INTERFEROMETRIC STUDIES OF LIGHT SCATTERING IN MOBILE LIQUIDS

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1. *Introduction*

A DESCRIPTION of the liquid state requires a knowledge of the distribution of molecules within the fluid, as also of their movements and their mutual interactions. Spectroscopic studies on the scattering of light afford probably the most direct method of obtaining information regarding these matters and are thus of great importance in enabling us to reach an understanding of the liquid state.

The molecular movements in liquids are of three different kinds in diminishing order of frequency, namely, the molecular vibrations, the molecular rotations which in the vast majority of liquids are hindered by collisions, and the translatory movements which are also restricted in the same way. It is with the two latter types of motion alone and especially with the last that we are here concerned. The present paper deals, in fact, with the scattering of light in dust-free liquids, occurring either without change of frequency or with frequency changes so small that they require interferometric aid for detection.

The translatory motions of the molecules in liquids would obviously give rise to Doppler shifts of frequency in the scattered light of the same nature as those observable in gases and dealt with in an earlier paper in these *Proceedings*. In the vast majority of liquids, however, such changes of frequency would, in view of the large molecular masses involved, be too small to be detectable. Considerably larger shifts falling within the range of interferometric study may, however, arise in a manner first indicated by Brillouin (1922). In the theory of the latter, the discrete molecular structure of the fluid is ignored and the scattering is regarded as a monochromatic reflection of the incident light waves by trains of sound waves of appropriate wave-length and orientation forming regular stratifications in the medium. Such reflections would be accompanied by Doppler shifts of frequency in one direction or the other and of a magnitude determined by the velocity

322

of the sound waves in the fluid. That the effect contemplated by Brillouin does arise in liquids is now a well-established experimental result. It is evident, however, that Brillouin's theory cannot completely describe the effects observable with liquids. The discrete molecular structure and especially the random molecular orientations may be expected to give rise to phenomena of a different kind. The assumption that the thermal agitation takes the form of extended wave-trains propagated in a regular way may also depart from the truth in certain circumstances, e.g., in highly viscous liquids and even in mobile liquids at high temperatures. A point of view which embraces Brillouin's theory but which is sufficiently general to include all such possibilities is to regard the changes of frequency in every case as a Raman effect. The translatory movements of the molecules would evidently produce local fluctuations of optical density in the medium. These would, in general, vary with time and the resultant scattering should accordingly exhibit frequency changes which are both positive and negative. Variations of optical density which are of a quasi-static or stationary character, if present, would give a scattering of unaltered frequency. On the other hand, pulsations of optical density taking place over an extended domain in the fluid and travelling through the medium in the manner of sound waves would give a coherent modified scattering, the resultant effect being spectral shifts varying with the direction of observation in the manner indicated by the Brillouin theory. The interferometric study of light scattering thus, in effect, enables us to make a frequency analysis of the fluctuations of optical density and to find whether they occur with or without correlation of phase at different points within the fluid (Raman and Venkateswaran, 1938).

2. *Experimental Arrangements*

A : Choice of Interferometer.—Among the previous investigators who have worked on the present subject, Gross (1930), Meyer and Ramm (1932), Ramm (1934) and Birus (1937) have used an echelon grating, while Rafalowski (1931) has used the Lummer-Gehrcke plate; but the most satisfactory patterns have been obtained with a Fabry-Perot etalon by Rao (1934). The author has examined the efficiency of various interferometers in order to choose the instrument best suited to the problem. For this purpose the 4047 A.U. radiation of a mercury arc scattered by ethyl alcohol was examined by (1) a 36-step reflection echelon having pressure and temperature controls supplied by Adam Hilger and Co., (2) a Lummer plate of glass of length 130 mm. and thickness 4·48 mm. and (3) a Fabry-Perot etalon of quartz with invar separation specially made by Heinrich Lesche of Potsdam. It was observed that the etalon gives the maximum separation and definition

of the fine structure components though with a low intensity, while the Lummer plate gives the maximum intensity but with the disadvantage that the fringes are somewhat broad. The echelon is unsuited for these investigations in spite of its high resolving power, on account of its low luminosity and dispersion and the complicated nature of the interference pattern. In the following investigations, the etalon has been used for recording the fine structure components and the Lummer plate for polarisation studies in which the intensity of scattered light is necessarily low.

B : The Source.—In most of the previous investigations, the 4046 and 4358 A.U. radiations of a high vacuum mercury arc were used for the incident light. These lines suffer from the disadvantage that they are accompanied by intense hyperfine structure satellites which make their appearance in the scattered light. 4358 A.U. is particularly unsuited as the principal maxima themselves are excessively broad. 4078 A. U. is comparatively more satisfactory, as the hyperfine structure components are of relatively low intensity. These features of the mercury radiations are illustrated in Fig. 5, Pl. X. For the purpose of getting reliable data, it is of the utmost importance to use highly monochromatic and sufficiently intense radiation. The blue triplet, 4680, 4722 and 4810 A.U. of the zinc arc satisfies these requirements.* As however, a pure zinc arc requires a high current density for continuous running, these lines become broad and undergo self-reversal. Therefore, a zinc-mercury amalgam arc in pyrex glass was developed in the course of the investigation here described. The amalgam contains 12–15% by weight of zinc and the arc is run at a current of about 4 amps. The blue triplet is then sharp and moderately intense and nearly as efficient as 4078 A.U. for scattering and gives a structureless interference pattern with fairly sharp maxima (Fig. 6, Pl. X). For studying the longitudinal scattering of light, the arc is made in a semicircular form.

C : The Choice of Scattering Direction.—The previous authors have observed the scattered light in a direction either at right angles to the incident beam or at an angle of 180° to the latter. While the transverse direction has the advantage that a perfect background free from any back reflection could be provided for scattered radiations, it suffers from the drawback that the magnitude of the Brillouin shift is only $1/\sqrt{2}$ times that at 180° and that an

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* Lyshede and Rasmussen (1937) have observed weak satellites for the zinc lines, two satellites at ·108 and ·146 cm.$^{-1}$ for 4680, four at ·127, ·086, ·063 and ·128 cm.$^{-1}$ for 4722 and four at — ·096, — ·056 and ·100 and ·163 cm.$^{-1}$ for 4810. These satellites are so weak that 1 to 4 hours are required to record them with observable intensity, whereas the principal line is registered strongly in a minute. In view of the extreme feebleness of the satellites and the small shift from the principal line, they do not appear in the scattered light.

error of 10° in the scattering angle due to the convergence of the incident beam, which is unavoidable, broadens the displaced components by about 10%. The result of it is that even in the most favourable cases, the Brillouin components cannot be well separated from the central component. In fact, Birus (1938) who adopted the transverse direction in conjunction with an echelon grating for the determination of the relative intensities of the three components, obtained patterns in which the displaced components could be scarcely identified. It is difficult to obtain from his published photographs any reliable information about the actual breadth or the intensity of the components. Both as regards the intensity of scattering and the frequency shift, the backward direction is the most favourable for observation. It is then possible to restrict the convergence of the incident beam to about 5°; but even if the error be as much as 10°, the broadening of the shifted lines is quite negligible and it therefore becomes possible to estimate the actual breadth of the components accurately. Further, as the method enables a great depth of liquid to be utilised, the patterns are more intense and can be recorded in a shorter time than is required for the transverse direction. The chief disadvantage in making observations at 180° to the incident direction is the difficulty for avoiding completely the reflections from the rear end of the experimental tube, which give a spurious central component. But by carefully choosing the glass tube and painting the whole of it except the observation end with enamel black, it is found possible to secure a suitable background, which is nearly as satisfactory as that obtainable with transverse scattering. The experimental tube is made of pyrex glass tubing, 1½″ in diameter and 12″ long, and free from streaks. To one end is fused a thin pyrex glass plate, the other end being drawn out in the form of a horn. The liquid is introduced into the tube and rendered dust-free by distilling it in vacuum by the usual method, the process being repeated until no dust particles could be detected with the tube placed in the track of a narrow beam of sunlight. The rest of the experimental arrangement is the same as that described in the previous paper except that the tube containing the liquid replaces the gas container. During the exposure, the liquid is maintained at room temperature by surrounding it with a water jacket kept in contact with the Wood's tube by metal foils. For experiments at higher temperatures, the water jacket is replaced by a cylindrical electric heater wound on a wide glass tube. The time of exposure for recording the zinc radiations varied from two to four days.

D: Polarisation Measurements.—Gross (1930), Rao (1935), Birus (1938) and Mitra (1940) have measured the state of polarisation of the components by interposing a Nicol in the path of the transversely scattered

light and recording two spectra, one with the vibration axis of the Nicol vertical and the other with the axis horizontal. This arrangement is, however, unsatisfactory as it is difficult to maintain the source at constant running condition for both the exposures, especially as the time of exposure required is very great. It is of advantage to record both the horizontal and the vertical components simultaneously. For this purpose, the experimental arrangement given in Fig. 1 is made use of. The light from a high-vacuum horizontal
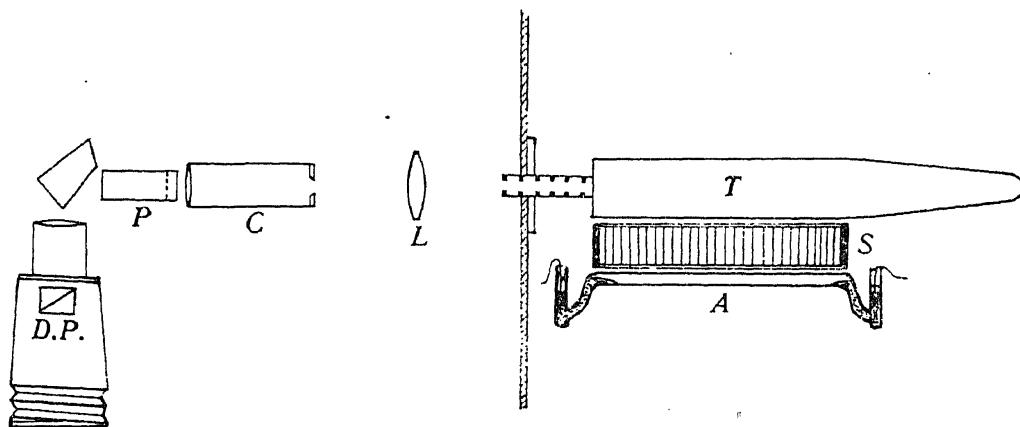

FIG. 1. Experimental Arrangement for Polarisation Studies

mercury arc A with water-cooled cathode is let fall on a Wood's tube T as a parallel beam by using a series of parallel vanes of blackened metal plates arranged in layers. The breadth of the metal plate is 50 mm. and the distance between successive plates 4 mm., giving a convergence angle of $4\frac{1}{2}°$. The corresponding convergence error is less than ·1%. Light scattered in the transverse direction by the liquid is focussed by means of a condenser L on the widely opened slit of a Hilger constant-deviation spectroscope. A glass Lummer plate P is placed between the collimating lens and the prism and a double-image prism D.P., of 1″ aperture, is interposed between the camera lens and the photographic plate in such a way that the horizontal and the vertical components are recorded side by side. As the dispersions of the Lummer plate and of the constant-deviation prism are mutually perpendicular, the polarisations produced at the optical surfaces partially annul each other, the resultant effect as determined experimentally using unpolarised light being the enhancement of the intensity of the vertical component by 12%. As a test of the arrangement, the horizontal and the vertical components of the interference pattern for light diffused transversely by a critical mixture of methyl alcohol—normal hexane were recorded. The pattern obtained is reproduced as Fig. 10 a in Pl. XI. It will be noticed that the horizontal component is extremely weak in comparison with the vertical; a microphotometric estimate of the intensity of the central components in the

two pictures gives the depolarisation ratio as ·02, which is in good agreement with the value obtained non-spectroscopically (Krishnan, 1935). The state of polarisation of the spectral components in the light scattered by cyclohexane, benzene, tetralin, phenol and glycerine has been studied.

3. *General Statement of Results*

The present paper describes the results obtained with four liquids having low depolarisation and low viscosity, *viz.*, carbon tetrachloride, cyclohexane, ethyl ether and ethyl alcohol; three others with moderate depolarisation and viscosity, *viz.*, acetone, iso-butyric acid and water, and also three liquids with a high depolarisation, *viz.*, benzene, phenol and tetralin. The interference patterns have been recorded with all radiations of the zinc-mercury arc scattered by these liquids. The records for the 4810 A.U. line of zinc obtained with a 5 mm. separation for the etalon are reproduced in Figs. 7 and 8 in Pl. X. The main features of the patterns are the following:

(1) At room temperature, the Brillouin components are fairly sharp and well defined in all liquids except in the case of carbon tetrachloride and ethyl ether in which they are broader.

(2) In well-exposed spectra of carbon tetrachloride, ether, alcohol, acetone and iso-butyric acid, a feeble component having twice the usual Brillouin displacement is observed. Its intensity is not more than 1/50th of the first order component as may be seen from the microphotometric records for alcohol (Fig. 4 *a*). Patterns of these liquids taken with 3 mm. separation for the interferometer plates showed no indication of any component with thrice the usual Brillouin displacement. In the case of aromatic liquids, the general background due to orientation scattering prevents the observation of any spectral components other than those of the first order.

(3) The general background superposed on the pattern increases in intensity with increasing depolarisation of total scattering. It however, shows a marked decrease with increasing viscosity of the liquid. For instance, with tetralin which has a high viscosity, the background is less intense than for benzene, even though the depolarisation of the light scattered by the former liquid is greater than that by benzene.

(4) In all liquids there is an undisplaced central component, the intensity of which relative to the Brillouin components varies from liquid to liquid.

(5) There is a continuum connecting the central and the Brillouin components of which the intensity is greater, the nearer the temperature of the liquid is to its boiling point. It may be clearly seen in the interference

C. S. Venkateswaran

patterns for ethyl alcohol, ethyl ether and isobutyric acid in which the central component is weak (see Figs. 7 e, f, i, k and l). This continuum is to be distinguished from the general background covering the whole pattern due to orientation scattering; in fact, in aromatic liquids in which the rotation wing is very intense, the continuum between the components is masked by the latter.
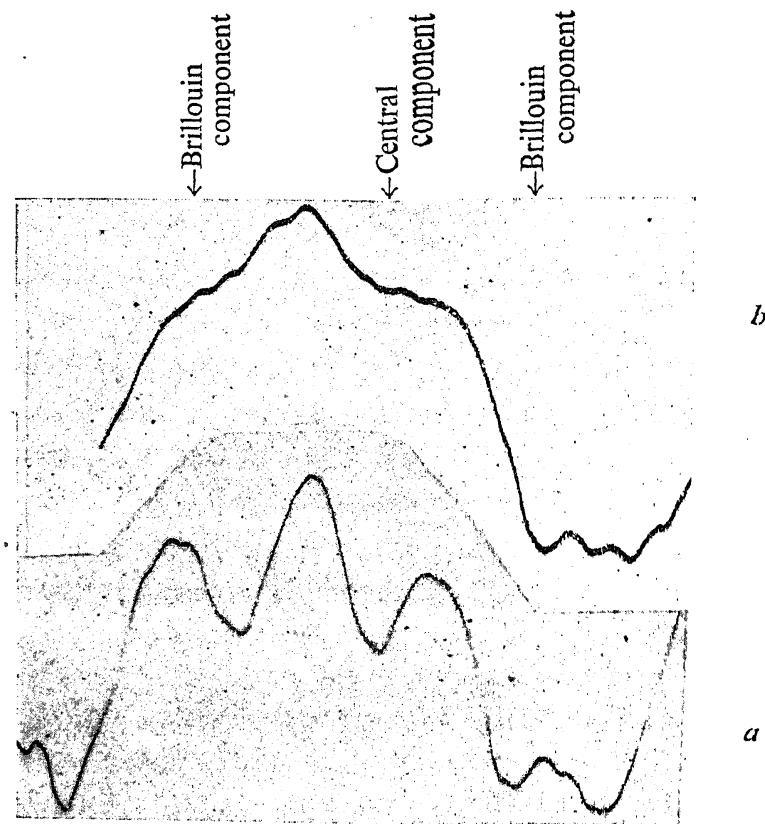

FIG. 2.   Microphotometric Curves for Acetone Scattering
(a) at 28° C. and (b) at 54° C.

4.   The Effect of Temperature

In order to study the effect of temperature on the breadth and the intensity of the three components, four typical liquids namely, acetone, ethyl alcohol, isobutyric acid and phenol have been chosen. The patterns of the scattered light were recorded for the high and low temperatures under exactly identical conditions. The effect of an increase of temperature in the four cases may be summarised as follows:—

(1) The Brillouin components in acetone become broader and shift towards the centre. As the separation between the components is small at the higher temperature, they appear as if they merge into one another (see Fig. 2). This effect is similar to the one observed by Raman and Rao (1935) in carbon tetrachloride.

(2) In the case of ethyl alcohol and iso-butyric acid (see Fig. 7 *e* and *f*, *k* and *l*), the rise of temperature is followed by a decrease in the intensity of the central component and an increase in that of the continuum. Both these liquids were examined near the boiling point; this involved raising the temperature of the acid by more than 120°. At the boiling point, the central component is scarcely present. The Brillouin components which are very intense even at the room temperature do not show any perceptible increase of intensity, though a slight broadening and a smaller frequency shift are noticeable. It appears reasonable to conclude that the increased density scattering which should result from the increased compressibility of the liquid at the higher temperature appears as an intensified continuum between the components.

(3) The intensity of the shifted components for supercooled phenol at 25° C. is very low. As the temperature is raised to 162° C., they gain considerably in intensity and remain sharp and well-defined except, at the very highest temperature, as illustrated in Fig. 8 in Pl. XI. At the same time, the central component becomes weaker. At the higher temperatures, a continuum also makes its appearance.

These results indicate that with increase in the mobility of the molecules following an increase of temperature, a part of the intensity of the central component gets released in all cases. The scattering power of the liquid also increases due to an increase in the compressibility of the medium. When the viscosity of the liquid is great as in phenol, the intensity thus made available is divided between the two coherent modified components. If the liquid is highly mobile, there is no enhancement of the intensity of the Brillouin components; but the released intensity gets distributed in the continuum. The diminution in the frequency shift of the Brillouin components with rise of temperature may be explained as due to the increase in the adiabatic compressibility and consequent diminution of the sound velocity in the medium.

5. Polarisation Studies

The polarisation pictures obtained for cyclohexane, benzene, tetralin, glycerine and phenol at 45°C. and with 4358 A.U. radiations of the mercury arc are reproduced in Figs. 10 *a*–10 *f* in Pl. XI. Similar pictures were recorded for the 4046 A.U. as well. In all the patterns the fringes are seen in the horizontal component but their intensity is always less than in the vertical component. They are extremely weak in cyclohexane as we should expect from the low depolarisation in the liquid. In benzene, phenol and tetralin, the fringes in both the components are superposed on an intense background due to rotational scattering. In none of these pictures are the Brillouin

components clearly separated from the central line; the fact that these are polarised may only be inferred from the relative intensities of the edge of the fringes in the horizontal and vertical patterns. The present investigation however, clearly establishes that the central component in the spectral pattern of the scattered light is partially polarised, the extent of depolarisation being very small either when the liquid is very mobile (*e.g.*, benzene) or when the depolarisation of the total scattering is low (*e.g.*, cyclohexane). This result modifies the conclusions of Rao (1935) and Birus (1938) based on observations with carbon tetrachloride, toluene and carbon disulphide, namely, that all the three spectral components are completely polarised. That the above authors have failed to notice the fringes in the horizontal component is probably because the pictures obtained by them are not sufficiently intense to record them; the fringes are very weak owing to the low viscosity in the case of toluene and carbon disulphide and owing to the low depolarisation in the case of carbon tetrachloride.

For making a correct estimate of the partial polarisation of the undisplaced central component, it is evidently necessary to deduct from the maximum intensity in the fringes that of the background, namely the intensity at the point of minimum illumination in the same, this being done separately for the vertical and the horizontal components. The ratio of the maxima of illumination in the horizontal and vertical components thus duly corrected, is indicated by P_{centre} and shown in column 6 of Table I, while $P_{background}$ which is the ratio of the minima of illumination in the two components is

TABLE I

Liquid	Prismatic Spectroscope			Lummer Plate			
	Temperature	$P_{wide\ slit}$	$P_{narrow\ slit}$	Temperature	P_{centre}	$P_{background}$	Intensity ratio of background to centre
Benzene ..	30°	·44	·12	45°	·10	·70	·90
Tetralin ..	30°	·54	·45	45°	·38	·78	·84
Phenol ..	45°	·66	·46	45°	·42	·75	·79
Glycerine ..	20°	·33	·30	45°	·25	..	..

shown in column 7. The intensity ratio of the minimum to the maximum of illumination in the horizontal component is also given in column 8; complete invisibility of the fringes in this component would correspond to a value unity for this ratio. To enable the significance of these results to be appreciated, the depolarisation of the transversely scattered light as determined

by an ordinary prismatic spectroscope, using a wide and a narrow slit respectively, are shown in columns 3 and 4 of the table. The former expresses the depolarisation of the total scattering by the liquid, and the latter the depolarisation after the rotational Raman scattering is excluded as completely as is possible with the instrument employed. These values are taken from papers by Saxena (1938) and Sunanda Bai (1941).

The data shown in Table I are very significant. It is seen that the depolarisation of the central fringe in the Lummer pattern is of the same order of magnitude as the depolarisation of the total scattering by the liquid *after excluding the rotational Raman scattering by its molecules.* The values of ρ_{centre} are on the other hand, smaller than $\rho_{wide\ slit}$. It will be noticed that the latter is greater for benzene than for glycerine, while the situation is reversed in respect of both ρ_{centre} and $\rho_{narrow\ slit}$. It is thus clear that the partial polarisation of the central component in the Lummer pattern has the same physical origin as the partial polarisation of the light diffused by the liquid when the rotational Raman scattering is excluded, in other words that it arises from the presence in the scattered light of a completely depolarised Q-branch of the rotational scattering. The fraction of the latter appearing as a Q-branch instead of in the rotation " wings " may be expected to become larger with increasing viscosity of the liquid, as a greater proportion of the molecules would then occupy practically fixed orientations. It follows that with increasing viscosity of the liquid, the intensity ratio of the background to the centre in the pattern should decrease. This is in agreement with observations as shown both by the photographs and by the measurements of intensity recorded in column 8 of Table I.

It is evident from the photographs that the Brillouin components for all the liquids exhibit a high degree of polarisation. The experimental evidence is however, not decisive on the question whether their polarisation is *total* as suggested by theory. Indeed, there is some evidence suggesting that in the case of mobile liquids with anisotropic molecules they may be only partially polarised. This is shown, for instance, by the fact that the central bright fringe in the horizontal component of the Lummer pattern in aromatic liquids is not very sharp. Further, if the Brillouin components had been fully polarised, we should have found that ρ_{centre} is somewhat greater than $\rho_{narrow\ slit}$ in every case; for these components are included in the latter and excluded from the former. Actually, the measurements show that ρ_{centre} and $\rho_{narrow\ slit}$ are about equal for all the liquids. Another anomaly which is noticed is that $\rho_{background}$ in every case is somewhat smaller than the value 6/7 indicated by theory for pure orientation scattering. Whether these anomalies are real or whether they arise from the unavoidable difficulties of

experimentation must remain for the present an open question. These remarks however, do not affect the validity of the main conclusions already stated.

6. The Relative Intensity of the Brillouin and the Undisplaced Components

In order to test the experimental arrangements, the light scattered by water, ethyl alcohol and benzene were examined both in the transverse and the longitudinal directions. Figs. 3 *a-c* are the microphotometric records of

(a) Longitudinal
    Hg 4358 A.U.

(b) Transverse
    Hg 4358 A.U.

(c) Longitudinal
    Zn 4810 A.U.
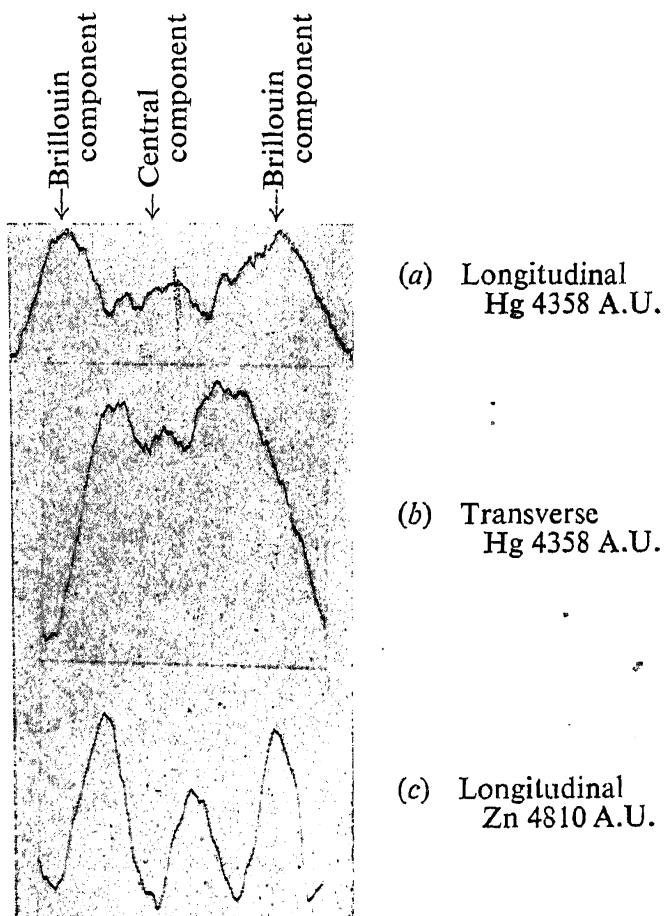
FIG. 3. Microphotometer Curves for Water Scattering

the 4358 A.U. of the mercury arc after scattering by water in these two directions and that of 4810 A.U. of the zinc arc for the backward direction only. The transverse scattering could not be examined by the zinc lines, as it is very weak. The corresponding spectra are reproduced in Fig. 9, Pl. XI. It will be seen that the central component is clearly present in all the three patterns, though definitely weaker than the displaced components. For reasons already mentioned, with the mercury radiations, the background for the transverse direction is very intense; it is distinctly less with the longitudinal scattering and almost completely absent when the zinc arc is employed. Due to the presence of this background of varying intensity, it is not possible

to make an accurate estimate of the ratios of intensities of the central components to the displaced components and compare them in the three cases. For benzene and ethyl alcohol the frequency shifts of the Brillouin components are relatively small and hence in the transverse scattering, the three components are scarcely separated from each other. However, an examination of these pictures due to the mercury and the zinc radiations gives the impression that the central line is present in them with almost the same relative intensity as in the backward scattering. In view of these results it is assumed that parasitic reflections do not sensibly affect the relative intensity of the components even in weakly scattering liquids.

Figs. 4 *a* to *g* give the microphotometric tracings of the 4810 A.U. of the zinc arc after it is scattered backwards by carbon tetrachloride, cyclohexane,
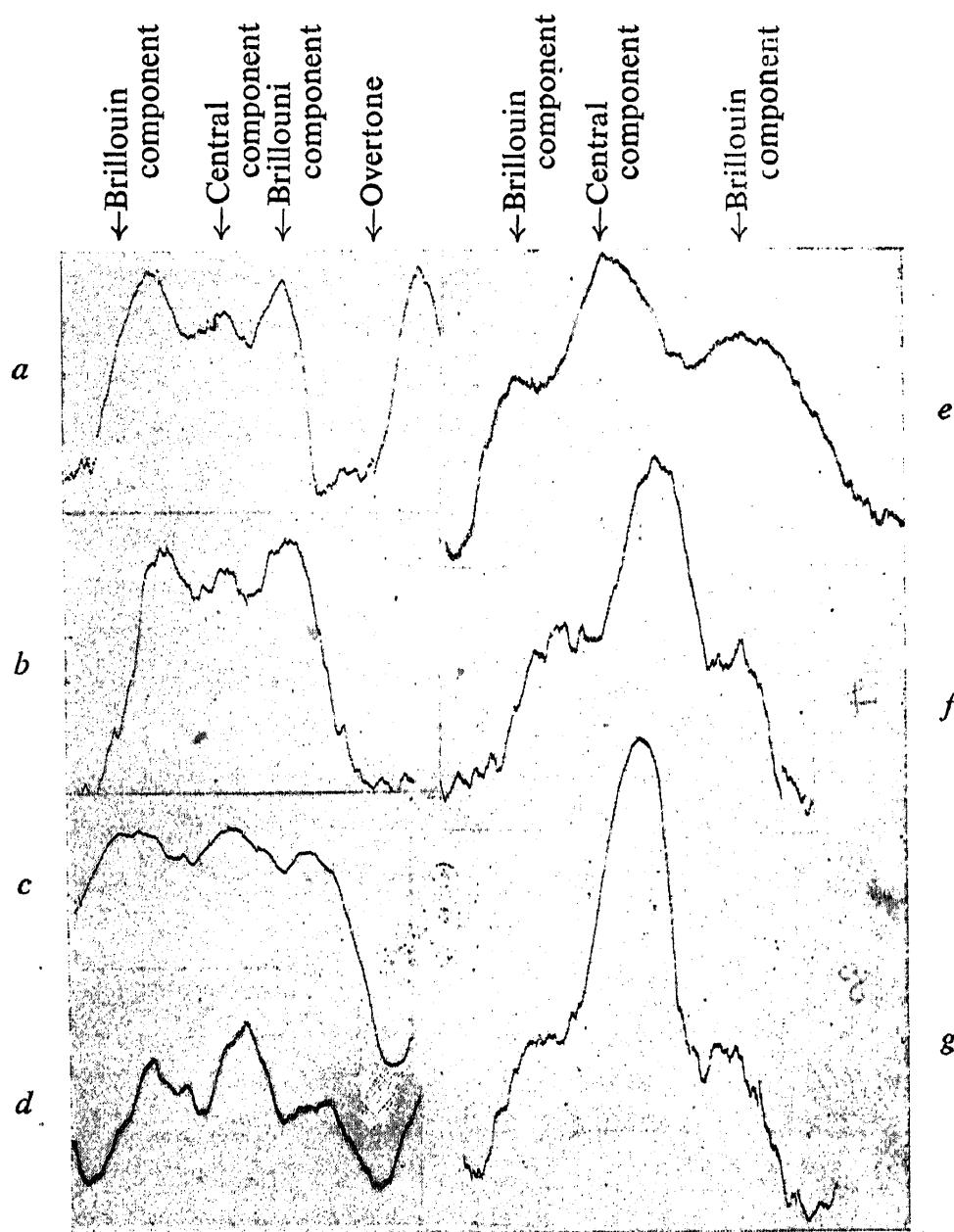

FIG. 4. Microphotometric Curves for Liquids

(*a*) Ethyl alcohol.   (*e*) Carbon tetrachloride.
(*b*) *iso*-Butyric acid.   (*f*) Tetralin.
(*c*) Ethyl ether.   (*g*) Benzene.
(*d*) Cyclohexane.

ethyl ether, ethyl alcohol, *iso*-butyric acid, benzene and tetralin. The ratio of the intensity of the central component I_C to the total intensity of the two Brillouin components, I_B, is estimated from these records by making use of the appropriate density-log intensity curve and listed in the last column of Table II. The adiabatic and isothermal compressibilities β_ϕ and β_t, η, the

TABLE II

Temperature of Observation $\backsim$ 25° C.

Liquid	η in poises	$\beta_t \times 10^6$	$\beta_\phi \times 10^6$	ρ_∞	ρ_0	$\dfrac{I_0}{I_D}$	$\dfrac{I_Q}{I_D}$	$\dfrac{I_C}{I_B}$
Carbon tetrachloride	·0084	115	78	·052	·013	·12	·03	·84
Ethyl alcohol ..	·0099	115·8	97·2	·054	·015	·13	·04	·39
Ethyl ether ..	·0021	196·7	146·9	·081	·026	·20	·06	·45
Cyclohexane ..	·0082	125	85	·080	..	·20	..	·65
Water ..	·0089	46·0	45·6	·088	..	·21	..	·36
Acetone ..	·0029	127	91	·20	·06	·56	·135	·79
Iso-butyric acid ..	·0133	..	91·5	·35	·15	1·28	·4	·45
Benzene ..	·0059	98·7	68·6	·44	·12	1·92	·29	·97
Tetralin ..	·0200	..	48·1	·54	·45	3·16	1·95	1·20

viscosity of the liquid at 25° C. and the wide and narrow slit values of depolarisation ρ_∞ and ρ_0 are given in columns 2 to 6. I_0/I_D the ratio of the orientation scattering to the density scattering and I_Q/I_D, where I_Q is the fraction of the former falling on the Q-branch calculated from the ρ_∞ and ρ_0 values, are given in columns 7 and 8. It should be mentioned that the ratio I_C/I_B is determined from the peak intensities of the three components and does not take into account their relative breadths. As the displaced components in some of these liquids are broader than the central one, these values give only the upper limit to the intensity ratio. Moreover, the presence of the continuum between the components makes the determination of the intensities less accurate. Due to these causes and the possibility of traces of parasitic light still being present, the experimental values may be slightly higher than the true ones.

7. Discussion of the Results

It will be seen from Table II that several of the liquids listed in it, *viz.*, carbon tetrachloride, cyclohexane, acetone, benzene and tetralin, give a central component which is brighter than either of the Brillouin components. As the scattering power is large for these liquids, the probability of parasitic light vitiating the results is much less than in the case of ether, alcohol and water for which weaker central components are registered. The intensity of the central component may evidently be due in part to the Q-branch of the orientation scattering coinciding with it. A comparison of the values

shown in the seventh and ninth columns of the table shows that while for liquids of low depolarisation, *viz.*, carbon tetrachloride, ether, alcohol and tetralin, I_C/I_B is much greater than I_O/I_D, the position is reversed in the case of *iso*-butyric acid, benzene and tetralin. An estimate of the intensity of the Q-branch, which may be regarded as an upper limit, may be made from the known values for ρ_∞ and ρ_0. The ratio $\dfrac{I_Q}{I_D}$ thus calculated is shown in column 8. It is seen that in every case except the two most viscous liquids *iso*-butyric acid and tetralin which have also the largest depolarisation, $\dfrac{I_Q}{I_D}$ is always much less than $\dfrac{I_C}{I_B}$. We are thus obliged to conclude that in the case of most ordinary liquids, the appearance of a central component cannot be explained as being principally due to the presence of a Q-branch for the orientation scattering. For the greater part, therefore, it must arise in some other way.

Landau and Placzek (1934) have tentatively suggested that the ratio of the central component to the total intensity of the Brillouin components arising from density fluctuations alone is given by $\gamma - 1$, where γ is the ratio of the isothermal to the adiabatic compressibility or the ratio of the specific heats of the liquid. This suggestion appears to be based on the assumption that the total density scattering of light in a liquid takes place in two steps; the first part arising as a result of the fluctuations taking place adiabatically, which appears as two equally intense displaced components and the second part due to a further increase in the density fluctuations during the restoration of thermal equilibrium in the medium, which falls on the central component. The total scattering will thus be represented as an isothermal process. Birus (1938) claims to have verified the relation of Landau and Placzek in the cases of ethyl alcohol, water and toluene. He finds support to his results from his observation that the central component is completely polarised. We have already seen that the latter statement is incorrect, and that the experimental method adopted by him cannot give the true estimate of the relative intensitities of the central and the Brillouin components.

The fundamental assumption on which the theory of Landau and Placzek is based, namely that the total intensity of the scattering in a liquid is given by the isothermal formula has been shown by Sunanda Bai in a paper appearing in these *Proceedings* to be not in accord with the actual facts. She has observed that the intensity of light scattered transversely in many liquids is intermediate between those calculated on the assumption that the density fluctuations take place in an adiabatic and an isothermal fashion.

That the considerations on which the Landau-Placzek theory is based are invalid is also indicated by the figures in Table III giving respectively the observed ratio of intensities and that calculated from the specific heats.

TABLE III

Ratio of Intensity of the Central Components to those of the Brillouin Components

Liquid	As calculated* from the Placzek-Landau Formula	As Observed	Liquid	As calculated from the Placzek-Landau Formula	As Observed
Carbon tetrachloride	·48	·84	Cyclohexane	·47	·65
			Water	·01	·36
Ethyl alcohol ..	·18	·39	Acetone	·39	·79
Ethyl ether ..	·34	·45	Benzene	·32	·97

* γ is calculated from the adiabatic and isothermal compressibilities given by Philips (1939) and Tyrer (1914).

Summary

A detailed interferometric study has been made of the spectral character of the monochromatic light in several dust-free liquids. The structureless radiations of the zinc arc, namely 4680, 4722 and 4810 A.U. are employed, and the light scattered backwards is analysed by a Fabry-Perot etalon. The liquids used are carbon tetrachloride, cyclohexane, ethyl ether, ethyl alcohol, water, *iso*-butyric acid, acetone, benzene, tetralin and phenol. Besides the two Brillouin components and an undisplaced central component, a continuum extending between these components is also noticed. This continuum is particularly intense in low boiling point liquids, like ethyl ether and ethyl alcohol. The displaced components are sharp and well defined for most of the liquids at the room temperature; but they become broad when the temperature is raised to the boiling point. The first overtone of the displaced component is also observed to be weakly present in ether, ethyl alcohol, acetone and carbon tetrachloride. The effect of increase of temperature is to lower the intensity of the central component. When the viscosity of the liquid is high, as in the case of phenol, the intensity thus set free enhances the intensity of the displaced components. When the liquid is very mobile, there is no appreciable increase in the intensity of the Brillouin components; but the continuum between these components gains intensity considerably. In the light scattered transversely by cyclohexane, benzene, tetralin, phenol and
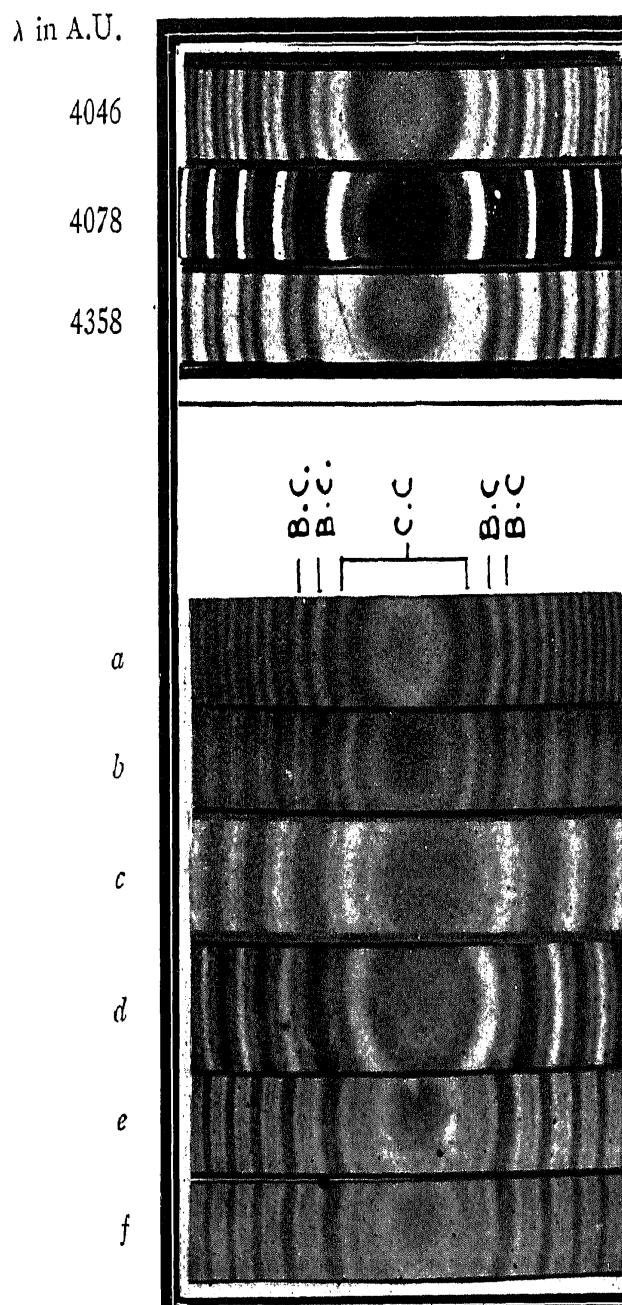
FIG. 5 Hg ARC.   FIG. 6 Zn ARC.
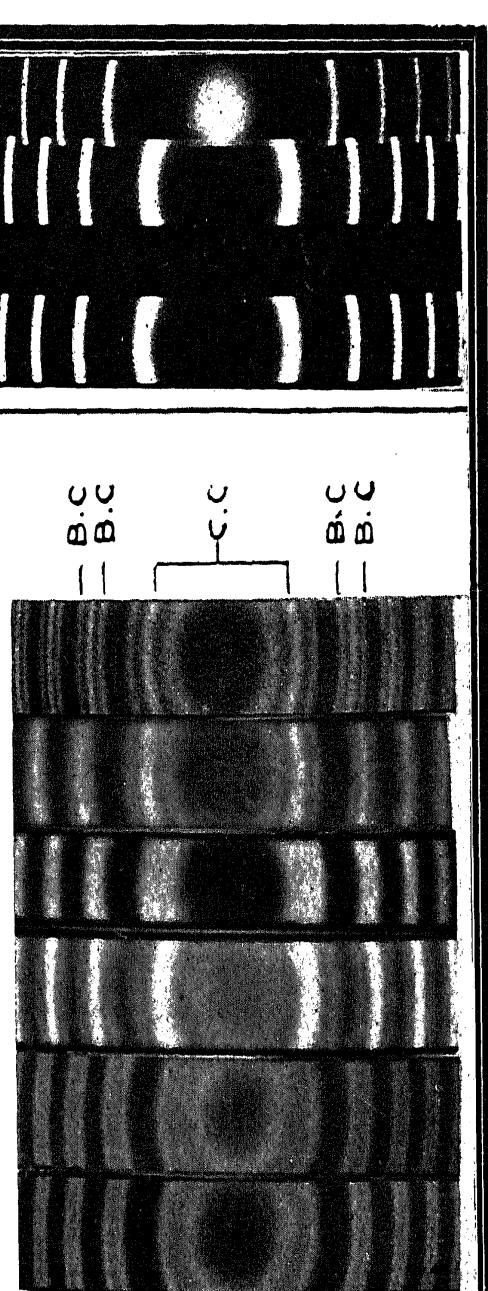
FIG. 7. Interferometer Patterns of Liquid Scattering

a. Water (30°)
b. Cyclohexane (30°)
c. Carbon tetrachloride (25°)
d. Benzene (28°)

e. Ethyl alcohol (25°)
f. Ethyl alcohol (65°)
g. Acetone (28°)
h. Acetone (54°)

i. Ethyl ether (30°)
j. Tetralin (30°)
k. iso-Butyric acid (25°)
l. iso-Butyric acid (154°C.)

C. S. Venkateswaran

Proc. Ind. Acad. Sci., A, vol. XV, Pl. XI

FIG. 8
Phenol

FIG. 9. Water
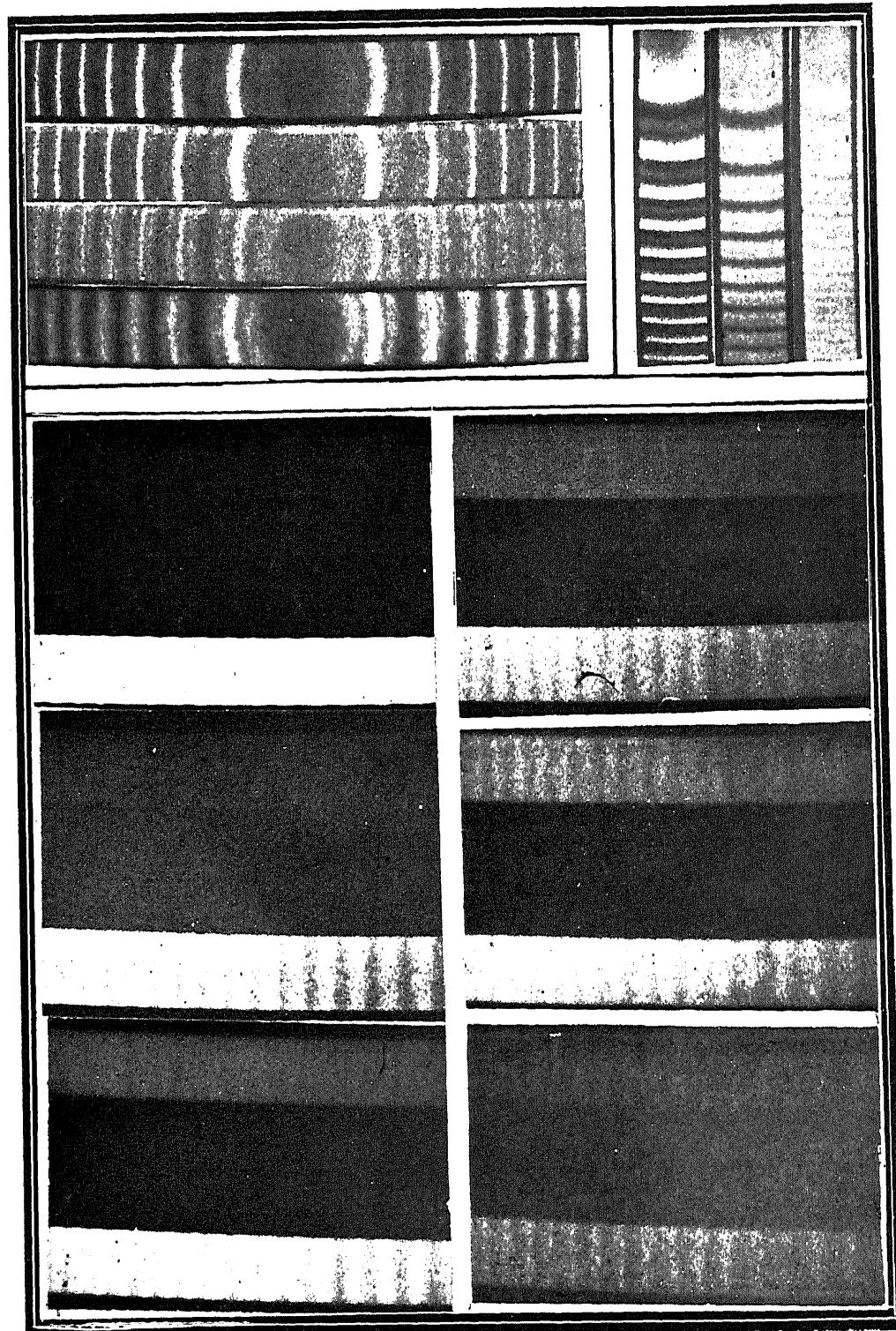
a. Hg. 4358

b. Transverse
   Scattering

c Longitudinal
   Scattering
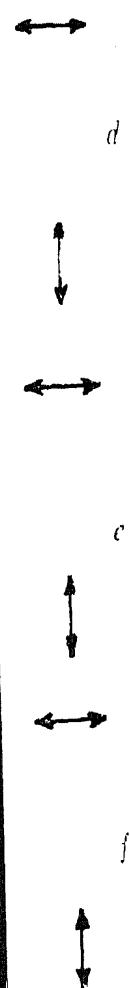
FIG. 10. Polarisation Pictures

a. Methyl alcohol-n.Hexane mixture.   b. Cyclohexane.   c. Benzene (45°).
d. Phenol (45°).   e. Tetralin (45°).   f. Glycerine (45°).