Depolarization of circularly polarized light in the Mie resonance region

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Abstract. We show that a disordered ensemble of dielectric particles near the Mie resonances has anomalous depolarizing properties. Under the first Kerker condition the depolarization length of circularly polarized light reaches its peak value, and can be ten times greater than the transport mean free path. The second Kerker condition is shown to be satisfied as the refractive index of particles increases. In this case, the depolarization length is minimum and almost coincides with the mean free path.

1. Introduction
The effect of slow decay of circular polarization is inherent in wave propagation through media with large scattering inhomogeneities [1] (see also [2] and references therein) and, as has been shown recently [3], through media with resonant Mie particles. The latter case is considered below.

Between the first two Mie resonances a single scattered wave can retain its circular polarization [3]. This effect arises provided that the electric dipolar contribution to the scattering amplitude becomes equal to the magnetic dipolar one (e.g., for silicon spheres, refractive index $n = 3.5$, the circular polarization memory can be observed at size parameter $k_0a = 0.784$, $k_0$ and $a$ are the wavenumber and the particle radius). The equality between the electric and magnetic dipolar contributions corresponds to the so-called first Kerker condition [3] under which the differential scattering cross-section is equal to zero in the backward direction. Numerical simulations [3] have indicated that multiply scattered light remains also completely polarized. However, as shown below, the results [3] can be considered as a first approximation. The inclusion of the quadrupolar and higher order contributions violates the exact fulfillment of the Kerker condition. The depolarization cross section proves to be small, but nonzero, and the circular polarization decays at scales that are more than ten times greater than the transport mean free path.

2. General relations
Consider a beam of polarized light incident on the surface of a scattering medium. The medium is assumed to be a statistically isotropic disordered ensemble of particles. The polarization state of scattered light can be described by the Stokes vector $\hat{S} = (I, Q, U, V)$ which obeys the vector radiative transfer equation [4],

$$\left\{ \mathbf{n} \frac{\partial}{\partial r} + n_0\sigma_{tot} \right\} \hat{S}(z, \mathbf{n}) = n_0 \int d\mathbf{n}' \hat{Z}(\mathbf{n}, \mathbf{n}') \hat{S}(z, \mathbf{n}') \tag{1}$$
where $\sigma_{\text{tot}} = \sigma + \sigma_a$ is the cross section of total extinction, $\sigma$ and $\sigma_a$ are the the cross sections of scattering and absorption, respectively; vectors $\mathbf{n}$ and $\mathbf{n}'$ denote the directions of wave propagation, $n_0$ is the number of scattering particles per unit volume. The phase matrix $\hat{Z}(\mathbf{n}, \mathbf{n}')$ entering into Eq.(1) can be expressed in terms of the scattering matrix (see [4]),

$$
\hat{Z}(\mathbf{n}, \mathbf{n}') = \hat{L}(\pi - \beta) \hat{F}(\mathbf{nn}') \hat{L}(-\beta')
$$

(2)

The scattering matrix $\hat{F}(\mathbf{nn}')$ describes the intrinsic properties of the medium. The matrices $\hat{L}(-\beta')$ and $\hat{L}(\pi - \beta)$ describe the transformation of the Stokes parameters under rotations in the space of directions $\mathbf{n}$ (regarding the definition of the matrix $\hat{L}$ and angles $\beta, \beta'$, see [4]).

For a macroscopically isotropic and symmetric medium, the scattering matrix $\hat{F}(\mathbf{nn}')$ appearing in Eq.(2) has the block-diagonal structure [4]:

$$
\hat{F}(\mathbf{nn}') = \begin{pmatrix}
    a_1(\mathbf{nn}') & b_1(\mathbf{nn}') & 0 & 0 \\
    b_1(\mathbf{nn}') & a_1(\mathbf{nn}') & 0 & 0 \\
    0 & 0 & a_2(\mathbf{nn}') & b_2(\mathbf{nn}') \\
    0 & 0 & -b_2(\mathbf{nn}') & a_2(\mathbf{nn}')
\end{pmatrix}
$$

(3)

The element $a_1$ in matrix (3) is the differential cross section of elastic scattering. It is subject to the condition

$$
\int d\mathbf{n}' a_1(\mathbf{nn}') = \sigma
$$

(4)

For spherical particles of given radius and refractive index, the matrix elements $a_1$, $a_2$ and $b_1$, $b_2$ are equal to

$$
a_1(\cos \gamma) = \frac{1}{2} \left( |A_{||}(\cos \gamma)|^2 + |A_{\perp}(\cos \gamma)|^2 \right), \\
b_1(\cos \gamma) = \frac{1}{2} \left( |A_{||}(\cos \gamma)|^2 - |A_{\perp}(\cos \gamma)|^2 \right),
$$

$$
a_2(\cos \gamma) = \text{Re} A_{||}(\cos \gamma) A_{\perp}^*(\cos \gamma), \\
b_2(\cos \gamma) = \text{Im} A_{||}(\cos \gamma) A_{\perp}^*(\cos \gamma)
$$

(5)

where $\cos \gamma = \mathbf{nn}'$, $A_{||}$ and $A_{\perp}$ are the amplitudes of the components polarized parallel and perpendicular to the scattering plane. The values of $A_{||}$ and $A_{\perp}$ are calculated with the Mie theory [5].

In the case of the normal incidence of the circularly polarized light the Stokes parameters $I, Q, U$ and $V$ obey two independent systems of transfer equations

$$
\begin{align*}
\{ & \frac{\partial}{\partial z} + n_0 \sigma_{\text{tot}} \} \\
I(z, \mu) & = n_0 \int d\mathbf{n}' \begin{pmatrix} a_1 \\ b_1 \cos \beta' \\ a_1 \cos \beta \cos \beta' - a_2 \sin \beta \sin \beta' \end{pmatrix} \begin{pmatrix} I(z, \mu') \\ Q(z, \mu') \\ V(z, \mu') \end{pmatrix}, \\
Q(z, \mu) & = n_0 \int d\mathbf{n}' \begin{pmatrix} a_2 \\ b_2 \cos \beta' \\ a_2 \cos \beta \cos \beta' - a_2 \sin \beta \sin \beta' \end{pmatrix} \begin{pmatrix} I(z, \mu') \\ Q(z, \mu') \\ V(z, \mu') \end{pmatrix},
\end{align*}
$$

(6)

$$
\begin{align*}
\{ & \frac{\partial}{\partial z} + n_0 \sigma_{\text{tot}} \} \\
V(z, \mu) & = n_0 \int d\mathbf{n}' \begin{pmatrix} a_2 \\ b_2 \cos \beta' \\ a_2 \cos \beta \cos \beta' - a_2 \sin \beta \sin \beta' \end{pmatrix} \begin{pmatrix} I(z, \mu') \\ Q(z, \mu') \\ V(z, \mu') \end{pmatrix},
\end{align*}
$$

(7)

where $\mu = \mathbf{nn}_n$, $\mathbf{n}_n$ is the inward normal to the surface, $\psi = \varphi - \varphi'$ is the difference between the azimuthal angles of vectors $\mathbf{n}$ and $\mathbf{n}'$. The degree of circular polarization of light is defined as $P_C = V/I$.

For a unit incident flux, the boundary conditions to Eqs.(6) and (7) have the form

$$
I(z = 0, \mu) = V(z = 0, \mu) = \frac{1}{2\pi} \delta(1 - \mu)
$$

(8)

The other quantities, $Q$ and $U$, should be put equal to zero at $z = 0$. 

Figure 1. Angular dependence of the elements $a_1$ (solid black line), $|a_2|$ (dashed red line), $|b_1|$ (dash-dotted green line) and $|b_2|$ (dotted blue line) at the first Kerker point. The numerical calculations were carried out with the Mie theory for a silicon sphere ($n = 3.5, k_0a = 0.784$).

3. First Kerker point. Basic mode approximation

Between the first two Mie resonances the differential scattering cross section can tend to zero in the backward direction [6]. This corresponds to the so-called first Kerker condition [6, 7] and occurs when the electric and magnetic dipolar contributions to the scattering amplitude coincide to each other.

Under the first Kerker condition the elements $a_1$ and $a_2$ appearing in Eqs.(3) and (5) prove to be very nearly equal to each other and vastly greater than the elements $b_1$ and $b_2$ (see Fig. 1). Therefore, in the vicinity of the first Kerker point we can neglect the off-diagonal elements of the scattering matrix, and the systems (6) and (7) reduce to two independent equations [2]. Within such an approximation, the specific intensity $I$ and the fourth Stokes parameter $V$ are subject to the transfer equations [2]

$$\begin{align*}
\left\{ \mu \frac{\partial}{\partial z} + n_0\sigma_{\text{tot}} \right\} I(z, \mu) &= n_0 \int d\mu' a_1(\mu\mu') I(z, \mu') \\
\left\{ \mu \frac{\partial}{\partial z} + n_0\sigma_{\text{tot}} \right\} V(z, \mu) &= n_0 \int d\mu' a_2(\mu\mu') V(z, \mu')
\end{align*}$$

Equations (9) and (10) correspond to the basic mode approximation [2] in the vector radiative transfer equation. The difference between the elements $a_1$ and $a_2$ is responsible for attenuation of ratio $V/I$ due to depolarization of circularly polarized light in the medium. The cross-section of depolarization [2, 9]

$$\sigma_{\text{dep}} = \int d\mu' (a_1(\mu\mu') - a_2(\mu\mu')) = \frac{1}{2} \int d\mu' |A_{\parallel}(\mu\mu') - A_{\perp}(\mu\mu')|^2$$

acts as the cross section of an "additional absorption" in Eq.(10). The elements $a_1$ and $a_2$ differ from each other only in the vicinity of the backward direction (see Fig. 1).

If only the electric and magnetic dipolar contributions to the scattering amplitudes are taken into account, the amplitudes $A_{\parallel}$ and $A_{\perp}$ and, consequently, the elements $a_1$ and $a_2$ can be expressed in terms of the electric $\alpha_e$ and magnetic $\alpha_m$ particle polarizabilities and written in the form

$$A_{\parallel} = k_0^2 (\alpha_e \cos \gamma + \alpha_m), \quad A_{\perp} = k_0^2 (\alpha_e + \alpha_m \cos \gamma)$$

(12)
depolarization length $l_{circ}$ for an ensemble of silicon spheres as a function of size parameter $k_0a$. The upper curve is the result of numerical calculations with the characteristic transfer equation. The lower curve is the diffusion result (15). The inset shows the peak value of $l_{circ}/l_{tr}$ as a function of the refractive index of scattering particles.

\[
\alpha_{1,2} = \frac{k_0^4}{2} \left( |\alpha_e| + |\alpha_m| \right)^2 (1 + \cos \gamma)^2 \pm |\alpha_e - \alpha_m|^2 (1 - \cos \gamma)^2
\]

From Eqs.(12) and (13) it follows that under the first Kerker condition, $\alpha_e = \alpha_m$ [6], the equalities $A_\parallel = A_\perp$ and $a_1 = a_2$ are valid. In this approximation the cross section of depolarization $\sigma_{dep} = 0$, and the circular polarization is retained at an arbitrary depth $z$ [3].

The difference between $a_1$ and $a_2$ shown in Fig. 1 is due to the higher-order multipolar contributions to the scattering amplitudes. The cross section $\sigma_{dep}$ is small but nonzero. The wavelength dependence of $\sigma_{dep}$ is illustrated in Fig. 2. The numerical calculations were carried out with the Mie theory [5]. A sharp dip in $\sigma_{dep}$ corresponds to the first Kerker point. In the vicinity of this point, the value of $\sigma_{dep}$ is much less than the transport cross-section $\sigma_{tr}$, resulting in the effect of circular polarization memory.

As $z$ increases, the fourth Stokes parameter decays as $V(z, \mu) \sim \exp(-z/l_{circ})$, where the depolarization length $l_{circ}$ can be found from the characteristic equation corresponding to Eq.(10) (for details see [2]). For a medium with no absorption ($\sigma_a = 0$) this equation has the form

\[
\det \left( n_0 (\sigma - a_2(l)) \delta_{l,m} - \frac{1}{(2l + 1)l_{circ}} (l\delta_{l-1,m} + (l + 1)\delta_{l+1,m}) \right) = 0
\]

where $a_2(l)$ ($l = 0, 1, \ldots$) are the expansion coefficients of the element $a_2(\cos \gamma)$ in the Legendre polynomials.

For silicon spheres, the results of numerical calculations of $l_{circ}$ in the vicinity of the first Kerker point are shown in Fig. 3. For a number of materials with high refractive index [8], the values of $l_{circ}$ are also presented in Table 1. From the obtained results it follows that ratio $l_{circ}/l_{tr}$ at the first Kerker point increases with the refractive index of particles. For different materials, the position of the first Kerker point is well approximated by the relation $k_0an = 2.744$. 

Figure 2. Depolarization cross section $\sigma_{dep}$ for silicon spheres as a function of size parameter $k_0a$. The inset illustrates the smooth behavior of $\sigma_{tr}$ near of the first Kerker point.

Figure 3. Depolarization length $l_{circ}$ for an ensemble of silicon spheres as a function of size parameter $k_0a$.
Table 1. Depolarization length at the first Kerker point for high refractive index materials

| Material | Refractive index $n$ ($\lambda = 1.6 \ \mu m$) [8] | $k_0n$ | $\sigma_{dep}/\sigma_{tr}$ | $l_{circ}/l_{tr}$ |
|----------|-----------------------------------------------|--------|----------------|-----------------|
| AlAs     | 2.9                                           | 2.743  | 0.0016         | 14.6            |
| GaP      | 3.05                                          | 2.745  | 0.0012         | 16.5            |
| InP      | 3.15                                          | 2.744  | 0.0010         | 17.9            |
| AlSb     | 3.28                                          | 2.745  | 0.0009         | 19.3            |
| GaAs     | 3.37                                          | 2.743  | 0.0008         | 20.8            |
| Si       | 3.47                                          | 2.745  | 0.0007         | 22.0            |
| Ge       | 4.24                                          | 2.743  | 0.0003         | 32.6            |

Under conditions of the circular polarization memory, $\sigma_{dep} \ll \sigma_{tr}$, the fourth Stokes parameters $V$ falls off at scales that are much greater than the transport mean free path $l_{tr}$, and the diffusion approximation can be applied to calculations of $V(z, \mu)$ [9]. Within such an approximation, the depolarization length $l_{circ}$ of circularly polarized light is equal to [2,9]

$$l_{circ} = \sqrt{l_{tr}l_{dep}/3} \quad (15)$$

where $l_{dep} = (n_0\sigma_{dep})^{-1}$ is the mean free path with respect to depolarizing collisions. Comparison of Eq.(15) with the results of numerical calculations is illustrated in Fig. 3. As follows from Fig. 3, the diffusion formula (15) is valid wherever the inequality $l_{circ} > l_{tr}$ is fulfilled.

In practice, spread of particles in size reduces the peak value of $l_{circ}/l_{tr}$. However, for relatively small deviations from the Kerker condition, the circular polarization memory effect remains observable.

4. Second Kerker point

The second Kerker condition implies that the scattering cross section tends to zero in the forward direction [6]. Owing to the optical theorem, this condition can be fulfilled only approximately.

With allowance for the electric and magnetic dipolar contributions, the optical response of the Mie particles was studied in [6,10]. Within such an approximation, the scattering amplitudes $A_\parallel$ and $A_\perp$ at the second Kerker point, $\alpha_e = -\alpha_m$ [6], satisfy the equality $A_\parallel = -A_\perp$. In this case $a_1 = -a_2$ and the cross section $a_1$ is proportional to $(1 - \cos \gamma)^2$ (see Eq.(13)). As a result, the scattering to the backward hemisphere dominates, and the mean cosine of single-scattering angle reaches its minimum value $<\cos \gamma> = -0.5$. The cross section of depolarization $\sigma_{dep}$ peaks at the second Kerker point. The maximum value of $\sigma_{dep}$ is equal to $2\sigma$.

If, in addition to the electric and magnetic dipolar contributions, the higher-order multipolar contributions are taken into account, the second Kerker condition can not be satisfied exactly. As follows from our calculations, the suppression of the forward scattering turns out to be slightly pronounced for relatively moderate values of the refractive index. However, the fulfillment of the second Kerker condition is achieved asymptotically as the refractive index increases, $n \gg 1$ (see Fig. 4). The values of $\sigma_{dep}$ and $<\cos \gamma>$ tend to their limiting values at $n \geq 5$ (see Fig. 5).

Owing to rather great values of $\sigma_{dep}$, multiple scattering near the second Kerker point is accompanied by fast depolarization of light. The results of numerical calculations of $l_{circ}$ with the characteristic equation (14) are presented in Fig. 5. As the refractive index increases, the depolarization length tends to the value close to the mean free path, $l_{circ} = 1.038l$. This value corresponds to the limit $n \gg 1$ where $a_1(\cos \gamma) = a_1(180^0)(1 - \cos \gamma)^2/4$. 


From the obtained results it follows that an ensemble of Mie particles near the second Kerker point can be considered as the medium that maximally scatters and depolarizes the electromagnetic radiation.

5. Conclusions
In conclusion, we have shown that a disordered ensemble of dielectric spheres with a relatively high refractive index exhibits anomalous depolarizing properties in the Mie resonance region. For the first and second Kerker points, the depolarization lengths of circularly polarized light have been first calculated. The decay of polarization in the vicinity of the first Kerker point has been shown to occur deep in the diffusion regime. It has also been found that the second Kerker condition can be satisfied as the refractive index of particles increases. In this case, the depolarizing ability of the medium is maximum. The depolarization length of circularly polarized light is very close to the mean free path.

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