

**Symmetry breaking induces room-temperature interface ferroelectricity in two-dimensional metastable mosaic-like heterostructures**

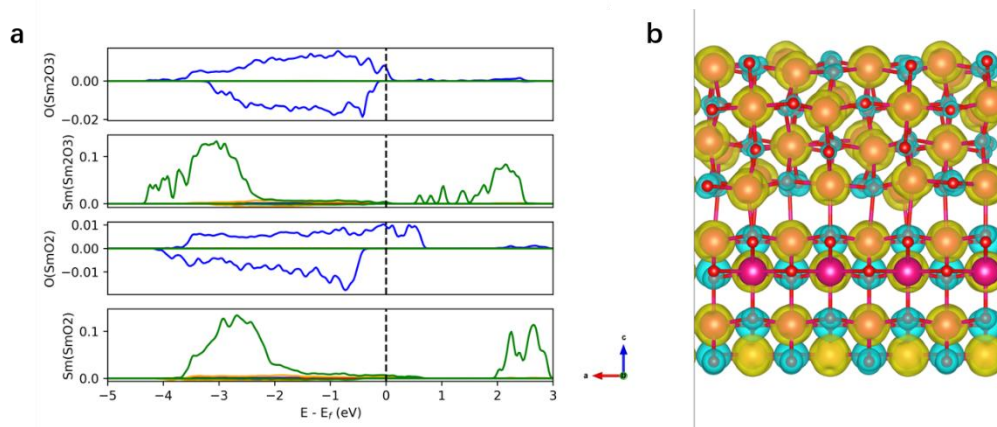
## **Supplementary Information includes:**

Supplementary Figures 1 to 26

Supplementary Tables 2

Supplementary Notes 2

Supplementary References

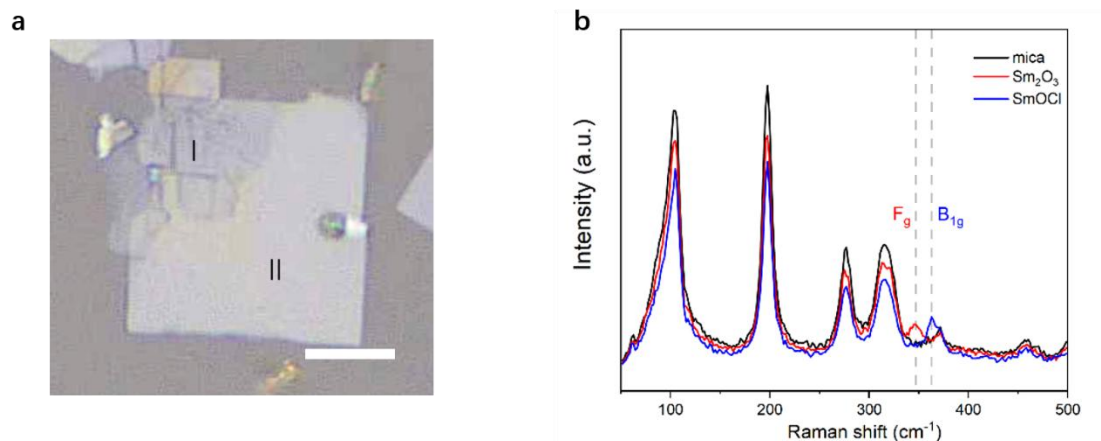


**Supplementary Fig. 1 | Theory calculations of model.** **a**, Calculated DOS of  $\text{Sm}_2\text{O}_3$  and  $\text{SmO}_2$  crystals. **b**, Calculated Spin-polarized charge distribution and atomic magnetic moment of heterostructure model.

For the Sm ions, the density of states below the Fermi level is dominated by the spin-up  $f$  orbitals, which indicates that the Sm ions are all in a spin-polarized state and their spin magnetic moments are primarily contributed by  $f$  electrons. In contrast, for the O ions, the density of states below the Fermi level is dominated by the O  $p$  orbitals. Furthermore, this asymmetry between the spin-up and spin-down  $p$  orbitals suggests that the magnetic moments of the O ions are mainly driven by spin polarization of the  $p$  orbitals.

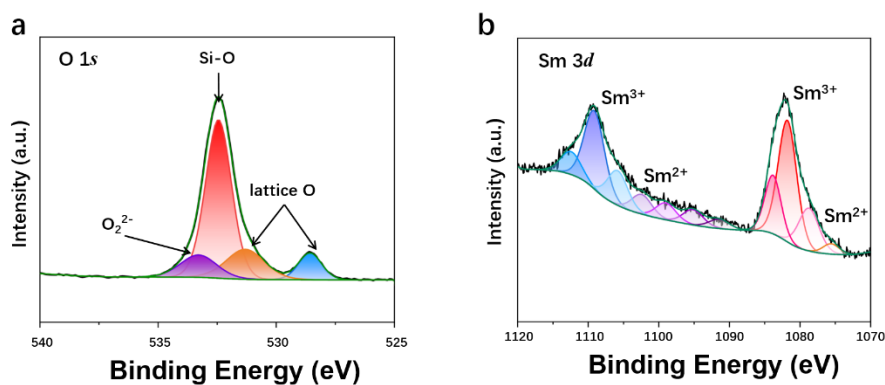
The yellow surfaces correspond to spin-up polarization, while the blue surfaces correspond to spin-down polarization. Thus, electrons around the Sm ions exhibit predominantly spin-up polarization, whereas electrons around the oxygen ions exhibit predominantly spin-down polarization.

Quantitative analysis of these polarizations reveals distinct magnetic moments for different samarium oxides. For monoclinic  $\text{SmO}_2$ , the Sm ions exhibit an average magnetic moment of  $5.0 \mu\text{B}$ , while the O ions contribute  $-0.44 \mu\text{B}$ . In comparison, cubic  $\text{Sm}_2\text{O}_3$  shows a slightly higher average magnetic moment for Sm ions at  $5.1 \mu\text{B}$ , and a reduced contribution from O ions at  $-0.11 \mu\text{B}$ . These quantitative magnetic moment values consistently reflect the described spin polarization trends, albeit with slight variations influenced by the specific crystalline structure.

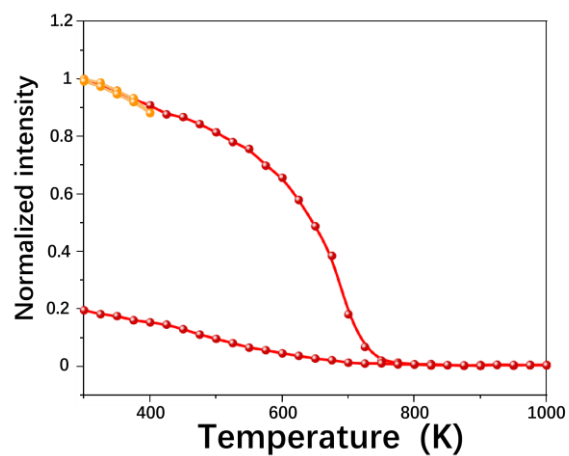


**Supplementary Fig. 2 | Further oxidation phase transition.** **a**, OM image of SmOCl and Sm<sub>2</sub>O<sub>3</sub>. Scale bars, 5  $\mu$ m. **b**, Raman of SmOCl and Sm<sub>2</sub>O<sub>3</sub>.

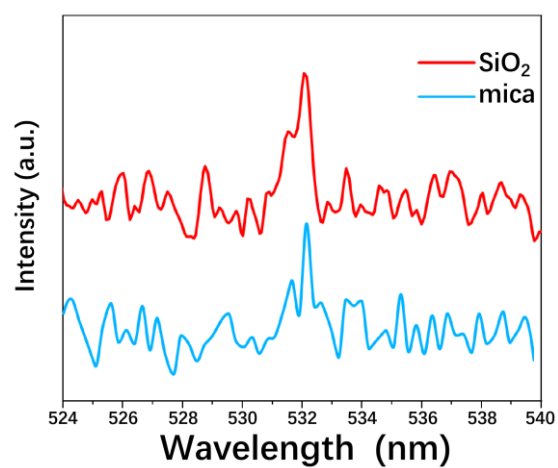
Raman spectroscopic analysis of the optical image of the nanosheet reveals two distinct regions. Region I is assigned to Sm<sub>2</sub>O<sub>3</sub>, with a characteristic Raman peak at 343 cm<sup>-1</sup> (F<sub>g</sub> mode). Region II is assigned to SmOCl, with a characteristic Raman peak at 361 cm<sup>-1</sup> (B<sub>1g</sub> mode). The coexistence of Sm<sub>2</sub>O<sub>3</sub> and SmOCl suggests that SmOCl undergoes further oxidation to Sm<sub>2</sub>O<sub>3</sub> during the growth process.



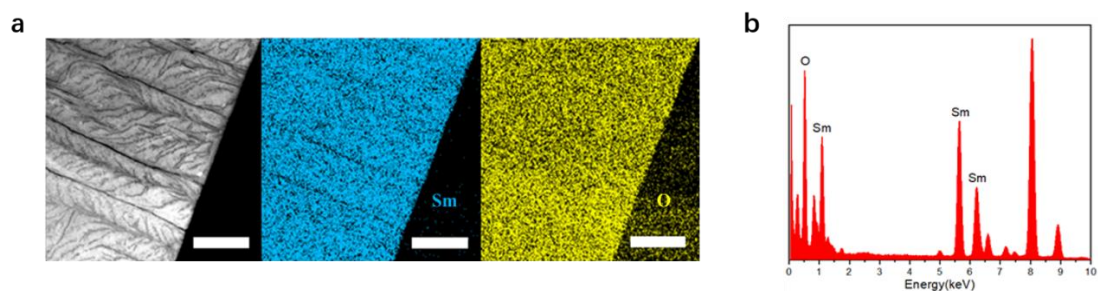
**Supplementary Fig. 3 | XPS characterization.** **a**, XPS spectra of O 1s. **b**, XPS spectra of Sm 3d.



**Supplementary Fig. 4 | Temperature-dependent SHG intensity during heating-cooling cycles.**

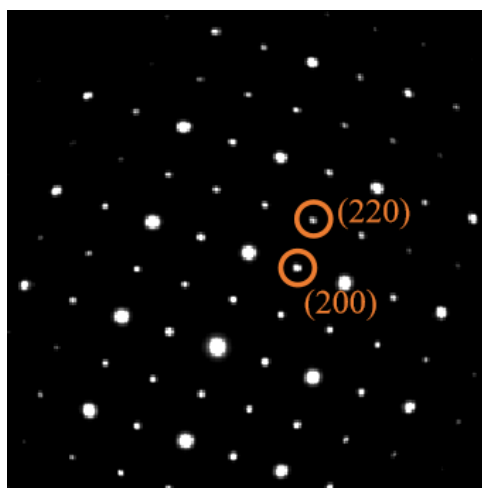


**Supplementary Fig. 5 | SHG signal of the nanosheets on the different substrates.**



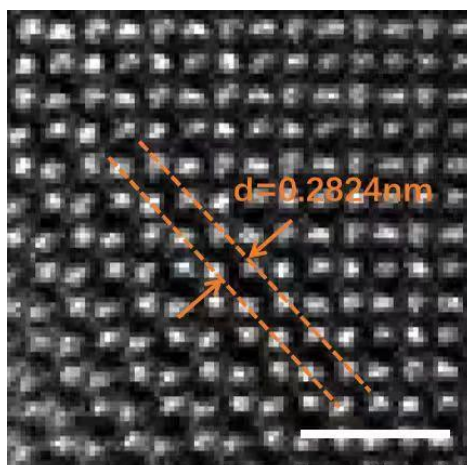
**Supplementary Fig. 6 | Elemental analysis of samarium oxide nanosheets. a,** EDS mapping. Scale bars, 2  $\mu\text{m}$ . **b,** EDS spectrum.

Elemental analysis demonstrates a uniform distribution of Sm and O throughout the nanosheets. Notably, the TEM-EDS spectrum of the nanosheets confirmed the absence of Fe elements in the final product.

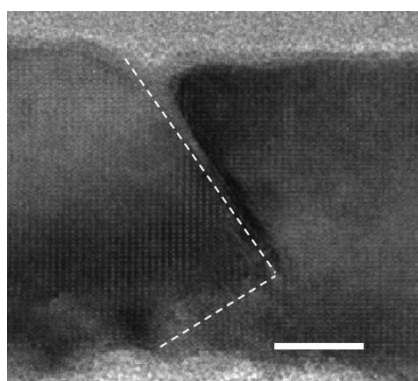


**Supplementary Fig. 7 | Selected area electron diffraction (SAED) image of  $\text{Sm}_2\text{O}_3$ .**

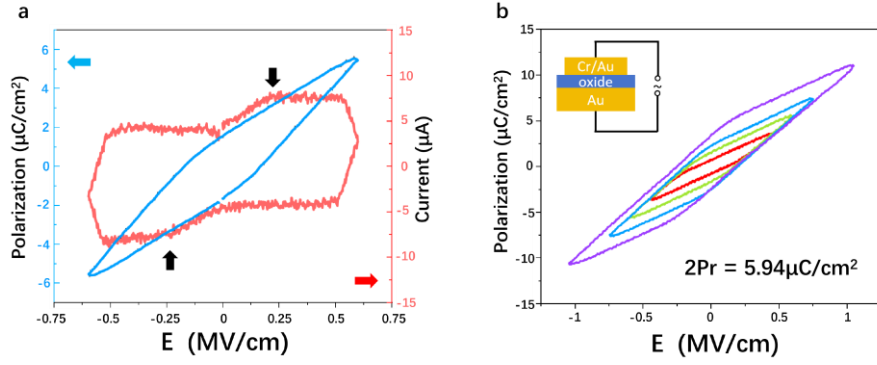
Thin nanosheets along the  $[001]$  axis shows a single set of rectangular diffraction spots indicative of a high-quality single-crystalline structure. The circled bright spots can be assigned to (200) and (220), confirming its 4-fold symmetric structure.



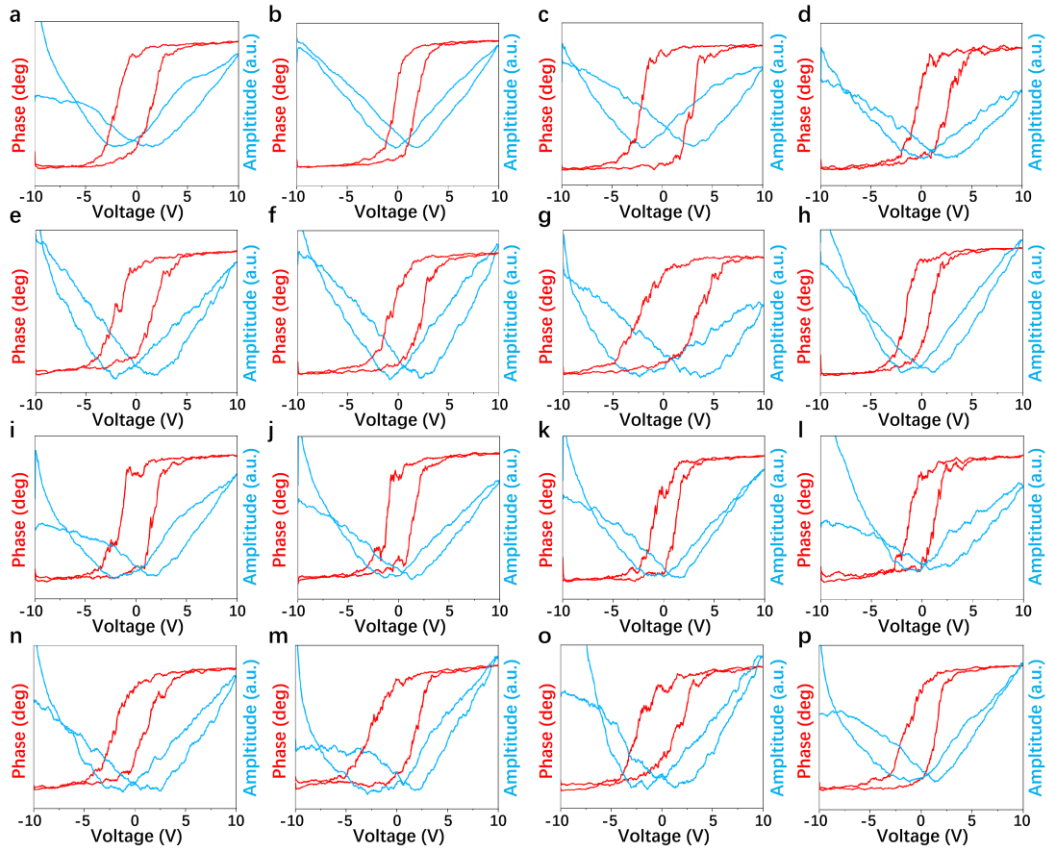
**Supplementary Fig. 8 | High-magnification HAADF-STEM image of the interface of  $\text{SmO}_2/\text{Sm}_2\text{O}_3$  heterostructure. Scale bars, 1 nm. The interlayer distance of interface was found to be 0.2824 nm.**



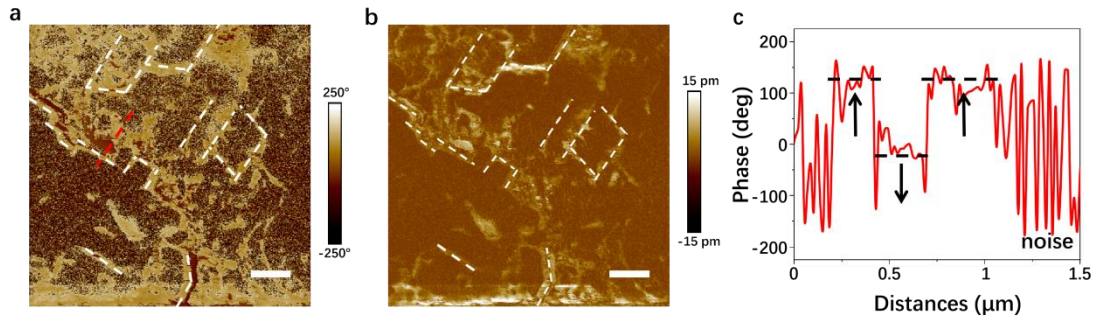
**Supplementary Fig. 9 | Cross-section TEM image of  $\text{SmO}_2/\text{Sm}_2\text{O}_3$  heterostructure. Scale bar, 20nm**



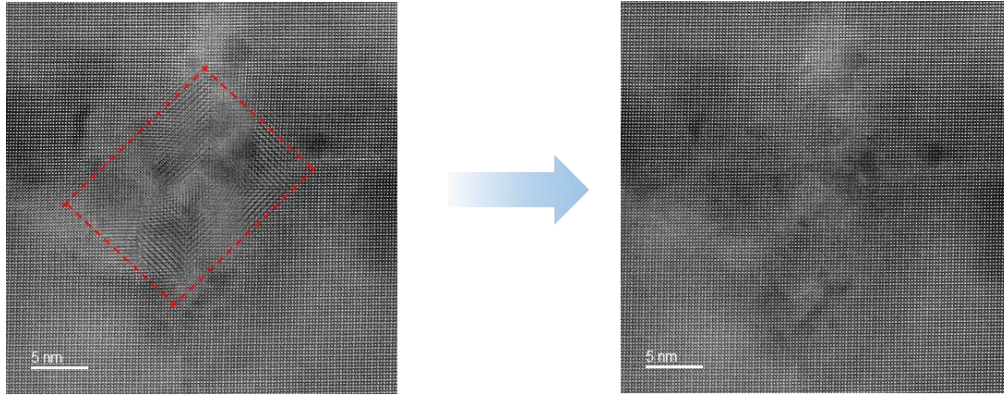
**Supplementary Fig. 10 | Macroscopic ferroelectric measurements** **a**, Polarization and current versus electric field for the  $\text{SmO}_2/\text{Sm}_2\text{O}_3$ . **b**, Polarization measured under increasing maximum electric field.



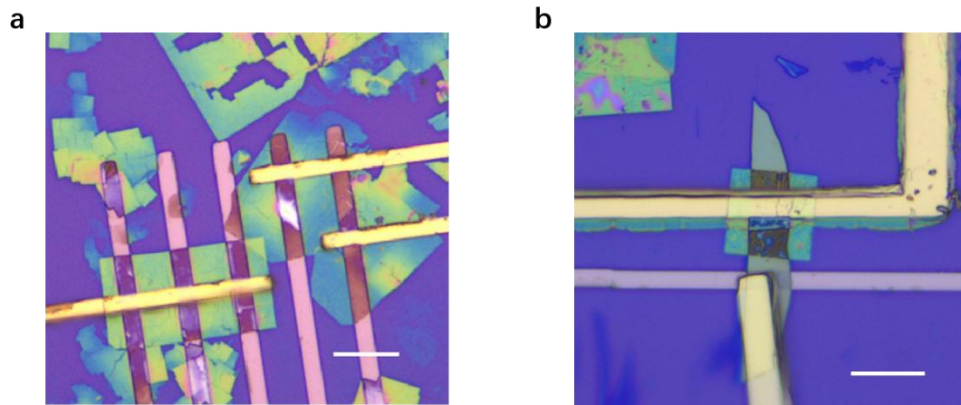
**Supplementary Fig. 11 | PFM characteristic of multiple flakes.**



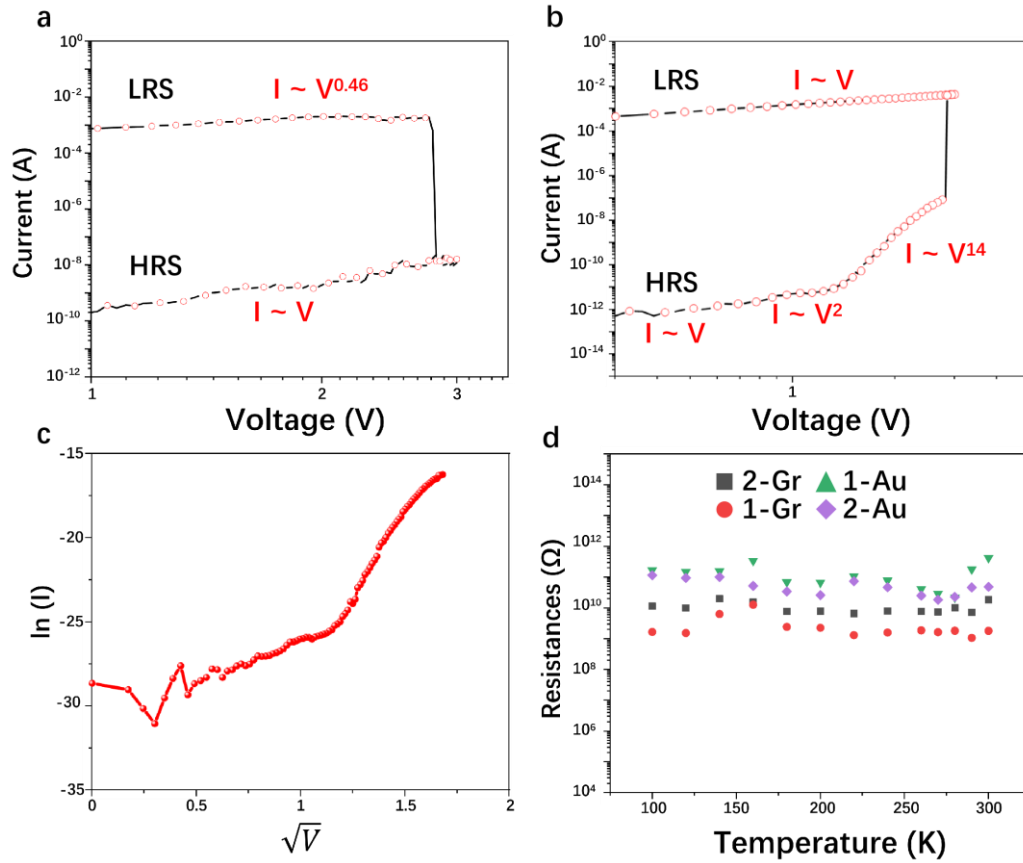
**Supplementary Fig. 12 | Phase characterization** **a**, PFM phase image. Scale bar,  $1 \mu\text{m}$ . **b**, PFM amplitude image. Scale bar,  $1 \mu\text{m}$ . **c** The PFM phase profile along the red dashed line shown in **a**.



**Supplementary Fig. 13 | TEM image of atomic structure.** Under electron-beam irradiation during TEM, the metastable  $\text{SmO}_2$  phase gradually transforms into the more stable  $\text{Sm}_2\text{O}_3$ . The  $\text{SmO}_2$  is preferentially oriented and embedded within the  $\text{Sm}_2\text{O}_3$  matrix, thereby constituting a mosaic-like lateral heterostructure.



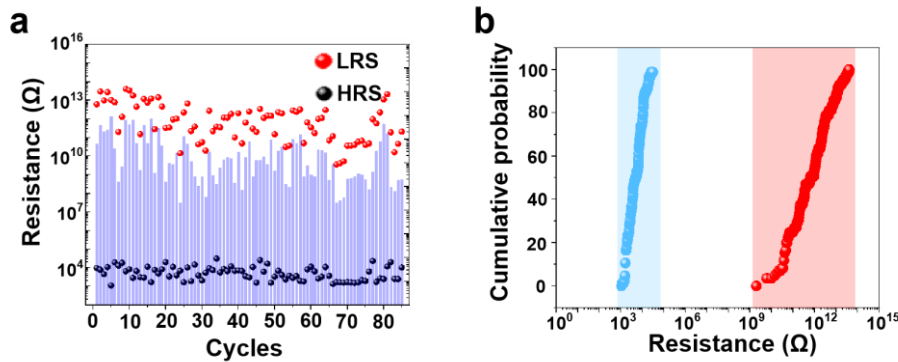
**Supplementary Fig. 14 | OM image of memristors.** **a**,  $\text{Ti}/\text{SmO}_2/\text{Sm}_2\text{O}_3/\text{Au}$ . Scale bars,  $5\ \mu\text{m}$ . **b**,  $\text{Ti}/\text{SmO}_2/\text{Sm}_2\text{O}_3/\text{Gr}$ . Scale bars,  $5\ \mu\text{m}$



**Supplementary Fig. 15 | Transport mechanism analysis of the Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub> memristor with Au and Gr bottom electrodes.** **a** and **b**,  $I$ - $V$  characteristics of Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Gr and Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Au during the RESET operation. **c**, The  $\ln(I)$  versus  $\sqrt{V}$  for the Au-device in the HRS. **d**, Temperature-dependent resistance of the Gr- and Au-devices in HRS measured from 300 K to 100 K.

The  $I$ - $V$  characteristics of Gr-bottom-electrode device is shown in Fig. S15a. As a semimetal, graphene possesses conduction and valence bands that meet at the Dirac point, exhibiting a linear dispersion relation rather than the parabolic dispersion characteristic of conventional metals<sup>1</sup>. This electronic structure allows the Fermi level of graphene to be modulated via external stimuli, such as electric fields or chemical doping. Unlike conventional metal electrodes, electron transfer between graphene and semiconductors mitigates pinning, enabling the Fermi level of graphene to align naturally with the semiconductor, thereby eliminating the potential barrier<sup>2</sup>. Consequently, the experiment observed Ohmic contact behavior in the HRS. Furthermore, the heterogeneous electron transfer (HET) dynamics are determined not only by the driving force (potential difference) but also by electronic coupling<sup>3</sup>. Electronic coupling reflects the probability of electron tunneling across the interface and stems from the density of states (DOS) of the electrode material. In contrast, upon switching to the LRS where oxygen vacancy conductive filaments penetrate the interface, localized states on the graphene surface introduce intermediate energy levels and modulate the interface barrier. These modifications cause the electron injection efficiency to vary non-linearly with voltage, leading to the sublinear transport characteristic of  $I \sim V^{0.46}$  observed experimentally. This sublinear transport characteristic serves as a macroscopic manifestation of HET kinetics being modulated by localized defect states.

Conversely, for the Au-electrode devices (Fig. S15b), the double-logarithmic  $I$ - $V$  plot reveals a clear three-regime behavior: an Ohmic region ( $n \approx 1$ ) at low voltages, a Child's law region ( $n \approx 2$ ) at intermediate voltages, and a trap-filled limit (TFL) region with a steep exponent of  $n \approx 14$  at high voltages. This sequential transition is characteristic of space-charge-limited conduction (SCLC) mediated by an exponential distribution of bulk traps<sup>4</sup>. The high exponent in the TFL region is suggestive of rapid filling of deep-level traps by injected carriers, leading to a sudden current surge once all traps are occupied. Crucially, temperature-dependent resistance measurements for the Au-electrode devices (Fig. S15d) conducted from 300K down to 100 K show negligible variation in the HRS. This weak temperature dependence, combined with the absence of a linear regime in the  $\ln(I)$  versus  $V$  plot across the entire voltage range, does not support thermally activated processes such as Schottky or Poole-Frenkel emission, and is more compatible with defect-mediated tunneling transport<sup>5</sup>. While we acknowledge that the transport analysis alone does not uniquely prove the switching mechanism, these observations are consistent with the presence of a high density of oxygen vacancies, which are the likely candidate defects responsible for the valence-change switching<sup>6</sup>.



**Supplementary Fig. 16 | Statistical distribution of HRS and LRS during 85 cycles for Gr-based device(CC = 50μA).** **a**, High/low resistance states (H/LRS) and switch ratio of Au/Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Gr memristor under 85 cycles(CC= 50μA). **b**, Cumulative probability of the resistance levels at the LRS and HRS(CC= 50μA).

## Supplementary Note 1: Thermal Stability Assessment of the Conductive Filament During Device Operation

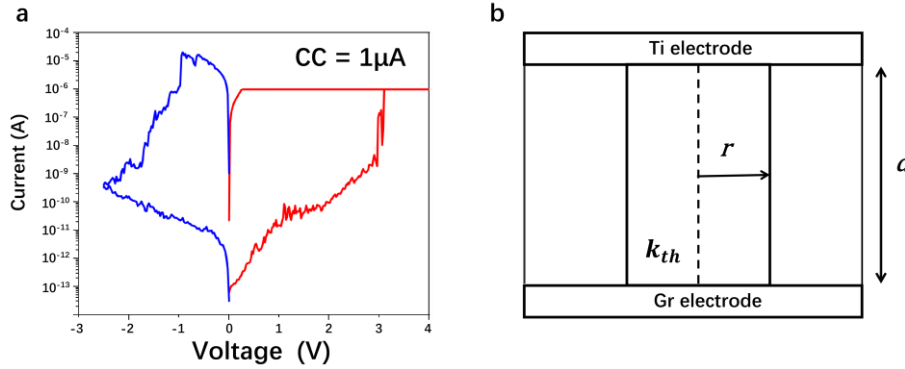
The I-V characteristics were measured with a 1  $\mu$ A compliance current to prevent excessive heating and a slow voltage sweep to ensure quasi-steady-state conditions. To preliminarily estimate the operating temperature of the device, a simplified thermal model was constructed based on the vertical structure, where the oxygen-vacancy conductive filament is approximated as a cylindrical channel bridging the two electrodes. Under these quasi-steady-state conditions, the filament temperature is derived from the following equations:

$$P = I \times V \quad (1)$$

$$T = T_0 + \frac{P}{R_{th}} \quad (2)$$

$$R_{th} = \frac{d}{k_{th} \times A} \quad (3)$$

where  $P$  is the Joule heating power,  $I$  and  $V$  are the measured current and voltage respectively,  $T_0$  is the ambient temperature of 300K,  $d$  is the oxide thickness (filament length, 35nm),  $R_{th}$  is the thermal resistance,  $k_{th}$  is the thermal conductivity, and  $A = \pi r^2$  represents the filament horizontal cross-section area,  $r$  denotes the filament radius which is estimated from the current distribution measured by C-AFM, giving  $r \approx 25$ nm.

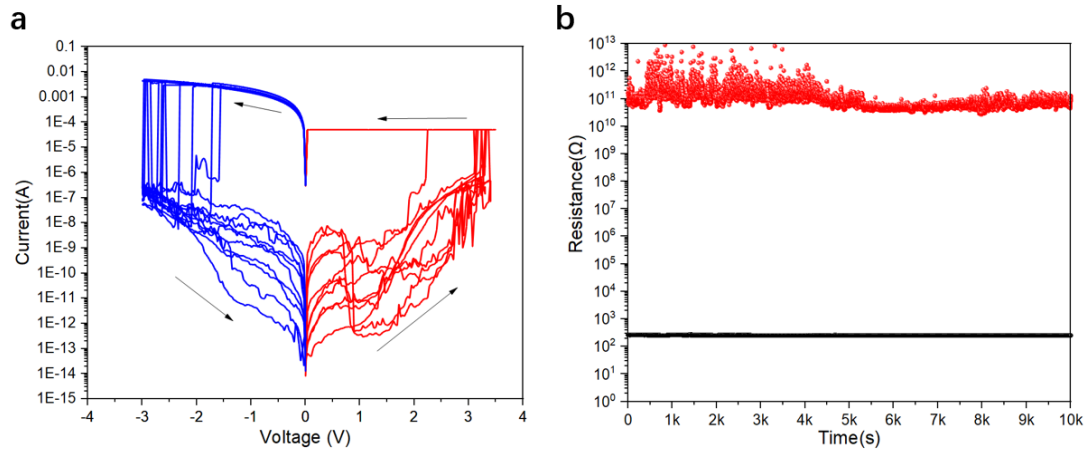


**Supplementary Fig. 17 | Thermal Stability Assessment During Device Operation** **a**, I-V switching curve of a Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Gr memristor at compliance current(CC) = 1  $\mu$ A. **b**, Schematic diagram of one-dimensional simplified thermal model for memristor.

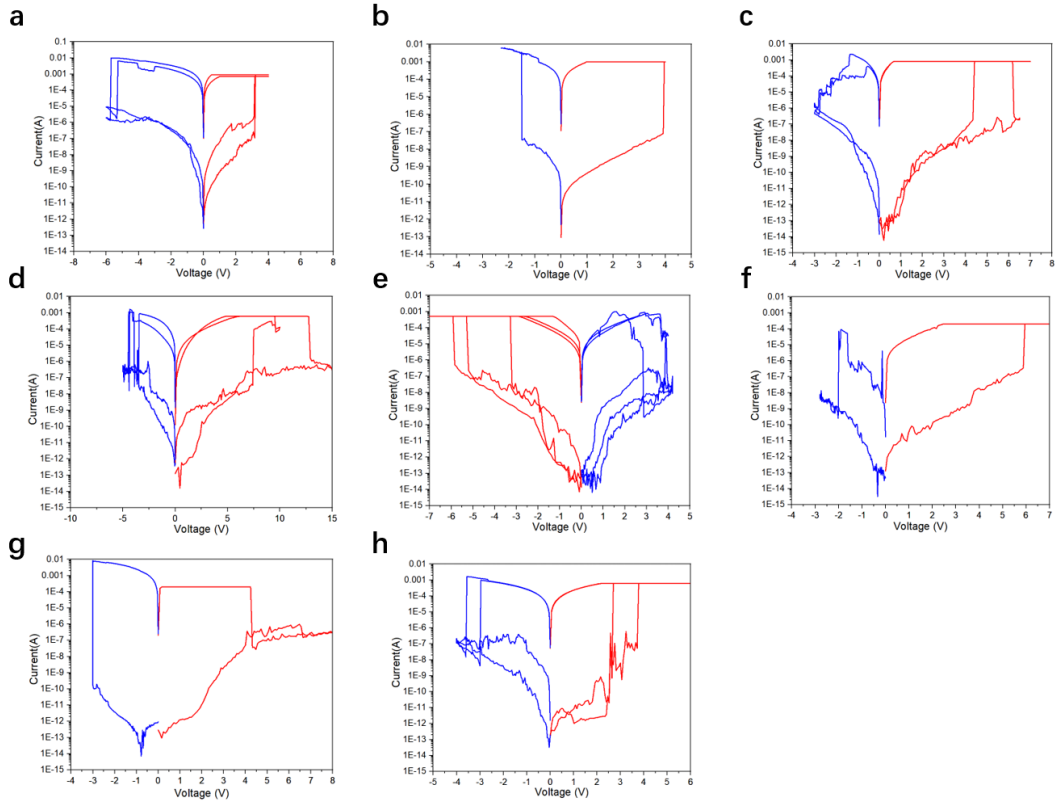
The thermal conductivity of epitaxially grown single-crystalline rare-earth oxide films is approximately five times that of SiO<sub>2</sub><sup>7</sup>. Given a SiO<sub>2</sub> thermal conductivity of  $\sim 1.3$  W/m·K<sup>8</sup> the conductivity is estimated at  $\sim 6.5$  W/m·K. However, in oxygen-vacancy-rich filaments, high defect concentrations induce strong phonon scattering. According to the Klemens model and prior simulations on oxide memristor filaments<sup>9</sup>, the local thermal conductivity in such regions can decrease by at least one order of magnitude relative to the pristine lattice. To ensure a rigorous assessment, we adopt a thermal conductivity of  $k_{th} = 0.65$  W/m·K as a reasonable baseline for the filament region. Regarding the power parameters used in the model, although the maximum RESET power extracted from the measured I-V cycles is  $\sim 1.89$   $\mu$ W, we utilize a value of  $P \approx 10$   $\mu$ W to account for device-to-device variations.

The peak filament temperature is estimated at 574 K, representing a conservative upper bound for the actual operating temperature because the one-dimensional model neglects radial heat loss

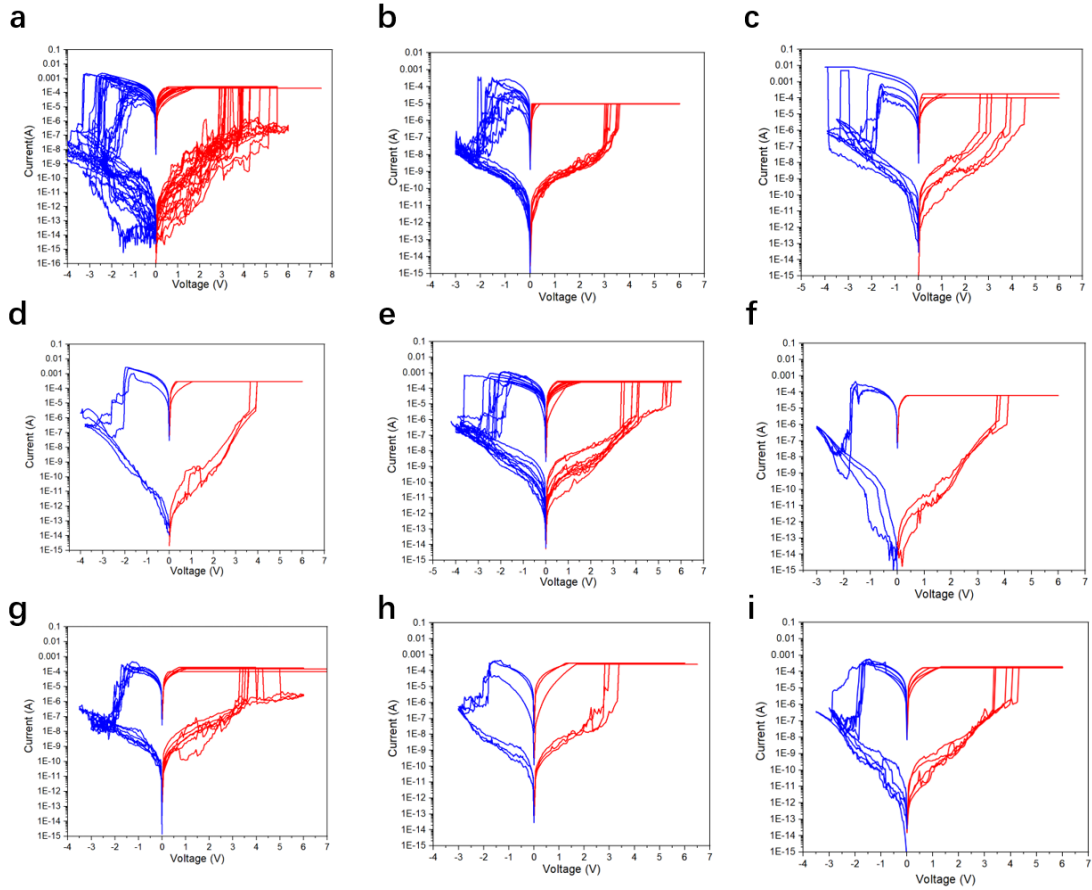
and electrode heat-sink effects. Serving as a qualitative assessment rather than an exact measurement, this estimate aims to determine whether thermal effects could trigger irreversible phase transformations. Under these assumptions, the conclusion that the peak temperature remains below the phase-transition threshold remains credible, suggesting the potential for thermally stable device operation under low-compliance-current measurement conditions. We have added the relevant content to the supplementary information.



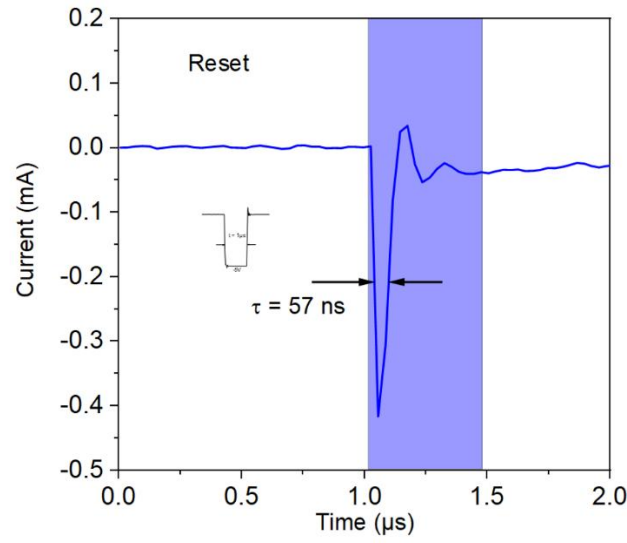
**Supplementary Fig. 18 | Electrical characteristics of the Ti/Sm<sub>2</sub>O<sub>3</sub>/Au. a,  $I$ - $V$  switching curves of a Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Au memristor. b, Retention time characteristic of Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Au memristor.**



**Supplementary Fig. 19 |  $I$ - $V$  characteristics of multiple Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Au memristors.**



**Supplementary Fig. 20 |  $I$ - $V$  characteristics of multiple Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Gr memristors.**



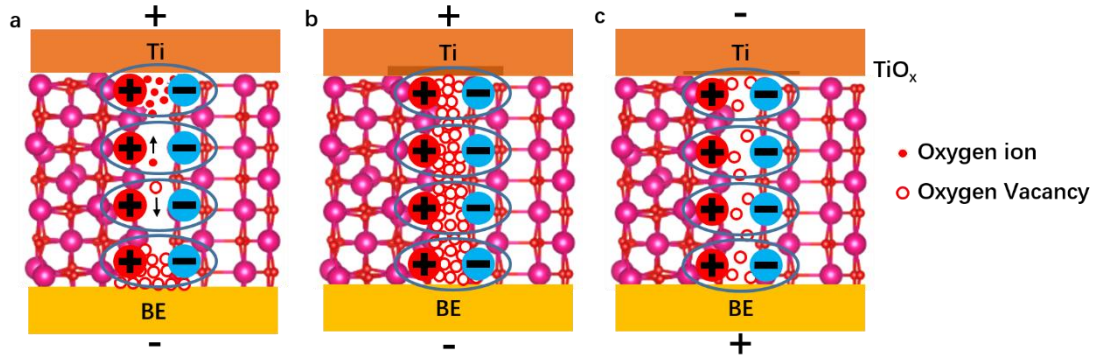
**Supplementary Fig. 21 | Reset behavior of Ti/SmO<sub>2</sub>/Sm<sub>2</sub>O<sub>3</sub>/Gr memristor at -5V(1μs) pulse.**  
The current response confirming a transition to HRS, shows the highest speed with a switching time of 57 ns.

**Table S1 Comparison of on/off ratio and switching time with various devices**

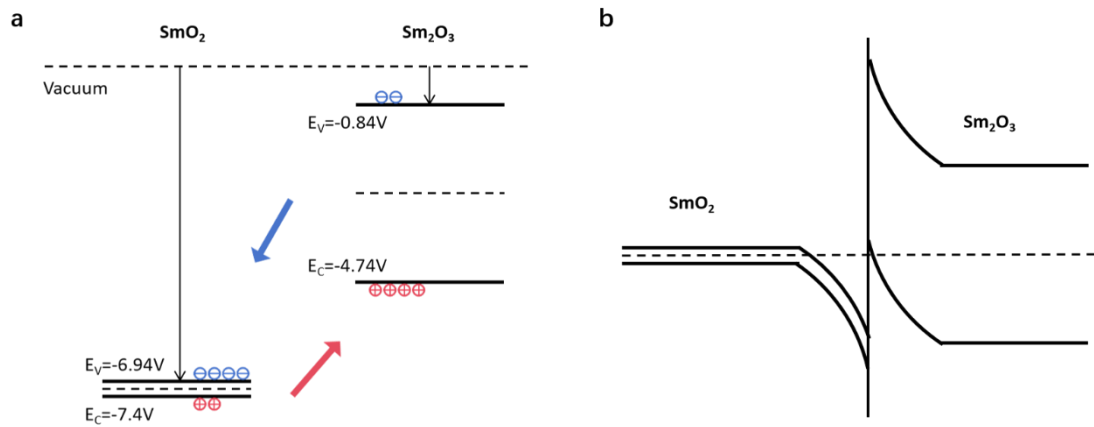
| No. | Device                                                     | on/off ratio     | Switching time | Reference |
|-----|------------------------------------------------------------|------------------|----------------|-----------|
| 1   | F-doped TiO <sub>2-x</sub>                                 | >10 <sup>4</sup> | 77.7 ns        | 10        |
| 2   | (MTS)h-BN/HfO <sub>2</sub>                                 | ~100             | 4.2 ns         | 11        |
| 3   | Gr/MoS <sub>2-x</sub> O <sub>x</sub> /Gr                   | 12               | <100 ns        | 12        |
| 4   | h-BN                                                       | ~10 <sup>3</sup> | 680 ns         | 13        |
| 5   | Ti/h-BN/Au                                                 | 50               | 120 ps         | 14        |
| 6   | MoS <sub>2</sub>                                           | ~10 <sup>6</sup> | 330 ps         | 15        |
| 7   | Ti/SiN <sub>x</sub> /p+Si                                  | ~10              | 500 ns         | 16        |
| 8   | Ag/TiO <sub>2</sub> /Pt                                    | ~10 <sup>3</sup> | 20 ns          | 17        |
| 9   | Ta/Ta <sub>2</sub> O <sub>5</sub> /AlN/Gr                  | ~100             | 50 ns          | 18        |
| 10  | Pd/SiO <sub>x</sub> /Gr                                    | >10 <sup>6</sup> | 35 ns          | 19        |
| 11  | Ta/HfO <sub>2</sub> /Pt                                    | >10              | 5 ns           | 20        |
| 12  | Pt/HfO <sub>2</sub> /Pt                                    | ~10 <sup>6</sup> | 120 ns         | 21        |
| 13  | Ti/HfSe <sub>x</sub> O <sub>y</sub> /HfSe <sub>2</sub> /Au | ~10 <sup>3</sup> | <50 ns         | 22        |
| 14  | Ti/PdSe <sub>2</sub> /Au                                   | ~10 <sup>3</sup> | 50 ns          | 23        |
| 15  | Ti/SmO <sub>2</sub> /Sm <sub>2</sub> O <sub>3</sub> /Gr    | >10 <sup>9</sup> | 47.3 ns        | This work |

**Table S2 Comparison of on/off ratio with various samarium oxide-based memristors**

| Device                                                                                      | Synthesis method        | on/off ratio      | retention time(s) | year | reference |
|---------------------------------------------------------------------------------------------|-------------------------|-------------------|-------------------|------|-----------|
| Pt//Sm <sub>2</sub> O <sub>3</sub> /TiN                                                     | RF magnetron sputtering | 10 <sup>2.2</sup> | 10 <sup>4</sup>   | 2011 | 24        |
| Pt//Sm <sub>2</sub> O <sub>3</sub> /TiN                                                     | RF magnetron sputtering | 10 <sup>2.5</sup> | 10 <sup>4</sup>   | 2011 | 25        |
| Ru/SmO <sub>x</sub> /TaN                                                                    | RF magnetron sputtering | 10 <sup>3</sup>   | 10 <sup>5</sup>   | 2012 | 26        |
| Ni/Sm <sub>2</sub> O <sub>3</sub> /ITO                                                      | RF magnetron sputtering | 10 <sup>2</sup>   | 10 <sup>5</sup>   | 2013 | 27        |
| Ni//Sm <sub>2</sub> O <sub>3</sub> /ITO                                                     | RF magnetron sputtering | 10 <sup>2</sup>   | 10 <sup>5</sup>   | 2014 | 28        |
| Pt/V <sub>2</sub> O <sub>5</sub> //Sm <sub>2</sub> O <sub>3</sub> /TiN/SiO <sub>2</sub> /Si | RF magnetron sputtering | 10 <sup>2</sup>   | -                 | 2018 | 29        |
| Al/V <sub>2</sub> O <sub>5</sub> //Sm <sub>2</sub> O <sub>3</sub> /TiN/SiO <sub>2</sub> /Si | RF magnetron sputtering | 10 <sup>3</sup>   | 10 <sup>4</sup>   | 2019 | 30        |
| Ag/Sm <sub>2</sub> O <sub>3</sub> /Pt                                                       | -                       | 10 <sup>4</sup>   | 10 <sup>4</sup>   | 2019 | 31        |
| Ti/SmO <sub>2</sub> /Sm <sub>2</sub> O <sub>3</sub> /Gr                                     | Eutectic Salt-Assisted  | >10 <sup>9</sup>  | 10 <sup>4</sup>   | 2026 | This work |

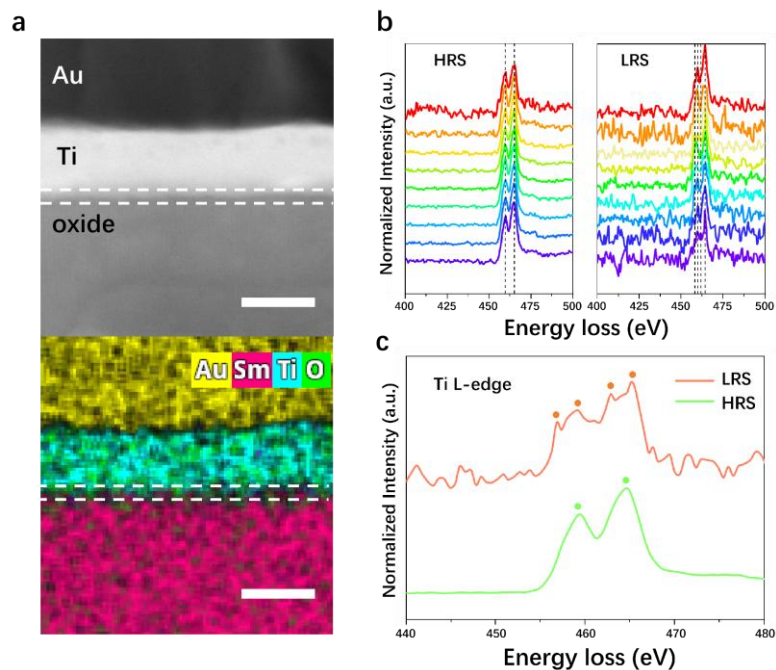


**Supplementary Fig. 22 | Schematic of the resistive switching mechanism.**

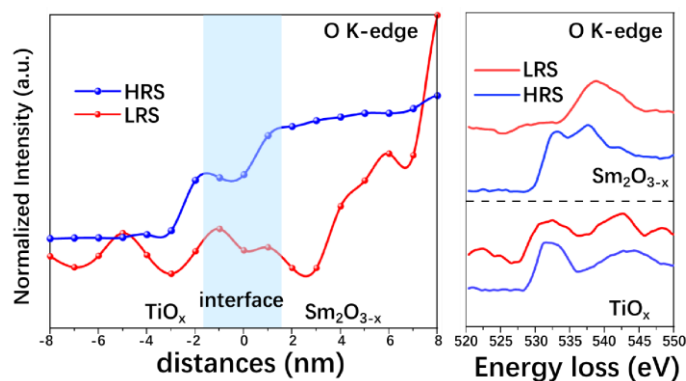


**Supplementary Fig. 23 | Calculated band structure of  $\text{Sm}_2\text{O}_3$  and  $\text{SmO}_2$  crystals. a,** Energy band and carrier injection of  $\text{SmO}_2$  and  $\text{Sm}_2\text{O}_3$  crystals. **b,** Energy band diagram near the interface of  $\text{SmO}_2/\text{Sm}_2\text{O}_3$  heterostructure.

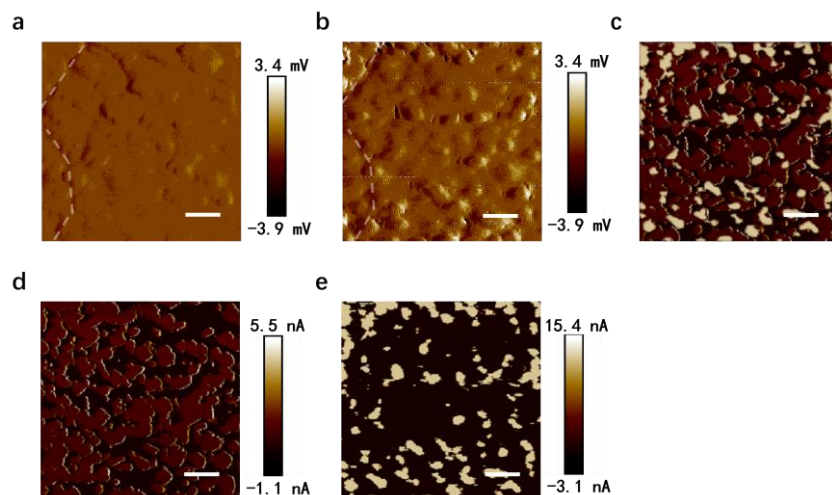
Upon contact between  $\text{Sm}_2\text{O}_3$  and  $\text{SmO}_2$ , charge redistribution occurs to equalize their Fermi levels. This process involves the transfer of electrons from  $\text{Sm}_2\text{O}_3$  into  $\text{SmO}_2$ , driven by the energetically favorable alignment where the conduction band minimum of  $\text{SmO}_2$  lies approximately 2.2 eV lower than the valence band maximum of  $\text{Sm}_2\text{O}_3$ . Consequently, the difference in Fermi levels between  $\text{SmO}_2$  and  $\text{Sm}_2\text{O}_3$  leads to charge transfer, where holes transfer from  $\text{SmO}_2$  to  $\text{Sm}_2\text{O}_3$  and electrons transfer from  $\text{Sm}_2\text{O}_3$  to  $\text{SmO}_2$  respectively. This process consequently induces downward energy band bending at the  $\text{SmO}_2$  interface and upward bending at the  $\text{Sm}_2\text{O}_3$  interface.



**Supplementary Fig. 24 | Cross-sectional TEM-EELS chemical analysis of Ti at the electrode interface in HRS and LRS.** **a**, TEM image and EDS mapping of the interface. Scale bar, 10nm. **b**, Ti L-edge EEL spectra as a function of position for the device in the HRS and LRS. The spectra were acquired across the Ti electrode from the bottom to the top. **c**, Comparison of Ti L-edge EEL spectra between HRS and LRS. (c) O K-edge normalized intensity for the same region in the HRS and LRS.



**Supplementary Fig. 25 | Cross-sectional TEM-EELS chemical analysis of O at the electrode interface in HRS and LRS**



**Supplementary Fig. 26 | C-AFM characterization of the memristive device in HRS and LRS.**

**a**, The C-AFM surface deflection at HRS. **b**, The C-AFM surface deflection acquired immediately after the forming process. **c**, Spatially superimposed C-AFM current maps acquired from the same device region in the HRS and LRS. **d**, The C-AFM current mapping images of device at HRS. **e**, The C-AFM current mapping images of device at LRS. All scale bar, 200nm.

## Supplementary Note 2: Computational details

The calculations of density functional theory are done with the projector augmented plane-wave basis, which is implemented in Vienna *ab-initio* simulation package<sup>32</sup>. And the plane-waves are cut-off at 550 eV. The exchange-correlations of electrons are described by the generalized gradient approximations with the form proposed by Perdew, Burke, and Ernzerhof<sup>33</sup>. The 4f electrons of Sm were treated as part of the ionic core. Vacuum layer of 20 Å is set in *z*-direction which is supposed to be nonperiodic to eliminate the interactions between images. Dipole correction is considered. The energy converge criterion for solving self-consistent Kohn-Sham equations is  $10^{-7}$  eV. The Brillouin zone is sampled with resolutions better than  $0.03 \text{ \AA}^{-1}$ , using the scheme of Monkhorst-Pack<sup>34</sup>. All the structures in this study are fully relaxed until the Hellman-Feynman smaller than  $0.05 \text{ eV/\AA}$ .

## Reference

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