Imprint issue during retention tests for HfO2 -based FRAM: An industrial challenge?

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ABSTRACT

For ferroelectric random access memory (FRAM) with HfO_2-based materials, the wake-up effect and the imprint have to be limited. Here, the electrical behavior of different samples is investigated during retention tests on woken-up samples at room temperature. Retention properties are compared during tests with or without alternations of voltage pulses with opposite signs. First, during retention tests with alternations, the imprint oscillates between two values that are believed to be too high for the reading operation of industrial FRAM memories. This imprint oscillation is not the sole cause of remanent polarization loss. Second, the wake-up effect and retention loss appear to be closely linked. Finally, two retention fitting models are tested: the first one follows a power law t^{-n} and the second one corresponds to a stretched exponential behavior $\exp(-(t/\tau)^{\beta})$. The data cannot be fitted by the power law at all, while the stretched exponential can fit the data after $t > 100$ s. In fact, the stretched exponential model highlights that the remanent polarization reduction during retention tests can be separated into at least two parts: a behavior for a short period of elapsed time and a behavior after a long period of elapsed time. The origins of this two-part remanent polarization loss behavior are discussed.

Since the discovery in 2011 by Böscke *et al.*[1] of ferroelectricity in Si-doped HfO_2, there has been a great deal of effort to overcome reliability issues in HfO_2-based materials. The most addressed phenomenon is the wake-up effect, but recently, imprint has also drawn a great deal of attention,[2–8] being considered the main cause of polarization reduction during data retention. Both the imprint and wake-up effects are temperature-dependent phenomena.[9,10]

Different definitions of the imprint have been given across the years.[11–13] In this paper, for simplification, imprint will be defined as the shift of the P–E hysteresis loop when the number of cycles increases during endurance tests or when the time increases throughout retention tests.

Retention is the ability of ferroelectrics to store a state over time. According to Park *et al.*,[14] HfO_2-based ferroelectric field effect transistors (FeFETs) have a retention higher than 10 years, greater than perovskite-based FeFETs, because of the difficulties involved in integrating perovskites on silicon.[15] Nonetheless, the present article is focused on ferroelectric random access memory (FRAM) in the one transistor–one capacitor (1T–1C) architecture since metal/ferroelectric/metal (MFM) capacitors are investigated here. In this architecture, retention is measured for quantities linked to the remanent polarization (P_r) over time. For instance, retention can be described by $2P_r$ or by the switching polarization (P_{sw}), which is equal to $|P_r^+| + |P_r^-|$. Only four papers have explicitly mentioned a retention higher than 10 years for HfO_2-based FRAM,[3,16–18] while other articles have mentioned good retention properties without extrapolation.[3,5,19,20] The underlying reason is the lack of satisfying models for polarization retention in HfO_2-based MFM capacitors.

Retention characteristics are generally considered to be influenced by two parameters: polarization relaxation and imprint. The root cause of imprint is considered to be a stronger charge trapping at one of the interfaces between the ferroelectric and one of the two electrodes, while polarization relaxation might be caused by a depolarization built-in bias, which allows the domains to switch back into the opposite state.[21]

Additionally, many works making the connection between retention and imprint rely on two protocols. One using baking is similar to the PUND technique and requires four identical capacitors. This procedure has been developed by Rodriguez *et al.*[22] The second, performed in Ref. 23, consists of cycling, sending a writing pulse, waiting, sending a reading pulse, and then reinitializing the electrode by cycling once again. The last procedure could be problematic because there is no measurement when the data are modified, and for HfO_2-based materials, it is not certain that the ferroelectric can be reinitialized.

In this work, a different procedure is used. This procedure is described in the following paragraphs. To begin, retention tests are performed on three woken-up samples having different electrical behaviors: a reference sample, one with a very low wake-up effect, and another one with a very high wake-up effect. The reference sample is investigated in order to compare the different retention tests, while the other two samples are studied in order to compare samples with high and low wake-up effects. The origins of the different electrical behaviors are still unknown. It is believed that they arise from the different architectures between the two samples during the annealing step, but further studies should be carried out to precisely identify the root causes of the high and the low wake-up effect. In a second part, the relevance of two different mathematical retention models is assessed.

Samples are stacks of $Pt/TiN/(Hf,Zr)O_2/TiN/Si$ with 100 nm of Pt and 50 nm of TiN. All materials are grown by RF magnetron sputtering at room temperature after cleaning and etching of the silicon native oxide by the buffer oxide etch procedure. Reactive sputtering is used for TiN, and nonreactive sputtering is used for $(Hf,Zr)O_2$ (HZO) and Pt. After rapid thermal annealing, three samples are investigated. They are called NM-50 W, NM-100 W, and M-100 W. "NM" stands for nonmesa and "M" for mesa, which refers to two different architectures already described in Ref. 24. Consequently, NM-50 W and NM-100 W correspond to the same architecture. The only difference between both samples is that "50 W" and "100 W" correspond to the power at which the HZO layer is deposited. M-100 W is etched with an injection of the SF_6/O_2 gas mixture during reactive ion etching. The thicknesses of the HZO layer for the three samples are given in Table I.

The setup for the electrical characterization has already been described in Ref. 25.

During the first step of the experiments, tests mimicking endurance tests are carried out, except that they are stopped before breakdown. These tests are named "wake-up procedures" in the following paragraphs, and they are performed using successively cycling and measurement sequences. The cycling sequences are performed with bipolar square pulses. The measurement sequences allow us to plot the P–E hysteresis loop. They are performed with a five triangular pulse sequence: a setting pulse and positive, up, negative, down pulses. This sequence is termed the "PUND measurement."

After the samples' awakening, retention tests are conducted. They consist of measurement sequences at regular time intervals. Two

TABLE I. Thicknesses of HZO layers for each sample.

Name of the sample	HZO layer thickness
NM-100 W	11.0 nm
NM-50 W	11.3 nm
M-100 W	12.3 nm

different retention tests are conducted. One is performed with the PUND measurement and is called the "retention test without alternations." The second category of test is carried out by alternating the PUND measurement with another measurement sequence using electrical pulses of opposite signs. The sequence is, therefore, called the "NDPU measurement," and the retention test is named the "retention test with alternations."

These last ones are believed to be complementary with the procedures presented in the introduction. Indeed, they allow us to skip the reinitialization step and they imitate several readings, but also rewriting operations when alternations occur.

All measurements are carried out at room temperature on circular pads of 20 μm diameter.

To characterize the imprint, a mathematical parameter is introduced. It is called the "imprint parameter" (IP) and defined as follows:

$$IP = \frac{E_c^+ + E_c^-}{E_c^+ - E_c^-}. \quad (1)$$

IP is dimensionless, and in the extreme cases, it takes the following notable values:

- if the hysteresis is symmetric ($|E_c^+| = |E_c^-|$), $IP = 0$,
- if $E_c^+ = 0$, $IP = -1$,
- if $E_c^- = 0$, $IP = +1$.

Moreover, IP is of the same sign as the hysteresis shift and describes "how much" the hysteresis is shifted.

Furthermore, imprint is often described by the mean coercive voltage[26] or mean coercive electrical field. Another parameter is, therefore, introduced. It is named the "mean switching coercive field" and written as $E_{sw}/2$. E_{sw} is equal to $|E_c^+| + |E_c^-|$ and called the "switching coercive field" by analogy with the switching polarization, whether E_{sw} is or is not linked to switching phenomena.

The first retention test is performed on the NM-50 W sample. After 10^5 cycles, P_{sw} is measured during a retention test without alternations. Only the PUND measurement is used to plot the P–E hysteresis loop, from which P_{sw} is extracted. Results are shown in Fig. 1.

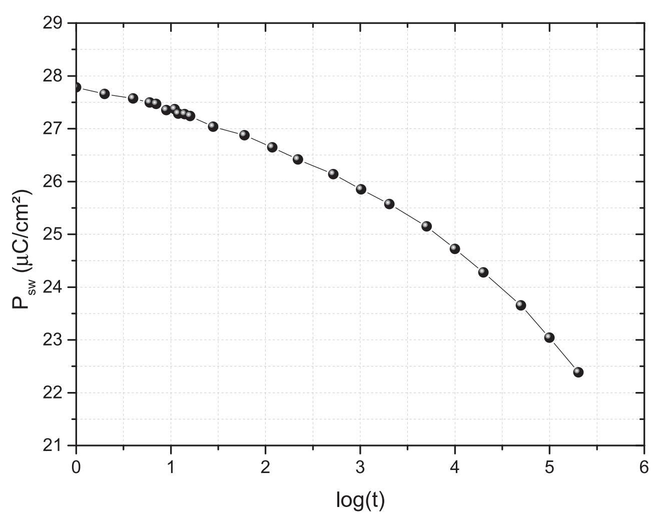

FIG. 1. Retention test without alternations for the NM-50 W samples.

Another experiment is carried out on this sample, constituted by alternations of PUND and NDPU measurements. Sequences are changed after 10^4 s, and the total time of the retention test is 10^5 s.

Results are shown Fig. 2. Contrary to the retention test without alternations, when alternations are added, P_{sw} is not monotonic anymore. This can be explained as follows: when the last programmed pulse is negative (PUND), the P–E hysteresis is shifted on the positive side until $IP \approx 0.4$ after 10^4 s and P_{sw} decreases. After 10^4 s, the measurement sequence is changed. The hysteresis moves to the other side: the negative side, until $IP \approx -0.4$ after another 10^4 s because the last programmed pulse is now positive (NDPU). Consequently, P_{sw} increases when the hysteresis is recentered at the beginning of each PUND or NDPU sequence, except the first one (PUND here). It can, thus, be expected that P_{sw} is maximum when $IP = 0$. After reaching $IP = 0$, IP continues its shift on the negative (positive) side during NDPU (PUND, respectively) measurements and P_{sw} decreases.

During these tests, $E_{sw}/2$ is always increasing during one specific sequence (PUND or NDPU) and $E_{sw}/2$ is reinitialized when the sequence is changed. $E_{sw}/2$ is generally higher for the PUND sequence than for the NDPU because the total width of the hysteresis is higher for PUND than it is for NDPU. If the width of the hysteresis decreases, the P–E loop is more saturated, which implies that during NDPU sequences, the maximum P_{sw} ($IP = 0$) is higher than that during the previous PUND sequence.

IP oscillations could be a real issue for FRAM applications since the operation voltage is assumed to be related to the coercive electrical field. Moreover, this experiment mimics not only reading operations but also rewriting operations in the opposite state for one capacitor. The high E_c shift seems to not be suitable for a commercial system where data can regularly be rewritten.

If the imprint was the sole origin of P_r loss, P_{sw} should reach the same maximum when $IP = 0$ for each equivalent sequence (PUND or NDPU). In Fig. 2, new sequences do not allow the total P_r recovery by recentering the hysteresis loop, meaning that imprint is not the single contribution to P_r loss.

Actually, P_r recovery has already been observed by Higaishi *et al.*,[27] but the authors' experiments were very different. After a wake-up procedure, the authors bake the sample to induce a high imprint. After a certain amount of time, they apply five cycles without changing the pulse signs. Just after the second cycle, they already observe a recentered P–E hysteresis loop.

One could wonder if the high IP oscillations are not linked to a high wake-up effect. As a result, the same experiment is performed on the M-100 W sample, which possesses a reduced wake-up effect (see Ref. 24 for more details). In Fig. 3, IP oscillations lead to the same results when sequences are alternating every 10^5 s instead of 10^4 s for the NM-50 W sample. Although IP oscillations are, therefore, smaller for the M-100 W sample, it is still too high for a commercial FRAM, meaning that reducing the wake-up effect does not prevent the E_c shift.

In the same experiment, it was also observed that P_r loss seemed to be very small compared to the previous sample. So it might possible that reducing the wake-up effect impacts the retention properties. However, the conditions of the experiment were different.

Consequently, the same measurements were performed with a third sample: NM-100 W. It has a comparable P_{sw}, and the same number of cycles was used. NM-100 W has a really pronounced wake-up effect compared to the M-100 W sample (see Ref. 24).

The results are plotted for all samples in Fig. 4. For a short period of elapsed time, P_r loss is minimal for the NM-50 W sample. But after

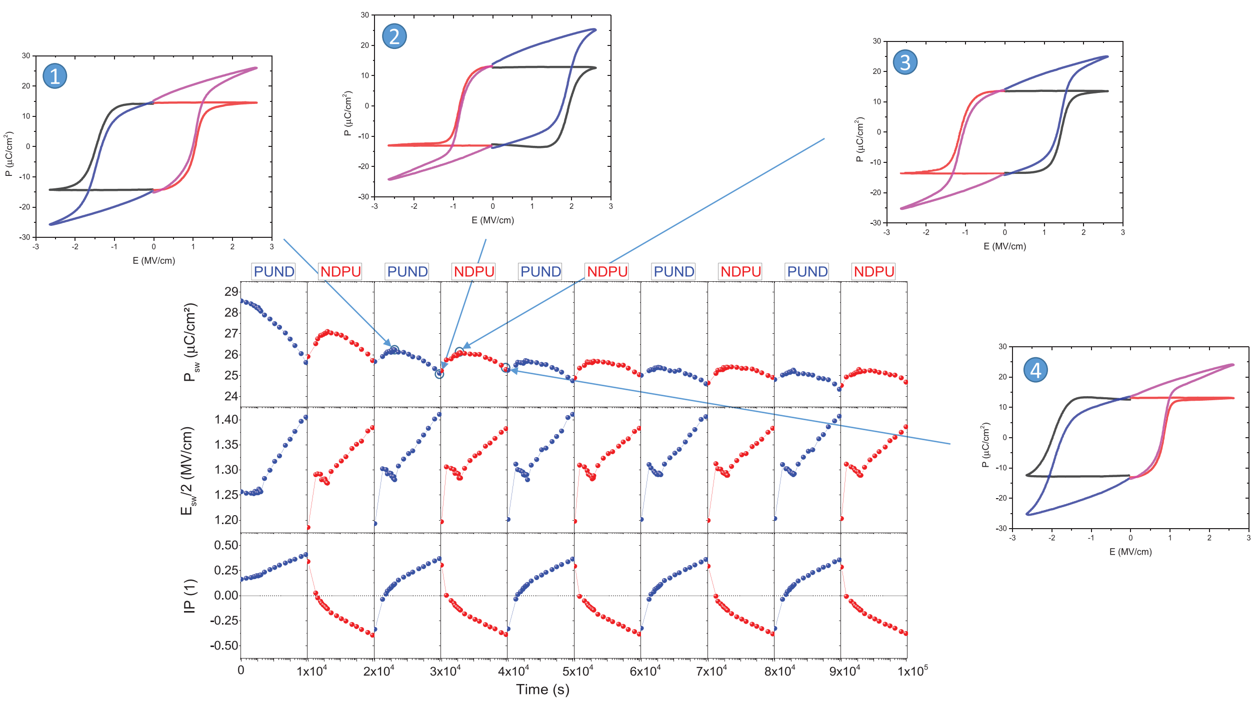

FIG. 2. Retention test for the NM-50 W sample with alternations of PUND and NDPU measurements.

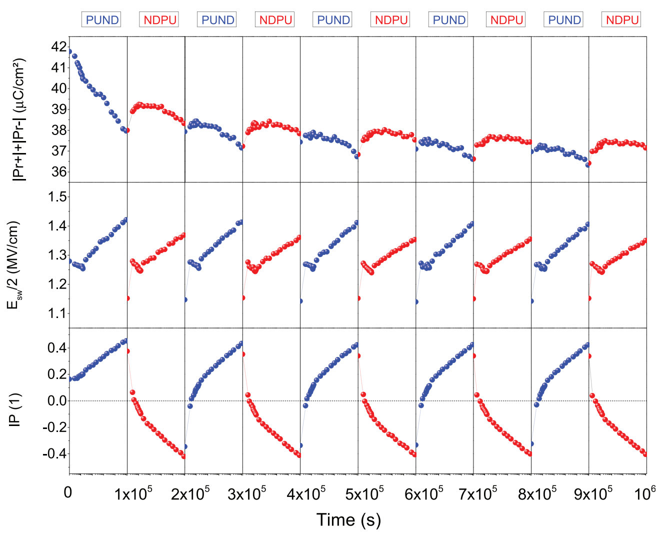

FIG. 3. Retention test with alternations of PUND and NDPU measurements for the M-100 W sample, which possesses a reduced wake-up effect.

$t \approx 20$ s, the slope of P_{sw}/P_0 is rather small for the M-100 W sample compared to the two other samples, confirming that retention loss is exacerbated by a high wake-up effect.

The same E_c shift was also observed for the NM-100 W sample ($-0.4 < IP < 0.4$) during the alternations of PUND and NDPU measurements, confirming that IP oscillates for all samples whatever the electrical behavior during endurance tests is.

Finally, in Fig. 4, in order to compare the curves with or without alternations, retention curves are plotted keeping only the last point of each PUND sequence, except the first sequence where all points are plotted. For the NM-50 W sample, the curve with alternations (in red) has a P_r loss smaller than the one without alternations (in black). As a consequence, alternations cause an improvement of the retention properties. But the difference seems rather small, probably meaning that imprint is not the only cause for P_r loss.

To simplify, in the following, P_{norm} is defined as the normalization of P_{sw} by P_0, the switching polarization at $t = 0$, i.e.,

$$P_{norm} = \frac{P_{sw}(t)}{P_0}. \quad (2)$$

In Fig. 4, it is clear that the retention fitting model is not a simple linear function, contrary to what can be sometimes found in the literature.

Thus, we looked for nonlinear fitting models for ferroelectric HfO_2-based capacitors in the literature. Two fitting equations were found. The first one have been utilized by Lyu *et al.*[16] for HZO/LSMO ($La_{2/3}Sr_{1/3}MnO_3$) thin layers and corresponds to a power law model,

$$P_{norm} = t^{-n}. \quad (3)$$

This equation was borrowed to Refs. 28 and 29 where the authors performed empirical fittings on $SrRuO_3/BaTiO_3/SrRuO_3$ capacitors.

When Eq. (3) is reduced to a linear fitting, the data curve does not fit our data at all (see supplementary material A for more details). Consequently, this model was discarded.

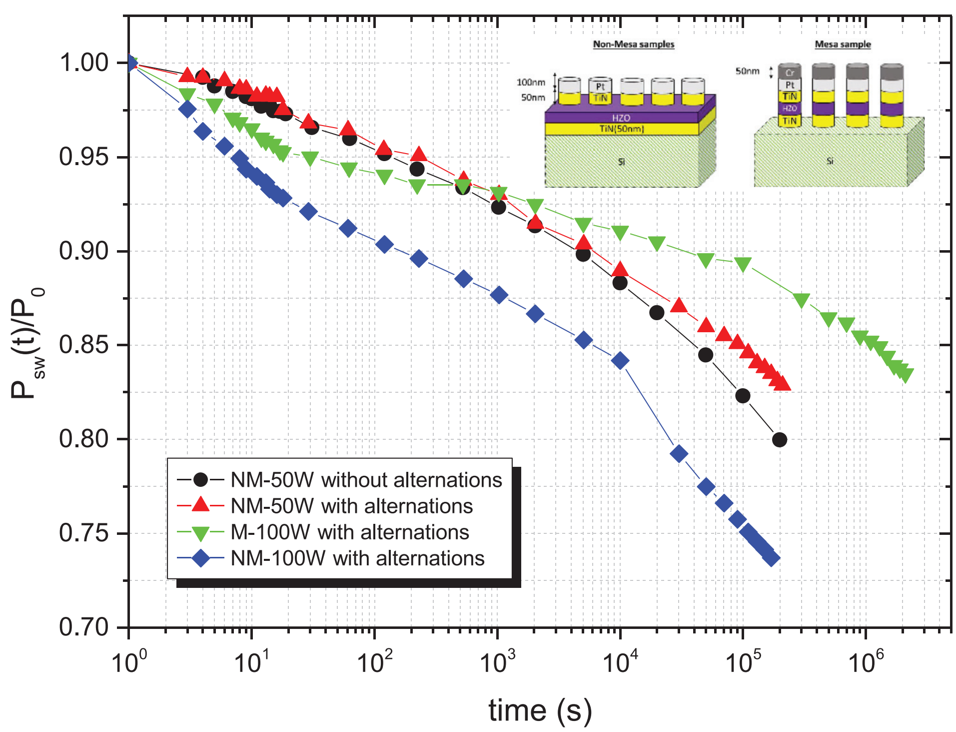

FIG. 4. Retention tests for all samples. For tests with alternations, only the last measure of the PUND sequence has been kept to avoid oscillations and keep only the envelope of the retention curves.

A further model derived from this equation was also investigated. Details are given in supplementary material C.

The second model is a stretched exponential model used by Lee *et al.*[19] to describe ferroelectric Y-doped HfO_2 thin films. The fitting model equation is derived from Refs. 30 and 31 where the authors are investigating the reversal area of domain walls by PFM (Piezo Force Microscopy) and c-AFM in $PbTiO_3$ and $PbZr_{1-x}Ti_xO_3$ thin films, respectively. In the stretched exponential model, $P_{norm}(t)$ is assumed to be equal to $\exp[-(t/\tau)^{\beta}]$. More details can be found in supplementary material B.

The stretched exponential decay $\phi(t) = \exp[-(t/\tau)^{\beta}]$ with $0 < \beta < 1$ is also known as the Kohlrausch–Williams/Watts (KWW) relaxation.[32,33] Here, the β value is not assumed to be in the interval $]0; 1[$ yet, since in Ref. 30, β is found to be greater than 1.

To reduce the problem to a linear fitting, the equation can be rewritten such as

$$\log_{10}\left[\log_{10}\left(\frac{1}{P_{norm}}\right)\right] = \beta \log_{10}(t) + C, \qquad (4)$$

where C is a constant (calculation details can be found in supplementary material B). This equation is not valid for $t = 0$ s since $\log(1/P_{norm}) = 0$. $P_{norm} = 1$ is then excluded from data and fits.

In Fig. 5, retention data are plotted according to Eq. (4). The curve is not linear at the beginning, but around $t = 100$ s, there is an inflexion point at which it becomes linear. Since $t = 100$ s is rather a low value to characterize retention, it could explain why it is generally ignored in the literature.

Thus, the simple stretched exponential model does not entirely describe retention data either. Nevertheless, the curve in Fig. 5 acts as if there were two or more exponentials in the expression of P_{norm}.

For such a mathematical expression, if β_1 is the smallest exponent in the expressions of P_{norm}, as explained in supplementary material B, when $t \to +\infty$, then

$$P_{norm} \underset{t\to+\infty}{\sim} C_1 e^{-(t/\tau)^{\beta_1}}. \qquad (5)$$

Hence, to extrapolate $P_{sw}(10yr)$, Eq. (4) is still valid if only the points after 100 s are taken into account. In fact, a model with two exponantials was already discussed by Lohkämper *et al.*[34] in 1990 for $BaTiO_3$.

In Ref. 26, Schorn *et al.* noticed that the imprint has a linear part at the beginning of retention and then a logarithmic part. It might be possible that imprint influences the rapid decrease in polarization at the beginning; then, as the imprint slows down, the depolarization phenomena take over.

Another possible explanation to the two-part retention characteristic could be linked to the retention behavior reported in Ref. 30 on $PbTiO_3$ thin films where $\beta > 1$. Since $\beta > 1$ corresponds to a very high slope of $\log[\log(1/P_{norm})]$, the coexistence of both processes could correspond to the two-exponential representation.

In Fig. 6, only the points after 100 s have been kept. The y-error at each point is plotted, but it is so small that it can barely be seen. The curve fits very well since the y-error is smaller than the 95% confidence interval.

Traditional US specifications, derived from magnetic memory parameters, are that the retention must be longer than 10 yr.[11,35] Then, the same procedure is used to fit the retention tests of all samples.

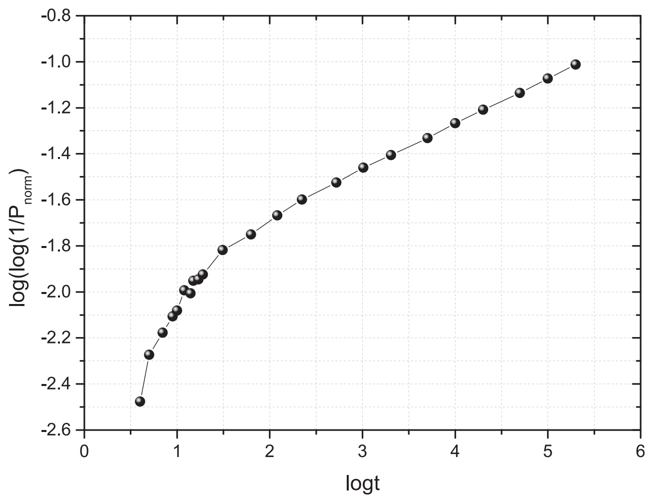

FIG. 5. Retention test plotted according to Eq. (4) for the NM-50 W sample without alternations.

Fitting results are summarized in Table II. For tests with alternations, the same data treatment that was used to plot Fig. 4 is carried out.

The results found previously are in full adequacy with the physical values in Table II. Indeed, the retention loss is very high for all the NM samples, whereas it is very low for the M-100 W sample, corroborating the correlation between the wake-up effect and long-term retention properties. For the NM-50 W sample, retention properties are improved by alternations, which may be a good news for FRAM applications since the data should be rewritten over time in FRAM cells. But despite the fact that imprint is recentering P–E curves during retention tests, the values are still similar, confirming that imprint during alternations has little impact on the long-term retention properties. Therefore, P_r loss might originate from leakage or depolarization field phenomena.

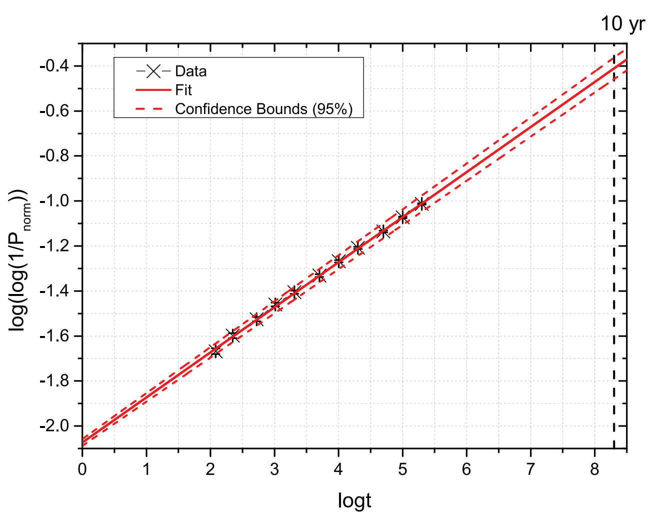

FIG. 6. Retention test plotted according to Eq. (4) for the NM-50 W sample. Data points before 100 s have been excluded in order to keep only the linear part of the curve.

TABLE II. Extrapolated data for the retention tests of all samples according to the stretched exponential model. Only the data points after 100 s have been taken into account.

Sample	Extrapolated data
NM-50 W without alternations	$P_{sw}(10\,yr) = 11.46 \pm 1.13\,\mu C/cm^2$ Retention loss: 59% ± 4% $\beta = 0.20 \pm 0.01\,SI$ $\tau = 3.47 \times 10^8$ s
NM-50 W with alternations	$P_{sw}(10\,yr) = 14.63 \pm 2.52\,\mu C/cm^2$ Retention loss: 49% ± 9 % $\beta = 0.18 \pm 0.01\,SI$ $\tau = 4.14 \times 10^9$ s
M-100 W with alternations	$P_{sw}(10\,yr) = 31.96 \pm 1.69\,\mu C/cm^2$ Retention loss: 25% ± 4% $\beta = 0.11 \pm 0.01\,SI$ $\tau = 8.33 \times 10^{13}$ s
NM-100 W with alternations	$P_{sw}(10\,yr) = 16.68 \pm 4.72\,\mu C/cm^2$ Retention loss: 60% ± 11% $\beta = 0.16 \pm 0.01\,SI$ $\tau = 1.37 \times 10^9$ s

In any case, this model enables a precise phenomenological description of the data. For further discussion about the physical meaning of β and τ, see supplementary material B.

In this article, a correlation between long-term retention properties and the wake-up effect is established: the sample with a reduced wake-up effect has a higher extrapolated P_{sw} value and a smaller retention loss after ten years. As a great deal of research has been conducted to reduce the wake-up effect since its first description in 2013,[36] polarization retention for FRAM applications might not be a problem in the years to come.

However, it was not found how to limit the imprint during retention tests with alternations. The oscillations of the P–E hysteresis loop during these series of tests are believed to be highly detrimental for FRAM memories.

Finally, a two-part retention characteristic has been highlighted for all samples. Since imprint is a highly temperature-dependent process, because it should originate from charge trapping, and industrial FRAM specifications generally require that the device works at $85\,°C$,[22] a more precise investigation including temperature dependence of data retention should be carried out. Moreover, retention is also linked to leakage mechanisms. Since the physical meaning of the fit parameters remains rather elusive here, temperature-dependent measurements and leakage measurements could help to find a better retention model.

See the supplementary material for the power law model fitting, for calculation details of the stretched exponential model, and for another trial of a further fitting model.

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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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