Ferroelastic Domain Patterns in Free-Standing Nanoferroelectrics

Phase Field Studies with Comparisons to Experimental Observations

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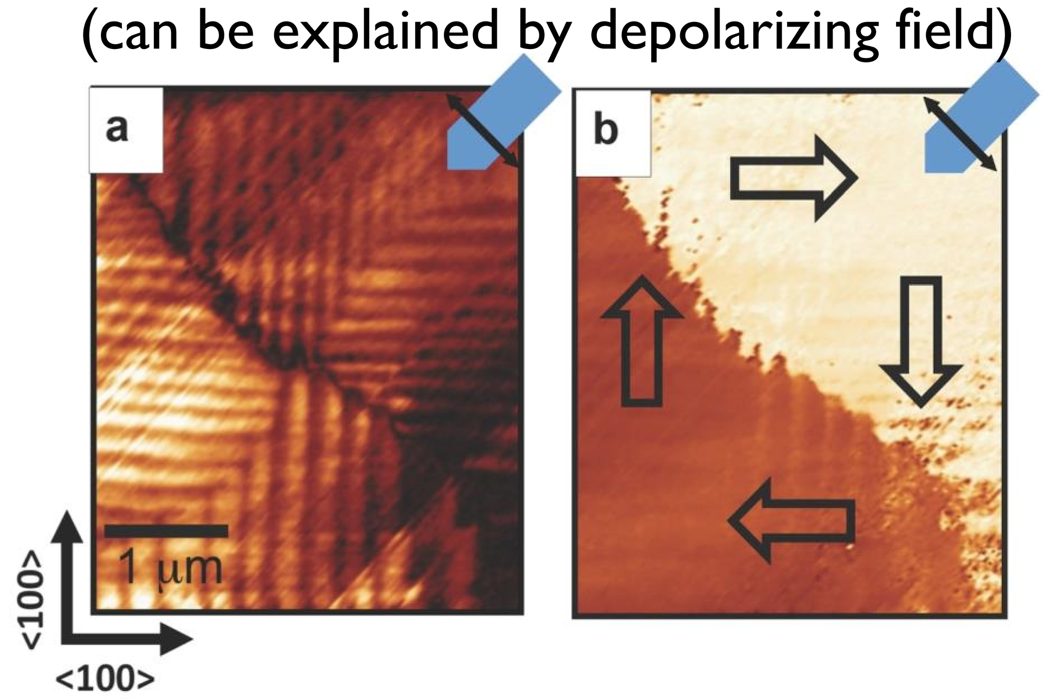
Motivation

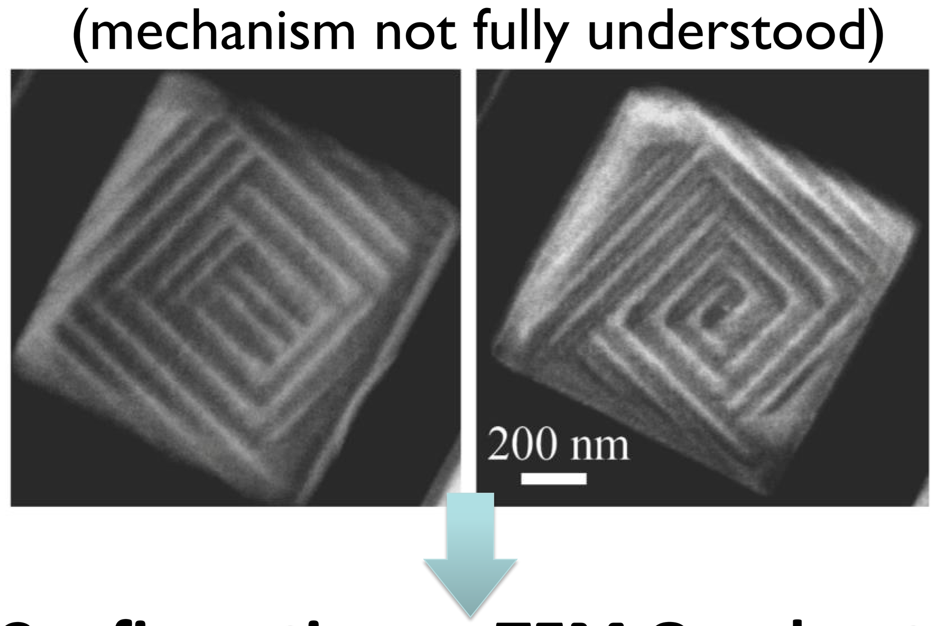
- Ferroelectric nanostructures – underlying physics
- Explain experimentally observed quadrant features in TEM images[1], and why they could differ from PFM (Piezoresponse Force Microscopy) images.
- Applications: next generation ferroelectric devices

Piezoresponse Force Microscopy (PFM) vs TEM

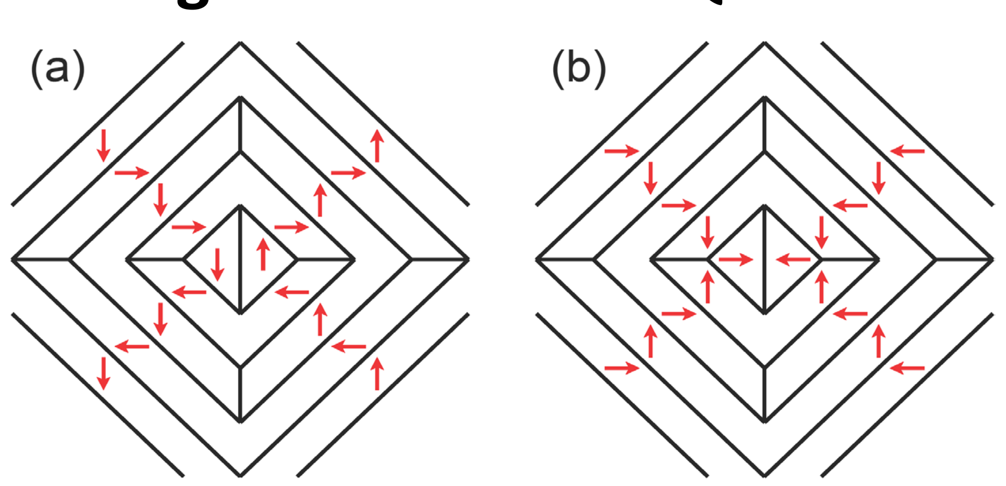
PFM images show flux closure domain patterns [2]
(can be explained by depolarizing field)



But TEM images show quadrant domain patterns [2]
(mechanism not fully understood)



Possible Configurations – TEM Quadrant Patterns



Model

- Devonshire-Ginzburg-Landau energy for polarizations
- Real Space approach: key to modeling nanostructures, boundary conditions, free surfaces
- Finite difference, explicit time stepping for polarization evolution, electrostatic and elastic fields [3]
- 3D, parallel code [4]

Free Energy

$$F_T = \int d\mathbf{r} \left[f_L + \frac{K}{2} |\nabla P_i|^2 + f_{\text{elastic}} \right]$$

$$\begin{aligned} f_L = \; & a_{ij} P_i P_j + a_{ijkl} P_i P_j P_k P_l \\ & + a_{ijklmn} P_i P_j P_k P_l P_m P_n \\ & + a_{ijklmnop} P_i P_j P_k P_l P_m P_n P_o P_q \\ f_{\text{elastic}} = \; & \tfrac{1}{2} C_{ijkl} \left(\varepsilon_{ij} - Q_{ij} P_i P_j \right) \left(\varepsilon_{kl} - Q_{kl} P_k P_l \right) \end{aligned}$$

Equation of Motion

$$\frac{\partial \mathbf{P}}{\partial t} = -\Gamma \left[\frac{\delta F_T}{\delta \mathbf{P}} + \nabla \phi \right]$$

Mechanical Equilibrium [3,4]

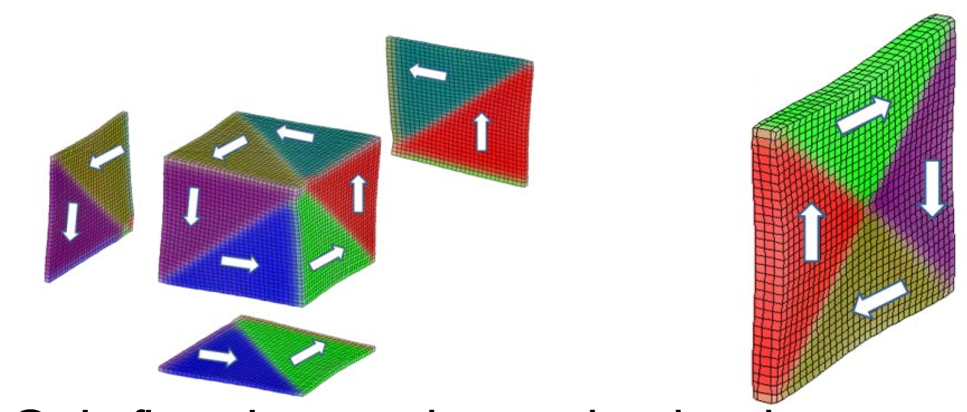
$$\rho \ddot{u}_i = \frac{\partial \sigma_{ij}}{\partial x_j} + \eta \nabla^2 \dot{u}_i \xrightarrow{\text{as } t \to \infty} 0$$

Maxwell Equation

$$\nabla \cdot \left(-\epsilon_0 \nabla \phi + \mathbf{P} \right) = -q N_e$$

[1] A. Schilling et. al., *Nano Lett*, **9**(9), 3359 (2009).
[2] R. Ahluwalia, N. Ng, A. Schilling, R. G. P. McQuaid, D. M. Evans, J. M. Gregg, D. J. Srolovitz, and J. F. Scott, *Physical Review Letters*, **111**(16), 165702 (2013).
[3] N. Ng et. al., *Acta Materialia*, **57**(7), 2047, (2009).
[4] N. Ng et. al., *Acta Materialia*, **60**(8), 3632 (2012).

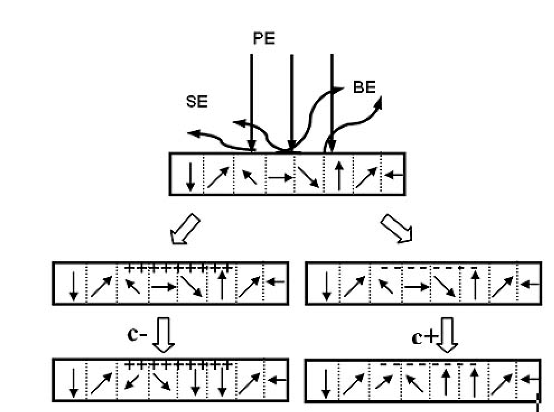
Free Standing Nanostructures [4]



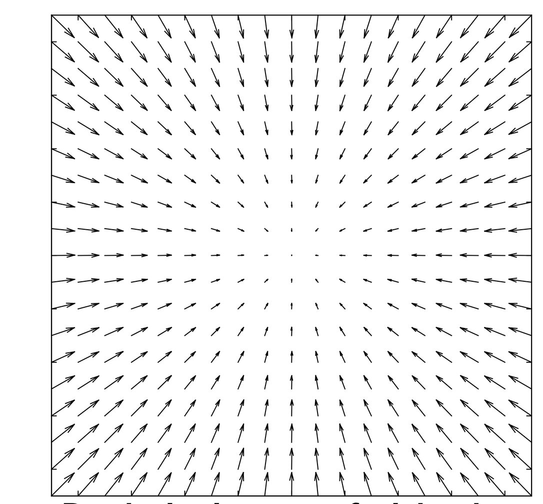
Flux closure domains form due to depolarizing fields

Only flux closure observed – this does not explain quadrant patterns

Is Electron Beam Charging Responsible?

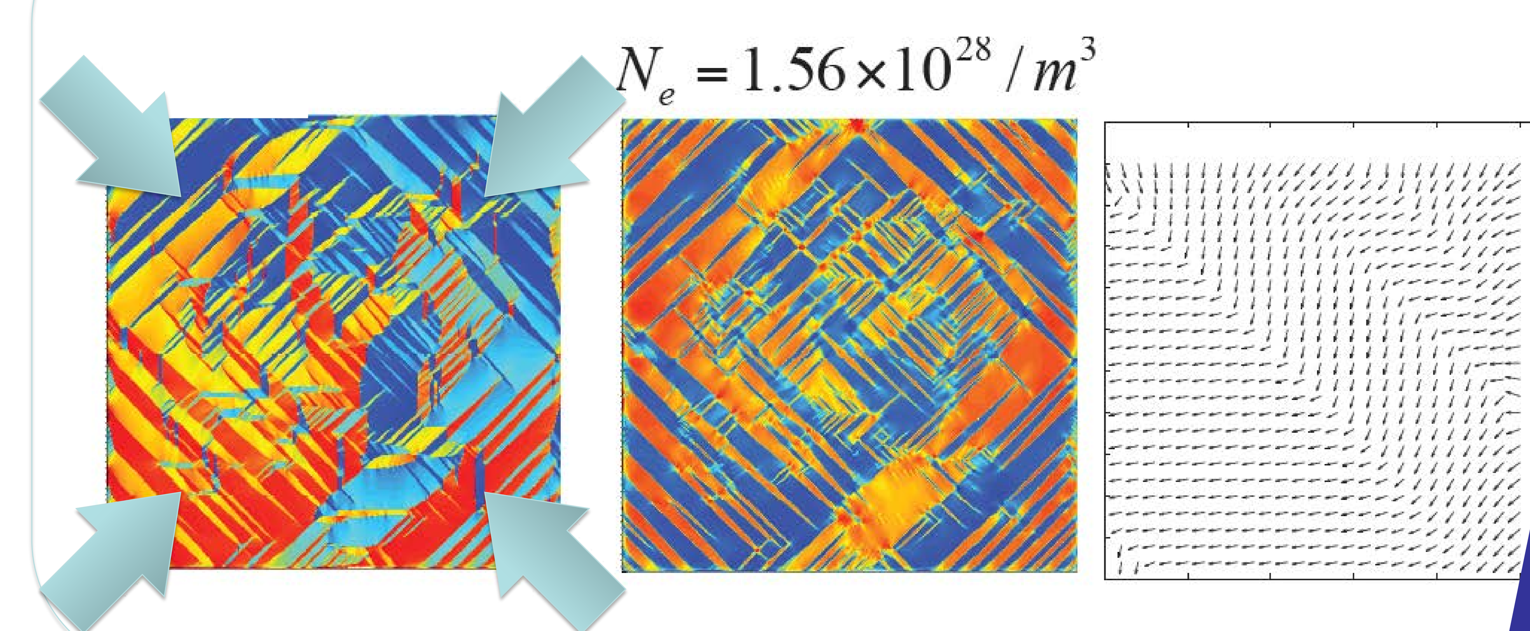


Li and Bonnell, *Annu Rev Mater Res*, 2008



Radial electric fields due to uniform charge

Radial Electric Fields Create Quadrant Domain Patterns [2]



Conclusion

- Differences between PFM and TEM images
- Depolarizing fields lead to flux closure in PFM imaging
- Electron beam charging leads to quadrant patterns with radial polarizations in TEM imaging

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