



# Physical Mechanism and Performance Factors of Metal Oxide Based Resistive Switching Memory: A Review

Cong Ye<sup>1</sup>, Jiayi Wu<sup>1</sup>, Gang He<sup>2,\*</sup>, Jieqiong Zhang<sup>3</sup>, Tengfei Deng<sup>1</sup>, Pin He<sup>1</sup>, Hao Wang<sup>1,\*\*</sup>

<sup>1</sup> Hubei Collaborative Innovation Center for Advanced Organic Chemical Materials, Hubei Key Laboratory of Ferroelectric and Dielectric Materials and Devices, Faculty of Physics and Electronic Science, Hubei University, Wuhan 430062, China

<sup>2</sup> School of Physics and Materials Science, Radiation Detection Materials & Device Lab, Anhui University, Hefei 230039, China

<sup>3</sup> Department of Electrical Engineering, City University of Hong Kong, Hong Kong Special Administrative Region, China

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This review summarizes the mechanism and performance of metal oxide based resistive switching memory. The origin of resistive switching (RS) behavior can be roughly classified into the conducting filament type and the interface type. Here, we adopt the filament type to study the metal oxide based resistive switching memory, which considers the migration of metallic cations and oxygen vacancies, as well as discuss two main mechanisms including the electrochemical metallization effect (ECM) and valence change memory effect (VCM). At the light of the influence of the electrode materials and switching layers on the RS characteristics, an overview has also been given on the performance parameters including the uniformity, endurance, the retention, and the multi-layer storage. Especially, we mentioned ITO (indium tin oxide) electrode and discussed the novel RS characteristics related with ITO. Finally, the challenges resistive random access memory (RRAM) device is facing, as well as the future development trend, are expressed.

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## 1. Introduction

The resistive switching (RS) phenomenon was firstly reported in a series of binary oxides by Hickmott in 1962<sup>[1]</sup>. Until now, the scientific and industrial interests on the resistance random access memory (RRAM) have already lasted for more than 50 years. Extensive research on the physical analyses and mechanisms has been performed to understand the electrical phenomena in the 1960s and 1970s<sup>[2–6]</sup>. In fact, there are various candidates (e.g. magnetic random access memory [MRAM], ferroelectric random access memory [FRAM], phase change random access memory (PRAM) and resistive random access memory [RRAM]<sup>[10–16]</sup>) for next-generation memory to replace current flash memory for its disadvantages of high operating voltage, low operating speed, high power consumption and the scaling limitation to 16 nm<sup>[7–9]</sup>. Fortunately, RRAM has been considered to be the most promising one, owing to its advantages of simple structure, low operating power and fast switching speed<sup>[17,18]</sup>. The RRAM structure is a switching layer sandwiched between two metal electrodes, which is a metal–insulator–metal (MIM) structure. It has been found that a great variety of materials

including transition metal oxides, perovskite, organic materials have shown RS characteristics<sup>[19–22]</sup>. Owing to the extreme diversity of RS materials, it is not possible to review all the related issues. Among them, metal oxide based resistive switching memory is widely investigated due to its simple composition, low cost, and compatibility with CMOS (complementary metal–oxide–semiconductor transistor) technology, such as NiO<sup>[23–26]</sup>, TiO<sub>2</sub><sup>[27–31]</sup>, ZrO<sub>2</sub><sup>[32,33]</sup>, Al<sub>2</sub>O<sub>3</sub><sup>[34,35]</sup>, HfO<sub>2</sub><sup>[36–41]</sup>, ZnO<sup>[42–44]</sup>, and WO<sub>3</sub><sup>[45]</sup>. It has been reported that memory cells such as TiN/TiO<sub>x</sub>/HfO<sub>x</sub>/TiN have shown switching speed of ~5 ns<sup>[46]</sup>. Lee et al.<sup>[47]</sup> demonstrated a TaO<sub>x</sub>-based asymmetric passive switching device with extreme cycling endurance of over 10<sup>12</sup>. Furthermore, the structure of TiN/HfO<sub>x</sub>/Pt exhibited data retention properties of 10<sup>4</sup> s at 85 °C<sup>[48]</sup>. In summary, research has indicated that excellent performance of ultrahigh switching speed, good switching endurance and reliable data retention has been achieved in metal oxide based resistive switching memory.

In this review, we focused on the metal oxide based RRAM. Although we limit to the metal oxide in this review, there are still some meaningful work needed to be summarized. Recent research on RRAMs still faces challenges such as wide resistance distribution and non-uniformity of SET/RESET voltage. Also, the physical mechanism in the SET/RESET process for RRAM is still unclear. Therefore, at first, the basic working principles of the device and the switching mechanism will be highlighted in Sections 2 and 3. Secondly, the effect of electrode materials and the structure of switching layers on the performance parameters including uniformity and power

\* Corresponding author. Ph.D.; Tel.: +86 551 63861793.

E-mail address: [ganghe01@issp.ac.cn](mailto:ganghe01@issp.ac.cn) (G. He).

\*\* Corresponding author. Ph.D.; Tel.: +86 27 88665004.

E-mail address: [nanoguy@126.com](mailto:nanoguy@126.com) (H. Wang).

consumption will be critically discussed in Sections 4 and 5. Finally, an outlook for RRAM application and its challenges current being faced such as high power consumption, operating stability and controversial RS switching mechanisms will be presented.

## 2. RS Behavior of the Metal Oxide Based Resistive Switching Memory: Unipolar and Bipolar

The RS characteristics of metal oxide RRAM can be classified into two switching modes: unipolar and bipolar. Unipolar RS behavior refers to the switching direction depending on the amplitude of the applied voltage rather than the polarity, as illustrated in Fig. 1(a). Bipolar RS behavior refers to the switching direction dependent on the polarity of the applied voltage, and thus the set process can only occur at one polarity and the reset process can only occur at reverse polarity, as shown in Fig. 1(b). The compliance current is an important parameter to be considered in the set process for both unipolar and bipolar RRAM. It is recommended to enforce a set compliance current to avoid a permanent breakdown of the device<sup>[49]</sup>. In addition, changing the value of compliance current and stop voltage can realize multilevel storage for the RRAM. Section 5 will describe it in detail<sup>[46]</sup>.

As for a unipolar RRAM, the device usually has a symmetric structure, which means that the same material has been used as the top electrode (TE) and the bottom electrode (BE), such as Pt/NiO/Pt<sup>[50]</sup>, Pt/TiO<sub>2</sub>/Pt<sup>[51]</sup>, Pt/ZnO/Pt<sup>[52]</sup>, Pt/ZrO<sub>2</sub>/Pt<sup>[53]</sup>, Pt/HfO<sub>2</sub>/Pt<sup>[54]</sup>. In contrast, the device structure of bipolar switching is usually asymmetric, which means that different materials are used as TE and BE, such as Pt/NiO/SrRuO<sub>3</sub><sup>[55]</sup>, Pt/TiO<sub>2</sub>/TiN<sup>[56]</sup>, TiN/ZnO/Pt<sup>[52]</sup>, Ti/ZrO<sub>2</sub>/Pt<sup>[57]</sup>, and TiN/HfO<sub>2</sub>/Pt<sup>[48]</sup>. For different switching modes, the reset mechanism is a controversial issue. Based on the conducting filament (CF) theory, it can be classified into two main CF disruption mechanisms: thermal dissolution model<sup>[26]</sup> and ionic migration model<sup>[58]</sup>. The former is often explored to explain the unipolar RS characteristics, whereas the latter can be normally applied to explain the bipolar RS characteristics. It is reported that the reset mechanism is greatly dependent on the electrode/oxide interfaces<sup>[38]</sup>. For unipolar RRAM, noble metals such as Pt or Ru are often applied as both top and bottom electrodes. On the other hand, the bipolar RRAM active materials such as TiN or Ti are normally used as electrodes, and the interfacial layer is easy to be formed in the operating process. Yu and Wong<sup>[59]</sup> proposed a universal reset mechanism for both unipolar/bipolar modes. Firstly, for the unipolar device, the mainstream viewpoint of reset mechanism is due to a thermal dissolution of CFs by local Joule heating<sup>[60–62]</sup>. Oxygen ions (O<sup>2-</sup>) would be

accumulated near the anode with a concentration gradient during the set process. Subsequently, the Joule heating would activate O<sup>2-</sup> to combine with the oxygen vacancies (V<sub>O</sub><sup>2+</sup>) during the reset process to make the CF disconnect. This thermal dissolution model results in the unipolar reset process. However, Yu et al.<sup>[63]</sup> put forward a question about this: if the CF is considered to be “dissolved” by local Joule heating, the CF may rupture in the middle of the filament where the temperature is the highest<sup>[64]</sup> rather than occur at the region near the anode.

As concerned as the bipolar device, the interfacial layer may act as an oxygen diffusion barrier, and thus the thermal diffusion mentioned above is not sufficient to result in the reset process. Only by the application of a reversed electric field can the oxygen ions drift back to cause the CFs to rupture. This is a so called ionic migration model. As the interfacial oxide layer was observed, Yu et al.<sup>[63]</sup> maintained that it may act as an “oxygen reservoir”, which stores the oxygen ions during the set process and then drives them back during the reset process. Besides, the “oxygen reservoir” can prevent the oxygen from escaping from the device to ambient. Therefore, this concept of an “oxygen reservoir” is envisioned, which can explain some switching characteristics for bipolar RRAMs. For example, in Fig. 2(a), the formed interface of ZrON can serve as an oxygen reservoir to hamper the oxygen ions’ diffusion further away from the RS layer HfOx and provide sufficient oxygen ions in the reset process, which is attributable to the robust cyclic endurance property of Pt/HfO<sub>x</sub>/ZrN<sub>x</sub> memory cell, as demonstrated in Fig. 2(b)<sup>[65]</sup>.

## 3. Physical Mechanism of Metal Oxide Based Resistive Switching Memory

The origin of RS behavior can be roughly classified into the conducting filament type and the interface type<sup>[66]</sup>. Here, we adopt the filament type to study the metal oxide based resistive switching memory, which considers the migration of metallic cations and oxygen vacancies. Accordingly, we discuss two main mechanisms: the electrochemical metallization effect (ECM) and the valence change memory effect (VCM).

In general, ECM can be adopted to explain RRAM which consists of an electrode made from an electrochemically active metal, such as Ag<sup>[19]</sup>, Cu<sup>[67]</sup>, or Ni<sup>[68]</sup>, and an electrochemically inert metal counter electrode such as Pt<sup>[69]</sup>, Au<sup>[70]</sup>, or Ir<sup>[71]</sup>. Yang et al.<sup>[19]</sup> studied Ag/ZnO:Mn/Pt memory cell and successfully observed nanoscale Ag conductive bridge penetrating through the storage thin film by scanning transmission electron microscopy (STEM) with high-resolution energy dispersive X-ray spectroscopy (EDX), which could account

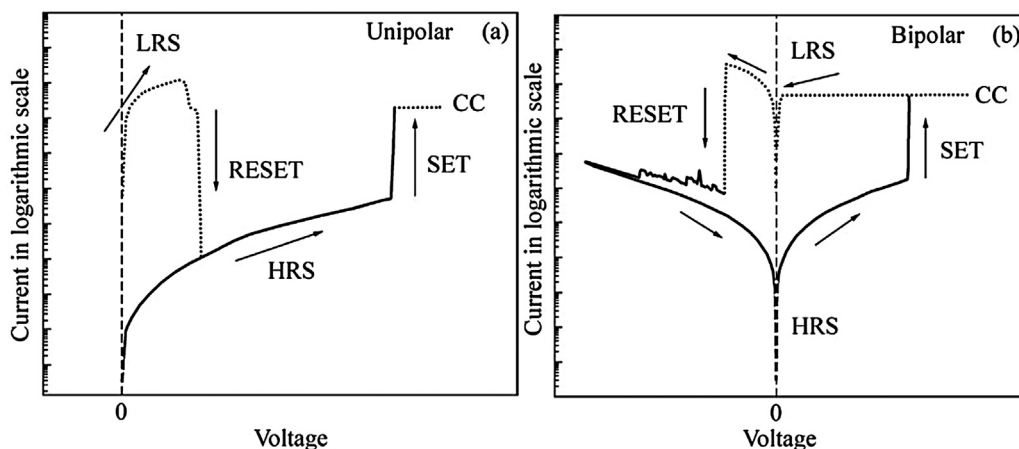


Fig. 1. Typical  $I$ - $V$  curves of unipolar (a) and bipolar (b) switching modes and the current compliance (CC) is adopted to avoid permanent dielectric breakdown of devices<sup>[49]</sup>.

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