Experimental Evidence for Recurrent Multiple Scattering Events of Light in Disordered Media

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The coherent backscattering of light from disordered samples was studied with a new technique, which allowed for a very accurate determination of the enhancement factor in exact backscattering. Using circularly polarized light, the enhancement factor in the polarization conserving channel was found to be 2.00 ± 0.01 in the weak scattering regime and was found to drop gradually below this value for stronger scattering. We argue that this behavior of the enhancement factor is the first direct experimental manifestation of recurrent multiple scattering events.

The interest in the field of multiple light scattering in disordered media has gone through an enormous revival after it was recognized that interference effects can be important, even after many scattering events. Several interference phenomena like weak localization [1] and short and long range spatial correlations in intensity fluctuations are the focus of today's research [2]. The phenomenon of weak localization is sometimes seen as a precursor to strong or Anderson localization of light [3], which would be the light counterpart of Anderson localization of electrons. The approach to a strong localization transition in a medium with strong disorder manifests itself as a reduction of the diffusion coefficient, eventually to zero. Localization in three dimensions of microwaves has been reported [4]. It is still a challenge to find experimental evidence for strong localization of light.

In all multiple scattering phenomena that have been observed so far, recurrent scattering of light waves can be disregarded. Recurrent events are events in which a wave is scattered more than once by the *same* scatterer. The neglect of recurrent scattering will be referred to as the self-avoiding multiple scattering (SAMS) approximation, and is valid in the weak scattering limit. However, for strong scattering recurrent events can become important. They could play a role in strong localization of light.

In order to find evidence for the existence of recurrent scattering we studied coherent backscattering or weak localization of light. Coherent backscattering manifests itself in the form of an enhancement of the intensity in the back direction for the light scattered from a disordered sample. This enhancement originates from constructive interference between multiply scattered amplitudes and their time reversed counterparts. Moving away from the exact backscattering direction, phase differences develop that average out this interference effect. The result is a "cone" of enhanced backscattering on top of the diffuse background, which has a width of the order of $(k\ell)^{-1}$ where ℓ is the (transport) mean free path of the light in

the medium and $k = 2\pi/\lambda$ is the magnitude of the wave vector of the light in vacuum [1].

In the SAMS approximation the enhancement factor in the helicity conserving channel is exactly 2 because every scattered wave has a distinct time reversed counterpart of equal amplitude. (In the polarization conserving channel of linearly polarized light the enhancement factor is somewhat lower because a single scattering contribution is present that does not contribute an interference term.) We have studied the enhancement factor, because in the strong scattering regime the enhancement factor could be influenced by recurrent scattering events.

In this Letter we report on coherent backscattering experiments that show an enhancement factor in the helicity conserving channel of 2.00 ± 0.01 for weak scattering. The enhancement factor decreases below 2.00 once the mean free path becomes of the order of one wavelength. We will argue that this is a manifestation of recurrent scattering events. A calculation of the first order density correction on the enhancement factor confirms this interpretation.

An accurate experimental determination of the enhancement factor requires much care. Stray light from surfaces and optical components was carefully eliminated. Where windows were used to index match the sample, these were thick and Ar coated on the front side, as to eliminate backreflections of scattered light onto the sample. After eliminating common experimental artifacts, the (slightly) angular dependent response of the standard setup [2] still can introduce a systematic error in the cone shape. Therefore, we modified it such that the optical components rotate only relative to the reference frame and not relative to each other. In this way a completely flat response over a large scanning range (500 mrad) could be achieved. Circularly polarized light was used, and the helicity conserving channel was detected [5].

The samples were prepared starting from finely divided TiO_2, ZnO, or $BaSO_4$. The first two were ground as a

suspension in methanol or chloroform to break possible clusters and were allowed to dry on plastic sheet. Some dry samples had $\lesssim 1\%$ PMMA added to them as a binder. The TiO_2 was also used to form a suspension in 2-methylpentane 2,4-diol or methanol. The interface of some of the dry TiO_2 samples and the TiO_2 suspension was largely index matched using a thick glass window.

As is known from previous experiments and confirmed by our experiments, the angular intensity distribution of both diffuse background and backscattering cone in the helicity conserving channel are very well described by diffusion theory [6]. In the *weak scattering limit*, the total normalized angular intensity distribution is given by [6]

$$I(\theta, k\ell) = \frac{\gamma_c(\theta, k\ell) + \gamma_\ell(\theta)}{\gamma_\ell(0)},$$

with ℓ the (transport) mean free path, k the wave vector of the incident light in vacuum, θ the scattering angle, and γ_c and γ_ℓ the bistatic coefficients for cone and diffuse background, respectively. [Note that $\gamma_c(0, k\ell) = \gamma_\ell(0)$.] The (weak) angular dependence of the diffuse background is a kinematic factor (light emerging under a larger angle has to cover a larger distance in the sample) and is not a function of $k\ell$. The expression for the incoherent background was tested separately by comparing it to the backscattered intensity from a sample with a very long mean free path. Perfect agreement was found.

As we want to study the strong scattering regime, we want to allow for (almost angular independent) beyond SAMS contributions to the intensity. If we make the realistic assumptions that (a) these contributions have the same (kinematic) angular dependence as γ_ℓ and (b) the shape of the cone remains the same, we find for $I(\theta, k\ell)$ in the *strong scattering regime*,

$$I(\theta, k\ell) = \frac{[1 - \eta(k\ell)]\gamma_c(\theta, k\ell) + \gamma_\ell(\theta)}{\gamma_\ell(0)}, \qquad (1)$$

where η is the relative contribution of beyond SAMS events. The enhancement factor E may now differ from 2, $E(k\ell) \equiv I(0, k\ell) = 2 - \eta(k\ell)$.

In Fig. 1 two examples of recorded backscattering cones are compared. The broader cone corresponds to the sample with the shortest mean free path. The solid line in Fig. 1 is $I(\theta)$ from Eq. (1); the dashed line is the corresponding $\gamma_\ell(\theta)$. The data are normalized to $\gamma_\ell(0)$. We have calculated the residues by substracting measured and theoretical curves, and found no structural deviation whatsoever. This supports assumptions (a) and (b), and shows that Eq. (1) describes the cone accurately.

If we compare the top of both cones in Fig. 1, we see that only the enhancement factor of the broader cone is significantly smaller than 2. In Fig. 2 the enhancement factor E is plotted against the inverse full width at half maximum of the cone W^{-1} for various measurements. The mean free path ℓ is proportional to W^{-1}. The sets of data in Fig. 2 include results obtained using various sample materials, sample preparation methods,
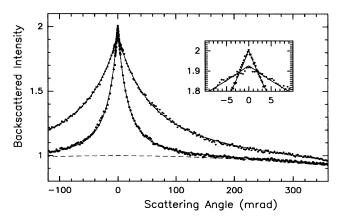

FIG. 1. Backscattered intensity plotted against the scattering angle. Zero corresponds to exact backscattering. Narrow cone: $BaSO_4$ sample, (scaled) mean free path $k_{med}\ell = 22.6 \pm 1.0$. Broad cone: TiO_2 sample, $k_{med}\ell = 5.8 \pm 1.0$. Angular resolution: 0.25 mrad. Solid and dashed lines are, respectively, cone and diffuse background from diffusion theory. The inset shows the top of both cones. The enhancement factor of the broad cone clearly deviates from 2.00.

and wavelengths. Two classes of measurements, i.e., with index-matched and with non-index-matched sample interfaces, are shown separately. Notice that in both series E is 2.00 ± 0.01 for large W^{-1} and drops below 2.00 for very small W^{-1} (i.e., for small ℓ). We have checked our method of determining E in various ways: by using a different theoretical cone shape [7], by trying to map theoretical and experimental cones forcing $E = 2$ keeping other parameters free, and by cutting down the scanning range at large W^{-1}. The lowering of E at small W^{-1} survived all these tests.

We are interested in the dependence of the enhancement factor on $k_{med}\ell$ with k_{med} the wave vector of the light inside the sample. For the index-matched series,
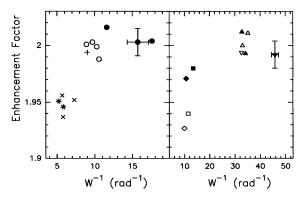

FIG. 2. Enhancement factor as a function of W^{-1}, where W is the width (FWHM) of the cone. The left graph: series with index-matched sample interface, right graph: series with non-index-matched interface. For a definition list of used symbols see the caption of Fig. 3.

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ℓ can be found from W^{-1} by $\ell = 0.7k^{-1}W^{-1}$ [6]. For the non-index-matched series we must take into account the narrowing of the cone due to internal reflection [8]. From the estimated bulk refractive index (n_{med}) (TiO_2: 1.35, $BaSO_4$: 1.2, ZnO: 1.2) [9], the reflectivity R of the sample interface is calculated [8]. From R a narrowing by a factor 0.62 ± 0.07 for TiO_2 and 0.76 ± 0.10 for $BaSO_4$ and ZnO is found [8].

We have plotted the enhancement factor against $k_{med}\ell$, with $k_{med} = n_{med}k$, in Fig. 3. The enhancement factor is 2.00 ± 0.01 for $k_{med}\ell \gtrsim 10$ and gradually drops below 2.00 for smaller values of $k_{med}\ell$ (strong scattering). In the SAMS approximation the enhancement factor in the helicity conserving channel is exactly 2 because first and last scattering will always occur on different scatterers and every event has a time reversed counterpart of equal amplitude. Therefore a deviation from 2 at small $k\ell$ must arise from other than self-avoiding (i.e., recurrent) scattering events. We propose two new classes of recurrent events that can reduce the enhancement factor at strong scattering. The first we call the class of recurrent "loop" events. These are recurrent multiple scattering events where the scattered wave has a distinct time reversed counterpart, but in which first and last scattering occurs on the same scatterer. The interference contribution of such events has the same angular dependence as their background contribution, and is therefore *observed* as background. The second we call the class of recurrent "folded" events. These are recurrent multiple scattering events that are identical to their time reversed counterpart. They only contribute to the diffuse background and give no interference term. Both classes influence the enhancement factor.

In the following, we will calculate the first order density correction on the enhancement factor that is due to recurrent scattering between two particles, and show that this correction indeed reduces the enhancement factor. In general, the total backscattered intensity is determined by the total four-point vertex $\Gamma(r_1, r_2, r_3, r_4)$ being the solution of the Bethe-Salpeter equation [10]. Following Ref. [10] this vertex is written as $\Gamma = R + \overline{R} + S$. Here R is the reducible vertex which obeys long range diffusion and $\overline{R}$ is *self-consistently* obtained from R by time reversing either its bottom or top line. The bistatic coefficients associated with R and $\overline{R}$ are called $\gamma_r(\theta)$ and $\overline{\gamma}_r(\theta)$. Since R is long range, $\overline{\gamma}_r(\theta)$ dephases rapidly at angles $\theta > (k\ell)^{-1}$ away from backscattering, and the sum of $\gamma_r(\theta)$ and $\overline{\gamma}_r(\theta)$ gives rise to the backscattering cone. Time-reversal symmetry guarantees that $\overline{\gamma}_r(0) = \gamma_r(0)$ (exact backscattering) for the polarization conserving channel [11] yielding an enhancement factor of ex-
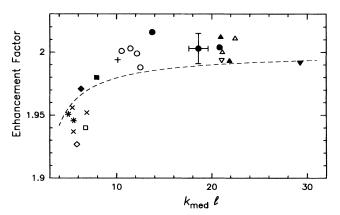

FIG. 3. Enhancement factor plotted against $k_{med}\ell$. Filled symbols: $\lambda = 632.8$ nm, others (open symbols and stars): $\lambda = 514.0$ nm. The dashed line is the calculated enhancement factor if recurrent scattering from two particles is incorporated. Measurements with non-index-matched interface: $\triangle = BaSO_4$, $\nabla = ZnO$, $\square = TiO_2$, $\Diamond = TiO_2$ with PMMA. Measurements with index-matched interface: $\bigcirc = TiO_2$ in 2-methylpentane 2,4-diol, $\times$, $*$ $= TiO_2$, $+ = TiO_2$ in methanol. The measured enhancement factor drops below 2.00 for $k_{med}\ell \leq 10$.

actly 2. This notion follows from the self-consistent treatment required by time-reversal symmetry. It is not necessary to make the familiar "ladder + most-crossed" approximation.

The set of events covered by S is closed under a time-reversal operation of either bottom or top line, $S = \overline{S}$. Furthermore, this set does not contribute to long range diffusion. The bistatic coefficient γ_s that corresponds to S consists of ordinary single scattering γ_{s_0} and beyond SAMS contributions $\delta\gamma_s$ ($\gamma_s \equiv \gamma_{s_0} + \delta\gamma_s$). The events in S are short range on the scale of ℓ and thus give rise to an (almost) angle independent contribution to the intensity. In that case, the enhancement factor becomes

$$E = \frac{\gamma_r + \overline{\gamma}_r + \gamma_s}{\gamma_r + \gamma_s} = 2 - \frac{\gamma_s}{\gamma_r + \gamma_s},$$

where the bistatic coefficients are taken at $\theta = 0$. To arrive at concrete results we will use Rayleigh vector scatterers and evaluate this equation in the helicity conserving $(++)$ channel. Because in the helicity conserving channel $\gamma_{s_0} = 0$, the enhancement is (to leading order in the density) given by $E = 2 - \delta\gamma_s/\gamma_r(0)$, where $\gamma_r(0) = 1.736$ [12]. This means we have to calculate $\delta\gamma_s$.

In lowest order of the density $\delta\gamma_s$ consists of recurrent scattering between only two scatterers [13]. The full scattering amplitude for a system of two Rayleigh point scatterers (1 and 2) separated by r is given by $f(kg, k'g') = g \cdot F_{kk'}(r) \cdot g'^*$ with [14]

$$F_{kk'}(r) = \frac{2}{1 - t_1 t_2 G(r)^2}\left[\frac{t_1 \exp(if \cdot r) + t_2 \exp(-if \cdot r)}{2} + t_1 t_2\, G(r)\cos(b \cdot r)\right].$$

Here, $\mathbf{f} \equiv (\mathbf{k} - \mathbf{k}')/2$, $\mathbf{b} \equiv (\mathbf{k} + \mathbf{k}')/2$, and t_i is the t matrix of particle i [15]. The incoming and outgoing wave vectors and polarization vectors are, respectively, $\mathbf{k}$ and $\mathbf{k}'$ and $\mathbf{g}$ and $\mathbf{g}'$. The vector Green's function $\mathbf{G}(\mathbf{r})$ can be separated into a longitudinal part $Q(r)$ and a transverse part $P(r)$ with P and Q known [14]. For the helicity conserving channel at exact backscattering we put $\mathbf{k} = -\mathbf{k}' = k\hat{\mathbf{z}}$ and $\mathbf{g} = \mathbf{g}'^* = (\hat{\mathbf{x}} + i\hat{\mathbf{y}})/\sqrt{2}$. The cross section for recurrent events $\sigma_s(\mathbf{r})$ is found by substracting the (bare) two particle ladder and most-crossed contributions from the total two particle cross section $|f(\mathbf{k}\mathbf{g}, -\mathbf{k}\mathbf{g}^*)|^2$,

$$2\sigma_s(\mathbf{r}) := |f(\mathbf{k}\mathbf{g}, -\mathbf{k}\mathbf{g}^*)|^2 - 4|\mathbf{g} \cdot \hat{\mathbf{r}}|^4 |t_1 t_2|^2 \left| \frac{e^{ikr}}{4\pi r} \right|^2 .$$

Given a volume V with front surface A, containing N scatterers, the bistatic coefficient $\delta\gamma_s$ can be calculated from $\sigma_s(\mathbf{r})$. In the limit $N/V \rightarrow n$ this yields [14]

$$\delta\gamma_s = \frac{3}{4} n \int d\mathbf{r} \frac{\langle \sigma_s(\mathbf{r}) \rangle}{\langle |t|^2 \rangle} + \mathcal{O}(n^2 \ln n) \sim (k\ell)^{-1}.$$

(Note that $n = 6\pi/\ell\langle |t|^2 \rangle$.) The particles of our samples are strongly polydisperse so we average t_1 and t_2 independently over a frequency window that is large compared to the width of the resonance. The integral over $\mathbf{r}$ was performed numerically. The outcome is plotted in Fig. 3 as a dashed line. We see that the recurrent two particle events indeed manifest themselves as a reduction of the enhancement factor at short mean free paths.

In conclusion, our coherent backscattering experiments show an enhancement factor of 2.00 ± 0.01 in the helicity preserving channel for mean free paths much larger than the wavelength, and a decrease below this value for very strong scattering. We have demonstrated that this is a manifestation of recurrent multiple scattering. Future experiments could involve the search for more efficient scatterers to yield samples with smaller $k\ell$. This way the strong scattering regime could be explored further.

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