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Stability of ferroelectric bubble domains

Vivasha Govinden^{1*}, Suyash Rijal^{2*}, Qi Zhang^{1†}, Yousra Nahas², Laurent Bellaiche²,
Nagarajan Valanoor¹, Sergei Prokhorenko^{2†}

¹ *School of Materials Science and Engineering, University of New South Wales, Sydney, NSW
2052, Australia*

² *Physics Department and Institute for Nanoscience and Engineering, University of Arkansas,
Fayetteville AR 72701, USA*

* these authors contributed equally

† corresponding authors:

Qi Zhang: peggy.zhang@unsw.edu.au

Sergei Prokhorenko: sprokhor@uark.edu

Abstract

Nanoscale ferroelectric topologies such as vortices, anti-vortices, bubble patterns etc. are stabilized in thin films by a delicate balance of both mechanical and electrical boundary conditions. A systematic understanding of the phase stability of bubble domains, particularly when the above factors act simultaneously, remains elusive. Here we present first-principle-based simulations in combination with scanning probe microscopy of ultrathin epitaxial (001) $\text{PbZr}_{0.4}\text{Ti}_{0.6}\text{O}_3$ heterostructures to address this gap. The simulations predict that as-grown labyrinthine domains will transform to bubbles under combinations of reduced film thickness, increased mechanical pressure and/or improved electrical screening. These topological transitions are explained by a common fundamental mechanism. Namely, we argue that, independently of the nature of the driving force, the evolution of the domain morphology allows the system to conserve its original residual depolarization field. Thereby, the latter remains pinned to a value determined by an external or built-in electric bias. To verify our predictions, we then exploit tomographic atomic force microscopy to achieve the concurrent effect of reducing film thickness and increased mechanical stimulus. The results provide a systematic understanding of phase stability and demonstrate controlled manipulation of nanoscale ferroelectric bubble domains.

Topological structures in ferroelectric systems have recently drawn immense attention. For example, topological configurations such as flux closures[1-4], vortex pairs[5-8], skyrmions[9, 10] or nanoscale bubble domains[11, 12] have led to emergent properties including high conductivity[13], chirality[14, 15], negative capacitance[16-19], and giant electromechanical response[11]. These attributes make polar topologies promising candidates for various device applications, especially where they can be deterministically controlled.

The stability of dipolar topological patterns is subject to a subtle balance of mechanical and electrical boundary conditions[11-20]. For instance, in the case of polar vortices we now know there is a fine balance between long-range electrostatic and elastic versus short-range polarization gradient interactions that determine vortex stability[21]. Damodaran et al. exploited this balance to practically realize phase co-existence where interconversion from the topological vortex phase to a ferroelectric a_1/a_2 phase resulted in giant piezoelectric and non-linear optical response[22]. More than a decade ago, Balke et al. reported the controlled creation of a ferrotorroidal order parameter by exploiting the trailing fields of a scanning probe tip[23]; this approach was most recently used to create a vortex-anti vortex pair by Kim et al. in an otherwise topologically trivial ferroelectric canva [5].

In contrast, understanding of the phase stability of polar bubbles in ferroelectric films still has open questions. On the one hand, we know that electric bubbles share many similarities with their spin siblings – magnetic bubbles[24] and skyrmions[25, 26]. For instance, electric bubbles were reported to emerge from modulated phases [27] (stripes[12] or labyrinths[28, 29]) under an external electric field following the well-documented transformation paths of magnetic stripes, conical or helical spirals and spin labyrinths into bubbles or skyrmions exposed to a magnetic field bias[25]. Moreover, polar bubbles result from the frustration induced by long-range dipolar interactions[29] which is also one of the first reported mechanisms of magnetic bubble stabilization[24, 30]. On the other hand, the electric dipolar interactions are much stronger than their magnetic counterpart and can be significantly tuned by dielectric spacers, charge doping or changing films thickness[31]. Additionally, ferroelectrics are arguably more sensitive to mechanical boundary conditions and stimuli. As a result, the physics of electric bubbles is largely determined by the balance of electrostatic interactions and electrostriction and is less affected by short-range interactions that play an important role in magnetic materials.

At the structural level, this difference manifests in the nanoscale $<10\text{nm}$ radius of polar bubbles (compared to $>100\text{nm}$ radius of magnetostatically stabilized bubbles in magnetic films) and their characteristic vortex ring geometry[12] inherited from the polar stripe phase (Fig. 1a-c). The achiral bubbles shown Fig. 1c are typical of $\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3/\text{SrTiO}_3/\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$ (PZT/STO/PZT) heterostructures and are invariably associated with Néel skyrmion textures within the in-plane cross sections of the ferroelectric layer [28][29]. A homotopy equivalent polar skyrmion bubbles schematically shown in Fig. 1d were reported in $\text{PbTiO}_3/\text{SrTiO}_3$ (PTO/STO) superlattices[32]. The latter structures are chiral due to an additional Bloch component arising at the center of 180° domain walls in PTO. Consequently, these two types of polar topologies appear under different external conditions[9, 28, 33].

At a more practical level, the size and sensitivity of polar bubbles to boundary conditions poses challenges but also offers new levers to control and functionalize these nanoscale domains. Indeed, experimentally stabilizing bubble domains is not a trivial task; even a modest change in the external boundary conditions triggers a rapid phase transition to labyrinthine or monodomains[11, 33]. The bubble domains have significant importance for technology as (i) they are a precursor to the much sought- after electrical skyrmion[32], (ii) promise giant piezoelectric response[11], (iii) can be realized in free-standing films[34] and (iv) possess dynamic intrinsic conductivity[35] that could be harnessed in neuromorphic computing[28]. Although we have captured the individual effects of factors such as thermal and bias profiles[28], mechanical pressure[33], thickness[36] and screening[37], exploiting these properties for a topological device requires knowledge of what happens when several of these external tuning parameters act in tandem.

This report provides an in-depth understanding of the phase stability of bubble domains in ferroelectric thin films under such simultaneous stimuli. Starting with first-principle-based effective Hamiltonian simulations as guide, we investigate the simultaneous effects of film thickness, external mechanical force and screening conditions on the topological defect transition mechanisms in epitaxial (001) $\text{PbZr}_{0.4}\text{Ti}_{0.6}\text{O}_3$ (PZT) thin films. Our simulations show that the mechanical stress normal to the film's surface, the decreased films thickness as well as enhanced electric screening have the same effect: the thinning and disconnecting of the domain pattern. We further show that, in all cases, the resulting sequence of labyrinth-bubble-monodomain transitions[28] can be explained by an unusual “conservation law” – an intrinsic tendency of the ferroelectric film to preserve its original depolarizing field. These predictions

are then verified experimentally by exploiting the newly developed tomographic atomic force microscopy (TAFM) technique[38, 39] on $\text{PbZr}_{0.4}\text{Ti}_{0.6}\text{O}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3//\text{SrTiO}_3$ (PZT-single layer) and PZT/STO/PZT/LSMO//STO (PZT-sandwich) films respectively under a variety of mechanical forces and at different thicknesses. Under compressive stress (applied through the TAFM tip), as-grown labyrinthine-shaped domains transform to disconnected labyrinths (PZT-single layer) or nanoscale bubbles (PZT-sandwich) dependent on the screening conditions. On the other hand, TAFM milling, i.e., which reduces thickness, results in bubble domains in both film configurations. The experiments confirm that the stability of these different ferroelectric topologies is ultimately linked to how mechanical stress, screening or thickness of the system modulate the residual depolarization field[40, 41] in the thin film heterostructure.

We begin with the effective Hamiltonian simulations of $\text{PbZr}_{0.4}\text{Ti}_{0.6}\text{O}_3$ ultrathin films[20] that allow us to understand how each of the boundary conditions govern the topological defect structures. For all cases we assume the films are (001) oriented with an imposed in-plane biaxial epitaxial strain of -2%[42] and a built-in bias field E_b of 0.18 V/nm. Full details of simulations are provided in Supplemental Material [43] and the references [44-45] therein. Figure 2a-e shows various simulated domain patterns for the investigated range of external boundary conditions.

In the virgin state, our simulations predict that the domains have a disconnected labyrinthine-like morphology (Fig. 2a). Upon applying an increasing homogenous stress (σ) perpendicular to the film surface, the labyrinthine domains progressively disconnect, transform into bubbles and eventually disappear to yield a monodomain state. Fig. 2b demonstrates the simulated domain pattern precursor to the bubble phase at $\sigma = 1$ GPa compressive stress. The same sequence of labyrinth-bubble-monodomain transitions is predicted by simulations when either enhancing the screening β (e.g., from $\beta=0.86$ in Fig. 1a to $\beta=0.88$ Fig. 2c) or decreasing the film thickness h (e.g., from $h=8$ u.c. in Fig. 2b to $h=7$ u.c. in Fig. 2d). Moreover, this chain of domain pattern transformations is not altered when tuning several external parameters. All factors work hand-in-hand and adjusting one variable allows to ease the transitions triggered by the other. For example, ultra-fine bubbles (Fig. 2e) are seen with just half the applied stress ($\sigma=0.5$ GPa) when the PZT thickness is reduced ($h = 7$ u.c) and screening conditions are enhanced ($\beta=0.88$). Notably, the polar structure remains achiral for all observed domain morphologies (see Supplemental Material [43]).

To understand the physics of the described transitions, we first recall that the non-trivial domain topologies in ultra-thin PZT films fall within the family of universal self-organized patterns commonly found in phase separation kinetics[28]. The link is granted by the condition of the bias field (E_b) compensation by the residual depolarizing field (E_d) that holds up to the monodomain state (i.e., it is valid for the labyrinthine and bubble patterns) (see Supplemental Material [43]). Here, since the bias magnitude E_b always remains fixed, the $E_d = E_b$ relation simply means that the residual depolarizing field is conserved under any change of external parameters.

The signatures of conserved depolarization are seen in all of our present simulations. For example, we find that, under mechanical loading, the out-of-plane polarization P_z only weakly responds to the applied stress despite a strong suppression of the average magnitude of local electric dipoles $|\mathbf{d}|$ (Fig. 1f). This assures a steady level of the bound charge at the interfaces thereby guaranteeing a constant E_d magnitude. Microscopically, the only way to maintain P_z under reducing $|\mathbf{d}|$ is by switching more dipoles along the bias. Such switching progressively disconnects labyrinth domains and leads to bubble formation when σ increases. The scenario of β -engineering is somewhat opposite. Unlike stress, the screening strength directly affects the depolarizing field and to compensate for a linear drop of E_d with increasing β (Fig. S1 of the Supplemental Material [43]) the polarization grows as $P_z \sim 1/(\beta_{SC} - \beta)$ (Fig. 2g). Here, β_{SC} denotes the screening strength at which the residual depolarizing field vanishes. In the linear approximation, β_{SC} also corresponds to critical screening at which the monodomain pattern forms. At the same time, our simulations show that $|\mathbf{d}|$ remains basically invariant up to the bubble-monodomain transition (Figure 2g). Therefore, the growth of P_z is, once again, achieved by switching the dipoles so as to increase the area of the domain polarized along the bias. Finally, for the thickness driven transitions both microscopic mechanisms of depolarizing field conservation come into play. Namely, when the thickness is reduced, the dipole magnitude $|\mathbf{d}|$ decays (Fig. 2h) just as in the case of increasing mechanical force. Such weakening of $|\mathbf{d}|$ is due to an enhanced role of fluctuations at reduced thickness and dimensional confinement. Moreover, increasing fluctuations enhance the dielectric permittivity ϵ and, in order to conserve $E_d = P_z/\epsilon$, the polarization is forced to increase when the thickness is reduced (Fig. 2h). The latter is similar to the enhanced screening scenario.

Testing the validity of the simulations requires an experimental method that facilitates the simultaneous application of electric field and mechanical pressure, change in thickness as

well as screening conditions. Fortunately, recent advances in scanning probe microscope (SPM) techniques[38, 46-48] allow us to image, characterize, as well as manipulate the topological domain patterns under a variety of external boundary conditions with a resolution of up to a few nanometers[6, 33, 46, 49, 50]. In this work we exploit the recently developed TAFM[38, 39]. Briefly, a stiff scanning probe with a large force constant is employed such that the applied force is sufficient to mechanically remove a layer of the thin film during scanning and hence access a fresh tomographic surface[38] (A tomographic surface is defined as the surface acquired after milling off its above layers). As milling takes place, the thickness of the residual layer decreases (Fig. S2 of the Supplemental Material [43]). To incorporate the effect of screening, we compared two candidate (001) epitaxial heterostructures- viz. (i) PZT-single layer and, (ii) PZT-sandwich (synthesis details found in Supplemental Material [43]). The TAFM calibration parameters for both systems can also be found in Supplemental Material [43]. Suffice to state here, that a critical force of 7 μN is needed to mill the layers in the PZT-sandwich and, and typically each 7 μN scan causes approximately 4 ± 2 nm depletion. On the other hand, a force of 15 μN is required to mill through the PZT-single layer. Each TAFM scan is followed by an individual piezoresponse force microscopy (PFM) scan to acquire the domain features at the specific surface depth.

We start with the effects of mechanical force and film thickness on the domain patterns of the PZT-single layer. Fig. 3a reveals the single layer possesses labyrinth domains in the as-grown state with a width of approximately 50 ± 10 nm; care was taken to acquire this image under 100 nN force, which has negligible influence on the domain pattern[33] The full images of PFM amplitude and phase are given in Fig. S3 of the Supplemental Material [43]. When the scanning force is increased by an order of magnitude or two (i.e., from 100 nN to ≥ 1 μN), the as-grown poled-up labyrinthine domains break down into smaller patterns domain transitions, as shown in Fig. 3b. Note that no topography change is observed, suggesting that the transitions are not correlated with either surface damage or ferroelastic switching. Under 15 μN , the wide labyrinth domains shrink into either nanoscale bubble, or disconnected labyrinthine domains (with a length and width of approximately 100 nm and 20 ± 5 nm, respectively). Repeated scans at 15 μN cause PZT milling and when 4 nm of PZT is milled, domains further shrink into a bubble domain only configuration (Fig. 3c).

On the other hand, as-grown domain patterns in PZT -sandwich film are narrower labyrinthine domains with a width of 30 ± 5 nm (Fig. 3d, full images are given in Fig. S4 of the

Supplemental Material [43]). Given that both PZT-single layer and PZT-sandwich film have similar out-of-plane lattice parameters (X-ray diffraction and structural data given in Fig. S5 of the Supplemental Material [43]) and comparable screening conditions from electrodes (as they have same LSMO thickness, i.e., 40 nm), the contrast in domain size between two films arises from the differences in their respective electrical boundary conditions (i.e., internal screening) induced by the presence (or absence) of the STO spacer[11]. Under a relatively smaller force (i.e., $\sim 5 \mu\text{N}$), the as-grown narrow labyrinthine domain patterns change into nanoscale bubble-like domains. The average planar domain size is approximately 10 nm as shown in their amplitude images (Fig. 3e).

The above data agree with the simulations under mechanical force (Fig. 2a-b), demonstrating a simple route of acquiring stable nanoscale bubble domains with the application of external mechanical force on as-grown labyrinthine domains. Under the external mechanical force applied using the SPM probe, the film is subject to an out-of-plane compressive strain [46]. As discussed in the simulations (Fig. 2f), this compressive strain drives the polarization switching to increase the area of the matrix domain. An increased force to $7 \mu\text{N}$ results in milling off 4 nm PZT in the PZT-sandwich layer, as well. Although the ultimate topological structure in both systems corresponds to bubble-like domains (Fig. 3c and Fig. 3f), the evolution pathway and driving forces are markedly different. Particularly for the sandwich film, we found the bubbles seen in Fig. 3e undergo a transition to short labyrinths with reducing thickness. This evolution pathway is addressed next.

TAFM of the sandwich film under progressive mechanical loading was carried out to reveal the evolution of the ferroelectric topology when reduced film thickness and mechanical force act simultaneously. Milling precise layer-by-layer of the film surface materials using TAFM conditions followed by PFM imaging facilitates a direct comparison of the domain patterns of the as-grown surface with tomographic surfaces at different depths. The PFM amplitude figures of PZT -sandwich film under $1 \mu\text{N}$ scanning (Fig. 4a), after $4 \mu\text{N}$ scanning (Fig. 4b), and after $7 \mu\text{N}$ scanning for one to four times (Fig. 4c-f), are compared in Fig. 4. The dashed rectangular box locks in the reference location. Three typical regions (I, II and III) are selected to facilitate the understanding of domain evolutions as a function of scanned surface depth (as highlighted in Fig. 4a and schematically illustrated in Fig. 4g).

Below $4 \mu\text{N}$ mechanical force, no milling takes place and the domain patterns (Fig. 4a and 4b) obtained correspond to those on the film surface. As-grown labyrinthine domains (e.g.,

in Regions I and II) break down into nanodomains when the force increases from 1 μN to 4 μN . With the scanning force further increased to 7 μN , the film starts to be milled off and the tomographic surface reaches the depth of 4 ± 2 nm (Fig. 4c). At this stage, some nano domains remain as fuzzy bubble configurations with even smaller planar sizes of approximately 3-10 nm (e.g., in Region I), while newly nucleated nanodomains are found in Region II (indicated by white arrows). Thus, when the scanning surface is above the STO spacer, domains evolve from labyrinthine towards smaller nanoscale domains due to the decreasing thickness of the PZT layer. With the removal of STO spacer (i.e., from Fig. 4d), domains evolve significantly due to the variation of the screening conditions[11]. Particularly in Region III (dashed oval in Fig. 4a-f), new labyrinthine-like domains are formed in the bottom PZT layer (Fig. 4d-f) which are completely different from the ones at film surface (i.e., no domains are observed in Region III surface, Fig. 4a).

In addition, we highlight most of the nanodomains from Region II in 4 nm depth layer are absent in the next 8 nm depth layer (Fig. 4d). Also, some nanodomains in surface layer are found to disappear in the 4 nm tomographic surface layer. This suggests such nanodomains domains only exist within the 4 nm thickness. All such nanodomains also exhibit fuzzy wall features consistent with bubble domains[11]. Therefore, using TAFM technique, the domain patterns can be changed along labyrinthine $\rightarrow$ disconnected labyrinthine $\leftrightarrow$ nanoscale bubble domains pathway through applying the external force and changing residual film thickness. The results confirm the universal breakdown of labyrinthine (Fig. 4a) into bubble domains (Fig. 4b) under external mechanical pressure, in alignment with the numerical simulations of Fig. 2a-b. When this mechanical pressure is accompanied by milling, i.e., the effective ferroelectric thickness is being reduced, the bubble domains become finer (Fig. 4c), confirming thickness reduction in tandem with external compressive mechanical force, results in finer bubbles (Fig. 2d). Hence domains with specific topological configurations (or transitions) can be deterministically induced by precise control of the depth penetration of the mechanical force, highlighting the opportunity for tailored external condition within TAFM.

These above observations in conjunction with our previous work now allow us to provide a complete understanding of the phase stability of ferroelectric bubble domains:

- Topology of polar domains is determined by the balance of the depolarization and bias fields. The former can be indirectly tuned by adjusting the thickness of the PZT layers, changing the screening conditions or exerting an out-of-plane

mechanical stress. Modifying any of these parameters shifts the balance which triggers the domain pattern evolution needed to restore the residual depolarization.

- Ferroelectric bubble domains are an intermediary topological state between the stripes/labyrinth domains and the monodomain state[11, 28]. This “Goldilocks” state is most readily realized experimentally by the insertion of an ultrathin dielectric spacer layer between two c-axis oriented epitaxial ferroelectric layers which are under modest in-plane compressive strains.
- Local mechanical pressure can be exploited to trigger topological defect transitions; the sequence of phase stability goes from labyrinth→bubbles→monodomain under a compressive strain applied along the polarization direction. Under the influence of mechanical pressure, as-grown bubbles can be erased to form a monodomain state[11], analogous to mechanical writing of polarization[44]. On the other hand, mechanical pressure can also trigger the breakdown of as-grown labyrinthine domains into bubbles.
- Thermal quenching leads to either configuration depending on the strength of the applied or built-in electric fields. Thermal quenching as a route to capture non-equilibrium states, results into labyrinthine domains (or bubbles) under the absence (or presence) of an electric field[28].
- Interface effects can be further exploited to engineer topological defect transitions. Varying the thickness of the ferroelectric and/or dielectric layer in a ferroelectric/dielectric/ferroelectric heterostructure modifies the strain, screening conditions as well as the bare depolarization field and thus plays a significant role in the resulting domains. Under varying thicknesses of these layers, a transition from labyrinthine to bubble domains can be triggered[36, 37]. A similar domain transition occurs upon milling through the ferroelectric layer.

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43. See Supplemental Material at [URL will be inserted by publisher] for the description of materials and methods employed in this work, derivations supporting conservation of residual depolarizing field as well as additional simulations data and experiments characterizing the domain and crystallographic structures of PZT-single layer and PZT/STO/PZT-sandwich heterostructures.
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Figure captions

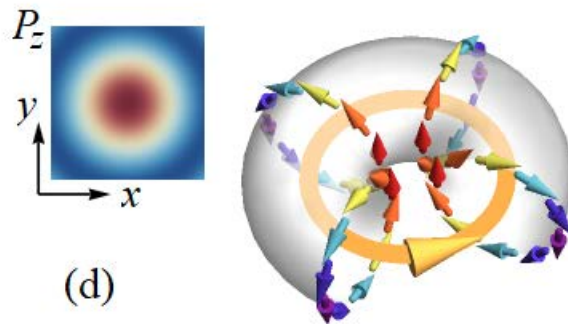
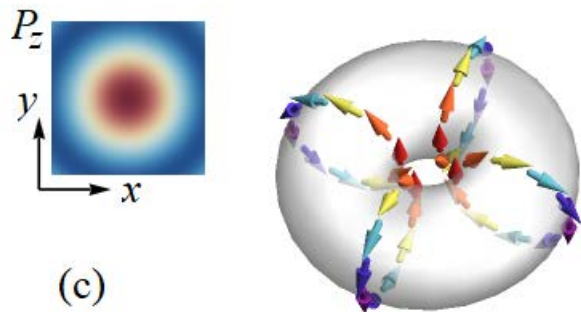
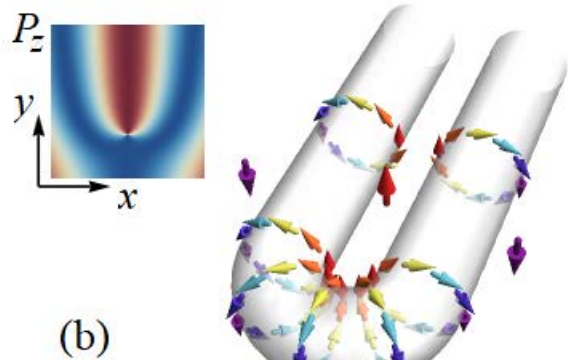
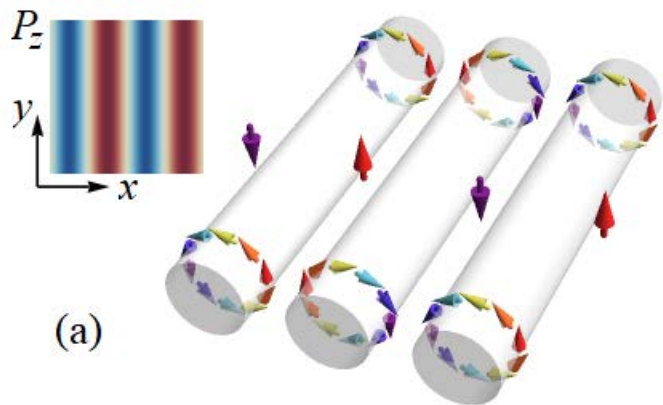
Figure 1. (a) Dipolar structure of the stripes domains in ultrathin PZT films[20]. The upper left inset shows the distribution of the out-of-plane component of polarization P_z within the middle (001) plane of the PZT film where one can see the “up” (red) and “down” (blue) polarized domains. The corresponding nanostripe regions are separated by polar vortex tubes as schematically shown in the right subpanel where the arrows represent local electric dipoles. Such tubes are bounded from the top and bottom by films interfaces. (b) An electric field bias causes the stripe domains to break creating polar disclinations or merons[28] shown in the upper left inset. This leads to reconnection of vortex tubes as shown in the right subpanel. (c) The breaking of stripes eventually creates achiral polar bubbles. The corresponding polarization pattern can be described as the closure of vortex tubes into polarization vortex rings. (d) Chiral bubble domains were reported in PTO/STO superlattices[18]. The latter structures possess an additional Bloch component of polarization (golden arrow) inherited from the ferroelectric transition at 180° domain walls in PTO[51].

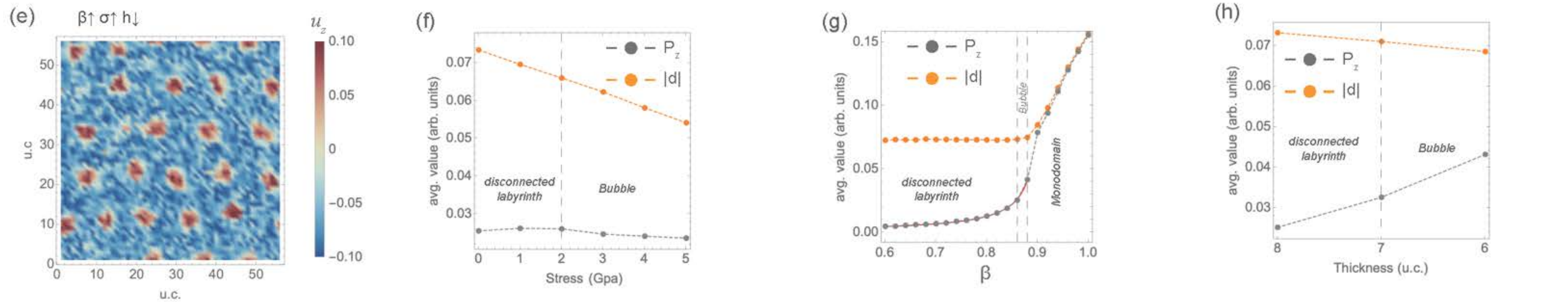
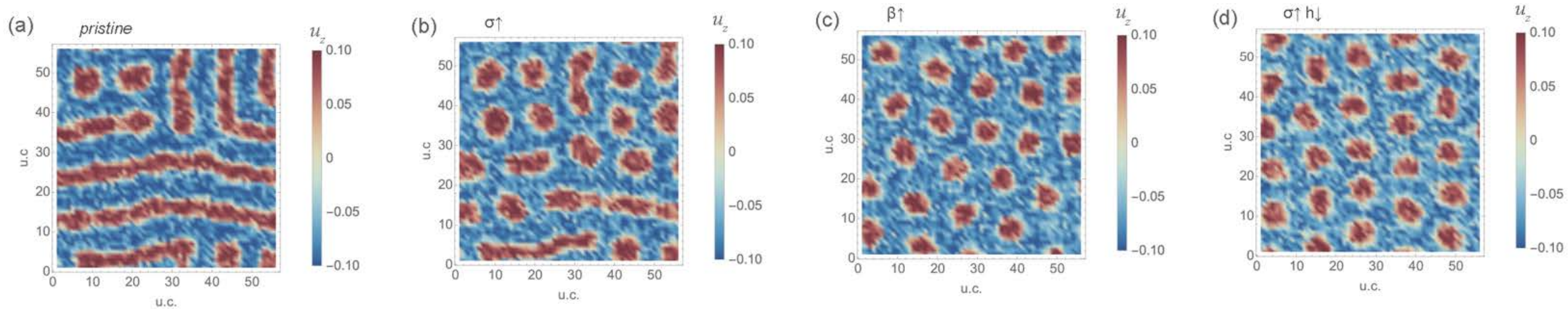
Figure 2. Effective Hamiltonian simulations of domain structure for different PZT film thickness h , screening β and the out-of-plane stress σ . (a) Labyrinth domains at $\sigma = 0$ GPa, $\beta=0.86$ and for $h=8$ u.c. (b) Disconnected labyrinth pattern with bubble domains at $\sigma = 1$ GPa, $\beta=0.86$, and $h=8$ u.c. Simulated arrays of bubble domains for (c) $\sigma = 0$ GPa, $\beta=0.88$, $h=8$ u.c and (d) $\sigma = 1$ GPa, $\beta=0.86$, $h=7$ u.c. (e) Ultra-fine polar bubbles at $\sigma = 0.5$ GPa, $\beta=0.88$, $h=7$ u.c. Dependence of the average dipole magnitude $|d|$ and the out-of-plane polarization P_z on (f) applied stress σ for $h=8$ u.c and $\beta=0.86$ (g) screening β for $h=8$ u.c. and $\sigma = 0$ GPa and (h) thickness h for $\sigma = 0$ GPa and $\beta=0.86$. Dashed lines indicate transitions. Solid red line in (g) shows the fit $P_z = C/(\beta_{SC} - \beta)$ with fitting parameters C and β_{SC} . Panels (a-e) show the middle-(001)-plane cross-sections of a supercell where red (blue) colors indicate dipoles pointing along $[001]$ ($[00\bar{1}]$) pseudo cubic direction.

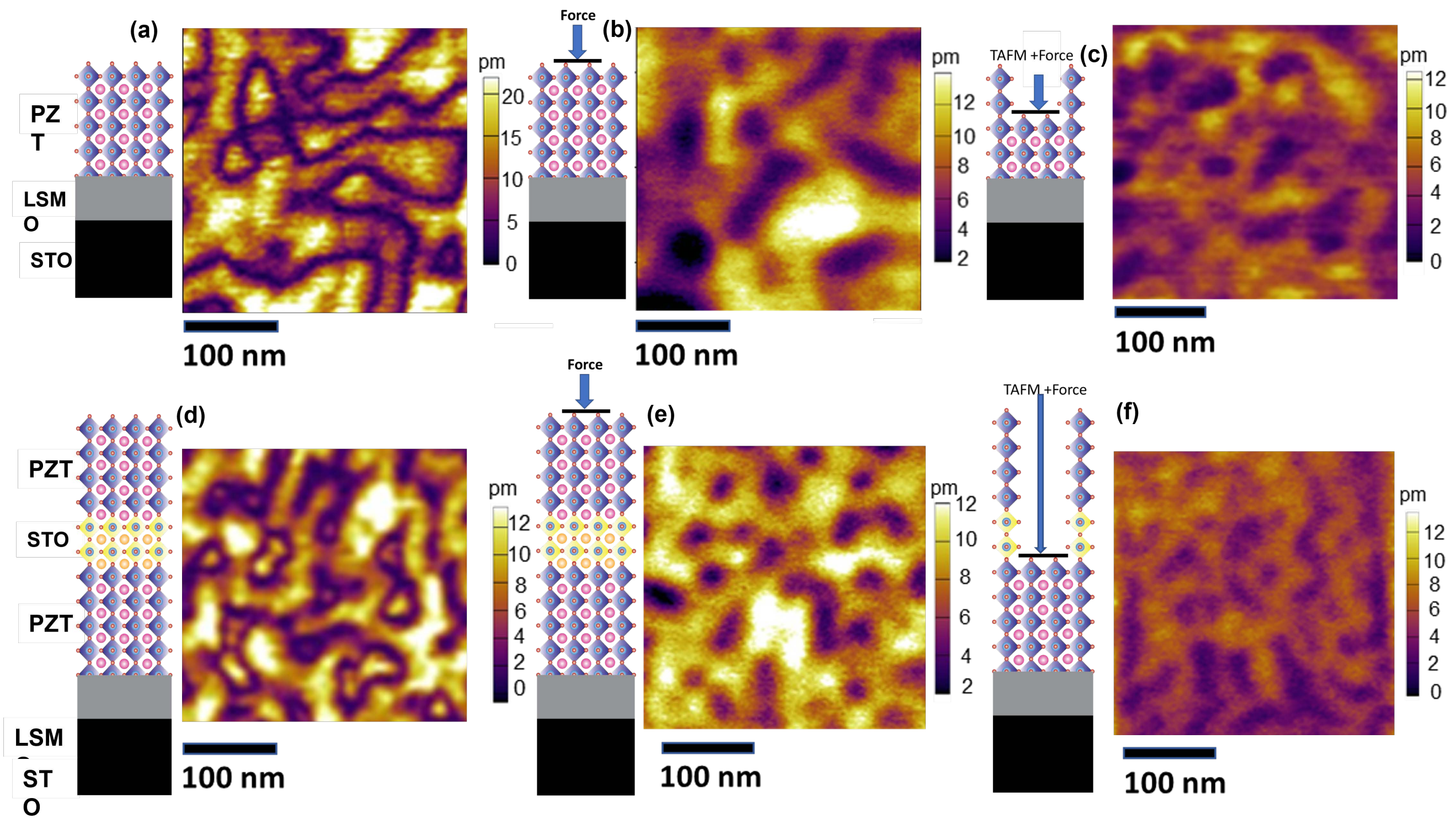
Figure 3. (a)-(c) domain patterns of in PZT-single layer under various conditions: (a) As-grown large labyrinthine domains. (b) Smaller labyrinthine domains are obtained under mechanical force. (c) Bubble domains are formed when ~ 4 nm of PZT is milled off. (d)-(f) domain patterns of in PZT/STO/PZT-sandwich film under various conditions: (d) As-grown

(thinner) labyrinthine domains. (e) Smaller labyrinthine domains and bubbles are obtained under mechanical force. (f) After milling through first PZT layer, film configuration is similar to (b) and domain patterns also look alike (small labyrinthine + bubble domains). After further milling through bottom PZT, bubble domains are formed. The scanning surfaces are indicated by the red line in the corresponding schematics. Note the schematics are a guide and do not reflect the real unit cell numbers.

Figure 4. Domain configuration as a function of force and depth: (a) 1 μN , (b) 4 μN , (c) 7 μN after 1 scan, (d) 7 μN after 2 scans, (e) 7 μN after 3 scans, and (f) 7 μN after 4 scans. The white box showing the reference domain area. With the force increases, the domains first changes into bubble -like domains, and then labyrinthine domains were observed again at a lower depth. (g) Schematic of domain evolutions as a function of force and depth in Region I, Region II and Region III in (a).





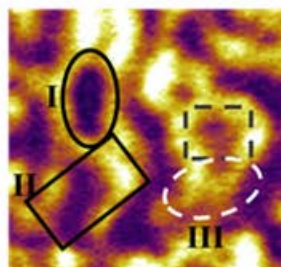


Top review

(a)

Force 1 μN

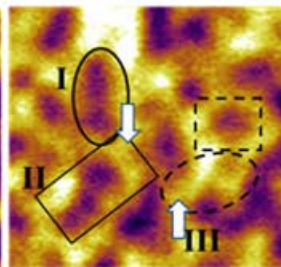
Depth 0 nm



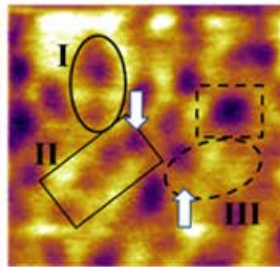
(b)

4 μN

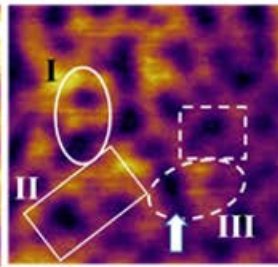
0 nm



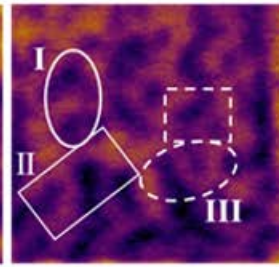
(c)

7 μN x 1 scan 4 ± 2 nm

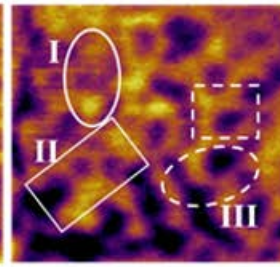
(d)

7 μN x 2 scans 8 ± 2 nm

(e)

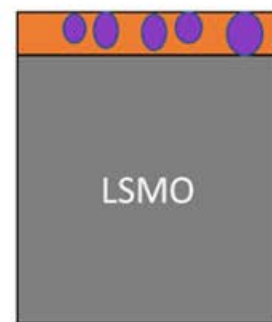
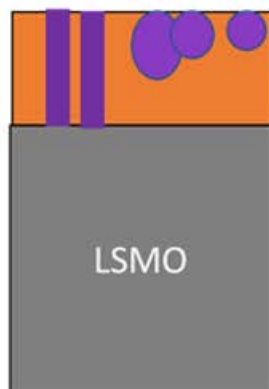
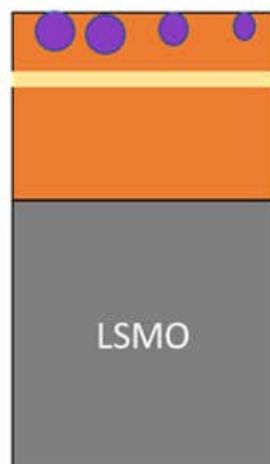
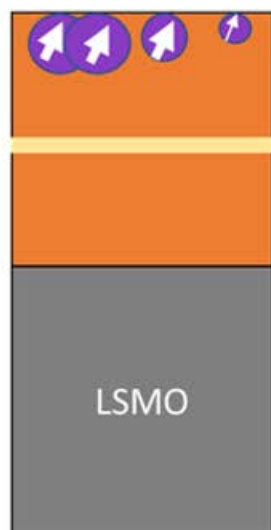
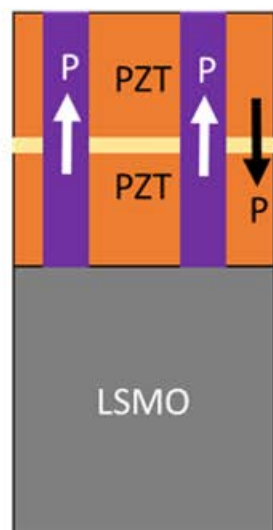
7 μN x 3 scans 12 ± 2 nm

(f)

7 μN x 4 scans 16 ± 2 nm

100 nm

Side review

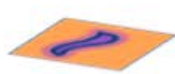
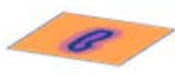


(g)

Region I

Region II

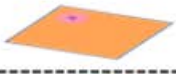
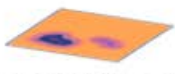
Region III

Surface
(1 μN)

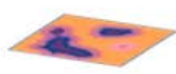
As-grown: Labyrinth

Surface
(4 μN)

Strong Force: bubbles

depth: 4 ± 2 nm
(7 μN x1)Surface above STO spacer:
Smaller bubblesdepth: 8 ± 2 nm
(7 μN x2)Surface approach or below
STO spacer:

Short Labyrinthine + bubbles

depth: 12 ± 2 nm
(7 μN x2)depth: 16 ± 2 nm
(7 μN x2)Deeper surface:
Smaller bubbles

PZT

STO

PZT

LSMO