

## State of Polarization of Light Scattered by Colored Spheroids

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Using the light-scattering theory of Rayleigh-Gans for spheroids, theoretical expressions were derived for the polarization state of light scattered by randomly oriented small and colored spheroidal particles in the case of linearly polarized incident light with an angle of inclination of  $\psi$  to the scattering plane. The azimuth angle  $\zeta$  of the ellipse of the scattered light did not depend on the axial ratio  $p$  and the relative complex refractive index  $m$  ( $=m_0 - ik_0$ ), but was a function of only the scattering angle  $\theta$  and  $\psi$ ; also, the ellipticity of the ellipse of the scattered light was always equal to 0 and the scattered light was partially linearly polarized for linearly polarized incident light. The degree of polarization of the scattered light for spheroidal particles,  $P$ , defined as the ratio of the polarized component to the intensity of the scattered light, depended strongly on  $p$ ,  $m_0$ ,  $k_0$ , and  $\theta$  together with the angle  $\psi$ . This  $P$  was found to be smaller than 1. In the case of spherical particles,  $P$  was always equal to 1 at any  $\theta$ ; this means that the scattered light was perfectly polarized. It has been concluded that a determination of  $P$  as a function of  $\psi$  and  $\theta$  gives good information regarding the shape and the optical constants ( $m_0$  and  $k_0$ ) of the colored spheroidal particles.

The light-scattering of colorless systems has been studied quite extensively by many investigators.<sup>1,2)</sup> However, some biocolloids such as cytochrome C, hemoglobin, ferritin et al., dyes, membrane fragments or metal colloids are colored. These colored biocolloids or colloids often exhibit absorption maxima in the visible region. Under usual experimental conditions, incident light over a visible region is used. Therefore, the light-scattering phenomena are very complicated since we must take into consideration the effects of the absorption together with light scattering. In the case of colored spherical particles, the extinction, scattering and absorption of light have been theoretically studied by means of the Mie theory.<sup>3,4)</sup> For absorbing spheroidal particles, the electrochromic ratio has also been calculated as well as the effect of a complex refractive index and of the axial ratio of the spheroids upon the electrochromic ratio.<sup>5)</sup> However, to the author's knowledge no theory exists for taking into consideration the anisometry effect on the scattering and absorption of light by colored nonspherical particles.

Even if we use completely polarized incident light, the scattered light by anisotropic scatterers will generally be partially polarized and will contain an unpolarized component, since the direction of the induced dipole in an anisotropic particle is different from the direction of the electric vector of incident light. To give a complete description of a partially elliptically polarized beam of scattered light, we must state the intensity associated with both the polarized and unpolarized components, and also give the full specification of the polarized ellipse.

Three parameters must be used to completely define the elliptical polarization of the scattered light: that is, the ratio of the minor and the major axes of the ellipse,  $\eta$  (called the ellipticity), the inclination  $\zeta$  of the major semi-axes of the ellipse with respect to the scattering plane (called the azimuth), and the direction of the motion of the end point of the electric vector of the

light (similarly defined as for circular polarization). Ellipticity can vary between zero and unity, and the azimuth angle varies between 0 and  $\pm 90^\circ$ . Linear and circular forms of polarization can be defined as special cases of elliptical polarization, that is, the ellipticity is 0 or 1, respectively.

The purpose of this work was to study the light scattering of a colored system having a complex index of refraction due to absorption in the above-mentioned visible region. For this purpose, the state of polarization of light scattered by randomly oriented, identical colored spheroidal particles was calculated on the basis of Rayleigh<sup>6)</sup> and Gans<sup>7)</sup> theorem. This will always be referred to as the Rayleigh-Gans theory of spheroids (RGS theory) in order to avoid any confusion with the well-known Rayleigh-Gans (RG) scattering theory involving the restrictive assumption that the refractive index of a particle is close to that of the surrounding medium.

### Theory

**General Consideration.** Let us consider a system of colored and relatively small spheroidal particles dispersed in a colorless medium, where the dispersed phase (consisting of the particles) is optically isotropic and has neither intrinsic birefringence nor intrinsic dichroism. When linearly polarized incident light with its electric vector oriented obliquely to the scattering plane (observation plane) reaches the scattering volume element, the scatterer changes the state of light polarization and the scattered light generally becomes elliptically polarized, but contains unpolarized component as well.

All the possible states of the scattered light can be most easily analyzed by making use of Stokes parameters, and a Mueller or scattering matrix which depends only on the characteristics of the scattering medium.<sup>8)</sup> The Stokes parameters ( $I$ ,  $Q$ ,  $U$ , and  $V$ ), which completely characterize the intensity and the polarization properties of a beam of light, including

partially polarized and unpolarized light, is defined in terms of time averages of the electric-field components of an electromagnetic wave.

$$\begin{aligned} I &= \langle E_\pi E_\pi^* + E_\sigma E_\sigma^* \rangle, \\ Q &= \langle E_\pi E_\pi^* - E_\sigma E_\sigma^* \rangle, \\ U &= \langle E_\pi E_\sigma^* + E_\sigma E_\pi^* \rangle, \\ V &= i \langle E_\pi E_\sigma^* - E_\sigma E_\pi^* \rangle. \end{aligned} \quad (1)$$

The subscripts  $\pi$  and  $\sigma$  refer to components of the electric field parallel and perpendicular, respectively, to the scattering plane, the brackets denote time averages, and an asterisk denotes a conjugate complex value. The four elements of the Stokes vector can be thought of as describing the total intensity ( $I$ ), the excess  $\pi$ -polarization over the  $\sigma$ -polarization intensity ( $Q$ ), the excess intensity of  $+45^\circ$  linear polarization over  $-45^\circ$  linear polarization ( $U$ ), and the excess of right (as opposed to left) circular polarization intensity ( $V$ ).

The Stokes vectors describing incident,  $S_0$ , and scattered,  $S$ , light are connected by a  $4 \times 4$  matrix called Mueller's matrix or scattering matrix,  $M$ ,

$$S = MS_0, \quad (2)$$

where  $S = [I, Q, U, V]$  and  $S_0 = [I_0, Q_0, U_0, V_0]$  are the Stokes vectors of the incident and scattered light.

**Scattering Matrix.** The matrix elements are functions of Euler's angles, the scattering angle  $\theta$  and the optical constants of the particle. If a particle is sufficiently small compared to the wavelength of light, it can be assumed that one dipole is introduced in the particle and that the oscillation of the dipole radiates scattered light according to a theorem proposed by Rayleigh<sup>6</sup> and Gans<sup>7</sup> (RGS theory). The scattering matrix  $M$  for randomly oriented spheroids in suspension is given by the following form (discussed by Van de Hulst<sup>9</sup>):

$$M = \begin{pmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{22} & 0 & 0 \\ 0 & 0 & S_{33} & 0 \\ 0 & 0 & 0 & S_{44} \end{pmatrix}, \quad (3)$$

where the matrix elements  $S_{11}$ ,  $S_{12}$ ,  $S_{22}$ ,  $S_{33}$ , and  $S_{44}$  depend upon the shape of the particle and its complex refractive index. Averaging over Euler's angles for a completely random orientation of particles gives<sup>10</sup>

$$\begin{aligned} S_{11} &= F_1(1 + \cos^2\theta) + 2|B|^2/15, \\ S_{12} &= F_1(\cos^2\theta - 1), \\ S_{22} &= F_1(\cos^2\theta + 1), \\ S_{33} &= 2F_1 \cos\theta, \text{ and} \\ S_{44} &= (2F_1 - 2|B|^2/15) \cos\theta, \end{aligned} \quad (4)$$

where

$$F_1 = \{|A|^2 + 2[\text{Re}A \cdot \text{Re}B + \text{Im}A \cdot \text{Im}B]/3 + 2|B|^2/15\}/2. \quad (5)$$

$\text{Re}A$  and  $\text{Im}A$  are the real and imaginary parts of  $A$ , respectively. In the case of a spheroidal particle,  $A$  and  $B$  are calculated as:

$$\begin{aligned} A &= \alpha^3(m^2 - 1)/\{3p^2[1 + (m^2 - 1)L_b]\}, \text{ and} \\ B &= \alpha^3(m^2 - 1)/\{3p^2[1 + (m^2 - 1)L_a]\} - A, \end{aligned} \quad (6)$$

where the parameter  $\alpha$  is the relative particle radius defined by

$$\alpha = 2\pi a/\lambda. \quad (7)$$

Here,  $a$  is the length of semiaxis symmetry for the spheroid, and  $\lambda$  is the wavelength of light in the medium. The shape parameters  $L_i$  in the  $i$ -direction ( $i=a$  or  $b$ ) are functions of the axial ratio  $p$  ( $=a/b$ ) for a prolate ( $p > 1$ ) and an oblate ( $p < 1$ ) spheroid<sup>11</sup>

$$L_b(p > 1) = (1/2)[p^2/(p^2 - 1)]\{1 - (1/\{p(p^2 - 1)^{1/2}\}) \times \ln\{p + (p^2 - 1)^{1/2}\}\}, \quad (8)$$

$$L_b(p < 1) = (1/2)[p^2/(1 - p^2)]\{1/\{p(1 - p^2)^{1/2}\}\} \arccos p - 1\},$$

and

$$L_a + 2L_b = 1. \quad (9)$$

The refractive index,  $n_2$ , of a colored particle is complex and can be written<sup>9</sup> as

$$n_2 = n_{20}(1 - ik). \quad (10)$$

Then, the relative complex refractive index,  $m$ , is

$$m = n_2/n_0 = m_0 - ik_0, \quad (11)$$

where  $n_0$  is the refractive index of the colorless medium; also,

$$m_0 = n_{20}/n_0 \text{ and } k_0 = m_0 k. \quad (12)$$

If the particle is colorless,  $k$  and  $k_0$  are both zero.

**State of Polarization of the Scattered Light.** The scattered light is, in general, partially polarized. The partially polarized light behaves as an incoherent sum of the totally polarized,  $I_p$  (subscript " $p$ " signify the polarized component), and the unpolarized,  $I_u$  (subscript " $u$ " signify unpolarized), components. Both components are related to the Stokes parameters of the scattered light ( $I, Q, U, V$ ) as follows:

$$\begin{aligned} I_p &= (Q^2 + U^2 + V^2)^{1/2}, \text{ and} \\ I_u &= I - I_p. \end{aligned} \quad (13)$$

In general, the polarized component is elliptically polarized, i.e., the end point of the electric vector moves periodically along an ellipse, which may, of course, reduce in special cases to a circle or a straight line. It is often convenient and sufficient for many experiments to give what is known as the degree of polarization of the light, together with a description of the shape,

azimuth and handedness of the ellipse of the polarized component of the scattered light. The degree of polarization,  $P$ , is defined as the ratio of the intensity of the polarized portion to the total intensity<sup>12)</sup>

$$P = I_p/I. \quad (14)$$

It is generally known that the description of the polarization ellipse of the scattered light is also related to the Stokes parameters of the scattered light as follows:<sup>12)</sup>

(1) The azimuth angle of the ellipse of the polarized component is

$$\zeta = (1/2) \arctan (U/Q), \text{ and} \quad (15)$$

(2) the ellipticity,  $\eta$ , of the ellipse is deduced from

$$2\eta/(1+\eta^2) = V/(Q^2+U^2)^{1/2}. \quad (16)$$

The linear polarization corresponds to  $\eta=0$ .

If we consider linearly polarized incident light with an electric vector that subtends an angle  $\psi$  with the scattering plane, the normalized Stokes parameters of the incident light beam are

$$I_0 = 1, Q_0 = \cos 2\psi, U_0 = \sin 2\psi, \text{ and } V_0 = 0. \quad (17)$$

By substituting  $S_0$  in Eq. 2, we can obtain the Stokes parameters of scattered light, and we can calculate the characteristics of each parameter of a partially polarized scattered light as follows:

$$\begin{aligned} I_p &= I_p^0(1 - \sin^2\theta \cos^2\psi), \\ I &= I_p^0(1 - \sin^2\theta \cos^2\psi) + I_u, \\ I_u &= 2|B|^2/15, I_p^0 = 2F_1, \\ \tan \zeta &= \tan \psi / \cos \theta, \text{ and} \\ \eta &= 0. \end{aligned} \quad (18)$$

According to the RGS theory for an electric dipole, the  $I_u$  term is only a function of  $B$ ; this indicates an optical anisotropy. The term  $I_p$  is linearly polarized when the incident light is linearly polarized. This is because the direction of the induced dipole is different from the direction of the incident electric vector due to the anisotropy of the particle.

The total intensity  $I$  during RGS scattering can be divided into two parts: one is the polarized component  $I_p$  which depends on the scattering angle  $\theta$  together with  $\psi$ ,  $m_0$ ,  $k_0$ , and  $p$ , and the other is the unpolarized component  $I_u$  which is independent of the scattering angle and the angle of inclination of the incident light  $\psi$ .

According to Eqs. 6, 11, and 18, the intensities  $I$  and  $I_p$  are zero if  $k_0$  is zero and  $m_0$  is unity. That is, scattering does not occur in a colorless system if the refractive index of particles and the medium are equal. In the case of a colored system, however, scattering occurs even when  $m_0$  is equal to unity. Generally, there exists no value of  $m_0$  which can make the inten-

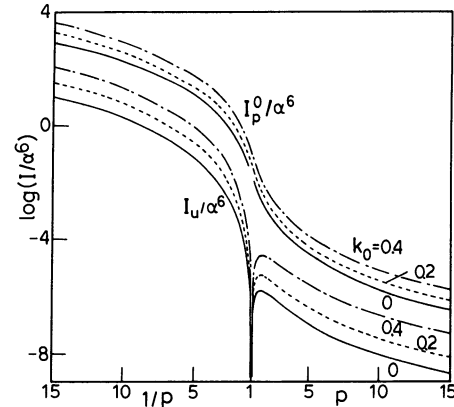


Fig. 1. Variation of  $\log(I_u/\alpha^6)$  and  $\log(I_p^0/\alpha^6)$  with the axial ratios  $p$  for spheroids of  $m_0=1.20$  and various  $k_0$  values;  $k_0=0$  (—),  $k_0=0.2$  (---), and  $k_0=0.4$  (---). For the oblate ( $p<1$ ) spheroids,  $1/p$  (instead of  $p$ ) is plotted on the abscissa to the left direction.

sities  $I$  and/or  $I_p$  equal to zero when  $k_0$  is not zero.

For an isotropic, spherical, colloidal particle, that is for Rayleigh scattering, the degree of polarization is always equal to 1 over the entire range of  $\theta$  since  $B=0$ . In the case of spheroids, the degree of polarization is smaller than 1 and depends on  $m_0$ ,  $k_0$ , and  $p$ , but it is independent of  $\alpha$  in the RGS approximation.

In the case of linearly polarized incident light, the ellipticity,  $\eta$ , of the polarized component of the scattered light is equal to 0; this means that the component is linearly polarized. The azimuth angle of the polarized component,  $\zeta$ , for the spheroids as well as for the spherical particles is equal to  $90^\circ$  in the case of  $\theta=90^\circ$  (according to Eq. 18) for both colored and colorless systems. The  $\zeta$  is equal to  $\psi$  when  $\theta=0^\circ$ ; this means that the direction of the electric field of the forward-scattered light is equal to that of the incident light for both Rayleigh scattering as well as the RGS approximation (if the orientation of particles is completely random).

## Numerical Results and Discussion

**Variation of the Intensity of the Scattered Light.** Numerical calculations were made with an ACOS/20 computer at the Computer Center of the Ministry of Agriculture and Forestry. Figure 1 shows the variation of the intensities of the scattered light for the unpolarized component  $I_u$  and the maximum of the polarized component  $I_p^0$  for prolate and oblate spheroids in the case  $k=m_0/m_0<1$  (that is  $k_0<m_0$ ) as a function of the axial ratio  $p$  and of a given  $m_0$  value (the case  $m_0=1.20$  is given as an example). We call the quantity  $I$  (as well as  $I_p$  and  $I_u$ ) the "intensity" of the scattered light; however, it is actually a dimensionless quantity which is equal to the Rayleigh ratio multiplied by  $(2\pi/\lambda)^2$  per particle. The scattered intensities,  $I_u$  and  $I_p^0$ , for a colored system become greater with increasing  $k_0$  since they include absorp-

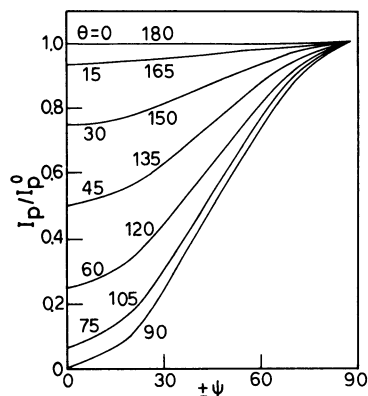


Fig. 2. Variation of  $I_p/I_p^0$  with  $\pm\psi$  (degree) for various values of  $\theta$ . Dependences on  $m_0$ ,  $k_0$ ,  $\alpha$ , and  $p$  are cancelled by taking the ratio of  $I_p$  to  $I_p^0$ , in the RGS approximation.

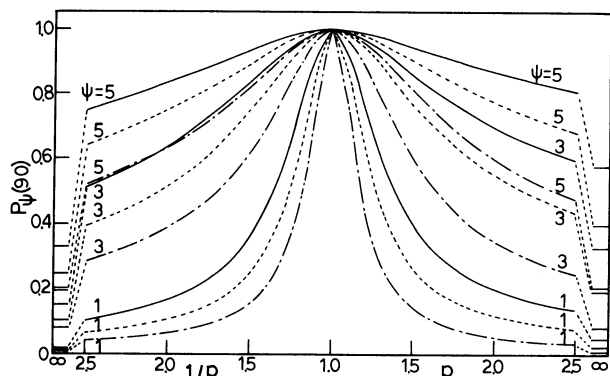


Fig. 3. Variation of the degree of polarization at  $\theta=90^\circ$ ,  $P_\psi(90)$ , with axial ratio of the prolate spheroid ( $p$ ) or of the oblate spheroid ( $1/p$ ) at  $m_0=1.20$  and various  $k_0$  values;  $k_0=0$  (—),  $k_0=0.2$  (---), and  $k_0=0.4$  (— · —). Horizontal lines on both sides indicate the limiting values at  $p \rightarrow \infty$  (infinitesimally thin rod) and  $p \rightarrow 0$  (infinitesimally thin disk), respectively.

tion contributions to the scattering.

Figure 2 shows the variation of the polarized component  $I_p$  with the azimuth angle of the linearly polarized incident light  $\psi$ . By taking the ratio  $I_p/I_p^0$ , all dependencies on  $m_0$ ,  $k_0$ ,  $\alpha$ , and  $p$  can be cancelled just as in an RGS approximation. As shown in Eq. 18, the intensities  $I_p$  and  $I$  are functions of  $\cos^2\theta$ ; therefore, they are symmetric around the  $\theta=90^\circ$  axis.

**Variation of the Degree of Polarization.** The degree of polarization  $P$  defined by Eq. 14 is, together with Eq. 18,

$$P_\psi(\theta) = (1 - \sin^2\theta \cos^2\psi) / [(1 - \sin^2\theta \cos^2\psi) + I_u/I_p^0]. \quad (19)$$

Thus, when  $\theta=90^\circ$ ,

$$P_\psi(90) = \sin^2\psi / (\sin^2\psi + I_u/I_p^0). \quad (20)$$

Figure 3 shows the variation of  $P_\psi(90)$  with the axial ratio  $p$  of the spheroids for various  $k_0$  and  $\psi$  at

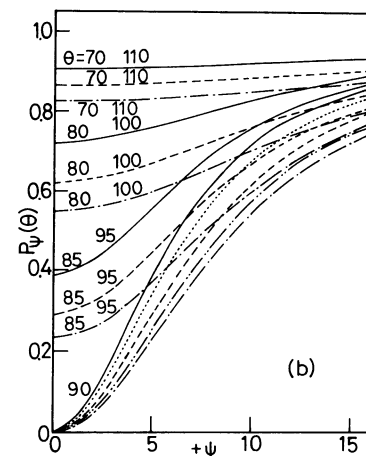
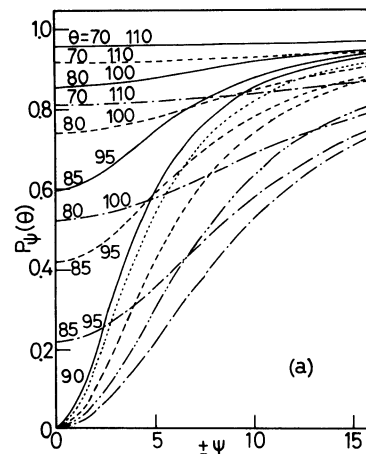


Fig. 4. Variation of the degree of polarization,  $P_\psi(\theta)$  with  $\psi$  (degree) for (a) the prolate spheroid at  $p=15$  and (b) the oblate spheroid at  $p=1/15$  and  $m_0=1.20$ . Parameters are scattering angle  $\theta$  (degree). The  $P_\psi(\theta)$  for various  $k_0$  values at  $\theta=90^\circ$  indicates  $k_0=0$  (—),  $k_0=0.1$  (---),  $k_0=0.2$  (--- · ---),  $k_0=0.3$  (— · —), and  $k_0=0.4$  (— · — · —).

$m_0=1.20$ . The  $P_\psi(90)$  depends strongly on  $p$  and  $\psi$ , especially for a  $\psi$  value smaller than  $5^\circ$ . The  $P_\psi(90)$  is nearly equal to 1.0 if  $\psi$  is larger than  $45^\circ$  since  $I_u$  is about  $1/100$  of  $I_p^0$  (Fig. 1). If  $p=1$ ,  $P_\psi(90)$  is equal to 1.0 irrespective of the value of  $\psi$  and  $k_0$  since  $I_u$  is equal to zero. The dependence of  $P_\psi(90)$  on  $p$  becomes greater as  $k_0$  increases. Also,  $P_\psi(90)$  decreases monotonously upon increasing of  $p$  or  $1/p$ .

The horizontal lines in Fig. 3 are the limiting values for  $p \rightarrow \infty$  ( $L_a=0$  and  $L_b=1/2$ ) on the right side and for  $p \rightarrow 0$  ( $L_a=1$  and  $L_b=0$ ) on the left side. The  $P_\psi(90)$  for an infinitesimally thin disk is much smaller than that for the rodlike particle.

Figure 4 shows the variation of the degree of polarization,  $P_\psi(\theta)$ , in Eq. 19 with  $\pm\psi$  at various scattering angles  $\theta$ . The cases of a prolate spheroid at  $p=15$  and of an oblate spheroid at  $p=1/15$  are shown here as examples. The intensities,  $I$  and  $I_p$ , are functions of  $\sin^2\theta$ ; therefore, the degree of polarization is also symmetric around  $\theta=90^\circ$ . At  $\theta=90^\circ$ ,  $P_\psi(\theta)$  increases monotonously from 0 to almost 1 with an

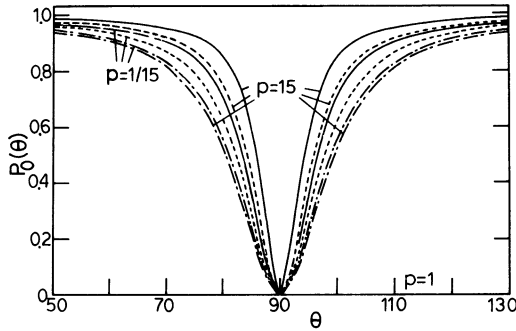


Fig. 5. Variation of the degree of polarization, at  $\psi=0$ ,  $P_0(\theta)$ , for the prolate spheroid at  $p=15$ , the oblate spheroid at  $p=1/15$ , the spherical particle ( $p=1$ ) of  $m_0=1.20$  and various  $k_0$  values;  $k_0=0$  (—),  $k_0=0.2$  (---), and  $k_0=0.4$  (- - -).

increase in  $\pm\psi$ ; its value at a given  $\psi$  decreases with increasing  $k_0$ . The  $P_\psi(\theta)$  at constant  $\theta$  increases monotonously with an increasing  $\pm\psi$ . The dependence of  $P_\psi(\theta)$  on  $\pm\psi$  becomes smaller as the deviation of  $\theta$  from  $90^\circ$  becomes larger and is larger as  $k_0$  becomes larger. The variation of  $P_\psi(\theta)$  for an oblate spheroid is shown in Fig. 4(b). The value of the degree of polarization  $P_\psi(\theta)$  for an oblate spheroid at  $p=1/15$  is a little smaller than that for a prolate spheroid at  $p=15$  since an oblate spheroid has two major axes ( $b$ ) which are responsible for producing an unpolarized component of scattered light while the prolate spheroid has only one major axis ( $a$ ).

For  $\psi=0^\circ$ , Eq. 19 can be rewritten as:

$$P_0(\theta) = \cos^2\theta / [\cos^2\theta + I_u/I_p^2] \quad (21)$$

Figure 5 shows the variation of  $P_0(\theta)$ ; its value is symmetric around  $\theta=90^\circ$  and increase from 0 to almost 1. The  $P_0(\theta)$  at  $\theta=85^\circ$  (or  $95^\circ$ ) for a colorless system ( $k_0=0$ ) is about 0.6, since  $\cos^2 85^\circ = 0.0076$  (which is of the order of magnitude of  $I_u/I_p^2$ ). For a colored system  $P_0(85)$  is about 2/3 of that for a colorless system at  $k_0=0.2$  and about 1/3 at  $k_0=0.4$ .

**Variation of the Degree of Depolarization.** It has been said that the various kinds of degrees of depolarization to be defined can be given as combinations of two fundamental degrees of depolarization,  $\rho_\pi$  and  $\rho_\sigma$ :

$$\begin{aligned} \rho_\pi(\theta) &= I_{\pi\pi}(\theta)/I_{\pi\sigma}(\theta), \text{ and} \\ \rho_\sigma(\theta) &= I_{\sigma\pi}(\theta)/I_{\sigma\sigma}(\theta), \end{aligned} \quad (22)$$

where  $\pi$  and  $\sigma$  indicate the parallel and perpendicular directions, respectively, of the electric vector of light to the scattering plane. The first and the second subscripts are for incident and scattered light, respectively.<sup>13</sup> For example, the degree of depolarization for unpolarized incident light,  $\rho_u$ , is given by  $\rho_\pi$  and  $\rho_\sigma$  as

$$\rho_u = \frac{I_{u\pi}}{I_{u\sigma}} = \frac{I_{\pi\pi} + I_{\sigma\pi}}{I_{\sigma\sigma} + I_{\pi\sigma}} = \rho_\sigma \frac{1 + \rho_\pi}{1 + \rho_\sigma} \quad (23)$$

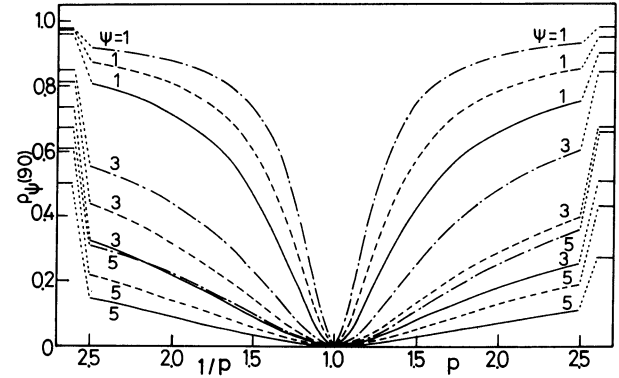


Fig. 6. Variation of the degree of depolarization at  $\theta=90^\circ$ ,  $\rho_\psi(90)$ , with axial ratio  $p$  or  $1/p$  for  $m_0=1.20$  and various  $k_0$  values;  $k_0=0$  (—),  $k_0=0.2$  (---), and  $k_0=0.4$  (- - -).

Horizontal lines on both sides indicate the limiting values at  $p \rightarrow \infty$  (infinitesimally thin rod) and  $p \rightarrow 0$  (infinitesimally thin disk), respectively.

if the particles are either spherical or spheroids oriented completely randomly so that the following Krishnan reciprocity theorem holds.

$$I_{\pi\sigma} = I_{\sigma\pi}. \quad (24)$$

In the RGS approximation for completely random orientation,<sup>14</sup>

$$\begin{aligned} I_{\pi\pi}(\theta) &= I_p^\circ \cos^2\theta + (1/2)I_u, \\ I_{\sigma\sigma}(\theta) &= I_{\pi\sigma}(\theta) = (1/2)I_u, \text{ and} \\ I_{\sigma\pi}(\theta) &= I_p^\circ + (1/2)I_u. \end{aligned} \quad (25)$$

Therefore, for unpolarized incident light

$$\rho_u(\theta) = [\cos^2\theta + I_u/I_p^\circ] / [1 + I_u/I_p^\circ]. \quad (26)$$

This is equal to  $\cos^2\theta$  and is not equal to zero, even if the unpolarized component  $I_u$  is zero (if the angle  $\theta$  is other than  $90^\circ$ ). For  $\theta=90^\circ$ ,

$$\rho_u(90) = I_u/[I_p^\circ + I_u] = 1 - P_{90}(90), \quad (27)$$

where  $P_{90}(90)$  is the degree of polarization for  $\psi=90^\circ$  and  $\theta=90^\circ$ .

Instead of unpolarized light, if linearly polarized light with the azimuth angle of  $\psi$  is used as the incident light, the degree of depolarization is

$$\rho_\psi(\theta) = \frac{I_{\phi\pi}(\theta)}{I_{\phi\sigma}(\theta)} = \frac{I_{\pi\pi}(\theta) \cos^2\theta + I_{\sigma\pi}(\theta) \sin^2\theta}{I_{\sigma\sigma}(\theta) \cos^2\theta + I_{\pi\sigma}(\theta) \sin^2\theta} \quad (28)$$

In the RGS approximation,

$$\rho_\psi(\theta) = [2\cos^2\psi \cos^2\theta + I_u/I_p^\circ] / [2\sin^2\psi + I_u/I_p^\circ], \quad (29)$$

and, for  $\theta=90^\circ$

$$\rho_\psi(90) = I_u/[2I_p^\circ \sin^2\psi + I_u]. \quad (30)$$

These equations show that for  $\psi=0^\circ$ ,  $\rho_0(90)=\rho_\pi(90)=1$ ; for  $\psi=90^\circ$ ,  $\rho_{90}(\theta)=\rho_\sigma(\theta)=I_u/[2I_p^\circ + I_u]$  which is independent of  $\theta$  and is very small because  $I_u/I_p^\circ$  is the order of  $10^{-2}$  as was shown in Fig. 1; for  $\psi=45^\circ$ ,  $\rho_{45}(\theta)=\rho_u(\theta)$  is approximately equal to  $\cos^2\theta$  if  $\theta$  is different

from  $90^\circ$ , but at  $\theta=90^\circ$ ,  $\rho_u(90)$  is nearly equal to  $2\rho_o$  and is very small at the order of  $10^{-2}$ .

In the RGS approximation, the variation of  $\rho_\psi(90)$  for various  $\psi$  values as a function of the axial ratio  $p$  of the particle for  $m_0=1.20$  is shown in Fig. 6. It seems to be very similar to Fig. 3 if the direction of the ordinate is inverted.

**For the Determination of Parameter Values.** As is shown in Figs. 3–6, the degree of polarization or degree of depolarization strongly depends on the axial ratio  $p$ ,  $m_0$ , and  $k_0$  of the spheroids, and is independent of  $\alpha$ . If we measure  $P_\psi(\theta)$ ,  $P_\psi(90)$ , or  $P_\psi(90)$ , the axial ratio for a colorless system is determined directly if the value of  $m_0$  is known. For a colored system, if we measure  $P_\psi(\theta)$  at several values of  $\psi$  and  $\theta$ , it is possible to determine both  $k_0$  and  $p$  from the curve-fitting method to the theoretical values and to determine whether the particle is prolate or oblate. On the contrary, if the value of the axial ratio  $p$  of the colored spheroid is known from the other physicochemical techniques, it is also possible to determine both  $m_0$  and  $k_0$  values of the colored spheroid. In conjunction with the knowledge of the axial ratio, the value of  $\alpha$  is also obtained from the values of the intensities ( $I$ ,  $I_p$ , and  $I_u$ ), which are obtained simultaneously. Knowledge about  $\alpha$  and  $p$  immediately gives information regarding the molecular weight  $M$  of the spheroid (for both colored and colorless systems) if the partial specific volume is known separately.

The value of  $k_0$  can be large if the particles are colored. It can even be infinitely larger if the particles are made of a good conductor; however, the extinction of the dispersed system as a whole can be made as small as possible in experiments by diluting the dispersed system. Therefore, extremely small  $k_0$  values are experimentally interesting. In particular, the value of the refractive index of a colored system is not practically equal to that of a colorless system if the value of  $k_0$  is larger than  $10^{-4}$ .<sup>3)</sup>

**Measurement of Degree of Polarization with the Analyzer.** The intensity of the irradiance transmitted through the analyzer with a transmission axis inclined by  $\chi$  to the scattering plane,  $I_{\psi\chi}(\theta)$ , is calculated by using the Mueller matrix for an ideal linear analyzer as follows:<sup>9)</sup>

$$\begin{aligned} I_{\psi\chi}(\theta) &= (I_u/2) + I_p \{1 + \cos 2(\chi - \zeta)\}/2 \\ &= I \frac{1 + P_\psi(\theta) \cos 2(\chi - \zeta)}{2}, \end{aligned} \quad (31)$$

where  $\zeta$  is the azimuth angle of the scattered polarized component. In the case of the degree of depolarization the two special cases of  $\chi=0^\circ$  (parallel) and  $\chi=90^\circ$  (perpendicular to the scattering plane) is generally discussed.

The variation of  $I_{\psi\chi}(\theta)$  is shown in Fig. 7. When  $\chi$  is changed by rotating the analyzer, the intensity of the transmittance  $I_{\psi\chi}(\theta)$  has maximum,  $I_{\max}(\theta)$ , and minimum,  $I_{\min}(\theta)$ , values at  $\chi=\zeta$  and  $\chi=\zeta+\pi/2$ , respectively

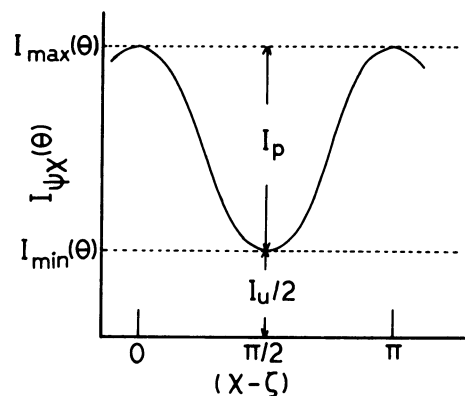


Fig. 7. Variation of transmitted intensity of the partially polarized light with analyzer orientation.

(Fig. 7). The degree of polarization can, therefore, be experimentally measured by rotating the analyzer by using

$$P_\psi(\theta) = [I_{\max}(\theta) - I_{\min}(\theta)]/[I_{\max}(\theta) + I_{\min}(\theta)]. \quad (32)$$

This is equivalent to thinking of the beam as being made up of two linearly polarized orthogonal incoherent beams with intensities  $I_{\max}(\theta)$  and  $I_{\min}(\theta)$ .

As mentioned in the INTRODUCTION, it is important to determine the size and shape of the colored biocolloids such as cytochrome C, hemoglobin, ferritin et al., dyes, membrane fragments prepared from the plant plasma membrane, and/or the various kinds of metal colloids. It is also important to determine the  $k_0$  values in order to determine the physico-chemical properties of these colored molecules. For this purpose, the theory presented in this paper can be efficiently applied to these colored systems.

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