New phenomena in quantum optics: photon antibunching, sub-Poisson photon statistics, and squeezed states

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The concepts listed in the title are introduced and clarified. The relations among them are pointed out. It is shown that the corresponding optical phenomena are definitely of a quantum nature. The basic quantities which are measured in correlation optical experiments are discussed. Manifestations of photon antibunching and of sub-Poisson photon statistics in the recording of delayed coincidences, in the distribution of photon counts, and in intensity fluctuation spectra are analyzed. (More precisely, what is involved is the spectrum of fluctuations in the photocurrent; all the results of the theoretical analysis in this review are expressed in terms of directly measurable quantities.) A diagram technique for calculating the correlation characteristics of the radiation from a polyatomic system is outlined. Photon correlations in nonlinear resonance fluorescence are analyzed. There is a discussion of approaches to the development of macroscopic sources of radiation with sub-Poisson photon statistics: the production of a squeezed state of a field in phase-sensitive parametric effects in nonlinear optics and the introduction of repulsive statistics in luminescence excitation events and in the pumping of a laser. Experiments in these directions which had been reported through April 1986 are discussed. The practical importance of reducing the quantum noise in radiation for extremely precise measurements and for optical data transmission is pointed out.

CONTENTS

I. INTRODUCTION

Research on the topics in the title of this review, which are associated with concepts and phenomena in optics, began in the 1970s and is presently in a rising stage of development. We can apparently conclude that we have reached the crest of this curve when we see the reliable experimental realization of macroscopic light sources with a photon flux of elevated regularity. It is this elevated regularity which is the primary characteristic of the electromagnetic-field states which we will be discussing. It means a lowering of the fluctuations in the radiation in comparison with the shot-noise level which can be attained in single-mode, steady-state lasing at a pump level far above the threshold.

Let us examine the basic concepts in the examples of known experiments on the temporal correlations of photons.[1,2] Figure 1a is a simplified diagram of an experiment carried out to count delayed binary coincidences of photons.[3] A light beam from a steady-state source S is split into two beams, which are sent to photodetectors D_1 and D_2. The arrangement records coincidences of photon detection events delayed by a time τ. The average count rate over the

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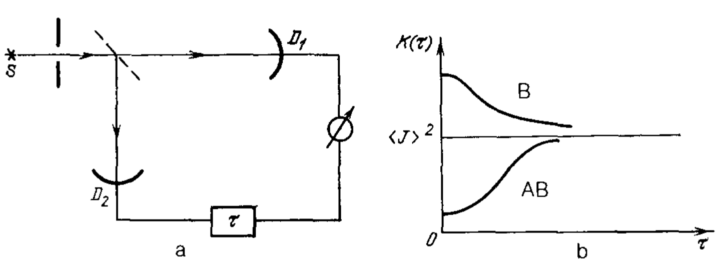

FIG. 1. Experiment on the statistics of delayed coincidences of photon counts. a—Simplified experimental layout; b—some representative photocurrent correlation functions. B) In the case of photon bunching; AB) in the case of photon antibunching.

ensemble of measurements is proportional to the correlation function of the photocurrent, $K(\tau)$, of the two photodetectors: $K(\tau) = \langle J_1(0)J_2(\tau)\rangle$ (the exact definitions of the quantities which are actually measured are discussed in Sec. 2). The term "photon bunching" (Fig. 1b) means an increase in the number of binary coincidences at small values of τ above the "random" number, i.e., above the number corresponding to the absence of correlations: $K(\tau) > \langle J\rangle^2$ at small values of τ ($\langle J_1\rangle = \langle J_2\rangle$ is assumed for simplicity). Photon bunching is characteristic of natural (quasithermal) light sources. It results from an interference of the waves from different points of the source (atoms) (Sec. 3). The random modulation of the intensity associated with this interference can frequently (but not always, as we will see below) be described in a semiclassical version of the theory (without quantization of the electromagnetic field—through a modelling of the stochastic field tailored to the particular light source and by means of a quantum-mechanical description of the interaction of this field with the atoms of the photodetector).

For single-mode, steady-state laser radiation, with a pump level well above the threshold, in which case the state of the field approaches a Glauber coherent state, the equality $K(\tau) = \langle J\rangle^2$ holds; i.e., the photon detection events are not statistically dependent.[3]

Already in the pioneering papers on quantum optics (Refs. 4–7; see also Ref. 3) it was pointed out that there could also be states of an electromagnetic field with "negative" binary correlations of photons, i.e., with a photon "antibunching." In this case, the relation $K(\theta) < \langle J\rangle^2$ would hold in an experiment recording delayed coincidences of photon detection events (AB in Fig. 1b). It might be said that when antibunching occurs the conditional probability for observing a "second" photon immediately after a "first" photon, with a short delay τ, averaged over the ensemble of pairs of delayed coincidences of photon detection events (for all values of τ), is smaller than the unconditional probability for the observation of a photon.

Let us examine the particular features of photon correlations in an experiment carried out to measure the distribution of photon detection events $p(n,T)$: the probability that precisely n photon pulses will be detected over a time T. We assume that radiation is incident on an ideal photodetector (Sec. 2). In this case the temporal realizations of the photocurrent can be represented as a train of short pulses. Figure 2 shows some realizations of this sort for various sources.[8] Let us consider the variance $D[n;T] = \langle (n-\bar{n})^2\rangle$ of the distribution $p(n;T)$ of the number of photon counts ($\bar{n}$ is the mean number of photon counts). A Poisson distribution $p(n) = (\bar{n}^n/n!)n^{-\bar{n}}$ with a variance $D[n] = \bar{n}$ is known to be characteristic of a field in a coherent state[4] (Fig. 2b). A bunching of photons corresponds to the inequality $D[n] > \bar{n}$, i.e., to the case in which the scatter in the photocurrent pulses which fall in a given time interval T is large in comparison with a Poisson distribution (Fig. 2a). An antibunching corresponds to smoother temporal distributions of the pulses (Fig. 2c). If $D[n] < \bar{n}$, one speaks in terms of sub-Poisson photon statistics (SPPS).

In a study of the statistics of radiation in a single mode from a natural source over a time $T \ll \tau_c$, where τ_c is the coherence time, one finds a Bose-Einstein distribution $p(n) = (\bar{n}^n)/(\bar{n}+1)^{n+1}$, and in this case we have $D[n] = \bar{n} + \bar{n}^2$. Spatial correlations play an important role. If the detector collects radiation from an area S which is much larger than the spatial-coherence area at the surface of the photocathode, σ_c, and if the time of a single measurement, T, exceeds τ_c, then random interference will lead to a variance $D[n] = \bar{n}(1+\delta')$, where $\delta' = (1/2)(\sigma_c/S)\langle J\rangle\tau_c = q\delta$, δ is the radiation degeneracy parameter (the mean number of photons which pass through the coherence area over the coherence time), and q is the quantum yield of the photodetector. Under the condition $\delta \ll 1$ (the number of photons in the coherence volume is small) the photon distribution differs only slightly from a Poisson distribution.

An interesting case of photon superbunching, $D[n] > \bar{n} + \bar{n}^2$, occurs in the case of two-photon emission. In this case, pairs of photons are correlated at least over a time interval of the order of ω_{21}^{-1}, where ω_{21} is the transition frequency.[9] In the parametric splitting ($\vec{\omega}_0 = \omega_1 + \omega_2$), in addition to the strong positive temporal correlation, there is also a hard correlation of the photons ω_1 and ω_2 in terms of directions (a "biphoton").[10] Some new applications of radiation with correlations of this sort for precise measurements and data transmission have already been found and are being discussed.[10,11]

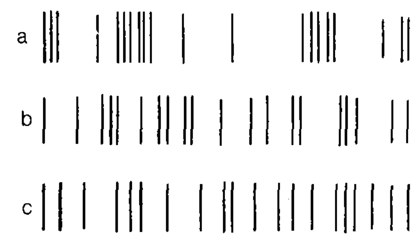

FIG. 2. Examples of realizations of photocurrent pulses for various statistical properties of the radiation. a—Bunching and super-Poissonian statistics; b—Poisson distribution; c—antibunching and sub-Poissonian statistics.

Photon antibunching was first observed experimentally in 1977, in the nonlinear resonance fluorescence of single atoms in a rarefied atomic beam (Ref. 12). The effect had been predicted in Refs. 13–15 (polyatomic effects were also discussed in Ref. 15, along with quantum-mechanical interference). After a refinement of the theory, an analysis of the experimental conditions, and the establishment of good agreement between theory and experiment,[16–18] a stream of theoretical papers appeared. In 1983 came a report[19] of the detection of SPPS in the nonlinear resonance fluorescence of a single atom (see also Ref. 20). The variance of the number of photon counts under these conditions had been calculated previously.[21–23] The overall distribution of photon counts was studied theoretically in several papers.[23–27] Figure 6 shows a typical result of the experiments of Refs. 12 and 17 on the count of delayed coincidences (this figure is taken from Ref. 18; see Sec. 3 of the present paper).

The Fermi features of the photon distribution in the presence of photon antibunching and SPPS of course do not contradict the requirements of Bose-Einstein statistics. The antibunching and the SPPS stem from the specific dynamics of the source—from "repulsive" factors in the emission or conversion of a flux of photons.[1)] In the case of the nonlinear resonance fluorescence of a single atom, with steady-state monochromatic excitation, the repulsive factor is the unavoidable delay in successive scatterings of incident photons. Under the given conditions in the preparation of the system and the observation, this delay agrees with the time-varying evolution of the population of the excited level, $\rho^{(11)}{}_{22}(\tau)$ (the superscripts on a density matrix element specify the initial condition at $\tau = 0$): The detection of the "first" photon is indirect, but the atom in state 1 is reliably detected (Fig. 6; see Sec. 3 for more details).

As another example from nonlinear optics we consider the propagation of a coherent wave in a medium with a two-photon absorption. In this case the antibunching and the SPPS can be represented as a consequence of a more probable absorption from a Poisson flux of pairs of photons which lie close together along the time scale (from regions of elevated intensity). The photon flux produced as a result must therefore be more regular than the incoming flux. Antibunching and SPPS in several effects of nonlinear optics are discussed in other reviews.[8,29,30]

Sub-Poisson photon statistics may be a consequence of the conversion of some "repulsive" statistics of excitation events into the statistics of radiation (Secs. 5,6). Teich and Saleh[176] have experimentally observed an SPPS effect (although a weak one) in the luminescence of atoms excited by an electron beam with shot noise suppressed.

Antibunching and SPPS may reveal a radiation field in "squeezed states," which are being discussed widely in the literature (Secs. 2,4; there is a good brief review on the subject[31]). In the squeezed state of a harmonic oscillator, the variance of one of two canonically conjugate observables is smaller than in a coherent state. For a single standing wave of a free electromagnetic field, we write the electric field operator in the Heisenberg picture:

$$\mathbf{E}(t,\ \mathbf{r}) = \frac{1}{2}\vec{\mathscr{E}}(\mathbf{r})\,[a\exp(-i\omega t) + a^{+}\exp(i\omega t)]; \qquad (1.1)$$

here a^+ and a are the photon creation and annihilation operators (more generally, they are the "slow"-amplitude operators), and $\mathscr{E}(\mathbf{r})$ is the spatial mode function of the field. We introduce the Hermitian operators (quadrature components)

$$X_1 = \frac{1}{2}(a^+ + a), \quad X_2 = \frac{i}{2}(a^+ - a), \qquad (1.2)$$

which are formally analogous to the dimensionless operators q and p for the harmonic oscillator. The field operator $\mathbf{E}$ takes the form

$$\mathbf{E}(t,\ \mathbf{r}) = \vec{\mathscr{E}}(\mathbf{r})\,(X_1\cos\omega t + X_2\sin\omega t). \qquad (1.3)$$

For the quadrature components X_1,X_2 we find the following results from definition (1.2) and the commutation relation $[a,a^+]$:

$$[X_1,\ X_2] = \frac{i}{2}, \quad \delta X_1\delta X_2 \geqslant \frac{1}{4}, \quad (\delta X)^2 \equiv D[X]. \qquad (1.4)$$

In a squeezed state we have, by definition,

$$\delta X_1 < \frac{1}{2} \quad \text{or} \quad \delta X_2 < \frac{1}{2}. \qquad (1.5)$$

Squeezed states and two-photon coherent states appeared as generalizations of coherent states.[32–34] (see Refs. 31, 34, and 206 for citations of the literature).

Disparities in the development of quadrature components (their mean values and fluctuations) in parametric processes are a familiar effect in classical nonlinear optics and radiophysics.[35] Such disparities set the stage for the suppression of amplitude or phase fluctuations of the resultant field. The situation is analogous to that regarding quantum fluctuations: The variance of one of the quadrature components can decrease, and a squeezed state and SPPS can be achieved as a result of phase-sensitive interactions of waves as they propagate through a nonlinear medium (Sec. 4).

The discovery and study of new states and new statistical properties of radiation have been regular features in the development of optics. We know that fluctuations in the energy of blackbody radiation played an important role in the development of quantum statistics[36] and our understanding of the particle-wave dualism.[37,38] In Einstein's formula[36,39] for the variance of the spectral energy density of blackbody radiation or in the equivalent formula for the variance of the number of photons in a single field mode,

$$D[m] = \overline{m} + \overline{m}^2, \qquad (1.6)$$

the first term corresponds to the statistics of independent (and distinguishable) particles, while the second can be related to an interference of waves (or the indistinguishability of photons).

The sharply increased interest in the statistical characteristics of radiation since the 1960s was stimulated by the development of ideas regarding the interference of partially coherent light beams,[40] the first (prelaser) experiments on intensity correlations[1,2] and optical mixing,[41] and (later) the development of lasers and the need for a detailed study of the unusual properties of laser light. New methods were developed for correlation measurements[42–45]; a systematic quantum theory of the statistical characteristics of radiation was derived[3–7,46,47]; and numerous applications of the new research methods were found in physics, chemistry, technology, and biology.[48–54] An ideology based on the theory of random processes and random fields,[55–59] combined with the methods of physical kinetics,[59–61] became the governing force in modern optics.[35]

Photon antibunching, SPPS, and the properties of squeezed states are optical effects which are definitely of a quantum nature. A comparison of quantum-mechanical and semiclassical approaches usually makes use of an "optical equivalence theorem"[7] which asserts that a representation of the field density matrix which is diagonal in the coherent states (the P representation) can be used to calculate and interpret the mean values of normally ordered products of field operators by means of relations which are formally similar to classical relations (if the latter have been written for analytic signals[40,7]). This theorem clearly does not apply to field states with photon antibunching or SPPS or to squeezed states.[2)] For such states, the weight function $P(\alpha)$, if it exists, is at any rate not positive definite, and it cannot be associated with the probability density of a classical complex field amplitude. As an example we consider the variance of the number of photons in a single radiation mode, assuming the existence of a P representation:

$$D[m] = \langle (a^+a - \langle a^+a\rangle)^2\rangle = \langle a^+aa^+a\rangle - \overline{m}^2$$
$$= \overline{m} + \langle :(a^+a - \overline{m})^2:\rangle = \overline{m} + \int d^2\alpha P(\alpha)(|\alpha|^2 - \overline{m})^2; \quad (1.7)$$

here $\overline{m} = \langle a^+a\rangle = \int d^2\alpha P(\alpha)|\alpha|^2$, and $:(\ldots):$ means the normal-ordering operation.[65] In the case of SPPS the second term in (1.7) is negative [but it must of course be greater than $(-\overline{m})$], so there exists a region of α values in which the condition $P(\alpha) < 0$ holds.

A theoretical description of photon antibunching and SPPS requires a thorough analysis of the measurement procedure and a correct determination of the operators of the quantum-mechanical observables which are associated with the measured quantities. Here it is particularly important to allow for the noncommutativity of the dynamic variables and discrete photon detection events.[3)] In Sec. 2 it is shown that the expression

$$K(\tau) = \frac{1}{2}[J(0), J(\tau)]_+ = \langle I\rangle\,\delta(\tau) + K_2(\tau) \quad (1.8)$$

[expression (2.5)] should be used for the photocurrent autocorrelation function (in an experiment with a single photodetector). The term with the δ-function reflects the discrete nature (shot noise) mentioned above. Only the regular part of (1.8), $K_2(\tau)$, contains a field correlation function of fourth order, but it is not proportional to the correlation function of the intensity of the incident light. In a classical description of the field in a single beam we would be dealing with a single function: the light intensity $I(t,\mathbf{r})$, which takes on (regardless of measurements) a definite value at each point $(t,\mathbf{r})$ (even if this is a random value before the measurement; the concept of a random field is unavoidable even in a classical approach). From the inequality $\langle (I(0) - I(\tau))^2\rangle \geqslant 0$ and the condition for a steady state, $\langle I(\tau)\rangle \equiv \langle I(0)\rangle$, we then find

$$K_{2,\mathrm{cl}}(\tau) = q^2\langle I(0)I(\tau)\rangle \leqslant q^2\langle I(0)^2\rangle = K_{2,\mathrm{cl}}(0). \quad (1.9)$$

This inequality is incompatible with antibunching. The function $K_2(\tau)$, which arises in a systematic quantum calculation of the photocurrent correlation function (1.8), does not necessarily obey condition (1.9), since it is not proportional to $\langle I(0)I(\tau)\rangle$. We could have $K_2(0) < K_2(\infty)$ and $K_2(0) = 0$ (strong antibunching).

Analogously, in an experiment with two photodetectors, in which the mutual correlation function of the intensities of two beams is measured, there are two functions, $I_1(t,\mathbf{r}), I_2(t,\mathbf{r})$, in the classical description. The mutual correlation function is

$$\langle I_1(t_1, \mathbf{r}_1) I_2(t_2, \mathbf{r}_2)\rangle^2 \leqslant \langle I_1^2(t_1, \mathbf{r}_1)\rangle\langle I_2^2(t_2, \mathbf{r}_2)\rangle. \quad (1.10)$$

Experiments on the photon correlations of two beams (ω_1) and (ω_2) which arise in spontaneous parametric frequency $(\omega_0 \doteq \omega_1 + \omega_2)$ down-conversion[68,69] and in cascade radiative transitions[8,70] have revealed a "violation" of inequality (1.10) at small values of $|t_2 - t_1|$ (the violation is particularly marked in the first of these cases[68]). In other words, it has been found that the classical description of the field does not hold.[4)]

The definitely quantum nature of photon antibunching and SPPS is sometimes thought of as peculiar to these effects. This is not the case. In the experiments which we mentioned above on the correlations of two beams, no photon antibunching occurred (it would be more appropriate to speak in terms of a superbunching in time). We would cite yet another important example, closely related to the topic of this review, which demonstrates the need for the quantization of a field in the analysis of the correlation properties of the ordinary spontaneous emission of independently excited and decaying atoms.[54,71] Aleksandrov *et al.*[71] measured the intensity fluctuation spectrum of this emission or, more precisely, the spectrum of the correlation function $K(\tau)$ [see (1.8)]. The concept of a classical wavepacket $\vec{\mathscr{E}} = \vec{\mathscr{E}}_0 \exp(-\gamma t/2)\cos\omega_{21}t$, emitted by each atom leads to the incorrect prediction that the intensity fluctuation spectrum should manifest correlations in a single train (from a single atom), in the form of a Lorentz line at $\omega \approx 0$, with a width γ and an intensity proportional to the number of atoms, N (not proportional to N^2). The appearance of such a line in the intensity fluctuation spectrum would have to be associated with the possibility of a "division" of a wave packet between two atoms of the photodetector. The experiment of Ref. 71 reliably established that there is no such peak in the intensity fluctuation spectrum above the spectral level of the shot noise. A systematic quantum theory[72] for the intensity fluctuation spectrum under these conditions yields results which agree entirely with the experimental results.[71] The variance of the number of photon counts, determined by the correct function $K(\tau)$, also has no traces of excess bunching over times of the order of[73] γ^{-1}.

There is a characteristic manifestation of photon antibunching in an intensity fluctuation spectrum. The strong-antibunching condition $K_2(0) = 0$ means that the spectrum of the function $K_2(\tau)$ has zero "area," i.e., contains negative components. In the spectrum of the photon correlation function $K(\tau)$ these components are seen as dips against the spectral background of the shot noise [the first term in (1.8)].[15] The spectrum of the overall autocorrelation function of $K(\tau)$ is of course nonnegative everywhere.[55,56] The suppression of shot noise in a certain frequency region is a general property of radiation with photon antibunching and SPPS. It may be important for applications. Measurements of intensity fluctuation spectra have several advantages over other methods for studying photon correlations.[15,22,54] It was in intensity fluctuation spectra that manifestations of a squeezed state were first observed.[167] (Sec. 4 of the present paper).

This review gives preference to the problem of the development of macroscopic light sources of elevated regularity. The studies carried out through December 1985 which have been of greatest importance, in the opinion of the authors, and also some later studies are considered. In some cases, the only source which we cite is a review which contains references to a group of earlier studies in a common direction.

2. MEASURABLE QUANTITIES

2.1. Photocurrent correlation function

A quantum theory of photodetection pertaining to research on photon correlations and the coherence of light was derived to a decisive extent by Glauber[4] (see also Refs. 6 and 7). Smirnov *et al.*[74] give several versions of the derivation of the expression for the photocurrent correlation function. Those derivations immediately reveal the (photodetector response)-field-(source dynamics) path. We will be concerned for the most part below with the statistical characteristics of the radiation which are associated with the photocurrent autocorrelation function:

$$K(t_1, t_2)=\left\langle \frac{1}{2}[J(t_1), J(t_2)]_+\right\rangle. \tag{2.1}$$

Here $[A,B]_+$ is the anticommutator,[39] and $J(t)$ [s^{-1}] is the photocurrent operator (in the Heisenberg picture), given by

$$J(t)=\frac{\mathrm{d}}{\mathrm{d}t}N(t)=\frac{\mathrm{d}}{\mathrm{d}t}\sum_k N_k(t), \tag{2.2}$$

where the operator $N(t)$ represents the number of atoms of the photodetector which are ionized by the light by time t, and $N_k(t)$ is a single atom operator with eigenvalues 0,1. The angle brackets in (2.1) and below mean the quantum-mechanical expectation value: $\langle A\rangle = \mathrm{Sp}\{\rho A\}$ where ρ is a statistical operator (density matrix) of the system, in this case the source-field-photodetector system. In accordance with expression (2.2), we write the correlation function $K(t_1,t_2)$ in the form $K(t_1,t_2) = K_1(t_1,t_2) + K_2(t_1,t_2)$, where

$$K_1(t_1, t_2)=\frac{\mathrm{d}^2}{\mathrm{d}t_1\mathrm{d}t_2}\sum_k\left\langle\frac{1}{2}[N_k(t_1), N_k(t_2)]_+\right\rangle, \tag{2.3}$$

$$K_2(t_1, t_2)=\frac{\mathrm{d}^2}{\mathrm{d}t_1\mathrm{d}t_2}\sum_{k\neq i}\sum\left\langle\frac{1}{2}[N_k(t_1), N_i(t_2)]_+\right\rangle. \tag{2.4}$$

The first term determines the shot noise; the second is the informative part of the correlation function. In order to express (2.3)–(2.4) in terms of the field characteristics we should use a perturbation theory in the interaction of the field with the atoms of the photodetector. We ignore any optical nonlinearity of the photodetection. The lowest nonvanishing order in the interaction for $K_1(t_1,t_2)$ is the second order [i.e., $K_1(t_1,t_2)$ is proportional to the mean light intensity); that for $K_2(t_1,t_2)$ is the fourth order. We assume that the spectral sensitivity function of the photodetector is considerably broader than the spectrum of the radiation under study. Under this condition, the photoabsorption event is localized in time (the particular instant of course remains random!), and a δ-correlated shot component of the signal is generated. This component is unrelated to the correlation properties of the light $K_1(t_1,t_2)\propto\delta(t_2-t_1)$.

Under these approximations we find the following expression for the correlation function[72,74] (2.1):

$$\begin{aligned}K(t_1, t_2)&=\frac{qc}{2\pi\hbar\omega_0}\sum_\mu\int\mathrm{d}S\,\langle E_\mu^{(-)}(t_1,\mathbf{r})\,E_\mu^{(+)}(t_2,\mathbf{r})\rangle\,\delta(t_2-t_1)\\&+\left(\frac{qc}{2\pi\hbar\omega_0}\right)^2\sum_{\mu,\nu}\int\mathrm{d}S_1\int\mathrm{d}S_2\,[\langle E_\mu^{(-)}(x_1)\,E_\nu^{(-)}(x_2)\,E_\nu^{(+)}(x_2)\\&\qquad\times E_\mu^{(+)}(x_1)\rangle\,\theta(t_2-t_1)\\&+\{x_1\leftrightarrow x_2\}].\end{aligned} \tag{2.5}$$

Here q is the quantum yield (efficiency) of the photodetection; c is the velocity of light; ω_0 is the central frequency of the radiation; the subscripts μ and ν represent Cartesian coordinates; $\mathbf{E}^{(\pm)}(x)$ are the positive- and negative-frequency parts of the operator representing the electric field of the light wave[5]; $x = (t,\mathbf{r})$, where ($\mathbf{r}$ are those points on the photocathode over which the integration is carried out; and $\theta(\tau) = 1$ at $\tau>0$ and $\theta(\tau) = 0$ at $\tau<0$. The interaction of the field with the photodetector atoms was already incorporated in the transformation from (2.3), (2.4) to (2.5). The operators $E_\mu^{(\pm)}(x_i)$ in (2.5) are thus written in the Heisenberg picture of the source-field system.

For experiments with two separate photodetectors, in which the correlation function $\widetilde{K}(t_1,t_2) = \langle(1/2)[J_1(t_1), J_2(t_2)]_+\rangle$ is measured, the expression for $\widetilde{K}(t_1,t_2)$ differs from (2.5) in that the first (shot) term is absent and in the circumstance that the integration is carried out separately over the surfaces of the first ($\mathbf{r}_1$) and second ($\mathbf{r}_2$) photocathodes.

In the steady state the correlation function $K(t_1,t_2)$ depends only on the difference $\tau = t_2 - t_1$.

In addition to direct measurement of the time evolution of the correlation function, it has become common to measure the intensity fluctuation spectrum, i.e., the spectrum of the function[45,54] $K(\tau)$:

$$G(\omega)=\int_{-\infty}^{\infty}\mathrm{d}\tau\exp(i\omega\tau)\,K(\tau). \tag{2.6}$$

The first term in (2.5) corresponds to the constant background-the spectral level of the shot noise—in the intensity fluctuation spectrum (in reality, this background is limited along the frequency scale by the inertia of the measurement apparatus). In a study of the radiation from a polyatomic system the second term in (2.5) contains a term $\langle J(t_1)\rangle\times\langle J(t_2)\rangle$ [$\langle E_\mu^{(-)}(x_1)E_\mu^{(+)}(x_1)\rangle\langle E_\nu^{(-)}(x_2)\times E_\nu^{(+)}(x_2)\rangle$ in the integral]. At any rate, it is useful to single out this "corpuscular" component (photons which are each definitely emitted by one of the elementary radiators; Subsection 3.3) even if it does not arise by itself. The remaining (cumulant) part of the correlation function reflects the statistical dependence of the photon counts.

2.2. Characteristics of photon antibunching and sub-Poisson photon statistics

The experimental criterion for photon antibunching is the inequality

$$K_2(t_1, t_2) < \{\langle J(t_1)\rangle\,\langle J(t_2)\rangle\} \tag{2.7}$$

at small values of $\tau = t_2 - t_1$; the more restricted condition for antibunching is the equality $K_2(t,t) = 0$. We again note that the first term in (2.5) bears no relation to photon correlations, but it does actually contribute to the correlation function of the photocurrent when two time intervals of the photodetector response overlap. At small values of τ this component is unavoidable, and it must be eliminated in the course of the statistical analysis.

To calculate the probability $p(n;T)$, for the detection of n photon counts over the observation time T, we need to determine the field correlation functions of all orders[4] (see also Subsection 3.2). Let us examine the variance of the number of photon counts in the steady state:

$$D[n,\ T] = \langle n^2\rangle_T - \langle n\rangle_T^2 = \int_0^T dt_1 \int_0^T dt_2\, K(t_1,\ t_2) - \langle J\rangle^2 T^2$$

$$= 2\int_0^T d\tau\,(T-\tau)\,[K(\tau) - \langle J\rangle^2]. \tag{2.8}$$

Here we have used $K(t_1,t_2) = K(\tau)$. It is useful to single out in $D[n,T]$ the universal Poisson term $\langle n\rangle_T$, which stems from the first term in (2.5) [the one with $\delta(\tau)$]:

$$D\,[n;\ T] = \langle n\rangle_T\{1 + \xi\,(T)\}. \tag{2.9}$$

The parameter $\xi(T)$, which is a measure of the deviation from the variance for a Poisson distribution, is

$$\xi\,(T) = \frac{2}{\langle J\rangle\, T}\int_0^T d\tau\,(T-\tau)\,[K_2(\tau) - \langle J\rangle^2]. \tag{2.10}$$

In the case $\xi(T)$ we shall call $\xi(T)$ the "SPPS parameter." We wish to emphasize that according to the precise meaning of the terms which we are using we should be speaking not about photon statistics "in general" but about the distribution of photon counts under definite measurement conditions, over a definite time interval T. We will use the notation $\xi_\infty = \xi(T)$ for $T \gg \tau_{\text{corr}}$ where τ_{corr} is the time scale of the variations in $K_2(\tau)$ [more precisely, the time scale of the irreversible decay of the correlations—for the vanishing of the integrand in (2.10)]. If $\xi_\infty < 0$, we will speak in terms of SPPS (without any special explanation).

The conversion of a flux of photons into photoelectrons which is performed by an ideal photodetector (which has no "dead time" or false counts) is a binomial conversion. In this case we would have $\bar{n}_T = q\bar{m}_T$ and $\xi^{(n)}(T) = q\tilde{\xi}^{(m)}(T)$ for any probability distribution $p(m,T)$, where m is the number of photons, n is the number of photoelectron counts, and q is the quantum yield. The Poisson distribution is preserved (with the replacement $\bar{m} \to \bar{n}$). It is useful to single out the parameter ξ_∞ for the following reason. As was mentionted in the Introduction, photon and antibunching leads to characteristic dips in the intensity fluctuation spectrum below the level of the shot noise. There is also a spectral expression of SPPS. At $T \gg \tau_{\text{corr}}$ we can write

$$\xi\,(T) \approx \xi_\infty = \frac{2}{\langle J\rangle}\int_0^\infty d\tau\,(K_2(\tau) - \langle J\rangle^2). \tag{2.11}$$

It is not difficult to see from expression (2.5) that $K_2(\tau)$ is an even function. The doubled integral in (2.11) is therefore the value of Fourier transform (2.6) at the point $\omega = 0$, after the spectral density of the shot noise, $\langle J\rangle$, and the part of the correlation function factorized in terms of the photocurrent, equal to $\langle J\rangle^2\delta(\omega)$, have been subtracted:

$$\xi_\infty = \frac{1}{\langle J\rangle}\,(G(\omega) - \langle J\rangle^2\,\delta(\omega) - \langle J\rangle)_{\omega=0}. \tag{2.12}$$

The parameter $|\xi_\infty|$ is therefore equal to the relative increase (in the case $\xi_\infty > 0$) or decrease ($\xi_\infty < 0$) in the photocurrent noise spectrum $G(\omega)$ in comparison with level of the spectral density of the shot noise at $\omega \approx 0$. This manifestation of SPPS after a long time may be important for extremely precise measurements by methods of correlation spectroscopy. The dips in the intensity fluctuation spectrum do not necessarily appear in the region $\omega \approx 0$; just where they appear depends on the dynamics of the system and the particular measurement version (Secs. 3, 4).

In general, different characteristics of radiation incorporated in $K_2(\tau)$ may prove useful for applications. For data trasmission in analog form the suppression of the shot noise in the frequency band of the signal modulation is important. In other words, we need to know the details of the shape of the dip in the intensity fluctuation spectrum. In digital transmission the reliability improves with decreasing "miss" probability $p(0,T)$, where T is the duration of a single packet.[75] For SPPS, $p(0,T)$ is smaller than for a Poisson distribution (for equal values of $\bar{n}$). In determining the instantaneous value of the signal-to-noise ratio one needs to know $K_2(\tau)$ at small values of τ.

Let us examine the relation between photon antibunching and sub-Poisson photon statistics.[76] We introduce the normalized correlation function (with a normal ordering of field operators)

$$\gamma^{(2)}(\tau) = \frac{K_2(\tau)}{\langle J\rangle^2} = \frac{\langle :J\,(0)\,J\,(\tau):\rangle}{\langle J\rangle^2}\,. \tag{2.13}$$

The following assertions are obvious from the dependence of the sign of the integral in (2.10) on the properties of the integrand:

1) It follows from the condition for SPPS for a given T, $\xi(T) < 0$, only that there is a time interval τ, $\tau \leqslant T$, in which the relation $\gamma^{(2)}(\tau) < 1$ holds [the condition $\gamma^{(2)}(0) = 0$ does not necessarily hold]. The converse of course is not correct.

2) If $\gamma^{(2)}(\tau) < 1$ at $\tau < T_1$, then for measurement intervals $T \leqslant T_1$ SPPS holds: $\xi(T) < 0$.

3) In particular, it follows from the antibunching in the restricted sense of the term, $\gamma^{(2)}(0) = 0$, by virtue of the continuity of $\gamma^{(2)}(\tau)$, that there exist intervals of T(which may be small) in which the relation $\xi(T) < 0$ holds (SPPS).

4) It follows from assertions 1) and 2) that photon antibunching and SPPS are equivalent if $\gamma^{(2)}(\tau)$ is a monotonic function and if $\gamma^{(2)}(\infty) = 1$ (and only under these conditions).

Consequently, photon antibunching and SPPS are generally not equivalent, and neither characteristic follows from the other. This assertion remains correct with regard to three characteristics, i.e., when we incorporate the "squeezed" nature of the field state.

The antibunching effect, i.e., the dip as $\tau \to 0$ on the curve of the photon-pair count rate (Fig. 1), can be important only for weak sources: $\langle I\rangle\tau_{\text{corr}} \lesssim 1$. We wish to stress that for a macroscopic source ($\langle I\rangle\tau_{\text{corr}} \gg 1$) the manifestations of SPPS may be significant, even though the photon antibunching is slight: There can be dips in the intensity fluctuation spectrum, which can in principle go to zero $\xi(T) \to -1$.

2.3 Squeezed field states and SPPS (examples)

Let us examine an example which demonstrates the relationship between squeezing and SPPS in a steady-state experiment.[64,77,78] We assume that we are detecting photon counts in a field consisting of a superposition of an intense field in a coherent state with a complex amplitude $\mathscr{E} = \mathscr{E}_0 e^{i\theta}$ and the radiation of interest, which is characterized by operators $E^{\pm}$. For sufficiently short observation time intervals T, and in the zeroth order approximation in the intensity ratio $\langle E^{(-)}E^{(+)}\rangle/\mathscr{E}_0^2$, the parameter $\xi(T)$ [Eqs. (2.9) and (2.10)] can be expressed in terms of the variances of the quadrature field components[64] $X_1 = (1/2)(E^{(+)} + E^{(-)})$, $X_2 = (i/2)(E^{(-)} - E^{(+)})$:

$$\xi(T) = 4\eta T\langle :(\Delta X_i)^2:\rangle,$$
$$i=1, \quad \text{if} \quad \theta=0, \text{ or } i=2, \quad \text{if} \quad \theta=\frac{\pi}{2}. \tag{2.14}$$

Here η is a dimensional constant which characterizes the photodetection efficiency. The condition for SPPS is related to the squeezing property: $\langle :(\Delta X_i)^2:\rangle < 0$. For these particular values of θ, these characteristics are equivalent. We recall that for a field in a coherent state we have $\langle (:\Delta X_i)^2:\rangle = 0$. The frequency-conversion method thus makes it possible in principle to establish the presence of a squeezed state in terms of the distribution of photon counts. We note that the phase of the reference wave, θ, must be determined with respect to the phase of the signal under study (Secs. 3, 4).

For long observation times, the parameter ξ and the photocurrent spectrum are expressed in terms of the correlation functions of the quadrature components. The observable effects also depend on the phase relations.

In Sec. 4 we will examine the conditions under which the variance of the number of photons of the resultant signal from a degenerate parametric amplifier can be written in the form

$$\langle(\Delta m)^2\rangle_t = 4\,\langle m\rangle_t\,\langle(\Delta X_i)^2\rangle_t, \quad i=1 \quad \text{or} \quad i=2, \tag{2.15}$$

where t is the signal formation time in the nonlinear medium. Using the identity

$$\langle(\Delta X_i)^2\rangle = \frac{1}{4} + \langle:(\Delta X_i)^2:\rangle, \quad i=1,\ 2, \tag{2.16}$$

we find the parameter $\tilde{\xi}_t$ (for the number of signal photons at the output from the amplifier):

$$\tilde{\xi}_t = 4\langle:(\Delta X_i)^2:\rangle, \quad i=1 \quad \text{or} \quad i=2. \tag{2.17}$$

In this case the conditions for SPPS and for squeezing are thus the same.

3. NONLINEAR RESONANCE FLUORESCENCE OF NONINTERACTING ATOMS; PHOTON ANTIBUNCHING AND SUB-POISSON PHOTON STATISTICS

Resonance fluorescence was one of the fundamental problems during the development of the quantum theory of radiation.[80,81] The advent of lasers was followed by the derivation of a theory for the nonlinear resonance response of matter to intense light, including a theory for nonlinear resonance fluorescence (see Ref. 82 for a detailed account and a bibliography). The predictions of the theory of nonlinear resonance fluorescence[83–86] (in particular, the appearance of an noncoherent background near the coherent-scattering line and—the most vivid effect—a triplet induced by an intense field) were confirmed quantitatively in the experiments of Refs. 87–89 (see also the literature cited in Refs. 82 and 89). Nonlinear resonance fluorescence also turned out to be the first type of radiation in which the new statistical effects in which we are interested here were observed.

The balance of this section of the paper is based on a calculation of the field correlation functions by a diagram technique in time-dependent perturbation theory for a density matrix.[90] This version of the diagram technique has proved to be very convenient in radiation theory when several interactions are taken into account, both coherent interactions and those which result in relaxation processes.[91] Instead of the corresponding diagrams, however, we will be using figures here which are similar to Young's experimental layout to illustrate the physical meaning of the various components of the correlation functions.

3.1. Group decomposition of correlation functions

We consider a system of N immobile, noninteracting, identical atoms in a region with a length scale L. The atoms are in a monochromatic plane light wave with an electric field

$$\vec{\mathscr{E}}(t,\ \mathbf{r}) = \frac{1}{2}\vec{\mathscr{E}}_0 \exp(-i\omega_0 t + i\mathbf{k}_0\mathbf{r}) + \text{c.c.} \tag{3.1}$$

The frequency ω_0 is close to the atomic transition frequency ω_{21} (level E_1 is the ground level; the levels are assumed to be nondegenerate). Secondary radiation is detected at a distance $R \gg L$. The difference between the light propagation times from different atoms to the surface of the photocathode (or of two photocathodes) is assumed to be much shorter than the important photocurrent fluctuation times which are determined by the evolution of the atom in the field:

$$\frac{L}{c} \ll \min\{\gamma^{-1},\ |\nu_0|^{-1},\ V_0^{-1}\}, \tag{3.2}$$

where γ is the radiative width of the upper level, $\nu_0 = \omega_0 - \omega_{21}$ is the deviation from the resonant frequency, $2V_0 = |\mathbf{d}_{21}\vec{\mathscr{E}}_0|/\hbar$ is the Rabi frequency, and $\mathbf{d}_{21}$ is the transition dipole moment. Under condition (3.2), the temporal retardation does not influence the photocurrent fluctuations and can be ignored.

Before we examine the field correlation functions in the integrands in (2.5), we would like to clarify a manifestation of the superposition principle for the field in the example of the expectation value $\langle \mathbf{E}^{(+)}(t,\mathbf{r})\rangle$ (in optics, this expectation value usually vanishes or is essentially the same as the field of the transmitted radiation from the external source; however, it is not devoid of meaning; Subsection 3.3). Using the expression

$$\mathbf{E}^{(+)}(\mathbf{r}) = i\sum_{\mathbf{k},\,\alpha=1,2}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\mathbf{e}_\alpha(\mathbf{k})\,e^{i\mathbf{k}\mathbf{r}}\,a_{\mathbf{k},\alpha} \tag{3.3}$$

for the field operator in the Schrödinger picture, we write the expectation value of a Heisenberg operator in the form

$$\langle \mathbf{E}^{(+)}(t,\ \mathbf{r})\rangle = \mathrm{Sp}\,[S^{-1}(t,\ t_0)\,\mathbf{E}^{(+)}(\mathbf{r})\,S(t,\ t_0)\,\sigma(t_0)]$$
$$= i\sum_{\mathbf{k},\alpha}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\mathbf{e}_\alpha(\mathbf{k})\exp(i\mathbf{k}\mathbf{r})$$
$$\times\,\mathrm{Sp}\,[S^{-1}(t,\ t_0)\,a_{\mathbf{k},\alpha}\,S(t,\ t_0)\,\sigma(t_0)]; \tag{3.4}$$

here $\omega = c|\mathbf{k}|$, $\mathbf{e}_\alpha(\mathbf{k})$ are unit polarization vectors ($\alpha = 1,2$); V is the nominal quantization volume; $S(t,t_0)$ is the operator representing the evolution of the system consisting of [atoms in an external coherent (classical) field] + (quantized field); and $\sigma(t_0)$ is the statistical operator of this system at the initial time.

In nonresonator problems a quantized electromagnetic field in an unbounded volume can be thought of as a reservoir at zero temperature with respect to the dynamic (atomic) system (despite the arbitrarily prolonged conversion of the light incident on the atoms into secondary radiation). We can thus write $\sigma(t) = \rho(t)|\Phi_0\rangle\langle\Phi_0|$, where the operator $|\Phi_0\rangle\langle\Phi_0|$ is the projection operator onto the ground state of the continuum of radiative oscillators (vacuum), and $\rho(t)$ is the statistical operator of the system of atoms. For the last factor in (3.4) we find

$$\mathrm{Sp}[\ldots] = \mathrm{Sp}_{(\mathrm{at})}\langle\Phi_0|S^{-1}(t,\ t_0)\,a_{\mathbf{k},\alpha}\,S(t,\ t_0)\,\rho(t_0)|\Phi_0\rangle, \tag{3.5}$$

where the trace (spur) is now taken over only the states of the atoms, and $\rho(t_0) = \prod_a (|1\rangle_{aa}\langle 1|)_a$ is the initial statistical operator of the atoms (for the steady-state response, the dependence on the initial state of the atoms essentially disappears after the limit $t\to\infty$ or $t_0\to-\infty$ is taken). The operator $a_{\mathbf{k},\alpha}$ in (3.5) must be paired with the creation operator for a photon of the same type,[92,93] $a^+_{\mathbf{k},\alpha}$. The operator $a^+_{\mathbf{k},\alpha}$ must be "taken" from the operator $S(t,t_0)$ in each order of perturbation theory. After this is done, however, the effect of the "newly assembled" operator $S(t,t_0)$ (more precisely, the evolution of the statistical operator of the atoms) does not change: The radiative oscillator of the type $(\mathbf{k},\alpha)$ in each term of the type in (3.5) in sum (3.4) is one of a continuum (after the limit $V\to\infty$ is taken). The energy operator of the free field contains $a^+_{\mathbf{k},\alpha}$ only in the product $a_{\mathbf{k},\alpha}$, and the operator representing the interaction of the field and the atoms, $H_I = -\Sigma_a \mathbf{d}^{(a)}\mathbf{E}(\mathbf{r}_a)$, is linear in $a^+_{\mathbf{k},\alpha}$. It is thus possible to make the (extremely useful!) assumption that $a_{\mathbf{k},\alpha}$ is paired with $a^+_{\mathbf{k},\alpha}$ from each $H_I^{(a)}$, while the summation over all the atoms, $\Sigma_a\cdots$, is preserved. The superposition principle begins to "work" in this step of the calculations.

We skip over the calculations to the result (which is correct in the wave zone):

$$\langle E^{(+)}\ (t,\ r)\rangle = i\sum_a \frac{k_0^2}{R_a}[\mathbf{n}_a[\mathbf{d}_{12}^{(a)}\mathbf{n}_a]]\,\bar{\rho}_{21}^{(a)} \times\exp(-i\omega_0 t + ik_0R_a + i\mathbf{k}_0\mathbf{r}_a), \tag{3.6}$$

where $\mathbf{r}_a$ are the radius vectors of the atoms, $\mathbf{R}_a = \mathbf{r}-\mathbf{r}_a$, $\mathbf{n}_a = R_a/R_a$, and $\bar{\rho}_{21}^{(a)}$ is the steady-state value of a nondiagonal element of the density matrix of atom a [a factor $\exp(-i\omega_0 t + i\,\mathbf{k}_0\mathbf{r}_a)$ has been singled out]. The quantity $\mathbf{d}_{12}\bar{\rho}_{21}$ is the positive-frequency complex amplitude of the steady-state expectation value of the dipole moment of the atom, so (3.6) agrees with the expression for the dipole radiation field in classical electrodynamics[94] [in this case, (3.6) is essentially the field of a coherently scattered wave]. The summation over the atoms and the average over their random positions make the quantity in (3.6) vanishingly small at all observation points $\mathbf{r}$ except those in a diffraction cone along the direction of the transmitted laser light, with a solid angle of the order of $(\lambda_0/L)^2$.

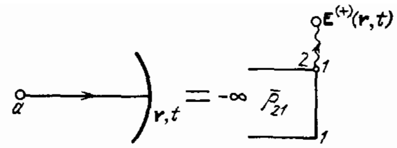

FIG. 3. Diagram illustrating the calculation of the expectation value of the positive-frequency part of the electric field of the radiation from an atom.

When this procedure is used in problems without coherent external radiation (spontaneous emission,[72,95] resonance transfer of excitation, and the formation of cooperative coherent emission[96,97]) the "classical" expressions are found for the probability amplitudes for radiative transitions (the average field vanishes in these cases).

We associate the quantity in (3.6) with the diagram (as an exception: "genuine") and the drawing in Fig. 3.

At this point we go back to photocurrent correlation function (2.5). The correlation function in the first term in (2.5) reduces to two diagrams (Fig. 4). The correlation function in which we are interested, in the second term, splits up into the sum of a rather large number of diagrams (Fig. 5). The nonshot part $[K_2(\tau)]$ of the photocurrent correlation function $K(\tau)$ is therefore represented as the sum of "two-photon" contributions of the secondary radiation of individual atoms and groups of two, three, and four atoms.

The circles labeled with letters on the diagrams represent atoms. A summation is understood to be carried out over the corresponding arrangements (over all possible combinations of the N atoms, taken 1, 2, 3, and 4 at a time; and over all permutations of the indices of the atoms within each combination). The average is taken over the positions of the atoms. The arcs are regions of the photocathode; 1 represents $(t,\mathbf{r}_1)$, and 2 represents $(t+\tau,\mathbf{r}_2)$. One might also think in terms of an experiment with two photodetectors in a delayed-coincidence arrangement. The lines running from atoms to points on the photocathode are the photon propagators "from creation to annihilation." The lines with backward arrowheads are complex-conjugate propagators.

The use of diagrams with definite rules governing the correspondence between their elements and the analytic expressions makes it possible to link each component of the correlation function of interest with elements of the density matrix of the system of atoms. Clearly, a similar group decomposition holds for correlation functions of all orders.

We believe that Figs. 3–5 explain the manifestation of the superposition principle for a quantum-field analysis of optical measurements. In any calculation of a measured sig-

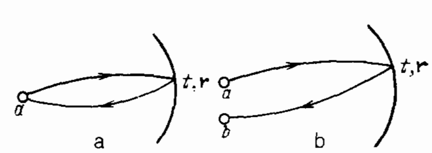

FIG. 4. Diagrams for the expectation value of the radiation intensity. a—Monatomic component; b—diatomic (interference) component of the intensity.

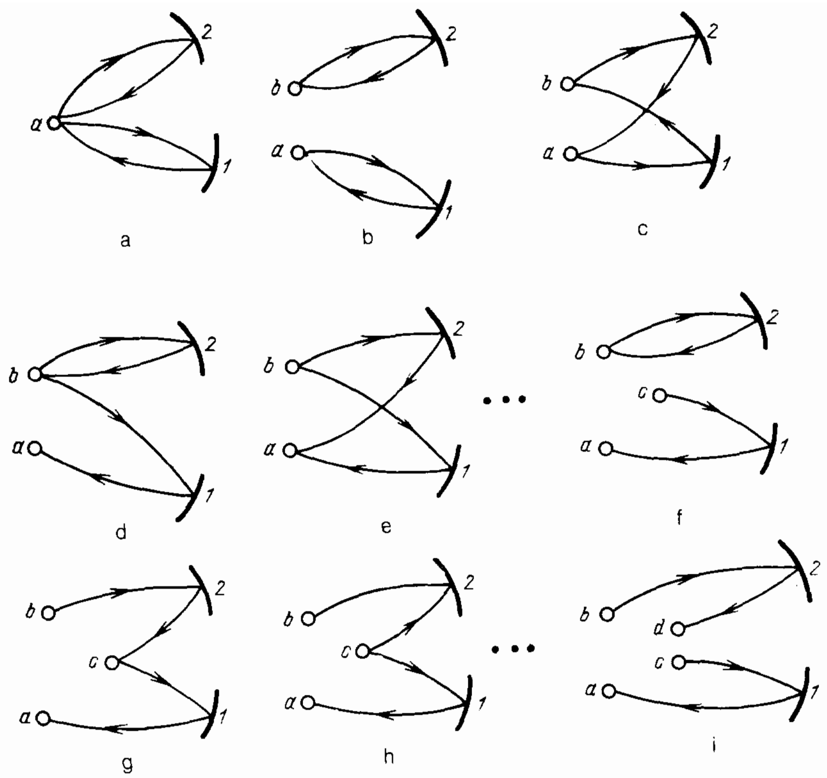

FIG. 5. Diagrams which form the fourth-order field correlation function of the radiation of a polyatomic system [expression (2.5); the second term is $K_2(\tau)$]. See Subsection 3.3 for a detailed analysis.

nal we are dealing with expectation values of Heisenberg field operators. The interaction of the field with the source links the photon propagators to the elementary radiators in a natural way. The interference laws for classical waves are reproduced at the level of such "directed" photon propagators or probability amplitudes. However, only events involving the detection of an increment in the energy (or of the momentum transfer or of the angular momentum) are elementary physical events (which are conveniently interpreted from the particle standpoint). It is to these events that we can directly apply the customary concepts of probability theory and the theory of random processes.[6)]

3.2. Photon antibunching in the fluorescence of a single atom

We have arrived at the first experimental study of photon antibunching.[12,18] Photon antibunching is an organic part of the nonlinear resonance fluorescence of a single atom. The diagram in Fig. 5a corresponds to the next component, $K_2^{(1)}(\tau)$ of the informative part, $K_2(\tau)$ of the photocurrent correlation function[12–15] [Eq. (2.5)]:

$$K_2^{(1)}(\tau) = N(q\alpha\gamma)^2 \bar{\rho}_{22}\rho_{22}^{(11)}(\tau), \tag{3.7}$$

where the coefficient α depends on the experimental geometry, $\bar{\rho}_{22}$ is the steady-state value of the probability for finding the atom in the upper level, and the superscripts in $\rho_{22}^{(11)}(\tau)$ specifiy the initial conditions $\rho_{ik}^{(mn)}(0) = \delta_{im}\delta_{kn}$ [δ_{pr} is the Kronecker delta)]. The equalities

$$\rho_{22}^{(11)}(0) = 0, \quad \rho_{22}^{(11)}(\infty) = \bar{\rho}_{22} \tag{3.8}$$

mean that there is an antibunching of photons emitted in succession by the same atom. At $N \gg 1$, the "single-atom" component of (3.7) is usually (but not always—see the discussion below) suppressed by the "polyatomic" components of a Gaussian nature. At $N = 1$, on the other hand, it is the sole component. In connection with expression (3.7) it is pertinent to note that a study of steady-state correlations can reveal the time-dependent behavior (dynamics) of the radiating system under specified external conditions (although this comment would be trivial from the standpoint of random processes, studies of this sort would not be trivial in the optical region and with an individual atom or group of atoms as the source of correlations in the radiation). In this case the photon antibunching associated with expression (3.7) reveals the process by which the upper level of the atom is populated. The detection of a photon at time t reliably fixes the atom (which is not subject to a destructive effect of the measuring instrument!) in the ground state. The probability (per unit time) for detecting a second photon at the time $t + \tau$ is proportional to $\gamma\rho_{22}^{(11)}(\tau)$. The quantity $\rho_{22}^{(11)}(\tau)$ serves as a conditional probability.

Kimble *et al.*[12], and Dagenais and Mandel[18] detected emission (nonlinear resonance fluorescence) of sodium atoms from a low-density beam along a direction approximately perpendicular to the beam. The exciting laser beam intersected the atomic beam at strictly right angles, minimizing the Doppler effect during the excitation. The laser light acting on the atoms was as resonance with the transition $(3^2S_{1/2}, F=2) - (3^2P_{3/2}, F=3)$. The intensity of this light was carefully stabilized (to within a few percent), as was its frequency (within 1–2 MHz). Two states, $(3^2S_{1/2}, F=2, m_F=2)$ and $(3^2P_{3/2}, F=3, m_F=2)$, were singled out with the help of a circularly polarized "preparatory" beam from the same laser, in the approach which was taken in Refs. 87 and 88 and in some earlier studies carried out to observe the spectrum of nonlinear resonance fluorescence (see Refs. 88 and 18 regarding the details of the method). The photons of the nonlinear resonance fluorescence were collected by a microscope objective. The light was then split into two beams, of approximately equal intensity, and delayed coincidences were counted (the distribution in τ of the number of photon-count pairs). Kimble *et al.*[12] estimated that in their first experiments there were simultaneously one or two atoms (almost certainly no more) in the observation region. In Ref. 17 Kimble *et al.* reported the average number of atoms to be 1/3 (within an uncertainty of 50%). The effect of fluctuations in the number of atoms (a ruinous effect for photon antibunching in this case) has been studied in several places.[16–18,73,101] A very careful analysis of the experiments carried out under various conditions, in particular, at various densities of the atomic beams[17,18] inspires confidence in the results.

Simple expressions for $\rho_{22}^{(11)}(\tau)$ can be written only for limiting relations among the parameters V_0, γ, $|\nu_0|$:

a) weak field, $V_0^2 \ll \max\{\gamma^2, \nu_0^2\}$,

$$\rho_{22}^{(11)}(\tau) \approx \frac{V_0^2}{(\gamma^2/4)+\nu_0^2}\Big[1+\exp(-\gamma\tau) - 2\exp\left(-\frac{\gamma\tau}{2}\right)\cos(\nu_0\tau)\Big]; \qquad (3.9)$$

b) strong field, $V_0^2 \gg \gamma^2, \nu_0^2$,

$$\rho_{22}^{(11)} \approx \frac{1}{2}\Big[1-\exp\left(-\frac{3\gamma\tau}{4}\right)\cos(2V_0\tau)\Big]. \qquad (3.10)$$

Figure 6 demonstrates the good agreement between the corrected experimental curves and the smooth functions $\rho_{22}^{(11)}(\tau)$ (Ref. 18). The corrections dealt with transit-time effects and correlations in the scattering from pairs of atoms and were based on theoretical expressions.

A theoretical study was made in Refs. 102–104 of the effect of a frequency spread, amplitude fluctuations, and phase fluctuations of the exciting laser light on the intensity correlations in nonlinear resonance fluorescence.

Figure 7 demonstrates the properties of the photocurrent noise spectrum [expression (2.6)] when the single-atom component is singled out: Against the background of the shot-noise spectrum $G_{\text{shot}}(\omega) = \langle J\rangle$ there are dips corresponding to antibunching.[15] The spectrum is less sensitive to fluctuations in the number of particles in the observation

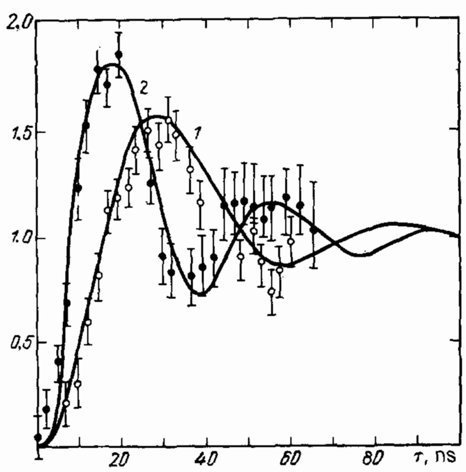

FIG. 6. Comparison of theoretical (solid lines) and corrected experimental results on the correlation function $\rho_{22}^{(11)}(\tau)\sqrt{\rho_{22}}$ (from Ref. 18). 1—$V_0 = 0.7\gamma$, $\nu_0 = -1.1$; 2—$V_0 = 1.1\gamma$, $\nu_0 = -1.7$.

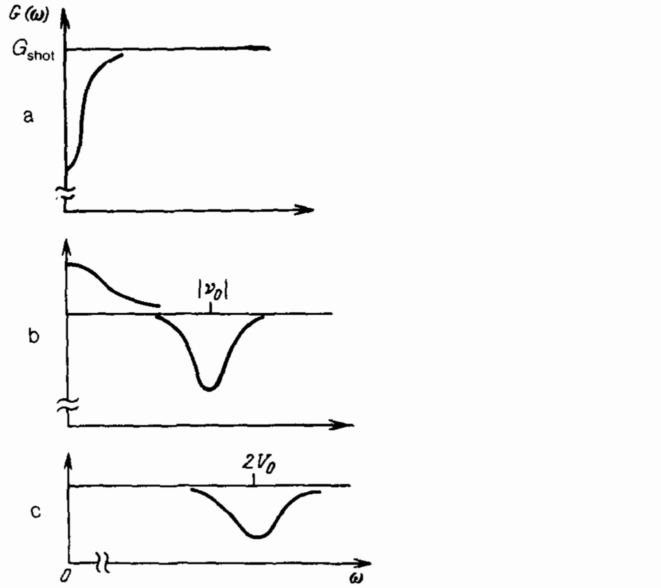

FIG. 7. Examples of intensity fluctuation spectra of the nonlinear resonant fluorescence of a single atom. a, b—weak field, $V_0^2 \ll (\gamma^2/4) + \nu_0^2$; a—$\gamma_0 = 0$; b—$\nu_0 = 3\gamma$; c—strong field, $V_0^2 \gg \gamma^2, \gamma_0^2$ ($q\alpha = 1$).

region (and less sensitive to the interaction of these particles with the exciting laser beam) than is the photon-count correlation function itself. Under the condition $\gamma\tau_{\text{tr}} \gg 1$ (τ_{tr} is the transit time), and with a large collection angle (Subsection 3.3), a peak with a width of the order of $\tau_{\text{tr}}^{-1} \ll \gamma$ appears in the region $\omega \approx 0$ in addition to $\langle J\rangle^2\delta(\omega)$ (from pairs of independently radiating atoms; Fig. 5b). The characteristic dip persists and can be seen even at[15,22] $N \gg 1$.

Short and Mandel[19,20] have reported establishing that the distribution of photon counts is of a sub-Poisson nature under the same conditions in the observation of the nonlinear resonance fluorescence of a single atom. The flux density in the atomic beam corresponded to an average distance of the order of 1 cm between atoms and to a time interval of about 10 μs, with a 200-ns duration of the count of photon pulses in a single "n" realization. Special measures were taken to monitor the "arrival" of an atom in the observation region. The statistics of the photon count were determined from the results of 24 725 000 realizations, in each of which from 0 to 3 photon counts were actually detected. The parameter $\xi(T)$ [see (2.9)] was found from these measurements to be $(-1.48 \pm 0.25)\cdot 10^{-3}$, and the value found after corrections was $(-2.20 \pm 0.23)\cdot 10^{-3}$. The corrections were made for the dark (background) counts, the dead time, false photon counts, and the possibility that two atoms were present in the observation region. Short and Mandel,[19,20] estimate the theoretical value of $\xi(T)$ [see expression (3.12) below] to be $\xi(T) \approx -(2.4 \pm 0.6)\cdot 10^{-3}$.

Using expressions (2.10) and (3.7), we find the following expression for the parameter $\xi(T)$ in the observation of the nonlinear resonance fluorescence of a single atom:

$$\xi(T) = q\alpha\gamma\cdot\frac{2}{T}\int_0^T d\tau\,(T-\tau)\,(\rho_{22}^{(11)}(\tau) - \bar{\rho}_{22}). \qquad (3.11)$$

Since we have $\rho_{22}^{(11)}(0) = 0$, there will always be SPPS if T is sufficiently small, $T \ll \min\{\gamma^{-1}, |\nu_0|^{-1}, V_0^{-1}\}$:

$$\xi(T) \approx -q\alpha\gamma T\bar{\rho}_{22}; \qquad (3.12)$$

in the saturation regime, with $V_0^2 \gg \gamma^2$, ν_0^2, at $T \ll V_0^{-1}$, we would have

$$\xi(T) \approx -\frac{1}{2} q\alpha\gamma T. \qquad (3.13)$$

At small values of T the relation $|\xi| \ll 1$ always holds.

For long observation time intervals, $T \gg \gamma^{-1}$, one will not always observe SPPS[21-23]:

$$\xi(T) \approx \xi_\infty = q\alpha \frac{2V_0^2[\nu_0^2-(3\gamma^2/4)]}{[(\gamma^2/4)+\nu_0^2+2V_0^2]^2}. \qquad (3.14)$$

The condition for SPPS is $\nu_0^2 < 3\gamma^2/4$, $\xi_\infty^{\min} = -3q\alpha/4$ (and is reached at $V_0^2 = \gamma^2/8$, $\nu_0 = 0$).

For a brief discussion of the photon-count distribution $p(n;T)$, we follow Ref. 25 (see also Refs. 24, 26, and 27). In order to calculate $p(n;T)$ we need to determine the field correlation functions of all orders: They contribute factorial moments from which the distribution is reconstructed.[4]

The situation is simplified in the case of the nonlinear resonance fluorescence of a single atom by the circumstance that each factorial moment $Q_m(T)$ is expressed as the $(m-1)$-fold convolution of the function $\rho_{22}^{(11)}(t_k - t_{k-1})$ $(k = 2,3,...,m)$ with a factor $(q\alpha\gamma)^m\bar{\rho}_{22}$. The Laplace transforms of the functions $Q_m(T)$ in the variable T, i.e., $\tilde{Q}_m(s)$, can thus be expressed in a fairly simple way in terms of the Laplace transform of the population of the upper level, $\tilde{\rho}_{22}^{(11)}(s)$. Taking the inverse Laplace transforms of course generally leads only to some extremely complicated expressions. However, it is entirely possible to study limiting cases and to carry out numerical calculations for small values of n. Figure 8 illustrates the narrowing of the $p(n;T)$ distribution in comparison with the corresponding Poisson distribution. For a long observation time T, under the condition $\langle n\rangle = q\alpha\gamma T\bar{\rho}_{22} \gg 1$, the central region of the distribution, $|n - \langle n\rangle| \ll \langle n\rangle$, can be approximated well by a Gaussian distribution with a variance $D[n] = \langle n\rangle \times (1 + \xi_\infty)$, where ξ_∞ is given by expression (3.14). This result agrees with the central limit theorem: At $T \gg \tau_{\text{corr}}$, a sample of duration T contains many uncorrelated regions of a random process.[7]

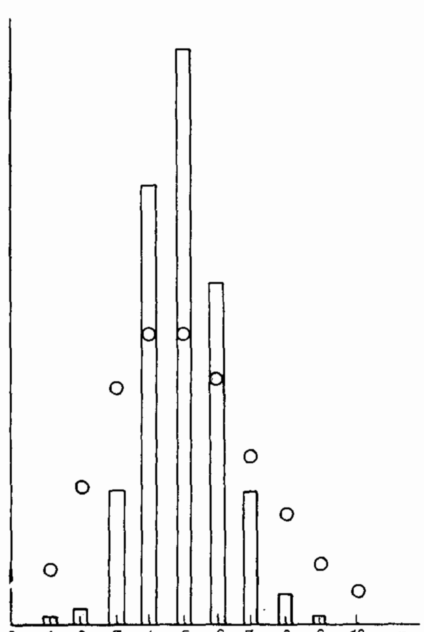

FIG. 8. Photon-count distribution in the detection of the nonlinear resonant fluorescence of a single atom (columns). $\bar{n} = 5$, $\nu_0 = 0$, $V_0^2 = \gamma^2/8$. The circles show a Poisson distribution with $\bar{n} = 5$.

In Refs. 23, 24, and 27 the distribution of the number of nonlinear-resonance-fluorescence photons was related to the statistics of momentum transfer to atoms from a laser wave. The problem is analyzed most comprehensively, and the literature reviewed, in Ref. 27.

3.3. Polyatomic effects in the nonlinear resonance fluorescence of a system of atoms

A classification of the contributions to the correlation function $K(\tau)$ from groups of two, three, and four atoms in the observation of the nonlinear resonance fluorescence of a system of atoms was given in Ref. 15. Despite the large number of interference processes which lead to intensity fluctuations, and despite the circumstance that most of the corresponding terms are small because of an averaging over the positions of atoms,[17] some of them may be distinguished by virtue of the characteristic correlations in terms of the propagation directions of the photons. Certain processes lead to a bunching of photons, while others promote photon antibunching and SPPS.[79,105,106] Under ordinary observation conditions, with $N \gg 1$, the direction $\mathbf{n}_0$ of the directly transmitted light of the exciting laser is eliminated from the photon collection angle. The dominant processes are then those depicted in Fig. 5b and c. Their sum corresponds to Gaussian field statistics and to a variance $D[n] = \bar{n}(1 + \delta')$ in the number of photon counts (see the Introduction). The diagram in Fig. 5b corresponds to the component of the correlation function which we have already considered back in Subsection 2.1. This is a trivial component in the absence of a statistical dependence of the excitation events and if the evolution of the excited atoms is independent (the opposite situation is dealt with comprehensively in Secs. 5 and 6). The diagram in Fig. 5c corresponds to a partial factorization of the fourth-order field correlation function [expression (2.5)] in the form $\langle E_\alpha^{(-)}(1)\, E_\beta^{(+)}(2)\rangle \langle E_\beta^{(-)}(2) \times E_\alpha^{(+)}(1)$. At the semiclassical level, this would be wave noise: beats of the spectral components of the radiation. In the intensity fluctuation spectrum, it is a convolution of the ordinary optical spectrum of the nonlinear resonance fluorescence. The pairs of photons which actually determine this part of the correlation function are strongly correlated in terms of propagation direction in a solid angle of the order of $(\lambda/L)^2$, where L is the size of the scattering region. On the whole, there is no sharp directionality in terms of the pair detection directions. Angular correlations lead to a decrease of this "two-atom" wave noise in comparison with the direct process (the diagram in Fig. 5b) in a ratio σ_c/S at $S > \sigma_c$, where σ_c is the area of the spatial coherence at the surface of the photocathode, and S is the area of the photocathode. The contribution of the wave noise can therefore be suppressed if observations are taken over a wide solid angle.[54] In the intensity fluctuation spectrum, it is also lowered, by a ratio $\gamma/\delta\omega_D$, by virtue of the Doppler effect; in measurements of the intensity fluctuation spectrum it is thus possible to distinguish a "single-atom" signal with photon antibunching (dips against a background of shot noise and wave noise), also in the case of the nonlinear resonance fluorescence of a polyatomic system.

All the other processes (all the diagrams from Fig. 5d on) are characterized by a sharp directionality or, more precisely, a selectivity in terms of the regions on the photocathode or the arrangement of two photodetectors (in an experiment on mutual correlations). For example, the diagram in Fig. 5d corresponds to a factorization $\langle E_\alpha^{(-)}(1)\rangle \langle E_\beta^{(-)}(2) E_\beta^{(+)}(2) E_\alpha^{(+)}(1)\rangle$. This diagram is the dominant one in an experiment with two photodetectors, one of which detects the radiation in a small solid angle in the direction $\mathbf{n}_0$, while the other detects the radiation in a wide solid angle, in directions quite different from $\mathbf{n}_0$ ("sideways-forward" photon correlations). The diagram in Fig. 5e (note the directions of the arrowheads) leads to so-called anomalous correlation functions [the partial factorization $\langle E_\alpha^{(-)}(1) E_\beta^{(-)}(2)\rangle \langle E_\beta^{(+)}(2) E_\alpha^{(+)}(1)\rangle$]. A method for identifying this signal in a mutual-correlation experiment was proposed in Ref. 106.

In an analysis of photon correlations or the intensity fluctuation spectrum in a solid angle Ω near the direction $\mathbf{n}_0$ (nonlinear resonance diffraction), the diagrams in Fig. 5(f)–(h), are dominant.[79] Physically, they correspond to beats of the diffractive (two-atom) component (Fig. 4b) with the single-atom fluorescence (Fig. 4a). The corresponding partially factorized terms in the fourth-order field correlation function are as follows [for convenience in comparison with the diagrams, the index of the atom whose evolution contributes the corresponding correlations is specified below; a summation over all sets $\{a \neq b \neq c\}$ is to be understood]:

for the diagram of Fig. 5f,

$$\langle E_\alpha^{(-)}(1)\rangle_a \langle E_\beta^{(-)}(2) E_\beta^{(+)}(2)\rangle_b \langle E_\alpha^{(+)}(1)\rangle_c,$$

for the diagram of Fig. 5g,

$$\langle E_\alpha^{(-)}(1)\rangle_a \langle E_\beta^{(-)}(2) E_\alpha^{(+)}(1)\rangle_c \langle E_\beta^{(+)}(2)\rangle_b,$$

for the diagram of Fig. 5h,

$$\langle E_\alpha^{(-)}(1)\rangle_a \langle E_\beta^{(-)}(2)\rangle_b \langle E_\beta^{(+)}(2) E_\alpha^{(+)}(1)\rangle_c$$

(the last quantity includes an anomalous correlation function). The role played by anomalous correlation functions in the statistics of radiation has been pointed out in several papers.[105–111,15,79] In this case, it is with the vanishing of these correlation functions at $\tau = 0$ that the photon antibunching and the SPPS are associated: The reconstruction of an induced dipole moment of the atom (not the population of an excited level!) is manifested after the atom "participates" in the emission of the "first" photon. A coherent contribution of this sort promotes a compensation for the Gaussian noise and tends to increase the regularity of the radiation. The correlation function $\langle E_\beta^{(+)}(2) E_\alpha^{(+)}(1)\rangle$ (+ c.c.) can be distinguished in a frequency-conversion scheme. On the diagrams in Fig. 5b, this case corresponds to the addition of all possible versions of the replacement of the photon propagators by the classical complex amplitudes $\mathscr{E}_h$, $\mathscr{E}_h^*$ of the field of the reference signal (from the same laser which is used to excite the fluorescence). Let us assume that its intensity, J_h is high in comparison with that of the fluorescence, and let us assume that the field $\mathscr{E}_h$ is shifted in phase by an amount θ with respect to the exciting field. We write the photocurrent correlation function[79]:

$$K(\tau) = \langle J\rangle \delta(\tau) + \langle J\rangle^2 + \langle J_h\rangle q\varkappa F_0\gamma \{[(\bar{\rho}_{12}\rho_{21}^{(11)}(\tau) - |\bar{\rho}_{12}|^2) \times \{1 + \exp[2i(\Delta - \varphi_h)]\} + \bar{\rho}_{22}[\rho_{12}^{(12)}(\tau) + \rho_{12}^{(21)}(\tau)\exp(2i\varphi_h)] + \text{c.c.}\}; \quad (3.15)$$

here $F_0 = (3/8\pi d_{21}^2)\ [\mathbf{d}_{21} - \mathbf{n}_0\ (\mathbf{d}_{21}\mathbf{n}_0)]^2$; $\varkappa = N\Omega$ if $\Omega < (\lambda/D)^2$ or $\varkappa = \nu_{at}\lambda^2 L$ if $\Omega > (\lambda/D)^2$; ν_{at} is the density of atoms; D and L are the dimensions of the interaction region respectively transverse and longitudinal with respect to the laser beam [under the condition $\Omega > (\lambda/D)^2$, the parameter $\varkappa$ determines the linear absorption at the center of the line]; and Δ is the phase of the induced dipole moment of the atom with respect to the exciting laser wave, i.e., $\bar{\rho}_{21} = |\bar{\rho}_{21}|\exp(i\Delta)$, where $\Delta = \text{arctg}\ [(\gamma/2)\times(\omega_{21} - \omega_0)^{-1}]$. Let us examine the variance of the number of photon counts over a small time interval T, using expression (3.15) (without the first term) in (2.10), and assuming $\rho_{ik}^{(mn)}(\tau) \approx \rho_{ik}^{(mn)}(0) = \delta_{im}\delta_{kn}$. We set $\varphi_h = \Delta$. We then find

$$\xi(T) \approx 2q\varkappa F_0\gamma T(\bar{\rho}_{22} - 2|\bar{\rho}_{12}|^2) = 2q\varkappa F_0\gamma T \frac{V_0^2[2V_0^2 - (\gamma^2/4) - \nu_0^2]}{[(\gamma^2/4) + \nu_0^2 + 2V_0^2]^2} \quad (3.16)$$

(we are using the steady-state values $\bar{\rho}_{22}$, $\bar{\rho}_{21}$). We find

$$\xi(T) < 0 \quad \text{for} \quad 2V_0^2 < \frac{\gamma^2}{4} + (\omega_0 - \omega_{21})^2; \quad (3.17)$$

here $\xi_{min}(T) = -(1/8)q\varkappa F_0\gamma T$ {this value is reached at $V_0^2 = (1/6)\ [(\gamma^2/4) + \nu_0^2]$}. It is useful to compare this result with expressions (2.14). According to the comments following those equations, we should identify the condition $\varphi_h - \Delta = 0$, under which (3.16) was derived, with the condition $\theta = 0$ in Subsection 2.3. We reach the conclusion that there is a squeezing of the quadrature component of $\mathbf{E}_1$ of the resonance-fluorescence field in the diffraction cone when inequality (3.17) holds. There is the further possibility that SPPS and squeezing in the nonlinear resonance diffraction of coherent radiation by a system of atoms could be observed under other conditions and in other types of measurements (squeezed states were not specifically mentioned in Ref. 79). Under optimum conditions, a dip with a relative depth of 0.28 can thus be achieved at $\omega \approx 0$ in the intensity fluctuation spectrum. In connection with the experimental possibilities, we would like to point out a subtle experiment[112] on the angular dependence of the intensity of nonlinear resonance fluorescence within the diffraction cone.

The relationship between SPPS and squeezing in nonlinear resonant fluorescence has been the subject of several studies.[63,64,113–116]

To conclude this section of the paper, we will mention some other studies of the statistics of photons in nonlinear resonance fluorescence and related experiments.

- Mutual correlations among the spectral components of the Stark triplet in the nonlinear resonance fluorescence of a two-level atom.[109,117–119]
- Intensity correlations, including photon antibunching and SPPS, in the nonlinear resonance secondary radiation of multilevel systems: systems with three levels[117,120–123] (the simplest model in which Raman scattering occurs); model systems of interacting boson modes[124]; electronic-vibrational systems (two-level molecules)[125,126]; and impurity centers in crys-

tals.[127–132] Of major interest here, in our opinion, is the possibility of achieving a spectral separation of the components of the secondary radiation and carrying out correlation studies of the various pathways and stages in the relaxation of complex systems.

- Time-varying[63,133] and cooperative[134–136] effects in photon antibunching and squeezing.
- Squeezing in the nonlinear resonance fluorescence of a regular chain of two-level atoms.[137]
- The effect of light in a squeezed state and/or with photon antibunching and SPPS on an atom and the transformation of the statistics of photons upon the propagation of light through a resonance medium.[138,139]

4. SQUEEZED STATES

Searches for sources of a radiation field in a squeezed state have now been the subject of many theoretical papers and several experimental studies. Models have been proposed which are based on several nonlinear optical effects: two-photon generation[34] (see the criticism in Refs. 31 and 140–142), parametric amplification,[34,143–146] harmonic generation,[147–149] parametric generation in a resonator,[150,151] lasing on free electrons,[152–154] Raman and hyper-Raman scattering,[155–158] and four-wave mixing.[159–167]

In this section we will examine two-photon coherent states and trace the relationship between squeezing and the dynamics of the source in the particular example of a degenerate parametric amplifier. We will discuss four-wave mixing, which is presently regarded as the most promising approach, and the experiments which have been carried out.

4.1. Two-photon coherent states

Two-photon coherent states, which were introduced by Yuen,[34] include a subset of states which minimize the product of the variances of the quadrature components and a subset of squeezed states [defined by inequalities (1.4)], but they do not include all squeezed states. The concept of two-photon coherent states has proved to be useful for analyzing the production and detection of squeezed states.

For a fixed mode of radiation field one constructs the operators

$$b=\mu a+\nu a^+,\quad b^+=\mu^* a^+ +\nu^* a \tag{4.1}$$

(a^+ and a are the photon creation and annihilation operators), where the complex numbers μ and ν satisfy the condition

$$|\mu|^2-|\nu|^2=1. \tag{4.2}$$

Transformation (4.1) under condition (4.2) is canonical (a Bogolyubov transformation); i.e., we have $[b, b^+]=1$. By introducing the operator b^+b, we can construct states with definite numbers of quasiparticles ("biphotons"). By analogy with ordinary coherent states $|\alpha\rangle$, the vectors $|\beta\rangle_{\mu,\nu}$ of two-photon coherent states, which are eigenvectors of the operator b, are defined:

$$b\,|\beta\rangle_{\mu,\nu}=\beta|\beta\rangle_{\mu,\nu}. \tag{4.3}$$

The variances of the quadrature components of the operator a [see (1.2)] in the state $|\beta\rangle\mu,\nu$ can be found easily by using the transformation which is the inverse of (4.1):

$$a=\mu^* b-\nu b^+,\quad a^+=\mu b^+-\nu^* b. \tag{4.4}$$

The results are

$$\langle(\Delta X_1)^2\rangle=\frac{1}{4}|\mu-\nu|^2,\quad \langle(\Delta X_2)^2\rangle=\frac{1}{4}|\mu+\nu|^2. \tag{4.5}$$

The minimum value of the product of uncertainties, $\delta X_1\ \delta X_2=\frac{1}{4}$, is reached in the limit $\mu,\nu\to\infty$ under condition (4.2) and with real (μ/ν). If $(\mu/\nu)\to 1$, the state $|\beta\rangle\mu,\nu$ becomes an eigenstate for X_1; if $(\mu/\nu)\to -1$, it becomes an eigenstate for X_2.

Canonical transformation (4.1) corresponds to a unitary transformation U such that $b=U^+aU$ (Ref. 168). We treat the operator U as an evolution operator governed by the Hamiltonian H: $U=\exp[-(i/\hbar)Ht]$. We can then find two-photon coherent states from the ground state or from any coherent state $|\alpha>$ (as an initial state) at $t>0$, provided only that H contains quadratic terms proportional to $(a^+)^2$, a^2. More-general transformations $b=\mu a+\nu a^+ +\varkappa$, where $\varkappa$ is a c-number or a c-function of the time, lead to a superposition of a coherent state and a two-photon coherent state. Equalities (4.5) remain in force. As we will see below, such states may be characteristic of SPPS.

4.2. Degenerate parametric amplifier

We treat the degenerate parametric amplifier here in an extremely simple and slightly formal model, but one which is popular in the literature: The fundamental signal—a wave at the frequency ω—is treated as an undamped oscillator excited parametrically at the frequency 2ω (actually, it is excited by a pump wave in the interaction of two waves in a medium with a quadratic polarization.[35,65,169] We choose the Hamiltonian of the system in the form

$$H=\hbar\omega a^+a+\hbar\,[f_1a^+ + f_2\,(a^+)^2+\text{H.a.}], \tag{4.6}$$

where the parameters f_1 and f_2 characterize the effect at the frequency ω (which simulates a wave at the entrance) and at the frequency 2ω (the parametric excitation by a pump wave), respectively. To calculate the variances of the quadrature components, we use the equations of motion in the Heisenberg picture. The relationship among the canonical transformation, a unitary transformation, and the Heisenberg picture of the motion in the problem of forced oscillations and the parametric excitation of a quantized oscillator was analyzed in Ref. 168 (Chapter VI), where there was essentially a tracking of the formation of states which were called "two-photon coherent states" in Ref. 34 and which were studied in detail. A solution of the equations of motion for the operator $a'(t)=a_{\rm H}(t)\exp(i\omega t)$ [$a_{\rm H}(t)$ in the Heisenberg picture] is[34]

$$a'(t)=a\,\mathrm{ch}\,(2R(t))-i\exp(-i\psi)\times a^+\,\mathrm{sh}\,(2R(t))+\varkappa(t), \tag{4.7}$$

where

$$R(t)=\int_0^t dt' r(t'),$$

$$\varkappa(t)=-i\int_0^t dt'\{f\,\mathrm{ch}\,[2R(t'-t)]-i\exp(-i\psi)\times f^*\,\mathrm{sh}\,[2R(t'-t)]\}, \tag{4.8}$$

$$f_1 = f \exp(-i\omega t), \quad f = |f| \exp(i\varphi)$$
$$f_2 = r(t) \exp(-i(2\omega t + \psi)) \tag{4.9}$$

Assuming $r(t) =$ const to simplify the problem, and choosing a specific pump phase $\psi = \pi/2$, we find

$$\langle(\Delta X_1)^2\rangle = \frac{1}{4}\exp(-4rt), \quad \langle(\Delta X_2)^2\rangle = \frac{1}{4}\exp(4rt). \tag{4.10}$$

Expressions (4.10) are identical to (4.5) in the case $\mu = \cosh(2rt)$, $\nu = \sinh(2rt)$. The ratio $(\mu/\nu) = \text{ctnh}\{2rt\}$ approaches unity at $rt \gg 1$. In other words, the state approaches an eigenstate for the quadrature component X_1. There is no squeezing in the case $r = 0$. At $r \neq 0$, a phase shift of π can transfer the squeezing from one component to the other. The expectation values of X_1 and X_2 are expressed in terms of the quadrature components $x_1 = (f+f^*)/2$, $x_2 = i(f^* - f)/2$:

$$\langle X_1\rangle = \frac{x_2}{2r}[1 - \exp(-2rt)],$$
$$\langle X_2\rangle = \frac{x_1}{2r}[\exp(2rt) - 1]. \tag{4.11}$$

To interpret the results and to make a comparison with the classical theory[35] we consider two operating regimes of a degenerate parametric amplifier. We choose the phase of the signal at the input in such a way that we have $x_1 = 0 (\cos\varphi = 0)$: a nonamplifying regime. Omitting factors which are not important for the present purposes, we write the field operator $E(t)$ in terms of quadrature components. In the latter we single out the expectation values and the operators representing deviations from the expectation values:

$$E(t) \sim (\langle X_1\rangle + \Delta X_1)\cos\omega t + \Delta X_2 \sin\omega t. \tag{4.12}$$

In the case of a strong parametric effect, with $rt \gg 1$, we find

$$\langle X_1\rangle \approx \frac{x_2}{2r}, \quad \langle(\Delta X_1)^2\rangle \ll 1;$$

i. e., the first term in (4.12) stabilizes, and acquires a semiclassical regular nature. Taking some license, we transform (4.12) as a nonoperator trigonometric expression:

$$E(t) \backsim \langle X_1\rangle \cos\left(\omega t - \frac{\Delta X_2}{\langle X_1\rangle}\right). \tag{4.13}$$

Despite the operator meaning of (4.13), it can be asserted that in this regime amplitude fluctuations are suppressed (and phase fluctuations are intensified). When the first term in (4.12) is singled out, the instantaneous signal-to-noise ratio $\langle X_1\rangle/\delta X_1$ can be made arbitrarily large (in this model). If we choose a different phase for the input signal, such that we have $x_2 = 0$, $\cos\varphi = 1$, we obtain a regime of maximum amplification. If $\exp(4rt) \gg 1$, we find that the phase fluctuations are suppressed and the amplitude fluctuations intensify:

$$E(t) \backsim (\langle X_2\rangle + \Delta X_2)\sin\omega t. \tag{4.14}$$

Expressions (4.12)–(4.14) can be used to trace the analogy with the classical theory of a degenerate parametric amplifier. In particular, there is a relationship with the classical "phase quantization" effect.[35,170] The difference in the results of the quantum theory stems primarily from the complementarity (noncommutativity) of the quadrature field components [see uncertainty relation (1.4)] and thus the complementarity of the amplitude and phase random modulation. On the other hand, we are speaking here in terms of a decrease in the quantum noise, to the extent allowed by the conditions for the discrimination of squeezed components of the signal.

Let us examine the observable quantities in this model. The expectation value and the variance of the number of photons in the fundamental mode are[171]

$$\langle m\rangle_t = \langle a^+a\rangle_t = |\varkappa|^2 + \text{sh}^2(2rt), \tag{4.15}$$

$$\langle(\Delta m)^2\rangle_t = \langle(a^+a)^2\rangle_t - \langle a^+a\rangle_t^2$$
$$= |\varkappa\,\text{ch}(2rt) - i\varkappa^* \exp(-i\psi)\,\text{sh}(2rt)|^2$$
$$+ 2\,\text{sh}^2(2rt)\,\text{ch}^2(2rt). \tag{4.16}$$

Under what conditions would SPPS follow from squeezing in this case? The result now depends on the relation between the phases of the input signal and the pump. We set $i\varkappa^* \exp(-i\psi) = \varkappa$. Under the condition $|\varkappa|^2 \gg \exp(4rt)$ we find

$$\langle m_t\rangle \approx |\varkappa|^2,$$
$$\langle(\Delta m)^2\rangle_t \approx 4|\varkappa|^2\langle(\Delta X_1)^2\rangle \approx \langle m\rangle\exp(-4rt). \tag{4.17}$$

If the condition $rt \gg 1$ also holds, the photon distribution is substantially sub-Poisson (Fig. 9). In contrast, the condition $|\varkappa|^2 \ll 1$ leads to an example of a squeezed state without SPPS.

4.3. Realization of squeezed states

The example discussed above shows how squeezed states can arise in phase-sensitive effects of nonlinear optics. However, there are serious difficulties in the realization of squeezed states. For example, the incorporation of a damping of the "ω" mode in a degenerate parametric amplifier leads to the following expressions[34] for the variances of X_1, and X_2 at $rt \gg 1$:

$$\langle(\Delta X_1)^2\rangle \approx \gamma[4(\gamma + 2r)]^{-1}, \quad \langle(\Delta X_2)^2\rangle \to \infty \tag{4.18}$$

(γ is the damping constant). If the squeezing is to be substantial, the "2ω" pump wave must be very intense; i.e., we must have $2r \gg \gamma$. The attainment of a squeezed state requires satisfying some stringent phase relations. For example, tak-

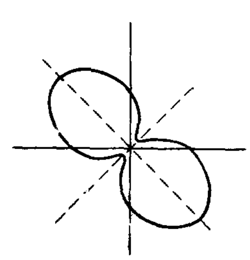

FIG. 9. Polar diagram of the Fano parameter $F = 1 + \xi t = \langle(\Delta m)^2\rangle/\langle m\rangle$ versus the phase of the resultant signal from a degenerate parametric amplifier, which is proportional to $\varkappa$ under the condition $|\varkappa|^2 \gg \exp(4rt)$ [expression (4.16)]. Here the values $4rt = 1.1$ and $\psi = 0$ are assumed. There is the possibility of SPPS here: $F_{min} = 0.34$. The experiment of Ref. 172 revealed a tendency toward this phase dependence of F.

ing an average over the pump phase ψ [see (4.16) and Fig. 9] disrupts the squeezing and the SPPS. Fluctuations in the amplitudes of the incoming waves are also pernicious. Photon correlation effects in the scattering by groups of atoms may mask a squeezing.

The greatest difficulties seem to stem from the small value of the nonlinear susceptibilities and the short interaction time (in the preceding section, t was actually the time of transmission through the nonlinear medium). The interaction time increases in a parametric generation arrangement.[150,151] There is the possibility in principle here of achieving squeezing in a steady state. Calculations[150,151] incorporating a resonator damping yield $\langle(\Delta X_i)^2\rangle = \frac{1}{8}$ as the minimal variance for the quadrature of the wave which is generated.

The possibility of achieving squeezed states during four-wave mixing has been discussed intensely in recent years.[159–167] The schemes which have been proposed share the following idea. From an interaction determined by a third-order polarization of the type $\mathbf{P} = \chi\mathbf{EEE}$, the part which can be associated with an effective interaction operator is singled out by the experimental conditions:

$$H_I = \hbar\,(k a_1^+ a_2^+ + \text{H.a.}). \tag{4.19}$$

Here $k \propto \chi E_{p1} E_{p2}$, where E_{p1} and E_{p2} are the complex amplitudes of the pump waves (the latter may be regarded as classical), and a_1^+, a_2^+ are operators representing the parametrically excited waves. We assume that the frequencies of all waves are the same (the case of complete degeneracy). We form a linear combination of the operators a_1^+ and a_2^+:

$$b^+ = s_1 a_1^+ + s_2 a_2^+, \quad |s_1|^2 + |s_2|^2 = 1. \tag{4.20}$$

Such a transformation might correspond to, for example, a mixing in a beam splitter. Operator (4.19), expressed in terms of b^+ and b, contains terms of the type B^+b^+, bb; i.e., it can generate squeezed states in the course of the evolution (Subsection 4.1). Four-wave parametric processes can thus lead to the production of a field in a squeezed state and to SPPS observable under special conditions.

The experiments of Refs. 160 and 161 used an arrangement of degenerate four-wave mixing involving the photon conversion $\mathbf{k}_{p1} + \mathbf{k}_{p2} \rightarrow \mathbf{k}_1 + \mathbf{k}_2$, with $\mathbf{k}_{p1} = -\mathbf{k}_{p2}$. A study was made of the statistics of the photons of the probing and phase-conjugate waves; these statistics turned out to be Poisson, in agreement with the predictions of the theory for the actual experimental conditions. It was thus demonstrated that there was a compensation for the Gaussian (wave) fluctuations of these waves separately as a result of the squeezing. The extent of the squeezing, however, was substantially limited by losses in the medium (sodium vapor). It was not possible to reduce the fluorescence background and thereby raise the efficiency of the four-wave mixing in order to bring out the SPPS in the mixing of two waves at the exit. For this reason, and also because of the inadequate stability of the interferometer, no analysis was made of the photon statistics for the superposition of the probing and phase-conjugate waves.

Levenson *et al.*[165] studied nondegenerate four-wave mixing in an optical fiber. In the photocurrent noise in the course of the frequency conversion, they singled out a spectral region which was free of the harmful manifestations of stimulated Brillouin scattering. They developed a strategy for future searches in experiments of this type. During the frequency conversion of the output radiation (with frequencies $\omega_p \pm \delta\omega$), one could expect a lowering of the spectral noise level at the frequency of the pump wave, ω_p, in the intensity fluctuation spectrum with respect to the shot noise in the region $\omega \approx \delta\omega$.

Slusher *et al.*[167] were the first to establish the characteristics of the squeezed state in the photocurrent fluctuation spectrum. We will examine this experiment in slightly more detail. They used an arrangement of nondegenerate four-wave mixing in an optical cavity (Fig. 10). A beam of sodium atoms served as a nonlinear converter. The deviation of the frequency of the exciting dye laser from the nearest resonance (the D_2 line) was substantially greater than the radiative width and the residual Doppler width (the latter resulting from the divergence of the laser beam). Cavity R_1 formed a standing wave of the laser light (the pump). In cavity R_2, a superposition of pairs of phase-conjugate waves of different frequencies appeared. The laser frequency was the same as one of the frequencies of cavity R_2 (the stabilization system is not shown in Fig. 10). They measured the photocurrent spectrum in a balanced arrangement[173] with frequency conversion at the frequency of the laser, ω_p, of the waves leaving the cavity with frequencies $\omega_p \pm 3\nu_2$ (ν_2 is the interval between the natural modes of cavity R_2).

This experimental arrangement made it possible to minimize the factors which would disrupt squeezing: spontaneous emission, amplification, and reabsorption. The loss was determined primarily by the exit of photons from cavity R_2. The situation can be described approximately by the equations of motion for the operators of two real waves with frequencies $\omega_{1,2} = \omega_p \pm 3\nu_2$, by making use of the effective interaction in the form in (4.19) and by allowing for damping (the constant C). Let us examine the mixing of the waves leaving the cavity with the field of the reference signal at the frequency ω_p. Using expression (2.5) we find

$$K(\tau) - \langle I\rangle^2 = q\,\langle I\rangle\,\delta(\tau) + \frac{1}{2} C q^2 I_h\,\{2\,\langle b^+ b(\tau)\rangle + [\exp(2i\varphi_h)\,\langle b^+ b^+(\tau)\rangle + \text{c.c.}]\}. \tag{4.21}$$

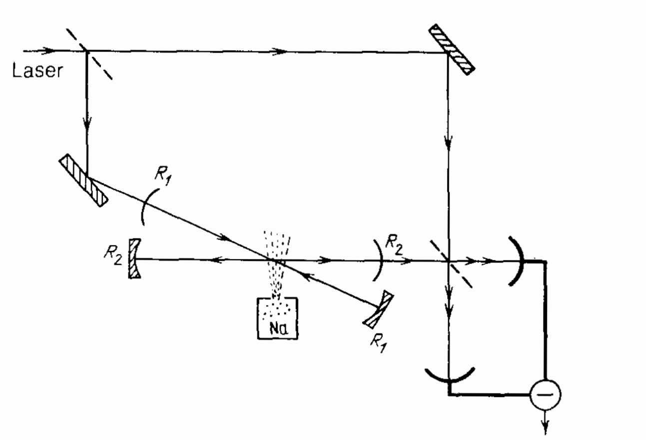

FIG. 10. Layout of the experiment of Ref. 167 on four-wave mixing in an optical cavity. See the text proper for an explanation. The stabilizing and control parts of the arrangement which were used in Ref. 167 are not shown here.

Here I_h and φ_h are the intensity and phase of the signal of the local oscillator (heterodyne), and $b^+ = (a_1^+ + a_2^+/\sqrt{2}$. The anomalous correlation functions depend on the phase of the complex coupling constant: $\langle b^+ b^+(\tau)\rangle \propto \exp(-2i\vartheta)$. The non-Poisson term in photocurrent correlation function (4.21) is minimized under the condition $\varphi \equiv \varphi_h - \vartheta = \pi/2$, and it is negative in this case. In the intensity fluctuation spectrum we should expect the appearance of a dip below the level of the shot noise, in the form of a Lorentz line centered at the frequency $3\nu_2 + |k|$, with a width $C/2$.

Figure 11 is a schematic drawing of the basic result of Ref. 167 (Slusher *et al.*[167] showed photographs of the signal on a spectrum analyzer at the output from the balanced arrangement, along with a detailed explanation of the various aspects of the experiment). Slusher *et al.*[167] estimate the relative depth of the dip to be 7%, which they say corresponds to a 20% squeezing. At any rate, there is no doubt that a positive effect was achieved, and this result strengthens the hope that it will be possible to achieve an effective lowering of the quantum noise in parametric processes.

There is a close analogy between result (4.21) and correlation function (3.15), which characterizes the photocurrent fluctuations during the frequency conversion of the light of resonant fluorescence in the diffraction cone.[79] The superficial difference between (4.21) and (3.15) results solely from the fact that the correlation functions for the field operators are expressed explicitly in terms of the elements of the density matrix of the source atoms in (3.15).

5. LUMINESCENCE IN THE CASE OF REPULSIVE STATISTICS OF ATOMIC EXCITATIONS

In this section we examine the possibility of achieving sub-Poisson photon statistics and photon antibunching in the spontaneous emission of a system of atoms. We assume that the pump does not create an atomic coherence and that it does not saturate the radiating transition. Among all the processes which form the photocurrent from the detected radiation we should then consider the two-atom direct and interference processes (Fig. 5b and c). We have already noted that the interference process (Fig. 5c) leads to a bunching of photons; the corresponding contribution to the parameter ξ [expression (2.9)] is $q\delta$, where δ is the degeneracy parameter of the spontaneous emission. For the problems which we will be discussing in this section of the paper the intensities are comparatively low, and the luminescence linewidth is comparatively large (in comparison with the radiative

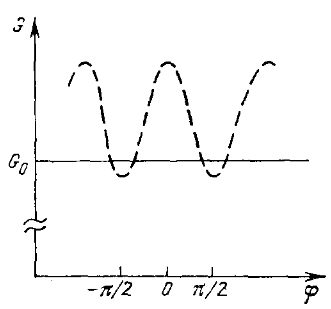

FIG. 11. The signal (the spectral component of the photocurrent fluctuations singled out near the frequency $3\nu_2$) versus the phase shift of the local oscillator (according to the data of Slusher *et al.*[167]). Here G_0 is the level of the shot noise.

width), so that with a relative photon collection angle $\Omega = S/4\pi R^2$ of the order of unity the expected effects of the direct process in the statistics will be greater than the contribution of the interference process ($\delta \ll 1$). We are thus examining the fluctuations in the radiation which correspond to Fig. 5b and which are related to correlations among excitation events. If the pump is assumed to be a wide-band Poisson process (if the spectral width of the pump exceeds that of the luminescence line), then if the spontaneous-emission events are statistically independent the photocurrent will also be a Poisson process, with a correlation function

$$K(\tau) = q\overline{M}\Omega\delta(\tau) + (q\overline{M}\Omega)^2; \tag{5.1}$$

here $\overline{M}$ is the average number of atoms which are excited per unit time over the entire volume (it is assumed that there is no quenching of the luminescence). The conversion of non-Poisson fluctuations of the pump into fluctuations of spontaneous emission has been studied experimentally and theoretically in several places (see the bibliography in Ref. 54 and also Refs. 95 and 174; see Refs. 76, 175, and 176 regarding sub-Poisson excitation).

The photocurrent correlation function is expressed in terms of the correlation function of the number of atoms excited at the time t, $N(t)$, in the following way:

$$K(\tau) = q\overline{M}\Omega\delta(\tau) + (q\overline{M}\Omega)^2 \langle N(t_1) N(t_2)\rangle \overline{N}^{-2}, \tag{5.2}$$

where $\overline{N}$ is the steady-state expectation value of $N(t)$. Here the angle brackets mean an average over the statistics of the pump and over the statistics of the radiating atoms. In particular, if the atoms do not interact with each other, we can write

$$\langle N(t_1) N(t_2)\rangle = \int_{-\infty}^{t_1} dt' \int_{-\infty}^{t_2} dt'' \langle M(t') M(t'')\rangle \times \exp[-\gamma(t_1 - t') - \gamma(t_2 - t'')], \tag{5.3}$$

where γ is a radiation constant. If the pump fluctuations are fast, the τ dependence is determined by the value of γ. In the spectrum of the correlation function $K(\tau)$, a line of width γ appears. This fact underlies a method for measuring the natural line width under conditions of a predominant inhomogeneous broadening of a luminescence line.[54]

In the case of repulsive statistics of excitations we can expect SPPS in the luminescence. Two specific mechanisms have been proposed for achieving SPPS by this approach: exciting a gas with an electron beam with a depression of shot noise[76,175,176] and de-exciting cooperative processes.[177]

5.1. Excitation of atoms by an electron beam with depressed shot noise

The depression of the shot noise of an electron beam (in a diode, for example) which occurs in the case of a space-charge limitation of the current has been studied quite thoroughly.[35,55,178] The variance in the number of electrons which pass through the region of the limiting space charge and which are detected over a time $T \gg \gamma_e^{-1}$ (γ_e^{-1} is the correlation time of the electrons in the beam) can be written

$$\langle(\Delta m_e)^2\rangle = \langle m_e\rangle \Gamma^2, \quad \xi_e = -(1 - \Gamma^2). \tag{5.4}$$

The depression factor Γ^2 may be of the order of 10^{-2}. It follows from (5.4) that there is a low-frequency dip in the fluctuation spectrum of the beam against the background of a shot noise with a relative depth $(1-\Gamma^2)$. Assuming for simplicity that this dip has a Lorentz shape, we write the electron current correlation function in the form

$$\langle I(0)\,I(\tau)\rangle=\langle I\rangle\,\delta(\tau)+\langle I\rangle^2-\frac{1}{2}\langle I\rangle\,\gamma_e\,(1-\Gamma^2)\times\exp(-\gamma_e\,|\tau|). \qquad (5.5)$$

Using (5.5) (without the first—shot—term!), we can find the correlation functions in (5.3). The variance in the number of atoms excited over a time $T\gg\gamma^{-1}$ is

$$\langle(\Delta N)^2\rangle_T=\langle N\rangle_T\,[1-\eta\,(1-\Gamma^2)]. \qquad (5.6)$$

This quantity turns out to be important in the generation problem of Ref. 179 (Sec. 6). From (5.2) and expressions (2.8)–(2.10) we then find the variance of the number of photoelectrons during the detection of the luminescence:

$$\langle(\Delta n)^2\rangle_T=\langle n\rangle_T\,[1-q\Omega\eta\,(1-\Gamma^2)]; \qquad (5.7)$$

here η is the atomic excitation efficiency ($\langle M\rangle=\eta\langle I\rangle$).

The dip in the intensity fluctuation spectrum against the background of the shot noise has a width γ and is smaller by a factor of $q\Omega\eta$ than the dip in the noise spectrum of the electron beam. The reason for the weakening of the correlations is a chain of transformations of the pump statistics: The excitation of an atom by an electron occurs with a probability η; the emitted photon falls within the collection angle of the photodetector with a probability Ω; and, finally, it is converted into a photoelectron with a probability q.

The idea of producing sub-Poisson radiation during excitation by an electron beam with a depression of shot noise[76] was implemented in the experiment of Ref. 176. A study was made of the statistics of the luminescence photons of mercury vapor (the $6^3P_1-6^1S_0$, with $\lambda=253.7$ nm) in a Franck-Hertz tube. Under the experimental conditions, the atomic excitation efficiency was $\eta\approx0.15$; the geometric collection factor was $\Omega\approx0.1$; and the depression of the shot noise of the electron beam was characterized by the parameter value $\xi_e=-(1-\Gamma^2)\approx-0.9$. The quantum yield ($q$) of the photomultiplier at the wavelength 253.7 nm was 0.15. In addition to these parameter values, which appear in expression (5.7), account was also taken of the filter transmission coefficient (0.83), the photon emission factor, which was reduced by secondary processes in the tube (0.8), and yet another factor of 0.3 due to the presence of Poisson illumination from the heater filament. Multiplying all these coefficients together, we find $\xi_\infty\approx-0.0007$ as the theoretical value of the sub-Poisson factor for photoelectrons. This value agrees satisfactorily with the experimental result when the dead time is taken into account.

5.2. Sub-Poisson statistics of the luminescence of impurity centers in crystals resulting from a cooperative de-excitation

A "repulsive" statistics of excitations can also arise as a result of cooperative processes caused by an interaction of impurity centers in a crystal. Among these processes are the summation of excitations, cooperative sensitization, and nonlinear quenching.[180]

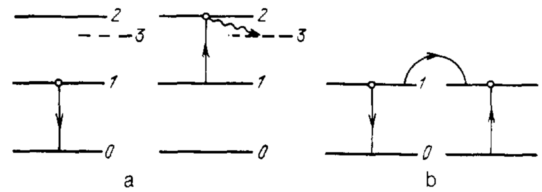

FIG. 12. a—Summation and b—transfer of excitations.

Let us consider a radiative transition with a frequency ω. If pairs of excitations which arise close together in time and space leave the radiative channel with a frequency ω with a high probability because of some cooperative process, the statistics of the excitations turns out to be "repulsive" and reminiscent of the statistics of particles of an incompressible fluid.[39] As an example we consider the well-studied phenomenon of cooperative luminescence[180]: a summation of excitations with an energy $\hbar\omega$ of two impurity centers at one of these centers (Fig. 12). The summation is rendered irreversible because of the fast $2\to3$ multiphonon relaxation. The subsequent emission of a quantum with a frequency $\omega_{3,0}$, i.e., the cooperative luminescence proper, is not of interest here: We are interested in the statistics of the photons corresponding to the 1–0 transition. A high summation efficiency is achieved by virtue of migration of excitations (Fig. 12b) and transport of two excitations over distances over which the summation occurs. If the migration is intense, a comparatively low degree of excitation is sufficient for achieving a significant SPPS effect. Saturation of the 0–1 transition is not required; this circumstance is an advantage of this method over the nonlinear-optics methods which have been proposed. Fluctuations in the number of atoms in the effective range of the pump do not disrupt the SPPS, in contrast with the case in other examples.

Let us assume that the pump is a steady-state Poisson process. To calculate the population correlation function $\langle N_1(t_1)N_1(t_2)\rangle$ in (5.2) we introduce the following definitions: $F(\mathbf{x}_1,\mathbf{x}_2)d^3x_1d^3x_2/V^2$ is the steady-state probability for finding two excitations in volume elements around the points $\mathbf{x}_1$ and $\mathbf{x}_2$; $f_0(\tau)\equiv\rho_{11}^{(00)}(\tau)$ is the probability for the excitation of some arbitrarily fixed atom by the pump in the time interval $(0,\tau)$; and $f_1(\tau)\equiv\rho_{11}^{(11)}(\tau)$ is the probability that an atom excited to level 1 at time $t_0=0$ will remain at this level at the time τ. We assume that the system is spatially homogeneous and that the interaction between impurity atoms is a central interaction. The functions introduced here have the properties

$$f_0(\tau)\underset{\tau\to\infty}{\longrightarrow}\bar f,\quad f_1(\tau)\underset{\tau\to\infty}{\longrightarrow}0,\quad F(|\mathbf{x}_1-\mathbf{x}_2|)\underset{|\mathbf{x}_1-\mathbf{x}_2|\to\infty}{\longrightarrow}\bar f^2, \qquad (5.8)$$

where $\bar f$ is the steady-state probability for finding a fixed atom excited. If the pump is weak, and if the time scale for the escape of an excitation from the interaction volume is long (in comparison with γ^{-1}), the correlation function for the number of excitations can be written in the form

$$\langle N_1(t_1)\,N_1(t_2)\rangle=n_0^2\int d^3x_1d^3x_2\,[F(|\mathbf{x}_1-\mathbf{x}_2|)\,f_1(\tau)+\bar f\,f_0(\tau)], \qquad (5.9)$$

where n_0 is the density. The first term in the integral corresponds to the appearance of two excitations, which have emitted detected photons, at times in the interval $(-\infty, t_1)$; the second term corresponds to the appearance of one excitation in the interval $(-\infty, t_1)$ and of the other in the interval $(t_1, t_2 = t_1 + \tau)$. In the absence of an interaction, $F = \bar{f}^2$, correlation function (5.7) will evidently be independent of the time, and we find (5.1), i.e., a Poisson statistics of photons.

In the case of repulsive statistics of excitations, we can define a correlation volume V_c (an effective summation volume) as follows:

$$V_c = -\int g(\rho)\, dV, \tag{5.10}$$

where $g(\rho)$ is a pair correlation function of excitations, which is directly related to the function $F(\rho)$ introduced above ($\rho = |\mathbf{x}_2 - \mathbf{x}_1|$):

$$F(\rho) = \bar{f}^2 [1 + g(\rho)]; \tag{5.11}$$

here $g(\rho) \to 0$ as $\rho \to \infty$. Using (5.11), we can find the correlation function for the concentrations of excitations and the variance in the number of excitations in the volume of interest:

$$\langle n_1(0)\, n_1(\rho)\rangle = \bar{n}_1 \delta^{(3)}(\rho) + \bar{n}_1^2 g(\rho); \tag{5.12}$$

$$\langle (\Delta N_1)^2 \rangle_V = \langle N_1 \rangle_V (1 - V_c \bar{n}_1). \tag{5.13}$$

The parameter ξ_T for the photon counts during the detection of the luminescence is

$$\xi_T = -q\Omega\gamma\bar{n}_1 V_c \frac{2}{T} \int_0^T d\tau\, (T - \tau)\, f_1(\tau). \tag{5.14}$$

Taking the integral in (5.14) (divided by T) as a definition of the average lifetime of an excitation (τ_1) at long times T, we find

$$\xi_\infty = -2q\Omega\bar{n}_1 V_c \gamma \tau_1. \tag{5.15}$$

We introduce the accumulation constant α [cm^3/s]: $\alpha\bar{n}_1$ is the average accumulation rate per atom excited to level 1. If we assume $V_c = \alpha\tau_1$ and $\tau_1^{-1} = \gamma + \alpha\bar{n}_1$, we can express ξ_∞ in a simple way in terms of the quantum yield of the $1 \to 0$ luminescence, $\eta_1 = \gamma\tau_1$, and the quantum yield in the summation channel, $\eta_2 = \alpha\bar{n}_1\tau_1$:

$$\xi_\infty = -2q\Omega\eta_1\eta_2. \tag{5.16}$$

It is not difficult to see that in this model the maximum SPPS effect $\xi_{\infty,\min} = -q\Omega/2$ is reached at $\alpha\bar{n}_1 = \gamma$, i.e., under the condition that the summation rate is equal to the rate of spontaneous emission.

As an example we cite data on the cooperative luminescence of Er^{3+} in crystals of the fluorite type[181]: $\gamma \approx 10^2$ s^{-1}, $\alpha \approx 10^{-14}$ cm^3/s. The condition $\alpha\bar{n}_1 = \gamma$ is reached at $\bar{n}_1 \approx 10^{16}$ cm^{-3}. Excitation concentrations at this level are observed at a moderate pump level (the typical concentration of impurity centers is $n_0 \approx 10^{22}$ cm^{-3}). A summation of excitations under corresponding conditions is used to depopulate the lower working level during generation.[182,183]

Cooperative processes occur in activated crystals, which can provide larger values of $|\xi_\infty|$. In nonlinear quenching,[184] for example, one of the atoms of a pair—that at which the sum excitation arises—can revert to level 1 as the result of a rapid radiationless relaxation (Fig. 12a). Of two excitations which appear at nearly the same time, only one is lost for the 1–0 radiative channel.

Other processes characterized by large values of the pair-de-excitation constant are collisions of excimer molecules[185] and the triplet-triplet annihilation of molecules.[186,187] In either case, a value $\alpha \approx 10^{-10}$ cm^3/s can be reached, and there is the hope of finding a specific system suitable for realizing SPPS.

6. GENERATION OF RADIATION WITH ENHANCED REGULARITY

So far, only a few studies have proposed theoretical models of radiation sources with a sub-Poisson photon statistics which use the principle of lasing. Some studies which do fall in this category are Refs. 188 and 189, which deal with a laser with a multiphoton loss, and Refs. 179 and 190, where the sub-Poisson statistics appears as a result of the statistical properties of the pump. The results of Refs. 179 and 190 are based on a refinement[190] of the quantum theory of generation[191,192] which is important for the statistics of the emission from a laser. We begin this section of the review with a brief description of the quantum theory of a laser which correctly incorporates the effects of the pump statistics. We then move on to a discussion of some specific models.

The methods discussed below for reducing intensity fluctuations (to SPPS) correspond to an "amplitude superstabilization," which goes beyond the semiclassical approach in the theory of generation. The optimum limit for the latter is a stabilization level which corresponds to a mixture of coherent quantum states with a random phase, a fixed amplitude, and a Poisson distribution of the number of photons. Sub-Poisson photon statistics in generation does not degrade the ordinary optical spectrum of single-mode generation in operation far above the threshold,[190] where the line width is determined primarily by phase diffusion.[191,192]

6.1. Incorporation of the pump statistics in the theory of single-mode generation

The possibility of constructing a closed kinetic equation for a field oscillator in the theory of a single-mode (gas) laser[191,192] is based on a relation between the relaxation time of the atom, τ_a, and that of the field C^{-1}: $\tau_a \ll C^{-1}$, where C is the resonator width. The starting point for the theory of a gas laser is an equation for the density matrix F of a four-level system (Fig. 13) and for a single mode of a quantized electromagnetic field of the generation on the 2–1 working transition. If an atom appears in level 2 in a time interval $\Delta t (\Delta t \gg \tau_a)$, it will certainly appear in the same interval in state (a) or (b), as a result of a relaxation mechanism (the possible appearance of an atom at the right end of the interval Δt in a region of the order of τ_a can be ignored). Defining the density matrix ρ as the trace of the matrix F in terms of atomic variables $\rho = \mathrm{Sp}_{\mathrm{at}} F$, we write the transformation of the matrix ρ due to the action of one atom on the interval Δt:

$$\rho(t + \Delta t) = F_{aa}(t + \Delta t) + F_{bb}(t + \Delta t). \tag{6.1}$$

Here F_{aa} and F_{bb} are matrix elements in terms of atomic variables and operators in terms of field variables. They can be found by solving a system of equations for the matrix F_{ij} ($i, j = 1, 2, a, b$) under the initial condition $F_{22}(t) = \rho(t)$. As a result, (6.1) can be written in the form

$$\rho(t + \Delta t) = (1 + \hat{u})\, \rho(t). \tag{6.2}$$

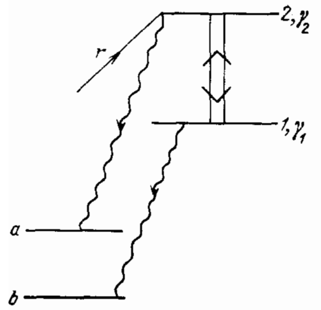

FIG. 13. Scheme of levels and transitions in the generation theory of Refs. 191 and 192. The working transition is the 2–1 transition; the pumping is shown in a simplified manner; the effective pump rate is a random process.

The operator $\hat{u}$ is expressed in terms of the coupling constants of the subsystems, the relaxation constants, the deviation of the gain line (in this case, a homogeneous line) from the center, and the creation and annihilation operators for the field mode (a^+, a). The operator $\hat{u}$ was found explicitly in Refs. 191 and 192. The contribution of one atom to the increment in the field density matrix, $\Delta\rho$, over a time Δt is small. The effect of many atoms on the field was taken into account in Refs. 191 and 192 by multiplying $\hat{u}\rho(t)$ by the number of active atoms, $r\Delta t$ (r is the pump rate):

$$(\Delta\rho)_+ = r\Delta t\, \hat{u}\rho(t). \tag{6.3}$$

The loss is described by a linear relaxation of the field operator with a damping constant C. Since the time scale of the variations in ρ is determined by the loss, we construct the large-scale derivative $\Delta\rho/\Delta t$ ($\Delta t \ll C^{-1}$, but $\Delta t \gg \tau_a$). The final equation is

$$\dot{\rho} = (\dot{\rho})_+ + (\dot{\rho})_-, \tag{6.4a}$$

$$(\dot{\rho})_+ = r\hat{u}\rho, \tag{6.4b}$$

$$(\dot{\rho})_- = C\left[a\rho a^+ - \frac{1}{2}(a^+a\rho + \rho a^+a)\right]. \tag{6.4c}$$

It was pointed out in Ref. 190 that the transformation of the field density matrix over a time Δt as a result of the excitation of many active atoms is multiplicative in the contributions of the individual atoms. A more detailed analysis of the effect of the pump leads to the expression

$$\rho(t+\Delta t) = \prod_{i=1}^{N} (1 + w_i u)\rho(t), \tag{6.5}$$

$$w_i = \int_t^{t+\Delta t} r_i(t')\,dt'. \tag{6.6}$$

It is assumed here that the pump is weak ($w_i \ll 1$) and of a broad-band nature; i.e., an excitation event occurs during a brief time interval (briefer than τ_a). We can write the pump $r = \Sigma_i r_i$ as a random pulsed process. For a correct incorporation of fluctuations in (6.5) we should retain terms in the expansion up to $\hat{u}^2$ inclusively. Terms with higher powers of $\hat{u}$ contribute little, because of the smoothness of the dependence of the matrix elements $\rho_{n,n+m}$ on the index n. We are assuming $\overline{(\Delta n)^2} \sim \bar{n}$, $\bar{n} \gg 1$, where $\bar{n}$ is the average number of photons in the resonator. The increment in the density matrix due to the active atoms is now different from the quantity $(\Delta\rho)_+$, given by (6.3):

$$(\widetilde{\Delta\rho})_+ = \left[N\hat{u} + \frac{1}{2}N(N-1)\hat{u}^2\right]\rho(t). \tag{6.7}$$

Here $N = N(\Delta t)$ is the total number of atoms which have been in level 2 during time interval ΔT. Expression (6.7) is written for a specific realization of excitation events. Let us assume that binary correlations of the pump are determined by the time τ_c, and let us choose $\Delta t \gg \tau_c$. In this case, we can ignore the statistical dependence of the cofactors in (6.7) which is associated with the ends of the interval Δt, and we can carry out an averaging over the pump statistics in accordance with the rule $\overline{[\ldots]}\bar{\rho}$ in (6.7). In constructing the large-scale derivative here we should again point out that the time scale of the variations in ρ is determined by the quantity $C^{-1} \gg \Delta t$. Incorporating the loss by the method described above, we find the equation

$$\dot{\rho} = (\dot{\beta})_+^{(1)} + (\dot{\rho})_+^{(2)} + (\dot{\rho})_-. \tag{6.8}$$

In (6.8), $(\dot{\rho})_+^{(1)}$ differs from the corresponding quantity in (6.4b) only in the replacement $r \to \bar{r}$; $(\dot{\rho})_-$ is given by (6.4c); and $(\dot{\rho})_+^{(2)}$ is the rate of change of the density matrix due to the binary correlation of pumping events,[81]

$$(\dot{\rho})_+^{(2)} = \frac{\bar{r}}{2}\zeta\hat{u}^2\rho. \tag{6.9}$$

The parameter ζ is related to the pump statistics:

$$\overline{(\Delta N)^2} = \bar{N}(1+\zeta). \tag{6.10}$$

The condition $\Delta t \gg \tau_c$, means that ζ is analogous to the parameter ζ_∞ (Subsection 2.2). In the case of a Poisson pump we would have $\zeta = 0$, and Eq. (6.8) would become Eq. (6.4a)—the equation of the conventional theory of a laser, with the obvious replacement of the pump rate by its average value over the Poisson statistics, $\bar{r}$. If the pump is sufficiently far above the threshold (and the pumping occurs exclusively to the upper working level) Eq. (6.4a) leads to a coherent state of the generation field with a variance $\overline{(\Delta n)^2} = \bar{n}$.

In the case of a sub-Poisson pump statistics ($\zeta < 0$) there can be an SPPS in the generation.

We introduce the simplifying conditions[190] $\gamma_2 = 0$ and the system is tuned to the center of the gain line. In this case the diagonal matrix elements of the operators $\hat{u}\rho$ and $\hat{u}^2\rho$ are

$$(\hat{u}\rho)_{nn} = -\rho_{nn} + \rho_{n-1,\,n-1}, \tag{6.11a}$$

$$(\hat{u}^2\rho)_{nn} = -\rho_{nn} + 2\rho_{n-1,\,n-1} - \rho_{n-2,\,n-2}. \tag{6.11b}$$

Using expressions (6.11a) and (6.11b), we find that in the continuum approximation (in n) Eq. (6.8) becomes a Fokker-Planck equation

$$\frac{\partial\rho}{\partial\tau} = \frac{\partial}{\partial n}[(n-\bar{n})\rho] + \bar{n}\left(1+\frac{\zeta}{2}\right)\frac{\partial^2\rho}{\partial n^2}, \tag{6.12}$$

where $\tau = Ct$, and $\bar{n} = \bar{r}/C$ is the average number of photons in the resonator. Equation (6.12) leads to a steady-state distribution function ρ_{nn}, the number of photons in the resonator, with a variance

$$\overline{(\Delta n)^2} = \bar{n}\left(1+\frac{\zeta}{2}\right). \tag{6.13}$$

We turn now to some specific examples.

6.2. Regular pump

A scheme involving a time-varying (periodic) excitation of atoms was examined in Ref. 190 in which it was possible to collect all the atoms in the upper working level during a pulse of length $T_0 \ll \tau_a$. The field energy was modulated at the pulse sequence period T_1; under the condition $T_1 \ll C^{-1}$, the modulation depth was small. This regular modulation is of no interest (it corresponds to discrete lines in the intensity fluctuation spectrum).

In the case with which we are concerned here, of a regular pump, we should set $\zeta = -1$ in (6.13). The variance of the distribution ρ_{nn} is then half that in the coherent state. The spectrum of fluctuations in the photocurrent from the output radiation, $G(\omega)$ is given in the case $\gamma_2 = 0$ by

$$G(\omega) = C_p \bar{n} [1 - C_p C (C^2 + \omega^2)^{-1}], \qquad (6.14)$$

where C_p gives the relative number of photons in the resonator which convert into photoelectrons in 1 s. In the case of ideal detection, i.e., if the losses due to diffraction and absorption are eliminated, and we have $q = 1$, $\gamma_2 = 0$, and $C_p = C$, the photocurrent spectrum vanishes at $\omega \ll C$. This vanishing implies the vanishing of the measurement noise, which limits the measurement accuracy. The parameters of the SPPS of photon counts from the output radiation, ξ_∞, and from the radiation in the resonator, $\xi_R = \zeta/2$, are related by

$$\xi_\infty = 2\xi_R C_p C^{-1} \qquad (6.15)$$

The factor of 2 stems from the anticorrelation of the instantaneous values of the numbers of photons inside and outside the resonator.[190]

Among the factors which lower the SPPS effect in more realistic models are the random nature of the events in which state 2 decays in the case $\gamma_2 \neq 0$, the deviation of the resonator frequency from the center of the gain line, and the circumstance that the detection is not ideal. These factors were taken into consideration in an analysis of SPPS in Ref. 190.

6.3. Repulsive pump statistics

Let us examine the use of the pumping mechanisms discussed in Sec. 5 to produce SPPS in generation. In the case of the excitation of atoms by an electron beam with depression of the shot noise, the variance of the number of excited atoms is given by (5.6). Correspondingly, the parameter ζ in (6.9), (6.12), and (6.13) is determined by the efficiency at which the energy of the electrons is converted into atomic excitations, i.e., $\zeta = -\eta(1 - \Gamma^2)$. For the case of ideal detection, under conditions corresponding to a maximum manifestation of SPPS, this mechanism would thus make it possible to reduce the photocurrent noise by a factor of $(1 - \eta)^{-1}$ in comparison with the noise from coherent radiation of the same intensity. The SPPS parameter of the output radiation is equal to $-\eta$ in the case in which the loss is due exclusively to the emission of radiation from the resonator.[179]

Let us examine the case in which repulsive pump statistics, associated with cooperative effects, is included. We introduce Poisson pumping to level 3, which lies above the upper working level, 2. We assume that the atoms in level 3 are capable of undergoing a pair de-excitation (Subsection 5.2). In the limiting case $\gamma_2 = 0$, an analysis leads to the following value for the parameter ζ in lasing equation[179] (6.12):

$$\zeta = -2\alpha\bar{n}_3\gamma_{3\to2}(\gamma_3 + \alpha\bar{n}_3)^{-2}; \qquad (6.15')$$

here $\gamma_{3\to2}$ is the rate of the $3\to2$ transition, γ_3 is the total decay constant of level 3, n_3 is the concentration of atoms in level 3, and α is the rate constant for pair de-excitation of atoms from level 3. The effective rate of pumping to the upper working level in (6.12) is

$$\bar{r} = \bar{r}_3\gamma_{3\to2}(\gamma_3 + \alpha\bar{n}_3)^{-1}. \qquad (6.16)$$

The maximum manifestation of SPPS is reached in the case $\gamma_3 = \gamma_{3\to2} = \alpha\bar{n}_3$, in which we have $\zeta = -1/2$. The comment which we made in Sec. 5 regarding a decrease in the SPPS parameter in other cooperative processes remains valid in the present case. In incorporating this pumping mechanism in the laser theory developed above, we should recall its applicability condition: $\tau_a \ll C^{-1}$. For cooperative effects in the emission of rare earth impurity ions in crystals, the times τ_a are typically long (10^{-2} s) and may not satisfy the relation $\tau_a \ll C^{-1}$. In order to implement this pumping mechanism we should thus seek systems with a high value of the summation constant, such that the relation $\alpha\bar{n}_3 \approx \gamma_3$ holds when the concentration $\bar{n}_3$ is not too high, and such that the relation $\gamma_3 \gg C$ holds. Examples of systems with a large value of the pair-summation constant ($\alpha \approx 10^{-10}$ cm^3/s), which could conceivably be used to implement this pumping mechanism, are given in Sec. 5.

6.4. Laser with a multiphoton loss

Herzog[189] has studied the photon statistics of a laser with an m-photon working transition and an l/photon loss. Pertinent to a study of the mechanisms for achieving SPPS is a laser with a single-photon working transition and a two-photon loss.[188,189] A two-photon loss is incorporated in the conventional quantum theory of a laser [Eq. (6.4a)] in place of a linear loss (6.4c):

$$(\dot{\rho})_{nn}^{(-)} = 2k[(n+2)(n+1)\rho_{n+2,\,n+2} - n(n-1)\rho_{nn}], \qquad (6.17)$$

where k is the two-photon loss constant.

It is legitimate to ignore the single-photon loss in comparison with the loss in a two-photon-absorbing cell if the number of photons in the resonator is large: $\bar{n}k \gg C$. At saturation of the one-photon transition, with a complete population inversion, one finds the variance of the distribution of the number of photons in the resonator to be $\overline{(\Delta n)^2} = (3/4)\bar{n}$; i.e., one finds an SPPS. The statistics of the output radiation was not studied in Refs. 188 and 189. It is important to examine this question since the discussion in Subsection 6.2 showed that the SPPS factor of the output radiation, ξ, is related to the SPPS factor in the resonator, ξ_R, by the relation $\xi = 2\xi_R C\Gamma^{-1}$ (in the case of ideal detection), where Γ^{-1} is the photon correlation time. We are thus confronted with the question of the optimum relation between the parameters C and Γ for a maximum manifestation of the SPPS of the output radiation.

7. CONCLUSION

After the first experiments[12,17-20] confirming the presence of photon antibunching and SPPS in the nonlinear resonance fluorescence of individual atoms, a weak SPPS effect was observed in the luminescence of a macroscopic system of atoms excited by an electron beam with depressed shot noise,[176] i.e., with repulsive statistics of excitation events. Active research by several groups[193] on parametric processes led to the establishment of the characteristics of the squeezed state of a field in an experiment on nondegenerate four-wave mixing in a resonator[167] (Subsection 4.3; see Ref. 194 for a detailed theory).

After this review had been accepted for publication, some new experimental results and some constructive theoretical suggestions appeared.

A characteristic of squeezing—a dip in the photocurrent noise spectrum with a depth of 12.5% of the level of the shot noise—was found[195] in nondegenerate four-wave mixing in an optical fiber (after some preliminary research[165]).

Antibunching in the nonlinear resonance fluorescence of a polyatomic system associated with the properties of anomalous correlation functions (Subsection 3.3) was observed in an experiment by Grangier *et al.*[196] An experiment by Kask *et al.*[197] demonstrated photon antibunching in the fluorescence of dye molecules.

A record squeezing was achieved by Wu *et al.*[198] in the course of degenerate parametric frequency down-conversion in a resonator. The role of a coherent pump (parametric effect; Subsection 4.2) was played by the second harmonic $\lambda_2 = 0.53\,\mu m$), produced by frequency doubling in the resonator of a Nd:YAG ring laser ($\lambda_1 = 1.06\,\mu m$). This light was coupled into a resonator containing a crystal with a quadratic optical nonlinearity ($MgO \cdot LiNbO_3$). The wave (λ_1) excited parametrically in this resonator was mixed with part of the laser beam. The spectrum of fluctuations in the difference between the photocurrents of two photodetectors, in a balanced arrangement, was measured. The maximum relative dip in the spectrum was about 50%. When the loss in the resonator (not associated with the useful output) and the efficiencies of the mixing and the detection were taken into account, a tenfold squeezing was achieved, according to estimates by Wu *et al.*[198] We should emphasize that that experiment used a sub-threshold regime of parametric excitation of a resonator mode. In the theory (Subsection 4.2), it is necessary to consider the damping and to set f_1 equal to zero in (4.6) (there is no wave at the input). The excitation of the mode (λ_1) remains at the level of quantum fluctuations. After coherent mixing with a reference signal, the resultant flux of photons turned out to be substantially sub-Poisson for certain phase relations. The depth of the dip (0.5) in the photocurrent spectrum was fixed at a frequency roughly one-fifth of the width. In other words, the SPPS parameter was essentially measured.

From the practical standpoint, coherent sources of sub-Poisson radiation—lasers—might be more efficient and more convenient (Sec. 6). The theoretical suggestions here deal primarily with regularization of the pump: a complete filling of the upper working level in each pump pulse,[190] pumping by an electron beam with depression of the shot noise of the beam, and the use of cooperative (pair) de-excitation in the pumping channel (see Refs. 199 and 200 for a further analysis). The authors of this review have shown that SPPS can also be achieved during steady-state single-mode generation by means of positive mutual correlations of the pump and the adjustable rate of in-resonator loss.[207]

The idea of suppressing pump fluctuations has been realized by a negative-feedback method in the single-mode operation of a GaAs injection laser.[201] Fluctuations in the photocurrent from the laser radiation, amplified and inverted in sign, were superimposed on the pump current. In the current fluctuation spectrum in the feedback circuit, a dip was observed in the frequency band 0–15 MHz, with a maximum relative depth of 0.81. A sub-Poisson factor of approximately this value was also found in measurements of the photocurrent pulse distribution. We wish to stress that the sub-Poisson statistics in this experiment were characteristic only of the photocurrent pulses in the feedback circuit; the photon counts in the tapped part of the laser beam were not sub-Poisson. It has been suggested that the negative-feedback principle might be used to generate sub-Poisson radiation which leaves the feedback circuit and which can be used for further (useful) transformations by means of a nondestructive quantum measurement based on the optical Kerr effect.[202]

A dip below the shot level in the intensity fluctuation spectrum of the output radiation from an injection laser was recently obtained in an experiment by Machida and Yamamoto.[208] The effect was achieved by virtue of the direct suppression of the noise in the pump current.

An intense search for sources of sub-Poisson radiation is being carried out because of the promising outlook for important practical applications. The advantages of field states with reduced fluctuations for data transmission were examined in Refs. 77, 78, and 203. The use of squeezed states for extremely precise measurements, in particular, for detecting gravitational waves, has been studied in Refs. 171 and 204, among other places. Kolobov and Sokolov[205] have shown that the use of sub-Poisson laser radiation in interference measurements would make it possible to reduce the measurement noise significantly.

The macroscopic manifestations of the new quantum properties of light which have already been demonstrated, the real possibility of lowering the noise in radiation, and important applications make further research an urgent matter.

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[1)]"The more we are together the merrier we'll be." This favorite song of the bosons[28] may not be heard (or, in this case, not seen) if the "singers" are forbidden to gather together.

[2)]This point has been mentioned in many papers, e.g., Refs. 12, 31, 34, and 62–64. It was some time ago that Glauber[4] set forth the case for that attitude toward the P representation in connection with the semiclassical description of the field which has now become generally accepted.

[3)]The inapplicability of the classical description of photon antibunching and sub-Poissonian photon statistics was analyzed by Reid and Walls[66] in connection with Bell's inequalities.[67] We will take up some related questions below.

[4)]A correct mathematical inequality can of course not be "violated" by physical experiments or arguments. We repeat that the matters involved here are differences in the structure of observables and differences in their interrelationships in classical and quantum descriptions of correlation measurements.

[5)]For one mode of a free field we would have $E_\mu^{(+)} \propto a \exp(-i\omega t)$.

[6)]Some inspiring papers for a discussion of the correspondence principle in the theory of radiation are those by Heisenberg,[98] Fermi,[99] and also Fano[100] and Glauber.[4]

[7)]The variance, however, exhibits traces of photon correlations over short times and may be greater than or less than $\langle n\rangle$.

[8)]It is possible to write $(p)^{(2)}_{+}$ in the form in (6.9) because of the balance between the gain and the loss.

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