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Multi-Scale 3D Imaging of Strain and Structure with Dark-Field X-Ray Microscopy

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Most materials and minerals have hierarchical structures whose dynamics span several length and time scales, giving rise to bulk properties that differ markedly from the properties of its foundational crystalline elements. Understanding and predicting this interplay therefore requires multi-scale experimental techniques that can rapidly swap between different length scales and make *in-situ* three-dimensional (3D) movies of these crystalline elements, including their symmetry, strain and orientation. Probing local crystallography favors diffraction-based approaches, yet existing 3D techniques are seldom multi-scale. Electron methods provide high spatial resolution but are destructive; limited to thin foils or involve serial sectioning. Scanning and coherent X-Ray methods are rapidly progressing towards 1 nm resolution, but tend to be slow and may cause radiation damage in delicate samples. Furthermore, all current methods face the challenge that the illuminated part of a bulk sample comprises many structural elements whose diffraction signals may overlap, preventing data analysis and interpretation.

Dark-field X-Ray Microscopy (DF-XRM) is a promising alternative (Figure 1) [1]. As a full-field diffraction microscopy technique, it is capable of non-destructively mapping lattice symmetry, orientation and strain in crystalline elements buried deep within mm-sized samples. The technique is easily combined with coarse-scale 3D techniques such as Diffraction Contrast Tomography, enabling users to intuitively progress from fast overviews of an entire specimen to details studies of local phenomena within a single experimental setting. The key feature of DF-XRM is its X-Ray objective lens, which generates a magnified image of the sample on a 2D detector. Placing the objective in the diffracted beam means that image contrast arises from the structural heterogeneity within the diffracting volume. The objective also acts as a collimator, excluding diffraction signals from other grains that would otherwise overlap with the signals of interest. The current implementation of DF-XRM at ID06 of the European Synchrotron Radiation Facility (ESRF) uses a compound refractive lens (CRL) as the objective, giving spatial and angular resolution of 100 nm and 0.001°, respectively. Combined with its speed and flexibility, this resolution makes DF-XRM an interesting new tool for measuring and understanding multi-scale structural dynamics in bulk materials. The following test cases from our recent work at the ESRF serve to demonstrate these capabilities.

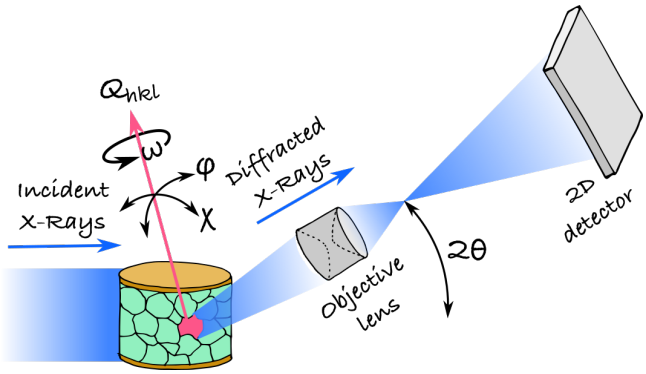

Figure. 1. DF-XRM uses an objective lens to create a magnified image from diffracted X-Rays. Maps of axial strain (ε_{33}) and lattice tilt involve scanning the objective and detector through (2θ), while maps of lattice tilt are obtained by scanning the sample tilt through (φ,χ). 3D data is obtained by acquiring projections around the scattering vector Q_{hkl}.

Case 1: Annealing:

Plastically-deformed metals are classically hierarchical. The deformation causes dislocations to proliferate and self-organize into extended networks defining grains and sub-grains from tens of µm to sub-µm in size. Heating the deformed structure then mobilizes the dislocations, causing the sub-grain structure to change. Predicting sub-grain structures in bulk materials is beyond the capabilities of bottom-up modelling approaches, and instead requires hybrid models that capturing the structural dynamics at specific length scales [2]. Such models must be closely guided by prior understanding of the structural dynamics, yet fundamental questions still remain regarding the deformation and annealing processes. The high angular resolution of DF-XRM provides a way to directly see these sub-grains and the dislocation networks that define them. Figure 2 shows the sub-grain structure in a single buried grain of annealed aluminum, made visible by the small but obvious changes in lattice tilt. The tilt difference across the sub-grain boundaries is of the order of 0.05 °, corresponding to dislocations separated by more than 0.5 µm. Unlike traditional methods such as electron backscatter diffraction, DF-XRM provides a measure of the angular distribution associated with each voxel in the sample, and a potential vehicle for improved estimations of local defect densities.

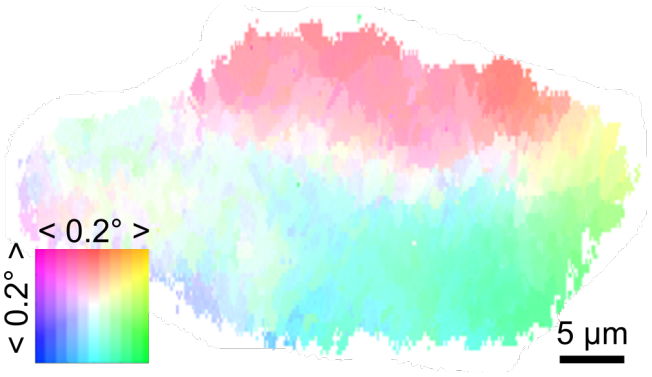

Figure. 2. Lattice tilt map from a single grain of aluminum after annealing at 325 °C for 20 minutes. Sub-grain structures are clearly visible as the pink, blue/green and blue/purple regions. From [3].

Case 2: Dislocations:

The ability to detect sub-grains raises the question of whether one could directly image the individual dislocations that separate them. Visualizing 3D dislocation structures is essential to understanding micromechanics and plasticity, and vital for optimizing the performance of microelectronics. Using TEM to visualize dislocations provides high contrast and spatial resolution, however the small samples are subject to spurious electromechanical boundary conditions and present a significant challenge for *in-situ* experiments such as deformation and creep. DF-XRM, on the other hand, provides similar contrast to TEM, greater flexibility regarding samples and environments, and, in favorable circumstances, sufficient spatial resolution to identify individual dislocations. Imaging dislocations using DF-XRM requires a "weak-beam" condition wherein the diffracted intensity is from the tails of the main Bragg peak (Figure 3). This greatly enhances contrast from the dislocation and ensures that the scattering is kinematical. A series of several images at different $(\varphi,\chi,2\theta)$ positions around the Bragg peak (i.e. positions in reciprocal space) can be constructed into quantitative maps of lattice tilt and strain corresponding to the axial component of the lattice displacement gradient in the x, y and z directions. Three of nine components of the dislocation density tensor may then be computed directly from the experimental data, revealing the character of the dislocation.

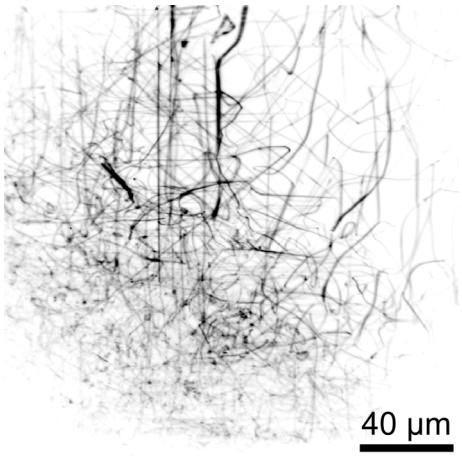

Figure. 3. DF-XRM inverse-intensity map of dislocation lines around an indentation in silicon.

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Case 3: Ferroelastic domains:

A defining feature of ferroelastic materials is the presence of domains: distinct regions separated by an atomically-coherent lattice misoritentation (e.g. inversion about a zone axis) that self-organize in response to stress via the collective motion of domain walls. This gives rise to a range of applications including shape-memory alloys, digital memory and solid-state capacitors. Furthermore, the discovery of emergent phenomena at the domain walls (e.g. photoelectricity) has renewed interest in the local structure at and around the walls. Here, DF-XRM was used to quantitatively map lattice symmetry and strain around individual domain walls buried within mm-sized samples. Figure 4 shows the local strain and lattice tilt around domain walls in a single crystal of $KNbO_3$. The stripe-like features are domains with <110>-type orientations (as one regularly sees using conventional methods). A closer inspection of the domain orientations, however, shows small orientation changes in the order of 0.01° - far below the expected twinning angles in $KNbO_3$ (0.2-0.3°). These unusually small orientation changes strongly suggest the presence of dislocations or potentially even "S-walls"; a rarely-seen type of domain wall with non-integer twinning planes. These observations are key to understanding and accurately modelling ferroelasticity, and are directly comparable to multi-scale models, such as phase-field and Monte-Carlo simulations.

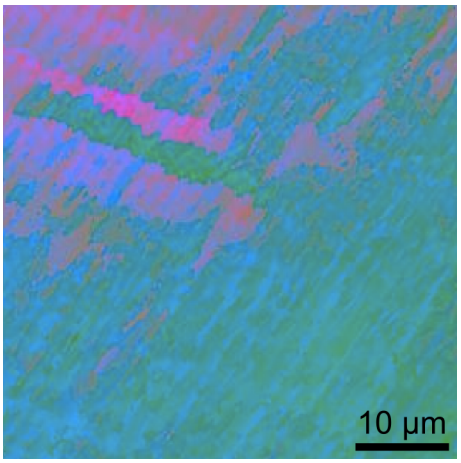

Figure. 4. Lattice tilt variations along and between domain walls in a single crystal of $KNbO_3$. See [4].

Future directions:

DF-XRM's ability to non-destructively map strain and structure in the bulk makes it a promising technique for studying multi-scale phenomena in a wide variety of systems. However, the relatively poor spatial resolution of DF-XRM limit studies to structures larger than 0.2-0.5 µm, while its acquisition rate of 0.1-1 s/image – though fast – is still too slow to observe many interesting processes (e.g. phase transformations). Current efforts are therefore directed towards improving the numerical aperture (and hence spatial resolution) and imaging rate. In particular, the use of multilayer Laue lenses (MLLs) as objectives are anticipated to yield a four-fold improvement in spatial resolution (to 25 nm), and a two-fold improvement in transmission efficiency (to 60%). The short focal lengths of MLLs (5 cm) reduce the space available for sample environments, however we believe the gains in performance will make it a favorable compromise. At the same time, diffraction-limited (i.e. "ultimate") synchrotron storage rings are expected to boost the imaging rate by a factor of 100-1000 more. The lower beam divergence would allow us to focus more X-Rays on the sample, while the reduced bandwidth from the undulators would potentially allow us to bypass the monochromator altogether. Combined with continued development of analysis methods and experimental design, we hope these improvements will open the door to real-time studies of complex multi-scale phenomena across a broad range of materials and systems.

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