Depolarization of multiply scattered waves by spherical diffusers: Influence of the size parameter

D. Bicout
Institut de Biologie Structurale, 41 Avenue des Martyrs, 38027 Grenoble Cedex 1, France

C. Brosseau
Laboratoire de Spectrométrie Physique, Université Joseph Fourier, Boîte Postale 87, 38042 Saint-Martin-d'Hères Cedex, France

A. S. Martinez
Laboratoire d'Expérimentation Numérique, Maison des Magistères, Centre National de la Recherche Scientifique, Boîte Postale 166, 38042 Grenoble Cedex 9, France

J. M. Schmitt
National Center for Research Resources, National Institutes of Health, Bethesda, Maryland 20892
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We study numerically, using the Mie theory, light transmission through a multiply scattering medium composed of a collection of uncorrelated, optically inactive spherical particles. The characteristic length over which a plane-wave field is depolarized depends on whether it is initially linearly or circularly polarized and on the size of the particles. In a medium containing particles small compared to the wavelength (Rayleigh regime), the characteristic length of depolarization for incident linearly polarized light is found to exceed that for incident circularly polarized light, while the opposite is true in a medium composed of particles large compared to the wavelength (Mie regime). A comparison of numerical results with the data from measurements on suspensions of polystyrene latex spheres in water is made. Agreement between these simulations and experiment is good for the range of sizes considered in this paper. We also discuss the relevance of the helicity-flip model to the analysis of these data.

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Recently, considerable interest has arisen in the polarization properties of waves scattered by inhomogeneous media, in connection with condensed-matter physics (i.e., weak localization) [1]. Multiple scattering of light from inhomogeneities in optically dense media randomizes the state of polarization and a wave propagating in such a medium becomes rapidly depolarized, but one knows little about the characteristic length scale on which this process proceeds. We have previously reported on the depolarization of optical waves propagating through a random multiple-scattering medium composed of a collection of nonabsorbing and optically inactive spherical particles whose size is very small compared to the wavelength of the scattered radiation (i.e., Rayleigh regime) [2]. For completeness, the main result of Ref. [2] will be rederived in a somewhat different form. For purposes of our model, we consider only the weak scattering limit (i.e., $kl \gg 1$) such that the elastic mean free path $l \equiv 1/\phi\sigma$ is much larger than the wavelength of the radiation. Consequently, the wave-field propagation may be described by a classical diffusion process. Here ϕ is the concentration of scatterers, σ is the scattering cross section, and k is the wave number in the scattering medium. Let us consider a quasimonochromatic plane wavefield that is incident normally along the z axis upon a plane-parallel slab, of finite thickness d ($d \gg l$) in the z direction, and of infinite extent in the x,y directions, composed of uncorrelated spherical particles of radius a. We begin by computing the degree of polarization of the light transmitted by the scattering medium for incident linearly (P_L) and circularly (P_C) polarized light. To do this, one must evaluate the different contributions of light following many different paths. Take a particular sequence of scattering events. The number of steps in this path of length s is $n \equiv s/l$. The number of scattering paths of length s is simply given by the Green's function $G(n,d)$ of the diffusion equation [2]. It follows from results of Ref. [2] that these degrees of polarization are given by a proper weighting $G(n,d)$ of scattering paths of length s. The resulting expression is

$$P_i = \frac{\int_0^\infty f_i(n) G(n,d)\,dn}{\int_0^\infty G(n,d)\,dn} , \tag{1}$$

where we have adopted the notation $i = L$ for linear and C for circular states of polarization. In the multiple-scattering regime, the functions f express the dependences of the output degrees of polarization for a number of scatterings equal to $n+1$. In the large-n limit, the f's reduce simply to

$$f_L(n) \cong \tfrac{3}{2} \exp[-n(l/\zeta_L)]$$

and $f_c(n) \cong \frac{3}{2} \exp[-n(l/\zeta_C)]$. The ζ's define characteristic lengths of depolarization for a path of $n+1$ scatterings: $\zeta_L = l/(\ln\frac{10}{7})$ and $\zeta_C = l/(\ln 2)$. From the numerical values of the ζ's, we find that $\zeta_L \cong 2\zeta_C$. Performing the integration in (1), it is straightforward to obtain

$$P_i = \frac{d}{l}\frac{\sinh\left[\dfrac{l}{\xi_i}\right]}{\sinh\left[\dfrac{d}{\xi_i}\right]}, \tag{2}$$

where $\xi_i = (\zeta_i l/3)^{1/2}$, with $i = L, C$, define characteristic lengths of depolarization for the slab geometry. Since $d \gg \xi_i$, the degree of polarization of the transmitted light in the far field can be approximated by

$$P_i \cong \frac{2d}{l}\sinh\left[\frac{l}{\xi_i}\right]\exp\left[-\frac{d}{\xi_i}\right]. \tag{3}$$

Thus we see that the characteristic length of depolarization for incident linearly polarized light is greater (by a factor of $\sqrt{2}$) than the corresponding length for incident circularly polarized light. This analysis should apply equally well for large spheres provided that l is changed into the transport mean free path

$$l^* = \frac{1}{\phi\sigma^*} = l/[1-\langle\cos(\theta)\rangle]$$

and that the appropriate size dependence of the f's is inserted therein. Here the transport scattering cross section for each scatterer is defined in the usual way as

$$\sigma^* = \int \sigma(\theta)[1-\cos(\theta)]d\Omega ,$$

where θ is the scattering angle.

The analysis of Ref. [2], derived from a maximum entropy principle, permitted us to characterize the irreversible evolution of the polarization state during scattering. These results were in agreement with the explicit computation done in the context of the Bethe-Salpeter equation handled in the diffusion approximation [3]. We were also able to derive a number of new results. For example, we showed that depolarization of light by multiple scattering is connected to a process of entropy production which falls off exponentially with the number of scattering events [2]. However, a fundamental question persists regarding this problem. It is not yet established how depolarization proceeds as the size of the particles increases from very small to large (i.e., Mie regime). To this end, we have studied numerically, in this Brief Report, the depolarization behavior as a function of the size of the scattering particles. These numerical results are compared to measurements on suspensions of polystyrene latex spheres in water.

We have used the SLAB Monte Carlo simulation code to analyze the depolarization behavior of a wave propagating through a slab of finite thickness and composed of uncorrelated spherical particles [4]. This simulation technique was developed to study the random-walk-like multiple-scattering process of the wave propagation. For details concerning the testing of the Monte Carlo program, we refer to Refs. [4,5]. In the SLAB code, the three-dimensional paths for waves are followed from one scattering to the next as the wave propagates into the medium. Each scattering is assumed to be elastic and is described by the standard Mie theory [6]. The input parameters are the relative refractive index, $m \equiv n_s/n_M = 1.20$, where n_S and n_M are the refractive index of the spheres ($n_S = 1.59$ for polystyrene) and of the surrounding medium ($n_M 1.33$ for water); the size parameter ka; and $kl^* = 1000$. These parameters were chosen for the purpose of comparison with experimental data. The experiments were carried out at room temperature, using a setup similar to that described in Ref. [7], which contains all relevant details. A semiconductor laser emitting at 0.67 μm was used as the source beam. The beam was normally incident on one side of the sample (3 mm thickness) and the scattered light transmitted through the back wall of the sample cell was detected within a collection angle of 2°. The multiply scattering samples were suspensions of polystyrene spheres (PolySciences, Inc.), with mean diameters of 0.22, 0.48, and 1.05 μm, in distilled water at different concentrations.

Results of simulations that were carried out are presented in Figs. 1–3. For a starting point, we refer to Eq. (3) and find, in the limit $ka \ll 1$, that $\xi_L = 0.967l$ and $\xi_C = 0.684l$. The wave becomes depolarized over a distance which is of the order of the mean free path. Now examine Fig. 1(a), which shows the degrees of polarization for linearly and circularly polarized light as a function of d/l for $ka = 1.19$ (i.e., intermediate region) and $ka = 6.43$ (Mie region) in a semilogarithmic plot. The two curves of Fig. 1(a) exhibit linear behavior in this plot. It is interesting to observe that the effect of the input polarization state in these regions is markedly different from that in the Rayleigh region. It is important to appreciate that for $ka \sim 1$, the slopes of these plots do not depend on the incident state of polarization. This is in contrast with the region $ka > 1$, for which these slopes ($\equiv l/\xi_i$) now depend strongly on polarization, the slope being greater for linearly polarized light than for circularly polarized light. Having considered the numerics, we now proceed to compare with data from measurements on suspensions of polystyrene latex spheres. Variations of the degrees of polarization for three values of the size parameter, viz., $ka = 1.23$, 2.69, and 5.89, are shown in Fig. 1(b). It is remarkable that the experiment gives an exponential decay over several decades. The behavior of these data is consistent with the simulation results of Fig. 1(a). Figure 2 shows the characteristic lengths of depolarization for incident linearly, ξ_L/l, and circularly, ξ_C/l, polarized light as a function of the dimensionless size parameter ka. In the case of particles large compared to the wavelength, a linearly or circularly polarized wave becomes depolarized over a distance which is significantly greater than the mean free path. Both the experimental data and simulations show that the ξ's are nearly equal in the region $ka \sim 1$ as can be seen in the inset of Fig. 2 where we have plotted the variation of the length ratio ξ_L/ξ_C with the size parameter ka. For the Rayleigh region, this ratio can be computed exactly using Eq. (3) and is equal to $\sqrt{2}$. In the range of sizes investigated, this ratio is a decreasing function of the parameter ka. As can be seen, the numerical calculations are in quantitative agreement with experimental data. Another interesting feature of Fig. 2

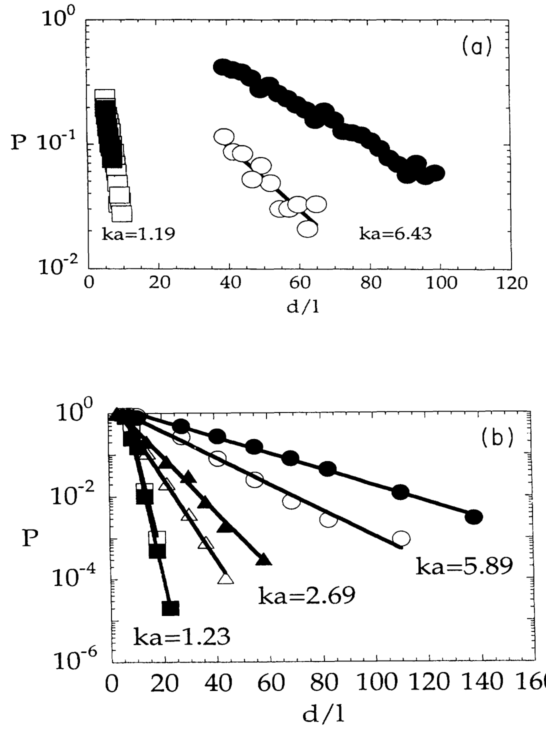

FIG. 1. (a) Semilogarithmic plot of the degrees of polarization for linearly polarized (open symbols) and circularly polarized (solid symbols) light as a function of d/l. Squares represent the intermediate region $ka=1.19$ and circles correspond to the Mie region $ka=6.43$. The lines are exponential fits to the data. (b) Same as (a). Experimental data correspond to measurements on suspensions of polystyrene latex spheres in water: 0.22, (□), 0.48 (△), and 1.05 (○) μm.

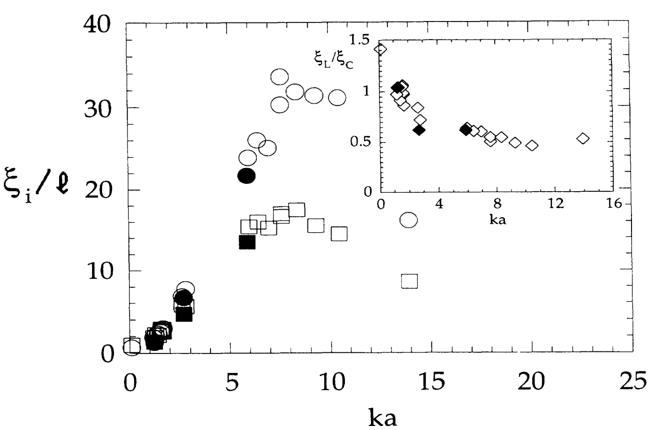

FIG. 2. The characteristic lengths of depolarization, ξ_L/l, for linearly polarized (□) and ξ_C/l, for circularly polarized (○) light as a function of the size parameter ka. The solid symbols indicate experimental data corresponding to measurements on suspensions of polystyrene latex spheres in water. The inset shows the ratio of the characteristic lengths ξ_L/ξ_C (◇) as a function of ka. Experimental data (◆) correspond to measurements on suspensions of polystyrene latex spheres in water.

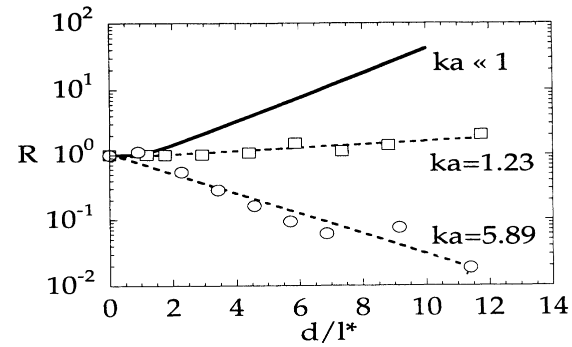

FIG. 3. Semilogarithmic plot of the ratio of the degrees of polarization $R \equiv P_L/P_C$ as a function of d/l^*, for three values of ka. The solid line corresponds to the Rayleigh region ($ka \ll 1$). The symbols indicate experimental data corresponding to measurements on suspensions of polystyrene latex spheres in water [0.22 (□) and 1.05 (○) μm]. The dashed lines are a fit to Eq. (3).

is the maximum of the ξ's observed at $ka \cong 8$ which is then followed by a decrease: We interpret this behavior as arising from Mie resonances. The resonance conditions that occur in large spheres have been examined in some detail [8]. By increasing the size parameter, one could thus hope to achieve a characterization of resonances in the multiple scattering from spheres. This is a difficult task we do not address here. In Fig. 3 we have shown the variations of the ratio of the linear to circular degree of polarization $R \equiv P_L/P_C$ as a function of d/l^*. We find that $\ln(R) \sim d/l^*$. Three behaviors are clearly seen in this plot. On the one hand, R increases according to Eq. (3) in the Rayleigh regime. On the other hand, it can be seen that R is constant in the intermediate regime and decreases in the Mie regime.

The difference between the two types of behavior corresponding to $ka \ll 1$ and $ka > 1$ stems from the anisotropy property of the scattering. In the Mie regime, the scattering is predominantly in the forward direction, while in the Rayleigh region, forward and backward scattered directions are treated on an equal footing. It is also appropriate to recall that the linear character of polarization states is not affected by backscattering, regardless of particle size. On the other hand, backscattering acts as an optical mirror (right $\leftrightarrow$ left) for circularly polarized states, i.e., helicity flip. This explanation forms the heart of the analysis of Mackintosh and co-workers [8]. The helicity being preserved over distances large compared to l^* for large scattering particles explains why the characteristic length for incident circularly polarized light is greater than the corresponding length for incident linearly polarized light. As pointed out in Ref. [8], two distinct mechanisms contribute to the depolarization of circularly polarized light: the randomization of the direction and the randomization of the helicity. However, it is difficult to infer from the preceding arguments what the contribution of these two mechanisms to the total length ξ_C will actually be.

In summary, an efficient Monte Carlo simulation that can provide reliable values of the characteristic lengths of depolarization of multiply scattered radiation from a slab-shaped medium composed of uncorrelated spherical particles has been presented. We focused attention on the polarization and size-parameter dependences of these lengths. A comparison with experimental data indicates a satisfactory agreement. This behavior may be exploited in many areas, including imaging [9], particle sizing [10], and lidar measurements of water droplet clouds [11]. It would be also interesting to extend the analysis described here to calculations aimed at determining the depolarization behavior of anisotropic scattering particles (e.g., circular cross-sectioned cylindrical fibers). Such work is currently in progress. Resonance effects not considered here may be very important, as well.

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