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Conical refraction in naphthalene crystals*

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1. Introduction

The phenomena of internal and external conical refraction in biaxial crystals predicted by Sir William Hamilton and observed by Humphrey Lloyd are amongst the most beautiful and striking effects arising in crystal optics. Following Lloyd's original experiments, these phenomena are usually exhibited with aragonite, a polished plate of this crystal suitably mounted between apertures and a viewing lens being employed for the purpose. The angles of internal and external conical refraction in aragonite are however small, ($\chi = 1^\circ 52'$ and $\psi = 1^\circ 42'$ respectively), and the use of other crystals, e.g., tartaric acid with $\chi = 3^\circ 54'$ and $\psi = 3^\circ 58'$, and of sulphur for which $\chi = 7^\circ 11'$ and $\psi = 7^\circ 33'$ has therefore been sometimes suggested. It may be pointed out, however, that organic crystals of the aromatic class are specially suitable for the purpose. Naphthalene, in particular, exhibits birefringence in an exceptional degree, having as its principal indices 1·525, 1·722 and 1·945 respectively for $\lambda = 5461$ Å and its conical angles ($\chi = 13^\circ 44'$ and $\psi = 13^\circ 51'$) are enormously larger than in aragonite. Large single crystals of naphthalene can easily be prepared (Hilmi-Benel, 1940; Nedungadi, 1941), and the substance is thus well suited for exhibiting the optical characters of biaxial crystals and especially conical refraction in a striking way.

It may be remarked that apart from the purely geometrical aspects of optical theory illustrated by the Hamilton-Lloyd experiments, certain physical aspects of the propagation of light in biaxial crystals arising in conical refraction are of great interest. One of these is the enormous concentration of energy which occurs along the axis of single-ray velocity within the crystal and along the axis of the cone of external refraction outside it (Raman 1921; Raman and Tamma 1922). The converse phenomenon associated with internal conical refraction has long been known and is referred to in the literature as the Poggendorff dark circle. This was explained by Voigt (1905) as due to the attenuation of the energy of the incident pencil which occurs in single-wave propagation within the crystal. Both of these phenomena are well shown by naphthalene and in such manner as to bring out clearly their theoretical significance.

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Conical refraction is often studied by viewing an illuminated pin-hole in focus through the crystal plate with a microscope or magnifying lens. It is generally supposed that what is then seen is internal conical refraction. That this is not quite correct was long ago pointed out (Raman, *loc. cit.*), but the matter was not then adequately discussed. Since the illuminated pin-hole is usually held close to the crystal and is backed by an extended source of light, the beam of light entering it is not restricted to any particular direction, and the effect observed is not therefore ascribable to internal conical refraction. Neither would it be altogether correct to ascribe it to external conical refraction; for, though with the pin-hole close to the crystal, a cone of light is incident on its first surface, no aperture limits the exit of the light from the second surface as in the Lloyd experiment. The focussed image of an illuminated pin-hole as seen in the microscope through the plate of crystal is formed by the entire bundle of rays issuing from the pin-hole and passing through the crystal and is thus a phenomenon distinct from either internal or external conical refraction, though related to both. As will be shown in this paper, the form of this image is determined by the curvature properties of the wave-surface in the crystal in the vicinity of the conical points. It is specially worthy of remark that when the microscope is focussed on the second surface of the crystal and not on the illuminated pin-hole, we see in the field of view an illuminated picture of the two sheets of the wave-surface, the conical point where they meet appearing as an intensely luminous centre, and the circle of contact with the tangent plane appearing as a dark ring (figure 7 in plate II).

2. Preparation of the specimen

A clear block of naphthalene can be grown by slow crystallization from a melt. Pure naphthalene redistilled several times is collected in a pyrex glass tube of about half-an-inch diameter, with its lower end drawn out tapering to a sharp point. The tube is suspended in a vertical furnace kept at a temperature from 10° to 15° C above the melting point of naphthalene and gradually lowered out of it automatically by clock-work mechanism. Crystallisation starts at the tapering end of the tube and develops upwards. By proper control of the temperature of the furnace and of the rate of lowering of the tube, it is possible to get clear flawless blocks of the single crystal of any desired length. It is removed from the container by momentarily heating the walls of the glass tube to a high temperature; the portion of the crystal in contact with it then melts and the crystal slips out.

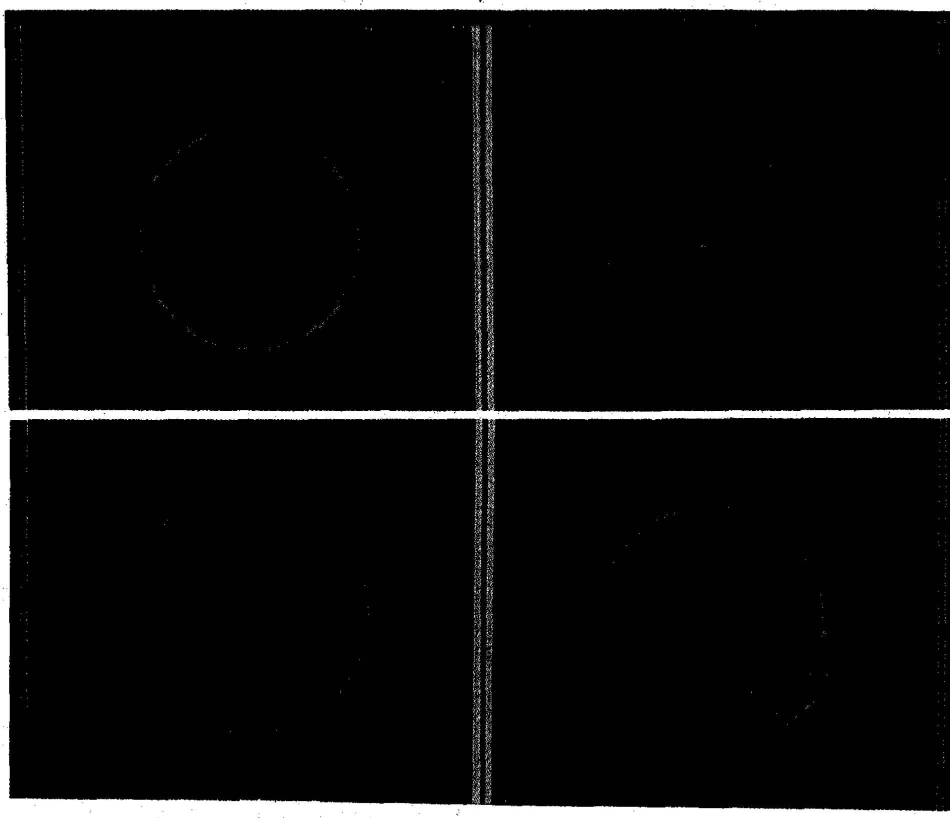
As the crystal blocks prepared in this way do not possess any natural faces, advantage is taken of the fact that the axes of the optical and magnetic ellipsoids of the crystal roughly coincide to determine their orientation. The three magnetic axes of the crystal block may be determined by marking its preferred orientations in a strong magnetic field with different modes of suspension. The crystal block may be then cut with faces making any desired angle with these axes. To exhibit conical refraction, the naphthalene block should have its faces approximately normal to one of the primary optic axes, these being inclined at 42° to the acute bisectrix of the angle between them. For mounting the cut crystal, a flat surface is first ground and then quickly pressed on to a microscope cover slip kept at a temperature of about 40° above the melting point of naphthalene. The crystal face melts and wets the glass plate and immediately cools, thus resolidifying the melted layer as part of the single crystal in addition to making good optical contact with the glass. The second face of the plate is then treated in the same way. An alternative method of mounting is to grind the surfaces of the block smooth on a ground glass plate and then to polish them by rubbing quickly on a soft cloth stretched over a glass plate and moistened with a drop or two of xylene. Thin microscope cover clips may then be stuck on the faces with Canada balsam. The mounted crystal may be conveniently fixed on a disc of aluminium having a central opening.

3. Method of observation and results

The angles of conical refraction in naphthalene are so large that with a fairly thick piece, the phenomena can be seen directly with the simplest possible arrangements. For a critical study of the effects, however, and especially for securing satisfactory photographs, it is convenient to use a microscope with a revolving and centering stage and a Federov universal stage attachment on which the crystal plate is placed so that it can be tilted and set with its optic axis accurately parallel to the axis of the microscope. A low-power objective and a high-power ocular should be employed so that, for the same effective magnification, the largest working distance between the upper surface of the crystal and the objective of the microscope can be secured. For observing conical refraction under the microscope, a relatively thin plate of naphthalene (say two to three millimetres thick) is quite suitable. It is easy with these arrangements to photograph the cone of external conical refraction and the cylinder of internal conical refraction outside the crystal in the manner of the Hamilton-Lloyd experiments. It is also possible to examine the relationships between these effects and the nature of the optical images obtained when a point-source of light is viewed through the crystal either in or out of focus.

Figures 1–4 in plate I and figures 5–8 in plate II reproduce a series of photographs obtained with the microscope camera attachment to illustrate the phenomena of conical refraction in naphthalene, the monochromatic green light $\lambda 5461$ Å of the mercury arc being employed to avoid all disturbances due to chromatic aberration or dispersion. Figure 1 shows the hollow cone of external conical refraction as seen above the crystal; to photograph this, both the upper and lower surfaces of the crystal are covered up except for small apertures situated at the ends of the axis of single-ray velocity, the lower aperture being illuminated by a convergent pencil of light. Figure 2 shows the cylinder of internal conical refraction seen outside the crystal when a parallel beam of light is incident in the direction of the optic axis on the lower face of the crystal; to observe this, the lower face is covered by a screen with a small aperture and the second face is left uncovered. Figures 3 and 4 in plate I and figures 5, 6, 7 and 8 in plate II reproduce a consecutive series of photographs of a point source of light held close to the first surface and viewed through the crystal; figure 3 is the image seen in focus, while the other photographs in the series are ultra-focal images obtained when the microscope objective is gradually drawn away from the crystal. As already mentioned in the introduction, figure 7 is the ultra-focal image of the point source of light obtained when the microscope is focussed on the second surface of the crystal. Figure 8 is the ultra-focal image obtained when the microscope is still further drawn up. In obtaining this series of six pictures, the source of light was an extremely fine hole (1 μ in diameter) in an aluminium foil covering the lower surface on the crystal and illuminated by a convergent beam of light, while the second face of the crystal was left uncovered. The extreme sharpness of the circular ring seen in figure 3 is particularly significant. *It is*

(1) (2)



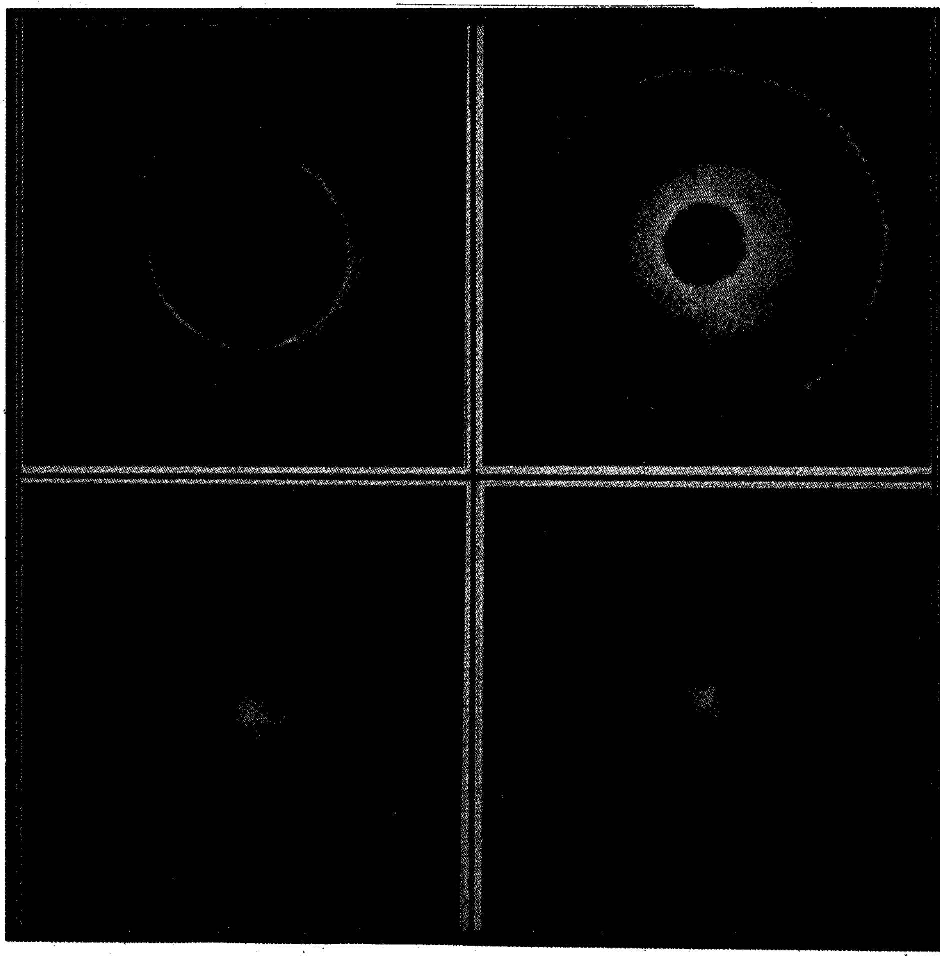
(3) (4)

Figures 1–4. Illustrating conical refraction in a naphthalene crystal.

Plate I

(5) (6)

(7) (8)



Figures 5–8. Illustrating conical refraction in a naphthalene crystal.

Plate II

noteworthy also that the so-called Poggendorff dark circle does not appear in the focal image of the point source and develops only in the ultra-focal images. The extremely bright point seen at the centre in figure 7 (as also in figure 8 and very feebly in figure 2) is a noteworthy feature. This bright point in the ultra-focal image coincides with the end of the axis of single-ray velocity meeting the second surface of the crystal. This is shown by the fact that the second aperture for observing the cone of external conical refraction above the crystal (figure 1) has to be placed exactly at the same point so as to admit the light passing through the crystal. It is evident from the series of pictures *that the axis of single-ray velocity and the conical point on the wave-surface are loci of intense concentration of energy* *within the crystal, while the circle of contact where the wave-surface touches the tangent plane is a locus of vanishingly small energy.*

4. Image formation with a biaxial crystal

It is well known (Stokes 1877; Walker 1904) that the image of a point source of light seen through a crystalline plate exhibits astigmatism, being drawn out into a line perpendicular to the plane of principal curvature. For a biaxial crystal there are, in general, no fewer than four distinct positions of best focus determined by the orientation of the plate and by the principal radii of curvature of each of the two sheets of the wave-surface. In our present problem, we are concerned with the curvature of the wave-surface in the vicinity of the conical point and especially along the circle of contact with the tangent plane. At the conical point, one of the principal radii of curvature for each of the two sheets of the wave-surface vanishes, while the other two radii are

$$\rho_1 = b \text{ and } \rho_2 = (a^2 + c^2 - b^2)^{3/2}/ac.$$

At points along the circle of contact, one of the principal radii of curvature of each of the two sheets becomes infinite, while the other radius of curvature is

$$\rho = b\cdot(a^2 - r^2)(c^2 - r^2)/(a^2 - b^2)(c^2 - b^2),$$

r being the length of the line joining the origin with any specified point on the circle of contact. At the two points where this circle cuts the circular and elliptic sections of the wave-surface respectively, the radii of curvature are

$$\rho'_1 = b \text{ and } \rho'_2 = b^3/a^2c^2.$$

In the case of naphthalene, b^2 and ac are practically identical, as is readily seen from the numerical values of the principal refractive indices. As a consequence of this, also, the angles of internal and external conical refraction are practically identical. Hence, while one of the principal radii of curvature of the wave-surface is infinite along the circle of contact, the other radius of curvature is practically constant and equal to b at all points on the circle and changes only slowly as we move away from the circle along the wave-surface either towards or away from the conical point. Accordingly, the astigmatism of the rays emerging from the plate results in an exceptionally simple form of the image, namely a sharply focussed circular ring having the same diameter as the circle in which the wave-surface makes contact with the second surface of the crystal. As the microscope objective is drawn away from the crystal, the ultra-focal image necessarily alters continually. The rays reaching the upper surface of the crystal within the circle of contact bend inwards, while those outside the circle bend outwards, a gap appearing between them owing to the vanishing intensity at points along the circle. The rays that bend inwards appear to gain rapidly in intensity as they approach the centre of the field; the latter appears as a luminous point from which the rays appear to diverge, when the focal plane of the microscope coincides with the upper surface of the crystal.

The radiations from the point source entering the crystal may be regarded as an assembly of plane waves with coherent phase-relationships crossing each other at that point. Entering the crystal, their directions of travel are altered, and the resultant distribution of the energy stream within the crystal is determined by their superposition. Along the axis of single-ray velocity, the lines of energy flow of numerous sets of plane waves coincide and the density of the energy flow is therefore a maximum on this line. On the other hand, along the so-called cone of inner conical refraction, the energy-flow of a single set of plane-waves is divided up and the energy flow is therefore a minimum. Since the disturbance emerging from the crystal is determined by the superposition of the plane waves refracted out from it, the energy flow outside would be closely related to the special character of the energy flow within the crystal. Actually, the bright spot at the centre of the field may be traced for a great distance outside the crystal. The bright spot is, in effect, a spectral image of the original point source, its position varying with the wavelength of the light used (Raman and Tamma, *loc. cit.*).

5. Summary

The angles of internal and external conical refraction for naphthalene are exceptionally large (both about 13° 45′), and the substance is therefore exceptionally well suited for exhibiting these phenomena as well as for a critical study of the same. A series of eight photographs is reproduced with the paper and is discussed in detail. The following noteworthy effects are exhibited by the crystal. The so-called Poggendorff dark circle appears only in internal conical refraction and is an ultra-focal phenomenon, disappearing when the image of a point source of light seen in exact focus through the crystal plate, the image being then a single circular ring which is extremely sharp. In external conical refraction we have an effect converse to the Poggendorff phenomenon, viz., a concentration of energy at the conical point of the wave-surface and therefore also along the axis of single-ray velocity. When the microscope is focussed on the second surface of the crystal and not on the source of light, the field of view exhibits a picture of the wave-surface in two sheets, their intersection appearing as an intensely luminous point and the tangent plane to the surface as a dark ring.

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