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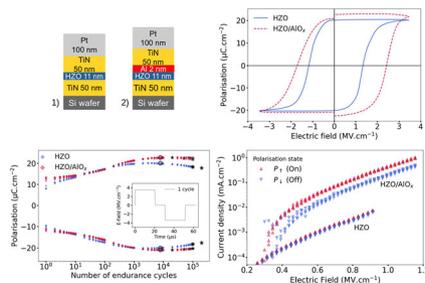
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Insertion of an Ultrathin Interfacial Aluminum Layer for the Realization of a $\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ Ferroelectric Tunnel Junction



Metal–insulator–ferroelectric–metal structure has recently been brought to the spotlight as a way to make Ferroelectric Tunnel Junction without the need for ultrathin 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

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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

$\text{Hf}_{0.5}\text{Zr}_{0.5}\text{O}_2$ (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

FTJs are tri-layer structures in which two 11
electrodes consisting either of metals or 12
highly doped semiconductors sandwich a 13
thin ferroelectric layer. The internal field 14
of the ferroelectric layer attracts or repels 15
charges at the interfaces with the electro- 16
des. Due to differences in screening 17

lengths and permittivities of the electrode materials, the polariza- 18
tion direction induces a modulation of the potential barrier. 19
The polarization state can then be probed through the measure- 20
ment of tunneling currents. Although ferroelectric HZO has 21
been demonstrated down to 1 nm thick layers,^[6] scaling HZO 22
thickness down to a few nanometers while maintaining good 23
ferroelectric properties remains a real fabrication challenge.^[7] 24
The resistance variation in an ideal FTJ is only due to direct 25
tunneling; however, real FTJs are complex devices where 26
several competing charge transport mechanisms can occur 27

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

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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 emis-
8 sion, 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 Al₂O₃ and TiO₂ 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 HfO₂ is the oxygen content. It has mainly been
20 studied in atomic layer deposition processes and the results indi-
21 cate that the formation of the orthorhombic and tetragonal
22 phases 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 oxy-
25 gen 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 ferroelec-
31 tric 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 interfacial
34 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 Al₂O₃ 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
stabilization of the ferroelectric orthorhombic phase.

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 Cu K α 1 and K α 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 com-
pared 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 ortho-
rhombic 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 cov-
ered 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 per-
mittivity versus voltage curve of the HZO/AlO_x sample, $\epsilon_r^{\text{computed}}$,

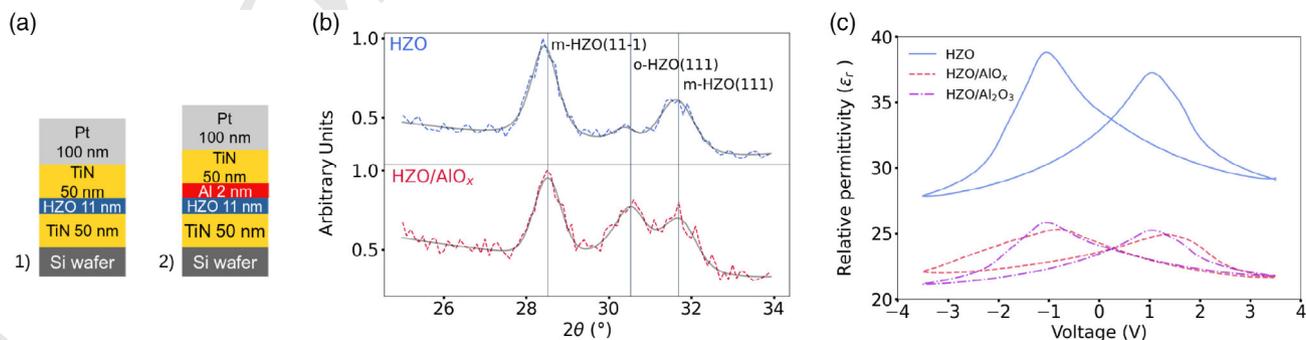


Figure 1. a) Structure of the two samples studied in this work, not to scale. Typical device diameters studied are 20, 50, and 100 μm . b) GIXRD diffraction pattern corresponding to each sample. The fits were obtained using pseudo-Voigt functions accounting for the Cu K α 1 and K α 2 radiations. c) Relative permittivity obtained from capacitance versus voltage sweeps taken between ± 3.5 V on both samples. The relative permittivity of the Hf_{0.5}Zr_{0.5}O₂ (HZO)/AlO_x corresponds to the average of the double layer. The HZO/Al₂O₃ curve is computed from the reference HZO by adding a theoretical 2 nm layer with a relative permittivity of 9.1 corresponding to Al₂O₃.

1 was computed from the permittivity curve of the HZO reference
2 sample, ϵ_r^{HZO} with thickness $d_1 = 11$ nm, by adding an interfacial
3 series capacitance of thickness $d_2 = 2$ 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

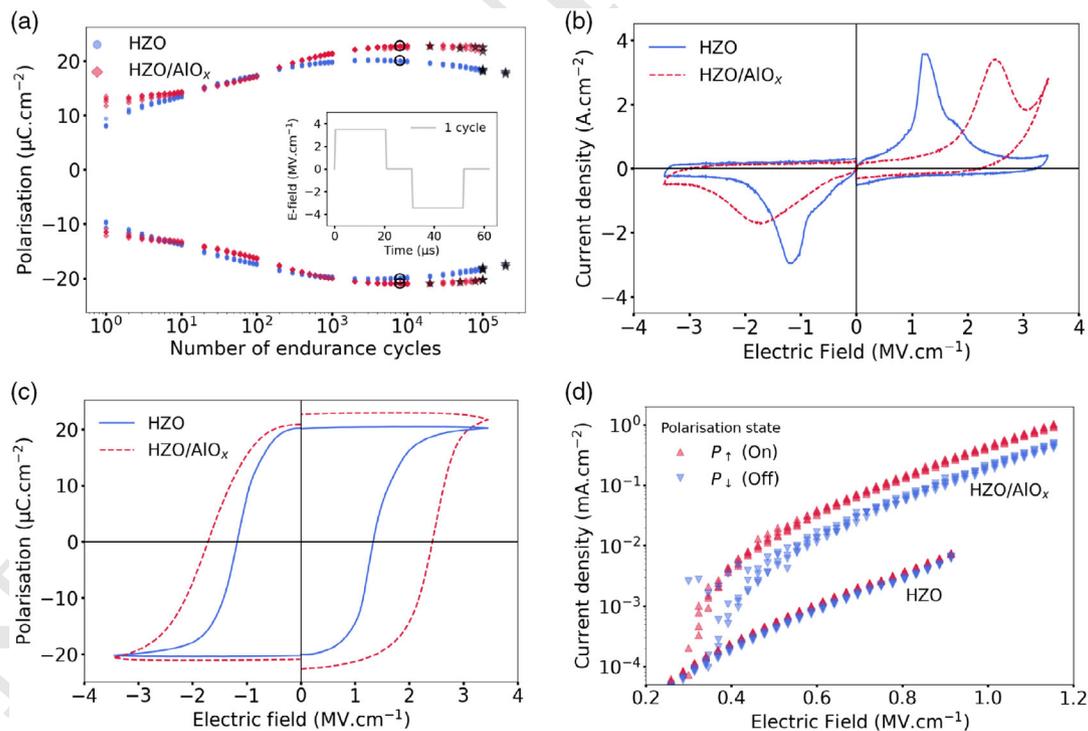


Figure 2. a) Remanent polarization for the reference and the AlO_x sample during cycling until dielectric breakdown. Five of each device were cycled. Stars mark dielectric breakdown. The inset represents one cycle with pulse parameters of ± 3.8 V for the reference and 4.5 V for the AlO_x sample $\approx 3.5 \text{ MV cm}^{-2}$, 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 μs 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 μm to reach a 1 nA sensing
current.

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

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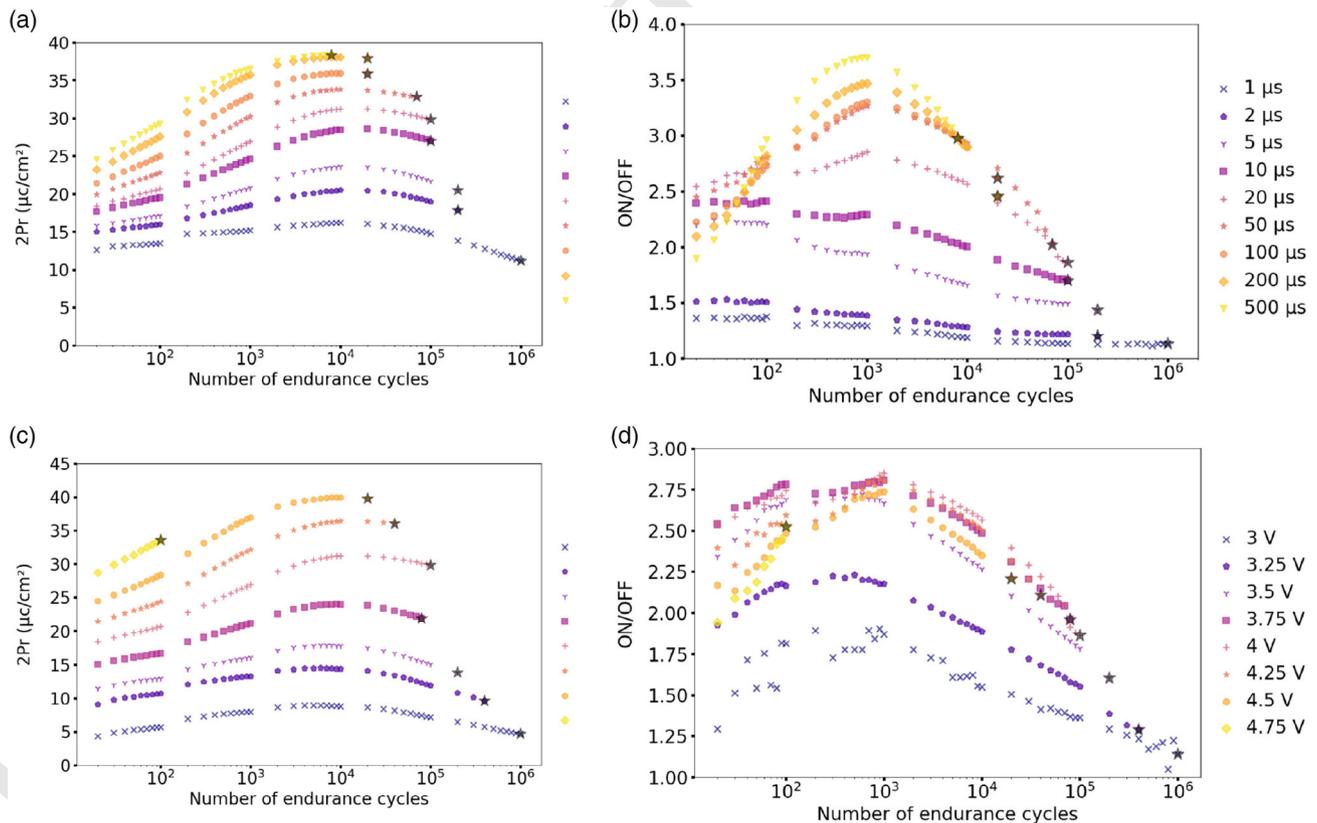


Figure 3. Evolution of remanent polarization a) and On/Off ratios for the AlO_x sample b) during endurance tests carried out with cycling pulse width varying from 1 to 500 μs at 4 V. Evolution of c) polarization and d) On/Off ratio while cycling with pulse heights ranging from 3 to 4.75 V with pulse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

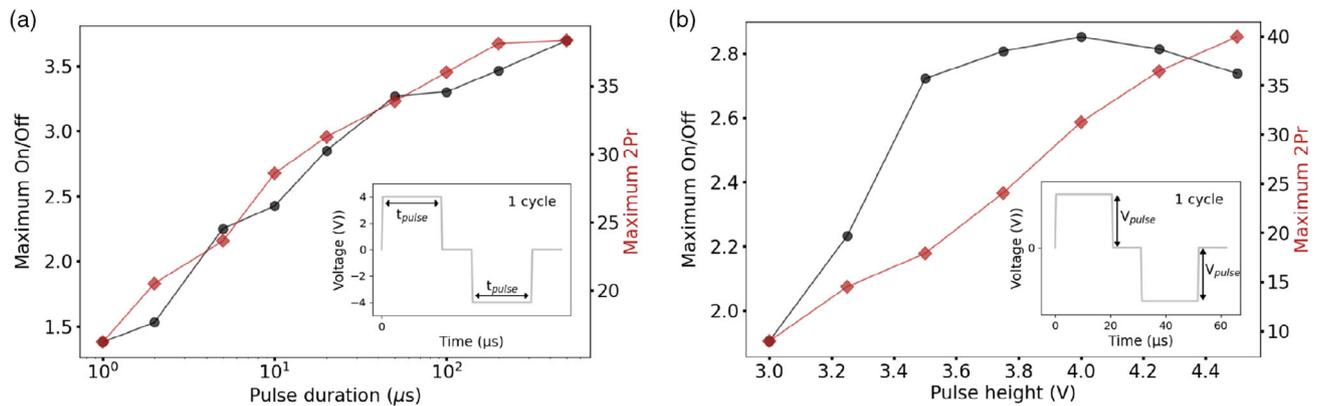


Figure 4. Maximum On/Off current ratios (dots) and remanent polarizations (diamonds) as a function of a) pulse duration and b) height. Insets represent the cycling pulses that were applied with the relevant parameters, as indicated.

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
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 N₂ 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 Hf_{0.5}Zr_{0.5}O₂ 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 N₂ atmo-
11 sphere 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 μs pauses at 0V between them. The duration was varied
22 first, from 1 to 500 μs with the height maintained at 4V. The voltage
23 was then increased from 3 to 4.75 V with a pulse width fixed at 20 μs.
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 μs 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]
32

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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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