



Multi-mode resistive switching behaviors induced by modifying Ti interlayer thickness and operation scheme



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ABSTRACT

To overcome the practical issues existed in RRAMs application, complementary resistive switching (CRS) and self-compliance effect are investigated to alleviate sneak current issue and avoid external compliance current (I_{comp}), respectively. It is remarkable that the two resistive switching (RS) behaviors are attempted to achieve in the same system of Pt/HfO₂(7.5– x)/Ti(2 x)/HfO₂(7.5– x)/Pt ($x = 0, 2.5, 3.5$ nm) device fabricated by magnetron sputtering, i.e., multi-mode resistive switching behaviors are modulated by varying Ti interlayer thickness and operation scheme, such as initial I_{comp} and applied voltage during reset switching. As indicated by the results, typical bipolar resistive switching (BRS) and CRS behaviors exhibit in Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt devices, and self-compliance effect appears in Pt/HfO₂(~5 nm)/Ti(~5 nm)/HfO₂(~5 nm)/Pt devices. After investigated the conductive mechanisms, it is proposed adopting conceivable switching mechanism associated with the formation of oxygen vacancy (V_{O}) and metallic conduction filament (CF) to understand CRS and self-compliance effect. The exploration of multi-mode RS behaviors paves the way to improve the flexibility and convenience of RRAMs in actual application.

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1. Introduction

In the recent years, resistive random access memories (RRAMs) have attracted much attention at home and abroad, due to their simple structures, high response speed, low energy consumption [1,2] and combinations with other functions such as ferromagnetic and optical [3,4]. As the most prospective candidate of storage devices for next generation, much better understanding of the switching mechanisms has been acquired for us [1,2,5]. However, there are still practical issues on the journey to achieve commercial application. Primarily, misreading problem originated by sneak current issue in the crossbar architecture is a tough obstacle for high density integration. In addition, I_{comp} is always essential for preventing RRAMs from perpetual breakdown during set process, inevitably increasing the complexity for actual operation to a certain extent. To address the problems, more effort has been paid and the research demonstrate that CRS cells can alleviate sneak current issue effectively [1,6–8], and self-compliance RS is an excellent choice for implementing switching from high resistance

state (HRS) to low resistance state (LRS) without I_{comp} applied [9,10]. Unfortunately, above two RS modes are carried out in standalone systems with different structures and storage/electrode materials, and there is only a few investigation on the multi-mode resistive switching behaviors in a same system [11], thus we are dedicated to design RRAMs with multi-mode RS behaviors in a same system to enhance the flexibility and convenience in practical application.

In this manuscript, device units composed of I/M/I (insulator/metal/insulator)-based functional stacks on Pt substrates with Pt top electrodes are designed to realize multi-mode RS phenomena. Generally speaking, constructing a CRS device, two BRS devices series back-to-back is a reliable route [12,13]. As for the layout of devices with self-compliance effect, a serial resistance [14,15] in switching layer can help to achieve it, and the key lies in the modification of defect distribution through various methods, e.g., nanowires [16], electrode engineering [17] and interface engineering [14] etc. Inspired by the considerations above, HfO₂/Ti/HfO₂ tri-layer structure is designed to serve as RS layers, which not only satisfies the structure criteria of CRS unit, but also introduces Ti interlayer as defect distribution regulator (V_{O} reservoir). HfO₂ is selected for its advantages of sub-ns operation speed and exceptional endurance [18,19], thus it becomes the most promising

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storage material in next-generation RRAMs. Moreover, its simple composition and fabrication in various methods, i.e., magnetron sputtering [20], ALD [19], EBPVD [21], make it more attractive for memories. As for metal interlayer, titanium becomes the candidate not only for its extensive usage as electrodes in various oxide switching layer [17,22,23], but also for V_O reservoir function originating from its chemical activity. Based on the investigation of I – V characteristics of the devices, the results show that multi-mode RS behaviors could be exhibited in a same system with the structure of Pt/HfO₂(7.5– x)/Ti(2 x)/HfO₂(7.5– x)/Pt through modifying the thickness of metal interlayer and adopting appropriate operation scheme. Specifically, the devices possessing a constant thickness (~15 nm) for total functional layer with different thickness of Ti layer are investigated, and through adjusting I_{comp} in initial switching, CRS and self-compliance effect were successfully presented in the device Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt and Pt/HfO₂(~5 nm)/Ti(~5 nm)/HfO₂(~5 nm)/Pt, respectively. What's more, assisted by inspecting the conductive and switching mechanisms of reference device Pt/HfO₂(~15 nm)/Pt, the RS mechanisms for multi-mode RS are thoroughly analyzed and proposed.

2. Experimental details

In this work, three types of device are fabricated. During preparation, HfO₂ films were firstly deposited on prefabricated Pt/Ti/SiO₂/Si substrates by radio frequency (RF) magnetron sputtering with a HfO₂ ceramic target at room temperature. The base vacuum of the chamber was better than 6×10^{-5} Pa and sputtering pressure was fixed at 0.8 Pa. Subsequently, titanium layers were deposited on HfO₂ films by direct current (DC) magnetron sputtering, and then grew the upper HfO₂ films with the same manufactured parameter as the lower fraction. Pt electrodes (~60 nm-thick) were deposited through a simple DC magnetron sputtering with ~50 μm radius produced by conventional ultraviolet lithography and lift-off processes. The schematic configurations of the three terminal devices are depicted in Fig. 1, in which 2 x represents the thickness of Ti interlayer, and x equals 0, 2.5, 3.5 corresponding to Pt/HfO₂(~15 nm)/Pt, Pt/HfO₂(~5 nm)/Ti(~5 nm)/HfO₂(~5 nm)/Pt, and Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt device, respectively.

All the electrical characteristics were conducted by an Agilent B1500A semiconductor parameter analyzer. The bias voltages were applied on Pt top electrodes (TEs) with Pt bottom electrodes (BEs) grounded at room temperature.

3. Results and discussion

The pristine Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt devices usually possess a high resistance of $\sim 5 \times 10^7 \Omega$, indicating good insulating property of sputtered HfO₂ film even though a Ti layer is introduced. The initial switching (shown in Fig. 2(a)) from HRS

(state ①) to LRS occurs when external voltages sweep from 0 V to –2 V, with a I_{comp} of 10 mA adopted to avoid permanent dielectric breakdown during set process. It's worth noting that the initial set process presents an obvious two-step current skip, occurring at voltages of ~ -1.3 V (marked as V_1) and ~ -1.6 V (marked as V_2), respectively. After initial process, stable switching behaviors are demonstrated in Fig. 2(a). It can be seen that the devices can be set to LRS (state ③) under negative voltage polarity (marked as 'NS'), and be reset to HRS (state ④) under same voltage polarity (marked as 'NR'). Also, it can set to LRS (state ③) and reset to HRS (state ⑤) under negative (marked as 'NS') and positive (marked as 'PR') voltage polarity, respectively. The stable RS under same and different voltage polarity reflects the devices possess typical nonpolar resistive switching characteristic. Since there is a two-step current skip with I_{comp} of 10 mA, a smaller I_{comp} (1 mA) was applied to control only a one-step current skip during the initial switching, and stable BRS between state ② and state ⑥ is achieved with external voltage of 2 V, as exhibited in Fig. 2(b). This indicates that unipolar and bipolar RS modes can co-exhibit in Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt devices, similar with the devices possessing M/I/M/I/M structure [24]. Further evaluation of the retention performance for the devices in unipolar and bipolar RS modes were conducted with a constant voltage of 0.1 V applied just followed the resistive switching for 2000 s (the inset of Fig. 2(a) and (b)), respectively. Excellent retention properties of the device in two switching modes manifest that the device possesses the potential for multi-mode resistive switching application.

In order to investigate the cycle-to-cycle stability and reproducibility of BRS of the devices, 300 successive switching cycles of a randomly selected device were recorded and are shown in Fig. 2(c). The red and blue curves in the figure represent the 10th cycle and the 300th cycle, respectively. It can be easily seen that all of the typical curves overlap very well to exhibit good cycle-to-cycle stability and reproducibility during the recorded 300 successive switching cycles. The cycle-to-cycle endurance performance is further analyzed by evolution of resistances of R_{LRS} and R_{HRS} of the device based on recorded 300 switching cycles, as shown in Fig. 2(d). It can be seen that both of HRS and LRS are stable as cycles running, and R_{LRS} keeps stable at $\sim 10^3 \Omega$ at 0.1 V, with the maximum current as low as $\sim 100 \mu\text{A}$, signifying the potential of low power consumption integrated application.

Stimulated by the small current fluctuation in negative-set curve in Fig. 2(a), where the current increases firstly and reduces subsequently, we tried to explore the existence of CRS behavior in the devices. With the method from Ref. [8], we obtained an intermediate resistance state by applying a 'soft' reset process (1 V), after one-step initial switching. Subsequently, CRS behavior emerges under voltage swept in a sequence of 0 V $\rightarrow$ –1 V $\rightarrow$ 0 V $\rightarrow$ 1 V $\rightarrow$ 0 V, and the stable switch curve with the state evolution as ② $\rightarrow$ ⑦ $\rightarrow$ ⑧ $\rightarrow$ ② $\rightarrow$ ⑦ $\rightarrow$ ⑧ $\rightarrow$ ② is plotted in Fig. 3(a). Different with the devices with similar stacks reported to exhibit unipolar or/and bipolar RS in the devices [20,24], CRS mode can achieve under proper operation scheme in our devices, besides unipolar and bipolar RS modes, indicating the potential of multi-mode RS for Pt/HfO₂(~4 nm)/Ti(~7 nm)/HfO₂(~4 nm)/Pt devices, along with the excellent retention property under a constant voltage stress, shown as inset of Fig. 3(a). In addition, I – V characteristics of the devices also manifest that RS behaviors can be modified by controlling initial I_{comp} and reset voltages [25].

In order to investigate the stability and reproducibility of the devices for CRS behavior, successive 35 cycles in a single device is provided in Fig. 3(b). The red and blue curves represent the first and the 35th cycle, respectively. It can be seen that excellent cycle-to-cycle endurance performance is confirmed for no destructive

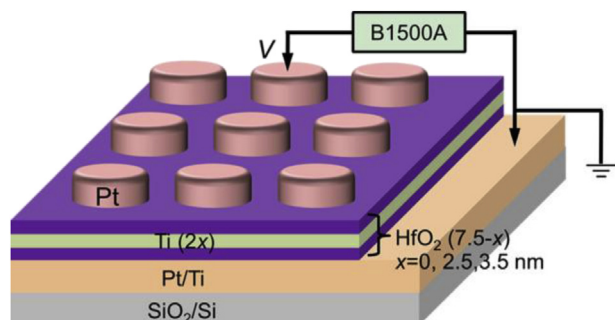


Fig. 1. Schematic configurations of HfO₂-based devices and measurement setup.

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