



Insertion of an Ultra-thin Interfacial Aluminium Layer for the Realisation of a Hf_{0.5}Zr_{0.5}O₂ Ferroelectric Tunnel Junction.

Benoît Manchon, Greta Segantini, Nicolas Baboux, Pedro Rojo Romeo, Rabei Barhoumi, Dominique Drouin, Infante Ingrid C., F. Alibart, Bertrand Vilquin, Damien Deleruyelle

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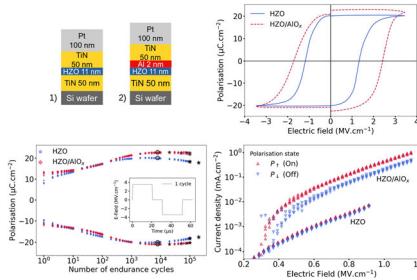
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3 B. Manchon,* G. Segantini, N. Baboux,
 4 P. Rojo Romeo, R. Barhoumi,
 5 I. C. Infante, F. Alibart, D. Drouin,
 6 B. Vilquin, D. Deleruyelle 2100585

7 **Insertion of an Ultrathin Interfacial
 8 Aluminum Layer for the Realization of a
 9 Hf_{0.5}Zr_{0.5}O₂ Ferroelectric Tunnel
 10 Junction**



Metal–insulator–ferroelectric–metal structure has recently been brought to the spotlight as a way to make Ferroelectric Tunnel Junction without the need for ultra-thin ferroelectric layer. Herein, the first use of an ultrathin Al layer is presented for the fabrication of an MFIM HZO-based FTJ through the scavenging of oxygen from the HZO during annealing. Herein, a small increase of remanant polarization, a large increase in conductivity and an On/Off ratio of up to 3.7, is reported.

1 Insertion of an Ultrathin Interfacial Aluminum Layer for 2 the Realization of a $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ Ferroelectric Tunnel 3 Junction

Q2 4 *Benoît Manchon,* Greta Segantini, Nicolas Baboux, Pedro Rojo Romeo,*
 5 *Rabei Barhoumi, Ingrid C. Infante, Fabien Alibart, Dominique Drouin, Bertrand Vilquin,*
 6 *and Damien Deleruyelle*

7 Herein, the effect of a 2 nm thin aluminum layer inserted between the ferro-
 8 electric layer and the top electrode in a TiN/ $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ /TiN stack deposited by
 9 reactive magnetron sputtering is investigated. The oxidation of the interfacial
 10 layer during annealing due to scavenging of the $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ impacts both the
 11 ferroelectric properties and the electrical conductivity of the junction. It is shown
 12 that the overall conductivity of the junction is boosted 20 folds while the
 13 resistance ratio between the positive and negative polarization states is increased
 14 from 1.3 up to 3.7. Through a systematic analysis of programming conditions,
 15 pulse duration, and height, we show that both the remanent polarization and
 16 On/Off current ratio can be enhanced at the expense of the endurance leading to
 17 a trade-off.
 18

1. Introduction

19 The unique properties of ferroelectric materials have been
 20 exploited to date in numerous integrated devices such as
 21 Ferroelectric Random Access Memories, Ferroelectric Field
 22 Effect Transistors, Negative Capacitance Field Effect
 23 Transistors, and Ferroelectric Tunnel Junctions (FTJs).^[1] The dis-
 24 covery of ferroelectricity in Si-doped hafnium oxide in 2011^[2]
 25 gave a significant boost to research on the topic. Although other
 26 successful dopants have been found,^[3] the solid solution

1 $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ (HZO) has attracted a lot of 1 interest.^[4] Hafnia-based thin films present 2 the double advantage of already being 3 widely used in the semiconductor industry 4 and of having a significantly lower crystal- 5 lization temperature as compared to perov- 6 skite ferroelectrics, with reported values as 7 low as 300 °C,^[5] allowing therefore full 8 compatibility with both front-end and 9 back-end of line integration. 10

11 FTJs are tri-layer structures in which two 12 electrodes consisting either of metals or 13 highly doped semiconductors sandwich a 14 thin ferroelectric layer. The internal field 15 of the ferroelectric layer attracts or repels 16 charges at the interfaces with the electro- 17 des. Due to differences in screening 18 lengths and permittivities of the electrode materials, the polariza- 19 tion direction induces a modulation of the potential barrier. 20 The polarization state can then be probed through the measure- 21 ment of tunneling currents. Although ferroelectric HZO has 22 been demonstrated down to 1 nm thick layers,^[6] scaling HZO 23 thickness down to a few nanometers while maintaining good 24 ferroelectric properties remains a real fabrication challenge.^[7] 25 The resistance variation in an ideal FTJ is only due to direct 26 tunneling; however, real FTJs are complex devices where 27 several competing charge transport mechanisms can occur 27

B. Manchon, N. Baboux, I. C. Infante, D. Deleruyelle
 INSA Lyon
 ECL
 CNRS
 UCBL
 CPE Lyon
 INL
 UMR5270
 Univ Lyon
 69621 Villeurbanne, France
 E-mail: benoit.manchon@insa-lyon.fr

B. Manchon, D. Drouin
 Institut Interdisciplinaire d'Innovation Technologique (3IT)
 Université de Sherbrooke
 Sherbrooke, Québec, Canada

B. Manchon, D. Drouin
 Laboratoire Nanotechnologies Nanosystèmes (LN2) CNRS UMI-3463
 Québec, Canada

Q3
 Q4

G. Segantini, P. Rojo Romeo, R. Barhoumi, B. Vilquin
 ECL
 INSA Lyon
 CNRS
 UCBL
 CPE Lyon
 INL
 UMR5270
 Univ Lyon
 69130 Ecully, France

F. Alibart
 Institute of Electronics
 Microelectronics and Nanotechnology (IEMN)
 Université de Lille
 Villeneuve d'Ascq, France

1 simultaneously. In addition, more than one physical mechanism
 2 can be involved in the resistance modulation. In this work, we
 3 will define FTJs as ferroelectric devices, which exhibit a change
 4 in their electrical resistance depending on polarization direction.
 5 Therefore, the term “tunneling currents,” in its broad sense, will
 6 refer to all charge transport modes across a dielectric (such as
 7 direct tunneling, Fowler–Nordheim tunneling, thermionic emission,
 8 and so on) rather than just direct tunneling. Recently, a
 9 slightly different structure has been brought in the spotlight.
 10 Theorized by Meyer et al.,^[8] the addition of an ultrathin dielectric
 11 layer, resulting in a metal/ferroelectric/insulator/metal (MFIM)
 12 structure, enables conductance modulation with thicker
 13 ferroelectric layers. The inserted dielectric layer plays the role
 14 of tunneling barrier while the ferroelectric layer acts as a
 15 polarization switch. In this context, several devices exploiting
 16 Al_2O_3 and TiO_2 interfacial layers have recently been reported
 17 in the literature.^[9–12]

18 An important parameter in the stabilization of the ferroelectric
 19 phase in doped HfO_2 is the oxygen content. It has mainly been
 20 studied in atomic layer deposition processes and the results indicate
 21 that the formation of the orthorhombic and tetragonal phases
 22 can be promoted by oxygen vacancies.^[13] In this work,
 23 the affinity of Al for oxygen atom was exploited to combine
 24 the fabrication of an MFIM junction with the generation of oxygen
 25 vacancies in HZO layer to promote the ferroelectric phase.
 26 A set of twin samples was fabricated to study the influence of
 27 an ultrathin interfacial Al layer. Their structure is depicted in
 28 **Figure 1**. A symmetrical TiN/HZO/TiN stack serves as reference
 29 to the second sample which had a 2 nm thick layer of Al inserted
 30 at the top electrode interface: TiN/HZO/Al/TiN. The ferroelectric
 31 HZO layer is 11 nm thick for both samples. Although
 32 symmetrical electrodes cannot, in theory, produce an On/Off
 33 ratio, symmetrical FTJs have been demonstrated due to interfa-
 34 cial differences between the top and bottom contact (interface
 35 reconstruction, exposure to air, and so on).^[14] It is theorized that
 36 the Al layer formed a dielectric AlO_x layer during the annealing
 37 step by scavenging oxygen atoms from the HZO layer. We report
 38 the first use of an oxygen scavenging ultrathin Al layer for the
 39 fabrication of an HZO-based FTJ. The aim was to obtain the
 40 same effect as an interfacial layer of Al_2O_3 to increase On/Off
 41 ratio of FTJ devices while also generating oxygen vacancies,

which play an important yet not fully understood role in the
 1 stabilization of the ferroelectric orthorhombic phase.
 2

2. Results and Discussion

The glancing incidence X-ray diffraction (GI-XRD) patterns shown in Figure 1a indicate the poly-crystalline nature of the HZO layers. The position of the peak at $2\theta \approx 30.5^\circ$ is consistent with the theoretical value of the orthorhombic/tetragonal phase for the (111) plane and confirms the presence of the ferroelectric phase. The monoclinic (11–1) and (111) peaks at $2\theta \approx 28.4^\circ$ and $2\theta \approx 31.6^\circ$, respectively, are also detected. Using pseudo-Voigt functions that account for $\text{Cu K}\alpha 1$ and $\text{K}\alpha 2$ radiations to fit the data shown in Figure 1b, an analysis of the integrated areas corresponding to the orthorhombic and monoclinic peaks was performed. The presence of orthorhombic phase in the sample with the Al layer was found to be higher by a factor 3 when compared to the one without Al, suggesting that the Al layer helped the formation of the ferroelectric phase. This is consistent with the idea of oxygen vacancies helping the formation of the orthorhombic phase. However, these statements are mitigated by the fact that the diffraction signal originates both from HZO capped and uncapped by the top electrode. The importance of tensile strain provided by top electrode capping in the stabilization of the orthorhombic phase is well documented.^[15,16] HZO not covered by the top electrode is unlikely to form the orthorhombic phase and the monoclinic phase is therefore overevaluated. Here, the surface ratio of HZO covered by the top electrode is about 50%. In addition, the overlap of the orthorhombic and tetragonal peaks makes the quantitative analysis of the two phases rather difficult.

The relative permittivity (ϵ_r) against voltage plots obtained from capacitance measurements plotted on Figure 1c exhibits the characteristic butterfly shape of ferroelectric capacitors. The extracted relative permittivity of the HZO/ AlO_x stack is lower than that of the reference as would be expected with the addition of a second dielectric layer. This finding is in favor of our assumption that the deposited Al has been oxidized during annealing. To further support this statement, the theoretical permittivity versus voltage curve of the HZO/ AlO_x sample, $\epsilon_r^{\text{computed}}$,

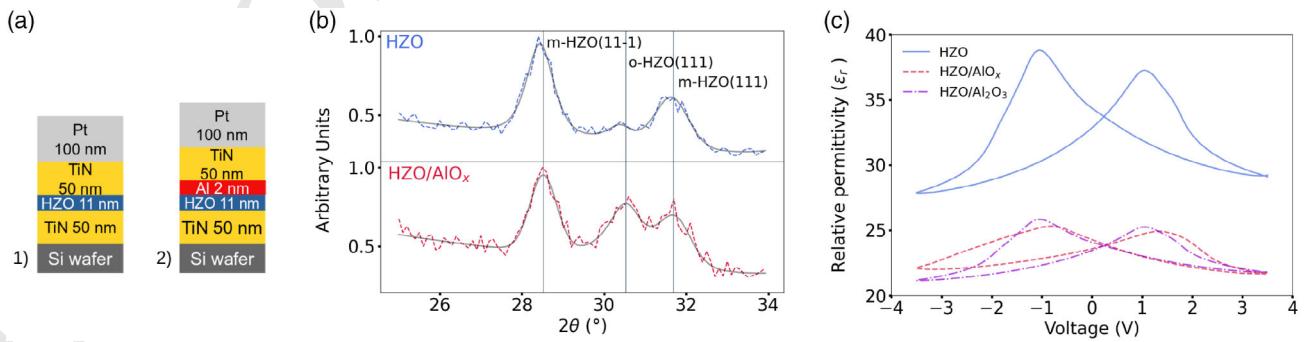


Figure 1. a) Structure of the two samples studied in this work, not to scale. Typical device diameters studied are 20, 50, and 100 μm . b) GI-XRD pattern corresponding to each sample. The fits were obtained using pseudo-Voigt functions accounting for the $\text{Cu K}\alpha 1$ and $\text{K}\alpha 2$ radiations. c) Relative permittivity obtained from capacitance versus voltage sweeps taken between ± 3.5 V on both samples. The relative permittivity of the $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ (HZO)/ AlO_x corresponds to the average of the double layer. The HZO/ Al_2O_3 curve is computed from the reference HZO by adding a theoretical 2 nm layer with a relative permittivity of 9.1 corresponding to Al_2O_3 .

1 was computed from the permittivity curve of the HZO reference
 2 sample, ϵ_r^{HZO} with thickness $d_1 = 11 \text{ nm}$, by adding an interfacial
 3 series capacitance of thickness $d_2 = 2 \text{ nm}$ and relative permittiv-
 4 ity ϵ_r^{IL} using Equation (1). As shown in Figure 1c, a very good
 5 match was achieved using $\epsilon_r^{\text{IL}} \approx 9.1$, a value fully consistent with
 6 the one of Al_2O_3 .^[17]

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$$\epsilon_r^{\text{computed}} = \frac{\epsilon_r^{\text{HZO}} \cdot \epsilon_r^{\text{IL}}}{d_2 \cdot \epsilon_r^{\text{HZO}} + d_1 \cdot \epsilon_r^{\text{IL}}} \quad (1)$$

7 The ferroelectric properties were then investigated by cycling
 8 the devices and regularly taking positive-up-negative-down
 9 (PUND) measurements.^[18] The assumption is made that the
 10 non-ferroelectric contributions to current are exactly the same
 11 in a switching and a non-switching pulse and thus the ferroelec-
 12 tric current can be isolated by simple subtraction. The positive
 13 and negative remanent polarizations (P_r) for five devices of each
 14 sample cycled at equal electric field are plotted **Figure 2a**.
 15 The device to device variation of the remanent polarization is
 16 very low with an average standard deviation to the mean of less
 17 than 1% for the five measurements on each sample. The maxi-
 18 mum $2P_r$ during cycling is slightly increased from 40.5 to
 19 $43.8 \mu\text{C cm}^{-2}$ with the addition of the Al interfacial layer.
 20 However, the overall endurance is impacted; the reference sam-
 21 ple devices all reached at least 1×10^5 cycles while three of the
 22 AlO_x devices broke down before, with one as early as 2×10^4 .

These changes may be attributed to the oxygen vacancies that
 1 were introduced, improving remanent polarization by increasing
 2 the orthorhombic phase content but precipitating breakdown
 3 due to a higher defect density. Oxygen vacancies are thought
 4 to help the formation of the orthorhombic phase by reducing
 5 the phases relative free energy, reducing the grain size or by play-
 6 ing the role of nucleation sites.^[19] However, oxygen vacancies are
 7 also associated with degraded ferroelectric properties past the sta-
 8 bilization process playing a key role in wake up, fatigue, and
 9 probably leading to an early dielectric breakdown.^[20] Here, it
 10 seems a compromise is reached in the AlO_x sample as remanent
 11 polarization was improved at the expense of endurance while the
 12 wake-up and fatigue behavior remained relatively unchanged.
 13 **Figure 2c** shows the current–voltage characteristics recorded dur-
 14 ing the switching pulses of a PUND measurement after 5×10^3
 15 cycles while **Figure 2b** shows the extracted polarization–voltage
 16 (P–V) curves after the current from the non-switching pulses was
 17 subtracted. The introduction of the interfacial layer has increased
 18 the coercive field and created a clear asymmetry in the device’s
 19 electrical properties. Polarization switching occurs at a higher
 20 (absolute) voltage than for the reference while being more
 21 gradual. This turns into an increased effective coercive field
 22 for both regions (from -1.2 to -1.7 MV cm^{-1} and from 1.3 to
 23 2.4 MV cm^{-1}) and a softer slope under negative field in the
 24 P–V loop. The increase of the coercive field can be related to
 25 the voltage drop through the interfacial dielectric layer making
 26

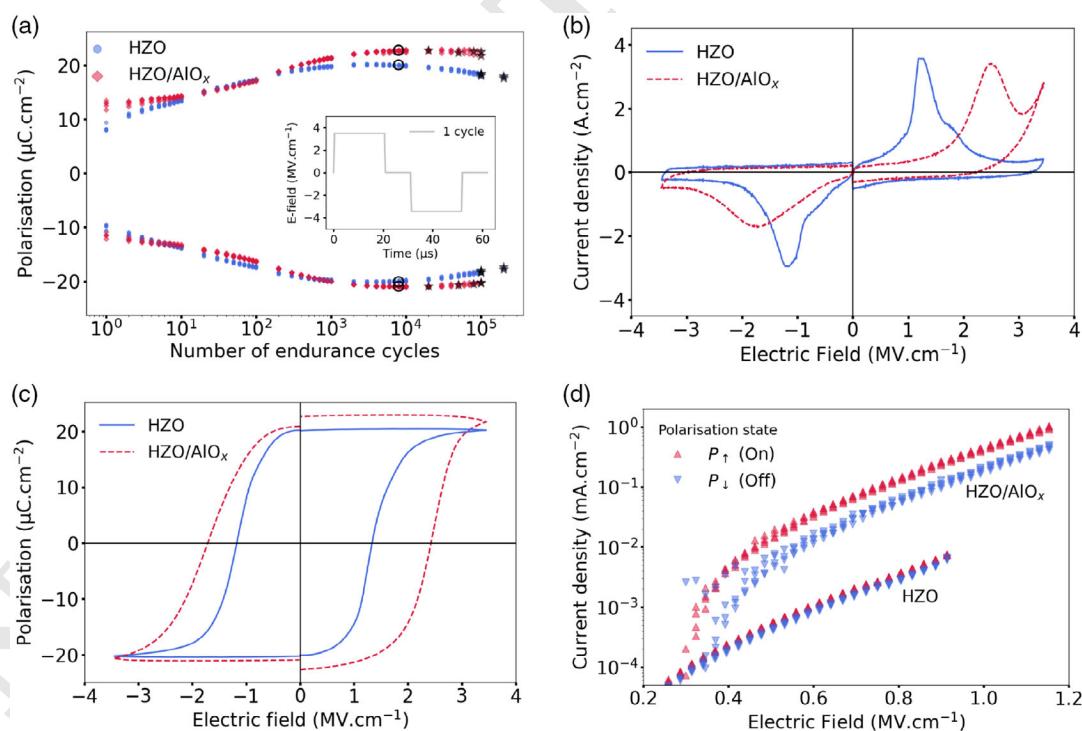


Figure 2. a) Remanent polarization for the reference and the AlO_x sample during cycling until dielectric breakdown. Five of each device were cycled. Stars mark dielectric breakdown. The inset represents one cycle with pulse parameters of $\pm 3.8 \text{ V}$ for the reference and 4.5 V for the AlO_x sample $\approx 3.5 \text{ MV cm}^{-2}$, 500 ns ramps, 20 ms plateau, and 10 ms pause between pulses. b) Current–voltage characteristics of the two samples under applied triangular pulses with $60 \mu\text{s}$ ramps. These data, as the following polarization curves, are measurements taken after 5×10^3 cycles (data point marked with an empty circle on the endurance graph). c) Polarization versus voltage loops for both samples extracted from the positive-up-negative-down (PUND) measurements. d) Voltage sweeps under the coercive field in the On and Off states for both devices. The measurements were repeated four times.

1 the voltage requirement higher to reach the coercive field in the
 2 HZO layer. One might wonder why Figure 1c does not show the
 3 same change in coercive field. It is because the electrical mea-
 4 surement conditions are rather different. The voltage range is
 5 not the same and the voltage sweep is quasi-static during the
 6 CV measurement as opposed to dynamic for the PUND.
 7 Interestingly, the tunneling currents are also drastically
 8 increased in the AlO_x sample, most likely due to the number
 9 of oxygen vacancies assisting conduction between electrodes.^[21]
 10 This feature is advantageous for FTJ applications since it allows
 11 larger sensing current at low voltages. The increased coercive
 12 field may also be beneficial for FTJ applications by allowing a
 13 wider range of accessible voltages for read operation without
 14 altering the polarization state, therefore giving access to even
 15 higher read currents.

16 Figure 2d shows the plots of current density against electric
 17 field taken at low voltages with the polarization facing one or
 18 the other electrode for both samples. The measurements were
 19 taken 4 times to show repeatability. The On/Off ratio is increased
 20 from ≈ 1.3 to ≈ 2.4 with the introduction of the Al layer. The
 21 small On/Off ratio of the symmetrical device is attributed to a
 22 slight interfacial difference due to exposure to air between
 23 HZO and top TiN deposition. The maximum current density
 24 available for operation in the Off state jumped up from 6.35
 25 to 451 mA cm^{-2} . This is a key parameter for dynamic sensing;
 26 for instance, such an increase would allow the reduction of

the FTJ diameter from 71 to 8.4 mm to reach a 1 nA sensing 1
 current. 2

3 The effect of pulse parameters on HZO/ AlO_x samples was 3
 4 then investigated by varying pulse widths and heights while mon- 4
 5 itoring both the remanent polarization and the On/Off current 5
 6 ratio. Both properties increase with pulse width (Figure 3a) and 6
 7 height (Figure 3c) at the expense of endurance. There is a 7
 8 trade-off that must be considered when choosing operation 8
 9 parameters. If the endurance appears to follow a more hectic 9
 10 dependency, it is due to the statistical nature of dielectric 10
 11 breakdown. The presence of defects can significantly impact 11
 12 the maximum endurance. The increase in maximum remanent 12
 13 polarization is monotonous for both pulse parameters (Figure 4). 13
 14 For best performances, the pulse parameters are 20 ms duration, 14
 15 as it offers the highest On/Off ratio while remaining relatively 15
 16 stable throughout the lifetime of the device, and 3.5 V since it 16
 17 provides the highest endurance. However, for the On/Off ratio, 17
 18 the pulse duration has the biggest influence with not only the 18
 19 value increasing from less than 1.5 to more than 3.5 but also 19
 20 the value becoming increasingly unstable during cycling. 20
 21 Voltage only influences the On/Off ratio up to 3.5 V where a form 21
 22 of saturation is reached. While maximum polarization more than 22
 23 doubles when voltage is increased from 3.5 to 4.5 V, the On/Off 23
 24 ratio remains relatively unchanged. This suggests the origin of 24
 25 the conductivity modulation is not directly dependent on the 25
 26 polarization field or at least not linearly. The oxygen vacancies 26

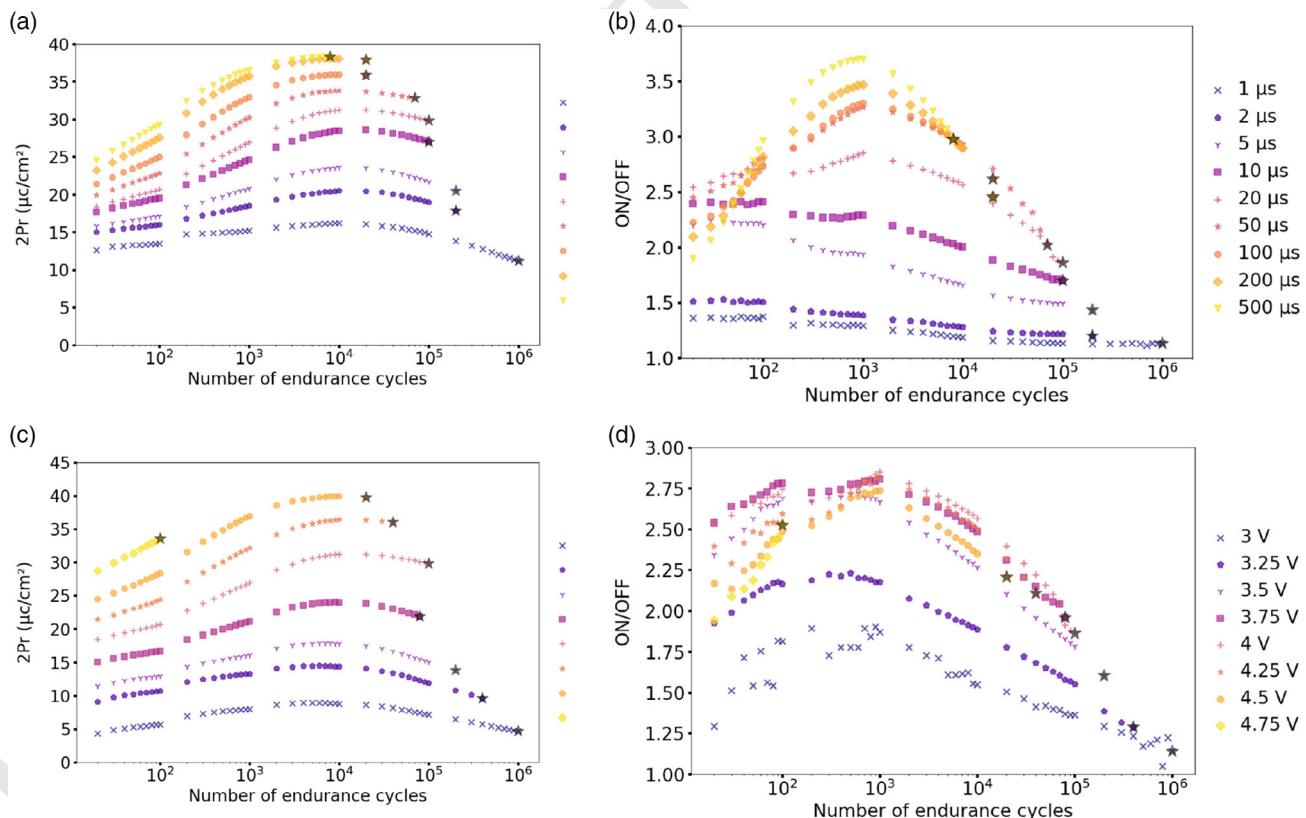


Figure 3. Evolution of remanent polarization a) and On/Off ratios for the AlO_x sample b) during endurance tests carried out with cycling pulse width varying from 1 to 500 μs at 4 V. Evolution of c) polarization and d) On/Off ratio while cycling with pulse heights ranging from 3 to 4.75 V with pulse duration of 20 μs . Stars mark dielectric breakdown.

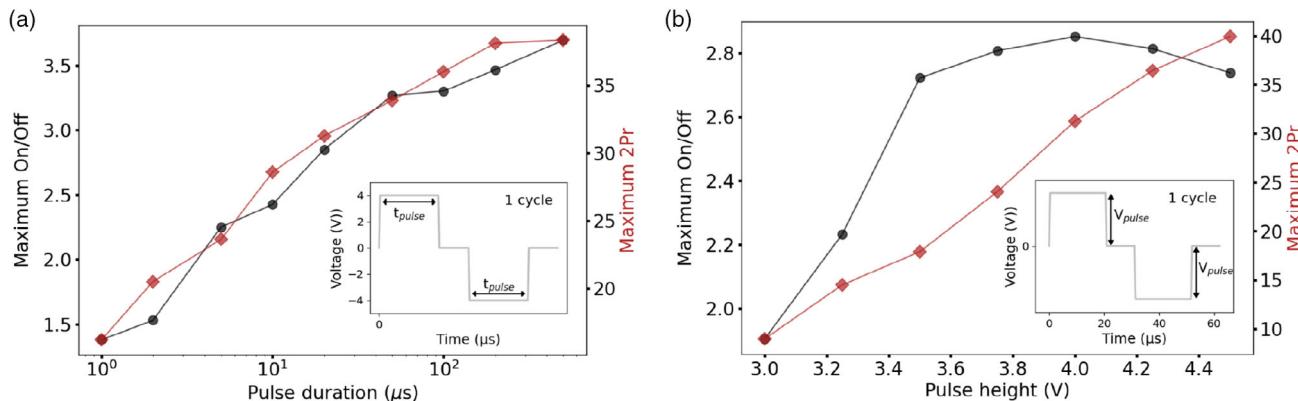


Figure 4. Maximum On/Off current ratios (dots) and remanent polarizations (diamonds) as a function of a) pulse duration and b) height. Insets represent the cycling pulses that were applied with the relevant parameters, as indicated.

1 migrating under the effect of the applied external field and the
 2 ferroelectric polarization are thought to be an important factor at
 3 play. We suspect that, similarly to the memristor model
 4 presented by Strukov et al.,^[22] the oxygen vacancies which are
 5 packed at the AlO_x/HZO interface migrate in and out of the
 6 ferroelectric layer under the effect of the applied field and of
 7 the ferroelectric polarization, lowering and increasing resistivity
 8 by creating available states for trap-assisted tunneling. Carrying
 9 out current–voltage measurement over an extended temperature
 10 range would be helpful to elucidate the prevalence of the differ-
 11 ent resistance modulation mechanisms and it will be the subject
 12 of further investigations.

13 3. Conclusion

14 The impact of inserting an ultrathin Al layer in HZO-based FTJ
 15 was investigated. With respect to reference devices, it is shown
 16 that the insertion of the Al layer leads to an increase of both the
 17 remanent polarization and tunneling currents. Oxidation of the
 18 Al layer into interfacial AlO_x by scavenging oxygen ions was
 19 found to be consistent with CV measurements. It is therefore
 20 believed that oxygen vacancies generated through oxidation of
 21 the Al layer promoted the formation of the orthorhombic phase
 22 of HZO while actively participating to the increase of leakage cur-
 23 rents. The On/Off current ratio was improved from 1.3 to up to
 24 3.5 in conjunction with a drastic increase of electrical conductiv-
 25 ity, which is beneficial for FTJ operations. Through studying the
 26 effect of cycling pulse amplitude and duration, it is shown that
 27 both the remanent polarization and On/Off current ratio can be
 28 tailored but they are increased at the expense of endurance.

29 4. Experimental Section

30 *Device Fabrication:* The fabrication process of the two samples was
 31 thought out to have as little difference between them as possible. A piece
 32 of n^+ Si (100) Si wafer was first cleaned using acetone and ethanol fol-
 33 lowed by a bath in a buffer oxide etching (BOE) solution to remove the
 34 native oxide layer. The 50 nm thick TiN bottom electrode was then depos-
 35 ited from a Ti target and a N_2 plasma by reactive magnetron sputtering
 36 inside an AC450-sputtering machine by Alliance Concept (all deposition

steps were carried out in this equipment). HZO of 11 nm was deposited
 1 by nonreactive sputtering of a ceramic $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ target. The piece of Si
 2 was then cleaved in halves for the UV lithography step preparing for the
 3 liftoff. On one half, 50 nm of TiN and 100 nm of Pt thick circular pads were
 4 added as top electrode. The purpose of the Pt was to improve the electrical
 5 contact with the metallic tips used for electrical characterization. An ultra-
 6 thin Ti layer was necessary for the adhesion of Pt on TiN. The other half
 7 was subjected to the same fabrication steps with the addition of a 2 nm Al
 8 layer between the HZO and the top electrode TiN. The exact sputtering
 9 parameters could be found in the work by Bouaziz et al.^[23] Both resulting
 10 samples were annealed in a tubular furnace at 450 °C under N_2 atmos-
 11 phere for 30 min.

12 *Device Characterization:* The thicknesses of the layers were verified by
 13 X-ray reflectometry (XRR) and structural properties were measured by
 14 GI-XRD with a Rigaku Smartlab diffractometer.

15 Electrical characterization was carried out using a Keithley 4200-SCS
 16 Semiconductor Parameter Analyzer Characterization System equipped
 17 with SMUs, RPMs, and CVUs. Voltage was applied through the bottom
 18 electrode by contacting a dielectrically broken-down device and the top
 19 electrode of the device of interest. For the study of the impact of pulse
 20 width and height, the cycling pulses were square shaped with 500 ns
 21 ramps and 10 μ s pauses at 0 V between them. The duration was varied
 22 first, from 1 to 500 μ s with the height maintained at 4 V. The voltage
 23 was then increased from 3 to 4.75 V with a pulse width fixed at 20 μ s.
 24 Each pulse form was applied once to one device until breakdown. Two
 25 reading protocols were used during cycling: a PUND sequence to measure
 26 polarization followed by two low voltage pulses to read the current value
 27 with a programming pulse in between to switch polarization state. The
 28 PUND pulses were triangular with ramps of 20 μ s and maximum voltage
 29 equal to cycling voltage. Read pulses were 1 V high and 5 ms long with
 30 500 ns ramps. The devices were 100 μm in diameter. The exact setup
 31 for electrical characterization was also described by Bouaziz et al.^[23]

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40 Conflict of Interest

41 The authors declare no conflict of interest.

1 Data Availability Statement

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

4 Keywords

5 aluminum, ferroelectric HZO, ferroelectric tunnel junction, thin films

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