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Analysis, by the Monte Carlo method, of the validity of the diffusion approximation in a study of dynamic multiple scattering of light in randomly inhomogeneous media

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Abstract. A numerical simulation is reported of dynamic multiple (isotropic and anisotropic) scattering of light in a randomly inhomogeneous medium representing a suspension of light-scattering Brownian particles. The results of a simulation of the temporal autocorrelation function of backscattered light are compared with calculations carried out within the framework of the diffusion approximation for the radiation transport equation. It is shown that, although the diffusion approximation describes incorrectly low-order scattering, it is quite acceptable for dealing with high-order scattering.

1. Introduction

Multiple scattering of waves in randomly inhomogeneous media has been investigated extensively in the last few decades [1]. Recently these investigations have expanded greatly both because of a considerable progress in analytic description of multiple scattering [2] and because of successful application of the Monte Carlo methods in the simulation of such scattering [3, 4]. This type of problem is of major practical importance, partly as a result of modern applications of optical diagnostic methods in medicine [5, 6].

In the last decade, investigators have been attracted by the feasibility of obtaining information on the dynamics of particles in a randomly inhomogeneous medium from an analysis of light scattered multiply in this medium. This is possible not only under conditions of spatially homogeneous (in the statistical sense) motion of particles in a medium [7], but also in the presence of dynamically inhomogeneous regions [8, 9] or of particle flow [9 – 11] in a medium. The results obtained suggest potential applications of optical waves in the diagnostics of the dynamics of scatterers in randomly inhomogeneous media under conditions of multiple scattering of light. However, in theoretical analyses of the problem most authors rely on the diffusion approximation to the transport equation for on the temporal correlation function of the electric field of optical waves in a medium [8, 12], which imposes certain restrictions on the validity of the theoretical results obtained.

We shall analyse in detail the dynamic multiple scattering of optical waves in a randomly inhomogeneous medium by the Monte Carlo method. We shall use the scalar approximation to consider the scattering by particles with an isotropic scattering diagram, as well as strongly anisotropic scattering. We shall assume that the weak-scattering condition ($\lambda \ll l$, where λ is the optical wavelength and l is the mean free path of a photon) is satisfied by the medium. We shall compare the results obtained by the Monte Carlo method with those deduced from an analysis of the problem on the basis of the diffusion approximation.

It has been shown that the diffusion approximation describes satisfactorily that fraction of the scattered radiation which is the result of high-order scattering. The diffusion approximation is unacceptable for a complete description of the characteristics of the scattered radiation in the case of diffuse reflection because then the contribution of low-order (single, double, etc.) scattering is important. Since the solution of the transport equation describing correctly the scattering of all orders is not yet available, stochastic simulation is essentially the only method capable of predicting experimental results in those cases when an important role is played by low-order scattering as well as by multiple scattering.

2. Spatial diffusion of radiation by the scattering of light in a randomly inhomogeneous medium

Let us consider a layer of a randomly inhomogeneous medium bounded by the planes $z = 0$, $z = L$ and consisting of a large number of tiny spherical particles, with the diameter a, which are distributed at random and which are moving in space. We shall describe the properties of this medium by the absorption coefficient μ_a, the scattering coefficient μ_s, and the reduced scattering coefficient $\mu_s' = (1 - g)\mu_s$. Here, g is the scattering anisotropy parameter which has to be calculated taking account of the specific form of the phase function $p(\cos\theta)$ characterising the diagram representing the scattering of an optical wave by a single particle:

$$g = 2\pi \int_0^{\pi} \cos\theta p(\cos\theta) \sin\theta \, d\theta \,. \tag{1}$$

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We shall use the Heney–Greenstein model of the phase function [1]

$$p(\cos\theta) = \frac{1}{4\pi}\frac{1-g^2}{(1+g^2-2g\cos\theta)^{3/2}}\,, \tag{2}$$

employed widely in multiple-scattering problems. The phase function $p(\cos\theta)$ has a simple physical meaning: the probability of the scattering through an angle in the interval $[\theta, \theta + d\theta]$ is equal to $2\pi p(\cos\theta)\sin\theta\, d\theta$. Isotropic scattering corresponds to $g = 0$ (scattering by point particles), whereas $g \approx 1$ characterises strongly anisotropic scattering [13] (scattering by large particles with the scattering diagram elongated in the forward direction).

The quantities μ_a, μ_s, μ_s' have the dimensions of reciprocal length. It is sometimes more convenient to use the reciprocals of these quantities, namely $l_a = \mu_a^{-1}$, $l = \mu_s^{-1}$, $l_{tr} = (\mu_s')^{-1}$. The absorption length l_a is the average distance which a photon travels in a medium before it is absorbed. The mean free path of a photon l is the average distance travelled by a photon between successive scattering events. Finally, the transport mean free path l_{tr} is the distance required for isotropisation of the radiation, which initially has a definite direction, as a result of the scattering. For point scatterers ($a \ll \lambda$), we have $g = 0$, $l_{tr} = l$, i.e. the direction of propagation of a photon becomes random after the very first scattering event. If $a \sim \lambda$, then $0 < g < 1$ and $l_{tr} > l$. This means that isotropisation requires $n > 1$ scattering events, which on the average is given by $n = l_{tr}/l = (1-g)^{-1}$.

If $l_a \gg l_{tr}$, then over distances much greater than l_{tr} the spatial propagation of radiation is diffusive. Such propagation in a medium can then be described within the framework of the diffusion approximation for the transport equation regarding the propagation as diffusive. This approach gives very accurate results far from a radiation source and far from the boundaries of a randomly inhomogeneous medium, i.e. in the situations when the role of low-order scattering is negligible. In the opposite case the diffusion approximation is insufficient and more accurate results may be obtained by stochastical stimulation of the scattering of light in a medium by the Monte Carlo method.

3. Elementary theory of the dynamic multiple scattering

Since light becomes strongly depolarised as a result of multiple scattering, we shall simplify the calculations by adopting the scalar approximation. We shall be interested in the temporal autocorrelation function of the electric field $G_1(\boldsymbol{r}, \boldsymbol{k}, \tau) = \langle E(\boldsymbol{r}, \boldsymbol{k}, t)E^*(\boldsymbol{r}, \boldsymbol{k}, t+\tau)\rangle$ of a multiply scattered wave, emerging from the investigated medium in the direction of the wave vector $\boldsymbol{k}$. The angular brackets are used here and later to denote averaging over an ensemble of realisations, which is equivalent to time averaging of an ergodic system. It is shown in Ref. [1] that under the conditions of spatial diffusion of light the properties of light become practically independent of the direction. We shall therefore assume that G_1 is independent of the direction of $\boldsymbol{k}$ and that the wave number k remains constant in the course of scattering (coherent scattering). We shall discuss the case when the velocities of particles in the investigated medium are much lower than the velocity of light c.

We shall consider, in our medium, a single path of a photon along which it is scattered n times at points $\boldsymbol{r}_1, \ldots, \boldsymbol{r}_n$. The photon generates, at the point of its exit from the medium at a time t, an electric field of intensity $E_n(t)$. The changes of the photon wave vector in consecutive scattering events will be denoted by $\boldsymbol{q}_1, \ldots, \boldsymbol{q}_n$. Then, as can easily be demonstrated, the product of the electric field of the scattered wave at a time t and of the same field at a moment $t+\tau$ is [7, 14]

$$E_n(t)E_n^*(t+\tau) = |E_n(t)|^2 \exp[-i\Delta\Phi_n(\tau)]\,. \tag{3}$$

Here,

$$\Delta\Phi_n(\tau) = \sum_{i=1}^{n} \boldsymbol{q}_i \cdot \Delta\boldsymbol{r}_i(\tau)\,; \tag{4}$$

$\Delta\boldsymbol{r}_i = \boldsymbol{r}_i(t+\tau) - \boldsymbol{r}_i(t)$ is the displacement of the ith particle in the time τ.

We shall assume that the particles in the medium do not interact with one another and that their motion is Brownian. Then, the displacement $\Delta\boldsymbol{r}_i(\tau)$ of each of the light-scattering particles is a random quantity distributed in accordance with the Gaussian law with zero average and the variance $\langle[\Delta\boldsymbol{r}_i(\tau)]^2\rangle = 6D_B\tau$ where D_B is the diffusion coefficient of the particles. Since we are considering here the case when $\lambda \ll l$, it follows that the fields created by photons travelling along different paths are summed incoherently. Averaging expression (3) over all the paths with the same number n of the scattering events and over all possible microscopic configurations of the light-scattering particles, and then summing the contributions of the scattering processes of different orders, we obtain

$$G_1(\boldsymbol{r}, \tau) = I_0(\boldsymbol{r}) \sum_{n=1}^{\infty} P(\boldsymbol{r}, n)\langle \exp[-i\Delta\Phi_n(\tau)]\rangle\,, \tag{5}$$

where $P(\boldsymbol{r}, n)$ is the fraction of the average intensity $I_0(\boldsymbol{r})$ of the scattered field at a point $\boldsymbol{r}$ resulting from nth-order scattering. The above expression can be simplified analytically within the framework of the diffusion approximation if additional assumptions are made (in particular, that $\mu_a \ll \mu_s'$). For example, if a plane wave is incident on a layer of a nonabsorbing randomly inhomogeneous medium, the autocorrelation function of the diffusely scattered light is described, apart from an unimportant numerical factor, by [14, 15]

$$G_1(\tau) \propto \frac{\sinh[\alpha(L + z_1 - z_0)]}{\sinh[\alpha(L + 2z_1)]}\,, \tag{6}$$

where $\alpha = [3\tau/(2\tau_0 l_{tr}^2)]^{1/2}$; $z_0 \approx l_{tr}$ is the distance in which the collimated light incident on the medium is converted into diffuse light; $z_1 \approx (2/3)l_{tr}$ is the distance from the boundary of the medium to a plane (extrapolated boundary) in which the zero boundary condition is imposed on G_1. The characteristic time τ_0 is related to the diffusion coefficient of the particles in the medium by the expression $\tau_0 = (4k^2 D_B)^{-1}$.

4. Monte Carlo method in the problem of multiple scattering

As shown in Ref. [16], the analytic result represented by expression (6) is largely approximate and it describes incorrectly the contribution of low-order scattering. However, it is possible to calculate the sum in expression (5) accurately by numerical simulation. We shall therefore simulate the propagation of photons in a medium exactly as was done in the simulation of scattering by an ensemble of immobile scatterers [17, 18]. At the entry to the medium the direction of the photon wave vector $\boldsymbol{k}$ is governed by the light source and it can be random (a point source or a bounded laser beam) or specified precisely (a plane wave).

The change in the direction of $\boldsymbol{k}$ in each scattering event is random and the probability density of the scattering of a photon through an angle θ is given by the phase function (2). Between consecutive scattering events the photon travels a distance z_l and the probability that this distance lies within the interval $[z, z + dz]$ is $p(z) = l^{-1}\exp(-z/l)dz$. These assumptions make it possible to simulate the propagation of a photon along any of the possible paths in the medium and to calculate the contribution of each of the paths to the correlation function $G_1(\boldsymbol{r}, \tau)$ of light emerging from the medium at a point $\boldsymbol{r}$.

The absorption of light can easily be taken into account in our model. If the scattering and absorption coefficients are μ_s and μ_a, respectively, the single-scattering albedo is $W_0 = \mu_s/(\mu_a + \mu_s)$. It is then obvious that the probability of absorption of a photon in a 'collision' with a particle in the medium is equal to $1 - W_0$. We can take into account the possibility of absorption of a photon by the particle itself or by a liquid or a gas in which the investigated particles are suspended.

5. Scattering of a plane wave on a layer of a randomly inhomogeneous medium

We shall apply the Monte Carlo method described above to analyse the scattering of a plane wave on a layer of a randomly inhomogeneous medium of thickness L. First of all, we shall consider how the transmission (T) and reflection (R) coefficients, equal to the ratios of the intensities of the transmitted and reflected light to the intensity of the incident light, depend on L. We shall do this by simulating the propagation in this medium of a certain number of N photons. We shall find the number of photons N_t transmitted by the medium, as well as the number of photons N_r reflected by the medium. The required transmission and reflection coefficients are obviously $T = N_t/N$ and $R = N_r/N$. These quantities can be calculated by substituting $\tau = 0$ in all the formulas of the preceding section. The Monte Carlo method then reduces to the familiar algorithm for numerical solution of the transport equation [18]. Here and later we shall limit ourselves to $N = 5 \times 10^4$. A calculation of an ensemble of realisations for $L = 10l_{tr}$ and of the scattering with an arbitrary phase function takes about one hour on a personal computer with a Pentium MMX 166 MHz processor.

When the scatterers are point-like and they do not absorb light ($g = 0$, $l_{tr} = l$, $\mu_a = 0$), the results obtained by the Monte Carlo method agree well with the theoretical formulas [19] $T \approx l/L$ and $R \approx 1 - l/L$ (Fig. 1), derived in the diffusion approximation.

However, for all values of L the Monte Carlo method gives results which differ somewhat from the analytic results. Therefore, we can say that in the statistical case the diffusion approximation provides a qualitatively valid description of multiple scattering, but the results are quantitatively quite wrong. This is supported also by the results obtained for anisotropic scatterers (we investigated the range $0 < g < 0.95$), which we shall not give here. The curves obtained for anisotropic scatterers coincide with the curves shown in Fig. 1. We must recall that $l_{tr} > l$ when $g > 0$ and, therefore, for large anisotropy parameters the simulation was carried out for layers of larger *physical* thickness.

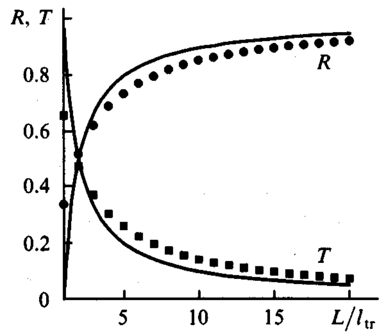

Figure 1. Transmission (T) and reflection (R) coefficients of light plotted as a function of the thickness L of a layer of a randomly inhomogeneous medium consisting of point scatterers (the dots and the squares are the numerical simulation results and the continuous curves are analytic).

We shall now consider the behaviour of the temporal correlation function of light backscattered by a layer of a randomly inhomogeneous medium. If a plane wave is incident on this medium, then G_1 is independent of the position of the point $\boldsymbol{r}$ on the surface of the medium at which the measurements are carried out, i.e. $G_1(\boldsymbol{r}, \tau) \equiv G_1(\tau)$ is determined. The analytic result corresponding to this case is given by formula (6). We shall assume specifically that $L = 10l_{tr}$ and begin with point scatterers that do not absorb light ($g = 0$, $l_{tr} = l$, $\mu_a = 0$). The Monte Carlo method enables us to study the role of the various scattering orders in this particular case.

Fig. 2 gives the normalised temporal autocorrelation function of diffusely scattered light $g_1(\tau) = G_1(\tau)/G_1(0)$. We can see that an increase in the highest of the included scattering orders increases the rate of fall of the correlation function with increase in τ. Moreover, the correlation function for $n = 1$, which corresponds to single scattering, falls almost exponentially with increase in τ/τ_0 (the curve for $n = 1$ is nearly a parabola) and the function g_1 obtained by including all the scattering orders falls almost exponentially as a function of $(\tau/\tau_0)^{1/2}$ (the curve for $n = \infty$ is almost a straight line). This behaviour of the temporal autocorrelation functions is in good agreement with the published theoretical and experimental results [7, 14].

In reality, the measured quantity corresponds to the curve for $n = \infty$ since it is experimentally impossible to separate

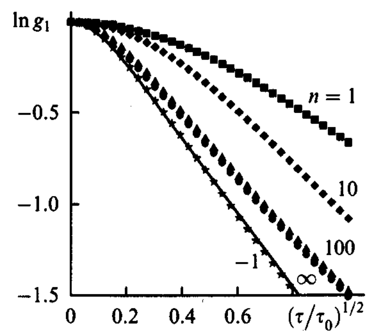

Figure 2. Normalised temporal autocorrelation functions of diffusely reflected light, obtained for $L = 10l_{tr}$ by numerical simulation [single scattering (■), scattering with $n \leqslant 10$ (◆), with $n \leqslant 100$ (▲), including all scattering orders (●), and ignoring single scattering (★)] and by analytic calculation (continuous curve).

the contributions of various scattering orders. It is evident from Fig. 2 that analytic results (continuous curve) and the curve for $n = \infty$ diverge quite considerably, i.e. the diffusion approximation is unacceptable for the description of the temporal correlation function of backscattered light. On the other hand, if we exclude the contribution of single-scattering processes (curve for $n = -1$), the Monte Carlo results are in excellent agreement with the analytic expressions. In other words, the diffusion approximation describes well the temporal correlation of diffusely scattered light in the situations in which the role of the low-order scattering is negligible.

We shall now consider anisotropic scattering ($0 < g < 1$). The transport mean free path of a photon l_{tr}, which is a characteristic scale of the problem, is no longer equal to the mean free path l. It follows from formula (6) that there is a definite similarity: the results for the case when $g \neq 0$ can be derived from the results corresponding to $g = 0$ by replacing l with l_{tr}. This conclusion is supported by the results of our numerical simulation, which are presented in Fig. 3. If the ratio of the thickness L of a layer of a randomly inhomogeneous medium to l_{tr} remains constant (for the curves in Fig. 3, we have $L/l_{tr} = 10$), a simulation carried out for different scattering anisotropy parameters ($g = 0, 0.5$, and 0.9) gives almost identical results. It should be stressed that for $g = 0, 0.5$, and 0.9, the simulation was carried out for different *physical* thicknesses L of the layer ($L = 10l$, $20l$, and $100l$, respectively).

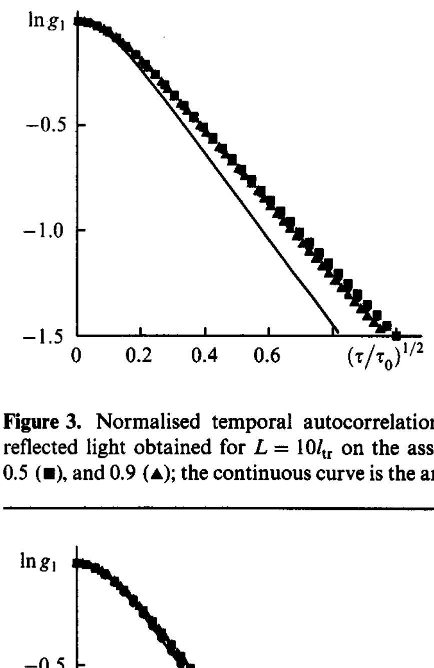

Figure 3. Normalised temporal autocorrelation functions of diffusely reflected light obtained for $L = 10l_{tr}$ on the assumption that $g = 0$ (●), 0.5 (■), and 0.9 (▲); the continuous curve is the analytic result.

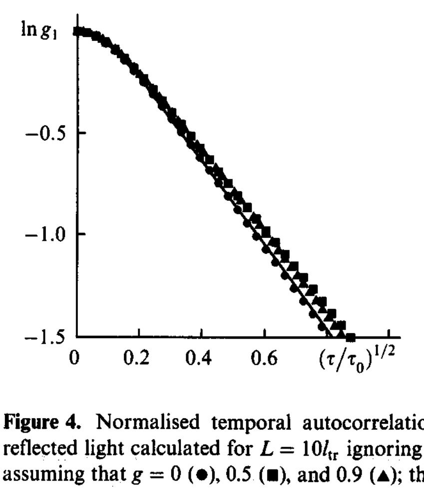

Figure 4. Normalised temporal autocorrelation functions of diffusely reflected light calculated for $L = 10l_{tr}$ ignoring low-order scattering and assuming that $g = 0$ (●), 0.5 (■), and 0.9 (▲); the continuous curve is the analytic result.

It is clear from Fig. 3 that the analytic formula (6), obtained on the basis of the diffusion approximation, gives different results from those obtained by numerical simulation employing the Monte Carlo method. As before, this occurs because in the diffusion approximation the contribution of low-order scattering is described incorrectly. However, if we artificially remove the photons which are scattered only a few times — $n \leqslant n_0 = l_{tr}/l$ ($n_0 = 1$ for $g = 0$, $n_0 = 2$ for $g = 0.5$, and $n_0 = 10$ for $g = 0.9$) — the analytic results agree quite well with the numerical data (Fig. 4). This makes it possible to generalise the conclusions on the degree of validity of the diffusion approximation, stated above for the isotropic scattering case, to anisotropic scattering situations when the dimensions of the light-scattering particles are comparable with the wavelength of light.

6. Conclusions

The Monte Carlo method of statistical simulation can be used successfully to tackle a number of problems relating to dynamic multiple scattering of laser radiation in randomly inhomogeneous media. The Monte Carlo method makes it possible to include the influence of various orders of the scattering for point particles as well as for particles whose size is comparable with the wavelength, which cannot be done analytically. On the other hand, if the influence of the low-order scattering ($n \leqslant l_{tr}/l$) is eliminated, the numerical simulation results are in good agreement with those derived within the framework of the diffusion approximation for the transport equation.

It follows that, in the situations when the role of the low-scattering is small, the diffusion approximation gives quite acceptable results. Under these conditions this approximation may even be preferable, since calculations carried out with the aid of analytic formulas are much less time-consuming than statistical simulation. In some situations we may, however, find that it is important to include the low-order scattering, which is described incorrectly within the diffusion approximation framework. In such cases the Monte Carlo method has indisputable advantages. In the majority of situations of practical importance it would be reasonable to use simultaneously both analytic and numerical methods. In fact, it is possible to calculate the contribution of multiple scattering within the framework of the diffusion approximation, and to find the contribution of low-order scattering by the Monte Carlo method. The results obtained in the course of the present investigation support the correctness of this approach.

We shall conclude by noting that in the applications it may prove very important to include such factors as the interaction between particles in a medium, deviation from the Brownian nature of their motion, structure of a sample, etc. However, these effects lead to considerable and so far unresolved difficulties in the analytic approach, which can easily be taken into account by numerical simulation using the Monte Carlo method. The algorithm described above is a convenient, and in many respects universal, method for investigating dynamic multiple scattering of light in randomly inhomogeneous media.

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