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Relation between Deformation and Integrated Intensity of X-Ray in Aluminium Single Crystals*

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Synopsis

The relation between the deformation and the integrated intensity of X-ray was investigated with aluminium single crystals, and it was found that with increasing slip deformation the reflected intensity first increased rapidly, and then gradually reached a saturation at about 2 per cent elongation, irrespective of the tensile direction relative to the crystal orientation. The preferential deformation at the surface layer was deduced from the change in integrated intensity of X-ray reflected from specimen of various thicknesses prestrained by a few per cent. It was seen that the region giving a maximum reflected intensity was not the outermost surface of crystal but the inner layer about 10μ from the surface, and that with increasing deformation this region extended into the crystal. Further, the orientation dependence of the reflecting power in a plastically bent state was examined on (111) reflection by a Geiger counter diffractometer, and it was seen that the change in the reflected intensity related to the crystallographical relation of stress direction.

I. Introduction

Some works on the internal structure of a real crystal have been reported(1), and recently, such a problem has been developed by the introduction of dislocations. Mott(2) proposed a model of net work of dislocations in crystal from the suggestion by Frank, which was verified by an ingeneous experiment on AgCl(3) single crystal. Furthermore, the dislocations and their arrangements and motions were directly observed by the aid of modern techniques on electron-microscope. Nowadays, the defect in crystal is explained by mutual actions of dislocations, stacking faults, vacancies, interstitial atoms and impurity atoms, and so a more precise explanation of the mechanism of deformation and the interpretation of the mechanical properties of a material will be obtained from the internal structure of a real crystal.

When metals and alloys are cold-worked, the changes in the integrated intensity, the line broadening, the peak shift and the asymmetrical line shape of X-ray are generally observed. It was difficult to separate the effects of the various defects in a crystal induced by deformation. However, the effects have recently been separated by the method of Fourier analysis(5). In the case of a single

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* The 985th report of the Research Institute for Iron, Steel and Other Metals.
(1) P. B. Hirsch, Progress in Metal Physics, **6** (1956), 236.
(2) N. F. Mott, Proc. Roy. Soc., **B64** (1951), 729.
(3) J. M. Hedges and J. W. Mitchell, Phil. Mag., **44** (1953), 223.
(4) P. B. Hirsch, *Internal Stress and Fatigue in Metals*, p. 139.
(5) B. E. Warren and B. L. Averbach, J. Appl. Phys., **21** (1950), 595.

crystal, the effect of deformation is mainly characterized by the change in integrated intensity, because it is very sensitive to the degree of perfection of a crystal. The present author(6) has investigated this problem with some metal single crystals, and the following results have been obtained: (1) a large effect of primary extinction is observed in aluminium single crystals, and the increase of integrated intensity of X-ray due to deformation from the active slip plane is larger than that from the latent slip planes: (2) this increase is saturated at a very early stage of the deformation. In these results, the former will show that the disturbance of crystal lattice due to slip deformation is related to the orientation relative to the tensile axis or to some minute difference in structural defects, and the latter will signify that the crystal soon becomes very imperfect by the slight deformation. However, the hardness is dependent upon the orientation, and also preferential perturbation of strain at the surface layer of crystal is clearly observed in an early stage of deformation.

Hence, the present experiment was carried out with some metal single crystals to throw light upon the relation between the deformation and the integrated intensity of X-ray.

II. Experimental procedure

In order to investigate the change in integrated intensity due to deformation, a special tensile testing machine which is capable of the measurement of integrated intensity of X-ray by the method of crystal rotation and of the microscopic observation under loading. The testing machine was rotated at the rate of two revolutions per minute by a synchronous motor. The coincidence of the rotation axis and the tensile axis of a specimen was obtained by a flexible grip. Time required for one set of experiment was about 8 hrs. The irradiation of X-ray was made under loading, but no creep was observed. The slits used was of a rectangular form $1\times2\,\mathrm{mm}^2$, and X-ray tubes of copper and iron targets made by Philps in Holland were used at 20 kV, 20 mA and 15 kV, 20 mA. The voltage regulator was used in order to stabilize the tension, the fluctuation being controlled within 1 per cent. The constancy of supplied energy at every exposure was confirmed by the ratio of blackness of a film by X-ray incident from another window, and the intensity of X-ray was reduced by nickel or manganese plate of suitable thickness. The photographic density-exposure curve on the film used was examined preliminarily, and the linear portion of the curve was used in the present experiment. From these results of preliminary experiment the time of suitable exposure was seen to be 50 min. The distance from the specimen to the film was 120 mm. In order to keep conditions constant during the treatment, the X-ray films were developed simultaneously in a large bath. The change in the reflected intensity was denoted by the relative value, that is, by the ratio of the

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(6) T. Sutoki and K. Nakajima, Sci. Rep. RITU, **A10** (1958), 73.
K Nakajima and S. Koda, Nature, recently be appeared.

intensity reflected from the crystal in a deformed state to that in the annealed state. The reflected intensity was measured by a microphotometer.

The crystal specimens were made by the following process: aluminium 99.995 per cent in purity was cold-rolled into strip or cold-drawn into wire, and after etching and washing, these samples were annealed at 500°C for 2 hrs, and after subjecting to 2 per cent elongation they were heated at 600°C for 150 hrs. Of single crystals thus obtained those of the following three kinds of size were used: 40 mm × 1.5 mmϕ, 40 mm × 4 mmϕ and 7 mm × 0.5 mm × 20 mm.

The distribution of strain in a crystal at an early stage of tensile deformation was first examined. It is well-known that the surface of a metal is strained preferentially by an external force. The heterogeniety of deformation can be deduced from the change in the integrated intensity provided a suitable wavelength of radiation is used. Intensity changes of X-ray in the specimens prestrained by a few per cent were measured with reducing diameter by dilute aqua regia. In this case, circular form in the original state of the specimen could not be expected because of the configurational change by the prestrain; in fact, the circular section of the specimen changed into elliptic form. The difference of the length between the major and the minor axis was about 0.2 mm after about 3 per cent elongation, and accordingly, the mean value of them was taken as the effective diameter of the specimen, and the variation of the intensity due to such a configurational change was not corrected. The observations of integrated intensity were made on the active slip plane of {111}.

Some measurements on the orientation dependence of the change in the integrated intensity on (111) plane due to simple bending were also carried out by Geiger counter diffractometer of XRD-5 made by General Electric Co. in U.S.A. X-ray tube of copper target was used at 15kV 7mA.

III. Experimental results and discussion

The reflected intensity may generally vary with irradiated position, and consequently, for the present purpose the same position of a crystal should always be irradiated before and after the deformation, which was almost realized in the whole course of the experiment by the aid of the above-mentioned tensile machine. Figs. 1 and 2 show the intensity change of the X-ray for various orientations of the specimen. From these results it may be concluded that (1) the integrated intensity increases rapidly irrespective of the orientation relative to the tensile axis, and is saturated at a few per cent elongation; (2) the saturation value is, however, different with different specimens. Fig. 3 shows the stress-strain curves of the above specimens. The integrated intensities were observed at the states indicated by solid circles in these curves, the applied load being maintained for 50 min. at these states. It will be seen

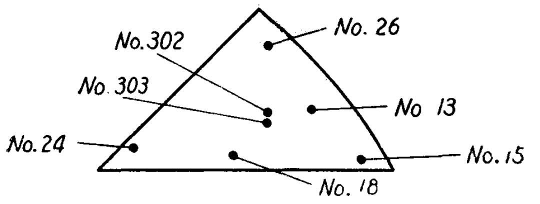

Fig. 1. Orientation of crystals.

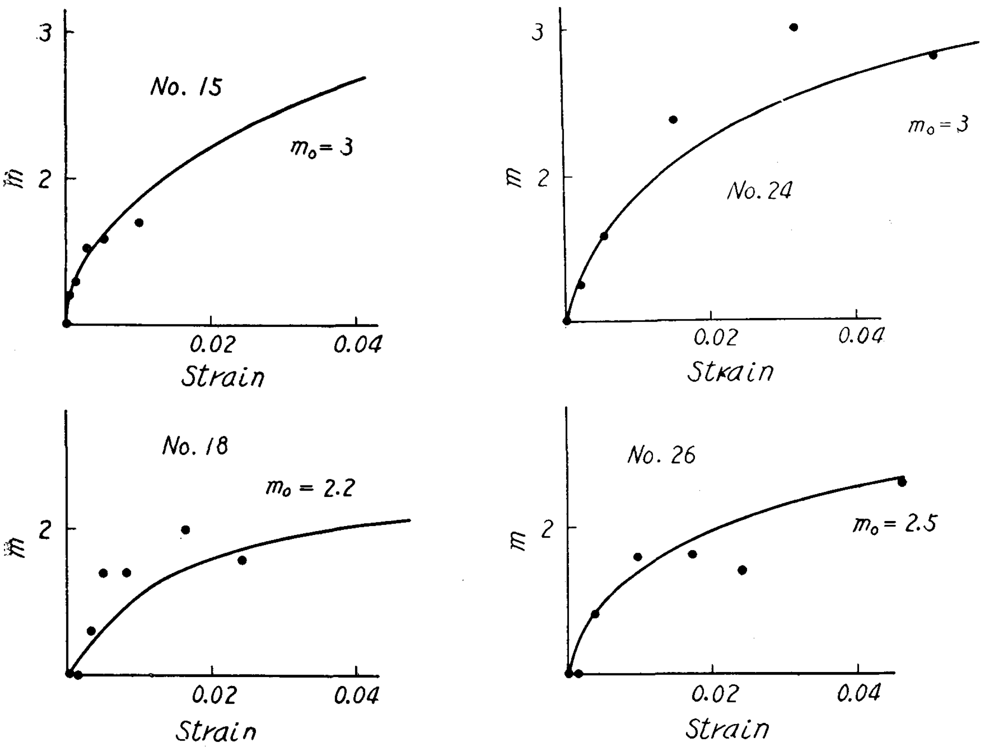

Fig. 2. Relation between integrated intensity and resolved shear strain. m_0 of each specimen was obtained from the ratio of the polished and the original state.
The full line shows the result of calculation by Eq. (1)

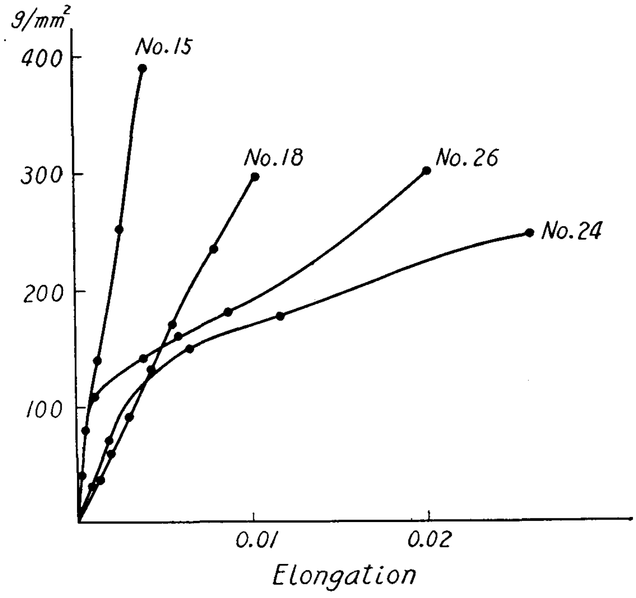

Fig. 3. Load-elongation curve.

clearly that the four stress-strain curves show the well-known behaviours characteristic to the orientation of specimen, whereas the change in the integrated intensity is almost independent of the orientation of the specimen. With increasing tension the heterogeneous deformation of crystal will appear by the formation of locally strained regions, for example, by deformation bands etc., and accordingly, the reflection from these regions will be enhanced strongly compared with other regions of the crystal. The direct evidence for these phensmena was shown clearly by the method of X-ray microscopy by Nishiyama and Hayami.(7) However, the increase of the reflected intensity begins at a very early stage of

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(7) Z. Nishiyama and S. Hayami, Mem. Inst. Sci. and Ind. Research, Osaka Univ., **11** (1958), 129.

deformation, and reaches rapidly a maximum as shown in the present experiments, that is, at this stage the strained region observable microscopically does not generally appear. It is important to note that not all kinds of strains are detectable by X-ray diffraction, that, is, the crystal lattice are not disturbed by a perfect glide, and accordingly, the change in the diffraction of X-ray due to such deformations should not be observable. In the interpretation of the relation between the integrated intensity and the deformation of crystal, the present author has pointed out(8) that the saturation value is taken as the relative intensity reflected from the ideally mosaic crystal, and that if the reduction in the reflected intensity is due only to primary extinction, the increase of reflecting power of crystal plane due to deformation will be explained as caused by the break up of elementary block. According to the present results, the relation between the relative intensity reflected and the deformation is shown by the following equation:

$$m=m_0\frac{\tanh L\cdot q/d}{L\cdot q/d}, \qquad m_0=\left(\frac{\tanh L_0\cdot q/d}{L_0\cdot q/d}\right)^{-1}$$

$$=m_0\frac{\tanh q/d\cdot\dfrac{a}{S+a/L_0}}{q/d\cdot\dfrac{a}{S+a/L_0}}.$$

$$q=\frac{e^2}{2mc^2}\,\frac{\lambda^2}{\sin^2\theta}\cdot N\cdot|F|, \qquad L=nd, \tag{1}$$

where n is the number of net planes contributing to X-ray diffraction, N the number of unit cell per unit volume, L_0 the block size in the original state, and d the spacing of the reflecting planes. The relation between the resolved shear strain, S, and the block size, L, is given experimentally as shown in Fig. 4, and the following experimental formula is obtained

$$\frac{1}{L}=\frac{S}{a}+\frac{1}{L_0}, \tag{2}$$

where a is a constant concerning the block size in final state and of the order of 5×10^{-6} cm. The relation similar to Eq. (2) was obtained by Taylor. Fig. 2 shows the result obtained by the above equation for each specimen. From this equation it will be seen that the relative intensity is expressed only by the strain. The assumptions contain some questions; nevertheless, the experimental results were interpreted satisfactorily by this equation.

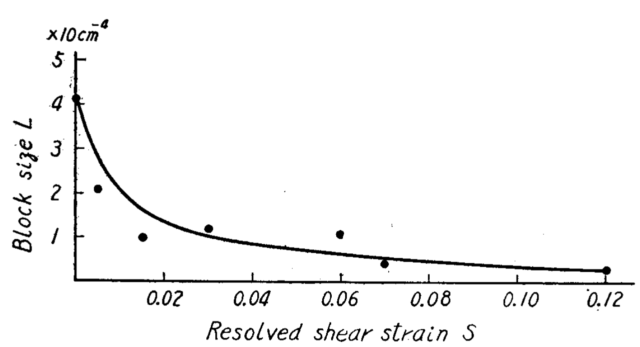

Fig. 4. Relation between block size and resolved shear strain obtained from the specimen No. 13. The full line shows the result of calculation by Eq. (2).

The penetration of X-ray into any material is dependent

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(8) K. Nakajima, Symposium on Phys. Soc. Japan, Oct., 1958.

upon the wavelength of the radiation, for instance, in the case of Cu-K_α radiation the half penetrating depth is about 20μ for aluminium, which is also the thickness of layer contributing to X-ray diffraction. Therefore, the structural change with the change in the thickness of specimen may be seen by utilizing a suitable wavelength of radiation; in fact, some theoretical and experimental works(9) on the preferential deformation at the surface layer due to external force have been reported for various metals and alloys. Such an inhomogeneity of deformation may effectively be observed by thick specimens. Figs. 5 and 6 show the change in the integrated intensity from the active slip plane of the specimens prestrained by 0.3 and 1.5 per cent respectively; in these cases Fe-K_α radiation was used, and the half depth of penetration for aluminium was calculated to be about 10μ. The reduction in thickness of the specimen by etching was slightly different with different orientations of the crystal as shown in Table 1, and so the mean length of the major and the minor axis was taken as the effective diameter of the specimen. Two interesting results were obtained: (1) the layer giving a maximum reflected intensity was situated about 15μ inward from the surface of the specimen as shown in Fig. 5; (2) the increase in the relative intensity reflected was measured only up to about 30μ in thickness, and then with decreasing thickness of specimen the intensity decreased rapidly up to the value of the original state. On the other hand, with increasing prestrain these changed partly as shown in Fig. 6, from which it will be seen that the

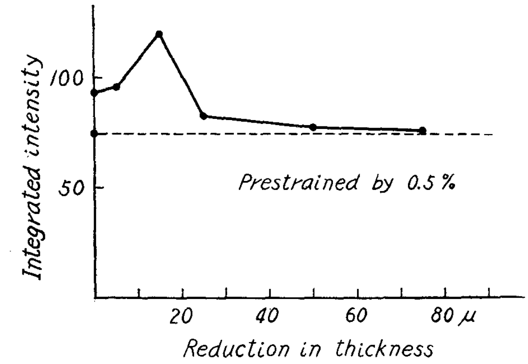

Fig. 5. Change in integrated intensity as the function of reduction in thickness.

Table 1. The reduction in thickness of specimen by etching.

No. 302 prestrain 0.5%			No. 303 prestrain 1.5%		
max.	min.	mean	max.	min.	mean
4.28	4.07	4.18	4.68	4.44	4.56
4.27	4.06	4.17	4.66	4.42	4.54
4.25	4.05	4.15	4.64	4.41	4.52
4.22	4.03	4.13	4.62	4.38	4.50
4.18	3.98	4 08	4.60	4.36	4.48
4.13	3.92	4.03	4.58	4.34	4.46
			4.54	4.31	4.43
			4.50	4.26	4.38

in mm.

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(9) A.H. Cottrell, *Dislocations and Plastic Flow in Crystals* 1954, p. 86.
K. Sumino, Unpublished

region giving a maximum reflected intensity extends toward the interior of crystal compared with the case shown in Fig. 5. These results will be interpreted as follows: the thin layer of the surface of specimen is first strained, and then the strained region advances gradually toward the interior of the crystal. As mentioned above, the region corresponding to the maximum reflected intensity is not the outermost surface of the specimen but the inner part about 10μ from the surface; this will be explained as caused by the rapid releasing of strain in the surface.

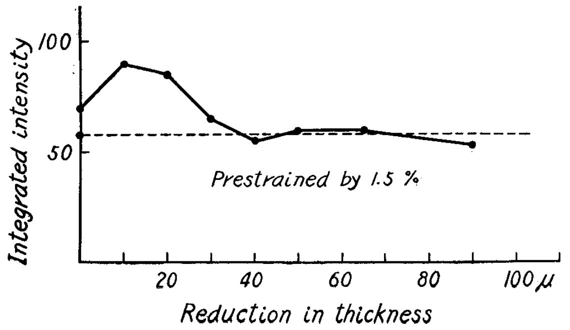

Fig. 6. Change in integrated intensity as the function of reduction in thickness.

From the above results it is clearly seen that the increase in the reflected intensity of X-ray due to the tensile deformation relates to the strained surface layer of the specimen. However, the distortion in the slip plane is different with different orientations in the plane. The observation of this variation will give some informations about the mechanism of deformation, but as shown in the above results, the strained region is restricted within a narrow limit, and therefore, it is difficult to perform such an examination. However, the above purpose will be achieved, if the orientation dependence of the reflected intensity is examined with a simply bent crystal. The crystallographical relation of the specimen used is shown in Fig. 7, Photo. 1 being its Laue photograph. The deviation of the surface normal from [111] was about 1 degree. The preparation of the single crystal of preferable orientation needs generally some special technique, for instance, as shown in Fujiwara's method.(10) However, the preparation of such a specimen from the materials of high purity was difficult. The single crystal referring to Fig. 7 was selected from the many samples. The specimen was bent around [11$\bar{2}$] axis, the bending being carried out by using

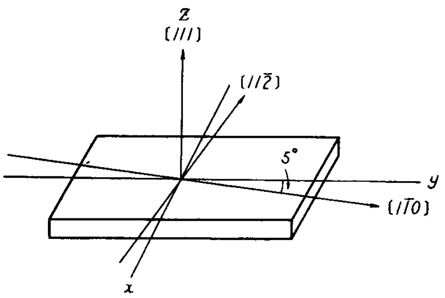

Fig. 7. Crystallographical relation of the specimen No. 203.

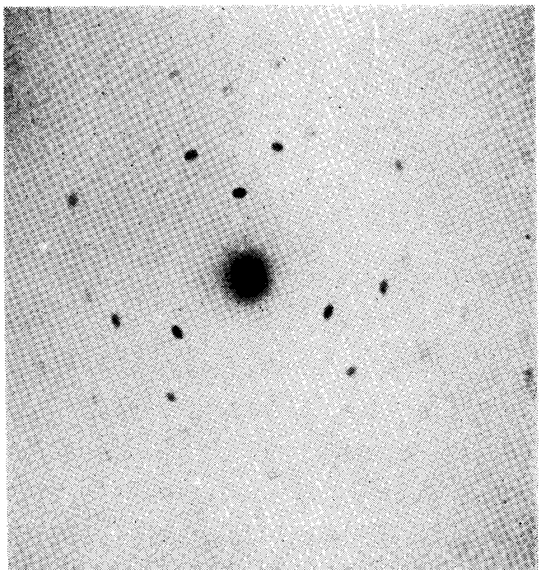

Photo. 1. Laue photograph of the specimen No. 203.

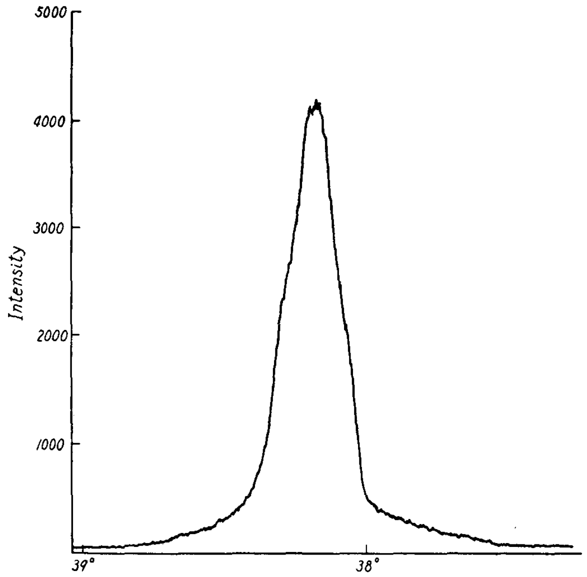

Fig. 8. Rocking curve from the aluminium single crystal before bending. $\varphi=-15°$

the surface of an iron cylinder 300 mm in diameter. A slight change in the diffraction with the incident direction is generally observed because of the imperfection of crystal. The detailed investigation of the diffraction as a function of incident direction was carried out with a specimen of the original state. The standard slit was used, the angular extent of divergency of X-ray beam being 0.4° and the irradiated area being of the order of 3mm^2. The rocking curves were recorded automatically, an example of which is shown in Fig. 8. The changes in the integrated intensity with the angle φ between $[1\bar{1}0]$ direction and the direction of the projection of the incident beam on the surface are shown in Fig. 9. The true half width of the reflection is estimable from the equation by Lambot et al.(11) :

$$L=\left|D\frac{\sec\theta}{2d}\lambda\right|+\left|f\frac{\sin(\theta+\alpha)}{\sin(\theta-\alpha)}\sin(\theta-\alpha+\beta)\right|$$

$$+\left|E\frac{\sin2\theta}{\sin(\theta-\alpha)}\right|+\frac{1}{8}Dw'\tan\theta\,, \qquad (3)$$

where α : angle between the reflecting plane and the surface of specimen,
β : angle between the surface of crystal and the plane parpendicular to the incident beam,
f : the breadth of incident beam on the surface of specimen,
E : effective thickness of the specimen,
D : the distance between film and specimen,
w' : angular extent of divergent beam.

The half width was of the order of 30″ in some cases. The orientation dependence of the diffraction due to simple plastic bending is shown in Figs. 9 and 10. Fig. 9 shows the changes in the integrated intensity with φ, from which a large increase in the integrated intensity compared with the original state of the specimen, and the change in intensity with increasing φ will be seen clearly, that is, the reflected intensity increases gradually with the increase of φ, and reaches a maximum value at $\varphi=0°$ or $[1\bar{1}0]$ direction, and then decreases gradually. This result will show that the distortion after simple plastic bending is larger

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(10) T. Fujiwara, J. Phys. Soc. Japan, **10** (1955), 355.
(11) M. Lambot, L. Vassamillet and J. Dejage, Acta Met., **1** (1953), 711.

in the orthogonal direction (or $[1\bar{1}0]$ direction) than in the bending axis (or $[11\bar{2}]$ direction), and that the distortion reaches a minimum at the direction of the bending axis. The horizontal line in this figure shows the integrated intensity obtained from the surface polished mechanically. It is interesting to note that a large change in the integrated intensity in $[11\bar{2}]$ direction, which is the bending axis, is observed as shown in Fig. 9.

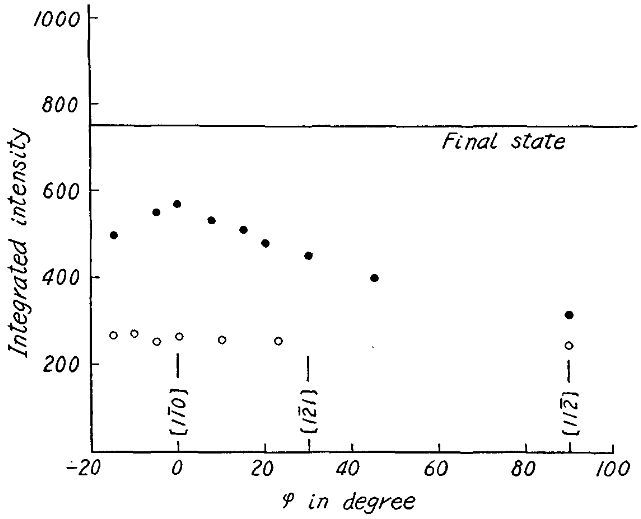

Fig. 9. The orientation dependence of the diffraction due to simple-plastic bending. Open circle refers to the integrated intensity of original state.

Wilson(12) has reported a theory of X-ray diffraction from the elastic field around a screw dislocation, and recently, Suzuki(13) has reported the same in the case of edge dislocation. Fortunately, recent techniques make it possible to obtain substances of high purity, and consequently, the preparation of single crystals of very good quality is becoming easy without serious difficulty. In fact, direct observations of dislocations in such materials have been reported by some workers.(14) However, it may probably be difficult to obtain the strain distribution due to such crystal defects, because the volume occupied by such defects is extremely small compared with the volume irradiated by X-ray.

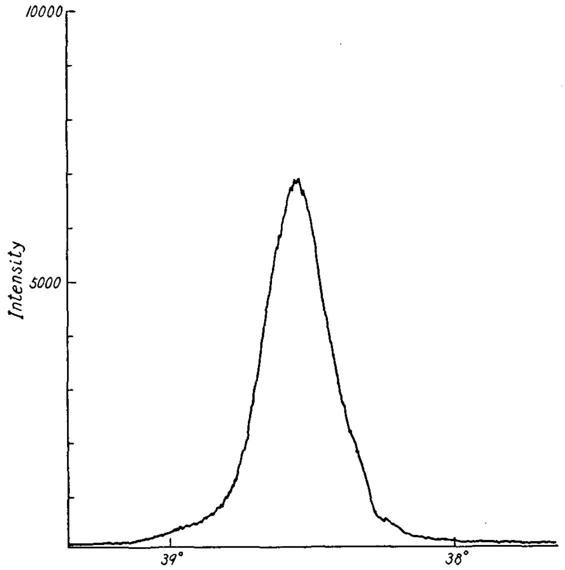

Fig. 10. Rocking curve from the aluminium single crystal after bending. $\varphi = -15°$

Summary

(1) The relation between the deformation and the change in integrated intensity of X-ray was investigated with aluminium single crystals.

(2) The intensity of the ray reflected from the active slip plane first increased

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(12) A. J. C. Wilson, Research **2** (1949), 541; **3** (1950), 387.

(13) T. Suzuki, Symposium on Phys. Soc. Japan, (1958) October.

(14) For examples. A. R. Lang, J. Appl. Phys., **29** (1958), 597; G. Borrmann, W. Hartwig and Irmler, Z. Naturforsch., **13A** (1958), 423.

rapidly and then gradually reached a saturation at about 2 per cent elongation, irrespective of the direction of tensile axis.

(3) The region giving a maximum reflected intensity after slip deformation was not the outermost surface but an inner layer about 10μ from the surface, and with increasing deformation this region extended toward the interior.

(4) In the case of simple plastic bending, the reflected intensity from (111) increased gradually with increasing angle between the direction of the bending axis (or $[11\bar{2}]$ direction) and the direction of the projection of incident beam on the surface (or (111) plane), and reached a minimum at the direction orthogonal to the bending axis.

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