doi:10.1017/S1431927613005047
Microsc. Microanal. 19 (Suppl 2), 2013

Viability of HAADF-STEM Imaging Contrast and Simulations as a Measure of B-site Ordering for Double Perovskites

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Spintronics has emerged as a promising technology that exploits both the intrinsic spin of the electron and it associated magnetic moment in a solid-state device. Utilization of the spin degree of freedom in metals and semiconductors has potential to create significant technological advances over current, charge-based technologies[1]. Therefore, accurate, atomic level characterization of half-metallic spin injectors and magnetic semiconductors will be essential to realizing these next generation technologies.

A significant barrier to this technological breakthrough was finding materials that exhibit ferromagnetic ordering and spin polarized transport above room temperature. One system that possesses two characteristics needed for practical spintronic applications, the A_2 B B' O_6 double perovskites (DP), has been studied extensively due to their high spin polarization (P) and Curie temperatures (TC) well above room temperature[2,3]. The fabrication of high-quality DP epitaxial films with high levels of B/B' ordering is essential to achieving desirable spintronic properties, historically however, fabrication and characterization has proven challenging. [4].

High-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) image contrast intensity is sensitive to atomic number (Z) and ideal for investigating local atomic arrangements. Therefore, the ability to collect quantitative HAADF images provides potential to collect both chemical and structural information with sub-Angstrom resolution in the time required for a single image acquisition[5]. Quantification of HAADF image contrast in terms of Z is not trivial, crystalline materials simulated along a zone axis requires separating out the effects of channeling, contributions of inelastic and elastic scattering, and the influence of thermal diffuse scattering and Debye-Waller factors[6]. Recent results show that even for single crystals, image simulations are required to correctly interpret HAADF-STEM image contrast[7].

For this work HAADF-STEM images of a Sr_2CrReO_6 ⟨001⟨ film were collected using a FEI probe-corrected HAADF-STEM Titan 80-300 microscope. Imaging along the ⟨110⟨ direction of Sr_2CrReO_6 reveals clear variation in column intensity with three distinct levels corresponding to independent atomic columns of Sr, Cr, and Re. The intensity of each atomic column is proportional to Z, for which Cr (Z = 24, darkest), Re (Z = 75, brightest), and Sr (Z = 38, intermediate) were identified. Image comparison and validation were performed against frozen-phonon based multi-slice simulation of a <110> SCRO with simulations results exhibiting intensity ratios as expected in the ratio of $\sim Z^{1.8}$. Simulations were also conducted of various B-site disorder conditions to compare against experimental image intensity variations. Results and comparisons of experimental images to simulated images for quantitative determination of B-site order/disorder will be discussed.

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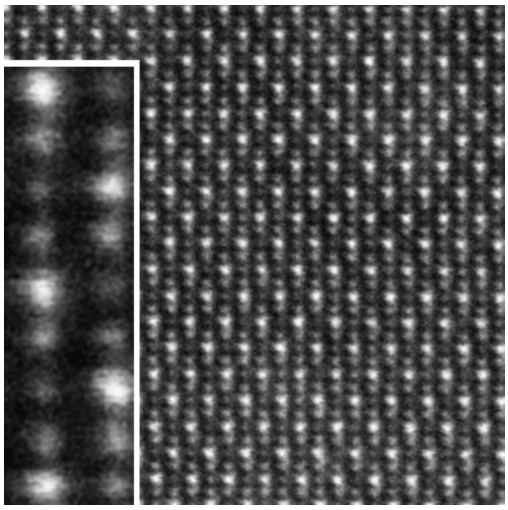

Fig 1: HAADF-STEM image of <110> SCRO exhibiting Z-contrast.

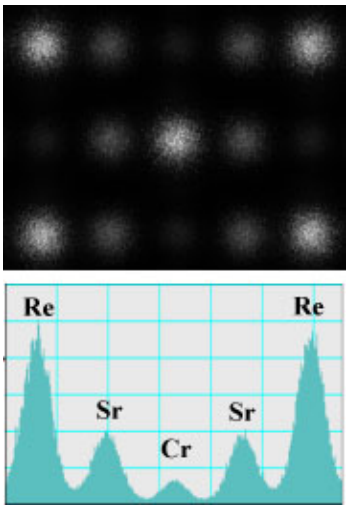

Fig 2: HAADF-STEM multislice image simulation of <110> SCRO showing ideal column intensity ratios.

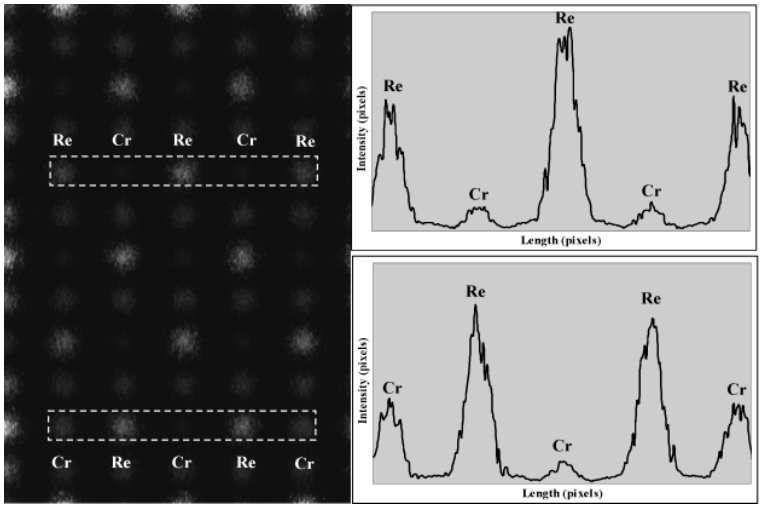

Fig 3: HAADF-STEM multislice image simulation of <110> SCRO showing non-stoichiometric column intensity resultant from B-site disorder.

https://doi.org/10.1017/S1431927613005047 Published online by Cambridge University Press