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To cite this article: A Herklotz *et al* 2013 *New J. Phys.* **15** 073021

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Strain-controlled switching kinetics of epitaxial $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$ films

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New Journal of Physics **15** (2013) 073021 (7pp)

Received 17 January 2013

Published 9 July 2013

Online at <http://www.njp.org/>

doi:10.1088/1367-2630/15/7/073021

Abstract. We investigate the effect of biaxial strain on the switching of ferroelectric thin films. The strain state of epitaxial $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$ films is controlled directly and reversibly by the use of piezoelectric $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})_{0.72}\text{Ti}_{0.28}\text{O}_3$ (001) substrates. At small external electric fields, the films show switching characteristics consistent with a creep-like domain wall motion. In this regime, we find a huge decrease of the switching time under compressive strain. For larger external electric fields, the domain wall motion is in a depinning regime. The effect of compressive strain is more moderate in this region and shows a reduction in the switching kinetics.

Ferroelectric thin films have been studied intensively in the last few decades because of their practical and fundamental importance. Of particular technological interest is the kinetics of domain switching in that it determines the reading and writing speeds of ferroelectric



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devices. Traditionally, the time-dependent change of the polarization $\Delta P(t)$ of capacitor-like heterostructures has been recorded and explained by appropriate switching models [1–4], the most well known being the Kolmogorov–Avrami–Ishibashi (KAI) model [5, 6]. Recently, piezoresponse force microscopy studies have provided grounds for new insights into the microscopic aspects of ferroelectric domain nucleation and growth [7–10]. It was found that domain wall motion can be understood in terms of a nonlinear dynamic process resulting from competition between elastic and pinning forces [10] and thus can be classified into creep, depinning and flow regimes [7].

Advances in epitaxial growth techniques have led to well-defined strain states of ferroelectric oxide films, and thus opened another way of tailoring ferroelectric thin film properties by ‘strain tuning’. However, investigating the influence of epitaxial strain on the domain dynamics has been virtually impossible until now, since the common approach of growing films in different strain states inevitably leads to varying film conditions, such as different domain configurations or defect concentrations. We have used a piezoelectric monocrystalline substrate of $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})_{0.72}\text{Ti}_{0.28}\text{O}_3$ (001) (PMN-PT) to reversibly strain an epitaxial BiFeO_3 film and, thereby, to directly study the strain dependence of the ferroelectric polarization P_S and the coercive field E_C [11]. A decrease of E_C and an increase of P_S were observed under compressive strain, which is in contradiction to thermodynamic expectations. Hence, a dynamic origin for the observed strain-dependent coercivity was proposed.

Very recently we have shown that the same approach can be used to directly study the strain effect on the switching *kinetics* of ferroelectric BiFeO_3 thin films [12]. This strain effect was found to be strongly dependent on the dominant domain wall propagation regime. In the creep regime, compressive strain resulted in a slowing down of domain dynamics, whereas an acceleration of domain dynamics was observed in the depinning regime. Here, PMN-PT substrates are employed to demonstrate that the switching kinetics $\Delta P(t)$ of epitaxial ferroelectric $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$ (PZT) films is also altered by strain. We show that the effect of strain is essentially opposite to BiFeO_3 . In the creep regime, compressive strain leads to an acceleration of domain dynamics, whereas domain wall motion is slowed down by compressive strain in the depinning regime. The results are discussed by pointing out the differences between the BiFeO_3 and PZT films.

An approximately 200 nm thick film of PZT was grown by pulsed laser deposition from a stoichiometric target in 0.1 mbar O_2 at 610 °C onto a polished PMN-PT(001) substrate covered with a 20 nm thick SrRuO_3 (SRO) electrode layer. Voltages up to 450 V were applied between the SRO layer and a gold electrode on the backside of the substrate, resulting in a maximum reversible biaxial in-plane strain of 0.15% [13, 14]. The lattice parameters of the film were determined by four-circle x-ray diffraction. Polarization measurements were performed in a parallel plate geometry with 0.14 mm² diameter Pt top electrodes using an AixACCT TF 1000 analyzer.

The PZT film exhibits a pseudocubic cube-on-cube epitaxy on SRO/PMN-PT. Figure 1 shows a reciprocal space map around the pseudocubic (103) reflection of the PMN-PT substrate. A tetragonal distortion of the PZT structure is evident with lattice parameters of $a = 4.04 \text{ \AA}$ and $c = 4.10 \text{ \AA}$. The appearance of only one peak suggests that the PZT film is single domain with the c -axis oriented out of plane. Nevertheless, the presence of a very small fraction of a -domains or a similarly small admixture of a rhombohedral phase cannot be excluded by our x-ray diffraction measurements.

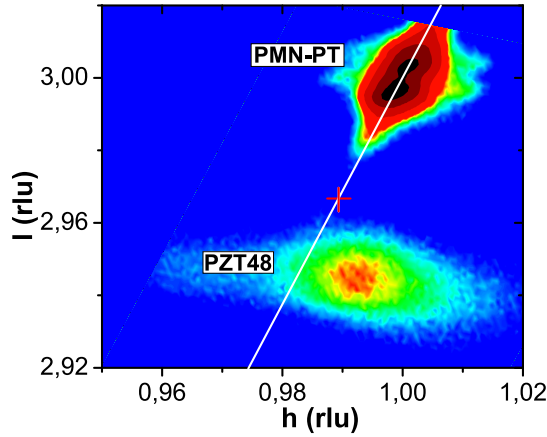


Figure 1. XRD reciprocal space maps around the (103) reflection of the PZT/SRO/PMN-PT heterostructure. The reciprocal lattice units are in terms of the PMN-PT substrate lattice. The white line denotes the positions of cubic unit cells, i.e. where the condition $a = c$ is fulfilled. The SRO layer peak is outside of the shown scan range.

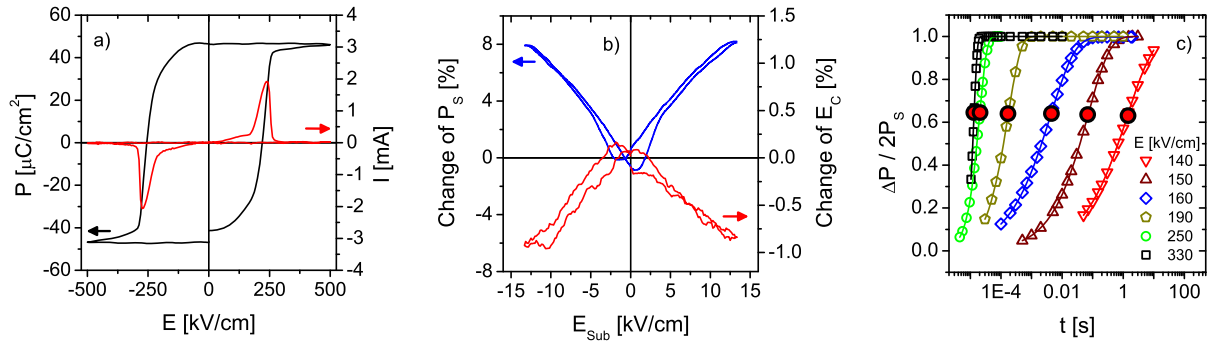


Figure 2. (a) Polarization loop (black) of the PZT capacitor and corresponding switching current (red). (b) Relative change of P_s (blue) and E_C (red) versus the electric field applied at the piezoelectric substrate. (c) Normalized polarization switching curves $\Delta P(t)/2P_s$ for different applied fields E . The solid lines and red dots display the fits to equation (1) and the obtained switching times t_{sw} , respectively.

Polarization measurements were performed in order to study the strain dependence of the ferroelectric properties. Figure 2(a) shows a polarization loop observed at room temperature and a frequency of 1 kHz. Well-defined switching and an almost negligible leakage current contribution allow for an accurate determination of the saturated polarization P_s and the coercive field E_C , which are in agreement with earlier reports [15].

In the following, the direct strain dependence of both characteristic values, P_s and E_C , was measured by application of a bias field E_{sub} to the piezoelectric substrate. In the aforementioned experiment, this method has been shown to provide strain-dependent P_s data in agreement with other experimental and theoretical investigations [11]. Figure 2(b) shows that P_s is enhanced with increasing E_{sub} . Here it should be noted again that an application of $E_{sub} = 10 \text{ kV cm}^{-1}$

corresponds to an in-plane compression of the PZT film of about 0.1% for either sign of the substrate field, since the substrate switches at a coercive field of about 1.5 kV cm^{-1} [13, 14]. Thus, our observation of an increased P_S is perfectly in line with the general tendency that the out-of-plane polarization component of a ferroelectric film is enhanced under in-plane compression. To be more specific, we find a strain dependence $dP_S/d\epsilon$ of $30 \pm 5 \mu\text{C cm}^{-2} \%^{-1}$. In a simple model considering thermodynamic aspects alone [16], one would expect E_C and P_S to have a similar dependence on strain since switching involves the nucleation of domains, and domain-wall energies increase with polarization [11]. However, as depicted in figure 2(b), our experimental finding clearly shows the opposite behavior of E_C . A similar result was found in our earlier work on BiFeO_3 and was explained by the dominance of domain kinetics, which is not included in present thermodynamic models [11]. It is the subject of this work to study the influence of strain on domain kinetics directly.

The polarization reversal during a ferroelectric switching event and domain kinetics are directly correlated. Here, pulse measurements are used to determine the time evolution of the polarization change $\Delta P(t)$ due to domain switching and thus study the switching kinetics of ferroelectric thin films. A series of pulses is applied to determine the switching P_{sw} and non-switching P_{ns} components of the polarization and calculate $\Delta P(t) = P_{\text{sw}} - P_{\text{ns}}$, a method that has been used before [1, 4, 17]. Figure 2(c) shows several $\Delta P(t)/2P_S$ curves at room temperature and different values of E . We find that our $\Delta P(t)$ curves can be well described by the KAI model, which is based on stochastic nucleation and unrestricted domain growth [5, 6]:

$$\Delta P(t) = 2P_S f(t) = 2P_S (1 - e^{(-t/t_{\text{sw}})^n}), \quad (1)$$

where n and t_{sw} are an effective geometric dimension and a characteristic switching time, respectively. Fitting our data to this switching model (solid lines in figure 2(c)) allows for the determination of the characteristic switching time $t_{\text{sw}}(E)$ (red circles in figure 2(c)). We note that the other fit parameter, n , depends on the growth and coalescence mechanisms of the reversed domains, which are not known in detail here. n strongly depends on E and takes a value of 0.5–2, which is near the earlier reported values for similar PZT films.

We now discuss the effect of strain on the switching kinetics. According to equation (1), the value $\Delta P(t)$ is a product of the local spontaneous polarization P_S within a domain and a time-dependent switching function $f(t)$, which represents the domain kinetics of the film. The strain effect on $\Delta P(t)$ can therefore be separated into the strain effect on P_S , which was previously discussed (figure 2), and the strain effect on $f(t)$. Figure 3 shows the measurements for two different values of E in two different strain states, i.e. under a substrate field of $E_{\text{sub}} = 0$ (black) and 15 kV cm^{-1} (red). In figure 3(a) the absolute value of $\Delta P(t)$ is plotted, while figure 3(b) depicts the strain-induced relative change $[\Delta P(t, E_{\text{sub}} = 15 \text{ kV cm}^{-1}) - \Delta P(t, E_{\text{sub}} = 0)]/\Delta P(t, E_{\text{sub}} = 0)$. The latter is useful, as small changes of $\Delta P(t)$ can be revealed. Here, the strain effect on P_S is reflected by a constant offset of $\approx 8\%$. The strain effect on the switching kinetics $f(t)$ is imposed on this offset of 8% in P_S , as illustrated by the colored areas. We find that domain dynamics can be both accelerated or slowed down, as indicated by the areas above and below the 8% offset level under compressive strain depending on the magnitude of switching field E . Under a small applied electric field, the switching time is decreased by compressive strain, thus giving a positive contribution (green area). In contrast, when a high E is applied the ferroelectric switching is slowed down under compressive strain, which in turn leads to a negative contribution (blue area) to the relative change of the polarization.

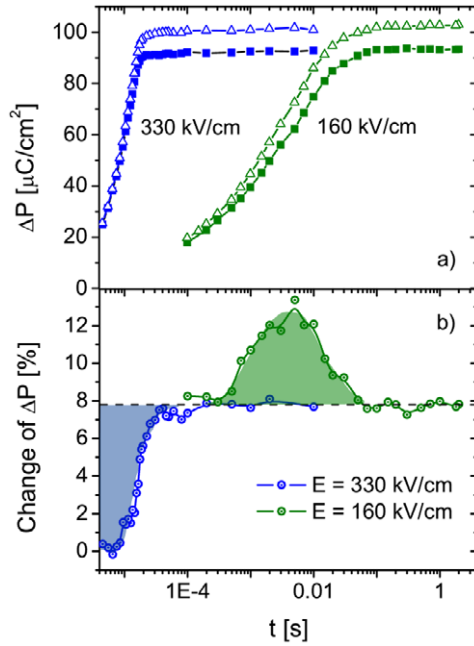


Figure 3. (a) Switched polarization versus writing time, $\Delta P(t)$, for two different electric fields E (330 and 160 kV cm $^{-1}$) applied to the capacitor in two different strain states (substrate electric field of $E_{\text{sub},1} = 0$ (squares) and $E_{\text{sub},2} = 15 \text{ kV cm}^{-1}$ (triangles)). (b) Strain-induced relative change $[\Delta P(E_{\text{sub}} = 15 \text{ kV cm}^{-1}) - \Delta P(E_{\text{sub}} = 0)] / \Delta P(E_{\text{sub}} = 0)$ of the same data.

The switching time of a ferroelectric capacitor is determined by two factors, the nucleation rate and the (average) domain wall velocity. The measured capacitors have a relatively large area. Therefore, a large number of nucleation centers can be expected to appear immediately after the field application, and the wall velocity is likely to be the limiting process for the switching [18]. In similar large-area PZT capacitors, the domain wall velocity v was found to be approximately proportional to $1/t_{\text{sw}}$ [10]. Therefore, we assume here that t_{sw}^{-1} represents the average domain wall velocity in order to enable the identification of the different regimes of wall motion. To further demonstrate the intriguing difference between high- and low-field behavior, figure 4 shows the strain effect on the switching time t_{sw} as a function of E in three different representations. Figure 4(a) depicts the strain-induced relative change of t_{sw} . It is evident that at low external voltages the switching time is reduced strongly under piezoelectric compression, whereas at high voltages a moderate increase is observed. We suggest that this behavior is related to different domain wall motion regimes. Recent studies revealed that the domain wall propagation in ferroelectric films is strongly nonlinear with electric field and can be described in terms of a pinning scenario [8, 10, 19]. At small fields the domain walls are strongly pinned by local disorder and the propagation is a thermally activated motion between pinning sites. In this creep regime the domain wall velocity v depends exponentially on the external electric field E : $v \sim e^{-U_{\text{eff}}/E^\mu}$, where U_{eff} is an effective energy barrier and μ is an exponent reflecting the nature of the pinning potential. The effect of strain in the creep regime is demonstrated in figure 4(b). In this Arrhenius-like plot the domain wall creep motion is reflected in a linear behavior, as stressed by linear fits in the graph, when μ is assumed to be 1.0. Similar values of μ have been

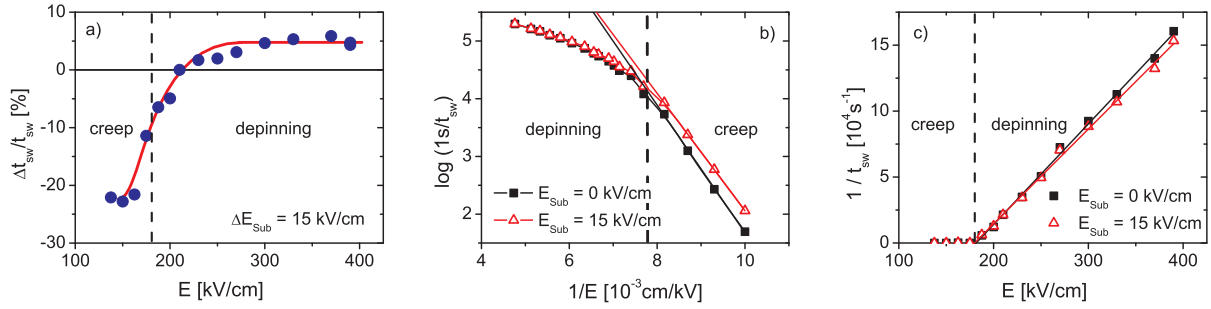


Figure 4. (a) Relative change of the switching time t_{sw} under 0.15% of biaxial compression for various electric fields E . (b), (c) Switching times t_{sw} in two strain states (same data set, substrate fields indicated in the plots) plotted for analysis of the creep regime (b) and the depinning regime (c). Fit curves in (b) explained in the text. Linear extrapolation lines in (c) used to estimate the crossover between the creep regime and the depinning regime (dashed vertical lines in all panels).

reported before for PZT films [7, 10]. Evidently, strain is affecting the slope of the curve, i.e. the magnitude of the energy barrier U_{eff} is decreased under compression.

At an external field E_{C0} , the driving force is large enough to overcome the energy barriers and a pinning–depinning transition occurs, with v increasing considerably with E . It can be concluded from figure 4(c) that creep-like domain wall propagation is present below $E_{C0} \approx 180$ kV cm $^{-1}$ (as estimated from a linear extrapolation of the high-field data to $1/t_{sw} = 0$), and the depinning regime is reached above. Note that the fields examined here are not high enough to enter the flow regime. In the depinning regime, compressive strain is found to reduce the domain wall mobility v/E .

The unexpected negative strain response of E_C upon compressive strain (figure 2(b)) can now be understood better. The electric field during the hysteresis measurement (figure 2(a)) runs through creep and depinning ranges, leading to a partial compensation of the strain effects in both regimes. In the present sample, the larger part of the polarization switches in fields where the switching is accelerated according to figure 4(a), resulting in a reduction of E_C under compressive strain. We note, however, that a quantitative evaluation of the strain-dependent E_C would be quite complex.

Finally, we want to compare our results on PZT films with our studies on BiFeO $_3$ films [12]. We stress that the changes of the strain effect with the external field E are strikingly similar, i.e. it strongly depends on the domain wall motion regime. However, the sign is opposite for both cases. Presently, we can only speculate about this observation, as the origin of the strain effect itself needs further exploration. The differences might be governed by intrinsic properties of the ferroelectric BiFeO $_3$ and PZT materials or might be induced by extrinsic contributions from defects in the films or from the interface to the electrodes. We note that the crystal structures of both materials are different. While PZT is assumed to be tetragonal, BiFeO $_3$ exhibits a pseudorhombohedral structure. The kinetics of the motion of domain walls varies significantly between tetragonal and rhombohedral domains. Thus, the strain effect on the domain wall motion may have an opposite sign due to different domain configurations.

In summary, applying reversible strain from a piezoelectric substrate to an epitaxial PZT film allows us to directly probe the strain effect on ferroelectric switching kinetics. We show that the influence of strain crucially depends on the regime of domain wall motion. Under small E , creep-like domain wall motion is present and switching is accelerated considerably under compressive strain. This finding might offer a new way of improving the reliability and endurance of thin film applications, as retention loss due to domain wall creep can be reduced by appropriate strain tuning. In the depinning regime, compressive strain reduces the domain wall mobility and thus slows down ferroelectric switching of our PZT film. We note here that there is a lack of theoretical understanding for the strain effect in both the creep regime and the depinning/flow regime. However, our experimental approach now provides the necessary access to meaningful data that will eventually help us identify the relevant intrinsic and defect-dominated parameters.

Acknowledgments

We acknowledge support from DFG FOR520 and the Scientific User Facilities Division, Office of Basic Energy Sciences, US DOE.

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