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Electronic Supplementary Information for

Electric Field Induced Reversal of Spin Polarization, Magnetic Anisotropy and Tailored Dzyaloshinskii-Moriya Interaction in Underoxidized $SrRuO_3/SrTiO_3$ Heterostructures

Zengjie Li, Xiang Liu, Jiawei Jiang, Wenbo Mi,* Haili Bai*

Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparation Technology,

School of Science, Tianjin University, Tianjin 300354, China

*Author to whom all correspondence should be addressed.

E-mail: miwenbo@tju.edu.cn (W.B. Mi) and baihaili@tju.edu.cn (H.L. Bai)

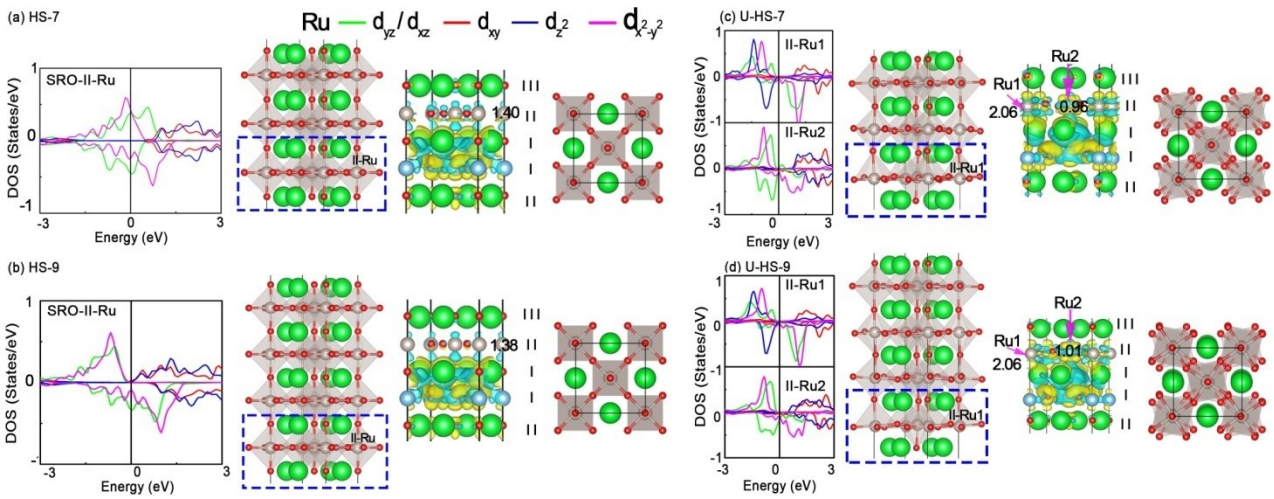

Fig. S1 (a-d) orbital-resolved density of states (DOS) of interfacial Ru atoms, the side view of Ru-O octahedral, charge density difference (isosurface value 0.002 e Å^{-3}), and the top view of Ru-O octahedral in $SrRuO_3$ (SRO)/$SrTiO_3$ (STO) heterostructures with 7 SRO atomic layers at pure (HS-7) and underoxidized (U-HS-7) SRO interface. The yellow and blue areas represent the accumulation and depletion of electrons, respectively.

As shown in Fig. S1, in the underoxidized SRO interface, the Ru-O octahedral structure of the interfacial Ru1 atoms is significantly destroyed. The Ru-O bond length is compressed in the *xoy* plane, which decreases the energy level of d_{z^2} orbital and increases the energy level of d_{xy} orbital. Therefore, by comparing to the pure interface (see Fig. S1(a) and (b)), in the underoxidized SRO interfaces, the spin-up Ru1 d_{xy} orbital near Fermi level moves to a higher energy level and the d_{z^2} orbital changes from the unoccupied state to occupied state (see Fig. S1(c) and (d)). In the underoxidized SRO interface, the charge transfer mainly occurs around the O atoms that have not been removed (Fig. S1(c) and (d)). Meanwhile, the Ru1 and Ru2 atoms exhibit different magnetic moment in the underoxidized SRO interface, which can be mainly ascribed to the redistribution of d_{xz}/d_{yz} and $d_{x^2-y^2}$ orbitals.

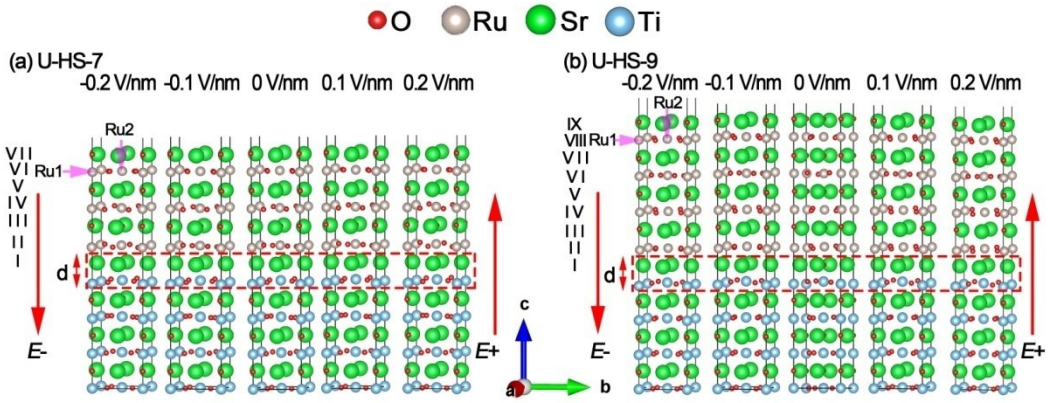

Fig. S2 Optimized lattice structure of underoxidized SRO/STO heterostructures with (a) 7 (U-HS-7) and (b) 9 SRO atomic layers (U-HS-9) at different electric fields. The up and down arrows represent the direction of the positive and negative electric fields, respectively.

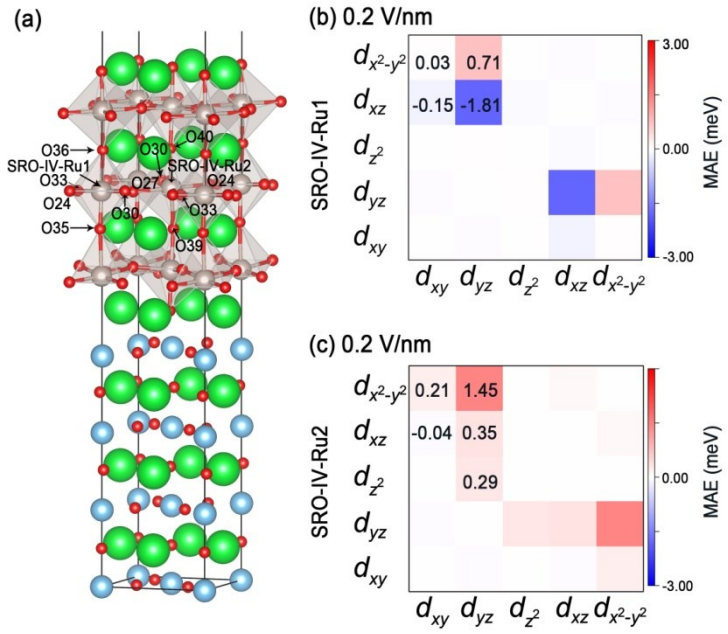

Fig. S3 (a) Schematic diagram of Ru-O octahedron structure in the U-HS-7 model. (b-c) Orbital-resolved MAE of Ru atoms in the U-HS-7 model at 0.2 V/nm.

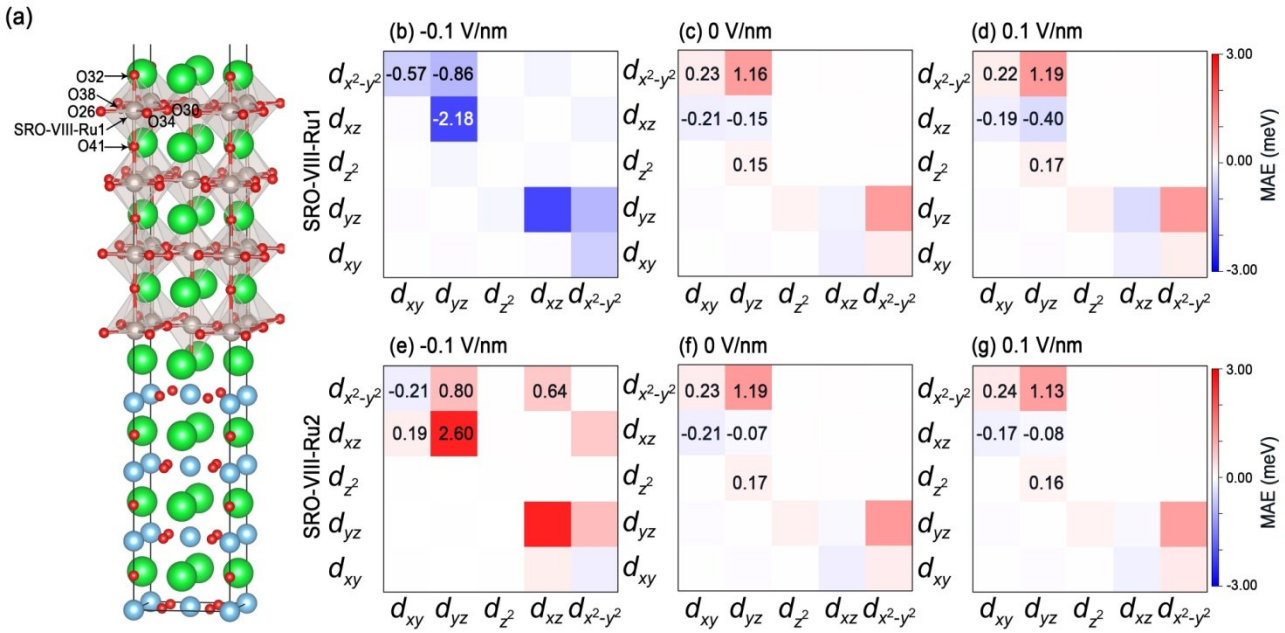

Fig. S4 (a) Schematic diagram of Ru-O octahedron structure in the U-HS-9 model. (b)-(g) Orbital-resolved MAE of Ru atoms in the U-HS-9 model at different electric fields.

Table S1 Magnetic moments (μ_B) of the Ru1/Ru2 atom (M_{Ru}) in U-HS-*X* (*X*=7, 9) model at different electric fields (*E*). II, IV, VI and VIII represent the RuO_2 layer number of SRO in U-HS-7 and U-HS-9 models, as labeled in Figure S2.

		RuO_2-Layer (M_{Ru1}/M_{Ru2})			
Model	*E*	II	IV	VI	VIII
	-0.2	2.13/0.62	-1.59/-1.64	-1.44/-1.54	
	-0.1	2.10/1.03	1.58/1.88	-1.38/-1.36	
U-HS-7	0	2.06/0.96	1.59/0.62	1.45/1.42	
	0.1	2.09/1.09	-1.83/-1.42	-1.46/-1.11	
	0.2	2.10/0.45	-1.72/-1.41	1.40/1.49	
	-0.2	2.05/0.99	1.74/1.43	-1.57/-1.56	-1.45/-1.44
	-0.1	2.06/1.02	1.65/1.66	1.48/1.43	1.48/-0.50
U-HS-9	0	2.06/1.01	1.72/1.51	1.57/1.52	1.44/1.43
	0.1	-2.07/-0.97	-1.76/-1.19	-1.57/-1.53	-1.45/-1.41
	0.2	-1.98/-1.17	-1.61/1.44	1.31/-1.77	1.68/-0.30

Table S2 The spin polarization of U-HS-7 and U-HS-9 models at different electric fields.

Electric Field	-0.2 V/nm	-0.1 V/nm	0 V/nm	0.1 V/nm	0.2 V/nm
U-HS-7	-46.86%	-13.28%	-40.34%	32.23%	34.65%
U-HS-9	-52.63%	-39.26%	-73.01%	55.65%	27.57%

Table S3 The Ru-O bond length of the (SRO-IV-Ru1)-O and (SRO-IV-Ru2)-O octahedral structure in the U-HS-7 model with different electric fields. The schematic diagram of lattice structure of U-HS-7 model is shown in Fig. S3(a).

-0.2 V/nm			0 V/nm			0.2 V/nm		
(SRO-IV-Ru1)-O/(SRO-IV-Ru2)-O			(SRO-IV-Ru1)-O/(SRO-IV-Ru2)-O			(SRO-IV-Ru1)-O/(SRO-IV-Ru2)-O		
O(36)	2.09 Å/O(40)	2.03 Å	O(36)	2.02 Å/O(40)	1.98 Å	O(36)	2.09 Å/O(40)	2.05 Å
O(35)	1.93 Å/O(39)	2.08 Å	O(35)	1.91 Å/O(39)	2.04 Å	O(35)	1.91 Å/O(39)	2.01 Å
O(24)	1.98 Å/O(27)	2.01 Å	O(24)	2.04 Å/O(27)	1.98 Å	O(24)	2.00 Å/O(27)	1.99 Å
O(27)	2.01 Å/O(24)	2.00 Å	O(27)	2.04 Å/O(24)	1.97 Å	O(27)	2.02 Å/O(24)	1.98 Å
O(30)	1.99 Å/O(33)	2.00 Å	O(30)	2.04 Å/O(33)	1.98 Å	O(30)	1.98 Å/O(33)	2.02 Å
O(33)	2.00 Å/O(30)	1.99 Å	O(33)	2.04 Å/O(30)	1.97 Å	O(33)	1.99 Å/O(30)	2.01 Å

Table S4 The Ru-O bond length of the (SRO-VIII-Ru1)-O octahedral structure in the U-HS-9 model with different electric fields. The schematic diagram of lattice structure of U-HS-9 model is shown in Fig. S4(a).

	-0.1 V/nm	0 V/nm	0.1 V/nm
O(42)	2.00	2.04	2.05
O(41)	2.04	2.12	2.11
O(26)	2.01	1.97	1.97
O(30)	2.01	1.97	1.97
O(34)	2.01	2.02	2.02
O(38)	2.01	2.02	2.02