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Resistive random access memories fabricated by using solution-processed AlZnSnO semiconductor films and indium ball electrodes

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ABSTRACT

Resistive random access memories (ReRAMs) using solution-processed AlZnSnO (AZTO) oxide semiconductor films as resistive switching layers were fabricated. Electrodes were prepared by applying Ag paste or by pressing indium balls onto the AZTO surface. Vacuum processes were not required. The resistive switching behavior was not found for the ReRAM using a Ag paste electrode. However, a significant resistive switching characteristic could be obtained when using an In ball as the electrode. In addition, by adjusting compliance current from 80 mA to 20 mA, a stable resistance difference between high and low resistance state of 1839 Ω could be obtained over 500 operations. The ReRAM also exhibited good data retention capability and read disturb immunity for at least 10^3 s. Physical mechanisms of current conduction and resistance switching behavior were also investigated.

1. Introduction

Resistive random access memories (ReRAMs) have attracted great interest because of their advantages of low operation voltage and simple structures. ReRAMs using amorphous metal oxide semiconductors (AOS) as resistive switching layers have been reported to exhibit remarkable resistive switching characteristics. However, AOS and metal electrodes are conventionally prepared by radio-frequency magnetron sputtering [1–6]. The high-vacuum systems and fabrication facilities result in substantial manufacture cost and much time consumption.

Solution processes are known to have advantages of high throughput, low cost, and fast fabrication. Zinc salts have been widely used as precursors to obtain ZnO-related oxide semiconductors. Pugliese et al. used zinc nitrate hexahydrate to obtain ZnO flowerlike microstructures for applications to dye-sensitized solar cells [7]. For exploitation of solar light, Mostoni et al. investigated photocatalyst properties of bismuth-doped ZnO nanoparticles that were prepared by an impregnation method and by a sol gel synthesis [8]. One-Dimensional ZnO/Au junction obtained by dielectrophoresis for simultaneous and versatile multi-sensing measurements was investigated in Ref. [9]. Application of solution-processed AlZnSnO (AZTO) films to thin film transistors was studied in Ref. [10].

In this work, a ReRAM using a solution-processed AZTO oxide semiconductor as a resistive switching layer was fabricated. Moreover, the metal electrodes were prepared by just placing silver paste or In balls on the surface of the spin-coated AZTO layer. A rapid, simplified, and low-cost process to fabricate ReRAMs was demonstrated. Although the ReRAM using silver paste as the electrode cannot exhibit resistive switching behavior, the ReRAM with an In ball electrode can give a significant resistance switching characteristic. Effect of compliance current (I_C) on ReRAM performance was also studied. The AZTO ReRAM has good endurance, data retention capability, and read disturb immunity. The physical mechanisms for carrier transport and resistive switching behavior were suggested and verified by X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS) analysis. This novel fabrication process may be applicable to mass production of high-performance and low-cost ReRAM devices in the future.

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2. Experiments

First, 5 ml of anhydrous ethanol was added to 0.1 M zinc acetate dehydrate [$Zn(CH_3COO)_2 \cdot 2H_2O$] (ZnAc), 0.1 M tin chloride dehydrate [$SnCl_2 \cdot 2H_2O$], and 0.01 M aluminum nitrate nonahydrate [$Al(NO_3)_3$], respectively. Second, mono-ethanolamine (MEA) was added to the ZnAc solution with ZnAc:MEA molar concentration of ~1:1. These solutions were then stirred at 60 °C for one hour. The AZTO precursor solution was obtained by mixing the above solutions and followed by stirring at 70 °C for 30 min. Finally, the precursor solution was aged at room temperature for 24 h. After cleaning the n^+ Si substrate, a AZTO film was spin-coated on the substrate at 3000 rpm for 20 s followed by a curing process at 120 °C for 10 min to remove the solvent and organic residuals. Second and third AZTO films were subsequently spin-coated on the first AZTO layer by using the same spinning and curing condition. The top electrode was prepared by applying silver paste to the AZTO surface to obtain an Ag/AZTO/n^+ Si device. For In/AZTO/n^+ Si ReRAMs, electrodes were fabricated by pressing In balls with a diameter of 500 μm onto the AZTO surface. The electrical characteristics of the ReRAMs were measured by using a semiconductor parameter analyzer. The crystallinity and the chemical structure of the AZTO film was analyzed by XRD and XPS, respectively.

3. Results and discussion

The I-V curves of the Ag/AZTO/n^+ Si device for 400 voltage cycles are shown in Fig. 1(a). The voltage cycle is $0 \rightarrow V_{max} \rightarrow 0 \rightarrow -V_{max} \rightarrow 0$. The I_C is set to be 80 mA. The I-V curves coincide and resistance switching behavior is not found in both positive and negative voltage regions. This is due to Ag diffusion in the AZTO film. A conducting filament formed by internal diffusion of Ag atoms into oxide semiconductor ZnO layer with thickness of 100 nm was proposed in Ref. [11]. A conduction path in 35 nm-thick SiO_2 layer formed by accumulated Ag atoms was found in Ref. [12]. In addition, oxygen vacancies will enhance the internal diffusion of Ag atoms [11]. The AZTO thickness in this work is only 15 nm. The conducting paths caused by Ag diffusion are hard to rapture and the bottom electrode is easily electrically connected to the top electrode via the Ag atoms. Thus, resistance switching characteristic cannot be obtained. When the top electrode of the Ag paste is replaced by an In ball, significant bipolar resistance switching behavior can be found as shown in Fig. 1(b). However, the resistance window is unstable and the switching characteristic is nearly absent when the cycle reaches ~350 times. Interestingly, when the I_C is lowered to 20 mA, stable bipolar resistive switching characteristic can be obtained, as shown in Fig. 1(c). Compared with Fig. 1(b) and (c), the observation that a lower I_C of 20 mA led to a smaller resistance window was also found in a ReRAM with a sputtered AZTO resistance switching layer [13]. Endurance characteristic shown in Fig. 2(a) reveals a stable resistance difference of 1839 Ω between high resistance state (HRS) and low resistance state (LRS). Obvious degradation is not found up to 500 switching cycles. This resistance window is observably higher than 402 Ω of a pristine IGZO ReRAM with a sputtered IGZO layer and an evaporated electrode during 100 switching cycles [5]. For the data retention test (Fig. 2(b)), significant degradation is not found up to 10^3 s. Even under constant voltage stress (CVS), stable data retention ability can also be observed ,as shown in Fig. 2(c).

The carrier conduction mechanisms of the ReRAMs are explored in Fig. 3(a). Conduction current density dominated by trap-filled space charge limit conduction (SCLC) can be described as

$$J = \frac{9}{8}\epsilon_i \mu \theta \frac{V^2}{d^3} \tag{1}$$

where ε_i is the permittivity, θ is the ratio of free charge density to total charge density, V is the voltage, and d is the distance [2]. In LRS, when I_C is 80 mA the slope of the linear fitting line is ~2, which is in accordance with equation (1). This means that traps are filled by abundant injected carriers caused by the high I_C of 80 mA and the conduction current is dominated by trap-filled SCLC [14]. When the ReRAM switches to HRS, the slope of linear fitting line in low electric field regime is still ~2. However, the slope of the fitting line changes to 2.7 in high electric field regime. It implies that the conduction mechanism departs from trap-filled SCLC when the voltages increases to ~1 V. Poole-Frenkel (P-F) effect describes that a high electric field can lower the potential barrier and the trapped electrons can have a higher possibility for being thermally excited from the traps to the conduction band [15]. It can be expressed by the following equation [16]

$$J = q\mu N_C E \exp\left[\frac{-q\left(\Phi_T - \sqrt{qE/\pi\epsilon_i}\right)}{kT}\right] \tag{2}$$

where q is the elementary charge, N_C is the effective density of states of the conduction band, E is the electric field, Φ_T is the barrier height for electron emission from the trapping states, k is Boltzmann's constant, and T is temperature. Based on equation (2), linear dependence of ln(I/V) on ($V^{1/2}$), as shown in Fig. 3(b), verifies the conduction current is dominated by P-F emission in the high electric field regime [17]. It is noted that the curve lower than voltage of 1 V departs from linearity. This confirms that the carrier transport in low electric field regime is not dominated by P-F mechanism. In the low electric field regime, the trapped carriers

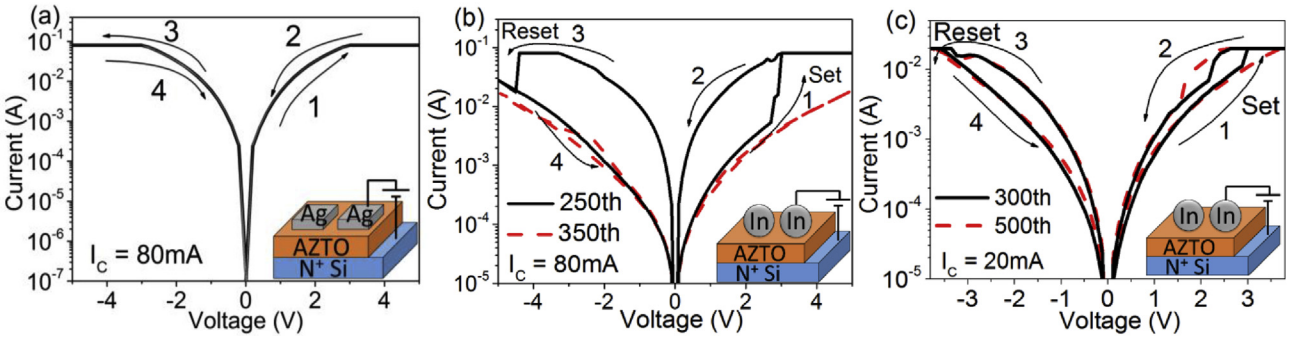

Fig. 1. I-V curves of (a) Ag/AZTO/n^+ Si measured with a I_C of 80 mA and In/AZTO/n^+ Si devices measured with I_C of (b) 80 mA and (c) 20 mA.