Broadening of the x-ray emission line due to the instrumental function of the double-crystal spectrometer

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The influence of the instrumental function on the Cu $K\alpha_1$ emission line was investigated for the case of a double-crystal spectrometer. The magnitude of broadening for both Si(220) and Si(440) was calculated for a Lorentzian emission line with the width of 1–5 eV; the broadening for Si(220) is 0.12–0.18 eV while that for Si(440) is only 0.015–0.043 eV. The former is too large to be neglected, so the correction for the instrumental function is important. The spectrum affected by the instrumental function seems to keep the shape of Lorentzian though its width is larger. The fact indicates that the Lorentzian fitting analysis is effective if the appropriate correction for width is done.

I. INTRODUCTION

The double-crystal spectrometer is one of the most valuable tools for the measurement of the x-ray emission spectrum, and many investigations have been performed with this type of spectrometer so far. In spite of the frequent use of the double-crystal spectrometer, the influence of its instrumental function still remains uncertain. The contribution of the instrumental function of the double-crystal spectrometer is usually considered to be so small that it is often neglected. But the measurement with different crystal planes gives different observed widths as we shall show later. This fact indicates that the proper correction is necessary when we discuss the linewidth with precision of a few tenth eV or more. In 1954, with confirmation in his subsequent work in 1963, Brogren derived one formula for estimating the true linewidth from his measurements [1,2]. His formula was as follows:

$$W_T = W_0 - W_C. \tag{1}$$

W_T, W_0, and W_C mean the true width of the line, the width of the observed emission line, and the width of the rocking curve of the employed crystal in $(+ -)$ position, respectively. All widths referred to half-maximum intensity. His formula seems valid because the true width of a certain line derived from his measurements utilizing Eq. (1) is consistent when the plane for diffraction changes. But the difference between the measured width and the true width is in fact, as we shall show later in the present work, usually smaller than the width of the rocking curve of the crystal employed. As for the general formulation for the instrumental function of the double-crystal spectrometer, it was precisely described by Azaroff in 1974 referring even to the tilt of the crystal and the size of the detector [3].

In the present work we have simulated the influence of the instrumental function of the double-crystal spectrometer on the emission spectrum to estimate to what extent the instrumental function practically affects the spectrum. The magnitude of the influence seems to greatly depend on both the wavelength investigated and the plane of the crystal employed. The simulation is applied to the case for radiation having the energy of Cu $K\alpha_1$ (8048.1 eV) observed with Si(220) and Si(440).

II. CALCULATION

A. Geometry

The geometry of the system for the double-crystal spectrometer is shown in Fig. 1. In the following, the tilts of the two crystals were not taken into consideration because they are unknown so they cannot be taken into calculation. The radiation emitted from the sample goes into the first crystal after it passes through the Soller slit, which restricts the vertical divergence. The glancing angle of this radiation for the first crystal, denoted by η_1, is given by

$$\eta_1 = \frac{\pi}{2} - \arccos(\cos\varphi\,\sin\theta) \equiv \eta(\theta,\varphi). \tag{2}$$

Here η is a function that calculates the glancing angle from two arguments. The first argument θ means the angle between the projection of the radiation on the horizontal plane [(x,y) plane] and the crystal plane for reflection. The second argument φ is the angle between the radiation and the horizontal plane. When the positions of the two crystals are for the Bragg angle of β, the glancing angle for the second crystal is expressed as follows:

$$\eta_2 = \eta(2\beta - \theta,\varphi) = \frac{\pi}{2} - \arccos[\cos\varphi\,\sin(2\beta - \theta)]. \tag{3}$$

Assuming that the radiation emitted from the sample is isotropic, as is the case for K emission spectrum induced by the

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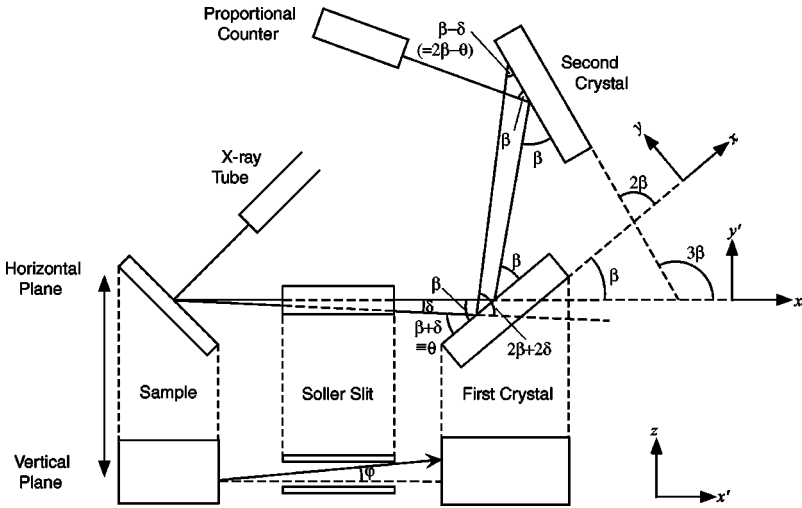*Author to whom correspondence should be addressed.

FIG. 1. Schematic diagram for double-crystal spectrometer.

secondary excitation, the intensity of the radiation depends only on energy. In this paper, the distribution of the intensity on the energy of radiation is denoted by $F(E)$. The slit function described in detail later is denoted by $S(\varphi)$. If the window of the detector is so large that all the reflected beam from the second crystal could be caught, then $I(\beta)$, the intensity with the crystal positions at the Bragg angle β, is

$$I(\beta) = \int_0^\infty \int_{-\varphi_0}^{\varphi_0} \int_0^{\pi/2} F(E)S(\varphi)R(E,\eta_1)$$

$$\times R(E,\eta_2)dE\,d\varphi\,d\theta, \tag{4}$$

where $R(E,\eta)$ is the rocking curve for the radiation having the energy of E and φ_0 is the maximum value of φ, which is limited by the Soller slit. What we observe in our measurement is $I(\beta)$ at each point of β. The spectrometer we used is equipped with a goniometer that makes it possible to change β by steps of 0.01°, 0.005°, 0.002°, 0.001°, and 0.0005°. In the rest of this section, the slit function and rocking curve used in the calculation are briefly presented.

1. Slit function

The Soller slit is used to restrict the vertical divergence in the spectrometer. The interval of each layer, denoted by w, is 1 mm and the length of this Soller slit, denoted by ℓ, is 100 mm (see Fig. 2). Each layer corresponds to a pair of slits that are separated by a distance ℓ and whose widths are both w. In that case, under the condition that w/ℓ is so small that we can approximate $\tan w/\ell$ to w/ℓ, the slit function $S(\varphi)$ is given by

$$S(\varphi) = \begin{cases} 1 - \dfrac{\ell}{w}|\varphi| & \left(|\varphi| \leq \dfrac{w}{\ell} \equiv \varphi_0\right) \\ 0 & (|\varphi| > \varphi_0). \end{cases} \tag{5}$$

The Soller slit used gives φ_0 of 2062.6″, which raises the divergence of the glancing angle by 4.52″ and 13.88″ for Si(220) and Si(440), respectively, at energy of Cu $K\alpha_1$ (8048.1 eV). Thus the vertical divergence leads to fairly small divergence of the glancing angle while the horizontal divergence causes the divergence of the glancing angle almost of its magnitude. The strong point of the double-crystal spectrometer lies in the fact that the horizontal divergence, which dominantly affects glancing angle, diminishes due to the second crystal. The vertical divergence cannot be restricted by the second crystal because the sum of η_1 and η_2 is not 2β when φ is not zero.

2. Rocking curve

We calculated the rocking curve of Si(220) and Si(440) at the energy of Cu $K\alpha_1$ (8048.1 eV) on the basis of dynamical theory with the effect of absorption described by Zachariasen [4]. The effect of absorption appears in the rocking curve as the asymmetric shape. Reflectivity depends on the polarization of incident beam and rocking curves are usually calculated for two specific cases, that is, normal polarization and parallel polarization. In the simulation, where it is assumed that the incident beam is unpolarized, the average of the rocking curve for normal polarization and that for parallel polarization was adopted as $R(E,\eta)$ in Eq. (4). This is based on the assumption that there is no relation in phase between the reflection on the first crystal and that on the second crystal. Besides, the rocking curve depends on the energy of the
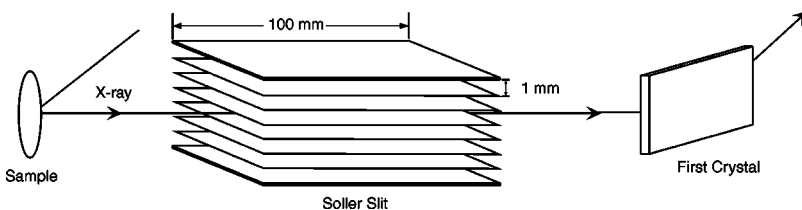

FIG. 2. Soller slit that we employed for our measurement ($\ell = 100$ mm, $w = 1$ mm).
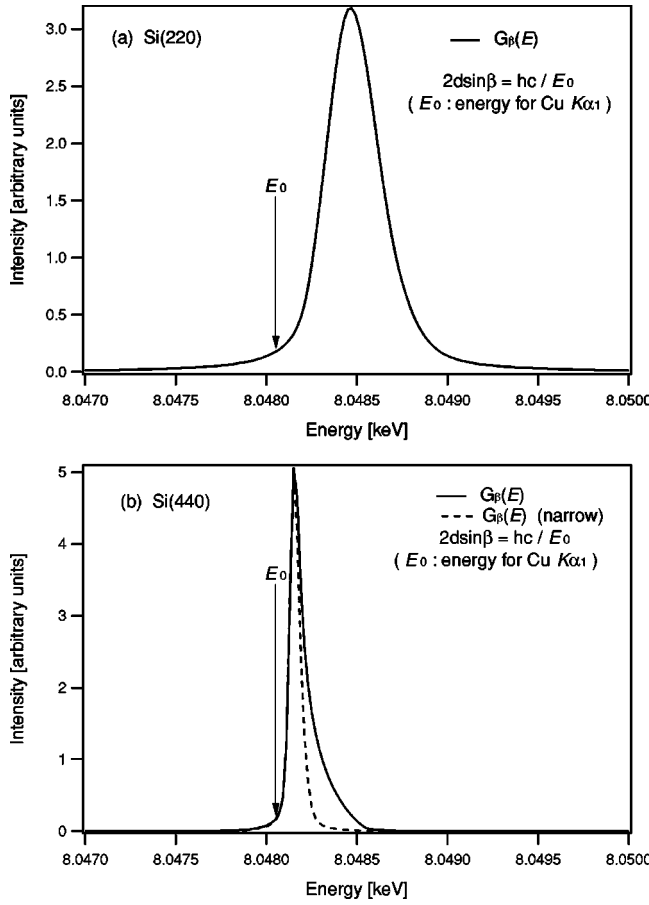
FIG. 3. (a) $G_\beta(E)$ for Si(220). (b) $G_\beta(E)$ for Si(440). [The solid curve shows $G_\beta(E)$ with the slit we employed, and the dashed curve shows that with the narrower slit ($\ell = 100$ mm, $w = 0.5$ mm).]

radiation. But the change of the rocking curve in the region for energy in the calculation [$(E_0 - 12.5)$ eV to $(E_0 + 12.5)$ eV; E_0 means the energy investigated] is so small that we are allowed to use a common rocking curve (the rocking curve at E_0 is likely the best) at any energy. We calculated the rocking curve at the step of 1 eV in the region of $(E_0 - 25)$ eV to $(E_0 + 25)$ eV. In the integration with respect to φ and θ, the obtained rocking curve for the energy closest to E was used with the shift of η by $(\theta_{B_m} - \theta_B)$ in order to improve the precision of the calculation. Here m is an integer between -25 and 25. θ_{B_m} and θ_B correspond to the Bragg angle for the energy of E_m [$E_m = (E_0 + m)$ eV] and for the energy investigated, respectively. For example, $R(E_m, \eta + \theta_{B_m} - \theta_B)$ was employed to give the value of $R(E, \eta)$.

B. Computation

In the procedure of calculating $I(\beta)$, the integration with respect to φ and θ was done to get $G_\beta(E)$ for every position, β of two crystals, which is defined as
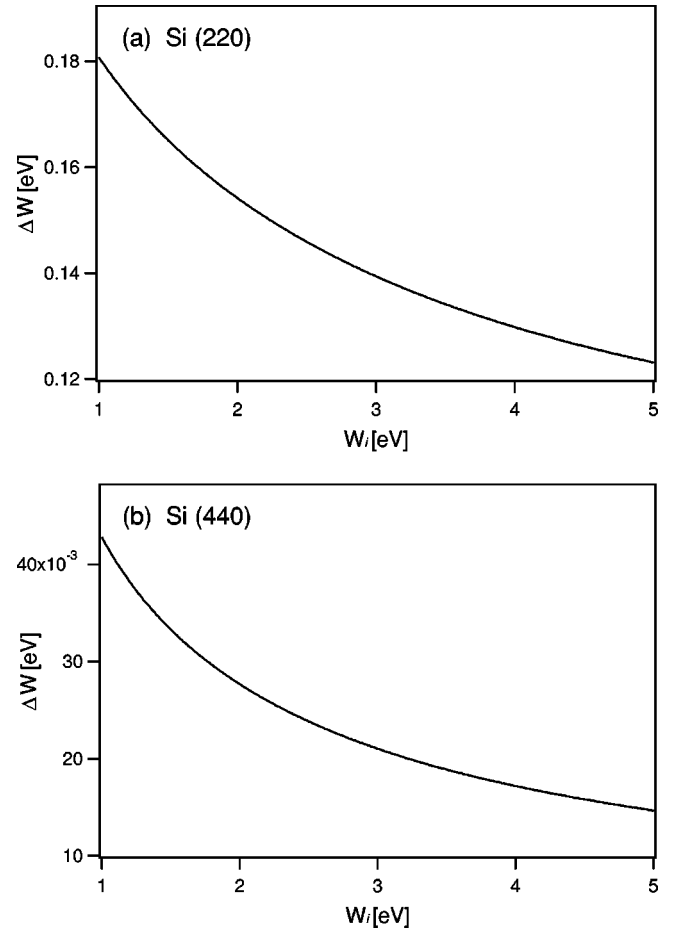

FIG. 4. Dependence of ΔW on W_i for (a) Si(220) and (b) Si(440).

$$G_\beta(E) = \int_{-\varphi_0}^{\varphi_0} \int_0^{\pi/2} S(\varphi) R(E, \eta_1) R(E, \eta_2) d\varphi \, d\theta, \quad (6)$$

where $G_\beta(E)$ corresponds to the window function on energy E of the spectrometer. To shorten the computation, the range of θ was reduced to such an extent that almost all the points at which the reflectivity on the first crystal [$R(E, \eta_1)$] is less than 10^{-3} were excluded. $G_\beta(E)$ for β satisfying the Bragg condition for the energy of Cu $K\alpha_1$ is shown in Figs. 3(a) and 3(b). It is easily seen that $G_\beta(E)$ for Si(440) is rather asymmetric and has its tail on the high-energy side. $G_\beta(E)$ with a narrower vertical Soller slit ($\ell = 100$ mm, $w = 0.5$ mm) shows that this is due to the comparatively wider dispersion on the lower side of glancing angle η that makes the contribution from the radiation having higher energy larger [see Fig. 3(b)]. In the present work, the step angle of β in the simulation for Si(220) was $4''$ while that for Si(440) was $8''$.

$I_\beta(E)$ was obtained by integrating $F(E)G_\beta(E)$ with respect to E for each β. $F(E)$ was defined only for the energy between $(E_0 - 12.5)$ eV and $(E_0 + 12.5)$ eV and the integration of $F(E)G_\beta(E)$ with respect to the energy were only performed on this range. We substituted Lorentzian [denoted by $L_i(E)$] for $F(E)$ in Eq. (4). The width of $L_i(E)$ was
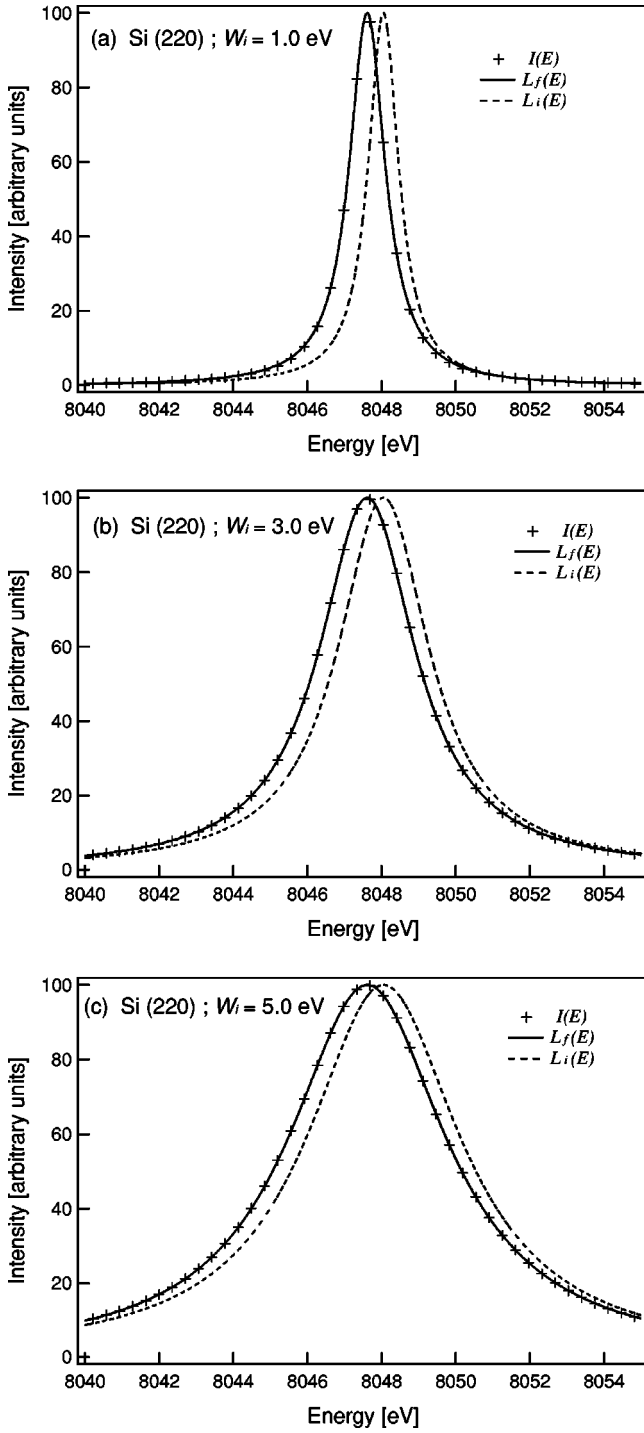
FIG. 5. Change in profile of original Lorentzian with widths of (a) 1.0 eV, (b) 3.0 eV, (c) 5.0 eV for Si(220).

FIG. 6. Change in profile of original Lorentzian with widths of (a) 1.0 eV, (b) 3.0 eV, (c) 5.0 eV for Si(440).

varied in the range of $1-5$ eV. In addition, to see how much the shape of $I(E)$ is deviated from Lorentzian, we fitted a Lorentzian [denoted by $L_f(E)$] to $I(E)$, which is obtained from $I(\beta)$ by converting β into the energy that satisfies the Bragg condition, $2d \sin \beta = hc/E$. We examined the broadening [by comparing the width of $L_i(E)$ and that of $L_f(E)$] and how much each point of $I(E)$ is deviated from $L_f(E)$. It should be noted that several points, which are close to the edges, were eliminated in the fit procedure because the
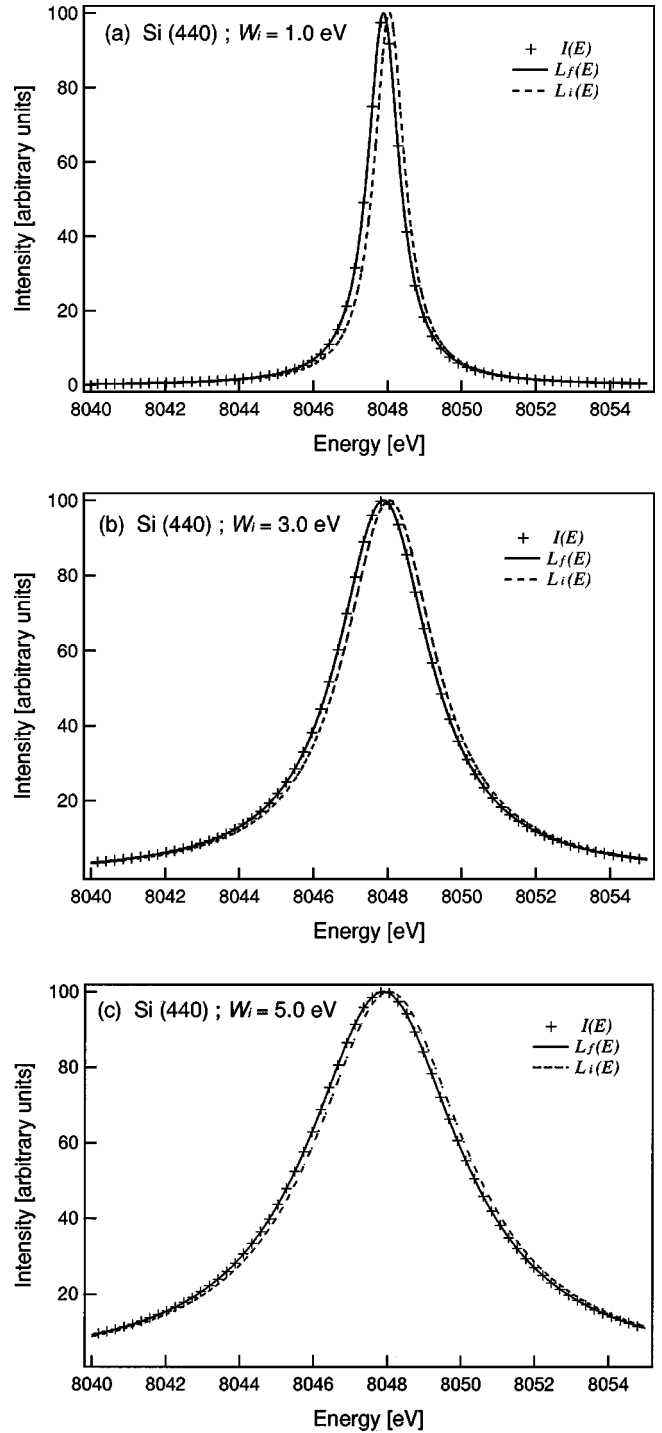
points near the edges are subjected to the influence of missing the curve in the further region.

III. RESULTS AND DISCUSSION

Figures 4(a) and 4(b) shows the broadening $\Delta W (\equiv W_f - W_i)$ determined by the calculation. It is easily seen that the larger width of $L_i(E)$ leads the smaller broadening both for Si(220) and for Si(440). In the region of $1-5$ eV for W_i, the
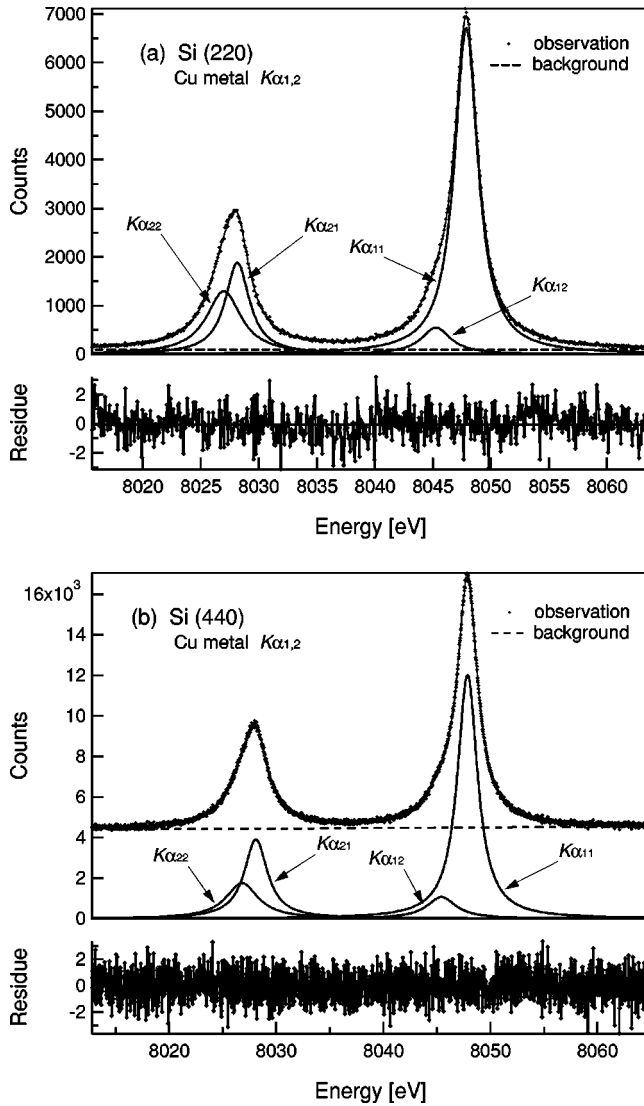
FIG. 7. Spectra of Cu $K\alpha$ measured using double-crystal spectrometer with double crystal of (a) Si(220) and (b) Si(440).

The deviation of $I(E)$ from $L_f(E)$ can be seen in Figs. 5(a)–5(c) for Si(220) and Figs. 6(a)–6(c) for Si(440), respectively. It appears that all the points of $I(E)$ are very close to $L_f(E)$ for all cases. This means that it may be allowed to fit Lorentzian to the spectrum observed.

For reference, the measured Cu $K\alpha$ spectra are shown in Fig. 7. Please note that the following discussion has no relation with various phenomena involved in the Cu $K\alpha$ spectra. Both spectra were measured using the same double-crystal spectrometer, but with Si(220) for one and Si(440) for the other. We fitted four Lorentzians to these spectra and compared the widths of the fitted Lorentzians. In the measurement, a Rh x-ray tube was used for exciting Cu K radiation. It was operated at 40 kV, 70 mA. The results for $K\alpha_{11}$, which are considered to be most reliable, were compared. As can be seen from Table I, the width of the fitted Lorentzian W_f for $K\alpha_{11}$ is 2.454 eV and the calculation gives 2.305 eV for W_i in the case of Si(220). Similarly, in the case of Si(440), the calculation gives 2.254 eV for W_i when W_f is 2.280 eV. For comparison, we tabulated, with ours, the true widths of Cu $K\alpha_{11}$ obtained by other researchers [5,6] using a double-crystal spectrometer (Table II). The deviation among them is less than 0.07 eV. Considering the fact that the difference between the widths deduced from the two independent measurements with the same reflection Si(333) is about 0.06 eV, the values tabulated seem close to one another.

One of the ways to evaluate the validity of our method is comparing our two corrected widths. These should be the same ideally. However, as can be seen from Table II, there still remains the difference of 0.05 eV. This difference might not be attributed only to the error in calculation but to that in measurement, which includes the aberrant alignment of the optical system. In either case, it is necessary to investigate a specific line using the same spectrometer with various conditions and see how the corrected widths agree with one another. It is desirable that the line to be investigated is intense and symmetric.

IV. CONCLUSION

We investigated the influence of the instrumental function of a double-crystal spectrometer by calculating the broadening of emission line having the energy corresponding to Cu $K\alpha_1$. The result was that the contribution of the instrumental function to the emission spectrum cannot be neglected and that it is possible to estimate its magnitude with calculation. Our results also show that the broadening of Cu $K\alpha_{11}$ for Si(220) and Si(440) are roughly 0.149 eV and 0.026 eV,

magnitude of broadening is in the range 0.12–0.18 eV and 0.015–0.043 eV for Si(220) and Si(440), respectively. On the other hand the width of the rocking curve (average of normal and parallel polarization) for Si(220) and Si(440) are 0.46 eV and 0.07 eV, respectively. This indicates that the magnitude of broadening is much smaller than the width of the rocking curve for both cases and the correction using Eq. (1) may underestimate the true width. The broadening for Si(440) is so small that the width without any correction is almost the same as the true width, though for Si(220) this is not the case.

TABLE I. Widths of four fitted Lorentzians for Cu $K\alpha_{1,2}$ observed with the crystal of Si(220) and Si(440). All values are in units of eV. The values in parentheses are errors in the Lorentzian fitting.

Crystal	$K\alpha_{11}$	$K\alpha_{12}$	$K\alpha_{21}$	$K\alpha_{22}$
Si(220)	2.454(0.013)	3.071(0.113)	2.602(0.091)	3.571(0.078)
Si(440)	2.280(0.008)	3.290(0.093)	2.612(0.061)	3.661(0.083)

TABLE II. True widths (corrected widths) of Cu $K\alpha_{11}$ obtained by using a double-crystal spectrometer. All values are in units of eV.

	Härtwig *et al.* [5]	Deutsch *et al.* [6]	Present work	
Reflection	Si(333)	Si(333)	Si(220)	Si(440)
Full width at half maximum	2.24	2.298	2.305	2.254

respectively. The former cannot be neglected for the observed linewidth and the correction for the broadening is important. As to the change in profile, it seems that the curves affected by the instrumental function are almost Lorentzian with wider width than that of the original. In this case, we can effectively use the Lorentzian fitting with the correction for the width. It is necessary to apply our calculation to various lines with various crystal planes.

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