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4D-STEM Measurement of Thickness and Orientation by Bloch Wave Dynamical Diffraction Matching

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Determination of sample thickness in the electron microscope is a common challenge, traditionally solved using electron energy loss spectroscopy (EELS) measurements and known scattering cross sections. While this method is straightforward and flexible, it gives no information on the structure of the material. Using four-dimensional scanning transmission electron microscopy (4D-STEM), thickness mapping has been demonstrated at unit-cell resolution using position averaged converged beam electron diffraction (PACBED) [1]. Though PACBED can simultaneously measure structural features such as polarization and mistilt from a zone axis, it is limited to small fields of view by the need for an atomic resolution probe, requires large scale simulations, and generally requires tilting the sample to a high-symmetry zone axis. In this work, we develop an approach for mapping crystal orientation and thickness from 4D-STEM datasets that is applicable to unknown and low-symmetry orientations, large fields of view, and compatible with simultaneous strain and phase mapping; we demonstrate its effectiveness by measuring the thickness of randomly oriented collection of nanoparticles using a simulated 4D-STEM dataset.

Using the automated crystal orientation mapping (ACOM) module of py4DSTEM described in [2], we obtain an initial guess for the orientation of the particle at each scan position by comparing the detected Bragg peak locations to a library of kinematic diffraction patterns. This information is used to index each detected diffraction disk and compute a list of potentially excited Bragg beams to include in a dynamical diffraction calculation. Using the Bloch wave method [3], we efficiently compute a series of diffraction patterns using those beams for a range of thicknesses and mistilts from the kinematic guess. The best matching pattern based on the sum squared difference from the experimental intensity ratios is used to assign a thickness and a precise orientation at each pixel of the scan.

For testing, we simulated 4D-STEM data from MgO nanoparticles with cubic shape and side lengths of 5-10 nm, which are randomly oriented and sitting on an amorphous carbon substrate, using the abTEM multislice code [4]. The atomic model and selected diffraction patterns are shown in Figure 1, and a virtual ADF image is shown in Fig. 2a. py4DSTEM [5] was used to detect the diffracted disks from each pattern, and the ACOM module was used to obtain the initial guess of the crystal orientation on a 1° mesh, shown in Fig. 2b. Some instability in the measured orientation is observed due to particles being aligned far from a high-symmetry zone axis that would provide unambiguous orientation information. The thickness was then computed using the dynamical diffraction matching approach while simultaneously refining the orientation angle over a search range of ±1° from the initial guess. The estimated thickness is shown in Fig. 2d and compared to the actual thickness of the MgO in Fig. 2c. The thickness measurements are accurate in the larger particles and correctly show the variation in the projected thickness caused by the tilt of the particles. In the thinner particles, and near the edges of some

of the larger ones, the thickness measurements are observed to be less reliable and result in large overestimates of the thickness. The sources of the thickness error will be discussed and we will present strategies to improve the method in the future [6].

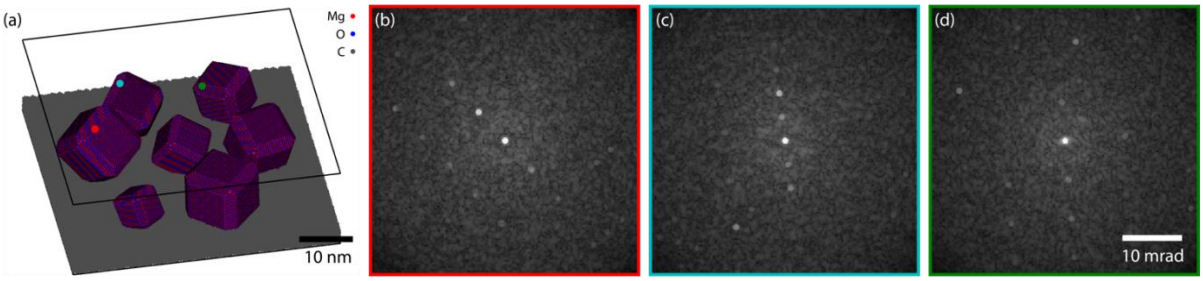

Figure 1. (a) Atomic model of cubic MgO nanoparticles sitting on amorphous carbon used for simulations. (b-d) Selected diffraction patterns from the 4D-STEM dataset taken from the locations marked by colored circles on the atomic model.

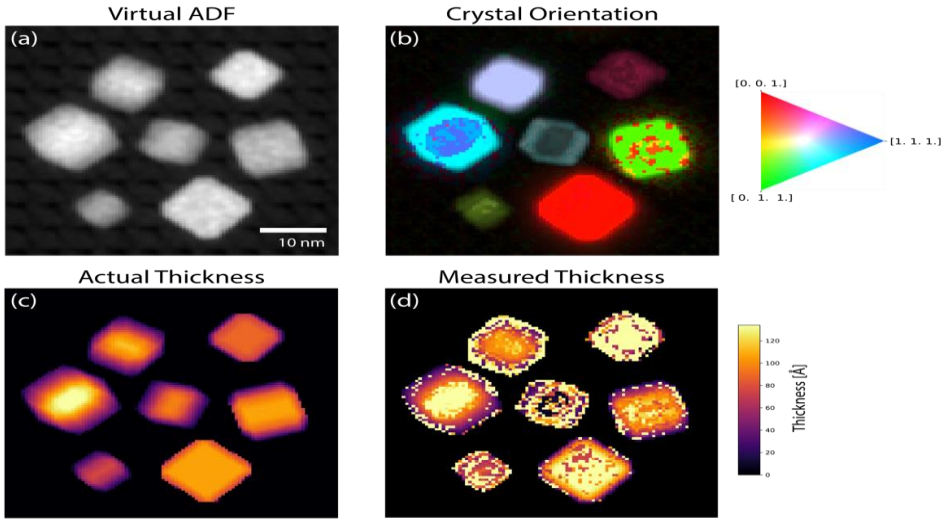

Figure 2. (a) Virtual annular dark field generated from 4D-STEM dataset (b) Automated crystal orientation mapping results from py4DSTEM. (c) Actual projected thickness of the MgO and (d) MgO thickness obtained from the dynamical refinement approach. Panels (c) and (d) share the same color bar.

References:

[1] LeBeau et al., Phys Rev B **79**(21), p. 214110.
[2] Ophus et al., Microscopy and Microanalysis (2021), p. 1.
[3] DeGraef in "Introduction to Conventional Transmission Electron Microscopy" (2003).
[4] Madsen and Susi, Open Research Europe **1** (2021), p. 24.
[5] Savitzky et al., Microscopy and Microanalysis **27**(4) (2021), p. 712.
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