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Unipolar resistive switching behavior of amorphous $SrMoO_4$ thin films deposited at room temperature

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ABSTRACT

Amorphous $SrMoO_4$ (SMO) thin films were deposited on Pt/Ti/SiO_2/Si substrates at room temperature by pulsed laser deposition and the resistive switching (RS) behavior of the Au/SMO/Pt devices was investigated. The Au/SMO/Pt devices exhibit typical unipolar RS behavior with excellent switching parameters as follows: high resistance ratio (~10^5) between the low resistance state (LRS) and high resistance state (HRS), non-overlapping switching voltages, and good endurance and retention characteristics. Detailed analysis of their current-voltage characteristics reveals that the conduction mechanisms are Ohmic conduction in the LRS and lower voltage region of HRS, and Poole-Frenkel emission in the higher voltage region of the HRS. Temperature dependent resistance measurements, combined with x-ray photoelectron spectroscopy and model analysis indicate that the unipolar RS behavior of the Au/SMO/Pt devices could be understood by a conical conducting filaments (CFs) model in which the conical CFs are composed of oxygen vacancies. The conical CFs extend from the cathode to anode during the forming process and the observed RS behavior occurs in the localized region near the anode. These results suggest that the room-temperature- deposited amorphous SMO thin films could find potential application in nonvolatile RS memory.

1. Introduction

Resistive random access memory (RRAM) based on resistive switching (RS) behavior has attracted a great deal of attention for next generation nonvolatile memory applications due to its outstanding features of simple structure, high switching speed, low power consumption, long retention time, and good compatibility with complementary metal oxide semiconductor (CMOS) technology [1–4]. The RRAM devices possess a simple metal/insulator/metal (MIM) structure and store information through reversible switching between a high resistance state (HRS) and a low resistance state (LRS) under external electrical stimuli. By applying appropriate voltages, a cell in its HRS can be "Set" to LRS, then again "Reset" back to HRS. The RS behavior can be classified into unipolar and bipolar types in terms of the polarity of the applied voltages for switching [2–4]. For the unipolar RS, the switching procedures involve the amplitude of the applied input (voltage/current) and are independent of the bias polarity. In contrast, the bipolar RS requires opposite electrical polarities for the resistance change between HRS and LRS.

A large variety of materials have been found to show RS behaviors, including binary oxides, ternary and more complex oxides, carbon based materials, and organic materials [5–10]. The conducting filaments (CFs) model has been proposed to be responsible for the reversible switching between HRS and LRS [2–4,11,12]. Structural inhomogeneities, such as those introduced by defects in crystalline systems, are thought to play an important role in the formation of CFs. For example, the grain boundaries in NiO thin films and the quasi-one-dimensional defects of dislocations in a single crystal of $SrTiO_3$ could act as easy and fast diffusion paths for the Ni interstitial and the oxygen vacancy [13,14]. However, material granularity would limit scalability since devices of size comparable to the grains themselves will result in performance instability and variability. Amorphous thin films could be advantageous for down scaling for nonvolatile memory application because they lack grain boundaries [9,15–18]. In addition, the growth of thin films at low temperatures (< 200 °C) is preferred for integrating the RRAM devices with the existing semiconductor device technology. There have been several recent reports on investigations of RS behaviors of oxide thin films fabricated at low temperatures to

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investigate their suitability for nonvolatile memory applications [18–23].

Molybdenum can have multiple valence states from +3 to +6. Molybdenum oxides are therefore expected to have various structural and electronic phases, which might facilitate the RS behavior in the molybdenum oxide-based RRAM devices. It has been reported that $MoO_3$and barium molybdate $BaMoO_4$exhibit excellent RS behaviors [24–26]. However, a relatively high temperature (~500 °C) is needed to deposit the MoO_3thin films showing RS behavior, and that may hinder its practical applications in RRAM devices. In contrast, $BaMoO_4$can be prepared at a low temperature of 140 °C [26]. These results motivate us to investigate whether other molydates such as $SrMoO_4$(SMO) deposited at low temperatures also show excellent RS behavior. SMO is an important member of molybdates family, which belongs to the scheelite structure family with a C_{4h} symmetry having two formula units per unit cell. Although SMO has been investigated for various applications such as luminescence hosts, white LEDs, and biomedical materials [27–29]. However, there is no report on utilizing SMO thin films as the RS material to construct nonvolatile memory cells. This paper describes investigations of the RS behaviors of the Au/SMO/Pt memory devices made of amorphous SMO thin films deposited by pulsed laser deposition at room temperature. The Au/SMO/Pt devices exhibit unipolar RS behavior with excellent switching parameters: a high resistance ratio (~10^5) between LRS and HRS, non-overlapping switching voltages, and good endurance and retention characteristics. Temperature dependent resistance measurements, x-ray photoelectron spectroscopy, and model analysis are combined to show that the unipolar RS behavior of the Au/SMO/Pt devices can be understood by a conical conducting filaments (CFs) model and that the conical CFs are composed of oxygen vacancies. The conical CFs extend from the cathode to the anode during the forming process, and the RS behavior occurs in a localized region near the anode.

2. Experimental procedure

An SMO sintered ceramic target with a diameter of 2.5 cm was ablated with a KrF (248 nm) excimer laser (Coherent COMPexPro102). The laser beam was focused onto the target within the vacuum chamber, through a quartz glass window using a spherical lens with a focal length of 50 cm. The beam was incident at an angle of 45° on the rotatable target positioned at a distance of 5 cm from the substrate. The laser operated at a repetition rate of 4 Hz and produced an energy density of 2 J/cm^2. Each laser pulse vaporized or ablated a small amount of the SMO material. The ablated SMO material was ejected from the target in a forward-directed plume that provided the material flux for film growth. The base pressure of the vacuum chamber before film deposition was about 5×10^{-4} Pa. Amorphous SMO thin films of thickness ~180 nm were grown on commercially available Pt/Ti/SiO_2/Si substrates at room temperature and oxygen pressure of 10 Pa. The oxygen used as the reactive gas was controlled by a mass-flow controller (MFC, Sevenstar D07-7B). The structural properties of the films were characterized by x-ray diffraction (XRD, Philips X'Pert Pro MPD) operating at 40 kV and 40 mA using K_α radiation (λ=0.154 nm). The films' thickness was measured by a field emission scanning electron microscope (SEM, FEI Sirion 200). The surface morphological properties of the films were investigated by a atomic force microscopy (AFM, Cypher ES). X-ray photoelectron spectroscopy (XPS, Thermo ESCALAB 250) with monochromatic Al K_α ($h\nu$=1486.6 eV) was used to evaluate the chemical bonding states of the elements in the SMO thin films. To fabricate the MIM sandwich structures, the Au top electrodes (200 μm in diameter) were deposited at room temperature by dc-magnetron sputtering with a shadow mask. The switching characteristics and conduction mechanisms of the Au/SMO/Pt devices were studied through current-voltage (*I-V*) measurements in top-bottom configuration using a Keithley 2400 source-measure unit.

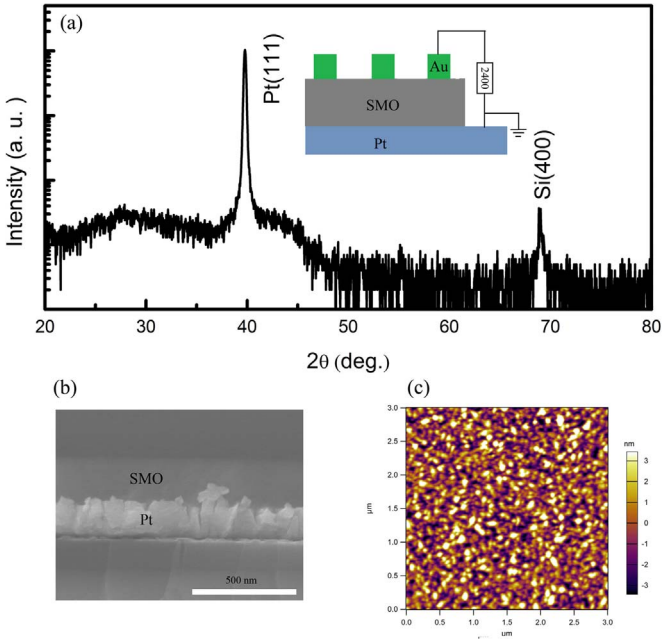

Fig. 1. (a) XRD pattern of SMO thin film. The inset shows the schematic diagram of Au/SMO/Pt device. (b) Cross-section SEM image of the SMO thin film on Pt/Ti/SiO_2/Si substrate. (c) AFM topography image of the SMO thin film.

3. Results and discussion

The XRD pattern of the SMO thin film is shown in Fig. 1(a). The absence of any characteristic XRD peak other than the (111) and (400) diffraction peaks of Pt and Si from the SMO thin film indicates that it is amorphous. The formation of the amorphous SMO thin film can be ascribed to the low substrate temperature, which restrains the surface mobility of the species adsorbed from the target and leads them to be frozen in random positions. Fig. 1(b) shows a cross-section SEM image of the SMO thin film grown on the Pt/Ti/SiO2/Si substrate. The thickness of the SMO thin film is about 180 nm. Fig. 1(c) presents the AFM topography image of the SMO thin film. It demonstrates that the surface of the SMO thin film is smooth with a root-mean-square roughness of about 2 nm. The schematic diagram of the Au/SMO/Pt device structure is also depicted in the inset of Fig. 1(a).

Fig. 2 shows the typical *I-V* characteristics of the Au/SMO/Pt device

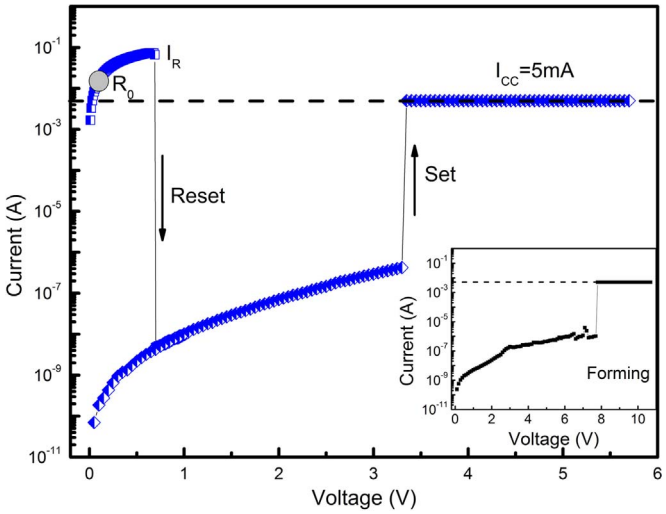

Fig. 2. Typical *I-V* characteristic in semi-logarithmic scale of the Au/SMO/Pt devices. The inset shows the forming process. R_0 is the resistance of the device in LRS and I_R is the reset current.

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