Dynamic multiple scattering of light in multilayer turbid media

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Abstract

We present a theoretical calculation of time autocorrelation functions of multiply scattered light in a turbid multilayer medium. The general expressions for correlation functions in transmission and backscattering geometries are presented. Useful examples of multilayer systems are considered.

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During the last decade a novel technique for the analysis of multiple light scattering in turbid media, the so-called diffusing wave spectroscopy (DWS), was introduced [1], and it has developed very rapidly [2–4] (see Ref. [12] for detailed discussions). Although this technique was first used in macroscopically homogeneous media where scatterers undergo pure Brownian motion [1–3], later the framework of DWS was extended to incorporate situations of shear [5,6] and inhomogeneous [7,8] laminar flows and those of large-scale heterogeneous media [9,10]. In the latter case DWS allows imaging of dynamical heterogeneities since methods similar to those used for imaging of absorbing objects [11] can be applied. In this contribution we report theoretical results on dynamic multiple light scattering in a turbid multilayer medium, where the scatterers are mobile either by Brownian motion like particles in suspension, or by flow drag of laminar type. Besides the intrinsic importance of our results for light scattering problems, they can also be applied to scattering of any classical waves (e.g., acoustic waves) in a strong multiple scattering regime.

We consider a thick slab of width \( L \) limited by the planes \( z = 0, z = L \) and composed of \( N \) layers filled with a turbid medium. The layer number \( n \) is situated between the planes \( z = z_{n-1} \) and \( z = z_n \) and characterized by its width \( \Delta_n = z_n - z_{n-1} \). Particle \( (D_B^{(n)} = (4k_0^2\tau_0^{(n)})^{-1}) \) and photon \( (D_p^{(n)} = vt^{(n)}/3) \) diffusion coefficients are assumed to be constant inside a given layer, but can vary from layer to layer. In the previous expressions \( \tau_0^{(n)} \) and \( t^{(n)} \) are the characteristic time of the Brownian motion of scatterers and the transport mean free path for elastic scattering inside the \( n \)th layer, respectively, \( k_0 \) and \( v \) denote the wave number of the light and the speed of light in the medium. Light absorption is neglected in these expressions for the sake of simplicity.

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Under conditions of strong multiple scattering ($k_0 t^{(n)} \gg 1$, $\Delta_n \gg t^{(n)}$) inside each layer and for an extended plane wave incident on the slab surface at $z = 0$, the unnormalized time autocorrelation function of the depolarized scattered electric field inside the $n$th layer, $G_1^{(n)}(r, \tau) = \langle E(r, t) E^*(r, t + \tau) \rangle$ in the scalar approximation\(^2\), obeys the well-known steady-state diffusion equation \([9,12]\),

\[
[\nabla^2 - \alpha^{(n)^2}(\tau)] G_1^{(n)}(r, \tau) = -S\delta(z - z_0), \quad (1)
\]

where $\alpha^{(n)^2}(\tau) = 3\tau / 2\tau^{(n)^2}$ for the case of pure Brownian motion of scatterers inside the layers, $\delta(z - z_0)$ is a Dirac delta function and $z_0$ is the position of the first scattering event of photons entering in the slab: this position is assumed to be inside the first layer and at a distance of the order of the mean free path from the slab surface ($z_0 \approx l_s^{(1)}$) \([13]\).

Solutions of Eq. (1) inside the layers have the following simple forms,

\[
G_1^{(1)}(z, \tau) = A^{(1)}(z) \exp(\alpha^{(1)}z) + B^{(1)}(z) \exp(-\alpha^{(1)}z) + (S/\alpha^{(1)}) \exp(-\alpha^{(1)}|z - z_0|),
\]

\[
G_1^{(n)}(z, \tau) = A^{(n)}(z) \exp(\alpha^{(n)}z) + B^{(n)}(z) \exp(-\alpha^{(n)}z), \quad n = 2, \ldots, N. \quad (2)
\]

Here $A^{(n)}$ and $B^{(n)}$ are unknown coefficients which one determines from the boundary conditions on the surfaces $z = 0$, $z = L$ of the slab, and on the boundaries $z_n$ between the layers \([9,13]\),

\[
G_1^{(1)}(0, \tau) = G_1^{(N)}(L, \tau) = 0,
\]

\[
G_1^{(n)}(z_n, \tau) = G_1^{(n+1)}(z_n, \tau),
\]

\[
D_p^{(n)} \frac{dG_1^{(n)}(z_n, \tau)}{dz} = D_p^{(n+1)} \frac{dG_1^{(n+1)}(z_n, \tau)}{dz}, \quad n = 1, \ldots, N - 1. \quad (3)
\]

Simultaneous analysis of Eq. (2) and Eq. (3) leads to a system of linear equations for $A^{(n)}$ and $B^{(n)}$ which we describe in matrix notation. The interface between the $n$th and $(n + 1)$-th layers is characterized by the matrix $T^{(n)}(\tau)$ given by

\[
T^{(n)}(\tau) = \frac{1}{2} \times \begin{pmatrix}
\beta^{(n)}(+) \exp(\alpha^{(n)}z_n) & \beta^{(n)}(-) \exp(-\alpha^{(n)}z_n) \\
\beta^{(n)}(-) \exp(\alpha^{(n)}z_n) & \beta^{(n)}(+) \exp(-\alpha^{(n)}z_n)
\end{pmatrix}, \quad (4)
\]

for $n = 1, \ldots, N - 1$, with

\[
\alpha^{(n)}(\pm) = \alpha^{(n+1)} \pm \alpha^{(n)}, \quad (5)
\]

\[
\beta^{(n)}(\pm) = 1 \pm \frac{D_p^{(n+1)} \alpha^{(n+1)}}{D_p^{(n)} \alpha^{(n)}}. \quad (6)
\]

Adding a definition of $T^{(n)}$ for $n = N$ in the form

\[
T^{(N)}(\tau) = \begin{pmatrix}
-\exp(-\alpha^{(N)}L) & -\exp(-\alpha^{(N)}L) \\
\exp(\alpha^{(N)}L) & \exp(\alpha^{(N)}L)
\end{pmatrix}, \quad (7)
\]

one obtains general expressions for time autocorrelation functions of backscattered ($G_1^B(\tau)$) and transmitted ($G_1^T(\tau)$) light in the limit $\alpha^{(1)}t^{(1)} \ll 1$,

\[
G_1^B(\tau) = \frac{\text{Tr}(S_B T_N)}{\text{Tr}(T_N)}, \quad (8)
\]

\[
G_1^T(\tau) = \frac{\text{Tr}(S_T)}{\text{Tr}(T_N)}, \quad (9)
\]

where constant factors proportional to $S$ (see Eq. (1)) are omitted. Here $T_N(\tau) = T^{(1)}(\tau) T^{(2)}(\tau) \ldots \times T^{(N)}(\tau)$, $T_N$ denotes the trace of the corresponding matrix, $S_B$ and $S_T$ are diagonal matrices determined by the exact positions of first ($z = z_0$) and last ($z = z_0$) for backscattering and $z = L - z_0$ for transmission) scattering events of diffusing photons,

\[
S_B(\tau) = \begin{pmatrix}
\exp(\alpha^{(1)}z_0) & 0 \\
0 & \exp(-\alpha^{(1)}z_0)
\end{pmatrix}, \quad (10)
\]

\[
S_T(\tau) = \begin{pmatrix}
\exp(\alpha^{(N)}z_0) & 0 \\
0 & -\exp(-\alpha^{(N)}z_0)
\end{pmatrix} \quad (11)
\]

Here we supposed that photons leave the slab if they approach closer than $z_0 \approx l_s^{(n)}$ (with $n = 1$ for backscattering and $n = N$ for transmission) to its boundaries. More precise estimations of $z_0$ appropriate for backscattering and transmission geometries are discussed in Refs. \([8,14]\).
The formulas given by Eqs. (8), (9) provide a useful framework for analysis of correlation properties of light backscattered from or transmitted through a multilayer slab. First, it appears that all the multiple scattering properties of the medium relevant for the correlation function of backscattered or transmitted light are contained in the $2 \times 2$ matrix $T_N(\tau)$ defined as a product of “local” matrices $T^{(n)}(\tau)$, related to the variations of these properties inside the medium. Next, from Eqs. (8), (9) it follows that it is the very same matrix $T_N(\tau)$ which determines both considered correlation functions. From general properties of matrix $T_N(\tau) = \| T_N^{lm}(\tau) \|^2$, we would like to outline that det($T_N$) = 0 and, more precisely, that $T_N^{11}(\tau) = T_N^{12}(\tau)$ and $T_N^{22}(\tau) = T_N^{21}(\tau)$, as follows from Eqs. (4), (7).

Note that when an analytical expression for $T_N(\tau)$ is known for given $N$, the matrix $T_{N+1}(\tau)$ that corresponds to the system with one more layer added, can be found by a simple multiplication of the matrix describing this additional layer by $T_N(\tau)$. Using this property one finds particular expressions for $T_N(\tau)$,

$$T_1^{11} = T_1^{12} = - \exp(-aL),$$
$$T_1^{21} = T_1^{22} = \exp(aL),$$

$$T_2^{11} = T_2^{12} = \exp(-a^{(1)}\Delta_1) \left( \sinh(a^{(2)}\Delta_2) - \frac{D_p^{(2)}\alpha^{(2)}}{D_p^{(1)}\alpha^{(1)}} \cosh(a^{(2)}\Delta_2) \right),$$

$$T_2^{21} = T_2^{22} = \exp(a^{(1)}\Delta_1) \left( \sinh(a^{(2)}\Delta_2) + \frac{D_p^{(2)}\alpha^{(2)}}{D_p^{(1)}\alpha^{(1)}} \cosh(a^{(2)}\Delta_2) \right),$$

$$T_3^{11} = T_3^{12} = \exp(-a^{(1)}\Delta_1) \left( \cosh(a^{(2)}\Delta_2) \sinh(a^{(3)}\Delta_3) - \frac{D_p^{(2)}\alpha^{(2)}}{D_p^{(1)}\alpha^{(1)}} \sinh(a^{(2)}\Delta_2) \sinh(a^{(3)}\Delta_3) \right) + \frac{D_p^{(3)}\alpha^{(3)}}{D_p^{(1)}\alpha^{(1)}} \sinh(a^{(2)}\Delta_2) \cosh(a^{(3)}\Delta_3) + \frac{D_p^{(3)}\alpha^{(3)}}{D_p^{(1)}\alpha^{(1)}} \cosh(a^{(2)}\Delta_2) \cosh(a^{(3)}\Delta_3) \right),$$

and so on. Although an analytical expression for $T_N(\tau)$ can be obtained for any given $N$, this leads to involved calculations when $N$ is large. In contrast, “local” matrices $T^{(n)}$ for $n = 1, \ldots, N$ can easily be calculated for any $N$ and their product can be obtained numerically. Therefore, Eqs. (8), (9) provide a simple numerical algorithm for the calculation of correlation functions in backscattering and transmission geometries.

For $N = 1$ one can use Eqs. (8), (9) in conjunction with Eq. (12) to obtain expressions for correlation functions of light transmitted through or reflected from the homogeneous slab. After simple calculations we obtain exponential decays of both correlation functions with characteristic times $\sim \tau_0$ in reflection and $\sim \tau_0(1/L)^2$ in transmission, that coincide with results previously obtained by other authors [4,14].

A surprisingly simple result for the correlation function of backscattered light can be obtained for a semi-infinite medium composed of two thick layers, the first of which is rigid ($\tau_0^{(1)} \to \infty$) and has a finite width ($\Delta_1 < \infty$), while the second is filled with fluid medium ($\tau_0^{(2)} < \infty$) and occupies the half-space $z > \Delta_1$ ($\Delta_2 \to \infty$). Using Eq. (8) and Eq. (13) and assuming for simplicity $D_p^{(1)} = D_p^{(2)}$, we obtain

$$G_1^B(\tau) = 1 - \frac{z_0}{\Delta_1 + a^{(2)} - 1}. \quad (15)$$

As follows from this equation, $G_1^B(\tau)$ varies from 1 for $\tau = 0$ to $1 - z_0/\Delta_1$ for $\tau \to \infty$. Therefore, one can determine the depth $\Delta_1$ of the semi-infinite turbid layer by measuring this asymptotic value and then $D_p^{(2)}$ from the curve defined by Eq. (15).

As another example of a possible application of the presented theoretical analysis, we consider a layer
of turbid medium characterized by particle and photon diffusion coefficients $D_B^{(2)}$ and $D_p^{(2)}$, respectively, embedded in an otherwise homogeneous turbid slab $(D_B^{(1)} < D_B^{(2)}, D_p^{(1)} = D_p^{(2)})$ of width $L$. The width of the layer is $A \gg L^*$ and its position inside the slab is $z$, that is, equal to the distance between the slab surface $z = 0$ and the nearest to it layer side. In order to analyze the correlation function of transmitted or backscattered light for such a medium, we employ Eqs. (8), (9) with matrix $T_3(\tau)$ given by Eq. (14), where we suppose $A_1 = z$, $A_2 = A$, $A_3 = L - A_1 - A_2$ and $D_B^{(1)} = D_B^{(3)}$, $D_p^{(1)} = D_p^{(2)} = D_p^{(3)}$. We consider the particular case of a larger particle diffusion constant inside the layer than in the surrounding medium, i.e. the situation when a heterogeneous layer leads to a more rapid decay of the correlation function than in the homogeneous case. We assume $L = 100 L^*$ and $A = 10 L^*$ and for simplicity we are interested in the normalized time autocorrelation function $g_1(\tau) = G_1(\tau)/G_1(0)$. Calculations show that correlation functions of backscattered and transmitted light both suffer perturbations relative to the homogeneous case. To describe these perturbations we determine the correlation time $\tau_{\text{max}}$ for which the maximum deviation from the normalized correlation function $g_1^0(\tau)$ for a homogeneous slab occurs, and in the absolute value of this deviation itself,

$$ \Delta g = \max_{0 < \tau < \infty} \left| g_1(\tau) - g_1^0(\tau) \right| = \left| g_1(\tau_{\text{max}}) - g_1^0(\tau_{\text{max}}) \right|. \tag{16} $$

In Fig. 1 $\tau_{\text{max}}$ and $\Delta g$ are shown as functions of heterogeneous layer position (depth). The inset of this figure shows that the influence of the same heterogeneous layer is nearly always greater in transmission, except in the region of extremely small depths $z$.

In transmission, both $\tau_{\text{max}}$ and $\Delta g$ have a symmetric form, the layers placed at $z$ and at $L - z$ produce exactly the same perturbations of the correlation function, that can also be deduced directly from Eqs. (9), (14). Hence, when the correlation function is measured in transmission, these two situations are indistinguishable. The exact position of the layer can be found only when it is placed in the middle of the slab. In this case the deviation of the correlation function from the homogeneous case is the largest, which can easily be explained using the notion of density of the diffusing photons paths distribution [7]. This density is obviously the largest at $z = L/2$ and therefore, when a heterogeneous layer is situated near this plane, perturbations of the correlation function reach the maximum, since the greatest part of the diffusing photons scans the region of heterogeneity. For similar reasons, the maximum deviation in the case of $z = L/2$ is achieved for the shortest time $\tau_{\text{max}}$. The photons that gather the largest additional dephasing, pass at least several times through the heterogeneous layer, which is possible only if they have extremely long diffusion paths. Since, in accordance to the basic principle of DWS, long diffusion paths determine the decay of the correlation function at short times and vice versa [4], the largest deviation from the unperturbed correlation function is achieved for the shortest time $\tau$ if the layer is placed exactly in the middle of the slab.

The correlation function of backscattered light exhibits quite a different behaviour. Now deviations from the homogeneous case correlation function are the largest for the smallest depths $z$ of the heterogeneous layer, since in this case the greatest part of the photons scans its volume. These deviations reduce rapidly with an increase of $z$ and become smaller than 1% already for $z \approx 25 L^*$. In practice this means that for heterogeneous layer depth greater than $25 L^*$, for considered dynamic contrast $(\tau_{\text{max}}^0 = 0.25 \tau_0^{(1)})$ and layer width $(A = 10 L^*)$, the layer cannot even be detected. This conclusion qualitatively coincides with a similar
result obtained from multiple light scattering experiments with cylindrical heterogeneity [15]. The correlation time $\tau_{\text{max}}$ for which maximum deviation occurs, decreases with an increase of $z$. When the layer is placed near the slab surface $z = 0$, even the photons that have relatively short diffusion paths scan its volume and are additionally dephased. These short paths correspond to large correlation times as shown in Fig. 1. When the depth $z$ of a heterogeneous layer is sufficiently large, only photons that have long diffusion paths reach the layer and gather the largest additional dephasing. Since they determine a decrease of the correlation function mainly at short times $\tau$, the $\tau_{\text{max}}$ decreases with an increase of $z$.

In conclusion, we have derived general expressions for correlation functions of light transmitted or reflected from a multilayer slab filled with turbid medium in a strong multiple scattering regime. These results allow us to analyze scattered light correlation properties in general as well as provide simple analytical expressions for corresponding correlation functions in particular cases, two of those were examined in order to show the way in which further analysis based on reported general results can be performed. In addition, one can imagine different interesting problems (such as randomly distributed along the $z$-axis particle or/and photon diffusion coefficients, complex planar flows, etc.), that can be analyzed within the framework provided. In particular, in the case of laminar flow of scatterers inside the $n$th layer, the corresponding $\alpha^{(n)}$ becomes $3\tau / 2 T_0^{(n)} \tau_{\text{d}}^{(n)} + 6 (\tau / \tau_{\text{d}}^{(n)})^2$, where the characteristic time introduced by the flow $\tau_{\text{d}}$ is roughly determined by the average velocity gradient inside this layer [10]. Thus, any complex distribution of flows that appears in particular problems (e.g., in meteorology, where wind velocity profiles can be quite sophisticated), when the multilayer approximation is good, can be analyzed by using our general result. Moreover, when applied to multiple scattering of other classical (not electromagnetic) waves, the presented analysis can also lead to interesting applications of diffusing waves in various areas of human activity (e.g., for detection of petrol layers by measuring the correlation function of acoustic waves generated by an earth-surface-based transmitter and backscattered from the ground). Finally, possible medical applications must also be mentioned.

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References