Supporting Information

Infinite-Layer/Perovskite Oxide Heterostructure Induced High-Spin States in $SrCuO_2/SrRuO_3$ Bilayer Films

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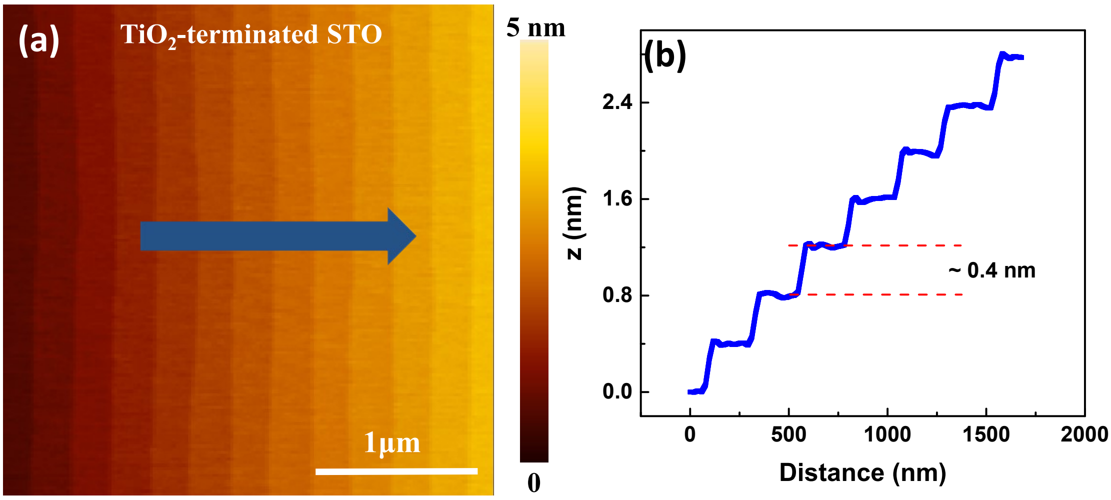
Figure S1. (a) Atomic force microscope (AFM) topography (3 μm × 3 μm) of the SrTiO3 (001) surface annealed at 1000 °C. Surface with regular terraces and steps is shown. (b) Height profile along the blue arrow marked in (a). Red dashed lines mark the height of a typical step.



In the process of sample preparation, the $SrTiO_3$ sample was treated by the standard chemical etching and thermal annealing method. All $SrTiO_3$ (001) substrates were first etched in aqua regia-based solution and then annealed in oxygen flow at 1000 ℃ for 2h. This procedure causes a recrystallization of substrate surface, leading to TiO_2-terminated surface. As shown by Fig. S1(a), regular-structured substrate surface appears, with relatively flat terrace planes and uniformly-spaced steps. The height of the step is about 0.4 nm, close to the lattice constant of $SrTiO_3$.[1] According to the literature using similar thermal treatment,[2,3] we believed that a single TiO_2 termination layer was formed for each terrace plane.

1. M. R. Castell, Surf. Sci 505, 1 (2002).
2. N. P. Guisinger et.al, ACS Nano 3, 4132 (2009).
3. W. H. Zhang et. al, Chin. Phys. Lett. 31, 017401 (2014).

Figure S2. X-ray reflectivity (XRR) of the (a) SRO and (b) SCO bare films grown on the (001)-oriented STO substrates with the same deposition time of 4 min. Good agreements between fitting curves (red) and experimental curves (black) are clearly demonstrated. The simulation curve is realized by the commercial software of DIFFRAC^{plus} LEPTOS 7. The deduced thickness of SRO layers is 13 uc and the thickness of SCO layer is 18 uc.

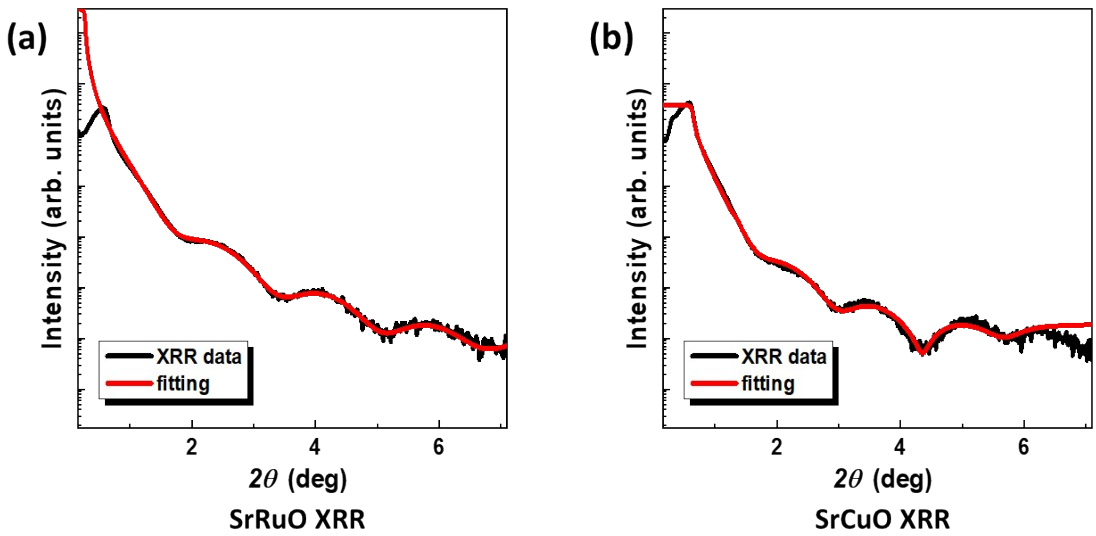


Figure S3. (a) ~(h) *M-T* curves of 5, 6, 7, 8, 9, 11, 13 and 16 uc SRO bare layers (red line) and bilayers (blue line) covered with the same SCO_{40} layer. *M-T* curves were measured in the field cooling mode with an out-of-plane magnetic field of 500 Oe. We can see that the SCO capping layer has induced a remarkable increase in *Tc* for the SRO layers thinner than 11 uc. The magnetization extracted from the *M-T* curves for the bilayer films is smaller than that of corresponding bare films. It is because the SCO capping layer has also enhanced the *Hc* of the bottom SRO layer. (i) The out-of-plane *M-H* curves of SRO_{16} bare layer (red line) and SCO_{40}/SRO_{16} bilayer (blue line) at 10K.

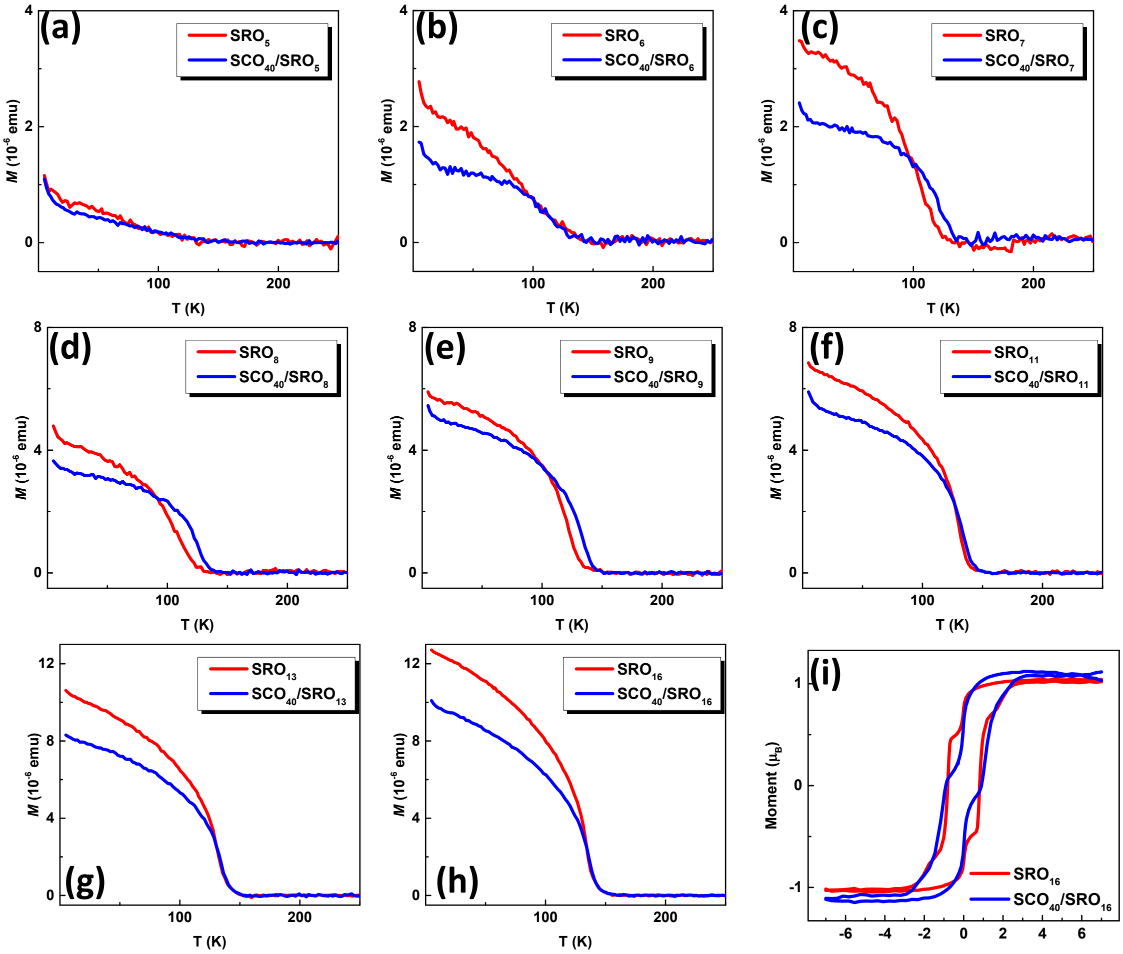


Figure S4. (a) Structural model of a SCO_4/SRO_5 heterostructure with the $[CuO_2]$-[Sr]-$[RuO_2]$ type interface for DFT calculations. The green, gray, blue and red dots represent the Sr, Ru, Cu and O ions, respectively. (b) DFT-resulted DOS on Ru, O and Sr sites for the five SRO layers, from the interfacial 1st layer to the 3rd, 4th and 5th inner layers. (c) Total *Ms* for each SRO layer. The largest *Ms* reaches ~2.8 μB/f.u. in the 1st SRO layer and is gradually reduces to ~1.8 μ_B in the 5th SRO layer, in good agreement with magnetic measurements.

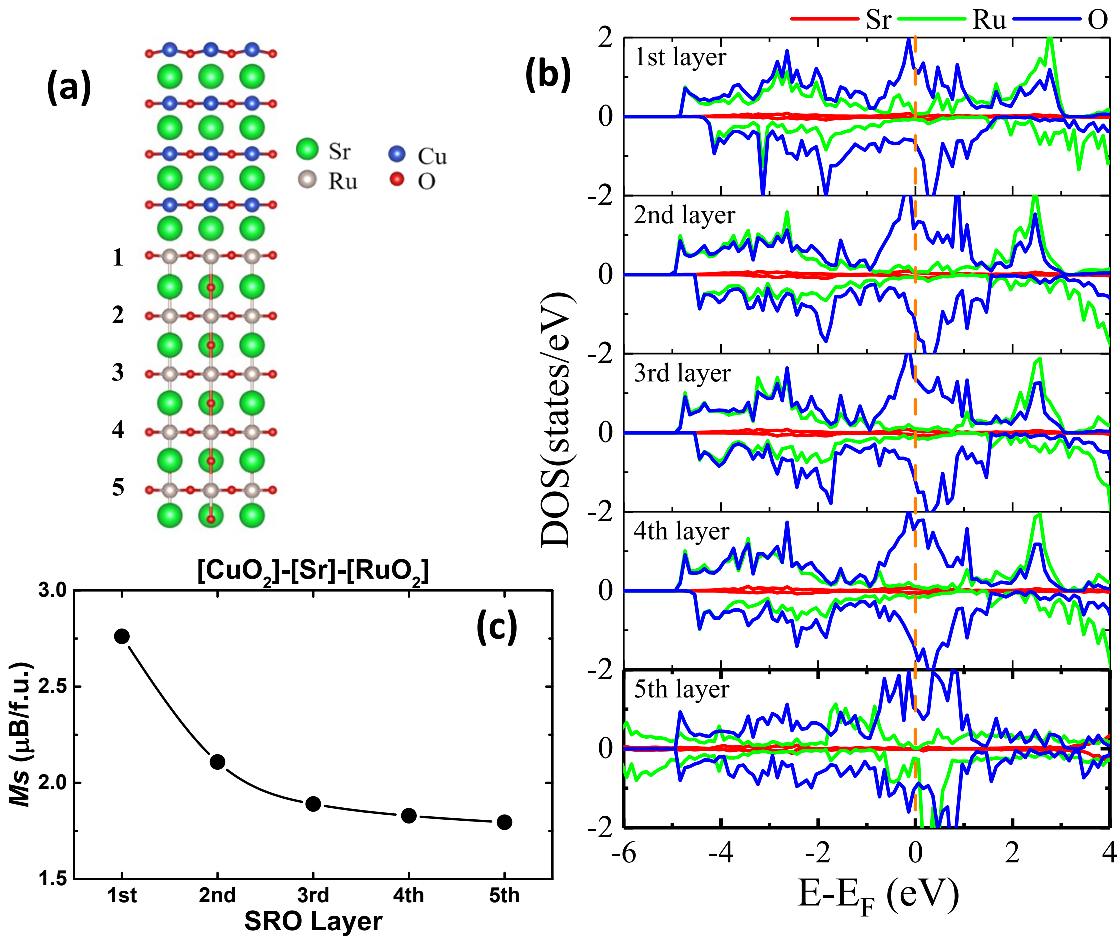


Figure S5 XAS at O K-edge for the SRO_9 bare, SRO_{17} single layer and SCO_5/SRO_9 and SCO_6/SRO_{17} bilayer. All curves were normalized according to its e_0, pre-edge range, and normalization range parameters. The O K-edge peaks from 528 to 535eV was divided into three parts, representing the hybridization between O *2p* states and the Ru t_{2g}, Ti t_{2g} and Ru/Ti e_g states, respectively. With increased film thickness, the Ti t_{2g} peak disappears.