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Observation of multiple scattering of laser radiation from a light-induced jet of microparticles in suspension

A.V. Kondrat'ev

Abstract. **Variation in the correlation function of light multiply scattered by a random medium was observed with increasing the incident beam power. The light-induced motion of microparticles in suspension, caused by a high-power laser radiation, serves as an additional factor in the decorrelation of the scattered light. The experimental data are in good agreement with the results of theoretical analysis.**

Keywords: *diffusing wave spectroscopy, multiple scattering of light, light-induced motion.*

Following the development of the diffusing wave spectroscopy in the last decade, a new impetus has been imparted to the study of properties of random media (microparticle suspensions, foams, etc.) [1]. Measurement of the time correlation function (CF) $C_1(t) = \langle E(0)E^*(t)\rangle$ of light scattered by a random medium provides information about parameters of the medium such as the size of scattering particles, their characteristic velocity, and localisation of the flows of scatterers in a homogeneous medium [2].

In the diffusion approximation, the correlation function of light scattered by a suspension of spherical Brownian particles can be written for the backscattering geometry in the form [1]

$$C_1(t) = \exp(-\gamma t/\tau_0), \qquad (1)$$

where $\tau_0 = (4k_0^2 D)^{-1}$; $k_0 = 2\pi/\lambda$; λ is the wavelength of light in the medium; $D = kT/(6\pi\eta r_0)$ is the diffusion coefficient of particles of radius r_0 in a liquid with viscosity η at a temperature T; and γ is a factor of the order of unity whose value depends on the size of the particles and the polarisation of light being detected. This experiment was initiated by the results presented in Ref. [3], where the authors predicted and analysed theoretically the effect of multiple scattering of light by a light-induced jet of microparticles in suspension. For a sufficiently high intensity of the light incident on the suspension, the momentum imparted by the light to the medium must induce a movement of the scatterers, which accelerates the attenuation of the CF of the scattered light. Apparently, this effect could not be discovered so far because broad laser beams of comparatively low power were used in the diffusing wave spectroscopy.

Measurements were made in the standard backscattering geometry [1]. A linearly polarised laser beam (wavelength $\lambda = 532$ nm, beam radius $a \approx 1$ mm) was focused by a lens (focal length $f = 15$ cm) on the surface of aqueous suspension (having a volume fraction, i.e., the ratio of the particle volume to the volume of the liquid $\Phi = 0.0183$) of spherical latex particles. The spot size was of the order of $\lambda_0 f/(2a) \approx 40$ µm. The output power of the laser was varied in the interval 0.07 – 2 W. The suspension was held in a glass container of size 8 mm × 20 mm × 5 mm. The particles had the following parameters: radius $r_0 = 90$ nm, mass density and refractive index of latex $\rho = 1.05$ g cm^{-3} and $n = 1.51$ respectively.

The characteristic time of the CF attenuation due to the Brownian motion of particles is $\tau_0 = 0.372 \pm 0.005$ ms. According to the Mie theory of scattering, the transport mean free path l^* of photons is ~ 0.16 mm, which was confirmed by experimental measurements of the function $C_1(t)$ of radiation transmitted by the sample (the scattering layer thickness was 5 mm) for an incident beam power $P = 0.01$ W without focusing.

A single-mode optical fibre of core diameter 3 µm was used to collect the backscattered light. To reduce the contribution of lower-order scattering, a polariser was used for separating from the scattered light only the part having a polarisation perpendicular to the polarisation of the incident beam (VH configuration). After passing through the fibre, light was incident on a photoelectric multiplier working in the photon-counting regime. After this the signal was processed by a multichannel correlator. The quantity to be measured was the normalised autocorrelation function of the scattered light intensity $g_2(t) = \langle I(0)I(t)\rangle / \langle I\rangle^2$, related to $C_1(t)$ by the expression $g_2(t) = 1 + \beta|C_1(t)|^2$, where $0 < \beta \leqslant 1$ is a factor inversely proportional to the number of speckles falling on the detector window. The detector was placed at a distance of 15 cm from the sample surface so that light from only one speckle-pattern spot of scattered light fell on the fibre. The coefficient β was equal to unity.

Figure 1 shows the results of measurement of the CF $C_1(t)$. For low incident beam powers, the CF is described by standard expression (1). In this case, the motion of particles is Brownian. For $P \gtrsim 0.1$ W, the form of the CF changes noticeably. Apart from the Brownian motion, the ordered motion of particles under the action of high-power laser radiation also becomes significant. As the power of the incident laser beam increases, the CF attenuates more rapidly.

A.V. Kondrat'ev Laboratoire de Physique des Matériaux Divisés et des Interfaces, UMR8108 du CNRS, Université de Marne-La-Vallée, F-77454 CNRS, Marne-La-Vallée Cédex 2, France
e-mail: ollandr@yandex.ru

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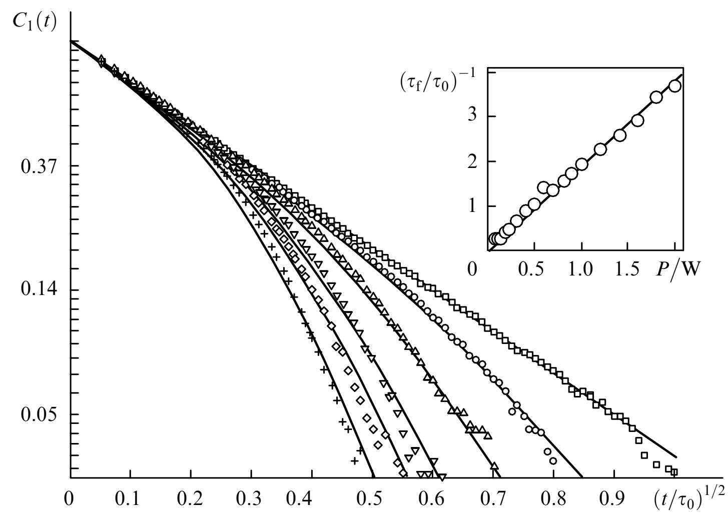

Figure 1. Normalised CF $C_1(t)$. The symbols correspond to the experimental points and the curves are the results of calculations using expression (3) for an incident beam power $P = 0.07$ (squares), 0.1 (circles), 0.4 (triangles), 0.8 (inverted triangles), 1.2 (diamonds), and 1.6 W (+). The inset shows the normalised time τ_f characterising the light-induced motion of particles as a function of the incident beam power.

The calculation of the CF $C_1(t)$ taking into account the light-induced motion of the scatterers requires the solution of a hydrodynamic problem on the emergence of velocity field in the medium [3]. It is quite difficult to obtain an analytic solution of this problem taking into account the boundary conditions for the experimental geometry considered by us. However, an approximate expression for $C_1(t)$ can be derived as follows. The random motion of particles in the medium is a result of the superposition of the Brownian motion and the light-induced ordered motion. Let us assume that the root-mean-square displacement $\langle \Delta r^2(t) \rangle$ of the scatterers can be expressed in terms of a certain effective velocity $v_f(P)$ characterising the light-induced motion averaged over the entire sample:

$$\langle \Delta r^2(t) \rangle = 6Dt + v_f^2(P)t^2. \tag{2}$$

The velocity v_f determines the characteristic time $\tau_f = \lambda/v_f$ of displacement of scatterers over a distance of the order of wavelength of the light due to their forced motion. Calculating the CF $C_1(t)$ using the standard technique [1], we obtain the following expression for the backscattered signal:

$$C_1(t, P) \approx \exp\left\{ -\gamma \left[\frac{t}{\tau_0} + \left(\frac{t}{\tau_f(P)} \right)^2 \right]^{1/2} \right\}. \tag{3}$$

Such a computational procedure implies that the contribution to the correlation function from lower orders of scattering, where the inhomogeneity of the velocity fields emerging in the medium is quite significant, is small. In experiments, this condition is satisfied when the VH configuration is used for detection. It can be shown from dimensional analysis [3] that, irrespective of the geometry of the problem, the characteristic time τ_f is determined by the relation $\tau_f \sim \eta \lambda l^* c/P$ accurate to a numerical factor. The solid curves in Fig. 1 show the results of approximation of the experimentally measured CFs (3). The coefficient $\gamma = 3.28 \pm 0.07$ was determined from (1) for a CF measured for the beam power $P = 0.07$ W, and was then assumed to be constant. The values of τ_f obtained from (3) for various beam powers correspond to the predicted dependence $\tau_f \sim P^{-1}$. This fact indicates that the change in CF is caused by the light-induced motion of scattering particles. The ratio τ_f/τ_0 of the times determining the contributions of Brownian and light-induced motions becomes equal to unity for the incident beam power $P = 0.5$ W. In this case, the mean intensity in the laser spot is $\sim 10^4$ W cm^{-2}. An estimate of the acceleration of particles caused by the light pressure is $\sim 10^5$ cm s^{-2} (for a value of reflectivity equal to unity). Probably, the light pressure is one of the main reasons behind the emergence of the particle movement, although other mechanisms cannot be ruled out either [4, 5].

The existence of multiple scattering by a light-induced jet of microparticles and the simplicity of its observation raise hopes for the development of the 'force' diffusive wave spectroscopy proposed by the author of Ref. [6]. Measurement of the dependence $\tau_f(P)$ leads to additional information on the scattering medium without changing the sample. For example, the viscosity of the medium or the photon transport mean free path can also be determined in addition to the diffusion coefficient of the particles (as in the standard version of the diffusing wave spectroscopy). Naturally, this requires a well-substantiated dependence of the CF $C_1(t, P)$ on the incident beam power taking into account the geometry of the experiment.

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