Application of the 3D X-Ray Crystal Microscope to Study Mesoscale Structure of Materials

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ABSTRACT

The 3D x-ray crystal microscope is an emerging tool for the study of mesoscale structure in polycrystalline materials. With this nondestructive device, local crystalline orientation, phase, elastic and plastic strain tensors can be measured with submicron spatial resolution in three dimensions. A key step in analyzing the Laue patterns from the 3D microscope is indexing the reflections, which determines the orientation of the sub-grain. With current algorithms, the angles between pairs, triplets and quadruplets of reflections are compared to theoretical angles to make guesses as to the reflection indices. The ability to index a pattern can however be compromised by both elastic and plastic deformation of a grain; elastic deformation changes the angles between reflections and plastic deformation increases the uncertainty in the centroid of each reflection. Here we report on the use of an indexing algorithm that simultaneously fits all peaks from a subgrain. This algorithm is more robust than previous methods and allows for indexing of deformed or strained grains. Some applications to studies of mesoscale materials properties are described.

INTRODUCTION

The 3D x-ray crystal microscope is a fundamentally new approach to the study of mesoscale structure in polycrystalline materials (Fig.1) With this nondestructive penetrating, scanning microprobe, local crystalline orientation, local phase, elastic and plastic strain tensor

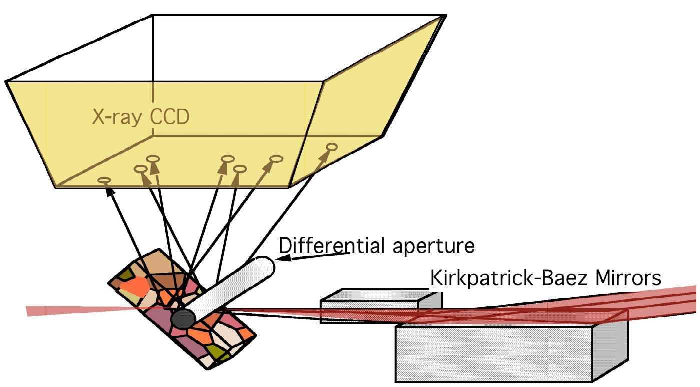

Figure 1. Schematic of the 3D X-ray Crystal Microscope. A polychromatic beam enters from the right and is focused by a nondispersive Kirkpatrick-Baez mirror pair [6] onto the sample. The Laue patterns generated by subgrains sampled the submicron beam are collected by an x-ray sensitive CCD located directly above the sample. The origin of the overlapping Laue patterns along the incident beam are decoded using a high Z wire that runs in front of the sample and acts as a virtual pinhole camera [3] or differential aperture microscopy.

distributions and defect structures can all be measured with submicron spatial resolution in three dimensions. Polychromatic Laue diffraction is used to determine the local unit cell parameters and orientation, which gives the local deviatoric strain tensor and texture. If the energy of one of the Laue reflections is measured than the full strain tensor can be recovered [1,5].

A key step in analyzing the Laue patterns is indexing the reflections, which determines the orientation of the sub-grain [1,4]. From precision measurements of grain orientation, the grain and subgrain structure can be determined and grain-boundary types can be characterized. Indexing is also a prerequisite for the measurement of elastic and plastic strain. Here we report on the application of the 3D x-ray crystal microscope to study mesoscale structure in materials. In particular we describe a new indexing algorithm that simultaneously fits all peaks. This algorithm is more robust than previous methods and allows for automated indexing of deformed and strained samples.

INDEXING SAMPLES WITH SHARP LAUE SPOTS

In many samples, Laue spots are sharp and the angles between reflections are well defined by the centroid of the spots. For these samples, established methods exist for finding and fitting the Laue spots and for determining the orientation of the grains from the angles between the reflections [1,2]. Typically a good angular match to four reflections is sufficient to determine orientation. This approach has been widely and successfully applied to a number of materials systems including electronic interconnects, polycrystalline Ni, Al and Cu and high temperature superconducting, Sn and diamond films.

Recent measurements of Sn provide a good example of the kind of unique information that can be obtained using the 3D x-ray crystal microscope. In these measurements, the local orientation of individual Sn grains were characterized in a thin Sn film with whiskers. The measurements are aimed at testing conflicting theories of whisker growth in these materials. Figure 2 shows a false color image of two adjacent slices through the Sn film. Whiskers can be seen projecting out of the film near the left and center of the image. From these and similar images we conclude that the orientations of whiskers can be different, that whiskers can grow on top of grains with distinct orientations and that the orientation of the whiskers can change abruptly during growth. We can also determine the grain boundary types surrounding each

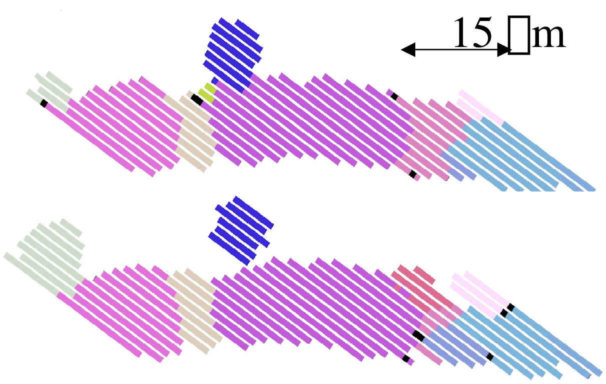

Figure 2. Adjacent slices- 2 μm apart- through the surface of a Sn film. Whiskers are clearly visible, projecting out of the surface of the film near the left edge and center of the images.

whisker. Additional information that can be obtained includes the local strain tensor distribution and the local plastic deformation tensor [8,9].

INDEXING DEFORMED SAMPLES

Although many samples have sharp Laue spots with minimal deformation of the unit cell, plastic/elastic deformation and/or elastic-strain gradients can smear the Laue spots and/or change the angles between reflections. Smearing can be reduced by differential aperture microscopy [3], but residual smearing and uncertain angles between planes can complicate automated indexing methods.

For example, in fusion-welded samples, smearing of the Laue reflections varies with crystal orientation and with distance from the weld core. We have studied iridium weld material where smearing is indicative of local plastic deformation. In order to index the smeared Laue patterns, a more robust approach to automated indexing was developed. Here the entire Laue pattern from a subgrain is predicted based on an assumed index for a single "primary" reflection. A similar approach has been suggested previously [7]. The pattern is calculated for all possible crystallographic orientations assuming a correct index assignment for the strong reflection. The fit is then compared at different orientations and for all possible primary reflection indices. A best fit with this method correlates all predicted reflections simultaneously with the measured Laue pattern.

The technique is illustrated in Fig. 3. Here the bright reflection near the center of the image is used as the primary reflection. As illustrated in the figure the Laue pattern rotates around the primary reflection depending on the possible orientation of the grain. The calculated pattern is compared with the measured pattern and a goodness of fit is determined based on overlaps of the calculated pattern and the measured pattern. Various indices are tried for the primary reflection and the best fit is assumed to represent a single subgrain contribution to the overlapping Laue patterns. The process is repeated with residual strong reflections that are not indexed in previous fits to account for multiple subgrain contributions.

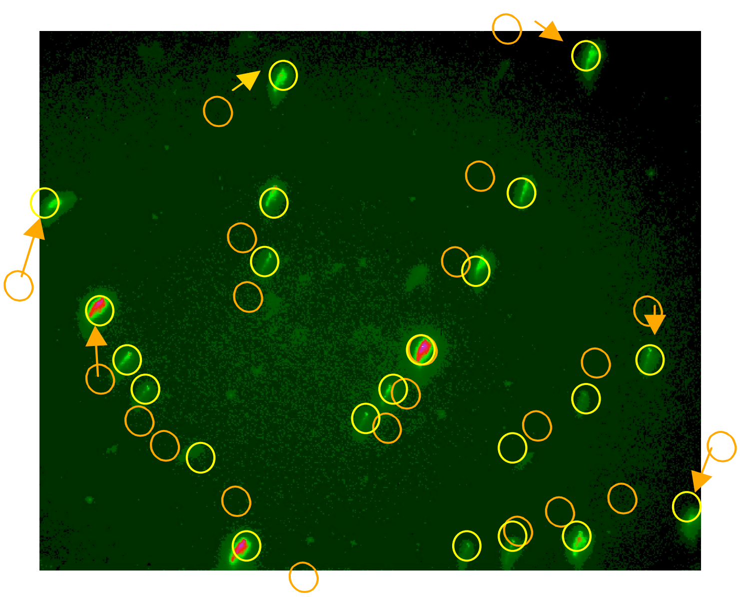

Figure 3. Laue image showing streaking and a schematic of how all reflections can be fit simultaneously. Using this approach, the grain structure, deformation and local orientation of even highly deformed materials can be determined.

Because virtually all Laue spots from a single subgrain are fit simultaneously, the angular precision does not need to be as high as with pair, triplet or quadruplet fits to the Laue pattern. For this reason the method is well suited for cases where the sample may be highly deformed (uncertain angles between reflections) or plastically deformed (uncertain position of Bragg peaks).

A study of an iridium weld (Fig. 4) provided the first example application of this new approach to indexing polychromatic Laue patterns. As shown in the micrograph of Fig. 4a, the iridium weld contained large (20-100 μm) grains. The Laue patterns generated from various regions of the sample showed significant streaking due to plastic deformation. This deformation is believed to arise from elastic inhomogeneities between the grains with different orientations.

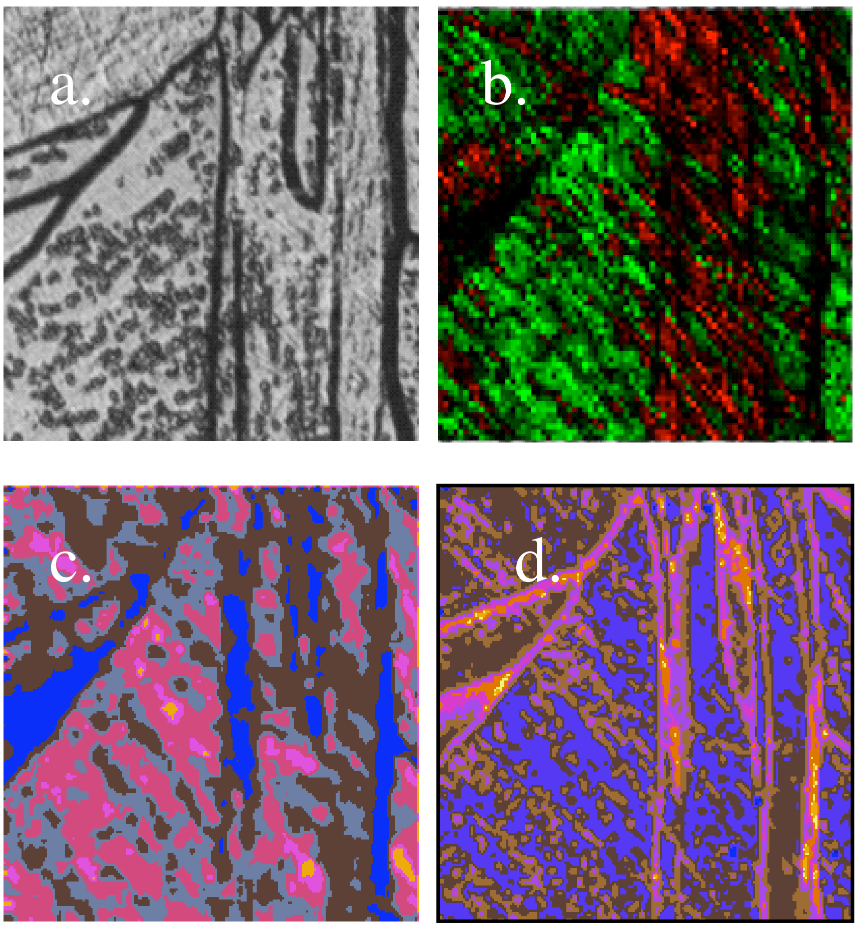

Figure 4. Various contrast images from an iridium weld. (a) Upper left- optical photomicrograph; (b) upper right -Laue spot streak direction; (c) lower left-Laue spot streaking magnitude; (d) lower right-"number" of Laue spots.

Although the Laue patterns were difficult to fit using pair and triplet matches, the rotation method was found to be very robust and the orientations of the various grains is found to match closely with the photomicrography image that highlights the grain boundaries Fig. 4a and with the number of Laue spots, which is sensitive to grain boundary location Fig. 4d.

The direction of the streaking is indicative of the primary active dislocation system(s). A false color image indicating the direction of streaking is shown in Fig. 4b. The length of the streaking is indicative of the total number of unpaired dislocations Fig. 4c.

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