

Supplementary Materials

Control of magnetic anisotropy in epitaxial SrRuO₃ via oxygen octahedral engineering

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Section 1. More detailed experimental details

Sample Preparation

The epitaxial SRO thin films were grown on (001)-oriented TiO₂-terminated STO substrates via pulsed laser deposition (PLD). Deposition was carried out at 680 °C under an oxygen partial pressure of 13 Pa. A KrF excimer laser (wavelength 248 nm) operating at a repetition rate of 13 Hz and a laser fluence of 2 J cm⁻² was used to ablate a high-purity SRO ceramic target (99.99%, Kurt J. Lesker Company). The target–substrate distance was maintained at 4.5 cm. Film growth was monitored in situ by reflection high-energy electron diffraction (RHEED). Following deposition, the samples were cooled to room temperature at a rate of 20 °C min⁻¹ in an oxygen atmosphere at a partial pressure of 5 Torr.

Sample characterization

The crystalline structure of the epitaxial SRO films was characterized by X-ray diffraction (XRD) 2θ – ω scans of the (002) reflection using a SmartLab diffractometer. The surface morphology and roughness were examined by atomic force microscopy (AFM) in tapping mode, performed with an Asylum Research MFP-3D-SA system over a scan area of 5 × 5 μm². The measured root-mean-square (RMS) roughness was

100 pm. Magnetic properties were evaluated using a Quantum Design superconducting quantum interference device (SQUID) magnetometer (MPMS), with magnetization versus field ($M-H$) curves at 10 K.

Hall Measurements

Hall measurements were carried out in Quantum Design Physical Property Measurement System (PPMS) with the 5×2 mm sample size. The Hall resistivity was extracted at 10 K and in a field range of ± 3 T.

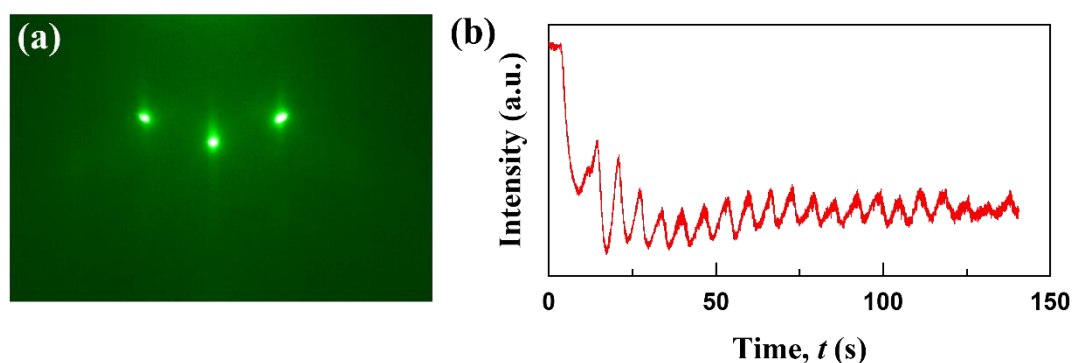


Figure S1. (a) The RHEED pattern of the deposited SRO film. (b) RHEED intensity oscillations recorded during deposition of SRO films in 20 uc thicknesses.

Section 2. Rocking curves of SRO (002) peaks with variable thicknesses.

We further performed XRD rocking curve measurements on all SRO thin films, as presented in Figure S3A–E. Narrow full width at half-maximum (FWHM) values of 0.0105° , 0.098° , 0.093° , 0.082° , and 0.076° were obtained for the SRO (002) diffraction peaks corresponding to film thicknesses of 10, 15, 20, 30, and 50 uc, respectively. The presence of pronounced Laue oscillations and sharp rocking curves demonstrate the high crystalline uniformity of the SRO films on STO substrates. The FWHM gradually decreases as the SRO film thickness increases, which agrees well with the results from $2\theta-\omega$ XRD scans.

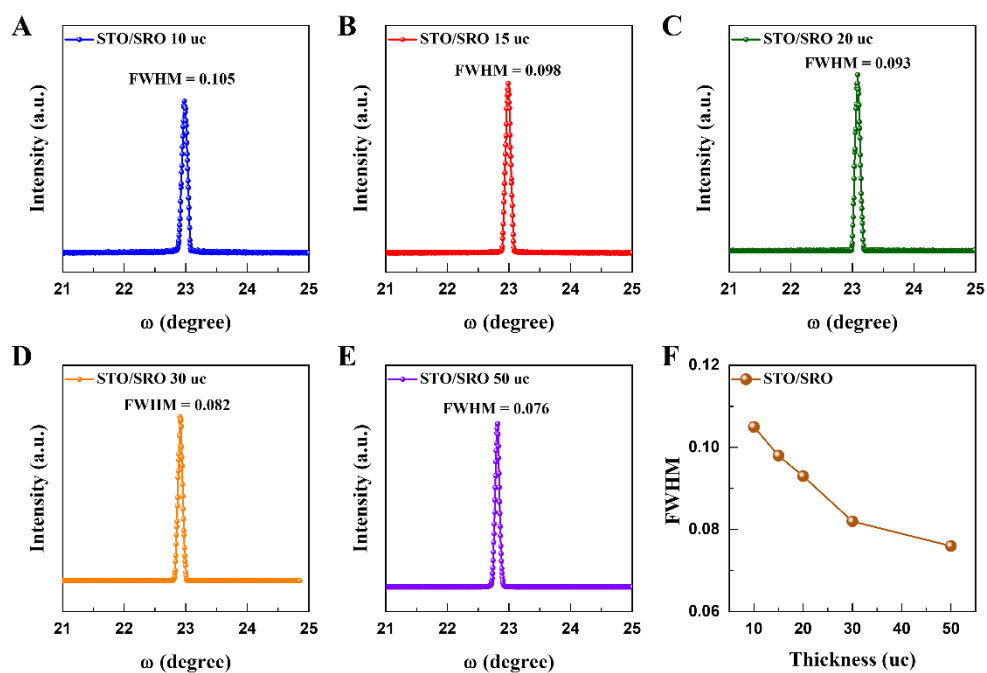


Figure S2. XRD rocking curves for STO/SRO films with thicknesses of (A) 10, (B) 15, (C) 20, (D) 30, and (E) 50uc. (F) Plot of FWHM versus SRO film thickness.

Section 3. AFM topography images of the SRO films with different thicknesses.

AFM characterization was performed on SRO thin films grown on STO substrates. All samples exhibit atomically flat surfaces featuring well-defined step-terrace morphologies. The variation in terrace width across films is attributed to differences in the miscut angle of the STO substrates.

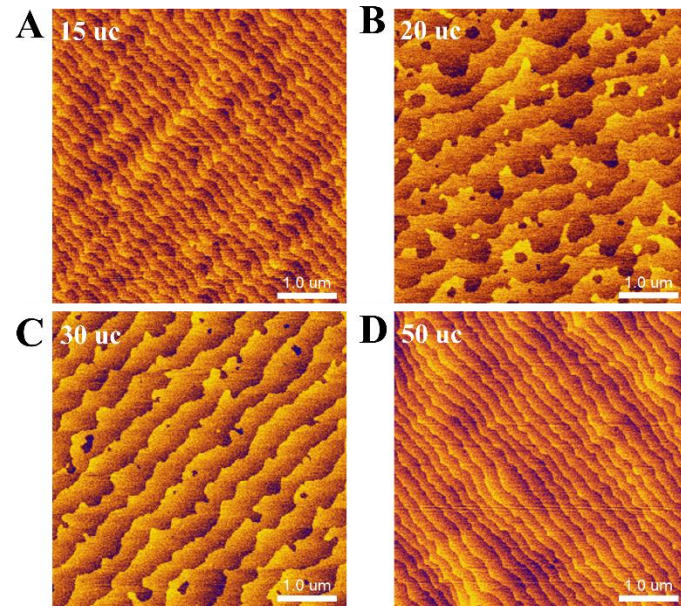


Figure S3. Atomic force microscopy (AFM) topography maps of SRO thin films with thicknesses of (A) 15, (B) 20, (C) 30, and (D) 50 uc.

Section 4. The M-H curves for the SRO film with variable thickness.

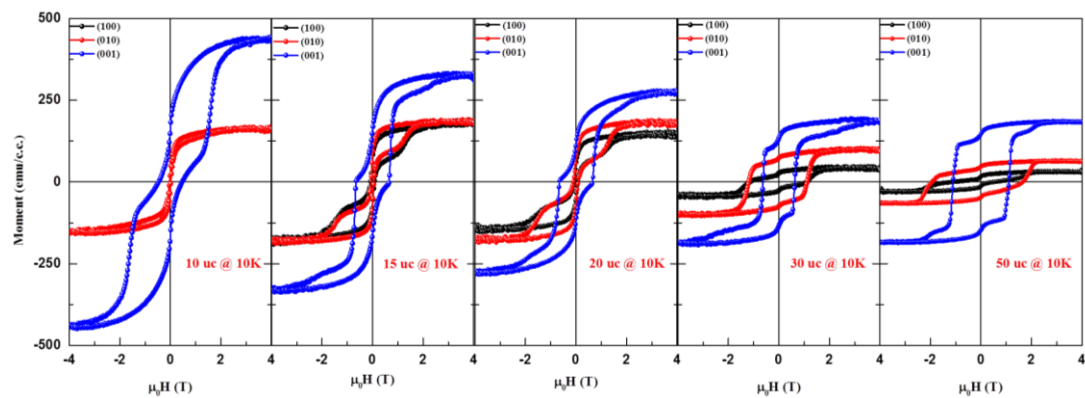


Figure S4. Magnetization vs magnetic field ($M-H$) curve of 10 uc-thick SRO thin film with variable thickness from 10 uc to 50 uc.

Section 5. The fitting result of out-of-plane M-H curves for 10 uc SRO.

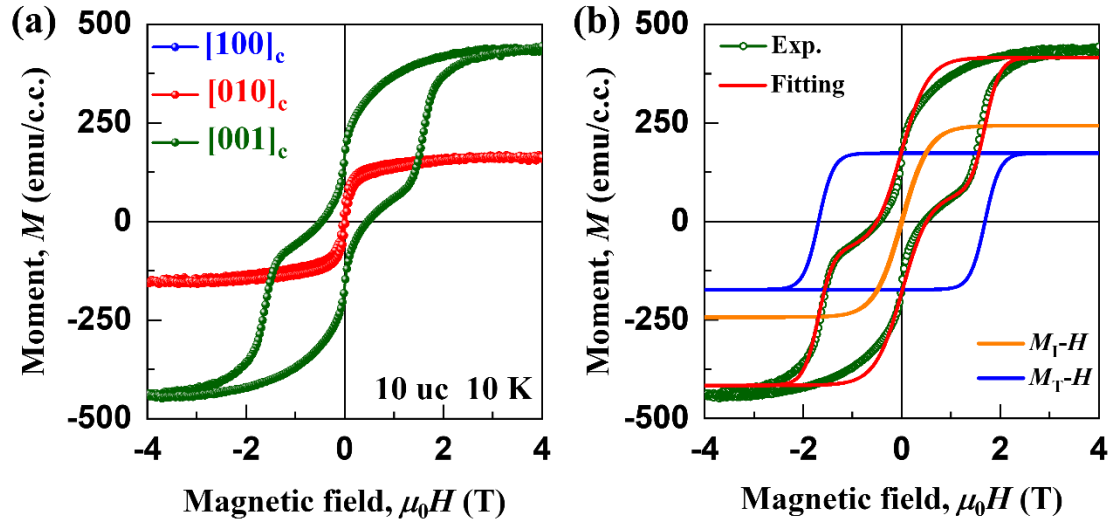


Figure S5. $M-H$ curve of 10 uc-thick SRO thin film. This out-of-plane $M-H$ curve can be fitted by two hysteresis loops (M_I-H and M_T-H), which correspond to the magnetic signals from interfacial insulating SRO_I and metallic SRO_T regions, respectively.