Multiple dynamic scattering of laser radiation on a light-induced jet of microparticles in suspension

S E Skipetrov, S S Chesnokov, S D Zakharov, M A Kazaryan, N P Korotkov, V A Shcheglov

Abstract. A self-consistent theoretical analysis is made of multiple scattering of coherent laser radiation in a random medium under conditions of formation of a light-induced jet of scatterers. It is shown that the laser particle acceleration leads to a qualitative change in the nature of the temporal auto-correlation function of scattered light, as compared with the case of scattering on chaotically moving Brownian particles. The effect of radiation absorption on the temporal coherence of the multiple-scattered light under conditions of light-induced particle motion in the scattering medium is studied.

1. Introduction

When a powerful laser pulse is incident on a suspension of microparticles, the particles are accelerated in the field of electromagnetic radiation. In the present paper we shall consider dynamic multiple scattering of light on a light-induced jet of microparticles in suspension, i.e. the scattering of laser radiation in a random medium under conditions of particle acceleration by the incident laser beam. A self-consistent analysis of the problem is made having regard to the fact that the particle acceleration is produced by the same laser beam that undergoes multiple scattering in the medium. It will be shown below that the correlation properties of scattered light change radically as the incident beam intensity increases to a value sufficient for a light-induced jet of microparticles to appear in suspension.

Multiple scattering in random media itself attracts the attention of investigators [1]. The angular distribution [2–4], the angular [5], spatial [6], and temporal [7, 8] correlations, as well as the degree of polarisation [4, 9] of the scattered light have become a subject of detailed analysis. Moreover, consideration is being given to the possibility of locating and diagnosing both absorbing [10] and purely dynamic [11, 12] inhomogeneities in random media, based on an analysis of the statistical characteristics of the multiple-scattered light measured at the boundary of a medium.

Additionally, the laser acceleration of microparticles has also been actively studied for years [13,14]. The possibility of creating optical ‘traps’ has been demonstrated, the laser levitation of particles has been realised [15], various mechanisms of microparticle acceleration in the laser field as well as the conditions of their realisation have been examined [16, 17]. However, the studies made so far deal essentially with fairly dilute suspensions of particles. Furthermore, of particular interest is the investigation of the laser particle acceleration in concentrated suspensions. The problem is of great practical importance, since many biological media may be classified as suspensions with high concentrations of suspended particles [18].

The present paper addresses the problem of dynamic multiple scattering of a coherent light wave from a semi-infinite randomly inhomogeneous medium containing particles of size less than or of the order of the radiation wavelength λ. A similar problem has been studied previously assuming the Brownian motion of particles [7, 8], as well as their laminar [19] or turbulent [20] flows. Until now, the problem of multiple scattering with due regard for the particle motion induced by the radiation being scattered has not been given a rigorous theoretical treatment. It is obvious that the laser particle acceleration will play a considerable role only if the power of the incident light is sufficiently high. For example, the motion of particles of micron size with supersonic velocities at a peak intensity \( I \) of a strongly focused laser pulse of the order of \( 10^8 \text{ W cm}^{-2} \) was observed [16]. We stress that here we shall not consider nonlinear optical phenomena, which may occur at high values of \( I \).

The first attempt to analyse the problem, similar to the one considered in the present article, was undertaken in Ref. [21]. However, the adopted model of microparticle motion in a laser field was too crude and yielded only qualitatively correct results. In the present paper, we shall give a self-consistent analysis of the multiple scattering of a powerful laser pulse in a randomly inhomogeneous medium using a physically justified model of the particle motion in the medium. The model is based on a solution of the hydrodynamic equations for a suspension of microparticles.

2. Formulation of the problem and estimates of the characteristic parameters

Suppose that the half-space \( z > 0 \) is filled with a concentrated suspension of microparticles in a liquid (the particle volume density is \( 0.01 < \phi < 0.1 \)). We shall characterise this medium by the photon mean free path \( l \), the photon transport mean free path \( l_{\text{tr}} \) [22], the absorption coefficient \( \mu_a \ll l_{\text{tr}}^{-1} \), and the dynamic viscosity (see Ref. [23]),

\[
\eta = \eta_0 \left( 1 + \frac{5}{2} \phi \right),
\]

(1)
where \( \eta_0 \) is the viscosity of the liquid. In the absence of external disturbances, the particles in the medium are said to be at rest, i.e., we shall neglect their Brownian motion.

A laser pulse (wavelength of light in the liquid \( \lambda \), pulse duration \( \tau_p \), peak intensity \( I_0 \)), which is directed along the \( z \) axis and focused in a spot of transverse size \( d \) (\( \lambda < d \ll \lambda_0 \)), is incident on the medium at a point \( r_0 = \{0, 0, 0\} \) (see Fig. 1). We shall assume that \( \tau_p \) is much longer than the photon lifetime in the medium [for nonzero absorption the latter can be estimated as \( (\mu_a c)^{-1} \), where \( c \) is the velocity of light in the medium]. Then, if \( I_0 \) is sufficiently low, the pulse undergoes multiple scattering on the particles of the medium; a certain portion of its energy is absorbed (resulting in slight heating of the medium), while the rest leaves the medium. The temporal coherence of the scattered light is less than that of the incident laser beam.

\[
V_r = \frac{P}{4\pi\eta} \frac{\cos \theta}{r}, \quad V_\theta = -\frac{P}{8\pi\eta} \frac{\sin \theta}{r}, \quad V_\phi = 0, \quad (3)
\]

where \( P \) is the net flux of the mechanical momentum in the jet. Under conditions being considered, \( P \) equals the momentum transferred from the laser radiation to the particles of the medium in a unit time. The flow lines corresponding to expressions (3) are presented in Fig. 2a. A photograph of a stream of particles moving under the influence of the laser radiation is shown in Fig. 2b [15].

If, however, \( I_0 \) becomes sufficiently high for the incident laser beam to initiate motion of the particles in the medium, the radiation is scattered on moving rather than on immobile particles. This causes degradation of the temporal coherence of the scattered light field. Various mechanisms of the laser particle acceleration are possible [13–17]; we shall not specify the mechanism in the present paper. We shall assume, however, that the mass, shape, and structure of any of the microparticles do not change significantly in response to the laser radiation in a time of the order of the photon lifetime in the medium.

We shall now determine the character of the particle motion under the influence of laser radiation. Since we are considering strong focusing of a light beam (\( \lambda < d < \lambda_0 \)), the point of incidence of the beam on the medium may be approximately regarded as a source of particles flying away in the direction of the \( z \) axis. The hydrodynamic interaction results in drag of the neighbouring particles by those initially accelerated. Consequently, a complex field \( V(r) \) of the particle velocities in the medium (a jet of particles) is created. We shall assume that the boundary of the medium at \( z = 0 \) does not affect significantly the velocity field \( V(r) \) far from the boundary (at \( z > \lambda_0 / 2 \)). Then, in order to find \( V(r) \), we shall consider the hydrodynamic equation for a viscous incompressible suspension filling the whole space (see Ref. [23]):

\[
\frac{\partial}{\partial t} \text{curl} V + (V \nabla) \cdot \text{curl} V - (\text{curl} V \nabla) \cdot V = \nu \Delta \text{curl} V. \quad (2)
\]

Here, \( \nu = \eta/\rho \) is the kinematic viscosity of the suspension and \( \rho \) is its density. This equation can be solved analytically only in a limited number of cases. However, a solution can be found for a thin jet of microparticles originating from \( \{0, 0, 0\} \) and directed along the \( z \) axis, as described above (the problem of a submerged jet). When the jet velocity is not too high, the solution in the spherical coordinates is (see Ref. [23])

\[
V_r = \frac{P}{4\pi\eta} \frac{\cos \theta}{r}, \quad V_\theta = -\frac{P}{8\pi\eta} \frac{\sin \theta}{r}, \quad V_\phi = 0, \quad (3)
\]

where \( P \) is the net flux of the mechanical momentum in the jet. Under conditions being considered, \( P \) equals the momentum transferred from the laser radiation to the particles of the medium in a unit time. The flow lines corresponding to expressions (3) are presented in Fig. 2a. A photograph of a stream of particles moving under the influence of the laser radiation is shown in Fig. 2b [15].

Let us estimate the characteristic coherence time of the backscattered light in the case under study by dimensional analysis. For simplicity, we shall restrict ourselves to the case of point-like scatterers (\( \lambda_0 = \lambda \)). Then the parameters relevant to our problem are \( P, \lambda, \eta, \mu_a \), and \( \lambda \). The radiation absorption in the medium can affect the net momentum flux \( P \) in the jet, but since \( \mu_a \ll \lambda_0^{-1} \), absorption can be ignored in the estimate of the coherence time of the scattered light. Then the simplest expression, which has the dimension of time and can be constructed of the remaining parameters, is \( \eta \lambda / P \). Thus we may expect from the dimensional considerations that the coherence time of the backscattered radiation will be of the order of \( \eta \lambda / P \).

3. Calculation of the temporal correlation function

Since it is well known that radiation becomes significantly depolarised as a result of the multiple scattering [1, 22], we shall consider only a scalar wave propagating in a randomly inhomogeneous medium. The electric field \( E(r, t) \) of the wave obeys the wave equation

\[
\left[ \nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right] (1 + \epsilon(r, t)) E(r, t) = 0. \quad (4)
\]

where \( \epsilon(r, t) \) is the random part of the permittivity. The
permittivity of the liquid containing the suspended particles is taken to be unity. Our concern is with the temporal auto-correlation function

\[ C_1(r, \tau) = \langle E(r, \tau) E^*(r, \tau + \tau) \rangle \]

(5)
of the electric field of a multiply scattered wave measured at a point \( r \) on the surface of a semi-infinite medium (from now on the angular brackets will denote the ensemble averaging).

First, we shall consider the case of scattering particles whose size is much less than the radiation wavelength \( \lambda \). Then \( l_r = l \) and the propagation of light in the medium can be regarded as a random walk of the photons. Hence, the analysis of Eqn (4) simplifies [24] and in the case of interest \( (\lambda < l) \) yields the following result for \( C_1(r, \tau) \) [7, 19, 25]:

\[ C_1(r, \tau) = \sum_{n=1}^{\infty} G(r_0, r, n) \exp \left[ -\frac{1}{2} \langle \Delta \phi_n^2(\tau) \rangle \right] , \]

(6)

where \( G(r_0, r, n) \) is the Green function for the geometry under consideration (i.e. the average radiation intensity at \( r \) generated by a point source of coherent light at \( r_0 \), corresponding to multiple scattering paths involving \( n \) single scattering events) and \( \langle \Delta \phi_n^2(\tau) \rangle \) is the variance of the phase difference between two waves scattered along the same path but separated in time by \( \tau \) (the averaging is first made over all possible paths corresponding to the \( n \)-fold scattering and then over all possible microscopic configurations of scatterers in the medium).

For the laminar flow of particles, which occurs at moderate values of \( I \), assuming that the flow velocity does not change significantly over distances of the order of \( l \), we obtain [19]

\[ \langle \Delta \phi_n^2(\tau) \rangle = \frac{2}{15} k^2 \tau^2 \eta^2 \zeta(n) , \]

(7)

where \( k = 2\pi/\lambda \) and \( \zeta(n) \) is determined by the viscous stress tensor

\[ \sigma_{ik}(r) = \eta \left[ \frac{\partial V_i}{\partial x_k} + \frac{\partial V_k}{\partial x_i} \right] . \]

(8)

We can show [19] that

\[ \zeta(n) = \frac{1}{\tau} \left( \sum_{n,k} \sigma_{ik}(r_1) \right) \rho_n(r_0, r_1, r) d^3r_1 , \]

(9)

where the integration is carried out over the whole volume of the randomly inhomogeneous medium and \( \rho_n(r_0, r_1, r) \) is the fraction of the \( n \)-fold-scattered photons travelling from the source of radiation at \( r_0 \) to \( r \) through the point \( r_1 \). We shall assume specifically that in the case being considered, the scattered light is detected in the immediate vicinity of the point of laser incidence on the boundary of the medium. Then, we can approximately set \( r \approx r_0 = \{0, 0, 0\} \) and use an expression for \( \rho_n(r_1) \equiv \rho_n(0, r_1, 0) \) derived in Ref. [19] in the diffusion approximation:

\[ \rho_n(r_1) = \frac{3}{2\pi^2} \frac{nr_1}{\eta^2} \exp \left( -\frac{3r_1^2}{\eta^2 n} \right) . \]

(10)

Finally, calculation of the viscous stress tensor, given by expression (8), carried out taking account of expression (3), yields

\[ \sigma_{rr} = \frac{P \cos \theta}{2\pi r^2} , \quad \sigma_{\theta r} = \sigma_{\theta \theta} = \frac{P \cos \theta}{4\pi r^2} , \]

\[ \sigma_{\theta \theta} = \sigma_{\phi \phi} = \sigma_{\phi r} = 0 . \]

(11)

Substituting these expressions into expression (9) and integrating over the region \( r > l \), where the scattering of light is adequately described in the framework of the diffusion approximation, and bearing in mind expression (7), we obtain

\[ \langle \Delta \phi_n^2(\tau) \rangle = \left( \frac{\tau}{\tau_c} \right)^2 f(n) , \]

(12)

where

\[ \tau_c = \frac{1}{10} \frac{\eta l^2}{P} \]

(13)
is the characteristic coherence time associated with the light-induced jet;

\[ f(n) = \exp \left( -\frac{3}{n} \right) + \frac{3}{n} \text{Ei} \left( -\frac{3}{n} \right) ; \]

(14)

\text{Ei}(x) is the exponential integral [26]. First, we note that the characteristic time given by expression (13) is identical with the result obtained from the dimensional analysis in Section 2, apart from a constant numerical factor. It is interesting to note that \( f(n) \), which describes the relative role of scattering of different orders, proves to be independent of the parameters of the problem and is only controlled by the scattering order \( n \).

Let us consider the dependence of \( f(n) \) on \( n \) (see Fig. 3). Asymptotic expressions for the functions occurring in expression (14) yield

\[ f(n) \approx \frac{n}{3} \exp \left( -\frac{3}{n} \right) , \quad n \to 0 , \]

(15)

\[ f(n) \approx 1 - \frac{3}{n} \ln n , \quad n \to \infty . \]

(16)

Hence, \( f(n) \) vanishes for \( n = 0 \) and tends to a constant limit for \( n \to \infty \). Since the limit equals unity, \( \tau_c \) represents the characteristic coherence time of the radiation associated with, strictly speaking, the scattering of infinite order. The coherence times associated with the scattering of finite order are greater than \( \tau_c \). It is evident from Fig. 3 that \( f(n) \) is a monotonically increasing function of \( n \). It means that the photons which are scattered more times contribute more to the decorrelation of the radiation.

Finally, making use of the expression for the Green function \( G(r_0, r, n) \) derived in the diffusion approximation

![Figure 3: Factor \( f(n) \) which determines the variance of the phase difference between two photons scattered on the same particles inside the medium but separated in time by \( \tau \) [see expression (12)].](image-url)
and assuming that the point of laser incidence on the medium $r_0$ is separated from the point $r$, where the temporal autocorrelation function is measured, by a distance of the order of $l$, we arrive at the final expression for $C_1(\tau)$:

$$C_1(\tau) \propto \sum_{n=0}^{\infty} \frac{1}{n^{3/2}} \exp \left( -\frac{1}{2} \left( \Delta \Phi_n^2(\tau) - \mu_n \ln n \right) \right), \quad (17)$$

where $\langle \Delta \Phi_n^2(\tau) \rangle$ is given by Eqn (12).

We recall that our analysis applies to the scattering of particles whose size is much smaller than the wavelength of the radiation (model of point-like scatterers). If the scatterer size is of the order of $\lambda$ (or greater than $\lambda$), the analysis becomes much more complicated. However, relying on the conclusions of the authors of Refs [28, 29], we may argue that approximate calculations can again be carried out on the basis of the diffusion model of propagation of light in the medium. This makes it possible to obtain formulas where, in contrast to the preceding expressions, $l$ is replaced by $l_n$.

4. Discussion of results

The normalized temporal autocorrelation functions of back-scattered light $C_1(\tau)/C_1(0)$ are presented in Fig. 4 for various values of the product $\mu_n l$ (specifically, we shall assume that $l$ is the same for all the three curves in Fig. 4, but the absorption coefficient $\mu_n$ is different). As expected, the absorption coefficient $\mu_n$ has a relatively small effect on the normalized correlation function of the scattered light. Absorption mainly demonstrates itself by the enhancement of $P$ [and, consequently, by a reduction in $r_0$, see expression (13)] as $\mu_n$ increases. The difference between the curves associated with different values of the absorption coefficient stems from the fact that for small $\mu_n$ ($\mu_n \ll l^{-1}$) a sufficiently large fraction of the scattered light comes from the high-order scattering. The latter contributes significantly to the decorrelation of the radiation.

For a relatively large $\mu_n$ ($\mu_n \sim l^{-1}$), as is seen from expression (17), negligible intensities are associated with large $n$ (due to the term $-\mu_n \ln n$ in the argument of the exponential function), and therefore most of the scattered light comes from the low-order scattering. Although the case of significant absorption is not properly described in our diffusion approximation, expression (17) again gives a physically clear result: the correlation function does not now decrease as fast as for small $\mu_n$, since the photons undergoing fewer scattering events are less decorrelated. The curves corresponding to different $\mu_n$ become parallel for $\tau/t_0 \gg 1$. This is due to the fact that, for large $\tau$, the main effect of the scattered radiation on the temporal autocorrelation function is produced by the photons scattered only a few times. These photons are nearly insensitive to weak absorption of light in the medium.

It can be seen from Fig. 4 that the coherence time of the scattered radiation is almost an order of magnitude longer than $\tau_0$. The reason for this is that $\tau_0$ represents the coherence time associated with the scattering of infinite order, as discussed above, while in accordance with expression (17) the temporal autocorrelation function is given by a sum of contributions from the scattering events of various orders $n$. A contribution from the scattering of order $n$ is proportional to $n^{-3/2}$ in the absence of absorption [see expression (17)]. Therefore, the main role in the sum of expression (17) is played by the terms corresponding to small $n$. The characteristic coherence times associated with these terms may be considerably longer than $\tau_0$ because the factor $f(n)$ proves to be much smaller than unity [see expression (12)].

It is instructive to compare our results, describing the laser particle acceleration, with the corresponding results for the random Brownian motion of the scatterers. In the latter case, we have $\langle \Delta \Phi_n^2(\tau) \rangle \propto n \tau$, i.e. the variance of the phase difference between two waves scattered from the same scatterers but separated in time by $\tau$ is proportional to $n$. The photons corresponding to $n \to \infty$ therefore acquire an infinitely large variance of the phase difference. The reason for this is that all the particles in the medium (independently of their spatial position) are moving identically in the statistical sense. Therefore, even the particles situated infinitely far from the boundary of the medium contribute to the decorrelation of the scattered radiation.

In the investigated case of the light-induced motion the value of $\langle \Delta \Phi_n^2(\tau) \rangle \propto \tau^{\alpha} f(n)$ tends to a finite limit for $n \to \infty$. This stems from the fact that the velocity of the light-induced motion decreases as $1/\tau$ with the distance from the point of the laser incidence on the boundary of the medium [see expressions (3)]. Hence, the particles sufficiently remote from the point of the laser incidence contribute considerably less to the decorrelation of the scattered light compared with the particles which are close to the point of incidence. Consequently, though the photons corresponding to large $n$ penetrate deep into the medium, they are scattered there virtually without loss of coherence. The loss of coherence is largely controlled by the scattering in the vicinity of the point of the laser incidence on the boundary of the medium.

The different temporal dependencies of $\langle \Delta \Phi_n^2(\tau) \rangle$ (a $\propto \tau$ for the Brownian motion and $\propto \tau^2$ for a light-induced jet of scatterers) are associated with the different nature of the particle motion in the two cases. In the first case, the motion of each particle represents a random walk and its root-mean-square displacement in a time $\tau$ is proportional to $\tau^{1/2}$. In contrast, in the case of a light-induced jet each particle of the suspension has a definite velocity, which is uniquely related to its spatial position [see expression (3)]. This velocity can be considered constant, at least on time scales of the order of $\tau_0$ (in this time a given particle is displaced by a distance considerably less than $\lambda$). The displacement of a given particle in a time $\tau$ is therefore proportional to $\tau$. It is this difference between the temporal dependences of the root-mean-square
displacement that gives rise to the qualitatively different dependences of \( \Delta \phi^2(\tau) \) (and, consequently, of the autocorrelation functions) on the delay time \( \tau \).

In real experiments the scatterers are, as a rule, not immobile even in the absence of external disturbances. If one approximately regards their motion as the Brownian random walk with a diffusion coefficient \( D_B \), an additional term \( \tau /2t_0 \), where \( t_0 = (4k^2D_B)^{-1} \), appears in expression (12) [7, 8]. The resultant expression holds as long as the different types of particle motion (the random Brownian walk and the directed light-induced motion) can be considered independent. This is possible at least at a sufficiently low power of the radiation incident on the medium.

5. Conclusions

We hope that the theoretical analysis carried out in the present paper will lead to experiments on observation of the predicted physical effect. Moreover, our results indicate that the optical correlation diagnostics can provide an effective tool for the study of the laser acceleration of microparticles in concentrated suspensions, especially when the motion is difficult to observe directly due to multiple light scattering.

References

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