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著者	NAKAJIMA Koichi, SUTOKI Tomiya
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Sub-Boundaries in Aluminium Single Crystals*

Kôichi NAKAJIMA and Tomiya SUTOKI
The Research Institute for Iron, Steel and Other Metals

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Synopsis

Single crystals of aluminium 99.97 per cent pure were made by the Bridgman method under various conditions, and the Laue spots were precisely studied with specimens which, on being etched, showed no macroscopic inhomogeneity. It was found that some spots were frequently splitted, whereas those diffracted from certain lattice planes were of the usual form, and that these special planes were {113} or planes lying close to them. From these results it was concluded that the splitting of Laue spots was due to a small angle interface of a bicrystal grown along the maximum temperature gradient, and that this boundary could be formed by the relative rotation of the crystals around $[1\bar{1}2]$ about one degree. Such boundary was explained by the mechanism of polygon wall in the the theory of dislocation.

I. Introduction

Aluminium single crystals frequently show splitted Laue patterns, whatever method may be utitized in their preparation. From his observation that an aluminium single crystal containing some lineage structures showed a splitted Laue pattern, Lacombe(1) explained the splitting as being caused by the lineage structure happening to form during the growth of crystal. But a macroscopically homogeneous region of the crystal frequetly shows a splitted Laue pattern, and sometimes contains abnormalities extending almost along the maximum temperature gradient, namely, the direction of specimen axis. Usually, the angular extent of such a Laue spot is of the order of one degree.

Kabata(2) observed sub-structures in aluminium single crystals made by the strain-annealing method, and explained them by the rotation about 20∼30′ around <112>. Recently, some imperfections were microscopically observed also by Lacombe(3) in aluminium single crystals made by the strain-annealing method.

The small angle crystal boundaries extending along the direction of specimen axis are observable in the case of the Bridgman method, but not in the case of the strain-annealing method. This will be due to the following reason. In general, the circumstances under which crystal grains grow are not the same in recrystallization and in solidification, that is, in the former the principal factor controlling the growth is a local inhomogeneity of stress field, whereas in the latter it is a temperature gradient. Hence, it was desirable to investigate the splitting of Laue

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* The 805th report of the Research Institute for Iron, Steel and Other Metals.
This paper was read on October 31, 1954 at the Autumn Meeting of the Physical Society of Japan held at Ôsaka.

(1) P. Lacombe, Mêtaux et Corrosion, **26** (1951), 392.

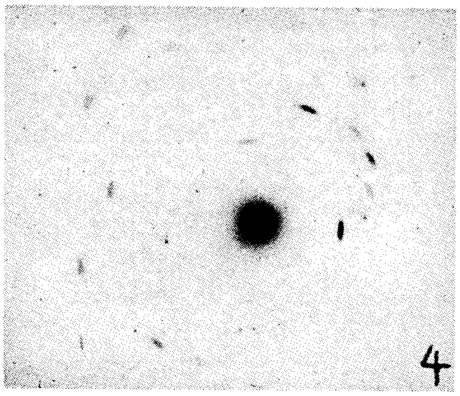
(2) M. Kabata, Memoirs of the Colleage of Science. Kyoto Imp. Univ., **A16** (1936), 223.

(3) P. Lacombe et L. Beaujard, Rev. Mêtal., **45** (1948), 317.

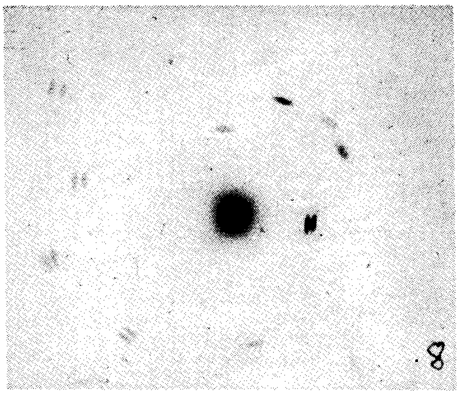
spots from the crystallographical point of view, apart from the lineage structure, and to clarify the effects of these controlling factors.

II. Method of experiments

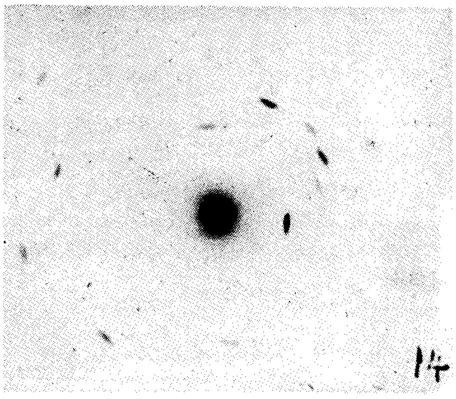
Single crystals of aluminium 99.97 per cent pure were made by the Bridgman method under various conditions, the size of specimen being $10\times1\times(70\sim100)\mathrm{mm}^3$. For the present purpose a Laue camera with a special specimen holder was devised, with which not only the irradiation of specimen could be carried out in any direction, but also the specimen could smoothly be displaced parallel to itself in the direction parpendicular to the primary beam. In this way, it was possible to detect the direction and shape of the small angle grain boundary. The distance from the directly irradiated point to each Laue spot was measured with a comparator, and the angular extent of the splitted spots was measured with a photometer. A copper target was used at 40 kV and 3 mA, the distance between specimen and photographic film being 38 mm. The specimens were all polished electrolytically but not mechanically. A slit, 0.8 mm in diameter, was used, and Laue photographs were taken at every 0.5 mm over the width of the specimen. For example, the patterns of the specimen M_1 are shown in Photos. 1, 2 and 3, in which the direction of the primary beam was exactly perpendicular to the surface of the specimen. Photo. 1 was taken at the point 2 mm from one edge of the specimen. Photo. 2



Phot. 1



Phot. 2



Phot. 3

Laue photographs of the specimen M_1

was taken in the region in which the interface of a bicrystal lay; the irradiated volume of each part of the bicrystal might be estimated from the width of the splitted spot. Photo. 3 was taken at the point displaced successively by 5 mm. It may be remarked that the superposition of Photos. 1 and 3 will result in the same as Photo. 2. Indices of lattice planes were determined by the method by Schiebold and Sachs.

III. Results of experiments

The region showing the splitted Laue pattern extended almost in the direction of the specimen axis, and the amount of splitting was different with different spots, there being some spots showing no splitting. Some lattice planes corresponding to unsplitted spots are shown in Table 1. The angular extents of some splitted spots in the spcimhn M_3, for example, are shown in Table 2. Of course, there were spots whose intensities were so weak that the angular extent of splitting could scarcely be determined.

Table 1. Lattice plane corresponding to unsplitted spots.

Specimen	Cooling rate	Plane of unsplitted spots	Angle between the normal of the plane and the specimen axis
M_1	0.1 cm/hour	$(1\bar{1}3)$ $(01\bar{2})$ $(1\bar{1}5)$	13° 14° 32°
M_2	"	$(1\bar{1}3)$ $(1\bar{1}5)$	14° 30°
M_3	5 cm/hour	$(\bar{2}11)$ $(\bar{3}13)$	17° 17°
M_4	"	$(1\bar{1}3)$ $(1\bar{1}2)$ $(1\bar{1}5)$	24° 18° 31°
M_5	"	$(1\bar{1}3)$ $(\bar{1}20)$ $(\bar{1}31)$	66° 57° 76°
M_6	"	(102) (115)	28° 18°
M_7	"	$(2\bar{1}2)$ (102)	16° 25°
M_8	"	$(\bar{3}11)$ $(2\bar{1}0)$	32° 22°

From Tables 1 and 2 it will be seen that the angular extent of splitting is different with different lattice planes, and that the lattice planes corresponding to unsplitted spots are not arbitrary ones but {113} or those lying close to them. Further, unsplitted spots were situated almost in the vicinity of the specimen axis

Table 2. Comparison of observed value $\Delta\theta$ and calculated value $\Delta\alpha$ in the specimen M_3.

No. of Laue spot i	Net plane	Observed value $\Delta\theta$	Calculated value		Difference	
			$\Delta\alpha_{1\bar{1}2}$	$\Delta\alpha_{1\bar{1}3}$	$\|\Delta\alpha_{1\bar{1}2}\|-\Delta\theta$	$\|\Delta\alpha_{1\bar{1}3}\|-\Delta\theta$
1	$01\bar{1}$	0	3′	−7′	3′	−7′
2	$11\bar{3}$	29′	33′	11′	4′	−18′
3	$11\bar{2}$	30′	32′	23′	2′	−7′
4	$10\bar{1}$	40′	52′	47′	12′	7′
5	$2\bar{1}\bar{1}$	35′	57′	43′	22′	8′
6	$2\bar{1}0$	35′	49′	54′	6′	19′
7	$5\bar{3}1$	37′	43′	49′	6′	12′
8	$1\bar{1}1$	?	19′	33′		
9	$1\bar{1}2$	0	0	11′		
10	$1\bar{1}3$	0	−10′	0		
11	$1\bar{1}5$	?	−20′	−10′		
12	001	28′	−25′	−26′	−3′	−2′
13	$\bar{1}13$	37′	−52′	−45′	−15′	8′
14	$\bar{1}11$	58′	−58′	−57′	0	−1′
15	$\bar{1}20$	40′	−44′	−50′	4′	10′
16	$\bar{1}3\bar{1}$	37′	−28′	−52′	−11′	15′

and the normals of corresponding lattice planes were, on the whole, parallel to the specimen axis.

When the crystal was prepared at a relatively high rate of solidification, the splitting was generally large and irregular and, therefore, lay quite outside the present purpose.

IV. Discussion

The present experiments showed clearly that one of {113} was almost the interface of a bicrystal, that is, a slight misorientation between the adjacent crystals was due to their small relative rotation around one of <113>.

Suppose that a block of single crystal is cut into two parts, and rotate one part relative to the other by a small angle φ around a certain crystallographic axis. Then, the angular displacement of any plane of the same indices in the two parts of the crystal can be calculated. In Fig. 1, let OP be the direction of the incident beam and ON be the normal of any crystal plane, which will come into the position ON′ by the rotation of a small angle φ around OZ, a certain crystallographic axis $[hkl]$ in question. Denote the angles which OP makes with ON and ON′ by α_{Ai} and α_{Bi}, respectively, then the angular displacement $\Delta\alpha_{hkl}$ of the crystal plane with respect

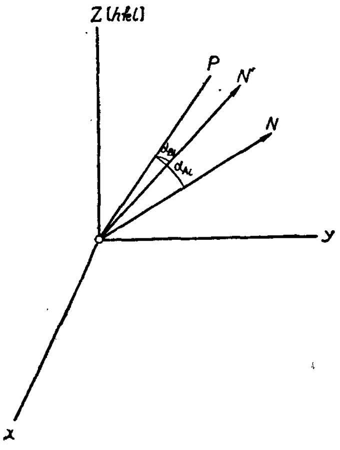


Fig. 1.

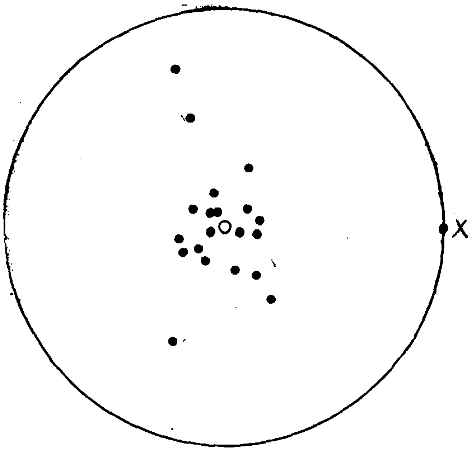
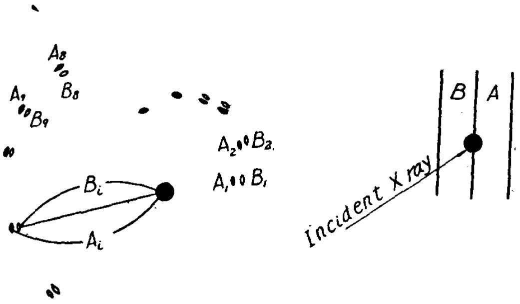


Fig. 2. Stereographic projection of specimen axis and unsplitted spots. The center of the circle represents the specimen axis and X the direction of incident beam.



(a) (b)

Fig. 3. Splitting of Laue spots of the specimen M_1.
A_1, A_2 : the Laue spots from the crystal A
B_1, B_2 : the Laue spots from the crystal B
Ai and Bi are the planes of the same indices in the respective crystals.

to OP will be

$$\Delta\alpha_{hkl} = \alpha_{Ai} - \alpha_{Bi}$$

The correlation of the axis of specimen and the lattice plane giving unsplitted spot can be seen by the aid of a stereographic projection as shown in Fig. 2, in which X is the direction of incident beam. It is clear that the poles of these planes are close to the specimen axis, that is, each part of a bicrystal is relatively rotated by a very small angle around a certain crystallographic axis lying close to the specimen axis. The axis of rotation, however, could not be determined exactly only by the above procedure, and hence, $\Delta\alpha$ was tentatively calculated on <112>, <113> and <115>, all lying close to the specimen axis, and was compared with the observed value $\Delta\theta$. For example, the results of the specimen M_3 are shown in Table 2, in which φ was put at 1°. The case of <115> was omitted because of relatively large scattering. As shown in Table 2, in some lattice planes $\Delta\alpha$ was negative, which is of an important significance in deciding whether the present results can be interpretated by such a rotation or not. In the splitted Laue pattern shown in Fig. 3, for example, let r_{Ai} and r_{Bi} be the distances of the splitted parts of a spot from the center of the photograph, and put $r_{Ai}=r_{Bi}+\Delta r$; of course, the two parts correspond respectively to the planes of the same indices in the adjacent crystals A and B. In case the present results can be interpretated by the above-stated rotation, the signs of $\Delta\alpha$ and Δr must coincide with each other. In general, it was not possible, in the splitted spots, to distinguish the Laue pattern of crystal A from that of crystal B, but as explained in Photos. 1, 2 and 3, r_{Ai} and r_{Bi} could be obtained respectively from the Laue patterns of crystals A and B. Thus, the sign of Δr was examined in each case of <112>, <113> and <115>. In every specimen, the above-mentioned relation held good both in $[1\bar{1}2]$ and $[1\bar{1}3]$, but not in $[1\bar{1}5]$. Consequently, it might be said that the crystals were relatively rotated around either $[1\bar{1}2]$ or $[1\bar{1}3]$ by about 1°. By comparing the corresponding values of $(|\Delta\alpha_{1\bar{1}2}|-\Delta\theta)$ and $(|\Delta\alpha_{1\bar{1}3}|-\Delta\theta)$ with each other, it will be seen that the former is, on the whole, less than the latter. As φ is small and the angle between $[1\bar{1}2]$ and $[1\bar{1}3]$

is only about 10°, the superiority of the former over the latter cannot be seen only from this comparison, and hence, will be considered from another point of view.

With the decreasing rate of solidification, the splitting became inconspicuous and in the case of a very slow rate of cooling, for example, of 0.1 cm/hr, the observed crystal boundaries were almost parallel to the axis of specimen. This will be natural from the point of view of surface energy and in such a case the boundary may be assumed to be a plane. For, as stated already, the Laue spots of the specified region extended almost along the direction of maximum temperature gradient and were splitted, and the widths of the two parts of a splitted spot linearly changed, one decreasing while the other increasing, as the irradiation travelled from one crystal to the other over the boundary region. Therefore, the boundary might be assumed to be a single plane. The boundary planes in 7 kinds of specimens were examined by the aid of the stereographic projection. As shown in Fig. 4, the boundary surfaces lay within the limit of about 15° from (110). Therefore, if the boundary plane is (110) these results will be explained consistently by the dislocation model, according to which, a simple grain boundary in well-annealed state is accommodated by a row of dislocations of the edge type. In other words, as $[1\bar{1}2]$ lies in (110), the most probable axis of rotation in question may be said to be $[1\bar{1}2]$.

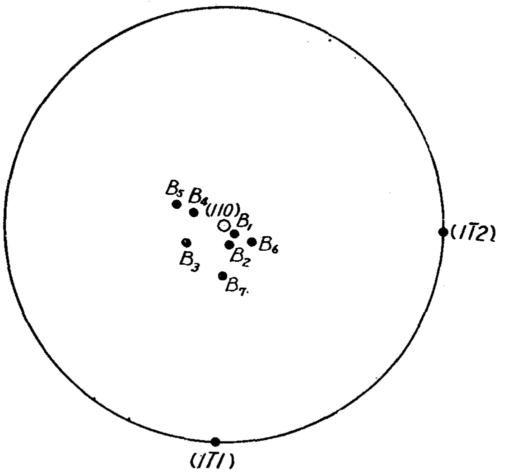


Fig. 4. Stereographic projection of the normals of boundary planes.

In the case of a quick rate of solidification, the difference between the orientations of adjacent crystals seemed to become relatively large. Some experiments were carried out with such specimens, but no definite result was obtained, due probably to complicated structures of the boundary. The nature of such a boundary, however, may not differ essentially from that of a small angle grain boundary. In fact, Crussard(4) concluded statistically that many nuclei growing in a deformed aluminium single crystal were rotated by 30~70° around <112> relative to the mother crystal. Further, Beck(5) showed that among many unclei in a deformed grain, those rotated around <111> by 30~40° relative to their neighbours grew most rapidly. When precisely inspected, the observed points were scattered comparatively widely, extending beyond the region of <112> and, accordingly, it might be said that Beck's conclusion was not necessarily correct, and that the rotation around <112> frequently took place in his experiment. It follows, therefore, that the relative rotation of adjacent crystals around <112> must be inherent in a crystal growth, irrespective of the amount of the misorientation between them.

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(4) Mlle Alalceuf et C.H.Crussard, Rev. Mêtal, **48** (1951), 415.

(5) P. A. Beck, J. App. Phys., **21** (1950), 520.

Of course, in many actual cases, this fundamental characteristic may variously be modified by different circumstances, resulting in apparently complicated phenomena.

Summary

(1) Splitting of Laue spots in aluminium single crystals made by the Bridgman method was studied.

(2) The interface between a bicrystal extended almost along the direction of temperature gradient, namely, the axis of specimen.

(3) Two crystals forming a bicrystal were relatively rotated by a small angle of the order of 1° around $[\bar{1}12]$, which lay close to the axis of specimen.

(4) In the case of a very slow rate of solidification the small angle grain boundary grew approximately parallel to (110).

Acknowledgement

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