

Role of interface termination in SrRuO₃/PbTiO₃/SrRuO₃ capacitors under epitaxial strain

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Ferroelectric devices have seen large advances in terms of speed, density and resistance to degradation allowing them to potentially become a standard for non-volatile memory¹. A recent study² has looked at ultrathin films of SRO/PTO mainly in terms of total energy and Schottky barriers, however we believe not everything is yet fully understood about these systems.

In this work we use density functional theory (DFT) within the local density (LDA) and local spin density with spherically averaged Hubbard U (LSDA+U) approximations using numerical atomic orbitals implemented in the SIESTA³ code. The systems under consideration are symmetrically terminated SRO/(PTO)_m bulk and SRO/(PTO)_m/SRO slabs, where m is an integer. There are two possible geometries, either we have RuO₂-SrO-RuO₂/(PbO-TiO₂)_m-PbO/[RuO₂-SrO-RuO₂] or SrO-RuO₂-SrO/TiO₂-(PbO-TiO₂)_m/[SrO-RuO₂-SrO], as illustrated in Fig. 1, where the term in the square brackets is absent from the supercell of the bulk system. We varied the amount of PTO unit cells from 2 and 4, the supercell area, A , from 1×1 to $\sqrt{2} \times \sqrt{2}$ and the in-plane lattice constant ($a = b$) from 3.87 Å to 3.67 Å.

For open circuit boundary conditions (OCBC) a dipole moment, for the whole SRO/PTO/SRO system, develops when the lateral lattice constant, a , is 3.67 Å and 3.77 Å for SrO and RuO₂ terminated surfaces, respectively (Fig. 2a). This result can be understood from the fact that when the surface is terminated by RuO₂ rather than SrO, there are twice as many metallic states to provide better screening. The exception to this is RuO₂ terminated surface with parameters $A = \sqrt{2} \times \sqrt{2}$ and $m = 2$, where not only does a polar order appear at increased strain, but also disappears as additional strain is introduced.

For closed circuit boundary conditions (CCBC), we measure Δ_{norm} a parameter that is zero when no polar order is present and unity when the polar order is equal to that of bulk

PTO with the same a . Even with perfect screening the development of polar order with respect to a coincides with OCBC, with interfaces terminated by RuO_2 exhibiting a larger polar order (Fig. 2b). We believe the explanation lies in the interfacial layer, PbRuO_3 , which plays a role in driving the minimization of the total energy (similar to the anomaly in the OCBC case) due to the fact that it is under the largest compressive strain compared to other layers.

Our analysis also includes the effects of interfaces on structural relaxations, spin polarization, metal-induced gap states and generalized capacitance (a different measure of instabilities due to polar order).

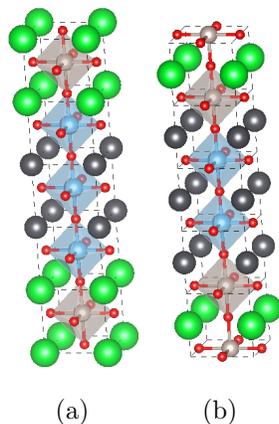
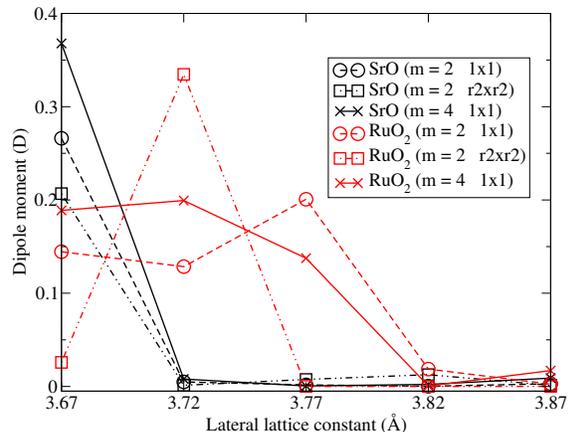
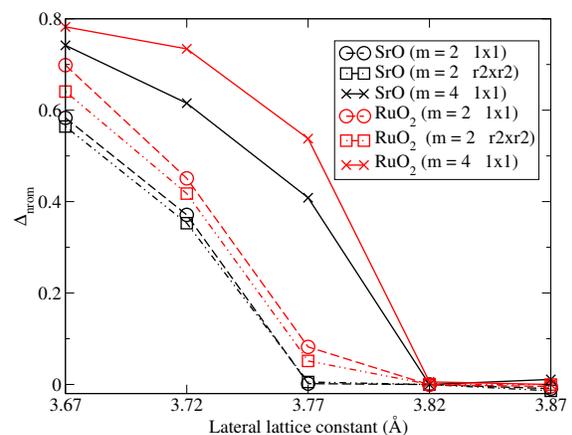


FIG. 1: Supercell of the slab for $\text{SRO}/(\text{PTO})_2/\text{SRO}$ when the termination plane is either (a) SrO or (b) RuO_2 . Green, grey, brown, blue and red correspond to Sr, Pb, Ru, Ti and O, respectively. The images were generated using VESTA⁴.



(a)



(b)

FIG. 2: Calculations of the polar order in (a) open and (b) closed circuit boundary conditions. Black (Red) refers to SrO (RuO_2) interface terminated.

¹ J. F. Scott and C. A. P. de Araujo, *Science* **246** (1989), 10.1126/science.246.4936.1400.

² Y. Umeno, J. M. Albina, B. Meyer, and C. Elssser, *Phys. Rev. B* **80** (2009), 10.1103/PhysRevB.80.205122.

³ J. M. Soler, E. Artacho, J. D. Gale, A. García, J. Junquera, P. Ordejón, and D. Sánchez-Portal, *J. Phys. Condens. Matter* **14** (2002), 10.1088/0953-8984/14/11/302.

⁴ K. Momma and F. Izumi, *J. Appl. Crystallogr.* **44** (2011), 10.1107/S0021889811038970.