

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

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have addressed my questions with additional data and discussions. The manuscript has been well improved. I would recommend the publication.

Reviewer #3

(Remarks to the Author)

See attached.

Version 1:

Reviewer comments:

Reviewer #3

(Remarks to the Author)

The revised Supplementary Information is substantially improved and now addresses my main concerns regarding scope, definitions, and internal consistency: the authors clearly state that the modeling is intended as a qualitative/phenomenological multiscale consistency check; they explicitly separate the (e_g) orbital interpretation (supported via charge-transfer multiplet calculations of the XLD) from the single-orbital nonequilibrium Hubbard model; and they now provide explicit formulations for the single-band TB model and the STT/LLG analysis, including a clearer explanation of what is meant by “TB-calculated STT” and how the “area” normalization is treated. Overall, this is close to sufficient for a qualitative-theory support role. However, a small round of technical editing is still needed to remove remaining inconsistencies/typos (e.g., “0.48 eV” vs “0.48 V” for the bias threshold; inconsistent bilayer/trilayer descriptions in the microspin section/figure; and coordinate/transport-direction wording in the TB section), as these directly affect clarity and reproducibility.

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Responses to the reviewer

We sincerely thank the Editor and all the Reviewers for the careful evaluation of our manuscript and for their valuable comments and suggestions. We are grateful to **Reviewer #1** for recommending acceptance of the revised manuscript for publication. We also particularly thank **Reviewer #3**, whose detailed comments have helped us substantially improve the clarity, scope, and internal consistency of the theoretical framework. We would like to emphasize that this work is primarily experimental in nature, while the theoretical analysis is included mainly to provide qualitative support and physical insight into the experimental observations. Because strongly correlated oxide systems, especially under non-equilibrium conditions, are intrinsically complex, developing a fully quantitative, material-specific theoretical description remains highly challenging at present. In response to these comments, we have revised both the main manuscript and the Supplementary Information to make the modeling more explicit, transparent, and appropriately scoped. In particular, we now state clearly that the theory is intended as a multiscale qualitative consistency check, rather than as a fully predictive, material-specific treatment of this non-equilibrium correlated oxide system. We have also added explicit model definitions, clarified the provenance of the parameters, separated the orbital interpretation from the single-orbital Hubbard model, and improved the discussion of conventions, units, and normalization throughout the Supplementary Information.

In addition, we have incorporated a new charge-transfer multiplet analysis of the Mn $L_{3,2}$ -edges XLD spectra. These calculations, performed within a D_{4h} crystal-field framework, reproduce the characteristic spectral signatures of the two orbital-polarization states and support our interpretation of the XLD data (see Supplementary Information, Fig. S1 and Section A). We have further reorganized the theoretical sections so that they now form a more transparent multiscale framework spanning the XLD multiplet analysis, the atomistic nonequilibrium Hubbard model, the phenomenological tight-binding transport model, the microspin simulations, and the STT analysis.

For ease of reference, all changes in the revised manuscript and Supplementary Information have been highlighted. Below, we provide our point-by-point responses to each comment, together with a brief description of the corresponding revisions made in the manuscript and Supplementary Information.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

Comment: *The authors have addressed my questions with additional data and discussions. The manuscript has been well improved. I would recommend the publication.*

Response: We sincerely thank the Reviewer for the positive evaluation of our revised manuscript. We are grateful for the Reviewer's careful reading and constructive comments, which have helped us improve the manuscript significantly. We appreciate the Reviewer's recommendation for publication.

Reviewer #3 (Remarks to the Author):

Referee comments (theory-focused)

Generic Comment # *The comments below are restricted to the theoretical part of the manuscript and its connection to the experimental interpretation. My main goal is to ensure that (i) the theoretical models are stated unambiguously (Hamiltonians/equations, conventions, units), and (ii) the claims made from the theory are supported by the level of modeling employed.*

Response: We sincerely thank the Reviewer for the rigorous evaluation and constructive feedback on the theoretical framework presented in our manuscript. The Reviewer's precise and insightful comments have significantly helped us clarify the mathematical formulations, parameter choices, conventions, and the intended scope of our computational models. To address the Reviewer's central concern directly, we now state explicitly that our theoretical framework is intended primarily as a qualitative consistency check and a phenomenological multiscale illustration of the underlying physical mechanism. Owing to the intrinsic complexity of non-equilibrium strongly correlated oxide systems, a fully predictive, first-principles quantitative treatment remains computationally prohibitive at present. Instead, our objective is to show that the experimentally observed electrically driven inverse metamagnetic transition is physically well grounded and consistently supported across multiple theoretical length scales. Accordingly, we have thoroughly revised both the main manuscript and the Supplementary Information. In particular, we have added explicit Hamiltonians and equations, clarified the physical assumptions and parameter provenance, distinguished clearly between qualitative and phenomenological modeling, and corrected typographical and notation-related issues throughout. We have also reorganized the theoretical discussion so that the different modeling components now form a more

transparent and coherent multiscale framework. We believe that, with these revisions, the theoretical section is now substantially clearer, better scoped, and more rigorously connected to the experimental interpretation. Below, we provide a point-by-point response to the Reviewer's comments on Sections (A), (B), (C), and (D).

(A) Tight-binding NEGF transport

The authors introduce a tight-binding transport model (combined with NEGF/Keldysh) intended to capture resistance changes between different magnetic configurations of the barrier. The setup is described as a multilayer stack (FM / I / SF11 / SF12 / NM) with prescribed on-site energies, barrier heights, layer counts, and a uniform nearest-neighbour hopping.

Response: At the mesoscopic scale, we use a single-band tight-binding model combined with the nonequilibrium Green's function (NEGF/Keldysh) formalism to connect the current-driven magnetic-state transition in the barrier to the experimentally observed transport response. In this framework, the FM and AFM-like barrier states are incorporated phenomenologically through their different effective spin-dependent barrier heights. This allows us to show how the suppression of exchange splitting in the barrier leads to an abrupt increase in resistance, thereby providing a qualitative explanation for the step-like R - I switching observed experimentally. The purpose of this model is therefore not quantitative prediction of the absolute device resistance, but rather a transparent mesoscopic illustration of how the magnetic-state change in the barrier is reflected in the transport characteristics.

Questions/points requiring clarification

Reviewer Comment A1: *Is the TB model explicitly defined (Hamiltonian + geometry)? The manuscript/SI provides a parameter list, but I could not find a clearly written Hamiltonian and geometry (dimensionality, lattice, how bias enters, how lead self-energies are implemented). Please write the explicit TB Hamiltonian used and define all conventions so the model is reproducible.*

Response: We thank the Reviewer for pointing out the need for a more explicit definition of the tight-binding model. In the revised Supplementary Information, we now describe the transport problem as a single-band one-dimensional chain along the transport direction (z -axis), with the transverse momentum $k_{\parallel}$ integrated out. The explicit single-band tight-binding Hamiltonian is now written as:

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

Here $c_{i\sigma}$ ($c_{i\sigma}^\dagger$) denotes the annihilation (creation) operator for an electron with spin σ at layer i . The layer-dependent spin-polarized onsite energy ϵ_i^σ includes the local electrostatic potential profile under applied bias and is specified according to the corresponding region in the multilayer stack (NM, FM, SF1, SF2, or I). The nearest-neighbour hopping parameter is taken as $t = -0.83$ eV. We have also clarified the model geometry, the lead assignment, and the bias convention in the revised Supplementary Information so that the full transport setup is stated unambiguously. These changes have been incorporated in the Supplementary Information (Section C) and highlighted.

Reviewer Comment A2: *Single-band nature and scope of claims. Please clarify the intended scope: are the TB results meant to be purely qualitative (illustrative trends), or are they used as quantitative support for the experimental magnitudes?*

Response: We thank the Reviewer for raising this important point. We have now clarified in the revised Supplementary Information that the single-band tight-binding model is intended purely as a qualitative, phenomenological illustration. Its role is to demonstrate how the suppression of exchange splitting in the barrier, corresponding to the transition from a ferromagnetic insulating (FI) state to an antiferromagnetic insulating (AFI) state, modifies the effective spin-dependent barrier heights and thereby produces a macroscopic resistance jump. It is therefore not intended as a quantitative, first-principles prediction of the absolute experimental resistance values or switching magnitudes. These clarifications have been incorporated in the Supplementary Information (Section C) and highlighted.

Reviewer Comment A3: *Bias polarity and mapping to the experimental convention. Please state clearly which lead corresponds to which experimental electrode and how the sign of bias/current in the model maps to the experimental bias convention.*

Response: We thank the Reviewer for this helpful suggestion. We have now clarified the bias convention explicitly in the revised Supplementary Information. In our model, a negative bias corresponds to the condition $\mu_L > \mu_R$, i.e., the chemical potential of the left lead is higher than that of the right lead. The left lead represents the ferromagnetic LSMO electrode, while the right lead represents the nonmagnetic LNO electrode. Therefore, under negative bias, electrons flow from LSMO to LNO, which is fully consistent with the polarity convention used in our experimental measurements. This

clarification has been incorporated in the Supplementary Information (Section C) and highlighted.

(B) Atomistic NEGF–Hubbard

Generic Comment # *The authors also employ an “atomistic” NEGF–Hubbard approach on a finite 2D lattice treated at Hartree–Fock level, coupled to leads (including a magnetic lead) and to an additional bath intended to model thermalization/relaxation.*

Response: At the microscopic scale, we employ a single-orbital nonequilibrium Hubbard model on a finite two-dimensional square lattice, treated within the Hartree–Fock approximation and coupled to left and right leads. The purpose of this atomistic NEGF–Hubbard framework is to examine whether a bias-driven redistribution of nonequilibrium carrier density can qualitatively account for the experimentally observed suppression of ferromagnetic order and the emergence of a high-resistance AFM-like state. We emphasize that this model is intended as a qualitative microscopic consistency check, rather than as a fully predictive, material-specific description. Accordingly, in the revised Supplementary Information, we have clarified the model assumptions, stated the single-orbital nature of the model unambiguously, and refined the discussion so that orbital interpretations from XLD are clearly separated from what is computed within the Hubbard framework.

Questions / points requiring clarification

Reviewer Comment B1: *Single-orbital nature of the Hubbard model. As written, the Hubbard model appears to include only site and spin indices (no orbital index), i.e. it is effectively single-orbital. Please confirm this explicitly.*

Response: This is correct. The Hubbard model used in our work includes only site and spin indices and therefore, is indeed a single-orbital model. We now state this explicitly in the revised Supplementary Information and clarify that the atomistic NEGF–Hubbard framework is intended as a qualitative microscopic consistency check for the experimentally observed electrically driven magnetic phase transition. This clarification has been incorporated in the Supplementary Information (Section B) and highlighted.

Reviewer Comment B2: *Consistency with claims about e_g orbital splitting / inversion. In the discussion around Fig. S1 the text argues in terms of a hierarchy (and even*

inversion) between $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$. Please clarify how such orbital-resolved statements are supported by a single-orbital Hamiltonian. If the e_g discussion is an experimental interpretation, please clearly separate it from what is computed in the model and soften any wording that implies the model computes orbital inversion.

Response: We thank the Reviewer for this important and helpful comment. We agree that a single-orbital Hubbard model cannot directly describe the hierarchy or inversion between the $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$ orbitals. The discussion of orbital polarization in the manuscript is therefore intended as an experimental interpretation based on the XLD measurements, rather than as a direct output of the single-orbital NEGF–Hubbard model. To make this distinction clear, we have revised the manuscript and the Supplementary Information so that the orbital interpretation is explicitly separated from the atomistic Hubbard-model results. In particular, we now state clearly that the single-orbital Hubbard model is used only to capture the qualitative current-driven magnetic phase transition and is not intended to compute orbital hierarchy or orbital inversion. We have also softened the wording wherever necessary to avoid implying that the Hubbard model directly yields orbital-resolved information.

In addition, to support the orbital interpretation of the XLD data more appropriately, we have incorporated charge-transfer multiplet calculations of the Mn $L_{3,2}$ -edges XLD spectra. These calculations, performed within a D_{4h} crystal-field framework, reproduce the characteristic spectral signatures of the two orbital-polarization states and support our interpretation of the XLD spectra (see **Supplementary Information, Fig. S1 (as shown below) and Section A**). These revisions have been incorporated into the revised manuscript and Supplementary Information (Sections A and B) and highlighted.

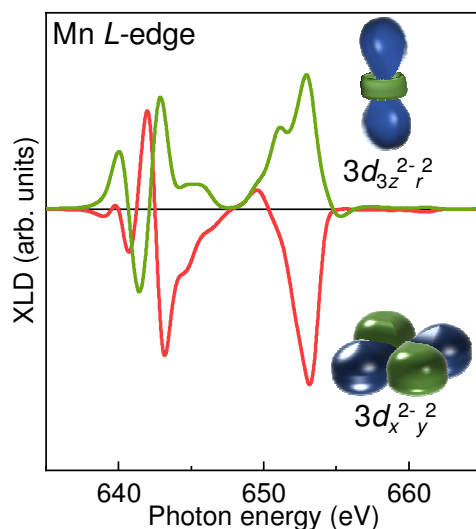


Fig. S1: Charge-transfer multiplet simulations of Mn $L_{3,2}$ -edges XLD spectra, illustrating the distinct spectral signatures of $3d_{3z^2-r^2}$ and $3d_{x^2-y^2}$ orbital polarizations.

Reviewer Comment B3: Parameter justification and mapping to experiment. The SI provides a particular parameter set (e.g. t , U , intersite terms, temperature) and lead/bath couplings. Please state whether these are constrained by material scales (or fitted to experiment), or whether they are illustrative. If illustrative, please avoid language implying “microscopic evidence” for orbital-level rearrangements.

Response: We thank the Reviewer for this important suggestion. The parameters used in the Hubbard Hamiltonian are illustrative and were not directly fitted to the experimental data. Their values were chosen to qualitatively reproduce the occurrence of an electrically-driven phase transition within the experimentally relevant voltage/current range. In the revised manuscript and Supplementary Information, we have clarified this point explicitly and have revised the wording accordingly to avoid implying that the model provides microscopic evidence for orbital-level rearrangements. Instead, the atomistic NEGF–Hubbard framework is now presented as a qualitative microscopic consistency check for the experimentally observed electrically driven magnetic phase transition. These clarifications have been incorporated in the Supplementary Information (Section B) and highlighted.

(C) Micromagnetic (MuMax3) simulations: what the authors did

Generic Comment # The SI reports micromagnetic simulations aimed at reproducing the field-dependent magnetoresistance (and a current-threshold-induced transition) by modelling the magnetic layers and the barrier in different magnetic states.

Response: At the macroscopic scale, we employ the MuMax³ micromagnetic simulation package to examine the competition between the externally applied magnetic field and the current-stabilized antiferromagnetic insulating (AFI) state in the barrier. The purpose of this simulation is to provide a phenomenological illustration of how the field-dependent magnetic configurations can give rise to the unconventional hysteretic R – H behavior observed experimentally in our SFTJ devices for $I > I_c$. In this way, the micromagnetic model serves as a qualitative bridge between the current-induced AFI state inferred from the microscopic picture and the macroscopic magnetoresistance response measured in the devices.

Questions / points requiring clarification

Reviewer Comment C1: Please state clearly whether the micromagnetic parameters (M_s , exchange stiffness A , anisotropy K_u , interfacial coupling) are taken from literature for the relevant materials/temperature, extracted from experimental loops, or tuned to reproduce specific features of the measured curves.

Response: The micromagnetic simulation is intended as a phenomenological, qualitative illustration of the field-driven switching behavior under an external current, rather than as a quantitative fit to the experimental hysteresis loops. Accordingly, the micromagnetic parameters—such as the saturation magnetization ($M_{sat} = 100$ kA/m) exchange stiffness ($A_{FM} = 3 \times 10^{-16}$ J/m³) and uniaxial anisotropy energy (3 kJ/m³) were chosen to reflect realistic material scales and to capture the fundamental competition between the current-stabilized AFI state and the external magnetic field. They were not extracted through direct quantitative fitting of the SQUID or transport data. We have clarified this point explicitly in the revised Supplementary Information. These changes have been incorporated in the Supplementary Information (Section D) and highlighted.

Reviewer Comment C2: Model choices and physical consistency. The simulations neglect demagnetization fields and use simplified boundary conditions. Please justify these choices and discuss how they impact hysteresis/switching fields.

Response: We thank the Reviewer for raising this important point. The neglect of demagnetization fields and the use of simplified boundary conditions are motivated by the specific geometry of our junction and the qualitative purpose of the micromagnetic simulation. In our heterostructure, the spacer layer is ultrathin (1 nm STO) and the SSMO barrier layers are also very thin (5–8 nm). Under these conditions, the switching sequence is expected to be governed primarily by the local interfacial exchange coupling (A_{inter}) and the strong local anisotropy, while the contribution from long-range dipolar fields is comparatively weaker. Accordingly, we disabled periodic boundary conditions and neglected the demagnetization field in order to focus on the dominant local interactions that control the hysteresis sequence. We have now clarified this physical justification in the revised Supplementary Information and have also noted that, within this phenomenological treatment, the simulation is intended to capture the qualitative form of the hysteresis and switching behavior rather than the exact quantitative values of the coercive or switching fields. These changes have been incorporated in the Supplementary Information (Section D) and highlighted.

Reviewer Comment C3: How the FI→AFI transition is implemented. Please clarify whether the FI→AFI transition is emergent from a coupled model or imposed by switching parameters/state by hand. If imposed, the results should be framed as phenomenological illustration rather than predictive validation.

Response: In the present MuMax³ treatment, the FI→AFI transition is imposed phenomenologically, rather than emerging from a fully coupled transport–magnetization model. Specifically, the transition is implemented by changing the sign/magnitude of the exchange stiffness parameter A to represent the AFI state in the barrier. The purpose of this micromagnetic simulation is therefore not predictive validation of the transition itself, but rather a phenomenological illustration of how the magnetic hysteresis and the resulting R – H behavior would evolve once the barrier is driven into the AFI state. We have revised the text accordingly to make this scope explicit and to avoid overstating the predictive role of the micromagnetic model. These changes have been incorporated in the Supplementary Information (Section D) and highlighted.

(D) Spin-transfer torque (STT) analysis

Generic Comment # The SI includes an STT-based estimate framed in terms of a generalized LLG equation and introduces a torque-like quantity to assess whether STT can account for the observed switching thresholds.

Response: Finally, we carried out a semi-quantitative spin-transfer torque (STT) analysis to assess whether conventional current-driven torque alone could account for the observed switching thresholds. Within this framework, we compare the experimentally required effective damping-like field with the STT-induced field estimated from the model. This analysis shows that the pure STT torque is several orders of magnitude too small to overcome the magnetic anisotropy of the switching layer at the experimentally relevant current densities. The purpose of this section is therefore to demonstrate that, although STT may contribute to the overall dynamics, it is insufficient by itself to explain the observed switching, thereby supporting the interpretation in terms of an electrically driven inverse metamagnetic transition.

Questions / points requiring clarification

Reviewer Comment D1: Equations: please write them explicitly. I could not find the

generalized LLG equation written out with the exact STT term used (damping-like only? any field-like term? sign conventions. Would the term be Slonczewski-like?). Please provide the explicit equation(s) of motion and define every symbol.

Response: We thank the Reviewer for pointing out the need for a more explicit presentation of the STT formalism. In the revised Supplementary Information, we now write the generalized Landau–Lifshitz–Gilbert (LLG) equation explicitly as:

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

Here, $\hat{m}$ is the unit magnetization vector of the free domain (SF2), γ is the gyromagnetic ratio, and α is the Gilbert damping constant. Under an applied current density, the current-induced effective fields are defined by a damping-like field,

$H_{damp} \sim \frac{H_{STT}}{\alpha}$, and a field-like (precession) field, $H_{pre} \sim -\alpha H_{STT}$. The current-driven

spin torques (STT) through

$$\mu_0 H_{STT} = \frac{STT}{M_S L_{SF2}},$$

where M_S is the saturation magnetization and L_{SF2} is the thickness of the switching domain (SF2). Since the spin torques are proportional to the applied current density, we can define the current-driven spin torque as:

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

We have also clarified in the revised text that this formulation is used as a semi-quantitative measure of the damping-like and field-like contributions within our TB-NEGF-based STT estimate. These revisions have been incorporated in the Supplementary Information (Section E) and highlighted.

Reviewer Comment D2: *What exactly is “TB-calculated STT” in Fig. S4? Fig. S4 refers to TB-calculated STT. Please clarify how STT is extracted from the TB-NEGF framework (e.g. from spin-current divergence, a specific Q or β quantity, etc.), and provide the explicit definition used in the figure caption or the text.*

Response: We thank the Reviewer for requesting this clarification. In the revised Supplementary Information, we now explain explicitly that the “TB-calculated STT” is obtained within the tight-binding nonequilibrium Green’s function (TB-NEGF) framework from the spin-current accumulation across the switching layer. Specifically, the spin current accumulation (Q_{iy}) under an external bias is evaluated using the

nonequilibrium Keldysh formalism⁵:

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

where i denotes the spin direction, t is the hopping parameter, $G^<$ is the lesser Green's function, and σ_i is the corresponding Pauli matrix. The spin-transfer torque (STT) acting on the switching layer SF2 is then defined from the relevant spin-current component as

$$STT = Q_{zy}.$$

In the revised figure caption and text, we now state this definition explicitly. Figures S5(a) and S5(b) present the calculated charge current density $J(\text{A/area})$, the corresponding STT (eV/area), and the associated 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. These clarifications have been added to the revised Supplementary Information and figure caption so that the meaning of “TB-calculated STT” is now stated unambiguously. These changes have been incorporated in the Supplementary Information (Section E) and highlighted.

Reviewer Comment D3: Parameter justification (e.g. damping α and material inputs). The STT conclusion depends on assumed parameters. Please justify the chosen values (literature/experiment) and, ideally, provide a sensitivity range showing that the conclusion (STT too small / too large) is robust.

Response: In our STT estimate, we use a Gilbert damping constant of $\alpha=0.01$, which represents a standard and conservative value for oxide magnetic layers. To assess whether conventional STT can account for the experimentally observed switching, we compare the experimentally required torque with the TB-calculated torque. From the experimental device parameters, the required torque is estimated as:

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

where $A = 1.8 \times 10^{-13} \text{ m}^2$ is the lateral area of the device, $M_s = 3.7 \times 10^5 \text{ A/m}$ and $L_{SF2} = 4 \times 10^{-9} \text{ m}$ is the thickness of the switching layer (SF2, SSMO). The corresponding experimentally required effective damping-like field is then

$$\mu_0 H_{damp}^{exp} \sim (\tau)^{exp} / \alpha \sim 1.42 \times 10^8 \cdot \frac{e}{\hbar} \text{ eV/\AA}$$

This required value is three to four orders of magnitude larger than the theoretically calculated $\mu_0 H_{damp}^{TB}$. Therefore, even allowing for reasonable variation in the damping constant and other magnetic parameters, the central conclusion remains unchanged: conventional STT alone is far too weak to account for the observed switching threshold. We have clarified this point in the revised Supplementary Information and emphasized that the STT analysis is intended as a robust semi-quantitative comparison rather than an exact threshold fit. These changes have been incorporated in the Supplementary Information (Section E) and highlighted.

Reviewer Comment D4: Units and normalization. The figure(s) normalize by “area” without specifying what unit/normalization is used (device area, cross-section in the TB model, unit-cell area, etc.). Please state the area definition and provide standard units (e.g. A/cm2 for current density; torque per unit moment, etc.) so the plot is interpretable.

Response: We thank the Reviewer for pointing out the need for greater clarity regarding the normalization units. In the revised Supplementary Information, we now state explicitly that, in Fig. S5(a), the tight-binding (TB) calculated charge current density J (green line) and spin-transfer torque STT (red line) are expressed in units of A/area and eV/area, respectively. Here, “area” refers to the lateral cross-sectional area of the user-defined two-dimensional unit cell in the computational model. Importantly, the current-driven spin torque is defined as:

$$(\tau)^{TB} = \frac{STT}{J}.$$

Because both STT and J are normalized by the same unit-cell area, this geometrical factor cancels exactly. As a result, the corresponding damping-like field,

$$\mu_0 H_{damp}^{TB} \sim \frac{\mu_0 H_{STT}}{\alpha} = \frac{(\tau)^{TB}}{\alpha},$$

is an intensive quantity and is therefore independent of the specific area normalization used in the calculation.

To remove ambiguity, we have now added this explanation explicitly to both the revised text and the caption of Fig. S5, including the definition of the area normalization and the reason why the final effective field does not depend on it. These clarifications have been incorporated in the Supplementary Information (Section E) and highlighted.

Generic Comment # *Typos / symbol encoding issues. The SI contains multiple typos and symbol-encoding artifacts (e.g. corrupted characters / question marks). Please correct these throughout.*

Response: We thank the Reviewer for highlighting these presentation issues. We have carefully revised the Supplementary Information to correct typographical errors, improve notation consistency, and remove symbol-encoding artifacts throughout the document.

Generic Comment # *Final remarks. In its present form, the theoretical part of the SI reads as a set of largely independent modelling exercises rather than a unified framework tied transparently to the experiment. Critical elements are missing for reproducibility and evaluation: explicit model equations (TB Hamiltonian and generalized LLG with STT), parameter provenance (fit vs literature vs illustrative), consistent unit normalization (including the “area” used in Fig. S4) etc. As a result, it is difficult to determine whether the theory is intended as a qualitative consistency check or a quantitatively predictive description. If the paper’s conclusions depend on quantitative, material-specific support, a major revision of the theoretical sections is required.*

Response: We thank the Reviewer for this important overarching comment. We would like to emphasize that this work is primarily experimental in nature, while the theoretical analysis is included mainly to provide qualitative support and physical insight into the experimental observations. Strongly correlated oxide systems, particularly under non-equilibrium conditions, are intrinsically complex, and developing a fully quantitative, material-specific theoretical description remains highly challenging at present. Accordingly, the purpose of the theoretical framework in this study is not to provide an exact quantitative simulation of the experimental system, but rather to demonstrate that the experimentally observed electrically driven inverse metamagnetic transition is physically plausible and qualitatively consistent across multiple theoretical length scales.

In the revised version, we now state explicitly that the theoretical framework is intended as a multiscale qualitative consistency check, rather than as a quantitatively predictive, material-specific description of this non-equilibrium correlated oxide system. To make this scope clear, we have added an introductory framing paragraph describing the role of each modeling level and its connection to the experiment. We have also included

explicit model equations, including the tight-binding Hamiltonian and the generalized LLG equation with STT terms; clarified parameter provenance by distinguishing between illustrative/phenomenological choices and experimentally motivated quantities; improved the discussion of conventions, units, and normalization, including the definition of the “area” used in the STT analysis; and corrected typographical and notation-related issues throughout.

In addition, we have revised the wording in both the main manuscript and the Supplementary Information to ensure that the claims made from each model are fully consistent with the actual level of modeling employed. We believe that these revisions make the theoretical section substantially clearer, more transparent, and more rigorously connected to the experimental interpretation. These changes have been incorporated in the revised manuscript and Supplementary Information and highlighted.

Responses to the reviewer

We sincerely thank the Editor and **Reviewer #3** for the careful evaluation of our revised manuscript and Supplementary Information. We are grateful that Reviewer #3 finds the revised Supplementary Information substantially improved and considers the theoretical framework close to sufficient for its intended qualitative-support role. We also appreciate the reviewer's remaining technical comments, which helped us further improve the clarity, consistency, and reproducibility of the theoretical sections.

In response to the reviewer's comment, we have carefully checked the revised manuscript and Supplementary Information and corrected the remaining inconsistencies and typographical issues. All corresponding changes have been highlighted in the revised manuscript and Supplementary Information. Below, we provide our point-by-point response of reviewer comment.

REVIEWER COMMENTS

Reviewer #3 (Remarks to the Author):

Comment: *The revised Supplementary Information is substantially improved and now addresses my main concerns regarding scope, definitions, and internal consistency: the authors clearly state that the modeling is intended as a qualitative/phenomenological multiscale consistency check; they explicitly separate the (e_g) orbital interpretation (supported via charge-transfer multiplet calculations of the XLD) from the single-orbital nonequilibrium Hubbard model; and they now provide explicit formulations for the single-band TB model and the STT/LLG analysis, including a clearer explanation of what is meant by “TB-calculated STT” and how the “area” normalization is treated. Overall, this is close to sufficient for a qualitative-theory support role. However, a small round of technical editing is still needed to remove remaining inconsistencies/tipos (e.g., “0.48 eV” vs “0.48 V” for the bias threshold; inconsistent bilayer/trilayer descriptions in the microspin section/figure; and coordinate/transport-direction wording in the TB section), as these directly affect clarity and reproducibility.*

Response:

We sincerely thank Reviewer #3 for the positive assessment of the revised Supplementary Information and for recognizing the improvements made to the scope, definitions, and internal consistency of the theoretical framework. We are pleased that the reviewer considers the revised theoretical analysis close to sufficient for its intended

qualitative and phenomenological support role. We also thank the reviewer for pointing out the remaining technical inconsistencies and typographical issues. We have now carefully revised the Supplementary Information to address these points.

Referee comments (theory-focused)

The comments below are restricted to the *theoretical part* of the manuscript and its connection to the experimental interpretation. My main goal is to ensure that (i) the theoretical models are stated unambiguously (Hamiltonians/equations, conventions, units), and (ii) the claims made from the theory are supported by the level of modeling employed.

(A) Tight-binding NEGF transport

The authors introduce a tight-binding transport model (combined with NEGF/Keldysh) intended to capture resistance changes between different magnetic configurations of the barrier. The setup is described as a multilayer stack (FM / I / SFI1 / SFI2 / NM) with prescribed on-site energies, barrier heights, layer counts, and a uniform nearest-neighbour hopping.

Questions / points requiring clarification

- **Is the TB model explicitly defined (Hamiltonian + geometry)?** The manuscript/SI provides a parameter list, but I could not find a clearly written Hamiltonian and geometry (dimensionality, lattice, how bias enters, how lead self-energies are implemented). Please write the explicit TB Hamiltonian used and define all conventions so the model is reproducible.
- **Single-band nature and scope of claims.** The TB approach is stated as single-band. Please clarify the intended scope: are the TB results meant to be purely qualitative (illustrative trends), or are they used as quantitative support for the experimental magnitudes?
- **Bias polarity and mapping to the experimental convention.** Please state clearly which lead corresponds to which experimental electrode and how the sign of bias/current in the model maps to the experimental bias convention.

(B) Atomistic NEGF–Hubbard

The authors also employ an “atomistic” NEGF–Hubbard approach on a finite 2D lattice treated at Hartree–Fock level, coupled to leads (including a magnetic lead) and to an additional bath intended to model thermalization/relaxation.

Questions / points requiring clarification

- **Single-orbital nature of the Hubbard model.** As written, the Hubbard model appears to include only site and spin indices (no orbital index), i.e. it is effectively single-orbital. Please confirm this explicitly.
- **Consistency with claims about e_g orbital splitting / inversion.** In the discussion around Fig. S1 the text argues in terms of a hierarchy (and even inversion) between $3d_{x^2-y^2}$ and $3d_{3z^2-r^2}$. Please clarify how such orbital-resolved statements are supported by a single-orbital Hamiltonian. If the e_g discussion is an experimental interpretation, please clearly separate it from what is computed in the model and soften any wording that implies the model computes orbital inversion.
- **Parameter justification and mapping to experiment.** The SI provides a particular parameter set (e.g. t , U , intersite terms, temperature) and lead/bath couplings. Please state whether these are constrained by material scales (or fitted to experiment), or whether they are illustrative. If illustrative, please avoid language implying “microscopic evidence” for orbital-level rearrangements.

(C) Micromagnetic (MuMax3) simulations: what the authors did

The SI reports micromagnetic simulations aimed at reproducing the field-dependent magnetoresistance (and a current-threshold-induced transition) by modelling the magnetic layers and the barrier in different magnetic states.

Questions / points requiring clarification

- **Parameters (fit vs literature vs assumed).** Please state clearly whether the micromagnetic parameters (M_s , exchange stiffness A , anisotropy K_u , interfacial coupling) are taken from literature for the relevant materials/temperature, extracted from experimental loops, or tuned to reproduce specific features of the measured curves.
- **Model choices and physical consistency.** The simulations neglect demagnetization fields and use simplified boundary conditions. Please justify these choices and discuss how they impact hysteresis/switching fields.
- **How the FI→AFI transition is implemented.** Please clarify whether the FI→AFI transition is emergent from a coupled model or imposed by switching parameters/state by hand. If imposed, the results should be framed as phenomenological illustration rather than predictive validation.

(D) Spin-transfer torque (STT) analysis

The SI includes an STT-based estimate framed in terms of a generalized LLG equation and introduces a torkance-like quantity to assess whether STT can account for the observed switching thresholds.

Questions / points requiring clarification

- **Equations: please write them explicitly.** I could not find the generalized LLG equation written out with the exact STT term used (damping-like only? any field-like term? sign conventions. Would the term be Slonczewski-like?). Please provide the explicit equation(s) of motion and define every symbol.
- **What exactly is “TB-calculated STT” in Fig. S4?** Fig. S4 refers to TB-calculated STT. Please clarify how STT is extracted from the TB-NEGF framework (e.g. from spin-current divergence, a specific $Q_{\alpha\beta}$ quantity, etc.), and provide the explicit definition used in the figure caption or the text.
- **Parameter justification (e.g. damping α and material inputs).** The STT conclusion depends on assumed parameters. Please justify the chosen values (literature/experiment) and, ideally, provide a sensitivity range showing that the conclusion (STT too small / too large) is robust.
- **Units and normalization.** The figure(s) normalize by “area” without specifying what unit/normalization is used (device area, cross-section in the TB model, unit-cell area, etc.). Please state the area definition and provide standard units (e.g. A/cm² for current density; torque per unit moment, etc.) so the plot is interpretable.
- **Typos / symbol encoding issues.** The SI contains multiple typos and symbol-encoding artifacts (e.g. corrupted characters / question marks). Please correct these throughout.

Final remarks. In its present form, the theoretical part of the SI reads as a set of largely independent modelling exercises rather than a unified framework tied transparently to the experiment. Critical elements are missing for reproducibility and evaluation: explicit model

equations (TB Hamiltonian and generalized LLG with STT), parameter provenance (fit vs literature vs illustrative), consistent unit normalization (including the “area” used in Fig. S4) etc. As a result, it is difficult to determine whether the theory is intended as a qualitative consistency check or a quantitatively predictive description. If the paper’s conclusions depend on quantitative, material-specific support, a major revision of the theoretical sections is required.