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Evolution of resistive switching with switching cycles in Pt/$BiFeO_3$/NiO/Pt memory cell

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Abstract. $BiFeO_3$/NiO heterostructure is prepared using chemical solution deposition method. The X-ray result indicates that a perovskite structure has been formed in this thin film, and the images of cross-section and surface show a good thin film quality. The current-voltage curves with switching cycles have also been measured in Pt/$BiFeO_3$/NiO/Pt structure, and two steps of resistive switching can be clearly observed. According to the analysis of resistances and switching voltages in these two steps, the evolution of resistive switching with switching cycles can be demonstrated by conducting filament mechanism.

1. Introduction

Multiferroic bismuth ferrite (BFO), can be found to show prominent characteristics in area of the ferroelectricity and the antiferromagnetism[1-3]. In Recent years, BFO is observed to demonstrate the better resistive switching (RS) properties in the application of resistive random access memory, and attracted broad attentions as promising solid state memory[4-8]. Kawachi et al. [5] investigated the effects of electric fields perpendicular to thec-axis of the trigonal cell in BFO single crystals through magnetization and resistance measurements, attributed to the reorientation of the magnetoelectric domains. Liu et al. [6] also studied the resistive switching properties in the bismuth ferrite thin films with different electrodes, which can be rationalized in terms of conducting filaments and the change of the Schottky barrier at the film/electrode interfaces due to ferroelectric polarization. He et al. [7] observed the direct evidence of purely interfacial effects on the resistive switching in Au/BFO/Nd:$SrTiO_3$(001) Schottky junctions through reducing the thickness of BFO interlayer, and attributed it to the ferroelectric polarization modulation of the barrier and the depletion width of p-n junction which is formed at the BFO/NSTO interface. Kumari et al. [8] fabricated Ag/BFO/FTO resistive random access memory device, and found that the device was robust against environmental conditions, exhibiting good reproducibility, retention and endurance. From the above, it can be seen that the resistive switching behaviors in BFO films are quite complicated. For the practical applications of BFO memory cell, the research of RS mechanism is still a hard work. In this work, the evolution of resistive switching with switching cycles has been studied in Pt/BFO/NiO/Pt structure, and two steps of resistive switching behavior can be clearly observed, which is ascribed to the formation and rupture of conducting filament varied from the BFO layer to the NiO layer.

2. Experimental

BFO/NiO thin film was grown on Pt/Ti/SiO_2/Si substrate by chemical solution deposition. The solution of BFO was compounded with $Fe(NO_3)_3\cdot 9H_2O$ and $Bi(NO_3)_3\cdot 5H_2O$, and the solution of

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NiO was prepared by $NiC_4H_6O_4{\cdot}4H_2O$. All the stoichiometric solutions were spin-coated and then dried, and finally annealed at the temperature of 550 °C. The structure of film was characterized by X-ray diffraction. The surface and cross-section image were characterized by a scanning probe microscope and a scanning electron microscopy, respectively. The current-voltage curves in Pt/BFO/NiO/Pt structure were measured by a semiconductor characterization system.

3. Results and Discussion

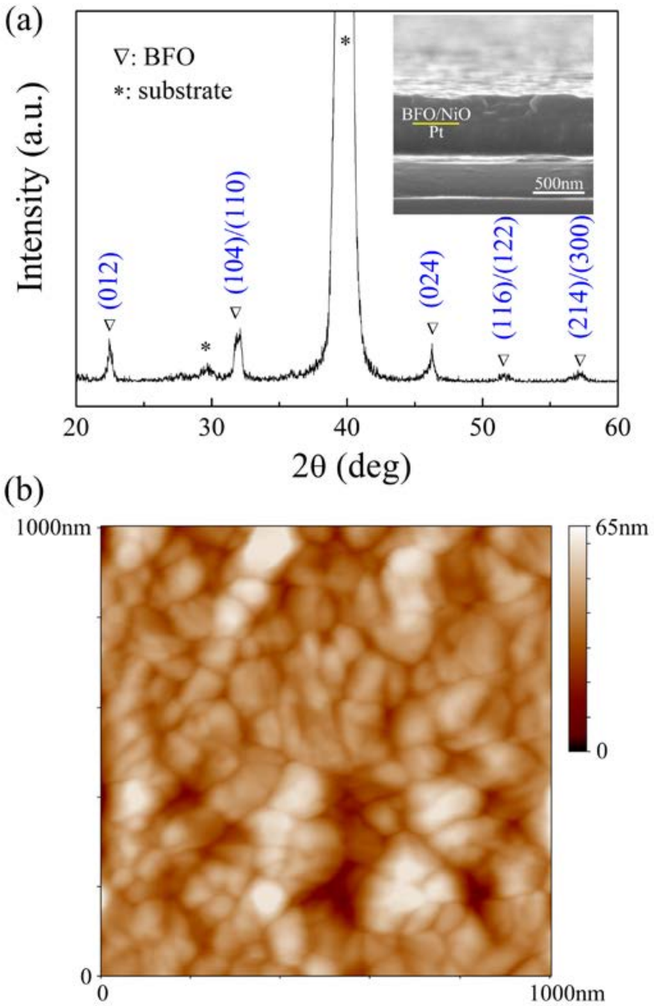

Figure 1. (a) X-ray diffraction patterns and (b) Scanning probe microscope morphology of BFO/NiO thin film. The inset of (a) displays the cross-section image.

Fig. 1(a) demonstrates the X-ray diffraction patterns of BFO/NiO film which is grown on Pt/Ti/SiO_2/Si substrate. The results of the diffraction peaks show the perovskite BFO structure with no other secondary phases. The cross-section image characterized by scanning electron microscopy in the inset of Fig. 1(a), revealing that the BFO/NiO film has been better grown on the Pt substrate. Additionally, the corresponding surface image of scanning probe microscope with a scanning area of $1000 \times 1000\,\mathrm{nm}^2$ is also characterized in Fig. 1(b), showing a good thin film quality.

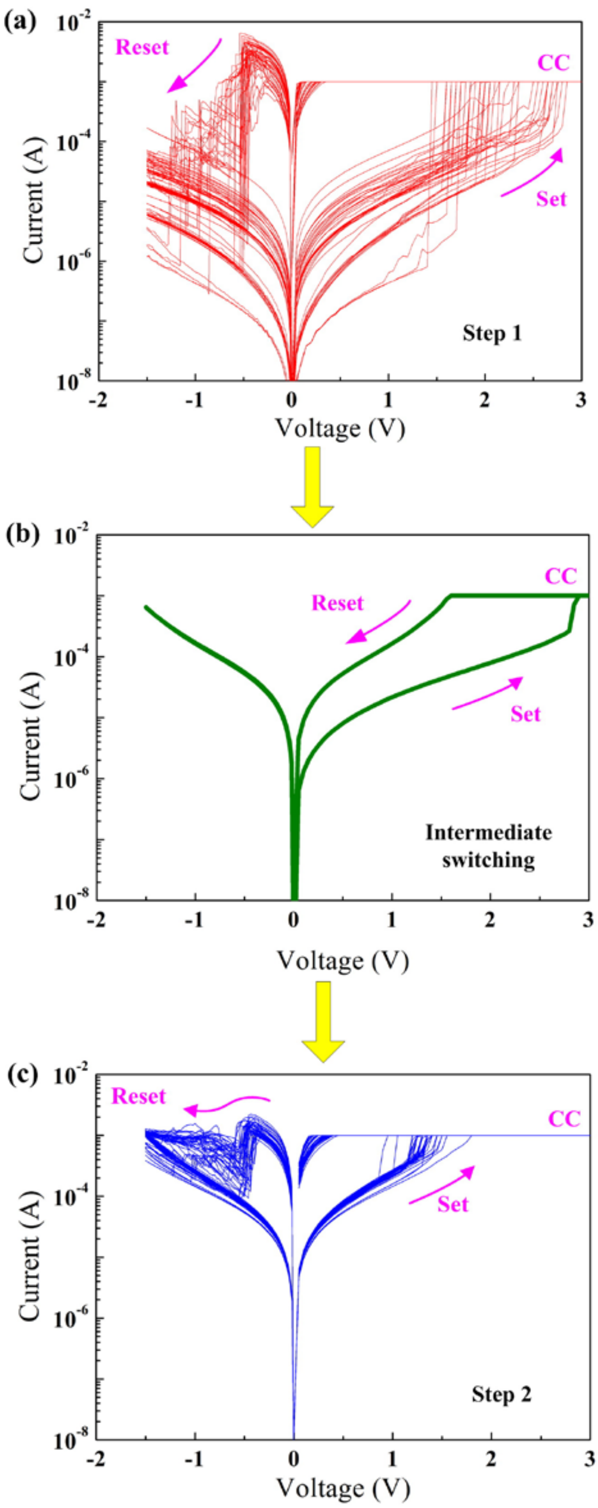

Figure 2. Current-voltage curves of resistance switching. (a) Step 1; (b) Intermediate switching; (c) Step 2.

After a forming process at the high voltage, the current-voltage curves of RS in Pt/BFO/NiO/Pt memory cell were measured for about 90 switching cycles. Based on the analysis of these current-voltage curves, the evolution of RS has been revealed in Fig. 2. Initially, the unstable current-voltage curves for 50 switching cycles can be observed as shown in Fig. 2(a). When the positive voltage increases, the set process where the resistance changes from a high resistive state (HRS) to a low resistive state (LRS) happens in a large range of applied voltage; when the negative voltage is applied, the reset process where the LRS switches back to the HRS occurs in an appropriate voltage range. This unstable state of resistive switching can be described as Step 1. Afterwards, an intermediate switching could be happened which is shown in Fig. 2(b), when scanning the positive voltage from zero to a high value, the resistance varies from one HRS to LRS; however, when scanning the voltage back to zero, the resistance changes from LRS to another HRS, in which the resistance is lower than that of the first one. After the intermediate switching, Step 1 of RS in Pt/BFO/NiO/Pt structure could be developed into Step 2. From Fig. 2(c), it can be seen that the current-voltage properties are greatly stabilized and reproduced in Step 2.

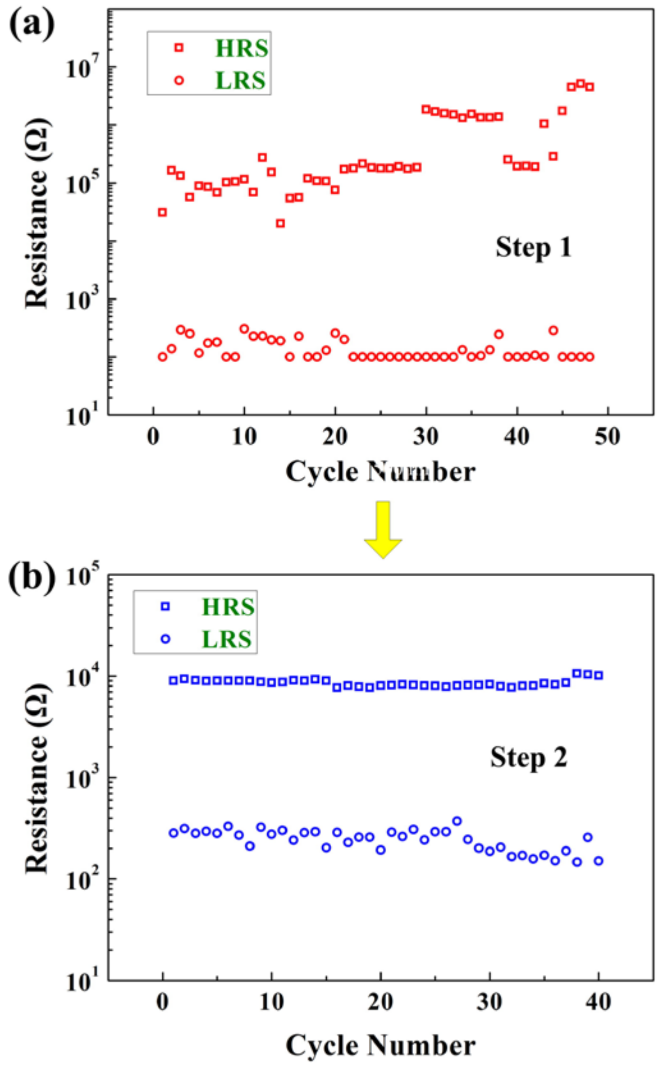

Figure 3. Resistance variation with switching cycles for HRS and LRS. (a) Step 1; (b) Step 2.

To further study the RS characteristics in Step 1 and Step 2, the resistance variation with switching cycles in HRS and LRS is shown in Fig. 3(a) and (b) respectively. In Step 1, the resistance of HRS is found to distribute within more than 100 times, and the resistance of LRS shows no evident dispersion. Compared with Step 1, the resistance of HRS in Step 2 is much smaller with little dispersion, but not much change in LRS, indicating that the resistance characteristic of RS can be greatly stabilized when Step 1 is developed into Step 2. Moreover, the switching voltage distributions in Step 1 and Step 2 are also shown in Fig. 4(a) and (b). In Step 1, the distribution of set voltage is from 1.5 to 2.5 V, and the reset voltage is around -0.5 V. Compared with Step 1, the distribution of set voltage from 1 to 1.5 V in Step 2 is quite narrow, but the reset voltage is almost the same. Obviously, the switching voltages of Step 2 also exhibit much better stability than that of Step 1.

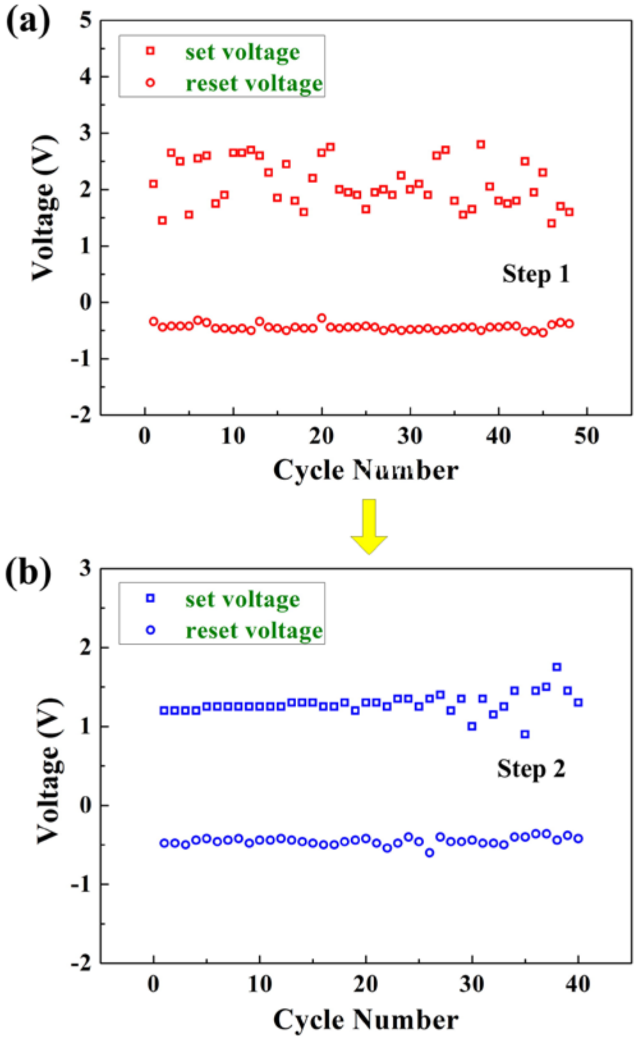

Figure 4. Distribution of the set and reset voltages. (a) Step 1; (b) Step 2.

Based on the above mentioned results, the evolution of RS in Pt/BFO/NiO/Pt structure is sketched in Fig. 5. In view of conductive filament mechanism [9], when applied the positive voltage, the ionized oxygen vacancies (OVs) can move from the anode to the cathode. These OVs capture the electrons injected from the cathode and become neutral, leading to the formation of filamentary conducting channel. Once the metallically conducting channel is created, the set and reset processes could be occurred at the thick layer of BFO film shown between Fig. 5(a) and (b), and the resistance as well as the set voltage are random and inhomogeneous, which is described as Step 1. After successive switching cycles of Step 1, the intermediate switching happens. In this switching, the set process occurs at the step-up voltage depicted from Fig. 5(a) to (b); while the reset process happens at the step-down voltage depicted from Fig. 5(b) to (c). Considering of the low resistivity of NiO film, the reset process could happen at the NiO film from the experimental data. The reason for this behavior is still unclear, probably owing to the Joule-heating effect according to the literatures [10,11].

After the intermediate switching, the set and reset processes may easily occur at the NiO film shown between Fig. 5(b) and (c), and the filamentary conducting channel can be locally confined better, resulting in the stable switching characteristics such as the resistance and the set voltage, which is described as Step 2. It is worth mentioning that, assuming the resistance of metallic-like filamentary channel is

$$R = \rho \frac{l}{S} \tag{1}$$

where ρ is the filament resistivity (almost constant in the thin film), l is the filament length (approximately equal the thickness of thin film) and S is the filament area. And then, the power per unit volume for the rupture of filamentary conducting channel due to the Joule heating can be shown as follows [12]

$$E = \frac{1}{Sl}\frac{V^2}{R} = \frac{1}{\rho l^2}V^2 \tag{2}$$

where V is the reset voltage. From equation (2), it is found that the reset voltage could be independent of resistive switching cycles, which can be inferred from the observation of Fig. 4. However, more investigations still need to be carried out based on large quantities of experimental data.

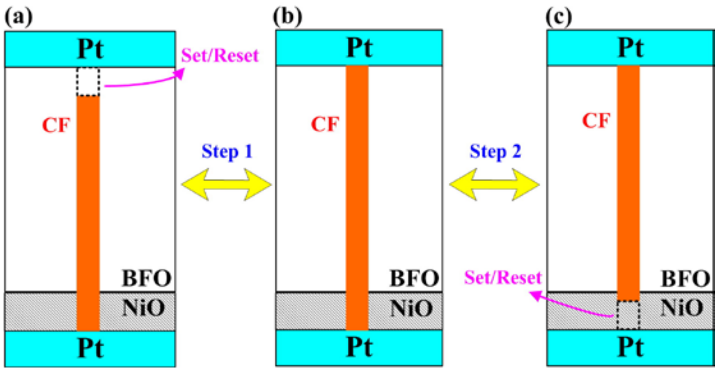

Figure 5. Sketches of evolution of resistive switching characteristic with switching cycles.

4. Conclusion

Pt/$BiFeO_3$/NiO/Pt memory cell is prepared using chemical solution deposition method. According to the studies of X-ray diffraction and surface properties, a good thin film quality of $BiFeO_3$/NiO heterostructure can be demonstrated. In addition, the evolution of resistive switching has been investigated by measuring the current-voltage switching curves, which is divided into the two steps: The resistive switching in Step 1 can be random and inhomogeneous; after the intermediate switching, the resistive switching in Step 2 becomes stable and uniform. This behavior could be ascribed to the formation and the rupture of filamentary conducting channel varied from the BFO thick layer to the NiO thin layer.

Acknowledgments

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