Supporting Information

Ultra-high resistive switching current ratio and improved ferroelectricity and dielectric tunability performance in a BaTiO<sub>3</sub>/La<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub> heterostructure by inserting a SrCoO<sub>2.5</sub> layer

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Fig. S1 The XRR of (a) BTO, (b) SCO and (c) LSMO.

The thickness of the single-layer film could be determined by fitting the XRR data. The fitting results showed that the thicknesses of BTO, SCO and LSMO

deposited for 1h were 42 nm, 50 nm and 34 nm, respectively.



Fig. S2 The local (a) phase hysteresis loop and (b) piezoresponse of BTO/SCO/LSMO.

The local phase and amplitude loops were measured in various areas in Fig. 2d. All the loops show standard ferroelectric behaviors. Therefore, the SCO intercalation does not reduce the ferroelectric properties of BTO film.



Fig. S3 Polarization-electric field hysteresis loops of the BTO/SCO bilayer (green line) and the BTO film (orange line) with the same electric field at room temperature.



Fig. S4 O 1s XPS experimental data and fitting results of BTO films in the (a) BTO/LSMO/STO and (b) BTO/SCO/LSMO/STO.

For the BTO in the BTO/LSMO/STO, the main peak at 529.5 eV corresponds to the lattice oxygen while the satellite peak at 531.7 eV is regarded as the oxygen vacancies. For BTO in the BTO/SCO/LSMO/STO, the main peak located at 529.2 eV and the satellite peak located at 531.0 eV. These results are close to the 529.2 eV and 531.1 eV reported in the literature <sup>[1]</sup>, indicating that the data collected and the results of the fit are credible. The XPS test is a semi-quantitative test method, and the area ratio of the peaks in the fitting result is approximately equal to the content ratio of the elements, which shows that the oxygen vacancy concentration of BTO in BTO/LSMO/STO is 14.9%, while the oxygen vacancy concentration of BTO in BTO/SCO/LSMO/STO is 28.7%. The increased concentration of oxygen vacancies may contribute to the enhancement of ferroelectricity <sup>[2]</sup>, while more oxygen vacancies can form thicker conductive filaments, thus enhancing the resistance properties <sup>[3]</sup>.

## References

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