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In situ TEM Study of Ferroelectric Oxide Heterostructures

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Atomically engineered heterostructures of ferroelectric oxides are promising avenues to study rich interface physics and to design nonvolatile functionalities for future energy-efficient nanoelectronics. Towards designing such ferroelectric-based devices, not only achieving precise control of domains and domain walls, but also understanding interfacial charge-lattice coupling phenomena are essential [1]. *In situ* transmission electron microscopy allows to observe real-time domain switching and to probe atomistic lattice responses at ferroelectric/correlated oxide heterostructures. In this presentation, two *in situ* TEM experiments performed on ferroelectric-oxide-based heterostructures are discussed.

Rich correlated-electron physics in nickelates, including metal-insulator transition, charge and magnetic ordering, is due to intimate competition among electronic bandwidth, on-site Coulomb repulsion, charge transfer energy, and a wide range of structural variants [2]. Pursuing understanding and controlling the interplay between structural distortions and electronic correlations in nickelates, we investigated ferroelectric $PbZr_{0.2}Ti_{0.8}O_3$ (PZT)/$LaNiO_3$ (LNO, 5 unit cell) heterostructures grown on (001) plane of $SrTiO_3$ substrate using *in situ* electrical biasing and atomic resolution imaging [3]. Two distinct polarization states at the PZT/LNO interface were induced by *in situ* biasing (Fig. 1(a)). The presence of metallic LNO layer ensures uniform bidirectional bias fields within the PZT film, enabling complete domain switching from upwards (P_{up}) to downwards (P_{down}) polarization and vice versa. Atomic resolution images taken from areas with P_{up} and P_{down} at the interface show two distinctive atomic structures in ultrathin LNO (oxygen displacements and strain along the out-of-plane direction), as shown in Fig. 1(b-d), providing an important insight into the question on how structural distortions related to polarization across interfaces under various strain conditions affect the overall electronic properties of the ultrathin nickelates.

Unusually large electromechanical responses were reported in ferroelectric oxide bilayers consisting of two polymorphs, tetragonal $PbZr_{0.2}Ti_{0.8}O_3$ (T-PZT)/rhombohedral $PbZr_{0.55}Ti_{0.45}O_3$ (R-PZT) [4]. Using *in situ* electrical biasing in TEM (Fig. 2), we found the *ferroelastic* domain switching at the bilayer interface is the physical origin for enhanced electromechanical responses [5]. The ferroelastic domain switching occurs in a deterministic manner where the *c/a* domain wall orientation is rotated by 90^o, as shown in Fig. 2(b). Our atomic resolution imaging as well as phase field simulation revealed the triple junctions where the T/R-PZT interface meets the *c/a* domain wall are a prominent nucleation site for domain switching.

Our works show that *in situ* electrical biasing in TEM combined with atomic resolution imaging allows to directly observe unit-cell-scale charge-lattice coupling and their responses to the external electric fields, revealing the property-dictating structural information at the interfaces of ferroelectric oxide heterostructures [6].

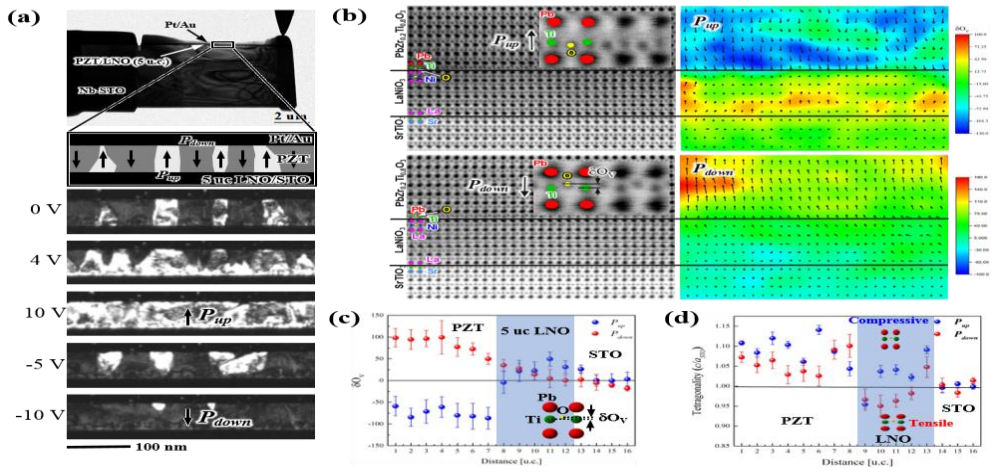

Figure 1. In situ domain switching and imaging of interfacial atomic structures at the PZT/LNO interface. (a) TEM and dark-field TEM images showing in situ domain switching in PZT thin films under various external biases. (b) Annular bright-field STEM images from the areas with downwards (P_{down}) and upwards (P_{up}) polarizations, respectively, and oxygen displacements maps. (c) Profiles of oxygen displacements and (d) tetragonalities across the interface.

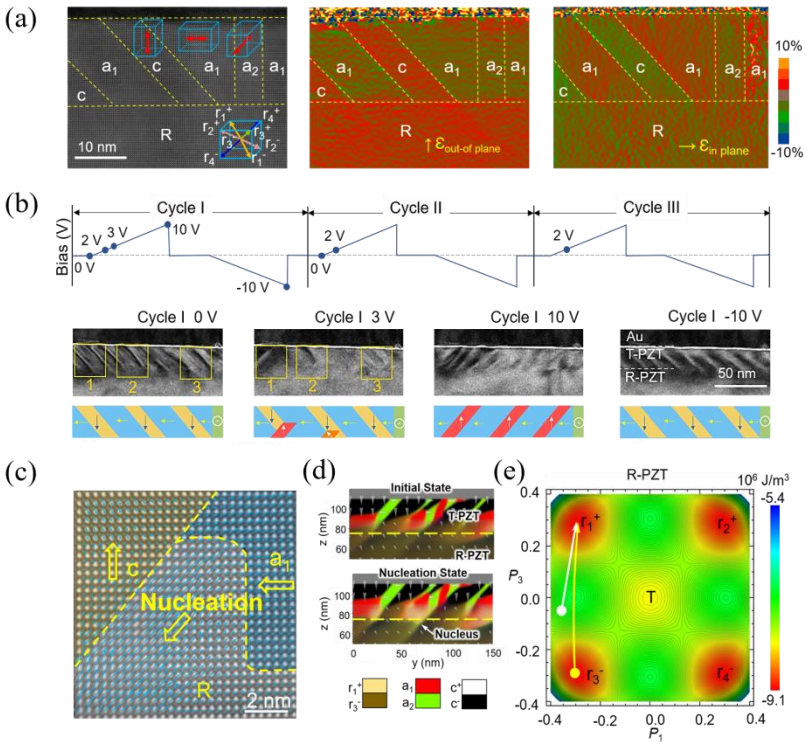

Figure 2. (a) STEM image of T/R-PZT bilayer and out-of-plane/in-plane strain maps. (b) TEM images in three voltage cycles. Schematics of domain structures are shown below each image. (c) HAADF STEM image showing the nucleation site near the interface. The Zr/Ti displacements (blue arrows) are overlaid. (d) Simulated domain structures at initial and nucleation states. (e) Energy landscape showing the switching paths, indicated with white arrows.

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