

## Nanoscale Studies of Ferroelectric Phenomena in Perovskite Thin Films

### Scientific Achievement:

We investigated ferroelectric domain dynamics at the nanoscale, through evaluation of domain structure, stability, and relaxation as a function of ferroelectric layer microstructure and controlled capacitor size and geometry.

We used an AFM piezoresponse imaging method based on the detection of the local electromechanical vibration of polarized domains in a ferroelectric sample, caused by an external AC voltage applied between an atomic force microscope (AFM) tip (movable top electrode) and an electrode layer underneath the ferroelectric layer. The AC electric field with a frequency  $\omega$  causes a localized sample vibration with the same frequency due to the piezoelectric effect. Simultaneously with the AC excitation, a conventional topographic image can be obtained. The ferroelectric domain structure is visualized by monitoring the first harmonic signal (piezoresponse signal). The phase of the piezoresponse signal depends on the sign of the piezoelectric coefficient, such that regions with opposite polarization orientation, vibrating in counter phase with respect to each other, under the applied AC field, appear as regions of different contrast in the piezoresponse image.

Theory indicates that an enhanced piezoelectric response should be observed if  $90^\circ$  ferroelastic domain walls move under electric field excitation in ferroelectric materials. Considering that removal of film/substrate interaction constraints may enable phenomena not hitherto observed in ferroelectric thin films laterally constrained by substrate-induced clamping, we decided to investigate polarization dynamic phenomena in LSCO/PZT/LSCO nanocapacitors fabricated with a focused ion beam method down to  $70 \times 70$  nm structures where film lateral constraints are removed. We have found that nanostructuring of the ferroelectric layer can dramatically alter the electro-mechanical interactions between the film and the substrate thereby enabling the movement of the ferroelastic  $90^\circ$  domain walls. This movement can be facilitated by a break or large weakening in the film clamping to the substrate. Piezoresponse scanning force microscopy images clearly show that  $90^\circ$  domain walls can move. Furthermore, measurements of the  $d_{33}$  parameter for  $1 \times 1 \mu\text{m}^2$  capacitors with such unclamped ferroelectric layers reveal values of up to 250 pm/V at remanence, which is approximately 3-4 times the predicted value of 87 pm/V for a single domain ferroelectric single crystal. These results may have a major impact in the development of the next generation of high-density ferroelectric memories. We also developed an e-beam lithography technique to produce nanocapacitors and obtained piezoresponse images.

### Significance:

The work currently performed on this topic is opening new avenues of research on ferroelectric domain dynamics at the nanoscale and it will play a major role in the projected center for nanomaterials at ANL. In addition, The understanding of the nanoscale ferroelectric thin film domain dynamics shown above will have a critical impact in the design of the next generation of high density (Mbit to Gbit) non-volatile ferroelectric random access memories (NVFRAMs), which can replace current DRAM and Flash memories, creating a new multibillion dollar market.

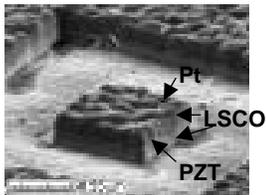
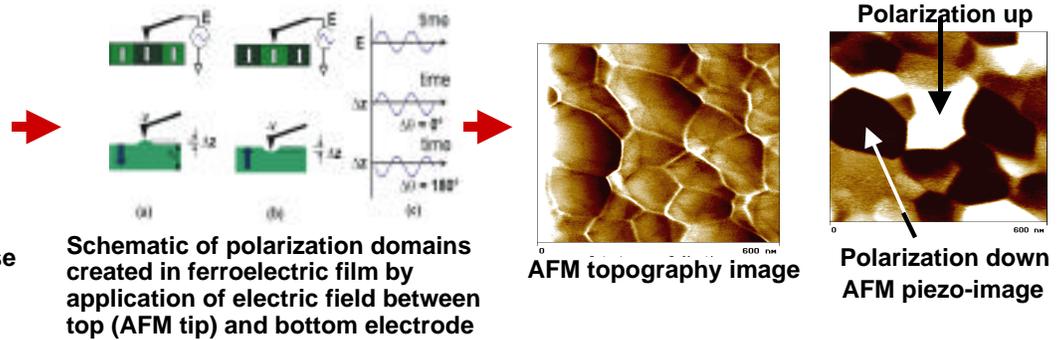
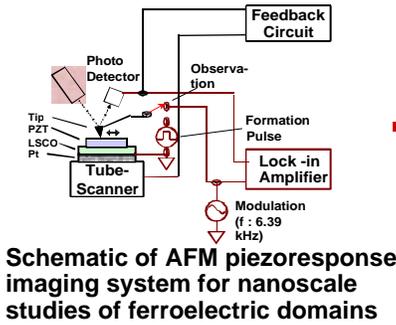
### Performers:

O. Auciello and D.J. Kim (ANL), V. Nagarajan and R. Ramesh (U. of Maryland).

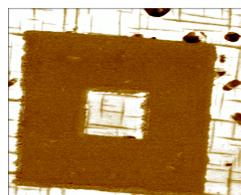
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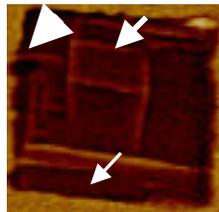
## AFM Piezoresponse Imaging of Ferroelectric Domains



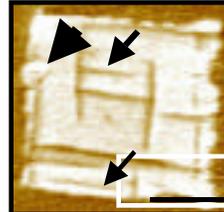
PZT nanocapacitor fabricated via FIB etching



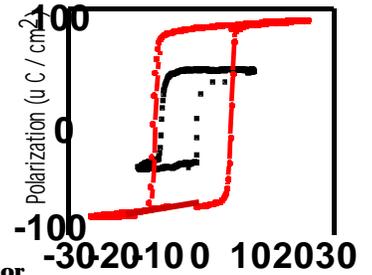
PZT nanocapacitor with polarization up showing 90° domains (lines)



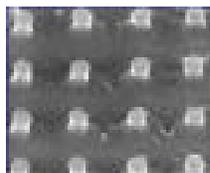
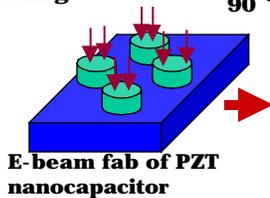
Polarization switching in PZT nanocapacitor fabricated via FIB etching



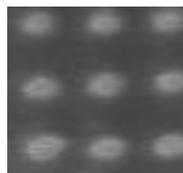
Piezoresponse image of PZT nanocapacitor array



Polarization loops for PZT nanocapacitor (—) and for constrained capacitor (—)



PZT nanocapacitor array via FIB



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