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# Analysis of Optical Properties and Image Patterns Due to Twinning in Calcite Crystals

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#### Abstract

The anomalous birefringent behavior and image patterns of select Calcite crystals were studied to determine whether its cause is trirefringence or mechanical deformation twinning. Calcite is a material known for its strong birefringence which gives rise to substantial double refraction, the property of a material to bend light into two separate beams due to an anisotropic index of refraction. Calcite samples have been studied that clearly produce multiple images beyond that of typical birefringence. The mechanism by which these image patterns are produced was studied to test a model describing this behavior. Image patterns and twin law are characterized for each sample indicating that the model adequately describes behavior for one twin plane but does not for two twin planes.

#### Keywords: Calcite, Twinning, Optics

### 1. Background

The Iceland Spar variety of calcite, also known as optical calcite, is a calcium carbonate mineral known for its strong birefringence. Birefringence is a property of an anisotropic material which has two indices of refraction. Incident light will refract at two angles as it passes through an anisotropic material depending on its angle incidence and polarization. For uniaxial materials, such as calcite, there is one optic axis for which incident light parallel does not undergo double refraction. Two separate rays are produced if light is not traveling parallel to the optic axis and two distinct images can be seen when looking through birefringent materials. The separate rays are called the ordinary ray (O-ray) and extraordinary ray (E-ray) which arise from corresponding ordinary and extraordinary indices. The measure of birefringence is the difference between the ordinary index and the extraordinary indices. The value for calcite is negative which means calcite is uniaxial negative<sup>1</sup>. Since the ordinary index is larger, it is the slow ray and the extraordinary index is the fast ray<sup>2</sup>. The O-ray and the E-ray are polarized at 90° angles to each other.

Optical calcite can be found as natural rhombohedral crystals which readily display double refraction. A set of samples were studied which do not exhibit typical optical behavior for calcite. These samples produce more than two images. The phenomenon can be seen easily when shining a laser through the face of a sample and projecting the image onto a surface.

Four samples were studied, three of which show an atypical image pattern. The fourth sample is typical and used for comparison of characteristic properties in the set of samples. An example of the atypical image pattern is seen (Figure 1) below. In this sample 12 images are present.



Figure 1. An atypical image pattern is seen for sample C12A. The photograph was taken using a DSLR camera looking at an approximate point source of light. The center image accounts for 4 of the images despite appearing as one image. Notice the alignment of the spectrum.

It was initially thought that the behavior of light in the atypical samples was due to trirefringence or multifringence. Trirefringence, or biaxial birefringence, is not subsequently suspected because calcite does not possess a crystal structure which corresponds to biaxial birefringence<sup>1 2</sup>. Trirefringence also does not mean a material will produce more than two images, but the two images are both E-rays and are distinguished as a fast and slow ray<sup>2</sup>.

A possible answer to why the atypical samples produce such image patterns is that the behavior is due to twinning in the crystals. Twinning is a common occurrence in many crystals where multiple crystal lattices grow into one another in a symmetrical fashion. This behavior was taken advantage of to produce a calcite gemstone, residing in the Harvard Mineralogical Museum, which was cut from a twinned crystal whose facet yielded a large number of images<sup>3</sup>. The optics involved with this facet is diagrammed in Figure 4 within the article<sup>3</sup> with a single incoming ray entering the facet and 8 rays exiting. Half of these are O-rays and the other half are E-rays.

The behavior seen in atypical samples is thought to be due to the interaction of light at the twinning interface between two crystals. The twinning interface or twin plane appears to have the effect of doubling the amount of rays as they pass through it. This is believed to be a consequence of the optic axis of each crystal being at a different orientation. The mechanism here was studied and a model was hypothesized and tested to explain the phenomena for the case of one twin plain. The model is referred to as the calcite twin refraction (CTR) model which makes predictions about the optics for rays interacting with a single twin plane. For each sample, the following were measured: image pattern, produced image count, number of O-rays and E-rays, polarization angle difference, visible twin planes, and twin law. Discrepancies between the CTR model and what is seen for atypical sample with two twin planes were analyzed.

# 2. Method

For each sample, the image patterns and twin planes were characterized. Four samples were characterized which have the following names: C2A, C4A, C12A, and C12B. All samples are a single rhombohedral crystal. C2A is a typical sample with the rest being atypical. Image patterns were produced by sending a laser beam through a particular cleavage plane, or crystal face, and projecting the image pattern onto a surface. The laser was approximately 5mW of wavelength 405nm. An aperture is used to reduce the glare of the laser which insures that the image pattern contrasts the background surface. The projection surface was an area of paper of approximately 3600 cm<sup>2</sup>. Because of the brightener in the paper, an arrangement of blue dots on the projection surface is observed due to fluorescence. The optical staging used is diagramed (Figure 2) below



Figure 2. Optical staging for analyzing the image patterns. Location 1 is the 405nm laser. Location 2 is an iris aperture. Location 3 is the sample. Location 4 is the 360 degree polarizer. Location 5 is the projection surface.

In the atypical samples, a majority of the dots in the image pattern diverge as the distance between the sample and the projection surface increases. Image separation is normally only dependent upon the incident angle and crystal thickness, but for typical samples it depends also upon the distance to the projection surface. Once the diverging behavior of the image pattern is taken into account the image pattern can be formed.

### 2.1. Characterizing The Image Pattern

The properties of the image pattern in the atypical samples are dependent on the incident crystal face. Even though there are three optically distinct crystal positions, only the position which yielded the apparent most number of images in the image patterns were used. The exact number of images in the image pattern was counted for each sample. The light emitted by the laser is polarized and therefore needs to be rotated about the optical axis of the staging system such that the light projected on the surface is distributed evenly amongst all produced images.

Seen before in the image (Figure 1), the second row of lights makes it appear that there are only nine images. However, the use of the polarizer in the optical staging reveals that there are indeed two dots. When rotating the polarizer to extinguish images some always persist. Since the light in these images should be polarized<sup>2</sup>, there should be two dots. Further evidence which supports this is shown below (Figure 3) which shows an image pattern for C12A. There are two sets of images for which all of the images in the set are extinguished at some polarization angle.



Figure 3. The distinct sets of images are shown in green and red in the false color image with each being numbered (Left). The green set extinguishes at a polarization angle that is different from that of the red set. Pairs (4, 5), (6, 7),

and (8, 9) appear to overlap and merge into a single image. The original image before enhancement (Right). Notice the center image appears to be merged into one completely.

### 2.1.1. sets of o-rays and e-rays

The number of O-rays and E-rays may be counted from the image as the number in a particular set. Whether or not a particular set was of fast rays or slow rays was not determined and it is assumed that the number of O-rays in a set equals the number of E-rays. There is a pair of O-ray and E-ray sets for each distinct position of incidence.

Once the set of rays are distinguished, the polarization angle difference may be found. The polarization angle is the angle at which a set of images is extinguished completely. Since there are two sets of images two distinct polarization angles corresponding to each set was found. The incident laser light was polarized and establishes  $0^{\circ}$  on the polarizer. Once we find those two angles we can calculate the difference as the polarization angle difference which is expected to be  $90^{\circ}$ .

### 2.2. Twin Planes

Before any measurements can be taken on twin planes, they first have to be identified. Measurements for twin planes were done for all the atypical samples and were not applicable to sample C2A because it contains no twin planes. Twin planes were recognized by looking for thin sheets, or lamellae inside samples along with thin lines/cracks on cleavage planes. The thin lines occur at the intersection between lamellae and a crystal face. The photograph (Figure 4) below is a sample highlighting twinning as it appears in sample C12B.



Figure 4. Twinning in samples. A photograph of sample C12B with a side (left) photograph indicating features. Location A corresponds to the red line which highlights a lamellae line, or thin crack. The dashed blue and orange shapes indicate the lamellae inside the crystal. The dashed green line indicates where they intersect which has its own crack within the crystal. Note the color bands which are thought to be interference bands.

The thin lines of concern are those that aren't parallel to the edge of a cleavage plane hereafter called lamellae lines. Lamellae cleavage planes will be the planes of the crystal which contain the lamellae lines. Several lamellae lines may be visible on a sample and some parallel to each other. The distinct orientations of lamellae lines correspond to distinct twin planes in a sample. For each sample the twin planes are identified and the obtuse angle, the twin angle, between the lamellae lines and the edge of the lamellae cleavage plane will be measured. The twin angle will then

be used to determine the twin plane's associated twin law<sup>4</sup> in analysis. Two methods were developed for measuring the twin angle: Point to point (PPP) and photographic projection (PGP).

# 2.2.1. point to point method

The PPP method requires the use of a pencil/pen, paper, and protractor. A sample is placed with the face that is the lamellae cleavage plane facing flat on the paper. The edges of this face are traced out as illustrated (Figure 5) below. On the lamellae cleavage plane that is face down, the endpoints of a lamellae line are plotted and then connected with a straight edge so that the resulting line passes through both lines on the trace that it intersects. Both obtuse opposite pairwise angles for the resulting line are measured. The average of these two angles is calculated as the twin angle. This process was repeated for each twin plane in all of the atypical samples.



Figure 5. Enhanced photograph of the PPP method for sample C4A and with the traced outline of the lamellae cleavage plane. Location A is the angle measured along with its opposite pairwise angle. The intersection line is highlighted by the dashed green and the dashed red line is the corresponding lamellae line used for the trace.

# 2.2.2. photographic projection method

The PGP method uses a DLSR camera to perform a similar process as the PPP method. A photograph of the lamellae cleavage plane is taken and then imaging techniques are used to determine the twin angle. A tripod and level were used to align the aim of the camera to be parallel as possible to the normal of the lamellae cleavage plane. This insures that the distortion do to perspective is minimized. As with the PPP method, a line is drawn digitally from one end of a lamellae line to another. Another line is drawn that is along both distinct edges. The twin angle can then be measured by using a clear protractor and placing it on the used display monitor. The PGP method being used on sample C12A can be seen (Figure 5). Using the PGP method, multiple lines (seen as yellow) can be obtained which provides more data points to determine the twin angle. The number traced lamellae lines depends on the number of lamellae lines in the sample and the number that are of good quality for a trace. Sample C12B only has two lamellae lines to project from. For each twin plane, the average angle found is the twin angle. This process is repeated for each sample.



Figure 7. PGP Method for sample C12A. The yellow lines are the line projections. The obtuse angle between the yellow lines and the horizontal grey line is the twin angle. Many thin cracks can be seen which appear to fork due to double refraction and are indicative of twinning.

# 3. Data And Results

Table 1 shows the data for the characterization of the image patterns for each of the samples. A photograph of the image pattern that produced the most images is included in the table.

Sample		C2A	C4A	C12A	C12B
Image Pattern		*	* * *		••••
Image Count		2	4	12	12
O-Rays		1	2	6	6
E-Rays		1	2	6	6
Polarization Angles	Set 1	$0\pm0.5$ $^{\mathrm{o}}$	$2\pm0.5$ °	$359\pm0.5^{\circ}$	$342\pm0.5^{\:o}$
	Set 2	$92\pm0.5$ $^{\mathrm{o}}$	$96\pm0.5$ $^{\rm o}$	$89\pm0.5^{\mathrm{o}}$	$74\pm0.5^{\rm o}$
Polarization Angle Difference		$92\pm0.7^{\:o}$	$94\pm0.7$ °	$90\pm0.7$ °	$92\pm0.7$ °

Table 1. data for characterization of image patterns in samples

# 3.1. Characteristic Description Of Each Sample

Each sample inspected for other features which were noted alongside the characterization data. The image pattern that is described refers the data on the particular incident cleavage plane which produces the most images.

# 3.1.1. C2A description

The sample is typical, and produces two images. The image pattern contains one O-ray and one E-ray. Twinning is not present in the sample.

# 3.1.2. C4A description

The sample is atypical and produces four or two images depending on the incident cleavage plane. The image pattern contains two O-rays and two E-rays. There is one distinct set of lamellae lines of which there are many lines. A single twin plane is present.

# 3.1.3. C12A description

The sample is atypical and produces 12 or 4 images depending on the incident cleavage plane. The image pattern contains 6 O-rays and 6 E-rays. There are two distinct set of lamellae lines. Two twin planes are present. The sample is quite clear and prismatic. All of the surfaces of the sample are polished.

# 3.1.4. C12B description

The sample is atypical and produces 12, 4, and 2 images depending on the incident cleavage plane and orientation of the plane that light passes through. The sample possesses only two lamellae lines, one for each of the two twin planes present. One of the twin planes spans the length inside the sample and the other is only in a small section toward the end of the sample. As seen before (Figure 4), the lengthwise twin plane shows bands of color which increase frequency as the sample is turned about an axis perpendicular to the lamellae cleavage plane.

# 3.2. Characterization Of Twinning

Table 2 and Table 3 show the data for the separate measurements of twin angle in degrees and concluded twin law for each sample. A twin laws associated twin angle that was closest to the one measured is indicated. C4A does not have a second twin plane so some entries are not applicable to it.

Sa	mple	C4A	C12A	C12B
Visible Twin Planes		1	2	2
Twin Angle	1	$142\pm0.5^{\mathrm{o}}$	$142\pm0.5^{\mathrm{o}}$	$140\pm0.5^{\:\mathrm{o}}$
	2	n/a	$140\pm0.8^{\mathrm{o}}$	$139\pm0.5^{\circ}$
Twin Law	1	{01.8}	{01.8}	{01.8}
	2	n/a	{01.8}	{01.8}

Table 2. twinning data from PPP method

Table 3. twinning data from PGP method

Sample		C4A	C12A	C12B
Visible Twin Planes		1	2	2
Twin Angle	1	$141.3 \pm 0.5^{\circ}$	$141.5 \pm 0.5^{\circ}$	$141.0 \pm 0.5^{\circ}$
	2	n/a	$139.8 \pm 0.5$ °	$141.8 \pm 0.5^{\circ}$
Twin Law	1	{01.8}	{01.8}	{01.8}
	2	n/a	{01.8}	{01.8}

# 4. Analysis

The Calcite twin refraction model (CTR) was developed using the data collected in Table 1 and insight from the Ross calcite facet<sup>3</sup>. The CTR model aims to explain the interaction of an incident ray of light through calcite sample with a single twin plane. It is diagrammed below (Figure 7) with an incident ray of arbitrary polarization that will split into the two vector components of the polarization via double refraction and are the O-ray and E-ray components.



Figure 7. Ray diagram of the CTR model with a single twin plane. Each numbered stage of the diagram has an associated polarization diagram below of each ray involved. When a ray crosses a boundary double refraction occurs and polarization components are shown as dashed red or blue lines in the polarization diagram. There are two anisotropic mediums which have birefringence orientations and optic axes that are different in spatial orientation from one another.

The way that the polarization vector splits into components is dependent upon the orientation of the unit vectors associated with the O-ray and E-ray components. This orientation will be referred to as the orientation of the birefringence or birefringence orientation. According to the CTR model, each unique optic axis has an associated unique birefringence orientation. The incident ray is split twice due to the different birefringence orientations of the anisotropic mediums on each side of the twin plane.

# 4.1. Case Of One Twin Plane

Since calcite is uniaxial, light parallel to the optic axis does not experience birefringence. However, according to the CTR model there should be no way to send a straight beam of light through a sample with a twin plane and the light not experience birefringence since the path of the beam will always be anti-parallel to at least one of the optic axes. This is supported in samples when we make the light incident to the lamellae cleavage plane. In this scenario, the image pattern produced contains two images which do not diverge as is the case with C4A. At any other incidence, light is split into four images in this sample. This means that the optic axis of one of the crystals is likely to lie parallel to the twin plane itself.

### 4.2. Case Of Two Twin Planes

In the case of two twin planes we may attempt to apply the CTR model as if the light passes through an additional second twin plane. If we assume this, then after incident light passes through the first twin plane, four rays will have been produced. The light passes through a second twin plane and split according to a third unique birefringence orientation. In the two twin plane case, we would expect three unique birefringence orientations and two doublings of the incident ray by the CTR model. With four incident rays to the second twin plane we would expect 8 distinct rays with half being O-rays and the other half being E-rays. However, this is not what we see in the samples with two twin planes. There is some unknown difference between the paths of rays in Figure 7 and the paths of rays in samples C12A and C12B. While the CTR model predicts that eight images are produced in this case, twelve images are observed in two twin plane samples.

As with the two optic axes with one twin plane, for two twin planes there are allegedly three optic axes. A consequence of this is that even if incident light is parallel to one of these axes it will still always be at an angle to at least two optic axes. This means that we will get behavior similar to C4A if we send light into C12A or C12B parallel to one of the optic axes. Again, the optic axes seems to lie in the twin planes themselves because when light is parallel to a twin plane in C12A or C12B the image pattern produced matches C4A and also produces four images.

#### 4.3. Twin Law

A twin law indicates the geometric relation of the components of a twinned crystal specific to a given crystal system. Calcite has only four known twin laws<sup>2</sup> and each has a corresponding twin plane associated with a Miller index. Calcite has a hexagonal lattice defined by four unique axes and four indices which are together called a Miller-Bravais index given by { $h \ i \ l$ } where h,k,i, and l are the inverse of the intersection positions of the twin plane with the axes of a hexagonal coordinate system. Peter's syntax convention<sup>4</sup> will be used in which the index of *i* is omitted with a period since it is the sum of *h* and *l*. The twin law for all of the twin planes was found to be the Miller-Bravais index of {01.8} or equivalently {0118} which is a common type of twinning <sup>2</sup>. The twinning in the calcite facet literature also had a Miller-Bravais index {01.8}<sup>4</sup>. There are two relevant types of twinning calcite which can be applicable to the set of atypical samples: growth and mechanical deformation twinning. Growth twinning relies on the samples being multiple conjoined rhombs and is not the case with these samples. Mechanical deformation is described by internal lamellae and thin lines or cracks on the exterior of the crystal<sup>4</sup>. This description matches that of most of the samples, especially C4A.

#### 4.4. Generalizations

Twinning and birefringence can occur in many materials. From the CTR model, the notion of the orientation of the birefringence having consequence for optical behavior should apply to all anisotropic materials which exhibit a kind of twinning. The optical behavior and nature of the image patterns will not express birefringence as pronounced as calcite however.

Since the phenomena is primarily a consequence of the birefringence orientation and optic axis in the crystal, one might expect that placing two singular rhombs of calcite in contact face to face at in various arrangements would emulate the data seen in Table 1. For example, two samples like C4A placed in series in location 3 in Figure 2 accordingly may produce an image like that of C12A due to aligning two twin planes in series. Experimentation has been done on this set of samples in this way but only by inspection of samples.

Additionally, there are possible applications in synthetic anisotropic materials which fit the aforementioned criteria. Such synthetic crystals may be designed so that certain image patterns are formed for whatever means or optical devices.

#### 5. Conclusion

From comparison of results and predictions stemming from the CTR model, the multiple images in the atypical samples is due to the optical behavior of optic axes at different angles two one another inside of the samples. The

type of twinning for samples C4A, C12A, and C12B is concluded to be of Miller-Brevais index {01.8} and of the mechanical deformation type.

However, the CTR model is not satisfactory in describing how samples C12A and C12B produce 12 images due to two twin planes. Further developments on the model should assume that these samples are not demonstrating biaxial birefringence, or trirefringence. The crystal twinning in these samples may be because there are two crystals super imposed in one another or interpenetrated.

### 6. Future Work

There are many venues for future experimentation. A model should be developed using two uniaxial indicatrix to describe the paths of light through the samples. To assist with this, an X-ray crystal diffraction study of a slice of these samples would allow for the molecular structure of the calcite in these samples to be determined and to check the alignment of unit cells at lamellae lines and twin boundaries.

The phenomenon seen here is concluded to be dependent upon the orientation of optic axes in the samples. This implies that other samples may be oriented next to each and a laser beam sent through them in sequence to produce similar image patterns. Alternatively, different sample materials such as clear quartz crystals, could have a similar experimental procedure applied to them if samples of good quality are found containing suitable twin planes. Analogous results should be seen in other anisotropic materials.

Another experiment would be to test incident light different wavelengths to understand the interplay of the indices of refraction in these samples and the alignment of the spectrum in each image position in the image pattern shown in Figure 1.

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