Photocount statistics in the backscattering enhancement effect

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The photocount statistics of the backscattering enhancement effect are studied for a system of polystyrene latex in water. The photocount distribution function is obtained for incoherent scattering and in the presence of a coherent component for which a few of the first few moments are analyzed. It is shown that with decrease of the cumulation time the photocount distribution function goes over from the Poisson to the Bose–Einstein form.

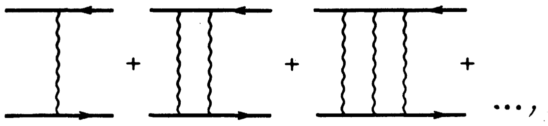
In the study of the scattering and propagation of light in strongly inhomogeneous media a great deal of attention has been paid to incoherent multiple scattering. However, as it turns out, to obtain a more complete picture of light scattering it is necessary to take into account the influence of interference at distances significantly greater than the extinction length σ. Such an analysis has been carried out in a number of articles,[1-3] from which it follows that coherent scattering can be observed by investigating the light scattered directly backward (180°). Indeed, the enhanced backscattering effect, which is manifested in the appearance of a peak with a half-width of a few tenths of a degree in the directly backscattered ($\theta = 180°$) fraction of the scattered light in strongly scattering systems with $\sigma \sim 10^2$–10^3 cm^{-1} against the intensity background of multiple incoherent scattering, was experimentally detected relatively recently.[4-6]

The most widely used material in the published studies of this subject is an aqueous solution of polystyrene latex. According to the reports, the coherent backscattering peak in these suspensions was observed by varying the dimensions of the suspended particles from 0.1 to 5 μm and the volume concentration from $5\cdot 10^{-3}$ to 0.15. It turns out that the magnitude and width of the peak are sensitive to the dimensions and concentration of the latex particles. Most strongly manifested is the dependence of the magnitude and width of the peak on the concentration of the suspended particles. The variation of the parameters of the peak in response to variation of the dimensions of the particles is very complicated.

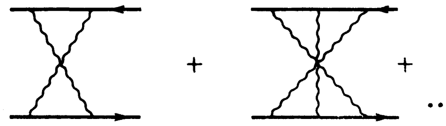
This effect is also observed very clearly in other physical systems, such as ceramics[7,8] and disordered liquid crystals in the nematic phase.[9]

A qualitative explanation for the appearance of this peak is as follows. Let multiple scattering in a strongly inhomogeneous medium containing a large number of scatterers be modeled as a sequence of re-radiations described by the wave vectors $\mathbf{K}_0, \mathbf{K}_1,\ldots,\mathbf{K}_m = \mathbf{K}_b$, where $\mathbf{K}_j$ is the wave vector of the jth scattering and $\mathbf{K}_m = \mathbf{K}_b$ is the vector of the last scattering. If the description assumes that the scattering processes are independent, all of the sequences of m scattering events can be taken to be independent, and the total scattered fraction is found by summing over the intensities. Such an approach, for example, is used in the description of scattering in the radiative transfer equation. However, any sequence of m scattering events can be put into correspondence with its inverse ($\mathbf{K}_0, -\mathbf{K}_{m-1},\ldots, -\mathbf{K}_1 = \mathbf{K}_b$), in which the light is scattered by the same inhomogeneities, but in the opposite direction. The fields in these two cases are coherent, and for direct backscattering will give an interference maximum with a width of the order of λ/L, where L is the mean free path of the photons, i.e., of the order of the extinction length.

A quantitative calculation of the angular distribution of this peak reduces to the summation of a series[10] over scattering multiplicities. In contrast with the usual situation in which only ladder diagrams are summed, here diagrams of two types must be taken into account—ladder diagrams and cyclic diagrams. Ladder diagrams are depicted in the following way


(1)

where the arrows denote the incident field, the straight lines—the propagation function, and the wavy lines—the correlations. These diagrams make the main contribution at all scattering angles. Cyclic diagrams


(2)

are important only near $\theta = 180°$.

It is possible to sum series (1) and (2) successively, and thereby obtain the shape of the peak, only in the case in which the correlation function $\varphi(\mathbf{r}_1,\mathbf{r}_2) \sim \delta(\mathbf{r}_1 - \mathbf{r}_2)$, i.e., for point scatterers with a circular indicatrix.[11-13] For the case of scatterers of finite dimensions summation of series (1) and (2) cannot be strictly carried out and one uses some kind of approximation.[14,15] Since on the other hand all of the measurements, as a rule, have been made in latex solutions with particle dimensions of the order of the wavelength of light, in this case a cumulation and systematization of most varied experimental data takes on a special significance. First, this is important in order to gain a more complete understanding of the phenomenon and, second, to provide a basis for an adequate theoretical description.

The overwhelming majority of the experimental data is obtained from measurements of the light scattering intensity during times long enough that statistical averaging takes place over all possible configurations of the system of scattering particles during the process of multiple scattering, i.e., these data comprise integrated light-scattering intensities.

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Along with a study of the integrated intensities, one method of analyzing systems with random inhomogeneities is to study their photocount statistics.[16,17] This problem has been attacked from various points of view, and its most important application is to provide a theoretical basis for correlation spectroscopy. By measuring the light scattering intensity over very short times—of the order of the lifetime of a concrete configuration— we obtain information about the instantaneous picture of the distribution of scatterers. From such data it is possible to judge, in particular, the statistics of different types of configurations—those loops that make the main contribution to the weak localization effect. It is impossible to obtain such information in any other way than by measuring the photocount statistics or, possibly, by computer experiment or modeling.

In any study of the photocount statistics the final result, as a rule, is the distribution function of the recorded intensities, which depends not only on the properties and dimensions of the investigated media, but also on the statistical characteristics of both the source and the receiver. Therefore, the problem of obtaining the statistical properties of the irradiated medium is to a significant degree analogous to the problem which arises in spectral investigations when it is impossible to neglect the apparatus function of the instrument. For example, the transmission of light through a turbulent atmosphere was studied in this way. Here the statistics of the intensity are determined by convolving the distribution function of the receiver (as a rule, a Poisson distribution) with the distribution function of the inhomogeneous atmosphere.[18] The statistics of laser radiation have been studied experimentally in complete detail for various generation regimes.[19,20]

As of the present time an analogous systematic study of strongly scattering systems has only been begun.[21,22] This problem was first discussed in Ref. 21, where successive instantaneous spatial intensity distributions in the speckle structure of a backscattered signal were recorded with the help of a fast videocamera with exposure time $\tau \sim 10^{-5}$ s. It was found that along with small variations about the mean of the intensity there are observed regions with sharply increased scattering intensity, analysis of which requires a knowledge of the photocount distribution function.

Measurements of the distribution function were carried out in Ref. 22 on a device intended for measurements of the temporal correlation function for cumulation times τ much shorter than the characteristic time needed by the particles to move a distance of the order of the wavelength of the light. Backscattering was investigated outside the cone of the coherent peak. The experimentally obtained distribution function was approximated by the gamma distribution

$$P(I)=\frac{\alpha^\alpha}{\Gamma(\alpha)}\left(\frac{I}{\langle I\rangle}\right)^{\alpha-1}\exp\left(-\frac{\alpha I}{\langle I\rangle}\right)$$

with distribution parameter $\alpha = 1.3$, where I is the intensity measured over a time τ, $\langle I\rangle$ is its mean value, and $\Gamma(\alpha)$ is the gamma function. It is believed in Ref. 22 that the closeness of α to unity testifies to the Gaussian character of the experimentally observed distribution function.

In the present work we have carried out a detailed analysis of the photocount statistics both inside and outside the backscattering cone for various cumulation times in the channels of a multichannel analyzer. Our results agree with

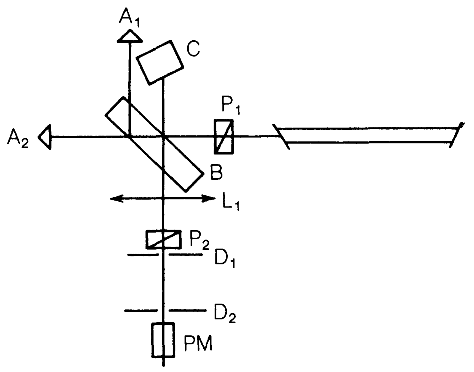

FIG. 1. Diagram of experimental setup for investigating the enhanced backscattering effect: C—cell, B—beam splitter, L_1—lens ($f = 50$ cm), P_1 and P_2—polarizers, PM—photomultiplier, D_1 and D_2—circular diaphragms ($\varnothing = 0.2$ mm).

the current picture of the photon statistics in randomly inhomogeneous media.

The measurements were carried out on the device depicted in Fig. 1. Argon and helium-neon lasers with beam divergence angles ≈ 0.6 mrad were used as the light sources. The main element of devices used to study the enhanced backscattering effect is a beam splitter, with the help of which part of the laser radiation is directed into the investigated medium. Since the scattering is into angles close to and including 180°, the light scattered along the path to the photodetector must pass through the beam splitter. The main requirement on this optical element is that it not distort the incident beam or increase its divergence. As the beam splitter in our setup we used a plane-parallel plate 2 cm in thickness with a parallelism discrepancy not greater than 30″. A plate of such large thickness is quite suitable for aligning the device and allows one to determine the position of the focal plane of the long-focal-length lens L_1 with high accuracy.

The scattered radiation passed through a polarizer and a diaphragm of 0.2 mm diameter to a photomultiplier. The photocount statistics were analyzed with the help of a multichannel analyzer and subsequent mathematical reduction. The magnitude of the recorded radiation was chosen to be contained with sufficient margin in the region of linearity of the photon counter.

The intensity of the scattered light was measured in the center of the peak ($\theta = 180°$) and on the background ($\theta = 170°$). Figure 2 shows a combined photograph of the display for these two angles. The ordinates of the points correspond to the scattering intensity accumulated in the channels after a time $\tau \sim 10^{-3}$ s. It is clear that, first of all, the integrated intensity at the peak is greater than in the background. The photograph clearly demonstrates that the scatter of the instantaneous photocounts in both the peak and the background is strongly asymmetric due to the large number of large peaks in the intensity. This is manifested particularly clearly for the direct backscattering, where there exist excursions, for example, 4–5 times higher than the mean values.

A systematic cumulation of photocounts was carried out, and the so-obtained data were subjected to mathematical analysis. Figure 3 shows typical distribution functions in the peak and in the background $P_p(n)$ and $P_{bg}(n)$, where n

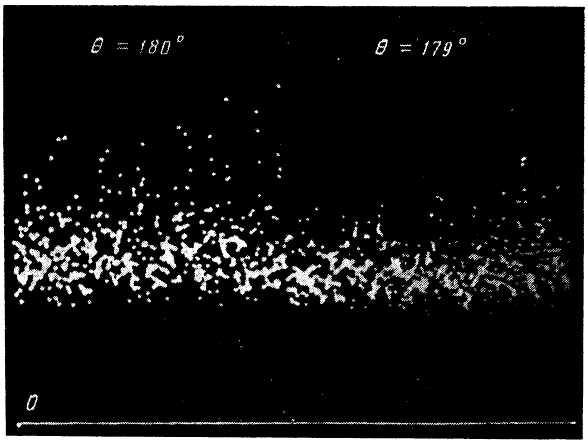

FIG. 2. Photograph of multichannel analyzer display. For each scattering angle 512 values were recorded. Diameter of latex particles $d_l = 1.2\ \mu$m. Cumulation time in both cases $5\cdot 10^{-3}$ s.

is the number of pulses recorded during the cumulation time τ. For convenience of analysis and comparison of various experimental data for different τ the mean values $\langle n\rangle$ in the peak were reduced in the processing of the data to one value. The magnitudes of $\langle n\rangle$ in the background were recalculated correspondingly. It is clear that $P_p(n)$ is much wider than $P_{bg}(n)$ mainly because of the part corresponding to larger values of n.

The most important and interesting problem is to separate the contribution to the direct backscattering intensity from the cyclic diagrams (2) corresponding to the coherent component. Since the solution of such problems is a separate and complicated mathematical problem, we will restrict ourselves to an analysis of a few of the higher moments of the distribution function. We assume that the direct backscattering is a sum of two random processes, described by the cyclic (cr) and ladder (ld) diagrams.

As is well known from probability theory,[23] the moments of random processes are connected in such a case by simple relationships:

$$M^{tot}_{(i)}=M^{cr}_{(i)}+M^{ld}_{(i)},\quad i=1,2,3, \tag{3}$$

$$M^{tot}_{(4)}=M^{cr}_{(4)}+M^{ld}_{(4)}+2M^{cr}_{(2)}M^{ld}_{(2)},$$

. . . .

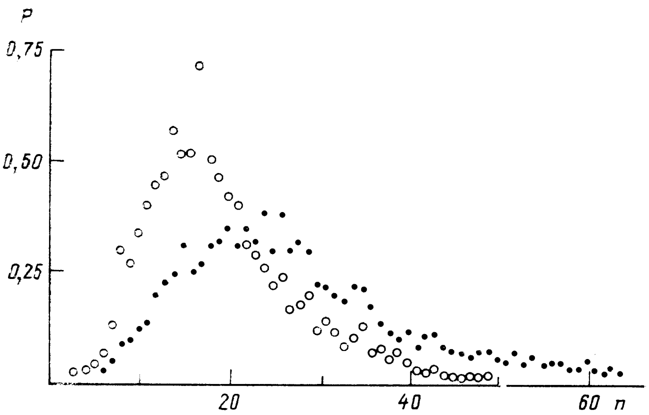

FIG. 3. Distribution function $P(n)$ for the peak and the background. ●– scattering angle 180°. ○– scattering angle 170°, $\tau = 10^{-3}$ s, $d_l = 5.1\ \mu$m.

The intensity fluctuations due to incoherent scattering described by the ladder diagrams can be found from measurements outside the backscattering cone. As experiment has shown, their values depend weakly on scattering angle.

The moments in the peak and in the background were calculated from the data sets $\{n_p\}$k and $\{n_{bg}\}$. The moments corresponding to the cyclic diagrams were then calculated from formulas (3). This procedure was carried out for times τ in the interval from $5\cdot 10^{-5}$ to $5\cdot 10^{-2}$ s. As could be expected, as a result of partial averaging all of the higher moments ($i = 2,3,4$) decay as τ grows. For the sake of comparison they were reduced to dimensionless form by dividing by the corresponding powers of the mean intensities. From the ratios of the moments for different scattering angles it is possible to judge the nature of the physical processes which give rise in the given case to the light scattering. If the intensity fluctuations had one cause, then the relative fluctuations in the peak and background would be identical.

In our case the relative fluctuations in the peak are 1.5–2 times higher than in the background. A still greater difference is obtained for the calculated values of the moments due only to the coherent part of the scattered light. For example, the relative mean square fluctuation in the peak is 5–7 times greater than the corresponding fluctuation in the background. For the fourth moment this difference is greater yet by almost an order of magnitude. Hence one may conclude that the distribution function $P(n)$ for coherent scattering should be highly asymmetric with a large half-width. If we consider the scattering in the observed volume from this point of view, then we are led to the conclusion that the intensity fluctuations of the scattered light at $\theta = 170°$ are determined mainly by the variation of the number of particles N as a consequence of Brownian motion. For coherent scattering the existence of a wide asymmetric distribution function indicates that important for the cyclic diagrams is not only the value of N but also the mutual arrangement of the particles.

In order to construct the intensity distribution function in its explicit form we must take the shot noise produced by the receiver into account. It is well known that this leads to a convolution of the intensity of the investigated light with the apparatus function of the receiver, which is a Poisson distribution. All of the results presented above belong to the case in which the volume from which the radiation is incident upon the receiver is larger than the coherence region. This leads to the result that even for the shortest cumulation times $\approx 5\cdot 10^{-5}$ s there is an additional averaging over space.

To eliminate the need for spatial averaging, a series of measurements were carried out with an additional diaphragm which decreased the observed volume by almost an order of magnitude. The measurements were carried out for the scattering angle 180°. For such experimental conditions it is possible to verify that the limiting behavior of $P(n)$ obtains for small and large cumulation times τ. As is well known, at very small times, under the condition that the system under investigation obeys Gaussian statistics, the function $P(n)$ should, as a result of convolution with the Poissonian shot noise of the receiver, have the form of the Bose–Einstein distribution:

$$P(n)=\frac{\langle n\rangle^n}{(1+\langle n\rangle)^{n+1}}. \tag{4}$$

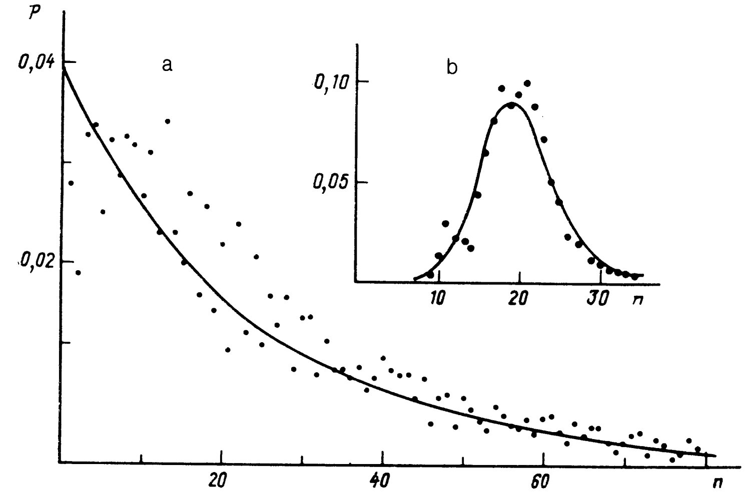

FIG. 4. Distribution function: a) $\tau = 5\cdot 10^{-5}$ s, b) $\tau = 5\cdot 10^{-2}$ s. Points—experiment, solid curve—calculated according to formula (4) in a), according to formula (5) in b); $d_l = 1.2\,\mu$m.

In the case of large times τ, when the averaging is carried out over all possible realizations of the Gaussian system, an analogous convolution will reproduce the apparatus function, i.e., the Poisson distribution:

$$P(n)=\frac{\langle n\rangle^n}{n!}e^{-\langle n\rangle}. \qquad (5)$$

Figure 4 demonstrates the validity of these ideas. Note that the solid lines, which correspond to the theory, are plots of formulas (4) and (5), which contain no adjustable parameter other than $\langle n\rangle$, which is determined from experiment.

For intermediate times we calculated the first four factorial moments:

$$\frac{M_i}{M_1^i}=\frac{\langle n^i\rangle}{\langle n\rangle^i},\quad i=2, 3, 4. \qquad (6)$$

As is well known,[17,24] for random processes described by the Bose–Einstein distribution, in the case of limitingly small cumulation times these moments should equal $i!$. In the limit of large times all of the factorial moments approach unity.

Figure 5 shows the dependence of $\ln[M_i/M_1^i]$ on τ. It is clear that the character of the dependence of the corresponding moments agrees with the theoretical predictions, although the limiting values of M_i/M_1^i are not obtained. This is explained by the fact that even if two diaphragms are used the observed volume somewhat exceeds the coherence region. Such a result is also confirmed by the form of the experimentally obtained distribution function at small times (Fig. 4a), in which the maximum is shifted slightly from $n=0$.

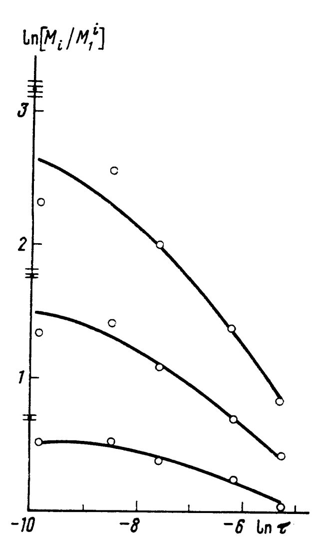

FIG. 5. Normalized factorial moments as functions of cumulation time τ. Points—experiment; two hatch-marks on the vertical axis—$M_2/M_1^2 = 2!$, three—$M_3/M_1^3 = 3!$, four—$M_4/M_1^4 = 4!$ show the theoretical limiting values.

The results show the implemented approach to be promising for studies of the enhanced backscattering effect. A more detailed investigation of the photocount distribution function under various experimental conditions should make it possible to obtain detailed information about the parameters of scattering systems and the kinetics of optical inhomogeneities.

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