

Demonstration of the corpuscular nature of light scattered by equilibrium fluctuations in a resonant medium

Yu.M. Golubev and L. I. Plimak

A. A. Zhdanov State University, Leningrad

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The evolution of the statistics of light scattered by the intrinsic noise of an unexcited medium is investigated theoretically. It is shown that the noise spectrum of scattered radiation contains a number of characteristic features (in particular, valleys), the appearance of which is uniquely related to the corpuscular properties of light. The possibility of experimental detection of these features is examined.

INTRODUCTION

There is undoubted interest in experiments in which the corpuscular properties of light can manifest themselves. It is already clear that such experiments are feasible, in particular, within the framework of intensity fluctuation spectroscopy (IFS).^{1–3} The fluctuations can be naturally recognized, for example, by the appearance of a discrepancy between predictions based on wave and corpuscular descriptions of light. Let us examine the following experiments from this point of view. Suppose that polarized light is scattered by an unexcited resonant medium and that intensity fluctuations are observed in light scattered in the direction perpendicular to the incident beam. The excited light is σ_+ -polarized and observations involve the detection of π -polarization (the quantization axis lies along the incident beam). We shall assume, for simplicity, that the angular momenta of ground and excited states are both equal to $1/2$. In accordance with the selection rules, the only atoms that will participate in producing the scattered light will be those occupying the sublevel of the ground state with magnetic quantum number $\mu = -1/2$.

We shall show that the wave and the corpuscular phenomenologies led to diametrically opposite predictions. From the semiclassical point of view (in which light is looked upon as a wave, i.e., is treated quasiclassically, and the medium quantum-mechanically), the scattered-light intensity will follow continuously the population of the $\mu = -1/2$ level, so that fluctuations in its population should lead to the appearance of a characteristic feature in the noise spectrum of the scattered light. From the corpuscular point of view, on the other hand, the experiment is naturally treated as scattering of a Poisson stream of incident photons by atoms that are not correlated with one another. Correlations in the scattered photon flux can then appear only to the extent to which one atom can scatter several photons. Moreover, an atom that has scattered a photon with the required polarization is found to occupy the $\mu = 1/2$ sublevel and cannot again interact with the exciting light. Consequently, the scattered photon flux will also be Poissonian, and the noise spectrum of the scattered light will not contain any special features. This contradiction between the two predictions shows that at least one of the two models of the electromagnetic field (the wave or the corpuscular model) is not acceptable. Since it is not clear *a priori* which is to be preferred, a calculation

has to be performed within the framework of quantum electrodynamics.

Precisely the same situation was encountered in the analysis of noise in the spontaneous emission of a gas.² Three methods were used in that case, namely, wave, corpuscular, and quantum-mechanical, to show that the corpuscular model of light was the most suitable for the qualitative explanation of the experiment (although it does not provide an explanation of *all* the details in the noise spectrum). On the other hand, the semiclassical description is valid for the noise in the transmission of an unexcited medium.^{4,5} It is therefore important to emphasize that an *a priori* choice of the model of light is not actually possible.

The experiment discussed in Ref. 2 is outwardly similar to our own: in both cases, one is essentially concerned with the observation of light scattered at right-angles. Our experiment differs in that, firstly, we shall consider polarized radiation and, secondly, we shall be interested in the possibility of using this method to detect fluctuations in the ground state of the medium. (The authors of Ref. 2 attempted the detection of fluctuations in the excited state. Such fluctuations are readily separated experimentally because of the difference in the characteristic times.)

IFS SIGNAL FOR SCATTERED LIGHT

We shall suppose that a Boltzmann gas of atoms placed in a constant magnetic field \mathbf{H} is illuminated by light of broad spectral composition and polarization \mathbf{e}_0 , and that there is a resonance between this radiation and a transition from the ground state. Observations are performed in a direction that is not parallel to the wave vector \mathbf{k} of the exciting radiation, and the detector responds to polarization \mathbf{e} . The angular momenta of the ground and excited states are, respectively, j_0 and j , and the Zeeman splitting is Ω_0 and Ω .

In IFS, the recorded signal is the power spectrum measured by a photodetector, and quantum electrodynamics predicts that this is given by⁶

$$G_{\mathbf{e}} = \int d\tau e^{i\omega\tau} G(\tau), \quad G(\tau) = \int_S d^2\mathbf{x}_1 d^2\mathbf{x}_2 \langle E^+(\mathbf{x}_1, t) E^+(\mathbf{x}_2, t+\tau) E(\mathbf{x}_2, t+\tau) E(\mathbf{x}_1, t) \rangle, \quad (1)$$

where S is the surface area of the photocathode and E is the positive-frequency part of the Heisenberg field operator corresponding to the polarization \mathbf{e} . This signal is observed

against the background of shot noise produced by the photodetector, which turns out to be important in estimates of the signal-to-noise ratio.

To evaluate (1), it is convenient to use the graphical method of Perel' and Konstantinov,⁷ as was done in our previous paper.⁴ We emphasize that, in Ref. 4, we considered the correlation between scattered and exciting light (the former measured in transmission), whereas here we are concerned with the self-correlation of scattered light (the incident beam does not reach the detector). In contrast to Ref. 4, the important graphs will be

(2)

Before (2) can be evaluated, we must introduce a number of assumptions about the temporal intervals of the problem. We shall suppose that the shortest will be $(kU)^{-1}$ and $(\Delta k)^{-1}$, where kU is the inhomogeneous width of the atomic line and Δk is the width of the illuminating spectrum, whereas the longest times will be the times taken by an atom to traverse the diameter of the beam, γ^{-1} , the ground-state coherence lifetime, $(\gamma^\kappa)^{-1}$, and the characteristic time for the excitation of the atom by the incident radiation, τ_0 , where γ^κ are the multipole moment relaxation constants in the ground state. For the unexcited gas, $\gamma^0 = 0$ and the γ^κ can be nonzero for $\kappa \geq 1$; τ_0 is the reciprocal of the excitation probability of an atom per unit time, which is of the order of the product of the line strength and the spectral density of the radiation. The homogeneous width γ_1 (longitudinal) of the upper state will be considered to be of intermediate magnitude, i.e.,

$$(kU)^{-1}, (\Delta k)^{-1} \ll \gamma_1^{-1} \ll \gamma^{-1}, (\gamma^\kappa)^{-1}, \tau_0.$$

These conditions not only simplify the calculations but also enable us to separate in the spectrum given by (1) the effects due to the lower level from those due to the upper level, according to their spectral widths. We shall also assume that $\Omega \ll \gamma_1$ and $\tau_0 \gg \gamma^{-1}$. The latter condition will enable us to use perturbation theory and confine our attention to the lowest nonvanishing approximation in the incident intensity.

Evaluation of the graphs (2) leads to the following expression for the power spectrum:

$$G_\omega = \frac{\langle P \rangle^2}{N} \sum_{q, \kappa} \{ S^{\kappa q} L_\kappa(\omega - q\Omega_0) + R^{\kappa q} D_\kappa(\omega - q\Omega_0) \}, \quad (3)$$

where $\langle P \rangle$ is the total mean scattered power at the photodetector, N is the characteristic number of atoms in the region of observations, i.e., in the illuminated portion of the cell that is "seen" by the photodetector, and $|q| \leq \kappa \leq 4$.

The expressions for the spectral profiles L_κ and D_κ depend on the geometry of the experiment, the constants γ^κ , and the mean free path of an atom. They are given in the Appendix for simple physical situations, since they may turn out to be useful in the design and analysis of an actual experiment. The specific form of $L_\kappa(\omega)$ and $D_\kappa(\omega)$ is unimportant for our purposes here. We shall suppose that

$$L_\kappa(\omega) + iD_\kappa(\omega) = 2(\tilde{\gamma}^\kappa + i\omega)^{-1}, \quad (4)$$

where $\tilde{\gamma}^\kappa = \gamma^\kappa + \gamma$. This is equivalent to a model in which the escape of the atom from the beam as a result of drift can be simulated by exponential decay with the constant γ .

The relative line intensities in the noise spectrum are described by the coefficients $S^{\kappa q}$ and $R^{\kappa q}$, for which

$$S^{\kappa q} + iR^{\kappa q} = Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}) [Z^{\kappa q}(\mathbf{e}, \mathbf{e}_0)]^*. \quad (5)$$

The quantity $Z^{\kappa q}$ contains the polarization conditions of the experiment:

$$Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}) = \frac{1}{(2\kappa+1)^{1/2}} \frac{\sum (\mathbf{d}\mathbf{e}_0^*)_{\mu m} (\mathbf{d}\mathbf{e})_{m\mu'} (\mathbf{d}\mathbf{e}^*)_{\mu' m'} (\mathbf{d}\mathbf{e}_0)_{m' \mu} C_{j\mu' m' \kappa q}^{j\mu \mu \kappa}}{\sum (\mathbf{d}\mathbf{e}_0^*)_{\mu m} (\mathbf{d}\mathbf{e})_{m\mu'} (\mathbf{d}\mathbf{e}^*)_{\mu' m'} (\mathbf{d}\mathbf{e}_0)_{m' \mu}}, \quad (6)$$

where $C_{j\mu' m' \kappa q}^{j\mu \mu \kappa}$ are the Clebsch-Gordan coefficients, the sums are evaluated over the indices of the lower (μ) and upper (m) levels, and d is the dipole moment.

In general, the noise spectrum (3) consists of five lines at frequencies $k\Omega_0$, $k = 0, 1, 2, 3, 4$. Thus, it contains information not only about the population, orientation, and alignment constants in the ground state as, for example, in Ref. 4, but also about the relaxation constants of the octupole ($\kappa = 3$) hexadecapole ($\kappa = 4$) moments of the density matrix. This is related to the fact that the absorption and emission of polarized photons can be accompanied by a resultant change in the magnetic number of up to $|\Delta\mu| = 4$. It is readily shown that both R^{00} and $R^{\kappa 4}$ are zero, i.e., the lines at zero frequency and the frequency $4\Omega_0$ are always symmetric. For practical calculations, it is useful to note that the quantities $Z^{\kappa q}(\mathbf{e}_0, \mathbf{e})$ and $Z^{\kappa q}(\mathbf{e}, \mathbf{e}_0)$ behave as irreducible tensors of rank κ and transform in terms of the D -functions⁸ under rotations of the magnetic field. Hence, it follows that $\Sigma_q S^{\kappa q}$ and $\Sigma_q R^{\kappa q}$ do not change under rotation of the magnetic field. When illumination and (or) observation are performed with mixed polarization, the averaging over \mathbf{e}_0 and (or) summation over \mathbf{e} should be performed separately for the sums in the numerator and denominator of (6).

For the case described briefly in the Introduction (exciting beam with σ_+ -polarization and observations of π -polarization, $j_0 = j = 1/2$), the expression given by (3) assumes the very simple form

$$G_\omega \propto L_0(\omega) - L_1(\omega). \quad (7)$$

It vanishes when the decay of the coherence of the ground state of the atom is unimportant (for $\gamma^1 = 0$).

CLASSICAL APPROACH TO THE DESCRIPTION OF THE SCATTERED-LIGHT NOISE

The above results exhaust the problem from the formal point of view. Nevertheless, we shall also provide a semiclassical calculation in order to elucidate the reasons for the difference between the predictions of quantum electrodynamics and semiclassical physics. The point is that this calculation involves certain methodological points that are of independent interest.

The semiclassical analysis is based on the following considerations. Let us suppose, for simplicity, that the spectral

density of the exciting radiation $I(\mathbf{x})$ is constant within the Doppler absorption profile. If $\rho_{\mu\mu'}(\mathbf{x})$ is the density matrix of ground-state atoms at the point \mathbf{x} , the power emitted into the solid angle Φ with polarization \mathbf{e} is given by the obvious formula (see Ref. 9)

$$P = \frac{\Phi}{4\pi} \frac{1}{\gamma_1} \sum_{m,\mu} (\mathbf{de}_0)_\mu (\mathbf{de})_{\mu\mu'} (\mathbf{de}')_{\mu'\mu} (\mathbf{de}_0)_{\mu'\mu} \times \int_V d^3\mathbf{x} I(\mathbf{x}) \rho_{\mu\mu'}(\mathbf{x}). \quad (8)$$

Assuming that fluctuations in $\rho_{\mu\mu'}$ are adiabatically slow (this is undoubtedly the case for the chosen ratios of the characteristic times of the problem), we shall use (8) not only for the mean scattered power, but also for its instantaneous value. This is completely justified within the framework of existing traditions. It is important to recognize, however, that it is precisely at this point that we have a departure from the quantum-mechanical description because the formulation of the theory in terms of random c -number functions is, of course, essentially classical.

Transforming to the tensor representation of the density matrix, and using the same assumptions as in (3)–(5), we find from (8) that

$$\frac{\delta P}{\langle P \rangle} = (2j_0 + 1)^{1/2} \sum_{\kappa q} Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}) \frac{\delta \rho_q^{\kappa*}}{N}. \quad (9)$$

From the classical point of view, the IFS signal is identical with the power spectrum of the scattered radiation:

$$G_\omega = \int d\tau e^{i\omega\tau} \langle \delta P(t) \delta P(t+\tau) \rangle = (2j_0 + 1) \frac{\langle P \rangle^2}{N^2} \sum_{\kappa q} Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}) Z^{\kappa' q'}(\mathbf{e}_0, \mathbf{e}) \times \int d\tau e^{i\omega\tau} \langle \delta \rho_q^{\kappa*}(t) \delta \rho_{q'}^{\kappa'}(t+\tau) \rangle. \quad (10)$$

As can be seen, this formula relates the dynamics of natural fluctuations of the scattering medium and of the scattered radiation. The semiclassical calculation is thus essentially based on the idea of light as the detector of the state of the medium.

The explicit expression for the mean $\langle \delta \rho_q^{\kappa*}(t) \delta \rho_{q'}^{\kappa'}(t+\tau) \rangle$ was derived in Ref. 4 from the consistency conditions for the semiclassical and quantum-mechanical approaches. We shall show that it can actually be derived from very general considerations. In the case under consideration, the equation for the density matrix is

$$(\partial/\partial t + \tilde{\gamma} + i\Omega_0 q) \rho_q^{\kappa}(t) = 0. \quad (11)$$

According to the fluctuation-dissipation theorem,¹⁰ this equation is also satisfied by $\langle \delta \rho \delta \rho \rangle$, so that

$$\langle \delta \rho_q^{\kappa*}(t) \delta \rho_{q'}^{\kappa'}(t+\tau) \rangle = \exp[(-\tilde{\gamma} - i\Omega_0 q')\tau] \langle \delta \rho_q^{\kappa*}(t) \delta \rho_{q'}^{\kappa'}(t) \rangle. \quad (12)$$

Consideration of spherical symmetry determines the simultaneous correlator in the form

$$\langle \delta \rho_q^{\kappa}(t) \delta \rho_{q'}^{\kappa'}(t) \rangle = (-1)^q \delta_{-q, -q'} \delta_{\kappa, \kappa'} F^{\kappa}. \quad (13)$$

To find F^{κ} , we must use the fact that $\delta \rho_0^{\kappa}$ is a linear combination of fluctuations in the populations of Zeeman sublevels, so that, provided the temperatures are not too low, ($kT \gg j_0 \Omega_0$), we find from classical considerations that $F^{\kappa} = N/(2j_0 + 1)$. We have thus shown that the dynamics of fluctuations in the medium can always be determined by starting with the fluctuation-dissipation theorem, the considerations of spherical symmetry, and correspondence with classical statistics.

From (10)–(13), we obtain ($|q| \ll \kappa \ll 4$)

$$G_\omega = \frac{\langle P \rangle^2}{N} \sum_{q, \kappa} |Z^{\kappa q}(\mathbf{e}_0, \mathbf{e})|^2 L_\kappa(\omega - q\Omega_0). \quad (14)$$

Let us now compare the results of the semiclassical (14) and the quantum-mechanical (3) calculations. Both predict that the noise spectrum of the scattered radiation has a structure consisting of five components with frequencies $k\Omega_0$, $k = 0, 1, 2, 3, 4$. Both predict the same order of magnitude for the strength of the signal. However, even cursory inspection of (3) and (14) will show that they are different: (14) consists only of Lorentz peaks, whereas (3) contains dispersion profiles and, secondly, the Lorentz peaks can be nonpositive, i.e., they can become valleys. A valley of this kind is indicated, for example, by (7). For this case, the semiclassical theory yields

$$G_\omega \propto L_0(\omega) + L_1(\omega), \quad (15)$$

i.e., instead of the difference as in (7), we have the sum of the profiles. If we neglect relaxation of atomic coherence in the ground state ($\gamma^1 = 0$), the quantum-mechanical calculations predict the absence of structure in the noise spectrum, whereas the semiclassical treatment predicts that structure should be present. This confirms the discussion given in the Introduction.

We note that (3) and (14) differ not only quantitatively, but also qualitatively in their interpretation. In accordance with (10), each κq -component in (14) is the consequence of fluctuations in a component of the tensor ρ_q^{κ} , that occur in the medium independently of light. We recall that this interpretation was also valid in the description of forward scattering.^{4,5} On the other hand, for light scattered laterally, the κq -components do carry information about the kinetics of the tensor ρ_q^{κ} , but are not the consequence of only the natural fluctuations of the medium, but represent the mutual interaction between light and medium in the scattering process, which is connected with the quantum-mechanical nature of the process of measurement.

We also draw attention to the fact that our results are essentially connected with the assumption that the one-atom correlations in the medium play a dominant role. Were the multi-atomic correlations to play the dominant role, the transition from (8) to (9) could turn out to be correct, and the semiclassical treatment would yield the same result as the quantum-mechanical. This happens, for example, if we introduce correlations into the medium by an external noise agency, as in Ref. 2. From this point of view, the experiment reported in Ref. 2 constitutes a demonstration of the different role of single-atom and many-atom correlations.

POSSIBILITY OF OBSERVATION

At present, it is unlikely that a detailed experimental examination could be made of the structure of (3), which would enable us to consider a quantitative comparison between theory and experiment. We shall therefore discuss yes-no type experiments in which the absence or presence of some particular detail of the noise spectrum will enable us to draw a qualitative conclusion. For example, it may be possible to verify the absence of the structure (7) under collisionless condition (for $\gamma^1 = 0$). Although this experiment is attractive by virtue of its simplicity, it is of little practical interest because it involves measurements at near-zero frequencies, which are technically difficult. The other possibility of this kind is connected with the presence, in general, of negative components in (3). For example, if the magnetic field is at an angle ϕ to the incident beam of light, Eq. (7) assumes the form

$$G_{\omega} \propto L_0(\omega) - L_1(\omega) \cos^2 \varphi^{-1/2} [L_1(\omega - \Omega_0) + L_1(\omega + \Omega_0)] \sin^2 \varphi, \quad (16)$$

i.e., a valley appears at the Zeeman frequency. When the level angular momenta differ from 1/2, the conditions for the appearance of valleys at frequencies that are multiples of Ω_0 may be different, but they are characteristic only of the quantum-mechanical calculation, and it is natural to look for them. The detection of a valley of this kind, even with the minimum precision, would be a possible way of demonstrating the corpuscularity of light that we are looking for.

In an actual experiment, detection noise is connected with fluctuations in the level of the shot background, and the signal-to-noise ratio is expressed¹¹ in terms of the ratio of the strength of the useful signal to the average level of the shot background η . This ratio is found to be equal to $\eta(\Delta\omega\Delta T)^{1/2}$, where $\Delta\omega$ is the transmission band of the spectrum analyzer, ΔT is the signal acquisition time, $\eta \sim qG_{\omega}/\langle P \rangle k$, and q is the quantum yield of the photodetector (see, for example, Ref. 6).

To estimate η , it is convenient to write

$$\eta \sim q\xi w\Phi/4\pi. \quad (17)$$

where $w = (\gamma\tau_0)^{-1}$ is the nonlinearity parameter of our problem (assumed small) and ξ is a coefficient representing the fact that not all the atoms are excited by light of polarization e_0 and not all atoms emit polarization e [the coefficient ξ is actually the denominator in (6) divided by d^4]. In deriving (17), we used the approximate relationships

$$\langle P \rangle \sim Nk \frac{\Phi}{4\pi} \xi \tau_0^{-1}, \quad G_{\omega} \sim \frac{\langle P \rangle^2}{N\gamma}.$$

When $\Delta\omega \sim 10^5 \text{ s}^{-1}$, which was the case in the experiments described in Refs. 2 and 5, and $\Delta T \sim 10^3 \text{ s}$, the detection level (signal-to-noise ratio of the order of unity) corresponds to $w\Phi/4\pi \sim 10^{-2}$. This is a relatively stringent condition because when observations are performed in polarized light, $\Phi/4\pi$ is probably no more than 10^{-1} . If, on the other hand, observations are performed in unpolarized light, one can achieve a signal-to-noise ratio of the order of 10, since $\Phi/4\pi \sim 1$ is then acceptable. (It is readily verified by direct calculation that, for observations in unpolarized light,

the lines at nonzero frequencies remain, although they become symmetric and possibly positive.) This possibility may be useful for the primary detection of the signal.

Another possible way of increasing the signal-to-noise ratio may be to increase the output of the source of light and go over to the nonlinear region $w \gtrsim 1$. Since perturbation theory cannot then be used, we must introduce certain corrections into the calculation. If we suppose that $w \ll \gamma_1/\gamma$, it is sufficient to take into account the effect of the field on the density matrix of the ground state. The graphical representation of G assumes the form

where the shaded block corresponds to the development of the density matrix of the ground state. It satisfies the equation

When $\gamma \ll \gamma_1$ this equation can be written down for both a broad excitation spectrum and a monochromatic spectrum.

Equation (18) was analyzed for the simple cases $j_0 = j = 1/2$ and $j_0 = 1, j = 0$. It was found that, in the nonlinear region, the signal-to-noise ratio was of the order of $q\xi\Phi/4\pi$ for lines at nonzero frequencies. (The fact that the estimate does not contain the dimensionless power is due to the fact that, for $w \gtrsim 1$, the increase in the linewidth in the noise spectrum begins to set in and compensates the increase in the useful signal.) The signal to noise ratio for observations in unpolarized light can thus be of the order of 100.

Of course, unless specific calculations are performed, it is not clear whether the valleys in the noise spectrum remain for $w \gtrsim 1$, although this is expected for $w \sim 1$. In any case, the use of powerful exciting radiation should also improve the signal-to-noise ratio for the primary detection of the signal.

APPENDIX

When the actual geometry of the experiment and the motion of the atoms are taken into account, the profiles L_x and D_x have the form

$$L_x(\omega) + iD_x(\omega) = \int d\tau K(\tau) \exp[(-\gamma^* + i\omega)\tau],$$

where

$$K(\tau) = \text{const} \int d^3x d^3x' I(x) I(x') \frac{1}{\pi^{3/2} \sigma_\tau^3} \exp\left[-\frac{(x-x')^2}{\sigma_\tau^2}\right],$$

$$K(0) = 1.$$

In these expressions, $I(x)$ is the spatial distribution of the spectral intensity of the incident light, which is assumed to be the same for all the lines, i.e., the spatial and spectral

dependence of the intensity can be factorized out. The space integrals are evaluated over the illuminated part of the cell that is seen by the photodetector. Its dimensions are assumed to be small both in comparison with the dimensions of the cell and with the distance between the cell and the photodetector. The quantity σ_τ depends on the assumptions made about the kinetics of the atomic density matrix. If the relaxation time of the momentum distribution is long in comparison with the time of relaxation of atomic coherence, the motion of the atoms may be looked up on as rectilinear and $\sigma_\tau = U_\tau$. When the opposite assumption is made (this occurred, for example, in the experiments reported in Ref. 5), the motion of the atoms can be looked upon as diffusion and $\sigma_\tau = 2a\tau^{1/2}$, where a^2 is the diffusion coefficient. These two cases correspond precisely to the transit and diffusion approximations in Ref. 4, where all the technical details of the way in which the motion of the atoms is taken into account can be found.

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