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Atomic-Resolution Z-Contrast Imaging and EELS Study of Ferroelastic and Ferromagnetic Ordering in $LaCoO_3$

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The perovskite oxide $LaCoO_3$ has attracted increasing attention due to its multiple ferroic transitions including ferroelastic and ferromagnetic ordering. At room-temperature $LaCoO_3$ exhibits creep, which is usually only observed at temperatures close to a material's melting point, while strained $LoCoO_3$ thin films have been shown to be ferromagnetic at low temperatures. To advance our understanding of these unusual properties, a combination of electron diffraction, atomic-resolution Z-contrast imaging and electron energy-loss spectroscopy (EELS) has been used to study the $LaCoO_3$ microstructures as a function of applied strain and temperature. A ferroelastic material exhibits a hysteretic stress-strain behavior similar to the hysteresis found in ferromagnetic materials.[1] The coercive stress marks the point where the stress-strain relationship starts to deviate from the expected linear relationship. It has been suggested that the formation of ordered domains or defects occurs at the coercive stress. Therefore, we study $LaCoO_3$ samples compressed at room temperature above and below the coercive stress, and compare them with untreated samples. In the $LaCoO_3$ samples compressed above the coercive stress we find superlattice-domains with lattice constant $3a_0$ along the (100), (010) and (001) orientations (see Figure 1). These have been attributed to monoclinic distortions within the rhombohedral lattice.[2] In untreated $LaCoO_3$ and samples compressed below the coercive stress we do not find any superstructure, but twin boundaries due to the slight rhombohedral distortion of bulk $LaCoO_3$. In contrast to these results, it has been previously shown that epitaxial $LaCoO_3$ films grown in $LaAlO_3$ and $(LaAlO_3)(Sr_2AlTaO_6)$ undergo a ferromagnetic ordering transition at temperature close to 80 K.[3] We have used a combining of electron diffraction, atomic-resolution Z-contrast imaging, EELS and in-situ cooling experiment to show that the biaxial strain induced by the substrate on the $LaCoO_3$ film stabilized the intermediate Co^{3+}-ion spin state at low temperature.[3] Using energy-loss magnetic circular dichroism (EMCD) method,[4] we have further obtained angular-resolved EELS of Co L-edges at low- and room-temperature (Figure 2a).[3] We can show that there is a ferromagnetic transition in the $LaCoO_3$ thin film at low temperature, while neither our atomic-resolution Z-contrast image nor the electron diffraction pattern show any sign of a structural transition that can explain the observed magnetic ordering transition (Figures 2b and c).[3] Yet, some areas of the strained $LaCoO_3$ thin films exhibit superlattice domains at room temperature (Figure 3) similar to those of strained $LaCoO_3$ grains above the coercive stress (Figure 1). In this presentation, we will explore the relationship between the observed ferroelastic and ferromagnetic properties of $LaCoO_3$ and examine how the observed superlattice domains influence the mechanical and magnetic properties of highly strained $LaCoO_3$ bulk and thin films.

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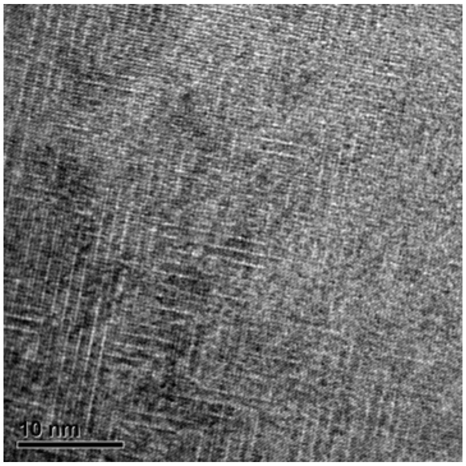

Fig 1: High-resolution phase contrast image of the superlattice domains in $LaCoO_3$ sample compressed at 110MPa at room temperature

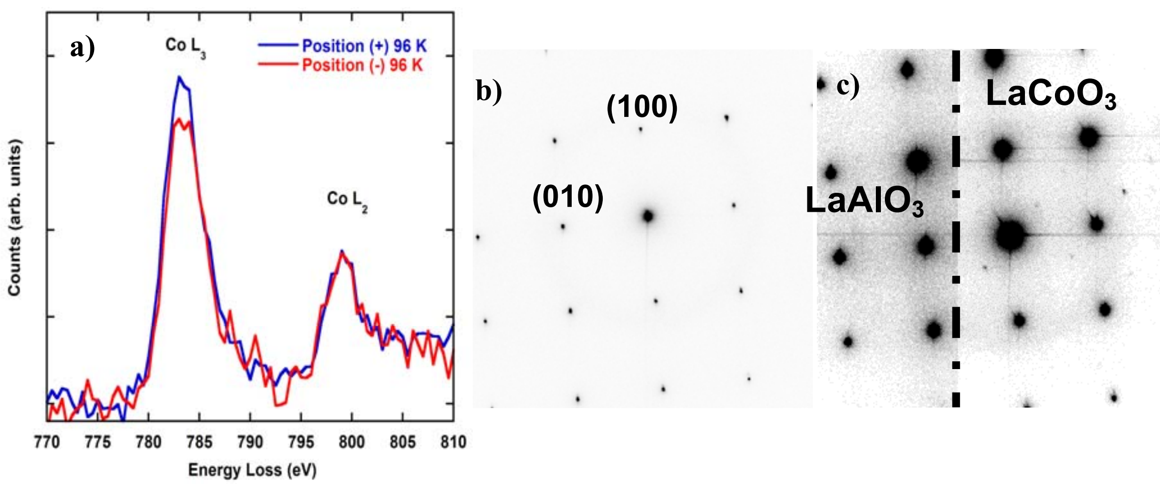

Fig 2: a) Angular-resolved EELS of epitaxial $LaCoO_3$ films grown in $LaAlO_3$ according to EMCD method at 94K; the electron diffraction pattern b) at room temperature and c) 94K

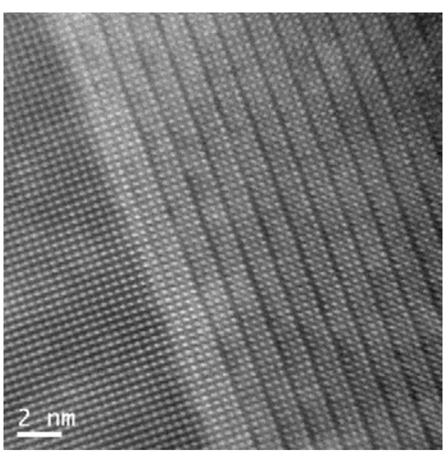

Fig 3: Atomic-resolution Z-contrast image of epitaxial $LaCoO_3$ films grown in $LaAlO_3$ showing a $3a_0$ superstructure.

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