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Polarization switching in Hf$_{0.5}$Zr$_{0.5}$O$_2$-dielectric stack: The role of dielectric layer thickness

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Abstract

Understanding the role of the dielectric (DE) layer in ferroelectric (FE) Hf$_{0.5}$Zr$_{0.5}$O$_2$ (HZO) based devices (e.g., ferroelectric-field-effect-transistors, FE-FETs) is important to enable their application-driven optimizations. To that end, in this work, we systematically investigate the polarization switching mechanisms in FE–DE stacks and analyze their dependence on the dielectric layer thickness ($T_{DE}$). First, we fabricate a HZO–Al$_2$O$_3$ (FE–DE) stack and experimentally demonstrate a decrease in remanent polarization and an increase in coercive voltage with an increase in $T_{DE}$. As such dependencies are out of the scope of commonly used single domain polarization switching models, therefore, we argue that the consideration of the multi-domain model is essential for analyzing the polarization switching in HZO. Then, using phase-field simulations of the FE–DE stack, we show that an increase in $T_{DE}$ results in a larger number of reverse domains in the FE layer to suppress the depolarization field, which leads to a decrease in the remanent polarization and an increase in the coercive voltage. Furthermore, our analysis signifies that the polarization switching mechanism in HZO can be modulated from domain-nucleation based to domain-wall motion based by increasing the $T_{DE}$. Therefore, it is important to bridge the gap between the theoretical understanding and experimental observations regarding the role of the DE layer thickness in the P-switching characteristics of FE HZO. To that end, in this Letter, we experimentally and theoretically analyze the P-switching in the FE–DE stack with HZO as the FE and Al$_2$O$_3$/HfO$_2$ as the DE layer have been demonstrated to exhibit an increasing coercive voltage ($V_C$) with the increase in $T_{DE}$. Therefore, it is important to bridge the gap between the theoretical understanding and experimental observations regarding the role of the DE layer thickness in the P-switching characteristics of FE HZO. To that end, in this Letter, we experimentally and theoretically analyze the P-switching in the FE–DE stack with HZO as the FE and Al$_2$O$_3$/HfO$_2$ as the DE layer. Our results signify an increase in $V_C$, a decrease in remanent-P ($P_R$), and a decrease in the P-switching slope with the increase in $T_{DE}$. By employing phase-field simulations, we show that such dependencies can be attributed to the multi-domain (MD) phenomena in FE, which cannot be captured in the single-domain P-switching model. To further study the role of the DE layer, we analyze the dependence of $V_C$, $P_R$, and the switching slope of the FE–DE stack through phase-field simulations. It is worth noting that

Ferroelectric (FE) hafnium-zirconium-oxide (HZO), by virtue of its CMOS process compatibility$^{12}$ and rich domain dynamics,$^{13-15}$ has been identified as one of the most promising candidates for future electronic devices. By integrating HZO as the FE layer in the gate stack of a transistor (ferroelectric-field-effect-transistors, FE-FET), nonvolatile memory (NVM),$^{16}$ neurons,$^{17}$ and synaptic$^{18,19}$ functionalities have been demonstrated. Such diverse functionalities demand different characteristics of polarization ($P$) switching in the HZO layer. For example, an abrupt $P$-switching is beneficial for neurons and binary NVMs, while a gradual $P$-switching is favorable for multi-bit memories and synapses. Therefore, it becomes essential to appropriately design FE-FETs for application-specific device behavior, for which gate stack optimization plays a key role. In the FE-FET gate stack, a dielectric (DE) layer exists between the FE and the semiconductor channel,$^{20}$ which can significantly impact the FE-FET characteristics.$^{21}$ According to the single-domain (SD) Landau–Khalatnikov (LK) model of FE, an increase in the DE thickness ($T_{DE}$) should increase the depolarization field and reduce the coercive voltage ($V_C$) of the FE–DE stack$^{22}$ (see the supplementary material). However, the FE–DE stack with HZO as the FE and Al$_2$O$_3$/HfO$_2$ as the DE layer have been demonstrated to exhibit an increasing coercive voltage ($V_C$) with the increase in $T_{DE}$. Therefore, it is important to bridge the gap between the theoretical understanding and experimental observations regarding the role of the DE layer thickness in the P-switching characteristics of FE HZO. $^{23,24}$ To that end, in this Letter, we experimentally and theoretically analyze the P-switching in the FE–DE stack with HZO as the FE and Al$_2$O$_3$/HfO$_2$ as the DE layer. Our results signify an increase in $V_C$, a decrease in remanent-P ($P_R$), and a decrease in the P-switching slope with the increase in $T_{DE}$. By employing phase-field simulations, we show that such dependencies can be attributed to the multi-domain (MD) phenomena in FE, which cannot be captured in the single-domain P-switching model. $^{25}$ To further study the role of the DE layer, we analyze the dependence of $V_C$, $P_R$, and the switching slope of the FE–DE stack through phase-field simulations. It is worth noting that...
while the multi-domain formation in FE in the presence of a dead layer and in FE–DE stack is a well-explored topic in the context of perovskite-based FE, the considered HZO–Al2O3 system and the corresponding analysis in this paper are significantly different. For instance, the fundamental nature of P-switching dynamics in FE HZO (fluorite based) is expected to be significantly different from the perovskite-based FE materials [i.e., lead zirconate titanate (PZT), barium titanate (BTO), etc.] because of the substantially less elastic coupling in the fluorites compared to the perovskites. In addition, the previous works were mostly focused on the temperature-driven phase transition and the dependence of the domain configuration on stress and other factors. In contrast, in our work, we focus on the applied voltage-driven polarization switching and the influence of microscopic interactions (electrostatic and elastic) and how the DE thickness plays an important role to modulate the multi-domain configuration and, thus, the corresponding P-switching mechanism.

For fabrication of the FE–DE stacks, we start with standard solvent cleaning of heavily p-doped Si substrates. Then, a 30 nm TiN layer is deposited by atomic layer deposition (ALD) at 250 °C, using [(CH3)2N]4Ti and NH3 as the Ti and N precursors, respectively. After this, an HZO film is deposited by ALD at 200 °C, using [(CH3)2N]4Hf, [(CH3)2N]4Zr, and H2O as the Hf, Zr, and O precursors, respectively. A HfO2:ZrO2 cycle ratio of 1:1 is used to form the 10 nm Hf0.5Zr0.5O2 film. Similarly, on top of HZO, an Al2O3 layer is deposited followed by 30 nm TiN layer deposition. After that, the samples are annealed at 500 °C in the N2 environment for 1 minute by rapid thermal annealing. Then, Ti/Au top electrodes are fabricated using photolithography, e-beam evaporation, and lift-off process (area = 5024 μm²). The material level characterization of the HZO and HZO–Al2O3 stacks (i.e., XRD and SEM/TEM) can be found in Refs. 31 and 32. The average polarization \( P_{\text{avg}} \) vs applied voltage \( V_{\text{app}} \) measurement is carried out using a Radiant RT66C FE tester at room temperature at a very low frequency (50 Hz). Considering the polarization switching time in HZO (<1 μs), such low-frequency measurements can be considered as quasi-static. Figure 1(a) shows the \( P_{\text{avg}}-V_{\text{app}} \) characteristics of the FE–DE stack for 10 nm HZO and 1/3/5 nm Al2O3. Our results show a decrease in \( P_{\text{avg}} \) at \( V_{\text{app}} = 0 \) V, an increase in \( V_C \) (\( V_{\text{app}} \) at \( P_{\text{avg}} = 0 \)), and a decrease in the \( P \)-switching slope (\( dV_{\text{avg}}/dV_{\text{app}} \)) with the increase in \( T_{\text{DE}} \). To explain such dependencies, we now analyze the \( P \)-switching in the FE–DE stack based on multi-domain phase-field simulation.

In our 2D phase-field simulation, we self-consistently solve the time-dependent Ginzburg–Landau (TDGL) equation [Eq. (1)] and Poisson’s equation [Eq. (2)] in 2D space (xz plane) employing the finite difference method.

\[
-\frac{\partial P}{\partial t} = \alpha P + \beta P^3 + \gamma P^5 - \frac{\partial^2 P}{\partial x^2} - \frac{\partial^2 P}{\partial z^2} + \frac{\partial \phi}{\partial x} - \frac{\partial \phi}{\partial z} \tag{1}
\]

\[
-\varepsilon_0 \left( \frac{\partial}{\partial x} \left( \varepsilon_x \frac{\partial \phi}{\partial x} \right) + \frac{\partial}{\partial z} \left( \varepsilon_z \frac{\partial \phi}{\partial z} \right) \right) = -\frac{dP}{dz}. \tag{2}
\]

Here, \( \alpha, \beta, \) and \( \gamma \) are Landau coefficients; \( \varepsilon_{x(\varepsilon_z)} \) is the gradient coefficient; \( \varepsilon_{\text{rel}} \) is the relative background permittivity; \( \Gamma \) is the

![Image](https://example.com/figure1.png)

**FIG. 1.** (a) Measured and (b) simulated \( P_{\text{avg}}-V_{\text{app}} \) characteristics of FE–DE stacks for different \( T_{\text{DE}} \). (c) Average \( V_C \) (\( V_{\text{app}} \) at \( P_{\text{avg}} = 0 \)) and \( P_{\text{R}} \) (\( P_{\text{avg}} \) at \( V_{\text{app}} = 0 \)) for different \( T_{\text{DE}} \). (d) Table showing the simulation parameters.
that the rhombic HZO crystal) is parallel to the film thickness (z-axis).18–22

multi-domain (MD) state, the shown in Fig. 1(b), illustrating a good agreement with the experiments simulating multiple grains (discussed later).

near the domain-walls (DWs) and (ii) electrostatic energy $P_{\text{grad}}$. Note that the variation in coercive fields, a multi-grain simulation is needed, such as variation in coercive fields, a multi-grain simulation is needed, and hence, reduced $|P|$ with the increase in $T_{\text{DE}}$.

Now, let us discuss $V_{\text{app}}$-induced $P$-switching. In the MD scenario, the $P$-switching can take place locally if $f_{\text{grad}} + f_{\text{dep}} + f_{\text{ffree}} > \max(f_{\text{free}})$. Therefore, for this discussion of local $P$-switching, we will use the notion of local electric-fields and energy components. First, note that the local $E_{\text{DE}}$ is maximum away from DW near the FE–DE interface, which leads to maximum $f_{\text{dep}}$. In contrast, $f_{\text{grad}}$ is maximum near the DW due to the largest variation in $P$ at the DW. Now, with an increase in $V_{\text{app}} (>0 \text{ V})$, the local (depolarizing) $|E_{\text{DE}}|$ increases in $-P$ domains and decreases in $+P$ domains leading to a local change in the $P$ magnitude ($|P|$ decreases in $-P$ domains and increases in $+P$ domains). This $V_{\text{app}}$ induced local change in $E_{\text{DE}}$ and $P$ yields an increase in $f=f_{\text{grad}}+f_{\text{dep}}+f_{\text{ffree}}$ in the $-P$ domains.18 If the increase in $f$ is dominant near the DW, then $P$-switching occurs through DW motion. However, if the increase in $f$ is dominant away from the DW, then $P$-switching occurs through the nucleation of new domains. $P$ profiles at different $V_{\text{app}}$ are shown in Fig. 3(a-i) for $T_{\text{DE}} = 1 \text{ nm}$. With the increase in $V_{\text{app}}$ $P$-switching starts through DW motion (at $V_{\text{app}} = 1.5 \text{ V}$) and at $V_{\text{app}} > 3 \text{ V}$ several new domains nucleate causing a denser domain pattern. The transient nature of domain nucleation is shown in Fig. 3(a-ii), signifying their formation starting from the FE–DE interface and a little away from the existing DW. Once, the domain pattern becomes denser, a significant portion of $E_{\text{DE}}$ is suppressed by the stray fields, albeit at the expense of some increase in $f_{\text{grad}}$. In this case, the maximum of $f$ occur near the DW, and hence, with further increase in $V_{\text{app}}$ P-switching takes place through DW motion leading to complete switching ($-P$ to $+P$) of several reverse domains. Similarly, for $T_{\text{DE}} = 3 \text{ nm}$ [Figs. 3(b-i) and 3(b-ii)], P-switching initiates through DW motion (at $V_{\text{app}} = 1.83 \text{ V}$) followed by domain nucleation (at $V_{\text{app}} > 3.15 \text{ V}$) and then DW motion. However, for $T_{\text{DE}} = 5 \text{ nm}$ [Fig. 3(c)], the initial domain pattern is much denser, which suppresses $E_{\text{DE}}$ at the cost of an increased $f_{\text{grad}}$. Hence, nucleation of new domains is not observed, and P-switching takes place only through DW motion (at $V_{\text{app}} > 3.6 \text{ V}$). It is important to note that, for both the domain-nucleation and DW motion we discussed here, the increase in $f_{\text{grad}}$ (or an increase in depolarizing $E_{\text{DE}}$) is the dominant component for satisfying the $P$-switching condition, $f > \max(f_{\text{free}})$. This is because the $f_{\text{grad}}$ component is significantly less in HZO (due to a lower gradient energy coefficient compared to the conventional perovskite-based FE materials). As $f_{\text{grad}}$ is the dominant energy component for the P-switching in HZO, hence, there is a correlation between the domain nucleation and DW-motion. For instance, the DW-motion in HZO can be thought of as the nucleation of a region just in the vicinity of the DW.
Now, let us analyze the effective polarization–voltage characteristics of the FE layer in the FE–DE stack. To obtain the average voltage drop across the FE layer ($V_{FE}$) for a particular $P_{avg}$, we compute the average voltage drop across the DE layer ($V_{DE}$) from its capacitance ($C_{DE} = \varepsilon_0 \varepsilon_r / T_{DE}$) and displacement continuity condition ($V_{DE} = P_{avg} / C_{DE}$). Then, we compute the $V_{FE} (= V_{app} - V_{DE})$ and plot the $P_{avg}$-$V_{FE}$ curves from both experimental and simulated characteristics as shown in Figs. 4(a) and 4(b). It should be noted that these $P_{avg}$-$V_{FE}$ characteristics are not an explicit or intrinsic representation of the $P_{avg}$-$V_{FE}$ characteristics of a standalone FE layer. Rather, this is an effective and/or apparent representation of the $P_{avg}$-$V_{FE}$ characteristics of the FE layer that appears in an FE–DE stack. It is interesting to note that these extracted $P_{avg}$-$V_{FE}$ characteristics signify a negative $dP_{avg}/dV_{FE}$ region in the multi-domain $P$-switching in the FE layer, which is similar to the earlier works on perovskite-based FE.\(^{35-37}\)

While such effects have been interpreted in some earlier works as either the transient effects\(^{13}\) or the stabilized single-domain negative capacitance effects,\(^{14}\) here our simulations suggest that the appearance of the negative slope in the hysteretic $P_{avg}$-$V_{FE}$ characteristics are quasi-static in nature originating from the multi-domain $P$-switching in the FE layer. To understand the physical origin of this negative $P_{avg}$-$V_{FE}$ slope, first, recall that the local (and average) $E_{z,FE}$ in the FE layer is depolarizing, i.e., opposite to the direction of local and average $P$. Now, let us consider the FE–DE stack is in the $P_R < 0$ states (average $E_{z,FE} > 0$). When $V_{app}$ is increased (>0 V) and leads to MD $P$-switching, the $P_{avg}$ increases either through the formation of new +P domains (nucleation) or through the size increase in +P domains (DW displacement). Both of these phenomena lead to a large decrease in local $E_{z,FE}$ (from positive to negative) in the newly switched +P region leading to a decrease in average $E_{z,FE}$ (i.e., the average $E_{z,FE}$ becomes less positive). As the increase in $P_{avg}$ accompanies the decrease in average $V_{FE} (= T_{DE} E_{z,FE})$, thus the $dP_{avg}/dV_{FE}$ becomes negative. A similar negative $dP_{avg}/dV_{FE}$ region can be obtained for $P_R > 0$ and $V_{app} < 0$. It is important to note that the appearance of negative $dP_{avg}/dV_{FE}$ is an electrostatic effect as each of the points in the $P_{avg}$-$V_{FE}$ curve is electrostatically stable. However, the actual slope of $dP_{avg}/dV_{FE}$ can certainly be impacted by the frequency of the applied $V_{app}$ due to the time-dependency of domain-nucleation and DW-motion. Moreover, the $P_{avg}$-$V_{FE}$ characteristics are not independent of $T_{DE}$ [Fig. 4(a)], and hence, the possibilities for single domain $P$-switching to be the source of negative $dP_{avg}/dV_{FE}$ are unlikely in this considered scenario.

Further, the DW motion occurs via lattice-by-lattice propagation yielding a gradual increase in $P_{avg}$. In contrast, nucleation of a
new domain involves simultaneous $P$-switching in several lattices leading to a sharper change in $P_{\text{avg}}$. Since, with an increase in $T_{\text{DE}}$, the dominant $P$-switching mechanism changes from nucleation to DW-motion-based, $P$-switching becomes more gradual—Figs. 1(a) and 1(b). Furthermore, our simulations show a stepwise $P$-switching behavior for $T_{\text{DE}} = 5$ nm [Fig. 1(b)], where each step jump signifies the DW displacement, and the flatter region corresponds to no DW displacement. The non-zero slope of the flatter region is due to the background permittivity and the change in $P$ magnitude. In this flat region, with an increase in $V_{\text{app}}$, $E_{Z,\text{FE}}$ first increases. If the increase in $E_{Z,\text{FE}}$ is beyond a critical value so that $\alpha > \max(\alpha_{\text{free}})$, then the $P$-switching takes place via DW displacement. Recall that the $P$-switching leads to an increase in $P_{\text{avg}}$ and a simultaneous reduction in $E_{Z,\text{FE}}$. This yields a negative slope in the $P_{\text{avg}}$-$V_{\text{FE}}$ characteristics ($dP_{\text{avg}}/dV_{\text{FE}} < 0$) and a step jump in the $P_{\text{avg}}$-$V_{\text{app}}$ characteristics. Now, after each $P$-switching step, to induce further DW motion, $V_{\text{app}}$ needs to be increased to increase $E_{Z,\text{FE}}$ beyond a (new) critical value. Consequently, we observe a stepwise $P$-switching behavior in Figs. 1(b) and 4(b). However, such step-jumps are absent in the measured characteristics because of the larger area (lots of grains) of the fabricated sample compared to our simulation (~one grain). Thus, even though the DW motion may be absent in some of the grains of the experimental sample, it may be present in other grains (due to the variation in grain size and/or crystallographic angle) leading to a continuous increase in $P_{\text{avg}}$ and decrease in $E_{Z,\text{FE}}$. Hence, we expect that simulation of a larger system considering multiple grains may reduce this mismatch between the simulation and experimental results.

Let us now explain the effect of $T_{\text{DE}}$ on the average coercive voltage, $V_C$ (defined as the $V_{\text{app}}$, where $P_{\text{avg}} = 0$). For that, we use the notion of the average voltage drop across the FE layer ($V_{\text{FE}}$). As an increase in $T_{\text{DE}}$ and corresponding formation of the denser domain pattern in the FE layer suppress the local and average $E_{Z,\text{FE}}$ (discussed before), it leads to a decrease in $V_{\text{FE}}$ at $V_{\text{app}} = 0$ V. With the decrease
in initial $V_{FE}$ a higher $V_{app}$ is required to achieve a critical $V_{FE}$ (as well as local $E_{DE}$) to trigger $P$-switching. Therefore, the DW motion initiates at $V_{app} = 1.5$ V for $T_{DE} = 1$ nm and at 1.85 V for $T_{DE} = 3$ nm. Similarly, the domain nucleation takes place at $V_{app} > 3$ V for $T_{DE} = 1$ nm and $V_{app} > 3.15$ V for $T_{DE} = 3$ nm. Furthermore, $dP_{avg}/dV_{app}$ decreases with the decrease in $T_{DE}$ (discussed before) leading to an increase in required $V_{app}$ to achieve $P_{avg} = 0$. Due to the decrease in initial $V_{FE}$ (at $V_{app} = 0$ V) and lower $dP_{avg}/dV_{app}$, $V_C$ of the FE–DE stack increases with an increase in $T_{DE}$. Note that the increase in $V_C$ for larger $T_{DE}$ cannot be captured by the SD model \cite{12} but can be described well considering the MD effects (as explained above).

So far, we have discussed different attributes of $P_{avg}$-$V_{app}$ characteristics of FE–DE stacks with respect to different $T_{DE}$. This can also be regarded as the influence of different DE capacitances ($C_{DE} = \epsilon_{DE}/T_{DE}$) in the FE-DE stack. Now, one might argue that the $P_{avg}$-$V_{app}$ characteristics can be tuned similarly by using a different DE material ($\epsilon_{DE}$). While this is indeed possible (and can be an important design knob), the effect of $\epsilon_{DE}$ is not just changing $C_{DE}$ but involves some more physical processes that mandate further analysis. To decouple the effect of $\epsilon_{DE}$ on $C_{DE}$, we theoretically analyze the FE–DE characteristics in the supplementary material by simultaneously and proportionally changing $\epsilon_{DE}$ and $T_{DE}$ to keep the same $C_{DE}$. We show that the $P_{avg}$-$V_{app}$ characteristics are not unique to $C_{DE}$ rather they depend on the choice of $\epsilon_{DE}$. Such dependency originates due to the electrostatic boundary condition of the in-plane electric field at the FE–DE interface. Considering different $\epsilon_{DE}$ (but the same $C_{DE}$), our simulation results suggest that the $V_C$ decreases and $P_g$ increases with the decrease in $\epsilon_{DE}$. We discuss such an $\epsilon_{DE}$ dependency on the $P$-switching in the FE–DE stack in the supplementary material.

In summary, we show that the FE layer forms a denser domain pattern with increasing $T_{DE}$ by suppressing the depolarization field and leading to a larger hysteresis in the FE–DE stack. Simultaneously, the mechanism of $P$-switching can be modulated from nucleation to DW-motion dominant by increasing $T_{DE}$. In addition, we show that the coercive voltage and remanent polarization can further be modulated by $\epsilon_{DE}$ while keeping the same $C_{DE}$. Such $T_{DE}$ and $\epsilon_{DE}$ dependency can serve as the potential knobs to deploy the application-driven optimization of the FE-FET gate stack. For instance, FE-FETs with low $T_{DE}$ (high switching slope) can be used for the design of binary NVMs and neurons, while high $T_{DE}$ can be utilized for multi-bit memories and synapse designs.

See the supplementary material for the $P$-switching characteristics in the FE–DE stack (i) with the single-domain approximation and (ii) the impact of dielectric permittivity on multi-domain $P$-switching.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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