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. © 1994 American Institute of Physics.

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 Twiss² 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=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 Δω=ωS−ωA decreases, where ωS and ωA are the pump, Stokes, and anti-Stokes wave frequencies respectively, the value of the contrast drops to a value m=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

\[
\begin{align*}
|k_i^s - k_i^a| &= \omega_s^2 + \omega_a^2 - 2\omega_s \omega_a, \\
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\end{align*}
\]

where \(k_i^s\) and \(k_i^a\) are wave vectors for the incident and scattered waves respectively. This correlation can be explained within the classical theory of simple scattering,⁶ in which the statistical correlation between the fields in modes \(k_i^s\) and \(k_i^a\) arises from the same Fourier component of the dielectric constant fluctuations \(\Delta \epsilon_r\). Here \(\Delta \epsilon_r\) is a fluctuation in the dielectric constant, \(\epsilon_r = \epsilon_r + \Delta \epsilon_r\), and \(\Delta \epsilon_r\) is the spatial harmonic in the Fourier expansion of the real function \(\Delta \epsilon(r)\).

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

\[
m = \left( \frac{\langle E_s^2 \rangle \langle E_a^2 \rangle}{\langle E_s^4 \rangle \langle E_a^4 \rangle} \right) - 1,
\]

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 \(\epsilon_r\) and the wavelength \(\lambda\) are much smaller than the dimensions of the scattering volume \(R_s\) (i.e., the fluctuations \(\Delta \epsilon_r\) 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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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.
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:

\[
\Delta q = \pi R_i, \quad (4)
\]

where \( \Delta q = k_1^s - k_1^t - k_2^s + k_2^t \) is the detuning from wave phase matching. Then

\[
\Delta \Phi = \lambda / R_i, \quad (5)
\]

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 \( k_s^1 \) and \( k_t^1 \) when a turbid medium is illuminated by a two-beam pump with wave vectors \( k_s^2 \) and \( k_t^2 \). These directions fulfill the condition (1).

We note that a similar scattering geometry was investigated both theoretically and experimentally in previous papers by Fillies\(^{2,9}\) 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 \((a>\lambda)\) 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 \( k_s^2 \) is determined by the difference in optical path lengths:

\[
\Delta_1 = AD + AF, \quad \Delta_2 = AF - BC
\]

(6)

and in the direction \( k_t^2 \):

\[
\Delta_3 = BC + BE, \quad \Delta_4 = BE - AD.
\]

(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 \( k_s^2 \) and \( k_t^2 \). For an arbitrary number of particles in the scattering volume, the total intensities in modes \( k_s^2 \) and \( k_t^2 \) are determined by the expression:

\[
I_s = N I_0 \left( 1 + \frac{1}{N} \sum_{i \neq j} \exp \left( \mp i q \cdot (R_i - R_j) \right) \right), \quad (8)
\]

where \( N \) is the number of particles, \( I_0 \) is the incident wave intensity, \( q \) is the scattering vector, and \( R_i \) is the radius vector of the \( j \)-th particle.

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 \( k_s^2 \) and \( k_t^2 \) 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 interferometers.\(^{13}\) 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 undergo 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 \( k_s^2 \) and \( k_t^2 \) pass through long-focus lenses \( F \) 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) \( T \), operating in photon counting mode. The photocurrent pulses are fed to two counters 9, 10, and to a coincidence circuit \( \Phi \) 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 \( T_{\text{eff}} \) was
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 $R_s = 0.8$ 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 ($\phi = 0.8$ 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

$$x = a^2 R_s / L < 1,$$

where $a$, $I$, $R_s$ 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

$$m = 0.07 \pm 0.01,$$

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

$$\delta \varphi = 2R_s / L = 0.2^\circ.$$

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

$$\Delta \varphi = \lambda / R_s = 0.03^\circ.$$

Thus, the detection volume $V_{det} = cT_0R_s^2 / (\Delta \varphi)^2$ (see Ref. 5) significantly exceeds the coherence volume $V_{coh} = (\Delta \varphi)^2$ of the scattered field, and the contrast is roughly

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

The factor $V_{det} / V_{coh}$ 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 related to a decrease in the signal-to-noise ratio.

1. By using rosin vapor as the scattering medium, i.e., small particles with dimensions $a - \lambda$, 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 ($0.1 - 0.5$ mm), and consequently a smaller value of $V_{det}$, the value of the contrast was

$$m = 0.17 \pm 0.04.$$

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

2. 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.

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 $k_s$ and $k_s$.

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 $N = 10$. This implies that the number, and consequently the size of the scattering volume $R$, 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

$$m = 0.9 \pm 0.06,$$

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

$$\Delta \Phi \approx 0.6' > \delta \phi.$$

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

$$\lambda = \frac{\lambda}{\Delta \Phi} \approx 40 \ \mu m.$$

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 $k_r$), we observe no change in contrast for long laser coherence lengths (for the Ar$^+$ we used, $\lambda_{coh} = 30$ 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 ($k_i = -k_i$) in the field of a standing-wave laser pump ($k_r = -k_r$), 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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