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LB-881

# DETERMINATION OF ORIENTATION AND DEFORMATION OF GERMANIUM CRYSTALS


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Approved

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## Introduction

There is a need for a method which will supply crystallographic information as to the orientation and deformations of large single crystals of germanium. One technique is metallographically to polish the surfaces of the ingot and use etching solutions to check the absence of grain boundaries, and to use etch-pit methods to supply orientation information.[1] Although such methods are rapid and useful, a more direct method is described in this bulletin which uses back-reflection Laue x-ray diffraction techniques. The technique of taking pictures and interpreting patterns are described in this bulletin so that persons not trained in crystallography may have a ready reference to this method.

## General Discussion

Back-reflection Laue patterns are obtained by sending a beam of x-rays, comprising a wide range of wave-lengths (white x-ray radiation*), through a pinhole system and then through a punched hole in the center of a film. Diffracted back from a sample placed at a fixed distance from the film, the rays appear as spots on the film. The arrangement is illustrated in Fig. 1. The sample S is placed at a distance of 3.00 ± 0.05 cm from the film. The x-ray beam hits the sample and "reflections" occur off planes which can "reflect"**. For example, plane XY in Fig. 1 diffracts a spot at point L on the film. This diffraction can be treated as a light reflection, in that the angle of incidence equals the angle of reflection. Thus, the intersection of the normal (dotted line) to the plane XY locates a point, P, on the film. It is apparent, from the geometry, that it is possible to determine all such points from the Laue spots. As the normals lie closer to the center of the film than the Laue spots, and the film diameter is only 12.5 cm, it is customary to multiply the radial distance (CP) by ten. This spreads out the pattern, making measurements for orientation purposes more accurate. To facilitate this "projection", a special scale is prepared which can be laid on a Laue pattern so that the "projected" pattern can be determined directly from the ruled scale.

[1] C. S. Barrett, STRUCTURE OF METALS, McGraw-Hill (1943).

*White x-ray radiation is best obtained from a tungsten-target x-ray tube operated at about 40-60 kv; however, if such a tube is not available, any tube giving appreciable general radiation with or without characteristic radiation will serve (for example, a copper- or iron-target tube).

**Only planes which satisfy Bragg's Law "reflect". The geometric relations are explained in this bulletin; detailed description and use of Bragg's Law is adequately explained in references (1) and (5).

## Experimental Technique

A General Electric XRD X-Ray Diffraction Unit is used. It contains an x-ray tube which has two port holes for radiation emission, so that two experiments may be conducted at the same time. There are two camera tracks on the unit, which align the cameras with the x-ray tube. For the Back Reflection Laue method, the back-reflection cassette with pinhole system

(of 0.025 inch in diameter) and specimen mount is used. The specimen is mounted with a template 3.00 ± 0.05 cm from the film, and in a position which would fix its position definitely relative to the film. A position could be fixed by mounting a face parallel to the film, or the length parallel to the x-ray beam, and by making a fiducial mark on the film and sample to fix the position around the x-ray beam. An exposure of about one hour is made with a copper-target tube, or about 1½ hours with an iron-target tube, and the film is then developed.

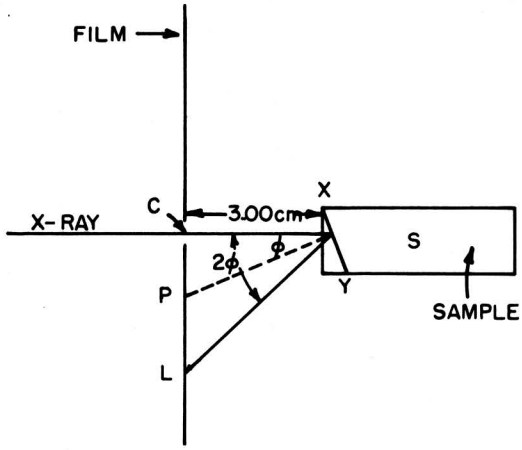


Fig. 1 – Geometrical relation of sample, film, and x-ray beam.

An easily made tracing of the Laue spots on translucent paper may be used for the projection. The scale is made by graduating the left side of a ruler in centimeters, the distance CL, from a central point C, and the right side is graduated so that for each graduation on the left side a corresponding mark is made on the right side such that the distance is 10XCP or 10 x 3 tan φ, where φ is the angle as shown in Fig. 1. Table I gives a tabulation of distances on the left side of ruler CL in centimeters and the corresponding lengths for marking off in centimeters on the right side, 10CP from C, in the preparation of such a ruler. Each graduation on the right side is given the same number as the corresponding length on the left side. Fig. 2 is an illustration of a ruler constructed by this method.

**Table I—Data for Construction of Graduated Scales (Rulers)**

| CL | 10CP | CL | 10CP | CL | 10CP |
|---|---|---|---|---|---|
| 0.1 | 0.50 | 2.4 | 10.53 | 4.7 | 16.44 |
| 0.2 | 1.00 | 2.5 | 10.86 | 4.8 | 16.63 |
| 0.3 | 1.49 | 2.6 | 11.20 | 4.9 | 16.80 |
| 0.4 | 1.99 | 2.7 | 11.52 | 5.0 | 16.98 |
| 0.5 | 2.48 | 2.8 | 11.93 | 5.1 | 17.15 |
| 0.6 | 2.97 | 2.9 | 12.13 | 5.2 | 17.32 |
| 0.7 | 3.45 | 3.0 | 12.43 | 5.3 | 17.50 |
| 0.8 | 3.93 | 3.1 | 12.71 | 5.4 | 17.65 |
| 0.9 | 4.40 | 3.2 | 12.99 | 5.5 | 17.81 |
| 1.0 | 4.87 | 3.3 | 13.27 | 5.6 | 17.97 |
| 1.1 | 5.33 | 3.4 | 13.53 | 5.7 | 18.11 |
| 1.2 | 5.78 | 3.5 | 13.80 | 5.8 | 18.26 |
| 1.3 | 6.22 | 3.6 | 14.05 | 5.9 | 18.41 |
| 1.4 | 6.66 | 3.7 | 14.30 | 6.0 | 18.54 |
| 1.5 | 7.08 | 3.8 | 14.54 | 6.1 | 18.68 |
| 1.6 | 7.50 | 3.9 | 14.77 | 6.2 | 18.81 |
| 1.7 | 7.91 | 4.0 | 15.00 | 6.3 | 18.94 |
| 1.8 | 8.31 | 4.1 | 15.22 | 6.4 | 19.08 |
| 1.9 | 8.70 | 4.2 | 15.44 | 6.5 | 19.20 |
| 2.0 | 9.09 | 4.3 | 15.65 | 6.6 | 19.32 |
| 2.1 | 9.45 | 4.4 | 15.85 | 6.7 | 19.45 |
| 2.2 | 9.82 | 4.5 | 16.06 | 6.8 | 19.56 |
| 2.3 | 10.17 | 4.6 | 16.25 | | |

To use the ruler, a tracing of the Laue pattern is pasted on the center of a sheet of drawing paper about 35 x 35 cm. The point C of the ruler is placed on the center of the traced Laue pattern, and for each Laue spot, as measured on the left side of the ruler, a corresponding mark is made on the paper using the right side of the ruler at an equivalent graduation mark. This procedure, when carried out for all Laue spots, will give a projection which has a simple geometric relation.

## Interpretation

In order to interpret Laue patterns some knowledge of crystallography is essential. The minimum of subject matter required is outlined below.

Germanium has the same intermeshed face-centered-cubic structure as diamond. The

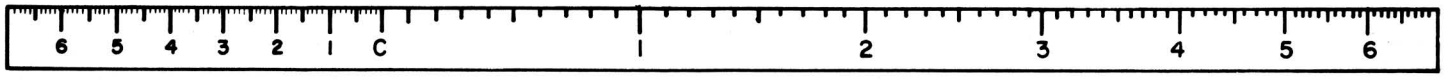


Fig. 2 – Scale for preparing projection of back reflection pattern.

smallest volume of this arrangement, which when repeated in space will give a diamond cubic lattice, is called a unit cell. The three edges of the unit cell designated as X, Y, and Z are used as a base for a system of notation for the planes of the crystal. These notations are called Miller indices and are universally used to designate planes. These indices are based on the intercepts of a plane with the three edges of the unit cell. The intercepts are measured in terms of the dimensions of the unit cell, which are unit distances along the three axes. For example, a plane that cuts the X axis at a distance from the origin equal to half the cell dimension is said to have the X intercept $\frac{1}{2}$; and if it cuts the Y and Z at one-half the cell dimension, then the Y and Z intercepts are $\frac{1}{2}$. If a plane is parallel to an axis, it intersects it at infinity. To determine the Miller indices of a plane, the following steps are necessary:

1. Find the intercepts on the three axes in multiples or fractions of the unit distance on each axis.

2. Take the reciprocals of these numbers.

3. Reduce to the three smallest integers having the same ratio, and enclose in parentheses (h k l), h being the integers for the X axis, k for the Y axis, and l for the Z axis.

Thus, the intercepts 1,1,1 have reciprocals 1,1,1 and indices (111); the intercepts 2,$\infty$,1 have reciprocals $\frac{1}{2}$,0,1 and indices (102). If a plane cuts any axis (for example, the X axis) on the negative side of the origin, the corresponding index will be negative and is indicated by placing a minus sign above the index: ($\bar{h}$ k l). The parentheses around the Miller indices signify a single plane, or set of parallel planes. Braces signify planes of a "form", that is, those planes which are equivalent in the crystal, such as the cube faces of a cubic crystal: $\{100\}$ = (100)+(010)+(001)+($\bar{1}$00)+(0$\bar{1}$0)+(00$\bar{1}$).

A direction in the lattice may also be expressed by a system of notation. The direction may be thought of as an extension of a line from the origin of the axes X, Y, and Z to any point in the lattice. The notation here is that the coordinates of a point on this line are expressed in the smallest whole integers. The indices of direction are written [u v w], where u is the X coordinate of a point on the line, v the Y coordinate, and w the Z coordinate when the point is chosen such that [u v w] are smallest whole integers.

For example, the X axis has indices [100], the Y axis [010], and the Z axis [001]; a face diagonal of the XY face of the unit cell has indices [110], and a body diagonal the indices [111]. Negative indices occur if any of the translations are in the negative directions of the axes, for example, the -X direction has indices [$\bar{1}$00]. It should be noted that reciprocals are not used in computing indices of a direction. In the cubic system a direction will always be perpendicular to a plane having the same indices. For example, the [111] direction is perpendicular to the (111) plane.

Other necessary concepts required for understanding Laue patterns are those of zones and zone axes. A zone axis in a crystal is a direction, the direction being formed by planes intersecting. All planes which intersect and have equivalent intersecting direction have the same zone axis, and all the planes having this axis belong to the same zone. On a Laue pattern, the spots which lie on a hyperbola belong to the same zone, and the zone axis is represented by the hyperbola. There is a simple relation between the indices of planes and zones which is useful in interpreting x-ray patterns; the plane (h k l) belongs to the zone [u v w] (i.e., parallel to [u v w] if hu + kv + lw = 0).

The Laue spots on a film give information only on the structure of the material where the x-rays hit, that is, a volume of about 0.025 inch diameter and a depth of about 1/32 inch. To obtain information on any other portion of the ingot, additional patterns must be obtained. To determine if a material is a single crystal, it is therefore necessary to check different sections of the sample to determine if the orientation is consistent for the crystal.

If a major axis is parallel to the x-ray beam, a symmetrical pattern will be obtained. For a four-fold symmetry the beam would be parallel to [100] axes; for a three-fold symmetry, parallel to [111]; and for a two-fold symmetry, parallel to [110]. In most cases a major axes will not be parallel to the beam. An unsymmetrical back-reflection Laue pattern of a quasi-symmetrical arrangement with spots laying on hyperbolas will be obtained.

**Table II—Angles between Planes of Forms $\{h_1k_1l_1\}$ and $\{h_2k_2l_2\}$ in the Cubic System[3]**

| $\{h_1k_1l_1\}$ | $\{h_2k_2l_2\}$ | | | | | | |
|---|---|---|---|---|---|---|---|
| 100 | 100 | 0° | 90° | | | | |
| | 110 | 45° | 90° | | | | |
| | 111 | 45°44' | | | | | |
| | 210 | 26°34' | 63°26' | 90° | | | |
| | 211 | 35°16' | 65°54' | | | | |
| | 221 | 48°11' | 70°32' | | | | |
| | 310 | 18°26' | 71°34' | 90° | | | |
| | 311 | 25°14' | 72°27' | | | | |
| | 320 | 33°41' | 56°19' | 90° | | | |
| | 321 | 36°43' | 57°42' | 74°30' | | | |
| 110 | 110 | 0° | 60° | 90° | | | |
| | 111 | 35°16' | 90° | | | | |
| | 210 | 18°26' | 50°46' | 71°34' | | | |
| | 211 | 30° | 54°44' | 73°13' | 90° | | |
| | 221 | 19°28' | 45° | 76°22' | 90° | | |
| | 310 | 26°34' | 47°52' | 63°26' | 77°5' | | |
| | 311 | 31°29' | 64°46' | 90° | | | |
| | 320 | 11°19' | 53°58' | 66°54' | 78°41' | | |
| | 321 | 19°6' | 40°54' | 55°28' | 67°48' | 79°6 | |
| 111 | 111 | 0° | 70°32' | | | | |
| | 210 | 39°14' | 75°2' | | | | |
| | 211 | 19°28' | 61°52' | 90° | | | |
| | 221 | 15°48' | 54°44' | 78°54' | | | |
| | 310 | 43°5' | 68°35' | | | | |
| | 311 | 29°30' | 58°31' | 79°58' | | | |
| | 320 | 61°17' | 71°19' | | | | |
| | 321 | 22°12' | 51°53' | 72°1' | 90° | | |
| 210 | 210 | 0° | 36°52' | 53°8' | 66°25' | 78°28' | 90° |
| | 211 | 24°6' | 43°5' | 56°47' | 79°29' | 90° | |
| | 221 | 26°34' | 41°49' | 53°24' | 63°26' | 72°39' | 90° |
| | 310 | 8°8' | 58°3' | 45° | 64°54' | 73°34' | |
| | 311 | 19°17' | 47°36' | 66°8' | 82°15' | | |
| | 320 | 7°7' | 29°45' | 41°55' | 60°15' | 68°9' | 75°38' 82°53' |
| | 321 | 17°1' | 33°13' | 53°18' | 61°26' | 70°13' | (83°8' 90° |
| 211 | 211 | 0° | 33°33' | 48°11' | 60° | 70°32' | 80°24' |
| | 221 | 17°43' | 35°16' | 47°7' | 65°54' | 74°12' | 82°12' |
| | 310 | 25°21' | 49°48' | 58°55' | 75°2' | 82°35' | |
| | 311 | 19°8' | 42°24' | 60°30' | 75°45' | 90° | |
| | 320 | 25°9' | 37°37' | 55°33' | 63°5' | 83°30' | |
| | 321 | 10°54' | 29°12' | 40°12' | 49°6' | 56°56' | |
| | | 70°54' | 77°24' | 83°44' | 90° | | |

[3] R. M. Bozorth, "The Orientations of Crystals in Electrodeposited Metals", *Phys. Rev.*, Vol. 26, pp. 390–400, (1925).

Table II— Cont.

| $\{h_1k_1l_1\}$ | $\{h_2k_2l_2\}$ | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 221 | 221 | 0° | 27°16' | 38°57' | 63°37' | 83°37' | 90° | |
| | 310 | 32°31' | 42°27' | 58°12' | 65°4' | 83°57' | | |
| | 311 | 25°14' | 45°17' | 59°50' | 72°27' | 84°14' | | |
| | 320 | 22°24' | 42°18' | 49°40' | 68°18' | 79°21' | 84°42' | |
| | 321 | 11°29' | 27°1' | 36°42' | 57°41' | 63°33' | 74°30' | |
| | | 79°44' | 84°53' | | | | | |
| 310 | 310 | 0° | 25°51' | 36°52' | 53°8' | 72°33' | 84°16' | |
| | 311 | 17°33' | 40°17' | 55°6' | 67°35' | 79°1' | 90° | |
| | 320 | 15°15' | 37°52' | 52°8' | 74°45' | 84°58' | | |
| | 321 | 21°37' | 32°19' | 40°29' | 47°28' | 53°44' | 59°32' | |
| | | 65° | 75°19' | 85°9' | 90° | | | |
| 311 | 311 | 0° | 35°6' | 50°29' | 62°58' | 84°47' | | |
| | 320 | 23°6' | 41°11' | 54°10' | 65°17' | 75°28' | 85°12' | |
| | 321 | 14°46' | 36°19' | 49°52' | 61°5' | 71°12' | 80°44' | |
| 320 | 320 | 0° | 22°37' | 46°11' | 62°31' | 67°23' | 72°5' | 90° |
| | 321 | 15°30' | 27°11' | 35°23' | 48°9' | 53°37' | 58°45' | 63°36' |
| | | 72°45' | 77°9' | 85°45' | 90° | | | |
| 321 | 321 | 0° | 21°47' | 31° | 38°13' | 44°25' | 50° | 60° |
| | | 64°37' | 69°4' | 73°24' | 81°47' | 85°54' | | |

Table III—Reflecting Planes of Diamond Cubic Structure

| $h^2+k^2+l^2$ | hkl | $h^2+k^2+l^2$ | hkl |
|---|---|---|---|
| 3 | 111 | 51 | 711; 551 |
| 8 | 220 | 56 | 642 |
| 11 | 311 | 59 | 731; 553 |
| 16 | 400 | 64 | 800 |
| 19 | 331 | 67 | 733 |
| 24 | 422 | 72 | 822 |
| 27 | 511; 333 | 75 | 751; 555 |
| 32 | 440 | 80 | 840 |
| 35 | 531 | 83 | 911; 753 |
| 40 | 620 | 88 | 664 |
| 43 | 533 | 91 | 931 |
| 48 | 444 | 96 | 844 |
| | | 99 | 933; 711; 755 |

On every Laue pattern of a cubic crystal there is a prominent row of spots from the zones [100], [110], and [111], and less prominent ones from the zone [113]. Spots at the intersections of these principal hyperbolas are "reflections" from the planes common to the principal zones in the crystal. They have low indices, high intensity, and are somewhat isolated from their neighbors on the hyperbola.

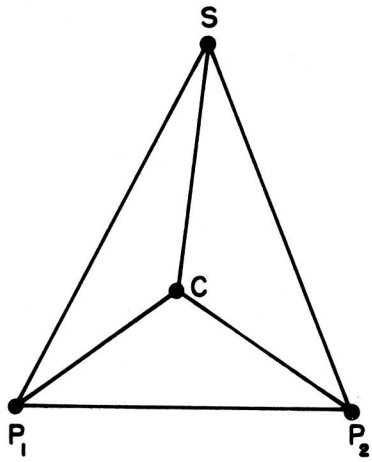


Fig. 3 - Three-dimensional representation $P_1CP_2$ in plane of projection.

The angles between planes represented by prominent spots are used to index the pattern. The angles are determined from a projection made of the Laue pattern by simple geometric relations. For example, in Fig. 3, if C is the center of the projection, and $P_1$ and $P_2$ are projections of prominent spots, and S is the sample now located at the projected distance of 30.0 cm, then the angle $P_1SP_2$ is the angle between the planes reflecting the spots $P_1$ and $P_2$. To determine this angle accurately, $P_1C$ and $P_2C$ are measured in centimeters, and the distance $P_1S$ and $P_2S$ are easily calculated because $P_1CS$ and $P_2CS$ are right triangles. The angle $P_1SP_2$ is then calculated from the geometric relation for a plane triangle, $a^2 = b^2 + c^2 - 2bc \cos A$; that is,

$$\overline{P_1P_2}^2 = \overline{P_1S}^2 + \overline{P_2S}^2 - 2(P_1S)(P_2S)\cos P_1SP_2$$

Having calculated the angles between the most prominent spots on the projection, one need only consult Table II to determine which planes have their angles between them equivalent to the calculated angles obtained. The indexing may be checked by having the angle relations amongst three spots check. The angles given in the table between any two planes are calculated from the relation

$$\cos \alpha = \frac{h_1h_2+k_1k_2+l_1l_2}{\sqrt{(h_1^2+k_1^2+l_1^2)(h_2^2+k_2^2+l_2^2)}}$$

where $\alpha$ is angle between two planes $(h_1k_1l_1)$ and $(h_2k_2l_2)$. Table II gives values of angles between planes of forms $\{h_1k_1l_1\}$ and $\{h_2k_2l_2\}$ for crystals of cubic system. In the case of the diamond cubic structure not all reflections occur. Table III lists the planes which reflect for the diamond cubic structure. This table should be used in conjunction with Table II to make certain the reflection is possible.

The angles between the spots may also be determined by the method of A. B. Greninger[2], in which a chart is used to obtain an estimate of the angles between the spots. Fig. 4 is a Greninger chart for a 3.0 cm distance between specimen and film. To use the chart, a tracing of the film is placed on the chart so that a row of spots, that is, spots of a zone, lay on a hyperbola extending across the chart horizontally. The center of the film trace is placed on the center spot of the chart, turning the tracing till a zone lies on a hyperbola. Then the angles between any spots on such a zone are read directly by determining the intervals on the chart between two spots. Each interval is equivalent to 2 degrees. This chart may be used together with Tables II and III to index a pattern without making a projection. A projection is still necessary, however, if an accurate orientation is required.

Once the projection is indexed, the orientation is effectively determined. For example, the orientation of a major axis, say [100], [110], or [111], is obtained directly. This may be done by finding the (400) Laue spot on the projection; then the line drawn from this spot to the projected sample 30.0 cm away on the perpendicular from the center of the projection is the [100] direction. The (100) plane does not reflect in a diamond cubic lattice; however, the (400) is parallel to (100) and the [100] direction is perpendicular to the (400). For

[2] A. B. Greninger, "A Back-Reflection Laue Method for Determining Crystal Orientation", *Zeitschrift fur Kristallographie*, Vol. 91, pp. 424-432 (1935).

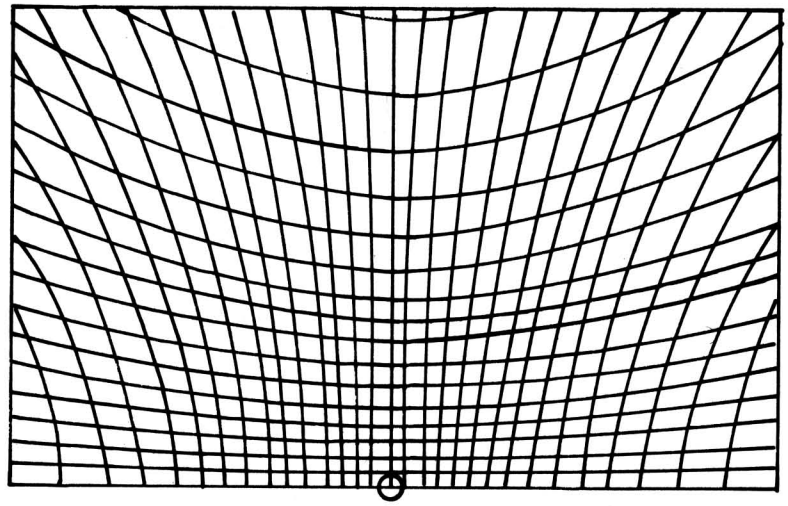

Fig. 4 - Greninger chart for Laue back-reflection pattern--3-cm distance from specimen to film graduated in 2° intervals.

the [110] direction the (220) reflection is found, as the (110) plane does not reflect, and for the [111] direction the (111) plane.

If deformations are present in a single crystal, these deviations from a perfect lattice become evident in the Laue pattern. The most obvious deviation from a single crystal is polycrystalliinity, which is recognizable from the pattern in that the spots seem randomly arranged, and do not lie on hyperbolas. If the crystals are smaller than 50 microns, spots will be located on rings, and, as the crystal size becomes smaller the spots increase in number. In the crystal-size region of about 5 microns (and smaller) the spots are so numerous that they are not discernible, and only continuous rings are visible on the pattern.

If the Laue spots are sharp and circular, the reflections come from a nearly perfect crystal; however, if the spots are elongated, then some deformation must be present. Such elongation of spots is called asterism.

Another form of deformation may occur which is known as twinning. In such cases, portions of the lattice are mirror images of each other. The junction, that is, the plane of symmetry, relating one portion of the lattice to the other, is called the twinning plane. In the case of germanium crystals, the twinning plane appears in most cases as a line on the polished face of an ingot. If back-reflection Laue patterns are taken on each side of the twinning plane with the x-ray beam parallel to it, the two patterns will be exact mirror images.

The occurrence of double spots on a Laue pattern indicates another type of deformation, which in some cases is caused by striations. The difference in orientation of the striations would explain the observed double spots[4].

The procedures described for obtaining orientation and deformation information are the methods used to date. Other procedures may be used to supplement the techniques. These procedures are adequately described in the references (1) and (5).

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[4] E. Teghtsoonian and B. Chalmers, "The Macromosaic Structure of Tin Single Crystals", *Canadian Journal of Physics*, Vol. 29, pp. 370-381 (1951).

[5] Henry, Tipson, and Wooster, THE INTERPRETATION OF X-RAY DIFFRACTION PHOTOGRAPHS, D. Van Nostrand, (1951).

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