

Investigation of Ferroelectric Switching Mechanism and Improvement of Switching Speed in Si doped HfO₂ for FeRAM Application

Hyang Keun Yoo¹, Joong Sik Kim¹, Zhongwei Zhu², Matthew R. MacDonald³, Xinjian Lei³, Tae Yoon Lee⁴, Donghoon Lee^{5,6}, Seho Lee¹, Alex Yoon², Seung Chul Chae⁴, Jungwon Park^{5,6}, David Hemker², John G. Langan³, Yoshio Nishi⁷ and Jin Kook Kim¹

¹ SK Hynix Inc., Icheon-si, Gyeonggi-do, Korea

Phone: +82-31-639-7432 E-mail: hyangkeun.yoo@sk.com

² Lam Research Corp., Fremont, CA, USA

³ Versum Materials Inc., Carlsbad, CA, USA

⁴ Dep. of Phys. Edu., Seoul Nat. Univ., Seoul, S. Korea

⁵ Dep. of Chem. & Bio. Eng., Seoul Nat. Univ., Seoul, S. Korea

⁶ CNR, Institute for Basic Science (IBS), Seoul, S. Korea

⁷ Dep. of Elec. Eng., Stanford Univ., Stanford, CA, USA

Abstract

Ferroelectric (FE) HfO₂ has recently drawn much interest due to new device applications such as FeRAM, FeNAND, and neuromorphic memory. FE HfO₂ has a relatively high coercive field (E_c). This makes a stable and robust FE state, good for storage devices. However, it also induces a slow polarization switching speed. This is critical for fast speed FeRAM memory device. Thus, it is one of the most crucial issues to improve the FE switching speed. Here, we investigated the ferroelectric switching dynamics of Si doped HfO₂. The polarization switching behavior can be explained by the domain-wall growth mechanism, which implies that the large E_c limits the switching speed. By manipulating the grain size, we found that the Si:HfO₂ consisting of nano-grains have a FE property with $E_c \sim 0.44$ MV/cm, half of the normal Si:HfO₂, and the domain switching speed becomes ~ 3 times faster.

1. Introduction

HfO₂ based materials have been widely used in the CMOS technology. The recently discovered ferroelectric (FE) property has extended the applications of HfO₂ material for new types of non-volatile memory applications including 1T-FeRAM [1], 3D FeNAND [2] and advanced neuromorphic computing device [3]. For memory application, the FE domain switching speed should be compatible with the conventional DRAM or at least other non-volatile new memories. Emerging FE property is considered as the phonon mediated collective phenomena, so the switching speed becomes as fast as the speed of sound [4]. However, it is usually much slower in real materials due to extrinsic disorders [5]. In HfO₂ based FE materials, the understanding of the switching mechanism and its properties should be conducted before device applications. Nevertheless, there are almost no systematic investigations of its switching dynamics. Here, we investigated the switching speed of FE Si:HfO₂ as well as the governing switching mechanism.

2. Results and discussion

A Si 4.2 mol% doped HfO₂ (4.2Si:HfO₂) film is an almost

optimally doped sample [6]. Figure 1(a) reveals P - E curves of 4.2Si:HfO₂ in as-grown and after-cycling of 10^4 . We used a dynamic hysteresis measurement function in TF 3000 with 1kHz, 3V amplitude. After cycling, the remnant polarization, P_r , and coercive field, E_c , values become slightly larger in P - E curve, often called the “wake-up” effect [7].

The switching dynamics of the FE 4.2Si:HfO₂ is understood by the domain-wall growth mechanism. We measured the frequency dependence of P - E hysteresis loop in the woken-up condition. When the frequency increases from 10Hz to 50kHz, both the positive and negative E_c increase. The $E_{c\pm}$ versus frequency curves are depicted in fig. 1(b). We found that the E_c is proportional to $\log f$ with two-scaling

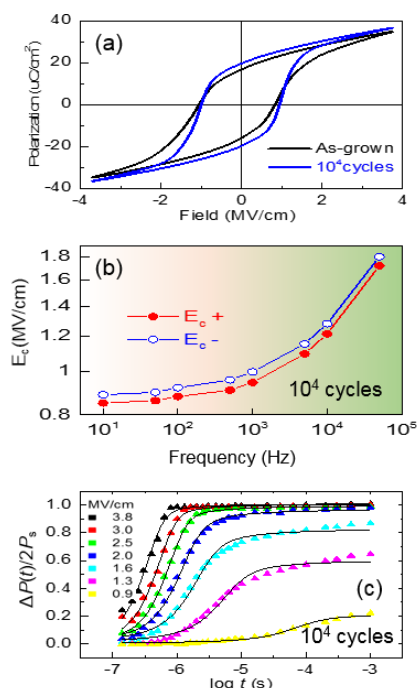


Fig. 1 [6] (a) P - E hysteresis loop for 4.2 mol% Si:HfO₂ in as-grown and after- 10^4 cycles. (b) E_{c+} and E_{c-} versus frequency curves after 10^4 -cycle. (c) Time-dependent change of polarization, $\Delta P(t)$, by varying the amplitudes in 10^4 cycled condition.

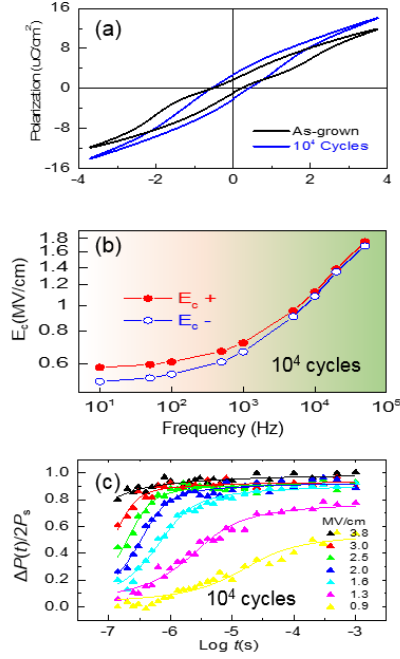


Fig. 2 [6] (a) P - E hysteresis loop for nano-grain Si:HfO₂ in as-grown and after-10⁴ cycles. (b) E_{c+} and E_{c-} versus frequency curves after 10⁴-cycle. (c) Time-dependent change of polarization, $\Delta P(t)$, by varying the amplitudes in 10⁴ cycled condition.

regions corresponding to creep and flow motions in domain-wall growth model [8]. Additionally, we measured the time (t)-dependent change in polarization $\Delta P(t)$ by varying the amplitudes of the applied external fields (E_{app}) from 0.9 to 3.8 MV/cm. The $\Delta P(t)$ normalized by 2 times of saturated polarization P_s versus $\log t$ are shown in fig. 1(c). The polarization switching behaviors can be analyzed in the classical Kolmogorov, Avrami and Ishibashi (KAI) model although the defects are slightly accompanied [9]. These results indicate that the FE domain dynamics in the 4.2Si:HfO₂ are mainly determined by domain-wall growth motion, and are not driven by extrinsic defects [5].

The switching speed of FE domain in 4.2Si:HfO₂ is limited by a large E_c . As shown in fig. 1(c), when the E_{app} increases, the $\Delta P(t)$ becomes increased in the same t . However, we found that the switched $\Delta P(100\text{ns})$ is less than 30%, even with very high E_{app} , such as 3.8 MV/cm. This indicates that the FE HfO₂ is not applicable for high-speed devices operated within 100ns. In the FE domain-wall growth model, switching speed is strongly affected by E_c [9]. The E_c of the 4.2Si:HfO₂ is ~1 MV/cm which is 10~20 times larger than that of perovskite-based FE materials [10]. This might be the origin of the slow switching speed, so the controllability of E_c is one of the prerequisites of the development of high-speed FE memory devices.

The strategy to achieve the fast switching property in the FE Si:HfO₂ is lowering the E_c without severe defect generation. Not only is the E_c generally considered as an intrinsic property of the material, but also the conventional methods of changing the E_c , such as doping control, always induce the

unavoidable disorder effects. It is well-known that FE property becomes unstable with reduction of E_c when the crystal size reaches to the critical size for FE [6]. This suggests that the reduction of HfO₂ grain size can be a new avenue to lower the E_c without significant defect generation.

The crystallization condition of Si:HfO₂ is changed by the Si-content and post-annealing temperature. This indicates that, if we make the film in the condition near the phase boundary between crystalline and amorphous, we can get the Si:HfO₂ film consisting of nano-grains [6]. Figure 2(a) reveals P - E curves of nano-grain HfO₂. The as-grown sample shows a little anti-FE like P - E curve. However, after 10⁴ cycles, the P - E curve becomes symmetric with $E_c \sim 0.44$ MV/cm.

The switching dynamics of the nano-grain HfO₂ can also be explained by the domain-wall growth mechanism. We found that, in fig. 2(b), the E_c is also proportional to $\log f$ with two-scaling regions consistent with the results of FE 4.2Si:HfO₂. This result supports that making the nano-grain sample of FE Si:HfO₂ is not accompanied with significant defect generations. Additionally, we also measured the $\Delta P(t)$ versus $\log t$ in fig. 2(c). We found that the amount of the switched $\Delta P(t)$ is enlarged by reduction of E_c . With 0.44 MV/cm of E_c , the switched $\Delta P(100\text{ns})$ becomes ~80% with 3.8 MV/cm of E_{app} .

3. Conclusions

We investigated the polarization switching dynamics in FE Si:HfO₂. In the optimal doping, Si:HfO₂ shows domain growth driven switching dynamics. The switching speed of Si:HfO₂ becomes limited by large E_c . We found that the E_c can be lowered by making a Si:HfO₂ films consisting of nano-grains. We experimentally proved that the FE switching speed can be enhanced by grain size control without critical disorder effects. Our results provide a new way to realize the high speed 1T-FERAM device.

Acknowledgements

The authors gratefully acknowledge the contributions of all of joint development (JD) members in Lam Research, Versum Materials and SK Hynix. J. P. and D. L. acknowledges support by IBS-R001-D1.

References

- [1] J. Muller *et al.*, IEDM (2013) 10.8.1.
- [2] K. Florent *et al.*, VLSI (2017) T12-4.
- [3] H. Mulaosmanovic *et al.*, VLSI (2017) T13-3.
- [4] D. S. Rana *et al.*, Adv. Mater. **21** (2009) 2881.
- [5] D. J. Jung *et al.*, Integr. Ferroelectr. **48** (2002) 59.
- [6] H. K. Yoo *et al.*, IEDM (2017) 19.6.1.
- [7] M. Pešić *et al.*, Adv. Func. Mater. **26** (2016) 4601.
- [8] S. M. Yang *et al.*, Phys. Rev. B **82** (2010) 174125.
- [9] J. Y. Jo *et al.*, Phys. Rev. Lett. **102** (2009) 045701.
- [10] J. Miller *et al.*, ECS J. Solid State Sci. Tech. **4** (2015) N30.