

SOME ILLUSTRATIONS OF THE THEORY OF THE CHRISTIANSEN OPTICAL FILTERS

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1. INTRODUCTION

THE functioning of the well-known Christiansen optical filters for obtaining an approximately monochromatic beam of light depends on the behaviour of an optically isotropic solid, *e.g.*, glass, when powdered and immersed in a flat-sided cell containing a liquid mixture of suitably adjusted refractive index. The principles of geometrical optics indicate that since the contents of the cell are optically heterogeneous, a pencil of white light entering the cell would suffer reflections and refractions at the boundaries between the solid and the liquid and would therefore fail to emerge in its original direction. An exception however arises when the refractive indices of the solid and the liquid in which it is immersed are identical for a particular wave-length in the spectrum. A beam of light having such wave-length would accordingly traverse the cell and emerge from it without any disturbance. Beams of other wave-lengths for which the refractive indices are unequal would be turned aside as already stated. It follows that the cell would function as a monochromatizer for the light transmitted through it.

The foregoing account of the principles on which a Christiansen filter works is based on the ideas of geometrical optics. That such an account is inadequate is shown by an observation made by Christiansen himself and described in his original paper (1884). He noticed that the regularly transmitted light when examined spectroscopically has a finite observable spectral width and that such width is notably influenced by the size of the particles of the solid powder. As the particle size is diminished, the band of transmission in the spectrum widens out, till finally with the smallest particles of all obtained by prolonged grinding and elutriation, practically the whole of the spectrum appears in the regularly transmitted light. Other factors besides the particle size also influence the observed behaviour of a Christiansen cell. Rayleigh (1899) noticed that the spectral width of the light transmitted by powdered glass immersed in a mixture of carbon disulphide and benzene increased fourfold when the thickness of the cell is diminished from

20 to 5 mm. He also found that the performance of the cell is influenced notably by the relation between the dispersive powers of the powder and the liquid. For instance, the spectral width of the regularly transmitted light increased threefold when the mixture of carbon disulphide and benzene was replaced by cedar-wood oil with a little of carbon disulphide added to it.

For a correct appreciation of the functioning of a Christiansen filter, it is necessary to approach the subject from the standpoint of the wave-theory of light. The operation of the filter clearly depends upon the relative retardations in phase of the different parts of the wave-front of light when it passes through the cell and traverses the media of different refractive indices. If the differences in phase arising in the passage of light through a single layer of particles are small enough in comparison with the wave-length of the light, the contents of the cell would behave as an optically homogeneous medium in spite of the difference in the refractive indices of the media contained in the cell. The shape and size of the particles of the solid powder, the fraction of the total volume which they occupy, the difference in the dispersive powers of the two media, the total thickness of the cell, and finally also the wave-length of the light determine the magnitude of the phase differences which arise and hence also the spectral character of the transmitted light. The energy which fails to appear in the transmitted light would naturally be found as diffracted radiation.

Basing himself partly on the results of his experimental observations and partly on theoretical considerations, N. K. Sethi (1920) suggested the following formula for the transmission coefficient of a Christiansen cell.

$$I_z = I_0 e^{-k \left(\frac{\mu_1 - \mu_2}{\lambda} \right)^2 \Delta \cdot Z}$$

where I_z is the intensity emerging from a cell of thickness Z containing particles of diameter Δ , for an intensity I_0 incident upon it, $(\mu_1 - \mu_2)$ is the difference in the refractive indices of the solid and liquid media for the wave-length λ , and k is an empirical constant. A formal derivation of Sethi's formula on purely theoretical grounds has been given by Sir C. V. Raman (1949) in these *Proceedings*, the constant k being now a quantity capable of being computed theoretically when the size, shape and the disposition of the particles in the cell are known. It should be remembered in this connection that in the derivation of the formula the particles are assumed to be much larger than the wave-length of light and therefore very far indeed from being of colloidal dimensions. The appearance of λ^{-2} instead of λ^{-4} in the exponential is therefore readily understood.

Very few of the accounts of the working of a Christiansen filter which have appeared in the literature make any reference to Sethi's fundamental contributions to the subject. In view of this, it has appeared desirable to publish in these *Proceedings* by way of a supplement to the theoretical paper of Sir C. V. Raman already mentioned, a series of spectrograms illustrating the dependance of the functioning of a Christiansen cell on the five factors mentioned above, *viz.*, the relative dispersive powers of the solid and the liquid, the wave-length of the light, the size of the particles, the thickness of the cell and the disposition of the particles within the cell, the latter including especially the proportion of the total volume of the cell occupied by the particles of the solid.

2. SPECTRAL CHARACTER OF THE TRANSMITTED LIGHT

As was observed experimentally by Sethi and is also indicated by his formula, the curve of the distribution of intensity in the spectrum of the light transmitted by a Christiansen filter has a Gaussian form. It exhibits a peak of maximum intensity at the wave-length for which the powder and liquid have equal refractive indices, while on either side it falls off with the increasing difference in these indices. The sharpness of the peak of transmission is accordingly determined by the magnitude of the difference in the dispersive powers of the two media in the vicinity of the wave-length of maximum transmission. If this difference is small, the band of transmission may be expected to extend over a wide range of the spectrum, while, *per contra*, if the powder and liquid differ widely in their dispersive powers, the transmission band should be sharp. Strictly speaking, the width of the band is not a definable quantity unless we specify the fraction of the maximum intensity at its centre which is regarded as setting a limit to its extension on either side. The particular wave-length at which the powder and its surrounding liquid have identical indices can be shifted along the spectrum by the addition of an appropriately chosen second component to the liquid contained in the cell. The shift in the spectral position of the transmission band thus brought about should be accompanied by a notable sharpening of the band if the shift is towards the violet end of spectrum. For, the dispersive powers of transparent solids and liquids usually increase rapidly with diminution in the wave-length of the light, and hence the difference in the dispersive powers of the two media may normally be expected to behave likewise. Such sharpening of the transmission band accompanying its shift in position should, according to the Sethi formula, become more pronounced by reason of the diminishing value of λ which appears in the inverse square power in the exponential of the formula.

The role played by the difference in the dispersive powers of the powder and liquid filling its interstices on the optical behaviour of a Christiansen filter is most strikingly illustrated by choosing substances for which this difference is very small. A very convenient example is furnished by powdered KCl immersed in a cell containing sym- $C_2H_2Cl_4$ to which a few drops of CCl_4 have been added to equalise the indices of the powder and liquid for any chosen part of the spectrum.

TABLE I
Refractive Indices

Substance	$n_{G'}$	n_B	n_D	n_C	$n_{G'} - n_C$
	$\lambda = 4340.6$	$\lambda = 4861$	$\lambda = 5893$	$\lambda = 6563$	
CCl_4 (20° C.) ..	1.47290	1.46755	1.46072	1.45789	.01501
KCl (18° C.) ..	1.50493	1.49835	1.49036	1.48721	.01772
sym- $C_2H_2Cl_4$ (15° C.) ..	1.50954	1.50381	1.49678	1.49302	.01652

It will be seen from Table I that in the visible spectrum, KCl has a slightly lower index than sym- $C_2H_2Cl_4$ but a slightly higher dispersive power. The addition of a little CCl_4 brings the refractive indices of the powder and liquid into coincidence almost simultaneously throughout the whole spectrum. Actually, such coincidence first occurs at the violet end of the spectrum and then shifts towards the red. If the experiments are made with the material placed inside a flat cell, colours are indeed noticeable in the transmitted light, but they are not very vivid, while the halo of diffracted light seen surrounding the source exhibits hardly any chromatic effects. The great extension of the spectral band of transmission in the present case becomes directly evident to observation if the powder and liquid are placed together in a hollow glass prism of 60° angle and the straight bright filament of an electric lamp is viewed through the mixture. The filamentary source is then seen drawn out into a spectral band of considerable extension whose position alters with the quantity of CCl_4 present or with the temperature of the cell. On the other hand, if a similar experiment be made with materials exhibiting a large difference in the dispersive powers, e.g., hexamethylene-tetramine powder immersed in a mixture of CS_2 and C_6H_6 in a flat cell, brilliant chromatic effects are observed in the transmitted light, and the complementary colours are seen in the diffraction halo; viewed through a hollow prism, the filament source is seen as a sharp monochromatic line.

A progressive narrowing of the spectral band of transmission occurs when powdered KCl is placed in a flat-sided cell and the same is filled up with one or other of a series of liquid mixtures the principal component of which has a higher dispersive power than KCl in ascending sequence. Tetrachloroethylene (C_2Cl_4), xylene, toluene, benzene and carbon disulphide form such a series. As the refractive indices of these liquids are also greater than those of KCl, the addition of a second component of lower index is necessary to obtain a transmission band in the visible spectrum. Carbon tetrachloride is suitable for this purpose in the case of the first four liquids of the series, while acetone is a more suitable diluent for CS_2 . The addition of the second component diminishes the dispersive power of the mixture, but it nevertheless remains higher than that of KCl. The residual difference is small when C_2Cl_4 is the principal component, larger with xylene, and quite large for the CS_2 -acetone mixture. The progressive sharpening of the band of transmission is illustrated in Figs. 1, 2 and 3 (Plate I) for these three cases respectively. Each figure exhibits a set of spectra in which the position of the transmission band had been shifted towards the violet by graduated additions of the second component. It will be seen that very broad transmission bands appear in Fig. 1, and that they are much sharper in Fig. 2 and quite sharp in Fig. 3. Each series of spectra also indicates the notable sharpening of the bands when they are shifted towards the violet by additions of the second component. Such sharpening is actually more striking than might seem at first sight, since the dispersive power of the spectrograph employed increases greatly towards the shorter wave-lengths. A comparison with the wave-length scale recorded in the pictures enables the situation to be better appreciated. It should be mentioned that the spectrograms reproduced in Plate I were obtained by focussing the image of a narrow slit illuminated by an electric arc on the slit of the instrument itself, the Christiansen cell being placed immediately in front of the focussing lens.

A notable feature in all the three sets of spectrograms reproduced in the Plate is that the transmission band is accompanied by a faint continuous spectrum extending unsymmetrically on either side of it. Such extension can be traced right up to the limit of the sensitivity of the photographic plate at the red end of the spectrum, whereas it falls off very quickly towards the violet. It does not appear likely that this feature is due to an admixture of the light regularly transmitted by the cell with the light diffracted in other directions. For, the optical arrangements described above were designed to prevent such an admixture. It would seem rather that the feature referred to is characteristic of the Christiansen effect itself. Indeed, one could have anticipated it from the Sethi formula, since the expression appearing as

the exponential in the formula increases much more slowly with increasing than with diminishing wave-lengths.

3. INFLUENCE OF PARTICLE SIZE

The dependence of the spectral width of the transmitted light on the size of the particles of the powder is illustrated in the series of spectra reproduced in Figs. 4, 5 and 6, Plate II. These spectrograms were obtained with glass powder immersed in a flat cell containing a mixture of CS_2 and C_6H_6 . The thickness of the cell in all the cases was the same, *viz.*, 3 mm., but the particle sizes however were very different. The series of spectrograms in Fig. 4 were taken with particles of an average dimension of $9\ \mu$, obtained by prolonged grinding and elutriation of the powdered material with water. The spectrograms reproduced in Fig. 5 were obtained with particles of larger size averaging $100\ \mu$, and those in Fig. 6 with particles still larger in size, *viz.*, $270\ \mu$. These three sets of spectra strikingly exhibit the broadening of transmission band with diminishing particle size indicated by theory. The six spectra in each set exhibit the effect of successive increases of the proportion of C_6H_6 in its mixture with CS_2 . The notable sharpening of the band thus resulting will be evident on a reference to the wave-length scale reproduced with the spectra.

Figs. 7 and 8, Plate III similarly illustrate the effect of increasing the thickness of the Christiansen filter. The spectra reproduced in it were recorded with powdered glass of the same average particle size, *viz.*, $100\ \mu$ in both cases, but with two different thicknesses of the cell, *viz.*, 3 mm. in Fig. 7 and 10 mm. in Fig. 8. It will be noticed that the wave-length range of transmission is reduced roughly in inverse proportion to the thickness of the cell employed. These spectrograms likewise illustrate the very great spectral sharpening when the transmission band is shifted towards the violet end of the spectrum by additional quantities of the second component in the liquid mixture.

It should be mentioned in this connection that the influence of the thickness of the transmitting layer in the Christiansen experiment can be very well demonstrated by using a hollow prism instead of a flat cell and viewing a bright linear source of light with the eye placed close to the second surface of the prism. As the observer moves his eye along the surface of the prism from the edge towards the base, the image of the source seen through the prism and drawn out into a spectrum rapidly sharpens into a monochromatic band of very small width.

4. INFLUENCE OF LOOSE PACKING

The spectrograms reproduced in Figs. 1 to 8 in Plates I, II and III were recorded when the solid powder in the Christiansen cell had settled down

completely and hence the liquid occupied only the interstices between the solid particles. In such a case the powder is rather closely packed, and the volume occupied by it exceeds that of the liquid. A more open order of distribution of the solid particles in which the volume occupied by the liquid is comparable with or in excess of that occupied by the solid can be obtained as follows. The materials placed in the Christiansen cell are so chosen that the solid and the liquid mixture do not differ greatly in density, *viz.*, hexamine powder and a $\text{CS}_2\text{-C}_6\text{H}_6$ mixture. An excess of the liquid is put into the cell and a suitable quantity of the powder is distributed uniformly through its volume by vigorously shaking the cell. The powder remains suspended in the liquid for an appreciable time, since the densities of the two media are not different. As the powder settles down slowly, the quantity of the solid in the upper portion of the liquid column decreases gradually and the packing of the solid particles becomes increasingly less dense in this part of the liquid. Such a change should be accompanied by a gradual broadening of the spectral width of transmission through this part of the cell. For, it follows from the theoretical considerations leading to the Sethi formula that the constant k diminishes as the quantity of powder in suspension in the liquid is reduced, ultimately tending to zero when the liquid is nearly free of all the suspended material. The successive stages of the increase in the spectral width of transmission can be readily observed by employing a hollow prism as the container, as mentioned above. In the initial stages, when all the solid in the cell is suspended in the liquid, the image of the source appears as almost perfectly monochromatic when viewed through the prism. As more and more of the powder settles down, the image spreads out and finally extends over the whole spectrum. These changes can be recorded as follows. Light emerging from a narrow illuminated slit passes through the upper part of a column of the mixture contained in a flat cell and is focussed on the slit of a spectrograph. The cell is at first vigorously shaken and the transmission spectrum is recorded at successive short intervals of time as the powder settles down slowly. Such a series of spectrograms is reproduced in Fig. 9 in Plate IV and exhibits conspicuously the progressive widening of the spectral region of transmission. It is interesting to note the pronounced asymmetry in the distribution of intensity in the spectrum. In the final stages of the experiment, only very fine particles will remain suspended in the liquid. Their small size would naturally tend to enhance to a certain extent the width of the spectral band of transmission resulting from loose packing of the solid. Hence we have to bear in mind that the observed spectral character of the transmitted light is determined by the joint effect of these two factors, and not by the looseness of the packing alone.

5. THE CASE OF VERY LARGE PARTICLES

The Sethi formula indicates that the light transmitted by a Christiansen cell would have a continuous spectral distribution of intensity. It is not difficult to understand why such a result is reached despite the assumptions made in deriving the formula, *viz.*, that the particles in the cell are approximately uniform in size and are not small in comparison with the wavelength of light. In the first place, it is assumed that each of the successive layers into which the contents of the cell can be conceived to be divided contains a great number of particles. It is assumed further that the cell contains a great number of such layers acting independently. As a consequence of these assumptions, the fluctuations of intensity with wavelength which would be exhibited in the light diffracted by a single particle are completely smoothed-out when the integrated effects of all of them acting together are evaluated. If, however, the particles contained in the cell are very large and the total thickness of the cell is sufficient to include only a few layers of such particles, it is not to be expected that a smoothed-out distribution of intensity would be observed in the spectrum of the transmitted light. Fluctuations of intensity dependent on the size of the particles as well as their number and arrangement in the cell may be expected to be observed. Effects of the kind indicated above have actually been observed in the transmission spectrum of a Christiansen cell containing particles of diameter of 1, 2 and 3 mm. respectively in a cell 10 mm. thick filled with a CS_2 -acetone mixture. A group of spectrograms of the transmitted light in the particular case of particles of average size 3 mm. as recorded with increasing exposures is reproduced in Fig. 10, Plate IV. With small exposures, only the most intense part of the transmitted light is recorded. This is found to extend over a very narrow range of wave-lengths as is to be expected from the Sethi formula. In the succeeding spectrograms taken with increasing exposures, the transmission spectrum is found to broaden out asymmetrically, showing less extension towards the violet end of the spectrum. This of course is due to the more rapid increase of the difference in the dispersive powers of the solid and liquid media on the violet side of the maximum of transmission than on the red side. These spectrograms clearly exhibit quasi-periodic fluctuations in the spectral distribution of the transmitted light. In the first few spectrograms, close to the most intense part of the transmission we can detect a series of maxima and minima of intensity. Observations with particles respectively of 1, 2 and 3 mm. diameter show that the spacing of these bands varies inversely with the particle size. Such a result is to be expected from theoretical considerations based on wave-optical principles.

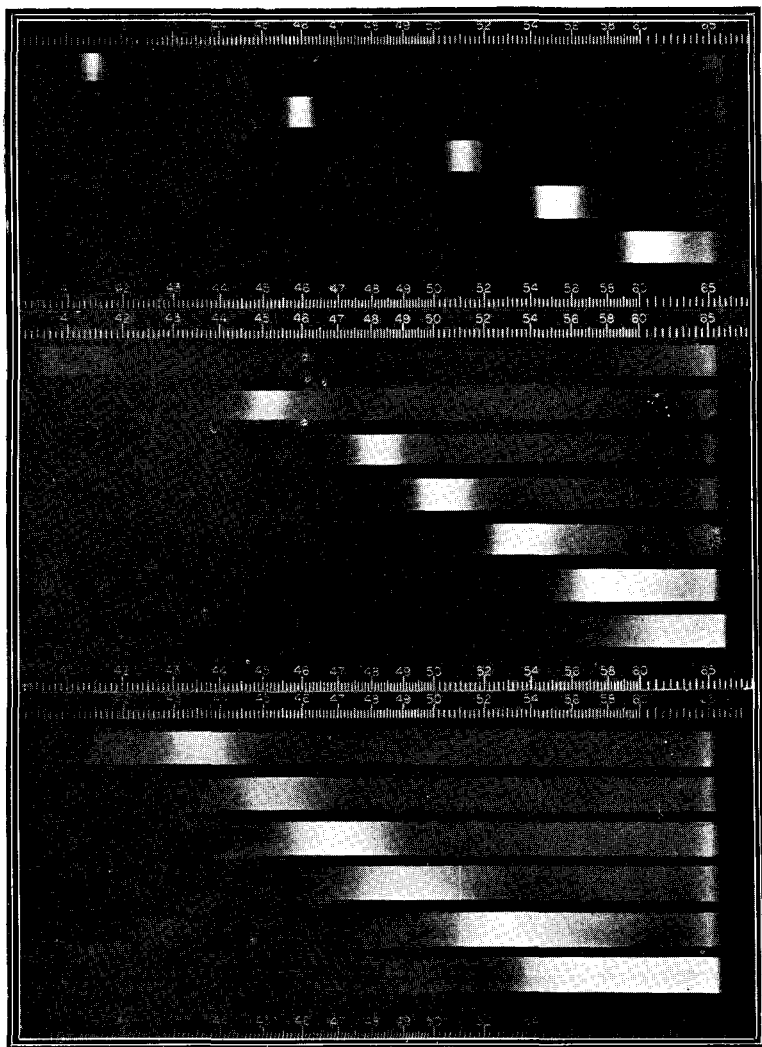


FIG. 3

FIG. 2

FIG. 1

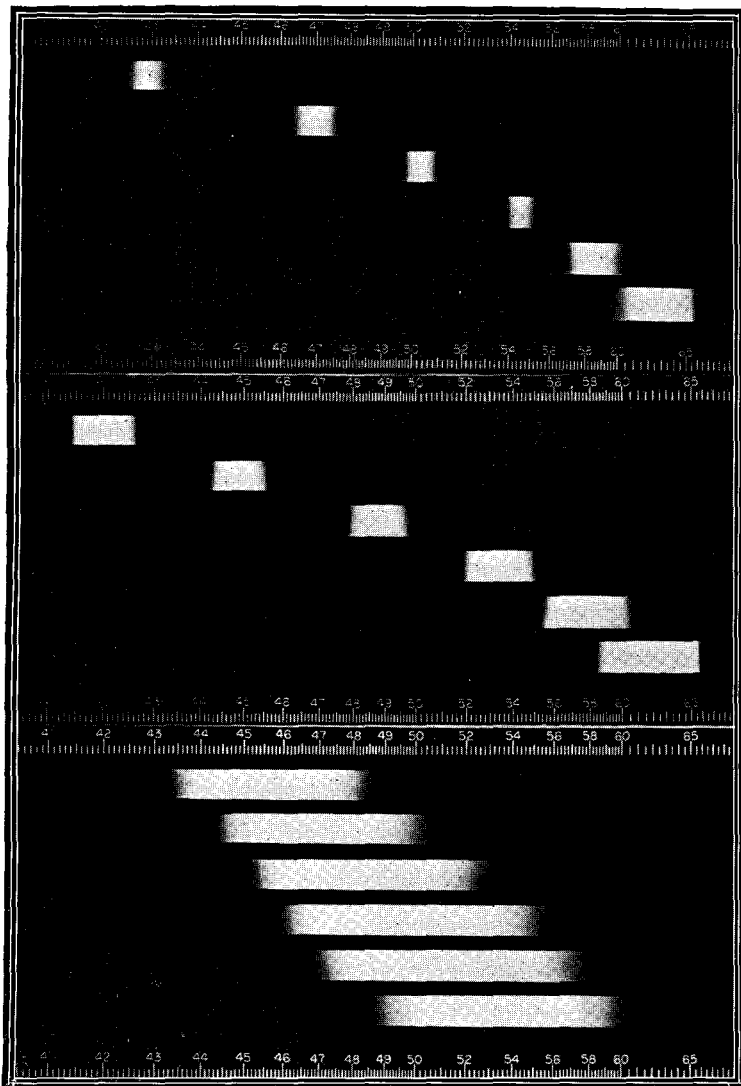


FIG. 6

FIG. 5

FIG. 4

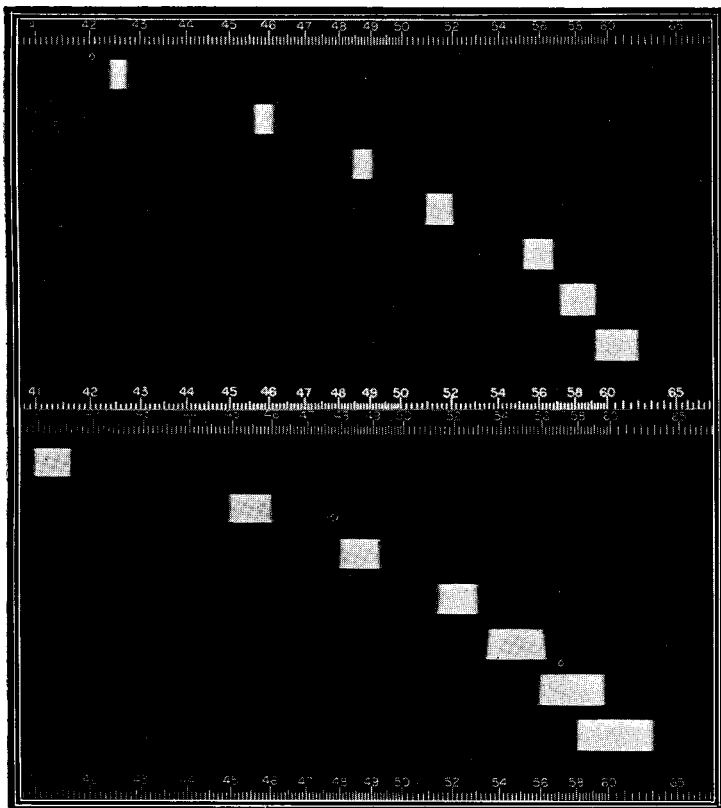


FIG. 8

FIG. 7

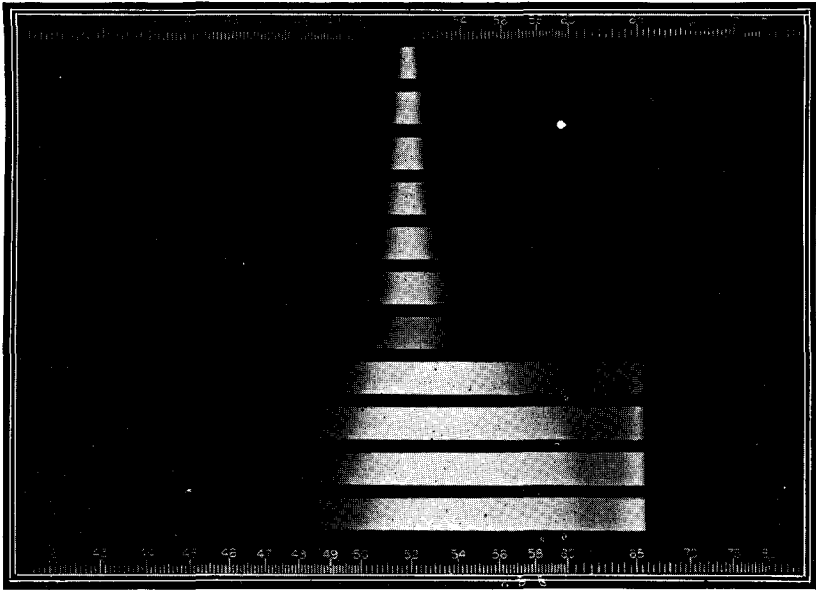


FIG. 9

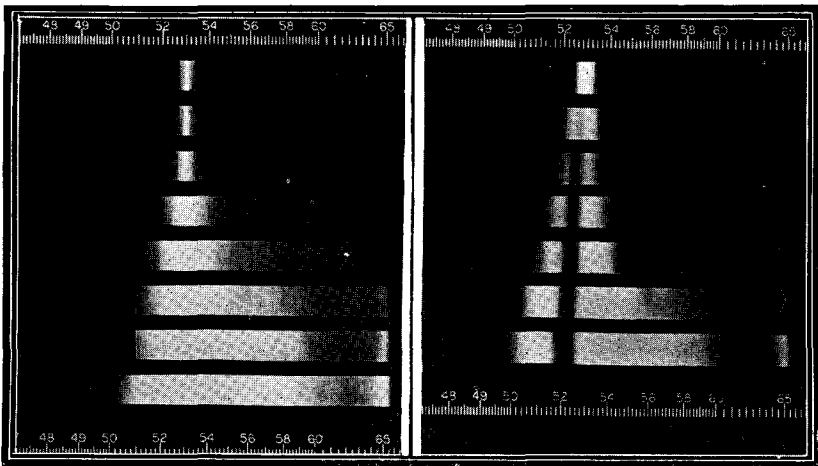


FIG. 10

FIG. 11

6. SPECTRAL CHARACTER OF THE DIFFRACTED LIGHT

The first spectrogram in the group of spectra reproduced in Fig. 11, Plate IV is of the light transmitted by particles of glass of average size 270μ contained in a cell 10 mm. thick filled with a mixture of CS_2 and acetone. The succeeding spectrograms show the changes observed in the spectral character of the diffracted light as the direction of observation becomes increasingly inclined to the direction of the incident light. It will be seen that the spectrum consists of a central dark band flanked on either side by two bright bands. Both the dark and the bright bands are found to broaden out with increasing obliquity of observation. But the bright band on the side of greater wave-lengths ultimately extends to the very limit of the sensitivity of the plate, while on the side of the shorter wave-lengths the extension is comparatively little. Essentially similar observations were made by Sethi and are to be found illustrated in his paper. He also correlated these spectral characteristics of the diffracted light with the colours observed in the halo in different directions.

To understand the spectral character of the diffracted light in different directions, we have to take note of the following points. In the first place, the transmitted and diffracted radiations should exhibit complementary effects; the more freely light of a given wave-length is transmitted through the cell, the less would be the fraction of the same appearing as diffracted light. Secondly, we have to consider the distribution of the diffracted radiation in different directions. As was noticed by Sethi, the angular spread of the diffracted light is least for wave-lengths in the vicinity of that most freely transmitted and increases progressively as the wave-length is altered in either direction, the more so towards shorter wave-lengths since the phase differences arising from the passage of light through the particles of the powder are largest in that case. Such increase in the angular spread of the diffracted light is necessarily accompanied by a fall of its intensity as observed in particular directions. In each case, there is fairly a well-defined angle beyond which the intensity of the diffracted light becomes negligible. Taking these factors into account, the features observed in the spectrograms reproduced in Fig. 11, Plate IV are readily intelligible.

In conclusion, I wish to express my grateful thanks to Prof. Sir C. V. Raman for suggesting the problem and his guidance in the course of the work.

SUMMARY

Studies of the spectral character of the light transmitted by Christiansen filters in various circumstances have been carried out and are discussed in

the light of wave-optical theory. The results are illustrated by a series of spectrograms reproduced with the paper. When the dispersive powers of the solid powder and of the liquid in which it is immersed are nearly equal, the transmission extends over a wide range of the spectrum. *Per contra*, it narrows down with increasing difference in the dispersive powers. The influence of the same factor is also strikingly evident when the wave-length of maximum transmission is shifted towards shorter wave-lengths by changing the composition of the liquid. A pronounced asymmetry is also noticeable in the spectral distribution of intensity of the transmitted light. The size of the particles of the powder and the total thickness of the filter are other factors which influence the spectral range of transmission. Of particular interest are the results obtained when the powder is only loosely suspended in the liquid and not tightly packed.

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