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A Method of Obtaining an Extremely Parallel X-ray Beam by Successive Asymmetric Diffractions and Its Applications

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For obtaining X-ray beams of extremely narrow angular spread, a monochromator system consisting of three crystals, prepared from a block of silicon single crystal, in which asymmetric 422 diffractions of Cu $K\alpha$ are repeated, is constructed. The narrowest angular width of the beam thus obtained is estimated to be 0·01″. By utilizing this monochromator system in place of the monochromator crystal in a double-crystal spectrometer of parallel setting, rocking curves are measured for both the Bragg- and Laue-case diffractions. No appreciable broadening of the half-value width is observed for the Bragg-case diffraction, compared with that of the intrinsic curve. The narrowest half-value width observed is 0·16″. Several subsidiary maxima are observed in the rocking curve of Laue-case diffraction from a thin crystal.

1. Introduction

Observation of an intensity curve of diffraction as close as possible to the so-called intrinsic curve, which is expected to be obtained with an ideally monochromatic and parallel incident beam, has been one of the most important subjects in experimental studies of X-ray diffraction by single crystals relating to the dynamical theory of X-ray diffraction. For this purpose, the exploring beam, that is to say, the beam incident on a crystal to be examined, should be sufficiently narrow in angle compared with the angular range of selective reflection for the crystal examined, which is usually as small as a few seconds.

In double-crystal spectrometers of parallel setting, which have been used since the 1920's, the influence of the spectral spread of primary X-rays is practically eliminated because the angular dispersion corresponding to the spectral width is cancelled out by successive diffractions from two crystals. However, in most of experiments using double-crystal spectrometers till the middle of the 1950's, the exploring beam obtained by a monochromator crystal had the characteristic due to the same intrinsic curve as that of the specimen crystal, because in them the same diffraction condition, say, that of the symmetric Bragg-case on a diffracting plane of the same spacing, was employed for both crystals. Under such a condition the observed rocking curve does not represent the intrinsic curve of the specimen crystal, but the self-convolution of it.

Brogren & Adell (1954) used different diffraction conditions, namely, the Bragg- and Laue-case diffractions, for the respective crystals in a double-crystal spectrometer, and observed for the first time the tendency to asymmetry of rocking curves.

Since about ten years ago, several methods for obtaining the exploring beam of the angular spread narrower than the angular range of selective reflexion have been devised, and by the use of double-crystal spectrometers applying these methods it becomes possible to measure the rocking curves which reproduce fairly closely the characteristics of the intrinsic ones. Broadly these methods are classified as follows:

(i) Limitation of a diffracted beam of the Laue case, which is spread in a crystal like a fan, by a slit placed behind the monochromator crystal (Borrmann & Kohra, 1959; Authier, 1960; Nakano, Kikuta & Kohra, 1964; Nakano, 1965).

(ii) Use of a diffracted beam of the Laue case, which is transmitted through a sufficiently thick crystal by the effect of anomalous transmission (Borrmann & Kohra, 1959).

(iii) A combination of (i) and (ii), in which a slit broader than that in (i) is used because, as pointed out by Nakano (1965), the diffraction effect due to the slit restricts the angular spread of the beam obtained in (i).

(iv) Use of a diffracted beam of the asymmetric Bragg-case (Renninger, 1961; Kohra, 1962).

Here it is worth while to mention the work of DuMond and his coworkers (DuMond, 1937; Bollman, Bailey & DuMond, 1938). He proposed the use of the beam diffracted successively by two crystals, the diffracting planes of which had the same spacing and were so slightly inclined that the reflective regions overlap only partly with each other. The angular width of the beam thus obtained is narrower than that of the intrinsic curve of the crystals concerned. Using this beam as the exploring beam in the arrangement corresponding to the double-crystal spectrometer of parallel setting*, they made some measurements of the rocking curves, but were able to observe neither the asymmetric profile nor the nearly flat region. This was, as they themselves presumed, probably because the tail of the

* Strictly speaking, the arrangement used by them is a triple-crystal spectrometer of the $(n, -n, n)$ setting.

exploring beam was so much extended that the integrated area of the part of its profile below the half-value of the peak was even larger than that above it. He considered also the case in which the both diffracting planes are exactly parallel and multiple successive reflexions take place between them, and pointed out that the half-value width of the exploring beam does not become narrower, while the tail part diminishes.

On the other hand, Renninger (1955) made use of a triple-crystal spectrometer instead of a double-crystal spectrometer and gave conclusive evidence for the asymmetry of the rocking curve. Later, detailed studies using triple-crystal spectrometers were made by Bubáková, Drahokoupil & Fingerland (1961 *a*,*b*, 1962*a*,*b*), Bubáková (1962) and Fingerland (1962). More recently Kohra, Kikuta, Annaka & Nakano (1966) applied a triple-crystal spectrometer using asymmetric reflexions for the two monochromator crystals, for studying the temperature effect on the profile of the rocking curve of the Bragg- or Laue-case.

As a technical development of a different aspect, Okkerse (1963, 1965*a*,*b*) made a series of precise measurements with the arrangement corresponding to the double-crystal spectrometer using successive diffractions by two parts of one block of single crystal. Bonse & Hart (1965, 1966*a*) obtained an exploring beam of tailless intensity profile, by utilizing a monochromator system prepared from one block of single crystal, in which multiple diffractions of the symmetric Bragg-case take place, and successfully applied it to the measurement of small-angle scattering. This may be considered as a realization of the method suggested previously by DuMond (1937).

In the present study we have constructed a monochromator system composed of three crystals prepared from one block of silicon single crystal, in which two or three successive diffractions of the asymmetric Bragg-case take place. This system has been able to give an exploring beam of extremely narrow angular spread, and has made possible the measurements of rocking curves which are satisfactorily close to intrinsic ones, for both the Bragg and Laue cases.

2. Theory

In the present study the exploring beam was obtained from a monochromator system as schematically shown in Fig. 1. It consists of three crystals, the diffracting planes of which are parallel and with the same reflexion indices corresponding to the condition of parallel setting. The surfaces of the crystals are inclined to the diffracting plane so that the asymmetric reflexion takes place successively.

According to the dynamical theory (Laue, 1960; Kohra, 1962), the angular widths of the incident and diffracted beams, ω_0 and ω_h, for selective reflexion in the asymmetric Bragg-case are given as

$$\omega_0=\sqrt{\frac{\sin\theta_h}{\sin\theta_0}}\,\omega_s \quad \text{and} \quad \omega_h=\sqrt{\frac{\sin\theta_0}{\sin\theta_h}}\,\omega_s \qquad (1)$$

respectively, where ω_s is the width for the symmetric case, θ_0 (or θ_h) the angle between incident (or reflecting) direction and the surface. θ_0 and θ_h are given as $\theta_0=\theta-\alpha$ and $\theta_h=\theta+\alpha$, respectively, where θ is the Bragg angle and α the angle between the diffracting plane and the surface. From (1) we have

$$\omega_h=b\omega_0\,, \qquad (2)$$

where $b=\sin(\theta-\alpha)/\sin(\theta+\alpha)$. The parameter b will hereafter be called the asymmetry factor. It is to be noted that relation (2) is valid not only for the selective region but also for the range near it.

Now we consider successive reflexions with the same asymmetry factor, b. Angular ranges of the beams incident on and diffracted from each crystal are given as follows,

$$\left.\begin{aligned}&\omega_{h1}=b^{1/2}\omega_s=\omega_{02}\\&\omega_{h2}=b\omega_{02}=\omega_{03}\\&\cdots\cdots\cdots\cdots\\&\omega_{hn}=b\omega_{0n}=\omega_{0n+1}\end{aligned}\right\} \qquad (3)$$

where subscripts $1,2,3,\ldots n$ denote the sequence of diffraction. From (3) we have

$$\left.\begin{aligned}&\omega_{h2}=b^{3/2}\omega_s=b^2\omega_{01}\\&\cdots\cdots\cdots\cdots\cdots\\&\omega_{hn}=b^{n+1/2}\omega_s=b^n\omega_{01}\end{aligned}\right\}. \qquad (4)$$

As seen from (4), the angular divergence decreases with the increase of the repeated number of diffractions so long as $b<1$. In the present study the diffracted beam corresponding to $n=2$ or 3 was employed as the exploring beam.

On the other hand, the horizontal cross section of the reflected beam becomes broader with the repeated number of diffractions. The ratio of the horizontal width of the cross section of the nth reflected beam to that of the incident beam is b^{-n}.

By the refraction effect, the directions of the incident and diffracted beams corresponding to the centre of selective reflexion do shift from the directions given by the kinematical theory, towards the outward normal of the crystal surface respectively by the magnitudes

$$\frac{\varphi_o}{2\sin 2\theta}(b+1) \quad \text{and} \quad \frac{\varphi_o'}{2\sin 2\theta}\left(\frac{1}{b}+1\right), \qquad (5)$$

where φ_o is the oth Fourier component of the polarizability of the crystal. Now let us consider the case that the beam diffracted from the nth crystal is incident on the $(n+1)$th crystal. When the asymmetry factor b is nearly unity, the angular range of the diffracted beam from the nth crystal appreciably overlaps the range of selective reflexion for the $(n+1)$th crystal, so that the intensity of the diffracted beam is expected to be fairly strong, but the sharpening of the beam cannot be satisfactory. On the other hand, if the value of b is small, the diffracted beam from the $(n+1)$th crystal may deviate from the selective region for the $(n+1)$th crystal. In this case, a considerable decrease in reflected intensity from the $(n+1)$th crystal is unavoidable, but a diffracted beam with a sufficiently

narrow angular spread may be obtained. In the present experiment such a condition is applied.

3. Experimental

The monochromator system, which consisted of three parts I, II and III, as shown in Fig. 1, was prepared from one block of silicon single crystal of high purity and free from dislocation. The surface of each part was cut so as to make the angle of 38° (=α) to the plane ($2\bar{1}\bar{1}$). The 422 asymmetric reflexion of Cu $K\alpha$ radiation was used, the Bragg angle, θ, being 44°1′. Thus the incident and reflexion angles are 6° and 82°, respectively, and $b=0{\cdot}106$.

The angular width of the beam diffracted from the second part is calculated to be 0·10″ from (4), and that from the third part 0·01″. The beam thus obtained is almost polarized because the scattering angle, 2θ, is near 90°. The horizontal magnification of the cross section of the diffracted beam relative to that of the incident beam is about 89 and 843 with respect to the second and third parts, respectively.

The beam diffracted from the second or third part of the monochromator system was used as the exploring beam, and the specimen crystal was set in the position corresponding to the parallel setting. The apparatus for rotating the specimen was the same as the one used in a previous investigation (Kohra, Kikuta, Annaka & Nakano, 1966). The rotation was made by a lever mechanism with a precision micrometer. The angular reading for the specimen crystal was calibrated by a Hilger autocollimator of reading 0·1″.

The intensity of X-rays was measured by a scintillation counter with a pulse height analyser. The spectrometer was set in a box, and the temperature in it was kept constant within ±0·1 °C by circulating water. All measurements were made by remote control.

4. Results

By using the exploring beam obtained in the way described in § 3, we made some measurements of diffraction curves from silicon single crystals for both the Bragg- and Laue-cases.

(i) *Measurements of the Bragg-case diffraction curves*

The half-value width of the intrinsic curve of 422 diffraction in the symmetric case is calculated to be 2·93″. This width is very large compared with the angular spread of the exploring beam of the order of 0·01″ or 0·10″, and, therefore, is too large to detect the broadening of a rocking curve due to the angular spread of the exploring beam. In order to distinguish this effect sensitively, we measured rocking curves of asymmetric reflexion because for them the half-value width of the intrinsic curve is considerably small.

Three specimens were prepared from silicon crystals free from dislocation and of high purity. The values of α, θ_h and b for each specimen are given in Table 1. Examples of observed rocking curves are given in Fig. 2 with schematic diagrams showing experimental arrangements. The observed half-value widths of the rocking curve and the calculated ones of the intrinsic curve are also given in Table 1.

As seen in Table 1, the agreement in half-value width between experiment and theory is satisfactory for each case except specimen 3 with beam II. It can be concluded that the broadening effect is negligibly small for not only beam III but also II when the half-value width of the intrinsic curve is larger than ~0·5″. As to the case of specimen 3 with beam II, it would not be very meaningful to argue seriously the discrepancy between theory and experiment, because the error for this case given in the Table, which is a formal standard deviation determined from five observed values, seems to be underestimated, considering the following facts: the half-value width varies very sensitively with the value of b, and further the accuracy corresponding to the magnitude of the calculated error almost exceeds that of the present measurement.

On the other hand, it is to be noted that the observed values of the reflexion per cent, that is the peak intensity of the rocking curve expressed as a percentage of the incident intensity, are noticeably low compared with those calculated. This phenomenon is probably related to the unneveness of the crystal surface.* This effect is expected to become appreciable when the reflecting angle is small.

It is also to be noted that there appears an extra small peak in the low-angle side of the main peak in each of the observed rocking curves. The peak is the most conspicuous for specimen 3. The appearance of such a peak seems to be also due to the unevenness of the surface, because the refraction effect is expected to become small at some irregular parts of the surface.

(ii) *Measurements of the Laue-case diffraction curves from a thin crystal*

Finally, the Laue-case diffraction from a thin crystal was studied. The specimen was wedge-shaped with the edge parallel to [111] and both surfaces are nearly parallel to ($1\bar{1}0$). A diffraction topograph of this spec-

* The unevenness of the surface was experimentally confirmed by X-ray diffraction topographs taken on some specimens by the Berg–Barrett technique. Many spots were in fact seen to exist in these topographs.

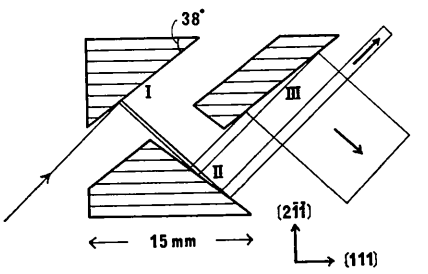

Fig. 1. Schematic diagram of the monochromator system.

imen taken by the Lang technique with the 422 reflexion of Cu $K\alpha$ showed Pendellösung interference fringes. The thicker part of the specimen was mounted on the holder to avoid the strain, and the thinner part was subject to X-ray diffraction. The 422 reflexion of Cu $K\alpha$ was used as in (i). A beam, which was diffracted from the second crystal of the monochromator system and limited by a slit 0·1 mm wide and 1·5 mm high, was incident on the thinner part of the specimen. Rocking curves were measured at various positions of the crystal. The profile and intensity of the curves varied sensitively with position.

A typical example of the rocking curves is shown in Fig. 3, in which one can see that the main peak is accompanied by several subsidiary peaks on both sides.

This curve is presumed to correspond to the intrinsic curve of Laue-case diffraction, because the exploring beam is sufficiently narrow in angular divergence and broad in cross section compared with the thickness of the crystal.

According to the dynamical theory, the intrinsic curve of the Laue-case diffraction for the symmetric case is given by

$$\frac{P_h}{P_0}=\frac{1}{4}\frac{1}{1+W^2}\left[\exp\left\{-\frac{\mu H}{\cos\theta}\left(1-\frac{\kappa_0}{\sqrt{1+W^2}}\right)\right\}+\exp\left\{-\frac{\mu H}{\cos\theta}\left(1+\frac{\kappa_0}{\sqrt{1+W^2}}\right)\right\}-2\exp\left(-\frac{\mu H}{\cos\theta}\right)\cos\frac{2\pi H}{d}\sqrt{1+W^2}\right], \quad (6)$$

where W is the parameter representing the deviation from the diffraction condition, $-1\leq W\leq 1$ corresponds to the region of selective reflexion, μ is the linear absorption coefficient, H is the thickness of the crystal, and d is the extinction distance and

$$\kappa_0=\varphi_{hi}|\varphi_{hr}|/\varphi_{0i}\varphi_{hr}\,.$$

The third term of (6) gives the oscillation term of intensity, with the angular period depending on H and W. The values of W corresponding to the maxima are given by

$$\frac{2H}{d}\sqrt{1+W^2}=2n+1 \quad (n=\text{integer})\,. \quad (7)$$

It is also seen that the intensity at the centre of the curve ($W=0$) is maximum when

$$H=\frac{(2n+1)d}{2} \quad (8)$$

and minimum when

$$H=nd\,. \quad (8')$$

The thickness H is estimated to be 44 μ from the angular interval of the subsidiary maxima by using equation (7). Since d is calculated to be 16·5 μ, the present case corresponds approximately to condition (8) with $n=2$.

From the features above mentioned, it is concluded that the observed rocking curve shown in Fig. 3 is no doubt the one which is close to the intrinsic curve of Laue-case diffraction, although a detailed analysis of the intensity profile has to be made further.

5. Discussion and conclusions

From the measurements of the rocking curve for various cases as reported in the preceding section it may be concluded that the angular spread of the exploring beam used is sufficiently narrow. Although there is no conventional method at present for determining absolutely such a narrow angular spread, the observed half-value width 0·16″, which was obtained for specimen 3, confirms well the high resolution attained in the present experiment.

As mentioned already, the small extra peak observed in the rocking curve of the Bragg-case diffraction as shown in Fig. 2(*a*), (*b*), and (*c*) is probably due to the unevenness of the surface. It is conceivable that the extra peak may be eliminated or diminished if a more flat surface is obtained with an improved surface treatment. In any case, however, it is certain that the observation of this kind of peak has become possible

Table 1. *Half-value widths of observed rocking curves and calculated intrinsic curves*

Specimen	$-\alpha$	θ_h $(\theta+\alpha)$	b	Half-value width: Observed	Calculated: Intrinsic*	Calculated: Broadening†	Exploring‡ beam
1	40° 5′	3° 56′	14·5	0·76±0·02″	0·78″	≪0·01″	III
				0·65±0·03	0·67	≪0·01	III
2	41 7	2 54	19·7	0·64±0·02	0·67	<0·01	II
3	43 51	10	344	0·16±0·01	0·17	0·03	II

* The calculated values were determined from the intrinsic intensity curve on the basis of a table given by Nakano (1965), assuming the Debye temperature $\Theta_D=543°$, $\varphi_{hr}=-6{\cdot}73\times10^{-6}$ (*International Tables for X-ray Crystallography*, 1962), $\varphi_{hi}=-3{\cdot}01\times10^{-7}$ (Bubáková *et al.*, 1962*b*) and $\varphi_{0i}=-3{\cdot}48\times10^{-7}$ (Bonse & Hart, 1966*b*), where φ_{hr} and φ_{hi} are the real and imaginary parts of the Fourier components of the polarizability of the crystal, respectively.

† Assuming both the intensity profiles to be Gaussian, the values of the broadening are calculated by $\Delta\omega=\sqrt{\omega_1^2+\omega_2^2}-\omega_2$, where ω_1 and ω_2 are the angular widths of the exploring beam and intrinsic curve, respectively.

‡ The exploring beams II and III are the beams diffracted by the second and third parts of the monochromator system, respectively.

owing to the high resolution of the exploring beam.

Several subsidiary peaks in the rocking curves of the Laue-case diffraction shown in Fig. 3 have been observed probably for the first time by the present experiment. It is to be noted that the peaks observed here are due to the interference of two wave fields excited by a plane wave, being in contrast to the interference fringes usually observed in the section or traverse topographs from a wedge-shaped crystal (Kato, 1961*a*,*b*), which are due the to interference of two wave fields excited by a spherical wave. The interference fringes due to the wave fields excited by a plane wave were recently observed topographically by Malgrange & Authier (1965) by applying the slit method to the double-crystal spectrometer. On the other hand, a group in Poland (Godwood, Lefeld-Sosnowska & Zielińska-Rohozińska, 1964; Lefeld-Sosnowska, 1964; Zielińska-Rohozińska, 1965) made measurements of diffracted intensity from a wedge-shaped crystal as a function of thickness to determine the coefficients of normal and abnormal absorptions, using a triple-crystal spectrometer. The intensity oscillations observed in their experiments, however, are also due to the interference of two wave fields excited by a spherical wave.

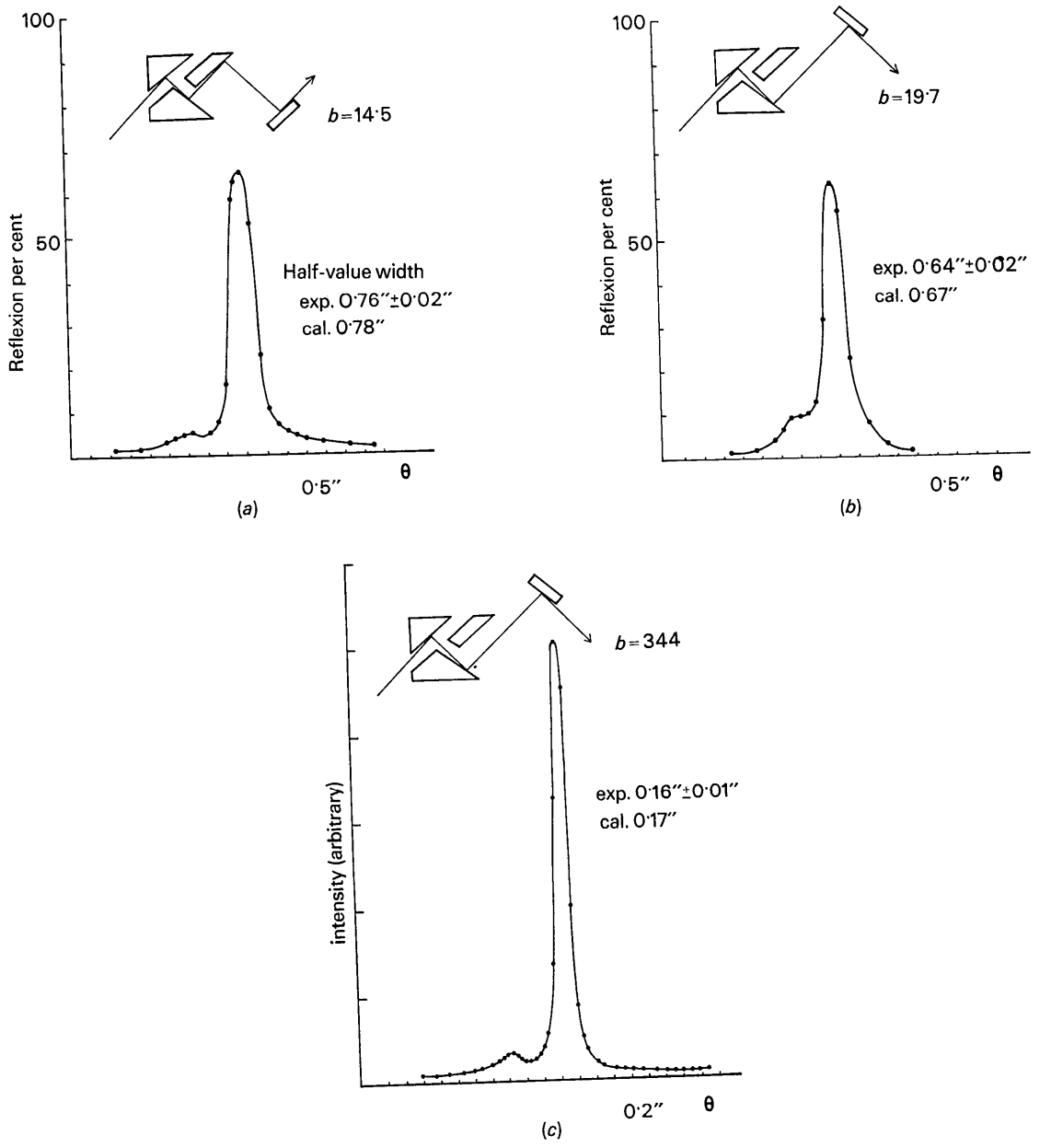

Fig. 2. Observed rocking curves from silicon single crystals of 422 Bragg-case diffraction, with Cu $K\alpha$ radiation. (*a*) Specimen 1 ($b=14{\cdot}5$), exploring beam III. (*b*) Specimen 2 ($b=19{\cdot}7$), exploring beam II. (*c*) Specimen 3 ($b=344$), exploring beam II.

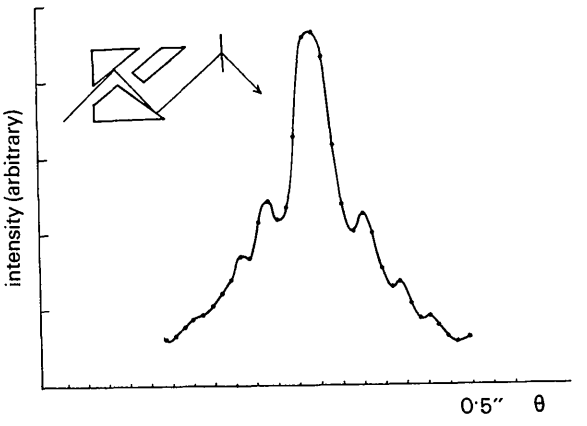

Fig. 3. Observed rocking curve from a thin silicon crystal of 422 Laue-case diffraction of Cu $K\alpha$ radiation. Exploring beam II.

The monochromator system used in the present study may be utilized in place of one or two crystals in a triple-crystal spectrometer. In spectral analysis using such an arrangement for example, the attainable resolution is expected to be sufficiently high for determining the energy loss or gain of X-ray photons due to the interaction with phonons, the order of which is a few hundredths of 1 eV. It is also possible to determine accurately the structure factors on single crystals. For such purposes, however, the technical problem to have a high power X-ray source must be solved.

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