Giant polarization sustainability in ultrathin ferroelectric films characterized by Cs-corrected transmission electron microscopy

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Ferroelectricity is generally deteriorated or even vanished when the ferroelectric films are downsized to unit cell scale, which hampers the potential minimization of ferroelectric-based electronic devices. To maintain and enhance the polarization in nanoscale ferroelectrics is of importance in both scientific and application aspects. Very recently, by tailoring the interface engineering, the polarization in $PbTiO_3$ layer can be greatly increased at $PbTiO_3/BiFeO_3$ bilayers grown on $SrTiO_3$ substrates, which attributes to the oxygen vacancy accumulation at $PbTiO_3/BiFeO_3$ interfaces.[1] In addition, oxygen vacancy is found to play an important role in improving the polarization in $BaTiO_3$ films grown on $SrTiO_3$ substrates as well.[2]

Here we report giant polarization sustainability in a series of ultrathin $PbTiO_3$ films scaled down to several unit cells grown on $NdGaO_3(110)$ substrates with $La_{0.7}Sr_{0.3}MnO_3$ as bottom electrodes. Atomic mappings via aberration-corrected scanning transmission electron microscopy demonstrate the robust ferroelectricity even for the film with the thickness of unit cell scale, as further confirmed by the piezoelectric response in the 3.6nm-thick film. For the 1.2 nm thick film, the calculated polarization reaches ~50 μC cm^{-2}. The 2 nm-thick film possesses a polarization as high as the bulk value. The films ranging from 10nm to 35nm thick display a giant out-of-plane lattice parameter, which corresponds to a polarization of 100μC·cm^{-2}, 20% larger than that of the bulk $PbTiO_3$. The giant polarization sustainability in the present films is proposed to result from the charge transfer at the $La_{0.7}Sr_{0.3}MnO_3/PbTiO_3$ interface, as supported by the anomalous decrease of Mn valence measured from X-ray photoelectron spectroscopy. This present study reveals the significant role of charge transfer to enhance the large polarization in ultrathin ferroelectrics particularly in the films below 10nm, which will be very helpful for the development of future electronic devices.

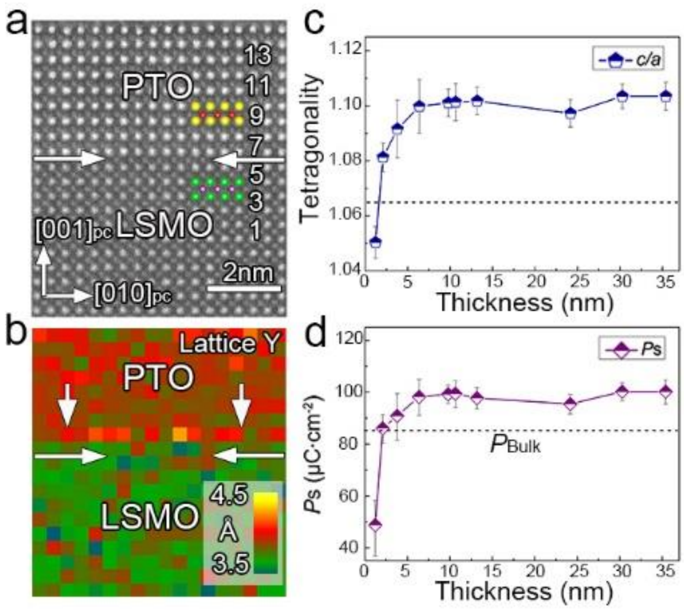

Figure 1. (a) Atomically resolved HAADF STEM image of $PbTiO_3/La_{0.7}Sr_{0.3}MnO_3$ interface; (b) Strain distribution along the interface; (c) A plot of tetragonality vs thickness of $PbTiO_3$ layer; (d)A plot of calculated polarizations as a function of $PbTiO_3$ film thickness.

References [1] Y. Liu, Y.L. Zhu, Y.L. Tang et al., *Nano Lett,* 17, 3619-3628 (2017).

[2] Q. Qiao, Y. Zhang, R. Contreras-Guerrero, R. Droopad, S. T. Pantelides, S. J. Pennycook, S. Ogut, R. F. Klie, *Appl. Phys. Lett.* 107, 201604(2015).