Resistive Switching Effect in Co-Doped ZnO Thin Films for Nonvolatile Memory Applications

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***Abstract*— Resistive switching phenomena have attracted considerable attention because of their potential application in the next generation of nonvolatile memory. In this circumstance, resistive-change random access memory (RRAM) has got extensive interests due to its high response speed, high scalability, multibit storage potential and simple structure. Co-doped ZnO is diluted magnetic semiconductor and is would be of great interesting to realize the resistive switching property.**

In this work, we have studied the cobalt doped ZnO thin films-based RRAM structure which exhibit reproducible bipolar resistive switching behavior. The cobalt doped ZnO films were deposited on the heavily doped n-type silicon (100) substrates by ultrasonic spray pyrolysis method. The values of both a high resistance state (HRS) and a low resistance state (LRS) have little fluctuation during the cycling test, indicating the excellent switching stability. The resistance ratios of HRS to LRS are in the range of 2-3 orders of magnitude within the 80 cycles of test. To sum up, steady and reversible bistable resistive switching behavior in Au/Co:ZnO/n+-Si device provides the possibilities for nonvolatile memory applications.

Keywords—Cobalt doped ZnO, Resistive switching, HRS, LRS

I. Introduction

Resistive switching phenomenon with different performance characteristics have been reported in various materials, including solid electrolyte [1], perovskites [2], organics [3] and binary transition metal oxides [4]. Among those materials, the metal oxides represent with many advantages of low cost, facile sample preparation. As one of the most studied wide bandgap semiconductors, ZnO thin films [5] and nanorods [6] as well as Mn-ZnO films [7] have been reported to shown stable resistive switching behavior.

However, study of the resistive switching property in cobalt-doped ZnO is few. It is well known that Co-doped ZnO is diluted magnetic semiconductor and is would be of great interesting to realize the resistive switching property. In this work, we demonstrate that 1% cobalt-doped ZnO films based metal-insulator-metal (MIM) structure exhibit reproducible bipolar resistive switching behavior.

II. Methods

The cobalt doped ZnO films were deposited on heavily doped n+-type silicon (100) substrates by ultrasonic spray pyrolysis method at ambient atmosphere, as described in details in [8]. All chemical reagents utilized in the present study were of analytical grade and used without further purification. The aqueous solution of zinc acetate (0.5 mol/l) and cobalt acetate (0.005 mol/l) were used as the sources of Zn and Co, respectively. The substrate temperature was set at 450 °C and the thickness of ZnO:Co film was about 200 nm. In order to measure the electrical properties of the Co:ZnO films, Au top electrodes of 220 μm in diameter with 100nm thickness were thermally evaporated with a metal shadow mask.

The current-voltage (I-V) characteristics of Au/Co:ZnO/n+-Si structure were measured by a Keithley 2400 source-measure unit. During the measurement in voltage sweeping mode, the positive bias was defined by the current flowing from the top electrode to bottom electrode, and the negative bias was defined by the opposite direction.

III. Results

Typical I-V characteristics of RRAM device with Au/Co:ZnO/n^+-Si structure at room temperature (RT) is shown in Fig. 1(a), and the semi-log value of the current is shown in Fig. 1(b). Reversible bipolar resistance switching between a high resistance (HRS) and a low resistance (LRS) was repeatedly observed.

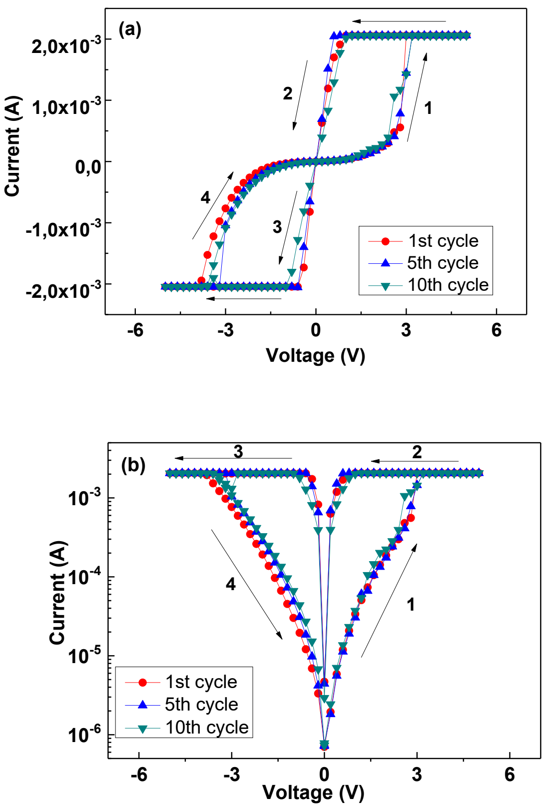

Fig. 1. Current-voltage (I-V) characteristics of Au/Co:ZnO/n$^+$-Si structure reproducibility for different cycles: (a) linear scale, (b) semi-log scale. The arrows indicate the voltage sweep direction.

The forming process with a compliance current of 3.7 mA is necessary to active the switching properties of Au/Co:ZnO/n$^+$-Si structure. The forming voltage for this structure is about 5.2 V. After forming process, the device switches to LRS. Even the external voltage is removed, the device keeps in the LRS state, displaying a non-volatile property. When the negative bias was future swept back to zero, the devices switch from LRS to HRS at about -3.2 eV, which is termed as reset process. In the subsequent cycles, the devices would switch to LRS at a lower threshold voltage at 2.8 eV, which is called set process. After forming process, the programming voltage sweep 0→5→0→−5→0 V was set to the memory cell for obtaining the reproducibility bipolar resistive switching, as shown by the numbered arrows. The little different asymmetry threshold voltage might be resulted from the different contact between top and bottom electrodes. During the switching process, a current compliance is undertaken to protect the device from the permanent breakdown.

To evaluate the switching properties of Au/Co:ZnO/n$^+$-Si device, the retention and endurance tests were then conducted , as it is shown in Fig. 2.

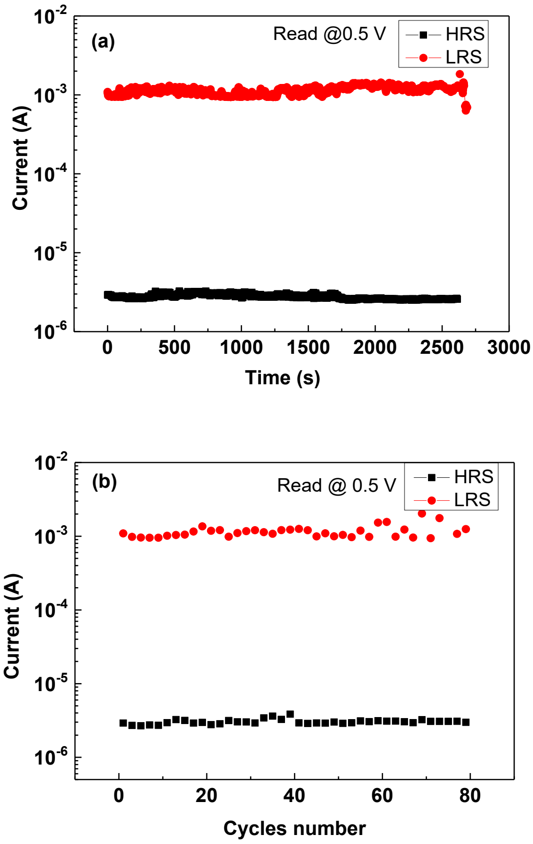

Fig. 2. (a) Retention test of Au/Co:ZnO/n$^+$-Si device read at 0.5 V. (b) Endurance performances of the Au/Co:ZnO/n$^+$-Si device during 80 sweep cycles.

Figure 2(a) shows the retention behavior of this memory device, with current values of both LRS and HRS states obtained at a reading voltage at 0.5 V. Both states show negligible shifts for more than 10^3 s. The endurance property (Fig. 2(b)) of the device was conducted with steady operations for 80 cycles. The current values were read out at 0.5 V in each dc sweep. The values of both HRS and LRS have little fluctuation during the cycling test, indicating the excellent switching stability. The resistance ratios of HRS to LRS are in the range of 2-3 orders of magnitude within the 80cycles of test. To sum up, steady and reversible bistable resistive switching behavior in Au/Co:ZnO/n+-Si device provides the possibilities for nonvolatile memory applications.

In order to clarify the origin of the switching characteristics of Au/Co:ZnO/n+-Si structure, the curve fittings of conduction mechanism for LRS and HRS were performed. The relation of current values versus voltage for the positive sweep region was drawn in double logarithmic plot, as it is shown in Fig. 3(a).

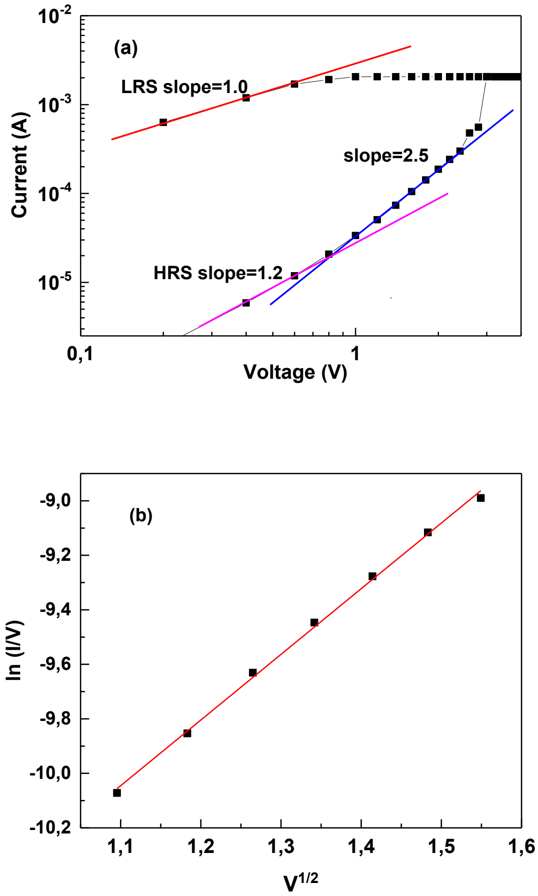

Fig. 3. (a) The double logarithmic plot of I-V characteristics for Au/Co:ZnO/n$^+$-Si structure, (b) ln(I /V) as a function of $V^{1/2}$ at the HRS region.

It can be seen from Fig. 3(a) that the conducting mechanism for the LRS currents is dominated by Ohmic conduction because the slope is close to 1.0, which is believed to be the formation of conductive filament connecting the two electrodes. For HRS curve, the nonlinear characteristic is observed, with the slope of 1.2 in low filed and 2.5 in the high filed. The classical nonlinear conduction mechanisms including Schottky emission, space charge limited current, and Poole-Frenkel (PF) emission were adopted to fit the nonlinearity HRS plot [10]. The relation ln(I/V) versus $V^{1/2}$ is fitted most linear as shown in Fig. 3(b), indicating the PF emission conducting mechanism for HRS. The PF conducting mechanism is bulk effect, which comes from a lowering of the Coulomb potential barrier of a trap site due to applied electric field. The electrons hopping between the trap states is the main contribution to the conduction of at HRS, which exists at grain boundaries or structural defects induced by oxygen vacancies and doped cobalt atoms [9].

To elucidate the conduction mechanism furthermore, we carried out the temperature dependence of dc current in HRS and LRS states as shown in Fig. 4.

It is observed from Fig. 4 that the positive temperature coefficient for the currents of LRS and HRS as measured at a reading voltage at 0.5 V, indicating a semiconductor behavior rather than the metallic in character. So the forming and rupture of metallic nanofilament as conductive channels for resistive switching can be ruled out.

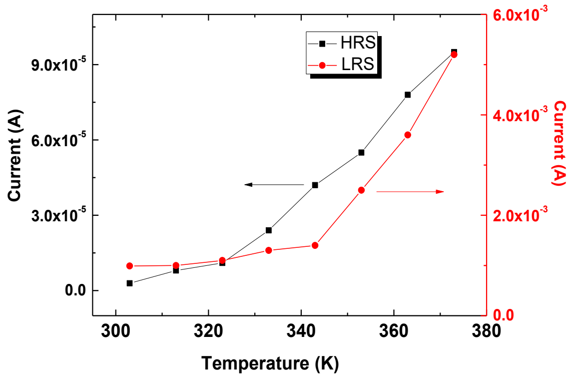

Fig. 4. The temperature dependence of dc current in HRS and LRS states measured at room temperatures.

IV. Conclusion

The RRAM structure on the base of the ZnO thin films doped by Co has been studied. The cobalt doped ZnO films were deposited on the silicon (100) substrates by ultrasonic spray pyrolysis method. The reproducible bipolar resistive switching behavior was observed. The values of both the HRS and LRS show a little fluctuation during the cycling test, indicating the excellent switching stability. The conducting mechanism for the LRS currents is dominated by Ohmic conduction, while for the HRS current the Poole-Frenkel emission mechanism was demonstrated. To sum up, steady and reversible bistable resistive switching behavior in Au/Co:ZnO/n+-Si device provides the possibilities for nonvolatile memory applications.

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