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Study of partially-switched states of ferroelectrics in relation to the spatial inhomogeneity of their domain structure

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Abstract In this paper we reexamine the possibility of using ferroelectric materials for adaptive learning artificial intelligence applications, by exploiting their capability to be set in electrically-controlled multivalue polarization states. Our experiments on a Pb(Zr,Ti)O_3 ceramic sample consisted in conveniently selecting the input pulse electric field and temperature of the sample during field application, in order to create partially-switched states. Employing strong/weak electric field pulses allows to control the analog polarization levels by a linear or logarithmic addition of pulses. The partially-switched states are mapped with enhanced resolution when domains with dissimilar evolution stages are present. Applying electric fields while heating the sample allows to reduce the switching time and shifts down the switching threshold. Thus, in addition to artificial intelligence applications, these results provide hints for energy-saving devices that exploit the intrinsic high mobility of small fluctuating domains.

Keywords Ferroelectrics · Switching · Domains

1 Introduction

With the advances of nonvolatile memory applications of ferroelectric thin films, clarifying the switching mechanisms and simulating the electric response of nonvolatile memory cells has become imperative. Historically, a few seminal papers on this topic established a nucleation-growth mechanism of polarization reversal [1–3] and this picture is being validated and completed by contemporary data as well [4–5]. Although the conventional proof of ferroelectricity has nearly always been the so-called major hysteresis loop, corresponding to complete switching processes between the two remanent polarization states, for practical reasons one needs to also examine the intermediate switching stages. In case of integrated nonvolatile memories, due to the device architecture and circuitry needed to ensure the reliability of memory cells' operation, the polarization may not necessarily be completely switched. In addition, for artificial intelligence applications where appropriate control of the learning curve is desired, the partially-switched stages are particularly relevant. Therefore, study of not only the binary ferroelectric states and electrically-induced transitions between them is required, but the analog intermediate levels must be characterized as well. Several studies in this direction have been done by the group of Ishiwara and Tokumitsu, who proposed adaptive learning devices based on Pb(Zr,Ti)O_3 (PZT) thin films [6]. Polarization reversal dynamics and the associated domain structure change during switching are routinely studied experimentally by the Piezoresponse Force Microscopy method [7].

In this work we reexamine the possibility of using ferroelectric materials for adaptive learning neurodevice applications, by exploiting their capability to be set in electrically-controlled multivalue polarization states. Fundamentally, ferroelectrics have two remanent polarization states associated to the stable minima of the total energy, obtained for certain correlated ionic displacements in their unit cell. These states allow the use of ferroelectrics as binary nonvolatile memory cells. However, in principle analog polarization levels are also possible if the ferroelectric domain structures associated to partially-switched

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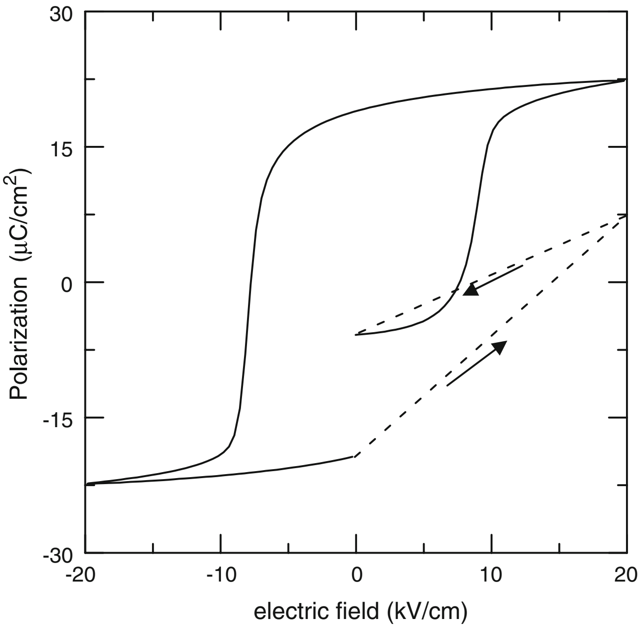

Fig. 1 Illustration of experimental procedure used for creating and measuring the partially-switched states. The *dashed lines* are not measured data, but eye-guidelines of possible polarization evolution during and after the application of a pulse electric field to a PZT ceramic capacitor, before measuring the hysteresis loop visible in the figure

stages of polarization reversal can be stabilized for a sufficient time in presence of dopants, inhomogeneities or other defects that inherently exist in real materials. In order to emulate a learning process, creating as many intermediate switching states as possible is desired. In what follows we will study in detail these partially-switched levels of the polarization reversal process and will aim to elucidate the conditions necessary for creating a large number of them in view of adaptive learning device applications.

2 Results and discussion

The experiments have been done using ceramic capacitors containing a commercial PZT plate of 50 μm thickness (P-7B of Murata Manufacturing Co. Ltd. Japan). A few material properties of this sample are listed in [8]. The experimental procedure is summarized in Fig. 1. First the sample is pre-poled into the negative remanent polarization state and then a rectangular pulse (or a sequence of rectangular pulses) with variable height and duration is applied. This pulse will induce a complete or partial polarization reversal, depending upon the above-mentioned parameters. The switching process itself is not directly measured in our experiments. Rather the memorized polarization state *after* switching is detected from a triggered single hysteresis loop measurement, using a triangular 100 Hz waveform with amplitude high enough to completely reverse the polarization of the sample. It is important to stress out that the measured hysteresis loop should be the very first after the switching pulse, because the subsequent ones will no longer detect the memory of the previous state (as the measuring triangular signal will have already erased it).

Using the above procedure we have investigated the partial switching processes as defined by Tokumitsu et al. as Pulse Width Modulation (PWM) and Pulse Frequency Modulation (PFM) [6]. Figure 2 reveals that upon varying the duration of the single pulse used for switching (PWM), the polarization can be reversed either by a strong 20 kV/

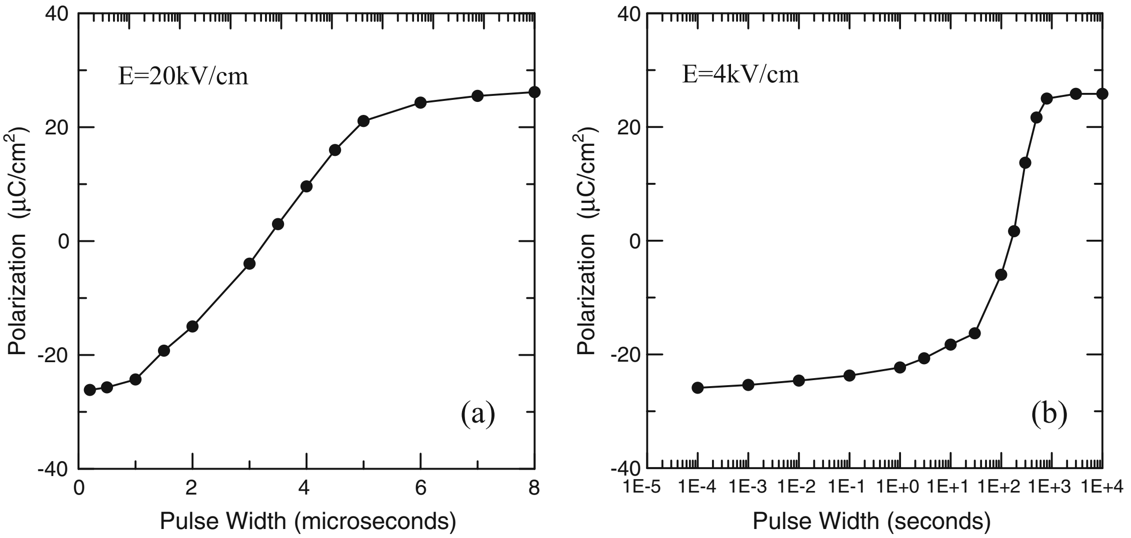

Fig. 2 Polarization evolution during its reversal induced by a single pulse electric field (PWM sequence) of (**a**): 20 kV/cm and (**b**): 4 kV/cm and width indicated on the abscissa

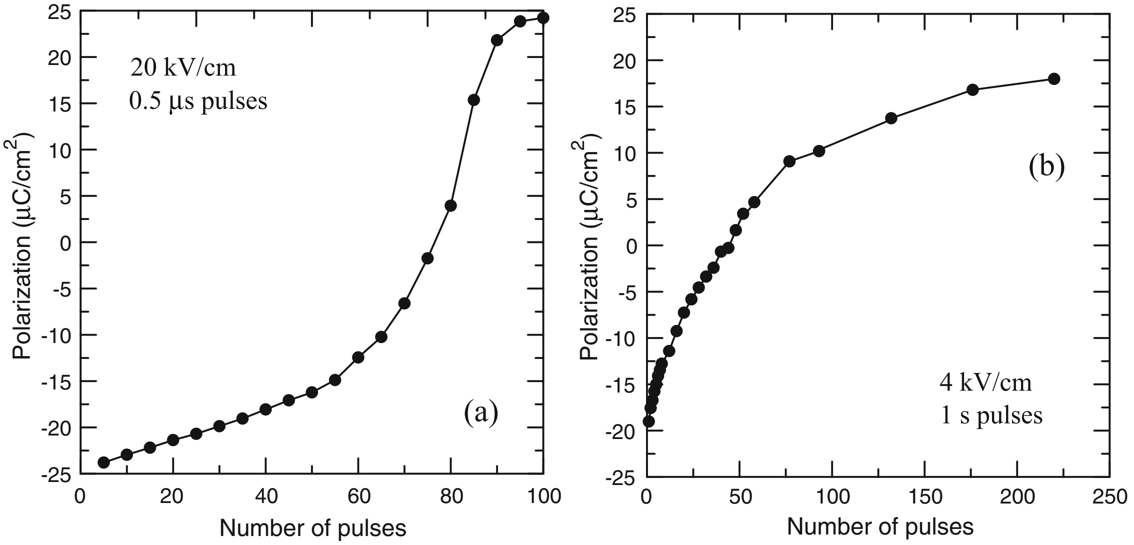

Fig. 3 Polarization evolution during its reversal induced by a PFM pulse electric field sequence of (**a**): 20 kV/cm, 0.5 µs pulses and (**b**): 4 kV/cm, 1 s pulses

cm electric field or by a weak 4 kV/cm electric field. Polarization retention preliminary tests revealed that the partially-switched levels are reasonably stable for durations comparable to experimental times of this study (actual data will be shown elsewhere). What is very different in the switching behavior is the total time needed to achieve complete polarization reversal: while in the strong field case it is in the microsecond range, it extends to tens of minutes in the weak field case. In fact, in the later case the switching process extends over several time decades, a behavior that has also been found (albeit on a larger extent) in ferroelectric thin films [9]. The origin of this phenomenon was proposed to be the widely-distributed time to nucleation in independent areas of the thin film sample [9]. Our sample is a bulk ceramic and so one should not expect it to behave completely similar with thin films. Indeed, while the switching time tends to increase to minutes or even hours range, complete polarization reversal can be achieved even for very weak fields with smaller intensity than the coercive field apparent on the ferroelectric hysteresis loop in Fig. 1. As this does not seem to comply with the assumption of domain evolution in independent nucleation-deprived regions, in what follows we will invoke other qualitative arguments related to the peculiarities of domain switching.

Next we have measured the switching behavior by the PFM procedure that involves application of many short length pulses. As with PWM, Fig. 3 reveals that the difference between the strong field and weak field measurements is still related to the switching time: in the former (later) case individual pulses with 'microsecond' ('second') duration are needed in order to complete the switching process, respectively. We also remark that the resolution of the partial switching states, defined as the switched polarization per pulse, is larger (smaller) in the initial stages for the strong (weak) field case, respectively, and the opposite is true for the late stages. If one uses 'microsecond' duration single pulses of electric fields with smaller intensity, Fig. 4 shows that the number of pulses required to completely switch the polarization dramatically increases up to 10^{10} and the partial switching states resolution increases as well. Thus, in artificial intelligence applications

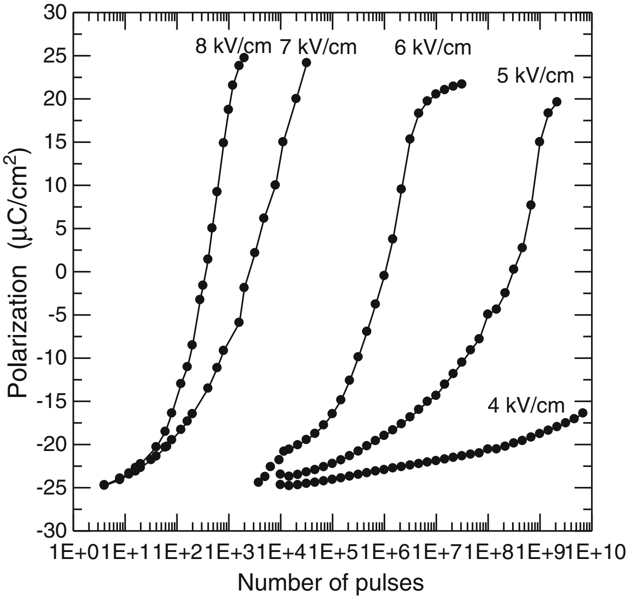

Fig. 4 Polarization evolution during its reversal induced by a PFM pulse electric field sequence with various field intensities

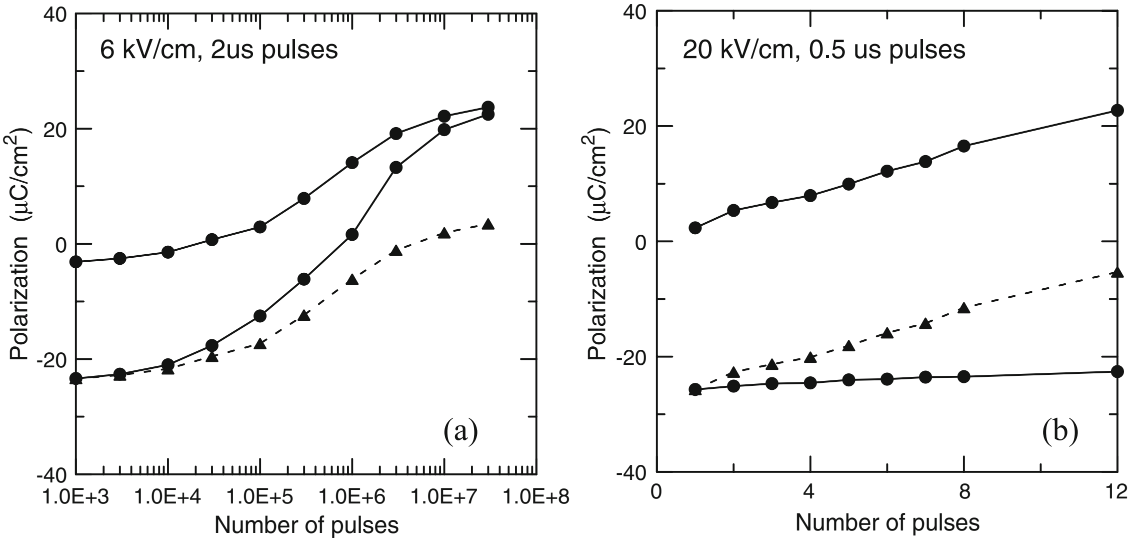

Fig. 5 Polarization evolution during its reversal induced by a PFM pulse electric field sequence of (**a**): 6 kV/cm, 2 μs pulses and (**b**): 20 kV/cm, 0.5 μs pulses, from the $P=-P$r state (*bottom*) and $P\sim0$ state (*top*), the latter induced by a (**a**): strong and (**b**): weak electric field. The *dashed line* is an eye-guideline, obtained by shifting the $P\times0$ data to start from $P=-P$r, enabling to assess the different slopes of the corresponding switching curves

where the learning curve should pass through a large number of intermediate stages, weak pulses with microsecond duration are suitable.

In order to explain these experimental findings, we will refer to the results of model simulations of the polarization reversal process, done under the framework of a Landau theory based lattice model, and discussed in some of our previous publications [10–12]. The calculations have revealed that switching induced by a strong electric field involves an evolution starting from many small reversed domains, while in case of a weak field the polarization reversal is triggered from a few domains that tend to become very large in size. The large domains have a large inertia and consequently require a very long time to be able to extend over the entire sample volume (according to the model, ferroelectric regions lacking initial nuclei with favorable orientation of polarization will remain unreversed until the internal electric fields associated to coupling to neighbors become significantly large). When one applies strong external electric fields, it becomes possible for reversed domains to appear in many regions (now the role of the coupling between neighbors is not as critical as in the weak field case) and so the PWM switching is faster or a smaller number of PFM pulses are required to achieve complete polarization reversal. Therefore, whenever many small domains are likely to be formed, one can expect the polarization reversal speed to be increased.

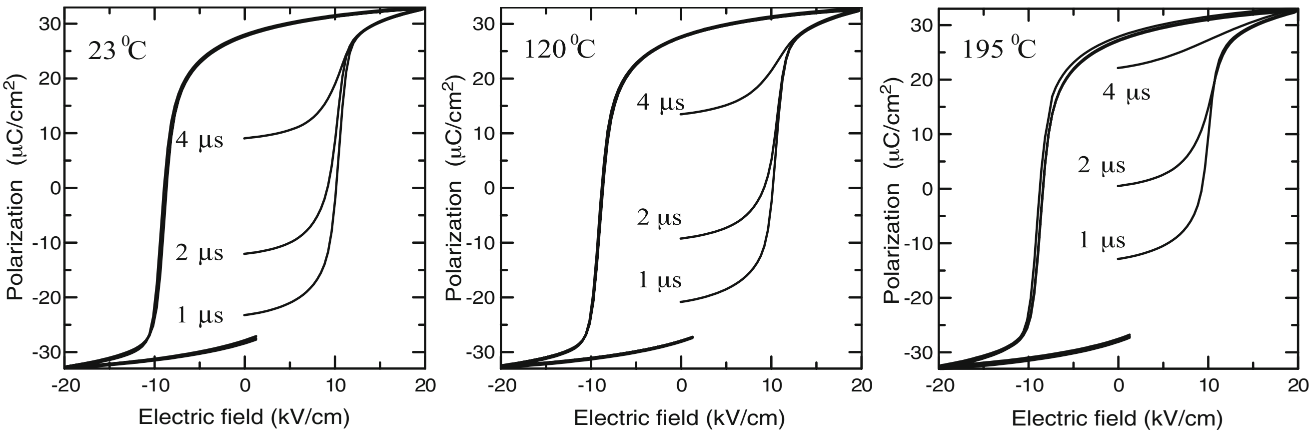

Fig. 6 Hysteresis loops starting from partially-switched states obtained by application of single pulses of 20 kV/cm amplitude and indicated widths, in an experiment at room temperature (*left*), after heating to 120 °C and cooling (*middle*) and after heating to 195 °C and cooling (*right*)

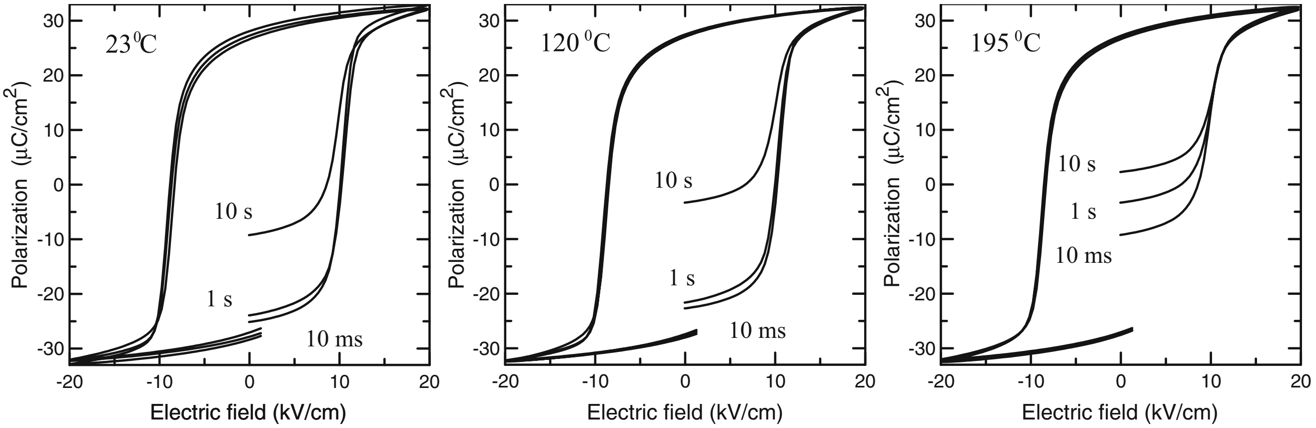

Fig. 7 Hysteresis loops starting from partially-switched states obtained by application of single pulses of 4 kV/cm amplitude and indicated widths, in an experiment at room temperature (*left*), after heating to 120 °C and cooling (*middle*) and after heating to 195 °C and cooling (*right*)

In order to confirm these mechanisms, we have performed partial switching experiments starting from a "designed" domain state with neutral polarization. Specifically, we have designed a "$P=0$ state" in three ways: (1) by a strong electric field, (2) by a weak electric field and (3) by heating the sample towards the Curie point (T_C). As a matter of fact, by this procedure we induce artificial nucleation sites in the sample. Upon setting the sample in the neutral polarization state according to above (1)–(3), we have studied the efficiency of creating subsequent partially-switched levels and compared it to the case of using $P=-Pr$ as starting point. After establishing initial partially-switched states with small (large) domains by means of a strong (weak) electric field that reverses the polarization up to the $P\sim 0$ state, the switching is continued by applying a weak (strong) field, respectively. In this way we additionally create large (small) domains that would otherwise never be formed when switching exclusively with strong (weak) fields, respectively. Rather contrary to expectations, Fig. 5(a) shows that in the (1) case, in weak fields, the small domains (acting as artificial nucleation sites) previously created by the application of a strong field do not accelerate the polarization reversal, compared to the case of switching based on large domains starting from the $-Pr$ state. Indeed, pulses increasing in number on a logarithmic scale are still needed to create subsequent partial switching levels. However, the partially-switched states resolution increases compared to that in the $-Pr$ state, due to the presence of small domains that are at an earlier evolution stage than the large ones.

Conversely, Fig. 5(b) shows that, in strong fields, the large domains that were previously formed under a weak

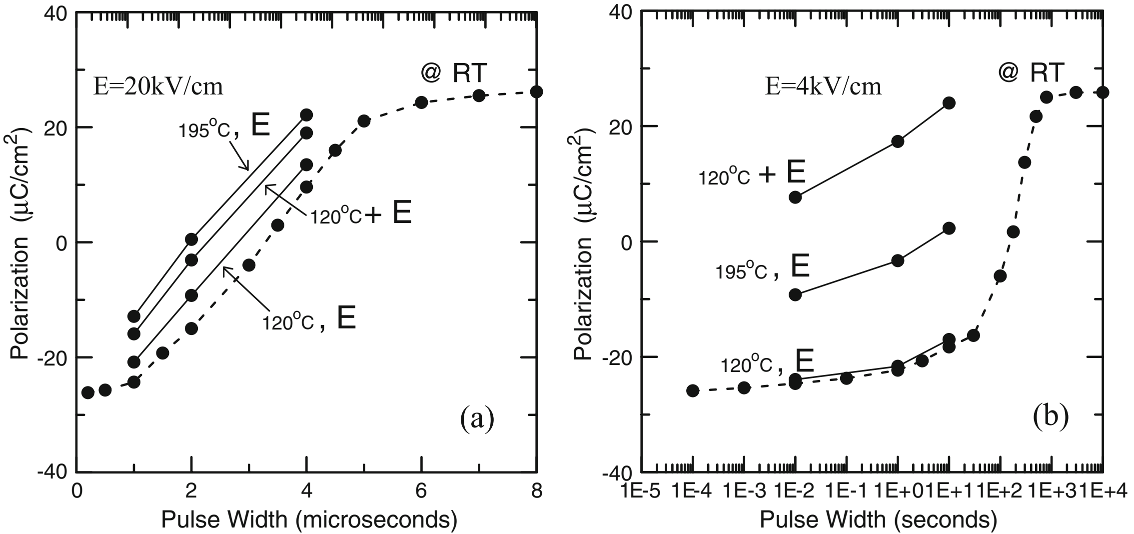

Fig. 8 Polarization dependence on the pulse width in a PWM experiment at room temperature and specified temperatures, either applying the electric field after previous sample heating and cooling or simultaneously with heating the sample. The electric field intensity is (**a**): 20 kV/cm and (**b**): 4 kV/cm

field [(2) variant] are effective for accelerating the polarization reversal when starting from the $P=0$ state, compared to $-Pr$ state. Also, an enhanced resolution of partially-switched states starting from the $P=-Pr$ state is obtained due to the absence of large domains. From these results we can conclude that pulse-induced ferroelectric switching is more efficient when a large density of domains at the same evolution stage exists (switching appears to become ballistic in this case) and partially-switched states are mapped with enhanced resolution when either small domains or domains with dissimilar evolution stages are present.

In the (3) case, instead of setting the sample into the $P\sim0$ state by electric field, we have generated randomly-distributed small domains by heating. A first series of experiments involved pre-poling the sample as before, followed by inserting the sample into an oven set to the chosen temperature and finally applying the switching pulses after cooling. We have made these experiments by heating the sample up to 120 and 195 °C, respectively. Figures 6 and 7 reveal that whenever small domains are thermally induced, switching with both strong and weak fields of a given duration (PWM pulse sequence) is completed to a larger extent, especially for temperatures approaching the Curie temperature (for our PZT ceramic sample $T_C\sim200$ °C). Another series of experiments has been done by applying the reversing electric pulse simultaneously with the sample heating up to 120 °C. The measurements are summarized in Fig. 8. We observe that heating simultaneously with the electric field application is most efficient in inducing fast polarization reversal and allows the switching threshold to be shifted to lower values of electric field intensity. In fact, in case of weak electric fields, this procedure leads to more complete switching than in case of heating near the Curie temperature and the switching time decreases from tens of minutes down to 10 s (see Fig. 8(b)). The influence of thermally-induced domains is not so pronounced in case of strong fields (see Fig. 8(a)), as switching in that case is already based on evolution of many small domains.

3 Conclusions

The performed experiments have shown that interesting properties of ferroelectrics, that are relevant for applications, relate not only to their binary memory states, but also to the intermediate levels corresponding to partially-switched states. Analog polarization levels can be created by conveniently selecting the operation conditions, either electrically by pulses with variable amplitude and/or duration or thermally by heating towards the Curie point. Employing strong/weak electric field pulses allows to control the analog polarization levels by a linear or logarithmic addition of pulses. The mapping of partially-switched states can be achieved with enhanced resolution (1) with narrow/strong field pulses in initial stages and wide/weak field pulses in late stages, on a linear scale and (2) with narrow/weak field pulses, on a logarithmic scale. The partially-switched states are mapped with enhanced resolution when domains with dissimilar evolution stages are present, in agreement to previous model calculations that revealed the role of domain dynamics for establishing the characteristics of partially-switched states that can be created. Simultaneous application of electric fields and heating the sample allows to reduce the switching time and shifts down the switching threshold, due to the creation of randomly-distributed small domains that behave similarly with those created by stronger electric fields. In addition to their relevance for artificial intelligence applications, these findings offer insights for energy-saving applications that exploit the intrinsic high mobility of such small fluctuating domains.

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