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Abnormal resistance switching behaviours of NiO thin films: possible occurrence of both formation and rupturing of conducting channels

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Abstract
We report a detailed study on the abnormal resistance switching behaviours observed in NiO thin films which show unipolar resistance switching phenomena. During the RESET process, in which the NiO film changed from a low resistance state to a high resistance state, we sometimes observed that the resistance became smaller than the initial value. We simulated the resistance switching by using a random circuit breaker network model. We found that local conducting channels could be formed as well as ruptured during the RESET process, which result in the occurrence of such abnormal switching behaviours.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Resistance switching phenomena have recently regained intensive research efforts due to their potential for realizing high-density resistance random access memory (RRAM) devices [1]. There have been many reports that resistance values in several binary transition metal oxides [2–5] and perovskite oxides [6–10] could be switched between a high resistance (HR) state and a low resistance (LR) state by applying an external electrical voltage.

Among these resistance switching materials, NiO has been studied extensively because of its high HR/LR ratio and its ease of integration with the available semiconductor process for creating high-density RRAM devices [11, 12]. NiO thin films normally showed a unipolar resistance switching characteristic, where the resistance could be switched at two values of applied voltage with the same polarity. The unipolar resistance switching in NiO has been described by the formation and rupture of localized metallic conducting filaments based on the observance of conducting channels from conductive atomic force microscope and high resolution transmission electron microscope studies [13–15]. Generally, the SET process, which corresponds to the change from the HR state to the LR state, has been described by the formation of conducting channels, whereas the RESET process, which is the change from the LR state to the HR state, has been described in terms of the rupturing of conducting channels. Detailed behaviours involved in the switching process, however, have not been well understood yet.

Recently, a percolation model based on a random circuit breaker (RCB) network has been reported to explain the unipolar resistance switching behaviour [16]. A switching

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medium composed of circuit breakers was proposed in the model, and the circuit breakers can be switched deterministically between bistable resistance states depending on the voltage applied to each component.

In this paper, we report abnormal resistance switching behaviours during the RESET processes of NiO thin films, which cannot be explained simply by the rupturing of conducting channels alone. The RCB network model was employed to simulate the resistance switching. Our detailed analysis found that the observed abnormal switching behaviours occur when the formation as well as rupturing process of conducting channels happen at the same time during the RESET process.

2. Experiment

We fabricated polycrystalline NiO thin films with a thickness of 200 nm on Pt/Ti/SiO_2/Si substrates using dc magnetron reactive sputtering [11, 14]. The NiO thin films were deposited at 300 °C and 5 mTorr in a gas mixture of Ar and O_2. The partial pressure of O_2 was 5%. We measured the electrical properties of the Pt/NiO/Pt capacitors in both voltage-driven and current-driven modes using an HP/Agilent 4155C Semiconductor Parameter Analyzer. In the voltage-driven mode, the applied voltage across the Pt/NiO/Pt capacitor was swept and the current was measured, and vice versa in the current-driven mode. A compliance current (I_{COMP}) was set to limit the maximum current value for the SET process to prevent complete dielectric breakdown during current–voltage (I–V) measurements. For current-driven measurements, we set a compliance voltage (V_{COMP}) in the RESET process.

3. Results and discussion

(colour online)

We observed different types of resistance switching behaviour during the RESET process. Figure 1 shows the resistance switching behaviours of our NiO films under dc voltage-driven mode. The red (grey) and black symbols correspond to the RESET and SET processes, respectively. In most cases, resistance switching during the RESET process occurred rather sharply, as shown in figure 1(a). However, we also observed random occurrence of an abnormal resistance switching behaviour, where the current value became larger than the onset value, denoted by 'S' in figure 1(b). Among the 50 Pt/NiO/Pt capacitors tested, this type of abnormal switching was observed in near 70% of them. For a Pt/NiO/Pt capacitor, the abnormal switching behaviour could be observed randomly in any RESET process, but not necessarily in every RESET process. This abnormal behaviour is quite surprising when the common understanding of the RESET process is considered. If the RESET process only involves the rupturing of conducting channels, the film resistance should keep increasing, so the anomalous peak in figure 1(b) should not occur. Therefore, the abnormal switching behaviour implies that in addition to rupturing, formation of new conducting channels may also be involved in the RESET process.

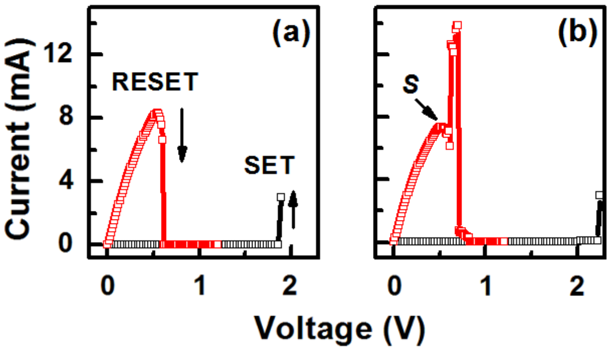

Figure 1. Current–voltage (I–V) characteristics of NiO capacitors during SET and RESET processes for dc voltage-driven I–V sweeps. The black and red (grey) symbols represent the SET and RESET processes, respectively. (a) Typical resistance switching. (b) Abnormal resistance switching with large resistance fluctuations during the RESET process. 'S' indicates the onset of the RESET process where the resistance value begins to change. (Colour online.)

We also observed similar abnormal RESET switching behaviours in current-driven switching measurements. We found a strong dependence of the abnormal switching on the compliance voltage (V_{COMP}). In current-driven mode, the voltage drop (V_{DROP}) across the capacitor is determined by the applied current and the capacitor resistance. The resistance starts to increase after the RESET process starts, and V_{DROP} will increase accordingly till it reaches V_{COMP}. We observed that when V_{COMP} was lower than 1.0 V, the capacitor was always smoothly switched to the HR state with no resistance oscillation. Figure 2(a) shows a typical I–V curve for the RESET process with V_{COMP} of 1.0 V. When V_{COMP} was increased to 2.0 V, we observed the abnormal RESET switching shown in figure 2(b), which exhibits several resistance value changes. Note that, after the first change, the resistance became lower than that of the original LR state while it was still undergoing the RESET process. When V_{COMP} was increased further to 2.5 V, we always observed a series of abnormal switchings before the NiO film experienced a hard-breakdown, as shown in figure 2(c).

The dependence of current-driven RESET behaviour on V_{COMP} could be understood if we compare the values of set voltage (V_{SET}) and V_{COMP}. V_{SET} is the voltage required for the SET process to occur. Note that V_{SET} for most of the capacitors in this experiment is between 1.5 and 2.5 V. With increasing V_{COMP}, V_{DROP} could be close to or larger than V_{SET}, making it possible to form new conducting channels in the NiO film. This is presented by the resistance oscillation in figure 2(b). When V_{COMP} is set to 2.5 V, which is definitely higher than V_{SET}, the capacitor is SET to an LR state with lower resistance as soon as V_{DROP} reaches 2.5 V, so the I–V curve showed the resistance switching back and forth between the HR and LR states, and finally in the breakdown state. The effect of V_{COMP} on the current-driven RESET behaviour implies that the generation of new conducting channels in the voltage-driven RESET process (figure 1(b)) may originate from the voltage distribution inside the NiO thin film.

To better understand the abnormal switching behaviours, we simulated the resistance switching by using the percolation

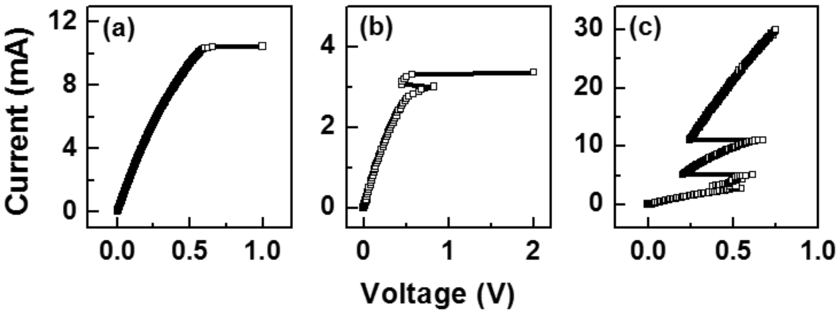

Figure 2. (*a*), (*b*) and (*c*) Experimental I–V curves for RESET processes under current-driven mode with $V_{\mathrm{COMP}} = 1.0$ V, 2.0 V and 2.5 V, respectively.

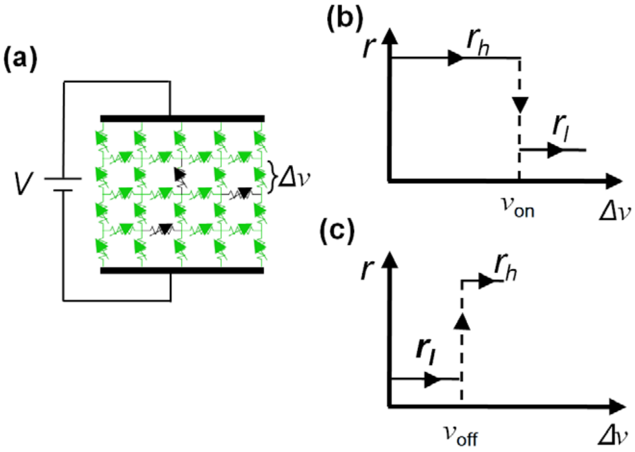

Figure 3. (*a*) Schematic diagram of the RCB network model for a two-dimensional bond percolation. Each bond represents one circuit breaker. The black and green (grey) colours represent the on- and off-states, respectively. The upper and lower black bars represent top and bottom electrodes. (*b*) The switching condition of the circuit breaker's state from off to on. (*c*) The switching condition of the circuit breaker's state from on to off. (Colour online.)

RCB network the model [16], in which formation and rupturing of conducting channels are represented by the turning-on and turning-off operations of circuit breakers. Figure 3(*a*) shows a schematic diagram of the RCB network composed of bistable circuit breakers. The resistance value of each circuit breaker will be either r_{l} for the on-state (i.e. 'connected') or r_{h} for the off-state ('disconnected'). We assumed that the state of each circuit breaker can be switched between two resistance values, i.e. r_{l} and r_{h} depending on its biased voltage, Δv, and values of threshold voltages, v_{off} and v_{on}. As shown schematically in figure 3(*b*), a circuit breaker that is initially in the off-state will be switched to the on-state when $\Delta v > v_{\mathrm{on}}$. Conversely, as shown in figure 3(*c*), a circuit breaker that is initially in the on-state will be switched to the off-state when $\Delta v > v_{\mathrm{off}}$. Note that detailed resistance switching behaviours of the whole RCB network will be determined by the distribution of Δv values across each circuit breaker.

We used two-dimensional square lattices of 150×30 circuit breakers for our simulations, with $r_{\mathrm{h}}/r_{\mathrm{l}} = 1000$ similar to the experimentally observed resistance difference between LR state and HR state. In the pristine network, we *randomly* selected 0.5% of the circuit breakers to be in the on-state, which resulted in the initial distribution of Δv values. In real situation, this could correspond to the nanometre-sized conductive defects exist in the as-grown NiO films [15]. The simulation is performed with the following procedure. For the formation process which generates the conducting percolation cluster, after increasing V from 0 by a given voltage step, we calculated Δv for each circuit breaker and checked the switching conditions shown in figures 3(*b*) and (*c*). If switching occurred in at least one circuit breaker, we reevaluated the Δv distribution and checked the switching conditions again, repeating this iterative process until the network reached a stable state. Then, we increased V by another voltage step and repeated the calculations until the total current through the network, i.e. I, reached I_{COMP}. The resulting configuration should correspond to the LR state, where there will be a percolating cluster of on-state circuit breakers.

The percolating network in the LR state and the resulting I–V curve of the RESET process were found to be dependent on the initial distribution of the Δv values in the pristine state. Figures 4(*a*) and (*b*) show two snapshots of the percolating cluster of on-state circuit breakers in LR states, formed with $I_{\mathrm{COMP}} = 1.0$. Although the on-state breakers in the pristine state were selected with the same number fraction (i.e. 0.5%), the detailed Δv distribution in the resulting percolating cluster could vary as a consequence of the initial *random* selection process. After I reached I_{COMP}, we set V to zero and began to increase it again, which corresponds to the RESET process. The I–V curves in figures 4(*c*) and (*d*) correspond to the RESET processes started from the initial LR states in figures 4(*a*) and (*b*). Note that these simulated I–V curves are quite similar to the experimental I–V curves in figures 1(*a*) and (*b*).

The abnormal switching behaviours observed in the current-driven I–V measurements were also well reproduced in simulation using the RCB network model. Figures 5(*a*)–(*c*) show the I–V curve simulations for current-driven resistance switching with increasing values of V_{COMP}. These simulation results agreed quite closely with the experimental observations shown in figure 2. We also studied the details of snapshots for the abnormal switching, and found that both rupturing and formation of conducting channels could also occur during the current-driven measurements.

We could obtain further insights on the RESET process by studying how circuit breakers were turned on and off. We investigated the snapshots of the RCB network for the values of V, marked with numbers 1–5 in figure 4(*d*). Figure 6 shows the detailed snapshots of the portion of the RCB network enclosed by the thick solid (black) box in figure 4(*b*). No configuration change was observed in other parts of the network during this simulation. Solid (black) and dashed (blue) lines are used in figure 6 to indicate regions where rupture and formation of conducting filaments occur, respectively, to clarify the progressive changes in the RCB network. It is interesting to note that although this simulation result corresponds to a RESET process, both rupturing and formation of conducting channels are clearly observed. Specifically, new conducting channels were generated in the network shown in numbered 3, forming a larger cluster, which results in the sudden increase in current in figure 4(*d*). In number 5, although there are also new conducting channels formed, due to the rupturing of more conducting channels, the resistance of number 5 is much higher than that of number 4. The close agreement between figures 1(*b*) and 4(*d*) and the detailed switching process shown in figure 6 indicate that rupturing and formation of conducting channels are not exclusive during the RESET process. The reason is that the switching of at least one conducting channel can bring about a significant change in the voltage distribution in the NiO thin film, which then leads to an avalanche of formation and rupturing in other nearby conducting channels.

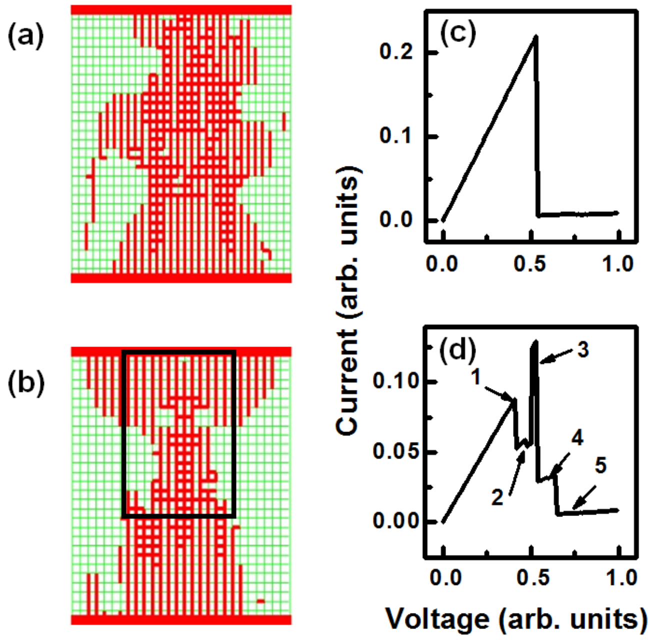

Figure 4. Simulation results for voltage-driven RESET processes using the RCB network model. All the simulations were performed in 150 × 30 square lattices of circuit breakers with 0.5% of the circuit breakers randomly turned on in the pristine state. (*a*) and (*b*) Snapshots of the percolating cluster of on-state circuit breakers in the LR state. Note that the thick (red) and thin (green) lines correspond to on- and off-state circuit breakers, respectively. (*c*) and (*d*) Voltage-driven RESET I–V sweep results from the configurations shown in (*a*) and (*b*), respectively. (Colour online.)

It is not yet clear what type of initial distribution of the Δv values can result in the observed abnormal switching behaviours, whose suppression will be highly desirable in RRAM applications. However, enhancements in suppressing abnormal switching might be feasible either by introducing defects or anisotropic conducting channels since it was reported that insertion of defects in the RCB network could reduce the fluctuations of RESET and SET voltages significantly [16]. More systematic investigations are highly desirable.

In addition to NiO, similar abnormal resistance switching also appeared in TiO_2 thin films used in [16], and Fe_2O_3 thin films in [5]. Both of the polycrystalline TiO_2 and Fe_2O_3 thin films were deposited on Pt/Ti/SiO2/Si substrates using pulsed laser deposition. Based on these observations, the abnormal RESET switching behaviour can be considered as a common characteristic of RRAM devices based on polycrystalline transition metal oxides.

4. Conclusions

In summary, we reported an abnormal resistance switching behaviour during the RESET process of NiO thin film capacitors, a phenomenon that should be overcome for practical applications of RRAM. The RCB network model was used to explain the mechanism of the abnormal resistance switching. It was revealed that the simultaneous process of formation and rupturing of the conducting channels can result in such abnormal RESET switching behaviour.

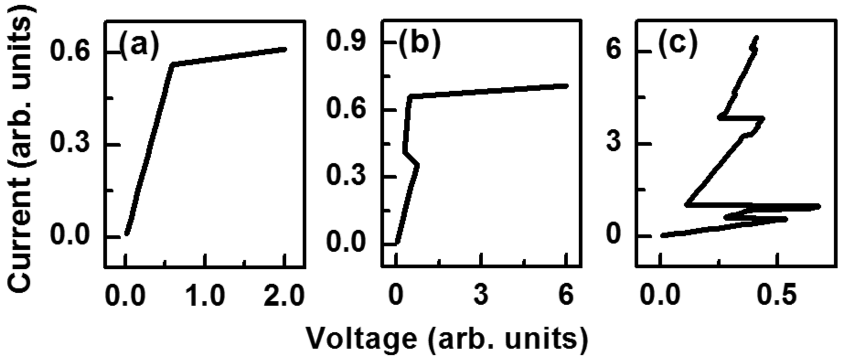

Figure 5. (*a*), (*b*), (*c*) Simulated I–V curves with different V_{COMP} values of 2, 6 and 10, respectively. Note that V_{COMP} values in the simulation are unitless.

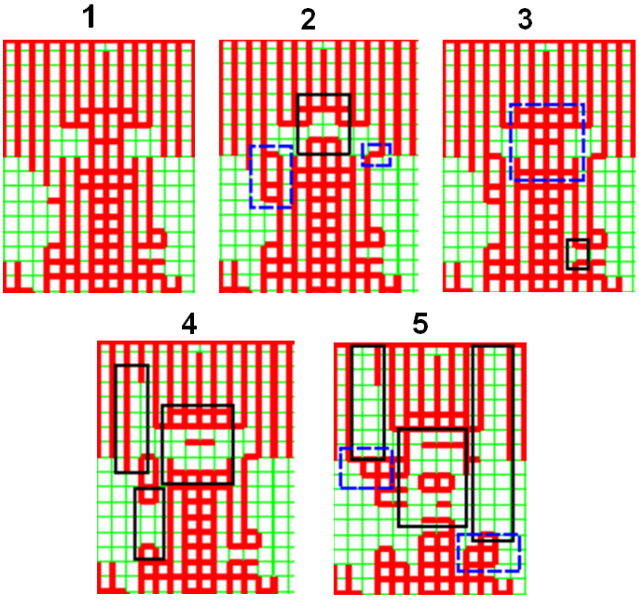

Figure 6. Snapshots of circuit breaker network configuration at each voltage, labeled in the I–V curve in figure 4(d). In each figure, the regions where conducting channels are formed and ruptured are marked by the dashed (blue) and solid (black) lines, respectively. (Colour online.)

Acknowledgments

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