Domain Wall Interface Density Control For Tunable Thermal Conductivity

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Motivation

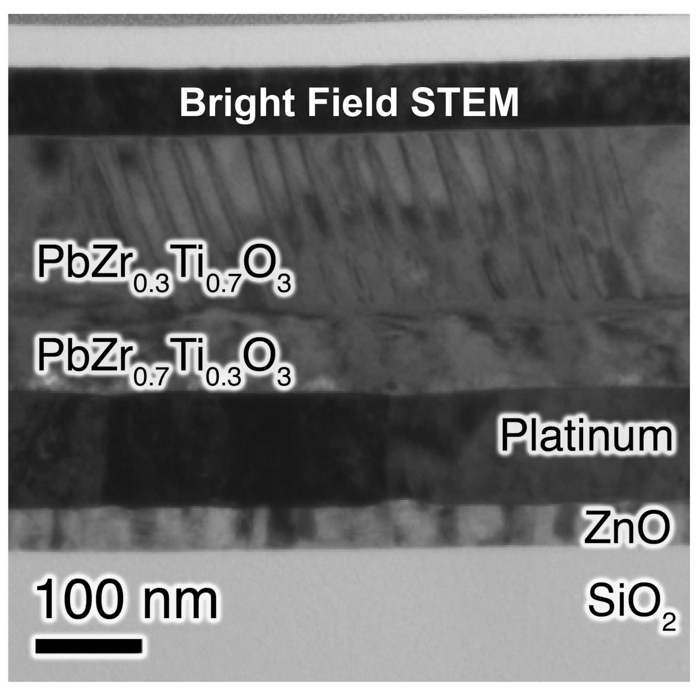
Ferroelectrics are ideal materials for ***tunable*** solid state thermal conductivity. The size and density controllable ferroelastic domain walls act as mobile coherent interfaces to scatter phonons. By modifying the domain features, one can either enhance or subdue phonon scattering, which opens a new pathway to control phonon transport. Potential applications include controlled heat transport and thermal diodes and switches.

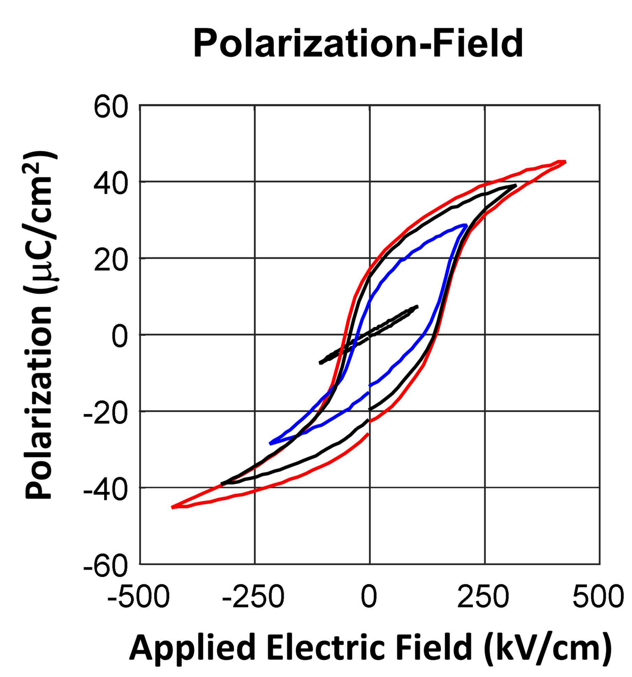
We have recently demonstrated room temperature thermal conductivity tuning of 11% with applied electric fields in bilayer lead zirconate titanate thin films.[1] The purpose of this work is to correlate the observed bias controlled thermal conductivity with the ferroelastic domain structure and density of the samples.

[1]Room-Temperature Voltage Tunable Phonon Thermal Conductivity via Reconfigurable Interfaces in Ferroelectric Thin Films, Nano Lett., 2015, 15 (3), pp 1791–1795

PZT Bilayer Films

Solution-derived polycrystalline PZT bilayers:
142 nm $PbZr_{0.3}Ti_{0.7}O_3$/60 nm $PbZr_{0.7}Ti_{0.3}O_3$/100 nm Pt/ 40 nm ZnO/SiO_2/Si

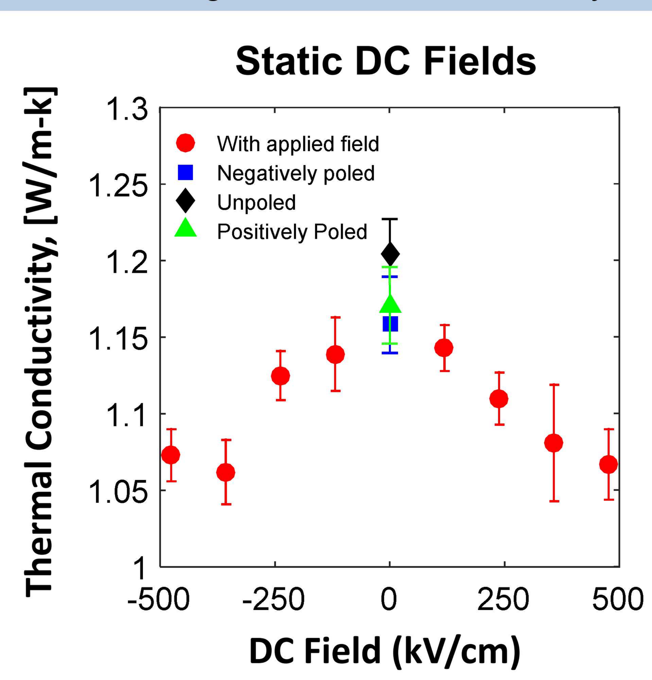


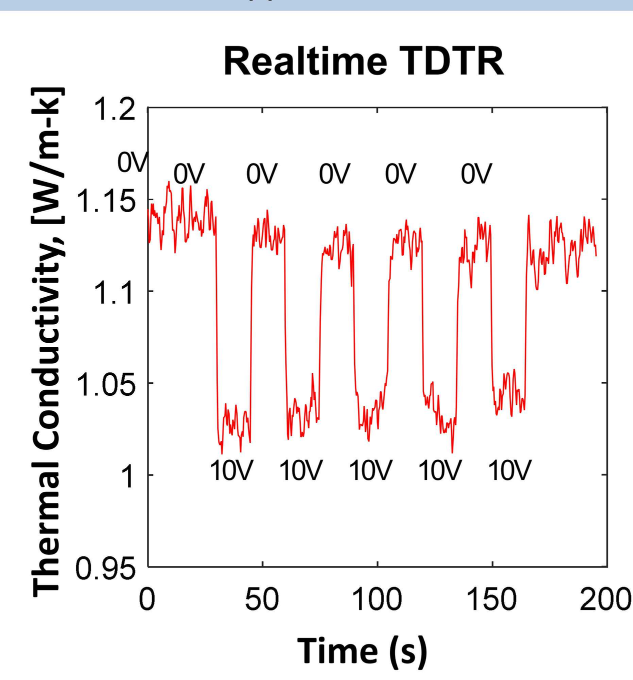


- Domain boundaries clearly visible in bright field STEM in the PZT 30/70 layer
- SAED reveals domain boundaries to be of 90° type
- No domain boundaries observed in the PZT 70/30 layer

Field Tunable Thermal Properties in PZT

Thermal measurements via Time Domain Thermoreflectance (TDTR) using 90 nm platinum top electrode transducers. Changes of thermal conductivity of 11% was achieved with the application of electric fields.





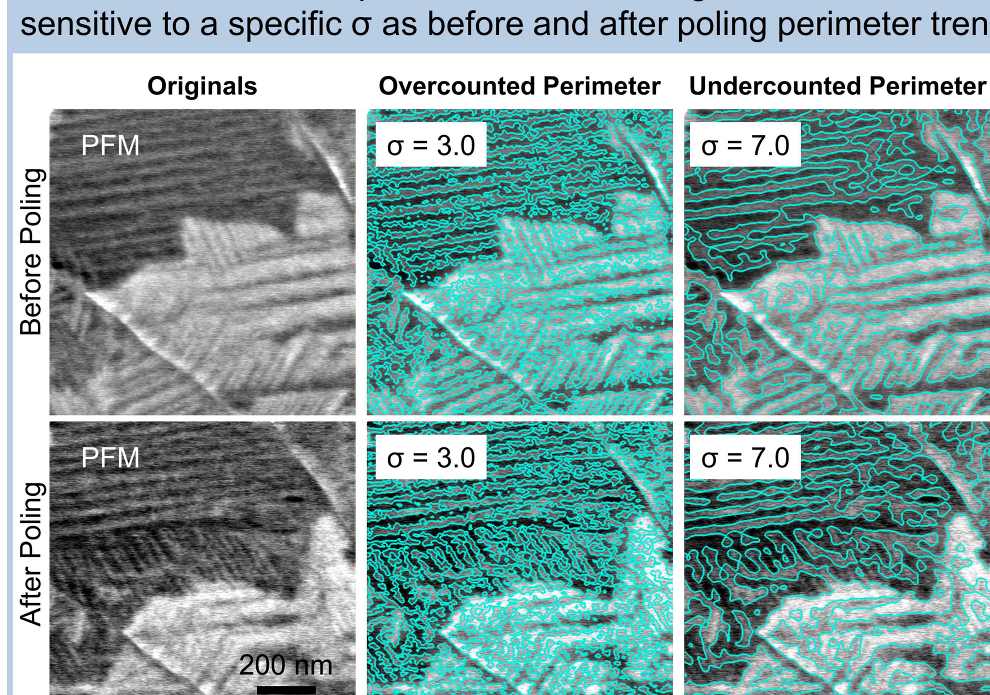
- Thermal conductivity decreases with applied electric field
- Thermal conductivity decreases after ferroelectric poling
- Tuning effect is reversible and rapid

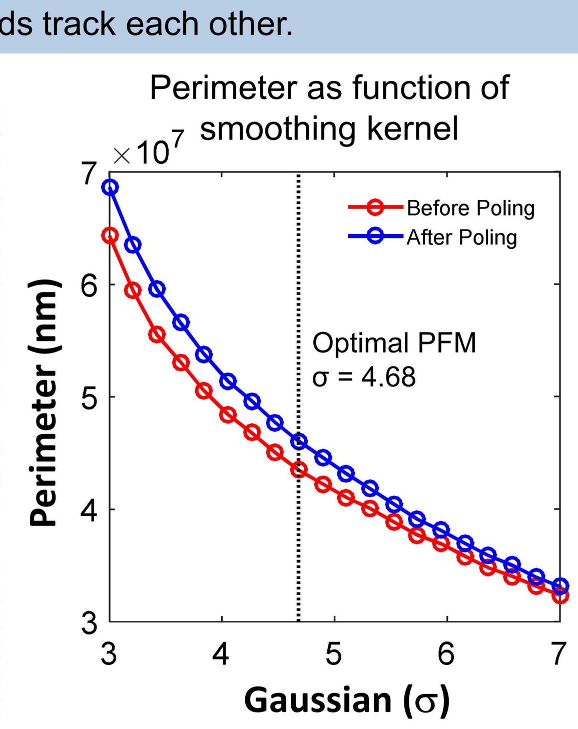
→ **Implies domain wall density increases post poling !**

Domain Perimeter Detection Algorithms

The domain perimeters of different domain orientations were found using an intensity based edge detection algorithm, the Laplacian of Gaussian (LoG) Method, implemented in Matlab. The algorithm creates closed continuous contours, and uses a single variable, σ, the standard deviation of the LoG filter that determines the amount of smoothing.

The optimal σ was chosen to best visually match the domain boundaries. For PFM, σ=4.68, and for SEM, σ=6.58, which corresponded best to recognizable domain features. The results, however, are less sensitive to a specific σ as before and after poling perimeter trends track each other.



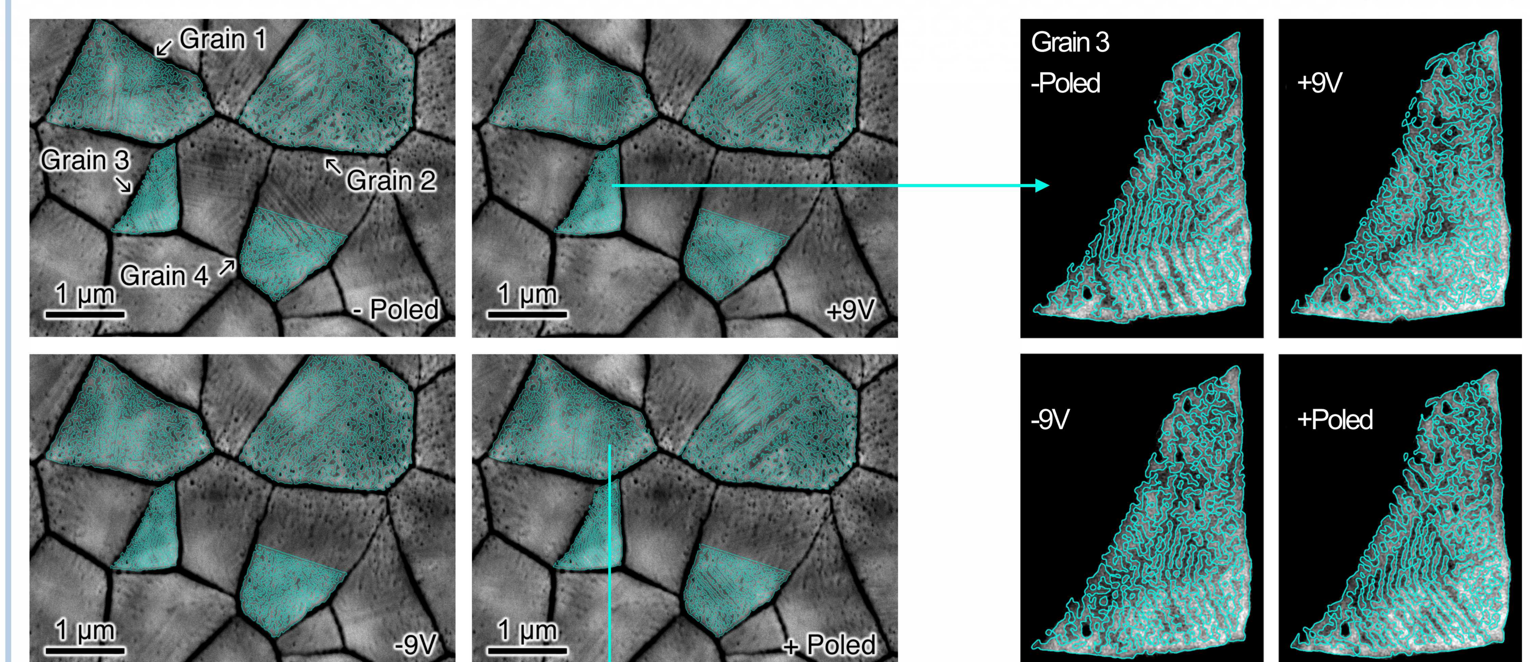


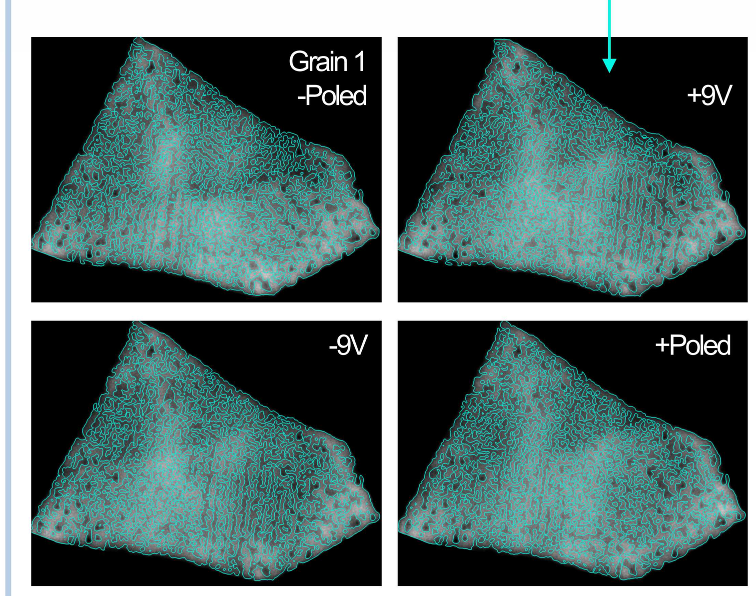
Domain Imaging Techniques

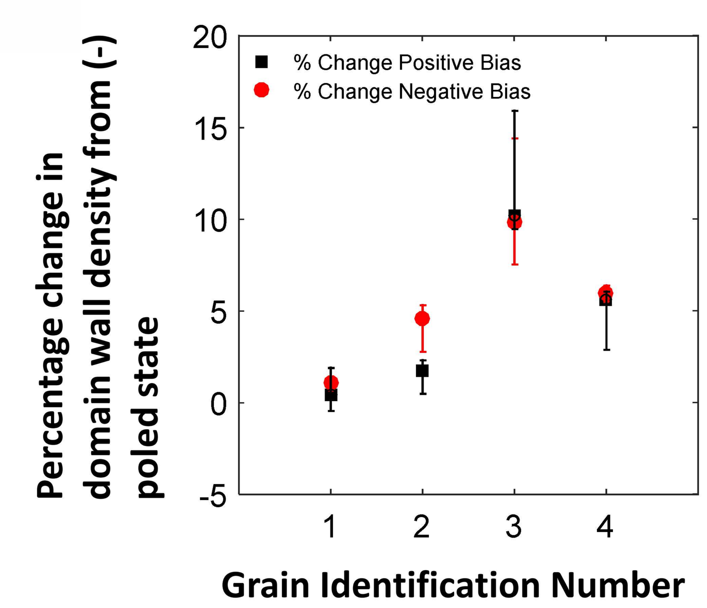
- Piezoresponse Force Microscopy (PFM) – a scanning probe method which images the local piezoelectric response of the surface, clearly indicating domain features at resolution down to ~10 nm
- In Operando Channeling Contrast Scanning Electron Microscopy (CC-SEM) - contrast of the backscattered electrons due to electron channeling that depends on the domain orientation relative to input beam

Channeling Contrast SEM

Electric fields were applied to the sample through 3 nm thick top Pt electrode. Domain contrast and clear voltage-induced domain structures changes are observed in each state. Four grains that unambiguously showed domain features were analyzed by edge identification. Changes in domain wall density of up to +10% where measured.

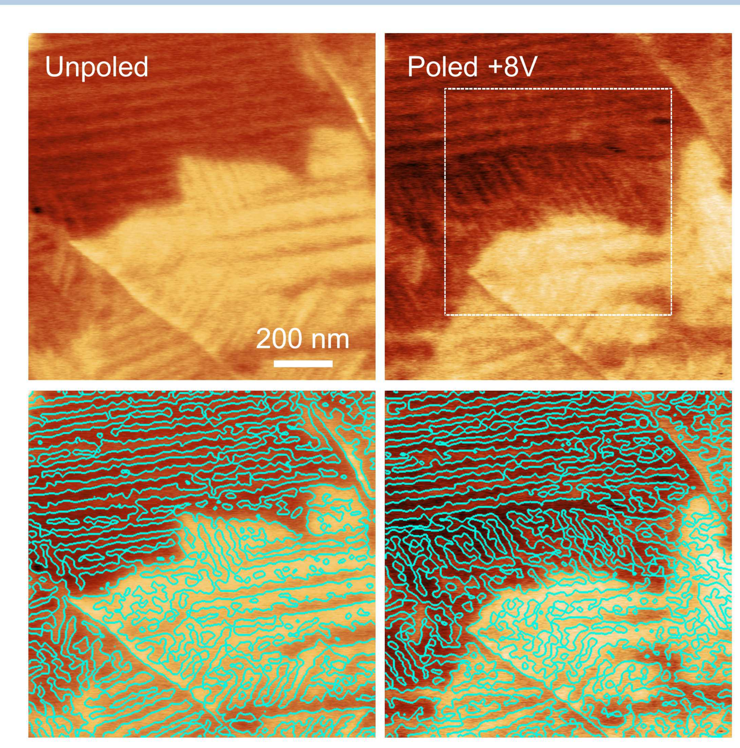


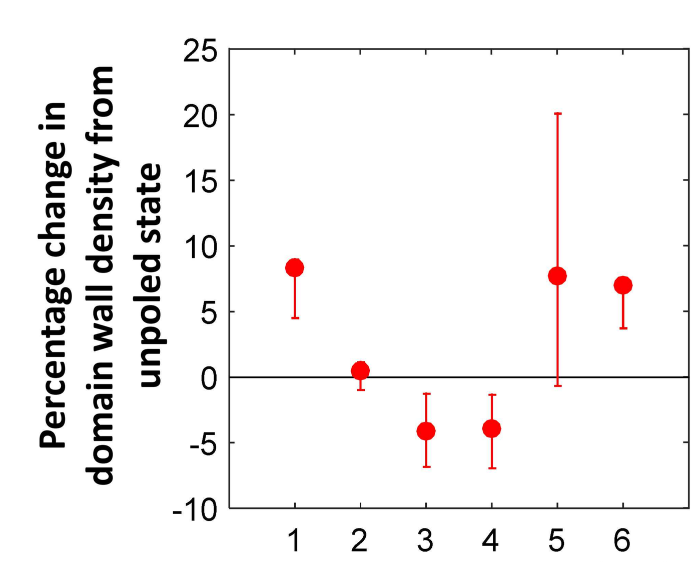




Piezoresponse Force Microscopy

Local poling experiments using the AFM tip were analyzed using the edge detection. A sub region of a larger area was poled at +8V in various locations on the film. Depending on local grain orientation and strain, regions either increase or decreased in domain wall density after poling. The example below shows an increase of 7.0%.





Results

- In aggregate, domain wall density increases after poling in PFM by 2.6%
- In operando CC-SEM reveals domain wall density increasing after poling by >2%

Conclusions

- Density of ferroelastic domains in ferroelectric thin films can affect thermal conductivity at room temperature
- Domain wall density increases with applied field in clamped PZT-based films
- Increased domain wall density results in decreased thermal conductivity due to phonon scattering at domain walls

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