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著者	SUTOKI Tomiya, NAKAJIMA Koichi
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X-Ray Study of Cold-Worked Metal Single Crystals*

Tomiya SUTOKI and Kôichi NAKAJIMA

The Research Institute for Iron, Steel and Other Metals

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Synopsis

The change in the reflected intensity of X-ray due to the deformation of metal single crystals of aluminium and zinc was examined with microphotometer by using Cu-K_α radiation. In aluminium single crystals, a large change in the relative intensity of {111}-reflections was observed with an increase of the deformation, and almost saturated at a few per cent elongation. The increasing rate of the relative intensity reflected from the active slip plane was larger than that from the latent slip planes. Similar results were obtained with zinc single crystals. The increase in the integrated intensity was explained as caused by the reduction in the extinction due to the fragmentation of crystallite blocks, from which it was concluded that the distortion after slip deformation was larger in the active slip plane than in the latent slip planes. In other words, the strain-hardening is larger in the active slip plane than in the latent slip planes. Further, the recovery of the relative intensities was examined, and it was seen that in aluminium crystal it was incomplete even after long-time annealing at high temperature, whereas in zinc crystal it was almost perfect at short-time annealing.

I. Introduction

It is well known that when a nearly perfect cyrstal is deformed elastically, the integrated intensity greatly increases(1). Recently, White(2) studied the integrated intensity of an elastically bent quartz crystal as the function of the radius of curvature of bending, and showed that an approximate theory was in good accord with the experimental results. Such an increase in the intensity may be explained as caused by the reduction in the extinction. In metal single crystals, the perfectness of the crystal structure is generally low and, accordingly, the extinction may be negligibly small compared with that in quartz crystal. The effect of extinction has been studied by James(3) with imperfect crystals, and it is also seen in aluminium crystal.

It has been shown(4) that in aluminium single crystals the relative intensity of the X-ray is greatly increased by small deformation, and that the change in the intensity of {111}-reflections is dependent on the tangential component of the external stress. Such an increase in the reflected intensity was different with different specimens, the increasing rate being, in some cases, about 3 times the ordinary value. This increase may be caused by the reduction in the extinction due to the distortion of crystal. As the extinction is related, in wide sense, to

* The 900th report of the Research Institute for Iron, Steel and Other Metals.

(1) E. Fukushima, Riken Iho, **14** (1935), 1105; 1199; **15** (1936), 1.
(2) J. E. White, J. App. Phys., **21** (1950), 855.
(3) R. W. James, Z. Krist., **89** (1934), 295.
(4) K. Nakajima and T. Hikage, Sci. Rep. RITU, **A9** (1957), 41.

the defect of crystal structure, it is natural that the increasing rate of the relative intensity is smaller in aluminum crystal than in quartz crystal.

Hitherto, X-ray investigation of lattice distortion was performed mostly on the diffusion of Debye-Scherrer lines or on the asterism of Laue spots, and scarcely on the change in the reflected intensity. Hence, the present experiment was carried out to throw light on lattice distortion from the extinction of X-ray on the basis of the above consideration.

II. Method of experiment

Single crystals of aluminium and zinc, respectively 99.995 and 99.96 per cent in purity, were made by the strain-annealing method and the Bridgman method, the size being about 1.4 mm in diameter and 30 mm in length. Specimens were etched by dilute aqua regia. The reflected intensity may generally vary with irradiated positions. 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. Crystal rotation method was used with Cu–K_α at 20 kV and 7 mA, the slit being 2×1 mm^2 in size; the exposure time was 30 minutes. The voltage regulator was used to stabilize the operation of X-ray tube, the fluctuation in the energy of incident radiation being controlled within 3 per cent over the whole exposure time. The reflected intensity was measured by microphotometer. 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 intensity reflected from the crystals in deformed state to that in the annealed state.

III. Results of experiment

The relative intensity greatly increased with increasing deformation. Figs. 1~3 show respectively the results on aluminium and zinc single crystals. From these figures it will be seen that in the case of aluminium the relative intensity first increases rapidly with deformation, and then slowly, becoming almost constant at a few per cent elongation, and that in the case of zinc, though the general tendency is similar to the former, the saturation value of the increase is considerably small. In both cases, the saturation value was generally different with different specimens. Owing to the slip deformation, the difference in the relative intensities between the crystallographically equivalent planes could be seen, as reported in the previous paper(4). The respective intensity changes in {111}-reflections with the deformation are shown in Figs. 1 and 2 and Photo. 1. From the figures, it will be seen that both the increasing rate of the intensity and the saturation value are not quite the same in these equivalent planes, being larger in the active slip plane $(1\bar{1}\bar{1})$ than in the latent slip planes. The strain-hardening of the latent planes is usually accepted to be the same as that of the active plane, but according to the present results, this is not the case.

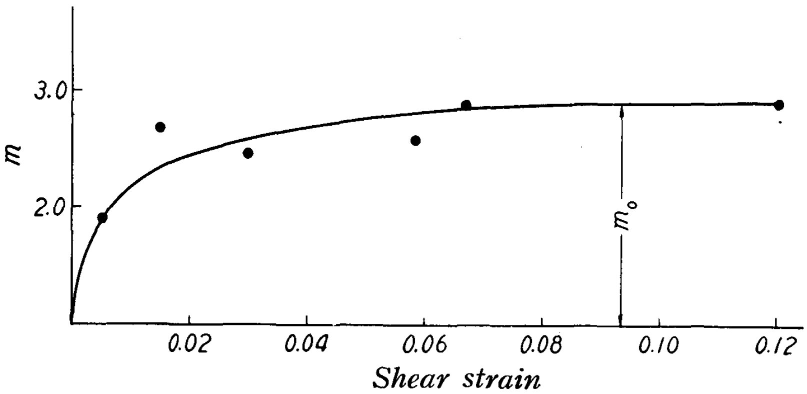


Fig. 1. Relation between reflected X-ray intensity from the active slip plane $(11\bar{1})$ of aluminium single crystal and strain.

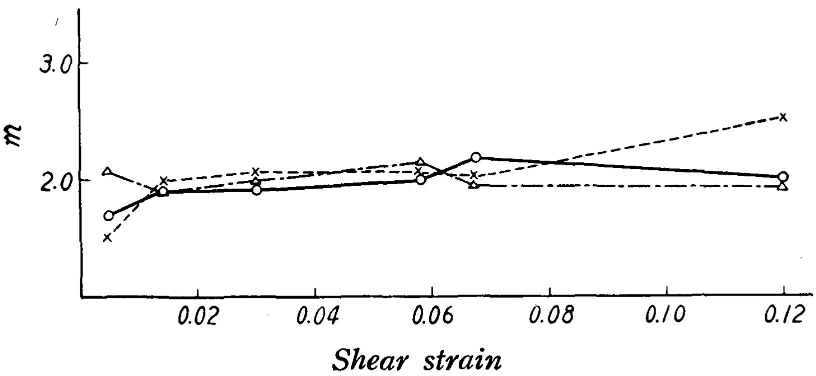
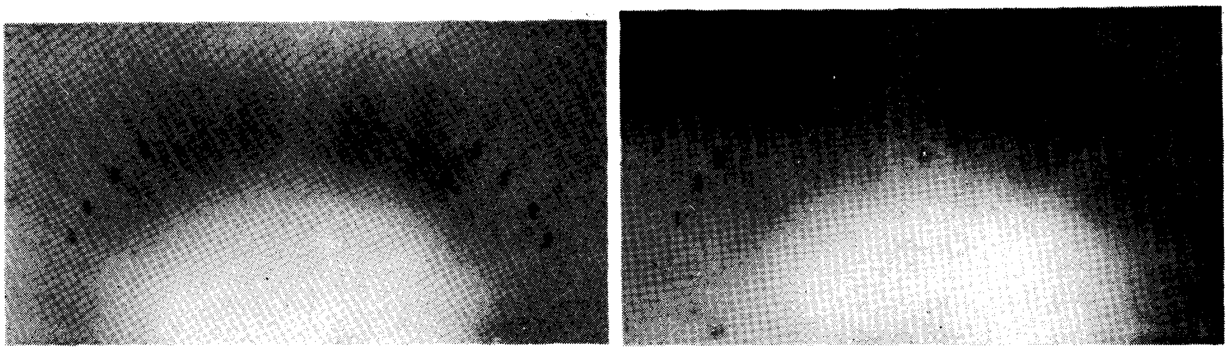


Fig. 2. Relation between reflected X-ray intensities from the latent slip planes of aluminium single crystal and strain.

--- × --- $(\bar{1}11)$ reflection
-·—△—·- $(1\bar{1}1)$ "
——○—— (111) "



(a) (b)

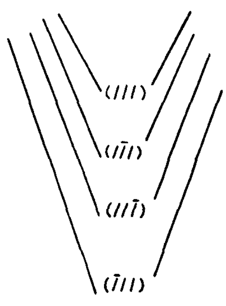


Photo. 1. Rotation photographs of aluminium single crystal.
(a) after 0.5 % strain
(b) " 6.2 % "

Slip deformation is generally accompanied with crystal rotation, and the displacements in diffraction spots must correspondingly take place. The rotation angle calculated from the displacement was only a few degrees in the present case, and hence, no correction to be made for the crystal rotation was taken into account

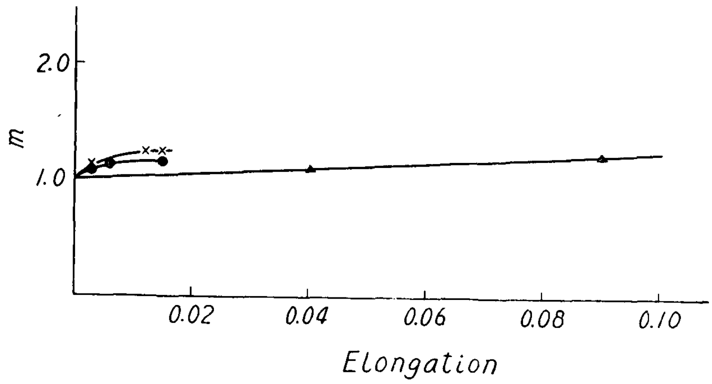


Fig. 3. Relation between reflected X-ray intensities from (0002) of zinc single crystal and elongation.

Further, the recovery of the relative intensity was examined. Aluminium single crystal was cut into two parts after a few per cent elongation, and they were annealed respectively at 300 and 600°C, and then the intensity measurements were carried out. The results are shown in Fig. 4, from which it will be seen that the relative intensity does not return to the original state even after long-time anneal

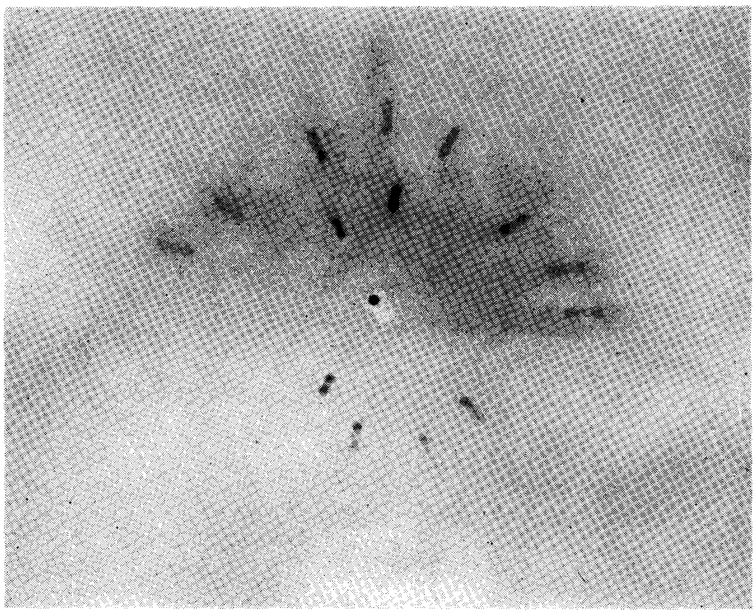


Photo. 2. X-ray photograph of aluminium single crystal showing double spots.

ing at this high temperature. In the case of zinc single crystal, however, it was almost completely restored to the original state after the annealing at 210°C for about 20 minutes as shown in Fig. 5.

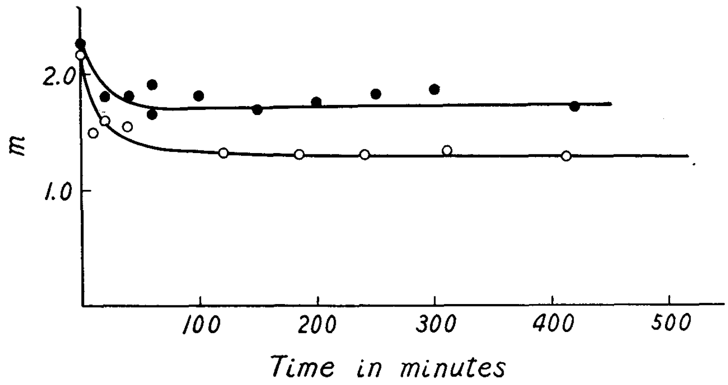


Fig. 4. Recovery of reflected X-ray intensity from the active slip plane of aluminium single crystal.
● annealed at 300°C
○ " 600°C

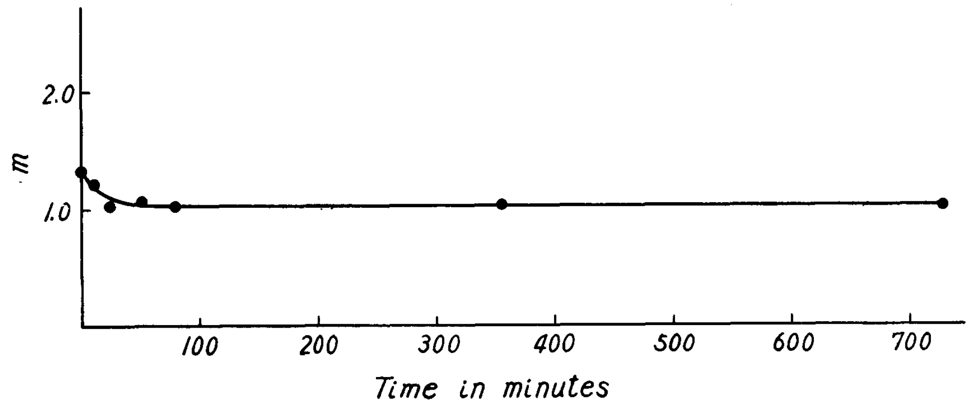


Fig. 5. Recovery of reflected intensity of (0002) of zinc single crystal annealed at 210°C.

IV. Discussion

As well known, every Laue spot diffracted from a thick crystal of quartz is double(5). This is because the intensity of reflected ray from the inner part of the crystal is greatly reduced by the extinction due to the perfectness of crystal structure, whereas the surface layer strongly reflects X-ray, owing to its imperfect structure. A great increase in the integrated intensity reflected from an elastically deformed quartz has been investigated extensively(1)(2). In aluminium used in the present experiment, double spots were also observed when the crystal was thick, as shown in Photo 2, which was obtained from a crystal of 5.53 mm in thickness, either surface of which was ground carefully with emery paper. The X-ray tube was operated at about 40 kV and 7 mA. The effective focussing diameter of electron beam was squeezed to about 0.5 mm by electric lens, the opening of the

(5) C. S. Barrett, Phys. Rev., **38** (1931), 832.
Y. Sakisaka and I. Sumoto, Proc. Math. Japan, **13** (1931), 211.

X-ray being about 10′. The relation between the interval s of a pair of reflections and the distance r from the central spot is shown in Fig. 6, which is clearly linear.

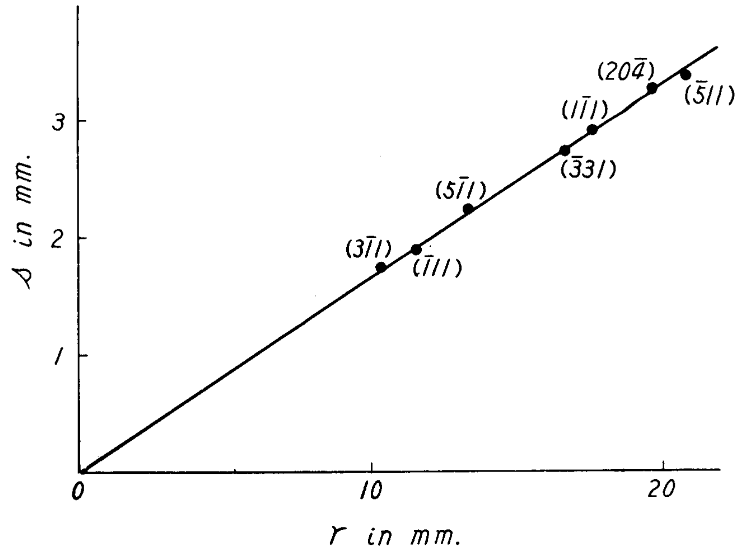


Fig. 6. Relation between interval s of a double spot and distance r from the central spot.

Therefore, the splitting can be concluded to have been caused by a strong reflection from surface layers of the crystal. When the incident beam is geometrically parallel, the mean effective thickness x of the surface layer can be obtained by simple calculation as follows:

$$x = \frac{rD - sR}{r + s/2}, \qquad (1)$$

where D is the thickness of a crystal, and R the distance between the film and the crystal surface. The results from the aluminium crystal is shown in Table 1,

Table 1. Mean effective thickness of aluminium crystal ground by emery paper of 0.3. R: 24.5 mm. D: 5.53mm

Plane	r (mm)	s (mm)	x (mm)
$\bar{1}11$	11.50	1.89	1.3_6
$\bar{3}31$	16.58	2.76	1.3_3
$1\bar{1}1$	17.57	2.96	1.2_9
$20\bar{4}$	19.53	3.29	1.2_9
$5\bar{1}1$	13.31	2.27	1.2_3
$3\bar{1}1$	10.27	1.76	1.2_6
$\bar{5}11$	20.20	3.38	1.3_1

1.3 on the average

the mean value being 1.3 mm. The mean effective thickness of quartz obtained by Suzuki[(6)] with similar technique is 0.2 mm, which is very small compared with the above. Sakisaka and Sumoto[(5)] studied the extinction effect of quartz crystal

(6) M. Suzuki, Sci. Rep. Tohoku Univ., **35** (1951), 137.

and concluded that the thickness of the surface layer contributing to X-ray reflection was within 0.1 mm, and that the interior part of the crystal had nothing to do with it, owing to large primary extinction. In the case of aluminium, the mean effective thickness is very larger than that of quartz, as stated above. Therefore, the effect of mechanical polishing may be said to extend to the depth of some millimeter, that is, the surface layer of aluminium crystal is easily broken by grinding. Moreover, the splitted Laue spots are not perfect, as shown in Photo. 2. This may be due to the imperfect structure of aluminium.

The relative intensity was considerably increased by tensile deformation, and the smaller the indices of lattice plane, the larger the rate of increase in the relative intensity became. As shown in Figs. 1 and 2, {111}-reflected intensities are not equal to one another, that is, the increase in the relative intensity reflected from the active slip plane is the largest of all {111}-reflections. It may, therefore, be concluded that the active slip plane suffers most severely the effect of deformation. Further, the change in the relative intensity reflected from the latent slip planes is somewhat irregular compared with that from the active plane, and different from each other. Contrary to common concept, this will, therefore, imply the circumstances that all the latent planes may not necessarily be equally distorted and hardened, owing to the orientation relation to the tensile axis or to some minute difference in structural defects. If the saturation value is taken as the relative intensity reflected from the ideally mosaic crystal, and if the reduction in the reflected intensity is due only to primary extinction, then the block size of crystal will be calculated from the theory of mosaic crystal. According to the theory of Darwin, the integrated intensity from ideally imperfect crystal is given by

$$I = I_0 Q \delta V ,$$

where Q is the fraction reflected per unit volume of crystal and δV the volume of crystal irradiated. The reduction of intensity due to primary extinction is corrected by substituting $Q \tanh nq/nq$ for Q, where

$$q = \frac{e^2}{2mc^2} \frac{\lambda^2}{\sin^2\theta} \cdot N \cdot |F| ,$$

n is the number of net planes contributing to X-ray diffraction, and N the number of unit cell per unit volume. The ratio of the intensities before and after the deformation of crystal will be given by

$$m = \frac{I'}{I} = \frac{\tanh n'q/n'q}{\tanh n_0 q/n_0 q} \tag{2}$$

When the crystallite block is broken, the correction factor $\tanh n'q/n'q$ tends to 1, and accordingly,

$$m = m_0 \frac{\tanh L/d \cdot q}{L/d \cdot q} , \quad m_0 = \left(\frac{\tanh L_0 q/d}{L_0 q/d} \right)^{-1} , \tag{3}$$

where $n = Ld$, m_0 the saturation value of relative intensity as the function of the block size, L_0 the initial block size and d the spacing of the reflecting planes. From Eq. (3) the block size L_0 can be calculated; in the case of aluminium crystal, we may take $q = 1.6 \times 10^{-4}$, $d = 2.3 \times 10^{-8}$cm and $m_0 = 2.9$, which results in

$$L_0 = 4.1 \times 10^{-4}\text{cm},$$

which is consistent with the results obtained by several workers with various techniques(7).

In the case of zinc, the characteristic of the change in the reflected intensity is the same as that in aluminium, but the rate of increase of intensity is considerably small compared with the latter. This may be due to low perfectness of zinc crystal used in the present experiment.

In the case of aluminium single crystal the reflected intensity was not recovered even after long-time anealing at high temperature. In the case of zinc the reflected intensity was recovered almost perfectly after short-time annealing. This is probably due to the simple type of deformation of zinc; on the other hand, the slip deformation of aluminium is multiple and the residual strain is present even after long-time annealing at high temperatures, as may be inferred from sessile dislocation.

Summary

(1) The relative intensities of X-ray reflected from aluminium and zinc single crystals subjected to tensile deformation were measured by microphotometer, and the recovery of the intensity by annealing was also examined.
(2) In the case of aluminium crystal, the increasing rate of the X-ray intensity reflected from active slip plane with increasing defomation was the largest of all {111}-reflections; in other words, the active slip plane is more distorted than the latent slip planes.
(3) In the case of zinc crystal, the characteristic tendency was qualitatively the same as in the case of aluminium, but quantitatively, the change was considerably small.
(4) The cange in the X-ray intensity reflected from zinc crystal almost perfectly recovered after short-time annealing at 210°C.
(5) In the case of aluminium, the complete recovery was not obtained even after long-time annealing at high temperature.

The present investigation was supported partly by the Grant in Aid of the Fundamental Scientific Research of the Ministry of Education.

(7) P. B. Hirsch, *Progress in Metal Physics*, **6** (1956), 236.