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The influence of the scattering anisotropy parameter on diffuse reflection of light

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Abstract

We discuss the impact of the scattering anisotropy parameter on the path length distribution of multiply scattered light. A quantitative treatment based on the radiative transfer equation is compared to an extended photon diffusion formalism. We compare both models with diffusing-wave spectroscopy measurements using randomly polarized light.

Keywords: Multiple scattering; Anisotropy; Diffusing-wave spectroscopy

1. Introduction

Probing diffuse light in the multiple scattering regime has become a very active field of research [1]. The backscattering regime is of particular importance for biomedical imaging techniques using visible or near-infrared diffuse light [2,3] and for the characterization of complex fluids [4]. At sufficiently large length scales (larger than the transport mean free path l^*) and at long times (larger than the collision time l^*/c, where c is the energy velocity), the transport of intensity is well described by the diffusion approximation [5]. The simplicity of the diffusion approximation makes it an important tool in the analysis of experimental data in practical situations. Nevertheless, it suffers from drawbacks which limit its validity. On the one hand, boundary conditions can only be introduced approximately, using extrapolation distances and angle-averaged reflection factors when internal reflections cannot be neglected [6,7]. On the other hand, the scalar diffusion approximation overestimates the contribution from short paths, the error becoming more severe as the anisotropy of scattering increases [8–10].

The purpose of this paper is to study experimentally and theoretically the diffuse reflection of randomly polarized light, and to show how the limits of the diffusion approximation can be overcome using improved models. Using randomly polarized light allows to focus on the impact of the scattering anisotropy parameter g. This parameter is defined as the average cosine of the scattering angle $g = \langle \cos \Theta \rangle$. The parameter is a direct measure of the scattering anisotropy. Predominant forward scattering, as observed in Mie scattering, leads to values close to 1 whereas for Rayleigh scattering $g = 0$. Negative values can be observed in the presence of short range structural order [11]. The value of g also defines the ration between the scattering mean free path ℓ and the transport mean free path $\ell^* = \ell/(1-g)$.

In this work, experimental data are obtained from diffusing-wave spectroscopy (DWS) measurements [12], an important tool in the characterization of complex fluids. DWS provides access to the distribution of light paths via the time correlation function of light fluctuations. The standard DWS theory relies on the diffusion approximation [13,14]. In the non-diffusive regime, the modelling

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can be improved by using more refined transport equations in order to compute the photon path-length distribution. The telegrapher equation has been used to study the effect of ballistic photons and scattering anisotropy [9]. More recently, the radiative transfer equation (RTE) has been introduced to model the transition from the single-scattering to the diffusive regime in DWS [15]. It has also been shown that measuring the path-length distribution, and using the measured data in the standard DWS theory (instead of calculations based on the diffusion approximation) improves the interpretation of the results [16].

In the present study, we show experimentally that for backscattered light, anisotropic scattering strongly influences the time decay of the field correlation function, a behavior which is not described by the standard DWS theory. Good agreement with the DWS data is obtained using an RTE calculation of the path-length distribution while the standard diffusion model display substantial deviations. Based on earlier work of Mackintosh and John [8], it has been suggested by Rojas et al. that in a reflection geometry, a cutoff in the path-length distribution can be introduced in order to improve the accuracy of the diffusion approximation [10]. The modified diffusion model significantly improves the description of DWS experimental data in the most relevant regime of intermediate time scales but gives unphysical predictions for extremely long paths.

2. Diffusing-wave spectroscopy

Dynamic light scattering allows the dynamic properties of complex media (such as biological tissues) to be characterized by measuring temporal fluctuations of scattered light [23,24]. In the multiple scattering regime, the DWS technique has been developed in order to relate light fluctuations to microscopic dynamic properties such as particle diffusion [12,13].

In the continuum approximation, the DWS normalized field correlation function $g_1(\tau)$ can be directly related to the path-length distribution $P(s)$ [13,14]

$$g_1(\tau) = \int_0^\infty P(s)\exp[-2(\tau/\tau_0)s/\ell^*]\mathrm{d}s. \tag{1}$$

In our case the characteristic relaxation time for diffusive particle motion $\tau_0 = (k_0^2 D)^{-1}$ (with D the diffusion constant of Brownian particles and k_0 the wave number in the medium) is a known quantity. The solution based on the scalar diffusion theory (non-reflecting boundaries) is well known [13]. For a non-absorbing medium, it reads

$$g_1(x(\tau)) = \left\{\exp[-\gamma_\mathrm{p}x(\tau)] + \exp[-(\gamma_\mathrm{p}+2\gamma_\mathrm{e})x(\tau)]\right\}/2 =: h(x(\tau)) \tag{2}$$

where $x(\tau) = \sqrt{6\tau/\tau_0}$, $\gamma_\mathrm{p} = z_\mathrm{p}/\ell^*$ and $\gamma_\mathrm{e} = z_\mathrm{e}/\ell^*$ with $\gamma = \gamma_\mathrm{p} + \gamma_\mathrm{e} = 5/3$ for scalar waves [5]. In the limit $x \ll 1$ this expression reduces to $g_1(x) \simeq \exp(-\gamma x)$. It is very common to use this expression when modelling experimental data (even outside the range of validity of the first-order development in x). In the derivation of Eq. (2) both residual absorption and limited container size have been neglected. Both processes lead to a loss of photons for long paths (for a detailed discussion see [24]).

The key quantity in DWS is the path-length distribution $P(s)$. The scalar diffusion approximation $[P_\mathrm{diff}(s)]$ is known to overestimate the contribution from the short paths of the distribution, the error becoming more and more severe as the anisotropy of scattering increases. Rojas et al. suggested a semi-empirical correction to the distribution of paths length by introducing a g-dependent cutoff in the distribution [10]

$$P_\mathrm{corr}(s) \propto P_\mathrm{diff}(s)[1 - 3/2g\exp(-s/\ell^*)], \tag{3}$$

where $P_\mathrm{diff}(s)$ is the distribution calculated from scalar diffusion. For a quantitative description, this distribution has to be normalized to satisfy the condition $\int_0^\infty P_\mathrm{corr}(s)\mathrm{d}s = 1$. It is worthwhile to point out that in the previous work of Rojas-Ochoa et al. [10] the modified path distribution has been used to study the polarization dependence of diffusely reflected light [10]. The empirical relation found between the depolarization length ℓ_p and the scattering mean free path ℓ has been confirmed in a subsequent theoretical study of Xu and Alfano [25].

The modified path-length distribution given by Eq. (3) can be easily included in the DWS framework. It follows directly from Eqs. (1)–(3):

$$g_1(x(\tau)) = \frac{2h[x(\tau)] - 3gh\left[\sqrt{x(\tau)^2+3}\right]}{2 - 3gh[\sqrt{3}]}. \tag{4}$$

3. Modelling by the radiative transfer equation

The accuracy of the computation of the path-length distribution $P(s)$ can be improved by using more refined transport equations for the light intensity (compared to the diffusion equation). In this work, we will use the time-dependent RTE. The use of the RTE allows to avoid a number of fundamental and practical drawbacks of the diffusion approximation. (i) The validity of the RTE is not restricted to large systems and long times. (ii) The boundary conditions can be handled exactly with the RTE approach [17]. This is particularly important in systems where internal reflections play a crucial role [18,19]. (iii) The RTE allows to deal with arbitrary scattering properties and an arbitrary level of absorption [20].

In this work, we consider a semi-infinite medium with the z-axis normal to the boundary. The medium $z > 0$ is filled with a scattering medium, and illuminated from the left at normal incidence by a plane wave. The specific intensity $I(z,\mu,t)$ inside the scattering medium obeys the RTE [21]

$$\frac{1}{c}\frac{\partial I(z,\mu,t)}{\partial t} + \mu\frac{\partial I(z,\mu,t)}{\partial z} = -(\mu_s+\mu_a)I(z,\mu,t) + \frac{\mu_s}{2}\int_{-1}^{+1} p^{(0)}(\mu,\mu')I(z,\mu',t)\mathrm{d}\mu', \tag{5}$$

where c is the energy velocity and $\mu = \cos\theta$, with θ the angle between the propagation direction and the z-axis. $p^{(0)}$ is the phase function averaged over the azimuthal angle $p^{(0)}(\mu,\mu') = (2\pi)^{-1}\int_0^{2\pi} p(\mathbf{u}\cdot\mathbf{u}')\mathrm{d}\phi$ where $\mathbf{u}$ and $\mathbf{u}'$ are unit vectors corresponding to directions (θ,ϕ) and (θ',ϕ'). The specific intensity $I(z,\mu,t)$ is integrated over the azimuthal angle ϕ. μ_s and μ_a are the scattering and absorption coefficients, respectively. The associated scattering and absorption mean-free paths are $\ell = \mu_s^{-1}$ and $\ell_a = \mu_a^{-1}$. The transport mean-free path is $\ell^* = \ell/(1-g)$. The real part of the medium effective index, accounting both for the homogeneous background medium and the scattering particles, is denoted by n.

In order to compute the field correlation function given by Eq. (1), one could compute the path-length distribution $P(s)$ using the RTE, and then perform the integration numerically. The calculation of $P(s)$ can be deduced from a calculation of the time-resolved diffuse reflected intensity $\phi_{\mathrm{RTE}}(t)$, resulting from an illumination with an incident pulse of negligible width. Assuming a constant velocity, one simply has $P(s) = \phi_{\mathrm{RTE}}(t = s/c)/(cU_{\mathrm{inc}})$, where U_{inc} is the energy density of the incident pulse. In this work, we have followed a different approach, taking advantage of the Laplace-transform structure of Eq. (1). Instead of solving the RTE for the specific intensity $I(z,\mu,s = ct)$, we solve the transport equation satisfied by the Laplace transform of the specific intensity with respect to the variable s

$$\mathscr{I}(z,\mu,\tau) = \int_0^\infty I(z,\mu,s)\exp[-2(\tau/\tau_0)s/\ell^*]\mathrm{d}s. \tag{6}$$

This quantity obeys an equivalent steady-state RTE with an effective extinction coefficient

$$\mu\frac{\partial\mathscr{I}(z,\mu,\tau)}{\partial z} = -\left(\mu_s + \mu_a + \frac{2\tau}{\tau_0\ell^*}\right)\mathscr{I}(z,\mu,\tau) + \frac{\mu_s}{2}\int_{-1}^{+1} p^{(0)}(\mu,\mu')\mathscr{I}(z,\mu',\tau)\mathrm{d}\mu'. \tag{7}$$

This equation can be solved numerically using standard procedures for the steady-state RTE. In the present study, we have used a discrete–ordinate scheme [22]. Details about the numerical method can be found in Ref. [18]. From the Laplace transform of the specific intensity $\mathscr{I}(z,\mu,\tau)$, the field correlation function $g_1(\tau)$ is readily obtained by computing the radiative steady-state energy flux at the boundary $z = 0$, which is simply the first-moment of the specific intensity with respect to the angular variable μ

$$g_1(\tau) = \int_{-1}^{+1}\mu\mathscr{I}(z=0,\mu,\tau)\mathrm{d}\mu. \tag{8}$$

4. Experiments and results

In our experiments we study laser light (wavelength $\lambda = 532$ nm) back reflected from a highly opaque "white" medium. The sample cell (10×5 mm base dimensions) is filled with a suspension of monodisperse polystyrene latex particles (refractive index $n = 1.59$) in water, with a volume fraction of 4%. The cuvette is immersed in a water bath to both keep the temperature constant at $T = 22\,^\circ$C and suppress reflections at the cuvette wall. We vary the diameter of the particles in the range $d = 110$–990 nm, with corresponding anisotropy factors varying in the range 0.1–0.95. The scattered light is measured in the backscattering direction. Typical measurements of the field time correlation function ($\ln g_1(x)$) are shown in Fig. 1.

In order to study the influence of anisotropic scattering we define an effective time decay parameter γ_{eff} by

$$\gamma_{\mathrm{eff},x'} = -\left.\frac{\partial[\ln g_1(x)]}{\partial x}\right|_{x=x'}. \tag{9}$$

We obtain the experimental values of $\gamma_{\mathrm{eff}}(x')$ by a linear fit to $\ln g_1(x)$ in the range $x = 0.15$–$0.25 \simeq 0.2$ and $x = 0.45$–$0.55 \simeq 0.5$. The linear fits on the experimental data are shown in Fig. 1 (solid lines).

The RTE calculations are carried out matching the experimental conditions. The phase function and mean free paths are computed using Mie theory in the independent scattering regime (low volume fraction of particles). Fig. 2 shows the measured γ_{eff} (symbols), as well as calculations using the different models for $g_1(\tau)$: DWS with corrected path-length distribution $P(s)$ (dash-dotted line) and with RTE calculation of $P(s)$ (solid line) as well as the constant value of γ_{eff} expected from the standard diffusion theory (dotted line). For comparison we have also included the predictions by Lemieux et al. (dashed line) which are based on a photon propagation model using a telegrapher equation approach [9]. The latter model explicitly takes in to account g and further provides analytic predictions for g_1 [Eq. (4.12) in Ref. [9]]

For values of $x \geqslant 0.2$ the experiments and the RTE calculations show a clear dependence on the anisotropy

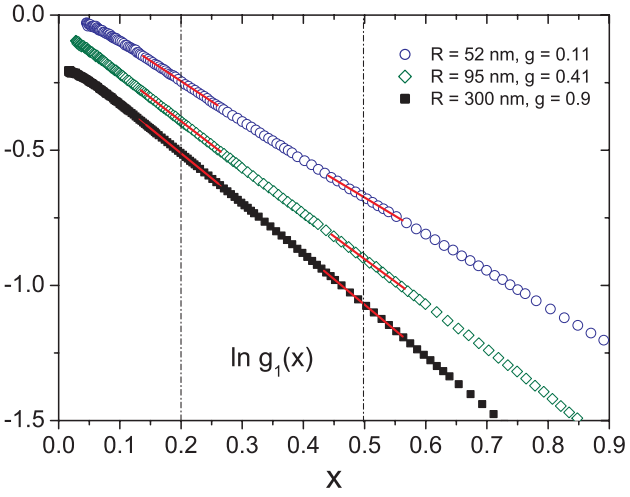

Fig. 1. DWS autocorrelation function for different g-values. Curves for $g = 0.41$, 0.9 are shifted for clarity. Solid lines: Linear fit to the data over the range $x = 0.15$–$0.25 \simeq 0.2$ and $x = 0.45$–$0.55 \simeq 0.5$.

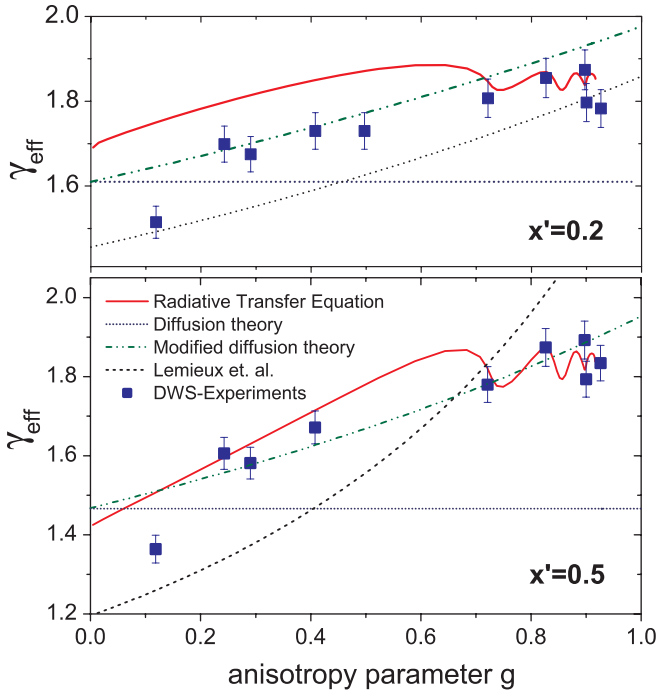

Fig. 2. Decay parameter γ_{eff} for backscattered light versus the anisotropy factor g. Symbols: experimental data. Solid line: DWS with RTE calculation of $P(s)$, Dash-dotted line: DWS with corrected path-length distribution $P_{corr}(s)$, Dotted line: standard DWS with $P_{diff}(s)$. Dashed line: predictions based on the model of Lemieux et al. [9].

factor g. The effective decay rate γ_{eff} increases with g. This behavior, which is not included in the standard DWS model Eq. (2), is well described by the RTE calculations. Also the modified diffusion model captures the essential features of the observed behavior. In particular at intermediate values of x the agreement is excellent. In practical applications of DWS this good agreement should turn out very beneficial. The region $x = 0.3$–0.7 is usually analysed in a DWS experiment. Smaller values are difficult to access since for $x < 0.2$ photon loss (absorption and/or limited container size) is often dominant whereas values of $\ln g_1(x)$ for $x > 1$ are not very reliable due to experimental noise (the measured signal has decayed to less than two per cent) and the contribution from very short paths. The model of Lemieux et al. [9] deviates substantially from the data in this regime of practical interest ($x = 0.3$–0.7). Deviations are smaller as x is decreased. Finally, we note that for values of $g > 0.7$, the RTE calculation displays oscillations. These oscillations are the signature of Mie resonances in the spherical particles.

Approaching $x \to 0$ the results are expected to become g independent which is reflected by the standard diffusion model, the telegrapher equation model of Ref. [9] as well as our RTE calculations (data not shown). Physically these long paths are well modelled by the diffusion approximation and in the limit of $P(s \to \infty)$ the distribution cannot be affected by the scattering anisotropy. In this regime the modified diffusion equation displays an artefact: While the overall agreement is still reasonable the remaining g-dependence of γ_{eff} is unphysical (data not shown).

5. Summary and conclusions

We have analysed the diffuse reflection of light based on DWS measurements. The time decay of the field correlation function substantially depends on the scattering anisotropy parameter g, a behavior that is not predicted by the standard DWS theory. A rigorous treatment based on the RTE describes the g dependence, in good agreement with the experimental data. An *ad hoc* modification of the path-length distribution of the standard DWS approach, using a g-dependent cutoff in the path-length distribution derived from the diffusion approximation, has been checked against experiments. It provides a useful simple approach for practical applications. Our results should help improving optical probes for soft matter studies and imaging in biological tissues.

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