Interface Structures and Associated Magnetic Properties of Perovskite Oxide Thin Films Controlled by Substrate Symmetry

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In addition to well-recognized effects of strain and chemical composition, substrate symmetry can determine electronic and magnetic properties in epitaxially grown ABO₃ type perovskite oxide thin films [1]. Thus symmetry becomes a valuable tool which, by altering the network of interconnected oxygen octahedra, can be utilized to tune the properties of the thin films. The atomic-scale mechanisms of this influence, which are essential to be able to predict the behaviour of the new materials systems, is manifested primarily in the details of the interface structure and the local structural changes in the film. These changes can be very localized and thus not amenable to X-ray and neutron scattering methods. At the same time, aberration corrected scanning transmission electron microscopy (STEM) can provide direct localized information on the structural distortions in thin films and at the interfaces.

Here, we report direct observations of the interface atomic structures and local structural modulations in epitaxially-grown perovskite oxide thin films exhibiting substrate symmetry effect. We studied $La_{0.5}Sr_{0.5}CoO_3$ (LSCO) films grown on $La_{0.3}Sr_{0.7}Al_{0.65}Ta_{0.35}O_3$ (LSAT) and NdGaO₃ (NGO) substrates, which are cubic and orthorhombic, respectively, with nearly identical lattice parameter of a = 3.87 Å. However, from the result of polarized neutron reflectometry, the film on NGO shows much stronger magnetization interaction with polarized neutrons than the film on LSAT.

Fig. 1 shows ADF images of the LSCO thin film structure taken using ORNL's VG Microscopes HB603U equipped with Nion® aberration corrector and operated at 300kV. Both films show structural modulation in the direction normal to the substrate (can be linked with oxygen vacancy ordering and Co spin-state ordering in the LSCO films [2]); in the film grown on LSAT the modulation appears to be regular while in the film grown on NGO modulation exhibits multiple defects (Fig.1 c,d). When interatomic spacings are examined quantitatively from STEM images (Fig. 2), it can be seen that the modulations are similar in the two films, but the interface behaviour is different. For both films the lattice spacings show a smooth transition at the interface, for La/Sr (A) as well as for Co (B) sublattices, however, in LSCO on LSAT we also

observed a local increase in the ADF intensity on the B site near the interface (Fig. 2(c)) that was not present in LSCO/NGO film and is not strain-related. This implies complex interplay between structure, chemistry, and magnetic properties of the film. Column-resolved EELS data and octahedral tilt behaviour at the interface will also be discussed.

References

[1] J. He et al., *Phys. Rev. Lett.* **105** (2010) 227203.

[2] J. Gazquez et al., Nano Lett., dx.doi.org/10.1021/nl1034896.

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images of the LSCO thin film structures grown on the LSAT and NGO substrates, respectively. (c, d) The highresolution ADF images of the LSCO atomic structure near the interface for the corresponding areas marked as dotted squares in a and b, respectively (after maximum entropy deconvolution).

FIG. 2. Interface structure and lattice spacings: ADF images of (a) the LSAT-LSCO interface and (b) the NGO-LSCO interface. (c, d) Line profiles of the out-of-plane interatomic distances (A-A and B-B distances) and ADF intensity graphs for the B site (averaged over 20-25 atomic layers) across the LSAT-LSCO (c) and the NGO-LSCO (d) interfaces.