Injection charge assisted polarization reversal in ferroelectric thin films

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The authors have investigated the polarization reversal on ferroelectric thin films caused by a grounded tip on 50-nm-thick Pb(Zr,Ti)O\textsubscript{3} films. Backswitching occurred when the grounded tip recontacted a “freshly” switched area. It is believed that the upper part of the film switches back due to the field between the grounded tip and previously injected charges. During dynamic operation, partial backswitching was observed during pulsed writing using pulse widths of 1 ms. The results show that polarization reversal is an issue, which has to be addressed in the writing scheme of future probe-based storage devices. © 2007 American Institute of Physics. [DOI: 10.1063/1.2679902]

The increasing demand for high density memory devices has triggered a significant interest for the use of an atomic force microscope to read/write nanoscopic domains in a ferroelectric media.\textsuperscript{1,2} Among them, many researchers have investigated the piezoresponse of the ferroelectric domain using piezoelectric force microscopy (PFM).\textsuperscript{3-6}

Recently, a peculiar polarization reversal phenomenon\textsuperscript{6-10} has been reported, where switching seemed to happen “against the field.” Bühlmann \textit{et al.}\textsuperscript{9} found that this phenomenon occurred when a grounded tip was recontacted on an area, which had just been switched previously. It is believed that charges are injected into the film during the switching process. When recontacting that area with the grounded tip, the field between the injected charges and the upper part of the film switches back due to the field between the grounded tip and previously injected charges. During dynamic operation, partial backswitching was observed during pulsed writing using pulse widths of 1 ms. The results show that polarization reversal is an issue, which has to be addressed in the writing scheme of future probe-based storage devices.

The 3×3 μm\textsuperscript{2} PFM image in Fig. 2 has been obtained as follows: First, a large area of 5×5 μm\textsuperscript{2} was poled downwards at +5 V. This back poling was done at 1 Hz and the scanning was done from top to bottom and lasted for 4 min. Then, the tip was moved to the bottom left corner of the big box where it was grounded and then moved toward the center. On the way there, the tip traveled on an area, which was poled downwards between 0 and 2 min ago. At the center, a smaller 1×1 μm\textsuperscript{2} box was written at −5 V and 2 Hz, which took 2 min. Then the tip was moved to the top left corner of the smaller box. There it was grounded and moved to the top left corner of the larger box. On that way, the tip traveled on an area, which was poled downwards between 4 and 6 min ago.

The phase image in Fig. 2 shows the “correct” polarizations of the box patterns. Polarization reversal occurred where the grounded tip traveled on an area, which was poled less than 2 min ago. As such, the diagonal line at the bottom left was created. No corresponding line can be seen on the top left, where the grounded tip traveled on an area, which was poled at least 4 min ago. This indicates that injected charges are responsible for the phenomena, as described above. Due to the dissipation of the injected charges, there was no polarization reversal on the 4–6 min “old” back-poled area. From this finding and from the fact that the tip loading force was kept constant at 50 nN throughout all experiments, we can exclude that polarization reversal was induced by lattice deformation below the tip.\textsuperscript{12,13} The width of the backswitched line is about 30 nm. The line shows a weak piezoelectric response in the amplitude image. This may partially be due to the low resolution. Another reason is the signal coming from the lower part of the film, which is down...
polarized, and therefore counteracts the signal from the upper polarized, upper part. At some places on the line, the phase image indicates unswitched parts. There, it is thought that the contribution of the lower part is dominating. This is an indication that the charges are injected very close to the surface there.

The polarization reversal effect was previously observed in 400-nm-thick epitaxial PZT and it was reported that it did not happen below 130-nm-thick PZT. A critical minimum thickness between 50 and 200 nm was estimated for epitaxial PZT. In single crystalline lithium tantalate thin films, a minimum thickness of 350 nm was reported. In contrast to those previous observations, we observed the effect up to a maximum of 2 min. Whereas polarization reversal by a grounded tip was observed even 6 min after charge injection in 400-nm-thick epitaxial PZT and it was reported that it did not happen below 130-nm-thick PZT. A critical minimum thickness between 50 and 200 nm was estimated for epitaxial PZT. In single crystalline lithium tantalate thin films, a minimum thickness of 350 nm was reported. In contrast to those previous observations, we observed the effect up to a maximum of 2 min. We believe that this time scaling is due to a faster disappearance of the injected charges in thinner films. In the polycrystalline films, the injected charge can easily migrate to the bottom electrode via the grain boundary which acts as a conduction path. In the previous work, more than 20 V, which was well above the coercive voltage, were necessary to inject charges in 400-nm-thick PZT films. We observe the phenomenon after poling at only 5 V, which is near the coercive tip voltage of about 2 V. This should be addressed in the writing scheme of future probe storage devices.

Results from pulsed writing can be seen in Fig. 3(a). The image was obtained as follows: First, a $5 \times 5 \mu m^2$ large area was back-poled downwards from top to bottom at a tip voltage of $+5 V$. In order to get rid of the injected charges, we waited 2 min before we started the pulsed writing process. The pulse base was 0 V (ground), the pulse width was 1 ms, and the voltage was $-20 V$. The scanned line length was 5 $\mu m$, and the line spacing was 300 nm. The scan speed was 1 Hz. At that scan speed, the tip travel distance during the pulse is equal to 10 nm. The diameter of the obtained dots was about 140 nm. In about 70% of dots, polarization reversal was observed by the appearance of a smaller, back switched area inside the dot. Figure 3(a) shows dots, which were written when the tip traveled from left to right. In the first dot of the third line, it can be seen that the backswitched area describes a dot or line inside the right side of the dot. This is explained as follows: During the application of the pulse, the dot is switched to the upward polarization and at the same time, charges are injected. Here, we suppose that the whole dot area was affected by charge injection. After the pulse, the tip is located only 10 nm to the right of the center. The now grounded tip travels rightwards on the upward polarized dot where the buried injected charges induce polarization reversal to the downward state. This leads to a backswitched dot or line, which runs from the dot-center to the right dot-border [schematically shown in Fig. 3(b)]. It is noted that not all dots showed that pattern. In the other dots, the polarization reversal was observed in the form of an isolated dot of about 30 nm in the center, indicating that the charge injection was very much localized to an area just below the tip in those cases [the second dot of fourth line in Fig. 3(a)]. However, in all cases, the backswitched areas were much smaller than the dot size. Considering the field distribution around the tip, this shows that the injected charges must be buried to the upper part of the sample. The dot poling experiments showed that charge injection from the tip is very efficient.
In summary, we observed the polarization reversal produced by a grounded tip in 50-nm-thick PZT films on Pt. In addition, the phenomenon was produced during pulsed writing, which has severe implications for the writing scheme in future probe-based storage devices.

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