

when all three cross vectors are in the set of measured reflexions, and

$$G = \frac{2N^{-1}R_h R_k R_l R_{h+k+l} (R_{h+k}^2 + R_{h+l}^2 - 1)}{1 + \tanh [(R_{h+k}^2 + R_{h+l}^2)/2]} \quad (9)$$

when only two cross-vectors, $h+k$ and $h+l$, are present. In particular, the trends of the average error and of the average magnitude of the errors seem to be similar in triplets and quartets when (8) and (9) are used. The simultaneous use of triplet and quartet relationship in tangent procedures is thus justified.

A further remark may be useful. The empirical scaling factor

$$SC = 1 + \tanh \left(\sum R_j^2/j \right), \quad j=2,3,$$

proposed in this paper is of course not unique. It occurred to the author both by analogy with the

scaling factor successfully used for centrosymmetrical quartets (Giacovazzo, 1976a) and by its functional simplicity. SC nevertheless involves the magnitudes of the cross vectors alone, whereas the theoretical conditional variance given by the distribution function

$$P(\Phi_{h,k,l} | R_h, R_k, R_l, R_{h+k+l}, R_{h+k}, R_{h+l}, R_{k+l})$$

suggests a scaling factor which takes all the magnitudes into account. It is hoped that further work in this direction will improve present results.

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X-ray Intensity Measurements on Large Crystals by Energy-Dispersive Diffractometry. I. Energy Dependences of Diffraction Intensities near the Absorption Edge

BY T. FUKAMACHI, S. HOSOYA AND M. OKUNUKI*

Institute for Solid-State Physics, University of Tokyo, 22-1 Roppongi 7-chome, Minato-ku, Tokyo, Japan

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The intensity variations of X-rays diffracted from a nearly perfect GaAs plate have been measured in symmetrical Laue and Bragg cases in the energy region near the As *K* absorption edge with small energy intervals, by the use of an energy-dispersive diffractometer and continuous X-rays from a sealed-off tube. The corresponding intensity variations have been calculated with the dynamical theory. These measurements and calculations have shown a good agreement. Moreover, the curve measured for the Bragg case on the same crystal, but after polishing, has shown good agreement with the corresponding curve calculated by the kinematical theory. However, there is a minor discrepancy in the energy region very near the absorption edge. This is probably due to the fact that the values of anomalous-scattering factors used for calculation are not precise enough to explain fine structures at the edge.

Introduction

Energy-dispersive diffractometry, with a solid-state detector (SSD) and continuous radiation from a normal X-ray tube, has various merits of its own, complementary to those of traditional angle-dispersive diffractometry. One of the merits is, as has been well known since early work (Giessen & Gordon, 1968), the possibility of carrying out rapid measurements comparatively easily even under extreme conditions. According to recent papers, the interplanar distances (Fukamachi, Hosoya & Terasaki, 1973), and the intensity values both for single crystals (Buras, Olsen, Gerward, Selsmark & Andersen, 1975) and for powder samples (Uno & Ishigaki, 1975) have been measured with considerable accuracy under normal

conditions and even in extreme conditions (Inoue, 1975). Another merit is the possibility of carrying out the measurements with radiations of desired energy values. This has already been utilized for a rapid determination of polarity sense (Hosoya & Fukamachi, 1973), and for experimental determination of anomalous scattering factors at the energy values near the absorption edge (Fukamachi & Hosoya, 1975). Including the latter, various possibilities of determining the anomalous scattering factors and phases of reflexions have preliminarily been reviewed (Hosoya, 1975). In these papers, however, full formulation of the expressions for diffraction intensities and other quantities was not required. In the present work, such a formulation has been described in order to explain the measured energy dependences of intensities near the absorption edge in typical Laue and Bragg cases for mosaic and perfect crystal plates; for a mosaic

* Permanent address: JEOL Ltd.

crystal, the kinematical theory, and for a perfect crystal, the dynamical theory have been used for calculations with absorption taken into account. The observed energy dependences due to anomalous scattering have been explained by the relevant calculations.

Notation

For later use, the necessary notation is summarized here. Most of symbols and formulations follow Miyake (1969) but, for simplicity, the atomic unit $m = \hbar = e = 1$ is used.

The symbols to be used are as follows:

$F_{\mathbf{h}}$: the crystal structure factor for the \mathbf{h} reflexion,

$$F_{\mathbf{h}} = F_{\mathbf{hr}} + iF_{\mathbf{hi}} = \sum_j (f_j^0 + f_j' + if_j'') \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_j), \quad (1)$$

f^0 : atomic scattering factor without any anomalous scattering part included,

f_j', f_j'' : the real and the imaginary parts of the anomalous scattering factor,

$\varphi_{\mathbf{h}}$: the electron polarizability per unit volume ($\times 4\pi$); this is complex and we have

$$\varphi_{\mathbf{h}} = \varphi_{\mathbf{hr}} + i\varphi_{\mathbf{hi}}, \quad (2)$$

$$\begin{aligned} \varphi_{\mathbf{hr}} &= -(4\pi/v\omega^2) \sum_j (f_j^0 + f_j') \\ &\times \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_j) = |\varphi_{\mathbf{hr}}| \exp(i\alpha_{\mathbf{hr}}), \quad (3) \end{aligned}$$

$$\begin{aligned} \varphi_{\mathbf{hi}} &= -(4\pi/v\omega^2) \sum_j f_j'' \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_j) = |\varphi_{\mathbf{hi}}| \exp(i\alpha_{\mathbf{hi}}), \\ &\quad (4) \end{aligned}$$

ω : energy of an X-ray photon,

v : the volume of a unit cell,

$$\delta = \alpha_{\mathbf{hi}} - \alpha_{\mathbf{hr}}. \quad (5)$$

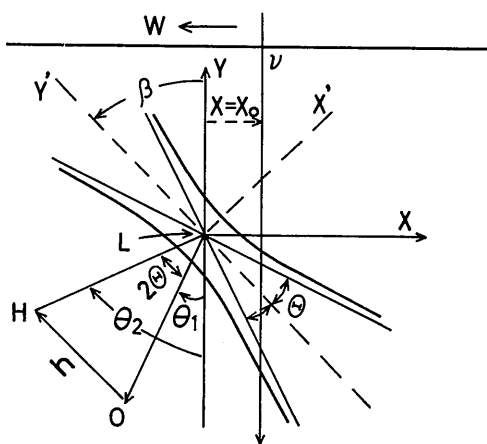


Fig. 1. Dispersion surfaces: O is the origin of the reciprocal lattice, H , the reciprocal-lattice point, L , the Laue point and ν , the normal to the crystal surface.

As shown in Fig. 1 by a pair of dispersion surfaces,

$$W = -X_0 \frac{\sin 2\Theta}{|\cos \theta_1 \cos \theta_2|^{1/2}} \frac{1}{\kappa_{0r} C |\varphi_{\mathbf{hr}}|}, \quad (6)$$

where X_0 , θ_1 , θ_2 and Θ are given in Fig. 1, Θ is equal to the usual Bragg angle θ_B if refraction is neglected, and $C=1$ for normal and $=|\cos 2\theta_B|$ for parallel polarization,

κ_{0r} : the wave vector inside the crystal;

$$g = \varphi_{0i}/(C|\varphi_{\mathbf{hr}}|), \quad (7)$$

where φ_{0i} is $\varphi_{\mathbf{hi}}$ with $\mathbf{h}=0$, and

$$g' = g \sin \Theta \cos \beta / |\cos \theta_1 \cos \theta_2|^{1/2}, \quad (8)$$

where β is given in Fig. 1.

$$|k| = |\varphi_{\mathbf{hi}}|/|\varphi_{\mathbf{hr}}|, \quad (9)$$

$$|\varphi_{\mathbf{hr}}/\varphi_{\pm\mathbf{h}}|^2 = 1/(1 + |k|^2 \mp 2|k| \sin \delta), \quad (10)$$

and $\mu(\omega)$ is the linear absorption coefficient which is equal to $-\omega/c \varphi_{0i}(\omega)$ at energy ω .

Formulation

The integrated intensity from a large fixed crystal obtained by energy-dispersive diffractometry is described.

(1) Ideally mosaic crystal

Let us assume that the incident continuous rays are smooth in intensity distribution and can be expressed by $I_0(\omega_B)d\omega$. Then the corresponding diffraction intensity $dI_{\mathbf{h}}$ is given as

$$dI_{\mathbf{h}} = (r_e/R)^2 p(\theta_B) \cdot G \cdot |F_{\mathbf{h}}|^2 I_0(\omega_B) d\omega, \quad (11)$$

where R is the distance between the crystal and the SSD, r_e the radius of a classical electron, $1/c^2$, c being the velocity of light, $p(\theta_B)$ the polarization factor, and $G (=G_1 G_2 G_3)$ the Laue function. The integrated intensity $J_{\mathbf{h}}$ is given by

$$J_{\mathbf{h}} = r_e^2 p(\theta_B) \int \int I_0(\omega_B) |F_{\mathbf{h}}|^2 \cdot G \cdot d\omega d\Omega, \quad (12)$$

where (see Fig. 2)

$$d\omega d\Omega/(2\pi c) = d\kappa d\Omega = dA_1 dA_2 dA_3 / (2v\kappa_B^2 \sin^2 \theta_B), \quad (13)$$

θ_B in $p(\theta_B)$ is approximated to be constant over the small irradiated area and $dA_1 dA_2 dA_3/v$ is an element of reciprocal volume. As is usually done, assuming $|F_{\mathbf{h}}|$ and $I_0(\omega_B)$ to be constant during the integration in (12), we get

$$\begin{aligned} J_{\mathbf{h}} &= 2\pi c r_e^2 p(\theta_B) |F_{\mathbf{h}}|^2 I_0(\omega_B) (2v\kappa_B^2 \sin^2 \theta_B)^{-1} \\ &\times \int \int G_1 G_2 G_3 dA_1 dA_2 dA_3 \\ &= 2\pi c r_e^2 p(\theta_B) |F_{\mathbf{h}}|^2 I_0(\omega_B) N_1 N_2 N_3 / (2v\kappa_B^2 \sin^2 \theta_B). \end{aligned} \quad (14)$$

Now, $N_1N_2N_3$ is the number of unit cells in the volume δV of the small crystal, so that $N_1N_2N_3 = \delta V/v$. Equation (12) then becomes

$$J_h = I_0(\omega_B) Q_h \delta V, \quad (15)$$

where

$$Q_h = (2\pi c)^3 r_e^2 p(\theta_B) |F_h|^2 / (2v^2 \omega_B \sin^2 \theta_B). \quad (16)$$

The integrated reflecting power R_h in the symmetrical Bragg case is expressed by

$$R_h = \frac{Q_h}{2\mu(\omega_B)} \quad (17)$$

for a plate with infinite thickness, and in the symmetrical Laue case is expressed by

$$R_h = \frac{Q_h H}{\cos \theta_B} \exp[-\mu(\omega_B) H / \cos \theta_B] \quad (18)$$

for a plate crystal with thickness H .

(2) Perfect crystal (under the two-wave approximation)

Integrated intensity J is given in general by

$$J = \int P_h(W, C) I_0(\omega_B) d\omega, \quad (19)$$

where the function of W , $P_h(W, C) = I_h(W, C)/I_0$, varies with the crystal forms and diffraction conditions. In the case of symmetrical reflexion, the following relation holds between $d\omega$ and dW , like the reduced form of (6):

$$d\omega = \frac{\omega_B C |\varphi_{hr}|}{2 \sin^2 \theta_B} dW. \quad (20)$$

Therefore,

$$J = \frac{\omega_B C |\varphi_{hr}|}{2 \sin^2 \theta_B} I_0(\omega_B) \int P_h(W, C) dW \quad (21)$$

holds, and the integrated reflecting power is expressed as

$$R_h = \frac{\omega_B C |\varphi_{hr}|}{2 \sin^2 \theta_B} \int P_h(W, C) dW. \quad (22)$$

Later, R_h will be obtained by using the function $P_h(W, C)$ relevant to the experimental conditions and then by carrying out the integration. When the incident X-rays are not polarized, R_h can be obtained as

$$R_h = \frac{1}{2}(R_h^{\perp} + R_h^{\parallel}), \quad (23)$$

a simple average of two terms R_h^{\perp} and R_h^{\parallel} with $C=1$ and $C=|\cos 2\theta_B|$ respectively.

The white radiation from an X-ray tube is considered to be more or less polarized. However, as seen later, the formulae with uniform polarization explain the present measurements without showing any appreciable deviation. This implies that the radiation is not strongly polarized.

The way of calculating anomalous scattering factors of Ga and As which is used in the present work is similar to that shown by broken lines for Ga in Fig. 5 of Fukamachi & Hosoya (1975).

Apparatus

The SSD diffractometer used has already been described (Fukamachi *et al.*, 1973; Fukamachi & Hosoya, 1975); the diffraction tube was rotated while the SSD with a heavy cryostat was fixed. In the present work, a fine-focus Philips tube with Cr target was operated.

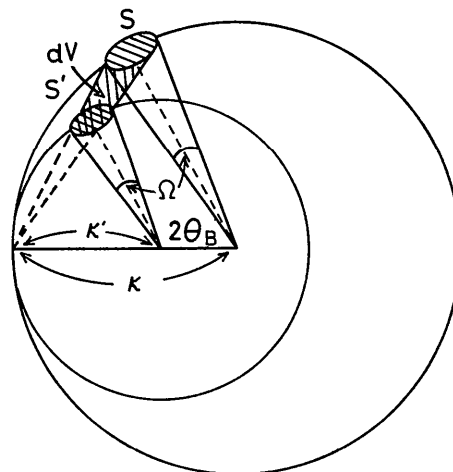


Fig. 2. The volume element over which the reflexion intensity is integrated in the Laue method: dV is the volume element in reciprocal space, $d\Omega$, the solid angle, S and S' , areas cut out from the sphere of reflexion by the solid angle $d\Omega$. The following relations hold: $S = \kappa^2 d\Omega$, $SS' = 2d\kappa \sin \theta_B$, and $dV = 2\kappa^2 \sin^2 \theta_B d\Omega d\kappa = dA_1 dA_2 dA_3/v$.

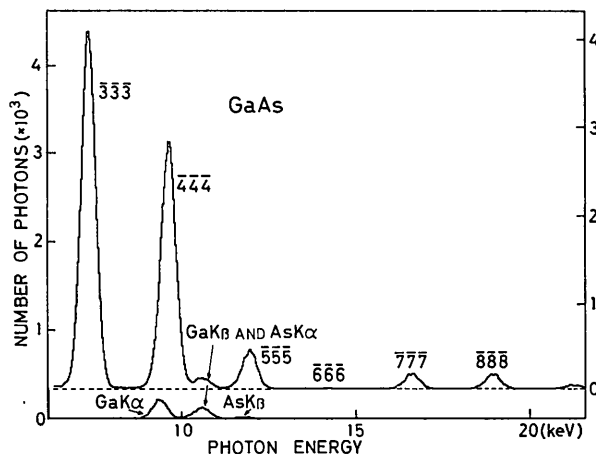


Fig. 3. The fluorescence X-ray peaks (lower curve) can be subtracted from the diffraction pattern (upper curve). $2\theta_B = 106^\circ$.

at 30 kV and 40 mA. In order to get high energy resolution, two pairs of long collimators were used (Fukamachi & Hosoya, 1975). They are 30 cm long with a hole either 0.4 mm or 1 mm in diameter. Later, another pair of collimators was used which have interchangeable holes different in size and shape.

Ge(Li) and Si(Li) detectors were used for the measurements. The software was prepared for controlling the system through a small computer: one of the programs was written in assembler language so that each proce-

dures could be fulfilled through conversational software by the use of various subroutines. The program on a higher level has also been prepared so that Fortran can be used.

As has already been described (Hosoya & Fukamachi, 1973), fluorescence X-rays from Ga and As were measured by rotating the specimen very slightly to an off-Bragg position. This spectrum was then subtracted from the measured hhh spectra, both being measured for the same length of time. Fig. 3 shows these spectra, and Fig. 4 the spectra with fluorescence peaks subtracted.

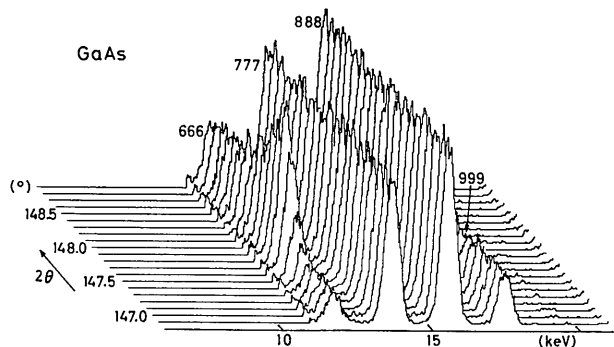


Fig. 4. Intensity change of the 666 reflexion from GaAs in the energy region near the As K absorption edge. $K\alpha_1$: Ga 9.251 keV, As 10.543 keV; $K\beta_1$: 10.263 keV, 11.725 keV.

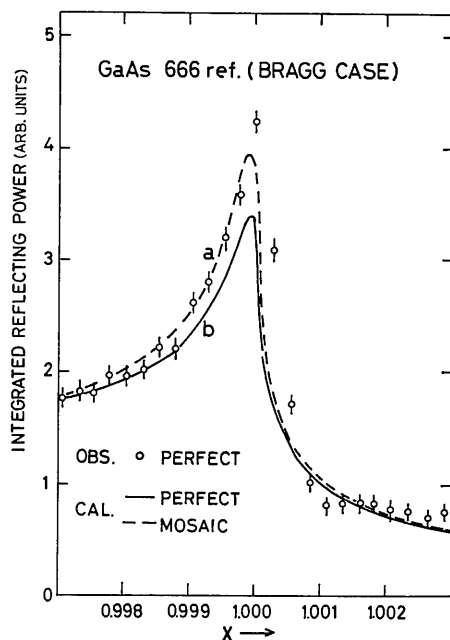


Fig. 5. Comparison between measured and calculated values of the 666 reflexion from GaAs in the energy region near the As K edge. The measured values shown by open circles are scaled so that the best fit may be obtained. Curves a and b are calculated for mosaic and perfect-crystal models respectively. The abscissa in Figs. 5, 6 and 7 is $X = \omega/\omega_K$, where ω_K is the energy of the As K absorption edge. The energy resolution is about ± 2 eV.

Intensity variation in the region near the absorption edge

The work by Cole & Stemple (1962) was more extensively repeated both theoretically and experimentally. The experiments were much improved by the present technique of using continuous radiation.

In the present calculations of structure factors, the origin is taken so that the Ga atoms are at $(0, 0, 0) + \text{f.c.c.}$ positions and the As atoms at $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4}) + \text{f.c.c.}$ positions based thereon.

(1) The intensity of the 666 reflexion from GaAs (Bragg case)

As shown in Fig. 3, the 666 reflexion is very weak, because $F_{666} = 4(f_{\text{Ga}} - f_{\text{As}})$. However, in the region near the absorption edge of As, this reflexion is not weak but nearly comparable to the 777 reflexion as shown in Fig. 4. As has already been pointed out (Cole & Stemple, 1962), the intensity of a weak reflexion does not vary very much between mosaic and perfect-crystal models in the energy region where the absorption is heavy. In Fig. 5, the measured values are compared with the calculated curves for the symmetrical Bragg case. The curve a has been calculated with (17) for the mosaic model, and the curve b for the perfect-crystal model. For the latter, the integration in (22) has been carried out numerically using

$$P_h(W, C) = \frac{1 + |k|^2 - 2|k| \sin \delta}{\{(1 - |k|^2)^2 + 4|k|^2 \cos^2 \delta\}^{1/2}} [\mathcal{L}(W) + \{\mathcal{L}^2(W) - 1\}^{1/2}]^{-1}, \quad (24)$$

where

$$\mathcal{L}(W) = \frac{\sqrt{(W^2 - 1 - g^2 + |k|^2)^2 + 4(gW - |k| \cos \delta)^2 + W^2 + g^2}}{\{(1 - |k|^2)^2 + 4|k|^2 \cos^2 \delta\}^{1/2}}. \quad (25)$$

These two formulae are generally expressed in terms of g' (Miyake, 1969). However, g' was replaced by g in (25), because $g = g'$ in the present symmetrical Bragg case. Moreover, for reflexions such as 666, for which Friedel's law holds, $\sin \delta = 0$ and $\cos^2 \delta = 1$ and then both (24) and (25) have much simpler forms.

The sample giving the data in Fig. 5 is free from dislocations as far as could be determined with the etch pit technique. Besides, the angular width of the 333 reflexion observed by the double-crystal method with Cu $K\beta$ is almost $6''$, which is comparable to the intrinsic value, $4.6''$. It will clearly be shown in part II that the measured data agree with the calculated curve much better for a perfect crystal than for a mosaic one. The curves *a* and *b* in Fig. 5 are more or less similar, as was described by Cole & Stemple (1962), but the data as a whole seem to agree with the curve *a* somewhat better than with *b*. More precisely, the discrepancy between the measured data and those curves is still not negligible. This minor discrepancy may come from the fact that the calculations for the anomalous scattering factors were carried out for the model not corresponding well to the real crystal; in fact, such fine structure of the absorption curve near the edge, which will be published later, was not taken into account.

It can at least be said that the weak reflexion can be dealt with in terms of a mosaic model without much error. On this ground, the anomalous scattering can be used without serious difficulty when the present method is applied, for instance, to the quantitative measurement of weak superstructure lines of partially ordered structures, in order to distinguish between atoms close together in the periodic table, such as Cu and Zn, as has been carried out previously (Jones & Sykes, 1937).

(2) Influence of the crystalline state of the surface (Bragg case)

In order that the effect of imperfection in a GaAs crystal could be investigated, a 110 plate was cut out and the surface polished with corundum powder of #2000; the measured data then changed as shown in Fig. 6. The data were explained by the calculations of (17) for the mosaic model. After the etching, the sample showed a very different curve, like that calculated with (24) for a perfect crystal. This fact has already been mentioned by Cole & Stemple (1962) and demonstrated preliminarily by Matsui & Takasu (1972), who used the conventional apparatus. The present results show that this type of study can be carried out far more easily and precisely with an SSD diffractometer.

(3) Intensity variation of ± 333 reflexions in the Laue case in the region near the As K edge

Cole & Stemple (1962) did not study the Laue case at all, while Datsenko, Skorokhod & Vasilkovski (1968) applied anomalous scattering to the study of the crystal imperfection inside crystals. In the present work, the intensity measurement was carried out in the symmetrical Laue case for the above-mentioned nearly perfect crystal. The results are shown for 333 and $\bar{3}\bar{3}\bar{3}$ reflexions in Fig. 7. The relevant calculations of (22) have been carried out by the use of the following integrand expressed by Miyake (1969) as

$$P_h(W, C) = I_h/I_0 = \exp(-\mu H/\cos \theta_B) \times (1 + |k|^2 - 2|k| \sin \delta) \times [\sin^2 \{s(C)H \operatorname{Re} \sqrt{L(W)}\} + \sinh^2 \{s(C)H \operatorname{Im} \sqrt{L(W)}\}] / |\sqrt{L(W)}|^2, \quad (26)$$

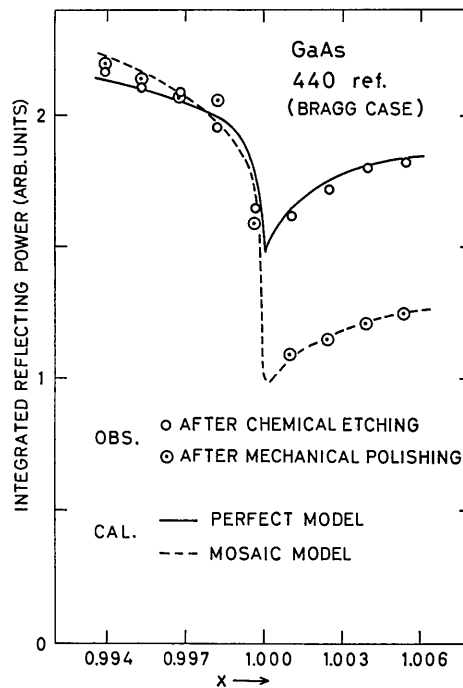


Fig. 6. Intensity change of the 440 reflexion from GaAs near the As K edge.

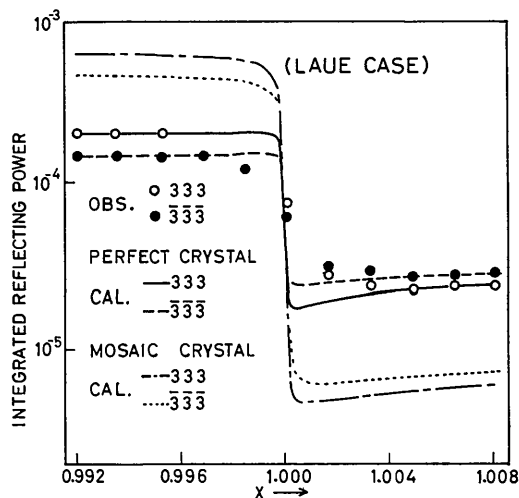


Fig. 7. Intensity change of 333 and $\bar{3}\bar{3}\bar{3}$ reflexions from GaAs in the Laue case in the region near the As K edge. The experimental energy resolution is about ± 12 eV. The curves for a mosaic crystal have been calculated with (18), while those for a perfect crystal with (22) and (26).

where

$$s(C) = (\pi k_{or} C |\varphi_{hr}|) / \cos \theta_B \quad (27)$$

and

$$|\sqrt{L(W)}|^2 = \{(W^2 + 1 - |k|^2)^2 + 4|k|^2 \cos^2 \delta\}^{1/2}. \quad (28)$$

The expression $\sqrt{L(w)}$ is written more explicitly as follows:

$$\begin{aligned} \text{Re } \sqrt{L(W)} &= 2^{-1/2} \{(W^2 + 1 - |k|^2) \\ &+ \sqrt{(W^2 + 1 - |k|^2)^2 + 4|k|^2 \cos^2 \delta}\}^{1/2} \quad (29) \end{aligned}$$

and

$$\begin{aligned} \text{Im } \sqrt{L(W)} &= \pm 2^{-1/2} \{- (W^2 + 1 - |k|^2) \\ &+ \sqrt{(W^2 + 1 - |k|^2)^2 + 4|k|^2 \cos^2 \delta}\}^{1/2}, \quad (30) \end{aligned}$$

where the sign is + if $|k| \cos \delta > 0$ and - if $|k| \cos \delta < 0$.

In this case, the measured intensity changes agree well with the curves calculated for a perfect-crystal model. It is interesting to note that the calculated integrated reflexion power is larger for a perfect crystal than for a mosaic model in the region $x > 1$, being due to the anomalous-transmission effect. The difference in the intensity change near the absorption edge between a perfect model and a mosaic model is bigger in the Laue case than in the Bragg case. It is, therefore, favourable for the study of dislocation density to use the present SSD diffractometry instead of the conventional method (Datsenko *et al.*, 1968). The dependence of intensity change upon the polarity will be extensively described in part II.

Conclusion

In the present paper, the integrated reflecting power has been calculated theoretically for diffraction of continuous X-rays, and this calculation has been compared with the results of measurements. Previous results [for example of Cole & Stemple (1962)] obtained with the usual diffractometry with a counter were not precise enough to be qualitatively compared with the theoretical calculations. In this sense, the present method has proved to be so useful that the results may even be compared with the theoretical ones in the energy region near the absorption edge. The following features are to be noted:

(1) The general features of intensity variation of integrated reflecting power show a good agreement between the experimental values measured for different energy values and the theoretical values calculated for the corresponding diffraction conditions and crystal models.

(2) The agreement between theory and experiment is a little worse in the energy region very near the absorption edge than elsewhere. This is considered to be due to the fact that the anomalous scattering factors have not yet been calculated directly with the effect of the fine energy structure characteristic of each atom in the relevant compound taken into account, but simply calculated as mentioned in the last part of the section on formulation. The disagreement as a result of this factor has already been found in previous work concerning the anomalous scattering factor curves of Ga in GaP.

(3) Because of the situation described in (2), it may be stated that the dynamical theory is also valid in this interesting energy region around the absorption edge.

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