

Characteristic noise of polarized light that has traversed a resonant gaseous medium

Yu. M. Golubev and L. I. Plimak

Leningrad State University

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It is shown that the noise spectrum of polarized light that has traversed a resonant medium must contain magnetic resonances due to the thermal fluctuations of the medium.

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INTRODUCTION

To observe magneto-optical effects such as quantum beats, the Hanle effect, level crossings, etc. one must have an artificially prepared medium in which there is an initial population difference or definite phase relationships between the Zeeman sublevels.¹ The relaxation of this initial nonequilibrium condition manifests itself in the observed resonances. A real medium that has not been specially prepared is a thermal ensemble of moving atoms. Such a system is statistical in nature and all its parameters fluctuate. In this paper we consider the fluctuations of the elements $\rho_{\mu\mu'}$, of the atomic density matrix (μ and μ' label the Zeeman sublevels of the ground state). We may hope that an appropriate measuring procedure will reveal resonances due to relaxation of the random population differences and Hertzian coherences between the Zeeman sublevels. Such relaxation is directly related to the thermal-fluctuation spectrum of the medium, and it is natural to expect it to manifest itself in the noise spectrum of radiation that has passed through the medium.

Aleksandrov and Mamyrin² have observed a resonance associated with fluctuations of the total population of a level. In a paper published in the present issue of the Journal,³ Aleksandrov and Zapasskii report observation of magnetic resonances associated exclusively with parameters differing randomly from zero. Sodium vapor was chosen as the test material in that study, the vapor being traversed by plane polarized light from a dye laser at a frequency for which the vapor is transparent. The measuring system recorded random rotations of the polarization plane (random circular birefringence). The effect was recorded so reliably that the authors could speak of the birth of a new specific method for magnetic resonance research.

Before undertaking their experiment, Aleksandrov and Zapasskii made a qualitative estimate of the magnitude of the effect to be expected (see Ref. 3) and obtained quite encouraging results. At the same time, it is known from results of other studies^{4,5} that only predictions based on quantum electrodynamics (QED) can be regarded as entirely reliable in studies of radiation noise. That is one of the reasons why we undertook the calculations presented here. The agreement of our results with the qualitative estimates served as an additional reason for undertaking the new experiment.³

Let us discuss a possible mechanism for the appear-

ance of resonances in the noise spectrum of light that has traversed the medium. An effect may be expected from correlations between two events in which light is scattered by the same atom.¹⁾ In probing in the transparent region, when an atom is excited virtually to the upper level, its state after the first scattering event for a specified polarization of the light is unambiguously related to the initial state, so that the two scattering events are correlated. This effect has an essentially classical nature and is related to the excitation of a dipole at an optical frequency. The situation in an absorption band is more complicated. Here it is necessary in principle to take into account the real excitation of an atom to the upper level followed by spontaneous emission, and in this case the atom may partially or completely "forget" its initial state (the loss of memory will be complete, for example, if the upper level has zero angular momentum).

Additional terms in the noise spectrum must depend on the angle throughout which the spontaneous emission is collected. We shall assume this angle to be small (in Ref. 3, for example, it was of the order of 10^{-3} – 10^{-5}) and shall neglect corrections necessitated by re-emission (see the Appendix). The effect we are interested in thereby becomes the same, in principle, both in the transparent region and in the absorption band. Qualitatively, of course, the results will be different. We note that in practice, the difference between the absorption and the transparent region is associated primarily with the fact that an intense light source cannot be used in the absorption band since nonlinear effects that we do not take into account could substantially alter the results.

THEORY

Here we present detailed calculations of the noise spectrum of polarized radiation that has passed through a gaseous medium; the calculations will be carried through for two cases: 1) the medium is transparent to the incident radiation (the spectrum of the radiation is concentrated in the Lorentz wing of the absorption band), and 2) the incident radiation falls in the absorption band (the spectrum of the incident radiation is symmetric with respect to the absorption band). The following assumption will be essential in both cases: all the characteristic frequency intervals of the problem (relaxation constants, Zeeman splittings, the Doppler width, the reciprocal of the characteristic time

for an atom to pass through the beam) are small as compared with the large parameter—the frequency difference between the spectrum of the incident radiation and the absorption band for case 1, and the spectral width of the incident radiation for case 2. In carrying through the calculations we shall use a two-level model of the atom with arbitrary angular momenta j and j_1 for the lower and upper states and shall ignore the fine and hyperfine structures.

As was noted in the Introduction, our calculations must be consistently based on quantum electrodynamics (QED). The QED calculation is presented in the Appendix. One of its consequences turns out to be that a simple semiclassical model, which we shall now describe, is adequate. We express the single-atom statistical tensor ρ_q in the form

$$\rho_q^*(\mathbf{R}, t) = \langle \rho_q^*(\mathbf{R}, t) \rangle + \delta \rho_q^*(\mathbf{R}, t), \quad (1)$$

i. e., as the sum of the average and the random part. For a gas in equilibrium

$$\langle \rho_q^* \rangle = \frac{n}{2j+1} \delta_{q0} \delta_{q0},$$

where n is the equilibrium atomic density. We shall use the polarization-moment representation¹ from the very beginning. The quantity $\delta \rho_0^0$ is the fluctuation of the number of atoms in the ground state. The fluctuations $\delta \rho_{0, \pm 1}^1$ and $\delta \rho_{0, \pm 1, \pm 2}^2$ we shall call the random orientation and the random alignment. Our problem will be, in particular, to determine what types of fluctuations contribute to the noise spectrum in various situations.

Let light with polarization e_0 (the unit polarization vector of the light) be incident on the medium. The fluctuations $\delta \rho$ of the medium lead, generally speaking, to fluctuations of the total power and polarization of the transmitted light. If a polarization e_r (the recorded unit polarization vector) is singled out, the instantaneous power of the recorded light will acquire a noise component:

$$I(\mathbf{x}, t) = \langle I(\mathbf{x}) \rangle + \delta I(\mathbf{x}, t). \quad (2)$$

The spectrum of the photocurrent is proportional to the Fourier transform of the quantity

$$G(\tau) = \int d^2\mathbf{x} d^2\mathbf{x}' \langle I(\mathbf{x}, t) I(\mathbf{x}', t+\tau) \rangle = \left(\int d^2\mathbf{x} \langle I(\mathbf{x}) \rangle \right)^2 + \int d^2\mathbf{x} d^2\mathbf{x}' \langle \delta I(\mathbf{x}, t) \delta I(\mathbf{x}', t+\tau) \rangle. \quad (3)$$

Here the integration is taken over the photocathode surface.

The relation between the fluctuations δI and $\delta \rho$ can be obtained by solving the classical problem of the passage of a plane wave through a thin layer of a medium having a spatially nonuniform dielectric tensor of the form

$$\epsilon_{ik} = \epsilon^{(0)} \delta_{ik} + \delta \epsilon_{ik}.$$

Here $\epsilon^{(0)}$ is the average complex dielectric constant and $\delta \epsilon_{ik}$ is the instantaneous random anisotropy of the medium, which is linearly related to the fluctuations $\delta \rho$ (see the Appendix for more details).

As a result, we obtain the following relation:

$$\int d^2\mathbf{x} \delta I(\mathbf{x}, t) = -2 \operatorname{Re} \sum_{\mu, \mu', m, m', q} (\mathbf{de}^{(0)})_{\mu m} (\mathbf{de}^{(r)})_{m' \mu'} \times e_0^* e_r (-1)^q (T_{-q})_{\mu \mu'} \int d^3\mathbf{R} \delta \rho_q^*(\mathbf{R}, t) \int dk \frac{I_k(\mathbf{R}_\perp)}{\gamma + i(k_0 - k)}. \quad (4)$$

Here T_q^* is an irreducible tensor operator,¹ V is the volume of the cell containing the gas under study, k_0 is the resonant frequency of the atomic transition, γ is the homogeneous width of the luminescence line at the transition, I_k is the average spectral power of the radiation, which is related to the complete integral

$$2\pi \langle I \rangle = \int dk I_k,$$

\mathbf{d} is the operator for the dipole moment of the atom, and the subscript m labels the Zeeman sublevels of the upper level.

In deriving (4) we took into account only single scattering from inhomogeneities but did not make use of the approximation of an optically thin layer for the passage of the field through the medium; the quantity I_k in the integrand is therefore taken at the point \mathbf{R}_\perp , which is the projection in the direction of the beam of the point \mathbf{R} in the medium onto the photodetector. In addition, in deriving (4) we neglected the static dichroism and birefringence, as is quite permissible in the case of a gas.

By substituting (4) into (3), we express the noise spectrum of the radiation that has traversed the medium in terms of the thermal-fluctuation spectrum of the medium, which is precisely the Fourier spectrum of the correlator $\langle \delta \rho_q^*(\mathbf{R}, t) \delta \rho_q^*(\mathbf{R}', t+\tau) \rangle$. To calculate this we must make certain assumptions concerning the dynamics of the fluctuations in the medium. This is fairly simple for an experimental situation such as was used in Ref. 2, which Kozlov⁶ has discussed theoretically and in which only the fluctuations in the total population of the level were important. In our case this expression arises automatically in the consistent QED calculation presented in the Appendix:

$$\langle \delta \rho_q^*(\mathbf{R}, t) \delta \rho_q^*(\mathbf{R}+\mathbf{r}, t+\tau) \rangle = (-1)^q \delta_{m' m} \delta_{q, -q} \times \frac{(2\kappa+1)n}{(2j+1)^2 \pi^3/2\sigma_z^2} \exp\left(-\frac{r^2}{\sigma_z^2} - i\Omega q \tau - \gamma_m |\tau|\right). \quad (5)$$

Here $\sigma_z = U|\tau|$, U is the most probable thermal velocity of the atoms, Ω is the Zeeman splitting of the level, and the γ_m are relaxation constants that arise as a result of collisions with buffer-gas atoms (and as a result of spontaneous decay if the lower level is not the ground state).

Expression (5) is valid for a classical gas in the weak-isotropic-collision approximation. In deriving it, the diameter of the beam was assumed to be small compared with the mean free path, and this means, in particular, that it must be small compared with the diameter of the cell. In experiments with alkali metal vapors (Refs. 1 and 3, for example) even a small admixture of a buffer gas resulted in the mean free path becoming small compared with the diameter of the beam. In this case we shall assume that the correlator $\langle \delta \rho \delta \rho \rangle$ satisfies the diffusion equation ($\tau > 0$)

$$\left(\frac{\partial}{\partial \tau} + \gamma_m + i\Omega q - \frac{2}{a^2} \frac{\partial^2}{\partial \mathbf{x}^2} \right) \langle \delta \rho_q^*(\mathbf{R}, t) \delta \rho_q^*(\mathbf{R}+\mathbf{r}, t+\tau) \rangle = 0, \quad (6)$$

in which allowance is made for the possibility of relaxa-

tion. This equation is valid for times that are long compared with the mean time τ_0 between collisions. Formula (5) is valid when $\tau \ll \tau_0$, and it should be used as the initial condition in solving Eq. (6). If the mean free path is short compared with the beam diameter, the solution of Eq. (6) is represented by the same function (5), but with $\sigma_r = a|\tau|^{1/2}/2$. It is also applicable only if the diameter of the beam is small compared with that of the cell.

The rest of the calculation was carried through using only standard assumptions. As a result, we obtain

$$\frac{G(\tau)}{I_0^2} = 1 + 9(2j+1)^2 \xi^2 \frac{2S}{\pi\sigma_r^2 + 2S} \times \sum_{\kappa=0}^2 \left\{ \begin{matrix} 1 & 1 & \kappa \\ j & j & j \end{matrix} \right\} \sum_{q=-\kappa}^{\kappa} Z^{*q}(\mathbf{e}_0, \mathbf{e}_r) \cos \Omega q \tau \exp(-\gamma_{\kappa}|\tau|). \quad (7)$$

Here

$$I_0 = \int d^2\mathbf{x} \langle I(\mathbf{x}) \rangle$$

is the total average light power at the photodetector with allowance for absorption in the medium. We assumed a Gaussian intensity distribution in the beam cross section and defined the effective area S of the cross section by the relation $I_0 = S \langle I(0) \rangle$, where $\langle I(0) \rangle$ is the intensity at the center of the beam. The parameter ξ differs, depending on whether the frequency of the incident light lies in the transparent region or in the absorption band. For the transparent region $\xi = \Delta\Phi N^{-1/2}$, i. e., it is the product of the total average phase advance in the medium by the relative fluctuation of the number of particles in the beam [$\Delta\Phi = k_0(n_0 - 1)L$, $N = nSL$, n_0 is the refractive index, and L is the thickness of the layer]. For the absorption band, $\xi = \beta LN^{-1/2}$, i. e., it is the product of the total absorption in the medium by the relative particle-number fluctuation

$$\beta = \frac{1}{2\pi\Delta\omega} \int dk \beta_k,$$

where β_k is the spectral absorption coefficient per unit length and $\Delta\omega$ is the spectral width of the radiation.

The dependence on the polarizations of the incident and recorded radiations, as well as the dependence on the direction of the magnetic field, is included in the quantity

$$Z^{*q}(\mathbf{e}_0, \mathbf{e}_r) = \left| \mathbf{e}_0 \cdot \mathbf{e}_r \sum_{\mu, \mu'} C_{1\mu' 1\mu}^{*q} e_0^{\mu} e_r^{\mu'} \pm \mathbf{e}_0 \cdot \mathbf{e}_r \sum_{\mu, \mu'} C_{1\mu' 1\mu}^{*q} e_0^{\mu} e_r^{\mu'} \right|^2. \quad (8)$$

The ambiguous sign is to be taken as negative for probing in the transparent region and positive for probing in the absorption band. The components of \mathbf{e}_0 and \mathbf{e}_r should be given in a coordinate system with the Z axis parallel to the magnetic field; the $C_{1\mu' 1\mu}^{*q}$ are Clebsch-Gordon coefficients, and $\{ . . . \}$ is the 6j symbol.

Some useful characteristics of the noise spectrum follow from the identities

$$Z^{*q}(\mathbf{e}_0, \mathbf{e}_r) = Z^{*q}(\mathbf{e}_0, \mathbf{e}_r) = Z^{*q}(\mathbf{e}_r, \mathbf{e}_0) = Z^{*q}(\mathbf{e}_0^*, \mathbf{e}_r^*). \quad (9)$$

DISCUSSION

In the general case, when the polarizations and the magnetic-field direction are arbitrary, the noise spec-

trum has three components: a zero-frequency component, and components at frequencies Ω and 2Ω . The level-population fluctuations ($\kappa=0$) have an effect only when probing in the absorption band ($Z^{00} \neq 0$ in the transparent region) and contribute to the line at zero frequency. The random orientation ($\kappa=1$) contributes at frequencies 0 and Ω , and the random alignment ($\kappa=2$) contributes at frequencies 0, Ω , and 2Ω . The frequency dependence of the contributions is given by the formula

$$g_{*q}(\omega) = \int_{-\infty}^{\infty} \frac{\cos \Omega q \tau}{\pi\sigma_r^2 + 2S} \exp(-\gamma_{\kappa}|\tau| + i\omega\tau) d\tau. \quad (10)$$

The line shape depends on the intensity distribution in the beam cross section; formula (10) is for a Gaussian distribution. If the mean free path is long as compared with the beam diameter, we have $\sigma_r = U|\tau|$, and (10) becomes the convolution of a Lorentz contour of width γ and an exponential with the argument $(2S/\pi)^{1/2}/U$. For the diffusion variant the line shape is even more complicated.

Let us consider a few specific situations in which the shape of the spectrum turns out to be relatively simple. We shall assume the static magnetic field to be either longitudinal or transverse. The incident radiation will be always assumed to be linearly polarized. In the case of a transverse magnetic field, the polarization vector makes the angle φ_0 with the magnetic field vector. Either linear or circular polarization may be recorded. When linear polarization is recorded in the case of a transverse magnetic field, the recorded polarization vector makes the angle φ_r with the magnetic field vector. In the case of a longitudinal magnetic field the angles φ_0 and φ_r are reckoned from an arbitrary direction. In the transparent region these two variants have obvious meanings: recording linear polarization corresponds to recording random rotations of the polarization plane (random circular birefringence), while recording circular polarization corresponds to random ellipticity (random linear birefringence). In the absorption band the situation is more complicated. In this case one cannot associate a definite type of random dichroism with the recording of linear or circular polarizations since here the dichroism is inseparable from the fluctuations of the total absorption of the medium for polarized light (see below).

Below we present the nonvanishing coefficients Z^{*q} for all the experimental variants listed above. We note that the identities (9) permit us to extend the results to several additional situations that arise when the polarizations of the incident and recorded light are interchanged. It follows from this that the result is independent of whether we are dealing with right- or left-hand polarized light.

In the transparent region with a transverse magnetic field and recording linearly polarized light (the case corresponding to the experiment of Ref. 3) we have

$$Z^{*1} = \frac{1}{4} \sin^2 2(\varphi_0 - \varphi_r).$$

For the same situation but recording circularly polarized light,

$$Z^{00} = 1/2 \sin^2 2\varphi_0, \quad Z^{11} = 1/2 \cos^2 2\varphi_0, \quad Z^{22} = 1/2 \sin^2 2\varphi_0.$$

For the transparent region with a longitudinal field and recording linearly polarized light,

$$Z^{00} = 1/2 \sin^2 2(\varphi_0 - \varphi_r).$$

For the same situation but recording circularly polarized light,

$$Z^{22} = 1/2.$$

For the same four cases but in the absorption band, we have: for a transverse field and linear polarization ($\varphi_0 = \varphi_r \equiv \varphi$),

$$Z^{00} = 1/2, \quad Z^{20} = 1/2(1 + 3 \cos 2\varphi)^2, \\ Z^{21} = \sin^2 2\varphi, \quad Z^{22} = \sin^4 \varphi;$$

for a transverse field and circular polarization,

$$Z^{00} = 1/2, \quad Z^{11} = 1/2, \quad Z^{20} = 1/2(1 - 3 \cos^2 \varphi_0)^2, \\ Z^{21} = 1/2 \sin^2 2\varphi_0, \quad Z^{22} = 1/2 \sin^4 \varphi_0;$$

for a longitudinal field and linear polarization,

$$Z^{00} = 1/2 \cos^4(\varphi_0 - \varphi_r), \\ Z^{20} = 1/2 \cos^4(\varphi_0 - \varphi_r), \quad Z^{22} = \cos^2(\varphi_0 - \varphi_r);$$

and for a longitudinal field and circular polarization,

$$Z^{00} = 1/2, \\ Z^{20} = 1/2, \quad Z^{22} = 1/2.$$

Thus, in the transparent region, both in a longitudinal magnetic field and in a transverse one, the random rotations of the polarization plane are due to orientation fluctuations, while the random ellipticity is due to alignment fluctuations. Population fluctuations do not manifest themselves in the transparent region since they do not alter the initial polarization of the light. In the absorption band, the noise spectrum arises both from population fluctuations ($\kappa = 1, 2$). The first are the least interesting since they have already been investigated^{2,6}; moreover, they always contribute at zero frequency, and this is the more difficult region to observe. When linear polarization is recorded, the noise spectrum contains contributions not only from the population fluctuations, but also from alignment fluctuations, and when circular polarization is recorded, it contains contributions from orientation fluctuations, too.

In the absorption band one can observe fluctuations in the total absorption of the medium. This corresponds to an experimental setup in which the light incident on the medium is polarized but the total intensity of the transmitted light is recorded. It can be shown that the spectrum is then still described by Eq. (7), but with $Z^{*q}(e_0, e_0)$ in place of $Z^{*q}(e_0, e_r)$, i. e., an experiment for recording fluctuations of the total absorption turns out to be equivalent to an experiment in which the polarizations e_0 and e_r are identical. This is not accidental. It is not difficult to see that when $e_0 = e_r$ the part of the power fluctuations associated with changes in the polarization corresponds to double (and multiple) scattering by the inhomogeneities of the medium and therefore vanishes in our approximations. This is most clearly evident when $e_0 = e_r$ and both represent linear polarization. It is evident from simple trigonometric considerations that in this case the intensity fluctuations δI

are proportional to $(\delta\varphi)^2$, where $\delta\varphi$ is the instantaneous value of the rotation angle of the polarization plane and is linearly related to the instantaneous inhomogeneity $\delta\rho$ of the medium. Thus, to $\langle(\delta I)^2\rangle$ the polarization fluctuations contribute a term of the order of $\langle(\delta\rho)^4\rangle$, and this corresponds to taking double scattering into account. [We recall that in the calculation we retained only terms of the order of $(\delta\rho)^2$].

It can also be shown that an experiment in which the incident light is unpolarized but light with a definite polarization e_r is recorded corresponds to Eq. (7) with the substitution

$$Z^{*q}(e_0, e_r) \rightarrow Z^{*q}(e_r, e_r),$$

i. e., the feature of interchanging the polarization conditions of the incident and recorded radiations turns out to be valid even in this case.

In concluding, we note that the results of the experiment reported in Ref. 3 can be regarded from the following point of view. The content of the fluctuation-dissipation theorem⁷ amounts to the statement that the dynamics of the relaxation of a system from an artificially produced initial state is identical with the dynamics of fluctuations in that system. Thus, the work of Aleksandrov with Zapasskii³ can be regarded as experimental confirmation of this theorem.

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APPENDIX

In quantum theory, the informative part of the photocurrent spectrum is given by the formula⁸

$$G(\tau) = \int d^3x d^3x' \langle E^+(x, t) E^+(x', t+\tau) E(x', t+\tau) E(x, t) \rangle, \quad \tau > 0. \quad (\text{A. 1})$$

The following is a convenient classification of all the possible beats given by this formula in the approximation of an optically thin layer⁹: beats of the incident radiation with the spontaneous and induced emission of the medium, beats of the induced emission with the induced emission, beats of the spontaneous emission with the induced emission, and beats of the spontaneous emission with the spontaneous emission. In this terminology the principal beats turn out to be those of the induced emission with the induced emission, for those are the ones that produce the structure in the noise spectrum discussed in the main text. The other beats are either small as compared with these or are uninformative. Thus, the beats of the incident radiation with the induced emission produce a structure in the noise spectrum only in higher orders in the intensity of the incident radiation, and in accordance with the conditions of the problem we treat that intensity as a small parameter. The beats of the incident radiation with the spontaneous emission⁹ yield a structure whose characteristic width is determined by the lifetime of the excited state of the atom. In our problem we treat the absorbing state of the atom as the ground state, i. e., its lifetime is long as compared with that of the excited state. Under such conditions the beats of the in-

cident radiation with the spontaneous emission produce an essentially uninteresting background for the main spectrum, whose width is determined by the lifetime of the lower level.

The beats of the induced emission with the spontaneous emission (or of the spontaneous emission with itself) are small, as compared with the principal beats, of the order of the solid angle (or its square) throughout which the spontaneous emission is collected (the acceptance solid angle of the photodetector); moreover, a direct calculation shows that the spectral composition of these beats is roughly the same as that of the principal beats (except for differences in the relative intensities of the lines). We shall assume the acceptance solid angle of the photodetector to be small and shall therefore reject all beats involving spontaneous emission.

For the calculation we shall use the graphical technique of Ref. 10. We shall require the spectral width $\Delta\omega$ of the incident radiation to be large as compared with all the characteristic spectral widths of the problem (Zeeman splitting of the levels, relaxation constants, the reciprocal of the characteristic time for an atom to pass through the beam), except, possibly, the Doppler width kU and the frequency difference Δ . Then, neglecting collisions, the informative part of the noise spectrum corresponds to the following diagrams:

$$G(\tau) - G_0(\tau) = \text{diagram 1} + \text{diagram 2} + \text{c.c.} \quad (\text{A. 2})$$

Here G_0 is the value of G in the absence of the medium; it specifies the noise spectrum of the incident radiation. The full lines in (A.2) represent atomic propagators, and the wavy lines, photon propagators. The short arrows directed into and out from the graphs correspond to the analytic signal of the incident light and its complex conjugate. The open circles represent arguments of the integrand in (A.1), i.e., points on the photocathode; it is understood that they are to be integrated over. In the final analysis, the diagrams must be averaged over the statistics of the incident light.

We are required to establish a correspondence between the semiclassical treatment given in the main text and the expressions presented here. To do this we rewrite the expression corresponding to the diagrams (A.2) in the form

$$G(\tau) - G_0(\tau) = \int d1 d2 d1' d2' \frac{\delta\langle I(t, \rho) \rangle}{\delta\rho(1, 2)} Q(1, 2; 1', 2') \frac{\delta\langle I(t+\tau, \rho) \rangle}{\delta\rho(1', 2')} \quad (\text{A. 3})$$

Here

$$Q(1, 2; 1', 2') = \text{diagram 3}$$

one line being regular and the other irregular

$$\langle I(t, \rho) \rangle = \frac{1}{2} + 2\text{Re} \text{diagram 4} \quad (\text{A. 4})$$

is the average intensity of the transmitted light regarded as a functional of the irregular line, and

$$\rho(1, 2) = \langle \Psi^+(1) \Psi(2) \rangle, \\ 1 = \{R_1, t_1, \mu_1\}, \quad 2 = \{R_2, t_2, \mu_2\}.$$

The variation with respect to ρ must be carried through with allowance for all possible states—not only the symmetric ones. The following expression can be obtained for the variation $\delta\langle I \rangle$:

$$\delta\langle I(t, \rho) \rangle = \frac{ik}{(2\pi)^3} \int dk d^3v d^3R \frac{I_k(R_L)}{k_0 + vk - k + i\gamma} \\ \times \sum_{\mu, \mu'} (\mathbf{de}_0)_{\mu\mu'} (\mathbf{de}_{r'})_{\mu\mu'} (\mathbf{e}_0 \cdot \mathbf{e}_r) \delta\rho_{\mu'\mu}(\mathbf{R}, t, \mathbf{v}) + \text{c.c.}, \quad (\text{A. 5})$$

where \mathbf{v} and M are the velocity and mass of an atom and $\delta\rho_{\mu'\mu}(\mathbf{R}, t, \mathbf{v}) = M^3 \int d^3r e^{iM\mathbf{v}\cdot\mathbf{r}} \delta\rho(1, 2)$ [in which $1 = \{R_1, t, \mu\}$, $2 = \{R_2, t, \mu'\}$, and the integration is taken over $\mathbf{r} = \mathbf{R}_1 - \mathbf{R}_2$ and $\mathbf{R} = (\mathbf{R}_1 + \mathbf{R}_2)/2$] is the variation of the density matrix with respect to the Zeeman sublevels for atoms with the specified momentum $\mathbf{p} = M\mathbf{v}$ and coordinate \mathbf{R} at the time t . All the other notation is the same as in Eq. (4). The fact that the intensity is taken at the photocathode itself and not at an arbitrary cross section of the beam results from allowing for absorption by the medium (see below). Expression (A.4) corresponds to classical forward scattering by a thin layer of a medium having a tensor dielectric constant determined by the subdiagram



This can be shown in essentially the same way as Zubarev⁷ showed it for an isotropic medium. Expression (A.5) reduces to Eq. (4) if one can neglect $\mathbf{k} \cdot \mathbf{v}$ as compared with the frequency difference or the spectral width of the incident radiation, as we did in the main text.

It is now obvious that for complete correspondence between the quantum and semiclassical calculations it is sufficient to introduce the formula

$$\langle \delta\rho_{\mu\mu'}(\mathbf{R}, t, \mathbf{v}) \delta\rho_{\mu''\mu'''}(\mathbf{R}', t', \mathbf{v}') \rangle = M^6 \int d^3r d^3r' e^{iM(\mathbf{v}\cdot\mathbf{r} + \mathbf{v}'\cdot\mathbf{r}')} Q(1, 2; 1', 2'). \quad (\text{A. 6})$$

The digits 1 and 2 have the same meaning here as in (A.4), and 1' and 2' represent analogous sets of primed variables. We can go over to the main text according to the formula

$$\langle \delta\rho(\mathbf{R}, t) \delta\rho(\mathbf{R}', t') \rangle = \int d^3v d^3v' \langle \delta\rho(\mathbf{R}, t, \mathbf{v}) \delta\rho(\mathbf{R}', t', \mathbf{v}') \rangle.$$

Up to now we have completely neglected collisions in all the calculations. In this case, when $k/MU \ll 1$ (the condition for classical motion of the atoms) we have

$$\langle \delta\rho_{q^*}(\mathbf{R}, t, \mathbf{v}) \delta\rho_{q'^*}(\mathbf{R}', t', \mathbf{v}') \rangle = (-1)^{\alpha\delta_{\alpha\alpha'}} \delta_{q-q'} (2\pi)^3 \delta^{(3)}(\mathbf{v}-\mathbf{v}') \\ \times D^{(3)}(\mathbf{R}' - \mathbf{R} - \mathbf{v}(t' - t)) \frac{(2\kappa+1)n}{(2j+1)^2} D^{(3)}(\mathbf{v}) \exp(-i\Omega q(t' - t)), \quad (\text{A. 7})$$

$$D^{(3)}(\mathbf{v}) = \frac{1}{(\pi U)^{3/2}} \exp\left(-\frac{\mathbf{v}^2}{U^2}\right), \quad \delta\rho_{q^*} = \sum_{\kappa, \alpha} (-1)^{\alpha} \delta\rho_{q^*}(T_{-q^*})_{\kappa^* \mu^*} [1],$$

(Ref. 1), which simply corresponds to the transport of an atom with velocity \mathbf{v} from one point to another in the time $t - t'$.

In the approximation of weak isotropic collisions, it is sufficient to multiply (A.7) by $\exp(-\gamma_{\kappa} |t - t'|)$. By convolution of the resulting expression with respect to

the initial and final velocities, we reach formula (5) of the main text for the "straight-through" case with $\sigma_r = U|\tau|$. In the "diffusion" case we shall assume that the atom "forgets" its initial momentum after N collisions and that the time $N\tau_0$, where τ_0 is the characteristic time between collisions, is small as compared with the characteristic dwell time of the atom in the beam. Then the Brownian motion of the atom must obviously be independent of the initial and final momenta. The desired correlator is obtained by multiplying (5) with $\sigma_r = a|\tau|^{1/2}/2$ by $D^{(s)}(\mathbf{v})D^{(s)}(\mathbf{v}')$.

The rest of the calculations are fairly simple. The summations over indices are performed according to formulas given in Ref. 11. It is not difficult to see that in all cases the momentum and spatial integrations in (A.3) can be separated. This is obvious for the diffusion form of the correlator, but it is also true of the "straight-through" form (A.7), since for a plane wave the longitudinal component of the velocity of the atom, which is responsible for the Doppler shift, is independent of the transverse velocity components, which are associated with the spatial correlations. It is also easy to see that the spatial integration, which determines the shape of the spectrum lines, is always the same, and that it is independent of the spectral composition of the incident radiation, but depends only on the intensity distribution in the cross section of the beam and on the ratio of the diameter of the beam to the mean free path (formula 9).

As we have already noted, the calculations can actually be carried through with allowance for real absorption in the medium. Formally, this can be done by replacing formula (A.4) for $I(t, \rho)$ by the series

$$\langle I(t, \rho) \rangle = \frac{1}{2} + 2 \operatorname{Re} \left(\text{diagram 1} + \text{diagram 2} + \dots \right). \quad (\text{A. 8})$$

It is essential that in this case the corresponding diagram series for $G(\tau)$ can be written in the form (A.3). Expression (A.5) has actually been calculated, starting from expression (A.8), for a plane wave. We neglected

the static birefringence and dichroism in that calculation. This is quite permissible for a gas.

¹The situation considered here differs in principle from the case of the noise spectrum of spontaneous emission,⁴ where two different photons are necessarily emitted by different atoms.

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