Higher-order quantum fluctuations of light in nonlinear four-wave mixing

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Quantum fluctuations nonlinear in the laser field and time correlations of photons in four-wave mixing are obtained. Stationary propagation of two radiation-field modes in a resonant medium in the presence of an optical cavity is considered with allowance for vacuum fluctuations of the radiation, intermode correlation, and relaxations. The time correlation functions of the mode amplitudes and occupation numbers are calculated for different experimental setups within the framework of quantum electrodynamics. It is shown that the laws governing the damping of optical correlations in time depend strongly on the radiation spectral range, on the mode-locking conditions, and on a parameter that characterizes resonance interaction between an atom and a laser field. Suppression of the fluctuations of the quadrature amplitudes of a radiation field in a squeezed state is attributed to growth of the intensity correlation function. The spectral-line intensities of parametric fluorescence in one-photon and two-photon excitation laser fields are also calculated.

1. INTRODUCTION

Problems involving quantum fluctuations of an electromagnetic field and photon correlations have recently become important topics in nonlinear optics. One of the reasons is the observation of novel unconventional optical quantum effects that appear in quantum fluctuations of light.[1–4] These include results on quantum statistics of light, on generation and detection of electromagnetic fields in squeezed states with suppressed quantum-fluctuation level. Another reason is the better experimental facilities available to the existing spectroscopy of intensity fluctuations,[5,6] which calls for a more accurate quantum theory of optical mixing of light.

Foremost among the optical phenomena in a nonlinear atomic medium which exhibits strong quantum properties is nondegenerate four-wave mixing, or parametric fluorescence, which is a source of an electromagnetic field in a squeezed state (see the experimental results in Refs. 7 and 8). This process is due to interaction, in a nonlinear atomic medium, between the laser field and two radiation-field modes having frequencies ω_1 and ω_2 symmetric about the laser frequency ω ($\omega_1 - \omega = \omega - \omega_2$).

The theoretical results of Refs. 9 and 10 on squeezed states in four-wave mixing deal with calculations of second-order radiation-field moments, viz., the mode occupation numbers and the mean squared amplitude fluctuations.

The present paper is devoted to a systematic and consistent study of radiation-field correlation functions of order higher than second, under conditions of four-wave interaction in a nonlinear atomic medium. We consider a stationary mode-propagation regime which is realized in the presence of an optical cavity, with account taken of the intermode-correlation quantum effects. Section 4 is devoted to parametric fluorescence in the cavity for the two typical cases of atomic media in one-photon and two-photon laser-field excitation. In Secs. 5 and 6 are calculated the photon-number correlation functions of two coupled modes $\omega_1 + \omega_2 = 2\omega$. In Sec. 7 we calculate the correlation function of the radiation-field intensities in a squeezed state with suppressed quantum amplitude fluctuations. Optical correlation effects that are nonlinearly dependent on the laser-field intensity are observed in the cavity, including photon superbunching, temporal oscillations of intensity fluctuations, and others. The dispersion about the mean of higher-order quadrature amplitudes of a two-mode field is considered in Sec. 8.

A consistent treatment of these questions is possible only within the scope of a quantum-electrodynamic theory of nonlinear wave mixing, allowing for both stimulated and spontaneous emission processes. Nondegenerate four-wave mixing has been thoroughly investigated for a two-level atomic medium in the semiclassical approximation (see, e.g., Ref. 11). Different quantum theories are proposed in Refs. 10 and 12. We use here the quantum approach proposed in our preceding paper.[13] Its salient feature is that the coefficients of the radiation-field mode-density matrix equations and the diffusion coefficients of the Langevin kinetic equations it uses are written in the quasi-energy-state (QES) representation.[1)] This makes it relatively easy to take into account effects nonlinear in the laser field (see Secs. 2 and 3) in addition to electromagnetic-field vacuum fluctuations, parametric mode interaction in a medium, and relaxations.

2. QUANTUM DESCRIPTION OF FOUR-WAVE MIXING IN THE QES REPRESENTATION

We consider four-wave interaction in an atomic medium in which the frequencies and momenta of a laser monochromatic field are transformed in accordance with the scheme $2\omega = \omega_1 + \omega_2$, $\mathbf{k}_{1l} + \mathbf{k}_{2l} = \mathbf{k}_1 + \mathbf{k}_2$.

The radiation-field modes produced in the nonlinear medium, with frequencies ω_1 and ω_2, propagate in an optical cavity along an axis that differs from that of the cavity that forms the laser-radiation wave $\mathbf{k}_{1l} = -\mathbf{k}_{2l}$ (Refs. 7–9).

The time evolution of the two radiation-field modes is described by the Langevin equations for the boson creation and annihilation operators of the mode occupation numbers in the QES representation of the "atom + monochromatic laser field" system.

We present first the semiclassical equations for the quantum-mechanical average mode amplitudes $A_i(t)$

$= \langle\psi_0|a_i(t)|\psi_0\rangle$ in an atomic medium of density N in a cavity volume v. Here

$$a_i(t)=S^+(t)a_iS(t) \tag{2.1}$$

are the annihilation operators for the photons of mode i ($i=1,2$), $S(t)=S(t,-\infty)$ is the scattering in the QES representation[15] with the interaction operator in the dipole approximation

$$W=-\mathbf{d}(t)\mathbf{E}=-i\mathbf{d}(t)\sum_i\left(\frac{2\pi\omega_i\hbar}{v}\right)^{1/2}(a_i\mathbf{e}_ie^{-i\omega_it}-a_i^+\mathbf{e}_i^*e^{i\omega_it}), \tag{2.2}$$

$\mathbf{e}_i$ are polarization vectors, and $\mathbf{d}(t)$ is the dipole-moment operator in the QES representation: $|\psi_0\rangle=|\varphi_0\rangle|\psi_F\rangle$, where $|\varphi_0\rangle$ and $|\psi_F\rangle$ are respectively the initial QES and the initial state of the modes.

Using the equation for the scattering matrix, we get

$$\frac{\partial}{\partial t}A_i(t)=\left(\frac{2\pi\omega_iv}{\hbar}\right)^{1/2}Ne^{i\omega_it}\mathbf{e}_i\langle\mathbf{D}(t)\rangle, \tag{2.3}$$

where

$$D_n(t)=S^+(t)d_n(t)S(t).$$

Using this expression in an approximation linear in the field $E(t)$:

$$D_n(t)=\frac{i}{\hbar}\int_{-\infty}^{t}dt_1\,E_m(t_1)\langle0|[D_n(t),D_m(t_1)]|0\rangle, \tag{2.4}$$

where $|0\rangle$ is the vacuum of the radiation field, we obtain for the four-wave-mixing scheme the following equations for the slowly varying amplitudes of two parametrically coupled modes $2\omega=\omega_1+\omega_2$;

$$\frac{\partial}{\partial t}A_1(t)=\alpha_1A_1(t)+\mu_2^*A_2^*(t),$$
$$\frac{\partial}{\partial t}A_2^*(t)=\alpha_2^*A_2^*(t)+\mu_1A_1(t). \tag{2.5}$$

In these equations, $\alpha_i=\alpha_{0i}-\Gamma_i$, where α_{0i} are the nonlinear susceptibilities in the presence of the laser field, Γ_i are the cavity absorption widths for the frequency ω_i and are introduced as usual phenomenologically; μ_i are the mode-coupling coefficients. The coefficients of the equations are calculated using the formulas

$$\alpha_{0i}=\frac{2\pi\omega_iN}{\hbar}e_n^*(i)e_m(i)[\Delta_{nm}^{(0)*}(-\omega_i)-\Delta_{nm}^{(0)}(\omega_i)],$$
$$\mu_1=\frac{2\pi N}{\hbar}(\omega_1\omega_2)^{1/2}e_n(2)e_m(1)[\Delta_{nm}^{(-2)*}(-\omega_1)-\Delta_{nm}^{(2)}(\omega_1)], \tag{2.6}$$

where

$$\Delta_{nm}^{(q)}(\omega_i)=\frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega}dt\int_{-\infty}^{t}dt_1\,e^{-iq\omega t}e^{i\omega_i(t-t_1)}$$
$$\times\langle\varphi_0,0|D_n(t)D_m(t_1)|0,\varphi_0\rangle. \tag{2.7}$$

The quantum Langevin equations of motion are obtained from (2.5) by the standard transition to the corresponding operators and adding noise operators $f_{1,2}$ with zero mean values $\langle f_i\rangle=0$. We limit ourselves to writing down the solutions of these equations in matrix form for the quantity $b=\begin{pmatrix}a_1\\a_2^+\end{pmatrix}$:

$$b_i(t+\tau)=\sum_kB_{ik}(\tau)b_k(t)+\sum_k\int_t^{t+\tau}dt_1\,B_{ik}(t-t_1+\tau)f_k(t_1), \tag{2.8}$$

where

$$B(t)=\frac{1}{g_+-g_-}\begin{pmatrix}(g_+-\alpha_2^*)e^{g_+t}-(g_--\alpha_2^*)e^{g_-t} & \mu_2^*(e^{g_+t}-e^{g_-t})\\ \mu_1(e^{g_+t}-e^{g_-t}) & (g_+-\alpha_1)e^{g_+t}-(g_--\alpha_1)e^{g_-t}\end{pmatrix} \tag{2.9}$$

$$2g_\pm=\alpha_1+\alpha_2^*\pm[(\alpha_2^*-\alpha_1)^2+4\mu_2^*\mu_1]^{1/2}. \tag{2.10}$$

We proceed now to calculate the field amplitude correlators containing averages over the initial state $|\psi_0\rangle$. Using the well known fluctuation regression theorem[16] based on relations of the type $\langle a_1(t+\tau)f_i(t)\rangle=0$ for $\tau>0$, we obtain from (2.8)

$$\langle a_1^+(t+\tau)a_1(t)\rangle=B_{11}^*(\tau)n_1(t)+B_{12}^*(\tau)g(t),$$
$$\langle a_2(t+\tau)a_1(t)\rangle=B_{22}^*(\tau)g(t)+B_{21}^*(\tau)n_1(t),$$
$$\langle a_1(t+\tau)a_1^+(t)\rangle=B_{11}(\tau)[n_1(t)+1]+B_{12}(\tau)g^*(t). \tag{2.11}$$

Here $n_i(t)=\langle a_i^+(t)a_i(t)\rangle$ are the mode occupation numbers and $g(t)=\langle a_1(t)a_2(t)\rangle$ is the anomalous correlator.[2)] The other correlation functions are obtained by the subscript exchange $1\rightleftarrows2$ in (2.11) with a corresponding exchange in the coefficients (2.9) and (2.10).

The contributions of the spontaneous radiation processes are contained in the quantities n_i and $g(t)$, which can be calculated with the aid of the solutions (2.8) and the equal-time mean values of the Langevin operators. We present expressions for these mean values in terms of the coefficients (2.7). Using the Einstein relations to determine the diffusion coefficients,[16] we obtain in the framework of the field-density-matrix formalism[13] for the radiation

$$\langle a_i^+(t)f_i(t)\rangle=(1-2\alpha_i/\beta_i)^{-1}\langle a_i(t)f_i^+(t)\rangle=\beta_i/2,$$
$$\langle a_1(t)f_2(t)\rangle=\left(\frac{\lambda-\mu_2}{\lambda-\mu_1}\right)^*\langle a_2(t)f_1(t)\rangle=\frac{1}{2}(\lambda-\mu_1)^* \tag{2.12}$$

(the averages for distinct t vanish). The correlation of the noise operators is a delta function with diffusion coefficients[18]

$$\langle f_i^+(t)f_i(t')\rangle=(1-2\alpha_i/\beta_i)^{-1}\langle f_i(t)f_i^+(t')\rangle=\beta_i\delta(t-t'),$$
$$\langle f_1(t)f_2(t')\rangle=\left(\frac{\lambda-\mu_1}{\lambda-\mu_2}\right)^*\langle f_2(t)f_1(t')\rangle=(\lambda-\mu_1)^*\delta(t-t'). \tag{2.13}$$

The physical meaning of the coefficients α_{0i} and μ_i in (2.6) is well known from an analysis of the semiclassical solutions of the four-wave mixing problem. The other coefficients describe one- and two-photon spontaneous emission processes[19] and are equal to

$$\beta_i = \frac{4\pi\omega_i N}{\hbar} \operatorname{Re}[e_n(i) e_m^*(i) \Delta_{nm}^{(0)}(-\omega_i)],$$

$$\lambda = \frac{2\pi N}{\hbar}(\omega_1\omega_2)^{1/2}[e_n(2)e_m(1)\Delta_{nm}^{(-2)*}(-\omega_1) + e_n(1)e_m(2)\Delta_{nm}^{(-2)*}(-\omega_2)]. \tag{2.14}$$

They characterize the quantum theory of nonlinear field mixing and were calculated for various atomic systems in our preceding paper.[13]

It must be noted that the solutions (2.8) of the quantum equations and the mean values (2.12) and (2.13) for the coupled modes are given in the general case for arbitrary atomic systems in which the four-photon process $2\omega = \omega_1 + \omega_2$ takes place. It is important that the diffusion coefficients are calculated in the proposed method microscopically and are not specified phenomenologically.

Another equivalent approach, more convenient for the investigation of a stationary regime, is to solve directly the equations for $n_i(t)$ and $g(t)$. These equations were obtained in Ref. 20; in our approach they are also easily derived by using the equation for the density matrix of two modes in the QES representation,[13] and take the form

$$\frac{\partial}{\partial t} n_{1,2}(t) = 2\operatorname{Re}\alpha_{1,2} n_{1,2}(t) + 2\operatorname{Re}[\mu_{2,1} g(t)] + \beta_{1,2},$$

$$\frac{\partial}{\partial t} g(t) = (\alpha_1+\alpha_2) g(t) + \mu_1^* n_1(t) + \mu_2^* n_2(t) + \lambda^*. \tag{2.15}$$

3. HIGHER-ORDER CORRELATION FUNCTIONS

The system of equations for the fourth-order moments are easiest to obtain with the aid of Eqs. (2.15) and the fluctuation-regression theorem mentioned earlier. According to the latter, equal-time mean values of the type $\langle a_i(t) B(t+\tau) a_j(t)\rangle$ with an arbitrary function $B(t)$ of the operators a_i and a_i^+ satisfy at $\tau > 0$ the same equations as the mean value $\langle B(t+\tau)\rangle$. As a result we get the equations

$$\frac{\partial}{\partial\tau} G_{11}(t,t+\tau) = 2\operatorname{Re}\alpha_1 G_{11}(t,t+\tau) + 2\operatorname{Re}[\mu_2 R_1(t,t+\tau)] + \beta_1 n_1(t), \tag{3.1}$$

$$\frac{\partial}{\partial\tau} G_{12}(t,t+\tau) = 2\operatorname{Re}\alpha_2 G_{12}(t,t+\tau) + 2\operatorname{Re}[\mu_1 R_1(t,t+\tau)] + \beta_2 n_1(t), \tag{3.2}$$

$$\frac{\partial}{\partial\tau} R_1(t,t+\tau) = (\alpha_1+\alpha_2) R_1(t,t+\tau) + \mu_2^* G_{12}(t,t+\tau) + \mu_1^* G_{11}(t,t+\tau) + \lambda^* n_1(t), \tag{3.3}$$

in which

$$G_{ij}(t,t+\tau) = \langle a_i^+(t) a_j^+(t+\tau) a_j(t+\tau) a_i(t)\rangle,$$

$$R_1(t,t+\tau) = \langle a_1^+(t) a_1(t+\tau) a_2(t+\tau) a_1(t)\rangle \tag{3.4}$$

for normally ordered correlators.

Consider stationary propagation of two modes excited in a cavity at time $t \gg (\operatorname{Re}\alpha_i)^{-1}$. In this regime $A_i(t)$ is zero, but the stationary solution $n_i(t) \to n_i$, $g(t) \to g$ of the quantum problem, i.e., a solution of (2.15) with $\partial n_i/\partial t = \partial g/\partial t = 0$, which is independent of the initial state of the radiation field, does exist.

Let us find the stationary solution $G_{ij}(t,t+\tau) \to G_{ij}(\tau)$, $R_i(t,t+\tau) \to R_i(\tau)$ of the set of equations (3.1)–(3.3). It is necessary first of all to find expressions for the equal-time correlators. In the stationary regime, the moments of the radiation field are independent of its initial state and are governed by vacuum processes. The equal-time correlators below the threshold for lasing in the cavity have a structure typical of Gaussian fields:

$$G_{11} = 2n_1^2, \quad G_{12} = n_1 n_2 + |g|^2, \quad R_i = 2n_i g, \tag{3.5}$$

where n_i and g are stationary solutions of Eqs. (2.15). It is easy to verify that the solution of Eqs. (3.1)–(3.3) with the boundary conditions (3.5) take the form

$$G_{11}(\tau) = n_1^2 + |\langle a_1^+(\tau) a_1\rangle|^2, \tag{3.6}$$

$$G_{12}(\tau) = n_1 n_2 + |\langle a_2(\tau) a_1\rangle|^2, \tag{3.7}$$

$$R_1(\tau) = n_1 g + \langle a_1^+ a_1(\tau)\rangle\langle a_2(\tau) a_1\rangle \tag{3.8}$$

where $\langle a_i(t+\tau) a_j(t)\rangle \to \langle a_i(\tau) a_j\rangle$ in the stationary limit.

Another quantity of interest, describing the fluctuation of the number of photons, is similarly given by

$$\langle a_i^+(\tau) a_i(\tau) a_i^+ a_i\rangle - n_i^2 = \langle a_i^+(\tau) a_i\rangle\langle a_i(\tau) a_i^+\rangle. \tag{3.9}$$

The results are valid below the lasing threshold in the cavity and include nonlinear quantum fluctuations of light and photon correlations. They are analyzed in the sections that follow.

4. SPECTRAL-LINE INTENSITIES IN PARAMETRIC FLUORESCENCE

We begin the discussion of the indicated nonlinear effect by calculating the mode occupation numbers and the anomalous correlator for a medium of two-level atoms with transition frequency ω_0 in a resonant field. We consider the case $\Omega = (\varepsilon^2 + 4|V|^2)^{1/2} \gg \gamma$, when the emission lines of this system at the resonant $\omega_r = \omega + \Omega$ and "three-photon" frequencies $\omega_t = \omega - \Omega$ are separated in the spectrum. We use the following notation: $\varepsilon = \omega_0 - \omega$ ($|\varepsilon| \ll \omega$), $V = -\mathbf{d}\mathbf{E}_0/\hbar$ is the matrix element of the interaction of a two-level atom with the pump field of amplitude $\mathbf{E}_0$, $\mathbf{d}$ is the dipole moment of the atomic transition, and γ is the spontaneous width of the atomic transition. Calculation using Eqs. (2.6), (2.7), and (2.14) by the method described in Refs. 15 and 19 yield[3)] in the approximation $\Omega \gg \gamma$:

$$\alpha_{0i} = i\frac{\pi\omega_i N}{\hbar}|\mathbf{e}(i)\mathbf{d}|^2 \rho \left\{\frac{(1-\varepsilon/\Omega)^2}{\omega_i - \omega_t + i\Gamma_\perp} - \frac{(1+\varepsilon/\Omega)^2}{\omega_i - \omega_r + i\Gamma_\perp}\right\}, \tag{4.1}$$

$$\beta_i = \frac{4\pi\omega_i N}{\hbar}|\mathbf{e}(i)\mathbf{d}|^2 \frac{|V|^2}{\Omega^2}\left\{\frac{2\Gamma_\perp|V|^2}{\Omega^2+\varepsilon^2}\left[\frac{1}{(\omega_i-\omega_t)^2+\Gamma_\perp^2} + \frac{1}{(\omega_i-\omega_r)^2+\Gamma_\perp^2}\right] + \frac{(1-\rho^2)\Gamma_\parallel}{(\omega_i-\omega)^2+\Gamma_\parallel^2}\right\}, \tag{4.2}$$

$$\mu_1 = i\frac{2\pi N}{\hbar}(\omega_1\omega_2)^{1/2}(\mathbf{e}(1)\mathbf{d})(\mathbf{e}(2)\mathbf{d}) \times \frac{V^{*2}}{\Omega^2}\rho\left\{\frac{1}{\omega_1-\omega_r+i\Gamma_\perp}-\frac{1}{\omega_1-\omega_t+i\Gamma_\perp}\right\}, \quad (4.3)$$

$$\lambda = -i\frac{2\pi N}{\hbar}(\omega_1\omega_2)^{1/2}(\mathbf{e}(1)\mathbf{d})(\mathbf{e}(2)\mathbf{d}) \times \frac{V^{*2}}{\Omega^2}\left\{\frac{1+\rho}{2}\left(\frac{1}{\omega_1-\omega_t+i\Gamma_\perp}+\frac{1}{\omega_2-\omega_t+i\Gamma_\perp}\right) + \frac{1-\rho}{2}\left(\frac{1}{\omega_1-\omega_r+i\Gamma_\perp}+\frac{1}{\omega_2-\omega_r+i\Gamma_\perp}\right) + \frac{2i(1-\rho^2)\Gamma_\parallel}{(\omega_1-\omega)^2+\Gamma_\parallel^2}\right\}. \quad (4.4)$$

The spectral widths

$$\Gamma_\perp = \frac{\gamma}{2}\left(1+\frac{2|V|^2}{\Omega^2}\right), \quad \Gamma_\parallel = \frac{\gamma}{2}\left(1+\frac{\varepsilon^2}{\Omega^2}\right) \quad (4.5)$$

in these equations agree with the known widths of the resonance fluorescence spectrum,[21] while

$$\rho = 2\Omega\varepsilon/(\varepsilon^2+\Omega^2) \quad (4.6)$$

is the difference between the stationary populations of two quasi-energy states. Note that in this scheme of parametric fluorescence in a cavity the coefficients β_i and λ of the equations contain each a spectral component of unbiased Rayleigh scattering without a contribution of coherent forward scattering.

In the case of a two-level medium the quantities n_i and g have a structure with three peaks at the frequencies $\omega_i = \omega$, $\omega \pm \Omega$. We present approximate equations obtained for their peak values from the stationary solutions of Eqs. (2.15) in general form. It can be verified with the aid of (4.1)–(4.4) that the following relations hold for each of these frequencies:

$$\operatorname{Im}\alpha_{0i}=0 \quad \mu_1=-\mu_2, \quad \beta_1\alpha_{02}-\beta_2\alpha_{01}-2\mu_2^*\lambda=0, \quad (4.7)$$

and as a result the indicated solutions can be reduced to the following simpler form:

$$n_1 = \frac{1}{2(\alpha_1+\alpha_2)}\left\{\frac{\alpha_2(\beta_1\Gamma_2-\beta_2\Gamma_1)}{\alpha_1\alpha_2+|\mu_2|^2}-\beta_1-\beta_2\right\}, \quad (4.8)$$

$$g = -\frac{\lambda^*}{\alpha_1+\alpha_2}+\frac{\mu_2^*(\beta_1\Gamma_2-\beta_2\Gamma_1)}{2(\alpha_1+\alpha_2)(\alpha_1\alpha_2+|\mu_2|^2)}. \quad (4.9)$$

(Expressions for the average number n_2 of photons of the coupled mode are obtained for (4.8) by the exchange $1\rightleftarrows 2$.) As a result, the heights of the peaks $n_\omega = n_1|_{\omega_1=\omega}$, $n_t = n_1|_{\omega_1=\omega_t}$, $n_r = n_1|_{\omega_1=\omega_r}$ and the anomalous correlator $\tilde{g} = g|_{\omega_1=\omega_t,\omega_2=\omega_r}$ are found to be

$$n_\omega = \sigma_0(1-\rho^2)|V|^2/\Gamma(\varepsilon^2+\Omega^2), \quad (4.10)$$

$$n_t = n_r = \frac{\Omega^2-\varepsilon^2}{\Omega^2+\varepsilon^2}|\tilde{g}| = \frac{(\Omega^2-\varepsilon^2)^2}{8\varepsilon^2\Omega^2+4\Gamma(3\Omega^2-\varepsilon^2)(\varepsilon^2+\Omega^2)/\sigma_0}. \quad (4.11)$$

In these expressions, $\sigma_0 = 4\pi N\omega_0|d|^2/\hbar\gamma$ is the mode absorption coefficient at the resonance frequency $\omega_1 = \omega_0$ in the absence of a laser field; we have left out the dependence on the mode polarization vectors and assumed for the cavity widths the simplifying condition $\Gamma_1 = \Gamma_2 = \Gamma$.

The heights of the side peaks of the average number of photons in the modes are thus equal, and for weak fields or large displacement from resonance $|V| \ll \varepsilon$ we obtain under the condition $\varepsilon \gg \gamma$

$$n_\omega = \frac{|V|^2\sigma_0}{\varepsilon^2\Gamma}, \quad n_t = n_p = \frac{2|V|^2}{\varepsilon^2}|\tilde{g}| = \frac{2|V|^4}{\varepsilon^4}\left(1+\frac{2\Gamma}{\sigma_0}\right)^{-1}. \quad (4.12)$$

The peak ratios for $|V| \gg \varepsilon$ and $\Gamma \lesssim \sigma_0$ are $n_\omega/n_t = n_\omega/n_r = 3$, just as in the case of resonance fluorescence without a cavity.

We describe now the application of the results to parametric fluorescence in an atomic medium under the conditions $|\omega_{ba} - 2\omega| \ll \omega_{ba}$ of two-photon resonance of atomic levels of like parity. We assume that the system excitation $\omega_a \to \omega_b$ is described by a two-photon matrix element V_2 and the decay is predominantly via a level ω_c in accordance with the scheme $|b\rangle \to |c\rangle \to |a\rangle$ by dipole-allowed one-photon transitions. It is known[22] that under the conditions $\Omega_2 = (\varepsilon_2^2 + 4|V_2|^2)^{1/2} \gg \gamma_{bc}, \gamma_{ca}$, where $\varepsilon_2 = \omega_{ba} - 2\omega$, while γ_{bc} and γ_{ca} are the spontaneous partial widths of the respective atomic transitions $|b\rangle \to |c\rangle$ and $|c\rangle \to |a\rangle$, the spectral lines of the system

$$\nu_{1,2} = \omega_{bc}-(\varepsilon_2\pm\Omega_2)/2, \quad \nu_{3,4} = \omega_{ca}-(\varepsilon_2\pm\Omega_2)/2$$

are separated in frequency: $|\nu_i - \nu_j| \gg \gamma_{bc}, \gamma_{ca}$. Using the calculated[13] coefficients $\alpha_{0i}, \mu_i, \beta_i, \lambda$ it is easy to verify that relations (4.7) hold also in this case, so that at the frequencies $\omega_1 = \nu_i$, $\omega_2 = 2\omega - \nu_i$ the number of the photons in the mode $n_{\nu_i} = n_1|_{\omega_i=\nu_i}$ and the anomalous correlator are given by Eqs. (4.8) and (4.9).

We present the final results for coupled radiation-field spectral components $\nu_1 + \nu_4 = 2\omega$ in the limit of weak pump fields under the condition $|V_2|^2|\gamma_{bc} - \gamma_{ca}|/\varepsilon_2^2\gamma_{ca} \ll 1$ and in the simple case $\Gamma_{\nu_1} = \Gamma_{\nu_4} = \Gamma$:

$$n_{\nu_1} = \frac{|V_2|^2}{\varepsilon_2^2(\sigma_{\nu_4}+2\Gamma)}\left\{\sigma_{\nu_1}+\sigma_{\nu_4}-\frac{(\sigma_{\nu_4}+\Gamma)(\sigma_{\nu_4}-\sigma_{\nu_1})}{(\sigma_{\nu_4}+\Gamma)\gamma_{ca}/\gamma_{bc}-\sigma_{\nu_1}}\right\},$$

$$n_{\nu_4} = \frac{|V_2|^2}{\varepsilon_2^2(\sigma_{\nu_4}+2\Gamma)}\left\{\sigma_{\nu_1}+\sigma_{\nu_4}+\frac{\Gamma(\sigma_{\nu_4}-\sigma_{\nu_1})}{(\sigma_{\nu_4}+\Gamma)\gamma_{ca}/\gamma_{bc}-\sigma_{\nu_1}}\right\}, \quad (4.13)$$

where

$$\sigma_{\nu_1} = 4\pi N\nu_1|\mathbf{e}_{\nu_1}^*\mathbf{d}_1|^2/\hbar\gamma_{bc}, \quad \sigma_{\nu_4} = 4\pi N\nu_4|\mathbf{e}_{\nu_4}^*\mathbf{d}_2|^2/\hbar\gamma_{ca}$$

are the absorption coefficients in adjacent atomic transitions, while $\mathbf{d}_1 = \langle\varphi_c|\mathbf{d}|\varphi_b\rangle$, $\mathbf{d}_2 = \langle\varphi_a|\mathbf{d}|\varphi_c\rangle$. In these approximations, the anomalous correlator g in the region $\omega_1 = \nu_1$, $\omega_2 = \nu_4$ is equal to

$$g = -\frac{V_2}{\varepsilon_2}\left(\sigma_{\nu_1}\sigma_{\nu_4}\frac{\gamma_{bc}}{\gamma_{ca}}\right)^{1/2} \Big/ (\sigma_{\nu_4}+2\Gamma). \quad (4.14)$$

In contrast to the case of a two-level medium, the Stark-doublet peak heights are in our case asymmetric, $n_{\nu_1} \neq n_{\nu_4}$. This follows also from the general result (4.8). In fact, the equality $n_1 = n_2$ holds if the ratio of the emission rates of photons with frequencies ω_1 and ω_2 is equal to the ratio of the cavity absorption widths at the corresponding frequencies, i.e., $\beta_1/\beta_2 = \Gamma_1/\Gamma_2$. Note that the line intensities of the second Stark double (ν_2,ν_3) are smaller in the weak-field region by an additional factor $|V_2|^2/\varepsilon_2^2$ compared with (4.13).

5. QUANTUM STATISTICS WITH ALLOWANCE FOR INTERMODE CORRELATION

Let us investigate the correlator $G_{11}(\tau)$ for a two-level medium in the region of interest $\Omega \gg \gamma$, with an aim at an experiment in which the radiation field outside the cavity is filtered and is incident on a photodetector. We assume that the spectral bandpass of the filter is $\Delta\omega \ll \Omega$. Thus, we measure the photocurrent spectrum of single-mode light which can be expressed[6] in terms of a Fourier transform of $G_{11}(\tau)$.

Using (2.9), (2.11), and (3.6) we get the expression

$$G_{11}(\tau)-n_1^2=|b_+|^2 \exp(2\,\mathrm{Re}\, g_+\tau)+|b_-|^2 \exp(2\,\mathrm{Re}\, g_-\tau) -2|b_+b_-^*| \exp[(\mathrm{Re}\, g_+ + \mathrm{Re}\, g_-)\tau] \cos(\Delta\tau-\varphi), \tag{5.1}$$

in which

$$\Delta=\mathrm{Im}[(\alpha_1-\alpha_2^*)^2+4\mu_2^*\mu_1]^{1/2},$$

$$b_\pm=(g_+-g_-)^{-1}[(g_\pm-\alpha_2^*)n_1+\mu_2^* g^*],$$

$$\varphi=\mathrm{arctg}[\mathrm{Im}(b_+b_-^*)/\mathrm{Re}\,(b_+b_-^*)].$$

We consider first the frequency range of the spectral lines. Calculations using the results of the preceding sections [Eqs. (4.1)–(4.5) and (4.11)] show that the normalized second-order correlation function is the same for the frequencies $\omega_1 = \omega_t$, $\omega_1 = \omega_r$ and is equal to

$$G_{11}(\tau)/n_1^2=1+[\exp(g_+\tau)+\exp(g_-\tau)]^2/4, \tag{5.2}$$

where

$$g_+=-\Gamma, \quad g_-=-\Gamma-\frac{\sigma_0\Omega^2\varepsilon^2}{(\varepsilon^2+6|V|^2)(\varepsilon^2+2|V|^2)}. \tag{5.3}$$

Let us comment on the result (5.2). It is accurate to terms of order γ/Ω if $\Omega \gg \gamma$, when the resonance and three-photon lines are separated in the spectrum, if the mode polarizations are equal, $\mathbf{e}_1 = \mathbf{e}_2 = \mathbf{e}$, $\mathbf{e}\|\mathbf{d}$. It is easy to verify with the aid of (4.1) and (4.3) that the assumptions made for the four-wave mixing we are considering lead to the following useful relations between the coefficients:

$$[(\alpha_{01}-\alpha_{02}^*)^2+4\mu_2^*\mu_1]^{1/2}=-(\alpha_{01}+\alpha_{02}^*) =\frac{\sigma_0\varepsilon^2\Omega^2}{(\varepsilon^2+\Omega^2)(\varepsilon^2+6|V|^2)}\,\frac{\Gamma_\perp^2(1+i(\omega_1-\omega+\Omega)/\Gamma_\perp)}{(\omega_1-\omega+\Omega)^2+\Gamma_\perp^2} \tag{5.4}$$

over a wide frequency range $\omega_1 \approx \omega_t$ (the same relations hold also in the region $\omega_1 \approx \omega_r$). As a result we obtain the simple expressions (5.3) for the coefficients (2.10) of the exponents.

It follows from (5.2) that the correlation function of the occupation numbers of one mode is affected by the damping of the fluctuations of both modes ω_t and ω_r. This is due to the parametric interaction between the modes, particularly to allowance, in the correlator, for the contribution from the spontaneous two-photon emission $2\omega \to \omega_1 + \omega_2$ of the atoms of the medium. In the absence of parametric interaction in the medium (meaning in our scheme that there is no cavity for the pump field, in which case $\mathbf{k}_{1l} + \mathbf{k}_{2l} \neq 0$, so that the synchronism condition is not met, $\mathbf{k}_{1l} + \mathbf{k}_{2l} \neq \mathbf{k}_1 + \mathbf{k}_2$), we have $\mu_i = \lambda = 0$ and, as follows directly from Eqs. (3.1)–(3.3),

$$G_{11}(\tau)/n_1^2=1+\exp\{-2(\Gamma-\mathrm{Re}\,\alpha_{01})\tau\}. \tag{5.5}$$

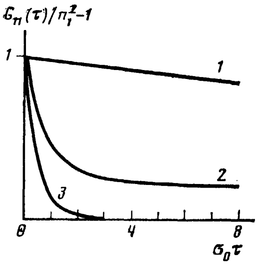

FIG. 1. Dependence of $G_{11}(\tau)/n_1^2 - 1$ on the parameter $\sigma_0\tau$ for the parameter values $\Gamma/\sigma_0 = 0.01$ and $2|V| = |\varepsilon|$. Curves *1* and *3* correspond to the absence of mode locking and are shown for the frequencies $\omega_1 = \omega - \Omega$ and $\omega_1 + \Omega$, respectively. Curve *2* describes the case of mode locking at $\omega_1 = \omega \mp \Omega$.

The fluctuation damping is determined in this case by the mode enhancement $\mathrm{Re}\,\alpha_{01} > 0$ in the region of the three-photon line and by the mode absorption $\mathrm{Re}\,\alpha_{01} \ll 0$ in the region of the resonance line, with

$$\mathrm{Re}\,\alpha_{01}|_{\omega_1=\omega_t} = -\left(\frac{\Omega-\varepsilon}{\Omega+\varepsilon}\right)^2 \mathrm{Re}\,\alpha_{01}|_{\omega_1=\omega_r} = \frac{\sigma_0}{4}\left(1-\frac{\varepsilon}{\Omega}\right)^2 \frac{\gamma\rho}{\Gamma_\perp}. \tag{5.6}$$

A graphical comparison of the correlators for the two cases of parametric interaction of the modes and in the absence of correlation is shown in Fig. 1.

For weak pump fields or large displacement from resonance we must put in (5.2) $g_+ = -\Gamma$ and $g_- = -\Gamma - \sigma_0$. In other frequency regions, the correlator (5.1) contains time-beat effects with a parameter Δ that is indicative only of a nonlinear medium and, as is important for spectroscopy problems, does not contain the cavity absorption width. In the region $\omega_1 \approx \omega_r$ or $\omega_1 \approx \omega_t$ we obtain with the aid of (5.1) an expression

$$|\Delta| = \frac{\sigma_0}{2}\,\frac{\varepsilon^2}{\varepsilon^2+2|V|^2}\,\frac{\gamma|\omega_1-\omega\mp\Omega|}{(\omega_1-\omega\mp\Omega)^2+{}^1/_4\gamma^2(1+2|V|^2/\Omega^2)^2}, \tag{5.7}$$

which vanishes in the region $|V| \gg |\varepsilon|$. The temporal oscillations in the correlator are due to parametric interaction; they are substantial on the emission spectral-line wings and vanish at $\omega = \omega_r$ or $\omega = \omega_t$ in agreement with (5.2), and also when mode locking is violated.

The temporal variation of the intensity correlation function depends also on the ratio $|V|/|\varepsilon|$ and on the parameter Γ/σ_0. For a cavity with large absorption width $\Gamma \gg \sigma_0$ the intensity-fluctuation damping is determined by the quantity

$$G_{11}(\tau)/n_1^2=1+\exp(-2\Gamma\tau). \tag{5.8}$$

We present numerical results obtained from Eqs. (5.1) and the exact stationary solutions of Eqs. (2.15) for a good cavity with small absorption width. The normalized correlator has the same dependence on the dimensionless parameter $\sigma_0\tau$ in either the frequency region $\omega_1 \approx \omega_r$ or $\omega_1 \approx \omega_t$, and is shown for two typical cases in Fig. 2. These data are evidence that the oscillation scale depends on the density of the medium via the absorption parameter σ_0 and via the detuning of the mode frequency from the side lobes of the triplet.

In the region $\omega_1 \approx \omega$ the gain and the absorption of the unshifted mode cancel each other, therefore $\alpha_{0i} = \mu_i = 0$ in accordance with (4.1) and (4.3), and $g_+ = g_- = -\Gamma$. As follows from (5.1), we obtain in this region for the normal-

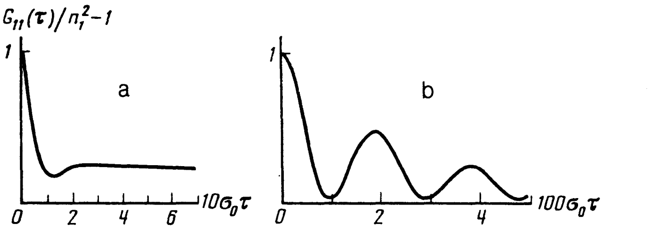

FIG. 2. Dependence of second-order normalized correlation function on the delay time for the following parameter values: a—$\Gamma/\sigma_0 = 0.001$, $2|V| = |\varepsilon|$, $\omega_t - \omega_1 = \gamma$, b—$\Gamma/\sigma_0 = 0.001$, $2|V| = |\varepsilon|$, $\omega_t - \omega_1 = 10\gamma$. The parameter σ_0 is equal to $1.3\cdot 10^6\ s^{-1}$ for the transition $(6s^2)\,^1S_0 \to (6s6p)\,^1P_1^0$ in atomic Ba (at a density $N = 10^8\ cm^{-3}$.

ized correlator expression (5.8) in which n_1 is given by Eq. (4.10).

6. NONLINEAR PHOTON SUPERBUNCHING

Let us investigate the temporal photon-number correlations between two modes, as determined by the quantity $G_{12}(\tau)$. These effects are due to parametric interaction between the modes; in its absence, in view of violation of the mode-locking condition, we obtain $G_{12}(\tau) = n_1 n_2$. In the stationary regime, Eqs. (2.9), (2.11), and (3.7) yield

$$G_{12}(\tau) - n_1 n_2 = |C_+ \exp(g_+\tau) - C_- \exp(g_-\tau)|^2,$$
$$C_\pm = (g_+ - g_-)^{-1}\{(g_\pm - \alpha_1)g^* + \mu_1 n_1\}. \qquad (6.1)$$

It is simplest to analyze this expression for a cavity with large damping, $\Gamma \gg \max\{|\alpha_{0i}|, |\mu_i|\}$. In this approximation, as follows from Eqs. (2.15) in the stationary regime, we have $n_i = \beta_i/2\Gamma$, $g = \lambda^*/2\Gamma$, and obtain for the normalized correlator the expression

$$\frac{G_{12}(\tau)}{n_1 n_2} = 1 + \frac{|\lambda|^2}{\beta_1\beta_2} e^{-2\Gamma\tau}, \qquad (6.2)$$

which is symmetric with respect to photon permutation in the two modes.

The ratio $|\lambda|^2/\beta_1\beta_2$ does not contain the atomic number density and is indicative of the difference between pairwise spontaneous emission of photons of frequency ω_1 and $2\omega - \omega_1$ and the corresponding independent emission by an atomic system. For resonance fluorescence by a two-level atom we have $|\lambda|^2 = \beta_1\beta_2$ in the frequency region $\omega_1 \approx \omega_2 \approx \omega$ and $|\lambda|^2/\beta_1\beta_2 \gg 1$ in the region $\omega_1 \neq \omega_2 \neq \omega$ for weak pump fields or large detuning from resonance [see Eqs. (4.2) and (4.4)].

Let us examine the functions

$$G_{12}(\tau)/n_1 n_2 = 1 + F(\tau) \qquad (6.3)$$

in the frequency region $\omega_1 = \omega_t$, $\omega_2 = \omega_r$ or $\omega_1 = \omega_r$, $\omega_2 = \omega_t$. They correspond to setting up a correlation experiment in which the statistics of delayed counts of photons having frequencies $\omega \pm \Omega$ separated by spectral filters are measured. The quantity $F_2(\tau) = F(\tau)|_{\omega_1 = \omega_r, \omega_2 = \omega_t}$ determines the probability of detecting photons of frequency ω_r at a time τ after recording photons of frequency ω_t. Similarly, the probability of detecting photons of frequency ω_t and ω_r in reverse chronological order is determined by the quantity $F_2(\tau) = F(\tau)|_{\omega_1 = \omega_r, \omega_2 = \omega_t}$. Calculations similar to those leading to (5.2), with account taken of the ratio $|\tilde{g}|/n_1 = (\Omega^2 + \varepsilon^2)/4|V|^2$ in (4.11), yield

$$F_{1,2}(\tau) = \frac{1}{16\varepsilon^2\Omega^2}\left\{\left[4|V|^2 - (\Omega+\varepsilon)^2\frac{\varepsilon^2 + 2|V|^2}{2|V|^2}\right]\exp(g_{-,+}\tau) - \left[4|V|^2 - (\Omega-\varepsilon)^2\frac{\varepsilon^2+2|V|^2}{2|V|^2}\right]\exp(g_{+,-}\tau)\right\}^2, \qquad (6.4)$$

where $g_{+,-}$ are given by Eq. (5.3). For $|V| \ll \varepsilon$ we get

$$F_2(\tau) = \frac{\varepsilon^4}{4|V|^4}\exp(-2\Gamma\tau),$$
$$F_1(\tau) = \frac{\varepsilon^4}{4|V|^4}\exp[-2(\Gamma+\sigma_0)\tau]. \qquad (6.5)$$

The results of calculations using (6.4) for the case $2|V| = \varepsilon$ and for the two values $\Gamma/\sigma_0 = 0.01$ and 1 are shown in Fig. 3. For clarity, the $F_1(\tau)$ and $F_2(\tau)$ curves are drawn on opposite sides of the ordinate axis.

It follows from the foregoing that at zero delay the probability of recording two photons is a maximum, with

$$\frac{G_{12}(0)}{n_1 n_2} = 1 + \left(1 + \frac{\varepsilon^2}{2|V|^2}\right)^2. \qquad (6.6)$$

The correlation between photons from different modes is therefore of the bunching or superbunching type if $|\varepsilon| \gg |V|$.

For $\tau \neq 0$, the time sequence in which the photons of the Stark triplet side peaks are recorded is not symmetric. Thus, the number of events in which photons ω_r of the resonance line are recorded prior to the photons ω_t of the "three-photon" line on leaving the cavity exceeds the number of events detected in the reverse sequence. The asymmetry vanishes in the case of a small absorption coefficient $\sigma_0 \ll \Gamma$, since the spectral component ω_r is absorbed while ω_t is amplified in a two-level resonance medium. It is important to note that the situation is different for resonance fluorescence by an indi-

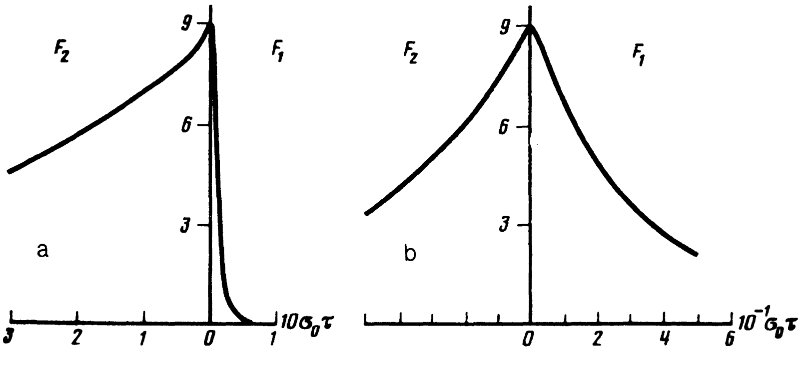

FIG. 3. Time-dependence asymmetry of normalized correlation between photons of two coupled modes at $2|V| = |\varepsilon|$ for the cases $\Gamma/\sigma_0 = 0.01$ (a) and $\Gamma/\sigma_0 = 1$ (b).

vidual atom.[4)] Namely, the number of pairs in which photons of the "three-photon" line are emitted prior to photons of the resonance line exceeds the number of pairs for the reverse emission sequence.

7. LIGHT-INTENSITY CORRELATION IN A SQUEEZED STATE

We consider now the correlation function of a two-mode field emerging from the cavity without the additional mode filtering considered in the problems of Secs. 5 and 6. If the interval between two frequencies ω_1 and ω_2 in the cavity is much less than the laser frequency ω, the field intensity [see Eq. (2.2)] can be written in the form

$$E(t)=E^{(+)}(t)+E^{(-)}(t)=E_1(t)+E_2(t),$$

$$E^{(+)}(t)=cA(t)e^{i\omega t},\quad E^{(-)}(t)=[E^{(+)}(t)]^+, \tag{7.1}$$

where

$$A(t)=2^{-1/2}i[a_1(t)e^{-i\delta t}+a_2(t)e^{i\delta t}],$$

$$c=(4\pi\hbar\omega/v)^{1/2},\quad \delta=\omega_1-\omega=\omega-\omega_2,\quad |\delta|\ll\omega. \tag{7.2}$$

The intensity of the resultant field is, in the stationary regime,

$$I=\langle E^{(-)}E^{(+)}\rangle=c^2(n_1+n_2)/2. \tag{7.3}$$

Our aim is to calculate the correlator

$$G(\tau)=\langle E^{(-)}(t)E^{(-)}(t+\tau)E^{(+)}(t+\tau)E^{(+)}(t)\rangle|_{t\to\infty}. \tag{7.4}$$

Leaving out intermediate steps, we present the following intermediate result:

$$4G(\tau)/c^4=G_{11}(\tau)+G_{22}(\tau)+G_{12}(\tau)+G_{21}(\tau)$$
$$+2\,\mathrm{Re}\{(\langle a_2^+(\tau)a_2\rangle\langle a_1^+(\tau)a_1\rangle^*$$
$$+\langle a_1(\tau)a_2\rangle\langle a_2(\tau)a_1\rangle^*)e^{i(\omega_2-\omega_1)\tau}\}, \tag{7.5}$$

where the $G_{i,j}$ are given by Eqs. (3.6) and (3.7). This expression was obtained by using, for arbitrary values of τ, the identities

$$\langle a_i a_i(\tau)\rangle=\langle a_2^+a_1(\tau)\rangle=0 \tag{7.6}$$

which can be easily verified using the solutions (2.8) and the equation cited in Sec. 2 for the density matrix.[5)] Note also that expression (7.5) corresponds to the following factorized form of the correlator:

$$G(\tau)=I^2+|\langle E^{(-)}(\tau)E^{(+)}\rangle|^2+|\langle E^{(+)}(\tau)E^{(+)}\rangle|^2. \tag{7.7}$$

There is no correlation for large delay-time intervals: $G(\tau)|_{\tau\to\infty}\to I^2$, and at zero time we obtain from (7.5)

$$G(0)/I^2=2+4|g|^2/(n_1+n_2)^2. \tag{7.8}$$

If the mode coupling is made to vanish by eliminating the mode locking, Eq. (7.8) describes the statistics of random light: $G(0)=2I^2$.

It was shown recently[25] that under certain conditions the field (7.1) is in a squeezed state. In this case the variance of any one of the quadrature components $A_1=A+A^+$, $A_2=-i(A-A^+)$:

$$\langle(\Delta A_{1,2})^2\rangle=1+n_1+n_2\mp2\,\mathrm{Re}\,g, \tag{7.9}$$

is smaller than in the coherent state, i.e., $\langle(\Delta A_1)^2\rangle<1$ or $\langle(\Delta A_2)^2\rangle<1$ for $\langle(\Delta A_1)^2\rangle\langle(\Delta A_2)^2\rangle\geqslant1$ ($\Delta A_i=A_i-\langle A_i\rangle$). The maximum suppression of the fluctuations of one of the quadrature amplitudes is realized when the condition $2|g|>n_1+n_2$ is met, if the phase of the normal correlator is specially chosen. For such a radiation field we have a ratio $G(0)/I^2>3$, i.e., substantially different from the ratio that characterizes random light. Recall that $G(0)=I^2$ for a coherent field.

We present the final result for the correlator in the spectral-line regions $\omega_1=\omega_t,\omega_2=\omega_r$, for four-wave mixing in a two-level medium. With the aid of Eqs. (2.9), (2.11), (4.11), (5.4), and (7.5) we obtain the expression

$$\frac{G(\tau)}{I^2}=1+\frac{1}{8}\left\{\left[1+\left(1+\frac{\varepsilon^2}{2|V|^2}\right)^2\right][1+\cos(2\Omega\tau)]\right.$$
$$\left.\times[\exp(g_+\tau)+\exp(g_-\tau)]^2\right\}+\frac{\Omega^2\varepsilon^2}{32|V|^4}[1-\cos(2\Omega\tau)]$$
$$\times[\exp(g_+\tau)-\exp(g_-\tau)]^2, \tag{7.10}$$

in which the coefficients $g_{+,-}$ are given by (5.3). Thus, the two-mode field intensity correlation attenuates with increase of τ and contains oscillations at the difference frequency $\omega_2-\omega_1=2\Omega$. For $\tau=0$ the intensity correlation has a bunching behavior

$$\frac{G(0)}{I^2}=3+\frac{\varepsilon^2}{|V|^2}\left(1+\frac{\varepsilon^2}{4|V|^2}\right), \tag{7.11}$$

and increases at small values of the parameter $|V|/|\varepsilon|$, which determines the resonance interaction of the atom with the pump field. For mutually coupled modes in another spectral region, $|\omega_1-\omega_t|\gtrsim\gamma$, $|\omega_2-\omega_r|\geqslant\gamma$, the ratio $G(0)/I^2$ depends also on the parameter Γ/σ_0 and has a rather complicated form. Analysis shows that in this region the dependence of the normalized correlator (7.8) on the parameter $|V|/|\varepsilon|$ differs little from (7.11) for the case of cavities with small damping $\Gamma_0\ll\sigma_0$. A substantial difference appears for four-wave mixing in a poor cavity, $\Gamma\gg\sigma_0$. In this case the radiation spectrum is similar to the fluorescence spectrum in free space [see the results preceding Eq. (6.2)]. The correlation function (7.8) is determined by the ratio $4|\lambda|^2/(\beta_1+\beta_2)^2$, which increases, as follows from (4.2) and (4.4), in proportion to $(\omega-\omega_{t,r})^2/\gamma^2$ on the spectral-line wings.

8. DISPERSION OF HIGHER-ORDER QUANTUM FLUCTUATIONS

We turn now to an analysis of the moments of the higher-order quantum fluctuations of the quadrature amplitudes of two-mode field (7.1)

$$E(t)=c[A_1(t)\cos\omega t+A_2(t)\sin\omega t]. \tag{8.1}$$

To calculate the higher moments we use Eqs. (3.5) and the relation $\langle a_1a_2a_2a_1\rangle=2g^2$, which express the fourth-order moments in terms of second-order moments. As a result we obtain the following simple result:

$$\langle(\Delta A_{1,2})^4\rangle=3\langle(A_{1,2})^2\rangle^2, \tag{8.2}$$

where the second-order dispersion is given by Eq. (7.9).

The condition for realizing squeezed states in fourth order can be obtained by the method of Ref. 26. As applied to four-wave mixing, it takes the form $\langle(\Delta A_1)^4\rangle<3$. The condition for squeezed states is met in fourth order together with the analogous condition $\langle(\Delta A_1)^2\rangle<1$ in second order.

Let us discuss briefly the question of quantum fluctuations of the square of the amplitude (7.2)

$$A(t)^2=d_1(t)+id_2(t), \quad d_1^+=d_1, \quad d_2^+=d_2. \tag{8.3}$$

Direct calculation of the dispersion with allowance for the equations that relate the fourth- and second-order moments yields

$$\langle(\Delta d_{1,2})^2\rangle={}^1/_2(1+n_1+n_2)+Q_{1,2},$$
$$Q_{1,2}={}^1/_4(n_1+n_2)^2\pm4[(\mathrm{Re}\, g)^2-(\mathrm{Im}\, g)^2]. \tag{8.4}$$

Using the commutator $[A,A^+]=1$ and Eq. (7.6) for $\tau=0$, we easily derive uncertainty relations for the dispersion of the amplitudes d_1 and d_2:

$$\langle(\Delta d_1)^2\rangle\langle(\Delta d_2)^2\rangle\geqslant{}^1/_4(1+n_1+n_2)^2. \tag{8.5}$$

This leads to a condition for squeezed states of the squared amplitude: either $Q_1<0$ or $Q_2<0$.

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[1)] Quasi-energies and QES of a composite "atom + monochromatic field" system were introduced in Ref. 14.

[2)] The anomalous correlator of a radiation field in a scattering medium was introduced in Ref. 17.

[3)] The coefficients (4.1)–(4.4) are more general than given in our preceding paper[13] and contain, in particular, the incoherent Rayleigh scattering.

[4)] A theory and experimental results on spectral–temporal correlations in resonance fluorescence are given in Refs. 23 and 24, respectively.

[5)] One way of verifying this is the following. Equations of type (2.15) for the mean values $\langle a_i^2(t)\rangle$, $\langle a_1^+(t)a_2(t)\rangle$ do not contain free terms, and have consequently zero stationary solutions.

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