Radiative transfer in chiral random media

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This paper is devoted to the investigation of polarization and radiative characteristics of coherent and diffused light beams in isotropic media with optically active particles. Simple solutions for the Stokes vectors of the direct and diffused beams are obtained in the framework of the vector radiative transfer theory. Results obtained can be used for the generalization of the circular dichroism and optical rotation dispersion spectroscopy for the case of disperse media. [S1063-651X(99)15210-3]

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I. INTRODUCTION

All natural products, which play an essential part in the phenomena of vegetable and animal life, are asymmetric [1]. As a result, left-handed and right-handed circularly polarized electromagnetic waves propagate through such media with different velocities. This produces the rotation of the polarization plane of incident linearly polarized light beams. Chiral media are also characterized by circular dichroism or different absorption of left-handed and right-handed circularly polarized waves. The circular dichroism (CD) spectra and optical rotatory dispersion (ORD) are standard tools in the stereochemistry of organic molecules [2].

The interpretation of both the circular dichroism and the optically rotatory dispersion curves becomes much more complex if molecules build agglomerates. The scattering of light plays an essential role in this case [3–8]. Thus, chemists try to avoid this complication and make measurements for uniform media. However, the scattering of light cannot be avoided in many cases. This is true, e.g., for bioparticles, including red blood cell membranes, viruses nuclei, mitochondria, and ribosoms. One cannot dilute such media without destroying their structural elements. It is important to have a chance to monitor the rearrangement of chemical groups in bioparticles during their lifecycles.

The interpretation of ORD and CD spectra of particulate media of any geometrical thickness can be done on the base of the radiative transfer theory (RTT) [9,10] that was initially applied in the field of astrophysics for studying the photon transport in planetary atmospheres, interstellar dust, and a great variety of astrophysical objects.

The scattered light transforms from the artifact to the valuable source of the information on the microstructure of media under investigation if one applies the RTT to the problem in question. It is possible to use both reflection and transmission schemes in the CD and ORD spectroscopy for different angles of observations, wavelengths, and polarization states of the incident light in the framework of the RTT, which provides more information for the solution of the inverse problem.

The main task of this paper is the introduction to the modern radiative transfer theory for the analysis of the optical properties of disperse media with optically active particles. We formulate the vector radiative transfer equation in chiral media and obtain its analytical solution for thin plane-parallel layers in terms of the elements of the extinction and scattering matrices.

II. THEORY

A. General equations

The change of the energy and state of the polarization of the photon flux in the chiral plane-parallel slabs can be described by the following Boltzman type equation [10–14]:

$$\mu \frac{d\vec{J}(\vec{\Omega},z)}{dz} = -\hat{\sigma}_{\rm ext}\vec{J}(\vec{\Omega},z) + \int_{4\pi} \hat{\sigma}_{\rm sca}(\vec{\Omega}' \to \vec{\Omega})\vec{J}(\vec{\Omega}',z)\,d\vec{\Omega}', \tag{1}$$

where $\vec{J}(\vec{\Omega},z)=(I,Q,U,V)$ is the Stokes vector of the light beam in the direction $\vec{\Omega}(\Theta,\phi)$ at the geometrical depth z, $\hat{\sigma}_{\rm ext}$ is the extinction matrix, $\hat{\sigma}_{\rm sca}(\vec{\Omega}'\to\vec{\Omega})$ is the differential scattering matrix, and μ is the cosine of the observation angle. The last term in Eq. (1) describes the process of multiple scattering of photons in chiral media. The positive Z direction is pointing from the top to the bottom of a layer.

Note that the components of the Stokes vector can be expressed via components of the electric vector of the scattered wave, propagating in the direction $\vec{e}_3=\vec{e}_2\times\vec{e}_1$,

$$\vec{E}=E_1\vec{e}_1+E_2\vec{e}_2. \tag{2}$$

They are defined by the following equations [9,15]:

$$I=E_1E_1^*+E_2E_2^*, \tag{3}$$

$$Q=E_1E_1^*-E_2E_2^*, \tag{4}$$

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$$U = E_1 E_2^* + E_2 E_1^*, \tag{5}$$

$$V = i(E_1 E_2^* - E_2 E_1^*), \tag{6}$$

where an asterisk denotes the conjugate complex value. We omit a common multiplier in Eqs. (3)–(6) for the sake of simplicity. One can see that Eq. (3) determines the intensity of the light field. The values of I, Q, U, and V completely characterize the arbitrarily polarized light beam in terms of the intensity, degree of polarization, and characteristics of the polarization ellipse (the ellipticity e, the direction of rotation, and azimuth ψ). The ellipticity $e \epsilon [0,1]$ is defined as the ratio of axes of the polarization ellipse. The value of $e = 0$ corresponds to the linearly polarized beam. The case $e = 1$ holds for the circularly polarized light. The azimuth ψ defines the orientation of the polarization ellipse. One can introduce the ellipticity angle φ as well. The absolute value of this angle is equal to $\arctan e$ and the sign defines the direction of the rotation of the polarization ellipse.

Light beams with the same values of the Stokes vector cannot be distinguished by polarization measurements determining quadratic quantities (e.g., $\langle \vec{E}\vec{E}^* \rangle$). However, these radiation fluxes can differ, since they can have different high-range field correlators.

The components of the Stokes vector can be rewritten in terms of the amplitudes a_1, a_2 and phases σ_1, σ_2 of a simple electromagnetic wave as well:

$$I = a_1^2 + a_2^2, \tag{7}$$

$$Q = a_1^2 - a_2^2, \tag{8}$$

$$U = 2 a_1 a_2 \cos(\sigma_1 - \sigma_2), \tag{9}$$

$$V = 2 a_1 a_2 \sin(\sigma_1 - \sigma_2), \tag{10}$$

where we used the following representation of the electric field components:

$$E_1 = a_1 e^{i(kz - \omega t + \sigma_1)}, \quad E_2 = a_2 e^{i(kz - \omega t + \sigma_2)}. \tag{11}$$

Here $k = 2\pi/\lambda$, λ is the wavelength, z is the distance along the propagation direction $\vec{e}_3$, $\omega = kc$ is the frequency, c is the speed of light, and t is the time. Note that amplitudes and phases in Eqs. (11) are not constants for real light beams. Thus, Eqs. (3)–(10) should be averaged taking into account many vibrations [9].

As it was mentioned before, the components of the Stokes vector completely define the characteristics of the ellipse of the polarization (ψ, φ). Stokes parameters are related to the values of ψ, φ with the following equations [9].

$$I = a^2, \quad Q = a^2 \cos 2\varphi \cos 2\psi, \quad U = a^2 \cos 2\varphi \sin 2\psi,$$
$$V = a^2 \sin 2\varphi, \tag{12}$$

where $a^2 = a_1^2 + a_2^2$. These formulas provide the geometrical interpretation of Eqs. (7)–(10). It follows from Eqs. (12)

$$\psi = \frac{1}{2}\arctan\frac{U}{Q}, \quad \varphi = \frac{1}{2}\arcsin\left(\frac{V}{IP}\right), \quad P = \frac{\sqrt{Q^2 + U^2 + V^2}}{I}, \tag{13}$$

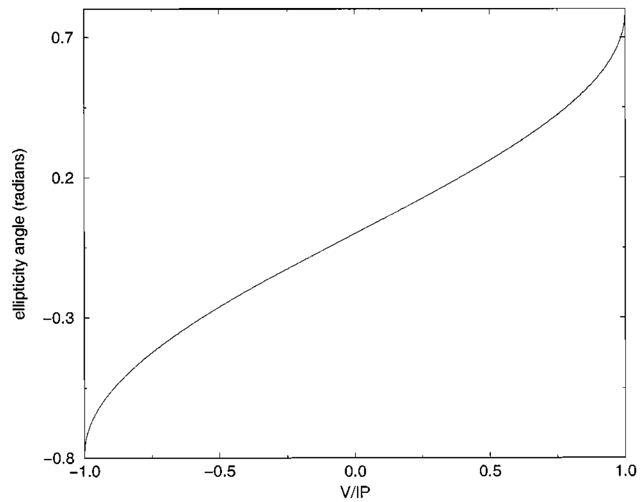

FIG. 1. The dependence of φ on V/IP.

where $\psi \in [0,\pi]$, $\varphi \in [-\pi/4,\pi/4]$, $\mathrm{sgn}(\cos 2\psi) = \mathrm{sgn}(Q)$, and the degree of polarization $P \in [0,1]$. The Stokes parameters (12) describe the completely polarized beam and $I = \sqrt{Q^2 + U^2 + V^2}$ in this case. Equations (13) can be applied for partially polarized beams ($P \neq 1$) as well. The dependencies of angles φ, ψ on the Stokes parameters are presented in Figs. 1 and 2. One can see that $\varphi \approx V/2IP$ at $|V/IP| \leqslant 0.4$, which is often the case. It follows from Fig. 3 that $e \approx |\varphi|$ at $|\varphi| \leqslant 0.2$.

The value of ψ determines the angle between the major axis of a polarization ellipse (maximum intensity component) and the arbitrary direction. Thus, it is coordinate dependent. The ellipticity represents the ratio of small to large axes of the polarization ellipse. This number is coordinate independent. There are two values of ψ, which satisfy Eq. (13) (see Fig. 2). The right value is selected from the condition $\mathrm{sgn}(\cos 2\psi) = \mathrm{sgn}(Q)$. This means that $\psi \in [0,\pi/4[$ or $\psi \in]3\pi/4,\pi]$ *for positive values of* Q and $\psi \in [\pi/4, 3\pi/4]$ (the middle line in Fig. 2) at $Q \leqslant 0$. The azimuth ψ is not defined at $U = Q = 0$, which means that there is not a special preferred oscillation direction in this particular case.

The positive values of φ mean that the polarization is right-handed or the electric vector traces the polarization el-

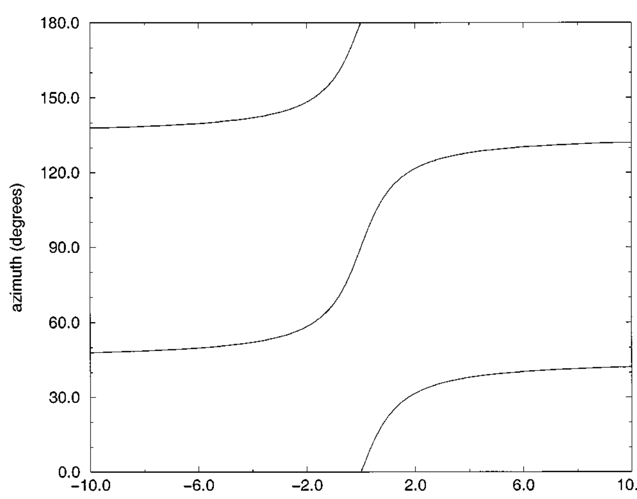

FIG. 2. The dependence of ψ on U/Q.

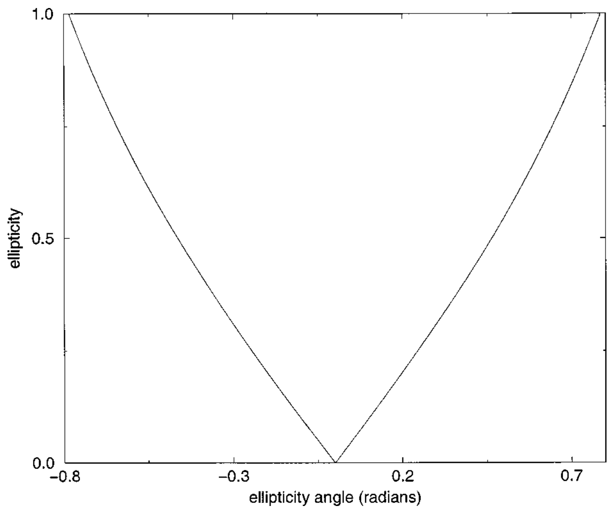

FIG. 3. The dependence of e on φ.

lipse in the clockwise sense when looking in the direction from which the light is coming [3]. It should be pointed out that there is not a unique mathematical definition of right-handed and left-handed polarized waves. Thus, one should be careful in this respect while comparing results of different authors.

Thus, the solution of Eq. (1) allows for the determination of values of a, φ, and ψ and the degree of polarization

$$P=\frac{\sqrt{Q^2+U^2+V^2}}{I}. \tag{14}$$

The information obtained can be used for the stereoochemical analysis of molecules inside small particles. It should be pointed out that Eq. (1) can be applied only for media with distances between particles being much larger than the wavelength and the size of scatterers. This often holds in the optical band of the electromagnetic spectrum.

Let us represent the Stokes vector in Eq. (1) as the sum of two components:

$$\vec{J}(\Omega',z)=\vec{J}_c(\vec{\Omega}',z)\,\delta(\vec{\Omega}'-\vec{\Omega}_0)+\vec{J}_d(\vec{\Omega}',z), \tag{15}$$

where $\vec{J}_c(\vec{\Omega}',z)$ is the Stokes vector of the direct (or coherent) beam, δ is the delta function, $\vec{\Omega}_0$ is the direction of propagation of an incident beam, and $\vec{J}_d(\vec{\Omega}',z)$ is the Stokes vector of the diffused light in the direction $\vec{\Omega}'$ at the depth z. From Eqs. (1) and (15) it follows

$$\mu\frac{d\vec{J}_d(\vec{\Omega},z)}{dz}=-\hat{\sigma}_{\rm ext}\vec{J}_d(\vec{\Omega},z)+\int_{4\pi}\hat{\sigma}_{\rm sca}(\vec{\Omega}'-\vec{\Omega})\vec{J}_d(\vec{\Omega}',z)d\vec{\Omega}'+\hat{\sigma}_{\rm sca}(\vec{\Omega}_0-\vec{\Omega})\vec{J}_c(\vec{\Omega}_0,z) \tag{16}$$

and

$$\mu\frac{d\vec{J}_c(\vec{\Omega}_0,z)}{dz}=-\hat{\sigma}_{\rm ext}\vec{J}_c(\vec{\Omega}_0,z). \tag{17}$$

Equation (17) describes the transformation of the intensity and polarization charateristics of a direct beam. It can be solved analytically [13]:

$$\vec{J}_c(\vec{\Omega}_0,z)=\exp\left\{-\frac{\hat{\sigma}_{\rm ext}z}{\mu}\right\}\vec{J}_c(\vec{\Omega}_0,0), \tag{18}$$

where we assumed that the medium is uniform and $\vec{J}_c(\vec{\Omega}_0,0)$ is the Stokes vector of the incident wave $\vec{J}_0(\vec{\Omega}_0)$ at the top of a layer ($z=0$):

$$\vec{J}_c(\vec{\Omega}_0,0)=\vec{J}_0(\vec{\Omega}_0).$$

The solution of the four integrodifferential equations (16) for the diffused light is more complex. It can be done, e.g., with the doubling method [16]. In the framework of this method one should calculate the radiation characteristics of a very thin layer with thickness z_1 in a single scattering approximation, neglecting the integral in Eq. (16):

$$\mu\frac{d\vec{J}_d(\vec{\Omega},z)}{dz}=-\hat{\sigma}_{\rm ext}\vec{J}_d(\vec{\Omega},z)+\hat{\sigma}_{\rm sca}(\vec{\Omega}_0\to\vec{\Omega})\vec{J}_c(\vec{\Omega}_0,z). \tag{19}$$

The radiation characteristics of the combined layer with thickness $2z_1$ can be found, accounting for the interaction between the second and first layers. Repeating this procedure to the point $z=z_0$, where z_0 is the thickness of a layer, one can solve Eq. (16). Note that the thickness of adding layers can be different in principle.

Thus, it is important to have the analytical solution of Eq. (19) as a starting point for the numerical procedure. It is also of general importance due to the possibility of preparing a thin layer in a laboratory, making the account for the integral term in Eq. (16) unnecessary. The solution of Eq. (19) can be presented in the following formal form:

$$\vec{J}_d^{\downarrow}(z)=\int_0^z e^{-\hat{\sigma}_{\rm ext}z/\mu}\vec{B}(z)\frac{dz}{\mu}, \tag{20}$$

$$\vec{J}_d^{\uparrow}(z)=\int_{z_0}^z e^{-\hat{\sigma}_{\rm ext}z/\mu}\vec{B}(z)\frac{dz}{\mu}, \tag{21}$$

where $\vec{J}_d^{\uparrow}$ and $\vec{J}_d^{\downarrow}$ are Stokes vectors of light fields propagated to the top and bottom of a layer, respectively, $\vec{B}(z)=\hat{\sigma}_{sca}(\vec{\Omega}_0\to\vec{\Omega})\vec{J}_c(\vec{\Omega}_0,z)$, and we used the boundary conditions

$$\vec{J}_d^{\downarrow}(0)=0, \tag{22}$$

$$\vec{J}_d^{\uparrow}(z_0)=0, \tag{23}$$

which state that there is no diffused light field incident on the top [Eq. (22)] and on the bottom [Eq. (23)] of a layer from outer space. Note, that Eqs. (20), (21), and (1) are equivalent, if one includes the integral multiple scattering term in the source function $\vec{B}$ in Eqs. (20) and (21).

Integrals (20) and (21) can be found analytically [13] for some specific source functions $\vec{B}(z)$. However, calculations with Eqs. (18), (20), and (21) are complex in the general case. Thus, our primary task will be a derivation of simplified formulas both for the direct $\vec{J}_c$ [see Eq. (18)] and singly scattered diffused $\vec{J}_d$ [see Eqs. (20) and (21)] components in the special case of disperse media with spherical chiral particles, surrounded by a uniform isotropic symmetric nonabsorbing host medium.

B. Direct light

The general structure of matrices $\hat{\sigma}_{\rm ext}$ and $\hat{\sigma}_{\rm sca}(\vec{\Omega}'-\vec{\Omega})$ for isotropic media with spherical chiral scatters can be presented in the following form [4,10–16].

$$\hat{\sigma}_{\rm ext}=\varepsilon\hat{\varepsilon},\quad \hat{\varepsilon}=\begin{pmatrix}1 & 0 & 0 & \alpha\\ 0 & 1 & \beta & 0\\ 0 & -\beta & 1 & 0\\ \alpha & 0 & 0 & 1\end{pmatrix}, \tag{24}$$

$$\hat{\sigma}_{\rm sca}=\hat{L}(\pi-i_2)\hat{\sigma}^s_{\rm sca}\hat{L}(-i_1),\quad \hat{\sigma}^s_{\rm sca}=\sigma\hat{\sigma},$$

$$\hat{\sigma}=\begin{pmatrix}1 & b_1 & b_3 & b_5\\ b_1 & a_2 & b_4 & b_6\\ -b_3 & -b_4 & a_3 & b_2\\ b_5 & b_6 & -b_2 & a_4\end{pmatrix}. \tag{25}$$

Note that rotation matrices $\hat{L}(-i_1)$ and $\hat{L}(\pi-i_2)$ perform the transformation of the coordinate system related to the scattering plane. The matrix $\hat{\sigma}^s_{\rm sca}$ is defined in the coordinate system, related to a scattering plane. One can find definitions for these matrices and angles i_1 and i_2 in the Appendix. The relationships between elements of matrices (24) and (25) with parameters of particles (their size, complex refractive indices) are presented in the Appendix as well.

First of all, let us consider the solution of four linear differential Eqs. (17) with the extinction matrix (24). Analytical solution of this system can be obtained with standard methods [11,17]:

$$\vec{J}_c(\Omega_0)=\hat{T}\vec{J}_0(\Omega_0), \tag{26}$$

where $\hat{T}=\hat{P}\hat{\Phi}\hat{P}^{-1}$. The matrix $\hat{P}$ is composed of eigenvectors of the extinction matrix $\hat{\varepsilon}$ [Eq. (24)].

$$\vec{\Xi}_1=\begin{pmatrix}-1\\0\\0\\1\end{pmatrix},\quad \vec{\Xi}_2=\begin{pmatrix}1\\0\\0\\1\end{pmatrix},\quad \vec{\Xi}_3=\begin{pmatrix}0\\-i\\1\\0\end{pmatrix},$$

$$\vec{\Xi}_4=\begin{pmatrix}0\\i\\1\\0\end{pmatrix}, \tag{27}$$

namely,

$$\hat{P}=\begin{pmatrix}-1 & 1 & 0 & 0\\ 0 & 0 & -i & i\\ 0 & 0 & 1 & 1\\ 1 & 1 & 0 & 0\end{pmatrix} \tag{28}$$

and

$$\hat{\Phi}=\begin{pmatrix}e^{-\lambda_1 s} & 0 & 0 & 0\\ 0 & e^{-\lambda_2 s} & 0 & 0\\ 0 & 0 & e^{-\lambda_3 s} & 0\\ 0 & 0 & 0 & e^{-\lambda_4 s}\end{pmatrix}. \tag{29}$$

Here, $s=\tau/\xi$, $\tau=\varepsilon z$, $\xi=\cos\vartheta_0$, ϑ_0 is the incidence angle, and $\lambda_1=1-\alpha$, $\lambda_2=1+\alpha$ and $\lambda_3=1+i\beta$, $\lambda_4=1-i\beta$ are eigenvalues of the extinction matrix $\hat{\varepsilon}$ [Eq. (24)]. Note, that it follows for the inverse matrix $\hat{P}^{-1}$ [see Eq. (28)]:

$$\hat{P}^{-1}=\frac{1}{2}\begin{pmatrix}-1 & 0 & 0 & 1\\ 1 & 0 & 0 & 1\\ 0 & i & 1 & 0\\ 0 & -i & 1 & 0\end{pmatrix}. \tag{30}$$

Thus, the transformation matrix $\hat{T}$ in Eq. (26), which coincides with the matricant in Eq. (18), has the following form [see Eqs. (28)–(30)]:

$$\hat{T}=e^{-s}\begin{pmatrix}\cosh\alpha s & 0 & 0 & -\sinh\alpha s\\ 0 & \cos\beta s & -\sin\beta s & 0\\ 0 & \sin\beta s & \cos\beta s & 0\\ -\sinh\alpha s & 0 & 0 & \cosh\alpha s\end{pmatrix}. \tag{31}$$

Solutions (26) and (31) for the direct component $\vec{J}_c(I_c,Q_c,U_c,V_c)$ are much simplier than Eq. (18). They have the following explicit form:

$$I_c=(I_0\cosh\alpha s-V_0\sinh\alpha s)e^{-s}, \tag{32}$$

$$Q_c=(Q_0\cos\beta s-U_0\sin\beta s)e^{-s}, \tag{33}$$

$$U_c=(Q_0\sin\beta s-U_0\cos\beta s)e^{-s}, \tag{34}$$

$$V_c=(-I_0\sinh\alpha s+V_0\cosh\alpha s)e^{-s}. \tag{35}$$

Equations (32)–(35) and Eq. (13) can be used for the investigation of the polarization characteristics of the direct beam under different types of the illumination of a turbid layer.

It is interesting that the transformation of components (Q, U) and (I, V) is independent in the case under investigation. Thus, circularly polarized waves ($Q_0=U_0=0$, $V_0=\pm I_0$) propagate in isotropic chiral media without changing the state of the polarization. For instance, it follows for the left-handed circularly polarized beam,

$$I_0=c,\quad V_0=-c,\quad Q_0=U_0=0, \tag{36}$$

where c is constant. One obtains from Eqs. (32)–(36),

$$I_c=c',\quad V_c=-c',\quad Q_c=U_c=0, \tag{37}$$

where $c=ce^{-\lambda_1 s}$. It follows for the right-handed circularly polarized circular incident wave,

$$I_0=V_0=c,\quad Q_0=U_0=0. \tag{38}$$

Thus, the components of the direct beam will be

$$I_c=V_c=c'',\quad Q_c=U_c=0, \tag{39}$$

where $c''=ce^{-\lambda_2 s}$.

Note that only two eigenvectors in Eqs. (27) have nonzero first elements. They correspond to left-handed and right-handed circularly polarized waves. These waves can be defined as eigenwaves in chiral isotropic media.

Let us consider now the case of the illumination of a layer by a vertically polarized light beam ($\psi=\pi/2$) with Stokes parameters: $I_0=b$, $Q_0=-b$, and $U_0=V_0=0$, where $b=\text{const}$. It follows from Eqs. (32)–(35) in this case,

$$I_c=b\cosh(\alpha s)e^{-s}, \tag{40}$$

$$Q_c=-b\cos(\beta s)e^{-s}, \tag{41}$$

$$U_c=-b\sin(\beta s)e^{-s}, \tag{42}$$

$$V_c=-b\sinh(\alpha s)e^{-s}, \tag{43}$$

and [see Eq. (13)]

$$P=1, \tag{44}$$

$$\psi=\psi_0+\frac{\beta s}{2}, \tag{45}$$

$$\varphi=-0.5\arcsin(\tanh\alpha s), \tag{46}$$

where $\psi_0=\pi/2$. Equations (44)–(46) hold for a horizontally polarized incident beam as well ($I_0=b$, $Q_0=b$, $U_0=V_0=0$, and $\psi_0=0$). Thus, the linearly polarized beam is transformed to the elliptically polarized beam in the case under investigation. The major axes of the polarization ellipse are shifted from the direction of the oscillations of the incident linearly polarized beam. This shift is characterized by the value $\beta s/2$. Note that it follows as $\alpha s\to 0$ from Eq. (46):

$$\varphi=-\frac{\alpha s}{2}. \tag{47}$$

One can see that matrix elements α and β in Eq. (24) are responsible for producing the ellipticity (CD) and the rotation of the polarization plane (ORD) of the direct beam, respectively.

The main interest of the CD and ORD spectroscopy is the spectral dependence of the value $\Delta m=m_L-m_R$, where m_L and m_R are refractive indices for left-handed and right-handed circularly polarized waves, respectively. This difference can be retrieved from measurements of the ORD and CD spectra.

Let us introduce the complex number

$$\Gamma(\lambda)=\beta'(\lambda)-i\alpha'(\lambda),$$

where

$$\alpha'(\lambda)=\alpha\varepsilon=-\frac{2\varphi\cos\vartheta_0}{z_0},\quad \beta'(\lambda)=\beta\varepsilon$$
$$=\frac{2(\psi-\psi_0)\cos\vartheta_0}{z_0}.$$

One can see that the value of Γ can be obtained from measurements of CD and ORD spectra. On the other hand, it follows from results presented in the Appendix for chiral spheres with radii a,

$$\Gamma(\lambda)=\frac{4\pi N}{k^2}\int_0^\infty A_{12}(0)f(a)da, \tag{48}$$

where

$$A_{12}(0)=\sum_{n=1}^{\infty}(2n+1)c_n(a,\lambda,m_L,m_R),$$

$f(a)$ is the particle size distribution, and N is the number concentration of scatterers. Coefficients c_n for uniform spheres are presented in the Appendix. There is a similar expression for layered chiral scatters [7]. Thus, the value of $\Delta m=m_L-m_R$ can be retrieved from the solution of the inverse problem associated with integral equation (48). This solution is simplified for monodispersed particles, media with known particle size distributions, and/or special types of particles when the kernel $A_{12}(0)$ can be represented as a simple analytical function [e.g., it follows for Rayleigh scatters [8]: $A_{12}(0)=(ka)^3\Delta m/(2+m^2)$, $m=2m_Lm_R/(m_L+m_R)$].

C. Diffused light

The intensity of the direct beam reduces considerably with an increase in the optical thickness. The characteristics of the diffused light are of importance in this case. Let us consider the diffused light now. The analytical solution for the value $\vec{J}_d$ in the framework of the single scattering approximation can be obtained from the system of linear nonuniform ordinary differential equations (19).

Let us rewrite Eq. (19) in the following form:

$$\dot{\vec{X}}=-\hat{\varepsilon}\vec{X}+\vec{W}, \tag{49}$$

where

$$\dot{\vec{X}}=\frac{d\vec{J}_d}{ds},\quad \vec{X}=\vec{J}_d,\quad \hat{\varepsilon}=\hat{\sigma}_{\text{ext}}/\varepsilon,\quad s=\varepsilon z/\mu,$$

$$\vec{W}=\gamma\hat{\sigma}_s\vec{J}_c,\quad \hat{\sigma}_s=\hat{\sigma}_{\text{sca}}/\sigma,\quad \gamma=\frac{w_0p(\theta)}{4\pi}. \tag{50}$$

We introduced the single scattering albedo

$$w_0=\sigma_{\text{sca}}/\varepsilon, \tag{51}$$

and the phase function

$$p(\theta)=\frac{4\pi\sigma}{\sigma_{\text{sca}}}, \tag{52}$$

normalized by the following condition:

$$\frac{1}{2}\int_0^\pi p(\theta)\sin\theta d\theta=1, \tag{53}$$

which follows from the definition of the scattering coefficient,

$$\sigma_{\text{sca}}=2\pi\int_0^{\pi}\sigma(\theta)\sin\theta d\theta. \tag{54}$$

Here, θ is the scattering angle.

Let us introduce a new vector

$$\vec{Y}=\vec{P}^{-1}\vec{X}. \tag{55}$$

It follows from Eqs. (49) and (55),

$$\hat{P}\dot{\vec{Y}}=-\hat{\varepsilon}\hat{P}\vec{Y}+\vec{W} \tag{56}$$

or

$$\dot{\vec{Y}}=-\hat{\Lambda}\vec{Y}+\hat{P}^{-1}\vec{W}, \tag{57}$$

where

$$\hat{\Lambda}=\begin{pmatrix}\lambda_1 & 0 & 0 & 0\\ 0 & \lambda_2 & 0 & 0\\ 0 & 0 & \lambda_3 & 0\\ 0 & 0 & 0 & \lambda_4\end{pmatrix} \tag{58}$$

and $\dot{\vec{Y}}=d\vec{Y}/ds$.

One can see that we have four decoupled equations [see Eqs. (57)] instead of more complex system of Eqs. (49) now. These equations can be solved with familiar techniques [17]:

$$Y_i^{\downarrow}=e^{-\lambda_i s}\int_0^s e^{\lambda_i \nu}f_i(\nu)d\nu, \tag{59}$$

$$Y_i^{\uparrow}=e^{-\lambda_i s}\int_{s_0}^s e^{\lambda_i \nu}f_i(\nu)d\nu, \tag{60}$$

where we accounted for boundary conditions (22) and (23) and

$$s_0=\varepsilon z_0/\mu, \tag{61}$$

$$\vec{f}(\nu)=\hat{P}^{-1}\gamma\hat{\sigma}_s\hat{P}\hat{\Phi}\hat{P}^{-1}\vec{J}_0. \tag{62}$$

It follows after substitution of Eqs. (61) and (62) into Eqs. (59) and (60) that

$$\vec{Y}^{\uparrow\downarrow}=\hat{P}^{-1}\gamma\xi\hat{\sigma}_s\hat{B}^{\uparrow\downarrow}\vec{J}_0, \tag{63}$$

where

$$\hat{B}^{\uparrow\downarrow}=\hat{P}\hat{F}^{\uparrow\downarrow}\hat{P}^{-1}. \tag{64}$$

The nonzero elements of the diagonalized matrices $\hat{F}^{\uparrow\downarrow}$ have the following analytical forms:

$$\hat{F}_{ii}^{\uparrow}=\frac{e^{-\lambda_i\tau/\xi}-e^{-\lambda_i[(1/\xi+1/\eta)\tau_0-\tau/\eta]}}{\lambda_i(\eta+\xi)}, \tag{65}$$

$$\hat{F}_{ii}^{\downarrow}=\frac{e^{-\lambda_i\tau/\eta}-e^{-\lambda_i\tau/\xi}}{\lambda_i(\eta-\xi)}, \tag{66}$$

where $\tau=\varepsilon z_0$ and $\eta=|\mu|$. It follows from Eq. (66) at $\eta\to\xi$ that

$$\hat{F}_{ii}^{\downarrow}=\frac{\tau e^{-\lambda_i\tau/\xi}}{\xi^2}. \tag{67}$$

Thus, one obtains for the diffused intensity [see Eqs. (55), (59) and (60)],

$$\vec{J}_d^{\uparrow\downarrow}=M^{\uparrow\downarrow}\hat{N}^{\uparrow\downarrow}\vec{J}_0, \tag{68}$$

where

$$M^{\uparrow}=\frac{\gamma\xi}{\eta+\xi},\quad M^{\downarrow}=\frac{\gamma\xi}{\eta-\xi},\quad \hat{N}^{\uparrow\downarrow}=\hat{\sigma}_s\hat{B}^{\uparrow\downarrow}, \tag{69}$$

$$\hat{B}^{\uparrow\downarrow}=\begin{pmatrix}b_{11} & 0 & 0 & b_{14}\\ 0 & b_{22} & b_{23} & 0\\ 0 & -b_{23} & b_{22} & 0\\ b_{14} & 0 & 0 & b_{11}\end{pmatrix}, \tag{70}$$

$$b_{11}=\frac{[\cosh(\alpha p)+\alpha\sinh(\alpha p)]e^{-p}-[\cosh(\alpha q)+\alpha\sinh(\alpha q)]e^{-q}}{1-\alpha^2}, \tag{71}$$

$$b_{14}=\frac{[\sinh(\alpha q)+\alpha\cosh(\alpha q)]e^{-q}-[\sinh(\alpha p)+\alpha\cosh(\alpha p)]e^{-p}}{1-\alpha^2}, \tag{72}$$

$$b_{22}=\frac{[\cos(\beta p)-\beta\sin(\beta p)]e^{-p}-[\cos(\beta q)-\beta\sin(\beta q)]e^{-q}}{1+\beta^2}, \tag{73}$$

$$b_{23}=\frac{[\sin(\beta q)+\beta\cos(\beta q)]e^{-q}-[\sin(\beta p)+\beta\cos(\beta p)]e^{-p}}{1+\beta^2}, \tag{74}$$

$p=\tau/\xi$, $q=\tau_0(1/\eta+1/\xi)-\tau/\eta$ for the upward light ($\hat{B}^{\uparrow}$) and $p=\tau/\eta$, $q=\tau/\xi$ for the downward light ($\hat{B}^{\downarrow}$). At $\xi=\eta$ it follows for the transmitted light [see Eqs. (63), (67), and (30)] that

$$M^{\downarrow}=\gamma\tau\exp(-\tau/\xi),\quad b_{11}=\cosh(\alpha p),$$

$$b_{22}=\cos(\beta p),\quad b_{14}=-\sinh(\alpha p),\quad b_{23}=-\sin(\beta p).$$

Equations (32)–(35) and Eqs. (68)–(74) are much more useful for applications than general solutions (18), (20) and (21). One should just multiply the Stokes vector of the incident light by the matrix $\hat{N}$ and scalar M to find the intensity and polarization characteristics of a diffused light field inside optically thin chiral diperse media. The characteristics of the direct beam can be found from Eqs. (26) and (31) or Eqs. (32)–(35).

Note that it follows for the transmitted diffused light at a bottom of a layer ($\tau=\tau_0$): $p=\tau_0/\eta$, $q=\tau_0/\xi$. General equations (71)–(74) do not simplify in this particular case. However, they do simplify for the reflected light [$\tau=0$, $p=0$, and $q=\tau_0(1/\eta+1/\xi)$]:

$$b_{11}=\frac{1-[\cosh(\alpha q)+\alpha\sinh(\alpha p)]e^{-q}}{1-\alpha^2},\tag{75}$$

$$b_{14}=\frac{[\sinh(\alpha q)+\alpha\cosh(\alpha q)]e^{-q}-\alpha}{1-\alpha^2},\tag{76}$$

$$b_{22}=\frac{1-[\cos(\beta q)-\beta\sin(\beta q)]e^{-q}}{1+\beta^2},\tag{77}$$

$$b_{23}=\frac{[\sin(\beta q)+\beta\cos(\beta q)]e^{-q}-\beta}{1+\beta^2}.\tag{78}$$

Let us check Eq. (68) for the special case when the extinction matrix is reduced to a scalar value. It follows from Eqs. (71) and (74) in this case that

$$b_{11}=b_{22},\quad b_{14}=b_{23}=0\tag{79}$$

and [see Eq. (68)]

$$\vec{J}_d^{\uparrow\downarrow}=b_{11}M^{\uparrow\downarrow}\hat{\sigma}_s\vec{J}_0,\tag{80}$$

where $b_{11}=\exp(-p)-\exp(-q)$. This formula coincides with well-known equation [18], derived for isotropic symmetric media (e.g., water clouds).

One obtains from Eqs. (71)–(74) at the small optical depth

$$b_{11}=b_{22}=q-p,\quad b_{14}=b_{23}=0.\tag{81}$$

Thus, the diffused intensity does not depend on the extinction matrix in this case. It is determined only by the differential scattering matrix as it should be.

III. CONCLUSION

The mirror symmetry is broken in living things. Proteins are constructed only from ''left-handed'' amino acids, whereas nucleic acids (DNA or RNA) contain only ''right-handed'' sugars. Thus, most of biological media are asymmetric.

Light beams can be used for monitoring bioparticles during their lifecycles. Polarization characteristics of transmitted and reflected light are of special value. The power of chirooptical methods was well demonstrated for uniform media [2]. However, application of same schemes for particulate media of a biological origin is not widespread due to complexities related to the accounting for single and multiple scattering of photons in such media.

This paper presents a system of analytical formulas, which can be used in studies of light interaction with particulate optically active media. The presentation is based on the vector radiative transfer theory. Simple analytical solutions for the polarization characteristics of direct and diffused light (in the single scattering approximation) are presented.

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APPENDIX: EXTINCTION AND SCATTERING MATRICES

Extinction and scattering matrices in radiative transport equation (1) specify the local optical properties of a scattering medium. They depend on the wavelength, size of particles, and their refractive indices. One can calculate these matrices with the following system of equations in the case of optically active spheres [3,4]:

$$\hat{\sigma}_{\text{ext}}=\varepsilon\begin{pmatrix}1&0&0&\alpha\\0&1&\beta&0\\0&-\beta&1&0\\\alpha&0&0&0\end{pmatrix},$$

$$\hat{\sigma}_{\text{sca}}=\hat{L}(\pi-i_2)\hat{\sigma}^s_{\text{sca}}(\theta)\hat{L}(-i_1),$$

where

$$L(-i_1)=\begin{pmatrix}1&0&0&0\\0&\cos 2i_1&-\sin 2i_1&0\\0&\sin 2i_1&\cos 2i_1&0\\0&0&0&1\end{pmatrix},\quad L(\pi-i_2)=\begin{pmatrix}1&0&0&0\\0&\cos 2i_2&-\sin 2i_2&0\\0&\sin 2i_2&\cos 2i_2&0\\0&0&0&1\end{pmatrix},$$

$$\cos 2i_j=2\cos^2 i_j-1,\quad \sin 2i_j=2\sqrt{1-\cos^2 i_j}\cos i_j,$$

$$\cos i_1 = \frac{-\mu + \mu \cos\theta}{s\sqrt{(1-\cos^2\theta)(1-\mu'^2)}}, \quad \cos i_2 = \frac{-\mu' + \mu\cos\theta}{s\sqrt{(1-\cos^2\theta)(1-\mu^2)}},$$

$$s = \mathrm{sgn}(\phi - \phi' - \pi), \quad j = 1,2, \quad \cos\theta = \mu\mu' + \sqrt{(1-\mu^2)(1-\mu'^2)}\cos(\phi - \phi'), \quad \mu = \cos\vartheta, \quad \mu' = \cos\vartheta,$$

$$\varepsilon = \frac{4\pi}{k^2}\mathrm{Re}[S_{11}(0)], \quad \alpha = -\frac{\mathrm{Im}[A_{12}(0)]}{\mathrm{Re}[A_{11}(0)]}, \quad \beta = \frac{\mathrm{Re}[A_{12}(0)]}{\mathrm{Re}[A_{11}(0)]},$$

$$\hat{\sigma}^s_{\mathrm{sca}} = k^{-2}\begin{pmatrix} \frac{|A_{11}|^2+|A_{22}|^2}{2}+|A_{12}|^2 & \frac{|A_{11}|^2-|A_{22}|^2}{2} & \mathrm{Re}[(A_{11}-A_{22})A_{12}^*] & \mathrm{Im}[(A_{11}+A_{22})A_{12}^*] \\ \frac{|A_{11}|^2-|A_{22}|^2}{2} & \frac{|A_{11}|^2+|A_{22}|^2}{2}-|A_{12}|^2 & \mathrm{Re}[(A_{11}+A_{22})A_{12}^*] & \mathrm{Im}[(A_{11}-A_{22})A_{12}^*] \\ -\mathrm{Re}[(A_{11}-A_{22})A_{12}^*] & -\mathrm{Re}[(A_{11}+A_{22})A_{12}^*] & \mathrm{Re}(A_{11}^*A_{22})-|A_{12}|^2 & \mathrm{Im}(A_{11}A_{22}^*) \\ \mathrm{Im}[(A_{11}+A_{22})A_{12}^*] & \mathrm{Im}[(A_{11}-A_{22})A_{12}^*] & -\mathrm{Im}(A_{11}A_{22}^*) & \mathrm{Re}(A_{11}^*A_{22})+|A_{12}|^2 \end{pmatrix}$$

$$A_{11}(\theta) = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\{a_n\tau_n(\cos\theta)+b_n\pi_n(\cos\theta)\},$$

$$A_{22}(\theta) = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}\{a_n\pi_n(\cos\theta)+b_n\tau_n(\cos\theta)\},$$

$$A_{12}(\theta) = \sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}c_n(\pi_n+\tau_n),$$

$$\pi_n(\cos\theta) = \frac{P_n^{(1)}(\cos\theta)}{\sin\theta}, \quad \tau_n(\cos\theta) = \frac{dP_n^{(1)}(\cos\theta)}{d\theta},$$

$$a_n = \frac{V_n(R)A_n(L)+V_n(L)A_n(R)}{W_n(L)V_n(R)+V_n(L)W_n(R)}, \quad b_n = \frac{W_n(L)B_n(R)+W_n(R)B_n(L)}{W_n(L)V_n(R)+V_n(L)W_n(R)}, \quad c_n = i\frac{W_n(R)A_n(L)-W_n(L)A_n(R)}{W_n(L)V_n(R)+V_n(L)W_n(R)},$$

$$W_n(J) = m\psi_n(m_J x)\xi_n'(x) - \xi_n(x)\psi_n'(m_J x), \quad V_n(J) = \psi_n(m_J x)\xi_n'(x) - m\xi_n(x)\psi_n'(m_J x),$$

$$A_n(J) = m\psi_n(m_J x)\psi_n'(x) - \psi_n(x)\psi_n'(m_J x), \quad B_n(J) = \psi_n(m_J x)\psi_n'(x) - m\psi_n(x)\psi_n'(m_J x),$$

Here,

$$\psi_n(x) = \sqrt{(\pi x/2)}J_{n+1/2}(x),$$

$$\xi_n(x) = \sqrt{(\pi x/2)}H_{n+1/2}(x),$$

$J_{n+1/2}$ and $H_{n+1/2}$ are Bessel and Hankel functions, and $P_n^{(1)}(\cos\theta)$ is the associated Legendre polynomial. Vectors $\vec{\Omega}(\Theta,\phi)$ and $\vec{\Omega}'(\Theta',\phi')$ define the observation and propagation directions [see Eq. (1)]. Note that it follows $\pi_n(0) = \tau_n(0) = n(n+1)/2$. Values of J are equal to L or R, $m_L = N_L/\bar{n}$, $m_R = N_R/\bar{n}$, $m = m_L m_R/\bar{m}$, and $\bar{m} = (m_L+m_R)/2$, $\bar{n}$ is the refractive index of a host medium, N_L and N_R are refractive indices of particles for left-handed and right-handed polarized waves, and $x = 2\pi a\bar{n}/\lambda$ is the size parameter. Note that it is supposed that the magnetic permittivity of particles and a host medium is the same.

It follows for polydispersed media with the particle size distribution $f(a)$ that

$$\overline{\varepsilon} = N\int_0^\infty \varepsilon(a)f(a)da, \quad \overline{\varepsilon\alpha} = N\int_0^\infty \varepsilon(a)\alpha(a)f(a)da,$$

$$\overline{\varepsilon\beta} = N\int_0^\infty \varepsilon(a)\beta(a)f(a)da,$$

$$\overline{\zeta} = N\int_0^\infty \zeta(a)f(a)da,$$

where values of ζ represent elements of the differential scattering matrix. The number concentration of particles N is related to the volumetric concentration C_v by the following formula:

$$N = \frac{C_v}{\frac{4\pi}{3}\int_0^\infty a^3 f(a)da}.$$

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