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Accuracy, Reproducibility, and Calibration in 4D-STEM

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In four-dimensional scanning transmission electron microscopy (4D-STEM), a two-dimensional diffraction pattern is collected at every scan position of a two-dimensional raster of the electron beam. Calibrating these large datasets generally involves locating the origin of diffraction space, correcting any elliptical distortions, finding the diffraction space pixel size, and determining any rotational misalignment between real and diffraction space [1]. Errors in any of these calibrations can result in loss of precision and accuracy of varying severity. For instance, miscalibration of the origin or elliptical distortions will result in reduced accuracy and precision, while miscalibration of the real/diffraction space rotation can result in measurements of oriented quantities, such as strain [2] or flowlines [3], which are uninterpretable or incorrect. See Figure 1.

Additionally, algorithms for performing the various 4D-STEM calibrations are not universal: an algorithm which works on one dataset may fail entirely on another. Calibration of the origin, for instance, is comparatively simple for data with no beamstop and a thin specimen, but becomes more complicated with thick samples, and more complex still for data with a beamstop, necessitating entirely different algorithms in each case. Similarly, elliptical distortions can be calibrated any number of different ways. With a preponderance possible of algorithms and multiple necessary calibrations, standards are crucial to ensure that measurements are not only accurate, but reproducible.

Acquiring a 4D-STEM scan of a standard calibration sample, like polycrystalline gold or aluminum, ensures accurate and reproducible measurements. If a calibration scan is not acquired, calibrations are often performed using the experimental data itself, which may contain mixed phases, unknown structures, uncertain lattice parameters, or simply be poorly suited for calibration. Figure 2 shows the dangers of elliptical calibration using a complex experimental dataset, while Figure 3 shows the superiority of performing the same calibrations using a 4D-STEM scan of an Aluminum standard sample. Pixel calibrations are dependent on accurate assessment of the origin and elliptical distortions, and will suffer from inaccuracies in these prior calibrations – see Figure 4. We will discuss best practices for achieving accurate, precise, and reproducible measurements from 4D-STEM data, common pitfalls to avoid when performing calibrations, and demonstrate use of algorithms suited to calibrating various kinds of data using the open source python package py4DSTEM [4, 5].

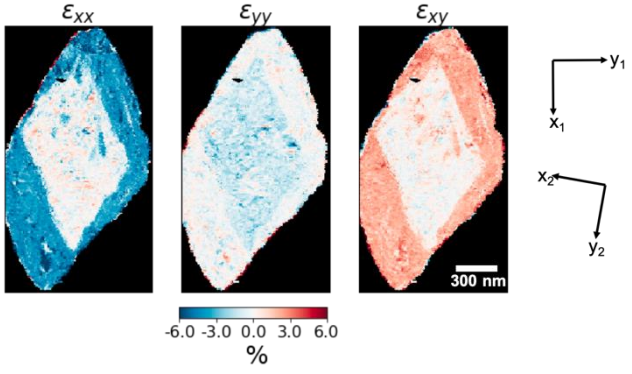

Figure 1. Strain map of Li_xFePO_4 nanoparticle [2] which can only be interpreted with proper rotational calibration. ε_{xx} and ε_{yy} represent tension and compression of the lattice along the x- and y-directions – but which directions are these? If the rotation between real and diffraction space has not been calibrated, the answer would appear to be x_1 and y_1, whereas in fact, the strain shown is along x_2 and y_2.

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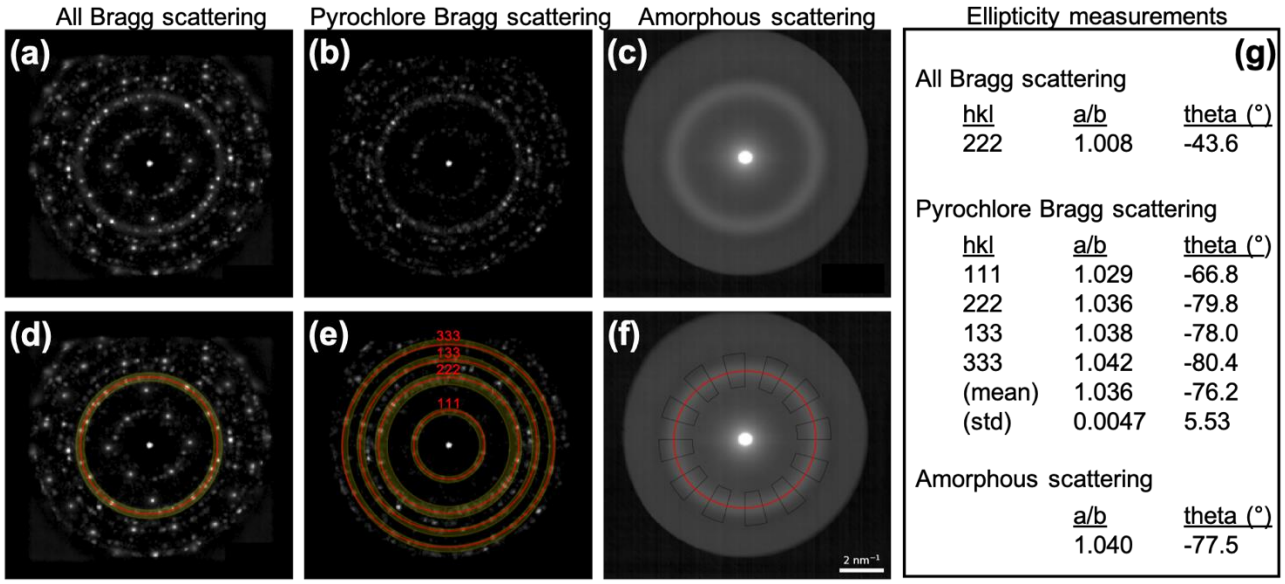

Figure 2. Elliptical calibration of a $GdTiO_3$ dataset containing a mix of a single crystal fluorite phase, polycrystalline pyrochlore phases, and amorphous phases [1], using several different methods. The data used for calibrations are shown in (a-c), the measurements are shown in (d-f), and the results are summarized in (g). 2D histograms of Bragg scattering positions for the entire dataset (a) and the polycrystalline pyrochlore regions only (b) are used to calibrate the data using the brightest ring (d) and using several different d-spacings (e). An averaged amorphous diffraction pattern is also used (c,f). The ground truth is unknown, however, the approximate consistency between the mean answer using the various pyrochlore rings and the amorphous data suggests these may be the most accurate. The answer using the brightest ring from all the Bragg scattering is almost certainly incorrect, and is also a likely result from a naïve attempt at calibration using this data.

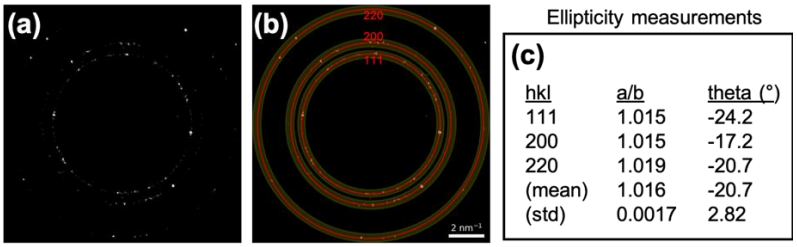

Figure 3. Elliptical calibration of a polycrystalline Aluminum standard dataset. A 2D histogram of Bragg scattering positions (a) is used to measure the elliptical distortion using each of the three available diffraction rings (b). The results (c) are highly consistent, lending confidence to the measurements.

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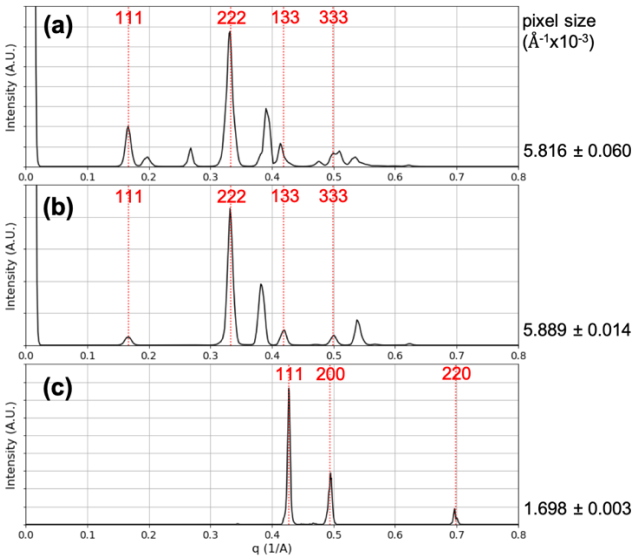

Figure 4. Detector pixel size calibrations depend on the accuracy of prior calibrations. (a) Pixel calibrations performed using (a) the $GdTiO_3$ elliptical calibrations from Fig. 2a,d, using (b) the $GdTiO_3$ elliptical calibrations from Fig. 2b,e, and using (c) the Aluminum calibrations shown in Fig. 3. The two datasets are unrelated, thus the different pixel sizes between (a,b) and (c) are expected; note, however, the significant improvement in precision in (c) over (a,b).

References:

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[2] HD Deng et al., Nature Materials (2022), p. 1.
[3] O Panova et al., Nature materials **18**(8) (2019), p. 860.
[4] https://github.com/py4dstem/py4DSTEM (accessed February 24, 2022)

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