

RESEARCH ARTICLE

Nonvolatile Electrical Control of Spin-Polarized Tunneling via Sublattice-Asymmetric Interfacial Magnetoelectric Coupling

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Received: 16 May 2026 | **Revised:** 16 July 2026 | **Accepted:** 31 July 2026

Keywords: condensed matter physics | electric-field control | ferrimagnet | multiferroic tunnel junction | spintronics | tunnel magnetoresistance

ABSTRACT

Achieving nonvolatile, all-electric control of spin-dependent transport properties remains a fundamental challenge in spintronics. Here, we demonstrate nonvolatile electric-field-driven control of spin-polarized tunneling in a multiferroic tunnel junction based on a compensated CoGd ferrimagnet. By switching the ferroelectric polarization, we achieve a clear sign inversion of the tunneling magnetoresistance (TMR), clearly indicating the all-electric control of the spin-polarized tunneling. Crucially, the opposite polarity of the TMR sign switching observed above and below the magnetic compensation temperature unambiguously proves that this behavior is intrinsically tied to ferrimagnetism. This phenomenon is governed by a synergistic mechanism: the electric field disparately modulates the antiferromagnetically coupled sublattices while simultaneously driving an interfacial electronic reconstruction that inverts the spin polarization at the Fermi level. Exploiting this intrinsic sublattice asymmetry in ferrimagnets elegantly circumvents traditional magnetic switching paradigms, establishing a framework for next-generation spintronic architectures.

1 | Introduction

Spintronics relies on the manipulation of spin-dependent transport for next-generation information processing [1–4]. In conventional ferromagnetic (FM) devices, such as magnetic tunnel junctions (MTJs) [5], controlling spin-polarized currents necessitates switching the magnetization of the free layer [6]. This switching is typically driven by external magnetic fields [7] or spin torques [8–10], both of which intrinsically demand high current densities and lead to significant energy dissipation.

While utilizing electric fields for magnetic control offers a pathway to drastically reduce energy consumption [11–16], a fundamental challenge persists: electric fields do not break time-reversal symmetry [17] and thus cannot directly switch magnetization. This makes nonvolatile, electric-field control of spin-polarized transport a critical fundamental challenge in spintronics.

Ferrimagnets (FIMs) offer a unique paradigm to circumvent this limitation [18, 19]. These materials comprise two

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antiferromagnetically coupled sublattices (e.g., rare-earth (RE) and transition-metal (TM) elements) with distinct valence states and local environments [20–22], yielding in a density of states (DOS) at the Fermi level that receives asymmetric contributions from both sublattices [22, 23]. When integrated into a multiferroic tunnel junction [24, 25] (MFTJ, e.g., FM/ferroelectric (FE)/FIM heterostructure), the interfacial symmetry breaking enables a new degree of control. Reversing the FE polarization fundamentally alters the interfacial atomic structure [26–28] and orbital hybridization [29, 30], asymmetrically modulating the sublattice contributions to the interfacial DOS. If the majority carriers near the Fermi level are dominated by a specific sublattice [31, 32], this interfacial magnetoelectric coupling can electrically reverse the relative orientation between the net magnetization and the interfacial spin polarization.

Here, we demonstrate nonvolatile electrical control of spin-dependent tunneling utilizing asymmetric interfacial magnetoelectric coupling in an MFTJ composed of a $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.8}\text{Ru}_{0.2}\text{O}_3$ FM electrode and a compensated $\text{Co}_{82}\text{Gd}_{18}$ FIM electrode, separated by a $\text{Pb}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ FE tunnel barrier. We observe that FE polarization switching robustly reverses the sign of the tunneling magnetoresistance (TMR). Importantly, we find that this TMR reversal polarity is intrinsically tied to the compensation temperature (T_M) of the CoGd layer, where the sign reversal is notably absent near T_M due to the net magnetization simultaneously reversed by the FE switching. First-principles calculations corroborate that FE polarization reversal strongly modifies the interfacial hybridization, flipping the momentum-dependent spin polarization of the interfacial states. Our device uniquely enables in situ electric-field control of spin-polarized tunneling with three temperature-programmable TMR regimes, achieving functionality beyond conventional ferrimagnetic tunnel junctions.

2 | Results

2.1 | The Fundamental Operation of an MFTJ

The fundamental operation of our hybrid MFTJ relies on synergistic interfacial magnetoelectric coupling governed by the intrinsic sublattice asymmetry of the ferrimagnet, that is, the profound structural and chemical differences of the two antiferromagnetically coupled sublattices (e.g., RE and TM). Therefore, the polarization-induced interfacial effects, such as orbital hybridization [33, 34] and charge transfer [35, 36], inevitably modify the two sublattices asymmetrically (Figure 1a,b). The interfacial perturbation may disparately modulate the spin-up and spin-down density of states (DOS), which may invert the sign of the interfacial spin polarization at the Fermi level (E_F) upon polarization reversal and switch the MFTJ's spin-dependent tunneling properties (Figure 1c,d).

Beyond reshaping the interfacial DOS, this asymmetric modulation simultaneously induces imbalanced magnetic moments δM of the two sublattices (Figure 1e,f). While negligible in uncompensated magnets, this δM becomes decisive in a compensated FIM near its T_M , enabling the ferroelectric field to directly reverse the macroscopic net magnetization.

This dynamic interplay makes the compensated FIM-based MFTJ an ideal testbed to verify our dual-control mechanism, predicting three distinct temperature-dependent TMR regimes. Far from T_M , the robust intrinsic magnetization pins the net magnetic orientation, allowing the ferroelectric field to exclusively flip the interfacial spin polarization and induce a definitive TMR sign reversal. Furthermore, because the dominant sublattice inherently transitions from RE to TM across T_M , the polarity of this FE-driven TMR modulation, such as switching from positive to negative versus negative to positive, is inverted between the low- and high-temperature limits. Conversely, near T_M , the ferroelectric switching simultaneously flips both the interfacial spin polarization and the net magnetization. These concurrent reversals dynamically offset each other, leaving the overall spin-dependent tunneling configuration intact and yielding no sign reversal in the TMR.

2.2 | Characterization of an MFTJ

To experimentally demonstrate this proposal, we consider an MFTJ comprising a $\text{La}_{0.7}\text{Sr}_{0.3}\text{Mn}_{0.8}\text{Ru}_{0.2}\text{O}_3$ (LSMRO) (LSMRO) ferromagnetic electrode and a $\text{Pb}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{O}_3$ (PZT) ferroelectric barrier grown on $(\text{La,Sr}) (\text{Al,Ta})\text{O}_3$ (LSAT) substrates by pulsed laser deposition (PLD), alongside a $\text{Co}_{88}\text{Gd}_{12}$ (CoGd) ferrimagnetic electrode deposited via magnetron sputtering (Figure 2a). Detailed growth and fabrication procedures are provided in the Methods.

We first characterized the films of the individual components to confirm their structural and magnetic properties. Anomalous Hall effect (AHE) measurements on the LSMRO (12 nm) and CoGd (4 nm) layers confirm the robust perpendicular magnetic anisotropy (PMA) in both (Figure 2b,c). Specifically, for the LSMRO layer (Figure 2b), the strong PMA at low temperatures originates from a certain amount of 4d or 5d transition-metal dopants [37]. The coercive field (H_C) monotonously decreased from 2650 to 1300 Oe as temperature increased from 15 to 70 K. In addition, the magnetic hysteresis loops of the LSAT/LSMRO at 50 K also confirm the PMA of LSMRO (Figure S1).

For the ferrimagnetic CoGd layer (Figure 2c), the AHE hysteresis loops exhibit clockwise polarity below 30 K, transitioning sharply to anticlockwise polarity above 30 K. Additionally, the H_C is enhanced at the temperature close to 30 K and reduced at the temperature away from it. Correspondingly, the magnitude of the anomalous Hall resistance change ($|\Delta R_{\text{AHE}}|$) decreases from 0.31 Ω at 15 K to nearly 0 Ω at 30 K, and recovers to 0.30 Ω at 70 K. These features unambiguously identify the T_M of the CoGd layer to be ~ 30 K, with the Gd (Co) sublattice dominating the net magnetization below (above) this temperature.

The ferroelectricity of the PZT layer is verified using out-of-plane piezoresponse force microscopy (PFM). As shown in Figure 2d, applying a writing voltage of ± 5 V generates distinct domain patterns, where the dark (bright) contrast corresponds to downward (upward) ferroelectric polarization in the central (peripheral) regions. The atomic force microscopy image, the x-ray diffraction pattern, and reciprocal space mapping of the bilayers confirm the atomically flat surface of PZT and z-orientation epitaxial structures (Figure S2).

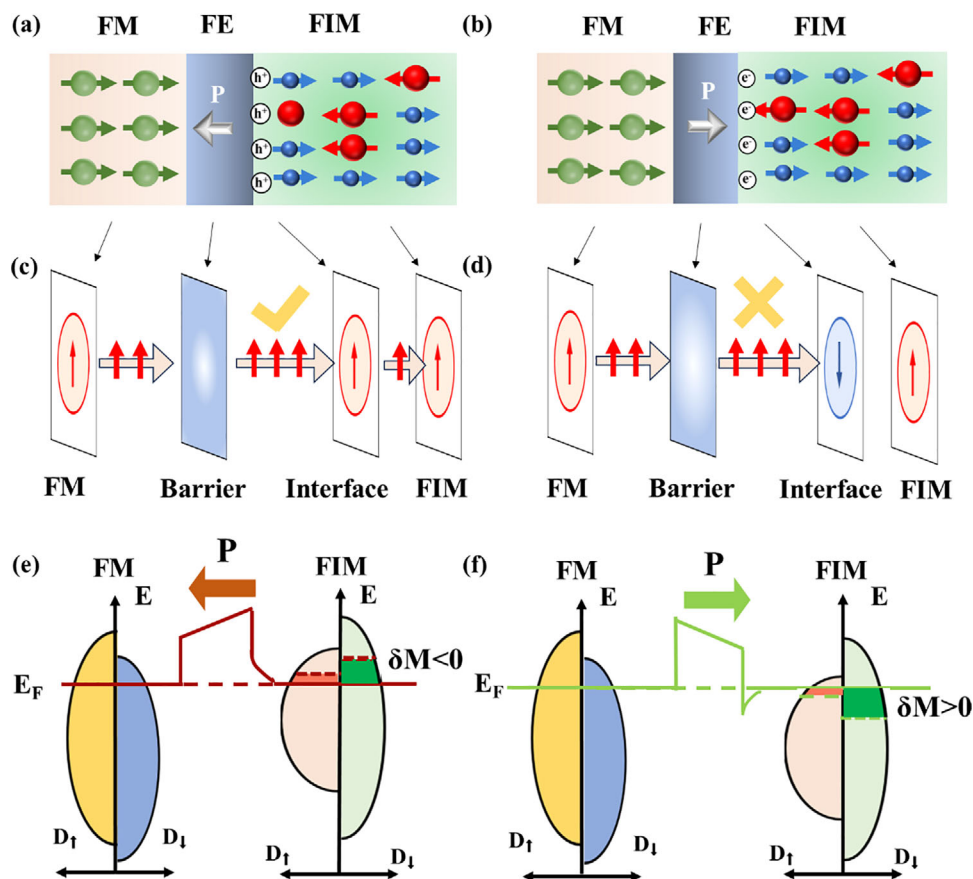


FIGURE 1 | (a,b) A FM/FE/FIM MFTJ. Reversing the ferroelectric polarization (P) induces opposite bound charge accumulations at the FE/FIM interface. (c,d) The corresponding spin-polarized tunneling processes. The polarization-driven interfacial electronic reconstruction inverts the sign of the interfacial spin polarization. This toggles the interfacial spin configuration from a low-resistance state (c) to a high-resistance state (d) relative to the FM electrode, triggering a distinct sign reversal in the TMR. (e,f) Schematic illustrating the asymmetric modulation of the density of states (DOS) of the antiferromagnetically coupled sublattices. The ferroelectric field shifts the Fermi level (E_F), asymmetrically influencing the contributions of the transition-metal (TM) and rare-earth (RE) sublattices. This disparate tuning modulates the intrinsic moment difference δM , dynamically shifting the macroscopic magnetic compensation temperature (T_M) of the ferrimagnet and enabling the direct electrical reversal of the dominant magnetic sublattice.

We then fabricate an LSMRO (12 nm)/PZT (5 nm)/CoGd (4 nm) MFTJ. The high-quality interfaces of the heterostructure were corroborated by representative cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) and annular bright-field (ABF) imaging (Figure 2e), alongside the corresponding energy-dispersive x-ray spectroscopy (EDS) elemental mapping (Figure S3).

To confirm full ferroelectric polarization switching in the MFTJ via applied electric fields, we measured I - V curves at 35 K (Figure 2f). After down polarization by -5 V, I increases slowly with increasing V , indicating a large tunneling resistance. Conversely, after up polarization by $+5$ V, I increases dramatically with increasing V , and the tunneling resistance decreases by at least one order of magnitude compared with that of the down-polarized state. These resistance changes are characteristic of a typical tunneling electroresistance (TER) effect [38, 39]. The bipolar resistive switching (Figure S4) and retention behaviors (Figure S5) further demonstrate that the TER in our devices originates predominantly from ferroelectric polarization switching, rather than from oxygen vacancy migration. Consequently, we applied pulsed gate voltages of $+5$ or -5 V (100 μ s) to deterministically

polarize the PZT barrier upward or downward for all subsequent measurements.

2.3 | TMR of an MFTJ Under Opposite Ferroelectric Polarizations

We next investigated the TMR signals under out-of-plane magnetic field sweeps for opposite FE polarization states. The TMR ratio is defined as $TMR = \frac{R_{AP} - R_P}{R_P} \times 100\%$, where R_P and R_{AP} denote the resistances for the parallel (P) and antiparallel (AP) magnetization alignments of the two magnetic electrodes, respectively. It is known that $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is a half-metal with positive spin polarization at the E_F [40], whereas Co exhibits negative spin polarization at E_F [41]. Given their low doping concentrations, we expect that our LSMRO and CoGd electrodes largely inherit these intrinsic spin polarizations. Therefore, at temperatures above T_M where the net magnetization of CoGd is dominated by the Co sublattice, the P state should intrinsically exhibit a high-resistance state, yielding a negative TMR. Indeed, a negative TMR of -18.3% is observed at 35 K under the upward PZT polarization (Figure 3a). Notably,

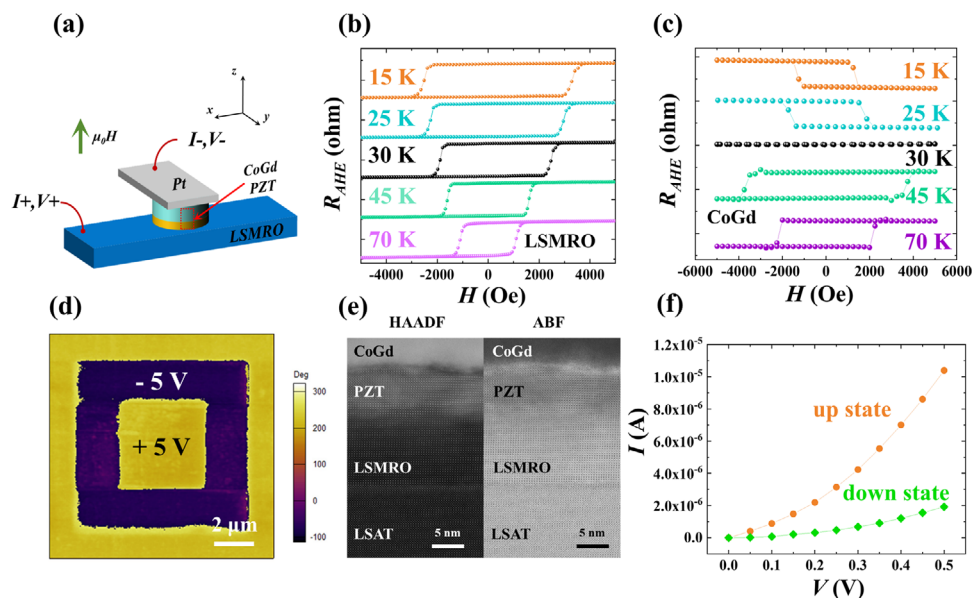


FIGURE 2 | (a) Schematic illustration of the LSMRO (12 nm)/PZT (5 nm)/CoGd (4 nm)/Pt (100 nm) MFTJ. (b,c) Anomalous Hall resistances (R_{AHE}) of the independent films of LSMRO (b) and CoGd (c) as functions of the out-of-plane magnetic field (H) at various temperatures (15 to 70 K). (d) Out-of-plane PFM image of the LSMRO/PZT bilayer. The dark and bright regions correspond to the downward and upward ferroelectric domains, respectively, written by ± 5 V gate voltages. (e) Cross-sectional HAADF-STEM image and the corresponding ABF image of the LSMRO/PZT/CoGd MFTJ. (f) I-V characteristic curve of the MFTJ measured at 35 K.

reversing the ferroelectric polarization to downward, the P state transitions to a low-resistance state, yielding a positive TMR of +5.2% (Figure 3b). This clear sign inversion unambiguously indicates the effective FE-controlled spin-polarized tunneling.

Crucially, FE polarization switching also strongly modulates the H_C of the TMR peaks. Owing to the considerable thickness of the LSMRO layer (12 nm), the ferroelectric field only slightly modifies its H_C . In stark contrast, the ultrathin CoGd layer (4 nm) allows the FE polarization to effectively modulate its perpendicular magnetic anisotropy and significantly shift its T_M point [17, 29, 42]. Consequently, switching the polarization of PZT drastically modulates the H_C of CoGd. Under upward polarization, the CoGd H_C becomes exceptionally large due to being close to the T_M of the upward polarization state. Conversely, because the T_M point is substantially lowered under downward polarization [29], the H_C of CoGd decreases drastically. As a direct result, during magnetic field sweeps from negative to positive, the magnetization reversal initiates in the LSMRO layer and subsequently in the CoGd layer under upward polarization (Figure 3a), whereas this reversal sequence is completely inverted under downward polarization (Figure 3b). Taken together, these observations definitively demonstrate that the all-electric control of spin-polarized tunneling arises from the asymmetric interfacial magnetoelectric effect associated with the CoGd ferrimagnet. In addition, first-principles calculations and electric-field control experiments were performed to rule out the possibility that the observed modulations originate from changes in the interfacial electronic or magnetic states of the LSMRO layer (see details in Supporting Information S6). The retention tests demonstrated that our MFTJ signals are nonvolatile (Figure S7). And the pulse-width dependence experiment further reflected that the

resistive switching is governed by ferroelectric polarization reversal (Figure S8).

To further analyze the TMR behavior in our device, we systematically measured the TMR signals under opposite ferroelectric polarizations across a broad temperature range (Figure 3c–f). For both polarizations, the resistance at 0 Oe shows a similar temperature dependence: it rises slightly at low temperatures and then gradually decreases with increasing temperature (Figure S9).

For upward polarization (Figure 3c,e), the TMR is negative above 30 K and positive below 30 K, indicating that the T_M of the CoGd layer is only marginally influenced by the upward ferroelectric field. In contrast, under downward polarization (Figure 3d,f), the TMR is positive above 20 K and negative below 20 K, demonstrating a substantial T_M reduction consistent with previous reports [29] (see Supporting Information S10 for details). This temperature-driven TMR sign reversal originates from the dominant sublattice crossover in CoGd, which reverses the interfacial spin polarization via the opposite spin characters of the two antiferromagnetically coupled sublattices. Comparing the TMR signs between the two polarization states shows a robust FE polarization-driven TMR sign reversal in both the high- and low-temperature limits, but this all-electric reversal is absent in the 20–30 K intermediate-temperature window. The emergence of these three distinct temperature-dependent TMR regimes aligns perfectly with our proposed physical picture, unambiguously confirming the phenomenon fundamentally originates from the asymmetric interfacial magnetoelectric effect. Additionally, the modulated magnitude of our ferrimagnet-based perpendicular hybrid heterostructure is comparable to that of the traditional in-plane MFTJ devices at temperatures below 40 K and surpasses them at temperatures above 40 K (Figure S11) [38, 43].

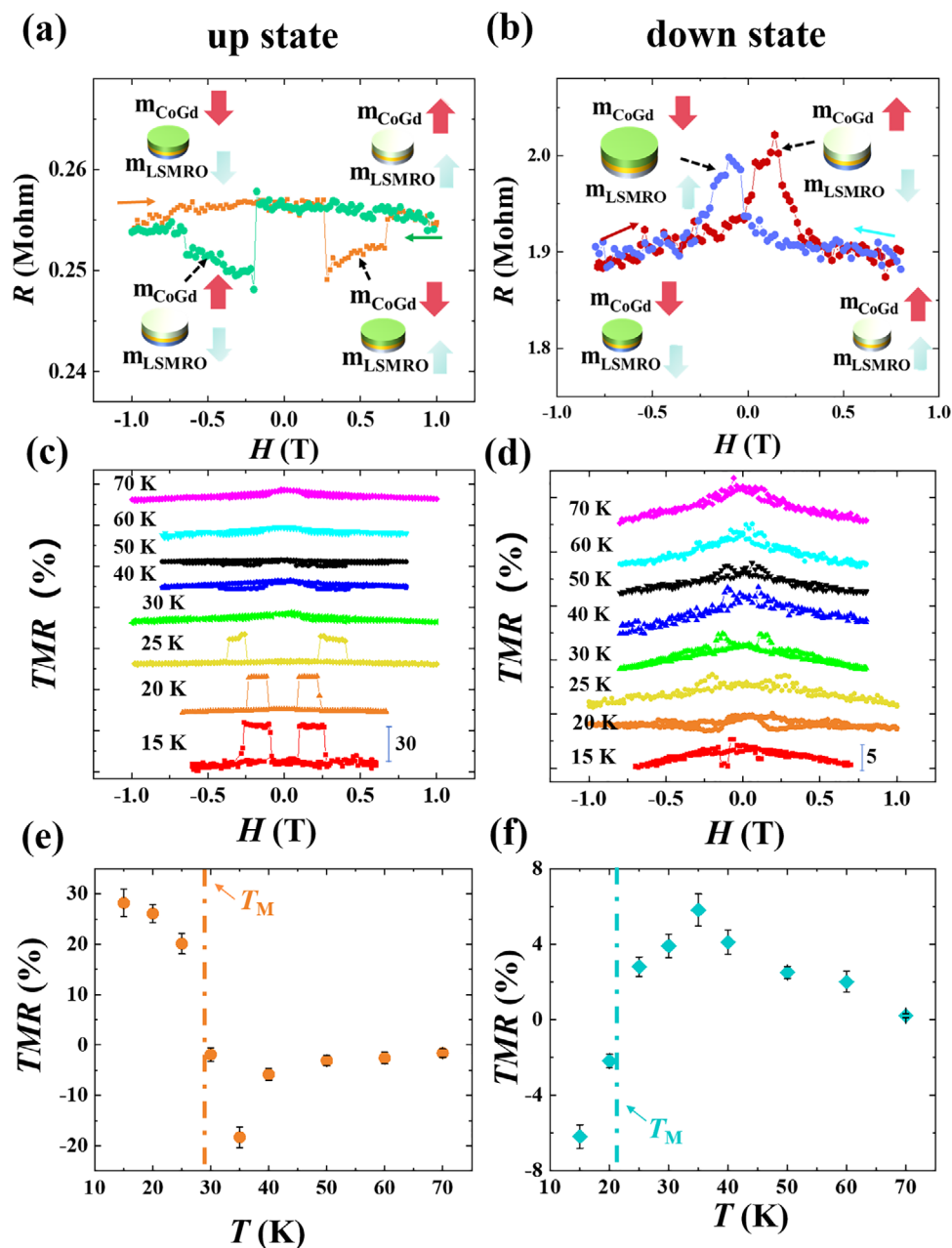


FIGURE 3 | (a,b) TMR curves as a function of the out-of-plane magnetic field (H) measured at 35 K under (a) upward and (b) downward ferroelectric polarization states. The insets schematically illustrate the magnetic configurations of the LSMRO and CoGd layers during the magnetization reversal process. (c,d) TMR curves measured at various temperatures (from 15 to 70 K) under (c) upward and (d) downward polarizations. (e,f) Temperature dependence of the TMR ratio under (e) upward and (f) downward polarizations. The vertical dashed lines denote the corresponding magnetic compensation temperatures (T_M) of the CoGd layer under the respective polarization states. Error bars are defined as standard deviations from three independent devices on the same device array under different magnetic fields.

2.4 | Calculations of FE-Driven Interfacial Spin Polarization

To elucidate the underlying microscopic mechanism of FE-driven interfacial spin polarization, we performed first principles calculations on a heterostructure comprising a 4-unit-cell-thick PbTiO_3 layer (serving as a simplified model for PZT) and a Co_5Gd layer (Figure 4a,b). The composition of the Co_5Gd layer ($\sim\text{Co}_{83.3}\text{Gd}_{16.7}$) was specifically chosen to closely approximate the experimental $\text{Co}_{82}\text{Gd}_{18}$ stoichiometry. Our results reveal that robust interfacial hybridization metallizes the PbTiO_3 layer adja-

cent to the interface, whereas the layers further away maintain a density of states similar to that for bulk PbTiO_3 (Figure S12). Crucially, by comparing the relaxed atomic structures for the two opposite polarization states, we find that the ferroelectric switching induces substantially larger displacements of the Gd atoms relative to the Co atoms. This disparate structural response clearly implies the asymmetric magnetoelectric modulation of the two sublattices.

We calculated the layer-dependent, transverse-wave-vector-resolved density of states ($\mathbf{k}_\parallel$ -resolved DOS, $N_\parallel^\uparrow$ and $N_\parallel^\downarrow$) at E_F and

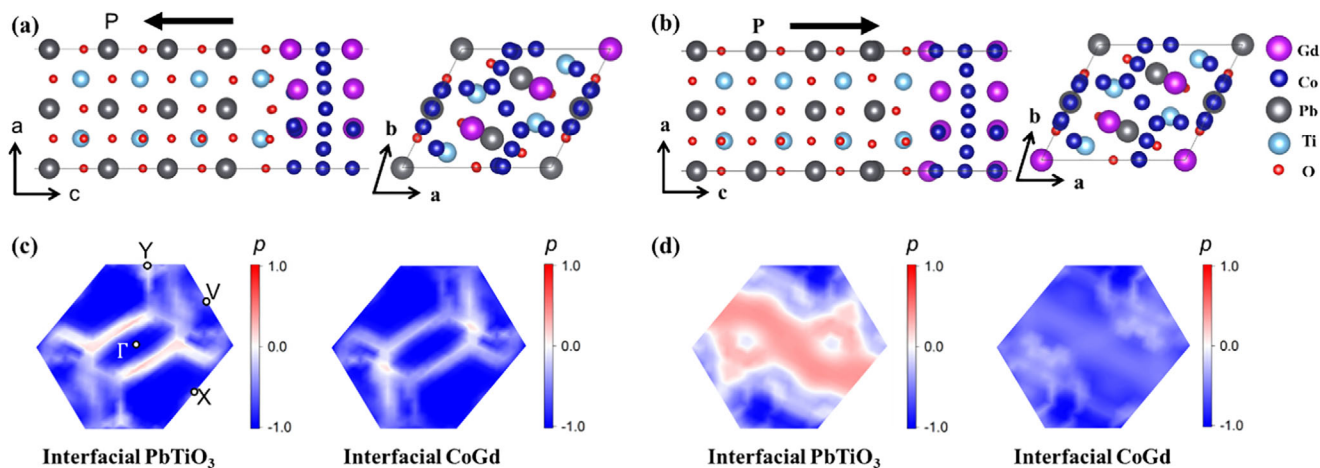


FIGURE 4 | (a,b) Relaxed atomic structures of the PbTiO₃/CoGd heterostructure with the ferroelectric polarization (P, black arrows) pointing (a) away from and (b) toward the CoGd interface. (c,d) Calculated transverse-wave-vector-resolved spin polarization ($p_{\parallel}$) at E_F within the two-dimensional Brillouin zone (2DBZ) for the interfacial PbTiO₃ and CoGd layers. Panels (c) and (d) correspond to the polarization configurations in (a) and (b), respectively. The color scale indicates the magnitude and sign of $p_{\parallel}$. Note the drastic sign inversion of $p_{\parallel}$ for the interfacial PbTiO₃ layer near the zone center (Γ point) upon polarization reversal, which dictates the spin matching/mismatching across the junction and governs the switching of the tunneling magnetoresistance.

derived the momentum-resolved spin polarization: $p_{\parallel} = \frac{N_{\parallel}^{\uparrow} - N_{\parallel}^{\downarrow}}{N_{\parallel}^{\uparrow} + N_{\parallel}^{\downarrow}}$. The ferroelectric switching of PbTiO₃ profoundly dictates the $p_{\parallel}$ distribution within these interfacial layers. For downward polarization (polarization points away from the interface, Figure 4c), the $p_{\parallel}$ distributions for both the interfacial CoGd and PbTiO₃ layers are predominantly negative. This negative polarization is particularly pronounced at the two-dimensional Brillouin zone center (Γ point), which inherently possesses the smallest evanescent decay rate and therefore overwhelmingly dominates the tunneling transmission.

Conversely, upward polarization (polarization points toward the interface, Figure 4d) induces severe local distortions via interfacial Ti atom displacement, altering the $p_{\parallel}$ landscape. While the interfacial CoGd layer retains an overall negative $p_{\parallel}$, the electronic reconstruction drives the interfacial PbTiO₃ layer to exhibit broad regions of positive $p_{\parallel}$, particularly at Γ point. Consequently, when the spin-polarized current from the LSMRO electrode tunnels across the barrier, the transmission is sustained (suppressed) when the $p_{\parallel}$ of the interface layer matches (mismatches) the spin polarization of the tunneling current as described by the Jullière's picture [44], effectively toggling the junction between low and high resistance states. We note that because the Co₈₂Gd₁₈ layer in our experiments is a polycrystalline alloy, our first-principles calculations based on an idealized periodic Co₅Gd crystal lattice primarily offer qualitative insights into the interfacial magnetoelectric coupling. Nevertheless, this momentum-resolved electronic reconstruction is in good agreement with our phenomenological model (Figure 1), confirming the proposed asymmetric interfacial magnetoelectric mechanism is broadly applicable in FIM alloy systems.

Beyond compensated ferrimagnets, the physical insights gained from exploiting intrinsic sublattice asymmetry offer a transformative approach for the field of antiferromagnetic spintronics. In collinear antiferromagnets, driving non-identical responses

from sublattices can inherently break macroscopic symmetries that typically restrict spin manipulation. A prominent example of this emerging paradigm is the recently proposed asymmetric spin torque, where sublattice-asymmetric spin accumulations enable the deterministic switching of the Néel vector without requiring exotic crystal symmetries [45]. By designing asymmetric magnetoelectric coupling in such antiferromagnetic systems, future research can harness similar frameworks for the purely electrical control of spin-dependent transport.

3 | Conclusion

Overall, we have realized the nonvolatile, electric-field-driven control of spin-dependent tunneling, evidenced by TMR sign reversal in a hybrid ferrimagnet-based multiferroic tunnel junction. This striking capability originates from asymmetric interfacial magnetoelectric modulation of the compensated ferrimagnet's two antiferromagnetically coupled sublattices. This approach elegantly circumvents the traditional time-reversal symmetry constraints that inherently hinder all-electric magnetic manipulation. Ultimately, exploiting or designing the sublattice asymmetry in magnets with two or more sublattices establishes a transformative physical paradigm, paving the way for the development of next-generation, ultra-low-power spintronic architectures and multifunctional multistate logic devices.

4 | Methods

4.1 | Sample Growth and Device Fabrication

The bilayers LSMRO (12 nm)/PZT (5 nm) were epitaxially deposited on (100)-oriented LSAT substrates using a KrF excimer laser ($\lambda = 248$ nm) in the pulsed laser deposition (PLD) system. The growth conditions for the LSMRO and PZT layers were maintained at 700°C and 600°C substrate temperature, 1.0 J/cm²

laser energy density, and 3 Hz pulse repetition rate. The oxygen pressure during the deposition process was set as 200 mTorr for LSMRO and 100 mTorr for PZT, respectively. After the deposition, the multilayers were in situ annealed for 15 min and then cooled down to room temperature under the oxygen pressure of 2000 mTorr to reduce oxygen vacancies.

After deposition of the heterostructures, the MTJ device array crisscrossed with strip-shaped bottom electrode LSMRO and top electrode $\text{Co}_{82}\text{Gd}_{18}$ (4 nm)/Pt (100 nm) were further fabricated, and a 60 nm silicon oxide layer was deposited to insulate them. The junction devices (5 μm in diameter) were patterned using ultraviolet lithography and inductively coupled plasma (ICP) etching. The $\text{Co}_{82}\text{Gd}_{18}$ (4 nm) layer was deposited using a double-target co-sputtering process in a magnetron sputtering system with a base pressure less than 5×10^{-8} Torr. A 100 nm Pt capping layer was subsequently deposited in situ on the CoGd layer to ensure electrical contact. Both layers were deposited under an Ar pressure of 0.8 mTorr at room temperature.

4.2 | Magnetoresistance Characterizations

AHE measurements and the magnetoresistance characterizations for our devices at low temperatures were performed using the Lakeshore Probe Station. The atmosphere is dry, and the pressure is below 1.0×10^{-7} Torr. A Keithley 2403 was used to provide current source, and a Keithley 2182 was used to detect the Hall voltage. In Hall measurements, the applied current was 0.1 mA unless otherwise noted.

4.3 | Polarized Methods

A Keithley B1500 provided a pulsed voltage to polarize the PZT layer. The LSMRO and CoGd layers were used as top and bottom electrodes. Two probes were used to connect the electrodes with the Keithley B1500. The application of a positive (negative) gate voltage $V_g \sim 5$ V (100 μs) was used to get non-volatile upward (downward) polarization in the MTJ structure. After the polarization, Keithley B1500 and electrodes were disconnected, and further measurements went on. All polarized processes of our samples were applied in the Lake Shore probe station.

4.4 | Calculation Methods

The density functional theory calculations (DFT) were performed using the projector augmented plane-wave (PAW) basis set, which was implemented within the Vienna ab initio simulation package (VASP). The plane-waves were truncated at 500 eV, while the exchange correlations of electrons were described using generalized gradient approximations (GGA) based on the functional proposed by Perdew, Burke, and Ernzerhof (PBE). To effectively model the interfacial coupling, we constructed $\text{PbTiO}_3/\text{CoGd}$ heterostructures comprising a $\sqrt{5} \times 2$ in-plane supercell of PbTiO_3 matched to a $\sqrt{3} \times \sqrt{3}$ supercell of CoGd. The PbTiO_3 slab thickness was set to 4 unit cells (u.c.), while the CoGd slab was 1.5 u.c. thick. To accurately reflect the experimental alloy composition ($\text{Co}_{82}\text{Gd}_{18}$), we adopted a stoichiometric Co_5Gd configuration (i.e., $\sim\text{Co}_{83.3}\text{Gd}_{16.7}$) in our calculations. A

vacuum region thicker than 20 Å was inserted along the out-of-plane direction to eliminate spurious periodic interactions. The atomic positions of the heterostructures under both up and down ferroelectric polarizations were fully relaxed until the Hellmann-Feynman forces on each atom were less than 0.02 eV/Å. The energy convergence criterion for solving the self-consistent Kohn-Sham equations was set to 1×10^{-4} eV. For structural relaxation, the Brillouin zone was sampled using a Γ -centered k -point mesh with a resolution of $0.06 \text{ \AA}^{-1}$. The layer-resolved density of states (LDOS) and the $k_{\parallel}$ -resolved density of states (KDOS) were subsequently calculated using a denser $21 \times 21 \times 1$ Monkhorst-Pack k -grid.

Author Contributions

K.W. conceived the work. P.L., Q.L. and Z.B. fabricated the devices, carried out the electrical transport measurements, and measured the magnetic properties. Q.L. grew the oxide films. D.S. and X.G. conducted first-principles calculations. K.W., D.S., L.C. and P.L. analyzed the data, wrote the manuscripts. All the authors discussed the results and commented on the manuscript.

Acknowledgements

This work was supported by the National Key Research and Development Program of China (Grant Nos. 2022YFA1405100 and 2024YFB3614100), the National Natural Science Foundation of China (Grant Nos. 12427805, 12241405, 12304154, 52072400, 52250402 and 12274411), Beijing Natural Science Foundation (Grant No. 2212048), the Basic Research Program of the Chinese Academy of Sciences Based on Major Scientific Infrastructures (Grant No. JZHKYPT-2021-08), and the CAS Project for Young Scientists in Basic Research (Grant No. YSBR-084). The theoretical calculations were performed at Hefei Advanced Computing Center.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section.

Supporting File: adma74583-sup-0001-SuppMat.pdf.