doi:10.1017/S1431927618000818
Microsc. Microanal. 24 (Suppl 1), 2018

Direct Imaging of Tilt Relaxation from the Interface in Epitaxially Strained Ca_2RuO_4 Thin Films using ABF-STEM.

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The metal-to-insulator transition (MIT) temperature of 84℃ makes Ca_2RuO_4 (fig.1a) a strong candidate for post-CMOS electronic devices. This electronic transition is strongly coupled to, and driven by, the tilt and rotation of the RuO_6 octahedra in the bulk material [1]. Introducing an interface to the bulk can frustrate the distortions of the octahedral, altering the electronic properties. This frustration can lead to dead layers of unresponsive atoms near the interface. The smaller the device, the larger its volume fraction dominated by the dead layer. Knowing the length scale of the frustration is key to predicting the ultimate device size and performance. To answer this, we have grown thin film Ca_2RuO_4 on $NdAlO_3$, $NdGaO_3$, and $LaAlO_3$ substrates (fig. 1b) showing that tuning epitaxial strain leads to change in electronic properties such as the d-band bandwidth, electrical resistivity (fig. 1c), and the MIT temperature.

In this work we used Annular Bright Field (ABF) imaging on a 300keV Titan Themis Scanning Transmission Electron Microscopy (STEM) to directly locate the atom positions with picometer precision [2]. Image distortions were suppressed by cross-correlating stacks of ABF-STEM images (fig. 2a). We calculated the average projected Ru-O-Ru bond angles layer by layer to characterize the tilt relaxation and decay lengths as we move away from the strained interface (fig. 2b). We observed for the insulating -1.6% and +0.3% strained films that the tilts tended to relax monotonically as we moved further away from the strained interface. For the conducting -3.0% strain film, although the tilt angles oscillate from layer to layer, the average tilts tended to decay at a slower rate than the previous cases.

We also compared the flattening of the octahedra using the measured projected apical and basal Ru-O distances for the differently strained states (fig. 2c). As strain increases, the apical distances decreased while basal distances tended to increase. This agrees well with 4d-2p orbital overlap scheme because less overlap of the orbitals would lead to a more metallic state. Furthermore, rotation of the octahedra will be discussed in terms of detecting two different values for projected basal Ru-O bond lengths.

Strained thin films can relax when prepared as TEM cross-sections, depending on the thickness of the cross-section [3]. TEM sample preparation of anisotropically strained films can also result in curving of the film. This can generate different tilt relaxation trends across the interface depending on the sample thickness. ABF imaging is known for its high sensitivity to tilt and sample thickness [4]. To identify and eliminate specimen geometry dependent features from intrinsic features in the ABF signal we will explore the effects of sample thickness and the tilt relaxations across the interface and how it affects on ABF images [5].

References:

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[5] Funded by the Cornell Center for Materials Research Shared Facilities which are supported through the NSF MRSEC program (DMR-1719875) and NSF fellowship (DGE-1650441).

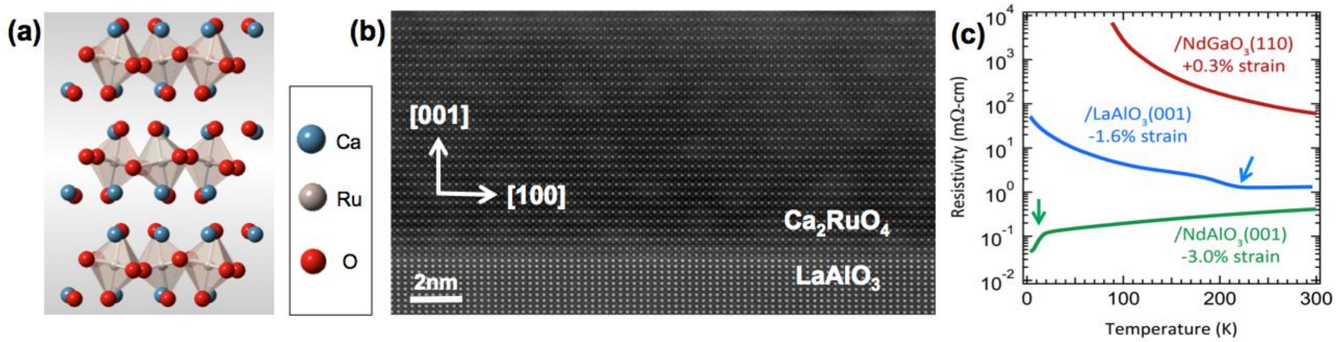

Figure 1. (a) Crystal structure of Ca_2RuO_4 in [010] zone axis showing the projected Ru-O-Ru tilts. (b) HAADF image showing uniformly grown Ca_2RuO_4 film on $LaAlO_3$ by Molecular Beam Epitaxy (MBE). (c) Electrical transport measurements on strained Ca_2RuO_4 thin films are shown as a function of temperature. Ca_2RuO_4 grown on $NdAlO_3$ shows metallic behavior with a kink at 17K. The MIT occurs at 230K for a -1.6% strain, and shifts up to 550K when strained to +0.3%. The bulk MIT temperature is 357K. (J. Ruf 2017, APS)

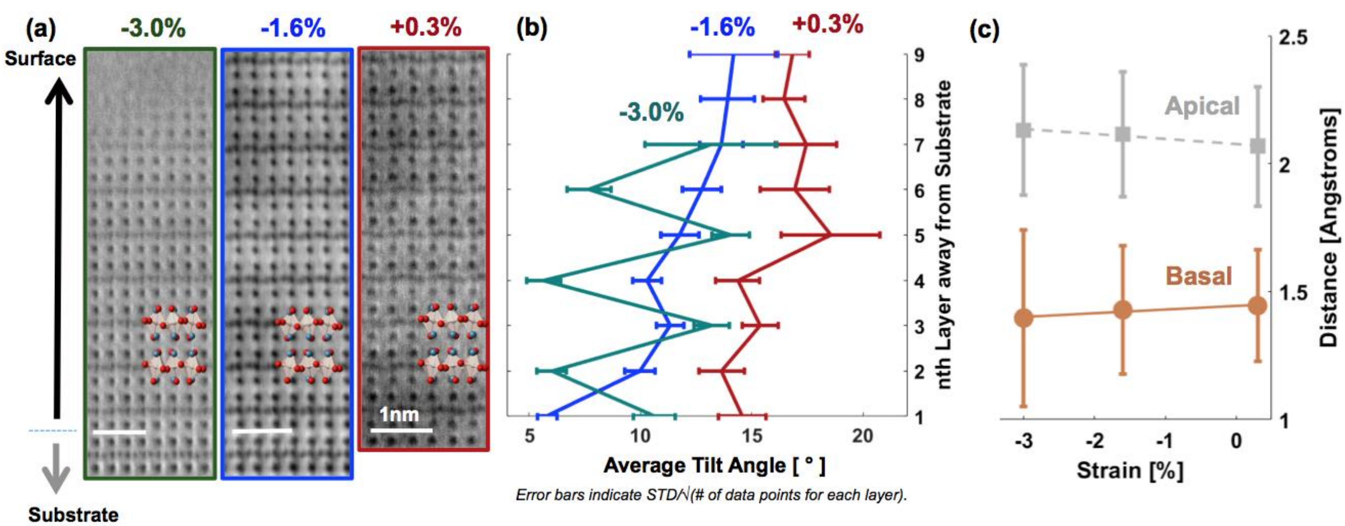

Figure 2. (a) Cross-correlated ABF images of each strained film states viewed at [010] zone axis are shown, taken under Titan in 300keV. (b) Average tilt angles are measured layer by layer, and are plotted in different colors for each of the different substrates. The data points are aligned with the layers shown in fig. 2a. (c) Measured apical and basal Ru-O distances are compared for three different strain states.