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We report the observation of weak localization of light in a random medium. In a sample consisting of a highly concentrated suspension of polystyrene particles in water, polarization-dependent enhanced backscattering of laser light was found within a cone of approximately 0.2° (half angle) for the highest concentration.

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The concept of Anderson localization, that is, localization due to randomness, is one of the most interesting new concepts of contemporary physics.[1] This strong type of localization pertains to the absence of diffusion in random materials as a result of interference of all the scattered waves. A precursor of Anderson localization, sometimes referred to as weak localization, concerning enhanced backscattering in a strongly scattering random medium, was first pointed out for electrons by Abrahams *et al.*[2] Related effects for electromagnetic waves have also been discussed but have hitherto not been associated with localization.[3] Although the vast majority of theories of localization have been developed for the Schrödinger wave equation, it is quite clear that the concept of localization is much broader. In principle, in almost any wave equation localization solutions can be obtained when solved for a random medium. This has been recognized by several workers in the field, and experiments have been suggested to measure (strong) localization for acoustic and electromagnetic waves, especially in one dimension.[4–6] Several aspects of electromagnetic waves make them very suitable to study important localization effects. In the first place their equations of propagation are well known, and the scattering of electromagnetic waves has been intensively studied for almost a century.[7] Furthermore, their vector character adds a new component to the localization problem which shows itself, for instance, as the importance of the polarization of the waves. In one dimension the localized character of solutions to the Maxwell equations can be established because they can be described in terms of a product of transfer matrices.[8] An extremely interesting situation would be the observation of localization of light in two dimensions. Generalizing the present theories on localization to the Maxwell equations implies that light is always localized in two dimensions.

In our laboratory we decided a considerable time ago to try to measure Anderson localization of light in a three-dimensional medium. Localization effects are very general and should be present in many random materials. The medium we opted for was a highly concentrated suspension of polystyrene spheres. Advantages of this medium are (i) the absorption coefficient for light is extremely small, (ii) the concentration can easily be varied up to a very high concentration (up to 30 vol%), and (iii) its single-scattering properties (Mie scattering) are known rigorously.[9] The high concentration is necessary to reduce the mean free path of light as much as possible, preferably down to its wavelength, which is necessary to induce localization effects. The most effective scattering occurs if the particle size is of the order of the wavelength of light.

At lower concentration one should be able to observe the precursor of (strong) localization, a phenomenon which is now known as weak localization and concerns the occurrence of enhanced backscattering. A simple picture of weak localization in real space has been given by Khmel'nitskii.[10] In this approach the transport from point A to B is considered in terms of propagation of wave packets along paths, each path contributing to the total probability of reaching B from A. This total probabiliy is the sum of all contributions squared (coherent sum). Phase differences between the paths will cancel out all interference terms so that the coherent sum will be equal to the incoherent sum, except when A and B *coincide*; for then contributions from the same path but traversing in opposite directions will interfere constructively. Application of this model to a scattering experiment yields a prediction of enhanced backscattering. A similar reasoning has been given in the wave-vector domain.[2] The enhancement factor is predicted by theory to be 2 (doubling of the intensity), and the width of the backscatter cone is predicted to be of order λ/l, where λ is the wavelength, and l is the (transport) mean free path.[3,11] The numerical prefactor is calculated to be $\frac{3}{4}\pi$.[11] In none of the calculations (partial) have depolarization effects been explicitly considered.

We have observed *polarization-dependent* weak localization of light in a small cone in concentrated suspensions. We find that the enhancement factors are substantially lower than 2 for our suspensions. The size of the cone is in good agreement with a theory using a renormalized multiple-scattering expansion. In this theory the length scale is set by the *transport* mean free path.

Our experimental setup is drawn schematically in Fig. 1. By expansion of the beam of a 5-mW He-Ne laser, its divergence was reduced to less than 0.1 mrad. The expanded beam was reflected onto the sample from a beam splitter. The backscattered light was measured as a function of the angle of scattering by a detector that could be moved in the focal plane of a lens placed immediately behind the beam splitter. Earlier versions of the experiment in which no beam splitter was used were not successful because the enhanced-backscatter angle turned out to be too small to be detectable without a beam splitter. A pinhole was fitted directly to the detector housing. The field of view of the detector, as determined by the focal length of the lens and the size of the pinhole, was 0.37 mrad (half angle). In those experiments where the state of polarization of the backscattered light was studied, the Nicol prism P2 was also fitted directly to the detector housing.

Great care was taken to block all alternative light paths leading from the source to the detector: Screens were placed to shield both the stray radiation coming from the chopper and the beam expander, and light backscattered from the cell and reflected from the mounting of the beam splitter and the lens holder. The cell was tilted off-axis in order to keep its window reflections well away from the detector. The fraction of the original beam that passes through the beam splitter was very carefully damped as its reflection from the wall would coincide with the 180° backscattered light from the cell. Background signals were recorded, both without a cell and with an empty cell in the light path. In all cases essentially flat curves (as a function of angle) were obtained. In the experiments where no polarizers were used, background levels were well below 1% of the signal level. With use of polarizers, the highest recorded background did not exceed 10% of the lowest recorded signal.

Alignment is obtained by adjusting the orientation of the beam splitter. The following procedure was used: the diaphragm D was replaced by a pinhole, the cell was replaced by a flat mirror, and the detector was put in its middle position. The positions of the mirror and the beam splitter were then alternately adjusted so as to make one part of the split reflection of the mirror return through the pinhole and the other part enter the detector.

A 10% by weight suspension of polystyrene spheres in water (Latex 5100) was obtained from Dow Chemical. Less dense samples were prepared by dilution with de-ionized water. A more concentrated sample was prepared by means of allowing the particles to settle and then decanting part of the water. All samples were homogenized with an ultrasonic bath before being transferred to the cell.

In Fig. 2 some of the experimental results at various densities are presented. The critical cones are very easily visible. In Table I the characteristics of some of the measured cones are presented. With increasing ρ, the density of the dielectric spheres, the cone broadens, and the ratio of the maximum intensity of the cone and the multiple-scattering intensity outside the cone seems to saturate at $I_{\max}/I_{\mathrm{MS}} \simeq 1.4$ as its

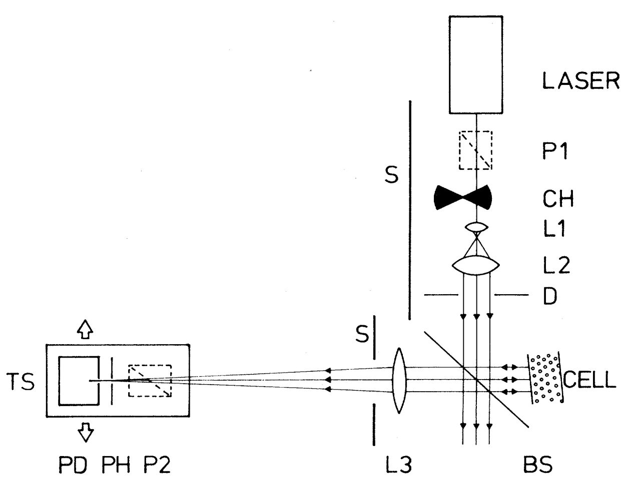

FIG. 1. Schematic representation of experimental setup. P1,P2, polarizers; CH, chopper; L1,L2,L3, lenses; D, diaphragm; BS, beam splitter; S, screen; TS, translation stage; PD, photodetector; and PH, pinhole.

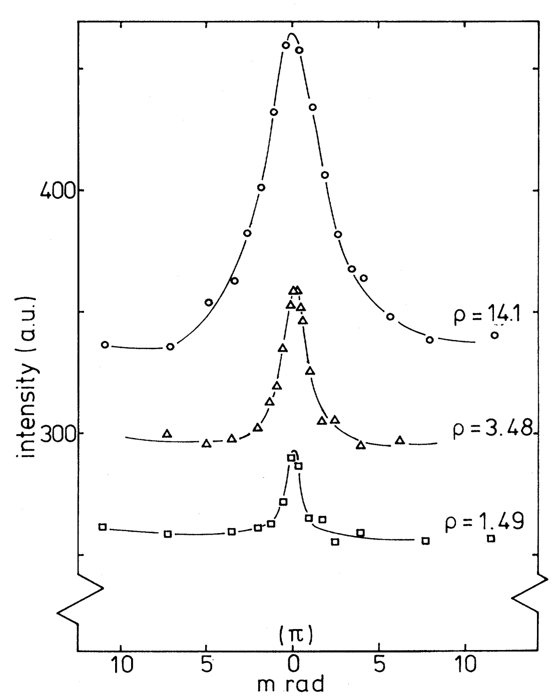

FIG. 2. Backscattering intensity as a function of angle for several concentrations. Density ρ in 10^{16} m^{-3}.

TABLE I. Critical cone in the backscattered light of 1.091-μm-diam polystyrene spheres in water at $\lambda = 633$ nm. I_{MS} refers to the intensity of the multiple scattering outside the region of the cone.

ρ (10^{16} m^{-3})	I_{max}/I_{MS} [a]	Width [b] (mrad)
1.49	1.13	0.11
3.48	1.21	0.45
6.79	1.36	0.98
14.1	1.37	1.58
48.8	1.38	3.8

[a]As measured with a beam splitter with transmission coefficients $T_s = 0.525$ and $T_p = 0.776$.
[b]Half angle at half maximum after subtraction of the field of view (half angle) of the detector.

TABLE II. Partial retention of polarization in the critical cone of $\rho = 14.1 \times 10^{16}$-m^{-3} suspension of $d = 1.091$-μm polystyrene spheres in water at $\lambda = 633$ nm. I_{MS} refers to the intensity of the multiple scattering outside the region of the cone.

Orientation of Nicol prisms[a]		I_{MS} (a.u.)		$I_{max} - I_{MS}$ (a.u.)	
P1	P2	Measured	Corrected [b]	Measured	Corrected [b]
...	s	126	240	58	110
...	p	183	236	66	85
s	s	74	141	47	90
s	p	106	137	32	41
p	s	29 [c]	56	9 [c]	16
p	p	53	68	34	44

[a]Relative to the plane of incidence and reflection of the beam splitter.
[b]Measured intensity corrected for passage through the beam splitter.
[c]Poor S/N ratio.

width becomes large compared to the field of view of the detector.

Studying the state of polarization of the backscattered light, we found that starting with a linearly polarized beam the backscatter light outside the region of the cone was completely depolarized, whereas inside this region some polarization remained. In Table II data are presented on the state of polarization of backscattered light. The measured intensities (columns 3 and 5) were corrected for passage through the beam splitter (columns 4 and 6). From the corrected values it follows that for backscattering outside the region of the cone the s/p ratio is 1 and independent of the state of polarization of the incident beam. From column 6 it can be seen that inside the region of the cone the backscattering enhancement is larger for the original polarization than for the perpendicular polarization.

We have calculated some single-scattering properties of the spheres. The scattering efficiency, which is defined as the ratio of the actual cross section and the geometrical cross section (πa^2), is equal to 2.89 (almost equally shared by the p and s scattering). With these numbers we calculate the scattering mean free path of the 14.1×10^{16}-m^{-3} suspension for light having a wavelength of 633 nm to be 2.6 μm. The average $\langle \cos\theta \rangle$, where θ is the scattering angle, is equal to 0.93, which can be used to calculate the transport mean free path l. Using the prediction of Akkermans and Maynard for the half angle of the cone, one would predict for this solution a half angle of 3.3 mrad which is in satisfactory agreement with the experimental number of 1.6 mrad.

In a simple picture the scattering of vector waves is described with the help of transfer matrices. If we denote the two-dimensional polarization vector of light with $\mathbf{p}$, then the relation between the incoming and outgoing light for a certain scattering diagram (light path) can be represented by a 2×2 matrix $\mathbf{M}$, $\mathbf{p}_{out} = \mathbf{M}\mathbf{p}_{in}$. In principle the resulting light intensity should be obtained from the square of the sum over all possible light paths. Outside the cone it is allowed, however, to use an incoherent summation. Experimentally, we find that the multiple-scattering background (that is outside the cone) is totally depolarized. From this one has to conclude that $\langle M_{11}^2 \rangle = \langle M_{22}^2 \rangle = \langle M_{12}^2 \rangle = \langle M_{21}^2 \rangle$, where the triangular brackets denote a summation over all light paths. Inside the critical cone one has to add to each light path its reverse coherently. The reverse light path is obtained from a given path by reversing all its momenta. To calculate the effect of the reverse path, we use the result that the matrix for the reverse path $\tilde{\mathbf{M}}$ is given by $\tilde{M}_{ij} = M_{ji}$. Symmetry requirements result in the vanishing of all cross products: $\langle M_{ij} M_{kl} \rangle \sim \delta_{i,k}\delta_{j,l}$. If we use this result to add the two backscatter diagrams coherently, we end up with an enhancement factor which is polarization dependent. The enhancement factor is 2 for the backscattered light polarized parallel to the incident light and 1 for the perpendicular component.

Our results are in qualitative agreement with these predictions. Indeed, the observed parallel enhancement is considerably larger than the observed perpendicular enhancement. However, the enhancement factor for the parallel component is about 1.6 and thus smaller than the predicted factor of 2, while the observed enhancement of the perpendicular component of about 1.3 is larger than the predicted factor of 1.

We do not have an interpretation of the quantitative difference between theory and experiment yet. An obvious explanation would be the presence of an (elastic) phase-relaxation process; however, it is difficult to propose a reasonable candidate. A much more likely explanation is that several different classes of light paths contribute, for instance, short and long paths. More theoretical work is needed before definitive statements can be made.

In localization experiments a relevant length scale is the inelastic length. An upperbound for the inelastic length is set by the dimensions of the cell. Thus far we have no reliable estimate of the inelastic length. A long inelastic length would open up some very spectacular possibilities. Strong localization might be observed if a long inelastic length could be maintained in an effectively two-dimensional cell. Another interesting effect is the possible occurrence of large fluctuations when the diffusion of the spheres could be made slow compared to the time scale of the measurement.[12]

The occurrence of light-localization effects is probably a general phenomenon in many more random materials. We hope that our findings will stimulate new theoretical and experimental work.

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