Ferroelectric Domain Walls Physics and Function

Lane W. Martin

Department of Materials Science & Engineering, University of California, Berkeley

Materials Sciences Division, Lawrence Berkeley National Laboratory

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Selected References

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Disclaimer: This topic is a rich one! There is not time cover all of it in detail, some great work is thus not included here. No offense is meant, just a result of the time limits! For those new to the field, know there is more beyond these slides to explore...

Question?

Imagine we are walking around the Corsican countryside and find a piece of magnetite (Fe_3O_4) which is magnetic (specifically ferrimagnetic). Could we stick this piece of Fe_3O_4 to the refrigerator?

Why not?

Domains – One of the most important considerations for property evolution in ferroics!

Ferroic Materials

- *Ferroic crystals* posses two or more orientation states or *domains* and under the right driving force, the *domain walls* move, switching the crystal from one domain state to another
    - Force can be stress (X), electric field (E), magnetic field (H), or a combination thereof
- *Ferroelectric* → Spontaneous polarization P_S
- *Ferroelastic* → Spontaneous strain x_s
- *Ferromagnetic* → Spontaneous magnetization M_S
- Not necessary that the orientation states differ in primary quantities (x, P, M) for the appropriate field to develop a driving force for domain motion
    - Example → Domains have different orientations of elastic compliance tensor, suitably chosen stress can produce different strains in 2 domains
    - Same stress may act upon the difference in induced strain to produce wall motion, domain reorientation
    - This is called *ferrobielastic* response as opposed to *ferroelasticity*

What is a Ferroelectric Material?

The Hierarchy of Materials: Symmetry

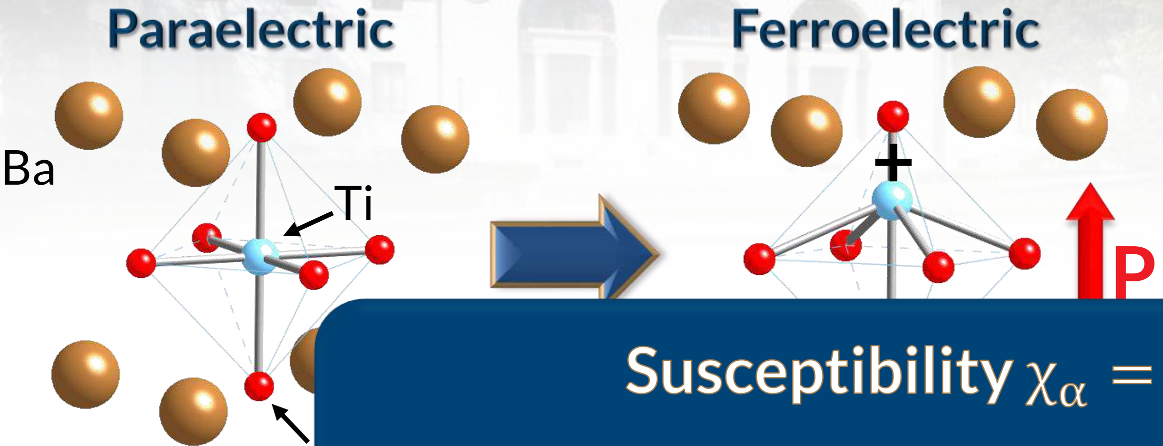
32 Crystal Classes → 11 possess center of symmetry → No polar properties

Of remaining 21 → 20 exhibit polarity when subjected to a stress (*Piezoelectric*)

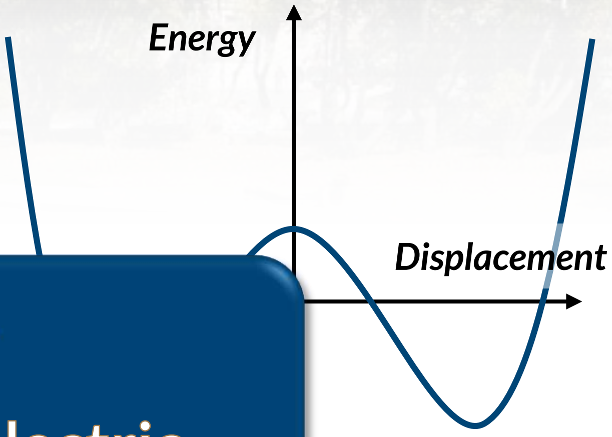
Of these 20 → 10 show unique polar axis → possess spontaneous polarization (*Polar*)

- Temperature dependence → changes in polarization result in flow of charge to-and-from surface (*Pyroelectric*)
- When a *polar* material has two or more orientation states in the absence of an *E* and can be shifted from one to another of these states by an electric field → *Ferroelectric*

What is a Ferroelectric Material?



Switchable Polarization



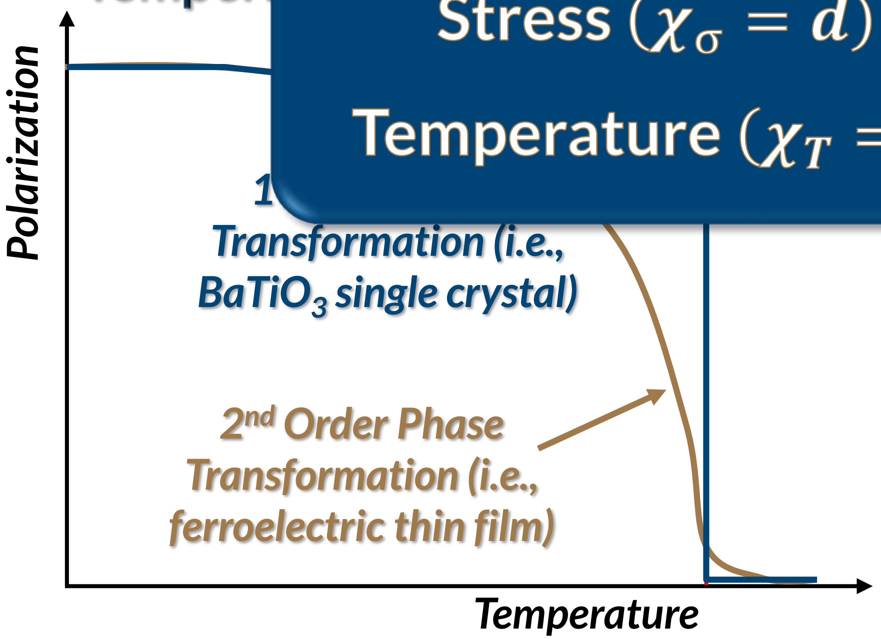
Susceptibility $\chi_\alpha = \frac{dP}{d\alpha}$

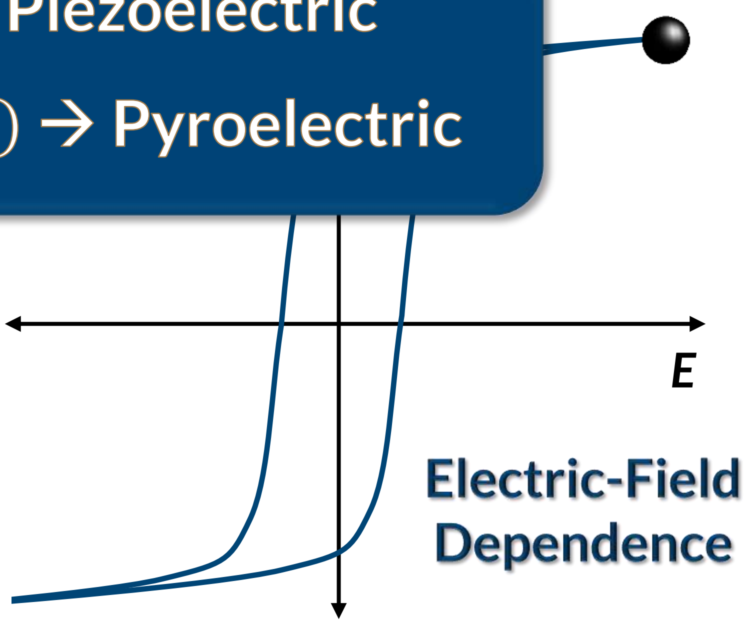
Electric Field ($\chi_E = \varepsilon$) → Dielectric

Stress ($\chi_\sigma = d$) → Piezoelectric

Temperature ($\chi_T = \pi$) → Pyroelectric

Temper

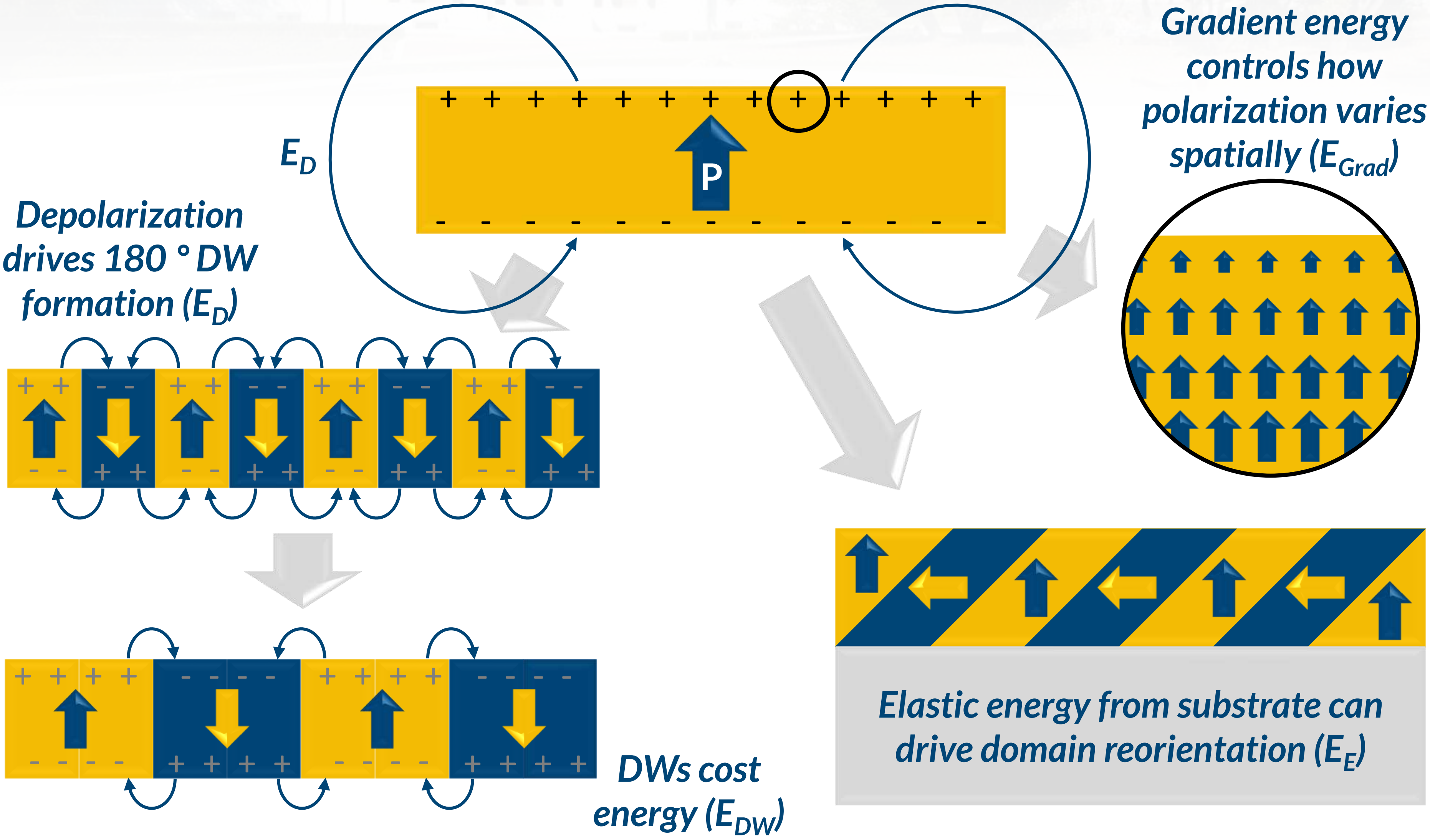




Electric-Field Dependence

Energies in Ferroelectrics

$$E_{Total} = E_P + E_D + E_{DW} + E_{Grad} + E_E$$



Depolarization drives 180 ° DW formation (E_D)

Gradient energy controls how polarization varies spatially (E_{Grad})

DWs cost energy (E_{DW})

Elastic energy from substrate can drive domain reorientation (E_E)

Ferroelectric Domains

- We are concerned with how polarization changes at inhomgeneities…
- Recall that $D = \varepsilon_0 E + P$ and that *P* arises from both the polarizability of the material in the presence of field $P_E = \chi E$ and from the spontaneous alignment of dipoles (P_S)

- If there is a spatial variation of the *D* the free charge density ***ρ*** must satisfy Poisson's Equation → $\nabla D = \rho$ so that $\nabla E = \frac{1}{\varepsilon\varepsilon_0}(\rho - \nabla P_S)$

- In an ideal infinite ferroelectric P_S is uniform so that $\nabla E = \frac{\rho}{\varepsilon\varepsilon_0}$ as in ordinary dielectrics
- At a surface where P_S decreases to zero (or near defects where P_S may differ from perfect xtal) ∇P_S acts as the source for a E_D

- This field can be compensated by the flow of free charge within the crystal → $\rho = \int_0^t \sigma \cdot E dt$ (where ***σ*** is the electrical conductivity)

Ferroelectric Domains

- Alternatively, free charge in the surrounding medium can compensate for the depolarization field at the crystal surface
  - In air this is usually slow compared to crystal conductivity
- Accumulation of surface charge by conduction satisfies the requirement that the *E* vanish both outside the crystal and inside the bulk (but not necessarily just below the crystal surface)
- The energy associated with this depolarization field is $\boldsymbol{W_E = \int_V D \cdot E dV}$
  - Energy is zero for a totally compensated crystal in =m
  - In an insulating xtal (and environment), =m is reached very slowly and at short times, the energy W_E can reach very high values
- When we under go the FE transition, there are at least 2 equivalent directions along which the spontaneous polarization may occur
- In order to minimize W_E different regions of the xtal polarize in each of these directions

Ferroelectric Domains

- Each volume of uniform polarization is a ***domain***

- The depolarizing field (E_D) that arises during cooling from PE phase is usually enough to *prevent any polarization in a virgin crystal*

- Boundaries separating domains are domain walls → they have an energy associated with them (E_{DW})

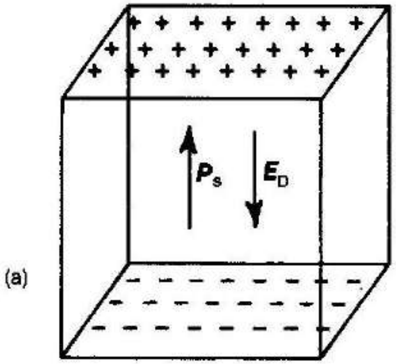
- Final domain configuration is determined by minimizing an appropriate free energy including both of these energy terms

- Final domain structures rely on things like…

crystal symmetry | electrical conductivity | defect structure
magnitudes of the polarization and elastic and dielectric compliances
history of the crystal preparation | sample geometry | etc.

Ferroelectric Domains

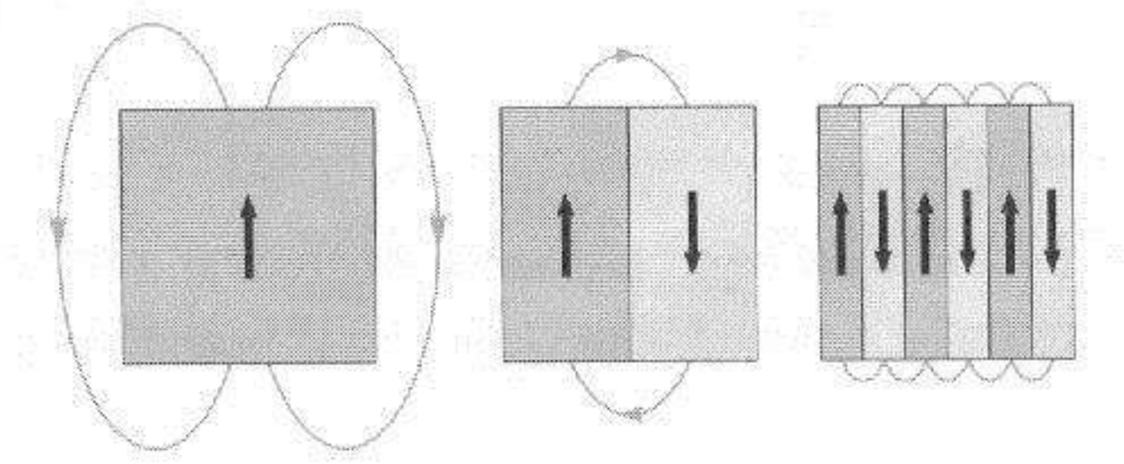
- The onset of spontaneous polarization results in the appearance of surface charge density and an accompanying depolarization field (E_D)



- Thus we can write $E_{Tot} = E_{Ext} + E_D$ where E_D points opposite to E_{Ext}
- The energy associated with the P in the E_D can be minimized by driving the crystal into regions of oppositely polarized regions → Domains
- Domains result in a reduction in E_D and in electrostatic energy for the system, but cost the energy to create a domain wall (generally 180° domain walls)

Ferroelectric Domains

- Configuration of domains follows a head-to-tail condition in order to avoid discontinuities in the polarization at the domain boundary → $\nabla\vec{P} = \sigma$



- Build-up of domain walls, elastic stress fields, and free charge carriers all counteract the process of domain formation (don't forget vacancies, defects, dopants, etc.)
- A single domain structure can be achieved by applying the right *E* → domains aligned along the E direction grow at the expense of the others

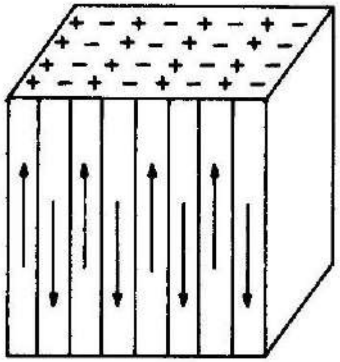
Ferroelectric Domains: Free Energy

- Static domain configuration of a FE is obtained by minimizing the total free energy of the crystal including the energy associated with the crystal surfaces and domain walls
- We can write: $G_1 = G_1^0 + \int \left(\frac{\alpha}{2} D^2 + \frac{\gamma}{4} D^4\right) dV + W_W + W_E$
  - Where we neglect higher order powers

- **Depolarization Energy (W_E)**
  - To minimize the depolarization energy at the crystal surfaces before compensation by free charge takes place domains form throughout the volume of the crystal
  - Without domain formation the energy associated with the crystal surfaces can be greater than the energy associated with the ferroelectric ordering → could turn FE order off!
  - Magnitude of W_E depends on the crystal and domain geometry

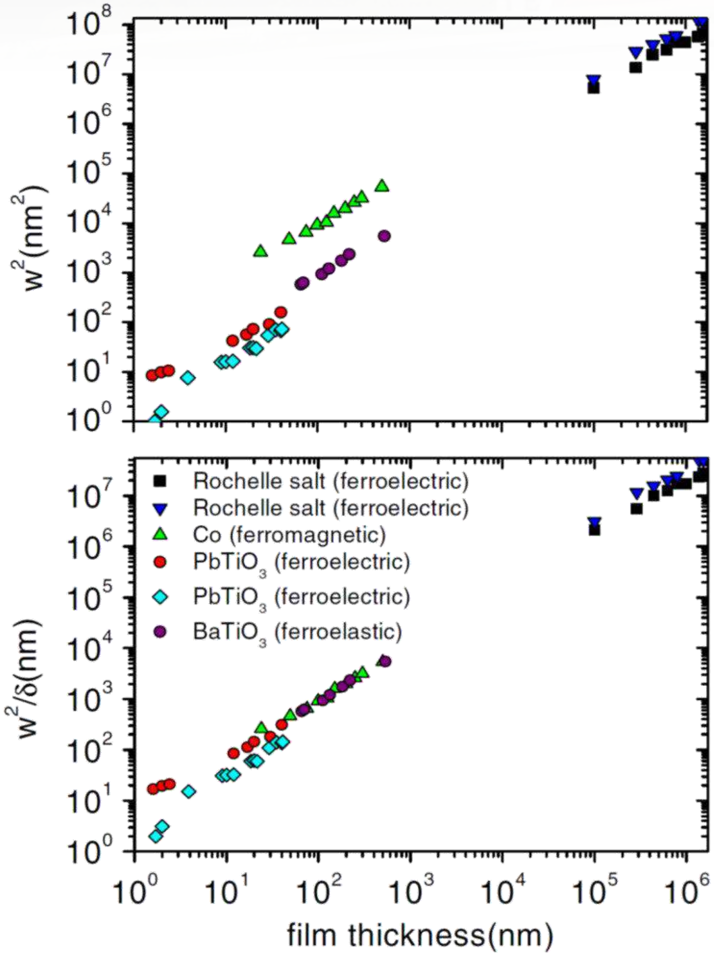
Ferroelectric Domains: Free Energy

- **Domain Wall Energy (W_W)**
  - If the *energy per unit area of a domain wall* is **σ** then for a domain geometry like this → $W_W = (\sigma/_d)V$
  - Minimizing the energy $W_E + W_W$ yields the =m value of the domain width as $\boldsymbol{d} = \left(\frac{\sigma t}{\varepsilon^* P_0^2}\right)^{1/2}$ where P_0 must be obtained from self-consistent solution free energy
  - Domains of *d* greater than this are suppressed by the W_E, domains smaller are suppressed by the W_W
  - *d* varies quadratically with thickness → thickness ↓, *d* approaches domain thickness → W_E can no longer be minimized by domain formation, FE may cease to exist
  - Contributions to W_W →
    - W_E due to the ∇P at domain boundaries
    - Dipolar energy (misalignment of dipoles on either side of DW)
    - Elastic energy



Ferroelectric Domains: Kittel’s Law

- Kittel’s law → Domain width $w = \sqrt{\frac{\sigma}{U}d}$ where σ is the energy density per unit area of the wall, U is the volume energy density of the domain, and d is the thickness



Catalan *et al.*, *J. Mater. Sci.* **44**, 5307 (2009)

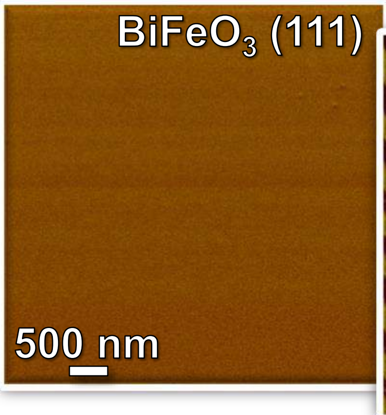
- Comparisons between stripe domains of different ferroic materials shows…
  - All of them scale with the same ***square root dependence*** of domain width on film thickness;
  - Kittel’s law holds true for ferroelectrics down to small thickness;
  - When the square of the domain size is normalized by the domain-wall thickness, the different ferroics fall on the same curve

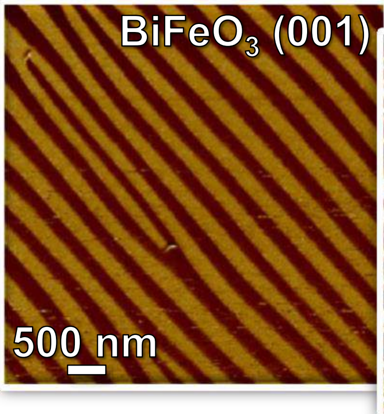
One can engineer domain structures (periodicities) with film thickness

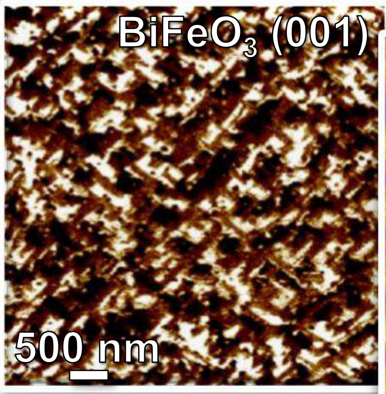
Ferroelectric Domains: Result

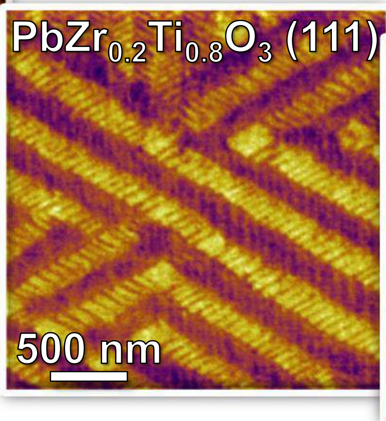
Combining all effects…

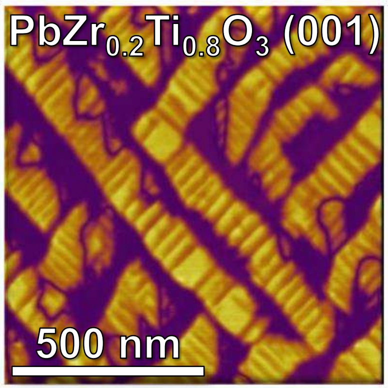
- Domain-wall thickness: 1-10 nm (2-20 unit cells) → 100s nm in FM
- Energy of domain wall: ~10 erg/cm^2 (10^{-6} J/cm^2) → 1-10 erg/cm^2 in FM
- FE domain walls are much less wide than magnetic domain walls →
  - Magnetic exchange energy is much larger than the elastic energy and slow rotation of the magnetization vector occurs over hundreds of unit cells
  - Magnetization magnitude remains constant, FE polarization reduces to zero in some domain walls (paraelectric character)









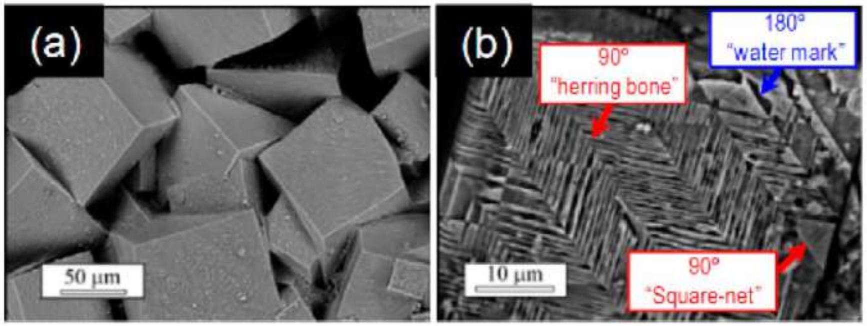


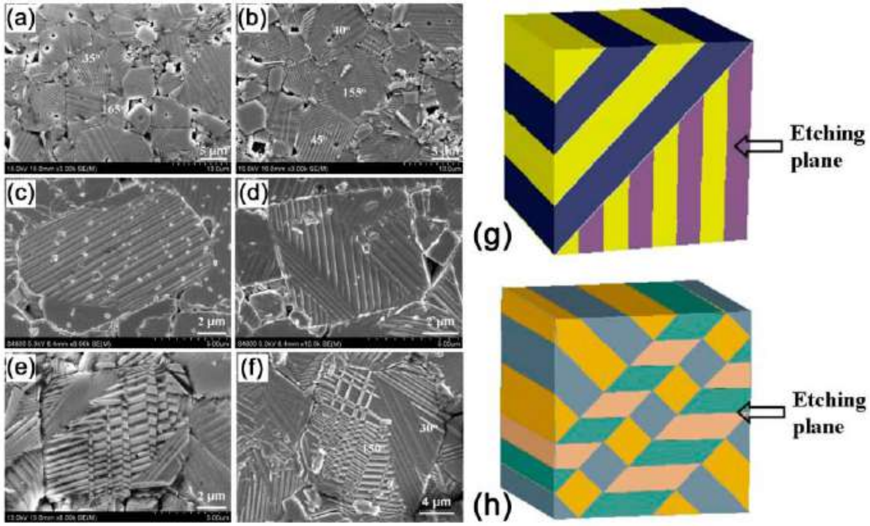
All films ~ 100 nm thick, all with $SrRuO_3$ bottom electrodes

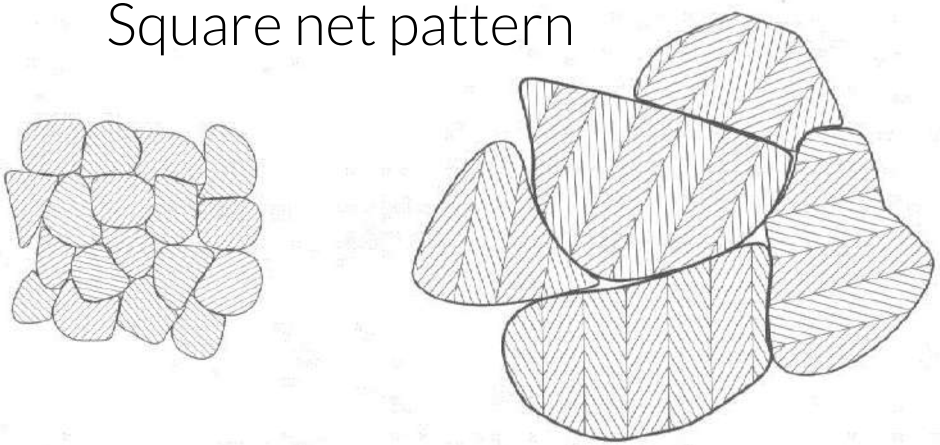
Ferroelectric Domains

- Polycrystalline, bulk ceramics → pattern of domains is quite different because the domain structure in each grain is formed under elastic, clamped conditions by surrounding domains
- Domain Wall Types in Single Xtals
  - Tetragonal – 90° (ferroelastic), 180° (ferroelectric)
  - Rhombohedral – 71°, 109° (ferroelastic), 180° (ferroelectric)
- Domain Structures in Poly-Xtals
  - Herringbone (more common) and Square net pattern

Alikin *et al.*, *Materials* 10, 47 (2017)





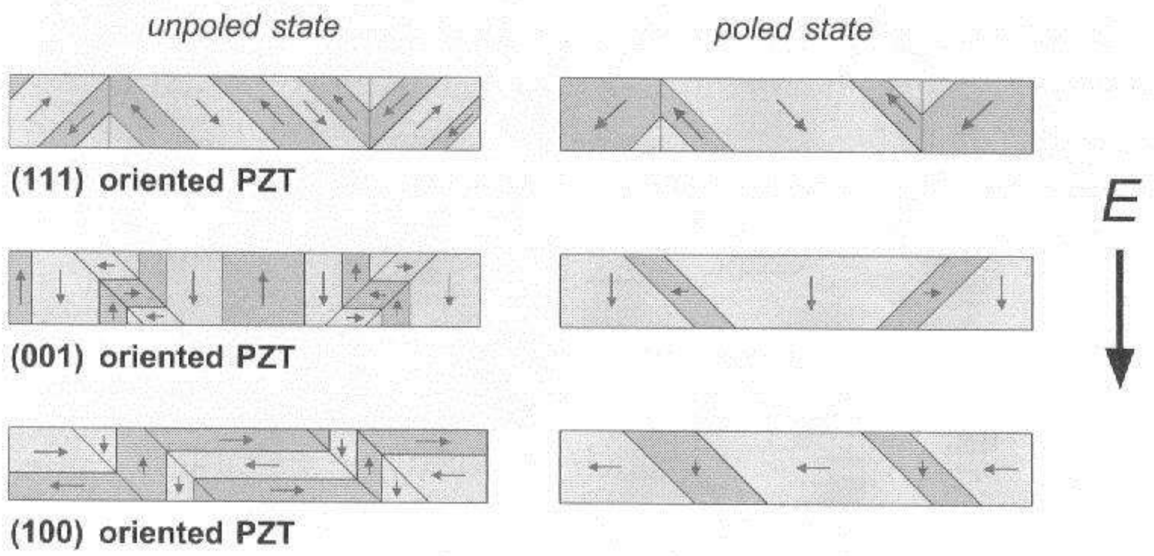


Ferroelectric Domains

Thin Film Domain Structures

- Ultra-thin films – Single-domain

- Films >~100 nm – Polydomain
  - Example tetragonal $PbZr_{1-x}Ti_xO_3$

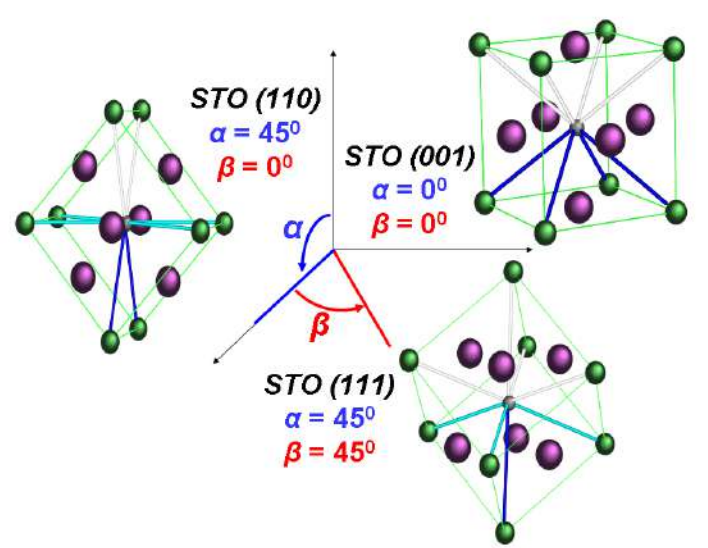


  - Varies with film/substrate orientation, applied field, etc.
  - Predominant structure consists of 90°

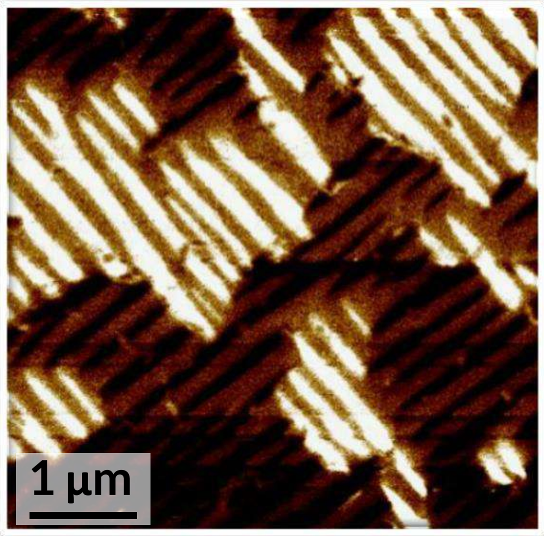
Ferroelectric Domains

Thin Film Domain Structures

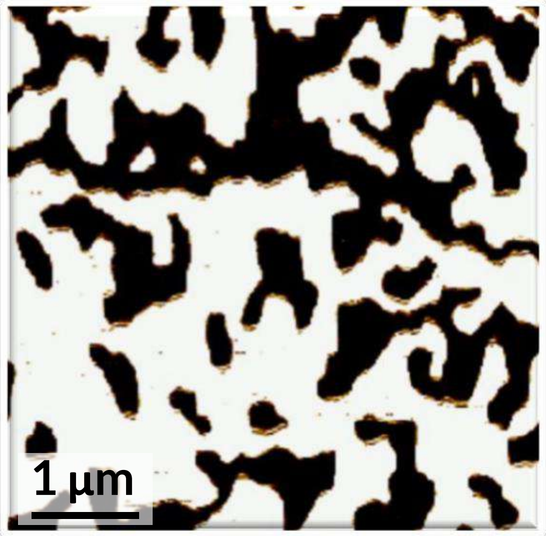
- Example: rhombohedral $BiFeO_3$
  - (001) – Predominantly 71° domain walls (could have all types)
  - (110) – Predominantly 71° domain walls (could have all types)
  - (111) – Can only have 180° domain walls



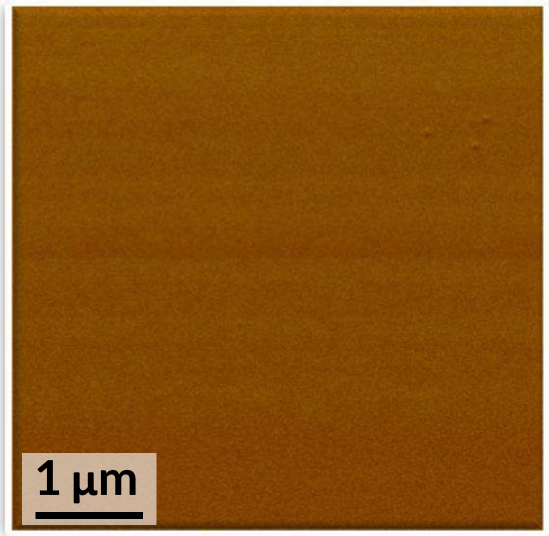
$BiFeO_3$ / $SrTiO_3$ (001)



$BiFeO_3$ / $SrTiO_3$ (110)



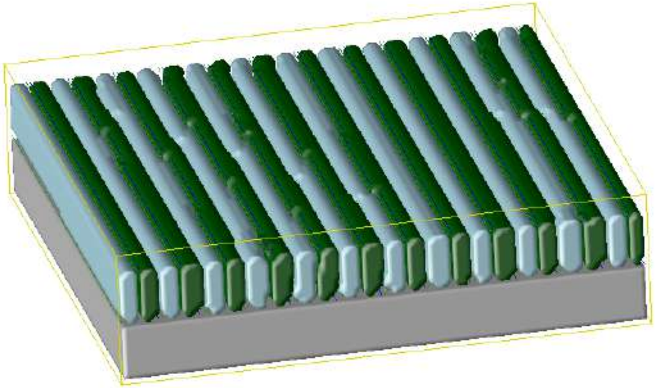
$BiFeO_3$ / $SrTiO_3$ (111)



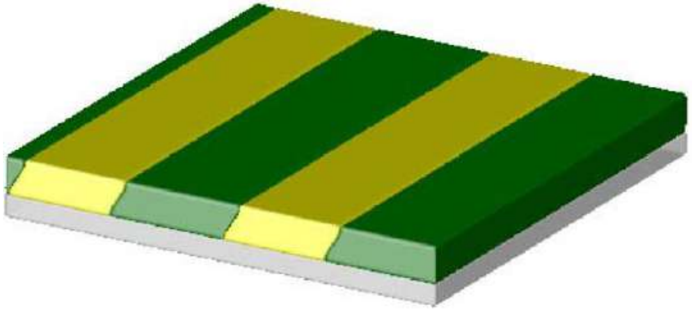
Controlling Ferroelectric Domain

Interplay between Strain and Electrostatics

Phase Field Models
(Yulan Li, Long-Qing Chen,
Penn State)

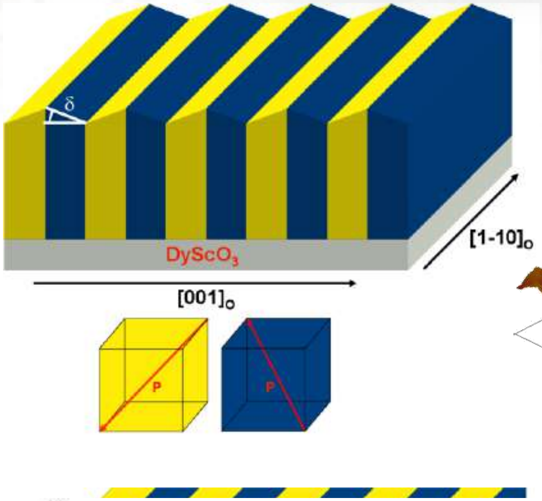


Open circuit boundary condition

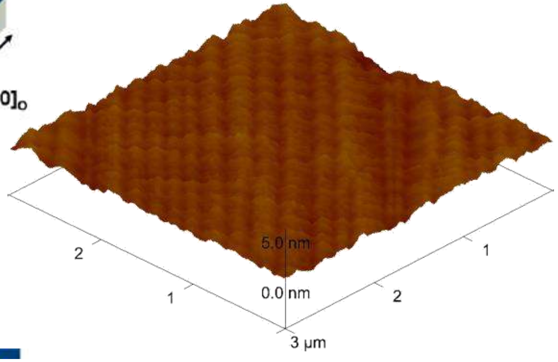


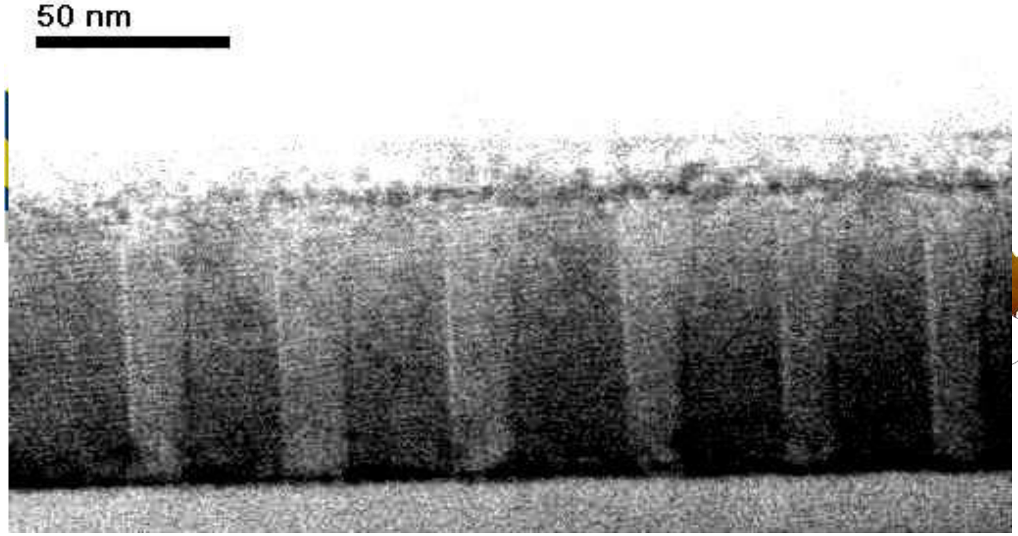
Short circuit boundary condition

S. K. Streiffer, *et al. J. Appl. Phys.* 83, 2742 (1998)



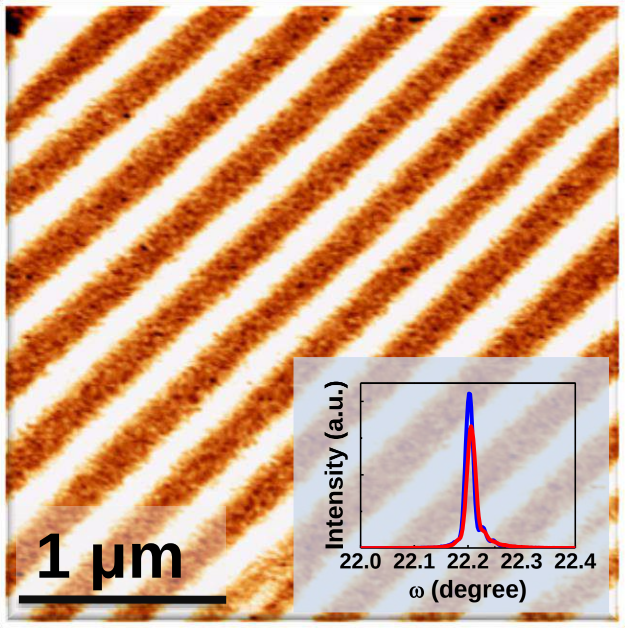
109° Domain Wall Arrays

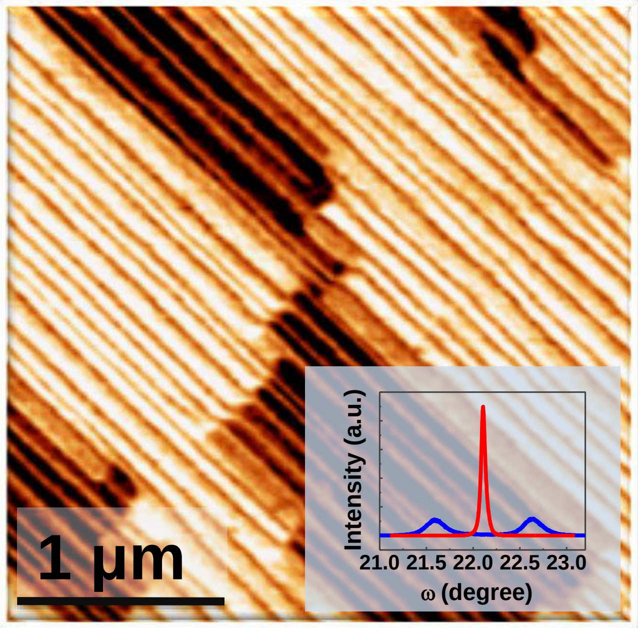




Chu *et al.*, *Nano Lett.* 9, 1726 (2009)

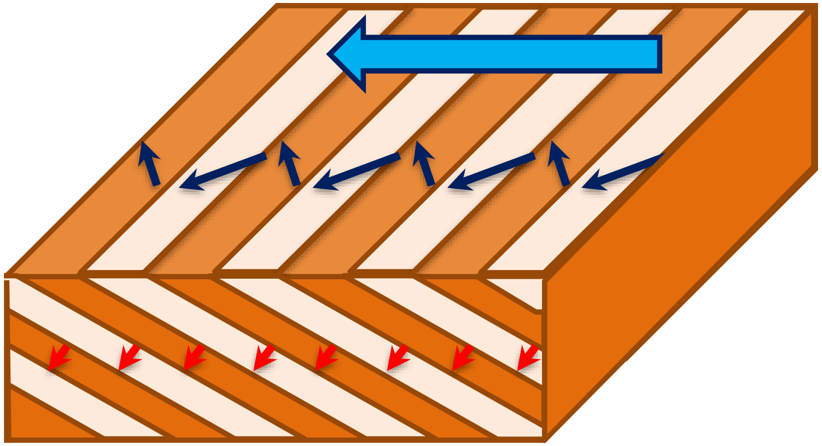
Model Systems – Ordered DW Arrays





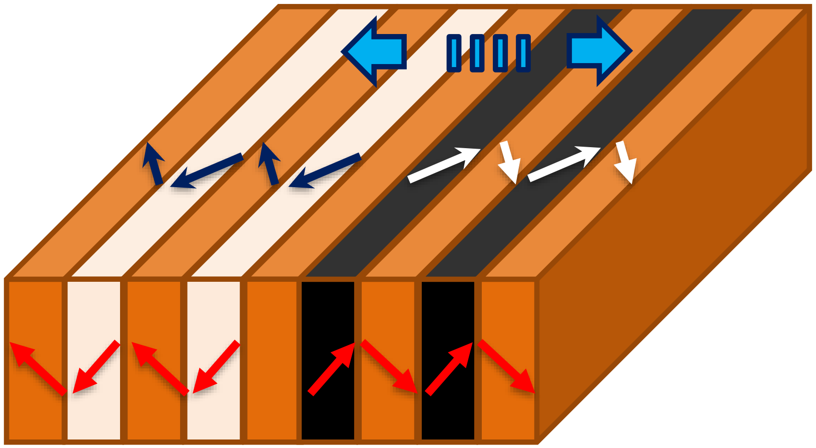
71° Domain Walls

$\vec{P}_{net}$



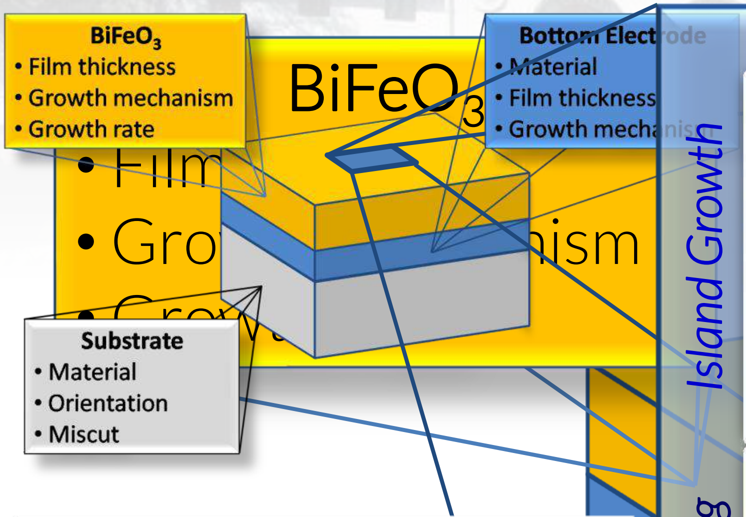
109° Domain Walls

$\vec{P}_{net}$



Yang *et al.*, *Nature Nanotechnol.* 5, 143 (2010)

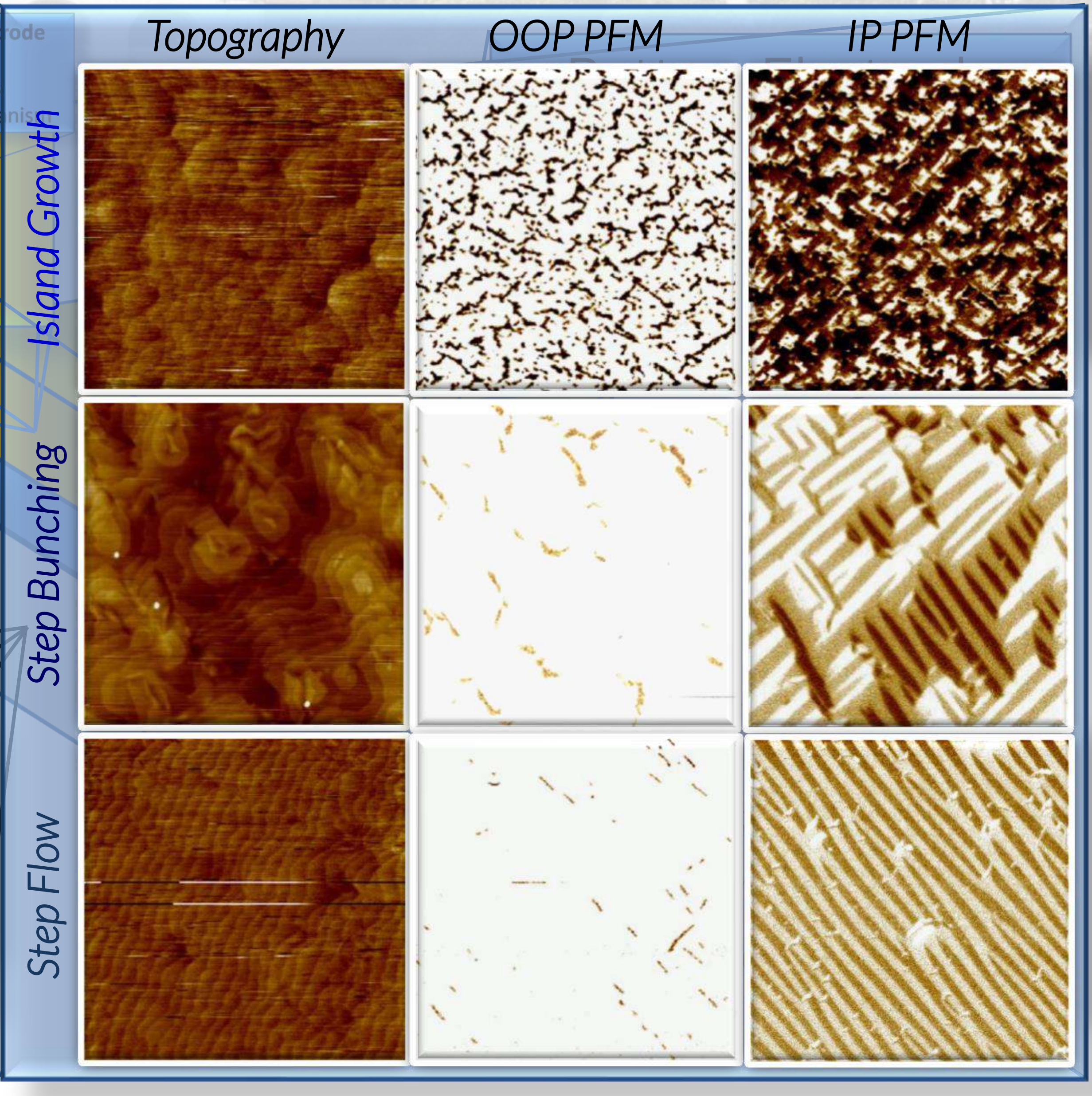
Engineering Domain Structures: BiFeO$_3$



BiFeO$_3$
- Film thickness
- Growth mechanism
- Growth rate

Bottom Electrode
- Material
- Film thickness
- Growth mechanism

Substrate
- Material
- Orientation
- Miscut



Controlling domain structures in BFO

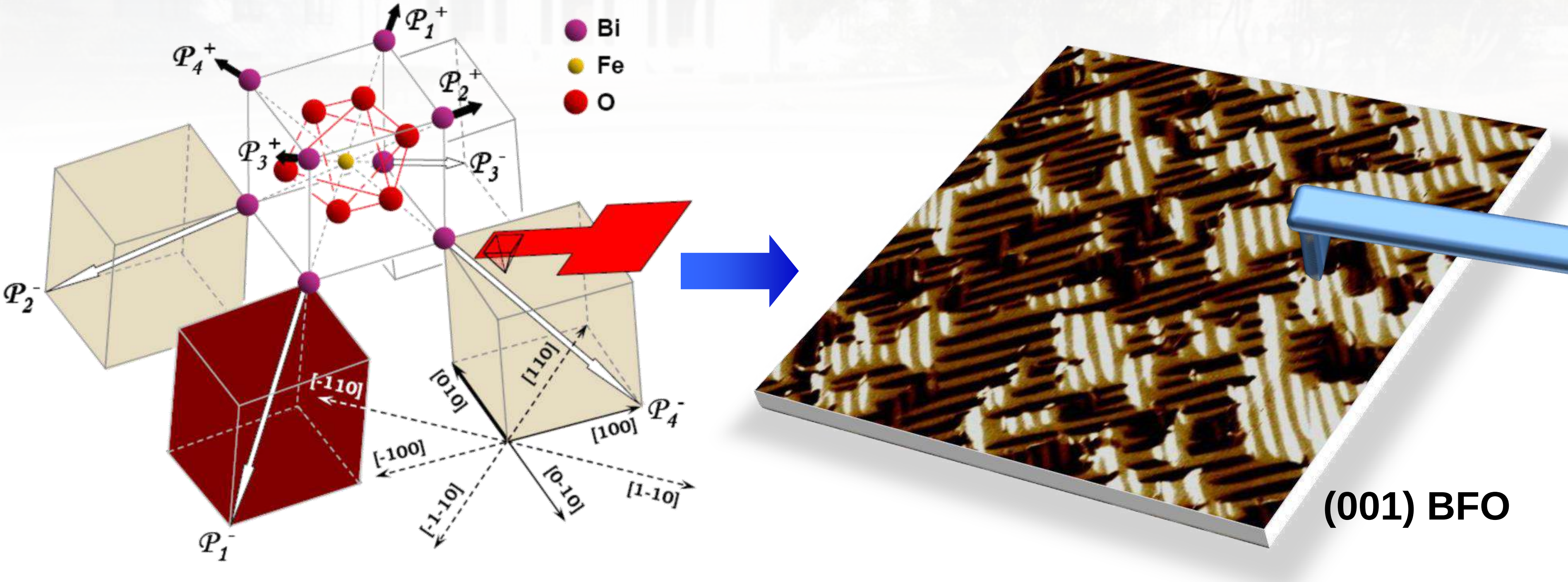
- BiFeO$_3$ – Growth rate, underlying bottom electrode structure
- SrRuO$_3$ – Structural variants
- Substrates - vicinality

• Miscut

Chu *et al.*, *Adv. Mater.* **18**, 2307 (2006); *Adv. Mater.* **19**, 2662 (2007)

Imaging Ferroelectric Domains

Piezoresponse Force Microscopy (PFM)

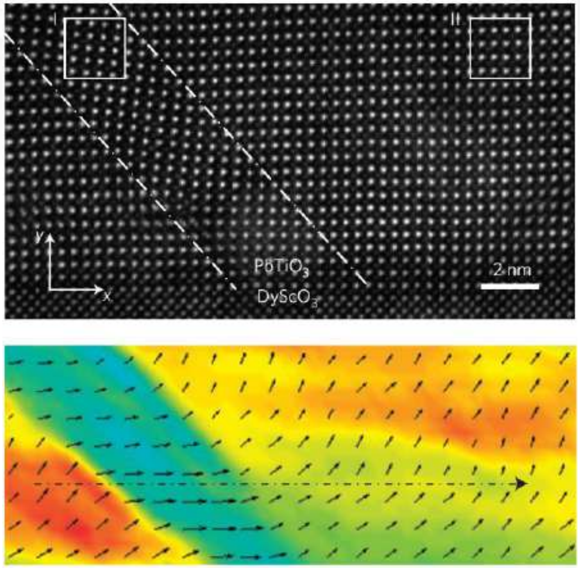


OOP Contrast	IP Contrast		Possible Orientations
	[010]	[0-10]	
Dark (up)	Dark (left)	Light (right)	[-111], [-1-11]
Light (down)	Light (right)	Dark (left)	[1-1-1], [11-1]

S. Kalinin, A. Gruverman, *Scanning Probe Microscopy* (Vol II), Springer: New York (2007); Kalinin *et al.*, *IEEE Trans. Ultrason. Ferroelect. Freq. Cont.* **53**, 2226 (2006); Soergel *J. Phys. D. Appl. Phys.* **44**, 464003 (2011); Gruverman *et al.*, *Nature Commun.* **10**, 1661 (2019)

Direct Imaging of Domain Walls

(Scanning) Transmission Electron Microscopy (STEM)

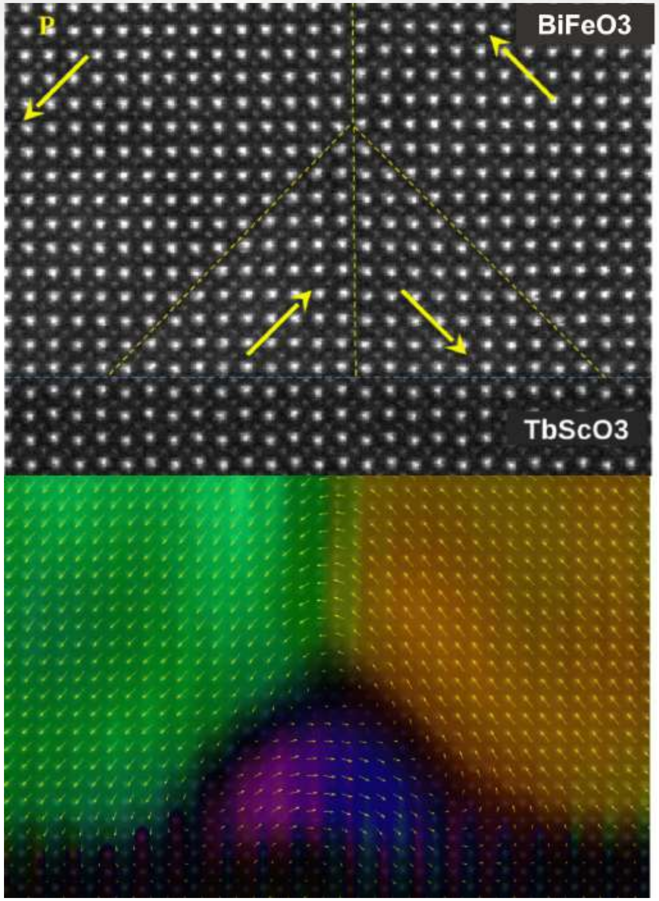


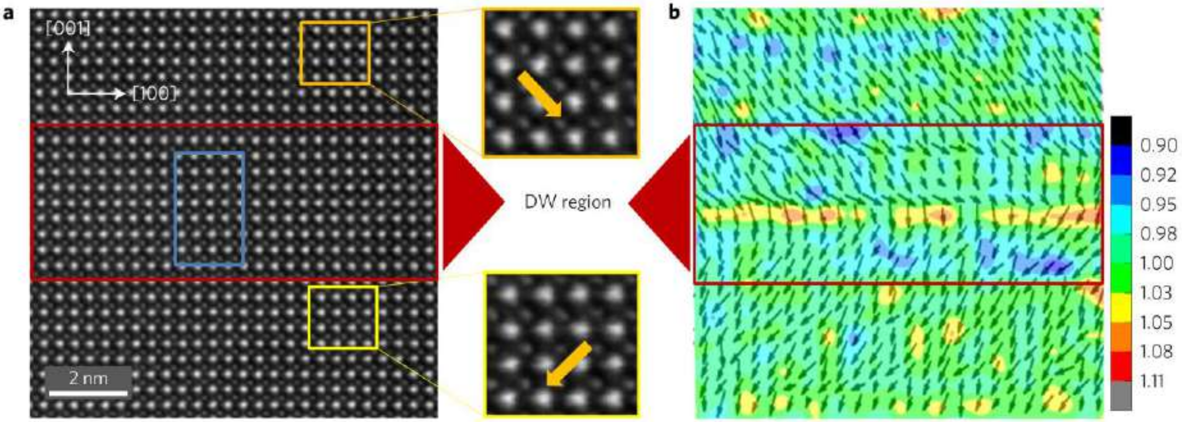
$PbTiO_3$ → Strain gradients (flexoelectricity) @ domain walls results in polarization rotation

Catalan *et al.*, *Nature Mater.* **10**, 963 (2011)

$BiFeO_3$ → Atomic resolution polarization mapping, unusual nanodomain structures

Nelson *et al.*, *Nano Lett.* **11**, 828 (2011)





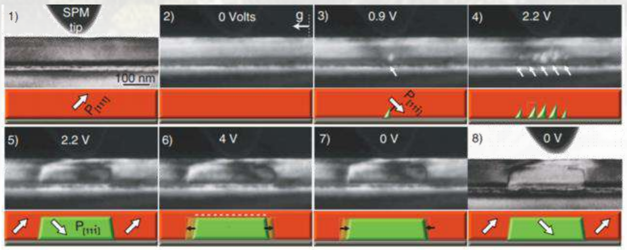
$BiFeO_3$ → Direct mapping of defects/charge accumulation at domain walls that results in domain-wall conductivity

Rojac *et al.*, *Nature Mater.* **16**, 322 (2017)

In operando Studies of Domains

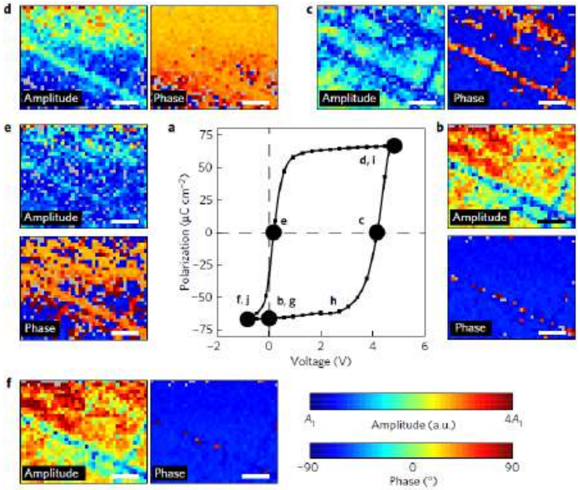
- STEM → Can probe ferroelectrics at the timescales of real processes important for device operation; increasing frame rates, data-acquisition rates during high-resolution imaging to probe ferroelectric switching

Nelson *et al.*, *Science* **334**, 968 (2011); Gao *et al.*, *Nature Commun.* **2**, 591 (2011); Winkler *et al.*, *Nano Lett.* **14**, 3617 (2014)

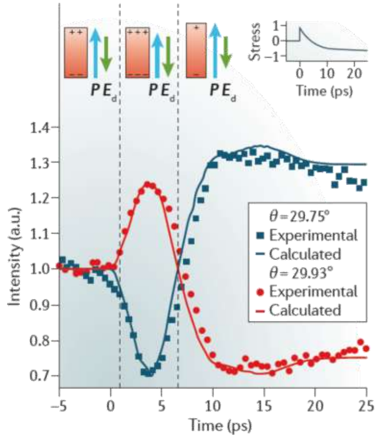


- PFM → Band excitation is changing the way FEs are studied under bias; real-time, *in operando* investigations of FE switching with nanoscale resolution and with the quantification and extraction of extensive data

Jesse, Kalinin *J. Phys. D* **44**, 464006 (2011); Agar *et al.*, *Nature Mater.* **15**, 549 (2016)



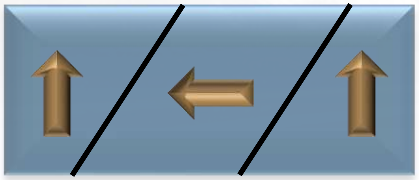
- Synchrotron → Undergoing rapid development; bright-light sources enable *in operando* studies of devices under applied fields, study of ultrathin films, the evolution of FEs under environmental conditions, and ultrafast probing of dynamic responses

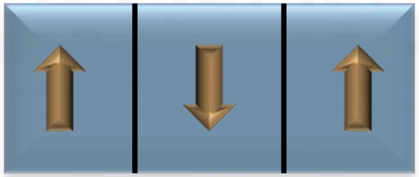


Evans *et al.*, *Am. Ceram. Soc. Bull.* **92**, 18 (2013); Do *et al.*, *Nature Mater.* **3**, 365 (2004); Fong *et al.*, *Phys. Rev. Lett.* **96**, 127501 (2006); Grigoriev *et al.*, *Phys. Rev. Lett.* **96**, 187601 (2006); Highland *et al.*, *Phys. Rev. Lett.* **105**, 167601 (2010); Jo *et al.*, *Phys. Rev. Lett.* **107**, 055501 (2011); Highland *et al.*, *Phys. Rev. Lett.* **107**, 187602 (2011); Daranciang *et al.*, *Phys. Rev. Lett.* **108**, 087601 (2012); Wen *et al.*, *Phys. Rev. Lett.* **110**, 037601 (2013); Hruszkewycz *et al.*, *Phys. Rev. Lett.* **110**, 177601 (2013);

Domain-Wall Contributions to Properties

Domain Walls in Ferroelectrics


90° DW


180° DW

$$\chi_\alpha = \frac{d\langle P\rangle}{d\alpha} = \frac{d(\phi_c P_s)}{d\alpha}; \alpha = T, E, \sigma$$

$$= \phi_c \frac{dP_s}{d\alpha} + P_s \frac{d\phi_c}{d\alpha}$$

Intrinsic **Extrinsic**

Domain wall displacement → Extrinsic contribution to χ

Total Pyroelectric Coefficient (π)

$$\pi = \pi_i + \pi_e + \pi_s$$

Intrinsic π → ΔT gives rise to ΔP$_s$ $\phi_c\left(\frac{dP_s}{dT}\right)$

Extrinsic π → Temp. dependent movement of domain walls $P_s\left(\frac{d\phi_c}{dT}\right)$

Primary Pyroelectric Effect

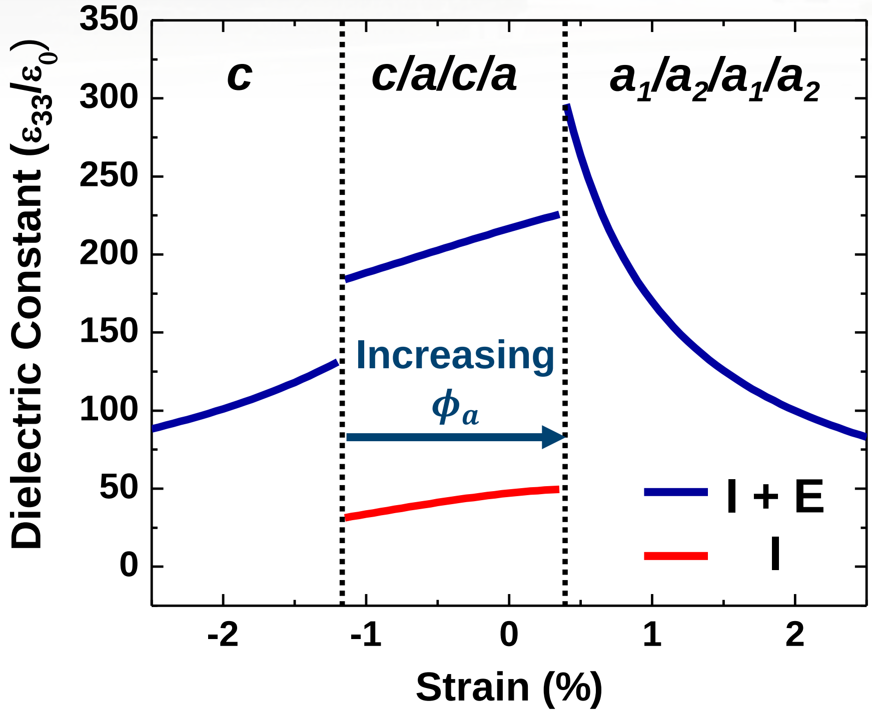
Secondary π → Diff. in thermal expansion between film/sub.
$\propto d_{31}(\alpha_f - \alpha_s)$

Zook and Liu, *J. Appl. Phys.* **49**, 4604 (1978)

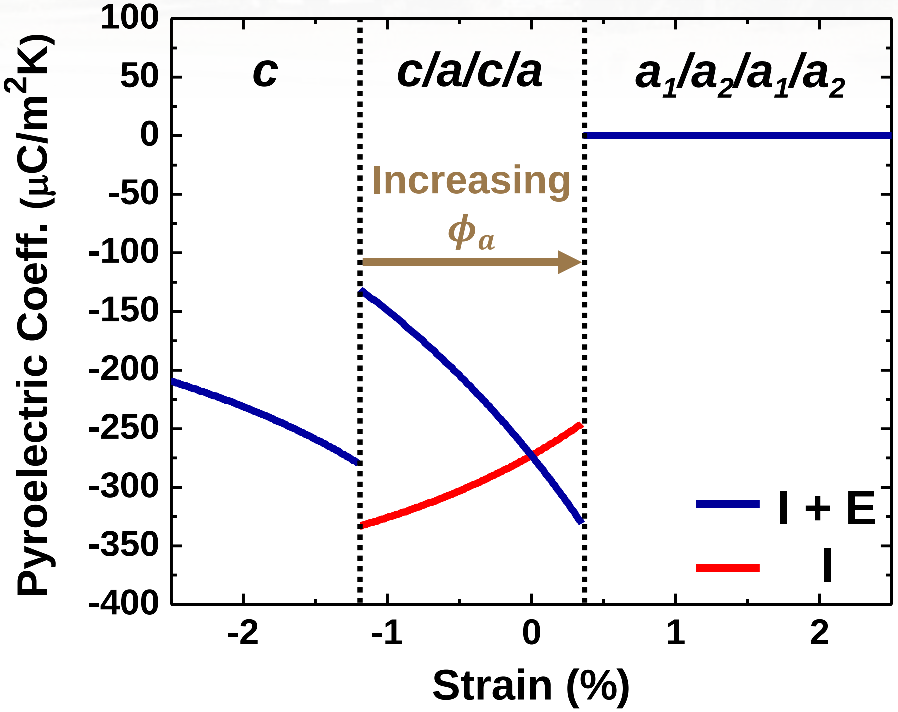
Very little work on extrinsic contributions to π

DW Effects in $PbZr_{0.2}Ti_{0.8}O_3$ Thin Films

Dielectric permittivity $\varepsilon = \left(\frac{dP}{dE}\right)_T$



Pyroelectric coefficient $\pi = \left(\frac{dP}{dT}\right)_E$



Models suggest that 90° domain walls…

- **Increase dielectric and pyroelectric response**
- **Can dramatically change nature of response**

Karthik *et al.*, *Phys. Rev. Lett.* **108**, 167601 (2012)

Karthik *et al.*, *Phys. Rev. Lett.* **109**, 257602 (2012)

DW Effects in $PbZr_{0.2}Ti_{0.8}O_3$ Thin Films

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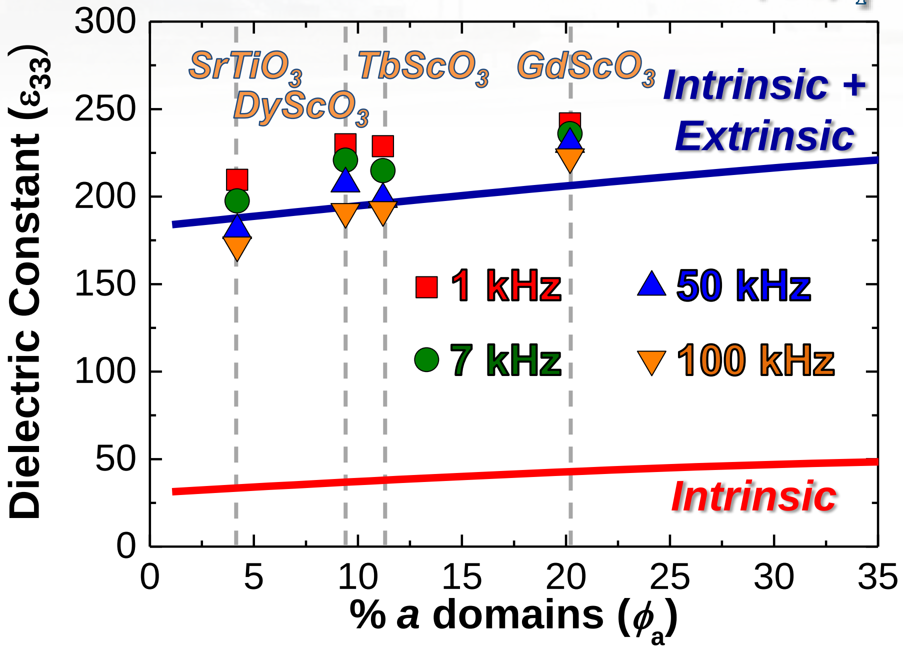
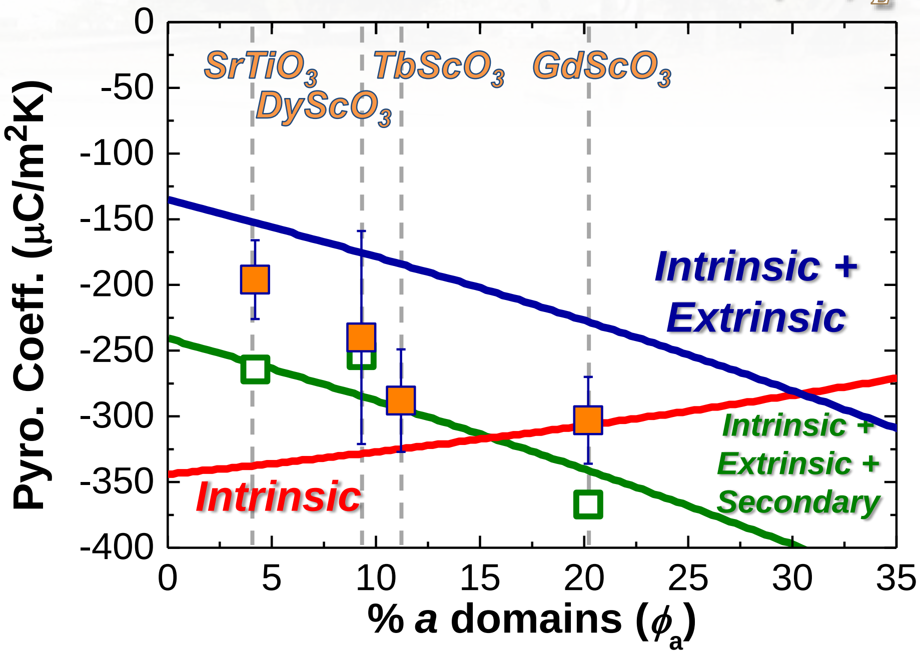


Karthik *et al.*, *Phys. Rev. Lett.* **108**, 167601 (2012)

Pyroelectric coefficient $\pi = \left(\frac{dP}{dT}\right)_E$

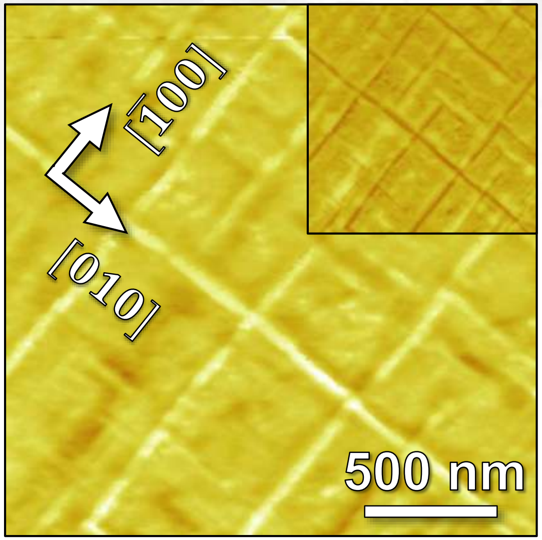


Karthik *et al.*, *Phys. Rev. Lett.* **109**, 257602 (2012)

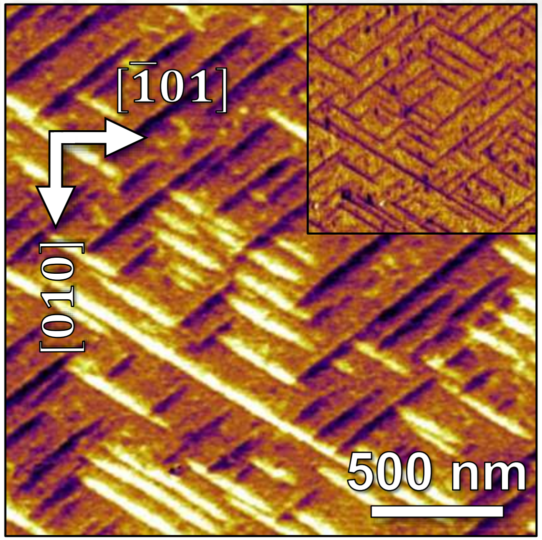
- Optimization of χ_α requires knowledge of domain structure
- 90° domain walls → impact the evolution of properties
- Subtle (but important) differences btw *E* & *T* susceptibilities

Orientation & Domains: $PbZr_{0.2}Ti_{0.8}O_3$

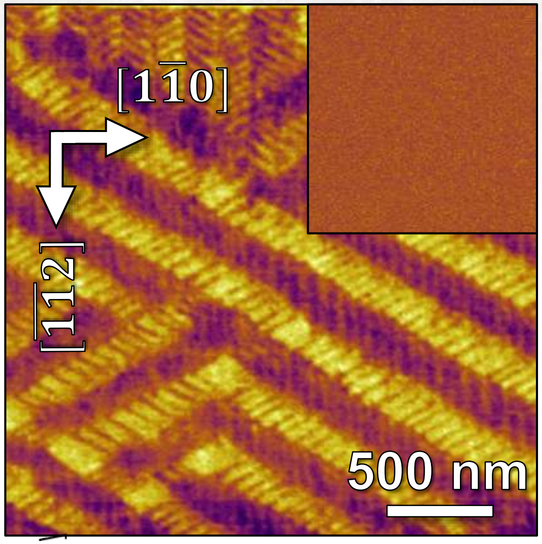
(001)



(101)



(111)



- Orientation provides a knob by which we can control the domain structure of materials
- All samples possess 90° domain walls → controlled structures
- Advantage of thin films → direct observation and quantification of domain structure features

Volume fraction of minority domains

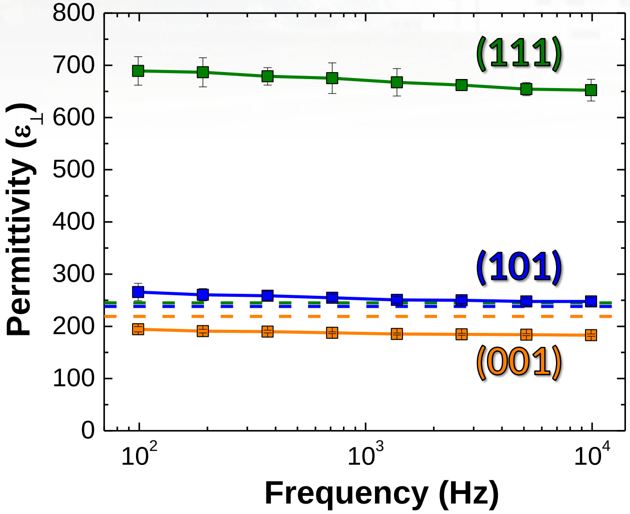
Line density of domain walls

Orientation	λ (µm^{-1})	ϕ (%)
(001)	8.91	15.3
(101)	16.3	19.9
(111)	48.9	33.3

Xu *et al.*, *Nature Commun.* **5**, 3120 (2014)

Exotic Low-/High-Field Effects

Stationary (Frozen) Permittivity

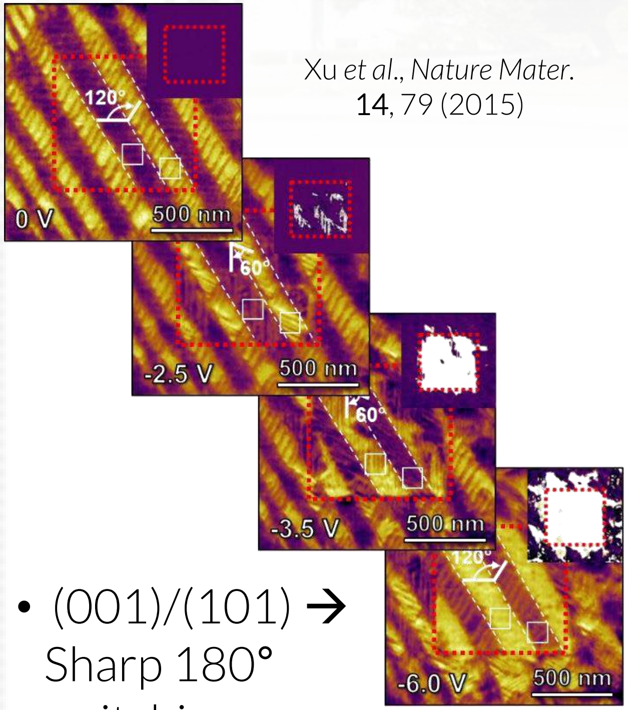


Xu *et al.*, *Nature Commun.* **5**, 3120 (2014)

- Response of the volume of the ferroelectric material within the finite width of the domain walls (non-motional)

ε_{dw} ≈ 1,500-19,000 (for 1-10 nm) → 6-78x ε_b

Multi-Step Polarization Reversal

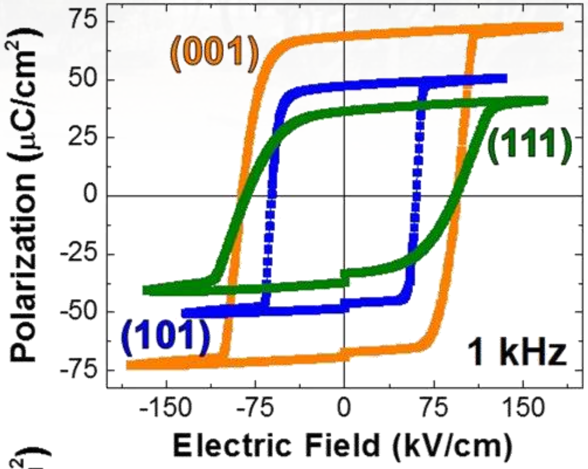


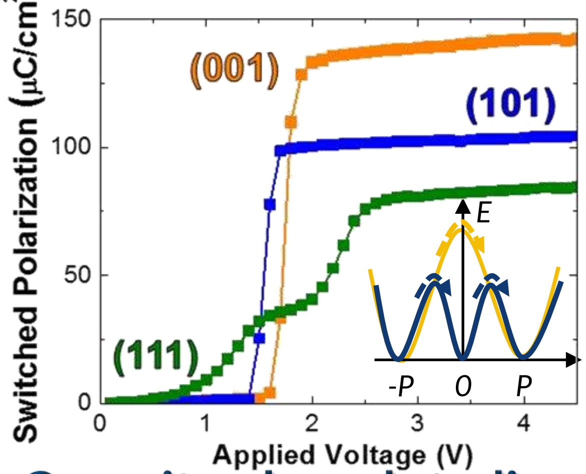
Xu *et al.*, *Nature Mater.* **14**, 79 (2015)

- (001)/(101) → Sharp 180° switching process
- (111) → Broad switching, 90° switching events that match models

Observation of multi-step 90° switching process → intermediate states?

Indications of Intermediate States

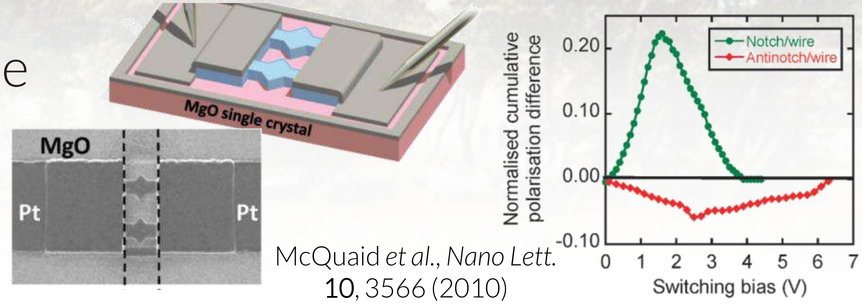




Capacitor-based studies suggest potential for intermediate states…

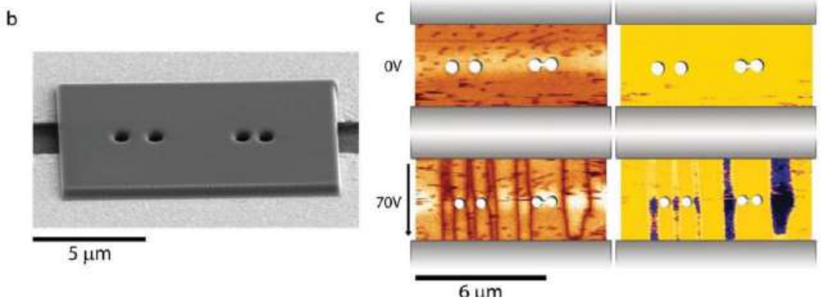
Controlling DW Formation/Motion

- Micro- and nano-machining → Controlling the boundary conditions controls how domains respond, tune mobility with “notches” and “antinotches”



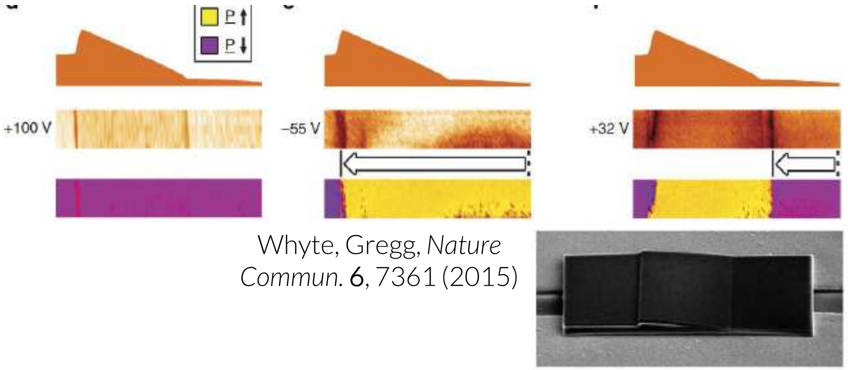
McQuaid *et al.*, *Nano Lett.* **10**, 3566 (2010)

- Making something from nothing → Researchers used “air holes” w/ FIB to create hot-spots to manipulate domain nucleation/growth



Whyte *et al.*, *Adv. Mater.* **26**, 293 (2014)

- Ferroelectric domain-wall diode → Single direction of motion for domain walls, irrespective of polarity, under a series of alternating electric field pulses → Sawtooth morphology is central to its function

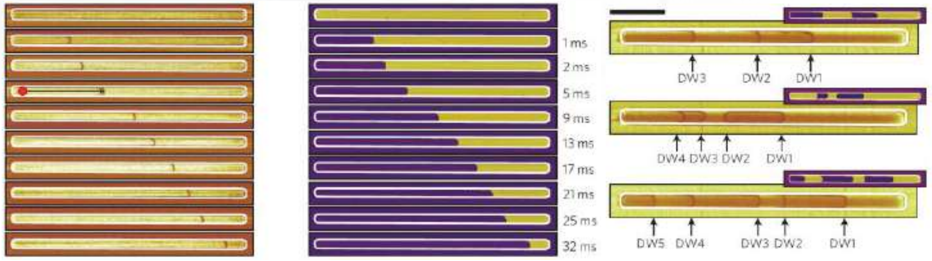


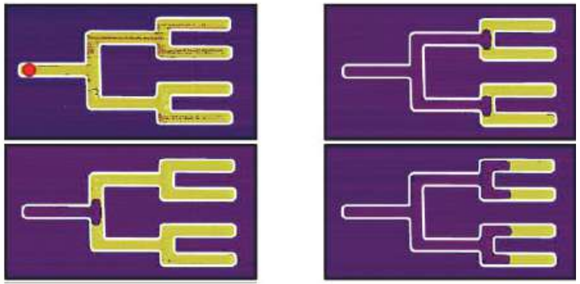
Whyte, Gregg, *Nature Commun.* **6**, 7361 (2015)

Understanding the energies at play enables one to grab deterministic control over domain production and motion under applied fields

Controlling DW Formation/Motion

To fully harness potential of DWs functional entities → Essential to achieve reliable, precise control of their nucleation, location, number, and velocity

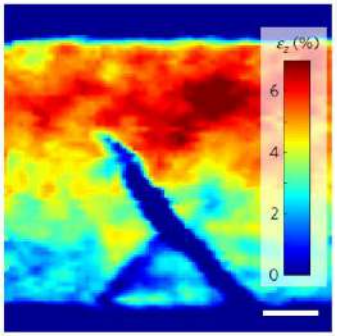




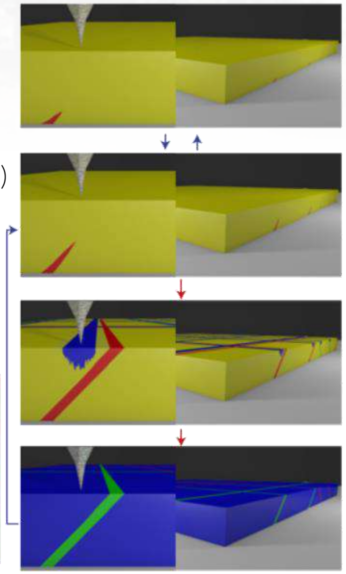
McGilly *et al.*, *Nature Nanotechnol.* 10, 145 (2015)

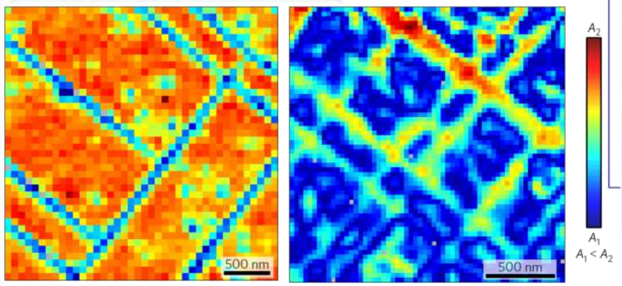
Emulates magnetic logic devices…

- Ability to "pick" a nucleation site
- Position controlled by tuning of voltage pulses; multiple domain walls can be nucleated and handled reproducibly
- A step towards the realization of domain-wall nanoelectronics utilizing ferroelectric thin films



Agar *et al.*, *Nature Mater.* 15, 549 (2016)





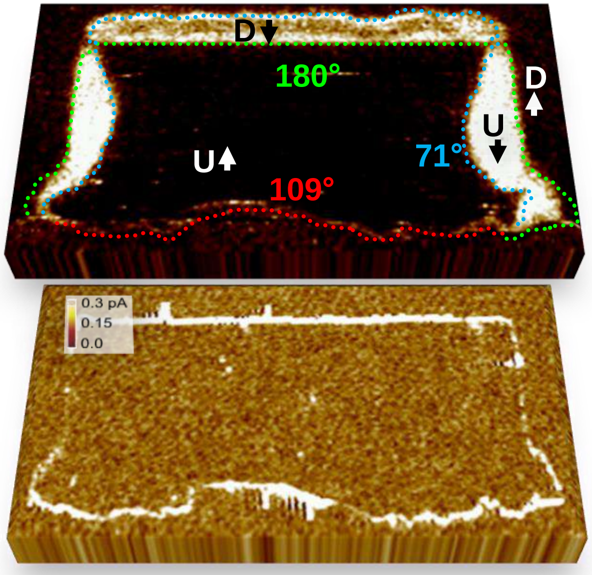
- Strain-gradients → manipulate domain structure, "needle" domains
- Such domains are found to be highly mobile → out-of-plane fields drive reduction/growth of the *a* domain
- Produces large piezoelectric responses at domain walls → not seen in classic

Domain Wall as the Device

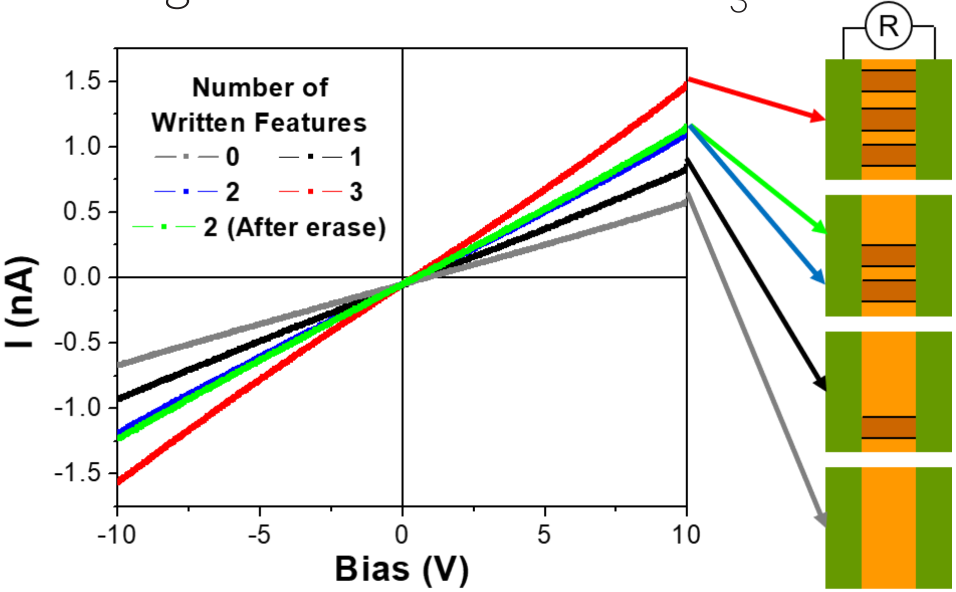
- Domain walls can contribute to properties via *extrinsic* contributions…
- But what about making the domain wall itself the functional unit?
- Ferroelectric domain walls as device elements

Catalan *et al.*, *Rev. Mod. Phys.* **84**, 119 (2012);
Vasudevan *et al.*, *Adv. Funct. Mater.* **23**, 2592 (2013)

- Domain walls can exhibit properties that differ from those of the bulk
  - Domain-wall conductivity in insulating ferroelectrics → $BiFeO_3$



Seidel, Martin *et al.*, *Nature Mater.* **8**, 229 (2009)



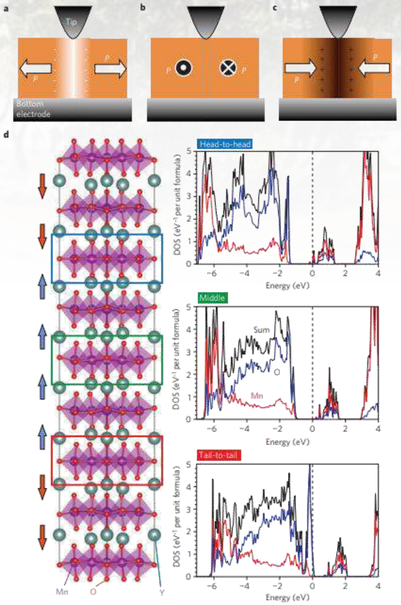
- Resistance of a single wall ~ 8 x 10^{10} Ω → Estimated resistivity ~2.5 Ω·m
  - Bulk resistivity ~ 10^7 Ω·m
- Conductivity was ascribed to an increased carrier density resulting from an electrostatic potential step and a decrease in the bandgap within the wall

Eliseev *et al.*, *Phys. Rev. B* **83**, 235313 (2012); Eliseev *et al.*, *Phys. Rev. B* **85**, 045312 (2012); Morozovska *et al.*, *Phys. Rev. B* **86**, 085315 (2012)

Function at Domain Walls

- Conduction at domain walls in…
  - $BiFeO_3$ (all types) Farokhipoor *et al.*, *Phys. Rev. Lett.* **107**, 127601 (2011)
  - $PbZr_{1-x}Ti_xO_3$ Guyonnet *et al.*, *Adv. Mater.* **23**, 5377 (2011)
  - $BaTiO_3$ Sluka *et al.*, *Nature Commun.* **4**, 1808 (2013)
  - $Er(Y)MnO_3$ Meier *et al.*, *Nature Mater.* **11**, 284 (2012)
  - $HoMnO_3$ Wu *et al.*, *Phys. Rev. Lett.* **108**, 077203 (2012)
  - $LiNbO_3$ Schroder *et al.*, *Adv. Funct. Mater.* **22**, 3936 (2012)

Probe-based control of head-to-head and tail-to-tail domain wall configuration and band structure in $YMnO_3$

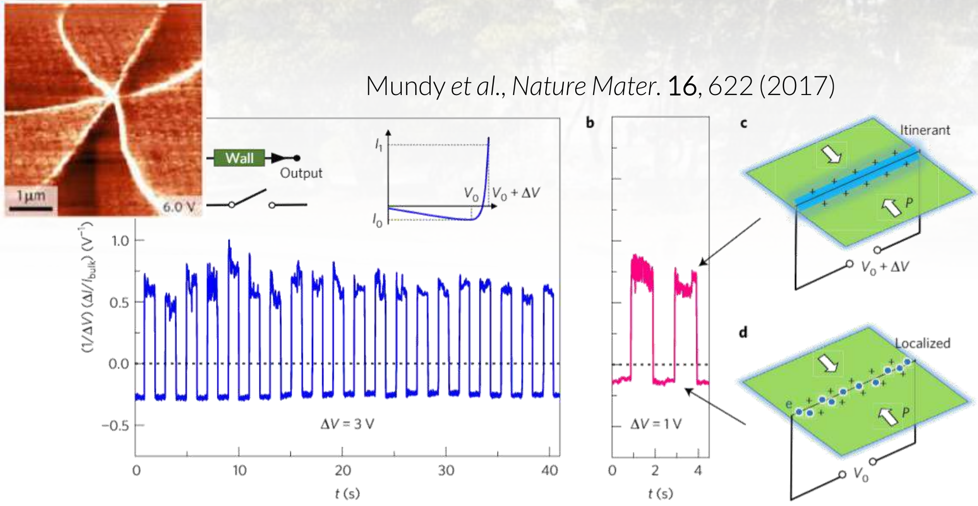


- Studies have probed…
  - Role of oxygen vacancies in the conduction behavior Seidel *et al.*, *Phys. Rev. Lett.* **105**, 197603 (2010); Gaponenko *et al.*, *Appl. Phys. Lett.* **106**, 162902 (2015)
  - How domain-wall curvature can give rise to large conductivity changes (~500%) Vasudevan *et al.*, *Nano Lett.* **12**, 5524 (2012)
  - How certain conducting domain walls in multiferroics can exhibit large magnetoresistance (~60%) He *et al.*, *Phys. Rev. Lett.* **108**, 067203 (2012)
  - Topological defect states and exotic properties in vortex domain structures Balke *et al.*, *Nature Phys.* **8**, 81 (2012)
  - Memristor-like functionality Maksymovych *et al.* *Nano Lett.* **11**, 1906 (2011)

Advances in synthesis/characterization have exposed new phenomena…
Was the ultimate nanoscale device there all along?

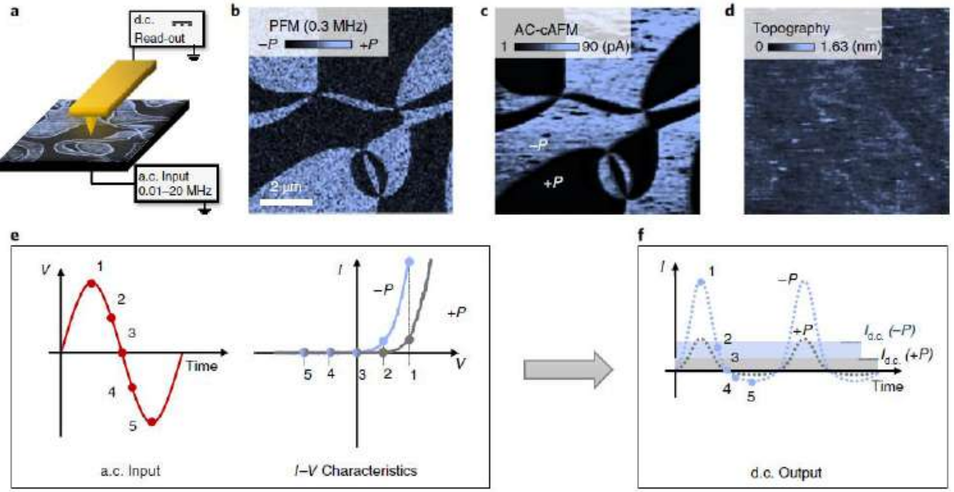
Domain-Wall Devices

- *E-field control of the electronic transport at FE domain walls*
- Reversibly switch resistive/conductive behavior at charged walls in $ErMnO_3$
- Transition → formation (activation) of inversion layer that acts as the channel for the charge transport
- "Foreshadow the possibility to design elementary digital devices for all-domain-wall circuitry"



Mundy *et al.*, *Nature Mater.* **16**, 622 (2017)

- ***Diode-like alternating-to-direct current conversion*** based on neutral FE domain walls in $ErMnO_3$
- Showed rectification at the tip-wall contact for frequencies at which the walls are effectively pinned
- Mechanism → Transport behavior at the walls arises from oxygen defects
- Frequency regime/magnitude of the direct current output controlled by bulk conductivity → electrode–wall junctions



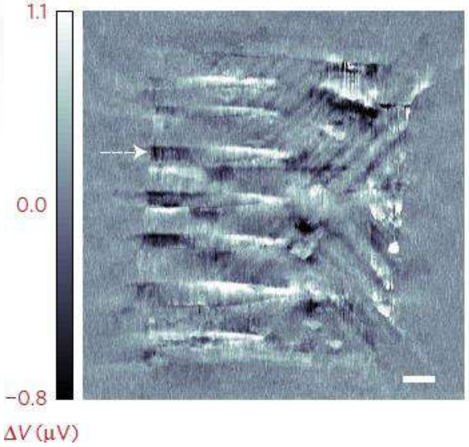
Schaab *et al.*, *Nature Nanotechnol.* **13**, 1028 (2018)

Domains in Ferroelastic $SrTiO_3$

Domain-wall Nanoelectronics Using Ferroelastic Materials

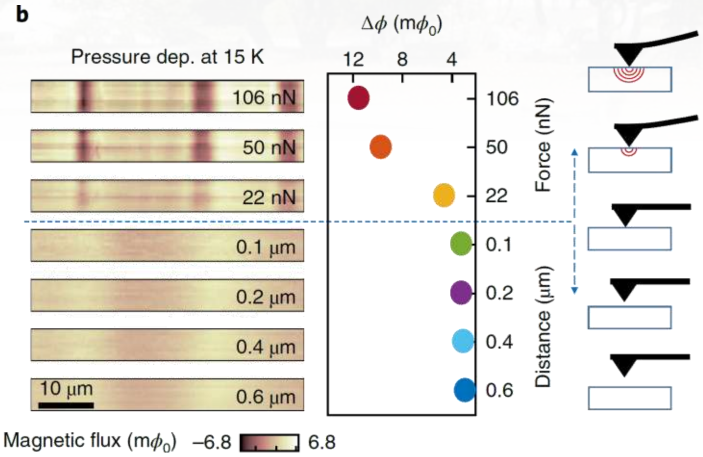
Local strain tuning of conductivity

Frenkel *et al.*, *Nature Mater.* **16**, 1203 (2017)



- Twin boundaries, with properties that are different from their surrounding bulk, tune $LaAlO_3/SrTiO_3$ interface 2DEG
- $SrTiO_3$ ferroelastic domain boundaries → Remaining highly mobile @ low-T, recently suggested to be polar
- Localized pressure to a twin boundary, detect a change in $LaAlO_3/SrTiO_3$ interface
- Polarity at the twin boundaries, control of conducting state

Local strain tuning of magnetism



Christensen *et al.*, *Nature Phys.* **15**, 269-274 (2019)

- Ferroelastic domain walls display strain-tunable polarity and enhanced conductivity
- Long-range magnetic order with modulations along the ferroelastic domain walls in $SrTiO_3$/heterointerfaces, manifests as a striped pattern in scanning SQUID
- Conducting interfaces → clear signatures in magnetotransport measurements
- Magnetic state coupled dynamically to the lattice, reversibly tuned by local forces

New Multiferroic (Ferroelastic + Ferroelectric + Ferromagnetic)

Where do we go from here?

High-throughput discovery and design of next-generation functional materials → Can we design new domain-wall function?

Non-traditional ferroelectrics → Domain walls in polymer, 'stuffed wurtzite' structures, 2D materials, 'hybrid perovskite' crystals, thiophosphates, HfO_2,...

Real-time studies → *Simulation/experiment* can do exciting things; data analysis/big-data/machine learning will extract new understanding of domain walls/structures

Ferroelectrics for energy → Domain walls can play a roll in impacting photovoltaic response, catalysis, physisorption rates of reactants,...

True domain-wall devices → emulate real device components with domain walls? Can we deterministically place/control them?

Domains & domain walls result from energy minimization in ferroic materials...

Their configuration can have profound impact on the macroscale properties...

Domain walls themselves can contribute properties unique from the bulk...