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X-ray Intensity Measurements on Large Crystals by Energy-Dispersive Diffractometry. IV. Determination of Anomalous Scattering Factors near the Absorption Edges of GaAs by the One-Intensity-Ratio Method

BY T. FUKAMACHI,* S. HOSOYA, T. KAWAMURA† AND M. OKUNUKI‡

Institute for Solid-State Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo, Japan

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Abstract

The imaginary part $f''(\omega)$ of each scattering factor for Ga and As has been measured through the absorption coefficient on a nearly perfect GaAs crystal plate in the energy regions near the K absorption edges of both Ga and As atoms. The intensity ratio r_{555} between 555 Friedel-pair reflexions has been measured in the same energy regions. From these data, the real part $f'(\omega)$ has been determined in the same way as has already been published [Fukamachi & Hosoya (1975). *Acta Cryst.* A**31**, 215–220]. The $f'(\omega)$ values above the edges thus obtained show fairly good agreement in fine structure with those calculated from $f''(\omega)$ values with the dispersion relation, but some discrepancies are found in the regions below the edges.

Introduction

In the papers of the present series (I: Fukamachi, Hosoya & Okunuki, 1976*a*; II: Fukamachi, Hosoya & Okunuki, 1976*b*; III: Fukamachi, Hosoya, Kawamura & Okunuki, 1977), the energy-dispersive intensity measurement method has been developed with a solid-state detector (SSD) diffractometer and in particular a study has been made on the energy dependence of diffraction intensity including the anomalous scattering factors (III, 1977). Measurements were carried out on the integrated reflexion powers R_h from a perfect GaAs crystal in the Laue and Bragg cases. The results could be interpreted by the dynamical theory of X-ray diffraction including the absorption effect (Miyake, 1969). The agreement between the measurements and the calculations was fairly good in energy regions not very near the edges. The following relation

$$r_h = R_h/R_{\bar{h}} = |F_h|^2/|F_{\bar{h}}|^2 \quad (1)$$

was also confirmed to hold experimentally and theoretically for both perfect and mosaic polar crystals in the symmetrical Laue and Bragg cases (II, 1976*b*). Equation (1) is very useful for determining the anomalous scattering factors (Hosoya, 1975; Fukamachi & Hosoya, 1975) using a crystal with a known simple polar structure without parameters.

In the present paper, we report on the determined f' values of the atoms in a perfect GaAs crystal in the energy regions near Ga and As K edges by the measurement of the r_h values. This was carried out earlier by Fukamachi & Hosoya (1975) for GaP, but the energy resolution is improved in the present case. The results are compared with other f' values such as those calculated by Hönl's theory (Hönl, 1933*a*,*b*) and by the dispersion relation (III, 1977). The discrepancies in f' values obtained by these methods are also discussed.

* Present address: Saitama Institute of Technology.
† Permanent address: Faculty of Education and Liberal Arts, Yamanashi University.
‡ Permanent address: JEOL Ltd.

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Measurements

The integrated powers of 555 and $\bar{5}\bar{5}\bar{5}$ reflexions in the symmetrical Bragg case have been measured on a GaAs single-crystal plate in the energy regions near the Ga and As K absorption edges. The specimen crystal is the same one used in the previous papers (I–III): it is free from dislocation as far as can be observed by the etch-pit technique. The energy resolution is 0·9 eV around the Ga K edge and 1·5 eV around the As K edge. The X-ray tube was operated at 30 kV and 38 mA. The measurement was repeated at each energy value at least four times, where the counts were accumulated for 400 s each time. The $K\beta$ fluorescence X-rays from Ga and As were measured by rotating the specimen slightly to an off-Bragg position and by subtracting the obtained spectrum from the diffraction intensities, as was reported earlier (Hosoya & Fukamachi, 1973). The energy dependence of each integrated reflexion power R_h is shown by open circles in Figs. 4 and 5 in Part III for regions near the Ga and As K edges, respectively. From these measured values, the intensity ratio was determined in the region near Ga and As K edges as shown in Figs. 1 and 2, respectively.

Determination of $f'(\omega)$

By the same method as described previously (Hosoya, 1975; Fukamachi & Hosoya, 1975), the values of $f'(\omega)$ have been determined. In GaAs, any effect due to the temperature factors does not appreciably affect the results because the temperature parameters are almost the same for Ga and As (Uno, Okano & Yukino, 1970). In order to determine f'_{Ga} values near the Ga K edge, for example, it is necessary to know other values, f^0_{Ga}, f^0_{As}, f'_{As}, f''_{Ga} and f''_{As}. The normal scattering factors f^0_{Ga} and f^0_{As} were taken from the table calculated by Fukamachi (1971) using Clementi's (1965) wave functions. The values of f'_{As} and f''_{As} depend upon energy, but they were approximated to be constant because they change very slowly in the relevant energy region far from the As edge. These values are listed in Table 1. The value of $f''_{\mathrm{Ga}}(\omega)$ was also taken from Part III (1977).

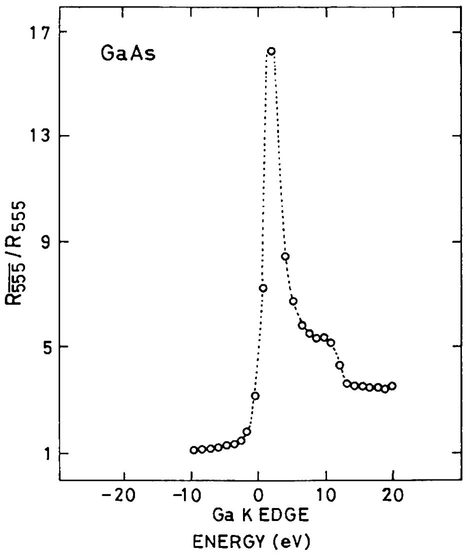

Fig. 1. Intensity ratios measured on Friedel-pair reflexions from GaAs in the energy region near the Ga K absorption edge.

On the other hand, f'_{As} values can be determined near the As K absorption edge, in a similar way as in the above, with the values of f''_{As}, f'_{Ga} and f''_{Ga} as parameters. However, in Fig. 2, there is an energy value for which R_{555} is equal to $R_{\bar{5}\bar{5}\bar{5}}$. At this point, the above method is not applicable. Instead, using relation (4) in Part II, the intensity ratio is expressed as

$$r_h = \frac{R_h}{R_{\bar{h}}} = \frac{1 + |k|^2 - 2|k| \sin \delta}{1 + |k|^2 + 2|k| \sin \delta} = \frac{|F_h|^2}{|F_{\bar{h}}|^2}, \qquad (2)$$

where

$$|k| = |\varphi_{hi}|/|\varphi_{hr}|, \qquad (3)$$

as given by (9) of Part I. Because $k \neq 0$, the condition $r_h = 1$ leads to

$$\sin \delta = 0, \qquad (4)$$

from which the relation

$$f''_{\mathrm{Ga}}/f''_{\mathrm{As}}(\omega) = (f^0_{\mathrm{Ga}} + f'_{\mathrm{Ga}})/\{f^0_{\mathrm{As}} + f'_{\mathrm{As}}(\omega)\} \qquad (5)$$

Table 1. *Scattering factor values*

The normal structure factor for the 555 reflexion	f^0_{Ga}	10·00	f^0_{As}	11·21
The anomalous scattering factor of As atoms at the Ga K edge			f'_{As} f''_{As}	−2·19 0·78
The anomalous scattering factor of Ga atoms at the As K edge	f'_{Ga} f''_{Ga}	−1·25 3·32		

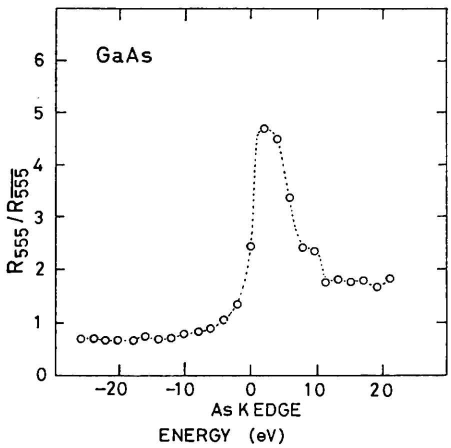

Fig. 2. Intensity ratios measured on Friedel-pair reflexions from GaAs near the As K edge.

is derived at a point near the As K edge. From (5), the value of $f'_{As}(\omega)$ can be determined as indicated by a solid circle in Fig. 4.

Results and discussion

The present results obtained using the values of one-intensity-ratio (IR) are shown in Figs. 3 and 4 for the energy regions near the Ga and As K edges. In these figures, the calculated values of f' by Hönl's theory (HT) and those by the dispersion relation (DR) discussed in Part III are shown for comparison.

Above the edges, the values obtained by IR are more or less in good agreement with those by DR. As for $f'_{Ga}(\omega)$ in Fig. 3, the shoulders denoted by α and β appear in both cases (IR and DR); $f'_{As}(\omega)$ in Fig. 4 has only one shoulder, α. On the other hand, below the Ga K edge, the value by IR is not in good agreement with the value either by DR or by HT. The reason for this may be due to the fact that f''_{Ga} is very small and therefore the intensity ratio r_h is almost equal to unity, which makes it difficult to determine $f'_{Ga}(\omega)$ precisely by the present method.

Moreover, as is shown in Fig. 1, the value of $(r_{555})^{-1}$ changes very rapidly in the region lower than the Ga K edge. If there is any slight misalignment, the error of the measured ratio can become really large. In addition, the error of $f''(\omega)$ due to the lack of energy resolution becomes large. These two reasons may well cause a relatively large discrepancy between the measured values and the calculated ones in the region with energy slightly lower than the Ga K edge.

Below the As K edge, however, f''_{Ga} is still large enough to give a large intensity ratio. This makes it easier to determine f'_{As} than f'_{Ga} in the Ga K edge. Accordingly, the value obtained by IR is about 5% smaller, in absolute terms, than the one calculated by DR, but agrees with the one by HT fairly well. Recently, Bonse & Materlik (1976) measured $f'(\omega)$ values of Ni by an optical method with an X-ray interferometer and a synchrotron radiation source. Their results show a good agreement with the values by HT. In view of this fact, the method of DR used in the above includes the experimental errors in the absolute values of $f''(\omega)$ and this should be improved.

Thus these two methods are complementary in various respects: the interferometer method can be applied to an elemental sample but is difficult to use with an ordinary X-ray source, while the present method needs a polar crystal of a suitable compound but can be used with an ordinary X-ray source and a SSD.

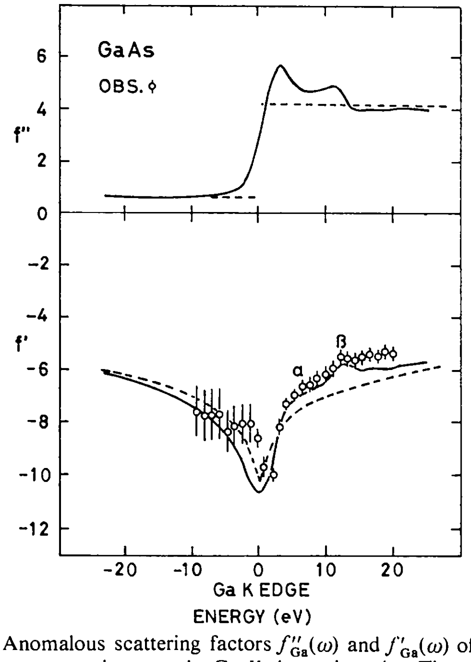

Fig. 3. Anomalous scattering factors $f''_{Ga}(\omega)$ and $f'_{Ga}(\omega)$ of GaAs in the energy region near the Ga K absorption edge. The solid line for $f''(\omega)$ was obtained from the measured values of the absorption coefficient $\mu(\omega)$; the broken lines for $f'(\omega)$ and $f''(\omega)$ are from Hönl's (1933*a*,*b*) theory; the solid line for $f'(\omega)$ was calculated from the $f''(\omega)$ curve by the dispersion relation. The error bars on $f'_{Ga}(\omega)$ values are due to the statistical fluctuation of the counts.

Conclusion

The subjects of the present series are the studies on the energy-dispersive integrated reflexion powers and on the energy dependence of the anomalous scattering

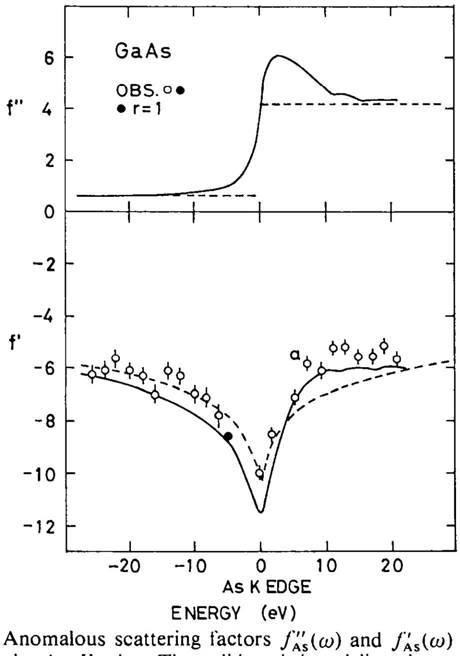

Fig. 4. Anomalous scattering factors $f''_{As}(\omega)$ and $f'_{As}(\omega)$ of GaAs near the As K edge. The solid and dotted lines have the same meaning as in Fig. 3.

factors near the K absorption edges in GaAs. In these papers, we applied the dynamical theory of diffraction to an absorbing crystal. The results have shown a relatively good agreement between theoretical calculations and the experimental results in the integrated reflexion intensity curve.

The fine structures of the anomalous scattering factor, namely shoulders of $f'_{\rm Ga}(\omega)$, $f''_{\rm Ga}(\omega)$, $f'_{\rm As}(\omega)$ and $f''_{\rm As}(\omega)$ curves above respective edges are different from each other. This seems to suggest that the structures of anomalous scattering factors should be attributed to the wave functions of Ga and As, or in other words, the matrix element of the photoabsorption does not simply depend upon the density of final states in GaAs, as was discussed for GaP (Fukamachi, Shimamoto, Ohtsuki & Hosoya, 1975). It is, therefore, necessary to find better wave functions in order to discuss the anomalous scattering factors more precisely.

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Temperature Dependence of X-ray Reflection Intensity from an Absorbing Perfect Crystal near an Absorption Edge

BY TAKAAKI KAWAMURA

Physics Institute, Faculty of Education and Liberal Arts, Yamanashi University, Kofu, Yamanashi 400, *Japan*

AND TOMOE FUKAMACHI

Department of Electrical Engineering, Saitama Institute of Technology, Okabe, Saitama 369-02, *Japan*

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Abstract

The temperature effect of X-ray integrated reflection intensities is discussed theoretically in the energy region near the absorption edge of Ga in GaAs perfect crystal. From the calculation based on the dynamical theory of diffraction, it is concluded that the integrated intensity of $\bar{5}\bar{5}\bar{5}$ in the Bragg case shows a complicated temperature effect depending on the change of the anomalous scattering factor across the absorption edge. Calculated results are compared with a preliminary measurement, which shows a qualitative agreement with the theory.

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Introduction

The integrated reflection intensities of X-rays (IRIX) from a crystal involve the effect of anomalous scattering in the energy region near the absorption edge of the specimen atom. Cole & Stemple (1962) first pointed out that the IRIX show different variations across the absorption edge depending on perfection and polarity of the crystal. They pointed out, in particular, that the intensity ratio of hkl to its polar opposite $\bar{h}\bar{k}\bar{l}$ for a polar crystal is independent of crystal perfection. In this connection, they measured the IRIX of 111 and $\bar{1}\bar{1}\bar{1}$ for perfect and mosaic crystals of GaAs across Ga