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State of Polarization of Light Scattered by Anisotropic Spherical Particles

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Using the light-scattering theory of Rayleigh-Gans, theoretical expressions were derived for the state of the polarization of light scattered by anisotropic spherical particles in the case of linearly polarized incident light with its electric vector inclined by an angle ψ to the scattering plane. The azimuth angle of the ellipse of the light scattered is a function of only the scattering angles, θ and ψ; the ellipticity of the ellipse of the scattered light is always equal to zero, meaning that the scattered light is linearly polarized for linearly polarized incident light. The degree of polarization of the scattered light, P, defined as the ratio of the polarized component to the total intensity of the scattered light, depends on the intrinsic anisotropy, p_m, the relative refractive index of the sphere to the solvent, m, and θ together with the angle ψ. Although P for isotropic spherical particles is always equal to 1 at any θ, P for anisotropic spherical particles is smaller than 1.0, but close to 1.0 if the incident light is unpolarized or linearly polarized with ψ=30°. If ψ is smaller than 30°, P varies from 1.0 to 0, depending strongly on the p_m and m of the anisotropic sphere. It has been suggested that the determination of P as a function of ψ and θ would provide good information about the optical intrinsic anisotropy of anisotropic spherical particles.

A particle may have two kinds of optical anisotropy: one is an "intrinsic" anisotropy and the other is a "form" anisotropy.[1] The form anisotropy is an optical anisotropy due to an anisometry. In other words, it is due to the anisotropic shape of a particle made of the isotropic phase. If the shape of the particle is nonspherical and is assumed to be a spheroid, the anisometry can be expressed by the ratio of the length of semiaxis of symmetry to the length of semitransverse axis of the spheroid. For an anisometric particle, the refractive index of the dispersed phase may be independent of the vibration direction of the electric vector. On the other hand, an intrinsic anisotropy occurs when the dispersed phase (the material composing the particle), itself, has an optical anisotropy because of, e.g., the orientation of valence bonds in the phase. For an intrinsic anisotropic particle, the refractive index of the dispersed phase depends on the vibration direction of the electric vector, and, even if the particle is spherical, the effect of the intrinsic anisotropy must be taken into consideration.

The light scattered by optically anisotropic scatterers will generally be partially polarized and contain some unpolarized component. To give a complete description of a partially elliptically polarized state of scattered light, we should analyze the intensities associated with both polarized and unpolarized components, and also give full specifications of the polarization ellipse.

In the case of randomly oriented, optically isotropic and anisometric particles, the state of the polarization of scattered light had already been calculated[2,3] on the basis of the Rayleigh[4]-Gans[5] theory (referred to as the Rayleigh-Gans theory of spheroids (RGS theory)). The RGS theory is used to avoid any confusion with the well-known Rayleigh-Gans (RG) theory of scattering, which involves a restrictive assumption that the refractive index of the particles is close to that of the surrounding medium.[6]

In the present paper, the effect of the intrinsic optical anisotropy to the state of polarization of light scattered by optically anisotropic spherical particles, such as spherical plant viruses, is discussed on the basis of the RGS theory and a set of parameters: i.e., the degree of polarization, ellipticity, azimuth, and handedness of the ellipse of the polarized component of scattered light is examined.

Theory

General Consideration. Let's consider light scattering by an optically anisotropic spherical particle. If a dispersed sphere is optically uniaxial, the excess polarizability can be expressed by a spheroid.[7] If the principal axes of the spheroid are α_a, α_b, α_b, the optical anisotropy is given by the ratio p_g,

$$p_g = \alpha_a/\alpha_b = g_a/g_b, \tag{1}$$

where

$$g_i = (3/4\pi)\,(m_i^2-1)/(m_i^2+2) \quad (i = a \text{ or } b), \tag{2}$$

and m_i is the relative refractive index of the i-direction.

When linearly polarized incident light with an electric vector oriented obliquely to the scattering plane (observation plane) reaches the scattering-volume element, the light scattered will be, in general, elliptically polarized and contain some unpolarized component as well.

All possible states of the scattered light are most easily analyzed by making use of the Stokes parameters as well as the Mueller or scattering matrix, which depends only on the characteristics of the scattering medium. The Stokes parameters(I, Q, U, and V) which completely characterize the intensity and polarization properties of a beam of light, including partially polarized and unpolarized light, are defined in terms of time averages of the electric field components of an electromagnetic wave.[8]

$$\begin{aligned} I &= \langle E_\pi E_\pi^* + E_\sigma E_\sigma^* \rangle, \\ Q &= \langle E_\pi E_\pi^* - E_\sigma E_\sigma^* \rangle, \\ U &= \langle E_\pi E_\sigma^* + E_\sigma E_\pi^* \rangle, \\ V &= \mathrm{i}\langle E_\pi E_\sigma^* - E_\sigma E_\pi^* \rangle. \end{aligned} \tag{3}$$

The subscripts π and σ refer to components of the electric field parallel and perpendicular, respectively, to the scattering plane, the brackets denote time averages, and the asterisks denote conjugate complex values. The four elements of the Stokes vector can be thought of as describing the total intensity (I), the excess of π-polarized over σ-polarized intensity (Q), the excess intensity of $+45°$ linearly polarized over $-45°$ linearly polarized light (U), and the excess of right (as opposed to left) circularly polarized intensity (V).

The Stokes vectors describing the incident and scattered light, $\boldsymbol{S}_0$ and $\boldsymbol{S}$, respectively, are connected by a 4×4 matrix called a Mueller's matrix or a scattering matrix, $\boldsymbol{M}$,

$$\boldsymbol{S} = \boldsymbol{M}\boldsymbol{S}_0, \tag{4}$$

where $\boldsymbol{S}=[I, Q, U, V]$ and $\boldsymbol{S}_0=[I_0, Q_0, U_0, V_0]$

Scattering Matrix. If the particle is sufficiently small compared to the wavelength of light, the scattering matrix $\boldsymbol{M}$ for a randomly oriented spheroid is given by RGS theory:[9)]

$$\boldsymbol{M} = \begin{pmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{22} & 0 & 0 \\ 0 & 0 & S_{33} & 0 \\ 0 & 0 & 0 & S_{44} \end{pmatrix}, \tag{5}$$

where the matrix elements S_{11}, S_{12}, S_{22}, S_{33}, S_{44} are given by

$$\begin{aligned} S_{11} &= F_1(1+\cos^2\theta)+2F_2, \\ S_{12} &= F_1(\cos^2\theta-1), \\ S_{22} &= F_1(\cos^2\theta+1), \\ S_{33} &= 2F_1\cos\theta, \text{ and} \\ S_{44} &= 2\,(F_1-F_2)\cos\theta, \end{aligned} \tag{6}$$

with

$$\begin{aligned} F_1 &= (\alpha^6/15)\,(p_g^2+3p_g+7/2)\,F_0, \\ F_2 &= (\alpha^6/15)\,(p_g-1)^2\,F_0, \text{ and} \\ F_0 &= [(1+2p_m)\,(m^2-1)]^2/[(m^2-1)+p_m(2m^2+7)]^2. \end{aligned} \tag{7}$$

The α is the size parameter defined by

$$\alpha = 2\pi a/\lambda, \tag{8}$$

where a is the radius of the sphere and λ is the wavelength of light in the medium.

If the principal refractive indices in the a- or b-directions of the excess polarizabilities n_a and n_b are divided by the refractive index of the solvent n_0 and are represented by (m_a, m_b, m_b), the intrinsic (structural) anisotropy p_m is defined by

$$p_m = (m_a^2-1)/(m_b^2-1), \tag{9}$$

and the mean relative refractive index m of the spherical particle is defined by[1)]

$$1/(m^2-1) = [1/(m_a^2-1)+2/(m_b^2-1)]/3; \tag{10}$$

therefore, the optical anisotropy p_g defined by Eq. 1 for spherical anisotropic particles is given by

$$p_g = [p_m(2m^2+7)+(m^2-1)]/[2p_m(m^2-1)+(m^2+8)]. \tag{11}$$

State of Polarization of the Scattered Light. The scattered light is, in general, partially polarized and is an incoherent sum of the polarized, I_p (subscript "p" signifies the polarized component), and the unpolarized, I_u (subscript "u" signifies unpolarized), components. Both components are related to the Stokes parameters of scattered light by

$$I_p = (Q^2+U^2+V^2)^{1/2}, \tag{12}$$

and

$$I_u = I - I_p.$$

If we consider linearly polarized incident light with an electric vector which makes an angle ψ with the scattering plane, the Stokes vector can be expressed by the normalized Stokes parameters of the incident light beam as

$$\boldsymbol{S}_0 = [1, \cos2\psi, \sin2\psi, 0]. \tag{13}$$

By substituting the value of $\boldsymbol{S}_0$ in Eq.4, we obtain the values for each of the Stokes parameters of partially polarized scattered light as follows:

$$\begin{aligned} I_p &= I_p{}^\circ\,(1-\cos^2\psi\,\sin^2\theta), \\ I &= I_p{}^\circ(1-\cos^2\psi\,\sin^2\theta)+I_u, \\ I_p{}^\circ &= 2F_1,\ I_u = 2F_2, \text{ and} \\ \eta &= 0. \end{aligned} \tag{14}$$

The ellipticity, η, defined by the ratio of the semiminor to the semimajor axis of the ellipse of the polarized component of scattered light is always equal to 0, which means that the polarized component is linearly polarized in the case of linearly polarized incident light.

The azimuth angle, ζ, of the linearly polarized component of the scattered light, is given by

$$\tan\zeta = \tan\psi/\cos\theta, \tag{15}$$

and is only a function of ψ and θ.

The degree of polarization, P, is defined as the ratio of the intensity of the polarized portion to the total intensity,

$$P_\psi(\theta) = I_p/I. \tag{16}$$

Therefore, we obtain with Eq. 14

$$P_\psi(\theta) = (1-\sin^2\theta\cos^2\psi)/[(1-\sin^2\theta\cos^2\psi)+(F_2/F_1)]. \tag{17}$$

Since

$$F_2/F_1 = (p_g-1)^2/(p_g^2+3p_g+7/2), \tag{18}$$

and p_g is given by Eq. 11, the $P_\psi(\theta)$ depends on p_m, m, θ, and ψ. For anisotropic spherical particles ($p_m \neq 1$), the $P_\psi(\theta)$ is smaller than 1.0 and is independent of α, as far as RGS theory holds.

A particle composed of isotropic material has a relative refractive index m equal in all directions of the electric vector of light, independently of the particle form. Therefore, in this case, $p_m=1$ (and $p_g=1$) and

$$\begin{aligned} F_1 &= (\alpha^6/2)\, F_0, \\ F_2 &= 0, \text{ and} \\ F_0 &= [(m^2-1)/(m^2+2)]^2. \end{aligned} \tag{19}$$

The $P_\psi(\theta)$ is always equal to 1 over the entire range of θ in this case.

Numerical Results and Discussion

Variation of the Intensity of the Scattered Light. Numerical calculations were made with an ACOS 850/20 computer at the Computer Center of the Ministry of Agriculture and Forestry. Figure 1 shows the variation in the intensities of the scattered light for the unpolarized component I_u and the maximum of the polarized component $I_p{}^\circ$ as a function of the intrinsic anisotropy p_m for $m=1.20$. Here, the quantity I (and I_p and I_u as well) is called the "intensity" of the scattered light; however, it is a dimensionless quantity which is equal to the Rayleigh ratio per particle multiplied by k^2 (where $k=2\pi/\lambda$), and is usually expressed by i.

Bovine serum albumin is known to be somewhat asymmetrical and to possess an intrinsic (structural) anisotropy. The intrinsic anisotropy parameter of bovine serum albumin in the native state was calculated to be $p_m=1.46$.[10] Spherical plant viruses such as tomato bushy stunt virus, southern bean mosaic virus, satellite tobacco necrosis virus et al. contain different amounts of nucleic acids and proteins, although some of these viruses produce nucleic acid-free isometric particles. In some of the viruses the particles consist of various kinds of amino acid residues and in others the particles consist of a long backbone of alternating sugar and phosphate residues with one of five different purine or pyrimidine bases attached to each sugar residue. These residues have an anisotropic structure. Therefore, spherical plant viruses possess an intrinsic anisotropy due to the orientation of valence bonds of the amino acid and/or nucleic acid residues. Taking into account the experimental results of these spherical plant viruses, it is satisfactory to think about the values of $p_m<1.9$ and also to think the case of $m_a>m_b$, that is, the values of $p_m>1$.

Variation of the Azimuth Angle ζ. Figure 2 shows the variation of the azimuth angle, ζ, with ψ for various θ. As is shown in Eq. 15, the value of ζ is independent of m, α and p_m. The ζ has the same sign as ψ for forward scattering ($0<\theta<90°$) and has the opposite sign for the backward scattering ($90°<\theta<180°$). ζ becomes greater as θ approaches 90°, and at $\theta=90°$ the value of ζ is equal to 90° for any value of ψ.

Variation of the Degree of Polarization. The degree of polarization at $\theta=90°$ given by Eq. 17 is

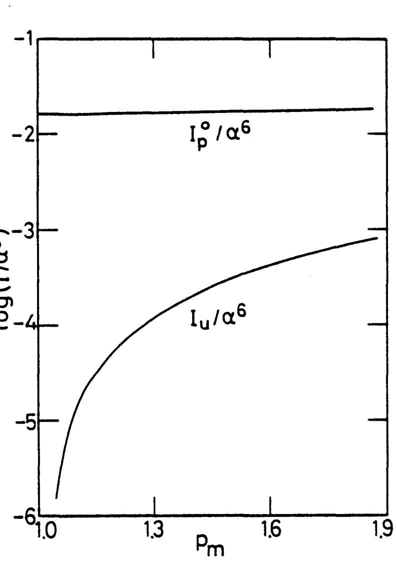

Fig. 1. Variation of $\log(I_u/\alpha^6)$ and $\log(I_p{}^\circ/\alpha^6)$ with the intrinsic anisotropy $p_m>1$ for $m=1.20$.

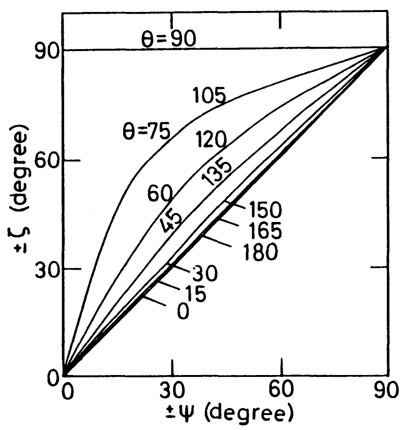

Fig. 2. Variation of the azimuth angle, ζ(degree), with ψ(degree) at various scattering angles θ(degree). For $\theta<90°$, ζ and ψ have the same sign, and for $\theta>90°$, ζ and ψ have the opposite sign.

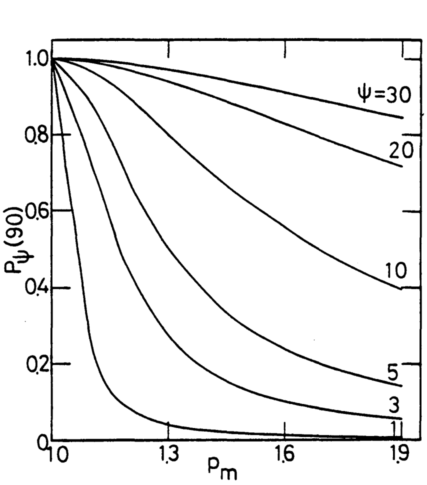

Fig. 3. Variation of the degree of polarization at $\theta=90°$, $P_\psi(90)$, with the intrinsic anisotropy $p_m>1$ and $m=1.20$ for various values of ψ(degree).

$$P_\psi(90) = \sin^2\psi/(\sin^2\psi + F_2/F_1). \tag{20}$$

Figure 3 shows the variation of $P_\psi(90)$ with the intrinsic anisotropy p_m and ψ, at m=1.20. $P_\psi(90)$ depends strongly on p_m and ψ, especially in cases in which the ψ is smaller than 10°. When ψ is larger than 30°, the change of the $P_\psi(90)$ is relatively small. The $P_\psi(90)$ for isotropic spherical particles (p_m=1) is equal to 1.0 irrespective of the value of ψ.

Figure 4 shows the variation of $P_\psi(90)$ with $\pm\psi$. The $P_\psi(90)$ increases monotonously from 0 to almost 1 with the increase of $\pm\psi$, and its value becomes smaller as the p_m deviates from the isotropic materials (p_m=1).

Figure 5 shows the variation of $P_\psi(\theta)$ with $\pm\psi$ at various scattering angle θ for p_m=1.4. The $P_\psi(\theta)$ is symmetric around θ=90° since the intensities, I and I_p, are functions of $\sin^2\theta$, as is shown in Eq. 17. The dependence of $P_\psi(\theta)$ on $\pm\psi$ becomes smaller as the deviation of θ from 90° becomes larger. In the case of θ=0° or 180° the $P_\psi(\theta)$ is constant independent of $\pm\psi$.

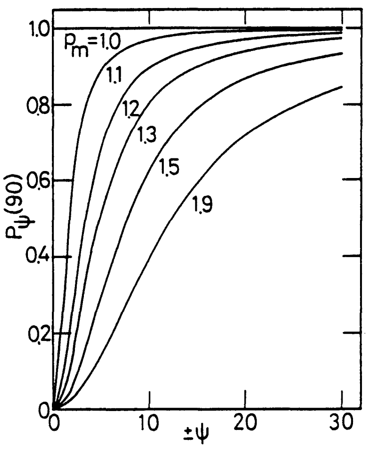

Fig. 4. Variation of the degree of polarization at θ=90°, $P_\psi(90)$, with ψ(degree) and m=1.20 for various values of $p_m \geq 1$.

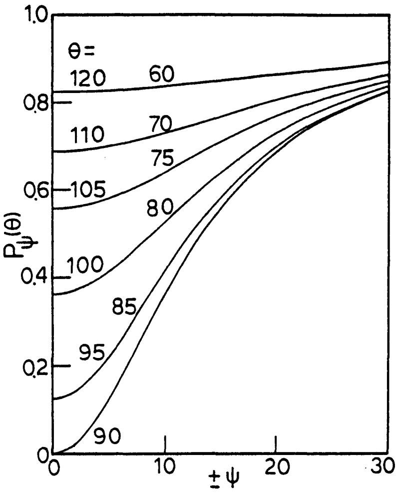

Fig. 5. Variation of the degree of polarization, $P_\psi(\theta)$, with ψ(degree) for p_m=1.4 and m=1.20. Parameters are scattering angle θ(degree).

Variation of the Degree of Depolarization. The intensity of the scattered light is measured practically by the irradiance transmitted through the analyzer with a transmission axis inclined by χ to the scattering plane. The degree of depolarization is defined by the ratio of the intensity of the parallel (i.e., χ=0°) to the perpendicular (i.e., χ=90°) component of the scattered light.

For unpolarized incident light, the degree of depolarization is given by

$$\rho_u(\theta) = [\cos^2\theta + F_2/F_1]/[1 + F_2/F_1], \tag{21}$$

which is equal to $\cos^2\theta$ and is not equal to zero when the scattering angle θ is other than 90°, even if the unpolarized component I_u is zero for the optically isotropic particles (p_m=1). At θ=90°, it becomes

$$\rho_u(90) = F_2/(F_1 + F_2) = 1 - P_{90}(90), \tag{22}$$

where $P_{90}(90)$ is the degree of polarization for ψ=90° and θ=90°.

Instead of unpolarized light, if linearly polarized light with an azimuth angle of ψ is used as incident light, the degree of depolarization for θ=90° is

$$\rho_\psi(90) = F_2/[2F_1 \sin^2\psi + F_2]. \tag{23}$$

The variation of $\rho_\psi(90)$ with p_m is shown in Fig. 6, which seems to be very similar to Fig. 3 if the direction of the ordinate is inverted. The value of $\rho_\psi(90)$ is very small if ψ is 30° or larger. The value, however, could become larger, if ψ is decreased.

For the Determination of Parameter Values. As is shown in Figs. 3—6, the degree of polarization, or degree of depolarization, depends strongly on the intrinsic anisotropy p_m, and is independent of α. If we measure a $P_\psi(\theta)$ or $P_\psi(90)$ value, and/or $\rho_\psi(\theta)$ or $\rho_\psi(90)$ value, the intrinsic anisotropy can be determined directly if the value of m is known. If we measure $P_\psi(\theta)$ or $\rho_\psi(\theta)$ values at several values of ψ and θ, it is possible to determine both m and p_m by fitting these values to the theoretical curves.

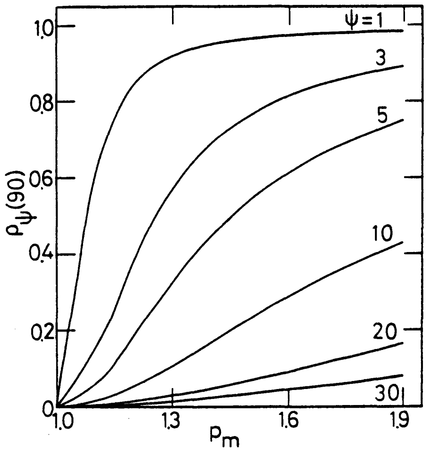

Fig. 6. Variation of the degree of depolarization at θ=90°, $\rho_\psi(90)$, with the intrinsic anisotropy p_m>1 and m=1.20.

Knowledge about p_m and m immediately gives information regarding the refractive index of both the a- and b-axis of the intrinsic (structural) anisotropic spherical particle, n_a and n_b (according to Eqs. 9 and 10) if the refractive index of the solvent n_0 is known.

In conjunction with knowledge concerning the intrinsic anisotropy, p_m, the value of α is also obtained from the intensities (I, I_p, and I_u) according to Fig. 1. Also, knowledge concerning α immediately gives information on the molecular weight, M, if the partial specific volume is known.

The theory presented in this paper can be efficiently applied to small spherical plant viruses. However, plant virus particles sometimes form by a process, analogous to crystallization, by an orderly aggregation of similarly sized units into three-dimensional arrays. The nucleic acid of viruses also has a definite structure and is not simply a randomly coiled thread, stimulating the crystallization structure. In these cases the effect of the intrinsic anisotropy is large, and a slight modification is necessary for any determination of the intrinsic anisotropy, due to the nonrandom orientation of the segments.

Measurement of Degree of Polarization with the Analyzer. When the transmission axis of the analyzer χ is changed, the intensity of the scattered light $I_{\psi\chi}(\theta)$ is given by[8,11]

$$I_{\psi\chi}(\theta) = (I_u/2) + (I_p/2)[1 + \cos^2(\chi - \zeta)], \\ = (I/2)[1 + P_\psi(\theta)\cos^2(\chi - \zeta)], \tag{24}$$

where ζ is the azimuth angle of the linearly polarized, scattered component. The $I_{\psi\chi}(\theta)$ has a maximum, $I_{\max}(\theta)$, and a minimum, $I_{\min}(\theta)$, values at $\chi=\zeta$ and $\chi=\zeta+\pi/2$, respectively. The degree of polarization can, therefore, be experimentally measured using the equation

$$P_\psi(\theta) = [I_{\max}(\theta) - I_{\min}(\theta)]/[I_{\max}(\theta) + I_{\min}(\theta)]. \tag{25}$$

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

If another kind of the degree of linear polarization is considered, which is defined directly by the experimentally measurable intensities of the irradiance through the transmission axis of the analyzer at χ and at $(\chi+\pi/2)$, respectively,

$$P_{\psi\chi}(\theta) = [I_{\psi\chi}(\theta) - I_{\psi(\chi+\pi/2)}(\theta)]/[I_{\psi\chi}(\theta) + I_{\psi(\chi+\pi/2)}(\theta)], \tag{26}$$

the deviation of $P_{\psi\chi}(\theta)$ from $P_\psi(\theta)$ is given by

$$P_{\psi\chi}(\theta) = P_\psi(\theta)\cos 2(\chi - \zeta). \tag{27}$$

The maximum and minimum values of $P_{\psi\chi}(\theta)$ occur when χ is equal to the azimuth angle ζ and is equal to the orthogonal direction to the azimuth angle, respectively. The $P_{\psi\chi}(\theta)$ is equal to zero at $\chi=(\zeta+\pi/4)$.

Radiation Diagram of Intensity of the Transmitted Light through the Analyzer. The intensity of the irradiance transmitted through the analyzer can also be rewritten as

$$I_{\psi\chi}(\theta) = F_2 + F_1 \cdot F_{\psi\chi}(\theta), \tag{28}$$

and

$$F_{\psi\chi}(\theta) = (1 - \sin^2\theta\cos^2\psi) + (\cos^2\theta\cos^2\psi - \sin^2\psi)\cos 2\chi,$$

where both F_2 and F_1 are functions of p_m, m, and α (as explained before), while $F_{\psi\chi}(\theta)$ is a function which depends only on angles θ, ψ, and χ. Figures 7 and 8 are radiation diagrams of the function, $F_{\psi\chi}(\theta)$, which shows a maximum and a minimum, depending on ψ, χ, and θ.

For $\theta=0°$, the $F_{\psi\chi}(0)$ can be simplified as

$$F_{\psi\chi}(0) = 2\cos^2(\psi - \chi). \tag{29}$$

If the angle between the transmission direction of the

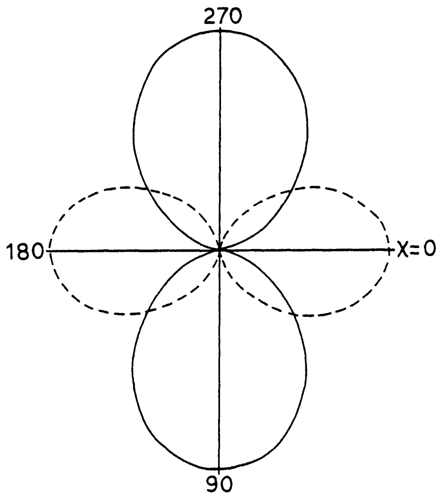

Fig. 7. Radiation diagram of the function of $F_{\psi\chi}(\theta)$. Parameters in the figure are transmission angles of analyzer χ(degree) to the scattering plane. Solid line represents the case for $\psi=90°$ without depending on θ. Broken line represents the case for $\psi=0°$ and $\theta=30°$ or $150°$. The $F_{\psi\chi}(\theta)$ at $\theta=0°$ and $\theta=90°$ is situated on an origin.

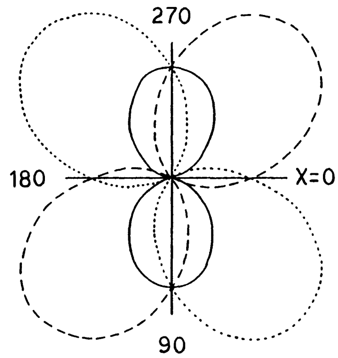

Fig. 8. Radiation diagram of the function of $F_{\psi\chi}(\theta)$ for $\psi=45°$ or $\psi=-45°$. Parameters in the figure are transmission angles of analyzer χ(degree). Solid line represents the value at $\theta=90°$ for $\psi=\pm45°$. Broken line indicates the value at $\theta=150°$ for $\psi=+45°$, and at $\theta=30°$ for $\psi=-45°$. Dotted line represents the value at $\theta=30°$ for $\psi=+45°$, and at $\theta=150°$ for $\psi=-45°$.

polarizer and the analyzer is $\pm\delta$,

$$I_{\psi x}(0)=(I_u/2)+I_p^{\circ}\cos^2\delta. \tag{30}$$

The second term in Eq. 30 is called Malus' law, which was discovered experimentally by Etienne Louis Malus in 1808.[8] Only the polarized component of the forward scattered light ($\theta=0°$) follows Malus' law; half of the intensity of the unpolarized component of the scattered light must, furthermore, be introduced to the total intensity of the forward scattered light.

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