

An accurate technique to record the angular distribution of backscattered light

D. S. Wiersma^{a)} and Meint P. van Albada

FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

Ad Lagendijk

FOM-Institute for Atomic and Molecular Physics, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands and Van der Waals-Zeeman Laboratorium, Valckenierstraat 65-67, 1018 XE Amsterdam, The Netherlands

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We report on a new technique to record the angular distribution of light which is scattered from a (disordered) material around the backscattering direction. The technique is accurate over a large scanning range (500 mrad) which includes the exact backscattering direction, and the angular resolution is high (100 μ rad). The technique is particularly suitable for the study of coherent backscattering from very strongly scattering random media, but it can be applied in any situation where the angular distribution of backscattered light is studied. It allowed us to measure for the first time the theoretical value of two of the enhancement factor in coherent backscattering. © 1995 American Institute of Physics.

I. INTRODUCTION

The technique to record backscattered light which we present in this article was developed for the study of coherent backscattering from random media. The experimental study of coherent backscattering puts high demands on the accuracy of a setup. A technique suitable for coherent backscattering experiments can therefore be applied to any situation where the angular distribution of backscattered light must be known accurately, for instance for backscattering experiments from rough surfaces or for the characterization of retroreflectors. We start with an introduction on coherent backscattering and on experimental configurations that have been used previously in the study of this phenomenon. In the next section, we will introduce the new technique which we call "off-centered rotation."

A. Coherent backscattering

Coherent backscattering is a general effect for waves scattered by random media.¹⁻³ It originates as follows. Consider a disordered sample that is illuminated by a (spatially broad) light beam. A partial wave that propagates over some distance through the sample and then leaves the illuminated spot in the backscattering direction will have a counterpropagating counterpart that follows the same path in the opposite direction. These counterpropagating partial waves travel over the same distance in the sample and will therefore interfere constructively in the exact backscattering direction. Moving away from the exact backscattering direction, a phase difference will develop between the counterpropagating waves that depends on the relative orientation of the points where the waves enter and leave the sample. For the ensemble of light paths, relative phases will therefore gradually randomize. After averaging over all light paths, this leads to a cone of enhanced backscattering. In the exact backscattering direction the intensity is twice as high as expected from an

incoherent addition of the scattered waves. (This enhancement factor is precisely 2 only for circularly polarized light in the helicity conserving channel. We will come back to polarization effects later.) The width (FWHM) of the backscattering cone is proportional to λ/l with l the (transport) mean free path for light in the medium and λ its wavelength.¹⁻³

The shape of the backscattering cone reflects the path length distribution of the light inside the sample and therefore reveals information about the internal structure of the sample. If the sample consists of a random collection of small particles, the mean free path l is inversely proportional to the density n and scattering cross section σ of the particles. In that case, the width of the cone is a measure for the particle density. For more complex, e.g., spongelike, random structures, it is difficult to identify the individual scattering elements. Here the width of the cone is just a measure for the scattering strength of the material. The shape of the backscattering cone is sensitive to the sample structure at large depth because the top of the cone is determined by very long light paths that have penetrated deep into the sample (features that are due to $>10^4$ scattering events are experimentally accessible). In the theoretical case of zero absorption, the top of the backscattering cone is a cusp. If absorption is present either at large depth or throughout the sample, the contribution from the longer light paths is reduced and consequently the top becomes rounded.

B. Principle of previous setups

To study coherent backscattering, one must accurately record the angular distribution of the intensity backscattered from a sample which is illuminated by a spatially broad light beam. In the experimental studies that have been published so far, two different schemes were used. In the simplest scheme, the incoming beam was reflected onto the sample by a mirror [see Fig. 1(a)].^{4,5} The scattered light is collected with a positive lens. The detector is placed in the focal plane

^{a)}Electronic mail: wiersma@amolf.amolf.nl

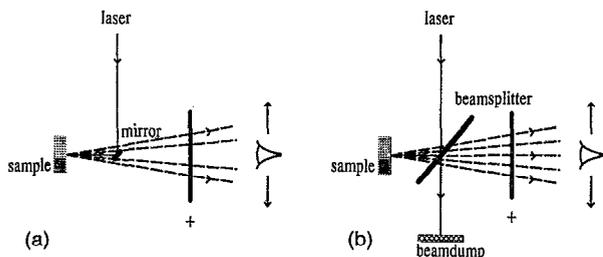


FIG. 1. Principle of setups used previously to record coherent backscattering. (a) Simplest scheme. The incoming beam is reflected onto the sample with a small mirror. The backscattered intensity can be recorded over a large angular range but the region close to exact backscattering cannot be observed. A positive lens is used to collect the scattered light. The detector is placed in the focal plane of the lens so that one scattering direction corresponds to one position of the detector. (The refraction of the waves by the lens is not shown.) (b) By using a beamsplitter, the exact backscattering direction can be observed but one has to correct for the angular dependent transmission of the beamsplitter, the positive lens, and other detection optics.

of the lens so that one scattering direction corresponds to one position of the detector. The use of a mirror allows for a determination of the scattered intensity over a large angular range. However, the mirror will mask the scattered light in and near the exact backscattering direction, so with this method the top of the backscattering cone cannot be observed. Another approach was to use a beamsplitter to reflect the light onto the sample [see Fig. 1(b)].^{2,3,6} The scattered light is recorded through the beamsplitter, so the exact backscattering direction can be monitored. Again a positive lens is used to collect the scattered light. The disadvantage of this scheme is, however, that one has to correct for the angular dependence in the transmission characteristics of the beamsplitter, the positive lens, and other detection optics (e.g., a polarizer).

In both schemes it is difficult to shield stray light in a satisfactory manner. The fundamental problem arises from parallax: the screen that is used to shield stray light must be placed at some distance in front of the sample. As the detector moves, the light path from sample to detector changes. Therefore, the field of view of the detector has to be larger than the solid angle under which it sees the illuminated area on the sample. In practice this means that an amount of stray light adds to the detected signal. This yields an extra background in the signal which is even likely to show some angular dependence. As a result the shape and enhancement factor of the backscattering cone cannot be recorded accurately.

II. EXPERIMENTAL CONFIGURATION

A. Principle of the setup

We have developed a new method, which we call off-centered rotation (OCR), to solve the experimental problems described above. The outline of the setup is drawn in Fig. 2(a). The incoming light beam is reflected via a beamsplitter onto the sample. The scattered light is collected with a positive lens ($f=1$ m). In the focus of the lens is the head of an optical fiber that transports the light to a photomultiplier tube. Detection optics, beamsplitter, and sample are placed

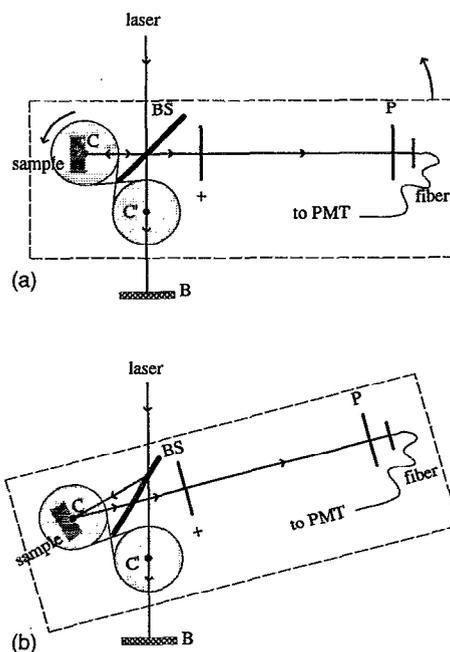


FIG. 2. (a) Outline of the setup to record the angular distribution of a backscattered intensity. The essence of the setup is that all optical components are placed on a frame which rotates around C' , which is the mirror image of the beamsplitter (BS) of the center C of the front sample surface. In this way, the path from sample to detector of the scattered light is always the same. The sample rotates around C to keep the incoming direction perpendicular to the sample surface. The scattered light is focused by a positive lens (+) on the head of an optical fiber which is placed in the focal plane of the lens. The fiber transports the light to a photomultiplier tube (PMT). The polarizer P is used to select one polarization channel of the scattered light. The laser beam transmitted through the beamsplitter is blocked by a beam dump (B), that must be of high quality as backreflections would reach the detector. (b) Same setup after rotation to another detection angle.

on a rotating frame. The center of rotation (C' is the center C of the sample surface, mirrored with respect to the plane of the beamsplitter. Figure 2(b) shows the setup after rotation. The incoming beam is directed at C , so after rotation it still arrives at the center of the sample surface C' . With respect to the frame, the incoming direction has changed and the direction of detection is still the same. By rotating the sample around C , the sample surface is kept at a constant angle with respect to the incoming beam. The rotation is obtained by placing the sample on a (rotatable) plateau, which is connected by a twisted band to a disk of the same size that is fixed to the laboratory frame. Thus the incoming direction on the sample is fixed and the angular distribution of the scattered light is recorded.

The scattered light always follows the same path through the beamsplitter and the detection optics. This is the major advantage of our setup: it avoids any corrections for angular dependence in the transmission of the components. The one remaining angular dependence in the setup is in the reflection of the incident beam by the beamsplitter, but this dependence can easily be measured to any desired accuracy by replacing the sample with a detector.

In order to observe a coherent backscattering cone it is necessary to average over many configurations of the sample. This eliminates the speckle pattern which is formed

if a coherent beam is scattered by a (stationary) random sample.⁶ In a fluid sample, Brownian motion of the particles provides the required ensemble averaging. Solid samples are mounted on a small motor which spins them around their axis. The most efficient ensemble averaging is obtained if the sample surface is not exactly perpendicular to the axis of rotation, so that the sample “wiggles.”

B. Polarization

The polarization of the incoming laser beam is linear, and a polarizer is mounted in front of the optical fiber head. The parallel or perpendicular polarization channel in the scattered light is selected by means of this polarizer. If experiments with circularly polarized incident light are desired, a quarter waveplate is mounted in front of the sample to convert the incoming linear polarization to circular polarization. The two circular components of the outgoing scattered light are converted back by this quarter-waveplate to mutually perpendicular linear components. The polarizer in front of the fiber is used in that case to select the linear polarization that corresponds to either of the helical channels of the scattered light.

In experiments with circularly polarized light, the incident and scattered waves will pass the quarter waveplate under necessarily different angles. Since only at perpendicular incidence the relative phase shift corresponds to exactly a quarter of a wave, some mixing of polarizations will occur in directions that are far from exact backscattering. (Note that a setup with two quarter-waveplates—one in the incident beam, and one in front of the detector—is not possible in view of angular dependent polarization characteristics of the beamsplitter.) We used a zero-order quarter-waveplate because its angular tolerance is far greater than that of a multiorder one. We found that even at the largest angles studied, the intensity coupled into the opposite polarization channel remained $<1\%$. In directions far from exact backscattering, our random samples scramble the polarization almost completely. Errors from mixing of polarization due to oblique incidence in the quarter-waveplate will therefore essentially cancel. We estimate the overall error in the measured curves due to oblique incidence in the quarter-waveplate to be $\leq 0.1\%$.

C. Angular resolution and scanning range

The scanning range of the setup is determined by the size of the beamsplitter. We used a beamsplitter of 5 cm diameter, an incoming beam diameter of 5 mm, and obtained a scanning range of about 500 mrad. The angular resolution of the setup was diffracted limited by the diameter d of the incident beam. With $d=5$ mm and using visible light, this limit is $\approx 100 \mu\text{rad}$. The detection optics (50 μm core fiber with its tip at a 1 m distance from the sample) allows in principle a resolution of 50 μrad .

D. Elimination of important artifacts

Liquid samples are contained in a sample cell. The front window of this sample cell can give rise to artifacts, mainly due to back reflections from its front face. (The back face is

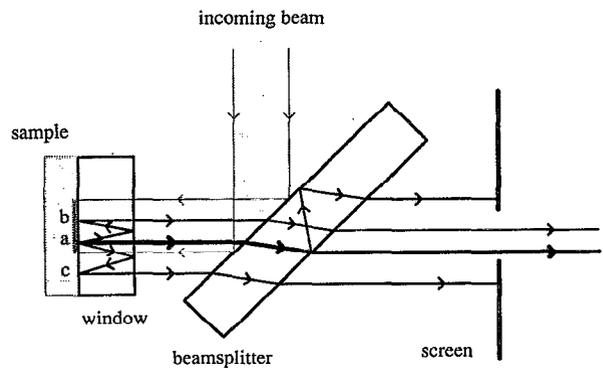


FIG. 3. Overview of some important ghost reflections that can give rise to experimental artifacts. The scattered light from the sample at point (a) can be reflected by the front face of the sample window. If the light is reflected to a point (b) inside the illuminated area, the scattered light from (b) will be recorded by the detector. If the light is reflected to a point (c) outside the illuminated area, the scattered light from (c) is blocked by the screen. The screen also blocks the ghost reflections from the rear side of the beamsplitter.

largely index matched with the sample.) We have to distinguish between light that is reflected back on the illuminated region of the sample and the light which is reflected outside this area (see Fig. 3). In the first case, the light is scattered again by the sample and contributes in backscattering to the backscattering cone. Whether these contributions can be resolved depends on what the limiting factor on the angular resolution of the setup is. If the limiting factor is the size of the illuminated area, these contributions can be resolved. They will affect the observed shape of the backscattering cone. Because the reflection will on average increase the distance between first and last scattering event, the cone will become narrower. If the angular resolution is smaller, these contributions cannot be (fully) resolved, and will lead to a lowering of the observed enhancement factor. The light that is reflected by the front window surface to regions outside the illuminated area cannot contribute to the backscattering cone because these waves have no counterpropagating counterparts. This light will however contribute to the background and will therefore also lower the observed enhancement factor.

To reduce the above effects, the glass window must be thick (1 cm) and its front side must be antireflection coated. Moreover, the second effect can be eliminated by placing a diaphragm between the beamsplitter and the positive lens. The aperture of this diaphragm is only slightly larger than the illuminated region on the sample. It is aligned such that only scattered light from the illuminated region can reach the detector. Note that this resolution is possible because in our setup the light path from sample to detector is always the same. This property of the setup also allows to shield other sources of stray light in a convenient way without the risk of partly masking the field of view of the detector during part of an angular scan (“clipping”).

A glass window can also be used to index match the front interface of a rigid sample, in order to eliminate internal reflection.⁷ The same considerations hold as for the front window of a liquid sample cell. Great care has to be taken to ensure good optical contact between window and sample

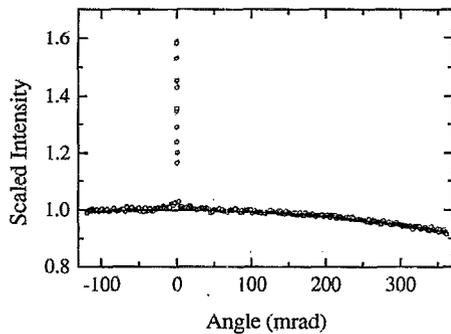


FIG. 4. Backscattered intensity plotted against the scattering angle, where zero corresponds to exact backscattering. Sample material Teflon, sample interface index matched. The intensity is scaled such that the diffuse background in the exact backscattering direction is 1. The solid line is the theoretical shape of the diffuse background from diffusion theory.

surface. We obtained good results if we coated the rear side of the glass window with collodium and pressed this side with force on the sample.

The beamsplitter is a slightly wedged thick window with a 50% reflectance coating on the front side and an antireflection coating on the back side. All (multiple) reflections that arise from the small remaining reflection of the back surface will be blocked by the diaphragm placed between the beamsplitter and the positive lens if the beamsplitter is sufficiently thick (about 1 cm). Due to its wedged shape, étalon effects in the beamsplitter are also avoided.

III. RESULTS

To check the response of the setup over a large angular range, we have recorded the backscattering from a very weakly scattering sample (i.e., a sample with a very large mean free path) (see Fig. 4). The backscattering cone of this (Teflon) sample is extremely narrow so the major part of the scan yields the (almost angle independent) diffuse background. The solid line in Fig. 4 is the diffuse background as calculated from diffusion theory.^{8,9} Agreement between data and theory is very good.

The setup enables us to determine the shape of a backscattering cone accurately over a large angular range including exact backscattering. Because the top is also resolved to high accuracy, it provides a reliable method to determine the enhancement factor of a backscattering cone. (For an application see Ref. 10.) If circularly polarized light is used and the helicity conserving channel is monitored, the theoretical enhancement factor is exactly 2.⁶ In the case of linear polarization, the scattered intensity includes a single scattering component that does not contribute to the coherent backscattering cone and therefore lowers the observed enhancement factor. Several experimental studies on the backscattering cone have been published^{1-6,9,11} but due to experimental difficulties, the reported enhancement factors have always been considerably lower than the theoretical value of 2. In Fig. 5, a backscattering cone from a sample with mean free path $l=2.1 \mu\text{m}$ is shown, which was recorded using the setup described in this article. The helicity conserving channel is monitored. From the inset we see that the enhancement factor is 2.00. The solid line is the theoretical curve:

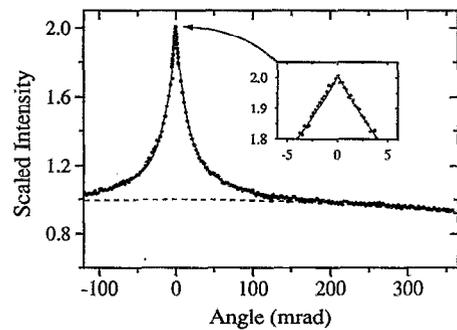


FIG. 5. Backscattered intensity plotted against the scattering angle, where zero corresponds to exact backscattering. Sample material BaSO₄ diffuse reflectance coating [Spectrafect™ (Ref. 12)], (transport) mean free path $l=2.1 \pm 0.1 \mu\text{m}$ (determined from cone width). The intensity is scaled such that the diffuse background (dashed line) in the exact backscattering direction is 1. The solid line is the theoretical backscattering cone plus diffuse background from Eq. (1). Inset: enlargement of the top of the cone. The enhancement factor is 2.00 ± 0.01 .

$$I(\theta) = \frac{\gamma_c(\theta) + \gamma_l(\theta)}{\gamma_l(0)}, \quad (1)$$

where $\gamma_c(\theta)$ and $\gamma_l(\theta)$ are the bistatic coefficients describing, respectively, the backscattering cone and the diffuse background, calculated using diffusion theory.^{8,9} Agreement between data and theory is very good.

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