

## Highly Accurate Real Space Nanometrology Using Revolving Scanning Transmission Electron Microscopy

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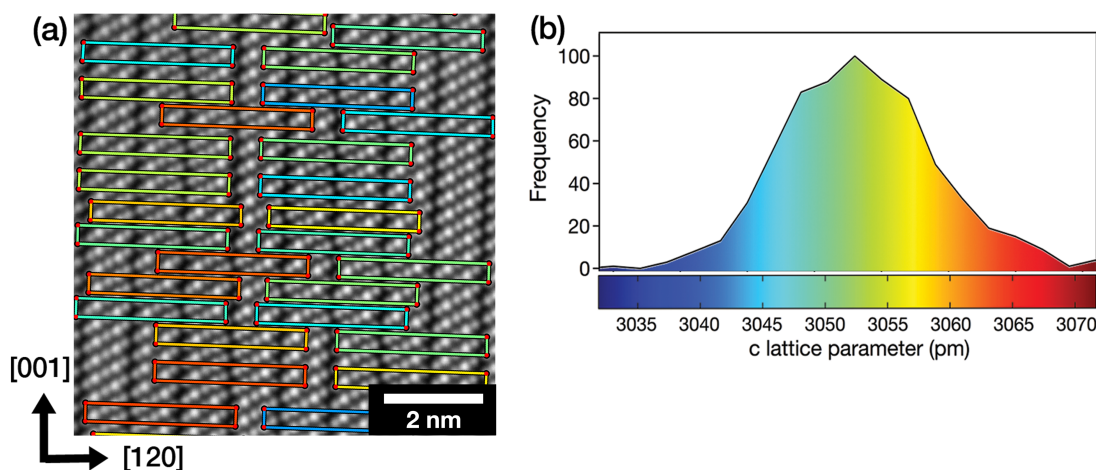
Accurately determining crystallography at the nanoscale provides key understanding of materials behavior. X-ray and neutron based diffraction methods provide highly accurate and precise measurements, but are typically limited in their application for nanoscale materials by poor spatial sensitivity. On the other hand, scanning transmission electron microscopy (STEM) is capable of spatial resolutions below an angstrom, making atomic scale analysis routine. Moreover, high-angle annular dark-field STEM produces images that are directly interpretable with intensities scaling to the atomic number and total number of atoms in a column [1-2]. While, real-space distance measurements are possible with STEM, the effects of thermal drift and scan distortion hinder *accurate* metrology.

In this talk, we will combine revolving STEM (RevSTEM) with a method for scan distortion correction to show accurate and precise real space length measurements for a nanostructured  $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$  alloy. We will show the effects of thermal drift can be corrected via measuring the drift parameters from multiple frames in an image series [3]. By using  $\langle 100 \rangle$  silicon as a reference standard, we correct the effects from distortions introduced from the scan system, which can then be used for imaging samples of unknown crystallography. The atom columns in drift corrected image series are then indexed and assigned to a matrix representation, which yields information such as the lattice parameters on a unit cell-by-unit cell basis, shown in Figure 1a [4]. To validate the accuracy of the technique, samples of pure  $\text{Bi}_2\text{Te}_3$  and  $\text{Bi}_2\text{Se}_3$  are analyzed using XRD and the real-space STEM imaging technique. In each case, the error is below 0.1 %. A nanostructured sample of unknown composition,  $\text{Bi}_2\text{Te}_{3-x}\text{Se}_x$ , is then investigated with errors again below 0.1 % between XRD and STEM. Further, we will show that the sample composition can then be determined to within 1 at% by Vegard's Law.

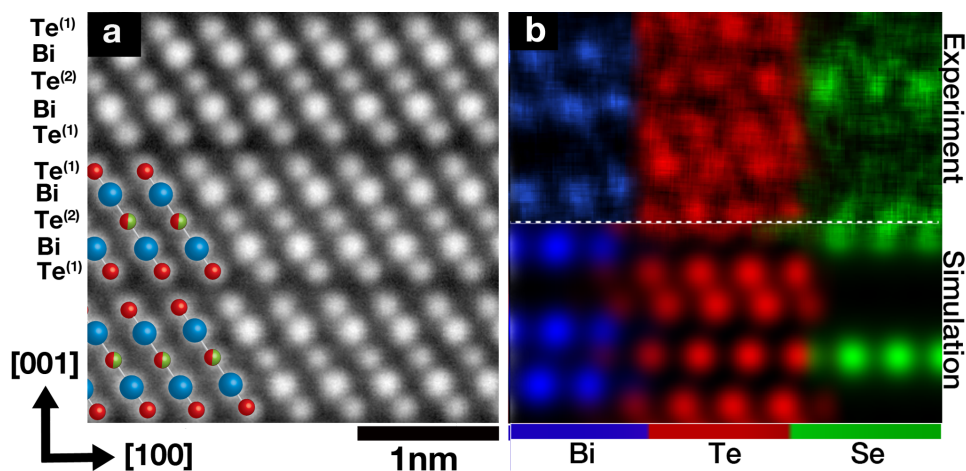
To relate structure and chemistry, atomic resolution EDS is then performed to determine the site-specific segregation of impurity atoms. The  $\text{Bi}_2\text{Te}/\text{Se}_3$  crystal structure consists of quintuple layers in the sequence Te(1)-Bi-Te(2)-Bi-Te(1) with a van der Waals interaction between Te(1) columns, shown in Figure 2a. It is observed that Se impurities reside in the Te(2) position in the structure, as shown in Figure 2b. We will then discuss how bond distances and atomic positions can be measured. Finally, we will discuss how the experimentally observed variation in the bond lengths and an anomalously large van der Waals gap can be explained using density functional theory (DFT) [5].

## References:

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 [5] JHD, WX, XS, and JML gratefully acknowledge support from the Air Force Office of Scientific Research (Grant No. FA9550-14-1-0182). SDF and LJA acknowledge support under the Australian Research Council's Discovery Projects funding scheme (DP140102538 and DP110102228). JHD acknowledges the National Science Foundation Graduate Research Fellowship (Grant DGE-1252376). The authors acknowledge the Analytical Instrumentation Facility (AIF) at North Carolina State University, supported by the State of North Carolina and the National Science Foundation.



**Figure 1.** (a) RevSTEM image series of pure  $\text{Bi}_2\text{Te}_3$  with boxes representing individual measured unit cells. (b) Histogram of the measured lattice parameters with the same color scale used for the box color in (a).



**Figure 2.** (a) RevSTEM image series of the nanostructured  $\text{Bi}_2\text{Te}_{2.7}\text{Se}_{0.3}$  with a schematic of the structure overlaid. (b) Experimental and simulated atomic resolution EDS showing segregation of Se to the Te(2) position.