Electrical Transport and Magnetic Properties of PbTiO₃/SrRuO₃ superlattices

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Artificially structured oxides present exciting opportunities to design materials with specified or novel properties [1,2]. With advanced epitaxial growth techniques, multicomponent perovskite heterostructure with desired dielectric, ferroelectric and magnetic behavior can be achieved experimentally. Currently, we have successfully fabricated PbTiO₃/SrRuO₃ (PTO/SRO) superlattices with ultra thin SrRuO₃ layers through RF magnetron off-axis sputtering [3]. The superlattice thin films (100nm) with top and bottom SrRuO₃ electrodes were deposit on a TiO₂ terminated SrTiO₃ substrate. The structure and quality of the films were examined through transmission electron microscope (HRTEM), X-ray diffraction (XRD) and atomic force microscopy (AFM). In addition, the electrical properties were obtained using a home-built probe station, and the magnetocapacitance measurement of the samples is done with a superconducting magnet system.

Here, our samples show a large anisotropy in their electrical resistvity, which can be tuned by changing the $PbTiO_3$ layer thickness. Along the out of plane direction, the samples demonstrate ferroelectricity and tunneling current characteristics that confirm the metallicity of the SrRuO₃ layers but can also act as a novel dielectric component. From the HRTEM cross-sectional image, Figure 1(a), we show evidence of thin SRO layers within the superlattice and that good quality is achieved throughout the sample. One important characteristic of our superlattices is the compositional breaking of inversion symmetry. The superlattice system of PbTiO₃/SrRuO₃ has both A site and B site variation that can break the inversion symmetry, as seen in Figure 1(b), resulting in a preference for a particular polarization direction. Perovskite-oxide superlattice systems demonstrating broken inversion symmetry have been both theoretically and experimentally studied in recent years. However, experimentally the systems are mostly deposited with three different materials to achieved symmetry breaking [4]. Figure 2(a) shows the calculated double well potential for LDA (local density approximation). To compare structures with a different composition, the energy is plotted in energy per unit cell instead of total energy. When the PTO layer is around 7 unit cells or less, the potential double-well is asymmetric and indicates a favored polarization state. Once the PTO layer increases to 9 unit cells and above, the PTO becomes the dominant phase and the system is not affected by the breaking of inversion symmetry, so the potential double-well remains symmetric.

In this work, we studied the impact of the compositionally broken inversion symmetry and magnetic field on the capacitance-voltage characteristic of our superlattices. As seen in Figure 2(b), the 5/1 and 7/1 PTO/SRO samples, the effect of the compositional inversion symmetry breaking is significant and the two peaks in the dielectric constant butterfly loop shifts to the right. Once the PTO layer thickness increases to 9 unit cells or more, the PTO becomes dominant and the system is a conventional ferroelectric material with the butterfly loop centered close to 0V. To investigate potential magnetoelectric coupling, we have applied a magnetic field to our samples at low temperature while measuring the dielectric constant. Acknowledgement: This work was funded by NSF DMR1334867.



Figure 1. (a) HRTEM cross-sectional image of 8/1 PTO/SRO superlattice. (b)Superlattice interface of only A site changes (left) and where both the A and B site changes (right) resulting in the breaking of inversion symmetry.



Figure 2. (a)) Total energy of several PbTiO3/SrRuo3 superlattices plotted as a function of polarization. (b) Comparison of dielectric constant between different PTO/SRO superlattices.

Reference

C. H. Ahn, K. M. Rabe and J.-M. Triscone, "Ferroelectricity at the Nanoscale: Local Polarization in Oxide Thin Films and Heterostructures," *Science* **303**, 488 (2004).
M. Dawber, K. M. Rabe and J. F. Scott, "Physics of thin-film ferroelectric oxides," *Rev. Mod. Phys.* **77**, 1083 (2005).

[3] S.J. Callori, J. Gabel, D. Su, J. Sinsheimer, M.V. Fernandez-Serra and M. Dawber, "Ferroelectric *PbTiO3/SrRuO3* Superlattices with Broken Inversion Symmetry," *Phys. Rev. Lett.* **109**, 067601 (2012).

[4]H.N. Lee, H.M. Christen, M.F. Chisholm, C.M. Rouleau and D.H. Lowndes, "Strong polarization enhancement in asymmetric three-component ferroelectric superlattices," *Nature* **433**, 395 (2005).