



Multi-level resistive switching characteristics of W/Co:TiO₂/fluorine-doped tin oxide (FTO) structures

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ABSTRACT

In the present work, multi-level resistive switching (RS) in W/Co:TiO₂/FTO structures induced by a multi-mixed mechanism was studied. It was found that the devices could be reproducibly programmed into three non-volatile resistance states. And the directly switching between any resistance states was realized. This increases the operation speed and lowers the complexity of controlling circuit of multi-level non-volatile memory.

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1. Introduction

Oxide-based resistance random access memory (RRAM), which is very promising for replacing silicon-based Flash memory when its scaling ends, has attracted considerable interest recently [1–3]. Multi-level RRAM may be the most economical and effective way to achieve high storage density, compared to decrease the physical devices size [1,4]. Generally, multi-level RS, which induced either by limiting the current compliance (CC) [5] or by controlling the stop voltage [6], has been largely reported by several groups. In CC control mode, multi-level resistance states originate from the variation of width of formation conductive filaments [7]. On the other hand, in stop voltage control mode, multi-level resistance states come from the variation of length of rupture conductive filaments [7]. However, most of these multi-level RS devices could not be switched from one intermediate resistance state (IRS1) to another intermediate resistance state (IRS2) directly. For example, once the conductive filaments are formed, it is hard to control the size of filaments in CC control mode except an extra erase operation. These extra erase operations decrease the operation speed and increase the complexity of control circuit.

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There are usually two types of switching modes with respect to the operating electrical polarity [8]. The one is called unipolar, and the switching procedure is independent of the polarity of operating voltage. The physical mechanism of the unipolar RS is usually induced by the bulk filaments of forming and Joule heating rupture [1], and thus the RS operating polarity does not depend on the polarity of the operating voltage. Another type is called bipolar, where the set process and reset process must operate with the voltage of the different polarities. This is connected with the drift of ions/oxygen vacancy under electric field [1]. And the energy barrier at the interface between electrode and switching media also plays important role [9]. Previous investigations in RRAM have demonstrated that bulk filaments and interface effects could be co-existence in the same RRAM devices [10], and devices emerged stepped I-V curves [11]. Hence, it is reasonable that unipolar and bipolar RS phenomenon can be co-existence in the same devices by controlling electrical polarity and CC, and this property might enhance the multi-level RS behaviors.

In this paper, W/Co:TiO₂/FTO devices were fabricated and their RS characteristics were investigated. Enhanced multi-level RS behaviour was observed, and three resistance states could be transformed into each other freely. This multi-level RS phenomenon has not been reported yet. Detail analysis indicated that the multi-level RS originates from a multi-mixed effect of W/Co:TiO₂ Schottky-like interface and the formation/rupture of oxygen vacancy filaments in Co:TiO₂ film.

Co:TiO₂ thin films were fabricated by sol-gel spin coating technique. TiO₂ sol was prepared by a modified hydrolysis process [12]. Co(CH₃COO)₂·4H₂O was used as the doping source and mixed with the sol at a molar ratio of Co:Ti = 1:100, which corresponds to a nominal doping concentration of about 1 at.%. Positively charged oxygen vacancy are necessary to maintain charge neutrality as Co²⁺ substitutes for Ti⁴⁺ [13]. Co-doping help us to control the oxygen vacancy in switching layer and improve the device performance. Thin films with desired thicknesses (about 120 nm) were deposited onto FTO glass followed by annealing in air ambient at 550 °C for 30 min. X-ray diffraction shows that our TiO₂ film is polycrystalline with anatase structure. A rounded tungsten tip with the diameter of 20 μm was used as top electrode, FTO conductive glass as bottom electrode. The resistivity of FTO is below 10 Ω/sq. The change in resistivity was negligible after directly annealing in air below 600 °C. We tested the resistance of FTO was about 20 Ω at room temperature. Its resistance increased to about 60 Ω after annealing at 550 °C and this change was acceptable.

All devices were electrically characterized in the dark with computer controlled Keithley 2400 at room temperature. A forward (positive) bias applied to the device was defined as the current flowing from the top W electrode into the thin film.

Both bipolar and unipolar switching behavior were observed depending on the controlling process (Fig. 1). Three resistance

states were achieved. As shown in Fig. 1, the directly switching between any resistances states of the device was realized. (1) Switching between HRS and LRS (Fig. 1(a)). This is a unipolar switching. The sequence of voltage-sweeping were shown as arrows 1–4 in Fig. 1(a). A switching from HRS to LRS was achieved by applying a negative voltage of −3.4 V (V_{set1}) with a CC of 4 mA. Subsequently, the reset process which switched the device from LRS to HRS was performed by sweeping the negative voltage to −1 V (V_{reset1}) without CC. (2) Switching between HRS and IRS (Fig. 1(b)). This is bipolar switching. The positive set process which switched the device from HRS to IRS was observed by applying a positive voltage of 2.6 V (V_{set2}) with a CC of 5 mA. One obvious difference between IRS and LRS is that the I-V curves is linear in LRS while it is nonlinear in IRS. This implies that the transport mechanisms are different in these two states. And the IRS could be reset to HRS by applying a negative bias (V_{reset2}) without CC. (3) Switching between IRS and LRS (Fig. 1(c)). A bipolar switching between IRS and LRS could be achieved with a CC of 5 mA. If the device was set to IRS with a positive bias (V_{set2}), it would transfer to LRS under a negative bias of about −1 V (V_{set3}), and switch back to IRS under a positive bias of about 0.7 V (V_{reset3}).

Although the switching voltage and resistance in different samples are different and these parameters may change slightly in different cycle even in the same device, most devices showed similar

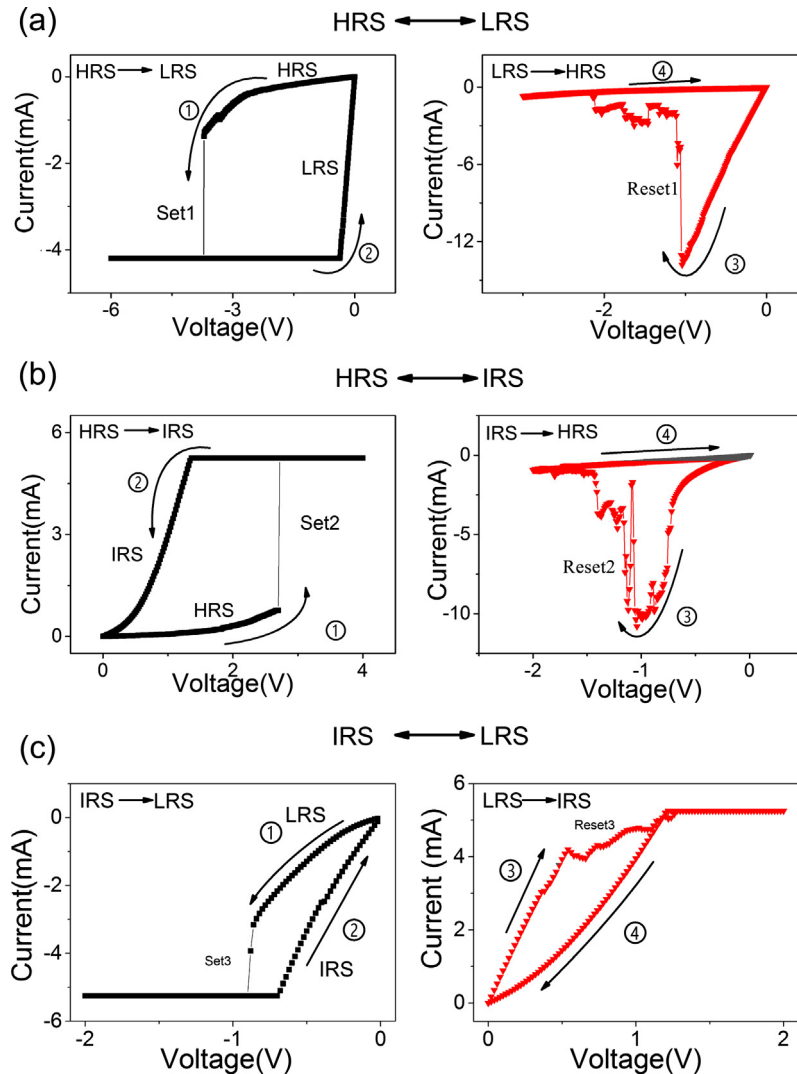


Fig. 1. (a) Switching between HRS and LRS. (b) Switching between HRS and IRS. (c) Switching between IRS and LRS.

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