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Research on resistive switching mechanism of multi-filaments formation/rupture in nickel oxide thin films

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ABSTRACT

Resistive switching (RS) effect has attracted enormous attention due to its potential for application of resistive random access memory with fast speed, good scalability and high endurance. NiO is a model system of RS effect, in which the formation/rupture of nickel filaments is directly related to the RS effect. In fact, it is still controversial that the RS effect in NiO films should be attributed to whether a single filament or multi-filaments. In this work, we demonstrate that multi-filaments are involved in the RS process by means of conventional I-V measurements, conductive atomic force microscopy and transmission electron microscopy. Furthermore, we observed the interesting tunneling magnetoresistance in this system for the first time, which not only reveals the mechanism of multi-filaments, but also provides a new way for multifunctional devices.

1. Introduction

In the past decade, resistive switching (RS) effect has attracted much attention due to its applications in resistive random access memory (RRAM) [1–5]. It has been observed in all kinds of materials, including oxides [6,7], organic [8,9], solid electrolytes [10,11] and so forth. Various mechanisms have been proposed to explain RS effect, such as formation/rupture of filament [7,12–14], interfacial barrier alteration [15], migration of oxygen vacancies [16,17], trapping/detrapping of charge carriers [18], etc. Among them, conducting filament-based RRAM has unique advantages in write/erase speed, retention time, and thermal stability, which make it promising for practical memories. However, the mechanism of RS effect in NiO films is still unclear, especially whether a single filament or multi-filaments contribute to the RS process [19].

In this study, we report a direct observation of multiple Ni filaments with the low resistance state (LRS) by using transmission electron microscopy (TEM). Multistep RESET process in I-V measurements was obtained by both conventional I-V measurements and conductive atomic force microscopy (C-AFM). In addition, we observed the interesting tunneling magnetoresistance (TMR) in the NiO RS films for the first time. The anomalous and symmetrical bulges in TMR curve prove that the high resistance state (HRS) originates from the rupture of multi-filaments. Moreover, the observation of TMR behavior demonstrate the electromagnetic coupling in this system, which provides an opportunity for multifunctional devices.

2. Experimental details

The NiO thin films were grown on Pt/Ti/SiO_2/Si substrates at 500 °C by pulsed laser deposition with 1 Torr oxygen pressure, a laser fluence of 1.2 J/cm^2 and 3 Hz repetition rate. The Au top electrodes were deposited on NiO thin films using magnetron sputtering. The microstructure of the NiO in LRS was investigated by TEM (FEI Tecnai F20, 200 kV).

Atomic force microscopy (AFM) measurements of NiO thin films were preformed in scanasyst mode. C-AFM measurements were accomplished with the help of a Bruker MultiMode Nanoscope V system at room temperature. Conductive Pt/Ir coated Si tips were used with the current sensitivity of 1 nA/V.

Magnetoresistance (MR) measurements were carried out by using a superconducting quantum interference device magnetometer at room temperature.

3. Results and discussion

The surface topology of NiO film was probed by AFM, as shown in Fig. 1(a). The little surface roughness of the film provides a guarantee for good contact between top electrodes and NiO film.

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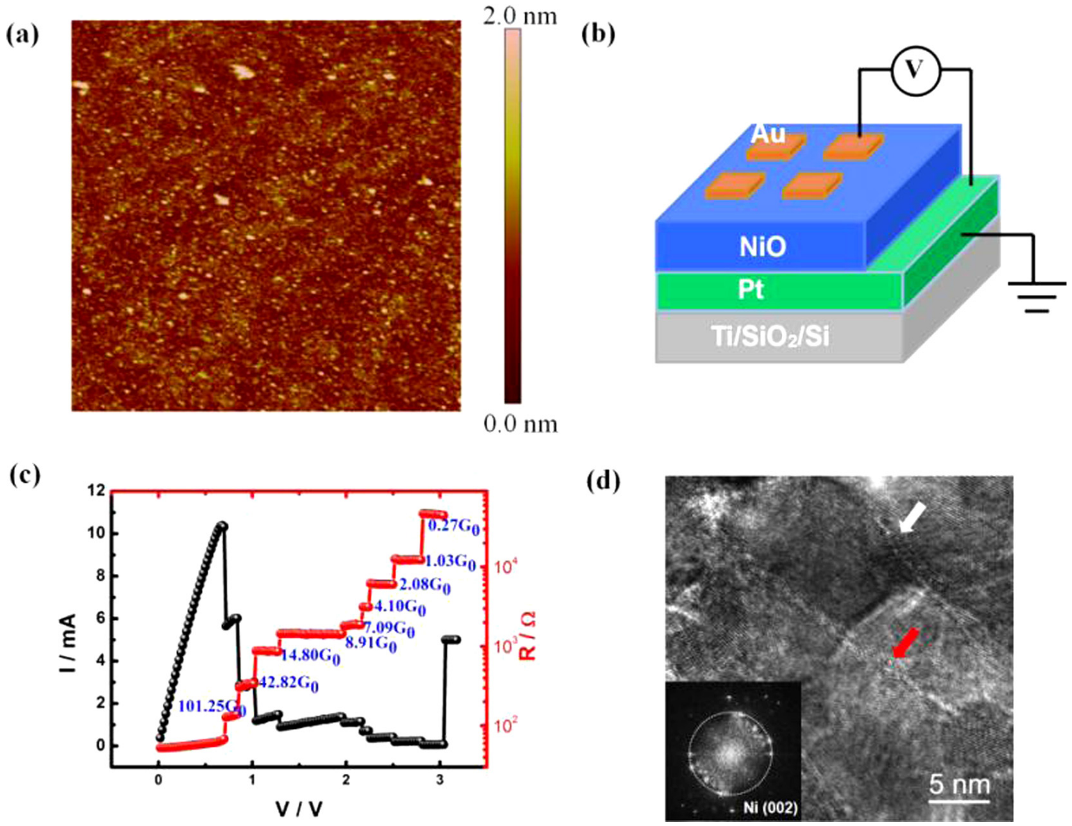

Fig. 1. (a) Surface topology of NiO film; (b) Schematic of the device structure; (c) Multistep RESET process in I-V measurements; (d) Multi-filaments observation in the LRS by TEM.

Sample configuration and the I-V curves are shown in Fig. 1(b) and (c), respectively. Interestingly, the I-V sweep exhibits multistep RESET process. Kim et al. reported the behavior of multistep transition [20]. However, it only occurred in the SET process and the origin is unclear. It is generally believed that the RS effect in NiO results from the formation/rupture of Ni filament. The multistep RESET process can be understood in terms of Ni multi-filaments instead of quantized conductance (As shown in Fig. 1(c), the conduction of the step transition is too large to be quantized conductance). In order to maintain the repetitive resistive switching, we need to set a current limit to avoid the permanent dielectric breakdown in SET process. We removed the voltage when current reached the compliance current of 5 mA, and the reversible RS process continued when the external voltage was applied from zero again.

We report a direct observation of Ni multi-filaments in NiO film with the LRS by using high-resolution TEM in Fig. 1(d). Tree-branch-like filaments (white arrow and red arrow) close to the electrode is similar to the previous theoretical assumption [21]. By applying the fast Fourier transfer (FFT), we obtained the Ni (002) diffraction spots in the inset of Fig. 1(d), which verify that the observed filamentary structures are composed of Ni atoms. No obvious Ni spots of FFT can be found in the area without filaments. The diffused FFT spots also indicate the variation of inter-atom spacing, which suggests the presence of the local atomic-structural distortion in the filaments.

Microscopically, we also make the I-V measurements by C-AFM, as shown in Fig. 2(a). The I-V curves of unipolar RS show similar behavior to the previous work [19,22,23]. In the meanwhile, the multistep RESET process was observed as well. In detail, the first RESET process starts at around 3 V, and the subsequent step transition is at about 4 V and 7 V, respectively. At last, the SET process occurs at around 10 V. Sweep the film in an area of 4 × 4 μm^2 using a tip as the top electrode in different voltages, including 2 V, 4 V, 6 V and 8 V, and the corresponding zone is in the frame with the color of viridity (LRS), light blue (HRS1), pink (HRS2) and orange (HRS3), respectively, as shown in Fig. 2(b). In order to be more intuitive, 3D image of Fig. 2(b) is shown in Fig. 2(c). We can see that the viridity frame shows the best conduction with many conducting paths while the orange frame has the poorest conduction with almost no conducting path and the zones in light blue and pink frames are in intermediate states. Here, the RS effect can be understood in terms of Ni multi-filaments instead of a single filament. Fig. 2(d) shows the current value along the blue dashed line and red dashed line in Fig. 2(b), and the current ratio of LRS/HRS3 is 87.8.

In view of Ni filaments should be ferromagnetic, the MR of NiO film in the HRS is an interesting issue and conduce to understand Ni filaments. The MR curve for the HRS with a resistance around 122 Ohm is shown in Fig. 3(a). No obvious MR is observed in the HRS with higher resistances. The MR curve shows obvious TMR effect with the ratio of 2.1%. Interestingly, the anomalous and symmetrical bulges come forth in the smooth step of resistance with the magnetic field from −5000 Oe to 5000 Oe (Three pairs of bulges can be found in the frame marked by black dashed lines). It verifies that the HRS roots in the rupture of multiple filaments. As we know, every filament is not same in the shape, thickness and structure. What's more important is that the filaments rupture in different position and the size of rupture gaps differs from each other, just as shown in Fig. 3(b). Therefore, the MR curve of every rupture filament tend to be diverse. Fig. 3(a) shows the superposed result of all MR curves.