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Recording and Using 4D-STEM Datasets in Materials Science

Colin Ophus[1], Peter Ercius[1], Michael Sarahan[2], Cory Czarnik[2] and Jim Ciston[1]

1. National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, Berkeley, USA
2. Gatan Inc., Pleasanton, USA

Traditional scanning transmission electron microscopy (STEM) detectors are monolithic and integrate a subset of the transmitted electron beam signal scattered from each electron probe position, shown schematically in Figure 1. These convergent beam electron diffraction patterns (CBED) are extremely rich in information, containing localized information on sample structure [1], composition [2], phonon spectra [3], three-dimensional defect crystallography [4] and more. Many new imaging modes become possible if the full CBED pattern is recorded at many probe positions with millisecond dwell times. Such a four-dimensional dataset would be comprised of a 2D CBED pattern at each point in a 2D STEM raster, hence the name 4D-STEM.

We have used a Gatan K2-IS direct electron detection camera installed on an uncorrected FEI Titan-class transmission electron microscope to record 4D-STEM probe diffraction patterns on a variety of samples at up to 1600 frames per second. The microscope voltage was set to 200 kV and the STEM probe convergence angle to 10.5 millirads. The high-angle annular dark field (HAADF) signal was recorded simultaneously using a monolithic Fischione detector. All data processing was performed using the Gatan Digital Micrograph software package and custom analysis code written in MATLAB.

As an example, a 4D-STEM dataset for a multilayer stack of epitaxial $SrTiO_3$ and mixed $LaMnO_3$-$SrTiO_3$ [5] is plotted in Figure 2. Figure 2A shows a HAADF micrograph of the multilayer along a (001) zone axis. Only the A sites (Sr and La) are visible in this micrograph and the composition can be roughly determined from the relative brightness. One possible 4D-STEM technique is position-averaged convergent beam electron diffraction (PACBED) described by LeBeau et al. [6]. We can easily construct ideal PACBED patterns by averaging the probe images over each unit cell fitted from Figure 2A, which is shown in Figure 2B. By matching these patterns to PACBED images simulated with the multislice method [7] we can precisely determine parameters such as sample thickness and composition, the latter of which is plotted in Figure 2C. For comparison, the composition has also been determined with electron energy loss spectroscopy (EELS) in a separate experiment, shown in Figure 3. The composition range of 0-85% $LaMnO_3$ measured by PACBED is in good agreement with the EELS measurements. In this talk we will demonstrate several other possible uses for 4D-STEM datasets [8].

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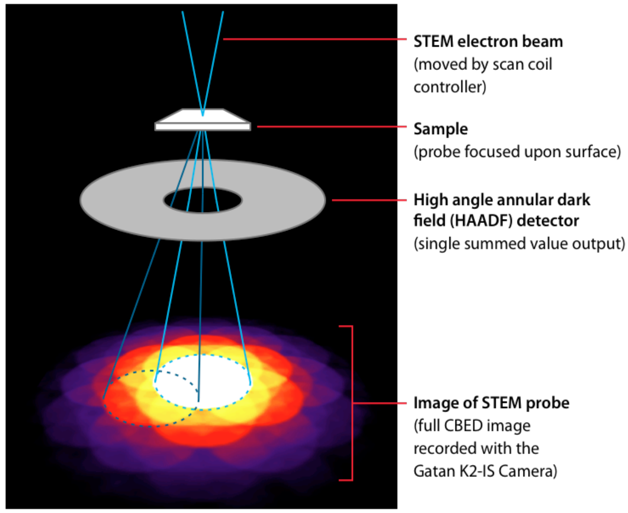

Figure 1. Experimental configuration of 4D-STEM experiments.

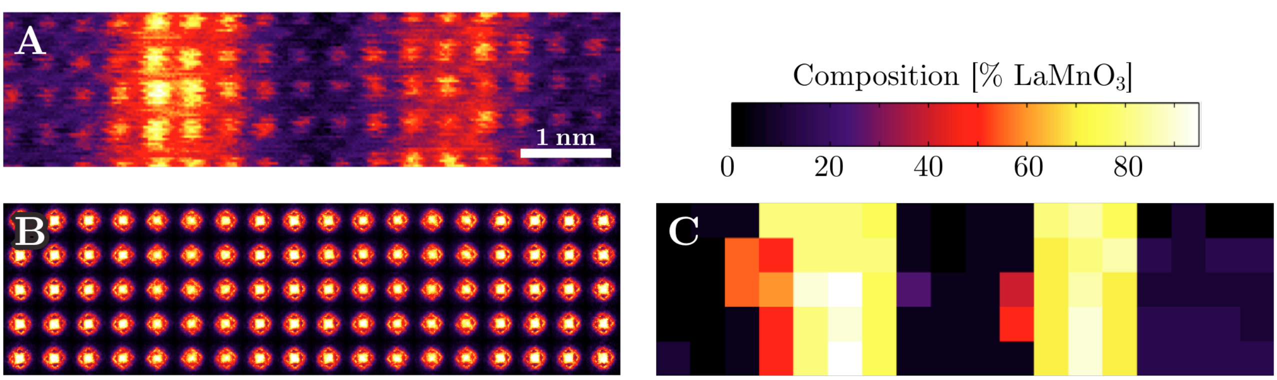

Figure 2. (A) HAADF micrograph of $SrTiO_3$-$LaMnO_3$ multilayers, generated from a 256 x 64 grid of probe positions. (B) Mean CBED patterns averaged over each unit cell from 4D-STEM dataset. (C) Composition map generated by PACBED pattern matching for each unit cell.

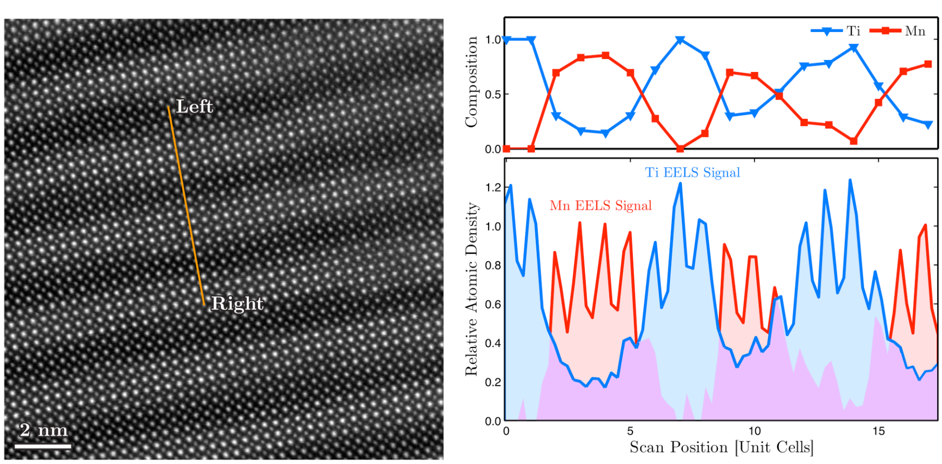

Figure 3. $SrTiO_3$-$LaMnO_3$ multilayer composition measured from EELS line scan of B sites.

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