

1.6. CLASSICAL LINEAR CRYSTAL OPTICS

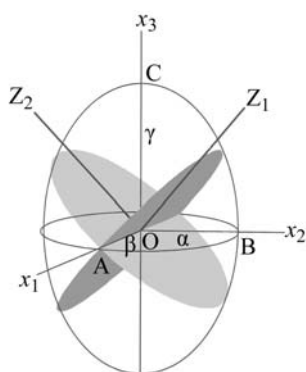


Fig. 1.6.3.3. Biaxial indicatrix, showing the two optic axes and corresponding circular cross sections.

ellipsoid, *i.e.* one in which all three semi-axes are different from one another (Fig. 1.6.3.3).

It is conventional to label the three axes according to the size of the refractive index by $n_\gamma > n_\beta > n_\alpha$ (or simply $\gamma > \beta > \alpha$). In such an ellipsoid, there are always two special directions lying in the γ - α plane, known as the *optic axial plane*, and perpendicular to which there are circular cross sections (shown shaded) of radius β . Thus these two directions are optic axes down which the crystal appears to be optically isotropic, with a measured refractive index β for light of any polarization. For this reason, crystals with this type of indicatrix are known as *biaxial*. When the angle between the optic axes, denoted conventionally as $2V$, is acute about the γ axis, the crystal is *positive biaxial*, and when it is acute about α the crystal is *negative biaxial*. Note that as $2V$ becomes smaller, the biaxial indicatrix becomes closer to a uniaxial indicatrix (positive or negative). In all general directions the crystal is optically anisotropic. Thus, for light along x_3 , the measured refractive indices will be α and β for light polarized along x_1 and x_2 , respectively; for light along x_2 , α and γ are measured for light polarized along x_1 and x_3 , respectively; and along x_1 , β and γ are measured for light polarized along x_2 and x_3 , respectively. There are therefore three different linear birefringences to measure: γ - β , β - α and γ - α .

The different indicatrices are oriented in the crystal according to symmetry considerations (Table 1.6.3.1), and so their observation can form valuable and reliable indicators of the crystal system.

1.6.3.3. The dielectric impermeability tensor

It has been seen how the refractive indices can be described in a crystal in terms of an ellipsoid, known as the indicatrix. Thus for orthogonal axes chosen to coincide with the ellipsoid axes, one can write

$$\frac{x_1^2}{n_1^2} + \frac{x_2^2}{n_2^2} + \frac{x_3^2}{n_3^2} = 1, \quad (1.6.3.22)$$

where $n_1 = (\epsilon_{11})^{1/2}$, $n_2 = (\epsilon_{22})^{1/2}$ and $n_3 = (\epsilon_{33})^{1/2}$. One can write this equation alternatively as

$$\eta_{11}x_1^2 + \eta_{22}x_2^2 + \eta_{33}x_3^2 = 1, \quad (1.6.3.23)$$

where the $\eta_{ii} = 1/\epsilon_{ii}$ are the *relative dielectric impermeabilities*. For the indicatrix in any general orientation with respect to the coordinate axes

$$\eta_{11}x_1^2 + \eta_{22}x_2^2 + \eta_{33}x_3^2 + 2\eta_{12}x_1x_2 + 2\eta_{23}x_2x_3 + 2\eta_{31}x_3x_1 = 1. \quad (1.6.3.24)$$

Thus the dielectric impermeability tensor is described by a second-rank tensor, related inversely to the dielectric tensor.

1.6.4. Practical observation of crystals

1.6.4.1. The polarizing microscope

There are countless applications of polarizing microscopy. One of the largest fields of use is in mineralogy and petrology, where the requirement is to identify naturally occurring minerals, the optical properties of which have already been determined elsewhere. Medical applications of a similar sort exist, for instance in the identification of the minerals present in bladder or kidney stones. The chemist or materials scientist who has synthesised a crystalline material may also wish to identify it from known properties, or it may be a new substance that needs to be described. For other purposes it might, for example, be necessary to determine the orientation (relative to crystallographic axes) of mineral specimens, *e.g.* in the cutting of synthetic corundum for the manufacture of watch jewels. This section explains the point of view of an observer who wishes to record and measure optical properties, for whatever reason. Although much of what follows is discussed in terms of mineral crystals, it is equally valid for crystals in general, whether organic or inorganic.

The polarizing microscope incorporates five major features not found in ordinary microscopes. These are:

(i) A *polarizer*, normally a sheet of Polaroid, which is part of the microscope substage assembly. This produces plane-polarized light before the light reaches the specimen. In some microscopes, the polarizer can be rotated, though applications of this technique are rare. In the commonly used petrological microscope, the vibration direction of the polarizer is set in what is called the E–W direction, that is, as the user of the microscope sees the field of view, the vibration direction is from side to side.

(ii) An extra, high-power *condenser* situated in the substage immediately below the specimen. The condenser is switched in and out of the optical path as required.

(iii) A *rotating stage*, circular in plan and graduated in degrees. For a number of purposes, specimens can be rotated through known angles.

(iv) An *analyser*, a second polarizing device, situated in the microscope tube above the specimen. Its vibration direction is set at right angles to that of the polarizer, *i.e.* usually N–S. Like the condenser, this can be inserted into the optical path as needed.

(v) A *Bertrand lens*, also in the microscope tube and insertable as required, which has the function of transferring images from the back (upper) focal plane of the objective to the front (lower) focal plane of the eyepiece. The Bertrand lens and the extra substage condenser are used together to convert the microscope from the *orthoscopic* to the *conoscopic* configuration (see later).

In addition, polarizing microscopes have slotted tubes that allow the insertion of a variety of extra devices generally known as accessory plates. Most common amongst these are the sensitive-tint plate (or 1λ plate) and the quartz wedge.

Objective lenses of various magnifying powers are mounted in a rotating turret. Apart from magnification (typically $ca \times 5$ for low power, and $\times 40$ or more for high power), the *numerical aperture* (n.a.) of a lens is an important feature. This is defined as the diameter divided by the focal length. This is a measure of the angle of the cone of light that can enter the objective. In the

Table 1.6.3.1. Symmetry constraints on the optical indicatrix

Crystal system	Indicatrix	Orientation constraints
Cubic	Isotropic (sphere)	None
Tetragonal Trigonal Hexagonal	Uniaxial	Circular cross section perpendicular to c
Orthorhombic	Biaxial	All indicatrix axes aligned along a , b and c
Monoclinic	Biaxial	One indicatrix axis aligned along b (second setting)
Triclinic	Biaxial	None

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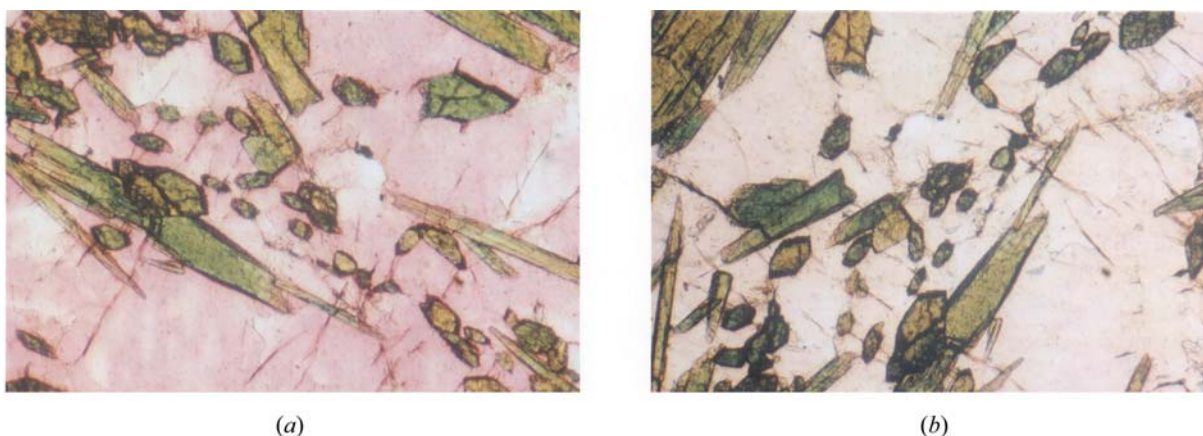


Fig. 1.6.4.1. A thin section of a rock containing the minerals aegirine (elongated crystals) and eudyalite (the matrix) viewed in plane-polarized light in two positions at right angles to each other [(a) and (b)]. Aegirine shows pleochroism from grass-green to yellow-green (compare specific crystals in the two photographs). Eudyalite shows pleochroism from pink to almost colourless.

conoscopic use of the microscope (see Section 1.6.4.11), this angle is required to be as large as possible so that the properties of rays travelling through the crystal in a variety of directions can be observed. Numerical apertures of more than *ca* 0.9 can not be achieved with 'dry' objectives, but higher values are obtained by inserting a drop of immersion oil between the specimen and the lens.

Eyepieces in polarizing microscopes are set in a short tube, at the lower end of which is mounted a set of cross wires, which lie in the front focal plane of the lens. When the microscope is properly focused, a real image of the specimen, created by the objective, is made to coincide with the cross wires. The cross wires are conventionally oriented vertically (N-S) and horizontally (E-W) in the field of view, and coincide with the vibration directions of the polarizer and analyser.

1.6.4.2. Specimen preparation

Specimens for examination with the polarizing microscope are usually of two different sorts: collections of small crystals or grains (and individual crystals), and thin sections cut from larger solid samples. In the first category, the material is often crushed to a fine sand, scattered on a microscope slide, a drop of immersion oil is applied, and a cover slip is placed on top. For special applications, an individual crystal may, for example, be mounted on the end of a glass fibre, and similarly examined under immersion oil. The thin-section technique is more widely used in petrology. Here a rock sample is cut into a section of standard thickness (0.03 mm) and mounted on a glass slide, using a resinous mounting material, formerly Canada Balsam but now synthetic. Small single crystals can also be used by mounting them on a spindle stage, or similar device, which allows one to orient the crystal in the microscope.

1.6.4.3. The indicatrix as an aid to practical microscopy

It is convenient for the microscopist to imagine the indicatrix sitting inside the crystal under observation. The indicatrix ellipsoid is fixed with respect to the crystallographic axes, according to different crystal systems (Table 1.6.3.1). The radial dimensions of the ellipsoid, in whatever direction, are a measure of the refractive index of a ray vibrating in that direction. Hence, because ray directions are approximately parallel to the microscope axis, a planar section across the indicatrix at right angles to the axis approximately contains the vibration directions of the ray. Such a section is in general an ellipse, the magnitudes of the major and minor axes of which represent the relative refractive indices, and the directions of which represent the vibration directions of the two transmitted rays. In all cases, the ray with the greater refractive index is known as the *slow ray* and the other as the *fast ray*. In uniaxial crystals the ordinary ray may be

slow or fast, depending on the optic sign. This mental image of the indicatrix acts as the microscopist's guide in the practical operations that follow.

1.6.4.4. Vibration directions

For the microscopist, the overriding feature of the behaviour of light transmitted through crystals is double refraction. Many of the observations that can be made with the polarizing microscope depend on the ability to distinguish the *individual* properties of the two rays. In general, they have different refractive indices, and in coloured crystals they may show different absorptions (*i.e.* the rays show different colours after transmission, the phenomenon known as *pleochroism* – see Fig. 1.6.4.1).

When the microscope is used for observations in *plane-polarized* light, the polarizer is inserted into the optical path but the analyser is not. The light before entering the crystal is polarized E-W. On entry, in the general case where the vibration directions of the crystal happen to lie in a random orientation relative to the vibration direction of the polarizer, the light is transmitted as two rays resolved into the two vibration directions of the specimen. These reach the observer's eye unchanged, and the effects observed are an average of what each ray would individually demonstrate.

Two cases can be used to illustrate this fundamental concept. The mineral calcite (CaCO_3) is always given as an example of extreme double refraction. In sections cut parallel to the *c* crystallographic axis, the two rays have refractive indices of 1.486 and 1.658, and this is the maximum difference they can show. Crystals in other orientations always show a smaller difference. The numerical difference between the two refractive indices, when we have identified the maximum difference, is what is quoted as the *birefringence* of the mineral, and for calcite it is unusually large (0.172) by the standards of other commonly occurring minerals (*cf.* quartz = 0.009). Birefringence is an important quantitative optical property, because in thin sections it is easily measurable. It is also useful to refer to the birefringence of individual grains, but this should not be confused with the use of the same term, based on maximum birefringence, as a diagnostic optical property of the material concerned.

Consider a calcite crystal lying with its *c* axis in the plane of the microscope slide. The vibration directions are parallel and at right angles to the crystallographic axis. On rotation of the stage, the *c* axis may be brought into parallelism with the vibration direction of the polarizer, in which case all the light is transmitted using only one of the vibration directions, which in this case has a refractive index of 1.486. On turning the stage through a right angle, all the effects seen are completely due to the other ray, with a refractive index of 1.658. The mounting medium in standard thin-section preparations has a refractive index of *ca* 1.54.

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Hence, in the first position there is quite a large contrast between the refractive index of the crystal grain and the mounting medium, and in the other it is considerably smaller. Calcite is usually a highly transparent and virtually colourless substance, and whether the observer can even see the crystals depends on the refractive index difference between it and the mounting medium. In the first case, it is highly visible and in the second case it is harder to see. It is of course the roughness of the surface of the thin section, or the angularities of the form of the crushed crystal, which create small refractions, if there is a refractive-index contrast between it and the mounting medium, and thus make the crystal visible. The general property is known as *relief*. High relief means highly visible in this sense. Calcite shows such extreme changes in relief on rotation of the stage that the phenomenon is known as *twinkling*. Relief is not a measurable property, but to the experienced microscopist it is a subtle and useful guide to identification.

As a second example consider the mineral biotite, which shows extremely strong pleochroism. Biotite is for practical purposes a hexagonal mineral having a platy form dominated by a strong cleavage on (001.1). In thin sections, biotite crystals cut across the cleavage, *i.e.* parallel to the direction [00.1], are typically very dark brown in plane-polarized light when the cleavage lies parallel to the polarizer direction. When turned through 90° , the same crystals are only a very pale brown, pale yellow, or almost colourless. The colour contrast, a consequence of the different absorbing properties of the two rays, is striking. Examples of pleochroism are shown in Fig. 1.6.4.1.

From these examples it should be clear that the different rays show different things. However, the microscopist needs to be certain that the observations made refer exactly to one ray or the other. When the analyser is inserted into the optical path, most crystals (excluding those of the cubic system, and crystallites lying in special orientations) will show *polarization colours*, a consequence of the interference between the two rays once they are recombined vibrating in the analyser plane. Details will be discussed below, but for the present purpose it is the *extinction position* that is of importance. On rotation, a crystal viewed with both analyser and polarizer in position (a configuration known as *crossed Nicols*, after the Nicol prism, the forerunner of Polaroid) will transmit no light in two positions at right angles to each other. This happens when the vibration direction of a transmitted ray coincides with that of the polarizer. In this case, all the light transmitted through the crystal vibrates in a single direction, at right angles to the vibration direction of the analyser, and it is consequently unable to reach the observer. Thus, for all observations of the effects of a particular ray, crossing the polars (inserting the analyser), turning the stage to an extinction position, and then uncrossing the polars (removing the analyser) leaves the specimen showing the pure effects of whichever ray vibrates in the same direction as the polarizer. Much that follows depends on this simple operation.

1.6.4.5. Measuring refractive indices

Refractive indices measured carefully can be extremely useful aids in crystal identification, as well as being of importance as physical properties of interest. Apart from distinguishing crystalline species that may look similar under the microscope but have widely different refractive indices, the precise composition of crystalline materials belonging to important solid-solution series (*e.g.* the plagioclase feldspars or the olivines, in geological applications) can also be determined.

The direct measurement of refractive indices is often made by the examination of crystal grains mounted in an immersion oil, using the so-called *Becke line* test. This is observed in plane-polarized light with the substage diaphragm closed down to produce a narrow beam of essentially parallel rays. A medium-

power lens is usually suitable. When the oil has a refractive index different from that of the crystal, the Becke line appears as a bright rim of light around the edge of the crystal. However, as the microscope tube is racked up and down slightly (*i.e.* the position of focus is changed), the Becke line moves in or out relative to the crystal edge. As the position of focus is *lowered* the line moves towards the medium with the *lower* refractive index, *e.g.* if the oil has a higher refractive index than the crystal, lowering the focus (racking down) causes the Becke line to contract into the crystal. It is of course important to set the specimen in an extinction position before making the observation. If the oil has a refractive index between those of the two rays passing through the crystal, then the behaviour of the Becke line will reverse if the crystal is rotated to the other extinction position. In cases where there is a very large contrast between the crystal and the surrounding medium, a line as such may not be observed, but rather the specimen may appear to glow with concentrated light. The equivalent of Becke line movement is then the expansion or contraction of the light pool with changing focus.

The general objective of the observations is eventually to achieve an exact match between the immersion medium and the crystal. This is done by choosing different oils, or mixtures of oils, in sequence, the refractive indices of which are measured by a suitable refractometer. Ideally monochromatic or near-monochromatic light (*e.g.* the Na doublet with $\lambda = ca\ 590\text{ nm}$) is used, in which case the Becke line simply disappears when the crystal and the oil match. In white light however, because of dispersion by the oil, a match is shown by the presence of *two* faint Becke lines, one red and one greenish blue, which migrate in opposite directions as the focus is changed.

The general strategy of refractive-index determination is perfectly straightforward for cubic crystals, but requires the separate determination of values of n_e and n_o in uniaxial crystals, and n_α , n_β and n_γ in biaxial crystals. The most general case is that of the biaxial crystal. If a large number of crystal grains in the mount are examined, a number of cases may be distinguished.

(i) All grains have both refractive indices higher than the oil. The oil has a refractive index below n_α .

(ii) All grains have both refractive indices below that of the oil. The oil has a refractive index above n_γ .

(iii) Some grains have both refractive indices above that of the oil, while others have one above and one below. The oil has a refractive index between n_α and n_β .

(iv) Conversely, some grains have both refractive indices below the oil, while others have one above and one below. The oil has a refractive index between n_β and n_γ .

Uniaxial crystals present a simpler series of cases, in which the crystal may show both refractive indices higher than the oil (*i.e.* the refractive index of the oil is less than that of the fast ray), both lower than the oil, or one higher and one lower.

Systematic application of the above techniques leads to the determination of all the refractive indices required, and constitutes one of the most powerful methods of crystal identification or description. However, it is useful to make an additional check using the fact that, in anisotropic crystals, any specimen that fails to show polarization colours between crossed polars (*i.e.* remains dark in all stage positions) must lie with an optic axis parallel to the microscope axis. Such a crystal directly shows n_o (uniaxial crystals) or n_β (biaxial crystals). Furthermore, crystal grains showing maximum birefringence (see below) can be checked to see if they give a centred flash figure (see later), and if they do, their two vibration directions will show n_e and n_o , or n_α and n_γ , that is, the optic axis or axes lie in the plane of the microscope slide.

In larger crystalline specimens, several other techniques are available for measuring the refractive index. Perhaps the simplest and also the most convenient is to cut the crystal into a prism, and use minimum-deviation measurements on a spectrometer table. In addition, large plates can be inserted directly into a commer-

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cial refractometer, in order to measure the refractive index directly.

1.6.4.6. Determination of linear birefringence

The numerical determination of linear birefringence gives less information than a full set of refractive-index measurements, but is nevertheless highly useful in crystal identification, particularly in mineralogical and petrological applications where the thin section is the norm. It is also a most sensitive indicator of changes in the crystal structure at a phase transformation or as a function of temperature, pressure *etc.* Refractive-index determination is tedious, but birefringence determination is quick and easy.

Double refraction generates polarization colours when crystals are viewed between crossed polars, except where the crystal is by chance in an extinction position, or cut normal to an optic axis. The colours result from the interference of the two transmitted rays when they are combined into one vibration direction in the analyser. Polarization colours are best observed with the substage diaphragm moderately closed down, so that the transmitted light corresponds to a roughly parallel bundle of rays (if the diaphragm is wide open, and the supplementary condenser is inserted, the resultant rays are far from parallel, and the polarization colours will immediately be seen to degrade in the direction of whitening).

Considering a section showing two refractive indices, n_1 and n_2 , the time difference required for a ray to traverse the section is

$$t = \frac{nz}{c}, \quad (1.6.4.1)$$

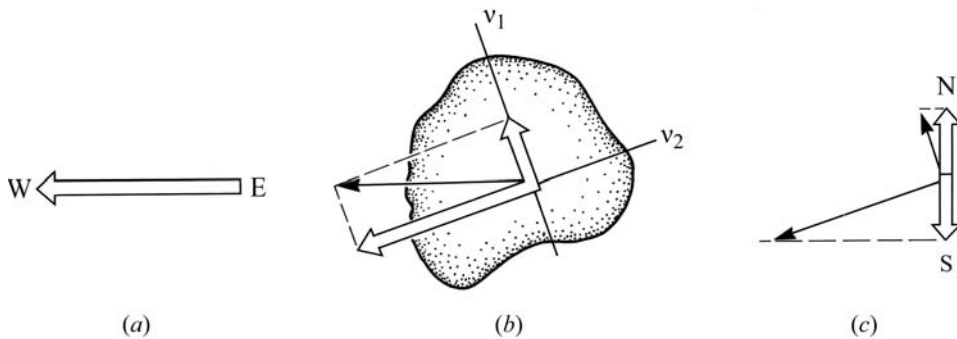


Fig. 1.6.4.2. Successive sections across the optical path of the microscope. (a) Above the polarizer, beneath the crystal. Polarized light vibrates E-W. (b) Within the crystal. Transmitted light is resolved into the two vibration directions of the crystal (v_1 and v_2). (c) Above the analyser. The two transmitted rays are resolved into the N-S direction. Amplitudes are shown, but phase difference is not.

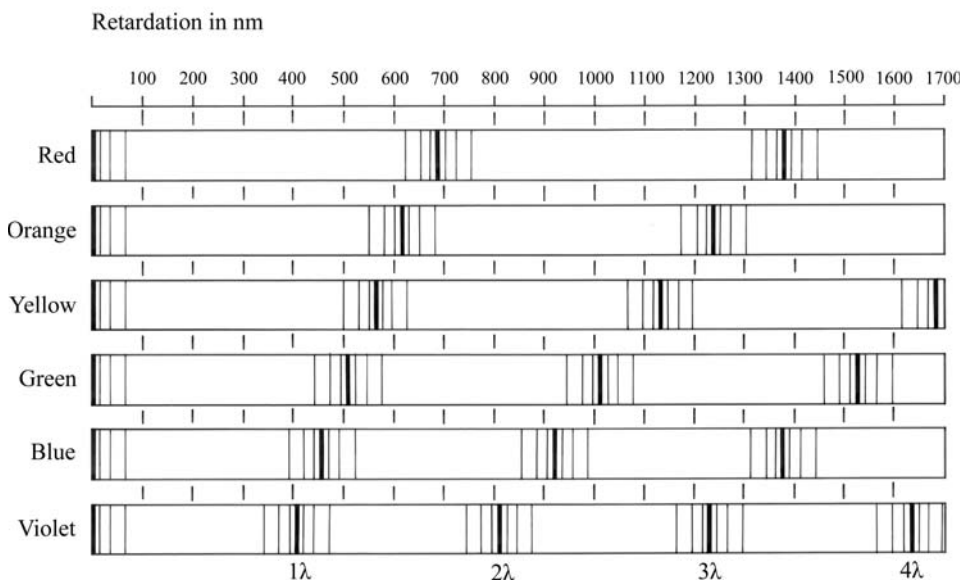


Fig. 1.6.4.3. Plot of retardation, R , versus wavelength, showing how polarization colours are formed.

where z is the thickness of the section. The time difference between the two rays is then

$$t_1 - t_2 = \frac{(n_1 - n_2)z}{c}, \quad (1.6.4.2)$$

which is referred to as the *retardation*. Multiplication by c gives the *relative retardation* or *optical path difference*, R , where

$$R = c(t_1 - t_2) = (n_1 - n_2)z. \quad (1.6.4.3)$$

R is usually expressed in nm (formerly $\mu\mu$).

The possibilities of interference clearly depend on R , but also on wavelength. For complete destructive interference, because of the way the transmitted rays are resolved into the vibration direction of the analyser (see Fig. 1.6.4.2), R must either be zero (as in cubic crystals, and sections normal to optic axes in anisotropic crystals) or a whole number of wavelengths. Thus as R changes, either with thickness, orientation of the crystals or with variation in birefringence in different substances, a variety of colours are produced, essentially formed from white light with various wavelengths *subtracted*. There is a good discussion of this point in Wahlstrom (1959).

Fig. 1.6.4.3 shows the effect of increasing R on a variety of visible-light wavelengths. When R is zero, no light is transmitted since all wavelengths show total destructive interference. As R increases a little, all wavelengths continue to show interference, and the polarization colours are essentially greys, which decrease in darkness until the middle of the first order where the grey is very pale, almost white. Most wavelengths at this stage are showing relatively strong transmission.

With increasing R , the region is reached where the shortest wavelengths of the visible light spectrum (violet) are beginning to approach a phase difference of 1λ . The transmitted light then takes on first a yellow tinge and then bright orange, as violet light (at $R \approx 400$ nm) and then blue are completely removed. Next, the removal of green light ($R \approx 500$ nm) results in the transmitted light being red and at $R \approx 560$ nm the top of the *first-order* colours is reached with the removal of yellow light. The resultant polarization colour is a distinctive magenta colour known as sensitive tint. The accessory plate known as the 'sensitive-tint' or ' 1λ ' plate is made to have $R = 560$ nm. Its use will be explained below, but meanwhile note that the polarization colour is so-called because with only very slight changes in R it becomes obviously red (falling R) or blue (rising R).

Between $R = 560$ nm and $R = 1120$ nm, the second-order colours are produced, and are similar in appearance to the colours of the rainbow (blue, green, yellow, orange and red in sequence), as orange, red, violet, blue, and green are successively cut out. The *third-order* colours ($R = 1120$ – 1680 nm) are essentially a repeat of the second-order, but there is a subtle change of quality about them, as they take on slightly garish hues compared with rainbow colours (the red at the top of the order has for example a distinct air of 'shocking pink' or even lipstick about it). This effect is a consequence of the increasing chances that two wavelengths will be cut out simulta-

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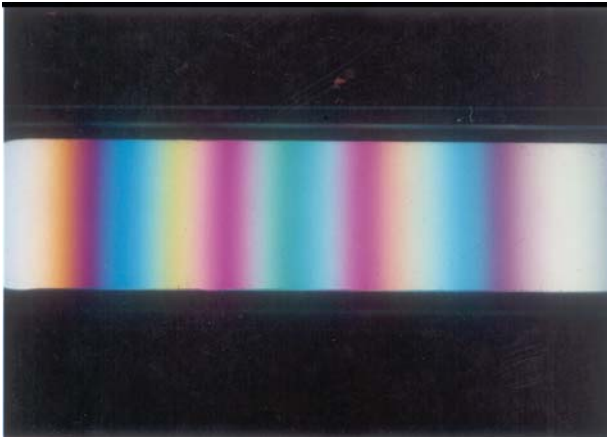


Fig. 1.6.4.4. A quartz wedge between crossed Nicols. The thin end is on the left. The colours shown vary between about the middle of the first order up to high fifth order.

neously (see Fig. 1.6.4.3), *e.g.* while first-order red results from the removal of green, second-order red results from the removal of both violet and yellow.

With increasing R , the distinction of colours within each order becomes weaker as the number of wavelengths simultaneously removed increases. Colours are diluted towards grey or white, so that from the fifth order upwards the range is little more than an alternation of pale pinkish and pale greenish tints. Eventually, at higher orders the polarization colours become a more-or-less uniform dull white. Fig. 1.6.4.4 shows the colours produced by a quartz crystal cut into a wedge shape.

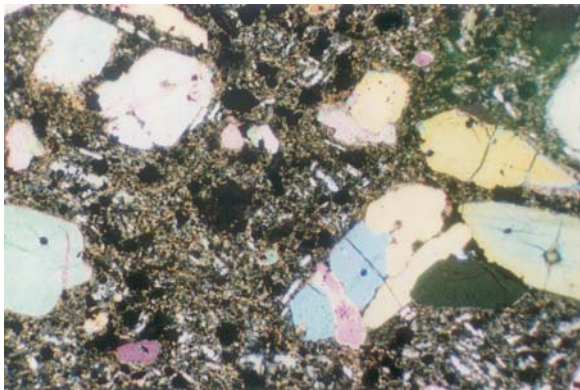
1.6.4.7. Identification of polarization colours

Birefringence can be determined quantitatively if the polarization colour can be correctly identified, and the section is of known thickness (see Fig. 1.6.4.6). It is of course necessary to distinguish between the birefringence of an individual grain, which will depend on orientation (see Fig. 1.6.4.5*a*), and the maximum birefringence (highest polarization colour) shown by the crystals concerned. It is the latter that is diagnostic, and in general it will be necessary to examine as many grains as possible to determine it.

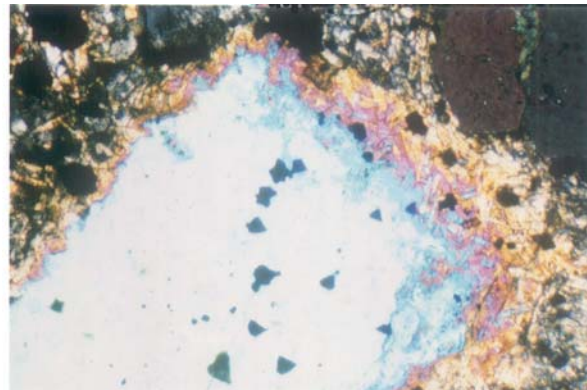
The grey colours of the lower part of the first-order colours are unique and immediately identifiable, but all other colours are at least superficially ambiguous. Even first-order white may be confused with a high-order white. There are essentially two methods of determining the order of an ambiguous colour, the first of which, fringe counting, is discussed here. Other methods depending on the use of the quartz wedge and sensitive-tint plate will be considered later.

1.6.4.8. Fringe counting

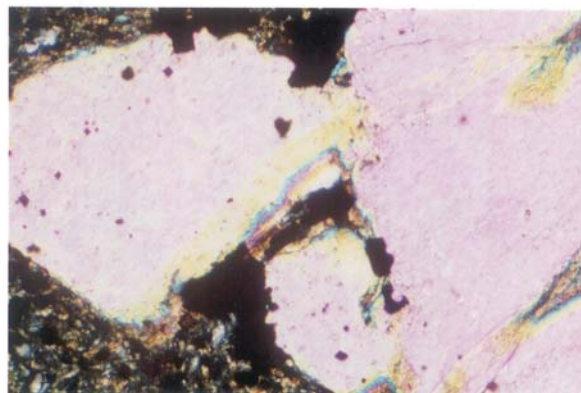
In a well made thin section, most crystals show a uniform polarization colour because of uniform orientation and thickness. Crystal edges, however, frequently taper off to near-zero thickness. In this case, the edge acts as a natural wedge of variable R and displays a series of fringes corresponding to the orders within the complete spectrum of interference colours, from first-order grey up to the main colour shown by the body of the crystal (Figs. 1.6.4.5*b* and *c*). Using a moderately high power objective, it is often easy to identify the individual bands and thus count up to the order concerned. The colour band red-sensitive tint-blue



(a)



(b)



(c)

Fig. 1.6.4.5. (a) Photomicrograph between crossed polars of a thin section of rock containing large olivine crystals set in a fine-grained matrix. The olivine crystals show a variety of polarization colours (first-order to high second-order) depending on their orientation. (b) Thin section between crossed polars of a rock containing a large pyroxene crystal, intergrown with other phases marginally. The edges of the crystal are wedge-like, and therefore thinner than the main body. The first-order white to orange colours make a prominent fringe on the right, grading inwards to sensitive tint and then to blue. The main body of the crystal is thus determined as second-order blue. (c) A similar section to that in (a) (centre of photograph) but here the second-order blue fringe is narrow and is succeeded by a second-order yellow fringe. Hence the main body of the crystal must be showing second-order pink.

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marking the first/second-order change is particularly easy to spot because of its generally dark colour. The method is also usable when the crystal grain has a sloping contact with a different crystal, and is especially easy when the latter has significantly lower birefringence. In this case, there are effectively two superimposed wedges (leading to some interesting addition or subtraction effects on R), but counting the fringes in ascending order will lead correctly to the identification of the polarization colour of the high-birefringence grain.

1.6.4.9. Fast and slow vibration directions

Before discussing other methods of identifying polarization colours, it is necessary to explain the use of the sensitive-tint plate and the quartz wedge. Both devices consist of a crystal mounted in an elongated holder that can be inserted into a slot in the microscope tube set at 45° to the vibration directions of the polarizer and analyser. The vibration directions of the plates themselves are normally oriented so that the slow ray vibrates NE–SW and the fast ray NW–SE.

The sensitive-tint (or 1λ) plate is made from a cleavage sheet of the white mica, muscovite, and has a thickness such that $R = 560$ nm. To determine fast and slow directions in an unknown specimen, the crystal grain is set to an extinction position and then rotated 45° in either direction. Thus its own vibration directions lie in diagonal positions, and when the tint plate is inserted, the vibration directions of plate and grain are parallel to each other. There are two possible cases depending whether the fast and slow directions coincide or not, *i.e.* slow vibration direction of plate parallel to the slow direction of the crystal (and fast parallel to fast) or slow direction of plate parallel to the fast direction of the crystal.

In the first case, the relative retardation is increased by 560 nm and the observed polarization colour jumps up the scale by one complete order as the plate is inserted. Thus, for example, first-order white ($R = 230$ nm) changes to second-order yellow/green ($R = 790$ nm), second-order blue changes to third-order blue *etc.*

When this effect is seen, it shows that the slow direction of the crystal lies NE–SW.

In the converse case, the observed polarization colour also changes, and, if the original colour is at least as high as second-order, will move down the scale by one complete order, *e.g.* second-order orange ($R = 950$ nm) changes to first-order yellow/orange ($R = 390$ nm). If the original colour is, however, within the first order (*i.e.* $R < 560$ nm), the new colour is still a consequence of subtracting 580 nm from R , but it is the absolute value of the new R (not the sign) which is relevant. For example, if the original colour is a first-order grey ($R = 100$ nm), the new colour corresponds to $R = 460$ nm, *i.e.* first-order orange. As a rapid mental aid, it is useful to think of the original colour falling to the bottom of the scale ($R = 0$) and then ‘bouncing’ back up until a change of one order has been reached, *e.g.* a first-order white, in the middle of the first order, hardly changes (half an order down followed by half an order up); first-order red changes to first-order grey (a fall of 90% of an order, followed by a rise of 10%).

If there is any doubt about the identification of the new colour, the crystal should be rotated through 90° and the second new colour examined. A comparison of the two options available, before and after rotation, rarely leaves any doubt about which is the higher colour (*i.e.* the slow-parallel-to-slow case). In all cases, whether or not the original colour is in the first order, one of the two new colours is higher than the other.

The sensitive-tint plate is so-called because it allows investigation of crystals showing very low birefringence (*e.g.* dark greys with $R \approx 50$ nm or less). In the parallel position, the new colour will lie just on the blue side of sensitive tint, and in the crossed position, just on the red side. These two colours are very easy to distinguish even though they represent only a small change in R .

From the above, it should be clear what an important aid the sensitive-tint plate can be in the actual identification of an unknown polarization colour, whether it be the body colour of the crystal or something observed in a set of grain-margin fringes. There are always two other colours that can be generated using the plate, and their relationship to the original colour is known in terms of R change, so that there are altogether three colours providing information.

The quartz wedge (Fig. 1.6.4.6) is an elongated wedge-shaped plate of progressively increasing thickness, usually cut parallel to the c axis so that the slow vibration direction is parallel to the length of the wedge. In this form it is inserted into a NE–SW slot so that the slow direction has this orientation. Some microscopes are, however, fitted with a NW–SE slot, and are provided with wedges (and sensitive-tint plates) that are ‘length fast’ rather than ‘length slow’. The optical effects are of course the same in both cases, but it is always a good idea to examine an accessory plate for its vibration directions (marked by the manufacturer) to be on the safe side.

The wedge varies in thickness from almost zero to about 0.2 mm, and typically shows a range of polarization colours from dark first-order grey up to the fourth order or so, as it is progressively inserted into the slot. The wedge can thus be used to change the polarization colour of an observed crystal by any desired amount of R within the available range (roughly 0–2500 nm). By using the two vibration directions of the crystal, these changes can be made additive or subtractive at will. In its simplest possible

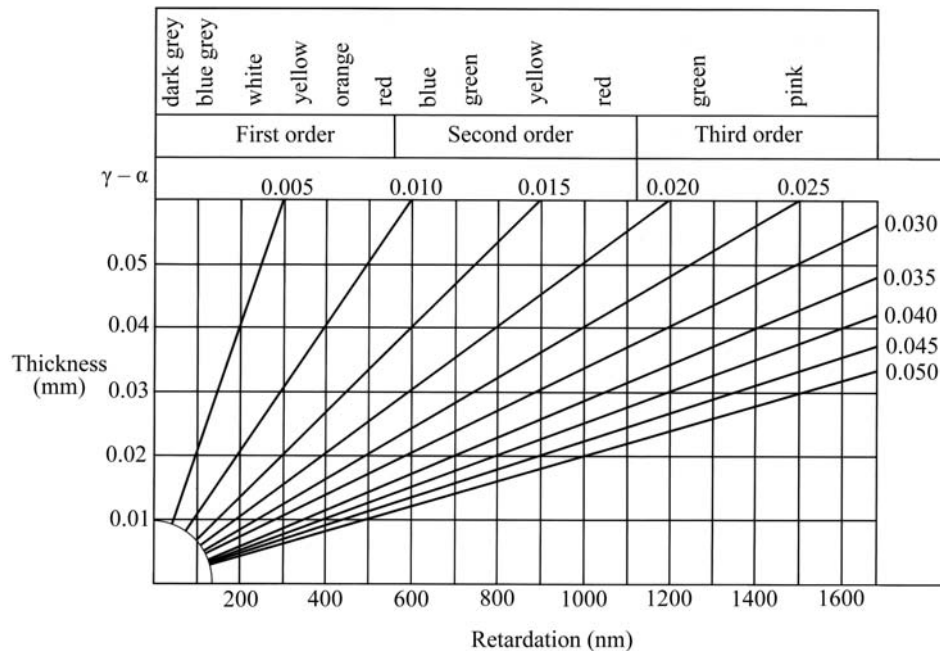


Fig. 1.6.4.6. Polarization colours *versus* thickness. The lines radiating from the origin are of equal birefringence (numerical values are given at the top and the right-hand side of the figure). The diagram may be used to determine thickness if birefringence is known, or birefringence if thickness is known. For example, the diagonal line for birefringence = 0.009 (*e.g.* quartz) intersects the standard thin-section thickness (0.03 mm) on first-order white. This is the polarization colour shown by quartz in a standard thin section.

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application, the crystal is set so that slow is against fast and the wedge is inserted until the crystal shows as close to zero birefringence as possible, *i.e.* the relative retardations of the wedge and the crystal are equal and opposite (this is called *compensation*). Next, the specimen is removed, the colour shown by the wedge noted, and the wedge is slowly pulled out, counting the orders as they go past. This is an accurate and simple alternative method of determining polarization colours.

The accessory plates are useful in identifying the order of polarization colours, but their most frequent application is in determining which of the vibration directions shown by a crystal is fast and which is slow. For example, in a specimen of a biaxial crystal lying with the optic axial plane in the plane of the slide, the slow ray represents γ and the fast ray α . To determine which is which, the vibration directions are set in the 45° position and the tint plate is inserted. If the polarization colour goes up by an order, then the slow direction of the plate is parallel to the slow direction of the crystal. Conversely, if the colour goes down by an order, or goes up by less than a complete order (when the original $R < 560$ nm), fast in the crystal is parallel to slow in the plate.

1.6.4.10. Other methods of measuring birefringence

While the use of compensating plates is convenient, more precise techniques have been developed for the measurement of linear birefringence, both in an absolute and in a relative sense. The main methods of making absolute measurements use commercially available compensators mounted on a microscope. The main types are used with a polarizing microscope with crossed polars:

(i) *Babinet compensator*: This is mounted instead of the eyepiece of the microscope, and uses two quartz wedges sliding in opposite directions to each other. The wedges are so designed that when they fully overlap, but without a birefringent specimen in the microscope, a black compensation band is seen in the centre of the field of view. Then when the specimen is placed on the microscope stage in one of the two possible 45° positions, the compensation band is shifted. When in the correct 45° position, as found by trial, the lower wedge is then screwed out to recentre the compensation band, and the distance moved is read from an internal scale. This distance is calibrated in terms of relative retardation.

(ii) *Berek and Ehringhaus compensators*: These use a rotating birefringent crystal to change their effective retardation in order to compensate against the retardation of the specimen. The Berek compensator uses a calcite plate 0.01 mm thick, whereas the Ehringhaus compensator has compound compensating plates of either quartz or calcite, made of two sections of equal thickness cut parallel to the optic axes and cemented above one another at right angles. The compensator is inserted in the slot used for accessory plates with the specimen in one of the two 45° positions. Then by tilting the compensator plate, the apparent retardations are varied until the combined retardation matches that of the specimen, thus giving rise to the compensation band appearing in the centre of the field of view. The angle of tilt can then be converted to relative rotation by the use of suitable tables provided by the manufacturer.

In order to measure birefringence in a relative sense, the following techniques have been devised. All are capable of phenomenal precision in measuring *changes* in birefringence, in some instances to one part in 10^7 .

(i) *Sénarmont compensator*: A $\lambda/4$ plate is inserted above the specimen, with one of its principal vibration directions, say the slow direction, parallel to the vibration direction of the polarizer. The analyser is rotatable with a divided circle so that the angle of rotation can be measured. It can be shown that the phase shift of the light δ is given in terms of the angle θ through which the analyser is turned to achieve extinction by

$$\delta = \frac{2\pi}{\lambda} \Delta n z = 2\theta.$$

Thus if the birefringence, or more correctly the relative retardation, of the specimen is changed, say by altering the temperature, one can follow the change simply by monitoring the angle θ . This can be done either manually, or electronically using a phase meter attached to a photomultiplier to measure the intensity as a function of the angle of the analyser, which is rotated at some frequency by a motor.

(ii) *Intensity between crossed polars*: In this case the specimen is placed in the 45° position between crossed polars and the intensity of the light through the system is measured by a photomultiplier and presented typically on a recorder. On changing the retardation of the specimen, say by heating, this intensity changes according to

$$I = I_o \sin^2 \delta/2.$$

Thus on heating a set of \sin^2 fringes is drawn out, and by counting the fringes exact measurements of δ can be made. This technique is of great sensitivity, but suffers from the fact that the specimen must be maintained throughout in the 45° position.

(iii) *Rotating analyser*: In this system (Wood & Glazer, 1980), a $\lambda/4$ plate is inserted below the substage but above the polarizer in order to produce circularly polarized light. On passing through a birefringent crystal specimen, this is generally converted to elliptical polarization. This then passes through a Polaroid analyser set to rotate about the axis of the light at a predetermined frequency ω . The resulting intensity is then given by

$$I = (I_o/2)[1 + \sin(2\omega t - 2\varphi) \sin \delta],$$

where φ is the angle between the analyser at any time and an allowed vibration direction of the specimen. Thus by measuring the light intensity with a photomultiplier and then by using, say, phase-sensitive detection to examine the signal at 2ω , a plot of $\sin \delta$ can be made as the specimen's retardation is changed. The fact that circularly polarized light is incident on the specimen means that it is not necessary to align the specimen to any particular angle. Recently, a new type of optical microscope (Glazer *et al.*, 1996) has been developed using this principle, in which false colour images representing I_o , φ and $|\sin \delta|$ can be formed (Fig. 1.6.4.7).

1.6.4.11. Interference figures

Interference figures provide one of the most powerful tools for obtaining information about a crystal. The simplest applications allow the division of crystals into uniaxial and biaxial classes, hence constraining the crystal system. Within these classes, crystals are easily divided into the subclasses of positive and negative. More advanced techniques enable the orientation of the indicatrix relative to crystallographic features to be fully established.

Interference figures are obtained when the microscope is used in its *conoscopic* configuration. This means that light is made to pass through the specimen in as wide a range of directions as possible. A powerful substage condenser is inserted to produce a wide cone of incident light and the substage diaphragm is set wide open. A diffusing screen makes a good substitute for the condenser, as it also produces light with a variety of ray directions. However, some brightness is inevitably lost. In either case, a high-power objective with a large numerical aperture is used to collect the light above the specimen, and the polarizer and analyser are crossed.

The distinction between conoscopic and orthoscopic (parallel-light) applications of the microscope is important. For observations of birefringence, the orthoscopic arrangement is required so that optical path lengths through the crystal are constant. The

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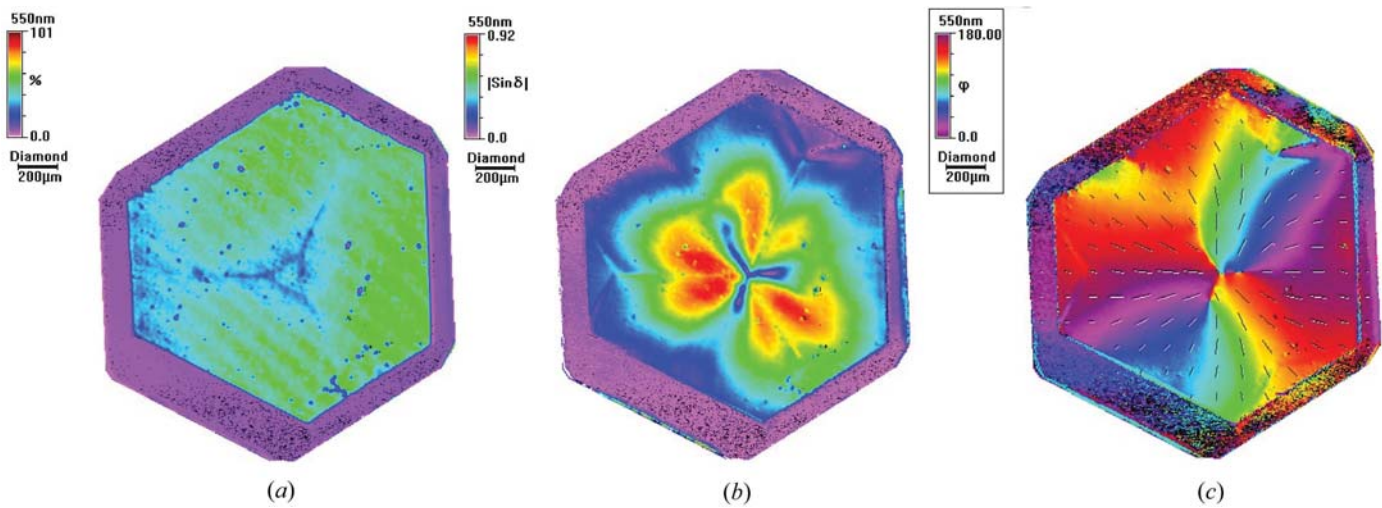


Fig. 1.6.4.7. Three birefringence images of industrial diamond viewed along [111] taken with the rotating analyser system. (a) I_0 ; (b) $|\sin \delta|$; (c) orientation φ of slow axis with respect to horizontal.

images observed in plane-polarized light rely on scattering from point sources within the specimen, and do not depend strictly on whether the configuration is conoscopic or orthoscopic. Nevertheless, relief and the Becke line are much more clearly observable in orthoscopic use.

The principle of conoscopic use is quite different. Here, the image is formed in the *back focal plane* of the objective. Any group of parallel rays passing through the specimen is brought to a focus in this plane, at a specific point depending on the direction of transmission. Hence every point in the image corresponds to a different transmission direction (see Fig. 1.6.4.8). Moreover, the visible effects are entirely caused by interference, and there is no image of the details of the specimen itself. That image is of course also present, towards the top of the tube at or near the cross wires, but the two are not simultaneously visible. The conoscopic image may be viewed simply by removing the eyepiece and looking down the tube, where it appears as a small but bright circle. More commonly however, the Bertrand lens is inserted in the tube, which has the effect of transferring the conoscopic image from the back focal plane of the objective to the front focal plane of the eyepiece, where it coincides with the cross wires and may be examined as usual.

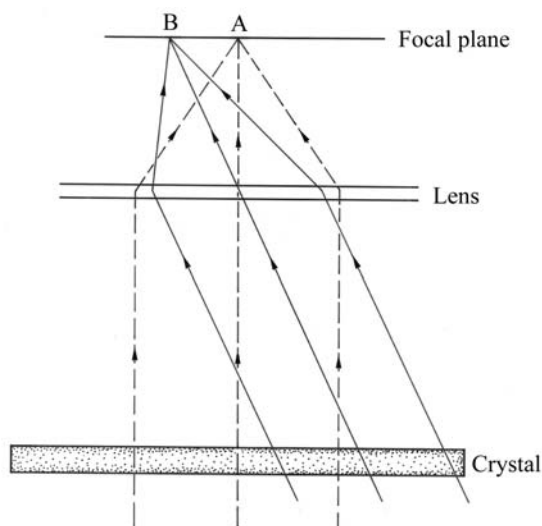


Fig. 1.6.4.8. Formation of the interference figure. The microscope axis lies vertically in the plane of the paper. A bundle of rays travelling through the crystal parallel to the microscope axis (dashed lines) is brought to a focus at A in the back focal plane of the objective. This is the centre of the interference figure. A bundle of oblique rays (solid lines) is brought to a focus at B, towards the edge of the figure.

It is useful to think of the conoscopic image as analogous to the gnomonic projection as used in crystallography. The geometrical principles are the same, as each direction through the crystal is projected directly through the centre of the lens into the back focal plane.

1.6.4.12. Uniaxial figures

To understand the formation of an interference figure, consider a simple example, a specimen of calcite cut at right angles to the c crystallographic axis. Calcite is uniaxial negative, with the optic axis parallel to c . The rays that have passed most obliquely through the specimen are focused around the edge of the figure, while the centre is occupied by rays that have travelled parallel to the optic axis (see Fig. 1.6.4.8). The birefringence within the image clearly must increase from nil in the centre to some higher value at the edges, because the rays here have had longer path lengths through the crystal. Furthermore, the image must have radial symmetry, so that the first most obvious feature of the figure is a series of coloured rings, corresponding in outward sequence to the successive orders. The number of rings visible will of course depend on the thickness of the sample, and when birefringence is low enough no rings will be obvious because all colours lie well within the first order (Figs. 1.6.4.9a and b). Fig. 1.6.4.10(a) illustrates, by reference to the indicatrix, the way in which the vibration directions of the o and e rays are disposed. Fig. 1.6.4.10(b) shows the disposition of vibration directions in the figure. Note that o rays always vibrate tangentially and e rays radially. The o -ray vibration directions lie in the plane of the figure, but e -ray vibration directions become progressively more inclined to the plane of the figure towards the edge.

The shaded cross on the figure illustrates the position of dark 'brushes' known as *isogyres* (Fig. 1.6.4.10b). These develop wherever vibration directions lie N-S or E-W, hence corresponding to the vibration directions of the analyser and polarizer. As the stage is rotated, as long as the optic axis is truly parallel to the microscope axis, the figure will not change. This is an example of a centred uniaxial optic axis figure, and such a figure identifies the crystal as belonging to the tetragonal, trigonal or hexagonal systems (see Fig. 1.6.4.11a).

From the point of crystal identification, one can also determine whether the figure coincides with the uniaxial positive ($n_e > n_o$) or uniaxial negative ($n_e < n_o$) cases. Inserting the sensitive-tint plate will move the coloured ring up or down the birefringence scale by a complete order. Fig. 1.6.4.11(c) shows the centred optic axis figure for calcite, which is optically negative. The insertion of a tint plate with its slow vibration direction lying NE-SW lowers the colours in the NE and SW quadrants of the figure, and raises

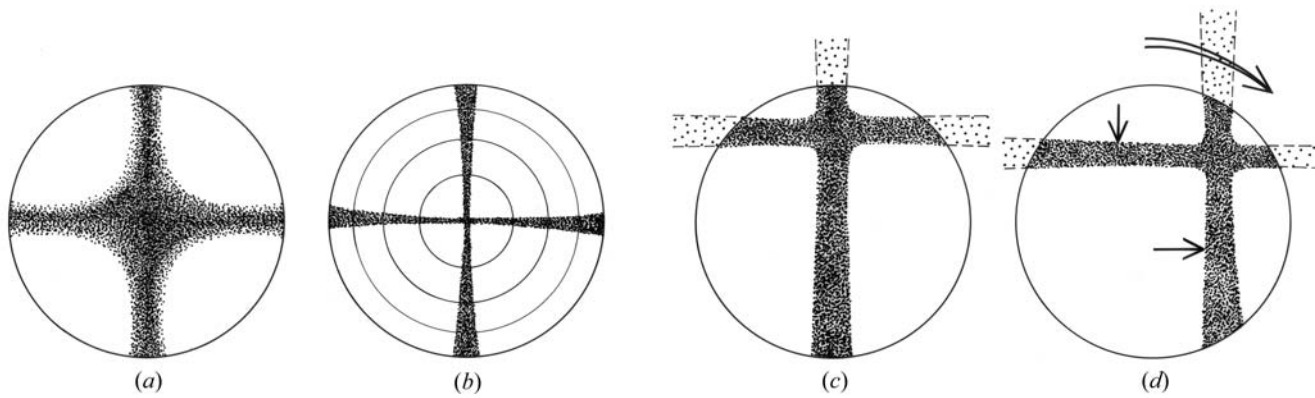


Fig. 1.6.4.9. (a) Centred uniaxial optic axis figure of a crystal with low birefringence. The isogyres are diffuse and the polarization colours in the quadrants are likely to be first-order grey. (b) As above, but for a crystal with high birefringence. The isogyres are narrower, and circular rings of polarization colours are seen, progressing outwards from first order to higher orders. (c) Off-centre uniaxial optic axis figure in the straight position (extinction position of crystal). (d) The same rotated to 45° . The curved arrow shows the rotation of the figure as the stage is turned clockwise. Arrows inside the field of view show directions of motions of isogyres. If the optic axis lies outside the field of view, such figures are difficult to interpret and may easily be confused with an off-centre biaxial optic axis figure when $2V$ is small to moderate.

those in the other quadrants (Fig. 1.6.4.11b). The simplest general rule is to look at the dark first-order grey in the original figure, lying immediately adjacent to the optic axis, *i.e.* the centre of the cross formed by the isogyres (Figs. 1.6.4.11b and c). If the crystal is optically negative, this colour changes to first-order yellow in the NE quadrant, if positive to blue. When the crystal has low birefringence, these colours may occupy the whole quadrant.

An *off-centre* uniaxial optic axis figure is obtained when the optic axis is inclined to the microscope axis by an amount which is small enough for it still to be visible within the figure (roughly within 25° of the microscope axis, using a normal high-power objective). Such figures show an isogyre cross with attendant rings, but the centre of the cross does not lie in the centre of the figure, and as the stage is rotated the centre of the cross moves round the figure in a circle (Figs. 1.6.4.9c and d). The isogyres remain NS and EW throughout. If the figure is so off-centre that the centre of the cross is not visible, the behaviour of the figure becomes difficult to interpret, and may easily be confused with some sorts of off-centre biaxial figures (see below). In the extreme case, when the optic axis lies in the plane of the slide, a quite different figure, known as a *flash figure*, is obtained. This is similar to many of the figures obtained from biaxial crystals, and will be considered further below.

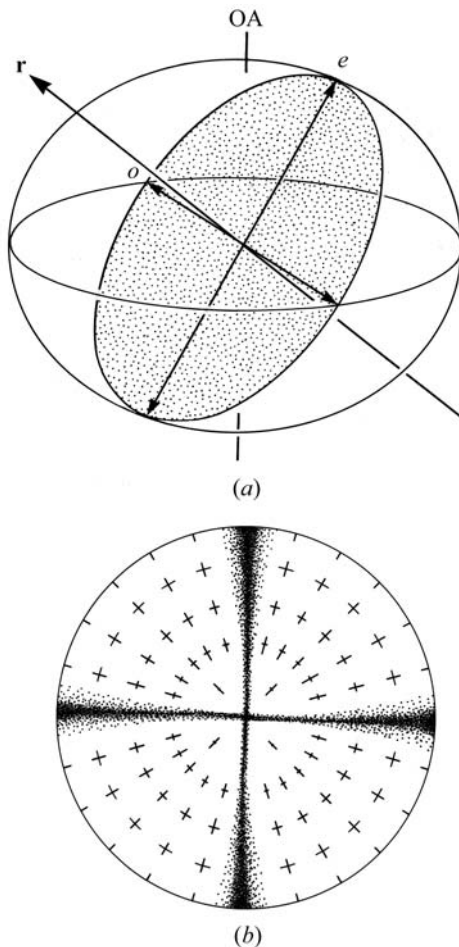


Fig. 1.6.4.10. (a) Vibration directions of an oblique ray passing through calcite, viewed relative to the indicatrix. OA is the optic axis, normal to the circular cross section of indicatrix, \mathbf{r} is the ray direction, e and o are, respectively, the vibration directions of the extraordinary and ordinary rays in the plane of the elliptical cross section of the indicatrix normal to \mathbf{r} . (b) Vibration directions as seen in the figure. The tangential set represents the o rays, the radial set represents e rays. The isogyres are also shown.

1.6.4.13. Biaxial figures

(i) *Acute bisectrix figures*: Biaxial figures may be introduced by considering a crystal with a small $2V$ angle, oriented so that the acute bisectrix is parallel to the microscope axis. The conoscopic figure will have much in common with the uniaxial case already discussed (and indeed will become identical as $2V$ tends to zero), and some form of bilateral rather than radial symmetry is to be expected.

Vibration directions within the figure can be deduced by consideration of the *Biot-Fresnel* construction. Given that the form of the indicatrix is known, this is used to determine vibration directions in any orientation required. A general case presented in the form of a stereographic projection is given in Fig. 1.6.4.12(a). It is first necessary to know the positions of the optic axes and the position of a ray direction. Two planes are constructed, each containing one optic axis and the ray direction. Two *additional* planes are constructed, each containing the microscope axis and *bisecting* the angles between the first two planes. The plane in which the vibration directions lie is plotted normal to the ray direction, and the two vibration directions in this plane are fixed by the intersections of this plane with the two additional planes. This is also a very powerful construction to use in conjunction with orthoscopic methods. If crystallographic features, *e.g.* traces of cleavage planes, crystallographic axes, twin planes *etc.* (twinning is discussed in Chapter 3.3) are added to the stereogram, it becomes possible to determine the predicted angles between vibration directions and crystallographic features. One of the most fundamental aims of polarizing microscopy is the determination of the complete orientation of the indicatrix relative to crystallographic directions. Familiarity with the Biot-Fresnel construction is a substantial aid.

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Returning to the interference figure, a modified version of the Biot-Fresnel construction is illustrated in Fig. 1.6.4.12(b). Since the interference figure is analogous to a gnomonic rather than a stereographic projection, the various constructional planes required are straight lines. The vibration directions for the complete figure are given in Fig. 1.6.4.12(c).

Considering first the isogyres (brushes), if the optic axial plane lies N-S, as in Fig. 1.6.4.13(a), clearly the isogyres form a cross, as in the uniaxial case. There is a small difference, however. Because the vibration directions curve round more rapidly near the optic axial plane, the N-S isogyre is sharper than the E-W, which is relatively broadened and less dark.

If the slide is rotated so that the optic axial plane lies NE-SW, inspection of the vibration directions in Fig. 1.6.4.13(b) shows that the isogyres must part in the direction shown. Rotation of the stage through a full 180° will thus show two positions at which the brushes cross – once when the optic axial plane lies N-S, the other when it lies E-W – and two positions of maximum separation of the isogyres – when the optic axial plane lies NE-SW and NW-SE (see Fig. 1.6.4.14a). The position of the broader, more diffuse, brush will alternate. The amount by which the two

isogyres are separated in the 45° position depends on the size of 2V. Since the field of view is usually about 60°, when 2V is 60° the isogyres will retreat right to the edge of the field of view in the 45° position, and then come back as rotation continues. When 2V > 60°, the brushes will entirely leave the field of view for part of the rotation (Fig. 1.6.4.14c). Approximate estimates of 2V can be made on this basis.

As in the uniaxial case, coloured fringes are also part of the figure, and are most obvious when birefringence is high (Fig. 1.6.4.14a and Fig. 1.6.4.15a). The outermost fringes have an elliptical form, but towards the centre of the figure they are distorted into a dumbbell-like or figure-of-eight form. This arises

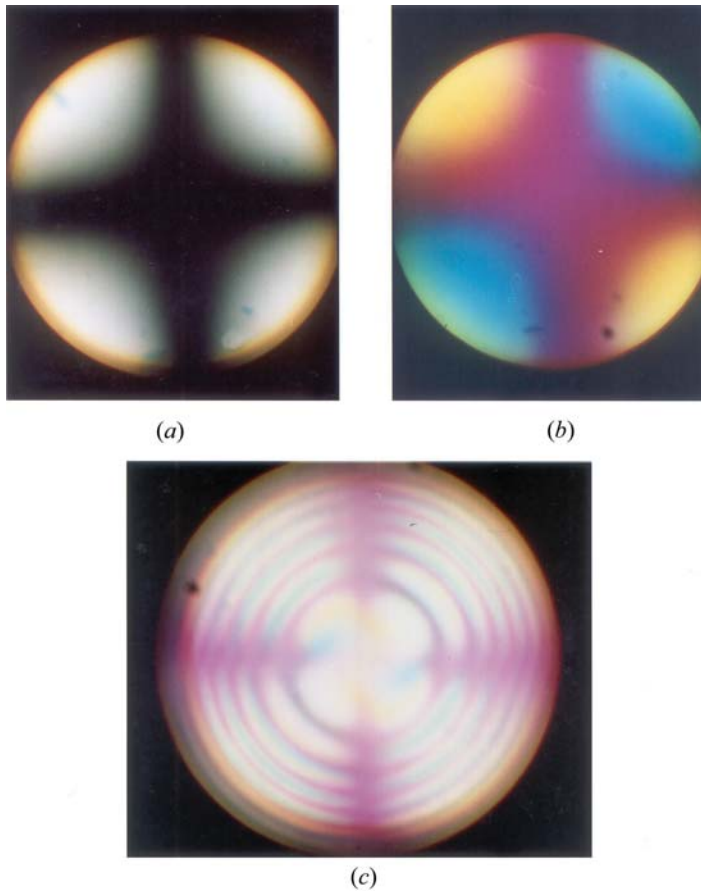


Fig. 1.6.4.11. (a) Centred optic axis figure of quartz (uniaxial). The birefringence is low, resulting in a diffuse cross and quadrant colours in the lower half of the first order. (b) The same figure with the sensitive-tint plate inserted (slow vibration direction NE-SW). The isogyres take on the sensitive-tint colour. Compensation takes place in the NE and SW quadrants (slow directions of crystal and plate parallel to each other) resulting in second-order blue, that is, one whole order above the original. In the other quadrants, the colours also apparently rise, but are restricted to yellow (high first-order). Thus the crystal is optically positive, that is, *e* rays (radial) are slow and *o* rays (tangential) are fast. (c) Centred optic axis figure of calcite with sensitive-tint plate (slow direction lying NE-SW) inserted. The birefringence is very high so there are many coloured rings. In the top right quadrant, the colour immediately adjacent to the cross is first-order yellow, whereas in the top left it is second-order blue. Calcite is hence optically negative. It can be observed that all the coloured rings in the top right and bottom left quadrants are an order lower than their counterparts in the top left and bottom right.

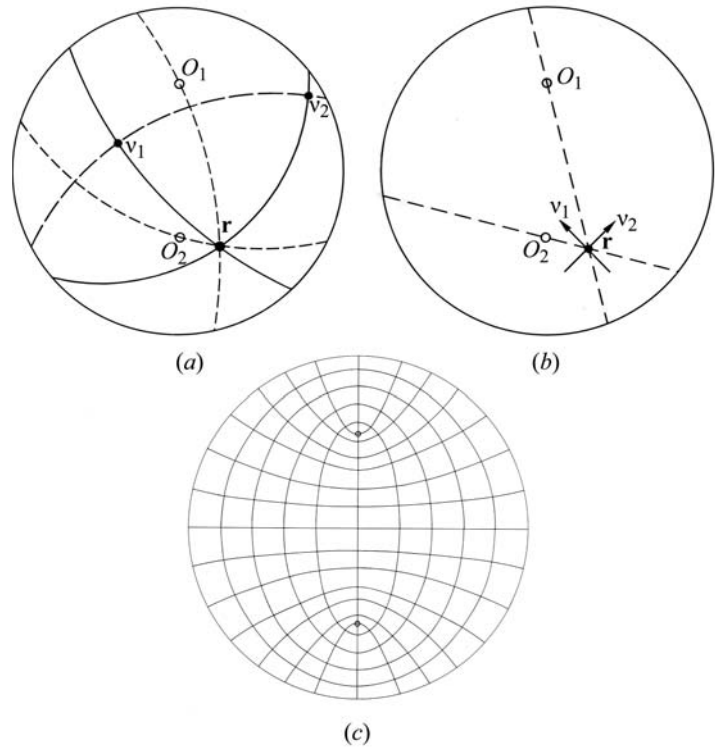


Fig. 1.6.4.12. (a) The Biot-Fresnel construction illustrated on the stereographic projection. (b) Method of determining vibration directions in a biaxial acute bisectrix figure using the Biot-Fresnel construction and a gnomonic projection. (c) Complete vibration directions of a figure similar to the above. As noted in the text, the directions shown are components resolved into the plane of the figure, and the true vibration directions are in general somewhat inclined.

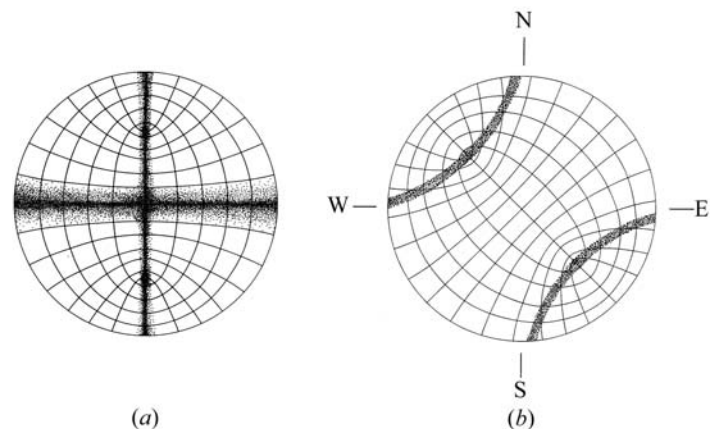


Fig. 1.6.4.13. (a) Illustration of the formation of crossed isogyres when the optic axial plane of a biaxial figure lies N-S. The shaded lines illustrate the zones in which vibration directions are N-S and E-W, leading to the formation of the isogyres. Along the E-W line, the zone across which vibration directions depart little from N-S and E-W is broad, hence this isogyre is wider than the N-S isogyre. (b) Illustration of the formation of separated curved isogyres when the same crystal is in the 45° position (optic axial plane NE-SW).

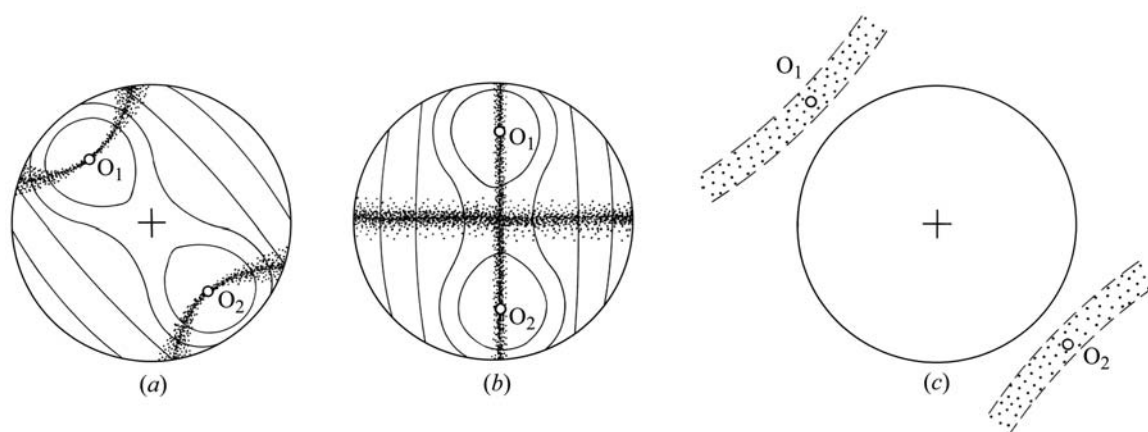


Fig. 1.6.4.14. (a) Drawing of biaxial optic axis figures set in the 45° position with the optic axial plane set NW–SE. $2V$, represented by the angle O_1 – O_2 , is about 40° (cf. total field of view of ca 60°). The distribution of coloured fringes is represented, with near-circular areas of first-order colours around the optic axes, progressing outwards to higher orders (see also Fig. 1.6.4.15a). (b) The same figure in the straight position (optic axial plane N–S). Note that the isogyre along the optic axial plane is sharper than the E–W isogyre. (c) Representation of a biaxial figure in the 45° position when $2V > 60^\circ$. The isogyres leave the field completely.

because the areas of minimum birefringence must lie adjacent to the optic axes, and birefringence, although generally increasing outwards, must also increase towards a point midway between the optic axes. This effect is slight when birefringence is low, but is prominent when birefringence is high, in which case each optic axis is surrounded by its own set of almost-circular fringes, set within the general elliptical array.

Optic sign can be determined from the acute bisectrix biaxial figure in a way analogous to the uniaxial case. The stage is rotated to the 45° position, *i.e.* with maximum separation of the isogyres, and the analogues of the uniaxial quadrants are identified. The difference here is that in one of the 45° positions the NE and SW quadrants join through the space between the optic axes, and in the other the NW and SE quadrants are connected. The sensitive-tint plate is employed just as in the uniaxial case, and the same rules apply to change of colour (Fig. 1.6.4.15a and b). For example, suppose that the stage is turned so that the NE and SW quadrants are separated from each other. Optically positive crystals show second-order blue close to the isogyre within the quadrant (on the concave side of the isogyre), while first-order yellow appears on the convex side, which is part of the combined NW and SE quadrants. Optically negative crystals show the reverse.

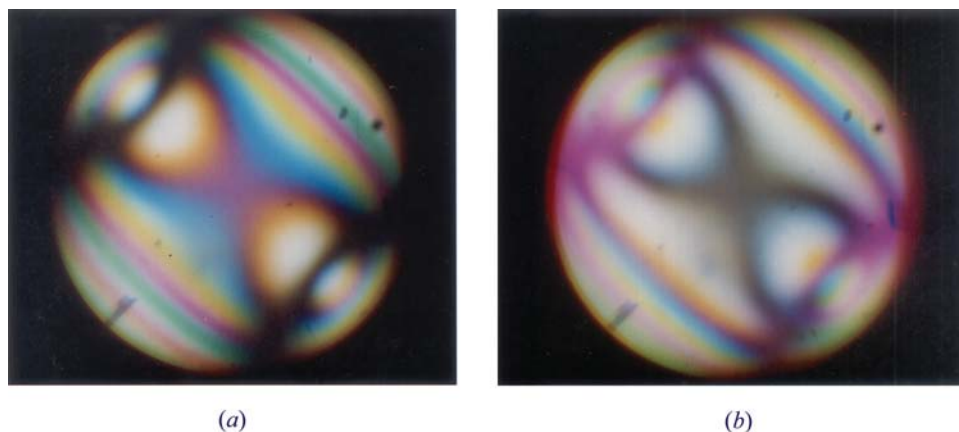


Fig. 1.6.4.15. (a) Acute bisectrix figure of a biaxial crystal with $2V \simeq 40^\circ$. The optic axial plane is oriented NW–SE. First-order colours are confined to the two circular ‘eyes’ or melatopes around the optic axes, with sensitive tint itself making a figure-of-eight shape (‘dumbbell’). At the edge of the figure in the NE and SW directions the coloured fringes reach third-order pink. (b) The same figure with the sensitive-tint plate inserted (slow direction NE–SW). The isogyres have taken on the sensitive-tint colour. Between the optic axes (*e.g.* the centre of the figure), the original sensitive-tint dumbbell has become black, and to the NW and SE of the centre all the fringes have dropped by one order. Conversely, in the extreme NW and SE directions all fringes have gained an order. The little patches of yellow lying just inside the optic axes, and the accompanying blue patches just outside, indicate by analogy with the uniaxial figure that the crystal is optically negative.

Slightly off-centre acute bisectrix figures are easy to interpret if $2V$ is low, but as the degree of off-centring increases, and as $2V$ increases, it is obvious that interpretational difficulties will rapidly increase, and recourse must be made to the optic axis figure.

(ii) *Optic axis figures*: Although the acute bisectrix figure forms the most convenient introduction to the subject, it is in practice quite difficult to find a suitable section without trial and error. In monoclinic and triclinic crystals, for example, the acute bisectrix section is not in general identifiable by reference to crystallographic features (cleavage, faces *etc.*), nor by any special optical properties observable under orthoscopic conditions. As in the uniaxial case, however, the optic axis section is identifiable by its low-to-nil birefringence. The optic axis figure is the workhorse of the microscopist.

The essential feature of this figure is that, as the stage is rotated, one of the optic axes is positioned permanently in the centre of the field of view, while the other rotates around it. One of the isogyres is always visible. When $2V$ is small, on rotation of the stage the figure simply looks like a wobbly version of the acute bisectrix figure. Both isogyres are visible, and determination of optic sign, using the sensitive-tint plate, is as described above.

As $2V$ increases, however, the second isogyre leaves the field during part of the rotation, and the point where the isogyres cross, when the optic axial plane lies N–S or E–W, moves towards the edge of the field of view. Eventually, with further increase of $2V$, the second isogyre does not enter the field of view at all, and the figure consists of a single brush rotating about the optic axis. The brush changes from straight, when it lies N–S or E–W, to curved, when the crystal is in the 45° position, and the degree of curvature decreases with rising $2V$. When $2V = 90^\circ$, the brush remains straight in all positions. With experience, the maximum curvature of the isogyre can be used for rough estimation of $2V$.

Optic sign is readily determined from any sort of optic axis figure by setting the crystal in the 45° position and using the curvature of the isogyre to identify the various ‘quadrants’ as described above. This of course is not possible when

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$2V = 90^\circ$, and such crystals are described as optically neutral.

A further piece of useful information is derivable from optic axis (and acute bisectrix) figures, *i.e.* the direction of the optic axial plane, that is, the line joining the two optic axes. Even if the second isogyre is never visible, the position of the second optic axis, outside the field of view, is easily judged. By reverting to orthoscopic observation without moving the specimen, it is easy to see how the optic axial plane relates to crystallographic features such as faces, cleavages or twin planes.

(iii) *Other biaxial figures*: Several special figures are occasionally useful, and their behaviour is readily explained by comparison with the acute bisectrix figure. The obtuse bisectrix figure behaves like the acute bisectrix figure, but as if $2V$ were very large. The isogyres form a diffuse cross in the centre of the field, and then, on rotation, rapidly depart from the field, either *via* the NE and SW quadrants or *via* the NW and SE. As rotation approaches 90° , they re-enter the field of view from the other direction. When $2V$ approaches 90° , there is of course very little difference between the acute and obtuse bisectrix figures, and to determine which is which it is necessary to measure the speed at which isogyres leave the field of view during rotation (for a given rotation the obtuse bisectrix isogyres move further).

The optic normal figure (often known as a *flash figure* because of the extreme speed of isogyre movement) is formed when the optic axial plane lies in the plane of the slide. Its behaviour on rotation of the stage is an extremely rapidly moving version of the obtuse bisectrix figure. The isogyres are very broad and diffuse, and when the cross is formed there is little more than a momentary darkening of the whole field of view. The brushes vanish completely from the field with only a very small rotation from this position. Often it requires careful inspection even to be certain that the cross splits into two brushes that vanish diagonally from the field.

The optic normal figure is useful in orientation studies and, as mentioned above, grain mounts of orthorhombic crystals may naturally give rise to suitably oriented crystals. In thin sections, the figure is useful to confirm that grains apparently showing maximum birefringence do in fact lie with their optic axial planes normal to the microscope axis. When only a small number of grains are available, this check is important.

In the most general sense, this exercise illustrates that all figures are variations on a single theme, in which the vibration direction diagram of Fig. 1.6.4.12(c) can be thought of as being viewed from different distances (delimiting the size of the area viewed) and different directions (*i.e.* where the centre of the field of view is, relative to the centre of the diagram). For, example, the figure shown represents a centred uniaxial optic axis figure if viewed from infinite distance in a direction corresponding to the centre of the diagram. On the other hand, if viewed from close up in the same direction (so that the field of view is thought of as a rather small circle in the middle of the diagram), the figure becomes analogous to the acute bisectrix figure of a crystal with a large $2V$, the obtuse bisectrix figure, or even the uniaxial flash figure (but see below), with increasing distance of the viewpoint. Off-centre figures can be equally well explained by placing the viewpoint non-centrally.

Only the uniaxial flash figure and the biaxial optic normal figure seem, at first sight, to defy exact interpretation, because the optic axes, lying in the plane of the figure, seem to require to be plotted at infinity in the plane of the paper. This then requires that the map of the vibration directions consists of two linear sets at a right angle to each other. No explanation of the rapid separation of the diffuse isogyres accompanying a small rotation from the extinction position is forthcoming. Rather, a simple extinction of the whole field is predicted at every 90° .

In fact, the vibration directions seen in such figures still retain the 'barrel-shaped' configuration seen in Fig. 1.6.4.12(c), though degrees of curvature can be very small. This arises because the

true vibration directions of, for example, the e rays in the uniaxial case, do not lie in the plane of the figure (except at the centre). A curved surface is required for the true description of the vibration directions. We have up to this point regarded the diagram as flat, but in fact the vibration directions, as we view them, become distorted into the barrel shape.

1.6.4.14. Orientation studies

The full skills of the microscopist are required in the production of an accurate description of the optical orientation of a sample, that is the relationship of the indicatrix to the crystallographic axes.

It is best to start by determining the crystal system. Completely isotropic crystals are cubic, uniaxial crystals are tetragonal, trigonal or hexagonal, while biaxial crystals are orthorhombic, monoclinic or triclinic. Crystallographic features such as edges, faces, crystal outlines, shapes, cleavages and twin planes give the additional information required to subdivide the uniaxial and biaxial classes.

In uniaxial cases, a grain giving a centred optic axis figure is required. If the crystals look square, or have cleavages and/or twin planes intersecting at right angles, then the system is most likely to be tetragonal. Features disposed as equilateral triangles indicate triclinic, and hexagonal arrangements may indicate triclinic or hexagonal. The cases are often impossible to distinguish.

Orthorhombic crystals are usually fairly easy to identify because, although biaxial, they still show parallelism between many optical and crystallographic properties. For example, vibration directions commonly lie parallel to the traces of cleavage planes or crystal outlines (so-called *straight extinction*). Alternatively, vibration directions may bisect the angles between such features (*symmetrical extinction*). Furthermore, crystals with obviously special orientations can be identified, *e.g.* a section showing two sharply defined cleavages (*i.e.* lying at right angles to the plane of the slide) perhaps at right angles to each other, or producing a diamond pattern, is obviously cut normal to an important crystallographic direction, perhaps containing two of the crystallographic axes. The interference figure of such a section should be examined carefully as it is likely to be a centred version of the acute bisectrix, obtuse bisectrix or flash figure.

Monoclinic crystals are extremely common, and, while biaxial, do not generally show the parallelism of optical and crystallographic features typical of the orthorhombic system. There is no general recipe for success in determining the optical orientation of such crystals, other than systematic observation of crystals in different orientations. The most important observations are the relationships between extinction positions and crystallographic features, and the nature of the interference figures. All monoclinic crystals have one plane that, if at right angles to the slide, shows symmetrical or straight extinction. This plane is observed when the twofold symmetry axis lies in the plane of the slide. In crystals that show two cleavages, their intersection is also likely to mark a crystallographic axis. From consideration of such features it is often possible to identify a crucial special section, that lying perpendicular to the twofold axis. This is an important section, because two crystallographic axes now lie in the plane of the slide and their directions may be indicated by cleavage traces, crystal edges *etc.* Determination of the angles between vibration directions and supposed crystallographic axis directions then gives the important angle (*e.g.* n_γ , c) which expresses the tilt of the indicatrix within the plane normal to the twofold axis.

If everything fails, and no relationship can be found between crystallographic and optical directions, the crystal is probably triclinic, and it is not possible to say very much about its orientation using the flat-stage microscope. Recourse must then be had

to the *universal stage*, a device that allows rotation of the slide in three dimensions. This is rarely done these days.

Orientation studies are completed by assigning specific axes of the indicatrix to specific crystallographic axes. The identification of the principal axes of the indicatrix is easy. For example, in uniaxial cases, sections showing maximum birefringence contain the unique crystallographic axis, which is parallel to the n_e direction. Knowledge of the optic sign shows which of the two vibration directions coincides with n_e , on the basis of being fast or slow. In biaxial cases, the maximum birefringence section has n_α and n_γ lying in the plane of the slide, and of these n_γ corresponds of course to the slow ray. In biaxial crystals, the identification of the optic axial plane direction in a figure enables immediate identification of the n_β direction, which is normal to it.

1.6.4.15. Absorption colours

Many crystalline substances are coloured in transmitted light under the microscope, as a result of the absorption of certain visible-light wave bands. Though there may be much variability between one sample of a substance and another, colour is nevertheless often a great aid in identification.

Absorption, like other optical phenomena, is capable of showing marked anisotropy, the phenomenon known as pleochroism. Pleochroism is usually obvious in both grain mounts and thin sections, because grains change colour when rotated in plane-polarized light. The effect can be subtle (*e.g.* the mineral hypersthene is almost colourless in thin sections but often shows pleochroism from a very faint pink to a very faint green), or very marked (*e.g.* dark brown to colourless or pale yellow in the mineral biotite). The full description of pleochroism involves the assigning of different colours to specific axes of the indicatrix. Uniaxial crystals are *dichroic*, that is, two colours describe the effects. Sections showing centred optic axis figures do not exhibit pleochroism, showing only the pure absorption colours of light vibrating normal to the optic axis (n_o). In biotite for example (which is for practical purposes uniaxial), this colour is usually dark brown. Maximum birefringence sections, or those showing centred flash figures in grain mounts, show maximum pleochroism, one vibration direction exhibiting the n_o absorption already noted, and the other the pure effects of absorption on n_e (in the case of biotite, colourless to pale yellow). Random sections show less pleochroism, and colours that are combinations of the pure end members.

Biaxial crystals can be notably trichroic, and colours are readily assigned by looking at maximum birefringence (flash figure) sections to obtain n_α (fast ray) and n_γ (slow ray) absorptions, and optic axis sections to obtain that for n_β .

The other notable thing about absorption colours is that they can occasionally severely mask polarization colours. Once this is appreciated, the use of the sensitive-tint plate is often sufficient to identify the latter.

1.6.4.16. Dispersion

Dispersion, that is to say variation of refractive index depending on wavelength, is a common phenomenon in crystals, and occasionally an important aid in identification. In extreme cases, it results in the production of highly anomalous polarization colours, often browns, blue greys or brownish purples, or bright colours which just look slightly unusual compared with normal second- or third-order colours. Anomalous colours are highly diagnostic of certain substances (*e.g.* the minerals chlorite, zoisite and epidote).

Dispersion also results in a lack of definition of extinction positions, because in biaxial crystals there may effectively be differently oriented indicatrices for different wavelengths. In extreme cases, it may be impossible to locate the extinction position with any accuracy. As an example of a milder case, the very common mineral plagioclase (triclinic) shows subtle, though

highly characteristic, features between crossed polars in sections cut approximately normal to the b crystallographic axis. The normal polarization colours seen in between the extinction positions are pure first-order greys and whites, but as a grain is rotated slowly through an extinction position the colours darken and take on a bluish (cold) tinge before going black, and then lighten again with a yellowish (warm) tinge. This is the result of a slight mismatch of the orientations of the indicatrices for long and short wavelengths.

Such dispersion is also often obvious (at least, when looked for) in the interference figures of biaxial crystals. Isogyres become edged with 'cold' (*i.e.* bluish) and 'warm' (tending to red or orange) fringes on opposite sides, indicating that optic axes for different wavelengths have slightly different positions.

Dispersion is a difficult phenomenon to investigate fully with the polarizing microscope. In cubic crystals, it can only be studied systematically by the determination of refractive indices using a number of monochromatic light sources of different wavelengths. Uniaxial crystals can show anomalous polarization colours, but they do not show fringes in interference figures, nor vagueness in extinction position, because the indicatrices for different wavelengths all have the same orientation. Like cubic crystals, however, the dispersion can be investigated by monochromatic light studies of refractive index. Biaxial crystals present the most complex cases. Not only can the shape of the indicatrix vary with wavelength (*i.e.* the relative values of the principal refractive indices), but so can orientation relative to the crystallographic axes. There are even substances known (admittedly with small $2V$) in which the optic axial plane for red light is at right angles to the optic axial plane for violet light. The phenomenon is known as 'crossed axial plane dispersion', a real challenge for the microscopist.

1.6.5. Optical rotation

1.6.5.1. Introduction

Optical rotation or gyration, as it is sometimes known, was first recorded by Arago in 1811. Since then, a great deal of work has been done to try to explain this phenomenon, and at the present time it is one of the few physical properties of a crystal that can be successfully understood in terms of the underlying crystal structure (Glazer & Stadnicka, 1986). Lowry (1935) has given a good historical account of the subject.

Optical rotation is the phenomenon observed in some crystals (and in some solutions of, usually, organic compounds) of the rotation of plane-polarized light on passing through the crystal. If the rotation, as seen by the observer, is to the left or counter-clockwise it is known as *laevorotation*; if to the right or clockwise it is known as *dextrorotation*. A crystal that shows this effect is sometimes called *optically active* or *gyrotropic*. It is therefore clear that, from a symmetry point of view, optical rotation can only occur in a crystal in which one direction is not equivalent to its opposite, *i.e.* there are no inversion symmetry operations that can change chirality.

One of the earliest theories to explain the origin of optical rotation was given by Fresnel. This was based on the idea that a plane-polarized wave can be equally described by two opposing circularly polarized waves propagating along the same direction. In Fig. 1.6.5.1(a), the polarization vector **OA** is simply given by the vector sum of **OB** and **OC**, polarization vectors at some instant of time belonging to a right-circular and a left-circular wave, respectively. Here it is assumed that the light propagates in a direction towards the viewer. If the light now passes through a gyrotropic material, one of the circularly polarized waves will be slowed down with respect to the other (this corresponds to having a small difference in refractive indices for the two waves). Thus on emerging from the crystal, there will be a phase difference between the circular waves, so that on recombining, the resulting