

## Supporting Information

### Convertible resistive switching characteristics between memory switching and threshold switching in a single ferritin-based memristor

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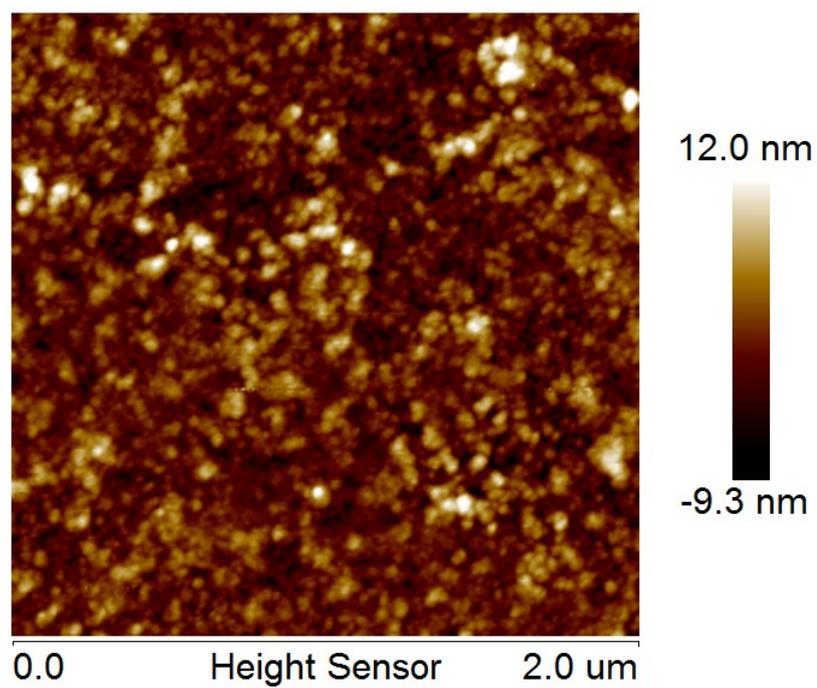
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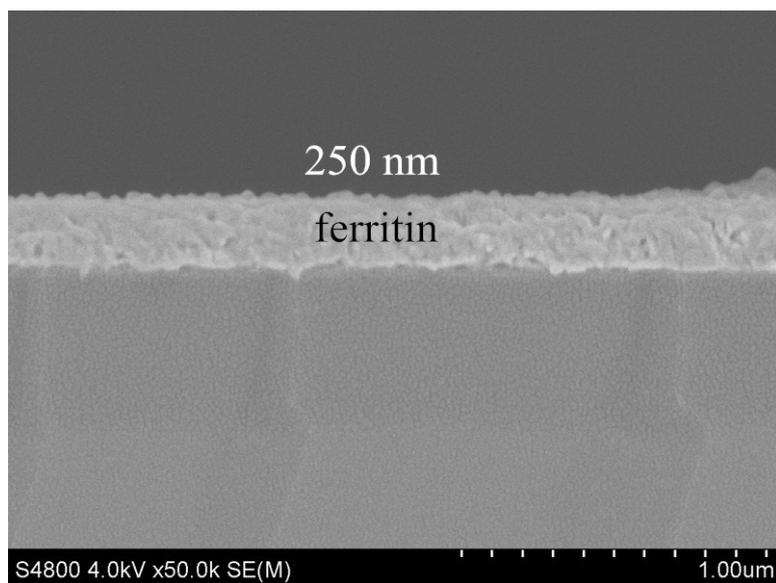
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## 1. Experimental Section

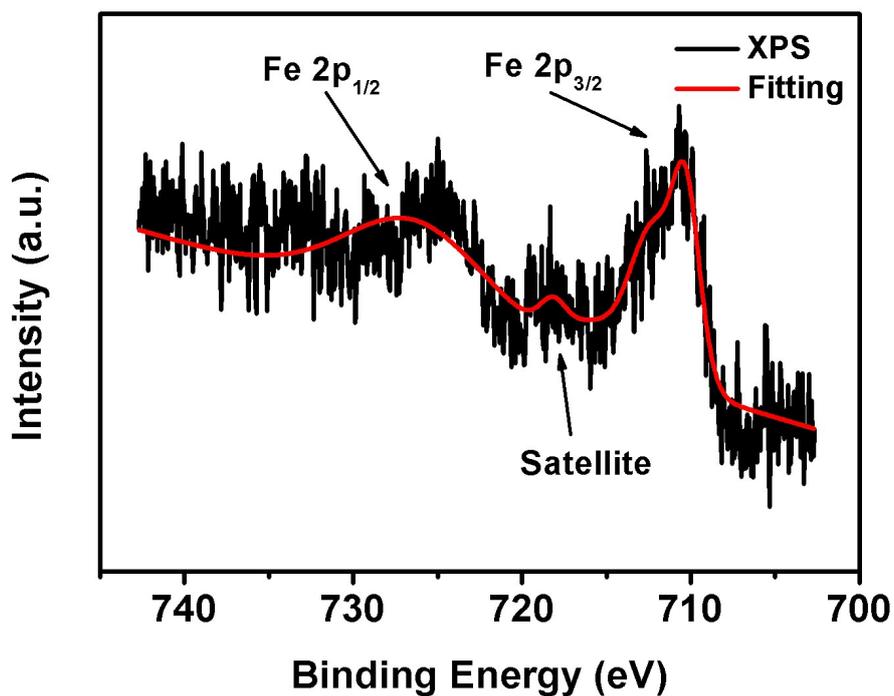
**Device fabrication and characterization:** The resistive switching behaviours of ferritin film were measured in the Pt/ferritin/Pt device. The Pt/Ti/SiO<sub>2</sub>/Si substrate was pre-cleaned for 30 mins in the order of the acetone, ethanol and deionized water in an ultrasonic bath, followed by drying in a nitrogen flow. Ferritin from equine spleen was purchased from Sigma-Aldrich (Shanghai, China) and used directly without further purification. The ferritin layer was then deposited on the Pt/Ti/SiO<sub>2</sub>/Si substrate by employing spin coater at a rotational speed of 500 rpm for 10 s and then at 1000 rpm for 60 s, followed by drying and annealing in a vacuum oven at 70 °C for 2h. The Pt electrode of ~100 nm thickness was deposited on the ferritin layer by radio frequency (RF) magnetron sputtering through a shadow mask. The chemical state of Fe in ferritin film was analyzed by the X-ray photoelectron spectroscopy (XPS, SHIMADZU, AXIS ULTRA DLD). The cross-sectional image of the ferritin film was visualized with a Hitachi S-4800 field-emission scanning electron microscope. The surface topography and the electrical characteristics of ferritin film were investigated by atomic force microscopic (AFM) and conductive atomic force microscopic (C-AFM) technique, respectively, utilizing a Bruker Dimension Icon scanning probe microscope. The current-voltage (I-V) characteristics were recorded in dc sweep or voltage list (equivalent to pulse) mode by Keithley 4200 semiconductor characterization system with a Lakeshore probe station under an ambient atmosphere.



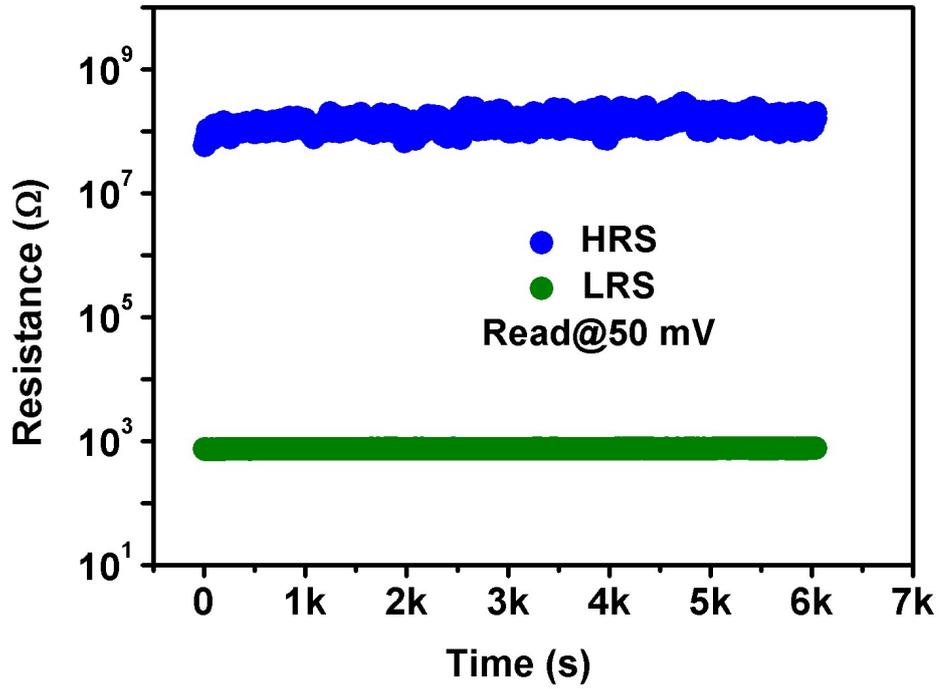
**Figure S1.** AFM image of the ferritin film with a scanning size of  $2 \times 2 \mu\text{m}^2$ . The rms roughness is 2.92 nm.



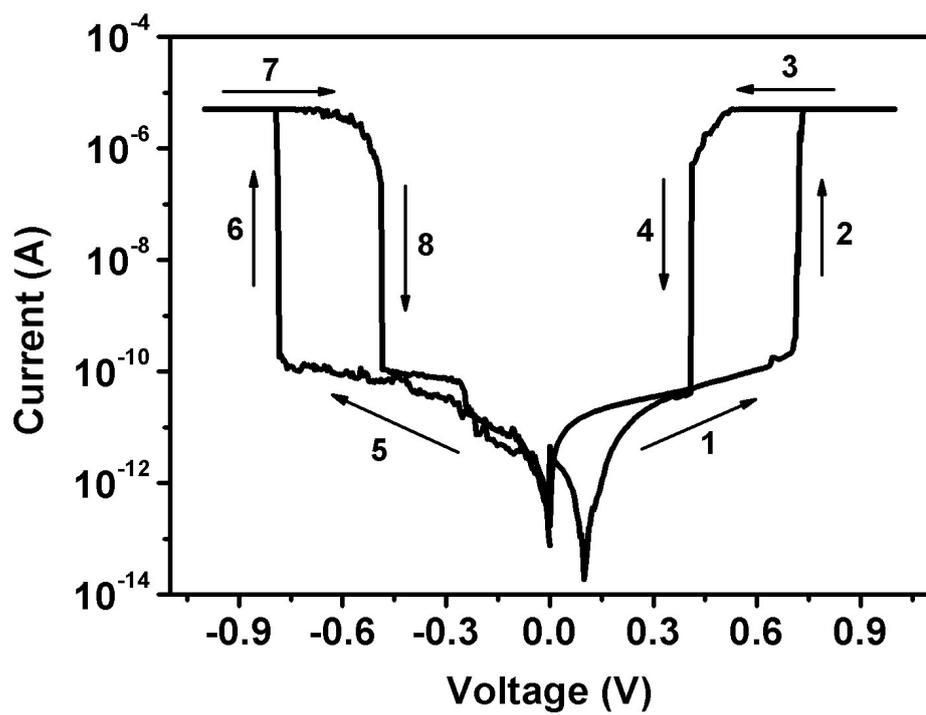
**Figure S2.** Cross-sectional scanning electron microscopic image of the ferritin film.



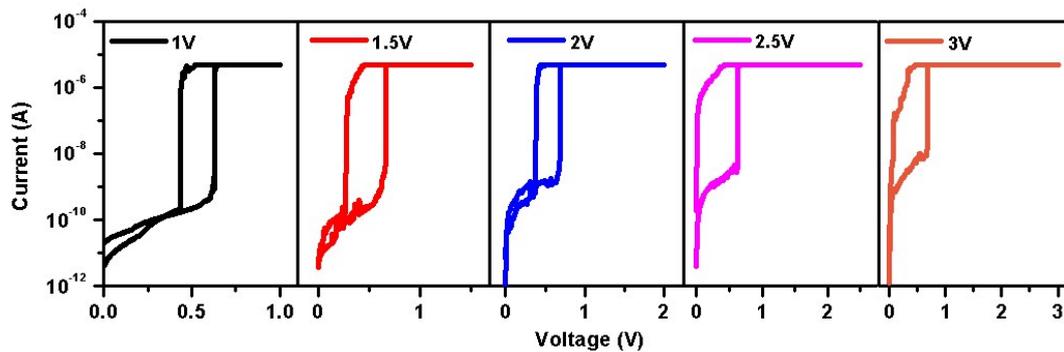
**Figure S3.** The XPS spectrum of Fe 2p obtained from the surface of ferritin film. The binding energies of Fe 2p<sub>3/2</sub> and Fe 2p<sub>1/2</sub> are 710.5 eV and 727.1 eV, respectively. The satellite peak is 718.2 eV, which is located ~8 eV higher than Fe 2p<sub>3/2</sub> peak, indicating the Fe ions contained in ferritin film are trivalent ions<sup>1</sup>.



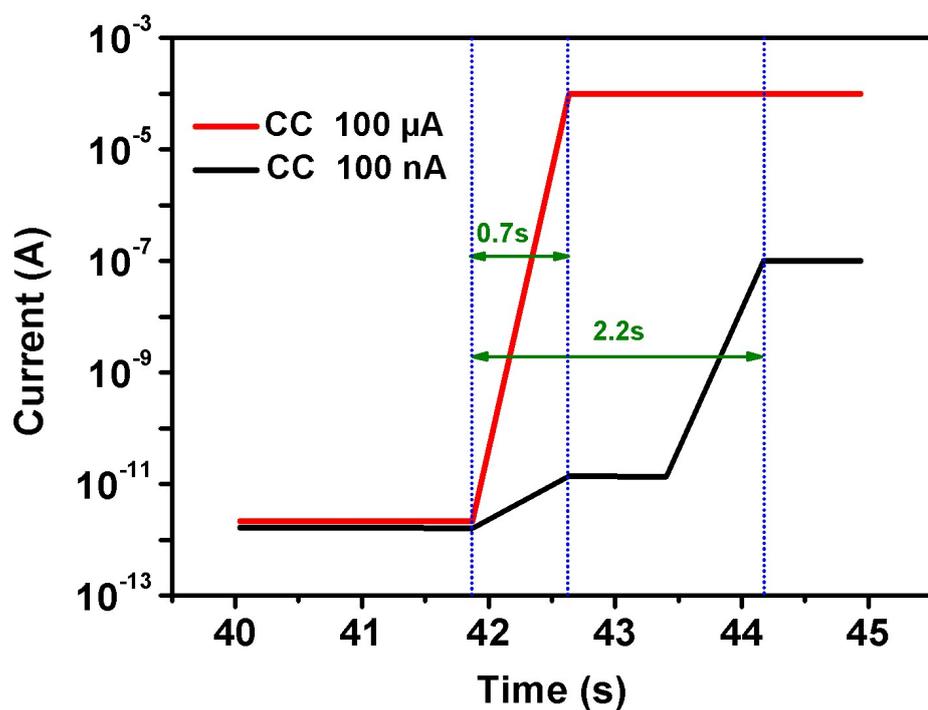
**Figure S4.** Retention characteristics of the Pt/ferritin/Pt device measured with the CC of  $100 \mu\text{A}$ .



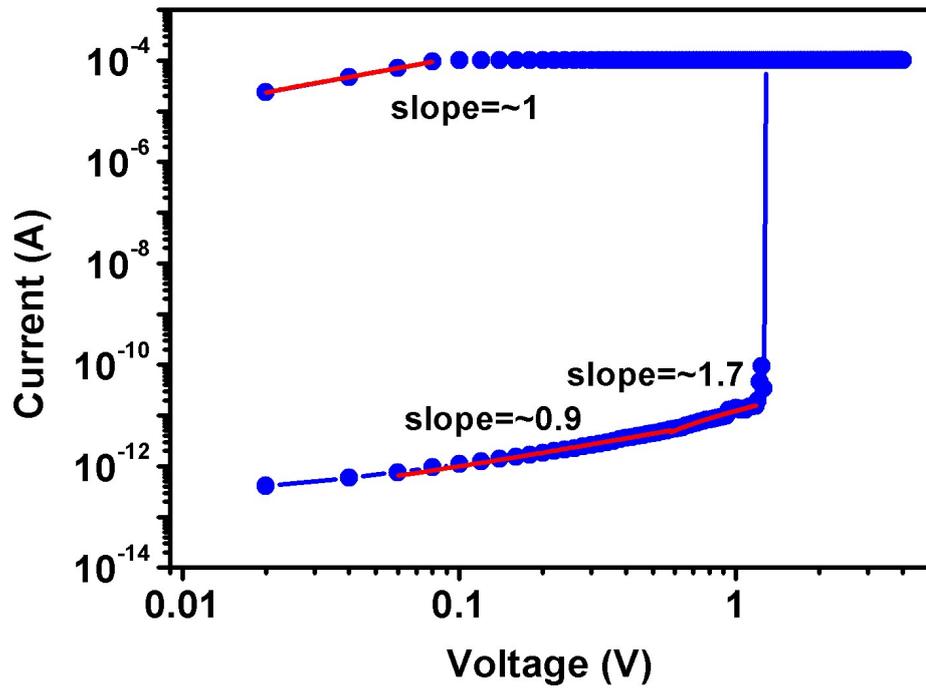
**Figure S5.** The symmetric threshold switching behaviour with the CC of  $5 \mu\text{A}$ .



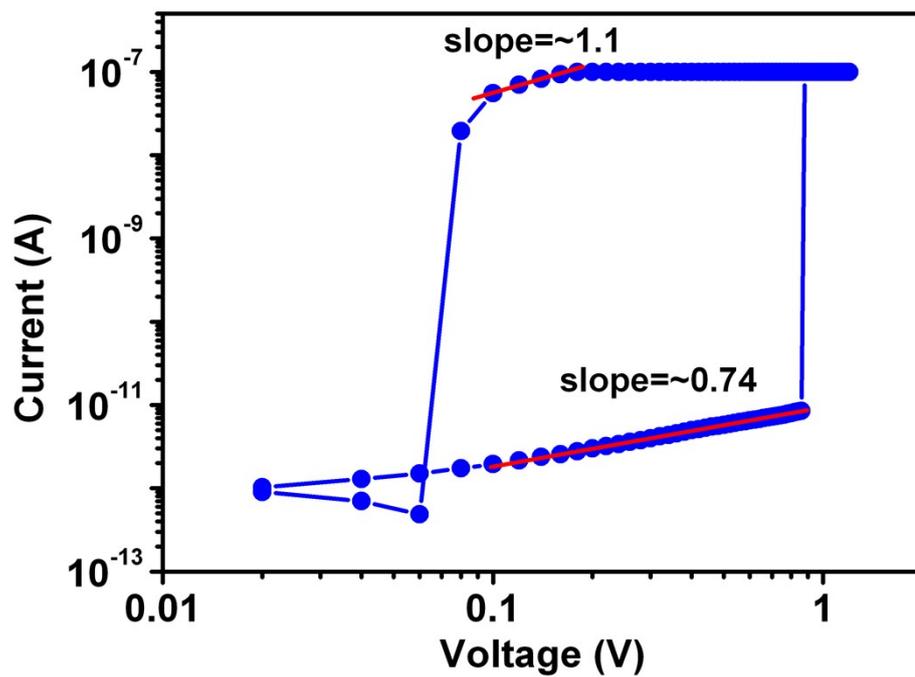
**Figure S6.** The volatile threshold switching of the device measured at different sweeping voltages of 1 V, 1.5 V, 2 V, 2.5 V and 3 V with the fixed CC of 5  $\mu$ A, respectively.



**Figure S7.** The responding current of a stimulating pulse with the CCs of 100  $\mu$ A and 100 nA. The data comes from a part (X-axis from 40 s to 45 s) of Fig. 3b. It shows that the response time is 0.7 s with CC of 100  $\mu$ A and 2.2 s with CC of 100 nA, respectively.

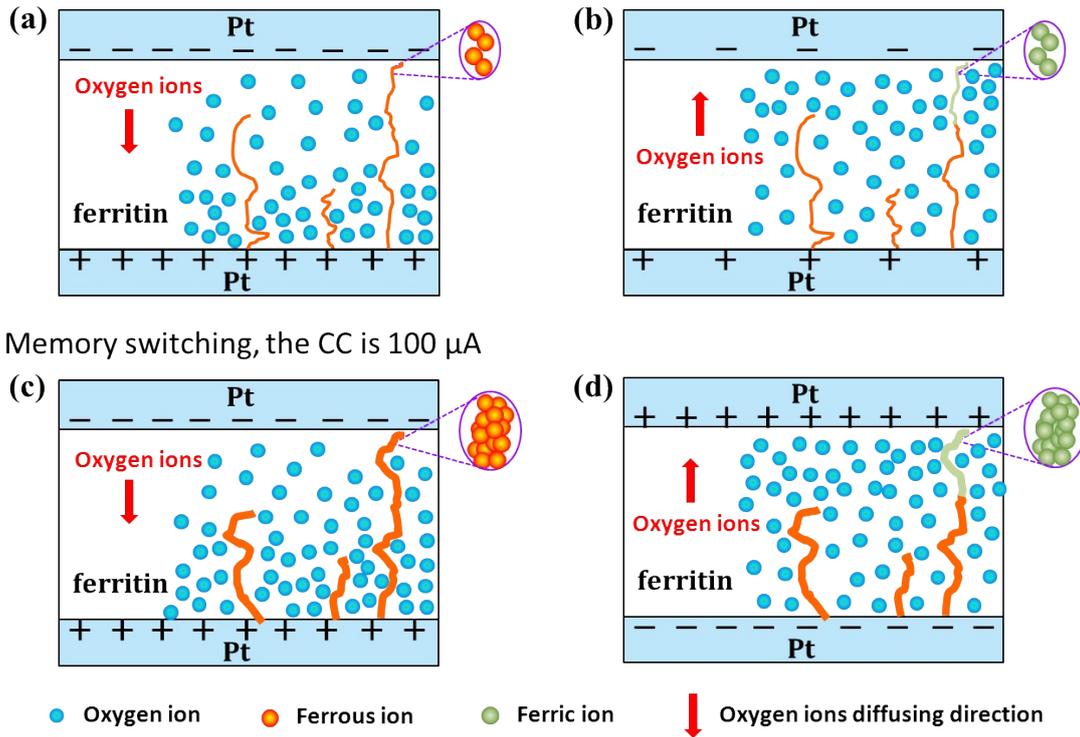


**Figure S8.** Experimental and fitted  $I$ - $V$  characteristics of ferritin film in non-volatile LRS and HRS states. The curve is plotted in log-log scale.



**Figure S9.** Experimental and fitted  $I$ - $V$  characteristics of ferritin film in volatile LRS and HRS states. The curves are plotted in log-log scale.

Threshold switching, the CC is less than  $5 \mu\text{A}$



**Figure S10.** Schematic illustration of the conductive filament formation and rupture of the ferritin-based memristor in LRS and HRS for (a,b) threshold switching, and (c,d) memory switching, respectively.

To further clarify the mechanism of the resistive switching effect of the ferritin-based memristor, the schematic illustration of resistive switching mechanism is shown in Fig. S10. Based on our findings, the device exhibits threshold switching when the preset CC is low due to the thin and unstable conductive filament formation at a sufficient voltage in LRS and easy rupture at a low voltage in HRS (Fig. S10a and 10b); the device exhibits memory switching when the preset CC is high enough due to the strong and stable conductive filament formation at a sufficient voltage in LRS and rupture at a reverse voltage in HRS (Fig. S10c and d). According to previous works, when external voltage is applied to the ferritin-based device, a solid state electrochemical reaction occurs in the hydrous ferric oxide of ferritin<sup>2,3</sup>. And ferric ion can be reduced to ferrous ion through electrochemical reaction. Since the monoatomic ferrous ion drifts more easily than ferric ion and the ferrous region has better conductivity than ferric region<sup>3</sup>, the conductive filament consists of ferrous ions and rupture of conductive filament is caused by oxidation of ferrous ions to ferric ions<sup>4</sup>.

## 2. References

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