Quantum statistics and photon correlation effects in parametric fluorescence

S. T. Gevorkyan and G. Yu. Kryuchkov

Institute of Physics Research, Academy of Sciences of the Armenian SSR, Erevan
(Submitted 12 December 1986)
Zh. Eksp. Teor. Fiz. **92**, 2034–2049 (June 1987)

A quantum electrodynamic theory is given of parametric mixing of two modes of a radiation field in an atomic gaseous medium subjected to a strong monochromatic pump field. The problems of quantum fluctuations and spatial evolution of photon statistics are considered as well as nonlinear (with respect to the pump field) effects of photon correlations in parametric fluorescence. Full allowance is made not only for stimulated emission processes, but also for spontaneous processes and in particular for the emission of photon pairs. Calculations are made of the mode intensities in a medium, correlation functions of the field amplitudes,and occupation numbers of two modes with Gaussian or Poisson photon statistics at the point of incidence, and also in the case of spontaneous parametric fluorescence. The calculations are carried out for atomic media under one- and two-photon resonance conditions, allowing for saturation effects.

§ 1. INTRODUCTION

The phenomenon of parametric mixing of weak waves or parametric fluorescence in an atomic gaseous medium subjected to a strong optical field has been thoroughly investigated mainly in connection with applications in frequency conversion.[1,2] Problems of this kind are usually solved by a semiclassical approach based on the Maxwell-Bloch equations which have been investigated in detail under steady-state conditions without expansion in terms of the monochromatic pump field and only for the simplest atomic systems (see, for example, Refs. 3 and 4). In the semiclassical approach the passage to the case of spontaneous parametric fluorescence, when there are no weak fields at the entry to a medium, is made in a formal manner by replacing the energy density of the input signal with the energy density of zero-point oscillations of the electromagnetic field. To the best of our knowledge, no systematic quantum analysis of this effect has yet been made without applying perturbation theory to the pump field.

Much less work has been done on the statistics of quantum fluctuations which originate from spontaneous noise in the interaction of a radiation field with atoms and are amplified parametrically to macroscopic amplitudes. Recent theoretical and experimental investigations of these topics are attracting much attention because of photon correlation under the influence of a laser field[5,6] and because of the widely discussed possibilities, in principle, of suppressing quantum fluctuations in nonlinear optics[7–9] and constructing macroscopic light sources characterized by sub-Poisson photon statistics[10,11] (i.e., with less dispersion of the number of photons than in the Poisson case). Nevertheless, the present level of understanding of statistical characteristics of quantum fluctuations in parametric processes is not fully satisfactory. One of the reasons for this situation is that the approaches already employed[5,12,13] are based on the effective Hamiltonian method in nonlinear optics and do not allow fully for spontaneous processes, particularly for two-photon emission. Moreover, no allowance is made for the effects of the influence of the pump field. However, it is known that these processes have a decisive influence in the case of photon correlation effects involving single atoms.[14–16]

The purpose of the present investigation will be to investigate the topics mentioned above and some other unsolved problems associated with parametric fluorescence under the influence of a monochromatic pump field.

We shall develop a quantum electrodynamic theory of parametric mixing of radiation field modes in which one- and two-photon spontaneous processes are described in a natural manner and we shall allow also for the dependence on the intensity of the pump field (see § 2). The specific results will be obtained without allowance for the contribution of atomic collisions or Doppler broadening in two cases (see § 3): a) an atomic gaseous medium composed of identical two-level atoms in the presence of a monochromatic resonance field of frequency ω ($|\omega_{ba}-\omega|\ll\omega_{ba}$, where ω_{ba} is the frequency of an atomic transition); b) a medium consisting of multilevel atoms subjected to a monochromatic field in the case of a two-photon resonance between levels of the same parity ($|\omega_{ba}-2\omega|\ll\omega_{ba}$, see Fig. 1 and § 3.2). It is known[3,4] that in both cases a wave of arbitrary frequency ω_1 incident on a medium generates a "mirror wave" of frequency $\omega_2=2\omega-\omega_1$.[1)]

We shall obtain solutions for the steady-state case giving the average number of photons in modes and the correlation function of the amplitudes of two parametrically coupled modes (§§ 4 and 5), as well as fourth-order moments, particularly of the correlation functions of the number of photons used to determine the statistics of paired photo-

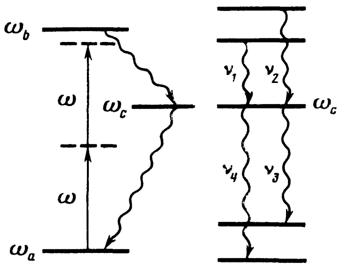

FIG. 1. Atomic energy levels (a) and quasienergy states of an atom (b) under the conditions of a two-photon resonance $|\bar{\omega}_{ba}-2\omega|\ll\bar{\omega}_{ba}$ between levels ω_b and ω_a of the same parity. The two-photon transition ($|\varphi_a\rangle\rightleftarrows|\varphi_b\rangle$) is induced by a pump field of frequency ω and the wavy lines in the diagram b represent the frequencies of transitions between quasienergy states.

0038-5646/87/061145-09$04.00

counts (§ 6). Next, in §§ 7 and 8 we shall consider the cases of spatial evolution of modes with Gaussian, Poisson, or sub-Poisson statistics on entering the medium.

§ 2. KINETIC EQUATIONS FOR CORRELATION FUNCTIONS

In this section we shall provide a general statistical description of a subsystem containing two modes of a quantized radiation field in an atomic medium in the presence of a classical pump field. This field is assumed to be monochromatic with frequency ω; the states of the two modes in a volume v with momenta $\mathbf{k}_i$, frequencies ω_i, and polarization vectors $\mathbf{e}_{\lambda i}(\mathbf{k}_i) = \mathbf{e}(i)$ will be denoted by the indices 1 and 2 ($i = 1,2$). In a quantum description the starting stage will be application of the density matrix method (see, for example, Refs. 5 and 19). We shall formulate this method in the quasienergy state representation[20] of a system comprising an atom and a monochromatic pump field. Such a modification of the method made it possible to allow for the effects of the influence of the pump field outside the perturbation theory framework when describing the evolution in time of the statistical characteristics of radiation field modes.

The moments of the radiation field (necessary for the description of interference experiments and measurements of photocount statistics), i.e., the quantum averages $\langle M(t)\rangle$ over the initial state of the complete system, of the products

$$M(t)=a_1^+(t)^m a_2^+(t)^n a_2(t)^k a_1(t)^l \tag{2.1}$$

of the Heisenberg creation and annihilation operators

$$a_i^+(t)=a^+_{\mathbf{k}_i\lambda_i}(t), \qquad a_i(t)=a_{\mathbf{k}_i\lambda_i}(t),$$

can be represented by the formula

$$\langle M(t)\rangle = \mathrm{Tr}_F(\rho_F(t)M_0(t)). \tag{2.2}$$

Here, the operator $M_0(t)$ is described by Eq. (2.1) written in terms of free field operators

$$a_{0i}(t)=a_i\exp(-i\omega_i t),$$

whereas Tr_F is a trace of the states of the two modes in question. The density matrix of the radiation field modes

$$\rho_F(t) = \mathrm{Tr}'(\rho(t))$$

is a sum of diagonal elements of the density operator $\rho(t)$ of the complete system over the states of the subsystem comprising the atom, the monochromatic field, and the radiation field minus the two modes $\omega_{1,2}$. In this formulation we have

$$\rho(t)=S(t)|\psi_0\rangle\langle\psi_0|S^+(t), \tag{2.3}$$

where the quantity $S(t) = S(t, -\infty)$ represents the scattering matrix in the representation of quasienergy states[21] with the interaction operator in the dipole approximation

$$W(t)=-i\sum_j\left(\frac{2\pi\hbar\omega_j}{v}\right)^{1/2}(a_j\mathbf{e}_je^{-i\omega_jt}-a_j^+\mathbf{e}_j^*e^{i\omega_jt})\mathbf{d}(t). \tag{2.4}$$

Here, $\mathbf{d}(t)$ is the operator of the dipole moment of the atom in the quasienergy state representation, v is the quantization volume, the initial state considered in the limit $t\to-\infty$ is $|\psi_0\rangle = |\Phi_0\rangle|\psi_F\rangle$, and it contains the initial quasienergy state $|\Phi_0\rangle$ and the initial state $|\psi_F\rangle$ of the radiation field.

The equations for $\rho(t)$ and, consequently, for $\rho_F(t)$ follow from the equations for the scattering matrix. In the Markov approximation, retaining only the terms of second order in the interaction $W(t)$, we obtain

$$\begin{aligned}\frac{\partial}{\partial t}\rho_F(t)=&\frac{2\pi\omega_1}{\hbar v}\{[a_1\rho_F(t),a_1^+]e_n^*(1)e_m(1)\Delta^{(0)}_{nm}(\omega_1)\\&+[a_1^+\rho_F(t),a_1]e_n(1)e_m^*(1)\Delta^{(0)}_{nm}(-\omega_1)+\text{H.c.}\}\\&+\frac{2\pi}{\hbar v}(\omega_1\omega_2)^{1/2}\{[a_2,a_1\rho_F(t)]e_n(2)e_m(1)\Delta^{(2)}_{nm}(\omega_1)\\&+[a_2^+,a_1^+\rho_F(t)]e_n^*(2)e_m^*(1)\Delta^{(-2)}_{nm}(-\omega_1)+\text{H.c.}\}\\&+(\omega_1,e(1))\rightleftharpoons(\omega_2,e(2)).\end{aligned} \tag{2.5}$$

This equation is symmetric under photon transposition, n and m are vector indices, and the coefficients are given by

$$\Delta^{(q)}_{nm}(\omega_i)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}dt\exp(-iq\omega t)\Delta_{nm}(\omega_i,t), \tag{2.6}$$

$$\Delta_{nm}(\omega_i,t)=\int_{-\infty}^{t}dt_1\exp[i\omega_i(t-t_1)]\langle\Phi_0,0|\mathscr{D}_n(t)\mathscr{D}_m(t_1)|\Phi_0,0\rangle. \tag{2.7}$$

In Eq. (2.7) we introduced the notation $\mathscr{D}_n(t) = S^+(t)d_n(t)S(t)$ and carried quantum averaging out over the state $|\Phi_0,0\rangle = |\Phi_0\rangle|0\rangle$, where $|0\rangle$ is the vacuum state of the radiation field. We now comment on the method used to derive the kinetic equation (2.5). It applies to problems of parametric mixing of two modes of a radiation field. Therefore, the terms containing exponential functions oscillating after long time intervals are eliminated from the above equation using the relationship $\omega_1+\omega_2=2\omega$ when $\omega_1\neq\omega_2\neq\omega$. In problems of this kind the condition for the phase matching of modes results in selection of photon pair emission processes, so that the restriction to second order in the interaction W in the weak-mode approximation is quite satisfactory.

Equations (2.6) and (2.2) allow us to derive kinetic equations for arbitrary moments. We shall give them for a medium in a volume v where the atomic density is N. We shall consider a one-dimensional model of the propagation of radiation field modes along the x axis in the constant-monochromatic-pump-field approximation under steady-state conditions. We shall ignore also the cooperative effects, i.e., we shall assume that the averages of Eq. (2.7) are small for atoms at different points in the medium. When these approximations are made, the transformation from a temporal to a spatial description of the averages of the field operators $a_i(x)$ varying slowly along the x axis can be made in the usual way[19] employing the substitution $t\to x/c$ and reconstructing the dependences on the momenta in the field amplitudes $a_i\to a_i(x)e^{ik_ix}$ (or more rigorously using the technique of wave packets—see, for example, Ref. 5). Consequently, the average number of photons in the modes

$$n_i(x)=\langle a_i^+(x)a_i(x)\rangle\cdot$$

and the correlation function of the field amplitudes

$$g(x)=\langle a_1(x)a_2(x)\rangle$$

are described by

$$\frac{\partial}{\partial x} n_{1,2}(x) = 2\,\mathrm{Re}\,\alpha_{1,2} n_{1,2}(x) + 2\,\mathrm{Re}[\mu_{2,1} e^{-i\Delta kx} g(x)] + \beta_{1,2}, \tag{2.8}$$

$$\frac{\partial}{\partial x} g(x) = \mathrm{Re}(\alpha_1+\alpha_2) g(x) + [\mu_1^* n_1(x) + \mu_2^* n_2(x) + \lambda^*] e^{i\Delta kx} \tag{2.9}$$

(a comma is used to separate the equations for the two modes). The coefficients of these equations are

$$\alpha_i = \frac{2\pi\omega_i N}{\hbar c} e_n^*(i) e_m(i) \left(\Delta_{nm}^{(0)*}(-\omega_i) - \Delta_{nm}^{(0)}(\omega_i)\right), \tag{2.10}$$

$$\beta_i = \frac{4\pi\omega_i N}{\hbar c} \mathrm{Re}\left(e_n(i) e_m^*(i) \Delta_{nm}^{(0)}(-\omega_i)\right), \tag{2.11}$$

$$\mu_1 = \frac{2\pi N}{\hbar c} (\omega_1\omega_2)^{1/2} e_n(2) e_m(1) \left(\Delta_{nm}^{(-2)*}(-\omega_1) - \Delta_{nm}^{(2)}(\omega_1)\right), \tag{2.12}$$

$$\lambda = \frac{2\pi N}{\hbar c} (\omega_1\omega_2)^{1/2} \left(e_n(2) e_m(1) \Delta_{nm}^{(-2)*}(-\omega_1) + e_n(1) e_m(2) \Delta_{nm}^{(-2)*}(-\omega_2)\right), \tag{2.13}$$

where μ_2 is obtained from Eq. (2.12) by the substitution $[\omega_1, e(1)] \rightleftarrows [\omega_2, e(2)]$. It should be pointed out that the imaginary parts Im α_i, which determine the dispersion of the waves, are included in the momenta $\tilde{k}_i = k_i + \mathrm{Im}\,\alpha_i$, and the momenta of a pump field in a medium $\tilde{k}$ are taken out from the quantities $\Delta^{(2)}$, and $\Delta^{(-2)*}$ (this will be explained further in the next section). Here, $\Delta k = 2\tilde{k} - \tilde{k}_1 - \tilde{k}_2$ represents the x component of the difference between the wave vectors in the medium.

We shall conclude this section by using Eq. (2.2) to give the kinetic equations for the moments $\langle M(x)\rangle$,

$$M = a_1^+(x)^m a_2^+(x)^n a_2(x)^k a_1(x)^l,$$

of the field operators $a_i(x)$ $[a_i(0) = a_i]$ varying slowly along the x axis:

$$\frac{\partial}{\partial x}\langle M(x)\rangle = \sum_{i=1}^{2}\Big\{\mathrm{Re}\,\alpha_i\Big(\Big\langle\frac{\partial M}{\partial a_i(x)} a_i(x)\Big\rangle + \Big\langle a_i^+(x)\frac{\partial M}{\partial a_i^+(x)}\Big\rangle\Big) + \beta_i\Big\langle\frac{\partial^2 M}{\partial a_i^+(x)\,\partial a_i(x)}\Big\rangle\Big\} + \Big\{\mu_1\Big\langle\frac{\partial M}{\partial a_2^+(x)} a_1(x)\Big\rangle + \mu_2\Big\langle\frac{\partial M}{\partial a_1^+(x)} a_2(x)\Big\rangle + \lambda\Big\langle\frac{\partial^2 M}{\partial a_1^+(x)\,\partial a_2^+(x)}\Big\rangle\Big\} e^{-i\Delta kx} + \Big\{\mu_1^*\Big\langle a_1^+(x)\frac{\partial M}{\partial a_2(x)}\Big\rangle + \mu_2^*\Big\langle a_2^+(x)\frac{\partial M}{\partial a_1(x)}\Big\rangle + \lambda^*\Big\langle\frac{\partial^2 M}{\partial a_1(x)\,\partial a_2(x)}\Big\rangle\Big\} e^{i\Delta kx}. \tag{2.14}$$

The above system of equations describes the evolution of the moments of two modes due to their interaction with the atoms in the medium in the presence of a pump field and allowing for relaxation processes.

§ 3. CALCULATION OF THE COEFFICIENTS

The coefficients in the system of equations (2.14) described by Eqs. (2.6), (2.7), and (2.10)–(2.13) are given in their general form for arbitrary atomic systems. In the present section we shall calculate these coefficients for atomic systems mentioned in the Introduction in those cases when the separation between spectral lines is much greater than their width. We shall carry out these calculations using the density matrix method applied to quasienergy states, which can be described as follows[2]:

$$\sigma_{ij}(t) = S^+(t)\,|\Phi_i\rangle\langle\Phi_j|\,S(t). \tag{3.1}$$

Writing down the operator $\vec{\mathscr{D}}(t)$ in terms of the matrix element $\mathbf{d}_{ij}(t)$ of a dipole transition between quasienergy states

$$\vec{\mathscr{D}}(t) = S^+(t)\,\mathbf{d}(t)\,S(t) = \sum_{i,j}\sigma_{ij}(t)\,\mathbf{d}_{ij}(t), \tag{3.2}$$

we find that determination of the function (2.7) reduces to calculation of averages of the type

$$\langle\Phi_0,\ 0|\sigma_{ij}(t)\sigma_{pk}(t_1)|\Phi_0,\ 0\rangle. \tag{3.3}$$

This calculation can be carried out using the quantum theorem on regression of fluctuations,[22] according to which the quantities of Eq. (3.3) satisfy in the $t > t_1$ case the same equations as the averages $\langle\Phi_0, 0|\sigma_{ij}(t)|\Phi_0, 0\rangle = \bar{\sigma}_{ij}(t)$.

3.1. Two-level atom in a resonant field. We shall describe this system employing the following notation: $\Delta = \omega_{ba} - \omega$ is the detuning from resonance, $\Omega = (\Delta^2 + 4|V|^2)^{1/2}$ is the Rabi frequency, $V = \mathbf{E}_0\mathbf{d}/\hbar$ is the matrix element of the interaction of an atom with the pump field, where $\mathbf{E}_0$ is the amplitude of the field intensity and $\mathbf{d} = \langle\varphi_b|\hat{\mathbf{d}}|\varphi_a\rangle$ is the matrix element of the dipole transition; γ is the spontaneous width of the atomic transition. We shall consider the case of a "strong field" corresponding to $\Omega \gg \gamma$, when the system has separate spectral lines at a resonance frequency $\omega_r = \omega + \Omega$ and at a "three-photon" frequency $\omega_t = \omega - \Omega$.

The averages of Eq. (3.3) obtained for the case $|\Phi_0\rangle = |\Phi_a\rangle$ calculated to within terms of order γ/Ω are given in Ref. 15. Using these results and the known matrix elements of the transitions between quasienergy states of a two-level atom $\mathbf{d}_{ij}(t)$ (Ref. 23), we find that in the relaxed regime $t \gg \gamma^{-1}$ when $\Delta > 0$, we obtain

$$\mathrm{Re}\,\alpha_i = \frac{\pi\omega_i N}{2\hbar c}|\mathbf{e}(i)\mathbf{d}|^2\Gamma(\sigma_{aa}-\sigma_{bb}) \times\left[\frac{(1-\Delta/\Omega)^2}{(\omega_i-\omega_t)^2+\Gamma^2} - \frac{(1+\Delta/\Omega)^2}{(\omega_i-\omega_r)^2+\Gamma^2}\right], \tag{3.4}$$

$$\beta_i = \frac{8\pi\omega_i N}{\hbar c}|\mathbf{e}(i)\mathbf{d}|^2 \times\frac{\Gamma|V|^4}{\Omega^2(\Omega^2+\Delta^2)}\left[\frac{1}{(\omega_i-\omega_t)^2+\Gamma^2} + \frac{1}{(\omega_i-\omega_r)^2+\Gamma^2}\right], \tag{3.5}$$

$$\mu_1 = i\frac{2\pi N}{\hbar c}(\omega_1\omega_2)^{1/2}(\mathbf{e}(1)\mathbf{d})(\mathbf{e}(2)\mathbf{d})\frac{(V^*)^2}{\Omega^2}(\sigma_{aa}-\sigma_{bb}) \times\left[\frac{1}{\omega_1-\omega_r+i\Gamma} - \frac{1}{\omega_1-\omega_t+i\Gamma}\right], \tag{3.6}$$

$$\lambda = -i\frac{2\pi N}{\hbar c}(\omega_1\omega_2)^{1/2}(\mathbf{e}(1)\mathbf{d})(\mathbf{e}(2)\mathbf{d})\frac{(V^*)^2}{\Omega^2}\left[\sigma_{aa}\left(\frac{1}{\omega_1-\omega_t+i\Gamma} + \frac{1}{\omega_2-\omega_t+i\Gamma}\right) + \sigma_{bb}\left(\frac{1}{\omega_1-\omega_r+i\Gamma} + \frac{1}{\omega_2-\omega_r+i\Gamma}\right)\right]. \tag{3.7}$$

The spectral width which occurs in these expressions is

$$\Gamma=\gamma(1+2|V|^2/\Omega^2)/2,$$

whereas the difference between the steady-state values of the populations of quasienergy states

$$\sigma_{ii}=\bar{\sigma}_{ii}(t)|_{t\gg\Gamma^{-1}}$$

considered in the $\gamma/\Omega\ll 1$ approximation is

$$\sigma_{aa}-\sigma_{bb}=2\Omega\Delta/(\Delta^2+\Omega^2). \tag{3.8}$$

In the constant-pump-field approximation we have adopted,

$$E(x)=E_0\exp(i\tilde{k}x),$$

where $\tilde{k}$ is the momentum taken allowing for the phase modulation of the field,[3] we find that only the field phase changes in the medium. This dependence on x is separated in the kinetic equations [it is contained in the terms $\exp(i\Delta kx)$] and it is therefore omitted from the coefficients μ_i and λ.

The coefficients (3.4)–(3.7) depend nonlinearly on the pump field amplitude. In the limiting case of $|V|\gg\Delta$, and $\sigma_{aa}\approx\sigma_{bb}$, we consequently have $\operatorname{Re}\alpha_i=\mu_i=0$ to within terms of order γ/Ω. In this range the nonzero coefficients (3.5) and (3.7) will be denoted by $\beta_{i,\mathrm{sat}}$ and λ_{sat}.

3.2. Atomic system under two-photon resonance conditions. It is assumed that the conditions are favorable for coherent and not multistage two-photon excitation of a level ω_b accompanied mainly by decay via a level ω_c (Fig. 1). The transition amplitude is governed by a two-photon matrix element

$$V_2=\sum_k V_{2k}V_{k1}/(\omega_{k1}-\omega), \tag{3.9}$$

where V_{2k} and V_{k1} are matrix elements of one-photon transitions. In the range $\Omega_2\gg\gamma_{bc}$, γ_{ca}, where $\Omega_2=(\varepsilon^2+4|V_2|^2)^{1/2}$ is the two-photon Rabi frequency and γ_{bc} and γ_{ca} are the spontaneous partial widths of the transitions $|\varphi_b\rangle\to|\varphi_c\rangle$, and $|\varphi_c\rangle\to|\varphi_a\rangle$, respectively, the four spectral lines of the system are separated on the frequency scale. They are governed by dipole transitions via quasienergy states and their frequencies are[24]

$$\nu_1=\bar{\omega}_b-\bar{\omega}_c-(\varepsilon+\Omega_2)/2,\qquad \nu_2=\bar{\omega}_b-\bar{\omega}_c-(\varepsilon-\Omega_2)/2,$$
$$\nu_3=\bar{\omega}_c-\bar{\omega}_a-(\varepsilon+\Omega_2)/2,\qquad \nu_4=\bar{\omega}_c-\bar{\omega}_a-(\varepsilon-\Omega_2)/2, \tag{3.10}$$

where $\bar{\omega}_i$ are the frequencies of atomic levels with non-resonant Stark shifts and $\varepsilon=\bar{\omega}_{ba}-2\omega$.

The approximation in which the spectral lines do not cross, $|\nu_i-\nu_j|\gg\gamma_{bc},\gamma_{ca}$, is used in Ref. 25 to obtain equations for the density matrix of quasienergy states $\bar{\sigma}_{ij}(t)$. We shall give the final results of calculations of the coefficients (2.10)–(2.13) in accordance with the scheme proposed above and we shall do this using the equations given above as well as the familiar expressions[24,25] for dipole transitions between quasienergy states:

$$\operatorname{Re}\alpha_i=\frac{2\pi\omega_iN}{\hbar c}\left\{|\mathbf{e}^*(i)\mathbf{d}_1|^2\left[\frac{(\sigma_{bb}-\sigma_{cc})\cos^2\theta\Gamma_a}{\Gamma_a^2+(\omega_i-\nu_2)^2}+\frac{(\sigma_{aa}-\sigma_{cc})\sin^2\theta\Gamma_b}{\Gamma_b^2+(\omega_i-\nu_1)^2}\right]\right.$$
$$\left.+|\mathbf{e}^*(i)\mathbf{d}_2|^2\left[\frac{(\sigma_{cc}-\sigma_{aa})\cos^2\theta\Gamma_b}{\Gamma_b^2+(\omega_i-\nu_4)^2}+\frac{(\sigma_{cc}-\sigma_{bb})\sin^2\theta\Gamma_a}{\Gamma_a^2+(\omega_i-\nu_3)^2}\right]\right\}, \tag{3.11}$$

$$\beta_i=\frac{4\pi\omega_iN}{\hbar c}\left\{|\mathbf{e}^*(i)\mathbf{d}_1|^2\left[\frac{\sigma_{aa}\sin^2\theta\Gamma_b}{\Gamma_b^2+(\omega_i-\nu_1)^2}+\frac{\sigma_{bb}\cos^2\theta\Gamma_a}{\Gamma_a^2+(\omega_i-\nu_2)^2}\right]\right.$$
$$\left.+|\mathbf{e}^*(i)\mathbf{d}_2|^2\sigma_{cc}\left[\frac{\cos^2\theta\Gamma_b}{\Gamma_b^2+(\omega_i-\nu_4)^2}+\frac{\sin^2\theta\Gamma_a}{\Gamma_a^2+(\omega_i-\nu_3)^2}\right]\right\}, \tag{3.12}$$

$$\mu_1=\frac{2\pi N}{\hbar c}(\omega_1\omega_2)^{1/2}\frac{V_2^*}{\Omega_2}\left\{(\mathbf{e}(1)\mathbf{d}_1^*)(\mathbf{e}(2)\mathbf{d}_2^*)\left[\frac{\sigma_{bb}-\sigma_{cc}}{\Gamma_a+i(\nu_2-\omega_1)}+\frac{\sigma_{cc}-\sigma_{aa}}{\Gamma_b+i(\nu_1-\omega_1)}\right]\right.$$
$$\left.+(\mathbf{e}(2)\mathbf{d}_1^*)(\mathbf{e}(1)\mathbf{d}_2^*)\left[\frac{\sigma_{cc}-\sigma_{bb}}{\Gamma_a+i(\nu_3-\omega_1)}+\frac{\sigma_{aa}-\sigma_{cc}}{\Gamma_b+i(\nu_4-\omega_1)}\right]\right\}, \tag{3.13}$$

$$\lambda=\frac{2\pi N}{\hbar c}(\omega_1\omega_2)^{1/2}\frac{V_2^*}{\Omega_2}\left\{(\mathbf{e}(1)\mathbf{d}_1^*)(\mathbf{e}(2)\mathbf{d}_2^*)\times\left[\frac{\sigma_{bb}}{\Gamma_a+i(\nu_2-\omega_1)}-\frac{\sigma_{aa}}{\Gamma_b+i(\nu_1-\omega_1)}\right]\right.$$
$$+(\mathbf{e}(2)\mathbf{d}_1^*)(\mathbf{e}(1)\mathbf{d}_2^*)\left[\frac{\sigma_{cc}}{\Gamma_a+i(\nu_3-\omega_1)}-\frac{\sigma_{cc}}{\Gamma_b+i(\nu_4-\omega_1)}\right]$$
$$\left.+(\omega_1,e(1))\rightleftarrows(\omega_2,e(2))\right\}. \tag{3.14}$$

In these expressions the spectral widths and the steady-state values of the populations of quasienergy states are given by

$$2\Gamma_{a,b}=\gamma_{ca}+\gamma_{bc}(1\pm|\varepsilon|/\Omega_2)/2, \tag{3.15}$$

$$\sigma_{aa}=\frac{n_a^2}{n_b^2}\sigma_{bb}=\frac{n_a\gamma_{ca}}{n_b\gamma_{bc}}\sigma_{cc}=\frac{n_a\gamma_{ca}}{n_b\gamma_{ca}+(n_a+n_b^2/n_a)\gamma_{bc}}. \tag{3.16}$$

Here the following notation is employed:

$$\mathbf{d}_1=\langle\varphi_c|\hat{\mathbf{d}}|\varphi_b\rangle,\qquad \mathbf{d}_2=\langle\varphi_a|\hat{\mathbf{d}}|\varphi_c\rangle,\qquad \cos\theta=(1+\varepsilon/\Omega_2)^{1/2},$$
$$n_{a,b}=(1\pm|\varepsilon|/\Omega_2)/2.$$

We shall also show that for this system the phase modulation of the pump field is due to nonresonant transitions and is unimportant (i.e., $\tilde{k}=k$),[4,18] in contrast to the case of a two-level atom.

§ 4. GROWTH OF PARAMETRIC FLUORESCENCE FROM SPONTANEOUS PROCESSES

It is convenient to discuss this topic and to establish the correspondence with the results for single atoms, starting with an analysis of Eqs. (2.8) and (2.9) for short propagation lengths. We shall consider the case when only one of the modes of frequency ω_1 enters a medium. The average number of photons in this mode is $n_1(0)$, whereas the number of photons of a new mode which appears at a frequency $2\omega-\omega_1$, is $n_2(x)$, where $n_2(0)=0$ is obtained in the lower orders of the expansion in the length, and these numbers are described by

$$n_1(x)=n_1(0)+(\beta_1+2\operatorname{Re}\alpha_1n_1(0))x+\dots, \tag{4.1}$$

$$n_2(x)=\beta_2x+(\beta_2\operatorname{Re}\alpha_2+\operatorname{Re}(\mu_1^*\lambda)+|\mu_1|^2n_1(0))x^2+\dots. \tag{4.2}$$

The correlation function of the amplitudes of the two modes is

$$g(x)=(\lambda^*+\mu_1^*n_1(0))x+\dots. \tag{4.3}$$

The physical meaning of these results is quite clear. The quantities $\beta_{1,2}$ and λ describe spontaneous processes which in the vacuum case with $n_1(0) = 0$ give rise to weak "bare" fields at frequencies $\omega_{1,2}$. The coefficient β with the factor c/Nv describes the rate of emission of photons of frequency $\omega_i \neq \omega$ by a single atom in the course of resonance fluorescence in the relaxed regime. The quantity λ is the amplitude of the process of spontaneous decay of the pump photons into a pair of photons of frequencies ω_1 and ω_2 related by the conservation law $\omega_1 + \omega_2 = 2\omega$; this amplitude is taken per unit length.

We shall now consider reduced propagation equations for slowly varying creation and annihilation operators for photons in the two modes:

$$\frac{\partial}{\partial x} a_1(x) = \mathrm{Re}\,\alpha_1 a_1(x) + \mu_2^* \exp(i\Delta k x) a_2^+(x) + f_1(x),$$
$$\frac{\partial}{\partial x} a_2^+(x) = \mathrm{Re}\,\alpha_2 a_2^+(x) + \mu_1 \exp(-i\Delta k x) a_1(x) + f_2^+(x). \tag{4.4}$$

The form of these equations differs from the semiclassical equations because of the contribution of the Langevin noise operators $f_{1,2}$, where $\langle f_{1,2}(x)\rangle = 0$.

The coefficients $\mathrm{Re}\,\alpha_i$ and μ_i represent the polarizabilities of an atom experiencing an external weak field. A generalization of the Einstein formula for the diffusion coefficients[22] and Eqs. (2.8) and (2.9) for nonzero correlation functions of the Langevin operators yield

$$\langle f_i^+(x) f_i(x')\rangle = \frac{\beta_i}{\beta_i - 2\,\mathrm{Re}\,\alpha_i} \langle f_i(x) f_i^+(x')\rangle = \beta_i \delta(x-x'), \tag{4.5}$$

$$\langle f_2(x) f_1(x')\rangle = \frac{\lambda^* - \mu_2^*}{\lambda^* - \mu_1^*} \langle f_1(x) f_2(x')\rangle$$
$$= \exp(i\Delta k x)\,(\lambda^* - \mu_2^*)\,\delta(x-x'). \tag{4.6}$$

In a phenomenological analysis (see, for example, Ref. 8) the correlation functions $\langle f_1 f_2\rangle$, and $\langle f_2 f_1\rangle$ are usually assumed to be zero and $\langle f_i^+ f_i\rangle$ does not allow for the frequency dependence and the effects of the intensity of the pump field. However, using the results of Eqs. (3.4)–(3.7) and (3.11)–(3.14), we can readily show that the correlation functions of Eq. (4.6) are comparable in magnitude with those given by Eq. (4.5).[3)] We shall now give the formal solutions of the operator equations:

$$a_1(x) = \varphi_2(x) a_1(0) + \mu_2^* \exp\left(i\frac{\Delta k}{2}x\right) \varphi_0(x) a_2^+(0)$$
$$+ \int_0^x dx_1 \Big[\varphi_2(x-x_1) f_1(x_1) + \mu_2^* \exp\left(i\frac{\Delta k}{2}(x+x_1)\right) \varphi_0(x-x_1) f_2^+(x_1)\Big], \tag{4.7}$$
$$a_2^+(x) = \varphi_1(x) a_2^+(0) + \mu_1 \exp\left(-i\frac{\Delta k}{2}x\right) \varphi_0(x) a_1(0)$$
$$+ \int_0^x dx_1 \Big[\varphi_1(x-x_1) f_2^+(x_1) + \mu_1 \exp\left(-i\frac{\Delta k}{2}(x+x_1)\right) \varphi_0(x-x_1) f_1(x_1)\Big].$$

The following notation is used in these solutions:

$$\varphi_0(x) = (g_+ - g_-)^{-1}[\exp(g_+ x) - \exp(g_- x)],$$

$$\varphi_{1,2}(x) = (g_+ - g_-)^{-1} \exp\left(\mp i\frac{\Delta k}{2}x\right) \times \Big[\left(g_+ - \mathrm{Re}\,\alpha_{1,2} \pm i\frac{\Delta k}{2}\right)\exp(g_+ x) - \left(g_- - \mathrm{Re}\,\alpha_{1,2} \pm i\frac{\Delta k}{2}\right)\exp(g_- x)\Big],$$

and the gains are given by the expressions

$$g_\pm = \tfrac{1}{2}\,\mathrm{Re}\,(\alpha_1 + \alpha_2) \pm \tfrac{1}{2}[(\mathrm{Re}\,\alpha_2 - \mathrm{Re}\,\alpha_1 + i\Delta k)^2 + 4\mu_2^*\mu_1]^{1/2}. \tag{4.8}$$

(Here and below, we use a combined notation for two quantities separated by a comma.) Averaging of the expressions in the system (4.7) over the initial coherence states of the radiation field $|\psi_F\rangle = |z_1\rangle|z_2\rangle$, and $a_i(0)\ |z_i\rangle = z_i|z_i\rangle$ gives the familiar solutions[3] of the semiclassical propagation equations.

We shall represent the average number of photons in a mode as a sum of two parts, one of which is "stimulated" and depends on the initial state of the two modes and the other is "spontaneous" and is equal to zero at the entry to the medium:

$$n_1(x) = n_{1\mathrm{st}}(x) + n_{1\mathrm{sp}}(x), \tag{4.9}$$

where these parts can be described by the following expressions if we use Eqs. (4.5) and (4.6) and assume that $\Delta k = 0$:

$$n_{1\mathrm{st}}(x) = A_1 \exp(2\,\mathrm{Re}\,g_+ x) + B_1 \exp(2\,\mathrm{Re}\,g_- x) - 2\,\mathrm{Re}\,[C_1 \exp((g_+^* - g_-)x)], \tag{4.10}$$

$$n_{1\mathrm{sp}}(x) = A_{10}[\exp(2\,\mathrm{Re}\,g_+ x) - 1] + B_{10}\frac{\mathrm{Re}\,g_+}{\mathrm{Re}\,g_-}[\exp(2\,\mathrm{Re}\,g_- x) - 1] - 4\,\mathrm{Re}\,g_+\,\mathrm{Re}\left[\frac{C_{10}}{g_+^* + g_-}[\exp((g_+^* + g_-)x) - 1]\right], \tag{4.11}$$

where the coefficients in front of the exponential functions are

$$A_1, B_1 = \left|\frac{g_{+,-} - \mathrm{Re}\,\alpha_2}{g_+ - g_-}\right|^2 \left\{ n_1(0) + \frac{|\mu_2|^2 n_2(0)}{|g_{+,-} - \mathrm{Re}\,\alpha_2|^2} + 2\,\mathrm{Re}\left(\frac{\mu_2 g(0)}{g_{+,-}^* - \mathrm{Re}\,\alpha_2}\right)\right\}, \tag{4.12}$$
$$C_1 = |g_+ - g_-|^{-2}\{(g_+^* - \mathrm{Re}\,\alpha_2)(g_- - \mathrm{Re}\,\alpha_2) n_1(0) + |\mu_2|^2 n_2(0) + \mu_2^*(g_+^* - \mathrm{Re}\,\alpha_2) g^*(0) + \mu_2(g_- - \mathrm{Re}\,\alpha_2) g(0)\},$$

and the quantities $g_\pm$ are given by Eq. (4.8) in the case when $\Delta k = 0$.

The component $n_{1\mathrm{st}}(x)$ with the boundary condition $n_{1\mathrm{st}}(0) = n_1(0)$ depends on $n_{1,2}(0)$ and on the correlation function $g(0)$. The component $n_{1\mathrm{sp}}(x)$, independent of the initial state of the modes of the radiation field, describes the change in the average number of photons emitted in one- and two-photon spontaneous processes. The coefficients A_{10}, B_{10}, and C_{10} are obtained from the coefficients A_1, B_1, and C_1 as a result of the following substitutions:

$$n_{1,2}(0) \to \beta_{1,2}/2\,\mathrm{Re}\,g_+, \quad g(0) \to \lambda^*/2\,\mathrm{Re}\,g_+, \tag{4.13}$$

where the first of these coefficients is given by

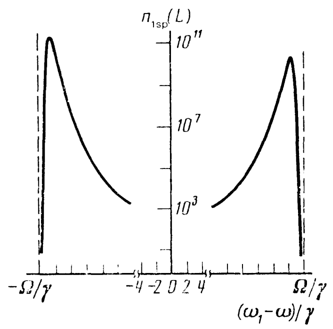

FIG. 2. Frequency dependence of the average number of photons at a distance $L = 1000\alpha_0^{-1}$ in a two-level medium obtained for the case when $2|V| = |\Delta|$. The parameter $\alpha_0 = 4\pi N\omega_{ba}|\mathbf{d}|^2/\hbar c\gamma$ is equal to 426 cm^{-1} for the $(6s^2)^1S_0 \to (6s6p)^1P_1^0$ transition in Ba atoms ($\omega_{ba} = 3.39\times10^{15}$ sec^{-1}, $|d| = 7.7\times10^{-18}$ cgs esu, $\gamma = 1.88\times10^8$ sec^{-1}) at atomic densities $N\approx10^{12}$ cm^{-3}. The curve is not given in the frequency range $|\omega_1-\omega| > \Omega$, where the phase-matching condition is not obeyed, and between the frequencies $\omega_1 = \omega \pm (\Omega - 12\gamma)$, i.e., far from the resonance and "three-photon" lines.

$$A_{10} = \frac{|g_+ - \mathrm{Re}\,\alpha_2|^2}{2\,\mathrm{Re}\,g_+|g_+-g_-|^2}\left\{\beta_1 + \frac{|\mu_2|^2\beta_2}{|g_+ - \mathrm{Re}\,\alpha_2|^2} + 2\,\mathrm{Re}\left(\frac{\mu_2\lambda^*}{g_+^* - \mathrm{Re}\,\alpha_2}\right)\right\}. \quad (4.14)$$

In the range $2\,\mathrm{Re}\,g_+x \gg 1$ the average number of photons in a mode rises exponentially along the propagation length

$$n_1(x) = (A_1 + A_{10})\exp(2\,\mathrm{Re}\,g_+x). \quad (4.15)$$

We shall now give numerical results for the case of spontaneous parametric fluorescence $n_1(0) = n_2(0) = g(0) = 0$ in a medium of two-level atoms. The dependence of the quantity $n_{1\mathrm{sp}}(x) = A_{10}\exp(2\,\mathrm{Re}\,g_+x)$ on the parameter $(\omega_1-\omega)/\gamma$ in the region of the resonance and "three-photon" lines is shown in Fig. 2. The curve is asymmetric near the frequencies $\omega_1 \approx \omega \pm \Omega$ [$n_{1\mathrm{st}}(L)$ corresponding to $\omega_1 = \omega - \Omega + 1.5\gamma$ is ten times greater than $n_{1\mathrm{st}}(L)$ corresponding to $\omega_1 = \omega + \Omega - 1.5\gamma$]. This asymmetry is due to the circumstance that in the case of a two-level atom in the field of resonant pumping the weak field is absorbed at the resonant frequency $\omega + \Omega$ and is amplified at the "three-photon" frequency $\omega - \Omega$.

§ 5. CORRELATION FUNCTIONS OF THE FIELD AMPLITUDES

The conditions under which we may encounter the correlation function of field amplitudes, which contains additional information on the scattering medium, are discussed in Ref. 27 together with the spatial and temporal properties of this function. In the present section we shall study the correlation function of the field amplitudes allowing for the parametric interaction between the modes in the case when $\Delta k = 0$, when the interaction is most effective. We shall give the final result written in the form of a sum of the parts which depend on the states of the modes on entry into the medium:

$$g(x) = g_{\mathrm{st}}(x) + g_{\mathrm{sp}}(x), \quad (5.1)$$

$$g_{\mathrm{st}}(x) = D_1e^{2\,\mathrm{Re}\,g_+x} + D_2e^{2\,\mathrm{Re}\,g_-x} - D_3e^{(g_++g_-^*)x} - D_4e^{(g_+^*+g_-)x}, \quad (5.2)$$

$$g_{\mathrm{sp}}(x) = D_{10}(e^{2\,\mathrm{Re}\,g_+x}-1) + \frac{\mathrm{Re}\,g_+}{\mathrm{Re}\,g_-}D_{20}(e^{2\,\mathrm{Re}\,g_-x}-1) - \frac{2\,\mathrm{Re}\,g_+}{g_++g_-^*}D_{30}(e^{(g_++g_-^*)x}-1) - \frac{2\,\mathrm{Re}\,g_+}{g_+^*+g_-}D_{40}(e^{(g_+^*+g_-)x}-1). \quad (5.3)$$

This result is obtained from the solutions (4.7) and the averages (4.5) and (4.6). The coefficient in front of the exponential functions in Eq. (5.2) are as follows:

$$D_{1,2} = |g_+-g_-|^{-2}\{\mu_1^*(g_{+,-} - \mathrm{Re}\,\alpha_2)n_1(0) + \mu_2^*(\dot{g}_{+,-} - \mathrm{Re}\,\alpha_1)n_2(0) + (g_{+,-} - \mathrm{Re}\,\alpha_2)(\dot{g}_{+,-} - \mathrm{Re}\,\alpha_1)g(0) + \mu_2^*\mu_1^*g^*(0)\},$$
$$D_{3,4} = |g_+-g_-|^{-2}\{\mu_1^*(g_{+,-} - \mathrm{Re}\,\alpha_2)n_1(0) + \mu_2^*(\dot{g}_{-,+} - \mathrm{Re}\,\alpha_1)n_2(0) + (g_{+,-} - \mathrm{Re}\,\alpha_2)(\dot{g}_{-,+} - \mathrm{Re}\,\alpha_1)g(0) + \mu_2^*\mu_1^*g^*(0)\}, \quad (5.4)$$

and the coefficients D_{j0} are deduced from D_j by the transformations of Eq. (4.13).

In the region of the strongest amplification, we obtain

$$g(x) = (D_1 + D_{10})\exp(2\,\mathrm{Re}\,g_+x), \quad (5.5)$$

whereas for short lengths we find from Eqs. (5.1)–(5.3) that the result is given by Eq. (4.3).

In the range $\Delta kx \gg 1$, if $\Delta k \neq 0$, the parametric coupling between the modes is unimportant; it then follows from Eq. (2.9) that there is no spontaneous component of the correlation function.

In the case of a medium of two-level atoms in the saturation range where $|V| \gg |\Delta|$, we find that, to within terms of order γ/Ω, the amplification of a mode is balanced by its absorption (see § 3.1) and the correlation function is governed entirely by the spontaneous process of emission of photons in pairs:

$$g(x) = g(0) - i\frac{\lambda^*_{\mathrm{sat}}}{\Delta k}[\exp(i\Delta kx) - 1]. \quad (5.6)$$

§ 6. SOLUTION OF EQUATIONS FOR FOURTH-ORDER MOMENTS

We shall now consider statistical characteristics of modes described by the correlation functions of the numbers of photons in the form of the normal product of the operators

$$G_{ij}(x) = \langle a_i^+(x)a_j^+(x)a_j(x)a_i(x)\rangle. \quad (6.1)$$

The fourth-order moments which follow from Eq. (2.14) satisfy the following closed system of equations:

$$\frac{\partial}{\partial x}G_{11}(x) = 4\,\mathrm{Re}\,\alpha_1G_{11}(x) + 4\,\mathrm{Re}[\mu_2R_1(x)e^{-i\Delta kx}] + 4\beta_1n_1(x), \quad (6.2)$$

$$\frac{\partial}{\partial x}G_{12}(x) = 2\,\mathrm{Re}(\alpha_1+\alpha_2)G_{12}(x) + \beta_1n_2(x) + \beta_2n_1(x) + 2\,\mathrm{Re}[(\mu_1R_1(x) + \mu_2R_2(x) + \lambda g(x))e^{-i\Delta kx}], \quad (6.3)$$

$$\frac{\partial}{\partial x}R_1(x) = \mathrm{Re}(3\alpha_1+\alpha_2)R_1(x) + \mu_2R(x)e^{-i\Delta kx} + 2\beta_1g(x) + [2\mu_2^*G_{12}(x) + \mu_1^*G_{11}(x) + 2\lambda^*n_1(x)]e^{i\Delta kx}. \quad (6.4)$$

$$\frac{\partial}{\partial x}R(x)=2\mathrm{Re}(\alpha_1+\alpha_2)R'(x)$$

$$+2[\mu_1^* R_1(x)+\mu_2^* R_2(x)+2\lambda^* g(x)]e^{i\Delta kx}. \quad (6.5)$$

The following notation is used in this system:

$$R_1(x)=\langle a_1^+(x)a_1(x)a_1(x)a_2(x)\rangle,$$

$$R_2(x)=\langle a_2^+(x)a_2(x)a_2(x)a_1(x)\rangle,$$

$$R(x)=\langle a_1^2(x)a_2^2(x)\rangle;$$

where the quantities $n_i(x)$ and $g(x)$ are the solutions of Eqs. (2.8) and (2.9), and the equations for the moments G_{22} and R_2 are obtained from Eqs. (6.2) and (6.4) when the indices are replaced.

We shall now consider how to solve the above system of equations for arbitrary boundary values of the moments which are determined by the photon statistics in the two modes on entry into the medium. We shall first consider the problem in the semiclassical approximation ignoring spontaneous processes. In this approximation we can drop the terms with the coefficients β_i and λ from the equations and this—as we can readily see—gives a homogeneous system of equations. The general quantum electrodynamic solution is a sum of linearly independent solutions of the homogeneous system, which we shall denote by $\widetilde{G}_{ij}$, $\widetilde{R}_i$, $\widetilde{R}$, and of a particular solution of the complete system. Direct substitution readily shows that the particular solution may be selected in the following form

$$G'_{ii}=2(n_i^2(x)-n_{ist}^2(x)), \quad R'=2(g^2(x)-g_{st}^2(x)),$$

$$G'_{12}(x)=n_1(x)n_2(x)+|g(x)|^2$$

$$-n_{1st}(x)n_{2st}(x)-|g_{st}(x)|^2,$$

$$R'_i(x)=2(n_i(x)g(x)-n_{ist}(x)g_{st}(x)) \quad (6.6)$$

with zero boundary solutions at $x=0$, where the components n_{ist} and g_{st} satisfy Eqs. (2.8) and (2.9) without the free terms β_i and λ. Thus the solution of the quantum problem is described by the following expressions:

$$G_{ii}(x)=\widetilde{G}_{ii}(x)+2(n_i^2(x)-n_{ist}^2(x)), \quad (6.7)$$

$$G_{12}(x)=\widetilde{G}_{12}(x)+n_1(x)n_2(x)$$

$$+|g(x)|^2-n_{1st}(x)n_{2st}(x)-|g_{st}(x)|^2, \quad (6.8)$$

$$R_i(x)=2(n_i(x)g(x)-n_{ist}(x)g_{st}(x))+\widetilde{R}_i(x), \quad (6.9)$$

$$R(x)=\widetilde{R}(x)+2(g^2(x)-g_{st}^2(x)), \quad (6.10)$$

where the quantities $n_i(x)$ and $g(x)$ for the two atomic systems under consideration are given by Eqs. (4.9)–(4.11) and (5.1)–(5.3). We can find $\widetilde{G}$ and $\widetilde{R}$ in the semiclassical approximation if we take the solutions given by the system (4.7) and multiply them directly by the operator equations from which the Langevin terms are omitted. We then have to drop the spontaneous contributions [which are allowed for fully in the particular solution (6.6)] also in the final stage of the calculations, writing down the averages of the products of the operators $a_i(0)$ and $a_i^+(0)$ in the normal form without allowance for their commutators. An analysis of the solutions (6.7) and (6.8) for different cases will be made in the next two sections.

§ 7. GAUSSIAN STATISTICS WITH CORRELATION BETWEEN MODES

We shall consider the case of spontaneous parametric fluorescence as the simplest application of the results obtained above. In the case when the vacuum initial state of two modes is $|\psi_F\rangle=|0\rangle_1|0\rangle_2$, the homogeneous system of equations has solution zero. Consequently, Eqs. (6.7) and (6.8) give the expressions

$$G_{ii}(x)=2n_{isp}^2(x), \quad (7.1)$$

$$G_{12}(x)=n_{1sp}(x)n_{2sp}(x)+|g_{sp}(x)|^2, \quad (7.2)$$

where the spontaneous components n_{isp} and g_{sp} corresponding to $\Delta k=0$ are given by Eqs. (4.11) and (5.3).

The mean square of the fluctuations of the number of photons in a mode of frequency ω_i is given by the following expression derived from Eq. (7.1):

$$\langle(\Delta n_i(x))^2\rangle=n_{isp}(x)+n_{isp}^2(x), \quad (7.3)$$

which describes the fluctuations of a single mode of chaotic light.[28] It follows from Eq. (7.2) that photons in the modes ω_1 and $2\omega-\omega_1$ are correlated by a bunching effect $(G_{12}-n_1n_2>0)$.

In the case of short propagation lengths the amplification and absorption effects are unimportant, so that the normalized correlation function $g_{12}(x)=G_{12}(x)/(n_1(x)n_2(x))$ considered in the lowest order of the expansion in x is given by

$$g_{12}(x)=1+|\lambda|^2/\beta_1\beta_2+\dots \quad . \quad (7.4)$$

We shall consider the ratio $|\lambda|^2/\beta_1\beta_2$, which describes the difference between the process of simultaneous emission of pairs of photons of frequencies ω_1 and $\omega_2=2\omega-\omega_1$ and the corresponding process in which the photons are emitted independently of one another.

Two-level atom. Using Eqs. (3.5) and (3.7) we find that in the region of the frequencies $\omega_1\approx\omega\pm\Omega$, and $\omega_2\approx\omega\mp\Omega$, the above ratio is given by

$$\frac{|\lambda|^2}{\beta_1\beta_2}\approx\left(1+\frac{\Delta^2}{2|V|^2}\right)^2\left(1+\frac{(\omega_1-\omega\pm\Omega)^2}{\Gamma^2}(\sigma_{aa}-\sigma_{bb})^2\right), \quad (7.5)$$

where for the values $|V|\gg|\Delta|$ we have $|\lambda|^2=\beta_1\beta_2$, and for $|V|\ll|\Delta|$ we obtain $|\lambda|^2/(\beta_1\beta_2)\approx\Delta^4/4|V|^4\gg1$. Numerical values of the quantities $|\lambda|^2$ and $\beta_1\beta_2$ are given in Fig. 3 for the characteristic value $2|V|=|\Delta|$. We must bear in mind that $|\lambda|^2\geqslant\beta_1\beta_2$. This inequality is a manifestation of the effect, which is nonlinear in the pump field. The quantity $|\lambda|^2$

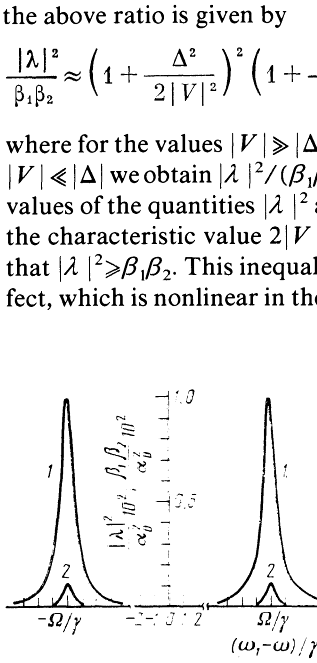

FIG. 3. Dependences on the parameter $(\omega_1-\omega)/\gamma$: 1) $|\lambda|^2$; 2($\beta_1\beta_2$; calculations made on the assumption that $2|V|=|\Delta|$ in the frequency range $\omega_1\approx\omega\pm\Omega$ employing the units $\alpha_0^2\times10^{-2}$ cm^{-2} where the parameter is $\alpha_0=4\pi N\omega_{ba}|\mathbf{d}|^2/\hbar c\gamma$; the frequencies between $\omega_1=\omega\pm(\Omega-4\gamma)$ are not considered.

near the poles is propertional to the sum of the populations $(\sigma_{aa}+\sigma_{bb})^2=1$, and the product $\beta_1\beta_2$ contains the factor

$$\sigma_{aa}\sigma_{bb}=|V|^4/(\Delta^2+2|V|^2)^2<1,$$

the smallness of which is due to filling of the state $|\Phi_b\rangle$, prior to the emission of a resonant frequency.

System with a two-photon resonance. In the region of the lines described by Eq. (3.10) we have $\omega_1\approx\nu_1$ or ν_2 and $\omega_2\approx\nu_4$ or ν_3, which are separated by the Rabi frequency, and then Eqs. (3.12) and (3.14) yield

$$(|\lambda|^2/\beta_1\beta_2)_{\omega_1\approx\nu_1,\nu_2}$$

$$\approx\frac{1}{4}\left(1+\frac{\varepsilon^2}{4|V_2|^2}\right)\left[\left(1\pm\frac{\varepsilon}{\Omega_2}\right)\gamma_{ca}+\left(1\mp\frac{\varepsilon}{\Omega_2}\right)\gamma_{bc}\right]^2\Big/\gamma_{ca}\gamma_{bc}. \quad (7.6)$$

It follows from the above formula that the measured values of Eq. (7.4) at two frequencies yield two relations for finding the two partial widths of the transitions.

The results given by Eqs. (7.1) and (7.2) represent a special case of the more general formulation of the problem when a weak field incident on a medium has Gaussian statistics, i.e., when the initial moments of higher orders can be expressed in terms of the second-order moments. A direct check shows that the system of equations (2.14) for the moments $\langle a_1+(x)^m a_2{}^+(x)^p a_2(x)^k a_1(x)^l\rangle$ for $m+k=p+l$ is closed, finite, and has the following solution

$$\langle a_1{}^+(x)^{m+l-k}a_2{}^+(x)^m a_2(x)^k a_1(x)^l\rangle$$

$$=\sum_f\frac{(m+l-k)!m!k!l!}{f!(k-f)!(l-f)!(m-k+f)!}$$

$$\times n_1(x)^{l-f}n_2(x)^{k-f}g(x)^f g^*(x)^{m-k+f}. \quad (7.7)$$

These expressions describe the evolution of a field with Gaussian statistics and a correlation between photons in two modes. They are valid for arbitrary boundary values $n_{1,2}(0)$ and $g(0)$, and also in the case of spontaneous parametric fluorescence.

§ 8. QUANTUM FLUCTUATIONS OF TWO-MODE COHERENT LIGHT

We shall now consider the contribution of spontaneous processes to parametric mixing of two modes which are described by the coherent states $|\psi_F\rangle=|z_1\rangle|z_2\rangle$, and $a_i(0)|z_i\rangle=z_i|z_i\rangle$, on entry into the medium, i.e., we shall consider the case of Poisson statistics. We shall use the method of solution of Eqs. (6.2)–(6.5) described in § 6. We can easily demonstrate that the solution of the semiclassical problem with the boundary conditions $\tilde{G}_{ii}(0)=|z_i|^4$, $\tilde{G}_{12}(0)=|z_1|^2|z_2|^2$, $\tilde{R}_i(0)=|z_i|^2z_1z_2$, $\tilde{R}(0)=z_1^2z_2^2$ is as follows:

$$\tilde{G}_{ii}(x)=n_{ist}^2(x),\quad \tilde{G}_{12}(x)=n_{1st}(x)n_{2st}(x),$$

$$\tilde{R}(x)=g_{st}^2(x),\quad \tilde{R}_i(x)=n_{ist}(x)g_{st}(x). \quad (8.1)$$

Then, using Eqs. (6.7)–(6.10), we obtain

$$G_{ii}(x)=n_i^2(x)+n_{isp}(x)(n_{isp}(x)+2n_{ist}(x)), \quad (8.2)$$

$$G_{12}(x)=n_1(x)n_2(x)+2\mathrm{Re}(g_{st}(x)g_{sp}^*(x))+|g_{sp}(x)|^2, \quad (8.3)$$

$$R_i(x)=2n_i(x)g(x)-n_{ist}(x)g_{st}(x), \quad (8.4)$$

$$R(x)=g^2(x)+2g_{st}(x)g_{sp}(x)+g_{sp}^2(x). \quad (8.5)$$

These expressions differ from the corresponding Gaussian case and they describe the evolution of statistical characteristics of two-mode coherent light due to spontaneous noise and parametric interaction.

It follows from Eqs. (8.3) that bunching or antibunching occurs between photons of different modes $(G_{12}-n_1n_2\lessgtr 0)$, for a special set of the phases of the probe fields $z_i=|z_i|\exp(i\theta_i)$ and of the phase of the intensity of the pump field $E_0=|E_0|\exp(i\theta)$. In particular, in the case of a resonant two-level medium when $\Delta k=0$ and $|V|\gg|\Delta|$, we obtain

$$G_{12}(x)-n_1(x)n_2(x)$$

$$=2\mathrm{Re}[\exp(i(\theta_1+\theta_2-2\theta))\lambda_{sat}]|z_1z_2|x+|\lambda_{sat}|^2x^2, \quad (8.6)$$

where λ_{sat} is the value of the coefficient of Eqs. (3.7) in the saturation region with a specified dependence on the pump field phase.

The dispersion of the number of photons in a mode differs from the Poisson law and is described by

$$\langle(\Delta n_i)^2\rangle=n_i(x)+n_{isp}(x)(n_{isp}(x)+2n_{ist}(x)). \quad (8.7)$$

The condition that the coherent component of the mode ω_1 is much greater than the random component can be written in the form $(G_{11}(x)/n_1^2(x))-1\ll 1$, and hence we obtain the condition $n_{1st}(x)\gg n_{1sp}(x)$. Application of this condition to the single-mode case $n_1(0)=|z_1|^2$ yields the condition for the average occupation number of a mode at the entry:

$$n_1(0)\gg l_1(x)=n_{1sp}(x)/n_{1st}(x)|_{n_1(0)=1}. \quad (8.8)$$

In the limit of short lengths the condition for the coherence of a mode is $n_1(0)\gg\beta_1x$, and in the amplification range we have $x\gg(\mathrm{Re}\,g_+)^{-1}$ and

$$n_1(0)\gg l_1=\frac{1}{2\,\mathrm{Re}\,g_+}\Big[\beta_1+\frac{|\mu_2|^2\beta_2}{|g_+-\mathrm{Re}\,\alpha_2|^2}$$

$$+2\,\mathrm{Re}\Big(\frac{\mu_2{}^*\lambda}{g_+-\mathrm{Re}\,\alpha_2}\Big)\Big]. \quad (8.9)$$

For a resonant two-level medium (see § 3.1) if $|V|\ll|\Delta|$, we obtain $l_1\approx\Delta^4/|V|^4\gg 1$ if $\omega_1\approx\omega_r$ and $l_1\gtrsim 1$, if $\omega_1\approx\omega_t$. The reason for this asymmetry is given in § 4. In the saturation region where $|V|\gg|\Delta|$ the threshold values of the occupation numbers become comparable: $l_1|_{\omega_1\approx\omega_t}\approx l_1|_{\omega_1\approx\omega_r}\approx 4|V|^2/\Delta^2\gg 1$. In the case of the frequencies in the wings of the spectral lines described by $\Omega\gg|\omega_1-\omega_{r,t}|\gg\gamma$, we generally have $l_1\gtrsim 1$.

We shall now consider briefly the feasibility of amplification of single-mode light with $n_1(0)\neq 0$, $n_2(0)=0$, and sub-Poisson statistics, when the dispersion of the number of photons in the length x

$$\langle(\Delta n_1(x))^2\rangle=n_1(x)+G_{11}(x)-n_1{}^2(x) \quad (8.10)$$

is less than in the Poisson case, i.e., when $G_{11}(x)<n_1^2(x)$.[4] We shall now turn to Eq. (6.7). In the amplification range we find from the solutions (4.7) that

$$G_{11}(x)-n_1{}^2(x)$$

$$=\frac{A_{10}{}^2}{l_1^2}[G_{11}(0)-2n_1{}^2(0)+(n_1(0)+l_1)^2]\exp(4\,\mathrm{Re}\,g_+x). \quad (8.11)$$

An analysis shows that the coefficient in front of the exponential function is positive even if $G_{11}(0) < n_1^2(0)$. This is due to the fact that $l_1 \gtrsim 1$, and it can be seen also from general considerations; in the amplification region the main asymptote of the dispersion (8.10) is equal to the difference (8.11). Hence, we can see that (8.11) is positive, and this is true also of $\langle(\Delta n_1)^2\rangle$. Therefore, in this formulation of the problem the amplification of light observing sub-Poisson photon statistics is impossible.

Continuous noise converts a sub-Poisson mode into a Poisson mode even if the propagation length is only $\sim\beta^{-1}$. It is important to note that if we ignore the two-photon spontaneous contribution [$\lambda = 0$ in Eq. (8.9)] in the wings of the spectral lines of a two-level atom where the frequencies are $\Omega \gg |\omega_1 - \omega_{r,t}| \gg \gamma$, we obtain $l_1 \sim \gamma/|\omega_1 - \omega_{r,t}| \ll 1$, which is in conflict with the above reasoning.

Both authors are grateful to B. V. Kryzhanovskiĭ, Yu. P. Malakyan, and M. L. Ter-Mikaelyan, and one of them (G.Yu.K.) is also indebted to A. P. Kazantsev and V. I. Ritus for discussions.

1) Experimental investigations of parametric scattering for the two systems discussed above were reported in Refs. 17 and 18.

2) The relationship between the quantities $\sigma_{ij}(k)$ in Ref. 21 and the density matrix of the complete system is given by the formula $[\psi_0|\sigma_{ij}(t)|\psi_0\rangle = \mathrm{Tr}(\langle\Phi_j|\rho(t)|\Phi_i\rangle)$, where the trace is summed over the variable radiation fields.

3) A mixed correlation function is allowed for in the problem of quantum fluctuations in the degenerate four-wave mixing considered in Ref. 26.

4) The sub-Poisson photon statistics was observed experimentally in resonance fluorescence of single atoms.[29]

1. S. A. Akhmanov and R. V. Khokhlov, *Nonlinear Optics*, Gordon and Breach, New York (1965).
2. N. Bloembergen, *Nonlinear Optics*, Benjamin, New York (1965).
3. R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A **24**, 411 (1981).
4. Yu. P. Malakyan, Kvantovaya Elektron. (Moscow) **12**, 1365 (1985) [Sov. J. Quantum Electron. **15**, 905 (1985)].
5. D. N. Klyshko, Photons and Nonlinear Optics [in Russian], Nauka, Moscow (1980), p. 207.
6. H. Paul, Rev.Mod. Phys. **54**, 1061 (1982).
7. D. F. Walls, Nature (London) **306**, 141 (1983).
8. P. Kumar and J. H. Shapiro, Phys. Rev. A **30**, 1568 (1984).
9. R. M. Shelby, M. D. Levenson, D. F. Walls, A. Aspect, and G. J. Milburn, Phys. Rev. A **33**, 4008 (1986).
10. Yu. M. Golubev and I. V. Sokolov, Zh. Eksp. Teor. Fiz. **87**, 408 (1984) [Sov. Phys. JETP **60**, 234 (1984)].
11. D. F. Smirnov and A. S. Troshin, Opt. Spektrosk. **59**, 3 (1985) [Opt. Spectrosc. (USSR) **59**, 1 (1985)].
12. S. A. Akhmanov, Yu. E. D'yakov, and A. S. Chirkin, Introduction to Statistical Radiophysics and Optics [in Russian], Nauka, Moscow (1981).
13. J. Peřina, Prog. Opt. **18**, 127 (1980).
14. P. A. Apanasevich and Ya.(Ja.)S. Kilin, Phys. Lett. A **62**, 83 (1977).
15. G. Yu. Kryuchkov, V. E. Mkrtchyan, M. L. Ter-Mikaelyan, and V. O. Chaltykyan, Zh. Eksp. Teor. Fiz. **88**, 30 (1985) [Sov. Phys. JETP **61**, 17 (1985)].
16. G. Yu. Kryuchkov and M. L. Ter-Mikaelyan, Dokl. Akad. Nauk SSSR **286**, 621 (1986) [Sov. Phys. Dokl. **31**, 64 (1986)].
17. D. J. Harter, P. Narum,M. G. Raymer, and R. W. Boyd, Phys. Rev. Lett. **46**, 1192 (1981).
18. J. Krasinski, D. J. Gauthier, M. S. Malcuit, and R. W. Boyd, Opt. Commun. **54**, 241 (1985).
19. Y. R. Shen, Phys. Rev. **155**, 921 (1967).
20. V. I. Ritus, Zh. Eksp. Teor. Fiz. **51**, 1544 (1966) [Sov. Phys. JETP **24**, 1041 (1967)].
21. G. Yu. Kryuchkov, Zh. Eksp. Teor. Fiz. **83**, 1992 (1982) [Sov. Phys. JETP **56**, 1153 (1982)].
22. M. Lax, Fluctuations and Coherence Phenomena [Russian translation], Mir, Moscow (1974).
23. M. L. Ter-Mikaelyan and A. O. Melikyan, Zh. Eksp. Teor. Fiz. **58**, 281 (1970) [Sov. Phys. JETP **31**, 153 (1970)].
24. V. P. Kraĭnov, Zh. Eksp. Teor. Fiz. **74**, 1616 (1978) [Sov. Phys. JETP **47**, 843 (1978)].
25. G. Yu. Kryuchkov, Dokl. Akad. Nauk Arm. SSR **29**, 209 (1984); Izv. Akad. Nauk Arm. SSR Fiz. **20**, 245 (1985).
26. M. D. Reid and D. F. Walls, Opt. Commun. **50**, 406 (1984).
27. A. P. Kazantsev, V. S. Smirnov, V. P. Sokolov, and A. N. Tumaĭkin, Zh. Eksp. Teor. Fiz. **81**, 888 (1981) [Sov. Phys. JETP **54**, 474 (1981)].
28. R. Loudon, *The Quantum Theory of Light*, Clarendon Press, Oxford (1973).
29. R. Short and L. Mandel, Phys. Rev. Lett. **51**, 384 (1983).

Translated by A. Tybulewicz