

Multilevel Set/Reset Switching Characteristics in Al/CeOx/Pt RRAM Devices

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Abstract—Al/CeOx/Pt based resistive random access memory (RRAM) devices were fabricated and investigated. The CeOx RRAM devices show self-compliance set switching without a requirement of high voltage electric forming process. Multilevel set and reset switching processes were observed in the CeOx RRAM devices. Based on the unique distribution characteristic of oxygen vacancies in CeOx films, the possible mechanism of multilevel resistive switching (RS) in the CeOx RRAM Devices was discussed.

Keywords— multilevel resistive switching, RRAM, CeOx

I. INTRODUCTION

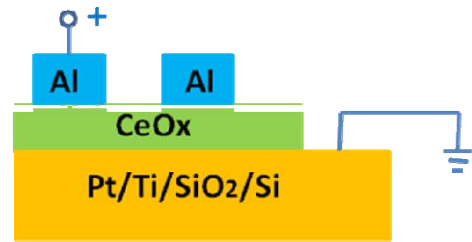
Resistive random access memory (RRAM) based on metal oxides has been actively studied due to its low power consumption, high speed, high scalability, multi-bits storage ability and good CMOS compatibility [1, 2]. Multilevel memory ability is important for the ultra-high-density memory device application. Multilevel resistive switching (RS) characteristics have been observed in some oxide-based RRAM devices [3, 4]. However, the deep understanding of characteristics and mechanism of multilevel RS in oxide-based RRAM is still a lack, which hampers the performance improvement of multilevel memory devices. In this paper, we report multilevel RS characteristics including set process and reset process in the Al/nonstoichiometric CeOx /Pt RRAM devices and discuss the possible mechanism of multilevel set/reset characteristics.

II. EXPERIMENTAL

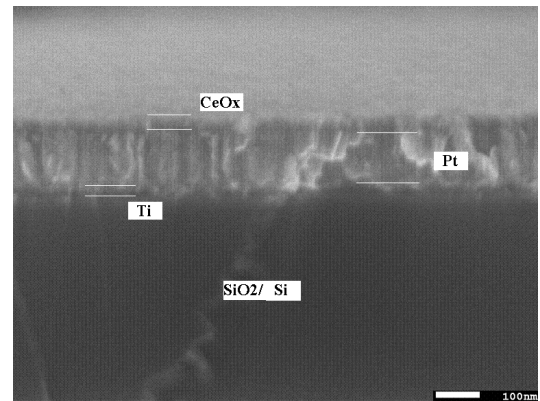
RRAM devices with the structure of Al/CeOx/Pt were fabricated. First, about 130nm Pt bottom electrode (BE) with a 20nm Ti adhesion layer was sputtered on SiO₂/Si substrates. Then, CeOx films with the thickness of about 40 nm were deposited on Pt/Ti/SiO₂/Si substrates at 450 °C in a 30 Pa flowing oxygen ambient by pulsed laser deposition. Finally, about 100nm Al top electrode (TE) with an area of 0.25mm² was evaporated on the CeOx films using a shadow mask. The electric measurements of RRAM devices were performed with an Agilent 4156C semiconductor analyzer. The microstructure and composition characteristics of the CeOx films were analyzed by the scanning electron microscope (SEM), X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS) measurements.

III. RESULTS AND DISCUSSION

A. Material characteristics



(a)



(b)

Fig.1 (a) schematic device structure of Al/CeOx/Pt RRAM, (b) the cross-sectional SEM image of CeOx/Pt/Ti/SiO₂/Si.

The schematic device structure of Al/CeOx/Pt RRAM is shown in Fig.1 (a). Fig.1 (b) shows the cross-sectional SEM image of a CeOx/Pt/Ti/SiO₂/Si stack. XRD measurements were performed to examine the microstructure of the CeOx films. The XRD result indicates that the CeOx film has polycrystalline fluorite structure.

Fig.2 shows the Ce 3d core level XPS spectra of a CeOx film. It can be found from the XPS spectra that Ce³⁺ and Ce⁴⁺ ions coexist in the CeOx film. It indicates that the as-deposited CeOx film is nonstoichiometric and oxygen vacancies are formed in the as-deposited film.

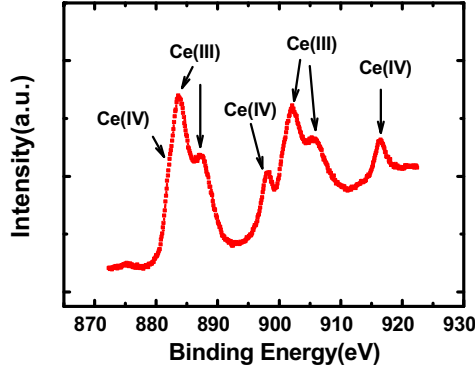


Fig.2 the Ce 3d core level XPS spectra of a CeOx film.

B. Electrical characteristics

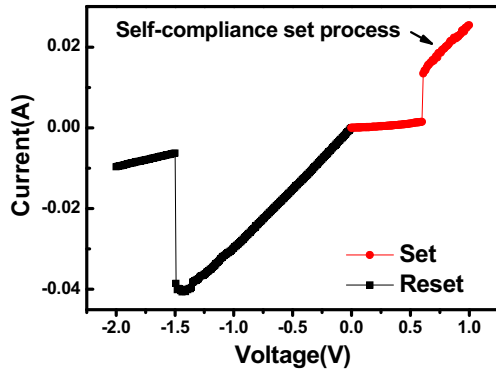


Fig.3 typical bipolar resistive switching in the CeOx RRAM device.

Electrical characteristics were measured by using an Agilent 4156C under dc voltage sweep mode. The sweeping voltage was applied on Al TE while the Pt BE was grounded. Typical bipolar RS was observed, as shown in Fig.3. The initial resistive state is at high resistive state (HRS). When the positive sweep voltage was applied to Al TE, the CeOx RRAM device switches to low resistive state (LRS) without a current compliance. Set process (transition from HRS to LRS) can self-stop in LRS. It indicates that the CeOx RRAM device shows self-compliance set process. Self-compliance set/reset resistive switching characteristics have been reported in some oxide-based RRAM devices [5, 6]. The self-compliance resistive switching is helpful to simplify the RRAM circuit design. Moreover, no high voltage electric forming process was needed during initial set process.

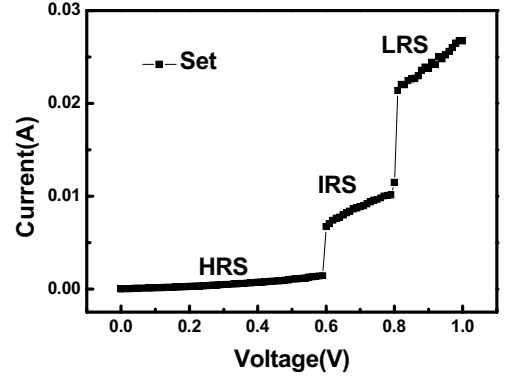


Fig.4 multilevel set process in the CeOx RRAM Device.

Multilevel set switching process was observed in the CeOx RRAM device, as shown in Fig.4. It implies that there are intermediate resistive states (IRS) in the CeOx RRAM Devices. Reset process (transition from LRS to HRS) only took place when negative voltage bias was applied to Al TE. Multilevel reset characteristic was also observed, as shown in Fig.5. It can be seen that the intermediate resistive states also exist during reset process.

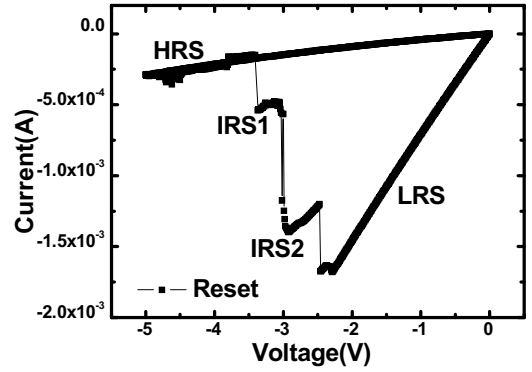


Fig.5 multilevel reset process in the CeOx RRAM Device.

These results suggest that the as-deposited CeOx RRAM devices have some metastable intermediate resistive states. IRS becomes higher from IRS2 to IRS1 and finally switches to HRS with the increasing of reset voltage. The switching from IRS to HRS was studied during reset process. Fig.6 shows the relationship between the resistance value of IRS (R_{IRS}) and I_{reset} . I_{reset} means the highest current value in reset process. It can be found that the I_{reset} linearly decrease with the increasing of IRS value. Therefore, the multilevel resistive states can be obtained by controlling the reset voltage and current. The multilevel set/reset characteristics are helpful for the CeOx RRAM devices to obtain multi-bits memory ability.

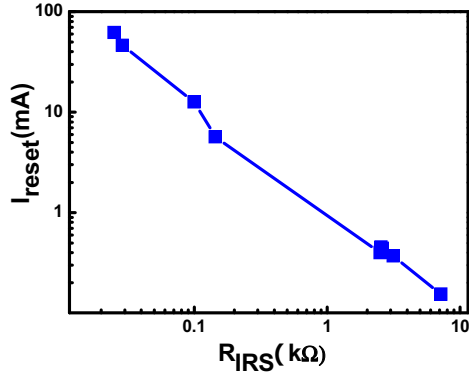


Fig.6 R_{IRS} dependence of I_{reset} during reset process.

C. Possible mechanism of multilevel switching

Oxygen vacancy conducting filament mechanism has been widely used to explain the resistive switching characteristics in oxide-based RRAM devices. In as-deposited CeOx films, oxygen vacancies preexist according to the XPS result. The high density pre-existing oxygen vacancies may be responsible for the absence of high voltage electric forming process in the fresh CeOx RRAM devices.

Oxygen vacancies in CeOx incline to distribute in line pattern and separate with each other due to impulsive interactions [7-9]. Therefore, independent multiple filaments can be formed under appropriate voltage bias, which lead to intrinsic multilevel resistive states. The intrinsic multilevel resistive states could be ascribed to the unique separated line-pattern distribution characteristics of oxygen vacancies in the nonstoichiometric CeOx films. The density of oxygen vacancy near Al reactive electrode/CeOx interface is higher than that inside a bulk because of the lower formation energy at surfaces and the reductive ability of Al electrode, resulting in the nonuniform distribution of oxygen vacancies. The nonuniform distribution of oxygen vacancies can illustrate the bipolar resistive switching in CeOx RRAM devices. The multilevel set and reset characteristics can be well explained based on the separated line-pattern distribution characteristics of oxygen vacancies in the CeOx film. Fig.7 shows the schematic multilevel set and reset process. Under positive voltage bias, the positive charged oxygen vacancies are pushed from the Al/CeOx interface to CeOx bulk to form the separated line-pattern filaments. Under negative voltage bias, oxygen vacancies are pulled back to Al/CeOx interface and the filaments ruptured. These oxygen vacancy conducting filaments in CeOx films are formed or ruptured sequentially during set or reset process, which can account for the multilevel set and reset characteristics.

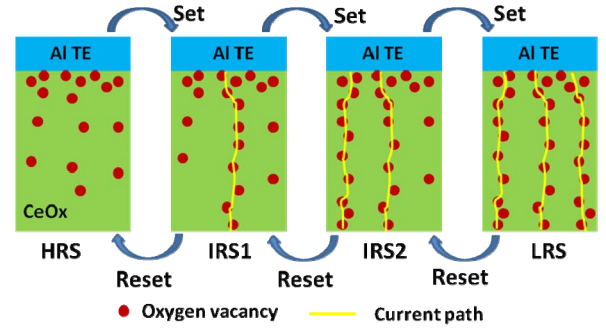


Fig.7 schematic multilevel set/reset process in the CeOx RRAM.

IV. CONCLUSION

RRAM devices based on the nonstoichiometric CeOx films were fabricated and studied. The CeOx RRAM devices show self-compliance set process and have no need of high voltage electric forming process. Multilevel set and reset switching process were observed in the CeOx RRAM device. An oxygen vacancy multifilament conduction mechanism was applied to explain the multilevel resistive switching characteristics.

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