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Time-resolved light transport in structurally anisotropic media

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ABSTRACT

Structurally anisotropic materials are ubiquitous in several application fields, yet their accurate optical characterization remains challenging due our incomplete understanding of how anisotropic light transport properties arise from the microscopic scattering coefficients. In fact, even when the dynamics of light transport is directly measured, coarse simplifications are often introduced due to a lack of established theoretical models or numerical methods. Here, we apply a general Monte Carlo implementation capable of handling direction-dependent scattering to the analysis of light transport in a sample of polytetrafluoroethylene (PTFE) tape. Using only a set of transient transmittance intensity profiles, the analysis retrieves the tensor components of the diffusive rates and the scattering coefficients along all three directions, in excellent agreement with Monte Carlo simulations.

Keywords: multiple scattering, time-resolved transmittance, anisotropic diffusion, scattering tensor, optical gating, fibrous materials

1. INTRODUCTION

Diffusion processes are often described using random walk models. For materials where the scattering properties do not depend on the direction of propagation, a well-known relation exists between the transport mean free path ℓ^* and the (experimentally observable) diffusive constant D, expressed as $D = v\ell^*/3$ with v as the energy velocity in the medium. At the microscopic level, the transport mean free path is related to the scattering mean free path ℓ (defined as the inverse of the scattering coefficient $\ell = 1/\mu_\mathrm{s}$) by a so-called "similarity relation" $\ell^* = \ell/(1-g)$, which depends on the asymmetry factor g representing the average cosine of the scattering angle relative to the incoming direction. Therefore, in the context of isotropic diffusion, the relation between the macroscopic observable diffusion rate and the microscopic scattering property can be written as:

$$D = \frac{1}{3}v\ell^* = \frac{1}{3}\frac{v}{\mu_\mathrm{s}'} = \frac{1}{3}\frac{v}{\mu_\mathrm{s}(1-g)}, \tag{1}$$

where μ_s' is the reduced scattering coefficient. This relation allows to infer directly microscopic structural information of a scattering material from the experimentally observed diffusivity of light.

This simple connection breaks down, unfortunately, for anisotropic materials. In this case, all parameters of interest become 3×3 tensor quantities referred to a fixed reference frame, including $\boldsymbol{D}$, $\boldsymbol{\mu}_\mathrm{s}$ $\boldsymbol{g}$ and $\boldsymbol{v}$. In many

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cases, a reference frame can be found where all these tensors are diagonal, and similar relations can in principle be defined for the scattering and transport mean free paths as:

$$\boldsymbol{\mu}_{\mathrm{s}} = \begin{pmatrix} \mu_{\mathrm{s},x} & 0 & 0 \\ 0 & \mu_{\mathrm{s},y} & 0 \\ 0 & 0 & \mu_{\mathrm{s},z} \end{pmatrix} = \begin{pmatrix} \frac{1}{\ell_x} & 0 & 0 \\ 0 & \frac{1}{\ell_y} & 0 \\ 0 & 0 & \frac{1}{\ell_z} \end{pmatrix}, \tag{2}$$

and

$$\boldsymbol{D} = \begin{pmatrix} D_x & 0 & 0 \\ 0 & D_y & 0 \\ 0 & 0 & D_z \end{pmatrix} = \begin{pmatrix} \frac{1}{3}v\ell^*_x & 0 & 0 \\ 0 & \frac{1}{3}v\ell^*_y & 0 \\ 0 & 0 & \frac{1}{3}v\ell^*_z \end{pmatrix}. \tag{3}$$

However, these quantities are no longer related to each other via simple analytical relationships, and the diffusive constant along a certain direction will not depend solely on the scattering properties along the same direction. The non-trivial interdependence between the observed diffusion rate along a given direction and the microscopic scattering parameters prevents their accurate retrieval.[1, 2] The lack of a comprehensive theory for the derivation of the anisotropic diffusion tensor exacerbates this issue, requiring their study via numerical methods.

In the following, we present experimental data for the time-resolved transmittance through a highly anisotropic medium, with the aim of measuring its diffusive tensor components and retrieving the corresponding scattering coefficients. This result is obtained by combining an optical gating experimental technique[3] with a newly developed implementation of anisotropic light transport, which has been integrated into the open-source Monte Carlo Python package PyXOpto.[4]

2. METHODS

2.1 Experimental setup

The experimental setup is based on a Ti:Sa laser source and a parametric oscillator producing two near-infrared synchronous trains of pulses at 820 nm and 1525 nm, with a 80 MHz repetition rate and a typical duration of about 150 fs (Fig. 1a). In the experiment, a motorized delay line is present on one of the two arms of the setup, which can either assume the role of the probe or the gate arms. The probe beam is focused on the sample to generate the diffused transmittance, which is then recombined with the collimated gate beam onto a 2 mm-thick β-barium borate (BBO) crystal. A sum-frequency signal is generated at 533 nm, proportional to the cross-correlation between the diffuse transmittance and the gate pulse at a given delay, which can be controlled with µm precision (corresponding to time steps of a few fs) using the motorized stage.

The resulting time-resolved signal can be then either integrated by a photomultiplied detector, or resolved spatially with a CCD camera recording the transient intensity profiles transmitted through the scattering medium. Thanks to the spatially uniform upconversion efficiency guaranteed by the expanded and collinear gate beam, the generated images provide quantitative information on the transverse propagation light on the exit surface of the sample, from which the diffusive rates along different directions can be measured directly.

2.2 Data analysis and Monte Carlo simulations

We measure transmitted intensity profiles at different time delays, as exemplified in Fig. 1b. Before analyzing them, the frames are rotated for convenience so that the observed diffuse profile has its main axes aligned to the laboratory reference frame. Then, the individual intensity profiles are fitted with bi-variate Gaussian distributions to retrieve the evolution of its Mean Square Displacement (MSD) or variance along the y (vertical) and x (horizontal) axes. The spread rate of the intensity profiles provides direct access to the x and y diffusion rates, independently of the presence of absorption. Thus, performing a linear regression on the MSD evolution after the initial transient allows to retrieve the diffusive constants D_x, D_y or, equivalently, their corresponding transport mean free path ℓ^*_x, ℓ^*_y. The spatially-integrated time-resolved curve completes the 3D picture by providing information on the diffusion rate along the depth direction z, and on the absorption coefficient μ_{a} (which we assume to be a scalar quantity).

In order to retrieve the microscopic scattering coefficients associated to the observed diffusive rates, the experimental MSD and time-resolved transmittance data are simultaneously fitted with a single anisotropic MC

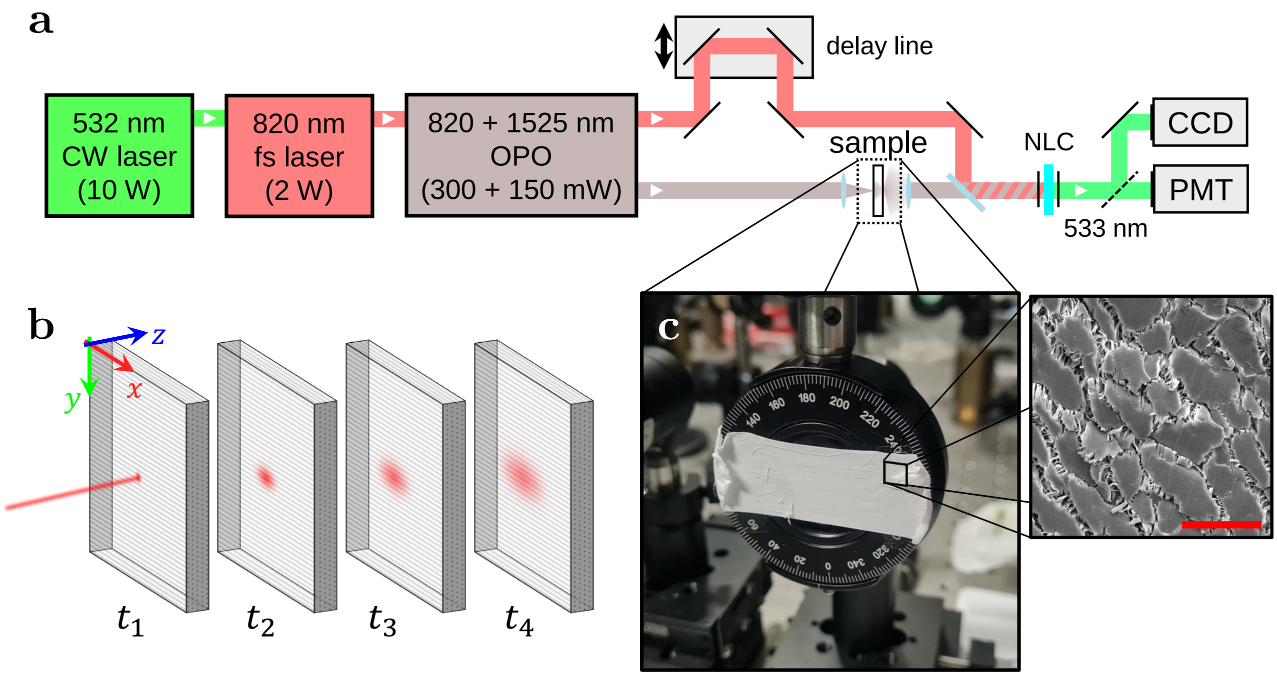

Figure 1: (a) Sketch of the experimental setup for time-resolved measurements. (b) Schematics of the transient imaging measurement on a slab sample. The spatial distribution of transmitted intensity is imaged at different time delays. In presence of anisotropic transport, the intensity distribution is an evolving ellipsoid. (c) Picture of the PTFE tape sample on a rotation mount with an insert of a scanning electron microscope image. Scale bar corresponds to 30 µm.

simulation. To the best of our knowledge, despite the prevalence of anisotropic materials, up to date there are no openly available MC software tools capable of handling anisotropic transport. To this end, we implemented a general description of anisotropic transport into the open-source MC Python package PyXOpto, which allows to generate spatio-temporal intensity distributions that can be directly compared to the experimental data, or analyzed in terms of their instantaneous MSD. In the simulations, a general anisotropic material can be described by the 3 independent components of the scattering tensor $\mu_{s,x}$, $\mu_{s,y}$ and $\mu_{s,z}$ and/or 3 asymmetry factors g_x, g_y and g_z in the framework of the Henyey-Greenstein phase function.

3. RESULTS

We illustrate our experimental and numerical analysis of anisotropic light transport using a strip of Teflon tape commonly used for sealing pipe threads. Teflon tape is made by polytetrafluoroethylene (PTFE) fibers exhibiting a preferential alignment, which endow it with a marked structural anisotropy. This anisotropy affects also light transport properties, and indeed anisotropic diffusion of light in PTFE has been already observed at different wavelengths.[5,6] However its actual scattering tensor components are still unknown. We measure the transmittance through a free-standing strip of tape at a probe wavelength of 1525 nm. The tape sample is attached on a rotating mount, to allow its alignment with the laboratory reference frame (Figure 1c). All measurements are averaged over multiple sample positions in an area of $\sim 4\,\mathrm{mm}^2$, to reduce the effect of possible inhomogeneities. This operation also allows to average out the speckle pattern, leaving only the incoherent diffused intensity profile. A preliminary set of measurements was performed at a fixed delay and different rotation angles of the samples to verify that a corresponding rotation was observed for the elongated transmittance profile, allowing to rule out the presence of direction-dependent biases in the non-linear up-conversion imaging process. This analysis confirmed the presence of a direction of faster light diffusion at an angle of ∼45° with respect to the direction of the tape strip.

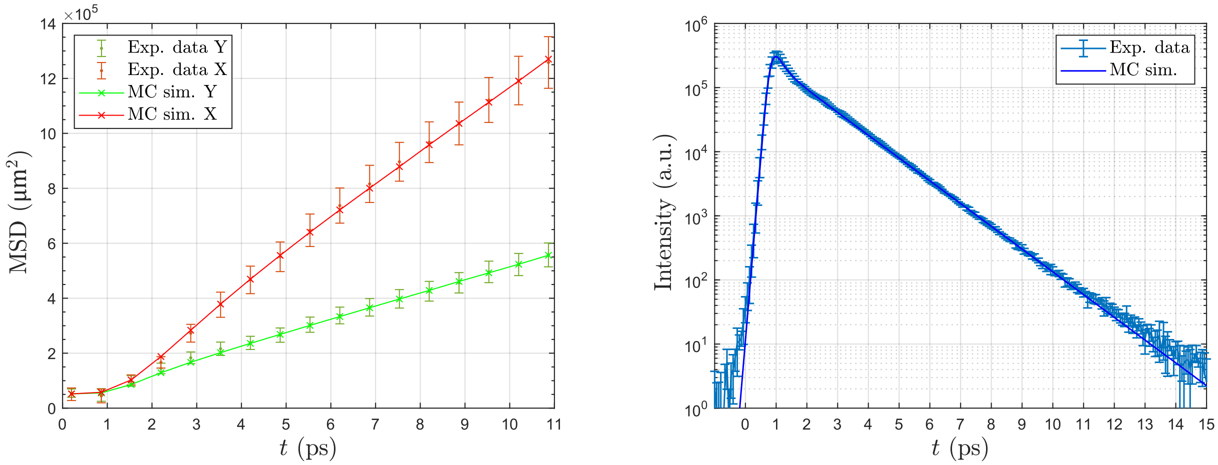

Figure 2: Mean square displacement (left) and time-resolved transmittance (right) data of Teflon tape. Solid lines represent the best MC fit, obtained with: $\mu_{\mathrm{s},x} = 48\,\mathrm{mm}^{-1}$, $\mu_{\mathrm{s},y} = 105\,\mathrm{mm}^{-1}$, $\mu_{\mathrm{s},z} = 77\,\mathrm{mm}^{-1}$, $g = 0.9$, $\mu_{\mathrm{a}} = 0$.

This angle corresponds to the direction perpendicular to the alignment of the PTFE fibers, as determined by scanning electron microscopy images, in agreement with previous reports.[5,6] Closer inspection of the electron microscopy picture shows that the polymer fibers have a flattened shape along the main plane of the tape, and are fused together to form planar scales on its external surface. This morphology suggests that different scattering coefficients could be found along each direction, resulting in a fully anisotropic sample.

The main set of measurements was performed in a fixed orientation configuration, rotated so that the faster axis of diffusion coincides with the x axis in the laboratory reference frame. We assumed an effective refractive index of $n = 1.05$ based on the PTFE/air volume fraction estimated by a weighting method, and a thickness of $L = 200\,\mu\mathrm{m}$ as declared from the tape manufacturer. The in-plane diffusive constants can be derived directly with a linear regression on the MSD evolution (Fig. 2a), returning values of $D_x = (289 \pm 15) \times 10^2\,\mu\mathrm{m}^2\,\mathrm{ps}^{-1}$ and $D_y = (117 \pm 5) \times 10^2\,\mu\mathrm{m}^2\,\mathrm{ps}^{-1}$.

In order to limit the number of free parameters, we assumed for simplicity a direction-independent asymmetry factor g, while the absorption coefficient μ_{a} was independently measured to be consistent with zero. The resulting best-fit MC simulation is in excellent agreement with both the time-resolved transmittance and the MSD evolution (Figure 2), closely matching also fine experimental features such as the more prominent transmittance peak at $t \sim 1\,\mathrm{ps}$ or the slight slope change of the fast-diffusing axis after $t \sim 5\,\mathrm{ps}$.

Even though the similarity relation is not expected to hold in the anisotropic case, it may still be convenient to evaluate an "effective" transport mean free path $\tilde{\ell}_i^*$, or equivalently an "effective" reduced scattering coefficient $\tilde{\mu}'_{\mathrm{s},i}$, by applying a similarity relation along each direction

$$\tilde{\ell}_i^* = \frac{1}{\tilde{\mu}'_{\mathrm{s},i}} = \frac{1}{\mu_{\mathrm{s},i}(1 - g_i)}, \tag{4}$$

for $i = \{x, y, z\}$. This quantity provides also a useful initial guess for the MC fitting procedure. In fact, for an asymmetry factor ~ 1, a small uncertainty on g can translate into a very large uncertainty on the scattering coefficients. Conversely, the effective transport mean free path is more stable against small variations of g, and also facilitates the comparison with the actual diffusive rates expressed in terms of transport mean free paths ℓ_i^*.

We obtain effective transport mean free paths values of $\tilde{\ell}_x^* = (210 \pm 20)\,\mu\mathrm{m}$, $\tilde{\ell}_y^* = (95 \pm 7)\,\mu\mathrm{m}$, $\tilde{\ell}_z^* = (130 \pm 14)\,\mu\mathrm{m}$, which remain largely unmodified even when changing the value of g around its fitted value of 0.9. It is interesting to note that the ratio of the diffusive constants $D_x/D_y = 2.47$ is different from the ratio $\tilde{\ell}_x^*/\tilde{\ell}_y^* = 2.21$, which shows that the direct observation of the macroscopic diffusion process is not sufficient to assess the microscopic anisotropy due to the non trivial mixing of the scattering along different directions, in contrast with previous published results.[7]

At this stage, the fitted value of the asymmetry factor should be considered only a coarse estimation. A more complete fit should also consider anisotropy in the asymmetry factor, even though the fitted value ($g = 0.9$) is probably representative of the most relevant asymmetry contribution (i.e., along the fast diffusing axis). As a prospect, it can be expected that any attempt to fit a model with 7 (or more) free parameters would also correspondingly require independent measurements of additional observables than the ones considered here.

4. DISCUSSION

In this work, we presented optical gating imaging as a robust and versatile method for the study of the direction-dependent diffusion of light in structurally anisotropic materials. Combined with a general Monte Carlo implementation of anisotropic light transport, we demonstrated the retrieval of the full scattering tensor in a representative scattering material exhibiting different scattering coefficients along each axis. Our results revealed their complex interplay in determining the observable diffusive rates, and a breakdown of the familiar similarity relation that is typically considered valid in the isotropic case.

Compared with previous studies, which were typically limited to the study of only two directions, and which resorted to coarse simplifications for their quantitative characterization, our work demonstrates the feasibility and importance of accurate modeling of anisotropic light transport, providing the experimental and numerical tools for their characterization.

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