Supporting Information

Electric field control of phase transition and tunable resistive switching in $SrFeO_{2.5}$

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Film growth

The growth dynamics of $La_{0.7}Sr_{0.3}MnO_3$-$SrFeO_{2.5}$ (LSMO-$SFO_{2.5}$) bilayer was monitored by reflection high energy electron diffraction (RHEED) as given in Figure S1. First, 7.8 nm thick LSMO (20 unit cell) was deposited on $SrTiO_3$ (100) (STO) substrate, then 30 nm $SFO_{2.5}$ was deposited on the top of LSMO film. In figure the left and right side of the dashed line clearly shows the periodic intensity oscillation of RHEED during the growth of LSMO and $SFO_{2.5}$ respectively. It confirms the layer by layer deposition of crystal unit cells. For $SFO_{2.5}$, one oscillation correspond to the half of the brownmillerite unit cell growth. The thickness of $SFO_{2.5}$ was optimized by estimating the time of single unit cell growth. The inset of Figure S1 shows the RHEED diffraction patterns obtained from STO substrate, LSMO, and $SFO_{2.5}$ films. The strips of diffraction patterns also indicate the high quality epitaxial growth.

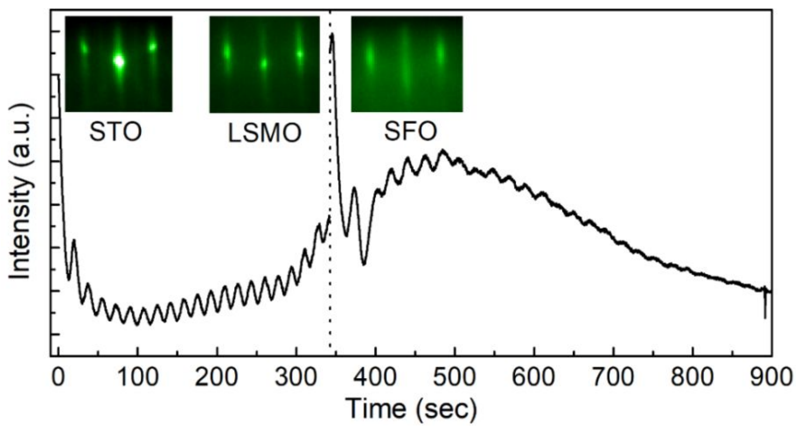

Figure S1. RHEED intensity oscillation during the growth of LSMO and $SFO_{2.5}$. Left side from dashed line for LSMO and right side for $SFO_{2.5}$. Insets are the diffraction pattern obtained from STO substrate, LSMO and $SFO_{2.5}$ films.

Reproducibility of gating experiments

The surface of bilayer sample was investigated by atomic force microscope before and after gating. The sample was carefully cleaned by acetone and alcohol before the surface scanning. The collected images at three states "pristine, $-V_g$, and $+V_g$" are given in Figure S2. We do not observe any notable change on the surface in reversible gating process. The observed roughness (R_a) in the 5×5 μm^2 area are 0.12 nm for pristine sample (before gating), 0.26 nm for $-V_g$ state, and 0.33 nm for $+V_g$ state. The small value of roughness at each state confirms that the gating experiment is repeatable.

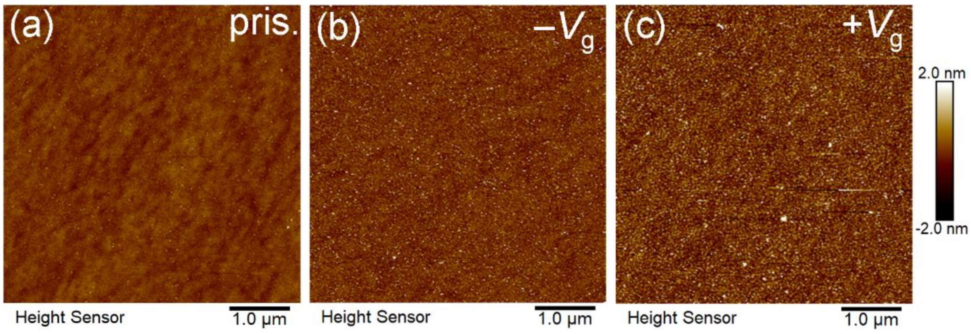

Figure S2. (a) AFM image of pristine LSMO-$SFO_{2.5}$ sample, (b) and (c) after negative and positive gating.

XAS spectra of oxygen *K*-edge and iron *L*-edge

Figure S3 shows the oxygen *K*-edge and iron *L*-edge spectra of single layer $SFO_{2.5}$ and $SFO_{3-\delta}$ sample. The single layer samples were fabricated directly by controlling the oxygen partial pressure during the film growth in the PLD system. The detail is given in experimental part of main text. The oxygen *K*-edge and iron *L*-edge spectra of single layer samples are very similar to the spectra of $+V_g$ and $-V_g$ gated samples given in main text Figure 3.

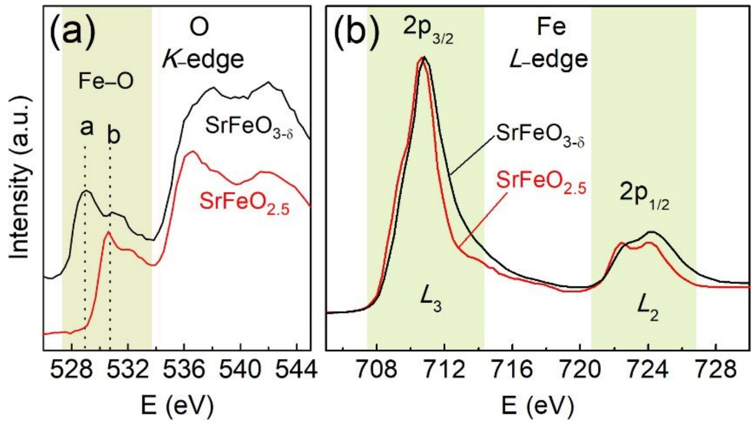

Figure S3. (a) Oxygen *K*-edge (b) Iron *L*-edge XAS curves of single layer $SFO_{2.5}$ and $SFO_{3-\delta}$ sample. The shaded region of oxygen *K*-edge spectra denote the Fe-O hybridization, and in iron $L_{2,3}$-edge spectra the shaded area denotes the shoulders of Fe-$2p_{3/2}$ and Fe-$2p_{1/2}$ region.

Magnetic structure of $SrFeO_3$

Figure S4 shows the 3D visualization of helical magnetic structure of cubic unit cell $SrFeO_3$. The yellow vectors represent the moments of iron atoms. Here, the magnetic moments of the iron atoms are tilted with respect to the *c* axis by angle of −35.3 degrees which are lying perpendicular to the direction [111] using the cubic setting[1].

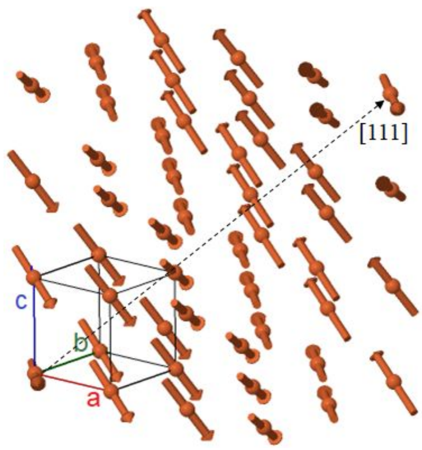

Figure S4. Visualization of helical magnetic structure of $SrFeO_3$. Vectors are representing the moments of iron atoms. The structure dimension consists of 3a, 3b, 3c unit cell.

Temperature dependent magnetization measurement (*M-T*)

To find out the effect of ionic liquid gating on LSMO layer, in-plane temperature dependent magnetic measurements (*M-T*) were performed for bilayer samples. The recorded data during the heating process from 10 K to 400 K is given in Figure S5. A strong magnetic signal was detected in both $-V_g$ and $+V_g$ gated state. As the temperature increases from 10 K to 400 K, the moment of LSMO starts to decrease and becomes zero nearly at 300 K, indicating the Curie temperature (T_C) of LSMO. The value of T_C is almost the same for both $-V_g$ and $+V_g$ states, because the controlled experiment was performed by optimizing the voltage and time of IL gating that only modulate the oxygen content in top $SFO_{2.5}$ layer without inducing prominent difference in magnetic properties of LSMO.

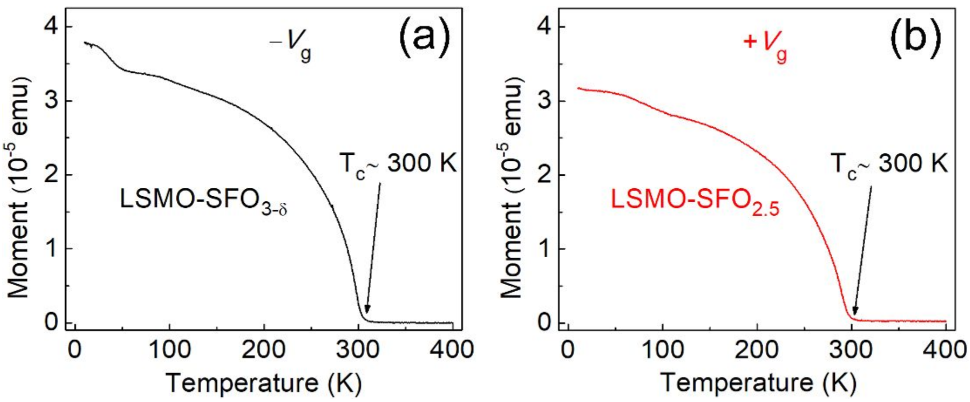

Figure S5. (a) and (b) Temperature dependent magnetization data after the $-V_g$ and $+V_g$ state of LSMO–$SFO_{2.5}$ sample.

$SrFeO_{2.5}$ film on Nb-doped $SrTiO_3$ and $SrTiO_3$ substrate.

Figure S6 shows the XRD patterns of $SFO_{2.5}$ film on Nb-doped $SrTiO_3$ (NSTO) substrate and LSMO-$SFO_{2.5}$ film on STO substrate. The NSTO and STO substrates, with almost the same lattice parameter, allow the coherent growth of the $SFO_{2.5}$ film with almost identical lattices. The XRD patterns clearly exhibit (080) plane diffraction peaks and half order (020), (040), (060) and (0<u>10</u>0) diffraction peaks in both samples, which are the characteristic signals of the BM phased film. The film thicknesses are the same on both substrates.

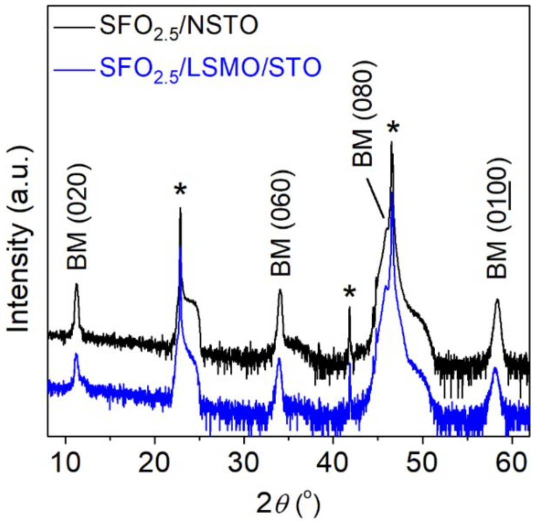

Figure S6. The XRD patterns of $SrFeO_{2.5}$ film on Nb-doped STO substrate and LSMO-$SrFeO_{2.5}$ film on STO substrate. Steric indicate the peaks from substrates.

Oxygen *K*-edge spectra at different gating time

The details of oxygen stoichiometry was investigated by *K*-edge spectra after applying negative gating up to 40 minutes on the $SFO_{2.5}$/NSTO sample. In Figure S7, a gradual change at Fe-O hybridization can be easily observed with increasing $-V_g$ time. For the initial state (no IL gating), the O *K*-edge spectrum shows only one peak at position *b*, indicating the loss of spectral feature associated with O 2*p*−Fe 3*d* hybridization[2, 3]. As the applied gating time increases gradually from 10 to 40 minutes, the signals at position *a* start to increase gradually, indicating the raise of oxygen content in $SFO_{2.5}$. Logically, it supports the extending and overlapping of O 2*p*−Fe 3*d* bands that favours the conductive behaviour, and reflect the PV phase of

$SFO_{3-\delta}$ [2, 3].

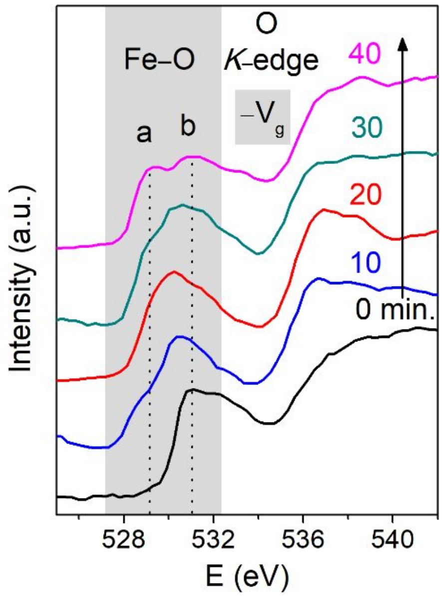

Figure S7. Oxygen *K*-edge XAS curves of negative gated $SFO_{2.5}$/NSTO sample at different time. The arrow indicates the sequence of $-V_g$ time in minutes. The shaded region denotes the ion-oxygen 3*d*-2*p* band hybridization.

References

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