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X-Ray Diffraction Measurement of Intragranular Misorientation in Alpha Brass Subjected to Reversed Plastic Strain

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A monochromatic misorientation goniometer was built and used to examine the character of X-ray diffraction spots from 70 percent copper, 30 percent zinc alpha (cartridge) brass specimens subjected to unidirectional and reversed plastic deformation. A two-exposure, moving and stationary film technique was employed. Numerical measurements of the average subgranular misorientation, amounting to several hundredths of a degree, were obtained at different stages in one complete cycle of 1 percent plastic strain amplitude. About 60 percent of the misorientation was recovered when the plastic strain was reversed, leaving a residual misorientation at zero net strain that increased linearly with the cumulative strain.

1. Introduction

Since an X-ray diffraction technique can provide information relating to the structure of metals on the atomic lattice scale, there is a continuing search for applications of this kind of tool in the study of structure-sensitive properties. The usefulness of the techniques has often been lost because of the difficulty of separating the numerous factors that can affect the diffraction of X-rays from crystals. The purpose of the investigation described in this paper was to devise a relatively simple method for measuring a single one of these factors—misorientation—and observe how it varied during reversed plastic deformation of a ductile polycrystalline metal. By misorientation in a grain, or disorientation as it is sometimes called, is meant the angle through which the atomic lattice planes are elastically or plastically bent or fragmented.

The distortion in Laue patterns of X-ray diffraction spots, commonly called "asterism," from single crystals subjected to shear strain has long been observed [1] [1]. Such spot blurring is a qualitative indication of lattice distortion. In 1925, J. Czochralski [2] reported that the asterism in a Laue pattern of a twisted aluminum crystal was decreased when the specimen was twisted backwards. Similar qualitative experiments were made in the present study. Both transmission and back-reflection Laue patterns were made from a single grain in a very large-grained aluminum specimen that was subject to forward and reversed bending. The set of transmission Laue patterns may be seen in figure 1. The greatest recovery of sharpness of the spots came not at the recovered zero bending position but at the negative bending position past zero; this interesting behavior remains to be explained. In recent years a more sophisticated Laue pattern technique, the focused spot method, has been employed by C. A. Julien and B. D. Cullity [3] to show the partial recovery of Laue spots after reversed twist or bending in aluminum crystals.

For studies of polycrystalline metals, much work has been done with the phenomenon of the broadening of characteristic X-radiation diffraction rings or lines in Debye-Scherrer patterns [4]. If the material is not so fine-grained as to give continuous lines in the pattern, a large number of discrete spots may be observed in the line position. Each spot is a diffraction from a known set of atomic lattice planes in an individual crystallite, and the blurring of such spots in cold-worked polycrystalline metals may yield information of interest. A study reported in 1940 by W. A. Wood and P. L. Thorpe [5] on the behavior of the crystalline structure of brass under slow and rapid cyclic stresses showed no recovery of tangential spot blurring on the Debye-Scherrer diffraction rings upon reversal of the static strains of the specimens. In 1953, however, W. A. Wood and R. B. Davies [6] showed that if the strains were small enough, about 1 percent in copper, there was a partial recovery in the spot blurring when the strain was reversed. In the present study, similar behavior was observed in the diffraction patterns from aluminum, copper, and steel rods subjected to torsional straining. The set of patterns from the copper rods at various stages in one cycle of strain may be seen in figure 2.

In 1951, A. J. Reiss, J. J. Slade, and S. Weissmann [7] described a method for studying the breadth of the reflecting range of individual crystallites in a polycrystalline specimen. Using a specimen with grains coarse enough that a spotty Debye-Scherrer pattern was obtained, they recorded the stationary

[1] Figures in brackets indicate the literature references at the end of this paper.

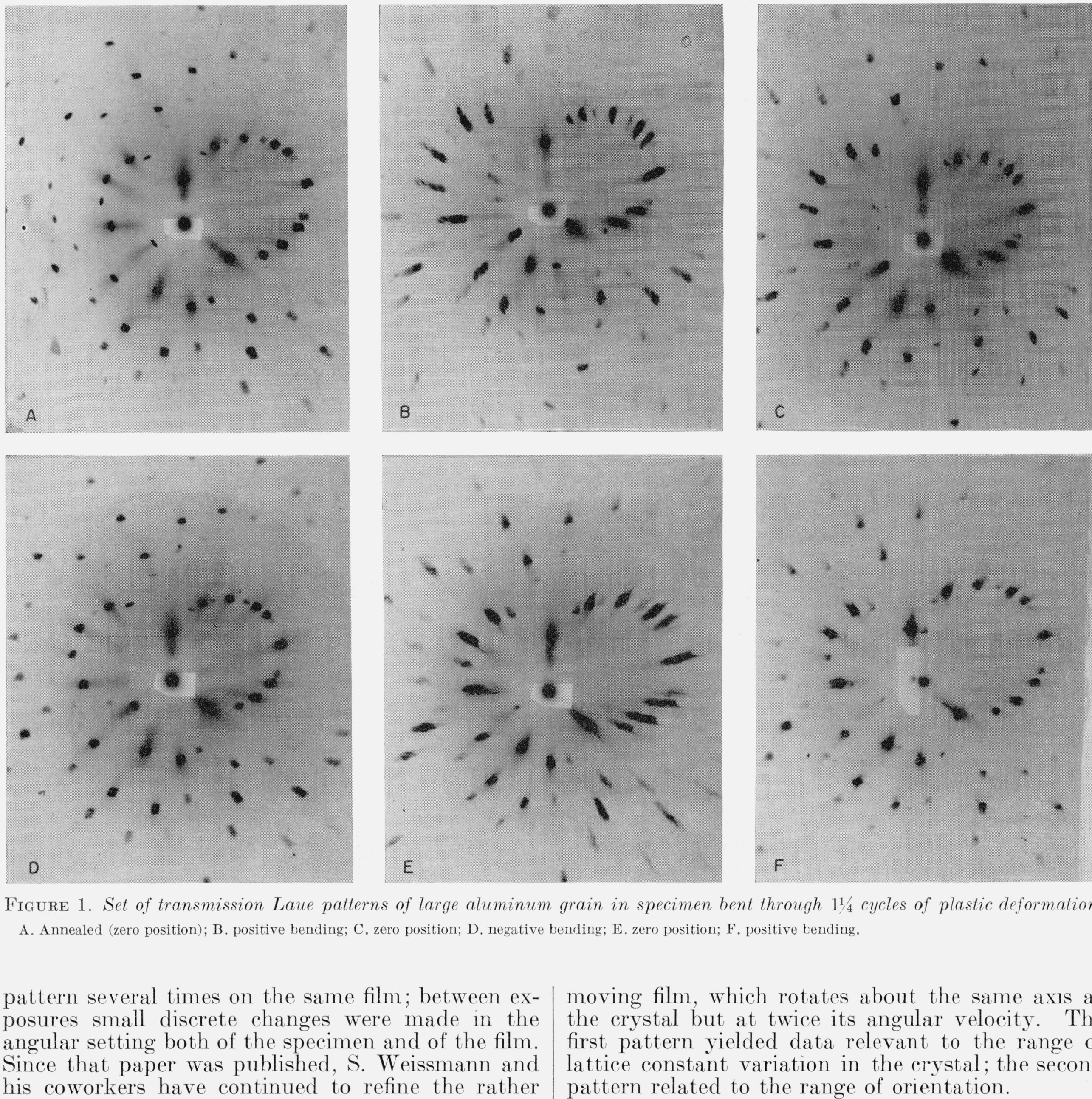

Figure 1. *Set of transmission Laue patterns of large aluminum grain in specimen bent through $1\frac{1}{4}$ cycles of plastic deformation.* A. Annealed (zero position); B. positive bending; C. zero position; D. negative bending; E. zero position; F. positive bending.

pattern several times on the same film; between exposures small discrete changes were made in the angular setting both of the specimen and of the film. Since that paper was published, S. Weissmann and his coworkers have continued to refine the rather complex instrumentation and technique to contribute many studies based upon it [8], which were not concerned directly, however, with the effects of reversed plastic strain.

C. S. Barrett [9] has described a scheme for rotating a film and off-center specimen together, "permitting the spread of a Laue spot in all directions without altering the wavelength of the reflected beam." In 1954, E. Kappler and R. Mock [10] described a somewhat similar method for obtaining the range of reflection from a single crystal. Two diffraction patterns were called for in their procedures. First, the single crystal specimen was rotated and the diffraction pattern was recorded on a stationary film; and second, the diffraction pattern was recorded on a moving film, which rotates about the same axis as the crystal but at twice its angular velocity. The first pattern yielded data relevant to the range of lattice constant variation in the crystal; the second pattern related to the range of orientation.

In recent years there have been several studies related to the problem of fatigue of metals, in which essentially ordinary Debye-Scherrer X-ray patterns have been made of materials subjected to cyclic stresses. In 1957, a study was made by L. M. Clarebrough, M. E. Hargraves, G. W. West, and A. K. Head [11] on polycrystalline copper that had been reduced 25 percent by rolling and then fatigued to fracture. High-angle diffraction patterns were made during the course of fatigue. At first the patterns showed very diffuse lines; but, as the fatiguing process continued, the lines grew sharper until the α_1–α_2 doublet was clearly resolved. This phenomenon, sometimes called "work softening," has been reported by many workers, including T. Brown and R. K.

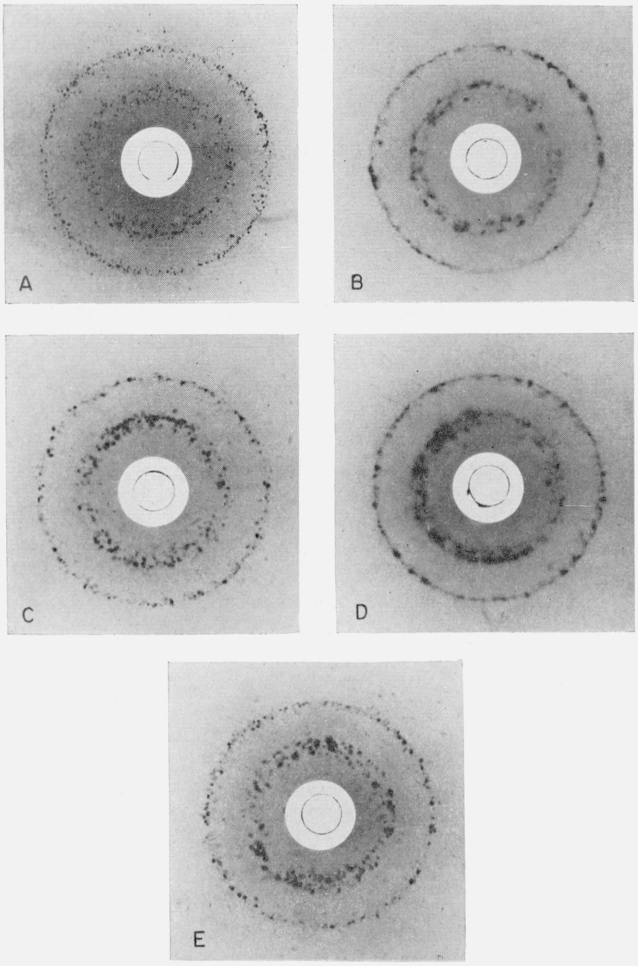

FIGURE 2. *High-angle X-ray diffraction patterns from copper rod at stages in one cycle of torsion.*

A. Annealed, zero twist; B. twisted clockwise 45°; C. twisted counter-clockwise to zero position; D. twisted counter-clockwise to −45°; E. twisted clockwise to zero position.

Ham [12], W. A. Wood and R. L. Segall [13], and M. L. Ebner and W. A. Backofen [14]. These studies are among those that emphasize the need for quantitative study of individual X-ray diffraction spots as affected by reversed plastic deformation.

Although the instrument and related procedure developed by Weissmann [8] and colleagues give the most complete information in this type of investigation, it was thought that a less complex method would suffice for the quantitative determination of misorientation. The interesting effects of reversed deformation do not appear to have been studied adequately in a quantitative manner, so the emphasis in this investigation was on measurements of misorientation at different stages of plastic straining, both forward and reversed.

2. Quantitative Measurement of Misorientation

In order to obtain a measure of the angle of misorientation, that is, the range of orientation in the distorted crystallite over which diffraction may occur, a special instrument, based upon the ideas of a double-crystal spectrometer, as may be seen in the diagram in figure 3, was designed and built. Although the procedure developed was for the measurement of the intragranular misorientation in coarse-grained polycrystalline specimens, it could be modified for work with single crystals. Shown in the photograph, figure 4, the instrument, which might be called a monochromatic X-ray misorientation goniometer, was built around three essential parts: (1) A manually adjustable mount for a stationary first crystal, M, ordinarily a strongly diffracting, monochromating single crystal; (2) a motor-driven oscillating mount for a second crystal, ordinarily the polycrystalline specimen, S, under study; and (3) a film holder circle with its center coincident with the axis of oscillation of the second crystal. The holder, F, might remain stationary or might oscillate synchronously with the specimen at the same angular velocity. Interchangeable film holders were provided, one for a semicylindrical strip film, the other for a small flat film. Most of the work was performed with the latter, using dental film because of its single-layer emulsion and resolving power higher than that of the usual X-ray film.

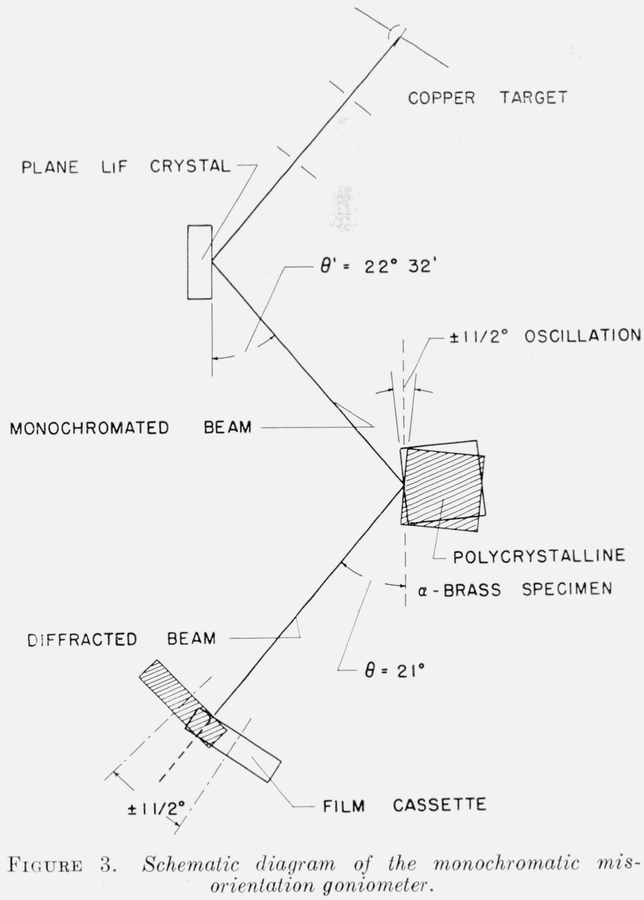

FIGURE 3. *Schematic diagram of the monochromatic misorientation goniometer.*

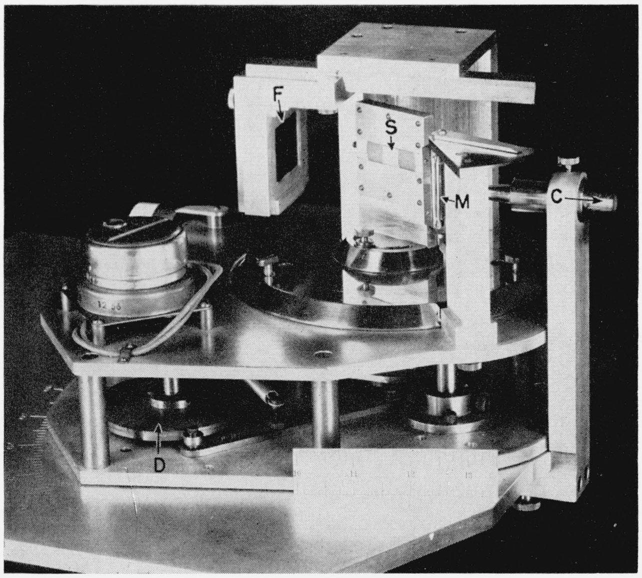

FIGURE 4. *Photograph of the monochromatic misorientation goniometer.*

M. Monochromating crystal; S. mounted brass specimen; F. flat-film holder; C. collimator; D. driving cam.

In this discussion, the term "broadening" of a diffraction spot means an increase of radial distance on the film over which blackening occurs; that is, it is an apparent increase in the range of Bragg angle over which diffraction takes place. Many factors contribute to the breadth of a spot in an X-ray diffraction pattern. Among the most important of these are (1) size of the diffracting crystallite, (2) lack of fine collimation, (3) lack of monochromatization, (4) randomly directed elastic strains of the lattice, and (5) micromisorientation. The first four factors, and possibly other instrumental factors, may all contribute in varying degrees to the broadening of spots whether the film is stationary or oscillating with the specimen. Only, however, in the latter case, that is, when the film is moving, does the fifth factor, misorientation, become important. By subtracting one-half the average breadth of many spots measured on a stationary film pattern from the average measured on a moving film pattern from the same specimen, a figure is obtained that represents the misorientation broadening. The average breadth on the stationary film must be divided by two because a linear distance on it corresponds to twice the angle of diffraction as compared to the same distance on the moving film.

In principle, the goniometer could also be used to determine the average microstrain, or range of atomic layer spacing, by comparing the stationary film spot breadths on the pattern from an annealed specimen with the pattern from the plastically deformed condition under study. It was observed, however, that this strain broadening was very small and probably within the limits of uncertainty in the measurements. It was not pursued in the present study.

Since the measurement of misorientation depended on the difference in broadening between stationary and moving film patterns, it was important to minimize the broadening due to the first four factors listed above; this consideration influenced many of the experimental details, including the choice of sample material. The metal used for the quantitative investigation was annealed alpha brass (70% Cu–30% Zn) with an average grain diameter of 0.035 mm. This grain size was sufficiently large so that it did not cause "small particle" broadening, but small enough so that the quantity it added to the "reflection breadth" (about 0.007 mm) was much less than the probable error of measurement of spot broadening. X-ray examination of the material indicated no preferred orientation.

The broadening due to lack of collimation and lack of monochromatization was minimized by using copper radiation and a lithium fluoride monochromating crystal. The Bragg angle for the strong (200) diffraction from this crystal is about 22½°, while the diffraction angle for the (111) ring from alpha brass is about 21°. Since these were the diffractions used in this study, the diffracting planes in the monochromator and specimen could be set nearly parallel to each other. This setting minimized dispersion [15] in the X-ray beam. The specimen was continuously oscillated about this position by a motor-driven, 3° cam mechanism. For comparable amounts of film density, exposure times for stationary film patterns were 6 hr and for moving film patterns, 10 hr, except for some few special cases of very diffuse patterns. Development of the films was carried out under as nearly constant conditions as practical. A typical pair of films, stationary and moving, is shown in figure 5.

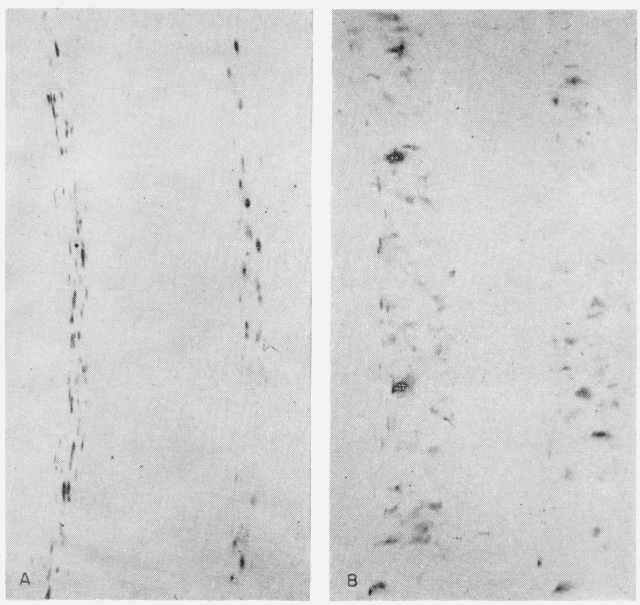

FIGURE 5. *A pair of patterns made with the monochromatic misorientation goniometer with* Cu Kα *radiation showing the (111) and (200) diffractions from brass, which had been extended 1 percent.*

A. Stationary film; B. moving film.

The brass was received in 1 by ⅜ in. bars, from which specimens 7½ in. long, with a gage length of 1.250 in. and a reduced cross section measuring 0.625 by 0.373 in., were machined. These dimensions were chosen so that a single specimen could be both plastically extended and compressed, could carry resistance wire strain gages nearly an inch long, and could provide sufficient test volume to allow the preparation for X-ray studies of three sections, with surface normals making angles of 0, 45, and 90°, respectively, with the axis of the applied load. It was desired to make observations on different sections in order to ascertain any directional effects in the misorientation that might occur due to the angle of the surface with respect to the axis of plastic deformation or due to the sign of the deformation.

The plastic extension was performed using Templin grips; the plastic compression was made possible by the careful machining of the ends of the specimens and by the use of massive steel guides around the specimen undergoing compression. The routine of the mechanical testing and strain measurements, along with a discussion of the difficulties that sometimes arise because of false negative permanent strains observed at small values of permanent set with resistance wire strain gages, has been previously reported [16].

The specimens from which the X-ray diffraction patterns were made were cut from the uniformly deformed gage length of the test specimens, with the normals to the surfaces to be polished each making a known angle, 0, 45, or 90°, with the axis of the applied load. The sections were mounted without heat in acrylic cement and carefully electropolished until, as verified by back-reflection X-ray diffraction patterns, all surface disturbance was removed.

On each diffraction film, 25 spots were measured. These were selected at random except that none was more than 12° from the equatorial region so that any azimuthal correction could be neglected and none was so near the edge of the diffraction band as to be obviously incomplete. Their breadths were measured parallel to the equatorial plane of diffraction on each pattern with a traveling microscope of about four-power magnification, reading directly to the nearest 0.01 mm. This type of diffuse spot measurement requires considerable practice, and allowance must be made for subjective factors, such as judgment of the edge of a spot, when data taken by different observers are to be compared.

From the difference between the average of spot breadths on the moving film and one-half the average on the stationary film, one obtained the contribution to the broadening from the misorientation present within grains in the polished surfaces of X-ray specimens sectioned at each of the three angles specified above from the deformed test specimens. This misorientation is given in the data table. It is difficult to state a meaningful value for the reliability of these data; they were arrived at from readings of two observers and involve several averaging procedures. A reasonable value for their significance is an average uncertainty of about $\pm 0.02°$, except for one set of patterns made after an extension of the specimen of 2.2 percent. The spots on the stationary film patterns in this case were not significantly different from those observed on the other stationary films and were measured in the same manner. On the moving film patterns from this specimen, however, the spots were so diffuse that they could not be measured even after prolonged exposure. Only a visual estimate could be made of their average breadth, which was about 1.5°.

The average misorientation is plotted in figure 6 as a function of the cumulative plastic strain for one cycle of 1 percent amplitude.

Results of intragranular misorientation measurements

State of plastic deformation			Average misorientation			
			45° Section	Cross section	Longitudinal section	Condition average
Annealed initial condition						6
Cycle position	Net strain	Cumulative strain				
Positive cycle	%	%	1/100 *deg*	1/100 *deg*	1/100 *deg*	1/100 *deg*
¼ (E)	+1	1	33	27	23	28
½ (E-C)	0	2	13	12	10	12
¾ (E-C-C)	−1	3	67	38	28	44
1 (E-C-C-E)	0	4	25	18	11	18
Negative cycle						
¼ (C)	−1	1	32	18	28	26
½ (C-E)	0	2	18	9	15	14
Large strain, unidirectional, 2.2 percent						150

Probable error in average about $\pm 0.02°$, except for case of 2.2 percent strain where it is about $\pm 0.50°$.
E—Extended 1 percent.
C—Compressed 1 percent.

3. Discussion

The misorientation reported in the data table and on the graph is the average contribution to the breadth of the diffraction spots arising from the misorientation within the diffracting grains. The fact that a small amount was observed with the material in the initial condition indicates the sensitivity of the method, since the evidence of mechanical tests, metallography, and the sharpness of ordinary back-reflection X-ray patterns indicated that the brass was well annealed. This misorientation is actually the range of angle of the family of atomic planes in the metal grain from which diffraction takes place. Both plastic lattice bending and small-angle fragmentation may contribute to this spot-broadening factor; their effects are not separable by this technique.

The maximum average values of misorientation were observed in specimens cut so that their surfaces made an angle of 45° with the loading axis. The

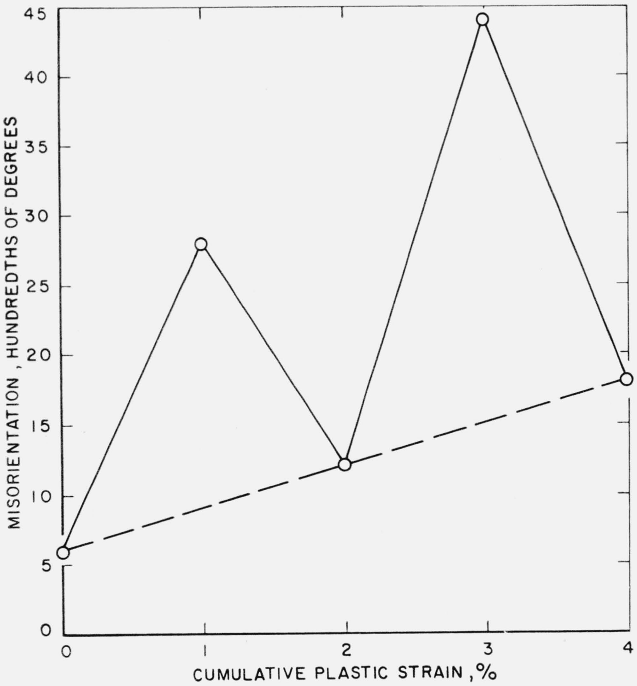

FIGURE 6. *Average misorientation versus cumulative strain for one cycle of plastic strain of 1 percent amplitude.*

directions of maximum shear stress were also at 45° to this axis. Because the angles of incidence and diffraction of the X-ray beam with respect to the polished surface were maintained equal, the diffraction planes (111) were parallel to this surface. The (111) plane is also the preferred plane of slip in this material. Hence, in the 45° section the observed diffraction spots came from crystals in which the angle between the slip plane and the maximum shear stress could be as small as zero. Since the smallest angle between (111) planes is 70°32′, in the diffracting crystals of the longitudinal and transverse sections the angle between the slip plane and the direction of maximum shear stress could not be smaller than about 25°; and it is significant that the misorientation observed in the latter cases was always smaller than that of the 45° specimens. These results emphasize the sensitivity of the method, and show that the lattice distortion of individual crystals in a polycrystalline matrix is related to their orientation.

The partial recovery of the misorientation with reversal of strain measured here in a quantitative manner is in agreement with those earlier qualitative observations on X-ray diffraction patterns cited in the introduction to this paper. These observations demonstrate that at least part of the lattice distortion is recoverable, and imply that, when the stress that has led to the generation and flow of dislocations in a crystal is reversed, at least part of the behavior of the dislocation system may also be reversed.

The average values of misorientation, 0.26° and 0.14°, observed in the two stages of the negative half cycle (i.e., begun with compression), as compared to the values 28 and 12 for the corresponding stages of the positive cycle, are symmetric, as expected in view of the shear nature of plastic deformation by slip in face-centered cubic metals. It was not felt necessary to carry the negative cycle any further under these circumstances.

The linear increase in the average residual misorientation at zero net strain is an interesting phenomenon. It indicates a permanent amount of disorder not recoverable by reversal of stresses on the dislocation system and is perhaps ascribable to the capture of dislocations at permanent pinning points due to plastic lattice bending carried so far that dislocations in other slip planes are generated. It would then be expected that the residual misorientation would be associated with the cumulative work-hardening of the metal, which Wood [17] found to increase nearly linearly with the first few cycles of reversed plastic deformation. Such a correlation is obscured by the reversible misorientation and by the Bauschinger effect, but it is hoped that further investigation will clarify the relationship.

The increase in misorientation when the unidirectional plastic strain was doubled was surprisingly large. It may reflect the increasing number of slip systems that must become active when individual crystals are forced to accommodate to the highly inhomogeneous strain in the polycrystalline aggregate.

4. Conclusions

The instrument and procedure described, while of only moderate complexity, permit quantitative measurements of intragranular misorientation in polycrystalline metals that are believed to be sufficiently precise for many purposes. Application of the technique to alpha brass specimens deformed in tension and compression showed that:

(1) Misorientation was highly sensitive to plastic deformation, the average value increasing from 0.06 to 0.28° when the annealed material was deformed plastically 1 percent.

(2) A large part of the misorientation was eliminated when the plastic strain was reversed.

(3) The residual misorientation increased linearly with increasing cumulative strain.

(4) The misorientation measured on surfaces parallel to directions of maximum shear stress was larger than that measured on surfaces either parallel or perpendicular to the loading axis.

The authors acknowledge the cooperation of colleagues at the National Bureau of Standards who made significant contributions to this investigation: in particular, A. N. Greaf, who built the goniometer, and H. E. Utech and P. D. Sarmiento, who assisted with the X-ray specimens and patterns.

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