STRUCTURE FACTOR DETERMINATION OF SINGLE CRYSTALS WITH HIGH EXTINCTION

By B. Buras*, T. Giebułtowicz, W. Minor, A. Rajca, I. Sosnowska, D. Śledziewska-Błocka and K. Wojtczak

Institute of Experimental Physics, Warsaw University**

(Received January 29, 1972)

A method of structure factor determination of single crystals with high secondary extinction is proposed. Spinning single crystal samples are used and absolute values of structure factors are obtained. Experimental results for bismuth, zinc and aluminium single crystals are presented and discussed.

1. Introduction

A method for determination of structure factors of single crystals with high secondary extinction is proposed and the preliminary rough experimental check of this method is described.

The method is based on the observed increase of intensity of neutrons reflected from a rotating single crystal when the speed of rotatio nincreases [1, 2]. Fig. 1 shows the general character of the dependence of the integrated intensity $E_{hkl}(\omega)$ on the angular velocity ω of the spinning single crystal. As it has been previously shown in some detail [2] the increase of intensity is a combined result of two effects. One is of a "geometrical" nature: while the neutron travels within the spinning single crystal, the angle of incidence related to a certain crystallographic plane (*hkl*) changes and thus the probability that the neutron once reflected will be once more reflected decreases with the speed of rotation. The second effect is due to the Doppler effect. While travelling within the rotating single crystal, the neutron passes crystal regions of different velocities. As the Bragg condition for the neutrons depends on the velocity of the crystal lattice, the Bragg angle is different in each of these regions. Thus both effects make even a perfect crystal look like a real one to the neutron. In the case of a real single crystal the (apparent) mosaic spread increases with the speed of rotation and thus an increase of reflectivity is observed.

* Present address: Department of Physics, H.C. Oersted Institute, University of Copenhagen, 2100 Copenhagen ∅, Denmark.

** Address: Instytut Fizyki Doświadczalnej, Uniwersytet Warszawski, Warszawa, Hoża 69, Poland.

By increasing the angular velocity of the crystal "saturation" of the measured integrated intensity may be obtained, as shown in Fig. 1, and the saturation value $E_{hkl}(\infty)$ can then be used as the extinction free integrated intensity. In other words the ratio $E_{hkl}(\infty)/E_{hkl}(0)$ gives the extinction factor y, as defined by Zachariasen [3]. If however, the saturation cannot be achieved for the available angular velocities then the fitting

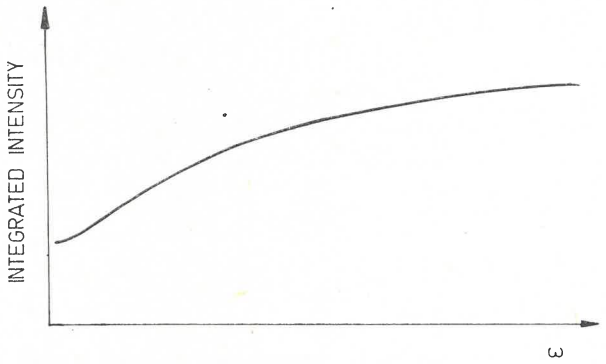

Fig. 1. General character of the dependence of the integrated intensity $E_{hkl}(\omega)$ on the angular velocity ω of the rotating single crystal

procedure, outlined below, can permit the determination of the absolute values of the structure factor.

The formula describing the curve shown in Fig. 1 has been derived in the Appendix of the paper [2] using the classical theory of secondary extinction [4]. In the case of a monochromatic incident neutron beam and nonabsorbing single crystal slab in symmetrical transmission geometry (Fig. 2) this formula has the following form:

$$E_{hkl}(\omega) = \frac{T\omega P_0(0)}{4\pi} \int_{-\infty}^{+\infty} \left[1 - \exp\left(-2Q \int_{-x_0}^{x_0} W\left(-\omega\tau - \frac{2\omega}{v_0} x + \frac{\omega}{v_0} x_0 \right) dx \right] d\tau \qquad (1)$$

where $P_0(0)$ — power of the incident beam, v_0 — neutron velocity, ω — angular velocity of the single crystal, $W(\Delta\theta) = \frac{1}{\eta\sqrt{2\pi}} \exp\left(-\frac{(\Delta\theta)^2}{2\eta^2} \right)$ — Gaussian distribution function for the mosaic blocks, $\Delta\theta$ — angular deviation of a mosaic block from the mean position, η — standard deviation of the mosaic blocks, x_0, $-x_0$ — x-coordinates of the slab surfaces (see Fig. 2), τ — time at which the neutron enters the crystal, $Q = \frac{\lambda^3 |F|^2}{V^2 \sin 2\theta_B}$ — crystallographic quantity, λ — neutron wavelength, θ_B — Bragg angle for the crystal at rest, V — unit cell volume, F — structure factor including the Debye-Waller factor, T — duration of the measurement.

In the case the time-of-flight (TOF) method we obtain a similar formula. In both cases the function $E_{hkl}(\omega)$ can be calculated by means of numerical methods. Only two

parameters of this function are unknown: the structure factor F and the standard deviation of the mosaic blocks η. By fitting the theoretical curve to the experimental points one can, at least in principle, find both the structure factor and the mosaic spread. In

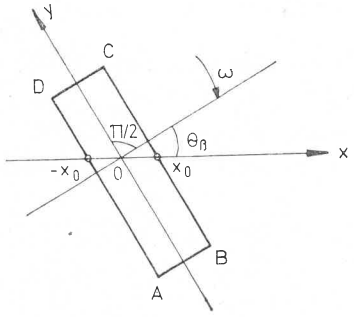

Fig. 2. Geometry of the experiment. The neutron beam is parallel to the x-axis. $ABCD$ — the cross-section of the single crystal slab spinning around the axis perpendicular to the plane of the figure

Section 2 we transform the function (1) into a form suitable for numerical calculations. Section 3 contains a brief description of the measurements and a discussion of the results.

2. *Formula for numerical calculations*

Equation (1) can be transformed by simple mathematics into the following one

$$E_{hkl}(\omega) = \frac{1}{2} A\omega \int_{-\infty}^{+\infty} \left[1 - \exp\left(-\frac{Qv_0}{\omega\sqrt{\pi}} \int_{z-\xi}^{z+\xi} e^{-k^2} dk\right)\right] d\tau \tag{2}$$

where

$$A = \frac{P_0(0)T}{2\pi}, \quad z = \frac{\omega}{\eta\sqrt{2}}\left(\tau - \frac{x_0}{v_0}\right) \tag{3}$$

$$\text{and } \xi = \frac{\sqrt{2}\, x_0 \omega}{v_0 \eta}.$$

In order to simplify the numerical calculations we approximate the internal integral in Eq. (2) by

$$\Phi(a) = \begin{cases} \dfrac{1}{\sqrt{\pi}} \displaystyle\int_0^a e^{-k^2} dk & \text{for } 0 \leqslant a < 5 \\ \dfrac{1}{2} & \text{for } a \geqslant 5 \end{cases}$$

and we obtain finally

$$E_{hkl}(\omega) = A\sqrt{2}\,\eta \int_0^{5+\xi} \left[1-\exp\left\{-\frac{Qv_0}{\omega}\times\right.\right. \tag{4}$$

$$\left.\left.\times\left(\Phi(z+\xi)-\operatorname{sign}(z-\xi)\Phi(|z-\xi|)\right)\right\}\right] dz.$$

The function $\Phi(a)$ is tabulated for values of $a = 0.002\,n$ ($n = 1, 2, 3 \ldots 2500$). For other values of a in the interval 0–5 the linear interpolation method was used.

The values of the parameters λ, v_0, V, θ_B and x_0 are known for given experimental conditions. The values of the parameters F and η were determined from the least squares fit of the theoretical values $E_{hkl}(\omega)$, described by Eq. (4), to the experimental values $E_{hkl}(\omega_i)$ for the angular velocities $\omega_i (i = 1, 2, 3, \ldots)$.

The procedure outlined above was performed with the help of the GIER computer.

3. Experiment and results

The majority of the measurements was performed on standard double axis spectrometers (DAS) located at the 8 MW reactor EWA at Świerk[1]. Fig. 3 illustrates the principle of the experimental set up. A collimated monochromatic neutron beam with

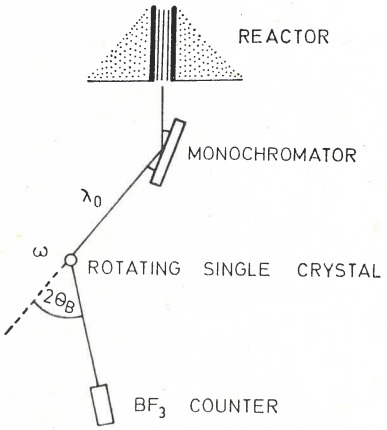

Fig. 3. Experimental set up (schematically) in the case when a monochromatic neutron beam is used (Double Axis Crystal Spectrometer — DAS)

$\lambda = (1.19 \pm 0.01)$ Å is scattered by a spinning single crystal with the zone axis parallel to the axis of rotation and perpendicular to the plane of the figure.

The intensities of neutrons scattered by the single crystal are measured by the BF_3 proportional counter placed in the Bragg position for the reflection under investigation.

[1] The authors are obliged to Drs J. Leciejewicz, A. Murasik and A. Oleś for permission to use the spectrometers.

Because of the lack of collimator in front of the detector, it was possible to measure the integrated intensity for a given reflection without moving the arm of the spectrometer.

Some of the single crystals were also investigated by the TOF technique. The principle of this method is described in Ref. [2].

The samples, prepared from different single crystals, were in the form of cylinders or slabs, as shown in Table I. In the case of cylinders a special set of cadmium slits was

TABLE I

	Reflection	Method	Shape	$F_{calc} \times 10^{-12}$ cm	$F_{exp} \times 10^{-12}$ cm	$\frac{\lvert\Delta F\rvert}{F_{calc}} \times 100\%$
Bi	$1\bar{1}0$	TOF	slab	1.67	1.86	17
	$1\bar{1}0$	DAS	cylinder	1.67	1.67	1
	$1\bar{2}1$	DAS	cylinder	1.51	1.51	1
	$2\bar{2}0$	DAS	cylinder	1.26	1.48	16
Zn	110	TOF	cylinder	1.12	1.16	4
	110	DAS	cylinder	1.12	1.39	24
	220	DAS	cylinder	1.12	1.39	24
Al	222	TOF	slab	1.21	1.09	10
	222	DAS	slab	1.21	1.09	10

$\Delta F = F_{calc} - F_{exp}$.

used in order to permit the application (with satisfactory approximation) of the formula (1) derived for a slab in symmetrical transmission geometry. All measurements were made at room temperature.

Typical results for both methods are shown in Fig. 4 and 5. They show the integrated intensity of the reflections $(1\bar{1}0)$, $(2\bar{2}0)$ of a rotating bismuth single crystal as a function of the spead of rotation. The circles are the measured values, whereas the solid curves are

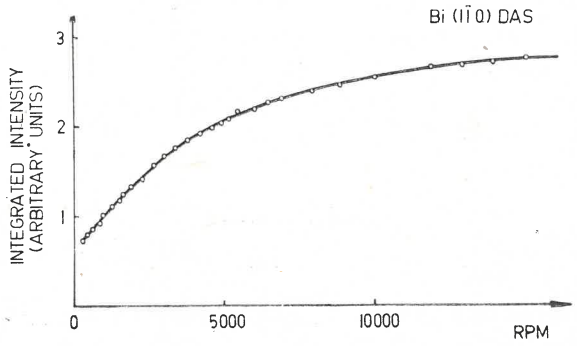

Fig. 4. Integrated intensity (arbitrary units) *versus* frequency of rotation (RPM) for the $(1\bar{1}0)$ reflection of bismuth. The circles represent the experimentally measured integrated intensities (DAS method) and the curve is calculated by the least squares fit (see text)

calculated as described in Section 2. Similar curves were obtained for aluminium and zinc single crystals.

Table I shows the structure factors F_{exp} obtained from the least squares fit (with the use of formula (4)) and the structure factors F_{calc} calculated from the known structure (the latter ones contain simple Debye-Waller factors).

The relative deviations[2] $\frac{|\Delta F|}{F_{calc}}$ from the calculated values are quite large. However, it should be remembered that the deviation is related to the arbitrary chosen single reflection. In addition, as mentioned in the introduction, the measurements were made in a very

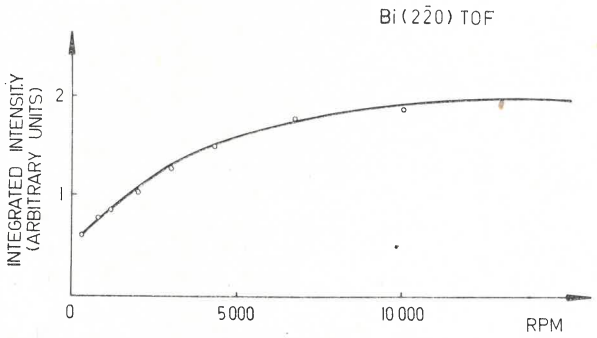

Fig. 5. Integrated intensity (arbitrary units) *versus* frequency of rotation (RPM) for the (2$\bar{2}$0) reflection of bismuth. The circles represent the experimentally measured integrated intensities (TOF method) and the curve is calculated by the least squares fit (see text)

crude way. No corrections for thermal diffuse scattering, absorption *etc.* were made. It should also be recalled that the classical theory of extinction [4] was applied, which is not the most advanced one [3]. Moreover, the Debye-Waller factors have been estimated in a very simple way and most probably they are not very accurate.

Taking all this into account the agreement seems to be not too bad. Thus we believe that the proposed method may present a useful tool for the determination of the absolute structure factor of single crystal with high secondary extinction. However, this conclusion must be checked by much more precise measurements and calculations.

The authors are obliged to Dr Bente Lebech for reading the manuscript and for interesting comments. Thanks are due to Miss B. Czerwińska and Miss B. Wentowska for their help in the numerical calculations.

REFERENCES

[1] B. N. Brockhouse, *Inelastic Scattering of Neutrons in Solids and Liquids*, International Atomic Energy Agency, Vienna 1961, p. 113.
[2] B. Buras, T. Giebultowicz, W. Minor, A. Rajca, *Nuclear Instrum. Methods*, **77**, 13 (1970).
[3] W. H. Zachariasen, *Acta Cryst.*, **23**, 558 (1967).
[4] W. H. Zachariasen, *Theory of X-Ray Diffraction in Crystals*, J. Willey, New York 1945.

[2] $\Delta F = F_{calc} - F_{exp}$.