Conductive filaments controlled ferromagnetism in Co-doped ZnO resistive switching memory device

Sih-Sian Li¹ and Yan-Kuin Su^{1,2,3}

¹ Department of Photonics, National Cheng Kung University, No.1, University Road, East District,

Tainan 701, Taiwan

Phone: +886-6-275-7575 E-mail: SihSianLi@gmail.com (S. S. Li) and yksu@mail.ncku.edu.tw (Y. K. Su)

² Institute of Microelectronics, Department of Electrical Engineering, Advanced Optoelectronic Technology Center, National Cheng Kung University, Tainan 701, Taiwan

³ Department of Electrical Engineering, Green Energy Technology Research Center, Kun Shan University, Tainan 710, Tai-

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Abstract

Both the resistive switching characteristic and ferromagnetism of Co-doped ZnO device were investigated in this study. The bipolar resistive switching behavior has been observed, and the mechanism of device operation can be attributed to the defects. The M-H curves of the device with the resistive switching behavior in initial, high resistance, and low resistance states were analyzed, and all of the states show ferromagnetic. Interestingly, the strength of saturation magnetization in the device at the initial state and the low resistance state were found to be weakest and strongest, respectively. X-ray photoelectron spectra demonstrated that the presence of oxygen vacancies in the device, and the oxygen vacancies concentration at low resistance state was larger than that at high resistance state and initial state. Especially, the depth profiling XPS spectra show that the change of conductive filaments was consistent with resistance states and room temperature ferromagnetism. The results clearly demonstrated that the resistive switching effect accompanied with ferromagnetism might be attributed to the film defects.

1. Introduction

The resistive random access memory and magnetoresistive random-access memory have been investigated as a nextgeneration nonvolatile memory device, and both the electrical and magnetic properties have been comprehensive assessed. The resistive switching (RS) behavior has been attributed to oxygen vacancies (V_O)-based conductive filaments (CFs). Meanwhile, the origin of room temperature ferromagnetism (RT-FM) has also ascribed to the defects.

In recent years, the voltage-controlled saturation magnetization (Ms) change has been investigated.[1-3] However, no literatures reported that correlation between resistance states and RT-FM was analyzed via depth profiling XPS method. In this study, the CFs controlled Ms modification was investigated and discussed. To study the mechanism of the RS effect and to clarify the possible origin of RT-FM, we have prepared the Co-doped ZnO (CZO) RS device. This device was comprised of Pt/CZO/In-doped ZnO (IZO) multilayers structure, as shown in inset of Fig. 1. The physical properties of the device, such as electrical, magnetic, and chemical properties, was evaluated by utilizing extensively characterization techniques.

2. Experimental Results and Discussions

The Pt/CZO/IZO multilayer thin films were deposited by using a magnetron co-sputtering method in an ultra-high vacuum system. Figure 1 shows the current-voltage (I-V) curves of the device with a cell area of 40×40 µm recorded using dc sweeping mode. The bipolar RS behavior was observed in the CZO device. No forming process was occurred due to presence of rich V₀ in the as-deposited thin film. These V₀ contribute to a quick formation of CFs, leading to RS effect without a forming process. The set and reset voltages of CZO device were 1.13 V and -0.75 V, respectively. The on/off current ratios of CZO device were found to be 5.73 × 10², which assist to distinguish different storage information.



Fig. 1 Typical I-V curve of CZO device. The inset illustrates a schematic configuration of the CZO RS memory device.

In order to elucidate the correlation between different resistance states and RT-FM in the ZnO-based device, the M-H curves were measured at initial state (IS), high-resistance state (HRS), and low-resistance state (LRS), respectively. Figure 2 shows the M-H curves of device in the IS, HRS, and LRS at RT. FM signal of a very weak Ms was about 0.2 emu/cm³ in the resistance state at the IS. As the device was switched to HRS and LRS, an increase of the Ms has been observed at HRS (0.79 emu/cm³) and LRS (1.92 emu/cm³), thereby indicating the lowest and highest Ms corresponding to the RS state of IS and LRS, respectively.



Fig. 2 The M-H curves of the CZO device with the RS behavior at IS, HRS, and LRS.

Figure 3 shows the O 1*s* state XPS spectra of the device at IS, HRS, and LRS. The asymmetric peak of O 1*s* state in device was analyzed by using Gaussian-fitting. Clearly, all of the signals were comprised of three peaks at about 529.7 eV, 530.9 eV, and 532.3 eV. The peak value at 529.7 eV has been allocated to the O²⁻ ions in a wurtzite lattice structure surrounded by Zn²⁺ ions, while the peak value at 532.3 eV has been assigned to O-H or O-O bonds, such as surface adsorbed H₂O or O₂. Additionally, the peak value of the O 1*s* state at 530.9 eV has been attributed to the existence of abundant V₀ in the thin film. It is worth noting that both the intensity and integral area of V₀-related peak at LRS were larger than that at HRS and IS. This results demonstrated that the V₀ concentration of device at LRS was higher than that at HRS and IS.

In order to verify the chemical bonding states further, depth-profiling XPS spectra were examined at different depths of thin film. Figure 4(a) shows the XPS spectra of O 1*s* state of device at HRS after etching for 1, 2, 3 minutes. This signal includes two peaks at 529.7 eV (O^{2-}) and 530.9 eV (V_0). Noting that the integral area of the peak at 530.9 eV increased with increasing etching time. On the other hand, at LRS, the integral area of the peak at 530.9 eV was no obvious change for all of the signals, as shown in Fig. 4(b).



Binding energy (eV)

Fig. 3 O 1s XPS spectra of CZO device with the RS behavior at IS, HRS, and LRS.

As the literature mentioned, the magnetic moments of ZnO could originate from the V₀. For the XPS results, the V₀-related peak intensity and integral area corresponding to LRS was higher than that in HRS, demonstrating that an increase of the V₀ concentration was occurred at LRS by the set process. The RS effect of CZO device due to the formation of V₀-based CFs was highly corroborated via the depth-profiling XPS spectra. In addition, the Ms was increased with switching to LRS in the CZO device. The variation of Ms was closely followed the change of resistance states. Therefore, this results evidenced that the set process increases the V₀ concentration, leading to an enhancement of RT-FM at LRS.



Fig. 4 The depth-profiling XPS spectra of O 1s state of CZO device at (a) HRS and (b) LRS after different etching times.

3. Conclusions

The CFs controlled Ms switching in the CZO RS memory device was investigated in this study. Clearly, the mechanism of RS effect and RT-FM have been attributed to the V₀-based CFs. Our device not only shows excellent bipolar RS behavior, but also exhibits RT-FM, and the highly stability and reliability have been found. Especially, the magnitude of Ms value can be controlled via different resistance states. The results evidently demonstrated that the defect is a key factor that controls Ms and CFs change in CZO RS device for novel spintronic device applications in the future.

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