

Automated Grain Segmentation for Crystal Orientation mapping in 4D Scanning Transmission Electron Microscopy

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Four-dimensional scanning transmission electron microscopy (4D-STEM) is transforming the field of electron microscopy by allowing quantitative, spatially resolved crystallographic analyses with exceptional precision [1-3]. The 4D-STEM procedure involves collecting hundreds to thousands of electron diffraction images where the probe position for each image is accurately recorded, allowing diffraction measurements to be resolved to real space. While the increased popularity of 4D-STEM in microscopy can be largely attributed to engineering advances in computing hardware, the management of the enormous datasets produced by this method remains challenging [4]. Even moderately large scans can generate upwards of 100 GB of data, which on top of logistical concerns for storing such quantities, also raises practical matters of post-processing since common random access memory card capacities fall far short of this scale. Concomitant with hardware constraints, the time required to complete these scans is excessive. In addition to throughput reasons for desiring a shorter scan time, a more indirect consequence of longer scanning in a serial diffraction image collection sequence is the increased effect of sample drift, which can distort the spatial mapping of nanostructural detail.

This work is designed to overcome the feasibility concerns of inflated datasets and extensive STEM scan times by using a mindful approach when choosing probe positions for the acquisition a priori. Our solution uses segmentation and other software tools to limit the quantity of data collected, while minimizing the loss of useful information in the case of 4D-STEM scans used in crystal orientation mapping. By assuming a consistent lattice orientation throughout each grain in a polycrystalline sample, a preset number of diffraction measurements can be assigned to automatically segmented crystal regions, significantly reducing the required workload.

Diffraction patterns of ultrafine-grained NiCo thin films were imaged using two probe positioning schemes: a 128 by 128 grid (16,384 points), and a non-uniformly spaced segmented position pattern (3,157 points). The larger scan took a total time of 3 hours to complete, while the smaller scan finished in 30 minutes. The probe positions overlaid on top of darkfield (DF) images is shown in Fig. 1 to provide a visual comparison of the two plans. 4D-STEM data collection was done on a Hitachi HT7700 TEM/STEM using the Azorus software package developed by Hitachi High-Tech Canada. The segmentation of the image was performed by a plugin in Azorus, custom developed by the authors, and the corresponding 4D diffraction data of each segment were collected by the software.

Crystal orientation mapping was achieved using py4DSTEM, an open-source Python package that contains a variety of 4D-STEM data analysis tools [5]. Following calibrations of the diffraction set origin location, pixel sizes, and corrections of elliptical and rotational distortions, diffraction images

were used to assign each probe position a rotational orientation. Diffraction patterns were matched to best-fitting predicted patterns, generated assuming a face-centered cubic NiCo lattice.

Reconstructed maps showed clusters of similarly oriented positions, with object features closely resembling visible facets in reference DF images. Fig. 2a shows the DF image of the NiCo sample prior to the electron diffraction sequence, with an overlay of segmentation boundaries. The same overlay is used in Fig. 2b, showing the distortion of features in the larger 128 by 128 grid orientation map. The reason for these discrepancies is sample drift, which occurred over the course of the lengthy scan. Compared to maps generated by the larger dataset, results from the efficiently segmented scan (Fig. 2d) were highly consistent, finding nearly identical crystal orientations for each grain common to the two sets. It should be noted, some unique orientation clusters were overlooked by the segmented scan, highlighting the need to carefully choose the number of diffraction images per segment. Examining the match of features in this map to the overlay generated by segmenting the respective DF image (Fig. 2c), good correspondence indicates negligible effects of image shift in this scan. Additionally, the exclusion of low intensity segments in the acquisition prevented unnecessary measurements where diffraction patterns did not emerge (voids in the sample), as confirmed by results for the larger scan. These findings show that scan sizes can be reduced by as much as 80%, while still retaining comparable accuracy and avoiding the drawbacks of extended scan times [6].

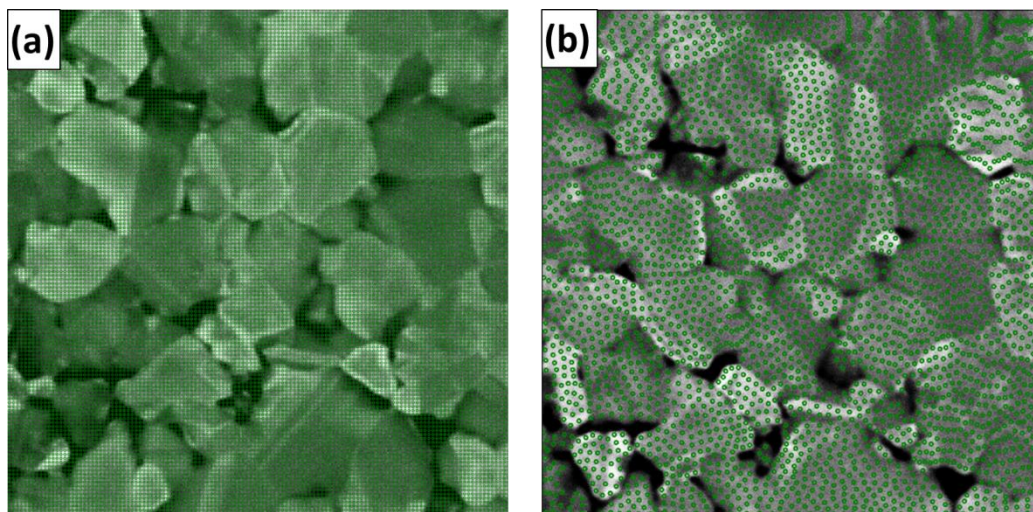


Figure 1. Difference between the two data collection schemes for crystal orientation mapping. (a) A uniform 128 by 128 mesh grid containing 16,384 diffraction data points which took around 3 hours to complete. (b) A non-uniformly spaced pattern after segmenting the image containing 3,157 data points which took around 30 minutes to complete.

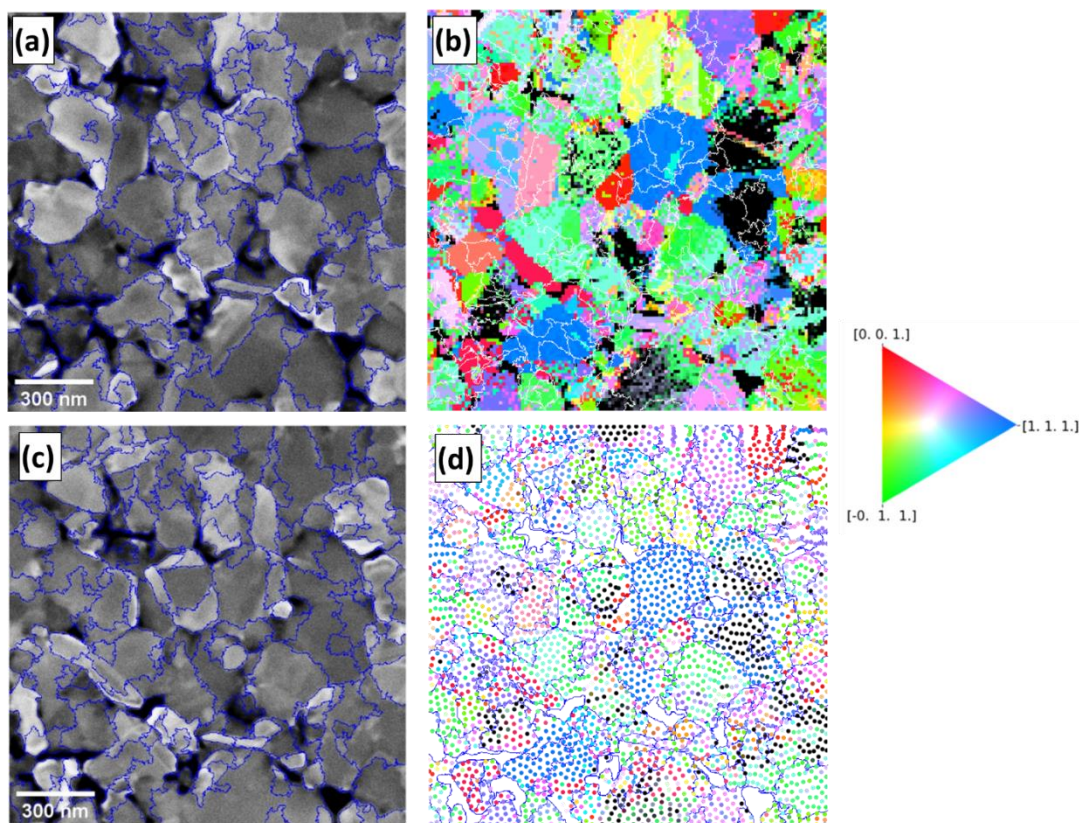


Figure 2. DF images captured before the start of each 4D-STEM acquisition and corresponding orientation maps, with segmentation overlays showing common boundaries. Orientation maps show alignment of the zone axis for each point with respect to [100], [110] and [111] directions; the legend on the right indicates the color coding of data points for these alignments. Black data points represent zero-correlation patterns, which were typically voids between grains or very thin sample regions where little diffraction occurred. (a) DF image taken before the start of the larger 128 by 128 scan. (b) Zone axis orientation map generated using 128 by 128 probe positions. The same segmentation lines in a) are overlaid. (c) DF image taken immediately following the first scan. Compared to a), a translation of features (downwards) can be seen, again supporting evidence of sample drift. (d) Zone axis orientation map created using the shorter 3,157 point scan with probe positions assigned by segmentation method. The same segmentation lines in c) are overlaid.

References:

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