Resistive switching in two-terminal HfO₂/SiO₂ stack with interface dipole modulation

Noriyuki Miyata^{1,*}, Kyoko Sumita¹, Akira Yasui², Reito Wada³, and Hiroshi Nohira³

¹National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki, Japan ²Japan Synchrotron Radiation Research Institute (JASRI), Sayo-gun, Hyogo, Japan ³Tokyo City University, Setagaya-ku, Tokyo, Japan

*Phone: +81-29-861-2511 E-mail: nori.miyata@aist.go.jp

Abstract

We propose a new metal–insulator–metal (MIM) resistive switching device using a HfO₂/SiO₂-based interface dipole modulation (IDM). Rectification characteristics useful for selector function and resistance change were demonstrated by the IDM MIM stack with an asymmetric tunnel barrier. *In situ* hard x-ray photoelectron spectroscopy (HAXPES) was applied to investigate the IDM MIM structure under bias conditions.

1. Introduction

The interface dipole modulation (IDM) observed from the HfO₂/SiO₂ stack structure has been reported to be applicable as a 3-terminal flash type memory device [1]. On the other hand, incorporating such a potential modulation into a metal-insulator-metal (MIM) structure is expected to lead to a two-terminal resistance change device. One of the advantages of two-terminal devices is the high-density cross-point memory array architecture. In this case, a selector device is required for suppressing the snake currents flowing through the adjacent memory cells. Various selector devices have been proposed, and one of them is a rectifier such as a p/n junction or MIM diode [2, 3]. In this paper, we introduce an idea of an IDM-based MIM device that achieves both resistance change and rectification. We report the electrical and physical characteristics of the fabricated IDM MIM stack structures.

2. Device concept and fabrication

A HfO₂/SiO₂/HfO₂ stack is investigated, where a monolayer (ML) TiO₂ modulation layer is inserted at the bottomside SiO₂/HfO₂ interface [Fig 1 (a)]. When a positive bias is applied to a top electrode (TE), the tunnel current is expected to increase because the potential barrier near the bottom-side SiO₂/HfO₂ interface decreases due to the IDM effect. Under the negative bias conditions, electrons tunnel through the topside HfO₂/SiO₂ barrier. Choosing a thicker top-side HfO₂ layer than the bottom-side HfO₂ layer is expected to reduce tunnel current compared to the positive bias conditions. This means that resistance change and rectification can be achieved in the same MIM stack.

The 4-nm HfO₂/2.5-nm SiO₂/1-ML TiO₂/1.5-nm HfO₂ stack was prepared on TiN bottom electrode (BE) by electron-beam evaporation [1]. The post deposition annealing (PDA) was performed at 350°C, and then Ir TEs were fabricated. For the photoelectron spectroscopy measurements, IDM stacks were fabricated on TaN BEs. The hard x-ray photoelectron spectroscopy (HAXPES) measurements were performed using synchrotron radiation (hv=7940 eV) at BL47XU at SPring-8 [4].

3. Results and discussion

3. 1. Electrical characteristics of IDM MIM

The current-voltage (I-V) curves of the fabricated IDM MIM cell exhibit the resistance change in the positive bias range [Fig. 2 (a)], as expected. Under this bias sweep condition, more than one-digit difference can be obtained. The current in the negative bias range is small and increases slightly below -3 V. This current change is opposite of the positive bias range and is consistent with the IDM operation. The current change and rectifying characteristics are stable under the cyclic *I-V* measurements [Figs. 2 (a) and (b)].

The above *I-V* characteristics can be observed for virgin cells, and the forming process that is usually required for conductive filament type memory is not required for the IDM device. The current difference (Δ I) between the SET and RE-SET states depends on the bias sweep range [Figs. 3 and 4]. This tendency is consistent with the previously proposed IDM mechanism [1], *i.e.*, the IDM magnitude gradually changes according to the electric field. This analog-like resistance change is expected to be useful in neuromorphic applications.

3. 2. XPS and HAXPES measurements

The key structural element in the TiO_2 -modulator-based IDM was reported to be Ti-O bonding [5]. Even though IDM oxide stacks on metal electrodes, we were able to confirm that the Ti 2p suboxide component could be suppressed after the appropriate PDA [Fig. 5]. Thus, we can reasonably expect that IDM occurs in the fabricated IDM MIM cells.

Figure 6 shows the HAXPES results obtained from the IDM MIM stack, where TE was connected to ground. By applying the BE bias, Ta 3*d* photoelectron spectrum shifts significantly and the Hf 3*d* spectra also show slight shifts. These characteristics can be reasonably explained by the potential changes in the MIM stack. Figure 7 shows Hf 3*d* spectra with and without bias, which can be explained by the band diagrams shown in the insets. This result suggests that the Hf sub-oxide components are negligible. Therefore, we believe that the resistance change caused by oxygen vacancy of HfO₂ is not the major switching mechanism of the IDM MIM cells.

4. Conclusions

We proposed a unique approach which provides both resistive switching and rectification function in the same MIM stack cell. We have fabricated MIM cells with the HfO₂/SiO₂ IDM stack and confirmed both characteristics. Photoelectron spectroscopy studies support that the intended IDM MIM stack can be fabricated.

Acknowledgements

This work was supported by JSPS KAKENHI Grant Number 19H02178. Part of the device fabrication was conducted at the AIST Nano-Processing Facility (AIST-NPF). The experiments were partly performed at SPring-8 with the approval of the program review committee (2019A1552).







Fig. 4 Effect of maximum applied voltage (V_{app}) on current difference (ΔI) between SET and RESET states.



Fig. 6 In situ HAXPES measurement of $Ir/HfO_2/1-ML TiO_2/SiO_2/TaN$ MIM stack under bias conditions.

References

[1] N. Miyata, Sci. Rep. 8, 8486 (2018).

HfO₂/SiO₂/HfO₂ IDM MIM stack

for different bias sweep widths.

- [2] J. S. Meena, S. M. Sze, U. Chand and T. -Y. Tseng, *Nanoscale Res. Lett.* 9, 526 (2014).
- [3] N. Alimardani and J. F. Conley, Appl. Phys. Lett. 102, 143501 (2013).
- [4] N. Miyata, J. Nara, T. Yamasaki, K. Sumita, R. Sano, and H. Nohira, IEEE International Electron Devices Meeting (IEDM), 7.6.1 (2018).
- [5] N. Miyata, Appl. Phys. Lett. 113, 251601 (2018).



Fig. 2 Current change and rectification characteristics of $HfO_2/SiO_2/HfO_2$ IDM MIM stack. (a) Cyclic *I-V* curves and (b) SET and RESET currents at +1.5 V.



Fig. 5 Ti 2p photoelectron spectrum of SiO₂/1-ML TiO₂/HfO₂ stack on TaN electrode.



Fig. 7 Hf 3*d* photoelectron spectra and predicted band diagrams.