Acta Cryst. (1979). A35, 962–965

Simultaneous Diffraction: *Umweganregung* Peaks in the Case of a Vibrating Single Crystal

BY P. MIKULA, R. T. MICHALEC, M. VRÁNA AND J. VÁVRA

Nuclear Physics Institute of the Czechoslovak Academy of Sciences, 250 68 Řež near Prague, Czechoslovakia

(*Received* 1 *November* 1978; *accepted* 18 *June* 1979)

Abstract

The positive diffraction intensity changes on forbidden (222) planes of a vibrating single crystal of Si, which are produced by simultaneous diffraction, are experimentally treated for the symmetric Laue case. The neutron diffraction experiment was performed for a flexurally vibrating as well as a non-vibrating perfect silicon bar. Several *Umweganregung* peaks were observed after exciting the bar into vibration while no *Umweganregung* peak was observed in the case of a non-vibrating crystal.

1. Introduction

It is known that the effect of simultaneous reflections can be observed when a crystal lattice is oriented in such a way that more than one set of planes are operative for a given wavelength of the X-ray or neutron beam. Using the terminology of reciprocal-lattice space, the effects of simultaneous reflections occur when a second lattice point (or more) defined by a scattering vector $\mathbf{g}_2$ is brought onto the sphere of reflection, *e.g.* by crystal rotation around the scattering vector $\mathbf{g}_1$ of a particular reflection for which the diffractometer is set. The occurrence of simultaneous reflections may reduce the intensity of a strong primary reflection (*Aufhellung*) or, on the other hand, increase the intensity of a weak reflection (*Umweganregung*). The extreme form of the *Umweg* effect is a simulation of a forbidden particular reflection in the following way. The reflected beam produced by the second set of planes associated with $\mathbf{g}_2$ may be in turn reflected by a third such set of planes for which the corresponding scattering vectors $\mathbf{g}_3$ are related to $\mathbf{g}_1$ and $\mathbf{g}_2$ by

$$\mathbf{g}_1 - \mathbf{g}_2 = \mathbf{g}_3.$$

So the double reflected beam has the same direction as a beam that could have been reflected by the particular set of planes. Of course, both the second and third set of planes must have non-zero structure factors.

Positive *Umweg* peaks were first observed by Renninger (1937, 1955, 1960). The principal theoretical investigation in this field was first performed by Ewald (1937). After the famous experimental work of Borrmann & Hartwig (1965), the dynamical theory, which has no restrictions on the number of excited reflections, has usually been applied to multiple diffraction so that the situations in which more than one set of atomic planes are in position to transmit X-rays anomalously could be investigated (Saccocio & Zajac, 1965; Hildebrand, 1967; Joko & Fukuhara, 1967; Penning, 1968; Iveronova, Katsnelson & Runova, 1977; Post, Chang & Huang, 1977). In the paper of Ewald & Héno (1968), there is also discussed the problem of direct *Umweganregung* phenomena. The problem of simultaneous reflections in the mosaic crystals is theoretically and experimentally treated in the papers of Moon & Shull (1964) for neutrons and Caticha-Ellis (1969) for X-rays. The indexing of the *Umweg* peaks for the crystals of diamond structure is reported in the papers of Cole, Chambers & Dunn (1962) and Kotwitz (1968).

Usually, the flux of thermal neutrons is not sufficient to produce on perfect crystals anomalous effects such as *Aufhellung* and *Umweganregung* and the changes of the anomalous transmission cannot be experimentally observed. Greater success may be achieved in the case of a mosaic crystal. Assuming the possibility of an observation of the positive peaks on the characteristics of the observed pattern, the decisive influence of the sample geometry was pointed out by Moon & Shull (1964). On a mosaic single crystal of Fe, they observed only one *Umweg* peak in Laue geometry.

Besides a very few experiments performed with neutrons on mosaic crystals, up to now we have not found any paper dealing with exciting *Umweg* peaks on perfect crystals by an elastic deformation, especially in the Laue case. The homogeneous bending (Klar & Rustichelli, 1973), vibrations (Michalec, Sedláková, Chalupa, Čech, Petržílka & Zelenka, 1974; Mikula, Michalec, Chalupa, Sedláková & Petržílka, 1975) and temperature gradient (Seiler & Dunitz, 1978) are

0567-7394/79/060962-04$01.00

usually used for elastic deformation on single crystals. Being interested in the effects in the symmetric Laue case, we decided to use flexural vibrations to exclude the direct promoting of simple 222 as well as higher order reflections by elastic deformation.

According to the dynamical theory of diffraction for a distorted crystal where a deformation is represented by a displacement **u**, the change of the integral intensity diffracted by a sample is a function of a scalar product $(\mathbf{g}.\mathbf{u})$. In our case the scalar product $(\mathbf{g}_1.\mathbf{u})$ is equal to zero for both 222 and its higher order reflections. Consequently, for these reflections the crystal behaves as a perfect one (Takagi, 1969). It follows from this fact that the 'background' to the possible *Umweg* effects may not depend on the value of the vibration amplitude. On the other hand, for $\mathbf{g}_2.\mathbf{u} \neq 0$, as well as $\mathbf{g}_3.\mathbf{u} \neq 0$, for the second and third set of planes, the crystal behaves as a distorted one. So the distortion in our case may entail an extension of the range of the wave vectors from the incident beam participating in the process of simultaneous and double diffraction.

2. Experimental arrangement

The measurements were carried out on a double-axis spectrometer (Michalec, Vavřín, Chalupa & Vávra, 1967). Using a mosaic monochromator Zn, a beam of nearly monoenergetic neutrons with wavelength $\lambda \simeq 1{\cdot}05$ Å impinged on the flexurally vibrating silicon bar at the Bragg angle for the forbidden 222 diffraction. A schematic drawing of the experimental arrangement is given in Fig. 1. The crystal bar was cut from a perfect single crystal of Si and was worked by mechanical polishing as well as by chemical treatment. The dimensions of the bar were 200 × 30 × 6 mm. The bar was excited into flexural mode vibrations with the fundamental resonance frequency 1·4 kHz. Vibrations were excited by pondermotive forces between the excitation electrode and the bar. This method of excitation is well described in the paper of Petržílka, Vrzal, Michalec, Chalupa, Mikula & Zelenka (1970). The elastic displacement **u** took place in the direction parallel to the reciprocal-lattice vector $[1\bar{1}0]$. The value of the vibration amplitude was measured with a microscope in the middle of the bar, *i.e.* at the point of maximum elastic displacement.

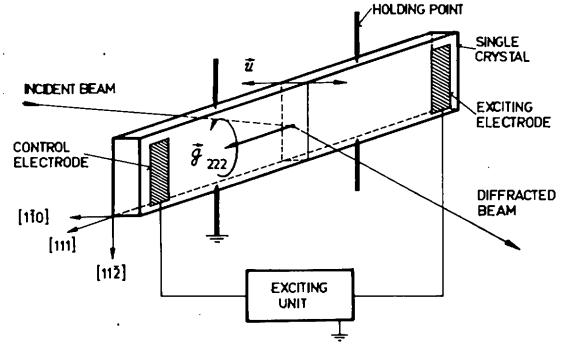

Fig. 1. Schematic arrangement for neutron diffraction by flexurally vibrating a silicon single crystal in the 222 diffraction setting with respect to the crystallographic system of coordinates. $\vec{g}_{222}$, reciprocal-lattice vector, $\vec{u}$, elastic displacement of the lattice planes.

The first part of the experiment was performed with an unscreened beam with dimensions 40 × 40 mm with a rather poor angular and vertical divergence. Then the collimated and screened beam of 10 × 30 mm with the horizontal divergence of 20′ was used.

For the rotation of the bar around the scattering vector [222] (see Fig. 1), the goniometer head was used.

3. Experimental results

Fig. 2 shows the rocking curves of non-vibrating (*a*) and vibrating (*b*) crystals for the angle $\varphi = 0°$ of rotation of the crystal round the diffraction vector [222]

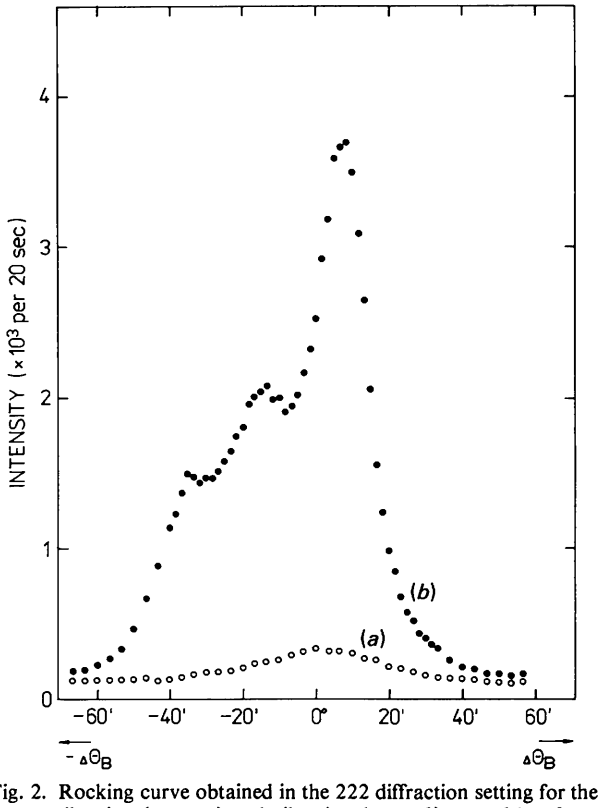

Fig. 2. Rocking curve obtained in the 222 diffraction setting for the non-vibrating (curve *a*) and vibrating (curve *b*) crystal bar for a wide neutron beam with horizontal and vertical divergence of about 1° and for azimuthal angle $\varphi = 0°$. Curve (*a*) corresponds to the diffraction of $\lambda/2$ neutrons on the (444) set of planes. Vibration amplitude u_0 was 30 μm.

(here, φ will be called the azimuthal angle). The wide uncollimated beam 40 × 40 mm with the horizontal divergence of 1° was used. In our experiment the azimuthal angle φ means a deviation of the lattice vector [1$\bar{1}$0] from the plane determined by the incident beam and the lattice vector [222].

The curve corresponding to the non-vibrating crystal is due to the diffraction of $\lambda/2$ neutrons on the set of planes (444). This higher order reflection was used for correct setting of the experimental apparatus. The rocking curve of a vibrating single crystal illustrates the *Umweg* effect on the rocking curve which is formed by several simultaneous reflections.

Fig. 3 presents the dependence of the diffracted intensity on the azimuthal angle φ for the uncollimated beam. Curve (*a*) corresponds to the non-vibrating crystal and curves (*b*), (*c*), (*d*) to the vibrating crystal with vibration amplitudes 4, 18 and 30 μm, respectively.

Fig. 4 shows the rocking curve of the vibrating single crystal obtained after the incident beam had been collimated to 20′ and screened to the width of 10 mm and for $\varphi = 0°$. The rocking curve of the non-vibrating crystal, corresponding to the $\lambda/2$ neutrons, was not observable.

Fig. 5 presents the intensity dependence on the azimuthal angle φ for the collimated and screened beam. Curves (*a*) and (*b*) correspond to the non-vibrating and vibrating crystal, respectively.

The use of the vibrating crystal in connection with a time-of-flight method in our experiment enabled us to verify very simply if the newly excited peaks corresponded to wavelength λ or $\lambda/2$ *etc.* As the intensity of neutrons diffracted by the vibrating single crystal is time modulated (see Fig. 6), it is possible to determine the wavelength very easily by a simple change of the flight path between the vibrating crystal and the detector. Such an investigation was performed for every peak corresponding to the vibrating crystal in the above mentioned figures. It was found that the whole

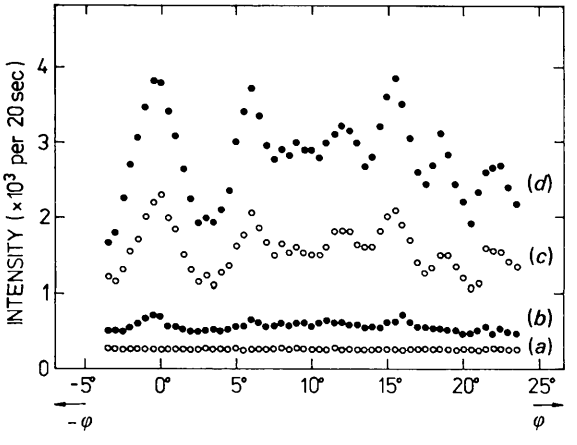

Fig. 3. The dependence of the diffracted intensity on the azimuthal angle φ in the 222 diffraction setting for different vibration amplitudes of the crystal bar. (*a*) Non-vibrating crystal; (*b*) (*c*) and (*d*) vibrating crystal with vibration amplitudes 4, 18 and 30 μm, respectively. In this case the wide beam with 1° collimation was used.

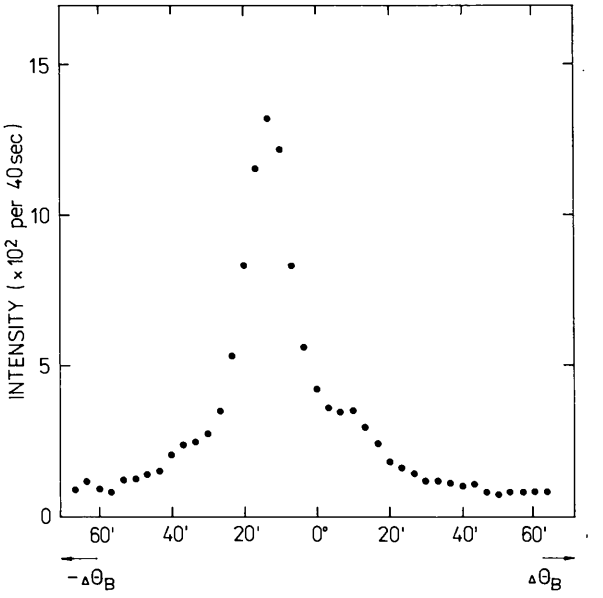

Fig. 4. Rocking curve obtained in the 222 diffraction setting for the vibrating crystal bar with the improved horizontal collimation of 20° and azimuthal angle $\varphi = 0°$. The vibration amplitude was 30 μm.

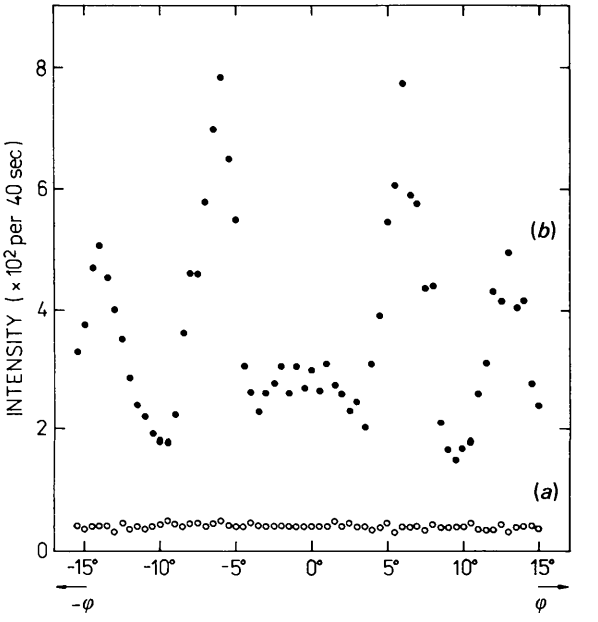

Fig. 5. The dependence of the diffracted intensity on the azimuthal angle φ in the 222 diffraction setting using the beam with the improved collimation of 20′. Curve (*a*) non-vibrating crystal: curve (*b*) vibrating crystal with the vibration amplitude of 30 μm.

pattern of the azimuthal dependence could be assigned to the neutrons of wavelength λ.

Fig. 6 illustrates the time modulation taken in the position of the *Umweg* peak at $\varphi = 18° 30'$.

4. Discussion

Comparing Figs. 2 and 4 and Figs. 3 and 5, we can see the dependence of the occurrence of a member of *Umweg* peaks on the width of the wavelength band given by the wavelength resolution. We performed a computer calculation of the azimuthal positions of the secondary reflections in the interval from 1·03 to 1·07 Å which just corresponds to the wavelength resolution $\Delta\lambda$ of the unscreened and uncollimated beam. About 60 operative secondary reflections were determined in the interval of the azimuthal angles from 0 to 30°. Their mutual azimuthal operative positions usually change strongly with their dependence on λ in the above-mentioned range $\Delta\lambda$. So the detailed comparison of the calculated results with the experimental ones is not possible. The azimuthal positions of the maximum and minimum density of the operating points in the $\Delta\lambda$ range were found to be in good agreement with the experimentally observed positions of peaks and minima on the *Umweg* patterns shown in Figs. 3 and 5. As was pointed out in the *Introduction*, the higher orders of the forbidden 222 reflection do not depend on the vibrating amplitude because for them the crystal behaves as a perfect one, so the diffracted intensity corresponding to the higher orders is included in the background to the possible *Umweg* effects. It follows from this and from our experimental results that the diffracted intensity corresponding to the excited *Umweg* peaks may be many times higher than the one corresponding to the higher order reflections.

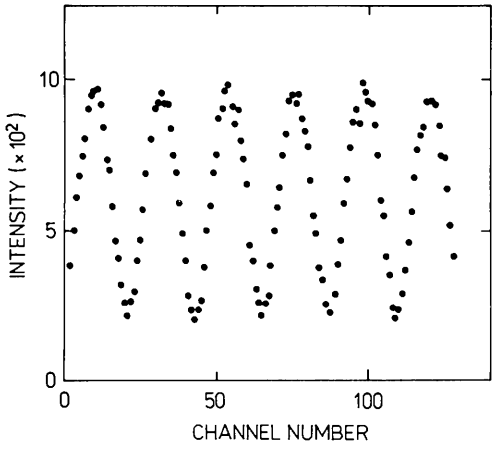

Fig. 6. Time modulation of neutrons diffracted in the 222 diffraction setting for azimuthal angle $\varphi = 18° 30'$ using the neutron beam with 1° collimation. The vibration amplitude was 30 μm and measuring time was 15 min.

5. Conclusions

The present data indicate a significant influence of crystal deformation on the process of simultaneous diffraction and the necessity of considering the presence of simultaneous reflections in accurate measurements of particular reflection intensities from single crystals. On the other hand, we give a simple scheme to show how the effects of such a process can be made more observable.

Further theoretical and experimental investigations in this direction are being carried out.

The authors wish to thank Š. Rabatin for his valuable help throughout the measurements and to B. Hašková and A. Dvořák for their help in preparing the manuscript.

References

BORRMANN, G. & HARTWIG, W. (1965). *Z. Kristallogr.* **121**, 401–409.
CATICHA-ELLIS, S. (1969). *Acta Cryst.* A**25**, 666–673.
COLE, H., CHAMBERS, F. W. & DUNN, H. M. (1962). *Acta Cryst.* **15**, 138–144.
EWALD, P. P. (1937). *Z. Kristallogr.* A**97**, 1–27.
EWALD, P. P. & HÉNO, Y. (1968). *Acta Cryst.* A**24**. 5–15, 16–42.
HILDEBRAND, G. (1967). *Phys. Status Solidi*, **24**, 245–261.
IVERONOVA, V. I., KATSNELSON, A. A. & RUNOVA, T. K. (1977). *Kristallografiya*, **22**, 939–945.
JOKO, T. & FUKUHARA, A. (1967). *J. Phys. Soc. Jpn*, **22**, 597–604.
KLAR, B. & RUSTICHELLI, F. (1973). *Nuovo Cimento*, **13B**, 249–271.
KOTWITZ, D. A. (1968). *Acta Cryst.* A**24**, 117–126.
MICHALEC, R., SEDLÁKOVÁ, L., CHALUPA, B., ČECH, J., PETRŽÍLKA, V. & ZELENKA, J. (1974). *Phys. Status Solidi A*, **23**, 667–673.
MICHALEC, R., VAVŘÍN, J., CHALUPA, B. & VÁVRA, J. (1967). Report ÚJV 1562, Nuclear Research Institute, Prague-Řež.
MIKULA, P., MICHALEC, R., CHALUPA, B., SEDLÁKOVÁ, L. & PETRŽÍLKA, V. (1975). *Acta Cryst.* A**31**, 668–693.
MOON, R. M. & SHULL, C. G. (1964). *Acta Cryst.* **17**, 805–812.
PENNING, P. (1968). *Philips Res. Rep.* **23**, 1–11, 12–24.
PETRŽÍLKA, V., VRZAL, J., MICHALEC, R., CHALUPA, B., MIKULA, P. & ZELENKA, J. (1970). *Phys. Status Solidi*, **42**, 895–902.
POST, B. CHANG, S. L. & HUANG, T. C. (1977). *Acta Cryst.* A**33**, 90–97.
RENNINGER, M. (1937). *Z. Phys.* **106**, 141–176.
RENNINGER, M. (1955). *Acta Cryst.* **8**, 606–610.
RENNINGER, M. (1960). *Z. Kristallogr.* **113**, 99–103.
SACCOCIO, E. J. & ZAJAC, A. (1965). *Phys. Rev.* **139**, A255–A264.
SEILER, P. & DUNITZ, J. D. (1978). *Acta Cryst.* A**34**, 329–336.
TAKAGI, S. (1969). *J. Phys. Soc. Jpn*, **26**, 1239–1253.