AIP | Applied Physics Letters

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Citation: Applied Physics Letters **83**, 2028 (2003); doi: 10.1063/1.1606100
View online: http://dx.doi.org/10.1063/1.1606100
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/83/10?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 10 8 SEPTEMBER 2003

Surface deformation and ferroelectric domain switching induced by a force microscope tip on a La-modified $PbTiO_3$ thin film

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(Received 16 December 2002; accepted 1 July 2003)

Surface deformation of a ferroelectric (111)-oriented thin film of La-modified $PbTiO_3$ is induced by contact with the tip of a scanning force microscope (SFM). The deformation is accompanied by switching of the out-of-plane polarization of ferroelectric domains revealed by simultaneous piezoresponse force microscopy. The effect shows up in topographic SFM images as strokes in the fast scan direction due to surface deformation occurring below the scanning tip, and is critically dependent on the contact force for which a threshold value is deduced that allows proper SFM characterization of such thin films. At higher force, SFM might be used as a nanoscale tool for investigating fundamental properties like phase transitions under applied stress in such systems. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1606100]

Ferroelectric thin films such as lead zirconate titanate (PZT) and lanthanum-modified lead titanate (PLT) attract particular interest in view of their potential application for nonvolatile memories based on their polarization reversal characteristics[1] as well as sensors and actuators in microelectromechanical systems (MEMS) that utilize their high piezoelectric activity.[2,3] Voltage-modulated scanning force microscopy (SFM), frequently termed piezoresponse force microscopy (PFM), which relies on the electromechanical response to an external ac voltage, has emerged as a powerful technique for noninvasive domain imaging and polarization dynamics study in ferroelectric thin films.[4–6] PFM with resonance contrast enhancement[7] has been exploited to determine the fraction of (100), (001) and (00-1) domains in PLT thin films and to assess local piezoelectric behavior.[8] In this letter, we report the results of a SFM/PFM study on a highly oriented (111) PLT thin film that reveals pronounced deformation of the surface topography along with ferroelectric domain switching induced by the SFM tip.

$Pb_{1-3x/2}La_xTiO_3$ thin films with $x=0.2$ and 350 nm thickness have been deposited by pulsed-laser deposition on $(111)Pt/TiO_2/SiO_2/(100)Si$ substrates following procedures previously described.[8,9] The PLT thin films show (111) preferential orientation, 96.2% vs 3.8% of (001) according to x-ray 2θ scan analysis, and a columnar microstructure revealed by scanning electron microscopy (SEM). SFM measurements were performed for two different setups: a commercial SFM, with no electrical polarization of the tip, in both contact and tapping mode, and a homemade SFM, adapted to implement voltage-modulated measurements.[10] Conducting n^+-doped silicon cantilevers with a force constant $k=0.17$ N/m and 10 nm tip radius have been used.

Typical topography maps obtained on the (111)-oriented PLT film, with no voltage applied to the tip–sample junction, are shown in Fig. 1. The images show an average grain size of about 85 nm, with average r.m.s. surface roughness of 10 nm. The image sequence was obtained by progressively increasing and decreasing the contact force. Horizontal strokes with varying lengths, ranging from 100 to 200 nm can be clearly evinced in the images acquired with the highest con-

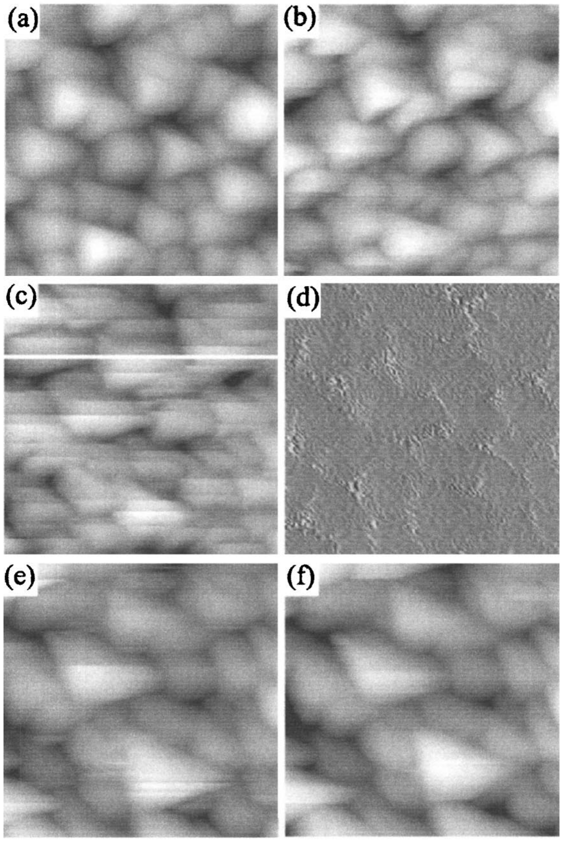

FIG. 1. Series of topographical images of the (111) PLT thin film taken successively when increasing and decreasing the loading force: (a) 1; (b) 5; (c) 25 nN; (d) error signal taken simultaneously to (c); and (e) 0.5 and (f) 0 nN. Scan size 400×400 nm^2.

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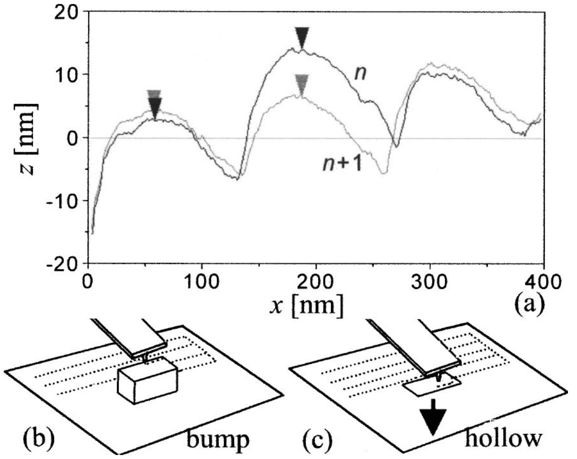

FIG. 2. (a) Line profile made at the position indicated by the white horizontal line in Fig. 1(c) of two subsequent scan lines. (b) Representation of a bump on the sample surface, with the SFM tip scanning it. (c) Bump turning into a hollow, while the tip goes on scanning.

tact force. Line profiles of two subsequent horizontal lines [Fig. 2(a)], performed at the position indicated in Fig. 1(c), show the presence of regions with varying heights besides regions with unchanged height. These discontinuities indicate that the sample topography undergoes deformation during the time that elapses from one scan line to the next. The deformation probability increases with the loading force as shown in the sequence of Fig. 1. Repeated scans of the same surface area show almost exactly the same features, excluding the occurrence of material dragging by the tip, which usually happens at random positions. Figure 1(d) shows the error signal measured simultaneously to the image of Fig. 1(c). The constant error signal demonstrates that strokes are not due to feedback instabilities. Rather, the way to read such images is the following. Let us suppose our sample is composed of a single bump [Fig. 2(b)] on a flat surface. The SFM tip is scanned onto the sample while exerting constant force. At some time during the nth scan line, the bump changes its shape or lateral position, for instance, by turning into a hollow like in Fig. 2(c). The trajectory of the tip during the $(n+1)$th scan line will follow a different path with respect to the previous one by imaging the new surface structure. The final appearance of the bump in gray tones will seem to be interrupted after the nth scan line, while the profiles of the nth and $(n+1)$th scan lines will appear coincident, outside the limited region that was concerned by deformation and/or displacement.

Superimposition of an ac with dc voltage $V(t)=V_{dc}+V_{ac}\cos(\omega t)$ between the tip and sample allows one to record the out-of-plane polarization component of the ferroelectric surface by detecting the normal cantilever vibration at frequency ω due to the local piezoresponse.[7] Figure 3 shows examples of topographic scans and the simultaneously acquired PFM images that show predominant two-level contrast corresponding to the antiparallel out-of-plane polarization components that prevail in small areas for highly oriented (111) PLT film. It is evident that morphological switching [strokes in Fig. 3(a)] involves significant modification of the PFM signal [Fig. 3(b)]. In several cases, topographical instabilities coincide with reversal of the out-of-plane polarization that may occur even for a single grain, but most importantly may propagate to adjacent grains, as is evident, for instance, in the bottom part of Fig. 3(b). These images were taken using a low resolution SFM scanner, thereby the poorer image resolution with respect to that in Fig. 1. Contact force of 250 nN was used here, probably causing the observed multiple switching. PFM contrast has been shown to depend strongly on the sample mechanical properties and imaging conditions.[7,11] Relying on the use of ''soft'' cantilevers ($k=0.17$ N/m), the tip loads were always below the PLT indentation limit (1–2 GPa),[12] in the so-called weak indentation limit of PFM,[11] where electrostatic contributions to the contrast mechanism opposite in sign to that of the piezoelectric effect[13] can be predicted. However, the high resolution of PFM images at the domain borders [Figs. 3(b), 3(d) and 3(f)] indicates the dominant contribution to be the piezoelectric one, since in that case resolution is limited by the tip radius (see, for instance, Ref. 11), while for electrostatic interaction, with long-range nature, the resolution expected is worse. For the (111)-oriented film, modulation techniques that exploit lateral force might be used to determine the in-plane polarization component and clarify the domain switching process.[5,6]

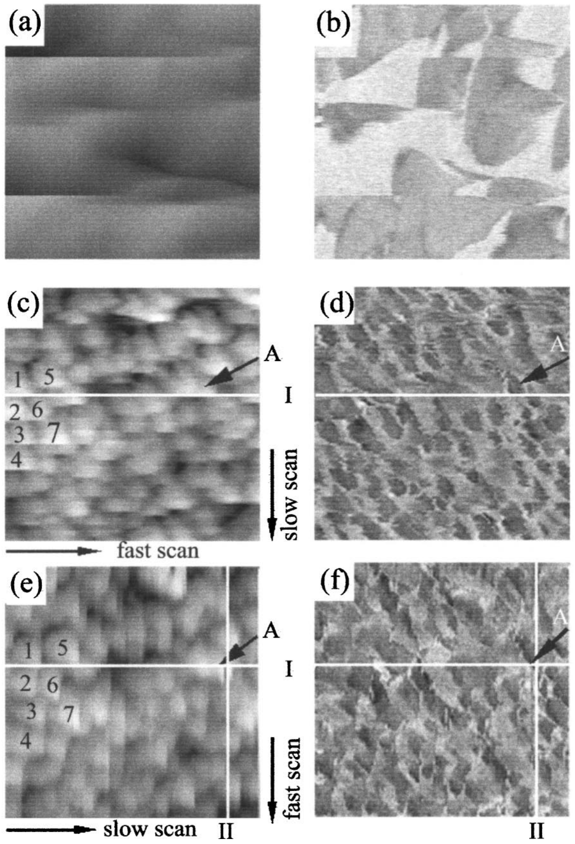

FIG. 3. SFM and PFM scans on the (111) PLT thin film showing the switching of morphology and polarization. (a) Topography and (b) PFM image of a 460×460 nm^2 region with $V_{dc}=0$ and $V_{ac}=2$ V$_{rms}$. (c) Topography and (d) PFM image of a 920×920 nm^2 region with $V_{dc}=0$ and $V_{ac}=2.8$ V$_{rms}$; scan rate 30 points/s. (e) Topography and (f) PFM image with slow and fast scan directions reverted.

The dependence on the scanning direction has been further studied in order to exclude artifacts due to tip shape effects, which are geometrical and should not depend on the way the scan is performed. Figure 3(c) shows morphological changes that involve only one or two grains, since the contact force was reduced to 35 nN and a higher resolution scanner was used. The corresponding polarization switching is visible in Fig. 3(d). The same area has been scanned by reversing the slow and fast scan directions [Fig. 3(e)], where a number of features can be recognized. The same correspondence can be pointed out between the PFM images [Figs. 3(d) and 3(f)]. It is evident how the shape of the grains appears completely different upon changing the scan direction. As an example, the grain denoted by ''A'' in Fig. 3(c) switches from a ''dark'' to a ''bright'' polarization state with respect to reference position ''I'' at the top of grain 6, whereas the same grain A in the orthogonal scan [Fig. 3(e)] extends well below the same reference point, and switches to the bright state at position ''II.'' Then, it looks like grain A has moved ''upward'' during the scan in Fig. 3(c). By crossing reference I and II, one could infer the location of a sensible site for domain switching by stress.

Further verification of the observed effect was provided by scans carried out by changing the imaging mode as well as the tip material. Tapping mode scans performed with the commercial SFM did not succeed in inducing switching. This was expected due to the lower average force exerted in intermittent contact. In general, it was observed that for very low force the effect is suppressed; after a certain threshold, single-grain switching events were recorded, with ''jumps'' of 4–7 nm. Multiple switching events, where higher jumps with lateral spreading of grain switching up to 500–1000 nm extension [visible in Fig. 3(a)], were achieved only by the homemade SFM, probably due to the higher contact force used and by the noise level of the low-resolution scanning system that likely led to higher force fluctuations. Measurements conducted with insulating Si_3N_4 tips as well as with highly conductive gold-coated tips have confirmed the occurrence of the effect, although with higher force threshold and lower switching probability. Influence of PFM parameters V_{dc}, V_{ac} and ω has been observed, suggesting a possible role of electric fields which, however, is beyond the scope of this letter.

It has been suggested[4,14] that the mechanical stress exerted by the SFM tip on the film surface, which for typical loading forces of 1–10 nN and contact area of $\sim$100 nm^2 translates into stress in the range of 10–100 MPa, may cause domain reorientation, while the shear deformation of the grain may be transferred to adjacent grains and facilitate polarization reversal in neighboring film regions. Recent studies have also shown that interfacial stress between the film and substrate is capable of inducing phase separation with reverse out-of-plane polarization within single micron-size ferroelectric capacitors made of (111) PZT films.[15] Surface deformations due to domain switching during application of uniform compressive stress up to 35 MPa have been recently reported for $BaTiO_3$ single crystals.[16] Accordingly, it can be suggested that both morphological deformation and polarization switching are mainly due to the compressive stress exerted by the tip on the PLT film. The columnar microstructure of the (111)-oriented film could promote collective switching, since each column is formed by an ensemble of coherent grains, which likely belong to the same domain type. The coherent grain boundaries would then serve as the appropriate medium for stress-induced switching propagation. On the other hand, for (111)-orientation, the stress-induced 90° domain switching does not produce additional in-plane and out-of-plane lattice cell deformation. Therefore, the morphological deformation observed here cannot originate from 90° domain switching, but most likely is due combined effect of the strain of a number of adjacent grains under stress. This effect could be much less effective for thinner PLT films strongly clamped to the substrate in which no instabilities have been observed.[8]

In conclusion, we have observed the effect of domain switching in a PLT thin film induced by the presence of the SFM tip that results in surface deformation in the vicinity of the tip. Polarization switching of the domains comprising the deformed region is revealed by piezoresponse force microscopy. This effect demonstrates the possibility of using SFM as a nanoscale tool for imaging phase transitions under mechanical stress. If loading forces are higher than a threshold value, thin film characterization becomes invasive. This rather macroscopic phenomenon caused by relatively small stress looks promising for low-power-consumption microtransducers in piezoelectric-based MEMS applications.

Note added in proof. After submission of this letter, we became aware of a PFM study on sol–gel PLT (8%) films, where the piezoelectric response was found to be reversed or even suppressed at tip loads in the range of 5–20 μN.[17] There, high internal strain that develops due to the sol–gel route, as well as contact area enlargement likely to occur with SFM tips at such force levels, might increase the force regime for stress-induced switching.

The authors thank the EC (Contract ERBFMRXCT98-0242) and CAM (Spain) (Contract 07N/2004/2001) for financial support, O. Kappertz and M. Wuttig for kindly providing the pictures of Fig. 1, and G. Tarabella and E. Andreoni for valuable assistance.

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