

《Original》 The Measurement of the Mosaic Spread of the Single Crystal*

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Abstract

A method of finding the mosaic spread of a single crystal is proposed by working with secondary extinction. With the mosaic spread of a crystal obtained by this method, the mosaic spread of another crystal is measured using a double axis single crystal diffractometer by Caglioti's method. The mosaic spread of the same crystal is also measured with a powder diffractometer by Epstein's method for the sake of the comparison of two methods.

요 약

단결정의 mosaic spread를 측정하는 방법 한가지를 제안하였다. 이 방법을 사용해서 하나의 단결정체의 mosaic spread를 측정하고 이 값을 토대로 다른 단결정체의 mosaic spread를 양측 단결정회절 장치를 이용하여 Caglioti의 방법으로 측정하였다. 동일한 단결정체에 관한 실험을 분말회절 장치를 이용하는 Epstein의 방법을 사용하여 두가지 방법을 비교할 목적으로 재차 수행하였다.

1. Introduction

The monochromating crystal is the most important part of a neutron diffractometer. As is well known, the intensity and the resolution of the spectrum from the sample of a diffractometer depend on the reflectivity of the monochromating crystal, the collimation divergences, and the dispersion parameter. The reflectivity of the single crystal for the monochromator has been discussed in connection with the mosaic spread and the collimation or the secondary extinction¹⁻⁶⁾. The

relations among the resolution or the intensity of neutron diffractometer, the mosaic spreads of crystals involved and the angular divergences of the collimations were studied⁷⁻¹⁴⁾, using the method proposed by Sailor *et al*¹⁵⁾. It is essential to know the mosaic spread of the monochromator to be used, when a neutron diffractometer is planned to build. The various ways of measuring the mosaic spread of a single crystal can be found from the above mentioned works⁷⁻¹⁴⁾.

In all but Epstein's case¹³⁾ which will be

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discussed in Sec. 2.3 and Sec. 3.3, to measure the mosaic spread, a double axis single crystal diffractometer with a monochromator of known mosaic spread has to be used. Since we do not have any crystal with known mosaic spread, a method of finding the mosaic spread of a crystal with relatively small secondary extinction is proposed in Sec. 1. The mosaic spread of a NaCl (200) crystal is measured by this method. This crystal is put as the monochromator in a double axis single crystal diffractometer mechanism¹¹⁾ to measure the mosaic spread of a copper crystal(111) by Calioti's method⁹⁾. The measurement of the mosaic spread of the copper crystal is also carried out by recent Epstein's method¹³⁾.

2. Theoretical

About the mosaic spread of a single crystal, there have been some misunderstanding as pointed out by Hamasaki¹⁴⁾. The value of experimentally determined mosaic spread is the so called effective mosaic spread, β' , which is different from the real mosaic spread. β' is such a quantity that the crystal reflectivity is approximated to being proportional to $\exp(-\Delta^2/(\beta')^2)$. Here, Δ is the deviation of a mosaic block from the mean orientation of the crystal plane. But β' will be mainly treated since it is not difficult to convert β' to real mosaic spread.

The meaning of symbols appearing in this paper are as follows;

- α_1 : the horizontal angular divergence of the first collimator defined by the width of one of its channels divided by the length
- α_2 : the corresponding value for the second collimator to α_1 ,
- α_3 : the corresponding value for the third collimator to α_1 ,
- β_1' : the effective mosaic spread of the first crystal,
- β_2' : the corresponding value for the second

crystal to β_1' ,

a : the dispersion parameter.

(1) The Measurement of the Mosaic Spread from the Secondary Extinction Coefficient.

We can define the secondary extinction coefficient, E_s , as $I_o = E_s I_c$ where I_c and I_o are the calculated and observed diffraction intensity respectively from a crystal. As discussed by Hamilton^{16, 17)}, E_s can be approximated to $\exp(-KI_c)$ when $E_s > 0.7$.

$$\text{Here, } K = \frac{8\lambda^3 A^{\frac{1}{2}}}{(3\pi)^{3/2} V^2} \quad (1)$$

where

λ : the wavelength of neutrons,

A : the cross section of the specimen,

V : the volume of the unit cell.

It is proposed here to measure the β' of a crystal, whose structure is exactly known, from the obtained value of E_s when $E_s > 0.7$.

(2) Caglioti's Method

Caglioti *et al*⁹⁾ discussed the Full Width at Half Maximum (FWHM) of the intensity distribution from a single crystal sample in a diffractometer with three different Soller collimators. There are two ways of moving the second crystal and the detector, that is, one-to-two coupling and just rocking the crystal with fixed detector. Since there is not much difference between these two cases, let us take the case of rocking. The formula for FWHM of the diffraction curve for this case has been worked out as follows;

$$\begin{aligned} (\text{FWHM})^2 = & (\beta_2')^2 + \{ \alpha_2^2 \alpha_3^2 (\alpha_1^2 + 4(\beta_1')^2) \\ & - 2a\alpha_2^2 \alpha_3^2 (\alpha_1^2 + 2(\beta_1')^2) \\ & + a^2 [\alpha_3^2 (\alpha_1^2 \alpha_2^2 + \alpha_1^2 (\beta_1')^2) \\ & + \alpha_2^2 (\beta_1')^2 + \alpha_1^2 \alpha_2^2 (\beta_1')^2] \} / Z \quad (2) \end{aligned}$$

where

$$\begin{aligned} Z = & \alpha_3^2 (\alpha_1^2 + \alpha_2^2 + 4(\beta_1')^2) + \alpha_2^2 (\alpha_1^2 + 4(\beta_1')^2) \\ & - 4a\alpha_2^2 (\alpha_1^2 + 2(\beta_1')^2) + 4a^2 (\alpha_1^2 \alpha_2^2 \\ & + \alpha_1^2 (\beta_1')^2 + \alpha_2^2 (\beta_1')^2). \end{aligned}$$

(3) Epstein's Method

Epstein *et al*¹³⁾ devised the way of finding out the mosaic spread of a crystal without having any other crystal of known mosaic spread. They put the crystal in question as monochromator to assemble a powder diffractometer. The formula by Caglioti *et al*⁷⁾ for the FWHM of diffraction pattern from the sample in a powder diffractometer is as follows

$$\begin{aligned} \text{FWHM} = & [\alpha_1^2 \alpha_2^2 + \alpha_1^2 \alpha_3^2 + \alpha_2^2 \alpha_3^2 \\ & + 4(\beta_1')^2 (\alpha_2^2 + \alpha_3^2) - 4\alpha_2^2 \alpha_3^2 (\alpha_1')^2 \\ & + 2(\beta_1')^2] + 4\alpha^2 (\alpha_1^2 \alpha_2^2 + \alpha_1^2 (\beta_1')^2 \\ & + \alpha_2^2 (\beta_1')^2)]^{1/2} / (\alpha_1^2 + \alpha_2^2 + 4(\beta_1')^2)^{1/2} \quad (3) \end{aligned}$$

Since β_1' is not known it is used as a parameter to obtain the best fit of the equation (3) to the experimental data. The FWHM is nearly linear in the region, $1 < a < 2$. The slope of this line and the minimum value of FWHM can be used as criteria to the curve fitting.

3. Experimental and Result

The crystal in question was a disk with the radius of 3 inches and the thickness of 1/2 inch. The face of the crystal was approximately parallel to (111) plane. The experiment was carried out at one of the radial beam port of TRIGA Mark-II reactor with a double axis neutron diffractometer consisting of BNL-W-34 monochromating mechanism and US-1 diffractometer installed in 1965¹¹⁾. The reactor was operated at the power of 250 KW during the experiment. Soller collimators were made

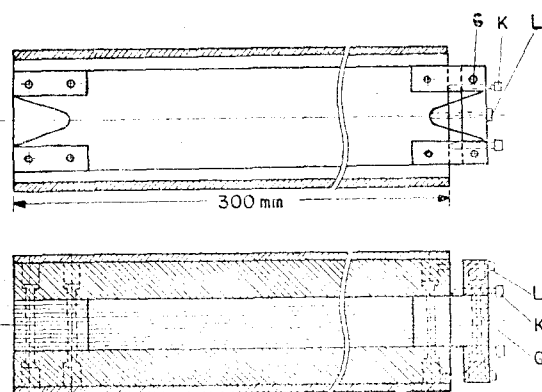


Fig. 1. Drawing of Soller collimator

of 0.1mm thick stainless steel plates using the technique of Bally *et al*¹⁸⁾. The thinnest spacers used are 1mm thick. Care was taken to keep parallel every plates together while these collimators were making. The drawing of the collimator is shown in Fig. 1.

(1) The Determination of β_1' and α_1

Following the method discussed in Sec. 2.1, the measurement to find the mosaic spread of the NaCl crystal (200) was carried out. The value of K in Eq. (1) was found to be 0.015cm² and that of β_1' turned out to be 2.9×10^{-4} . This value was very small as usual with ionic crystals. It is well known that the effective angular divergence of a collimator is slightly different from the value determined from its dimensions because of the total reflection and the transparency at the collimator wall. The effective divergence of collimators, α' s, were measured by the method of Sailor¹⁵⁾ from the rocking curve of our NaCl (200) crystal at the wavelength of 0.975 Å. The results and the calculated divergences for collimators used in the experiment are shown in table 1,

Table 1. Angular divergences of collimators

length (mm)	width (mm)	divergence from dimensions	effective divergence
300	1	0.0033	0.0035
300	2	0.0066	0.0069

(2) Caglioti's Method

A double axis single crystal diffractometer was assembled by putting the NaCl crystal, whose mosaic spread had been found to be 2.9×10^{-4} , as the monochromator and putting the crystal in question at the sample position. The values of α_1 , α_2 , and α_3 were 0.0035, 0.042, and 0.0069 respectively in the experiment. The rocking curve of the sample taken

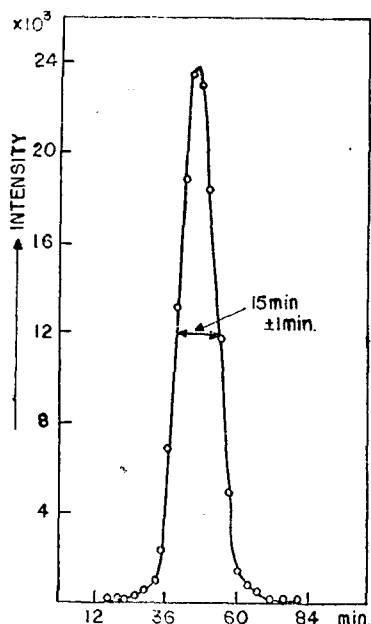


Fig. 2. Rocking curve of Cu (111) crystal with 0.975 Å neutrons

with Caglioti's condition using the monochromatic neutrons of wavelength of 0.975 Å from NaCl crystal is shown in Fig. 2. The value of β_2' , which was obtained from the FWHM of the rocking curve, was 0.0031 ± 0.0003 . This value was converted to be 0.0028 ± 0.003 using the method of Hamaski⁽⁴⁾.

(3) Epstein's Method

Two powder diffractometers were set up with different α_1 's of 0.0035 and 0.0069 using the same crystal of question as the monochromator in both cases. The values of α_2 and α_3 are the same with the case of Sec. 3.2 respectively. The experimental FWHM's of various reflections were taken from three polycrystalline samples (NaCl, CaF₂, Ni) at the wavelengths of 0.975 Å and 1.304 Å using above two diffractometers. The results are shown in Fig. 3 and Fig. 4 respectively with the dispersion parameters. The series of values of FWHM's with certain β_1' were calculated by Eq. (3) varying a with a IBM330 digital computer.

Each one of these series was fitted to the

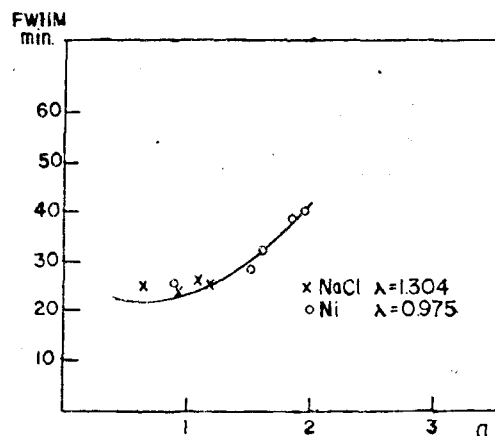


Fig. 3. Experimental FWHM's. $\alpha_1=0.0035$, $\alpha_2=0.042$, $\alpha_3=0.0067$. The continuous curve is one of theoretical curve.

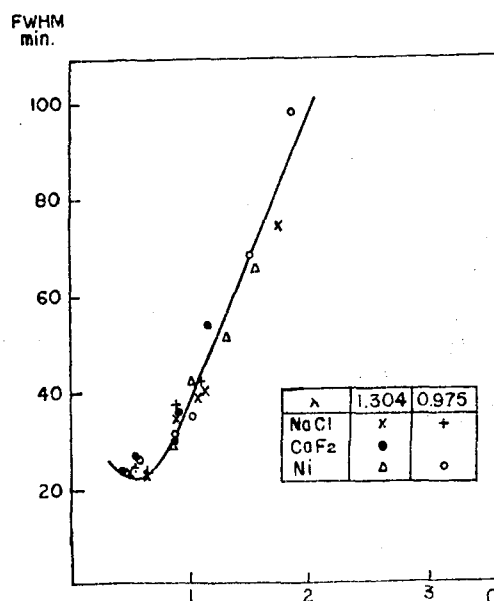


Fig. 4. Experimental FWHM's. $\alpha_1=0.0069$, $\alpha_2=0.042$, $\alpha_3=0.0067$. The continuous curve is one of theoretical curve.

experimental data of different conditions (Fig. 3, 4). The above series of values of FWHM's were plotted against a 's and the slopes were taken from these graphs in the range of $1 < a < 2$. These slopes vs. (β_1') 's are shown in Fig. 5 as the curve, slope. The FWHM's for each a , at the point where the experimental FWHM is minimum, have been calculated by

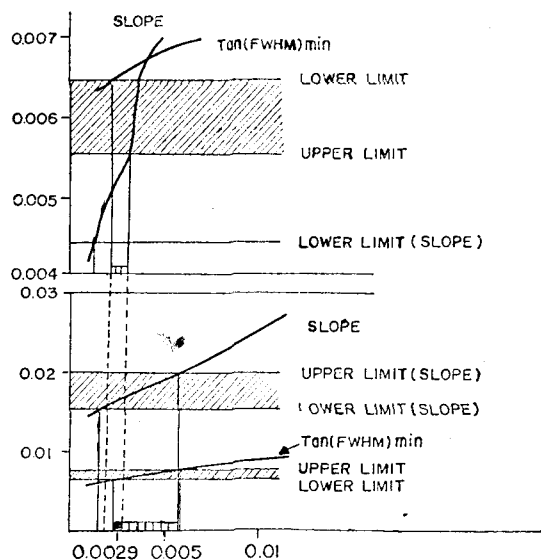


Fig. 5. Illustration of the method to find the values of

Eq. (4) varying β_1' 's and are also shown in Fig. 5 as the curve, $\tan(\text{FWHM}) \min$. Upper and lower limit of possible slope and minimum of the curve were evaluated from the experimental data. These possible regions defined a probable region for β_1' for every one of these graphs. The intersection of the four regions defined β_1' with dependable accuracy on the width of this intersection. The procedure is described in Fig. 5. The value of β_1' obtained is 0.0029 ± 0.001 and the real mosaic spread converted from this value becomes 0.0026 ± 0.001 which is equivalent to 10 ± 0.5 min. arc.

4. Conclusion

The errors in the measurement of β_2' by Caglioti's method on which our method is based, is $\Delta\beta_2' = \frac{\text{FWHM}}{\beta_2'} (\Delta\text{FWHM})$ as discussed by Rauch¹⁹⁾ if the error of β_1' is ignored. On the other hand, the degree of accuracy in the measurement of β_2' by Epstein's method can not be predicted theoretically because it is determined by the width of the

intersection of the four regions as mentioned in Sec. 3.3.

It can be imagined that the accuracy for Epstein's method is better than for our method though the accuracy of Epstein's probably. Even favorable, this method is more complicated to work with than our method.

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