Understanding Polarization Asymmetry and Precise Tuning of the Built-in Bias in $PbTiO_3$ based Superlattice Thin Films

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The artificial layering in ferroelectric superlattices is a potential source of polarization asymmetry, where one polarization state is preferred over another. One manifestation of this asymmetry is a built-in electric field associated with shifted polarization hysteresis loops and capacitive butterfly curves. While the built-in bias maybe generated from inhomogeneous strain through artificial layering or other composition gradients, it may also be influenced by extrinsic effects such as asymmetric electrodes or the inhomogeneous distribution of defects, such as oxygen or lead vacancies. Due to all the intrinsic and extrinsic factors mentioned above, it is typically very difficult to decisively identify the source of polarization asymmetry in any given sample or material system.

In this work, we comprehensively studied the origin of polarization asymmetry for ferroelectric $PbTiO_3$ based superlattice films. Using off-axis RF-magnetron sputtering, we prepared several compositions of $PbTiO_3/SrTiO_3$ (PTO/STO) superlattice thin films; and for comparison $PbTiO_3/SrRuO_3$ (PTO/SRO) superlattices, which have an additional intrinsic compositional asymmetry due to A-site B site variations at the interface. All samples were deposited with in-situ symmetric $SrRuO_3$ top and bottom electrodes to neglect the influence electrodes may have on the built-in potential. Through theoretical modeling and experiments such as heat treatment and repetitive electrical cycling on the samples, we were able to identify and study the significance of Pb-O vacancy defect dipoles on the built-in electric field. In addition, we were able to detect the presence of oxygen vacancies at the PTO/STO interfaces in the superlattice with STEM-EELS. Finally, we show the ability to tune this built-in bias by depositing a hybrid superlattice that combines PTO/STO and PTO/SRO superlattices. By tuning the composition of the PTO/STO/PTO/SRO superlattice, the built-in bias in this combo-superlattice can be reduced to zero.

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