Observation of Anomalous Transport of Strongly Multiple Scattered Light in Thin Disordered Slabs

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We present results of experiments on the transport of light through thin random media. Total transmission and time-resolved propagation measurements were performed using strongly scattering samples of varying thickness L. For $L/\ell < 8$, where ℓ is the transport mean free path, the observed decay times from the long-time exponential behavior exhibit strong deviations from diffusion theory and radiative transport theory, whereas the total transmission measurements do not. For the thinnest sample ($L/\ell \simeq 2$) a reduction of the diffusion coefficient with a factor of 2 was observed. [S0031-9007(97)04617-6]

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The interest in the field of multiple scattering of classical waves in random media was initially led by the analogy with electron scattering in disordered metals. The observation of the counterpart for light of Anderson localization [1] was the ultimate goal. However, experimental parameters to attain this effect have not yet been achieved. Many other interesting phenomena in light scattering have since been investigated and observed, among others weak localization and universal conductance fluctuations in light transmission [2,3]. Recently, interest has arisen in medical imaging applications to determine optically the location of objects in strongly scattering biological tissues. Optical imaging may serve as an important substitute for x-ray based techniques as optical radiation is nonionizing. Pulse transmission measurements on very short time scales already give promising results [4].

An exact description of propagation of light through a medium that scatters and absorbs is quite complicated. A tremendous simplification is obtained if the propagation can be described with transport theory, where all interference effects are neglected. If the sample size (L) is considerably larger than the transport mean free path ℓ, there will be no coherent beam left and the equation of radiative transfer reduces to a diffusion equation [5,6]. The study of localization of light is concerned with the breakdown of diffusion theory for strongly scattering thick samples.

Here we report on an observed breakdown of radiative transfer theory for relatively thin ($L/\ell < 8$) strongly scattering samples. For large thicknesses these samples behave according to diffusion theory. Using a new experimental technique [7] we studied the dynamics of light propagation in thin disordered samples and found surprisingly large deviations from time-dependent diffusion theory and radiative transfer theory. The diffusion coefficient following from the long-time exponential decay of the transmitted pulse appears to depend on the sample thickness and is considerably reduced for $L/\ell \simeq 2$.

The experiments were performed on strongly scattering samples made of TiO_2 particles (with refractive index $n \approx 2.8$) on the basis of commercially available rutile pigment. The particles have a size distribution characterized by $d = 220 \pm 70$ nm. We prepared samples with a thickness ranging from 1.43 to 18 μm. Mean values and standard deviations of L were determined by measuring on several positions on the samples using a microscope. For all samples the estimated standard deviation of L was 0.3 μm.

In the time-resolved experiments the transport of an ultrashort light pulse through the sample is investigated employing a new time-resolved interferometric approach [7] in which, contrary to custom in ultrafast spectroscopy, no nonlinear techniques are used. This approach enables a measurement of transmitted pulses with an exceptionally high dynamic range and high time resolution. A 70 fs pulse from a Ti:Sa laser with a central wavelength of $\lambda = 781$ nm is focused on the sample with a lens with a focal length of 10 cm. A second lens with a focal length of 2.5 cm is used to collect the scattered light in transmission. The interferometric autocorrelation function of a double pulse consisting of the scattered pulse and a delayed undisturbed pulse is recorded using a Fourier-transform (FT) spectrometer (BioRad FTS-60A). The FT spectrometer enables us to measure the autocorrelation function with a time resolution of the order of 10 fs. A Mach-Zehnder interferometer at the front end of the FT spectrometer is used to create the double-pulsed signal. The reference pulse is amplitude modulated at 100 kHz using a photoelastic modulator (Hinds) and two polarization filters. Using a lock-in technique, the autocorrelation function coming from the FT spectrometer is demodulated enabling measurements with an exceptionally large dynamic range (5 orders of magnitude). The autocorrelation function is the combination of the autocorrelation function of the two pulses and their cross correlates. The cross-correlation functions contain all information on the pulse transport through the disordered sample as function of time.

We first show that the utilized experimental interferometric technique essentially measures the averaged intensity, a quantity that is easily compared with results

from transport theories. The interferometrically measured cross-correlation function is related to a spatial amplitude-amplitude correlation function valid *inside* the sample, which can be written as

$$C_{\mathbf{r}}(\Delta L/c) = \mathrm{DC} + \frac{1}{\Delta t}\int_{-\Delta t/2}^{\Delta t/2} dt \times A_{\mathrm{sig}}(t,\mathbf{r})A^{*}_{\mathrm{ref}}(t + \Delta L/c,\mathbf{r}), \quad (1)$$

where $\Delta L/c$ is the variable delay in the Michelson interferometer of the FT spectrometer, Δt is the integration time, and A_{ref} and A_{sig} are the reference and signal amplitudes, respectively. The term designated by DC is a constant given by the mean intensity of the pulses. For the signal amplitude we can write

$$A_{\mathrm{sig}}(t,\mathbf{r}) = \int_{-\infty}^{t} d\tau \int d\mathbf{r}' G(\tau,\mathbf{r},\mathbf{r}')S(\tau - t,\mathbf{r}'), \quad (2)$$

where $G(\tau,\mathbf{r},\mathbf{r}')$ is the Green's function which satisfies the wave equation and describes the propagation of scattered light through the disordered sample. $S(t,\mathbf{r})$ is a source term for which we will use a point source $S(t,\mathbf{r}) = j(t)\delta(\mathbf{r} - \mathbf{r}_{\mathrm{source}})$ with j having a Gaussian profile. The amplitude A_{sig} exhibits strong fluctuations as a function of position $\mathbf{r}$, which is a behavior known as speckle. The correlation function $C_{\mathbf{r}}(\Delta L/c)$ oscillates with a period of $\Delta L/c$. To eliminate both the speckles and the fringes, we have averaged the square of the cross-correlation function using 50 to 100 measurements by focusing the impinging pulse at different positions of the sample. For the average of

$$C^{(2)}_{\mathbf{r}\mathbf{r}'}(\Delta L/c) \equiv C_{\mathbf{r}}(\Delta L/c)C_{\mathbf{r}'}(\Delta L/c), \quad (3)$$

one can write

$$\langle C^{(2)}_{\mathbf{r}\mathbf{r}'}(\Delta L/c)\rangle \propto \frac{1}{\Delta t}\int_{-\Delta t/2}^{\Delta t/2} dt \times \exp\left\{-2\left(\frac{t + \Delta L/c}{\Delta\tau}\right)^2\right\}F(t;\mathbf{r},\mathbf{r}'), \quad (4)$$

with

$$F(t,\mathbf{r},\mathbf{r}') \propto \int_{-\infty}^{t} d\tau \exp\left\{-2\left(\frac{t - \tau}{\Delta\tau}\right)^2\right\}\langle|G(\tau,\mathbf{r},\mathbf{r}')|^2\rangle, \quad (5)$$

where the width of the incoming Gaussian pulse is $\Delta\tau$ ($\approx$70 fs in our experiments). From Eq. (5) we see that the signal intensity is a convolution of the incoming intensity with the average of the squared Green's function. The latter is a slowly varying function which can be related directly to results from diffusion theory [6,8].

A typical result of the time-resolved transmission of a sample of 1.43 μm thickness is shown in Fig. 1, where we have plotted the intensity on a logarithmic scale as function of time. An exponential decay for long times is observed over 3 orders of magnitude. The maximum at short times is mainly determined by the coherent (unscattered) pulse,

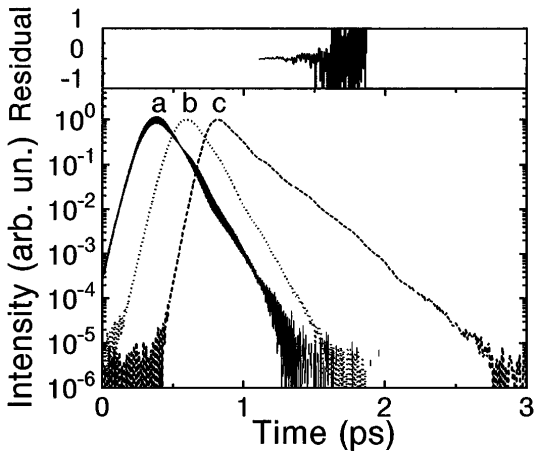

FIG. 1. Typical result of a time-resolved measurement of the transmitted pulse on a 1.43 μm sample [solid line, indicated with (*a*)]. The intensity of the pulse is plotted as function of time. The dotted line (*b*) shifted in time represents the Fourier filtered signal to which an exponential function was fitted: decay time $\approx$70 fs. The relative residual between the determined and measured signal is shown on top of the figure. The dashed line (*c*) shows the result of a measurement on a 4.4 μm sample, demonstrating the exceptionally high dynamic range of 5 orders of magnitude.

while the exponential decay is due to multiple scattering in the sample. Apparently not all the fringes of the cross-correlation function have been averaged out. They were removed using an additional Fourier filter. The filtered intensity is also given in Fig. 1 and has been used for an exponential fit yielding a decay time of 70 fs, which demonstrates the high time resolution of our method. To show the quality of the fit, we have included the relative residual. We estimated the error in the decay time using two independent data sets from one sample and found a value of $\approx$1 fs. We also presented in the same figure results of a measurement on a 4.4 μm sample exhibiting the exceptionally high dynamic range that can be obtained.

To characterize our samples, stationary experiments were performed. The transport mean free path ℓ of the samples was determined by measuring the total transmission at the wavelength of interest ($\lambda = 781$ nm) nm. In the setup the sample is illuminated at one side and all the transmitted light is collected using an integrating sphere. In Fig. 2 the results for the total transmission as function of sample thickness are presented.

To infer the transport mean free path from these measurements we have to take into account the effect of refractive index mismatches. When applying diffusion theory to finite samples, some refinements are required. To describe the fact that the diffusive intensity is actually nonzero at a boundary an extrapolation length [6,9] $z_0 \simeq \ell$ is introduced defining the effective slab thickness $L_{\mathrm{eff}} = L + 2z_0$ to be used in diffusion theory. To account for the influence of internal reflection due to the refractive index mismatch at the boundaries [10], the extrapolation

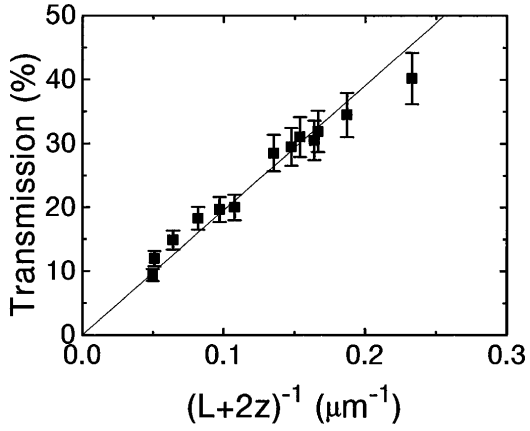

FIG. 2. Measured total transmission as function of sample thickness L. The effective thickness of the sample is adjusted for internal reflection with an extrapolation length $2z = 2.3\ell$. The solid line is the theoretical description based on diffusion theory of the total transmission using a transport mean free path $\ell = 0.95\,\mu$m.

length has to be modified according to [11] $z = z_0(1 + \overline{R})/(1 - \overline{R})$, where $\overline{R}$ is the angle-averaged reflection coefficient at each boundary. The extrapolation length can also be incorporated in diffusion theory by modifying the boundary conditions of the diffusion equation [6,10,11]. For the stationary case and for isotropic scattering, solutions of the Milne equation show [9] that the value of z_0 for an semi-infinite medium becomes smaller by only a few percent for slab thicknesses $2 \leq L/\ell \leq 16$. Comparison of numerical simulations with diffusion theory has shown [12] that the total transmission for varying L and $\overline{R}$ is very well described by the diffusion theory result $T = (\ell + z)/(L + 2z)$, in which $z_0 = 2\ell/3$ [6,11].

Our samples are prepared on a quartz substrate ($n = 1.46$) leading to two boundaries with different reflection coefficients: $\overline{R}_{\text{as}}$ (air sample) and $\overline{R}_{\text{sq}}$ (sample-quartz substrate) giving rise to two extrapolation lengths: z_{as} and z_{sq}. Experiments performed to study the effect of refractive index contrast [13] showed that Mie theory in the independent scattering approximation gives reliable results for the mean index of refraction of the sample. We estimated for the mean index of refraction $n_{\text{eff}} = 1.34$ leading to $\overline{R}_{\text{as}} = 0.4$ and $\overline{R}_{\text{sq}} = 0.02$. The total transmission data were fitted employing results from diffusion theory with extrapolation length $z = (z_{\text{as}} + z_{\text{sq}})/2$. The fit for the total transmission is shown in Fig. 2 for the obtained value for the transport mean free path: $\ell = 0.95 \pm 0.1\,\mu$m. In our experiments $k\ell \approx 8$ so our samples are indeed strongly scattering.

The transport mean free path can also be determined by another, independent, stationary experiment. Measuring enhanced backscattering cones for thick samples yields a value for the scattering mean free path $0.65 \pm 0.1\,\mu$m. This is in good agreement with the value obtained from the total transmission data and therefore gives an independent consistency check on both the measured sample thickness and transport mean free path. The absorption length has been determined and was found to be $\ell_{\text{abs}} \approx 80\,\mu$m. The maximum thickness of our samples was 18 μm, so absorption plays no role in our experiments. All our stationary experiments could be interpreted with conventional stationary diffusion theory.

We will now try to interpret our dynamic results with time-dependent diffusion theory focusing on the *long-time* behavior of the transmitted pulses. For extremely thin disordered samples ($L/\ell \leq 1$) diffusion theory will clearly break down. Although for $L/\ell \leq 10$ deviations in the short-time behavior of transmitted pulses have been shown experimentally [14] one expects for the long-time diffusion theory still to hold. Assuming that the light propagating through the disordered sample can be described by a diffusion process, the long-time exponential decay of the transmitted pulse is characterized [8] by a decay time τ_D

$$1/\tau_D = \pi^2 D/L_{\text{eff}}^2\,, \tag{6}$$

where the effective sample size is given by $L_{\text{eff}} = L + 2z$. In conventional diffusion theory, the decay time τ_D exhibits a length dependence according to Eq. (6). The diffusion constant D follows from (6), the experimentally obtained decay times and thicknesses of the samples. The results are presented in Fig. 3, where the diffusion constant is plotted as a function of sample thickness. We observe a strong dependence on sample thickness of the apparent diffusion constant for $L/\ell < 8$. The thick samples ($L/\ell > 8$) are characterized by the diffusion constant $\overline{D}$ that does not depend on thickness. We found $\overline{D} = 32 \pm 2\ \text{m}^2/\text{s}$. Using this value as a reference one

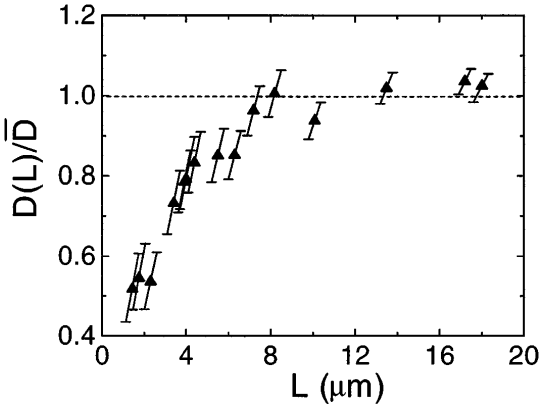

FIG. 3. The measured apparent diffusion constant (triangles) as function of sample thickness L. The thick samples ($L/\ell > 8$) are characterized by the diffusion constant $\overline{D}$. The plotted data are scaled with this $\overline{D}$ (dashed line). Within standard diffusion theory the diffusion constant should not depend on thickness. A strong reduction of the diffusion constant of about 50% can be seen for the thinnest sample. The error bars are tilted since the errors on D are mainly due to the uncertainty in determining L.

observes a deviation in the diffusion constant of about 50% for the thinnest sample. Obviously, $1/\tau_D$ does not obey the diffusion result (6).

We now try to explain our experimental results with transport theory that goes beyond the diffusion description. To investigate the length dependence of $1/\tau_D$ numerical calculations have been performed on the time-resolved Bethe-Salpether (BS) equation [3,15] for the intensity of multiple-scattered light for several slab thicknesses. Previous work [15] indicates that the transition of transport by coherent waves to transport obeying the diffusion equation can be studied as a function of slab thickness employing the time-resolved BS equation. For thick samples the BS equation can be shown to be equivalent to the classical equation of radiative transfer for $k\ell > 1$ [5,6]. Refractive index mismatches can be introduced in the BS equation quite straightforwardly [16]. We solved the wave equation in particular for finite slabs using the complex propagation constant $K = \omega/c + i/(2\ell)$ (assuming isotropic scattering for simplicity). One then obtains finite-size corrections depending on $\overline{R}$ to (a) the inhomogeneous or source term in the BS equation and (b) the BS integral kernel described by the Green's function of the wave equation. For $L/\ell > 2$, our computed long-time exponential behavior gave values for $1/\tau_D$ which agreed within numerical accuracy with results from diffusion theory. Hence, no length dependence of $1/\tau_D$ differing from Eq. (6) was found in this extended approach and our numerical results do not explain our experimental data for $L/\ell < 8$.

As is the standard practice in this field we assumed isotropic scattering in all the above calculations. Taking into account anisotropic scattering may possibly explain our experimental data. To numerically study dynamic finite-size effects for anisotropic scattering, however, is rather involved since one probably also has to focus on the specific intensity which also depends on the solid angle [6].

An alternative explanation for the length dependence of $1/\tau_D$ may lie in a renormalization of D related to a proper formulation of the scattering properties of TiO_2 particles in a finite geometry. When L, ℓ, and λ are of the same order of magnitude, the scattering amplitude depends on the position of the scatterer in the slab, which may result in a modulation of the speed of light giving an effective reduction of the diffusion coefficient.

In conclusion, we have introduced a new technique to study transient phenomena in multiple scattering media. It has been demonstrated here that this technique is characterized by a very high time resolution and an exceptionally high dynamic range. Using this method new and surprising results have been obtained in measurements of transmitted pulses through thin multiple scattering media.

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