

Intensity-fluctuation spectroscopy of optical fields with non-Gaussian statistics

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Intensity-fluctuation spectroscopy (IFS) is usually considered to be complementary to conventional spectroscopy and capable of removing technical restrictions on the resolving power. However, the information provided by field spectroscopy is identical to that obtained by IFS only for fields with Gaussian statistics. For non-Gaussian fields, IFS yields essentially new information, and the present review is devoted to this aspect of IFS. It surveys experiments concerned with the investigation of the noise spectrum of resonance fluorescence and of coherent forward scattering by an atomic vapor, which provide data on the width and the structure of levels involved in atomic transitions under the conditions of dominant Doppler broadening. Fundamental and technical limitations of the method are examined. Analogous studies of fluctuation spectra of radiation scattered by macroparticles in liquids can be used to determine the time dependence of the particle form factor independently of the characteristics of translational diffusion.

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1. INTRODUCTION

The intensity of any electromagnetic radiation always fluctuates in time. Until recently, such fluctuations were looked upon as mere noise, affecting problems in research, communications, radar, and so on. However, work performed in the course of the last few years has shown that studies of the statistical properties of these fluctuations can be successfully used to yield information not only equivalent to that produced by conventional field spectroscopy, but also complementing it in a fundamental way. The present review is devoted to researches in which fluctuation spectroscopy was used to obtain data on the dynamics of radiating systems that cannot be obtained by conventional spectral analysis.

The particular branch of spectral analysis that we shall survey here has a number of different designations. We shall frequently refer to it as "intensity-fluctuation spectroscopy (IFS)" or "noise spectroscopy." A number of other designations is used in the literature, for example, "optical mixing spectroscopy," "light beating spectroscopy," "optical heterodyne and homodyne spectroscopy," and so on. None of these phrases is all-embracing, and each is employed in appropriate situations, so that we reserve the right to use them as the need arises.

Let us compare IFS with traditional spectroscopy. In the traditional spectroscopic experiment, radiation is resolved into a spectrum by some dispersing element such as a prism, diffraction grating, and so on, or it is made monochromatic by a suitable filter, such as a resonator, a selective absorber, and so on. The result of perfect monochromatization is described by the quantity

$$E_{\omega} = \frac{1}{2\pi} \int E(t) e^{i\omega t} dt, \quad (1)$$

i. e., the Fourier transform of the field strength $E(t)$. Spectral analysis is concluded by using a photodetector to determine the intensity of the spectrum which, in accordance with the Wiener-Khinchin theorem, is given by

$$I_{\omega} = \int \langle E(t) E^*(t + \tau) \rangle e^{i\omega\tau} d\tau. \quad (2)$$

In contrast to the *spectral intensity* I_{ω} recorded in conventional spectroscopy, noise spectroscopy investigates the *power spectrum*. This transposition of words reflects the transposition of functional elements in the procedure of measurement: whilst, in conventional spectral analysis, the analytical element is followed by the intensity-measuring photodetector, in noise spectroscopy the radiation is directly converted into the photodetector signal, and it is this signal that is subjected to spectral analysis. The quantity that is

measured in the latter case is

$$i_{\omega}^2 = \int \langle i(t) i(t+\tau) \rangle e^{i\omega\tau} d\tau, \quad (3)$$

where $i(t)$ is the instantaneous photocurrent. If we suppose that $i(t)$ is proportional to the light intensity $I(t) = |E(t)|^2$, the procedure used to measure the photocurrent spectrum i_{ω}^2 turns out to be equivalent to the measurement of the light power spectrum

$$I_{\omega}^2 = \int \langle I(t) I(t+\tau) \rangle e^{i\omega\tau} d\tau \quad (4)$$

(in actual fact, this is not entirely true because the relationship is more complicated; this question will be examined in the next section).

It is clear that the power spectrum I_{ω}^2 is determined, at least partly, by the field spectrum I_{ω} . For example, if the field consists of only two monochromatic waves, the power spectrum consists of two lines (as does the field spectrum), one of which has zero frequency and the frequency of the other is equal to the frequency difference between the two superimposed fields. Of course, the absolute frequencies of the radiation remain unknown.

The IFS problem has a precise analogy in radiofrequency engineering. If we have a high carrier frequency that is modulated by the relatively low-frequency process under investigation, its spectrum can be obtained by performing a direct spectral analysis of the high-frequency field, and the measuring equipment that is necessary need only have sufficiently high resolution. If the demands imposed on this equipment are excessive, one can begin by first detecting the signal and, having removed the high carrier frequency, perform the analysis of the signal in the low-frequency region with correspondingly lower resolution, but only at the price of a new requirement, namely, sufficiently rapid response of the detector.

Forrester, Gudmundsen and Johnson¹ were the first to investigate the light noise spectrum, using the heterodyne principle. They exposed their receiver to two neighboring mercury lines and found that the photocurrent spectrum contained a peak at the frequency of the beats between these two lines. The purpose of this experiment was purely to demonstrate the effect: the noise peak amounted to only 10^{-4} of the "white" shot-noise background, and was only just detectable. The results obtained by Forrester *et al.*¹ are obvious in the light of present knowledge. However, at the time, they gave rise to considerable discussion in which, in particular, doubts were cast on the very possibility of detection of beats between two independent sources of light. An analogous controversy arose soon after as a result of the investigations of intensity correlations begun by Hanbury-Brown and Twiss,² which became widely known. It is useful to note here that, although the work of Forrester *et al.* and Hanbury-Brown and Twiss² is essentially equivalent, it was discussed quite independently until recently.

The experiments noted above are very difficult to perform because the intensity correlation effect is small. The above ratio of 10^{-4} between the beats and the shot background is typical for conventional sources

of radiation. Noise spectroscopy with such sources did not, therefore, become widely used in practice, except for determinations of correlation areas for stellar light.²

The advent of the laser gave rise to a rapid increase in interest in noise spectroscopy. Because of the high spectral intensity, the power associated with beats produced with laser radiation exceeds the shot-noise level by several orders of magnitude, and this was immediately exploited in the analysis of the mode composition of laser radiation. However, noise spectroscopy found its greatest development in problems involving the analysis of the velocity distribution of particles suspended in liquids and gases, which scatter the incident laser radiation (see Ref. 3 in this connection). Because of the Doppler effect, the scattered radiation is found to be broadened, and this broadening is most effectively analyzed by the IFS method which, in this particular application, is referred to as optical heterodyne or homodyne spectroscopy.

In the foregoing applications, IFS is used to obtain data that can also be obtained by traditional spectroscopy. The difference between noise spectroscopy and conventional spectroscopy is then purely methodological, although the former has definite advantages in the analysis of highly coherent radiation (because practical limitations prevent the use of conventional methods in the analysis of spectral structures of width less than 100 MHz) and in the analysis of spatial correlations over baselines in excess of 10 m.

In addition to methodological advantages, noise spectroscopy has, in some situations, definite fundamental advantages in that it provides information that is not contained in the field spectrum. In fact, the noise spectrum is essentially the spectrum of beats between the different harmonics of the field spectrum, so that it is capable of exhibiting the presence of phase correlations between them, which are entirely absent from the field spectrum. Consider the simple example of harmonically intensity-modulated "white" light, i. e., radiation whose power spectrum has, by definition, a peak at the modulation frequency. When the modulation frequency is less than the total width of the spectrum, conventional spectral analysis will not distinguish between modulated and unmodulated light.

The field spectrum and the power spectrum are described by first- and second-order correlation functions, respectively. Complete description of an arbitrary statistical process involves an infinite number of correlation functions of all orders. On the other hand, it is known that, when the field fluctuations are Gaussian in character, all the correlation functions can be expressed in terms of the pair correlator. Thus, the information content of field and noise spectra is essentially the same in the case of Gaussian fields. It is only for non-Gaussian fields that the noise spectrum yields new information.

The present review is devoted to researches whose aim was to investigate the properties of noise spectra that are connected with the non-Gaussian character of the processes under investigation.

The evolution of these ideas has given rise to an unusual branch of atomic spectroscopy that is distinguished by ultrahigh resolution and is unaffected by the Doppler broadening of spectral lines. Essentially, it is concerned with the internal dynamics of atomic states (beats in the electron density of atoms at frequencies corresponding to transitions between closely spaced energy levels, and the decay of states under the influence of perturbations), which can be studied by investigating fluctuations in the radiation interacting with the atoms.

Analogous problems have also been formulated as part of the investigation of the dynamics of macroparticles in solutions. Here again, the aim has been to study the internal dynamics of the scattering particles, for example, rotation or change of shape against the background of translational motion. Although the objects under investigation, the methodology, and the problems that arise in these two groups of topics are very different, they are, in fact, fundamentally analogous, and this is the reason for combining them in a single review.

2. SEPARATION OF THE NON-GAUSSIAN COMPONENT OF FLUCTUATIONS IN THE ELECTROMAGNETIC FIELD

A. Detection of light intensity fluctuations

When light intensity fluctuations are referred to, it is important to remember that the directly measured quantity is not light itself, but the signal produced by the photodetecting device, for example, the photocurrent. Although the average (over a sufficiently long period of time) photocurrent is proportional to the light intensity, this is hardly the case for the instantaneous values of these quantities. Their relationship has been frequently discussed in the literature.⁴⁻⁷

In quantum theory, the operator

$$i(t) = e \frac{d}{dt} \hat{N}(t) = e \frac{d}{dt} \sum_i \hat{N}_i(t), \quad (5)$$

can be associated with the photocurrent, where $\hat{N}(t)$ is the operator corresponding to the number of atoms in the photocathode that are ionized at time t , $\hat{N}_i(t)$ is an operator with eigenvalues 0 and 1, and e is the charge on the electron. The sum is evaluated over atoms that are ionized at time t .

Let us construct the correlation function

$$\langle i(t_1) i(t_2) \rangle = e^2 \frac{d^2}{dt_1 dt_2} \left(\sum_i \langle \hat{N}_i(t_1) \hat{N}_i(t_2) \rangle + \sum_{i,j} \langle \hat{N}_i(t_1) \hat{N}_j(t_2) \rangle \right). \quad (6)$$

The first term is due to the contributions of individual atoms and, in the final analysis, represents the shot background that is independent of frequency, but is proportional to the mean intensity radiation. The second term describes interference between absorption by different atoms, and represents the information-bearing part of the spectrum (it contains both the Gaussian and non-Gaussian components; see below).

If we solve the quantum-mechanical problem of interaction between radiation and absorbing atoms in the

photocathode within the framework of perturbation theory of the first order in the light intensity, we obtain the following expressions for the spectrum of the photocathode current:

$$i_{\omega}^2 = (i_{\omega}^2)_{\text{shot}} + (i_{\omega}^2)_{\text{inform}}, \quad (7)$$

$$(i_{\omega}^2)_{\text{shot}} = \frac{e^2}{2\pi\omega_0} q \int d^2s \langle E_{\mu}^*(\mathbf{r}, t) E_{\mu}(\mathbf{r}, t) \rangle, \quad (8)$$

$$(i_{\omega}^2)_{\text{inform}} = \frac{e^2}{4\pi^2\omega_0^2} q^2 \iint d^2s_1 d^2s_2 \int d\tau \cdot e^{i\omega\tau} \times \langle E_{\mu}^*(\mathbf{r}_1, t) E_{\nu}^*(\mathbf{r}_2, t + \tau) E_{\nu}(\mathbf{r}_2, t + \tau) E_{\mu}(\mathbf{r}_1, t) \rangle + \text{K.C.}; \quad (9)$$

where E_{μ} is the Cartesian component of the field vector E , the integrals are evaluated over the photocathode area S , repeated subscripts imply summation, and q is the quantum yield of the photodetector.

Comparison of (7)–(9) with (4) will show that the photocurrent spectrum i_{ω}^2 differs from the power spectrum I_{ω}^2 by the presence of the shot background. The latter is homogeneous in frequency and its appearance is related to the discrete nature of the absorption of light by the photocathode. Moreover, the information-bearing part of i_{ω}^2 is not completely equivalent to I_{ω}^2 because, strictly speaking, the correlators $\langle I(t_1) I(t_2) \rangle = \langle E^*(t_1) E(t_1) E^*(t_2) E(t_2) \rangle$ and $\langle E^*(t_1) E^*(t_2) E(t_2) E(t_1) \rangle$ are not identical. There is no difference between them in classical electrodynamics but, in quantum theory, the operators E and E^* can commute only in accordance with certain definite rules.

It follows from (7)–(9) that the photocurrent spectrum is centered not on the optical frequency but on zero frequency, and lies in the radiofrequency range. In many investigations, this has been the basic methodological advantage of IFS, namely, it has provided us with the possibility of analyzing relatively slow processes modulating the field E . The exploitation of this advantage has been facilitated in recent years by advances in instrumentation available for the analysis of low-frequency signals in radio-engineering.

B. Possible reasons for the non-Gaussian nature of radiation statistics

The information-bearing part of the noise spectrum is determined by the second-order correlator $\langle E^*(1) E^*(2) E(2) E(1) \rangle$. It is well known that, for a field with Gaussian statistics,

$$\begin{aligned} \langle E^*(1) E^*(2) E(2) E(1) \rangle \\ = \langle E^*(1) E(1) \rangle \langle E^*(2) E(2) \rangle + \langle E^*(1) E(2) \rangle \langle E^*(2) E(1) \rangle. \end{aligned} \quad (10)$$

Hence, it follows immediately that, in the case of a Gaussian field, the noise spectrum does not contain any information other than that contained in the field spectrum because the fourth-order correlator is expressed in terms of second-order correlators. The noise spectrum is then a convolution of the field spectrum with itself. Thus, if the field spectrum is Lorentzian, the noise spectrum is also a Lorentzian with twice the width.

We shall be interested in effects connected with non-Gaussian properties of radiation, for which (10) is not satisfied, so that the noise spectrum contains information that is complementary to what can be inferred from the field spectrum. Until recently, non-Gaussian effects

appeared to be totally negligible in atomic physics, and there was no doubt as to the reasons for this. In fact, the field due to N sources can be written in the form

$$E(t) = \sum_{i=1}^N E_i(t) \quad (11)$$

for which the average is $\langle I(0)I(\tau) \rangle$, where $I = |E|^2$. The average can be written in the form

$$\langle I(0)I(\tau) \rangle = \frac{1}{2} \sum_i \langle I_i(0)I_i(\tau) \rangle + \sum_{i,j} \langle E_i(0)E_i^*(\tau) \rangle \langle E_j^*(0)E_j(\tau) \rangle + \text{c.c.}, \quad (12)$$

where the first term is the non-Gaussian component of the radiation (of course, subject to the condition that the radiation emitted by each individual source is non-Gaussian), whereas the second term is Gaussian. It is readily seen that the Gaussian component is greater by a factor of N than the non-Gaussian, and this has always served as the basis for scepticism in relation to possible detection of the latter since, in practice, we usually have $N \gg 1$ (we shall later discuss certain situations for which $N \sim 1$ can be achieved).¹⁾ All this was regarded as quite indisputable prior to the advent of the laser. The nonlinear properties of the medium, which manifest themselves in the case of high-intensity electromagnetic fields, introduce essential corrections. First of all, under certain conditions, the laser itself generates non-Gaussian radiation (under mode-locking conditions). However, in addition, it turns out that, under the influence of high-intensity radiation, conventional thermal sources also produce non-Gaussian light. Thus, owing to the nonlinear phenomenon, the non-Gaussian nature of the radiation immediately ceased to be a purely theoretical abstraction. At the same time, it gradually became clear that, even in the absence of any nonlinear effects in the radiation from conventional thermal sources, the detection of non-Gaussian properties was a fully realistic proposition. It eventually turned out that the above ratio of $1/N$ between the non-Gaussian and Gaussian components could be appreciably improved under certain conditions. Some of these possibilities will be discussed below.

We emphasize that the non-Gaussian component of the radiation generated by independent sources is determined by fluctuations in the intensity of the individual sources and, consequently, does not contain any broadenings connected with phase fluctuations. In particular, it is independent of Doppler broadening. This fact governs the importance of non-Gaussian effects in the investigation of the characteristics of individual sources of radiation. In the traditional spectroscopic experiment, such characteristics either cannot be obtained in principle or, if they can, this can only be done at the expense of considerable effort. It is thus clear that the main advantage of IFS is that, when the field has non-Gaussian properties, this can be exploited to obtain information on the internal dynamics

¹⁾ This is in agreement with the central limit theorem, well known in statistical physics, according to which the field of N sources has Gaussian statistics if all the sources are independent and their number is large ($N \rightarrow \infty$).

of the system under investigation in pure form.

To illustrate the foregoing, consider the example of the monochromatic electromagnetic wave

$$E(\mathbf{r}, t) = E_0 e^{i\mathbf{k}\mathbf{r} - i\omega t} \quad (13)$$

scattered by a set of N independent particles. Scattering by the i th particle with position vector $\mathbf{r}_i(t)$ produces the spherical wave

$$E_i(t) = \alpha_i(t) \frac{1}{|\mathbf{R} - \mathbf{r}_i|} e^{i\mathbf{k}|\mathbf{R} - \mathbf{r}_i| + i\mathbf{q}_i \mathbf{r}_i - i\omega t}, \quad (14)$$

where

$$\mathbf{q}_i = \mathbf{k} - k \frac{\mathbf{R} - \mathbf{r}_i}{|\mathbf{R} - \mathbf{r}_i|},$$

\mathbf{R} is the radius-vector of the point of observation, and α_i is the scattering amplitude, which is proportional to the polarizability of the particle.

If we suppose that the observations are performed on the far field, so that the dimensions of the source can be neglected in comparison with the distance between the source and the point of observation, and if we further suppose that all the scattering particles are identical, we find, after substitution of (14) in (12), that

$$\langle I(0)I(\tau) \rangle = \frac{N}{R^4} \langle |\alpha(0)|^2 |\alpha(\tau)|^2 \rangle + \frac{N(N-1)}{R^4} \langle |\alpha(0)\alpha^*(\tau)|^2 \rangle \langle e^{-i\mathbf{q}(\mathbf{r}(\tau) - \mathbf{r}(0))} \rangle. \quad (15)$$

As can be seen, the Gaussian component, which is proportional to N^2 , is determined by the translational motion $\mathbf{r}(\tau) - \mathbf{r}(0)$. This can completely mask the dynamics of the individual particle, which is determined by the average $\langle \alpha(0)\alpha^*(\tau) \rangle$. At the same time, the non-Gaussian component, which is proportional to N , is independent of the translational motion throughout space, and is determined exclusively by the dynamics of the individual particle.

It was assumed in the derivation (15) that the scattering particles were entirely independent of one another. There is a variety of other situations in which the particle motions are correlated. For example, this occurs in turbulent flows, but we shall not consider such problems here. Collective motions are discussed in detail in a variety of papers and in a number of monographs.^{8,9}

C. Conditions necessary for the observation of non-Gaussian effects

Qualitatively, the conditions for the separation of non-Gaussian field effects can be established on the basis of the following considerations. In the region in which the observations are performed, we have the interference pattern due to fields of all sources, and we wish to observe the fluctuations in this pattern in the course of time. We may distinguish two types of fluctuations: random shifts of the interference pattern connected with phase fluctuations in the field which, as we have seen, are responsible for the Gaussian field component when the sources are independent, and random changes in the intensity of the interference pattern which are related to amplitude fluctuations and are responsible for the non-Gaussian field noise. The problem is therefore to separate one type of fluctuation from the other. When the characteristic times for these fluctuations are

very different, this presents no problem. When the characteristic time T_c (correlation time) for the phase fluctuations is greater than the characteristic time T_0 for the amplitude fluctuations, the problem is generally trivial: if we take the duration of observations to be much shorter than T_c , we can eliminate the effect of phase fluctuations on the output characteristics, i.e., the Gaussian field component. When $T_c \ll T_0$, the phase fluctuations are averaged during the time of observation, and the result of this is that the Gaussian component is suppressed by a factor of T_c/T_0 as compared with the non-Gaussian component.

All that we have said so far is best understood in spectroscopic language. The Gaussian and non-Gaussian components of the noise spectrum are readily separated if they have essentially different spectral widths.

The most difficult situation from the point of view of separating out non-Gaussian effects, is the intermediate case for which $T_c \sim T_0$. This often occurs, for example, when light is scattered by macroparticles in solution. There are two ways of separating amplitude fluctuations phase fluctuations in this case. Firstly, we can increase the solid angle Ω within which the radiation is collected, so that, when $\Omega \gg \Omega_c$ (Ω_c is the coherence solid angle which determines the region of phase correlations on the photocathode), the phase fluctuations will average out and hence the Gaussian component will be suppressed by a factor of Ω_c/Ω . Secondly, the measurements can be performed with two photodetectors, separated by a distance greater than the phase correlation length. Both methods rely on the fact that the phase fluctuations will average out over large distances. Another approach relies on the fact that the time T_c can sometimes be varied artificially and made very different from the amplitude-fluctuations time T_0 . One such method will be discussed in Section 4. It relies on the setting up of a transverse velocity gradient in the specimen with the result that the Gaussian component is again substantially reduced.

The above qualitative discussion shows that the most favorable conditions for separating the non-Gaussian component from the Gaussian background arise when the space-time field coherence is reduced as much as possible. It must, however, be remembered that the non-Gaussian component must be separated not only from the Gaussian background but also from the shot noise of the photodetector, and these two backgrounds sometimes dominate the situation.

To estimate the relative contribution of all three noise components, we turn to the formulas given by (7)–(9) for the photocurrent spectrum. Substituting (11) into these expressions, and recalling that all the sources are statistically independent, we obtain the following approximate formulas. The ratio of the non-Gaussian to the shot components is

$$\Delta_1 \sim qe^2 \frac{I}{N_c} \Omega T_0, \quad (16)$$

and the ratio of the non-Gaussian to the Gaussian component is

$$\Delta_2 \sim e^2 \frac{1}{N} \frac{\Omega}{\Omega_c} \frac{T_0}{T_c}; \quad (17)$$

where $\varepsilon = \Delta I/I$ is the intensity modulation depth of the radiation of an individual source, I is the average intensity of the radiation produced by an individual source per unit solid angle, and ΔI is the intensity fluctuation amplitude of an individual source.

The formulas given by (16) and (17) enable us to conclude that, in practice, one need not necessarily have to have a small number of particles participating in the scattering process in order to detect non-Gaussian effects. It is possible to choose the geometry of the experiment and to design the specimen so that the ratios Δ_1 and Δ_2 become sufficiently large even for $N \gg 1$.

We have throughout spoken of the separation of the non-Gaussian effect from the background of shot and Gaussian noise by comparing the respective averages. We must now note that the average level of shot and Gaussian noise cannot itself be a disturbing factor because it can always be simply subtracted from the resultant signal in one way or another. However, if the measurements are performed over a finite interval of time, we obtain only the statistical realization of the average during the time of observation, rather than the average value in the sense of mathematical expectation. These realizations fluctuate around their average value, producing "noise in noise," and this restricts the possibilities of the experiment. The uncertainties introduced in this way depend on the average noise (shot or Gaussian), the time interval Δt during which the measurements are performed, and the detection frequency band $\Delta\omega$. For example, when the background shot noise is taken into account, the signal-to-noise ratio can be written in the form¹⁰

$$\frac{S}{N} \sim \Delta_1 \sqrt{\Delta\omega \cdot \Delta t}. \quad (18)$$

3. NOISE IN ATOMIC SPECTROSCOPY

Studies of quantum beats in luminescence¹¹ served as the original stimulus to investigations of the efficacy of IFS in atomic spectroscopy. These studies clearly showed that atomic emission kinetics contains not only information on the lifetime of the radiating state but also on its structure that cannot be resolved in classical spectroscopy because of the dominating Doppler broadening of the spectral lines. However, kinetic measurements rely on the complex technique of selective pulsed excitation of atoms, so that searches for other solutions became more attractive. It seemed quite obvious that the kinetics of the decay of excited atoms should be manifest in the noise spectrum of luminescence emitted by steadily excited ensembles of atoms. This section is devoted to this and certain other possibilities of using IFS in atomic spectroscopy.

A. Searches for non-Gaussian properties in the spontaneous emission of a gaseous medium

1. Spontaneous emission noise under constant excitation of the medium.

In all cases in which it is required to produce a model of the photon—the product of the elementary atomic

emission event—it is usually presented as a damped train of classical electromagnetic waves. Until quite recently, this phenomenology was regarded as completely adequate and provided a means of avoiding precise calculations in quantum electrodynamics. This was used as the starting point in Ref. 12, in which an attempt was made to analyze the spectrum of intensity fluctuations (noise spectrum) in the spontaneous emission of a gaseous medium. The radiation field was represented by the sum of fields due to individual atoms:

$$\begin{aligned} E(t) &= \sum_j E_j(t) = \sum_j a_j \theta(t-t_j) e^{i\omega_j(t-t_j) - (1/2)\gamma(t-t_j)}, \\ \theta(t) &= \begin{cases} 0, & t < 0, \\ 1, & t > 0. \end{cases} \end{aligned} \quad (19)$$

The radiation intensity $I = |E|^2$ then consists of two terms, one of which is equal to the sum of intensities $I_i = |E_i|^2$ due to the individual atoms:

$$\sum_i I_i(t) = \sum_i |a_i|^2 \theta(t-t_i) e^{-\gamma(t-t_i)}, \quad (20)$$

and the other includes the interference between the emissions due to different atoms:

$$\sum_{i,j} E_i(t) E_j^*(t) = \sum_{i,j} a_i a_j^* \theta(t-t_i) \theta(t-t_j) e^{i(\omega_i - \omega_j)t - \gamma(t-t_i) - (1/2)\gamma t + i\phi_{ij}}. \quad (21)$$

The first term in (19) contributes to the intensity fluctuation spectrum in the form of a Lorentzian at zero frequency, whose width is equal to the width γ of the radiative state of the atom. The interference term (21) appears as a result of beats between all the components of the Doppler profile. This contribution is also centered on zero frequency and its width is equal to twice the Doppler width (under the conditions of dominant Doppler broadening).

The presence of a broad spectrum of "Doppler" beats in addition to shot noise had already been noted in Ref. 1. The novelty was the prediction of a narrow Lorentz peak with an *a priori* non-Gaussian origin and uniquely related to the kinetics of the elementary emission process. In contrast to the Gaussian component, the density of excess noise with the Lorentz spectrum was expected to be very high and could be comparable with the spectral density of shot noise. The experimental verification of this prediction was important both from the fundamental point of view and because it opened up a new way toward the determination of atomic constants.

The experiments reported in Ref. 12 (see also Ref. 13) clearly showed that the Lorentz feature at zero frequency was absent from the noise spectrum of spontaneous emission. This demonstrated that the traditional phenomenological description of the photon was invalid as part of the explanation of this experiment.

Let us explain this result in a qualitative manner. The description of the photon as a damped train of classical electromagnetic waves, i. e., a physically continuous process, implies the possibility of any degree of detailed analysis, and this is in conflict with the discrete nature of light. In fact, the photon is a discrete portion of energy that is wholly materialized in a discrete event such as the emission of a photoelectron.

The physical processes that occur in the course of photodetection are such that the photoelectron does not in itself carry any information on the kinetics of the wave packet representing the photon. Photoelectric emission is described on the time axis by a current delta-function with a white and, hence, uninformative spectrum. This is the situation in the case of a single emission event. We now recall that, in practice, we have to deal with emission by a *set* of atoms. Let us suppose, to begin with, that the excitation intensity is so low that the wave trains due to the individual atoms do not overlap in time. Figure 1a shows the envelopes of two trains of electromagnetic radiation corresponding to the excitation of two atoms at times t_i and t_k , respectively. When these trains interact with the photocathode, the most favorable situation is that of the emission of two photoelectrons at times t'_i and t'_k . These instants of time are statistically correlated with the instants of excitation t_i and t_k but, since the latter are assumed to be independent, there is no correlation between t'_i and t'_k , i. e., again, all information about the emission kinetics has been lost. Finally, let us suppose that the wave trains emitted by different atoms *do* overlap on the time axis (Fig. 1b). We then have the possibility of two photoelectrons being emitted in the overlap region as a result of the interaction between the resultant field and the photocathode. The creation of the first of these photoelectrons defines the origin of time, which enables us to establish the correlation with the appearance of the second electron, and this correlation should reflect the dynamics of the field generating these photoelectrons, including the exponential decay of the envelope. This is the case in principle but, in practice, this information cannot be extracted. In fact, in the overlap region, the photoemission probability exhibits beats in time with frequency $\omega_i + \omega_k$ where ω_i and ω_k are the random Doppler frequencies of the emission of the two atoms under consideration.²⁾ The exponential decay that reflects the required kinetics of the elementary emission process simply modulates these beats. This leads to the appearance of a wide spectrum that is practically indistinguishable from the Doppler spectrum, instead of the required narrow Lorentzian peak.

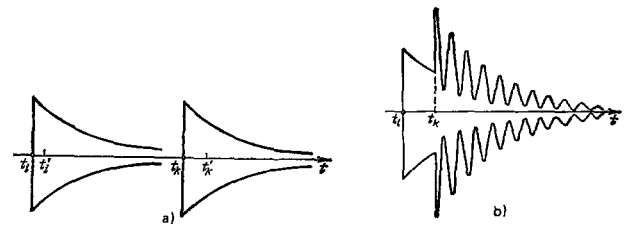


FIG. 1. Illustration of the absence of traces of spontaneous emission kinetics in the intensity fluctuation spectrum.

²⁾We emphasize that, in the overlap region, the photons that are recorded cannot be assigned to any particular atoms: they are the photons of the resultant field and do not have a definite energy. Were this not so, we would not be able to observe the beats since, for example, after the detection of the first photon of frequency ω_1 from the first atom, we would be left with the single-frequency field of the second atom, and the second photon would be uncorrelated with the first.

After it was demonstrated that classical photon phenomenology was inadequate as part of the explanation of experimental results, as attempt was made in Ref. 12 to perform the relevant calculations on the basis of a different and purely corpuscular phenomenology. It was assumed that, after excitation, the atom instantaneously emitted a particle—the photon—with a delay distributed in accordance with an exponential law. This led to the correct conclusion, namely, that the Lorentzian peak was not present in the noise but, of course, there was no way of describing the interference Gaussian noise which was of purely wave origin.

A unified description of all the noise components can only be developed on the basis of quantum electrodynamics.⁶ For atoms excited with Poisson statistics, the photocurrent noise spectrum has the form

$$i_{\omega}^2 = qF|u(\omega)|^2 \left(1 + \frac{1}{2} q\delta e^{-\omega^2/4\omega_0^2}\right), \quad (22)$$

where $u(\omega)$ is the frequency characteristic of the electronics, q is the quantum yield of the photodetector, δ is the degeneracy parameter which is numerically equal to the number of photons traversing the coherence area during the coherence time, F is the photon flux at the receiver, and ω_0 is the Doppler width.

The first term in (22) represents shot noise and the second the excess Gaussian noise. For spontaneous emission in the visible range, the degeneracy parameter δ is small ($\leq 10^{-3}$), so that the Gaussian noise spectrum consists of a very weak peak on the shot-noise background.

2. Coherent amplification of spontaneous emission.

An experimental situation in which the dynamics of atomic emission could not be described exclusively in terms of classical phenomenology had not been previously encountered in physics. This phenomenology is adequate for the description of regular emission processes such as quantum beats, superradiation, echoes, and so on. However, it turns out that the classical model can be invalid in the case of fluctuation processes. The above example of spontaneous emission by a gaseous medium serves as an illustration of this fact. Another illustration is afforded by attempts to modify the above experiment so that a study could be made of the noise spectrum of the spontaneous emission of a gaseous medium. The point is that the absence of the Lorentzian peak representing the excess noise in spontaneous emission is not in itself an argument for rejecting the description of the atom in terms of a continuous process of emission of an internally correlated train of electromagnetic waves. Experiment¹² merely shows that, even if such correlation were present, it would disappear without trace during photodetection, i.e., during the replacement of the photon with the photoelectron. It was therefore natural to try to “avoid” photodetection in searches for correlation, and turn to the coherent amplification of light. There were reasons to expect that a highly amplified primary wave packet was capable of participating in a number of absorption events in the photocathode, thus leading to correlation between photoelectrons over time intervals of the order of the lifetime of the atom. An experiment of this

kind was reported in Refs. 14 and 15. These results were subsequently reviewed¹⁶ and it was shown that the peaks found in the noise spectrum were due to the transformation of radiation fluctuations in the amplifying resonance medium and not due to excess correlations in the primary radiation. The observed structure was therefore ascribed to the amplifying medium and not to the source of primary radiation, as expected. A theoretical analysis of the coherent amplification process¹⁷ then shows that the amplification does not lead to the appearance of correlations corresponding to the classical model of the emission event.

The purely corpuscular approach was also found to be unsatisfactory in this case because it led to the expectation that coherent amplification should result in the appearance of a large number of pairs of photons with zero time delay. The authors of Ref. 18 described an experiment in which a search was made for zero-time excess correlations in emission after coherent amplification. The statistics of photon coincidences did not confirm the conclusion that there was an increased number of photon pairs at zero time, and showed only a slight increase in the number of photoelectron correlations within the coherence time of the primary radiation. The observed correlations were consistent with an effect of the type described by Hanbury Brown and Twiss,² the size of which is proportional to the spectral intensity and, therefore, increases during the light amplification process. These results can also be explained on the basis of Ref. 17. Thus, experiments on the coherent amplification of photons have shown that neither the classical nor the corpuscular phenomenologies are capable of explaining the noise spectrum. The correct results can only be obtained within the framework of quantum electrodynamics.

In conclusion, we note Ref. 19, which was published after the appearance of Refs. 6, 12, and 13.

The authors of Ref. 19 calculated the noise spectrum of spontaneous emission and again used two methods, namely: classical phenomenology of the photon and quantum electrodynamics. In contrast to Refs. 6 and 12, they found that these two approaches led to the same result, namely, the predicted appearance of the Lorentz feature in the noise spectrum. The reason for this was that they erroneously assumed that the noise spectrum was determined by the correlator $\langle I(t) \rangle \langle I(t+\tau) \rangle$ (angle brackets indicate quantum-mechanical averages and the bar represents averaging over the instants of excitation of the emitting atoms), whereas the correct correlator should have been $\langle I(t)I(t+\tau) \rangle$. These two quantities are identical in classical electrodynamics because there is no quantum-mechanical averaging but, in quantum theory, the difference between them is important in many cases, e.g., in spontaneous emission. This point was discussed in Refs. 6 and 12.

3. Transformation of excess noise in the excitation of the medium into non-Gaussian spontaneous emission.

Following the experimental and theoretical work discussed above, it may now be considered that it has been

reliably shown that the noise spectrum of spontaneous emission by a gaseous medium whose atoms are excited with Poisson statistics, i.e., completely independently of one another and with equal probability at all times, does not contain any features connected with the kinetics of the emission process. The situation is different if the excitation statistics is not of the Poisson type. For example, this is completely obvious in the case of pulsed excitation (i.e., completely non-Poissonian) after which the intensity of simultaneous emission decays exponentially. The question of spontaneous emission noise in the case of arbitrary excitation statistics was first discussed in Ref. 12, where it was shown that the non-Gaussian nature of the noise became more prominent with increasing departure of excitation statistics from the Poisson case. These calculations¹² were performed on the basis of the corpuscular description of the photon, so that the Gaussian noise component (usually very small), which was of pure wave origin, could not be taken into account. The complete expression that takes into account both wave and corpuscular properties of radiation was obtained in Ref. 6 by the methods of quantum electrodynamics. It has the form

$$i_{\omega}^2 = qF|u(\omega)|^2 \left(1 + q \frac{\Omega}{4\pi} \frac{M_{\omega}^2}{M} \frac{\gamma^2}{\gamma^2 + \omega^2} + \frac{1}{2} q \delta e^{-\omega^2/4\omega_0^2} \right). \quad (23)$$

This formula differs from (22) by the presence of the second term which contains the product of the power spectrum of the process, which describes the kinetics of spontaneous decay (Lorentzian of width γ), and the power spectrum M_{ω}^2 of the excitation-intensity fluctuations with mean intensity M . The magnitude of this term is also found to depend on the quantum yield q of the receiver and the solid angle Ω within which the radiation is collected. In deriving this formula, the random distribution of the excitation events in the neighborhood of each instant of time t was approximated by a Poisson process of intensity M which, however, was a function of time (random or determined), i.e.,

$$M_{\omega}^2 = \int_{-\infty}^{\infty} \langle \overline{M}(t) \overline{M}(t+\tau) \rangle e^{i\omega\tau} d\tau. \quad (24)$$

In accordance with (23), fluctuations in the excitation intensity are transformed into excess spontaneous-emission noise that carries information about the atomic constant γ . When the intensity is independent of time in the sense indicated above, i.e., the fluctuations are purely Poissonian in character, we have $M_{\omega}^2 \sim \delta(\omega)$ and the spontaneous-decay kinetics is not reflected in the noise spectrum.

In general, spontaneous-emission kinetics may differ from the exponential form and may contain beats at the upper-level splitting frequencies in external and intra-atomic fields (Zeeman and Stark structures, fine and hyperfine splitting). It is clear that, if they are to be extracted from the overall noise, the excitation fluctuation spectrum M_{ω}^2 and the spectral characteristic of the receiver $u(\omega)$ must overlap the width of the structure under investigation.

Experiments designed to demonstrate the charac-

teristic filtration of excitation noise are described in Refs. 12 and 13. In one of these experiments, mercury atoms were excited by an electron beam whose density was modulated by a broad-band noise generator. In another, the population of an excited state of xenon atoms was modulated in the course of the saturation of the transition by the very noisy degenerate emission produced by a single-pass laser. In both cases, the spontaneous-emission noise spectrum was found to contain a Lorentzian peak of width determined by the excited-state lifetime. In the second case, this time turned out to be greater than the characteristic time due to the trapping of the resonance emission.

B. Noise spectroscopy of light used to probe an atomic medium

There is a number of routes along which traditional spectroscopy can proceed: one can investigate fluorescence spectra, absorption spectra, and scattering and refraction spectra. An analogous situation is encountered in IFS as well: in addition to the fluorescence noise that we considered in the last section, we can also investigate absorption noise, i.e., noise characterizing radiation transmitted by the absorbing medium under investigation, and the scattered and refracted light noise. Moreover, since IFS investigates interference between two fields, there are other possible combinations, for example, we can investigate beats in the light transmitted by a medium with that scattered by it. Out of this great variety of possibilities, two approaches have been exploited so far, namely, the absorption noise of a gaseous medium has been investigated²⁰ and magnetic resonances have been observed in the noise spectrum of radiation refracted by a gas.²¹ The two experiments are unified by the theory developed in Ref. 22.

The experiments reported in Refs. 20 and 21 were the logical outcome of the searches for the non-Gaussian component of spontaneous emission, described above. As already noted, the basic result of these experiments was that the photodetection process could not be sensitive to the internal dynamics of the atom (provided only there was no correlation between the excitation of the different atoms), since the exchange of a photon for a photoelectron was accompanied by an irreversible destruction of the field state, and the internal correlation of the elementary spontaneous emission event could not be detected in this type of experiment. The situation is altered if the state of the atom is probed by external radiation. As it drifts across the light beam, each individual atom may scatter the external radiation twice. If, after the first scattering event, the atom does not completely "forget" its original state then, depending on how good this "memory" is, the second scattered photon will be correlated with the first, and this correlation can, of course, be established in the course of the photodetection process. This can be used to investigate the internal dynamics of an individual atom and, hence, an ensemble of independent atoms.

1. Absorption fluctuations.

The above probing procedure is not simple to demonstrate for short-lived excited states of atoms. Actually there are two conditions that must then be satisfied, which is difficult to achieve in this case. Thus, firstly, we need highly intense probing radiation, so that the atom can be excited several times during the lifetime of the initial state. Secondly, to ensure that the structure corresponding to the initial level does not become "contaminated" with the structure of the second level (the probing field interacts with both levels involved in the transition with which it is in resonance), we must ensure that the latter level, which is of no interest to us here, should be even more short-lived. However, these conditions can readily be satisfied if the state under investigation is a bound or a metastable state. Fluctuations in absorption from the ground state of an atomic vapor due to fluctuations in the number of atoms in the probing beam were investigated in the first experiment.²⁰ The decay of the state corresponded to the diffusional escape of the atom from the light beam.

Fluctuations in the intensity of transmitted light are proportional to the intensity itself. The fluctuations being investigated must be observed against the shot-noise background, which is proportional to the square root of the intensity, so that it is expedient to increase the intensity of the probing light but without entering the region in which nonlinear effects begin to arise that would complicate the situation. Optical pumping effects must be avoided for the same reason. It was found that these conditions could be satisfied by using unpolarized radiation from a gas-discharge lamp for the probing beam. As far as the maximum intensity is concerned this lamp is far from the optimal solution, but it has the advantage that it produces a very low noise level whose intensity is not very different from the shot-noise intensity in the frequency band above 10–100 Hz.

In the experiment, a layer of potassium vapor whose optical thickness for resonance radiation was of the order of unity was mixed with argon (~10 Torr) and was exposed to a narrow beam of unpolarized light from a potassium lamp. The transmitted light was detected by a photodiode whose output current was subjected to spectral analysis at low frequencies. Special calibration measurements were performed to determine the possible contribution due to the departure of the natural noise of the lamp from the shot-noise level at low frequencies. In fact, these deviations turned out to be small.

A characteristic increase in the noise level at low frequencies was observed for light transmitted by the cell containing the K + Ar vapor mixture. The excess noise component was approximately equal to the shot-noise background, which was in accord with the estimates reported in Ref. 20 and the theoretical work described in Refs. 22 and 23. Figure 2 reproduces the experimental results given in Ref. 20. The points show the frequency dependence of excess noise spectral density above shot noise. The solid curve is the calculated Lorentz noise spectrum with correlation time 4.5×10^{-3} s

(there is a misprint in Ref. 20 at this point). This time is of the order of magnitude of the estimated diffusion time of a potassium atom in argon across the light beam.

In a control experiment in which the cell contained potassium vapor without the buffer gas, the transmitted light noise was indistinguishable from shot noise. This was indeed expected because, in this case, the width of the excess noise spectrum should have been greater by a factor of the order of two (the time taken by the atom to cross the light beam was of the order of 10^{-5} s), and the spectral density of the excess noise should have been lower by a comparable factor.

The experiment demonstrated the possibility in principle of noise analysis of the state of a completely random ensemble of atoms. It led to proposals for further more penetrating experiments, which are described in the next section. The then newly discovered possibility of optical monitoring of the dynamics of the statistical motion of atoms appears to be of some practical interest in connection with studies of diffusion and self-diffusion of gases. Although the observed effect was small, it can be increased by some orders of magnitude by using a laser beam to explore the system. Moreover, these fluctuations in gas absorption must be borne in mind as a source of noise in devices using optical and radiooptical resonance discriminators, such as frequency standards using optically pumped alkali-metal atoms or laser-based optical standards of frequency. In the latter case, this noise may be particularly significant. Absorption noise can be reduced by a correct choice of the frequencies at which the useful signals are processed.

2. Probing in the transparency region: refraction fluctuations.

All the IFS work on atomic systems has been performed with the ambitious goal of extracting from radiation noise useful information on the internal dynamics of atomic states when they are completely disordered. The success achieved along this route in the last experiment has only been partly satisfactory because the information obtained on the translational motion of atoms is nowhere near the originally anticipated set of atomic characteristics (level structure, relaxation times of the components of the density matrix). This feeling of dissatisfaction gave rise to further efforts. It was suggested that IFS be used to de-

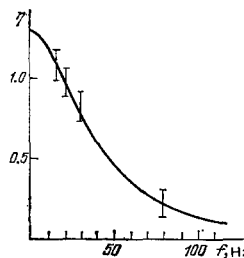


FIG. 2. Relative excess of noise spectral density of radiation used to probe an atomic vapor above the shot noise as a function of frequency.

tect the random Larmor precession of paramagnetic atoms in a magnetic field.

Larmor precession of the magnetic moment is the classical description of the coherence of the magnetic sublevels of a paramagnetic particle. For an ensemble of particles in thermal equilibrium, all forms of coherence should vanish on average, but this is only the case to within any fluctuations present in the system. Let us explain this by considering the example of the transverse component of the magnetization of a paramagnetic material placed in a magnetic field. Competition between the thermal energy of a particle and its energy of interaction with the field results in the establishment of a time-independent equilibrium magnetization along the external field. By virtue of symmetry, all orientations of the transverse component of the magnetic moment are equally probable, so that the average transverse magnetization is zero. However, the canceling of the individual microscopic magnetic moments is not perfect and, at each instant of time, the system should exhibit a randomly oriented transverse magnetization. Of course, this magnetization must be small to the extent to which the law of large numbers is satisfied, and must precess in the external field with the characteristic magnetic level splitting frequency.

In principle, this precession can be detected by observing the modulation of the absorption of circularly polarized light directed at right-angles to the magnetic field. This method of detecting the regular precession of spins is well known in the optical orientation technique as it is applied to atoms,²⁴ for which it is used to record magnetic resonance.

However, in contrast to population fluctuations, the possibility of noise detection of random precession is not so obvious. The perturbing role of the act of measurement is the key question. The presence of the atom in the beam is established by interaction with light, but this cannot remove it from the beam (the recoil momentum is small and can be neglected), so that its spatial state cannot be destroyed. On the other hand, when proton scattering is used to detect the orientation of the atom in space, this *can* alter the orientation, i. e., destroy the state; at any rate, the atom loses some of its "memory" of the initial state. Qualitative considerations of this kind drew the attention of the authors of Ref. 21 to the possibility of studying the ensemble within the transparency region, i. e., by examining refraction noise. Since refraction is not accompanied by real transitions in atoms, it may be supposed that this type of measuring procedure will be nondestructive.

Of course, the foregoing discussion cannot pretend to be complete and the ideas that it suggests must be verified by rigorous quantum theory. This verification was performed in Ref. 22, which confirmed estimates based on the semiclassical model of an ensemble of precessing magnetic moments.²¹ The theory introduced a measure of clarity also into the possibility of using light absorption as a way of detecting precession. We shall return to this question below but, for the moment, let us turn to the description of the ex-

periment.²¹

The medium under investigation was sodium vapor in a weak magnetic field of the order of 2 Oe. The vapor was illuminated at right-angles to the magnetic field by a linearly polarized laser beam tuned to a frequency lying in the neighborhood of the sodium resonance lines (Fig. 3). The precessing magnetization was recorded by observing the paramagnetic Faraday effect, i. e., the rotation of the plane of polarization of light transmitted by the magnetized medium. The rotation is proportional to the component of magnetization along the direction of the light beam. Precession of magnetization should lead to a periodic variation in the position of the plane of polarization at the Larmor frequency. This can be transformed by an analyzer into a periodic variation in the intensity of the transmitted light.

The photodetector output was amplified by a tuned amplifier operating at a fixed frequency (1.3 MHz). The position of the required noise peak was determined by using a scanning magnetic field. The output of the tuned amplifier was detected, amplified at low frequencies, and again synchronously detected in phase with the modulation of the magnetic field.

Simple estimates²¹ have shown that the expected magnetic peak in the noise spectrum should be higher by a factor of two than the shot-noise level.

Figure 4 shows examples of recorded signals. A clear resonant increase in noise density at 1.3 MHz was observed near the calculated magnetic field of 1.85 Oe for sodium vapor at a density of 10^{12} – 10^{14} cm⁻³. The signal intensity was found to increase as the laser frequency approached the sodium absorption lines. The signal was also reliably observed in the region in which the vapor was definitely transparent, i. e., when the laser line was placed between the resonance lines of sodium, at a distance of 3 Å from the absorption lines. In the transparency region, the signal was found to be a linear function of the laser-beam intensity. In all cases, the width of the resonance (of the order of 20 kHz) was determined by the bandwidth of the tuned amplifier. This was in agreement with the estimated phase relaxation time of sodium, which took into account transit times and collisions between the atoms.

This experiment demonstrates an essentially new approach to magnetic resonance studies. What is being

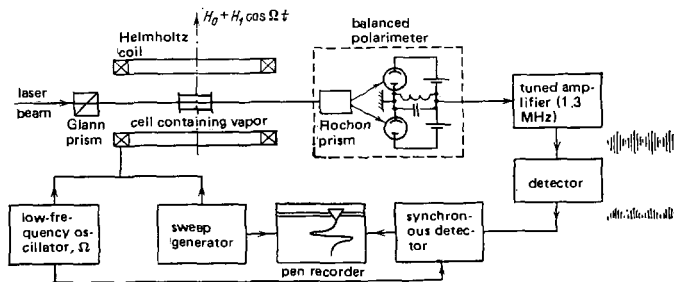


FIG. 3. Experimental arrangement used to observe magnetic resonance in the noise spectrum of Faraday rotation.

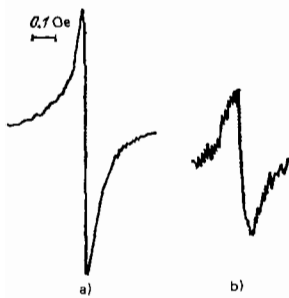


FIG. 4. Signals showing magnetic resonance in Faraday notation noise. The probing radiation was tuned to the edge of the D_1 absorption line of sodium (a) and to an intermediate point between the D_1 and D_2 absorption lines (b).

observed in this experiment is, in fact, magnetic resonance in sodium atoms, but under very unusual conditions: firstly, high-frequency fields are not used to induce resonance (the resonance may be referred to as spontaneous) and, secondly there is no need for the magnetic polarization of the medium, which is essential for the formation of the signal in conventional magnetic resonance. This point immediately leads us to a range of problems for which the new technique turns out to be preferable to traditional ESR methods. The intensity of the ESR signal is proportional to the product $N_\gamma H/kT$ where N is the total number of paramagnetic centers in the spectrometer resonator, γH is the splitting energy of the magnetic sublevels in the field H , and kT is the thermal energy (as usual, it is assumed that $\gamma H \ll kT$). The ESR signal tends to zero with increasing temperature and decreasing field strength. The signal associated with the noise resonance is proportional to \sqrt{N} , and for $\gamma H \ll kT$ is independent of both temperature and field strength. The noise method may therefore be indispensable in high temperature studies of paramagnetic relaxation, and also when the total number N of particles is anomalously low.

We note, by the way, that apart from fluctuations in transverse magnetization, the method can also be used (by directing the beam of light along the magnetic field) to investigate the fluctuation spectrum of longitudinal magnetization. It would appear that this may turn out to be promising in the study of longitudinal relaxation times as functions of external factors in condensed media.

3. Characteristic structure of the noise spectrum and its relation to kinetic processes in the medium.

The experiment described in Ref. 21 employed a special setup based on qualitative ideas about the nature of the effect. Theory²² not only confirms these qualitative ideas but, in addition, provides us with a means of examining the effect in the case of arbitrary polarization, arbitrary direction of the probing beam, and arbitrary type of analyzer.

The theoretical formula describing the information-bearing non-Gaussian part of the photocurrent spectrum when the probing beam is used within the trans-

parency region of the medium has the following form:

$$i_{\omega}^2 \sim \sum_{\kappa=0,1,2} \sum_{q=-\kappa}^{\kappa} I_0 \left(\frac{\varphi}{\sqrt{N}} \right)^2 \left\{ \begin{matrix} \kappa & \kappa & \kappa \\ j & j & j_1 \end{matrix} \right\}^2 Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}_H) g_{\kappa q}(\omega); \quad (25)$$

where I_0 is the total intensity of the probing radiation at the photodetector, ϕ is the total increase in the phase angle of the radiation in a medium of thickness l , N is the number of ground state atoms within the volume of the probing beam, $\left\{ \dots \right\}$ is the 6j-symbol, and j and j_1 are the angular momenta in the ground and excited states of the atom, respectively.

The dependence of the signal on the polarization of the incident and scattered radiation, and also on the direction of the constant magnetic field, is described by

$$Z^{\kappa q}(\mathbf{e}_0, \mathbf{e}_H) = |(\mathbf{e}_0, \mathbf{e}_H)^* \sum_{\mu, \mu'} C_{1\mu, 1\mu'}^{\kappa q} \mathbf{e}_{0\mu} \mathbf{e}_{H\mu'} - (\mathbf{e}_0 \neq \mathbf{e}_H)|^2 \quad (26)$$

where $C_{1\mu, 1\mu'}^{\kappa q}$ are the Clebsch-Gordan coefficients and \mathbf{e}_0 and \mathbf{e}_H are the polarization unit vectors of the incident and transmitted radiation whose components $e_{0\mu}$ and $e_{H\mu'}$ are specified in the coordinate frame whose z -axis lies along the magnetic field.

Each component κ provides its own contribution to the noise spectrum. The form of this contribution is given by

$$g_{\kappa q}(\omega) = \int_{-\infty}^{\infty} \frac{1}{\sigma(\tau) + S} \cos \Omega \tau e^{-\gamma_{\kappa} |\tau| + i\omega \tau} d\tau, \quad (27)$$

where Ω is the magnetic splitting frequency of the ground state, γ_{κ} are the damping constants of the ground state (determined by collisions with the buffer-gas atoms), S is the cross-sectional area of the probing beam $\sigma = (\mu\tau)^2$ when the mean free path is large in comparison with the transverse size of the light beam (μ is the root mean square thermal velocity), and $\sigma = a|\tau|$, when the mean free path is small in comparison with the transverse size of the beam (a is the diffusion coefficient).

As can be seen, the form of the spectral components is relatively complicated and, as noted above, is determined by all the possible spin relaxation processes. By associating each of the $\kappa=0, 1, 2$ with its own fluctuation process in the medium, we see that fluctuations in alignment ($\kappa=2$) may provide a contribution to the zero frequency and to the frequencies Ω and 2Ω , orientation fluctuations ($\kappa=1$) may contribute to the zero frequency and Ω , and population fluctuations ($\kappa=0$) provide no contribution since $Z^{00}=0$. The last effect can be understood from general physical considerations of this particular type of probing in the transparency region.

The contribution of the different components $g_{\kappa q}(\omega)$ depends on the particular experimental conditions. For example, in the experiment described in Ref. 21, in which linear polarization was used in both observation and illumination, whilst the magnetic field was pointed in the transverse direction, there was only one nonzero coefficient, namely, $Z^{11} = (1/4) \sin^2 \phi$, where ϕ is the angle between the polarizations of the incident and scattered radiation. Thus, only fluctuations in orientation ρ_1^{\pm} provide a contribution to the noise spectrum or,

more precisely, that part of the orientation that is related to coherence between neighboring Zeeman sublevels. This is accompanied by resonance at frequency Ω , which is in full agreement both with qualitative discussions and with experimental results. When, on the other hand, the magnetic field is made to point in the longitudinal direction, the other coefficient $Z^{10} = (1/2)\sin^2 2\varphi$ becomes nonzero, and we must conclude that, in this case, a peak connected with fluctuations in the orientation ρ_0^1 , i. e., with fluctuations in the population of Zeeman sublevels, should appear at zero frequency. Other variants of the experiment are also possible. For example, one can record the intensity of the circularly polarized component of transmitted radiation when the medium is illuminated by plane-polarized light, and so on. The above experiments cover all possible modifications.

In addition to formulas for the current spectrum given by (25), which apply to probing within the transparency region of the medium, the authors of Ref. 22 have also given formulas for probing within the absorption region. These turn out to be very similar to (25) except that the phase increase ϕ must be replaced by the average absorption coefficient of the medium (evaluated over the spectrum of the probing beam). In addition, the negative sign in the expression for $Z^{\kappa\alpha}$ must be reversed. The quantity Z^{00} then assumes a nonzero value, i. e., fluctuations in the total level population begin to contribute to the noise spectrum. In the case of unpolarized light, the data obtained by probing in the absorption region are in agreement with phenomenological analysis²³ and with experimental results.²⁰

Light-beam probing in the transparency region was found to be preferable in the experiment described in Ref. 21 because it was expected that the atom would retain a "memory" of its original state even after the scattering process. From this point of view, absorption probing appeared to be completely different because the extent of the memory was then determined, in addition to all other factors, by the upper state of the atom that was actually occupying it (in contrast to transmission probing). For example, it seems obvious that, if the upper state is spherically symmetric with angular momentum $j=0$, the atom occupying this level retains no "memory" of the symmetry of the initial state. In actual fact, there is no fundamental difference between scattering processes in different resonant probing techniques (i. e., so long as the difference between the illumination frequency and the atomic transition frequency is not of the order of the illumination frequency). One can identify two simple scattering mechanisms when the probing field is not too strong. One of them involves excitation of the dipole moment of the atom in the intermediate state, followed by emission. In this case, the atom is not actually excited to the upper state (two quantum scattering). The other mechanism involves the excitation of the atom to the upper level, followed by spontaneous decay of this level (two-step scattering). When the medium is illuminated within the transparency region, the upper level is unpopulated for practically all intensities. The contribution of the two-step scattering process to the

overall picture is therefore negligible, and need not be taken into account. On the other hand, when light beam probing is performed within the absorption region, the upper level is quite readily populated, and the contribution of the two-step scattering process may be considerable. However, when the angle within which the radiation is collected is made as small as possible, this contribution again becomes small in comparison with the contribution due to the two-quantum process, since the scattering function in the latter process is wholly connected with the divergence of the probing beam, whereas the scattering function in the two-step process is identical with that involved in spontaneous emission from the upper level, and this function is much broader. Thus, theory²² has shown that the characteristic structure will be manifest in the noise spectra of transmitted radiation recorded in both of the above probing methods and, from this standpoint, there is no fundamental difference between them.

We now make one other remark about the role of light in the evolution of non-Gaussian noise. It was assumed in the qualitative discussion before these experiments were performed that light was simply a means whereby intrinsic fluctuations in the medium are detected (passive role of light). In fact, the state of the medium can be described in the language of irreducible tensor operators

$$\rho_q^\kappa(t) = \rho_q^{-\kappa} + \delta\rho_q^\kappa(t),$$

where $\delta\rho_q^\kappa$ describe intrinsic fluctuations in level population ($\kappa=0$), orientation ($\kappa=1$), and alignment ($\kappa=2$), which arise because the ensemble of atoms is statistical in character. One or other variant of light-beam probing can be used to detect the corresponding type of fluctuation in the medium.

On the other hand, quantum-mechanical calculations show that light plays an active role. In particular, the state occupied by the atom between two successive scattering events is uniquely related to the initial state, and is different from it. For example, one can imagine a situation where these two approaches lead to diametrically opposite results. When only one of the magnetic sublevels is populated (for example, as a result of the "freezing" of the system, when $kT \ll \hbar\Omega$), fluctuations in the density matrix must be assumed to be absent, so that, in accordance with the passive light concept, one could expect that no characteristic fluctuations would be present. On the other hand, quantum-mechanical calculations (i. e., those based on the active light concept) predict the presence of fluctuations even in this case. Moreover, their intensity for spin 1/2 is the same as when the two sublevel populations are equal. This is not the first time that this type of situation has arisen. For example, the dual approach is used to describe Mandel'shtam-Brillouin scattering. Thus, on the one hand, one can consider scattering of light by two independently existing sound waves; on the other hand, in quantum theory, it is assumed that the sound waves can be excited during the scattering process. As in our case, these two approaches lead to the same result in the high-temperature limit $kT \gg \hbar\Omega$.

In conclusion of this section, we note the possibility of another equivalent approach to the interpretation of magnetic resonance in circular-birefringence noise. This noise may be interpreted as the result of interference between the original laser light and light that has undergone Raman scattering with spin flip in sodium vapor. This approach is discussed in detail in Ref. 55. From this point of view, the experiments described in Ref. 21 are closely related to studies of Brillouin scattering in toluene (3.56) and xenon (3.57). In both cases, the observed resonances are a consequence of Raman scattering. On the other hand, from the point of view of information theory, these are different experiments because one of them involves intra-atomic kinetics and the other the collective behavior of the medium. It is also relevant to note the similarity between Ref. 21 and direct optical studies of Raman scattering with spin flip in which "spontaneous" magnetic resonances was also observed, essentially, under the unusual conditions indicated above. The difference between these two versions and the new approach to the study of magnetic resonance are effectively the differences between conventional spectroscopy and IFS.

4. Nonlinear processes involved in the evolution of characteristic noise in the probing radiation.

Thus, when unexcited vapor atoms are probed with light, the noise spectrum of the transmitted radiation is found to acquire the characteristic non-Gaussian structure that originates from fluctuations in the parameters of the medium and is totally independent of the probing light itself (at sufficiently high thermal energies kT). This process will be referred to as *linear*, in contradiction to the *nonlinear* process in which the incident light has some effect on the medium. The concept of nonlinearity is somewhat broader in IFS than in conventional spectroscopy. For example, highly degenerate probing radiation can have relatively low average intensity, and thus produce purely linear phenomena in traditional spectroscopy, but it can give rise to large fluctuations that induce fluctuations in the parameters of the medium which, in turn, influence the noise in the original radiation.

Three problems have now been analyzed in some detail theoretically and to some extent experimentally:

- (1) linear probing of an unexcited medium²⁰⁻²³ (already discussed above).
- (2) nonlinear probing of an excited medium by "white" electromagnetic noise^{16,28}.
- (3) nonlinear probing of an excited medium by a monochromatic electromagnetic wave³¹⁻³².

Although these problems are the simplest that one can formulate, they nevertheless reflect the most typical experimental situations and, in the final analysis, provide us with a good demonstration of the possibilities of IFS. Moreover, they can be used to exhibit three different mechanisms producing characteristic features in noise, which we shall now briefly describe. The probing of an unexcited medium (problem 1) can

always be made linear, i.e., independent of the properties of the original radiation. Nonlinearity can be introduced by increasing the average intensity of the original light, in which case the noise spectrum acquires corrections that are small in comparison with the linear corrections. A more interesting situation is that where probing radiation is highly degenerate and of low mean intensity but exhibits considerable fluctuation (problem 2). This produces induced fluctuations in the parameters of the medium, which may become much greater than the intrinsic thermal fluctuations, and thus completely determine the final result. The nature of the excitation of the medium is then important only in so far as the sensitivity of the medium to the external perturbation is concerned. The effect is at a minimum when the difference between the populations of levels participating in the given atomic transition is zero, since the medium is then transparent and its parameters are difficult to "stimulate".

A different type of nonlinearity arises when a monochromatic wave is used to probe an excited medium (problem 3). In this case, atoms occupying the lower level generate linear noise (in accordance with problem 1) but, at the same time, the noise produced by atoms in the upper level cannot be described in the same order of perturbation theory in terms of linear process. One must then invoke combinational coupling in the medium between waves of different frequency³⁰ when a strong electromagnetic wave is present. The size of the nonlinear effect is of the same order of smallness as the linear effect when the two levels have roughly the same population. This may occur, for example, for two excited levels.

We note once again that, in both problems involving nonlinear probing, we have to deal not with corrections to linear effects but with effects of the same order, or even stronger.

Let us examine all these phenomena on the basis of a simple formal scheme that will enable us to achieve a better understanding of the correlations from which the useful signal originates in different cases. Let us suppose that the instantaneous intensity $I(t)$ of light transmitted by the medium can be written in the form

$$I(t) = I_0(t) + \delta I(t); \quad (28)$$

where I_0 is the original intensity and δI is an addition due to the medium. We now write the mean of the intensity product $G = \langle I(t)I(t') \rangle$ in the form

$$\left. \begin{aligned} G &= G_0 + G_1 + G_2, \\ G_0 &= \langle I_0(t)I_0(t') \rangle, \\ G_1 &= \langle I_0(t)\delta I(t') \rangle + \langle I_0(t')\delta I(t) \rangle, \\ G_2 &= \langle \delta I(t)\delta I(t') \rangle, \end{aligned} \right\} \quad (29)$$

which, as we already know, determines the excess noise (Gaussian and non-Gaussian). The quantity G_0 determines the noise in the original light, and G_1 and G_2 are in some way related to the influence of the medium.

When fluctuations in the original light are small, they cannot induce fluctuations in the medium, so that I_0 and δI must be regarded statistically as indepen-

dent and the quantity $\sim G_1$ becomes uninteresting, since it can only contribute to the constant component of the noise when the averages on which it depends are factorized out. The noise effect is determined by the mean $\langle \delta I \cdot \delta I \rangle$. If we now recall the expressions

$$E(t) = E_0(t) + \delta E(t), \quad I = |E|^2, \quad I_0 = |E_0|^2, \quad (30)$$

and (28), we find that the leading components of $\langle \delta I \cdot \delta I \rangle$ are $\langle E_0^* E_0 \delta E^* \cdot \delta E \rangle$ and $\langle E_0^* E_0^* \delta E \cdot \delta E \rangle$. Moreover, for atoms occupying the lower level, they are expressed in terms of thermal fluctuations in parameters of the medium, i. e., they determine linear phenomena for which light acts as the probe used to examine these fluctuations, but does not perturb the medium. On the other hand, for atoms occupying the upper level, the average $\langle E_0^* E_0^* \delta E \cdot \delta E \rangle$ can be treated exclusively from the point of view of the onset of nonlinear combinational coupling, i. e., as being due to a change in the kinetics of the medium under the influence of the external field.

When fluctuations in the original field are large, the quantities I_0 and δI cease to be statistically independent because induced δI are found to appear. The main contribution to the characteristic noise is then provided by the quantity G_1 . Thus, the useful signal is then formed as a result of correlation between secondary emission and the original radiation.

We note that the characteristic structure of noise spectra in nonlinear problems is not always connected with non-Gaussian statistics. For example, when monochromatic radiation is used as the probe, this type of structure appears even in the traditional spectrum I_ω ²⁵⁻²⁷ i. e., in the Gaussian component.

The papers cited above are largely concerned with the probing of gaseous media with external fields that are not too strong. The saturation properties of probing fields may introduce new elements into the mechanisms responsible for the evolution of both Gaussian and non-Gaussian noise components. We shall not consider this question here and merely call attention to some of the papers³³⁻³⁷ devoted to this topic.

* * *

The discussion given in this section demonstrates that IFS is a fruitful source of information on the dynamics of the states of quantum-mechanical objects. Since, however, this information can, as a rule, be obtained by other methods as well, it will be useful to note the conditions under which the new approach is more advantageous. The competing spectroscopic techniques (magnetic resonance, interference of states, nonlinear laser spectroscopy²⁹) are based on the registration of collective (coherent) responses of a system of particles to an external disturbance. The amplitude of these responses are proportional to the particle concentration N_0 . In IFS, the observed quantities are the fluctuations in the parameters of the system, the characteristic magnitudes of which are proportional to $\sqrt{N_0}$. For large values of N_0 , therefore, the noise signal usually has lower intensity than the response to a regular disturbance. Thus, whenever regular, ultrahigh-resolution spectroscopic methods can be employed, they are

to be preferred.³⁾

However, there are situations where regular methods cannot be employed because the necessary order cannot be introduced in the system, or it is undesirable to disturb the system. For example, the object under investigation may be inaccessible (astrophysical objects belong to this type). Another example is the application of IFS to the study of magnetic resonance, which was discussed above. When the magnetic field is reduced, or the temperature is increased, the regular ESR signal will become weak, whereas the noise signal will be independent of these variables and, under certain conditions, may become the dominant component. Moreover, noise spectroscopy can be used to investigate the system under conditions of complete disorder in the state under investigation, which may also be an important consideration in special cases.

4. EFFECTS ASSOCIATED WITH NON-GAUSSIAN STATISTICS IN THE SCATTERING OF LIGHT BY MACROPARTICLES IN SOLUTION

As noted in the Introduction, IFS has found particularly wide-spread application in studies of the motion of particles in liquids and gases. Such investigations are performed in the pursuit of different aims, e. g., in the examination of the velocity field of liquid or gas flows, measurement of the particle diffusion coefficients, determination of the active mobility of microorganisms, and so on. In all these cases, a measurement is made of the broadening and displacement of a laser line resulting from the scattering of laser radiation by moving particles, i. e., IFS is used as ultrahigh-resolution spectroscopy to obtain data on the spectrum of the radiation either by heterodyning or directly from the intensity fluctuation spectrum (and, as is usually the case, the recorded radiation has Gaussian statistics). In investigations concerned with microparticles in solution, IFS is, at present, the most accurate and convenient way of measuring the diffusion coefficient, so that the method can be employed to determine the dimensions of macromolecules, to detect conformational transitions, to investigate interactions between particles, and so on.

However, the spectrum of scattered radiation is not exclusively determined by the translational motion of the particles. Rotation of nonspherical particles, thermal fluctuations in their shape and state, and internal motions connected, for example, with the functioning of the organelles of microorganisms, all contribute to the characteristic amplitude modulation of the scattered field and, consequently, affect the spectrum of the scattered radiation. We shall combine these and other similar mechanisms responsible for fluctuations in the scattering power under the heading "internal particle dynamics."

³⁾ The dilemma that arises in this connection can be figuratively compared with that facing an archaeologist who has to decide between excavating an ancient midden or a whole town preserved in volcanic ash. The former must suffice when nothing better is available.

The internal dynamics of macromolecules and high-molecular complexes is of considerable interest, and there have been several attempts³⁸⁻⁴⁰ to investigate it by IFS. It is important to remember, however, that the contributions due to amplitude modulation connected with internal dynamics can only be distinguished from phase modulation connected with translational motion by their different angular distribution in the scattered spectrum. We shall see below that the correlation characteristics of amplitude and phase modulation in typical situations are very similar, so that quantitative data on the internal dynamics of these systems are very different to obtain by this method.

In the remainder of this section, we shall discuss a new approach to the study of the internal dynamics of macroparticles in solution, based on the exploitation of non-Gaussian effects. The point is that, in the case of non-Gaussian statistics, the amplitude and phase modulations are not equivalent. Thus, the non-Gaussian component of the intensity spectrum is not subject to diffusion broadening and is wholly determined by internal dynamics. Unfortunately, even in this case, we still have the problem of separating the non-Gaussian component from the Gaussian background with rather similar spectral characteristics. In the concluding section of the present review, we shall examine ways of overcoming this difficulty by methodologies based on differences between the nature of the two processes to be separated, and will give examples of a successful separation of the non-Gaussian component from the Gaussian background, including situations in which the spectral widths of the components are very similar.

A. Application of IFS to the study of the internal dynamics of macroparticles

In atomic spectroscopy, large Doppler broadening ensures that the spectral density of the Gaussian noise component is usually small in comparison with the shot component. The opposite situation is encountered when we consider macroparticles: the diffusion broadening of the spectrum scattered by relatively large, and hence slowly moving, particles is small, and the spectral density of the Gaussian component is usually greater than that of the shot background. Thus, we have the problem of separating the non-Gaussian component from the Gaussian background, and this dictates the methodology that has to be used in the study of macroparticle dynamics.

The complexity lies not only in the high spectral density of the Gaussian noise component, but also in the fact that the spectra of the Gaussian and non-Gaussian fluctuation components are frequently almost completely superimposed on one another and are difficult to separate.

We now turn to Eq. (15), which describes correlations in the intensity of light scattered by N independent particles. The Gaussian component consists of two characteristic factors. One of them is connected with the translational motion of the particles and, if we suppose that this motion constitutes diffusion (D is the coef-

ficient of translational diffusion), we may write

$$\langle e^{i\mathbf{q}(\mathbf{r}(\tau) - \mathbf{r}(0))} \rangle = e^{-Dq^2\tau}. \quad (31)$$

The second factor is determined by the internal dynamics of the particle, and is the correlation function for fluctuations in the formfactor $\alpha(\mathbf{q}, t)$, which (in the single-scattering approximation) takes the form

$$\alpha(\mathbf{q}, t) = \int \Delta\chi(\mathbf{r}, t) e^{i\mathbf{q}\cdot\mathbf{r}} d^3r, \quad (32)$$

where $\Delta\chi$ is the deviation of the polarizability from the mean and the integral is evaluated over the volume of the particle. Thus,

$$\langle i(0) i(\tau) \rangle_{\text{gauss}} \sim N^2 |\langle \alpha(0) \alpha^*(\tau) \rangle|^2 e^{-2Dq^2\tau}. \quad (33)$$

The non-Gaussian component, on the other hand, is described exclusively by formfactor fluctuations:

$$\langle i(0) i(\tau) \rangle_{\text{nongauss}} \sim N \langle |\alpha(0)|^2 |\alpha(\tau)|^2 \rangle. \quad (34)$$

When the characteristic size b of the particles under investigation is small ($qb \ll 1$), we can confine our attention to the first terms of the expansion of the formfactor in powers of qb , and verify that the intensity of the non-Gaussian component is proportional to $(qb)^4$. Thus, the exploration of internal dynamics is possible only for particles whose dimensions are not too small in comparison with the wavelength. On the other hand, the spectral width of the Gaussian component is determined by the reciprocal of the time taken by the particle to diffuse through a distance $1/q$, and the width of the non-Gaussian component is determined by the characteristic time for the change in the shape of the particle. It is clear from dimensional considerations that the latter time is equal to the time taken by the particle to diffuse through a distance of the order of its size. Thus, when $qb \sim 1$, the widths of the Gaussian and non-Gaussian components are of the same order. For example, when formfactor fluctuations are connected with the rotational diffusion of particles, the spectral width of the non-Gaussian component is proportional to the coefficient of rotational diffusion θ . For particles that can be regarded as spheres for hydrodynamic purposes, we have

$$D = \frac{kT}{8\pi\eta b}, \quad \theta = \frac{kT}{8\pi\eta b^3}, \quad (35)$$

where η is the viscosity of the solution and, when $qb \sim 1$, we have $Dq^2 \sim \theta$.

The precise shape of the Gaussian component of the noise spectrum is usually unknown. The point is that the real specimen usually consists of particles with different diffusion coefficients, both because extraneous impurities cannot be entirely eliminated and because of the natural polydispersity of the object under investigation. We thus have the situation where the non-Gaussian component cannot be separated by increasing the accuracy of the experiment, and some other steps have to be taken in order to ensure a sufficiently large ratio of non-Gaussian to Gaussian spectral densities. The Gaussian component can, of course, be simply subtracted once it has been measured in a preliminary experiment or by optical heterodyning, i.e., by directing a high-intensity monochromatic reference beam onto the photodetector, so that only the

second-order correlator for the scattered electromagnetic field is effectively measured or, in accordance with (17), by reducing the aperture within which the radiation is collected. However, this requires absolute calibration which is difficult to perform in practice with sufficient precision.

Finally, we note one other purely methodological feature encountered in the study of the dynamics of macroparticles in solution. If we use (35), we can readily show that, for particles whose sizes are in the range 10^2 – 10^4 Å, we have to measure spectral widths in the range 1 Hz–1 MHz. A considerable length of time is necessary to ensure sufficient precision in this range, especially at its low-frequency end. In practice, therefore, one is forced to use real-time analyzers.

B. Non-Gaussian effects for $N \sim 1$

The most obvious way of isolating non-Gaussian effects is to reduce the number of particles within the volume under investigation. In fact, until quite recently, it was considered that such effects were actually manifest only when $N \sim 1$. However, the main contribution to observed effects under such conditions is due to fluctuations in the number of particles in the scattering volume, which are connected exclusively with translational diffusion and carry no information on the internal dynamics of the particles under investigation. Let us consider this situation in greater detail.

There are two ways of reducing the number of particles in the scattering volume: one can either reduce the particle concentration without changing the scattering geometry, or one can reduce the scattering volume without changing the concentration. In the former case, the ratio of non-Gaussian to shot spectral densities remains unaltered whilst the Gaussian component is suppressed relative to the other two. However, the absolute magnitude of the non-Gaussian component falls in proportion to the concentration, so that it cannot be measured in the case of strong dilution because of parasitic scattering and dark noise in the recording equipment. In the second case, it is natural to reduce the size V of the scattering volume by focusing down the probing radiation. The intensity of light scattered by an individual particle is then proportional to $V^{-2/3}$, the number of particles is proportional to V , and the coherence solid angle is proportional to $V^{2/3}$. Thus, according to (16) and (17), the spectral density of Gaussian, non-Gaussian, and shot components is, respectively, constant, proportional to $N^{-1/3}$, and proportional $N^{1/3}$, i. e., focusing can be used to increase not only the non-Gaussian noise relative to the Gaussian and shot noise, but also to increase the absolute magnitude of the signal.

It would appear that maximum focusing of the probing radiation would ensure the best conditions for the investigation of internal dynamics with the aid of non-Gaussian effects. However, this is not the case. The point is that the recorded intensity scattered by an individual particle can vary not only as a consequence of its internal dynamics, but also as a result of its motion relative to the probing beam and the volume from which the light is collected. Since formfactor fluctua-

tions occur independently of the position of the particle, this phenomenon can be allowed for by multiplying the photocurrent correlation function (34) by the factor $e^{-|r|}/T_N$, where T_N is the time spent by the particle in the scattering volume, $T_N \sim L^2/D$, and L is the smallest characteristic linear size of the scattering volume. Thus, fluctuations in the number of particles lead to a broadening of the non-Gaussian component. In the case of maximum focusing, i. e., when $L \sim \lambda$, this broadening becomes comparable with the usual diffusion broadening, and any advantages of this particular method as compared with additional procedures for isolating the contribution due to the internal dynamics in the recorded spectrum are lost.

The broadening of the non-Gaussian component due to fluctuations in the number of particles in the scattering volume is a general feature. For example, when the non-Gaussian component of the polarization noise associated with scattered light is separated in atomic spectroscopy (Section 3), the magnetic resonances are broadened largely as a result of the drift of the atoms across the probing beam. It is clear that, when the time spent by the particle within the volume under investigation is small in comparison with the characteristic time for the process in which we are interested, the contribution of the latter will be small. This means that, as we have seen in the last section, the size of the scattering volume in studies of the internal dynamics of macroparticles must be much greater than the particle size. When $N \sim 1$, this condition can be met only for large and strongly scattering particles. Figure 5 shows measurements of the intensity correlation function for light scattered by a suspension of *E. coli*.⁴¹ Analysis of these data shows that, although the recorded noise is non-Gaussian, it is almost entirely determined by fluctuations in the number of particles in the scattering volume. According to the authors of Ref. 41, only the exponential decay over the initial segment of the correlation function is connected with orientational relaxation. Whatever the true interpretation of these data (one cannot exclude the possibility that this is the contribution of the Gaussian noise component with decay time Dq^2), it is worthy of note that the effect itself is small. It is therefore not surprising that non-Gaussian effects that arise for $N \sim 1$ cannot be used to obtain information on the internal dynamics of macromolecules and high-molecular complexes, i. e., particles that are

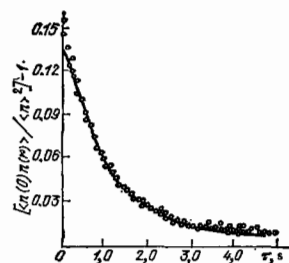


FIG. 5. Autocorrelation function for the intensity of light scattered by a suspension of *E. coli* AW 405. Solid line— theoretical, representing fluctuations in the number of particles within the scattering volume.⁴¹

much smaller than bacteria, and this approach has not been pursued much further.

C. Fluorescence IFS

The Gaussian and non-Gaussian components of the noise spectrum have very similar spectroscopic characteristics when the width of the spectrum of the recorded radiation is determined by diffusional motion in the sources.

Fluorescence IFS, in which one observes not the scattered light but the emission of fluorescing markers on macroparticles, is an example of a situation where there is no difficulty in separating the Gaussian from the non-Gaussian component. Two mechanisms are responsible for low-frequency fluctuations in the intensity of fluorescence emitted by an individual center. Firstly, the center drifts across the volume in which the excitation is produced (or the volume in which the observations are performed), and this leads to fluctuations in the number of observed centers. Secondly, there is the orientational motion of the center. The latter mechanism is important when the angular distribution of the emitted radiation and (or) of the excitation is an isotropic. Thus, by measuring the fluorescence noise spectrum, one obtains information both on the diffusional motion of objects containing a fluorescence center and on their internal dynamics. These mechanisms lead to intensity fluctuations with characteristic times that are large in comparison with the reciprocal width of the fluorescence spectrum. At the same time, the spectral width of the Gaussian component is so large that its spectral density is small in comparison with shot noise. The situation is analogous to IFS of the spontaneous emission of excited atoms (Section 3A). The difference is that the width of the spontaneous emission spectrum is determined by Doppler broadening, whereas the width of the fluorescence spectrum (band) is a characteristic of the fluorescing centers and is usually much greater than the Doppler broadening associated with the motion of these centers.

The important methodological advantage of fluorescence IFS as compared with scattered-light IFS is that it can be used for the selective observation of objects that are "labeled" by a fluorescing marker. In particular, by measuring the fluctuation spectrum of the number of such markers within a given volume, one can determine the diffusion coefficient for the labeled objects in complicated, multicomponent systems. Experiments of this kind have been successfully performed by a number of workers.^{42,43} The intensity of a fluorescing marker may also vary as a result of chemical reactions, so that fluorescence IFS can be used to investigate the rates of such reactions under equilibrium conditions.⁴⁴

An attempt has been reported in Ref. 45 to use fluorescence IFS to investigate internal macroparticle dynamics as well. However, in addition to its important advantage, namely, large spectral width, i. e., low degree of coherence of the recorded radiation, this method suffers from the basic disadvantage that the intensity due to an individual source is low.

The ease of separation of non-Gaussian effects from the Gaussian background has to be paid for by the complexity of separation of these effects from the shot-noise background. It would seem, at first sight, that reliable detection of fluctuations in the number of macroparticles in the above experiments is an indication that the intensity emitted by an individual center is sufficient. However, the important quantity in these experiments was not the intensity due to an individual marker, but the resultant intensity due to a macroparticle which, in practice, contains a large number of such markers. This type of amplification of non-Gaussian effects is absent in the case of orientational motion because of the random orientation of different markers in the object under investigation. The difficulty connected with insufficient intensity of an individual center has not been overcome and, so far as we know, it has not been possible to obtain information on the internal dynamics of macroparticles with the aid of fluorescence IFS. (We are not concerned here with experiments on the decay of fluorescence polarization after pulsed excitation, or the depolarization of fluorescence. Such experiments record not the fluctuating but the determined signal, so that they do not properly belong to the range of problems that we are discussing here.)

The Gaussian component is readily suppressed in fluorescence IFS because of the low degree of coherence of the radiation emitted by the markers. A similar effect can be achieved in the case of scattering if sufficiently nonmonochromatic radiation is used in the probing beam.⁴⁶ It is clear that, when the separation between the scattering particles is greater than the coherence length of the probing radiation, interference between fields scattered by different particles will vanish, and this will signal the absence of the Gaussian component. Here again, the main difficulty is to separate the non-Gaussian component from the shot-noise background, which means that a very bright, nonmonochromatic source of radiation is essential. We note that, when light with nondegenerate statistical properties is scattered, the recorded radiation is definitely non-Gaussian. In fact, since the statistics of the product of two Gaussian random quantities is non-Gaussian, the radiation scattered by a system with Gaussian statistical properties will have Gaussian statistics only for a determined probing radiation. For example, generally speaking, the statistics of singly-scattered light is non-Gaussian.⁴⁷

D. Artificial reduction in the spectral density of the Gaussian component

When the volume in which laser radiation is scattered contains a flow with velocity gradient W , the width of the scattered spectrum is determined not only by diffusion broadening but also by homogeneous Doppler broadening $\sim qWL$. The width of the Gaussian component can be substantially increased by ensuring that the velocity gradient is large enough. The spectral density of this component is then very low, and this immediately disposes of the problem of separation of the non-Gaussian component from its background. At the same time,

there is no difficulty with the shot background because the intensity due to an individual source remains high in this approach. The implementation of this idea appears to have led to the first observation of non-Gaussian effects associated with orientational motion of particles.⁵¹

Particles placed in a flow with a lateral velocity gradient acquire rotational motion. This phenomenon has a number of consequences, of which the best known is the dynamooptical Maxwell effect that consists of the appearance of optical anisotropy in a flow with a velocity gradient.

The dynamooptical effect is the basis of the method of flow birefringence, which is widely used to investigate the optical and hydrodynamic properties of macromolecules.⁴⁸ The orienting effect of the flow is also used to determine the particle shape from the angular distribution of the scattered radiation.⁴⁹

Non-Gaussian effects that arise in light scattering by particles in a flow with a lateral velocity gradient are, of course, connected not with the mean orientation but with the regular rotation of each individual particle, which modulates its formfactor. It can be shown¹⁵ that, for axially symmetric (from the hydrodynamic point of view) particles, and if we neglect rotational diffusion, the motion of the axis of the particle is strictly periodic, and the period T is independent of the initial orientation of the axis relative to the flow. It is given by

$$T = \frac{4\pi}{W\sqrt{1-a^2}}. \quad (36)$$

The anisotropy parameter a can be found for an ellipsoid of revolution for which it is given by $a = (1 - p^2)/(1 + p^2)$, where p is the ratio of the ellipsoid axes. Rotational diffusion and deviation of the particle shape from the axially symmetric shape will disturb the periodicity of the motion. When the coefficient of rotational diffusion is $\theta \ll T^{-1}$, and the departure from the axially symmetric shape is small, the deviation from periodicity is also small, and the non-Gaussian component of the scattered-intensity spectrum consists of a set of equidistant lines separated by $\Delta\omega = 2\pi/T$. Their width depends on θ and on the departure from the axial symmetry of the particles.

Experiments to demonstrate this phenomenon have been performed with *E. coli* bacteria, whose dimensions were sufficient to ensure a well-defined effect. A flow with a transverse velocity gradient was established in a gap of $d = 2$ mm between fixed and rotation coaxial cylinders of radii $\rho = 20$ mm and $\rho - d = 18$ mm, respectively. The moving cylinder was rotated at $f = 36$ rev/s, which produced a velocity gradient $W = 2\pi f\rho/d \sim 2 \cdot 10^3$ s⁻¹. The modulation of the formfactor due to the rotation of the particles was maximized by observing the scattered radiation in the plane perpendicular to the axes of the cylinders, i. e., in the plane of the flow. The diameter of the probing beam was 0.1 mm and the number of particles in the volume that was examined was $N \sim 10^4$.

Figure 6 shows the results of this experiment. A well-defined line can be clearly seen on the intensity

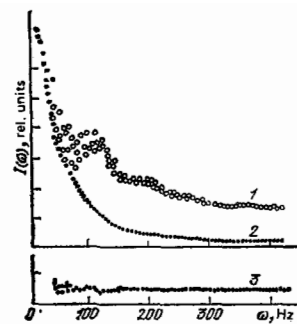


FIG. 6. Intensity fluctuation spectrum recorded for light scattered by particles in a flow with a lateral velocity gradient: 1—*E. coli* in a flow with a velocity gradient; 2—the same in the absence of flow; 3—spherical particles (latex) in a flow with a velocity gradient.

fluctuation spectrum of light scattered by bacteria in the flow with velocity gradient (curve 1). The splitting is independent of the angle of scattering and amounts to 120 Hz. According to (36), this corresponds to anisotropy parameter $a \approx 0.7$, or to an axial ratio of the equivalent ellipsoid of revolution $p \approx 2.3$, which is in excellent agreement with the known average dimensions of *E. coli* ($2 \times 0.8 \times 0.8 \mu\text{m}^3$). The width of the line is much greater than the expected broadening due to rotational diffusion. Since the shape of the *E. coli* is nearly axially symmetric, the observed width is probably connected with the presence of bacteria of different shape in the specimen, with p ranging from 2.0 to 2.5. This is consistent with visual observations. These data illustrate the possibilities of this type of experiment as a means of determining the elongation distribution function for particles in solution.

Curve 2 shows the photocurrent spectrum when the rotating cylinder is brought to rest. Here, one observes the usual Gaussian component, i. e., a Lorentz profile (90° scattering). As the cylinder is set in motion, homogeneous Doppler broadening produces a spreading of this profile over $dWq \sim 10^7$, so that non-Gaussian effects can be observed through the appearance of the detached line. The fact that this line is connected with the rotation of particles in the flow, i. e., with the modulation of the formfactor $\alpha(t)$, can be verified by performing a control experiment in which light is scattered by spherical particles of similar size (1.5 μm diameter latex particles), in which case, this modulation does not, of course, arise and characteristic lines of any kind do not appear. The results of this type of control experiment are shown by curve 3 in Fig. 6.

E. Cross-correlation method

We shall next discuss a method which now appears to be the most promising from the point of view of studies of the internal dynamics of macroparticles. It relies on the analysis of the correlation between photocurrents i_1 and i_2 produced by two different photodetectors rather than the autocorrelation of the photocurrent due to a single receiver as in all the above cases. It has been found that this leads to several

nontrivial advantages.

We have already noted that the Gaussian noise component is a consequence of interference between fields from different sources, which can occur only within the limits of the coherence solid angle Ω_c . It follows that, when the angle between the two directions drawn from the scattering volume to the two photodetectors is greater than the coherence angle, the Gaussian component should not appear. In typical optical experiments, the coherence angle is of the order of the ratio of the wavelength of the radiation to the linear dimensions of the volume in which the observations are performed, i. e., the suppression of the Gaussian component can be assured for practically any mutual disposition of the photodetectors. When two photodetectors are used, the shot noise will also disappear, which is readily understood on the basis of (6), and any other statistically independent noise will be absent from the two detection channels as well. It must be remembered that the precision of the experiment is not increased as a result of this (by precision, we mean the signal-to-noise ratio) because only the average levels of the Gaussian and shot components are zero in this type of detection. The statistical uncertainty in the measured $G^{b3}(\tau)$, on the other hand, is determined, as before, by all the intensity fluctuations. In fact, proceeding as in the derivation of (18), we can readily verify that the uncertainty in the measured cross-correlation functions and the autocorrelation function are practically equal. However, when the characteristic times of phase and amplitude correlations are of the same order, the very absence of the Gaussian component is very significant because its spectral shape is really unknown, which complicates the determination of the spectral characteristics and the non-Gaussian component.

It would appear that the possibilities of the cross-correlation method as a way of studying the internal dynamics of macroparticles in solution were first demonstrated in Ref. 53, although the use of two photodetectors had been suggested in Ref. 52. It is also shown in Ref. 53 that the cross-correlation method has a further methodological advantage in studies of rotational diffusion, namely, for fixed positions of photodetectors, the orientational fluctuations provide an anticorrelation contribution to $G^{b3}(\tau) = \langle i_1(0) i_2(\tau) \rangle$. It can be shown that, for single scattering by small ($qb \ll 1$) axially symmetric particles, $G^{b3} \sim (q_1 q_2)^2 - (1/3) q_1^2 q_2^2$, where q_1 and q_2 are the scattering vectors corresponding to the two photodetectors, respectively. Maximum anticorrelation occurs for $q_1 \perp q_2$. This condition is, of course, violated for particles whose dimensions are comparable with the wavelength.

The anticorrelation effect was used in Ref. 53 to detect the presence of orientational diffusion. Light was scattered by a solution of the tobacco mosaic virus (rods 3000 Å long and 50 Å thick). The experimental conditions corresponded to $N \sim 1$ under maximum focusing. Moreover, as in Ref. 41, the observations were largely confined to fluctuations in the number of particles in the scattering volume. Despite the optimum

disposition of the photodetectors (opposite to one another, at right-angles to the probing beam), the anticorrelation effect could be seen only over the initial segment of the cross-correlation function. Although this experiment did not yield quantitative data, the very detection of the rotational contribution to the non-Gaussian noise component was an important achievement.

We have already repeatedly noted that the requirement that $N \sim 1$ is not only not necessary but may even be undesirable if one is concerned with the investigation of rotation or internal motion of macroparticles. The number of particles in the scattering volume was therefore chosen in Ref. 54 to be quite large, namely, $N \sim 10^2$. The object under investigation was a solution of *E. coli*, which we have already encountered. These are large particles and their orientational diffusion time is relatively long: $T_0 \sim 0.1$ s, i. e., it is of the order of the characteristic time for intensity fluctuations in the probing laser beam. Special measures were undertaken in this experiment to avoid distortion connected with this fact. The photodetectors were mounted at 45° to the incident beam and were arranged symmetrically relative to the beam. For this particular object (its dimensions were of the order of the wavelength), this arrangement is optimal for observations of anticorrelation. Figure 7 shows the measured $G^{b3}(\tau)$. As can be seen, the scattering volume was sufficiently large and the observed situation was wholly determined by rotational diffusion of the scattering particles. The coefficient of rotational diffusion determined from this curve turns out to be $\theta \sim 0.25$, which is in good agreement with existing data on the size and shape of the bacteria.

Thus, the cross-correlation method can be used to perform the perfect subtraction of the average values of the Gaussian and shot components from the resultant intensity spectrum which, as already noted, is particularly important when the correlation times T_0 and T_c are of the same order. It is appropriate to mention here the direct analogy between the above cross-correlation technique and the Brown-Twiss type experiments² in which coincidences are recorded between two photomultipliers as functions of the angular separation of the two counters relative to the light source. In our terminology, the aim of this experiment is to separate the

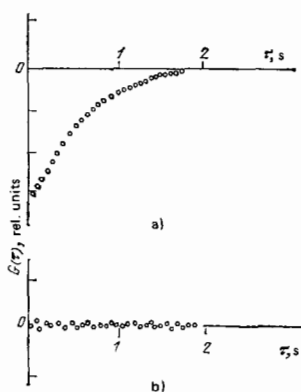


FIG. 7. (a) Cross-correlation function for light scattered by a suspension of *E. coli*; (b) the same for spherical latex particles.

integrated intensity of the Gaussian noise of the light field from the shot background. The separation of the components is possible because the shot noise of the two photodetectors is not cross-correlated, but the phase (Gaussian) noise is correlated so long as the two photodetectors are within the coherence area of the beam of diameter $l_c \sim \lambda/\varphi$, where φ is the angular size of the source. It follows that, by measuring the cross correlation between the photomultiplier counts as a function of separation between the receivers, it is possible to determine the angular size of the source (for example, a star).

In our case, the object under investigation is the non-Gaussian component of intensity fluctuations, which corresponds to fluctuations in the intensity of the source as a whole. To separate out this component, the detectors are taken well outside the limits of the coherence area. This destroys the cross correlation between photons that is connected with the wave noise in the radiation field, and the shot fluctuations are then always uncorrelated.

In conclusions of this section, we note two points. Firstly, all the above experiments were concerned with the investigation of orientational motion. This may have given the impression that observation of intrinsic internal dynamics, i. e., changes in the shape of the particle, involves some additional difficulty. In fact, this is not the case. The fact of the matter is that the theory describing the dynamics of changes in particle shape in a viscous medium is very complicated, and the shape of the formfactor fluctuation spectrum due to a flexible particle can only be determined within the framework of some particular simplified model whose physical reality is often problematic. It is therefore, not surprising that early demonstration experiments on the detection of non-Gaussian effects in IFS were concerned with the orientational motion of rigid particles because the situation is then quite clear from the theoretical point of view.

Secondly, most of this section was devoted to the situation in which the width of the formfactor fluctuation spectrum was very close to the diffusion width. The reasons why this situation is often encountered in practice was explained in Section 4A. Of course, it is possible to cite many cases in which these two widths are very different. For example, this occurs in the case of the spectrum of the depolarized component of light scattered through a small angle by optically anisotropic particles.⁴⁰ Although the non-Gaussian effects are then easier to observe, there is little point in such observations because the internal dynamics is much more readily detected directly in the scattered spectrum, i. e., within the framework of traditional IFS. This is why IFS techniques (which enable us to observe non-Gaussian effects) and, above all, the cross-correlation method, are the most promising for studies of internal dynamics precisely in the case of similar Gaussian and non-Gaussian widths.

CONCLUSIONS

We have tried to use a number of examples to demonstrate the possibilities of IFS, both in atomic physics

and in the physics of macroparticles. The basic idea that we have exploited was that the non-Gaussian component of intensity noise in radiation scattered or emitted by a medium can be a source of information about the internal motion of particles. The fact that such information can actually be extracted has now been recognized and demonstrated for objects that can in no way be regarded as unique.

When we consider the importance of methods developed within the framework of IFS, we must remember two aspects of this question. Firstly, the method is capable of yielding directly information about the objects that is important in practice (this includes atomic and molecular constants, rotational and translational diffusion constants, and so on). The other aspect is that the IFS approach generates new physical ideas and generalizations.

We have already noted that experiments on the application of IFS to microphysics are at present largely demonstrative in character. The practical advantages of IFS as compared with other methods have, indeed, been clearly demonstrated only for small-angle Mandel'shtam-Brillouin scattering. The possibilities of IFS in the study of the dynamics of macroparticles are much clearer. Such studies are now being widely conducted and the number of papers in this field continues to increase.

As regards the second aspect of the role of IFS, we note that this is not simply a new technique in spectroscopy but, above all, a new approach to the investigation of the electromagnetic field from a fundamentally new point of view. This is clearly seen in the example of the Hanbury Brown and Twiss effect, which, from the point of view of spectroscopy, provides traditional information on the spectrum and the spatial coherence of the source. However, this experiment involves a new feature of the electromagnetic field that is characterized by the bunching of photons. The new approach has created an interest in photon correlation and anticorrelation problems, in fields in quantum-mechanical states, and in questions relating to the limits of validity of classical electrodynamics. It is thus clear that IFS has turned out to be a very fruitful approach in fundamental respects as well.

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