Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Ferroelectric resistance switching in Pt/Fe/$BiFeO_3$/$SrRuO_3$/$SrTiO_3$ heterostructures

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S1. KPFM patterns

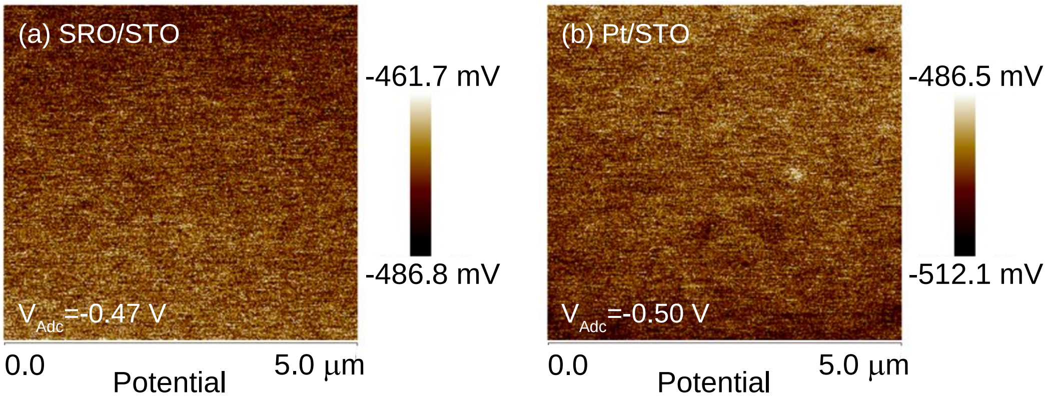

Fig. S1 KPFM patterns of (a) the SRO/STO and (b) the Pt/STO heterostructures in a 5×5 μm^2 area.

Fig. S1 shows the KPFM patterns of the SRO/STO and the Pt/STO heterostructures. The calculated work function φ can be written as $\varphi = \varphi_{tip} - eV_{Adc}$, where φ_{tip} (4.81 eV) is the work function of the tip and V_{Adc} is the average surface potential of the sample. In Fig. S1, the V_{Adc} of the SRO and the Pt layers are –0.47 and –0.50 V, respectively. Thus, the calculated work function of the SRO and the Pt are 5.28 and 5.31 eV. The calculated work function of the Pt is consistent with the ideal value (5.3 eV), but the calculated work function of the SRO is more than the ideal value (5.2 eV),[1] which is related to the quality of the SRO layer.

S2. *I–V* curves at different temperatures

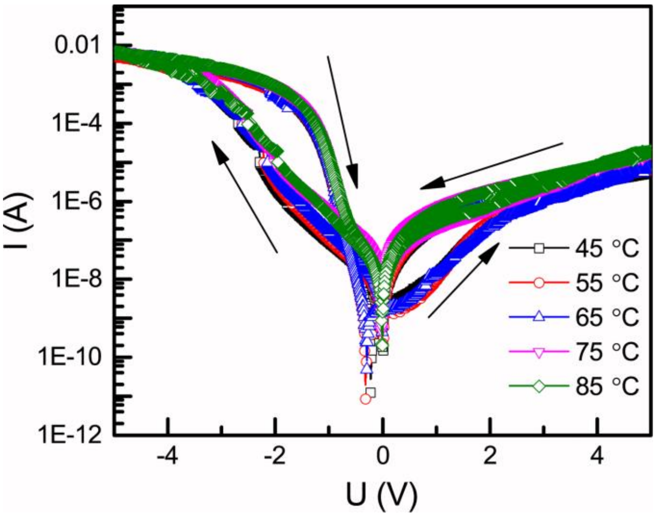

Fig. S2 *I–V* curves for the Pt/Fe/BFO/SRO heterostructure at 45, 55, 65, 75 and 85 ℃.

In this work, the temperature is controlled by the thermostatic heater HP-2525. Firstly, the Pt/Fe/BFO/SRO heterostructure is placed on the thermostatic heater HP-2525. Then, we set the temperature by the thermostatic heater HP-2525. When the temperature is stable for 10 minutes, the *I–V* curves of the Pt/Fe/BFO/SRO heterostructure are measured. Fig. S2 shows the *I–V* curves of the Pt/Fe/BFO/SRO heterostructure at different temperatures. The obvious bipolar RS behaviors are observed, demonstrating that the electrical property of the Pt/Fe/BFO/SRO heterostructure around room temperature is stable. When the temperature is higher than 75 ℃, an obvious signal vibration is observed in Fig. S2. Since the resistance in the Pt/Fe/BFO/SRO heterostructures is related to the ferroelectric polarization, the signal vibration may be resulted from the influence of the temperature on the ferroelectric polarization. With the increase of the temperature, the polarization

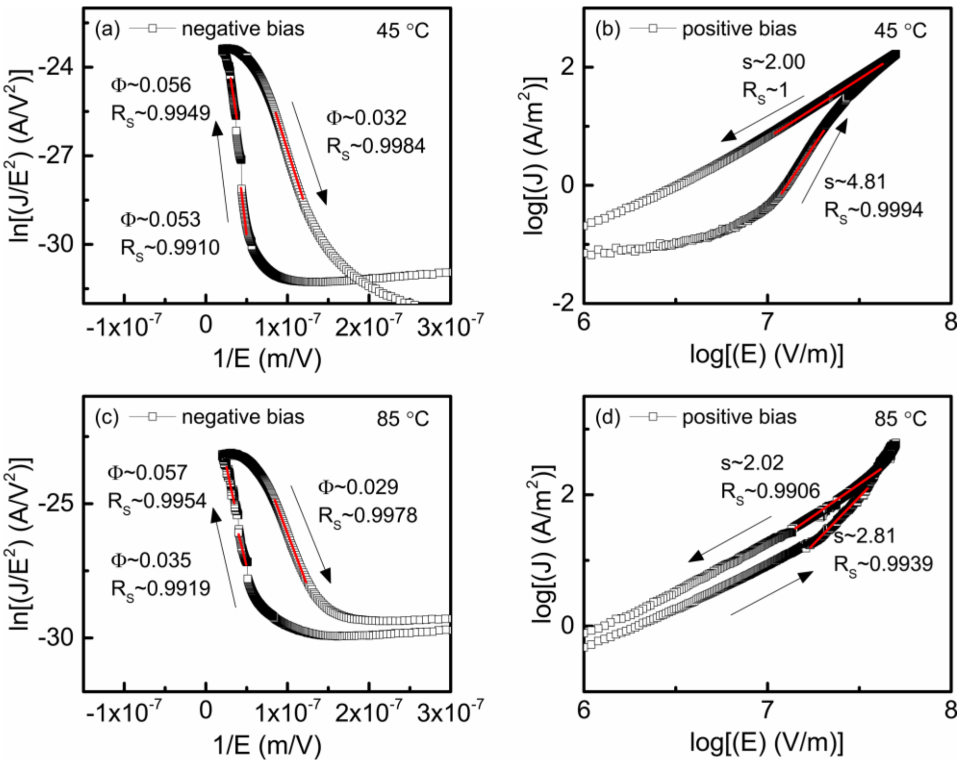

Fig. S3 (a) ln(J/E^2)–1/E and (b) log(J)–log(E) plots for the Pt/Fe/BFO/SRO heterostructures at 45 ℃. (c) ln(J/E^2)–1/E and (d) log(J)–log(E) plots for the Pt/Fe/BFO/SRO heterostructures at 85 ℃. The s represents the slope of the curve. The Φ is the barrier height. The R_s is the R-square value of the fitting result.

switching becomes easier.[2] Moreover, for the same applied voltage, the current at high temperature is more than the current at low temperature. Thus, with the increase of the temperature, the injected charges accumulate easily at the electrode/BFO interface, and induce the pinned ferroelectric domains at the electrode/BFO interface.[3] Further, the pinned ferroelectric domains form the channel, leading to the signal vibration.

In order to further investigate the conductivity mechanism, the I–V curves of the Pt/Fe/BFO/SRO heterostructure at 45 and 85 ℃ are fitted, as shown in Fig. S3 According to the fitting results, the conductivity mechanisms are the interface-limited

FN tunneling mechanism in the negative *V* and the SCLC mechanism in the positive *V*. Therefore, the temperature has no influence on the conductivity mechanisms of the Pt/Fe/BFO/SRO heterostructure.

S3. Retentivity testing

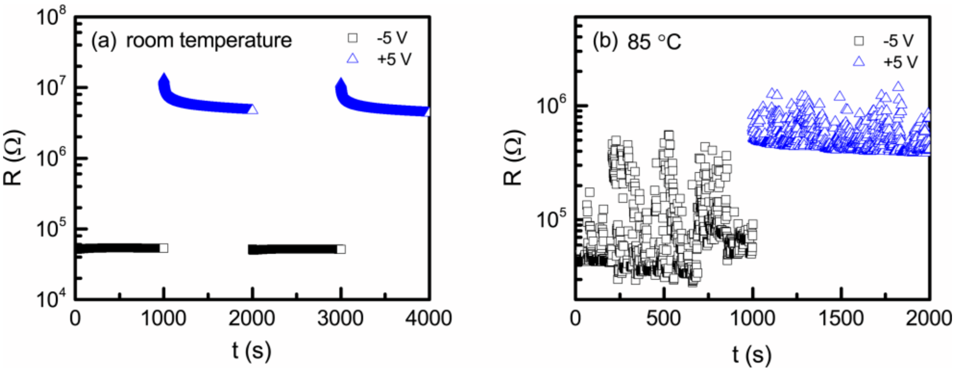

Fig. S4 Retentivity of the HRS and the LRS for the Pt/Fe/BFO/SRO heterostructure after applying +5 and –5 V pulse voltages at (a) room temperature and (b) 85 ℃.

Fig. S4 shows the retentivity of the HRS and the LRS for the Pt/Fe/BFO/SRO heterostructure at room temperature. Those results demonstrate that the resistance degradation for HRS is intrinsic and may be related to the oxygen vacancies migration. While the Pt/Fe/BFO/SRO heterostructure is poling by the positive pulse voltage, the positive polarization charges and the oxygen vacancies are migrating toward the BFO/SRO interface. When the positive pulse voltage is removed and the read voltage is applied, the oxygen vacancies are gradually far from the BFO/SRO interface due to the electrostatic repulsion interaction of the positive polarization charges and the impact of the read voltage. Since the positive polarization charges and the oxygen vacancies lead to the increase of the depletion layer width, the decrease of the oxygen vacancies around the BFO/SRO interface causes the decrease of the depletion layer width, which induces the resistance degradation for the HRS. Fig. S4(b) shows the retentivity of the HRS and the LRS at 85 ℃. The obvious signal vibration is observed, which may be resulted from the influence of the temperature on the ferroelectric polarization. Although the temperature affects the electrical property of the Pt/Fe/BFO/SRO heterostructure, the ratio of the HRS/LRS is ~6.1 is still observed in Fig. S4(b).

S4. *I–V* curves with different applied voltages

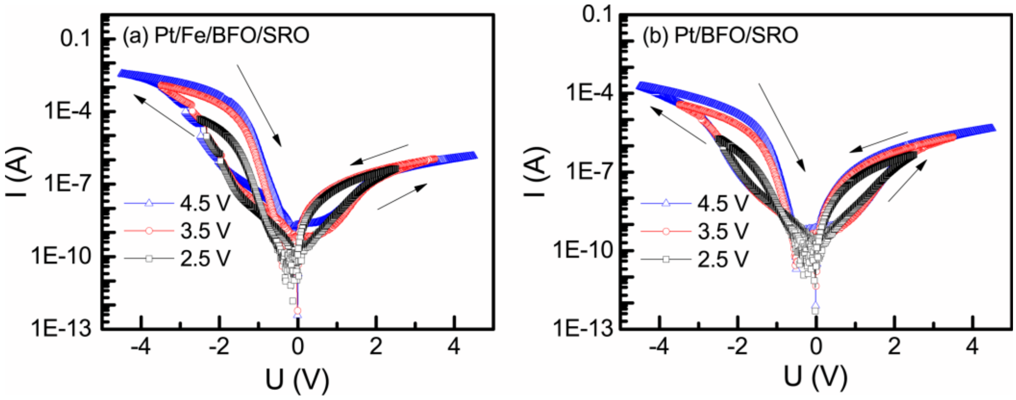

Fig. S5 *I–V* curves for (a) the Pt/Fe/BFO/SRO and (b) the Pt/BFO/SRO heterostructures. The applied voltage sweeps along $0 \rightarrow -V_{max} \rightarrow 0 \rightarrow V_{max} \rightarrow 0$. The V_{max} is 2.5, 3.5, 4.5 V, and the step is 10 mV. The arrows represent the sweep direction of the applied voltage.

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2. J. Wu and J. Wang, J. Appl. Phys., 2010, 108, 094107.
3. X. Zou, L. You, W. Chen, H. Ding, D. Wu, T. Wu, L. Chen and J. Wang, ACS NANO, 2012, 6, 8997–9004.