Intensity fluctuation spectroscopy using squeezed light

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In this paper we discuss the method of intensity fluctuation spectroscopy, which is based on spectral analysis of the photocurrent correlation function of probe light scattered by a medium under study. Starting from first principles, we estimate the sensitivity of the method, taking into account the quantum nature of photoabsorption, and examine the feasibility of using it to detect probe light with nonclassical photon statistics. We show that the measurement errors for the photocurrent spectrum are determined by fourth-order correlation functions of the photocurrent, which we express in terms of TN-ordered correlation functions of the field from first to fourth order. When the fluctuation spectrum is observed by using an optical heterodyning scheme, these measurement errors can be written in terms of the Mandel spectral parameter for the recorded light $\xi(\Omega)$, which characterizes the deviation of the spectrum of photocurrent fluctuations at a frequency Ω from the Poisson level. The use of squeezed light with $\xi(\Omega)\rightarrow -1$ for this detection can lead to a considerable increase in the sensitivity of the method of intensity fluctuation spectroscopy compared with the use of coherent light in an analogous recording scheme. As an example we consider two nonlinear processes: two-photon absorption by a two-level atomic transition, and parametric conversion of light in a four-wave process via scattering by atoms with oriented internal angular momenta. We show that the use of squeezed light leads to the possibility of observing a qualitatively new effect in fluctuation spectroscopy—a correlation-induced Faraday effect. [S1063-7761(96)00509-4]

1. INTRODUCTION

Traditional methods of atomic and molecular high-resolution spectroscopy are based on analysis of the intensity, spectral content, and polarization of light transmitted or scattered by a medium under study.[1] However, in addition to these characteristics, important information about physical processes taking place in the medium is also contained in the fluctuations of intensity and polarization of the probe light. The use of information-containing properties of fluctuations in the parameters of light to analyze the state of the medium under study constitutes a new type of intensity fluctuation spectroscopy (Refs. 2–6)[1)] The method of intensity fluctuation spectroscopy is based on measuring the correlation function of the photocurrent and then writing it in terms of the correlation function of the electromagnetic field and the correlation functions of the medium susceptibility. Once we have analyzed the measurement results, we can obtain important information about the dynamics of the fluctuations and various characteristics of the relaxation process in the medium under study. In a number of cases, the fluctuation spectrum contains information that is absent from the first-order field correlation function spectrum. A further interesting feature of the method is the fact that in combination with optical heterodyning it allows us in principle to observe the spectrum of correlations of the component of scattered light with the same frequency content, i.e., averages of type $\langle aa\rangle$ and $\langle a^+a^+\rangle$, whereas a measurement of the spectral intensity gives only a measurement of the average $\langle a^+a\rangle$. Here a and a^+ are annihilation and creation operators for the field oscillator respectively.

Optical mixing spectroscopy has been successfully used to solve a range of problems, for example, investigation of the velocity distribution of molecules in liquids and gases, analysis of the properties of various randomly nonuniform media, see Refs. 5 and 6, and the study of the internal dynamics of macromolecules.[4,7] The possibility of using intensity fluctuation spectroscopy to investigate the dynamics of fluctuations in the population and orientation of an ensemble of atoms with Zeeman splitting of their levels was demonstrated in the experiments of Refs. 8, 9. A number of theoretical papers[10–14] were initiated by these experiments, whose goal was to investigate the potential of the method in atomic spectroscopy applications. Especially interesting was the use of correlation methods to investigate nonclassical statistical effects such as antibunching of photons and generation of squeezed states of light, which are caused by the nonlinear interaction of light with matter and which reflect the quantum character of the evolution of the internal state of atoms of the medium under study.

Despite the successful use of the method of intensity fluctuation spectroscopy to address a number of problems and the great potential of the method for investigating the nonlinear scattering of light by various media, its use under more widespread circumstances is restricted by insufficient sensitivity due to the large experimental errors in measuring information-bearing fluctuations. This is connected with the fact that for optically thin media the fluctuation amplitudes are proportional to the square root of the particle concentration, and not the concentration itself as is the case for an ordinary optical detection signal in the traditional spectroscopic approaches,[1] and also with the fact that the

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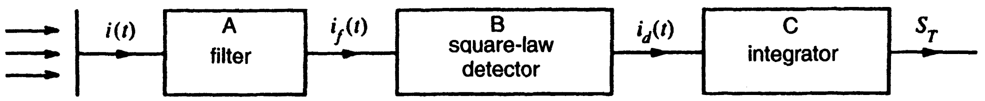


FIG. 1. Electronic detector channel. The photodetector current passes through a resonance filter tuned to frequency Ω_0. The current obtained as a result of a subsequent square-law transformation $i_d(t) \propto i_f^2(t)$ is averaged in an integrator, and S_T is the output signal of the integrator.

information-bearing fluctuations of interest must be separated from a background of shot noise and, in the general case, of Gaussian fluctuations that contain no additional information beyond that which is contained in the field spectrum, see Ref. 6. Since the average values of shot noise and Gaussian noise can always be subtracted from the total signal, the measurement potential of the method of intensity fluctuation spectroscopy is limited by fluctuations in the levels of these types of noise. Errors in the correlation measurements when the light has Gaussian statistics are discussed in Refs. 2 and 3. There it was shown that when an analyzer with a transmission bandwidth γ_f and a signal collection time T is used for the spectral analysis of photocurrent fluctuations, the ultimate sensitivity of the method is determined by the parameter $(\gamma_f T)^{-1/2}$. An analogous estimate was also made for probe light with Poisson statistics. Although this parameter can be made small, in real situations this smallness may be insufficient for precision measurements, which hinders the widespread practical application of the method.

In recent years, experimental and theoretical investigations have demonstrated the possibility of creating sources of so-called squeezed light, for which the manifestation of nonclassical statistical properties is characteristic.[15,16] The use of such sources for optical monitoring, which leads to a decrease in the photodetector noise and an increase in the measurement accuracy, was demonstrated in the experiments of Refs. 17–19. We can expect that in those cases where the quantum nature of fluctuations in the probe light parameters becomes important, improvements of this kind are possible for the method of intensity fluctuation spectroscopy as well, i.e., the estimate of the sensitivity of correlation measurements given above can in principle be improved. Furthermore, the newly discovered possibility of using the methods of optical mixing spectroscopy to observe effects based on the fundamental relations of quantum mechanics such as the Heisenberg uncertainty relation is interesting and important in its own right. The goal of the present paper is to investigate the sensitivity and to seek new applications for the method of intensity fluctuation spectroscopy, taking into account the quantum-mechanical peculiarities of the probe light.

In examining squeezed light as a possible source of probe light in intensity fluctuation spectroscopy experiments, it is necessary that the discussion be based on a fully quantum-mechanical theory of photodetection, both to calculate the signal and to estimate its errors. In this case we are required to calculate second- and fourth-order correlation functions of the photocurrent. The quantum theory of photodetection developed by Glauber,[20] (see also Ref. 21) assumes that quantum effects are manifest in real situations only when the "field-photodetector" interaction is taken into account. In this case, for a wideband photodetector the question of how to order the photocurrent operators in the second-order correlation functions of the photocurrent at different time arguments turns out to be unimportant. As the analysis we carry out below shows, a different situation arises when we calculate fourth-order correlation functions of the photocurrent. Their values depend in an important way on the ordering of the sequence of operators with different time arguments. We will show that the choice of true time ordering of the photocurrent operators leads to an expression for the measurement errors in intensity fluctuation spectroscopy in terms of *TN*-ordered correlation functions of the field from first to fourth order. We will discuss in detail the specific case of a heterodyne photodetector, which can be realized, e.g., by mixing squeezed light obtained from parametric scattering with a classical reference wave, or in using the light from a sub-Poisson laser as a detecting source. In this case we show that it is possible both to decrease the measurement errors and to observe certain qualitatively new effects.

The article has the following plan. In Secs. 2, 3 we analyze the observed quantities and errors in the method of intensity fluctuation spectroscopy, starting from classical and quantum representations of the photocurrent. In Sec. 4 the method of intensity fluctuation spectroscopy is discussed in combination with a scheme of optical heterodyning. We obtain a connection between measurement errors and the Mandel spectral parameter for the probing light. Section 5 is devoted to a discussion of concrete examples, one of which is two-photon absorption by a two-level atomic transition and parametric conversion of light in a fourth-order process mediated by scattering by oriented atoms.

2. QUANTITIES TO BE MEASURED AND THEIR ERRORS IN INTENSITY FLUCTUATION SPECTROSCOPY EXPERIMENTS

The electronic measurement sequence that is characteristic of intensity fluctuation spectroscopy experiments is shown in Fig. 1. The photodetector current $i(t)$ is subjected to a spectral analysis by a filter whose resonant frequency Ω_0 can be smoothly tuned. A current $i_d(t) \propto i_f^2(t)$ is obtained after a square-law transformation averaged over a time T. The resulting quantity S_T is the output current of an integrator or a voltage on a capacitor of the corresponding integration circuit.

The current at the output of the filter can be defined either in terms of the temporal characteristics of the filter $h(\tau)$ or its spectral transfer function $Y(\Omega)$[22,23]:

$$i_f(t)=\int_{-\infty}^{t} h(t-t_1)i(t_1)dt_1$$

$$=\int_{-\infty}^{\infty} Y(\Omega)i(\Omega)e^{-i\Omega t}\frac{d\Omega}{2\pi}, \tag{2.1}$$

where

$$i(\Omega)=\int_{-\infty}^{\infty} i(t)e^{i\Omega t}dt, \quad Y(\Omega)=\int_{-\infty}^{\infty} h(t)\theta(t)e^{i\Omega t}dt. \tag{2.2}$$

We have set the "turn-on" time of the filter equal to $-\infty$, which eliminates the effect of transient processes from our discussion. In this case the output signal S_T equals

$$S_T=\frac{1}{T}\int_0^T dt\int_{-\infty}^{t}\int dt_1dt_2h(t-t_1)h(t-t_2)i(t_1)i(t_2). \tag{2.3}$$

For a random process $i(t)$ of arbitrary type, a random variable defined in this way depends both on the initial time t_0 (which we take to be zero) and on the averaging interval T.

For a statistical ensemble of observations, the average value of the output signal $S=S(\Omega_0)=\langle S_T\rangle$ (which depends parametrically on the filter frequency Ω_0) is expressed in terms of the photocurrent correlation function:

$$G(t_1,t_2)=\langle i(t_1)i(t_2)\rangle. \tag{2.4}$$

In what follows we limit ourselves to the case of a stationary random process, for which the correlation function $G(t_1,t_2)$ is a function of the time difference $\tau=t_1-t_2$. In this case the average value S depends neither on the choice of initial time nor on the signal averaging time T:

$$S=S(\Omega_0)=\int_{-\infty}^{\infty} G(\Omega)|Y(\Omega)|^2\frac{d\Omega}{2\pi}$$

$$=\int_0^{\infty} G(\Omega)|Y(\Omega)|^2\frac{d\Omega}{\pi} \tag{2.5}$$

and is defined by the spectral density $G(\Omega)$ of the random process $i(t)$

$$G(\Omega)=\int_{-\infty}^{\infty} G(\tau)e^{i\Omega\tau}d\tau \tag{2.6}$$

and the transfer function $Y(\Omega)$ of the filter.

Let us assume the following relationships among the various temporal scales encountered in this problem: the averaging time T, the transmission bandwidth γ_f of the filter, and the characteristic time for photocurrent fluctuation correlations τ_0:

$$T\gg\gamma_f^{-1}\gg\tau_0, \tag{2.7}$$

In particular, this implies that the spectral density $G(\Omega)$ varies only slightly over a frequency interval of order γ_f, and in place of (2.5) we have

$$S=S(\Omega_0)\approx G(\Omega_0)\int_0^{\infty}|Y(\Omega)|^2\frac{d\Omega}{\pi}=\eta_0\gamma_fG(\Omega_0). \tag{2.8}$$

where the dimensionless parameter η_0

$$\eta_0=\frac{1}{\gamma_f}\int_0^{\infty}|Y(\Omega)|^2\frac{d\Omega}{\pi}\sim O(1), \tag{2.9}$$

is determined by the transfer function of the filter. Thus, the scheme shown in Fig. 1 actually measures the spectral density of the photocurrent $G(\Omega)$.

The measurement errors in the method of intensity fluctuation spectroscopy are determined by the dispersion of the quantity S_T

$$\Delta S^2=(S_T^2)-S^2, \tag{2.10}$$

which for the case of a stationary random process has the form

$$\Delta S^2=\iiint_{-\infty}^{\infty}\frac{d\Omega}{2\pi}\frac{d\Omega'}{2\pi}\frac{d\bar\Omega}{2\pi}\left(\frac{\sin(\bar\Omega T/2)}{\bar\Omega T/2}\right)^2$$

$$\times Y(\Omega+\bar\Omega/2)Y^*(\Omega-\bar\Omega/2)Y(\Omega'-\bar\Omega/2)Y^*(\Omega'$$

$$+\bar\Omega/2)(\mathcal{G}(\Omega,\Omega',\bar\Omega)-2\pi\delta(\bar\Omega)G(\Omega)G(\Omega')), \tag{2.11}$$

where the spectral function $\mathcal{G}(\Omega,\Omega',\bar\Omega)$ is defined by the Fourier transform of the photocurrent correlation function of fourth order

$$\mathcal{G}(\Omega,\Omega',\bar\Omega)$$

$$=\iiint_{-\infty}^{\infty} d\tau d\tau' d\bar\tau e^{i\Omega\tau+i\Omega'\tau'+i\bar\Omega\bar\tau}\mathcal{G}(\tau,\tau',\bar\tau),$$

$$\mathcal{G}(\tau,\tau',\bar\tau)=\langle i(t_1)i(t_2)i(t_3)i(t_4)\rangle. \tag{2.12}$$

In these relations we have taken into account that under stationary conditions the correlation function $\mathcal{G}$ depends only on three differences of the arguments, for which we have taken $\tau=t_1-t_2$, $\tau'=t_3-t_4$, and $\bar\tau=(t_1+t_2)/2-(t_3+t_4)/2$.

For the special case of Gaussian noise, where the fourth-order correlation function of the photocurrent separates into a sum of products of pairs of correlators, Eq. (2.11) is transformed into the well-known Rice relation[23]:

$$\Delta S^2=2\iint_{-\infty}^{\infty}\frac{d\Omega}{2\pi}\frac{d\Omega'}{2\pi}$$

$$\times\left(\frac{\sin[(\Omega+\Omega')T/2]}{(\Omega+\Omega')T/2}\right)^2|Y(\Omega)|^2|Y(\Omega')|^2G(\Omega)G(\Omega'). \tag{2.13}$$

However, in general, measurement errors of the spectral density $G(\Omega)$ are determined by the fourth-order correlation function (2.12) of the photocurrent, which we must be able to calculate or reliably estimate in order to determine the sensitivity of the method of intensity fluctuation spectroscopy.

We call the reader's attention to the fact that in using intensity fluctuation spectroscopy to address optical monitoring problems, the interest is not in the average value S_T of the observable itself, which is determined by Eqs. (2.8), (2.9), but rather its change due to interaction with the medium under study. When the photocurrent fluctuation spectrum in the absence of a medium is constant in the neighborhood of the frequencies under study, this implies that it is necessary to make at least two measurements for two different values of filter frequency Ω_{01} and Ω_{02}, and to compare these measurements. The difference in the values $S(\Omega_{01})$ and $S(\Omega_{02})$ can be reliably observed if it is considerably larger than the square root of the sum of the variances of the quantities S_T defined by relation (2.11) for the two resonant frequencies Ω_{01} and Ω_{02} of the filter.

3. PHOTOCURRENT CORRELATION FUNCTION

The results of the previous section were obtained by starting from classical representations of the photocurrent. As is well known,[20,21] a classical treatment does not lead to a completely correct description of the process of photodetection. The first-principles quantum theory of photodetection developed by Glauber[20] and further elaborated in a number of subsequent papers (see, e.g., Refs. 21, 15, 24) shows that in analyzing the results of a specific experiment it is important to precisely define the measured quantity and its quantum-mechanical analog. In particular, in defining the quantum correlation functions of the photocurrent it is important to address the question of ordering of the sequence of operators $i(t)$ with different time arguments. Thus, in papers that discuss correlation experiments of the type described above, see, e.g., Refs. 21, 15, it is normally assumed that the quantum-mechanical analog of the classical average $\langle i(t_1)i(t_2)\rangle$ is the autocorrelation function of the photocurrent $\langle 1/2\{i(t_1)i(t_2)\}_+\rangle$, where $\{A,B\}_+$ is an anticommutator, and the brackets $\langle\dots\rangle$ denote a quantum-mechanical averaging over the "source–field–photodetector" state. However, whereas for the pair correlation function the relation $\langle i(t_1)i(t_2)\rangle=\langle i(t_2)i(t_1)\rangle$ is satisfied as a rule[15] (because in this case a lack of commutativity can be associated only with δ-correlated shot-noise contributions to this function), i.e., the question of ordering of the photocurrent operators is not important, quantum-mechanical effects are manifest only when we take into account the "field–photodetector" interaction. A different situation arises when we compute the fourth-order correlation functions of the photocurrent, whose values depend significantly on the ordering of operators with different time arguments.

In order to define normal ordering of the photocurrent operators under the signs of statistical and quantum-mechanical averaging, we start from the fact that the integral transformation in the original relation (2.1) consists of an expression for the system (filter) response to an external perturbation (the input photocurrent) that satisfies the principle of causality. For this reason it is natural to assume that the first signal to arrive at the input of the square-law detector is the signal induced by the photocurrent at the earliest time, and to associate with the observed quantity S_T a quantum-mechanical operator of the following form:

$$S_T=\frac{1}{T}\int_0^T dt\int_{-\infty}^{t}\int dt_1 dt_2 h(t-t_1)h \times(t-t_2)T(i(t_1)i(t_2)), \tag{3.1}$$

where the photocurrents are quantum-mechanical Heisenberg operators and T denotes their time average. Since it is a quantum observable, the quantity S_T satisfies the necessary requirement of self-adjointness $S_T=S_T^\dagger$. In this case the expression for the mean-square fluctuations of S_T, as before, is determined by relations (2.11), (2.12), which contain the photocurrent correlation function $\mathcal{S}$, which now averages photocurrent operators that are time-ordered in the necessary way. Taking into account the self-adjointness of S_T, the function $\mathcal{S}$, which is a quantum generalization of (2.12), can be written in the form

$$\mathcal{S}(\tau,\tau',\bar{\tau})=\mathcal{S}(t_1,t_2,t_3,t_4) =\langle\tilde{T}(i(t_1)i(t_2))T(i(t_3)i(t_4))\rangle, \tag{3.2}$$

Here $\tilde{T}$ is the operator of anti-ordering in time. In this relation we take into account that for a stationary random process the correlation function $\mathcal{S}$ in fact depends only on the three argument differences τ, τ', $\bar{\tau}$ defined according to (2.12). However, in the general analysis of this section we will discuss the correlation function of all four temporal arguments independently, i.e., we will assume that $\mathcal{S}=\mathcal{S}(t_1,t_2,t_3,t_4)$.

The photocurrent operators $i(t)$ entering into the correlation function are expressed in terms of the operator of the number of atoms $N(t)$ ionized at time t:

$$i(t)=e\,\frac{dN(t)}{dt}=e\,\frac{d}{dt}\sum_k N_k(t). \tag{3.3}$$

Here $N_k(t)$ is a single-atom projection operator onto all the excited states that are evolving in accordance with the Heisenberg equations, while the summation with respect to label k runs over all atoms of the photodetector; e is the electron charge, which we set equal to unity in what follows $(e=1)$. Calculation of the correlation function for photocurrent is thus reduced to calculating the corresponding correlation function for the number of excited atoms, which can be done with the help of perturbation theory based on the assumptions of Glauber[20] for the model of an ideal photodetector. In this case it turns out that our approach to ordering the operators in the definition (3.2) is not the only one possible. We calculated the fourth-order correlation functions for various types of time ordering of the photocurrent operators, along with unordered and symmetrized averages. In this case, if it is approximately true that recombination of photoelectrons in the detector with the emission of photons does not occur, i.e., if the transfer of energy from the field to the detector is unidirectional, the following calculated averages coincide:

$$
\begin{aligned}
\langle\widetilde{T}(i(t_1)i(t_2))T(i(t_3)i(t_4))\rangle &= \langle T(i(t_1)i(t_2)i(t_3)i(t_4))\rangle \\
&= \langle\widetilde{T}(i(t_1)i(t_2)i(t_3)i(t_4))\rangle \\
&= \langle i(t_1)T(i(t_2)i(t_3)i(t_4))\rangle \\
&= \langle\widetilde{T}(i(t_1)i(t_2)i(t_3))i(t_4)\rangle. \qquad (3.4)
\end{aligned}
$$

A common feature of all these averages is the fact that they all correspond to various possible averages of the products of operators ordered along the contour introduced by Konstantinov and Perel' in their diagram technique.[25]

Substituting (3.3) into the definition (3.2), we can write the photocurrent correlation function in the form

$$
\mathscr{G}(t_1,t_2,t_3,t_4)=\frac{\partial}{\partial\{t\}}\sum_{k,l,m,n}K_{klmn}(t_1,t_2,t_3,t_4). \qquad (3.5)
$$

Here $\partial/\partial\{t\}=\partial/\partial t_1\,\partial/\partial t_2\,\partial/\partial t_3\,\partial/\partial t_4$, and

$$
\begin{aligned}
&K_{klmn}(t_1,t_2,t_3,t_4) \\
&\qquad=\langle\widetilde{T}(N_k(t_1)N_l(t_2))T(N_m(t_3)N_n(t_4))\rangle. \qquad (3.6)
\end{aligned}
$$

Let us break up the correlation function for the photocurrents into a sum of several terms, which describe one-, two-, three- and four-atom interactions with the field:

$$
\begin{aligned}
\mathscr{G}(t_1,t_2,t_3,t_4)&=\mathscr{G}_1(t_1,t_2,t_3,t_4)+\mathscr{G}_2(t_1,t_2,t_3,t_4) \\
&\quad+\mathscr{G}_3(t_1,t_2,t_3,t_4)+\mathscr{G}_4(t_1,t_2,t_3,t_4), \qquad (3.7)
\end{aligned}
$$

where

$$
\mathscr{G}_1(t_1,t_2,t_3,t_4)=\frac{\partial}{\partial\{t\}}\sum_{k}K_{kkkk}(t_1,t_2,t_3,t_4), \qquad (3.8)
$$

$$
\begin{aligned}
\mathscr{G}_2(t_1,t_2,t_3,t_4)=\frac{\partial}{\partial\{t\}}\sum_{k,l}{}'\,(&K_{lkkk}(t_1,t_2,t_3,t_4) \\
&+K_{klkk}(t_1,t_2,t_3,t_4) \\
&+K_{kklk}(t_1,t_2,t_3,t_4) \\
&+K_{kkkl}(t_1,t_2,t_3,t_4) \\
&+K_{llkk}(t_1,t_2,t_3,t_4) \\
&+K_{lklk}(t_1,t_2,t_3,t_4) \\
&+K_{lkkl}(t_1,t_2,t_3,t_4)), \qquad (3.9)
\end{aligned}
$$

$$
\begin{aligned}
\mathscr{G}_3(t_1,t_2,t_3,t_4)=\frac{\partial}{\partial\{t\}}\sum_{k,l,m}{}'\,(&K_{kklm}(t_1,t_2,t_3,t_4) \\
&+K_{klkm}(t_1,t_2,t_3,t_4) \\
&+K_{klmk}(t_1,t_2,t_3,t_4) \\
&+K_{lkkm}(t_1,t_2,t_3,t_4) \\
&+K_{lkmk}(t_1,t_2,t_3,t_4) \\
&+K_{lmkk}(t_1,t_2,t_3,t_4)), \qquad (3.10)
\end{aligned}
$$

$$
\mathscr{G}_4(t_1,t_2,t_3,t_4)=\frac{\partial}{\partial\{t\}}\sum_{k,l,m,n}{}'\,K_{klmn}(t_1,t_2,t_3,t_4). \qquad (3.11)
$$

The prime on the summation sign implies that we retain only terms corresponding to differing values of the summation labels: $k\neq l$, etc.

In order to express the correlation functions of the photocurrent (3.8)–(3.11) in terms of the correlation functions of the electromagnetic field, let us discuss the interaction of light with the photodetector atoms in the lowest orders of perturbation theory. The total Hamiltonian H of the system we write in the form of a sum of an unperturbed Hamiltonian H_0 that describes the independent evolution of the electromagnetic field and the detector and an operator V that describes their interaction:

$$
H=H_0+V. \qquad (3.12)
$$

Let us transform to the interaction picture and introduce the evolution operator for the complete system under discussion:

$$
\hat{S}=\hat{S}(\infty,-\infty)=T\exp\left(-\frac{i}{\hbar}\int_{-\infty}^{\infty}V(t)dt\right). \qquad (3.13)
$$

In this case the atomic correlation functions (3.6) can be written in the form

$$
\begin{aligned}
&K_{klmn}(t_1,t_2,t_3,t_4) \\
&\quad=\langle\widetilde{T}(\hat{S}^{-1}N_k^0(t_1)N_l^0(t_2))T(N_m^0(t_3)N_n^0(t_4)\hat{S})\rangle, \qquad (3.14)
\end{aligned}
$$

where the single-atom operator $N_k^0(t)$ is defined in the interaction picture and can be expressed in terms of creation operators $\Psi_s^{0\dagger}(\mathbf{r}_k,t)$ and annihilation operators $\Psi_s^0(\mathbf{r}_k,t)$ of an atom at the point $\mathbf{r}_k$ in state s, which is an eigenstate of the Hamiltonian of internal motion of the atom:

$$
N_k^0(t)=\sum_s\Psi_s^{0\dagger}(\mathbf{r}_k,t)\Psi_s^0(\mathbf{r}_k,t). \qquad (3.15)
$$

The operator $V(t)$ in the interaction picture has the following form in the dipole approximation:

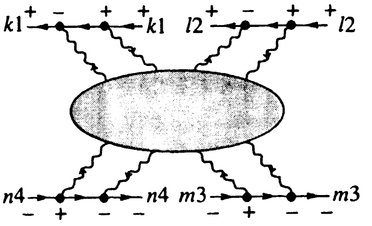
$$
V(t)=-\sum_{s,s_0}\sum_k(d^\mu)_{ss_0}E_\mu^{(-)}(\mathbf{r}_kt)\Psi_s^{0\dagger}(\mathbf{r}_kt)\Psi_{s_0}^0(\mathbf{r}_kt)+\text{H.c.} \qquad (3.16)
$$

Here $(d^\mu)_{ss_0}$ is the matrix element of the μth component of the transition dipole moment vector between the ground s_0 and excited s states, and $E_\mu^{(-)}(\mathbf{r}_k,t)$ is the operator for the negative-frequency component of the field intensity. Here we use a co(contra)variant system to describe the tensor indices and will adhere to the definition of the field frequency components used in Ref. 26: $E^{\pm}\sim e^{\pm i\omega t}$, i.e., the + and − signs are associated with photon creation and annihilation operators, respectively. The time evolution of the operators $\Psi_s^{0\dagger}(\mathbf{r}t)$ and $\Psi_s^0(\mathbf{r}t)$ is determined by the field and detector Hamiltonians: $H_0=H_d+H_f$. The Hamiltonian H_f also takes into account the interaction of the field with the scattering medium, so that the operators $E_\mu^{(\pm)}(\mathbf{r},t)$ are exact Heisenberg operators with respect to all the interactions in which the light participates except for its interaction with the detector.

Let us calculate the correlation functions $K_{klmn}(\ldots)$ (3.6), (3.14) and the photocurrent correlation functions (3.7)–(3.11) expressed in terms of them by using the Keldysh diagram technique.[27,28] In calculating the functions $K_{klmn}(\ldots)$ we make the usual assumptions corresponding to the model of an ideal Glauber photodetector.[20] The spectrum of the sensitivity function of the photodetector we assume to be considerably broader than the spectrum of the light under study, so that the process of photoabsorption can be treated as localized in time. We will also assume that the original state consists of photodetector atoms in their ground states, all of whose sublevels are uniformly occupied, and that the probability of light absorption does not depend on its polarization. We will neglect the recombination of photoelectrons and the resulting feedback of the photodetector to the incident light. In what follows, we will discuss as examples the diagram representations of the functions (3.6), (3.14) entering into the expression for the correlation functions $\mathscr{G}_4(\ldots)$ and $\mathscr{G}_3(\ldots)$, and point out distinctive features of the calculation for these examples.

Analysis of the possible diagrams shows that in the approximations discussed above the correlation function $K_{klmn}(t_1,t_2,t_3,t_4)$ with non-coinciding values of the atomic labels $k \neq l \neq m \neq n$, which determines the photocurrent correlation function $\mathscr{G}_4(\ldots)$, is given by the following graphical expression:

$$K_{klmn}(t_1, t_2, t_3, t_4) = \sum_{\{s\}}$$



(3.17)

Here the straight lines, whose ends are denoted by labels + or −, denote the Green's functions that appear in the Keldysh technique for the ground and excited states of the i-th atom, which are averages of atomic operators Ψ time-ordered in various ways:

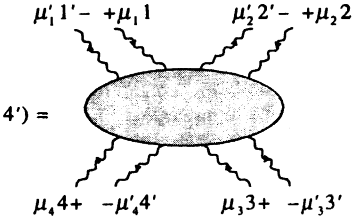
$$iG_{ss'}^{\sigma\sigma'}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=\langle T_{\sigma_1\sigma_2}(\Psi_s^0(\mathbf{r}_1t_1)\Psi_{s'}^{0\dagger}(\mathbf{r}_2t_2))\rangle, \tag{3.18}$$

where the ordering operators $T_{\sigma_1\sigma_2}$ (σ_1, $\sigma_2=\pm$) act according to the following rules: $T_{--}=T$ is the operator of time-ordering, $T_{++}=\tilde{T}$ is the operator of time-anti-ordering, T_{+-} is the identity operator, and T_{-+} is the operator of transposition. In the case of fermion statistics, Eq. (3.18) must be multiplied by -1 if the ordering operator $T_{\sigma_1\sigma_2}$ leads to a transposition of Ψ operators. In order to simplify the description we use abbreviated notations in the diagrams. For example, the label $k1$ denotes $k1=s_k$, $\mathbf{r}_k$, t_1, where s_k is an internal state of the kth atom; analogous notations are used in the remaining cases. We associate matrix elements of the dipole moment with vertices of the diagram, in accordance with the general rules of the Keldysh diagram technique.[27,28] The summation sign over s in the diagram (3.18) indicates that the summation should be carried out over all excited states of the atoms. We draw the reader's attention to the fact that when there are no excited atoms in the initial state of the photodetector, taking into account the δ-correlated nature of the process of photoabsorption for an ideal photodetector, the choice of signs for vertices and at the ends of the diagram (3.17) is the only one possible.

The internal portion of the diagram consists of a block that describes the fourth-order correlation function of the electromagnetic field, and which takes into account all of the interaction with the system "field plus source," including the interaction with the medium being probed:

$$\langle\tilde{T}(E_{\mu 1}^{(+)}(\mathbf{r}_1t_1)\ \ldots\ E_{\mu 4}^{(+)}(\mathbf{r}_4t_4))T(E_{\mu' 1}^{(-)}(\mathbf{r}_1'\,t_1'\,)\ \ldots\ E_{\mu' 4}^{(-)}(\mathbf{r}_4'\,t_4'\,))\rangle$$

$$=\mathcal{D}^{(4)}_{\{\mu,\,\mu'\}}(1,\,2,\,3,\,4;\,1',\,2',\,3',\,4')=$$



(3.19)

For an ideal photodetector, there is an analytic expression corresponding to diagram (3.17) that relates the photoresponse to the correlation function of the electromagnetic field (3.19) when the arguments and labels of the latter coincide: $\mu_i\mathbf{r}_it_i=\mu_i'\mathbf{r}_i't_i'$, where $i=1$ to 4. If we then substitute (3.17) into the photocurrent correlation function (3.11), the summation over all photodetector atoms can be replaced by integration over the surface of the photocathode, which leads to the following expression for the correlation function $\mathscr{G}_4$:

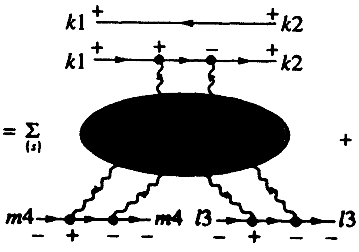
$$\begin{aligned}\mathscr{G}_4(t_1,t_2,t_3,t_4)&=\left(\frac{\zeta c}{2\pi\hbar\omega}\right)^4\int d^2r_1\ldots\int d^2r_4\\&\times\langle\tilde{T}(E_{\mu_1}^{(+)}(\mathbf{r}_1t_1)\ldots E_{\mu_4}^{(+)}(\mathbf{r}_4t_4)\\&\times T)(E_{\mu_1}^{(-)}(\mathbf{r}_1t_1)\ldots E_{\mu_1}^{(-)}(\mathbf{r}_4t_4))\rangle\\&=\left(\frac{\zeta c}{2\pi\hbar\omega}\right)^4\int d^2r_1\ldots\int d^2r_4\\&\times\mathscr{D}^{(4)}_{\{\mu,\mu\}}(1,2,3,4;1,2,3,4),\end{aligned} \tag{3.20}$$

where ω is the average frequency of the incident light, and ζ is the quantum efficiency of the photodetector. Repeated tensor indices (here defined in a Cartesian basis) imply summation.

This expression can also be obtained by starting from the well-known formulas of Glauber applied to an ideal four-atom photon detector.[20] The calculations of Ref. 20 (see also Ref. 24) were carried out for an arbitrary n-atom ideal photodetector as part of a discussion of the photoabsorption process to lowest nonvanishing order in ordinary time-dependent perturbation theory. In order to determine the probability for a photoabsorption at each of the photodetector atoms at "its own" instant of time t_i, the evolution operator for each of the atoms is also calculated at that time instant t_i. By direct comparison, it is not difficult to verify that the method of calculation used in Ref. 20 for the probability of recording n photons by an n-atom photodetector corresponds precisely to calculating the correlation function $K_{kl\ldots}(t_1,t_2,\ldots)$ with $k\neq l\neq\ldots$ defined by relation (3.14). We can also say that the physically transparent conclusion regarding the probability of an n-fold coincidence discussed by Glauber corresponds to calculating the time-ordered expression for the atomic correlation functions in the form (3.14), (3.6), and thereby is a basis for the time ordering in the expression for the quantum observable S_T; see (3.1).

Let us discuss the photocurrent correlation function $\mathscr{G}_3(t_1,t_2,t_3,t_4)$ defined by Eq. (3.10). The lowest nonvanishing order of perturbation theory in the interaction of detector atoms with the field that contributes to the atomic correlation functions $K_{kklm}(t_1,t_2,t_3,t_4)$, $K_{klkm}(t_1,t_2,t_3,t_4),\ldots$ with $k\neq l\neq m$, will be the sixth, with the main contribution coming from diagrams with one unconnected line. The correlation function $K_{kklm}(t_1,t_2,t_3,t_4)$ is determined by the following graphical expression:

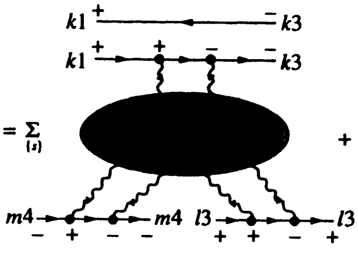
$$K_{kklm}(t_1,t_2,t_3,t_4)=\sum_{(s)}$$



$$+(k1\leftrightarrow k2), \tag{3.21}$$

where the second term denotes a diagram that differs from the first by the replacement $k1\leftrightarrow k2$. The correlation function $K_{klkm}(t_1,t_2,t_3,t_4)$ is determined by the following diagram:

$$K_{klkm}(t_1,t_2,t_3,t_4)=\sum_{(s)}$$



$$+(k1\leftrightarrow k3). \tag{3.22}$$

Note that, as in the discussion of the diagram (3.17), the choice of signs for the vertices of diagrams (3.21), (3.22) is the only one possible for an ideal photodetector. A graphical representation of the remaining four atomic correlation functions that enter into $\mathscr{G}_3(\ldots)$ can be obtained without difficulty by analogy with (3.21) and (3.22). The analytic calculations of all the diagrams that appear are straightforward and for the most part repetitive. When substituted into the photocurrent correlation function $\mathscr{G}_3(\ldots)$, the connected portions of the diagrams generate factors determined by the third-order correlation function of the electromagnetic field, and the disconnected ends lead to δ-functions in the corresponding temporal arguments.

Generalizing these discussions, we remark that the lowest nonvanishing perturbation orders with respect to interaction of detector atoms with the field for the correlation functions $\mathscr{G}_2(\ldots)$ and $\mathscr{G}_1(\ldots)$ will be fourth and second respectively. The primary contributions to the diagram expansion of the corresponding atomic correlation functions will come from diagrams with two and three disconnected atomic lines. In this case, just as in the case of $\mathscr{G}_3(\ldots)$, the connected parts of the diagrams will generate correlation functions of the electromagnetic field, while the disconnected atomic lines will lead to δ-correlated factors. Omitting simple but tedious intermediate computations, we present the final results for all the components of the photocurrent correlation function (3.8)–(3.11):

$$\mathscr{G}_1(t_1,t_2,t_3,t_4)=g_1(t_1)\delta(t_1-t_2)\delta(t_2-t_3)\delta(t_3-t_4), \tag{3.23}$$

$$\begin{aligned}\mathscr{G}_2(t_1,t_2,t_3,t_4)=&\,g_2(t_1,t_2)\delta(t_2-t_3)\delta(t_3-t_4)\\&+g_2(t_1,t_2)\delta(t_1-t_3)\delta(t_3-t_4)\\&+g_2(t_1,t_3)\delta(t_1-t_2)\delta(t_2-t_4)\\&+g_2(t_1,t_4)\delta(t_1-t_2)\delta(t_2-t_3)\\&+g_2(t_1,t_3)\delta(t_1-t_2)\delta(t_3-t_4)\\&+g_2(t_1,t_2)\delta(t_1-t_3)\delta(t_2-t_4)\\&+g_2(t_1,t_2)\delta(t_2-t_3)\delta(t_1-t_4),\end{aligned} \tag{3.24}$$

$$\begin{aligned}\mathscr{G}_3(t_1,t_2,t_3,t_4)=&\,g_3(t_1,t_3,t_4)\delta(t_1-t_2)\\&+g_3(t_1,t_2,t_4)\delta(t_1-t_3)\\&+g_3(t_1,t_2,t_3)\delta(t_1-t_4)\\&+g_3(t_1,t_2,t_4)\delta(t_2-t_3)\\&+g_3(t_1,t_2,t_3)\delta(t_2-t_4)\\&+g_3(t_1,t_2,t_3)\delta(t_3-t_4),\end{aligned} \tag{3.25}$$

$$\mathscr{G}_4(t_1,t_2,t_3,t_4)=g_4(t_1,t_2,t_3,t_4), \tag{3.26}$$

where we use the notation

$$g_n(t_1,\ldots,t_n)=\left(\frac{\zeta c}{2\pi\hbar\omega}\right)^n\int d^2r_1\ldots\int d^2r_n \times\mathscr{D}^{(n)}_{\{\mu,\mu\}}(1,\ldots,n;1,\ldots,n) \tag{3.27}$$

and define the nth order correlation function of the electromagnetic field as follows:

$$\mathscr{D}^{(n)}_{\{\mu,\mu'\}}(1,\ldots,n;1',\ldots,n')=\langle\tilde{T}(E^{(+)}_{\mu_1}(\mathbf{r}_1t_1)\ldots E^{(+)}_{\mu_n}(\mathbf{r}_nt_n)) \times T(E^{(-)}_{\mu'_1}(\mathbf{r}'_1t'_1)\ldots E^{(-)}_{\mu'_n}(\mathbf{r}'_nt'_n))\rangle. \tag{3.28}$$

In expression (3.27) we have used the same system of abbreviated notation as in Eq. (3.20).

The expressions we have obtained for the various components of the photocurrent correlation function (3.23)–(3.26) show that contributions are present in the full photocurrent correlation function (3.7) that are proportional to electromagnetic field correlation functions of various orders from first to fourth. For a real photodetector, the accuracy of the δ-correlated factors is determined by the spectral absorption bandwidth of the detector, which must be considerably wider than both the spectral content of the incident light and the frequency band of photocurrent fluctuations under study as defined by the spectrum of the correlation functions $g_n(\ldots)$. Note also that although the functions $\mathcal{G}_i(\ldots)$ with $i=1$ to 4 are determined by field correlation functions of different order, as we will see below, they give comparable contributions to the total correlation function of the photocurrent when the state of the field is close to coherent.

4. ERRORS IN MEASURING THE PHOTOCURRENT FLUCTUATION SPECTRUM BY OPTICAL HETERODYNING

When a medium is investigated by the method of intensity fluctuation spectroscopy, the amount and content of information obtained depend significantly on the choice of a specific optical scheme for detecting the probe light. The most complete information is given by polarization-sensitive correlation measurements, where the detected light is subjected to a polarization analysis, since this scheme allows us to measure not only the intensity correlations of the light, but also its polarization correlations, see Ref. 13. The use of probe light with quantum statistical properties, e.g., light in the squeezed state, imposes definite limitations on the possible methods for detection. The quantum statistical properties of squeezed light are significantly "spoiled" when the light passes through various kinds of polarization analyzers, due to partial absorption. In this paper we discuss the use of squeezed light to increase the sensitivity and extend the possibilities of the method of nonlinear correlation spectroscopy. Our scheme is based on the principle of optical heterodyning, in which light scattered by a medium is mixed with a strong coherent reference wave. In using this technique, we will assume that the light incident on the medium is in a state close to coherent, with quantum properties that are manifested only in its fluctuations. In this method the light scattered by the medium is recorded in the forward direction; therefore the reference wave is a coherent component of the light passing through the medium. Initially, the nonclassical (squeezed) light can be combined with the coherent light in a beam splitter with a high transmission coefficient, see Fig. 2.

The photocurrent correlation function $G(t_1,t_2)$ determined by Eq. (2.4), expressed in the notation (3.7), can be written in the form

$$G(t_1,t_2)=g_1(t_1)\delta(t_1-t_2)+g_2(t_1,t_2). \tag{4.1}$$

In the method of optical heterodyning, we can separate a group of high-intensity modes from the light incident on the photodetector in which the field is in a state close to coherent. The amplitude of the coherent component can be separated out in calculating the quantum averages, which formally reduces to replacing the field operator by the sum

$$\mathbf{E}(\mathbf{r}t)=\mathcal{E}(\mathbf{r}t)+\mathbf{e}(\mathbf{r}t), \tag{4.2}$$

where the first term, which is a unit operator, projects out the complex amplitudes of states that are eigenstates of the field operator (coherent modes of the reference wave), while the second term corresponds to the Heisenberg operator for field fluctuations.

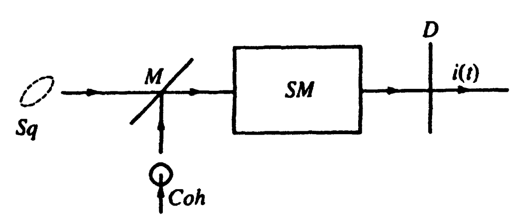


FIG. 2. Observation of the fluctuation spectrum of transmitted light using the method of optical heterodyning. The coherent reference wave (Coh) is mixed with the squeezed light (Sq) in a beam splitter (M) with a high transmission coefficient. The light passing through the scattering medium (SM) is absorbed by the photodetector D and is subject to further spectral analysis, see Fig. 1.

Taking into account (4.2), the spectrum of the function $G(\tau)$, $\tau=t_1-t_2$ determined by relation (2.6) can be written in the form

$$\begin{aligned}
G(\Omega)=&\zeta JS_0+\zeta^2(JS_0)^2 2\pi\delta(\Omega)\\
&+\zeta^2 JS_0\frac{c}{2\pi\hbar\omega}\int\frac{d^3\kappa}{(2\pi)^3}f(\boldsymbol{\kappa}_\perp)\\
&\times\{e^{-2i\theta_0}(\boldsymbol{\epsilon}^*)^{\mu_1}(\boldsymbol{\epsilon}^*)^{\mu_2}\Phi^{(--)}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)\\
&+e^{2i\theta_0}\epsilon^{\mu_1}\epsilon^{\mu_2}\Phi^{(++)}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)+\epsilon^{\mu_1}(\boldsymbol{\epsilon}^*)^{\mu_2}\\
&\times\Phi^{(+-)}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)+(\boldsymbol{\epsilon}^*)^{\mu_1}\epsilon^{\mu_2}\Phi^{(-+)}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)\}\\
=&\bar{i}^2 2\pi\delta(\Omega)+\bar{i}(1+\zeta\xi(\Omega)).
\end{aligned} \tag{4.3}$$

where S_0 is the area of the illuminated surface of the photocathode, J is the flux density of photons in the reference wave, $\bar{i}=\zeta JS_0$ is the average photocurrent, and ζ is the quantum efficiency of the photodetector.

In Eqs. (4.2) and (4.3), we take as a reference wave a wave packet with negative-frequency components of the following form:

$$\mathcal{E}^{(-)}_\mu(\mathbf{r}t)=\epsilon_\mu\int\frac{d^2k_\perp}{(2\pi)^2}\mathcal{E}(\mathbf{k}_\perp)\exp\left\{i\mathbf{k}_\perp\rho-i\omega\left(t-\frac{z}{c}\right)\right\}, \tag{4.4}$$

which defines a monochromatic light signal propagating along the z axis with a narrow range of wave vectors and a complex polarization vector $\boldsymbol{\epsilon}$. Let us neglect the effect of diffraction, i.e., we will not take into account the difference between longitudinal components of the wave vectors. The distribution of transverse wave vector components $\mathbf{k}_\perp$ is determined by the following packet amplitude function:

$$\begin{gathered}
\mathcal{E}(\mathbf{k}_\perp)=|\mathcal{E}(\mathbf{k}_\perp)|e^{i\theta_0},\\
|\mathcal{E}(\mathbf{k}_\perp)|^2=JS_0 f(\mathbf{k}_\perp)\frac{2\pi\hbar\omega}{c},\\
\int\frac{d^2k_\perp}{(2\pi)^2}f(\mathbf{k}_\perp)=1.
\end{gathered} \tag{4.5}$$

The functions $\Phi^{\sigma_1\sigma_2}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)$ (σ_1; $\sigma_2=\pm$) are spacetime Fourier components of the normal ($\sigma_1=\pm$; $\sigma_2=\mp$) and anomalous ($\sigma_1=\pm$; $\sigma_2=\pm$) correlation functions $\Phi^{\sigma_1\sigma_2}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)$ of the Heisenberg field intensity operators

$$\Phi^{(\sigma_1\sigma_2)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=\langle T_{\sigma_1\sigma_2}(e^{(\sigma_1)}_{\mu_1}(\mathbf{r}_1t_1)e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2))\rangle, \tag{4.6}$$

which we assume depend only on the coordinate differences $\tau=t_1-t_2$, $\rho=\mathbf{r}_1-\mathbf{r}_2$ after separating out the rapidly oscillating phase factors:

$$\Phi^{(\sigma_1\sigma_2)}_{\mu_1\mu_2}(\boldsymbol{\kappa},\Omega)=\int_{-\infty}^{\infty}d\tau\int d^3\rho\,\exp(i\Omega\tau-i\boldsymbol{\kappa}\rho)\times\exp\left\{-i\sigma_1\omega\left(t_1-\frac{z_1}{c}\right)-i\sigma_2\omega\left(t_2-\frac{z_2}{c}\right)\right\}\Phi^{(\sigma_1\sigma_2)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2). \tag{4.7}$$

The second equation in (4.3) defines the Mandel spectral parameter $\xi(\Omega)$, which measures the deviation of the fluctuation level from the shot-noise limit in the region of the spectrum under discussion. The spectral dependence of the Mandel parameter is determined by the corresponding dependences of the field correlation functions (4.6), and is a manifestation of quantum beating components of the field intensity in the photocurrent spectrum against the background of the classical reference wave. In accordance with relations (2.8), (2.9) (see also the discussion at the end of Sec. 2), we conclude that measurement of the Mandel parameter $\xi(\Omega)$ in the vicinity of the filter resonance frequency $\Omega\simeq\Omega_0$ corresponds to measuring the observable $S=S(\Omega_0)$:

$$S=S(\Omega_0)=\eta_0\gamma_f\bar{i}(1+\zeta\xi(\Omega_0)), \tag{4.8}$$

where the parameter η_0 is determined by Eq. (2.9). In this case, in order that the uncorrelated contribution $\bar{i}^2 2\pi\delta(\Omega)$ to the spectrum (4.3) arising from the product of photocurrent averages in the correlation function be absent from the output signal determined by the integral (2.5), it is necessary for the following condition to hold:

$$\bar{i}|Y(0)|^2\simeq\bar{i}\left(\frac{\gamma_f}{\Omega_0}\right)^4\ll\gamma_f, \tag{4.9}$$

which is a criterion for ideality of the filter that notches out the neighborhood of zero frequency. Our estimate of the spectral transfer function in (4.9) corresponds to a filter with temporal characteristics of the following form: $h(\tau)=\exp(-\gamma_f\tau)\cos(\Omega_0\tau)$. In this section we will assume in our analysis that the important condition (4.9) is satisfied.

The results of the previous section show that in order to calculate the measurement error of the observable S_T, whose variance ΔS^2 is determined by Eqs. (2.10), (2.11), (3.2), it is necessary to compute the correlation functions for the detected field to all orders from first to fourth; see Eqs. (3.7), (3.23)–(3.28). In this case, using the expansion (3.7) for the fourth-order photocurrent correlation function, we can make an analogous expansion for the variance (2.10):

$$\Delta S^2=\Delta S_1^2+\Delta S_2^2+\Delta S_3^2+\Delta S_4^2, \tag{4.10}$$

in which each term directly corresponds to one of the contributions (3.23)–(3.26) to the photocurrent correlation function. Let us analyze these contributions still further, taking into account distinctive features of the heterodyne detection system.

The contribution ΔS_1^2 to the variance is determined from the photocurrent correlation function (3.23), and consequently from the first-order correlation function of the field. The AC component is proportional to the first power of the heterodyne wave intensity, and in the case under discussion here of a strong reference wave it is small, of order $\gamma_f/\bar{i}\ll 1$ compared to terms that are second order in the intensity, which appear in $\Delta S_2^2-\Delta S_4^2$. In what follows we will neglect the contribution of ΔS_1^2 to the total variance.

The variance ΔS_2^2 is determined by a subgroup of the set of terms that enter into the correlation function $\mathcal{G}_2$; see (3.24). These terms are in turn determined by the second-order correlation functions of the detected field. In isolating the principal contribution to the variance ΔS_2^2 we can discard small corrections that appear because of the incoherent component of the field, and save only terms that involve the reference wave. In other words, we can ignore whatever correlations there are in the field in calculating ΔS_2^2. Taking into account the partial compensation of the second term entering into (2.11), ΔS_2^2 has the form

$$\Delta S_2^2=2\eta_1\frac{\gamma_f}{T}(\zeta S_0J)^2; \tag{4.11}$$

here we have introduced the dimensionless parameter η_1:

$$\eta_1=\frac{1}{\gamma_f}\int_{-\infty}^{\infty}|Y(\Omega)|^4\frac{d\Omega}{2\pi}, \tag{4.12}$$

which depends on the type of filter. Note that in deriving (4.11) we have discarded a small contribution proportional to the square of the filter transfer function evaluated at zero frequency $|Y(0)|^2$.

The variance ΔS_3^2 is caused by the contributions of six terms (3.25), which are determined by third-order field correlation functions. In these correlation functions terms are present that do not depend on time, proportional to the third power of the reference wave intensity J^3, and also terms proportional to δJJ^2, where δJ is the contribution to the photon flux density from the incoherent component of the light. However, all the terms of this kind can be ignored in ΔS_3^2, first of all because of the partial compensation by analogous terms contained in the product of second-order correlation functions of the photocurrent in (2.11), and secondly by virtue of the fulfillment of condition (4.9), which eliminates the effect of the neighborhood of zero frequency on the photocurrent fluctuation spectrum. The primary contribution to ΔS_3^2 comes from those terms that are proportional to the square of the reference wave intensity J^2 and which also contain the correlation functions of the incoherent component of the field (4.6), (4.7). Analysis of these terms shows that the corresponding contribution to the variance can be directly expressed in terms of the Mandel parameter:

$$\Delta S_3^2=4\eta_1\frac{\gamma_f}{T}(\zeta S_0J)^2\zeta\xi(\Omega_0), \tag{4.13}$$

where we have discarded small terms proportional to $|Y(0)|^2$.

Let us discuss the contribution ΔS_4^2 to the variance, which is determined by the photocurrent correlation function $\mathcal{G}_4$. It is easy to see that terms proportional to the fourth power of the reference wave intensity J^4 are completely compensated by the corresponding terms contained in the product of second-order photocurrent correlation functions in (2.11). An analogous compensation also takes place for four of the time-independent terms proportional to $\delta J J^3$. Other terms in the expansion of $\mathcal{G}_4$ in powers of the heterodyne wave intensity, which are third order in J, turn out to be proportional to the linear contribution to the second-order field correlation function; see the expansion (4.3). These terms are either cancelled by analogous terms entering into the product of second-order photocurrent correlation functions, or are unimportant due to condition (4.9), because the corresponding contribution to the variance is proportional to the fourth power of the filter transfer function at zero frequency $|Y(0)|^4$.

A tedious analysis of the contributions to ΔS_4^2 from all the terms of the expansion of $\mathcal{G}_4$ that are second order in the heterodyne intensity shows that only sixteen of them are unrelated to spectral fluctuations in the neighborhood of zero frequency and are not cancelled by terms of the same order entering into the product of pairwise photocurrent correlation functions in (2.11). These terms are determined by all possible correlation functions of the incoherent field components of the following form:

$$\begin{aligned}\Phi^{(\sigma_1\sigma_2\sigma_3\sigma_4)}_{\mu_1\mu_2\mu_3\mu_4}&(\mathbf{r}_1t_1,\mathbf{r}_2t_2,\mathbf{r}_3t_3,\mathbf{r}_4t_4)\\ &=\langle T_{\sigma_1\sigma_2\sigma_3\sigma_4}(e^{(\sigma_1)}_{\mu_1}(\mathbf{r}_1t_1)e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2)e^{(\sigma_3)}_{\mu_3}(\mathbf{r}_3t_3)e^{(\sigma_4)}_{\mu_4}\\ &\quad\times(\mathbf{r}_4t_4))\rangle,\end{aligned} \tag{4.14}$$

where the operator $T_{\sigma_1\sigma_2\sigma_3\sigma_4}$ orders field operators along the Konstantinov–Perel' contour[25]: the signs $\sigma_i=\pm$ correspond to positioning the operators on the upper and lower edges of the contour in a fashion corresponding to time anti-ordering or time ordering. In this case the operators that appear on the upper edge of the contour always come first in the product.

In this paper we are primarily interested in the case of probe light that is in the squeezed state. For squeezed light obtained from parametric scattering by a crystal, the fourth-order correlation function (4.14) breaks up into a sum of pairwise products of the averages (4.6). Physically, the reasons for this factorization are quite obvious. In all the processes in which two-mode squeezed light is generated, the photons are created in pairs, so that only photons of a given pair can be correlated and no other correlations between different pairs can exist. Thus, we can say that the probe light possesses no four-photon correlations that do not factor into products of pairwise correlations. Four-photon correlations can arise when the interaction between the light and the medium under study is mediated by processes like eight-wave mixing, caused, e.g., by successive scattering of four photons by a single atom. However, the probability of such processes, which is proportional to the number of scatterers N in the medium, is much smaller than the probability of two successive four-wave processes taking place on different atoms, which is proportional to N^2. Neglecting these multiphoton processes, we can approximately assume that

$$\begin{aligned}\langle T_{\sigma_1\sigma_2\sigma_3\sigma_4}&(e^{(\sigma_1)}_{\mu_1}(\mathbf{r}_1t_1)e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2)e^{(\sigma_3)}_{\mu_3}(\mathbf{r}_3t_3)e^{(\sigma_4)}_{\mu_4}(\mathbf{r}_4t_4))\rangle\\ &\approx\langle T_{\sigma_1\sigma_2}(e^{(\sigma_1)}_{\mu_1}(\mathbf{r}_1t_1)e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2))\rangle\langle T_{\sigma_3\sigma_4}(e^{(\sigma_3)}_{\mu_3}(\mathbf{r}_3t_3)e^{(\sigma_4)}_{\mu_4}\\ &\quad\times(\mathbf{r}_4t_4))\rangle+\langle T_{\sigma_1\sigma_3}(e^{(\sigma_1)}_{\mu_1}(\mathbf{r}_1t_1)e^{(\sigma_3)}_{\mu_3}(\mathbf{r}_3t_3))\rangle\\ &\quad\times\langle T_{\sigma_2\sigma_4}(e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2)e^{(\sigma_4)}_{\mu_4}(\mathbf{r}_4t_4))\rangle+\langle T_{\sigma_1\sigma_4}(e^{(\sigma_1)}_{\mu_1}\\ &\quad\times(\mathbf{r}_1t_1)e^{(\sigma_4)}_{\mu_4}(\mathbf{r}_4t_4))\rangle\langle T_{\sigma_2\sigma_3}(e^{(\sigma_2)}_{\mu_2}(\mathbf{r}_2t_2)e^{(\sigma_3)}_{\mu_3}(\mathbf{r}_3t_3))\rangle.\end{aligned} \tag{4.15}$$

Substituting this expansion into the expression for the correlation function $\mathcal{G}_4$, we can write ΔS_4^2 in the following form:

$$\Delta S_4^2=2\eta_1\,\frac{\gamma_f}{T}\,(\zeta S_0 J)^2(\zeta\xi(\Omega_0))^2. \tag{4.16}$$

We call the reader's attention to the fact that Eq. (4.15) has the same form as for Gaussian light. In this case, for a large number of scatterers N the neglected terms that describe higher-order correlations are small, of order $1/N$. However, in contrast to the purely Gaussian statistics, for the case of squeezed light the nonzero contribution to (4.15) comes from anomalous correlation functions. In this case the contribution ΔS_4^2 to ΔS^2, like the contribution ΔS_3^2, is determined by the Mandel spectral parameter $\xi(\Omega)$ at filter frequency Ω_0, which for squeezed light can be negative.

Combining the results (4.11), (4.13), (4.16), we obtain for the total variance (4.10) the following expression:

$$\Delta S^2=2\eta_1\,\frac{\gamma_f}{T}\,(\zeta S_0 J)^2(1+\zeta\xi(\Omega_0))^2. \tag{4.17}$$

This relation, which is valid in the limit $\gamma_f T\rightarrow\infty$, can be used to estimate the sensitivity of measurements of the Mandel parameter by optical heterodyning. Let us write the Mandel parameter in the form

$$\xi(\Omega)=\xi_0+\delta\xi(\Omega), \tag{4.18}$$

where ξ_0 is its value for light incident on the medium. This value is assumed to be constant in the spectral range of interest to us, where $\delta\xi(\Omega)$ is a small correction due to contributions from scattered light. Using relations (4.8), (4.17) and taking into account the comments we made at the end of the second section on the need for at least two independent measurements in order to determine the change in the Mandel parameter in the medium under study, we obtain the following estimate for the limiting value of $\delta\xi$ that can be observed by this method:

$$\delta\xi\simeq\frac{2\sqrt{\eta_1}}{\zeta\eta_0}\,\frac{1}{(\gamma_f T)^{1/2}}\,(1+\zeta\xi_0). \tag{4.19}$$

This relation shows that the sensitivity of measurements of the information-bearing component of the photocurrent spectral density can be greatly improved if the probe light is squeezed, i.e., the Mandel parameter $\xi_0 \to -1$ for the light incident on the medium, and the photodetector efficiency $\zeta \to 1$.

It is interesting to note that the "improvement" factor $1+\zeta\xi$ enters into Eq. (4.19) to first order. When squeezed light is used in linear spectroscopy to measure a small absorption coefficient, the analogous "improvement" factor enters to the 1/2 power; see Ref. 19. Thus, the use of squeezed light in nonlinear correlation spectroscopy can be considerably more effective. However, it is important to note that that the relations between measured quantities (e.g., the Mandel parameter) and the characteristics of the scattering medium are specific to correlation spectroscopy and are not universal, and in general require some additional analysis. The choice of an optimal experimental geometry depends critically in this case on the nature of the scattering medium and the type of scattering. We will illustrate these points in the next section for the example of nonlinear scattering of light by an atomic medium.

5. CORRELATION SPECTROSCOPY OF AN ATOMIC MEDIUM

Equation (4.3) relates the photocurrent statistics to the correlation characteristics of the field. The latter enter into this expression through the normal $(\Phi^{(+-)}_{\mu_1\mu_2}, \Phi^{(-+)}_{\mu_1\mu_2})$ and anomalous $(\Phi^{(--)}_{\mu_1\mu_2}, \Phi^{(++)}_{\mu_1\mu_2})$ correlation functions of the field intensity, which are defined by Eqs. (4.6), (4.7). In what follows, we will discuss the case of a single polarization component, and introduce the following functions:

$$\begin{aligned}
&\Phi^{(--)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=(\epsilon^*)^{\mu_1}(\epsilon^*)^{\mu_2}\Phi^{(--)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2),\\
&\Phi^{(++)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=\epsilon^{\mu_1}\epsilon^{\mu_2}\Phi^{(++)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2),\\
&\Phi^{(+-)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=\epsilon^{\mu_1}(\epsilon^*)^{\mu_2}\Phi^{(+-)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2),\\
&\Phi^{(-+)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=(\epsilon^*)^{\mu_1}\epsilon^{\mu_2}\Phi^{(-+)}_{\mu_1\mu_2}(\mathbf{r}_1t_1,\mathbf{r}_2t_2), \qquad (5.1)
\end{aligned}$$

which satisfy the symmetry relations

$$\begin{aligned}
&\Phi^{(++)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=[\Phi^{(--)}(\mathbf{r}_2t_2,\mathbf{r}_1t_1)]^*,\\
&\Phi^{(++)}(\boldsymbol{\kappa},\Omega)=[\Phi^{(--)}(\boldsymbol{\kappa},\Omega)]^*,\\
&\Phi^{(-+)}(\mathbf{r}_1t_1,\mathbf{r}_2t_2)=\Phi^{(+-)}(\mathbf{r}_2t_2,\mathbf{r}_1t_1),\\
&\Phi^{(-+)}(\boldsymbol{\kappa},\Omega)=\Phi^{(+-)}(-\boldsymbol{\kappa},-\Omega). \qquad (5.2)
\end{aligned}$$

These relations show that only two functions are independent; we pick $\Phi^{(--)}$ and $\Phi^{(+-)}$.

For quasimonochromatic light propagating in the z direction, when diffraction effects are neglected this pair of correlation functions can be represented as follows:

$$\begin{aligned}
\Phi^{(--)}(\boldsymbol{\kappa},\Omega)&=(2\pi)^2\hbar\omega\delta(c\kappa_z\\
&\quad-\Omega)e^{2i\theta(\boldsymbol{\kappa}_\perp,\Omega)}r(\boldsymbol{\kappa}_\perp,\Omega),\\
\Phi^{(+-)}(\boldsymbol{\kappa},\Omega)&=(2\pi)^2\hbar\omega\delta(c\kappa_z-\Omega)\lambda(\boldsymbol{\kappa}_\perp,\Omega), \qquad (5.3)
\end{aligned}$$

taking into account that in general the phase of the anomalous correlation function θ can also depend on the transverse wave vector $\boldsymbol{\kappa}_\perp$ and on frequency Ω: $\theta=\theta(\boldsymbol{\kappa}_\perp,\Omega)$. It is convenient to introduce a generalized spacetime spectral Mandel parameter

$$\begin{aligned}
\xi(\boldsymbol{\kappa}_\perp,\Omega)&=\lambda(\boldsymbol{\kappa}_\perp,\Omega)+\lambda(-\boldsymbol{\kappa}_\perp,-\Omega)\\
&\quad+2\cos 2(\theta(\boldsymbol{\kappa}_\perp,\Omega)-\theta_0)r(\boldsymbol{\kappa}_\perp,\Omega), \qquad (5.4)
\end{aligned}$$

in terms of which we can write the spectral Mandel parameter defined by Eq. (4.3) as follows:

$$\xi(\Omega)=\int\frac{d^2\kappa_\perp}{(2\pi)^2}f(\boldsymbol{\kappa}_\perp)\xi(\boldsymbol{\kappa}_\perp,\Omega). \qquad (5.5)$$

In this expression, the integration runs over possible values of the transverse heterodyne wave vectors $\boldsymbol{\kappa}_\perp$, which we assume are considerably smaller than the limiting value $\kappa_{coh}\sim(k/L)^{1/2}$, where L is the length of the scattering medium. The quantity κ_{coh}^{-1} determines the ultimate spatial scale for coherence of the scattered light that allows phase matching of light scattered by different volumes of the medium. This scale can also be interpreted as the ultimate spatial scale over which low-noise measurements are possible, see Ref. 29. Because there is a natural limit on the possible values of the parameter $\xi(\Omega)>-1$, the real positive-definite functions $r=r(\boldsymbol{\kappa}_\perp,\Omega)$ and $\lambda=\lambda(\boldsymbol{\kappa}_\perp,\Omega)$ satisfy the following inequality:

$$\lambda(\boldsymbol{\kappa}_\perp,\Omega)+\lambda(-\boldsymbol{\kappa}_\perp,-\Omega)-2r(\boldsymbol{\kappa}_\perp,\Omega)>-1. \qquad (5.6)$$

Thus, a measurement of the Mandel parameter $\xi(\Omega)$ is in fact a collection of measurements of the three quasi-independent quantities λ, θ, r. The effect of the scattering medium reduces to modifying these quantities as follows: λ, θ, $r \to \lambda'$, θ', r'. For squeezed light these parameters have a clear interpretation as quantities that specify the eccentricity and orientation of the ambiguity ellipse in the complex amplitude plane, within which quantum fluctuations take place in the amplitude of the negative-frequency components of the field intensity at frequencies $\boldsymbol{\kappa}_\perp$, Ω. In what follows, we will refer to this as the squeezing ellipse for brevity.

The evolution of the correlation functions (5.1) and the change in the parameters λ, θ, r can be described by the optical transport equations for a nonlinear medium. However, these equations do not have a universal form, since they depend considerably on the specific type of nonlinearity being discussed. As an example, in what follows we will discuss two-photon absorption by a two-level atomic transition and parametric conversion of light in a four-wave scattering process mediated by atoms polarized with respect to internal angular momentum.

The problem of propagation of light with arbitrary quantum statistical properties through a multiphoton and, in special cases, two-photon absorber was studied in Ref. 30. In Fig. 3 we show the results of a calculation of the Mandel parameter for resonant two-photon absorption as a function of the gain of the medium K. The calculations were made for the spectral region near zero frequency $\Omega\sim 0$, which in reality corresponds to measurement of the photocurrent spectrum in a band defined by the relaxation times in the problem. The two curves in Fig. 3 correspond to coherent input light with

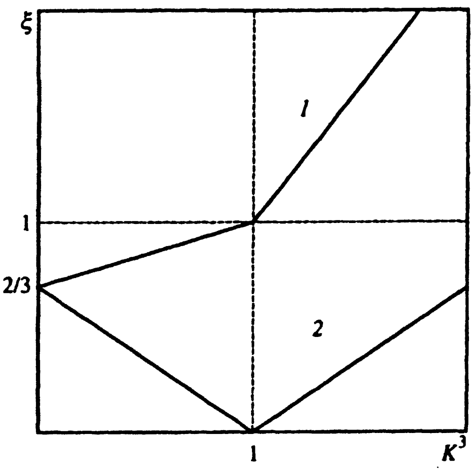


FIG. 3. Result of calculating the Mandel parameter $\xi(\Omega)$ in the low-frequency region as a function of the gain coefficient K of the medium for resonant two-photon absorption at the transition $2\omega_0$ according to the data of Ref. 30. Curves 1 and 2 correspond to probing by coherent and squeezed light.

Poisson statistics $\xi_0=0$ (curve 1) and sub-Poisson squeezed light with $\xi_0=-1$ (curve 2). These functions show that the changes in the Mandel parameter caused by the medium for weak absorption ($K\sim 1$) are first order in both cases. The specific expression that connects these changes with the macroscopic characteristics of the medium were given in Ref. 30. For us, the important thing is that when the probe is resonant or close to resonant, these measurements are determined by changes in the parameters λ and r, and do not involve the angle θ, which characterizes the orientation of the squeezing ellipse. If the two-photon absorber is used as an experimental setup to measure these characteristics by intensity fluctuation spectroscopy, then according to the results of Sec. 4, the use of squeezed light should increase the sensitivity of the scheme for measuring small changes in $\delta\xi$ in a significant way. This result can be clearly demonstrated by geometry. When the medium is probed by classical light using intensity fluctuation spectroscopy, rather small changes in the Mandel parameter are measured by identifying rather small distortions in the Gaussian ambiguity circle, which corresponds to coherent light. Of course, the problem with making such measurements is that we must isolate these small distortions against a background of random statistical versions of the original ambiguity circle. It is considerably easier to observe manifestations of two-photon absorption based on changes in the statistics of ideally squeezed input light. In this case, observing a change in the Mandel parameter corresponds to observing an increase (or appearance) of the small axis of the squeezing ellipse, and in the limit of "perfect squeezing" and an ideal photodetector there are no accompanying background fluctuations; see (4.19).

The second example—parametric conversion of light in a four-wave process mediated by a medium consisting of atoms that are oriented with respect to internal angular momentum—was discussed in Ref. 14. Transport equations that describe the evolution of the correlation functions (5.1) were derived in Ref. 14 for the case where the lower state of the driven atomic transition was the ground state. This state, which did not undergo relaxation during the motion of the atom within the beam, was split into a group of Zeeman sublevels by an external magnetic field $\mathbf{H}_0$ directed perpendicular to the probe light. The correlation function (5.1) in the medium acquires a dependence on the coordinate z:

$$\Phi^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa},\Omega)\to\Phi^{(\sigma_1\sigma_2)}(\boldsymbol{\kappa},\Omega;z), \tag{5.7}$$

and the transport equations[14] that describe its variation with z can be used when the frequency Ω is close to the frequency of the Zeeman splitting Ω_0 or twice that frequency $2\Omega_0$. We will use these equations for the case where the probe light is circularly polarized and quasiresonant with the atomic transition $j=1/2\to j'=1/2$, where j, j' are angular momentum quantum numbers for the ground and excited states; the level scheme and excitation geometry are shown in Fig. 4.

After transforming (5.1) using Eqs. (20), (21) of Ref. 14, the variation of the correlation functions $\Phi^{(--)}(\boldsymbol{\kappa},\Omega;z)$ and $\Phi^{(+-)}(\boldsymbol{\kappa},\Omega;z)$ when $\Omega\sim\Omega_0$ can be described by the equations

$$\begin{aligned}\frac{\partial}{\partial z}\Phi^{(--)}(\boldsymbol{\kappa},\Omega;z)={}&-\frac{1}{2}n_0[\sigma(\omega+\Omega)+\sigma(\omega\\&-\Omega)]\Phi^{(--)}(\boldsymbol{\kappa},\Omega;z)-e^{-2i\theta_0(z)}\\&\times[\chi(\boldsymbol{\kappa},\Omega;z)+\chi(-\boldsymbol{\kappa},\\&-\Omega;z)]\Phi^{(--)}(\boldsymbol{\kappa},\Omega;z)\\&-\chi(\boldsymbol{\kappa},\Omega;z)\Phi^{(+-)}(\boldsymbol{\kappa},\Omega;z)\\&-\chi(-\boldsymbol{\kappa},-\Omega;z)\Phi^{(-+)}(\boldsymbol{\kappa},\Omega;z)\\&+(2\pi)^2\hbar\omega\delta(c\kappa_z-\Omega)\\&\times R^{(--)}(\boldsymbol{\kappa},\Omega;z),\end{aligned} \tag{5.8}$$

$$\begin{aligned}\frac{\partial}{\partial z}\Phi^{(+-)}(\boldsymbol{\kappa},\Omega;z)={}&-n_0\sigma'(\omega-\Omega)\Phi^{(+-)}(\boldsymbol{\kappa},\Omega;z)\\&-[e^{-2i\theta_0(z)}\chi(-\boldsymbol{\kappa},-\Omega;z)\\&+e^{2i\theta_0(z)}\chi^*(-\boldsymbol{\kappa},-\Omega;z)]\\&\times\Phi^{(+-)}(\boldsymbol{\kappa},\Omega;z)-\chi(-\boldsymbol{\kappa},-\Omega;z)\\&\times\Phi^{(++)}(\boldsymbol{\kappa},\Omega;z)-\chi^*(-\boldsymbol{\kappa},-\Omega;z)\\&\times\Phi^{(--)}(\boldsymbol{\kappa},\Omega;z)+(2\pi)^2\hbar\omega\delta(c\kappa_z\\&-\Omega)\Lambda^{(+-)}(\boldsymbol{\kappa},\Omega;z).\end{aligned} \tag{5.9}$$

Here n_0 is the concentration of atoms, while $\sigma(\omega)$ is the complex cross section that describes linear absorption and dispersion of quasiresonant light at a frequency ω:

$$\begin{aligned}\sigma(\omega)&=\sigma'(\omega)+i\sigma''(\omega)\\&=\frac{4\pi\omega}{3\hbar c}\frac{|d_{jj'}|^2}{2j+1}\frac{1}{-i(\omega-\omega_0)+\gamma/2},\end{aligned} \tag{5.10}$$

where $d_{jj'}$ is the reduced matrix element of the dipole moment for the $j\to j'$ transition, ω_0 is the frequency of the unperturbed optical atomic transition, and γ is the natural atomic linewidth. We will neglect Zeeman mixing of the

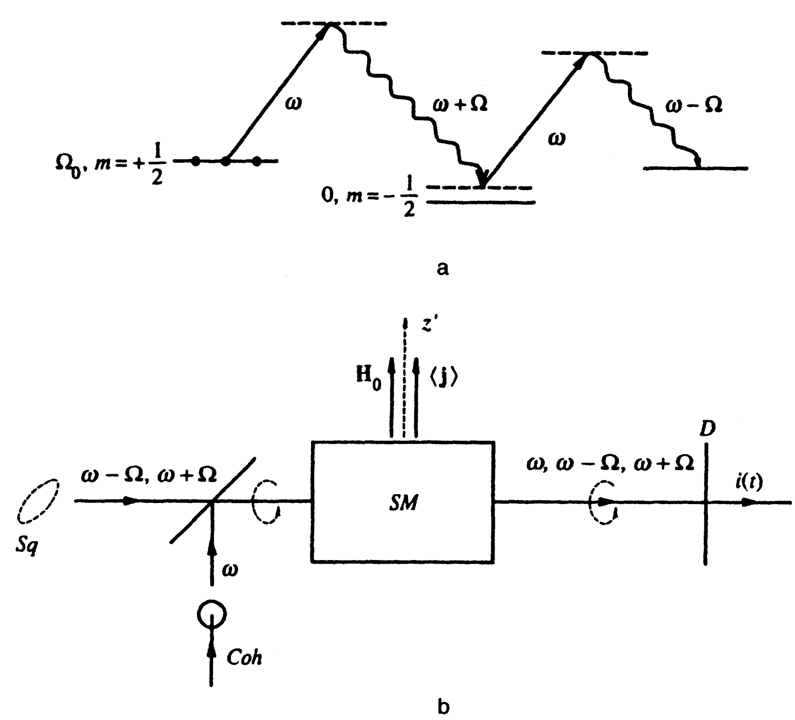


FIG. 4. Scheme of working levels (a) and experimental geometry for parametric conversion of light by an ensemble of oriented atoms (b). The average orientation of the atomic angular momenta $\langle\mathbf{j}\rangle$ is directed along an external magnetic field $\mathbf{H}_0$ and corresponds to preferentially population of one of the Zeeman sublevels, for example Ω_0, $m=1/2$. The light that illuminates the scattering medium (SM) has right-handed circular polarization, and is a superposition of the coherent component (Coh) at frequency ω and conjugate modes of squeezed light (Sq) at frequencies $\omega-\Omega$, $\omega+\Omega$. The light absorbed by the photodetector D is subject to further spectral analysis; see Fig. 1.

ground-state sublevels in this expression, assuming that $|\omega-\omega_0|\gg\gamma\gg\Omega_0$. The phase $\theta_0(z)=\theta_0-(1/2)n_0\sigma''(\omega)z$ is that of the reference wave propagating in the medium, which is the pump wave in the parametric process. The four-wave interaction of the pump wave $\mathbf{k}\omega$ with a pair of phase-conjugate modes $\mathbf{k}\pm\boldsymbol{\kappa}$, $\omega\pm\Omega$ is measured by the constant $\chi(\boldsymbol{\kappa},\Omega;z)$, while $R^{(--)}(\boldsymbol{\kappa},\Omega;z)$ and $\Lambda^{(+-)}(\boldsymbol{\kappa},\Omega;z)$ represent bulk sources of parametric and Raman scattering. The explicit form of these parameters is given in Ref. 14. Here we give only their asymptotic expressions, which are valid under the conditions $\Omega_0\gg|\Omega-\Omega_0|\gg\bar{\tau}^{-1}$, where $\bar{\tau}$ is the average time of flight of an atom through the probing beam:

$$\chi(\boldsymbol{\kappa},\Omega;z)e^{-2i\theta_0(z)}\approx -R^{(--)}(\boldsymbol{\kappa},\Omega;z)e^{-2i\theta_0(z)}\approx i\tilde{\chi}(\Omega),$$

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

$$\Lambda^{(+-)}(\boldsymbol{\kappa},\Omega;z)\rightarrow 0, \tag{5.11}$$

the parameter $p=\langle j_{z'}\rangle/j$ characterizes the degree of orientation of the atoms in the direction of the magnetic field, see Fig. 4. It is important to note that the asymptotic relations (5.11) are valid only when $p\neq 0$, i.e., only when the light interacts parametrically with the nonequilibrium atomic ensemble.

The solution to the system of Eq. (5.9) for an optically thin medium reduces to iteration. Transforming these equations to a form that describes the change in the parameters λ, θ, r when the light probes a transmission band $\sigma''(\omega)\gg\sigma'(\omega)$, and assuming ideal conditions for phase matching of the squeezing ellipse and the reference wave at the input to the medium $\theta(\boldsymbol{\kappa}_\perp,\Omega;0)-\theta_0(0)=\pi/2$, we obtain for the growth rates of these parameters

$$\Delta r(\boldsymbol{\kappa}_\perp,\Omega)\sim\Delta\lambda(\boldsymbol{\kappa}_\perp,\Omega)\sim 0,$$

$$\Delta(\theta(\boldsymbol{\kappa}_\perp,\Omega)-\theta_0)=\Delta\theta_l(\Omega)+\Delta\theta_{nl}(\boldsymbol{\kappa}_\perp,\Omega), \tag{5.12}$$

where the linear $\Delta\theta_l(\Omega)$ and nonlinear $\Delta\theta_{nl}(\boldsymbol{\kappa}_\perp,\Omega)$ growth rates for the relative angle have the form

$$\Delta\theta_l(\Omega)=-\frac{1}{2}n_0[\sigma''(\omega+\Omega)+\sigma''(\omega-\Omega)$$
$$-2\sigma''(\omega)]\Delta z,$$

$$\Delta\theta_{nl}(\boldsymbol{\kappa}_\perp,\Omega)=\tilde{\chi}(\Omega)\Delta z\,\frac{\xi_0(\boldsymbol{\kappa}_\perp,\Omega)+1}{2r(\boldsymbol{\kappa}_\perp,\Omega)}. \tag{5.13}$$

In the low-frequency range of the fluctuation spectrum $(\Omega\ll\gamma)$ the inequality $\Delta\theta_{nl}\gg\Delta\theta_l$ follows naturally. In this case the change in phase of the anomalous correlator, which can be interpreted geometrically as a rotation of the squeezing ellipse with respect to the reference wave vector in the complex amplitude plane (see Fig. 5), is determined primarily by the nonlinear interactions. The change in the space-time spectral Mandel parameter turns out to be proportional to the square of the angle of rotation:

$$\Delta\xi(\boldsymbol{\kappa}_\perp,\Omega)\approx 4\Delta\theta^2(\boldsymbol{\kappa}_\perp,\Omega)r(\boldsymbol{\kappa}_\perp,\Omega)$$
$$\approx\tilde{\chi}^2(\Omega)\Delta z^2\,\frac{(\xi_0(\boldsymbol{\kappa}_\perp,\Omega)+1)^2}{r(\boldsymbol{\kappa}_\perp,\Omega)}. \tag{5.14}$$

and the correction to the Mandel parameter $\xi(\Omega)$ is found by substituting (5.14) into the integral (5.5). Examining the explicit expression for the four-wave interaction constant $\tilde{\chi}$ (see (5.11)), we see that in this case the rotation of the squeezing ellipse is determined by the orientation of the angular momentum along the external magnetic field.

These results recall the classical Faraday effect, i.e., rotation of the plane of polarization of linearly polarized light as it propagates in a medium in a magnetic field. In this case,

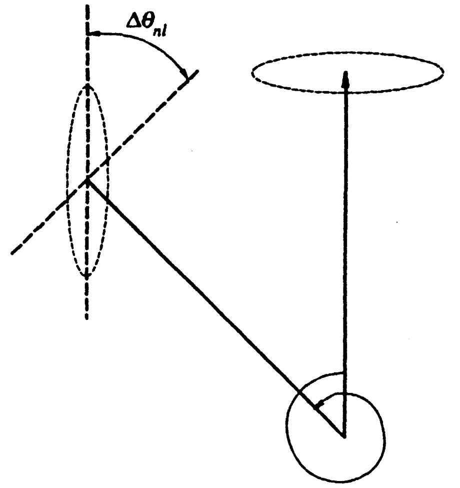


FIG. 5. Geometric illustration of the changes in the Mandel parameter for the parametric conversion shown in Fig. 4. The change in the orientation of the major axis of the squeezing ellipse with respect to a coherent reference wave, which is characterized by an angle of rotation $\Delta\theta_{nl}$, is proportional to the orientation of the angular momenta of the atoms.

the direction of the polarization vector is associated with the major axis of the squeezing ellipse in the complex amplitude plane. As in the ordinary Faraday effect, rotation of the direction of "polarization fluctuations" is initiated by the orientation of the angular (magnetic) moment of the medium. Pursuing this analogy, we can view a heterodyne detector that mixes the squeezed light with a reference wave as a unique type of "polarization analyzer." However, we call the reader's attention to the fact that in this case the direction of the average oriented angular momentum vector is orthogonal to, not parallel to, the direction of the probe beam; see Fig. 4. It is obvious that the occurrence of such a correlation-induced Faraday effect is entirely based on the quantum statistical properties of the light in the case of classical light and is impossible in principle. Thus, this example demonstrates the breadth of possibilities of the method of intensity fluctuation spectroscopy based on the use of quantum properties of light. From a practical standpoint, observation of the rotation of the squeezing ellipse in the complex amplitude plane induced by nonlinear parametric interaction of the light with the medium can be used for spectroscopic analysis of the medium characteristics.

A further interesting feature of the parametric process described by Eqs. (5.8), (5.9) is the fact that when the light incident on the medium is perfectly squeezed, i.e., $\xi_0(\kappa_\perp, \Omega) \rightarrow -1$, the medium becomes optically transparent and the Mandel parameter is unchanged. This result is a consequence of the fact that the intrinsic parametric conversion, which redistributes photons between conjugate modes, and the nonlinear dispersion, both of which are expressed in terms of the same interaction constant, affect the phase of the anomalous correlation function in a compensating manner.

In general, the transformation of the spectrum of fluctuations of squeezed light by a scattering medium reduces to a simultaneous distortion and rotation of the squeezing ellipse; by using the method of intensity fluctuation spectroscopy based on a heterodyne photodetector, we can observe these changes. If the light used to probe the medium were coherent, the distortion would infringe on the original Gaussian ambiguity circle for the quasiprobability distribution corresponding to the shot-noise background in the spectrum of photocurrent fluctuations. In general, it is worth noting that extending the possibilities of the method of intensity fluctuation spectroscopy by replacing coherent light with squeezed light can be just as productive as replacing unpolarized light by polarized light or using polarization-sensitive detection in linear spectroscopy.

6. CONCLUSION

In this paper we have estimated the sensitivity of the method of intensity fluctuation spectroscopy by using a quantum description of the light that probes the medium under study. The fourth-order photocurrent correlation function, which describes errors in measuring the photocurrent spectrum (which is what is observed in the method of intensity fluctuation spectroscopy) consists of the sum of contributions from *TN*-ordered field correlation functions from first to fourth order. For the case of heterodyne photodetection, if we assume quasi-Gaussian statistics when extracting operator averages from the incoherent field components, the variance of fluctuations in the observables is expressed in terms of the spectral Mandel parameter $\xi(\Omega)$ of the detected light. Our estimate shows that when the probe light is squeezed, which implies that a limiting value of $\xi(\Omega) \rightarrow -1$ is possible, we may expect a considerable increase in the sensitivity of the method of intensity fluctuation spectroscopy. We have found that the implications of replacing classical light by squeezed light in this case are more significant than for the analogous situation in linear absorption spectroscopy.

As an example of intensity fluctuation spectroscopy using squeezed light, we have discussed two-photon absorption by a two-level atomic transition and parametric conversion of light in a four-wave scattering process mediated by oriented atoms. In using the results of Ref. 30, we have shown that when two-photon absorbers are used to record small changes in the Mandel parameter, the sensitivity is considerably higher for squeezed light than it is for classical light. This circumstance is of considerable importance in applications, since two-photon and multiphoton absorption processes are widely used in atomic spectroscopy, molecular spectroscopy, and photochemistry.

In the case of parametric scattering, the use of squeezed light leads to qualitatively new ways to use the method of intensity fluctuation spectroscopy. Changes in the Mandel parameter of the transmitted light can be inferred in this case by observing a rotation of the squeezing ellipse in the complex amplitude plane relative to the coherent component of the reference wave. For this example the angle of rotation is found to be proportional to the orientation of the angular momenta of the atoms in the direction perpendicular to the probe beam. An interesting feature of this effect, which we refer to as a correlation-induced Faraday effect, is that it cannot be interpreted by using a classical representation of fluctuations in either the light intensity or the susceptibility of the scattering medium. This is because the latter, which mediates the generation of phase-conjugate photon pairs, in practice changes the state of the field only at the level of the quantum uncertainty relation. Also interesting is the fact (possibly associated with its quantum nature) that observation of this effect is accompanied by a considerably larger signal/noise ratio if the squeezing ellipse in the light incident on the medium is oriented at an angle of $\pi/4$ and not $\pi/2$. In this case, it is easy to verify that the variance of fluctuations of the Mandel parameter determined by (4.17) will be considerably higher (not lower) than for classical light. Note, however, that in this observation geometry, along with the rotation we will also observe a distortion of the squeezing ellipse. In order to observe a pure rotation, we can couple the reference wave at the output from the medium, i.e., just inside the detector channel. In this case we must introduce an additional phase shift for the reference wave that takes into account the linear dispersion of the medium.

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1)We note that the term "intensity fluctuation spectroscopy" is usually used in Russian literature. In the English literature, other names are more often used: correlation spectroscopy, optical mixing spectroscopy, and quantum beating spectroscopy. All of these terms are quite close in their physical meaning, and in this paper we will, for the most part, adhere to the term "intensity fluctuation spectroscopy."

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