CHARACTERIZATION OF FIELD CYCLING FATIGUE IN HfZrO$_x$ FERROELECTRIC CAPACITORS

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ABSTRACT

In this paper, the fatigue behavior of HfZrO$_x$ (HZO) ferroelectric (FE) capacitor is thoroughly characterized. By proposing an empirical model applicable to the entire process of fatigue, we found that the fatigue effect is dominated by the process of charge migration to non-switching regions and charge exchange with electrodes to form the localized built-in field and cause domain-pinning.

INTRODUCTION

In recent years, Hf-based ferroelectric (FE) material has been regarded as the candidate for high-density nonvolatile memory devices due to its CMOS compatibility, scalability, high speed, and low operation voltage [1]. However, the endurance problem poses significant challenges for their practical adoption for mass production. One of its endurance problems is fatigue effect, which refers to the degradation of remnant polarization (P_r) under the bipolar field cycles. The main cause of fatigue is generally attributed to pinning of seed domains or domain walls by charged defects. New defect generation, defect movement and the process of charge trapping/de-trapping are possible origins of these charged defects [2]-[3], but a consensus on the dominant factor has not been reached. Moreover, current fatigue models of ferroelectrics are only applicable to certain stages of the decay process [4] or have shown inadequate fitting to the fatigue trend [5]-[6]. A comprehensive model that can accurately describe the entire fatigue process is still missing. Developing a quantitative fatigue model can aid in understanding the underlying physical mechanism and predicting the degradation of ferroelectrics during operation.

This paper presents a thorough investigation of the fatigue effect in HfZrO$_x$ (HZO) ferroelectric capacitors, including the development of a quantitative model that accurately describes the entire fatigue process. The model is applied to analyze fatigue at different temperatures, voltage amplitudes, and operation frequencies in detail, and the dominant factors for fatigue are revealed.

DEVICE FABRICATION

The capacitors used in this work are TiN/Zr:HfO$_2$/TiN (MFM) structures. Its fabrication process starts with the magnetron sputtering onto the SiO$_2$/Si wafer to form a 60 nm thick TiN bottom electrode. Secondly, 12 nm HZO thin film with Hf:Zr ratio of 1:1 was deposited by atomic layer deposition (ALD) at 250°C. Thirdly, photolithography was conducted to form patterned electrodes, and then 60 nm top electrode TiN was fabricated by magnetron sputtering. Finally, the as-fabricated HZO film was crystallized at 450°C for 1 minute in N$_2$ ambient.

CHARACTERIZATION AND MODELING OF FATIGUE

The pulse sequences to examine the switching kinetics and endurance of the MFM capacitor are shown in Fig. 1(a) and (b), respectively. To exclude the leakage current, positive-up-negative-down (PUND) pulse sequences were applied to measure the FE component.

A typical FE polarization charge density-time relationship is shown in Fig. 1(c), from which the relationship of switched polarization and the corresponding operation frequency of square wave can be extracted. As shown in the inset of Fig. 1(c), a switching time distribution tail can be found between 100kHz and 500kHz. The evolution of P_r with bipolar stress cycling at

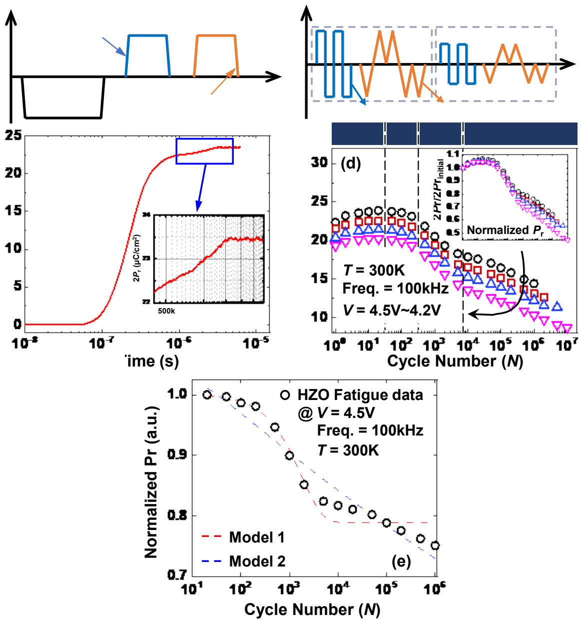

Figure. 1: The polarization-time relationship: (a) voltage setup and (c) experimental data. Inset: enlarged view of the box section. The switching time is transformed to the corresponding operation frequency. The evolution of P_r under different switching voltages: (b) voltage setup and (d) experimental data. Inset: the evolution of $Pr/Pr_{initial}$ with cycling. (e) Fatigue data fit by previous models, which are not applicable to the entire process of fatigue.

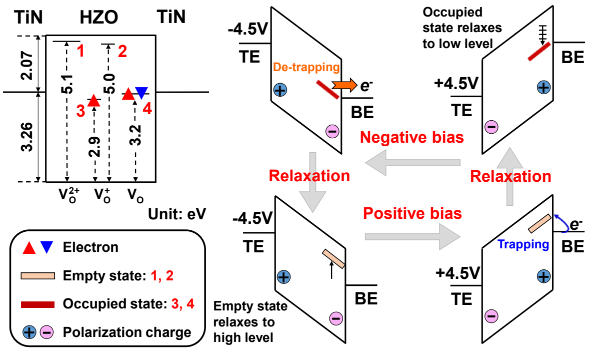

Figure 2: Energy level of oxygen vacancy in 0, +1 and +2 charge states [7], and the band diagram of the trapping and de-trapping process.

different voltage amplitudes are summarized in Fig. 1(d), which can be divided into four stages. Stage I is referred to as wake-up, where the P_r is increasing. Subsequently, the fatigue process starts with a reduction of P_r. Based on different degradation rates, this process can be further divided into three stages: stage II (slow), III (logarithmic), and IV (saturated). Our results indicate that lower pulse amplitudes lead to higher fatigue rates, which can be further confirmed by the normalized P_r values in the inset of Fig. 1(d). As shown in Fig. 1(e), fatigue models proposed for traditional FE materials fail in describing the fatigue data of HZO.

Defect generation, defect movement, and the trapping/de-trapping of charges are potential factors to induce charged defects and cause domain-pinning. However, if the first two ones are dominant factors, the fatigue rate should be faster at higher voltage, which is not the case in our experiments. Oxygen vacancy (V_O) is well recognized as the type of trap that can impact the FE properties during field cycling [7]. As the commonly used TiN electrode in the MFM capacitor is oxygen reactive, it is often partially oxidized during the ALD process and causes the aggregation of V_O at the interface. According to the formation energy given by DFT calculations [7], V_O is stable in the neutral or positively charged state in FE HZO. Hence, we assumed that FE domains are pinned by V_O^{2+} at the top or bottom interface and will be de-pinned upon the injection of electrons.

Charge redistribution within the ferroelectric film and charge exchange between the electrodes are two factors that can result in the charging of V_O and domain-pinning, which can help explain the lower fatigue rate at higher voltage amplitudes. Despite most domains are switched at the high electric field of ~3.5MV/cm, a distribution tail of switching field still exists, and some domains remain non-switched, as shown in Fig. 1(d). Thus, akin to the split-up effect [8], charges are redistributed to the non-switching region to form a localized built-in field with a direction that aligns with the FE polarization and cause the domain-pinning. This localized field will cause the FE domains in the adjacent switching region to align with the same direction, thereby enlarging the non-switching region. With smaller voltage, more non-switched regions exist, and the redistribution effect becomes more pronounced, resulting in a higher fatigue rate.

Another perspective to explain the fatigue trend is the charge exchange with electrodes. The band diagram in Fig. 2 shows an example of defects near the bottom electrode, while defects near the top electrode follow a symmetric process. When a negative bias is applied to the top electrode, electrons are de-trapped, and V_O tends to become positively charged, pinning the seed domains at the interface. When the polarity is changed, electrons tend to be trapped into the V_O^{2+} near the bottom electrode, and FE domains are de-pinned. Therefore, if bipolar stress is applied, trapping and de-trapping will occur simultaneously, and the defects will finally come to an equilibrium occupancy, resulting in a decrease in $2P_r$. Based on these analyses, we proposed an empirical model to describe the process of fatigue, as follows:

$$R(N) = \frac{2P_r\,(N)}{2P_{rmax}} = -Aexp(-BN^{-m}) + 1 \qquad (1)$$

$$\lim_{N\to 0} R(N) = 1 \qquad (2)$$

$$\lim_{N\to\infty} R(N) = 1 - A \qquad (3)$$

where R is the reliability function, representing the normalized P_r, A is a fitting parameter that decides the fatigue rate and ratio of pinned domains in the final state, and B and m are fixed parameters related to the energy and spatial distribution of defect. Next, fatigue behavior at different temperatures, voltages and frequencies is characterized and studied in detail with the proposed model.

The dependence of fatigue on temperature is first characterized. As shown in Fig. 3(a), the voltage amplitude and frequency are fixed at 4.5V and 100kHz, and our model can well reproduce the entire fatigue process as the temperature is varied from 275K to 350K. The fatigue rate increases with higher temperature, and $2P_r$ degrades to a lower final value. Since fatigue is a thermally activated process, it follows Arrhenius law:

$$A(T) \propto \exp\,(-E_a/\mathrm{k}_B T) \qquad (4)$$

where E_a is the activation energy, k_B is the Boltzmann constant, and T is temperature. As shown in Fig. 3(b), Ln(A) exhibits a good linear relationship with $1/T$, and E_a is extracted as 32meV, which is consistent to the calculated results in other studies [4]. Since the activation energy for defect generation (2eV [9]) and movement (0.7eV [10]) are both much higher than 32meV, they should not be the primary factor responsible for fatigue. Instead, this low E_a is not far from the phonon energy $\hbar\omega_0$ [11], indicating that P_r degradation is caused by the charged defects induced by electron de-trapping.

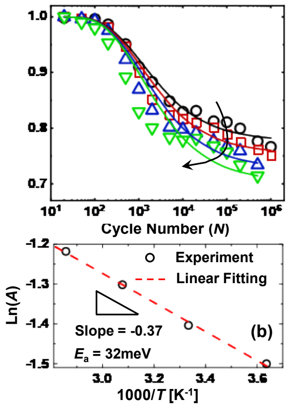

Figure. 3. (a) Experiment fatigue data and (b) extracted fatigue rate A at different temperatures.

Next, we analyze the fatigue data obtained at different voltage pulse amplitudes using the proposed model. Fig. 4(a) illustrates the voltage setup, and Fig. 4(b) shows the results. We observe that fatigue accelerates gradually as the voltage is lowered from 4.5V to 4.2V, and our new model can describe all the fatigue curves well. As shown in Fig. 4(c), the values of A we extract are linearly related to voltage, implying that fatigue rate is regulated by voltage amplitude through band bending. Since the defect energy level of empty state is higher than the occupied state [7], trapping at a positive bias is much harder than de-trapping at a negative bias (Fig. 2). With higher voltage amplitude, electron traps at higher energy levels can be pulled down closer to the Fermi level. This increases the possibility for electrons to be trapped and more domains to be de-pinned, resulting in a slower fatigue rate.

Finally, the effect of frequency on fatigue is investigated. Fig. 4(d)-(e) show the voltage setup and fatigue results from 100kHz to 500kHz, where our model can fit the fatigue curves well. As shown in Fig. 4(f), the extracted fatigue rate A follows a linear relationship with frequency within this specific range. At higher frequencies, electron trapping becomes more difficult because of the larger capture time constant induced by the higher energy level of the empty state. Therefore, de-trapping dominates and the fatigue rate increases. However, when the frequency drops below 10kHz, only very weak fatigue happens, which can even recover after a certain number of cycles, as shown in the inset of Fig. 4(e). This recovery can be attributed to the fact that a more complete switching and charge injection from the electrode can occur at low frequency, which reduces the non-switched region and inhibits the formation of charged V_O^{2+}.

Based on the analysis above, we can deduce that charge redistribution within the ferroelectric thin film and charge exchange with electrodes are dominant factors for P_r fatigue, instead of defect generation. Although new defects may be generated upon repeated field cycling, they would not affect the FE properties if they are in a neutral state. It is important to notice that breakdown is more likely to occur under higher voltage, which is different from the voltage dependence of fatigue. Therefore, the operating conditions should be chosen carefully to optimize the endurance of ferroelectric capacitor.

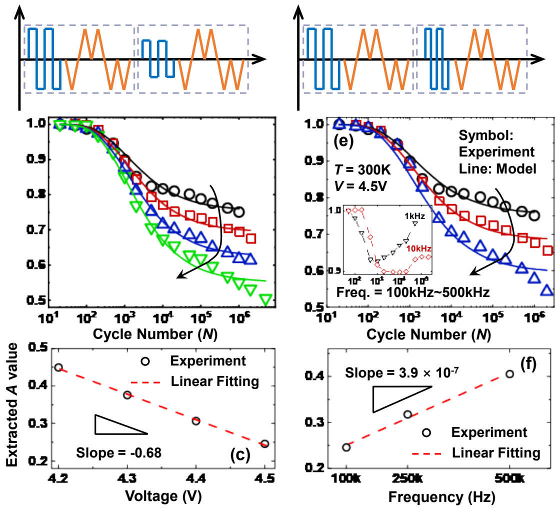

Figure. 4. (a) Voltage setup, (b) experiment fatigue data and (c) extracted fatigue rate A at different voltages. (d) Voltage setup, (e) experiment fatigue data and (f) extracted fatigue rate A at different frequencies.

CONCLUSION

In this work, we have conducted a comprehensive characterization of fatigue for HZO ferroelectric thin film. Our proposed empirical model has successfully captured the entire fatigue process, revealing that the fatigue rate is influenced by charge redistribution within the ferroelectric film and the charge exchange with the electrode, leading to the creation of charged defects and domain-pinning. Moreover, different voltage dependence of fatigue and breakdown implies that to find an appropriate operating condition is crucial for practical application.

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