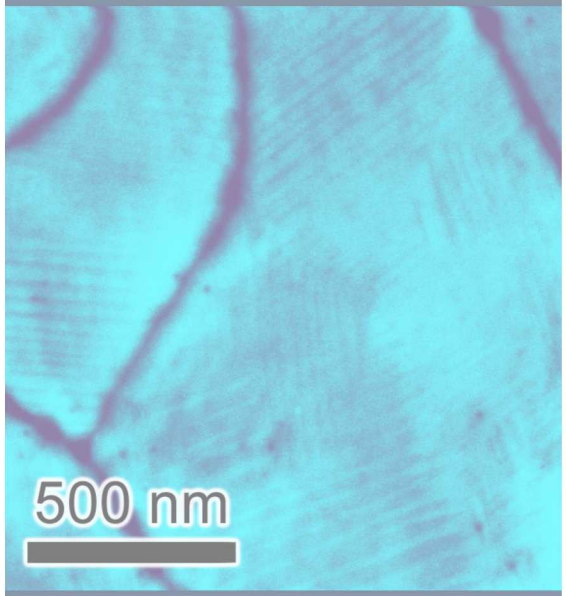
SAND2017-0935C



Positive Thermal Conductivity Change in PZT Thin Films Under Applied Fields via Substrate Clamping Relief

Jon Ihlefeld

Sandia National Laboratories

Albuquerque, NM, USA

jihlefe@sandia.gov

Sandia National Laboratories

Exceptional service in the national interest

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Collaborators

University of Virginia

- ***Brian Foley***
- ***John Gaskins***
- ***Patrick Hopkins***

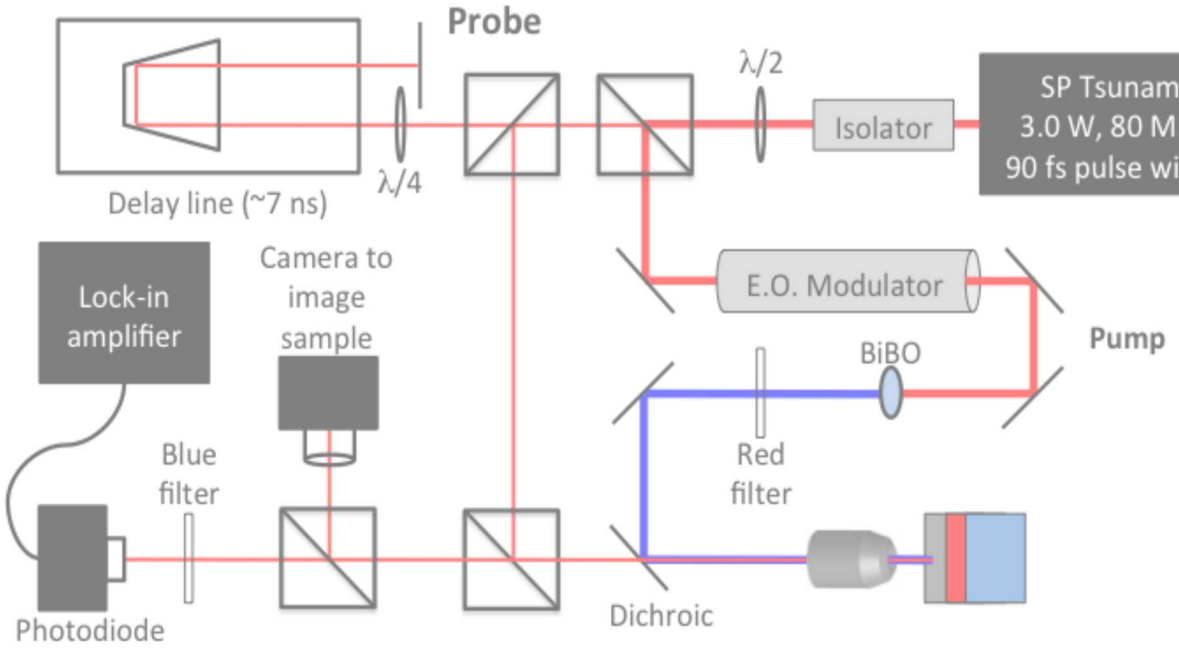
Penn State University

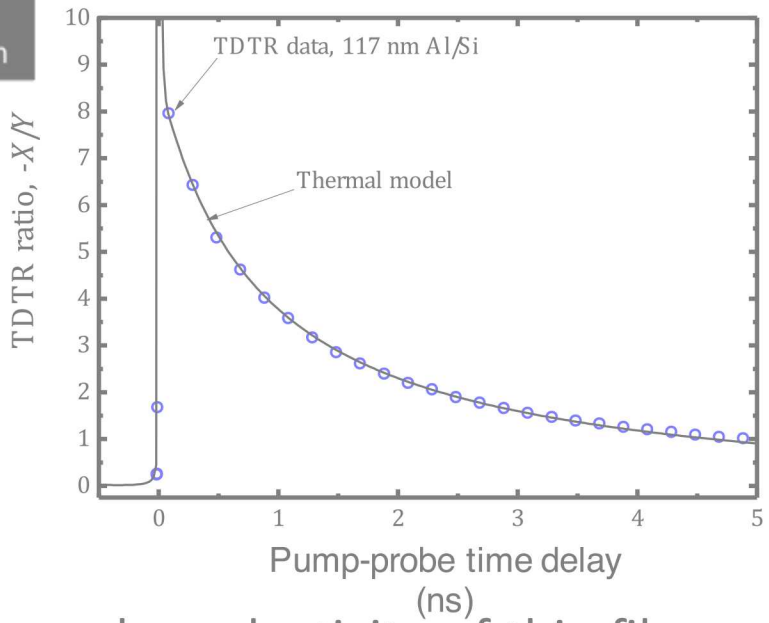
- ***Margeaux Wallace***
- ***Susan Trolier-McKinstry***

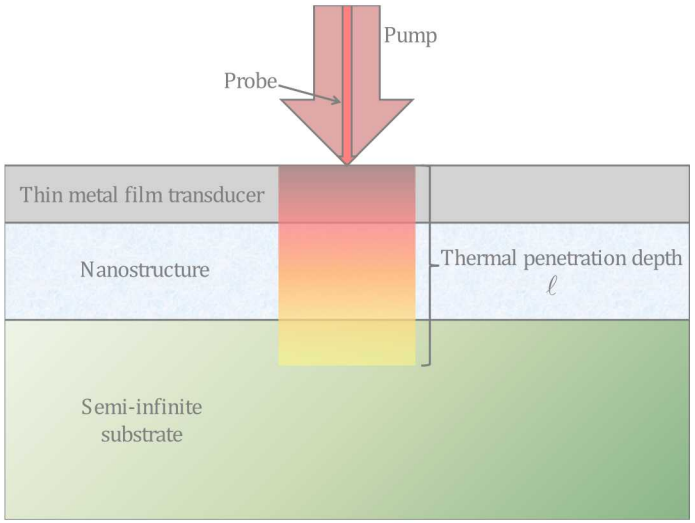
Sandia National Laboratories

- David Scrymgeour
- Joe Michael
- Bonnie McKenzie
- Douglas Medlin
- Elizabeth Paisley

Time Domain ThermoReflectance (TDTR)



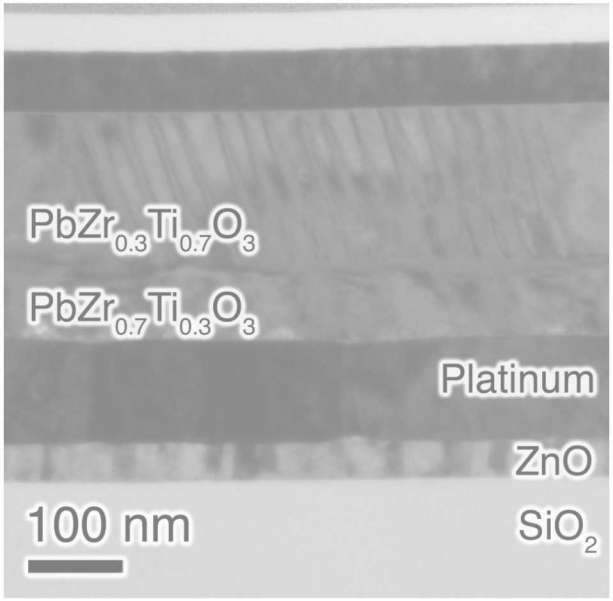


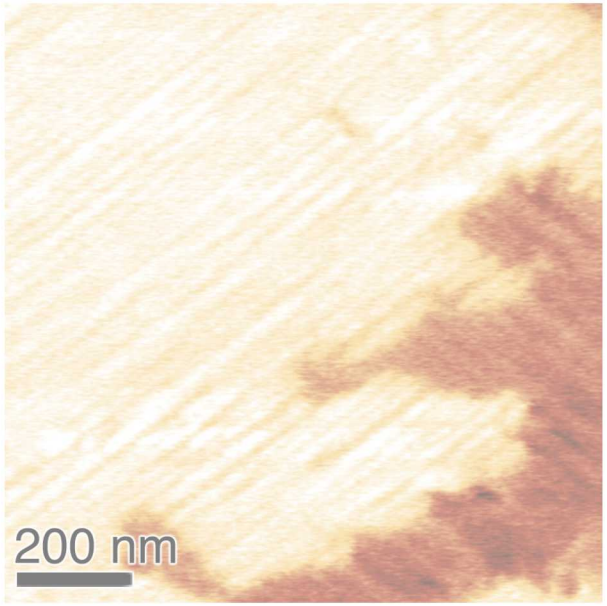


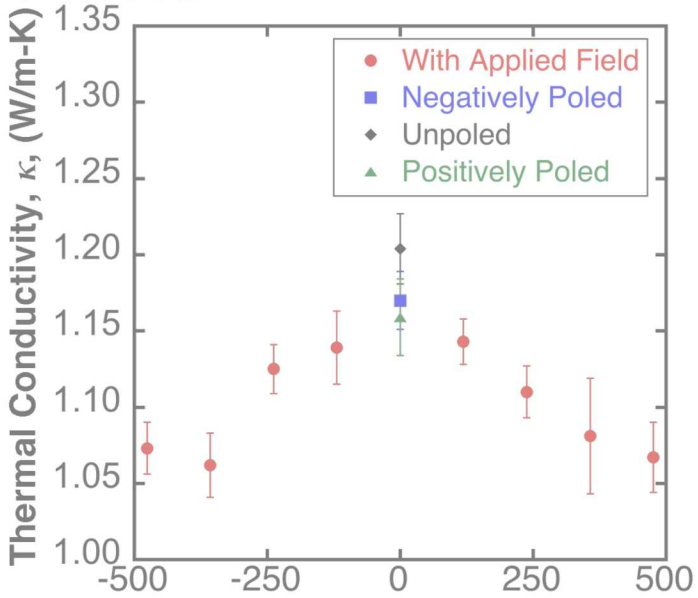
- Can measure thermal conductivity of thin films and substrates (κ) separately from thermal boundary conductance (h_K)
- Nanometer spatial resolution (~10's of nm)
- Femtosecond to nanosecond temporal resolution
- Noncontact

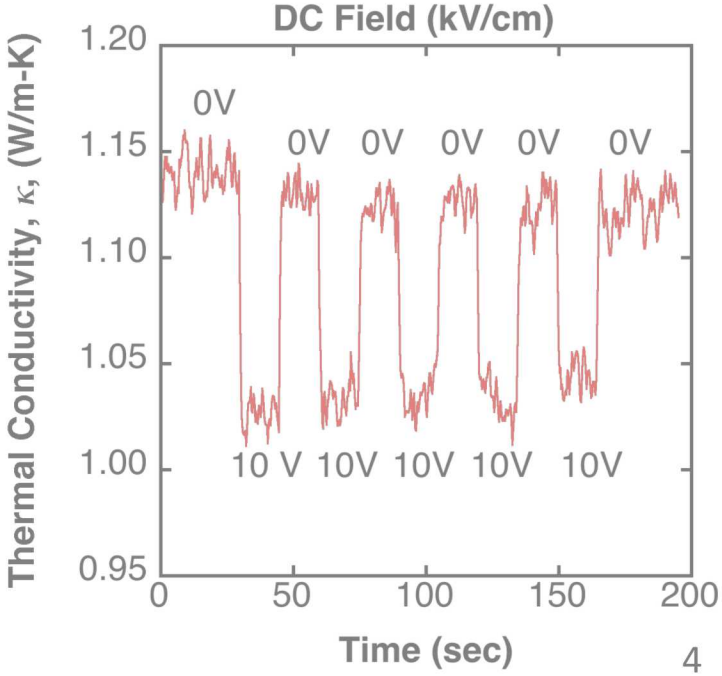
P. Hopkins *et al.*, *J. Heat Trans.* 132, 081302 (2010)

Domain Structure in PZT Bilayer Films







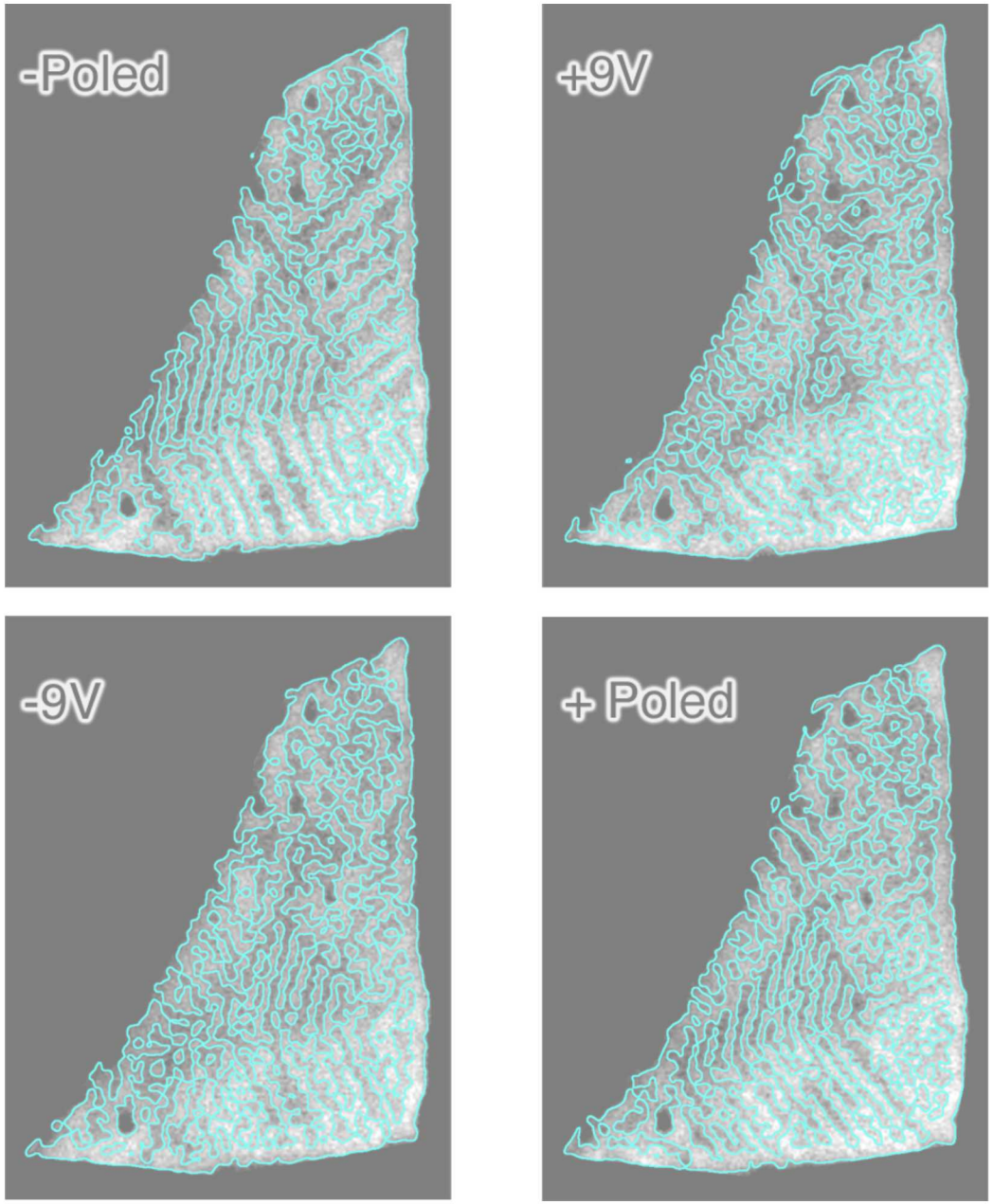


- Bilayer PZT films with high densities of ferroelastic domain walls have been previously shown to allow for voltage regulation of thermal conduction
- ***~12.5% decrease in thermal conductivity with ~500 kV/cm fields***

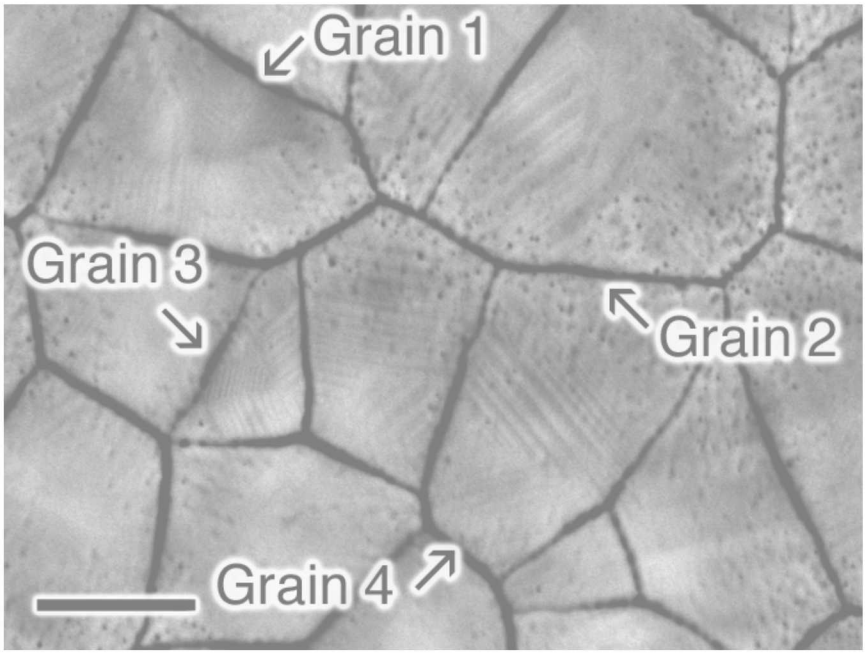
Ihlefeld, *et al., Nano Letters,* ***15****, 1791 (2015)*

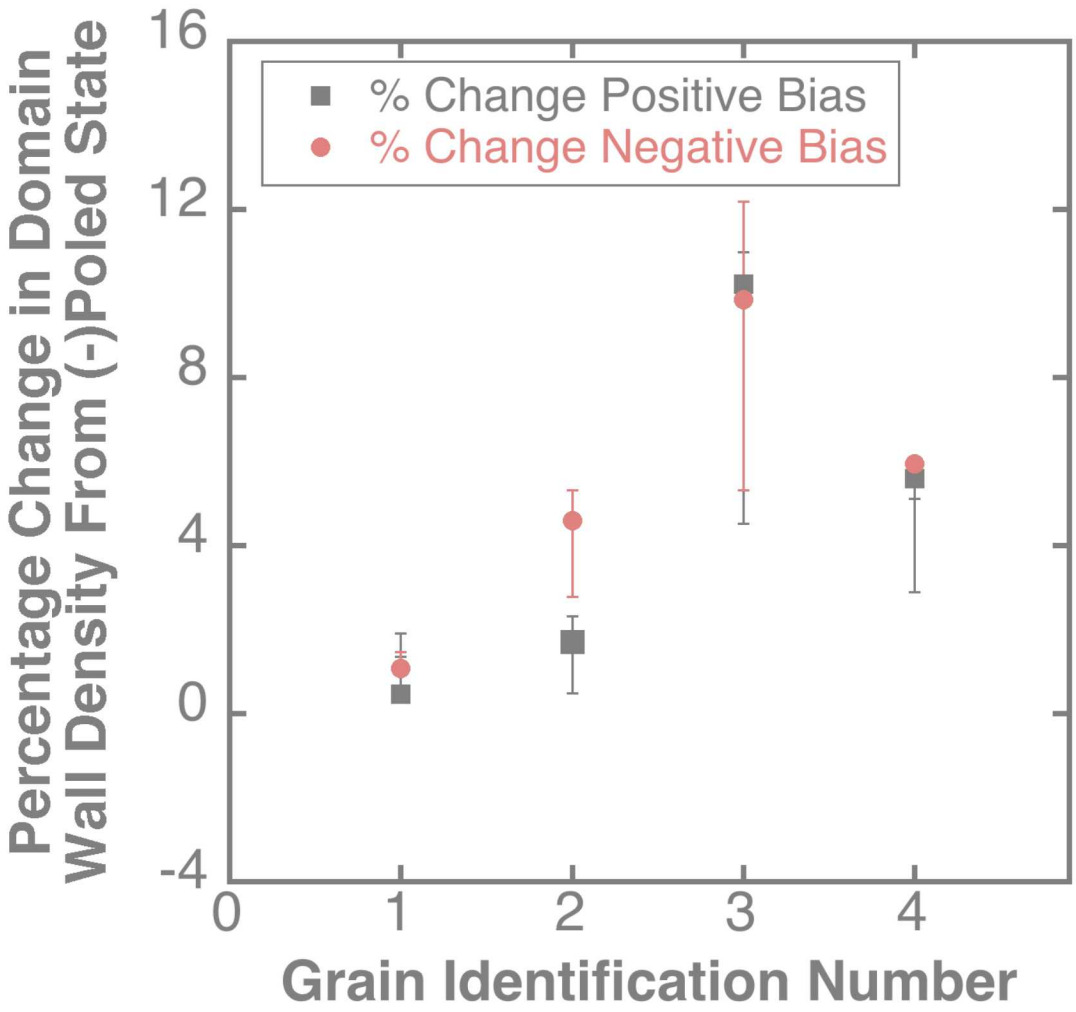
Why does thermal conductivity decrease?

- *In-situ* channeling contrast SEM enables observation of domain structure *during* application of applied voltage
  - 3 nm thick Pt electrode used to allow electron transparency
- Domain structure becomes ***more*** complex under applied fields



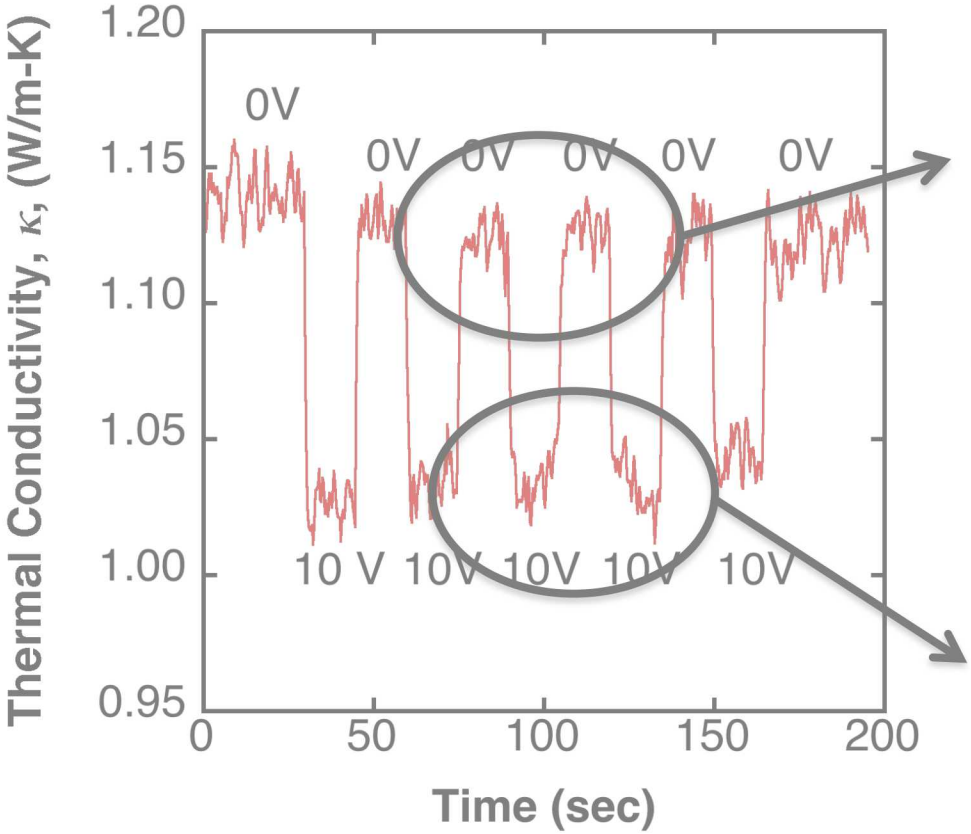
Why does thermal conductivity decrease?

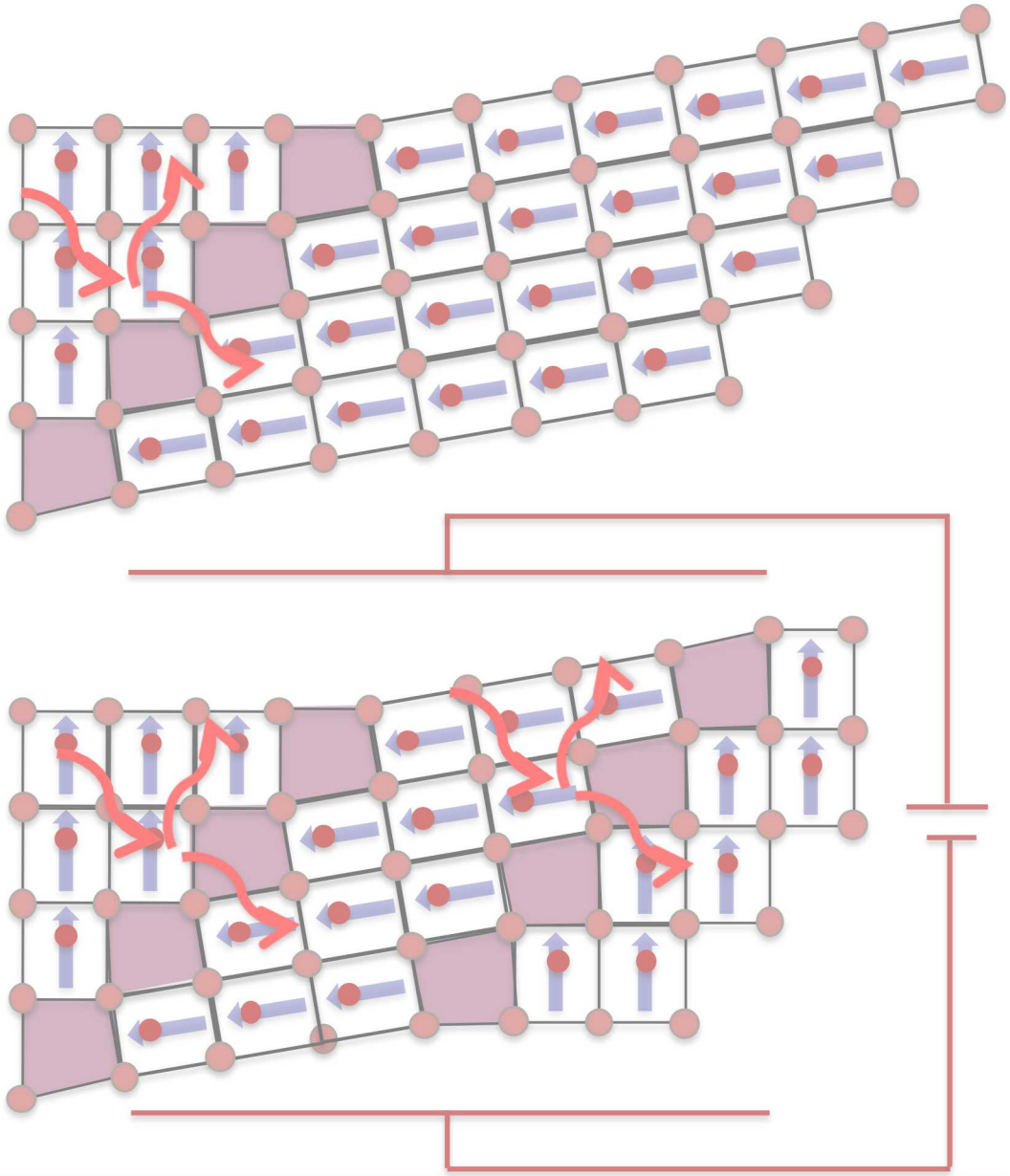




All grains with observable domain structure changes under field show increases in domain density

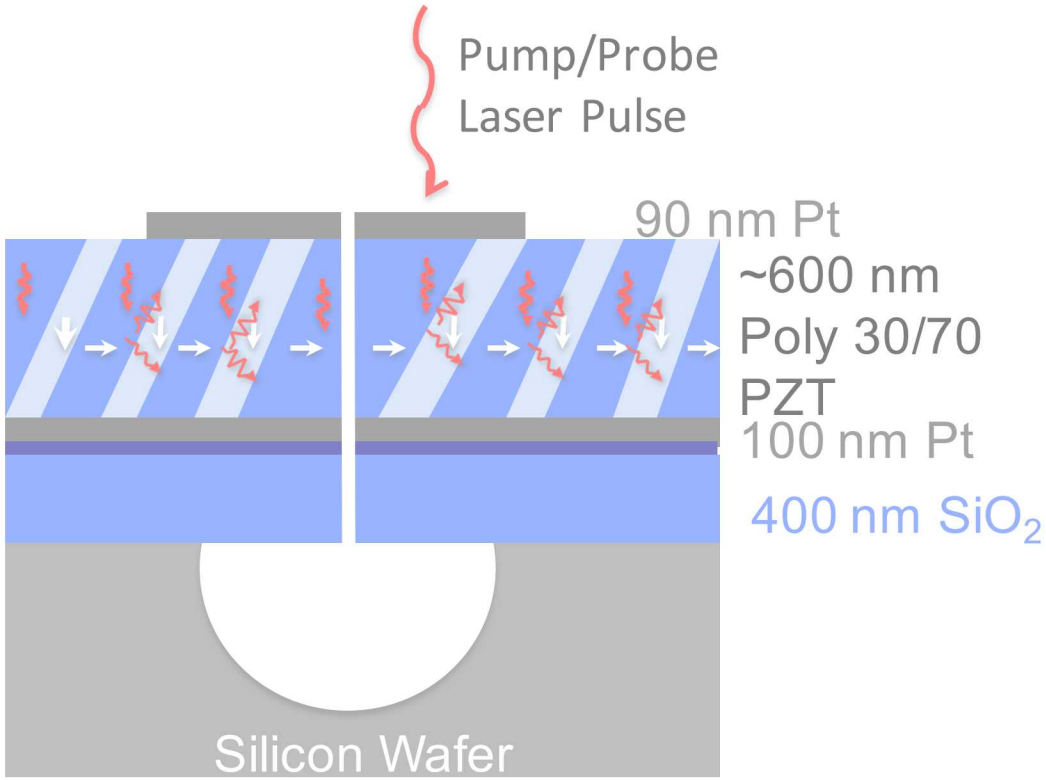
Ferroelastic Domain Walls to Modulate Thermal Conductivity



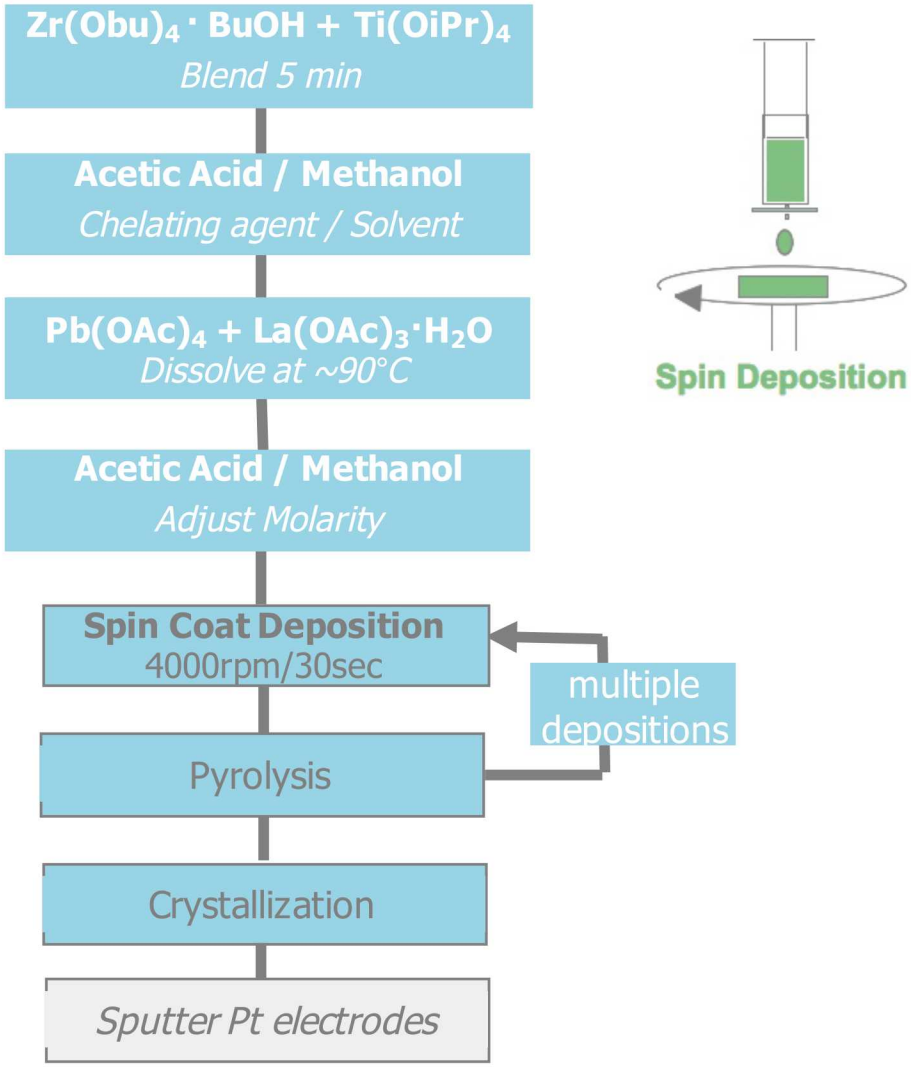


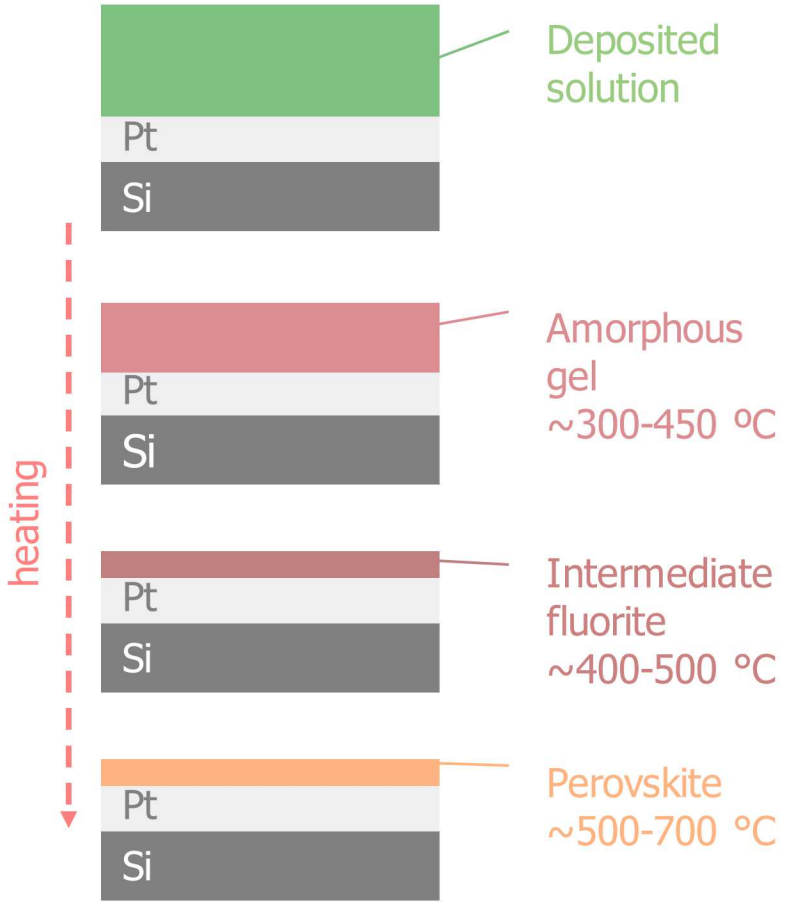
Can We Make Thermal Conductivity Increase?

- Substrate clamping minimizes ferroelastic domain wall motion
- Prevents growth of domains and decrease of domain wall density
- Relieving clamping is expected to increase ferroelastic domain wall mobility*
- Released films prepared by XeF_2 etching into a membrane structure*



SNL IMO $Pb(Zr,Ti)O_3$ CSD Film Preparation

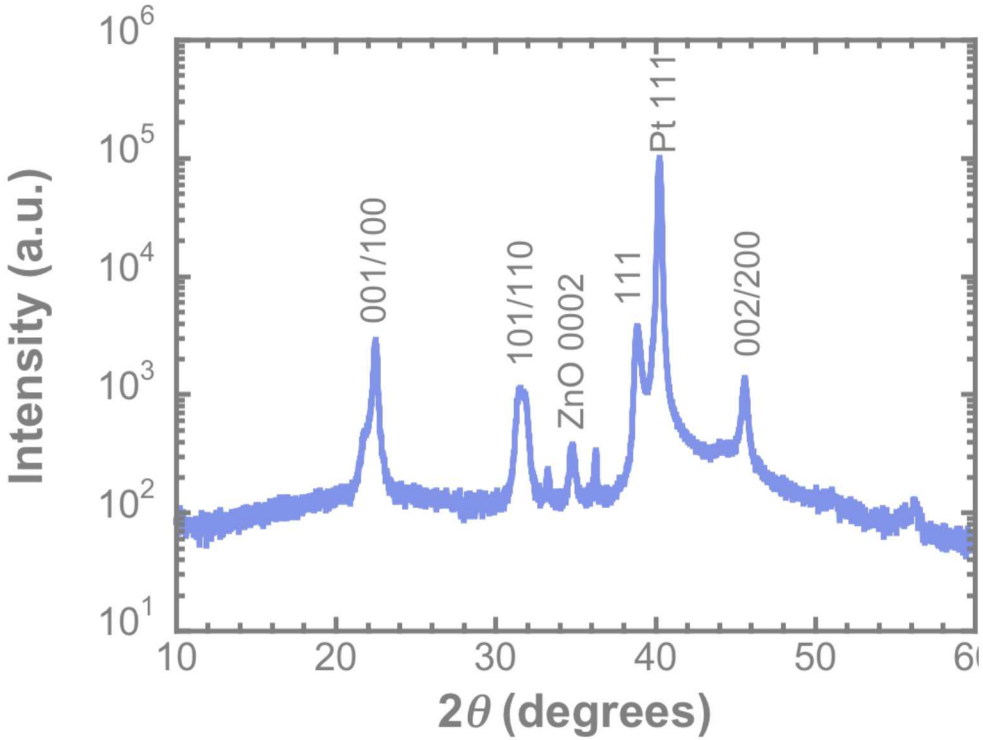


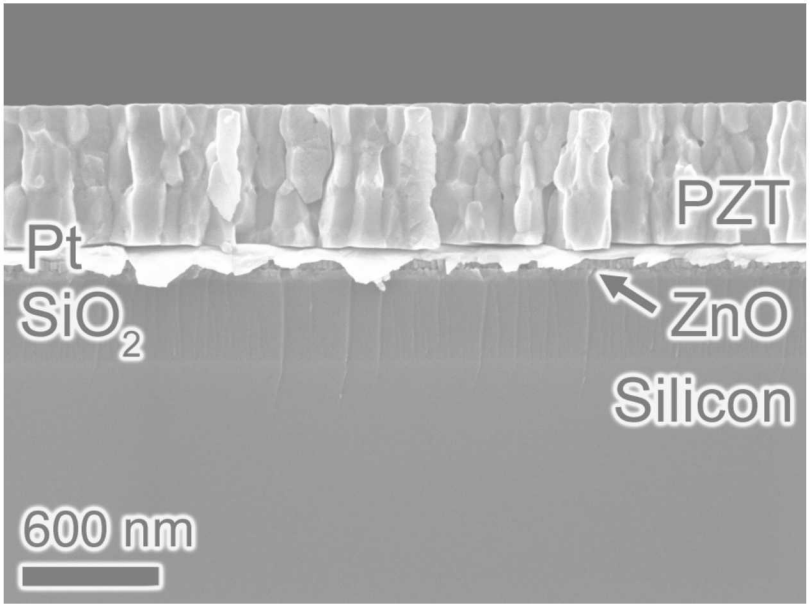


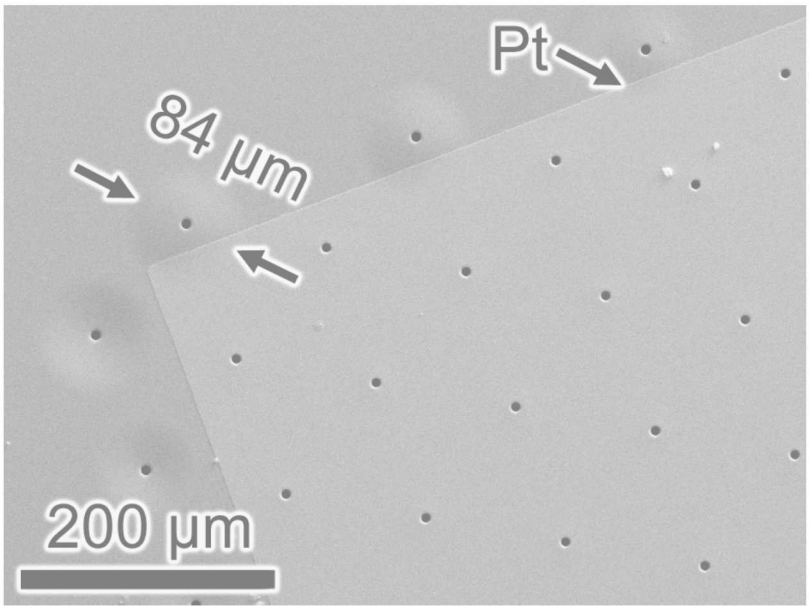
Assink & Schwartz *Chem. Mater.* (1993), G. Yi & M. Sayer. *J. Appl. Phys.* (1988)

Can We Make Thermal Conductivity Increase?

- $PbTiO_3$ layer deposited initially to favor 00*l* crystallographic texture
- Columnar, dense grain structure
- ~84 µm membrane diameter after release



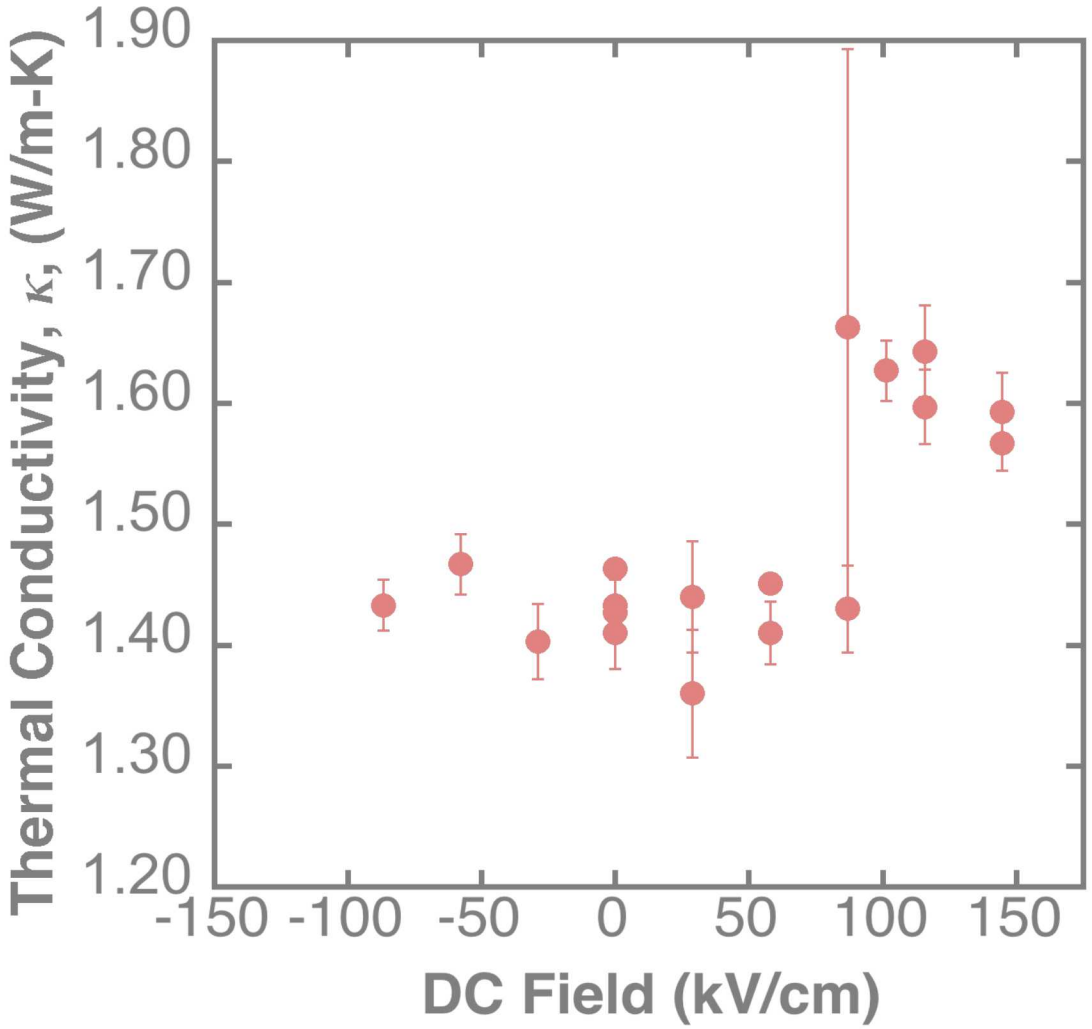




Can We Make Thermal Conductivity Increase?

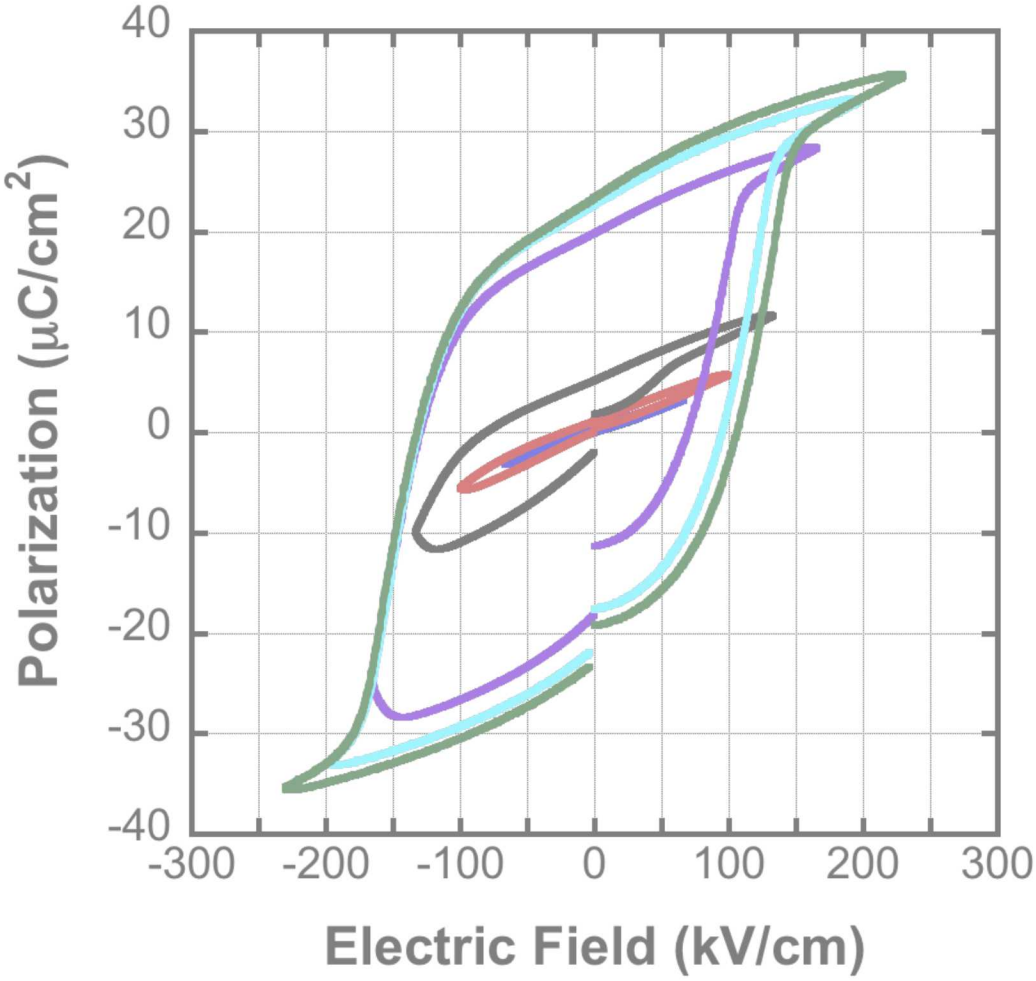
Sandia National Laboratories

- Measured thermal conductivity on membrane structure while applying fields
- Observe discontinuous 13% ***increase*** in κ at ~90 kV/cm
- Relieving strain results in increased thermal conduction
  - Is domain wall motion the origin?

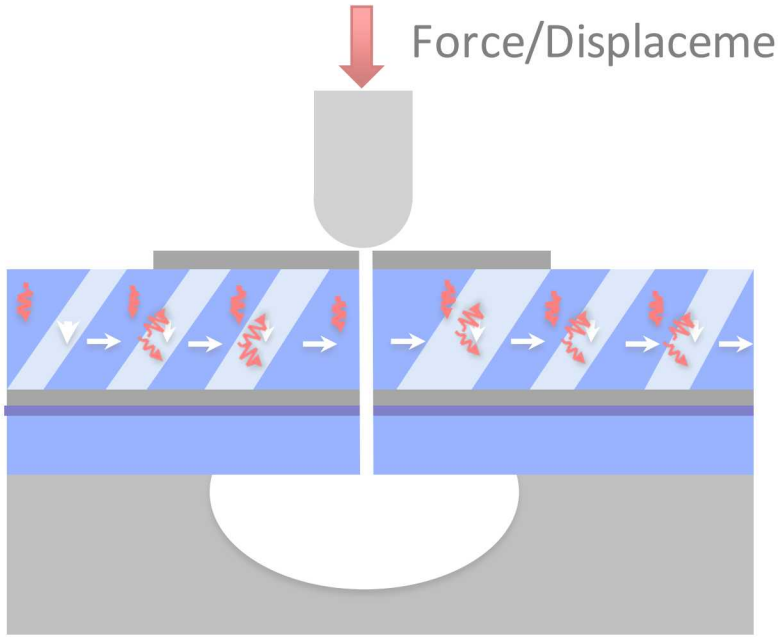


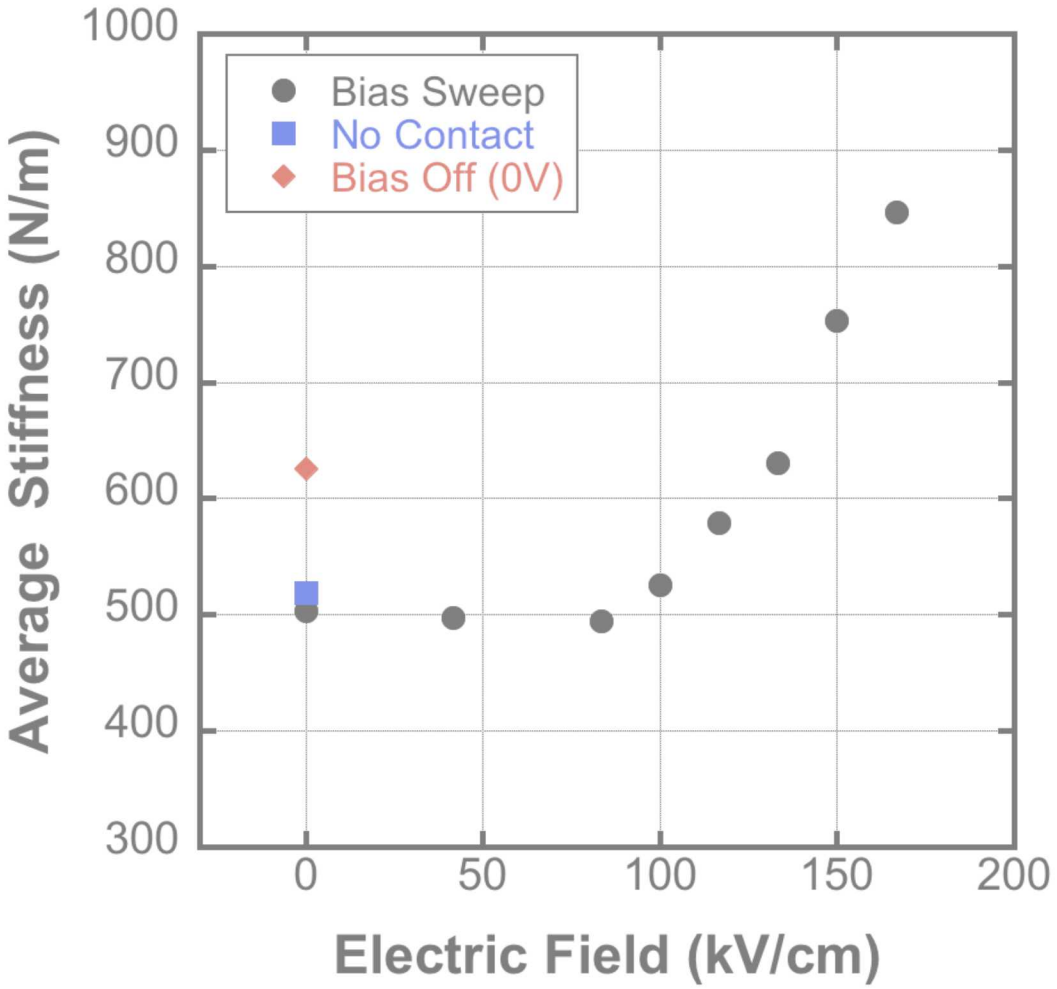
Can We Make Thermal Conductivity Increase?

- Polarization-field response measured
  - *Samples both clamped and released regions*
- E_c = ~100-110 kV/cm
  - E_c from P-E measurements slightly higher than that from the κ-E measurements
  - What is the role of releasing the film?



Can We Make Thermal Conductivity Increase?

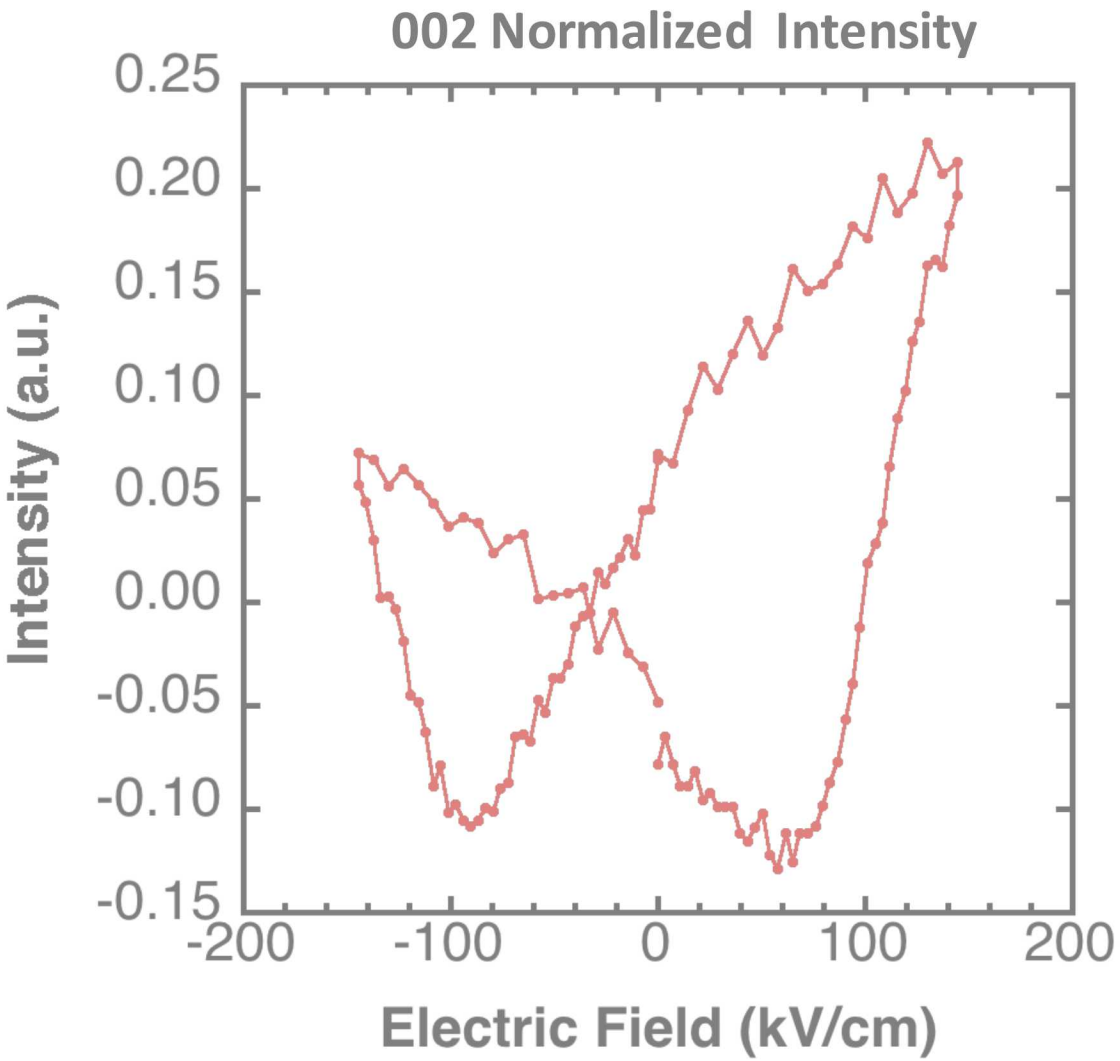




- Nanoindentation stiffness measurements performed on membrane
- Onset of stiffness increase is ~90-100 kV/cm
  - ***Correlates with discontinuous κ increase***

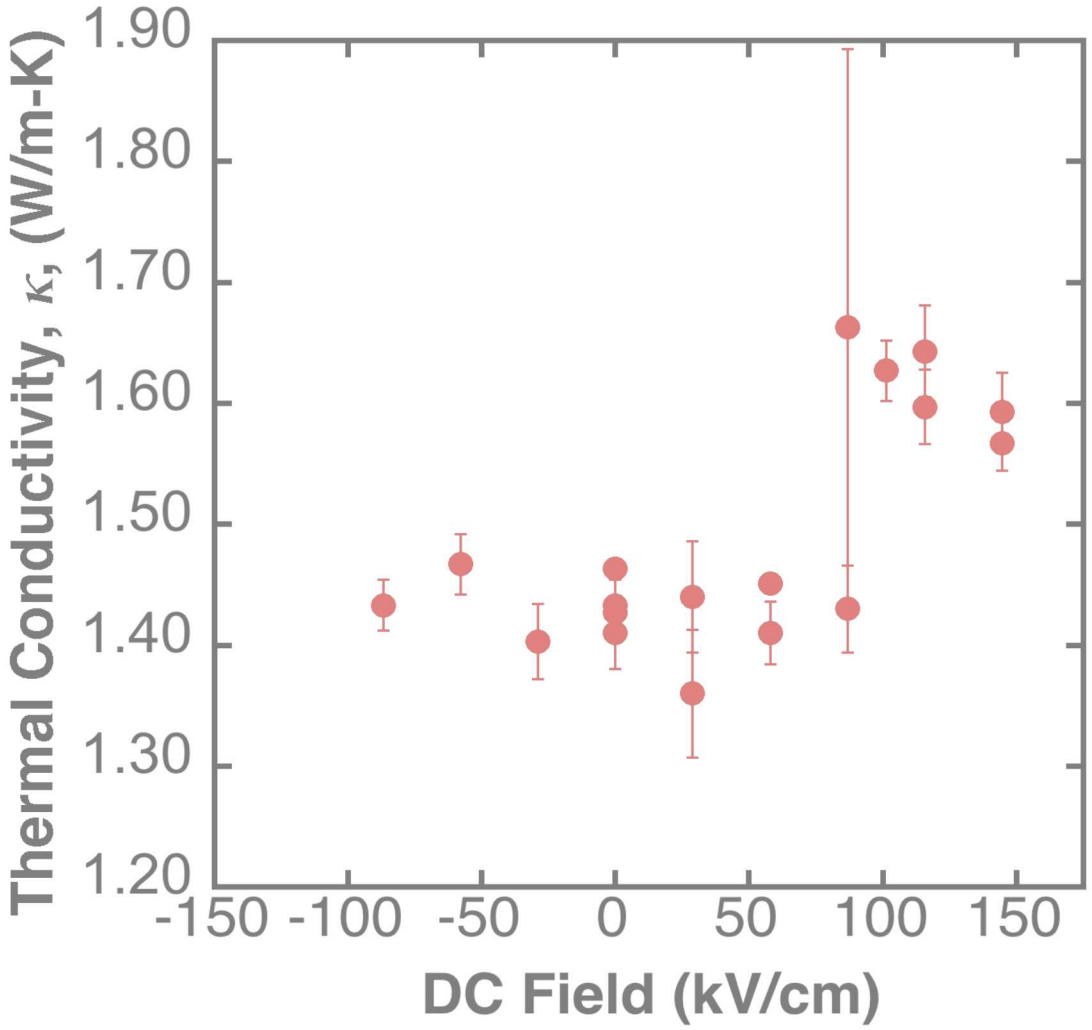
Can We Make Thermal Conductivity Increase?

- X-ray μ-diffraction performed on membrane
- Observe 'd_{33}-like' response
- E_c = ~90 kV/cm
  - ***Correlates with discontinuous κ increase***



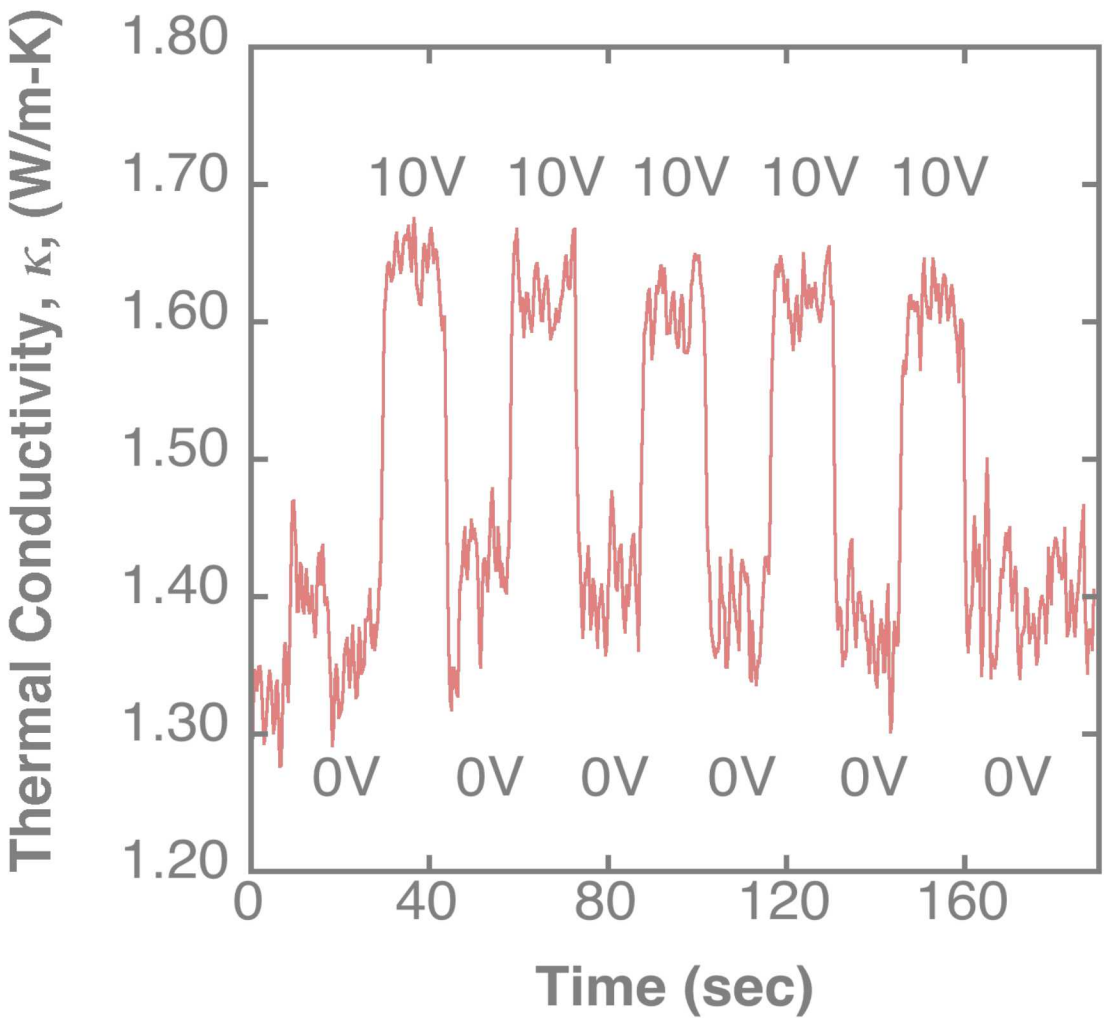
Ferroelastic Domain Wall Motion is Responsible for Thermal Switching Behavior

- Field at which thermal conductivity discontinuity occurs correlates with:
  - Coercive field as measured from P(E) response
  - Onset of stiffness discontinuity in membrane
  - Coercive field as measured from X-ray microdiffraction on membrane
- ***Ferroelastic Domain Wall Reconfiguration is Responsible***



Repeatability of Thermal Switching Behavior

- Temporal response of thermal regulation measured
- Thermal conductivity change for membrane structures is fast
  - Sub-300 ms timescale
- Response is repeatable
  - ***Indicates that domain wall densities decrease while under field and increase when field is removed***



Summary

- Both negative and positive voltage coefficients of thermal conductivity can be obtained in ferroelectric thin films
  - Bilayer structures form more domain walls in response to field
  - Strain-relieved structures reduce in domain wall density in response to field
- Ferroelectric films are a promising avenue toward active phonon transport regulation