

THE STRUCTURE AND OPTICAL BEHAVIOUR OF SOME NATURAL AND SYNTHETIC FIBRES

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1. INTRODUCTION

COTTON, silk and wool which are the fibres commonly used in the textile industry have recently found competitors in various synthetic products, viz., rayon, nylon and the rest. The technological importance of a knowledge of the properties of all such fibres has naturally resulted in a great deal of attention being devoted to the investigation of their molecular structure. The principal tools employed in such studies have been the polarisation microscope, the X-ray diffraction-camera and the electron microscope. In the present paper we describe a different method of study, which judging from the results so far obtained and now presented, appears to be very promising. The technique employed is to record the *optical diffraction pattern* of a single fibre with arrangements generally similar to those used in an X-ray diffraction camera, but with the difference that visible light is used instead of X-rays. The incident light beam may consist of either unpolarised light or of light polarised with its vibration direction making any desired angle with the fibre axis. It is also possible to place an analyser in the path of the light diffracted by the fibre, with its vibration direction making any desired angle with the fibre axis. A great variety of patterns exhibiting observable differences may thus be recorded even in the case of a single fibre. The two most interesting and characteristic patterns are those obtained respectively with unpolarised light and with the fibre placed between crossed polaroids with its axis bisecting the angle between their vibration directions.

2. EXPERIMENTAL TECHNIQUE

A narrow pencil of sunlight emerging through an aperture of 1.5 mm. diameter is focussed by means of a photographic lens placed 124 cm. away from it to a small brilliant image in the plane of the photographic film which is 40 cm. from the lens. The fibre itself is placed in the path of the condensed beam of light 12 cm. away from the plane of the recording film; a short length of the fibre is thus completely bathed in the pencil of incident light. Two stops in the path of the incident beam placed respectively before and

after the lens ensure that in the absence of the fibre, the field is completely dark except in the immediate vicinity of the focussed image of the source. The fibre itself is attached to a metal frame, and by manipulating a pair of screws can be held perfectly straight without any undue tension being exerted on it. An exposure of the order of a few seconds is adequate to record the principal feature in the diffraction pattern, namely a bright straight streak running transversely to the direction of the fibre. Longer exposures of the order of a few minutes are needed to record the fainter outlying features of the pattern which reveal the inner structure of the fibre. When recording them, it is useful to place a small blackened stop immediately in front of the photographic film to prevent its fogging by over-exposure of the central spot. Replacing the photographic film by a ground glass plate and viewing the latter through a polaroid held in front of the eye, one may look for evidence of polarisation in the diffracted light in various parts of the field. By inserting a polariser in front of the lens, the light falling on the fibre may itself be polarised in any chosen direction of vibration, *viz.*, parallel or perpendicular to the direction of the fibre or making an angle of 45° with it. The state of polarisation of the diffracted light may then be observed on the screen through a suitably held analyser. By placing the analyser immediately after the fibre in a crossed position with respect to the polariser, the depolarised component of the diffracted radiations may be recorded photographically. It is usually convenient to set the vibration directions of the polariser and analyser at an angle of 45° with the fibre axis on either side. One is then not troubled with the scattering of light by the material of the analysing polaroid.

Various improvements naturally suggest themselves, as for example, to use instead of a flat film a cylindrical camera with the diffracting fibre centred along its axis and providing arrangements for rotating the fibre or moving it up and down along its length. Such a cylindrical camera with appropriate arrangements for collimating the light beam should enable the complete diffraction pattern to be recorded. A further improvement which seems likely to prove useful is to employ, instead of sunlight, truly monochromatic light of sufficient intensity obtained with a pointolite mercury arc and a colour filter. We intend to return to the subject after effecting various improvements in technique, but have thought it desirable not to delay the publication of the present preliminary report.

3. SOME THEORETICAL CONSIDERATIONS

It will be useful here to recall briefly the nature of the diffraction pattern of a dielectric cylinder in the ideal case when it consists of homogeneous

and isotropic material and has a uniform circular cross-section. If the diameter of the cylinder is sufficiently large in relation to the wave-length, the diffraction pattern may be considered roughly as consisting of two patterns which are superposed on each other; the first part is the pattern produced by a narrow obstacle with parallel edges which by Babinet's principle is the same as the diffraction pattern of a narrow slit; the second part arises from light which finds entry into the cylinder and emerges therefrom after suffering refractions and possibly also one or more internal reflections. Interferences would necessarily arise between the radiations diffracted in any given direction but which have traversed different optical paths. Monochromatic light would, in general, have to be used to fully reveal the characters of these interferences. It is obvious, however, that in all cases the diffracted light would appear as a fan of rays lying in a plane strictly transverse to the direction of the fibre, the rest of the field remaining perfectly dark.

If in the ideal case under consideration the cylinder though homogeneous and uniform is of birefringent material, the same results as those stated above would still be valid except that the distribution of intensity as well as the phase of the vibration in the fan of diffracted rays in various directions would depend upon the direction of the electric vector in the incident light beam. Hence, if the incident light be polarised, the fan of diffracted rays would in general be elliptically polarised and would therefore not be extinguished when observed through an analyser. Exceptions, however, would arise in special cases, as for example if the vibration direction of the incident light is parallel or perpendicular to the fibre-axis and coincides with an axis of the optical indicatrix of the material of the cylinder. In such cases we may expect the fan of rays to be more or less completely extinguished when the polariser and analyser are crossed.

The foregoing statements are very elegantly illustrated by the photographs reproduced as Figs. 11 and 12 in Plate XII. Fig. 12 was obtained by enclosing fragments of glass wool orientated at random between two glass plates and viewing a small distant aperture illuminated by the light of a sodium vapour lamp with the plates held close to the eye of the observer or the lens of a camera. Each piece of glass wool gives rise to a streak of diffracted light running through the field in a direction transverse to its length. A great many radial streaks accordingly appear in the field of view. Owing to the fact that all the fibres are of nearly equal diameter, the individual streaks conspire to build up a diffraction pattern consisting of a bright central disk surrounded by a system of rings of gradually diminishing intensity. The

photograph shows at least two such rings clearly with a few faint and rather irregular ones further out. The entire pattern disappears when polaroids in a crossed position are placed one on either side of the plates containing the diffracting material. Such disappearance is a consequence of the fact that the material of the glass wool is optically isotropic.

If, instead of glass wool, a quantity of shredded rayon or cocoon silk is held between the two glass plates with the individual fibres orientated at random, we observe a diffraction pattern of the same general character as that reproduced in Fig. 12 with glass wool. But when the combination is placed between crossed polaroids, the pattern of rings seen surrounding the source vanishes; nevertheless, a notable amount of diffracted light in the form of bright streaks continues to be visible in the field and appears as a cross with bright and dark arms. This effect is illustrated for the case of rayon in Fig. 11, Plate XII. The bright arms of the cross in the figure bisect the angles between the vibration directions of the polaroids. Cocoon silk exhibits similar effects, the origin of which is clearly ascribable to the birefringence of the fibres. Surprisingly enough, however, the bright cross illustrated in Fig. 11 is not observed with either cotton or wool. That these fibres are birefringent is indeed shown by the presence of diffracted light in the field when viewed through crossed polaroids. Why the cross is not shown by either cotton or wool in these circumstances will be explained later.

Returning now to the elementary considerations set forth earlier, it is obvious that even if the dielectric cylinder consists of optically non-homogeneous material, the fan of diffracted rays would be confined to a sheet transverse to the direction of the fibre provided that both the dimensions and the structure of the cylinder are invariable along its length. Any departure from such uniformity may be expected to reveal itself by the appearance of diffracted streams of light in various other directions. From the character of the diffraction pattern observed, we can draw inferences regarding the nature of the non-uniformities giving rise to them.

4. EXPERIMENTAL RESULTS

(i) *Glass Wool*

As already indicated, glass wool gives with the arrangements described in Section 2 an intense straight streak running transversely to the direction of the fibre. However, with prolonged exposures, faint bands running nearly parallel to the course of the streak but exhibiting irregularities in their intensity are recorded photographically. These features indicate that though the material is in the main, isotropic and homogeneous, variations either in composition or diameter or of both exist of sufficient magnitude to give rise

to observable effects. This report being a preliminary one, we have not thought it worthwhile to pursue the matter further or to reproduce any of the photographs obtained with glass wool.

(ii) *Cocoon Silk*

Figs. 9 and 10 in Plate XI reproduce photographs obtained with cocoon silk, Fig. 9 being recorded with incident unpolarised light and Fig. 10 with crossed polaroids set at 45° to the direction of the fibre. The former was taken with 30 seconds exposure and the latter with about 3 minutes. The birefringence of the silk fibre is thus clearly exhibited. The diffraction pattern, however, is not essentially different from that expected for the ideal case of a cylinder of uniform cross-section and structure. Actually, the portion of the fibre bathed by the incident light was seen to be visibly uniform. It should be mentioned that the streak of diffracted radiation exhibited brilliant colours which, in general, were very different when observed respectively with unpolarised light and between crossed polaroids.

(iii) *Rayon*

The diffraction patterns of rayon are markedly different from those of cocoon silk (see Figs. 7 and 8 in Plate X). The central bright streak is bordered on either side by a series of bands running parallel to it, so much so that in heavily exposed photographs, the diffraction pattern appears as a broad band with fainter bands lying further out. In order to exhibit clearly the central bright streak in Fig. 7, the negative had to be heavily printed so that the fainter outlying bands were completely blotted out. The intensity of the bands varies notably along their length. This feature is even more conspicuous in the photograph recorded between crossed polaroids (Fig. 8).

(iv) *Chrysotile (asbestos)*

The finest possible fibre was isolated from a lustrous specimen of the mineral and its diffraction pattern was recorded (Figs. 5 and 6 in Plate IX). As seen visually, the brightest part of the diffraction pattern is the straight bright streak. When recorded with adequate exposures, however, it is evident that the pattern consists of a great many streaks of diffracted radiation running at various angles to the principal streak, their intensity decreasing with increasing distance from the centre of the pattern. With the exposures given, the central part of the pattern was heavily recorded while the outer streaks appear only faintly. Thus, it was not possible to make a print for reproduction which would exhibit both the inner and the outer streaks clearly. However, the general nature of the pattern can be seen from Fig. 5. The pattern as recorded with the crossed polaroids shows the central

streak very clearly, while the marginal parts were hardly visible, presumably due to their faintness. We note that in Fig. 6 the central streak appears curiously curved along its length.

(v) *Cotton*

By far the most interesting patterns recorded in the present investigation are those of single cotton fibres (Plate VII). Fig. 1 shows the pattern as observed in unpolarised light and Fig. 2 as observed between crossed polaroids; the former was recorded with about 30 seconds and the latter about 4 minutes exposure. The central bright streak which is clearly seen in Fig. 1 has almost completely vanished in Fig. 2. The radiating streaks of diffracted light which are very conspicuous in Fig. 1 are much less evident in Fig. 2: the latter seems to consist principally of wavy bands running approximately in a parallel direction.

(vi) *Wool*

The patterns of wool recorded respectively with unpolarised light and with crossed polaroids are reproduced in Figs. 3 and 4 in Plate VIII. They are very similar to each other except that the central streak which is very clearly seen in Fig. 3 has completely disappeared in Fig. 4. In the latter case, however, the general intensity is smaller and correspondingly longer exposures are needed for recording the observed pattern. We may mention here some interesting observations made by us visually. With the incident light polarised and its vibration direction parallel to the fibre, the central streak is almost completely extinguished while the outer parts of the pattern are visible though enfeebled in intensity when the pattern is viewed through a crossed polaroid. Much the same result is observed between crossed polaroids when the vibration direction of the incident beam is perpendicular to the fibre. On the other hand, when the vibration direction of the incident light is inclined at 45° to the fibre direction, the appearance of the field is complementary in the two cases when the analysing polaroid is respectively in the parallel and in the crossed position. In the former case, the central streak is brightly visible while the surrounding field is much enfeebled; in the latter case, the central streak disappears while the marginal parts brighten up. A striking feature in Figs. 3 and 4 to which attention may be specially drawn is the lack of symmetry of the pattern on the two sides of the principal streak. Such an effect is not noticeable in the case of the other fibres studied.

From a comparison of the diffraction patterns for the individual fibres, it will be readily understood why aggregates of rayon and silk exhibit the phenomenon illustrated in Fig. 11 of Plate XII, whereas cotton and wool

do not. In all the four cases, the diffraction disk and rings in the central part of the field (illustrated in Fig. 12 of the same Plate for the case of glass wool) disappear when the material is placed between crossed polaroids. The residual light in the field then consists of the diffracted radiations emerging after passage through the birefringent material of the fibres. In the cases of rayon and of silk, this radiation is mostly or entirely concentrated in the diametral streak; the intensity of this is negligible when the fibre axis is parallel to the vibration direction of either the polariser or the analyser, but is a maximum when it bisects the angle between them. The net result due to an aggregate of fibres orientated in various directions is thus the phenomenon of the cross illustrated in Fig. 11. On the other hand, in the cases of cotton and wool, the complex patterns seen in the field contain much of the diffracted light, and this is not extinguished when viewed between crossed polaroids, whatever be the setting of the fibres with respect to the vibration-directions of the polariser and analyser. It is not surprising that in these circumstances, cotton and wool aggregates do not exhibit the phenomenon under consideration.

5. INTERPRETATION OF THE RESULTS

We do not propose in this preliminary report to discuss in any detail the features observed and recorded in the Plates accompanying the paper. Such a discussion could be better undertaken when the techniques have been further improved and the results are expressed in quantitative terms. We shall content ourselves here with some remarks of a general nature.

The appearance of features other than the central diametral streak in the diffraction pattern of a fibre is connected, as already remarked, with variations in the structure of the material along the length of the fibre; perfect uniformity of structure would result in the absence of such features, while *per contra* they would be very pronounced when the structure varies conspicuously from point to point along the fibre. This inference is supported in the cases under study by evidence from other sources. It is well known, for example, that silk is a crystalline fibre, its X-ray pattern exhibiting well-defined spots. The uniformity of structure thus indicated is also evident from its optical diffraction pattern which is of the simplest type. *Per contra*, in the case of cotton, the X-ray diffraction pattern shows very clearly that the cellulose groups comprising the fibre are variously inclined to the fibre axis; the characteristic X-ray reflections appear spread out as arcs instead of as sharp spots. The optical diffraction pattern reproduced in Fig. 1, Plate VII, also indicates the presence of diffracting units inclined to the length of the fibre. That the diffraction pattern does not vanish when

viewed between crossed polaroids but continues to be visible with certain significant changes indicates that these diffracting units are themselves birefringent. Thus, while supporting the indications furnished by the X-ray data, the optical diffraction pattern of the cotton fibre gives us a fuller insight into its structure.

6. SUMMARY

When a narrow pencil of light which comes to a focus in the plane of a photographic film traverses a single fibre set transversely to it in its path, the light diffracted by the fibre in various directions records itself on the film when adequate exposures are given. Photographs thus obtained with single fibres of cotton, wool, silk, rayon and chrysotile are reproduced in the paper. The birefringence of the fibres exhibits itself by the non-disappearance of the pattern when two polaroids crossed with respect to each other are placed one on either side of the fibre, the axis of the latter bisecting the angle between their vibration directions. The diffraction patterns indicate the inner structure of the fibres and are strikingly different for the different materials.

REFERENCES

Detailed accounts of the structure of fibres of cotton, silk, wool and rayon as determined by the known methods are to be found in several recent treatises, amongst others, those listed below.

- | | |
|------------------|---|
| E. Ott | .. <i>Cellulose</i> , Chap. III, Interscience Publishers, 1946. |
| P. H. Hermans | .. <i>Physics and Chemistry of Cellulose Fibres</i> , Second Part, Elsevier, 1949. |
| G. L. Clark | .. <i>Applied X-Rays</i> , Chap. XXIII, McGraw-Hill, 1940. |
| A. Frey-Wyssling | .. <i>Sub-Microscopic Morphology of Protoplasm and Its Derivatives</i> , pp. 193-203, Elsevier, 1948. |

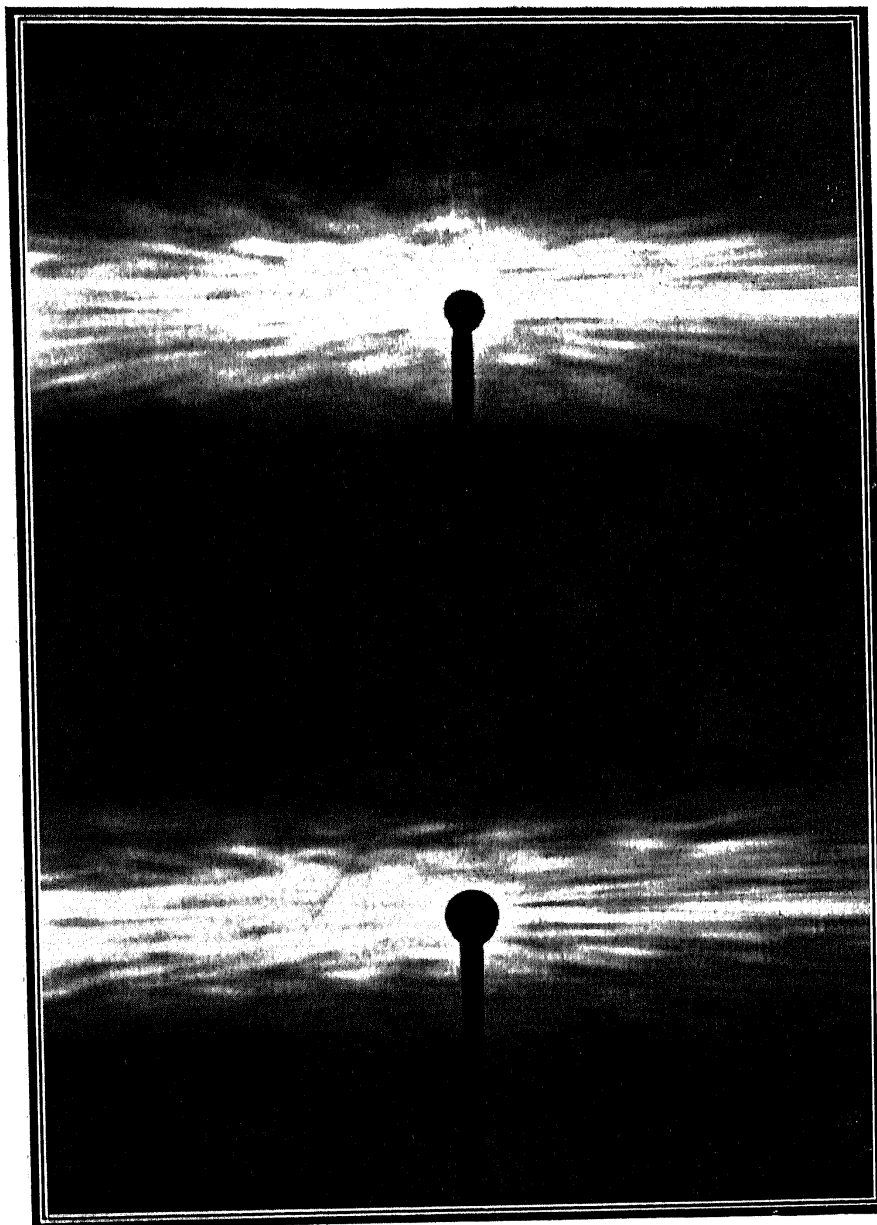


FIG. 1

FIG. 2

Diffraction Pattern of Cotton Fibre

FIG. 1. With unpolarised light; FIG. 2. Between crossed polaroids

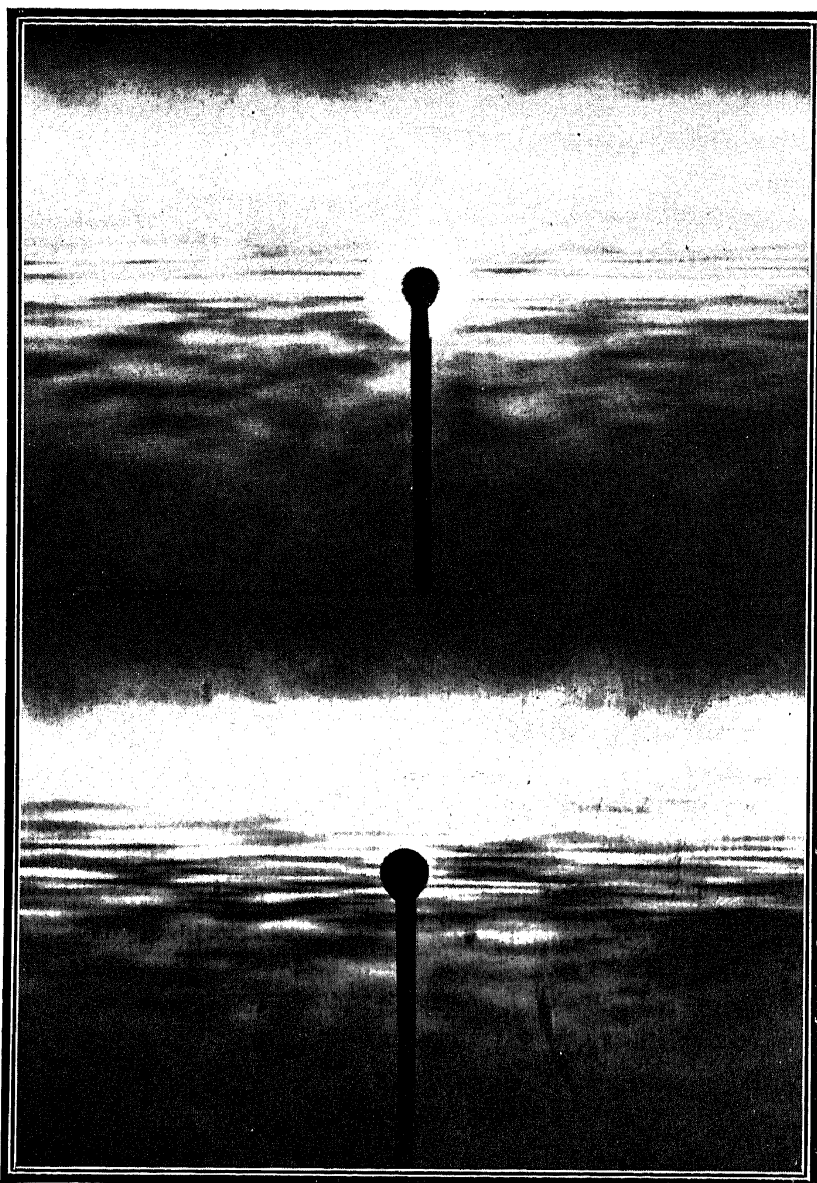


FIG. 3

FIG. 4

Diffraction Pattern of Wool Fibre

FIG. 3. With unpolarised light ; FIG. 4. Between crossed polaroids

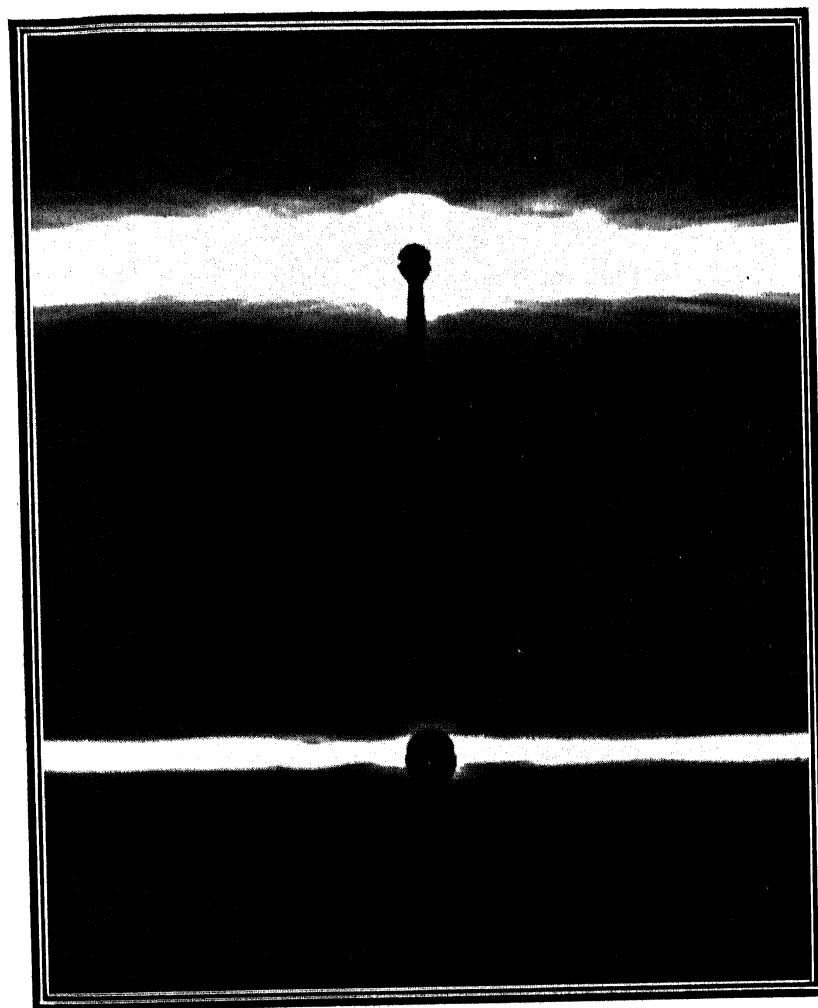


FIG. 5

FIG. 6

Diffraction Pattern of Chrysotile (Asbestos) Fibre

FIG. 5. With unpolarised light; FIG. 6. Between crossed polaroids

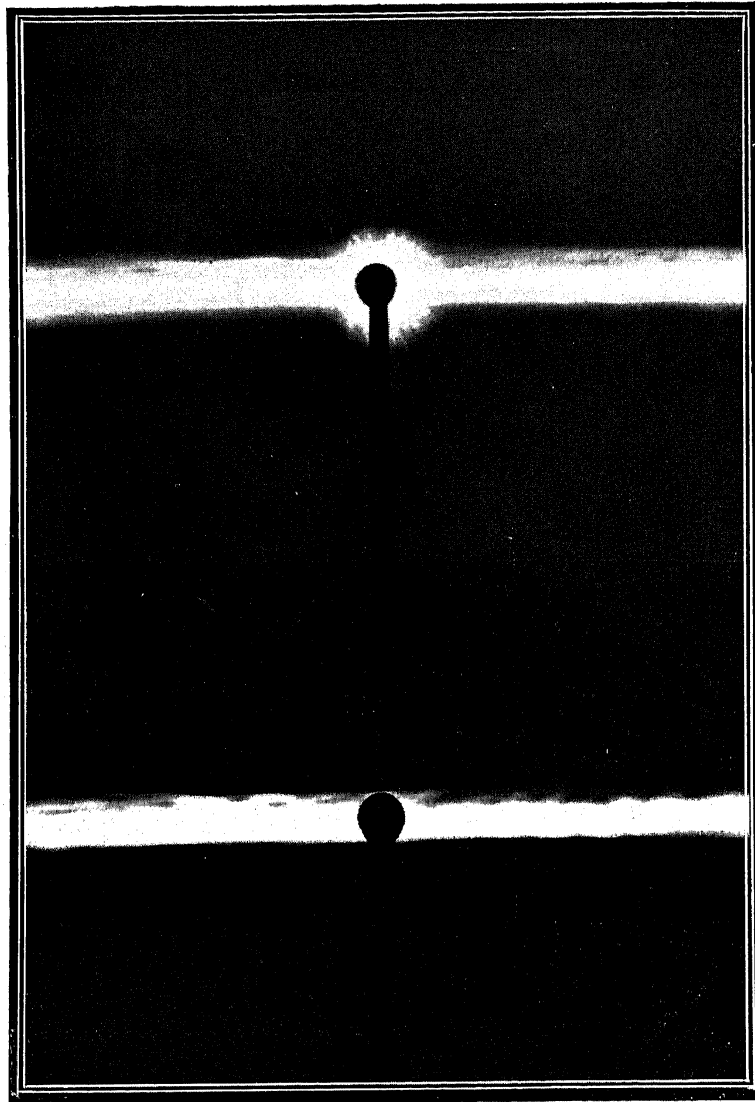


FIG. 7

FIG. 8

Diffraction Pattern of Rayon Fibre

FIG. 7. With unpolarised light; FIG. 8. Between crossed polaroids

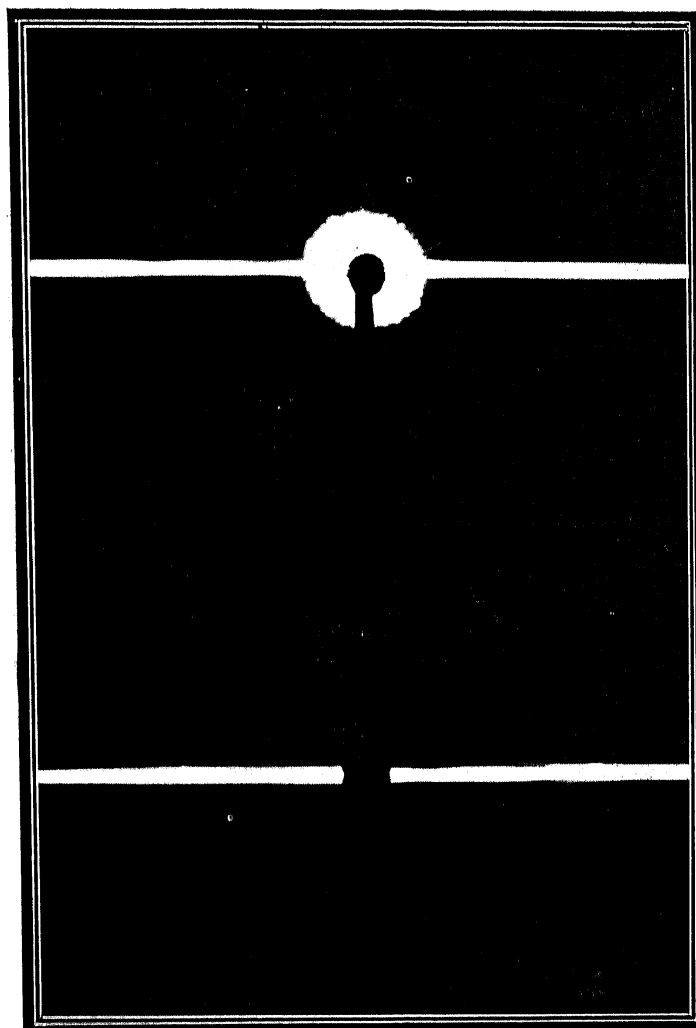


FIG. 9

FIG. 10

Diffraction Pattern of Cocoon Silk Fibre

FIG. 9. With unpolarised light ; FIG. 10. Between crossed polaroids

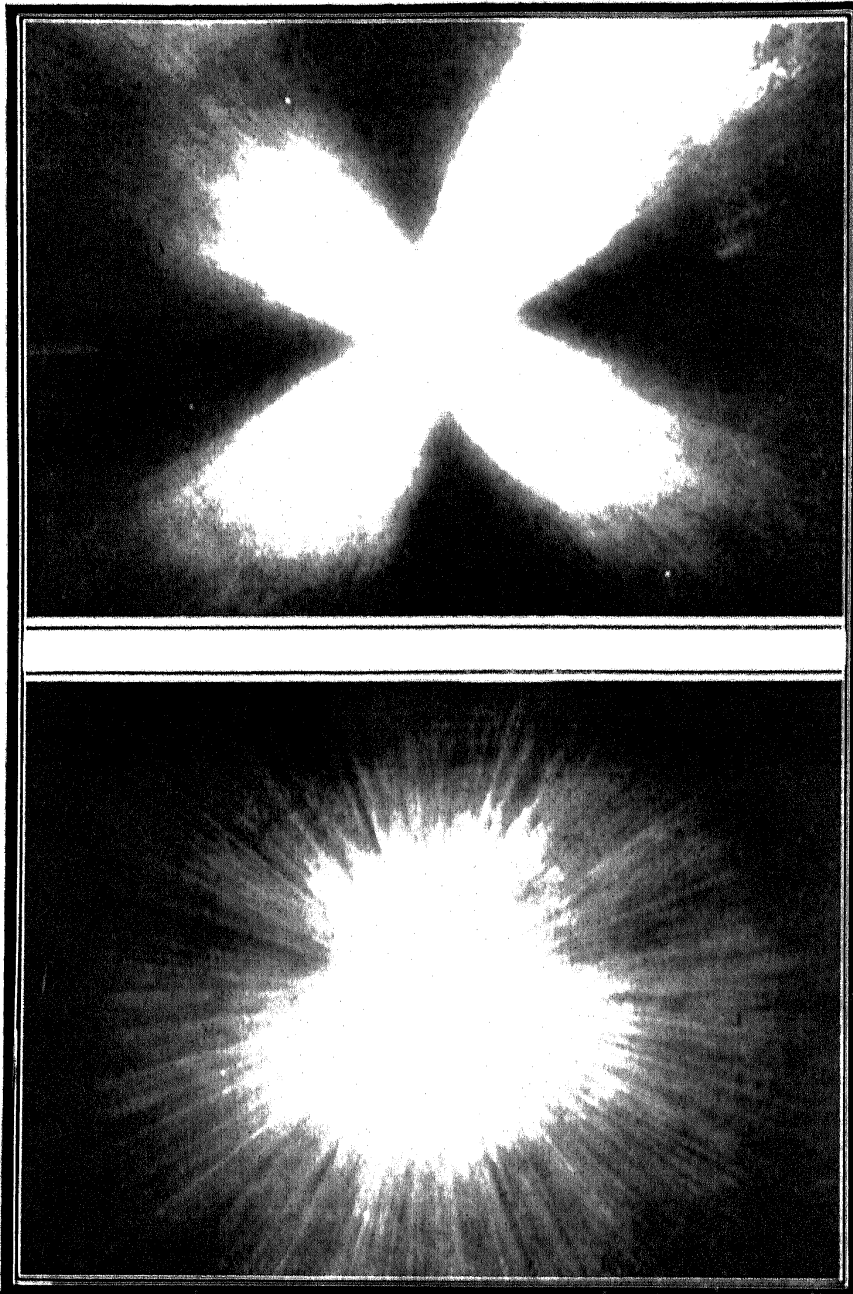


FIG. 11

FIG. 12

Diffraction Patterns of Fibre Aggregates

FIG. 11. Rayon between crossed polaroids; FIG. 12. Glass wool in unipolarised light