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Studying the effects of interfacial coupling in $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ thin films on $SrTiO_3$ using in-situ cooling experiments

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Hole-doped $La_xSr_{1-x}CoO_{3-\delta}$ has been attracted much attention due to its rich magnetic behavior and intriguing electronic transport properties [1]. At x=0.5, the heavily doped cobalt oxide not only exhibits a change in valence from Co^{3+} to Co^{4+}, which induces short-range ferromagnetic ordering, but also results in the formation of oxygen vacancies. In epitaxial thin films, the lattice mismatch between the substrate, such as $LaAlO_3$ (LAO) or $SrTiO_3$ (STO), the $La_xSr_{1-x}CoO_{3-\delta}$ film can stabilize oxygen vacancy ordering, which will have significant effects on the films' magnetism and electronic transport properties[2]. In addition, for films grown on STO, the anti-ferrodistortive phase transition at 105 K can further affect the structure and transport properties of the thin films. For example, $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ (LSCO) films grown on STO substrates have exhibited coupling between the out-of-phase TiO_6 octahedral rotation in STO at around 105 K and the interfacial CoO_6 octahedral network [3]. Furthermore, smaller Jahn-Teller distortions in CoO_6 make it possible for these coupling effects to propagate as far as 10 nm into the LSCO thin film and influence the magnetic/transport properties [4]. However, a detailed understanding of the local atomic and electronic structures, as well as the interplay between oxygen vacancy ordering and the low-temperature phase transitions at the atomic scale remains elusive.

In this contribution, we will combine in-situ cooling experiments with atomic-resolution STEM imaging and electron energy-loss spectroscopy (EELS) to study the local atomic and electronic structures of LSCO films grown on STO and LAO. LAO is used as a reference samples, due to the absence of a low temperature phase transition. The 12 nm thin LSCO samples were grown on STO (001) or LAO (001) substrate using molecular-beam epitaxial growth. We utilize the aberration-corrected JEOL ARM200CF, operated at both 200 kV and 80 kV, to acquire atomic-resolution high-angle annular dark field (HAADF) images and annular bright field (ABF) images, as well as EELS at room temperature and 90 K. The in-situ cooling experiments are conducted using a Gatan 636 liquid nitrogen cooling holder.

Oxygen vacancy ordering can be seen in LSCO films grown on STO at both room temperature and at 90 K. Figures 1a) and b) show a superlattice structure in the LSCO films that is oriented perpendicular to the LSCO/STO interface. This modulation in the CoO_2 column intensities is associated with oxygen vacancy ordering. Quantification of interatomic lattice spacing at room temperature and a 90 K shows that the lattice spacing for dark layer increases while it decreases for the bright layers (Figure 1c). ABF images along the (110) direction of the LSCO/STO interface (Figure 1d) demonstrate that the oxygen vacancy ordering structure is consistent with the brownmillerite structure [5]. The O *K*- and Ti *L*-edges of the STO substrate, as well as the Co *L*-edges of the LSCO films are shown in Figure 2. In STO, below 105 K, the oxygen *K*-edge pre-peak intensity increases reflecting the tilt of TiO_6 octahedral (Figure 2a). The corresponding O *K*-edges taken from LSCO show a shift and increase in the pre-peak intensity (Figure 2c) at low temperature; the Ti and the Co L-edge remain largely unchanged (Figure 2b and d). These changes in the EELS fine-structure will be correlated with the octahedral tilts and the corresponding structure symmetry breaking, as well as the electrostatic balance disruption at the STO/LSCO interface.

ABF imaging as well as electron magnetic circular dichroism (EMCD) measurements will be conducted to examine the impact of the interfacial oxygen octahedral tilts on the ferromagnetic ordering behavior [6].

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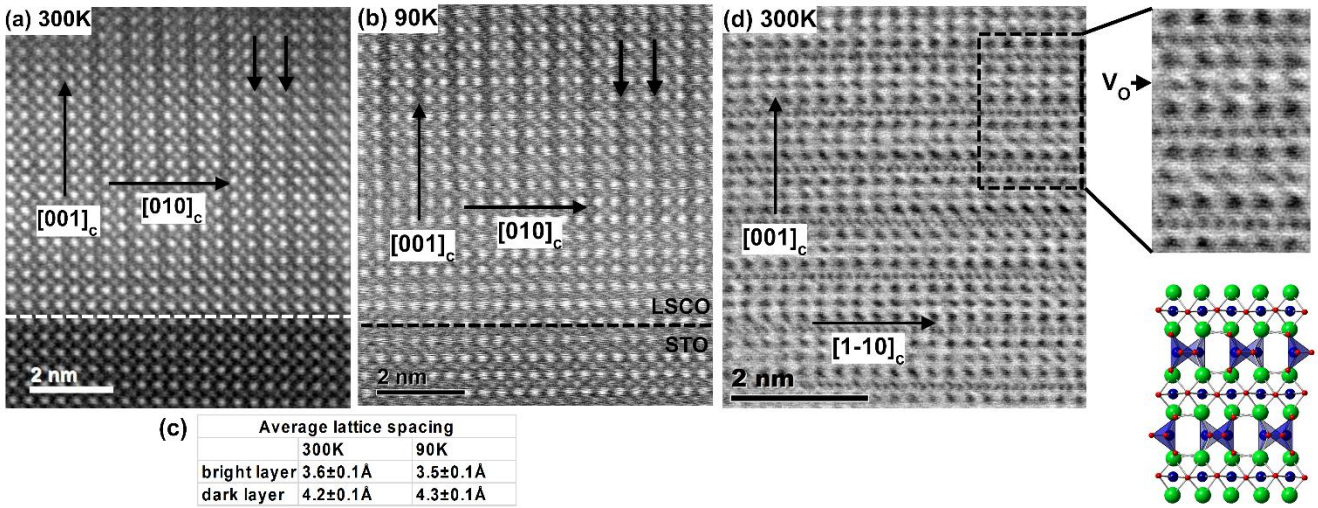

(c)

Average lattice spacing		
	300K	90K
bright layer	3.6±0.1Å	3.5±0.1Å
dark layer	4.2±0.1Å	4.3±0.1Å

Figure 1. (a) and (b) Representative HAADF image of the LSCO/STO interface viewed along the (100) directions 300K and 90K, black arrows mark the dark layer. (c) Average lattice spacing measurement. (d) ABF image of LSCO thin film along (110) at 300K, consistent with the brownmillerite structure.

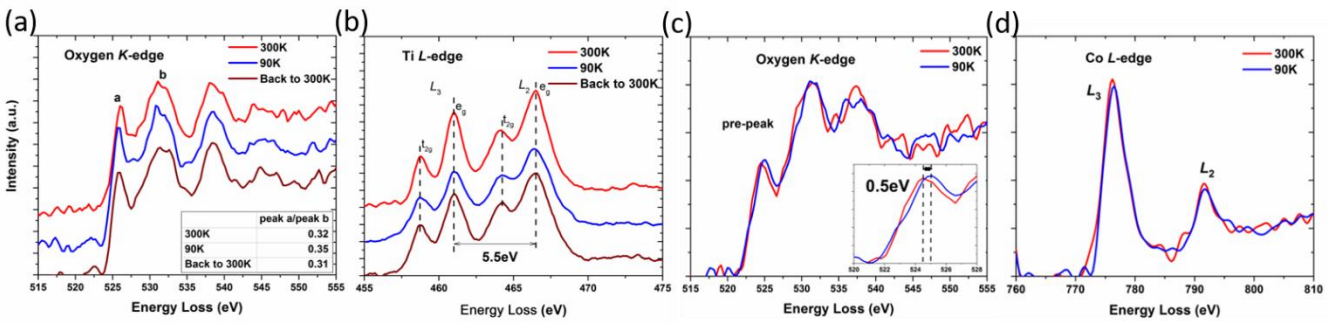

Figure 2. (a) and (b) Oxygen *K*-edge and Ti *L*-edge of the STO substrate above and below the anti-ferrodistortive transition temperature. (c) and (d) Oxygen *K*-edge and Co *L*-edge acquired from an area within 3.3nm from the interface.

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