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X-ray Interference Fringes in Berg–Barrett Micrographs

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Very fine lines, separated by distances in the range 3·0–7·5 microns, have been observed by examining, at high optical magnification (*ca.* 500 ×), Berg–Barrett micrographic diffraction patterns obtained in a study of bulk, cleaved zinc crystals. Bands of these lines which are clearly differentiable from dislocation images have been observed in cases where, effectively, the X-ray beam had passed through two non-parallel external crystal surfaces in order to give the recorded diffraction pattern. The bands of lines appear to be Pendellösung fringes which are produced because of the boundary conditions imposed by non-parallel crystal surfaces on the external diffracted X-ray intensity. The conditions under which the lines are observed in zinc indicate that the Pendellösung fringes occurred in crystals having a relatively high dislocation density ($\geq 10^6$ lines per cm^2) and that anomalous transmission of X-rays had occurred.

Introduction

In the course of a Berg–Barrett X-ray study of dislocations in melt-grown zinc crystals (Schultz & Armstrong, 1962), it was observed that sets of very fine lines were usually present near certain surface irregularities. The details of the appearance of the fine line structure indicated that these lines were different from the dislocation images that were observed: (1) the lines were observed as narrow strips of equally thick regions of enhanced and diminished intensity, as compared with comparably spaced dislocation images whose dark regions were relatively quite narrow; (2) the lines were extremely regularly spaced over substantial regions of the diffraction patterns, unlike any other structure observed; and (3) the regions of lines occurred only near surface steps rising away from the incident beam. The occurrence of the very fine lines is described in the following communication and a tentative explanation of their origin is given.

Experimental

The general features of the Berg–Barrett technique as it is used for observing dislocations in crystals has been described by Newkirk (1958, 1959). The Berg–Barrett X-ray fixture, the X-ray equipment, and the history of the zinc crystals used in the present study have also been described in a previous report on the observation of dislocations (Schultz & Armstrong, 1962). Here we briefly describe the experimental setup and conditions necessary for seeing the very fine lines. A schematic view of the apparatus is shown in Fig. 1. The specimens were squat cylindrical disks (0·8 cm dia. × 0·5 cm height and 2·5 cm dia. × 0·5 cm height) with their axes parallel to the 0001 direction. They were mounted in the X-ray fixture in a manner such that the incident X-rays bathed the 0001 plane(s) surface and a portion of the curved side of the specimen nearest to the incident beam. The crystals were oriented by first rotating them about an axis in the horizontal plane of the X-ray beam (plane of Fig. 1) so that a trace of the $11\bar{2}0$ family of directions was vertical (out of the plane of Fig. 1); and then the crystals were rotated about that vertical $11\bar{2}0$ direction until the Bragg angle was obtained in the horizontal plane of the beam for the appropriate plane in the $10\bar{1}3$ family of planes.

The diffraction micrographs were taken using iron characteristic radiation at 20 kV and 8 mA with exposures varying between one and two hours. Kodak High Resolution Photographic Plates were used to record the diffraction spots. A manganese foil was placed between the specimen surface and the photographic film in order to filter the radiation diffracted from the specimen. A horizontally mounted

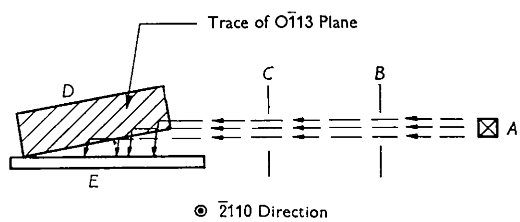

Fig. 1. Geometry of X-ray system. Radiation from source *A*, collimated by slits *B* and *C*, is diffracted from specimen *D*, and recorded on photographic plate *E*.

Machlett Laboratories Type A-2 Diffraction Tube with a spot focus 0·1 cm on a side provided the incident radiation with ~0·5° vertical divergence at the specimen placed 15 cm from the target. The specimen-to-film distance of points in the diffraction area varied between 0·01 and 0·20 cm. Diffraction spots of fairly uniform intensity over an area of

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$\sim 0{\cdot}25$ cm^2 were easily obtained. Chemical polishing experiments indicated that in the average case the reflected beam had penetrated to an effective depth of some 5 microns below the crystal surface. This effective depth of penetration compares, for a 90% decrease in beam intensity, with a calculated absorption depth of 5·3 microns and an extinction depth of 1·8 microns (James, 1953).

Results

The nature and scale on which the very fine lines were observed, in comparison with dislocations, is illustrated by Figs. 2 and 3. In Fig. 2, dislocations are revealed as the fine black lines distributed within subgrains whose boundaries, in turn, are shown as the larger scale, more intense black or white lines in the micrograph. Typical fine scale lines, unresolvable in the special regions where they occur in Fig. 2, are revealed in Fig. 3 which is an enlargement of the small area marked by the peculiar hook-like emulsion defect shown in both figures. Similar fine lines have been observed at crystal edges, cleavage steps, sub-boundaries, and etch hillocks or pits. These lines and those of Fig. 3 were interpreted as interference fringes rather than as dislocations because of the narrow line width, regular intensity, and rather uniform spacing, compared with the dislocation lines. Optical interference micrography showed that the lines in Fig. 3 occurred adjacent to a cleavage step of height 1·9 microns. The spacing of the fine structure observed in various regions ranged between 3·0 and 7·5 microns, most often nearer the smaller dimension. The fringe bands extended never less than 100 microns from the surface singularity. The dislocation densities of regions at which the bands were seen are strictly comparable to the dislocation densities of regions free of bands, *i.e.*, $\geq 10^6$ lines per cm^2. In fact, examination of optical interference fringes on the specimen surface near cleavage steps showed that some plastic deformation and bending had occurred in those regions where bands had been observed.

Discussion

The diffraction conditions which exist at the cleavage steps, crystal edges and other regions where fringes were observed are shown schematically in Fig. 4. Here, the path of X-rays through the crystal, which is bounded by surfaces with unit normals $\boldsymbol{\nu}_e$ and $\boldsymbol{\nu}_a$, is indicated at the various positions A, B and C. The incident X-ray beam having a wave vector $\mathbf{k}_0$ is reflected by the diffracting planes, whose reciprocal lattice vector is $\mathbf{B}_H$, to form the diffracted X-ray beam having a wave vector $\mathbf{k}_H$. It is well known that, when the diffracted beam exits through a crystal surface not parallel to the entrance surface, a splitting of the external beam into waves having two very slightly different wave vectors occurs to produce Pendellösung interference (Laue, 1941). Following the standard procedure outlined by Zachariasen (1945), the intensity of diffracted X-rays was obtained along an arbitrary surface behind and inclined to the incident crystal surface and, from the intensity fluctuation with position vector along this arbitrary surface, the spacing of Pendellösung fringes, S, was calculated as

$$S = \frac{\lambda}{K|\psi_H|\sqrt{((1+y^2)\cos\theta_G/\cos\theta_0)}\,.\,\cos\varphi/\cos\theta_G}$$

where λ is the incident radiation wave length, K is the polarization factor for the incident beam, ψ_H is the polarizability per unit volume associated with the crystal diffracting planes, y is a parameter measuring the deviation of diffracting planes from the Bragg angle, θ_G is the angle between the normal (pointing inward) to the incident surface and the diffracted beam, θ_0 is the angle between the incident beam and the incident surface normal, and φ is the angle between the position vector along the exit surface and the incident surface normal. These symbols are largely identical with those previously defined by Kato & Lang (1959); the fringe spacing is also similar to that derived by them, even though their expression for S was obtained directly from the difference in diffracted wave vectors. For the region of maximum intensity of reflection around the Bragg angle ($-1 \leq y \leq 1$) and for $\varphi = 90°$ at cleavage steps and crystal edges, a value of S ranging between 3·1 and 4·4 microns was calculated. These values of S are in qualitative agreement with the measured spacing of 3·0 microns. The experimental observation that the smallest fringe spacings occurred at crystal edges and cleavage steps is in agreement with the calculation given for S.

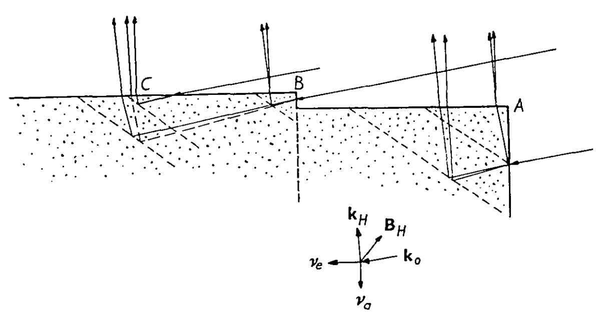
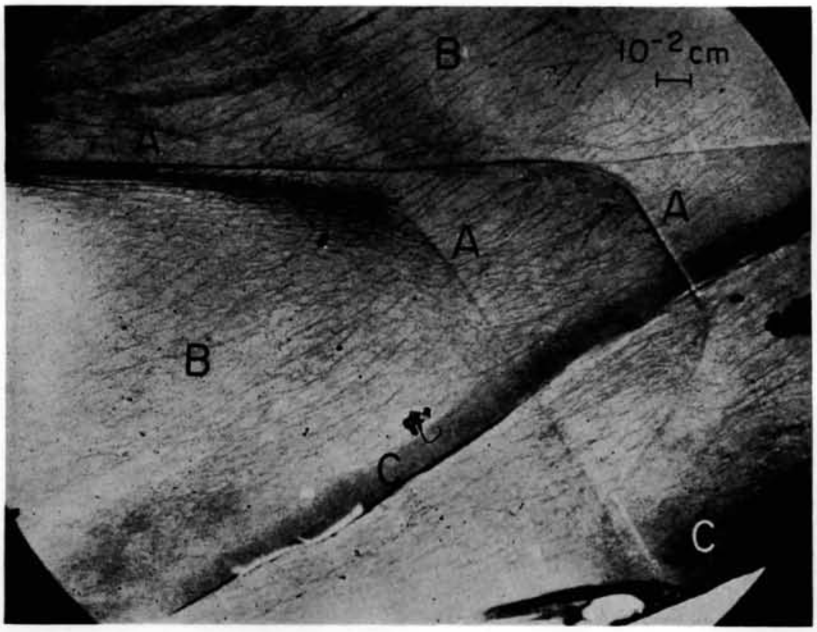


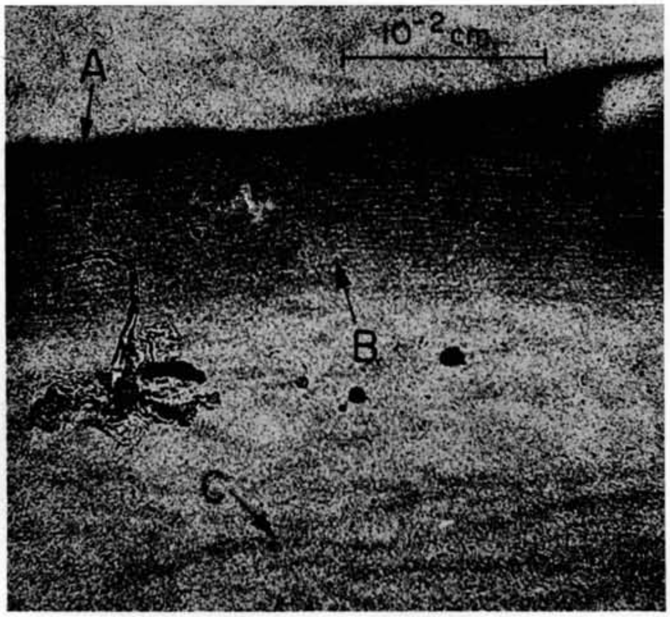
Fig. 4. Schematic view of X-ray interference effects. A-Laue diffraction at crystal edge, B-Laue diffraction at cleavage step, C-Laue and Bragg interaction at crystal surface.

Taking the interference fringes as Pendellösung fringes, then, their number and areal extent indicates that anomalous transmission of X-rays has occurred. That anomalous transmission is necessary is illustrated in Fig. 4. The positions A and B indicate respectively the crystal edge and a cleavage step in the diffracting face. It is clear that if absorption is high, X-rays



0$\bar{1}$13 Plane Reflection ; [$\bar{2}$110]

Fig. 2. Berg–Barrett X-ray micrograph of zinc crystal cleavage surface; Fe $K\alpha$ radiation. *A*-Dislocation sub-boundaries, *B*-areas containing dislocations, *C*-areas containing interference fringes.



0$\bar{1}$13 Plane Reflection; [2$\bar{1}\bar{1}$0]

Fig. 3. X-ray interference fringes at cleavage step in zinc; Fe $K\alpha$ radiation. *A*-Cleavage step on basal plane surface, *B*-area containing fringes, *C*-dislocations.

incident at one of the non-parallel faces, say at the specimen side surface at A, will contribute to the pattern only in a region quite close to the edge, since the X-ray path length would have to be very long to give a pattern extending far from the edge. On the other hand, if anomalous transmission occurs, a non-negligible area of the top face will be affected by radiation entering the side of the crystal. Thus, the incident X-ray beam must have traversed within the crystal volume a distance of several hundred microns in order to produce interference fringes at a distance equal to or greater than 100 microns from the 'wedge' vertex (Fig. 3). As mentioned earlier, the experimental measurement of an average depth of X-ray penetration and the calculated absorption and extinction distances were less than 10 microns. The conclusion to be drawn is that the X-rays have been anomalously transmitted.

It must be pointed out, however, that if anomalous transmission of X-rays has indeed occurred, then several difficulties arise in trying to match the experimental conditions for the observation of fringes to the general conditions associated with the anomalous transmission. A first difficulty is that to observe fringes one is concerned with having two reasonably intense diffracted waves of slightly different wave vectors at the exit crystal surface; but the theory of anomalous transmission as set forth by Borrmann (1941, 1950) and others (Hashimoto, Howie & Whelan, 1962) indicates that only one of the two internal diffracted waves within the crystal may have an anomalously low absorption factor, the other being absorbed before reaching the exit surface (see B, Fig. 4). Although this difficulty might be circumvented by interaction of the anomalously transmitted portion of the beam with the beam entering and exiting the same crystal surface (C, Fig. 4), a less reasonable (larger) fringe spacing was calculated for this case. A second difficulty is that the observed extent of fringe bands requires propagation of the energy in the X-ray beam in a direction which is not predicted. The band of fringes in Fig. 3 extends more than $100\,\mu$ from the cleavage step, while the height of the cleavage step is only $1{\cdot}9\,\mu$. If energy flow were along the diffraction plane (lying at $-35{\cdot}5°$ to the surface), then there would have to be an anomalously large component of energy flow nearly parallel to the surface to result in fringes as far from the step as is observed.

The relatively high imperfection content of the zinc crystals makes the observation of interference fringes and anomalous transmission of X-rays unexpected, even though quantitative conditions are not available on the crystal perfection required to obtain to observe these effects. Previously, Pendellösung fringes have been observed in a limited number of nearly perfect crystals using highly refined apparatus in which X-ray divergence is small (Kato & Lang, 1959; Kohra & Yoshimatsu, 1962). However, the anisotropic distribution of dislocations in zinc has been suggested by us (Schultz & Armstrong, in preparation) to be an important factor in allowing the observation of dislocations in zinc and has led Merlini (1961) to look directly for (and find) the anomalous transmission of X-rays in zinc by means of diffracted intensity measurements. Our results on observing fringes in an appreciably deformed crystal indicate that the degree of perfection of a crystal from the viewpoint of X-ray diffraction may be very different from the crystal being truly ideally perfect and, thus, further experiments must be performed before, as suggested by Kato & Lang, the observation of interference fringes may be used to prove the ideal perfection of a crystal.

Summary

X-ray interference fringes have been observed in a Berg–Barrett study of zinc crystals with the use of an unrefined apparatus and specimens of low perfection. The experimental conditions accompanying the observation of fringes and the nature of the fringes has been described. The fringes appear to be due to a Pendellösung effect. The areal extent of the fringes indicates that anomalous transmission of X-rays has occurred.

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