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LIGHT-INDUCED DYNAMIC BACKSCATTERING OF LASER PULSES IN A RANDOMLY INHOMOGENEOUS MEDIA

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Abstract

Backscattering of high-power laser pulses in a randomly inhomogeneous media containing microparticles suspended in a liquid is treated theoretically. Relationships for the temporal correlation function of multiply scattered radiation, which account for acceleration of particles in the field of optical radiation, are derived for the first time. A new method for determination of particle velocities based on correlation properties of scattered light is proposed.

Correlation properties of radiation under multiple-scattering conditions have been studied intensively during the last decade [1]. A large number of experiments have been carried out using particles of micrometer and submicrometer size (polystyrene spheres) suspended in liquid (water) [2, 3], under conditions of laminar flow of scatterers [4] and also when the dynamics of the light-scattering particles is nonuniform in the bulk of the medium [5, 6]. However, results available at present relate only to the case where the radiation is of rather low intensity, which makes it possible to perform theoretical analysis neglecting the effect of scattered light on the dynamics of scatterers in the medium. At the same time, it is well known that a laser pulse incident on a randomly inhomogeneous medium modifies the motion of particles in the medium if the laser-pulse power is sufficiently high. Several mechanisms of such modification exist [7–9]. In this connection, it is of interest to analyze correlation characteristics of scattered light with allowance for the light-induced motion of scatterers in the medium against the background of the scatterers' random walk.

In this study, we use a diffusion approximation to calculate the autocorrelation function $C_1(\tau) = \langle E(t)E^*(t+\tau)\rangle$ for the light backscattered from a semi-infinite, randomly inhomogeneous medium (with volume density of the light-scattering particles in excess of 1 %) with allowance for the particle motion induced by the focused-radiation laser pulse. It is demonstrated below that the function $C_1(\tau)$ depends on the power of radiation incident on the medium. Furthermore, a new method of measuring the characteristic velocities of microparticles in the experiments with laser-induced acceleration of these particles is proposed.

Let a high-power focused-radiation laser pulse (with wavelength in the medium λ, peak-power density I_0, duration τ_p, neck size d, and repetition rate f) be incident on the boundary of a semi-infinite medium that occupies the half-space $z > 0$ and consists of a suspension of microparticles (of sizes $a \gtrsim \lambda$) in a liquid. We assume that τ_p is much smaller than the characteristic lifetime of a photon in the medium, which is about 10^{-11} s [10]. Let the origin of coordinates be coincident with the point of incidence of the pulse onto the boundary of the medium and let us characterize the medium by the transport mean free-path length $l^* \gg \lambda$ of a photon and by the diffusion coefficient D of the particles [11]. The correlation function of backscattered radiation in the case where the medium does not absorb the radiation can be written [2] as

$$C_1(\tau) = \sum_{n=1}^{\infty}\langle E(t)E^*(t+\tau)\rangle_n = \sum_{n=1}^{\infty} I(n)\exp\left\{-\frac{1}{2}\langle\Delta\varphi^2(\tau)\rangle_n\right\}, \tag{1}$$

where the nth term of the sum represents the contribution of the nth-order scattering processes; $\langle\ldots\rangle_n$ signifies averaging over the above processes and also over all kinds of microscopic configurations of scatterers in the

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medium; $I(n)$ is the intensity related to the nth order scattering processes; and $\Delta\varphi(\tau)$ is the phase difference for two photons sequentially scattered by the same particles at instants separated by the time interval τ. The quantity $\Delta\varphi(\tau)$ can be represented as the sum of contributions stemming from (i) chaotic (Brownian) motion of particles and (ii) light-induced motion of the same particles. The first of these contributions is given [2] by

$$\langle \Delta\varphi_B^2 \rangle_n = \frac{\tau}{\tau_0} n, \tag{2}$$

where $\tau_0 = (4k^2 D)^{-1}$ and $k = 2\pi/\lambda$. In order to evaluate the second contribution, we assume that the laser pulse induces the motion of the particles in the direction of the z axis within a narrow cylinder of diameter $d < l^*$ with the root-mean-square velocity of this motion being $V \ll c$. Such an assumption is realistic when the pulse-repetition rate f is high and the laser radiation is sharply focused; in this case, the particles are accelerated under the effect of the incident-light pressure and are kept at the laser-beam axis by gradient forces [8]. The contribution to $\langle \varphi^2(\tau) \rangle_n$, related to light-induced motion of the particles, is then determined as

$$\langle \Delta\varphi_V^2(\tau) \rangle_n = \frac{1}{2} k^2 V^2 \tau^2 P_n, \tag{3}$$

where P_n is the average number of photon-scattering events in the zone of light-induced motion of particles when the photon was involved in a total of n scattering events and left the medium at the point $(0,0,0)$ of pulse incidence. In order to determine P_n, we assume that the conditions for applicability of the diffusion approximation to the transport equation are met in the medium [11]. Much the same as in [12], the quantity P_n is then expressed in terms of the Green function determined to a diffusion approximation [13] for the problem under consideration. In the limit of large n, we arrive at

$$P_n \approx \frac{3d^2}{8l^{*2}} \left[\frac{n+1}{2n} + \ln n \right]. \tag{4}$$

With allowance for the expression for $I(n)$ derived to the diffusion approximation [2, 3], formula (4) yields

$$C_1(\tau) \propto \sum_{n=1}^{\infty} \frac{1}{n^{5/2}} \exp\left\{ -\frac{\tau}{2\tau_0} n - \frac{\tau^2}{2\tau_V^2} \left[\frac{n+1}{2n} + \ln n \right] \right\}, \tag{5}$$

where $\tau_V = 4l^*/(\sqrt{3}kVd)$ and an insignificant numerical multiplier is omitted. It is noteworthy that typical values of τ_0 for suspensions of latex spheres of micrometer-scale sizes are on the order of $10^{-4} - 10^{-3}$ s [1] for $\lambda \sim 500$ nm, whereas $\tau_V \sim 10^{-5}$ s already for $V = 1$ cm/s. Expression (5) was derived with absorption of light in the medium neglected; however, the absorption can be easily taken into account if we introduce the substitution $\tau/\tau_0 \to \tau/\tau_0 + l^*/(2l_a)$, where l_a is absorption path. Thus, the role of absorption consists in suppressing the contribution of the scattering processes of high orders.

As is evident from formula (5), the contribution of Brownian motion to decorrelation of n-fold scattered photons increases in proportion to n, whereas the contribution of light-induced motion increases much more slowly (as $\ln n$ for large n). This is related to the fact that all particles of the medium are involved in the Brownian motion, whereas only a fraction of the particles are involved in light-induced motion. Nevertheless, the temporal correlation function is found to depend on the velocity of light-induced motion; this is illustrated in Fig. 1 where the function $C_1(\tau)$ is plotted for several velocities V of light-induced motion. As evident from Fig. 1, the characteristic decay time for $C_1(\tau)$ is directly related to V and hence to the peak-power density I_0 of laser pulse incident on the medium because $V \propto I_0$ [8]. In view of this, measuring $C_1(\tau)$ we can uniquely determine the velocity V of light-induced motion of particles of the medium. It is evident from Fig. 1 that the proposed method for measuring the velocity is bound to be sensitive to variation of V within a very wide

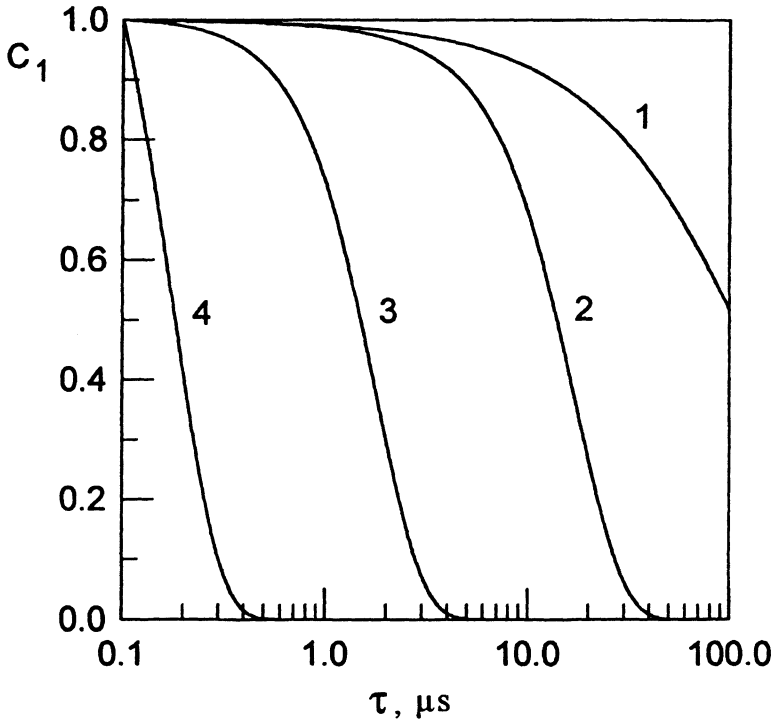

Fig. 1. Temporal autocorrelation function $C_1(\tau)$ for backscattered radiation (normalized so that $C_1 = 1$ for $\tau = 0.1$ s) in the absence of light-induced motion of particles ($V = 0$) (1), for $V = 0.01$ m/s (2), $V = 0.1$ m/s (3), and $V = 1$ m/s (4). The following values of parameters were used in calculations: $\tau_0 = 10^{-4}$ s, $\lambda = 387$ nm, and $d = l^*$.

range. In particular, we can foresee no basic restrictions on the use of this method also in the case where the particles are accelerated to supersonic velocities (in [9], velocities of particles up to $\sim 10^4$ m/s were reported).

It is worth noting in conclusion that the proposed method for measuring the velocity of light-induced motion of micrometer-size particles in dense media exposed to the field of high-power laser pulses seems to be, as of now, the only technique that does not affect the particle motion. This method can be widely used not only in studies of laser-induced acceleration of particles under laboratory conditions, but also in various systems for particle-mixture separation whenever laser-induced acceleration of particles is employed.

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