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Temperature-dependent resistive switching characteristics for Au/n-type $CuAlO_x$/heavily doped p-type Si devices

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ABSTRACT

Bipolar switching phenomenon is found for Au/n-type $CuAlO_x$/heavily doped p-type Si devices at temperatures above 220 K. For high or low resistive states (HRS or LRS), the electrical resistance is decreased with increasing temperature, indicating a semiconducting behavior. Carrier transport at LRS or HRS is dominated by hopping conduction. It is reasonable to conclude that the transition from HRS to LRS due to the migration of oxygen vacancies (V_O) is associated with electron hopping mediated through the V_O trap sites. The disappearance of the resistive switching behavior below 220 K is attributed to the immobile V_O traps. The deep understanding of conduction mechanism could help to control the device performance.

1. Introduction

Resistive random access memory (RRAM) has been widely viewed as a promising candidate for future data storage applications [1]. RRAM offers superior performance, such as high density, fast programming speed, low power consumption, good retention and simple structure. Resistive switching (R_S) characteristics have been discovered in various materials, especially in metal oxides (ZnO, InGaZnO, TaO_x, MnO_2, NiO, HfO_2, etc.) [2–10]. However, few reports on $CuAlO_x$ RRAMs are found. Zhang et al. [11] demonstrated high-performance, flexible RRAM prototype devices using amorphous p-type $CuAlO_x$ with different oxygen concentrations as the switching layer. For Cu:AlO_x/TaO_x/TiN memory devices, memory characteristics and switching mechanism were investigated by Roy et al. [12]. Therefore, it is necessary to take a deeper insight into the R_S mechanism of CuAlOx–based devices. In addition, $CuAlO_x$ could be used to make all-oxide (transparent) p–n junction and transistors by combining two-type transparent conductive oxides [13,14]. To our knowledge, there have been no reports on the fabrication the n-type $CuAlO_x$-based R_S devices. The motivation of present work is to explore the current–voltage (I–V) characteristics of the n-type $CuAlO_x$-based R_S devices for RRAM applications. In addition, the sensitivity of the R_S characteristics to temperature provides an opportunity to realize stable and reliable R_S properties in the $CuAlO_x$-based memory devices. In this study, we demonstrate that the temperature-dependent conduction mechanisms of Au/n-type $CuAlO_x$/heavily doped p-type Si (p^+-Si) devices and reveal how high resistive state (HRS) changes into low resistive state (LRS). These performance changes were shown to be due to electron hopping mediated through the $+2$ charged oxygen-vacancy (V_O) sites.

2. Experimental

Four-inch Si (100) wafers purchased from Woodruff Tech Company were used in the experiment. The resistivity of the p^+-Si wafer is about 0.001 Ω cm. The p^+-Si film thickness was about 525 μm. Before deposition of $CuAlO_x$, the p^+-Si samples were cleaned in chemical cleaning solutions of acetone and methanol, rinsed with de-ionized water, and blow-dried with N_2. Next, the p^+-Si sample was chemically etched with a diluted HF solution for 30 s, rinsed with de-ionized water and blow-dried with N_2. A SiO_2 layer was grown on the p^+-Si wafer using a dry oxidation process. The n-type $CuAlO_x$ films were prepared on p^+-Si and SiO_2/p^+-Si substrates by rf magnetron sputtering. The target size is 2 in and the target-substrate distance is 65 mm. A high-purity $CuAlO_2$ target (rf power was fixed at 80 W) was used for deposition of $CuAlO_x$ films. The sputtering pressure was fixed at 5×10^{-3} Torr. The substrate temperature was fixed at 500 °C. The target was used in conjunction with Ar/O_2 as an ambient gas for sputtering. The flow of Ar and O_2 was 70 and 10 SCCM (SCCM denotes standard cubic centimeter per minute), respectively. The film thickness of $CuAlO_x$ was estimated to be 100 nm. The structural properties were determined by X-ray diffraction (XRD) using Cu Kα radiation. The carrier concentration, mobility, resistivity and conduction type were obtained from the Hall

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measurements in the van der Pauw configuration for $CuAlO_x/SiO_2/p^+$-Si samples. For $CuAlO_x/p^+$-Si and $CuAlO_x/SiO_2/p^+$-Si samples, the electrodes were fabricated by depositing Au metal on the $CuAlO_x$ layer through a shadow mask. The $CuAlO_x$ thin films show n-type behavior. According to the Hall measurement at room temperature, the electron concentration and mobility were 4.1×10^{14} cm^{-3} and 0.8 cm^2 V^{-1} s^{-1}. For Au/$CuAlO_x/p^+$-Si devices, the R_S properties are studied using I–V measurements. The I–V curves were measured using a Keithley Model-4200-SCS semiconductor characterization system. The I–V characteristics were measured in the temperature range from 123 to 423 K in steps of 30 K using a temperature controlled cryostat. X-ray photoelectron spectroscopy (XPS) was employed to identify the chemical bonding state and the atomic concentration. XPS measurements were performed using a monochromatic Al Kα X-ray source. The photon energy was calibrated with the Au 4f core-level line.

3. Results and discussion

Fig. 1 shows the XRD patterns of $CuAlO_x$ films. All diffraction peaks in the patterns of both materials could be indexed by assuming the delafossite structure. In the XRD spectra shown in Fig. 1, peaks of the $CuAlO_2$ phases are respectively marked. Three peaks [corresponding to the (006), (101) and (018) planes] [15–18] were observed. No peaks corresponding to other Cu and Al oxides were found in the pattern.

Fig. 2 shows a typical I–V curve with set/reset processes at 303 K. The voltage was applied on the p^+-Si, while the Au electrode was grounded in all the tests. This configuration has no influence on the presented model for determining the resistance states. If the voltage is applied on the Au electrode and the p^+-Si is grounded, the set (reset) process will occur at the positive (negative) voltage. In Fig. 2, different resistance states occur during the voltage sweeping. It can be inferred that the V_O defects in the n-type $CuAlO_x$ layer are responsible for hysteresis behavior. As shown in Fig. 2, the Au/$CuAlO_x/p^+$-Si device shows bipolar switching phenomenon, with the transition from HRS to LRS occurring under negative voltage and reset process happening at the positive direction. For the RRAMs using metal oxides as switching layers, the conduction channels usually consists of oxygen vacancies [19–21].

To confirm the influence of oxygen vacancies on the conduction behaviors of HRS and LRS, electrical measurements were carried out on Au/$CuAlO_x/p^+$-Si structure with various temperatures. Transport mechanism analysis could be clearer and interesting phenomena could be observed if the ambient temperature is low. Fig. 3(a) and (b) show temperature-dependent I–V curves. In order to elucidate the R_S mechanism of Au/$CuAlO_x/p^+$-Si devices, the values of resistance (R) for both HRS and LRS for a bias voltage of +2 V were derived based on measurement data [Fig. 3(a) and (b)]. Fig. 3(c) shows R as a function of temperature (T). For HRS or LRS, the electrical resistance is decreased with increasing temperature, indicating a semiconducting behavior. In addition, the R_S behavior is observed at temperatures above 220 K. In metal oxide based devices the conduction is believed to be through conduction channels formed in regions with higher concentration of V_O [5, 22–24]. Changes in V_O distribution lead to changes in resistance states, with the LRS having a higher V_O concentration in the vicinity of the p+-Si electrode area compared with the HRS [6]. The intrinsic carrier concentration (n_i) is a strong function of temperature, so the Fermi energy level (E_F) is a function of temperature also [25]. As the temperature decreases, n_i decreases, and E_F moves closer to the conduction band. At temperatures below 220 K, E_F goes above the energy level of V_O and these V_O states below E_F are full, thus forming the neutral and immobile V_O traps. The diffusion of V_O has been found at temperatures above 200 K [26]. The R_S properties at temperatures above 220 K is linked to the V_O migration and is associated with electron hopping mediated by the mobile V_O traps.

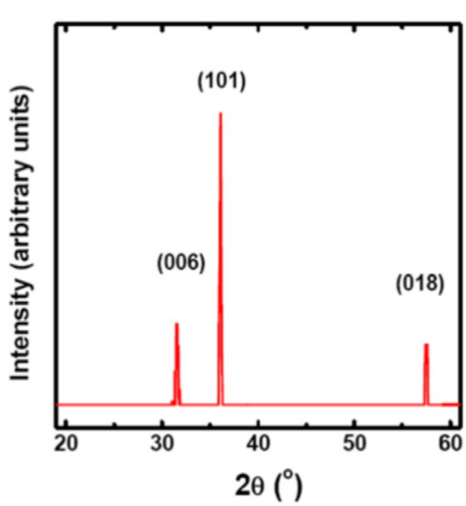

Fig. 1. XRD patterns of $CuAlO_x$ films.

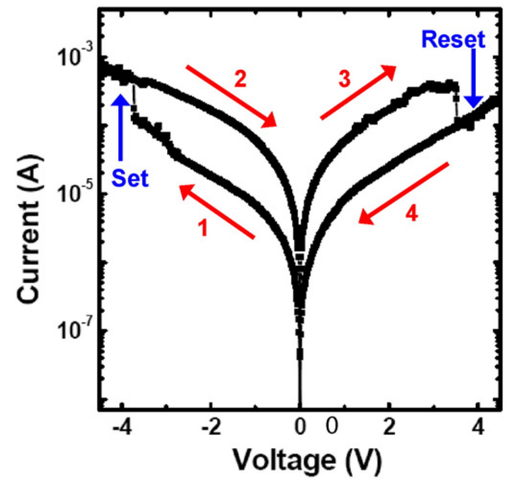

Fig. 2. Typical bipolar resistance switching I–V curve of Au/$CuAlO_x/p^+$-Si devices at 303 K (Note: the Au electrode is grounded).

In order to understand this phenomenon, an analysis using the temperature-dependent I–V characteristics was presented. Fig. 4 shows the temperature-dependent I–V curves in the voltage range of 1.5–2.5 V for Au/$CuAlO_x/p^+$-Si devices in the LRS and HRS regions at temperatures above 280 K, respectively. Fig. 4 also shows the temperature-dependent I–V curves in the voltage range of 1.5–2.5 V for Au/$CuAlO_x/p^+$-Si devices at temperatures below 220 K. The log (I)–V curves are almost linear and I increases as T increases, which indicate that the currents exhibit hopping conduction behavior [27]. For hopping conduction, the conduction current increases with T, because thermally excited electrons hop from one trap state to another trap state in the $CuAlO_x$ layer. In order to study the electrical conduction mechanism, the I–V curves are measured and compared at various temperatures. The areas of Fig. 4(a) [Fig. 4(b) and (c)] that are denoted with rectangles are used for the construction of Fig. 5(a) [Fig. 5(b) and (c)]. Fig. 5 shows ln(I)–(1/T) curves in the LRS (HRS) region at temperatures above 280 K for V = 1.50, 1.75, 2.00, 2.25 and 2.50 V, respectively. Fig. 5 also shows ln(I)–(1/T) curves for Au/$CuAlO_x/p^+$-Si devices at temperatures below 220 K for V = 1.50, 1.75, 2.00, 2.25 and 2.50 V, respectively. The ln(I)–(1/T) curve is linear (Fig. 5), which indicates that hopping conduction is the dominant process [27]. The hopping conduction can be expressed as [7,27–29]:

$$I = Sqa_h n_e v_f \left[\exp\left(\frac{qa_h V}{2dkT} - \frac{q\phi_t}{kT} \right) \right] \quad (1)$$

where S is the contact area, d is the film thickness, q is assumed to be the electronic charge, a_h is the mean hopping distance, $q\phi_t$ is the barrier