Advanced electron microscopy and spectroscopy on ferroelectric thin films

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We study the ferroelectric thin films by combing various advanced electron microscopy and spectroscopy, including the quantitative annular bright field (ABF) image analysis, *in situ* electrical biasing, and sub-10 meV resolution electron energy loss spectroscopy (EELS).

From quantitative ABF image analysis, we image the atomic structure of surface, interface, dislocations and grain boundaries with picometer precision and measure the polarization distribution around these defects to reveal the underlying screen mechanism of polarization bound charge. For example, we find that at the surface in $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT) thin films, the atomic structure strongly depends on the polarization underneath, i.e., there are six atomic layers with suppression of polarization at negatively poled surfaces, while no reconstruction exists at positively poled surfaces except the topmost atomic layer, and seven atomic layers with suppressed polarization exist at a 'neutral' surfaces in a ferroelastic domain [1]. At the bottom interface, interfacial layers with reduced polarization ranging from 3 to 6 unit cells in thickness exist, depending on the nature of polarization charge at the interfaces. The electric dipoles can even persist in the ultrathin PZT films, i.e., the residual polarization is about 16 μCcm^{-2} at 1.5-unit cell thick PZT film on bare $SrTiO_3$ (STO) and about 22μCcm^{-2} at 2-unit cells thick PZT film on STO with $SrRuO_3$ (SRO) electrode [2].

From the probe-based in situ TEM technique, we watch the domain switching with millisecond resolution and discover the interactions between the defects and domain walls. We observe the domain nucleation always starts from the interface between the electrode and ferroelectrics [3,4]. The domain wall motion can be strongly pinned by point defects in $BiFeO_3$ thin film [4] and weakly pinned by edge dislocations in PZT thin film [3]. The high-energy domain wall is not stable and spontaneous backstitching occurs after removal of the external electrical field. The switchingbility of ferroelastic domains in PZT thin film depends on their microstructure, i.e., the strength of interface clamping and dislocation pining, and active sites for nucleation.

From the 6 meV resolution EELS from monochromatic STEM, we measure the vibrational spectra from surface, bulk and interface, and band-gap at the surface, interface and other defects. I will also show our latest results of low-loss EELS measurements of ferroelectric thin films.

[1] P.Gao et al, Nature Communications 7, 11318 (2016).

[2] P.Gao et al, Nature Communications 8, 15549 (2017).

[3] P.Gao et al, Nature Communications 2, 591 (2011).

[4] C.T.Nelson, P.Gao et al, Science 334, 968 (2011).

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