Fluctuations of the polarization of the radiation transmitted by a nonequilibrium gaseous medium

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An investigation is reported of correlations of fluctuations of the polarization of the radiation transmitted by a nonequilibrium optically thin gaseous medium. It is shown that the correlator of photocurrents is related to symmetric and antisymmetric correlators of the polarization moment of the Wigner density matrix of particles in the medium. The resonances exhibited by the correlators at zero and Zeeman frequencies are governed by the average polarization moments of the density matrix of the medium of rank $\varkappa = 1$–4. In a weak magnetic field the contributions of the moments $\varkappa = 3$ and 4 can be separated explicitly.

1. INTRODUCTION

Spectroscopy of the intensity fluctuations (SIF)[1-7] is a new promising method for the investigation of the spectra of atoms and molecules and of interparticle interactions, the potentialities of which have not yet been realized in full. The essence of the SIF method is determination of a correlation between fluctuations of the intensities of the radiation transmitted by a gas cell at different moments in time and an analysis of the spectrum of these correlations. For example, experiments reported in Ref. 2 demonstrated that the spectrum of correlations of the angles of rotation of the plane of polarization of linearly polarized light transmitted by a cell containing Na vapor, studied in the optical transparency region in the vicinity of the D_1 line, exhibits a resonance at an rf frequency corresponding to the Zeeman splitting of the ground state of the sodium atom. A theory of the effect proposed in Refs. 2 and 3 accounts for the resonance as a consequence of correlations between fluctuations of the refractive index, which are due to fluctuations of the random optical orientation of atoms inside a beam. Subsequent investigations[5-7] have provided a theoretical generalization of the SIF method. Studies have been made of the correlation between fluctuations of the transmitted light, of fluorescence, and of scattered light. However, in all these cases a gaseous medium was assumed to be in equilibrium and a practical recommendation could be reduced basically to the feasibility of using the SIF method in a study of rf resonances in noise and determination of the relaxation constants of various polarization moments of atoms in the medium, including the relaxation parameters of high-rank ($\varkappa = 3$–4) moments when the fluctuations are manifested in the scattered radiation noise[6,7] in those cases when it is difficult to use directly optical pumping, rf resonance, and other methods.

We shall report an investigation of the feasibility of using the SIF method in experiments on a nonequilibrium gas containing atoms or molecules which have arbitrary polarization moments. The value of the correlator of fluctuations of different polarization moments of the density matrix is then determined not only by the concentration of the particles, as in an equilibrium gas,[1-3] but also by the average values of the polarization moments of nonzero ranks $\varkappa = 1$–4. In some cases the contribution of moments of higher rank $\varkappa = 3$ or 4 can be separated explicitly. This provides an opportunity for direct determination of these moments, which is difficult to carry out by conventional optical methods. The use of the SIF method in experiments on a nonequilibrium gas may therefore provide additional information on the interaction of particles with light or with one another, giving rise to high-rank polarization moments.

2. CORRELATOR OF PHOTOCURRENTS

In SIF method one measures the correlator of the photocurrents induced by radiation which probes a medium and passes through optical analyzers; this correlator is related to a second-order correlation function of an electromagnetic field (see Refs. 8 and 5)

$$\langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle = \sum_{\mu_1\mu_1'}\sum_{\mu_2\mu_2'} e^2 \iint d^2r_1\, d^2r_2\, \nu_1\nu_2 \int_{-\infty}^{\infty}\int d\tau_1\, d\tau_2, \tag{1}$$
$$f^{(1)}_{\mu_1\mu_1'}(\tau_1)\, f^{(2)}_{\mu_2\mu_2'}(\tau_2)\exp(-i\omega\tau_1 - i\omega\tau_2)\, D^{(E)}(1,2;2',1').$$

Here, $D^{(E)}(1,2;2',1')$ is the correlator of the Heisenberg operators of the electric field $E(i)\equiv E_{\mu i}(r_i t_i)$, and is expressed in the Coulomb gauge and quasimonochromatic case in terms of the correlator of the vector potential operators:

$$D^{(E)}(1,\ 2;\ 2',\ 1') \approx (\omega/c)^4 \langle \tilde{T}(A(2)A(1))\,T(A(1')A(2'))\rangle \equiv (\omega/c)^4 D(1,2;2',1'), \tag{2}$$
$$r_i = r_i', \qquad t_i' = t_i - \tau_i.$$

The angular brackets denote averaging over the density operator ρ_0 of the system; T and $\tilde{T}$ are the time-ordering and antiordering operators; $f^{(i)}_{\mu\mu_i'}(\tau_i)$ are the photodetector sensitivity functions the dependences of which on the polarization indices μ_i and μ_i' are governed by the analyzers standing in front of the photodetectors; ω is the average frequency of the probe radiation; e is the electron charge. The sensitivity functions are localized in time so that $f^{(i)}_{\mu\mu'}(\tau)\to 0$ if $\tau \gtrsim \tau_c$, where τ_c is the characteristic time scale of the investigated correlations. Integration with respect to $\mathbf{r}_1$ and $\mathbf{r}_2$ in Eq. (1) is carried out over the photocathode surfaces; ν_1 and ν_2 are the numbers of atoms per unit surface area.

In the Keldysh diagram technique[9,10] the two-photon correlator defined by Eq. (2) corresponds to a two-particle Green function the ends of which 1′ and 2′ are labeled with the minus sign and the ends 1 and 2 are labeled with the plus sign. Calculation of this function will be made subject to the

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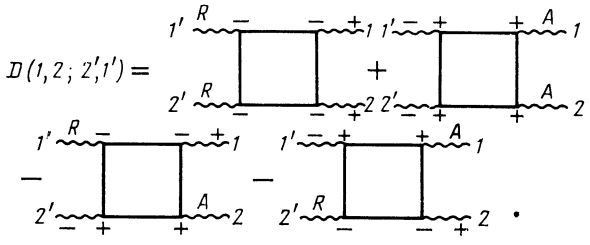
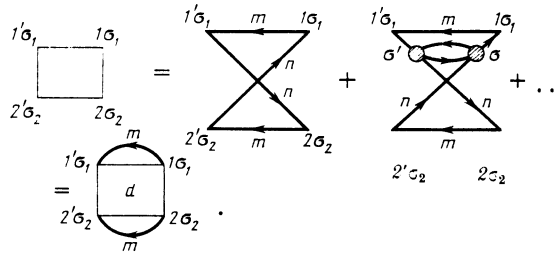
following assumptions. The light incident on a medium will be regarded as Gaussian radiation with the coherence time assumed short compared with τ_c. We shall consider an optically thin gas layer, so that a single interaction of a photon with a medium is important. We shall assume that the radiation is quasimonochromatic and its spectrum is in quasiresonance with the frequency of the transition being excited. The dimensions of the photodetector are assumed to be sufficiently small to ignore the contribution of the scattered light. The density matrix of the gas particles is assumed to be of nonequilibrium nature in respect of the internal state because of the interaction with external pump sources, collisional processes, etc. (for brevity, we shall consider primarily atoms, but the treatment is applicable also to a molecular gas). We shall assume that the probe radiation is weak and it does not influence the state of the medium.

An analysis of possible Keldysh diagrams shows that, subject to the adopted approximations, the following graphs contribute mainly to the correlation of the transmitted radiation:



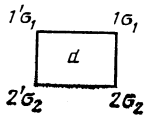
(3)

The wavy lines denote one-photon Green functions in the standard notation.[10] The indices R and A identify retarded and advanced Green functions. The internal parts of the diagrams in Eq. (3) represent blocks composed of atomic Green functions:



(4)

Here, $\sigma_1(\sigma_2) = -, +$ the continuous thick lines with indices n and m represent the exact Green functions for the ground and excited states of atoms, respectively. The terms of the expansion of Eq. (4) allow for all possible interactions of atoms in the ground state with one another and with buffer particles. The shaded circles represent a complete set of irreducible diagrams describing two-particle interactions. Using the fact that the Green functions of excited states decay in intervals of the order of the lifetime τ_m of these states and over distances of the order of $v\tau_m$ (v is the thermal velocity of an atom), we can describe the evolution of the atomic Heisenberg operators of the ground state over intervals of this kind by the free evolution approximation. The correlation block identified by the letter d in Eq. (4) is then described by



$$= \iint \frac{d^3P_1}{(2\pi\hbar)^3}\frac{d^3P_2}{(2\pi\hbar)^3} \times \exp\left[\frac{i}{\hbar}\mathbf{P}_1(\mathbf{r}_1-\mathbf{r}_1') + \frac{i}{\hbar}\mathbf{P}_2(\mathbf{r}_2-\mathbf{r}_2') - \frac{i}{\hbar}\frac{E_{n_1}(P_1)+E_{n_1'}(P_1)}{2}(t_1-t_1') - \frac{i}{\hbar}\frac{E_{n_2}(P_2)+E_{n_2'}(P_2)}{2}(t_2-t_2')\right]$$

$$\times K^{(\sigma_1\sigma_2)}_{n_1'n_1;\,n_2'n_2}(\mathbf{P}_1\mathbf{R}_1T_1;\,\mathbf{P}_2\mathbf{R}_2T_2), \quad K^{(\sigma_1\sigma_2)}_{n_1'n_1;\,n_2'n_2}(\mathbf{P}_1\mathbf{R}_1T_1;\,\mathbf{P}_2\mathbf{R}_2T_2) \tag{5}$$

$$= \mathrm{Sp}[\rho_0 T_{\sigma_1\sigma_2}(\delta\hat{\rho}_{n_2'n_2}(\mathbf{P}_2\mathbf{R}_2T_2)\,\delta\hat{\rho}_{n_1'n_1}(\mathbf{P}_1\mathbf{R}_1T_1))]. \tag{6}$$

Here $\delta\hat{\rho}_{n'n}(\mathbf{R}T)$ denotes the Heisenberg operator representing fluctuations of the Wigner density matrix:

$$\delta\hat{\rho}_{n'n}(\mathbf{PR}T) = \sum_{\mathbf{p}} e^{i\mathbf{pR}/\hbar}\, a^+_{n'\mathbf{P}-\mathbf{p}/2}(T)\, a_{n\mathbf{P}+\mathbf{p}/2}(T) - \rho_{n'n}(\mathbf{PR}T), \tag{7}$$

which evolves with time in accordance with the Hamiltonian of the atomic subsystem. The indices n denote the quantum numbers of the ground state; $\rho_{n'n}(\mathbf{PR}T)$ is the Wigner density matrix; $\mathbf{R}_i = (\mathbf{r}_i + \mathbf{r}_i')/2$; $T_i = (t_i + t_i')/2$; is the total energy of an atom; $a^+_{n'\mathbf{P}}(T)$ and $a_{np}(T)$ are the Heisenberg creation and annihilation operators for atoms in the ground state. The ordering operators $T_{\sigma_1\sigma_2}$ act in accordance with the following rules: T is the ordering operator of the "slow" times T_1 and T_2; T_{++} is the antiordering operator of T_1 and T_2; T_{-+} is the identity operator; T_{+-} is the operator representing transposition of fluctuations $\delta\hat{\rho}(1) \rightleftarrows \delta\hat{\rho}(2)$.

In calculation of the correlator of the photocurrents in accordance with Eq. (1) we shall temporarily assume that an optical analyzer records directly a certain polarization moment of the radiation and the dependence of the sensitivity functions in Eq. (1) on the correlation indices in coordinate system with the z axis along the direction of propagation is described by the relevant polarization operator:

$$f^{(i)}_{\mu_i\mu_i'}(\tau) = (-1)^{\mu_i}\langle 1-\mu_i|\,\hat{T}^{(i)}_{K-Q}(11)\,|1\mu_i'\rangle\, f^{(i)}(\tau), \tag{8}$$

where the polarization operator $\hat{T}^{(i)}_{K-Q}$ is defined in the standard manner[11]:

$$\hat{T}^{(i)}_{K-Q}(11) = \sum_{\mu_i'\mu_i} C^{K-Q}_{1\mu_i\,1-\mu_i'}\,|1\mu_i\rangle\langle 1\mu_i'|\,(-1)^{1-\mu_i'}. \tag{9}$$

It is convenient to introduce the polarization moment representation for the operators of fluctuations of the Wigner density matrix:

$$\delta\hat{\rho}_{j_i}{}^{\varkappa q}(\mathbf{PR}T) = \sum_{n'n} (-1)^{j_i-n'}\Pi_{j_i}\begin{pmatrix} j_i & \varkappa & j_i \\ -n' & -q & n\end{pmatrix}\delta\hat{\rho}_{n'n}(\mathbf{PR}T), \tag{10}$$

where j_i $(i = 1,2)$ are the total momenta of atoms in a ground state which, in principle, can be different for consecutive interactions with radiation if an atom has a hyperfine structure: n and n' are the values of the projection of the total angular momentum. We shall use the notation[11] $\Pi_{XY\ldots} = [(2X+1)(2Y+1)\ldots]^{1/2}$.

We shall consider later the steady-state case when the matrix representing the density of the gas atoms has a constant polarization moment. In this case the one-particle Green functions depend on the difference between their time arguments and one can avoid internal integration with respect to the time variables in Eq. (1) by adopting the Fourier representation the "fast" difference times. When all these approximations are allowed for, the current correlator is of the form

$$\langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle^{K_1Q_1;\ K_2Q_2} = \langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle^{K_1Q_1;\ K_2Q_2}_{(s)} + \langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle^{K_1Q_1;\ K_2Q_2}_{(a)}, \quad (11)$$

$$\langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle^{K_1Q_1;\ K_2Q_2}_{(s)} = e^2\zeta_1\zeta_2 \sum_{\varkappa_1 q_1}\sum_{\varkappa_2 q_2}\sum_{K_1'Q_1'}\sum_{K_2'Q_2'}\iint_V d^3R_1\,d^3R_2 \times \iint \frac{d^3P_1}{(2\pi\hbar)^3}\frac{d^3P_2}{(2\pi\hbar)^3}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi} S^{K_1Q_1}_{K_1'Q_1'}(\omega_1 P_{1z};\varkappa_1 q_1) \times S^{K_2Q_2}_{K_2'Q_2'}(\omega_2 P_{2z};\varkappa_2 q_2) \times K^{\varkappa_1 q_1;\varkappa_2 q_2}_{(s)\,j_1;j_2}(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2)\,(\hbar\omega)^{-2} I^{K_1'Q_1'}(\omega_1\mathbf{R}_1) I^{K_2'Q_2'}(\omega_2\mathbf{R}_2), \quad (12)$$

$$\langle \delta i_1(t_1)\,\delta i_2(t_2)\rangle^{K_1Q_1;\ K_2Q_2}_{(a)} = e^2\zeta_1\zeta_2 \sum_{\varkappa_1 q_1}\sum_{\varkappa_2 q_2}\sum_{K_1'Q_1'}\sum_{K_2'Q_2'}\iint_V d^3R_1\,d^3R_2 \times \iint \frac{d^3P_1}{(2\pi\hbar)^3}\frac{d^3P_2}{(2\pi\hbar)^3}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi} \times \{S^{K_1Q_1}_{K_1'Q_1'}(\omega_1 P_{1z};\varkappa_1 q_1) A^{K_2Q_2}_{K_2'Q_2'}(\omega_2 P_{2z};\varkappa_2 q_2) \times \theta(t_1-t_2) - A^{K_1Q_1}_{K_1'Q_1'}(\omega_1 P_{1z};\varkappa_1 q_1) S^{K_2Q_2}_{K_2'Q_2'}(\omega_2 P_{2z};\varkappa_2 q_2)\theta(t_2-t_1)\} \times K^{\varkappa_1 q_1;\varkappa_2 q_2}_{(a)\,j_1;j_2}(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2)\,(\hbar\omega)^{-2} I^{K_1'Q_1'}(\omega_1\mathbf{R}_1) I^{K_2'Q_2'}(\omega_2\mathbf{R}_2). \quad (13)$$

Integration in Eqs. (12) and (13) with respect to $\mathbf{R}_1$ and $\mathbf{R}_2$ is carried out over a volume V occupied by a beam; $I^{K_i'Q_i'}(\omega_i\mathbf{R}_i)$ is the spectral density of the polarization tensor of the radiation at the entry to the cell at a point $\mathbf{R}_i$, defined as the product of the polarization density matrix and the spectral intensity [in the case of the optically thin layer under discussion it depends only on the transverse (relative to the direction of propagation z) coordinates]; ζ_1 and ζ_2 are the quantum efficiencies of the photodetectors. Equations (12) and (13) contain the symmetric and antisymmetric correlators of the Wigner density matrix:

$$K^{\varkappa_1 q_1;\varkappa_2 q_2}_{(s,a)\,j_1;j_2}(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2) = \mathrm{Sp}\{\rho_0[\delta\hat{\rho}^{\varkappa_1 q_1}_{j_1}(\mathbf{P}_1\mathbf{R}_1 t_1) \times \delta\hat{\rho}^{\varkappa_2 q_2}_{j_2}(\mathbf{P}_2\mathbf{R}_2 t_2) \pm \delta\hat{\rho}^{\varkappa_2 q_2}_{j_2}(\mathbf{P}_2\mathbf{R}_2 t_2)\,\delta\hat{\rho}^{\varkappa_1 q_1}_{j_1}(\mathbf{P}_1\mathbf{R}_1 t_1)]\}. \quad (14)$$

The expressions for the matrices $S^{KQ}_{K'Q'}(\omega P_z;\varkappa q)$ and $A^{KQ}_{K'Q'}(\omega P_z;\varkappa q)$ are given in the Appendix.

An important difference between Eqs. (11)–(13), relating the photocurrent correlator to the correlator of the polarization moments of the Wigner density matrix, and the corresponding relationships in the equilibrium case,[3] is the presence in the former of an antisymmetric density correlator which vanishes for a gas in equilibrium (see below).

3. FLUCTUATIONS OF THE POLARIZATION MOMENTS OF THE WIGNER DENSITY MATRIX IN THE ABSENCE OF RELAXATION

A description of fluctuations of the polarization moments of the Wigner density matrix and a calculation of the corresponding correlator must allow for the processes of collisional relaxation in the ground state. Using the results of the theory of Refs. 10, 12, and 13, we may conclude that the correlator of the polarization moment of fluctuations should satisfy the same system of kinetic or diffusion equations as the polarization moments of the density matrix themselves. In various specific situations, such as that of atoms with hyperfine structure in the ground state, the system of equations may be quite complex because of the possible processes of redistribution of the polarization between the sublevels it is difficult to solve analytically. Since our aim is to demonstrate the potential applications of the SIF methods in the study of atoms or molecules polarized in respect of the internal angular momentum, we shall consider experiments in which the effects of relaxation are unimportant. In the case of atoms in the S state this allows us to consider free motion across a beam (in the absence of collisions) and diffusion, because the depolarization cross sections are then much smaller than the gas-kinetic cross sections. In the case of atoms in the states with $L \neq 0$ and molecules we shall assumeonly free motion, since the depolarization cross sections are then sufficiently large.

When these assumptions are allowed for, the correlator of the polarization moments of the Wigner density matrix becomes

$$K^{\varkappa_1 q_1;\varkappa_2 q_2}_{(s,a)\,j_1;j_2}(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2) \approx \delta_{j_1 j_2} k(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2) \times \sum_{\varkappa q}\frac{1\pm(-1)^{\varkappa_1+\varkappa_2+\varkappa}}{2}(-1)^{q+2j}\begin{pmatrix}\varkappa_1 & \varkappa_2 & \varkappa\\ q_1 & q_2 & -q\end{pmatrix}\Pi_\varkappa{}^2\Pi_j \times \begin{Bmatrix}\varkappa_1 & \varkappa_2 & \varkappa\\ j & j & j\end{Bmatrix}\exp\{-i\omega_0 q_1(t_1-t_2)\}\,\rho_j^{\varkappa q}, \quad j_1=j_2=j. \quad (15)$$

In the presence of a strong magnetic field when the frequency of the Zeeman splitting ω_0 is higher than the rate of relaxation Γ of the polarization moment of the ground state, Eq. (15) is valid only for the coordinate system with the z' axis directed along the magnetic field $\mathbf{H}_0$. The polarization moments of the density matrix are $\rho_j^{\varkappa q}\neq 0$ only if $q=0$. In the absence of a magnetic field we have $\omega_0 \ll \Gamma$, so that Eq. (15) is valid in any coordinate system. Since the average time taken by an atom to cross a light beam $\bar{\tau}$ is by definition less than the relaxation time Γ^{-1}, it follows that in a weak magnetic field the exponential factor is close to unity and can be ignored.

The derivation of Eq. (15) in the case of free motion is based on analytic interpretation of the first term of the expansion (4) allowing for the equality (5) and using expressions for single-particle Green functions of an ideal nonequilibrium gas in terms of the components of the Wigner density matrix (see Ref. 14). The function $k(\mathbf{P}_1\mathbf{R}_1 t_1;\mathbf{P}_2\mathbf{R}_2 t_2)$ represents a correlator of the classical distribution function of an ideal gas:

$$k(\mathbf{P}_1\mathbf{R}_1 t_1;\ \mathbf{P}_2\mathbf{R}_2 t_2) = n_0(2\pi\hbar)^3 f_0(P_1)\,\delta(\mathbf{P}_1-\mathbf{P}_2) \times \delta(\mathbf{R}_2-\mathbf{R}_1-\mathbf{P}_1(t_2-t_1)/m). \quad (16)$$

This result is derived assuming the Maxwellian distribution of the atomic linear momenta:

$$\rho_j^{\varkappa q}(\mathbf{PR}t)=n_0\rho_j^{\varkappa q}(\mathbf{R})f_0(P),$$
$$f_0(P)=(2\pi\hbar^3/mT)^{3/2}\exp(-P^2/2mT), \tag{17}$$

where n_0 is the concentration of the atoms. Then, since there is no relaxation in the course of the motion of atoms across the beam, we can ignore the dependence on the spatial coordinate under steady-state conditions.

In the case of diffusion when the interaction operator does not include the spin variables, the dependence of each term and of the whole block (4) on the quantum numbers n_i and n_i' is the same as for free motion, i.e., only the first term of the expansion need be retained and the thick lines are replaced with thin ones. The sum of the terms of the expansion (4) subject to Eq. (5) is independent of the internal quantum numbers and determines the correlator of the distribution function of a real gas for which we shall use the diffusion asymptote:

$$k(\mathbf{P}_1\mathbf{R}_1t_1;\ \mathbf{P}_2\mathbf{R}_2t_2)=n_0f_0(P_1)f_0(P_2)[4\pi D^2|t_2-t_1|]^{-3/2}\exp[-(\mathbf{R}_2-\mathbf{R}_1)^2/4D^2|t_2-t_1|], \tag{18}$$

where D is the diffusion coefficient.

It follows directly from Eq. (15) that if we allow for the adopted approximations in the case of an equilibrium gas, the antisymmetric correlator of the density matrix vanishes. In the case of fluctuations of the polarization moments which are longitudinal relative to a magnetic field **H**, we again have $K_{(a)j;\,j}^{\varkappa_1 0;\varkappa_2 0}=0$.

4. DISCUSSION OF POSSIBLE EXPERIMENTS

By way of example, we shall consider the experimental setup shown in Fig. 1. Unpolarized radiation crosses a resonantly absorbing cell containing the vapor of working atoms. A beam of this radiation is split by a semitransparent mirror M, light then passes through analyzers A_1 and A_2, and is recorded with photodetectors D_1 and D_2. After passing through a delay line the photocurrents are multiplied and are subjected to a spectral analysis with a correlator C. It should be pointed out that when such a medium is probed with resonant radiation characterized by a profile symmetric relative to the absorption line, the correlator $\langle\delta i_1(t_1)\delta i_2(t_2)\rangle_{(a)}^{K_1Q_1;K_2Q_2}$ vanishes. This follows directly from Eqs. (13) and Eqs. (A1) and (A2). We shall consider only this case.

We shall assume that the analyzers A_1 and A_2 pass only radiation with the linear polarization along the x and y axes. Allowing for the relationship between the degree of linear polarization p_x along the x axis and the polarization moment of the density matrix of the radiation

$$p_x=-2\mathrm{Re}\,\Phi^{22},\quad \Phi^{KQ}=I^{KQ}(\omega)/3^{1/2}I^{00}(\omega), \tag{19}$$

we find that the measured quantity is the current correlator:

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,p_x}=\sum_{Q_1,Q_2=\pm2}\langle\delta i_1(t_1)\delta i_2(t_2)\rangle_s^{2Q_1;2Q_2}. \tag{20}$$

If the direction of the magnetic field in the laboratory coordinate system xyz (Fig. 1) is represented by spherical functions θ and φ, then calculations based on Eqs. (12) and (A.1) give

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,p_x}=\sum_{\varkappa=0}^{4}\frac{1+(-1)^\varkappa}{2}\rho_j^{\varkappa 0}(-1)^{\varkappa+2j}\Pi_{\varkappa 22j}\times\begin{Bmatrix}2&2&\varkappa\\ j&j&j\end{Bmatrix}[{}^3/_{16}C_{2020}^{\varkappa 0}c_0(t_1-t_2)\sin^4\theta(1+\cos 4\varphi)$$
$$-{}^1/_4C_{212-1}^{\varkappa 0}c_1(t_1-t_2)\sin^2\theta(1+\cos^2\theta-\cos 4\varphi\sin^2\theta)$$
$$+{}^1/_{16}C_{222-2}^{\varkappa 0}c_2(t_1-t_2)(4\cos^2\theta+(1+\cos^2\theta)^2+\cos 4\varphi\sin^4\theta)]. \tag{21}$$

The functions $c_q(\tau)$ in Eq. (21) are defined as follows:

$$c_q(\tau)=e^2\xi_1\xi_2\int_V\int d^3R_1\,d^3R_2\int\int\frac{d^3P_1}{(2\pi\hbar)^3}\frac{d^3P_2}{(2\pi\hbar)^3}\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\times\frac{d\omega_2}{2\pi}\Pi_j^{-2}\begin{Bmatrix}1&1&2\\ j&j&j'\end{Bmatrix}^2\mathrm{Re}\,\sigma_{jj'}(\omega_1P_{1z})\,\mathrm{Re}\,\sigma_{jj'}(\omega_2P_{2z})$$
$$\times(\hbar\omega)^{-2}I(\omega_1R_1)I(\omega_2R_2)k(\mathbf{P}_1\mathbf{R}_1t_1;\ \mathbf{P}_2\mathbf{R}_2t_2)\cos q\omega_0\tau,$$
$$\tau=t_1-t_2, \tag{22}$$

where j and j' are the moments of the ground and excited states; the function $\sigma_{jj'}(\omega P_z)$ is defined by Eq. (A.3); $I(\omega, \mathbf{R})$ is the spectral intensity of the incident radiation. In the presence of a strong magnetic field, $\omega_0\bar{\tau}\gg1$ the components of the Fourier functions $c_q(\tau)$ represent resonance profiles centered on frequencies $q\omega_0$. The nature and width of the resonances are governed by the nature of the correlator $k(\mathbf{P}_1\mathbf{R}_1t_1;\ \mathbf{P}_2\mathbf{R}_2t)$, which under steady-state conditions is a function of the difference of its arguments $\tau=t_1-t_2$.

A nonequilibrium gas with atoms that have polarization moments of even rank represents an optically anisotropic medium in which the direction of the optic axis coincides with the direction of the polarization moment.[15,16] In the

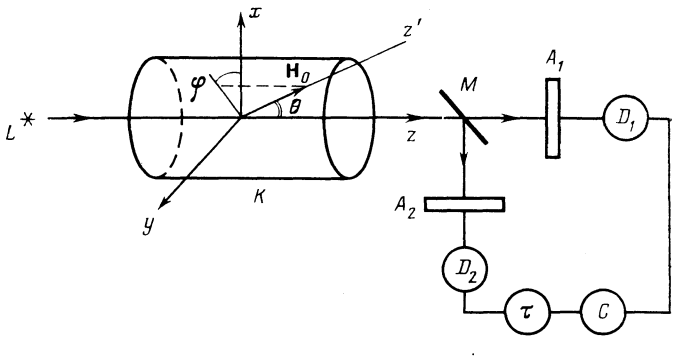


FIG. 1. Experimental setup for the determination of correlations of the transmitted radiation: L is the source of unpolarized radiation; K is a gas cell; M is a semitransparent mirror; A_1 and A_2 are optical analyzers; D_1 and D_2 are photodetectors; τ is the delay line; C is the correlating system.

case under discussion this is the direction of the magnetic field. It would be interesting to determine two independent correlations of fluctuations of the linear polarization: along the optic axis and at an angle of 45° to this axis. If the magnetic field is weak, $\omega_0\bar{\tau}\ll 1$, then bearing in mind that $c_0(\tau)\approx c_1(\tau)\approx c_2(\tau)$, we find the difference between these correlations is

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,p_x}|_{\varphi=0}-\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,p_x}|_{\varphi=\pi/4}$$
$$=\rho_j^{40}\Pi_{422j}(-1)^{2j}\begin{Bmatrix}2&2&4\\j&j&j\end{Bmatrix}c_0(t_1-t_2)\frac{\sqrt{70}}{4\sqrt{2}}\sin^4\theta. \quad (23)$$

This quantity is proportional to the moment $\varkappa=4$. This is due to the fact that the left-hand side of Eq. (23) represents the definition of the correlator $\langle\delta i_1(t_1)\delta i_2(t_2)\rangle$[22,23], which is related to the density correlator $K^{22;22}_{(s)j;j}$.. The density correlator is in its turn proportional to the atomic polarization moment ρ_j^{44} in the coordinate system xyz. The angular factor in Eq. (23) is proportional to the function $d^4_{40}(\theta)$ and relates this moment to the moment ρ_j^{40} in the coordinate system with the z' axis along the magnetic field $\mathbf{H}_0$. In a strong magnetic field, $\omega_0\bar{\tau}\gg 1$ Eq. (23) is satisfied only for short time intervals $t_1-t_2\ll\omega_0^{-1}$. In a study of the spectrum of correlations of the photocurrent in the vicinity of the frequencies ω_0 and $2\omega_0$ the moment $\varkappa=4$ appears against the background of the total population and alignment. We can estimate its value if we have additional information on the population and alignment, which can be obtained by conventional optical detection methods.

We shall now consider the possibility of detection of a polarization moment of odd rank. We shall assume that the analyzers A_1 and A_2 deal essentially with the linear (along the x and y axes) and circular polarizations. Bearing in mind the relationship between the Stokes parameter ξ_2, representing the degree of circular polarization, and the polarization moment of the radiation

$$\xi_2=-\sqrt{2}\Phi^{10}, \quad (24)$$

as well as Eq. (19), we find that the measured quantity is now the current correlator:

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,\xi_2}=\sqrt{2}\sum_{Q=\pm 2}\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{2Q,10}_{(s)}. \quad (25)$$

Consequently, calculations based on Eqs. (12) and (A.1) give

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,\xi_2}=\sum_{\varkappa=1}^{3}\frac{1-(-1)^\varkappa}{2}\rho_j^{\varkappa 0}(-1)^{2j}\Pi_{\varkappa j}$$
$$\times\begin{Bmatrix}2&1&\varkappa\\i&i&i\end{Bmatrix}\left[\frac{\sqrt{6}}{8}C^{\varkappa 0}_{2010}b_0^+(t_1-t_2)\cos 2\varphi\sin^2\theta\cos\theta\right.$$
$$+\frac{\sqrt{2}}{4}C^{\varkappa 0}_{211-1}b_1^+(t_1-t_2)$$
$$\left.\times\cos 2\varphi\sin^2\theta\cos\theta-\frac{\sqrt{2}}{4}C^{\varkappa 0}_{211-1}b_1^-(t_1-t_2)\sin 2\varphi\sin^2\theta\right]. \quad (26)$$

The functions $b_q^{\pm}(\tau)$ are defined by

$$\begin{pmatrix}b_q^+(\tau)\\b_q^-(\tau)\end{pmatrix}=e^2\zeta_1\zeta_2\int\int_V d^3R_1\,d^3R_2\int\int\frac{d^3P_1}{(2\pi\hbar)^3}\frac{d^3P_2}{(2\pi\hbar)^3}$$
$$\times\int\int_{-\infty}^{\infty}\frac{d\omega_1}{2\pi}\frac{d\omega_2}{2\pi}\operatorname{Re}\sigma_{jj'}(\omega_1,P_{1z})\operatorname{Re}\sigma_{jj'}(\omega_2,P_{2z})$$
$$\times\frac{\sqrt{30}}{\Pi_j^2}\begin{Bmatrix}1&1&2\\j&j&j'\end{Bmatrix}\begin{Bmatrix}1&1&1\\j&j&j'\end{Bmatrix}(\hbar\omega)^{-2}I(\omega_1,\mathbf{R}_1)I(\omega_2,\mathbf{R}_2)$$
$$\times k(\mathbf{P}_1\mathbf{R}_1t_1;\mathbf{P}_2\mathbf{R}_2t_2)\begin{pmatrix}\cos q\omega_0\tau\\\sin q\omega_0\tau\end{pmatrix},\quad \tau=t_1-t_2, \quad (27)$$

where $b_0^-(\tau)=0$. As in the case of the functions $c_q(\tau)$, defined by Eq. (22), the Fourier components of the functions $b_q^{\pm}(\tau)$ represent resonance profiles centered on frequencies $q\omega_0$. The functions $b_1^-(\tau)$ considered in the Fourier approximation correspond to a complex quantity. This is not unexpected because the correlator of two different photocurrents may be an odd function of the difference between the arguments. If the correlator of the photocurrent is measured using a spectrum analyzer, by analogy with Ref. 2, then only the real part of the correlator is recorded. We can show that a determination of the imaginary part of the fluctuation spectrum in this analyzer requires introduction of a delay line for one of the photocurrents and this line should induce a delay $\tau_0\sim\pi/2\Omega$, where Ω is the frequency in the vicinity of a resonance $\Omega\sim\omega_0$.

If the magnetic field is weak or if we deal with correlations separated by short time intervals we have $\omega_0\bar{\tau}\ll 1$, then bearing in mind that $b_+^0(\tau)=b_1^+(\tau)$ and $b_1^-(\tau)\to 0$, we obtain from Eq. (26)

$$\langle\delta i_1(t_1)\delta i_2(t_2)\rangle^{p_x,\xi_2}$$
$$=\frac{\sqrt{10}}{8}\rho_j^{30}\Pi_{3j}(-1)^{2j}\begin{Bmatrix}2&1&3\\j&j&j\end{Bmatrix}b_0^+(t_1-t_2)\cos 2\varphi\sin^2\theta\cos\theta. \quad (28)$$

This relationship is interpreted by analogy with Eq. (23). A signal recorded by the method described above is proportional to the sum of the density correlators $K^{2\pm 2;10}_{(s)j;j}$, which is proportional to the sum of the polarization moments $\rho_j^{3\pm 2}$ in the coordinate system xyz. The angular factor which occurs in Eq. (28) is proportional to the function $\operatorname{Re}D^3_{20}(\varphi,\theta,0)$ relating the quantity $\operatorname{Re}\rho_j^{32}$ to the polarization moment ρ_j^{30} in the coordinate system with the z' axis along the magnetic field. In the case of the nonzero magnetic field we can estimate the moment ρ_j^{30} from an analysis of the correlation spectrum at the frequency ω_0 if we have additional information on the degree of orientation.

We shall conclude this section by noting that in these calculations we are dealing with a specific fast atomic transition $j\to j'$ (j and j' are the total angular momenta of the ground and excited states). In the case of atoms characterized by a hyperfine structure these momenta are the sum of the total electron and nuclear momenta. If the hyperfine structure of an excited state is "masked" by the Doppler broadening, as is found frequently, Eqs. (A.1) and (A.2) should be supplemented by summation of the values of the momentum j' of the excited state. If the total electron angular momentum of the ground state is $j_{eL}=1/2$ (alkali atoms), we can easily show that it is not possible to use the SIF method to observe the polarization moment of rank $\varkappa>2$ for the hyperfine structure sublevels of the ground

state. The SIF method can then be used to record the orientation and alignment. It should be mentioned that under these conditions we can detect only the orientation if we use conventional optical methods.

NUMERICAL ESTIMATES. CONCLUSIONS

The potentialities of SIF experiments are limited primarily by fluctuations of the shot background level.[2,3,5] We can observe informative correlations only if the amplitude of the relevant resonances is considerably higher than the average value of these fluctuations. In the experiments considered here, when two photodetectors are used, the average value of the shot background vanishes because of the lack of correlation between the relevant Poisson photoabsorption processes. However, in real experiments the observations last a finite time. Using the general theory of the spectra of random processes,[17] it is easy to show that in a spectral analyzer with a pass band Δ, if the observation time is T a zero shot background level in the spectrum of a photocurrent correlator is achieved to within

$$T^{-1}i_1^T(\omega)\,i_2^T(-\omega)\sim e(\bar{i}_1\bar{i}_2/T\Delta)^{1/2}, \tag{29}$$

where $i_1^T(\omega)$ and $i_2^T(\omega)$ are the running spectra of the realizations for the first and second detectors in a finite observation time T (Ref. 17). The ratio of the amplitude of such informative correlations to the level of fluctuations of the shot noise is described by a dimensionless parameter

$$\eta=1/2(\zeta_1\zeta_2)^{1/2}lJ\sigma_0\tau_c(T\Delta)^{1/2}, \tag{30}$$

where $l=n_0\sigma_0L$ is the dimensionless optical thickness; σ_0 is the absorption cross section of resonant photons; L is the cell length; J is the density of the photon flux in a probe beam; τ_c is the characteristic time of the investigated correlations. In the case under discussion the value of τ_c is of the order of the time $\bar{\tau}$ taken by an atom to cross the light beam. The estimate represented by Eq. (30) is obtained on the assumption that the probabilities of photon absorption by the first and second detector are the same and amount to 1/2. Reliable detection of a resonance is possible when Δ is much less than τ_c^{-1}. Substituting in Eq. (33), by way of parameters, the values $\zeta_1\sim\zeta_2\sim1$, $l\sim0.1$, $J\sim10^{15}$ cm$^{-2}\cdot$s^{-1} (when the radiation source intensity is $I\sim1$ mW/cm^2), $\sigma_0\sim10^{-11}$ cm^2, $\tau_c\sim\Delta^{-1}\sim10^{-5}$ s (transit regime in the case of a beam with the transverse size $a\sim0.1$ cm), and $T\sim10$ s, we obtain for the parameter η an estimate $\eta\sim10$. There is therefore a considerable reserve in respect of the signal/noise ratio, which is necessary for experimental detection of the predicted resonances.

We shall conclude by noting that in experiments carried out at room or higher temperatures the SIF method has a technique for recording the polarization moments of higher rank which may prove useful in studies of the polarization of cesium atoms ^{133}Cs as a result of the D_1 transition, as well as atoms of rare-earth elements and molecules. In the case of alkali metals it is possible to achieve a resolution of the hyperfine structure of the excited states necessary for the observation of higher-rank polarization moments by performing experiments on atomic beams. At room temperature the SIF method can be recommended as suitable for detection of the alignment in the ground state of alkali atoms.

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APPENDIX

The matrices $S^{KQ}_{K'Q'}(\omega P_z;\varkappa q)$ and $A^{KQ}_{K'Q'}(\omega P_z;\varkappa q)$ occurring in the correlators of the photocurrents described by Eqs. (12) and (13) are defined by considering the interaction of radiation with a medium in the first order of perturbation theory[14-16]:

$$S^{KQ}_{K'Q'}(\omega P_z;\varkappa q)=(-1)^{j+j'+\varkappa}\frac{\Pi_\varkappa^2\Pi_{K'K}}{\Pi_j}\begin{pmatrix}K & \varkappa & K'\\ Q & -q & -Q'\end{pmatrix}\times\begin{Bmatrix}\varkappa & K & K\\ 1 & 1 & 1\end{Bmatrix}\times\begin{Bmatrix}j & j & \varkappa\\ 1 & 1 & j'\end{Bmatrix}\left[\frac{1}{2}\sigma_{jj'}(\omega P_z)+\frac{1}{2}(-1)^{\varkappa+K+K'}\sigma^*_{jj'}(\omega P_z)\right], \tag{A.1}$$

$$A^{KQ}_{K'Q'}(\omega P_z;\varkappa q)=(-1)^{j+j'+\varkappa}\frac{\Pi_\varkappa^2\Pi_{KK'}}{\Pi_j}\begin{pmatrix}K & \varkappa & K'\\ Q & -q & -Q'\end{pmatrix}\times\begin{Bmatrix}\varkappa & K & K\\ 1 & 1 & 1\end{Bmatrix}\times\begin{Bmatrix}j & j & \varkappa\\ 1 & 1 & j'\end{Bmatrix}\left[\frac{1}{2}\sigma_{jj'}(\omega P_z)-\frac{1}{2}(-1)^{\varkappa+K+K'}\sigma^*_{jj'}(\omega P_z)\right]. \tag{A.2}$$

Here $j=j_1$ and j_2 are the angular momenta of the ground state, j' is the total angular momentum of the excited state,

$$\sigma_{jj'}(\omega P_z)=4\pi k\hbar^{-1}|d_{jj'}|_2[i(\omega-\omega_{j'j}-kP_z)+(\gamma_{j'}+\gamma_j)/2]^{-1}, \tag{A.3}$$

$k=\omega/c$, $d_{jj'}$, is a reduced matrix element of the dipole moment, P_z is the component of the momentum of an atom along the direction of the wave vector $\mathbf{k}\|\mathbf{z}$;; $\gamma_{j'}$, and γ_j are the reciprocals of the lifetimes for j' and j.

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