

## Detection mechanism for ferroelectric domain boundaries with lateral force microscopy

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The contrast mechanism for the visualization of ferroelectric domain boundaries with lateral force microscopy is generally assumed to be caused by mechanical deformation of the sample due to the converse piezoelectric effect. We show, however, that electrostatic interactions between the charged tip and the electric fields arising from the surface polarization charges dominate the contrast mechanism. This explanation is sustained by quantitative analysis of the measured forces as well as by comparative measurements on lithium niobate and potassium titanyl phosphate. © 2006 American Institute of Physics. [DOI: 10.1063/1.2234303]

Lateral force microscopy (LFM) is usually utilized for the detection of friction forces between tip and sample while scanning the surface. LFM can therefore map topographical steps (roughness) but also different material compositions, whereby the contrast depends strongly on the scanning parameters (velocity, direction, contact pressure, etc.).<sup>1</sup>

In first LFM experiments on ferroelectric samples, investigating guanidinium aluminum sulfate hexahydrate (GASH), such dependencies were observed, indicating that the domain contrast was due to topographical features as these materials exhibit domain selective, hygroscopic properties.<sup>2,3</sup> Domains or domain boundaries have also been revealed by LFM in other crystals such as lithium niobate ( $\text{LiNbO}_3$ ),<sup>4,5</sup> potassium titanyl phosphate (KTP),<sup>4,6</sup> barium titanate,<sup>7</sup> and lead titanate zirconate,<sup>8,9</sup> using the very same experimental setup as for piezoresponse force microscopy (PFM).<sup>10</sup> In brief, an alternating voltage (amplitude  $U_{ac}$  and frequency  $f_{ac}$ ) is applied to the tip which leads to deformations of the sample due to the converse piezoelectric effect. The resulting vibrations of the surface cause oscillations of the cantilever which can be read out with a lock-in amplifier. For LFM, instead of the bending, the torsion of the cantilever is detected. In the case of antiparallel 180° domains, the alternating voltage leads to tilting vibrations of the surface at the domain boundaries. This tilting is supposed to cause lateral forces acting on the tip which result in torsional oscillations of the cantilever.<sup>4,5</sup> Our investigations show, however, that these torsional oscillations are dominated by the electrostatic interaction between the charged tip and the electric fields arising from the surface polarization charges. We therefore name this detection technique lateral electrostatic force microscopy (LEFM).

For the experiments we used a commercial scanning force microscope (Smena, NT-MDT), modified to allow application of voltages to the tip. The ac-voltage driven bending and torsion of the cantilever were recorded simultaneously as vertical and lateral responses, respectively. To obtain accurate experimental data, the alignment of the four-segmented photodetector with respect to the cantilever is crucial. In case of misalignment, a pure bending of the cantilever inevitably pretends a (nonexisting) torsion and vice versa. The magnitude of this cross-talk can be estimated

when exciting the cantilever in air at its resonance frequency and comparing the vertical with the lateral response: We measured cross-talks of typically 5%–20% why we developed a cross-talk compensator which electronically suppresses the cross-talk by a factor of 100. Therefore we add a 180° phase-shifted correction signal of adjustable amplitude to the error signal. Note that for every cantilever, the compensation has to be adjusted anew.

The LEFM setup was calibrated with a sample consisting of a glass plate with chrome electrodes which allow to generate electric fields parallel to the sample surface (Fig. 1). The chrome mask had a thickness of 250 nm and a slit width of  $w=20\ \mu\text{m}$ . Applying a voltage of  $U_{dc}=20\ \text{V}$  to the electrodes generates an electric field of  $E\approx U_{dc}/w=10^6\ \text{V/m}$  within the slit which at its center is mainly parallel to the surface. The tip ( $U_{ac}=10\ \text{V}_{pp}$  and  $f_{ac}\sim 33\ \text{kHz}$ ) senses this electric field  $E$  and performs oscillations which can be read out with a lock-in amplifier. The positioning of the tip was found to be uncritical along a section of at least  $10\ \mu\text{m}$  width in the middle of the slit. We have found a linear relationship between the magnitude of the lateral force and  $U_{ac}$  or  $U_{dc}$ , respectively, as expected from the theory.<sup>11</sup> This setup allows to determine the phase of the oscillations with respect to the direction of the electric field. Furthermore, the influence of friction between tip and sample surface on the oscillation amplitude can be measured. Altering the load up to  $30\ \mu\text{N}$  had no influence on the signal; however, when retracting the tip just above the surface an enlargement by a factor of 1.5 was measured.

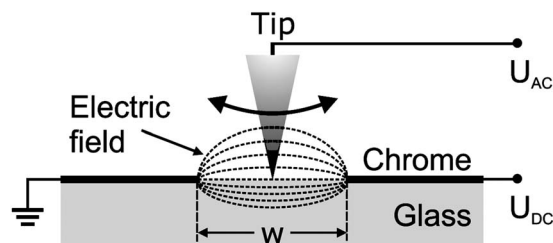


FIG. 1. Setup for LEFM calibration. A chrome mask with a slit of  $w=20\ \mu\text{m}$  width allows to generate electric fields parallel to the surface by applying a voltage  $U_{dc}$  to the electrodes. Simultaneously an alternating voltage  $U_{ac}$  is applied to the tip. The amplitude and phase of the enforced oscillations of the tip can be read out with a lock-in amplifier.

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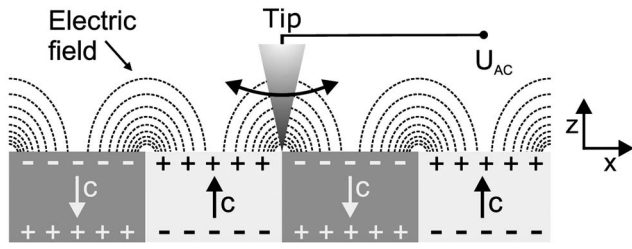


FIG. 2. Schematic drawing of the static electric fields above a periodically poled ferroelectric crystal. Here  $c$  denotes the polar axis and  $U_{ac}$  the alternating voltage applied to the tip.

In LFM the orientation of the cantilever with respect to the lateral force  $F_l$  acting on the tip is crucial. If  $F_l$  is perpendicular to the cantilever, the latter will be forced to twist which can be read out as a torsion signal. If, on the contrary,  $F_l$  is orientated parallel to the cantilever, this will lead to a buckling, which can be detected as a deflection signal. The LFM signals were calibrated taking into account the appropriate force constants of the cantilever and assuming that the photodetector has the same sensitivity for vertical as for lateral signals. Be aware that for PFM images the contrast information is height (in meters) whereas for LFM images it is force (in newtons). The dimensions of the cantilevers used for the measurements are length  $l=130\ \mu\text{m}$ , width  $w=35\ \mu\text{m}$ , thickness  $t=1.9\ \mu\text{m}$ , and tip height  $h=17\ \mu\text{m}$ . Therefore the lateral spring constants are  $k_t=(Gwt^3)/(3lh^2)\approx 106\ \text{N/m}$  for torsion and  $k_b=(Ewt^3)/(12lh^2)\approx 91\ \text{N/m}$  for buckling [shear modulus  $G=0.5\times 10^{11}\ \text{N/m}^2$  and Young's elasticity modulus  $E=1.7\times 10^{11}\ \text{N/m}^2$  for Si (Ref. 12)]. For the sake of completeness we give the deflection spring constant which is  $k_d=(Ewt^3)/(4l^3)\approx 4.6\ \text{N/m}$ .

We investigated a  $z$ -cut periodically poled lithium niobate (PPLN) crystal with a period length of  $8\ \mu\text{m}$  and a thickness of  $0.5\ \text{mm}$ . The appropriate piezoelectric coefficient is known to be  $d_{33}=7.6\ \text{pm/V}$  (Ref. 13) and the (uncompensated) surface polarization charge density is  $\sigma=0.7\ \text{C/m}^2$ .<sup>14</sup> Figure 2 shows a sidewise sketch of a PPLN crystal. Because of the surface polarization charges, electric fields build up whose strength parallel to the surface is maximum at the domain boundaries. The electric field  $E_x(x,z)$ , with  $x$  being the axis parallel to the surface and perpendicular to the domain boundaries and  $z$  denoting the distance from the sample surface (Fig. 2) for an infinite PPLN structure, is given by

$$E_x(x,z) = \frac{\sigma}{4\pi\epsilon_0} \ln \left\{ \prod_{n=-\infty}^{\infty} \frac{[(x+2na)^2+z^2]^2}{[(x+2na+a)^2+z^2]^2} \right\},$$

with  $a$  denoting the domain size (PPLN period of  $2a$ ) and  $n$  the number of domains being included. For the PPLN sample electric field strengths of  $10^{11}\ \text{V/m}$  are theoretically expected if no compensation of the surface charges is assumed.

In Fig. 3 the experimental results for deflection (a) and torsion (b) images of the end of a poled stripe of PPLN are shown, with the corresponding scan lines in (c). The orientation of the cantilever was chosen to be parallel to the stripe. At first sight it is obvious that the deflection image (a) shows the domain faces (due to the converse piezoelectric effect), whereas the torsion image (b) only shows the domain boundaries, at the left edge as a dark stripe and at the right edge as a bright stripe. The contrast inversion is due to the change of

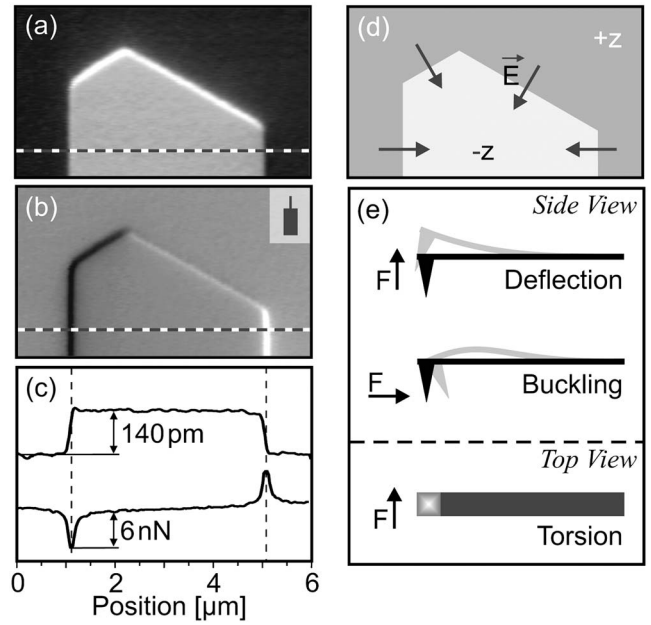


FIG. 3. Deflection (a) and torsion (b) images of the in-phase lock-in output simultaneously recorded on a  $\text{LiNbO}_3$  crystal (image size of  $6\times 3.5\ \mu\text{m}^2$ ) with the corresponding scan lines (c) with  $10\ \text{V}_{pp}$  applied to the tip. The orientation of the chip with the cantilever is shown as inset in (b). Schematic drawing (d) of the electric field distribution  $E$ . In (e) the possible movements of the cantilever are depicted. The deflection image (a) shows deflection (PFM) and buckling (LEFM) and the torsion image (b) the twisting of the cantilever (LEFM).

the direction of the electric field [Fig. 3(d)]. This is consistent with the results we obtained with the test sample from Fig. 1, where we determined the sign of the output of the lock-in amplifier with respect to the direction of the electric field. The contrast is reduced when the electric field vector perpendicular to the cantilever becomes smaller as can be seen on the tilted edges of the domain. The torsion signal on both domain faces is identical, which can be observed only when the cross-talk between vertical and lateral signals is suppressed. Looking more closely at Fig. 3(a), at the top edges of the domain a bright stripe is visible. When comparing with the schematic drawing of the electric field configuration in Fig. 3(d), at these edges the electric field has a component along the axis of the cantilever. This also leads to lateral forces acting on the tip which result in a buckling of the cantilever, adding to the observed vertical signal.

To summarize the experimental results of Fig. 3, the deflection image (a) shows the vertical movement of the cantilever due to the converse piezoelectric effect and at the top edges the electrostatic interaction of the periodically charged tip with the electric field which leads to a buckling of the cantilever [upper part of Fig. 3(e)]. The torsion image (b) only shows the electric field component perpendicular to the axis of the cantilever [lower part of Fig. 3(e)].

To support our explanation that lateral forces measured at the domain boundaries are due to electrostatic interactions of the periodically charged tip with the electric field arising from the surface polarization charges, we analyzed the data quantitatively. Assuming a tip radius of  $r=30\ \text{nm}$  and a voltage of  $U=10\ \text{V}_{pp}$  applied to it results in a charge of  $Q=U4\pi\epsilon_0r\approx 10^{-17}\ \text{C}$ . In an external electric field  $E=10^{11}\ \text{V/m}$  this leads to a force  $F=QE\approx 10^{-6}\ \text{N}$ . As the strength of the electric field  $E$  is directly proportional to the surface charge density  $\sigma$ , comparing this result with the mea-

sured value of  $F=6$  nN underlines that the surface charge is reduced by three orders of magnitude by compensation charge which agrees with other publications.<sup>15,16</sup>

For comparison, we estimated the lateral forces expected from the tilting of the surface due to the converse piezoelectric effect. From the tip size and the lateral resolution in PFM measurements, one can assume that the tilt of the surface occurs in a region of about  $\Delta x=100$  nm diameter. The maximum vertical surface displacement for  $\text{LiNbO}_3$  is  $\Delta h=76$  pm with  $U_{ac}=10$  V<sub>pp</sub> applied to the tip. The surface across the domain boundary can then be described as a inclined plane with an angle of  $\alpha=0.04^\circ$ . A vertical force  $F_v$  acting on this inclined plane can be vectorially devised in two components: one normal to the slanted surface ( $F_n$ ) and the other one parallel to it ( $F_l$ ) causing a torsion of the cantilever. From simple geometrical considerations, it is evident that  $\Delta h/\Delta x=F_l/F_n \approx F_l/F_v$  because of  $\alpha$  being very small. When operating a scanning force microscope in contact mode, the typical load of the tip acting on the surface is  $F_v \approx 10$  nN (Ref. 1) and thus the expected lateral forces are  $F_l \approx 0.008$  nN. This is smaller by three orders of magnitude than the values measured, e.g., in Fig. 3(b). Therefore the mechanical contribution to the lateral forces seems to be negligible. Furthermore, we have not observed any dependence of the lateral forces on the load of the tip, as it is required for this contrast mechanism.

Finally we carried out comparative LFM measurements on  $z$ -cut periodically poled potassium titanyl phosphate (KTP) crystals that also exhibit  $180^\circ$  domains only. The measured piezoelectric coefficient of  $d_{33}=6$  pm/V is lower than the reported values of  $10\text{--}26$  pm/V (Ref. 17) which can be attributed to the inhomogeneous electrical field at the tip causing clamping and thus a reduced piezoelectric deformation.<sup>18</sup> However, the ratio between lateral and vertical signals should be constant if both are originated by the converse piezoelectric effect. Of course this is valid only if the electromechanical response at the domain boundary is the same, i.e., the tilting takes place at equal length scales. We observed identical length scales for the electromechanical response on both  $\text{LiNbO}_3$  and KTP using the same tip. Thereby, we measured a 2.6 times smaller ratio between lateral and vertical signals for KTP. This agrees more likely with an electrostatic origin for the lateral forces as the sur-

face polarization charge density for KTP (Ref. 19) is 3.5 times smaller than for  $\text{LiNbO}_3$ .

In conclusion, we have shown that the origin of the contrast mechanism for detection of ferroelectric domain boundaries in lateral force microscopy is dominated by the electrostatic interaction of the charged tip with the electric field arising from the surface polarization charges. A quantitative estimate as well as comparative measurements on  $\text{LiNbO}_3$  and KTP crystals sustain this explanation.

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<sup>11</sup>Lateral force  $F_l=QE_x$ , charge  $Q \sim U_{ac}$ , and (in case of chrome mask) electric field  $E_x \sim U_{dc}$ .

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