Polarization asymmetry in waves backscattering from highly absorbant random media

A. Dogariu, M. Dogariu, K. Richardson, S. D. Jacobs, and G. D. Boreman

Within the range in which light penetration depth is approximately the same as or less than the diameter of the particles in the medium, particulate media with considerable absorption behave as two-dimensional, rough-surface structures. As penetration depth increases, a complicated transition between volume and surface effects is seen. For these media, low-order scattering sequences have small spatial extent, making observation of polarization characteristics difficult. We present an experimental technique to access the low-order scattered photons by artificially reinjecting them through total internal reflections. Using a dielectric layer in contact with the high-absorption medium, we are able to observe fourfold polarization asymmetry in backscattering from highly absorbant media. We discuss the origin of the polarization patterns in a ray-optics approximation and suggest possibilities for solving practical problems encountered in characterizing composites with appreciable absorption.

1. Introduction

Experimental[1] and analytical[2] efforts are being directed toward understanding the mechanism of material removal in magnetorheological (MR) finishing (MRF). The MR fluid consists of black spherical particles of carbonyl iron in an aqueous mixture with cerium oxide polishing abrasive particles. The solid concentration is more than 40%. Outside of a magnetic field, the MR fluid exhibits a relatively small viscosity, while in a magnetic field the viscosity can rise to tens of N/m^2. Because of the opacity of the suspension, the interaction of the magnetic field-stiffened MR fluid "lap" with the glass surface during finishing is difficult to study through direct observation. Scanning electron micrographs taken on dilute suspensions show that the carbonyl iron particles form chains that orient along gradients in the magnetic field.[3] Other work suggests that, in forming these chains, the magnetic particles force the nonmagnetic particles and carrier liquid to the interface with the glass part. The structure and composition in this zone of contact where polishing occurs are unknown. Understanding the MRF microstructure and its impact on the macroscopic rheological behavior is the basis for a predictive methodology for controlling the polishing process.

To achieve a better structural description, light scattering is one of the most attractive alternatives. By a remote, noninvasive, and nonperturbative assessment, light scattering is advancing the knowledge of microstructure changes induced by an external magnetic field. Light-scattering techniques also have the potential for developing high-performance instrumentation and sensing procedures for online MRF morphology assessment. From an optical-characterization viewpoint, MRF is a highly packed, random composite medium that, owing to its high iron content, manifests a strong absorption in the visible. The characterization of optical-wave propagation in these composites requires a better understanding of the static and dynamic properties of optical waves in the multiply scattering regime. Results of a considerable effort over the past decade have shown that, despite the inherent randomness of photon paths, structural information can be retrieved by means of multiple light scattering, and scalar-wave models for optical fields have been used successfully for explaining numerous light-scattering phenomena.[4]

However, the vector nature of light plays an important, sometimes dominant, role in many processes.[5–8]

A. Dogariu, K. Richardson, and G. D. Boreman are with the Center for Research and Education in Optics and Lasers (CREOL), University of Central Florida, 4000 Central Florida Boulevard, Orlando, Florida 32816-2700. M. Dogariu is with the Department of Physics, University of Central Florida, 4000 Central Florida Boulevard, Orlando, Florida 32816-2700. S. D. Jacobs is with the Laboratory for Laser Energetics, Center for Optics Manufacturing, University of Rochester, Rochester, New York 14623.

Received 5 February 1997; revised manuscript received 13 June 1997.
0003-6935/97/318159-06$10.00/0

The polarization state of multiply scattered light can be used to extract information about the scattering medium and the underlying propagation mechanisms.[9–14] Uniform random volume scatterers manifest fourfold symmetrical polarization return patterns (PRP) when a narrow-beam geometry is used and emergent light is collected in the cross-polarized channel. These patterns can be understood in terms of a deterministic rotation of the polarization vector and a competing statistical depolarization.[5,10,11] The structure of the patterns can be related to statistical descriptors of the scattering medium. Rotations of the polarization vector result from low-order, cornerlike scattering events and induce deterministic depolarization effects. Illumination in a pencil-beam configuration and detection of waves with zero lateral momentum transfer (waves retroreflected parallel to the direction of incidence) permit visualization of these effects.[10–11]

In a volume-scattering medium, a complex mechanism of multiple scattering occurs, and a distribution of scattering paths and scattering orders is developed. The resulting multiple-scattering mechanism causes statistical depolarization, decreasing the sharpness of observed PRP. A quantitative description of this phenomenon can be made in terms of pattern contrast.[10]

The wave depolarization is a material parameter that measures the weight of high-order scattering sequences compared with that of low-order ones. The degree of statistical depolarization ultimately determines the existence of polarization patterns. For media in which the characteristic scattering lengths are much smaller than the sample dimensions, depolarization is unity, and no polarization patterns can be observed. Accordingly, in attempting a structural characterization, it is necessary to observe low-order scattering sequences.

The contribution of higher-order sequences can be reduced by dilution of the media, finite optical depths, or absorption. These mechanisms act as a scattering-path cutoff filter and shift the balance in favor of the low-order scattering sequences. However, in dense media, the low-order scattering events correspond to small lateral spreading, reducing the spatial extent of the polarization patterns. We present a way to overcome these experimental restrictions by taking advantage of internal reflections on a dielectric interface. The internal reflection enlarges the spatial extent of the polarization patterns, facilitating quantitative data collection.

Because dense suspensions of large highly absorbing particles are of practical industrial interest, our goal is the optical characterization of their properties. Little information is available in the scientific literature about optical properties of suspensions with one or more components that have complex indices of refraction and appreciable solid loads. For opaque composites, investigation of bulk properties of highly packed random media is challenging because refractive or transmission techniques are inadequate and one has access only to reflection measurements. The characterization of dense media with high absorption eludes the classical approaches, which require high-order scattering, based on radiative transport theory in the diffusion approximation. The media in which low-order scattering sequences are dominant require different approaches for explanation of the observed phenomena and for development of analytical, albeit approximate, descriptions.

Characterization of densely packed absorbing composites is a challenging task because of the low-order scattering sequences involved and because of the reduced spatial extension of these scattering paths. To characterize these media, we describe an experimental technique that takes advantage of total internal reflections at the boundary of a random medium. When a dielectric layer is placed in contact with the medium, the order of scattering is artificially increased by adding the internal-reflection scattering events. The lateral spread is also substantially enlarged because of the scattering paths inside the dielectric layer. However, because of their deterministic nature, the additional scattering events do not add to the wave randomization process. These reinjections enforce a shift of the wave-scattering path length distribution toward larger values, permitting access to polarization properties of low-order scattered waves. Photons that, at the injection point, are scattered at large angles are reinjected into the medium and accumulate a lateral shift that is determined by the dielectric layer thickness and refractive index. After several interactions with the medium, reflected waves are collected at large distances from the injection point, where the spatial resolution is not an issue anymore. The high absorption ensures that the recorded scattering orders are low enough to enable a polarization analysis.

We show that the characteristic four fold polarization patterns obtained in retroreflectance from densely packed random suspensions with high absorption can be used to extract information about the medium and to extend our knowledge about the vector nature of wave propagation.

For highly absorbant media, the major scattering characteristics are determined by the boundary layer modeled as an irregular surface. We show that the cornerlike scattering events and the symmetry and isotropy of the scattering medium are responsible for the appearance of the polarization patterns and that a ray-optics analysis correctly describes the main polarization features.

2. Experiment

We investigated high-absorption random media in an experimental geometry described in Fig. 1. A highly absorbant suspension is contained in a cell with a 1-mm-thick window. A narrow beam, focused down to 0.3 mm, of linearly polarized light (wavelength $\lambda = 633$ nm) is normally incident on the interface b of the cuvette window as seen in Fig. 1. The light emergent from the same surface is imaged with a narrow field of view upon a CCD camera, and the images are acquired by a frame grabber (512×512). Typically 50 frames are recorded and averaged. A polarizer is

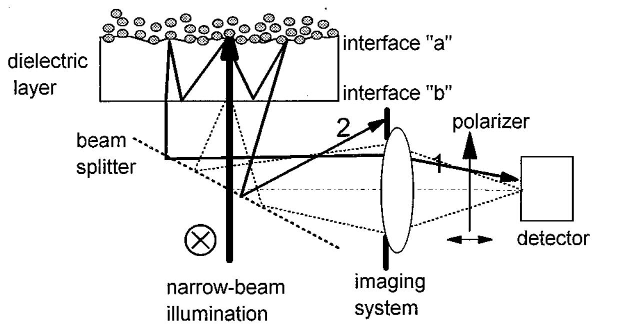

Fig. 1. Experimental setup used to record polarization return pattern from media with high absorption.

inserted in the detection path and provides the selection of the cross-polarized intensity. This monostatic geometry with normal incidence is particularly attractive for practical applications.

A sequence of patterns recorded under these experimental conditions is shown in Fig. 2 as contours of equal intensities. The media were aqueous dispersions of carbonyl iron (4.5-μm median size) and cerium oxide particles (3.5-μm median size) with various solid fractions. Assuming monodispersivity and spherical shapes for the two components, we used Mie scattering theory to evaluate the scattering and absorption coefficients. Subsequently, the photon penetration depths for these mixtures are estimated to be of the order of 7, 12, 20, and 40 μm for PRP in Figs. 2(a), 2(b), 2(c), and 2(d), respectively. As can be seen, when absorption is increased gradually, the fourfold polarization structure becomes visible, resembling the polarization patterns we

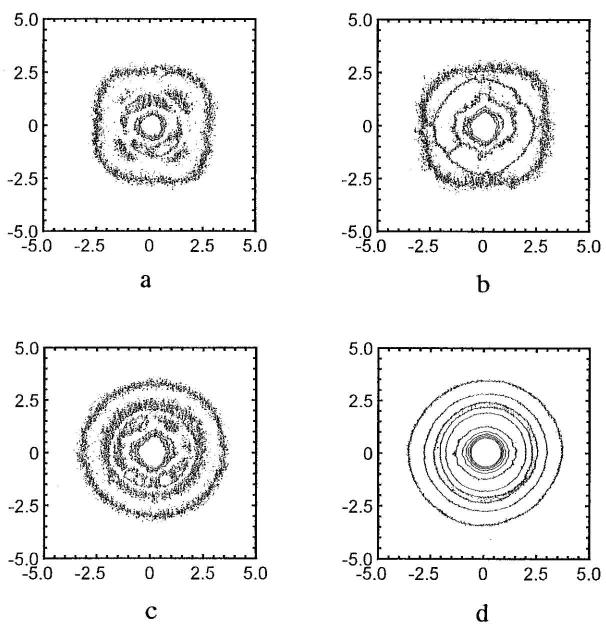

Fig. 2. Contours of equal intensity in backscattering from highly absorbant media. The penetration depths in panels a, b, c, and d are 7, 12, 20, and 40 μm, respectively. The scales are in millimeters.

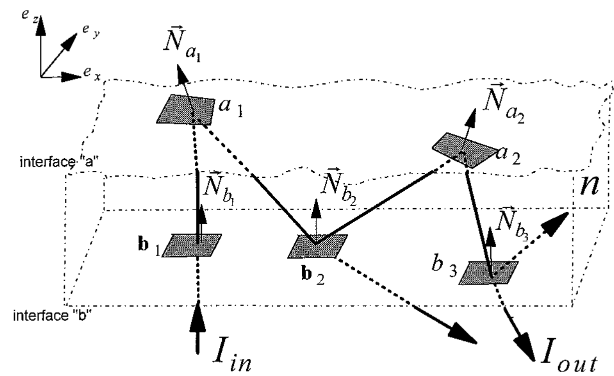

Fig. 3. Schematic of an in-plane, multiple-scattering sequence developed inside a dielectric layer in contact with a strongly absorbant medium.

obtained by scattering from dielectric layers with rough surfaces.[8] This corresponds to the geometric-optics limit of closely packed particles of a dimension much larger than the wavelength.

There are three distinct regions in the patterns. First, there is a bright central spot of depolarized light caused by the direct retroreflection of the incident finite-sized beam and imperfect polarizers. Second, the presence of a dark surrounding ring can be explained if the rays shown in Fig. 3 are followed. Because the rays exiting at points of the type b_2 are eliminated because of the small collection angle, only rays emerging at points of type b_3, which suffered cornerlike scattering sequences of two reflections on face a and one reflection on face b, are contributing to the recorded pattern. As long as the incidence angle on face b is smaller than the limit angle, most of the light is transmitted through face b, at large angles that make it ineligible for collection, and very little is reflected toward points of type a_2. At the limit angle, a jump in brightness is visible because all the light incident at b_2 is now reflected toward points a_2.

The main feature of the pattern is the clear four-lobe structure; the contrast decreases rapidly with the radial distance. We show that the observed four-lobe pattern can be explained by a geometric-optics approach, which is applicable to a wide class of practical situations. We also show that the statistics of the boundary influence the appearance and structure of the pattern. The origin of the patterns is prototypical three-step, cornerlike scattering sequences. As shown in Fig. 4, coplanar scattering sequences in the plane containing the incident polarization vector ($\varphi = 0$) and in the plane perpendicular to it ($\varphi = 90°$) either leave the polarization vector unchanged or flip it while the scattering sequences contained in planes at azimuthal angle φ lead to a rotation $\pi - 2\varphi$ of the polarization vector. These cornerlike scattering sequences were found to be the origin of similar polarization patterns observed for weak volume scattering media in a narrow-beam illumination geometry.[5,7] The randomization of the electric vector that occurs in multiple scattering was shown to be the competing effect that diminishes the

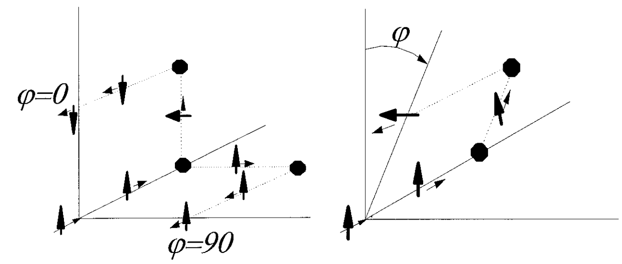

Fig. 4. Geometrical depolarization determined by two cornerlike scattering events.

contrast of the patterns. The contrast of the patterns was used to extract the scattering path length of waves emerging at a radial distance R from the injection point.

3. Ray-Optics Approximation

For media with small penetration depth, a ray-optics model can be developed to describe the vector behavior of the scattered light. When the characteristic length scales are appreciably larger than the wavelength, most of the energy is scattered in the specular direction, and the geometric-optics approximation is applicable. In the ray representation, the field emitted at one point after a single-scattering event is completely determined by the incident field and the local properties (geometry and complex refractive index) of the boundary. This restriction allows for a simple tracking of the light rays, without essentially limiting the validity of the approach. We monitor a simple, three-step scattering event such as the one shown in Fig. 3 and describe the outgoing electric vector orientation.

Because the collection angle of the imaging system is limited, single scattered rays like the type-2 ray in Fig. 1 are not collected by the detector, and only rays emerging close to the backscattering direction are monitored. The incident and the emergent directions are parallel to each other and also perpendicular to surface b, as can be seen in Fig. 3. These conditions define the cornerlike sequence of, at minimum, three coplanar reflection events.

Consider that the narrow incident beam has the electric vector oriented along the axis defined by e_y and that the analyzer has the direction along the axis defined by e_x. In the geometric-optics limit, single-scattering events can be described in a manner similar to scattering by a surface with the local slope specified by a unit random vector $\mathbf{N}(\theta, \varphi)$. The outgoing field at point b_3 will be defined by the deterministic reflection events governed by Snell's law and by the random process described by the variable $\mathbf{N}(\theta, \varphi)$.

In the axial symmetry conditions of the problem, it can be shown that the field that emerges after a cornerlike sequence at a point located on the face b at the azimuth φ will have the orientation

$$\mathbf{E}_{\text{out}} \sim e_x |\mathbf{E}_{\text{out}}| \sin 2\varphi + e_y |\mathbf{E}_{\text{out}}| \cos 2\varphi, \qquad (1)$$

and, because only light in the cross-polarized channel is recorded,

$$I_{\perp\text{out}}(\varphi) \sim \frac{|\mathbf{E}_{\text{out}}|^2}{2} (1 - \cos 4\varphi). \qquad (2)$$

As long as the quasi-surface descriptors are homogeneous and stationary random processes, the fourfold symmetry of the field is preserved. We note that in the cylindrical symmetry of the problem, any directional properties of the boundary break the fourfold symmetry of the patterns.

Obviously the trajectories of the rays that emerge at face b in Fig. 3 are more complicated than the simplest in-plane sequences described here. Multiple reflections also occur, and the field at any point on surface b depends also on nonlocal properties such as the nature and geometry of other points on the surface.

The scattering planes can be skewed and the reflection sequences can be nonplanar, inducing additional azimuth rotations of the electric vector; relative phase differences introduced by total internal-reflection events add a certain degree of ellipticity to the emerging light; differences between s- and p-reflection coefficients lead to rotations of the vibration plane with respect to the scattering plane. Moreover, the finite size of the incident beam creates alternative reflection channels that may contribute at the emerging point, distorting the structure of the lobes. However, one common feature of these effects is that, because they all depend on the random variables associated with the boundary, their overall effect will be a statistical random depolarization of the multiply scattered light. This scrambling of the polarization vector is responsible for decreasing the contrast of the polarization patterns.

In the geometric-optics limit, the above-mentioned processes lead eventually to random rotations of the polarization vector with respect to the scattering plane. This suggests that, because we are interested only in the polarization features of the backscattered light and not in the absolute radial dependence of intensity, we can include the depolarizing effects in a generic rotation γ of the electric vector. Therefore if, after one ideal cornerlike sequence, the cross-polarized component was $E_{\perp\text{out}} = \mathbf{E}_{\text{out}} \sin 2\varphi$, we can now rewrite it as

$$E_{\perp\text{out}} = \mathbf{E}_{\text{out}} \sin(2\varphi + \gamma). \qquad (3)$$

Although φ is determined by the geometry and the symmetry of the problem, γ should be regarded as the fluctuating part of the rotation angle, with a probability distribution function determined by the statistical characteristics of our quasi-surface.

In what follows we show that the fourfold symmetry is fully preserved as long as the fluctuation γ is a zero-mean random process, which is a natural assumption for isotropic systems. The cross-polarized intensity averaged over the random variable γ will be

$$\langle I_{\perp\text{out}} \rangle = (\langle |\mathbf{E}_{\text{out}}|^2 \rangle / 2)[1 + (1 - 2\langle \cos^2 \gamma \rangle) \times \cos 4\varphi + \langle \sin 2\gamma \rangle \sin 4\varphi]. \qquad (4)$$

An important observation is that γ is independent of the azimuth φ of the cornerlike scattering plane. Moreover, when the fluctuation γ is of zero mean, then $\langle \sin 2\gamma \rangle = 0$, and the resulting intensity is

$$\langle I_{\perp \mathrm{out}} \rangle = (\langle |\mathbf{E}_{\mathrm{out}}|^2 \rangle / 2)[1 + (1 - 2\langle \cos^2 \gamma \rangle)\cos 4\varphi]. \quad (5)$$

The cross-polarized intensity consists of an azimuth-independent term modulated by a factor with periodicity $\pi/2$. The azimuth-dependent factor is reduced when $\langle \cos^2 \gamma \rangle$ becomes large and, consequently, the fourfold structure disappears. If Eq. (2) predicted a pattern of unity contrast, Eq. (5) shows that the fourfold symmetry is preserved for random multiple-scattering processes, but the contrast of the patterns is diminished. As from Eq. (5), whenever the medium properties are not isotropic, the fluctuating γ ceases to have a zero mean and $\langle \sin 2\gamma \rangle \neq 0$, which leads to the distortion of the fourfold symmetry. By investigating the azimuth periodicity of the patterns, we can infer the directional features.

We experimentally observe that the contrast of the patterns decreases radially. At longer distances from the injection point, the numerous multiple reflections lead to a complete scrambling of the polarization vector, and the statistical depolarization becomes predominant. This effect is accounted for if the width of the probability distribution function for γ, centered at $\gamma = 0$, increases with the number of reflection events, i.e., with the radial distance on the surface. With the occurrence of many scattering events, at large radial distances γ becomes uniformly distributed between the limits $-\pi$ and π, when $\langle \cos^2 \gamma \rangle = 1/2$, and the polarization pattern vanishes. The value of the azimuth-independent term $\langle |\mathbf{E}_{\mathrm{out}}|^2 \rangle$ is determined by the reflection properties of the quasi-surface. Therefore both the contrast of the pattern and the azimuth-independent intensity decrease with the number of reflection events.

For the present case of a quasi-surface reflection, statistical depolarization occurs also for low-order reflections. When the penetration depth is considerable, the possibility exists for waves to spread laterally over distances comparable with the penetration depth. This is a complicated scattering mechanism governed by the delicate balance between surface- and volume-scattering processes. The lateral diffusion along interface a, around points such as a_1 and a_2 in Fig. 3, determines a tilt of the scattering planes that induces the fluctuation γ of the polarization vector at exit points such as point b_3 in Fig. 3.

A simplified description can be developed that considers the medium with a finite penetration depth as a facetlike rough surface, and therefore lateral wave spreading can be understood as a multiple-scattering process inside the valleys of the surface. Accordingly the direction of the polarization vector is randomized in a manner that is, in a first approximation, proportional with the penetration depth.

This is exemplified in Fig. 5, where we present contours of equal intensities as derived from Eq. (5) for fluctuations γ, which have a Gaussian distribution centered around $\gamma = 0$ and a rms value proportional to the penetration depth of the samples for which the patterns shown in Fig. 2 were recorded. As can be seen, the main features of the polarization patterns are correctly described by Eq. 5 on a geometric-optics basis. When the penetration depth increases the transition from surface scattering to volume scattering sets in, the actual number of scattering events increases together with the polarization fluctuations, and accordingly the polarization asymmetry characteristic vanishes.

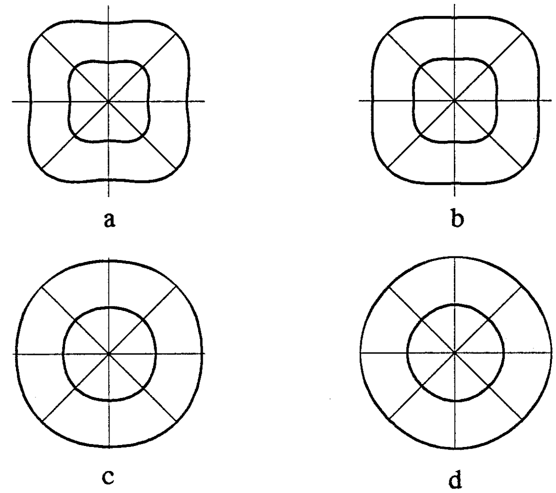

Fig. 5. Contours of equal intensity as derived from Eq. (5) for $\langle \cos \gamma \rangle$ proportional to the penetration depths in Fig. 4.

4. Conclusions

Polarization cross patterns resulting from low-order scattering sequences are visible in dense, multiple-scattering, and strongly absorbant media. In these media, the scattering characteristics are determined by the boundary layer that can be modeled as a rough surface. Internal reflections enlarge the lateral wave spreading, making possible the observation of polarization-dependent retroreflection patterns. In a narrow-beam illumination, fourfold polarization patterns are obtained and explained by a ray-optics model.

Because the symmetry and isotropy of the scattering medium play a major role in the appearance and the structure of the patterns, study of the cross patterns can reveal the local anisotropy or directionality of the medium, which is particularly useful for the investigation of structural reversible media such as electro- or MR fluids.

The contrast of the patterns is determined by the scattering characteristics of the medium, and it can be used as a quantitative descriptor for structural information such as the solid content or particle size.

We consider this a new and important result, which offers the possibility to investigate a wide class of scattering media with a simple technique based on relative intensity measurements.

This research was funded in part by the Center for Optics Manufacturing, University of Rochester, and U.S. Army Material Command under contract DAAA21-94-C-0003.

A. Dogariu acknowledges collaboration with NSF Engineering Research Center for Particle Science and Technology, University of Florida.

References

1. S. D. Jacobs, D. Golini, Y. Hsu, B. E. Pucherbner, D. Strafford, W. I. Kordonski, I. V. Prohorov, E. Fess, D. Pietrowski, and V. W. Kordonski, "Magnetorheological finishing a deterministic process for optics manufacturing," in *International Conference on Optical Fabrication and Testing*, T. Kasai, ed., Proc. SPIE **2576,** 372–382 (1995).
2. W. I. Kordonski and S. D. Jacobs, "Magnetorheological finishing," Int. J. Mod. Phys. B **10,** 2937–2948 (1996).
3. I. V. Prohorov, W. I. Kordonski, L. K. Gleb, G. R. Gorodkin, and M. L. Levin, "New high precision magnetorheological instrument-based method of polishing optics," in *Optical Fabrication and Testing*, Vol. 24 of 1992 OSA Technical Digest Series (Optical Society of America, Washington, D.C., 1992), pp. 135–136.
4. A. Ishimaru, *Wave Propagation and Scattering in Random Media* (Academic, New York, 1978).
5. D. Elyiahu, M. Rosenbluh, and I. Freund, "Angular intensity and polarization dependence of diffuse transmission through random media," J. Opt. Soc. Am. A **10,** 447–491 (1993).
6. R. Berkovitz and M. Kaveh, "The vector memory effect for waves," Europhys. Lett. **13,** 97–101 (1992).
7. M. P. Albada, M. B. Van der Mark, and A. Lagendjik, "Vector character of light in weak localization: spatial anisotropy in coherent backscattering from a random medium," Phys. Rev. B **36,** 2353–2358 (1987).
8. M. Dogariu and T. Asakura, "Photon pathlength distribution from polarized backscattering in random media," Opt. Eng. **35,** 2234–2239 (1996).
9. C. J. Solomon and J. C. Dainty, "Use of polarization in double-passage imaging through a random screen," Opt. Commun. **87,** 207–211 (1992).
10. M. Dogariu and T. Asakura, "Polarization dependent backscattering patterns from weakly scattering media," J. Opt. (Paris) **24,** 271–278 (1993).
11. M. Dogariu and T. Asakura, "Polarized light scattering in dielectric layers with rough interfaces," Opt. Commun. **131,** 1–7 (1996).
12. A. Carswell and S. R. Pal, "Polarization anisotropy in lidar multiple scattering from clouds," Appl. Opt. **24,** 3464–3471 (1980).
13. D. Bicout, E. Akkermans, and R. Maynard, "Dynamical correlations from multiple light scattering in laminar flow," J. Phys. I (Paris) **1,** 471–491 (1991).
14. D. Bicout, C. Brosseau, A. S. Martinez, and J. M. Schmitt, "Depolarization of multiply scattered waves by spherical diffusers: influence of size parameters," Phys. Rev. E **49,** 1767–1770 (1994).