P H Y S I C A L   R E V I E W   L E T T E R S

Observation of the Critical Regime Near Anderson Localization of Light

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The transition from diffusive transport to localization of waves should occur for any type of classical or quantum wave in any media as long as the wavelength becomes comparable to the transport mean free path ℓ^*. The signatures of localization and those of absorption, or bound states, can, however, be similar, such that an unequivocal proof of the existence of wave localization in disordered bulk materials is still lacking. Here we present time resolved measurements of light transport through strongly scattering samples with $k\ell^*$ values as low as 2.5. In transmission, we observe deviations from diffusion which cannot be explained by absorption, sample geometry, or reduction in transport velocity. Furthermore, the deviations from classical diffusion increase strongly with decreasing ℓ^* as expected for a phase transition. This constitutes an experimental realization of the critical regime in the approach to Anderson localization.

Diffusive, undirected transport is characterized by a linear increase of the mean square displacement $\langle r^2 \rangle$ with time [1]. As a consequence, transmission of particles through a slab of length L is proportional to $1/L$ known in the context of electrons in metals as Ohm's law. However, when the wave nature of the diffusing particles is taken into account, constructive interference of waves propagating on reciprocal multiple scattering paths may lead to a breakdown of diffusion and the particles are "trapped" on closed multiple scattering loops [2]. This means that the probability of returning to the starting point of such a closed loop is increased twofold due to the fact that the path has the same length in both counterpropagating directions. Thus diffusive transport away from this starting point is reduced. Anderson first predicted this destruction of diffusion in 1958 [3] to explain the metal-insulator transition. If the scattering power of the medium is high, i.e., the transport mean free path ℓ^* is small, the density of closed loops increases which leads to a reduced transport inside the material. This can be described by a rescaling of the diffusion coefficient [4]. For strong localization the rescaled diffusion coefficient becomes zero at finite length scales, such that the spread of $\langle r^2 \rangle$ comes to an end at a length scale corresponding to the localization length. The transition from a diffusive to a localized state should occur when the wavelength λ becomes comparable to ℓ^*, as quantified by the Ioffe-Regel criterion $k\ell^* \simeq 1$ where $k = 2\pi/\lambda$ is the wave number [5].

While localization of electrons in metals was widely studied [6], an unequivocal demonstration of the pure interference effect as predicted by Anderson turned out to be difficult in this case, as electrons bound in deep minima of a random potential are virtually impossible to distinguish from those in closed loops. However, photons in disordered materials revealed an excellent model system to study localization effects [2,7]: in contrast to electrons, they do not interact with each other, nor can they be bound in a random potential. Hence for an infinite random system,

all deviations from classical diffusion have to be due to interference when absorption is taken into account properly. Moreover, in order to create a strongly scattering medium, materials having a large optical refractive index combined with very small absorption are available for visible light.

Thus there have been many investigations in the past studying diffusive transport of visible [8,9] and infrared light [10]. For visible light, where reported values of $k\ell^*$ go down to 3.2 [8], measurements have mainly focused on static transmission experiments where a decay faster than $\propto 1/L$ was interpreted as the onset of localization [11]. It was noted, however, that absorption, which is always present in such samples, can lead to a similar decay [12,13]. Furthermore, recent investigations of time resolved transmission of these samples with similar values of $k\ell^*$ show no discernible deviation from purely diffusive behavior [9]. Localization of photons has also been studied by use of microwave scattering in disordered tubes [13,14], where indications of a decreasing diffusion coefficient have been found [14]. However, it has to be noted that these investigations were carried out in tubes of extent $\sim 2 \times 2 \times 20$ in units of ℓ^*, which constitute a finite, quasi-one-dimensional (1D) system. As localization is always present in 1D and 2D [4], theoretical descriptions of the quasi 1D geometry [15] indicate that observations of localization effects are in this case also possible for values of $k\ell^* \gg 1$. Similar investigations in 3D [16], where there is a phase transition to the localized state, find a negligible effect in the same regime of $k\ell^*$ values. Thus, an unequivocal proof of the transition to localization in three dimensions is still lacking.

Here we present measurements of time resolved photon transport through bulk powders of TiO_2 with typical sample dimensions of $10^5 \times 10^5 \times 10^4$ in units of ℓ^*. These measurements have the advantage that they allow a direct determination of the photon path length dependence of the diffusion coefficient, where absorption and localization

lead to different functional forms of the tail (exponential versus nonexponential) of the path length distribution [15,17]. Furthermore, we independently measure the turbidity of the samples using coherent backscattering [18,19]. This allows a systematic study of the approach to the localization transition as a function of turbidity.

Our samples consist of ground TiO_2 particles in its rutile structure with a refractive index of 2.8. Those particles are commercially available as pigments for white paint from DuPont (R101, R104, R700, R706, R900, and R902) or Aldrich (Ti-Pure). The resulting grains have various average particle diameters, closely spaced between 220 nm to 540 nm with a polydispersity of $\sim 25\%$. Electron micrograph images of R700 and Ti pure are shown in the insets of Fig. 1. In order to minimize ℓ^* these powders are compressed to obtain filling fractions of $\phi \simeq 0.4$. The values of ℓ^* are determined from coherent backscattering [18,19]. Constructive interference on reciprocal paths creates a twofold enhancement of the light in direct backreflection, as in this case every path has a reciprocal partner. The angular width of this enhancement to the incoherent background is inversely proportional to $k\ell^*$ [20].

For the highly scattering samples studied here, the width becomes very broad, which is why the distribution has to be measured to very wide angles for a good determination of the incoherent background. This incoherent background is not constant but depends on the backscattering angle θ as $\cos(\theta)$, such that the difference of the backscattering signal from this functional shape has to be determined. Our setup was custom designed for this purpose and consists of 256 photosensitive diodes attached to an arc with a diameter of 1.2 m in order to get sufficient angular resolution over a range of $|\theta| < 65°$ [21]. Here, the resolution is $\sim 1°$ for $|\theta| > 10°$ and $0.14°$ for $|\theta| < 10°$. In addition, the central part of the backscattering cone, $|\theta| < 3°$, was measured
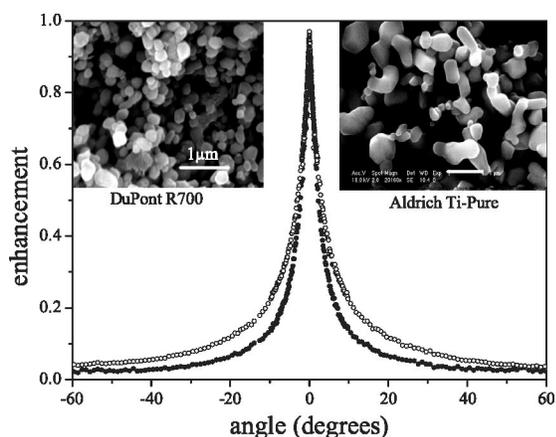

FIG. 1. Measurements of coherent backscattering for two different samples. Open symbols: R700 with an average particle diameter of 250 nm which yields $k\ell^* = 2.5$. Closed symbols: Ti-Pure with an average diameter of 540 nm and $k\ell^* = 6.3$. All measurements were done with circularly polarized light at a wavelength $2\pi/k = 590$ nm. The insets show electron micrographs of R700 and Ti-pure.

separately using a beam splitter and a charged coupled device camera to a resolution of $0.02°$. The measurements are done using circularly polarized light in order to reduce the influence of singly scattered light.

Figure 1 shows the backscattering angular distribution for two different samples, R700 and Ti-Pure, corresponding to average particle diameters of 250 nm and 540 nm, respectively. Using the value of the averaged refractive index, n, from numerical simulations based on the energy density coherent potential approximation [22], we determine the reflectivity of the surface and are hence able to correct for the overestimation of $k\ell^*$ due to internal reflections [23]. Thus the values of $k\ell^*$ are determined from the full widths at half maximum of the curves with a correction for the reflectivity of the surface. This gives values ranging from $k\ell^* = 6.3(1)$ for Ti-Pure to $k\ell^* = 2.5(3)$ for R700 at a wavelength of 590 nm, close to the Ioffe-Regel criterion. Such a direct determination of ℓ^* is of vital importance in the systematic investigation of sample properties with turbidity below. Time of flight (TOF) measurements cannot be used for this purpose as they only give information on the diffusion coefficient $D = v\ell^*/3$. Hence a reduction in transport velocity due to resonance scattering [24] leads to an underestimation of ℓ^* by a factor of up to five.

In order to measure the time resolved transmission we use a single photon counting method, where the time delay of a picosecond light pulse transmitted through the sample is measured. From a histogram of TOFs of many such pulses, the path length distribution inside the sample is obtained directly [9,25]. Our setup consists of a dye laser modified with a mode locker and a cavity dumper, which works at a wavelength of 590 nm with a pulse width of ~ 20 ps. In order to recover the pure path length distributions, the TOF histograms have to be deconvoluted with the pulse shape of the laser system. This is because after pulses, albeit strongly suppressed, nd indiscriminate noise may lead to disturbances in the TOF measurements. For this purpose we measured the pulse shape in the absence of a sample for each experiment, which also yields data on the indiscriminate background. This zero pulse was then deconvoluted with the TOF data in Fourier space to directly give the path length distribution for a supposed delta-peaked pulse as it is calculated theoretically. Figure 2 shows the path length distributions in the samples characterized in Fig. 1 compared to R902 with $k\ell^* = 4.3$. As can be seen in Fig. 2(a), classical diffusion theory including absorption [17] fits the data from Ti-Pure very well. In contrast, the path length distribution of R700 [Fig. 2(c)] shows marked deviations from diffusive behavior. In particular, we observe a nonexponential decay at long times, with photons staying inside the sample longer than expected from a purely diffusive process consistent with local scaling theory [17]. Note that this does, however, not imply an increased total transmission, as the distributions are normalized to total counts. Rather, the total transmission

as indicated by the count rate decreases with increasing turbidity.

A distribution of ℓ^* values in the sample due to, e.g., stratification, might lead to a similar change in D with time. However, if the sample is illuminated from the opposite direction, such a layering would lead to inverted
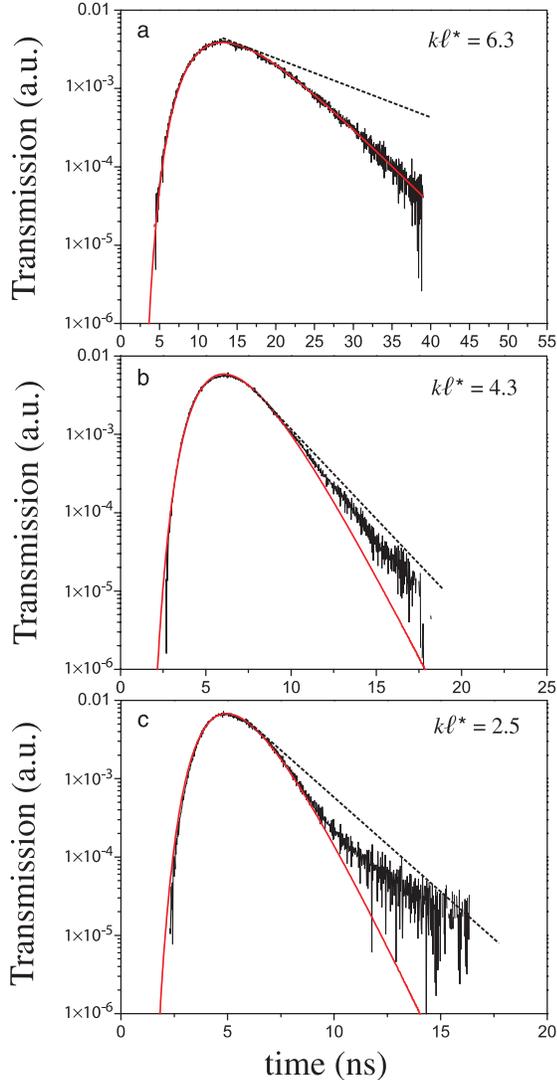

FIG. 2 (color online). Path length distributions from R700, R902, and Ti-Pure. The experimental results are compared to diffusion theory including absorption (full line). In (a), one can see that the data from Ti-Pure ($L = 2.5$ mm, $D = 22$ m^2/s, $\ell_a = 2600$ mm, $n = 1.28$) closely follow the diffusion fit, showing an exponential decay at long times. Part (c) in contrast shows strong deviations from the diffusion fit for R700 ($L = 1.48$ mm, $D = 15$ m^2/s, $\ell_a = 340$ mm, $n = 1.55$), with a clearly nonexponential decay at long times. These deviations can be explained by a time dependent diffusion coefficient in the sample. An intermediate case is shown in part (b) from R902 ($L = 1.51$ mm, $D = 13$ m^2/s, $\ell_a = 380$ mm, $n = 1.23$), with a value of $k\ell^* = 4.3$, where small deviations from the classical behavior can be observed. The respective values of the absorption length are indicated by the slope of the dashed lines.

deviations as well, i.e., a decay faster than exponential. We tested this by measuring the TOF distributions with sample R700 flipped. These measurements show the same tail at long times as Fig. 2(c), hence we can exclude the possibility of stratification. Furthermore, we have checked for the possibility of this long time tail arising from background illumination or light passing around the sample somehow. When looking at different samples of R700 with thicknesses ranging from 1 to 1.5 mm, no difference in the long time tail was observed, while the count rate decreased by a factor of roughly ten.

Diffusion in a slab of length L, which in the experiments below ranges from 1.3 mm to 2.5 mm, leads to an exponential decay of the time dependent transmission at long times as [17]

$$I(t) \propto e^{-[(\pi^2 D(t)/L^2)+(c/n\ell_a)]t}. \tag{1}$$

Here ℓ_a is the absorption length, which in our samples ranges from 0.3 m to 2.6 m and c is the speed of light in vacuum. Note that the values for ℓ_a are about a factor of 10^6 larger than typical values of ℓ^* in our samples. Any nonexponential decay of $I(t)$ at long times thus indicates a temporally varying diffusion coefficient $D(t)$. In that case, the exponential slope in Eq. (1), changes from the sum of the diffusive part and the absorption determined part to one which is solely determined by the absorption length. This can also be seen in Fig. 2, where the absorption length is indicated by a dashed line. $D(t)$ can be quantified by taking the negative time derivative of the logarithm of the intensity [14]. In the inset of Fig. 3 this is shown normalized to $[\pi^2 D(t=0)/L^2 + c/n\ell_a]$ for the samples of Fig. 2. At long times the classical Ti-Pure approaches the constant value of 1, whereas the curve for R700 shows a strongly decreasing diffusion coefficient and hence a temporally varying diffusion coefficient.

To quantify the nonclassicality of the diffusive light transport through the different samples we determined the average of the ratio of the measured data to the diffusion fits. This was done systematically for all powders over the time interval from t_{max} to $3t_{max}$, where t_{max} is the time of the peak maximum. This deviation is given as a function of $k\ell^*$ as determined from coherent backscattering in Fig. 3. The figure clearly shows that the deviations increase strongly when approaching the Ioffe-Regel criterion, as expected for a phase transition.

The data we have presented show clear deviations from diffusive transport through these highly scattering samples. The nonexponential decay of the path length distribution indicates a renormalized value of the diffusion coefficient at long times as was predicted by scaling theory [4,7]. Moreover, the deviations from diffusive behavior scale with the value of $k\ell^*$ consistent with the Ioffe-Regel criterion [5] and the approach to a phase transition.

These deviations cannot be explained by absorption as this only leads to an additional exponential decrease but cannot introduce a nonexponential path length distribution.
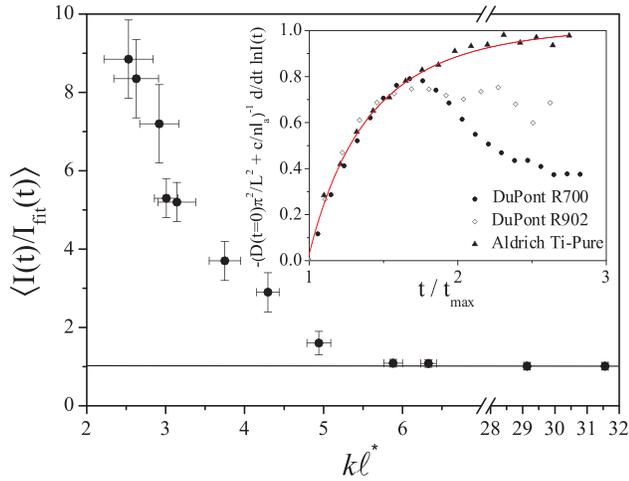
FIG. 3 (color online).   Measurements of the deviations from diffusion on several samples with different values of $k\ell^*$. The deviation is quantified by taking the ratio of $I(t)$, to the classical fit, $I_{fit}(t)$, and averaging it over the time interval from t_{max} to $3t_{max}$. The results were plotted over $k\ell^*$ from coherent back-scattering (see text). Samples that have a $k\ell^*$ nearer to the Ioffe-Regel criterion show a bigger deviation from diffusive behavior. The inset shows characterizations of the time dependence of the diffusion coefficient (see text) for the three samples presented in Fig. 2. R700 shows a marked decrease in D at long times in contrast to Ti-Pure, which follows the prediction for classical diffusion (full line). This demonstrates the existence of localized modes in R700.

There is also no systematic dependence of the values of the absorption length of the various samples on $k\ell^*$ in contrast to the monotonic dependence of the deviations from classical diffusion. A change in the transport velocity due to resonant scattering [24] can also be ruled out, since values of the diffusion coefficient from the classical fits are consistent with a value of the transport velocity of c/n for the most localizing sample [26]. Furthermore, such a change is not expected to be dependent on the length of the specific path and hence would not lead to a nonexponential decay as the data show. Moreover, sample inhomogeneities, such as a stratification, cannot account for the effect, as the path length distributions are independent of the direction of illumination. Finally, background illumination or spurious paths measured due to low count rates can also be ruled out as the deviations are independent of count rate as can, for instance, be seen by varying the sample thickness. We thus conclude that these observations constitute direct evidence for the slowing down of photon diffusion due to the approach to the Anderson localization transition and hence for the existence of the transition to strong localization of photons in three dimensions.

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