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Retention of intermediate polarization states in ferroelectric materials enabling memories for multi-bit data storage

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A homogeneous ferroelectric single crystal exhibits only two remanent polarization states that are stable over time, whereas intermediate, or unsaturated, polarization states are thermodynamically instable. Commonly used ferroelectric materials however, are inhomogeneous polycrystalline thin films or ceramics. To investigate the stability of intermediate polarization states, formed upon incomplete, or partial, switching, we have systematically studied their retention in capacitors comprising two classic ferroelectric materials, viz. random copolymer of vinylidene fluoride with trifluoroethylene, P(VDF-TrFE), and $Pb(Zr,Ti)O_3$. Each experiment started from a discharged and electrically depolarized ferroelectric capacitor. Voltage pulses were applied to set the given polarization states. The retention was measured as a function of time at various temperatures. The intermediate polarization states are stable over time, up to the Curie temperature. We argue that the remarkable stability originates from the coexistence of effectively independent domains, with different values of polarization and coercive field. A domain growth model is derived quantitatively describing deterministic switching between the intermediate polarization states. We show that by using well-defined voltage pulses, the polarization can be set to any arbitrary value, allowing arithmetic programming. The feasibility of arithmetic programming along with the inherent stability of intermediate polarization states makes ferroelectric materials ideal candidates for multibit data storage. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4953199]

A homogeneous ferroelectric single crystal exhibits a spontaneous polarization that can repeatedly be switched by an electric field. The polarization reversal is described by the Landau-Devonshire mean-field theory,[1,2] which treats a ferroelectric single crystal as a homogeneous domain with the polarization, P, as the order parameter. The free energy as a function of polarization has two minima. These two states exhibit a remanent polarization, $\pm P_r$, at zero electric field. Other states with intermediate values of polarization are thermodynamically instable. Hence, in a defect-free single crystal, polarization reversal at the coercive field, E_c, originates from a coherent, collective rotation of dipoles, generally referred to as intrinsic switching.[3] Such intrinsic switching has been experimentally reported in ultra-thin films of $BaTiO_3$[4] and of the random copolymer of vinylidene fluoride with trifluoroethylene, P(VDF-TrFE).[5,6]

Commonly used ferroelectric materials such as $BaTiO_3$, $Pb(Zr_xTi_{1-x})O_3$ (PZT), and P(VDF-TrFE) are, however, not homogeneous single crystals but inhomogeneous, multi-domain, polycrystalline thin films or ceramics that are by nature not defect-free.[7–9] There is energetic and spatial disorder due to, for instance, misfit dislocations,[10,11] defects in conformation and molecular packing of macromolecular chains,[12] and grain boundaries.[13,14] The presence of these defects lowers the barrier for polarization reversal. The collective rotation of dipoles is replaced by nucleation and anisotropic growth of individual domains, termed as extrinsic switching.[15–17]

In piezoelectric transducers and actuators, the ferroelectric material is poled at high electric field and operated below the coercive field. In ferroelectric memories for data storage, binary information is stored as remanent polarization. In all these applications, complete polarization is obtained by poling far above the coercive field. The retention of the two remanent polarization states has been investigated as function of temperature, residual internal electric field, film thickness, and type of electrode materials.[18–20] Optimized devices operated well below the Curie temperature, T_c, show an almost infinite data retention for both saturated polarization states.[18]

Application of an electric field close to the coercive field leads to incomplete switching and, hence, to intermediate polarization states. It is not *a priori* known whether these intermediate polarization states are thermodynamically stable. Experimental data on retention of intermediate polarization states are scarce. The Landau-Devonshire theory predicts that for a single domain, these states are not stable. However, this mean-field treatment for single crystals may be invalid for multi-domain ferroelectric materials, in which the free energy is no longer a single-valued function of the homogeneous polarization, but strongly depends on the configuration of domains.

Here, we systematically investigate the stability of intermediate polarization states of two classic ferroelectric materials, viz. P(VDF-TrFE) and PZT, by monitoring their retention characteristics. To exclude the influence of a depolarization field, we characterize the stability in a ferroelectric

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capacitor. We note that the formation of periodic domain stripes according to Kittel-Mitsui-Furuichi law,[21,22] as has been experimentally observed in free-standing ferroelectric materials,[23] can then be disregarded. Intermediate polarization states are obtained by applying electric fields close to the coercive field. The retention was measured as a function of time and temperature. We find that the intermediate polarization states are surprisingly stable over time, even up to the Curie temperature. We argue that this remarkable stability originates from the coexistence of local minima in the energetic landscape of multi-domain ferroelectric materials. A domain growth model is derived that quantitatively describes deterministic switching between intermediate polarization states. We show that by using a well-defined voltage pulse, the capacitor can be partially switched to a state with a polarization of any chosen value, allowing arithmetic programming. The consequences for the development of multi-bit memories for data storage are discussed.

The capacitors based on the random copolymer P(VDF-TrFE) (65%/35%) were prepared as previously described.[17] The P(VDF-TrFE) films with a thickness of about 500 nm were spin-coated from a 5 wt. % solution in methylethylketone on thermally oxidized silicon monitor wafers, on which 50 nm thick Au bottom electrodes on a 2 nm Ti adhesion layer were photo-lithographically defined. To enhance the crystallinity and hence the ferroelectric properties, the samples were subsequently annealed in vacuum at 140 °C for 2 h. As a last step, a Au top electrode was evaporated through a shadow mask. The device area was 0.16 mm^2. Soft PZT ceramics were obtained from Morgan Advanced Ceramics (PZT 507, $T_c = 438$ K, $d_{33} = 700$ pm/V). The ferroelectric hysteresis loops and data retention were measured with a Radiant Precision Multiferroic Test System (Radiant Technologies, Inc.).

Each experiment started from a discharged and depolarized ferroelectric capacitor, to ensure that the electrodes as well as the ferroelectric medium are electrically neutral. The experimental procedure to arrive at this fully depolarized state is schematically presented in Fig. 1(a). Any history due to a preceding measurement is erased by applying an alternating electric field whose amplitude gradually decreases from far above the coercive field to zero. Simultaneously, the electrical displacement is measured. An example is given in Fig. 1(d), where a capacitor polarized at $-P_r$ is fully depolarized. The electrical displacement upon applying the alternating electric field is presented by the fully drawn blue lines. The polarization starts at $-P_r$ and ends at about zero. We note that this procedure is similar to demagnetizing a ferromagnetic material.[24]

To set a certain polarization state, a monopolar triangular voltage pulse is applied, as depicted in Fig. 1(b). The corresponding electrical displacement is presented as the red line in Fig. 1(e). Polarization increases with the applied voltage and keeps increasing while scanning back. This delay depends on the width of the writing pulse and the time constant of polarization reversal. In this specific example of a P(VDF-TrFE) capacitor, the polarization is set to 5 μC/cm^2. Subsequently, the electrodes are shorted. After a given time, the polarization is read out by two bipolar triangular pulses, cf. Fig. 1(c). The first triangular pulse reads the retained

polarization, while the second results in a full loop that is recorded as a reference. The corresponding electrical displacement loops are depicted by the black lines in Fig. 1(f).

By changing the amplitude of the writing pulse, the polarization can be set to any intermediate value, as shown by the red lines in Figs. 1(g)–1(l). The applied electric field must be close to the coercive field in order to obtain a nonnegligible polarization. The retained polarization is subsequently read out either immediately after the writing pulse (Figs. 1(g)–1(i)) or after 1000 s (Figs. 1(j)–1(l)), as indicated by the black lines. The measurements show hardly any loss in polarization as the value, within the experimental uncertainty, does not change with time of shortening, regardless of whether the set polarization is saturated or incomplete. This excellent retention is clearly presented in Fig. 2(a), where the polarization is presented as a function of time. Each data point is a separate measurement starting from a fully depolarized capacitor. Fig. 2(a) shows that up to 10^4 s there is hardly any change in polarization. Similar measurements were performed for the PZT capacitors. The extracted polarization as a function of time is presented in Fig. 2(b). Also for PZT, the polarization is retained. The intermediate polarization states of both P(VDF-TrFE) and PZT do not depolarize but are stable over time.

The retention measurements were performed as a function of temperature for P(VDF-TrFE) capacitors (Fig. 2(c)). The retained polarization after 1000 s is presented as a function of temperature. The intermediate polarization states are remarkably stable up to the Curie temperature, T_c, of about 370 K. At higher temperatures, both intermediate and saturated polarization states immediately depolarize.

A ferroelectric material typically consists of an ensemble of domains, as has been confirmed by Piezoresponse Force Microscopy (PFM) measurements on both PZT and P(VDF-TrFE) thin-film capacitors.[25,26] The switching of each individual domain is extrinsic. When an electric field is applied, polarization reversal starts from nucleation sites, such as defects and grain boundaries. Subsequently, the domains grow driven mainly by electrostatic interactions. As the electrostatic energy is much larger than the thermal energy, k_BT, the intermediate states are not in thermodynamic equilibrium. When interactions between domains can be disregarded, domains are independent and, as a consequence, each individual domain is in itself bistable. The threshold for domain switching is the coercive field that differs from domain to domain, for instance, due to a different nucleation rate or different domain-wall creep motion.[16,17] Consequently, in a multi-domain material, there is a distribution of coercive fields, as is reflected in the polarization hysteresis loop, which is not perfectly square. We note that the distribution of the coercive field depends on frequency and can be determined as the derivative of the electrical displacement as a function of electric field. When the capacitor is switched with an electric field in the vicinity of the mean value of the coercive field, domains with different polarization coexist, yielding intermediate polarization states. Experimentally, intermediate polarization states are stable, which suggests that bistability of individual domains is maintained in a macroscopic ferroelectric material.

We note that the inherent stability of intermediate polarization states makes ferroelectric materials ideal candidates
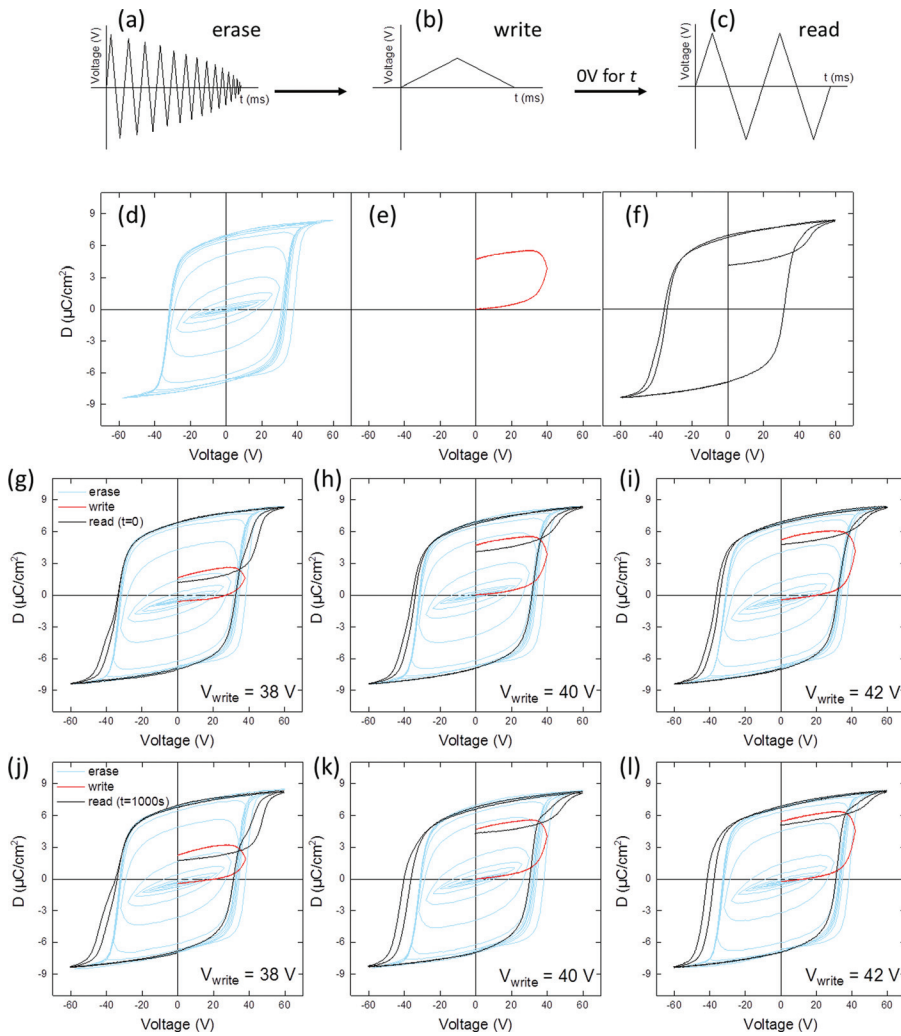
FIG. 1. Erasing, writing, and reading of intermediate polarization states. (a) Pulse train used to depolarize a ferroelectric capacitor. (b) Writing pulse to set an intermediate polarization state. (c) Readout pulse sequence. The first triangular pulse reads the retained polarization, while the second reads the full loop as a reference. Writing and reading pulses correspond to a frequency of 100 Hz. (d) Electrical depolarization of a P(VDF-TrFE) capacitor starting at a polarization of $-P_r$ and ending at zero. (e,f) Writing and reading out an intermediate polarization state. (g,h,i) Retained polarization immediately read out after the writing pulse. The labels show the amplitude of the triangular writing pulse. (j,k,l) Retained polarization read out 1000 s after the writing pulse. The amplitude of the triangular writing pulse is indicated.

for multibit data storage. Recently, eight polarization states corresponding to a 3-bit memory have been reported using capacitors based on PZT and BiFeO$_3$ ferroelectric thin films. Good retention and state reproducibility was demonstrated.[27] Multilevel information storage has been reported in patterned P(VDF-TrFE) capacitors,[28] as regions with different thicknesses throughout the ferroelectric layer have a different polarization upon biasing due to different coercive voltages. A 3-bit memory has been reported in a single P(VDF-TrFE) capacitor using a pulse sequence leading to partially polarized states.[29] The operation has been explained with the dipole switching theory.[29–31] Various multibit memories have also been reported for ferroelectric field-effect transistors (Fe-RAM) by controlling the local polarization. Stable multibit operation of a single-grain PZT 1T-FeRAM has been reported.[32] A 60 nm channel length PZT FeRAM showed multilevel programming, fast switching, and data retention up to 10^5 s.[33] Multibit flexible FeRAMs have been reported using gate-controlled polarization of the ferroelectric P(VDF-TrFE) gate.[34,35] A multibit dual gate transistor was realized by selectively programming the ferroelectric top gate.[36] Programming cycle endurance was up to 100 cycles[34] and data retention exceeded 10^3 s[36] and 10^5 s.[34] Apart from capacitors and field-effect transistors, multibit memories have been fabricated using modulation of the Schottky injection barrier by the ferroelectric polarization.[37,38] All these reports demonstrate the

stability of intermediate polarization states and pave the way for multibit memories. We note that this only holds for ferroelectric devices showing extrinsic switching. When switching is intrinsic,[4–6] then by definition the intermediate polarization states cannot be in thermodynamic equilibrium.

A key requirement for multibit data storage is a protocol to accurately and reproducibly set the intermediate polarization states. This deterministic switching can, for instance, be realized by setting the gate bias in a FeRAM, and by controlling the displacement current in a ferroelectric capacitor.[27] In the following, we perform deterministic switching by controlling the voltage instead of the current, which in a ferroelectric capacitor is experimentally much easier. We derive a protocol to set the intermediate polarization states and thus allow arithmetic programming.

As the intermediate polarization states are stable, arithmetic programming can be performed using well-defined voltage pulses. As an illustration, we first take a capacitor of P(VDF-TrFE) and set the initial polarization at a value of $P_r/2$, 0, $-P_r/2$, and $-P_r$. Then, we apply a square, stepwise voltage pulse. The measured polarization as a function of pulse width and pulse amplitude is presented in Fig. 3. The time dependent polarization from the initial states of $P_r/2$, 0, $-P_r/2$, and $-P_r$, indicated by the blue, green, red, and black symbols, respectively, is presented for various pulse amplitudes. For a pulse amplitude of 20 V, the polarization does
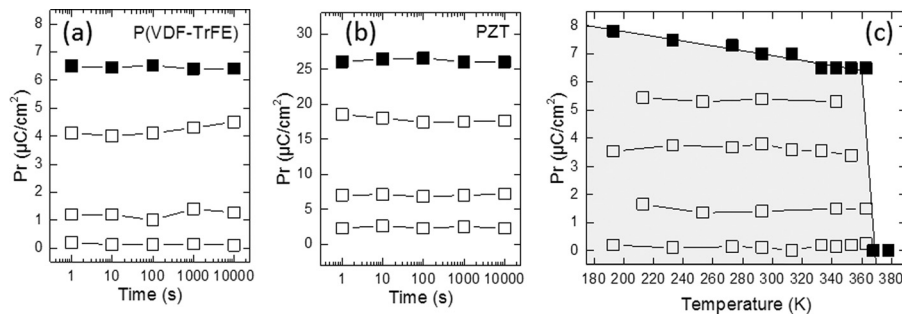
FIG. 2. Retention of intermediate polarization states. (a,b) Polarization as a function of time of P(VDF-TrFE) and PZT capacitors, respectively. Solid symbols represent the saturated polarization and open symbols represent intermediate, unsaturated polarization states. (c) Retained polarization after 1000 s as a function of temperature, as measured on P(VDF-TrFE) capacitors. Solid symbols represent the saturated polarization, and open symbols represent intermediate, unsaturated polarization states. The shaded area marks the range of temperature and polarization within which states are stable.

not change for any pulse width within the measurement window, here 3 s. In order to change the polarization, the pulse amplitude should be close to, or higher than, the coercive voltage, here 25 V. The higher the pulse amplitude is, the faster the polarization changes irrespective of the initial value of the polarization.

To quantify the measured change in polarization, we generalize the Kolmogorov-Avrami-Ishibashi (KAI) model, originally derived to describe complete polarization reversal[39] to

$$P(t) = P(0) + [P_r - P(0)] \times [1 - e^{-(t/t_0)^n}], \quad (1)$$

where $P(0)$ is the initial polarization, t_0 is the characteristic switching time, and n is the so-called Avrami index. The KAI model is based on the classical statistical theory of nucleation and unrestricted domain growth, and has been successfully applied to P(VDF-TrFE) thin films.[9,17] The phenomenological switching parameters t_0 and n depend on electric field and temperature and have been extracted previously from transient measurements.[17] By substituting the reported parameters in Eq. (1), the polarization as a function of time for each pulse height has been calculated and is presented as the solid lines in Fig. 3. A good agreement with measured polarization values is obtained.

With the deterministic knowledge of switching between the intermediate states, we can do arithmetic programming with pre-defined pulse trains. An example is given in Fig. 4.

We start from an initial polarization state of $-P_r$ and calculate a voltage pulse train to arrive at $-P_r/2$, 0, P_r, and $-P_r$. The calculated voltage train is shown at the top of Fig. 4, and the electrical displacement measured after each discrete voltage pulse is presented at the bottom of Fig. 4. The small spikes are due to the non-switching component of the ferroelectric capacitor. The plateaus correspond to the electrical displacement after removal of the electric field, equal to the (unsaturated) remanent ferroelectric polarization. A fair agreement with the expected set values is obtained. All measurements are highly reproducible as we carefully avoided fatigue, which for P(VDF-TrFE) is extrinsic, dominated by delamination of the top electrode.[40] Furthermore, within the time frame of the measurements, fatigue of PZT can be disregarded.[41,42] We note that a protocol for deterministic switching between any intermediate states can always be derived based on measured transients of polarization reversal, irrespective of the details of the switching mechanism.[29,43,44]

In summary, we have systematically investigated the retention of intermediate polarization states in capacitors comprising two classic ferroelectric materials, viz. P(VDF-TrFE) thin films and PZT ceramics. Each experiment started from a discharged and electrically depolarized ferroelectric capacitor. We show that the intermediate polarization states are stable over time, up to the Curie temperature. The remarkable stability originates from the coexistence of effectively independent domains, with different polarization due to different values of the coercive field. The stability explains
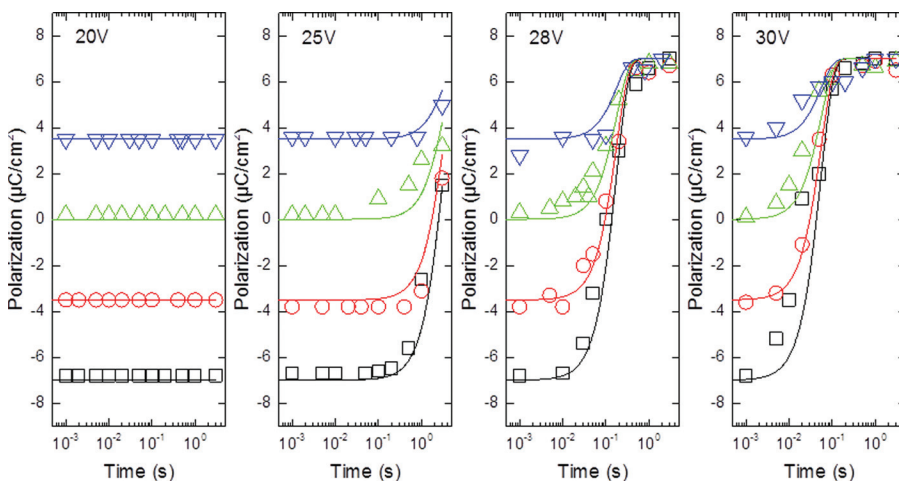

FIG. 3. Switching between intermediate polarization states. The polarization of a P(VDF-TrFE) capacitor is set to an initial value of $P_r/2$, 0, $-P_r/2$, and $-P_r$ denoted by the blue, green, red, and black symbols, respectively. A square, stepwise voltage pulse is applied and the measured polarization is presented as a function of pulse width for various values of the pulse amplitude. The fully drawn curves are calculated from Eq. (1) by substituting reported switching parameters.
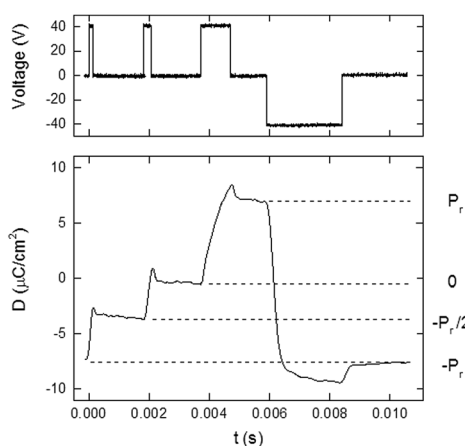
FIG. 4. Deterministic switching between intermediate polarization states. The polarization of a P(VDF-TrFE) capacitor is set at $-P_r$. The top shows the pulse train designed to arrive sequentially at polarization states of $-P_r/2$, 0, P_r, and $-P_r$. The bottom shows the measured polarization transient, in good agreement with the designed polarization values, as indicated by the dotted lines.

the reported excellent data retention of multibit ferroelectric memories using intermediate polarization states. The switching between intermediate polarization states is described by a generalized KAI model. Deterministic switching is quantitatively described using reported phenomenological switching parameters, which allows arithmetic programming, as demonstrated experimentally.

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