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Circular polarization memory of light

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Circular polarization memory of light multiply scattered by Mie particles is investigated. The mechanism of randomization of helicity is found, in general, to dominate light circular depolarization by particles of large size or a high refractive index while the mechanism of randomization of direction dominates for small particles of a lower refractive index. The characteristic length for circular polarized light to lose its helicity is determined for Mie scatterers of arbitrary size and refractive index and is used successfully to analyze circular depolarization of light transmission through a slab. Circular polarization memory of light is found to be most pronounced for not only large soft particles but also particles of smaller size and a high refractive index.

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The propagation of polarized light in a highly scattering turbid medium is characterized by the randomization of its direction and polarization due to multiple scattering. This randomization accounts for the remarkable success of scalar diffusion theory in describing the transport properties of multiple scattered light [1–6]. Recently, it is becoming increasingly apparent that the vector nature of light plays an important role in diverse phenomena such as coherent backscattering [3,7], diffusing wave spectroscopy [8,9], and the memory effect of circular polarization [8,10]. Polarization of light is important in many real world applications of considerable interest such as optical imaging and optical coherence tomography, and has been used to discriminate short-path photons in a highly scattering medium [11,12], in speckle spectroscopy [13], and to characterize and image objects in turbid media [14–18]. The polarization memory effect is an unexpected long preservation of the incident circular polarization of multiple scattered light by large particles where the wave's helicity (the right-handed or left-handed circular polarization state of light) is randomized less rapidly than is its direction [10]. This effect is in striking contrast to the isotropization of the electric field of linearly polarized light whose depolarization occurs simultaneously with the isotropization of its direction inside a system composed of uncorrelated and noninteracting scatterers [19]. The direction of photons in a scattering medium is randomized by the mean transport free path l_t and isotropized by the isotropization length l_p. The influence of the size parameter and the refractive index on the memory of circular depolarization was later analyzed by various authors using Monte Carlo simulations or the numerical solution of the radiative transfer equation in a slab geometry [20–23]. Two distinct mechanisms contribute to the depolarization of circularly polarized light: the randomization of the direction, and the randomization of the helicity of the field. MacKintosh *et al.* [8,10] analyzed the circular depolarization taking into account the randomization of the helicity. The interplay of the two mechanisms, which determines the memory of circular depolarization, is unclear and yet to be understood [20].

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In this paper we shall clarify the interplay of the randomization of the helicity and the randomization of the direction on the memory of light circular depolarization. We show that the loss of the helicity of multiple scattered light is characterized by one parameter λ_x for Mie scatterers of arbitrary size and refractive index taking into account of both mechanisms for circular depolarization. The decay of the helicity asymmetry follows a power law $(\lambda_x)^n$ with the increase of the number n of scattering events when $n \gg 1$. This introduces the uncoiling length $l_x = l_s / \ln(1/\lambda_x)$ for light to lose its helicity and become circular depolarized where l_s is the mean scattering free path. We shall show that strong circular polarization memory occurs for light scattering by large soft particles. Moreover, strong memory can also occur for light scattering by smaller particles of a high refractive index.

For circularly polarized light, the wave's helicity may preserve or flip at each scattering event depending upon the scattering angle. Denote the preserving and flipping probabilities of the circular polarization states as $p_\pm(\theta)$ where θ is the scattering angle. The probabilities do not depend on the azimuthal angle ϕ for circular polarized light. For Mie scatterers, the scattered electric field of positive and negative helicity is given by $(E'_+, E'_-)^T = A(E_+, E_-)^T$ where

$$A = \begin{pmatrix} \dfrac{S_2 + S_1}{2} & \dfrac{S_2 - S_1}{2} \\ \dfrac{S_2 - S_1}{2} & \dfrac{S_2 + S_1}{2} \end{pmatrix}, \quad (1)$$

in which $S_{2,1}$ are the diagonal elements of the well-known amplitude scattering matrix and responsible for the scattering of the parallel and perpendicular electric field components, respectively, with respect to the scattering plane [24]. The superscript "T" means transpose. As the electric field of positive (negative) helicity light propagating in one direction resides on the same plane and rotates counterclockwise (clockwise) in that plane, the interference between such light of opposite helicity vanishes over time much longer than one period of light, and therefore only the intensities of light of positive or negative helicity need to be considered [30]. The preserving and flipping probabilities of the wave's helicity are proportional to the magnitude square of the diagonal and off-diagonal elements of A, respectively, and given by

$$p_{\pm}(\theta) = \frac{|S_2 \pm S_1|^2}{2(|S_2|^2 + |S_1|^2)}. \quad (2)$$

An analysis of the first mechanism of light circular depolarization was given by Mackintosh *et al.* [8,10]. For an n-scattered photon which encounters a series of scattering of angles $\theta_1, \theta_2, \ldots, \theta_n$ where $\cos\theta_j = \mathbf{s}^{(j-1)} \cdot \mathbf{s}^{(j)}$ and $\mathbf{s}^{(j)}$ is the propagation direction of light after jth scattering, its helicity is either preserved or flips, determined by the even (or odd) number of spin flips in this path. The probability of preserved or flipped helicity of the n-scattered photon becomes

$$P_n^{\pm} = \sum_{\{\sigma_j\}}^{\substack{\text{even}\\ \text{odd}}} \prod_{j=1}^{n} p_{\sigma_j}(\theta_j), \quad (3)$$

where the σ_j is "+" or "−," and the symbols Σ^{even} and Σ^{odd} refer to sums with an even and odd number of "−" in $\{\sigma_j\}$. By an examination of the binomial expansions for $P_n^+ \pm P_n^-$ using Eq. (3), the probabilities $P_n^{\pm}$ were found to be $P_n^{\pm} = \frac{1}{2}(1 \pm a_n)$ where the helicity asymmetry a_n is given by

$$a_n = \prod_{j=1}^{n} [p_+(\theta_j) - p_-(\theta_j)] = \prod_{j=1}^{n} \frac{\mathrm{Re}[S_2(\theta_j) S_1^*(\theta_j)]}{\frac{1}{2}|S_2(\theta_j)|^2 + \frac{1}{2}|S_1(\theta_j)|^2} \quad (4)$$

for a path with the sequence of n specified scattering angles θ_j $(j=1,2,\ldots,n)$ and Re denotes the real part. Equation (4) represents the dependence of the helicity asymmetry on the randomization of the helicity.

The second mechanism for light to circular depolarize is the randomization of its propagation direction. A proper ensemble average must be taken for the probability of preserved or flipped helicity of the n-scattered photon over all the possible intermediate scattering angles, i.e., $\langle P_n^{\pm} \rangle = \frac{1}{2}(1 \pm \langle a_n \rangle)$. The ensemble averaged helicity asymmetry $\langle a_n \rangle$ over all possible series of n scattering events with the propagation direction of light varying from $\mathbf{s}^{(0)}, \mathbf{s}^{(1)}, \ldots,$ to $\mathbf{s}^{(n)}$ can be written as

$$\langle a_n \rangle = \frac{1}{p_n[\mathbf{s}^{(n)}|\mathbf{s}^{(0)}]} \int a_n p[\mathbf{s}^{(n)}|\mathbf{s}^{(n-1)}] \prod_{j=1}^{n-1} p[\mathbf{s}^{(j)}|\mathbf{s}^{(j-1)}] d\mathbf{s}^{(j)}, \quad (5)$$

where $p[\mathbf{s}^{(j)}|\mathbf{s}^{(j-1)}] = C_{\text{sca}}^{-1}\left[\frac{1}{2}|S_2(\theta_j)|^2 + \frac{1}{2}|S_1(\theta_j)|^2\right]$ is the phase function describing the probability of light being scattered from $\mathbf{s}^{(j-1)}$ to $\mathbf{s}^{(j)}$, and $C_{\text{sca}} \equiv \int d\Omega\left[\frac{1}{2}|S_2(\theta)|^2 + \frac{1}{2}|S_1(\theta)|^2\right]$ is the scattering cross section. The n-step transition probability $p_n[\mathbf{s}^{(n)}|\mathbf{s}^{(0)}]$ is given by

$$p_n[\mathbf{s}^{(n)}|\mathbf{s}^{(0)}] = \int p[\mathbf{s}^{(n)}|\mathbf{s}^{(n-1)}] \prod_{j=1}^{n-1} p[\mathbf{s}^{(j)}|\mathbf{s}^{(j-1)}] d\mathbf{s}^{(j)} \quad (6)$$

and approaches $1/4\pi$ when the propagation direction is isotropized after multiple scattering $(n \gg 1)$.

Plugging Eq. (4) into Eq. (5) and expanding $\mathrm{Re}[S_2(\theta_j) S_1^*(\theta_j)]$ into spherical harmonics, Eq. (5) can be integrated to obtain

$$\langle a_n \rangle = \frac{1}{4\pi p_n[\mathbf{s}^{(n)}|\mathbf{s}^{(0)}]} \sum_{l=0}^{\infty} (2l+1) \Lambda_l^n P_l[\mathbf{s}^{(0)} \cdot \mathbf{s}^{(n)}], \quad (7)$$

where P_l is the Legendre polynomial and Λ_l is given by

$$\Lambda_l = \frac{\int d\Omega\, \mathrm{Re}[S_2(\theta) S_1^*(\theta)] P_l(\cos\theta)}{\int d\Omega\left[\frac{1}{2}|S_2(\theta)|^2 + \frac{1}{2}|S_1(\theta)|^2\right]}. \quad (8)$$

Two special cases deserve attention. For very large particles for which $S_2 \simeq S_1$, the flipping probability $p_- = 0$ and hence $a_n = \langle a_n \rangle = 1$. For Rayleigh scatterers, $S_2 \propto \cos\theta$ and $S_1 \propto 1$, and hence $\Lambda_l = \frac{1}{2}\delta_{l1}$ and $\langle a_n \rangle = \left(\frac{1}{2}\right)^n 3\mathbf{s}^{(0)} \cdot \mathbf{s}^{(n)}$. In general, only the leading two terms of Λ_0 and Λ_1 for $l=0$ and 1 dominate Eq. (7) when $n \gg 1$ [25]. The helicity asymmetry for any Mie particle after multiple scattering $(n \gg 1)$ can now be written as

$$\langle a_n \rangle \simeq \Lambda_0^n + 3\Lambda_1^n \mathbf{s}^{(0)} \cdot \mathbf{s}^{(n)}, \quad (9)$$

where we have approximated $p_n[\mathbf{s}^{(n)}|\mathbf{s}^{(0)}]$ by $1/4\pi$.

Equations (7) and (9) give the helicity asymmetry of light emerging in the direction $\mathbf{s}^{(n)}$ after n scattering events taking into account of both mechanisms for light circular depolarization. The contribution due to randomization of helicity only is independent for each scattering event and can be computed by integrating Eq. (4) over the scattering angles weighted by the phase function to yield $\langle a_n \rangle_{\text{helicity}} = \Lambda_0^n$. The difference $\langle a_n \rangle - \langle a_n \rangle_{\text{helicity}} \simeq 3\Lambda_1^n \mathbf{s}^{(0)} \cdot \mathbf{s}^{(n)}$ can be regarded as the contribution from randomization of direction. Either mechanism may dominate light circular depolarization dependent on the size and refractive index of the particle and the helicity asymmetry can simply be written as

$$\langle a_n \rangle \simeq \alpha \lambda_x^n, \quad (10)$$

where $\alpha \lambda_x^n$ is the leading term in Eq. (9) when $n \gg 1$. The probability of preserved or flipped helicity of a photon traveling a path of length s, taking into both mechanisms of light circular depolarization, is then

$$P_{\pm}(s) = \frac{1}{2}(1 \pm \alpha \lambda_x^n) = \frac{1}{2}(1 \pm \alpha e^{-s/l_x}), \quad (11)$$

where

$$l_x \equiv l_s / \ln \frac{1}{\lambda_x} \quad (12)$$

is the uncoiling length, which is the characteristic length for light to lose its memory of circular polarization; l_s is the mean scattering free path, and s/l_s is the mean number of scattering events experienced by the photon.

The leading Λ_l $(l=0,1,2)$ are plotted in Fig. 1 for the spherical particles of moderate refractive index $m=1.1$ and 1.2. Λ_1 starts from 0.5. Λ_0 starts from 0 and increases much faster than Λ_1 and becomes the dominating one when the size parameter $x > x^* \sim 1.5$ where the size parameter x

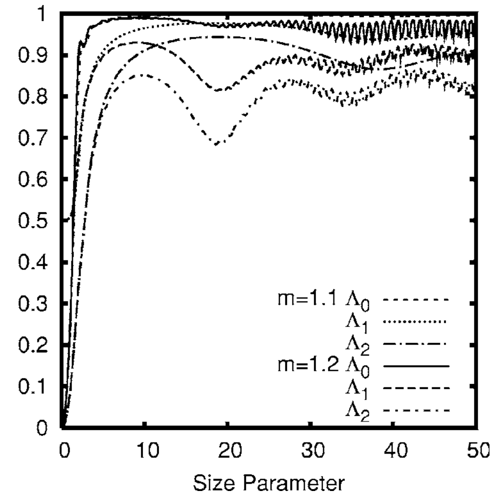

FIG. 1. The leading Λ_0 and Λ_1 for spherical particles of refractive index $m=1.1,1.2$. Λ_3 is also plotted for comparison.

$\equiv 2\pi a/\lambda$, a is the radius of the particle, and λ is the wavelength of light in the medium.

The helicity asymmetry for particles of moderate refractive index is dominated by the mechanisms of randomization of helicity and randomization of direction for large and small particles, respectively, and can be written as

$$\langle a_n\rangle \simeq \begin{cases} \Lambda_0^n, & x > x^* \\ 3\mathbf{s}^{(0)}\cdot\mathbf{s}^{(n)}\Lambda_1^n, & x < x^*, \end{cases} \tag{13}$$

except that $\langle a_n\rangle=\Lambda_0^n$ whatever the size of the particle when $\mathbf{s}^{(0)}\cdot\mathbf{s}^{(n)}=0$. Equations (11) and (13) tell that circular polarized light is depolarized uniformly ($\lambda_x=\Lambda_0$) across all observation directions (independent on the observation direction $\mathbf{s}^{(n)}$) for large particles ($x>x^*$). For smaller scatterers ($x<x^*$), circular polarized light is depolarized faster in the exact 90° direction ($\mathbf{s}^{(0)}\cdot\mathbf{s}^{(n)}=0$) with $\lambda_x=\Lambda_0$ than in the other observation directions with $\lambda_x=\Lambda_1>\Lambda_0$.

The ratio of the uncoiling length l_x over the transport mean free path $l_t=l_s/(1-g)$ where g is the average cosine of scattering angles vs the size parameter and the refractive index of the particle is displayed in Fig. 2. For Rayleigh scatterers, $l_x=l_s/\ln 2=1.44l_s$ and $l_t=l_s$, agrees with the previous analysis [8,20]. Figure 2 clearly demonstrates that l_x/l_t oscillates with the size and refractive index of the particle and a larger particle may not necessarily lead to a stronger preservation of circular polarization. The fine structure in l_x/l_t comes from Mie resonance scattering and l_x/l_t can be much larger than unity, indicating a strong circular polarization memory. For large soft particles, the ratio can be found to grow roughly as $2\ln x$ based on the approximate amplitude scattering matrix for soft particles [26], reducing to the result of Gorodnichev *et al.* in the large soft particle limit [27] [see Fig. 2(a)]. Surprisingly, however, a much stronger memory of circular polarization is observed for particles of smaller size and high refractive index; the ratio $l_x/l_t\sim 34$ for $x=1.5$ and $m=1.83$ [see Fig. 2(b)]. It should be noted that the value of l_x/l_t can be much higher for smaller x and with suitably larger m.

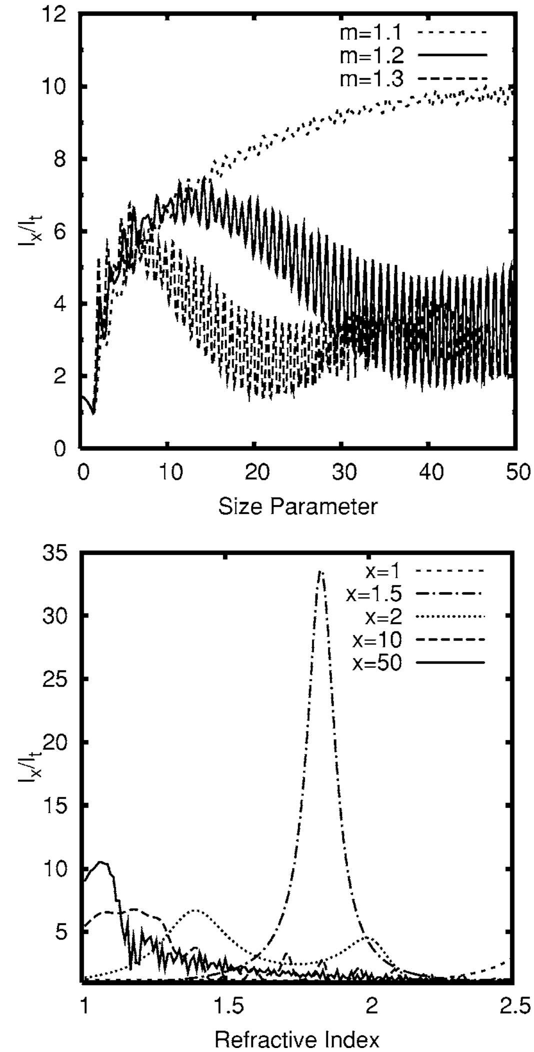

FIG. 2. The ratio of the uncoiling length l_x over the transport mean free path l_t vs (a) size parameter and (b) refractive index of the particle. This ratio can be much larger than unity, i.e., a strong circular polarization memory, for large soft particles and smaller particles of high refractive index.

An important quantity of interest is the circular depolarization of light transmission through a slab of scattering medium. A circular depolarization length ξ_C is commonly used to quantify the depolarization of circular polarized light [20,21]. The length ξ_C, unlike the uncoiling length l_x, is only appropriate for light transmission through a slab. ξ_C relates to l_x through the expression

$$\xi_C = \left(\frac{l_x l_t}{3}\right)^{1/2} \tag{14}$$

for diffusive light transmission through a slab [20]. The degree of circular polarization of light transmitted in the forward direction is given by

$$X_{\mathrm{DOCP}} = \beta\frac{L+2z_e}{l_s}e^{-L/\xi_C}, \tag{15}$$

assuming diffusion of light through the slab where L is the thickness of the slab, z_e is the extrapolation length [28] on

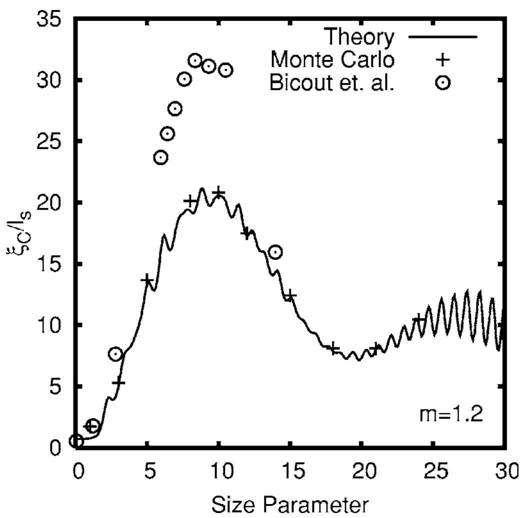

FIG. 3. The circular depolarization length ξ_C for light transmission through a slab scaled by the mean scattering free path l_s. The symbols "+" denote the ratio ξ_C/l_s obtained from Monte Carlo simulations and "○" from Fig. 2 in Bicout *et al.* [20].

the left and right boundaries in the diffusion approximation, and β is a scaling parameter. The difference between Eq. (15) and Eq. (3) given in [20] is that a more realistic extrapolation boundary condition is used here. Equation (15) can be used to extract ξ_C from experimental measurements or Monte Carlo simulations of forward light transmission through slabs of increasing thickness.

Figure 3 displays ξ_C as predicted by Eq. (14) using the uncoiling length Eq. (12) (solid curve) and also from fitting Eq. (15) to the degree of circular polarization of light transmission through slabs of increasing thickness and matched refractive index using Monte Carlo simulations [29] (with symbols "+"). We also plot here data reproduced from Fig. 2 of [20] (with symbols "○"). In fitting via Eq. (15), z_e is fixed at $\frac{2}{3}l_t$ [28]. Good agreement is found between the first two sets of data. However, the values of ξ_C given in [20] within the region of size parameter $5<x<10$ do not agree with our results. This may originate from the fact that a different boundary condition ($z_e=0$) was used in [20].

In conclusion, we have shown that the loss of the helicity of multiple scattered light is characterized by one parameter λ_x for Mie scatterers of arbitrary size and a refractive index and clarified the interplay of the randomization of the direction and the randomization of the helicity on the memory of light circular depolarization. The mechanisms of randomization of helicity and randomization of direction in general dominate circular depolarization by particles of large size or a high refractive index and small particles of a lower refractive index, respectively. A characteristic length scale, the uncoiling length l_x, for light to lose its helicity and become circular depolarized has been introduced and used successfully to analyze circular depolarization of light transmission through a slab. Strong memory of circular polarization is found to occur for light scattering by not only large soft particles but also smaller particles of a high refractive index.

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