

CRYSTALS OF QUARTZ WITH IRIDESCENT FACES

BY SIR C. V. RAMAN

(From the Raman Research Institute, Bangalore)

Received May 10, 1950

1. INTRODUCTION

THE optical phenomena forming the subject of this paper are exhibited in a very striking manner by two small crystals of colourless transparent quartz which were obtained by the author some years ago from a jeweller in Bombay. An enlarged photograph of one of the crystals is reproduced as Fig. 1 (*a*) in Plate XV accompanying the paper. As will be seen from the illustration, one of the pyramidal faces terminating the crystal exhibits a brilliant reflection. The colour of this at normal incidence is a bright green and changes to blue and then to bluish violet as the incidence is made more oblique. At the same time, the iridescence becomes distinctly weaker and much less saturated in hue. The coloured reflection is seen over the whole area of the face with fairly uniform colour and intensity. No trace of the phenomenon is exhibited by any of the other pyramidal faces of the crystal or by any other part of its surface. The second crystal similarly shows a brilliant iridescence on the largest of its pyramidal faces, its colour and intensity, remarkably enough, being practically indistinguishable from those of the first crystal. The second crystal shows, in addition, an iridescence of the same colour on two other pyramidal faces of smaller size, but not on a fourth which is adjacent to the largest iridescent face and of comparable area.

2. THE NATURE OF THE IRIDESCENCE

That the iridescence is not due to superficial films adhering to the crystals is obvious on an examination of its features. Had it had been of that nature, the area of colour would be that covered by the film and would not extend beyond it. Actually, when the crystal is held suitably with respect to the incident light, one observes a brilliant blue band of iridescence along the edge of the face but outside its area, while the face itself remains dark. This effect which is illustrated in Fig. 1 (*d*) in Plate XV is due to light which enters the crystal face near its edge and after suffering a reflection *within* the crystal, emerges through the *contiguous* face. When the crystal is held appropriately, it is possible also to observe the converse effect, *viz.*, light enters the crystal through the contiguous face, and after reflection within the crystal manifests itself as a bright blue band parallel to the edge of the face but

inside it, the face itself not displaying the usual iridescence. In suitable circumstances, also, both effects may be simultaneously observed, bright blue bands being seen alongside the edge on either side, the latter itself appearing as a dark line of separation. That the iridescence arises from reflection inside the crystal and not at the external surface is further evident from the fact that the outer margin of the coloured area is, in general, not exactly coincident with the edge of the face; between them appears a strip without colour and of width varying with the obliquity of incidence of the light and with the setting of the edge with respect to the direction of observation. This effect is illustrated in Fig. 1 (c) of Plate XV in which the lower edge of the face is seen distinctly doubled.

It is thus evident that the iridescence is due to a layer of material which lies below the surface of the crystal and forms an integral part of its structure. The observations indicate the thickness of the layer to be of the order of a quarter of a millimetre. It follows that the coloured reflection from within the crystal would be diluted by the reflection of white light from its exterior surface in an increasing measure as the incidence is made more oblique. This is actually observed, as has already been indicated, and the explanation given is confirmed by the effect of immersing the crystal in a beaker of water, or better still, xylene. By such immersion, we weaken or suppress the white-light reflection, and the iridescence is thereby notably improved in intensity as well as in respect of the saturation of its hue. The polarisation of the white-light reflection at oblique incidences may also be utilized to reduce its intensity by viewing the crystal face through a suitably orientated polaroid. At the polarising angle for quartz, the external reflection is completely suppressed, and the internal reflection which is then of a violet colour appears of a saturated hue, though to some extent it is enfeebled in intensity by its own partial polarisation.

As is to be expected from the intensity of the iridescent reflection, the crystal faces which show the phenomenon exhibit the complementary tint by transmitted light in a conspicuous manner. The transmission colour for normal incidence is a lively rose-pink; this fades away to pale yellow and finally to white as the incidence becomes more oblique.

3. ITS SPECTRAL CHARACTER

Spectroscopic examination of the iridescence exhibited by the crystal faces shows it to be essentially a *monochromatic reflection*. The reflected light at normal incidence exhibits a few narrow streaks close to each other in the green region; as the incidence is made more oblique, these streaks move towards the violet end of the spectrum. Examination of white light

transmitted normally through the crystal face reveals a set of narrow bands of extinction in the same position in the spectrum as the bright streaks observed by reflected light. These effects are illustrated in Figs. 2(a), (b) and (c) in Plate XV, the reflection and extinction bands being indicated by arrows. The photographs were taken with sunlight as the source. The diffuse background seen in the reflected spectra reproduced as Figs. 2(a) and (b) is due to the reflection of white light from the surface and some parasitic illumination returning from the rear of the crystal. The former effect becomes more important at oblique incidences. In Fig. 2(c), the extinction bands due to the crystal are those marked, the rest being the Fraunhofer lines in the solar spectrum.

From the facts stated, it is clear that the iridescent material is essentially a stratified medium consisting of a great many parallel layers of extreme thinness which is more or less exactly the same for all the layers. If we assume the iridescence observed at normal incidence to be a second-order reflection, it would follow that the individual laminae would be 0.34μ thick, and as the thickness of the entire iridescent layer is about 250μ , there would be room in it for about 700 layers. If these were all of exactly the same thickness, they would result in extremely sharp monochromatic reflections and extinctions.

4. THE NATURE OF THE STRATIFICATIONS

As is well known, quartz exhibits two principal kinds of twinning, *viz.*, the electrical and the optical, which may also be described as the orientational and chiral types of twinning respectively. We may evidently dismiss the electrical type of twinning without further consideration in relation to our present problem. Chiral twinning is very common in quartz and frequently exhibits itself as a polysynthetic or lamellar twinning in which the right-handed and left-handed layers are parallel to each other and to the rhombohedral planes of the crystal structure. Such polysynthetic twinning is indeed characteristic of "amethystine" quartz and exhibits itself prominently when a section cut normally to the optic axis of the crystal is viewed through a polarising microscope. The thickness of the twinning layers is known to be highly variable; it may be sufficiently large to be seen without optical aid, or so small as to be invisible even under the polarising microscope. Large variations have been observed even within a single section plate.

The two crystals now under study do not exhibit the slightest hint of amethystine colour. Nevertheless, in their external form, they show the same features as amethystine quartz, and indeed reproduce perfectly the features observed in a great number of crystals of that material collected by

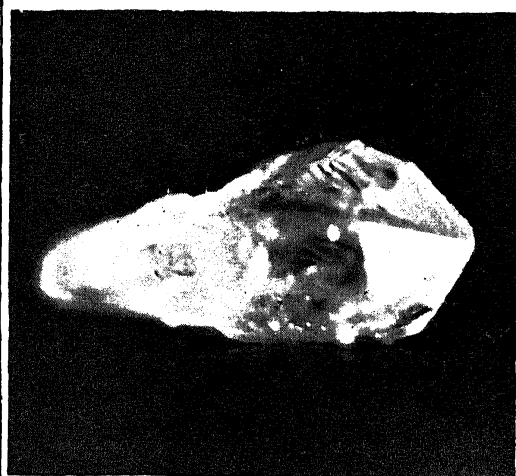
the author from the campus of the Osmania University at Hyderabad and deposited in the Museum of this Institute. The prismatic faces are completely missing, and their place is, for the greater part, taken by an array of terraces running in zig-zag fashion parallel to the rhombohedral faces of the crystal. One such set of terraces, lying below a pyramidal face which does not exhibit iridescence is visible in Fig. 1 (a) in Plate XV. Though the matter has not been tested by section-cutting, since this would have involved destruction of the specimens, it may be taken for granted that the crystals displaying iridescence consist of polysynthetically twinned quartz in which the right-handed and left-handed forms lie in layers parallel to the rhombohedral faces.

Chiral polysynthetic twinning in quartz would not, however, necessarily give rise to iridescence. Had this been the case, the phenomenon would have been noticed and reported long ago by many observers. It is obvious that a crystal face would not exhibit any visible colour by reflection unless the laminations contiguous to it have a sufficient reflecting power and are sufficiently fine and sufficiently numerous to build up a coherent monochromatic iridescence. Even in the two crystals forming the subject of the present study, the conditions necessary for iridescence evidently exist only for some of the crystal faces and not for the others. It may be remarked in this connection that an examination of their external forms reveals some striking differences between the pyramidal faces which display iridescence and those which do not. The latter are terminated by zig-zag lines below which lie a succession of terraces well separated from each other. On the other hand, in all the four cases in which iridescence is observed, we notice immediately below the iridescent face, an area having the correct crystallographic orientation for a prismatic face but presenting a peculiar grooved and eroded appearance; where this meets the iridescent face, we notice a step-like configuration, indicative of a laminated structure of the material parallel to the face. These features can be made out by a study of Figs. 1 (a), (b), (c) and (d) in Plate XV.

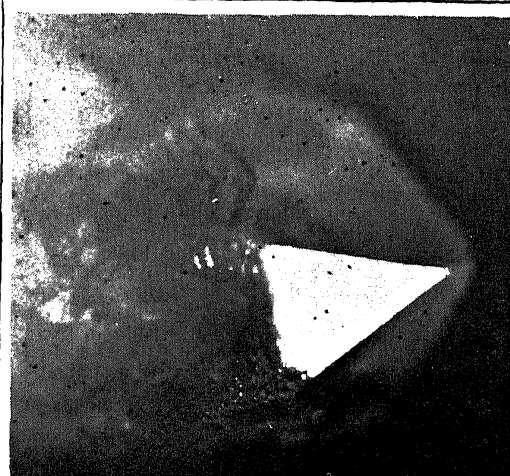
5. SOME FURTHER CONSIDERATIONS

The suggestion made above that the stratifications giving rise to the iridescence in quartz are of the polysynthetic chiral type raises some further questions. The iridescence exhibited by some specimens of potassium chlorate was explained long ago by Rayleigh as due to polysynthetic twinning in the monoclinic crystals of that substance. The special features observed in the case of potassium chlorate, *viz.*, the disappearance of the iridescence at normal incidence and its periodic appearance and disappearance at all incidences when the crystal is rotated in its own plane, are consequences

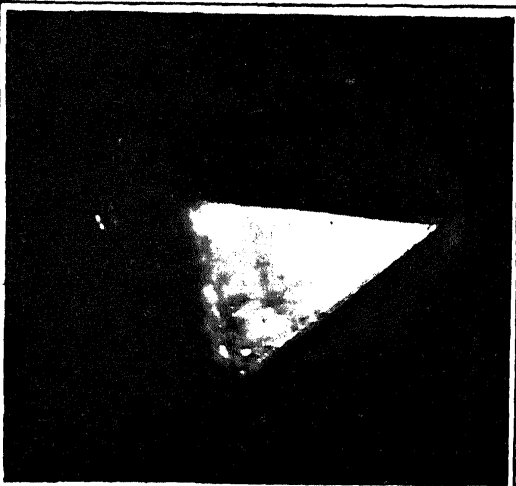
(a)



(b)



(c)



(d)

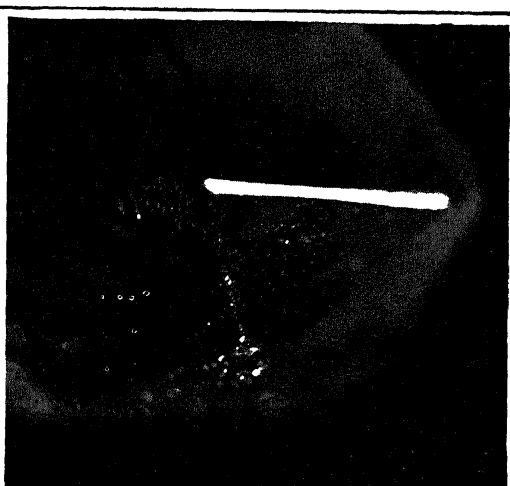
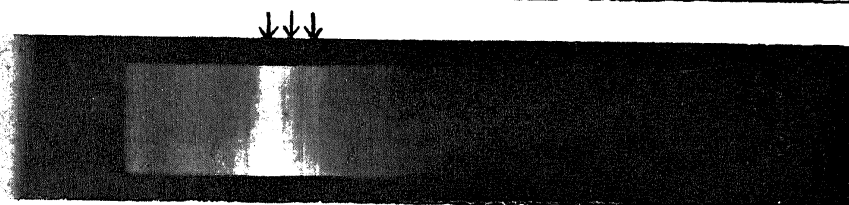
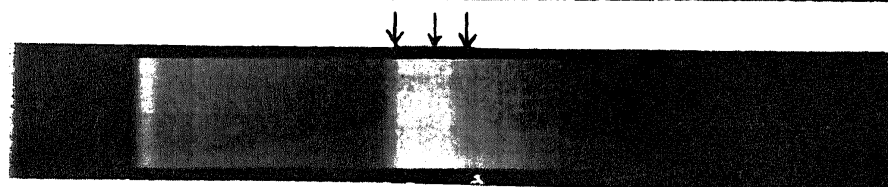


FIG 1

(a).



(b)



(c)

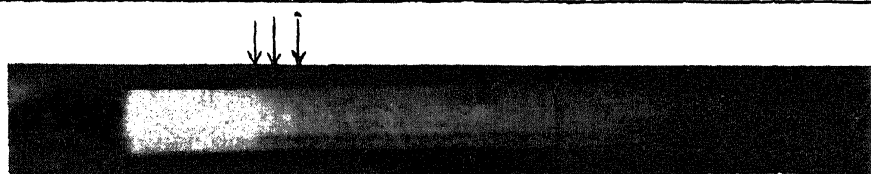


FIG. 2

124
113



of an orientational type of twinning. It is not surprising that these features are not met with in the case of quartz. The orientational twinning in the case of potassium chlorate also results in some peculiar polarisation effects. No such effects are observed in the case of the iridescence of quartz. Provided that the depolarisation due to the passage of the light through the quartz is avoided or duly taken account of, the iridescent reflections are found to exhibit a normal behaviour, in other words, they show, at least qualitatively, the degree and kind of polarisation to be expected for reflection at the boundary between two media of nearly equal refractive index.

Finally, we are faced with the question whether the optical difference between layers of right-handed and left-handed quartz is sufficient to give an appreciable reflection of light at the boundary of separation between them. If the answer to this question be in the negative, we would be faced with a dilemma, since it would be difficult to explain otherwise the iridescence which is actually observed and which is so clearly related to the architecture of the quartz crystal. It seems, however, just possible that the polysynthetic twinning may be accompanied by a segregation at the boundaries between the right-handed and left-handed quartz of extremely thin layers of impurity material. Such layers when regularly disposed could give rise to coherent reflections of sufficient intensity to explain the observed iridescence.

The photographs illustrating this paper (Plate XV) were obtained by Mr. A. Jayaraman, whose highly competent assistance has also otherwise been of value in the work described.

SUMMARY

The paper is a study of the iridescence exhibited by some of the pyramidal faces of two crystals of transparent quartz in the possession of the author. Spectroscopic examination shows the reflections and the corresponding extinctions to be monochromatic. The thickness of the iridescent layer may be directly observed, and the number of laminations inferred to be present is sufficient to explain the observed characters of the iridescence. The nature of the stratifications in the crystal giving rise to the iridescence is discussed. Photographs of one of the crystals and of the spectral character of the reflections and extinctions illustrate the paper.