ON THE POLYCRYSTALLINE FORMS OF GYPSUM AND THEIR OPTICAL BEHAVIOUR

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1. Introduction

Gypsum occurs in nature both as well-developed crystals and as polycrystalline masses. The best known of the massive forms is alabaster; this is a fine-grained white solid. Another naturally occurring variety of gypsum is satin-spar, which is a fibrous material presenting a highly characteristic sheen or lustre from which its name is derived. The present paper is chiefly concerned with a form of gypsum which has come under our notice and which is distinct from either alabaster or satin-spar. It is noteworthy by reason of the very beautiful and interesting optical effects which it exhibits. We have been unable to find any reference to these phenomena in the mineralogical literature, and it would seem that though the material is clearly a distinct species of gypsum, this has not been hitherto recognised and its remarkable optical behaviour has remained unnoticed. We shall proceed to describe the phenomena and explain them in terms of the structure of the material. We have thought it desirable also to include in this paper some observations we have made with alabaster and with satin-spar, and their interpretation in terms of the physical constitution of these varieties of gypsum.

2. Nature of the Material

The species of gypsum which forms the principal subject of this paper presents some external features which enable it to be readily distinguished from the other recognised varieties of gypsum, *viz.*, selenite, satin-spar and alabaster. Like selenite, it may be readily split into slabs or sheets of any desired thickness, but the surfaces thus exposed do not exhibit the smoothness and optical perfection characteristic of the cleavages of selenite. Indeed, they are more appropriately described as planes of parting rather than as cleavages. Another distinctive feature of the material which at once distinguishes it from selenite is that the edges of the blocks or slabs are not smooth but exhibit parallel ridges, suggesting that the material is an aggregate of rod-shaped crystals having a common orientation perpendicular to the planes of parting. Optically, the material does not show the perfect transparency in all directions characteristic of selenite; neither does it, on the other hand, exhibit the sheen or lustre characteristic of satin-spar. The fact that it readily splits into parallel sheets indicates a certain measure of similarity of its structure to that of the tabular forms of selenite. We infer that the planes of parting are parallel to the crystallographic symmetry plane of gypsum, while the rod-shaped crystals are orientated along the crystallographic symmetry axis. We shall see later that these inferences are supported by the results of a more detailed investigation and we accordingly propose to designate this species of gypsum (which appears to be by no means uncommon) as "fascicular gypsum".

3. THE CIRCLES OF INTERNAL REFLECTION

When a plate of this species of gypsum is held in front of the eye and a distant source of light is viewed *normally* through it, the phenomena illustrated in Fig. 1, Plate XIII are observed. A brilliant circle of light is seen; at the centre of this circle, the source of light appears, but is overlaid by a diffraction pattern consisting of a central disc with concentric surrounding rings. As the plate is tilted away from the normal setting, the outer circle enlarges while the inner diffraction pattern expands and becomes a second circle. This is initially somewhat diffuse by reason of its being a circular diffraction ring with fainter concentric rings on either side (Fig. 2 in Plate XIII). The circle, however, sharpens when the plate is more obliquely set. With a further tilt of the plate, a third circle emerges at the centre of the whole pattern, firstly as a diffraction disc accompanied by rings and later as a well-defined circle of light (Fig. 3 in Plate XIII). As the plate is held more and more obliquely, the angular diameters of all the three circles continue to enlarge, but they remain concentric with each other, and also tend to come closer together (Fig. 4 in Plate XIII). The source itself continues to be visible as a bright point on the second ring in every case. The brightest parts of all the three circles are the regions in the immediate vicinity of this image of the source. However, in a dark room, the complete circles can be seen, and their angular diameters may be increased to any desired extent by giving an appropriate tilt to the plate. At the most oblique settings of the plate, the angular separations of the three circles from each other reach a minimum and then increase once again. Except when they emerge at the centre of the field and display the colours associated with diffraction, the bright circles appear white, thus unmistakably indicating their origin as due to *internal reflection.*

It should be mentioned that the photographs reproduced in Figs. 1 to 4 in Plate XIII, were obtained using a sheet about 1 mm. thick cleaved from a block of the material. Thin glass plates were fixed on either side with a little canada balsam to eliminate the effect of the optical imperfections of the exterior faces. Essentially the same phenomena are observed with either thicker or thinner sheets. With thick plates, however, the rings become a little broad and the whole field is overlaid by diffuse light; the light source also ceases to be visible on the second ring when the plate is tilted sufficiently. This is not the case with thinner plates; the source of light continues to be visible even at very oblique incidences. The diffraction effects of the kind referred to above are also more conspicuous with the thinner plates.

4. Polarisation of the Internal Reflections

We shall now proceed to describe and illustrate the remarkable features of polarisation exhibited by the circles of internal reflection. These features may be observed by placing a polaroid between the eye and the light source either before or after the plate, and rotating either the polaroid or the plate in its own plane. Strangely enough, the effect of the introduction of the polariser is quite different according as it is set before or after the plate of gypsum. The extent to which the plate is tilted away from the normal is not a matter of importance, since a particular circle of reflection exhibits much the same features of polarisation irrespective of its angular diameter. On the other hand, the orientation of the plate in its own plane with respect to the vibration direction of the polaroid is of the utmost importance. One finds by trial that there are two directions in the plane of the plate perpendicular to each other which we may designate as OX and OZ respectively, with one or the other of which the vibration direction of the polaroid should coincide to produce the maximum effects. If the vibration direction of the polaroid bisects the angle between OX and OZ, the polarisation effects are not observed.

Figures 5, 6, 7 and 8 in Plate XIV illustrate the changes in the relative intensities of the three circles of reflection resulting from the insertion of a polaroid. (The plate of gypsum was set obliquely, and the camera recorded only the more intense parts of the circles.) The legends entered below each figure indicate the location of the polaroid and its setting. P indicates that the light was polarised before entering the plate of gypsum, while A indicates that light was unpolarised at entry but passed through a polaroid after reflection. On a comparison of Figs. 5 and 6 it will be seen that a transfer of the polaroid from the front to the back of the plate results in reversing the behaviour of the first and the third circles of reflection, leaving the second or middle circle unaffected. A comparison of Fig. 7 with Fig. 8 shows a similar change in the opposite direction, while a comparison of Fig. 5 with Fig. 7 or of Fig. 6 with Fig. 8 exhibits the effect of setting of the vibration direction of the polaroid along OZ instead of along OX.

The significance of the facts stated above is clear, namely, that in the internal reflections which give rise to the first and third circles, the vibration direction changes from OX to OZ or *vice versa* in the act of reflection: the former change gives us the first circle of reflection, while the third circle of reflection is associated with a change of the vibration direction from OZ to OX. On the other hand, in the middle or second circle of reflection, there is no change of the vibration direction, OX remaining as OX and OZ as OZ.

If both a polariser and an analyser are used, their effects are combined, as will be seen from Fig. 9 to Fig. 12 reproduced in Plate XIV; the legends below each figure give the settings of both the polariser and analyser. It will be noticed that when they are crossed, the second or middle circle is greatly weakened, while the first appears with maximum intensity and the third circle is totally extinguished or *vice versa*. On the other hand, when the polariser and the analyser are set parallel, both the first and the third circle appear greatly weakened, while the second or middle circle appears with maximum intensity.

It may be remarked that the photographs reproduced as Figs. 5 to 12 in Plate XIV, were recorded with a specimen which exhibited the polarisation effects most conspicuously. Other specimens indeed show similar effects but not always in such a striking manner The degree of perfection of polarisation was also found to vary over the area of the specimens.

5. ORIGIN OF THE REFLECTIONS

It is evident that the phenomena described above are all interconnected and that an explanation of the same has to be sought for on the basis of the geometric and optical characters of the polycrystalline aggregate. Earlier in the paper, we have suggested that the material consists of rod-shaped crystals having a common orientation for their crystallographic symmetry axis. The latter is an axis of the optic ellipsoid of gypsum. Had the two other optic vibration directions also been the same for all the rods, the material would be optically homogeneous and indistinguishable from selenite. Hence we are compelled to assume that the two other optic vibration directions in the plane perpendicular to the rods are not identical for all of them.

Holding a plate of the material between crossed polaroids and viewing an extended source of light through it, we find on rotating the plate in its own plane, two mutually perpendicular settings in which there is a maximum restoration of light and two others intermediate between them in which there is nearly but not quite complete extinction of light. A *small* bright source of light viewed through the combination at the latter settings is, however, extinguished more or less perfectly.

The situation indicated above may be summed up by the statement that the plate considered as a whole has two mutually perpendicular directions as its optic vibration directions, though the individual rods in the aggregate may deviate therefrom to a greater or less extent. That this is the actual situation is indicated by the appearance of the plate under a microscope when viewed between crossed polaroids. Rotating the plate in its own plane, two settings are found in which there is a minimum of light in the field but by no means complete extinction everywhere. The greater part of the plate appears dark, but there are areas in which there is a notable restoration of light. At these two settings, the directions in the plate designated as OX and OZ in the description of the polarisation effects are also the vibration directions of the polariser and the analyser. It is noticed further that the regions on the plate which give the most marked extinctions are those which give the most perfect polarisation of the circles of reflection, while the areas in which the extinctions are imperfect, also show marked imperfection of the polarisation of the circles of reflection.

6. The Geometric Character of the Reflections

It is a familiar result in geometrical optics that when a pencil of light is incident on a cylindrical rod in a direction making an angle θ with its generators, the rays reflected by its surface lie along the generators of a right circular cone whose semi-vertical angle is also θ and which has its axis parallel to the length of the cylinder. In the present case, however, the reflections occur at intercrystalline boundaries within a birefringent solid, and hence the angles of incidence and of reflection of the light at these boundaries are not necessarily equal. Actually, as a result of the birefringence of the material, an incident pencil of light would divide on entry into the plate into two pencils polarised differently which travel in different directions with different velocities. A further subdivision would occur in the act of reflection at the boundaries, giving us four reflected pencils in all. However, owing to the special circumstance that the external surfaces of the plate are normal to an axis of its optic ellipsoid, two of these pencils would obey the ordinary laws of reflection and would emerge from the plate in identical directions, though their planes of polarisation are mutually perpendicular. For the two other pencils, the angle of reflection would be greater or less than the angle of incidence as a consequence of the planes of polarisation and therefore also the wave-velocities being different for the incident and reflected pencils. Thus, in the final result, we would have, instead of a single cone of reflected rays, three such cones exhibiting the polarisation effects already described; the central or the middle one would have the direction of the light source as one of its generators, and hence the source would appear as a luminous point on that circle, The correctness of the explanations set forth above is confirmed by a few simple calculations and observations. The angles of incidence and of reflection of light inside a birefringent medium are connected by the relation $\mu_i \cos \theta_i = \mu_r \cos \theta_r$, where θ_i and θ_r are the glancing angles which incident and reflected rays make with the reflecting boundary, and μ_i and μ_r are the corresponding refractive indices. In our present problem, the two refractive indices under consideration are 1·521 and 1·530. For normal incidence on the surface of the plate, $\theta_i = 0$, while the values of θ_r comes out respectively as 6° 18′, 0°, and imaginary for the three circles of reflection. Accordingly, the angles of emergence for the first and second circles come out as 9° 39′ and 0° respectively while the third circle is non-existent, in close agreement with the observations. For the case in which the third circle is just visible, the angular radii of the first, second and third circles on emergence come out as 13° 42′, 9° 39′ and 0° respectively, once again in close agreement with the results of observation.

The reflections at the intercrystalline boundaries arise from the fact that the optical polarisabilities of the material on either side of such a boundary differ in magnitude or direction or both. The birefringence of gypsum being very weak, such differences would necessarily be small. The strong reflections actually observed are a consequence of the incidence of the light being very oblique in the cases under study. This is clear from the fact that the circles of reflection display their maximum intensity in the regions not too remote from the image of the light source appearing located on the second circle (see Figs. 2, 3 and 4, Plate XIII). The variation of intensity exhibited by the first circle of reflection over its circumference (Fig. 1 in the same plate) has a different origin : it exhibits the fact that the reflecting power is a function of the azimuth of incidence of the light on the boundary, besides being dependent on the angle of incidence which in this case has the maximum value of 90°.

A few brief remarks must suffice in explanation of other features which are observed and which have been referred to earlier. The diffraction phenomena associated with the circles of internal reflection are evidently a consequence of the limitation of the effective aperture due to the oblique incidence of light on the reflecting surfaces. They naturally become most conspicuous when such incidence is nearly grazing. Phenomena closely resembling them may be observed when light from a distant source is reflected obliquely at the surface of a short cylinder of glass.

The reflections of light at the intercrystalline boundaries in the material would necessarily result in a progressive extinction of the incident light beam in traversing the material. The greater the thickness of the plate and the more it is tilted away from the normal, the larger would be the number of reflecting surfaces traversed by the incident light, and hence also the more rapid would be its extinction, as is actually observed.

The diffuse radiation which appears overlying the circles of reflection, especially with thick plates, and the broadening of the circles of reflection which is noticed with such plates when they are held obliquely may be the result of a lack of perfect parallelism in the orientation of the rod-like crystals within the material. Another factor which would also result in a diffusion of light is the *refraction* of the light beams in their passage through the successive intercrystalline boundaries. That such refraction does play an important role is suggested by the partial polarisation of the second circle of reflection which is noticed when the plate is tilted considerably away from the normal.

7. Optical Behaviour of Transverse Sections

In the foregoing, it has been assumed that the rod-like crystals of which the material is composed are parallel to each other and to the crystallographic symmetry axis. If this were strictly the case, a plate of the material so cut that its faces are parallel to the rods should exhibit the following optical behaviour: (1) a light beam polarised with its electric vector parallel to the rods should be freely transmitted by the plate without diffusion or extinction; (2) on the other hand, if the electric vector is perpendicular to the rods, the optical heterogeneity of the material should exhibit itself and result in a strong diffusion and extinction of the light in its passage through the plate, the diffused radiation appearing as a fan of rays in a plane transverse to the rods.

These indications of theory can be tested by observation and are found to be in general accord with the facts but not completely so. A distant source of light viewed through such a plate held normally continues to be visible in focus. Further, it is also completely polarised with its electric vector parallel to the length of rods. However, one also observes a fan of diffracted rays accompanying the transmitted light spread out in a plane transverse to the rods, and this exhibits a partial polarisation in the same sense as the transmitted light. A tilt of the plate away from its normal setting results in extinguishing the transmitted light, if such tilt is in a plane parallel to the rods. On the other hand, a tilt in the perpendicular plane has no such effect.

From the foregoing observations one infers that the structure of the material is approximately but not quite accurately that assumed. This also becomes evident when a plate cut in the same fashion is examined between crossed polaroids under a microscope. A complete extinction is not obtained in any setting of the plate. There is, however, a notable diminution of intensity of the light seen in the field, when the direction of the rods coincides with the vibration direction of either the polariser or the analyser.

8. SOME OBSERVATIONS WITH ALABASTER

Alabaster is opaque to light, evidently as a consequence of the birefringence of gypsum, though light can penetrate by diffusion through very considerable thicknesses. The extent of such penetration is highly variable and such variations appear to be connected with the structure of the material including especially the size of the crystallites of which it is composed.

It appeared to be of interest to investigate whether a transmission of light in the true optical sense could be observed with alabaster if it be thinned down sufficiently. With coarse-grained varieties the test does not succeed, since we soon arrive at a stage at which the thickness of the test plate is comparable with the size of the individual crystallites which can then be observed to transmit light independently of each other. Fine-grained varieties of alabaster, however, can be thinned down sufficiently to enable a true optical image of a distant and brilliant source of light to be seen through the plate. The image appears overlaid by a diffuse halo, the structure and angular extension of which are determined by the thickness of the plate and the grain size of the substance. The optical transmission is readily distinguished from the overlying halo by its brilliance and its perfect sharpness. It is completely extinguished when the plate is held between crossed polaroids, whereas the halo continues to be visible in the same circumstances though with diminished intensity.

9. THE STRUCTURE AND OPTICAL BEHAVIOUR OF SATIN-SPAR

The fibrous nature of satin-spar is conspicuously evident from its very appearance. A beam of light is spread out by reflection at the surface into a fan of light in a plane transverse to the length of the fibres. A plate of the material having its faces parallel to the fibres differs very strikingly in its behaviour from that described in Section 7 above. A distant source of light is invisible when viewed through it, all that is observed being a fan of diffracted rays transverse to the fibres and this is found to be completely unpolarised. This behaviour is readily explicable as a consequence of the birefringence of gypsum and the known crystallographic orientation of the fibres.

As is to be expected, light is found to penetrate along the fibres rather more freely than in the transverse direction. A distant bright source of light can indeed be seen if viewed through a rod of the material exactly along

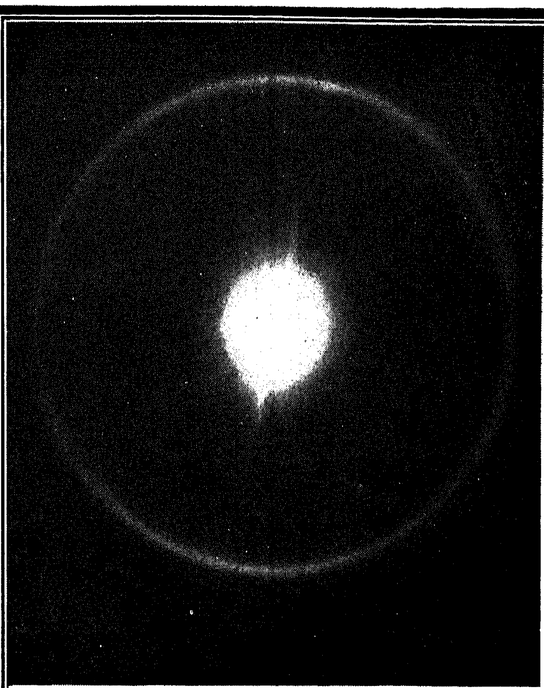


FIG. 1

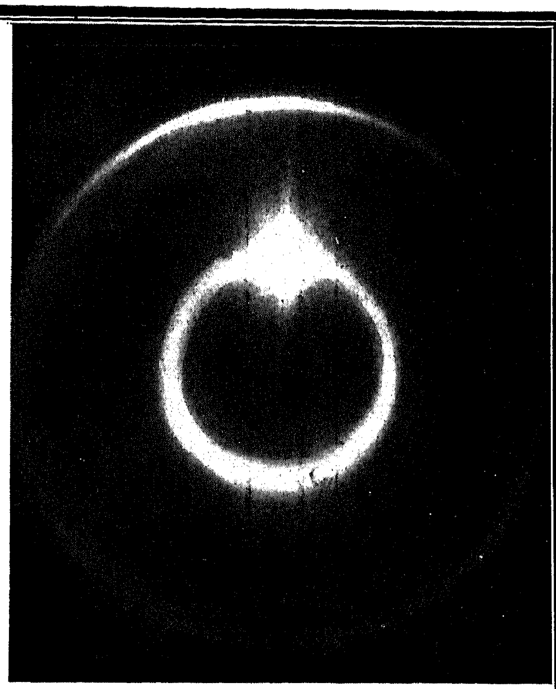


FIG. 2

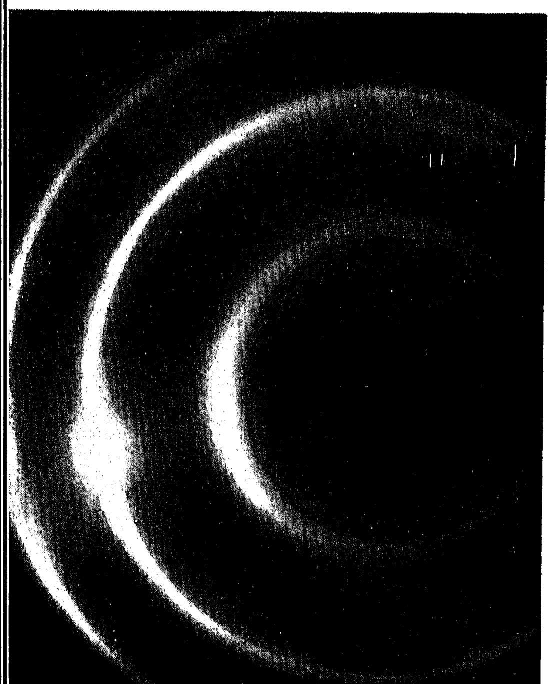


FIG. 3

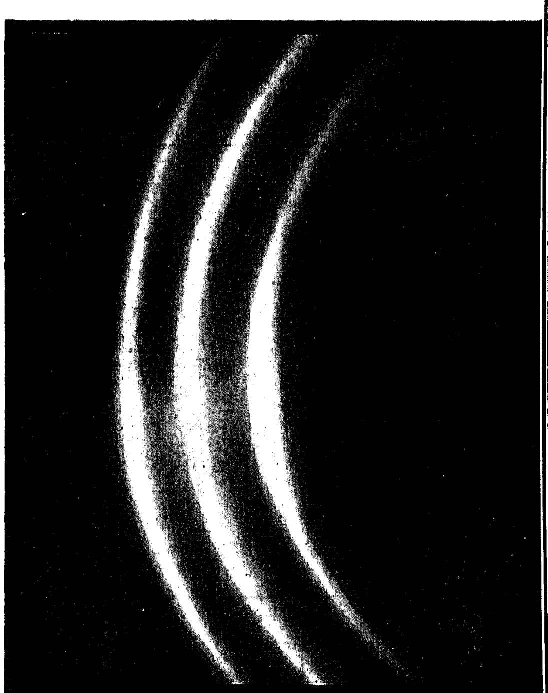
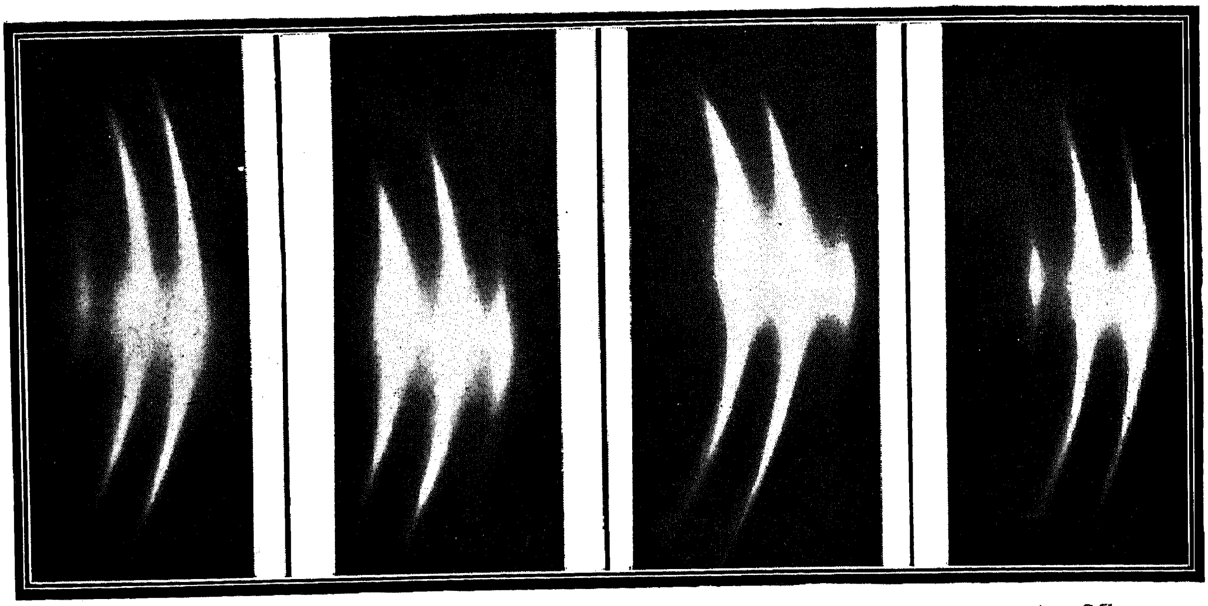


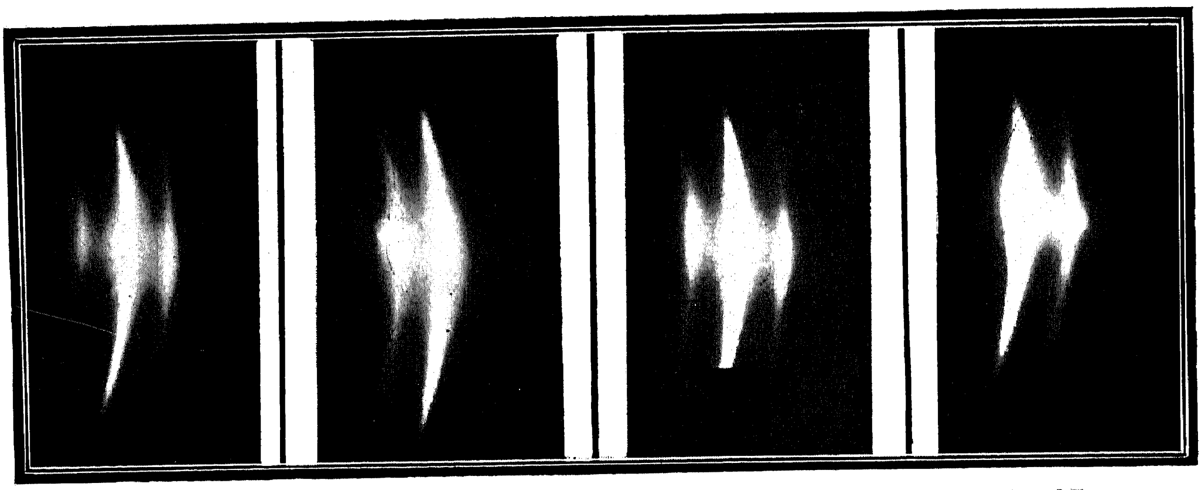
FIG. 4

FIG. 5 FIG. 6 FIG. 7 FIG. 8



P—OX A—OX P—OZ A—OZ

FIG. 9 FIG. 10 FIG. 11 FIG. 12



P—OX A—OX | P—OX A—OZ | P—OZ A—OZ | P—OZ A—OX its fibres, but appears spread out into a diffuse patch of light. If the rod be tilted a little, this transforms into a diffuse circle of light with a darker area within, the diameter of which increases with the tilt. The brilliancy and sharpness of the phenomena observed with fascicular gypsum are conspicuously absent in the present case, neither are any polarisation effects observed.

10. Summary

The paper brings to notice the remarkable optical effects exhibited by a polycrystalline form of gypsum which is different from both alabaster and satin-spar in its structure. It is not a fibrous material but consists of fine rods orientated nearly parallel to the *b*-axis of gypsum and exhibits a ready cleavage along planes perpendicular to that axis. A source of light viewed through a plate of the material exhibits, in general, three concentric circles which are polarised in a characteristic fashion. The source itself appears as a luminous point on the second or middle circle. It is shown that these circles arise by reason of the reflection of light at the boundaries between the rod-like crystals composing the material, for which the name "fascicular gypsum" is accordingly proposed. A theoretical explanation of the phenomena is given and photographs of the same are reproduced. Observations on the optical behaviour of alabaster and of satin-spar are also reported.