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MEASUREMENT OF TWIN MISORIENTATION BY USE OF FIRST-ORDER LAUE RINGS IN CBED PATTERNS

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Twin misorientation is conventioanlly determined via the measurement of spot splitting in selected area diffraction (SAD) patterns. However, due to its limited spatial resolution, it cannot be used in regions smaller than about 0.5 μm. In addition, two-dimensional microdiffraction patterns taken from such regions by a converged probe often fail to reveal such splitting. This paper discusses the use of first-order Laue zone (FOLZ) ring displacement to gauge such information.

Background

A selected-area diffraction pattern taken across alternating structural domains (twins) can be visualized as arising via a superposition of the reciprocal lattices of the individual domains. In the case of a tetragonal distortion, the angle between the c axes in adjacent twins deviates from 90° by an angle 2ε. The configuration is shown in Fig. 1, which yields[1]

$$c/a = \tan(45^\circ + \varepsilon) \tag{1}$$

where c/a is the tetragonality of the twinned phase.

In a [010] tetragonal zone axis diffraction pattern, 2ε is the angle subtended at the center of the reciprocal lattice by the split diffraction spots (Fig. 1b), and measurement of this spot splitting is a conventional method of determining the tetragonality of the sample. This determination is also possible with higher precision by analysis of Kikuchi line pairs.[2] However, the spatial resolution of both techniques is no greater than about 0.5 μm, owing to spherical aberration in the former case, and in the latter the broadening of the effective sampling area for inelastically scattered electrons, from which Kikuchi lines arise. Analysis with higher spatial resolution would be possible by measurement of high-resolution lattice or structure images, although only if the area of interest is very thin; furthermore, the method is highly demanding of both microscope and operator.

In this paper a method is proposed for measurement of small misorientations (≤1°) in more general cases with high spatial resolution by means of convergent beam electron diffraction (CBED). The high spatial resolution results from the use of a converged probe (∿ 10 nm), with beam broadening minimized since only eleastically scattered electrons are considered.

The fine dark higher-order Laue zone (HOLZ) deficiency lines in the central disk of a CBED pattern are sensitive to orientation, and superposed sets of lines from adjacent domains could in principle be used to gauge misorientation. However, these lines are frequently not visible (when of insufficient contrast above background intensity), and in addition can lose much contrast because of the dependence on exp $(-g^2)$ of the Debye-Waller effect and the large g-vector of HOLZ reflections.[3] The bright line-segment reflections in the HOLZ ring, which are the dark-field counterparts of the central-disk deficiency lines, usually have sufficient contrast above the dark background to be visible in most cases despite the Debye-Waller effect, and these are the pattern features on which the present method depends.

Recently, Tanaka et al.[4] have reported the measurment of misoriented regions in silicon via FOLZ ring displacement. However, the diffraction geometry was not considered in detail. The present paper discusses the situation in more depth.

Consider a CBED pattern taken across adjacent twins, looking down the a or b axis of *one* twin. Due to beam convergence, the Ewald sphere is spread out and intersects each twin reciprocal lattice (Fig. 2). As shown, the projected HOLZ ring radius in reciprocal space G will be shifted by a distance x with respect to that of the first twin. From geometry, we can relate the misorientation to the HOLZ ring displacement via:

$$\cos \varepsilon = \frac{(G - x)}{G} = 1 - \frac{x}{G} \tag{2}$$

with each term as defined above. For small ε, we can write

$$\cos \varepsilon = 1 - \varepsilon \tag{3}$$

with ε measured in radians. For x and G expressed in the same units, Eq. (3) becomes

$$\varepsilon = \frac{x}{G} \tag{4}$$

Thus, the angle ε can be measured from

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the HOLZ ring displacement. An examination of the configuration (Fig. 2) reveals that the beam convergence angle must be of the order of the misorientation angle, or greater, to observe this effect. If the foil orientation was not near to the a (or b) axis, unwanted inelastic intensity becomes visible in the ring (Fig. 4c), and the simplicity of the above geometry is lost.

Experimental

Figure 3 shows the evolution of the ring displacement with convergence angle. The analogy with spot splitting is clear. As an experimental example, Fig. 4(b) shows a [110] CBED pattern obtained from a twinned region in $Pb_{0.7}La_{0.2}O_3$ + 0.1 PbO (Fig. 4a). The misorientation angle ε was determined to be 0.65 ± 0.02° from Eq. (4), with x representing the maximum HOLZ ring displacement (Fig. 3b). This angle is similar to values obtained via the less accurate method of selected area diffraction spot splitting (1.2±0.08°) and compares favorably with an ε of 0.7°, calculated from reported c/a values for the bulk material.[5] Figure 5 demonstrates that this technique can reveal such information from localized regions when two-dimensional microdiffraction cannot.

Summary

The utility of the technique described above lies in situations where crystallographic misorientation (of the order of the beam convergence angle) information is desired from small (<0.5μm) regions. Such information can be obtained via a single convergent beam electron diffraction pattern at enhanced spatial resolution with respect to selected area diffraction techniques, and in regions where two-dimensional microdiffraction fails to reveal such data.

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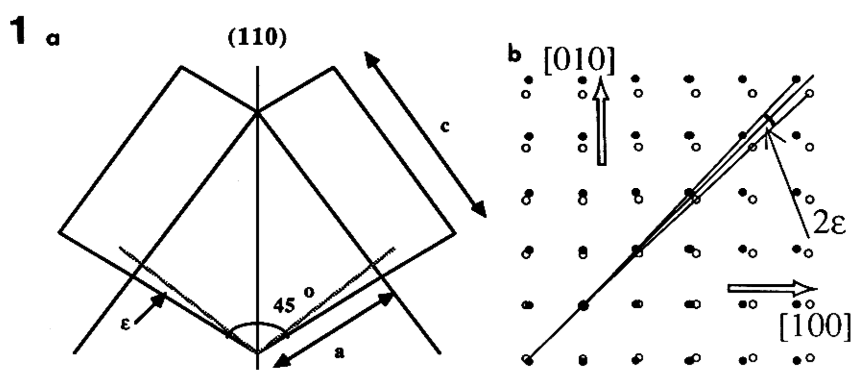

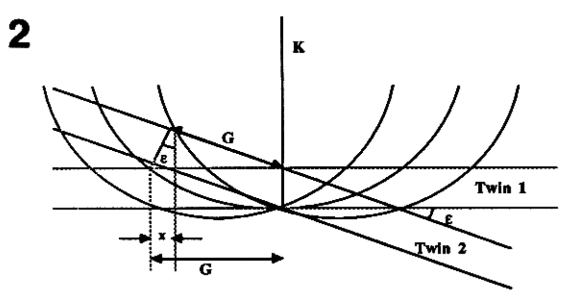

FIG. 1.-- (a) Schematic showing reorientation of a tetragonal cell by twinning; (b) corresponding diffraction pattern, showing origin of spot splitting.
FIG. 2.--Diffraction geometry leading to displacement of FOLZ ring.

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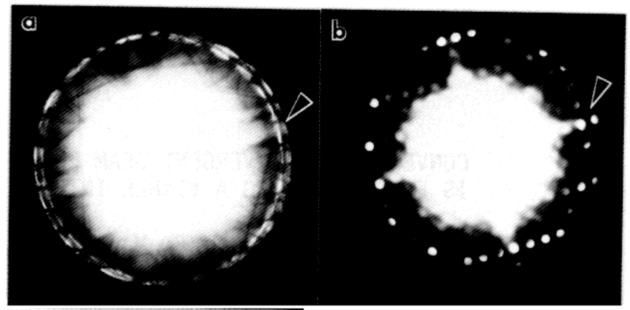

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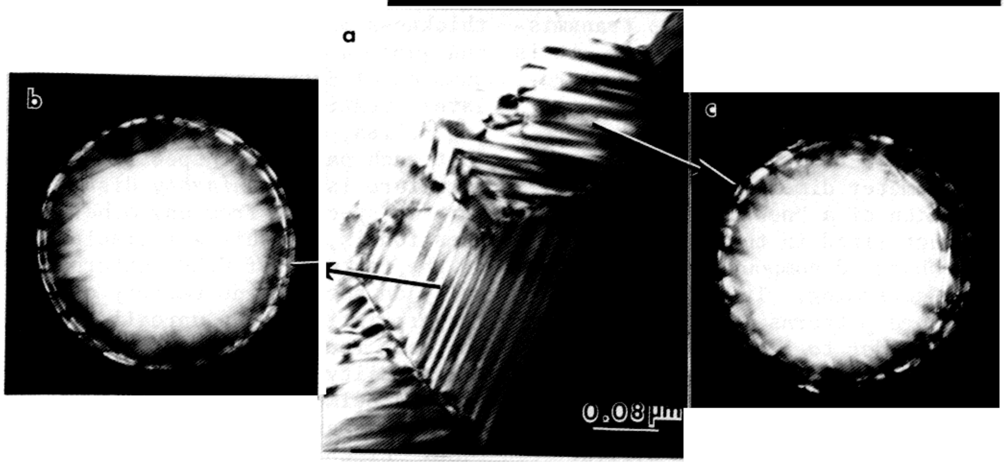

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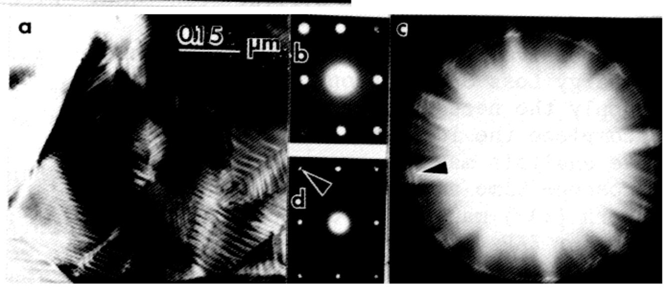

FIG. 3.--[110] CBED patterns from twinned region, showing evolution of FOLZ ring displacement with convergence angle; (a) 14 mrad, (b) 7.2 mrad.
FIG. 4. (a) Bright-field image of structural domains (twins) within single grain; (b) [110] CBED patterns taken across adjacent nearly edge-on twins, exhibiting a displacement of the FOLZ ring perpendicular to the twin boundaries, (c) across inclined twins, showing disjointed FOLZ ring sections arising from inelastic scattering (convergence angle 14 mrad).
FIG. 5.--Comparison of [110] patterns in revealing misorientation information from localized twinned region (a). (b) Microdiffraction pattern, convergence angle 27 mrad; (c) CBED pattern, convergence angle 18.7 mrad; (d) [110] SAD pattern are from same general area shown for comparison.