Electromagnetic field correlations between differently directed modes in elastic light scattering

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We have experimentally investigated the intensity correlations in light generated by elastic scattering of a standing pump wave by macroscopic objects, i.e., particles with sizes on the order of a micron. We have observed that the spatial correlation function is a maximum for oppositely directed modes of scattered light. We show that this phenomenon is analogous to the Brown–Twiss effect for a pseudothermal source.

1. INTRODUCTION

One of the most common methods used to investigate the properties of a randomly nonuniform medium is based on spectroscopy of intensity fluctuations in the light scattered by the medium.[1] Media of this kind, which are characterized by parameters that are random both in time and in space, cause additional fluctuations in the scattered optical fields. For this reason, measurements of the intensity correlation functions can yield valuable information on the scattering medium, information that is difficult to obtain by other methods. The first experiments of this kind were performed by Brown and Twiss[2] in 1955–57, who observed the fourth moment of the optical fields for light generated by a mercury lamp.

The information about the scattering medium that can be obtained by this method depends on the character of the medium. Thus, by studying the intensity correlation function of a non-Gaussian scattering medium we can to draw certain conclusions about the behavior of individual scatterers.[3,4]

There is also great interest in cases where the scattered light exhibits significantly nonclassical properties.[5] In Refs. 5, 6 it was shown that photon correlations exist at low temperatures and outside of Raman resonances, caused by simultaneous creation of a pair of Stokes (S) and anti-Stokes (A) photons as a result of spontaneous decay of two pump quanta. In this case the statistics of the scattered light should be nonclassical, i.e., the probability of creating such a pair will be considerably higher than the product of probabilities for the creation of individual photons in the S and A modes. The distinguishing feature of this effect is the possibility of a change in the statistics of the fields that are created from pure quantum-mechanical, i.e., with high contrast ($m \gg 1$, see below) to classical thermal ($m=1$) by varying parameters such as the frequency of the scattered light (i.e., its closeness to resonance) and/or the effective phonon temperature. Thus, as the combination-tone frequency shift $\Delta\omega=\omega_A-\omega_L=\omega_L-\omega_S$ decreases, where ω_L, ω_S, and ω_A are the pump, Stokes, and anti-Stokes wave frequencies respectively, the value of the contrast drops to a value $m_{\text{class}}=1$, which in the Raman-scattering limit becomes the molecular value for fluctuations in the orientation and concentration of the molecules. Analogous phenomena should also occur in continuous media that support quasielastic scattering; e.g., during phase transitions (critical opalescence) and in turbid media consisting of suspensions of macroparticles.

In this paper we experimentally investigate a limiting case of these correlations: correlations between differently directed modes when a two-beam pump is elastically scattered in a turbid medium.

The theory of this effect was developed in Ref. 7, where it was shown that when a two-beam pump is scattered in a turbid medium, correlations should be observed in the light intensity for light scattered in directions related by the phase matching conditions

$$\mathbf{k}_1^L+\mathbf{k}_2^L=\mathbf{k}_1^S+\mathbf{k}_2^S, \tag{1}$$

$$\mathbf{k}_1^L-\mathbf{k}_2^L=\pm(\mathbf{k}_1^S-\mathbf{k}_2^S), \tag{2}$$

where $\mathbf{k}_i^L$, $\mathbf{k}_i^S$ $(i=1,2)$ are wave vectors for the incident and scattered waves respectively. This correlation can be explained within the classical theory of simple scattering,[8] in which the statistical correlation between the fields in modes $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$ arises from the same Fourier component of the dielectric constant fluctuations $\Delta\varepsilon_q=(\Delta\varepsilon_{-q})^*$. Here $\Delta\varepsilon$ is a fluctuation in the dielectric constant, $\varepsilon=\langle\varepsilon\rangle+\Delta\varepsilon$, while $\Delta\varepsilon_q$ is the spatial harmonic in the Fourier expansion of the real function $\Delta\varepsilon(\mathbf{r})$; $\mathbf{q}=\mathbf{k}^S-\mathbf{k}^L$ is the scattering vector.

The value of the contrast, which characterizes the intensity correlations, is:

$$m=\frac{\langle E_1^S(E_1^S)^*E_2^S(E_2^S)^*\rangle}{\langle E_1^S(E_1^S)^*\rangle\langle E_2^S(E_2^S)^*\rangle}-1, \tag{3}$$

which equals zero for statistically uncorrelated fields, turns out to equal 1 and 1/4 in phase matching directions (1) and (2), respectively, when the following conditions are fulfilled:

1) the scattering is simple scattering;

2) both the correlation radius of fluctuations in the dielectric permittivity r_c and the wavelength λ are much smaller than the dimensions of the scattering volume R_s (i.e., the fluctuations $\Delta\varepsilon_q$ are independent and Gaussian in character).

It was also shown in Ref. 7 that in the case of a two-beam pump, the phase-matching condition for correlation between Stokes and anti-Stokes modes becomes condition (1) in the limit of quasielastic scattering.

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According to Ref. 7, we can estimate the angular width $\Delta\Phi$ of the maxima exhibited by the intensity spatial correlation function in the phase matching directions (1) and (2) from the relation:

$$\Delta q R_s \simeq \pi, \tag{4}$$

where $\Delta \mathbf{q} = \mathbf{k}_1^L - \mathbf{k}_2^L - \mathbf{k}_1^S + \mathbf{k}_2^S$ is the detuning from wave phase matching. Then

$$\Delta\Phi \simeq \lambda / R_s, \tag{5}$$

and the quantity $\Delta\Phi$ depends only on the size of the scattering volume (for a given wavelength).

In this work, we experimentally investigated the intensity correlation function for light scattered in the opposite directions $\mathbf{k}_1^S$ and $\mathbf{k}_2^S = -\mathbf{k}_1^S$ when a turbid medium is illuminated by a two-beam pump with wave vectors $\mathbf{k}_1^L$ and $\mathbf{k}_2^L = -\mathbf{k}_1^L$; these directions fulfil the condition (1).

We note that a similar scattering geometry was investigated both theoretically and experimentally in previous papers by Fillies[9,10] and Dhont *et al.*[11,12] These papers showed that in this geometry the diffusion coefficient of Brownian particles can be determined in terms of the time dependence of the cross correlation function for the intensity of scattered light, even in the case of multiple scattering.

In this paper we show that in the single-mode detection limit, the statistics of light elastically scattered by small particles ($a \simeq \lambda$) is thermal in character and does not depend on the size and number of particles over rather wide limits.

2. PHENOMENOLOGICAL MODEL OF THE EFFECT

Consider a situation where the scattering medium consists of a set of randomly moving particles whose dimensions are comparable to the wavelength of the light.

The appearance of correlated fluctuations in the directions determined by (1) can be explained in the following way. Assume that there happen to be two particles A and B in the scattering volume V_s (Fig. 1). The intensity of the field scattered in the direction $\mathbf{k}_1^S$ is determined by the difference in optical path lengths:

$$\Delta_1 = AD + AF, \quad \Delta_2 = AF - BC \tag{6}$$

and in the direction $\mathbf{k}_2^S$:

$$\Delta_3 = BC + BE, \quad \Delta_4 = BE - AD. \tag{7}$$

Because of the symmetry of the scattering geometry specified by condition (1), we have $\Delta_1 = \Delta_3$, $\Delta_2 = \Delta_4$. This leads to temporal phase matching of the intensity fluctuations (caused by the relative motion of particles A and B) in the directions $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$. For an arbitrary number of particles in the scattering volume, the total intensities in modes $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$ are determined by the expression:[8]

$$I_1^s \simeq N I_0 \left(1 + \frac{1}{N} \sum_{k \neq j} \exp[\pm i \mathbf{q} * (\mathbf{R}_j - \mathbf{R}_k)] \right), \tag{8}$$

where N is the number of particles, I_0 is the incident wave intensity, $\mathbf{q}$ is the scattering vector, and $\mathbf{R}_j$ is the radius vector of the jth particle.

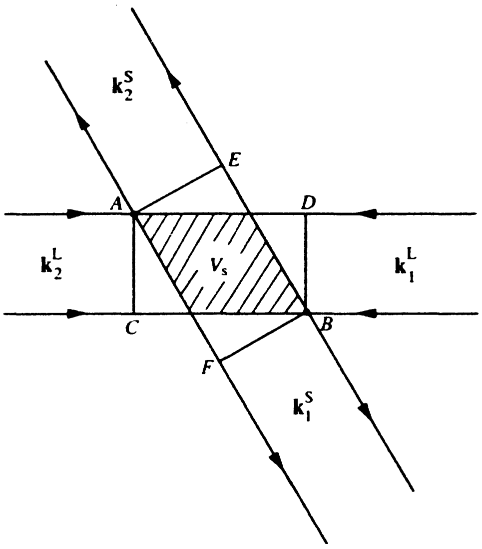

FIG. 1. Mutual orientation of pump-mode $\mathbf{k}_1^L$ and $\mathbf{k}_2^L$, ($\mathbf{k}_1^L = -\mathbf{k}_2^L$) and scattered-light wave vectors $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$, ($\mathbf{k}_1^S = -\mathbf{k}_2^S$) when the phase matching condition (1) is fulfilled. The detector volume contains two particles A and B.

Equation (8) contains interference terms from all possible pairs of particles (for N particles there are $(N^2 - N)/2$ of these terms). Once again, by virtue of the symmetry of the problem, the intensity fluctuations in modes $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$ are phase matched. We can explain the phase matching of fluctuations in the light intensity in the directions (1) for scattering by large ($a > \lambda$) particles or in a continuous randomly nonuniform medium.

Here it is necessary to mention the analogy between this phenomenon and correlations in intensities from the pseudothermal source (a nonspecular rotating disc) used in the Brown–Twiss interferometer.[13] In our case, the turbid medium plays the role of the thermal source, transforming the Poisson statistics of the laser pump photons into thermal statistics with a geometric distribution function, while the scattering geometry, which is specified by the phase matching conditions (1), acts as a beam-splitter that spatially separates the modes that undergoing correlation.

3. EXPERIMENTAL SETUP AND MEASUREMENT METHOD

Figure 2 shows a sketch of our experimental setup for investigating intensity correlations in elastic scattering. Collinear counterpropagating pump waves $\mathbf{k}_1^L$ and $\mathbf{k}_2^L$ pass through long-focus lenses *4* and converge in the scattering volume V_s. As scattering media we used:

1. droplets of milk suspended in distilled water,
2. rosin vapors blown through the volume V_s,
3. dust particles present in air.

The scattered light passes through a system of irises *5–5* and *6–6* and arrives at two photomultipliers (FEU-79) *7*, operating in photon counting mode. The photocurrent pulses are fed to two counters *9*, *10*, and to a coincidence circuit *8* which sends a pulse to the output whenever a pair of pulses from the photomultiplier happen to appear at both of its inputs within a time interval $T_{\text{eff}} = 5$ ns. The time T_{eff} was

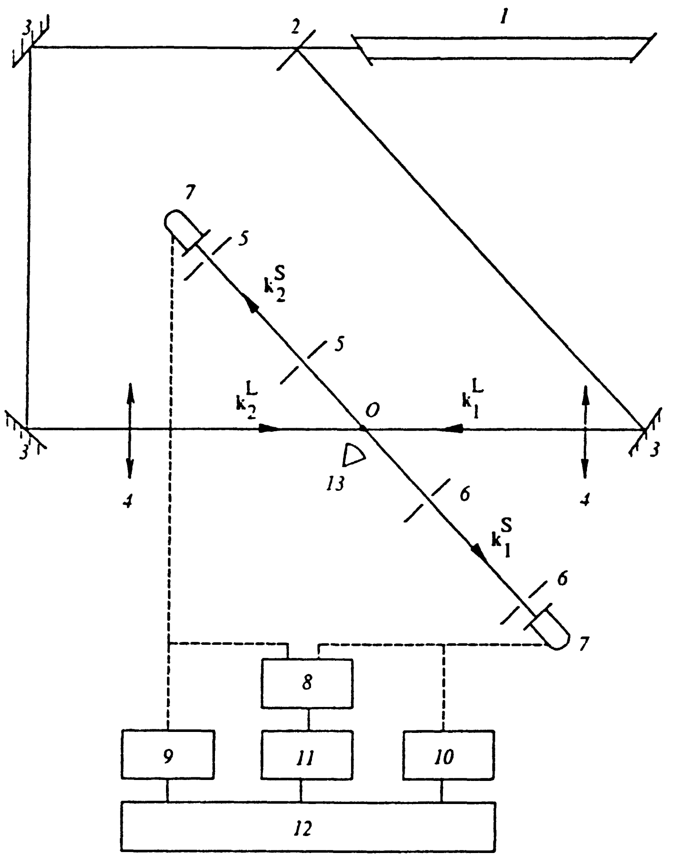

FIG. 2. Sketch of experimental setup for investigating intensity correlations for elastic light scattering. $\mathbf{k}^S_{1,2}$, $\mathbf{k}^L_{1,2}$ are the modes of scattered radiation and pump. *1*—Ar^+ laser; *2*—beam splitter; *3*—rotating mirror; *4*—long-focus lenses; *5, 6*—irises; *7*—photomultiplier; *8*—coincidence circuit; *9, 10, 11*—pulse counters; *12*—microcomputer; *13*—photodiode.

determined by calibration measurements in which both photomultipliers were illuminated by statistically independent fields. The source of these fields was emission from a photodiode *13* with a stabilized power supply. The microcomputer *12* samples the inputs of the counters with a sampling time T (0.1 s $<T<$1 s) and computes the normalized correlation function

$$g_2 \simeq \frac{\langle I_1 I_2\rangle}{\langle I_1\rangle\langle I_2\rangle} \simeq \frac{\langle N3\rangle}{T_{\text{eff}}\langle N1\rangle\langle N2\rangle}, \tag{9}$$

and contrast

$$m \simeq g_2 - 1, \tag{10}$$

where $N1$, $N2$, $N3$ are the counting rates for pulses from photomultipliers *9*, *10* and the coincidence circuit *11*, respectively.

By rotating the system of irises *5–5* and *6–6* and detectors *7* around the point O, which coincides with the center of the scattering volume, we were able to measure the angular dependence of the correlation function.

4. DISCUSSION

Let us discuss the results of our measurements for each of the scattering media listed below:

1. For light scattering in a weak solution of milk in distilled water placed in an optically flat quartz cell, the size of the scattering volume was $R_s \simeq 0.8$ mm. This size was determined by the diameter of the laser spot size, the distance from the scattering region to the photomultiplier ($L=500$ mm), and the diameter of the iris ($\phi \simeq 0.8$ mm). The size of the milk droplets suspended in water was determined by a microscope to be at most of the order of the wavelength, and the number of droplets present in the scattering volume was some tens of thousands. In this case, the scattering may be considered simple scattering, because the "multiple-scattering parameter" (see Ref. 8) is

$$x = a^2 R_s / l^3 < 1, \tag{11}$$

where a, l, R_s are the characteristic size of the particles, the average distance between them, and the scattering volume.

The value of the contrast when condition (1) is satisfied exactly turns out to be

$$m \simeq 0.07 \pm 0.01, \tag{12}$$

which agrees with the estimate based on Eq. (5). Actually, for $R_s \simeq 0.8$ mm, the angular width of the signal radiation recorded by the photomultiplier is determined by the diameters of irises 5, 6 and the distance from the photomultiplier to the scattering volume:

$$\delta\varphi \simeq 2R_s/L \simeq 0.2°. \tag{13}$$

However, the angular width of the correlation function from (5) was found to be

$$\Delta\Phi \simeq \lambda / R_s \simeq 0.03°. \tag{14}$$

Thus, the detection volume $V_{\text{det}} = cT_{\text{eff}} R_s^2 \sim (\delta\varphi)^2$ (see Ref. 5) significantly exceeds the coherence volume $V_{\text{coh}} \sim (\Delta\Phi)^2$ of the scattered field, and the contrast is roughly

$$m \simeq \frac{1}{V_{\text{det}}/V_{\text{coh}}} = \left(\frac{\Delta\Phi}{\delta\varphi}\right)^2 \simeq 0.03. \tag{15}$$

The factor $V_{\text{det}}/V_{\text{coh}}$ is essentially the effective number of independent modes recorded by the detector. Attempts to reduce the number of detected modes leads to familiar problems in experiments of this kind[14] related to a decrease in the signal-to-noise ratio.

2. By using rosin vapor as the scattering medium, i.e., small particles with dimensions $a \sim \lambda$, we were able to significantly increase the intensity of the scattered light (while nevertheless remaining within the range of applicability of the simple scattering model). For smaller irises and laser spot sizes ($\varnothing \simeq 0.5$ mm), and consequently a smaller value of V_{det}, the value of the contrast was

$$m \simeq 0.17 \pm 0.04. \tag{16}$$

The correlation time τ_c for this pseudothermal source turns out to be of the order of microseconds: $\tau_c \simeq \lambda / v$, where v is the linear velocity of a particle moving in the volume V_s ($v \simeq 10$ cm/s). However, measuring the temporal intensity correlation function, whose width determines the value of τ_c, lies outside the purview of this work.

3. An interesting way to obtain experimental information about the scattering medium is to measure the intensity cor-

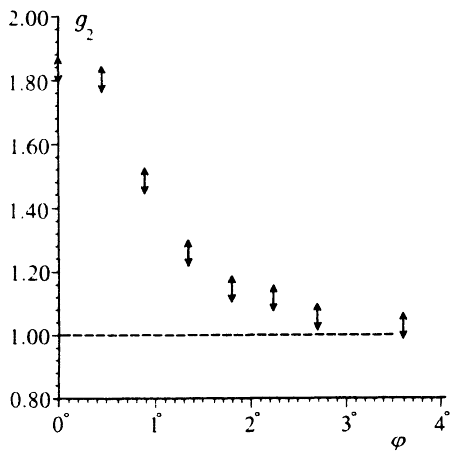

FIG. 3. Angular dependence of the normalized intensity correlation function for scattering by particles of dust (low concentration). The points are levels corresponding to completely uncorrelated fields in the modes $\mathbf{k}_1^S$ and $\mathbf{k}_2^S$.

relation function when $V_{det} < V_{coh}$. In this case, a suitable object of study is light scattering by ordinary room dust at rather low concentrations. The sizes of dust particles were determined to lie between a fraction of a micron and a few microns. It is important that the average number of scattering particles (in the scattering volume) should not be very large, say $N \simeq 10$. This implies that the number, and consequently the size of the scattering volume R_s, undergo significant fluctuations. For this reason, the average size of the scattering volume, and consequently the detection volume, turn out to be smaller than in the previous cases (with all other conditions equal). The value of the contrast was measured to be around

$$m \simeq 0.9 \pm 0.06, \tag{17}$$

decreasing smoothly to zero with detuning from the direction of phase matching (1). The angular width of the normalized correlation function (Fig. 3) was

$$\Delta\Phi \simeq 0.6° > \delta\varphi. \tag{18}$$

This allows us to estimate the average distance between dust particles:

$$\bar{l} \simeq \frac{\lambda}{\Delta\Phi} \simeq 40 \ \mu\text{m}. \tag{19}$$

Note that according to Ref. 7, the degree of mutual coherence of the pump beams does not affect the value of the contrast. Actually, when a phase delay is introduced into one of the arms of the pump beam (the mode $\mathbf{k}_1^L$), we observe no change in contrast for long laser coherence lengths (for the Ar^+ we used, $l_{coh} \simeq 30$ cm).

CONCLUSIONS

Thus, in this paper we have investigated the classical limit of the effect predicted in Ref. 6 which allows us to create a source of optical fields with adjustable statistics. By recording a rather small number of modes (in the limiting case, one) we have shown that the statistics of the quasielastic scattered fields are pseudothermal in character, which accords with the conclusions arrived at in Ref. 6 in the limit of zero Raman frequency shift $\Delta\omega$ (the Rayleigh limit); in this limit, the contrast we observed for the intensity correlation function is close to unity.

The scattering geometry used in this paper, i.e., with oppositely-directed modes of scattered light ($\mathbf{k}_1^S = -\mathbf{k}_2^S$) in the field of a standing-wave laser pump ($\mathbf{k}_1^L = -\mathbf{k}_2^L$), is analogous to the partially reflective mirror used in the Brown–Twiss interferometer, in that it allows us to spatially separate the correlating modes.

By using the measured width of the maximum of the correlation function, we can estimate medium parameters such as the concentration of independent scatterers.

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