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An X-ray Diffraction Method for the Study of Substructure of Crystals

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A double-crystal diffractometer method using a Geiger counter and supplementary film technique has been described. The analysis of the multi-peaked rocking curves obtained from bent, coarse-grained aluminum established a correlation between intensity maxima and adjacent lattice regions giving rise to them. The tilt between adjacent subgrains was determined and the existence of substructural domains within the subgrain was shown.

1. Introduction

Various workers studying the subgrain structure of single-crystal and polycrystalline aluminum as well as other metals have obtained data concerning the subgrain size which vary greatly in order of magnitude (Tate & McLean, 1951–2; Perryman, 1954; Hunter & Robinson, 1953; Delisle, 1953; Hirsch & Kellar, 1952). Refined microscopic and X-ray techniques have revealed substructural entities which are considerably smaller than those disclosed by more conventional techniques. The possibility therefore arises that under certain conditions of specimen preparation various orders of magnitude of substructural entities may coexist. The possible coexistence of such different substructural entities is investigated in coarse-grained bent aluminum specimens by means of an X-ray diffraction technique.

2. Experimental method

(*a*) *Preparation of specimens*

The aluminum specimens studied were furnished through the courtesy of Mr M. Metzger of Columbia University. The specimens were of 99·993% purity, containing 0·004% Fe and 0·002% Cu. The coarse grains were obtained by the strain-anneal method. Specimen Al–1 was bent to a radius of curvature of 20·5 cm. and subsequently heated at 600° C. for 18 hr. Specimen Al–2 was bent to a radius of curvature of 2 cm. and recrystallized upon annealing at 645° C. for 21 hr. and etched with 7% HCl solution containing some copper. This solution has been shown to attack subgrain boundaries strongly (Metzger & Intrater, 1954).

(*b*) *X-ray method*

As the principal research tool in this investigation a modified X-ray double-crystal diffractometer shown in Fig. 1 was employed. The X-ray beam emerging from a copper-target tube is reflected from the cleaved surface of a calcite crystal mounted on an adjustable holder (A) next to the beam port. The rocking curve of the cleaved calcite crystal exhibited a width at half maximum intensity of 8 sec. of arc. In order to remove the $K\alpha_2$ component from the reflected radiation, the beam passes through a 41 cm. long collimator (B) bolted to the sturdy base plate (C).

The collimator is provided with adjustable slit systems (D) at both ends. By proper adjustment of the vertical exit slit the $K\alpha_2$ component of the emerging beam was eliminated. The specimen is mounted on a goniometer (E), the axis of rotation being coincident with the crystal surface. The goniometer is equipped with a micrometer screw (F) which permits vertical scanning of the specimen surface. The specimen is rotated by a worm gear (G) which is connected to a synchronous 2 r.p.m. reversible motor (H) by means of interchangeable reduction gears (I) providing for a variety of speeds of specimen rotation. Attached to the reduction-gear system is a revolution counter (J) which enables one to relocate readily the angular position of the specimen. The reflection intensities of the specimen are detected by a Geiger counter (K) mounted on a track (L) which can be rotated independently around the specimen axis. Provisions are made for tilting the Geiger counter to register non-equatorial reflections. Attached to the front of the Geiger-counter window is a frame with inserted film holder (M). By means of the screw (N) small film

shifts can be made between each angular position of the specimen, thereby permitting one to obtain photographic details of the beam from positions corresponding to selected ordinates of the reflection curve.

3. Experimental results and discussion

(a) Multi-peaked rocking curves and analysis of intensity distribution

A portion of the rocking curve recorded for the (200) reflection of specimen Al−1 is shown in Fig. 3(*a*). This curve was obtained by slow specimen rotation and detection of the reflected beam by the stationary Geiger counter set at the proper Bragg angle. The irradiated area of the specimen was 1·6 mm. long and 0·22 mm. wide, and remained essentially constant over the small range of specimen rotation. The counter window was kept open so that every portion of the reflected beam could be registered.

The most salient features of the rocking curve are the multiple peaks of the intensity distribution. Similar multipeaked rocking curves have been observed for bent zinc single crystals (Conard, Averbach & Cohen, 1953).

The statistical fluctuations could be distinguished from the true crystallographic effects by running the rocking curve several times. The intensity maxima are about 8 min. of arc apart. Although the individual component curves are not fully separated they can nevertheless be analyzed graphically (Weissmann & Evans, 1954). The rocking-curve widths of the component curves were estimated to be 5–7 min. of arc.

A photographic analysis of the total distribution curve obtained by the Geiger-counter technique was made for the (200) reflection for specimen Al−1. The rocking curve of Fig. 3(*a*) was retraced in discrete, 1 min. intervals and the reflected beam was recorded on a film which was shifted between each specimen setting. This 'multiple exposure technique' has been described previously (Reis, Slade & Weissmann, 1951) and has been used to advantage in the study of silicon powder and silicon ferrite (Slade & Weissmann, 1952) and fine-grained aluminum (Weissmann & Evans, 1954).

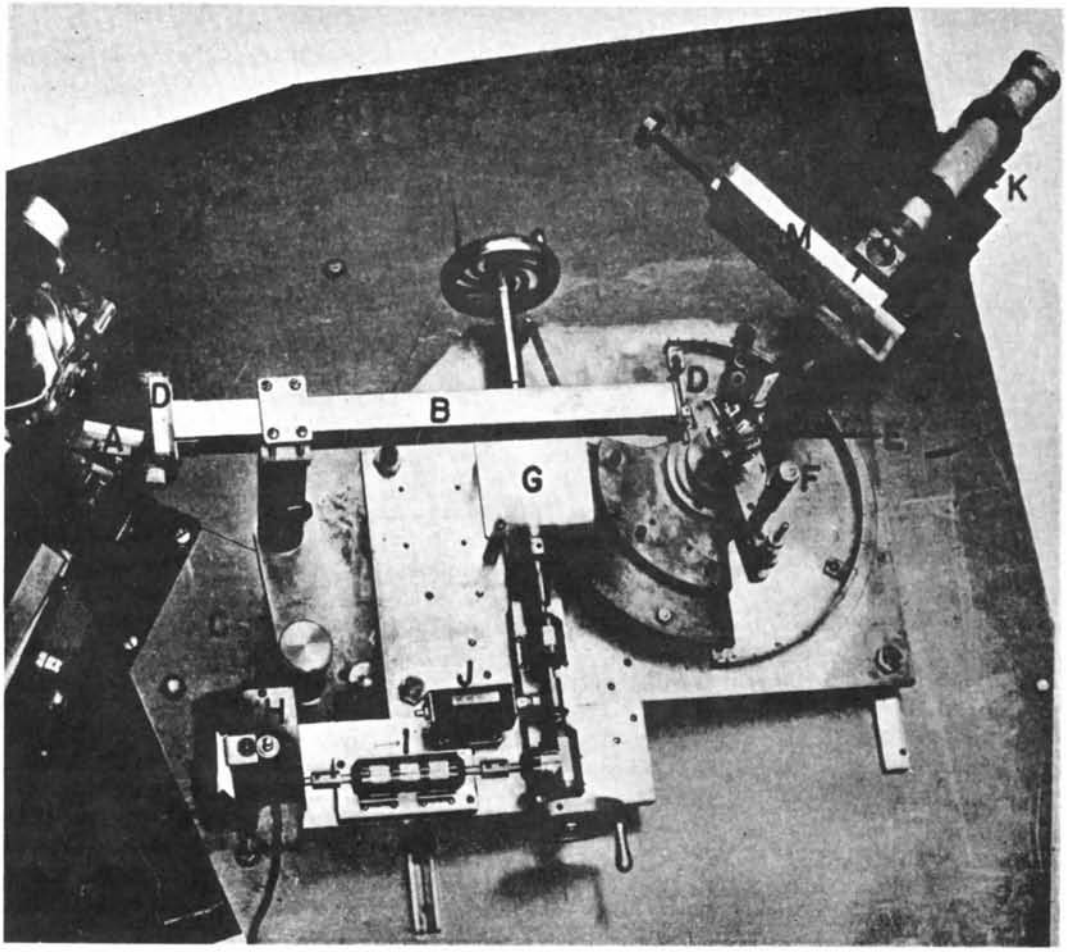
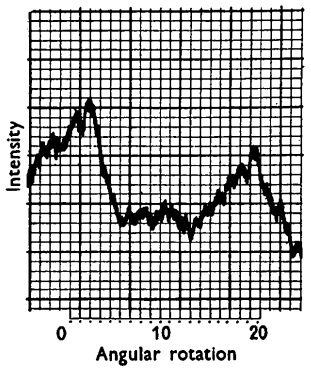
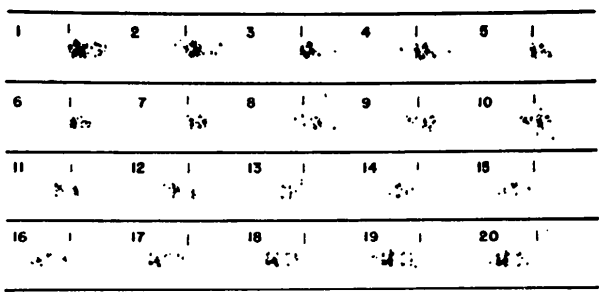


Fig. 1. Modified X-ray double crystal diffractometer.



(a)



(b)

Fig. 3. (a) Portion of rocking curve of Al−1 (200) reflection. 1 division of angular rotation corresponds to 1 min. of arc. The series of photographic spots was obtained by retracing the Geiger counter curve at 1 min. intervals.
(b) Same series of photographic images as shown in (a) (10×).

The photographic record obtained for Al−1 appears in Fig. 3(a) and (b) magnified $\frac{3}{4}$ times and 10 times, respectively. The reflection image obtained by replacing the aluminum specimen by a cleaved calcite crystal of optical quality showed a single spot of the shape of the moon a day or two beyond the first quarter, without any split-off intensity.

Each image in Fig. 3(b) consists of many spots, and it is possible to indentify the same spot in successive photographs by means of its fixed position with respect to the reference mark. These spots are found to persist through a rotation of 3–4 min. of arc. The width of the rocking curve of these spots is estimated to be of the order of 2 min. of arc.

Comparison of Fig. 3(b) with Fig. 3(a) reveals that the most dense clustering of spots 1, 10, 19 coincides with the maxima and the least dense with the minima of the intensity distribution curve. This variation in the density of spots as a function of specimen position indicates that the small substructural entities, which give rise to the individual spots, show a preferred grouping about a mean orientation. This mean orientation corresponds to a rocking curve maximum, whereas the spread of each rocking curve component is determined by the angular orientation range of the small individual substructural entities and the spread of the clusters into which these entities group. It follows that each component of the multipeaked rocking curve has an angular width W, at half maximum intensity that requires two parameters for definition: $W^2 = w_1^2+w_2^2$, where w_1 is the half-width of the distribution of substructural domains which cluster about a mean orientation. w_1 is therefore a measure of the outer or group misalignment. w_2, the half-width of the reflections, arising from the substructural entities, is a measure of the average inner or individual misalignment.

For specimen Al−1, W and w_2 are measured to be 6 and 2 min. respectively, and $w_1 = \sqrt{(W^2-w_2^2)} \approx 5{\cdot}6$ min. of arc.

(b) Correlation between intensity maxima and lattice regions

In order to establish the lattice regions in the specimen which give rise to the corresponding intensity maxima of a rocking curve one may consider the two simple cases of Fig. 4(a) and (b), which demonstrate possible orientation relationship of three neighboring regions.

The regions shown are simultaneously irradiated with a monochromatized, parallelized beam and all the areas satisfying the Bragg condition will reflect.

A reference position is established in the middle of region B and appears in the middle of the image of

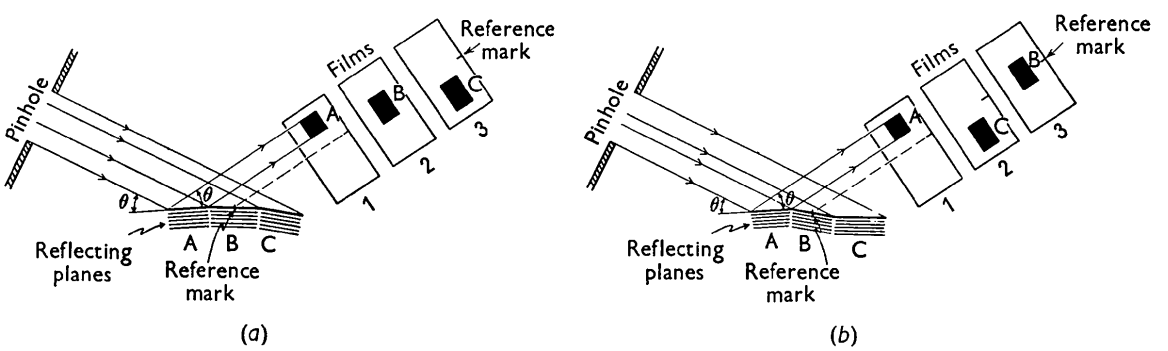


Fig. 4. Schematic representation showing the correlation between lattice orientation and sequence of reflection images.

that region when it is in its optimum reflecting position.

At a definite angular setting region A will be in reflecting position and the corresponding image will appear on film 1 to the left of the reference mark. By rotating the specimen counter-clockwise regions B and then C will be recorded on film 2 and 3 successively (Fig. 4(*a*)).

In Fig. 4(*b*), where region C is closer in orientation to A than it is to B, the sequence of images will be in the order A (film 1), C (film 2), B (film 3) and their positions on the film with respect to the reference mark remains unaltered. It will be noted that in Fig. 3(*b*) the sequence of spot clusters is consecutive and hence establishes that neighboring intensity maxima derive from adjacent regions of the specimen.

The metallographic examination of the etched specimen Al–2 revealed the presence of subgrains (Fig. 2(*a*)) of varying size ranging from 0·5 to 3 mm. The area irradiated by the X-ray beam was located by means of a film superimposed on the specimen surface and was determined to be 3 mm. long and 0·1 mm. wide. The Geiger counter rocking curve for the (200) reflection obtained for that specimen is shown in Fig. 2(*b*). The angular separation between intensity maxima A′ and B′ was measured to be 5·5 and between B′ and C′ 17·5 min. of arc. Decomposing the three-peaked distribution curve into three corresponding component curves and measuring their width at half-maximum intensity one obtains values of about 3 min. for the width associated with A′ and B′ respectively, and 4 min. for that associated with C′.

Since the intensity maxima A′, B′ and C′ could be related to metallographically revealed subgrains A, B and C respectively the angular tilt of adjacent subgrains was determined and found to be 5·5 min. between subgrains A and B and 17·5 min. between B and C.

Images shown in Fig. 2(*c*) were obtained by recording photographically reflections from subgrain A at angular positions corresponding to abscissas near A′, 1 min. apart. Those images are broken up into fine spots which derive from substructural entities of subgrain A. Similar images were obtained for subgrains B and C. These observations indicate that for the aluminum specimen investigated two orders of magnitude of substructural entities (subgrains and substructural domains within the subgrains) coexist.

4. Conclusions

The analysis of the multipeaked rocking curve carried out with the aid of the supplementary film technique disclosed the relationship of neighboring intensity maxima with respect to the lattice regions giving rise to them. In the case where the different lattice regions can be identified as subgrains the angular separation of intensity maxima reveals the subgrain tilt. It has been shown that the subgrains of the specimen investigated consist of substructural domains which in turn exhibit an inner angular misalignment as evidenced by their individual rocking curves. The substructural domains cluster preferentially around a mean orientation which can be correlated with an intensity maximum of the multipeaked rocking curve.

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