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Diffusion of light through a disordered ensemble of resonant Mie-particles embedded in a medium with circular dichroism

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Abstract. We study transmission of unpolarized light (incoherent superposition of right-hand circular and left-hand circular polarized waves) through an optically active medium doped by scattering Mie-particles. The medium is assumed to possess circular dichroism. Within the spatial diffusion approximation, the degree of circular polarization of the transmitted radiation is calculated. It is shown, that in the presence of scatterers a significant increase in the circular polarization compared to the medium with no scatterers can be observed.

1. Introduction

In an absorbing medium with optical activity, circular dichroism can be observed: the absorption coefficients for the right- and left-handed polarizations differ from each other (see, e.g., [1]). The circular dichroism is very sensitive to the configuration of complex molecules, as well as to conformational transitions in them. Therefore, measuring the circular dichroism is one of the most important methods of stereochemical analysis [2, 3]. The main experimental difficulty in quantifying the circular dichroism characteristics is due to a weakness of the effect [2–4].

In the last decade, in the context of nanophotonics (see, e.g., [5]), the optical properties of particles with a high refractive index in the visible and near infrared ranges have been studied in detail [6–9]. Near the first two Mie-resonances, the so-called first Kerker condition [10] has been shown to be fulfilled (see, e.g., [6, 11]). If the particle parameters satisfy the Kerker condition, scattering in the backward direction is suppressed and, in addition, circularly polarized light does not change its polarization upon scattering [7]. The depolarization of the waves becomes noticeable only after a great number of scattering events [8,9]. In the case of the spatial diffusion of light in such a medium, the attenuation of circular polarization occurs very slowly. The results [8, 9] enable us to expect that the addition of Mie-particles with the optical parameters and sizes satisfying the Kerker condition to a homogeneous medium, on the one hand, should lead to a significant increase in the photon path lengths in the medium, and, on the other hand, should not disrupt the initial circular polarization of the waves propagating in the medium. When the homogeneous medium possesses circular dichroism (the absorption coefficients of right-hand and left-hand polarized waves differ from each other), the addition of Mie-particles should result in a substantial increase in the difference between the intensities of the differently polarized waves, I_R and I_L. The degree of circular polarization of the transmitted radiation in the medium with no scattering $|I_R - I_L| / (I_R + I_L) = \Delta\varkappa L/2$ ($\Delta\varkappa$ is the difference between the corresponding absorption coefficients, L is the sample thickness), is replaced by $|I_R - I_L| / (I_R + I_L) = \Delta\varkappa S/2$, where S is the average path length of photons in the sample. In the diffusion regime, the inequality $S \gg L$ is satisfied, and a significant increase in the ratio $|I_R - I_L| / (I_R + I_L)$ should occur. Thus, similarly to such systems as a "random laser" (see, e.g., [12]) and a scattering host medium for measuring weak absorption in liquids and gases [13, 14], a disordered ensemble of nearly resonant Mie-particles embedded in a medium with circular dichroism acts as a spatially distributed resonator which increases the photon path lengths in the sample without violating their circular polarization.

Below we study multiple scattering of initially unpolarized light (incoherent superposition of right-hand and left-hand polarized waves) in a system of Mie-particles embedded in an optically active homogeneous medium. The optical parameters of the Mie-particles are assumed to satisfy the first Kerker condition. Within the diffusion approximation, the degree of circular polarization arising in transmission of light through the medium is calculated. The degree of circular polarization is shown to drastically increase in comparison with the case of a homogeneous medium. The dependence of the degree of polarization and the enhancement factor on the sample thickness are calculated.

2. Transport equations for circularly polarized components of light

As shown in Refs. [8, 9], near the first Kerker point, the single-scattering matrix takes the form:

$$\hat{d} = \begin{pmatrix} a_1 & 0 & 0 & 0 \\ 0 & a_1 & 0 & 0 \\ 0 & 0 & a_2 & 0 \\ 0 & 0 & 0 & a_2 \end{pmatrix} \tag{1}$$

where a_1 and a_2 are expressed in terms of the scattering amplitudes $A_\parallel$ and $A_\perp$ of the cross-polarized waves, $a_1 = \left(\left|A_\parallel\right|^2 + |A_\perp|^2\right)/2$ and $a_2 = \mathrm{Re}\, A_\parallel A_\perp^*$ [15]. Under the assumption that the optical activity effect is small, the amplitudes $A_\parallel$ and $A_\perp$ can be calculated without taking into account this effect.

For a medium with circular dichroism, right-hand and left-hand circularly polarized waves are the eigenmodes of the medium [16] and, therefore, multiple scattering of light in such a medium is conveniently described in terms of the Stokes vector in the circular representation [17–19]

$$\hat{I} = \frac{1}{\sqrt{2}} \begin{pmatrix} Q - \mathrm{i}U \\ I - V \\ I + V \\ Q + \mathrm{i}U \end{pmatrix} \tag{2}$$

where I, Q, U V are the Stokes parameters in the standard linear representation [15]. The values $I \pm V$ are proportional to the intensities of right-hand and left-hand polarized waves, $I_{R,L} = (I \mp V)/2$.

The vector radiative transfer equation with the scattering matrix (1) decouples into two independent systems of equations. One system involves the Stokes parameters $Q \mp \mathrm{i}U$ (Q and U characterize the linear polarization of light), the other system describes the circularly polarized components $I_{R,L}$ [8, 9]. So, if the first Kerker condition is satisfied, the linear and circular polarizations evolve independently of each other.

The intensities I_R and I_L of the circularly polarized components obey the system of equations (see, e.g., [18–20])

$$\left[\mu\frac{\partial}{\partial z}+n_0\sigma+\varkappa\right]I_R\left(z,\mu\right)-\frac{\Delta\varkappa}{2}I_R\left(z,\mu\right)= \quad n_0\int d\mathbf{n}'\;a_+\left(\mathbf{nn}'\right)\cdot I_R\left(z,\mu'\right)+ n_0\int d\mathbf{n}'\;a_-\left(\mathbf{nn}'\right)\cdot I_L\left(z,\mu'\right) \tag{3}$$

$$\left[\mu\frac{\partial}{\partial z}+n_0\sigma+\varkappa\right]I_L\left(z,\mu\right)+\frac{\Delta\varkappa}{2}I_L\left(z,\mu\right)= \quad n_0\int d\mathbf{n}'\;a_+\left(\mathbf{nn}'\right)\cdot I_L\left(z,\mu'\right)+ n_0\int d\mathbf{n}'\;a_-\left(\mathbf{nn}'\right)\cdot I_R\left(z,\mu'\right) \tag{4}$$

where $\mu=\mathbf{nn}_{int}$, $\mathbf{n}$ is the direction of wave propagation, $\mathbf{n}_{int}$ is the inner normal to the sample surface, $a_\pm\left(\mathbf{nn}'\right)=\left(a_1\left(\mathbf{nn}'\right)\pm a_2\left(\mathbf{nn}'\right)\right)/2$, σ is the cross-section of elastic scattering of light by Mie-particles, n_0 is their number per unit volume, $\varkappa$ is the average absorption coefficient of the medium, $\Delta\varkappa$ is the difference in the absorption coefficients of right-hand and left-hand circularly polarized waves. The value of $\Delta\varkappa$ is the magnitude of circular dichroism in a homogeneous medium [2, 16].

3. Spatial diffusion of light

A solution to the Eqs.(3) and (4) can be written in the form of expansion in a series in the Legendre polynomials (see, e.g., [18, 19]):

$$I_{R,L}\left(z,\mu\right)=\sum_{l=0}^{\infty}\frac{2l+1}{4\pi}\;I_{R,L}\left(z,l\right)P_l\left(\mu\right) \tag{5}$$

The coefficients $I_{R,L}\left(z,l\right)$ appearing in Eq.(5) are subject to the equations

$$\frac{l}{2l+1}\frac{\partial I_R\left(z,l-1\right)}{\partial z}+\frac{l+1}{2l+1}\frac{\partial I_R\left(z,l+1\right)}{\partial z}+\left(\sigma_{tot}^{(R)}-n_0a_+(l)\right)I_R\left(z,l\right)=n_0a_-(l)I_L\left(z,l\right) \tag{6}$$

$$\frac{l}{2l+1}\frac{\partial I_-\left(z,l-1\right)}{\partial z}+\frac{l+1}{2l+1}\frac{\partial I_-\left(z,l+1\right)}{\partial z}+\left(\sigma_{tot}^{(L)}-n_0a_+(l)\right)I_-\left(z,l\right)=n_0a_-(l)I_+\left(z,l\right) \tag{7}$$

where $\sigma_{tot}^{(R,L)}=n_0\sigma+\varkappa\mp\Delta\varkappa/2$ and

$$a_\pm\left(l\right)=\pi\int_{-1}^{1}d\mu\;\left(a_1\left(\mu\right)\pm a_2\left(\mu\right)\right)P_l(\mu) \tag{8}$$

The coefficient $a_-(l=0)$ is proportional to the depolarization cross section σ_{dep} introduced in [21], $a_-(l=0)=\sigma_{dep}/2$, where $\sigma_{dep}=\int d\mathbf{n}'\left(a_1\left(\mathbf{nn}'\right)-a_2\left(\mathbf{nn}'\right)\right)$. If the depolarization cross section of σ_{dep} is much smaller than the transport cross section of elastic scattering $\sigma_{dep}\ll\sigma_{tr}=\int d\mathbf{n}'\left(1-\mathbf{nn}'\right)a_1\left(\mathbf{nn}'\right)$, the effect of slow decay of circular polarization is observable [21]. In this case, the depolarization of circularly polarized light occurs after the onset of the regime of spatial diffusion of radiation. This effect is most pronounced, if the first Kerker condition is fulfilled and the ratio σ_{dep}/σ_{tr} reaches its minimum value [8, 9].

Under the conditions of spatial diffusion in a weakly absorbing medium (we assume that $\varkappa\ll n_0\sigma_{tr}$), the angular distribution of radiation turns out to be close to isotropic one, and only the first two terms with $l = 0$ and $l = 1$ should be left in the expansion (5) [18,19,21]. Then, Eqs.(6) and (7) are reduced to the following system of diffusion equations for $I_{R,L}(z) = I_{R,L}(z, l = 0)$:

$$\left[\frac{\partial^2 I_R(z)}{\partial z^2} - 3n_0\sigma_{tr}\Sigma_R I_R(z)\right] + \frac{3}{2}n_0\sigma_{tr}\sigma_{dep}I_L(z) = 0,$$
$$\left[\frac{\partial^2 I_L(z)}{\partial z^2} - 3n_0\sigma_{tr}\Sigma_L I_L(z)\right] + \frac{3}{2}n_0\sigma_{tr}\sigma_{dep}I_R(z) = 0 \qquad (9)$$

where the effective absorption coefficients appearing in the system (9) are equal to $\Sigma_{R,L} = \varkappa \mp \Delta\varkappa/2 + n_0\sigma_{dep}/2$.

In the limit $L \gg l_{tr}$ the boundary conditions to the diffusion equations (9) can be written as $I_{R,L}(z = 0) = I_{R,L}(z = L) = 0$. For unit flux of unpolarized light (incoherent superposition of right-hand and left-hand polarized light) incident on the boundary $z = 0$, the intensities $I_{R,L}$ of radiation transmitted in the forward direction has the form

$$I_R(L, \mu = 1) = \frac{1}{8\pi(1+\eta^2)}\left[(1+\eta)\frac{\varepsilon_- l_{tr}}{\sinh \varepsilon_- L} - \eta(1-\eta)\frac{\varepsilon_+ l_{tr}}{\sinh \varepsilon_+ L}\right] \qquad (10)$$

$$I_L(L, \mu = 1) = \frac{1}{8\pi(1+\eta^2)}\left[\eta(1+\eta)\frac{\varepsilon_- l_{tr}}{\sinh \varepsilon_- L} + (1-\eta)\frac{\varepsilon_+ l_{tr}}{\sinh \varepsilon_+ L}\right] \qquad (11)$$

where the parameter η entering into Eqs.(10) and (11) is equal to $\eta = \left[\sqrt{(n_0\sigma_{dep})^2 + (\Delta\varkappa)^2} - \Delta\varkappa\right] / (n_0\sigma_{dep})$. The attenuation coefficients ε_- and ε_+ are determined by

$$\varepsilon_{\mp} = \sqrt{3n_0\sigma_{tr}\left[\varkappa + \frac{1}{2}\left(n_0\sigma_{dep} \mp \sqrt{(n_0\sigma_{dep})^2 + (\Delta\varkappa)^2}\right)\right]} \qquad (12)$$

According to Eq.(12), the quantities $\varkappa + \frac{1}{2}\left(n_0\sigma_{dep} \mp \sqrt{(n_0\sigma_{dep})^2 + (\Delta\varkappa)^2}\right)$ act as the effective absorption coefficients for waves with different polarizations in the medium with circular dichroism. If the depolarization process can be neglected ($n_0\sigma_{dep} \ll \Delta\varkappa$), we return to the initial definition of the absorption coefficients for right-hand and left-hand polarized waves in a homogeneous medium with the circular dichroism, $\varkappa \mp \Delta\varkappa/2$. In the opposite case of noticeable depolarization ($n_0\sigma_{dep} \gg \Delta\varkappa$), the difference between the effective absorption coefficients turns out to be small (the difference is proportional to $(\Delta\varkappa/n_0\sigma_{dep})^2$).

The total intensity $I = I_R + I_L$ and the difference between the intensities of right-hand and left-hand polarized waves $I_L - I_R$ (i.e., the fourth Stokes parameter V) are determined by the relations

$$I(L, \mu = 1) = \frac{1}{4\pi(1+\eta^2)}\left[(1+\eta)^2\frac{\varepsilon_- l_{tr}}{\sinh \varepsilon_- L} + (1-\eta)^2\frac{\varepsilon_+ l_{tr}}{\sinh \varepsilon_+ L}\right] \qquad (13)$$

$$V(L, \mu = 1) = \frac{1-\eta^2}{4\pi(1+\eta^2)}\left[\frac{\varepsilon_- l_{tr}}{\sinh \varepsilon_- L} - \frac{\varepsilon_+ l_{tr}}{\sinh \varepsilon_+ L}\right] \qquad (14)$$

In the absence of circular dichroism ($\Delta\varkappa = 0$) Eq.(13) transforms to the well-known formula for the intensity I of the scalar theory (see, e.g., [21]). In this case the circular polarization does not arise, and the fourth Stokes parameter is zero, $V = 0$.

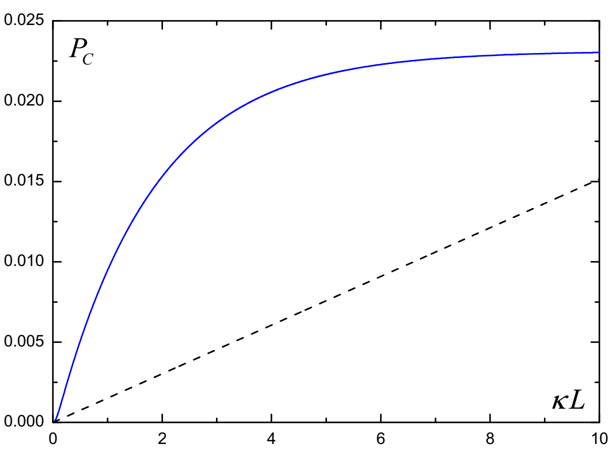

Figure 1. Degree of circular polarization for initially unpolarized light vs sample thickness L. The numerical calculations were carried out for $\Delta\varkappa/\varkappa = 3\cdot 10^{-3}$, $\sigma_{dep}/\sigma_{tr} = 10^{-3}$, $n_0\sigma_{tr}/\varkappa = 100$. The solid blue and dashed black lines correspond to the scattering medium and the medium with no scattering centers ($n_0 = 0$), respectively.

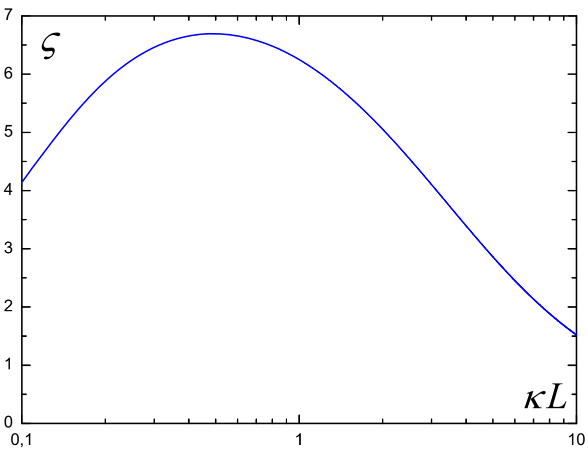

Figure 2. Enhancement factor ζ == $P_C/P_C(n_0 = 0)$ vs sample thickness L. The parameters of the scattering medium are the same as in Fig. 1.

4. Discussion

For the initially unpolarized beam, the degree of circular polarization of radiation transmitted through the medium can be calculated with the ratio $P_C = |V|/I$ where V and I are determined by Eqs.(13) and (14). For a relatively thin sample, $l_{tr} \ll L \ll l_D$ ($l_{tr} = (n_0\sigma_{tr})^{-1}$ is the transport mean free path, $l_D = (3n_0\sigma_{tr}\varkappa)^{-1/2}$ is the diffusion length (see, e.g., [22]) the degree of polarization increases with L as

$$P_C = \frac{\Delta\varkappa}{2}\left(\frac{L^2}{l_{tr}}\right) \tag{15}$$

In the limiting case of very thick samples, $L \gg l_{circ}$, ($l_{circ} = (\varepsilon_+ - \varepsilon_-)^{-1}$ is the length of circular polarization attenuation [18,19]), the growth of the degree of polarization saturates and P_C tends to the limiting value

$$P_C = \frac{\Delta\varkappa}{\sqrt{(n_0\sigma_{dep})^2 + (\Delta\varkappa)^2} + n_0\sigma_{dep}} \tag{16}$$

An example of numerical calculations for the L-dependence of P_C is shown in Fig. 1. We use the typical value of the ratio $\Delta\varkappa/\varkappa$ [2] and the value of σ_{dep}/σ_{tr} corresponding to the first Kerker point for Si particles [8, 9]. From the figure it follows that the addition of scattering Mie-particles to a homogeneous medium with circular dichroism leads to noticeable increase of the degree of polarization at relatively small thickness L. For characterizing the increase in the degree of polarization compared to the case of a homogeneous medium, we introduce the enhancement factor $\zeta = P_C/P_C(n_0 = 0)$. The L-dependence of this factor is illustrated in Fig. 2 for the same parameters of the medium as in Fig. 1. The enhancement factor peaks at $L \sim 1/\varkappa$. The maximum value of ζ can be estimated as $\zeta_{max} \sim \sqrt{n_0\sigma_{tr}/\varkappa}$.

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5. Conclusions

In conclusion we have studied the propagation of initially unpolarized light (incoherent superposition of right-hand and left-hand polarized waves) in a medium with circular dichroism containing randomly spaced Mie-particles. The degree of circular polarization in the transmitted beam has been shown to increase due to the presence of scatterers. This effect is most pronounced if the first Kerker condition for Mie-particles is fulfilled and the ratio of the depolarization cross section to the transport cross section reaches its minimum value. In this case, the disordered ensemble of Mie-particles acts as an optical resonator which leads to an increase in photon path lengths in the medium without changing the state of their circular polarization.

The results obtained above can be of interest for applications in studies of optically active liquids which can be easily doped by scattering centers.

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References

[1] Barron L D 2004 *Molecular light scattering and optical activity* (Cambridge University Press)
[2] Cantor C R and Schimmel P R 1980 *Biophysical chemistry: Part II "Techniques for the study of biological structure and function"* (WH Freeman and Co, Oxford)
[3] Berova N, Nakanishi K and Woody R W 2000 *Circular Dichroism: Principles and Applications* (Wiley).
[4] Preda F, Perri A, Réhault J, Dutta B, Helbing J, Cerullo G and Polli D 2018 *Opt. Letters* **43** 1882
[5] Kuznetsov A I, Miroshnichenko A E , Brongersma M L , Kivshar Yu S and Lukýanchuk B 2016 *Science* **354** 846
[6] Nieto-Vesperinas M, Gomez-Medina R and Saenz J J 2011 *J. Opt. Soc. Am.* **A28** 54
[7] Schmidt M K, Aizpurua J, Zambrana-Puyalto X, Vidal X, Molina-Terriza G and Saenz J J 2015 *Phys.Rev.Lett.* **114** 113902
[8] Gorodnichev E E, Kuzovlev A I and Rogozkin D B 2016 *JETP Lett.* **104** 157
[9] Gorodnichev E E, Kuzovlev A I and Rogozkin D B 2016 *Quantum Electronics* **46** 947
[10] Kerker M, Wang D-S and Giles C L 1983 *J. Opt. Soc. Am.* **73** 765
[11] Valuckas V, Paniagua-Dom?nguez R, Fu Y H, Lukýanchuk B and Kuznetsov A I 2017 *Appl. Phys. Lett.* **110** 091108
[12] Gottardo S, Sapienza R, Garcia P D, Blanco A, Wiersma D S and López C 2008 *Nat. Photonics* **2** 429
[13] Koman V B, Santschi C and Martin O J F 2015 *Anal. Chem.* **87** 1536
[14] Mupparapu R, Vynck K, Svensson T, Burresi M and Wiersma D S 2015 *Optics Express* **23** 1472
[15] Newton R. G 1982 *Scattering Theory of Waves and Particles* (2nd ed., New York, Springer)
[16] Bohren C F and Huffman D R 2008 *Absorption and Scattering of Light by Small Particles* (Wiley)
[17] Kuscer I and Ribaric M 1959 *Opt. Acta* **6** 42
[18] Gorodnichev E E, Kuzovlev A I and Rogozkin D B 2007 *JETP* **104** 319
[19] Gorodnichev E E, Kuzovlev A I and Rogozkin D B 2014 *Phys.Rev.* E **90** 043205
[20] Kokhanovsky A A 1999 *Phys.Rev.* **E4** 4899
[21] Gorodnichev E E, Kuzovlev A I and Rogozkin D B 1998 *JETP Lett.* **68** 22
[22] Ishimaru A 1997 *Wave Propagation and Scattering in Random Media* (Institute of Electrical and Electronics Engineers)