A Method of Obtaining a Highly Parallel and Monochromatic X-ray Beam by Successive Diffraction

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A beam of Cu Kα 1 with an angular spread $1 \times 10^{-5}$ radian and a wavelength spread $(\delta \lambda/\lambda) = 0.8 \times 10^{-5}$ was obtained by a silicon monochromator–collimator system. It consisted of two crystal elements in which the 620 and 333 reflexions occurred successively. The two elements, originally monolithic, were cut after they had been glued to two metal blocks connected by a leaf spring. In order to satisfy the diffraction conditions in the two elements, one element was rotated about 10° with respect to the other by bending the leaf spring. Using this beam, rocking curves of germanium single crystals were measured for the 220, 440 and 222 reflections. The half-value widths of the experimental curves were found to be $12.93 \pm 0.01$° for the 220 and $5.88 \pm 0.01$° for 440 which are in good agreement with those of calculated convolution curves, 12.94 and 5.88° respectively. The integrated intensity of 222 was $1.59 \pm 0.1 \times 10^{-8}$. From this value the structure factor of 222 was determined to be $1.05 \pm 0.08$.

Introduction

For detailed studies on X-ray diffraction from a single crystal, it is often desirable to use a highly parallel and monochromatic X-ray beam, and for this purpose the double- or triple-crystal arrangement is conventionally used. In the double-crystal arrangement of the parallel setting the X-ray beam diffracted by the first crystal is narrow in angular spread for any component of wavelength, while the angular dispersion due to the spread in wavelength is eliminated only if the diffracting plane of the second crystal has the same spacing as that of the first crystal. On the other hand, with the triple-crystal arrangement using $(+, +, +)$ setting, an X-ray beam of a narrow spread in wavelength as well as in angle is obtained by successive diffraction from the first and second crystals. This arrangement can be applied to any diffracting planes of any crystals, in contrast to the double-crystal arrangement, because the spacings of the diffracting planes are not required to be equal, although in this case the adjustment of crystals is more difficult and the intensity is much weaker than in the double-crystal arrangement of the parallel setting.

In previous papers (Kohra & Kikuta, 1968; Kikuta, 1971a; Matsushita, Kikuta & Kohra, 1970) we described the construction of a monolithic crystal collimator consisting of two or three components with the arrangement of the parallel setting, and obtained an X-ray beam of angular spread as narrow as $0.1$° or $0.01$° by successive asymmetric diffraction from each component. Because of its easy adjustment and stable performance, this collimator has been used intensively for measurements of diffraction curves in the double-crystal arrangement of the parallel setting (Kikuta, Matsushita & Kohra, 1970; Kikuta, 1971b; Hashizume Nakayama, Matsushita & Kohra, 1970). The idea of successive diffraction by components in a monolithic crystal can also be applied to the triple-crystal arrangement by replacing the first and second crystals in it with a monolithic crystal. Kotwitz (1968, 1969) proposed the use of Umweganregung for monochromatization and collimation of a neutron beam, in which a forbidden reflexion is stimulated by two successive reflexions. The beam thus obtained is expected to have narrow angular and wavelength spreads. For X-rays, Deslattes (1968) pointed out that some combinations of diffracting planes in silicon and germanium crystals approximately satisfy the condition of successive diffraction for several characteristic X-rays, although no combination exactly satisfying this condition has been found for characteristic X-rays in common use, such as the Kα radiations of copper, molybdenum and silver.

In the present study we constructed a new monochromator–collimator crystal system consisting of two crystal elements in such a way that each element was originally a part of the same monolithic crystal. However, one of them is purposely slightly rotated using a certain device, so that the condition of successive diffraction at (620) in one element and at (333) in the other is fulfilled for Cu Kα 1. The X-ray beam obtained by this system is theoretically expected to have the wavelength and angular spreads, $\delta \lambda/\lambda$ and $\delta \omega$, of about $10^{-5}$ and $10^{-5}$ radian respectively. The construction and applications of the system are reported in this paper.

Principle

First we briefly explain the function of the first and second crystals in a triple-crystal arrangement of the $(+, +, +)$ setting such as is schematically shown in Fig. 1(a). They have the function both of monochromator and collimator at the same time. The situation is schematically shown in Fig. 1(b) using the DuMond
In this diagram the Bragg condition is represented by a band, the horizontal width of which is given by

\[ 2|\vec{b}|X_n|/\sin 2\theta_B \]  

(1)

and corresponds to the angular width of selective reflection for the wavelength concerned, where \( \theta_B \) is the Bragg angle, \( X_n \) is the \( n \)th Fourier component of the real part of the polarizability of the crystal and \( b \) is the asymmetry factor given by

\[ b = \sin (\theta_B - \beta)/\sin (\theta_B + \beta) \]  

(2)

where \( \beta \) is the angle between the crystal surface and the diffracting plane. The band I is for the first crystal and the band II is for the second. After successive diffraction by the two crystals a very narrow range of wavelength and angular divergence, corresponding to the overlapping area of the two bands, is selected. The wavelength of the X-ray beam is determined from the following relations:

\[ 2d_1 \sin \theta_{11} = \lambda \]  

\[ 2d_2 \sin \theta_{12} = \lambda \]  

\[ \theta_{11} + \theta_{12} = \alpha \]  

(3)

where \( \alpha \) is the angle between two diffracting planes, \( \theta_{11} \) and \( \theta_{12} \) are the Bragg angles and \( d_1 \) and \( d_2 \) are the spacings of the diffracting planes. The suffices 1 and 2 of \( \theta_B \) and \( d \) represent the first and second crystals respectively.

Usually these two crystals are separate crystals. However, if the geometrical angle between two diffracting planes in a monolithic crystal were equal to the sum of the Bragg angles of the two diffracting planes, an X-ray beam of a fixed wavelength would successively be diffracted as schematically shown in Fig. 2(b). Although, as mentioned before, an ideal combination of two diffracting planes cannot be found with the \( K\alpha \) radiations of copper, molybdenum and silver for crystals such as silicon and germanium, there are several combinations of diffracting planes in a monolithic crystal which approximately satisfy the condition (3). Some examples for silicon are given in Table 1, where the value 5.430866 Å is used as the lattice constant of silicon, based on the value obtained by Bond (1960) with a correction for thermal expansion, and the values of the wavelengths given by Bearden (1967) are adopted.

The slight deviation from (3) in these combinations may be eliminated by various methods, for example, by changing the lattice spacing with thermal expansion or doped impurities, by inclining the normals of the diffracting planes and by rotating one of the diffracting planes. The first method is, however, not practical because very precise control and rigorous stability are required. The second method might not be difficult but will require severe limitation of the vertical angular divergence of the beam. The last method is relatively simple and suitable for precise adjustment. Accordingly, we adopted it as described in detail in the following section.

**Construction, adjustment and performance**

A monolithic crystal system as shown in Fig. 2 was prepared from a block of silicon single crystal* free.

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Table 1. Combinations of diffracting planes for successive reflexion in silicon single crystal at 20°C

<table>
<thead>
<tr>
<th>H1</th>
<th>H2</th>
<th>( \theta_{11} )</th>
<th>( \theta_{12} )</th>
<th>( \theta_{11} + \theta_{12} )</th>
<th>( \alpha )</th>
<th>( \theta_{11} + \theta_{12} - \alpha )</th>
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</thead>
<tbody>
<tr>
<td>Cu K( \alpha )</td>
<td>333</td>
<td>620</td>
<td>47°28'32&quot;</td>
<td>63°46'16&quot;</td>
<td>111°14'48&quot;</td>
<td>111°25'0&quot;</td>
</tr>
<tr>
<td>333</td>
<td>533</td>
<td>47°28'32&quot;</td>
<td>68°26'43&quot;</td>
<td>115°55'15&quot;</td>
<td>116°7'6&quot;</td>
<td>-11°51&quot;</td>
</tr>
<tr>
<td>620</td>
<td>533</td>
<td>63°46'16&quot;</td>
<td>68°26'43&quot;</td>
<td>132°12'59&quot;</td>
<td>132°27'54&quot;</td>
<td>-14°55&quot;</td>
</tr>
<tr>
<td>Mo K( \alpha )</td>
<td>422</td>
<td>51T</td>
<td>18°39'28&quot;</td>
<td>19°50'8&quot;</td>
<td>38°29'36&quot;</td>
<td>38°13'1&quot;</td>
</tr>
<tr>
<td>422</td>
<td>155</td>
<td>18°39'28&quot;</td>
<td>27°47'52&quot;</td>
<td>46°27'20&quot;</td>
<td>46°41'10&quot;</td>
<td>-13°50&quot;</td>
</tr>
<tr>
<td>440</td>
<td>513</td>
<td>21°40'45&quot;</td>
<td>22°43'36&quot;</td>
<td>44°24'21&quot;</td>
<td>44°10'53&quot;</td>
<td>13°28&quot;</td>
</tr>
</tbody>
</table>

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* The crystal was made and supplied to us by Mr N. Akiyama of Komatsu Electronic Metals Company.
from dislocations and of high purity, which consisted of two crystal elements \( C_1 \) and \( C_{II} \) connected with a thin crystal bridge \( T \). The surfaces of \( C_1 \) and \( C_{II} \) which are parallel within 5' to (310) and (111) respectively, were polished with carborundum of 3000 mesh and etched. Then the crystal elements \( C_1 \) and \( C_{II} \) were tightly glued to metal blocks \( A \) and \( B \) respectively, as shown in Fig. 3. The blocks \( A \) and \( B \) are connected by a thin metal plate \( D \). The monolithic crystal was cut at the thin bridge \( T \). Finally \( C_1 \) was rotated around a vertical axis \( Z \) by 609-9' by bending the thin metal plate in order that the condition of successive diffraction for the 620 and 333 of \( \text{Cu} \ K\alpha_1 \) was exactly satisfied. This angle was determined in the following way. As shown in Fig. 4, a highly parallel X-ray beam of \( \text{Cu} \ K\alpha_1 \) with a wide cross section produced by a collimator utilizing successive asymmetric diffraction of 511 of silicon (Matsushita, Kikuta & Kohra, 1971) impinged on the surfaces \( x,y \) and \( z \) of the present monochromator-collimator system. The two systems were arranged in such a way that the diffracting plane (511) of the former was nearly parallel to (333) of the latter. The lattice spacings of the two are equal so that the arrangement is equivalent to the double-crystal arrangement of the parallel setting. When the present crystal system was rotated, diffraction peaks from \( x \) and \( y \) were successively observed, and the angular separation between these peaks corresponds to the rotated angle of \( C_1 \). The normal of the diffracting plane of \( C_1 \) was kept in a horizontal plane within 5" for the rotation of \( C_1 \) over an angle of 10'. This was confirmed with an autocollimator and a mirror.

If the crystal surface were distorted, the deviation from the diffraction condition would differ from place to place and the beam obtained would have wide spreads in angle and wavelength. Although \( C_1 \) and \( C_{II} \) were made thick enough to avoid deformation due to gluing, the diffraction photograph from the surface \( z \) was taken in the position of the parallel setting, as shown in Fig. 4, in order to check whether the crystal was really deformed or not. The present crystal system was set near the angular position where the diffracted intensity was about half the maximum. As the slope of the curve was steepest here and a local change in the deviation from the diffraction condition on the crystal surface could drastically cause a local variation in the diffracted intensity, an extremely small distortion of the order of \( 10^{-7} \) in \( \delta d/d \) or in inclination of the lattice plane could be observed (Bonse, 1962). In the photograph no appreciable variation was observed between before gluing and after gluing, and it was concluded that the distortion \( \delta d/d \) or the inclination of the lattice plane was less than \( 10^{-7} \).

The wavelength of the beam obtained with the present crystal system and a rotation angle of \( C_1 \) of 609-9' is estimated to be 1·54055 Å, which is within 1·5 \times 10^{-5} Å of the peak of the \( \text{Cu} \ K\alpha_1 \) spectrum. The wavelength and angular spreads, \( \delta \lambda/\lambda \) and \( \delta \omega \), are estimated to be \( 0·8 \times 10^{-5} \) and \( 1·0 \times 10^{-5} \) respectively, from the DuMond diagram. In this estimation it would be necessary to consider the dynamical effect of simultaneous diffraction if condition (3) were exactly satisfied at the original angular positions of the crystal elements without rotating one of the crystal elements.
In the present study, however, the error is not serious because the exact condition of successive diffraction is not satisfied without rotating one of the crystal elements. The X-ray beam obtained is almost perfectly polarized because the Bragg angle for the second crystal element is close to 45°. The intensity ratio of the parallel component of polarization to the normal component is estimated to be 1.2%.

The present crystal system has the following characteristics compared with the usual two separate crystals. The alignment of the second element is easily made in the present system by slightly rotating one of the elements only around one axis, while the second crystal of the separate crystals has to be aligned on a completely independent goniometer from that of the first crystal. The angle between the two crystal elements can be fixed stably, and we can treat the system as a single crystal from which we can obtain an X-ray beam of a fixed wavelength with high accuracy as well as with constant spreads in angle and wavelength.

Application to measurements of diffraction curves

Employing the present monochromator–collimator system we measured diffraction curves of 220, 440 and 222 for germanium single crystals. Specimen crystals were prepared from a single crystal with a dislocation density of about 100/cm². The surfaces of the crystals were nearly parallel to the diffracting planes to be studied. Fig. 5 shows schematically the experimental arrangement. An X-ray source with apparent focal spot size 0.8 × 0.4 mm² was operated at 50 kV and 20 mA. The intensity was measured by a scintillation counter with a single-channel pulse-height analyser. The specimen crystal was rotated with a high-precision tangential bar system.

Fig. 6 shows the measured diffraction curve of the 220 reflexion together with the calculated curve taking account of convolution. In Table 2 the half-value widths and reflexion percentage are given for the 220 and 440 reflexions. As the diffracting plane makes an angle β with the crystal surface in each specimen as tabulated in Table 2 the half-value width differs from that in the symmetric case, i.e. β = 0. We corrected the experimental value, Δθ exp, to that for symmetric diffraction, Δθ sym, according to the relation

$$Δθ_{sym} = \sqrt{b} Δθ_{exp}. \quad (4)$$

The calculated values of convolution, Δθ calc, for symmetric diffraction are also given for comparison with Δθ sym. The agreement between calculation and experiment is satisfactory in half-value width and re-

<table>
<thead>
<tr>
<th>hkl</th>
<th>β</th>
<th>√b</th>
<th>Δθ exp</th>
<th>Δθ sym</th>
<th>Δθ calc</th>
</tr>
</thead>
<tbody>
<tr>
<td>220</td>
<td>5'</td>
<td>0.9965</td>
<td>12.97 ± 0.01 (0.03)</td>
<td>12.93</td>
<td>12.94</td>
</tr>
<tr>
<td>440</td>
<td>5'</td>
<td>0.9988</td>
<td>5.91 ± 0.01 (0.03)</td>
<td>5.90</td>
<td>5.88</td>
</tr>
<tr>
<td></td>
<td>−18'</td>
<td>1.0040</td>
<td>5.84 ± 0.01 (0.03)</td>
<td>5.86</td>
<td></td>
</tr>
</tbody>
</table>

* Standard deviation of the mean. † Standard deviation.
flexion percentage. In the calculation the values calculated by Dawson (1967) using Clementi's (1965) wave functions were adopted as atomic structure factors, those of Cromer (1965) as the dispersion-correction $\Delta f'$, those of Guttman & Wagenfeld (1967) as the imaginary parts of the atomic structure factors $\Delta f''$, and the value $290^\circ K$ obtained by Batterman & Chipman (1962) as the Debye temperature.

For the 222 reflexion, only the integrated intensity was measured because the peak intensity was extremely low. The azimuth of the crystal was carefully adjusted using the diagram of Cole, Chambers & Dunn (1962) so as to avoid the effect of simultaneous diffraction. The diffraction curve is shown in Fig. 7. The diffracted intensity was as weak as 10 c.p.s at the peak, but the curve was clearly observed because of the very low background. The half-value width of the curve is nearly $2^\circ$, which corresponds to the angular and wavelength spreads of the beam obtained by the present monochromator-collimator system. The integrated intensity was measured over the angular range from $-13^\circ$ to $13^\circ$ around the peak. Outside this range the diffracted intensity did not differ from the background intensity. The value of the integrated intensity was $1.59 \pm 0.1 \times 10^{-8}$. From this value the structure factor for the 222 reflexion of germanium was determined to be 1.05, which is close to those reported in previous studies (Renninger, 1960; Colella & Merlini, 1966; Weiss, 1966). The present value of the structure factor is 5% larger than that reported previously (Matsushita, Hayashi & Kohra, 1972). This comes from the following. When we determine the structure factor from the measured integrated intensity, the calculated integrated intensity $R_w$, which is a function of structure factor, is required on the $W$-scale (DeMarco & Weiss, 1965). In the previous case the value of Weiss was used for calculating $R_w$, while in the present case we used an iteration procedure beginning with the value of Weiss until the value of the structure factor $F(n)$ obtained agreed with $F(n-1)$ to within 1%, where $n$ represents the number of iteration cycles. Accordingly, the present result is a more consistent one.

**Discussion and conclusions**

As seen in the previous section, the agreement between experiment and calculation proves that the X-ray beam obtained from the present monochromator-collimator system is highly parallel and monochromatic, as theoretically expected. Moreover the crystal system is very compact in size and mechanically stable.

Many other combinations of two diffracting planes for successive diffraction of characteristic X-rays may be found for crystals such as silicon, germanium and quartz if an allowance of several degrees is allowed as the deviation from the condition of successive diffraction. Such a large deviation can be compensated without much difficulty by the present technique.

By some modifications of the crystal system it would be possible to make the angular and wavelength spreads of the X-ray beam still sharper or narrower. The use of successive symmetric diffraction between parallel walls of a groove (Bonse & Hart, 1965, 1966) for each crystal element, as shown in Fig. 8(a), would make the intensity distribution tailless both in angle and wavelength. Furthermore, the arrangement of asymmetric diffraction in such a way as shown in Fig. 8(b) would make them narrower. In the use of successive asymmetric diffraction, it is important to note that the number of successive diffractions between walls for the second crystal element should be even. Otherwise, the spectral window* is deformed to be like a line, as pointed by Nakayama (Nakayama, Hashizume, Miyoshi, Kikuta & Kohra, 1973).

With the stable performance of the system and the good quality of the beam, it is concluded that the present system can be extensively used for the measurement of diffraction curves of any diffracting plane of any crystal in place of the first and second crystals of the triple-crystal arrangement. An example of the application has been briefly reported previously (Matsushita, Hayashi & Kohra, 1972) and a more detailed study will be reported elsewhere in the near future.

* The overlapping area of the bands I and II in Fig. 1 is called the spectral window.
Fig. 8. The use of successive diffractions between parallel walls in each crystal element. One of the elements may be rotated by the present technique if necessary. (a) Symmetric diffraction. (b) Asymmetric diffraction.

The author would like to express his sincere gratitude to Professor K. Kohra for his guidance and continual encouragement. He would like to thank Drs S. Kikuta and K. Nakayama for their helpful discussions and Mr N. Akiyama for supplying a silicon single crystal of very high quality. The assistance of Messrs H. Ishida and J. Hayashi in the experiment is also acknowledged.

References

Distortion of X-ray Small-Angle Scattering Curves Measured by a Levelut–Guinier Camera*

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The applicability of the Levelut–Guinier camera to X-ray scattering measurements is considered with respect to the error introduced by the distortion of the true scattering curves. The distortion of different types of curves, representing different scattering systems, was found by calculation, and the character and amount of distortion are discussed.

1. Introduction

A camera for detecting weak X-ray scattering at small angles has recently been developed (Levelut & Guinier, 1967; Levelut, 1968). The constructors compared the possibilities of the camera with those of the usual diffractometric or photographic devices. The very good sensitivity of the new device makes it possible to study the distribution of point defects in crystals which previously were not detectable by conventional small-angle apparatus. However, the possible distortion of experimental curves has not been discussed quantitatively in detail. Until some method for correcting experimental scattering curves becomes available, knowledge of the character and amount of distortion may help in the evaluation of scattering curves.

The principle of measurement is presented in Fig. 1(a) for a perfectly collimated primary beam, i.e. a beam of negligible divergence and negligible cross section. The radiation scattered on the specimen S is limited by two circular screens, the radius of the inner one being \( r_1 \) and the outer one \( r_2 \), which determine the ring-