LETTER TO THE EDITOR

Applicability of Neutron Double Bragg Reflections at Small Scattering Angle to a Study on Deformation of Metallic Crystals

In the past the majority of studies on multiple reflections (double, triple and quintuple) of X-rays or neutrons from mosaic crystals have been carried out with the aim of carrying out the extinction corrections for large angle Bragg reflections^{(1)~(7)} and corrections for weak diffuse scatterings⁽⁸⁾ since the true scattering intensities will be superposed on the effects due to multiple scattering, in particular double Bragg reflections (D.B.R.). A complete survey of D.B.R. of X-rays was presented⁽⁹⁾ and the various aspects of them are now fairly well understood. In order to discuss quantitatively the effects of cold work on X-ray small angle scattering intensities, a general theory⁽¹⁰⁾ of the intensities due to D.B.R. has been developed for any powder samples that cold work in a sample breaks up the original grains into small sub-grains with a small variation in orientation and the enhanced inter-grain contribution from these groups of sub-grains can produce a strong small angle scattering. The theory also suggests the possibility that small angle scattering measurements can provide the information on the orientation of the fragments or sub-grains, that are not available from line broadening measurements. The purpose of this paper is to show that the characteristics of neutron D.B.R. at a small scattering angle due to the mosaic structure in crystal can be applied to a study on deformation of large metallic crystals as a function of crystallographic directions $[hkl]$ since the D.B.R. intensities are strongly changed by the application of pressure to the crystal.

The measurements were performed for a single crystal of lead ($\phi 50 \times T20$) by means of the time-of-flight technique, using "white" pulsed neutrons from a chopper installed at Kyoto University Research Reactor. In principle, this technique can simultaneously provide the D.B.R. characteristics for each $(hkl) - (\bar{h}\bar{k}\bar{l})^\dagger$ reflection since the variables in the Bragg equation are now λ , d instead of θ , d , where λ is the neutron wavelength, d is the interplanar spacing of the (hkl) planes and θ is the Bragg angle for the (hkl) reflection, respectively. The value of λ for any particular reflection can be determined by carrying out time-of-flight analysis for the time, while the neutrons travel the distance from the chopper, via the sample, to the detector, which is proportional to the neutron wavelength. The experimental arrangement is schematically shown in Fig. 1 together with the names, materials and dimensions used in this experiment. The divergent angle of incident neutrons seen by the sample B is fixed to about one minute of arc by a fine collimator D and a fine collimator E. The direction $[010]$ of crystal was set parallel to the direction of incident neutrons and the vertical ω -axis of the goniometer is parallel to the $[001]$ direction of crystal. The neutrons scattered at a definite angle of 5.82 mrad are detected by a Li-glass detector C through a fine collimator F, with a divergent angle of 5.24 mrad, as shown in Fig. 1. In this investigation, the wavelength resolution $\Delta\lambda/\lambda = \Delta t/t$ is about 5%, where Δt is the time resolution determined by the time-width of neutron burst and the channel width of the time analyzer and t is the time taken by the neutrons to travel the total flight path length.

[†] In this paper, the successive Bragg reflections from (hkl) and $(\bar{h}\bar{k}\bar{l})$ planes are denoted by the expression $(hkl) - (\bar{h}\bar{k}\bar{l})$.

Symbol	Name	Materials	Dimensions
A	Chopper	K-metal alloy	(Straight slit) $W2 \times H44 \times L250$, 8 slits
B	Sample	Lead	(Single crystal) $\phi 50 \times T20$
C	Neutron Detector	Li-glass	$\phi 50 \times T0.5$
D	Fine Collimator I	K-metal alloy	$W2 \times H44 \times L250$, 8 slits
E	Fine Collimator II	Iron (Cd-plate lining)	$W1.5 \times H30 \times L1000$, 1 slits
F	Fine Collimator III	Boric carbon	$W6 \times H25 \times L400$, 1 slits
G	Detector Shielding	Boric carbon, Boric acid Heavy concrete	$\sim 1 \text{ m}^3$

Fig. 1 Schematic description of the experimental arrangement together with the names, materials and dimensions (in mm) of apparatuses used in this experiment.

Fig. 2 Double Bragg reflection (D.B.R.) patterns of neutrons by a single crystal of lead at a scattering angle of 5.82 mrad. Arrows indicate the neutron wavelengths calculated for $(hkl) - (\bar{h}\bar{k}\bar{l})$ reflections. (a) D.B.R. pattern without stress. (b) D.B.R. pattern when a stress of 0.98 MPa is applied to the direction $[110]$ of crystal. (c) D.B.R. pattern when the stress is removed.

Table 1 Observed channel numbers of double Bragg reflection peaks at three flight path lengths, neutron wavelengths for the reflections and corresponding crystal lattice planes for the reflections.

Numbers of coherent peaks	Flight lengths from Sample, L_{1-3} (cm)			Neutron wavelength (nm) $\lambda_{\text{obs.}}, (\lambda_{\text{cal.}})$	Crystal lattice planes for double Bragg reflections (hkl)-($\bar{h}\bar{k}\bar{l}$)
	57	77	95		
(1)	78(80)	80(83)	82	0.33(0.33)	(111)-($\bar{1}\bar{1}\bar{1}$)
(2)	64~66(65)	67~69(68)	70	0.27(0.27)	(311)-($\bar{3}\bar{1}\bar{1}$)
(3)	61(61)	63(63)	65	0.26(0.25)	(040)-(040) (220)-(220)
(4)	51(51)	53(53)	55	0.21(0.20)	(420)-(420)

(): Channel numbers calculated on the basis of elastic scattering of neutrons.

A scattering pattern in time of flight made at a scattering angle of 5.82 mrad is shown in Fig. 2(a). Energies of scattered neutrons were estimated from channel shifts of peaks in the pattern when the flight path length from crystal was changed from 57 to 77 cm and 95 cm in order to examine the contributions of inelastic scattering. It is shown in Table 1 that the observed channel numbers and their estimated wavelengths for each peak at the three flight path lengths agree well with the calculated ones on the basis of elastic scattering for the Bragg reflections, (111)-($\bar{1}\bar{1}\bar{1}$), (311)-($\bar{3}\bar{1}\bar{1}$), (040)-(040), (220)-(220) and (420)-(420). It has been shown that these peak intensities result from the so-called double Bragg reflections from mosaic blocks and fall off rapidly with scattering angle, i.e. decrease to about 1/4 and 1/10 at scattering angles of 11.6 and 17.5 mrad, respectively.

To investigate the change of D.B.R. pattern due to crystal deformation, a compressive stress of 0.98 MPa was applied to the [110] direction of the crystal during the measurement. The pattern, as shown in Fig. 2(b), indicates that the peaks of (040)-(040) and (220)-(220), which are observed at the same neutron wavelength, disappears and the peak height of (420)-(420) becomes about three times higher. And the two peaks of (111)-($\bar{1}\bar{1}\bar{1}$) and (311)-($\bar{3}\bar{1}\bar{1}$) become fairly prominent. This observation shows that the mosaic structure varies depending upon the crystallographic directions due to the deformation. Successively, the same measurement was carried out after the stress was removed. As shown in Fig. 2(c), the pattern becomes almost completely the same one as in Fig. 2(a) except the peak of (111)-($\bar{1}\bar{1}\bar{1}$).

Some discussions for these experimental results may be summarized as follows:

(a) The D.B.R. in this experiment occurs only at a small scattering angle due to the mosaic spread in crystal since its intensity falls off rapidly with increasing scattering angle.

(b) Under such a low stress as 0.98 MPa, the mosaic structure of crystal can almost elastically change and its nature depends upon crystallographic directions. This means that the variation of mosaic spread depends upon the direction.

(c) When a single crystal is slightly deformed, crystal defects such as dislocations and foreign atoms move in the volume of crystal due to stress. This changes the correlation in orientations of mosaic blocks with slightly varying orientations and the dimensions of coherent regions (or blocks) so as to satisfy the balance conditions of stresses determined by the resolved shear stress on crystal planes. The disappearance of (040)-(040) and (220)-(220) reflections in the pattern is due to the decrease in the mosaic spread by the stress applied

to crystal. This situation produces a forward scattering since the mosaic blocks become rigorously parallel.

(d) Another point of interest is the large increment in (420)-(420) and (111)-($\bar{1}\bar{1}\bar{1}$) reflections. In these cases the cold work will make the mosaic spread a little bit broader, which is reverse to the case (c). This means that individual mosaic blocks are considered to be rotated by slightly different angles through the mean reflecting position. Consequently the incident beam will be able to penetrate much more deeply into the crystal. In addition, the first reflected beam now has a much higher probability of meeting suitably oriented blocks for the second reflection than was the case of a sample in which the neighbouring blocks have a fairly narrow spread. This effect can intensify D.B.R. over large angles including the scattering angle of 5.82 mrad. The distinguished nature of the (111)-($\bar{1}\bar{1}\bar{1}$) reflection is that its shape and intensity are irreversible even after the stress is removed. This reveals a plastic deformation of the mosaic structure related to a work of the slip system $\{111\}-\langle 1\bar{1}0 \rangle$ in *fcc* metals.

In conclusion, this experimental work shows that the information on the mosaic structure change in crystal can be provided for each direction through the measurements of double Bragg reflections of neutrons and be applied to the problems of the strength and fracture of crystals.

REFERENCES

- (1) S. Miyake: *X-ray Diffraction*, in Japanese, Asakura Syoten (1969), 202-221.
- (2) G. E. Bacon: *Neutron Diffraction*, 3rd ed., Clarendon Press, Oxford (1975), 65-93.
- (3) G. E. Bacon and R. D. Lowde: *Acta Cryst.*, **1** (1948), 303.
- (4) W. H. Zachariasen: *Acta Cryst.*, **18** (1965), 705.
- (5) W. H. Zachariasen: *Acta Cryst.*, **23** (1967), 558.
- (6) S. G. Bogdanov and A. Z. Menshikov: *Acta Cryst.*, **A32** (1976), 869.
- (7) M. J. Cooper and K. D. Rouse: *Acta Cryst.*, **A32** (1976), 806.
- (8) Y. Nakai: *J. J. Appl. Phys.*, **13**, No. 7 (1974), 1061.
- (9) *Small Angle X-Ray Conference Proceedings*, J. Appl. Phys., **30**, (1959), No. 5, 601-674.
- (10) B. E. Warren: *Acta Cryst.*, **12** (1959), 837.

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