EXPERIMENTS IN CRYSTAL OPTICS

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INTRODUCTION

In most crystalline materials, the speed of light varies with direction. This phenomenon, optical anisotropy, is the principal subject of optical crystallography. The most striking feature of optical anisotropy is double-refraction (or retardation). Macroscopically, it manifests in three ways:

- In traveling through the crystal, an incident light ray is broken into two rays,
- rays emerging from the crystal are polarized,
- a crystal, sandwiched between crossed polars, shows interference colors in transmitted light.

The classic physical optics textbook approach to double-refraction starts from Huyghens constructions of wave fronts and from the optical indicatrix. Optical indicatrices are useful for a systematic description of optical properties in crystals, but students do not usually consider them an easy subject, and, therefore, shy away from optical crystallography. This is unfortunate since a basic understanding of optical crystallography is prerequisite to a correct interpretation of phenomena observed with the polarizing microscope, the most commonly used tool for the detailed study of rocks.

Generally, students are comfortable with simple optical terms like reflection and refraction, while it is uncommon that they actually have seen double-refraction and noticed that crystals polarize light. Many have an unnecessarily complicated idea about vibration directions, interference colors, and interference figures; they assume such phenomena always require a microscope to observe. This is not so. Students well trained in thin section microscopy are often surprised that interference figures can be made visible macroscopically.

The purpose of the experiments below is to impart an intuitive understanding of the interaction between light and crystals and, thus, of optical crystallography. This will help to demystify what is seen in the polarizing microscope, and will better prepare the student for the introduction of optical indicatrices as 3-D models to describe the directional dependence of light velocities, and thus refractive indices in anisotropic crystals.

Background

The following demonstrations and exercises are designed for use in an undergraduate course in mineralogy or optical crystallography. If students are not familiar with elementary optics, I suggest a brief explanation, or recapitulation, of (a) Snell's Law (i.e. the Law of Refraction: rays at boundary surfaces, index of refraction, speed of light in matter) and (b) polarization of light (unpolarized light, polarized light, polarizer, analyzer, plane of polarization). Furthermore, students should have a basic knowledge about crystal systems, crystallographic axes, etc.

CLASS ROOM DEMONSTRATION OF DOUBLE REFRACTION

Materials needed:

- Optical bench, 100 cm
- Helium-neon laser#), 0.2/1.0 mW
- 1/4-lamda filter#)
- 2 polarizing filters with angular scale#)
- Lens, f=150 mm#)
- Table#) or ring*) to hold crystal
- Clear calcite cleavage rhombohedron (at least 30mm x 40mm x 60mm).
- Clear quartz prism (diameter at least 30mm)

Optical bench, laser, lens, filters etc. are standard equipment in physics departments and used in lab classes on geometric optics. At our university, we borrow this equipment from the Physics Department.

Experimental set-up and preparations

The experimental setup is illustrated in Figures 1A and 1B. I use a HeNe-laser for a light source (maximum output 1mW, with an integrated grey filter the output can be reduced to 0.2mW). The laser is clamped to the optical bench. The laser beam should be unpolarized. But since unpolarized lasers fluctuate their polarization state, it is best to make the beam circularly polarized by inserting a 1/4 lambda plate in the laser beam. Mount the calcite rhomb with the rhombohedron face normal to the incident ray. If the rhomb is held by a table, plasticine helps fix the crystal in the correct position.

Two parallel rays leave the crystal: the ordinary and extraordinary ray. Depending on the rhombohedron size, the spacing between these two rays is on the order of a few millimeters or, at most, a centimeter. In classes with a larger audience, most students will be too far away to actually see the two emerging rays as distinctly separate. This may be remedied by placing a convex lens in the passage of rays. The lens allows one to project the points of emergence on to a screen. (Points of emergence are the points where the two beams emerge from the crystal.) The blackboard or a wall will do as a screen. With sufficient distance from lens to screen (several meters), the images of the two points, and thus of the two rays, will become visible to students seated far away.

If the incident laser beam, ordinary ray and lens axis all are coaxial (they should be), the ordinary ray traverses the lens undeflected. The extraordinary ray is refracted by the lens and crosses the ordinary ray at the focal point. Beyond the focal point, the two rays diverge and can, therefore, appear as two clearly separate spots on the screen (Fig. 1B). The spacing between these two spots increases with decreasing focal length of the lens, and with increasing distance between lens and screen.

#) Laser, filters, lens and table are mounted on stems. These stems fit in holders clamped to the optical bench.

*) For a slightly more sophisticated version of the experiment, the table is replaced by a ring (Fig. 1) mounted on a stem.

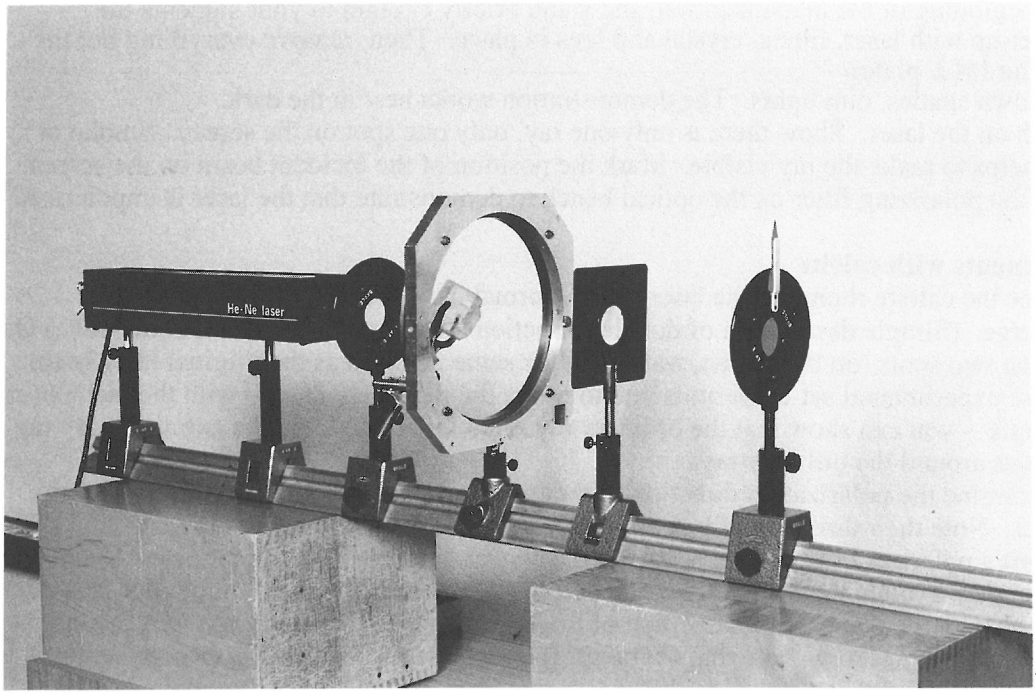

FIGURE 1A. Experimental set-up for the demonstration of double-refraction in calcite. From the left: laser, λ/4-plate, calcite rhomb, lens, analyzer.

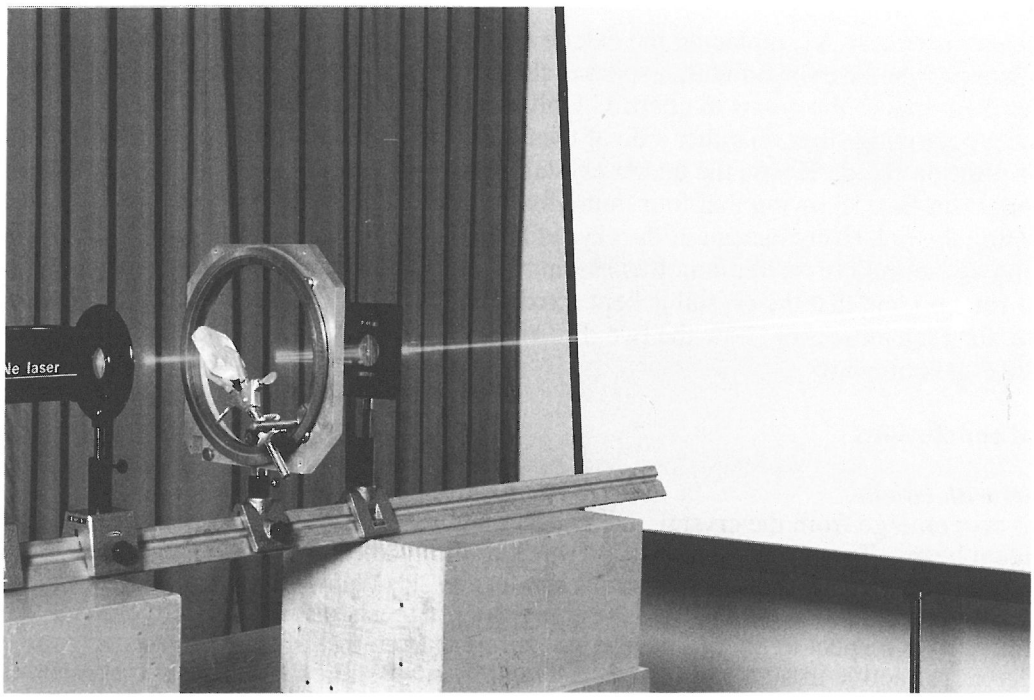

FIGURE 1B. Passage of rays. If the calcite is not perfectly clear and contains cracks, at very close distance, very thin beams may be noticed. From farther away, only the primary beam and the emerging beams are visible.

The emerging rays also pass through a rotatable polarizing filter.

At the beginning of the demonstration, show and briefly explain to your students the complete set-up with laser, filters, crystal and lens in place. Then, remove everything but the laser (and the 1/4 λ plate).

- Pull down shades, dim lights. The demonstration works best in the dark.
- Switch on the laser. Show there is only one ray, only one spot on the screen. Smoke or dust helps to make the ray visible. Mark the position of the incident beam on the screen.
- Place the polarizing filter on the optical bench to demonstrate that the laser is unpolarized.

A. Experiments with calcite

1. Place the calcite rhomb in the laser beam (normal incidence). One ray enters, two emerge. (Simple description of double refraction: "One ray goes in, two come out".) One of the two spots (on the screen, wall) is in the same position as the original laser beam.

1a. If the experimental set-up permits you to rotate the rhombohedron -- with the incident ray as axis -- you can show that the ordinary ray is stationary and that the extraordinary ray circles around the ordinary ray.

2. Determine the polarization direction of the two emerging rays by using the polarizing filter. Note their direction of polarization with respect to the c-axis of the crystal.
3. Insert a polarizer between the laser and the crystal, making the incident beam plane-polarized. Rotate the plane of polarization by turning the polarizer. Show that this does not change the planes of polarization of the emerging beams; it only changes the intensity of the two rays. Also, note that every 90° rotation one of the rays will become extinct.
4. Rotate the crystal about the incident beam (which is normal to the rhombohedral face). Show that planes of polarization of the emerging beams rotate with the crystal, (i.e., that they are fixed with respect to its c-axis.)

B. Experiments with quartz

1. Repeat experiment A1, replacing the calcite rhomb by a quartz crystal. Ensure normal incidence of the laser beam with respect to the prism face of the crystal (i.e., the laser beam is normal to the c-axis in quartz). Only one ray is visible.
2. Place a polarizing filter on either side of the quartz crystal. Cross the two filters and keep their position fixed. Rotate the quartz crystal (not the polars!) 360° about an axis normal to the prism face, showing that four mutually perpendicular extinctions occur.

2a. Rotating the polarizers instead of the crystal leads to the same result: Four extinctions during one complete revolution. It is essential that both polarizers remain crossed during their rotation and that the crystal is kept fixed.

3. Note, for each extinction, how the two directions of polarization are oriented with respect to the c-axis in quartz.

Results and conclusions

Experiments with calcite.

1. Two rays emerge from the crystal, one undeflected, the other deflected with respect to the incident beam. The undeflected ray is termed 'ordinary', because it follows Snell's Law. The deflected ray does not follow Snell's law and so it is called the 'extraordinary ray'. The different refraction behavior of these two rays suggests that the refractive index effective with respect to the ordinary ray is different from that of the extraordinary ray. Different refractive indices result from different light velocities. Thus light traverses the calcite at two different speeds, at the same time.
2. The emerging rays are examined with a polarizing filter. During one complete turn of this polarizer, each of the rays disappears twice; extinctions are 90° apart. This proves

that the emerging beams are linearly polarized, and their planes of polarization are mutually perpendicular.

3. Extinction always occurs in definite positions of crystal and polarizing filter relative to each other. Consequently, planes of polarization (vibration directions) in the crystal are fixed with respect to crystallographic directions: one is parallel to c, the other normal to c (Fig. 2A).

Experiments with quartz.

1. The incident beam is not broken into diverging beams.
2. Rotating the quartz prism between crossed polars (experiment B.2) yields four extinctions, 90° apart. The experiment proves that light emerging from the crystal has two planes of polarization and that these are mutually perpendicular.
3. The planes of polarization are fixed with respect to crystallographic directions. Their orientation relative to the c-axis is the same as in calcite: One is parallel to c, the other normal to c (Fig. 2B).

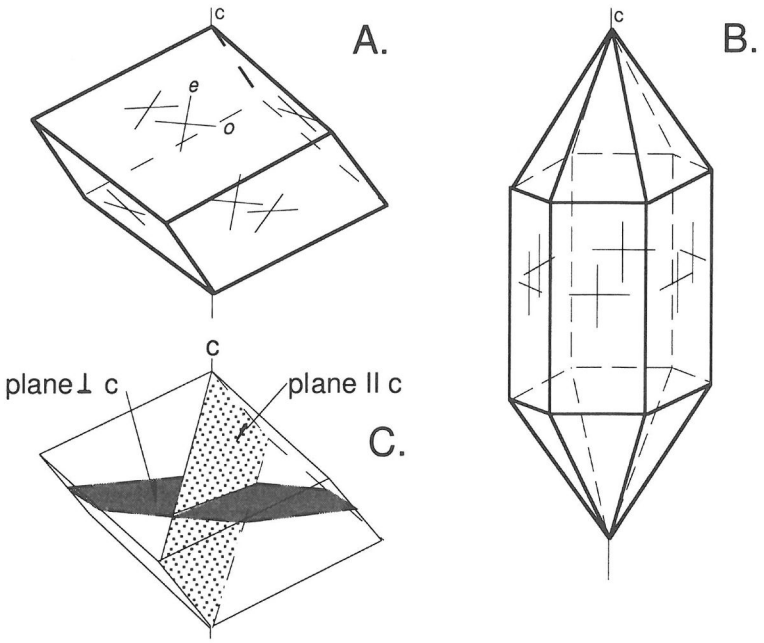

FIGURE 2. Directions of polarization (vibration directions) are marked as crosses on rhombohedral faces of calcite (A) and on prism faces of quartz (B). - Vibration directions of rays: e - extraordinary, o - ordinary. (C) Calcite rhombohedron with planes parallel to c and normal to c.

Explanations

The experimental results may be interpreted by using crystal structure drawings for calcite and quartz. In the calcite structure, carbonate ions form planar CO_3^{2-} triangles oriented normal to c (Fig. 3). Vibration directions (planes of polarization) were found to be parallel to c and normal to c (Figs. 2A, 2C). Consequently, waves traveling through calcite either oscillate parallel to the plane

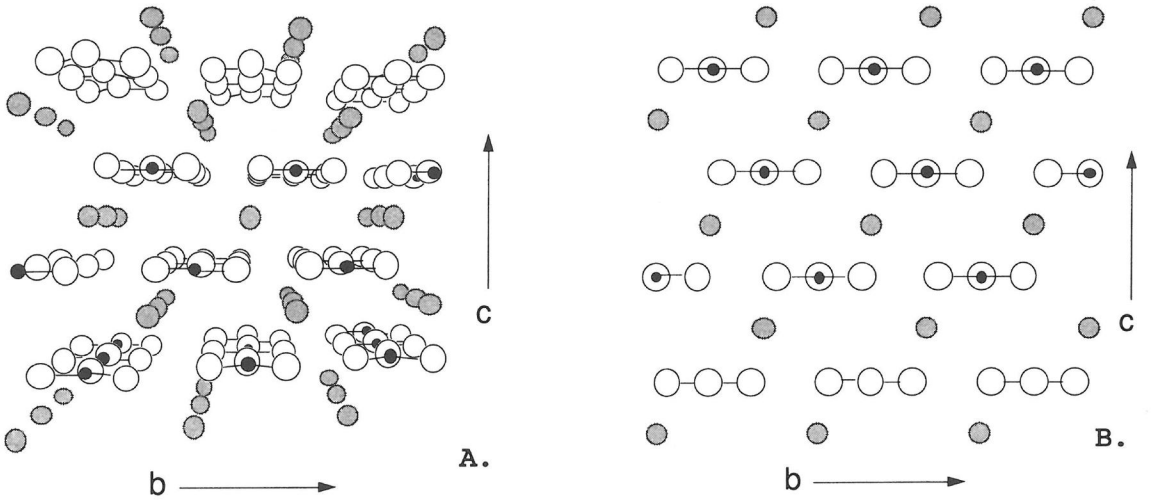

FIGURE 3. Crystal structure of calcite ($CaCO_3$). Spheres represent atoms: white - oxygen, black (small) - carbon, dark gray (large) - calcium. A. Perspective view along direction perpendicular to c (and b), [210]. B. Projection onto c, b plane, (100). CO_3^{2-} groups are arranged in planes normal to c. In directions perpendicular to c, oxygen atoms are densely packed, distances between them are relatively small. In directions parallel with c, distances between oxygen atoms are relatively large. (Figure generated with ATOMS).

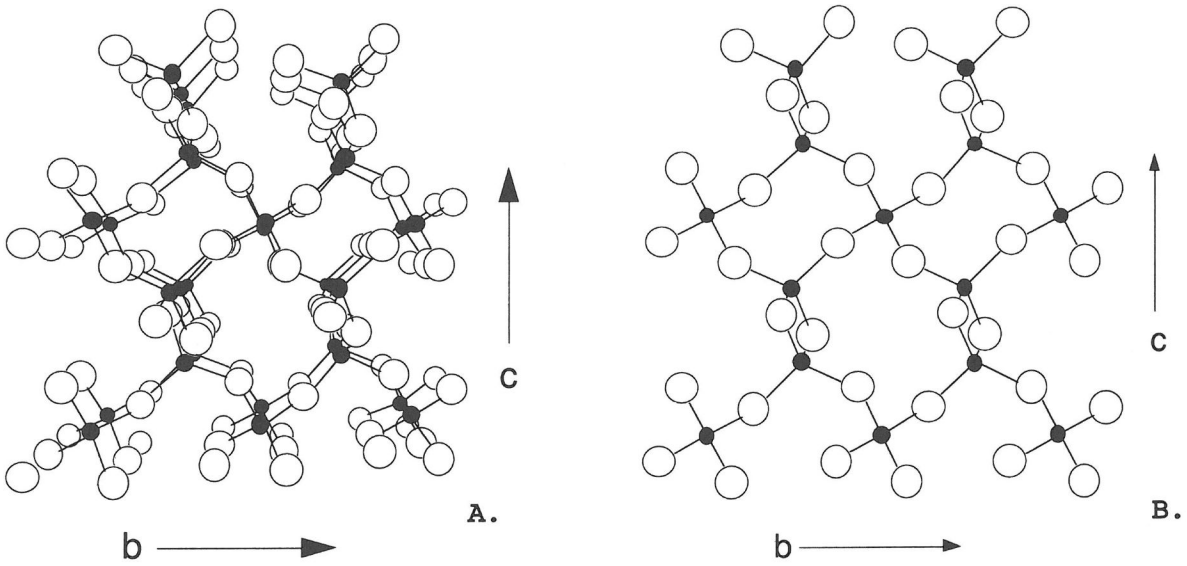

FIGURE 4. Crystal structure of quartz (SiO_2). Spheres represent atoms: white - oxygen, black - silicon. A. Perspective view along direction perpendicular to c (and b), [210]. B. Projection onto c, b plane, (100). - Oxygen atoms are distributed almost evenly in this structure, distances between them are almost the same in all direction. (Figure generated with ATOMS).

Of the atomic species in the structure, oxygen is the one with the highest degree of atomic polarizibility and, therefore, the one to most strongly interact with light. The influence of the other atoms is negligible. Light waves vibrating parallel to the CO_3^{2-} planes interact with many more oxygen atoms than waves normal to them. Interaction with (the highly polarizable) oxygen atoms slows light down. The lower the velocity, the higher the refraction. Thus, waves vibrating normal to c travel much slower and have a higher refractive index, than waves vibrating parallel to c. The crystal structure of calcite shows why the maximum double refraction in this mineral is very high ($|n_e-n_o| = 0.17$). In planes perpendicular to c, oxygens are much more closely packed than in planes parallel to c (Fig. 3).

Compare this to the crystal structure of quartz (Fig. 4): oxygens are almost evenly distributed in all directions. Therefore, it is plausible that light rays with their vibration directions perpendicular to c and parallel to c travel through the crystal at very similar speeds and thus differ very little with respect to their indices of refraction. The maximum difference of the refractive indices in quartz ($|n_e-n_o|=0.009$) is small in comparison to that observed in calcite.

PREPARATION OF HANDS-ON EXERCISES

Materials

Each student is given a kit with the following materials:

- 2 pieces of sheet polarizers (50mm x 50mm),
- quartz crystals with hexagonal prism, length > 4 cm, diameter > 2.5 cm,
- calcite rhombohedron, minimum length of smallest edge: 20mm (preferable are rhombs with distinctly different edge lengths, e.g., 20mm x 25mm x 30mm),
- muscovite sheet, minimum size: 30mm x 30mm, thickness to yield 2nd - 4th order interference colors,
- thin sections of rocks (e.g., basalt with phenocrysts, granite, dunite),
- felt tip pen (water soluble),
- interference color chart,
- quartz section cut normal to c (diameter: 25mm, thickness: 1mm)*)
- calcite section cut normal to c (diameter: 25mm, thickness: 1mm)*)

Expenses

Estimated expenses for 10 lab kits with the above items (without thin sections) range between $75 and $200. The cost depends largely on size and quality of the calcite and quartz samples. Suitable calcite and quartz samples are commercially available by the kilogram from mineral dealers. The clearer these minerals, the higher their price. Quartz and calcite crystals used in the exercises, need not be perfectly clear, but should be transparent enough to see through. Large polarizing sheets (ca. 300mm x 350mm) are available#) for less than $25. Cut these in squares of 50mm x 50mm, one sheet yields enough polarizers for 20 kits.

Time

The experiments below are easily completed within one and a half hours.

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*) If a microsaw is available, such sections are readily cut from ordinary specimen of calcite rhombs and quartz prisms. These sections need not be exactly normal to c. For the exercises, cuts approx. perpendicular to c will do.

#) e.g. Edmund Scientific, TECHSUP@EDSCI.COM.

HANDS-ON EXERCISES

A. Experiments with calcite

A1. Separation of images.

a) Place a calcite rhomb on a piece of paper (the paper and rhombohedral face are parallel). View a dot (or another object on the paper) through the calcite rhomb.
b) Note the horizontal and vertical separation of the two images. Use a ruler to measure the distance between the images.

A2. Effect of crystal thickness on separation of images.

a) Vary experiment (A1a) by using rhombs of different thickness. The point is to observe how the spacing of the images changes with crystal thickness.
b) Compare the horizontal separation of the images for different rhombs.

A3. Distinction between ordinary and extraordinary image.

a) Observe the images of the dot, while rotating the calcite about an axis normal to both the paper plane and the rhombohedral face.
b) Which of the two images is 'ordinary' (i.e. obeys Snell's law)? Which is 'extraordinary' (i.e. does not obey Snell's Law)? If it is not immediately obvious, how to recognize the 'ordinary' image, imagine the calcite rhomb replaced by a glass plate of similar thickness. You would then only see one dot, i.e., you would see an 'ordinary image' of the dot. Would you expect this (image of the) dot, upon rotation of the glass plate, to stay in place or to move in a circle?

A4. Polarization.

a) View an object (dot etc.) on a piece of paper through the calcite rhomb. Place a polarizing filter on top of the upper rhombohedral face. Observe the two images. Slowly rotate the polarizer one full circle about an axis normal to the paper (only the polarizer is rotated, not the crystal). What happens to the images?
b) Start with the extinction of the one image. By how many degrees do you have to turn the polarizer to make the other image disappear? Referring to rays instead of images, the question is: by how many degrees do the polarization directions of the emerging two rays differ?"

A5. Directions of vibration and c-axis. The images are formed by rays traveling through the rhomb. Extinction of images by the polarizer implies blocking of the image-forming rays (and, therewith, light waves) and proves that they are linearly polarized by the crystal. Blocking of rays (images) occurs when the direction of polarization (or direction of vibration) in the crystal is perpendicular to that of the polarizer.

a) Use the same set-up as in experiment A4. Turn the crystal back and forth to find the stationary (the ordinary) image and then bring it to extinction by the polarizer. On the upper face of the rhomb draw (use felt tip pen) a straight line for the ordinary, vibration direction in the crystal. Label it "o" for ordinary. (Remember: extinction occurs when vibration direction in the crystal and the direction of polarization in the polarizer are perpendicular to each other.)
b) Turn the polarizer until the other image (formed by the extraordinary ray) disappears. Plot a straight line for the extraordinary direction of vibration. Label it "e" for extraordinary.

c) Repeat experiment A5a and A5b for all the other faces of the rhomb.
d) Locate the c-axis in the calcite rhomb (see Fig. 2C). How are the vibration directions and c-axis oriented with respect to each other? Compare with Fig. 2A.

A6. Alternative determination of directions of vibration. Place the calcite rhomb between two polars in crossed position and view it in transmitted light. One way of doing this is to hold the two polarizing filters in the same hand, for instance by placing one filter between index and thumb and the other between middle finger and ring finger. Hold the calcite rhomb in the other hand and stick it between the polars so that these loosely sandwich the crystal and thus are parallel with two opposite rhomb faces. Use a window or a light bulb as light source.

a) Keep the filters fixed while turning the calcite rhomb about the axis normal to the polarizers and normal to the rhombohedral face parallel with them. Find the position of minimum light transmission. In this position vibration directions in the crystal are parallel and perpendicular to the directions of polarization of the crossed polars. Mark the directions of vibration on the rhomb faces (for instance with a felt tip pen).
b) Repeat experiment A6a for the other two sets of rhomb faces.
c) How are vibration directions and c-axis oriented with respect to each other? Compare with Fig. 2A.

B. Experiments with quartz

B1. Separation of images? Place a quartz crystal on a piece of paper (prism face and paper plane parallel). View a dot through the quartz prism.

B2. Polarization. Redo experiment A6 using a quartz prism. Place the polarizing filters in crossed position on either side of opposite prism faces. View the prism against the light. Keep the filters fixed while turning the quartz crystal about the axis normal to the polarizers and normal to the prism face. Find the position of minimum light transmission and mark the directions of vibration on all the prism faces. How are the vibration directions oriented with respect to the long axis of the prism (i.e. with respect to the c-axis)? Compare with Fig. 2B.

C. Experiments with muscovite

C1. Polarization. _Place a cleavage piece of muscovite between two polarizing filters in crossed position and view it against the light. Keep the filters fixed and rotate the muscovite piece about the axis normal to the polarizers and normal to the cleavage plane. Find the position of minimum light transmission and mark the directions of vibration on the cleavage face.

How are the vibration directions oriented with respect to the planar cleavage (and normal to the planar cleavage)? Which colors are seen in random positions?

C2. Interference color changes with variation of crystal thickness. Cleave the muscovite into thinner sheets. Observe the change in interference colors with changes in thickness. Try to locate the interference colors on an interference color chart.

C3. Interference colors and orientation. Hold the piece of muscovite in a position where cleavage plane and crossed polarizers are normal to the imagined line between the eye of the observer and the light source. Tilt the sheet against this line and observe the interference color. If the light traverses the muscovite at an angle to cleavage, it passes through more crystal. How do the increased angle and thickness affect the interference colors?

D. Observation of interference figures without microscope

Usually, interference figures of crystals are viewed in the polarizing microscope only. In principle, however, observing interference figures is quite simple and does not require a microscope. All one needs is a transparent mineral in form of a thin, sufficiently large[+)] plate or section, and two polarizers. A daylight light source (a window) is best, but a light bulb will do as well.

Interference figures are not images of the object, but represent interference patterns formed by light rays after passage through an anisotropic medium. These patterns become visible when a thin transparent crystal plate is placed between crossed polars and viewed against the light in such a way that the rays passing through the crystal form a large cone with a wide angular aperture (Fig. 5). This implies that the crystal is examined in many different directions at the same time. In other words, the bundle of light rays passing through the crystal is strongly divergent. This form of observation is called conoscopic.

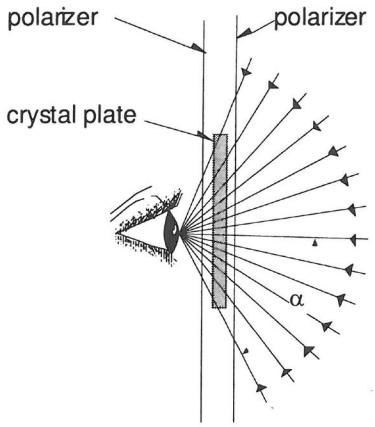

FIGURE 5. Macroscopic observation of interference figures. a - distance between eye and object, α - angular aperture.

At a larger distance to the eye (e.g., 20cm), the crystal plate appears with an overall homogenous interference color. This form of observation is called orthoscopic and implies that rays passing through the object are parallel, or almost parallel like in A, Fig. 6: either of the outer boundary rays, which pass at the very edge of the crystal, form a small angle with the central ray. This angle will become smaller yet if the distance between the eye is increased beyond position A, i.e. the rays will become 'more parallel'. Compare this with the opposite situation , the conoscopic passage of rays, where the same crystal plate is placed very close to the eye (Figure 6) and where the angle is thus very large between outer boundary rays and central ray.

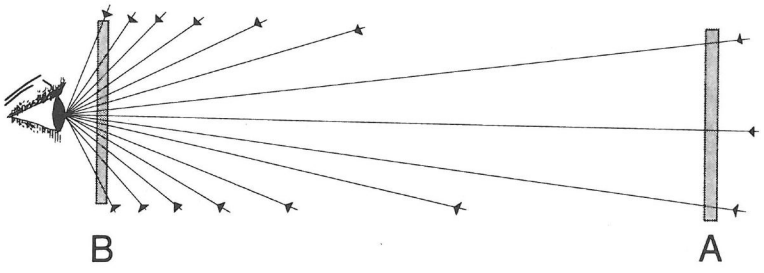

FIGURE 6. Crystal plate at different distances from the eye. A. Large distance. The angle between boundary rays is very small (small angular aperture), i.e. rays are almost parallel: orthoscopic passage of rays. B. Short distance. The angle between boundary rays is very large (large angular aperture): conoscopic passage of rays.

[+)] diameter > 2cm

By diminishing the distance between object and eye, it is possible to zoom from orthoscopic to conoscopic observation. Fig. 6 illustrates this transition: First, in position A, the bundle of rays through the crystal plate is almost parallel, and the angular aperture is very small (orthoscopic observation). With decreasing distance to the eye, rays through the crystal become more divergent, the angular aperture increases. Finally, in position B, the bundle of rays is strongly divergent, and the angular aperture very large (conoscopic observation).

D.1 Experiment with a muscovite cleavage sheet. Place a thin cleavage piece of muscovite between crossed polars. Hold it against the light. Bring it as closely to your eye as possible (Fig. 5, Fig. 6B). Note the dark bands (isogyres) and the curved color bands (isochromes).

D.2 Experiment with sections of quartz and calcite. Place an axial quartz section[§] between crossed polars and observe the interference figure. Note the dark cross (isogyres) and the circular color bands (isochromes). The interference figure observed is a uniaxial figure from a section cut normal to c.

Note that interference figures show interference colors of increasing order from the center outward. These interference colors follow the same sequence as in the interference color chart.

Redo the experiment with the calcite section.

E. Macroscopic observation of thin sections between crossed polars

Observe interference colors of mineral grains in thin sections of dunite, granite, and basalt. Try to locate these colors on the interference color chart.

Explanations

A structural interpretation of double refraction is given above (see explanations to lecture demonstrations). Here, a few additional comments on results, which at first, may appear puzzling.

Experiments A, B and C with calcite, quartz and muscovite show that all three minerals polarize light in two directions. In terms of light waves, this implies that an incident wave splits into two waves, which vibrate in mutually perpendicular planes and travel through the crystal at different speeds. This difference in speed and, thus, in refractive index is easier to explain in calcite than in the other two minerals.

Why is a separation of images/rays observed in the experiments with calcite (A1-A6), but not in the experiments with quartz (B1, B2) and muscovite (C1, C2)? In principle, separation of rays occurs in all of these minerals unless the incident ray is parallel to one of the "principal" vibration directions. In the above experiments with calcite, the incident ray is normal to the rhombohedral face, i.e., it is <u>not</u> parallel to one of the principal vibration directions. Therefore, one can observe a separation of the emerging rays and, correspondingly, a separation of images. In the experiments with quartz and muscovite the incident ray is parallel to one of the principal vibration directions, therefore there is only one ray emerges and no separation is observed.

Why does muscovite show distinct interference colors between crossed polars while calcite and quartz appear white? All three minerals do show interference colors between crossed polars. Yet, not all interference colors are colors in the ordinary sense of the word. - With increasing thickness, crystals produce interference colors of increasing order. Only interference colors of lower order (I-III) show distinct colors (see interference color chart). With increasing order,

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§ An axial section is a section cut normal to c. Recommended thickness: ca. 1 mm

interference colors become paler and, finally, merge into an "high order white", which is indistinguishable from ordinary white light.

Most minerals show distinct interference colors in thin crystal plates (10 μm to 100 μm thick). Muscovite cleaves readily into sheets of such thickness, and, therefore, shows distinct interference colors (see experiment C1-C3). This is not so with the calcite and quartz crystals used in the experiment. Their edge length and diameter measure several centimeters. These specimens are comparatively thick and, thus, yield interference colors of extremely high order (undifferentiated white). This is the reason why they appear colorless.

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