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On the iridescence of potassium chlorate crystals—Part II. Polarisation effects

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1. Introduction

A remarkable fact about the iridescence of potassium chlorate is that examples are frequently forthcoming in which the reflections are monochromatic, indicating a high degree of regularity of the stratifications as well as the presence of a great number of them. As we have seen in the preceding paper, an examination of the reflection spectra of such crystals reveals features that are in accord with the general theory of the optical behaviour of a regularly stratified isotropic medium. Nevertheless, it is not to be supposed that such a theory would suffice to explain all that is actually observed in the present case. Indeed the iridescence is itself a consequence of the birefringence of the medium coupled with the fact that the alternate layers of it are differently orientated. It follows that a proper theory has necessarily to be based upon a consideration of the propagation of light through a birefringent medium and of the specific characters of a reflection at a twin plane boundary.

2. Crystal form and birefringence

Potassium chlorate crystallises in the holohedral class of the monoclinic system; in other words the crystal has both a two-fold rotation axis and a plane of symmetry perpendicular to it. An examination of the tablet-shaped iridescent crystals in convergent polarised light readily enables one to convince oneself that the two-fold rotation axis and the plane of symmetry are oriented alike for all the components of the twinned crystal and that the former is parallel to the external faces of the tablet and that the latter is perpendicular to it. When the tablet is viewed normally between crossed polaroids and rotated in its own plane, two positions of extinction are obtained; in both these positions, when an extended source of light is viewed through the combination, a dark straight isogyre is seen crossing the field from end to end. It follows that the plane of observation marked out by this isogyre contains two of the principal optical vibration directions, while the third is perpendicular to it. In other words, the symmetry plane containing two of the principal vibration directions is normal to the face of the tablet, while the other principal vibration direction coinciding with the two-fold rotation axis lies in that face. Observations made with rhombus-shaped tablets show that the two-fold axis bisects the acute angle of the profile, while the symmetry plane bisects the obtuse angle in it.

The detailed discussion of the theory of twin-plane reflection given by Rayleigh in his pioneer investigation on the subject is based on the assumption that the crystal is feebly birefringent. This however is far from being actually the case for potassium chlorate. The three principal refractive indices of the crystal for sodium yellow light, are given in the literature as 1·5241, 1·5174 and 1·4099, and the crystal is thus strongly birefringent. On the other hand, one would not be far wrong in describing the substance as a close approximation to a uniaxial crystal in its optical behaviour. The angle between the two directions of single wave-velocity is given in the literature as 27° for sodium yellow light.

3. The vanishing of the reflection in the symmetry plane

Very simple considerations indicate that the reflection at the twin-plane boundaries should vanish—as is actually the case—when the plane of incidence coincides with the plane of symmetry.

The location of the principal optic directions in the symmetry plane is shown in figure 1, OX and OY referring to the upper side and OX′ and OY′ to the lower side of a twinning plane. The third vibration direction OZ is perpendicular to the plane of the paper. Since the upper and lower halves of the diagram are mirror images of each other, it follows that the coefficient of reflection at the boundary for any given angle of incidence would be the same whether the incidence is from above or from below. On the other hand, it follows as a consequence of the principal of reversibility that the reflection coefficients should be of opposite sign according as the wave is incident on one side or the other of the boundary. As these two results are inconsistent, it follows that the reflection coefficient should vanish for all angles of incidence and all states of polarisation of the incident light, in the case under consideration.

4. The polarisation of the reflection by a twin plane

The foregoing result may be interpreted physically in terms of the polarisation of the medium produced by the field of the light wave traversing it. If the vibration be perpendicular to the plane of symmetry, the polarisation is obviously the same on both sides of the boundary; there being no discontinuity, there is no reflection. The same situation subsists when the direction of vibration is in the symmetry plane and the incidence is normal. For oblique incidences, the polarisation does indeed exhibit a discontinuity at the boundary, but this lacks a component transverse to the direction of travel of a reflected wave and hence there is no reflection.

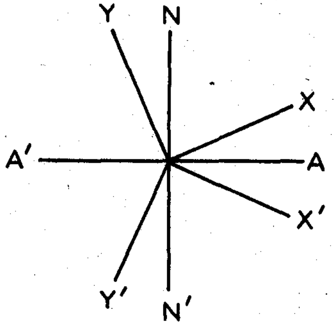


Figure 1

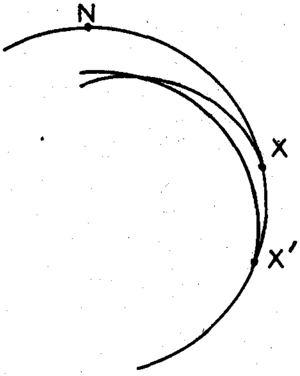


Figure 2

The foregoing considerations assist us to derive in a very simple manner the characters of the reflection at the composition plane of twinning when the direction of travel of the incident light-waves in the medium does not lie in the symmetry plane but makes a small angle with it. The latter condition allows the azimuths and obliquities of incidence to be varied subject only to the restriction that if the azimuthal angle is large, the angle of incidence should be small. In all such cases, the reflection would necessarily be weak and the incident and transmitted waves would therefore differ but little in amplitude. The corresponding directions of vibration would however be different on the two sides of the boundary, and the reflection occurring at the latter would be the result of a sudden change in the direction of the polarisation at the boundary and not of a discontinuity in its magnitude.

In figure 2, the normal to the boundary and the principal vibration directions OX and OX′ on either side of it are shown as points N, X, X′ on a great circle. Two other great circles have been drawn through X and X′ containing respectively the directions of travel of the incident and transmitted waves. These circles cut each other at a small angle and the possible directions of vibration may be assumed to be either parallel or perpendicular to them, since the optical behaviour of potassium chlorate closely approximates to that of a uniaxial crystal with OX or OX′ as the optic axis. The vector difference in the polarisation on the two sides of the boundary which results in a reflection is transverse to the mean direction of the polarisations due to the incident and transmitted waves. Hence, we deduce that if an ordinary wave is incident on the twin-plane boundary, it is reflected as an extraordinary wave and *vice versa.*

The validity of the result stated above is restricted to the cases considered in which the twin-plane reflection is weak in comparison with the incident and transmitted waves. More generally, a discontinuity in magnitude as well as in direction of the polarisation at the boundary would have to be considered. There would then be both an ordinary and an extraordinary reflected wave and not merely one or the other. If, for example, the plane of incidence is perpendicular to the symmetry plane, only in the particular case of nearly normal incidence of the light, and not at all incidences, could we expect the special law of polarisation stated above to be valid.

5. The case of nearly normal incidence

As already remarked, potassium chlorate is strongly birefringent and the refractive indices for waves incident nearly normally are very different for the two possible directions of vibration, namely, 1·52 and 1·47. In these circumstances it is distinctly surprising that the crystals give at such incidences, a single sharply defined monochromatic reflection or a series of such reflections. The reason for this is to be found in the special law of polarisation shown above to be valid for such incidences. Considering a pencil of light incident on the crystal, it divides into two beams travelling with very different velocities, the ordinary ray with the lower velocity and the extraordinary ray with the higher velocity. However, the ordinary ray is reflected at the successive twin boundaries and travels backwards as an extraordinary ray with the higher velocity, while similarly the extraordinary ray is reflected and travels back as an ordinary ray with a lower velocity. Hence the total light path is to a very close approximation the same in both cases, and we observe sharply defined monochromatic bands in the spectrum of the reflected light which give no indication that the stratifications giving rise to them are of a strongly birefringent material.

6. Explanation of the triplet bands

The situation stated above is radically altered when the light is incident obliquely in a plane perpendicular to the symmetry plane. For, in this case, an ordinary wave travelling downwards is reflected back both as an ordinary wave and as an extraordinary wave, while similarly the extraordinary wave is reflected back both as an ordinary wave and as an extraordinary wave. Hence we have four distinct trains of reflected waves, the light paths traversed by the first two of which are so widely different that we can expect them to manifest themselves independently in the spectrum of the reflected light, viz., (i) incident and reflected as ordinary waves, (ii) incident and reflected as extraordinary waves, and (iii) and (iv) incident as ordinary and reflected as extraordinary waves or vice versa. For the reasons stated above, the first and the second groups may be expected to appear as monochromatic reflections well removed from each other on either side of a central group comprised of the third and fourth set of reflections. These outer components would be of vanishingly small intensities when the incidence is nearly normal, but would gain in this respect as the incidence is made more oblique. The third and fourth groups would by reason of the geometric symmetry, have identical optical paths and would therefore not be spectroscopically separable.

7. Explanation of the doublet bands

The case in which light is incident obliquely in an azimuth nearly coinciding with the symmetry plane may now be considered. Here, as we have seen, an ordinary wave is reflected only as an extraordinary wave and vice versa. The light paths however lie on opposite sides of the normal to the face of the crystal. Hence, unless the alternate components of twinning are all of equal thickness, the total light path of a wave entering as an ordinary and reflected back as an extraordinary wave would not be the same as that of a wave entering as an extraordinary and reflected back as an ordinary wave. In these circumstances, the wavelengths for which the reflections reinforce each other would not be the same in the two cases, and the monochromatic reflection observed at nearly normal incidences would split into a doublet, and the doublet separation would increase progressively with increasing obliquity of incidence. As the reflections are weak, the components of the doublet may be expected to be fairly sharply defined. A further remark may be made. Since the appearance of the doublet is a consequence of the optical paths on the two sides of the normal being different, a rotation of the crystal through 180° about the normal should result in a reversal of the situation; the extraordinary and the ordinary reflections would interchange their positions in the spectral doublet when the crystal is rotated through 180°.

8. Some observational facts

We now return to the theoretical result deduced in section 4 above, viz., that in certain circumstances an ordinary wave incident at a twin plane boundary is reflected as an extraordinary wave and vice versa. An observational test of this may be readily made. A bright source of light is viewed by reflection in the crystal set at such an azimuth that the coloured reflection vanishes and the setting is then altered a little so that it reappears. It is then found that if the incident light be unpolarised, the reflected light is also unpolarised, indicating that the reflection coefficients are equal for the ordinary and the extraordinary waves. If however the incident light is plane-polarised in such manner that only either the ordinary ray or the extraordinary ray traverses the crystal, the coloured reflection is also plane-polarised but in a reciprocal fashion; it is quenched if the polariser and analyser are set in parallel positions, but comes through if they are crossed.

As is well known, a strongly birefringent crystal transforms a beam of plane-polarised light traversing it into elliptically polarised light which would not be quenched by an analyser set in any position. In the particular case under consideration, however, this difficulty does not arise, since the plane of incidence is nearly coincident with the symmetry plane of the crystal which contains two of the optical vibration directions while the third is perpendicular to it; the ellipticity introduced in these circumstances is therefore negligible. Its disturbing effect can however be made evident by setting the polariser so that the vibration direction is neither parallel nor perpendicular to the plane of incidence. It is then found that the reflection cannot be quenched at any setting of the analyser.

The foregoing considerations suggest that for a complete study of the laws of twin-plane reflection at all azimuths of incidence, it is necessary to use very thin plates in which the iridescent layer is not overlaid by untwinned material. Observations made with such crystals further confirm the theoretical results deduced in section 4 above and show that in the particular case when light is incident in a plane perpendicular to the symmetry plane, the special law of polarisation is only valid if the incidence of light on the crystal is nearly normal. The observations suggest that at more oblique incidences the vibration in the reflected light is neither parallel nor perpendicular to that in the incident waves. Such a result becomes readily intelligible when it is recalled that at such incidences the reflected light includes both an ordinary and an extraordinary component and not merely one or the other (*vide* section 6 above).

In his theoretical paper on the reflection of light at a twin-plane in a crystal, Rayleigh showed that when the plane of incidence is perpendicular to the plane of symmetry, the polarisation of the reflected ray would be reversed in the act of reflection; in other words if the incident light is polarised in the plane of incidence, the reflected light would be polarised in the perpendicular plane and vice versa. The theoretical derivation was based on the assumption that the birefringence of the crystal is very weak. This assumption is however not in agreement with the facts for the case of potassium chlorate. It is therefore not surprising that Rayleigh was unable to confirm the theoretical prediction by observations except in the particular case of nearly normal incidence.

9. Polarisation of the spectral components

The remarks in the foregoing section refer to the behaviour of the reflected light when examined without the aid of spectral analysis. We shall now pass on to consider the very interesting polarisation effects exhibited by the spectral components of the reflected light in various cases. The facts of observation will be succinctly described and the reader will have no difficulty in verifying that they afford a striking confirmation of the origin of the spectral components given earlier in this paper. It should be understood that the circumstances of observation are invariably those described in the second paragraph of section 4. When reference is made to a polariser or an analyser, it is to be understood that their vibration directions are set such that they are either parallel or perpendicular to the plane of incidence as the case may be.

The case of nearly normal incidence: The unresolved maximum in the spectrum of reflected light is unpolarised if the incident light is unpolarised. If the incident light is polarised, the unresolved maximum is polarised in the *reverse* fashion and is therefore extinguished by an analyser set *parallel* to the polariser.

The triplet bands: The outer components of the triplet described in section 6, when observed with incident unpolarised light are found to be plane-polarised in two mutually perpendicular directions. One or the other component is extinguished when viewed through an analyser appropriately set. If the incident light is polarised in one or another of two perpendicular directions, one or the other of the two components is found to be extinguished. The surviving outer component is also extinguished if viewed through an analyser in the *crossed* position. The central band of the triplet behaves in a fashion similar to the unresolved maximum referred to above in the case of normal incidence.

The doublet bands: With incident unpolarised light, the components of the doublet described in section 7 are plane-polarised in mutually perpendicular directions. In this case, if the crystal be turned round through 180°, the planes of polarisation of the two components are interchanged. With incident polarised light, one or the other component is extinguished. The surviving component is also extinguished if the analyser is set *parallel* to the polariser.

10. Description of the plates

The spectra reproduced in plates I and II illustrate the polarisation effects briefly described above. They were obtained with the crystals designated as A, B, C and D in part I of the present series of papers and whose spectra were illustrated in the plates accompanying that paper. For convenience of reference the crystal used and the setting in which it was placed have been entered at the head of each vertical column in plates I and II. In setting I, the plane of incidence was normal to the plane of symmetry and the angle of incidence was small. In setting II, the plane of incidence was nearly coincident with the plane of symmetry and the angle of incidence was large. P entered in the horizontal row of the figure denotes that the incident light was polarised, but that no analyser was employed, while A denotes that the incident light was unpolarised while the reflected light was analysed. If both P and A are entered, it signifies that incident light was polarised and that the reflected light was analysed. The angle which the direction of vibrations of the polariser and the analyser respectively, make with the plane of incidence have been entered after the letters P and A.

The following special remarks may be made.

Figure A I: The pictures of this crystal with polariser or analyser alone have not been reproduced as they were all similar to each other. The picture appearing in plate I was taken with both polariser and analyser. Notice that the reflection is weak when the polariser and analyser are parallel and strong when they are crossed.

Figure A II: Note the change in spectral width and the spectral shift of the reflection with the change in position of the vibration direction of the polariser or the analyser alone. Note also that the reflection vanishes when the polariser and analyser are parallel and that it appears when they are crossed.

Figure B I: Note the plane polarisation of the outer components of the triplet and fact that they almost disappear when the polariser and analyser are crossed and that one or the other of the outer components reappears strongly when they are parallel. The central band becomes very weak when they are parallel, whereas it comes out strongly when they are crossed.

Figure B II: Note the nearly perfect polarisation of the components of the doublet and that one or the other appears when polariser and analyser are crossed whereas both of them disappear when they are parallel.

Figure C I: The same remarks given for figure B I apply to this figure also.

Figure C II: The same remarks given for figure B II apply to this figure also.

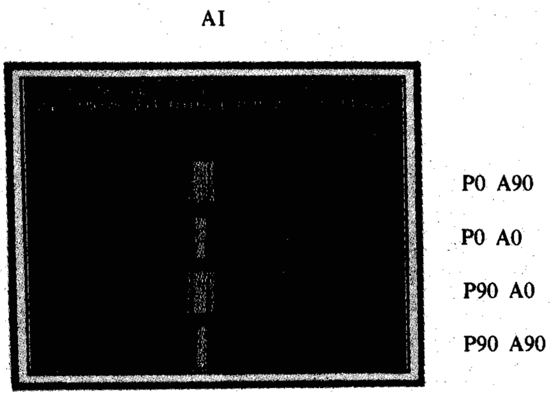
Figure D I: The outer components of the triplet are themselves double. Otherwise the same remarks apply to this figure as in the case of figures B I and C I.

Figure D II: Each of the components is now double and the two subsidiary components behave in opposite ways. Otherwise the remarks are the same as in the case of B II and C II.

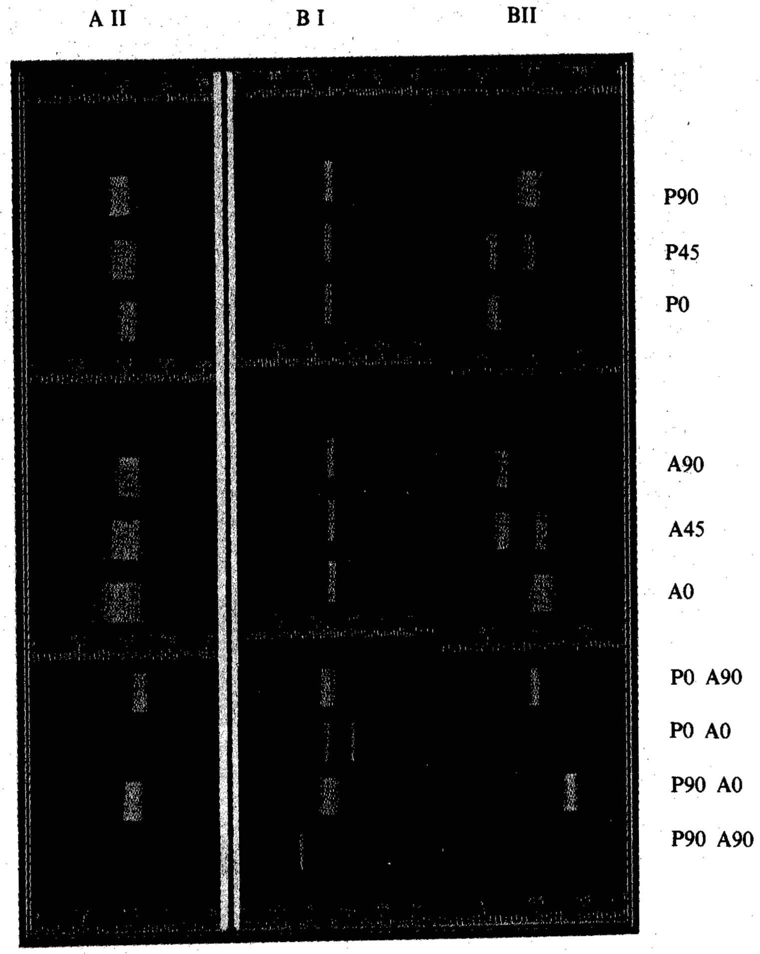
11. Summary

A simple explanation is given why the reflections vanish when the light is incident in the symmetry plane of the twinned crystals. Physical considerations enable the law of polarisation for twin-plane reflections to be derived for directions of incidence adjoining the plane of symmetry. It is further shown that the spectral character of the reflections stands in the closest relationship with the polarisation law thus derived. An explanation is given for the appearance of doublets and triplets respectively in the spectra observed when the incidence is oblique and the azimuth of such incidence is nearly zero and 90° respectively. It is a necessary consequence of the theory that the outer components in these two cases should be plane-polarised, in one case in the reciprocal fashion and in the other case normally. The theoretical results are strikingly confirmed by observations on the polarisation of the spectral components. Photographs are reproduced illustrating these effects.

(1)



(2)



Figures 1 and 2

Plate I

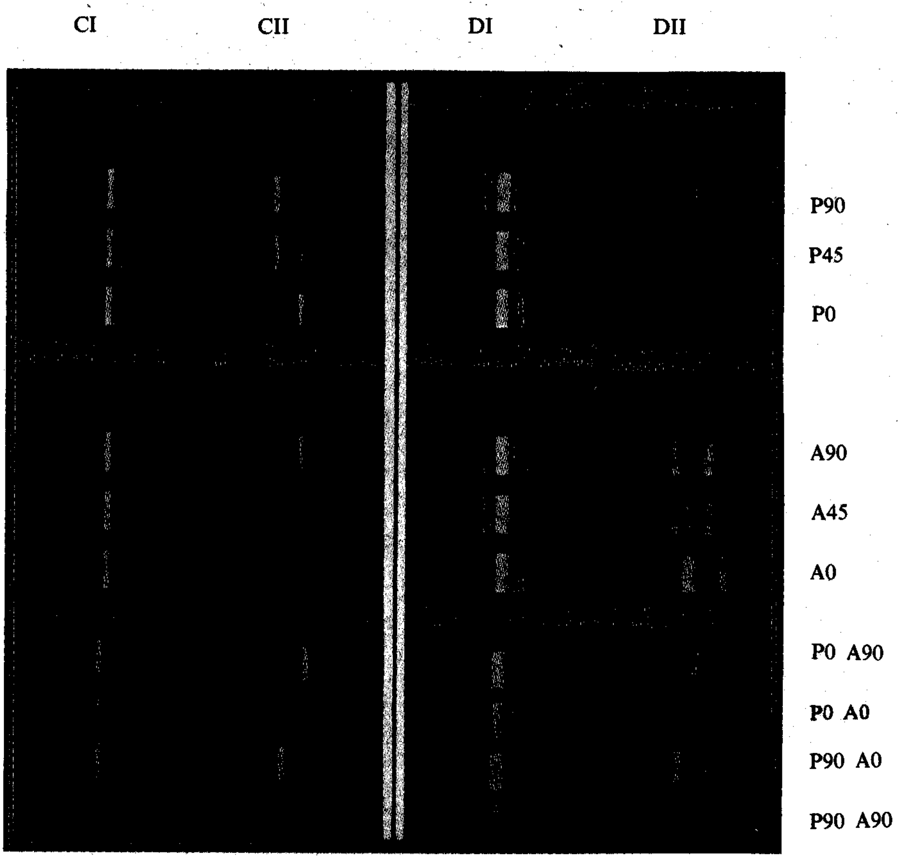


Figure 3

Plate II