New method for broadband spectroscopy of light transport through opaque scattering media.

O. L. Muskens and A. Lagendijk
Center for Nanophotonics, FOM Institute for Atomic and Molecular Physics AMOLF,
Kruislaan 407, 1098 SJ Amsterdam, The Netherlands.
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We present a new broadband technique for the measurement of diffuse light transport through opaque scattering media. Using the spectral correlations introduced by a scattering medium onto a white-light supercontinuum spectrum, the diffusion constant of light is determined over a wide spectral range in the visible and near infrared. Independent broadband measurements of both the transport mean free path and the diffusion constant are used to calculate the spectral dependence of the energy velocity in a porous GaP slab. Broadband correlation spectroscopy is found to be an excellent tool for the characterization of random scattering media.

Quantitative information on light transport in disordered media is of importance in many medical, chemical, and physical applications [1, 2]. Scattering plays a crucial role in the performance of novel nanophotonic structures such as photonic crystals, waveguides, and metamaterials [3–5]. Access to the spectral variation of light scattering properties is indispensable when studying different scattering regimes, effects of resonances or band structure [6], or eventually the transition to Anderson localization [7]. Until recently, many light transport experiments were limited by the use of single-wavelength or narrowband tunable lasers, hampering the interpretation of results. Recent developments in spatially coherent, broadband light sources allow for new experiments yielding unprecedented access to interference phenomena in random media [8].

Static light transport is characterized by a mean free path ℓ, which can be obtained from total transmission and enhanced backscattering experiments. Dynamic measurements, like time-resolved transmission of short optical pulses, give access to the diffusion constant D and to the transport velocity v_E [6, 9, 10]. Access to the dynamic transport parameters can also be gained through frequency correlations of light transmitted through the material, as first demonstrated by Genack et al [11, 12]. Further experiments have revealed both short-range and long-range correlations [13, 14], in close agreement with theory [15].

Here, we introduce a novel broadband method for measurement of dynamic transport parameters over a wide spectral range in the visible and infrared combining a supercontinuum white-light source and a Fourier transform (FTIR) spectrometer. A schematic overview of the experimental setup is shown in Fig. 1. The supercontinuum light source (Fianium SC-450) produces a 2-mm wide, collimated beam of 2 W optical power in a spectrum spanning the visible and infrared from 450 nm up to 1800 nm wavelength. The transmission of light through a slab of thickness L reduces the intensity roughly by a factor ℓ/L times the fraction of solid angle used for detection, resulting in efficiencies as low as 10^{-6} for single speckle measurements on optically thick samples. In order to maximize the available optical power in a single speckle spot behind the scattering medium, the beam was focused to a spot of approximately 5 μm in diameter onto the sample using a lens with a 3 cm focal length. An angular cone of 2.5° of the light diffusively transmitted through the sample was collimated and coupled into a FTIR spectrometer (Varian FS7000e). The transmitted optical power, in the μW range, was detected by a silicon detector (Thorlabs PDA55) with preamplifier. Polarization filters before and after the sample were used to select linear polarization channels. Spectra were collected with 0.5 cm^{-1} resolution and an integration time of 120 s per spectrum (i.e. 25 scans of the FTIR spectrometer), while the sample position was kept fixed. An average over the disorder was obtained by selecting different sample positions by rotating its azimuthal axis using a stepper motor. Each spectrum was normalized to a spectrum averaged over 30 different sample positions to divide out the instrumental response. An additional correction for small variations in sample transmission was made by normalizing the total integrated intensity over the spectral bandwidth to that of the averaged spectrum.

We have used the white-light frequency correlation technique to investigate some of the most strongly scattering materials to date. The material under study here is photoanodically etched porous GaP [16]. The material consists of a high density of pores in a crystalline GaP wafer with a volume fraction of about 50%. A typical normalized spectrum of light transmitted through a 10.2 μm thick slab of porous GaP is shown in Fig. 2. The strong variations of the intensity with frequency correspond to speckle resulting from interference of many

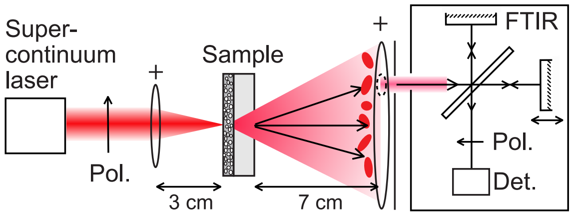

FIG. 1: Experimental setup.

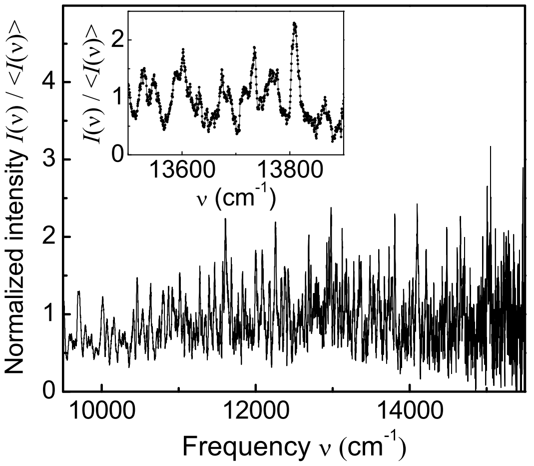

FIG. 2: Transmission spectrum through a 10.2 μm thick porous GaP slab, normalized to the spectrum averaged over 30 different sample positions. Inset: detail of the spectrum showing individual speckle.

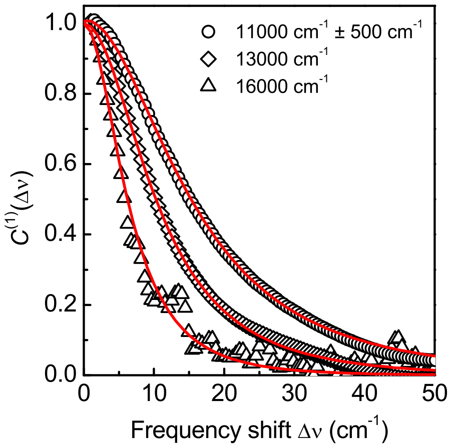

FIG. 3: Spectral correlation function $C_1(\Delta\nu)$ obtained from experimental speckle spectra over a bandwidth of 1000 cm^{-1} around center frequencies of 11000 cm^{-1} (circles), 13000 cm^{-1} (diamonds), and 16000 cm^{-1} (triangles). Lines denote fits using Eq. 2.

multiple scattering paths through the medium [17]. The typical spectral range of the variations amounts to tens of cm^{-1}, which is easily resolved by the FTIR spectrometer (cf. inset of Fig. 2). However, spectral overlap of speckle originating from different coherence areas reduces the intensity contrast. The second moment of the intensity distribution over the spectrum in Fig. 2 was calculated to be 1.17, i.e., below the value of two expected for intensity fluctuations in the diffuse scattering regime. It was verified that the influence of speckle overlap on the frequency range of correlations is small as long as the remaining fluctuations exceed those of the higher order long-range $C^{(2)}$ correlation [14].

We calculated the spectral correlation function $C^{(1)}(\Delta\nu)$ in a spectral bandwidth from $\nu_{\min}$ to $\nu_{\max}$, for single incoming and outgoing directions a and b, using the definition

$$C^{(1)}(\Delta\nu) \equiv \int_{\nu_{\min}}^{\nu_{\max}} \mathrm{d}\nu \, \frac{\langle \delta I_{ab}(\nu) \delta I_{ab}(\nu+\Delta\nu)\rangle}{\langle \sigma_{ab}(\nu)\rangle \, \langle \sigma_{ab}(\nu+\Delta\nu)\rangle}\,, \quad (1)$$

where $\delta I_{ab}(\nu) = I_{ab}(\nu) - \langle I_{ab}(\nu)\rangle$ denotes the intensity fluctuation and $\sigma^2_{ab}(\nu) = \langle I_{ab}(\nu)^2\rangle - \langle I_{ab}(\nu)\rangle^2$ is the variance of the intensity distribution, brackets denoting averaging over different realizations. For the case of transmission through a slab, factorization of the correlation function results in an analytical expression given by[12–15]

$$C^{(1)}(\Delta\nu) = \frac{L^2}{\ell^2} \frac{\cosh \ell\eta - \cos \ell\eta}{\cosh L\eta - \cos L\eta}\,, \quad (2)$$

with $\eta = \sqrt{4\pi\Delta\nu/D}$, where D denotes the diffusion constant of light in the medium. The effect of internal reflection can be introduced by setting $L = L_{\mathrm{slab}} + z_1 + z_2$, where L_{slab} denotes the slab thickness and z_1 and z_2 are the extrapolation lengths at its two interfaces.

We have evaluated the spectral correlation function for the GaP slab by integrating over spectral windows of 1000 cm^{-1} in width. It was found that the first two points in the correlation function accumulated all the uncorrelated noise of the setup, these points were therefore discarded. Subsequently all correlation functions were normalized to the third data point, i.e. $\Delta\nu = 1$ cm^{-1}. Resulting correlation functions are shown in Fig. 3 for three frequency ranges in the spectrum. A strong variation of the width of the correlation function is observed, which indicates a change of the diffusion constant of light in the random medium. Fits using Eq. 2 are shown by the red lines in Fig. 3. These fits yield the average diffusion constants in the selected spectral regions as shown in Fig. 4(b). In the fitting procedure, we used parameters of the slab $L_{\mathrm{slab}} = 10.2 \pm 0.2$ μm and $(z_1 + z_2)/\ell = 4.3 \pm 0.5$.[16] Values of the mean free path were taken from broadband enhanced backscattering spectroscopy described in Ref. 8, as shown in Fig. 4(a). Both ℓ and D are strongly reduced toward higher frequencies, indicating an increase in scattering strength of the material. To assess the effect of the uncertainty in extrapolation length, we have fitted both ℓ and D over the confidence interval of $z_1 + z_2$. As indicated by the shaded areas in Fig. 4(a,b), the diffusion constant D depends much less on the boundary conditions than ℓ, demonstrating that, for thick enough slabs, D can be obtained from speckle correlations independently from ℓ, even though the latter enters the fitting procedure through Eq. 2.

Using the values of D and ℓ obtained from broadband experiments, we estimate the energy transport velocity v_E using the relation $D = v_E\ell/3$. Resulting values of v_E are shown in Fig. 4(c). We obtain a value of v_E/c_0 of 0.73 ± 0.15 at 10000 cm^{-1}, which decreases slowly toward higher energies to a value of 0.51 ± 0.11 at 16000 cm^{-1}. Above 16000 cm^{-1} the combination of low transmission and detector sensitivity currently prevents spectral correlation measurements. Our low-frequency value of v_E is

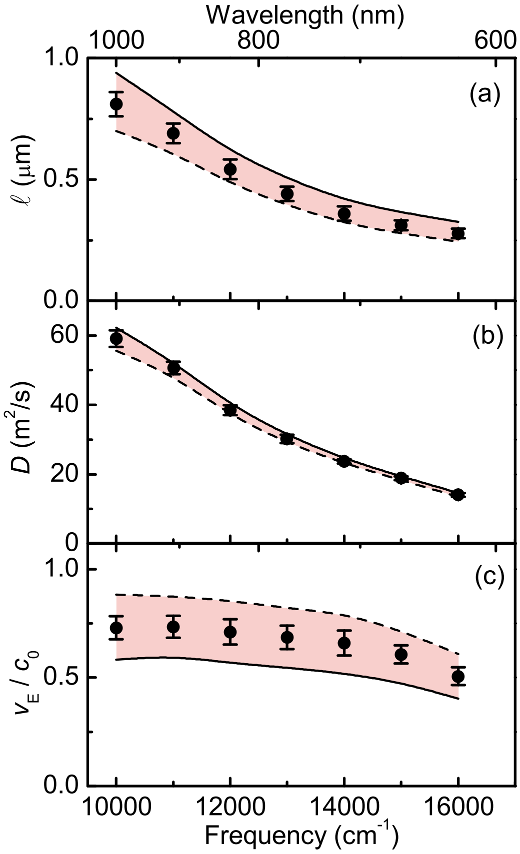

FIG. 4: Experimental values over a wide spectral range of (a) the transport mean free path ℓ (cf. Ref. 8) and (b) diffusion constant D obtained from the $C^{(1)}$ frequency correlation function for a 10.2 μm porous GaP slab. (c) Calculated energy velocity v_E using $D = v_E\ell/3$. Error bars denote fitting error at a fixed extrapolation length $z_1 + z_2 = 4.3$, while the shaded areas represents confidence interval (including fitting error) for $z_1 + z_2 = 3.8$ (solid lines) and 4.8 (dashed lines).

in good agreement with the value of $v_E/c_0 = 0.79 \pm 0.15$ obtained by Johnson et al. on the same series of samples [10]. Peeters et al. reported a $v_E/c_0 = 1.0 \pm 0.2$, which appears rather high given that v_E cannot exceed the speed of light [9]. The transport velocity in the low-frequency range of Fig. 4(c) lies close to the phase velocity $v_{\rm ph}/c_0 = (0.69 \pm 0.07)$ estimated using the effective refractive index of porous GaP [18]. The quantitative agreement between v_E and $v_{\rm ph}$ indicates that internal resonances do not play a role in the macroporous GaP network, in contrast to for example TiO$_2$ materials in which a strong reduction v_E has been reported [13]. The decrease of v_E toward higher frequencies in the GaP indicates a small but significant effect of internal resonances for larger size parameters.

In conclusion, we have demonstrated a new broadband technique for accurate measurement of the diffusion constant of light in opaque random scattering materials. We have studied a strongly scattering porous GaP slab and have extended previous results from time-resolved measurements. The broadband correlation method results in values for the diffusion constant that are as accurate as obtained in time-resolved interferometry experiments. White-light spectroscopy of random media holds promise as a versatile tool for detailed investigations of spatial interference phenomena and mesoscopic transport, where the optical frequency represents an important tuning parameter.

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