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Improved Magnetism in Mn-doped ZnO Thin Films by Inserting ZnO Layer

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Abstract. Compared to Ti/Mn doped ZnO/Pt (Ti/MZO/Pt) device, the Ti/MZO/ZnO/Pt device exhibits more excellent resistive switching (RS) performances and larger magnetic variation behaviors. The RS behavior is attributed to formation and rupture of oxygen vacancy (Vo) based conductive filaments (CFs), and magnetic variation was demonstrated by bound magnetic pole (BMP) model. Here ZnO buffer layer contribute to improve the RS and magnetism.

1. Introduction

Extensive studies have been investigated ferromagnetism (FM) at room temperature in many transition metal (TM) doped and undoped binary metal oxide structures, such as ZnO, NiO, TiO_2 [1-3]. However their room temperature FM is still too weak to practically apply in spintronics. It is generally believe that oxygen vacancies (Vos) played an important role in the FM of binary metal oxides. Many techniques, such as annealing in argon or vacuum and doping TM [4], have been used to enhance Vo density. Unfortunately those methods have no significant improvements in FM at room temperature. Recently it was reported that V_Os migrate easily in binary oxides under an electric field, generating Vo-based conductive filaments (CFs) during resistive switching (RS) process [5]. It is the Vos with high density in CFs that coupled each other, resulting in a large magnetism in a resistive state [6, 7]. It has been proven that inserting oxide buffer layer between metal electrodes and insulator is an effective method to improve the RS performances [8, 9]. So, it is interesting to study magnetism by inserting buffer layer. In this paper, we fabricate Ti/Mn doped ZnO/Pt (Ti/MZO/Pt) and Ti/MZO/ZnO/Pt memory devices for investigating the RS performances and magnetic variation.

2. Material and Methods

The 4 at.% Mn doped ZnO films (MZO films) with and without a ZnO buffer layer were deposited on a commercially available $Pt/Ti/SiO_2/Si$ substrate at 400°C using pulsed laser deposition (PLD) under an oxygen pressure of 5 Pa. The thicknesses of MZO films about 115 nm. ZnO buffer layers were deposited at 400 °C under a background pressure of 1.0×10^{-5} Pa in vacuum atmosphere. Top electrodes (TE) of Ti (diameter 200μm) were deposited through a metal shadow mask onto the MZO films by using magnetron sputtering.

The crystalline structure was determined by X-ray diffraction (XRD, X'pert PRO MPD). The current-voltage (I-V) characteristics were performed using a Keithley 2612A sourcemeter, and Schematic of the device layout and the current-voltage (I-V) measurement configuration, as shown Figure 1 (a). Hysteresis loops (M-H curves) at room temperature were measured by Quantum Design PPMS-6700.

3. Results and Discussion

Figure 1 (b) shows XRD spectra of the MZO films with and without a ZnO buffer layer. It can be seen that a sharp (002) peak of ZnO wurtzite structure appear in both the as-prepared films except for the diffraction peak form the substrate, and no peaks related to Mn metal was determined. Compared to MZO films, the crystalline quality of MZO films with a ZnO buffer layer (MZO/ZnO) have improved.

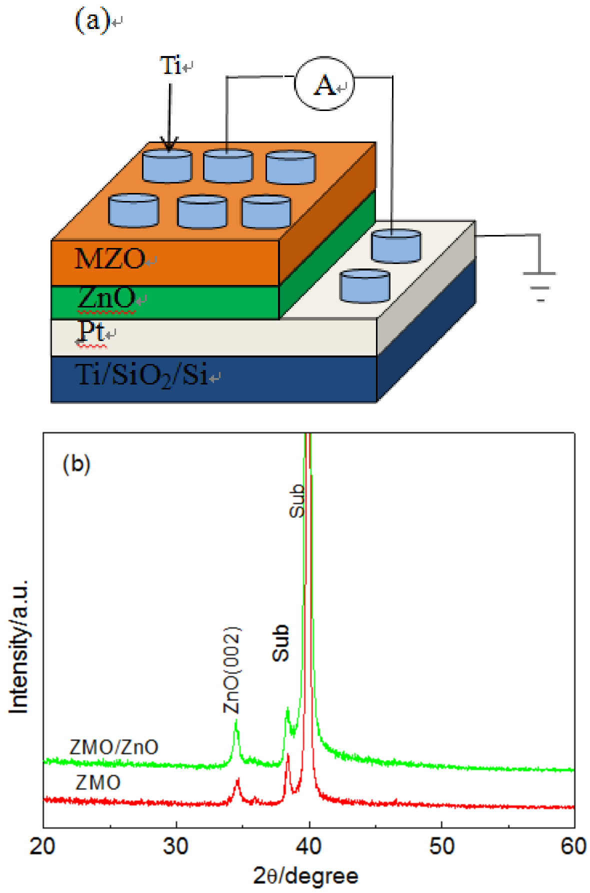

Figure 1. (a) Schematic of the device layout and the current-voltage (I-V) measurement configuration. (b) XRD patterns of the MZO films with and without a ZnO buffer layer

Figure 2 (a) and (b) shows typical I-V curves of MZO and MZO/ZnO film. Compared to Ti/MZO/Pt device, the Ti/MZO/ZnO/Pt device presents more stable and larger switching ratio. The top inset in Figure 2 (a) and (b) shows distributions of the set (V_{SET}) and reset (V_{RESET}) voltage. For Ti/ZMO/Pt device, the V_{SET}/V_{RESET} varies from 1.08V/-0.76V to 1.72V/-0.96V, and the ΔSET/ΔRESET reach up to 0.64 V and 0.2 V, respectively. Compared to the above Ti/ZMO/Pt device without ZnO inserting layer, the V_{SET} /V_{RESET} was small in the Ti/ZMO/ZnO/Pt device, changing from 0.49V/-0.50V to 0.52V/-0.54V, and ΔSET/ΔRESET are only 0.03V/0.04V. Obviously, MZO/ZnO bilayer films displays centralized distribution of V_{SET} and V_{RESET}. Figure 2 (c) and (d) shows the endurance of the above device with 300 cycles. It should be noted that the R_{HRS}/R_{LRS} ratio of Ti/MZO/ZnO/Pt device approximately attained 133, which is far higher than 3 of Ti/MZO/Pt device without a ZnO buffer layer. According to the sudden jump at V_{SET} and V_{RESET} in the both devices, it can be speculated that RS behavior may originate from the formation and fracture of CFs [10, 11]. It is confirmed by log-log scale plot in low resistance state (LRS), as shown in the Figure 2 (e) and (f). The fitting curve display Ohmic behavior with a slope close to 1 in the LRS.

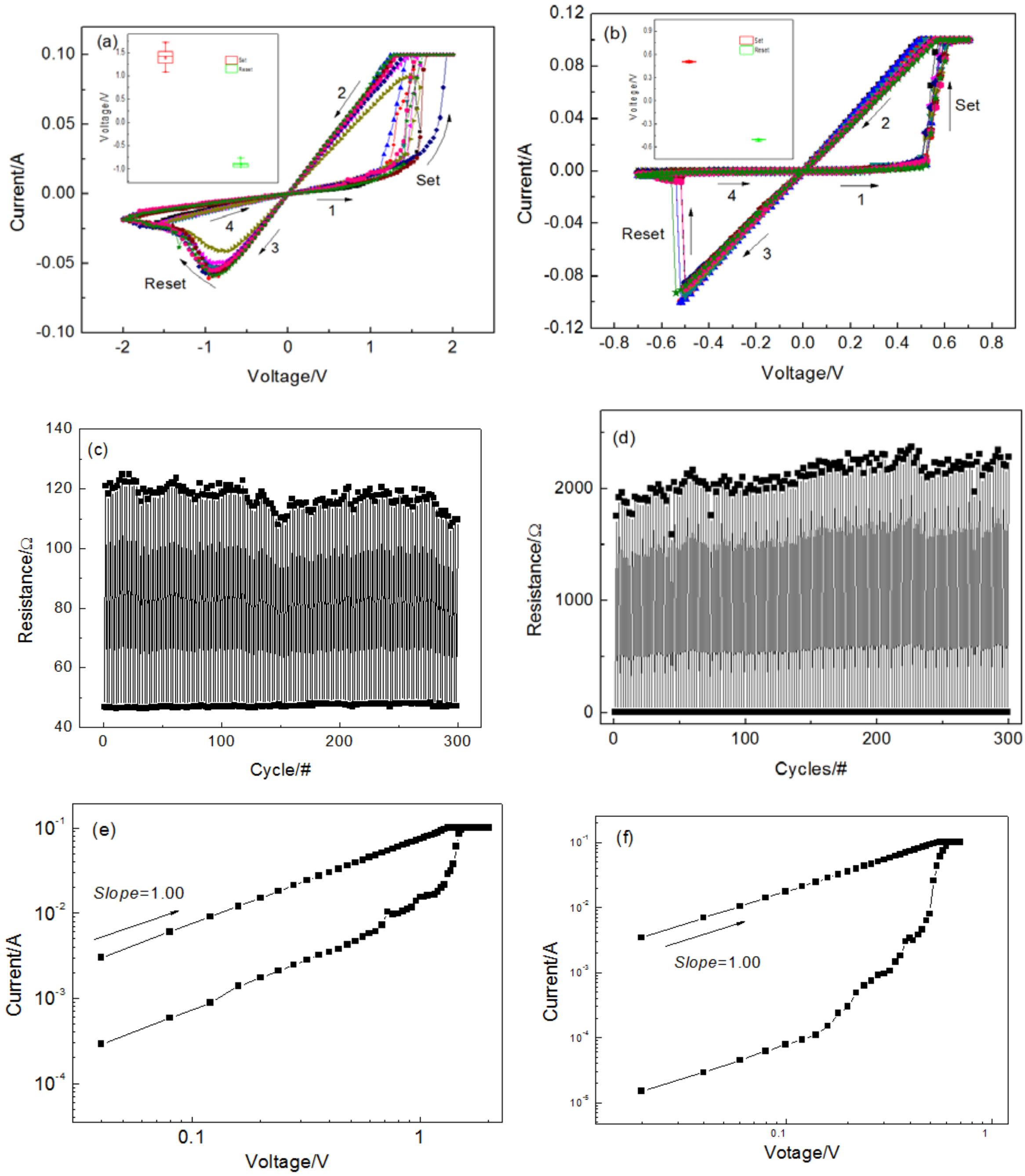

Figure 2. Typical I-V curves of Ti/ZMO/Pt (a) and (b) Ti/MZO/ZnO/Pt devices, endurance of Ti/ZMO/Pt (c) and Ti/MZO/ZnO/Pt (d) devices at a read voltage of +0.5 V; the Log-Log plot I-V for Ti/ZMO/Pt (e) and Ti/MZO/ZnO/Pt (f) devices.

To clarify the change of magnetism in the process of RS process, the M-H curves of MZO and MZO/ZnO films in the initial state (IS), LRS and HRS are determined at room temperature, as shown in Figure 3 (Signal from the substrates have been subtracted from the all M-H curves). The saturation magnetism (M_S) are only 0.87 μemu (in the IS), 2.47 μemu (in the HRS) and 3.93 μemu (in the LRS) for Ti/ZMO/Pt device without a ZnO buffer layer. While for Ti/ZMO/ZnO/Pt device with a ZnO buffer layer, the M_S value increase to approximately 2.44 μemu (in the IS), 6.00 μemu (in the HRS) and 16.08 μemu (in the LRS), respectively. Especially the ratio of M_{LRS}/M_{IS} reached up to 8.51, which is much higher than 4.78 of Ti/ZMO/Pt device. The above results show that ZnO buffer layer is favorable for improving magnetism and it variation of MZO film during the RS process.

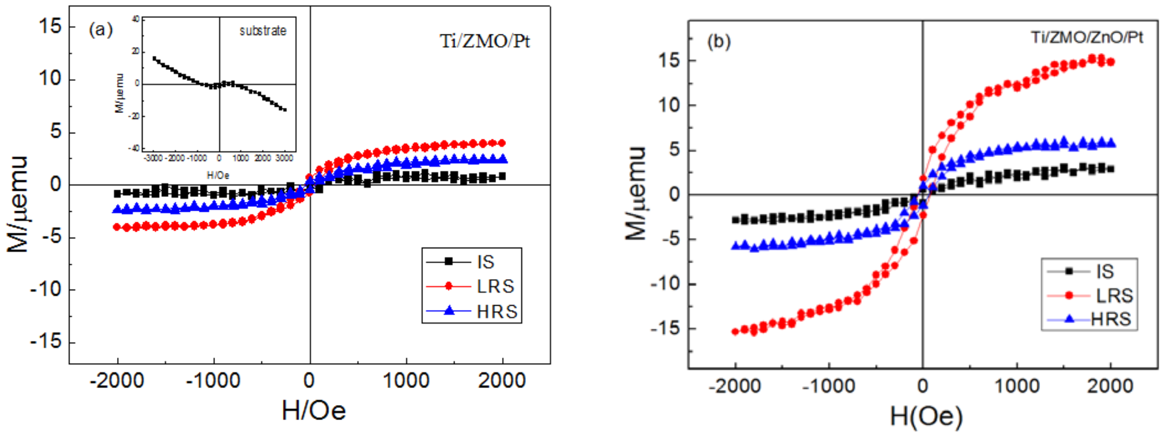

Figure 3. M-H curve of Ti/ZMO/Pt device (a) and substrate (inset) and Ti/ZMO/ZnO/Pt device (b) in the IS, HRS, and LRS.

The main view put forward at present is the bound magnetic pole (BMP) model about origin of ferromagnetism in binary metal oxide [7, 12]. The neighboring BMPs are overlapped to form BMPs clusters, thus showing the macroscopic ferromagnetism. In this paper, ZnO buffer layer was deposited in no-oxygen ambient, which is equivalent to introduce enough Vos which increase the thickness (or Vo density) of the formed CFs in the MZO films in the LRS. This makes more BMPs clusters overlapped each other, resulting in a stronger FM at room temperature.

4. Conclusions

In summary, Mn doped ZnO-based resistive switching devices with and without ZnO buffer layer are demonstrated in this paper. Compared to Ti/MZO/Pt device without ZnO buffer layer, the Ti/MZO/ZnO/Pt device exhibits more excellent resistive switching performances and larger magnetic variation behaviors. The R_{HRS}/R_{LRS} ratio (approximately 133) of Ti/MZO/ZnO/Pt device is far higher than 3 of Ti/MZO/Pt device, and the ratio of M_{LRS}/M_{IS} of the former reached up to 8.51, which is much higher than 4.78 of the latter. The ZnO buffer layer plays a prominent role in resistive switching and magnetic variation. We speculate the main reason is that buffer layer contribute to increase Vo density in the CF_S as well as reduce the randomness of filament formation.

5. References

[1] ChihChieh Hsu, WeiChieh Ting and YuTing Chen 2017 Effects of substrate temperature on resistive switching behavior of planar ZnO resistive random access memories *J Alloy Compd* 691 pp. 537-44 DOI: 10.1016/j.jallcom.2016.08.248
[2] GyoungHo Buh, Inrok Hwang and Bae Ho Park 2009 Time-dependent electroforming in NiO resistive switching devices *Appl Phys Lett* 95 (14) 142101 DOI: 10.1063/1.3242337
[3] Senthilkumara V, Kathalingama A, Kannana V, Karuppanan Senthilb and Jin-Koo Rheea 2013 Reproducible resistive switching in hydrothermal processed TiO_2 nanorod film for non-volatile memory applications *Sensor Actuat A-Phys* 194 pp. 135-139 DOI: 10.1016/j.sna.2013.02.009
[4] Yumin Hu, SihsSian Li, CheinHsiun Kuang, TaiChun Han and ChinChung Yu 2015 Post-annealing effect on the room-temperature ferromagnetism in Cu-doped ZnO thin films *J Appl Phys* 117 (17) 17B901 DOI: 10.1063/1.4906527
[5] H. Y Peng, G. P Li, J. Y Ye, Z. P Wei, Z Zhang, D. D Wang, G. Z Xing and T. Wu 2010 Electrode dependence of resistive switching in Mn-doped ZnO: Filamentary versus interfacial mechanisms *Appl Phys Lett* 96 (19) 192113 DOI: 10.1063/1.3428365
[6] C. Chappert, A. Fert and F. N. Van Dau 2007 The emergence of spin electronics in data storage

Nat Mater 6 pp. 813-23 DOI: 10.1038/nmat2024
[7] G. Chen, C. Song, C. Chen, S. Gao, F. Zeng and F. Pan 2012 Resistive switching and magnetic modulation in cobalt-doped ZnO Adv Mater 24 (26) pp. 3515-20 DOI: 10.1002/adma.201201595
[8] D. C. Kim, M. J. Lee, S. E. Ahn, S. Seo and J. C. Park 2006 Improvement of resistive memory switching in NiO using IrO_2 *Appl Phys Lett* 88 (23) 232106 DOI: 10.1063/1.2210087
[9] F. Yang, M. Wei and H. Deng 2013 Bipolar resistive switching characteristics in CuO/ZnO bilayer structure *J Appl Phys* 114 (13) 134502 DOI: 10.1063/1.4821237
[10] Nuo Xu, Lifeng Liu, Xiao Sun, Xiaoyan Liu and Dedong Han 2008 Characteristics and mechanism of conduction/set process in TiN∕ZnO∕Pt resistance switching random-access memories *Appl PhysH Lett* 92 (23) 232112 DOI: 10.1063/1.2945278
[11] Jian Zhang, Hui Yang, Qilong Zhang, Shurong Dong and J. K. Luo 2013 Bipolar resistive switching characteristics of low temperature grown ZnO thin films by plasma-enhanced atomic layer deposition *Appl Phys Lett* 102 (1) 012113 DOI: 10.1063/1.4774400
[12] Daqiang Gao, Yan Xu, Zhaohui Zhang, Hua Gao and Desheng Xue 2009 Room temperature ferromagnetism of Cu doped ZnO nanowire arrays *J Appl Phys* 105 (6) 063903 DOI: 10.1063/1.3079509