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Incoherent light transport in anisotropic media: Form factor influence for oriented prolate ellipsoids

Nadjim Moumini, Christophe Baravian *

Laboratoire d'Energétique et de Mécanique Théorique et Appliquée, Nancy University, CNRS–UMR 7563. 2, avenue de la forêt de Haye, B.P. 160 F-50504 Vandoeuvre Cedex, France

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ABSTRACT

In turbid media, the partial or global orientation of anisotropic particles induces anisotropic light transport. In this study, we discuss the anisotropic incoherent transport of light in media where prolate ellipsoids are oriented in the same direction. In these anisotropic media, incoherent light transport is investigated using Monte Carlo simulations where the influence of particle anisotropy, size and optical properties are explored in a systematic way, from the local scattering event up to the diffusion limit. The database allows inverting the anisotropy of the backscattered image to yield the form factor of the particles. We then illustrate the relevance of such an analysis to assess the deformability of human erythrocytes in blood samples under normal physiological conditions.

1. Introduction

In the last decade, numerous light scattering techniques have been developed for characterizing strongly scattering concentrated suspensions. Such techniques provide valuable insight on the microscopic properties of the concentrated suspensions from their spatial or temporal optical diffusive properties.

In isotropic random media, average particle size can be determined using diffusive wave spectroscopy techniques even in the limit of strong multiple scattering [1,2]. Alternatively, coherent backscattering techniques can also be used to derive optical diffusive properties [3]. Other techniques, based on incoherent light transport, yield the particle size or the volume fraction of particles [4–8], whereas both parameters can be obtained simultaneously using polarization transport [9]. Light-based techniques are very complementary to X-ray or neutron scattering methods, since they apply to particle sizes typically larger than a few tens or hundreds of nanometers, whereas small angle X-ray scattering (SAXS) [10] or small angle neutron scattering (SANS) [2] can be successfully applied to determine in a non-intrusive way the structure of concentrated suspensions, for particle sizes that are typically smaller than 100 nm. In the case of anisotropic media, SAXS and SANS are particularly useful as they provide information on both particle morphology and global orientation [11–13]. However, at present, very few techniques can be used for obtaining such an information for suspensions with larger particle sizes. The aim of the present paper is to propose a novel approach to investigate the anisotropy of concentrated suspensions using incoherent light transport. In order to obtain information about the morphology and/or orientation field of particles in concentrated dispersions, we first try to understand light transport in the diffusion limit in strongly scattering anisotropic media.

* Corresponding author. Tel.: +33 3 83 59 57 27.
E-mail address: christophe.baravian@ensem.inpl-nancy.fr (C. Baravian).

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A few experiments have already addressed light scattering in concentrated suspensions of anisotropic particles either for well oriented anisotropic particles under an external field (shear for instance) [4,14] or for partially oriented anisotropic particles. This was for instance the case of nematic liquid crystals where an anisotropic coherent backscattering cone [15] or anisotropic incoherent light transport properties have been reported [16–18]. Other experiments have dealt with human tissues such as dentin [19,20], skin [21], muscles [22] or red blood cells suspensions [14]. In this latter case, an anisotropic backscattered image was observed upon application of shear to red blood cells suspensions at physiological concentration (40–50% in volume). In other recent studies, the damaging of high-density polyethylene during tensile tests was followed by light scattering techniques [4]. The appearance of an anisotropic backscattered spot was linked to the formation and subsequent deformation of cavities oriented along the tensile axis [16,19,20,22,23]. Although the real particle morphology could not be deduced from the observed patterns [14].

As far as the theory of incoherent light transport in anisotropic random media in the diffusive limit is concerned, relatively few studies have addressed the question. Berdnik and Loiko [24] studied light scattering in media composed by spheroidal particles oriented along the same direction and Kienle et al. [25] convincingly showed that a diffusion equation (inspired from [26] in isotropic media) is not correct for describing incoherent light transport in anisotropic random media. It therefore appears relevant to perform Monte Carlo simulations to mimic the medium. Indeed, Kienle et al. [25] proposes three different approaches of the problem to compare their performances. In the first one a diffusion equation with anisotropic diffusion coefficients is written. The second approach consists in a Monte Carlo simulation of photon propagation in a mixture of oriented cylinders and spherical particles; the anisotropy of the photon transport being introduced through anisotropy of the scattering cross section. The third approach uses the real interaction between the electromagnetic wave and the oriented cylinders. The main conclusion of this comparative stuffy is that the diffusion approximation significantly differs from Monte Carlo results, and that the difference increases with medium anisotropy. Writing a diffusion equation to model anisotropic incoherent transport in the diffusion limit is therefore clearly not straightforward, although in some cases good agreements were obtained with experimental data [23]. Another approach to the problem is to develop a diffusion approximation on the basis of radiative transfer equations. Along such lines, Heino et al. [27] proposed an anisotropic diffusion approximation to resolve the radiative transfer equation. They assumed the media to be composed of oriented infinite cylinders. After derivation of the diffusion approximation, an anisotropic diffusion equation is obtained. This equation is then resolved by Monte Carlo simulations, based on the radiative transfer model by both finite elements and boundary element method, which basically leads to similar solutions. Even if a purely diffusive approach appears as a viable option for describing anisotropic media, some open questions remain about the nature and position of the diffusive source inside the medium as well as on the appropriate evaluation of the anisotropic diffusion coefficients, especially in steady-state experiments [18,23].

The present paper essentially focuses on anisotropic media where all particles are oriented in the same direction and progressively deformed, as in the case of concentrated red blood cells suspensions. By orienting all particles in the same direction, the influence of the order parameter on photon transport vanishes and photon transport then depends only on the particle form factor. Our aim is to obtain the real spheroidal shape of the particles from the analysis of the backscattered light pattern. To reach such a goal, we first develop a full Monte Carlo simulation database by systematically exploring the influence of varying particle size, optical refractive index and form factor.

In the first part, we present the interaction between an electromagnetic wave and a prolate ellipsoidal particle. The second part describes the Monte Carlo method used to resolve incoherent light transport. Finally, by analyzing the Monte Carlo simulations, the anisotropy of the incoherent backscattered light is compared to the form factor of particles for various refractive indexes ratios and sizes. These results are used to obtain the shear modulus of red blood samples under normal physiological conditions.

2. Plane electromagnetic wave interaction with an ellipsoidal particle

Light transport is described starting from the local scattering event. When photons enter a collection of particles, they experience many scattering events before being eventually backscattered by the medium. Therefore, in this section, we first describe the scattering event corresponding to the interaction between a plane electromagnetic wave and a single particle before looking at the complete interaction with a collection of particles. We will only consider the spheroidal prolate shape.

Various numerical techniques have been developed for many years [28–33] for solving the problem of electromagnetic scattering by non-spherical particles. All these techniques try solving Maxwell equations for the interaction between a non-spherical particle with an equivalent spherical radius r_v, an optical refractive index n_p, a form factor $\varepsilon = a/b$ (a minor axis and b major axis) and a plane electromagnetic wave (wavelength λ) in a surrounding medium with optical refractive index n_m. Various calculation techniques have then been proposed as for instance, the separation of variable method (SVM) introduced by Asano and Yamamoto [28,29] or the discrete dipole approximation (DDA) proposed by DeVoe [34] and subsequently reviewed by Draine and Flatau [30]. The *T*-matrix code used in this study was developed by Mischenko to calculate the interaction between a non-spherical particle and an electromagnetic wave. This code is based on the null field method (NFM) introduced by Waterman [35] and reviewed by Mishchenko [31–33,36–38]. Such an approach involves three parameters only, the size parameter $x = 2\pi r_v n_m/\lambda$,the refractive index ratio $m = n_p/n_m$ and the aspect ratio of the particle ε.