# Optical Transmission in Disordered Media

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The scale dependence of transmission in wedged random media containing titania microstructures is measured. The diffusion coefficient and absorption length are determined from the total transmission versus thickness. These parameters are observed to determine the scale dependence of the correlation frequency of fluctuations in intensity within the transmitted speckle pattern as the laser frequency is scanned. Mean free paths as short as 1.4 and 0.35 $\mu$m are observed.

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The study of optical propagation in random media is of broad interest because of the universal character of wave motion, whether classical or quantum mechanical. In this Letter, I show that the nature of optical propagation in disordered solids can be established by measurement of the scale dependence of transmission. Both the total transmission as a function of sample thickness, $T(L)$, and fluctuations in intensity $I(\nu;L)$ within a single coherence area of the speckle pattern in the transmitted light as the laser frequency $\nu$ is scanned for various values of $L$ are measured. From these measurements, the optical diffusion coefficient $D$, the transport mean free path $l$ in which the direction of propagation is randomized, and the imaginary part of the dielectric function, $\epsilon_2$, are obtained. The validity of these methods is established in the regime of diffusive transport in an absorbing medium. An important feature of this approach is that it can be readily extended to the study of nondiffusive coherent propagation[1,2] in more strongly scattering samples. Therefore a discussion of localization is given, though no evidence of it is reported here. Measurements are made in a sample of submicron titania crystals (average index of refraction, $n\sim 2.7$) randomly distributed in a porous wedged polystyrene matrix ($n=1.59$), as well as in a wedged sample of tightly packed and lightly sintered titania crystals. Though the structures and laser wavelength utilized here have not been optimized for maximum scattering strength, the values of $l$ measured are the smallest that have been reported. These results serve as a basis of comparison with theoretical estimates[3] and as a guide for the design of more intensely scattering structures which may localize light.

The change in the character of wave propagation as the scattering strength, characterized by $\lambda/l$, increases is given by the scaling theory of localization.[1,2,4,5] In the classical, diffusion limit, in which $\lambda/l \ll 1$, the phases of scattered partial waves are uncorrelated and propagation can be described in terms of the diffusion of the energy density of the wave, or equivalently, the density of quanta of the field. The diffusion coefficient is given by $D=vl/3$, where $v$ is the wave velocity. When scattering is sufficiently intense that $\lambda/l \lesssim 1$, extended correlations in scattered waves lead to destructive interference which reduces the average transport rate. Since transport then depends upon the spatial extent of wave correlations it can only be described in terms of a scale-dependent diffusion coefficient $D(L)$, which depends upon $\lambda$ as well as $l$.[4] When $l$ reaches a critical value, which was conjectured by Ioffe and Regel[6] to be $l^*=\lambda/2\pi$ for electrons, waves are localized in the Anderson sense[7] that $D(L)\to 0$ as $L\to\infty$. The transition between extended and localized states is predicted to occur for electromagnetic[1] and elastic waves[5] as well as for quantum-mechanical waves. The quantity $D(L)$ is the primary focus of the scaling theory of localization. Its determination from measurements of $T(L)$ and $I(\nu,L)$ would make possible a detailed test of the theory which was developed to describe electron motion in amorphous solids. It remains an open question, however, as to whether large enough index fluctuations and low enough absorption can be achieved in appropriate structures to localize light in three dimensions. The shortest value of $l$ reported here is still a factor of 5 larger than $l^*$.

The universal character of wave propagation in disordered media is borne out by the observation of coherent backscattering of electrons and light in the weak-localization regime.[8] Coherent backscattering of electrons is observed in magnetoresistance oscillations in disordered metallic rings with a period of $h/2e$ in the flux threading the ring.[8-10] These oscillations are due to interference of the time-reversed electron waves which travel completely around the ring. Coherent backscattering of a laser beam from a suspension of polystyrene spheres in water[11-13] and from a solid sample of silica beads[14] has also been observed recently. Polarized reflection is enhanced by a factor of nearly 2 compared with the incoherent background within a narrow cone about the backscattered direction of angle $\theta\sim\lambda/l$.

The experimental setup used to measure optical transmission is as follows: An argon-ion laser pumps a Coherent Radiation, Inc., model 599-21 single-frequency dye laser which can be scanned over 30 GHz. The laser wavelength is $\sim$5896 Å. The laser beam passes through

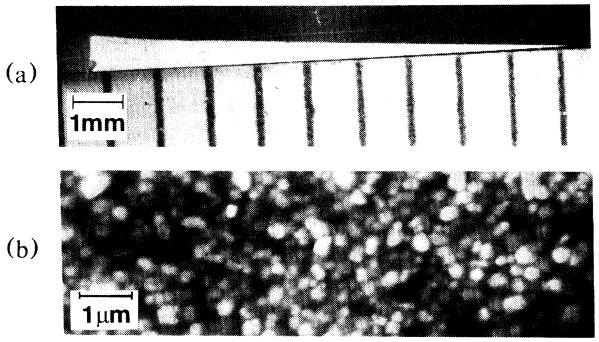


FIG. 1. Sample of $TiO_2$ embedded in polystyrene. (a) Photograph of wedge. (b) Electron micrograph of sample.

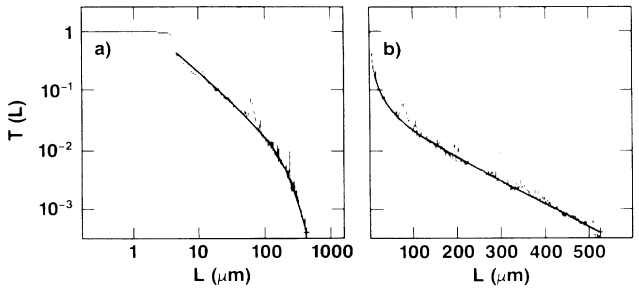


FIG. 2. (a) Log-log and (b) semilog plot of normalized transmission. The solid line is a fit by Eq. (1).

a Coherent Associates electro-optic intensity stabilizer and approximately 20 mW of laser power is focused to a 12-$\mu$m-diam spot on the sample. The wedged sample is mounted on a computer-controlled translation stage, which determines $L$. The transmitted intensity is a stable speckle pattern at a fixed laser frequency which fluctuates in a reproducible manner as the laser frequency is scanned under computer control. To observe high-contrast speckle spectra $I(\nu,L)$ as the laser is scanned with $L$ fixed, a single coherence area is selected with an aperture, and a single polarization component is chosen with a polarizer.

The intensity is detected in a Hamamatsu R9 28 photomultiplier tube (PMT) and stored in the computer after it is normalized to the laser intensity, which is monitored by a photodiode. To measure the normalized transmission $T(L)$, the sample is translated and the transmitted light is passed through a large-area opal diffuser placed immediately after the sample and before the PMT. For large values of $L$, the effect of light scattered around the edges of the sample is eliminated by imaging of the transmitted spot through an aperture before it is detected. Since the dependence of transmission on $L$ for the two configurations is the same for intermediate values of $L$, the data sets can be spliced together to yield $T(L)$.

Most of the results reported here are for the sample shown in Fig. 1. It is a 40%–60% mixture, by volume, of rutile $TiO_2$ microcrystals embedded in a continuous porous polystyrene matrix. The size distribution of the $TiO_2$ powder supplied by du Pont de Nemours and Company is shown in the electromicrograph of a cut surface in Fig. 1(b). The sample is produced by mixing the titania powder with a colloid of 0.091-$\mu$m polystyrene spheres, in water, provided by Dow Chemical. A layer of polystyrene spheres coats the oppositely charged $TiO_2$ surface to produce a structure in which $TiO_2$ particles do not touch. The mixture is allowed to dry and is then pressed into a pellet. It is embedded at an angle into a Lucite block at a temperature which melts the polystyrene into a porous matrix. The block is then polished and sawed in half to display the wedge of 0.075 rad, shown in the photograph of Fig. 1(a). An electromicrograph of the tip of the wedge indicates that the wedge starts at a thickness of $L\sim 5$ $\mu$m.

A measurement of $T(L)$ in the sample, shown in Fig. 1, is presented in Fig. 2 together with a fit by the expression in Eq. (1) for a smooth wedge. Jumps in transmission are caused by pits in an otherwise smooth surface. The lower envelope of the data, therefore, corresponds to transmission in a flat wedge which is found to start at $L=6$ $\mu$m. The log-log plot of $T(L)$ in Fig. 2(a) displays an initial inverse dependence upon $L$ which is the signature of diffusion. The semilog plot of the same data in Fig. 2(b) shows the eventual exponential decay of transmission due to absorption with decay length $L_{abs}=112\pm 5$ $\mu$m.

For a diffuse transport, the photon current density in the interior of the sample is given by $j(x,t)=-D\,\partial n(x,t)/\partial x$, where $n$ is the diffuse photon density associated with a nearly isotropic velocity distribution. For a thick slab with $L\gg l$, the maximum diffuse photon density $n'$ due to a collimated beam of photon flux $j_{in}$ incident normal to the surface at $x=0$ occurs near the front face and has a magnitude calculated from transport theory[15] of $n'=5j_{in}/v$. The steady-state current density transmitted through the slab $j_{trans}(L)$ can be closely approximated by solution of the diffusion equation including the effects of absorption, $D\,d^2n(x)/dx^2-n(x)/\tau_{abs}=0$, where the absorption rate is given by $1/\tau_{abs}=\omega\epsilon_2$ with $\omega=2\pi\nu$, with boundary conditions $n(0)=n'$ and $n(L)=0$. This solution of the diffusion equation is $n(x)=n'\sinh[\alpha(L-x)]/\sinh(\alpha L)$, with $\alpha=(\omega\epsilon_2/D)^{1/2}$. Evaluating the current in the limit $x\rightarrow L$ gives $j_{trans}(L)=n'\alpha D/\sinh(\alpha L)$. The normalized transmission $T(L)=j_{trans}(L)/j_{in}$, can therefore be written as

$$T(L)=5\alpha D/v\sinh(\alpha L)=\begin{cases}5D/vL, & L<L_{abs},\\ (10\alpha D/v)\exp(-\alpha L), & L>L_{abs},\end{cases} \quad (1)$$

where $L_{abs}=\alpha^{-1}$.

The velocity in the medium is found from a measurement of Brewster's angle in reflection of 55.75° to be $v=2.04\times10^{10}$ cm/s. A nonlinear least-squares fit of the lower envelope of the experimental values of $T(L)$ to Eq. (1) is shown in Fig. 2 and gives $D=(9.5\pm0.5)\times10^{5}$ cm$^2$/s and $\alpha=89$ cm$^{-1}$, which yields $\epsilon_2=2.3\times10^{-6}$. The total path traveled for $L=L_{abs}$ is $\sim v\tau_{abs}=3L_{abs}^2/l=2.7$ cm. This is less than the intrinsic absorption length of polystyrene or titania and is probably due to absorption by surface oxidation states of titania or surface adsorbed organic molecules.

John[1] showed that in the critical regime, the relation $L_{abs}=(D/\omega\epsilon_2)^{1/2}$ holds with $D(L)$ substituted for $D$, and Anderson[2] showed that the expression for $T(L)$ can be generalized in similar fashion. Thus, the measurements made here can in principle be used to study optical transport in the critical or localized regimes.

Optical transmission has been measured in a more strongly scattering wedged titania sample. The sample is produced from a sintered 99.999% $TiO_2$ sputtering target supplied by International Advanced Materials. It has a porosity of 30% and an average particle size of $\sim0.3$ $\mu$m. The roughness of the surface of the wedge is less than 1 $\mu$m. The use of data in the region in which $T(L)$ is inversely proportional to $L$ gives $l=0.35\pm0.1$ $\mu$m. This is the shortest optical mean free path that has been reported. It is anticipated that considerably smaller values of $l$ would occur in samples with greater porosity and with the wavelength closer to the band gap at 0.41 $\mu$m where $n\simeq3.4$. At the low porosity of this sample, scattering is from the interstices and Mie resonances do not enhance the scattering cross section within the sample.[3]

Fluctuations in $I(\nu;L)$ for various values of $L$ are presented in Fig. 3 for the sample shown in Fig. 1. These fluctuations arise because light at any point in the transmitted speckle pattern is a superposition of fields from all paths traversing the sample with relative phase shifts which depend upon $\lambda$. The width of the distribution of phase shifts, $\delta\phi$, may be expressed in terms of the spread in travel times through the sample, $\delta t$: $\delta\phi=2\pi v\delta t/\lambda$. The typical frequency difference between peaks in $I(\nu;L)$, $\delta\nu(L)$, is the frequency shift required to change $\delta\phi$ by $2\pi$ radians. When the changes in scattering cross section and $v$ as $\nu$ is varied are negligible, $\delta\nu$ is given by the uncertainty relation, $\delta\nu\delta t=1$. For diffusive transport, $\delta t\sim1/\delta\nu$ is proportional to the average travel time through the sample, and hence, to $L^2/D$.

Experimental values of the inverse of the half-width $1/\Delta\nu_{1/2}(L)$ of the autocorrelation function of $I(\nu;L)$ are plotted in Fig. 4. The values $\Delta\nu_{1/2}$ are obtained from the average of normalized autocorrelation functions of ten spectra for different speckle spots for each value of $L$. The data give

$$\Delta\nu_{1/2}(L)=0.45D/L^2,\quad L\lesssim L_{abs}. \tag{2}$$

For $L\gg L_{abs}$, $\Delta\nu_{1/2}$ is found to saturate because of the greater absorption of photons which take longer to traverse the sample.

These results are intimately related to universal conductance fluctuations which reflect the wave nature of electrons. Lee and Stone[16] have calculated the conductance correlation function in chemical potential $F(\Delta E)$ as a function of the electron diffusion coefficient $D$ and inelastic scattering length $L_{in}=(D\tau_{in})^{1/2}$, where $\tau_{in}$ is the inelastic-scattering time. The correlation function of transmitted optical intensity in $\Delta\nu$, $F(\Delta\nu)$, can be evaluated in the context of this theory by our making the identification $\Delta\nu=\Delta E/h$, using the optical diffusion coefficient for $D$, and substituting $L_{abs}$ for $L_{in}$ in Eqs. (4)–(7) of Ref. 16. An evaluation of $1/\Delta\nu_{1/2}(L)$ for these parameters for a sample with transverse dimensions much smaller than $L$ with reflecting side walls is

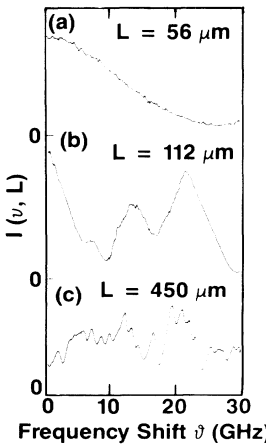


FIG. 3. Examples of intensity fluctuations within a coherence area of light transmitted through the sample.

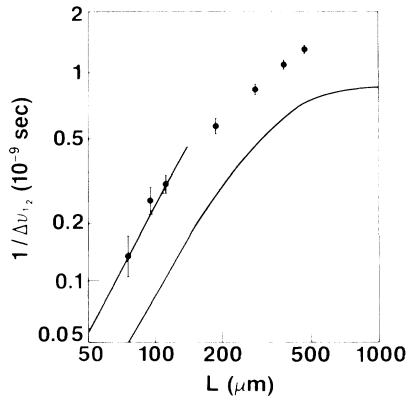


FIG. 4. Inverse of correlation frequency vs thickness, determined from average of autocorrelation function of intensity fluctuation. The solid curve is calculated from the theory in Ref. 16. The line through the data has slope $m=2$.

shown in the solid curve in Fig. 4. The data have the form of this curve but $1/\Delta\nu(L)$ is consistently larger by a factor of $\sim 2.7$. The reduced value of $\Delta\nu_{1/2}$ for optical diffusion may result from the frequency dependence of polarization randomization which is not a factor in the electronic case or partly from the choice of sample transverse dimensions. The value of $1/\Delta\nu_{1/2}(L)$ for a point a distance $L$ from a source embedded within an infinite medium is found from the calculation of Shapiro[17] to be an order of magnitude greater than given by Eq. (2). This is due to the longer lengths of paths that can connect two points in an unbounded sample. By the use of Eq. (2) it is possible to determine $D$ even in strongly scattering samples such as the sintered $TiO_2$ sample studied here for which no specularly reflected beam is observed, expect in grazing incidence. The saturation of $\Delta\nu_{1/2}$ for large $L$ excludes the possibility that the exponential decay of $T(L)$ is due to localization. These results establish the relationship between the average transmission and the frequency scale of fluctuations in transmission.

In conclusion, I have measured, for the first time, the basic parameters of optical propagation in strongly scattering media from a study of the scale dependence of transmission.

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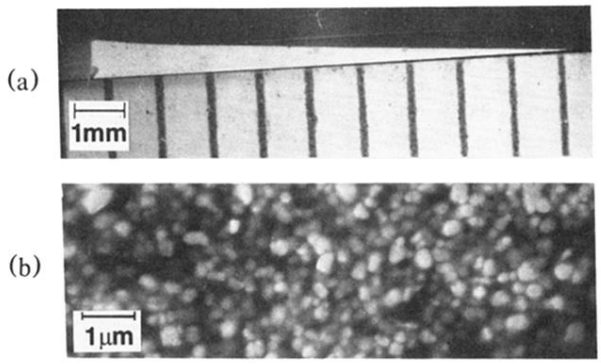


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