Acta Cryst. (1993). A**49**, 573-575

X-ray Dynamical Diffraction in Ge with a Zero-Real-Part Scattering Factor

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(*Received* 2 *July* 1992; *accepted* 24 *November* 1992)

Abstract

X-ray dynamical diffraction induced only by the imaginary part of the scattering factor was measured using a Ge perfect crystal. The 844 integrated reflecting intensities near the K-absorption edge were measured in both the Bragg and the Laue cases. The intensities show the characteristic variations for the scattering factor having no real part, which agree well with theoretical predictions. There remains a slight difference between the theoretical [Fukamachi & Kawamura (1993). *Acta Cryst.* A**49**, 384-388] and the experimental energy position at which this occurs, which is related to the fine structure of the anomalous scattering factor above the absorption edge.

1. Introduction

With synchrotron radiation (SR), it is easy to vary the X-ray energy across a specified absorption edge of an atom. Several studies have been carried out with SR to measure the reflected intensity from a perfect crystal across the absorption edge of the constituent atom. In these analyses, dynamical theories of diffraction that take the absorption effect into account were used. In some of the theories, however, the calculated reflected intensity diverges greatly from the measured value when the real part of the scattering factor becomes zero, which is expected to occur near the absorption edge. Fukamachi & Kawamura (1993) have derived simple formulae for reflection intensities that are applicable even when the real part of the scattering factor is zero. The formulae are given for both the Bragg case and the Laue case. The results show a very sharp rocking curve in the Bragg case and a *Pendellösung* fringe induced by the imaginary part of the scattering factor in the Laue case. Here, the integrated reflecting intensities (IRIs) are measured from a Ge perfect crystal across the K-absorption edge to study the IRIs when the real part of the scattering factor is zero: the IRIs are induced only by the imaginary part in both the Bragg and the Laue cases.

2. Theoretical basis

The atomic scattering factor f is given by

$$f = f^0(\mathbf{h}) + f'(\omega) + if''(\omega), \tag{1}$$

where $f^0(\mathbf{h})$ is the normal scattering factor and $f'(\omega)$ and $f''(\omega)$ are the real and imaginary parts of the anomalous scattering factor, respectively. $f^0(\mathbf{h})$ is a function of the reciprocal-lattice vector $\mathbf{h}$ in general and is approximated to be $f^0(\mathbf{h}) = f^0(|\mathbf{h}|)$ when the anisotropy is small enough. $f^0(|\mathbf{h}|)$ is a monotonically decreasing function of $|\mathbf{h}|$ and has its maximum value at $|\mathbf{h}| = 0$.

To satisfy the condition

$$f^0(|\mathbf{h}|) \le |f'(\omega)|, \tag{2}$$

we have to choose a high-index reflection (large $|\mathbf{h}|$) and use the X-ray energy ω very close to the absorption edge. For a Ge crystal, the highest measurable reflection hkl must satisfy $h^2 + k^2 + l^2 \le 100$ [with $|\mathbf{h}| = 2\pi/d(hkl)$, where d is the spacing of the (hkl) planes] for the X-ray energy around the K-absorption edge. $f''(\omega)$ and $f^0(|\mathbf{h}|) + f'(\omega)$ are shown in Fig. 1 for the 844 reflection ($h^2 + k^2 + l^2 = 96$). The values in Fig. 1 are calculated by the method of Parratt & Hempstead (1954) with the oscillator strength of Cromer (1965). The damping factor is 1 eV for the solid line and 3 eV for the dashed line. The value of 1 eV is the value calculated by the classical theory (Parratt & Hempstead, 1954). When the damping factor is 1 eV, the value of $f^0(|\mathbf{h}|) + f'(\omega)$ becomes zero at an energy ± 1.3 eV from the edge. Between -1.3 and 1.3 eV, $f^0(|\mathbf{h}|) + f'(\omega)$ is negative. When the damping factor

0108-7673/93/030573-03$06.00

is 3 eV, $f^0(|\mathbf{h}|)+f'(\omega)$ is always positive across the edge. Since the actual anomalous scattering is not known for a Ge crystal, we have to examine whether the relation $f^0(|\mathbf{h}|)+f'(\omega)\leq 0$ is satisfied or not by measuring the diffraction intensity. The anomalous scattering factors calculated by Cromer & Liberman (1970) are not used because the values show discontinuous change at the absorption edge and are not appropriate to study the intensity variation close to the absorption edge.

The integrated reflecting powers (IRPs) in the energy-dispersive mode are calculated for the 844 reflection from a Ge perfect crystal as a function of energy across the K-absorption edge. The IRPs in the symmetric Bragg case are shown in Fig. 2 (solid line) and those in the symmetric Laue case in Fig. 3 (solid and dashed lines). The IRPs were obtained by numerically integrating the intensity given by Fukamachi & Kawamura (1993). When the incident X-ray intensity does not vary by changing the X-ray energy, the IRI is proportional to the IRP and the comparison between theory and experiment is straightforward. The temperature factor in the calculation was the value measured by Batterman & Chipman (1962).

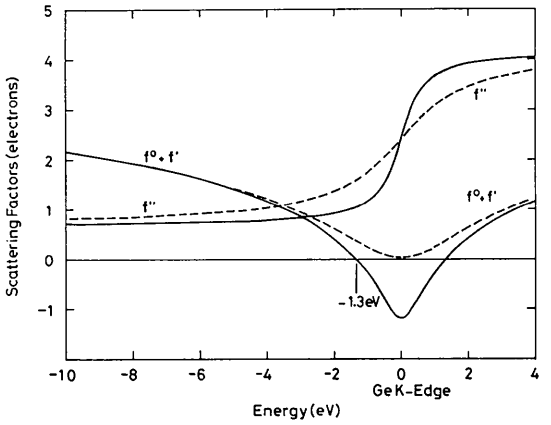

Fig. 1. The real $f^0(|\mathbf{h}|)+f'(\omega)$ and the imaginary $f''(\omega)$ parts of the scattering factor for the 844 reflection near the K-absorption edge in Ge. The unit of the scattering factor is the number of electrons.

In Fig. 2, the calculated IRP with the damping factor of 1 eV decreases monotonically as a function of energy below the absorption edge. The IRP has its minimum at 1.5 eV below the absorption edge, which is 0.2 eV lower than the energy at which the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ is satisfied. It shows a steep increase across the absorption edge due to the change of $f''(\omega)$. For the energy at which $f^0(|\mathbf{h}|)+f'(\omega)=0$ is satisfied above the edge, there is no significant change in the intensity. This is because the value of $f''(\omega)$ is large enough to smear out the change of $f^0(|\mathbf{h}|)+f'(\omega)$.

In Fig. 3, the solid line shows the IRPs calculated with the assumption that the crystal thickness is 39.3 μm and the damping factor is 1 eV. The IRPs decrease as a function of energy below the absorption edge. A shoulder appears at the point at which $f^0(|\mathbf{h}|)+f'(\omega)=0$ is satisfied. The IRP is quite small above the edge due to absorption. The dashed line is the IRP calculated with the damping factor of 3 eV, which shows no significant change compared with the solid line around the absorption edge.

The slowly varying modulation of the calculated IRP about 5 eV below the edge is the *Pendellösung* fringe induced by X-ray resonant scattering (Yoshizawa, Fukamachi, Ehara, Kawamura & Hayakawa, 1988; Fukamachi, Yoshizawa, Ehara, Kawamura & Nakajima, 1990).

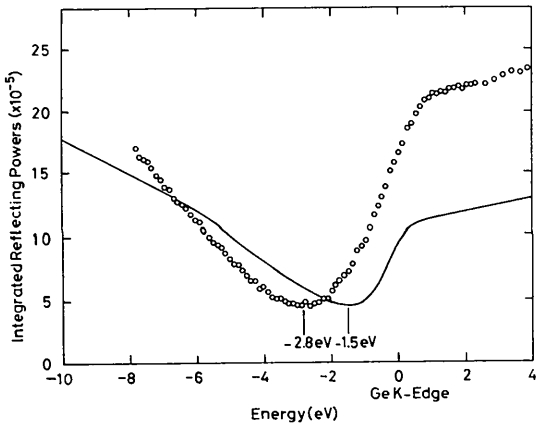

Fig. 2. The calculated IRPs in atomic units (solid line) and the measured IRIs in arbitrary units (open circles) of the Ge 844 reflection near the K-absorption edge in the symmetric Bragg case. The measured IRIs are scaled so that the minimum intensity is the same as the calculated one.

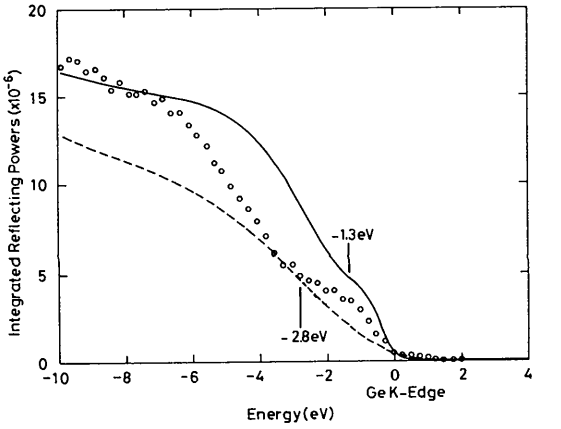

Fig. 3. The calculated IRPs in atomic units with damping factors of 1 eV (solid line) and 3 eV (dashed line) and the measured IRIs in arbitrary units (open circles) of the Ge 844 reflection near the K-absorption edge in the symmetric Laue case. The measured IRIs are scaled so that the intensity at the shoulder is the same as the calculated one.

3. Experiment and discussion

We measured the IRI with the experimental apparatus described by Fukamachi *et al.* (1990). The Ge crystal used was of high perfection with an EPD of 500 cm^{-2}. For the Bragg-case measurement, a (211) face was used. For the Laue-case measurement, a (100) parallel-sided plate was used and the crystal thickness was 39.3 (4) μm.

The experimental results of IRI for the Bragg case and the Laue case are shown in Figs. 2 and 3, respectively, by the open circles. The measured absorption edge is determined so that the intensity drop of IRI in Fig. 3 coincides with that of the calculated curve (the solid line). The energy resolution of the optical system, $\Delta\omega$, is determined by the relation

$$\Delta\omega = |\omega \cot\theta \, \Delta\theta|, \quad (3)$$

where θ is the Bragg angle and $\Delta\theta$ was 0.008° in the optical system used (Fukamachi *et al.*, 1990). The resolution $\Delta\omega$ of the experiment was ±0.2 eV.

In Fig. 2, the agreement between the theoretical curve and the experiment is generally good. The minimum intensity in the experiment is, however, 2.8 (4) eV below the edge, which is 1.3 eV lower than the theoretical value. In the Laue case (Fig. 3), the shoulder position in the experiment is also 2.80 (48) eV below the edge, which should correspond to the shoulder at the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ in the calculation.

Since the measured IRIs show a typical feature corresponding to the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ at 2.8 eV below the edge for both the Bragg and the Laue cases, we regard the IRIs at this energy as those induced by the imaginary part of the scattering factor only. The energy value of 2.8 eV is slightly larger than the value obtained by the above calculation (~1.3–1.5 eV).

The energy difference of the condition should come from the effect of XANES. The energy shift due to the change of damping factor (~0.5–2.0 eV) is quite small, of the order of 0.4 eV, which cannot explain the result. For a semiconductor, the value of $f''(\omega)$ just above the absorption edge is generally larger than the theoretical value given by the Parratt & Hempstead (1954) method. This results in a larger value of $|f'(\omega)|$ than the theoretical value through the dispersion relation between $f'(\omega)$ and $f''(\omega)$ (Kawamura & Fukamachi, 1978). Then the energy at the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ shifts to the lower side from the absorption edge (Fukamachi, Hosoya, Kawamura & Okunuki, 1977, 1979).

4. Summary

We have measured the IRIs from a Ge crystal across the *K*-absorption edge and found the IRIs due only to the imaginary part of the scattering factor $f''(\omega)$ when $f^0(|\mathbf{h}|)+f'(\omega)=0$. We have confirmed that the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ is satisfied at 2.8 eV below the *K*-absorption edge for the 844 reflection of Ge within the experimental error of ±0.24 eV. It is noted that at this condition the dynamical diffraction is caused only by the imaginary part of the scattering factor and the real part is zero. There remain some discrepancies between the theory and the experiment, which should be attributed to the value of the anomalous scattering factor used in the calculation.

We could not identify the energy position above the Ge *K*-absorption edge at which the condition $f^0(|\mathbf{h}|)+f'(\omega)=0$ was satisfied. In the Laue case, the reflected intensities were too weak to measure the variation. In the Bragg case, no significant intensity variation was observed, which was consistent with theoretical calculation. To clarify the energy position, it would be useful to measure the rocking curves rather than the IRIs (Fukamachi & Kawamura, 1993).

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