Supplementary Information

Learning processes modulated by the interface effects in a Ti/conducting polymer/Ti resistive switching cell

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Fig. S1. I-V properties of Ti/PEDOT:PSS/Ti junctions depending on the thickness of PEDOT:PSS. The thickness of the device was (a) 100 nm, (b) 85 nm, (c) 70 nm and (d) 55 nm. The baking time of PEDOT:PSS was 20 min. The diameter of the device was 300 μm.

The thickness of PEDOT:PSS was changed and its influence to the I-V properties can be shown in Fig. S1. Logarithmic scale was used for Y-axis for the convenience to see the overall variation trends. The first scans were plotted using bold green line. The red arrows indicate the scanning directions. Figure S1(a) was re-plotted from Fig. 2. One can easily find that the features of Figs. S1(b), S1(c) and S1(d) resemble Fig. S1(a), indicating the memory phenomena were not influenced qualitatively by the thickness of PEDOT:PSS. These results also indicated that the resistive switching should be located at the interfaces.



Fig. S2. The zoomed-in HRTEM images for (a) Ti BE/PEDOT:PSS and (b) PEDOT:PSS/ Ti TE for the pristine device.



Fig. S3. The zoomed-in HRTEM images for (a) the Ti BE/PEDOT:PSS and (b) PEDOT:PSS/the Ti TE for the device with the NDR behaviors appeared in both the first and the second quadrants (as shown in Fig. 3d).

The transition can be observed from crystalline structure to amorphous interface and amorphous polymer layer in the zoomed-in HRTEM images about the interface (shown in Figs. S2 & S3). The layers labelled by the letter '1' are of the Ti electrode in which the crystalline structures with stripe images can be observed. The layers labelled by the letter '2' should be the layers of Ti compound. The stripe images of the crystalline structure disappear and the contrasts become shallower in these layers. The variations of the contrasts suggest that the contents of the metallic Ti are reduced. The layers labelled by the letter '3' are of PEDOT:PSS, which is amorphous totally. The white contrasts in these layers suggest the lack of metallic element, which is also confirmed by Fig. S6.



Fig. S4. I-V properties of Ti/PEDOT:PSS/Ti junctions obtained from five voltage scans between (a) \pm 0.5 V, (b) \pm 1 V, (c) \pm 1.5 V and (d) \pm 2 V. The vertical axis is logarithmic in the main figure but linear in the inset; '1' and '2' indicate scanning process number as in Fig. 3.

The device is stable under low bias, such as ± 0.5 V. Its state is modified when the bias values are higher than 1 V.



Fig. S5. I-V curves obtained from 5 consecutive scans of ± 0.5 V after initial scans. The initial scans are (a) 5 scans of ± 2.0 V, and (b) 5 scans of ± 3.0 V.

The electrical characteristics after strong stimulation were tested using ± 0.5 V scans. It is seen that the 5 consecutive scans using ± 0.5 V are nearly superposed after 5 scans at ± 2.0 V, while there is very low divergence from each other after 5 scans at ± 3.0 V. This result demonstrates that even if the state of the device is modified by a strong stimulation, the modified state could be read out using small voltage.



Fig. S6. cross-sectional TEM image of the device having NDR in both first and second quadrants in I-V curves. The variations in the element concentrations of various elements obtained using EDX are also shown.

After electric manipulation, oxygen and sulfur elements accumulated at the interface near the electrode, which is confirmed by EDX spectra.