Quantitative Characterization of Interface Traps in Ferroelectric/Dielectric Stack Using Conductance Method

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Abstract—In this work, the conductance method with an optimized circuit model is established to investigate the trapped charges at the ferroelectric/dielectric (FE/DE) interface. The density of interface states is quantitatively characterized to be $\sim 4 \times 10^{12}$ to $10^{13}$ cm$^{-2}$ · eV$^{-1}$. And the injection and accumulation of these enormous interfacial charges play a key role in the operation of the FE/DE stack. The proposed measurement technique provides a comprehensive understanding of the FE/DE stack as well as some new insights of negative capacitance effect and ferroelectric field-effect transistor device operations.

Index Terms—Conductance method, ferroelectric/dielectric (FE/DE), hafnium zirconium oxide, interface traps, leakage-assist-switching mechanism.

I. INTRODUCTION

FERROELECTRIC-GATED field-effect transistor (FET) has been extensively studied for several years for its application on nonvolatile memory as ferroelectric FET (Fe-FET) [1]–[5]. The proposal of negative capacitance (NC) effect in ferroelectric devices exploits the development of NC-FET for low-power CMOS logic due to the steep sub-threshold swing below 60 mV/decade [6]–[10]. No matter in Fe-FETs or NC-FETs, the ferroelectric/dielectric (FE/DE) stack is commonly applied, where the DE layer is inevitable in the form of native oxide or passivation layer for a substrate. Therefore, the study of the FE/DE stack is of great importance. It is well-known that the FE/DE stack is fundamentally different from an FE capacitor and a DE capacitor in series [12],[13]. It becomes complicated due to the FE/DE interfacial coupling effect and charge accumulation at the FE/DE interface [14]–[22]. It is a fact that there is a gap of charge mismatch between remnant polarization in FE layers and maximum tolerable charge density in DE layers [18]. The remnant polarization $P_r$ of typical ferroelectric films is at the order of 10 μC/cm$^2$, whereas conventional dielectric insulators cannot support such a large charge density unless the existence of large leakage current and eventually the breakdown of dielectric. Thus, the leakage current and interfacial charges at the FE/DE interface are the only possible mechanism to make the polarization switching happen possible to satisfy the charge balance requirement [18]. In addition, the degradation of FeFETs has also been reported to be dominantly impacted by the interface properties, which is indicative of the significant FE/DE interfacial behavior during the operation of FE/DE stack [22]–[24]. However, the FE/DE interface trap properties have not been intensively and quantitatively investigated thoroughly.

Recently, a simultaneous $I_d-V_g$ and polarization-voltage ($P-V$) measurements by the positive up negative down (PUND) pulse was applied to Fe-FETs to extract the defect density of interface [23]. By calculating the undesired non-ferroelectric hysteresis loop of $I_d-V_g$ curves, a charge density of $\sim 10^{13}$ cm$^{-2}$ at the FE/DE interface was obtained, which accounts for the stress-induced imprint voltage shift in the $P-V$ loops. The charge density from this method is independent of polarization switching. Another novel split $C-V$ technique combined with Hall measurement has been introduced to separate interface traps from the ferroelectric polarization charges in Fe-FETs, shown a surprisingly large trapped charge of $\sim 10^{14}$ cm$^{-2}$ as the key role in the performance-boosting of transistors [20]. Nonetheless, the semiconductor/DE interface is also involved in the Fe-FETs, and the charge trapping at the semiconductor/DE interface might be confused with that at the FE/DE interface. To make the study of the FE/DE interface more distinct and straightforward, an FE/DE capacitor is more preferable.

In this work, the charge behaviors and interface trap properties at the FE/DE interface are quantitatively characterized in the structure of metal/FE/DE/metal capacitor. Hafnium...
zirconium oxide (Hf0.5Zr0.5O2, HZO) is chosen as the FE layer and high-k insulator Al2O3 is chosen as the DE layer. P–V and C–V properties at different temperatures and frequencies are first measured to examine the existence of interface traps. Then, the conductance method, which is widely used for the study of the metal-oxide-semiconductor (MOS) interface [25], is formulated to measure the trap density at the FE/DE interface (Dit). Our previous work [21] reported the conductance peak phenomenon in FE/DE stack in C–V measurement. In this work, taking the consideration of leakage current through FE/DE stack and band bending fluctuation effect into the circuit and mathematical model for an accurate characterization of FE/DE interface trap behaviors are developed based on the main results of [21]. Using this optimized model, a distribution of Dit and the corresponding time constant (τ) at different polarization states are finally extracted at different temperatures. It is found that the charge trapping at the FE/DE interface, which is supplied from the leakage currents through the ultrathin DE layer, dominates the polarization switching of FE/DE stack.

II. EXPERIMENTS

A. Capacitor Fabrication

Metal/FE/DE/metal capacitors were fabricated for the investigation of the FE/DE interface, as the cross-sectional schematic shown in Fig. 1(a). Heavily p-doped Si wafers with the resistivity less than 0.005 Ω·cm were adopted as the substrate after standard solvent cleaning. As the metal contact, TiN was deposited by atomic layer deposition (ALD) at 250 °C, using the [(CH3)2]4Ti and NH3 as the Ti and N precursors, respectively. Alternating monolayers of HfO2 and ZrO2 were deposited by ALD at 200 °C to achieve an overall film composition of Hf0.5Zr0.5O2 and a thickness of 10 nm, using [(CH3)2]4Hf, [(CH3)2]4Zr and H2O as the precursors of Hf, Zr, and O, respectively. To avoid cross-contamination, the TiN and HZO films were deposited in two separate chambers, which are connected externally by the Ar environment for 1 min by rapid thermal annealing to crystallize the HZO film. Then, Ti/Au top electrodes were fabricated by photolithography, e-beam evaporation, and a liftoff process. Dry etching was used to remove the top TiN layer by CF3/Ar.

B. Electrical Characterization

Electrical characterizations at room temperature (RT) were probed in a Cascade summit probe station and low temperature experiments were conducted in a Lakeshore cryogenic probe station. The capacitance–voltage (C–V) and conductance measurements were taken using an Agilent E4980A impedance analyzer and P–V measurements were carried out using a Radiant RT66 ferroelectric tester. The applied voltage ranges were maximized in P–V measurements before the leakage current had essential impacts. All devices have already been broken up before the P–V characterization.

III. RESULTS AND DISCUSSION

A. Temperature-Dependent P–V and C–V Properties

Fig. 1(b) shows the P–V curves of the HZO/Al2O3 capacitors with 10-nm HZO and different Al2O3 thicknesses from 1 to 5 nm. The ferroelectric hysteresis loops are observed which confirms the ferroelectricity of these HZO/Al2O3 capacitors. It is also found that the remnant polarization (Pr) decreases with the increasing Al2O3 thickness, which is due to the leakage-assist-switching mechanism [18]. The leakage currents through ultrathin Al2O3 layers with different thicknesses supply different quantities of mismatched charges for polarization switching, thus resulting in different Pr. These mismatched charges are essentially the trapped charges at the FE/DE interface.

To further demonstrate and investigate this charge trapping process, temperature-dependent P–V measurements were performed. Fig. 2(a) shows the P–V hysteresis loops for the 10-nm HZO/3-nm Al2O3 capacitor at low temperatures from 110 K to RT of 293 K. Temperature-dependent Pr is summarized in Fig. 2(b) for capacitors with different Al2O3 thicknesses. Pr decreases with the decreasing temperature, indicating the suppression of polarization switching. This phenomenon is the result of leakage current reduction through Al2O3 and the suppression of trap response at low temperatures, resulting in fewer trapped charges for polarization switching. It also indicates that the leakage-assist charge injection through the
FE/DE stack contains not only tunneling current but also thermionic emission process [26], [27]. Polarization switching at cryogenic temperatures suggests that both direct tunneling and trap-assisted tunneling are associated with the electron transport. Furthermore, \( P_r \) is found to decrease much more quickly in capacitors with thinner Al\(_2\)O\(_3\), suggesting that thermionic emission contributes sufficiently to the leakage current transport.

\( C-V \) characteristics were also measured at different frequencies and different temperatures. The frequency-dependent \( C-V \) properties, ranging from 1 kHz to 1 MHz at RT, are shown in Fig. 3(a) for 10-nm HZO/3-nm Al\(_2\)O\(_3\) capacitor. The capacitance decreases with the increasing frequency, as fewer traps respond to a higher frequency. The control sample, a capacitor with 20-nm Al\(_2\)O\(_3\) dielectric layer only was also measured and its capacitance value keeps almost constant at the whole frequency regime, as the red symbols and line shown in Fig. 3(b). Therefore, this frequency dispersion is not induced by the measurement setup or parasitic effect.

Fig. 3(b) shows the frequency-dependent capacitance value at low temperatures from RT down to 110 K. A smaller capacitance and larger frequency dispersion are observed at lower temperatures, which is attributed to the suppression of trap responses and charge injection. All these \( P-V \) and \( C-V \) measurement results under various temperatures and frequencies manifest the charge trapping at the FE/DE interface, supplied from leakage currents through the ultrathin DE layer, which is critical for the charge balance and polarization switching of FE HZO eventually.

### B. Principle of Conductance Method for FE/DE Stack

The conductance method, proposed by Nicollian and Goetzberger [25], is a sensitive technique to determine the trap density at the MOS interface. As a loss mechanism, interface trap capture and emission of carriers are represented by the conductance. This trap resulted loss process, as well as capacitances in the MOS structure, are lumped to the parallel of an equivalent conductance and an equivalent capacitor.

Applying a series of small-signal sine waves with different frequencies, this equivalent parallel conductance is measured, then the interface trap density and time constant can be obtained by fitting the measurement results to a theoretical circuit model. Comparing the possible equivalent circuit model of FE/DE stack with that MOS capacitor, the conductance method can also be applied to extract properties of the FE/DE interface traps.

Fig. 4(a) shows the proposed equivalent circuit of FE/DE stack, which is similar to that of MOS capacitors. The branch, including interface trap-related resistance \( R_t \) and capacitance \( C_{it} \), is parallely connected with the FE capacitance \( C_{FE} \) and then connected with the DE capacitance \( C_{DE} \) in series. Here, the interaction of interface traps with polarization charges by \( D_{it} \) (\( C_{it} = qD_{it} \)) is a lossy process, represented by \( R_t \). A conductance component \( G_L \) is introduced because of the leakage currents through the FE/DE stack. It will not affect the measurement result of interface traps, which will be explained later.

Because the impedance analyzers generally assume the device under test to consist of the parallel of measured conductance \( G_P \) and measured capacitance \( C_P \), one can replace the circuit of Fig. 4(a) by that in Fig. 4(b) for convenience, assuming negligible series resistance in the system. Thus, the total admittance \( Y_{it} \) of Fig. 4(a) can be normalized into the parallel connection of conductance \( G_P \) and capacitance \( C_P \) with

\[
Y_{it} = G_P + j\omega C_P
\]
and

\[
\frac{G_P}{\omega} = \frac{(\omega \tau_d)C_{DE}^2C_{it} + G_L}{(\omega \tau_d)^2(C_{FE} + C_{DE})^2 + C_A^2} + \frac{C_L}{\omega}
\]

\[
C_P = \frac{C_{DE}(C_{FE} + C_{it})}{C_{FE} + C_{DE} + C_{it}} (\omega \rightarrow 0)
\]

\[
C_P = \frac{C_{DE}C_{FE}}{C_{FE} + C_{DE}} (\omega \rightarrow \infty).
\]

where \(\omega\) is proportional to the measurement frequency \(f\), and \(\tau_d\) is equal to \(R_0C_{it}\). Here, for a clear description, \(C_A\) is the sum of all capacitances, defined as

\[
C_A = C_{FE} + C_{DE} + C_{it}.
\]

To demonstrate the validity of this circuit model, the component of \(C_P\) is analyzed first. It can be expressed as (5) when extending \(\omega\) to zero and as (6) when to infinity

\[
C_P = \frac{C_{DE}(C_{FE} + C_{it})}{C_{FE} + C_{DE} + C_{it}}.
\]

It is found that \(C_P\) is the serial connection of \(C_{DE}\) and \(C_{FE}\) at high frequency. However, at low frequency, \(C_P\) includes an extra branch of \(C_{it}\) in parallel with \(C_{FE}\). This changing tendency of \(C_P\) is consistent with the measurement result in Fig. 3(b), which can be theoretically explained by the frequency-related trap responses. The interface traps can respond to low-frequency signals but lag at high frequency. On the other hand, it is confirmed that for capacitors of 10-nm HZO/3-nm Al₂O₃, the numerical calculated \(C_P\) at RT, using (6) with the real \(C_{DE}\) (~1.9 \(\mu F/cm²\)) and \(C_{FE}\) (~2.3 \(\mu F/cm²\)), is approximate to the measured 0.95 \(\mu F/cm²\) as shown in Fig. 3(b). In addition, for all the measured devices with different Al₂O₃ thicknesses, the numerical calculated \(C_P\) using (6) with the real \(C_{DE}\) and \(C_{FE}\) have been confirmed to be approximate to the measured capacitance value at 1 MHz. Both phenomena indicate that the application of the conductance method with the optimized circuit model is valid to FE/DE capacitors.

As mentioned above, the relationship between \(G_P/\omega\) and \(f\) can be utilized to quantitatively extract the properties of interface traps. Note that \(G_P\) in (2) is composed of two terms. For the first one, it is always equal to zero when assuming \(\omega\) to be either zero or infinity, which means there must be a peak as the red dashed line shown in Fig. 4(c). As for the second term, \(G_{L}/\omega\) is inversely proportional to \(f\) as the blue dotted line shown in Fig. 4(c). Here, this \(G_{L}\) can be obtained by measuring the leakage current of FE/DE capacitors. Although this term is important in low-frequency regimes, it can be negligible in high-frequency regimes where the peak appears. Note, this conductance of the leakage current of \(G_{L}\) from top to the bottom electrode does not contribute to a conductance peak. Consequently, by adding these two together, the final curve of \(G_P/\omega\) versus \(f\) decreases first and then presents a peak with the generally increasing frequency, as the solid black line depicted in Fig. 4(c).

When carrying out the conductance measurement, a series of small ac signals with frequency changing over a wide range is applied to the FE/DE capacitors already biased at a constant voltage. Interfacial charges are resonances with the small ac signal at a certain frequency and then present a peak in the black solid line shown in Fig. 4(c). As (7) formulated, this peak value \((G_P/\omega)_\text{max}\) provides the information of \(D_{it}\) \((D_{it} \equiv C_d/\omega)\) at the FE/DE interface. And \(\tau_d\) can be obtained from the corresponding frequency \(\omega_{\text{peak}}\) where the peak occurs using (8)

\[
\left(\frac{G_P}{\omega}\right)_{\text{max}} = \frac{C_{DE}^2}{2(C_{FE} + C_{DE})C_{it}}
\]

\[
\omega_{\text{peak}} = \frac{C_{FE} + C_{DE} + C_{it}}{(C_{FE} + C_{DE})\tau_d}.
\]

However, a random distribution of discrete charges at the FE/DE interface makes the situation complicated and causes a deviation from the ideal case. With a nonuniform distribution of localized charges over the FE/DE interfacial plane, band bending fluctuation-induced \(\tau_d\) dispersion should be considered. The Gaussian distribution is the most common statistical distribution characterized by only mean value and variance, which can be applied to describe this \(\tau_d\) dispersion. Then, (2) for \(D_{it}\) extraction becomes

\[
\frac{G_P}{\omega} = \int_{-\infty}^{+\infty} \frac{(\omega \tau_d)C_{DE}^2C_{it}}{(\omega \tau_d)^2(C_{FE} + C_{DE})^2 + C_A^2} p(\tau) d\tau + \frac{G_L}{\omega}
\]

where \(P(\tau)\) is a probability distribution of \(\tau_d\) given by

\[
P(\tau) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left( -\frac{(\tau - \tau_{\text{it}})^2}{2\sigma^2} \right)
\]

where \(\sigma\) is the standard deviation of \(\tau_d\) and \(\tau_{\text{it}}\) is the mean value of \(\tau_d\). Finally, \(D_{it}\) and \(\tau_d\) at the FE/DE interface can be obtained by fitting the measured \(G_{L}\), \(G_P\), and \(C_P\) with the theoretical model given by (9) and (10).

It should be noted that the branch of \(R_0\) and \(C_{it}\) in Fig. 4(a) is connected to \(C_{FE}\) in parallel rather than \(C_{DE}\), because this scheme has the best fit to experimental data (shown in Section III-C). This circuit scheme reveals that the FE/DE interface traps are most likely to be intrinsically related to the FE layer. Meanwhile, another four schemes were also attempted: 1) connecting branch of \(R_0\) and \(C_{it}\) to \(C_{DE}\); 2) only considering \(G_{L}\) through DE layer; 3) only considering \(G_{L}\) through FE layer; and 4) separating \(G_{L}\) into two components including one through DE layer and another through FE layer. By modeling and calculating, all these schemes were examined to be against the experimental observations, which cannot follow the trend of \(C_P\) and even contribute to a peak of \(G_P/\omega\).

C. Extraction of \(D_{it}\) and \(\tau_d\)

Fig. 5(a) plots the measurement result of \(G_P/\omega\) as a relationship to log frequency with negative bias voltages applied. And the results under positive bias voltage are shown in Fig. 5(b). Clear conductance peaks with different peak values are observed at different bias voltages, which can be used for the extraction of \(D_{it}\) and \(\tau_d\). And this conductance method has also been applied to the control samples, the metal/ferroelectric/metal capacitors with pure HZO film and metal/insulator/metal capacitors with pure Al₂O₃ film. There is no conductance peak observed in these two capacitors.
Fig. 5. Experimental results of the $G_p/\omega$ versus $\log f$ measured at different (a) negative and (b) positive bias voltages in the 10-nm HZO/1-nm $\text{Al}_2\text{O}_3$ capacitor. (c) Leakage conductance $G_L$ of 10-nm HZO/1-nm $\text{Al}_2\text{O}_3$ under different bias voltages. (d) Experimental results of the $G_p/\omega$ versus $\log f$ at different voltages with 1 V/step, for the HZO/Al$_2$O$_3$ capacitors with the DE thickness of (d) 3 and (e) 5 nm.

Fig. 6. Summarized (a) conductance peak values $(G_p/\omega)_{\text{max}}$ and (b) measured frequencies at peaks under different bias voltages for the HZO/Al$_2$O$_3$ capacitors with different thicknesses of Al$_2$O$_3$.

Fig. 7. Experimental data (squares) and fitting results (solid lines) of the conductance curve as a function of $\log f$ taken from the 10-nm HZO/1-nm Al$_2$O$_3$ capacitor. The bias voltage ranges from $-3.5$ to $3.5$ V. Different color corresponds to that in Fig. 5.

Fig. 8. (a) Calculated Dit of the HZO/Al$_2$O$_3$ capacitors with different Al$_2$O$_3$ thicknesses. (b) Peak frequency values predicted by the model when $\tau_i$ is around 1.6 $\mu$s.
and [21] can be obtained. This accordance verifies that polarization switching, almost the same result with the data in section states. By integrating them into the energy band during the operation of FE/DE stack. Besides, the smaller $D_{it}$ and larger $\tau_{it}$ at low temperatures confirm the suppression of trap response. This conductance method provides a new approach and possible new insights to understand the mechanism of NC-FETs and Fe-FETs operations and their reliability degradations.

IV. CONCLUSION

In conclusion, the FE/DE interface is experimentally investigated in a simple capacitor structure using the conductance method with an improved circuit model. The components and connection of the circuit model for the conductance method are analyzed in detail, and the nonuniform trap distribution is also taken into consideration. With the proposed measurement technique, the $D_{it}$ at different static polarization states is obtained to be $\sim 4 \times 10^{12}$ to $10^{13}$ cm$^{-2}$ · eV$^{-1}$, indicating a critical role of charge trapping at the FE/DE interface during the operation of FE/DE stack. Stacks, the smaller $D_{it}$ and larger $\tau_{it}$ at low temperatures confirm the suppression of trap response. This conductance method provides a new approach and possible new insights to understand the mechanism of NC-FETs and Fe-FETs operations and their reliability degradations.

D. Behaviors of FE/DE Interface Traps at Low Temperature

To further understand the behaviors of FE/DE interface traps, the conductance method was also applied to 10-nm HZO/1-nm AlO$_3$ capacitors at low temperatures. Fig. 9(a) shows the experimental $G_{p/\omega}$ with a relationship to log frequency at zero bias voltage measured from RT to 110 K. Fig. 9(b) shows the extracted voltage-dependent $D_{it}$ at different temperatures. The peak value and frequency at peak both decrease with the decreasing measurement temperature.

As summarized in Fig. 10, $D_{it}$ at the FE/DE interface is smaller at lower temperatures, due to a weaker charge trapping/detrapping process, which suggests the reduction of leakage current through the DE layer and the suppression of trap response at the FE/DE interface. This phenomenon also agrees well with C–V measurement results in Fig. 3(b). In addition, $\tau_{it}$ was extracted and shown as the blue squares in Fig. 10. As the temperature decreases, a larger $\tau_{it}$ can be found. This can be attributed to the fact that charge injection is inhibited and trap response is slowed down.

References


