

Nonclassical optical effects induced by a bichromatic field

G. Yu. Kryuchkyan

Institute of Physics Research, Academy of Sciences of Armenia, Ashtarak

(Submitted 28 November 1990)

Zh. Eksp. Teor. Fiz. **99**, 1416–1430 (May 1991)

An investigation was made of the quantum statistics of an optical field established as a result of the interaction in an optical resonator of a system of atoms with a bichromatic laser field representing two modes with frequencies located symmetrically relative to the atomic transition frequency. A quantum electrodynamic theory is developed for resonant fluorescence and intermode correlation in a bichromatic field allowing for multiphoton processors, effects of vacuum fluctuations of radiation, and relaxation. The nonlinear absorption coefficient of a mode interacting with such an atomic medium and the intensity of light at the exit from a resonator are found to differ significantly from the corresponding results in the case of a monochromatic field. Calculations are also reported of the rms variance of the quadrature amplitudes and correlation functions of the intensities in the two cases of single- and two-mode excitation in the resonator. In particular, it is shown that a mode at the central frequency of a luminescence line is excited in a squeezed state with suppressed quantum fluctuations of the quadrature amplitude and it results in an interference between the intensities which exhibits nonclassical photon superbunching.

1. INTRODUCTION

The properties of quantum states of light and the various methods of obtaining these states are a subject of major topical interest. It is now known that various nonlinear optical phenomena in nonlinear crystals and atomic media can be used to generate squeezed states of light with quantum fluctuations of one of the quadrature amplitudes below the level of fluctuations corresponding to a vacuum or coherent state. Generation of squeezed light was first achieved in the course of nondegenerate four-wave mixing in a beam of sodium atoms.¹ This process generated two-mode squeezed light with a relatively large spectral width (for theoretical results see Refs. 2 and 3). The strongest degree of squeezing of the quadrature amplitude was achieved in parametric oscillation.⁴ In the majority of both theoretical and experimental investigations it is customary to consider generation of nonclassical light with the aid of monochromatic cw optical fields. Few investigations have been made of the squeezed states of optical pulses.^{5–7}

One of the aims of the present study was to consider the statistical properties of nonclassical light generated on interaction, in an optical resonator, between a system of atoms and a nonmonochromatic two-mode strong field. The results demonstrate another possibility of generation of an optical field in a single-mode squeezed state.

We shall consider a bichromatic field with two components and zero phase shift between their amplitudes and with frequencies $\omega_{1L} = \omega_0 - \delta$ and $\omega_{2L} = \omega_0 + \delta$ which are detuned symmetrically by δ from a resonance with the frequency of the atomic transition ω_0 . A matrix element of a transition between the states of a two-level atom subjected to a bichromatic field, described in a classical manner, is then

$$V(t) = V[\cos(\omega_0 - \delta)t + \cos(\omega_0 + \delta)t]. \quad (1)$$

The dynamics of an atom in a bichromatic field had been investigated already.^{8–10} The resonant fluorescence spectrum was calculated in Ref. 10. This spectrum has a fine structure with peaks at frequencies $\omega_q = \omega_0 + q\delta$, where

$q = 0, \pm 1, \pm 2, \dots$, provided the detuning from the resonance is much greater than the spontaneous width γ of an excited atomic level.

We shall show that when a system of atoms interacts with a bichromatic field in an optical resonator, the mode at the frequency ω_0 of the central fluorescence line is excited in a squeezed state. The physical mechanism responsible for this effect is as follows. The excitation of the mode with the frequency ω_0 in such a resonator is the result of a nonlinear process of two-photon emission from an atom in the bichromatic field. In the lowest order in respect of the interaction of an atom with this field and with the field of its own radiation, the atom can be represented by diagrams describing a four-photon process with the energy conservation law $\omega_{1L} + \omega_{2L} = 2\omega_0$ (Fig. 1). A strong pair correlation is established between the photons of frequencies ω_0 and this suppresses quantum fluctuations of the quadrature amplitudes of the mode ω_0 . This intermode correlation is manifested also in another nonclassical optical effect considered below, photon superbunching in the interference between the intensities.

Single-mode squeezed light may be generated in the following experiment. A beam of identical atoms interacts in a circular optical resonator with two bichromatic field modes of frequencies ω_{1L} and ω_{2L} . A mode of frequency ω_0 and a momentum \mathbf{k} is excited spontaneously in the resonator. In the circular resonator all these modes propagate in the same direction and we have $\mathbf{k}_{1L} + \mathbf{k}_{2L} = 2\mathbf{k}$. One can envisage another experiment in which a bichromatic field is a standing wave propagating at an angle (specifically θ) to atoms moving at a velocity v , whereas the mode ω_0 is excited in the resonator at right-angles to a plane formed by the directions of the motion of atoms and of the propagation of the field. In this case the detuning from a resonance is governed by the Doppler shift so that the two frequencies of the bichromatic field are

$$\omega_{1L} = \omega_0 \left(1 - \frac{v}{c} \cos \theta \right), \quad \omega_{2L} = \omega_0 \left(1 + \frac{v}{c} \cos \theta \right).$$

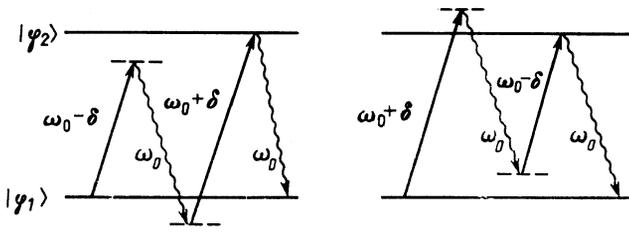


FIG. 1. Graphical illustration of the process of absorption of two laser photons of frequencies $\omega_0 - \delta$ and $\omega_0 + \delta$, accompanied by the emission of two photons of frequency ω_0 . This process results in photon bunching. An atom is transferred from the ground atomic state $|\varphi_1\rangle$ to an excited state $|\varphi_2\rangle$ by a three-photon process in a time of the order of δ^{-1} , followed by the emission of a second photon. Consequently, if the time resolution is $\Delta t \gg \delta^{-1}$, recording of photon coincidences shows that the ω_0 photons are emitted in pairs.

We shall also investigate quantum electrodynamic effects in a resonator when a resonant fluorescence is generated by a bichromatic field. The expressions obtained below for the nonlinear absorption coefficient and for the intensity of light at the exit from the resonator differ considerably from the corresponding familiar results applicable in the case of a monochromatic field.¹¹⁻¹³

We shall consider these topics by the density matrix method using the representation of quasienergy states of a system consisting of an atom and a bichromatic field. Selection of quasienergy states as the basis wave functions of an atom in a bichromatic field¹⁴ simplifies greatly the calculation procedure and still makes it possible to include most fully the effects which are nonlinear in the field. A detailed description of the application of this method to a quantum theory of parametric fluorescence and four-wave mixing can be found in earlier papers of the present author and a colleague.¹⁵⁻¹⁷

The paper is organized as follows. In Sec. 2 we shall give the main kinetic (transport) equations describing the dynamics of modes in a resonator. In Sec. 3 we shall calculate the coefficients in these kinetic equations and describe their properties. In Sec. 4 we shall consider the intensity of the optical field of resonant fluorescence in a bichromatic field at the exit from the resonator. Sections 5 and 6 will be devoted to nonclassical optical effects: the squeezing of the quadrature amplitude and the interference of the intensities of an optical field in the case of single- and two-mode generation configurations.

2. PRINCIPAL EQUATIONS DESCRIBING THE DYNAMICS OF MODES IN A RESONATOR

In this section we shall deal with the underlying assumptions made in considering the interaction of a system of atoms with a laser field and with the modes of the field generated in an optical resonator. The modes with the frequencies ω_i in the resonator are described by the creation a_i^+ and annihilation a_i operators. The laser field is described classically and not specified any further.

The Hamiltonian of the interaction of an atom perturbed by a laser field with the modes of the radiation field in the resonator is

$$H_{int} = -i\mathbf{d}(t) \sum_i \left(\frac{2\pi\hbar\omega_i}{v} \right)^{1/2} [a_i \mathbf{e}(i) e^{-i\omega_i t} - a_i^+ \mathbf{e}^*(i) e^{i\omega_i t}], \quad (2)$$

where $\mathbf{d}(t)$ is the dipole moment operator in the representation of quasienergy states, v is the resonator volume, and $\mathbf{e}(i)$ is the polarization vector.

The coupling between the fields inside and outside the resonator is described by the Hamiltonian

$$H_{SR} = \hbar \int \sum_k d\omega_k [q_i(\omega_k) a_i b_{\omega_k}^+ + \text{H.c.}], \quad (3)$$

where $b_{\omega_k}^+$ are the creation operators for a field photon outside the resonator, satisfying the condition $[b_{\omega_k}, b_{\omega_k'}] = \delta(\omega_k - \omega_k')$; $q_i(\omega_k)$ are the coupling constants.

In the Markov approximation the positive-frequency part of the operator representing the electric field intensity outside the resonator is

$$\mathbf{E}^{(+)}(x, t) = \mathbf{E}_0^{(+)}(x, t) + \left(\frac{4\pi\hbar}{cS} \right)^{1/2} \sum_i (\Gamma_i \omega_i)^{1/2} \mathbf{e}(i) a_i(t) \times \exp \left[-i\omega_i \left(t - \frac{x}{c} \right) - i\varphi_i \right], \quad (4)$$

where $\mathbf{E}_0^{(+)}$ is the operator of the free radiation field; x is a coordinate; S is the cross-section area of the resonator; φ_i is the phase shift due to the propagation of the field across the resonator; the quantity

$$\Gamma_i = \pi |q_i(\omega_i)|^2 \quad (5)$$

is the width of the absorption line of the mode ω_i in the resonator such that $q_i(\omega_i) = |q_i(\omega_i)| \exp(-i\varphi_i)$. The result represented by Eq. (4) is obtained in the standard manner by solving the Heisenberg equations of motion for the operator (see, for example, Ref. 18).

The dynamics of a radiation field mode inside the resonator is described by kinetic equations representing the average number of photons in a mode and by an anomalous correlation function, all of which are deduced using an equation for the density matrix $\rho(t)$ of the radiation field modes. For a medium whose volume is v and the density of atoms is N , the relevant equation is

$$\begin{aligned} \frac{d\rho}{dt} = & \frac{2\pi N}{\hbar} \sum_{ij} (\omega_i \omega_j)^{1/2} \{ [e_n(j) e_m(i) [a_j, a_i \rho] e^{-i(\omega_i + \omega_j)t} \\ & + e_n^*(j) e_m(i) [a_i \rho, a_j^+] e^{i(\omega_j - \omega_i)t}] \Delta_{nm}(\omega_i, t) \\ & + [e_n(j) e_m^*(i) [a_i^+ \rho, a_j] e^{i(\omega_i - \omega_j)t} + e_n^*(j) e_m^*(i) [a_j^+, a_i^+ \rho] \\ & \times e^{i(\omega_i + \omega_j)t}] \Delta_{nm}(-\omega_i, t) \} + \sum_i [a_i \rho, a_i^+] \Gamma_i + \text{H.c.} \end{aligned} \quad (6)$$

The coefficients in this equation are

$$\Delta_{nm}(\omega_i, t) = \int_{-\infty}^t dt_1 e^{i\omega_i(t-t_1)} \langle D_n(t) D_m(t_1) \rangle \quad (7)$$

and

$$D_n(t) = S^+(t) d_n(t) S(t) \quad (8)$$

is the Heisenberg operator of the dipole moment $\mathbf{D}(t)$, which in the representation of quasienergy states is expressed in terms of operators using a scattering matrix $S(t)$ (Ref. 19). The averaging in Eq. (7) is carried out over the ground quasienergy state and over the vacuum radiation field. Equation (6) is derived in the approximation which allows for terms of the second and lower orders in respect of the interactions (2) and (3) in the coefficients Γ_i and $\Delta_{nm}(\omega_i, t)$, by a method described in Refs. 15 and 16.

We shall now give the kinetic equations for the occupancy numbers of the modes $n_i(t) = \text{Tr}(a_i^\dagger a_i \rho(t))$, where $i = 1$ or 2 , and for the anomalous correlation function $g(t) = \text{Tr}(a_1 a_2 \rho(t))$ in the case when only two modes of frequencies ω_1 and ω_2 are excited in the resonator. Calculations carried out allowing for time evolution of the density matrix of (6) give

$$\partial n_i / \partial t = 2(\text{Re } \alpha_i - \Gamma_i) n_i + 2 \text{Re}(\mu_i^* g(t)) + \beta_i, \quad (9)$$

$$\partial g(t) / \partial t = (\alpha_1 + \alpha_2 - \Gamma_1 - \Gamma_2) g(t) + \mu_1^* n_1 + \mu_2^* n_2 + \lambda_{12}. \quad (10)$$

The coefficients in these equations can be expressed in terms of the averages of the product of the dipole-moment operators of Eq. (7) and are as follows:

$$\alpha_i = \frac{2\pi N \omega_i}{\hbar} e_n^*(i) e_m(i) \langle \langle \chi_{nm}(\omega_i, t) \rangle \rangle, \quad (11)$$

$$\mu_i = \frac{2\pi N}{\hbar} (\omega_1 \omega_2)^{1/2} e_n(2) e_m(1) \langle \langle e^{-i(\omega_1 + \omega_2)t} \chi_{nm}(\omega_1, t) \rangle \rangle, \quad (12)$$

$$\beta_i = \frac{4\pi N \omega_i}{\hbar} \text{Re} \{ e_n(i) e_m^*(i) \langle \langle \Delta_{nm}(-\omega_i, t) \rangle \rangle \}, \quad (13)$$

$$\lambda_{12} = \frac{2\pi N}{\hbar} (\omega_1 \omega_2)^{1/2} e_n(1) e_m(2) \times \langle \langle e^{-i(\omega_1 + \omega_2)t} [\Delta_{nm}^*(-\omega_1, t) + \Delta_{mn}^*(-\omega_2, t)] \rangle \rangle, \quad (14)$$

where

$$\chi_{nm}(\omega_i, t) = \Delta_{nm}^*(-\omega_i, t) - \Delta_{nm}(\omega_i, t).$$

The double angular brackets in Eqs. (11)–(14) denote averaging over a time interval which makes these expressions time-independent. One should therefore bear in mind that the coefficients described by (7) are expressed in our approach via the matrix elements of transitions between quasienergy states of an atom in a bichromatic field [see Eq. (17)] and contain, in the case of large time intervals, oscillatory exponential functions of type $\exp[i(\omega_{q_1} + \omega_{q_2})t]$, and $\exp[i(\omega_{q_1} - \omega_{q_2})t]$ with frequencies of the possible spontaneous transitions. Therefore, we can easily see that the coefficients of Eqs. (12) and (14) differ from zero in those cases when the frequencies of two modes in a resonator are related by $\omega_1 + \omega_2 = \omega_{q_1} + \omega_{q_2}$ or $\omega_1 + \omega_2 = \omega_{q_1} - \omega_{q_2}$. However, the coefficients α_i and β_i are governed by time-independent contributions to Eq. (7) and, consequently, they are independent of the condition of phase matching of the modes.

Equations (9) and (10) describe also the degenerate case when $\omega_1 = \omega_2$ and only one mode is excited in the reso-

nator [see Eq. (31)]. It should also be mentioned that the structure of Eqs. (9) and (10) resembles equations obtained in Refs. 15–17 and 20 for the four-wave mixing case, but differ from the latter in respect of the coefficients. We shall derive these equations in a more general form with coefficients which are valid in the case of an arbitrary laser field.

3. COEFFICIENTS OF KINETIC EQUATIONS AND THEIR RELATIONSHIP TO MULTIPHOTON PROCESSES IN A BICHROMATIC FIELD

We shall calculate the coefficients in Eqs. (11)–(14) for two cases: single-mode excitation in a resonator with an eigenfrequency ω_i ; two-mode excitation with frequencies ω_1 and ω_2 coupled by the phase-matching condition $\omega_1 + \omega_2 = \omega_q + \omega_{q_2} = 2\omega_0 + q\delta$, where $q = q_1 + q_2$. We shall do this using linearly independent wave functions of a two-level atom in a bichromatic field with amplitudes varying harmonically with time. In the resonant approximation, subject to the conditions $\delta \ll \omega_0$, and $|V| \ll \omega_0$, where V is the matrix element of a transition between the atomic states $|\varphi_1\rangle$ and $|\varphi_2\rangle$, these wave functions are as follows:

$$|\Phi_j(t)\rangle = c_{j1}(t) |\varphi_1\rangle + c_{j2}(t) e^{-i\omega_0 t} |\varphi_2\rangle, \quad j=1, 2, \quad (15)$$

where

$$c_{11}(t) = c_{22}(t) = \cos\left(\frac{\xi}{2} \sin \delta t\right),$$

$$c_{12}(t) = c_{21}(t) = -i \sin\left(\frac{\xi}{2} \sin \delta t\right),$$

$\xi = 2|V|/\delta$ is the intensity parameter, and the energy of the ground atomic level is assumed to be zero.

The coefficients of Eq. (7) are calculated using the fluctuation-regression theorem. The calculation method is described in Ref. 21 and it is applied to a bichromatic field in Ref. 10. The essence of the method is as follows. A two-time average of the dipole moment operators (8) is expressed in terms of matrix elements

$$\mathbf{d}_{ij}(t) = \langle \Phi_i | \mathbf{d} | \Phi_j \rangle = \mathbf{d}_{ij}^{(-)}(t) + \mathbf{d}_{ij}^{(+)}(t) \quad (16)$$

of dipole transitions between the states described by Eq. (15) and in terms of elements of the atomic density matrix considered in the representation of quasienergy states $\sigma_{ij}(l) = S^\dagger(t) |\Phi_i\rangle \cdot \langle \Phi_j | S(t)$; this average is thus described by

$$\begin{aligned} \langle D(t) D(t') \rangle &= \sum_{ij\sigma\sigma'} \langle \sigma_{ij}(t) \sigma_{\sigma\sigma'}(t') \rangle d_{ij}(t) d_{\sigma\sigma'}(t') \\ &= d_{11}(t) d_{11}(t') [\Delta^2 + (1 - \Delta^2) e^{-\tau_1(t-t')}] + 1/2 e^{-\tau_2(t-t')} \\ &\quad \times [d_{12}(t) d_{21}(t') (1 + \Delta) + d_{21}(t) d_{12}(t') (1 - \Delta)]. \end{aligned} \quad (17)$$

This expression contains

$$\Delta = \sigma_{11} - \sigma_{22} = 4J_0(\xi) / [3 + J_0(2\xi)] \quad (18)$$

the difference between the steady-state populations of quasienergy states $|\Phi_1\rangle$ and $|\Phi_2\rangle$, where $J_0(\xi)$ is a Bessel function, whereas the quantities

$$\gamma_1 = \frac{\gamma}{4} [3 + J_0(2\xi)], \quad \gamma_2 = \frac{\gamma}{8} [5 - J_0(2\xi)] \quad (19)$$

determine respectively the decay of the diagonal and off-

diagonal components of the density matrix $\langle \sigma_{ij} \rangle$. It should be noted that a purely spontaneous decay of atomic states is considered and no allowance is made for atomic collisions, but the decay constants γ_1 and γ_2 allow for the effects of the bichromatic field intensity. In the absence of this field we have $\gamma_1 = 2\gamma_2$.

The matrix elements (16) of the dipole transitions between the quasienergy states of Eq. (15) are

$$\begin{aligned} d_{ij}^{(-)}(t) &= \sum_q e^{-i\omega_q t} d_{ij}^{(q)}, \\ d_{ij}^{(+)}(t) &= \sum_q e^{i\omega_q t} d_{ij}^{(q)*}, \end{aligned} \quad (20)$$

$$d_{11}^{(q)} = -d_{22}^{(q)} = \frac{d}{4} J_q(\xi) (1 - \cos q\pi),$$

$$d_{12}^{(q)} = \frac{d}{2} \left[\delta_{q0} + J_q(\xi) \frac{1 + \cos q\pi}{2} \right],$$

$$d_{21}^{(q)} = \frac{d}{2} \left[\delta_{q0} - J_q(\xi) \frac{1 + \cos q\pi}{2} \right].$$

Using Eqs. (17)–(20) we can calculate the function $\Delta(\omega_i, t)$, and, consequently, the coefficients given by Eqs. (11)–(14). We must bear in mind that the function $\Delta(-\omega_i, t)$ cannot be obtained from $\Delta(\omega_i, t)$ by the substitution $\omega_i \rightarrow -\omega_i$, because the resonance approximation is used in the calculations. This function is equal to the complex conjugate of $\Delta(\omega_i, t)$ subject to an additional substitution $\Delta \rightarrow -\Delta$. We shall give only the final results.

a. Nonlinear absorption coefficient

We obtain the following expression for the coefficient α_i :

$$\alpha_i = -\frac{8\pi\omega_i N}{\hbar} \frac{J_0^2(\xi)}{3 + J_0(2\xi)} \frac{|\mathbf{de}(i)|^2}{i(\omega_0 - \omega_i) + \gamma_2}. \quad (21)$$

This quantity represents the polarizability of an atomic medium in the presence of a bichromatic field when the radiation field mode has the frequency ω_i . An analysis of Eq. (9) demonstrates that in the case of short time intervals the real part describes the absorption of the mode in the region of $\omega_i \approx \omega_0$. Near other spectral lines $\omega_i \approx \omega_0 + q\delta$, where $q \neq 0$, of a two-level atom in a bichromatic field there is no mode absorption followed by re-emission in the atomic medium. Some comments are due about this result.

The quantity $\text{Re } \alpha_i$ defined by Eqs. (7) and (11) generally describes both amplification and absorption of a mode whose frequency is ω_i . In the adopted approach this quantity is determined by the matrix elements of the dipole transitions between quasienergy states, which also determine the probabilities of elementary processes. In a bichromatic field for time intervals in the range $t \ll \gamma^{-1}$ the probabilities of spontaneous emission or absorption of a photon of frequency $\omega_0 + q\delta$, $q \neq 0$, by one atom as a result of a specific transition between quasienergy states are the same irrespective of whether it is emission or absorption [see Refs. 8–10 and the system of equations (20)]. Therefore, the mode amplifica-

tion and absorption effects cancel each other and in the vicinity of $\omega_i \approx \omega_0 + q\delta$ the polarizability vanishes. In the spectral range $\omega_i \approx \omega_0$ the situation is quite different. The matrix elements of the transitions (20) for $q = 0$ and, consequently, the probabilities of elementary processes of absorption or emission of photons of frequency ω_0 , are not equal. The absorption probability exceeds the probability of emission for the same intensity parameters. This results in the absorption of a mode of frequency $\omega_i \approx \omega_0$ followed by its re-emission in the investigated atomic medium. It should be noted that $\text{Re } \alpha_i < 0$ applies throughout the full range of the intensity parameter ξ .

It is necessary to stress also that the results described by Eq. (21) differ considerably from the corresponding results in the case of a monochromatic resonant field. In the latter case, as shown theoretically in Ref. 22 and experimentally in Ref. 11, in addition to the absorption of a weak field by a two-level atom in the presence of a resonant field, there is also amplification of the weak field at a frequency which is symmetric relative to the frequency of the resonant field. This effect is also manifested in resonant fluorescence inside an optical resonator.^{12,13}

b. Rate of one-photon spontaneous emission

Equations (7), (13), (17)–(20) lead to the following expression for the coefficient β_i :

$$\begin{aligned} \beta_i = \frac{\pi\omega_i N}{\hbar} |\mathbf{de}(i)|^2 \sum_q \left\{ \gamma_1 \frac{(1 - \Delta^2) J_{2q+1}^2(\xi)}{(\omega_i - \omega_{2q+1})^2 + \gamma_1^2} \right. \\ \left. + \gamma_2 \frac{\delta_{q0} [1 - 2\Delta J_0(\xi)] + J_{2q}^2(\xi)}{(\omega_i - \omega_{2q})^2 + \gamma_2^2} \right\}. \end{aligned} \quad (22)$$

When the above quantity is multiplied by $1/Nv$, it describes the steady-state rate of emission of photons of frequency ω_i in the course of resonant fluorescence in a bichromatic field. In accordance with our formulation of the problem, the contribution of the elastic forward scattering is omitted from the above result, but apart from that stipulation, Eq. (22) is identical with the expression derived in Ref. 10 by a different method. We can show that $\Delta^2 \leq 1$, and both the first and second terms in Eq. (22) are positive throughout all the investigated range of ξ .

c. Rate of spontaneous emission of a photon pair

In calculation of the coefficients μ_1 and μ_2 representing the coupling between the modes and the coefficient λ_{12} , which governs the anomalous correlation function in the case of short time intervals $g(t) \approx t\lambda_{12}^* + \dots$, we have to specify the conditions of the intermode frequency matching. We shall consider the situation when $\omega_1 + \omega_2 = 2\omega_0 + q\delta$, and q is an integer or zero. In this case the process of time averaging in Eqs. (12) and (14) leaves only the nonvanishing contributions to the function $\Delta(\omega_i, t)$ which contain an exponential function of the $\exp[i(2\omega_0 + q\delta)t]$ type. The result is

$$\begin{aligned}
 &= \frac{\pi N}{2\hbar} (\omega_1 \omega_2)^{1/2} (\mathbf{e}(1) \mathbf{d}) (\mathbf{e}(2) \mathbf{d}) \sum \left\{ \frac{1}{4} (1 - \Delta^2) J_{q_1}(\xi) J_{q-q_1}(\xi) \right. \\
 &\times (1 - \cos q_1 \pi) [1 - \cos(q - q_1) \pi] \left[\frac{1}{i(\omega_0 + q_1 \delta - \omega_1) + \gamma_1} \right. \\
 &+ \left. \left. \frac{1}{i[\omega_1 - (q - q_1) \delta - \omega_0] + \gamma_1} \right] + \left[\delta_{q_1,0} \delta_{q-q_1,0} - \frac{1}{4} J_{q_1}(\xi) J_{q-q_1}(\xi) \right] \right. \\
 &\times (1 + \cos q_1 \pi) (1 + \cos(q - q_1) \pi) \left. \left[\frac{1}{i(\omega_0 + q_1 \delta - \omega_1) + \gamma_2} \right. \right. \\
 &\left. \left. + \frac{1}{i[\omega_1 - (q - q_1) \delta - \omega_0] + \gamma_2} \right] \right\}. \quad (23)
 \end{aligned}$$

The above equation describes spontaneous emission of a pair of photons whose resonance frequencies are in the range $\omega_1 \approx \omega_0 + q_1 \delta$ and $\omega_2 \approx \omega_0 + (q - q_1) \delta$ or $\omega_1 \approx \omega_0 + (q - q_1) \delta$ and $\omega_2 \approx \omega_0 + q_1 \delta$. It should be pointed out that the pole contributions to Eq. (23) at frequencies $\omega_0 + q_1 \delta$ and an odd value of q_i (with the factor $1 - \cos q_1 \pi$ in the numerator) are due to diagonal transitions $|\Phi_1\rangle \rightarrow |\Phi_1\rangle$, $|\Phi_2\rangle \rightarrow |\Phi_2\rangle$ between quasienergy states of Eq. (15), whereas the pole contributions at frequencies $\omega_0 + 2q_1 \delta$ are due to off-diagonal transitions $|\Phi_1\rangle \rightleftharpoons |\Phi_2\rangle$.

We shall give the result for the case when the eigenfrequencies of the two resonator modes are linked by $\omega_1 + \omega_2 = 2\omega_0$:

$$\begin{aligned}
 \lambda_{12} &= \frac{\pi N}{\hbar} (\omega_1 \omega_2) (\mathbf{e}(1) \mathbf{d}) (\mathbf{e}(2) \mathbf{d}) \left\{ \frac{\gamma_2 [1 - J_0^2(\xi)]}{(\omega_1 - \omega_0)^2 + \gamma_2^2} \right. \\
 &+ \sum_{2q+1 > 0} (\Delta^2 - 1) \gamma_1 J_{2q+1}^2(\xi) \left[\frac{1}{[\omega_1 - \omega_0 - (2q+1)\delta]^2 + \gamma_1^2} \right. \\
 &+ \left. \left. \frac{1}{[\omega_1 - \omega_0 + (2q+1)\delta]^2 + \gamma_1^2} \right] \right. \\
 &- \sum_{q > 0} \gamma_2 J_{2q}^2(\xi) \left[\frac{1}{(\omega_1 - \omega_0 - 2q\delta)^2 + \gamma_2^2} \right. \\
 &+ \left. \left. \frac{1}{(\omega_1 - \omega_0 + 2q\delta)^2 + \gamma_2^2} \right] \right\}. \quad (24)
 \end{aligned}$$

It follows from the calculations that when the modes are phase-matched $\omega_1 + \omega_2 = 2\omega_0$, the coefficients μ_1 and μ_2 representing the coupling between the modes vanish throughout the frequency range (ω_1, ω_2) . This situation is specific to the optical-field configuration considered by us and is related to the mutual compensation, discussed above, of the mode amplification and absorption effects in a medium of two-level atoms in a bichromatic field.

It should be pointed out that Eq. (17) is derived in the approximation of nonoverlapping spectral lines when the condition $\delta \gg \gamma$ is satisfied and the broadening of each spectral line, which is of the order of γ , is small compared with the separation δ from the next line, so that the spectrum has a fine structure. Consequently, the results obtained in the present section are valid only in this approximation.

4. INFLUENCE OF A RESONATOR ON THE FLUORESCENCE OBSERVED IN THE BICHROMATIC FIELD

We shall investigate the intensity of light at the exit from the resonator in the case when one mode of frequency ω_i is excited in the resonator. It follows from Eq. (4) that in the case of vacuum generation of radiation this intensity is

$$I = \langle \mathbf{E}^{(-)} \mathbf{E}^{(+)} \rangle = \frac{4\pi\hbar}{cS} \Gamma_i \omega_i n_i. \quad (25)$$

The resonator gives rise to steady-state values of the number of photons in the mode. Using the steady-state solution of Eq. (9),

$$n_i = \beta_i / 2(\Gamma_i - \text{Re } \alpha_i), \quad (26)$$

and allowing for $\mu_1 = \mu_2 = 0$, we find that the intensity at the exit is

$$I = \frac{2\pi\hbar\omega_i}{cS} \beta_i / \left(1 - \frac{\text{Re } \alpha_i}{\Gamma_i} \right). \quad (27)$$

This expression describes the dependence of the intensity on the eigenfrequency ω_i and the width Γ_i of the absorption line in the resonator, on the intensity parameter ξ , and on a quantity $\sigma = 4\pi\omega_0 N |de|^2 / \hbar\gamma$, which is the absorption coefficient of the mode at the frequency of an atomic transition in the absence of a pump field. The nonlinear absorption coefficient is negative, so that Eq. (27) is valid also in the case of resonators with arbitrarily small values of Γ_i .

If the eigenfrequency of the resonator is equal to the frequency of the atomic transition, $\omega_i = \omega_0$, then using Eqs. (21) and (22) we find that the intensity I_0 of the central resonant fluorescence peak is

$$I_0 = \frac{\pi\hbar\omega_0\Gamma}{cS} \frac{3 + J_0(2\xi) [1 + J_0^2(\xi)] - 5J_0^2(\xi)}{[3 + J_0(2\xi)] [5 - J_0(2\xi)] \Gamma / 4\sigma + 4J_0^2(\xi)}, \quad (28)$$

where Γ is the width of the absorption line of the ω_0 mode in the resonator.

One further feature of the nonlinear absorption coefficient in the case of a bichromatic field should be noted. If $\omega_i = \omega_0$, it follows from Eq. (21) that

$$\text{Re } \alpha_0 = \text{Re } \alpha_i |_{\omega_i = \omega_0} = -\sigma \frac{16J_0^2(\xi)}{[3 + J_0(2\xi)] [5 - J_0(2\xi)]}. \quad (29)$$

For certain values of the intensity parameter equal to the roots of $J_0(\xi) = 0$ we find that Eq. (29) vanishes. We can easily see that this effect can be attributed to vanishing of the difference $\Delta(\xi)$ between the steady-state populations of quasienergy states for the same values of the parameter ξ because of the interference between different multiphoton transitions under the action of a bichromatic field (the lowest values in the sequence of roots are $\xi = 2.3, 5.5, 8.7, 11.7, \dots$). The dependence of the ratio $\text{Re } \alpha_0 / \sigma$ on the parameter ξ is plotted in Fig. 2.

This feature of the nonlinear absorption coefficient is manifested by the intensity of light at the resonator exit. It follows from the general expression (27) that in the region of $\omega_i = \omega_0$ the maximum intensity corresponds to those values of the intensity parameter for which the atomic medium is transparent: $\text{Re } \alpha_0 = 0$. Figure 3 shows the dependence of

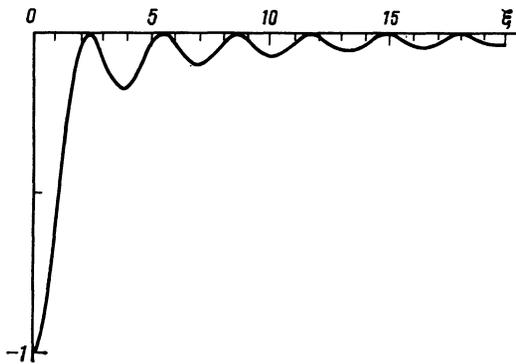


FIG. 2. Dependence of the ratio $\text{Re } \alpha_0/\sigma$ of the nonlinear absorption coefficient to the conventional absorption coefficient on the parameter ξ .

the dimensionless ratio $I_0 (4\pi\hbar\omega_0\Gamma/cS)^{-1}$ on the parameter ξ calculated for two values of the ratio Γ/σ using Eq. (28). We can easily see that a reduction in the ratio Γ/σ increases considerably the intensity.

The absorption coefficient σ is proportional to the number density of atoms. At low densities the intensity I_0 is low and it increases with N as $(1 + \text{const}/N)^{-1}$. If $\xi \ll 1$, the intensity tends to zero and in the range $\xi \gg 1$ the intensity is independent of the resonator width, as can easily be demonstrated.

We shall give also the expression for the intensity of light in the case when the eigenfrequency of the resonator is close to the atomic transition frequency but not identical with the latter:

$$I = \frac{\pi\hbar\omega_i\Gamma_i}{cS} \frac{1 - 2\Delta(\xi)J_0(\xi) + J_0^2(\xi)}{\Delta(\xi)J_0(\xi) + 2\Gamma_i[(\omega_i - \omega_0)^2 + \gamma_2^2]/\sigma\gamma_2}. \quad (30)$$

When the resonator eigenfrequency lies in a different spectral range, particularly when it coincides with the sidelines $\omega_i = \omega_0 + q\delta$, where $q \neq 0$, it follows from Eq. (21) that there is no absorption and the intensity of Eq. (27) is proportional to the rate of photon emission in the form of resonant fluorescence when a bichromatic field is acting in the absence of a resonator.

5. SUPPRESSION OF QUANTUM FLUCTUATIONS OF QUADRATURE AMPLITUDES

a. Single-mode case

Nonclassical optical effects in the emission spectrum are usually due to an anomalous correlation function. We shall begin by considering the case of excitation of a single-

mode field in a resonator and we shall use Eq. (10) for the correlation function $\langle a_i^2 \rangle$. Using $\mu_1 = \mu_2 = 0$, we obtain

$$\frac{\partial}{\partial t} \langle a_i^2 \rangle = 2(\alpha_i - \Gamma_i) \langle a_i^2 \rangle + \lambda^*(\omega_i). \quad (31)$$

The coefficient

$$\lambda(\omega_i) = \frac{4\pi N}{\hbar} \omega_i e_n(i) e_m(i) \langle \langle e^{-2i\omega_i t} \Delta_{nm}^*(-\omega_i, t) \rangle \rangle \quad (32)$$

differs from zero only when $\omega_i = \omega_0$ and it follows from Eq. (24) that this coefficient is given by

$$\lambda_0 = \lambda(\omega_i) |_{\omega_i = \omega_0} = \frac{8\pi N \omega_0}{\hbar \gamma} (\mathbf{de})^2 \frac{1 - J_0^2(\xi)}{5 - J_0(2\xi)}. \quad (33)$$

The quantity λ_0 describes the process of emission of a pair of photons with the same frequency ω_0 .

It follows that a steady-state solution of Eq. (31) exists only at a resonator frequency equal to ω_0 and is given by

$$g_0 = \langle a_i^2 \rangle |_{\omega_i = \omega_0} = \lambda_0 / 2(\Gamma - \alpha_0). \quad (34)$$

Finally, we obtain the following expression for the anomalous correlation function:

$$|g_0| = \frac{1}{4} \frac{1 - J_0^2(\xi)}{[5 - J_0(2\xi)]\Gamma/4\sigma + \Delta(\xi)J_0(\xi)}. \quad (35)$$

We know that the minimum value of the rms variance of the quadrature amplitude

$$A_i = a_i e^{-i\varphi} + a_i^+ e^{i\varphi} \quad (36)$$

of a single-mode radiation field inside a resonator corresponds to a specific phase φ and is given by

$$V_i = \langle (\Delta A_i)^2 \rangle_{\min} = 1 + 2(n_i - |\langle a_i^2 \rangle|). \quad (37)$$

Using Eqs. (26) and (35) for the average number of photons and the anomalous correlation function, we find that the variance $V_0 = V_i |_{\omega_i = \omega_0}$ of the quadrature amplitude of the mode ω_0 is

$$V_0 = 1 - \frac{1}{4} \frac{1 - J_0(2\xi)}{1 + \Gamma[3 + J_0(2\xi)][5 - J_0(2\xi)]/16J_0^2(\xi)\sigma}. \quad (38)$$

This result describes the level of rms quantum fluctuations of a single-mode field and it is below the level in vacuum: $V_0 < 1$. The level of the fluctuations is equal to the level of vacuum fluctuations ($V_0 = 1$) in both limiting cases of weak and strong fields when $\xi \ll 1$ and $\xi \gg 1$ and also for those val-

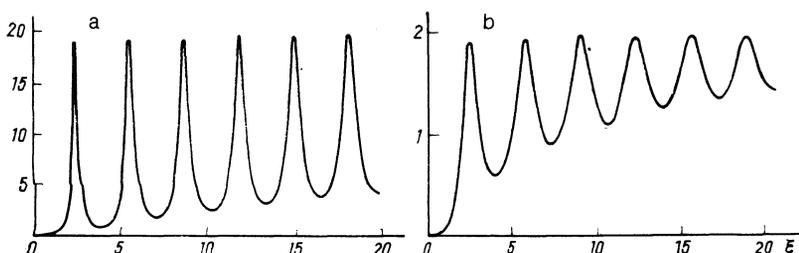


FIG. 3. Dependence, on the parameter ξ , of the normalized light intensity $I_0 (4\pi\hbar\omega_0\Gamma/cS)^{-1}$ of the mode ω_0 at the resonator exit, calculated for two values of the ratio Γ/σ : a) 0.01; b) 0.1.

ues of the intensity parameter which satisfy the condition $J_0(\xi) = 0$.

Figure 4 shows the dependences, based on Eq. (38), of the variance on the intensity parameter calculated numerically for two values of the ratio Γ/σ . We can easily see that the squeezing effect is significant in a good resonator when $\Gamma \ll \sigma$. If $\Gamma/\sigma = 0.01$, the minimum variance is $V_0 = 0.68$ and it corresponds to the intensity parameter $\xi = 1.8$. The intensity of single-mode light at the resonator exit is then $I_0 = 4\pi\hbar\omega_0\Gamma/cS$. In the regions where the intensity rises the variance tends to unity. In particular, if $\xi = 2.1$, we have $I_0(4\pi\hbar\omega_0\Gamma/cS)^{-1} = 4.2$, and the variance is $V_0 = 0.74$.

In the case of other frequencies $\omega_i = \omega_0 + q\delta$ and for side lines in the spectrum of resonant fluorescence in a bichromatic field the steady-state value of the anomalous correlation function $\langle a_i^2 \rangle$ vanishes and single-mode squeezed states are not obtained.

b. Two-mode case

We shall now consider generation of a two-mode optical field subject to the frequency matching condition $\omega_1 + \omega_2 = 2\omega_0$, $\omega_1 \neq \omega_2 \neq \omega_0$ in a squeezed state. The resonances of the radiation frequency then occur at $\omega_1 \approx \omega_0 + q\delta$ and $\omega_2 \approx \omega_0 - q\delta$, where q is an arbitrary even or odd value. In the same spectral range there is no absorption of modes in the atomic medium so that the steady-state solutions of Eqs. (9) and (10) are

$$n_1 = \beta_1/2\Gamma_1, \quad g = \lambda_{12}^*/2\Gamma_1. \quad (39)$$

It is recognized here that $\mu_1 = \mu_2 = 0$ and $\Gamma_1 = \Gamma_2$, and the quantities β_1 and λ_{12} are taken from Eqs. (22) and (24).

Comparison of Eqs. (22) and (24) also easily shows that in the regions of frequencies ω_1 and ω_2 which are not too close to the frequency of an atomic transition, and on the assumption that the polarizations of two modes coincide, we have

$$\beta_1 = \beta_2 = |\lambda_{12}|. \quad (40)$$

If the operator representing the intensity of a two-mode field of Eq. (4) is described by

$$E^{(+)} = E_0^{(+)} + (8\pi\hbar\omega_0\Gamma/cS)^{1/2}A(t)e^{-i\omega_0 t}, \quad (41)$$

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

and if we bear in mind that $|\omega_1 - \omega_2| \ll \omega_0$, where $\epsilon = \omega_0 - \omega_1 = \omega_2 - \omega_0$, we find that the quadrature amplitude

$$A_\theta(t) = A(t)e^{-i\theta} + A^*(t)e^{i\theta},$$

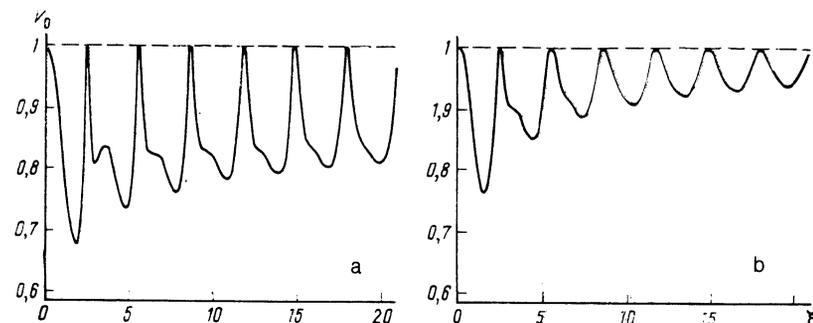


FIG. 4. Dependence, on the parameter ξ , of the rms variance of quantum fluctuations of the quadrature amplitude of a mode in a resonator, calculated for two values of the ratio Γ/σ : a) 0.01; b) 0.1.

as demonstrated in Ref. 23, has a time-invariant rms variance of quantum fluctuations. The minimum variance for a given phase θ is

$$R = \langle (\Delta A_\theta)^2 \rangle_{\min} = 1 + n_1 + n_2 - 2|g|. \quad (42)$$

Using the solutions (39) and the relationship (40), we find that $R = 1$, i.e., the level of rms fluctuations of a two-mode field is identical with the level of the vacuum state.

6. INTENSITY INTERFERENCE EFFECT

The nonclassical nature of single-mode light of frequency ω_0 at the resonator exit is manifested also in another optical phenomenon, intensity interference described by a second-order correlation function. For identical times the normalized correlation function is

$$g^{(2)} = \langle (E^{(-)})^2 (E^{(+)})^2 \rangle / I^2. \quad (43)$$

In the vacuum case the field of the mode ω_i in the resonator obeys the Gaussian statistics, so that the quantity (43) can be factorized and, if we use Eqs. (4) and (25), it becomes

$$g^{(2)} = 2 + |\langle a_i^2 \rangle|^2 / n_i^2. \quad (44)$$

If the resonator eigenfrequency is ω_0 , we can use Eqs. (26) and (35) to obtain the following result:

$$g^{(2)} = 2 + \left[\frac{1 - J_0^2(\xi)}{1 - J_0^2(\xi) [5 - J_0(2\xi)] [3 + J_0(2\xi)]^{-1}} \right]^2, \quad (45)$$

which does not include a dependence on the absorption width of the resonator.

In weak fields or in the case of a large detuning from a resonance, $\xi \ll 1$, we can use an expansion in Bessel functions. Including terms up to the order of ξ^2 , this expansion yields

$$g^{(2)} \approx 2 + (16/5\xi^2)^2, \quad (46)$$

which describes the nonclassical phenomenon of photon superbunching: $g^{(2)} \gg 2$.

In the range $\xi \gg 1$, the correlation function is $g^{(2)} = 3$. The dependence of the normalized correlation function on the intensity parameter is plotted in Fig. 5. We can easily see that $g^{(2)} \gg 3$ throughout the investigated range of the intensity parameter and the equality sign corresponds to those values of ξ for which we have $J_0(\xi) = 0$.

At other frequencies $\omega_i \neq \omega_0$ the anomalous correlation function vanishes and, consequently, we have $g^{(2)} = 2$ and the resonator field then obeys the statistics of single-mode chaotic light.

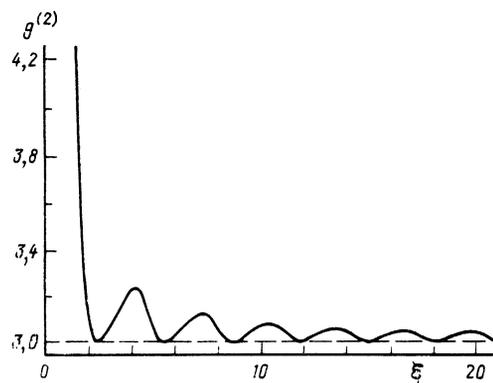


FIG. 5. Dependence, on the parameter ξ , of the normalized correlation function of the intensity of light of the mode ω_0 at the resonator exit.

We shall give the result for the two-mode field discussed earlier in Sec. 5b. The normalized correlation function of Eq. (43) for this case has the following factorized form:

$$g^{(2)} = 2 + 4|g|^2 / (n_1 + n_2)^2. \quad (47)$$

Using the solutions of Eq. (39) and the relationship (40), we find that $g^{(2)} = 3$ throughout the investigated range of the parameters.

7. CONCLUSIONS

These calculations confirm, as stated in the Introduction, that the process of resonant fluorescence in a bichromatic field inside a resonator results in the emission of light in a squeezed state. The experimental procedure suggested above should result, in contrast to the familiar nondegenerate four-wave mixing,¹⁻³ in the generation of single-mode squeezed light at the frequency of an atomic transition, but two-mode squeezed states are not obtained. The degree of squeezing depends on the ratio Γ/σ and on the intensity parameter $\xi = 2|V|/\delta$ [Eq. (38)]. In the case of a mode in a squeezed state the second-order normalized correlation function describing the interference of intensities of light beams is independent of the width of the absorption line of the resonator and for low values of the intensity parameter ξ it gives rise to the quantum effect of photon superbunching: $g^{(2)} > 2$.

In the author's opinion another important result of the present study relates to the amplification or absorption of modes of the radiation field in an atomic medium in the presence of a bichromatic laser field. It is shown that in contrast to a monochromatic laser field, there is no mode amplification in the presence of a bichromatic field. The mode is absorbed after its re-emission in an atomic medium only in the region of the atomic transition frequency and for certain specific values of the intensity parameter ($\xi = 2.3, 5.5, 8.7$, etc.) the nonlinear absorption coefficient vanishes, i.e., the medium is transparent. The minimum variance of the quadrature amplitude of single-mode compressed light inside the resonator is $V_0 = 0.68$ when $\Gamma/\sigma = 0.01$. The squeezing ef-

fect decreases in the region of the parameter ξ where the light intensity rises and the atomic medium is transparent.

One should also mention that the majority of the results obtained in the present study can be readily generalized to the case of a bichromatic laser field with a nonzero relative phase between the amplitudes. Calculations carried out for this case showed that the result given by Eqs. (21), (22), (27), (38), and (45) and representing, respectively, the nonlinear absorption coefficient, the rate of resonant fluorescence, the intensity of light at the exit from the resonator, and the rms variance and the normalized correlation function are not affected by such a generalization. If we allow for the phase ψ between the two component amplitudes of a bichromatic field, we find that the coefficients λ_{12} [Eqs. (23) and (24)] and λ_0 [Eq. (33)] are multiplied by the phase factor $\exp(i\psi)$ and this in particular is manifested in the result for a phase-dependent variance of the fluctuations of the quadrature amplitude [Eq. (36)].

The author is grateful to participants of a seminar of the Theoretical Division of the Institute of Physics Research of the Armenian Academy of Sciences and particularly to K. V. Kheruntsyan for discussions.

- ¹ R. E. Slusher, L. W. Hollberg, B. Yurke *et al.*, Phys. Rev. Lett. **55**, 2409 (1985).
- ² M. D. Reid and D. F. Walls, Phys. Rev. A **34**, 4929 (1986).
- ³ D. A. Holm and M. Sargent, Phys. Rev. A **35**, 2150 (1987).
- ⁴ L. A. Wu, H. J. Kimble, J. L. Hall *et al.*, Phys. Rev. Lett. **57**, 2520 (1986).
- ⁵ B. Yurke, P. Grangier, R. E. Slusher *et al.*, Phys. Rev. A **35**, 3586 (1987).
- ⁶ R. E. Slusher, P. Grangier, A. LaPorta *et al.*, Phys. Rev. Lett. **59**, 2566 (1987).
- ⁷ A. V. Belinskii and A. S. Chirkin, Zh. Eksp. Teor. Fiz. **98**, 407 (1990) [Sov. Phys. JETP **71**, 228 (1990)].
- ⁸ B. V. Klyachanovskiy and A. O. Melikyan, Opt. Commun. **29**, 164 (1979).
- ⁹ S. P. Goreslavskii and V. P. Kraĭnov, Opt. Spektrosk. **47**, 825 (1979) [Opt. Spectrosc. (USSR) **47**, 457 (1979)].
- ¹⁰ G. Yu. Kryuchkov, Opt. Commun. **54**, 19 (1985).
- ¹¹ F. Y. Wu, S. Ezekiel, M. Ducloy *et al.*, Phys. Rev. Lett. **38**, 1077 (1977).
- ¹² D. A. Holm and M. Sargent, J. Opt. Soc. Am. B **2**, 1456 (1985).
- ¹³ Yifu Zhu, A. Lezama, and T. W. Mossberg, Phys. Rev. A **39**, 2268 (1989).
- ¹⁴ V. I. Ritus, Zh. Eksp. Teor. Fiz. **51**, 1544 (1966) [Sov. Phys. JETP **24**, 1041 (1967)].
- ¹⁵ S. T. Gevorkyan and G. Yu. Kryuchkov, Zh. Eksp. Teor. Fiz. **92**, 2034 (1987) [Sov. Phys. JETP **65**, 1145 (1987)].
- ¹⁶ S. T. Gevorkyan and G. Yu. Kryuchkov, Zh. Eksp. Teor. Fiz. **94**(12), 125 (1988) [Sov. Phys. JETP **67**, 2461 (1988)].
- ¹⁷ S. T. Gevorkian and G. Yu. Kryuchkov, Opt. Commun. **67**, 451 (1988).
- ¹⁸ J. Peřina, *Quantum Statistics of Linear and Nonlinear Phenomena*, Reidel, Dordrecht, Netherlands (1981).
- ¹⁹ G. Yu. Kryuchkov, Zh. Eksp. Teor. Fiz. **83**, 1992 (1982) [Sov. Phys. JETP **56**, 1153 (1982)].
- ²⁰ D. A. Holm, S. An, and M. Sargent, Opt. Commun. **60**, 328 (1986).
- ²¹ G. Yu. Kryuchkov, V. E. Mkrtchyan, M. L. Ter-Mikaelyan *et al.*, Zh. Eksp. Teor. Fiz. **88**, 30 (1985) [Sov. J. Quantum Electron. **61**, 17 (1985)].
- ²² B. R. Mollow, Phys. Rev. A **5**, 2217 (1972).
- ²³ C. M. Caves and B. L. Schumaker, Phys. Rev. A **31**, 3068 (1985).

Translated by A. Tybulewicz