High on/off ratio (>10) ferroelectric tunnel junctions (FTJs) with ultrathin Y-doped HfO₂

Xuan Tian¹, Shigehisa Shibayama¹, Tomonori Nishimura¹, Takeaki Yajima¹, Shinji Migita² and Akira Toriumi¹

¹Department of Materials Engineering, The University of Tokyo, Tokyo 113-8656, Japan

²National Institute of Advanced Industrial Science & Technology (AIST), Tsukuba 305-8569, Japan

Phone: +81-3-5841-1907, E-mail: xuan@adam.t.u-tokyo.ac.jp

Abstract

We have investigated ultrathin ferroelectric HfO_2 for demonstrating high performance ferroelectric tunnel junctions. With the decrease in HfO_2 thickness below 5 nm, ferroelectricity was reduced gradually, while the tunneling current was exponentially enhanced. We found an optimum thickness to enhance rather high on/off ratio (>10), keeping a sufficient tunneling current. To further enhance the on/off ratio, it is required to understand the polarization reduction mechanism in 2-nm-thick region.

1. Introduction

Ferroelectric tunnel junction (FTJ) is a promising nonvolatile resistive memory in terms of fast operation speed, high density, low power consumption, and non-destructive reading process [1, 2]. Ferroelectric HfO₂ has provided a new opportunity in FTJ memories in terms of CMOS friendly material and process. However, HfO₂-based FTJs suffered from a small on/off ratio due to the poor remnant polarization [3] or a large operating voltage due to an additional interfacial layer [4]. Besides, for the practical application of FTJ devices, HfO₂ thickness needs to be ultra-thin to get a large reading current for high access speed, while the on/off ratio should keep high for sufficient reading margin. In this work, a high performance FTJ with ultrathin HfO₂ film is demonstrated.

2. Experiment

Heavily doped p-type Ge (N_A = 5×10¹⁸cm⁻³) was used as the bottom electrode. Y-doped HfO₂ films were prepared by co-sputtering of Y₂O₃ and HfO₂ on Ge substrate, then TiN was sputtered on HfO₂ as the top electrode. The stack was processed by post metallization annealing (PMA) at 700°C in N₂ for 30s to crystallize ultrathin HfO₂ films. Different sizes of electrodes were fabricated with the standard lithography process. Ferroelectric properties of all films were measured by the ferroelectric analyzer with Q-V mode at 10 kHz, and the double pulse measurements. I-V characteristics of HfO₂ FTJs were measured by the semiconductor analyzer at two different temperatures.

3. Results and Discussion

The evolution of switching polarization (P_{sw}) with HfO_2 thickness down to 5 nm has been reported in previous work [5]. In the present work, P_{sw} of Y-doped HfO_2 film gradually drops below a critical thickness around 5 nm, as shown in **Fig.**

1 (a). The ferroelectric polarization almost disappears below 2.4 nm. By reducing HfO₂ thickness, the reading current density at 0.2 V exponentially increases as shown in Fig. 1 (b). The double pulse measurement was adopted to accurately determine the ferroelectricity in ultrathin HfO₂ [5, 6]. Although the leakage current (I_L) is larger in thinner HfO₂ film as noted by the arrows in Fig. 1 (c, d), the double pulse measurement helped clearly distinguish the polarization current (I_P) from I_L, as shown in Fig. 1 (d). P_{sw} in 2.7 nm HfO₂ was around 6.9 μ C/cm² from the integration of I_P.



Fig. 1 (a) The evolution of P_{sw} with HfO₂ thickness. (b) Reading current density at 0.2 V in different thickness HfO₂ device. Double pulse measurement results of (c) 3.3 nm and (d) 2.7 nm Y-doped HfO₂ films.

The resistive switching behaviors of HfO₂-based FTJs are shown in Fig. 2. A relatively thick (3.3 nm) HfO₂ was firstly investigated. Two resistive states are demonstrated in a round-trip DC voltage sweeping in Fig. 2 (a). Before voltage sweeping, the device was written with -1.8 V, therefore the polarization direction was switched up, pointing to the top TiN electrode. Afterwards, voltage swept from -1.8 V to 1.8 V, leading to the high resistive (off) state in the I-V characteristic, then swept back to -1.8 V, resulting in a low resistive (on) state. The on/off ratio (defined by Ion/Ioff read at 0.2 V) was around 2 in the 3.3 nm HfO₂ FTJ. To achieve larger tunneling current for higher access speed in FTJ application, HfO₂ thickness was reduced to 2.7 nm, while a good ferroelectric polarization with $P_{sw} \sim 7 \ \mu C/cm^2$ was sustained. Two resistive states are clearly demonstrated by the DC voltage sweeping as noted by the arrows in Fig 2 (b).

Comparing to Fig. 2 (a), the tunneling current is considerably increased by two orders of magnitudes, and the on/off ratio is also largely enhanced to be over 10. Besides, the device can be written in 1.6 V and read by 0.2 V, showing a significant merit of low power consumption.



Fig. 2 I-V characteristics of (a) 3.3 nm and (b) 2.7 nm HfO₂ FTJ under DC voltage sweeping. Two colors represent results from different junctions. Arrows denote the voltage sweeping direction.

The ferroelectricity dependent device properties were investigated in 2.7 nm HfO₂ FTJs by varying P_{sw} of HfO₂, where the Y doping concentration was intentionally changed. The on/off ratio was found to be proportional to P_{sw} of HfO₂, as shown in Fig. 3 (a). This indicates that the resistive switching is determined by the ferroelectric strength in HfO₂ film. Next, the off state tunneling current was measured on different sizes of electrodes. Approximately the same current density was obtained in different electrode areas, as shown in Fig. 3 (b), which suggests a uniform current flow under an electrode. This fact excludes the possibility of filamentinduced resistive switching. I-V characteristics of 2.7 nm HfO₂ FTJs were measured at 300 K and 150 K. A similar resistive switching behavior with a stable on/off ratio is observed at both temperatures in Fig. 3 (c). The decrease of the tunneling current at 150 K indicates some temperaturedependent current flow process, like the thermionic or the Poole-Frenkel emission processes, might be involved. However, the ferroelectric tunneling performance is not affected for FTJ applications at room temperature.



Fig. 3 (a) The relationship between on/off ratio and P_{sw} . (b) The offstate current measured on different sizes of electrodes. (c) The I-V characteristics of 2.7 nm HfO₂ FTJ measured at 300K and 150 K.

The thickness dependence of reading current density and the corresponding on/off ratio in HfO₂ FTJs are summarized in **Fig. 4**. While reducing HfO₂ thickness from 3.3 nm to 2.7 nm, the reading current exponentially increases, and the on/off ratio is largely enhanced. The on/off ratio, however, dramatically drops as the HfO₂ thickness is further lowered. The P_{sw} remarkably decreases at the same time as shown in **Fig. 4 (a)**. This indicates that the marginal on/off ratio results from the substantially decreased Psw in thinner HfO2. Therefore, a trade-off occurs between large reading current (high access speed) and large reading margin (high on/off ratio) in FTJs with ~2 nm HfO2. Sustaining the good polarization intensity in $\sim 2 \text{ nm HfO}_2$ is the key to avoiding the decreased on/off ratio while achieving further higher tunneling current. Further study is necessary to understand this problem by considering the ferroelectric orthorhombic phase stability in ultrathin HfO₂ from the interface effect, which influences the depolarization field, or the film processing effect, like doping, annealing, and so on. On the other hand, the on/off ratio can be effectively enhanced by taking advantage of the electrode effect, that is, using asymmetric electrodes with widely different screening length [2, 7] in FTJ devices. Therefore, an appropriate selection of the electrode by taking account of the electron density, the electrode / HfO2 interface effect etc., will enable to further enhance the on/off ratio.



Fig. 4 (a) The HfO₂ thickness dependence of current density read at 0.2 V (black), and the P_{sw} (red). (b) The evolution of on/off ratio in HfO₂ FTJs with different ferroelectric layer thicknesses.

4. Conclusions

A high performance HfO_2 FTJ has been demonstrated for practical applications with sufficient tunneling current, low operation voltage, and large on/off ratio (>10). The on/off ratio, however, becomes marginal when further reducing HfO_2 thickness below 2.7 nm due to the substantially decreased P_{sw} in HfO_2 film. Finally, it is expected that the tradeoff between the tunneling current and the on/off ratio observed in FTJ devices with around 2 nm HfO_2 will be further improved to thinner region by employing appropriate electrode in addition to improving the processing conditions of HfO_2 .

Acknowledgement

This work was supported by JST-CREST Project (JPMJCR14F2).

References

- [1] A. Chanthbouala, et al., Nat Nanotechnol, 7, 101 (2012).
- [2] Z. Wen, et al., Nat. Mater., 12, 617 (2013).
- [3] A. Chouprik, et al., Microelectron. Eng., 178, 250-253, (2017).
- [4] S. Fujii et al., VLSI, 2016.
- [5] X. Tian, et al., Appl. Phys. Lett., 112, 102902 (2018).
- [6] X. Tian, et al. IEDM 2017, pp. 37.1.1-37.1.4.
- [7] H. Lu, et al. Nat. Commun. 5, 5518, (2014).