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Electric Field Cycling-mediated
Variations in Defect Distributions
Associated with Split-up
Behavior of a Ferroelectric Si-
doped HfO_2 Thin Film

전기장에 의한 결함분포 변화와 관련된 실리콘이
도핑 된 산화물 하프늄 박막의 Split-up 거동

2020년 8월

서울대학교 대학원

과학교육과 물리전공

송 명 섭

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지도 교수 채 승 철

이 논문을 교육학석사 학위논문으로 제출함
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서울대학교 대학원
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Abstract

We report variations in the characteristic ferroelectric switching time and interface free carrier concentration associated with the wake-up and split-up behaviors of a 4.2 mol % Si-doped HfO_2 thin film. Prior to the development of split-up behavior, the thin films exhibited the wake-up behavior; the remnant polarization increased on repeated external bias cycling. After the wake-up behavior, Si-doped HfO_2 films exhibited degradation of the remnant polarization values and splitting of the ferroelectric switching current peaks when the external bias was swept; this is referred to as split-up behavior. An investigation of the ferroelectric switching dynamics revealed retardation, followed by recovery, of the characteristic switching time for ferroelectric nucleation, coincident with the wake-up and split-up behaviors, respectively. We analyzed the interface free carrier concentration by deriving capacitance-voltage characteristics and revealed that split-up behavior was associated with increased numbers of defects as electric field cycling continued.

Keyword : : Ferroelectricity, HfO_2 , Split-up, thin film, defect

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I. Introduction

The ferroelectricity of ultra-thin HfO_2 films has attracted increasing attention given that the films are highly compatible with current complementary metal oxide semiconductor (CMOS) processes [1-4]. The increasing demand for non-volatile universal memory has driven a great deal of research on the maintenance of the ferroelectricity of such films, which have many potential commercial applications. When various dopants were included, HfO_2 films fabricated using the sub-10 nm CMOS process exhibited good ferroelectricity; the remnant polarization P_r was of the order $10 \mu\text{C}/\text{cm}^2$ [5, 6]. Such ferroelectric properties of HfO_2 may mitigate the blockage of ferroelectric memory caused by the perovskites [7, 8]. Thus, HfO_2 films exhibiting ferroelectricity may find immediate applications not only in the context of random access memory, but also in the construction of tunneling junctions [9, 10]. However, although non-volatile memory applications using HfO_2 film are maturing, the utility of such ferroelectricity remains debatable; it is unclear whether it is thermally stable and reliable during switching [11, 12].

A detailed understanding of ferroelectricity and the properties thereof in HfO_2 thin films is essential for practical applications. Stabilization of the non-centrosymmetric orthorhombic $\text{Pca}2_1$ phase has often been suggested to give rise to ferroelectricity [5, 13, 14]. Prior to commercialization, variations in ferroelectric switching mechanisms, and the characteristics thereof under external bias cycling, must be thoroughly understood. For example, the wake-up [15, 16], fatigue [11, 17-19], and split-up [20] behaviors may be of major concern in terms of the stability of a ferroelectric memory device. Regarding the wake-up behavior, enhancement of spontaneous polarization has been suggested to reflect structural

variations among ferroelectric orthorhombic and non-ferroelectric structural phases (i.e., monoclinic and tetragonal phases). In terms of fatigue behavior, various experimental methods, including first-order reversible curve (FORC) [21] construction, first-principle analysis [22], impedance spectroscopy [19], scanning tunneling electron microscopy [19], and harmonic analysis [17], have attributed the origin of fatigue to variations in bulk defect concentrations [19], modification of interface junction properties [18], or structural decomposition mediated by charge carrier injection [23, 24]. However, the split-up behavior, the splitting of the ferroelectric switching current peaks, has not previously been investigated rigorously, compared with the emergence and enhancement of ferroelectricity in HfO_2 via external field cycling. Thus, a deeper understanding of ferroelectric characteristics in terms of split-up behavior is essential from a practical viewpoint.

A comprehensive study on ferroelectric switching dynamics would clarify the factors governing split-up behavior associated with oxygen vacancies and other defects. Studies on polarization switching over time have aimed to define the types of defects in ferroelectric materials. For example, ferroelectric switching dynamics revealed the presence of dipole defects in polycrystalline $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (poly-PZT) films [25], and the development of distinct interfacial defects when the switching time distributions were asymmetric [26]. Although the split-up behavior of the HfO_2 thin-film ferroelectric switching current has been studied using conventional polarization hysteresis [17] and FORC construction [20], the underlying mechanism requires more attention; a thorough understanding of ferroelectric switching dynamics would reveal the mechanism underlying split-up behavior.

In this Letter, we report the ferroelectric switching dynamics of the split-up behavior of Si-doped HfO_2 thin films. We studied the evolution of ferroelectric properties using the standard polarization-voltage (P - V) hysteresis approach, and current-voltage (I - V) measurement, during electric field cycling; 8 nm-thick Si- HfO_2 thin films were fabricated via atomic layer deposition (ALD) and capacitance-voltage (C - V) data were obtained to explore the dielectric properties of, and defect distributions in, the interface. The C - V data revealed variations in dielectric properties and the effective interface carrier densities. The split-up behavior when the effective charge density was varied near interfaces was attributable to increased numbers of oxygen vacancies; these controlled the recovery of the characteristic switching time as the ferroelectric switching dynamics evolved.

II. Material and methods

Ferroelectric TiN/Si:HfO₂/TiN capacitors were fabricated on a Si (001) substrate. Prior to fabrication of Si-doped HfO₂ films, the bottom TiN layer was deposited via chemical vapor deposition. The Si-doped HfO₂ film was deposited onto the TiN/Si substrate via ALD. Tetrakis(dimethylamido) hafnium (TDMAH), tetrakis(dimethylamino) silane (4DMAS), and ozone were used for ALD. The top TiN electrode was then deposited via chemical vapor deposition. All specimens were post-annealed at 600°C for 20 s in N₂ to crystallize the Si:HfO₂ films. In terms of electrical and ferroelectric characterization, *P-V* hysteresis and time-dependent dynamic polarization switching [$\Delta P(t)$] were explored using a conventional semiconductor parameter analyzer (model 4200; Keithley) and ferroelectric tester (TF Analyzer 3000; aixACCT Systems).

III. Results and discussion

3.1. Electric field cycling behavior of ferroelectric HfO₂ thin film

Si-doped HfO₂ thin films exhibited initial wake-up and subsequent split-up behaviors. Samples were subjected to 1×10^4 external voltage cycles (3 V at 10 kHz) for wake-up. Next, samples were subjected to 1×10^{10} external voltage cycles (1.6 V at 100 kHz) to allow split-up to be studied [20]. The P - V hysteresis measurements were conducted after each wake-up/split-up test. Fig. 1a shows the P - V hysteresis loops of Si-doped HfO₂ films (pristine, preset, woken-up, and split-up). The as-grown specimens ("pristine") exhibited pinched hysteresis attributable to the interface pinning effect [17]. As external bias cycling commenced, the remnant polarization increased to 15 $\mu\text{C}/\text{cm}^2$ and the woken-up specimens developed clear rectangular hysteresis. Such enhancement of the remnant polarization was observed in HfO₂ films containing various dopants; structural phase transitions during cycling enhanced the remnant polarization [15, 19]. As shown in Fig. 1b, after wake-up, ongoing cycling of the external bias induced peak splits in the ferroelectric switching current (split-up behavior). In addition, the remnant polarization of split-up specimens fell as electric field cycling continued (Fig. 1e).

To further investigate in variations of switching current, we conducted first order reversal curves (FORCs) measurement. FORCs measurement shows switching current distribution based on the Preisach model. The bias voltage indicates the asymmetry of preferred direction of polarization by asymmetry of top and bottom

interfacial factors, defect distribution or material difference, etc. Figs. 1c-d show FORCs of ferroelectric Si doped HfO_2 thin film in woken-up and split-up specimen, respectively. The bias voltage merged to near zero voltage that indicates the distribution of switching current homogeneous and the switching peak enhanced in woken-up specimen. After split-up process, two peaks of switching current emerged near at -0.8 and 0.4V that could be contributed to disruption of ferroelectric polarization switching by defect distribution.

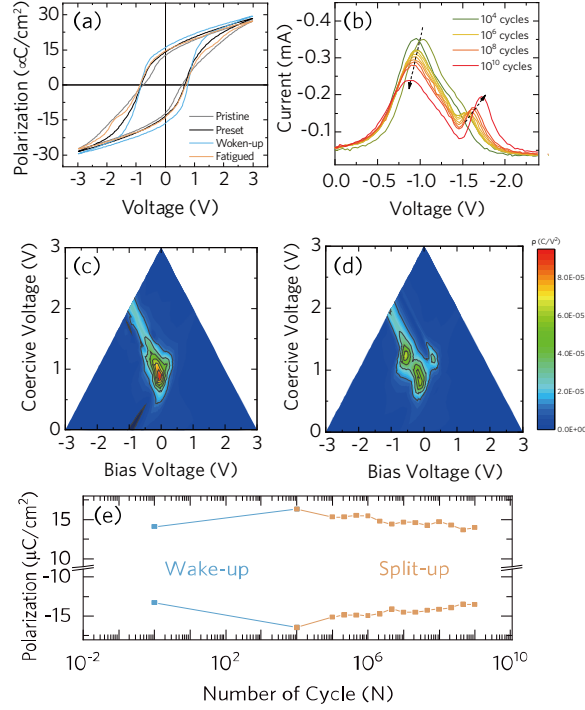


Fig. 1. (a) Polarization-voltage hysteresis of the pristine, preset, woken-up, and split-up cells (Pt/TiN/Si:HfO₂/TiN capacitors). (b) Current-voltage measurements of preset, woken-up, and split-up cells. Experimental switching density acquired from the (c) woken-up and (d) split-up cells. (e) The external field cycle number-dependence of the remnant polarization, which increased and decreased after wake-up and split-up, respectively.

3.2. Investigation of ferroelectric switching dynamics

To characterize the ferroelectricity of pristine, woken-up, and split-up specimens, we studied the ferroelectric switching dynamics under various external voltages. The dynamics of preset, woken-up, and split-up cells were compared in terms of time-dependent changes in the polarization switching $[\Delta P(t)]$ values. We investigated switching dynamics after 20 cycles of external bias were applied to pristine specimens; this was associated with a rapid wake-up effect and we refer to these specimens as "preset". We used a bundle of 3-V trapezoidal pulses with a rise time duration of 125 μ s; $\Delta P(t)$ values were derived by subtracting the non-ferroelectric from the ferroelectric switching currents (Fig. 2a). A rectangular pulse applied from 140 ns to 1 ms (i.e., a "write" pulse) was used to explore partial switching of ferroelectric polarization (Figs. 2b–d). The switched polarization value ($2P_s$) was measured using a read pulse with a bias opposite to that of the write pulse. After recording $2P_s$, a poling pulse was applied to completely switch the polarization again. To avoid incomplete polarization saturation, the $\Delta P(t)/2P_s$ curves were obtained using a voltage higher than that of the coercive field. The detailed measurement method has been described earlier [16]. Preset, woken-up, and split-up specimens exhibited typical polarization switching behaviors; polarization increased as the amplitude and application time of the external voltage increased (Figs. 2b–d). All specimens exhibited increased $\Delta P(t)/2P_s$ ratios as the external bias increased from 2.0 to 3.0 V.

Experimentally, ferroelectric switching is characterized by an inhomogeneous distribution of local nucleation times, followed by polarization reversal mediated

by limited nucleation switching [16, 27-29]. Ferroelectric switching is governed by both nucleation and subsequent domain growth [30]. Polarization switching kinetics have been modeled using (1) the Kolmogorov-Avrami-Ishibashi (KAI) [31, 32] method and (2) a nucleation-limited-switching (NLS) approach [33]. The KAI model assumes that both homogeneous nucleation and unrestricted domain growth are statistical in nature [31, 32]. The NLS model assumes that polarization switching is limited by domain nucleation, and considers that a polycrystalline film is an ensemble of microscopic regions that are independent in terms of their switching kinetics [33]. For polycrystalline ferroelectric HfO_2 thin films, experiments have suggested that polarization switching is best modeled using the NLS approach [8, 29, 34]. Therefore, we fitted the $\Delta P(t)/2P_s$ values using an NLS model featuring a Lorentzian distribution of the characteristic switching time, as shown in Equations (1) and (2) [25]:

$$\frac{\Delta P(t)}{2P_s} = \int_{-\infty}^{\infty} \left[1 - \exp\left\{-\left(\frac{t}{t_0}\right)^2\right\}\right] F(\log t_0) d(\log t_0) \quad (1)$$

with:

$$F(\log t_0) = \frac{A}{\pi} \left[\frac{w}{(\log t_0 - \log t_1)^2 + w^2} \right] \quad (2)$$

where A , w , and $\log t_1$ are a normalization constant, the half-width at half-maximum of the distribution, and the mean distribution value, respectively. The solid lines in Figs. 2b–d indicate that polarization switching was well-described by the NLS model when there was a Lorentzian distribution.

The switching time of ferroelectric nucleation was retarded, but then recovered, in woken-up and split-up specimens, respectively. Fig. 3a shows the deconvoluted Lorentzian distribution used when constructing the fit lines of the solid ferroelectric switching dynamics curves of Figs. 2b–d. The peak positions in Fig. 3a are the characteristic switching times ($\log t_0$ values) of the preset, woken-up, and split-up specimens. For woken-up specimens, the characteristic switching times of the peak positions were retarded, and the peaks were sharpened (blue dashed-dotted lines in Fig. 3a). For split-up specimens, the retarded times recovered and the distributions became broader again (yellow dashed lines in Fig. 3a). Fig. 3b shows the characteristic ferroelectric switching times extracted from Fig. 3a. Distribution sharpening and broadening, as well as switching time retardation and recovery, can be explained by the changes in defect concentrations associated with ferroelectric polarization switching.

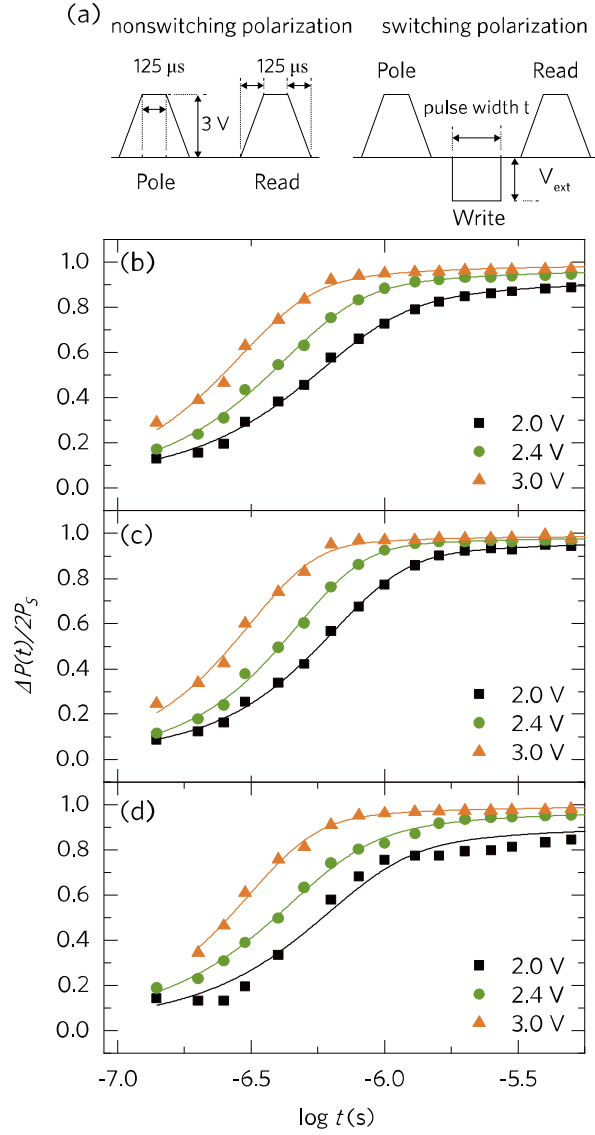


Fig. 2. (a) Schematic of the pulse trains used to measure non-switching and switching polarization. Time-dependent switched polarization $[\Delta P(t)]$ as a function of the external voltage (V_{ext}) from 2.0 to 3.0 V at room temperature for (b) preset, (c) woken-up, and (d) split-up cells. The solid lines correspond to the fitting results obtained using the nucleation-limited switching (NLS) model with a Lorentzian distribution of the characteristic switching time.

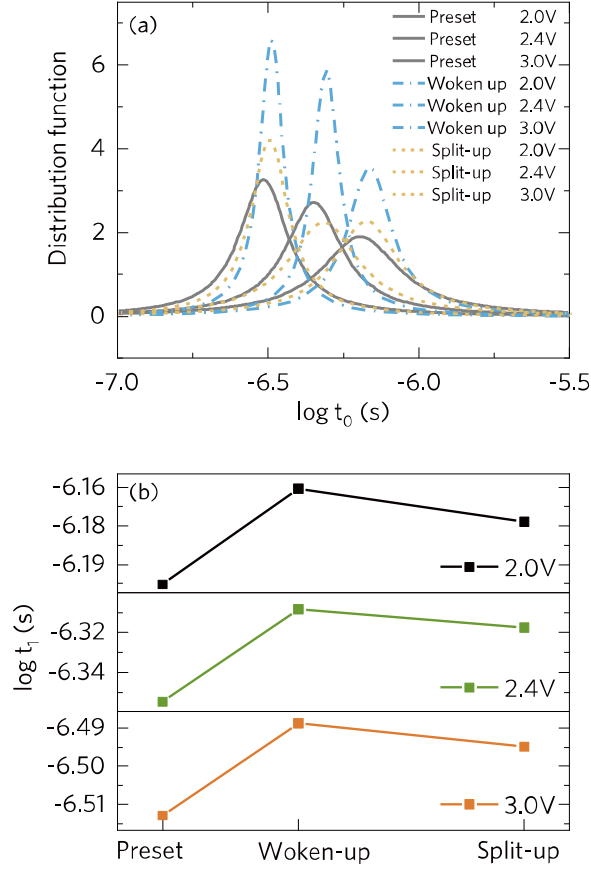


Fig. 3. (a) Lorentzian distributions of the fitting functions for the time-dependent switched polarization shown in Figs. 2b–d of the preset, woken-up, and split-up cells. (b) The extracted characteristic switching time values of the preset, woken-up, and split-up cells when the external voltage varied.

Inhomogeneous switching and changes in polarization switching time are attributable to changes in defect density; this affects local field distribution, where defects serve as pinning sites for domain wall motion inside films [18, 23-25]. For example, a dipole defect aligned antiparallel to the local lattice polarization impedes both nucleation and domain growth. However, a parallel defect dipole enhances both nucleation and domain growth [35]. Also, charge defects, oxygen vacancies, depletion layers, non-ferroelectric structural phases, and pinned domain orientations all trigger inhomogeneous switching behavior [15, 18, 36]. Such defects, and the associated charge-trapping, influence the field distribution over all film layers [37]. The ferroelectric layer encounters a reduced electric field, which either impedes domain propagation [38] or reduces the number of switching domains if the electric field is less than the coercive field [18]. Local defect-related variations in field distribution reduce the nucleation time and average domain size [39, 40]. Film disorders caused by defects serve as nucleation seeds and the amount of activation energy required for domain nucleation decreases [40]. For woken-up specimens, reduced defect density decreases disorder, in turn triggering a more homogeneous switching distribution, followed by retardation of the polarization switching time and marked redistribution of the characteristic ferroelectric switching time. These effects combine to render the inhomogeneity of the switching distribution, and to retard (and then allow recovery of) the characteristic switching time during wake-up and split-up, respectively.

3.3. Changes of interfacial property

During electric field cycling, defect distributions change especially at film interfaces. A defect-rich interfacial layer is formed during deposition of the HfO₂ onto bottom and top TiN electrodes [41, 42]. Repeat electric field cycling induces redistribution of initial oxygen vacancies [43]. It is well-known that repeat cycling generates defects, including oxygen vacancies [44]. It has been suggested that oxygen vacancies are created in, and then migrate from, film interfaces during electric field cycling [45, 46].

To explore variations in interface structure, we performed C-V measurements on preset, woken-up, and spilt-up specimens of Si-HfO₂ films. The free carrier concentration derived from the 1/C²-V plots has been investigated at the interface of the metal-semiconductor junctions. The metal-ferroelectric-metal junction has been considered as two back-to-back metal-semiconductor junctions [47, 48] where 1/C²-V plots also revealed the many metal-ferroelectric junction properties similar to the metal-semiconductor junction [49-54]. Accordingly, the split-up behavior with respect to the junction properties can be understood by utilizing the capacitance measurement.

Variations in free carrier concentration levels may be calculated via analysis of the interface depletion capacitance [47, 55, 56]. The depletion capacitance (C_d) attributable to a small AC signal is defined as [57]:

$$C_d = \frac{\epsilon_r \epsilon_0 d}{W} = \frac{\epsilon_r \epsilon_0 d}{\sqrt{\frac{2\epsilon_r \epsilon_0}{q N_a} (V_{bi} - V)}} \quad (3)$$

where ϵ_r , ϵ_0 , A , N_a , V_{bi} , V , and q are the dielectric constant of the film, permittivity of free space, electrode area, free carrier concentration, built-in potential, applied bias, and charge, respectively. When Equation (3) is used to analyze the $1/C^2$ - V curves, the free carrier concentration (N_a values) can be determined.

Fig. 4a shows the C-V hysteresis curves for preset, woken-up, and split-up specimens, respectively. The clear hysteresis evident in Fig. 4a reflects the ferroelectricity of the preset, woken-up, and split-up specimens. The peaks in C-V curves are associated with polarization reversal and indicate the transition of the depletion capacitance from the bottom to top and top to bottom Schottky contacts. The peaks are in fact same with the coercive voltage of polarization switching which is represented in Fig. 1. But the coercive voltage in C-V measurement is smaller than coercive voltage in P-V or I-V hysteresis because the C-V curve is a quasi-static measurement. Fig. 4b shows the free carrier concentration-voltage behaviors calculated from Equation (3) when sweep down from 2 V to -2 V for preset, woken-up, and split-up specimens, respectively. In Fig. 4b, the sign of calculated N_a values changes abruptly near 0.5 V which correspond with the coercive voltage in C-V measurement. This is not due to the change in the carrier type but the aforementioned transition of the depletion capacitance which result in sign change in the $d(1/C^2)/dV$ derivation. Except the region where the polarization switching occurs, the free carrier concentration was well estimated without the influence of the polarization reversal on the measured capacitance. Note that the linearity of $1/C^2$ - V plots and the constant N_a values indicate that the effect of ferroelectric polarization switching is negligible (inset in Fig. 4b). In the saturated region, the N_a value of woken up specimen is $2.79 \times 10^{18} \text{ cm}^{-3}$ which is lower than

the preset specimen $3.29 \times 10^{18} \text{ cm}^{-3}$. After the split-up process, the N_a value rebounded $2.92 \times 10^{18} \text{ cm}^{-3}$ (Table 1). Although the N_a values differed between sweep-up and sweep-down because of interface asymmetry, the trends associated with continued electric field cycling were identical.

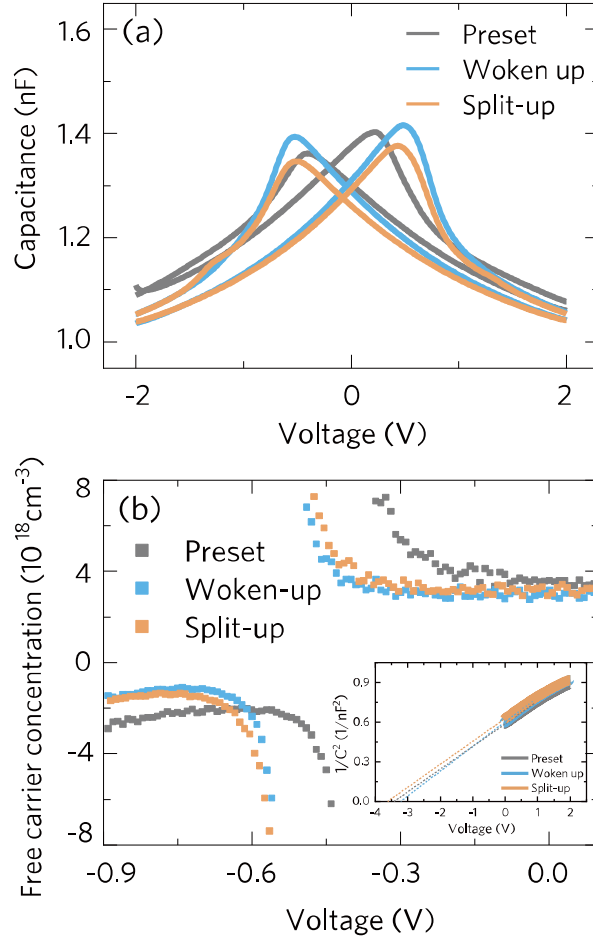


Fig. 4. (a) Capacitance-voltage characteristics and (b) free carrier concentration-voltage behavior of Si:HfO₂ thin film for the preset, woken-up, and split-up cells. Inset shows linear fitting of $1/C^2$ -V plots for preset, woken-up, and split-up cells in region where ferroelectric switching is negligible.

Table 1. Free carrier concentrations of Si:HfO₂ thin film capacitors according to the extent of electric field cycling

	Free carrier concentration (cm ⁻³)	
	Sweep up	Sweep down
Preset	3.29E+18	3.34E+18
Woken-up	2.79E+18	3.06E+18
Split-up	2.92E+18	3.29E+18

Variations in local electric fields that interfere with ferroelectric polarization switching are attributable to changes in the density of oxygen vacancies. Such vacancies near interfaces trap charges that interfere with free polarization switching [58]. A change in the number of oxygen vacancies can be estimated by measuring the free carrier concentration. We used Equation (3) to derive changes in the free carrier concentration of woken-up and split-up specimens, which indicate changes in the number of oxygen vacancies at the film interface [47]. Redistribution of such vacancies decreased free carrier concentration from 3.29×10^{18} to $2.79 \times 10^{18} \text{ cm}^{-3}$ during wake-up. However, after split-up process, the concentration increased from 2.79×10^{18} to $2.92 \times 10^{18} \text{ cm}^{-3}$. Thus, further electric field cycling generated oxygen vacancies near the interface, accompanied by split-up behavior. Such changes in the number of oxygen vacancies with continued field cycling causes both the initial retardation, and subsequent recovery, of the characteristic switching time.

IV. Conclusions

In summary, we investigated the polarization switching dynamics of, and obtained C-V data on, woken-up and split-up Si-doped HfO_2 thin films. The peak splits of ferroelectric switching currents were evident in the split-up specimens; the remnant polarization decreased on repeat electric field cycling. The polarization switching of polycrystalline ferroelectric Si-doped HfO_2 was well-described by the NLS model with a Lorentzian distribution of the characteristic switching time for nucleation. In split-up specimens, the lag in the characteristic switching time (attributable to defect redistribution during wake-up) was gradually eliminated, and the shape of the distribution broadened. We used C-V data to estimate defect densities near the interface. Given the observed ferroelectric switching dynamics and derived C-V measurements, we found that split-up behavior was attributable to changes in oxygen vacancy density; such changes induced local fields that interfered with ferroelectric polarization switching.

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Appendix

<A.1> Experimental method for first order reversal curves (FORCs) measurement

A.1.1. Derivation of FORC measurement

The first order reversal curves (FORCs) measurement shows the switching density (Preisach density) on the switching field distribution.

The simplest way to calculate this switching density is as follows: To derive the switching density, the polarization switching current have to be obtained by field sweeps starting at saturated field increasing the amplitude of reversal field as shown in Fig A1. The switching density is defined as the mixed second derivative of the polarization with respect to the reversal field E_r and actual field E , i.e., field region from the reversal field to saturated field:

$$\rho(E_r, E) = \frac{1}{2} \cdot \frac{\partial^2 P_{FORC}(E_r, E)}{\partial E_r \partial E} = \frac{1}{2 \cdot E} \frac{\partial j_{FORC}(E_r, E)}{\partial E_r} \approx \frac{1}{2E} \frac{j_{FORC}(E_{r,i}, E) - j_{FORC}(E_{r,i-1}, E)}{E_{r,i} - E_{r,i-1}} \quad (A1)$$

For each i -th reversal field $E_{r,i}$ plotted as the y -axis, the difference in the measured switching current response to the current for the previous reversal field is plotted versus the actual field E . It is recommended to adjust the measuring sample points as amplitude of reversal field in instrument system or fitting the switching current to aligning actual field E_i . In this paper we represented the FORC distribution versus the coordinates of coercive voltage V_c and the bias voltage V_{bias} representing respectively coercive field E_c and the bias field E_{bias} by using coordinate transformation:

$$V_c = \frac{V - V_T}{2}, V_{bias} = \frac{V + V_T}{2} \quad (A2)$$

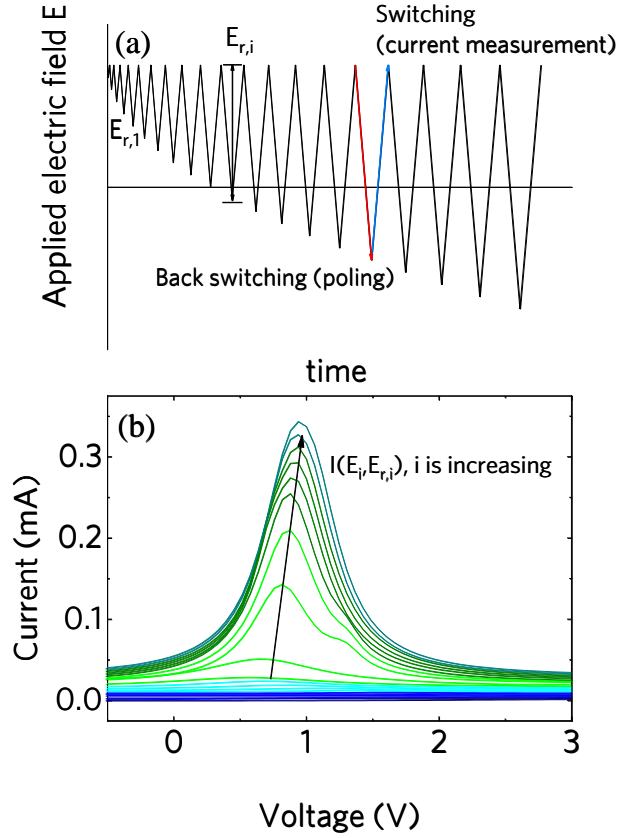


Fig. A1. (a) Schematic of electric field evolution along time for the first order reversal curve (FORC) measurement. Red arrow indicates the back switching field to pole the ferroelectric domain and blue arrow indicates the switching field for calculating switching density. (b) Corresponding transient current-voltage curves.

A.1.2. Detailed manual using keithley 4200 instrument

1. Make a new user test module to measure a capacitor structure using PMU modules and build the pulse train for back switching and switching by editing segment arb configuration (Fig. A2). It is recommended to make a same number of test module as i which represent sequence number of reversal field.

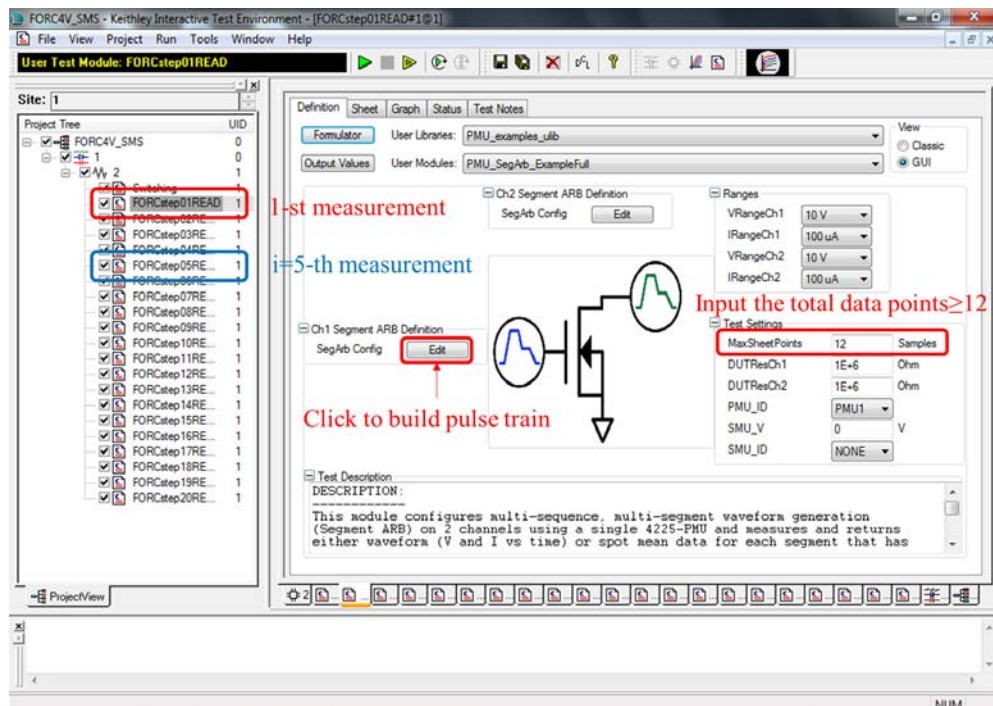


Fig. A2. User interface of keithley 4200. List of sequence of user test module for FORC measurement(Left) and parameter controller of 1-st measurement sequence.

2. The pulse train of i -th test module is made of a trapezoid pulse which starts at the saturated field and decreases to i -th reversal field and end at the saturated field again (Fig A3). The amplitude of reversal field should increase as arithmetic progression as increasing i until the reversal field equal to the opposite saturated field. The current data to calculate the switching density could be obtained by measuring the current during the switching process. The current data during the poling process is not necessary.

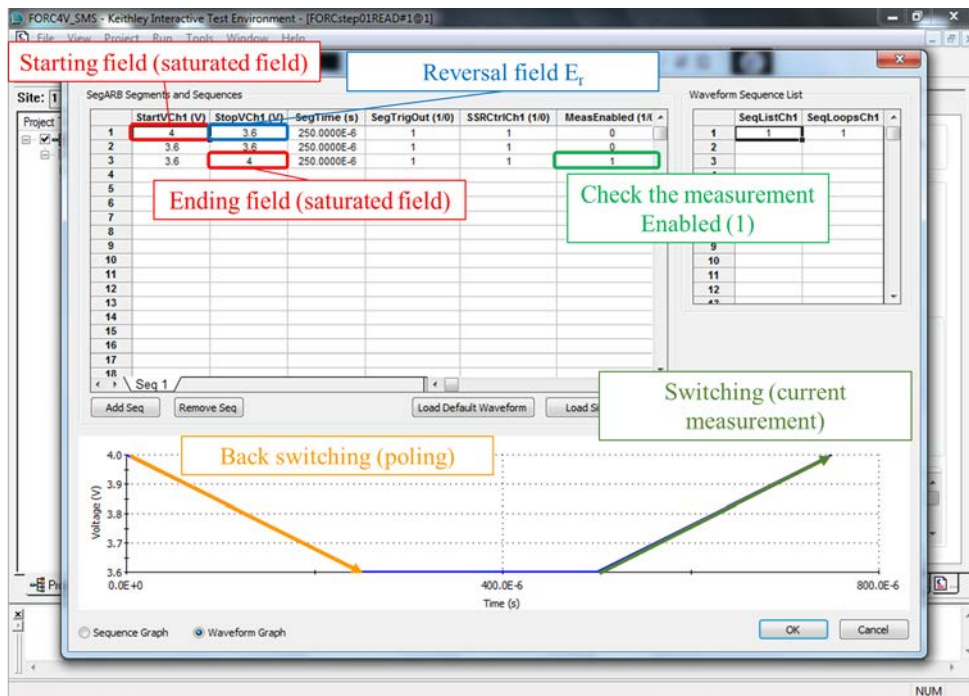


Fig. A3. Segment arb configuration of 1-st measurement sequence. The pulse train measurement can be manipulated by inputting amplitude, segment time, measurement enabled and etc.

3. Before applying pulse train, you have to adjust MaxSheetPoints in test setting. (Fig. A2.) Maxsheetpoints represent total points of data during the measurement and its minimum is 12. The current data should be obtained from each of the same actual field. Therefore, maxsheetpoints can be defined by multiplying of amplitude of reversal field by the maxsheetpoint of 1-st measurement, which represent how the FORC diagram shows distribution specifically. If you set the maxsheetpoints to $n \geq 12$ in 1-st measurement, the maxsheetpoints of i-th measurement should be $n \times i$.

4. Obtained current data should be aligned by actual field. It is recommended to calculate the FORC measurement with fitted data to minimize misaligning of the current data. The difference of the measured switching current response to the current for the previous reversal field can be calculated by subtracting the i -th data from $(i-1)$ -th data (Fig. A4).

VMeasCh1	IMeasCh1	VMeasCh1	IMeasCh1			
3.5934E+0	53.6267E-9	3.1935E+0	58.1807E-9			
3.6202E+0	47.7162E-9	3.2138E+0	57.3250E-9			
3.6517E+0	51.7703E-9	3.2412E+0	58.6888E-9			
3.6852E+0	52.4898E-9	3.2711E+0	60.8909E-9			
3.7184E+0	48.2888E-9	3.3027E+0	58.9035E-9			
3.7510E+0	51.5450E-9	3.3362E+0	62.7501E-9			
3.7841E+0	51.7819E-9	3.3689E+0	61.8363E-9			
3.8177E+0	46.8608E-9	3.4023E+0	58.4923E-9			
3.8506E+0	49.5830E-9	3.4354E+0	59.1301E-9			
3.8844E+0	51.4536E-9	3.4680E+0	58.1354E-9			
3.9174E+0	50.2197E-9	3.5018E+0	58.0813E-9			
	$I(E_i, E_{r,i})$		$I(E_i, E_{r,i})$	Actual Field E	Reversal Field E_r	$\Delta I(E_i, E_{r,i})$
3.5934E+0	53.6267E-9	3.5684E+0	60.8016E-9	3.5934E+0	3.5934E+0	7.1749E-9
3.6202E+0	47.7162E-9	3.6018E+0	62.1890E-9	3.6202E+0	3.5934E+0	14.4728E-9
3.6517E+0	51.7703E-9	3.6348E+0	60.6882E-9	3.6517E+0	3.5934E+0	8.9178E-9
3.6852E+0	52.4898E-9	3.6678E+0	58.6525E-9	3.6852E+0	3.5934E+0	6.1627E-9
3.7184E+0	48.2888E-9	3.7009E+0	62.9789E-9	3.7184E+0	3.5934E+0	14.6901E-9
3.7510E+0	51.5450E-9	3.7339E+0	62.0126E-9	3.7510E+0	3.5934E+0	10.4676E-9
3.7841E+0	51.7819E-9	3.7675E+0	60.7012E-9	3.7841E+0	3.5934E+0	8.9192E-9
3.8177E+0	46.8608E-9	3.8009E+0	56.9832E-9	3.8177E+0	3.5934E+0	10.1224E-9
3.8506E+0	49.5830E-9	3.8341E+0	59.7863E-9	3.8506E+0	3.5934E+0	10.2033E-9
3.8844E+0	51.4536E-9	3.8675E+0	61.4676E-9	3.8844E+0	3.5934E+0	10.0140E-9
3.9174E+0	50.2197E-9	3.9012E+0	63.4444E-9	3.9174E+0	3.5934E+0	13.2247E-9

Fig. A4. Current-Voltage measurement data obtained by 1-st and 2-nd sequence of FORC measurement.

5. For each i -th reversal field $E_{r,i}$ plotted as the y-axis, plot the calculated switching density as the z-axis versus the actual field E as the x-axis.

6. You can represent the FORC distribution versus the coordinates of coercive voltage/field and the bias voltage/field by using coordinate transformation in eq. A2.

국문 초록

이 논문에서는 강유전성 스위칭 시간과 계면의 자유 전하 밀도와 관련하여 wake-up 현상과 split-up 현상에 대해 4.2% mol 실리콘 도핑 된 산화물 하프늄을 이용하여 연구하였다. Split-up에 앞서, 박막은 전기장 전계에 따라 잔류 분극이 증가하는 wake-up 현상을 보였다. Wake-up 현상 이후, 외부 전기장에 의해 잔류 분극 값이 감소하였고 강유전성 스위칭에 의한 잔류 분포가 나누어지는 현상, 즉 split-up 현상을 보였다. Nucleation limited switching 모델을 기반으로 한 강유전성 스위칭 동역학을 통해 분석하였을때, 강유전성 핵생성을 위한 스위칭 시간은 wake-up과 split-up현상에 따라 각각 느려지고, 빨라졌다. 이와 함께 전압-전기용량 특성의 결과를 분석하였을 때, 박막내 결함의 증가가 split-up 현상에 영향을 주는 것으로 생각된다.

주요어 : 강유전체, 산화물 하프늄, Split-up, 박막, 결함

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