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The structure and optical behaviour of iridescent calcite

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1. Introduction

Christiaan Huygens[1] noticed that one of his specimens of iceland spar contained an inner layer exhibiting iridescence and remarked that this was parallel to one edge of the rhombohedral cleavage and equally inclined to the two others. It is now recognised that such layers arise from internal twinning and that they may be artificially produced by subjecting a crystal of calcite to suitable stresses. It is also well-known that when a distant luminous object is viewed through a rhomb of iceland spar in which such twinning is present, three images of the object are seen instead of only one; the two outer images often exhibit vivid colour, while the central or undeviated image is usually colourless. The brightness of the images, their angular separation, and the colours which they exhibit vary greatly with the direction in the crystal along which the source of light is viewed. An explanation in general terms of the origin of the two outer images and of the colour and polarisation which they display appears in Mascart's treatise.[2] Grailich[3] and later Osthoff[4] investigated the reflection and refraction of light by the twinning planes in calcite. A summary of their results is given by Pockels.[5] A noteworthy observation regarding the optical behaviour of twinned calcite is also to be found recorded in a paper by Rayleigh[6].

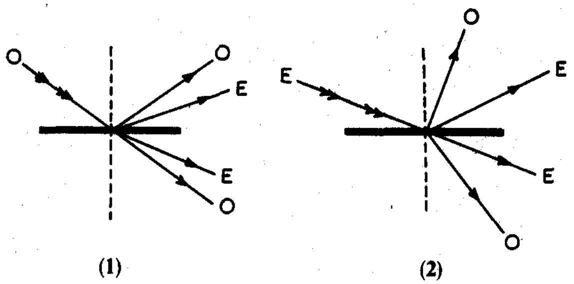
Numerous specimens of transparent calcite exhibiting internal twinning were available to us in the Museum of this Institute and this induced us to undertake the study of their optical behaviour. It was immediately noticed that specimens in which twinning layers traverse the crystal simultaneously in two or even three different directions exhibit gorgeous arrays of multi-coloured images arranged in geometric patterns when a source of light is viewed through them. Plates I and II accompanying this paper reproduce photographs obtained by us of a few of the many patterns observed with the material at our disposal. A detailed examination of the case of calcite containing only a single twinning layer also revealed many facts which had apparently been overlooked by the earlier investigators. We shall refer to these in the course of the paper, the purpose of which is to present the complete picture of the optical behaviour of iridescent calcite which has emerged from our studies.

2. Geometric theory

When a train of light-waves traverses a layer of calcite differing from the material on either side of it in crystallographic orientation, the waves would necessarily be reflected and refracted by the layer. Elementary considerations indicate that the angle of incidence θ_i and the angle of reflection or refraction θ_r would be related to the corresponding refractive indices μ_i and μ_r by the general formula

$$\mu_i \sin\theta_i = \mu_r \sin\theta_r. \tag{1}$$

Since the medium is birefringent, it follows that for a given angle of incidence, a wave-train of either the ordinary or the extraordinary species would, in general, give rise to *two* reflected and *two* refracted wave-trains. Other inferences from the formula are: (1) One of the refracted wave-trains emerging from the layer in each case would travel in the same direction as the incident wave-train; (2) the angles of reflection and of refraction of the ordinary wave-train would necessarily be identical; and (3) the angles of reflection as also of refraction would, in general, be greater for the extraordinary than for the ordinary wave-trains (*see* figure 1 and figure 2 below in which these results are embodied).



(1) (2)

Figures 1 and 2. Reflection and refraction by a twinning layer.

When a distant source of light is viewed through the opposing pair of faces of a cleavage rhomb of iceland spar, we observe only a single image of the source, though the wave-trains divide into the ordinary and extraordinary species and follow different paths within the crystal. It is readily seen from figures 1 and 2 that the same situation would subsist even when a twinning layer is present and is traversed by these waves. In such a case, however, the ordinary wave-train which emerges as an extraordinary one and the extraordinary wave-train which emerges as an ordinary one after passage through the layer, would both give rise to additional images, deviated respectively to one side and the other of the principal image. The magnitude of their angular deviations may be deduced at once from the general formula and is the smaller, the more nearly the direction of observation through the crystal approaches the optic axis. It is important to remark that while the central or undeviated image would appear in the same direction for all wavelengths, such would not be the case for the deviated images. The dispersion of the refractive indices in calcite is very different for the ordinary and the extraordinary waves. Hence, the two lateral images would appear drawn out into spectra, and it may be remarked that such dispersion would be large when the wave-train under consideration emerges nearly parallel to the twinning layer.

The general formula indicates other results of interest. If μ_i be the ordinary and μ_r the extraordinary index, θ_r would be real only if $\theta_i \leqslant \sin^{-1}\mu_r/\mu_i$. At the upper limit indicated, the reflected and refracted extraordinary waves would emerge parallel to the twinning layer, while for greater incidences they are non-existent. Likewise, when μ_i is the extraordinary and μ_r the ordinary index, $\theta_r \leqslant \sin^{-1}\mu_i/\mu_r$: the limiting value is reached when the incident waves graze the twinning layer. In both cases, the limiting angle may be written as $\sin^{-1}\mu_e/\mu_0$ where μ_0 is the ordinary index and μ_e is the extraordinary index for the direction of propagation parallel to the twinning layer in the plane of incidence. Thus, if light be incident on a twinning layer simultaneously in all directions, a boundary line may be drawn in the field separating the directions of incidence of the ordinary wave-trains which give rise to extraordinary reflections and refractions and of those which do not. Likewise, the optical fields traversed by the reflected and refracted wave-trains are, each of them, divided by a boundary line on the two sides of which ordinary wave-trains resulting from incident extraordinary ones respectively appear and do not appear.

It is evident from the foregoing remarks that we cannot always expect to be able to observe the deviated images on both sides of the principal image when a distant light source is viewed through a calcite rhomb. As the angle of incidence on the twinning layer approaches and ultimately exceeds the limiting value, the extraordinary refraction resulting therefrom would emerge parallel to the twinning layer and ultimately disappear.

3. Intensity considerations

A factor which greatly influences the observed phenomena is the azimuth of the plane of incidence of the light on the twinning layer. It has already been remarked that the layer is parallel to one of the edges of the rhombohedral cleavage and equally inclined to the two others. It follows that a plane through the edge first mentioned which is perpendicular to the lamina would contain the optic axis of the calcite crystal and the optic axis of the twinning layer as well. The plane under reference would thus be a plane of symmetry for the entire crystal including the twinning layer. As in the case of potassium chlorate recently discussed in these

Proceedings,[7] it can be shown from elementary considerations that when the plane of incidence of the light coincides with this symmetry plane, reflections and refractions would disappear and the incident light-waves would emerge from the twinning layer with their direction of propagation as well as their intensity unaltered.

The foregoing conclusion is readily tested by viewing a distant source of light through the calcite rhomb held in front of the eye of the observer in such manner that the twinning layer is vertical while the plane of observation is horizontal; the source should be viewed through the faces of the rhomb which are bounded by edges of which none is parallel to the twinning layer. Only the undeviated image of the source can then be seen. But if the crystal is slightly tilted about a horizontal axis so that the plane of incidence of the light ceases to be horizontal, the deviated images make their appearance, and rapidly gain in intensity as the plane of incidence is turned away from the horizontal in either sense.

An interesting modification of this experiment is to view an extended area of illumination through the faces of the rhomb held in the manner indicated above and to rotate the rhomb about a vertical axis so that the plane of the twinning layer remains vertical but alters its orientation with respect to the direction of observation. A division of the field of observation into two parts differing in their intensity of illumination then comes into view. A dark and sharply defined boundary is seen to separate the two parts of the field except where it is cut by the horizontal symmetry plane. This dividing boundary becomes more and more conspicuously observable as we move away from the symmetry plane in either direction. Interposing a polaroid in front of the rhomb, it is found that the division of the field into two parts and the dark boundary separating them are most conspicuous when the vibration direction of the polaroid is vertical, but disappear when the same is horizontal. It is thereby made evident that the boundary is the one indicated by the geometric theory as the limit of the angle of incidence of the ordinary waves on the twinning layer beyond which the extraordinary reflections and refractions disappear. That the boundary vanishes in the symmetry plane and becomes more conspicuous as we move away from it is to be expected in the circumstances. The boundary is sharply defined evidently as a consequence of the weakening of the ordinary transmission by reason of the extraordinary reflections and refractions reaching their maximum intensity just prior to their disappearance at the limiting incidence.

A slight modification of the arrangements described above enables us to observe the boundaries at which the reflections and refractions of the extraordinary waves as ordinary waves terminate. For this purpose, the illuminated screen is held to one side so that the light reflected or refracted by the twinning layer comes into the field of view. The boundary is seen then as an arc of light bordered by coloured fringes and separating a bright from a dark part of the field of view. The bright arc is however interrupted at the symmetry plane, and gains in intensity as we move away from that plane in either direction. By inserting a polaroid before and after the calcite rhomb respectively, it is readily demonstrated that the luminous arc has its origin in the trains of extraordinary waves which are transformed to ordinary ones by reflection or by refraction. The sharpness and brilliancy of the boundary are to be expected alike on geometrical and physical considerations.

4. Interference phenomena

As has been already remarked, the deviated images of a distant light source seen through the twinning layer are spread out into spectra by reason of the difference in the dispersive powers of calcite for the ordinary and extraordinary waves. Using a narrow slit as the source of light, these spectra can be readily observed and are found to be channelled by interference bands crossing them. The position and number of the interferences vary with the specimen under observation and thus evidently depend on the thickness of the twinning layer in each particular case. Careful examination reveals that we have two sets of interferences simultaneously present in the spectra, one of which is on a finer scale than the other. The explanation of these effects is not far to seek. Returning to the text-figures 1 and 2, it will be evident that in the passage of the incident waves through the twinning layer, the birefringence of the latter would come into play. Hence, each of the wave-trains reflected or refracted by the layer would itself be the resultant of the wave-trains of the two different species possible within the layer, the optical paths of which would be different. Interferences of the same nature as those exhibited by crystalline plates in polarised light would therefore arise and would manifest themselves in the spectral character of the reflected and refracted wave-trains. Effects of the same nature as the ordinary interferences of thin plates would also simultaneously appear. The colours observed with extended light sources are a consequence of both species of interference.

Interference phenomena are also strikingly manifested at and near the boundaries of transmission, reflection and refraction considered in the preceding section. They take the form of alternate dark and bright bands of varying sharpness and intensity fringing the boundaries referred to. There again, we observe two species of interference, one of which is on a finer scale than the other. These boundary effects are best studied using a diffusing screen illuminated by a monochromatic source, though indeed they can be also observed with white light.

5. Effects observed with multiply twinned calcite

We now turn to a consideration of the phenomena, typical examples of which are reproduced as figures 1, 2, 3 and 4 in plates I and II accompanying the paper. The photographs reproduced were obtained using a small aperture backed by a tungsten filament lamp as the source of light. A photographic camera placed about three metres away was focussed on the source, the calcite rhomb being placed immediately in front of the lens. Panchromatic plates were employed for recording the patterns. The brilliant colour effects actually observed are, of course, not reproduced.

Figure 1 shows the three images observed with a calcite rhomb having a single twinning layer. In addition to the bright images, we observe a system of bright streaks, as also some diffuse light surrounding the images. The surfaces of the cleavage rhombs had not been specially polished and their unevenness was no doubt in part responsible for some of the unwanted effects. Nevertheless, our studies with numerous specimens left little doubt that the long bright streaks appearing in the photographs and also seen visually had their origin within the calcite rhombs. It appears reasonable to attribute them to a lack of perfect smoothness of the twinning layers within the crystal.

Another remark may usefully be made with regard to figure 1 in the plate. This was recorded through the faces of the calcite rhomb which had two of their edges parallel to the twinning layer. In these circumstances, the plane of incidence of the light on the twinning layer was, in general, far removed from the symmetry plane. By turning the crystal about one edge, the images could be made to approach towards or recede from each other. Simultaneously, there were large changes in the colour and intensity of the images. At particular settings which were nearly but not quite coincident, the two outer images vanished but on further rotation they reappeared in positions closer still to the principal image. The observations indicated that for the settings at which the images disappeared, the plane of incidence was nearly coincident with the symmetry plane.

Figure 2 in plate I shows the pattern of a type frequently observed by us and which arises from the simultaneous action of twinning layers parallel to two of the edges of the rhombohedral cleavage. A noteworthy feature in the photograph is that the middle spot in the outer rows has split into two components. These were found to be polarised in perpendicular directions. Figures 3 and 4 represent more complicated cases. It is to be noted that the spots appear in groups of threes, with the middle one in each row distinctly separated into two components. All the patterns exhibited special features of polarisation analogous to those observed in the simple case of a single twinning layer.

6. Summary

Arrays of multi-coloured images of a distant light-source arranged in geometric patterns are exhibited by calcite rhombs traversed by twinning layers in several directions simultaneously. The paper reproduces four photographs of such patterns and also discusses the theory of the reflection and refraction of light by twinning layers in calcite. The large difference between the ordinary and

extraordinary dispersive powers results in a linear source observed through such a layer appearing drawn out into spectra channelled by interferences. Interference bands also fringe the sharply defined boundaries seen separating the different parts of the optical field of reflection and refraction.

7. References

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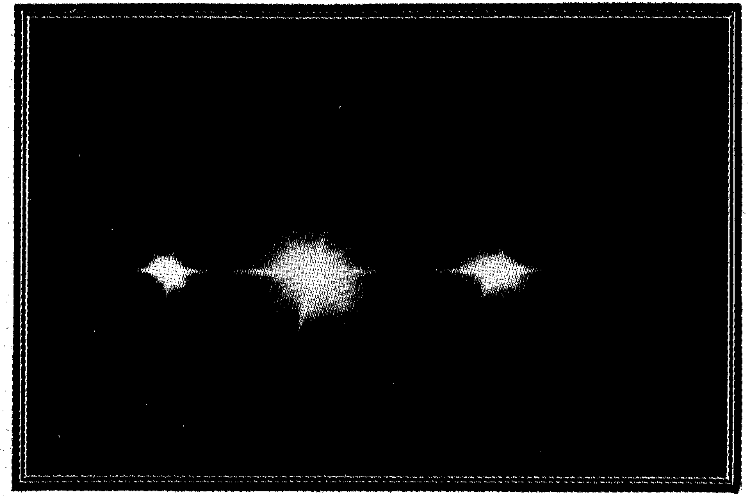

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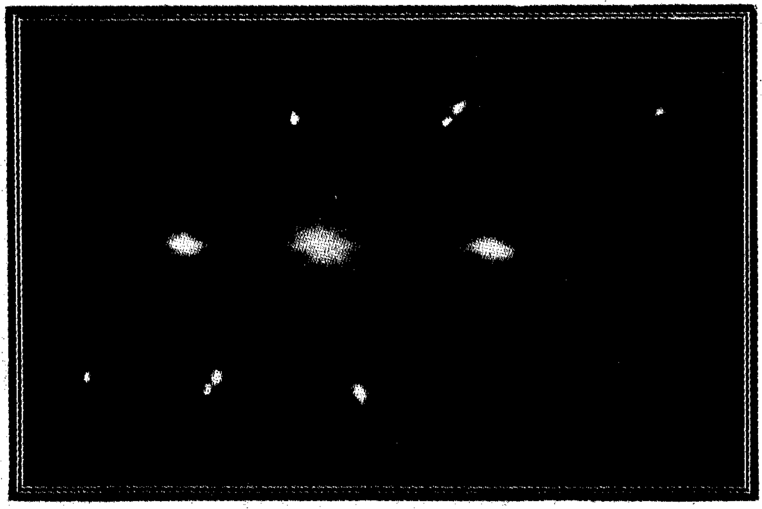
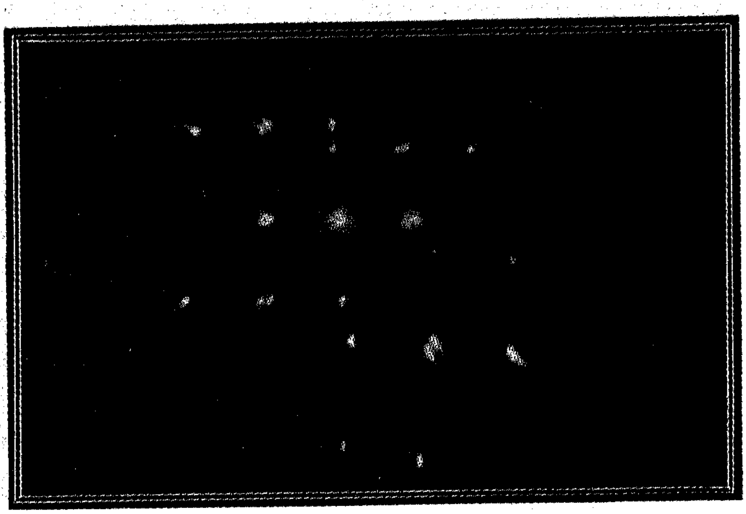


(2)

Figures 1 and 2

Plate I

(3)



(4)

Figures 3 and 4

Plate II