Diffusion-wave spectroscopy of light-induced fluxes

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Dynamic multiple scattering of laser radiation in a concentrated suspension of micron or submicron particles is considered with the allowance for particle acceleration in the field of (*a*) other (accelerating) laser beam and (*b*) probing radiation. It is shown that in both of the cases the methods of optical correlation spectroscopy allow estimation of the velocity of light-induced particle motion to be made by analyzing time autocorrelation function of the multiply scattered radiation. The method is proposed to measure the characteristic velocities of light-induced particle flows based on the principles of diffusion-wave spectroscopy.

Introduction

It is known that if laser radiation of a sufficiently high power is incident on a suspension of microparticles, the particles may be accelerated in the field of electromagnetic radiation. As a result, a complex field of particle velocities is formed in the medium. This phenomenon was many times observed in the experiments (see, for example, Refs. 1–6).

In this paper, we show that light-induced fluxes of particles in dense suspensions can be studied by use of the diffusion-wave spectroscopy (DWS) technique. This approach is now widely used to study the dynamics of turbid media (such as colloidal suspensions, gels, foams, emulsions, biological media, etc.; see Refs. 7–14 and reviews in Refs. 15–17), in which multiple scattering of light is quite an ordinary event. Here we present only the key items of the calculation scheme and discuss most interesting results. A more detailed consideration can be found in Ref. 18 and in our papers 19–29.

1. Effects of microparticles acceleration by laser radiation

Consider, for definiteness, a spherical particle of the radius a suspended in a liquid. Under the exposure to laser radiation, the particle is affected by the following forces:
- light pressure $\mathbf{F}_p$,
- gradient force $\mathbf{F}_\nabla$,
- convection entrainment,
- radiometric pressure,
- light-reactive pressure.

If the particle size is less than the wavelength of laser radiation, λ, the equations for the force due to light pressure and the gradient force have the following form:

$$\mathbf{F}_p \blacktriangleright \frac{2}{3}\,\alpha^2\, k^4\, E^2\, \hat{n}, \qquad (1)$$

$$\mathbf{F}_\nabla \blacktriangleright \alpha\nabla\,\langle E^2\rangle, \qquad (2)$$

where α is the particle polarizability; E is the strength of the electric field in the light wave; $k = 2\pi/\lambda$, $\hat{n}$ is the unit vector parallel to the direction of the light wave propagation. For a dielectric particle with the dielectric constant ε_0 suspended in the medium with the dielectric constant ε, we have $\alpha \blacktriangleright a^3\,(\varepsilon - \varepsilon_0)/(\varepsilon + 2\varepsilon_0)$.

In contrast to the forces described by Eqs. (1) and (2) and caused by the electromagnetic effect of laser radiation, the effects of convection entrainment and radiometric and light-reactive pressure are connected with light absorption by the medium. Convection entrainment arises at rather high absorption coefficient of the suspension. Because the laser beam heats a suspension inhomogeneously, liquid masses (being in the field of gravity) start to mix inside a sample and thus entrain suspended particles into this motion. The radiometric pressure arises because of radiation absorption by the particulate matter and inevitable heating of the particles. Since laser radiation is usually incident on the medium from a certain side, the temperature of one side is higher than that on the opposite side, and this gives rise to the radiometric force. Finally, the so-called light-reactive pressure is caused by evaporation of the particulate matter that produces the reactive force affecting the particulate frame.

There exist also other mechanisms of acceleration of microparticles suspended in liquid or gas by laser radiation (for example, laser radiation can initiate an acoustic shock wave in the medium, and then this wave affects the suspended particles[3]). We restrict our consideration to the case under the assumption that optical characteristics of the medium and the parameters of particles that scatter light only insignificantly change under the effect of laser radiation.

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2. Diagnostics of light-induced motion of particles in dense suspensions by DWS methods

Let us consider a cell filled with a concentrated suspension of micron and submicron particles in a fluid (Fig. 1).

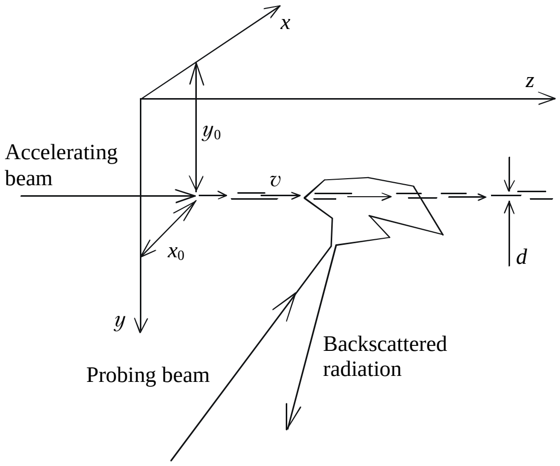

Fig. 1. Optical arrangement of the experiment.

A sufficiently high-power laser beam (wavelength λ_0, the radiation may be both continuous-wave and pulsed) focused onto the left wall of the cell accelerates the suspended particles along the axis z, as is shown in Fig. 1. The probing beam is focused onto the front wall of the cell. Multiply scattered radiation leaving the cell is recorded with a detector. Let any dimension of the cell far exceeds the mean photon free path in the medium ℓ^*.

We assume that the particles accelerated by laser radiation move within a narrow cylindrical zone $d < \ell^*$ in diameter, whose position is described by the coordinates x_0 and y_0. It was just this scenario of particle acceleration that was observed experimentally in Ref. 2. To simplify the analysis, we assume that the velocity v of particles is independent of their position inside the zone of light-induced motion.

We propose that the diagnostics of the light-induced flux of particles be performed with the other laser beam (wavelength λ, radiation is continuous-wave or pulsed with the pulse duration τ_p being much longer than the photon survival time in the medium $(\mu_a c)^{-1}$, where μ_a is the medium absorption coefficient and c is the speed of light) focused onto the front wall of the cell. The radius vector $\mathbf{r}_s = (x_s, y_s, 0)$ determines the point of incidence of the beam onto the cell wall. Unlike the first beam accelerating the particles, the second beam is called probing. Its power is assumed rather low, therefore its effect on the suspended particles can be neglected. Let $\mathbf{r} = (x_s, y_s, 0)$ is a point at a medium boundary, at which the time autocorrelation function $G_1(\tau) = \langle E(t)\, E^*(t + \tau)\rangle$ of the scattered probing radiation is measured. We describe the probing radiation in the scalar approximation, since it is thought to be significantly depolarized. Without loss of generality, consider first the particles, whose size is much smaller than the wavelength of the probing radiation λ. In this case, the particles can be considered approximately as point-like scatterers. Besides, we assume that the absorption coefficient of the suspension at the wavelength λ of the probing radiation is negligibly small. Then, to describe the multiple scattering of light in the suspension, we should know only the photon mean free path ℓ, which is assumed not only much longer than d (this assumption corresponds to the model of a narrow flux of particles in the suspension), but also exceeding the linear dimension of the focal spot of the probing beam on the cell wall (this corresponds to strong focusing of the beam).

With the accelerating beam turned off, the suspended particles take part in random Brownian motion characterized by the diffusion coefficient D_B depending on their size and suspension viscosity and temperature.[30] Turning on the accelerating beam gives rise to a directed flux of particles superimposed on the Brownian wandering. In this case, to calculate the time correlation function of the scattered radiation, we can use the equation[18]

$$G_1(\tau) = \sum_{n=1}^{\infty} I(\mathbf{r}_s, \mathbf{r}, n) \exp\left\{-\frac{1}{2}\langle\Delta\varphi_n^2(\tau)\rangle\right\}, \qquad (3)$$

where $\langle\Delta\varphi_n^2(\tau)\rangle$ should be calculated with the allowance for the specific geometry of the flux of scatterers. Assuming that the Brownian and directed motions of particles are independent, we have

$$\langle\Delta\varphi_n^2(\tau)\rangle = \langle\Delta\varphi_n^{(B)2}(\tau)\rangle + \langle\Delta\varphi_n^{(F)2}(\tau)\rangle, \qquad (4)$$

where the first term describes the effects of decorrelation due to the Brownian motion of scatterers, and the second term describes the effects of decorrelation caused by the light-induced motion. As was shown in Refs. 7–10,

$$\langle\Delta\varphi_n^{(B)2}(\tau)\rangle = \frac{\tau}{\tau_0} n, \qquad (5)$$

where $\tau_0 = (4k^2 D_B)^{-1}$ and $k = 2\pi/\lambda$.

The equation for the second term in Eq. (4) can be found by using the method of integrals over trajectories.[18] Since the cross size of the flux zone d is assumed small ($d < \ell$), then inside this zone we can use the single scattering approximation: every photon, whose trajectory crosses the flux zone, takes part in exactly one event of scattering on a particle accelerated by laser radiation. Certainly, some photons may cross the flux zone meeting no one particle, while others experience two scattering events, but the probability of such events is low and, moreover, we can expect that their effects on $G_1(\tau)$ compensate for each other.

Consider the photon trajectory including n scattering events with the mth event at an accelerated particle moving with the velocity $\mathbf{v}$ and other events at the resting scatterers. The phase difference between two photons scattered along the same trajectory at the time moments separated by the period τ is caused by the displacement of the moving scatterer for this period and, as can be readily seen, it is equal to

$$\Delta\varphi_{mn}^{(F)}(\tau) = k\,\tau(\hat{\mathbf{e}}_m\cdot\mathbf{v})\,\delta(x_m - x_0)\,\delta(y_m - y_0), \quad (6)$$

where $\hat{\mathbf{e}}_m$ is the unit vector setting the direction of the scattered wave after the mth scattering event in the chain of n events ($m = 1, \ldots, n$), $\{x_m, y_m, z_m\}$ are the coordinates of the point, at which the scattering event occurs, and we assume that the particle flux is parallel to the axis z, and its position is determined by the coordinates x_0 and y_0.

To find $\langle\Delta\varphi_n^{(F)2}(\tau)\rangle$, we should sum Eq. (6) over $m = 1, \ldots, n$, square the sum, and average the result over all possible photon trajectories in the medium. Since the vectors $\hat{\mathbf{e}}_m$ and $\mathbf{v}$ are independent, $\langle(\hat{\mathbf{e}}_m\cdot\mathbf{v})^2\rangle = v^2/2$, and finally we have

$$\langle\Delta\varphi_n^{(F)2}(\tau)\rangle = \frac{1}{2}\,(kv\tau)^2\sum_{m=1}^{n}\langle\delta(x_m - x_0)\,\delta(y_m - y_0)\rangle. \quad (7)$$

It can easily be understood that the sum in the right-hand side of this equation is the unnormalized probability $P_n(x_0, y_0)$ that the photon scattered n times experiences one scattering event inside the zone of the light-induced flux, and the mth term of this sum is the unnormalized probability $P_{mn}(x_0, y_0)$ that this scattering event has the number m. These probabilities are obviously related as

$$P_n(x_0, y_0) = \sum_{m=1}^{n} P_{mn}(x_0, y_0). \quad (8)$$

In its turn, $P_{mn}(x_0, y_0)$ can be presented as

$$P_{mn}(x_0, y_0) = \int_{-\infty}^{\infty} P_{mn}(x_0, y_0, z)\,\mathrm{d}z, \quad (9)$$

where $P_{mn}(x_0, y_0, z)\,\mathrm{d}z$ is the probability that the mth scattering event takes place in the vicinity of the point (x_0, y_0, z), and the integration over z reflects the fact that the velocity of the light-induced flux $\mathbf{v}$ is directed along the z axis.

In view of the above-said, Eq. (7) can be written in the following form:

$$\langle\Delta\varphi_n^{(F)2}(\tau)\rangle = 2\left[\frac{\tau}{\tau_F}\right]^2 P_n(x_0, y_0), \quad (10)$$

where $\tau_F = 2(kv)^{-1}$ is the characteristic time connected with the presence of the light-induced flow of scatterers in the medium.

In calculating $P_{mn}(x_0, y_0, z)$, $P_{mn}(x_0, y_0)$, and $P_n(x_0, y_0)$, we should take into consideration only those possible photon trajectories, which start from the radiation source (point $\mathbf{r}_s$), finish at the detector (point $\mathbf{r}$), and on their way between these points they do not cross the medium boundary. Note that $P_n(x_0, y_0)$ can be also considered as the mean number of scattering events inside the zone of the light-induced flux. Calculation of $P_{mn}(x_0, y_0, z)$ gives[18]

$$P_{mn}(x_0, y_0, z) = \frac{I(\mathbf{r}_s, \{x_0, y_0, z\}, m)\; I(\{x_0, y_0, z\}, \mathbf{r}, n - m)}{I(\mathbf{r}_s, \mathbf{r}, n)}, \quad (11)$$

where to find $I(\mathbf{r}_s, \mathbf{r}, n)$, we use the diffusion approximation, assuming the medium to be semi-infinite[18]

$$I(\mathbf{r}_s, \mathbf{r}, n) = \left(\frac{3}{4\pi\ell^2 n}\right)^{3/2}\times \left[\exp\left\{-\frac{3(x - x_s)^2}{4\ell^2 n}\right\} - \exp\left\{-\frac{3(x + x_s)^2}{4\ell^2 n}\right\}\right]\times \exp\left\{-\frac{3((y - y_s)^2 + (z - z_s)^2)}{4\ell^2 n}\right\}. \quad (12)$$

Then, upon substitution of Eq. (12) into Eq. (11) and making integration in Eq. (9), we obtain the following result:

$$P_{mn}(x_0, y_0) = \frac{9x_0^2\, n^2}{4\pi\ell^2\, m^2\,(n - m)^2}\times \exp\left\{-\frac{3x_0^2\, n}{4\ell^2\, m\,(n - m)}\right\}\times \exp\left\{-\frac{3(y_0 - y_s)^2}{4\ell^2\, m} - \frac{3(y_0 - y)^2}{4\ell^2\,(n - m)} + \frac{3(y - y_s)^2}{4\ell^2\, n}\right\}. \quad (13)$$

Note that the transition to the case of scatterers of a finite size (about the probing radiation wavelength λ) can be made by replacing the photon mean free path ℓ by the transport mean free path ℓ^* in all the above equations. But, now the role of a single scattering event in the above reasoning comes to a more complex event, namely, several successive scattering events, as a result of which the direction of propagation of scattered radiation becomes random (the number of single scattering events needed for this to occur is obviously equal to ℓ^*/ℓ). In this case, the considered theoretical model is correct only in the case of $\ell^* > d$.

Assume, for simplicity, that $y = y_s = 0$, i.e., the probing beam is focused onto the origin of coordinates on the front wall of the cell and the time autocorrelation function of the diffusely scattered light is measured at the same point. Besides, let $x_s = \ell^*$ assuming that the laser beam incident on the medium leads to formation of the isotropic source of radiation at the distance ℓ^* from the medium boundary.

Consider now the dependence $G_1(\tau)$ at different velocities v of the light-induced flux. Figure 2 shows the normalized time autocorrelation functions $g_1(\tau) = G_1(\tau)/G_1(0)$ calculated for $v = 0$, 1, 10, and 100 m/s. To derive them, equations (3)–(5), (8), (10), (12), and (13) were used at $\tau_0 = 10^{-4}$ s; this value is typical of water suspensions of polystyrene submicron spheres under normal conditions. To get an idea on the value of the characteristic times τ_F connected with the directed flux of scatterers, assume $\lambda = 0.5$ μm. Then τ_F varies from 16 μs at $v = 10^{-2}$ m/s to 1.6 ns at $v = 100$ m/s.

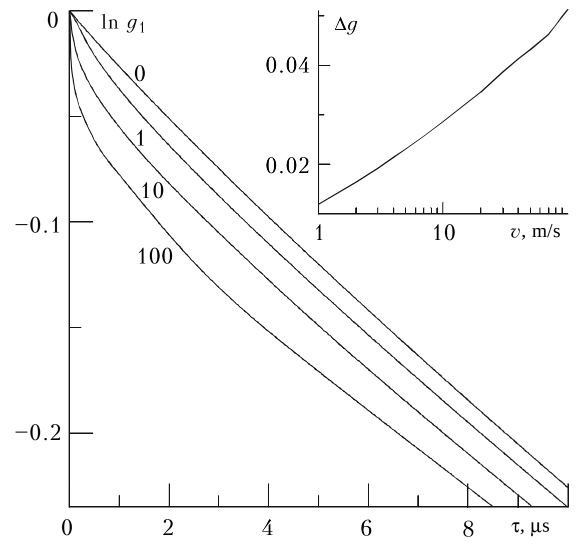

Fig. 2. Normalized time autocorrelation function of diffusely reflected radiation ($x_0 = 5\ell^*$, $y_0 = 0$, $\tau_0 = 10^{-4}$ s).

At $v = 0$ (absence of the flux) $g_1(\tau)$ decreases with the increasing τ only because of the Brownian motion of the suspended particles. In the presence of the flux ($v > 0$), the decrease of $g_1(\tau)$ becomes faster. As is seen from Fig. 2, the higher v, the faster $g_1(\tau)$ decreases. This means that measurements of $g_1(\tau)$ can be used for diagnostics of the light-induced fluxes in randomly inhomogeneous media (for example, to determine the velocities of such flows).

It should be noted that the main contribution to the decrease of the correlation function is due to the Brownian motion, whose intensity is the same for all curves in Fig. 2, rather than due to the scatterer flux. The cause of this is that the Brownian motion involves all particles, whereas the directed light-induced motion involves only a small fraction of the particles. Therefore, the directed motion affects only a part of scattered photons, whereas the Brownian motion affects all the scattered photons. The rate of decrease of $g_1(\tau)$ turns out to be sensitive to the velocity v of the flux at $\tau < \tau_F$ and even at $\tau \sim \tau_F$. At larger τ, as is seen from Fig. 2, the rate of decrease of $g_1(\tau)$ is largely determined by the Brownian motion of the scatterers.

The above analysis suggests at least two different methods for determination of the characteristic velocity of the light-induced flux from the measured dependences $g_1(\tau)$. First, at small τ, as is seen from Fig. 2, the derivative $\mathrm{d}g_1/\mathrm{d}\tau$ increases with the growth of v. Therefore, analyzing the behavior of $g_1(\tau)$ at small τ, we can determine the velocity of the flux. However, this method of measuring v may prove to be hard-to-implement at high v. Another method of determining v is to measure the maximum difference Δg of the autocorrelation function $g_1(\tau)$ in the presence of the flux from its value in the absence of the flux. This difference is shown in Fig. 2 as a function of v. One can see that Δg is almost a linear function of ln v.

3. Ponderomotive action of light in DWS problems

Let the half-space $z > 0$ be filled with the concentrated suspension of microparticles in a fluid (particle volume density $0.01 < \Phi < 0.1$). The medium is characterized by the photon mean free path ℓ and the transport mean free path ℓ^*, the absorption coefficient μ_a, and the dynamic viscosity[30]

$$\eta = \eta_0 \left(1 + \frac{5}{2}\Phi\right), \tag{14}$$

where η_0 is the viscosity of the fluid. In the absence of external forces, the particles of the medium are considered resting, i.e., their Brownian motion is neglected. Let the medium be exposed to a laser pulse *1* (wavelength λ in the fluid, pulse duration τ_p, peak intensity I_0, $\lambda < d < \ell^*$, direction along the z axis) focused onto the zone with the cross size d (Fig. 3).

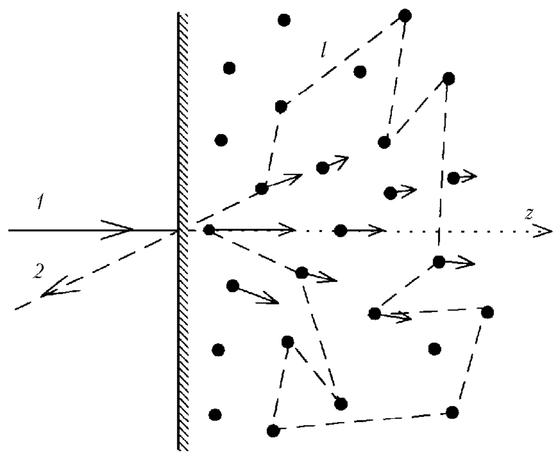

Fig. 3. Optical arrangement of the experiment.

This pulse gives rise to the light-induced flux. As earlier, we believe that τ_p is much longer than the photon survival time in the medium.

If I_0 is low enough, the pulse experiences multiple scattering on the particles of the medium, and a part of its energy is absorbed (causing insignificant heating of the medium), while the other part leaves the medium. As this takes place, the time coherence of the scattered radiation does not worsen as compared to the beam incident on the medium (here we neglect the thermal motion of the particles).

If I_0 is sufficient to give rise to the particle motion in the medium, then the radiation is scattered on moving particles, rather that resting ones. This leads to worsening of the time coherence of the scattered light field. The mechanism of acceleration of the particles in the field of a laser pulse can be different (see Section 1), and at this stage of analysis it makes no sense to concretize it. We assume, however, that the mass, shape, volume, and other parameters of microparticles do not change significantly under the effect of laser radiation on the time scales about the photon survival time in the medium.

Determine now the character of particle motion under the effect of laser radiation. Since we consider strong focusing of the beam ($\lambda < d < \ell^*$), then the point of the beam incidence on the medium can be approximately considered as a point-like source of particles outgoing in the direction of the axis z. Due to hydrodynamic interaction, these particles entrain neighboring ones, and this leads to formation of the complex field $\mathbf{v}(\mathbf{r})$ of particles' velocities in the medium (formation of a jet). We believe that the presence of the medium boundary at $z = 0$ has only insignificant effect on the field $\mathbf{v}(\mathbf{r})$ in the regions far from the boundary (at $z > \ell^*$). To find $\mathbf{v}(\mathbf{r})$, consider the equation of motion of a viscous suspension filling the entire space in the approximation of the suspension incompressibility[30]

$$\frac{\partial}{\partial t} \text{rot } \mathbf{v} + (\mathbf{v} \cdot \nabla) \text{ rot } \mathbf{v} - (\text{rot } \mathbf{v} \cdot \nabla)\, \mathbf{v} = \nu\, \Delta \text{rot } \mathbf{v}. \quad (15)$$

Here $\nu = \eta/\rho$ is the kinematic viscosity of the suspension; ρ is its density. This equation can be solved analytically only in a few cases. However, in the above situation of a thin jet of microparticles from the point {0, 0, 0} in the direction of the axis z (problem of submerged jet, see Ref. 30), the solution can be found. In the spherical coordinate system and not very high velocities of the jet, it has the form[30]

$$v_r = \frac{P}{4\pi\eta}\frac{\cos\theta}{r}, \quad v_\theta = -\frac{P}{8\pi\eta}\frac{\sin\theta}{r}, \quad v_\phi = 0, \quad (16)$$

where P is the total momentum flux in the jet. Under our conditions, it is equal to the momentum given by the laser radiation to the medium particles in a unit time. The flow lines corresponding to Eq. (16) are shown in Fig. 4.

To calculate the time correlation of radiation after multiple scattering in a randomly inhomogeneous semi-infinite medium, in which the field of scatterer velocities has the form (16), let us use the method of integration over trajectories.[18] First, consider the case of point-like scatterers (scatterer size $a \ll \lambda$).

For the velocity field (16), calculation of the stress tensor[30] gives

$$\sigma_{rr} = -\frac{P\cos\theta}{2\pi r^2}, \; \sigma_{\phi\phi} = \sigma_{\theta\theta} = \frac{P\cos\theta}{4\pi r^2},$$
$$\sigma_{r\theta} = \sigma_{r\phi} = \sigma_{\theta\phi} = 0. \quad (17)$$

To find $G_1(\tau)$, we now have to calculate the integral[12,13]

$$\xi(n) = \frac{1}{\eta^2}\int \left(\sum_{i\neq k} \sigma_{ik}^2(\mathbf{r}_1)\right) \rho_n(\mathbf{r}_0, \mathbf{r}_1, \mathbf{r})\, \mathrm{d}^3\mathbf{r}_1, \quad (18)$$

substitute it into the equation[12,13]

$$\langle \Delta\varphi_n^2(\tau)\rangle = \frac{2}{15} k^2 \ell^2 \tau^2 n\xi(n), \quad (19)$$

and then calculate the correlation function by the equation[12,13]

$$G_1(\tau) = \sum_{n=1}^{\infty} I(\mathbf{r}_0, \mathbf{r}, n) \exp\left\{-\frac{1}{2}\langle\Delta\varphi_n^2(\tau)\rangle\right\}, \quad (20)$$

where $I(\mathbf{r}_0, \mathbf{r}, n)$ is the mean intensity of radiation at the point $\mathbf{r}$ generated by the point-like source of coherent radiation situated at the point $\mathbf{r}_0$ and scattered n times.

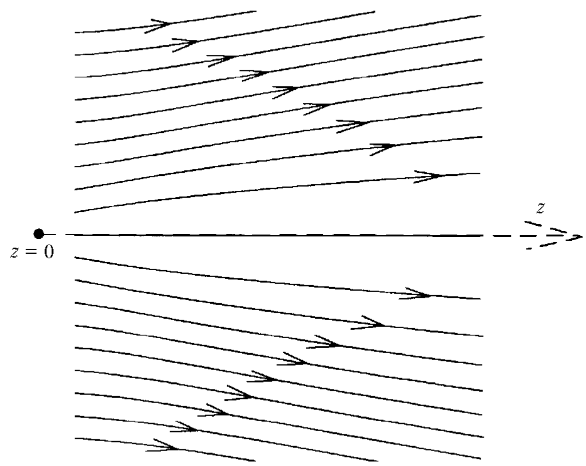

Fig. 4. Pattern of flow lines corresponding to the problem of a submerged jet.[30]

In Eq. (18) the integration is performed over the entire volume of the randomly inhomogeneous medium; $\rho_n(\mathbf{r}_0, \mathbf{r}_1, \mathbf{r})$ is the fraction of photons scattered n times and passing from the source at the point $\mathbf{r}_0$ to the point $\mathbf{r}$ through the point $\mathbf{r}_1$. For definiteness, we believe that the scattered light in the considered case is measured in the immediate proximity from the point of incidence of the laser beam onto the medium boundary. Then we can take approximately that $\mathbf{r} \blacktriangleright \mathbf{r}_1 = \{0, 0, 0\}$ and use the equation for $\rho_n(\mathbf{r}_1) = \rho_n(0, \mathbf{r}_1, 0)$ obtained in Ref. 14 in the diffuse approximation:

$$\rho_n(\mathbf{r}_1) = \frac{3}{2\pi\ell^2\, nr_1} \exp\left\{-\frac{3r_1^2}{\ell^2\, n}\right\}. \quad (21)$$

Substituting Eqs. (17) and (21) into Eq. (18) and extending the integration in Eq. (18) to the area $r_1 > \ell$, in which the light scattering is well described by the diffusion approximation, we find from Eq. (19)

$$\langle\Delta\varphi_n^2(\tau)\rangle = (\tau/\tau_c)^2 f(n), \quad (22)$$

where

$$\tau_c = \sqrt{10}\, \eta\ell\lambda/P \quad (23)$$

is the characteristic coherence time connected with the appearance of the light-induced jet;

$$f(n) = \exp\left\{-\frac{3}{n}\right\} + \frac{3}{n} Ei\left\{-\frac{3}{n}\right\}, \quad (24)$$

$Ei(x)$ is the integral exponential function.[31] The function $f(n)$ determining the role of the processes of different order turns out to be independent of the problem parameters and dependent only on the number of scattering events n. The function $f(n)$ vanishes at $n = 0$ and tends to unity at $n \to \infty$ (Fig. 5). Therefore τ_c is the characteristic coherence time corresponding, strictly speaking, to the infinite number of scattering events. The coherence time longer than τ_c corresponds to scattering processes involving a finite number of scattering events.

Using the equation for the Green's function $I(\mathbf{r}_0, \mathbf{r}, n)$ obtained in the diffusion approximation[13] [see also Ref. (12)] and assuming that the point of incidence of laser radiation onto the medium $\mathbf{r}_0$ and the point $\mathbf{r}$, at which the time correlation function is measured, are separated by the distance on the order of ℓ, we derive the final equation for $G_1(\tau)$:

$$G_1(\tau) \propto \sum_{n=1}^{\infty} \frac{1}{n^{5/2}} \exp\left\{-\frac{1}{2}\langle\Delta\varphi_n^2(\tau)\rangle - \mu_a\, \ell n\right\}, \quad (25)$$

where $\langle\Delta\varphi_n^2(\tau)\rangle$ is given by Eq. (22). As was noted above, the analysis performed above is valid for the case with the scatterer size $a \ll \lambda$. If $a \sim \lambda$ (or $a > \lambda$), the analysis becomes far more complicated. However, the approximate results corresponding to this case can be obtained by substituting ℓ^* for ℓ in all equations.

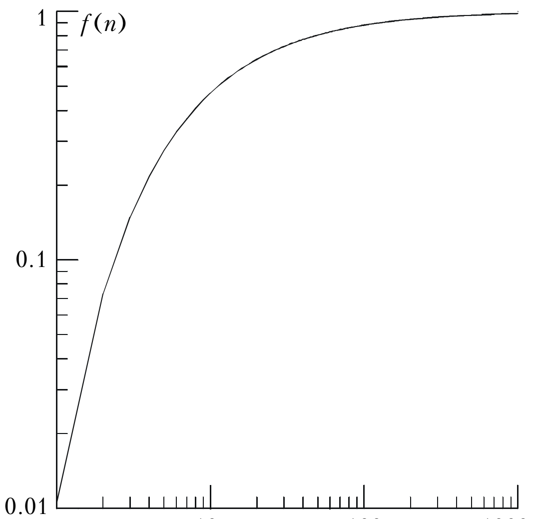

Fig. 5. The factor $f(n)$ determining the dispersion of the phase difference of photons scattered by the same particles of the medium at time moments spaced by τ [see Eq. (24)].

4. Discussion

Figure 6 shows the normalized time autocorrelation functions of the backscattered radiation $G_1(\tau)/G_1(0)$ for different values of the product $\mu_a\ell$ (for definiteness, we believe that ℓ is the same for all the three curves, and the absorption coefficient μ_a is different). Note that the absorption coefficient μ_a has a relatively weak effect on the normalized correlation function of the scattered light. The effect of absorption manifests itself largely through the increase of P [and, consequently, the decrease of τ_c, see Eq. (23)] with the increase of μ_a. The difference between the curves corresponding to different values of the absorption coefficient is caused by the fact that at small μ_a ($\mu_a \ll \ell^{-1}$) a sufficiently large portion of scattered radiation is due to high-order scattering processes, which contribute significantly to decorrelation of radiation. At a relatively large μ_a ($\mu_a \sim \ell^{-1}$), as is seen from Eq. (25), negligibly low intensities correspond to large n (because of the presence of the term $-\mu_a\, \ell n$ in the exponent); therefore, the main part of the scattered radiation is caused by low-order scattering processes. Although this case is not fully correctly described in the diffuse approximation we use, Eq. (25) gives the physically clear result as before: the correlation function now decreases not so fast as at small μ_a, because the photons scattered less times are decorrelated more weakly. Note that at $\tau/\tau_c \gg 1$ the curves corresponding to different μ_a become parallel. This is connected with the fact that at large τ the time autocorrelation function is affected most strongly by the photons scattered several times and, as a result, almost insensitive to low absorption in the medium.

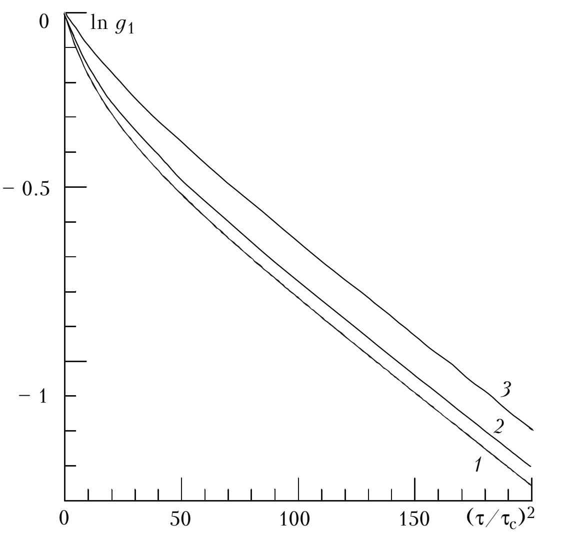

Fig. 6. Logarithm of the normalized time autocorrelation function $g_1(\tau) = G_1(\tau)/G_1(0)$ vs. the squared τ/τ_c ratio for three different values of the absorption coefficient μ_a at a fixed momentum P transferred from the electromagnetic field to particles of the medium in unit time: $\mu_a = 0$ (*1*), $\mu_a = 0.1\, \ell^{-1}$ (*2*), $\mu_a = 0.5\, \ell^{-1}$ (*3*).

It should be noted that the dependence of $\langle\Delta\varphi_n^2(\tau)\rangle$ on τ has different forms for the Brownian motion of scatterers [$\langle\Delta\varphi_n^{(B)2}(\tau)\rangle \propto \tau^2$] and the light-induced jet [$\langle\Delta\varphi_n^{(F)2}(\tau)\rangle \propto \tau$]. Thus, the effects of laser acceleration qualitatively change the form of the time correlation function of the diffusely reflected radiation. At a relatively low intensity, the Brownian wandering and the light-induced motion of scatterers can be considered independent. Then, the equation for $\langle\Delta\varphi_n^2(\tau)\rangle$ with the allowance made for both types of the motion can be written as a sum of two terms in Eq. (4).

Let us estimate the conditions under which the ponderomotive effect of the light is significant in calculation of the time coherence of the scattered light. To do this, we, obviously, should concretize the main mechanism of acceleration of the suspended particles in the field of laser radiation. For example, for polystyrene submicron balls suspended in water, light pressure plays the main role, because the absorption is low in polystyrene, and all other acceleration mechanisms are connected just with the absorption of light. Therefore $P \approx W/c$, where W is the radiation power, and c is the speed of light in the medium. The second term in Eq. (4) exceeds the first one, if the radiation power is higher than some critical value $W_c(\tau, n)$:

$$W > W_c(\tau, n) \approx 4c\ell \sqrt{\frac{5\pi\eta\, k_B T}{3a} \times \frac{n}{\tau f(n)}}, \quad (26)$$

where k_B is the Boltzmann constant, and T is the temperature of the suspension. In derivation of Eq. (26), the equation for the diffusion coefficient of spherical particles in suspension[30] $D_B = k_B T/(6\pi\eta a)$ was used.

As is seen from Eq. (26), the critical power depends on both the number of the scattering events n and the delay τ. Analysis shows that the ratio $n/f(n)$ is minimum at $n = 5$, therefore the effects of laser acceleration are maximum for the photons scattered several (about five) times. To estimate this, take $a \sim 0.1$ μm and the volume concentration of particles equal to 1%; then $\ell^* \sim 200$ μm. For water $\eta \sim 10^{-3}$ Pa·s; the radiation wavelength λ is taken 0.5 μm. As a result, at $n = 5$ the critical power W_c varies from 15 W at $\tau = 1$ μs to 0.5 W at $\tau = 1$ ms. These critical values of the radiation power are small enough for the effect described in this paper to be observed experimentally.

Conclusion

The results presented in this paper strongly suggest that the technique of the diffusion-wave spectroscopy can be used to study the fluxes of microparticles induced by high-power laser radiation in concentrated suspensions. If the power of a strongly focused laser beam exceeds 1 – 10 W, then the effects of laser acceleration of microparticles can markedly modify the time autocorrelation function of the light reflected diffusely from micron- and submicron particles. We hope that theoretical analysis performed in this work will stimulate experimental research in this field.

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