Weak Localization of Photons: Universal Fluctuations and Ensemble Averaging

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We have carried out an experimental study of weak localization of photons in a rigid, nonabsorbing, disordered medium. Unlike recent results reported for particles in a fluid, we observe large-amplitude fluctuations in the scattering intensity as a function of angle. Ensemble averaging of these sample-specific fluctuations decreases their intensity and uncovers a sharp peak in the backscattering direction. These results emphasize the common origin of weak localization of electrons, as a wave phenomenon, and of photons which are inherently wavelike.

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Propagation of coherent waves in highly disordered media is of interest in a wide range of disciplines. An essential feature in the theoretical framework for the analysis of the transport coefficient is the *phase-coherent* multiple scattering of waves. For wave scattering in the absence of absorption, there is for any path a time-reversed path that the wave can travel and undergo the same net phase shift. As a result, two coherent eigenstates of a wave traveling these time-reversed paths and arriving in the backscattering direction undergo the same phase shift together with the same wave-vector transfer. Therefore, they interfere constructively. This constructive interference leads to a peak in the backscattered direction. As discussed extensively in the context of the weak localization of electrons, only the sequence of scattering events that causes the wave to backscatter has this unique property.[1,2] This singular feature, which can be interpreted as an order in disordered media, is the origin of the anomalous manifestations in a variety of situations where one has multiple scattering of waves. While much of the recent literature has focused on electron transport where this phenomenon results in weak localization of electrons, it has recently been pointed out that propagation of photons through a disordered media gives a direct, graphic description of the enhanced backscattering cross section.[3–5]

The experiments on electrons have been fueled by the ability to make electronic structures small compared to the inelastic scattering length. Here substantial quantum corrections to the transport coefficient are obtained that are associated with the diffusive propagation of an electron that undergoes phase-coherent multiple-scattering events. Of particular interest to our work is the recent realization that, if the size of a structure is smaller than the inelastic scattering length, the electron coherently samples the entire volume, and any perturbation that alters its phase locally will lead to large fluctuations in resistance.[6,7] This phenomena is a direct consequence of quantum transport of electrons, rather than "finite-size effects." Weak localization and a coherent backscattering peak without these large fluctuations are expected to be obtained experimentally only in samples that are sufficiently large that a suitable ensemble average over many coherent volumes is possible.[8]

We have carried out studies of propagation of photons in a nonabsorbing, rigid, disordered medium and have shown experimentally for the first time the importance of fluctuations and ensemble averaging in weak localization of photons. Our medium is a "fluff" of fiber-optic–quality, submicron-size SiO_2 beads in air (a solid medium). High-resolution ($<$ 1-mrad) angular scans of reflectance (R) near the backscattering direction contain very large ($dR/R = 1$), reproducible fluctuations that originate from multiple-scattering events inside the bulk. Ensemble averaging of several scans averages out the "sample specific" fluctuations and reveals a sharp peak in the backscattering direction that was hidden under the large-amplitude fluctuations. We have also carried out the same experiments on suspensions of polystyrene balls in water (a liquid medium), and reproduced in more detail the previous results.[3–5] The backscattering peak is seen more readily in the liquid sample because the Brownian motion of the balls does the ensemble averaging very efficiently. *A key result that distinguishes this work from the previous work is that the backscattering peak can be revealed only after the dominant fluctuations in the scattering intensity have been decreased by an ensemble-averaging process.*

The experimental setup for the backscattering experiment is similar to the ones used in the previously reported results.[3,4] To expand the range of our angular scans we have placed the detector assembly and the focusing lens on a goniometer table. The center of the goniometer is defined by the point of incidence of light. The surface of the sample is at an angle to the incident beam to avoid the specular reflectance caused by the mismatch between the effective dielectric constant of the sample and air. The beam of a 5-mW low-divergence He-Ne laser is collimated and used as the source of light. The diameter of the collimated beam is 1 mm, corresponding to an inherent divergence of 0.6 mrad. A low-noise Si detector together with phase-sensitive detection technique provides a signal-to-noise ratio of better than 100 at the limit of our beam resolution.

The solid sample is a network of colloidal silica particles produced by flame hydrolysis of silicon tetrachloride vapor. A uniform thickness of monodispersed particles can be deposited on a variety of substrates. We can vary *d*, the thickness of the sample, from a few microns to a few millimeters allowing us to cover the full range from $d < l$ to $d >> l$, where l is the elastic scattering length. The size of the particles has been determined with a scanning electron microscope to be between 0.1 and 0.2 μm in diameter. The average initial solid fraction in the *as-grown* sample varies between 0.05 and 0.12. The fact that the fluff holds itself as a connected network at such a low solid fraction suggests that the formation process is governed by a diffusion-limited aggregation of silica particles. Sintering of the fluff can increase the solid fraction all the way up to 1.0. In fact, this is a procedure for making high-quality fiber optics. The absorption of the silica particles at the He-Ne laser wavelength is extremely small because of their purity. The liquid sample is similar to the one used in the other experiments.[3-5] It contains 0.3-μm-diam polystyrene balls suspended in deionized water. The liquid sample is provided at a volume fraction of 0.1 by Duke Scientific.

In Fig. 1 we show the scattering results from a few-millimeter-thick fluff. The average density for this sample is 0.1, and the average diameter of the SiO_2 particles is 0.15 μm. Figure 1(a) shows a typical scan near the backscattering direction. The noiselike structure is reproducible from scan to scan. It resembles the "speckle pattern" associated with the reflection of a coherent light from a rough surface, and contains both single- and multiple-scattering contributions to the reflection. To eliminate the component which is the result of a single-scattering event, we pass the beam through a linear polarizer followed by a quarter-wave plate, in that order, just before the surface of the sample. This combination modifies the polarization distribution by $\cos^2\theta$ and acts as an "isolator" decreasing the intensity of a beam that undergoes a single reflection by about 10^3. (2θ is the angle between the polarization of the backscattered light and the linear polarizer.) Although we are looking at a modified distribution of the polarization in the data of Fig. 1(b), we can be sure that this interference pattern originates from the multiple-scattering events inside the bulk of the sample.

The fluctuations in Fig. 1(b) that originate only from multiple-scattering events appear to have larger relative amplitude compared to the results shown in Fig. 1(a). The crossed bars in Fig. 1(a) and Fig. 1(b) are measures of the mean values and the standard deviation of the two patterns. The sharpness of the structure in both cases is limited to the resolution of our spectrometer, which is about 0.6 mrad. The reproducible structure of Fig. 1(b) can be changed by a lateral translation of the sample or by a very slight rotation about the point of entrance of the beam. We find that in order to change the pattern in Fig. 1(b) completely we have to change the position of the beam by almost its whole width. However, the interference pattern is considerably more sensitive to the angle of incidence. Less than a 1-mrad tilt of the sample is enough to produce a completely uncorrelated pattern.

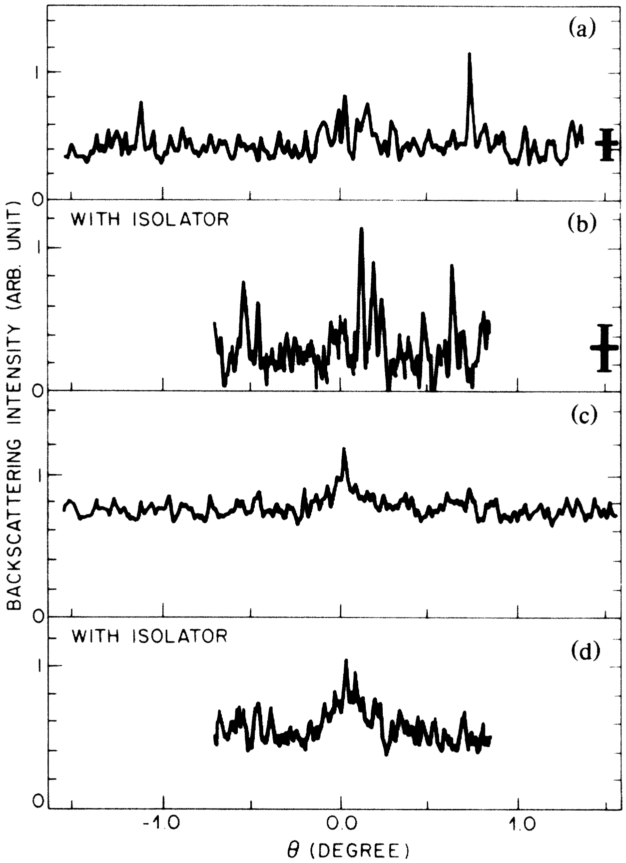

FIG. 1. (a) Backscattering intensity from a rigid fluff of submicron SiO_2 beads in air. (b) The same as (a) but with the single-scattering component removed. (c) Ensemble average of nine scans such as in (a). (d) Ensemble average of sixteen scans such as in (b).

Figure 1(c) shows an ensemble average of nine uncorrelated patterns such as the one in Fig. 1(a). These patterns are obtained by movement of the beam position. Ensemble averaging of N patterns decreases the size of the fluctuations by a factor of $\sqrt{N}$. The net result is the emergence of the sharp backscattering peak. Similar averaging of fifteen uncorrelated traces with the isolator is shown in Fig. 1(d). The relative size of the backscattering peak compared to the average background is larger in Fig. 1(d) than in Fig. 1(c). This is due to the absence of the uninteresting single-scattering contribution in the results of Fig. 1(d).

In Fig. 2 we compare the ensemble-averaged result of Fig. 1(d) with a *single* scan with the "isolator" from

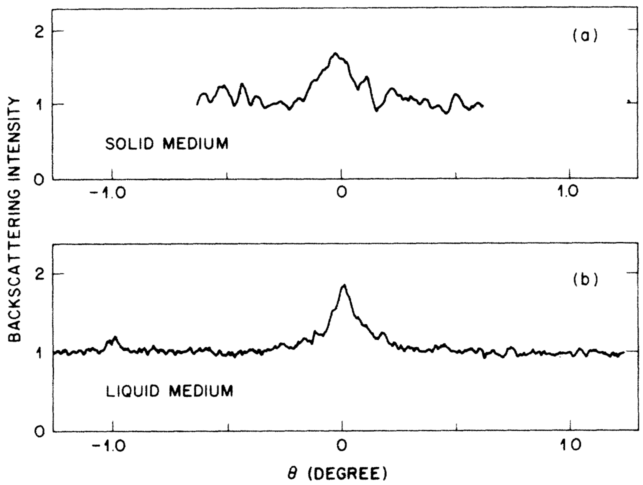

FIG. 2. (a) Numerically smoothed result of Fig. 1(d). (b) *Single* scan with the "isolator" from a suspension of polystyrene balls of comparable size and solid fraction.

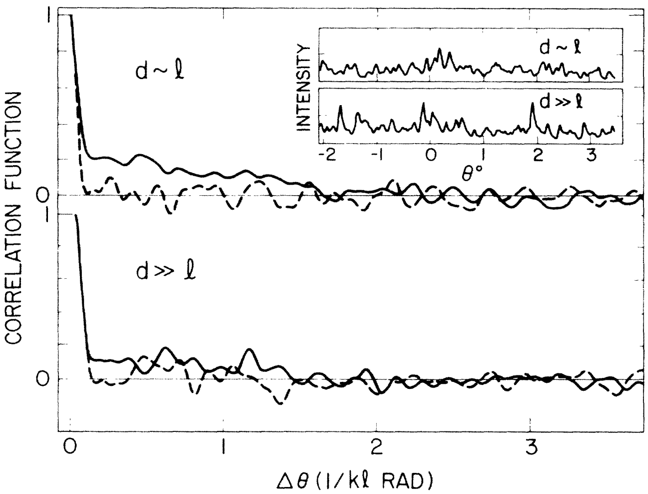

FIG. 3. Inset shows single scans of scattering intensity originating from the bulk of a thin ($d = l$) and a thick ($d >> l$) rigid fluff. Main frame shows the average angular correlation in (solid lines) and 20° away from (dashed lines) the backscattering direction for both samples.

a liquid sample of comparable solid fraction and particle size. The result from the solid sample has been smoothed numerically by the computer; nonetheless, the scan from the liquid sample clearly has much smaller fluctuations. It appears that motion of the polyballs in the liquid effects a very efficient ensemble averaging in the time that it takes the spectrometer to scan the time constant associated with the resolution. *The important observation is that coherent wave propagation in disordered media is inherently noiselike, and the backscattering peak is obtained only by an ensemble averaging.*

The smooth result from the liquid sample, however, enables us to carry out a much more detailed analysis of the line shape of the backscattering peak. For example, we find that the use of the "isolator" does not change the line shape of the backscattering peak. But, by elimination of the component that arises from diffuse specular reflectance the enhancement of the scattering cross section at the backscattering direction is exactly 2, which is in good agreement with the theoretical prediction.[2,9] Furthermore, our analysis, given in detail elsewhere, indicates that the backscattering peak is in fact composed of two characteristically different peaks.[10] These are important findings which were overlooked in the previous measurements.[3-5]

Within the weak localization of a scalar field, the width of the backscattering peak is equal to $1/kl$, where k is the wave vector, and l is the elastic mean free path.[2,9] Through the measurement of the direct transmission and a fit by its expected variation of $\exp(-d/l)$, we find $l = 9$ μm for our solid samples. On the basis of this result, the theoretical estimate of the width is within a factor of 2 of the experimental results. We note that by using the expression for the Rayleigh mean free path in terms of material properties[9]—dielectric mismatch, particle size, and concentration—we find a value for l that is considerably smaller than what is predicted from the width of the backscattering peak. This apparent discrepancy is also present in the results from the liquid sample and is due to aggregation of small particles.

In Fig. 3 we show a more detailed analysis of the intensity fluctuations. The insets to Fig. 3 compare the backscattering scans from a 2-mm-thick and a 14-μm-thick sample, which are in the regimes $d >> l$ and $d = l$, respectively. In both scans we have removed the single-scattering component in order to consider the interference effects that arise from multiple scattering in the bulk. Although the absolute intensity from the thin sample is decreased considerably, we find that the magnitudes of the relative fluctuations in both samples are comparable. This implies that in both mediums the photons sample the entire scattering volume coherently, and the magnitude of the fluctuations is not determined by the size of the medium. That is to say, these are universal fluctuations in the transport coefficients of a photon that undergoes phase-coherent multiple-scattering events. These fluctuations in the scattering intensity as a function of the angle are present at all angles, including the transmission direction.

In Fig. 3 we show the average autocorrelation functions of the reflection patterns from the thin and thick samples. The solid and dashed lines are from patterns in and 20° away from the backscattering direction, respectively. In both samples the correlation function decreases rapidly for angular separations up to an angle defined by the resolution of the spectrometer. As expected from the results of Fig. 1, there is a tail in the autocorrelation of the patterns which include the backscattering peak (the solid lines). This tail extends up to an angular separation of about $1/kl$, and is stronger for the thin sample. The stronger tail in the thin sample implies the backscattering peak from the thin sample is broader. This is consistent with refined studies of the backscattering peak carried out with use of a more detailed ensemble-averaging technique. These results will be reported separately.

The presence of large-amplitude fluctuations in the transport coefficient of photons is a direct consequence of its wave nature.[11,12] We referred to these fluctuations as "universal," because their behavior is independent of the scattering volume. They, however, should be distinguished from the universal fluctuations which are independent of both disorder and sample size, and are seen in the *total* transmission coefficient of electrons as a function of magnetic field or chemical potential.[6,7] This latter phenomena is found to arise from a long-range spatial correlation between the wave functions, and is discussed as evidence for the *quantum* diffusion of an electron.[12,13] Since the fluctuations in the transport coefficients are related to the buildup of correlation caused by multiple scatterings, there should be angular correlation in the scattering intensity. As seen in Fig. 3, we do not find any evidence for a tail in the angular correlations except in the region that includes the backscattering peak. This is also true for patterns obtained in transmission through the thin ($d = l$) and thick ($d = 300\ \mu\text{m} >> l$) samples. In fact the correlation functions of transmission patterns are very similar to the dashed lines in Fig. 3. It should be mentioned that, as a result of the limited size of the scans, a factor that prevents exact definition of the zero baseline, we cannot conclude if there is a weak correlation tail in these results. Therefore, in the absence of a quantitative calculation we cannot use these results to discuss the possible fluctuations in the total transmission.

In conclusion, we would like to emphasize the analogy between our study of weak localization of photons and the weak localization of electrons. The weak-localization effects, seen either as a backscattering peak in our experiments or as oscillations with periodicity of $\frac{1}{2}$ the flux quantum in the magnetoresistance measurements,[8] are present side by side with the large-amplitude sample-specific fluctuations. The presence of these fluctuations is a direct consequence of coherent sampling of the whole scattering volume. That their effects on an experiment can be decreased by an ensemble-averaging process is due to the random nature of the propagation of waves through a disordered medium.

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