Probing Magnetism and Ferroelectricity with Synchrotron X-ray Microdiffraction

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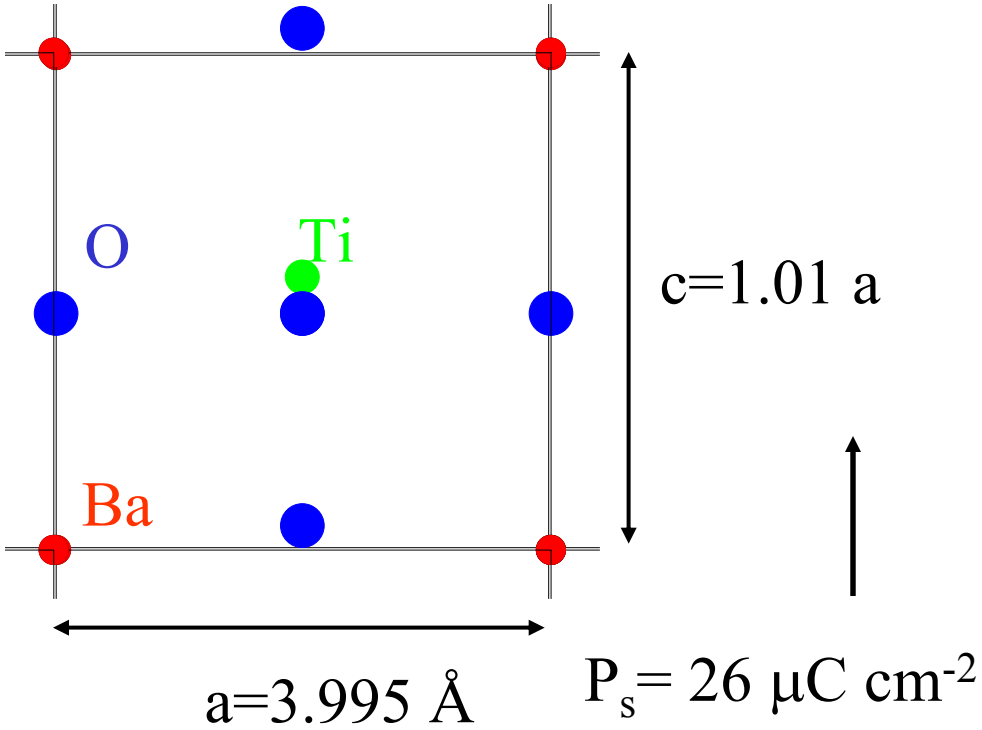
Outline

- Overview: Physical Phenomena, Motivation, and Tools
- Polarization Switching and Fatigue in Ferroelectric Thin Films
- What's the state of the art now?
- What would we like to be able to do?
- What can we do in the near future?

Ferroelectric Materials

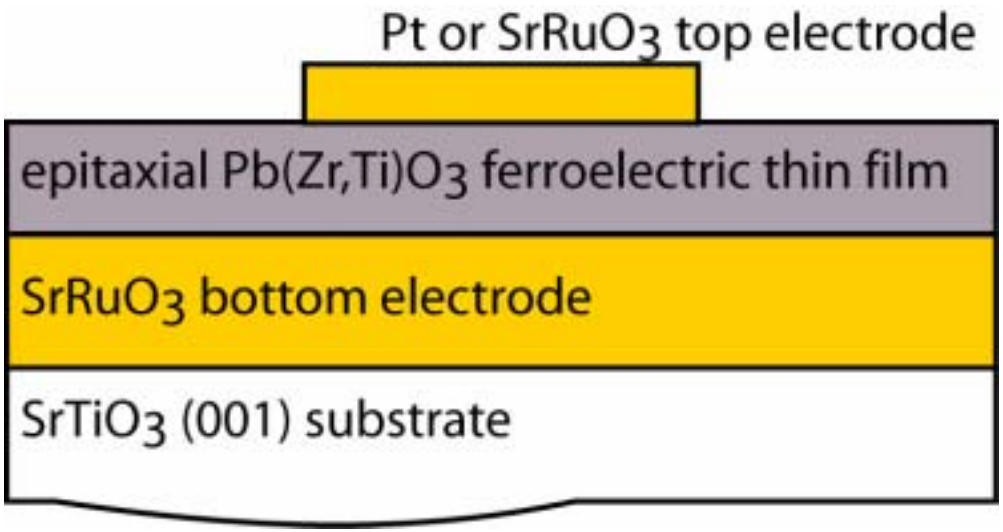
- Example: tetragonal phase of barium titanate ($BaTiO_3$)
- 6 possible polarizations
- Organized naturally or artificially into domains.



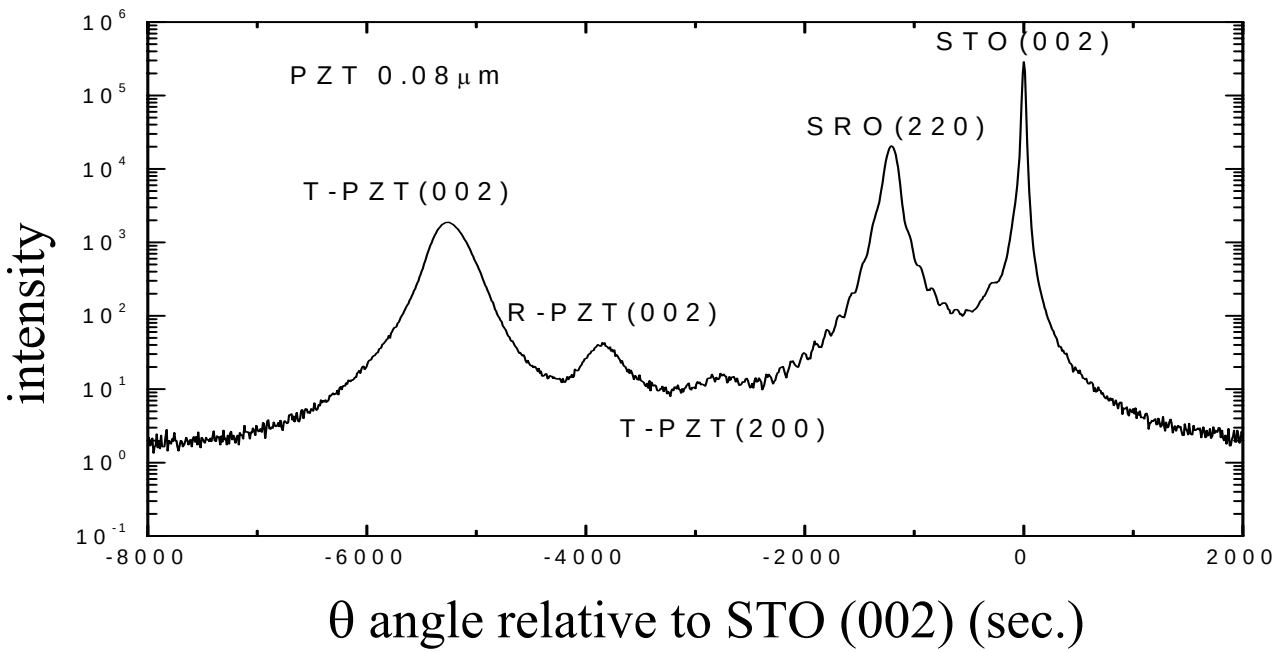
What can be learned about polarization switching in ferroelectric materials?

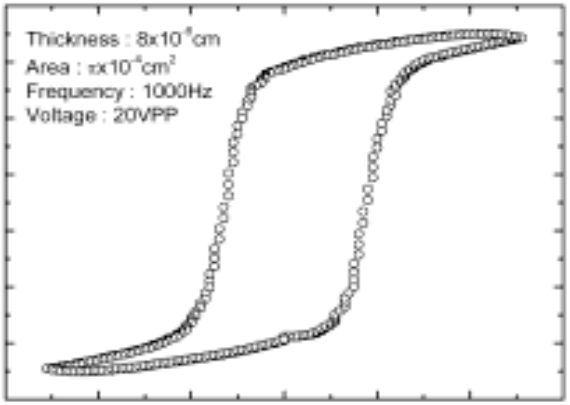
Problems with existing techniques: time resolution, electrodes, quantification.

Epitaxial PZT Thin Film Capacitors



tetragonal $PbZr_{1-x}Ti_xO_3$
x=0.55
80 nm or 160 nm PZT thickness



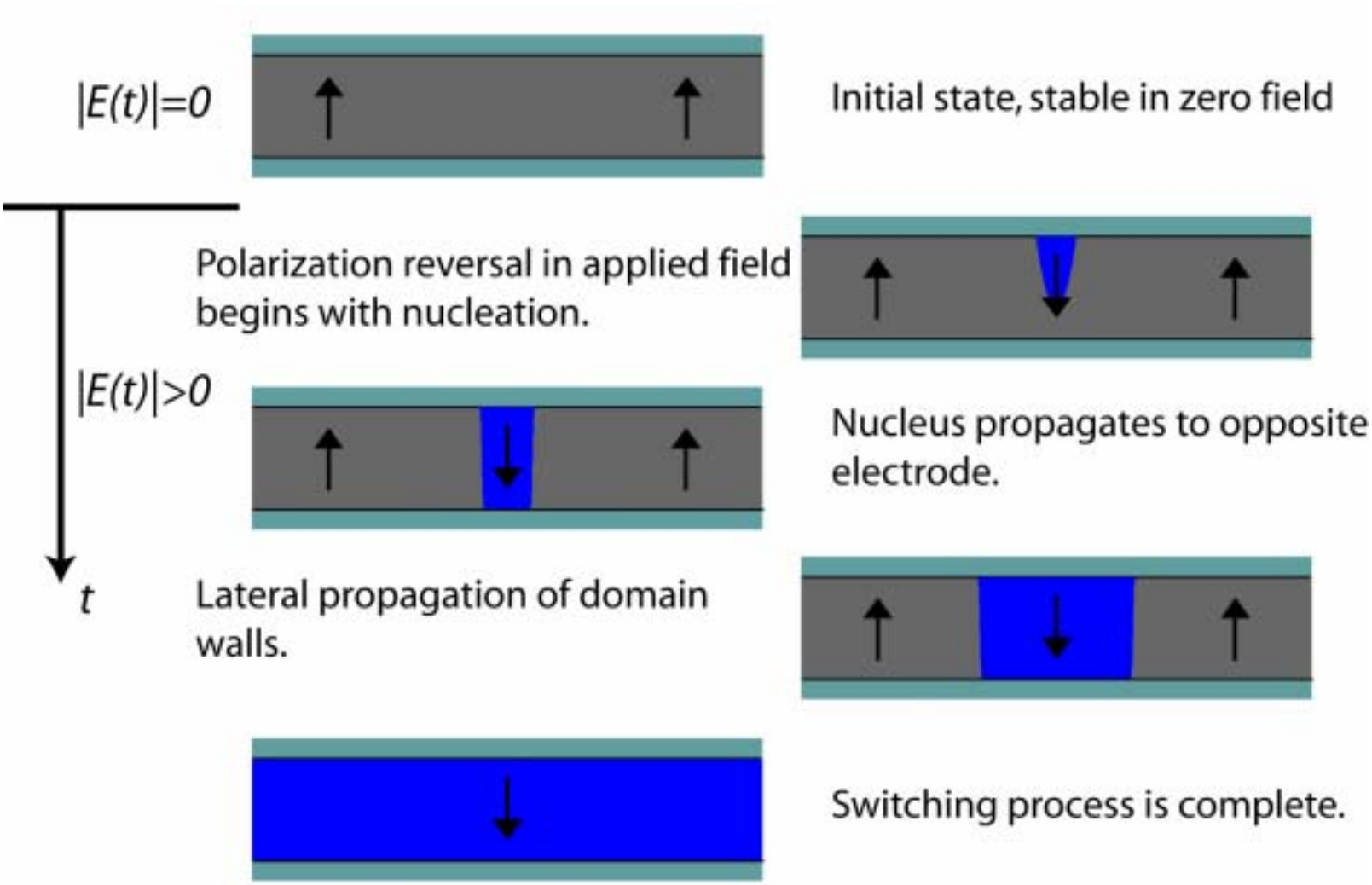


$2\,P_r \approx 95\ \mu C/cm^2$

Open Questions in the Physics of Ferroelectric Devices

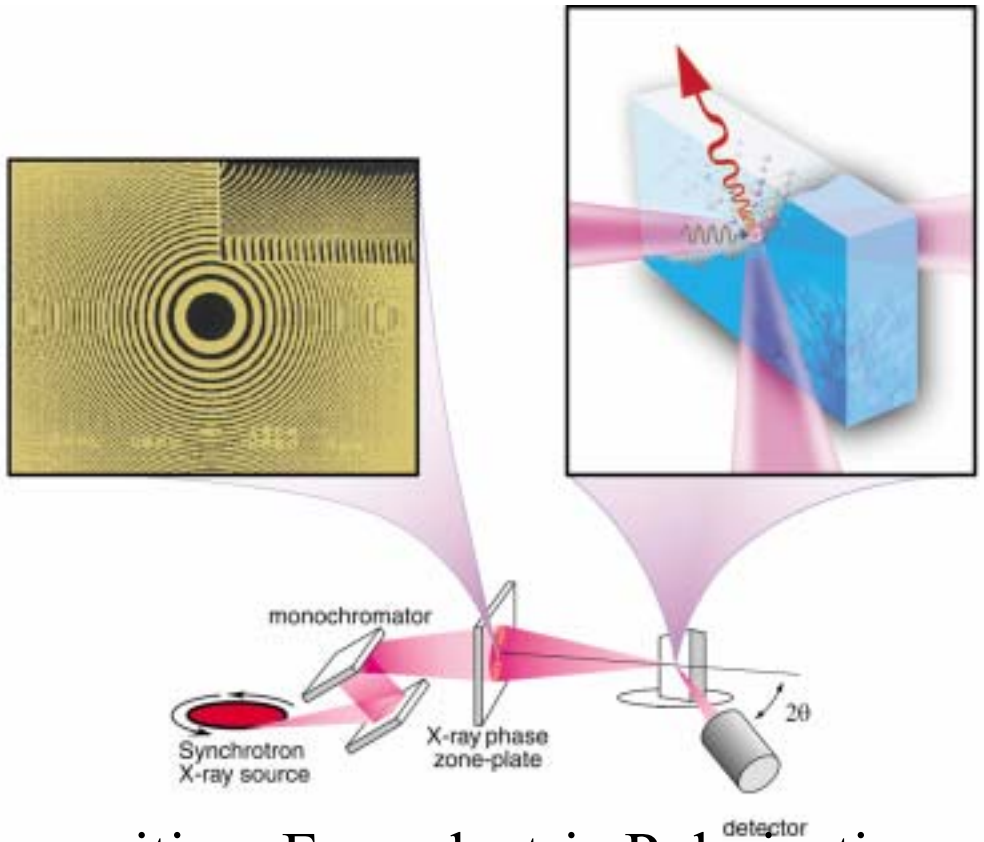
- How does polarization switching occur?
  - What is the speed of a moving domain wall?
  - How quickly can a ferroelectric thin film capacitor switch polarization?
  - Are there any intermediate structural states that are important to switching?
- How can the piezoelectric response of actuator materials be optimized?

“Standard Model” of Switching



X-ray Microdiffraction as a Tool for Physics and Materials Science

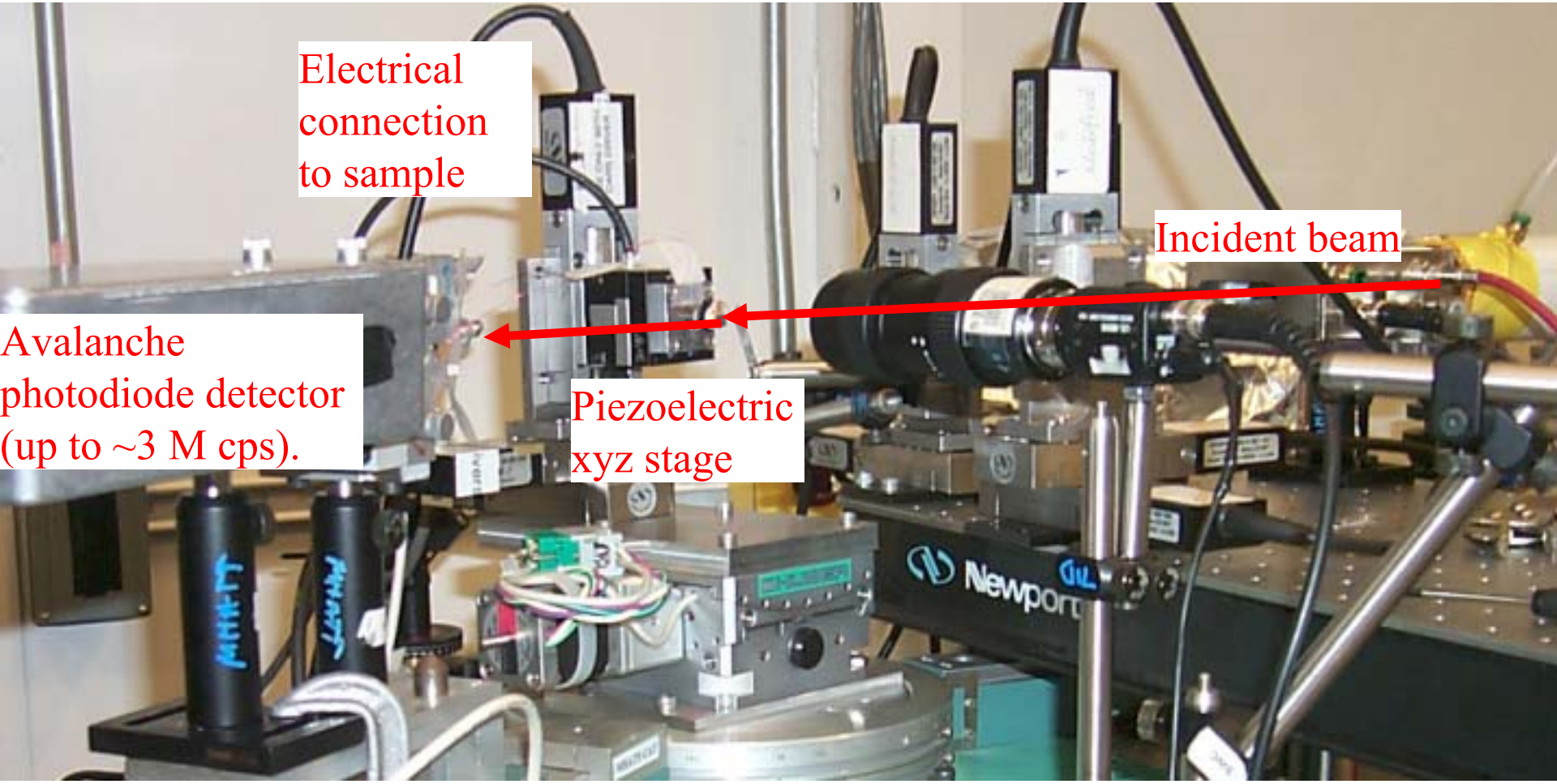
- 6 to 30 keV
- 10^{10} ph/s/0.01%BW
- Minimum spot (APS)- 0.1 x 0.1 μm^2
- < 1 ns time resolution



Contrast from: Diffraction, Composition, Ferroelectric Polarization, Magnetization.

Problems with existing techniques: time resolution, electrodes, quantification.

Time Resolved Microdiffraction

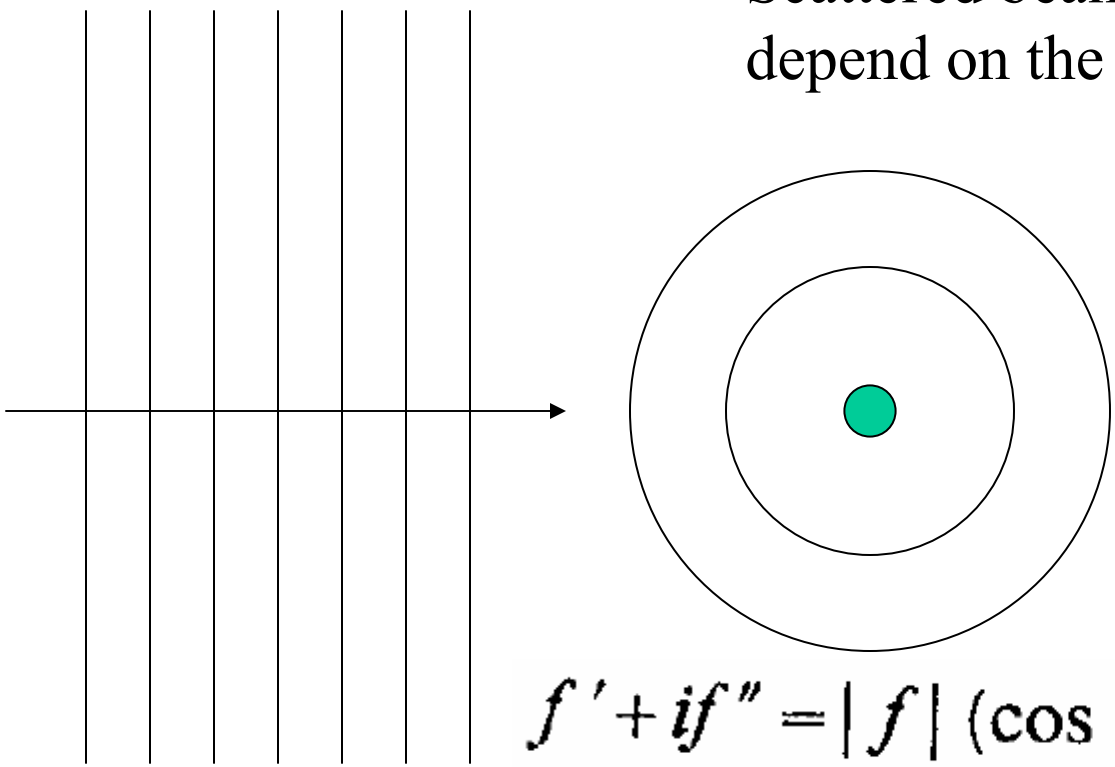


Use avalanche photodiode with multichannel scaler to time-resolve the diffraction signal during voltage pulses.

Reminder: X-ray Scattering From Atoms

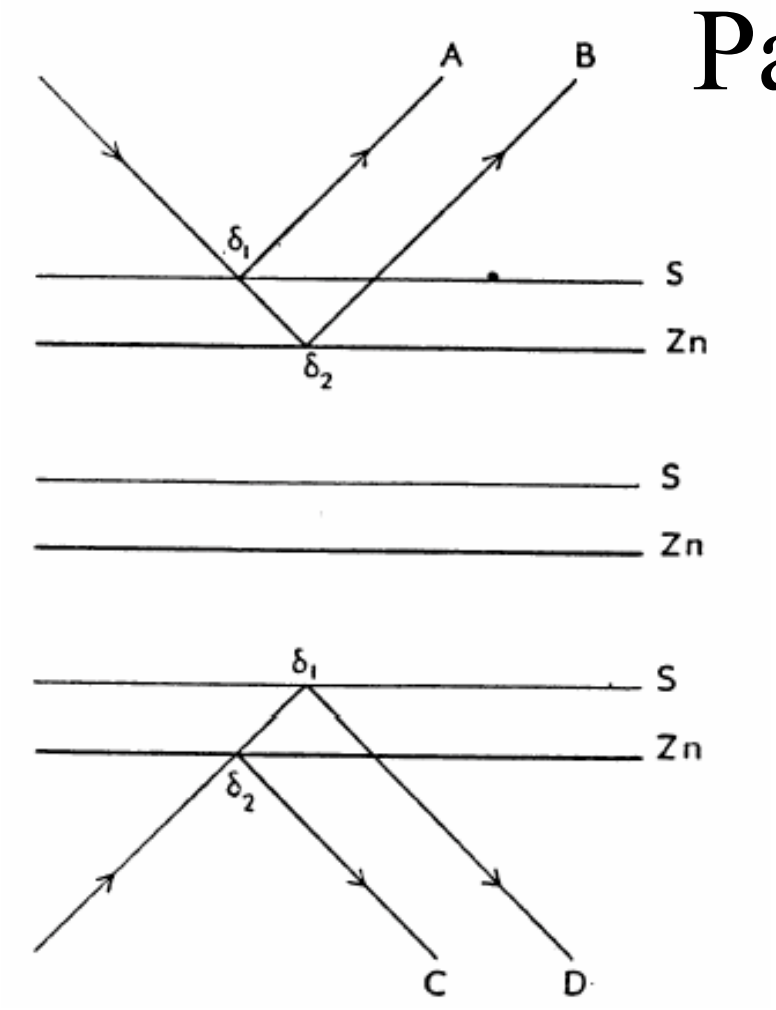
X-rays are scattered by atom into (approximately) all directions.

Scattered beam amplitude and phase depend on the atom.



$$f' + if'' = |f|(\cos\delta + i\sin\delta) = |f|\, e^{i\delta}$$

Symmetry of X-ray Diffraction Patterns



Bragg condition: Phase relationship between unit cells.

Intensity of peaks set by phase relationship between beams scattered within each unit cell.

Phase difference between B and A:

$\delta_0+\delta_2-\delta_1$

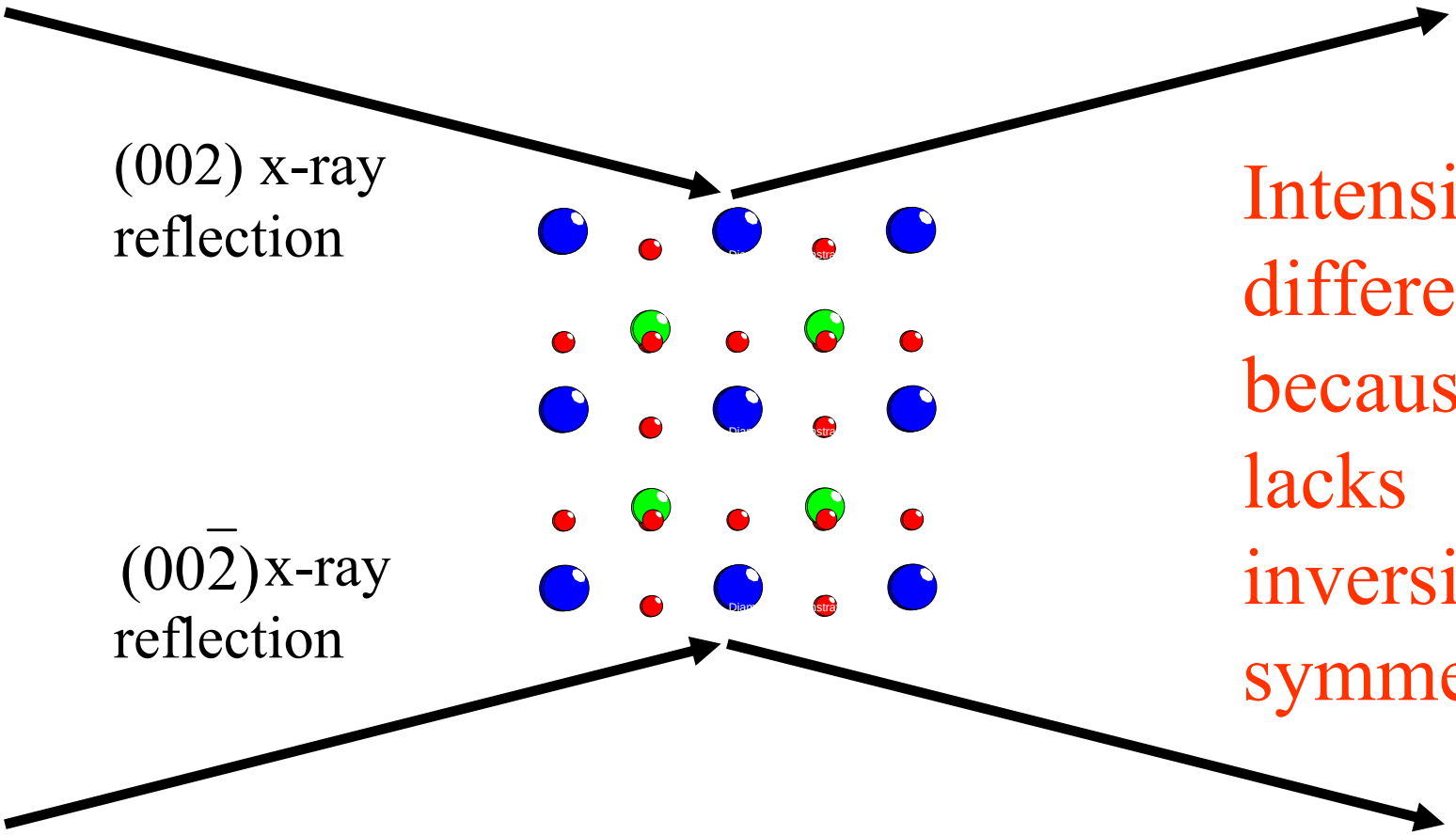
Phase difference between D and C:

$\delta_0+\delta_1-\delta_2$

Phase difference depends on polarity of crystal.

ZnS (111): Coster, Knol, and Prins (1930).

Anomalous scattering effects in PZT

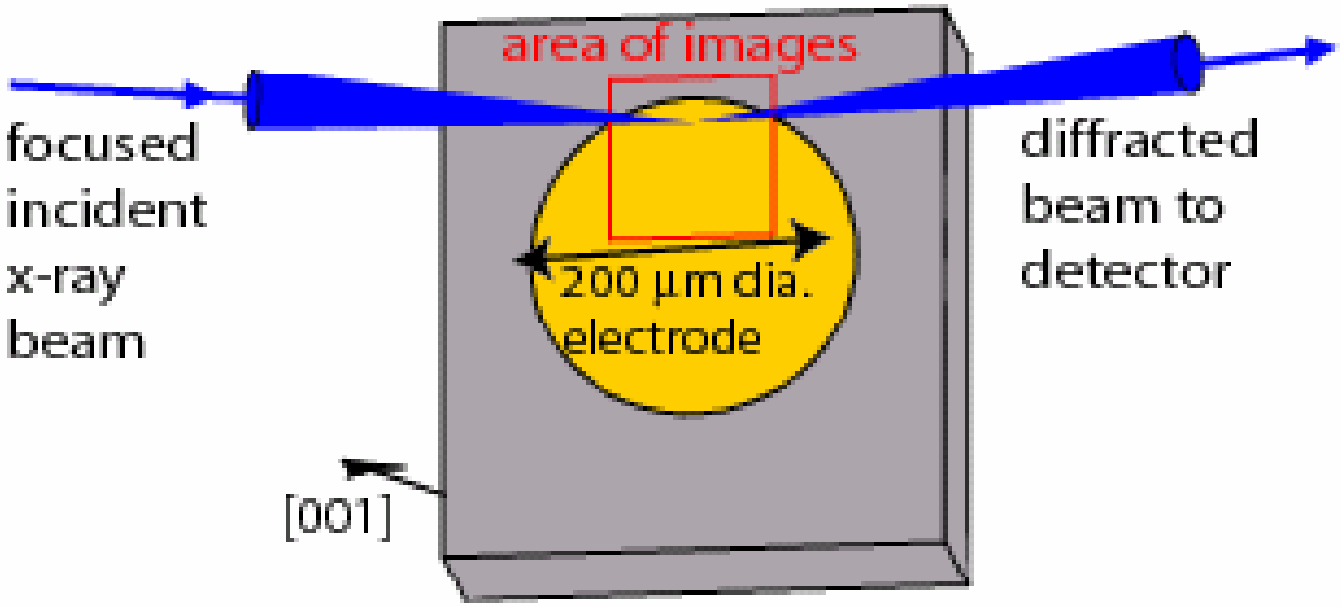


Intensities are different because PZT lacks inversion symmetry!

Outline

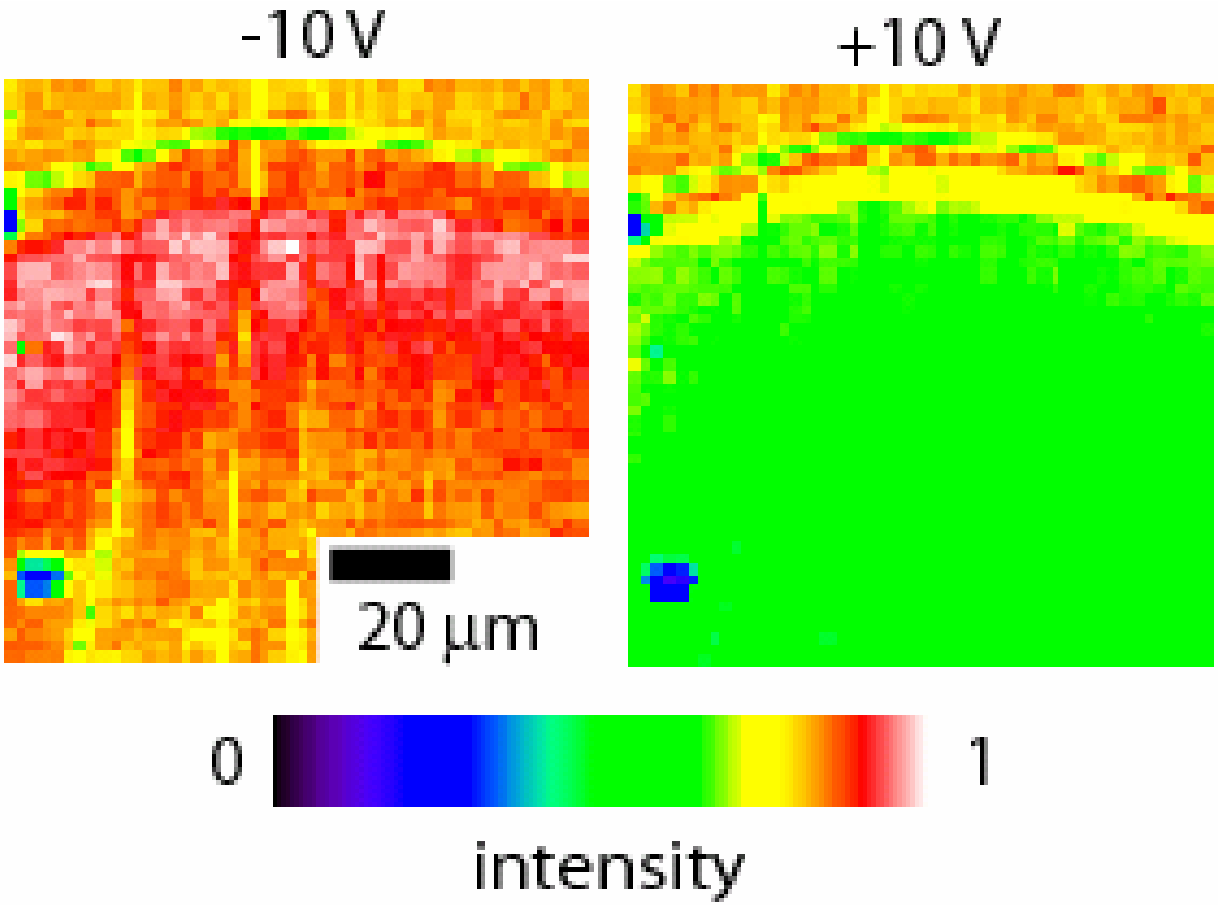
- Polarization Switching and Fatigue in Ferroelectric Thin Films
- Where are we now?
- What can we do?

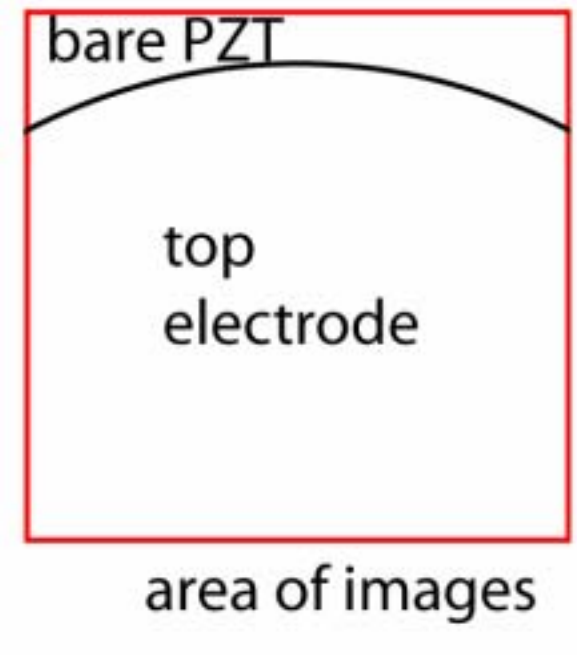
Imaging Ferroelectric Polarization Switching with X-ray Microdiffraction



X-ray photon energy
energy 10 keV.

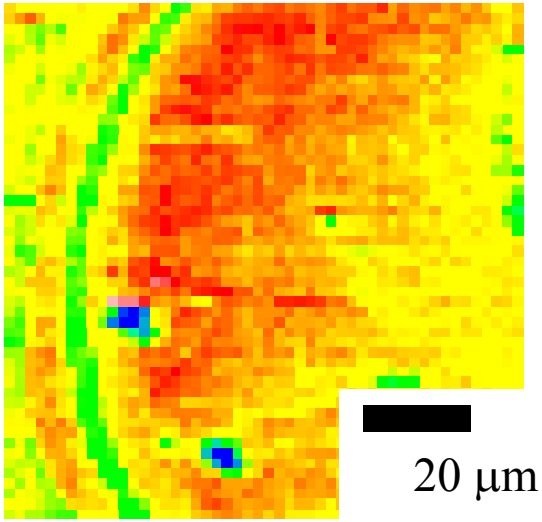
Imaging PZT Polarization Switching





Maps of the intensity of PZT (002) reflection vs. the position of the beam on the sample.
Intensity following –10 V pulse is 25% higher than

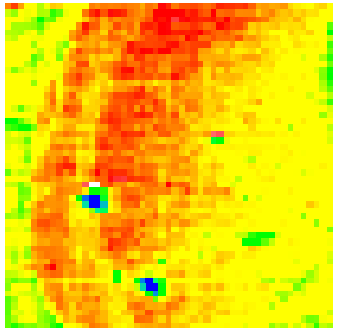
Switching with $E \approx E_c$



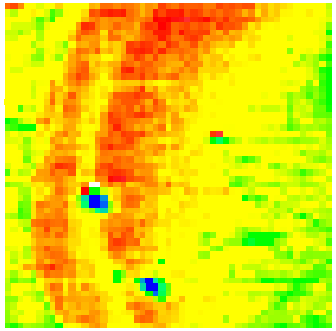
-10V

1. Apply negative voltage pulse to produce uniform polarization.

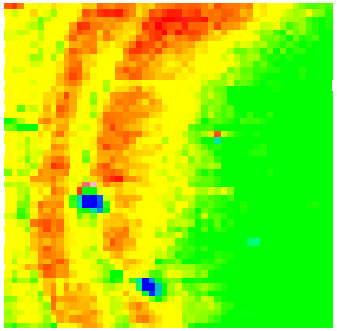
500 μs triangle pulses



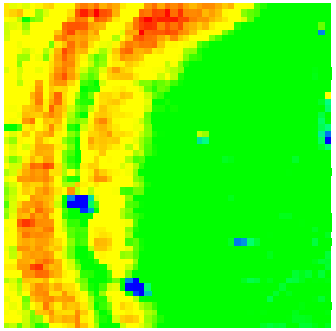
0.8V



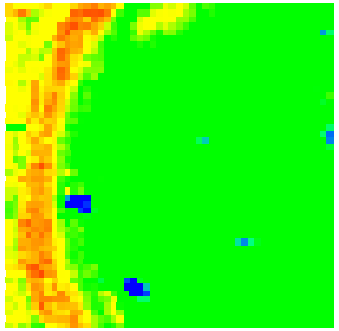
1.2V



1.6V



2.0V

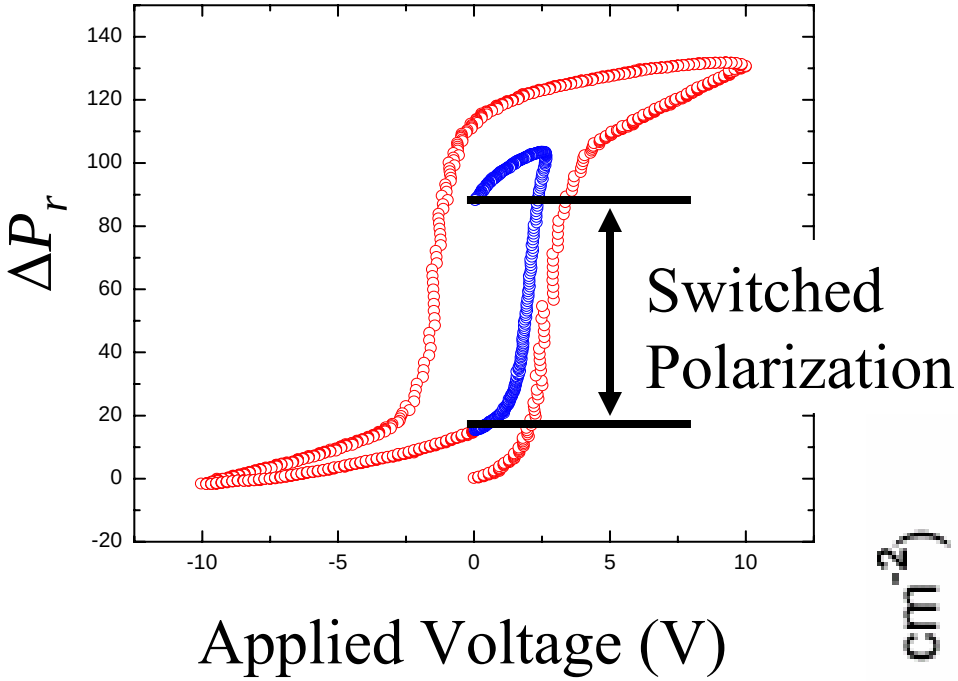


2.8V

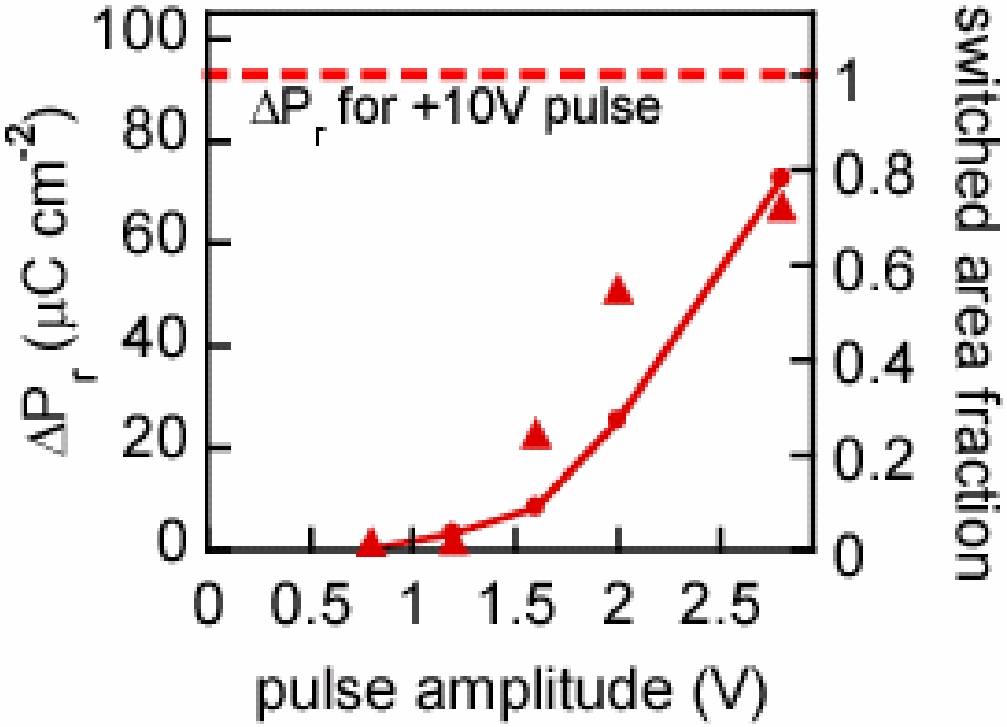
2. Apply a positive pulse near E_c.

Device switches in well defined areas.

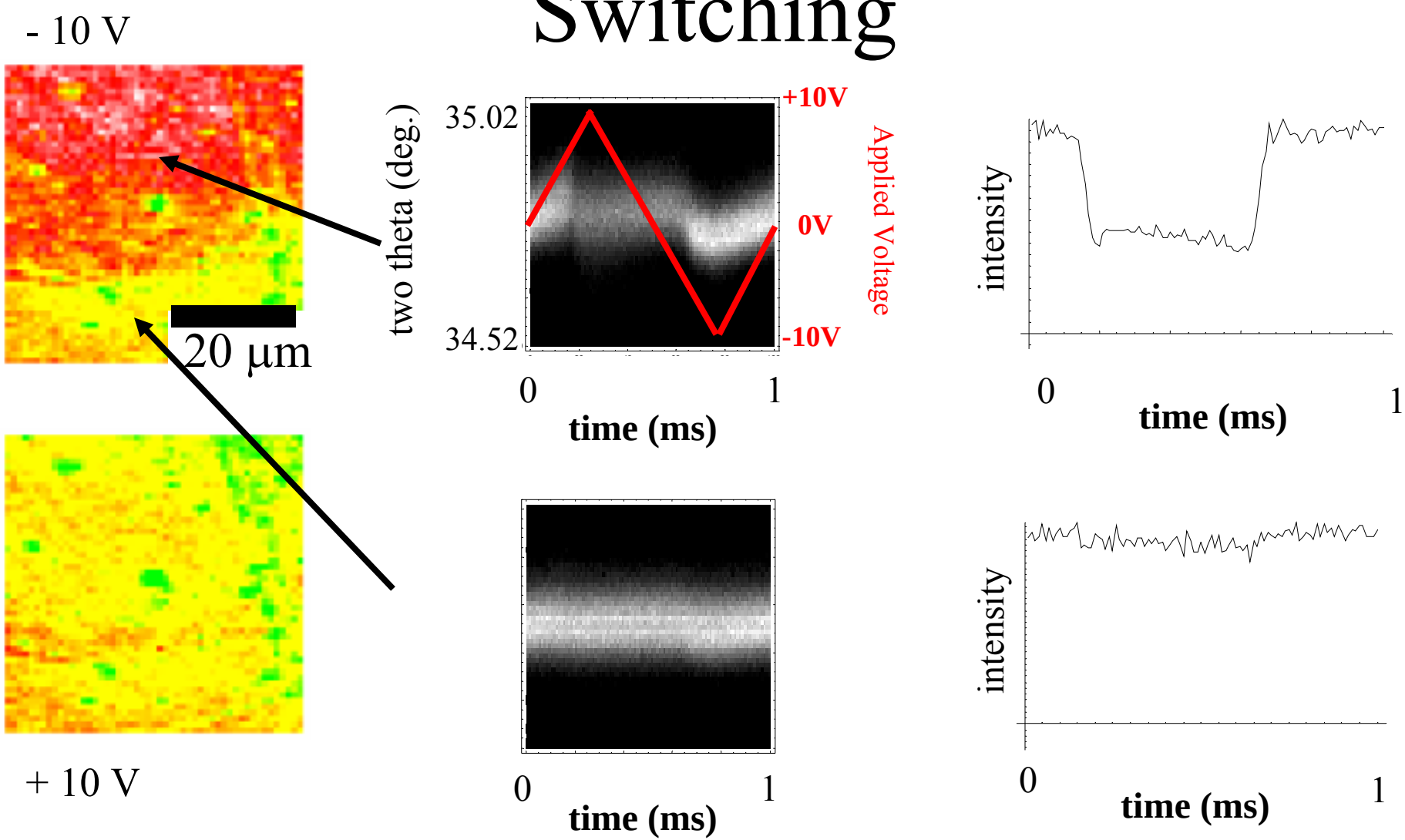
Relationship of Polarization to Switched Area



Switched Area Measured from X-ray Microdiffraction Maps is Proportional to the Total Switched Polarization.



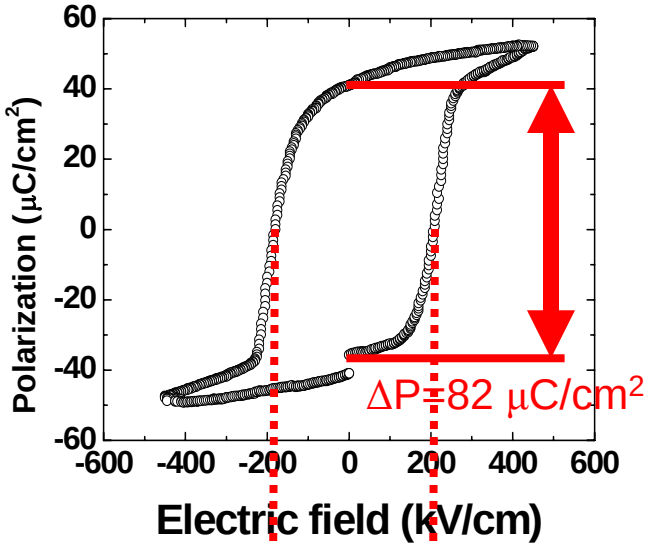
Time Resolved Diffraction During Switching



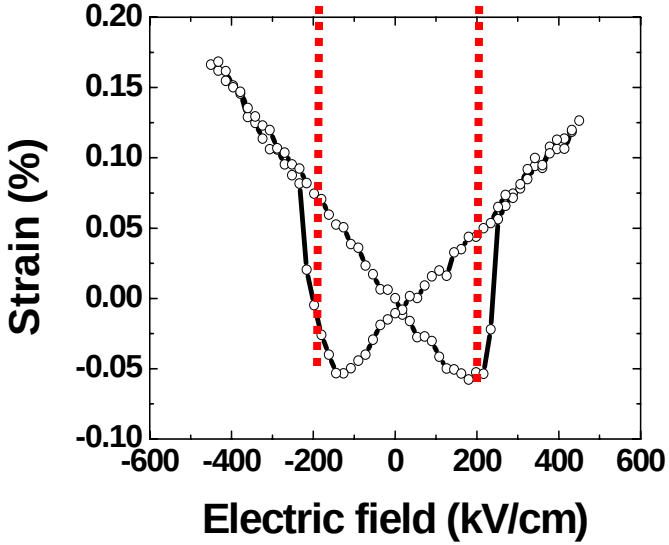
Switching of 200 μm diameter device limited by RC time constant.

Relating electrical and mechanical observations of polarization switching

Electrical hysteresis loop

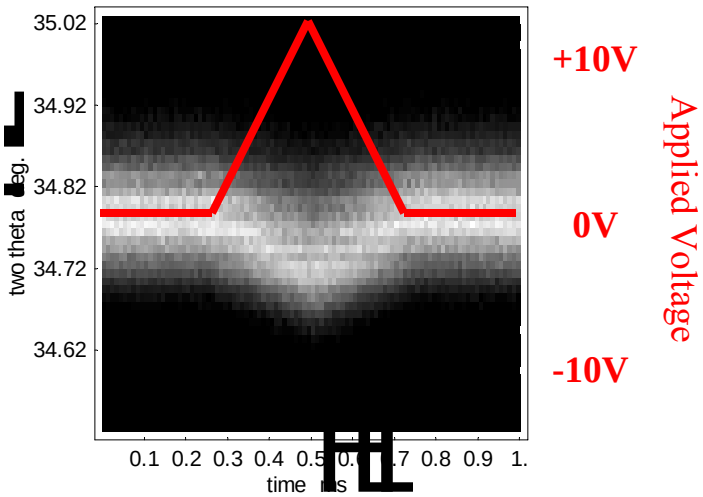


Mechanical strain curve

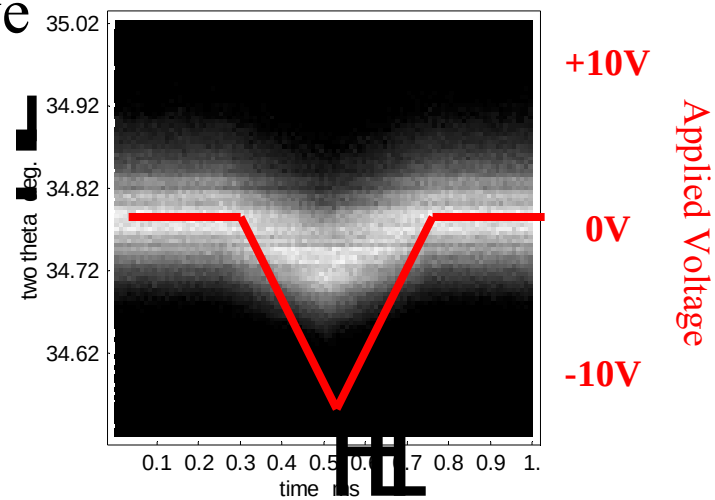


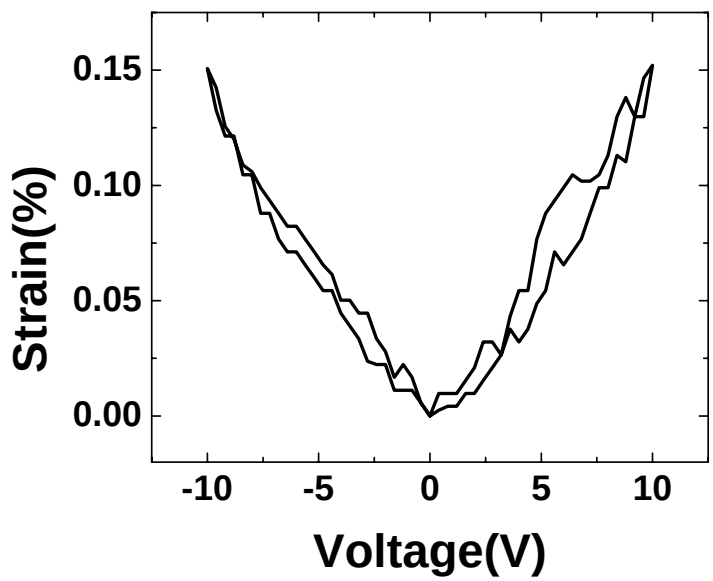
Direct Measurements of Piezoelectric Distortion

Positive pulses



Negative pulses





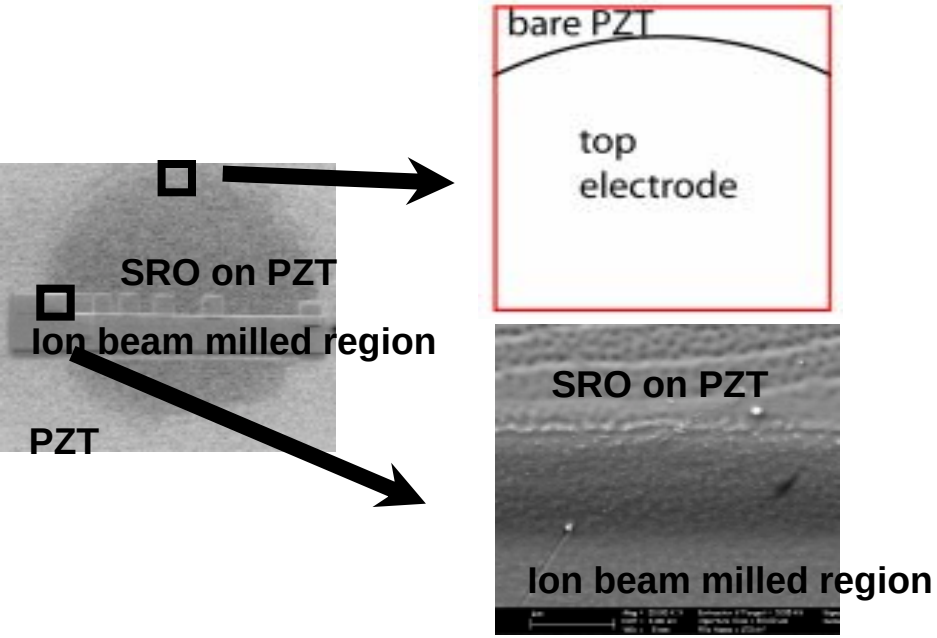
Both polarizations have d_{33}=60 pm V^{-1}.

Quantitative measurement: Requires No Standards.

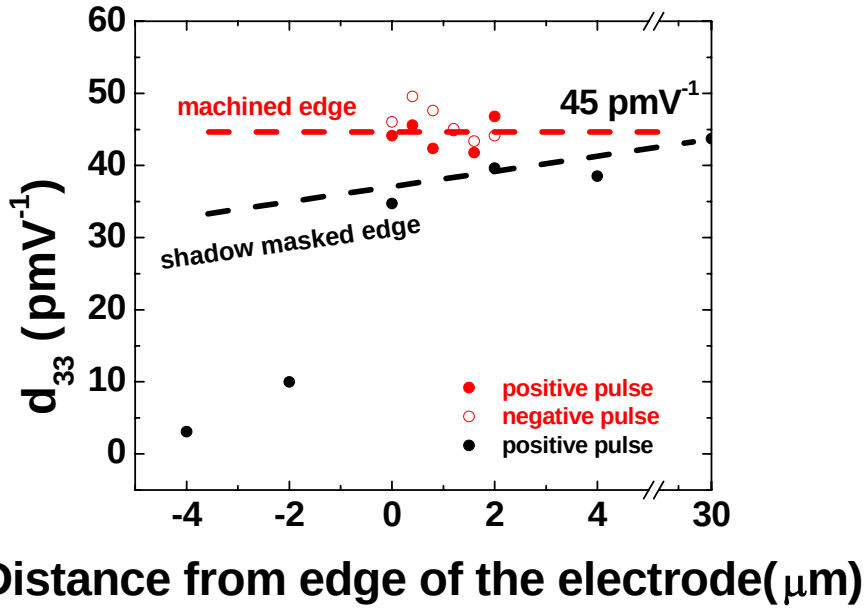
Mechanical boundary conditions are important!

- Piezoelectric displacement can be strongly constrained by either substrate or PZT film around the edge of a capacitor.

shadow masked edge



edge machined with focused ion beam



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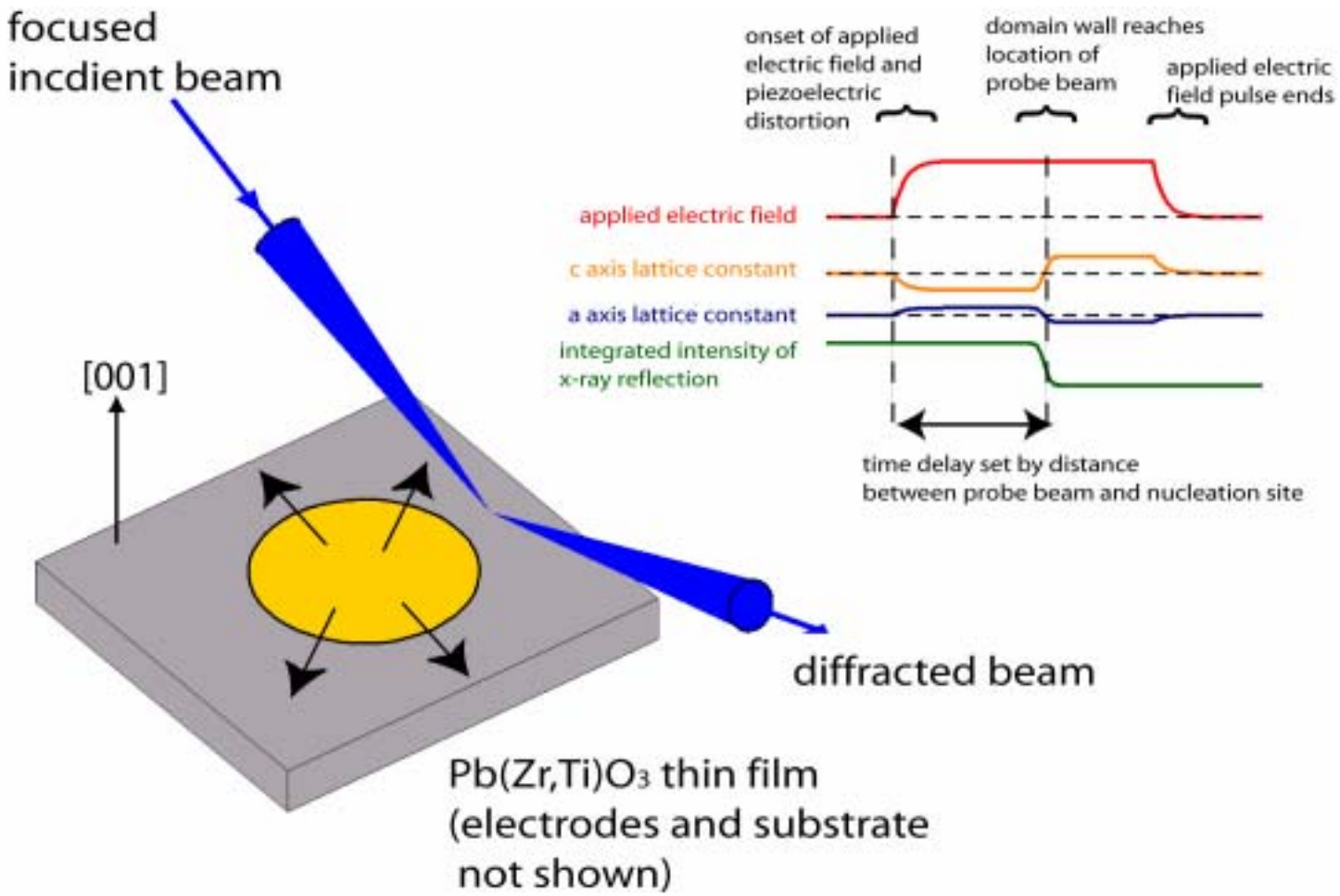
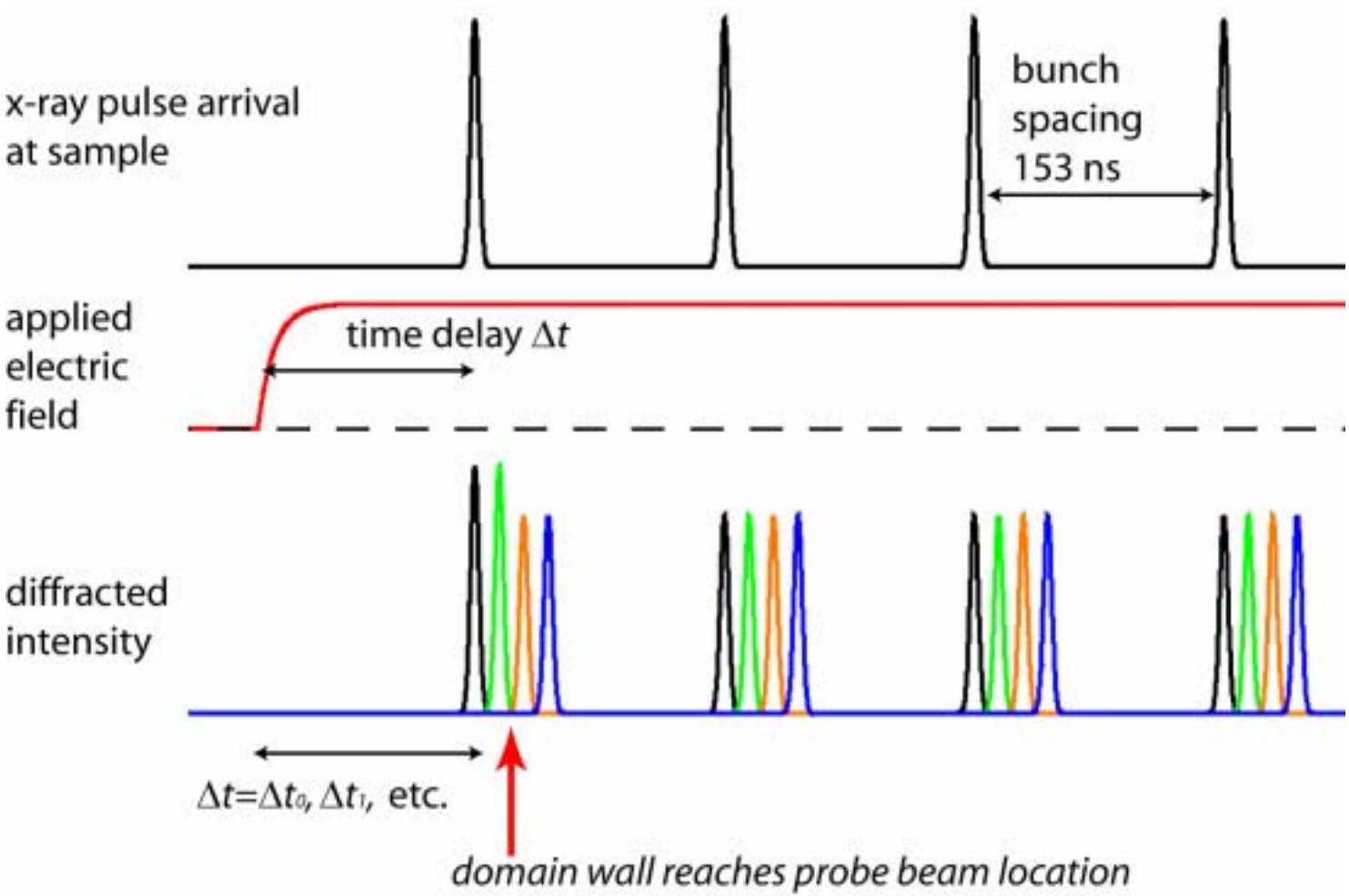
Time Dependent Phenomena in a Switching Capacitor



Proposed Timing Diagram

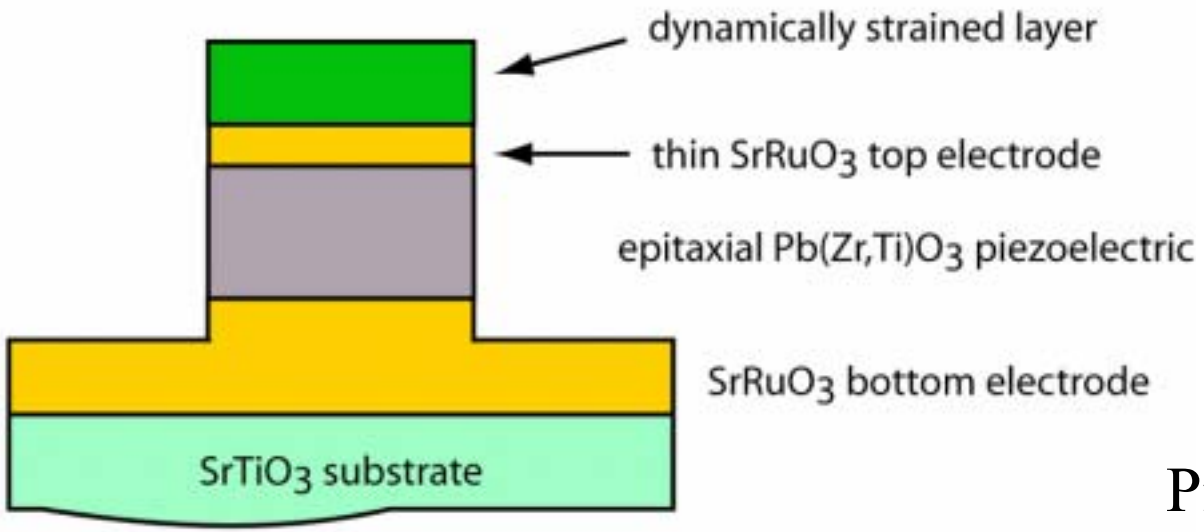


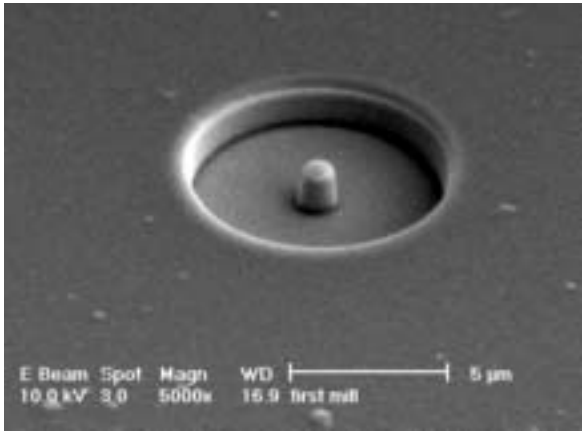
Conclusions: Ferroelectrics

Goals: Quantitative Imaging with Submicron Resolution

1. X-ray microdiffraction provides a new avenue to studying polarization switching in ferroelectric devices.
2. Quantitative agreement of x-ray observations with electrical measurements of switching.
3. Potential to take advantage of the flexibility of x-ray scattering techniques: resonant scattering, time resolved diffraction…

Future:





Pillar formed using FIB.

People

- Ferroelectric thin films
  - X-ray experiments: Dal-Hyun Do (UW), Eric Dufrsense (Adv. Photon Source)
  - PZT thin films: Dong Min Kim and Chang-Beom Eom (UW Materials Science)