

# Generation of squeezed states of an electromagnetic field in an interaction of electromagnetic radiation with optically oriented atoms

D. V. Kupriyanov and I. M. Sokolov

Leningrad Polytechnical Institute

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An analysis is made of the formation of squeezed states of an electromagnetic field as the result of a nonlinear interaction of classical electromagnetic radiation of frequency  $\omega$  with a system of optically oriented atoms in a magnetic field. A polarization of the atoms may, under certain conditions, intensify the parametric generation and thus the squeezing of the electromagnetic field near the frequencies  $\omega \pm \Omega_0$  and  $\omega \pm 2\Omega_0$ , where  $\Omega_0$  is the Zeeman splitting frequency of the atomic ground state. A high degree of squeezing is achieved in the scattered light when the intensity of the incident radiation is well below the saturation intensity in the wing of the excited atomic transition.

## 1. INTRODUCTION

The problem of the generation of squeezed states of electromagnetic fields is presently the subject of active research because of the promising outlook for the development of low-noise sources of optical radiation.<sup>1,2</sup> In particular, nonlinear scattering of classical pump radiation by a macroscopic system of atoms can be used to generate squeezed states.<sup>3,4</sup> This possibility was realized in the experiments of Ref. 5, in a layout of in-cavity nondegenerate four-wave mixing. The working medium was Na vapor. Some subsequent theoretical analyses<sup>6–8</sup> of this experiment considered the interaction of classical pump radiation with a system of “immobile” two-level atoms, in complete accordance with the experimental conditions of Ref. 5. An important conclusion reached in those papers is that an effective squeezing can be achieved in experiments of this type if the pump intensity is comparable to the saturation value in the wing of the atomic transition. A possible degeneracy of the excited state of originally equilibrium atoms and a polarization of the pump were taken into account in Ref. 9, but there was no fundamental change in the conclusions.

On the other hand, it is known from research in the spectroscopy of intensity fluctuations that the nature of the fluctuations in transmitted radiation which has interacted with a system of atoms is strongly influenced by the Zeeman splitting of the ground state of these atoms.<sup>10,11</sup> The fluctuation spectrum of the photocurrent becomes even richer when one considers the atoms which populate Zeeman sublevels in a nonuniform fashion.<sup>12,13</sup> The deviation from equilibrium which is required can be experimentally produced by a weak, incoherent, external optical-pump source, if the ground state is a spinor state and if its polarization is disrupted slightly by relaxation processes (the optical-orientation method). It was found in Ref. 13 that under certain conditions manifestations of sub-Poisson statistics may appear in the spectrum of photocurrent fluctuations when the radiation intensity is well below the saturation intensity.

In the present paper it is shown that the sub-Poisson effect in the case at hand stems from a specific manifestation of the squeezing of radiation which has been scattered in a nonlinear fashion by a system of nonequilibrium moving

atoms. General transport equations are derived to describe the evolution of the normal and anomalous correlation functions in a gas of oriented atoms. Conditions under which a high degree of squeezing can be achieved are determined.

## 2. PHOTOCURRENT FLUCTUATION SPECTRUM IN THE HETERODYNING OF SCATTERED RADIATION

Let us consider the relationship between the observed spectrum of fluctuations in the photocurrent of radiation scattered by a nonlinear medium and the correlation characteristics calculated for this radiation at a subsequent time. In the quantum theory of photodetection, the informative part of the fluctuation spectrum is determined by the fourth-order correlation function of the electromagnetic field.<sup>14</sup> In the case of the heterodyning of radiation incident on the surface of a photocathode by a group of modes which are in a classical coherent state and whose intensity is high with respect to the intensity of the light scattered by the medium, the photocurrent fluctuation spectrum can be written in the form

$$\begin{aligned} \langle |\delta i_a|^2 \rangle = & \zeta I S_0 + \zeta^2 I S_0 \\ & \times \int \frac{d^3 \kappa}{(2\pi)^3} f(\kappa_{\perp}) \frac{c}{2\pi \hbar \omega} \{ e^{-2i\theta_{\kappa}} (\mathbf{e}^*)^{\mu_1} (\mathbf{e}^*)^{\mu_2} \Phi_{\mu_1 \mu_2}^{(-)}(\kappa, \Omega) \\ & + e^{2i\theta_{\kappa}} e^{\mu_1} e^{\mu_2} \Phi_{\mu_1 \mu_2}^{(++)}(\kappa, \Omega) \\ & + e^{\mu_1} (\mathbf{e}^*)^{\mu_2} \Phi_{\mu_1 \mu_2}^{(+)}(\kappa, \Omega) + (\mathbf{e}^*)^{\mu_1} e^{\mu_2} \Phi_{\mu_1 \mu_2}^{(-)}(\kappa, \Omega) \}. \quad (1) \end{aligned}$$

The first term determines the level of the shot noise:  $I$  is the photon flux density in the beat-frequency wave,  $S_0$  is the area of the illuminated surface of the photocathode,  $\zeta$  is the quantum yield, and  $e = 1$  is the electron charge. The second term corresponds to the contribution to the fourth-order correlation function of the electromagnetic field from beats of the radiation scattered by the nonlinear medium with the beat-frequency wave. It is assumed that the beat-frequency radiation is represented by a set of plane waves with a negative-frequency component

$$\tilde{\mathcal{E}}_h^{(-)}(\mathbf{r}, t) = \varepsilon \int \frac{d^2 \boldsymbol{\kappa}_\perp}{(2\pi)^2} \mathcal{E}_{\boldsymbol{\kappa}_\perp} e^{i\boldsymbol{\kappa}_\perp \cdot \mathbf{r} + i\varphi(\mathbf{r}, t)}, \quad \varphi(\mathbf{r}, t) = \mathbf{k}\mathbf{r} - \omega t,$$

$$\mathcal{E}_{\boldsymbol{\kappa}_\perp} = |\mathcal{E}_{\boldsymbol{\kappa}_\perp}| e^{i\theta}, \quad (2)$$

$$|\mathcal{E}_{\boldsymbol{\kappa}_\perp}|^2 = \frac{c}{2\pi\hbar\omega} I f(\boldsymbol{\kappa}_\perp), \quad \int \frac{d^2 \boldsymbol{\kappa}_\perp}{(2\pi)^2} f(\boldsymbol{\kappa}_\perp) = 1.$$

The frequency  $\omega$  and the average wave vector  $\mathbf{k}$  coincide with the corresponding properties of the radiation incident on the medium;  $\varepsilon$  is the polarization vector of the beat-frequency wave.

The functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega)$  ( $\sigma_1, \sigma_2 = \pm$ ) are the space-time Fourier components of the normal ( $\sigma_1 = \pm, \sigma_2 = \mp$ ) and anomalous ( $\sigma_1 = \pm, \sigma_2 = \pm$ ) correlation functions of the Heisenberg operators of the scattered-radiation field. In the quasimonochromatic case, these functions can be expressed in terms of the photon Green's functions of the Keldysh diagram technique<sup>1)</sup> (Refs. 15 and 16):

$$\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega) = -\sigma_1\sigma_2 \int_{-\infty}^{\infty} d\tau \int d^3\rho e^{i\Omega\tau - i\boldsymbol{\kappa}\cdot\rho} \times \frac{\omega^2}{c^2} \exp[i\sigma_1\varphi(\mathbf{r}_1, t_1) + i\sigma_2\varphi(\mathbf{r}_2, t_2)] iD_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2), \quad (3)$$

where

$$\tau = t_1 - t_2, \quad \boldsymbol{\rho} = \mathbf{r}_1 - \mathbf{r}_2,$$

$$D_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = -i \langle T_{\sigma_1\sigma_2} A_{\mu_1}^{(\sigma_1)}(\mathbf{r}_1, t_1) A_{\mu_2}^{(\sigma_2)}(\mathbf{r}_2, t_2) \rangle,$$

and  $A_\mu^\pm(\mathbf{r}, t)$  are the positive- and negative-frequency components of the vector potential ( $\propto e^{\pm i\omega t}$ ). The operator  $T_{\sigma_1\sigma_2}$  is a chronological ordering operator in the case  $\sigma_1 = \sigma_2 = -$ ; an antichronological ordering operator in the case  $\sigma_1 = \sigma_2 = +$ ; an identity operator in the case  $\sigma_1 = +, \sigma_2 = -$ ; and an interchange operator in the case  $\sigma_1 = -, \sigma_2 = +$ . In (1) and below, we are using a cyclic basis to describe the polarization states and the covariant (contravariant) form for writing the tensor indices  $\mu_1, \mu_2$ .

Relation (3) is rigorous outside the scattering medium, in spatial regions where the field is free. The presence of an ordering in the anomalous correlation functions is unimportant, since the operators  $A_\mu^{(\pm)}$  of identical frequency commute. If we consider the steady-state case, however, and if the spatial scale of the correlations of interest is small in comparison with the length scale of the field variations in the medium (actually, in comparison with the distance over which temporal dispersion effects are manifested), relation (3) also holds approximately inside the scattering layer. The functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega)$  acquire a slow variation along the propagation direction of the pump radiation (along the coordinate  $z$ ) in this case:

$$\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega) \rightarrow \Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega; z). \quad (3')$$

Evolution equations describing the evolution along the "slow" variable  $z$  can be found for correlation functions (3), formed as a result of a nonlinear quaresonant scattering of classical laser radiation propagating through a gaseous medium. These equations, which are transport equations, describe the changes in the correlation functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega; z)$  over scales on the order of the dispersion length and the length scales of the nonlinear parametric and Raman scattering. These scales are assumed to be large in comparison with the beam radius  $a_0$ , which corresponds approximately to the spatial correlation length (more on this below).

In the following section of this paper we derive transport equations for the correlation functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega; z)$  in the case in which classical radiation (a pump in a parametric process) which is quaresonant with the atomic transition  $j \rightarrow j'$  ( $j, j'$  are the complete electron moments of the ground and excited states) is propagating through a medium. In contrast with the equations for four-wave mixing in a gas of immobile two-level atoms,<sup>6-8</sup> which are the equations usually discussed, we are interested here in the effect of space-time correlations and in the interaction of the pump wave with multilevel atoms which are oriented beforehand, and in which the sublevels of the ground state are populated in a nonuniform fashion.

### 3. TRANSPORT EQUATIONS FOR THE CORRELATION FUNCTIONS OF THE RADIATION

Let us assume that a cell holding a vapor of the working atoms is irradiated by monochromatic electromagnetic radiation of frequency  $\omega$  which is quaresonant with the electronic transition  $j \rightarrow j'$  of these atoms. The radiation is assumed to be propagating along the  $z$  direction. The cell is in an external magnetic field  $\mathbf{H}_0$ , which is directed perpendicular to the  $z$  axis (Fig. 1). The frequency of the Zeeman splitting of the ground state,  $\Omega_0$ , is small in comparison with the frequency detuning  $\omega_0 - \omega$ , where  $\omega_0$  is the frequency of the unperturbed atomic transition. The atoms of the medium are oriented by the incoherent external optical pump along the direction  $\mathbf{H}_0$ . In the coordinate system with  $z'$  axis running parallel to  $\mathbf{H}_0$ , they have nonzero polarization moments  $\rho_j^{\kappa 0}(z)$  ( $\kappa \geq 0$ ) given by

$$\rho_j^{\kappa 0}(z) = \sum_n (-1)^{j-n} \Pi_n \begin{pmatrix} j & \kappa & j \\ -n & 0 & n \end{pmatrix} \rho_{jn; jn}(z), \quad (4)$$

where  $\rho_{jn; jn}(z)$  is the density matrix of the ground state at the point  $z$ , and  $n$  is the projection of the magnetic moment onto the  $z'$  direction. We are using the notation

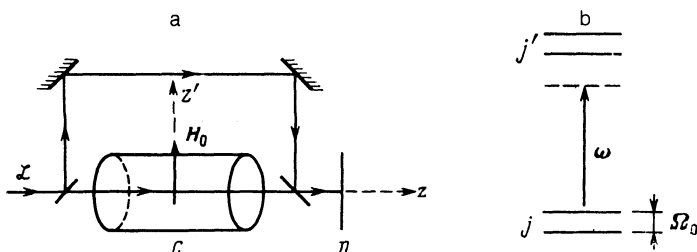
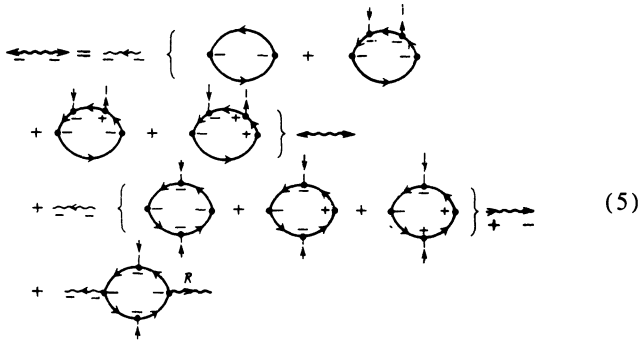


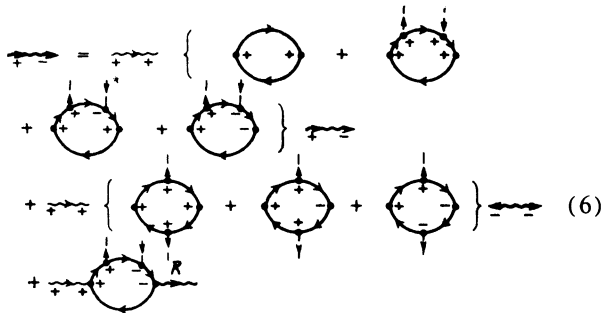
FIG. 1. a: Layout of the experiment to observe the fluctuation spectrum of the forward-scattered radiation.  $C$ —Gas-filled cell;  $L$ —laser beam (part of the radiation is tapped by a system of mirrors and used as the beat-frequency wave);  $\mathbf{H}_0$ —external magnetic field;  $D$ —photodetector. b: Diagram of the working levels for the case of the optical transition  $j = 1/2 \rightarrow j' = 1/2$ .  $\omega$ —Frequency of the incident laser radiation;  $\Omega_0$ —frequency of the Zeeman splitting of the ground state.

$\Pi_{XY\dots} = [(2X+1)(2Y+1)\dots]^{1/2}$  from Ref. 17. The length scale of the variation in the polarization moments along the  $z$  direction is much greater than the transverse beam dimension  $a_0$ . For simplicity we assume that the medium is optically isotropic and that the presence of nonzero polarization moments with  $\kappa > 0$  does not alter the polarization of the incident radiation. Correspondingly, there is no alignment in this experimental geometry:  $\rho_j^{20} = 0$ .

For the Green's functions  $D_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$  in (3) we have the following diagram equations, in which we are incorporating the assumptions made above:



for the anomalous Green's function  $D_{\mu_1\mu_2}^{(- -)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$  and



for the normal Green's function  $D_{\mu_1\mu_2}^{(+ -)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2)$ . The wavy light lines and the wavy heavy lines in the diagrams represent, respectively, the free photon Green's functions and the photon Green's functions "dressed" with the interaction with the medium. The dashed lines represent the classical field of the incident pump radiation in the medium.<sup>2)</sup> The arrows going into and coming out of a vertex on the photon Green's functions and the pump lines correspond to the negative- and positive-frequency components of the field, respectively. The letter  $R$  represents the retarded photon Green's function of linear electrodynamics. The internal lines correspond to the Green's functions of free atoms in the ground state (it is assumed that the atoms cross the pump beam without undergoing collisions) and to the Green's functions of the excited state which are dressed with the radiative interaction with the vacuum heat reservoir. Only those terms of the diagram expansions which do not vanish as a result of the operation of the wave operator in terms of the first argument (on the left) have been retained in Eqs. (5) and (6). We should add to Eqs. (5) and (6) the symmetry relations which determine the two other Green's functions:

$$D_{\mu_1\mu_2}^{(++)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = (-1)^{\mu_1+\mu_2+1} D_{-\mu_2-\mu_1}^{(--)}(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1),$$

$$D_{\mu_1\mu_2}^{(-+)}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) = D_{\mu_2\mu_1}^{(+ -)}(\mathbf{r}_2, t_2; \mathbf{r}_1, t_1). \quad (7)$$

Here are the basic approximations which we used in deriving Eqs. (5) and (6). Under the assumption that the length scale of the correlations of interest is small in comparison with that of the interaction of the field with a medium, and since the field operators of identical frequency commute at  $\mathbf{r}_1 = \mathbf{r}_2$ , we can ignore the ordering in the determination of the anomalous Green's functions, and we can assume

$$\overleftarrow{\text{---}} \approx \overrightarrow{\text{---}}, \quad \overleftarrow{\text{---}} \approx \overrightarrow{\text{---}}. \quad (8)$$

The polarization operators in diagram equations (5) and (6) were calculated with allowance for the first nonvanishing nonlinear corrections for the pump field. Accordingly, the pump field is assumed to be weak enough so that we can discard the contributions of higher orders in a perturbation theory. It follows directly from the calculation of the diagrams that the following condition must hold:

$$J\sigma_0\bar{\tau}\gamma|\omega_0-\omega|^{-1} \ll 1, \quad (9)$$

where  $J$  is the photon flux density in the pump wave,  $\sigma_0$  is the cross section for the absorption of the resonant photons in the absence of a Doppler broadening,  $\gamma$  is the natural width of the excited level,  $\bar{\tau} = a_0/\bar{v}$  is the mean transit time of an atom through the beam (this time is a rough measure of the correlation time scale in the interaction of the pump radiation with the atoms), and  $\bar{v}$  is the thermal velocity. Condition (9) means that the pump radiation cannot, strictly speaking, be thought of as a monochromatic plane wave with a given wave vector  $\mathbf{k}$  in this problem, since in this case we have  $\bar{\tau} \rightarrow \infty$ . The requirement that the pump be monochromatic can be reconciled with condition (9) if one examines correlations over distances  $|\mathbf{r}_1 - \mathbf{r}_2| \ll a_0$ .

What is the meaning of the other terms in diagram equations (5) and (6)? Terms which have been grouped in the first sets of curly brackets in these equations constitute an expansion of the retarded polarization operator, (5), and the advanced polarized operator, (6); low-lying nonlinear corrections for the pump field are being taken into account. The second terms in these equations correspond to that mutual coupling of the normal and anomalous correlation functions which results from the nonlinear parametric scattering of the pump wave in the medium. The free terms in Eqs. (5) and (6) describe correlation-function sources which are distributed over the volume: a quasis resonant Raman scattering for the normal correlation functions and a parametric scattering for the anomalous correlation functions.

Equations (5) and (6) ignore the influence of depletion and nonlinear effects on the evolution of the pump in the medium. Estimates show that the corresponding corrections to the retarded polarization operator for the pump field are small in comparison with the polarization operators of Eqs. (5) and (6), which shape the correlation functions near the resonant Zeeman frequencies ( $\Omega \sim \Omega_0, 2\Omega_0$ ) in the ratio  $\gamma/|\omega_0 - \omega|$ . In the group of terms in Eqs. (5) and (6) corresponding to the expansions of the retarded and advanced polarization operators for the photons of the conjugate parametric-scattering modes, we are also ignoring several correc-

tions which are correspondingly small. In a study of the spectrum of correlations of the electromagnetic field near a zero frequency ( $\Omega \sim 0$ ), all these corrections are comparable in magnitude to the polarization operators of Eqs. (5) and (6), and these approximations break down. Equations (5) and (6) are thus applicable for studying the spectrum of correlations only near the Zeeman frequencies  $\Omega \sim \Omega_0, 2\Omega_0$ .

To diagram equations (5) and (6) we should add corresponding equations with polarization operators which act on the right ends of the diagrams. Equations describing the slow evolution of the correlation functions along the variable  $z$  are found by the standard method, which is analogous to the method used to derive a kinetic equation in Ref. 16. As a result, using (3) and (3'), we find the following system of consistent transport equations for the correlation functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega; z)$ :

$$\begin{aligned} & \frac{\partial}{\partial z} \Phi_{\mu_1\mu_2}^{(--)}(\boldsymbol{\kappa}, \Omega; z) \\ &= -\frac{1}{2} n_0 [\sigma(\omega + \Omega) + \sigma(\omega - \Omega)] \Phi_{\mu_1\mu_2}^{(--)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ [\Pi_{\mu_1}^{\mu_1'}(\boldsymbol{\kappa}, \Omega; z) \delta_{\mu_2}^{\mu_2'} + \Pi_{\mu_2}^{\mu_2'}(-\boldsymbol{\kappa}, -\Omega; z) \delta_{\mu_1}^{\mu_1'}] \Phi_{\mu_1'\mu_2'}^{(--)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ X_{\mu_1}^{\mu_1'}(\boldsymbol{\kappa}, \Omega; z) \Phi_{\mu_1'\mu_2}^{(+)}(\boldsymbol{\kappa}, \Omega; z) + X_{\mu_2}^{\mu_2'}(-\boldsymbol{\kappa}, -\Omega; z) \\ &\times \Phi_{\mu_1\mu_2'}^{(+)}(\boldsymbol{\kappa}, \Omega; z) + (2\pi)^2 \hbar \omega \delta(c\boldsymbol{\kappa}_z - \Omega) R_{\mu_1\mu_2}^{(--)}(\boldsymbol{\kappa}, \Omega; z) \end{aligned} \quad (10)$$

for the anomalous correlation function and

$$\begin{aligned} & \frac{\partial}{\partial z} \Phi_{\mu_1\mu_2}^{(+-)}(\boldsymbol{\kappa}, \Omega; z) \\ &= -\frac{1}{2} n_0 [\sigma(\omega - \Omega) + \sigma'(\omega - \Omega)] \Phi_{\mu_1\mu_2}^{(+-)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ \{(-1)^{\mu_1+\mu_1'} [\Pi_{-\mu_1}^{-\mu_1'}(-\boldsymbol{\kappa}, -\Omega; z)] \cdot \delta_{\mu_2}^{\mu_2'} \\ &+ \Pi_{\mu_2}^{\mu_2'}(-\boldsymbol{\kappa}, -\Omega; z) \delta_{\mu_1}^{\mu_1'}\} \\ &\times \Phi_{\mu_1'\mu_2'}^{(+)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ (-1)^{\mu_1+\mu_1'} [X_{-\mu_1}^{-\mu_1'}(-\boldsymbol{\kappa}, -\Omega; z)] \cdot \Phi_{\mu_1'\mu_2}^{(--)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ X_{\mu_2}^{\mu_2'}(-\boldsymbol{\kappa}, -\Omega; z) \Phi_{\mu_1\mu_2'}^{(++)}(\boldsymbol{\kappa}, \Omega; z) \\ &+ (2\pi)^2 \hbar \omega \delta(c\boldsymbol{\kappa}_z - \Omega) \Lambda_{\mu_1\mu_2}^{(+)}(\boldsymbol{\kappa}, \Omega; z) \end{aligned} \quad (11)$$

for the normal correlation function. Repeated indices in (10) or (11) mean summation. In accordance with (7), the functions  $\Phi_{\mu_1\mu_2}^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa}, \Omega; z)$  satisfy the symmetry relations

$$\begin{aligned} \Phi_{\mu_1\mu_2}^{(++)}(\boldsymbol{\kappa}, \Omega; z) &= (-1)^{\mu_1+\mu_2} [\Phi_{-\mu_2-\mu_1}^{(--)}(\boldsymbol{\kappa}, \Omega; z)] , \\ \Phi_{\mu_1\mu_2}^{(+-)}(\boldsymbol{\kappa}, \Omega; z) &= \Phi_{\mu_1\mu_2}^{(+)}(-\boldsymbol{\kappa}, -\Omega; z). \end{aligned} \quad (12)$$

In Eqs. (10) and (11) we have introduced

$$\begin{aligned} \sigma(\omega) &= \sigma'(\omega) + i\sigma''(\omega) \\ &= \frac{4\pi k}{\hbar} \frac{|d_{j'j}|^2}{3(2j+1)} \left[ -i(\omega - \omega_0) + \frac{1}{2} \gamma \right]^{-1}, \end{aligned} \quad (13)$$

which is the cross section for the interaction of the quasire-

sonant photons with an atom,  $k$  is the wave number,  $d_{j'j}$  is the reduced matrix element of the dipole moment of the transition  $j \rightarrow j'$ , and  $n_0$  is the density of atoms. It is assumed that the frequency deviation  $\omega_0 - \omega$  is much larger than the Doppler frequency shift of the atomic transition, so that the inhomogeneous line broadening can be ignored in the calculations.

The covariant components of the tensor  $\Pi_{\mu_1\mu_2}(\boldsymbol{\kappa}, \Omega, z)$  (the nonlinear correction to the retarded polarization operator) are

$$\begin{aligned} \Pi_{\mu_1\mu_2}(\boldsymbol{\kappa}, \Omega; z) &= J e^{-l(z)} \sum_{\nu_1\nu_2} \sum_{\kappa_1q_1} \sum_{\kappa_2q_2} (-1)^{\nu_1+\nu_2} \left| \right. \\ &\times e_{\nu_1}(\mathbf{e}^*)_{\nu_2} (-1)^{\kappa_1-q_1+\kappa_2-q_2} \Pi_{\mu_1\mu_2}^2 \left( \begin{array}{ccc} 1 & \kappa_1 & 1 \\ -\mu_1 & -q_1 & \nu_1 \end{array} \right) \\ &\times \left( \begin{array}{ccc} 1 & \kappa_2 & 1 \\ \nu_2 & -q_2 & -\mu_2 \end{array} \right) \left\{ \begin{array}{cc} 1 & 1 \\ j & j \end{array} \right\} \left\{ \begin{array}{cc} 1 & 1 \\ j & j \end{array} \right\} \\ &\times \left[ \frac{1}{4} \sigma^2(\omega) G_{+j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\kappa}, \Omega; z) \right. \\ &\left. + \frac{1}{4} |\sigma(\omega)|^2 G_{-j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\kappa}, \Omega; z) \right]. \end{aligned} \quad (14)$$

Here  $\mathbf{e}$  is the polarization vector of the pump, and  $l(z) = n_0 \sigma'(\omega) z$  is the optical thickness of the medium at point  $z$  for the photons of frequency  $\omega$ . The functions  $G_{\pm j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\kappa}, \Omega; z)$  are related to the Fourier components of the symmetry and antisymmetric correlation functions introduced in Ref. 12 for the fluctuations of the polarization moments of the Wigner density matrix:

$$\begin{aligned} G_{\pm j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\kappa}, \Omega; z) &= \int d^3\rho \int_{-\infty}^{\infty} d\tau \theta(\tau) e^{-i\boldsymbol{\kappa}\rho + i\Omega\tau} \\ &\times \{K_{(s,a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\rho}, \tau; z) \pm K_{(a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\rho}, \tau; z)\}. \end{aligned} \quad (15)$$

The functions  $K_{(s,a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\rho}, \tau; z)$  are expressed in terms of the correlation functions introduced in Ref. 12 for the polarization moments by

$$\begin{aligned} K_{(s,a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\mathbf{r}_1, t_1; \mathbf{r}_2, t_2) \\ &= \iint \frac{d^3p_1}{(2\pi\hbar)^3} \frac{d^3p_2}{(2\pi\hbar)^3} K_{(s,a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\mathbf{p}_1, \mathbf{r}_1, t_1; \mathbf{p}_2, \mathbf{r}_2, t_2) \\ &\rightarrow K_{(s,a)j,j}^{\kappa_1q_1;\kappa_2q_2}(\boldsymbol{\rho}, \tau; z) \\ &= n_0 \sum_q D_{q_1q}^{\kappa_1} \left( 0, \frac{\pi}{2}, 0 \right) D_{q_2-q}^{\kappa_2} \left( 0, \frac{\pi}{2}, 0 \right) \sum_{\kappa} \frac{1 \pm (-1)^{\kappa_1+\kappa_2+\kappa}}{2} \\ &\times (-1)^{2j} \Pi_{\kappa}^2 \Pi_j \left( \begin{array}{ccc} \kappa_1 & \kappa_2 & \kappa \\ q & -q & 0 \end{array} \right) \left\{ \begin{array}{ccc} \kappa_1 & \kappa_2 & \kappa \\ j & j & j \end{array} \right\} \left( \frac{m}{2\pi T \tau^2} \right)^{3/2} \\ &\times \exp \left( -\frac{m\rho^2}{2T\tau^2} - iq\Omega\tau \right) \rho_j^{\kappa_0}(z), \end{aligned} \quad (16)$$

where  $\boldsymbol{\rho} = \mathbf{r}_1 - \mathbf{r}_2$ ,  $\tau = t_1 - t_2$ ,  $m$  is the mass of an atom,  $T$  is the temperature of the medium (the gas is assumed to be at equilibrium in terms of translational degrees of freedom), and  $D_{q_1q}^{\kappa_1}(\dots)$  are the Wigner  $D$ -functions.<sup>17</sup>

The matrix  $X_{\mu_1\mu_2}(\boldsymbol{\kappa}, \Omega; z)$  in Eqs. (10), (11), which couples the normal and anomalous correlation functions, corresponds to a nonlinear parametric conversion of pump pho-

tons into a pair of conjugate photons with frequencies  $\omega \pm \Omega$  and wave vectors  $\mathbf{k} \pm \boldsymbol{\kappa}$ :

$$\begin{aligned}
 X_{\mu_1, \mu_2}(\boldsymbol{\kappa}, \Omega; z) = & J e^{-l(z) + 2i\theta(z)} \sum_{\nu_1, \nu_2} \sum_{\kappa_1, q_1} \sum_{\kappa_2, q_2} (-1)^{\nu_1 + \nu_2} \\
 & \times e_{\nu_1} e_{\nu_2} (-1)^{\kappa_1 - q_1 + \kappa_2 - q_2} \Pi_{11, \kappa_1, \kappa_2}^2 \begin{pmatrix} 1 & \kappa_1 & 1 \\ -\mu_1 & -q_1 & \nu_1 \end{pmatrix} \\
 & \times \begin{pmatrix} 1 & \kappa_2 & 1 \\ -\mu_2 & -q_2 & \nu_2 \end{pmatrix} \begin{pmatrix} 1 & 1 & \kappa_1 \\ j & j & j' \end{pmatrix} \begin{pmatrix} 1 & 1 & \kappa_2 \\ j & j & j' \end{pmatrix} \\
 & \times \left[ \frac{1}{4} \sigma^2(\omega) G_{+, j; j}^{\kappa_1, q_1; \kappa_2, q_2}(\boldsymbol{\kappa}, \Omega; z) \right. \\
 & \left. + \frac{1}{4} |\sigma(\omega)|^2 G_{-, j; j}^{\kappa_1, q_1; \kappa_2, q_2}(\boldsymbol{\kappa}, \Omega; z) \right], \quad (17)
 \end{aligned}$$

where  $\theta(z) = \theta_0 - \frac{1}{2} n_0 \sigma''(\omega) z$  is the phase of the pump wave at point  $z$ , and  $\theta_0$  is the initial phase.

The sources of the correlation functions distributed over the volume,  $R_{\mu_1, \mu_2}^{(-)}(\boldsymbol{\kappa}, \Omega; z)$  and  $\Lambda_{\mu_1, \mu_2}^{(+)}(\boldsymbol{\kappa}, \Omega; z)$ , are

$$\begin{aligned}
 R_{\mu_1, \mu_2}^{(-)}(\boldsymbol{\kappa}, \Omega; z) = & J e^{-l(z) + 2i\theta(z)} \sum_{\nu_1, \nu_2} \sum_{\kappa_1, q_1} \sum_{\kappa_2, q_2} (-1)^{\nu_1 + \nu_2} e_{\nu_1} e_{\nu_2} (-1)^{\kappa_1 - q_1 + \kappa_2 - q_2} \\
 & \times \Pi_{11, \kappa_1, \kappa_2}^2 \begin{pmatrix} 1 & \kappa_1 & 1 \\ -\mu_1 & -q_1 & \nu_1 \end{pmatrix} \\
 & \times \begin{pmatrix} 1 & \kappa_2 & 1 \\ -\mu_2 & -q_2 & \nu_2 \end{pmatrix} \begin{pmatrix} 1 & 1 & \kappa_1 \\ j & j & j' \end{pmatrix} \begin{pmatrix} 1 & 1 & \kappa_2 \\ j & j & j' \end{pmatrix} \\
 & \times \frac{1}{4} \sigma^2(\omega) [G_{+, j; j}^{\kappa_1, q_1; \kappa_2, q_2}(\boldsymbol{\kappa}, \Omega; z) + G_{+, j; j}^{\kappa_2, q_2; \kappa_1, q_1}(-\boldsymbol{\kappa}, -\Omega; z)], \quad (18)
 \end{aligned}$$

$$\begin{aligned}
 \Lambda_{\mu_1, \mu_2}^{(+)}(\boldsymbol{\kappa}, \Omega; z) = & J e^{-l(z)} \sum_{\nu_1, \nu_2} \sum_{\kappa_1, q_1} \sum_{\kappa_2, q_2} (-1)^{\nu_1 + \nu_2} (\mathbf{e}^*)_{\nu_1} e_{\nu_2} (-1)^{\kappa_1 - q_1 + \kappa_2 - q_2} \\
 & \times \Pi_{11, \kappa_1, \kappa_2}^2 \begin{pmatrix} 1 & \kappa_1 & 1 \\ \nu_1 & -q_1 & -\mu_1 \end{pmatrix} \begin{pmatrix} 1 & \kappa_2 & 1 \\ -\mu_2 & -q_2 & \nu_2 \end{pmatrix} \\
 & \times \begin{pmatrix} 1 & 1 & \kappa_1 \\ j & j & j' \end{pmatrix} \begin{pmatrix} 1 & 1 & \kappa_2 \\ j & j & j' \end{pmatrix} \\
 & \times \frac{1}{4} |\sigma(\omega)|^2 [G_{+, j; j}^{\kappa_1, q_1; \kappa_2, q_2}(\boldsymbol{\kappa}, \Omega; z) + G_{-, j; j}^{\kappa_2, q_2; \kappa_1, q_1}(-\boldsymbol{\kappa}, -\Omega; z)]. \quad (19)
 \end{aligned}$$

Equations (10) and (11), found from diagram equations (5) and (6), describe the evolution of the correlation functions without allowance for the diffraction of the radiation. That approximation is justified if one is dealing with the forward-emitted photons of the conjugate parametric-scattering waves, so that the relation  $k\kappa^{-2} \gg L$  holds, where  $L$  is the length of the cell. In accordance with the requirement (stipulated above) that the spatial scale of the correlations of interest be small, and in accordance with the approximate description of the pump as a monochromatic plane wave, the conditions  $\kappa \gg a_0^{-1}, n_0 \sigma''(\omega)$  must hold. Together, these conditions put limits on the range of the wave-vector transfer  $\boldsymbol{\kappa}$  for which Eqs. (10) and (11) are valid. In evaluating matrices (14) and (17)–(19), we assumed that

the frequency  $\Omega$  was much smaller than the frequency deviation  $\omega_0 - \omega$ , and we ignored the corrections on the order of  $\Omega/|\omega_0 - \omega|$  in these expressions.

The system of transport equations for the normal and anomalous correlation functions in (10), (11) differs in a fundamental way, in terms of the structure of its coefficients, from the equations which are usually discussed in the literature, which correspond to the four-wave mixing in a gas of immobile two-level atoms.<sup>6–8</sup> The difference arises because the coefficients of these equations acquire terms proportional to the nonlinear polarization moments of the density matrix of the working atoms. As is shown below for the particular case of the transition  $j = 1/2 \rightarrow j' = 1/2$ , the deviation of the atoms from equilibrium strongly amplifies the effect of the phase-sensitive parametric-scattering process on the evolution of the correlation functions (and thereby amplifies the squeezing effect). An effective squeezing can be achieved if the pump intensity is well below the saturating value in the wing of the atomic transition. Under the assumption that the pump intensity is low, we ignored the small terms proportional to the ratio  $J/J_s$  (where  $J_s$  is the photon flux density in the pump wave corresponding to the saturation intensity in the line wing) in evaluating the polarization operators from the diagrams of equations (5), (6). To take these terms into account would be to go beyond the accuracy of our approximations [see Eq. (9)]. Note, however, that the terms which have been discarded are the terms which are responsible for the squeezing effect in the case of four-wave mixing involving two-level atoms.<sup>5–8</sup>

To conclude this section of the paper we note that if in solving Eqs. (10), (11) we restrict the discussion to the first iteration and evaluate the photocurrent fluctuation spectrum from (1), using the pump radiation which has passed through the gas-filled cell as the beat-frequency wave, then the expression for the photocurrent spectrum is found to be the same as that which was derived in Ref. 13 in the first nonvanishing approximation in the nonlinear interaction of the pump radiation with the scattering medium.

#### 4. CASE OF THE TRANSITION $j = 1/2 \rightarrow j' = 1/2$

As an example we consider the propagation along the  $z$  axis of circularly polarized pump radiation [ $\mathbf{e} = \mathbf{e}_{+1} = -(\mathbf{e}_x + i\mathbf{e}_y)/\sqrt{2}, e_{-1} = -\delta_{-1\nu}$ ], which interacts with the transition  $j = 1/2 \rightarrow j' = 1/2$  of the working atoms, whose ground state is optically oriented in the direction perpendicular to the  $z$  axis. The correlation functions for the electromagnetic field,  $\Phi_{\mu_1, \mu_2}^{(\sigma_1, \sigma_2)}(\boldsymbol{\kappa}, \Omega; z)$ , are considered near the Zeeman frequency ( $\Omega \sim \Omega_0$ ). In this case, Eqs. (10), (11) become

$$\begin{aligned}
 & \frac{\partial}{\partial z} \Phi_{-1-1}^{(-)}(\boldsymbol{\kappa}, \Omega; z) \\
 & = -\frac{1}{2} n_0 [\sigma(\omega + \Omega) + \sigma(\omega - \Omega)] \Phi_{-1-1}^{(-)}(\boldsymbol{\kappa}, \Omega; z) \\
 & \quad - e^{-2i\theta(z)} [\chi(\boldsymbol{\kappa}, \Omega; z) + \chi(-\boldsymbol{\kappa}, -\Omega; z)] \Phi_{-1-1}^{(-)}(\boldsymbol{\kappa}, \Omega; z) \\
 & \quad + \chi(\boldsymbol{\kappa}, \Omega; z) \Phi_{+1-1}^{(+)}(\boldsymbol{\kappa}, \Omega; z) + \chi(-\boldsymbol{\kappa}, -\Omega; z) \Phi_{-1+1}^{(-)}(\boldsymbol{\kappa}, \Omega; z) \\
 & \quad + (2\pi)^2 \hbar \omega \delta(c\boldsymbol{\kappa} - \Omega) R_{-1-1}^{(-)}(\boldsymbol{\kappa}, \Omega; z), \quad (20)
 \end{aligned}$$

$$\begin{aligned}
& \frac{\partial}{\partial z} \Phi_{+i-1}^{(+)}(\mathbf{x}, \Omega; z) \\
&= -n_0 \sigma'(\omega - \Omega) \Phi_{+i-1}^{(+)}(\mathbf{x}, \Omega; z) - (e^{-2i\theta(z)} \chi(-\mathbf{x}, -\Omega; z) \\
&+ e^{2i\theta(z)} \chi^*(-\mathbf{x}, -\Omega; z)) \Phi_{+i-1}^{(+)}(\mathbf{x}, \Omega; z) \\
&+ \chi(-\mathbf{x}, -\Omega; z) \Phi_{+i+1}^{(+)}(\mathbf{x}, \Omega; z) \\
&+ \chi^*(-\mathbf{x}, -\Omega; z) \Phi_{-i-1}^{(-)}(\mathbf{x}, \Omega; z) \\
&+ (2\pi)^2 \hbar \omega \delta(c\mathbf{x}_z - \Omega) \Lambda_{+i-1}^{(+)}(\mathbf{x}, \Omega; z). \quad (21)
\end{aligned}$$

The parametric conversion of the pump photons is characterized by the constant

$$\begin{aligned}
& \chi(\mathbf{x}, \Omega; z) \\
&= -\frac{1}{8} n_0 \sigma(\omega) J e^{-i(z) + 2i\theta(z)} \{ \sigma'(\omega) [K(\mathbf{x}, \Omega) + K(-\mathbf{x}, -\Omega)] \\
&- ip\sigma''(\omega) [K(\mathbf{x}, \Omega) - K(-\mathbf{x}, -\Omega)] \\
&- p\sigma''(\omega) [Q(\mathbf{x}, \Omega) + Q(-\mathbf{x}, -\Omega)] \\
&- i\sigma'(\omega) [Q(\mathbf{x}, \Omega) - Q(-\mathbf{x}, -\Omega)] \}. \quad (22)
\end{aligned}$$

Here  $p$  is the degree of polarization of the ground state [ $p = p(z) = 3^{1/2} \rho_j^{(0)}(z)$ ], and the functions  $K(\mathbf{x}, \Omega)$  and  $Q(\mathbf{x}, \Omega)$  are defined by

$$\begin{aligned}
& K(\mathbf{x}, \Omega) = \left( \frac{2\pi m}{T\kappa^2} \right)^{1/2} \exp \left[ -\frac{(\Omega - \Omega_0)^2 m}{2T\kappa^2} \right], \\
& Q(\mathbf{x}, \Omega) = 2 \int_0^\infty d\tau \exp \left( -\frac{T\kappa^2}{2m} \tau \right) \sin(\Omega_0 - \Omega) \tau \rightarrow \frac{2}{\Omega_0 - \Omega} + \dots \quad (23)
\end{aligned}$$

for  $|\Omega - \Omega_0| \gg (2T\kappa^2/m)^{1/2}$ . The expressions for the volume sources of the correlation functions which are due to parametric and Raman scattering are

$$\begin{aligned}
& R_{-i-1}^{(-)}(\mathbf{x}, \Omega; z) \\
&= \frac{1}{8} n_0 \sigma^2(\omega) J e^{-i(z) + 2i\theta(z)} \{ K(\mathbf{x}, \Omega) \\
&+ K(-\mathbf{x}, -\Omega) + ip [Q(\mathbf{x}, \Omega) + Q(-\mathbf{x}, -\Omega)] \}, \quad (24) \\
& \Lambda_{+i-1}^{(+)}(\mathbf{x}, \Omega; z) = -\frac{1}{8} n_0 |\sigma(\omega)|^2 J e^{-i(z)} \\
& \times \{ K(\mathbf{x}, \Omega) + K(-\mathbf{x}, -\Omega) - p [K(\mathbf{x}, \Omega) - K(-\mathbf{x}, -\Omega)] \}. \quad (25)
\end{aligned}$$

In Eqs. (20) and (21), the nonlinear corrections to the retarded polarization operator (these corrections are responsible for the space-time dispersion and the absorption) are expressed directly in terms of the parametric-interaction constant, while the polarization of the photons of the conjugate modes which are generated is the same as the polarization of the pump field.

The volume source in the equation for the normal correlation function in (21) describes an occupation of conjugate modes as a result of a quasiresonant Raman scattering. As can be seen directly from (23) and (25), this process, which

is insensitive to the phase of the pump radiation, is efficient in the immediate vicinity of the resonant peaks,  $\Omega \approx \pm \Omega_0$ . The parametric scattering described by constant (22) and by volume source (24), in contrast, is sensitive to the phase of the pump radiation. Its efficiency falls off comparatively slowly in the wings of the resonance lines of the correlation functions [see Eqs. (22)–(24)]. Consequently, examining Eqs. (20), (21) near frequencies  $\Omega$  such that  $|\Omega - \Omega_0| \gg (2T\kappa^2/m)^{1/2}$ , we can ignore the contribution from Raman scattering.

The occupation of the conjugate modes as a result of the nonlinear parametric scattering leads to the formation of an electromagnetic field in a squeezed state with nonequivalent fluctuations of the quadrature components. This statement means that the optimum choice of the phase  $\theta_h$  of the beat-frequency wave causes the second term in (1) to go negative and to partially cancel the contribution from the first term. The situation is complicated, however, by the need to deal with the nonlinear corrections to the space-time dispersion and by the absorption of photons in Eqs. (20) and (21). The additional dispersion makes it impossible to achieve a matching for arbitrary values of  $\Omega$  (the phases of the radiation generated by the parametric generation) in the different parts of a cell, so it leads to a decrease in the effectiveness of the squeezing.

The negative effect of a nonlinear dispersion is not seen for parametric generation in frequencies  $\omega \pm \Omega_1$ , where  $\Omega_1$  is the root of the equation

$$\begin{aligned}
& \sigma''(\omega + \Omega_1) + \sigma''(\omega - \Omega_1) - 2\sigma''(\omega) \\
&= -2 \operatorname{Im} e^{-2i\theta_0} [\chi(\mathbf{x}, \Omega_1; 0) + \chi(-\mathbf{x}, -\Omega_1; 0)], \quad (26)
\end{aligned}$$

where  $\chi(\mathbf{x}, \Omega; 0)$  is the value of constant (22) at the entrance to the gas-filled cell, under the condition that the frequency  $\Omega_1$  lies in the wing of functions (23). In the absence of an absorption [ $I(z) \rightarrow 0$ ], and under the condition  $p = \text{const}_z$ , condition (26) means an identical dispersion law for the pump wave and the anomalous correlation function [see (20)].

Let us assume that the conditions (stipulated above) which lead to ideal squeezing are satisfied. In the case of a heterodyning of the radiation incident on the photocathode by light with the same polarization as the pump ( $\epsilon = e$ ) and in the optimum phase,  $\theta_h = \theta(L) + \pi/4$  (in the case  $\Omega_1 < \Omega_0$  with  $p > 0$ ,  $\omega < \omega_0$ ) or  $\theta_h = \theta(L) - \pi/4$  (in the case  $\Omega_1 > \Omega_0$  with  $p < 0$ ,  $\omega < \omega_0$ ), the photocurrent fluctuation spectrum near the frequency  $\Omega_1$  is then given by the following expression:

$$\begin{aligned}
& \langle |\delta i_a|^2 \rangle \\
&= \zeta I S_0 + \zeta^2 I S_0 \left\{ \frac{(2\bar{\chi})^2}{(2\bar{\chi})^2 - (2\bar{\chi} + \alpha)^2} [\operatorname{ch}[(2\bar{\chi})^2 - (2\bar{\chi} + \alpha)^2]^{1/2} L - 1] \right. \\
&\quad \left. - \frac{|2\bar{\chi}|}{[(2\bar{\chi})^2 - (2\bar{\chi} + \alpha)^2]^{1/2}} \operatorname{sh}[(2\bar{\chi})^2 - (2\bar{\chi} + \alpha)^2]^{1/2} L \right\}, \quad (27)
\end{aligned}$$

where

$$\alpha = \alpha(\Omega) = \frac{1}{2} \sigma''(\omega + \Omega) + \frac{1}{2} \sigma''(\omega - \Omega) - \sigma''(\omega), \quad (28)$$

$$\bar{\chi} = \bar{\chi}(\Omega) = \frac{p}{4} n_0 \sigma''^2(\omega) J \left( \frac{1}{\Omega_0 - \Omega} + \frac{1}{\Omega_0 + \Omega} \right)$$

$$\leftarrow -\frac{i}{2} [\chi(\kappa, \Omega) + \chi(-\kappa, -\Omega)] e^{-2i\theta} \quad \text{for } |\Omega - \Omega_0| \gg \left( \frac{2T\overline{\kappa^2}}{m} \right)^{1/2}. \quad (29)$$

The mean square wave-vector transfer,  $\overline{\kappa^2}$ , which determines the limit in (29), is characterized by the spread of transverse wave vectors of the beat-frequency wave [see (2)]. Using (13), (26), (28), and (29), we find that the frequency  $\Omega_1$  is given approximately by

$$\Omega_1 \approx \Omega_0 + \frac{1}{2} p J \sigma''(\omega) \frac{(\omega_0 - \omega)^2}{\Omega_0^2}. \quad (30)$$

The expression for the photocurrent fluctuation spectrum in (27) and expression (29) clearly demonstrate the fundamental role played by the polarization of the working atoms in forming the squeezed states of the electromagnetic field. In the case  $p = 0$  there is no squeezing effect at all.

A solution of Eqs. (20), (21) which corresponds to ideal squeezing and which leads to expression (27) for the photocurrent spectrum is valid under the condition that the efficiency of the parametric scattering per unit length, characterized by the parameter  $2\tilde{\chi}(\Omega_1)$ , is substantially greater than the linear absorption coefficient  $n_0 \sigma'(\omega)$ . Using (13) and (28)–(30), we find that this condition holds if

$$\Xi = \frac{2\Omega^2}{\gamma |\omega_0 - \omega|} \gg 1. \quad (31)$$

This inequality, combined with the requirement  $\Omega_0 / |\omega_0 - \omega| \ll 1$ , can be satisfied experimentally. In the case of alkali atoms ( $\gamma \lesssim 10^8 \text{ s}^{-1}$ ), for example, with  $\omega_0 - \omega \sim 10^{11} \text{ s}^{-1}$  and  $\Omega_0 \sim 10^{10} \text{ s}^{-1}$ , we have  $\Xi \sim 10$ . In this case, as can be seen directly from (29) and (30), the conditions for an effective squeezing can be satisfied at pump intensities well below the saturating values in the wing of the atomic transition:

$$\left( \frac{\Omega_0}{\omega_0 - \omega} \right)^2 \ll |p J \sigma''(\omega) \tau_{\bar{x}}| \ll 1, \quad \tau_{\bar{x}} = \left( \frac{m}{2T\overline{\kappa^2}} \right)^{1/2}. \quad (32)$$

In order of magnitude, we have  $\gamma^{-1} < \tau_{\bar{x}} < \bar{\tau}$  [see Eq. (9)].

For a clear illustration of the possibilities of the method which we have been discussing here for generating squeezed states of an electromagnetic field, Fig. 2 shows the photocurrent fluctuation spectrum  $\langle |\delta i_{\Omega}|^2 \rangle$  as a function of the dimensionless frequency  $\delta = (\Omega - \Omega_0) \tau_{\bar{x}}$  for various optical thicknesses of the gas-filled cell,  $l = n_0 \sigma'(\omega) L$ . These results were found through a numerical solution of Eqs. (20), (21). We used the following parameter values: a quantum yield  $\zeta = 1$ , a polarization  $|p| = 1$ ,  $\Omega_0 / (\omega_0 - \omega) = 0.1$ ,  $J |\sigma''(\omega)| \tau_{\bar{x}} = 0.2$ , and  $\Xi = 5, 10$ , and  $15$ . Figure 3 shows the minimum fluctuation level  $\langle |\delta i_{\Omega}|^2 \rangle_{\min}$  characterizing the degree of amplitude squeezing as a function of the optical thickness  $l$ . It can be seen directly from Figs. 2 and 3 that the squeezing efficiency increases with increasing  $\Xi$  and that the minimum in the photocurrent fluctuation spectrum coincides with the frequency  $\Omega_1$  in the limit of strong squeezing ( $\delta_1 \approx \pm 10$  for the parameter values just listed). For the Na atoms which were used in the experiments of Refs. 5 and 10, the parameter values corresponding to the curves in Figs. 2 and 3 are reached at frequency deviations  $\omega_0 - \omega \sim 10^{10} - 10^{11} \text{ s}^{-1}$  in the wing of the  $D_1$  line, at

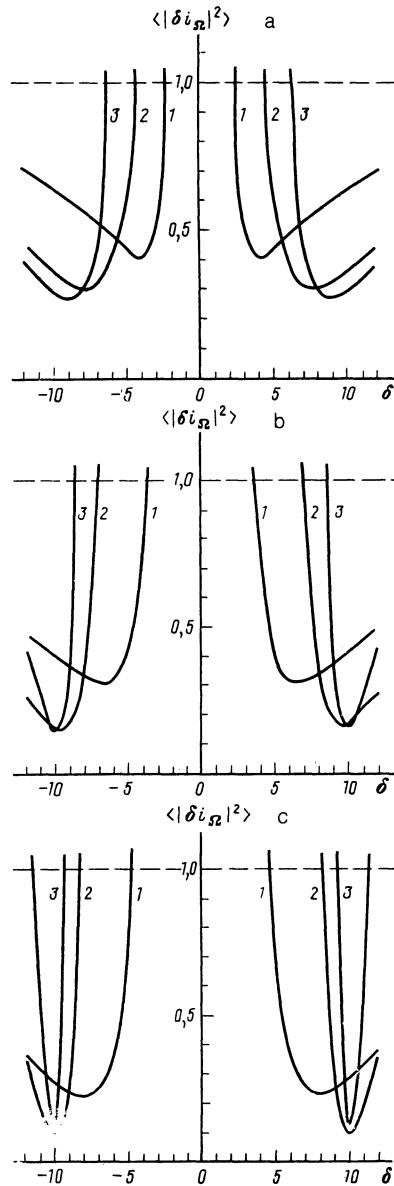


FIG. 2. Photocurrent fluctuation spectrum  $\langle |\delta i_{\Omega}|^2 \rangle$  as a function of the dimensionless frequency  $\delta = (\Omega - \Omega_0) \tau_{\bar{x}}$  for various values of the parameter  $\Xi$ . a—5; b—10; c—15. Here  $\zeta = 1$ ,  $\Omega_0 / (\omega_0 - \omega) = 0.1$ ,  $J |\sigma''(\omega)| \tau_{\bar{x}} = 0.2$ ,  $p = +1$  ( $\delta < 0$ ),  $p = -1$  ( $\delta > 0$ ). Curves 1–3—Optical thickness  $l = 0.1, 0.3, 0.5$ . The scale is drawn with respect to the level of the shot noise.

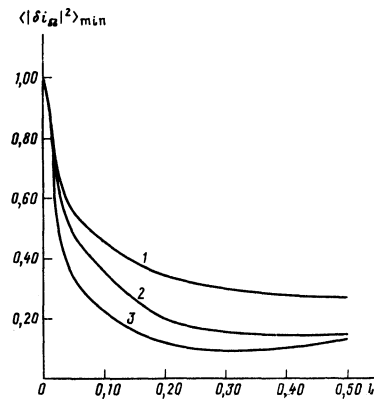


FIG. 3. Minimum fluctuation level  $\langle |\delta i_{\Omega}|^2 \rangle_{\min}$  as a function of the optical thickness of the cell,  $l$ , for  $\zeta = 1$ ,  $\Omega_0 / (\omega_0 - \omega) = 0.1$ ,  $J |\sigma''(\omega)| \tau_{\bar{x}} = 0.2$ ,  $|p| = 1$ . Curves 1–3— $\Xi = 5, 10, 15$ , respectively. The scale is drawn with respect to the level of the shot noise.

$\Omega_0 \sim 10^9 - 10^{10} \text{ s}^{-1}$ , at atomic densities  $n_0 \sim 10^{13} - 10^{14} \text{ cm}^{-3}$ , and at a cell length  $L \gtrsim 10 \text{ cm}$ . With  $\tau_{\pi} \sim 10^{-5} \text{ s}$  [ $(\kappa^2)^{1/2} \gtrsim 10 \text{ cm}^{-1}$ ], and  $|\sigma''(\omega)| \sim 10^{-12} \text{ cm}^2$ , we have  $J \sim 10^{16} \text{ cm}^{-2} \cdot \text{s}^{-1}$ . This figure corresponds to pump intensities  $\sim 1 \text{ mW/cm}^2$ .

## 5. DISCUSSION

How well do the results derived above correspond to results calculated previously<sup>13</sup> by perturbation theory on the nonlinear interaction of radiation with a scattering medium? In Ref. 13, the sub-Poisson statistics of the photocurrent arose in the heterodyning of the scattered radiation directly by the transmitted pump radiation. In the case of a weak interaction of the radiation with the atoms, the eccentricity of the ellipse characterizing the asymmetric distribution of the field fluctuations in the complex-amplitude plane is small. As can be seen from the analysis above, the position of the major semiaxis of the ellipse with respect to the complex amplitude of the transmitted pump radiation is determined by the angle  $\pm (\pi/4 - \delta\theta)$ , where  $|\delta\theta| \ll 1$ . When the transmitted pump radiation is used as the beat-frequency wave, and the fluctuations have a slight asymmetry, sub-Poisson statistics will be observed in the photocurrent fluctuation spectrum if  $\delta\theta > 0$ , while super-Poisson statistics will be observed if  $\delta\theta < 0$ . It follows from the expression for the parametric-conversion constant, (22), that we have  $\delta\theta \sim \sigma'(\omega)/\sigma''(\omega) \sim \gamma/2(\omega_0 - \omega)$  and that the sign of this quantity is determined by the signs of the cross section  $\sigma''(\omega)$ , the polarization  $p$ , and the difference  $\Omega - \Omega_0$ . Upon a change in the sign of the difference  $\Omega - \Omega_0$ , the sign of  $\delta\theta$  also changes, so the statistics of the fluctuations change. As a result, the photocurrent fluctuation spectrum assumes the qualitative shape shown in Ref. 13. In the limit of strong squeezing, a heterodyning of the scattered light by the transmitted pump radiation may lead to only super-Poisson statistics of the photocurrent.

It is quite natural that the multilevel nature of an atom would have a favorable effect on the process by which squeezed states of an electromagnetic field are generated. Again in the interaction of a pump with a system of two-level atoms,<sup>3,4,6-8</sup> we are essentially seeing a manifestation of a "multilevel nature," which stems from the appearance of quasienergy states in the strong external field of the pump radiation. An orientation of the atoms, which is a fundamental condition for the appearance of a squeezed state of the

scattered radiation in the problem of the present paper, can be associated in the case of two-level atoms with an additional phase matching in the evolution of quasienergy sublevels.

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<sup>1</sup> For brevity, the indices  $\sigma_1$  and  $\sigma_2$  are understood as the numbers  $\pm 1$  in (3).

<sup>2</sup> The word "pump," which is used below in connection with parametric scattering, has no bearing on external sources of an incoherent optical pump.

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