

Supplementary Information

Electrically Driven Inverse Metamagnetic Transition in $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$

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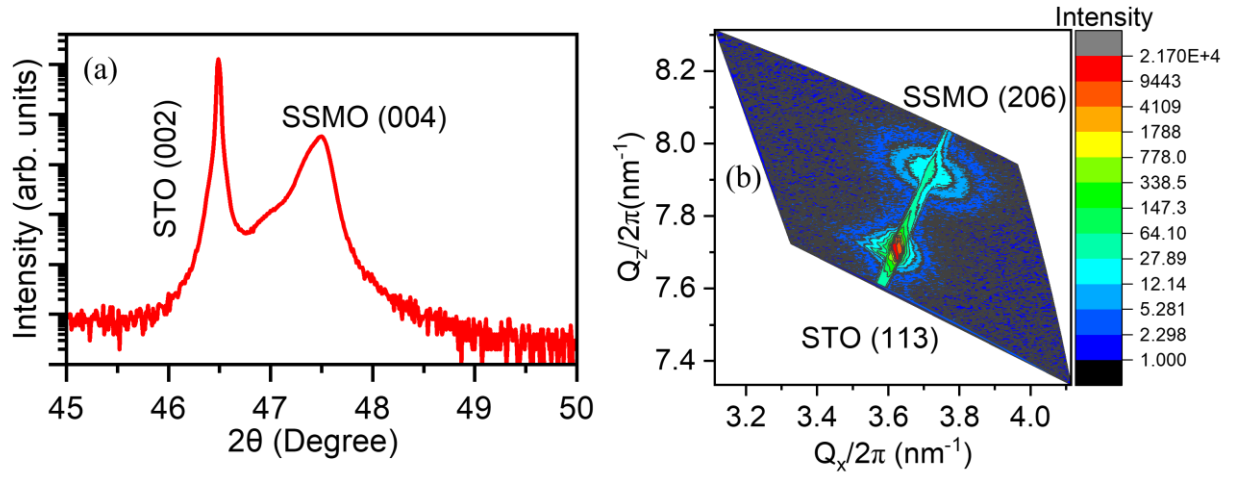


Fig. S1: X-ray diffraction pattern of the epitaxial thin film. (a) The X-ray diffraction pattern of the $\text{Sm}_{0.6}\text{Sr}_{0.4}\text{MnO}_3$ (SSMO) thin film grown on an STO (001) substrate confirms the epitaxial nature of the film. (b) Reciprocal space mapping data of the SSMO thin film along the STO (113) plane show that the (206) reflection of SSMO aligns epitaxially with the STO (113) plane, indicating that the film is in a nearly relaxed state.

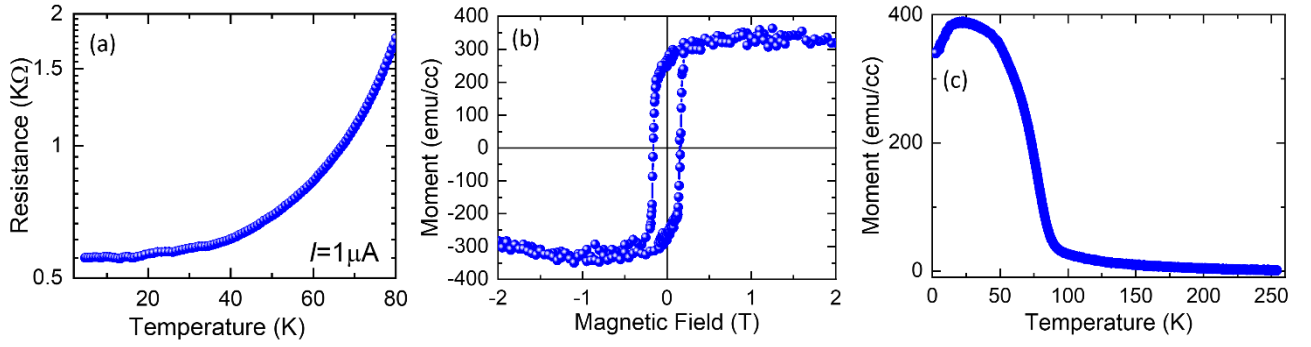


Fig. S2 | Electrical transport and magnetization measurements of the $\text{Sm}_{0.6}\text{Sr}_{0.4}\text{MnO}_3$ thin film. (a) Temperature-dependent resistance, measured under a 0 T magnetic field, confirms the metallic behavior of the SSMO thin film. (b) Magnetic moment as a function of magnetic field at 5 K exhibits a clear hysteresis loop, indicating robust ferromagnetic ordering. (c) Temperature-dependent field-cooled magnetization data show a paramagnetic-to-ferromagnetic phase transition, with a Curie temperature around 100 K.

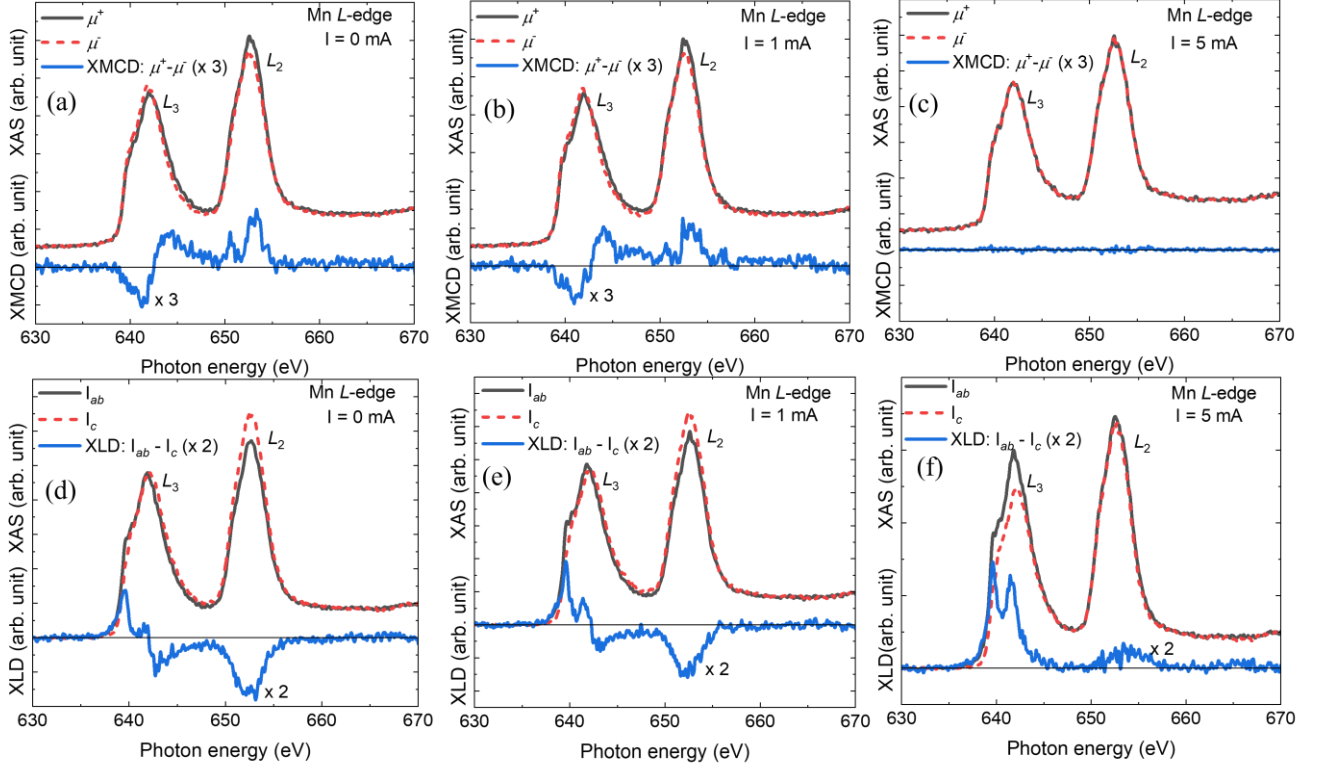


Fig. S3: X-ray absorption spectroscopy (XAS), X-ray magnetic circular dichroism (XMCD), and X-ray linear dichroism (XLD) measurements of the SSMO thin film under applied current. (a–c) Polarization-dependent Mn $L_{3,2}$ -edges XAS and XMCD spectra of the SSMO thin film measured at applied currents of 0 mA (a), 1 mA (b), and 5 mA (c). (d–f) Polarization-dependent Mn $L_{3,2}$ -edges XAS and XLD spectra acquired at 0 mA (d), 1 mA (e), and 5 mA (f). Multiplication factors for the XMCD and XLD signals are also noted in the respective panels.

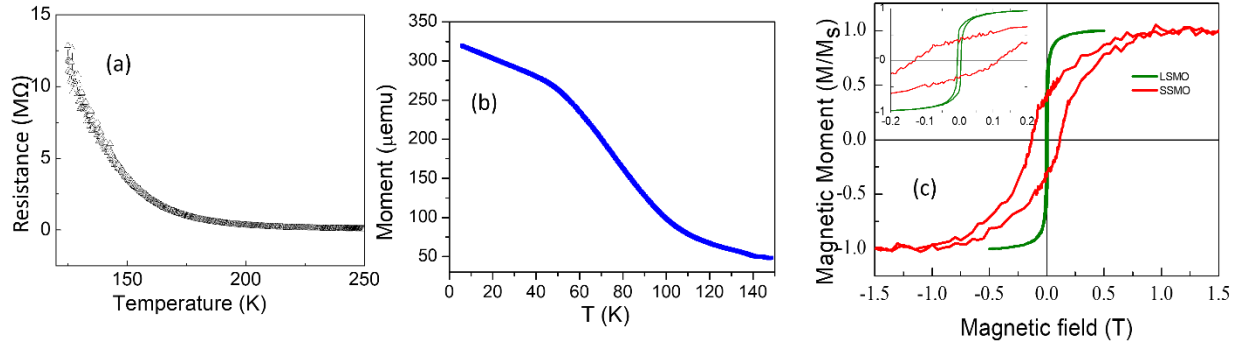


Fig. S4: Temperature-dependent transport and magnetization of SSMO-FI. (a) R–T and (b) M–T curves for $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x = 0.25$) films, demonstrating insulating behavior and a Curie temperature $T_C \approx 100$ K. (c) Normalized magnetic moment (M/M_s) as a function of applied magnetic field for LSMO (200 nm) and SSMO (100 nm) thin films measured at 5 K. The inset provides an enlarged view highlighting the coercive fields of the films. The coercive fields are determined to be 0.001 T and 0.14 T for LSMO and SSMO, respectively.

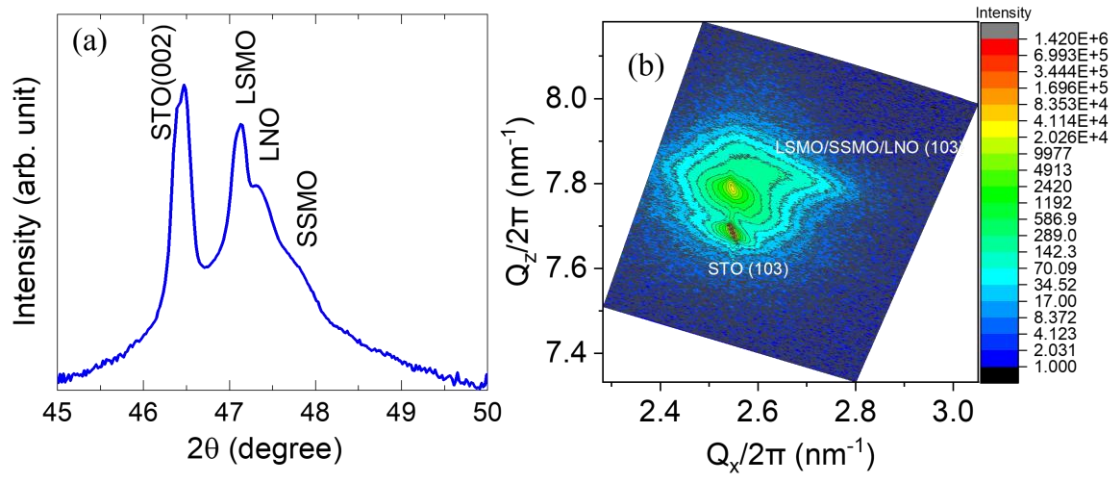


Fig. S5: X-ray Diffraction Analysis of Multilayer Thin Film Heterostructures. (a) XRD pattern of the LNO (200 nm)/SSMO (8.5 nm)/STO (1 nm)/LSMO (200 nm) film stack grown on an STO (001) substrate, confirming the high crystallinity and epitaxial nature of the multilayer structure. Distinct diffraction peaks corresponding to the LSMO, LNO, and SSMO layers are clearly observed. (b) Reciprocal space mapping (RSM) plot demonstrating nearly full lattice matching of the LNO/SSMO/STO/LSMO heterostructure with the underlying STO (001) substrate.

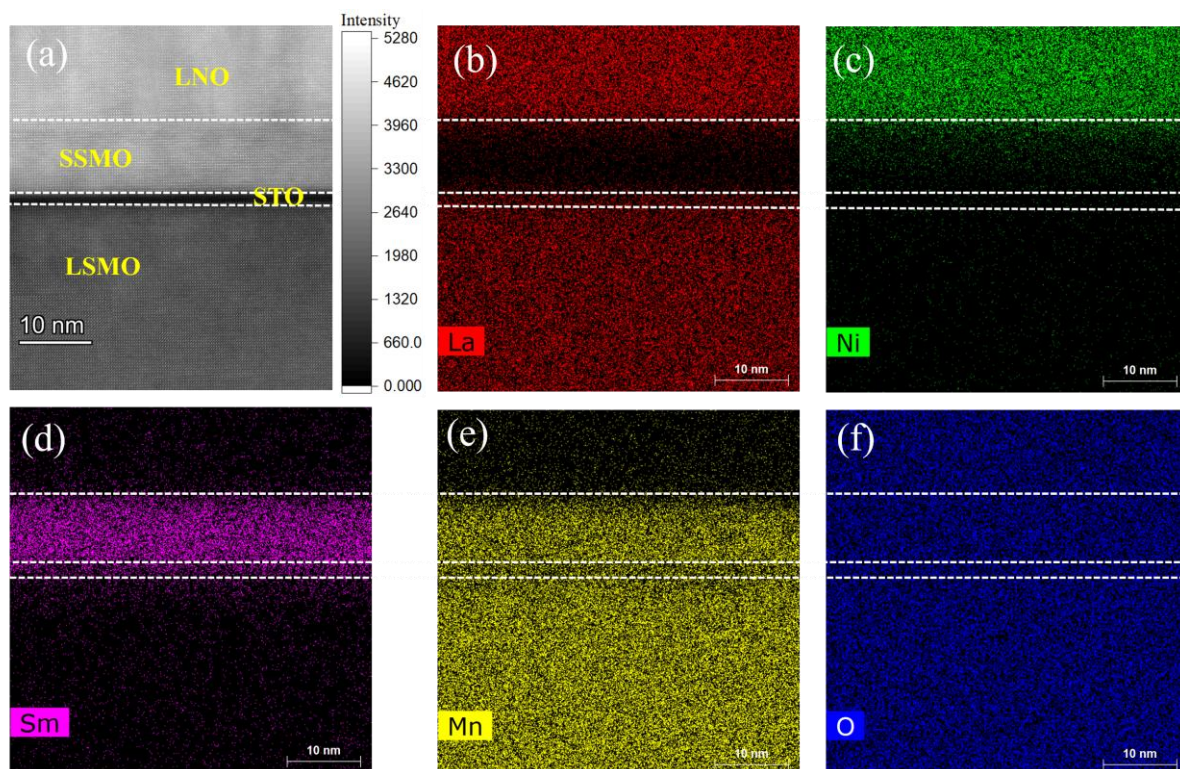


Fig. S6: High-Resolution TEM and Elemental Mapping of Oxide Heterostructures. (a) Cross-sectional high-resolution TEM image of the LNO/SSMO (8.5 nm)/STO (1 nm)/LSMO heterostructure grown on an STO (001) substrate. The image clearly shows distinct, well-defined interfaces between LSMO/STO and STO/SSMO, confirming high-quality epitaxial growth. The SSMO/LNO interface appears comparatively less sharp, indicating a relatively diffuse transition at that boundary. The colour scale represents the intensity scale, ranging from black (minimum intensity) to white (maximum intensity). (b–f) Elemental EDX maps for La (red), Ni (green), Sm (magenta), Mn (yellow), and O (blue), respectively. The colours in parentheses represent the pseudo-colour assigned to each element to visualise the spatial distribution and relative intensity. These maps confirm the spatial distribution and compositional uniformity of each element across the multilayer stack, validating the intended stoichiometry and structural integrity of the heterostructure.

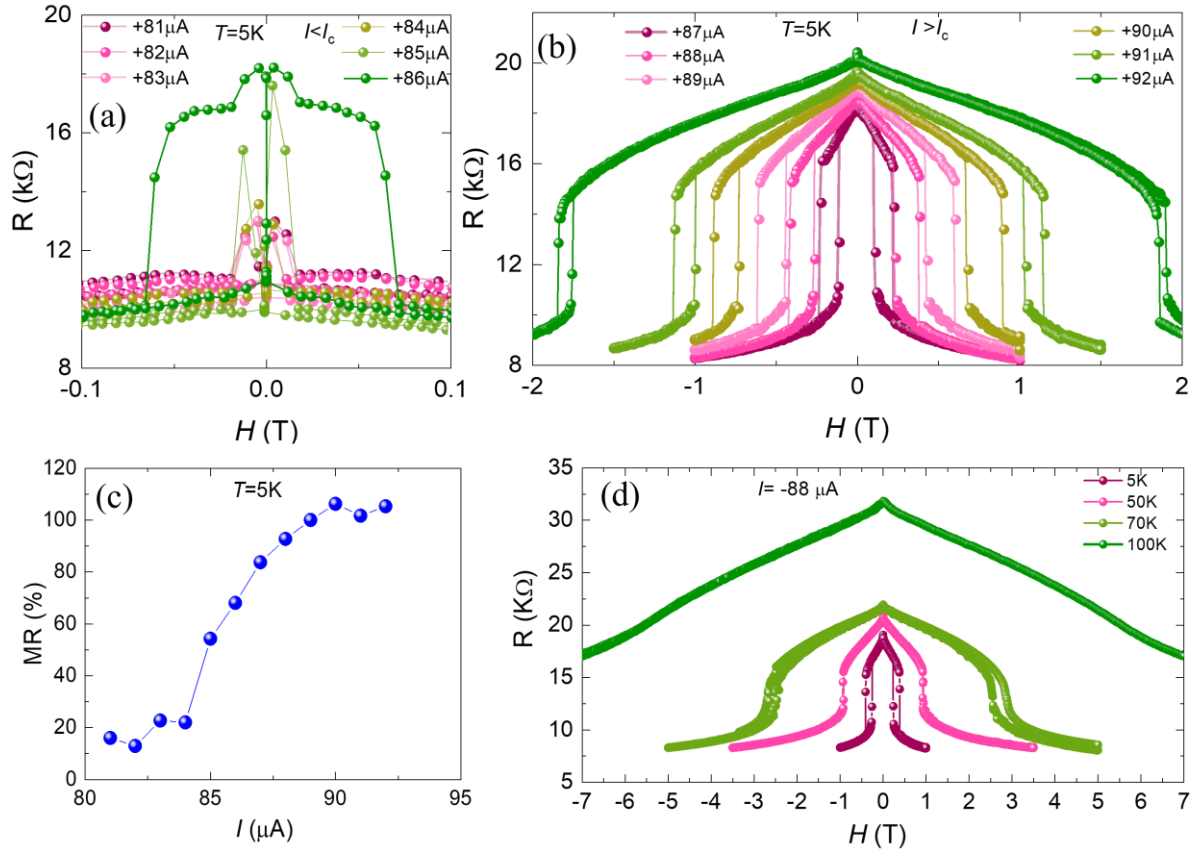


Fig. S7: Bias- and Temperature-Dependent Magnetoresistance in the SFTJ Device. (a, b) Magnetoresistance (MR) measured under positive bias currents exhibits a threshold behavior similar to that under negative bias: negligible MR below $86 \mu\text{A}$ and a notable increase above this current, reaching up to $\sim 100\%$. The asymmetry between positive and negative bias responses highlights unconventional, bias-dependent transport distinct from standard MTJs and SFTJs. (c) Quantitative MR analysis under positive bias, derived from panels (a) and (b), confirms current-driven switching and reflects the unique electronic pathways across the LNO and LSMO electrodes. (d) Resistance versus magnetic field at various temperatures under $-88 \mu\text{A}$ bias shows a monotonic decrease in MR with increasing temperature, correlating with the magnetic ordering in the SSMO barrier.

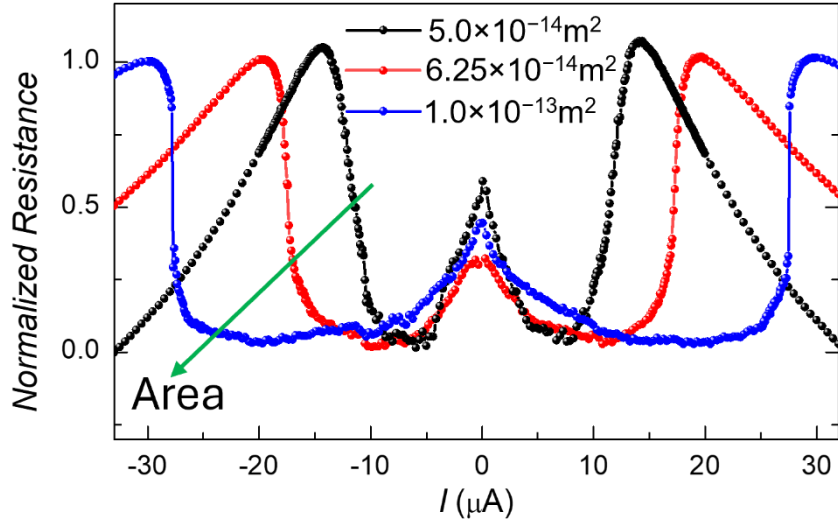


Fig. S8: Area scaling of the switching threshold. Normalized resistance R/R_0 versus current for three SFTJs (6.5-nm SSMO barrier) with varying lateral areas. The critical current I_c increases with device area (≈ 14 , 20, and 35 μA , respectively), so the critical current density $J_c \approx \frac{I_c}{A}$ remains nearly constant. This linear $I_c \propto A$ trend indicates a current-density threshold (flux-driven) switching mechanism.

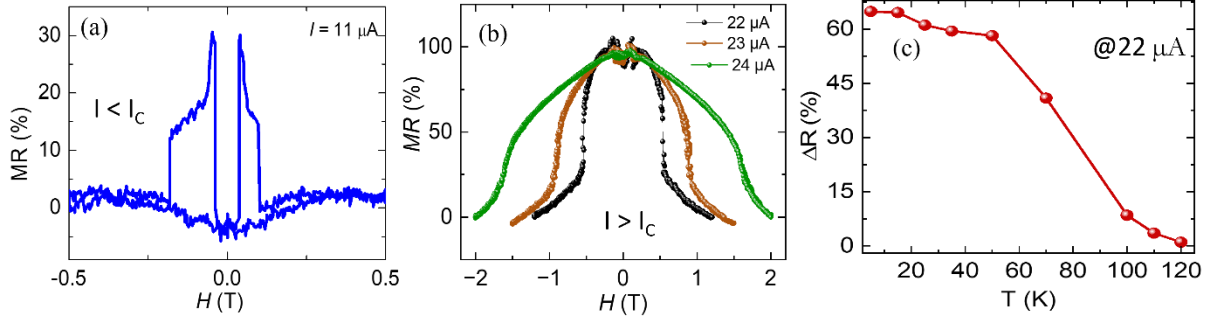


Fig S9: Current- and Temperature-Dependent Magnetoresistance in SFTJ Devices with a 6.5 nm SSMO-FI Barrier. (a) Magnetoresistance (MR) of the SFTJ device with a 6.5 nm $\text{Sm}_{0.75}\text{Sr}_{0.25}\text{MnO}_3$ (SSMO-FI) barrier layer, measured under a constant bias current of 11 μA . This behavior is characteristic of conventional SFTJs, indicating that below the critical current, the SSMO layer retains its ferromagnetic insulating (FI) character and facilitates spin-filtering. (b) MR profiles at various applied currents reveal an unconventional switching behavior, with a clear transition emerging above the critical current ($\sim 22 \mu\text{A}$), signaling a shift in the magnetic phase of the barrier. (c) Temperature-dependent MR measured at 22 μA shows a monotonic decrease with increasing temperature, vanishing near the Curie temperature (~ 100 K), consistent with the magnetic ordering in the SSMO-FI barrier layer.

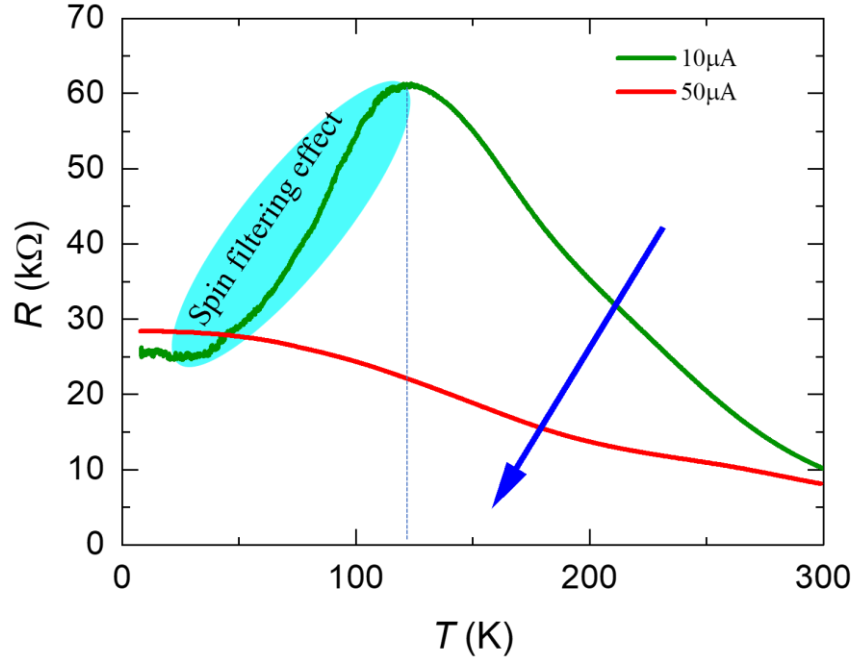


Fig. S10: Temperature-Dependent Transport in SFTJ Devices Demonstrating Spin-Filtering Behavior of the SSMO Barrier. This figure illustrates the fundamental operating principle and current-dependent transport characteristics of spin-filter tunnel junctions (SFTJs) employing a 6.5 nm $\text{Sm}_{0.75}\text{Sr}_{0.25}\text{MnO}_3$ (SSMO) ferromagnetic insulating (FMI) barrier. SFTJs exploit spin-dependent tunneling, where the exchange-split conduction band in the FMI barrier creates unequal potential barrier heights for up-spin and down-spin electrons. In the ferromagnetically ordered state, the barrier height is lower for up-spin electrons, leading to a highly spin-polarized tunneling current dominated by up-spin carriers. This mechanism gives rise to the spin-filtering effect. The main plot presents the electrical resistance of the SFTJ device as a function of temperature under two different current bias conditions: 10 μA (below the critical switching current, I_C) and 50 μA (above I_C). At 10 μA , the resistance decreases sharply below the Curie temperature (T_C) of the SSMO barrier (~ 100 K), consistent with the onset of ferromagnetic ordering and efficient spin filtering. In contrast, at 50 μA , the resistance increases and shows no pronounced drop near T_C , indicating that the current-induced phase transition suppresses the ferromagnetic order in the barrier. As a result, the spin-dependent tunneling is lost, and the device transitions into a non-spin-filtering regime. This comparison underscores the crucial role of magnetic phase and current bias in governing the transport and spin polarization in SFTJ devices.

A. Charge-Transfer Multiplet Calculations of Mn $L_{3,2}$ -Edge XLD Spectra

To support the orbital interpretation of the experimental Mn $L_{3,2}$ -edges XLD spectra, we performed charge-transfer multiplet cluster calculations using the CTM4XAS program¹ within the ligand-field and charge-transfer multiplet framework (Fig. S11). The calculations were carried out for Mn^{3+} ($3d^4$: $t_{2g}^3 e_g^1$), as Mn^{4+} ($3d^3$: $t_{2g}^3 e_g^0$) does not exhibit linear dichroism. The simulations were performed under D_{4h} symmetry. The cubic crystal-field splitting is described by $10Dq$, while the tetragonal distortion is parameterized by D_s and D_t , leading to energy splittings of $3D_s - 5D_t$ for t_{2g} and $4D_s + 5D_t$ for e_g levels. Preferential occupation of the $3d_{3z^2-r^2}$ ($3d_{x^2-y^2}$) orbital is achieved by positive (negative) values of D_s and D_t . The calculations include variation of key parameters: crystal-field splitting ($10Dq$), charge-transfer energy (Δ), on-site Coulomb interaction (U_{dd}), core-hole potential ($U_{pd} \approx 1.5U_{dd}$), and Mn $3d$ -O $2p$ hybridization strength $\sqrt{3}(pd\sigma)$. The parameter set used is: $10Dq = 1.7$ eV, $\Delta = 4$ eV, $\sqrt{3}(pd\sigma) = 2$ eV, $U_{dd} = 5$ eV, and $U_{pd} = 7.5$ eV. Orbital polarization is controlled via $D_s = \pm 0.16$ eV and $D_t = \pm 0.01$ eV for $3d_{3z^2-r^2}$ - and $3d_{x^2-y^2}$ - polarized states, respectively². To account for lifetime and instrumental broadening, Lorentzian (0.3 eV) and Gaussian (0.25 eV) functions were applied. Additional intrinsic broadening mechanisms likely contribute to the slightly broader experimental spectra compared to the simulations³.

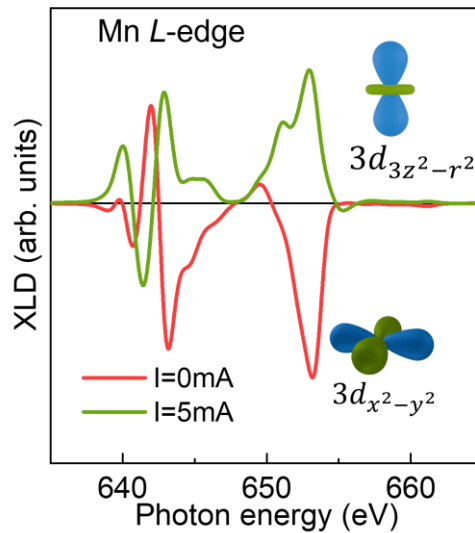


Fig. S11: Charge Transfer Multiplet Simulation: Charge-transfer multiplet simulations of Mn $L_{3,2}$ -edges XLD spectra, illustrating the distinct spectral signatures of $3d_{3z^2-r^2}$ and $3d_{x^2-y^2}$ orbital polarizations.

The following sections present a multiscale theoretical framework intended as a qualitative consistency check for the experimentally observed electrically driven inverse metamagnetic transition. The atomistic nonequilibrium Hubbard model addresses the electrically-driven evolution of magnetic order, the single-band tight-binding model connects the magnetic-state change to barrier-controlled transport, the microspin simulations illustrate the resulting field-dependent hysteresis phenomenologically, and the STT analysis shows that conventional torque alone is insufficient to account for the observed switching.

B. Atomistic Simulations

We employ a single-orbital nonequilibrium Hubbard model as a qualitative microscopic consistency check for the experimentally observed current-induced magnetic phase transition in spin-filtering devices. The Hubbard Hamiltonian model within the Hartree-Fock approximation represents a generalization of the tight-binding Hamiltonian whose on-site energies depend on the electron charge density through the Coulomb interaction term. The electron charge density, in turn, can be altered by the electric field, charge current, or temperature. The choice of the model parameters describes the electron tunnelling process, while the magnetic state depends on the critical current. To explain the main experimental features of current-induced switching of magnetic state in the magnetic insulator such a microscopic theory of magnetic phase transition has to include mechanisms of electron interactions out of thermodynamic equilibrium. For this reason, we adopt a Hubbard model on a 2d square lattice of a finite size that we treat within the Hartree-Fock approximation⁴.

$$H_C = - \sum_i \left(\frac{U_i}{2} \langle n_i^\uparrow - n_i^\downarrow \rangle + I_i + \mu_B h \right) (c_i^{\uparrow\dagger} c_i^\uparrow - c_i^{\downarrow\dagger} c_i^\downarrow) + \sum_{i,j,\sigma} t_{ij} c_i^{\dagger\sigma} c_j^\sigma,$$

where $I_i = \sum_j U_{ij} \langle n_j^\uparrow - n_j^\downarrow \rangle$, U_i and U_{ij} describe the strengths of electron-electron Coulomb interactions on an atomic site i and between atomic sites i and j respectively, $\langle n_i^\sigma \rangle$ is the average number of electrons with spin σ on an atomic site i , μ_B is the Bohr magneton, h is the applied magnetic field, t_{ij} is the electron hopping integral between atomic sites i and j , $c_i^{\dagger\sigma}$ and c_i^σ are Fermionic creation and annihilation operators with spin σ on an atomic site i . Here, we included the inter-site electron-electron interaction to describe the competing FM–AFM phase. For instance, without the inter-site Coulomb term U_{ij} , the magnetic state described by the Hamiltonian H_C is antiferromagnetic. However, when both onsite U_i and inter-site U_{ij}

interactions are present, the dominating state depends on the particular electron charge density distribution $\langle n_i^\sigma \rangle$ within the Hubbard cluster. In thermodynamic equilibrium, this quantity $\langle n_i^\sigma \rangle$ is given by the Fermi-Dirac statistic. In contrast, when the correlated electron system is out of equilibrium, this charge density distribution has to be found from non-equilibrium quantum statistics⁵.

To calculate physical properties under the charge current flow through the Hubbard cluster, we divide the system into the central region described by the Hubbard Hamiltonian H_C attached to the left and right semi-infinite leads. Therefore, the total Hamiltonian becomes

$$H = H_L + H_C + H_E + H_R + H_{bath+leads},$$

where H_L and H_R are non-interactive Hamiltonians of the left and right semi-infinite leads, $H_E = \sum_{i,\sigma} \varphi_i c_i^{\dagger\sigma} c_j^\sigma$, φ_i is the electrostatic potential across the Hubbard cluster and H_{bath} describes the coupling of H_C with the leads and thermal bath⁴. We find the electrostatic potential self-consistently by solving the Poisson equation at a given applied bias voltage. We employ the same approximation as in ref. ⁴, assuming infinite flat bands of the leads, and therefore, the coupling Hamiltonian is described by frequency-independent hybridization parameters in the leads $\Gamma_{L(R)}$ and the bath Γ_{bath} . We also assume that the left lead is magnetic, whose hybridization with the Hubbard cluster is set to be spin-dependent $\Gamma_L^\uparrow = 1.2 \text{ eV}$, $\Gamma_L^\downarrow = 0.8 \text{ eV}$, while the right lead is non-magnetic with $\Gamma_R^\uparrow = \Gamma_R^\downarrow = 1 \text{ eV}$. The coupling to the thermal bath is set to $\Gamma_{bath} = 0.01 \text{ eV}$. We calculate the retarded (advanced) Green Function from the Dyson equation

$$G(E) = (IE - H_C - H_E - \Sigma)^{-1} \equiv A^{-1},$$

where E is energy, I is the identity matrix and Σ is the self-energy due to connecting the Hubbard cluster to the leads and thermal bath. We obtain the Green function by diagonalizing the non-Hermitian matrix A .

$$G_{ij}(E) = \sum_n \frac{V_{in} V_{nj}^{-1}}{E - E_n}$$

here, E and V are eigenvalues and eigenvectors of the matrix A and from here, we omit writing the spin indices for simplicity's sake. Without applied current, the electron system in the thermodynamic equilibrium, and the charge density distribution can be obtained from the equilibrium solution⁵.

$$\langle n_i \rangle = -\frac{1}{\pi} \text{Im} \left[\sum_n V_{in} V_{nj}^{-1} \right] \int_{-\infty}^{+\infty} \frac{f_0(E - \mu) dE}{E - E_n}$$

where f_0 is the Fermi-Dirac distribution function. Integration over the energy was calculated analytically using Ozaki's representation of the Fermi-Dirac distribution function⁶. When charge current flows across the SSMO barrier, one needs to obtain the non-equilibrium Green Function to calculate the non-equilibrium charge density distribution. This can be done by solving the quantum kinetic equation⁵.

$$G^< = G_0^< + G_0 \Sigma^< G^\dagger + G_0^< \Sigma^\dagger G^\dagger + G_0 \Sigma G^<,$$

where G , $G^\dagger$ and $G^<$ are the retarded, advanced, and non-equilibrium (lesser) Green Functions and Σ , $\Sigma^\dagger$ and $\Sigma^<$ are their corresponding self-energies due to connection to the leads and thermal bath. Index 0 stands for the corresponding Green's functions without connection to the leads and the bath. The diagonal matrix elements of the non-equilibrium Green Function become

$$G_{ii}^<(E) = 2i \sum_j G_{ij}(E) \Gamma_j f_0(E - \mu_j) G_{ji}^*(E),$$

here $\Gamma_j = i(\Sigma_i - \Sigma_i^\dagger)$ and μ_j is the local electro-chemical potential. The expression for the non-equilibrium charge density in terms of the eigenvalues and eigenvectors reads

$$\langle n_i \rangle = -\frac{1}{\pi} \left[\sum_{n,m,j} V_{in} V_{nj}^{-1} \Gamma_j V_{jm}^* V_{mi}^{-1*} \right] \int_{-\infty}^{+\infty} \frac{f_0(E - \mu_j) dE}{(E - E_n)(E - E_m^*)},$$

where the integral over energy can also be evaluated analytically using Ozaki's method. This charge density has to be found by self-consistent calculations as follows. One starts from an initial guess to set up the Hamiltonian. After the Hamiltonian plus self-energies matrix is diagonalized, one calculates a new charge density, and this is continued iteratively until self-consistency is achieved. Finally, to prevent unphysical current leakage into the Fermion bath, one has to adjust electrochemical potentials after each iteration⁵. This condition for the leakage current vanishing is satisfied when

$$\sum_j \int dE G_{ij}(E) \Gamma_j f_0(E - \mu_j) G_{ji}^*(E) = - \int dE f_0(E - \mu_i) \text{Im}[G_{ii}(E)]$$

We calculate I-V characteristics of the 15x15 Hubbard cluster using the following set of model parameters: $U_i = 1 \text{ eV}$, $U_{ij} = 0.08 \text{ eV}$, $t = 0.5 \text{ eV}$, $h = 0 \text{ T}$, $T = 50 \text{ K}$. The corresponding R - V curve is shown in Fig. 4(a) of the main text.

To demonstrate that the observed resistance switching corresponds to FM – AFM phase transition, we also evaluate the averaged atomic magnetic moment

$$\langle m \rangle = \frac{\mu_B}{N} \sum_{i=1}^N \langle n_i^\uparrow - n_i^\downarrow \rangle,$$

and the averaged absolute value of the atomic magnetic moment.

$$\langle |m| \rangle = \frac{\mu_B}{N} \sum_{i=1}^N \langle |n_i^\uparrow - n_i^\downarrow| \rangle.$$

Using this framework, we performed atomistic simulations to calculate the bias-dependent evolution of both the electrical resistance and the underlying magnetic configuration, quantified through the averaged atomic magnetic moment $\langle m \rangle$ and the averaged absolute value of atomic magnetic moment $\langle |m| \rangle$ (Fig. S12a). At equilibrium (zero applied bias/no electron current), the system resides in the low-resistance state, consistent with the ferromagnetic phase inferred from our XMCD and transport measurements. As the applied bias voltage is turned on, conduction electrons start flowing through the system. In the low

bias regime, the calculated resistance remains relatively unchanged, and the charge current linearly increases with bias voltage. This spin-dependent electron current affects electron-electron interactions through the nonequilibrium electron density, leading to a metal-insulator phase transition at some critical current⁷. Thus, at a bias voltage near 0.48 V, the calculated resistance sharply rises, indicating a switch into a high-resistance state. This current-driven metal-insulator phase transition changes the magnetic configuration of the system. The magnetic moment analysis provides further insight into the underlying phase evolution. In a purely FM state, $\langle m \rangle = \langle |m| \rangle$, while in a purely AFM state, $\langle m \rangle = 0$ and $\langle |m| \rangle \neq 0$, reflecting compensated spin arrangements without net magnetization (Fig.S12b). Our simulations show that at equilibrium (no charge current) $\langle m \rangle \approx 0.3 \mu_B$, while $\langle |m| \rangle$ is only marginally ($0.05 \mu_B$) larger, indicating a dominant ferromagnetic state. As the applied bias voltage increases, but the system remains in a mostly low resistance state, both $\langle m \rangle$ and $\langle |m| \rangle$ evolve gradually until reaching a critical current at 0.48 V bias voltage, where the system switches to a mostly high resistance state where $\langle m \rangle$ abruptly decreases while $\langle |m| \rangle$ simultaneously increases. This regime, characterized by $\langle |m| \rangle \gg \langle m \rangle$, corresponds to a transition into an antiferromagnetic-like phase, consistent with the suppression of ferromagnetism observed experimentally in XMCD measurements.

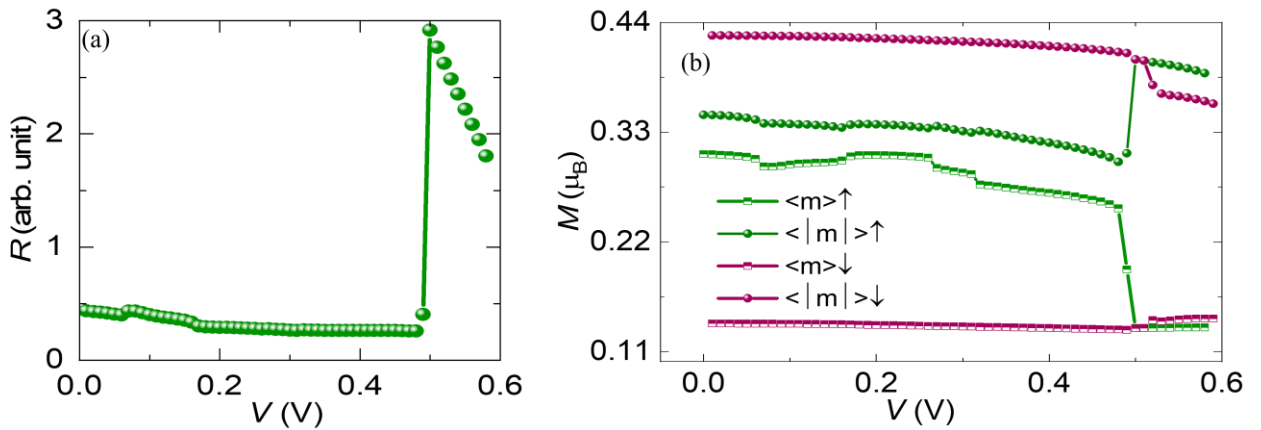


Fig. S12: Electrically driven resistance switching and Magnetization by Atomistic simulation. (a) Simulated resistance–voltage (R – V) curve for a 15×15 Hubbard cluster connected, showing a transition from a low- to high-resistance state near 0.48 V. (b) Bias dependence of the averaged atomic magnetic moment $\langle m \rangle$ and its absolute value $\langle |m| \rangle$. The

system evolves from a ferromagnetic state ($\langle m \rangle \approx \langle |m| \rangle$) to an antiferromagnetic-like state ($\langle |m| \rangle \gg \langle m \rangle$) around 0.48 V.

In equilibrium, the system is ferromagnetically ordered. When the applied current exceeds a threshold value, the nonequilibrium carrier density, ρ_{neq} , renormalizes the on-site potentials, suppressing FM ordering and stabilizing an AFM-like state. Using representative parameters (on-site $U_i = 1$ eV, intersite $U_{ij} = 0.08$ eV, hopping $t = 0.5$ eV, $T = 50$ K; hybridizations $\Gamma_L^{\uparrow,\downarrow} = 1.2, 0.8$ eV, $\Gamma_R^{\uparrow,\downarrow} = 1.0$ eV, $\Gamma_{\text{bath}} = 0.01$ eV) and solving self-consistently with the electrostatic potential, the calculated R - V curve develops a sharp jump near 0.48V, while the magnetic order parameters evolve from $\langle m \rangle \approx \langle |m| \rangle$ (uniform FM) to $\langle |m| \rangle \gg \langle m \rangle$ (compensated AFM-like), qualitatively mirroring our element-specific spectroscopy: under fixed field, Mn $L_{3,2}$ XMCD collapses above the drive threshold (loss of long-range FM) and XLD reverses sign from in-plane $3d_{x^2-y^2}$ to out-of-plane $3d_{3z^2-r^2}$, evidencing a current-induced e_g reordering in the film. This orbital interpretation, discussed in the main text, is derived from the XLD experiment and the charge-transfer multiplet analysis in Section A; it is not computed within the present single-orbital Hubbard model. The model parameters are illustrative and were chosen to reproduce the qualitative occurrence of an electrically-driven phase transition within the experimentally relevant bias range; they are not obtained from a material-specific fit.

Device scaling further points to a current (carrier-flux) threshold in the barrier: for 6.5-nm SSMO, the critical current grows approximately linearly with area ($200 \times 250 \text{ nm}^2 \rightarrow I_c \approx 14 \mu\text{A}$; $250 \times 250 \text{ nm}^2 \rightarrow 20 \mu\text{A}$; $400 \times 250 \text{ nm}^2 \rightarrow 35 \mu\text{A}$), while increasing the thickness to 8.5 nm at $250 \times 250 \text{ nm}^2$ pushes I_c to $\sim 88 \mu\text{A}$. This thickness trend is naturally captured if the transition is triggered when the injected nonequilibrium carrier density (n_{neq}), which decays into the insulator as $n_{\text{neq}}(z) \propto J e^{-z/\lambda}$ (with λ the injection/thermalization length), exceeds an integrated threshold N_{th} across the barrier: $N_{\text{th}} \approx (J\lambda/e) [1 - e^{-t/\lambda}] \Rightarrow J_c(t) \approx eN_{\text{th}}/\{\lambda[1 - e^{-t/\lambda}]\}$, so thicker barriers require larger J_c and hence larger $I_c = J_c A$. By contrast, a purely field-set threshold would give $V_c = E_c t$ and $J_c \propto V_c e^{-2\kappa t} \sim t e^{-2\kappa t}$, i.e., decreasing current density with thickness, opposite to experiment. Finally, the field dependence of the threshold—stronger fields required to drive AFM $\rightarrow$ FM at larger currents—follows from the same nonequilibrium picture: the driven carrier flux deepens the AFM free-energy minimum, so a larger Zeeman energy is needed to melt it.

Together, the XMCD/XLD signatures, NEGF–Hubbard simulations, and area/thickness scaling of I_c and J_c provide a unified, qualitatively consistent picture: an electrically driven inverse metamagnetic transition of the SSMO barrier in which current-induced redistribution of nonequilibrium carrier density suppresses FM ordering and stabilizes AFM order. Finally, we note that in a tunnel junction, current and voltage are coupled—the same bias that raises J also sets the interfacial field. The most accurate statement, therefore, is that we observe an electrically driven inverse metamagnetic transition. In practice, the phase boundary in (J, V, t, T, H) space can be crossed by ramping either J or V ; our data map it most clearly versus current density and geometry, and consistently support a flux-threshold mechanism.

C. Tight-binding calculation

To gain insight into the two-state resistance switching observed in our devices, we performed a single-band tight-binding model with the nonequilibrium Green's function formalism for a FM/I(1)/SFI1(4)/SFI2(4)/NM SFTJ as schematically presented in Supplementary Fig. S13, corresponding to the LSMO/STO(1 nm)/SSMO(8.5 nm)/LNO sample presented in Figure 2. This model bridges the microscopic phase transition to the macroscopic transport observables. By phenomenologically incorporating the FM and AFM states derived above into the tunnel barrier, we illustrate how the loss of exchange splitting leads to an abrupt increase in the effective barrier height. This qualitatively explains the step-like transition in our experimental R – I characteristics.

The system is modeled as a 1D chain along the transport direction (z-axis) with transverse momentum $k_{||}$ integrated out, described by the Hamiltonian⁵:

$$\hat{H}_{TB} = \sum_{i,\sigma} \epsilon_i^\sigma c_{i\sigma}^\dagger c_{i\sigma} - t \sum_{\langle i,j \rangle, \sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.)$$

where $c_{i\sigma}$ ($c_{i\sigma}^\dagger$) is the creation (annihilation) operator for an electron with spin σ at layer i . The spin-polarized on-site energies of left ferromagnetic (FM) and right nonmagnetic (NM) electrodes are $\epsilon_{FM}^{\uparrow(\downarrow)} = 2.6$ eV (6.0 eV) and $\epsilon_{NM} = 2.6$ eV, respectively. The barrier height of the insulator (I) barrier is $\phi_I = 1.0$ eV, and the number of atomic layers of I is $N_I = 1$. To demonstrate the Ferro-Antiferro transitions in the SSMO barrier, we consider two kinds of spin-filter insulating barriers (SF1 and SF2), and the atomic layers of SF1 and SF2 are $N_{SF1, SF2} = 4$. For the Ferromagnetic Insulating (FI) phase, the same spin-polarized barrier

heights are $\phi_{SF1,SF2}^{\uparrow,\downarrow} = 0.8 \text{ eV} \mp 0.2 \text{ eV}$. As for the Antiferromagnetic Insulating (AFI) phase, the opposite spin-polarized barrier heights are $\phi_{SF1}^{\uparrow,\downarrow} = 0.8 \text{ eV} \mp 0.2 \text{ eV}$ and $\phi_{SF2}^{\uparrow,\downarrow} = 0.8 \text{ eV} \pm 0.2 \text{ eV}$. The schematic energy diagrams for FI phase with low resistance ($I < I_c$) and AFI phase with high resistance ($I > I_c$) are displayed in supplementary Figs. S13 (a) and (b), respectively. The nearest-neighbour hopping energy is -0.83 eV in all regions.

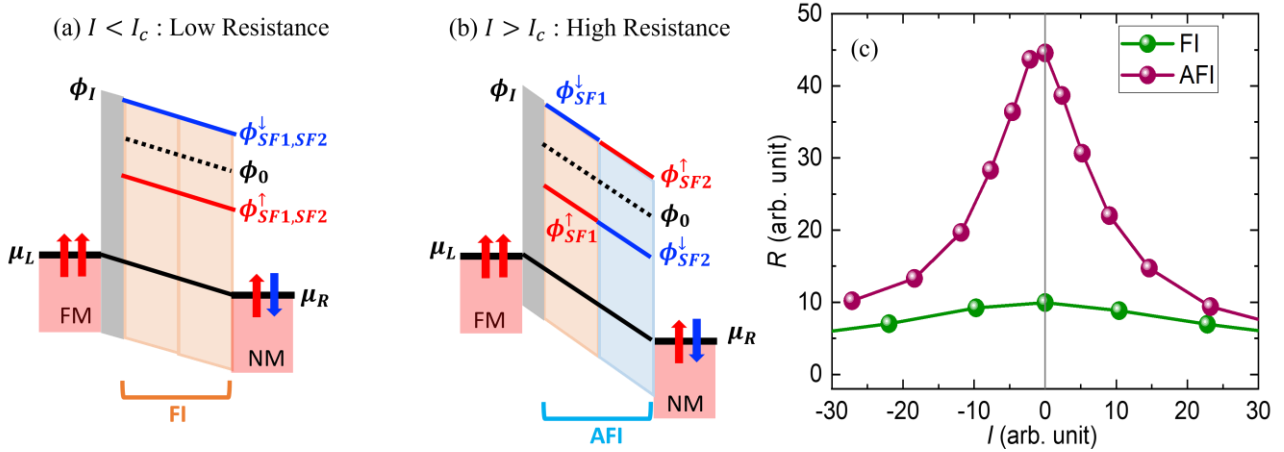


Fig. S13: Energy Band Diagrams and Transport Data for SFTJ Devices Using the Tight-Binding Calculations. To demonstrate the Ferro-Antiferro transitions in the SSMO barrier, we consider two kinds of spin-filter insulating barriers (SF1 and SF2), and the atomic layers of SF1 and SF2 are $N_{SF1,SF2} = 4$. (a) Schematic energy diagram of the spin-filter tunnel junction (SFTJ) device featuring a ferromagnetic insulating (FI) barrier under sub-critical current conditions ($I < I_c$). In this regime, the exchange-split conduction bands of the FI layer enable spin-selective tunneling, resulting in a low-resistance state. (b) Schematic energy diagram of the same SFTJ device when the applied current exceeds the critical threshold ($I > I_c$), driving the barrier into an antiferromagnetic insulating (AFI) state. The absence of spin polarization in the AFI phase leads to spin-degenerate tunneling, producing a high-resistance state. (c) Tight-binding simulations of R – I behavior for SFTJ devices with layered FM/I(1)/SF1(4)/SF2(4)/LNO structures, contrasting the transport characteristics in FI and AFI regimes.

When SFTJ is under an external bias, the spin-polarized current flows along the z direction, and the charge current, I can be obtained by the nonequilibrium Keldysh formalism⁸. The charge current density can be obtained by the non-equilibrium Keldysh formalism,

$$I = \frac{-et}{8\pi^3\hbar} \int \text{Tr}(G_{\lambda,\mu}^< - G_{\mu,\lambda}^<) dE d\mathbf{k}_{||} .$$

Here $G_{i,j}^<$ is the lesser Green's function between two sites λ and μ , $\mathbf{k}_{||}$ is the transverse component of the wave vector, and the energy integral is within the bias window. A negative bias in the model implies that the chemical potential of the left lead is higher ($\mu_L > \mu_R$), corresponding to electrons flowing from the FM (LSMO) to the NM (LNO) electrode, which perfectly matches our experimental setup. The resistance $R = dI/dV$ is calculated under an external bias between ± 0.6 V to ensure the direct tunnelling through SFI/I barriers. The simulated resistance–current characteristics (Fig. S13c) reveal a sharp increase in resistance when the barrier layer transitions from a FI to an AFI, consistent with our experimental observations. This single-band tight-binding model is intended as a phenomenological, qualitative illustration of how the loss of exchange splitting in the barrier produces the observed resistance jump; it is not intended to quantitatively reproduce the absolute experimental resistance values.

D. Microspin dynamics simulation

Building on the understanding that an external current stabilizes the AFM insulating (AFI) phase, we employ the MuMax³ package⁹ to model the competition between this current-stabilized AFI state and the external magnetic field. This phenomenological simulation successfully reproduces the asymmetric and hysteretic R–H loops observed in our SFTJ devices for $I > I_c$. The micromagnetic parameters were chosen to reflect realistic material scales but were not obtained by direct fitting to the experimental hysteresis loops. To qualitatively elucidate field-driven magnetic switching of an SFTJ device under an external current ($I > I_c$), the microspin dynamics simulation was applied to calculate the magnetic hysteresis loop of a FM/AFI bilayer. Here we simply consider two ferromagnetic LSMO (FM) layers and two SSMO layers with Antiferromagnetic Insulating (AFI) phase, the cell size of each layer is $1 \times 1 \times 1 \text{ \AA}^3$. For the FM layers, the saturation magnetization $M_{sat} = 100$ kA/m, the exchange stiffness $A_{FM} = 3 \times 10^{-16}$ J/m, and the uniaxial anisotropy energy $K_{eff} = 3$ kJ/m³. As for the SF layers, the saturation magnetization $M_{sat} = 100$ kA/m, the exchange stiffness $A_{AFI} = -3 \times 10^{-16}$ J/m for AFI phase, and the uniaxial anisotropy energy $K_{eff} = 30$ kJ/m³. Since an ultrathin STO (1nm) insulator is inserted to separate LSMO and SSMO, the much weaker exchange stiffness at FM/AFI interface is considered as $A_{inter} = 1 \times 10^{-17}$ J/m. Given the ultrathin nature of the spacer (1 nm STO) and barrier layers (5-8 nm SSMO), the local interfacial exchange coupling (A_{inter}) and the strong local anisotropy overwhelmingly dominate the switching sequence compared to the long-range

dipolar fields. To simplify the simulation and focus on the interaction between the FM and AFI layers, we disable the periodic boundary conditions and ignore the demagnetization field. The hysteresis loop was simulated by reducing an external field (H_{ext}) from $+1.5T$ to $-1.5T$ with a step size of $0.01T$. At each step, we use the function, `relax()`, of MuMax³ to optimize the magnetic configuration with minimum energy. In the present simulation, the FI→AFI transition is imposed phenomenologically by changing the sign/magnitude of the exchange parameter rather than emerging from a fully coupled transport–magnetization calculation.

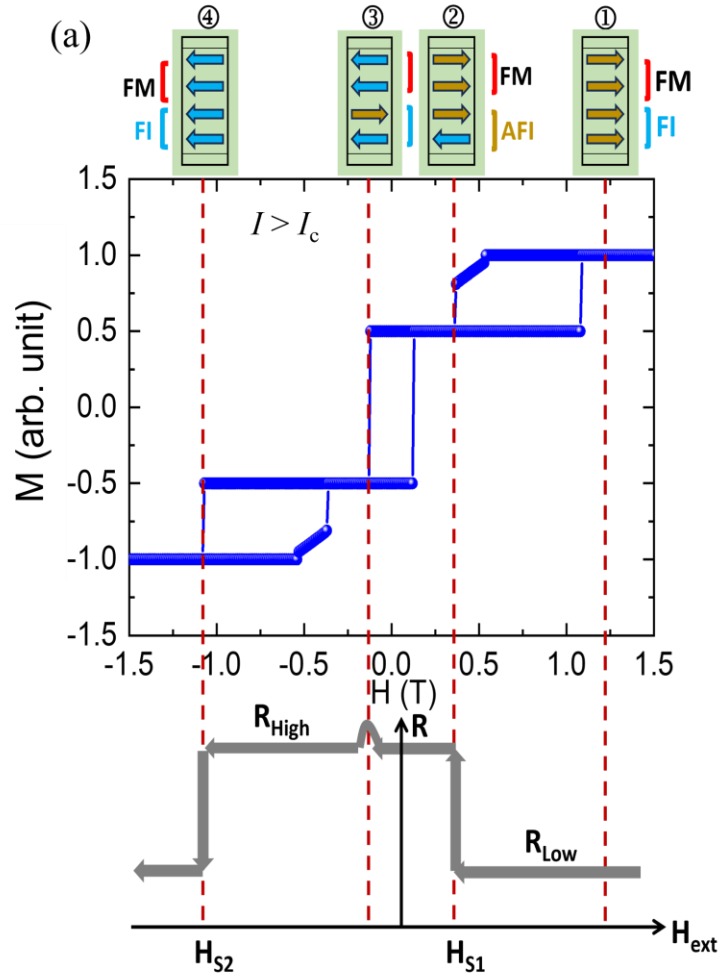


Fig. S14: Magnetization and Resistance Switching using Microspin Simulation. Microspin dynamics simulation of a magnetic hysteresis loop in a FM/AFI bilayer under $I > I_c$, showing field-driven switching between FM/I/FI and FM/I/AFI states at H_{S1} and H_{S2} , respectively. These transitions are consistent with the experimentally observed unconventional magnetoresistance (MR) behavior, as schematically depicted in the bottom panel.

Fig. S14 (top panel) presents the simulated magnetic hysteresis loop of an FM/FI bilayer under applied currents exceeding the critical threshold ($I > I_C$), where FM and FI correspond to the ferromagnetic LSMO electrode and the FM insulating SSMO-FI barrier, respectively. At large positive magnetic fields (~ 1.5 T), the moments of both LSMO and SSMO-FI align parallel, stabilizing the FM/FI configuration. As the external field is reduced to approximately 0.4 T (H_{S1}), a FI-to-AFI transition occurs within the SSMO-FI barrier, leading to an FM/AFI configuration that persists as the field is swept toward zero. Upon field reversal, the magnetization of the LSMO layer switches first due to its lower coercive field, but the FM/AFI configuration remains stable. Only when the field is further decreased beyond approximately -1.1 T (H_{S2}), the moments in the AFI barrier realign, restoring the FM/FI configuration. Such magnetic field-induced melting of the AFM phase has been widely reported in manganite systems⁷. The schematic resistance–field (R – H) characteristics, shown in the bottom panel of Fig. S14, closely replicate the unconventional MR response observed experimentally in our devices (Fig. 2e)

E. Spin-transfer torque (STT) calculations

Finally, we performed a semi-quantitative analysis of the spin-transfer torque (STT) to rigorously rule out conventional torque-driven magnetization reversal. We demonstrate that the pure STT torque is several orders of magnitude too weak to overcome the magnetic anisotropy and trigger switching at our experimental current densities.

To evaluate whether conventional current-driven STT can account for the observed transition in the SSMO barrier, we assume a two-magnetic-domain model (SF1 and SF2) and employ the generalized Landau-Lifshitz-Gilbert (LLG) equation¹⁰:

$$\frac{1+\alpha^2}{\gamma} \frac{d\hat{\mathbf{m}}}{dt} = -\hat{\mathbf{m}} \times (\mathbf{H}_{eff} + \mathbf{H}_{pre}) - \hat{\mathbf{m}} \times \hat{\mathbf{m}} \times \alpha (\mathbf{H}_{eff} + \mathbf{H}_{damp}),$$

where $\hat{\mathbf{m}}$ is the unit magnetization vector of the free domain (SF2), γ is the gyromagnetic ratio, and α is the Gilbert damping constant. At zero bias, the effective field ($\mathbf{H}_{eff}$) is governed by the external field and magnetocrystalline anisotropy.

Under an applied current density (J), the current-induced effective fields are defined as the damping-like field, $H_{damp} \sim \frac{H_{STT}}{\alpha}$, and the field-like (precession) field, $H_{pre} \sim -\alpha H_{STT}$. These current-induced H_{STT} is determined by the current-driven spin torques (STT) via $\mu_0 H_{STT} =$

$\frac{STT}{M_s L_{SF2}}$, where M_s is the saturation magnetization and L_{SF2} is the thickness of the switching domain (SF2). Within the TB-NEGF framework, the spin-transfer torque acting on SF2 is obtained from the divergence of the spin current across the switching layer. Since the spin torques are proportional to the applied current, we can define the current-driven spin torque as:

$$(\tau)^{TB} = \frac{\hbar}{|e|} \epsilon_{STT} = \frac{STT}{J}.$$

Theoretically, the spin current accumulation (Q_{iz}) under an external bias is obtained via the nonequilibrium Keldysh formalism⁵:

$$Q_{iz} = \frac{t}{16\pi^3} \int Tr[(G_{\lambda,\mu}^< - G_{\mu,\lambda}^<) \sigma_i] dE d\mathbf{k}_{||},$$

where i denotes the spin direction. The current-driven spin-transfer torque (STT) acting on SF2 is defined in the direction of $\hat{m}_{SF2} \times (\hat{m}_{SF1} \times \hat{m}_{SF2})$. We present in Figures S15(a) and S15(b) the calculated charge current density J (A/area) along with STT (eV/area) and the damping-like field, $\mu_0 H_{damp}^{TB} \sim \frac{\mu_0 H_{STT}}{\alpha} = \frac{(\tau)^{TB}}{\alpha}$, respectively. The calculated τ/α is in the order of $10^4 \cdot \frac{e}{\hbar}$ eV/A, where e is the elementary charge, $\hbar$ is the reduced Planck constant, and $\alpha = 0.01$ is chosen for the damping constant for SF2.

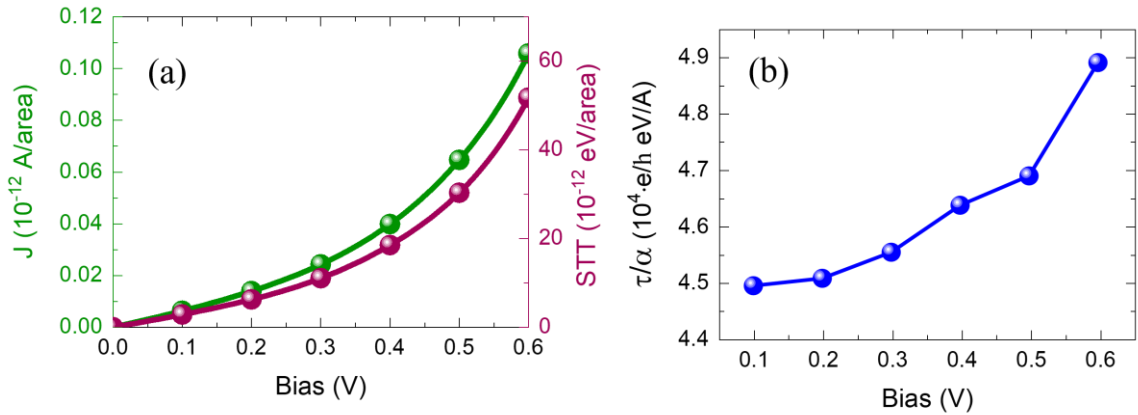


Fig. S15: TB calculated STT of M/I/FI1/FI2($\theta=\pi/2$)/NM SFTJ Device. The bias dependence of (a) charge current density J (green line) and spin-transfer torque STT (red line). Both quantities are presented in units normalized by the lateral cross-sectional area of the user-defined computational two-dimensional unit cell. (b) The bias dependence of the damping-like field, $\mu_0 H_{damp}^{TB} = \frac{(\tau)^{TB}}{\alpha}$ (blue line). Notice that the current-driven spin

torquance is defined as $(\tau)^{TB} = \frac{STT}{J}$. Because both STT and J are normalized by the exact same unit cell area, this geometric factor algebraically cancels out. Consequently, the resulting effective field shown in (b) is an intensive physical quantity that is strictly independent of the area normalization.

Conversely, to activate the transition (analogous to overcoming the barrier in an FI-to-AFI switching event), the current-driven damping field at the critical current (I_c) must overcome the effective anisotropy field of the SF2 layer, $\mu_0 H_{damp} \sim \mu_0 H_{SSMO} = 0.075$. From our experimental parameters, the required torquance is extracted as:

$$(\tau)^{exp} = \frac{\hbar}{|e|} \epsilon_{SF2} \sim \frac{\mu_0 H_{SSMO} \cdot M_s \cdot L_{SF2}}{I_c / A},$$

where $A = 1.8 \times 10^{-13} \text{ m}^2$ represents the lateral cross-sectional area of the device, $M_s = 3.7 \times 10^5 \text{ A/m}$ is the saturation magnetization, and $L_{SF2} = 4 \times 10^{-9} \text{ m}$ is the thickness of the SF2 (SSMO) layer.

Consequently, the experimentally required effective damping-like field is evaluated as $\mu_0 H_{damp}^{exp} \sim (\tau)^{exp} / \alpha \sim 1.42 \times 10^8 \cdot \frac{e}{\hbar} \text{ eV/A}$. This threshold value is three to four orders of magnitude larger than our theoretically calculated field, $\mu_0 H_{damp}^{TB}$. This massive discrepancy definitively demonstrates that conventional spin-transfer torque (STT) alone is fundamentally insufficient to actuate the magnetic switching observed in our SFTJ devices, thereby providing robust quantitative evidence for the electrically driven inverse metamagnetic transition mechanism. Reasonable variation of damping and magnetic parameters does not change the conclusion that the STT-generated effective field remains orders of magnitude below the experimentally required threshold.

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