Effect of absorption on temporal correlation of light scattered from a turbid medium

Serguei E. Skipetrov

Department of Physics, Moscow State University, 119899 Moscow, Russia

Received 19 January 1998; revised 19 March 1998; accepted 31 March 1998

Abstract

Theoretical analysis of the effect of absorption on the time autocorrelation function of light multiple-scattered in a turbid medium with large scale heterogeneous dynamics of scatterers is presented. A particular case of planar inclusion inside a slab filled with turbid medium is considered to demonstrate the main conclusions. © 1998 Elsevier Science B.V. All rights reserved.

PACS: 42.25.Bs; 82.70.Dd; 87.64.±t

Keywords: Turbid media; Diffusing wave spectroscopy; Absorption spectroscopy

1. Introduction

Diffusion approximation is currently shown to give sufficiently accurate results for the time autocorrelation function of light in a strong multiple-scattering regime [1–5]. Recent experimental and theoretical researches have demonstrated the applicability of the diffusion theory not only to macroscopically homogeneous samples but also to samples with large scale spatially varying dynamics [6–10]. In the latter case measurements of the field autocorrelation function \( G(r,\tau) = \langle E(r,t)E^\ast(r,t+\tau) \rangle \) (where \( \langle \rangle \) denotes time averaging for an ergodic system or ensemble averaging for a non-ergodic one) on the surface of a turbid sample allows to locate or “image” a hidden heterogeneous region of different particle dynamics (dynamic heterogeneity or inclusion for short) inside the sample. First demonstrated by Boas et al. [6] and then examined in more detail [7–10], this possibility was studied neglecting light absorption in the medium.

Absorption of light, however, is an intrinsic property of any medium and its effect is often enhanced in the multiple scattering limit because of the long diffusion paths of photons inside the scattering medium. Absorption reduces the contribution of the longer paths to the decay of the correlation function, as discussed in Refs. [4,5]. For macroscopically homogeneous samples it is easy to show that absorption just shifts the time scale of the time correlation function allowing one to apply mathematically the same analysis as in the case of no absorption [4,5].

In this paper we show that the effect of absorption on the time autocorrelation function of multiply scattered light in the case of a dynamically heterogeneous medium also appears to have a simple mathematical treatment. The theoretical analysis previously performed for dynamically heterogeneous media without absorption [6–10] is extended to the situation of turbid media with non-zero absorption coefficient. We discuss some new interesting features of \( G(r,\tau) \) introduced by the nonvanishing absorption coefficient inside and outside a dynamic heterogeneity and the way these features could be used in the analysis of experimental data. Expressions for the time autocorrelation functions of the depolarised scattered electric field of backscattered and transmitted light in the case of planar inclusion inside a turbid slab are derived and discussed.

E-mail: skipet@fort.phys.msu.su
2. General analysis

We consider a large sample of size $L$ filled with a turbid medium, such as a suspension of polystyrene beads of diameter $b - \lambda$, with $\lambda$ being the wavelength of light in the medium. The medium is characterised by the light $D_p^m$ and particle $D_p^m$ diffusion coefficients, the photon random walk step $l_{\text{in}}^o$, and the absorption coefficient $\alpha_{\text{in}}^o$. A region of characteristic size $d$ (an inclusion) inside the sample is assumed to have light scattering and absorption properties that differ from those of the surrounding medium, namely, $D_p^m$, $D_p^m$, $\mu^m_{\text{in}}$ and $\mu^m_{\text{in}}$. Sub- or superscripts “in” and “out” in our notation denote parameters corresponding to the medium inside and outside the inclusion, respectively. We assume small absorption ($\mu_{\text{in}}^m \ll \mu_{\text{out}}^m$, $\mu_{\text{out}}^m \ll \mu_{\text{in}}^m$), the photon diffusion coefficients $D_p^m$ and the photon random walk steps $l_{\text{in}}^o$ inside and outside the inclusion are related by $D_p^m = c l_{\text{in}}^o/3$ with $c$ being the speed of light in the medium. Instead of dealing with particle diffusion coefficients $D_p^m$ it appears convenient to introduce characteristic times $t_{\text{in}}^o$ describing decorrelation of light inside and outside the inclusion [2,3]: $t_{\text{in}}^o = (4k_0^2 l_{\text{in}}^o)^{-1}$, where $k_0 = 2\pi/\lambda$ is the wave number of light in the medium.

Within the weak scattering limit ($k_0 l_{\text{in}}^o \gg 1$, $k_0 l_{\text{out}}^o \gg 1$) and in the strong multiple scattering regime ($d \gg l_{\text{in}}^o$, $L \gg l_{\text{out}}^o$, $L - d \gg l_{\text{out}}^o$) throughout the sample the unnormalised time autocorrelation function of the depolarised scattered electric field inside and outside the inclusion $G_l(r,t) = \langle E(r)E^*(r,t) \rangle$ in the scalar approximation obeys the well-known steady-state diffusion equation [4,6]:

$$[\nabla^2 - \alpha_{\text{in, out}}^2(t)]G_l(r,t) = -S(r)D_p^m$$.

(1)

where $\alpha_{\text{in, out}}^2(t) = \frac{3\mu_{\text{in, out}}^m}{l_{\text{in, out}}^o} + \frac{\tau}{(2\alpha_{\text{in, out}}^m)^2 l_{\text{in, out}}^o}$ for the case of pure Brownian motion of scatterers and $S(r)$ describes the distribution of light sources inside the sample. Mathematical formulation of the problem is completed by the boundary conditions at the sample surface $S$ and at the surface $S_1$ of the inclusion [11]:

$$G_l(r,t) \equiv (2/3)l_{\text{in, out}}^o \nabla G_l(r,t) = 0, \quad r \in S,$$

$$G_l(r,t) = G_l^o(r,t), \quad r \in S_1,$$

$$D_p^m \nabla G_l^o(r,t) = D_p^m \nabla G_l^o(r,t), \quad r \in S_1.$$.

(2)

For further analysis it is convenient to write $\alpha_{\text{in, out}}^2(t)$ in the form

$$\alpha_{\text{in, out}}^2(t) = \frac{3(t + T_{\text{in, out}})}{2T_{\text{in, out}}^2 l_{\text{in, out}}^o},$$

(3)

where $T_{\text{in, out}} = 2\mu_{\text{in, out}}^m l_{\text{in, out}}^o$. It follows from Eq. (3) that the introduction of absorption ($T_{\text{in, out}} \neq 0$) shifts the arguments of $\alpha_{\text{in, out}}^2(t)$ on $T_{\text{in, out}}$, respectively. For a fixed spatial position $r$ of the detector at the sample surface, it means that if $T_{\text{in}} = T_{\text{out}} = T$, then the autocorrelation function of light emerging from a macroscopically heterogeneous absorbing sample is given by exactly the same expression as in the case of no absorption, but with $\tau + T$ substituted for $\tau$. In other words, the condition

$$\mu_{\text{in, out}}^m l_{\text{in, out}}^o = -\mu_{\text{out, in}}^m l_{\text{out, in}}^o$$

(4)

provides the applicability of the results obtained for nonabsorbing samples ($\mu_{\text{in, out}}^m = 0$) to the case of light absorption coefficients $\mu_{\text{in, out}}^m \neq 0$, provided that $\tau$ be replaced by $\tau + T$. In particular, theoretical analysis performed for spherical [6], planar [8] and cylindrical [9] dynamic heterogeneities can be also used if light scattering media inside and outside the inclusion possess absorption coefficients $\mu_{\text{in, out}}^m$ that obey Eq. (4).

In the case of $T_{\text{in}} \neq T_{\text{out}}$, however, the situation becomes more involved and cannot be reduced to a simple shift of the time scale. It appears, however, that for a special correlation time $\tau = \tau_0$, condition $\alpha_{\text{in}}^2(\tau) = \alpha_{\text{out}}^2(\tau)$ can be satisfied independently of the position, size and shape of the inclusion. From Eq. (3) we find

$$\tau_1 = \frac{T_{\text{in}} - p T_{\text{out}}}{p - 1} \quad \text{with} \quad p = \frac{\tau_{\text{in}}}{\tau_{\text{out}}}. \quad (5)$$

Using the definitions of $T_{\text{in, out}}$ and the relations between $l_{\text{in, out}}^o$ and $D_p^m$, we rewrite Eq. (5) as

$$\tau_1 = \frac{l_{\text{in}}^o}{l_{\text{out}}^o} \frac{\mu_{\text{in}}^m}{\mu_{\text{out}}^m} \frac{1}{D_p^m} l_{\text{in, out}}^o D_p^m l_{\text{out}}^o.$$.

(6)

Since $\alpha_{\text{in}}^2(\tau_1) = \alpha_{\text{out}}^2(\tau_1)$, the solution of Eq. (1) for $\tau = \tau_1$ has exactly the same numerical value for macroscopically homogeneous and heterogeneous cases independently of the position, size and shape of the heterogeneity. This somewhat unexpected point finds a simple physical explanation in the framework of the diffusion theory.

Indeed, Eq. (1) taken for $\tau = 0$ describes an average scattered intensity $G^s(r,0) = \langle |E(r)|^2 \rangle$ which depends on the absorption coefficients $\mu_{\text{in, out}}^m$, photon random walk steps $l_{\text{in, out}}^o$, as well as on the position, shape and size of the heterogeneity. Thus one gets different values of $G^s(r,0)$ for different absorption coefficients (and/or photon random walk steps) and sample geometries. Furthermore, the decay of $G^s(r,\tau)$ with time $\tau$ is controlled by $\tau_{\text{in, out}}^o$, sample geometry, and, again, by $\mu_{\text{in, out}}^m$ and $l_{\text{in, out}}^o$. The smaller the photon random walk step, the larger the number of scattering events at a fixed length of photon path in the medium, while the increase of absorption decreases the
effective lengths of photon paths that contribute to the decay of \( G_\tau(r, \tau) \).

Hence, for fixed parameters of random media inside and outside the inclusion, but for different sizes, shapes, and positions of the heterogeneous region, a set of \( G_\tau(r, \tau) \) curves is expected to be measured that consists of curves with different \( G_\tau(r, 0) \) values and different decay rates. The important consequence of our analysis is that all the curves will intersect at \( \tau = \tau_1 \), and that the value of \( \tau_1 \) is only controlled by \( l_{\text{in}, \text{out}}, D^\text{in}_{\text{out}}, \mu^\text{in}_{\text{out}}, k_0 \), and not by the sample geometry.

It must be emphasised that the above inference applies to the \textit{unnormalised} time autocorrelation function \( G_\tau(r, \tau) \). If each of \( G_\tau(r, \tau) \) is normalised to its own value at \( \tau = 0 \), the resulting curves will not coincide at any finite \( \tau \) apart from \( \tau = 0 \). Up to now, only the normalised time autocorrelation function has been used in experiments on imaging of dynamic heterogeneities in multiple-scattering media [6–10]. Normalisation does not reduce the sensitivity of the imaging techniques as long as the contrast between the hidden heterogeneity and the surrounding medium is purely dynamic (\( \mu^\text{in}_{\text{out}} = \mu^\text{out}_{\text{in}} \), \( l^\text{in}_{\text{out}} = l^\text{out}_{\text{in}} \)).

Once a difference in the absorption coefficients or/and photon random walk steps appears (\( \mu^\text{in}_{\text{out}} \neq \mu^\text{out}_{\text{in}} \) or/and \( l^\text{in}_{\text{out}} \neq l^\text{out}_{\text{in}} \)), additional information on the inclusion can be obtained from the values of \( G_\tau(r, 0) \) (average scattered intensities). In the limiting case of large difference between \( \mu^\text{in}_{\text{out}} \) and \( \mu^\text{out}_{\text{in}} \) (or \( l^\text{in}_{\text{out}} \) and \( l^\text{out}_{\text{in}} \)), there is no need to measure the correlation function at all: just the space-resolved measurements of the average scattered intensity allows for imaging of the inclusion [12,13].

In the present paper we discuss the situation when the medium inside a hidden inclusion differs from the surrounding medium in both the static scattering properties (absorption coefficient and photon random walk step) and the particle dynamics. This implies that combination of the techniques applied in the cases of absorbing/scattering and purely dynamic inclusions should be used. This line of reasoning leads to the conclusion that it is the unnormalised time autocorrelation function that could provide the most accurate information about the hidden inclusion in the case under consideration. Because of this, we are concerned with the unnormalised time autocorrelation function throughout the paper.

In the particular case of \( l^\text{in}_{\text{out}} = l^\text{out}_{\text{in}} = l^* \), Eq. (6) simplifies:

\[
\tau_1 = -\frac{l^*}{2k_0^2} \frac{\Delta \mu_s}{\Delta D_B},
\]

where

\[
\Delta \mu_s = \mu^\text{in}_{\text{out}} - \mu^\text{out}_{\text{in}} \quad \text{and} \quad \Delta D_B = D^\text{in}_{\text{out}} - D^\text{out}_{\text{in}}
\]

are the deviations of the light absorption and particle diffusion coefficients inside the inclusion from their values in the surrounding medium.

Hence, for known \( l^\text{in}_{\text{out}} = l^\text{out}_{\text{in}} = l^* \) and for a fixed wavelength \( \lambda \) the value of \( \tau_1 \) only depends on the ratio \( \Delta \mu_s/\Delta D_B \). This means that if a single inclusion is embedded inside an otherwise macroscopically homogeneous turbid sample and if \( l^\text{in}_{\text{out}} = l^\text{out}_{\text{in}} \) is known, one can determine the ratio \( \Delta \mu_s/\Delta D_B \) just by measuring \( \tau_1 \) as a correlation time for which \( G_\tau(r, \tau) \), measured at different positions \( r \) at the sample surface, intersect. Thus \( \Delta \mu_s/\Delta D_B \) can be found without particular knowledge of the inclusion position, size and shape.

### 3. Case of planar inclusion

To illustrate the above analysis we consider a particular case of planar inclusion of width \( d \) inside a slab of width \( L \). Inclusion is parallel to the slab surfaces and situated at a distance \( z \) from the front surface (see Fig. 1). Following Refs. [8,9] we assume an extended plane wave to be incident on the slab surface at \( z = 0 \) and write the source term \( S(\tau) \) in Eq. (1) as \( S_0 \delta(z - z^\text{in}_{\text{out}}) \). To solve Eq. (1) with the boundary conditions (2) for the considered geometry we use a general solution for the correlation function of light backscattered from or transmitted through a multilayer turbid sample obtained in Ref. [8]. Although the analysis of Ref. [8] is devoted to the case of negligible light absorption throughout the sample, it can be modified (as discussed in the previous section) to incorporate situations of absorbing media. This gives the following expressions for the time autocorrelation functions of the depolarised backscattered \( [G_\tau^0(\tau)] \) and transmitted \( [G_\tau^1(\tau)] \) light:

\[
G_\tau^0(\tau) = \frac{3S_0}{c} \frac{F(z - z^\text{in}_{\text{out}}, d, L - z - d)}{F(z, d, L - z - d)},
\]

\[
G_\tau^1(\tau) = \frac{3S_0}{c} \frac{\alpha^\text{in}_{\text{out}} \sinh(\alpha^\text{in}_{\text{out}} z^\text{in}_{\text{out}})}{\alpha^\text{in}_{\text{out}} \sinh(\alpha^\text{in}_{\text{out}} z)}
\]

\[
F(\xi_1, \xi_2, \xi_3) = \alpha^\text{in}_{\text{out}} \alpha^\text{in}_{\text{out}} \sinh(\alpha^\text{in}_{\text{out}} (\xi_1 + \xi_3)) + \left[ \alpha^\text{in}_{\text{out}} \alpha^\text{in}_{\text{out}} \cos(\alpha^\text{in}_{\text{out}} (\xi_1 - \xi_3)) \right] \sinh(\alpha^\text{in}_{\text{out}} (\xi_1 + \xi_3)) + \left[ \alpha^\text{in}_{\text{out}} \alpha^\text{in}_{\text{out}} \cos(\alpha^\text{in}_{\text{out}} (\xi_1 - \xi_3)) \right] \sinh(\alpha^\text{in}_{\text{out}} (\xi_1 + \xi_3)) \times \sinh(\alpha^\text{in}_{\text{out}} \xi_2).
\]

Here, for the sake of simplicity, we consider the case of

![Fig. 1. A sketch of the considered geometry. A layer of width \( d \) is embedded at a depth \( z \) inside a turbid slab of width \( L \).](image-url)
discussed in the previous section. In Fig. 2,\( G \)\( \text{ted}\) light has qualitatively similar behaviour in the sense that the value of \( \tau_1 \) is only determined by the ratio \( \Delta \mu / D_B \) as discussed in the previous section (see Eq. (7)).

4. Conclusion

In the present paper we discuss the statistics of time-dependent multiple-scattering speckles emerging from a turbid medium with large scale spatially varying dynamics and absorption. It is shown that this case can be treated in the framework of the diffusion theory similarly to the case of nonabsorbing samples. It follows from our analysis that if a single inclusion is embedded in an otherwise homogeneous sample, and once the condition \( \mu_n \tau_{in}^{out} = \mu_u \tau_{out}^{in} \) is satisfied, the unnormalised time autocorrelation function \( G(\tau) \) of scattered light measured at the sample surface should be equal to its value in the case of no absorption taken for a correlation time \( \tau + T \) with \( T = \mu_n \tau_{in}^{out} \). If the indicated condition is not satisfied, a special correlation time \( \tau_1 \) exists for which all \( G(\tau) \) have the same value independently of the position, size and shape of the inclusion. This correlation time is controlled by the light scattering and absorption properties of the media inside and outside the inclusion. In particular case of known \( l_{in}^{out} \) and \( l_{out}^{in} \) the value of \( \tau_1 \) is completely determined by the ratio \( \Delta \mu / D_B \), where \( \Delta \mu \) and \( D_B \) are the deviations of the absorption and particle diffusion coefficients inside the inclusion from their values in the surrounding medium.

The presented analysis can be modified to deal with the case of a spatially-localised, hidden laminar flow of scatterers inside a turbid sample. In this case \( a_n^2(\tau) = 3 \mu_n l_{in}^{out} + 3 \tau/(2 \mu_n l_{in}^{out}) + \sqrt{\tau^2/(4 \mu_n l_{in}^{out})} \) with \( \tau_1 \) being the characteristic time introduced by the flow [9]. In this case, however, one expects a more complex behaviour of the time autocorrelation function of the multiple-scattered light. If, for instance, a suspension flows inside a capillary embedded in a sample filled with a suspension having the same particle diffusion coefficient \( D_B = D_B^\text{inh} \), the same photon random walk step \( (l_{in}^{out})^2 = l_{out}^{in} \), but different absorption coefficient \( (\mu_n l_{in}^{out}) \), an intersection of \( G(\tau) \) curves, similar to the one discussed in the body of the text, will appear only for \( \Delta \mu = \mu_n l_{in}^{out} < 0 \), and we expect \( \tau_1 = \tau_1(-\Delta \mu l_{in}^{out}/2)^{1/2} \).

References