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

Supporting Information

'Stateful' Threshold Switching for Neuromorphic Learning

Zhijian Zhong[1,†], Zhiguo Jiang[1,†], Jianning Huang[1], Fangliang Gao[1,✉], Wei Hu[2], Yong Zhang[1] & Xinman Chen[1,✉]

[1] *Guangdong Engineering Research Center of Optoelectronic Functional Materials and Devices, Institute of Semiconductors, South China Normal University, Guangzhou 510631, PR China*

[2] *Key Laboratory of Optoelectronic Technology and System of Ministry of Education, College of Optoelectronic Engineering, Chongqing University, Chongqing 400044, PR China*

✉E-mail: gaofl@m.scnu.edu.cn (F. Gao); chenxinman@m.scnu.edu.cn (X. Chen).

† Z. Zhong and Z. Jiang equally contribute to this work.

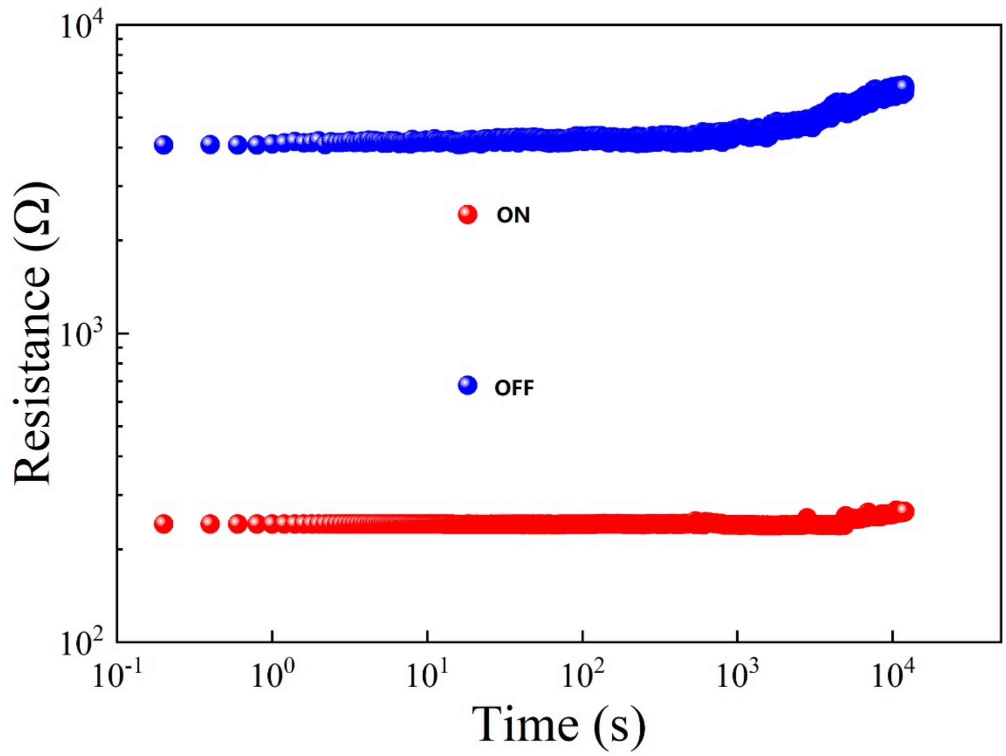

Fig. S1 Resistance retention of ON and OFF states in RS within 12000 seconds.

Figure S1 shows resistance retention of ON and OFF states in RS within 12000 seconds. Both states maintain for a long time with no resistance degradation, indicating that ON and OFF states are nonvolatile. The OFF/ON resistance ratio is ~20.

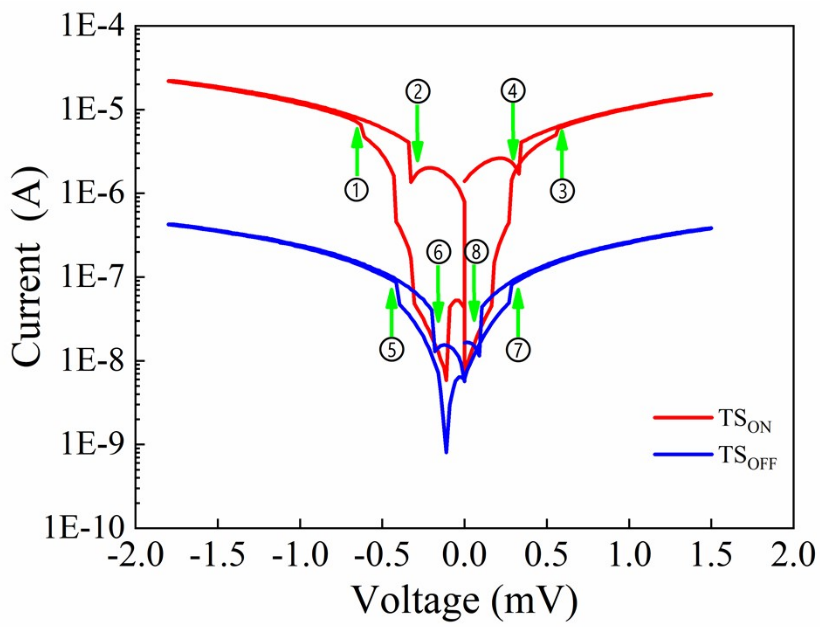

Fig. S2 I-V curves of threshold switching (−1.8 mV–1.5 mV–−1.8 mV) in OFF and ON states (TS_{OFF} and TS_{ON}) of RS. ①–⑧ signs all transition voltage in threshold switching.

Figure S2 shows the classic TS with four transition processes in both ON and OFF states. In TS_{ON} (red line), the device switches from OFF to ON state at ① and turns back to OFF state at ② during the negative sweeping process; During the positive sweeping process, the device switches to ON state at ③ and turns back to OFF state at ④. Similarly in the TS_{OFF} state, the device switches from OFF to ON state at ⑤ and turns back to OFF state at ⑥ during the negative sweeping process; During the positive sweeping process, the device switches to ON state at ⑦ and turns back to OFF state at ⑧.

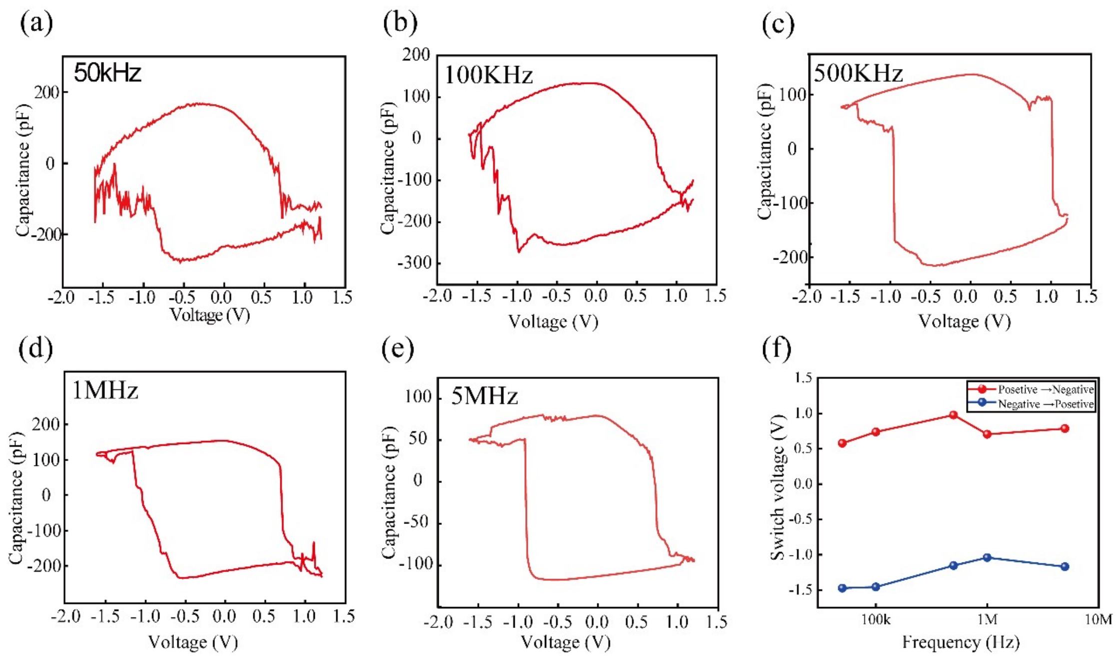

Fig. S3 (a)-(e) C-V curves of Ag/HfO_x/ITO memristor in the frequency from 50 kHz to 5 Mhz by applying 30 mV AC signal. (f) The switching voltages of capacitance polarity in different frequencies.

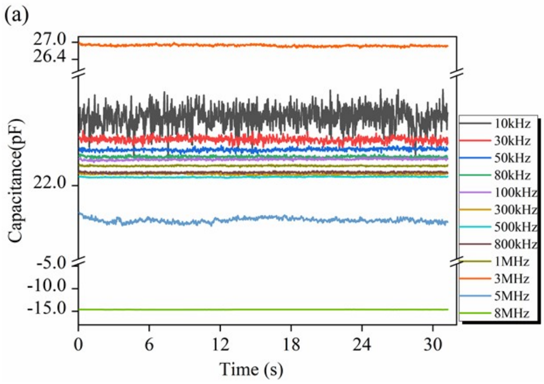

Fig. S4 Capacitance retention of Ag/HfO_x/ITO memristor in different frequencies (10 kHz–8 MHz) in (a) OFF and (b) ON states.

Figure S4 illustrates the capacitance retention of Ag/HfO_x/ITO memristor in frequencies from 10 kHz to 8 MHz in OFF (a) and ON (b) states. In the OFF state, positive capacitance declines as frequency increases from 10 kHz to 500 kHz, and enhances from 500 kHz to 3 MHz. Then the capacitance value drops sharply and turns to negative as the frequency is over 5 MHz. In the ON state, the negative capacitance value decreases monotonously from 10 kHz to 8 MHz.

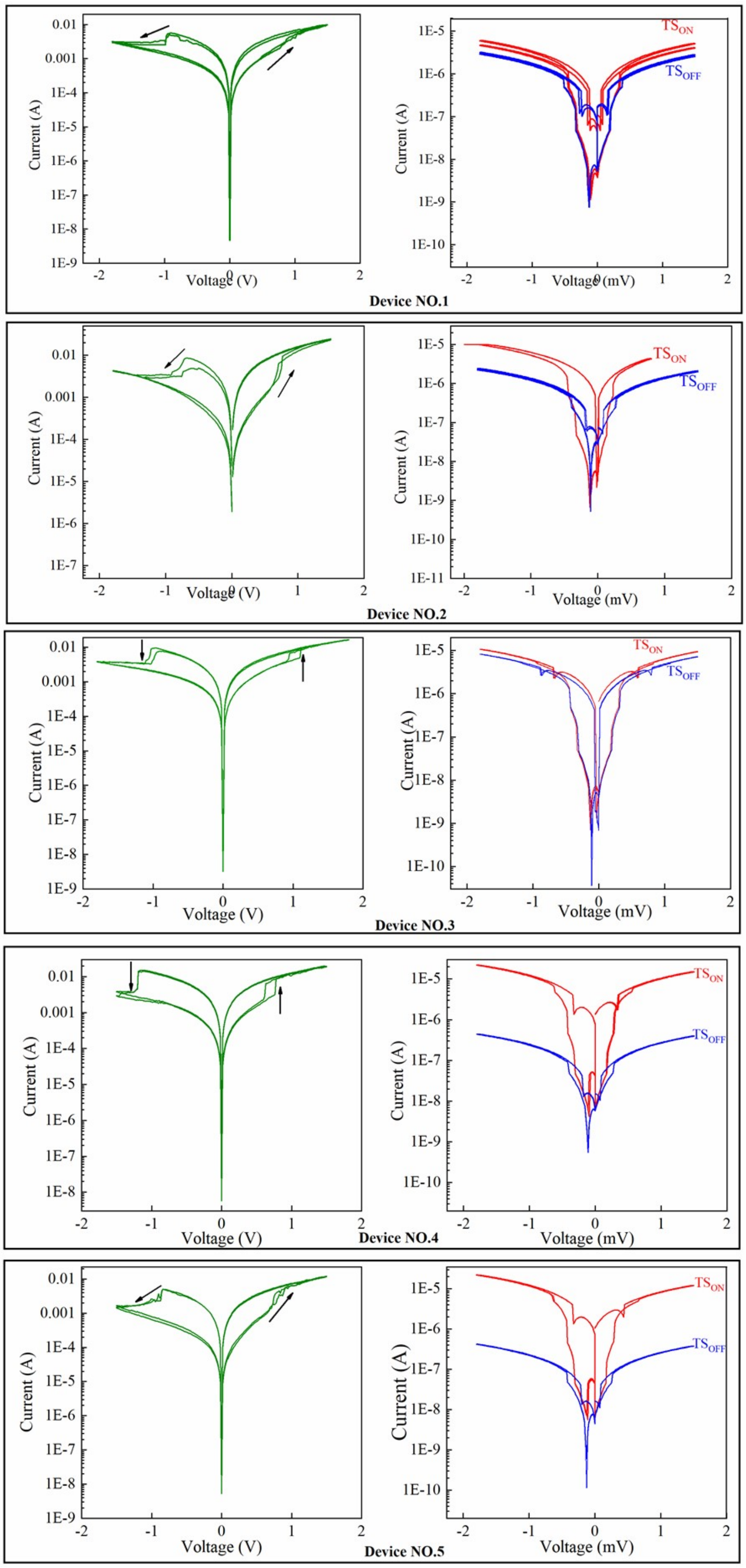

Fig. S5 '*Stateful' TS* of the five optional $Ag/HfO_x/ITO$ devices

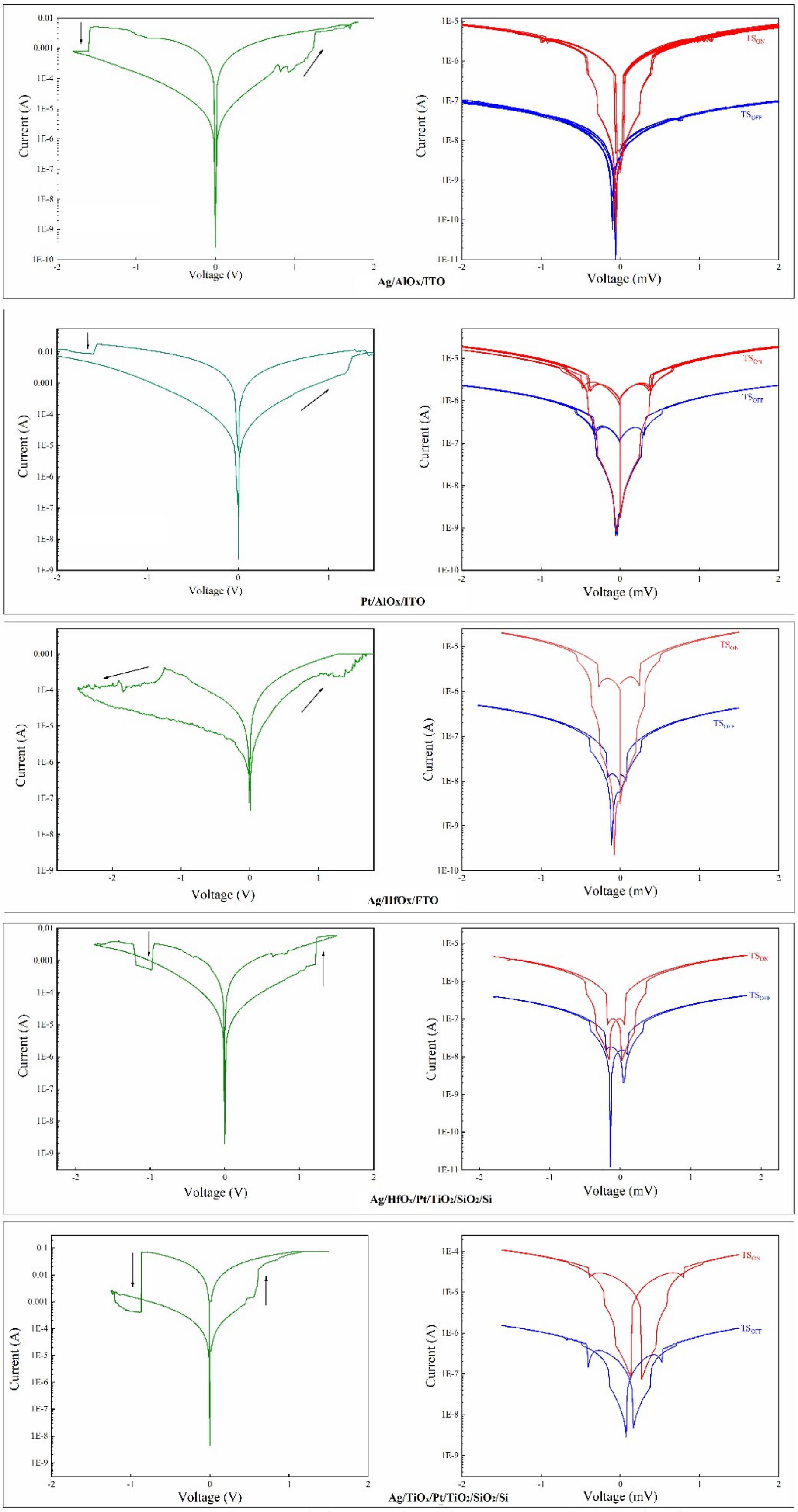

Fig. S6 '*Stateful' TS* of various memristive devices.

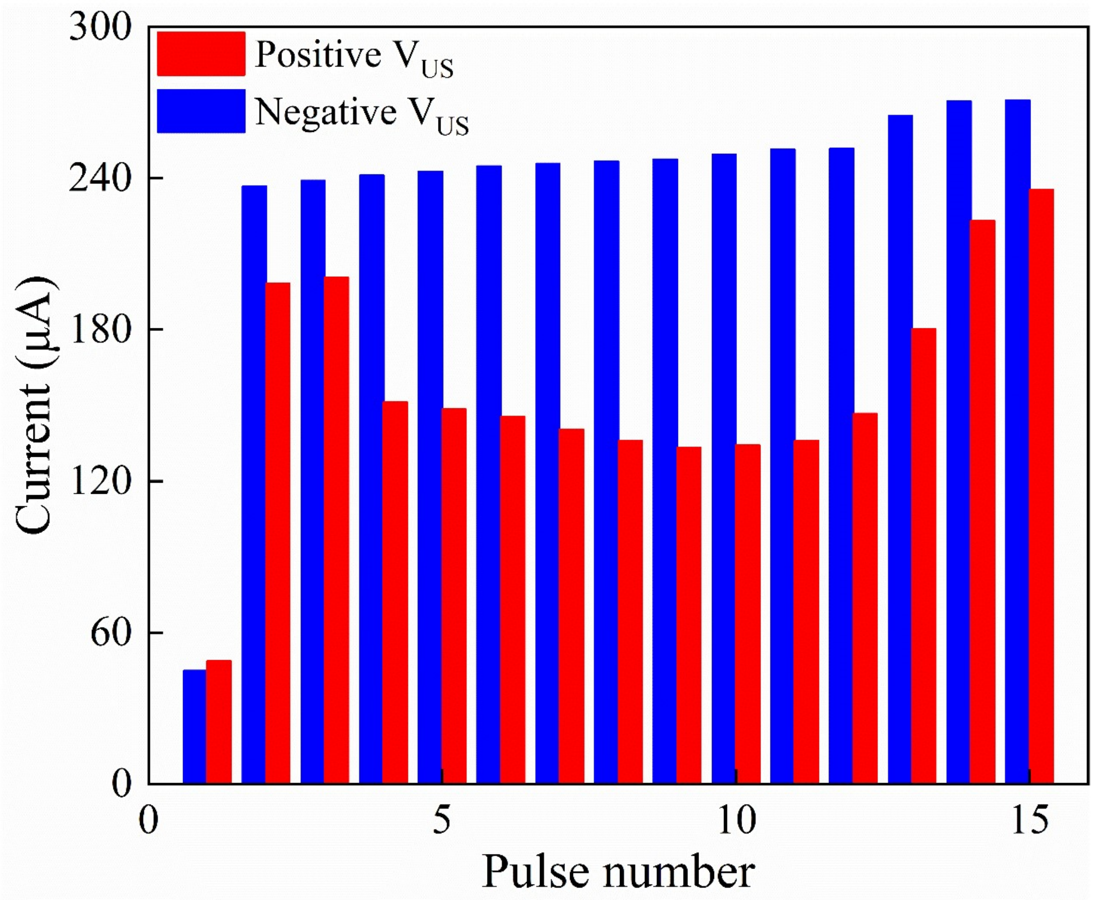

Fig. S7 Evolution of output current under the accompanied V_{US} pulse of 0.75 V during the training process.

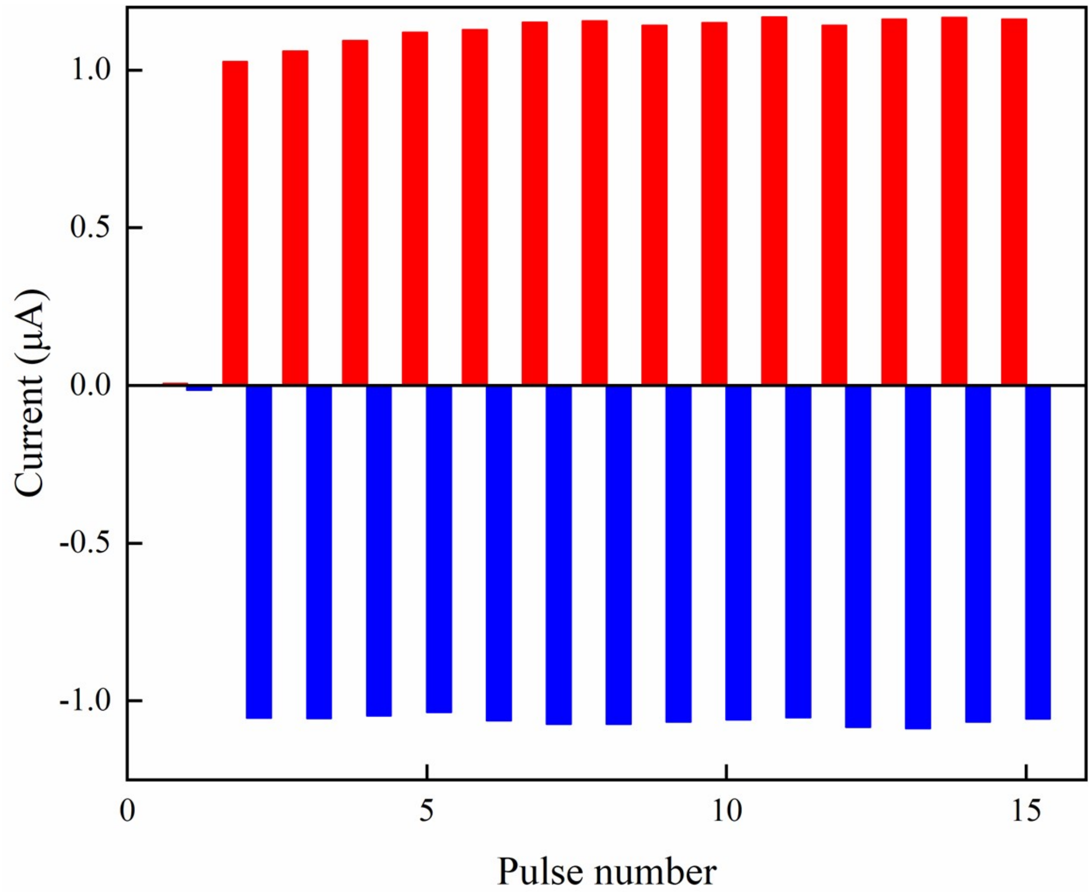

Fig. S8 Training processes by using V_{CS} pulses (0.4 mV, red; −0.4 mV, blue) and V_{US} pulses (1.2 V).

Figure S8 present the response current of a memristive synapse in training processes, which was train by V_{CS} pulses of ±0.4 mV and V_{US} pulses of 1.2 V. Obviously, the device reaches the criterion current of 1 μA by only one training cycle with V_{US} pulse of 1.2 V. It means that associative learning of a memristive synapse can be established by one time training if V_{US} pulse of 1.2 V is used.

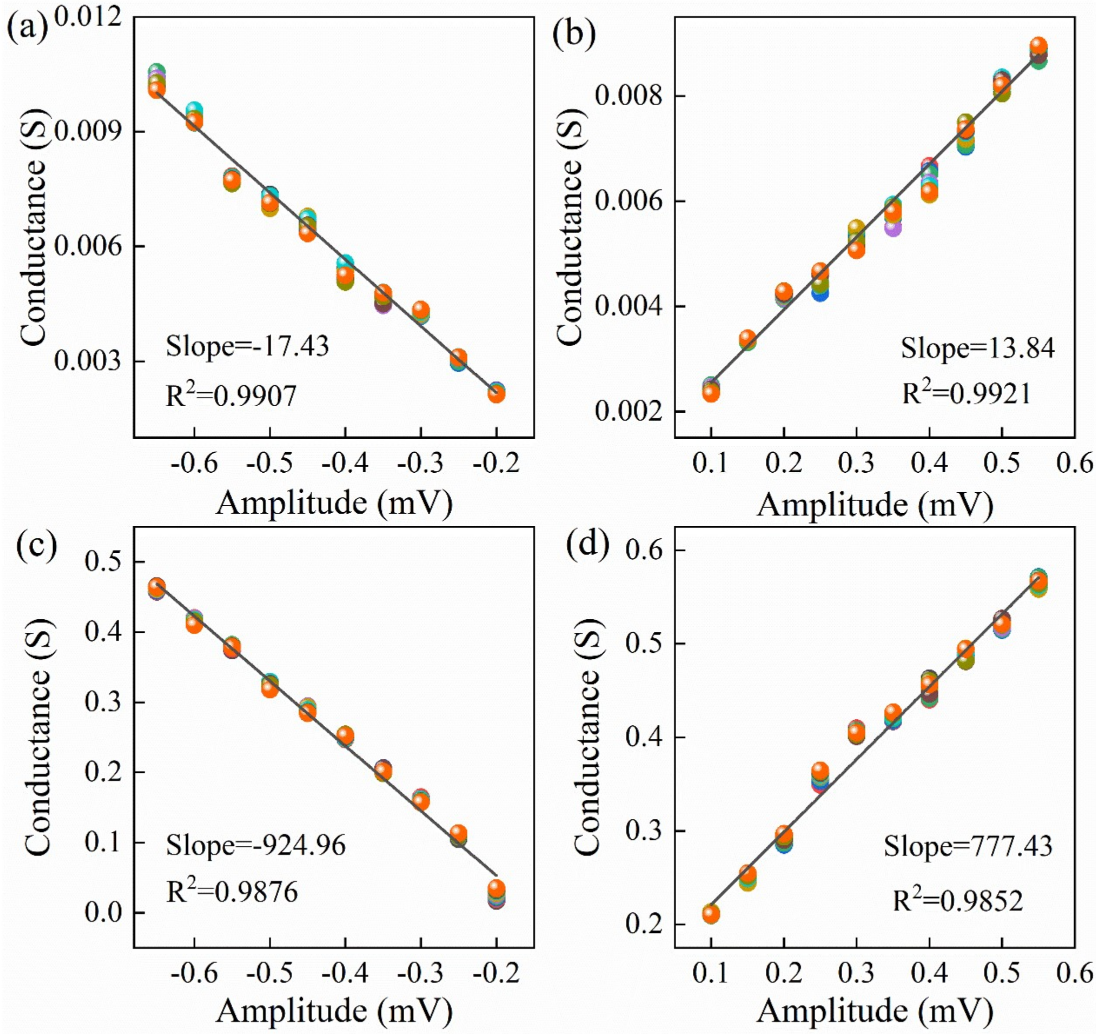

Fig. S9 Linear fitting results of conductance as function of potentiating pulse amplitude. (a) −0.2 mV–−0.65 mV in OFF state, (b) 0.1 mV–0.55 mV in OFF state, (c) −0.2 mV–−0.65 mV in ON state, and (d) 0.1 mV–0.55 mV in ON state.

Table S1 All resistance states available in a typical ‘stateful’ TS.

RS	TS	
	High resistive state	Low resistive state
OFF (0)	00	01
ON (1)	10	11

Table S1 summarizes all resistance states available in a typical 'stateful' TS by considering high and low resistive states in each switching performance, namely RS, TS_{OFF}, and TS_{ON}. To differentiate them, high and low resistance states were only noted as OFF and ON states in RS, they can seem as Boolean logic ‘1’ and ‘0’, respectively. Thus, high and low resistive states in TS_{OFF} can be utilized as ‘00’ and ‘01’, ‘10’ and ‘11’. While ‘00’ and ‘01’ states are the subset of ‘0’ state and ‘10’ and ‘11’ states are the subset of ‘1’ state.

Table S2 Detail parameter value of each components in ON and OFF states of RS.

State	Resistor (R)	Capacitor (C)	Inductor (L) or Resistor (R)	Time constant
ON	45 Ω	100 pF	(L) 0.8 μH	1.8×10^{-8} s
OFF	7000 Ω	200 pF	(R) 30 Ω	9×10^{-9} s

Table S2 lists the detailed parameter value of each parasitic component in OFF and ON states. The time constant (τ) of the charging process can be estimated by considering LR series circuit in ON state and RC parallel circuit in OFF state:

$$\tau_{OFF} = R \times C_{OFF} = 30\Omega \times 300 \times 10^{-12} F = 9 \times 10^{-9} s \tag{S-1}$$

$$\tau_{ON} = L_{ON} \div R_{ON} = 0.8 \times 10^{-6} H \div 45\Omega = 1.8 \times 10^{-8} s \tag{S-2}$$

It can be found that the charging time of the parasitic elements of the device in ON state is twenty times as that in OFF state, which enlarges the area of the non-linear region of the I-V curve in the sweeping process, that is, the storage window in ON state in the I-V curves is larger.