρ_k and ρ_s are the density matrices for the pump and the Stokes fields, respectively. From Eq. (22), we get

$$\partial\text{Tr}_s[\rho_s(t)a_s^\dagger a_s]/\partial t$$
$$=2\beta_R[(\rho_{1A}^0-\rho_{2A}^0)a_k^\dagger a_k-\rho_{2A}^0]$$
$$\times\text{Tr}_s[\rho_s(t)a_s^\dagger a_s]+2\beta_R a_k^\dagger a_k\rho_{1A}^0. \quad (24)$$

The solution of the above equation gives

$$\langle a_s^\dagger a_s\rangle(t)=\text{Tr}\rho_k(0)\{[\text{Tr}_s(\rho_s(0)a_s^\dagger a_s)+A/B]$$
$$\times\exp[B(a_k^\dagger,a_k)t]-A/B\},$$
$$B(a_k^\dagger,a_k)=[(\rho_{1A}^0-\rho_{2A}^0)a_k^\dagger a_k-\rho_{2A}^0]2\beta_R, \quad (25)$$
$$A(a_k^\dagger,a_k)=2\beta_R a_k^\dagger a_k\rho_{1A}^0.$$

By expanding $\exp(Bt)$ into power series, it is seen that $\langle a_s^\dagger a_s\rangle(t)$ is a function of the nth order correlation functions of $a_k^\dagger$ and a_k. Therefore, the Stokes generation must depend strongly on the statistical properties of the pump field. In particular, for a coherent pump field we have, assuming $\rho_{2A}^0\ll(\rho_{1A}^0-\rho_{2A}^0)a_k^\dagger a_k$,

$$\langle a_s^\dagger a_s\rangle(t)=[\langle a_s^\dagger(0)a_s(0)\rangle+(1-\rho_{2A}^0/\rho_{1A}^0)]$$
$$\times\exp[2\beta_R(\rho_{1A}^0-\rho_{2A}^0)\langle a_k^\dagger a_k\rangle t]-(1-\rho_{2A}^0/\rho_{1A}^0),$$

[12] R. W. Hellwarth, Phys. Rev. 130, 1852 (1963).

but for a chaotic pump field, since

$$\langle (a_k^\dagger)^n (a_k)^n \rangle = n!(\langle a_k^\dagger a_k \rangle)^n,$$

we have

$$\langle a_s^\dagger a_s\rangle(t) = \{[\langle a_s^\dagger(0)a_s(0)\rangle + (1-\rho_{2A}^0/\rho_{1A}^0)]/ [1-2\beta_R(\rho_{1A}^0-\rho_{2A}^0)\langle a_k^\dagger a_k\rangle t]\} - (1-\rho_{2A}^0/\rho_{1A}^0).$$

Clearly, the average Stokes generation by chaotic pumps is much more effective than that by coherent pumps.

The multimode case in Raman transitions is somewhat complicated. For simplicity, we assume a uniform medium which fills up the entire volume of quantization. Assume also a set of spatial modes associated with each frequency, or a band of frequencies with a bandwidth much smaller than the Raman linewidth. Then, if the pump field is not highly depleted, we can show, for $\rho_{2A}^0=0$,

$$\partial \operatorname{Tr}_s[\rho_s(t)E_s^{(+)}E_s^{(-)}]/\partial t = 2\gamma_R\{E_k^{(+)}E_k^{(-)}[\operatorname{Tr}_s(\rho_s(t)E_s^{(+)}E_s^{(-)}) + (\hbar\omega_s/2)\sum_\lambda |u_{s\lambda}|^2]\}, \quad (26)$$

$$\gamma_R = [\pi N\omega_s|\eta|^2 g(\omega_l-\omega_s)/\hbar],$$

assuming all fields to be polarized in the same direction. In deriving Eq. (26), we have used the approximation

$$\sum_\lambda u_{k\lambda}^*(\mathbf{r})u_{k\lambda}(\mathbf{r}) \approx \delta(\mathbf{r}-\mathbf{r}'),$$

where the summation is over modes at the frequency ω_k. This approximation is equivalent to relaxation of the momentum matching condition in the Raman transitions.[13] The solution of Eq. (26) gives

$$\langle E_s^{(+)}E_s^{(-)}\rangle(r,t) = [\langle E_s^{(+)}E_s^{(-)}\rangle(r,0)+S(r)] \times\langle \exp(2\gamma_R E_k^{(+)}E_k^{(-)}t)\rangle - S(r), \quad (27)$$

$$S(r) = (\hbar\omega_s/2)\sum_\lambda |u_{s\lambda}(r)|^2.$$

If the quantity in the square brackets is independent of r, then $\langle E_s^{(+)}E_s^{(-)}\rangle(t)$ can be regarded as the average Stokes intensity in the volume. Since the magnitude of $\langle\exp(2\gamma_R E_k^{(+)}E_k^{(-)}t)\rangle$ is usually larger for multimodes than for a single mode, the average Stokes intensity should be higher for the multimode case. In the quasi-probability distribution, we have

$$\langle \exp(2\gamma_R E_k^{(+)}E_k^{(-)}t)\rangle = \int d^2\mathcal{E}_k W(\mathcal{E}_k)\exp(2\gamma_R|\mathcal{E}_k|^2 t).$$

For stationary fields with large numbers of modes,[1]

$$W(\mathcal{E}_k) = \exp[-|\mathcal{E}_k|^2/\langle E_k^{(+)}E_k^{(-)}\rangle]/\pi \times\langle E_k^{(+)}E_k^{(-)}\rangle. \quad (28)$$

[13] N. Bloembergen and Y. R. Shen, Phys. Rev. Letters **13**, 720 (1964).

Such a distribution gives

$$\int d^2\mathcal{E}_k W(\mathcal{E}_k)|\mathcal{E}_k|^{2n} = n!\langle E_k^{(+)}E_k^{(-)}\rangle^n.$$

Therefore, we would get

$$\langle E_s^{(+)}E_s^{(-)}\rangle(t) = [\langle E_s^{(+)}E_s^{(-)}\rangle(0)+S]/ \times[1-2\gamma_R\langle E_k^{(+)}E_k^{(-)}\rangle t]-S. \quad (29)$$

Equation (28) also leads to the conclusion that the probability of having at least $(1/N)$ part of the ensembles with a gain coefficient $2\gamma_R|\mathcal{E}_k|^2$ larger than the average gain $2\gamma_R\langle E_k^{(+)}E_k^{(-)}\rangle$ by a factor $\ln N$ is $1-e^{-1}=0.63$, where N is the number of modes.[13] However, if the fields are nonstationary or there is phase correlation between modes, the factor $\ln N$ would be replaced by a much larger value, of the order of N for full phase correlation.

The statistical properties of the Stokes output in the Raman transitions are difficult to describe quantitatively. Qualitatively, they depend strongly on the initial statistical nature of both the pump and the Stokes field. If the pump is coherent and not appreciably disturbed, then the statistical properties of the Stokes output would be the same as those of a quantum oscillator.[14] In particular, if initially there is no Stokes input, the medium would appear as a Stokes noise generator.

III. INCOHERENT LINEAR AND NONLINEAR SCATTERING

Rayleigh and Brillouin scattering are often regarded as linear scattering processes. Nevertheless, they belong to the class of nonlinear optics in the sense that excitational waves in the medium actually play the equivalent role of light waves. Incoherent Rayleigh and Brillouin scattering are most frequently discussed in the classical language.[15] The transformation from classical to quantum terms is, however, straightforward.

Consider scattering due to density fluctuations in a dilute medium. The total Hamiltonian is

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}},$$

where $\mathcal{H}_0$, given by Eq. (4), includes the coherent interaction of light with the medium, and $\mathcal{H}_{\text{int}}$ describes solely the incoherent part of the interaction. In first order, with the trace taken over the atomic system, $\mathcal{H}_{\text{int}}$ can be written as

$$\mathcal{H}_{\text{int}} = -\sum_{i,k}[\mathbf{E}_k^{(+)}(\mathbf{r}_i)\cdot\mathbf{p}\cdot\mathbf{E}_{k_0}^{(-)}(\mathbf{r}_i) + \mathbf{E}_{k_0}^{(+)}(\mathbf{r}_i)\cdot\mathbf{p}\cdot\mathbf{E}_k^{(-)}(\mathbf{r}_i)], \quad (30)$$

[14] J. P. Gordon, L. R. Walker, and W. H. Louisell, Phys. Rev. **130**, 806 (1963).

[15] See, for example, L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, Inc., New York, 1960), p. 377.

where $\mathbf{p}$ is the atomic polarizability in the electric-dipole approximation, k_0 is the pump mode, and k is the mode of the scattered radiation. By assuming running modes with

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

Eq. (30) takes the form

$$\mathcal{H}_{\rm int}=\sum_k\left[a_k^\dagger(t)a_{k_0}(t)f_k^*+a_{k_0}^\dagger(t)a_k(t)f_k\right],\quad(31)$$

where in the Heisenberg representation

$$f_k=-\sum_i(2\pi\hbar\omega_k^{1/2}\omega_{k_0}^{1/2}/\epsilon_kL^3)\hat{e}_k\cdot\mathbf{p}^*\cdot\hat{e}_{k_0}\times\exp[i(\mathbf{k}-\mathbf{k}_0)\cdot\mathbf{r}_i],$$

the equation of motion is

$$da_k(t)/dt=i\omega_ka_k(t)-(i/\hbar)f_k^*a_{k_0}(t).\quad(32)$$

If the pump field is of high intensity, and is not disturbed appreciably by the incoherent scattering, we can treat a_k and a_{k_0} as c numbers. This is actually equivalent to treating the pump field in the classical limit. Then, Eq. (32) can be solved readily. In fact, the problem reduces to the one of radiation by a prescribed current distribution discussed by Glauber.[1] The solution of Eq. (32) leads to the expression of an electric field at a point $\mathbf{r}$ for the scattered radiation,

$$\begin{aligned}\mathbf{E}_{\rm sc}^{(-)}(\mathbf{r},t)&=-(1/c)\partial\mathbf{A}^{(-)}(\mathbf{r},t)/\partial t\\&=\left(\frac{-\partial}{c\partial t}\right)\left\{\hat{e}_k\left(\frac{iL^3}{8\pi^3}\right)\int d^3k\int_0^t dt'\left(\frac{2\pi c}{\hbar k\epsilon_kL^3}\right)^{1/2}\right.\\&\quad\times f_k^*a_{k_0}(t')\exp[i\mathbf{k}-\mathbf{r}-i\omega(t-t')]\\&\quad\left.+\text{complex conjugate}\right\}.\end{aligned}$$

The integration in the above equation can be carried out explicitly.[16] At a point $\mathbf{r}$ sufficiently far from the scattering region, the electric field is approximately given by

$$\begin{aligned}\mathbf{E}_{\rm sc}^{(-)}(\mathbf{r},t)&=a_{k_0}\mathbf{F}(r,t)\exp(i\mathbf{k}_0\cdot\mathbf{r})\\&\quad\times\int_V d^3r'N(\mathbf{r}',t)\exp[i(\mathbf{k}_0-\mathbf{k})\cdot\mathbf{r}'],\quad(33)\\\mathbf{F}(\mathbf{r},t)&=(\mathbf{k}\times i\mathbf{p}\cdot\hat{e}_{k_0})\times(\mathbf{k}/|\mathbf{r}-\mathbf{R}|)(2\pi\hbar\omega_0/\epsilon_kL^3)^{1/2}\\&\quad\times\exp(i\mathbf{k}\cdot\mathbf{r}-i\omega_0t),\end{aligned}$$

where V is the volume of interaction and $\mathbf{R}$ is the center of V. The calculation now follows essentially the same as the classical treatment.[15] Clearly, if the scattering medium is uniform and stationary, so that the density of atoms $N(\mathbf{r},t)$ is constant, the integral $\int_V d^3r$ in Eq. (33) would vanish if $\mathbf{k}_0\neq\mathbf{k}$, and, consequently, there is no scattering in the direction $\mathbf{k}\neq\mathbf{k}_0$. Thus, incoherent scattering appears as a result of density fluctuations. If we consider only one Fourier component of the total density fluctuations,

$$N(\mathbf{r},t)=\sum_q N_q\exp(i\mathbf{q}\cdot\mathbf{r}-i\omega_qt),$$

then we obtain from Eq. (33) the first-order correlation function

$$\begin{aligned}\langle\mathbf{E}_{\rm sc}^{(+)}(r,t_1)\cdot\mathbf{E}_{\rm sc}^{(-)}(r,t_2)\rangle&=|F(r)|^2 8\pi^3V\langle a_{k_0}^\dagger a_{k_0}\rangle\\&\quad\times\langle N_q(t_1)N_q(t_2)^*\rangle\Delta(\mathbf{k}_0-\mathbf{k}\pm\mathbf{q})\\&\quad\times\exp[-i(\omega_0\pm\omega_q)(t_1-t_2)]\\\Delta(k_0-k\pm q)&\xrightarrow[V\to\infty]{}\delta(\mathbf{k}_0-\mathbf{k}\pm\mathbf{q}).\end{aligned}\quad(34)$$

For $N(r,t)=$ constant, the scattered radiation in the direction $\mathbf{k}\neq\mathbf{k}_0+\mathbf{k}_0'$ vanishes. The Fourier transform of $\langle E_{\rm sc}^{(+)}(r,t_1)E_{\rm sc}^{(-)}(r,t_2)\rangle$ gives the power spectral density of the scattered radiation. Higher-order correlation functions can also be obtained from Eq. (33), and, hence the statistical properties of the scattered radiation can be described completely.

It is, however, interesting to note that for this case, an explicit expression of the density matrix for the scattered radiation can be written down immediately, following Glauber's treatment for the radiation by a prescribed current distribution.[1] If we assume P representation for both the pump field and the density fluctuations, such that

$$\begin{aligned}\langle(a_{k_0}^\dagger)^m(a_{k_0})^n\rangle&=\int d^2\alpha_{k_0}P(\alpha_{k_0})(\alpha_{k_0}^*)^m(\alpha_{k_0})^n,\\\langle(N_q)^m(N_q^*)^n\rangle&=\int d^2\sigma_qP(\sigma_q)(\sigma_q)^m(\sigma_q^*)^n,\end{aligned}\quad(35)$$

then we find for the scattered radiation

$$\rho_k(t)=\int d^2\alpha_{k_0}\int d^2\sigma_qP_{k_0}(\alpha_{k_0})P_q(\sigma_q)|\alpha_k(t)\rangle\langle\alpha_k(t)|,$$

with

$$\alpha_k(t)=(i/\hbar)\int_0^t dt\,\mathfrak{F}(\alpha_{k_0},\sigma_q),\quad(36)$$

where

$$\mathfrak{F}(\alpha_{k_0},\sigma_q)=-(2\pi\hbar\omega_{k_0}^{1/2}\omega_k^{1/2}/\epsilon_kL^3)\hat{e}_k\cdot\mathbf{p}\cdot\hat{e}_{k_0}\sigma_q.$$

This shows that statistical properties of the scattered radiation are determined by those of incident radiation and density fluctuations. Thus, measurements of statistical properties of the scattered radiation could yield information about the statistical properties of the density fluctuations, if those of the incident radiation are known. The analysis is particularly simple for coherent incident radiation.

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[16] E. Fermi, Rev. Mod. Phys. 4, 87 (1932).

Recently, the question whether intensities of scattered radiation may be different for coherent and incoherent incident radiation has arisen.[17] It is clear from Eq. (34) that with our assumptions for linear incoherent scattering, the average scattering intensity is directly proportional to the average number of photons in the pump modes, and is independent of the coherent property of the pump field.

The above calculation can be extended to the case of incoherent nonlinear scattering, which has recently been investigated by Terhune *et al.*[18] We shall again consider only nonlinear scattering due to density fluctuations, in which two photons in the pump modes k_0 and k_0' are scattered into a single photon in the scattered mode k. The corresponding interaction Hamiltonian can be written as

$$\mathcal{H}_{\text{int}}=-\sum_{i,k}\big[\mathbf{E}_k^{(+)}(\mathbf{r}_i)\cdot\mathbf{p}^{(2)}:\mathbf{E}_{k_0}^{(-)}(\mathbf{r}_i)\mathbf{E}_{k_0'}^{(-)}(\mathbf{r}_i)+\text{adjoint}\big],\quad(37)$$

where $\mathbf{p}^{(2)}$ is the second-order nonlinear polarizability.[19] Following the same procedure as in the linear case, one would find for the scattered radiation

$$\mathbf{E}_{\text{sc}}^{(-)}(\mathbf{r},t)=a_{k_0}a_{k_0'}\mathbf{F}(\mathbf{r},t)[\exp i(\mathbf{k}_0+\mathbf{k}_0')\cdot\mathbf{R}]\times\int_V d^3r'N(\mathbf{r}',t)\exp[i(\mathbf{k}_0+\mathbf{k}_0'-\mathbf{k})\cdot\mathbf{r}]$$

$$\mathbf{F}(\mathbf{r},t)=(\mathbf{k}\times\mathbf{p}^{(2)}:\hat{e}_{k_0}\hat{e}_{k_0'})\times\left(\frac{\mathbf{k}}{|\mathbf{r}-\mathbf{R}|}\right)\left(\frac{2\pi\hbar}{L^3}\right)\times\left(\frac{\omega_0\omega_0'}{\epsilon_{k_0}\epsilon_{k_0'}}\right)^{1/2}\exp[i\mathbf{k}\cdot\mathbf{r}-i(\omega_0+\omega_0')t].\quad(38)$$

If only one Fourier component of the density fluctuations is taken into account, the first-order correlation function of the scattered radiation is

$$\langle\mathbf{E}_{\text{sc}}^{(+)}(\mathbf{r},t_1)\cdot\mathbf{E}_{\text{sc}}^{(-)}(\mathbf{r},t_2)\rangle=|F(r)|^2 8\pi^3V\times\langle a_{k_0}^\dagger a_{k_0'}^\dagger a_{k_0}a_{k_0'}\rangle\langle N_qN_q^*\rangle\Delta(\mathbf{k}_0+\mathbf{k}_0'-\mathbf{k}\pm\mathbf{q})\times\exp[-i(\omega_0+\omega_0'\pm\omega_q)(t_1-t_2)],\quad(39)$$

where $F(r)$ is given in Eq. (38). Assuming Eq. (35) for both the pump modes and the density fluctuations, we find the density matrix for the scattered radiation

$$\rho_k(t)=\int d^2\alpha_{k_0}\int d^2\alpha_{k_0'}\int d^2\sigma_qP_{k_0}(\alpha_{k_0})P_{k_0'}(\alpha_{k_0'})\times P_q(\sigma_q)|\alpha_k(t)\rangle\langle\alpha_k(t)|\,,$$

$$\alpha_k(t)=\frac{i}{\hbar}\int_0^t dt\,\mathfrak{F}(\alpha_{k_0},\alpha_{k_0'},\sigma_q)\,,\quad(40)$$

$$\mathfrak{F}(\alpha_{k_0},\alpha_{k_0'},\sigma_q)=-(8\pi^3\hbar^3\omega_{k_0}\omega_{k_0'}\omega_k/\epsilon_{k_0}\epsilon_{k_0'}\epsilon_kL^9)^{1/2}\times\hat{e}_k\cdot\mathbf{p}^{(2)}:\hat{e}_{k_0}\hat{e}_{k_0'}\alpha_{k_0}\alpha_{k_0'}\sigma_q.$$

From Eq. (39) it is seen that the scattering intensity, $\langle|E_{\text{sc}}^{(+)}(r,t)|^2\rangle$, for $k\neq l$ is proportional to $\langle a_{k_0}^\dagger a_{k_0}\rangle\langle a_{k_0'}^\dagger a_{k_0'}\rangle$, but for $k=l$, it is proportional to $\langle a_{k_0}^\dagger a_{k_0}^\dagger a_{k_0}a_{k_0}\rangle$, which from Eq. (17) is two times larger for chaotic than for coherent fields.

In the actual experiments, the incident radiation may contain many modes. However, as long as the divergence and the linewidth of the incident radiation are small compared with the acceptance angle of the photodetector and the linewidth of the scattered radiation, conservation of energy and momentum as expressed in Eq. (39) can be relaxed. We therefore have for the multimode case,

$$\langle|E_{\text{sc}}^{(+)}(\mathbf{r},t)|^2\rangle\cong|F(r)|^2(2\pi VL^6/\hbar^2\omega_0\omega_0')\times\langle E_{k_0}^{(+)}E_{k_0'}^{(+)}E_{k_0}^{(-)}E_{k_0'}^{(-)}\rangle(R,t)\times\langle N_qN_q^*\rangle\Delta(\mathbf{k}_0+\mathbf{k}_0'-\mathbf{k}\pm\mathbf{q}),\quad(41)$$

where

$$E_{k_0}^{(-)}(\mathbf{R})=\sum_\lambda(2\pi\hbar\omega_0/\epsilon_{k_0}L^3)^{1/2}a_{k_{0\lambda}}\exp(i\mathbf{k}_{0\lambda}\cdot\mathbf{R}-i\omega_{0\lambda}t).$$

and $\mathbf{R}$ is the center of the volume V. Then, if $k_0=k_0'$, from Eqs. (20a) and (28) we find for stationary fields, if the number of modes is large, $\langle E_{k_0}^{(+)}E_{k_0}^{(+)}E_{k_0}^{(-)}E_{k_0}^{(-)}\rangle=2\langle E_{k_0}^{(+)}E_{k_0}^{(-)}\rangle^2$. This shows that the scattering intensity in the multimode case is two times higher than in the single-mode case. The second-order incoherent nonlinear scattering is closely related to the second-order coherent scattering, which gives rise to sum-frequency and second harmonic generation, as we shall now discuss.

IV. SUM-FREQUENCY AND SECOND HARMONIC GENERATION

The coherent sum-frequency generation can be described by the same interaction Hamiltonian in Eq. (37) for incoherent nonlinear scattering. It was shown in Sec. III that if there are no fluctuations in the medium, scattered radiation can only appear in the direction where the wave vectors of incident and scattered radiation are matched. This corresponds to coherent scattering. Thus, coherent sum-frequency generation described by the Hamiltonian of Eq. (37) appears in the direction $k_0+k_0'-2k\approx0$.

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[17] T. V. George, L. Goldstein, L. Slama, and M. Yokoyama, Phys. Rev. **137**, A369 (1965); R. D. Watson and M. K. Clark, Phys. Rev. Letters **14**, 1057 (1965); R. C. C. Leite, R. S. Moore, S. P. S. Porto, and J. E. Ripper, *ibid.* **14**, 7 (1965); D. H. Woodward, Appl. Opt. **2**, 1205 (1963).

[18] R. W. Terhune, P. D. Maker, and C. M. Savage, Phys. Rev. Letters **14**, 681 (1965); P. D. Maker, in *Proceedings of the Conference on Physics of Quantum Electronics, Puerto Rico, 1965*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 60.

[19] J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. Pershan, Phys. Rev. **127**, 1918 (1962).

The calculation follows essentially the same pattern as for the case of incoherent nonlinear scattering. Again, in the Heisenberg representation, the equation of motion is

$$da_k/dt=-i\omega_k a_k(t)-(i/\hbar)f_k a_{k_0}(t)a_{k_0'}(t),$$
$$f_k=-NV(8\pi^3\hbar^3\omega_0^{1/2}\omega_0'^{1/2}\omega_k^{1/2}/\epsilon_{k_0}\epsilon_{k_0'}\epsilon_k L^9)^{1/2}\times\hat{e}_k\cdot\mathbf{p}^{(2)}:\hat{e}_{k_0}\hat{e}_{k_0'}. \quad (42)$$

Here, we have assumed a uniform medium in a volume V. For intense pump fields, which have not yet been depleted appreciably by the sum-frequency generation, a_{k_0} and $a_{k_0'}$ can be treated as constant c numbers. Then, Eq. (42) yields

$$a_k(t)=[a_k(0)-(i/\hbar)f_k a_{k_0}a_{k_0'}t]\exp(-i2\omega_0 t). \quad (43)$$

From Eqs. (42) and (43), we find the average rate of sum-frequency generation;

$$\begin{aligned}d\langle a_k^\dagger a_k\rangle(t)/dt &=(i/\hbar)[f_k^*\langle a_{k_0}^\dagger a_{k_0'}^\dagger a_k\rangle(0)-f_k\langle a_k^\dagger a_{k_0}a_{k_0'}\rangle(0)]\\&+(\omega_k t/\hbar)[f_k^*\langle a_{k_0}^\dagger a_{k_0'}^\dagger a_k\rangle(0)+f_k\langle a_k^\dagger a_{k_0}a_{k'_0}\rangle(0)]\\&+(2|f_k|^2t^2/\hbar^2)\langle a_{k_0}^\dagger a_{k_0'}^\dagger a_{k_0}a_{k_0}\rangle(0), \quad (44)\end{aligned}$$

which can readily be integrated. Equation (44) shows that for $\mathbf{k}_0=\mathbf{k}_0'$, corresponding to second-harmonic generation, the average rate of generation depends on the initial statistical properties of the pump and the second-harmonic fields. In particular, if $\langle a_k(0)\rangle=0$, this rate is proportional to the second-order correlation function $\langle a_{k_0}^\dagger a_{k_0}^\dagger a_{k_0}a_{k_0}\rangle(0)$, and is therefore two times higher for a chaotic than for a coherent pump field.

Corresponding to the Hamiltonian of Eq. (37) with a_{k_0} and $a_{k_0'}$ treated as c numbers, the density matrix for the sum-frequency field is

$$\rho_k(t)=\int d^2\alpha_{k_0}d^2\alpha_{k_0'}P_{k_0}(\alpha_{k_0})P_{k_0'}(\alpha_{k_0'})\times D(\alpha_k)\rho_k(0)D^{-1}(\alpha_k),$$
$$D(\alpha_k)=\exp[\alpha_k a_k^\dagger-\alpha_k^* a_k], \quad (45)$$
$$\alpha_k=\frac{i}{\hbar}\int_0^t dt\, f_k\alpha_{k_0}\alpha_{k_0'},$$

where f_k is given in Eq. (42) and P representation is assumed for the pump fields,

$$\rho_{k_0}=\int d^2\alpha_{k_0}P_{k_0}(\alpha_{k_0})|\alpha_{k_0}\rangle\langle\alpha_{k_0}|,$$

$$\rho_{k_0'}=\int d^2\alpha_{k_0'}P_{k_0'}(\alpha_{k_0'})|\alpha_{k_0'}\rangle\langle\alpha_{k_0'}|.$$

If initially, $\rho_k(0)=\int d^2\beta_k P_k(\beta_k)|\beta_k\rangle\langle\beta_k|$, then Eq. (45) becomes

$$\rho_k(t)=\int d^2\alpha_{k_0}d^2\alpha_{k_0'}d^2\beta_k P_{k_0}(\alpha_{k_0})P_{k_0'}(\alpha_{k_0'})P_k(\beta_k)\times|\alpha_k+\beta_k\rangle\langle\alpha_k+\beta_k|, \quad (46a)$$

which, in the case of second-harmonic generation, reduces to

$$\rho_k(t)=\int d^2\alpha_{k_0}d^2\beta_k P_{k_0}(\alpha_{k_0})P_k(\beta_k)\times|\alpha_k+\beta_k\rangle\langle\alpha_k+\beta_k|, \quad (46b)$$

with $\alpha_k=(i/\hbar)\int_0^t dt\, f_k\alpha_{k_0}^2$. The above expressions lead to the following results. (1) For coherent pump fields, if $|\beta_k\rangle=|0\rangle$, the generated sum-frequency field is also coherent; but if $|\beta_k\rangle\neq|0\rangle$, the sum-frequency output has the same distribution function P_k as the input with $\rho_k(t)=\int d^2\beta_k P_k(\beta_k)|\alpha_k+\beta_k\rangle\langle\alpha_k+\beta_k|$. (2) If $|\beta_k\rangle=|0\rangle$, the sum-frequency output reflects the statistics of the pump fields. (3) In general, the sum-frequency output has the composite statistical properties of the pump fields and the sum-frequency input. Clearly, measurements of the statistics of the sum-frequency or second-harmonic output could yield information about the statistics of the pump fields. For example, if $|\beta_k\rangle=|0\rangle$, the nth-order correlation function of the second harmonics is proportional to the $2n$th-order correlation function of the fundamental.

The discussion can easily be extended to the multimode case. As discussed in the case of incoherent scattering, if the energy and momentum matching condition is relaxed, Eq. (44) gives

$$\begin{aligned}d\langle E_k^{(+)}E_k^{(-)}\rangle(\mathbf{r},t)/dt&=(i/\hbar)[g_k^*\langle E_{k_0}^{(+)}E_{k_0'}^{(+)}E_{k_0}^{(-)}\rangle(\mathbf{r},0)\\&-g_k\langle E_k^{(+)}E_{k_0}^{(-)}E_{k_0'}^{(-)}\rangle(\mathbf{r},0)]+(\omega_k t/\hbar)\\&\times[g_k^*\langle E_{k_0}^{(+)}E_{k_0'}^{(+)}E_k^{(-)}\rangle(\mathbf{r},0)\\&+g_k\langle E_k^{(+)}E_{k_0}^{(-)}E_{k_0'}^{(-)}\rangle(\mathbf{r},0)]+(2|g_k|^2t^2/\hbar^2)\\&\times\langle E_{k_0}^{(+)}E_{k_0}^{(+)}E_{k_0'}^{(-)}E_{k_0'}^{(-)}\rangle(\mathbf{r},0), \quad (47)\end{aligned}$$
$$g_k=-NV(2\pi\hbar\omega_k^{1/2}/\epsilon_k^{1/2}L^3)\hat{e}_k\cdot p^{(2)}:\hat{e}_{k_0}\hat{e}_{k_0'},$$

where $E_k^{(-)}(r,t)$ has the same expression as in Eq. (41). Again, for $k_0=k_0'$, if $\langle E_k(0)\rangle=0$, the average rate of second-harmonic generation is usually higher for multimode than for single-mode pump fields, since $\langle E_{k_0}^{(+)}E_{k_0}^{(+)}E_{k_0}^{(-)}E_{k_0}^{(-)}\rangle$ has a larger value in the former case. For stationary fields, there is a ratio of 2 in the rates of second-harmonic generation for the two cases.[20] The density matrix given in Eq. (46) can also easily be generalized to multimodes.

The above discussion is valid as long as there is no appreciable depletion of pump power by sum-frequency generation. For the more general case, the mathematics becomes much more complicated, since the reaction of the sum-frequency field on the pump fields must be

[20] J. Ducuing and N. Bloembergen, Phys. Rev. 133, A1493 (1964).

taken into account. The sum-frequency generation now depends on bigher-order correlation functions of the initial pump fields. The output is no longer coherent even if the initial pump fields are coherent. Ducuing and Armstrong[21] have discussed the statistical aspects of second-harmonic generation with high conversion using the classical approach. A corresponding quantum-statistical discussion would be extremely difficult, if the noncommutability of the operators a and $a^\dagger$ is to be taken into account.

V. PARAMETRIC AMPLIFICATION

One of the most important subjects in nonlinear optics is parametric amplification. It is not only because the parametric amplification may lead to tunable oscillators at light frequencies,[22] but because in a broader sense, it also describes such important nonlinear processes as stimulated Raman and Brillouin scattering by elementary excitations.[23] In the latter cases, the idler photon mode is replaced by the mode of elementary excitations. The calculations remain the same if the elementary excitations are bosons.

The statistical properties of a parametric amplifier have been discussed in detail by Gordon *et al.*[24] However, they have assumed a constant field strength for the pump mode. From our discussion in the previous sections, we expect that the statistics of the pump field should influence the statistical output of the amplifier. Their results are valid only when the pump field is in a coherent state. In the following, we shall follow their calculations, but take into account the statistical properties of the pump field.

The interaction Hamiltonian for parametric amplification is also the same as in Eq. (37).

$$\mathcal{H}_{\text{int}}=-\sum_{i,k}\left[\mathbf{E}_p^{(+)}(\mathbf{r}_i)\cdot\mathbf{p}^{(2)}:\mathbf{E}_s^{(-)}(\mathbf{r}_i)\times\mathbf{E}_I^{(-)}(\mathbf{r}_i)+\text{adjoint}\right]. \quad (48)$$

Here, however, the coherent scattering process is to destroy a photon in the pump mode p, and to create one photon in the signal mode s and another in the idler mode I, with $\omega_p=\omega_s+\omega_I$ and $\mathbf{k}_p=\mathbf{k}_s+\mathbf{k}_I$. The Heisenberg equations of motion are

$$\begin{aligned} da_s/dt&=-i\omega_s a_s(t)-i\kappa a_p(t)a_I^\dagger(t)\,,\\ da_I^\dagger/dt&=i\omega_I a_I^\dagger(t)-i\kappa^* a_p^\dagger(t)a_s(t)\,, \end{aligned} \quad (49)$$

where

$$\kappa=-NV(8\pi^3\hbar\omega_p\omega_s\omega_I/\epsilon_p\epsilon_s\epsilon_I L^9)^{1/2}\hat{e}_p\cdot\mathbf{p}^{(2)*}:\hat{e}_s\hat{e}_I.$$

If the pump field is of high intensity, and has not been depleted appreciably by the parametric process, then $a_p(t)\approx a_p(0)\exp(-i\omega_p t)$, where $a_p(0)$ and $a_p^\dagger(0)$ can be regarded as c numbers. Then the solution of Eq. (49) is

$$\begin{aligned} a_s(t)=\{&a_s(0)\cosh[|\kappa|(a_p^\dagger a_p)^{1/2}t]\\ &+[i\kappa a_p/|\kappa|(a_p^\dagger a_p)^{1/2}]a_I(0)\\ &\times\sinh[|\kappa|(a_p^\dagger a_p)^{1/2}t]\}\exp(-i\omega_s t)\,,\\ a_I(t)=\{&a_I(0)\cosh[|\kappa|(a_p^\dagger a_p)^{1/2}t]\\ &+[i\kappa a_p/|\kappa|(a_p^\dagger a_p)^{1/2}]a_s(0)\\ &\times\sinh[|\kappa|(a_p^\dagger a_p)^{1/2}t]\}\exp(-i\omega_I t)\,, \end{aligned} \quad (50)$$

$$\begin{aligned} \langle a_s^\dagger a_s\rangle(t)=\mathrm{Tr}\rho_p(0)\{&\langle a_s^\dagger a_s\rangle(0)\cosh^2[|\kappa|(a_p^\dagger a_p)^{1/2}t]\\ &+(\langle a_I^\dagger a_I\rangle(0)+1)+\sinh^2[|\kappa|(a_p^\dagger a_p)^{1/2}t]\\ &+i[\kappa a_p\langle a_s^\dagger a_I^\dagger\rangle(0)/|\kappa|(a_p^\dagger a_p)^{1/2}-\kappa^* a_p^\dagger\langle a_s a_I\rangle(0)/\\ &|\kappa|(a_p^\dagger a_p)^{1/2}]\tfrac{1}{2}\sinh 2[|\kappa|(a_p^\dagger a_p)^{1/2}t]\}\,, \end{aligned} \quad (51)$$

with a similar expression for $\langle a_I^\dagger a_I\rangle(t)$. Equation (51) shows that the output signal in the parametric amplification depends on the initial statistical properties of the pump field. Assume $\langle a_s a_I\rangle(0)=0$. Then, for a coherent pump field, we have

$$\begin{aligned} \langle a_s^\dagger a_s\rangle(t)=&\tfrac{1}{2}[\langle a_s^\dagger a_s\rangle(0)-\langle a_I^\dagger a_I\rangle(0)-1]\\ &+\tfrac{1}{2}[\langle a_s^\dagger a_s\rangle(0)+\langle a_I^\dagger a_I\rangle(0)+1]\\ &\times\cosh[2|\kappa|\langle(a_p^\dagger a_p)^{1/2}\rangle t]\,, \end{aligned} \quad (52)$$

but for a chaotic pump field, since

$$\langle(a_p^\dagger a_p)^n\rangle=n!\langle a_p^\dagger a_p\rangle^n\,,$$

we have

$$\begin{aligned} \langle a_s^\dagger a_s\rangle(t)=&\tfrac{1}{2}[\langle a_s^\dagger a_s\rangle(0)-\langle a_I^\dagger a_I\rangle(0)-1]\\ &+\tfrac{1}{2}[\langle a_s^\dagger a_s\rangle(0)+\langle a_I^\dagger a_I\rangle(0)+1]\\ &\times\sum_{n=0}^{\infty}[n!/(2n)!](2|\kappa|t)^{2n}\langle a_p^\dagger a_p\rangle_p. \end{aligned} \quad (53)$$

It is clear from the above expressions that the signal output is much larger for chaotic than for coherent pump fields.

For the multimode case, if the energy and momentum matching condition can be relaxed, as discussed in the previous sections, the calculations follow essentially the same as in the single-mode case with a replaced by $E^{(-)}(r,t)$, and κ in Eq. (49) by

$$\kappa'=-NV(2\pi/L^3)(\omega_s\omega_I/\epsilon_s\epsilon_I)^{1/2}\hat{e}_s\cdot\mathbf{p}^{(2)*}:\hat{e}_s\hat{e}_I. \quad (54)$$

The result is

$$\begin{aligned} \langle E_s^{(+)}E_s^{(-)}\rangle(r,t)=\mathrm{Tr}\rho_p(0)\{&\langle E_s^{(+)}E_s^{(-)}\rangle(r,0)\\ &\times\cosh^2[|\kappa'|(E_p^{(+)}E_p^{(-)})^{1/2}t]+\langle E_I^{(-)}E_I^{(+)}\rangle(r,0)\\ &\times\sinh^2[|\kappa'|(E_p^{(+)}E_p^{(-)})^{1/2}t]\}\,, \end{aligned} \quad (55)$$

assuming $\langle E_s^{(-)}E_I^{(-)}\rangle(r,0)=0$. Again, the output signal is usually larger for multimode than for single-mode

[21] J. Ducuing and J. A. Armstrong, in *Proceedings of the Third Quantum Electronics Conference, Paris, 1963*, edited by P. Grivet and N. Bloembergen, (Columbia University Press, New York, 1964), p. 1643.

[22] J. A. Giordmaine and R. C. Miller, Phys. Rev. Letters **14**, 973 (1965).

[23] Y. R. Shen and N. Bloembergen, Phys. Rev. **143**, 372 (1966).

[24] J. P. Gordon, W. H. Louisell, and L. R. Walker, Phys. Rev. **129**, 481 (1963). See also W. H. Louisell, *Radiation and Noise in Quantum Electronics* (McGraw-Hill Book Company, Inc., New York, 1964).

pump fields. For stationary fields with many modes, we have

$$\langle E_s^{(+)}E_s^{(-)}\rangle(r,t)=\tfrac{1}{2}[\langle E_s^{(+)}E_s^{(-)}\rangle(r,0)$$
$$-\langle E_I^{(-)}E_I^{(+)}\rangle(r,0)]+\tfrac{1}{2}[\langle E_s^{(+)}E_s^{(-)}\rangle(r,0)$$
$$+\langle E_I^{(-)}E_I^{(+)}\rangle(r,0)]\sum_{n=0}^{\infty}[n!/(2n)!](2\kappa' t)^{2n}$$
$$\times\langle E_p^{(+)}E_p^{(-)}\rangle^n(r,0). \quad (56)$$

In principle, all higher-order correlation functions of the signal and the idler fields can be obtained from Eq. (50). However, to describe the statistical properties of fields, an explicit expression of the density matrix for the fields is usually of great interest. For the case of parametric amplification, the density matrices $\rho_s(t)=\mathrm{Tr}_I\rho_{s,I}(t)$ and $\rho_I(t)=\mathrm{Tr}_s\rho_{s,I}(t)$ for the signal and the idler fields can be obtained through the use of the characteristic functions,[24] which are defined as[25]

$$X_s(\gamma,t)=\mathrm{Tr}_{s,I}\{\rho_{s,I}(t)\exp[\gamma a_s^\dagger(0)]\exp[-\gamma^* a_s(0)]\}$$
$$=\mathrm{Tr}_{s,I}\{\rho_s(0)\rho_I(0)\exp[\gamma a_s^\dagger(t)]$$
$$\times\exp[-\gamma^* a_s(t)]\}, \quad (57)$$
$$X_I(\gamma,t)=\mathrm{Tr}_{s,I}\{\rho_s(0)\rho_I(0)\exp[\gamma a_I^\dagger(t)]$$
$$\times\exp[-\gamma^* a_I(t)]\}.$$

Explicit expressions of X_s and X_I can be found by substituting into Eq. (57) the expressions of $a_s(t)$ and $a_I(t)$ in Eq. (50) and the known initial distribution $\rho_s(0)$ and $\rho_I(0)$.[24] Here, $a_p^\dagger$ and a_p are treated as c numbers. Then, the characteristic functions lead to the density matrices in the P representation,[25]

$$\rho_s(t)=\mathrm{Tr}_I\rho_{s,I}(t)=\int d^2\alpha_s P_s(\alpha_s,t)|\alpha_s\rangle\langle\alpha_s|,$$

$$P_s(\alpha_s,t)=\int d^2\alpha_p P_p(\alpha_p)\langle\alpha_p|$$
$$\times\left[\int X(\gamma,t)\exp(\alpha_s\gamma^*-\alpha_s^*\gamma)d^2\gamma/\pi^2\right]|\alpha_p\rangle, \quad (58)$$

with a similar equation for $\rho_I(t)$. As an example, consider the case where initially both the signal and the idler modes are in the vacuum state.

$$\rho_s(0)=|0_s\rangle\langle 0_s|, \quad \rho_I(0)=|0_I\rangle\langle 0_I|.$$

From Eqs. (50) and (57), the characteristic function X_s is

$$X_s(\gamma,t)=\exp\{\tfrac{1}{2}|\gamma|^2(\cosh^2[|\kappa|(a_p^\dagger a_p)^{1/2}t]$$
$$+\sinh^2[|\kappa|(a_p^\dagger a_p)^{1/2}t]-1)\}. \quad (59)$$

[25] R. Glauber, in *Proceedings of Conference on Physics of Quantum Electronics, 1965*, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 788.

Substitution of $X_s(\gamma,t)$ in Eq. (58) gives

$$P_s(\alpha_s,t)=\int d^2\alpha_p P_p(\alpha_p)(1/\pi\langle n_s\rangle)$$
$$\times\exp[-|\alpha_s|^2/\langle n_s\rangle], \quad (60)$$

where

$$\langle n_s\rangle(t)=\sinh^2[|\kappa\alpha_p|t].$$

If the pump field is coherent, this corresponds to a Gaussian probability distribution for a chaotic field with an average number of photons $\langle n_s\rangle$.[24] Thus, with no input to the amplifier, the parametric amplifier acts as a noise oscillator. Characteristic functions for various input conditions have been obtained by Gordon *et al.*[24]

More generally, we should also consider the loss in the modes due to absorption. However, in the first approximation, we can simply take ω_s and ω_I in Eq. (49) as complex quantities. The mathematics is straightforward, and will not be reproduced here. The above discussion is valid as long as the pump field is not appreciably disturbed. The general calculations, taking into account the reaction of the parametric process on the pump field, becomes extremely complicated.

VI. CONCLUSION

Nonlinear optical effects often depend on the statistical properties of the fields present. The rate of nonlinear absorption, emission, and amplification is higher for chaotic than for coherent, and higher for multimode than for single-mode pump fields. The statistics of the fields generated in the nonlinear effects is a partial function of the statistics of the pump fields. Measurements of the statistics of the output fields may yield information about the statistics of the input fields, and the statistical properties of the medium.

APPENDIX

Classically, a cavity problem of coherent scattering can usually be converted to a corresponding steady-state propagation problem by simply replacing t by $-z/c$ in the field amplitudes, where $\hat{z}$ is the direction of propagation. It is expected that the same is true in the quantum treatment. This can be realized by using a localized momentum operator instead of the Hamiltonian operator.

For steady-state propagation, the field amplitudes at fixed spatial points remain unchanged. The vector potential for a plane wave propagating in the z direction can be written as

$$A(z,t)=c\sum_k(\hbar/2\omega_k\epsilon_k L^3)^{1/2}$$
$$\times\{\psi_k(z)\exp(-i\omega_k t)+\psi_k^\dagger(z)\exp(i\omega_k t)\},$$
$$\psi_k(z)=b(z)\exp(ikz), \quad (A1)$$
$$[b_k(z),b_{k'}^\dagger(z)]=\delta_{kk'}.$$

For free fields, $\psi_k(z)=a_k\exp(ikz)$. Here, we have defined localized annihilation and creation operators $b(z)$ and $b^\dagger(z)$ under the assumption that $\langle(b_k^\dagger)^m(b_k)^n\rangle$ does not vary much in a distance d large compared with the wavelength. We also assume that $k=2\pi n/d$, where n is an integer. Thus, the corresponding localized photon number operator is[26]

$$\hat{n}(z)=(\mathcal{A}d/L^3)\sum_k b_k^\dagger(z)b_k(z)\,, \tag{A2}$$

where $\mathcal{A}$ is the cross-sectional area of the beam, and L^3 is the volume of quantization. We can also define a localized momentum operator,

$$\begin{aligned}\mathcal{P}(z_0,t)&=\hat{z}\mathcal{H}(z_0,t)/c\\&=\hat{z}\frac{L^3}{cd}\int_{z_0-d/2}^{z_0+d/2}H(z,t)dz\,,\end{aligned} \tag{A3}$$

where $H(z,t)$ is the Hamiltonian density, and $\mathcal{H}(z_0,t)$ is the Hamiltonian corresponding to a system which has the same Hamiltonian density

$$(1/d)\int_{z_0-d/2}^{z_0+d/2}H(z,t)dz$$

everywhere in the volume L^3. Therefore, $\mathcal{H}(z_0,t)$ here has the same form as given for the various cases discussed in this paper, but with $b_k(z_0)$ and $b_k^\dagger(z_0)$ replacing a_k and $a_k^\dagger$, assuming that the medium has a uniform density $N(z_0)$, which fills the entire quantization volume for free fields,

$$\mathcal{P}(z,t)=\hat{z}\sum_k \hbar k[b_k^\dagger(z)b_k(z)+\tfrac{1}{2}].$$

The momentum operator acts as a translation operator:

$$\begin{aligned}d\psi(z)/dz&=(-1/i\hbar)[\psi(z),\mathcal{P}(z)]\,,\\dE^{(-)}(z)/dz&=(-1/i\hbar)[E^{(-)}(z),\mathcal{P}(z)]\,.\end{aligned} \tag{A4}$$

Thus, for example, in the case of sum-frequency generation, Eq. (A4) yields

$$\begin{aligned}dE_k^{(-)}(z)/dz-ikE_k^{(-)}(z)&=i[2\pi\omega_k/c\epsilon(z)]\\&\quad\times N(z)\hat{e}_k\cdot\mathbf{p}^{(2)}:\hat{e}_{k_0}\hat{e}_{k_0'}E_{k_0}(z)E_{k_0'}(z)\,,\end{aligned} \tag{A5}$$

which agrees with the corresponding classical equation. According to Eq. (A4), the unitary translation operator is

$$U(z,z_0)=\left\{\exp\left[(i/\hbar)\int_{z_0}^{z}\mathcal{P}(z)dz\right]\right\}_+\,. \tag{A6}$$

Here, the space-ordered product $\{\ \}_+$ has the similar definition as the time-ordered product. Field operators at different spatial points are connected by this unitary operator:

$$E(z,t)=U^{-1}(z,z_0)E(z_0,t)U(z,z_0)\,. \tag{A7}$$

We can now define a localized density matrix operator,

$$\rho(z)=U(z,0)\rho(0)U^{-1}(z,0)\,, \tag{A8}$$

assuming free space for $z<0$. Then the correlation function of fields at different times is given by

$$\begin{aligned}&\langle E^{(+)}(z,t_1)\cdots E^{(+)}(z,t_n)E^{(-)}(z,t_n)\cdots E^{(-)}(z,t_1)\rangle\\&=\mathrm{Tr}[\rho(0)E^{(+)}(z,t_1)\cdots E^{(+)}(z,t_n)E^{(-)}(z,t_n)\\&\qquad\times\cdots E^{(-)}(z,t_1)]\\&=\mathrm{Tr}[\rho(z)E^{(+)}(0,t_1)\cdots E^{(+)}(0,t_n)E^{(-)}(0,t_n)\\&\qquad\times\cdots E^{(-)}(0,t_1)]\,.\end{aligned} \tag{A9}$$

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

$$\partial\rho/\partial z=(-1/i\hbar)[\mathcal{P}(z),\rho(z)]\,. \tag{A10}$$

With the help of these localized operators, the calculations for steady-state propagation in a medium become exactly the same as the corresponding calculations for a cavity with t replaced by $-z/c$.

Physically, the density matrix $\rho(z)$ describes an ensemble of photon systems which has all the statistical properties of fields at z. If a photon system is taken as the section of the light beam emerged from the plane at z in a time T, where T can be the counting time of photodetectors,[9] then $\rho(z)$ actually describes an ensemble of such photon systems. This is the ensemble we measure in experiments.

The problem of beam splitting has been deliberately avoided in this paper. It requires some modification of our formalism. Qualitatively, the split beams would have different statistical properties than the unpslit beam, and they are correlated with each other. The equivalent problem in the cavity case corresponds to the splitting of the photon ensemble with time.

[26] L. Mandel, Phys. Rev. **144**, 1071 (1966).