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Improved endurance in ultrathin Al_2O_3 film with a reactive Ti layer based resistive memory

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ABSTRACT

A 3-nm-thick Al_2O_3 based resistive memory with a Ti layer was prepared in this work. The Ti/HfO_x devices with the same thickness were also fabricated for comparison. The oxygen gettering of Ti form Al_2O_3 is lower than that from HfO_x. The Al_2O_3 devices with strong dielectric strength exhibit tight distribution of initial resistance state and high resistance state. The low resistance state, SET and RESET voltage of the devices seems insensitive to the dielectric film. Without an ideal current limiter, high forming voltage (V_F) leads to the Al_2O_3 device with poor endurance (<100 cycles). Reduction of current overshoot by an external resistor of 800 Ω during forming process, the endurance of the memory device can be improved and increase up to 10^4 cycles. The current overshoot and V_F in the Al_2O_3 device with high operational temperature during forming step can be also suppressed.

1. Introduction

Binary oxide based resistive memories (RMs) with excellent scalability, simple device structure, low power consumption and high speed: are considered as one of attractive candidates for next generation nonvolatile memory application [1]. Several binary oxides as the insulator with bistable resistive switching (RS), including NiO [2], TaO_x [3,4], TiO_2[5,6], HfO_x [7,8], ZrO_2 [9], CuO_x [10], and Al_2O_3[11], have been extensively addressed. The negative differential resistance effect was observed in anodic Al_2O_3 layer by Hickmott [12]. Atomic layer deposition (ALD) is one of the most promising technologies to grow high quality binary oxide and thin insulator [13]. With thin thickness, the forming voltage of the RM can be reduced [7]. In the structure of Pt/Al_2O_3/Ru, the filaments in the insulator were found to be more difficult to form and disconnect, the devices exhibited low resistance ratio and fluctuation in resistance states [14]. Recently, the Al_2O_3 based RM by ALD with an ultra-low RESET current were reported [13], however, the ALD prepared Al_2O_3 based devices with high bandgap and good dielectric strength suffers high forming voltage (V_F) and poor endurance.

The authors demonstrated a reactive Ti capping layer and an optimal HfO_x thickness to obtain a robust RS with high endurance [7]. The bipolar resistance switching in Pt/Ti/Al_2O_3/Pt device with a 40-nm-thick insulator by sputtering was demonstrated [15]. However, Wu et al. used a 1-nm Ti above 10-nm-thick Al_2O_3 layer, the device can be operated by unipolar mode [13]. These results suggested that the characteristics of the Al_2O_3 RM devices are strongly dependent on the selection of electrode layer, the preparation method, and the thickness of Al_2O_3 layer. In this work, we comprehensively studied the characteristics of ultra-thin (3 nm) Al_2O_3 devices with a reactive Ti layer. The device with a 3-nm-thick Al_2O_3 layer with an appropriate V_F and sufficiently high dielectric strength were prepared. This characteristics result in the device with a stable resistive switching and a relief of current overshoot during forming step. Thick Al_2O_3 devices with high V_F suffered serious current overshoot and exhibited unswitchable characteristic. The device with Al_2O_3 thickness less than 2 nm showed a high leakage current in the initial state. The devices with an identical thickness of HfO_x were also prepared for comparison. Suppression of the current overshoot by the loaded resistor or high operational temperature during the forming process can improve the cycling numbers of endurance for the Al_2O_3 devices.

2. Device fabrication

The bipolar RM devices were prepared through the stacked layers, which consisted of Al_2O_3 and HfO_x as shown in the inserted of Fig. 1, TiN were served as top electrode and bottom electrode (BE), the detail fabrication process of HfO_x can be checked elsewhere [7], ALD process were also adopted to prepare Al_2O_3 films with the precursor of $Al(CH_3)_3$ and water. The thickness of the reactive Ti and

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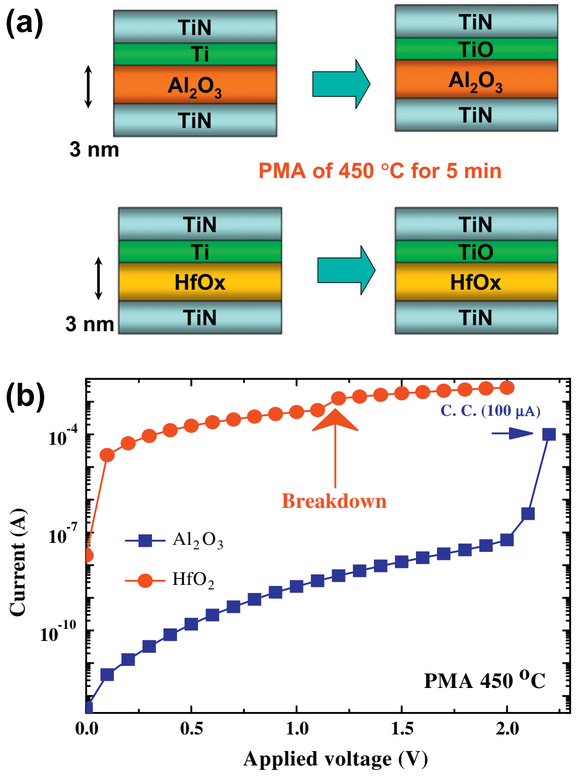

Fig. 1. (a) Schematic diagrams for the as-grown and PMA of Ti/Al_2O_3 and Ti/HfO_x stacked layer, and (b) typical *I*–*V* curves in the pristine Al_2O_3 and HfO_x devices.

dielectric layer is 10 and 3 nm, respectively. Standard lithography and dry etching were adopted to pattern the memory cell. Low temperature oxide was used as an encapsulation layer. The finished memory devices with cell size of 0.13 μm^2 after post-metal-annealing (PMA) of 450 °C were referred as the Al_2O_3 and HfO_x devices. The microstructure of the stacked structure was investigated by cross-sectional transmission electron microscopy (XTEM) operated at 200 keV. The crystal structure of 3-nm-thick Al_2O_3 film is amorphous and that of HfO_x is amorphous with embedded nanocrystals. X-ray photoelectron spectroscopy (XPS) was employed to examine the chemical bonds of the insulator. Two configurations of contacted pad (referred as short BE and long BE) were used to explore the effect of external resistor on the first RESET and RS of Al_2O_3 device. The electrical properties of the RM cells are characterized with short BE mode by HP 4156 and HP 4284 LCR meter. With long BE mode, an extra resistor of 800 Ω was only adopted during electrical measurement for the forming step. During the cycling test, all the devices are performed with short BE configuration. Fifty devices were evaluated to explore the electrical uniformity of the devices. Positive bias is referred as the top TiN electrode applied with positive voltage.

3. Results and discussion

In Fig. 1, the typical *I*–*V* curves in the pristine Ti/Al_2O_3 and Ti/HfO_x devices were presented. The initial state of the Al_2O_3 device shows a better insulator property than that of HfO_x device with the same thickness of dielectric film (3 nm). The theory of Gibbs free energy for metal oxide may give some clarification to the reactive electrode of Ti influence on the memory performance of the RMs. The standard Gibbs free energy of formation at 450 °C about TiO, Al_2O_3, and HfO_2 compound is respectively −955, −966, and −940 kJ/mol. The Ti layer can react with HfO_x easier than that of Al_2O_3 [15]. Hence, the Ti capping layer can capture more oxygen species in the buried HfO_x than Al_2O_3. This result is responsible for the leaky property of Ti/HfO_x device at the same bias. The permanent damage will occur in the HfO_x device with the operation bias higher than breakdown voltage (BV). The *J*–*V* curves of the pristine Al_2O_3 and HfO_x devices at different temperatures were also measured to explore their conduction mechanism (Fig. 2a). For the Ti/Al_2O_3/TiN device, the *J*–*V* curves show weak temperature dependence and a good linear relationship in the *J*–V^2 plot in the median voltage range from to 0.15 to 0.3 V. The ln(*J*)–ln(*V*) curve for the pristine Al_2O_3 device are presented in

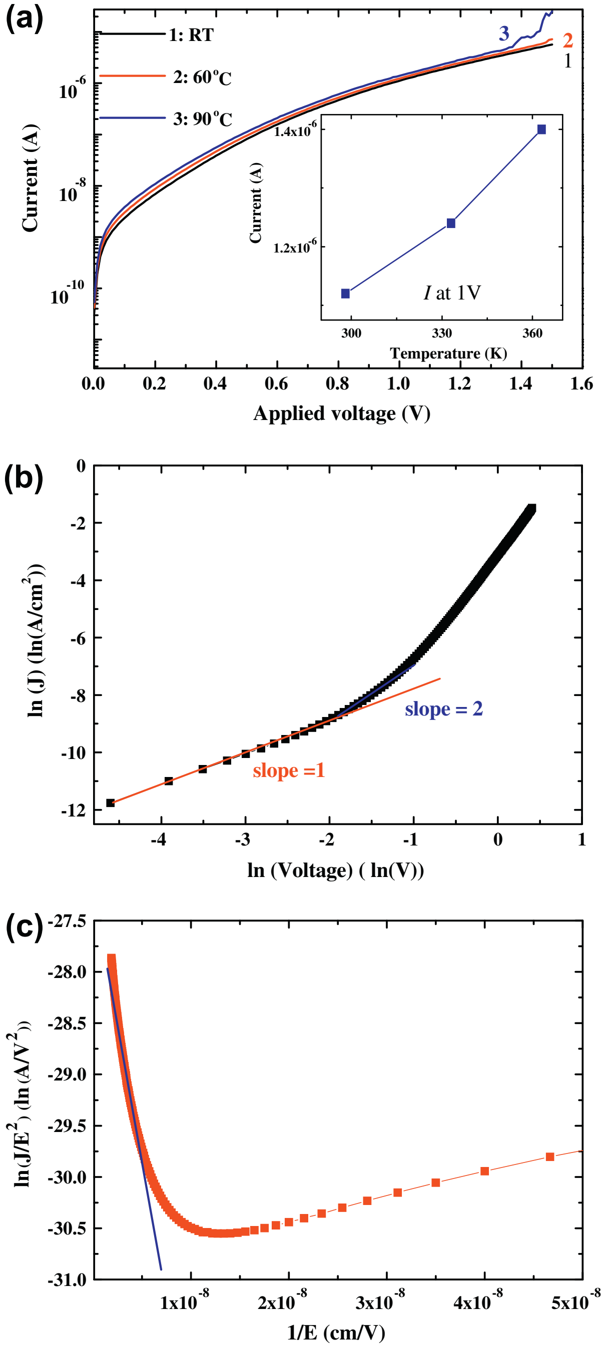

Fig. 2. (a) Temperature dependence of leakage current, (b) the *J*–*V* characteristic in a double-logarithmic plot, and (c) the Fowler–Nordheim tunneling plot of log(J/E^2) versus 1/*E* of the fresh Al_2O_3 devices. The measurement temperatures were varied between 27 and 90 °C, the inserted in (a) is the leakage current at different measurement temperature.