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Precision determination of the line shape for coherently backscattered light from disordered solids: Comparison of vector and scalar theories

M. Rosenbluh, I. Edrei, M. Kaveh, and I. Freund
Department of Physics, Bar Ilan University, Ramat Gan, Israel
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We present high-precision measurements of the line shape of coherently backscattered light from a disordered solid which strongly supports the recent vector theory of Stephen and Cwillich, and which cannot be adequately described by current scalar theories. Three different regions have been identified in the experimentally observed line shape which reflect three different types of photon trajectories inside the multiply scattering medium. They are (i) extremely long trajectories which are affected by finite sample size, (ii) trajectories much smaller than the sample dimensions but much larger than the optical mean-free path; for these trajectories the scattering process is essentially two dimensional, and (iii) short trajectories which give rise to an asymptotic falloff which is proportional to the inverse square of the momentum transfer.

We present high-precision measurements of the coherent backscattering of light which demonstrate the necessity for a vector theory of the scattering process, such as the one presented recently by Stephen and Cwilich.[1,2] Earlier measurements[3-7] had been analyzed using a scalar theory[8] which provided a good approximation to some of the main features of the experimental data, but which failed to explain the following important observations: (i) The scalar theory predicts a 2:1 ratio for the coherent-peak–diffuse-background intensity ratio. This is at variance with all the experimental data which show a significantly smaller ratio. In many of the previous measurements this ratio could not be determined accurately because of instrumental broadening and experimental difficulties and thus these data could not distinguish between the scalar and vector theories. (ii) The scalar theory predicts that the coherent backscattering maintains the same polarization as the incident laser beam, a prediction that is again at variance with all the experimental data. (iii) The scalar theory fails to provide an adequate fit to the high-precision measurements of the line shape presented in this Rapid Communication.

We also present experimental evidence for the rounding of the coherent peak due to finite size effects, and compare these observations with theory. Finally, the high signal-to-noise ratio of our data allows us to clearly show, for the first time, the theoretically expected crossover in angular (θ) dependence from $1/\theta$ to $1/\theta^2$, due to the crossover in the effective dimensionality of the scattering process.

The coherent backscattered peak is due to the interference between the trajectories of multiply scattered photons and their time-reversed trajectories. If one ignores the vector nature (polarization) of the electromagnetic wave, all time-reversed trajectories can be expected to interfere constructively, and result in a peak backscattered relative intensity of two. (In all our discussions the intensity of the backscattered peak is measured relative to the backscattered intensity at arbitrary large angles, where the scattered intensity is uniform and is defined as one.) However, for long photon trajectories, the polarization of the scattered light can be expected to change. This leads to a weakening of the interference and a marked difference between the predictions of scalar and vector theories at small backscattering angles, which correspond to long photon trajectories. In order to study this difference we have measured the backscattered line shape with high precision, and analyzed it using both scalar and vector theories.

The experiments were performed using the apparatus described previously.[7] Backscattering data were obtained at two different laser wavelengths, $\lambda_1=514.5$ nm and $\lambda_2=457.9$ nm, using thin films of $BaSO_4$ microparticles as the scatterer. The signal was measured for light polarizations both parallel and perpendicular to the incident laser beam polarization. The data exhibit the following important features: (i) For the parallel polarized component the maximum intensity of the coherent peak relative to the diffuse background is 1.87 for both laser wavelengths. (ii) For the perpendicular component the diffuse background intensity is ~ 0.8 and the coherent peak is both very weak and very broad. (iii) The peak at its maximum is parabolic in shape for $\theta \ll \lambda/l$, while for $\theta \simeq \lambda/l$ the peak intensity falls with a $1/\theta$ dependence, and then crosses over to a $1/\theta^2$ dependence for $\theta > \lambda/l$, where l is the transport, or momentum exchange mean-free-path length.

In Figs. 1(a) and 1(b) we show the experimental line shape for the 514.5 and 457.9 nm wavelengths, respectively, as well as the calculated line shape (solid line) based on the vector theory of Stephen and Cwillich[1] [Eq. (D3)]. Also shown is the calculated line shape (dashed line) based on the scalar theory of Akkermans, Wolf, and Maynard.[8] The adjustable parameter in fitting the line shapes was l/λ, with a value of ~ 12 and ~ 13 for the vector theory and ~ 5 and ~ 6 for the scalar theory for the two wavelengths, respectively. As can be seen, the vector theory provides a far superior fit to the general line shape as well as predicting a peak maximum of 1.87 in excellent agreement with experiment. We also attempted to fit the observations with a calculated line shape based on a fully generalized scalar theory[9] by adjusting l/λ, but this also failed to produce good quantitative agreement with the experiments, especially for $\theta < 10$ mrad. The major

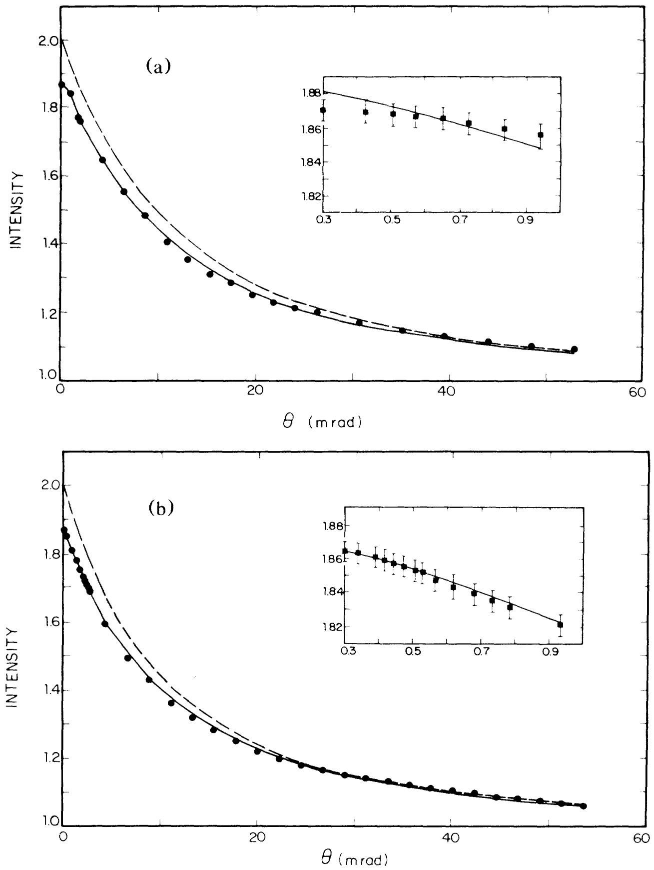

FIG. 1. (a) Line shape for 514.5-nm wavelength. The experimental data are indicated by filled circles, the solid line is from Eq. (D3) of Ref. 1 with $l/\lambda=12$ and $t=323\lambda_1$, and the dashed line is from Eq. (5) of Ref. 8 with $l/\lambda=5$. The inset shows the line shape at its peak for the smallest measured angles ($\theta>0.3$ mrad). The solid line in the inset is the vector theory calculation. (b) Line shape for 457.9 nm wavelength. The parameters used in the vector theory calculation are $l/\lambda=13$ and $t=399\lambda_2$. For the scalar theory calculation $l/\lambda=6$.

source of the discrepancy is the prediction by the scalar theory of a maximum peak intensity of 2, independent of the adjustable parameters of the theory.

The effects of finite sample size are not expected to be important for photon trajectories shorter than the sample thickness. Thus finite size effects are only important for small scattering angles because the coherent scattering for small θ is dominated by extremely long photon trajectories, which are truncated by the finite size of the sample. This truncation results in a rounding of the peak at small angles. Indeed for scattering angles greater than a few mrad, the calculated line shape assuming an infinitely thick sample or a finite thickness (such as was used in the vector calculation of Fig. 1) does not affect the line shape. At the peak of the coherent backscattering, however, finite size effects become important as shown in the insets in Figs. 1(a) and 1(b). The expanded scale of the insets shows the peak of the measured coherent backscattering for the smallest angles measured in our experiments (between 0.3 and 1 mrad), as well as the vector theory fit. The sample thickness resulting in the best fit for the two data sets is $t=324\lambda_1$ and $t=399\lambda_2$ which is self-consistent within the accuracy of the fitting and is in reasonable agreement with the measured value of the thickness of our sample (~0.15 mm). We note that the inclusion of corrections due to finite size effects does not actually require a vector theory, but can also be accommodated by a scalar theory,[10] with the important difference that for the scalar theory the rounded peak maximum still remains at 2.

The vector theory also predicts backscattering of perpendicularly polarized light, with a normalized intensity of ~0.5, and a superimposed, broad, coherently backscattered peak with a peak height of ~0.05. Our data[7] indicate that although this prediction is qualitatively correct, i.e., the perpendicular background component is less than 1 (about 0.8) and there is a weak and very broad coherent peak, the theory does not give an acceptable quantitative fit to the experimental data. Attempts to resolve this discrepancy by incorporating the anisotropic polarizability[1] of $BaSO_4$ into the theory have not been successful, since the anisotropy introduces only very small corrections to the line shape.

In Fig. 2 we show on a log-log plot the crossover from a $1/\theta$ to a $1/\theta^2$ dependence of the experimental line shapes for the two wavelengths. This behavior is due to a crossover in the effective dimensionality of the scattering process, and may be understood in the following way: As discussed by Akkermanns *et al.*,[8] the scattered intensity is essentially the Fourier transform of the probability distribution of the spatial separation of loop end points. The two end points of a loop mark, respectively, the beginning of a scattering sequence and the point at which the photon exits the sample without undergoing any further scattering. Accordingly, these end points will almost always lie within a few optical mean-free paths of the sample sur-

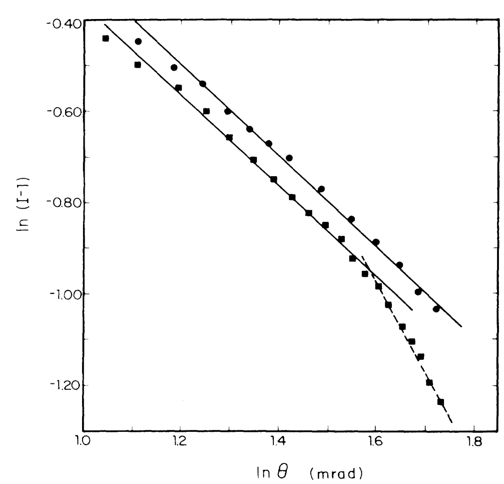

FIG. 2. The log of the backscattered intensity exceeding background $[\ln(I-1)]$ as a function of the log of backscattered angle. Filled circles represent measurements at 514.5 nm and the squares are for 457.9 nm. Solid lines have a slope of $1/\theta$, while the dashed line has a slope of $1/\theta^2$.

face. When the end-point separation is much larger than the optical mean-free path, the distribution is well approximated by a two-dimensional one along the entrance plane of the scattering volume, and its Fourier transform has a $1/\theta$ dependence. On the other hand, when the end-point separation is less than or of the order of the optical mean-free path, the distribution is fully three dimensional, and its Fourier transform falls like $1/\theta^2$. Since large end-point separations correspond to small values of $\sin\theta/\lambda$, while small separations correspond to large $\sin\theta/\lambda$, we expect that for a given wavelength the intensity will fall as $1/\theta$ for small scattering angles, and crossover to a $1/\theta^2$ behavior for sufficiently large scattering angles. As shown in Fig. 2 this is precisely what is observed for the shorter wavelength λ_2, while for the 20% longer wavelength λ_1, the crossover is expected to move out to an approximately 20% larger scattering angle, which is just beyond the range of the measurements. This crossover behavior is predicted by both the scalar and vector theories of the line shape.

In conclusion, we have shown that the experimentally observed line shape of coherent backscattering from a disordered solid medium is well predicted by a vector theory of the multiple scattering process for signal polarization parallel to the laser polarization. For perpendicular polarization, the theory predicts the right qualitative behavior but is not yet in satisfactory agreement with experiment. The line shape is shown to cross over from a $1/\theta$ to a $1/\theta^2$ dependence as expected from the change in dimensionality of the scattering process. Finally, the rounding of the line shape at its peak is understood in terms of finite sample size effects, which can be incorporated into both the scalar and vector theory of the line shape, and gives good agreement with experiment.

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[10]The detailed calculation will be published elsewhere; I. Edrei and M. Kaveh (unpublished).