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Published in:
Physical Review. B, Condensed Matter

DOI:
10.1103/PhysRevB.36.2353

PHYSICAL REVIEW B VOLUME 36, NUMBER 4 1 AUGUST 1987

Vector character of light in weak localization: Spatial anisotropy in coherent backscattering from a random medium

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(Received 30 April 1987)

We report on the first computer simulation of the effects of weak localization in the backscattering of light from a medium containing randomly distributed Rayleigh scatterers. In agreement with our earlier experimental results, the cone of enhanced backscattering in the polarized light component is found to be spatially anisotropic, and this is a consequence of the transverse character of electromagnetic waves.

Recently weak localization of light was detected,[1,2] manifesting itself in the form of a cone of enhanced intensity around the direction of pure backscattering in the light scattered from a random medium. From a more detailed study of the backscatter cone, two very striking effects were observed: (i) The enhancement is much more pronounced in the polarized than in the depolarized light component,[1-5] and (ii) if very small particles are used as scatterers, the "cone" of enhanced intensity shows spatial anisotropy,[6] its width being larger in the direction of the polarization vector of the incident beam than in the perpendicular direction. Both these effects are clearly related to the vector character of light and a complete description of the weak localization of light should therefore take into account this vector character. A lot of theoretical work has already been done to explain the shape and the width of the backscatter cone, and most of this work concerns the *isotropic* scattering of *scalar* waves: An exact theory[7,8] and diffusion approximations[8,9] have been developed. The shape and width of the backscatter cone predicted by these scalar theories agree quite accurately with those found experimentally in the *polarized* light component, but scalar theory cannot be used to describe the depolarized component. In other work, the vector character is considered: Akkermans[10] calculated the enhancement factor for the depolarized light component per order of scattering for a medium containing Rayleigh scatterers and found a total enhancement factor by summing the weighted contributions of the orders. [In the weight factors, the (partial) retention of polarization at low orders of scattering was not taken into account and the resulting value of $\simeq 1.5$ is therefore too high.] Stephen and Cwillich[11] developed a diffusion theory that takes polarization into account and this theory predicts an enhancement factor of $\simeq 1.12$ for the depolarized light component in Rayleigh scattering. By an extension of this theory, Cwillich and Stephen[12] predict spatial anisotropy in the backscatter cone as a result of oblique incidence. We believe that the anisotropy effect that we observed is too large to be explained by oblique incidence only. Furthermore, we demonstrated experimentally that the anisotropy results from lower-order scattering contributions.[6] Therefore a diffusion theory (which is inherently valid for high-order processes) cannot be expected to predict the effect. We think that the qualitative explanation that we gave in Ref. 6 which is based upon the symmetry of the Rayleigh phase function and independent of the angle of incidence, is correct. In this paper, we will show that Akkermans' method to calculate the enhancement factor in the depolarized light component leads to a correct result if the partial survival of polarization at low scattering orders is taken into account. Furthermore, we will present results of the first computer simulation of weak localization in multiple Rayleigh scattering, which demonstrate that the spatial anisotropy in the backscattering cone of the polarized component is a consequence of the vector character of light in combination with the symmetry of the Rayleigh phase function.

Let light with polarization vector $\mathbf{P}_i$ be incident on a random medium in the z direction, and let $s_1, s_2, \ldots, s_{n-1}, s_n$ be a "light path" formed by n scattering centers, along which a wave travels. A scattering event will normally change the polarization vector of the traveling wave. We observe the wave that is scattered from s_n, using a detector that is located in the $-z$ direction and denote the polarization vector of this wave by $\mathbf{P}_f^+$. Since $\mathbf{P}_i$ and $\mathbf{P}_f^+$ are both parallel to the $z=0$ plane, they are related by $\mathbf{P}_f^+ = \underline{M}^+ \mathbf{P}_i$, $\underline{M}^+$ being a 2×2 matrix. For the wave that travels along the same path but in the opposite direction (the time-reversed path) $\mathbf{P}_f^- = \underline{M}^- \mathbf{P}_i$, and $\underline{M}^+$ and $\underline{M}^-$ are related by $\underline{M}^- = (\underline{M}^+)^T$. For directions slightly different from $-z$, the relationship $\underline{M} = (\underline{M}^+)^T$ will still hold approximately. The phases of a time-reversed pair of waves are correlated and the waves will interfere: For the polarized component the interference will be constructive in the $-z$ direction, and in directions different from $-z$ the phase shift between the waves traveling in opposite directions along an individual path will depend on the angle θ between $-z$ and the direction of observation and on the relative coordinates of s_1 and s_n: if s_1 is situated in the origin and x_n and y_n are the x and y coordinates of s_n, then the phase shifts that result from a displacement of the detector over an angle θ in the x-z plane (parallel to $\mathbf{P}_i$) and in the y-z plane (perpendicular to $\mathbf{P}_i$) will be $\simeq x_n \sin\theta/\lambda$ and $\simeq y_n \sin\theta/\lambda$, respectively. If θ is sufficiently large, the phase shift for individual light paths will be random and the detected polarized and depolarized intensities will be $2\langle M_{11}^2\rangle$ and $\langle M_{12}^2\rangle+\langle M_{21}^2\rangle = 2\langle M_{12}^2\rangle$, respectively. (The triangular brackets represent summing over all light paths.) The intensities observed in the $-z$ direction will be $\langle (2M_{11})^2\rangle$ and $\langle (M_{12}+M_{21})^2\rangle$. For the enhancement factors in the polarized and depolarized components $E_\parallel$ and $E_\perp$ and for the polarized and depolarized fractions of the total backscattering $F_\parallel$ and $F_\perp$ it follows that

$$E_\parallel=\frac{\langle (2M_{11})^2\rangle}{2\langle M_{11}^2\rangle}=2,\quad E_\perp=1+\frac{\langle M_{12}M_{21}\rangle}{\langle M_{12}^2\rangle},$$
$$F_\parallel=\frac{\langle M_{11}^2\rangle}{\langle M_{11}^2\rangle+\langle M_{12}^2\rangle},\quad F_\perp=\frac{\langle M_{12}^2\rangle}{\langle M_{11}^2\rangle+\langle M_{12}^2\rangle}. \tag{1}$$

It is easily seen that for second-order backscattering $M_{12}=M_{21}$, and this already suffices to explain that $E_\perp>1$. Likewise, if in second-order backscattering the angle between the incident polarization vector and the normal to the scattering plane is ϕ, the backscattered polarized amplitude will have maxima for $\phi=0,\pi$, and minima for $\phi=\pi/2, 3\pi/2$, so that for the transport of the polarized component it will hold that the average lightpath has $y_n>x_n$. Consequently, a displacement of the detector over an angle θ in the y-z plane will affect the average phase shift to a larger extent than would an equal displacement in the x-z plane or, the second-order contribution to the polarized cone is broader in the x than in the y direction.

Having shown that the second-order contribution to the backscattering has the properties of making the depolarized enhancement factor $E_\perp$ larger than 1 and making the polarized cone spatially anisotropic, we will evaluate the sum of the contributions of all orders: If C_n is the fraction of the total backscattered intensity that is due to nth order scattering processes, the total depolarized enhancement factor will be

$$E_\perp^{\text{tot}}=\sum_{n=1}^{\infty} C_nF_\perp^{(n)}E_\perp^{(n)}\Big/\sum_{n=1}^{\infty} C_nF_\perp^{(n)} . \tag{2}$$

We will follow Akkermans'[10] method to find $E_\perp^{(n)}$ (since intermediate scattering directions will enter into the considerations, vectors and matrices will be in three dimensions from now on): After the first scattering event the polarization vector of the traveling wave is $\mathbf{P}_1=\underline{m}_1\mathbf{P}_1$, where

$$\underline{m}_1=\begin{vmatrix} 1-k_{1x}^2 & -k_{1x}k_{1y} & -k_{1x}k_{1z}\\ -k_{1x}k_{1y} & 1-k_{1y}^2 & -k_{1y}k_{1z}\\ -k_{1x}k_{1z} & -k_{1y}k_{1z} & 1-k_{1z}^2\end{vmatrix} . \tag{3}$$

After the nth scattering event the polarization vector of the wave traveling along a light path in one direction is $\mathbf{P}_f^+=\underline{M}^{(n)}\mathbf{P}_i$, where $\underline{m}^{(n)}=\prod_{i=n}^{1}\underline{m}_i$. Now $\underline{M}^{(n)}=\underline{m}_n\underline{M}^{(n-1)}$ and the nine linear equations contained in the latter matrix equation may be used to find the enhancement factor and the amount of depolarization per order of scattering. Consider the equations:

$$\begin{aligned} M_{11}^{(n)}&=(1-k_{nx}^2)M_{11}^{(n-1)}-k_{nx}k_{ny}M_{21}^{(n-1)}-k_{nx}k_{nz}M_{31}^{(n-1)} ,\\ M_{12}^{(n)}&=(1-k_{nx}^2)M_{12}^{(n-1)}-k_{nx}k_{ny}M_{22}^{(n-1)}-k_{nx}k_{nz}M_{32}^{(n-1)} ,\\ M_{21}^{(n)}&=-k_{nx}k_{ny}M_{11}^{(n-1)}+(1-k_{yn}^2)M_{21}^{(n-1)}-k_{ny}k_{nz}M_{31}^{(n-1)} ,\\ M_{22}^{(n)}&=-k_{nx}k_{ny}M_{12}^{(n-1)}+(1-k_{ny}^2)M_{22}^{(n-1)}-k_{ny}k_{nz}M_{32}^{(n-1)} .\end{aligned} \tag{4}$$

Multiplying the second equation by the third and averaging over all angles of scattering (the odd terms vanish) we get

$$\langle M_{12}^{(n)}M_{21}^{(n)}\rangle=\xi^{(n)}=\langle (1-k_{nx}^2)(1-k_{ny}^2)\rangle\xi^{(n-1)}+\langle k_{nx}^2k_{ny}^2\rangle\eta^{(n-1)} ,$$

where $\eta^{(n-1)}\equiv\langle M_{11}^{(n-1)}M_{22}^{(n-1)}\rangle$ and triangular brackets represent the averaging. Likewise, we get from the first and last one,

$$\langle M_{11}^{(n)}M_{22}^{(n)}\rangle=\eta^{(n)}=\langle (1-k_{nx}^2)(1-k_{ny}^2)\rangle\eta^{(n-1)}+\langle k_{nx}^2k_{ny}^2\rangle\xi^{(n-1)} .$$

From $k_{nx}=\sin\theta\cos\phi$, $k_{ny}=\sin\theta\sin\phi$, and $k_{nz}=\cos\theta$, we have $\langle (1-k_{nx}^2)(1-k_{ny}^2)\rangle=\frac{6}{15}$ and $\langle k_{nx}^2k_{ny}^2\rangle=\frac{1}{15}$. Solving the set

$$\begin{aligned}\xi^{(n)}&=\tfrac{6}{15}\,\xi^{(n-1)}+\tfrac{1}{15}\,\eta^{(n-1)} ,\\ \eta^{(n)}&=\tfrac{1}{15}\,\xi^{(n-1)}+\tfrac{6}{15}\,\eta^{(n-1)} ,\end{aligned} \tag{5}$$

we get

$$\xi^{(n-1)}=[(\tfrac{7}{15})^{(n-1)}-(\tfrac{5}{15})^{(n-1)}]/2 .$$

Now, since we observe from the $-z$ direction, it holds

$$\underline{m}_n=\begin{vmatrix}1&0&0\\0&1&0\\0&0&0\end{vmatrix} ,$$

so that for nth order backscattering

$$\langle M_{12}^{(n)}M_{21}^{(n)}\rangle=\xi^{(n-1)} .$$

Similarly, we can find

$$\langle (M_{12}^{(n)})^2\rangle=[(\tfrac{10}{15})^{(n-1)}-(\tfrac{7}{15})^{(n-1)}]/3 ,$$

and

$$\langle (M_{11}^{(n)})^2\rangle=[(\tfrac{10}{15})^{(n-1)}+2(\tfrac{7}{15})^{(n-1)}]/3 .$$

For the perpendicular enhancement factor in the nth order contribution $(1+\langle M_{12}^{(n)}M_{21}^{(n)}\rangle/\langle (M_{12}^{(n)})^2\rangle)$ we thus obtain

$$E_\perp^{(n)}=1+\frac{3}{2}\,\frac{0.7^{(n-1)}-0.5^{(n-1)}}{1-0.7^{(n-1)}} . \tag{6}$$

The perpendicularly polarized fraction of the nth order contribution to the backscattering is

$$F_{\perp}^{(n)}=\frac{\langle (M_{12}^{(n)})^2\rangle}{\langle (M_{11}^{(n)})^2\rangle+\langle (M_{12}^{(n)})^2\rangle}=\frac{1-0.7^{n-1}}{2+0.7^{n-1}} . \quad (7)$$

A computer simulation (see below) showed that the expression $2[(n+3)^{-1/2}-(n+4)^{-1/2}]$ is a good appoximation for the fraction C_n of the total backscattered intensity that is due to nth order scattering. From C_n, $F_{\perp}^{(n)}$ and $E_{\perp}^{(n)}$ we find $E_{\perp}$ to be $\simeq 1.11$, in agreement with the value of $\simeq 1.12$ found by Stephen and Cwillich. Among the particles that we used in our experiments[6] the 0.214-μm polystyrene spheres have a phase function that comes closest to the Rayleigh phase function (the difference is still substantial). Samples containing these particles as scatterers show a perpendicular enhancement factor of 1.12 ± 0.02.

In order to verify our explanation for the spatial anisotropy found in the polarized cone, we performed the following computer simulation: As a sample a finite slab was taken, of which the thickness (in units of the mean free path λ_{mf}) could be varied. The vacuum-sample interface was chosen in the $z=0$ plane, incidence took place in the origin, and the direction of incidence was z. For nth order scattering, random sequences of $n-1$ scattering directions were generated (the nth direction being $-z$). The distance of free travel of a photon before each scattering event was taken $-\ln(r)$, r being a random value between 0 and 1, corresponding to Beer's-law behavior. Record was kept of the coordinates of the photon, and those sequences in which the photon left the slab on either side before the nth scattering event were discarded. From the angles of scattering and from the Rayleigh phase function, 2×2 matrices relating $\mathbf{P}_f$ to $\mathbf{P}_i$ were calculated, and values for M_{11}^2, $M_{12}M_{21}$, and M_{12}^2, attenuated by a factor $\exp(-z_f)$ (z_f being the z coordinate of the last scattering center) were accumulated in two two-dimensional arrays of "channels" according to their x_f- and z_f-, respectively, their y_f- and z_f-coordinates. Thus, each channel comes to hold a contribution to the backscattered intensity for which the angle dependence of the phase shift between the waves traveling in opposite directions is known. The corresponding interference patterns were then calculated and added, yielding two distinct intensity patterns (one for x and one for y polarization) for both scanning directions (the x scan parallel to $\mathbf{P}_i$, and the y scan perpendicular) making a total of four patterns. We note that, by counting the interference between time-reversed pairs of paths but not the interference between different paths, we obtain a result that is equivalent to an ensemble average over different realizations of the random medium, i.e., the type of cone that is found from a liquid medium in which the scatterers are subject to thermal motion or from a solid medium that is spun. The strong intensity fluctuations that may be observed from stationary solid samples[4,5] are a result of interference between different light paths.

The simulation was tested in the following ways.

(i) Using an isotropic phase function, the simulation was found to reproduce the transmission and reflection characteristics calculated for different slabs from exact Milne theory.[13]

(ii) Still using an isotropic phase function, the simulation reproduced the dependence of full width at half maximum (FWHM) on λ/λ_{mf}, the order of scattering, and the slab thickness as calculated from exact isotropic[8] theory.

(iii) Using the Rayleigh phase function in combination with a constant step length, omitting the $\exp(-z)$ attenuation, and permitting z to become negative (i.e., evaluating exclusively the influence of the angles of scattering) the simulation reproduced the order dependence of the perpendicular enhancement factor as found from (6).

Simulated values of the full width at half maximum Γ (FWHM) for x and y scans of the nth order parallel cone for a slab of thickness $20\lambda_{mf}$ are given in Fig. 1. For low orders of scattering, the values are larger for the (parallel) x scan than for the (perpendicular) y scan. For increasing order the value Γ_x/Γ_y converges towards unity, so the

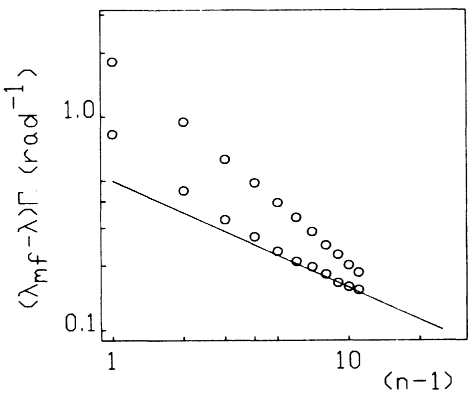

FIG. 1. (λ_{mf}/λ) Γ of the simulated contribution to the backscatter cone as a function of scattering order. Upper curve, scan $\parallel \mathbf{P}_i$; lower curve, scan $\perp \mathbf{P}_i$. Continuous line, (λ_{mf}/λ) $\Gamma=0.5(n-1)^{-1/2}$.

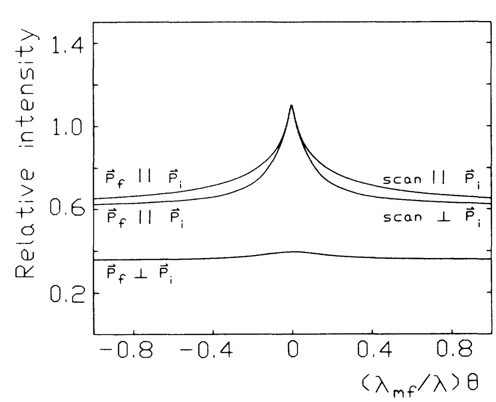

FIG. 2. Anisotropy in the enhanced backscattering of light from a random medium containing Rayleigh scatterers: total cones as found by summing the orders 1–1000 (coresponding to 94% of the total backscattering). The curves are normalized with respect to 100% of the total ($\parallel + \perp$) background intensity. Upper curve: scan $\parallel \mathbf{P}_i$, $\mathbf{P}_f \parallel \mathbf{P}_i$; middle curve: scan $\perp \mathbf{P}_i$, $\mathbf{P}_f \parallel \mathbf{P}_i$; lower curve: $\mathbf{P}_f \perp \mathbf{P}_i$ (curves for scans $\parallel \mathbf{P}_i$ and $\perp \mathbf{P}_i$ coincide).

high-order asymptotic behavior is expected to be the same for both scanning directions. An estimate of this behavior obtained by extrapolation of the FWHM values found in y scans is given in Fig. 1 as a continuous line. The implied empirical relationship between Γ and the order of scattering is $\Gamma = 0.5(\lambda/\lambda_{mf})(n-1)^{-1/2}$.

The fraction that each separate order contributes to the total backscattering was simulated in the following way: Photons were allowed to random walk in a slab. For every collision, the scattering angles θ and ϕ were chosen randomly, the former from a probability distribution imaging the Rayleigh phase function $\Phi(\theta) = \frac{3}{4}[1+\cos^2(\theta)]$, the latter from an interval $0-2\pi$. The step length was $-\ln(r)$, r being (as before) a random number between 0 and 1. Record was kept of the number of collisions n, the z coordinate, and the last direction of scattering. Backscattered photons were accumulated in channels, according to their n value. Tested with an isotropic phase function, the simulation reproduced the results of Milne theory. Using the Rayleigh phase function, we found very nearly the same results as in the isotropic case. The simulation showed that to an approximation sufficient for our purposes, the nth-order fraction of the total backscattering (for thick slabs) may be given by $C_n = 2[(n+3)^{-1/2} - (n+4)^{-1/2}]$ in both isotropic scattering and Rayleigh scattering.

Using the latter result, intensity profiles for the x- and y-polarized components in both x and y scans were calculated in the following way: For the orders 2–12, simulated profiles, weighted by a factor of $C_n F^{(n)}$ ($F^{(n)}$ being $F_{\parallel}^{(n)}$ or $F_{\perp}^{(n)}$) were added. The x-polarized profiles were increased by C_1 (=0.106) to account for the first-order contribution. The x- and y-polarized contributions of order > 12 were assumed to be Lorentzian shaped with a FWHM of $0.5(\lambda/\lambda_{mf})(n-1)^{-1/2}$ and flat, respectively, both with $F^{(n)} = \frac{1}{2}$. The total intensity profiles for the orders 1–1000 (corresponding to 94% of the total backscattering from a semi-infinite slab) are given in Fig. 2. It is seen that for the x-polarized component (parallel to the incident polarization) the x scan yields a wider profile than the y scan and this is a result of the anisotropic character of the Rayleigh phase function. For the y-polarized component the x- and y-scan curves coincide. This also follows from the symmetry of the Rayleigh phase function: The perpendicular component of the polarization vector varies with $\sin\phi\cos\phi$, making the x and y directions equivalent.

This work is part of the research program of the "Stichting voor Fundamenteel Ondezoek der Materie" (FOM), which is financially supported by the "Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek" (ZWO).

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