Electronic Supplementary Material (ESI) for Nanoscale Advances. This journal is © The Royal Society of Chemistry 2022

## **Supplementary Information**

## Uniform self-rectifying resistive random access memory based on MXene-TiO<sub>2</sub> Schottky junction

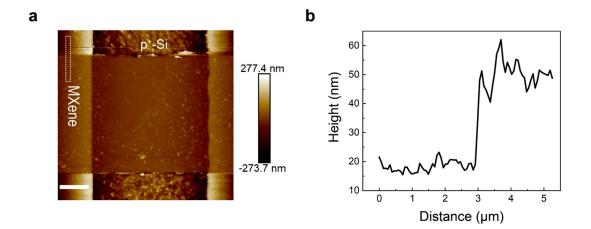
Chao Zang<sup>1,2#</sup>, Bo Li<sup>1,2#</sup>, Yun Sun<sup>1\*</sup>, Shun Feng<sup>1,3</sup>, Xin-Zhe Wang<sup>1,2</sup>, Xiaohui Wang<sup>1,2\*</sup>, Dong-Ming Sun<sup>1,2\*</sup>

<sup>1</sup>Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, 72 Wenhua Road, Shenyang, 110016, China.

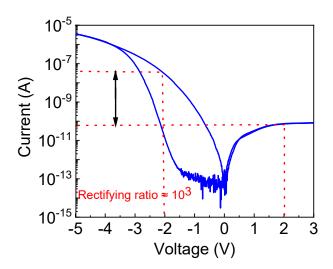
<sup>2</sup>School of Material Science and Engineering, University of Science and Technology of China, 72 Wenhua Road, Shenyang, 110016, China.

<sup>3</sup>School of Physical Science and Technology, ShanghaiTech University, 393 Huaxiazhong Road, Shanghai, 200031, China.

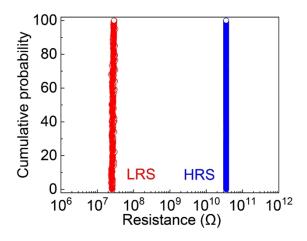
\*These authors were equal major contributors to this work.



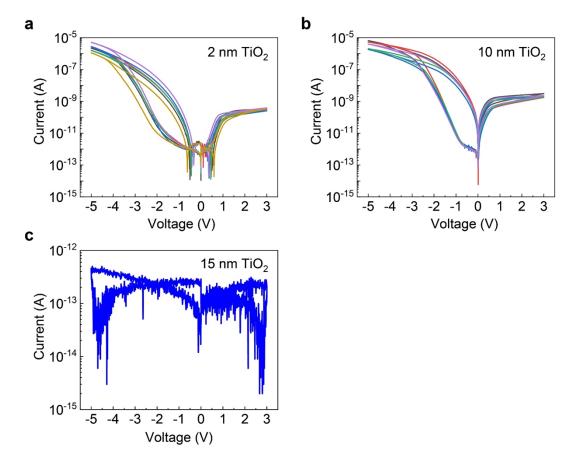
Supplementary Fig. 1 (a) AFM image of the morphology of MXene film. Scale bar, 2  $\mu$ m. (b) Height profile along the white dashed line in (a), indicating a thickness of 25 nm MXene.



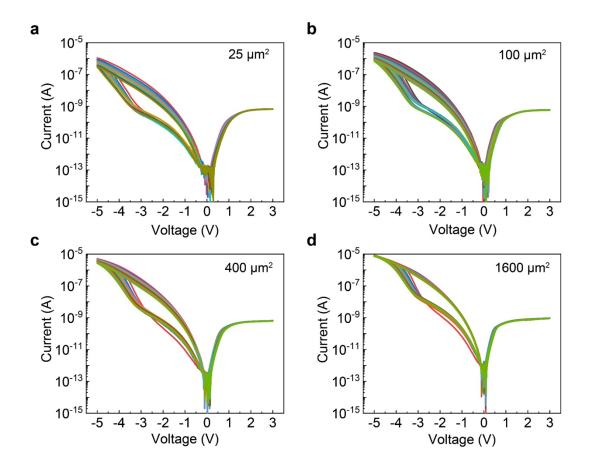
**Supplementary Fig. 2** Typical I-V characteristic of an MXene-TiO<sub>2</sub>-Si RRAM with a rectifying ratio of  $10^3$  obtained at 2 V and -2 V.



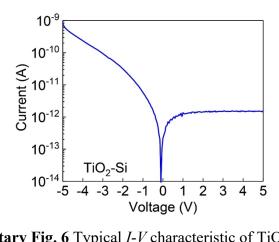
Supplementary Fig. 3 The read disturbance under 2 V/-2 V for HRS and LRS.



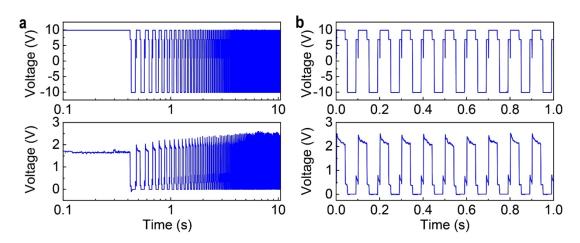
**Supplementary Fig. 4** *I-V* characteristics of MXene-TiO<sub>2</sub>-Si RRAMs with different thickness of TiO<sub>2</sub>. (a) 2 nm, (b) 10 nm, (c) 15 nm.



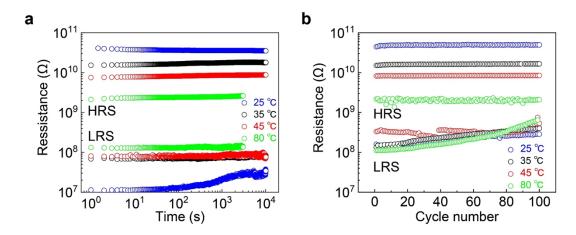
**Supplementary Fig. 5** Typical *I-V* characteristics of MXene-TiO<sub>2</sub>-Si RRAMs with different resistive area, indicating the bipolar SET and RESET processes. (a) 25  $\mu$ m<sup>2</sup>, (b) 100  $\mu$ m<sup>2</sup>, (c) 400  $\mu$ m<sup>2</sup>, (d) 1600  $\mu$ m<sup>2</sup>.



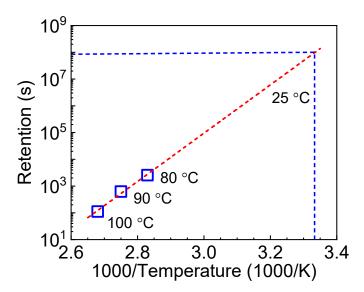
Supplementary Fig. 6 Typical I-V characteristic of TiO<sub>2</sub>-Si junction.



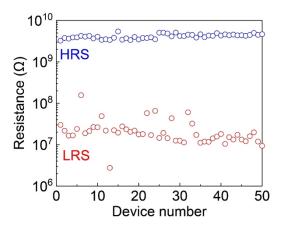
**Supplementary Fig. 7** (a) Endurance test performed with more than 100 voltage sweep cycles in 10 s. (b) About 10 cycles in 1s cut from (a).



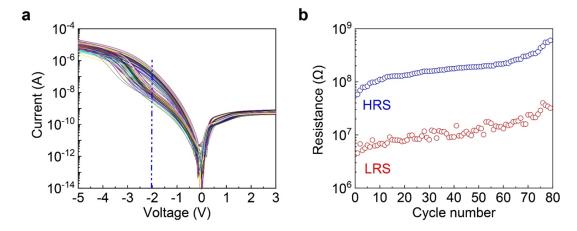
**Supplementary Fig. 8** (a) Retention and (b) endurance characteristics at the different temperatures.



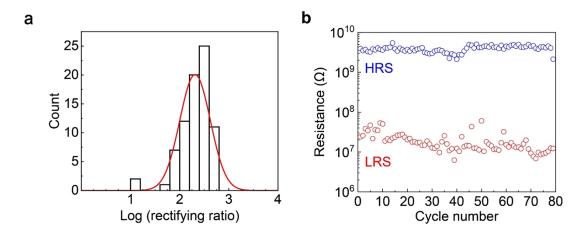
**Supplementary Fig. 9** Temperature dependence of the retention time (s) follows the Arrhenius relation. Retention time at 25 °C are extrapolated from the linear fitting of measurement results (blue squares).



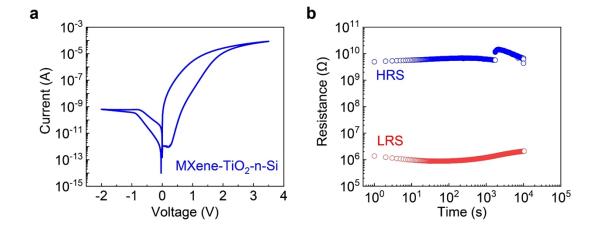
**Supplementary Fig. 10** Device-to-device variation of HRS and LRS at the read voltage of 2 V, respectively.



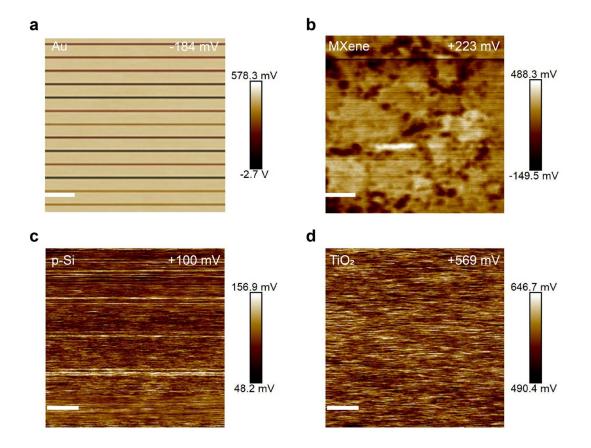
**Supplementary Fig. 11** (a) The *I-V* characteristics of a single RRAM device with 80 cycles. (b) The extracted cycle-to-cycle distribution of HRS and LRS at 2 V read voltage. The resistance states can be also distinguished through the current on-off ratio more than 10



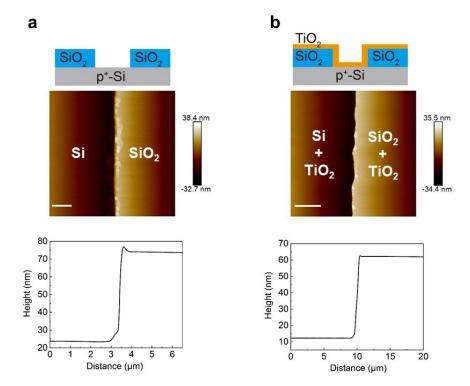
**Supplementary Fig. 12** The statistical cycle-to-cycle variation of (a) rectifying ratio performed by a logarithmic operation and (b) HRS and LRS, respectively.



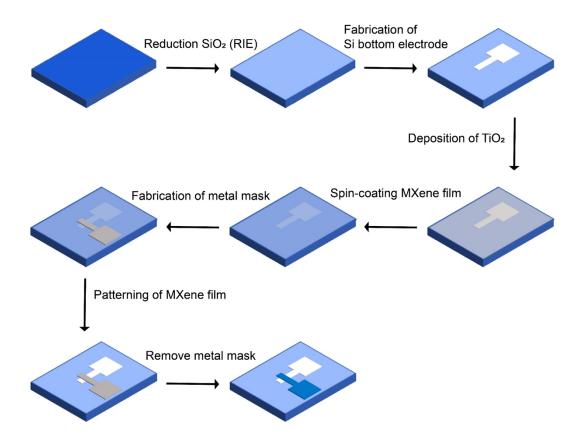
**Supplementary Fig. 13** (a) Self-rectifying behavior and (b) retention characteristics of the MXene-TiO<sub>2</sub>-n-Si.



Supplementary Fig. 14 The Kelvin probe force microscopy (KPFM) potential map images and numerical analysis of (a) Au (scale bar: 840 nm), (b) MXene (scale bar: 1  $\mu$ m). (c) p<sup>+</sup>-Si (scale bar: 1  $\mu$ m) and (d) TiO<sub>2</sub> (scale bar: 1  $\mu$ m). The potentials of Au, MXene, p<sup>+</sup>-Si, and TiO<sub>2</sub> were -184 mV, +223 mV, +100 mV and +569 mV, respectively. The work function of Au is 5.2 eV. Therefore, the work function of MXene is 5.2 – 0.184 – 0.223 = 4.893 eV, p<sup>+</sup>-Si is 5.2 – 0.184 – 0.1 = 5.016 eV and TiO<sub>2</sub> is 5.2 – 0.184 – 0.569 = 5.547 eV. (KPFM probe: SCM-PIT-V2, 0.01-0.025 Ohm-cm Antimony (n) doped Si).



Supplementary Fig. 15 The schematic, corresponding AFM and height of Si electrode (a) before and (b) after 5-nm-TiO<sub>2</sub> deposited by ALD. Scale bar, 1  $\mu$ m, 4  $\mu$ m.



**Supplementary Fig. 16** Fabrication flowchart of MXene-TiO<sub>2</sub>-Si self-rectifying RRAMs.

Table S1. Performance comparison about TiO<sub>2</sub>-based RRAM devices.

Structure	on/off ratio	rectifying ratio	Retention (s)	endurance
Ti/TiO <sub>2</sub> /Pt <sup>[1]</sup>	$10^{3}$	$10^{5}$		1000
$Cu/Ti/HfO_2/TiO_2/TiN^{[2]}$	$3 \times 10^3$			200
$TaO_x/TiO_2/TaO_x^{[3]}$	$10^{3}$		$10^{4}$	$10^{10}$
$TiN/Ti/TiO_x/HfO_x/TiN^{[4]}$	$10^{3}$		$10^{4}$	
$Pt/In,Sn_2O_3/TiO_2/Pt^{[5]}$	160			40
$TiN/TiO_x/Al_2O_3/IrO_x^{[6]}$	10	10	$10^{5}$	1000
$TiN/TiO_x/Al_2O_3/IrO_x^{[7]}$	30			$10^{6}$
My work	$10^3$	$10^3$	$10^{4}$	100

<sup>[1]</sup> Appl. Phys. Lett. 2020, 96, 262901.

<sup>[2]</sup> Advanced Electronic Materials 2020, 6 (9), 2000488 .

<sup>[3]</sup> ACS Nano 2012, 6, 9, 8166-8172.

<sup>[4]</sup> Appl. Phys. Lett. 2012, 101, 103506.

<sup>[5]</sup> Appl. Phys. Lett. 2008, 92, 162904.

<sup>[6]</sup> IEEE Transactions on Electron Devices 2018, 65 (3), 957-962 □.

<sup>[7]</sup> ACS omega 2017, 2 (10), 6888-6895□.