Weak Localization and Coherent Backscattering of Photons in Disordered Media

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Coherent backscattering of waves by a disordered scattering medium is responsible for weak localization. We have directly observed this effect for the first time using visible light and concentrated aqueous suspensions of submicron-size polystyrene spheres. The scattered intensity is found enhanced by up to 75% within a narrow cone centered at the backscattering direction. As predicted by theory, the aperture of the cone is inversely proportional to the light mean-free path; the latter was controlled by the concentration of spheres. The importance of light polarization and particle size is discussed.

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The phenomenon of weak localization of electrons[1] is known to arise from interference effects in the multiple scattering of electrons by impurities.[2] In particular, the so-called coherent backscattering process provides a simple physical picture of the weak localization mechanism.[2–4] This process, which occurs not only for electrons in an impure metal but for any wave propagating in a disordered scattering medium,[4,5] can be understood as follows: Consider a plane wave of wave vector $\mathbf{k}_0$ (wavelength λ) experiencing a series of m elastic scattering events ($m > 2$) according to the sequence $\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2, \ldots, \mathbf{k}_m$, where $\mathbf{k}_i$ is the wave vector after the ith scattering. For fixed directions of incidence ($\mathbf{k}_0$) and of observation ($\mathbf{k}_m$), each such loop (index a) has a reverse counterpart (index b) given by $\mathbf{k}_0, -\mathbf{k}_{m-1}, \ldots, -\mathbf{k}_1, \mathbf{k}_m$. For the backscattering situation ($\mathbf{k}_m = -\mathbf{k}_0$), the waves associated with these two paths have equal complex amplitudes ($A_a = A_b$) when leaving the medium (at least if the scattering potential is isotropic). They interfere constructively, so that the contribution of this pair of loops to the background intensity, $4|A_a|^2$, is exactly twice the incoherent signal calculated by neglect of interference effects ($2|A_a|^2$). When $\mathbf{k}_m$ differs from $-\mathbf{k}_0$, the phase shift between the waves a and b amounts to $(\mathbf{k}_0+\mathbf{k}_m)\cdot(\mathbf{r}_1-\mathbf{r}_m)$, where $\mathbf{r}_1$ and $\mathbf{r}_m$ are the respective positions of the 1st and mth scattering center. The average interference term due to all loops with m scattering events is then nonzero—and positive—for $|\mathbf{k}_m+\mathbf{k}_0| < L_m^{-1}$, where L_m is the average diameter of these loops (which are not necessarily closed). The minimum value of L_m is the average distance L_2 between two successive scattering events, that is, the elastic mean free path l. Hence, one expects an increase of the scattered intensity from the incoherent background value by up to a factor 2 inside a cone of angular width of order λ/l centered at the backscattering direction.[6] The overall effective cross section including the effect of coherent multiple scattering is thus larger than the classical one (σ_0) by a quantity of order $(\lambda/l)^2\sigma_0$ (in three dimensions) and the transport coefficient associated with the wave has a relative correction $\sim -(\lambda/l)^2$, a result confirmed by more sophisticated diagrammatic expansions.[7] While extensive measurements of conductivity and magnetoresistance in two- and three-dimensional normal metals have confirmed this picture of the weak localization,[8] the phenomenon of coherent backscattering had never been directly observed until now. However, as already stressed, this classical effect should occur for any wave propagating in a medium of randomly distributed elastic scatterers, and recent theoretical papers have urged experiments on this[4] and related[9] subjects. We report here the first observation of the coherent backscattering effect.

We have investigated the multiple scattering of visible light from an argon laser by aqueous suspensions of submicrometer-size monodisperse polystyrene spheres. These beads essentially do not absorb visible light; hence the scattering is purely elastic. Beads of diameters $d = 0.109$, 0.35, and 0.46, and 0.8 μm were used, covering the crossover from pure Rayleigh scattering (for d values much smaller than the wavelength in water $\lambda = 0.515/1.33 = 0.387$ μm) to strongly angle-dependent Rayleigh-Gans scattering (for larger diameters). Note that the beads can be considered stationary on the time scale of the passage of light through the loops.[10] This is a necessary condition for the existence of the coherent backscattering effect. The volume-fraction of beads, as purchased from Sigma Chemicals Corp., is 10% and was stepwise lowered by successive dilution.

The experimental setup is shown in Fig. 1. The horizontally or vertically linearly polarized laser beam passes first a 2-m focal distance lens, used in order to reduce the beam divergence to somewhat less than 1 mrad. A fraction of the beam is reflected onto the

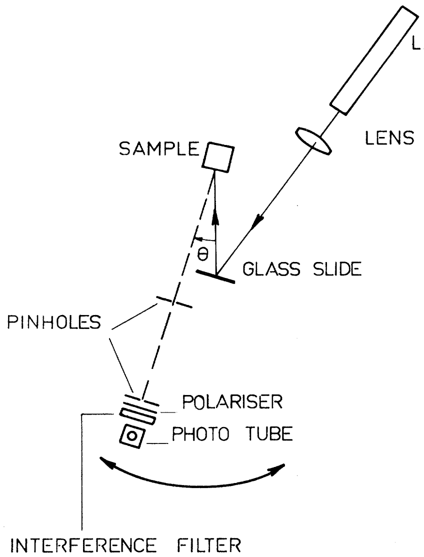

FIG. 1. Experimental setup for the study of backscattering (not on scale).

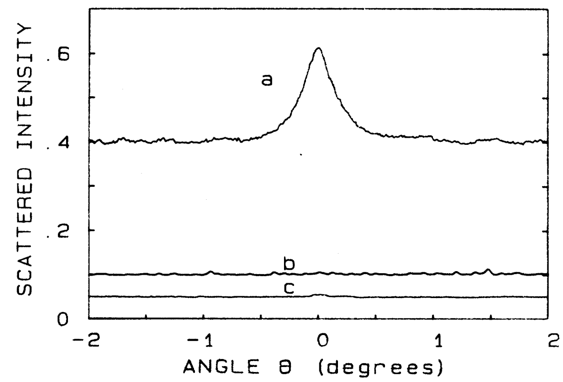

FIG. 2. Angular dependence of the scattered light intensity (curve *a*) by an aqueous suspension of 0.46-μm-diam polystyrene beads (solid fraction 10%), (curve *b*) by the same cell filled with water, and (curve *c*) in the absence of any cell. For these curves, no analyzer was used; scales are identical, but curves *b* and *c* are shifted by 0.1 and 0.05 vertical units, respectively.

sample by a thin glass slide (0.1-mm-thick microscope coverslip). The typical power incident on the sample is 10 mW. The sample itself is contained in a 10×10-mm^2 standard rectangular spectrometer quartz cell. For all samples used, the sample thickness as well as the beam diameter (about 2 mm) were much larger than the elastic mean free path l, i.e., the sample could be considered semi-infinite. The point of incidence of the beam on the sample is located at the center of a goniometer table. Two 0.8-mm-diam pinholes mounted on the goniometer arm at distances of 0.25 and 1 m, respectively, from the cell define the direction of detection within an angular resolution of about 3 mrad. Usually a linear-polarization analyzer is located behind the second pinhole. The scattered light then passes through a 3-nm-width interference filter, and the intensity is measured by a photomultiplier tube.

Figure 2 shows the scattered intensity obtained from an angular scan around the backscattering direction ($\theta=0$) for a suspension of 0.46-μm-diam beads at a solid fraction of 10%. We observe a sharp peak centered at backscattering (curve *a*). Signals obtained from the same cell filled with water (*b*) and without cell (*c*) are given for comparison. This shows that the peak does not result from some additional scattering by the quartz cell or the beam splitter. Figure 3(a) shows the evolution of the peak as the density n of beads is decreased. At high density, its height is found roughly constant, while its width W decreases with decreasing n (curves 1 and 2). This is consistent with the theoretical prediction $W\propto \lambda/l$, i.e., $W\propto n$ (for negligible interparticle interaction). At smaller n the peak height decreases (curve 3) because the peak width becomes close to our instrumental width, which was directly measured by replacenemt of the sample with a properly aligned mirror (curve 5). Finally, for a sufficiently small density, no measurable peak could be seen (curve 4). In order to test the scaling prediction $W\propto \lambda/l\propto n$ throughout the whole n range investigated, it is appropriate to define an effective width W_{eff} by the ratio of the area under the peak to the incoherent intensity. Indeed, this quantity, which, unlike the measured peak width, does not depend on the resolution, should vary with the density like the intrinsic peak width (since the intrinsic relative peak height is expected constant). Figure 3(b) shows that W_{eff} is proportional to n. These data demonstrate unambiguously that the observed peak is due to coherent backscattering.

In order to make a more quantitative comparison between our data and theory, we determined the elastic mean free path l of our samples. To this end, we have measured their optical transmission by using a commercial spectrophotometer. The fraction of the light transmitted without being scattered varies with the sample thickness D as $e^{-D/l}$. For the sake of precision, values of l have been extensively measured with use of various cells ($0.1\ \mathrm{mm} < D < 1\ \mathrm{mm}$) and low solid fractions (10^{-3} to 10^{-2}, depending on the bead size).[11] Under these conditions, l is relatively large ($\sim 300\ \mu$m) and the l values of the original solutions (solid fraction $\sim 10^{-1}$) were deduced by simple multiplication with the appropriately measured dilution factor. (Several measurements on the original solutions have also been carried out, and their results, although less precise, agree with those obtained by the above procedure.) Table I shows the experimental mean free path compared with the corresponding effective peak width W_{eff} for the four diameters of beads used (the solid fraction was about 10% in all four

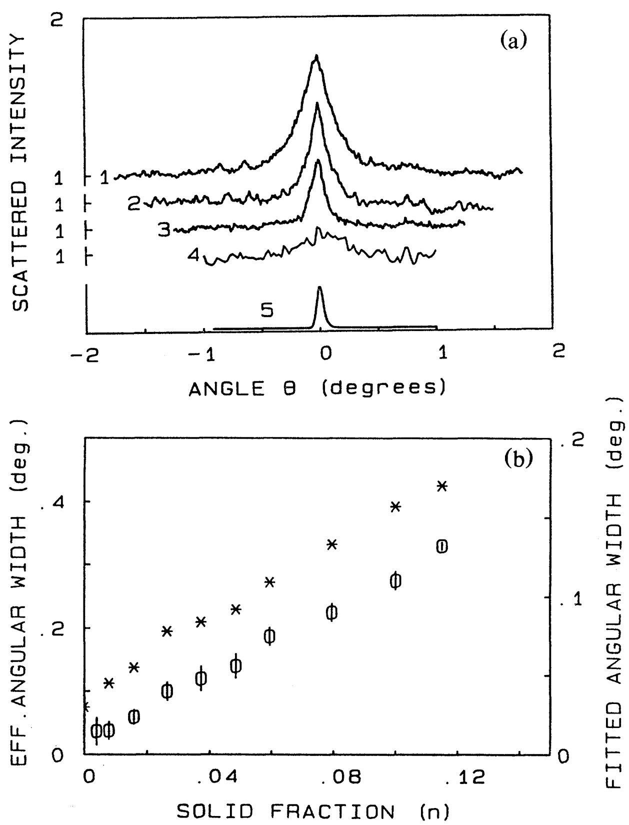

FIG. 3. (a) Dependence of the coherent backscattering effect on the solid fraction (n) of beads: curve 1, $n=0.11$; curve 2, $n=0.06$; curve 3, $n=0.026$; and curve 4, $n=0.004$. The bead diameter is 0.46 μm. The analyzer is parallel to the vertical incident polarization, VV. For each curve, the intensity is normalized to the incoherent background intensity (i.e., the intensity outside the peak). Curve 5: instrumental width (vertical scale is arbitrary). (b) Concentration dependence of the effective width (circles) of the coherent backscattering in VV configuration. Also plotted is the experimental width (asterisks), determined by fitting of the experimental curves by a Lorentzian line shape (this gives good results except near $\theta=0°$). The width extrapolates at zero density to the instrumental width.

cases). The simple relation $W_{\rm eff} \propto 1/l$ does not hold when the bead diameter d rather than the density n is changed. This is certainly related to the fact that the larger beads scatter most of the light in the forward direction. As d increases, the intensity scattered by one single bead becomes restricted to a cone of approximate aperture λ/d. For large d, we thus expect the minimum width of a loop to be proportional to dl/λ rather than to l only. As seen from the last column of Table I, the observed d dependence of $W_{\rm eff}$ seems consistent with this crude argument.

Since the current theory[4] only applies to the case of scattering of a scalar wave from pointlike particles, an absolute comparison with our data is meaningful at

TABLE I. Effective aperture $W_{\rm eff}$ of the coherent backscattering cone for various diameters d of beads. λ and l are the light wavelength (0.387 μm) and mean free path, respectively. Polarizers configuration is VV.

d (μm)	l (μm)	$W_{\rm eff}$ (mrad)	$lW_{\rm eff}$ (μm)	$dlW_{\rm eff}/\lambda^2$
0.109	33	2.2	7.2×10^{-2}	4.4×10^{-2}
0.35	5	4.4	2.2×10^{-2}	5.1×10^{-2}
0.46	2.8	4.9	1.4×10^{-2}	4.4×10^{-2}
0.80	1.9	4.1	0.76×10^{-2}	4.0×10^{-2}

best for the smallest beads used ($d=0.109$ μm). The theoretical correction to the diffusion coefficient of the wave[4,7] can be used to predict a value $3/\pi(\lambda/l)^2$ for the ratio η of the volume confined under the coherent backscattering cone to the incoherent intensity. With use of measured mean free path $l=33$ μm, this gives $\eta=1.3\times10^{-4}$. When extracting η from our data by rotating the peak around the backscattering direction, we find $\eta=0.2\times10^{-4}$. In terms of angular width, theory and experiment differ by a factor $6^{1/2}\sim2.4$, which is not surprising, considering that (i) the distance between scatterers is not large compared to the wavelength as assumed by the theory[4,7]; (iii) even for pointlike scatterers, Rayleigh scattering is not isotropic, as (iii) light is not a scalar wave. As outlined in the following section, polarization generally plays an important role, despite the fact that it has been previously considered irrelevant for weak localization phenomena.[5,12]

In Fig. 4, curve VV gives the backscattered signal from the same sample as in Fig. 2, when both polarizer

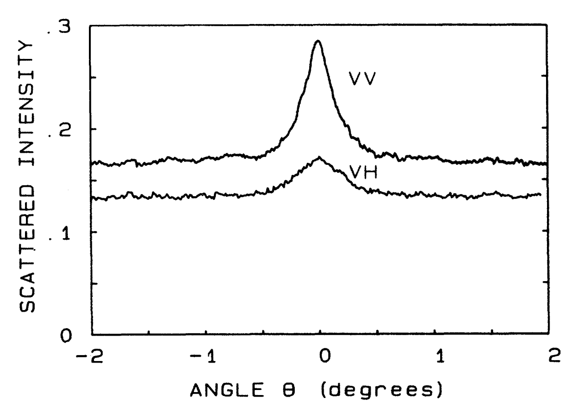

FIG. 4. Polarization dependence of the coherent backscattering effect for 0.46-μm-diam beads at a solid fraction of 10%. Directions of polarization are vertical-vertical (VV) and vertical-horizontal (VH). Curves are plotted at the same scale.

and analyzer are oriented parallel to each other and normal to the horizontal goniometer plane. Curve VH shows a scan with vertical polarizer and horizontal analyzer (for the studied case of small backscattering angles, VV is equivalent to HH and VH to HV). The incoherent part of the signal is found to be roughly the same in both cases. Hence, as expected, this light is depolarized by the multiple scattering. The height of the peak, however, is much larger to VV than for VH ($\sim 75\%$ above the incoherent background versus $\sim 25\%$). We think that this is due to the angular dependence of the light scattered from each individual sphere: The scattered amplitude in direction $\mathbf{k}'$ is proportional to the projection of the incident polarization $\mathbf{P}$ onto the plane normal to $\mathbf{k}'$, i.e., proportional to $\mathbf{k}' \times (\mathbf{P} \times \mathbf{k}')$. Hence the amplitudes corresponding to the loop $(\mathbf{k}_0, \mathbf{k}_1, \ldots, \mathbf{k}_m, \mathbf{k}_0)$ and its inverse are proportional to

$$\mathbf{k}_0 \times [\mathbf{k}_0 \times [\mathbf{k}_m \times [\mathbf{k}_m \cdots [\mathbf{k}_1 \times [\mathbf{k}_1 \times \mathbf{P}]] \cdots]]]$$

and

$$\mathbf{k}_0 \times [\mathbf{k}_0 \times [\mathbf{k}_1 \times [\mathbf{k}_1 \cdots [\mathbf{k}_m \times [\mathbf{k}_m \times \mathbf{P}]] \cdots]]],$$

respectively; these two expressions are generally not equal. However, their projections on the incident polarization $\mathbf{P}$ are easily seen to be strictly identical, so that full coherence between both loops is maintained for VV and HH (or any other parallel orientation). This is why the concentration dependence of the effect has been studied in such a configuration VV. On the other hand, the coherence between the two loops is not perfect for nonparallel polarizations (VH, HV, and others). Although a theoretical calculation for crossed polarizations seems complicated, the observation of a small effect (Fig. 4, curve VH) shows that the coherence is slightly positive in this case. Note that for both configurations VV and VH, the peak width is about the same. Finally, the fact that even for VV the experimental peak height is not exactly twice the incoherent background may be due to the finite angular resolution of our experiment.[13] Additional experiments should clarify this point.

In summary, our optical experiment is the first direct observation of the coherent backscattering effect for waves propagating in a disordered medium. Both order of magnitude of the aperture of the backscattering cone and its dependence on the density of scatterers are in fair qualitative agreement with the theoretical predictions. We believe that light is perhaps more convenient for further studies (such as the influence of absorption) than other previously suggested waves (ultrasound, neutrons, and microwaves).

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Note added.—A weak ($<15\%$) optical backscattering enhancement has been reported but differently discussed by Ishimaru *et al.*[14]

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[6]Interference effects in multiple scattering have been previously considered by D. A. de Wolf, IEEE Trans. Antennas Propag. **19**, 254 (1971). A backscattering cone was also predicted, but with an erroneous aperture, probably because only a special kind of loops was taken into account.

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[8]In particular, the weak-localization effect implies a characteristic oscillation of the magnetoresistance, first observed by D. Yu. Sharvin and Yu. V. Sharvin, Pis'ma Eksp. Teor. Fiz. **34**, 5, 285 (1981) [JETP Lett. **34**, 5, 273 (1981)]. For a recent review see G. Bergman, Phys. Rep. **107**, 1 (1984).

[9]P. W. Anderson, to be published.

[10]In the most unfavorable case of our smallest (i.e., fastest) beads ($a = 0.109$ μm) and largest loops ($L_m \sim$ cell size ~ 1 cm), the time for thermal diffusion over a distance $\lambda/2$ is about 1 msec as compared to a time of 10^{-11} sec for random travel of light through L_m (for $l = 30$ μm).

[11]Note that over the whole visible range $\lambda = 0.4$–0.8 μm (in vacuum), the measured mean free path was found in agreement with the theoretical value, deduced from the bead scattering cross section (computed in the Rayleigh-Gans regime) and from an original solid fraction of beads of 10%. This shows that the inelastic mean free path is at least 100 times larger than the elastic (Rayleigh) mean free path.

[12]S. John, H. Sompolinski, and M. J. Stephen, Phys. Rev. B **27**, 5592 (1983).

[13]It should be kept in mind that the contribution of single scattering to the backscattered intensity is not doubled by the coherence effect. For small beads, this also should reduce the peak height below twice the incoherent background value. However, for the 0.46-μm-diam beads used for Fig. 4, the estimated contribution from single scattering to the incoherent intensity is negligible, as confirmed by the observed depolarization of the signal outside the peak. Hence this contribution cannot be responsible for the experimental reduction of the peak height.

[14]A. Ishimaru *et al.*, J. Opt. Soc. Am. A **8**, 831, 836 (1984).