High-Precision Orientation of Crystals Using the Laue Method with Characteristic X-rays

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A method previously developed for high-precision orientation of low-index planes of perfect or nearly perfect crystals has been extended to planes with arbitrary indices. The extension has been accomplished by predicting the back-reflexion patterns of Laue spots due to characteristic X-rays and by identifying spots which should have equal intensity when the correct orientation is obtained. It is possible to orient the specimen crystal within 0·01° of the desired orientation in favourable cases. In cases where the equal-intensity criterion cannot be used the accuracy of the orientation using the predicted pattern is at least as good as that using the conventional Laue method. Furthermore there is no need to measure angular distances between the spots or the distance between the crystal and the film.

1. Introduction

The Laue method, as described in any textbook on X-ray diffraction in crystals, uses polychromatic radiation. Some of the diffraction spots in a Laue photograph may, however, be due to characteristic lines present in the X-ray spectrum. These spots, which will be called *characteristic spots* in the following, are generally of much higher intensity than the ordinary Laue spots. Slight changes in the direction of the incident beam have a strong influence on the intensity of these characteristic spots and may even introduce new spots. The ordinary spots, on the other hand, are not greatly affected by such changes. As has been reported by Mathiesen (1968), the characteristic spots can be used experimentally for a high-precision orientation of a crystallographic symmetry axis parallel to the incident beam. In this case the characteristic spots related by symmetry should appear with equal intensity. The method has been used recently for preparing crystal surfaces parallel to a low-index crystallographic plane (Mathiesen, Gerward & Pedersen, 1974).

The importance of extending crystal surface studies to high-index surfaces with ordered arrays of steps has been recognized for some time (Perdereau & Rhead, 1971). The present work is a systematic study of the characteristic spots in a Laue photograph. The results make it possible to predict the characteristic spots theoretically and to use them for preparing crystal surfaces parallel to a high degree of accuracy to a crystallographic plane with arbitrary indices. The method can be applied to perfect or nearly perfect single crystals.

2. Theoretical basis of the method

The mean wavelength, λ, and the wavelength range, $\Delta\lambda$, of the incident X-rays contributing to a particular reflexion in the Laue pattern are given by the Bragg equation and its differential:

$$\lambda = 2d_{hkl} \sin \theta, \tag{1a}$$

$$\Delta\lambda = \lambda \cot \theta \Delta\theta, \tag{1b}$$

where d_{hkl} is the interplanar spacing, θ is the grazing angle of incidence, given by the orientation of the lattice planes relative to the incident beam, and $\Delta\theta$ is the effective angular divergence (to be discussed in § 5). The *hkl* reflexion gives rise to a characteristic spot if the following condition is fulfilled:

$$|\lambda_c - \lambda| < \Delta\lambda, \tag{2}$$

where λ_c is a characteristic X-ray wavelength. The condition (2) can be visualized geometrically in reciprocal space: the reciprocal-lattice point *hkl* should be very close to the Ewald sphere with radius $1/\lambda_c$.

The characteristic spots are incidental phenomena and a computer program has been prepared in order to predict whether they occur in a given experimental situation. The program, which in the present form works for cubic crystals, runs through all reflexions in order of increasing value of $h^2 + k^2 + l^2$, *i.e.* decreasing value of the interplanar spacing. Those reflexions are selected which have a non-zero structure factor, and which are consistent with equation (2) for a given orientation of the crystal specimen and given values of λ_c and $\Delta\theta$. The program calculates Bragg angles, structure factors and related diffraction data.

The number of possible X-ray reflexions is restricted by the choice of the characteristic wavelength. This can be seen by inserting the wavelength λ_c in equation (1a) and noting that the Bragg angles are confined to the range $\pi/4$ to $\pi/2$ in the case of back-reflexion. Thus it is sufficient to consider interplanar spacings within the limits:

$$\lambda_c/2 < d_{hkl} < \lambda_c/\sqrt{2}. \tag{3}$$

The corresponding reciprocal-lattice points are situated within two spheres with radius $\sqrt{2}/\lambda_c$ and $2/\lambda_c$ respectively from the centre of reciprocal space.

Krahl-Urban, Butz & Preuss (1973) have shown that crystal orientation can be simplified considerably by using computed and plotted Laue patterns. In the present work the plot program produced by Preuss (1972) has been extended to take into account the intensity of the incident X-rays as a function of wavelength, including the characteristic lines of the anode.

3. Experimental and theoretical Laue patterns

In this section some Laue photographs of principal interest are shown together with the corresponding plotted spot patterns (Figs. 1, 2 and 3). The specimens are single crystals of silicon and germanium. The experimental patterns have been recorded in a back-reflexion Laue camera using Polaroid film and with the specimen mounted on a three-circle goniometer. The collimator of the incident beam has rotational symmetry. The angular divergence, $\Delta\theta_{\text{coll}}$, is $1{\cdot}2 \times 10^{-2}$ rad $= 40'$ (full width).

The experimental procedure begins with a *preliminary orientation* using the ordinary Laue spots produced by the continuous X-ray spectrum. A *more precise orientation* is then obtained by adjusting the specimen until the predicted pattern of characteristic spots appears. In many cases a *high-precision orientation* can be obtained by adjusting the specimen until some selected characteristic spots in the pattern have equal intensity. The appropriate spots are found either by symmetry considerations or by theoretical calculations.

4. Characteristic spots having equal intensity

The method of high-precision orientation discussed in this work requires that the Laue pattern contain a number of characteristic spots, which should have equal intensity when the exact orientation has been achieved. In order to orient the crystal completely around the three axes of the goniometer, one has to consider the intensities of at least three spots. The spots can be related by symmetry but this is not a necessary condition. The symmetric and the asymmetric cases are discussed separately in the following sections.

4·1 *Characteristic spots related by symmetry*

If the Laue projection symmetry of the desired orientation contains a three, four, or sixfold axis or two mirror lines, then it is sufficient for a high-precision orientation to consider the intensities of one set of symmetrically equivalent characteristic spots. If there is a twofold axis only, one has to consider at least four characteristic spots (related in pairs) not on the same line through the centre of the pattern.

Inspection of the possible Laue projection symmetries (*International Tables for X-ray Crystallography*, 1969) shows that for any Laue class there are at most three planes having the high projection symmetry described above. The Laue class *m3m*, to which silicon

and germanium belong, can be used as an example. The three planes (100), (111) and (110) have the Laue projection symmetries 4*mm*, 3*m* and 2*mm* respectively. They can be oriented perpendicular to the incident beam with a high precision provided that at least one set of characteristic spots is produced by the X-rays. The (*hk*0) and (*hhl*) planes have the lower projection symmetry of *m* only and all other planes have no projection symmetry. General symmetry considerations cannot give a sufficient number of characteristic spots for a high-precision orientation of these planes. However, groups of characteristic spots, which are singled out from a large number of spots in the Laue pattern, might exhibit internal symmetries higher than the pattern as a whole. Incidental symmetries of this kind can transform a low-symmetry case into a high-symmetry one with a sufficient number of symmetrically equivalent characteristic spots. An example is shown in Fig. 2 and is discussed in § 5.

4·2. *Characteristic spots without any symmetry relation*

The calculations in this work have shown that it is often possible to find groups of characteristic spots, which should have equal intensity, even if they do not form symmetric patterns. In this case it is necessary to be able to predict the pattern of the characteristic spots theoretically.

The spots belonging to a particular group are produced by reflexions of the same characteristic wavelength in lattice planes with the same spacing and the same structure factor. The Bragg condition (1) implies that the spots define a circle around the centre of the Laue pattern. The radius of the circle is the same as that of the corresponding Debye–Scherrer ring in a powder pattern. A complete high-precision orientation requires that the Laue pattern contain at least three characteristic spots on the same circle or four spots on two different circles. In the latter case the chords that can be drawn between the spots on each circle must not be parallel.

5. Discussion

The method for crystal orientation discussed in this work is based on the theoretical prediction of the characteristic spot pattern. There is no need to measure angular distances between diffraction spots in the Laue photograph or the distance between the crystal and the film. Thus a crucial source of error in the conventional Laue technique is eliminated.

In principle, the method is similar to the one used when a crystallographic plane is oriented in a diffractometer (Cullity, 1956) but the present method has some obvious advantages: the experimental arrangement is more simple; reflexions of characteristic X-rays in several planes are observed simultaneously, so that a complete orientation is possible in one experiment; a preliminary orientation can be obtained with the ordinary Laue spots produced by the continuous X-ray spectrum.

The effective angular divergence which appears in equation (1b) is given by

$$\Delta\theta = \Delta\theta_{coll} + \Delta\theta_{line} + \Delta\theta_{cryst} \qquad (4)$$

where the contributions on the right side are due to the collimator, the X-ray line width and the diffracting crystal respectively. The experimental value of $\Delta\theta_{coll}$ was given in § 3.

The spectral line width of molybdenum $K\alpha_1$ radiation is $\Delta\lambda(K\alpha_1) = 0\cdot00029$ Å, (Compton & Allison, 1934). Inserting this value in equation (1b) shows that $\Delta\theta_{line}$ is much less than $\Delta\theta_{coll}$ provided the back-scattered beam does not lie within about 4° of the incident beam.

The rotational symmetry of the incident beam requires $\Delta\theta_{cryst}$ to be small compared with $\Delta\theta_{coll}$. Thus the method is restricted to perfect or nearly perfect crystals. There are, however, an increasing number of materials which can be considered. Several semiconductors, like silicon, germanium and gallium arsenide, can be grown in the form of single crystals of high perfection. The reflexion range of such crystals is a few seconds of arc. Single crystals of metals, like aluminum, copper and lead, can be obtained with a mosaic spread less than 10'.

The energy current of a characteristic spot is schematically shown in Fig. 4 as a function of a misfit angle φ defined by

$$\varphi = \theta - \theta_c , \qquad (5)$$

where θ is determined by the orientation of the lattice planes relative to the incident beam [equation (1)] and θ_c is determined by the characteristic wavelength λ_c:

$$\theta_c = \text{arc sin } [\lambda_c/2d_{hkl}] . \qquad (6)$$

Characteristic spots related by symmetry in the Laue pattern will automatically have the same misfit angle when the crystal is oriented properly. Special care has to be taken when the characteristic spots do not form symmetric patterns. It has to be verified that the spots belonging to the same group, as defined in § 4·2, also have the same misfit angle. Considering the Ewald construction in the reciprocal lattice, it is obvious that this is generally the case, provided the incident-beam direction is parallel to a reciprocal-lattice vector.

In the present work the value of the misfit angle has been calculated for each characteristic reflexion. The reflexions belonging to a particular group are easily identified, because the results of the calculations are presented in order of decreasing interplanar spacing, as mentioned in § 2.

A slight change in the direction of the incident beam changes the misfit angles in different directions so that the reflexions belonging to a particular group no longer have equal intensities. Consider, for instance two spots related by mirror symmetry. Their intensities change in opposite directions as indicated in Fig. 4. Using the angular divergence given in § 3 and assuming that the smallest relative intensity change that can be

detected is 2 to 5% one should be able to orient the specimen within $0\cdot8 \times 10^{-2}$ deg to 2×10^{-2} deg. This range agrees well with the experimental value 1×10^{-2} deg reported by Mathiesen (1968).

Although the condition (2) for characteristic spots to occur is rather restrictive, there is a certain degree of freedom in choosing the characteristic X-ray wavelength. The K lines are most useful because they have high intensities compared with the continuous spectrum. The chance of getting characteristic reflexions increases with decreasing value of the characteristic wavelength. This follows from equation (3): the shorter the wavelength, the larger is the volume of reciprocal space containing possible reciprocal-lattice points for back-reflexion. The calculations have shown that the molybdenum K lines give a suitable number of reflexions in most cases.
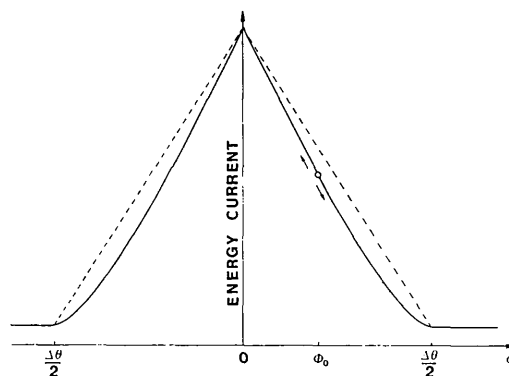

Fig. 4. Theoretical diffracted energy current *versus* misfit angle, φ, for a characteristic X-ray reflexion. $\Delta\theta$ = angular divergence of the collimator. Broken line is valid for a slit collimator, full line for a cylindrical collimator. φ_0 indicates the common misfit angle for a set of equivalent reflexions.
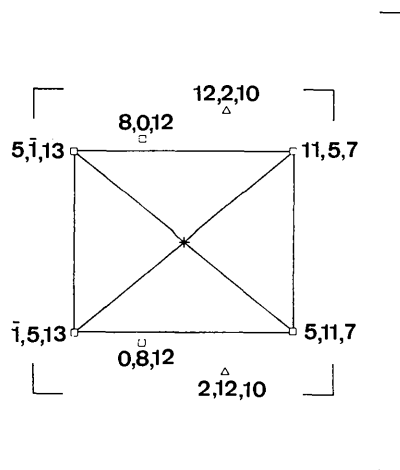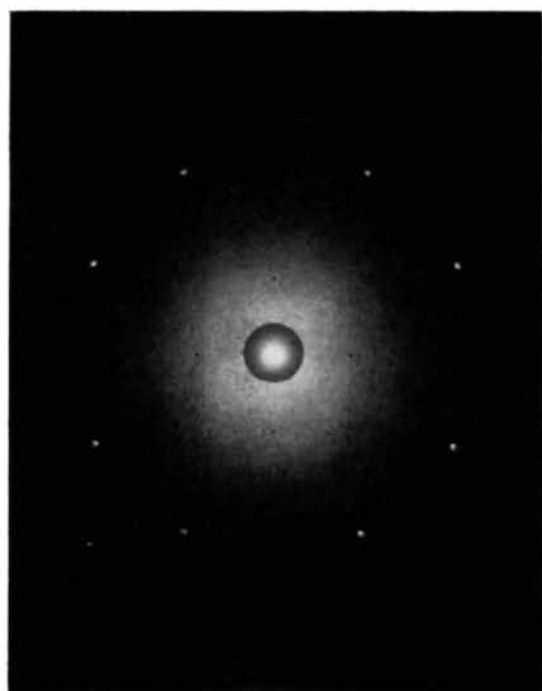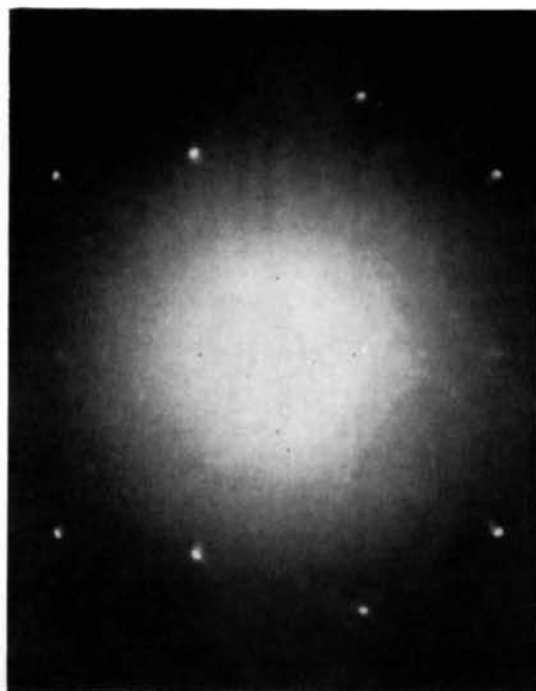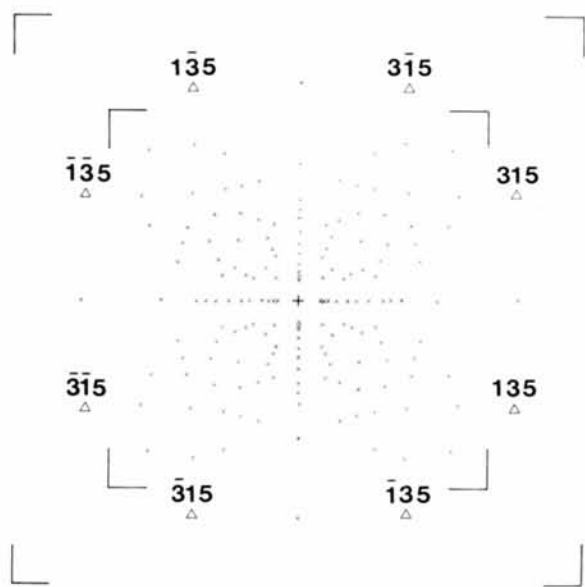

Fig. 5. Separate plot of the characteristic spots in Fig. 2. The group of four spots at the corner of a rectangle has the incidental symmetry 2mm.
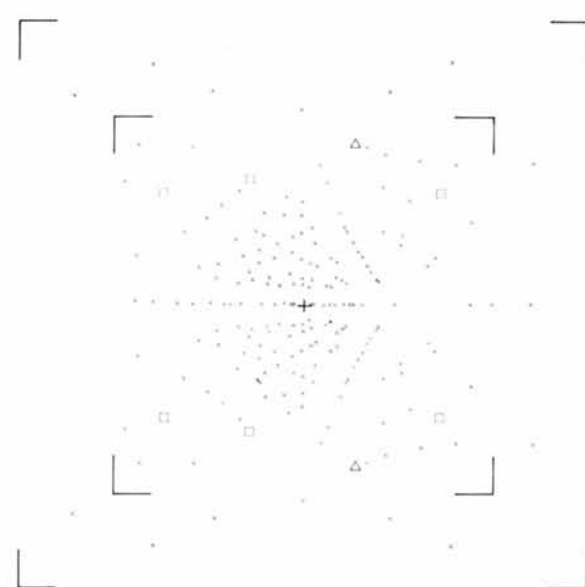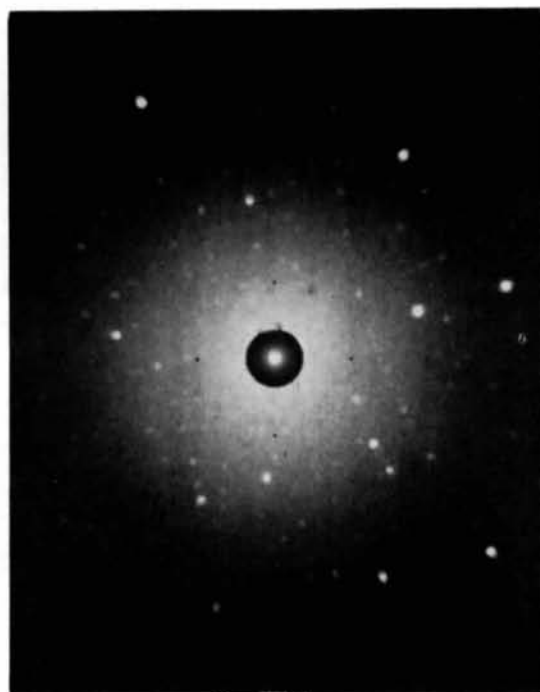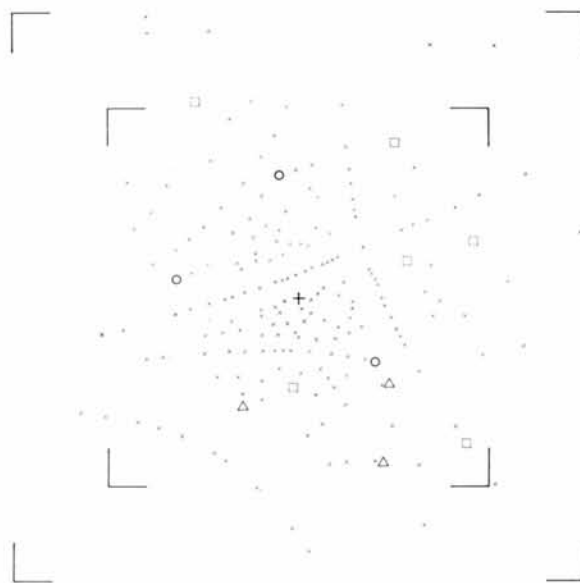
(a)

(a)

(b)

(b)

Fig. 1. Laue projection on the (001) plane of germanium. (a) Experimental photograph. Cobalt radiation. (b) Plotted figure. × = ordinary diffraction spots produced by the continuous spectrum. △ = diffraction spots produced by Co $K\beta_1$ radiation. An area of 10×10 cm is indicated for the distance between film and crystal equal to 3 and 2 cm respectively.

Fig. 2. Laue projection on the (112) plane of germanium. (a) Experimental photograph. Molybdenum radiation. The fluorescence radiation from the germanium sample has been reduced by an aluminum sheet (thickness 0·5 mm) in front of the photographic film. (b) Plotted figure. × = ordinary spots. □ = Mo $K\alpha_1$ spots. △ = Mo $K\beta_1$ spots. The indices of the spots are shown in Fig. 5.

(a)

(b)

Fig. 3. Laue projection on the (721) plane of silicon. (a) Experimental photograph. Molybdenum radiation. (b) Plotted figure. × = ordinary spots. □ = Mo $K\alpha_1$ spots. ○ = Mo $K\alpha_2$ spots. △ = Mo $K\beta_1$ spots. The indices of the spots are shown in Fig. 6.
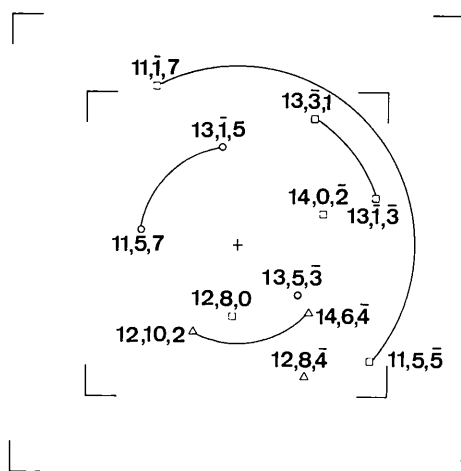
Fig. 6. Separate plot of the characteristic spots in Fig. 3. Spots joined by an arc of a circle should have equal intensity according to the calculations.

Fig. 1 shows the Laue projection on the (001) plane of germanium. It contains a fourfold axis so that a high-precision orientation can be performed with one set of symmetrically equivalent characteristic spots. In this particular case it was found that the molybdenum K lines failed to give any reflexions. The characteristic spots in Fig. 1 have been produced by cobalt $K\beta_1$ radiation.

Fig. 2 is an example of a low-symmetry pattern. It is a Laue projection on the (112) plane of germanium. The Laue pattern as a whole contains a mirror line. However, a sufficient number of symmetrically equivalent characteristic spots can be found because of incidental symmetries like the one shown in Fig. 5. Consider the group of four characteristic spots formed by reflexions from the $(5,\bar{1},13)$, $(11,5,7)$, $(5,11,7)$ and $(\bar{1},5,13)$ planes. They have an internal symmetry characterized by $2mm$. Thus a complete high-precision orientation is possible by adjusting the crystal until these four spots have equal intensity. Another group

of four spots, formed by reflexions from the $(\bar{3},5,11)$, $(5,\bar{3},11)$, $(3,11,5)$ and $(11,3,5)$ planes, appears outside the field of view of Fig. 5.

Fig. 3 shows the Laue projection on the (721) plane of silicon. A plane of this kind would be very difficult to orient within $\frac{1}{2}°$ or better by the conventional Laue technique. The Laue pattern contains no symmetry and none of the most close-packed zones formed by the ordinary Laue spots goes through the centre of the pattern. However, a complete high-precision orientation has been performed in this work by the method described in § 4·2. The pattern of characteristic spots in Fig. 3(b) is shown separately in Fig. 6. Spots joined by an arc of a circle should have equal intensity according to the calculations. Inspection of Fig. 3(a) shows that it has been possible to fulfil the intensity condition experimentally for all the spots indicated in Fig. 6. Thus a very accurate check on the crystal orientation has been achieved.

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