# Performance improvement by template-induced crystallization in ferroelectric HfO<sub>2</sub> tunnel junction memory for cross-point high-density application

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#### Abstract

We experimentally investigate impacts of template layer (TL) insertion on memory characteristics of ferroelectric  $HfO_2$  tunnel junction memory. The template layer reduces crystallization temperature and modulates crystal orientation of the  $HfO_2$ , which improves controllability of interlayer thickness and increases effective polarization perpendicular to the stack. As a result, simultaneous improvement of operation voltage, data retention and disturb immunity was successfully achieved, demonstrating the excellent suitability to cross-point high-density array operation.

## 1. Introduction

Discovery of ferroelectricity in HfO<sub>2</sub> opened a new world thanks to its functionality and CMOS compatibility. One of the promising applications is HfO<sub>2</sub>-based ferroelectric tunnel junction (FTJ) memory, in which the cell resistance can be switched by polarization reversal of the ferroelectric  $HfO_2$ , as depicted in Fig. 1(a). We demonstrated unique advantages of the HfO2-based FTJ [1], such as self-compliance, self-rectification, high non-linearity in I-V, high-speed switching, forming-free operation, excellent device-to-device uniformity and so on. These are attractive features for high-density cross-point array operation, and are hard to be realized in the conventional filament-type Ox-RAM or CBRAM. Fig. 1(b) shows several requirements for cross-point array applications. In addition to low power operation and non-volatility, immunity to voltage disturbance is required for reliable operation.

In this work, we demonstrate that the HfO<sub>2</sub>-FTJ with the template-induced crystallization technique improves these characteristics simultaneously and we show that the FTJ is quite suitable for cross-point high-density applications.



**Fig. 1** Schematic images of (a) resistive switching mechanism of ferroelectric tunnel junction and (b) large-scale cross-point array in write or read operation.

#### 2. Concepts of template-induced crystallization in FTJ

Fig. 2(a) summarizes the impacts of the template-induced crystallization (TIC). The template layer (TL) decreases crystallization temperature of the ferroelectric HfO<sub>2</sub> (FE-HfO<sub>2</sub>), which at the same time reduces the interlayer (IL) thickness. The depolarization field ( $E_{dep}$ ) is sensitive to the IL thickness [1, 2] and it is decreased with reducing the IL thickness as shown in Fig. 2(b). The decreased  $E_{dep}$  improves data retention. Meanwhile, the crystal orientation of the FE-HfO<sub>2</sub> is also modulated by the TIC. We expect that the effective remnant polarization (Pr), namely the vertical component of the Pr, is improved. We experimentally demonstrate that these effects are responsible for simultaneous improvement of various memory characteristics such as data retention, operation voltage, disturb immunity, on-current and ON/OFF ratio.



**Fig. 2(a)** Schematic image of our FTJ cell and concepts of template induced crystallization. (b) Calculated depolarization field as a function of IL thickness. Calculation was performed refferring to Ref. [3].

## 3. Physical analysis of TIC FE-HfO<sub>2</sub>

First, we investigate impacts of the TIC on crystallization temperature and crystal orientation of the FE-HfO<sub>2</sub>. The HfO<sub>2</sub> film was deposited by an ALD process, and it was amorphous state before crystallization annealing. Fig. 3 shows in-plane XRD spectra of the FE-HfO<sub>2</sub> w/ and w/o the TIC, before and after post metallization annealing (PMA) at various temperatures. Crystallization temperature of the FE-HfO<sub>2</sub> is clearly decreased with crystallized TL. In addition, (020) peak is found in the HfO<sub>2</sub> w/ TIC sample while not in that w/o TIC, meaning that b-axis of the FE-HfO<sub>2</sub>, which is perpendicular to polarization direction [4], is at the vertical direction in the w/o TIC sample. On the other hand, polarization direction is randomly oriented in the w/ TIC





sample. Therefore, the effective Pr is improved by the orientation control with TIC.

The reduction in crystallization temperature decreases the thickness of interlayer (IL) between FE-HfO<sub>2</sub> and bottom-electrode (BE), as confirmed by HAXPES (Fig. 4(a)). Here, oxide peak intensity is calibrated and proportional to the IL thickness. When crystallization of the HfO<sub>2</sub> is carried out at high temperature, the IL significantly grows compared to as-deposited specimen. The IL growth is suppressed by lowering the crystallization temperature. This behavior was confirmed by TEM as well (Fig. 4(b)-(d)). Meanwhile, it is observed in the TEM images that the lattice stripe in the FE-HfO<sub>2</sub> partly matches that in the TL. This also indicates that FE-HfO<sub>2</sub> crystallization is induced by the TL.



**Fig. 4 (a)** HAXPES spectra of FTJ stacks w/o PMA, w/ low-temperature (LT-) PMA and w/ high-temperature (HT-) HT-PMA. Signal intensity was calibrated by metallic peak of BE element. (**b-d**) TEM images of FTJ stacks (b) w/o crystallization annealing, (c) w/ LT-PMA and (d) w/ HT-PMA.

## 4. Electrical measurement of TIC-FTJ

DC I-V characteristics of the FTJ w/ and w/o the TIC are shown in Fig. 5(a). The ON/OFF ratio was clearly improved by the TIC, which can be attributed to the increase in the effective Pr of the FE-HfO<sub>2</sub>. Operation voltage and on-current were also improved by lowering the crystallization temperature thanks to the reduction in the IL thickness. Here, area-switching in the TIC-FTJ was confirmed by current density-voltage characteristics (Fig. 5(b)).



Fig. 5 (a) Current-voltage characteristics of  $HfO_2$ -based FTJ w/o and w/ TIC. (b) current density- voltage characteristics of  $HfO_2$ -FTJs with TIC, which have different device areas.

Pulse measurement was also performed, to check the memory properties in practical condition (Fig. 6(a)). Analog switching is realized even in short pulse measurement without current compliance. The operation voltage is slightly increased with shortening the pulse width. The slope of V-T characteristics is quite steep compared to that for the device w/o the TIC (Fig. 6(b)). This means that the operation voltage is kept relatively low even in short pulse condition, and TIC FTJ is robust for read- and set-disturb. These characteristics are mandatory for reliable cross-point array operation.

Improvement of disturb immunity is likely to come from the reduction in  $E_{dep}$ .



Fig. 6 (a) Write properties of TIC  $HfO_2$ -FTJ by pulse measurements with various pulse widths. (b) Required set time of  $HfO_2$ -FTJ w/ and w/o TIC, as a function of set voltage.

Retention properties of the TIC FTJ are shown in Fig. 7. While read current of the LRS rapidly decreases for the device w/o the TIC [1], the decay speed is much improved by introducing TIC. The memory window is estimated to be kept wide enough even after 10-years.



Fig. 7 Data retention properties of low and high resistance state, in our previous FTJ and TIC HfO<sub>2</sub>-FTJ with LT-PMA.

Furthermore, impacts of the TIC were confirmed regardless of the deposition condition of the  $HfO_2$  (data not shown). Further improvement is expected by adopting the TIC to optimized FE-HfO<sub>2</sub> ultrathin film which has large Pr like those reported in ref. [5].

## 4. Conclusions

Impacts of template layer insertion on memory properties of  $HfO_2$ -based ferroelectric tunnel junction memory were investigated experimentally. One is decreasing the crystallization temperature, which enables us to reduce IL thickness. The other is controlling crystal orientation of the FE-HfO<sub>2</sub> film. These effects lead to the simultaneous improvement of the operation voltage, data retention, disturb immunity, on-current and ON/OFF ratio, demonstrating the excellent suitability to cross-point array operation. Further improvement is expected by adopting template layer to optimized FE-HfO<sub>2</sub> thin film.

#### References

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