

THE OPTIC INTERFERENCE FIGURES OF AMETHYSTINE QUARTZ—PART I

BY S. PANCHARATNAM

(Memoir No. 107 of the Raman Research Institute, Bangalore-6)

Received April 11, 1958

(Communicated by Sir C. V. Raman)

§1. INTRODUCTION

AMETHYST is the familiar purple modification of quartz having an absorption band in the yellow-green region of the spectrum, a study of whose optical properties had been reported in an earlier paper.¹ Observations on untwinned amethystine sectors through a polaroid, showed that they exhibited pronounced pleochroism even when observed along the *c*-axis—showing that the *c*-axis was *not* an axis of uniaxial symmetry for the absorption ellipsoid. It followed that the material could not be strictly uniaxial. This was indeed confirmed, though only in the cases of very intensely coloured sectors, was the biaxial nature quite pronounced—the *c*-axis being the acute bisectrix. A sector of this type which was pre-eminently suited for the present study (*i.e.*, which did not possess certain extraneous complicating features characteristic of most quartz-amethyst plates) had been obtained by careful selection during our previous investigation. Of the various conclusions which emerged from that investigation it is sufficient for our present purpose to accept the following particular results, which may be independently deduced from certain observations described in the present paper itself (§§3 and 4):

(a) The amethystine sector used is an optically untwinned and genuinely biaxial medium possessing optical activity.

(b) The sector is pleochroic, the elliptic section of the absorption ellipsoid normal to the acute bisectrix having its major and minor axes lying respectively parallel and perpendicular to the axial plane.

The optic interference figures displayed in convergent light by this class of media (*viz.*, the class of biaxial media possessing both pronounced pleochroism and optical rotatory power) have not been investigated and are therefore of interest. In the present paper we reproduce and discuss the remarkable types of interference figures exhibited under various conditions by the selected amethystine sector. These interference figures are found to

differ considerably from those displayed both by inactive absorbing crystals and active transparent crystals. It will be shown however that the observations described become explicable in detail in the light of the theory of light propagation in absorbing crystals possessing optical activity; this theory was presented in an earlier paper² hereafter referred to as P-*a*. We shall also have frequent occasion to refer to a paper³ (hereafter referred to as P-IV) where the interference figures of absorbing biaxial crystals had been discussed.

Though it should certainly be possible to repeat our experiments with other deeply coloured sectors of comparable intensity, the impression should not be created that this can be done with any densely coloured area in a basal section of amethyst randomly selected. According to our previous investigation, this is because of certain very interesting—though, from our present point of view, extraneous—complicating factors which feature prominently in most quartz-amethyst plates. In the appendix at the end of Part II, attention is briefly drawn to these complicating factors, and also to the fact that it becomes difficult to compare our results directly with those of other workers.

§2. PRELIMINARY REMARKS

In the present study we have used an intensely coloured amethystine sector occurring in a roughly basal section-plate of the quartz-amethyst about 3 mm. thick. In our previous investigation, an examination of the etch patterns shown by the two surfaces of the plate had revealed that the portion to be used was untwinned and was *right-rotating*, the sector being associated with a secondary rhombohedral face. The tremendous depth of absorption may be estimated from the wedge-spectrograms published in that paper—taken with this coloured area. Because of the large absorption the optic interference figures reproduced in the present paper had to be photographed by focussing a reduced image of a sodium (or mercury) arc directly on the sector using a battery of lenses which also served as the converger. Only an area about 3 mm. square in the body of the sector was used: the remainder of the plate was blackened on both sides with paint to exclude light from adjoining uncoloured and twinned areas. In what follows (particularly in Part II) an acquaintance with the behaviour in convergent light of transparent biaxial crystals—both inactive and active—will be presumed.

§3. OBSERVATIONS IN BLUE LIGHT

Figures 1, 2 and 3 (Plate IX) exhibit the interference figures observed in blue light (λ 4358) at three different settings of *crossed* polaroids. They

prove clearly the biaxial nature of the material, the axial angle for this wavelength being about 8° ; the axial plane has been kept horizontal in these photographs—as well as in all the others reproduced in this paper. In Fig. 1 the vibration-directions of the polariser and the analyser lie along and perpendicular to the axial plane while in Fig. 2 they are at 45° to the axial plane. Fig. 3 has been obtained by interchanging the positions which the polariser and analyser occupy in Fig. 2. In these as well as in all other settings of the crossed polaroids it was observed that—unlike the behaviour obtaining in transparent inactive crystals—the optic axial directions were never extinguished; though at the border of the figure the isogyres were fairly dark and appeared at the same position as in non-active crystals, they became diffuse and vanished as the optic axial directions were approached. *This behaviour is characteristic of an ordinary optically active crystal*; and the fact that there is no sensible difference between Figs. 2 and 3 is only what is to be expected for a *transparent* crystal of this class. (This may be shown from reference 4, Eqns. 1 and 2.) In a transparent optically active crystal the optic axial directions may be extinguished by keeping the polariser setting fixed, and rotating the analyser away from the crossed position by an amount equal to the rotation of the plane of polarisation along the optic axial directions. This has been done in Fig. 4, where in addition—purely for achieving a symmetrical interference figure—the polariser and analyser have been adjusted so as to be symmetrically situated with respect to the axial plane. The mean value of the rotation measured in this approximate manner for six azimuths of the polariser was found to be about -115° . This being also the rotation for a plate of quartz of the same thickness (*viz.*, 0.28 cm.) the untwinned nature of the medium is again confirmed. (That the material was right-rotating was established—independent of the etch study—by the fact that the rings expanded when the analyser was rotated clockwise).

A curious property not displayed by transparent crystals was however detected. The measured rotation varied systematically with the azimuth of the incident vibration, deviating by as much as ten per cent. from the mean value. This phenomenon will be theoretically explained later (§5); it arises from the fact that the plate is so intensely coloured that even for the blue region of the spectrum the dichroism is not altogether absent, though it is small especially when compared to the optical rotation. Since the latter diminishes with increasing wavelength such effects of dichroism may be expected to be even more prominent for corresponding wavelengths on the long wavelength side of the absorption band. Before proceeding to describe such observations with red light, it is well to confirm also the property

(b) mentioned in the introduction: this can be best done by using a wavelength (λ 5890) lying in the heart of the absorption band.

§4. BREWSTER'S BRUSHES

Figure 5 shows the so-called Brewster's brushes as revealed without the use of either polariser or analyser in the unpolarised yellow light of a sodium lamp. It will be seen that they consist of two pairs of brushes, one pair of brushes being associated with each optic axis. For the present we may consider the phenomenon from the elementary standpoint that the behaviour should approximate to that of inactive absorbing crystals as we approach directions represented by points near the border of the field of view. The position of the brushes is in fact the same as exhibited in a crystal such as yttrium platinocyanide⁵ which has a small axial angle and is inactive.

In non-active crystals Brewster's brushes will be seen only when there is pronounced dichroism along the optic axial directions; and they will then lie along zones for which the principal planes of linear birefringence are inclined at 45° to the principal planes of linear dichroism. In the present case the brushes are seen to lie along the same zone as the isogyres of the biaxial interference figure when the polariser and analyser are at 45° to the axial plane (refer *e.g.* Fig. 2). This means that the principal radii of the elliptic section of the absorption ellipsoid (taken normal to the acute bisectrix) lie along and perpendicular to the axial plane, *i.e.*, along $O'X_k$ and $O'Y_k$ in Fig. 1, text, of Part II: this diagram represents the field of view of the convergent light figure, O' representing the acute bisectrix, while O_1 and O_2 represent the two optic axes.

Which of the directions $O'X_k$, $O'Y_k$ correspond to the less absorbed vibration in the absence of optical activity could be determined in the following manner. The plate was tilted first about a horizontal axis (so as to effectively eliminate the effects of optical activity) and the depth of absorption for the horizontal vibration direction was noted in *parallel* light. An analogous procedure was adopted for observing the depth of absorption for a vibration parallel to $O'Y_k$; this was seen to be enormously greater than that for the vibration $O'X_k$, thus independently confirming the statement (b) made in the introduction.

Though the explanation of Brewster's brushes given in this section appears to have completely neglected the presence of optical activity, it actually holds to a higher degree of approximation than is to be suspected from such a statement. According to P-a, §7 b, we may—for directions not too close to an optic axis—neglect the *squares* of the ellipticity of the waves

(and hence the square of the non-orthogonality factor $\cos \frac{1}{2}c$) even though the first power of these quantities may not be negligible. For such directions it has also been shown that the absorption coefficients of the waves propagated along any direction will be virtually the same as in the absence of optical activity. Referring now to Eqn. (11) of P-IV it follows that the intensity at any point in the field seen directly in unpolarised light will be the same as in an inactive crystal (neglecting the squares of the ellipticity of the waves, in both cases). Hence the explanation of Brewster's brushes as customarily presented for an inactive absorbing crystal (Pockels,⁶ p. 424) may be also applied in the present case—(see also Part II, §7).

In the present paper we shall be making the following approximation which is quite customary, *viz.*, that the elliptic section of the absorption ellipsoid made by the plane of the paper in Fig. 1, text, Part II, is practically the same (in orientation and dimension) as the section normal to any direction of propagation in the angular range considered. [That this approximation is not strictly true is shown by the asymmetry of the intensity of Brewster's brushes with respect to the axial plane (see Appendix)]. For the purpose of our qualitative discussion we shall also take the rotatory power to be constant over the range of directions under consideration.

§5. OBSERVATIONS IN RED LIGHT

Figures 6, 7 and 8 (Plate X) show the biaxial interference figures exhibited in red light (6450–6900 Å) at three different settings of *crossed* polaroids which correspond respectively to the three settings already used in Figs. 1, 2 and 3 with blue light. Apart from the very appreciable increase of the axial angle (relative to blue light), the most striking feature is that Figs. 7 and 8 are *not* identical since the brush apparently goes through the eyes in the latter. Any such alteration in the interference figure caused by a mere interchanging of crossed polaroids implies that the waves propagated along a general direction are in two general non-orthogonally polarised states, and that *the relation between these states of polarisation is necessarily different from that obtaining in inactive absorbing crystals* (see P-IV, §8 a).

It was observed that in the setting of Fig. 8—for which the vibration-direction of the polariser is inclined at -45° with respect to the axial plane—the portion of the brush which apparently passes through the eyes was really not perfectly dark; and that the optic axial directions could be more perfectly extinguished by turning the analyser clockwise, away from the crossed position, through a small angle—the mean value of this angle for obtaining best extinction being estimated to be 31° . On the other hand when the vibration-direction

of the polariser made an angle of $+45^\circ$ with respect to the axial plane (as in Fig. 7), the analyser had to be turned (from the crossed position) through the much larger angle of 50° to reach a position where the optic axis appeared best extinguished. For any general setting of the polariser the axial directions can be extinguished by a similar procedure, as illustrated in Fig. 9 (where, in addition, the polariser and analyser have been oriented symmetrically with respect to the axial plane).

Along an axial direction in a *transparent* active crystal two oppositely polarised circular vibrations are propagated with 'circular birefringence' $|2\rho|$. In the present case such a supposition cannot account for the phenomenon noted in the previous paragraph—which had also been exhibited less conspicuously in blue light, *viz.*, the variation of the apparent rotation with the azimuth of the incident vibration. But in P-*a*, §8 *a*, it had been theoretically deduced that in an optically active crystal with weak linear dichroism ($k < |2\rho|$) the waves propagated along an axial direction will be *elliptically* polarised: the two elliptic vibrations will be exactly similar in form and orientation but will be described in opposite senses. The major axes of the ellipses will be coincident and—since the rotatory power is negative in the present case—will make an angle of -45° with respect to the line $O'X_k$ (Fig. 1, text, Part II) which is the trace of the axial plane as well as the trace of a principal plane of linear dichroism.

Before proceeding further we shall describe an experiment confirming the theoretical results stated in the last paragraph. The crystal was as usual kept in the conoscopic arrangement between crossed polaroids which could be turned around *together*; but a mica retardation plate was set immediately behind the first polaroid and an *identical* retardation plate was set immediately in front of the second polaroid with its principal planes turned round by 90° with respect to the first. The two retardation plates were coupled so that they could be turned around *together*. The arrangement in front of the crystal constitutes an '*elliptic polariser*'. The arrangement behind the crystal constitutes a '*crossed elliptic analyser*' which, if the crystal plate were absent, would *automatically* cut out the elliptically polarised light produced by the arrangement in front—independent of the setting of the coupled polaroids and the coupled retardation plates. In the present case, $\lambda/4$ retardation plates were used, the principal planes being oriented at an angle of 45° to the axial plane. Thus the principal axes of the incident elliptic vibration were equally inclined to the axial plane, though the ellipticity could be altered by turning the coupled polaroids. It was observed that there were two settings of the coupled polaroids at which the optic axial

directions were extinguished—the figure for one of these settings being reproduced in Fig. 10. These were the two settings at which the vibration-direction of the first polaroid made angles of $+34^\circ$ and -34° with respect to O'M (Fig. 3 *b*, text, Part II) as roughly determined by the fact that the spots had moved exactly into the axial plane at these settings. *This confirms that the two waves propagated along each optic axis are elliptically polarised as described above—the numerical value of the ellipticity $|\omega|$ of both vibrations being about 34° . On the Poincaré sphere (Fig. 1, text), the states of polarisation of these two waves are denoted by the points A and B lying on the same meridian of longitude but at latitudes -2ω and $+2\omega$ respectively.*

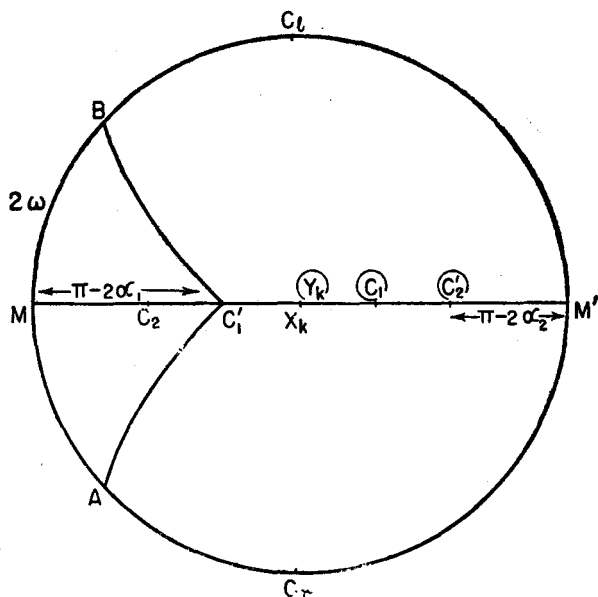
The variation of the measured rotation with the azimuth of the incident vibration may now be understood by applying the results of ref. 7, §§4 and 5, for discussing the dissolution of the incident vibration into two elliptic vibrations, and the compounding of the latter after their propagation through the plate with different velocities but equal absorption coefficients. The actual rotatory power along an axial direction can in fact be calculated from the apparent rotations α_1 and α_2 observed with the incident vibration lying respectively parallel and perpendicular to the major axes of the elliptic vibrations propagated. Considering first the former case the incident vibration will be represented on the Poincaré sphere by the point M on the equator (Fig. 1, text, Part I). It will be decomposed into two vibrations in states A and B whose intensities will be equal and whose initial 'phase difference' will be zero since M lies on the arc AB (reference 7, §4). Hence if the slower wave B suffers a phase retardation of ϕ relative to the other on propagation through the plate along an axial direction, then ϕ also represents the absolute phase advance of vibration A over B when they emerge from the plate. These vibrations will still be of equal intensity—their absorption on passage through the plate being equal according to P-*a*, §8 *a*—so that they will compound to give a *linear* vibration represented by a point C_1 on the equator (reference 7, §5); if E_1' be the area of the triangle $C_1'BA$, where C_1' is the point opposite to C_1 , then $\frac{1}{2}E_1'$ should be equal to the supplement of the absolute phase advance of vibration A over B (see reference 7, Eq. 5 *b*). The spherical excess $\frac{1}{2}E_1'$ of the right-angled triangle $C_1'BM$ may be obtained from spherical trigonometry:

$$\tan \frac{1}{2}E_1' = \tan \omega \tan \frac{1}{2}(\pi - 2\alpha_1).$$

Hence

$$\cot \frac{1}{2}\phi = \tan \omega \cot \alpha_1 \tag{1}$$

On the other hand, if the incident vibration is turned to the orthogonal state M' , the two vibrations into which it is split along an optic axial direction will have an initial phase difference of $-\pi$ and a final phase difference



TEXT-FIG. 1. The Poincaré sphere as used for explaining the apparent variation of optical rotation with the azimuth of the incident vibration. Symbols enclosed in circles represent points on the hemisphere below the plane of the paper.

of $(\phi - \pi)$, compounding finally to give the linear vibration in state C_2 . Hence if C_2' represent the state orthogonal to C_2 , then ϕ should be equal to half the spherical excess of the triangle $C_2'AB$. Proceeding as before we get from spherical trigonometry,

$$\tan \frac{1}{2}\phi = \tan \omega \tan a_2 \quad (2)$$

From (1) and (2) we obtain

$$\tan^2 \omega = \frac{\tan a_1}{\tan a_2} \quad (3)$$

$$\tan^2 \frac{1}{2}\phi = \tan a_1 \tan a_2 \quad (4)$$

The apparent rotations a_1 and a_2 observed in the respective cases when the incident vibration lies parallel to $O'M$ and $O'M'$ (Fig. 3 *b*, text, Part II) have been roughly measured to be 31° and 50° respectively (refer above). Inserting these values in (3) and (4) we find firstly that the numerical ellipticity $|\omega|$ of the vibrations propagated along an optic axis should be $35 \cdot 5^\circ$ —in

FIG. 1

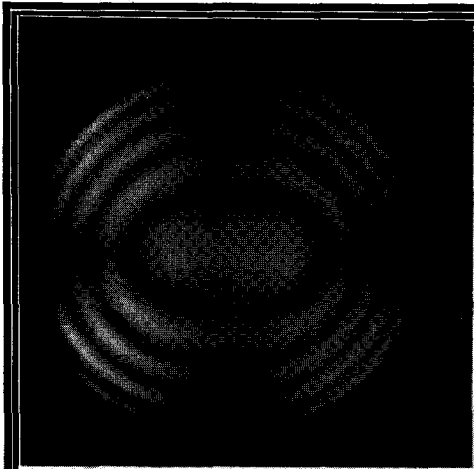


FIG. 2

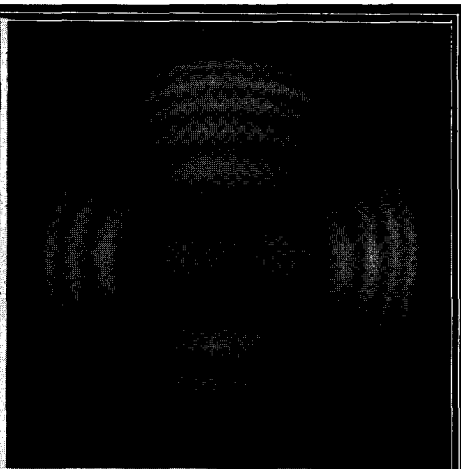


FIG. 3

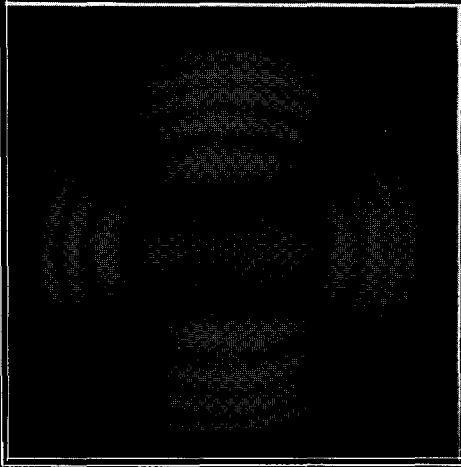


FIG. 4

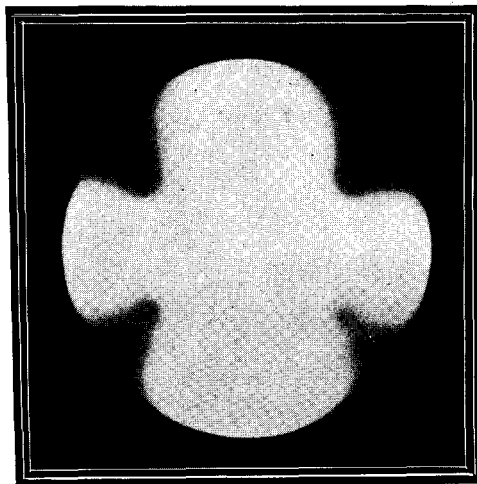
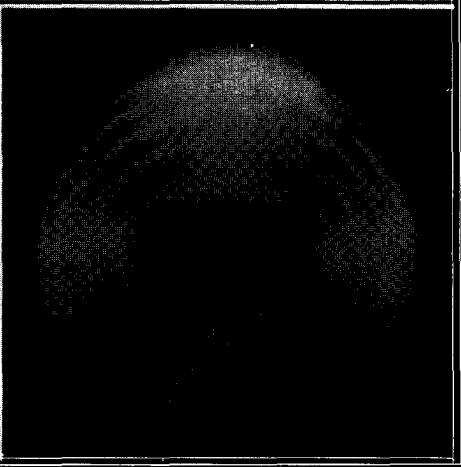


FIG. 5

FIGS. 1-5

FIG. 6

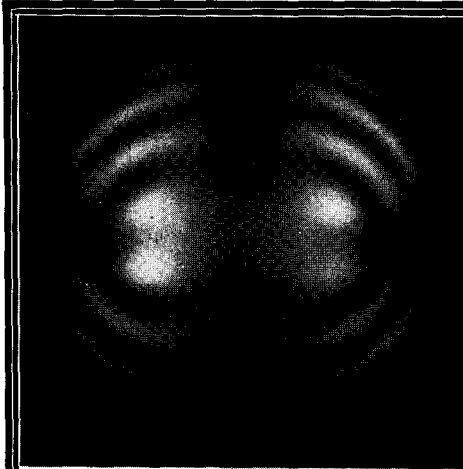


FIG. 7

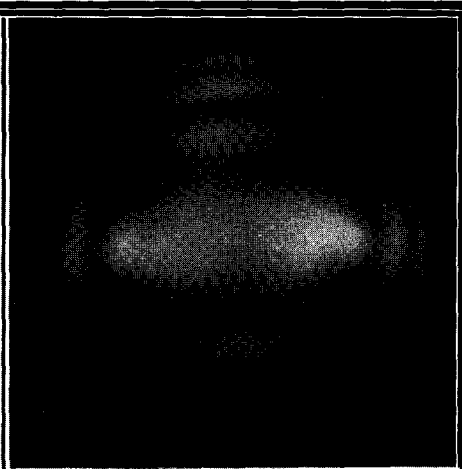


FIG. 8

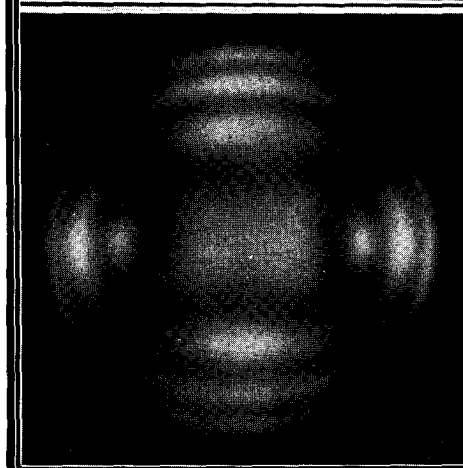


FIG. 9

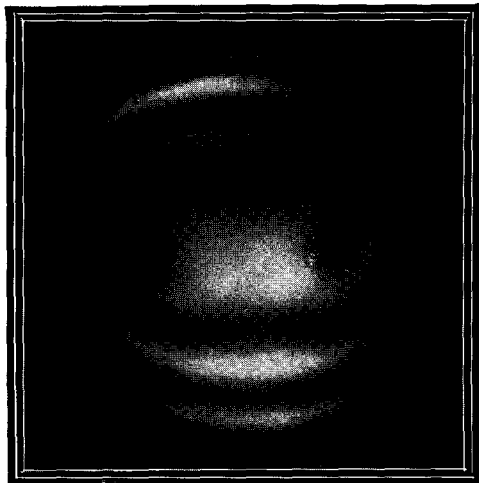
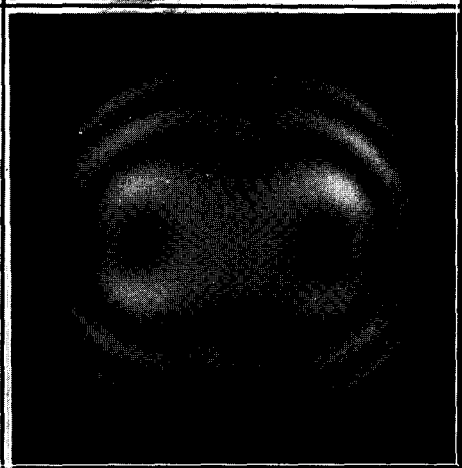


FIG. 10

agreement with the experimentally estimated value of 34° , previously quoted; secondly, the relative phase advance ϕ gained by the faster wave over the slower wave on passage through the plate is found to be 80.5° . Since the waves are not circularly polarised, the relative phase difference per unit path (ϕ/d) is *not* directly equal to $|2\rho|$ but is related to it by Eq. (17) of P-a:

$$|\rho| d = \frac{\frac{1}{2} \phi}{\sin |2\omega|} \quad (5)$$

The true optical rotation $|\rho| d$ which would be exhibited by the plate in the absence of dichroism may be calculated from (5) by substituting the values of ω and ϕ obtained from (3) and (4). In the present case it is found to be 42.5° . (Though this is somewhat lower than the optical rotation for a plate of transparent quartz, the difference may be entirely due to the large experimental errors involved).

§6. SUMMARY

The interference figures exhibited by an intensely coloured amethystine sector at two wavelengths which lie practically outside the absorption band are reproduced. In blue light the interference figures are characteristic of a transparent biaxial crystal possessing the rotatory power of quartz. In red light the axial angle is larger, and effects due to weak linear dichroism are also exhibited. Thus the rotation of the plane of polarisation along the optic axes varies with the azimuth of the incident vibration; and the method of determining the rotatory power from the apparent rotation is illustrated.

§7. REFERENCES

1. Pancharatnam .. *Proc. Ind. Acad. Sci.*, 1954, **40 A**, 196.
2. ————— (P-a) .. *Ibid.*, 1957, **46 A**, 280.
3. ————— (P-IV) .. *Ibid.*, 1957, **45 A**, 2.
4. ————— .. *Ibid.*, 1957, **45 A**, 402.
5. Boguslawski .. *Ann. der Physik*, 1914, **44**, 1084.
6. Pockels .. *Lehrbuch der Kristallogoptick*, Teubner, 1906.
7. Pancharatnam .. *Proc. Ind. Acad. Sci.*, 1956, **44 A**, 247.