SCATTERING OF LIGHT BY LARGE WATER DROPS—PART II

BY G. R. PARANJPE, Y. G. NAIK AND P. B. VAIDYA
(*From the Department of Physics, Royal Institute of Science, Bombay*)

Received October 19, 1938

IN Part I of this paper we gave detailed calculations of the intensity and polarisation of light-scattering by large water drops particularly with a view to study the angular distribution of intensity and its dependence on the particle sizes. The present part concerns itself with an experimental study of the subject.

As has been pointed out earlier that the available experimental data on the scattering of light are very meagre, the problem of scattering of light by dielectric spheres of sizes comparable to, or greater than, the wave-length of light has not been worked out before in detail.

Previous Work

The experimental works of Keen and Porter[16], Ray[9] and others are restricted mainly to small colloidal particles, and the scattering is measured either along the direction of light or in a transverse direction. Benedict and Senftleben[17] were the first to study systematically the scattering of light by small particles in different directions.

Pokrowsky[13] worked on water drops formed by sudden condensation of water vapour. The average size of drops in his experiments was considerably smaller than the wave-length of light, the greatest value of $\alpha = \frac{2\pi\rho}{\lambda}$ being $0 \cdot 67$. Konig-Marten's sector-photometer was employed to determine the intensity of the polarisation of the scattered light and the intensity of one of the components. The results of his observations for intensity and polarisation agree well with those calculated according to the theory. He also applied an approximation of Rayleigh's theory in the explanation of his experimental work on the scattering of light by water drops in different directions within 5 degrees from the forward direction.

Webb[18] has investigated recently the scattering of light by large drops of water and alcohol-water mixture. The drops were illuminated by a parallel beam of light from a carbon arc and the intensity of the scattered light was recorded on a photographic plate. His observations, however, are likely to have been affected by the inconstancy of the arc, the growth of the drops in the process of condensation, and uncertainty in the number of scattering drops.

Borne[19] used copper oxide photo-cells, one to measure the intensity of the scattered light by cigarette and ammonium chloride smokes illuminated by a 5000 watt lamp, and another to measure the intensity of the source. The particle size was, however, considerably smaller than the wave-length of light.

Work of this type has usually to contend with two difficulties. One of them refers to the production of fairly large, stationary and uniform dielectric spheres. The other is the estimation of their number and dimensions. The theoretical working out of the problem as mentioned in the first part is also very laborious and becomes unwieldy for large spheres. It is on account of these difficulties perhaps that this problem has not been pursued in much detail.

Production of steady clouds, the determination of the size of the drops and the transmission of light have been subjects of study for a long time by means of the apparatus used in the present work. It has been possible to overcome the difficulties mentioned before, and a fairly stable cloud of uniform density and of any desired particle size of radius, between 1 to 12 μ can be now produced. The instantaneous determination of the size of the drops by the corona ring method and the stability of cloud have been of special advantage in the present work. It must be emphasised that the uniformity of the size of water-drops in the cloud is of very great and fundamental importance in the work on scattering. A slight change in the size of the drops changes the general character of the scattering very much. The details of these experiments have been given in a number of papers[20].

Production of a Cloud of Water Drops

The clouds on which the present investigations were carried out were formed inside a flask of about 12 litres capacity by adiabatic expansion. This expansion was effected by connecting this flask suddenly to a large tank in which a suitable low pressure was created by means of a vaccum pump. This tank was connected with the flask through a tube of wide bore, whose cross-section was about 1 sq. inch. The instantaneous connection between the flask and the tank was made by means of a cock of a larger cross-section. A small quantity of water or liquid to be experimented upon was kept inside the flask.

When a sudden expansion of any desired magnitude is made, a fixed quantity of liquid is available for condensation and this liquid distributes itself on the number of nuclei available in the flask. Thus, by controlling the entry of dust nuclei into the flask and the expansion ratio, a cloud of any desired size of drops can be produced. In order that the vapour may not condense on the sides of the flask and that the air inside the flask may remain saturated, it was vigorously shaken every time so as to wet all the inside surface.

Care was taken during the experiments to see that the cloud produced was of uniform density and that the size of the drops for any particular set of measurements was kept the same. This was achieved by producing a large number of smoke particles (size less than about 50 $\mu\mu$) from a particular brand of incense stick in a large cupboard of about 4000 litres capacity. The smoke was allowed to be diffused and co-agulated in the initial stages, after being thoroughly mixed by means of a fan, for a period of at least an hour or more before the actual measurements were made. This ensured the density of smoke particles in the cupboard to be the same. It was found that the decrease in the number of smoke particles in the beginning is very rapid and that it becomes extremely slow after about an hour. As any set of measurements takes not more than an hour, one can neglect any small variation in the number of smoke particles. A measured quantity of this smoke-bearing air from the cupboard was allowed to enter the expansion flask every time.

With a little experience it was possible to control the entry of smoke-bearing air from the cupboard into the flask as also the expansion ratio, so that a cloud of uniform density and of desired size of drops could be produced.

The size of the drops in the cloud was determined or checked by the corona rings seen by looking at a distant source of light through it. The angular aperture of the corona ring in monochromatic light serves as a method of determining instantaneously the size of the drops. The wave-length of light, the size of the drops and the angular aperture of the corona are related with each other as follows :—

$$\text{Sin } \theta = (n + 0 \cdot 22) \frac{\lambda}{2\rho},$$

where 2θ = angle subtended by the diameter of a corona ring minimum at the eye of the observer.

n = the order of the ring.

λ = mean wave-length of the light forming the corona.

ρ = average radius of the drops.

A well-defined circular corona ring attains its final size in less than a second and remains stationary till the cloud settles down. The well-defined circular shape of the corona suggests that the drops are nearly of the same size. The final steady size of the corona indicates that the drops do not evaporate appreciably during the course of the experiment.

In some experiments, when the drops were smaller than 2 μ in radius, the corona method was used only as a check for the size of the drops, and not directly to determine it. It is found in actual practice that the size of the corona is well-defined for all drops of radii larger than 2 μ. Wilson[21] and Mecke[7] have also shown that the corona theory is applicable only to drops whose radii are greater than 2 μ. Even for drops of 2 μ radii, the corona is very large and diffuse and hence the size was checked by the Stokes' law as corrected by Bromwich[22] and Cunningham[23]. A check was also obtained by a subsequent experiment in which only a known fraction of the air from the cupboard was introduced and a well-defined corona was observed in the cloud. This experiment indicated how many nuclei were there in the large cupboard. Knowing this and the total volume of nuclei-bearing air that was introduced into the flask for actual experiment, the value of the size of the drops, when less than 2 μ in radii, was determined. It was found that the two values agreed well within an error of about 5%. The contribution by the nuclei within the drops to the scattered light is quite negligible. It was therefore, not necessary to make any correction in the intensity measurements on this account.

Intensity Measurement

In order to measure the intensity of light scattered by a beam of parallel rays in any given direction a source of light S_2 was taken. S_1 is another lamp, kept at a great distance and is used to obtain the corona rings in the cloud. A monochromator M is used to facilitate the measuring of the intensity of scattered light of a particular wave-length (Fig. 1).

In order that the intensity of the scattered light may be as large as possible, the source of light S_2 was kept within the wooden chamber in which the experimental flask was kept and the monochromator was also pushed as near to the flask as possible. In order to change and measure the direction of scattered light entering the monochromator a spectrometor table was used. The flask F in which the cloud was produced was placed at the centre of this table and along the axis of the collimator of the monochromator. The source of light S_2 along with the lens L_2 and diaphram DD′ was mounted on an arm of the spectrometer table. A heavy weight is attached to the other side of the arm as a counterpoise. The arm, being attached to a vernier scale moving on the main circular scale of the spectrometer table, can be rotated and fixed to any desired angle.

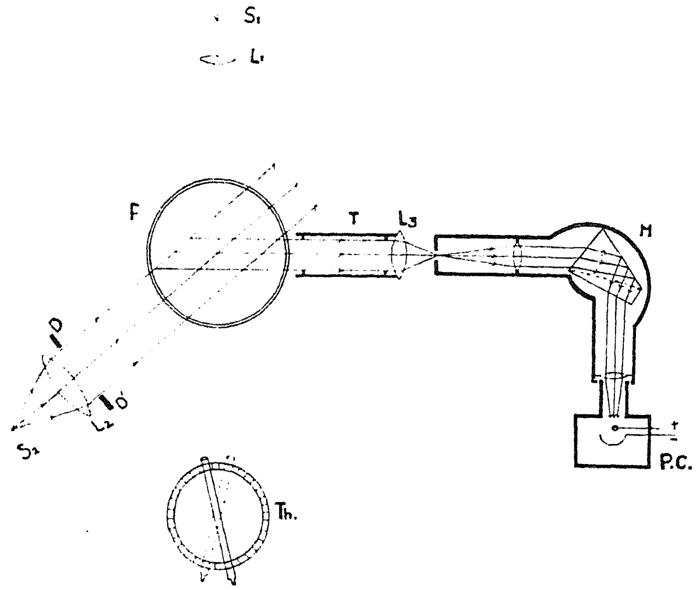


FIG. 1

S_1 & S_2—Sources of light; L_1 & L_2—Collimating lenses; DD'—Diaphram; F—Condensation flask; T—Collimating tube; L_3—Lens; M—Monochromator; $P.C.$—Photo-cell; Th—Theodolite.

The actual intensity of light is measured by means of a photo-cell P.C. attached to the monochromator. The diagrammatic sketch of the connections of the photo-cell is shown in Fig. 2.

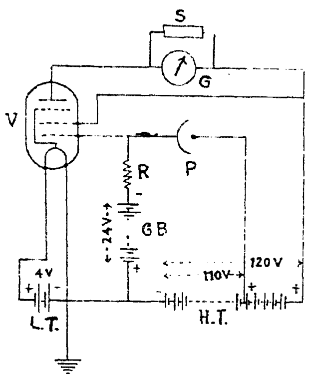


FIG. 2

The temperature of the air surrounding the cloud chamber was observed every now and then. As the cloud chamber is placed in a large enclosure there was very small variation of temperature in its neighbourhood. It was observed that if the lamp S_2 was kept on for a long time there was a small rise in the temperature of the enclosure and hence care was taken to switch on this lamp only for a short time for the measurement of the scattered intensity.

The coiled filaments of lamp S_2 were in one plane in the form of a square and hence gave a beam of uniform intensity. This light was made parallel by a lens mounted on the same arm of the spectrometer table as the source. The lens had a large aperture so that a rectangular beam of light was passing through the cloud. One more diaphram was placed between the lens and the cloud chamber. The lamp was so placed that the boundaries of this beam were in the horizontal and vertical planes. In order that the intensity of light of this lamp may remain constant, it was connected to a separate storage battery.

The scattered beam was allowed to pass through a tube with a slightly smaller cross-section and was then focussed by means of a lens on the slit of the monochromator. It was ascertained that no extra light fell on the slit. The aperture of the tube receiving the scattered light was kept small to facilitate the estimation of the correction for the volume and number of actual scattering particles, when the incident beam is rotated through different angles.

The incident direction of light was every time ascertained before beginning any set of observations by first adjusting the position of the lamp in such a way that the photo-cell gave a maximum deflection for direct light incident on the slit of the monochromator. As a further check on the measurements it was ascertained that with a given cloud, equal rotations of the lamp S_2 on either side of this position, gave the same current through the photo-cell. This adjustment ensured the correctness of the observations for different angles of scattering.

The photo-cell was found to work very satisfactorily. Initially there used to be a very large fluctuation in the dark current, but later on by earthing the box containing the cell as also the one containing the amplifying circuit, the fluctuation disappeared. The readings obtained after this improvement were consistent and could be reproduced. The photo-cell was most sensitive in the red region and hence the monochromator was adjusted to select a wave-length in this region.

Volume Correction

As the incident beam of light illuminates only a small portion of the cloud in the flask, the effective volume of cloud which scatters light varies with the angle of scattering. From Fig. 3 (*a*, *b*) it is clear that the volume of cloud responsible for the scattering of light in a given direction depends on the angle at which the scattered light is received in the monochromator. Evidently the volume responsible for scattering is more for angles nearer to 0° or 180°. For angles near about 90°, the volume of cloud scattering the light is a minimum. The volume of intersection of the incident beam and the scattered beam will be a function of the angle γ and it can be expressed by $\frac{V_0}{\sin \gamma}$, where V_0 is the volume of intersection at 90°. (This will hold good only for $\gamma > \frac{2D - b}{a}$, where $2D$ = breadth of the incident beam ; $2b$ = diameter of the scattered beam ; a = radius of the flask.)

This determination is necessary because in the present case the scattering of light is not due to an isolated sphere, but due to a large number of particles distributed uniformly in the cloud.

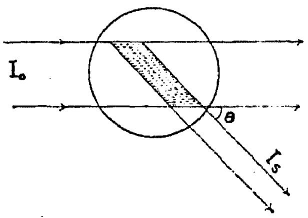


FIG. 3, *a*

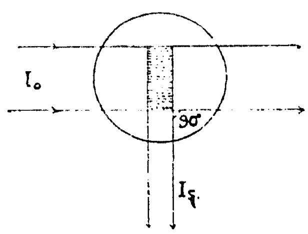


FIG. 3, *b*

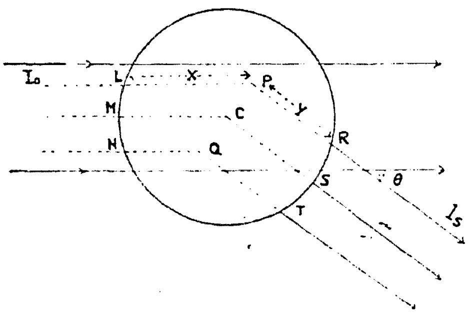


FIG. 4

If we assume the approximate formula of light-absorption in a cloud of drops as given by Jöbst[24], Debye[5], Anderson[25], and modified by Houghton[26] we find that the intensity of light I′ reaching a sphere P well within the flask (Fig. 4) will be given by

$$I' = I_0 \times e^{-kx}$$

where I_0 = intensity of incident light on P ;

k = constant ;

and X = distance of the sphere P from the boundary of the flask where the light enters the cloud.

This light will be scattered by that drop in all directions and the intensity of light coming out in any given direction γ will be further absorbed for a distance Y within the flask. The intensity of scattered light I″ by P in a direction γ will therefore be given by

$$I'' = I_0 \times e^{-kx} \times F(\rho, \lambda, m', \gamma),$$

where $F(\rho, \lambda, m', \gamma)$ is a scattering function depending on the radius of the drop, the wave-length, the refractive index and the angle of scattering, γ.

This scattered light will suffer further absorption due to a path Y within the flask and hence I_s the actual intensity of scattered light coming out along γ will be given by

$$\begin{aligned} I_s &= I'' \times e^{-kY} \\ &= I_0 \times e^{-kX} \times e^{-kY} \times F(\rho, \lambda, m', \gamma) \\ &= I_0 \times e^{-k(X+Y)} \times F(\rho, \lambda, m', \gamma). \end{aligned}$$

If there are N particles scattering the light, the total intensity coming out along a direction γ will be

$$I_s = I_0 \times N \times e^{-k(X+Y)} \times F(\rho, \lambda, m', \gamma).$$

If we try to find out the average length of the path $(X+Y)$ within the flask of the incident and the scattered light by various particles, we shall obtain $(X+Y) = 2a$ where a is the radius of the flask.

This becomes clear from Fig. 4 in which we find that for a particle P the length of path of light is (LP + PR) for one at C the path is (MC + CS) $= 2a$ and for Q it is (NQ + QT). Again $(LP + PR) > 2a > (NQ + QT)$, and hence the average value of $(X+Y)$ can be approximately taken equal to $2a$.

The number of scattering particles N will be given by $N = nV$,

where n = number of drops per c.c.

and V = volume of the cloud scattering the light.

If V_0 = volume of intersection of the incident and the scattered beams when they are at right angles to each other (*i.e.*, when $\gamma = 90°$),

$$V = \frac{V_0}{\sin\gamma}$$

$$\therefore N = \frac{n V_0}{\sin\lambda}$$

Thus the final formula giving the intensity of scattered light in the present experimental arrangement is

$$I_s = I_0 \times \frac{n V_0}{\sin\gamma} \times e^{-2ak} \times F(\rho, \lambda, m', \gamma)$$

by taking $nV_0 e^{-2ak} = C$ = a constant we get,

$$I_s = I_0 \times C \times F(\rho, \lambda, m', \gamma) \times \frac{1}{\sin\gamma}$$

$$\therefore \quad I_s \times \sin\gamma = I_0 \times C \times F(\rho, \lambda, m', \gamma).$$

Thus all observed values of I_s will have to be multiplied by sin γ in order to correlate them with the actual values of scattering I obtained from the scattering function F $(\rho, \lambda, m', \gamma)$.

Here it must be noted that the number of drops per c.c. is of the order of 2000 to 6000, and they little influence the final result on account of multiple scattering. Trincks[27] has pointed out in his recent paper that serious changes are required in Mie's result when two particles are very near. These changes become negligible when the separation of drops is greater than two or three particle diameter. The maximum average distance between two drops in the present investigations taking n to be equal to 8000, comes out to be 0·05 cm. The maximum diameter of the drops is 8 μ (·0008 cm.). Thus the ratio of the average distance to the maximum diameter turns out to be 62·5°. Thus it is clear that multiple-scattering does not affect the final result. As a further check, experiments were performed by collimating the incident light with apertures of different sizes in any fixed direction. Tests carried out at three different angles indicated that keeping all the factors constant and varying only the number of scattering particles by changing the aperture of the incident beam, the intensity of the scattered light was proportional to the number of particles.

Experimental Results

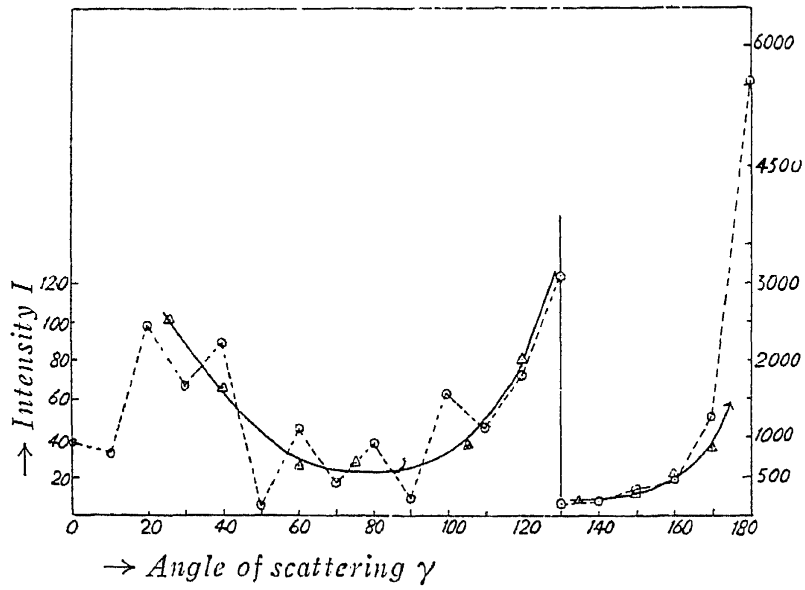
To verify the theoretical results, experiments were performed for three particle sizes, *viz.*, $a = 10$, 20, and 30. The results are expressed as factors proportional to the ratio of the scattered light to the incident light, and are given below after applying the necessary volume-corrections for different angles. The wave-length of light was taken to be 6800 A.U. for all the measurements. γ gives the angle of scattering and I is the final value of the scattered light corected for volume.

Results

TABLE I			TABLE II			TABLE III		
γ in degrees	I Theoretical $\alpha=10$	I Experimental $\alpha=10\cdot12$ $\rho=1\cdot096\mu$	γ in degrees	I Theoretical $\alpha=20$	I Experimental $\alpha=20\cdot22$ $\rho=2\cdot19\mu$	γ in degrees	I Theoretical $\alpha=30$	I Experimental $\alpha=30\cdot30$ $\rho=3\cdot28\mu$
25	80·0	103	40	97·0	92	40	130·0	125
40	89·9	67	50	116·3	75	50	78·5	95
60	45·1	30	60	36·1	45	60	47·5	60
75	25·0	27	70	8·1	36	70	40·0	52
90	9·7	28	80	47·8	30	80	14·5	35
105	55·0	36	90	59·9	45	90	48·4	38
120	72·2	80	100	32·9	50	110	130·0	125
135	140·0	125	110	45·5	76	120	195·8	212
150	328·5	286	120	134·0	130	130	666·2	690
160	475·0	492	130	257·0	216	140	1367·0	1275
170	1274·0	829	140	315·4	312	150	2068·0	2300
			150	453·1	500	160	10540·0	4810*
			160	1370·1	1140			

Discussion of Results

The experimental results for $\alpha = 10\cdot12$ are shown by a graph in Fig. 5 along with the theoretical curve for $\alpha = 10$. It is seen from the nature of the curve that the maxima and minima are not observed at all in the experimental curve. The average lay-out of the curve in the backward direction is however along a mean path of the theoretical curve. The maxima and minima cannot be expected to be observed experimentally for reasons described later. In the forward direction the curve is in very good agreement with the theoretical curve, except for angle 170°. This angle lies within the limit $\gamma = \dfrac{2\,D - b}{a}$ and hence the correction $\dfrac{V_0}{\sin\gamma}$ does not hold good for this value. For all small angles smaller than 12·5°, the same correction $\dfrac{V_0}{\sin 12\cdot5^\circ}$ holds good, in the present case. The discrepancy in the result therefore disappears as the value of the intensity as corrected above becomes 1033 instead of 829.

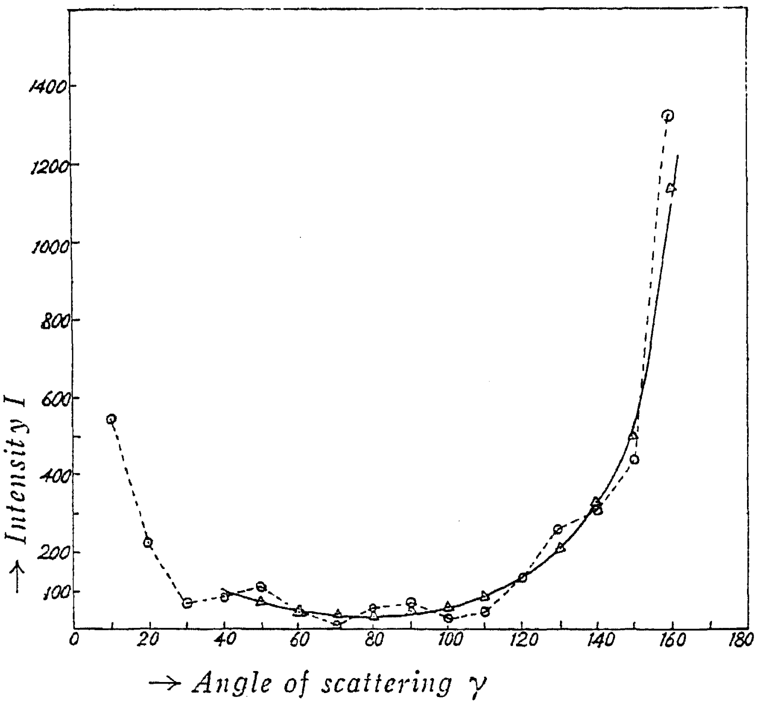


⊙ - - - - - - - - *Theoretical*; $\alpha = 10$

△ ———————— *Experimental*; $\alpha = 10 \cdot 12$

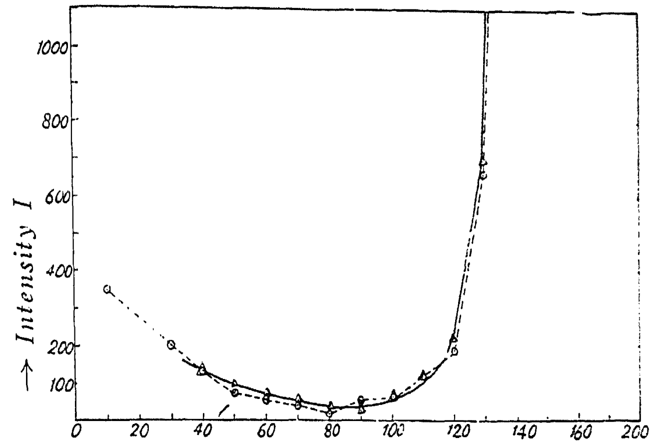
FIG. 5

The experimental curves for $\alpha = 20$ and 30 plotted in Figs. 6 and 7 respectively, lend further proof to the theory of Mie for larger drops. In the backward direction the intensity from 40° to 70° decreases rapidly, from 70° to 110° the decrease is small and from 120° onwards the increase is very rapid. The experimental curves appear to agree very well with the theoretical ones, in both the forward and the backward directions. It is found



⊙ - - - - - - - - *Theoretical* ; $\alpha = 20$

△ ———————— *Experimental* ; $\alpha = 20 \cdot 22$



→ *Angle of scattering* γ

⊙ - - - - - - - - *Theoretical*; $a = 30$

△ ———— *Experimental*; $a = 30 \cdot 30$

FIG. 7

that the general lay-out of the curves coincides with a mean hypothetical curve running through the theoretical one. The individual readings of the intensities differ from the theoretical ones to an extent of ± 17 per cent. These discrepancies can be attributed to various factors mentioned below. (The result marked with an asterisk in Table III deviates considerably from the theoretical value.)

The readings of intensities at least for three different angles were taken consecutively in one cloud formation. The adjustment of the angles had therefore to be done within a short time and hence could be adjusted correct to about 20 minutes. This gave rise to a maximum error of about 7 per cent. The collimation of light cannot be said to be perfect and hence some error due to the light being slightly diverging or converging is always likely to occur. Again the size of the drops may not be absolutely identical for all individual drops. The actual values of a in the experiments do not come out to be exactly in round numbers 10, 20 and 30 for which the theoretical curves have been obtained.

The above factors are also responsible for the fact that there were no maxima and minima observed in the experimental results. The general lay-out of all the curves however appear to verify Mie's theory quite well.

It can be observed from the curves that the quantitative agreement between the theoretical and experimental results is much closer in the forward direction than in the backward direction. As the intensity falls down considerably in the backward direction the errors in the intensity measurements are likely to be more than in the forward direction.

The experiments bring out clearly that for the study of scattering, or transmisson, of light it is essential that the size of the spheres in any scattering medium must all be the same, and that the particles should not be too dense to give rise to multiple scattering. The theory of Mie is thus applicable not only to small particles but also to particles of any size as postulated by him.

REFERENCES

1. Lord Rayleigh .. *Collected Papers*, pp. 104, 518 and 547 ; *Proc. Roy. Soc.*, 1911, **84,** 25 ; 1914, **90,** 219 ; 1917, **94,** 297.
2. Love .. *Proc. Lond. Math. Soc.*, 1899, **30,** 308.
3. Mie .. *Ann. d. Phys.*, 1908, **25,** 428.
4. Macdonald .. *Phil. Trans. Roy. Soc.*, 1910, **210,** 113.
   Nicholson .. *Proc. Lond. Math. Soc.*, 1910, **9,** 67 ; 1912, **11,** 277.
   Bromwich .. *Phil. Trans. Roy. Soc.*, 1920, **220,** 175.
   Ryde .. *Proc. Roy. Soc.*, 1931, **131,** 451.
   Kunz .. *Phil. Mag.*, 1930, **39,** 416.
   Rosenberg .. *Ann. d. Phys.*, 1922, **68,** 414.
5. Debye .. *Ibid.*, 1909, **30,** 57.
6. Ganz .. *Ibid.*, 1925, **76,** 29.
7. Mecke .. *Ibid.*, 1920, **62,** 623 ; 1921, **65,** 257.
8. Blumer .. *Zeit. f. Phys.*, 1925, **32,** 119 ; 1926, **38,** 308 and 942 ; 1926, **39,** 195.
9. Ray .. *Proc. Ind. Assn. Cult. Sci.*, 1921, **7,** 1 ; 1923, **8,** 23.
10. Wiener .. *Abh. d. Kaiser-Leopold-Crol Deutch Acad. d. Natur.*, 1907, **73,** Pt. I and 1909, **91,** Pt. II.
11. Lambert .. *Photometria*, Ausberg, 1860.
12. Lommel and Seeliger .. *Ann. d. Phys.*, 1889, **36,** 473.
13. Pokrowsky .. *Zeit. f. Phys.*, 1925, **31,** 14 and 514 ; 1927, **41,** 688 ; 1927, **43,** 395 ; 1929, **53,** 67 ; 1930, **60,** 850 ; 1930 ; **65,** 130.
14. Dorno .. *Phys. d. Sonnen-und Himmel Strahlung Braunchweig* 1919.
15. Shoulejkin .. *Phil. Mag.*, 1924, **48,** 307.
16. Keen and Porter .. *Proc. Roy. Soc.*, 1913, **89,** 370.
17. Benedict and Senftleben *Ann. d. Phys.*, 1919, **60,** 297.
18. Webb *Phil. Mag.*, 1935, **19,** 927.
19. Borne .. *Zeit. f. Phys.*, 1936, **99,** 73.
20. Paranjpe, Mirchandani and Naik *Bomb. Univ. Jour.*, 1932, **1,** Pt. II, 1.
    ——— and Naik .. *Ibid.*, 1934, **3,** Pt. II, 8.
    ——— and Bhagwat .. *Ibid.*, 1935, **4,** Pt. II, 28.
21. Wilson .. *Camb. Phil. Soc. Proc.*, 1929, **25,** 369.
22. Bromwich .. *Ibid.*, 1929, **25,** 369.
23. Cunningham .. *Proc. Roy. Soc.*, 1910, **83,** 367.
24. Jöbst .. *Ann. d. Phys.*, 1925, **76,** 165 and 863.
25. Anderson .. *The Aviation*, 1930, **38,** 930.
26. Houghton .. *Phys. Rev.*, 1931, **38,** 152.
27. Trincks .. *Ann. d. Phys.*, 1935, **22,** 561.