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existence of certain other compounds. Thus one would expect that the polarizability of the anion of the element 85 would be considerably higher than that of I^-. The discoverers[18] of the radioactive species of this element report that it does not co-precipitate with AgI and behaves like a metallic element rather than as a halogen. One can express this fact from the above point of view by saying that the large deformability expected for the anion of ekaiodine leads to the instability of its salts.

In $CO_3^=$ the refraction per oxygen octet (4.08) is smaller than in the analogous $SiO_3^=$ (4.42) because the field of the small C^{4+} (helium type) is stronger than that of Si^{4+} (neon type). Thus by analogy one would expect $O^=$ in FO_4^-, FO_3^+, or FO_2^{3+} to be deformed by the very small F^{7+} still more strongly than by Cl^{7+} in ClO_4^-. None of these complex ions of fluorine exists; however, F_2O is a stable compound, which can be considered as formed by interaction within the grouping $F^-O^{2+}F^-$. One can interpret the imaginary reaction $2KFO_4 \rightarrow F_2O + K_2O + 3O_2$ as the result of the strong deformation of the oxygen ions by F^{7+}. Obviously this is only a way of formulating the fact that the electron affinity of fluorine is larger than that of oxygen. However, the advantage of the above point of view becomes evident when one tries to explain why ClO_4^- exists but BrO_4^- is unknown. The probable reason is that Br^{7+} has a non-rare gas structure and must have a much larger deforming power[19] than Cl^{7+}, as does Cu^+ compared with Na^+. The ion I^{7+}, which also is of the non-rare gas type but which must be considerably larger than Br^{7+}, combines with $O^=$ to form relatively stable periodates. From our point of view, this would mean that I^{7+} exerts a smaller influence on the oxygen octet than does Br^{7+}, for the same reason the Cl^{7+} has a smaller influence than has F^{7+}.

[18] D. R. Corson, K. R. MacKenzie, and E. Segrè, Phys. Rev. **58**, 672 (1940).

[19] K. Fajans, J. Chem. Phys. **9**, 281, 378 (1941).

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Polarization of Light Scattered by Isotropic Opalescent Media*

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A general study is given of the polarization of light scattered by isotropic media whose elements of heterogeneity are not very small in comparison with the wave-length, (suspensions, colloidal solutions, solutions of large molecules, . . .). This includes an extension of a theory by R. S. Krishnan, who, considering certain particular states of polarization of the incident light and applying the law of reciprocity, had proved the equality of two of the four coefficients which are to be considered in these cases. Using Stokes' linear representation of the polarization of light beams, it is shown that the scattering through a given angle and for a given wave-length is characterized by the 16 coefficients of the linear forms which express the four polarization parameters of the scattered beam in terms of the four corresponding parameters of the incident beam and that the law of reciprocity leads to six relations between these sixteen coefficients. For an isotropic asymmetrical medium (having rotatory power), the scattering is thus characterized by *ten* independent coefficients. In the case of a symmetrical medium, four of these coefficients must be zero, leaving only *six* scattering coefficients, and if the scattering particles are spherical, there are two additional relations between these coefficients. The comparison with dipolar scattering by very small elements shows that the best test to prove multipolar scattering is the existence of some ellipticity in the scattered light when the incident beam is linearly polarized in a direction oblique to the scattering plane.

I

THE scattering of light by a macroscopically homogeneous medium is caused by some microscopical structure. If the dimensions of the elements of this structure are very small in comparison with the wave-length of the light, the scattering has the well-known simple characteristics of secondary dipolar emission.[17–20] But if

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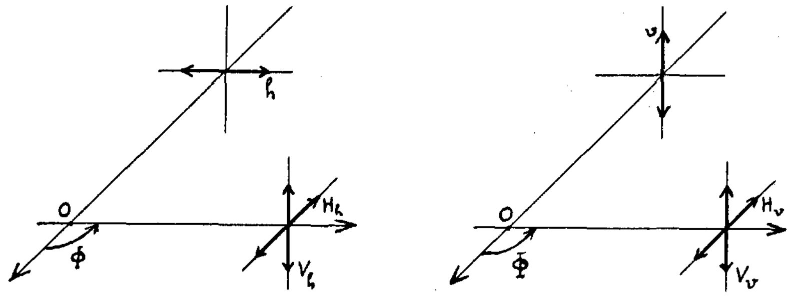


FIG. 1.

these elements have dimensions comparable to the wave-length, the phenomenon is more complicated, and has been experimentally studied only in particular conditions of excitation or observation, and theoretically only for spherical scattering particles.[7,21]

Our purpose is to extend a method used by R. S. Krishnan, and to point out the independent parameters which are necessary for specifying, in general, the intensity and polarization of the light scattered by any isotropic medium for given scattering angle and wave-length. A summary of this research has been read before the French Society of Physical Chemistry in May, 1939.[22]

We shall have to distinguish between *symmetrical* media, for which the center of any large spherical volume is a center of symmetry and any plane through this center a plane of symmetry, and the asymmetrical media, which usually have some optical rotatory power. The isotropy may be only statistical, as the result of an isotropic distribution of small anisotropic elements.

We shall see that for isotropic media, whose scattering elements are not very small compared with the wave-length, media which are more or or less turbid or opalescent, it is necessary to introduce several new parameters, whose values will contribute to the determination of the magnitude, shape, and optical properties of these elements. This method will be applicable in the study of smokes, fogs, suspensions, emulsions, colloidal solutions, solutions of large molecules, and also of media with widely extended fluctuations such as pure fluids, or liquid mixtures, near their critical state, glasses, etc.

My thanks are due to Dr. R. Wurmser with whom I had several discussions on these questions of scattering, which were the origin of this research.

II. THE RELATION OF R. S. KRISHNAN

In several papers published in 1938, R. S. Krishnan[1–5] has established theoretically, and has given the experimental verification of, a relation between the intensities of certain components of the light scattered by an isotropic symmetrical medium for some particular conditions of polarization of the incident light.

At a point of a horizontal light beam linearly polarized and of given intensity, he considered the scattering in a horizontal direction making an angle ϕ with the incident beam. He denoted by H_h and V_h the intensities of the horizontal and vertical vibrations of the scattered beam when the direction of vibration of the incident beam is horizontal, and by H_v and V_v these intensities when this direction is vertical (Fig. 1). He defined the corresponding depolarization factors of the scattered light by the ratios (generally smaller than 1)

$$\rho_h = V_h/H_h, \quad \rho_v = H_v/V_v. \tag{1}$$

The superposition without any phase relation, of the two considered incident beams, polarized at right angles and of the same intensity, gives an unpolarized incident beam to which corresponds a scattered beam whose horizontal and vertical intensities of vibration are $H_u = H_h + H_v$ and $V_u = V_h + V_v$. The depolarization factor of this scattered beam is defined by the ratio

$$\rho_u = H_u/V_u$$

and has thence the value

$$\rho_u = (H_h + H_v)/(V_h + V_v). \tag{2}$$

The measurements of the three depolarization factors ρ_v, ρ_h, and ρ_u thus give the ratios of the four quantities H_h, V_h, H_v, V_v.

Using a general law of reciprocity due to Lord Rayleigh, R. S. Krishnan obtained the relation

$$H_v = V_h \quad (3)$$

for any symmetrical isotropic medium, and proved that this relation must be true also for a medium with only axial symmetry around the vertical direction perpendicular to the plane of scattering.[2]

This relation is verified in two particular cases already known: (1) For very small scattering particles (dipolar scattering) V_v, H_v, V_h are independent of ϕ, and (Lord Rayleigh)[6]

$$H_v = V_h,$$
$$H_h = V_v \cos^2 \phi + H_v \sin^2 \phi,$$

which gives for transverse scattering

$$H_v = V_h = H_h \quad \text{(dipoles, } \phi = \pi/2). \quad (4)$$

(2) For spherical scattering particles of any dimension, and for all values of ϕ (G. Mie)[7]

$$H_v = V_h = 0 \quad \text{(spheres)}. \quad (5)$$

R. S. Krishnan gave, moreover, the experimental proof of Eq. (3) for various non-spherical large particles, for which the observed intensities H_v and V_h are always equal, but generally different from 0 and from H_h.

From the Eqs. (1), (2), and (3), the relation (6) results:

$$\rho_u = (1 + 1/\rho_h)/(1 + 1/\rho_v). \quad (6)$$

As a consequence of reciprocity it is thus unnecessary to measure the depolarization factor ρ_u for unpolarized excitation, if the depolarization factors ρ_h and ρ_v for horizontally and vertically polarized excitation have been measured.

Finally, in a paper published in November 1939, R. S. Krishnan[8] considered the case in which the direction of vibration of the linearly polarized incident beam makes an angle θ with the normal to the plane of scattering. Neglecting the correlation of phase which then exists between the horizontal and vertical components of vibration of the incident beam, he obtained for the ratio of the intensities of the horizontal and vertical components of vibration of the scattered light the equation,

$$\rho_\theta = H_\theta / V_\theta = (1 + \mathrm{tg}^2\, \theta/\rho_h)/(\mathrm{tg}^2\, \theta + 1/\rho_v). \quad (7)$$

This equation, though in agreement with the particular theoretical results of Lord Rayleigh (any small particles) and of G. Mie (large spheres) did not seem to him to be generally true, because of the arbitrariness of the hypothesis he had made to obtain it. He reported even a few measurements he had made of the scattering by large non-spherical particles showing some disagreement with it. However, we shall show that this equation must be valid for any symmetrical medium.

III. THE LAW OF RECIPROCITY IN OPTICS

In his book on the *Theory of Sound*, Lord Rayleigh established a theorem of reciprocity for the forces and displacements in the neighborhood of an equilibrium state of a mechanical system governed by linear equations.[9] Later on he extended to optics, without a new demonstration, the law of reciprocity; he merely indicated in a footnote the necessity of specifying the states of polarization.[10] R. S. Krishnan referred to this statement by Lord Rayleigh of the theorem of reciprocity to establish, in the case of light scattering, the relation we have given above. But the conditions he considered are particular, because he did not take into account the possible correlation of phases between the two components of vibration of each beam of light.

To apply the law of reciprocity to the most general phenomenon of scattering by an isotropic medium, it is necessary to start from a precise statement of this law.

Any monochromatic beam of light may be considered, in an infinite number of ways, as the superposition, with more or less phase correlation, of two completely polarized beams of complementary characters, for instance rectangular linear polarizations, or inverse circular polarizations. We shall choose as reference polarization states, for each direction of propagation, the states of linear polarization along two fixed rectangular axes.

Let us consider, given any system in which light can be scattered and absorbed, for an incident linearly polarized beam F_1 having an intensity I_1, coming from a linear polarizer N, a particular emerging beam from which we may separate a linearly polarized component F'_1, having an intensity I'_1, by means of a linear polarizer N'. Let us associate with these beams the inverse beams, that is to say, an incident polarized beam F_2 coming from the polarizer N', with an intensity equal to I_1, in the direction opposite to that of the emerging beam F'_1, and the corresponding emerging beam F'_2 coming out of the polarizer N in the direction opposite to that of the incident beam F_1. The law of reciprocity states that the intensity I'_2 of this last beam F'_2 is equal to the intensity I'_1 of the beam F'_1: *If two incident polarized beams have equal intensities, the inverse emerging beams of the same polarization, which are associated with them, also have equal intensities.*

This law is true only if the considered optical system is not affected by a reversal of time, so that the sense of propagation of light be immaterial. There must be no movements, no electrical currents, no magnetic fields. To extend it to more general cases it is necessary to reverse, together with the direction of light propagation, all movements, electrical currents, and magnetic fields. For instance, it is well known that the law of reciprocity, as stated above, is not true for a system in which magnetic rotatory power comes into account, if the magnetic fields are not reversed with the sense of propagation of light.

Moreover, only monochromatic beams of the same frequency must be considered. At least it is necessary that the mechanisms which modify the frequency can be reversed with the propagation of light, as for instance a change of frequency caused by scattering by a moving body. The law of reciprocity is not valid for fluorescence or for Raman effect, in which the change of frequency is irreversible. In scattering phenomenon it is only relevant for Rayleigh scattering, with no or small symmetrical frequency changes.

It is also interesting to give the corpuscular statement of the law of reciprocity: If a photon associated with the incident polarized beam F_1 has a probability p to come out of the optical system associated with the polarized beam F'_1, then inversely, a photon associated with the beam F_2, reverse, to F'_1, has the same probability p to come out associated with the beam F'_2 reverse to F_1.

The law of reciprocity is thus seen to be connected with the general principle of quantum mechanics asserting the equal probability of inverse transitions between two states of the same energy.

IV. STOKES' LINEAR REPRESENTATION OF STATES OF POLARIZATION

Let us first consider a completely polarized monochromatic beam of light, whose electrical vibration may be represented by its components along two rectangular axes

$$E_x = p_1 \cos(\omega t + \varphi_1),\quad E_y = p_2 \cos(\omega t + \varphi_2), \tag{8}$$

the amplitudes p_1, p_2, and the frequency $\omega/2\pi$ being positive. Let δ be the phase difference of these components

$$\delta = \varphi_1 - \varphi_2 \tag{9}$$

and I_e the total intensity of vibration

$$I_e = p_1^2 + p_2^2. \tag{10}$$

The terminal point of the oscillating vector **E** thus specified, describes, in the direct or reverse sense according to the positive or negative sign of $\sin\delta$, an ellipse with semi-axes a and b $(b \leqslant a)$, whose major axis makes an angle α with the x axis. Let us set

$$\operatorname{tg}\beta = \pm b/a, \quad -\pi/4 \leqslant \beta \leqslant \pi/4, \tag{11}$$

taking the sign + or − according to the sense of rotation, i.e., so that always

$$\operatorname{tg}\beta \sin\delta > 0.$$

With such definitions, it is known that

$$\begin{aligned} p_1^2 - p_2^2 &= I_e \cos 2\beta \cos 2\alpha,\\ 2p_1p_2 \cos\delta &= I_e \cos 2\beta \sin 2\alpha,\\ 2p_1p_2 \sin\delta &= I_e \sin 2\beta. \end{aligned} \tag{12}$$

These three quantities, which we shall name M_e, C_e, S_e, determine the elliptic vibration (except its phase). In Poincaré's representation they are considered as the rectangular components of a vector in space, whose length is I_e, longitude 2α, and latitude 2β.

No actual light is strictly monochromatic. The amplitudes and phases of the components of any light vibration undergo slow variations without strict correlation. The ellipse of vibration, which is still determined at each moment, changes its shape and magnitude, slowly in comparison with the period of vibration but extremely swiftly in comparison with the duration of any measurement. It is thus possible to measure only mean values.

The study of the polarization of a light beam requires the use of analyzers, each giving the mean intensity of a vibration E_a obtained as a linear combination, with given changes in phase, of the two components E_x and E_y of the initial vibration

$$E_a = c_1 p_1 \cos(\omega t + \varphi_1 + \eta_1) + c_2 p_2 \cos(\omega t + \varphi_2 + \eta_2). \quad (13)$$

This mean intensity has the value

$$\begin{aligned} I_a = {} & \tfrac{1}{2}(c_1^2 + c_2^2)(\langle p_1^2\rangle_{\mathrm{Av}} + \langle p_2^2\rangle_{\mathrm{Av}}) \\ & + \tfrac{1}{2}(c_1^2 - c_2^2)(\langle p_1^2\rangle_{\mathrm{Av}} - \langle p_2^2\rangle_{\mathrm{Av}}) \\ & + c_1 c_2 \cos(\eta_1 - \eta_2)\langle 2p_1p_2 \cos\delta\rangle_{\mathrm{Av}} \\ & - c_1 c_2 \sin(\eta_1 - \eta_2)\langle 2p_1p_2 \sin\delta\rangle_{\mathrm{Av}}, \end{aligned}$$

the notation $\langle\ \rangle_{\mathrm{Av}}$ denoting the mean value with respect to time. By the use of four different (linearly independent) analyzers it is thus possible to calculate the quantities

$$\begin{aligned} I &= \langle p_1^2\rangle_{\mathrm{Av}} + \langle p_2^2\rangle_{\mathrm{Av}} = \langle I_e\rangle_{\mathrm{Av}}, \\ M &= \langle p_1^2\rangle_{\mathrm{Av}} - \langle p_2\rangle_{\mathrm{Av}} = \langle M_e\rangle_{\mathrm{Av}}, \\ C &= 2\langle p_1p_2 \cos\delta\rangle_{\mathrm{Av}} = \langle C_e\rangle_{\mathrm{Av}}, \\ S &= 2\langle p_1p_2 \sin\delta\rangle_{\mathrm{Av}} = \langle S_e\rangle_{\mathrm{Av}}, \end{aligned} \quad (14)$$

and if these four quantities are known, it is possible to calculate the intensity that will be measured with any analyzer corresponding to certain values of the coefficients c_1 and c_2 and of the phase shift $(\eta_1 - \eta_2)$. That is to say, the four quantities I, M, C, S give a complete description of the polarization properties of the light beam (Stokes).[11]

It is easy to prove that for any beam of light the parameters I, M, C, S, verify the inequality

$$I \geqslant (M^2 + C^2 + S^2)^{\frac{1}{2}}, \quad (15)$$

since the equality is true only for completely polarized light; and if four quantities satisfy this condition they may be considered the polarization parameters of a light beam.

Any light beam, having a partial polarization specified by the values I, M, C, S of the Stokes' parameters, may be considered as the superposition, without any phase correlation, of a beam of natural light having an intensity

$$I_N = I - (M^2 + C^2 + S^2)^{\frac{1}{2}} \quad (16)$$

and of a beam of completely polarized elliptic light having an intensity

$$I_E = (M^2 + C^2 + S^2)^{\frac{1}{2}} \quad (17)$$

and whose ellipse of vibration is defined by the angles α and β given by the relations

$$\begin{aligned} I_E \cos 2\beta \cos 2\alpha &= M, \\ I_E \cos 2\beta \sin 2\alpha &= C, \\ I_E \sin 2\beta &= S. \end{aligned} \quad (18)$$

The ratio

$$p = I_E / I = (M^2 + C^2 + S^2)^{\frac{1}{2}} / I, \quad (0 \leqslant p \leqslant 1) \quad (19)$$

is called the degree of polarization.

The essential property of the Stokes' parameters is their additivity in the superposition of two independent beams of light, i.e., without any correlation between the perturbations of their phases or amplitudes. This additivity corresponds to the absence of any interference.

When a beam of light passes through some optical arrangement, or more generally, produces a secondary beam of light, the intensity and the state of polarization of the emergent beam are functions of those of the incident beam. If two independent incident beams are superposed the new emergent beam will be, if the process is linear, the superposition without interference of the two emergent beams corresponding to the separate incident beams. Consequently, in such a linear process, from the additivity properties of the Stokes' parameters, the parameters I', M', C', S' which define the polarization of the emergent beam must be homogeneous linear functions of the parameters I, M, C, S corresponding to the incident beam; the sixteen coefficients of these linear functions will completely characterize the corresponding optical phenomenon. This fundamental remark is due in its general formulation to P. Soleillet.[12] We shall use it in the next section in the study of scattering. Let us give here only the linear transformation formulas of the Stokes' parameters in two simple cases which we shall have to consider:

When the light beam is rotated through an angle ψ around its direction of propagation, for instance by passing through a crystal plate with simple rotatory power, we have

$$\begin{aligned} I' &= I, \\ M' &= M \cos 2\psi - C \sin 2\psi, \\ C' &= M \sin 2\psi + C \cos 2\psi, \\ S' &= S, \end{aligned} \tag{20}$$

and these equations also give the transformation of the Stokes' parameters when the reference axes are rotated through an angle $-\psi$.

When a difference in phase φ is introduced between the components of the vibration along the axes, for instance by a birefringent crystal plate with axes parallel to the reference axes (axes of maximum speed along Ox for $\varphi > 0$), we have

$$\begin{aligned} I' &= I, \\ M' &= M, \\ C' &= C \cos \varphi - S \sin \varphi, \\ S' &= C \sin \varphi + S \cos \varphi. \end{aligned} \tag{21}$$

It is interesting to note how the method used by Stokes' to characterize a state of polarization may be generalized and connected with the wave statistics of J. von Neumann:[13] Let us consider a system of n harmonic oscillations of the same frequency subjected to small random perturbations; we may represent them by complex expressions

$$E_k = P_k \exp (i\omega t), \quad P_k = p_k \exp (i\varphi_k), \tag{22}$$

the modulus p_k and the arguments φ_k varying in course of time, slowly in comparison with the period of oscillation, but quickly in comparison with the duration of any measurement. Let us suppose that we can measure the mean intensity of an oscillation E linearly dependent on these oscillations

$$E = \sum_k C_k E_k, \quad C_k = c_k \exp (i\eta_k). \tag{23}$$

The value of this mean intensity is (the asterisk indicating the change to the complex conjugate quantity)

$$\langle EE^* \rangle_{\text{Av}} = \sum_{kl} C_k C_l^* \langle P_k P_l^* \rangle_{\text{Av}}. \tag{24}$$

The mean intensity depends on the particular oscillations involved only through the von Neumann's matrix

$$\Gamma_{kl} = \langle P_k P^*_l \rangle_{\text{Av}}, \tag{25}$$

the knowledge of which determines all we can know about these oscillations by such measurements. Since this matrix is hermitic, we can set

$$\Gamma_{kk} = \mu_k, \quad \Gamma_{kl} = \gamma_{kl} + i\sigma_{kl}, \quad (k \neq l) \tag{26}$$

μ_k, $\gamma_{kl} = \gamma_{lk}$, $\sigma_{kl} = -\sigma_{lk}$ being real quantities. The diagonal terms μ_k are the mean intensities of the oscillations:

$$\mu_k = \langle p_k^2 \rangle_{\text{Av}} \tag{27}$$

and the other terms give the correlations between the oscillations:

$$\begin{aligned} \gamma_{kl} &= \langle p_k p_l \cos (\varphi_k - \varphi_l) \rangle_{\text{Av}}, \\ \sigma_{kl} &= \langle p_k p_l \sin (\varphi_k - \varphi_l) \rangle_{\text{Av}}. \end{aligned} \tag{28}$$

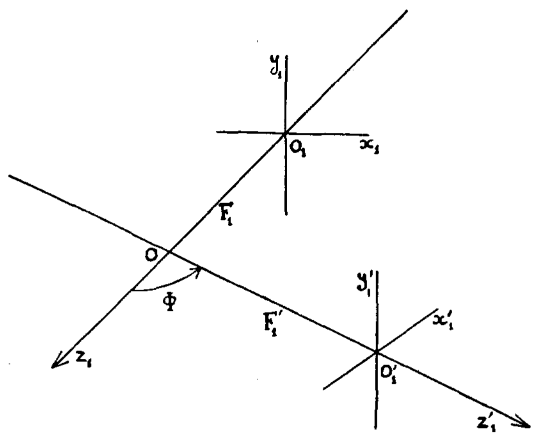
The state of excitation of n oscillators having the same frequency is thus defined by n^2 real quantities. For instance, a vectorial vibration in space having three components must be defined by *nine* quantities (P. Soleillet),[12] and the possibilities of interference between two beams of light of the same frequency depend on *sixteen* parameters, since there are four components (two for each beam).

When the different oscillations are the components of an oscillating vector $\mathbf{P} \exp (i\omega t)$, the matrix Γ_{kl} has, for any change of the reference axes, the variance of a tensor of the second order, since its elements are the mean values of the products of the components of two vectors ($\mathbf{P}$ and $\mathbf{P}^*$).

It is easy to prove that the determinant of the matrix Γ_{kl} and all its diagonal minors are always positive or zero.

When the ratios of the P_k's are independent of time, the oscillation state of the system is said to be *pure* (complete polarization in the case of a light beam); all the diagonal minors of the matrix Γ_{kl} are then zero, and conversely. When it is not so, the state is said to be *mixed* (partial polarization). In general, any mixed state of oscillation of a system of n oscillators may be considered, in an infinite number of ways, as the superposition of n pure states without correlation (for a state to be equivalent to the superposition of less pure states, it is necessary that the determinant of the Γ_{kl} be zero).

Still more generally, it is possible, in a similar way, to find the quantities which will appear in the linear investigation of a non-harmonic system whose motion is described by n statistical func-



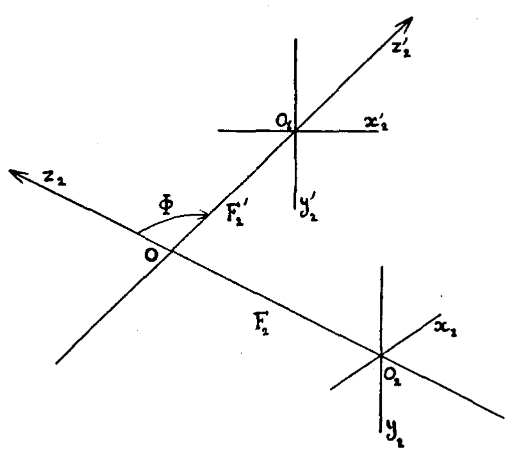


FIG. 2. (Above.) FIG. 3. (Below.)

tions of the time $E_k(t)$: These will be the *correlation functions* of M. Courtines[14] and J. Bernamont[15] defined by the relation

$$f_{kl}(\tau)=\langle E_k(t)E_l(t+\tau)\rangle_{\text{Av}}=f_{lk}(-\tau) \tag{29}$$

or the functions corresponding to them by the Laplacian transformation

$$\Gamma_{kl}(\nu)=\int_{-\infty}^{+\infty} f_{kl}(\tau)\exp(-2\pi i\nu\tau)d\tau, \tag{30}$$

which are the amplitudes of their representation by Fourier's integrals

$$f_{kl}(\tau)=\int_{-\infty}^{+\infty} \Gamma_{kl}(\nu)\exp(2\pi i\nu\tau)d\nu \tag{31}$$

and verify the condition of hermiticity

$$\Gamma_{lk}(\nu)=\Gamma_{kl}(\nu)^*. \tag{32}$$

It is thus seen that the state of motion of a system having n degrees of freedom is characterized, when linear methods of analysis are used, by n^2 continuous spectra giving, for each frequency, n spectral densities of intensity and $n(n-1)$ spectral densities of correlation.

V. THE SIXTEEN SCATTERING COEFFICIENTS OF AN ARBITRARY ISOTROPIC MEDIUM

If a monochromatic parallel beam of light F_1 is propagated along an axis O_1z_1, we consider the light scattered at a point O of this axis in a direction Oz'_1, making an angle ϕ (between 0 and π) with Oz_1. Let us specify the state of polarization of the incident beam F_1 by the values I_1, M_1, C_1, S_1 of its Stokes' parameters for the axes $O_1x_1y_1$, and that of the scattered beam F'_1 by the values I'_1, M'_1, C'_1, S'_1 of its Stokes' parameters for the axes $O'_1x'_1y'_1$. The two sets of rectangular axes $O_1x_1y_1z_1$ and $O'_1x'_1y'_1z'_1$ are right handed; the planes $O_1z_1x_1$ and $O'_1z'_1x'_1$ coincide; and the parallel axes O_1y_1 and $O'_1y'_1$ are orientated so that the rotation ϕ smaller than π which brings Oz_1 on Oz'_1 is positive around their common direction (Fig. 2).

We assume the linear character of light scattering for the superposition of non-coherent beams: If two independent light beams F and G which can be propagated along O_1z_1 give, when separate, the scattered beams F' and G' along Oz'_1, the scattered beam when the incident beams F and G are superposed without any phase relation, may be obtained by the superposition without phase relation of the two beams F' and G'. Since the Stokes' parameters are additive for the superposition of non-coherent beams, this requires that the quantities I'_1, M'_1, C'_1, S'_1 be linear homogeneous functions of the quantities I_1, M_1, C_1, S_1, i.e., that

$$\begin{aligned} I'_1&=a_{11}I_1+a_{12}M_1+a_{13}C_1+a_{14}S_1,\\ M'_1&=a_{21}I_1+a_{22}M_1+a_{23}C_1+a_{24}S_1,\\ C'_1&=a_{31}I_1+a_{32}M_1+a_{33}C_1+a_{34}S_1,\\ S'_1&=a_{41}I_1+a_{42}M_1+a_{43}C_1+a_{44}S_1. \end{aligned} \tag{33}$$

If the scattering is produced by an isotropic medium, the sixteen *scattering coefficients* a_{ik} will depend only, for a given medium, on the frequency of the light and on the scattering angle ϕ.

We shall see in the next sections that symmetry and reciprocity arguments prove that the number of independent scattering coefficients is actually less than sixteen.

VI. SYMMETRICAL MEDIUM

If the scattering medium is symmetrical, and consequently without any rotatory power, the plane of scattering $Oz_1z'_1$ determined by the directions of excitation and observation, is a plane of symmetry for the medium. Therefore, if the incident beam (I_1, M_1, C_1, S_1) is replaced by the symmetrical beam with respect to this plane, a beam whose parameters are (I_1, M_1, $-C_1$, $-S_1$), the new scattered beam must be symmetrical to the first scattered beam, and hence its parameters will be (I'_1, M''_1, $-C'_1$, $-S'_1$). In other words the relations (33) must hold if the sign of the parameters C_1, S_1, C'_1, S'_1 is changed whatever the values of I_1, M_1, C_1, S_1. This requires that for an isotropic symmetrical medium

$$a_{13}=a_{14}=a_{23}=a_{24}=a_{31}=a_{32}=a_{41}=a_{42}=0,$$

and consequently that for such a medium the relations (33) reduce to

$$\begin{aligned} I'_1&=a_{11}I_1+a_{12}M_1,\\ M'_1&=a_{21}I_1+a_{22}M_1,\\ C'_1&=a_{33}C_1+a_{34}S_1,\\ S'_1&=a_{43}C_1+a_{44}S_1. \end{aligned} \tag{33A}$$

For a symmetrical medium the number of scattering coefficients is only eight.

VII. RECIPROCITY

We must now apply the law of reciprocity, and for this purpose consider an incident beam being propagated along the axis Oz_2 opposite to the direction Oz'_1 of the first scattered beam, and the corresponding scattered beam F'_2 in the direction Oz'_2 opposite to the direction Oz_1 of the first incident beam (Fig. 3). We shall take as reference axes for the states of polarization of these new beams two sets of axes having, with respect to F_2 and F'_2 the same orientation as the axes previously used with respect to F_1 and F'_1: For beam F_2 the axis O_2x_2 in coincidence with $O'_1x'_1$ and the axis O_2y_2 opposite to $O'_1y'_1$, for beam F'_2 the axis $O'_2x'_2$ in coincidence with O_1x_1 and the axis $O'_2y'_2$ opposite to O_1y_1. With these reference axes the Stokes' parameters I'_2, M'_2, C'_2, S'_2 of the scattered beam F'_2 will be expressed as linear functions of the parameters I_2, M_2, C_2, S_2 of the incident beam F_2 by relations identical to the relations (33):

$$\begin{aligned} I'_2&=a_{11}I_2+a_{12}M_2+a_{13}C_2+a_{14}S_2,\\ M'_2&=a_{21}I_2+a_{22}M_2+a_{23}C_2+a_{24}S_2,\\ C'_2&=a_{31}I_2+a_{32}M_2+a_{33}C_2+a_{34}S_2,\\ S'_2&=a_{41}I_2+a_{42}M_2+a_{43}C_2+a_{44}S_2. \end{aligned} \tag{34}$$

The coefficients a_{ik} have the same values, since the scattering angle has the same value ϕ and since the medium is supposed to be isotropic.*

To be able to express reciprocity, we must introduce on the paths of the incident and scattered beams some polarizers transforming an initial beam having a fixed linear polarization into an incident beam of any elliptical polarization, and any component of the scattered beam into a final beam having, like the initial beam a fixed linear polarization.

For this purpose we may introduce on the path of the incident beam F_1: (1) a linear polarizer N with fixed orientation such that the electrical vibration of the light emerging from it is along O_1x_1, this light having thus the Stokes' parameters (1, 1, 0, 0) if its intensity is unity; (2) a crystal plate R with rotatory power turning the light vibration through an angle ψ; according to formulas (20) the Stokes' parameters of the beam after this plate will be (1, cos 2ψ, sin 2ψ, 0); (3) a birefringent crystal plate B with its axes parallel to the reference axes $O_1x_1y_1$ and producing a change in phase φ between the vibrations along O_1x_1 and O_1y_1. The incident beam thus obtained may have any complete elliptical polarization; its Stokes' parameters will be, according to Eq. (21),

$$\begin{aligned} I_1&=1,\\ M_1&=\cos 2\psi,\\ C_1&=\sin 2\psi\cos\varphi,\\ S_1&=\sin 2\psi\sin\varphi. \end{aligned} \tag{35}$$

* It is even sufficient that the direction Oz_1 and Oz_2 be equivalent in the scattering medium, which might, for instance, have only axial symmetry around the perpendicular Oy to the scattering plane.

Similarly, on the path of the scattered beam F'_1 whose Stokes' parameters I'_1, M'_1, C'_1, S'_1 will be obtained by relations (33) in which I_1, M_1, C_1, S_1 have the values given by (35), we shall introduce: (1) a birefringent crystal plate B' with its axes parallel to the axes $O'_1x'_1y'_1$ and producing a change in phase φ' between the vibrations along $O'_1x'_1$ and $O'_1y'_1$; after this plate the Stokes' parameters of the beam will be (Eq. 21)

$$\begin{aligned}&I'_1,\\&M'_1,\\&C'_1\cos\varphi'-S'_1\sin\varphi',\\&C'_1\sin\varphi'+S'_1\cos\varphi';\end{aligned}$$

(2) a crystal plate R' with rotatory power turning the light vibration through an angle ψ'; after this plate the Stokes' parameters will become (Eq. 20)

$$\begin{aligned}&I'_1,\\&M'_1\cos 2\psi'-(C'_1\cos\varphi'-S'_1\sin\varphi')\sin 2\psi',\\&M'_1\sin 2\psi'+(C'_1\cos\varphi'-S'_1\sin\varphi')\cos 2\psi',\\&C'_1\sin\varphi'+S'_1\cos\varphi';\end{aligned}$$

(3) a linear polarizer N' with fixed orientation selecting the electrical vibration along $O'_1x'_1$. The intensity J_1 of the light coming out of this polarizer will be equal to half the sum of the first two Stokes' parameters of the light entering it; thus

$$J_1=\tfrac{1}{2}[I'_1+M'_1\cos 2\psi'-C'_1\sin 2\psi'\cos\varphi'+S'_1\sin 2\psi'\sin\varphi']. \quad (36)$$

Let us consider now the light being propagated in the reverse direction through this arrangement, when the intensity of the beam F_2 coming out of the polarizer N' toward the scattering medium has an intensity equal to unity. The values of the Stokes' parameters are then for this beam (1, 1, 0, 0) and after the plates R' and B' they will be

$$\begin{aligned}&I_2=1,\\&M_2=\cos 2\psi',\\&C_2=\sin 2\psi'\cos\varphi',\\&S_2=\sin 2\psi'\sin\varphi'.\end{aligned} \quad (37)$$

The parameters I'_2, M'_2, C'_2, S'_2 of the corresponding scattered beam will be given by Eq. (34) I_2, M_2, C_2, S_2 having the values (37), and the intensity of the light coming out of the arrangement through the polarizer N, obtained as for the first scattered beam, will be

$$J_2=\tfrac{1}{2}[I'_2+M'_2\cos 2\psi-C'_2\sin 2\psi\cos\varphi+S'_2\sin 2\psi\sin\varphi]. \quad (38)$$

Since the incident beams F_1 and F_2 have the same intensity, and the initial and final states of polarization are the same complete linear polarization, the intensities J_1 and J_2 of the emergent beams must be equal, according to the law of reciprocity, whatever crystal plates are introduced in the path of the light.

We have, consequently,

$$I'_1+M'_1\cos 2\psi'-C'_1\sin 2\psi'\cos\varphi'+S_1'\sin 2\psi'\sin\varphi'=I'_2+M'_2\cos 2\psi-C'_2\sin 2\psi\cos\varphi+S'_2\sin 2\psi\sin\varphi, \quad (39)$$

whatever the values of the angles ψ, φ, ψ', φ'. Using Eqs. (33), (34), (35), and (37), we obtain

$$\begin{aligned}&(a_{12}-a_{21})(\cos 2\psi-\cos 2\psi')\\&+(a_{13}+a_{31})(\sin 2\psi\cos\varphi-\sin 2\psi'\cos\varphi')\\&+(a_{14}-a_{41})(\sin 2\psi\sin\varphi-\sin 2\psi'\sin\varphi')\\&+(a_{23}+a_{32})(\sin 2\psi\cos\varphi\cos 2\psi'-\sin 2\psi'\cos\varphi'\cos 2\psi)\\&+(a_{24}-a_{42})(\sin 2\psi\sin\varphi\cos 2\psi'-\sin 2\psi'\sin\varphi'\cos 2\psi)\\&-(a_{34}+a_{43})\sin(\varphi-\varphi')\sin 2\psi\sin 2\psi'=0.\end{aligned} \quad (40)$$

This identity can be maintained only if all the coefficients of the trigonometrical expressions in it are zero: For $\psi=0$, $\psi'=\pi/2$ it reduces to

$$a_{12}-a_{21}=0; \quad (41)$$

for $\psi=-\psi'=\pi/4$, $\varphi=\varphi'=0$, to

$$a_{13}+a_{31}=0; \quad (42)$$

for $\psi=-\psi'=\pi/4$, $\varphi=\varphi'=\pi/2$, to

$$a_{14}-a_{41}=0. \quad (43)$$

After the suppression of the three first terms in the identity (40), as a consequence of the zero value of their coefficients, the new identity obtained reduces, for $\psi=\pi/4$, $\psi'=\varphi=\varphi'=0$ to

$$a_{23}+a_{32}=0; \quad (44)$$

for $\psi=\pi/4$, $\psi'=0$, $\varphi=\varphi'=\pi/2$, to

$$a_{24}-a_{42}=0; \quad (45)$$

and for $\psi=\psi'=\pi/4$, $\varphi=\pi/2$, $\varphi'=0$, to

$$a_{34}+a_{43}=0. \quad (46)$$

The sixteen scattering coefficients must therefore obey six conditions of symmetry or antisymmetry, and the linear relations which give the Stokes' parameters of the scattered beam in terms of those of the incident beam, may be written

$$\begin{aligned} I' &= a_1 I + b_1 M - b_3 C + b_5 S, \\ M' &= b_1 I + a_2 M - b_4 C + b_6 S, \\ C' &= b_3 I + b_4 M + a_3 C + b_2 S, \\ S' &= b_5 I + b_6 M - b_2 C + a_4 S. \end{aligned} \tag{47}$$

The scattering of light, through a given angle, by an asymmetrical isotropic medium is characterized by ten independent coefficients.

If the scattering medium is symmetrical, and has therefore no rotatory power, the coefficients b_3, b_4, b_5, b_6 are necessarily zero, since in this case the relations (47) must have the form of the relations (33A), and the linear relations between the Stokes' parameters are reduced to

$$\begin{aligned} I' &= a_1 I + b_1 M, \\ M' &= b_1 I + a_2 M, \\ C' &= a_3 C + b_2 S, \\ S' &= -b_2 C + a_4 S. \end{aligned} \tag{47A}$$

The scattering of light, through a given angle, by a symmetrical isotropic medium is characterized by six independent coefficients.

R. S. Krishnan in his publications dated 1938[1–5] considered only incident beams with linear polarization either horizontal ($I=1$, $M=1$, $C=S=0$) or vertical ($I=1$, $M=-1$, $C=S=0$). These excitation conditions involve only the four coefficients a_{11}, a_{12}, a_{21}, a_{22}, and the relation obtained by him $H_v = V_h$ is equivalent to the first relation proved here $a_{12}=a_{21}$.

In the more general case considered in his paper of 1939, the incident beam has a linear polarization in a direction making an angle θ with the perpendicular to the scattering plane. The Stokes' parameters of the incident beam are then

$$I=1, \quad M=-2\cos 2\theta, \quad C=\sin 2\theta, \quad S=0$$

and, applying Eq. (47) we obtain

$$\rho_\theta = \frac{H_\theta}{V_\theta} = \frac{I'+M'}{I'-M'} = \frac{a_1 + b_1(1-\cos 2\theta) - a_2\cos 2\theta - (b_3+b_4)\sin 2\theta}{a_1 - b_1(1+\cos 2\theta) + a_2\cos 2\theta - (b_3-b_4)\sin 2\theta}.$$

When θ is 0 or $\pi/2$ this formula gives

$$\rho_v = \frac{H_v}{V_v} = \frac{a_1-a_2}{a_1+2b_1+a_2}, \quad \rho_h = \frac{V_h}{H_h} = \frac{a_1-a_2}{a_1-2b_1+a_2},$$

which shows that the expression of ρ_θ may be written

$$\rho_\theta = \frac{1+\mathrm{tg}^2\,\theta/\rho_h - 2(b_3+b_4)(a_1-a_2)^{-1}\,\mathrm{tg}\,\theta}{\mathrm{tg}^2\,\theta + 1/\rho_v - 2(b_3-b_4)(a_1-a_2)^{-1}\,\mathrm{tg}\,\theta}.$$

This proves that Eq. (7) proposed with some doubt by R. S. Krishnan, must be true for any symmetrical medium, since then $b_3=b_4=0$ but not for an asymmetrical medium.

VIII. FORWARD AXIAL SCATTERING

The light scattered in the same direction as the incident beam cannot be distinguished from the remaining light of this beam. However, if the incident beam has a small aperture ω, it is possible to define, as a limit when $\omega \to 0$, a transmitted beam T, with axis Oz_1 and aperture ω, and a scattered beam including all rays between two cones of apertures η ($\eta > \omega$) and $\eta + d\eta$, having also Oz_1 as axis. The scattered beam F'_0 thus defined may vary with the small angle η, but whatever this angle, it will have axial symmetry around Oz_1.

The law of reciprocity applies to the transmitted beam T as well as to the axially scattered beam F'_0. If reference axes parallel to those chosen for the incident beam are used for the polarization states of these beams, the linear relations between Stokes' parameters will have in both cases, the form obtained above. Writing them for beam F'_0, we will indicate the zero value of the scattering angle ϕ by marking the coefficients with a superscript 0

$$\begin{aligned} I' &= a_1^0 I + b_1^0 M - b_3^0 C + b_5^0 S, \\ M' &= b_1^0 I + a_2^0 M - b_4^0 C + b_6^0 S, \\ C' &= b_3^0 I + b_4^0 M + a_3^0 C + b_2^0 S, \\ S' &= b_5^0 I + b_6^0 M - b_2^0 C + a_4^0 S. \end{aligned} \tag{48}$$

In this case we must also express the axial symmetry around the common direction of the incident and scattered beams: If the incident beam is turned around itself through any angle, the axially scattered beam F'_0 will simply turn around itself through the same angle. It is sufficient to consider an infinitesimal rotation

$\frac{1}{2}d\alpha$ which changes the beam (I, M, C, S) into a beam F_1 with parameters

$$\begin{aligned} I_1&=I,\\ M_1&=M-Cd\alpha,\\ C_1&=Md\alpha+C,\\ S_1&=S, \end{aligned} \tag{49}$$

and the beam $F'_0(I', M', C', S')$ into a beam F'_{01} with the parameters

$$\begin{aligned} I'_1&=I',\\ M'_1&=M'-C'd\alpha,\\ C'_1&=M'd\alpha+C',\\ S'_1&=S'. \end{aligned} \tag{50}$$

The parameters I'_1, M'_1, C'_1, S'_1 must be expressed in terms of I_1, M_1, C_1, S_1 by the relations identical to relations (48). Eliminating I, M, C, S and I', M', C', S' between the relations (48), (49), and (50), we obtain

$$\begin{aligned} I'_1&=a_1{}^0I_1+(b_1{}^0+b_3{}^0d\alpha)M_1\\ &\quad-(b_3{}^0-b_1{}^0d\alpha)C_1+b_5{}^0S_1,\\ M'_1&=(b_1{}^0-b_3{}^0d\alpha)I_1+a_2{}^0M_1\\ &\quad-[b_4{}^0+(a_3{}^0-a_2{}^0)d\alpha]C_1\\ &\quad+(b_6{}^0-b_2{}^0d\alpha)S_1,\\ C'_1&=(b_3{}^0+b_1{}^0d\alpha)I_1+(b_4{}^0-(a_3{}^0-a_2{}^0)d\alpha]M_1\\ &\quad+a_3{}^0C_1+(b_2{}^0+b_6{}^0d\alpha)S_1,\\ S_1'&=b_5{}^0I_1+(b_6{}^0+b_2{}^0d\alpha)M_1\\ &\quad-(b_2{}^0-b_6{}^0d\alpha)C_1+a_4{}^0S_1. \end{aligned} \tag{51}$$

So that this linear system be identical with system (48) it is necessary and sufficient that

$$b_1{}^0=b_2{}^0=b_3{}^0=b_6{}^0=0, \quad a_2{}^0=a_3{}^0.$$

The linear transformation for the Stokes' parameter is thus, for axial scattering:

$$\begin{aligned} I'&=a_1{}^0I+b_5{}^0S,\\ M'&=a_2{}^0M-b_4{}^0C,\\ C'&=b_4{}^0M+a_2{}^0C,\\ S'&=b_5{}^0I+a_4{}^0S. \end{aligned} \tag{53}$$

These equations apply to any asymmetrical medium. They show that forward axial scattering involves in general *five* independent coefficients.

For a symmetrical medium $b_4{}^0$ and $b_5{}^0$ are necessarily zero; therefore, the linear transformation is reduced to

$$\begin{aligned} I'&=a_1{}^0I,\\ M'&=a_2{}^0M,\\ C'&=a_2{}^0C,\\ S'&=a_4{}^0S. \end{aligned} \tag{53A}$$

The forward axial scattering involves in this case only *three* independent coefficients.

For the transmitted beam T the linear transformation will have also the form (53) or (53A) but with other values of the coefficients, which will in this case correspond to simple classical properties: The quantity

$$1-a_1{}^0$$

will measure the absorption for natural light; the quantity

$$b_4{}^0 : a_2{}^0$$

the tangent of the rotation (rotatory power); the quantity

$$1-(a_2{}^{02}+b_4{}^{02}+b_6{}^{02})^{\frac{1}{2}} : a_1{}^0$$

the depolarization for linear light; the quantity

$$1-a_4{}^0 : a_1{}^0$$

the depolarization for circular light; and the quantity

$$b_5{}^0 : a_1{}^0$$

the circular dichroism.

IX. BACKWARD AXIAL SCATTERING

A similar argument may be applied to the scattering in the direction opposite to that of the incident beam, since there is also in this case axial symmetry. But then any rotation of the incident beam around its direction must produce an *inverse* rotation of the same magnitude of the scattered beam around its direction.

Marking with a superscript π the coefficients of the linear transformation (47) for the value $\phi=\pi$ which correspond to backward scattering, the invariance for an infinitesimal rotation leads to the conditions

$$b_1{}^\pi=b_2{}^\pi=b_3{}^\pi=b_4{}^\pi=b_6{}^\pi=0, \quad a_2{}^\pi=-a_3{}^\pi \tag{54}$$

and the linear transformation is

$$\begin{aligned} I'&=a_1{}^\pi I+b_5{}^\pi S,\\ M'&=a_2{}^\pi M,\\ C'&=-a_2{}^\pi C,\\ S'&=b_5{}^\pi I+a_4{}^\pi S. \end{aligned} \tag{55}$$

The backward scattering by an asymmetrical medium involves only *four* independent coefficients.

For a symmetrical medium b_5^{π} must be zero, so that the linear transformation is

$$\begin{aligned} I' &= a_1^{\pi} I, \\ M' &= a_2^{\pi} M, \\ C' &= -a_2^{\pi} C, \\ S' &= a_4^{\pi} S. \end{aligned} \tag{55A}$$

The backward scattering involves then *three* independent coefficients, like the forward scattering.

X. SCATTERING BY PARTICLES HAVING SPHERICAL SYMMETRY

When the incident light is completely polarized, the light scattered by one particle of any shape is completely polarized. Any depolarization of the light scattered by an isotropic distribution of similar particles is the result of the difference in polarization of light waves scattered by particles of different orientation; but if the scattering is caused by identical particles having spherical symmetry, the polarization will be the same for all the partial waves, and there will be no depolarization, if we suppose the emulsion sufficiently diluted, so that double scattering be negligible.

Thus, for such a diluted emulsion of identical spherical particles, whenever the incident light is completely polarized, the scattered light must be also completely polarized; consequently, whenever the Stokes' parameters of the incident beam verify the condition

$$I^2 - M^2 - C^2 - S^2 = 0$$

the Stokes' parameters of the scattered beam must verify the similar condition

$$I'^2 - M'^2 - C'^2 - S'^2 = 0.$$

In other words, the linear transformation (47) on the Stokes' parameters must correspond in this case to a rotation and a similarity in a Minkowsky four-dimensional space. Expressing this we obtain the relations

$$\begin{aligned} a_1 b_1 - a_2 b_1 - b_3 b_4 - b_5 b_6 &= 0, \\ -a_1 b_3 + b_1 b_4 - a_3 b_3 + b_2 b_5 &= 0, \\ a_1 b_5 - b_1 b_6 - b_2 b_3 - a_4 b_5 &= 0, \\ -b_1 b_3 + a_2 b_4 - a_3 b_4 + b_2 b_6 &= 0, \\ b_1 b_5 - a_2 b_6 - b_2 b_4 - a_4 b_6 &= 0, \\ -b_3 b_5 + b_4 b_6 - a_3 b_2 + a_4 b_2 &= 0, \end{aligned} \tag{56}$$

and

$$\begin{aligned} a_1^2 &= a_2^2 + b_3^2 + b_4^2 + b_5^2 + b_6^2, \\ a_1^2 &= a_3^2 + b_1^2 + b_2^2 + b_4^2 + b_5^2, \\ a_1^2 &= a_4^2 + b_1^2 + b_2^2 + b_3^2 + b_6^2. \end{aligned} \tag{57}$$

The three relations (57) are a consequence of the six relations (56), which are linear with respect to the four quantities a_1, a_2, a_3, a_4 and are coherent if*

$$b_2 b_3 b_4 + b_2 b_5 b_6 + b_1 b_4 b_6 - b_1 b_3 b_5 = 0. \tag{58}$$

They give then

$$\begin{aligned} 2a_1 &= \frac{b_3 b_4 + b_5 b_6}{b_1} + \frac{b_2 b_5 + b_1 b_4}{b_3} + \frac{b_2 b_3 + b_1 b_6}{b_5}, \\ 2a_2 &= -\frac{\qquad}{\qquad} + \frac{\qquad}{\qquad} + \frac{\qquad}{\qquad}, \\ 2a_3 &= -\frac{\qquad}{\qquad} + \frac{\qquad}{\qquad} - \frac{\qquad}{\qquad}, \\ 2a_4 &= \frac{\qquad}{\qquad} + \frac{\qquad}{\qquad} - \frac{\qquad}{\qquad}, \end{aligned} \tag{59}$$

from which follows a linear relation between the a's

$$a_1 - a_2 + a_3 - a_4 = 0. \tag{60}$$

There are thus only five independent parameters in the scattering by identical spherical particles without mirror symmetry.

In the case of an emulsion containing spherical particles differing in magnitude or optical properties, each scattering coefficient will be the sum of the corresponding coefficients for the various types of spherical particles in the mixture. The only relation between the scattering coefficients which will hold after this adding process, will be the linear relation (60), which is thus characteristic of scattering by any mixture of spherical particles without mirror symmetry.

For identical spherical particles with mirror symmetry we must have $b_3 = b_4 = b_5 = b_6 = 0$ and the general solution of Eqs. (56), (57) gives

$$a_1 = a_2, \quad a_3 = a_4 \tag{60A}$$

and

$$a_1^2 = a_3^2 + b_1^2 + b_2^2. \tag{61}$$

The two relations (60A) being linear will remain true for a mixture of different spherical particles

* It is also possible to solve the Eqs. (56) when the b's obey the two conditions

$$b_1 b_2 = b_3 b_6 = b_4 b_5,$$

but the singular solutions then obtained do not correspond to scattering by spherical particles.

with mirror symmetry: There are then four independent scattering coefficients.

The first of the relations (60A) is equivalent to the relation $H_v = V_h = 0$ resulting, in the case of homogeneous symmetrical spheres, from the theory of G. Mie.[7]

XI. COMPARISON WITH DIPOLAR SCATTERING

The general polarization properties of dipolar secondary light emission have been determined by P. Soleillet.[12] His theory results in the fact that the linear relations between the Stokes' parameters of the incident and scattered beams, must be in the case of dipolar scattering

$$\begin{aligned} I' &= (a - b\sin^2\phi)I - b\sin^2\phi M, \\ M' &= -b\sin^2\phi I + b(1+\cos^2\phi)M, \\ C' &= 2b\cos\phi C, \\ S' &= 2c\cos\phi S, \end{aligned} \tag{62}$$

in which a, b, c are independent of the angle of scattering ϕ. There is then no distinction between symmetrical and asymmetrical media.

For any angle of scattering, dipolar scattering is qualitatively characterized by the condition

$$b_2 = 0, \tag{63}$$

which expresses the non-existence of any ellipticity in the scattered light when there is no ellipticity in the polarization of the incident light ($S'=0$ when $S=0$).

For transverse dipolar scattering ($\phi = \pi/2$) we have, moreover,

$$a_3(\pi/2) = a_4(\pi/2) = 0 \tag{64}$$

and

$$b_1(\pi/2) + a_2(\pi/2) = 0, \tag{65}$$

which proves that whatever the polarization of the incident light, there is then neither obliquity of polarization ($C'=0$) nor ellipticity ($S'=0$), and that for an incident beam polarized in the plane of scattering ($I=M$, $C=S=0$) the scattered light is not at all polarized ($M'=C'=S'=0$).

This last test was used by R. S. Krishnan to prove the multipolar character of light scattering by some media, particularly by liquid mixtures in the neighborhood of the critical state. But this proof has been criticized,[16, 23] because convergence in the incident beam or secondary scattering is, at least qualitatively, a possible cause of the observed polarizations.

The existence of some ellipticity in the scattered light for an incident beam linearly obliquely polarized ($C \neq 0$, $S=0$) which would prove that the coefficient b_2 is not zero, would be a much more sure test of the multipolar character of the scattering, and consequently, of the non-negligible magnitude, in comparison with the wave-length, of the elements of heterogeneity of the scattering medium.

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