Coherent Interaction of Multiple Diffracted X-Rays in Crystals

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Dedicated to Prof. Dr. G. Hildebrandt at his 60th birthday

Coherent dynamical interaction in a 6-beam Borrmann diffraction experiment leading to an unusual intensity enhancement of X-ray transmission in crystals is investigated by using a synchrotron radiation source. A sharp enhanced intensity spot of two seconds of arc beam-divergence is observed on the direct beam at the exact 6-beam diffraction position. The plane-wave dynamical theory is employed to account for the experimental results. The polarization dependence of the crystal excitation by the incident beam is discussed.

Introduction

Coherent interaction of X-rays takes place in crystals through multiple diffraction, where several sets of atomic planes diffract simultaneously an incident beam. Under such conditions the transmission of X-rays through a perfect crystal is anomalously high [1–6]. In the (000) $(0\bar{4}4)$ $(2\bar{2}0)$ $(\bar{2}02)$ $(2\bar{4}2)$ $(\bar{2}\bar{2}4)$ Borrmann diffraction case unusually bright trans-beams suffering zero absorption should be, in principle, observable at the exact 6-beam diffraction position [2]. Attempts have been made to observe this predicted phenomenon, but only with limited success [7–9].

In this paper, we report on such an anomalous transmission of X-rays through a perfect silicon crystal on the direct incident beam of linearly polarized synchrotron radiation. The present results will be compared with those obtained with an unpolarized X-ray source [7], [9].

Experimental

The experiment was carried out at the Hasylab of DESY. Figure 1 shows the experimental arrangement, which is similar to that reported by Huang, Tillinger and Post [7]. The synchrotron radiation of the DORIS storage ring together with a collimation system produces an incident beam with an angular divergence of about 20 seconds of arc. The linear π-polarization lies in the plane of Figure 1. The experiment is performed by aligning a [111] cut, plate-like perfect silicon crystal for $(0\bar{4}4)$ reflection at the wavelength $\lambda = 1.0$ Å. The rest of the involved reflections then simultaneously satisfies Bragg's law. The crystal thickness is 3 mm. A Laue transmission photograph (see Fig. 2) is taken to check the crystal alignment at this wavelength. Figure 2 is obtained by placing a film 5 cm behind the crystal parallel to the crystal plate. Six-beam diffraction is seen. The direct beam, (000), is attenuated by a 1 mm thick lead absorber and a 3 mm thick aluminium plate. The camera is also shielded by aluminium and lead foils to supress the high background caused by the white radiation. The enhanced transmitted intensity on the direct beam (Fig. 2) is readily detectable on the original film. When the crystal is aligned for $\lambda = 1.54$ Å by setting $(0\bar{4}4)$ at the Bragg angle for this wavelength, the intensity enhancement of the direct transmitted beam at the 6-beam point is reinforced. This is shown in Fig. 3, which is taken by placing a film, perpendicular to the direct beam, 20 cm away from

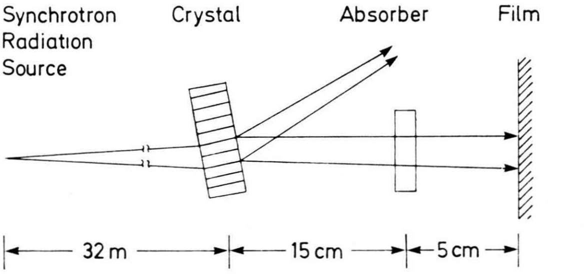

Fig. 1. Schematic representation of the experimental arrangement.

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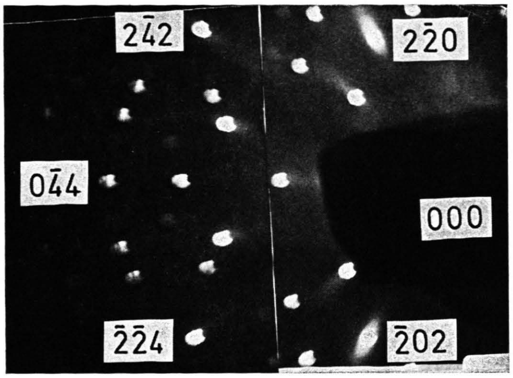

Fig. 2. Laue transmission photograph of a silicon crystal for the six-beam case at $\lambda = 1.0$ Å. The six diffracted spots form a regular hexagon.

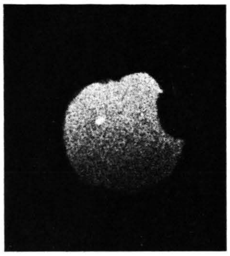

Fig. 3. Image of the direct (000) beam at the exact 6-beam diffraction position for $\lambda = 1.54$ Å.

the crystal. The angular divergence of the enhanced spot is estimated to be about 2 seconds of arc by comparing the diameter of the spot and that of the direct beam in Figure 3. The shape of the direct beam results from the geometry of the collimator at the exist of the synchrotron beam line.

Calculations

The computer program for n-beam dynamical calculations [6] is employed to account for the observed results. This program is based on the plane-wave dynamical theory of Ewald [10], modified by von Laue [11], and generalized by Kato [12] for the n-beam cases. The π-polarization of the synchrotron radiation is considered in the calculations.

Table 1. The linear absorption coefficient μ_0 along the direction of the incident beam, the percentage of mode excitation and the resonance failure g for a π-polarized beam ($\varphi = 0$).

Mode	μ_0 (cm^{-1})	Excitation (%)	g (cm^{-1})
1	0.03	0.0	246.57
2	0.19	10.1	251.33
3	0.19	0.1	251.33
4	0.87	16.7	275.90
5	13.17	4.0	441.15
6	13.17	6.6	441.15
7	16.54	18.0	514.68
8	16.54	5.1	514.68
9	24.28	16.7	637.34
10	521.66	0.0	2083.26
11	543.48	6.1	2111.65
12	543.48	16.6	2111.65

Table 1 gives, at the exact six-beam diffraction position for each mode of propagation, the linear absorption coefficient μ_0, excitation of modes (the fraction of the incident energy associated with a given mode), and the resonance failure g (see [10]). Only 9 of the 12 modes are appreciably excited by the incident beam during the occurence of the 6-beam diffraction. This selection of modes is due to the linear π-polarization. For a thick crystal, only the first four modes can contribute to the transmitted intensity. The rest of the modes is suppressed by the crystal absorption. In the present study, only modes 2 and 4 are effective. Mode 2 has minimum intensity at the atom sites, while mode 4 has maximum intensity near (but not at) the atom sites [9]. The wavevector difference of the two modes is only 24.55 cm^{-1} in the [111] direction. The intensity distribution of these two modes projected onto the (111) planes at the exact 6-beam point can be seen from Figs. 4(b) and 4(c) of [9], except that the intensity maxima are squeezed along the $[0\bar{1}1]$ direction due to the π-polarization.

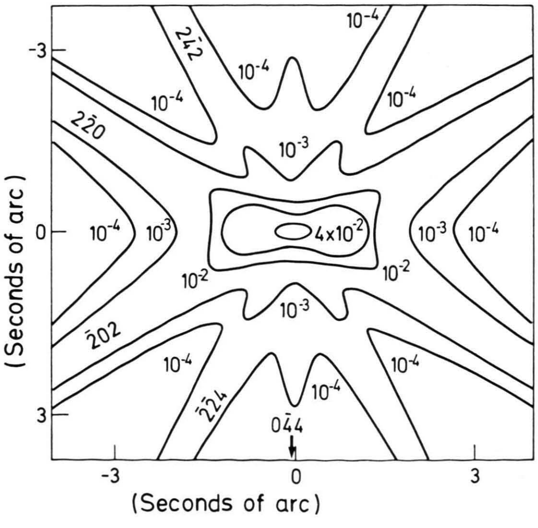

Fig. 4. Calculated intensity distribution of the direct (000) beam in the vicinity of the 6-beam point for $\lambda = 1.54$ Å.

The calculated transmitted intensity of the (000) reflection is shown in Figure 4. Diffracted intensities less than 10^{-2} are too weak to be observed in Figure 3. For the crystal settings slightly off the 6-beam position (i.e. 1 second of arc), the absorption of the two modes (modes 2 and 4) increases by a factor of 30. Thus, a sharp intensity enhancement within a 2 seconds of arc angular range is expected and observed experimentally in Figure 3. According to [9], this intense beam is nearly divergenceless, monochromatic and spatially coherent.

Discussions

Since the experiment involves a linearly polarized radiation source, it would be appropriate to discuss, based on the dynamical calculation, the effect of polarization on the mode-excitation of the crystal. On the other hand, the calculated absorption coefficients and the excitation of the modes depend strongly on the imaginary and the real parts of the structure factors, respectively. Therefore, the influence of the input structure factors on the calculation should also be discussed.

(*A*) *Polarization*

Suppose that φ is the angle between the reference π-vector (representing the π polarization) and the resultant electric field of the incident wave. The angle φ is zero for a π-polarized field and 90° for a σ-polarized field. For an unpolarized beam, $\varphi = 45°$. Table 2 shows the polarization effect on the excitation of the first four lowest absorbing modes with an linear absorption coefficient μ_0 along the incident beam of less than 1 cm^{-1}. For the π-polarization ($\varphi = 0$), only modes 2 and 4 are strongly excited, while for the σ-polarization modes 1 and 3 are effective. The transition of the excitation from modes 2 and 4 to modes 1 and 3 is clearly seen, when φ varies from 0° to 90°. For an unpolarized beam, these four modes are nearly equally excited. Since the absorption of these 4 modes is related to the absorption of the (220) reflection [9], it would be very difficult to observe only a sharp tiny enhanced intensity at the exact 6-beam point. The diffracted ($2\bar{2}0$) and ($\bar{2}02$) intensities will certainly appear in the vicinity of the 6-beam point [7].

Table 2. The relation between the incident polarization angle φ and the excitation of modes 1, 2, 3 and 4.

φ (deg.)	Excitation of modes (%)			
	1	2	3	4
0	0.0	10.1	0.1	16.7
10	0.5	9.8	0.8	16.2
20	1.9	9.0	2.8	14.7
30	4.2	7.7	5.8	12.5
40	6.9	6.0	9.5	9.8
45	8.3	5.2	11.5	8.3
50	9.8	4.3	13.4	6.9
60	12.5	2.7	17.1	4.2
70	14.7	1.4	20.2	1.9
80	16.2	0.6	22.1	0.5
90	16.7	0.3	22.8	0.0

Reflection	Structure Factor F at 25 °C	
(hkl)	F'	F''
000	113.952	2.604
202	69.534	2.524
242	50.759	2.372
440	44.524	2.308

Table 3. Structure factor $F = F' + i \cdot F''$ of the involved reflections in the 6-beam case, at $T = 25$ °C and $\lambda = 1.54$ Å.

(*B*) *Structure Factors*

The structure factors $F = F' + i \cdot F''$ (F': real part, F'': imaginary part) of the involved reflections listed in Table 3 are used for the dynamical calculations. As pointed out by Joko and Fukuhara [2], the lowest linear absorption coefficient μ at the exact 6-beam point is

$$\mu = \varkappa (F''_{000} - F''_{202} - F''_{224} + F''_{044}) \approx 0\,, \quad (1)$$

where $\varkappa$ is a proportionality constant. A slight variation in F'' can result in a negative absorption. In the calculation, a variation of 0.001 in F'' gives a deviation of approximately 0.2 cm^{-1} in μ. In addition, a deviation of 0.002 in the real part F' of the structure factor varies the excitation of the mode by a maximum factor of 4. In comparison to Ref. [7] and [9], the μ for silicon is different from that for germanium, mainly because silicon is less absorbing than germanium at the wavelength used.

(*C*) *Polarization of the* (*000*) *Transmitted Beam*

If the resultant transmitted wave in [000] direction is written as $|E|\, e^{i\beta}$, the polarization angle β should be closely related to the polarization of the incident beam. The relation between β and φ, which is a function of the crystal thickness, is given in

Table 4. The relation between the incident polarization angle φ and the transmitted polarization angle β for different crystal thickness t.

φ	β (deg.)		
(deg.)	$t = 0.1$ cm	$t = 0.2$ cm	$t = 0.3$ cm
0	− 180.0	0.0	180.0
10	− 138.3	5.8	158.4
20	− 118.5	11.8	140.8
30	− 108.9	18.3	127.7
40	− 103.3	25.6	118.0
45	− 101.2	29.7	114.0
50	− 99.4	34.3	110.5
60	− 96.5	44.7	104.4
70	− 94.1	57.5	99.2
80	− 92.0	72.9	94.5
90	− 90.0	90.0	90.0

Table 4. For $\varphi = 0°$ and $90°$, the transmitted beam is also linearly polarized.

Conclusion

The observation of the intensity enhancement at the exact 6-beam diffraction point is due mainly to the linearly polarized radiation source and the perfect silicon crystal. A similar intensity enhancement should be observable for a perfect germanium crystal. The experimental arrangement can be used as an X-ray mode-limiter. A single-mode excitation can be achieved if a thick perfect crystal is employed. The anomalously transmitted beam at the exact 6-beam diffraction position provides a new source of monochromatic X-rays with exceedingly low divergence and single-mode excitation for X-ray diffraction and imaging studies.

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