THE SCATTERING OF LIGHT BY PARTICLES SUSPENDED IN A MEDIUM OF HIGHER REFRACTIVE INDEX.

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1. *Introduction.*

The theory of light scattering developed by Lord Rayleigh[1] and perfectly confirmed by a series of precise measurements, relates to the case of light falling on very small particles whose diameter is many times smaller than the wave-length of light. Only in such cases the relations stated by Lord Rayleigh permit us to define the intensity of light scattered by a particle in various directions, as well as the polarisation of the scattered light.

In the case of colloidal particles observation shows that the scattering of light is quite unsymmetrical about the plane perpendicular to the incident ray. The scattering is more intense in the forward direction, that is, in the direction of the incident beam than in the opposite direction. If one studies the polarisation of scattered light, it appears that the maximum of polarisation corresponds to azimuths considerably greater than 90°.

The problem of scattering of light by particles of any dimension has been studied in general theoretically as well as experimentally by G. Mie[2] taking into account also the absorption of light by particles whose electrical conduction is not zero. Lord Rayleigh[3] has also applied Maxwell's equations to investigate the disturbance produced by the incidence of light upon a transparent sphere of dimensions comparable with the wave-length of light.

Lord Rayleigh studied experimentally the colour and polarisation of the light scattered in different directions by very fine suspensions of sulphur, and later the observations were pushed further by B. Ray[4] for the case of much larger particles. He found that for larger particles the curves representing the intensity of light scattered in different directions would

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[1] Lord Rayleigh, *Scientific Papers*, **1**, 104 ; **1**, 518 ; **4**, 397.

[2] G. Mie, *Ann. d. Physik.*, 1908, **25**, 377.

[3] Lord Rayleigh, *Scientific Papers*, **5**, 547.

[4] B. Ray, *Proc. Indian Assoc. for the Cultivation of Science*, 1922, **7**, 1.

become of an oscillatory character, the phenomena being markedly different for light having its electric vector parallel and perpendicular to the plane of scattering containing the incident and scattered rays. The reversal of polarisation observed by Lord Rayleigh with the blue light, was also observved in the case of larger particles and with red light. Theoretical formulæ were numerically computed for the case in which $\frac{2\pi\rho}{\lambda} = 5$ (ρ = radius of the particles).

The gradual changes of colour of the light transmitted through colloidal solutions with increase in size of the particles, the colours of metal-glasses and the axial colours seen through droplets of water, alcohol, etc., were theoretically considered by B. Ray.[5] A theoretical expression was also derived for the amplitude and phase of the secondary wave which was in agreement with the oscillatory character of the scattered light and the colour development observed in experiment.

A comprehensive mathematical exposition was developed using Mie's formula by W. Shoulejkin[6] for the case of spherical particles having the ideal properties of a dielectric from which he concluded that there was a smooth continuous change with increase of the size of the particles from the scattering of light to its reflection and refraction. The relative refractive index was taken to be equal to 1·32. A definite asymmetry of scattering was noticed in the case of particles having the size comparable with the wave-length of light.

Later, on the basis of Mie's exact formula, H. Blumer[7] calculated the intensity scattered in all directions by dielectric spheres of diameters from relative index of refraction $m' = 1\cdot01$ to $m' = \infty$. The intensity of light scattered in the direction of the incident ray is far greater than that in the opposite direction.

The results derived by Shoulejkin and Blumer agree with those obtained experimentally by Schaefer and Mirzkirch[8] and Schaefer and Wilmsen.[9]

Up till now, no calculations have been made of the intensity of light scattered by particles having a refractive index lower than that of the surrounding medium. In the present paper the author has tried to calculate the intensity and polarisation of light scattered by air bubbles of various

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[5] B. Ray, *Proc. Indian Assoc. for the Cultivation of Science*, 1923, **8**, 221.

[6] W. Shoulejkin, *Phil. Mag.*, 1924, **48**, 307.

[7] H. Blumer, *Zeits. f. Phys.*, 1925, **32**, 119; 1926, **38**, 304 and 920; 1926, **39**, 195.

[8] Schaefer and J. Mirzkirch, *Zeits. f. Phys.*, 1923, **13**, 166.

[9] Schaefer and Wilmsen, *Zeits. f. Phys.*, 1924, **24**, 345.

sizes suspended in liquids of different refractive indices. Experimental observations of the intensity were also made in the case of scattering of light by water particles dispersed in benzene prepared as a dilute emulsion of water in benzene.

2. *The Theory of Mie.*

A plane electromagnetic wave is incident on a sphere of radius ρ in a dielectric medium. The electromagnetic field which is characterised by material constants such as, dielectric constant, conductivity and magnetic permeability, is determined by Maxwell's equations. The integrals of the same could be represented as a sum of the products of spherical and cylindrical functions. The solution of the problem consists in the splitting of the integral into a series of partial waves which give the amplitudes of the waves radiated by the particle in all directions and superposed on the incident plane wave. By introducing the boundary conditions at the spherical surface, Mie obtained certain expressions for the components of the amplitude of the electric vectors inside and outside the sphere.

Mie derived the following intensity formula for the light scattered in a direction γ at a very great distance r from the sphere ($r \gg \rho$), when the incident beam was unpolarised the intensity being taken as unity.

$$\mathrm{I}_1 = \frac{\lambda^2}{4\pi^2 r^2} j_1 ; \quad \mathrm{I}_2 = \frac{\lambda^2}{4\pi^2 r^2} j_2 .$$

$$j_1 = \left| \sum_{\nu=1}^{\nu=\infty} \left\{ \frac{a_\nu}{\nu(\nu+1)} \Pi_\nu + \frac{p_\nu}{\nu(\nu+1)} [\Pi_\nu \cos \gamma - \Pi'_\nu \sin^2\gamma] \right\} \right|^2$$

$$j_2 = \left| \sum_{\nu=1}^{\nu=\infty} \left\{ \frac{a_\nu}{\nu(\nu+1)} [\Pi_\nu \cos \gamma - \Pi'_\nu \sin^2\gamma] + \frac{p_\nu}{\nu(\nu+1)} \Pi_\nu \right\} \right|^2 \qquad (1)$$

The sign $|\ |^2$ shows that the square of the absolute value of the complex in the parenthesis is taken.

I_1=the intensity of radiation whose electric oscillations are perpendicular to the plane of sight.

I_2=the intensity with oscillations in the plane of sight and perpendicular to the ray of light.

a_ν and p_ν are complex functions defined by boundary conditions depending upon the wave-length λ, radius of the particle ρ and the index of refraction of the material of the particle in relation to the surrounding medium m' and they signify the νth electrical and magnetic partial waves. The arguments of the cylindrical functions occurring therein are:

$$\alpha = \frac{2\pi\rho}{\lambda} \text{ and } \beta = m' \alpha$$

$m' = \frac{m_0}{m}$ where m_0 = refractive index of the particle and m=that of the surrounding medium.

IIν and II′ν are spherical functions on cos γ. The evaluated values for the first three terms are as follows :—

$II_1 = 1$; $II_2 = 3\cos\gamma$; $II_3 = 7{\cdot}5\cos^2\gamma - 1{\cdot}5$.
$II'_1 = 0$; $II'_2 = 3$ $II'_3 = 15\cos\gamma$.

$I_1 + I_2$ gives the full intensity of the rays scattered under angle γ and the difference $I_1 - I_2$ is the surplus of polarised light.

The number ν of the partial waves which should be considered as sufficient for calculation depends upon the size of the particle in relation to wave-length.

Using Mie's formula the intensity of scattered light is calculated in the present paper for the following cases :—

1. $m_0 = 1$; $m = 1{\cdot}33$; $m' = {\cdot}75$.
   (a) $\frac{2\pi\rho}{\lambda}$ is very small ; (b) $\frac{2\pi\rho}{\lambda} = 1$; (c) $\frac{2\pi\rho}{\lambda} = 3$.
2. $m_0 = 1$; $m = 1{\cdot}66$; $m' = {\cdot}6$.
   (a) $\frac{2\pi\rho}{\lambda}$ is very small ; (b) $\frac{2\pi\rho}{\lambda} = 1$; (c) $\frac{2\pi\rho}{\lambda} = 3$.
3. $m_0 = 1$; $m = 1{\cdot}5$; $m' = {\cdot}66$.
   (a) $\frac{2\pi\rho}{\lambda}$ is very small ; (b) $\frac{2\pi\rho}{\lambda} = 1$.

3. *Air Bubbles suspended in a Medium of Water.*

$m = 1{\cdot}33$; so $m' = 0{\cdot}75$.

(a) $\frac{2\pi\rho}{\lambda}$ is very small.

In this case radiation consists of the first partial wave ν=1. Besides $p_1 = 0$. Since α is small a_1 reduces to $2\alpha^3 \frac{m'^2-1}{m'^2+2}$

$$I_1 = \frac{\lambda^2}{4\pi^2 r^2}\left|\frac{a_1}{2}\right|^2 ; \quad I_2 = \frac{\lambda^2}{4\pi^2 r^2}\left|\frac{a_1}{2}\cos\gamma\right|^2$$

$$\therefore \quad I_1 + I_2 = A\,(1 + \cos^2\gamma) ; \quad I_1 - I_2 = A\,(1 - \cos^2\gamma)$$

where $A = \frac{16\pi^4\rho^6}{\lambda^4 r^2}\left[\frac{m'^2 - 1}{m'^2 + 2}\right]^2$.

The intensity distribution is symmetrical about the plane perpendicular to the incident beam. Maximum of polarisation corresponds to an angle of 90° to the incident beam. The polarisation is in the plane of sight and is complete.

(b) $\frac{2\pi\rho}{\lambda} = 1$; so $\alpha = 1$; $\beta = \frac{3}{4} = 0{\cdot}75$.

In this case only the first two partial waves are to be taken into consideration. Moreover, p_2 can be neglected. In the calculations the factor $\frac{\lambda^2}{4\pi^2 r^2}$ is dropped since the absolute intensity of light is not required. The common multiplicator $e^{\cdot i}$ is also dropped out from the coefficients a_1, p_1 and a_2.

$$a_1 = -0{\cdot}1564 + i.0{\cdot}203; \qquad p_1 = 0{\cdot}0131 - i.0{\cdot}0201.$$
$$a_2 = 0{\cdot}0158 - i.0{\cdot}0244;$$

TABLE I.

Angle γ ..	0°	20°	40°	60°	90°	120°	140°	160°	180°
j_1 ..	0·0103	0·0107	0·0116	0·0132	0·0164	0·02	0·0216	0·0229	0·0239
j_2 ..	0·0103	0·0095	0·007	0·0035	0·0001	0·0047	0·013	0·0206	0·0239
$j_1 + j_2$ ..	0·0206	0·0202	0·0186	0·0167	0·0165	0·0247	0·0346	0·0435	0·0478
$j_1 - j_2$ ..	0·0000	0·0012	0·0046	0·0097	0·0163	0·0153	0·0086	0·0023	0·0000
% Polarised light ..	0	5·9	24·7	58	99	62	24·8	5·3	0

$j_1 + j_2$ = Total Intensity. $j_1 - j_2$ = Polarised Light.

The law of radiation is graphically represented by Figure 1. The outer curve represents the full intensity of the scattered light $I_1 + I_2$ and the part

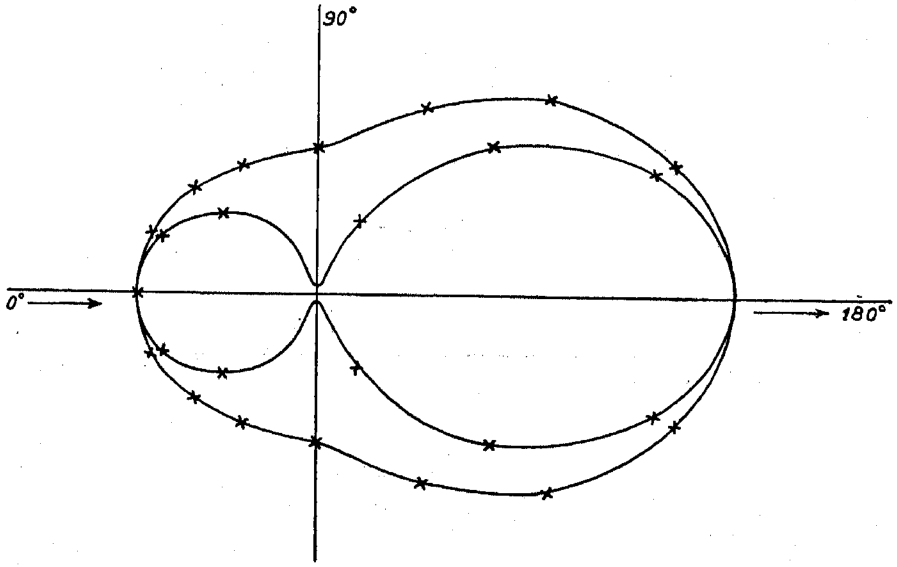

FIG. 1.

between the outer and the inner one represents the amount of polarised light $I_1 - I_2$. One notices from the figure a definite asymmetry of scattering so that the intensity of the scattered light along the forward direction is about 2·5 times greater than that in the opposite direction. The angle of maximum polarisation is no longer 90°. But it is displaced in the direction of the incident rays.

$$(c)\quad \frac{2\pi\rho}{\lambda} = 3\,;\quad \alpha = 3\,;\quad \beta = 2\cdot 25.$$

In this case one has to consider several partial waves. But as Shoulejkin has pointed out, for all practical purposes, only the first three need be taken into consideration.

$$\begin{aligned} a_1 &= 1\cdot 32 + i.1\cdot 137\,; & p_1 &= -1\cdot 138 - i.0\cdot 832. \\ a_2 &= -1\cdot 644 - i.0\cdot 992\,; & p_2 &= 0\cdot 776 + i.0\cdot 249. \\ a_3 &= 0\cdot 849 + i.0\cdot 234\,; & p_3 &= -0\cdot 215 - i.0\cdot 039. \end{aligned}$$

TABLE II.

Angle γ ..	0°	20°	40°	60°	90°	120°	140°	160°	180°
j_1 ..	0·001	0·003	0·015	0·04	0·311	2·632	5·94	10·2	12·25
j_2 ..	0·001	0·006	0·026	0·016	0·078	0·315	3·11	6·83	12·25
$j_1 + j_2$ ..	0·002	0·009	0·041	0·056	0·389	2·947	9·05	17·03	24·5
$j_1 - j_2$ ..	0·000	-0·003	-0·011	0·024	0·233	2·317	2·83	3·37	0·000
% Polarised light ..	0	− 33	− 26·8	42·7	60	78	31	19	0

The negative sign shows that the plane of polarisation is turned through 90° and that the electrical vibrations are in the plane of sight.

The total intensity of scattered light is plotted against the angle, see Figure 2.

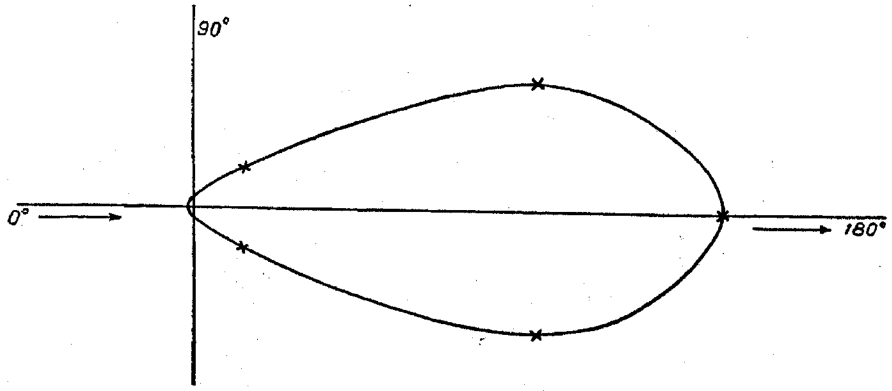

FIG. 2.

Here also the asymmetry is very striking. The forward scattering is by far more intense than the backward scattering.

4. *Air Bubbles suspended in a Medium of Refractive Index* 1·66.

$$m = 1{\cdot}66, \; m' = \frac{m_0}{m} = {\cdot}6$$

(*a*) For particles of diameter very small compared with λ just as in the previous case, a symmetrical distribution of intensity is got with maximum polarisation at 90°.

$$(b) \quad \frac{2\pi\rho}{\lambda} = 1\,; \quad \alpha = 1\,; \quad \beta = 0{\cdot}6.$$

The values of intensity for various angles are tabulated as shown below :—

$a_1 = -0{\cdot}25 + i.0{\cdot}2961\,;$ $\qquad p_1 = 0{\cdot}0189 - i.0{\cdot}0287.$
$a_2 = 0{\cdot}0246 - i.0{\cdot}0385\,;$

TABLE III.

Angle γ ..	0°	20°	40°	60°	90°	120°	140°	160°	180°
j_1 ..	0.0238	0·0247	0·0267	0·0302	0·0375	0·0457	0·0504	0·0534	0·0547
j_2 ..	0·0238	0·0210	0·0155	0·0079	0·0002	0·0101	0·0217	0·0450	0·0547
$j_1 + j_2$ ..	0.0476	0·0457	0·0422	0·0381	0·0377	0·0558	0·0721	0·0984	0·1094
$j_1 - j_2$ ..	0·000	0·0037	0·0112	0·0323	0·0373	0·0356	0·0287	0·0084	0·000
% Polarised light ..	0	8·1	26·5	84	99	63	39	8·5	0

As in the corresponding previous case asymmetry present in the distribution of intensity is quite clear here. The above generalisations also hold good in this case.

$$(c) \quad \frac{2\pi\rho}{\lambda} = 3\,; \quad \alpha = 3\,; \quad \beta = 1{\cdot}8.$$

$a_1 = 0{\cdot}855 + i.\,2{\cdot}515\,;$ $\qquad p_1 = -\,0{\cdot}837 - i.\,1{\cdot}307.$
$a_2 = -\,2{\cdot}071 - i.\,1{\cdot}701\,;$ $\qquad p_2 = 1{\cdot}031 + i.\,0{\cdot}365.$
$a_3 = 1{\cdot}215 + i.\,0{\cdot}416\,;$ $\qquad p_3 = -\,0{\cdot}298 - i.\,0{\cdot}061.$

TABLE IV.

Angle γ ..	0°	20°	40°	60°	90°	120°	140°	160°	180°
j_1 ..	0·003	0·04	0·118	0·35	1·104	4·28	9·88	16·8	20·1
j_2 ..	0·003	0·036	0·083	0·09	0·471	0·584	4·63	14·3	20·1
$j_1 + j_2$ ..	0·006	0·076	0·201	0·44	1·575	4·864	14·51	31·1	40·2
$j_1 - j_2$ ..	0·000	0·004	0·025	0·26	0·633	3·696	5·25	2·5	0·000
% Polarised light ..	0	5·3	12·4	56·5	41·9	76	36·1	8	0

5. *Air Bubbles in a Medium of Refractive Index 1·5.*

$$m = 1 \cdot 5,\ m' = \frac{m_o}{m} = \cdot 66.$$

(*a*) $\frac{2\pi\rho}{\lambda}$ is very small. Results are similar to that got in the previous cases 3*a* and 4*a*.

$$(b) \quad \frac{2\pi\rho}{\lambda} = 1\ ; \quad \alpha = 1\ ; \quad \beta = 0 \cdot 66.$$

$$a_1 = -\,0 \cdot 084 \;+ i.\,0 \cdot 225\ ; \qquad p_1 = 0 \cdot 013 - i.\,0 \cdot 039.$$
$$a_2 = 0 \cdot 0116 - i.\,0 \cdot 018\,;$$

TABLE V.

Angle γ ..	0°	20°	40°	60°	90°	120°	140°	160°	180°
j_1 ..	0·0079	0·0082	0·0093	0·0109	0·0145	0·0184	0·0207	0·0222	0·0228
j_2 ..	0·0079	0·0072	0·0048	0·0006	0·0001	0·0057	0·0130	0·0191	0·0228
$j_1 + j_2$ ..	0·0158	0·0154	0·0141	0·0115	0·0146	0·0241	0·0337	0·0413	0·0456
$j_1 - j_2$ ..	0·0000	0·001	0·0045	0·0103	0·0144	0·0127	0·0077	0·0031	0·0000
% Polarised light ..	0	6·5	31·9	89·5	98·6	52·6	22·8	7·5	0

From the above calculations it is quite clear that in the case of particles (dielectric in an ideal state) whose refractive index is lower than that of the surrounding medium a distribution of intensity of scattered light is given according to Rayleigh's formula, provided the diameters of the particles are small compared with the wave-length of light used. When the size of the particle increases, the symmetric nature of the scattering vanishes and the intensity of light scattered in the forward direction preponderates over that in the backward direction. The results obtained are in quite agreement with those of Shoulejkin and others for the case of colloidal particles or clouds having a refractive index higher than that of the surrounding medium.

6. *Experimental Observations.*

A clean dry bottle was first washed with a small quantity of benzene. About 200 c.c. of pure benzene was taken in it, and about 20 drops of water were added and the mixture was shaken well for about 20 minutes so that the water got well-dispersed in the medium of benzene. This emulsion of water in benzene was poured into a rectangular cell which had been previously cleaned and washed with benzene. This was placed in the path of a narrow parallel beam of light from a carbon arc. With the naked eye the distribution of intensity of the light scattered in various directions was noted. It was observed that the intensity in the forward direction was considerably greater than that in the backward direction.

Further experimental work on the same lines is in progress.

In conclusion, the author wishes to express his grateful thanks to Sir C. V. Raman, KT., F.R.S., N.L., for his keen interest and guidance during the progress of the work.

7. *Summary.*

A short review of the theoretical as well as experimental work done on the scattering of light by particles suspended in a medium of lower refractive index is given. Using the formula derived by Mie, calculations are extended to the case of particles such as air bubbles suspended in a medium of higher refractive index. Results are obtained for three different values of refractive indices and for three different sizes of the particles. When the particles are small in size, the distribution of intensity is symmetrical about the plane perpendicular to the incident beam. As the size of the particle increases, the intensity in the forward direction becomes greater than that in the backward direction. The angle of maximum polarisation is displaced from 90° in the direction of the incident rays and moreover, it is everywhere incomplete. Experimental observations are also made in the case of water particles dispersed in benzene.