



Effect of Cu ions assisted conductive filament on resistive switching memory behaviors in ZnFe₂O₄-based devices



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ARTICLE INFO

Article history:

Received 24 August 2016

Received in revised form

30 September 2016

Accepted 1 October 2016

Available online 5 October 2016

Keywords:

Cu ions

Memory device

Conductive filaments

Resistive switching

ZnFe₂O₄

ABSTRACT

Resistive random access memory (RRAM) has been developing as a most promising non-volatile memory in the current memory technology. In this work, ZnFe₂O₄ (ZFO) nano powder were firstly prepared by co-precipitation assisted hydrothermal process. Further, the resistive switching memory devices with Ag/Cu doped ZnFe₂O₄ (CZFO)/Au/Si and Ag/ZFO/Au/Si structures grown on silicon (Si) substrate were prepared by radio frequency magnetron sputtering, and their memory behaviors were contrastively investigated. It is observed the Cu doping can obviously affect the bipolar resistive switching memory characteristics. Thus a model concerning the formation and rupture of Cu ions assisted conductive filament inside the CZFO layer is suggested to explain the change of memory behaviors. These works provide a foundation for exploring the memory application of multifunctional material and further regulate their nonvolatile memory behaviors.

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1. Introduction

Current many electronic devices, such as mobile phones and computers, require increasingly high storage density memories for their operation [1]. Therefore, there is a great attention given to the exploration of new data storage approaches, which can increase storage capacity and reading/writing speed. Many conventional data storage technologies, such as dynamic random access memory (DRAM) and Flash, which is based on charge storage, are closing to their storage limits [1]. Therefore, there is an urgent need for development of new memory devices based on novel concepts. Among all of the emerging technologies, the resistive switching memory is one of the most promising candidates for next generation of logic and nonvolatile memory technology due to its simple structure, high storage density, low power and excellent scalability

[2–5]. At recently, resistive random access memory (RRAM) is defined as the reversible and non-volatile change of the resistance between high resistance state (HRS) and low resistance state (LRS) under applied electrical pulses [6,7]. Up to now, a great success and progress have made due to the development of new materials and structure design [8,9], and has become the basic phenomenon for development of novel non-volatile RRAM. It is known that the physical mechanism of bipolar resistive switching based on electrochemical metallization has been believed to be the formation and dissolution of conductive filaments in solid electrolyte, which is sandwiched by active electrode and inert counter electrode, and the growth of metal-elemental-based conducting filaments was clearly observed in As₂S₃ [10], Ag–Ge–Se [11], GeS [12], SiO₂ [13], ZrO₂ [14,15], TiO₂ [16] and ZnO [17,18], these materials have also been employed as oxide electrolytes in electrochemical metallization cell. In previous works, there are numerous mechanisms, including trap-controlled space charge limited current (SCLC) [19], ionic conduction [20], and metallic conduction filaments, have been proposed to explain the RRAM behaviors. But the conductive filament mechanism is widely used [21–23]. Generally speaking,

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there are two growth modes of the metal-elemental-based conductive filaments, one is the conducting filaments starting from active electrode and then growing towards counter electrode, and the other is the conducting filaments starting and growing inversely. The growth modes of the conducting filaments depend on the ions mobility and the microstructures of the oxide electrolytes. In addition, it has been reported that unipolar resistive switching, bipolar resistive switching, and transitions between unipolar resistive switching and bipolar resistive switching have been realized in the same sample under different electroforming conditions [24].

In recent reports, ZnFe_2O_4 , as an important member of spinel ferrites and chemically and thermally stable semiconductors, has recently been studied intensively for its wide applications including ferrofluid, photocatalyst, gas sensor, magnetic data storage, and so on [25–27]. In previous works, Hu et al. have reported that spinel-like ZnFe_2O_4 and MgZnO films exhibits superior resistive switching performance [28,29], which indicates spinel-like materials have also the potentiality as RRAM. This motivated us to explore whether normal spinel ferrite materials such as Cu doped ZnFe_2O_4 (CZFO) films as the resistive switching active layer to fabricate RRAM. In addition, ZFO have been prepared by many methods [30–32]. However, it is noteworthy that the hydrothermal method has such advantages as simple instrumentation, low cost, relatively low toxicity, abundant natural resources and easy manipulation [33,34]. Therefore, the development of facile synthetic methods for ZFO nano powder should be of great technical significance.

Although ZFO has been extensively studied but to the best of our knowledge, the resistive switching behaviors of Cu doped ZnFe_2O_4 (CZFO) have not been reported yet. Therefore, in this work, the ZFO nano powder was firstly prepared by a hydrothermal process, and the resistive switching behaviors of Cu doped ZFO (CZFO) and ZFO structures was further investigated.

2. Experiment process

2.1. Preparation of ZFO nano powder

ZFO nano powder were synthesized by a simple co-precipitation assisted hydrothermal method, which have been reported in the previous paper [35]. Raw materials and dosage for prepare ZFO is as follow: 0.0125 mol $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ and 0.05 mol $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ were dissolved in a proper quantity of deionized water, and continuously stirred for 5 h. Then proper amount 0.10 mol NH_4HCO_3 was added into the above solution, and stirred for 1 h again. Then it was transferred to a Teflon-lined stainless steel autoclave (50 mL), and it is heated at 180 °C for 24 h. The product was collected by centrifugation, washed with distilled water for few times until the PH value almost to be 7.0, dried in a vacuum at 80 °C overnight. Finally the powders were annealed at 600 °C for 2 h under the protection of nitrogen. Finally, we made one target of magnetron sputtering using as-prepared ZFO nano powder.

2.2. Preparation of Ag/CZFO/Au/Si and Ag/ZFO/Au/Si structure devices

ZFO and CZFO films were fabricated on Si substrates by radio frequency magnetron sputtering method at room temperature. We firstly deposited Au layer of ~120 nm on Si substrate. Then the ZFO and CZFO films were deposited and the power of the radio frequency was set at 80 W. The deposition rate of films was 0.01 nm s^{-1} . The typical base chamber pressure for the deposition was $5.0 \times 10^{-5} \text{ Pa}$ and the Ar working chamber pressure was 1.0 Pa. After ZFO and CZFO films deposition, the Ag electrodes with the

area of ~1 mm² were deposited on the same side for the electric measurement by direct current sputtering.

2.3. Material characteristics

Crystal structure of ZFO and CZFO films was characterized by X-ray diffraction (XRD) with Cu K α radiation at room temperature. Cross section morphology of films was characterized using scanning electron microscope (SEM). Energy dispersive X-ray spectroscopy (EDX) spectra of as-prepared films were observed by transmission electron microscopy (TEM) at an acceleration voltage of 200 kV. The chemical state of films was analyzed by X-ray photoelectron spectroscopy (XPS/ESCALAB250).

2.4. Device characteristics

In the test of resistive switching characteristics, Ag act the top and Au as the bottom electrodes, the experimental test circuit is shown in Fig. 1a. All the electric measurements were measured using the electrochemical workstation (CHI-660D) at room temperature.

3. Results and discussions

Fig. 1b exhibit the cross-sectional scanning electron microscope (SEM) images, which show the thickness of bottom electrode Au (~120 nm) and that of active layer (~150 nm) deposited on Si substrate.

Fig. 2a shows the X-ray powder diffraction (XRD) pattern of the MCZFO and ZFO layers at room temperature. The XRD profile of films matches very well with that in the reported works [36–38]. Obviously, no other impurity peaks are detected. In addition, the low background and sharp peaks suggest that the films retain their good crystallinity. We can see from the XRD that the diffraction patterns are identical for CZFO and ZFO, indicating the lattice structure of materials had not been changed after ZFO was doped with Cu. All diffraction peaks can be attributed to the pure CZFO and ZFO phase without trace of second phases, the (111), (220), (311), (422), (333), (440) peaks are all reflect the existence of the ZFO in a face-centered cubic (fcc) phase. We can find there is (200) peak of Cu for CZFO. At the same time, we can see the peaks are stronger and width of peaks is narrow, which indicates good crystallization of the product. Therefore, the crystallization is perfect for CZFO and ZFO. Fig. 2b and c shows the EDX spectrum of CZFO and ZFO. The EDX data confirm that the elements were Zn, Fe and O for ZFO and Cu, Zn, Fe and O for MCZFO without any other impurities. At the same time, the mass percentage of Cu is 4.32% for CZFO within experimental error.

XPS was used to further confirm the formation of CZFO film. As shown in Fig. 3a, the signals of Cu, Zn, Fe and O can be identified in the product, consistent with the formation of Cu doped ZnFe_2O_4 . The presence of carbon at 284.8 eV in the spectrum can be assigned to carbon contamination, and CO_2 adsorbed on the surface of the product when it was exposed to the air after synthesis. The signals at 1045.1 and 1022.4 eV can be attributed to Zn 2p_{1/2} and Zn 2p_{3/2} of Zn²⁺ [39]. There are four signals in the high-resolution spectrum of Fe 2p, as presented in Fig. 3d. The signals centered around 711.6 and 726.1 eV are caused by Fe³⁺ at octahedral sites. The satellite peaks, with binding energies 8 eV higher than the main peaks, confirm the oxidation state of iron is 3+ [39,40], consistent with the reported oxidation state (Fe³⁺) in ZnFe_2O_4 [40]. The O 1s peak can be found at 530.1 eV, which is characteristic of oxygen in metal oxides. The binding energy values of Cu 2p for the sample were found to be 936.7 eV. The obtained results obviously implied that Cu are present in CZFO film. The binding energy of Cu 2p_{3/2} is 934.8 eV, which is in

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