

## Highly efficient and fast modulation of magnetic coupling interaction in SrCoO<sub>2.5</sub>/La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub> heterostructure

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To verify the effect of HAc on the phase transition of B-SCO films, the single B-SCO films were fabricated by pulsed laser deposition technology. As shown in the Figs. S1(a-b), the B-SCO films (40 u.c., initial state) displays the negligibly small magnetization. The results are consistent with the previous reports. After immersing the B-SCO films into HAc solution for a short time of 2s, the sample shows distinct magnetic hysteresis loops and the coercive field is approximate 5000 Oe (Fig. S1(a)). Compared with the initial state of films, the B-SCO films treated by HAc exhibits larger saturation magnetization (as shown in Fig. S1(b)). And its  $T_C$  is approach to that of P-SCO films (Fig. S4(b)). This means that the antiferromagnetic B-SCO phase has been transited to perovskite ferromagnetic P-SCO phase. Fig. S4(a) demonstrates the result of magnetic measurement of single P-SCO films, which is similar with Fig. S1(a) (red line). Thus, there is no doubt that the sample would undergo a structural phase transition after treated by HAc. The next, the same sample was heated to 150°C in an atmospheric environment. The magnetic signs were hardly detected indicating the sample recovered to initial state.

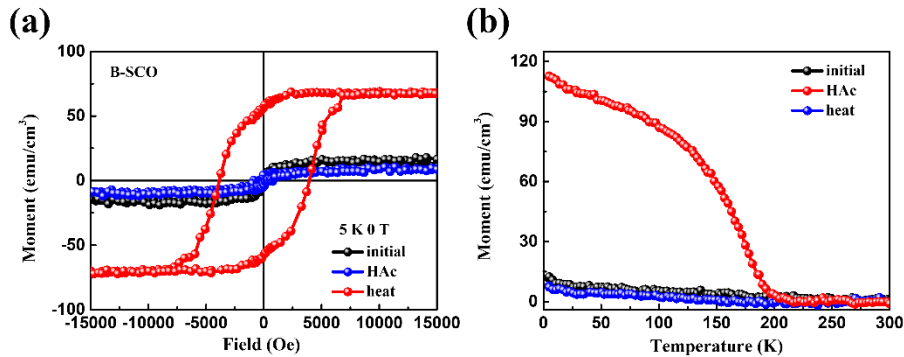


Fig. S1 In-plane moment versus magnetic field (a) and temperature (b) curve of B-SCO (40 u.c.) single films at different states.

The XRD measurement is conducted to further confirm the structure transition of B-SCO films (Fig. S2(a)). At initial state, the (002), (006), and (0010) characteristic diffraction peak of brownmillerite B-SCO is observed due to the alternating stacking of CO<sub>4</sub> tetrahedral layers and CO<sub>6</sub> octahedral layers. After treated by HAc, the half-order diffraction peaks disappear. And the new diffraction peak around 48° is detected, which confirms the successful conversion of brownmillerite to perovskite phase. When

the same sample was heated, it reverts to the brownmillerite structure. And the half-order peaks appear again. Further, Co valance state of films with different state were also investigated by XAS measurement to confirm the structure transition. As shown in Fig. S2(b), the Co  $L_3$ -edge shifts to higher energy direction compared with initial state B-SCO films. In conclusion, the above experimental results provide sufficient evidence for the topotactic phase transition of B-SCO films after HAc process.

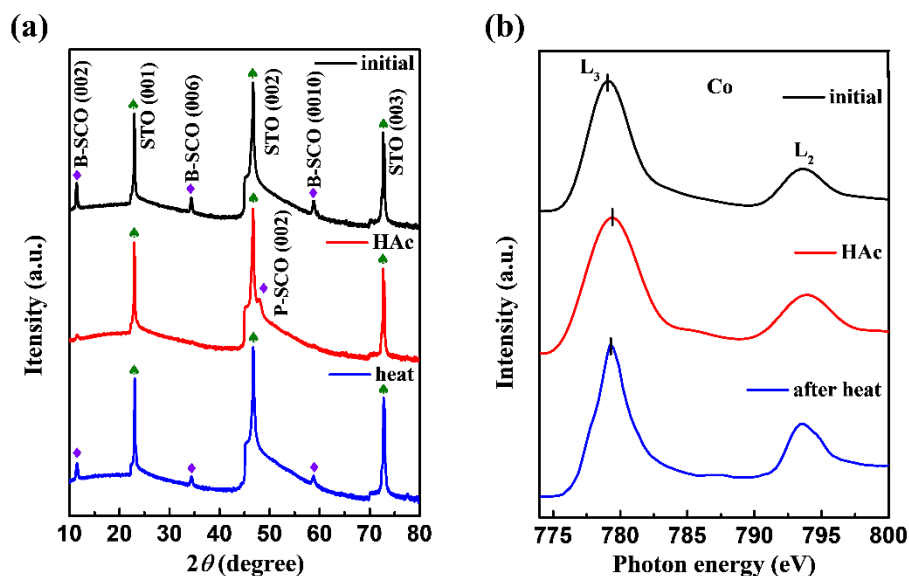


Fig. S2 XRD scans recorded (a) and XAS spectra of Co  $L$ -edge (b) for single B-SCO films undergoing topotactic phase transformation.

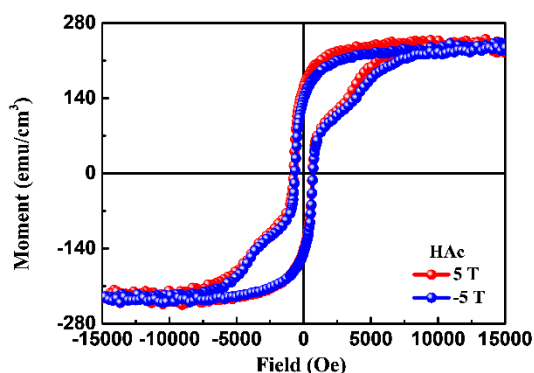


Fig. S3 Major magnetic hysteresis loop of B-SCO/LCMO bilayer after treated by HAc, measured at 5 K.

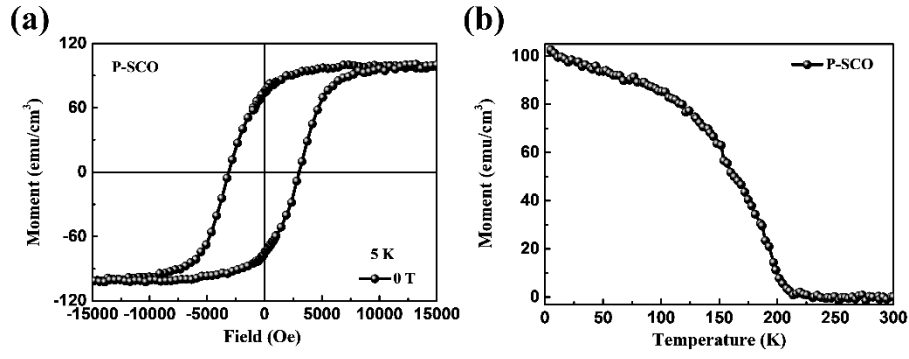


Fig. S4 Magnetic properties measurement of P-SCO (40 u.c.) single films on STO substrates.

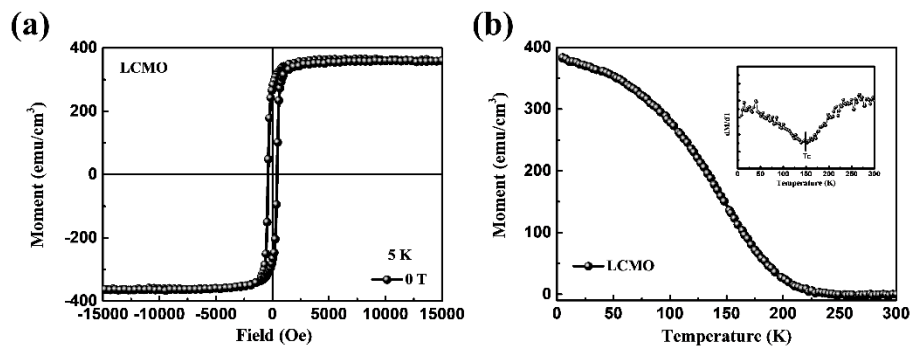


Fig. S5 Magnetic hysteresis loops (a) and moment as function of temperature (b) of 40 u.c. LCMO films. The inset is the temperature dependence of  $dM/dT$  curve, indicating that the Curie temperature is around 150 K.

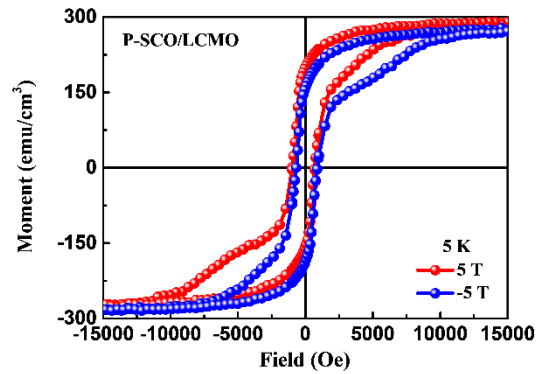


Fig. S6 Magnetic hysteresis loops of P-SCO (20 u.c.) / LCMO (40 u.c.) bilayer at 5 K with the magnetic field along in-plane direction.

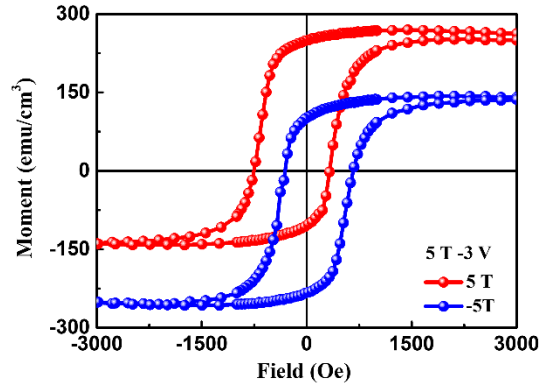


Fig. S7 Minor magnetic hysteresis loop of B-SCO/LCMO bilayer after gated by IL with -3 V.