

# Poling, Reversing, and Cycling

**Xiaoli TAN, Hanzheng GUO, Cheng MA**

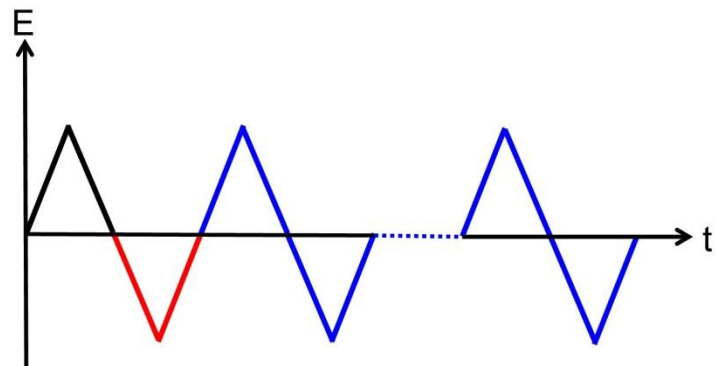
Department of Materials Science and Engineering

Iowa State University, Ames, IA50011, USA

1. Poling

2. Reversing

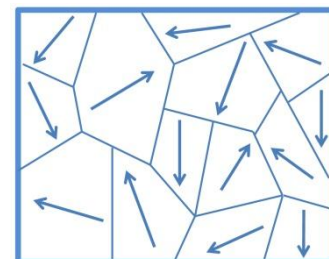
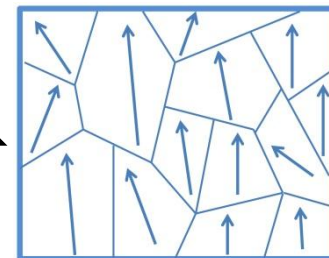
3. Cycling



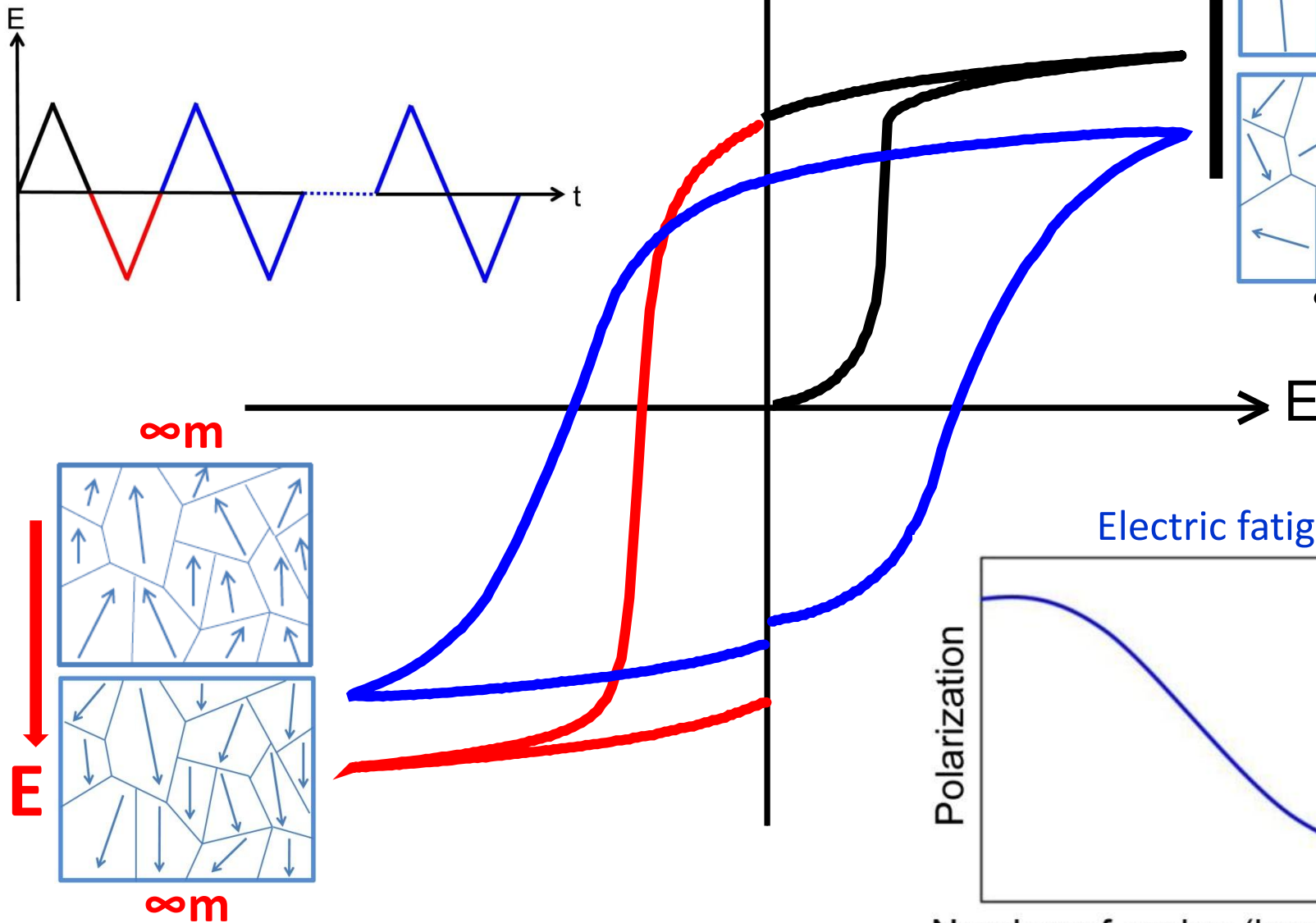
P

E

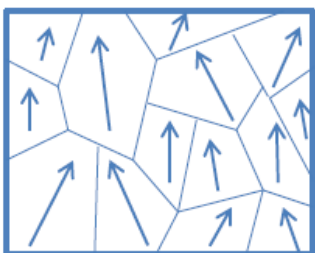
$\infty m$



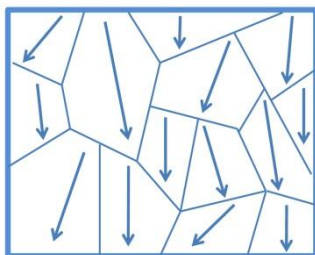
$\infty m$



$\infty m$

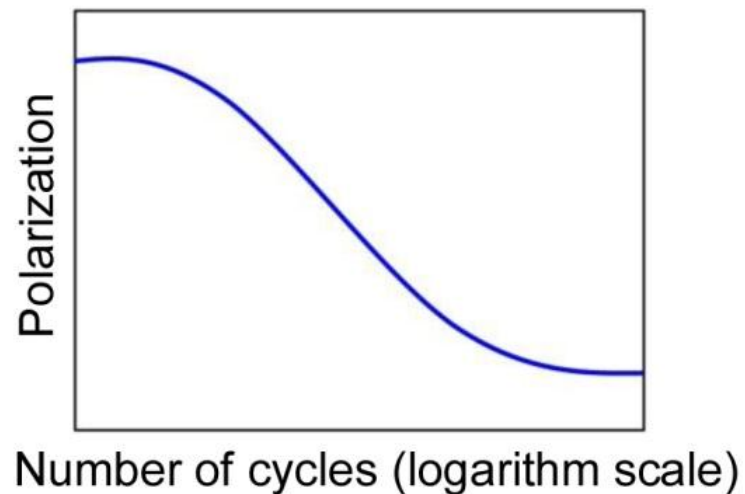


E

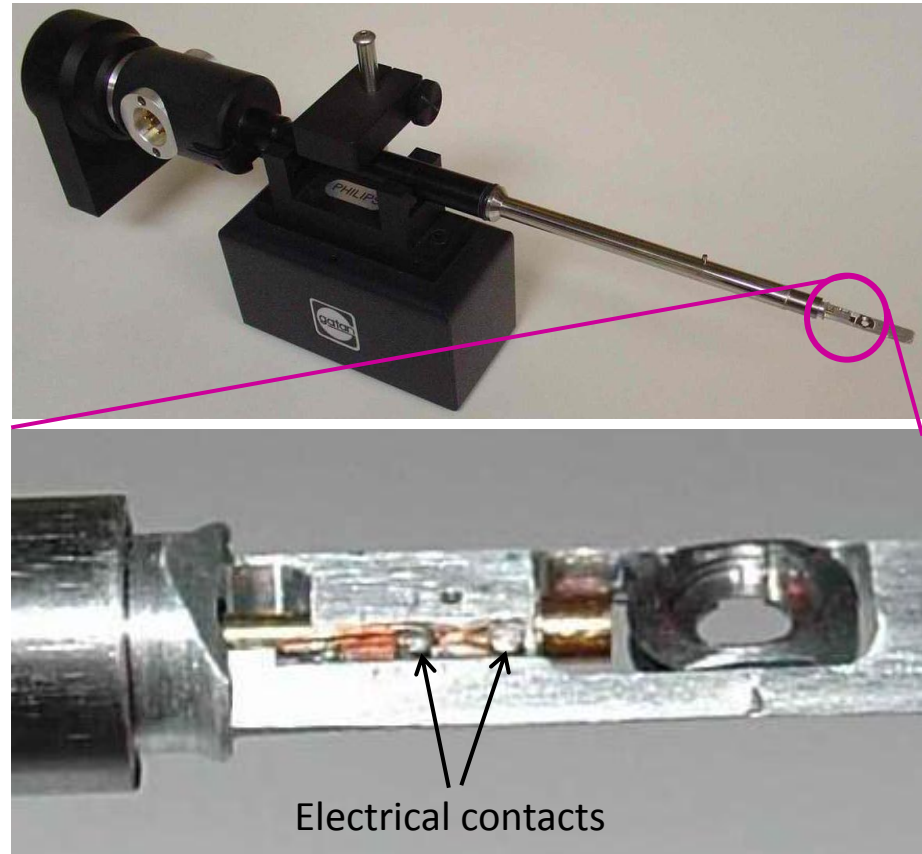
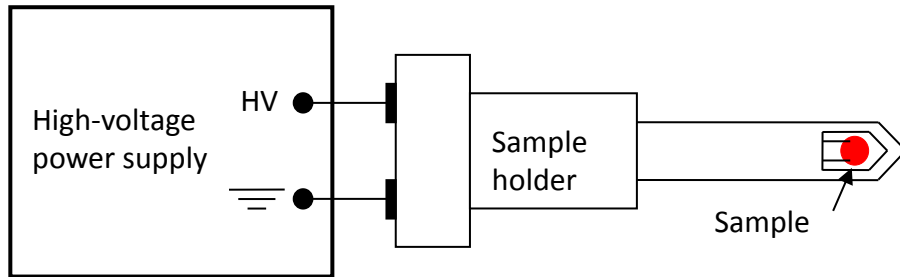
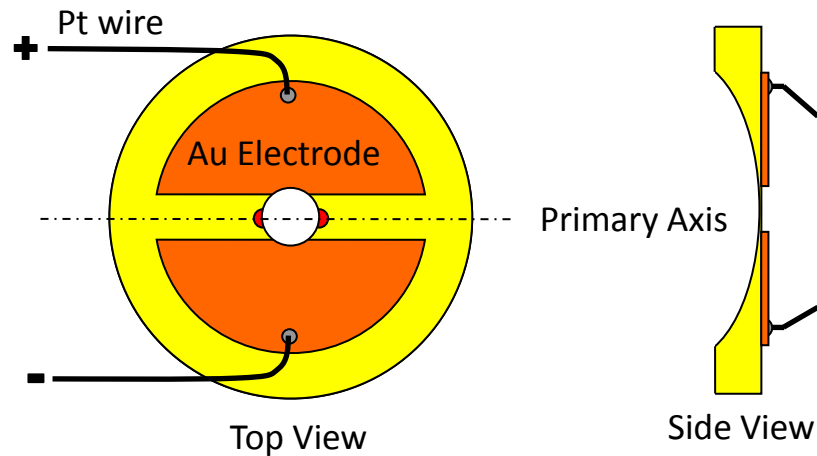


$\infty m$

Electric fatigue

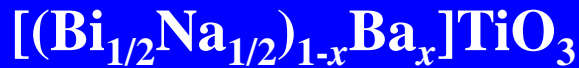
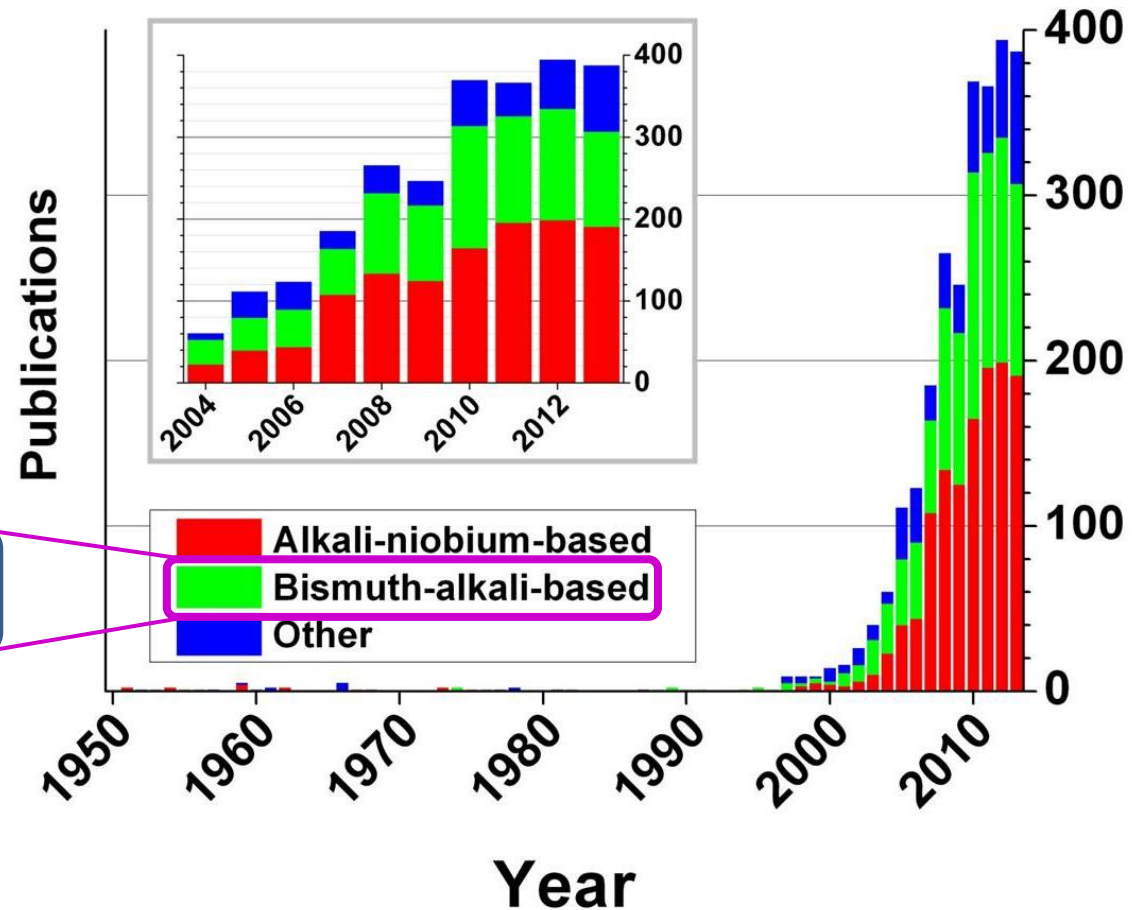


# The E-field *in situ* TEM technique: Double-tilt stage



- X. Tan, Z. Xu, J.K. Shang, and P. Han, *Appl. Phys. Lett.* 77, 1529-31 (2000).
- X. Tan, H. He, and J.K. Shang, *J. Mater. Res.* 20, 1641-53 (2005).
- H. He, and X. Tan, *Phys. Rev. B* 72, 024102/01-10 (2005).
- J. Kling, X. Tan, W. Jo, H.-J. Kleebe, H. Fuess, and J. Rödel, *J. Am. Ceram. Soc.* 93, 2452-55 (2010).
- C. Ma, H. Guo, S.P. Beckman, and X. Tan, *Phys. Rev. Lett.* 109, 107602 (2012).
- H. Guo, S.J. Zhang, S.P. Beckman, X. Tan, *J. Appl. Phys.* 114, 154102 (2013).
- H.Z. Guo, C. Zhou, X.B. Ren, and X. Tan, *Phys. Rev. B - Rapid Commun.* 89, 100104(R) (2014).
- H.Z. Guo, X. Tan et al., *Phys. Rev. B* 90, 014103/1-10 (2014).

# Annual refereed publications on lead-free piezoceramics

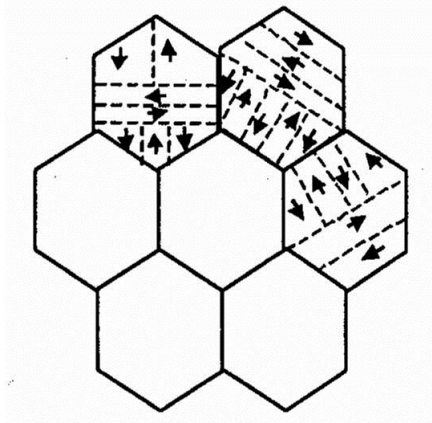


Courtesy of Prof. Jürgen Rödel

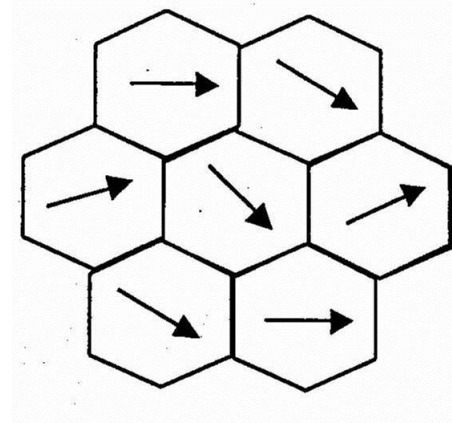
# The **poling** of ferroelectric polycrystals:

Breaking the space inversion symmetry

Transforming non-piezoelectric to piezoelectric



**DC electric field**



**Spherical symmetry**

**Curie group  $\infty\infty m$**

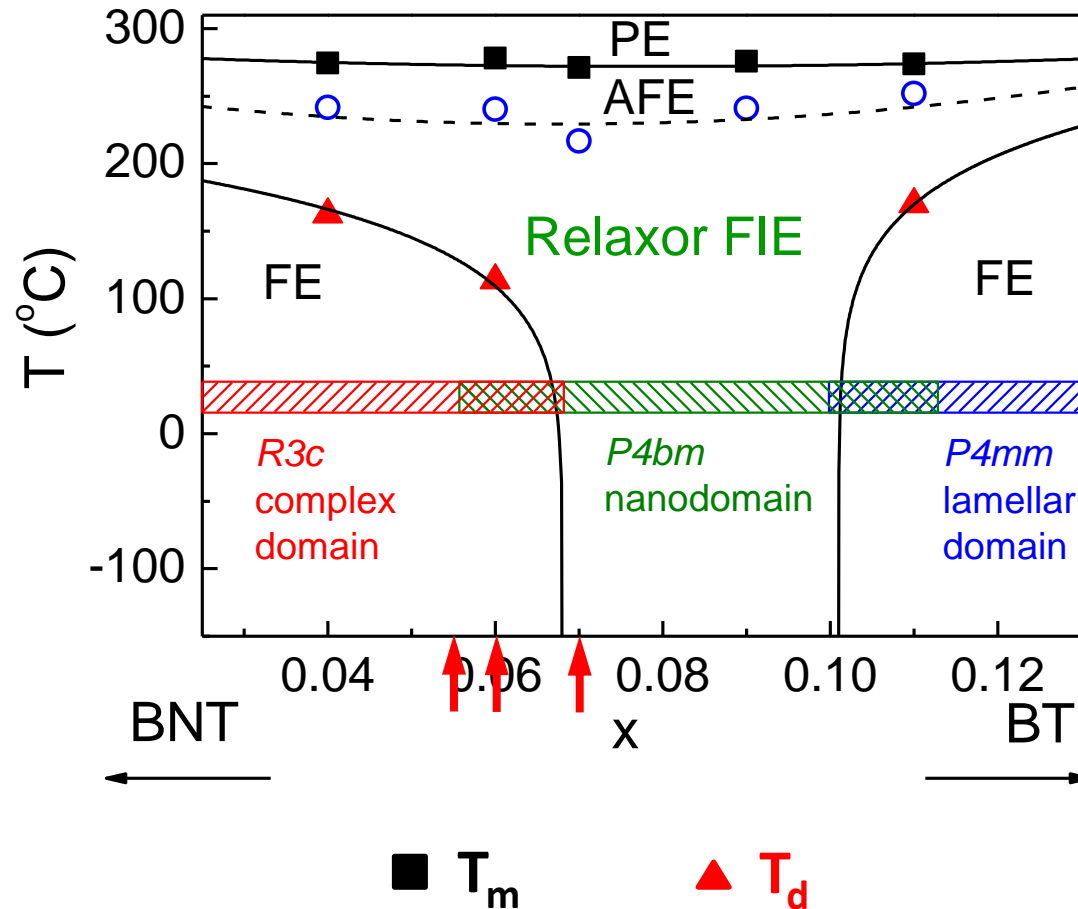
**Non-piezoelectric**

**Conical symmetry**

**Curie group  $\infty m$**

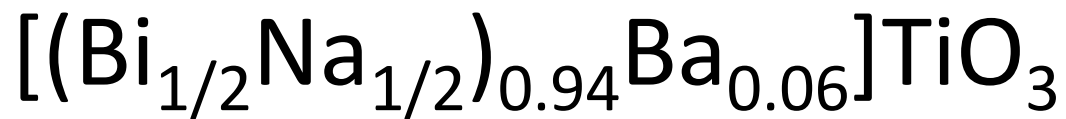
**Piezoelectric**

# The phase diagram of unpoled ceramics of $[(\text{Bi}_{1/2}\text{Na}_{1/2})_{1-x}\text{Ba}_x]\text{TiO}_3$

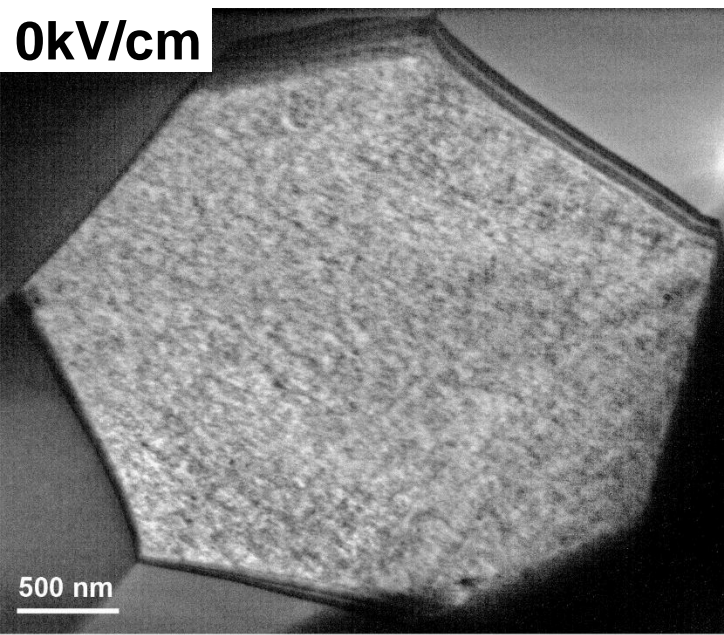


C. Ma, and X. Tan, *Solid St. Comm.* **150**, 1497 (2010).

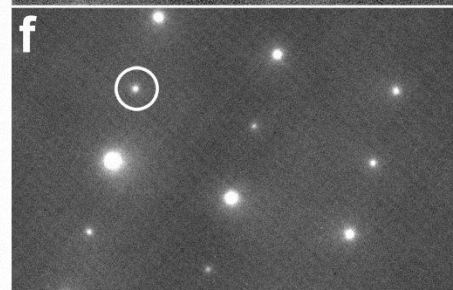
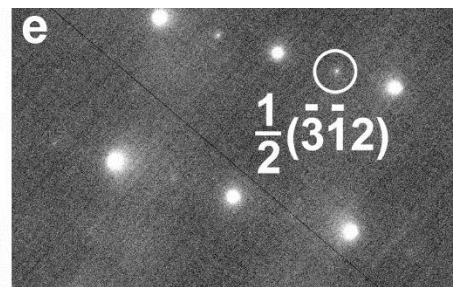
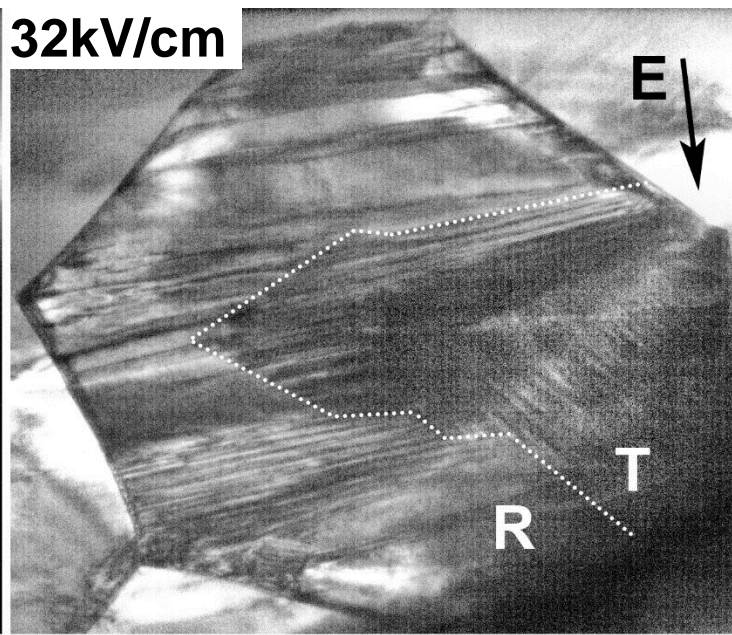
C. Ma, X. Tan, E. Dul'kin, and M. Roth, *J. Appl. Phys.* **108**, 104105 (2010).



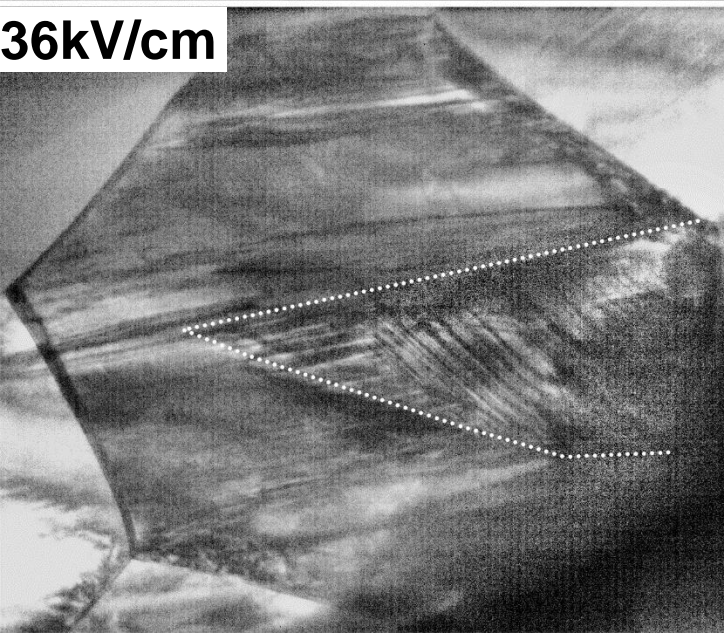
0kV/cm



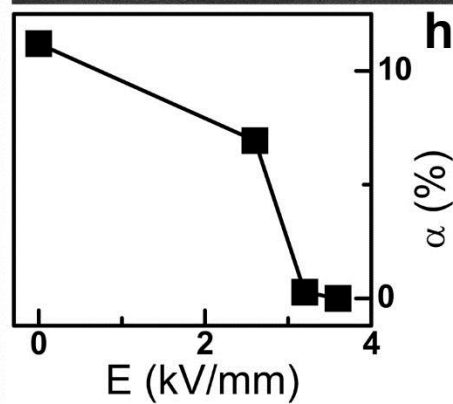
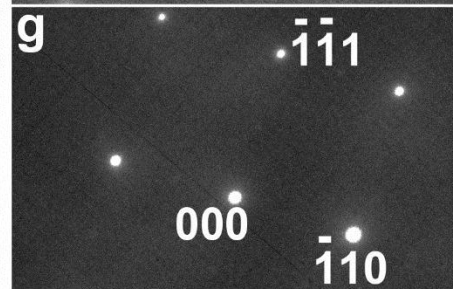
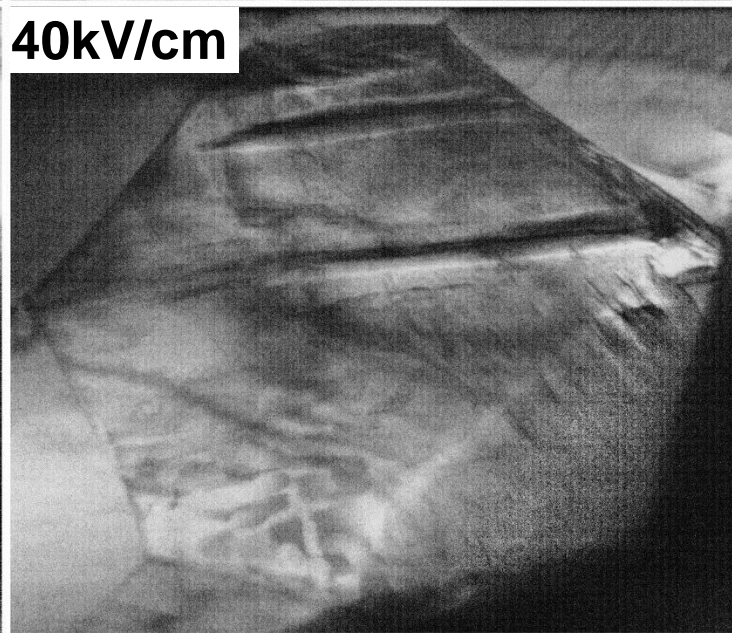
32kV/cm



36kV/cm



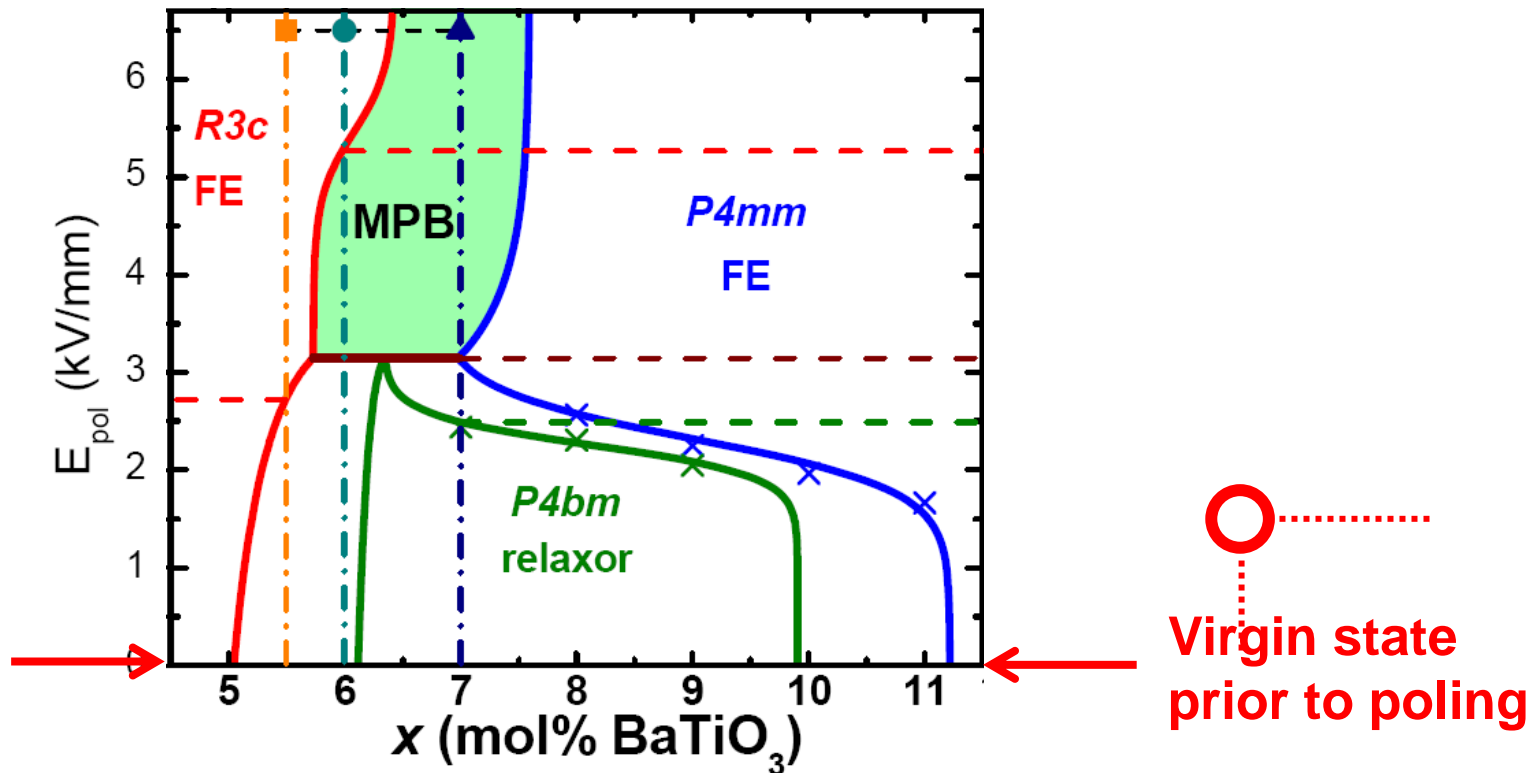
40kV/cm



# Phase transitions & the corresponding $d_{33}$

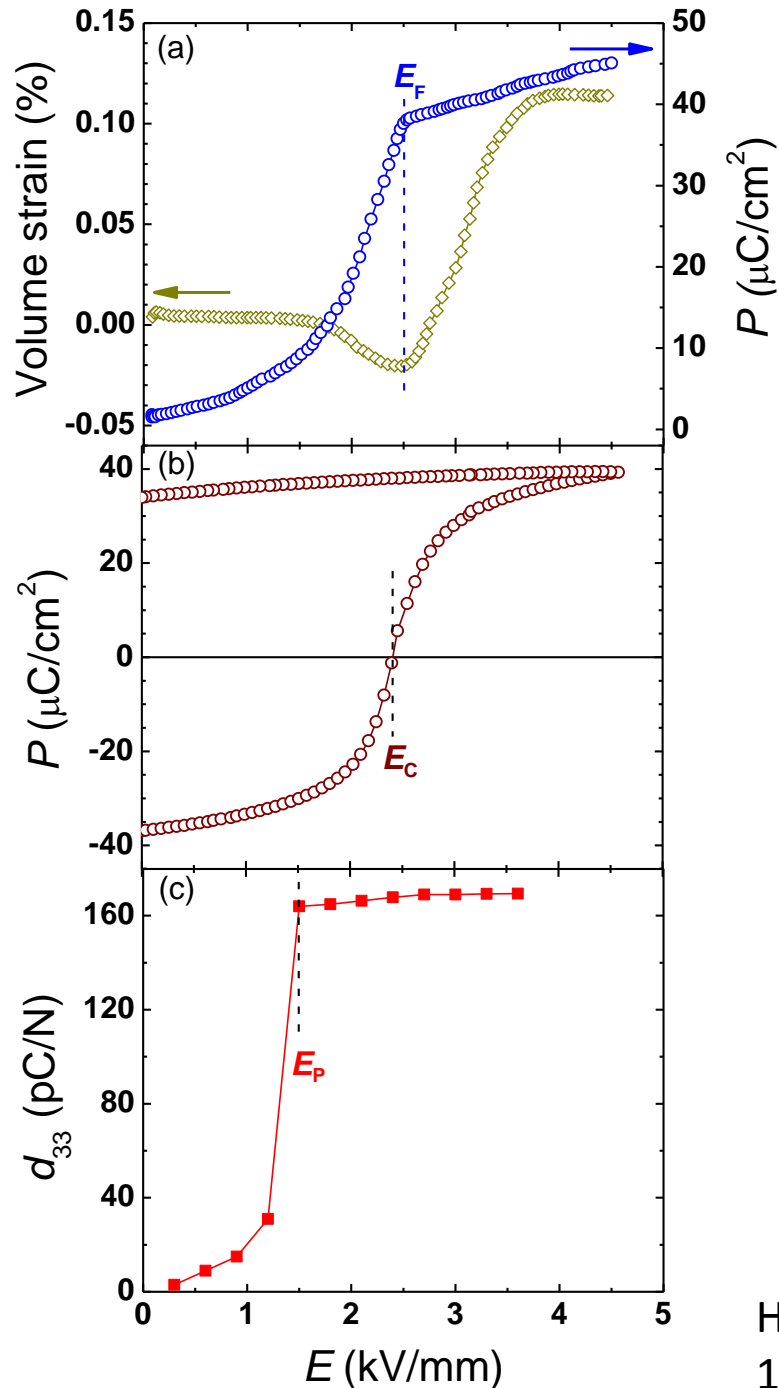
Poling: 25°C, at field levels from 1.5 to 6.5 kV/mm.

After 24 hours,  $d_{33}$  was measured at 10 spots across the electrode surface.





# Poling below $E_C$ ?



$E_F$ : the critical field to transform to ferroelectric phases

$E_C$ : the critical field to switch ferroelectric domains

$E_p$ : the critical field to develop saturating piezoelectric  $d_{33}$

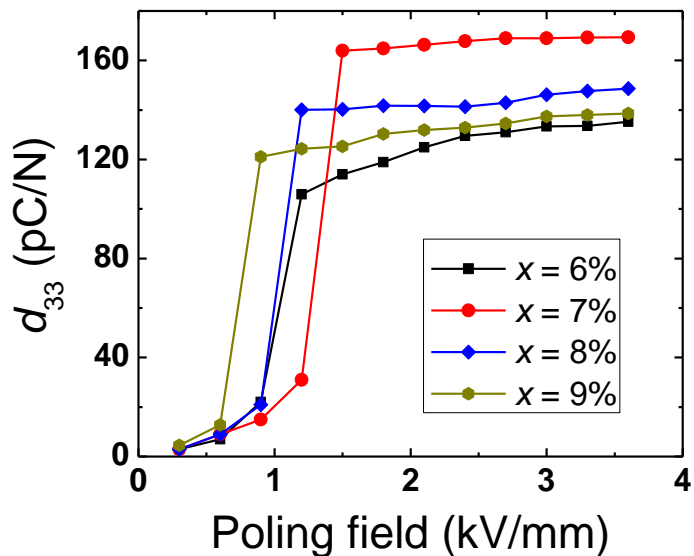
For  $[(\text{Bi}_{1/2}\text{Na}_{1/2})_{0.93}\text{Ba}_{0.07}]\text{TiO}_3$

$E_F = 2.5$  kV/mm

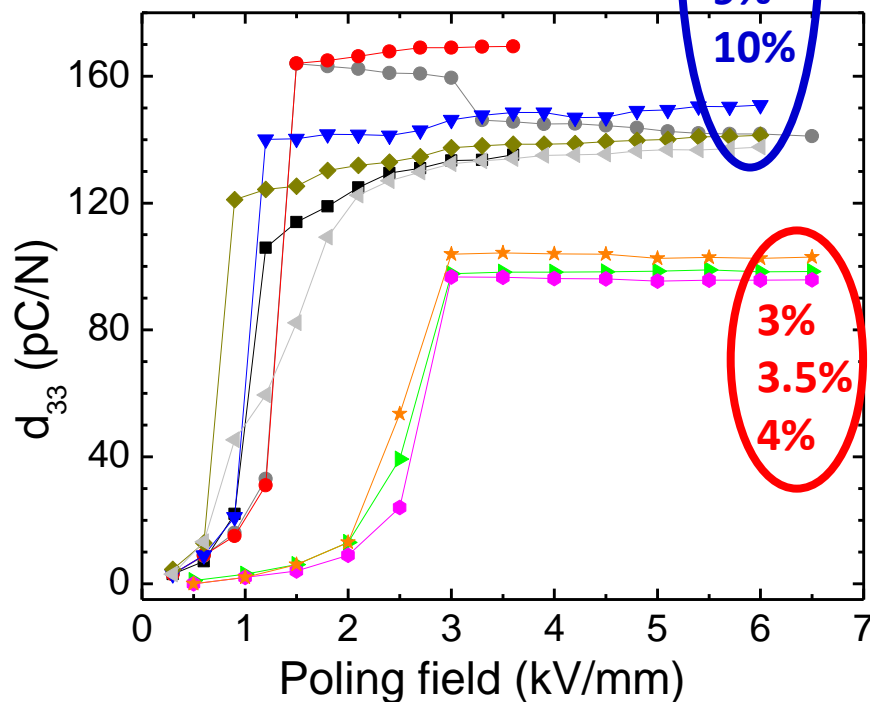
$E_C = 2.4$  kV/mm

$E_p = 1.5$  kV/mm

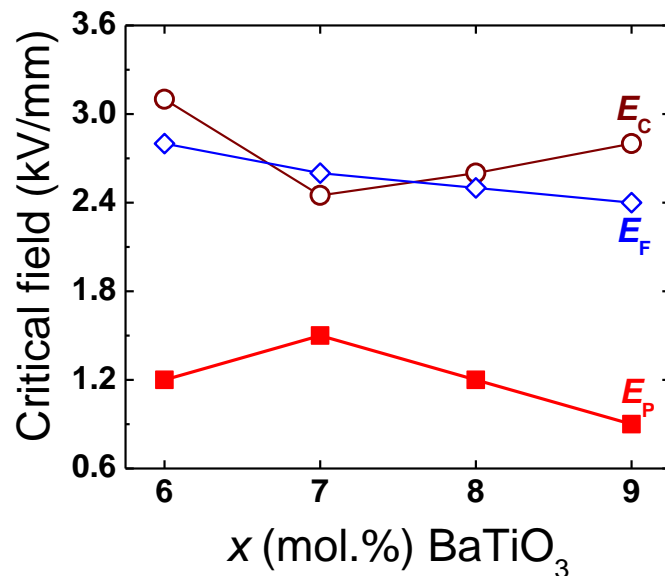
# Poling of the *P4bm* phase!



6%  
6.6%  
7%  
8%  
9%  
10%

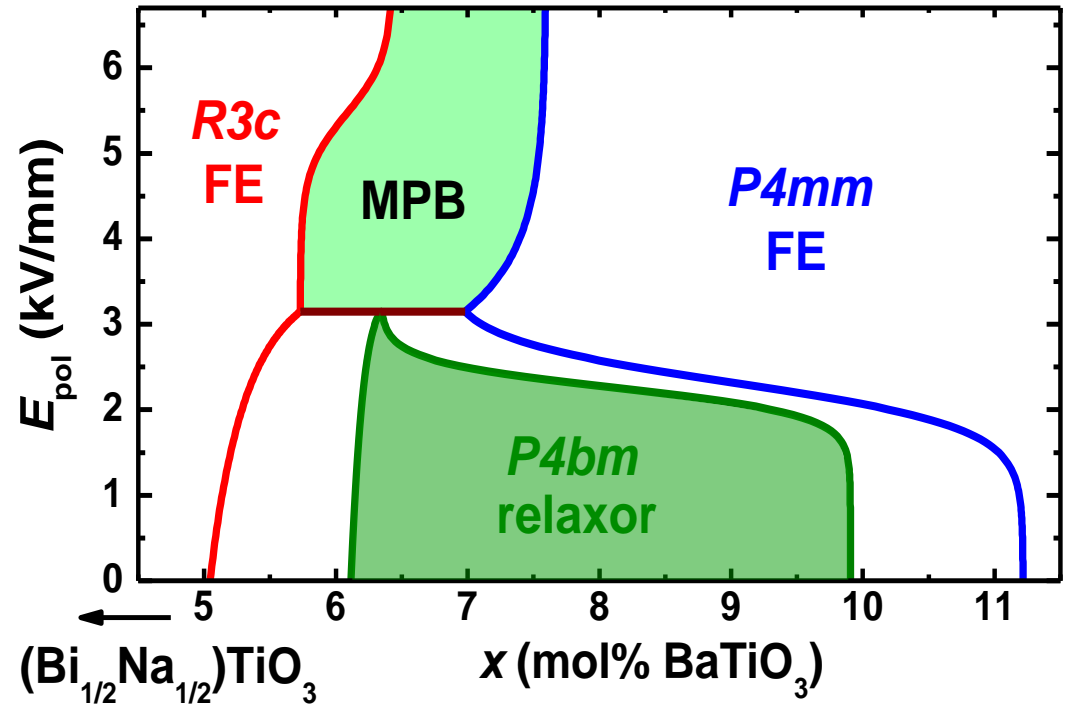
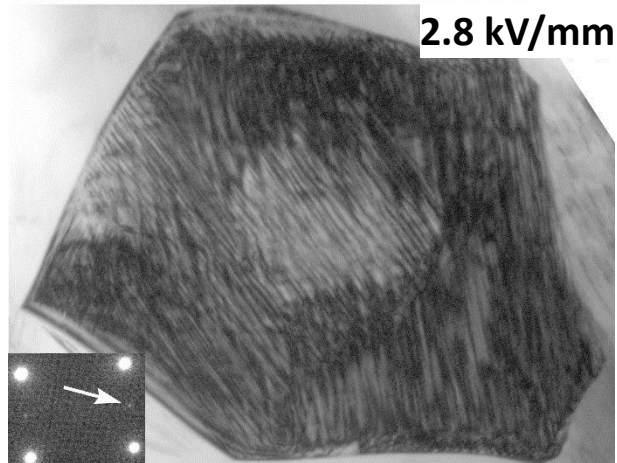
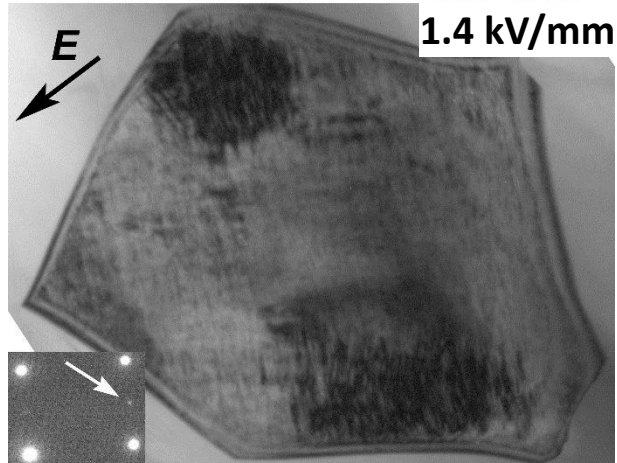
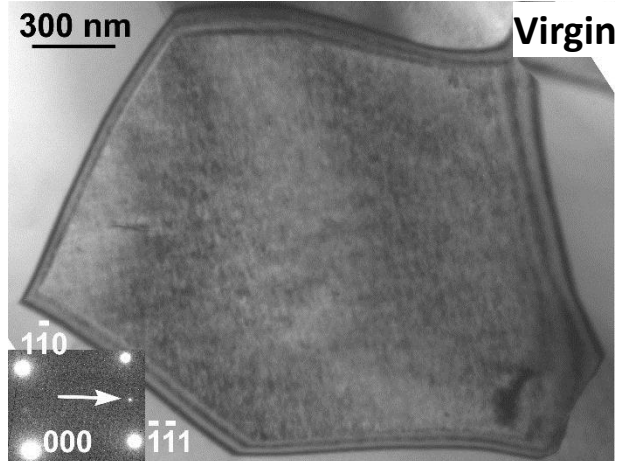


3%  
3.5%  
4%



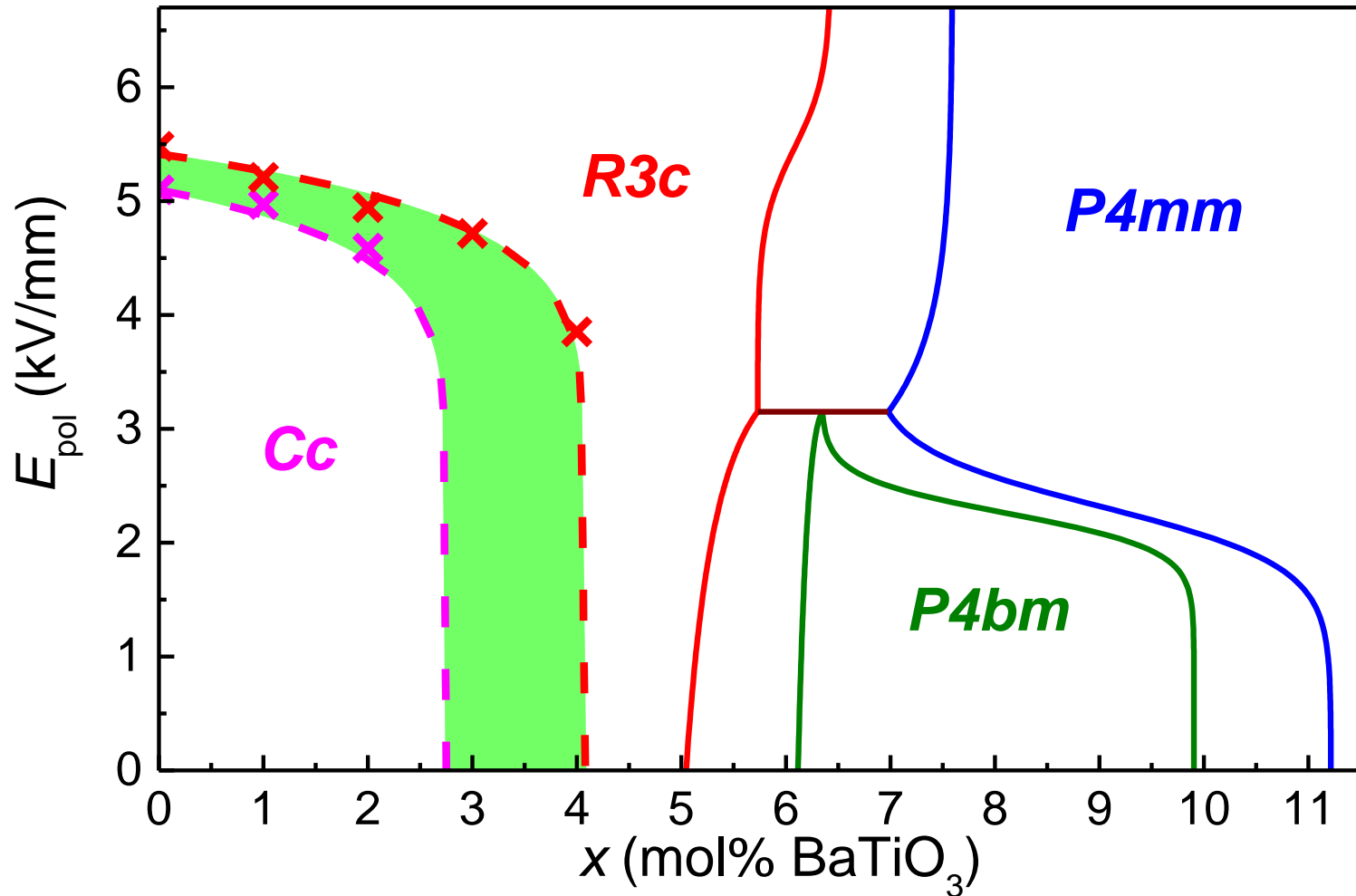
H. Guo, C. Ma, X. Liu, and X. Tan,  
*Appl. Phys. Lett.* 102, 092902 (2013).

# Poling of the *P4bm* relaxor phase



H. Guo, C. Ma, X. Liu, and X. Tan, *Appl. Phys. Lett.* 102, 092902 (2013).

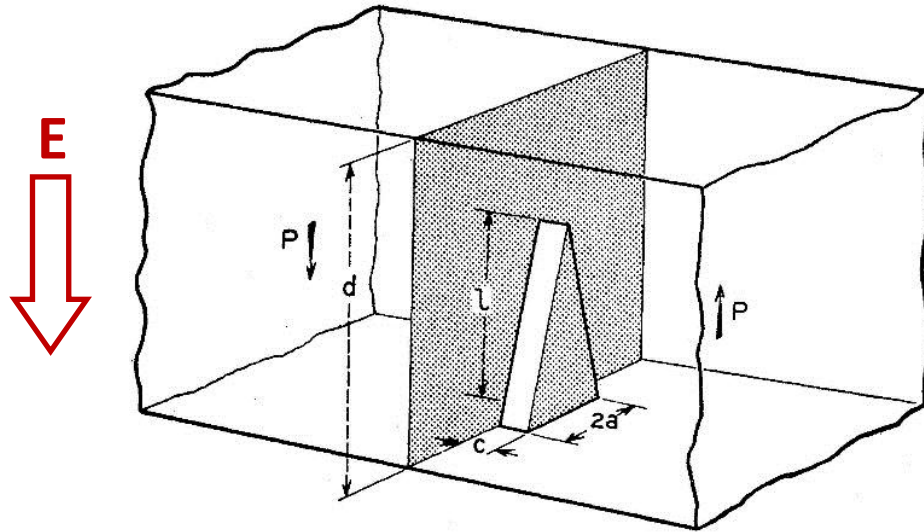
# The updated phase diagram with the $Cc$ phase



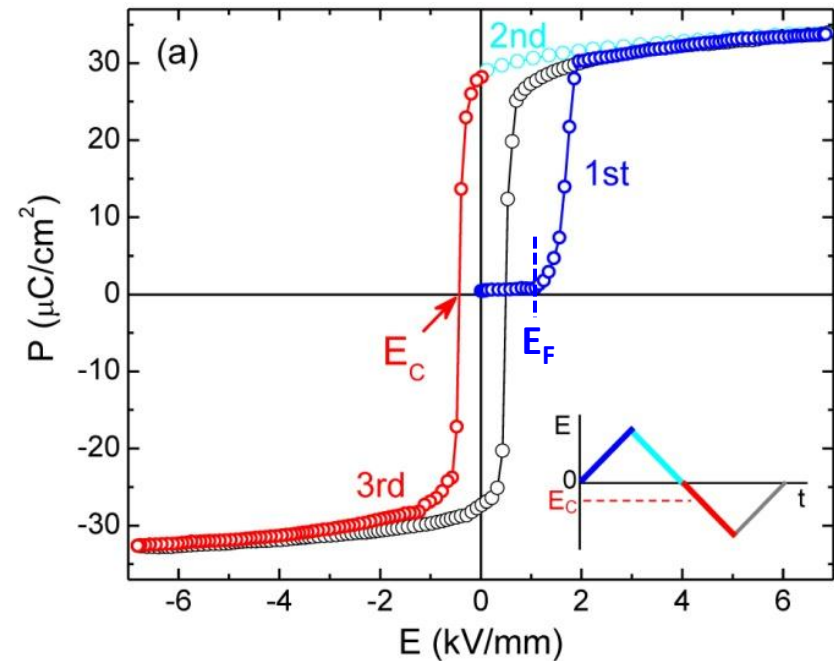
C. Ma, H. Guo, and X. Tan, *Adv. Funct. Mater.* 23, 5261-5266 (2013).

# The reversing of ferroelectric polarization

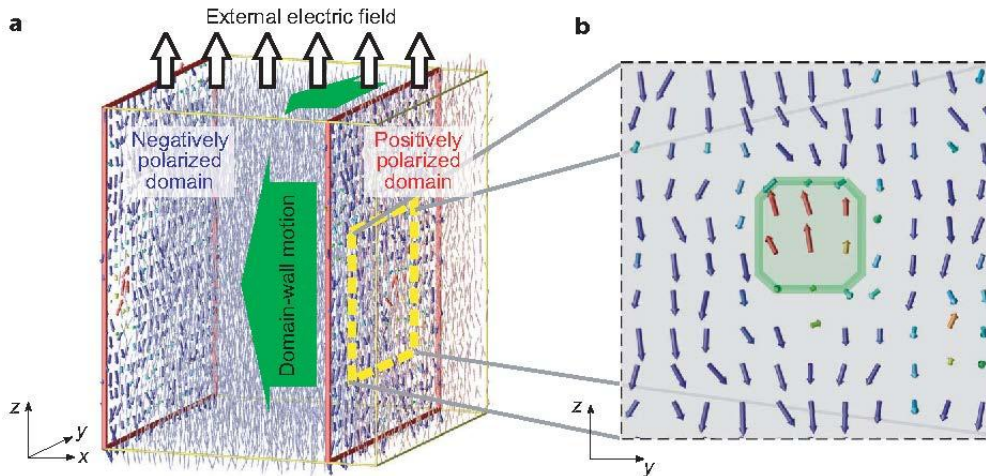
Normal ferroelectric



Induced ferroelectric

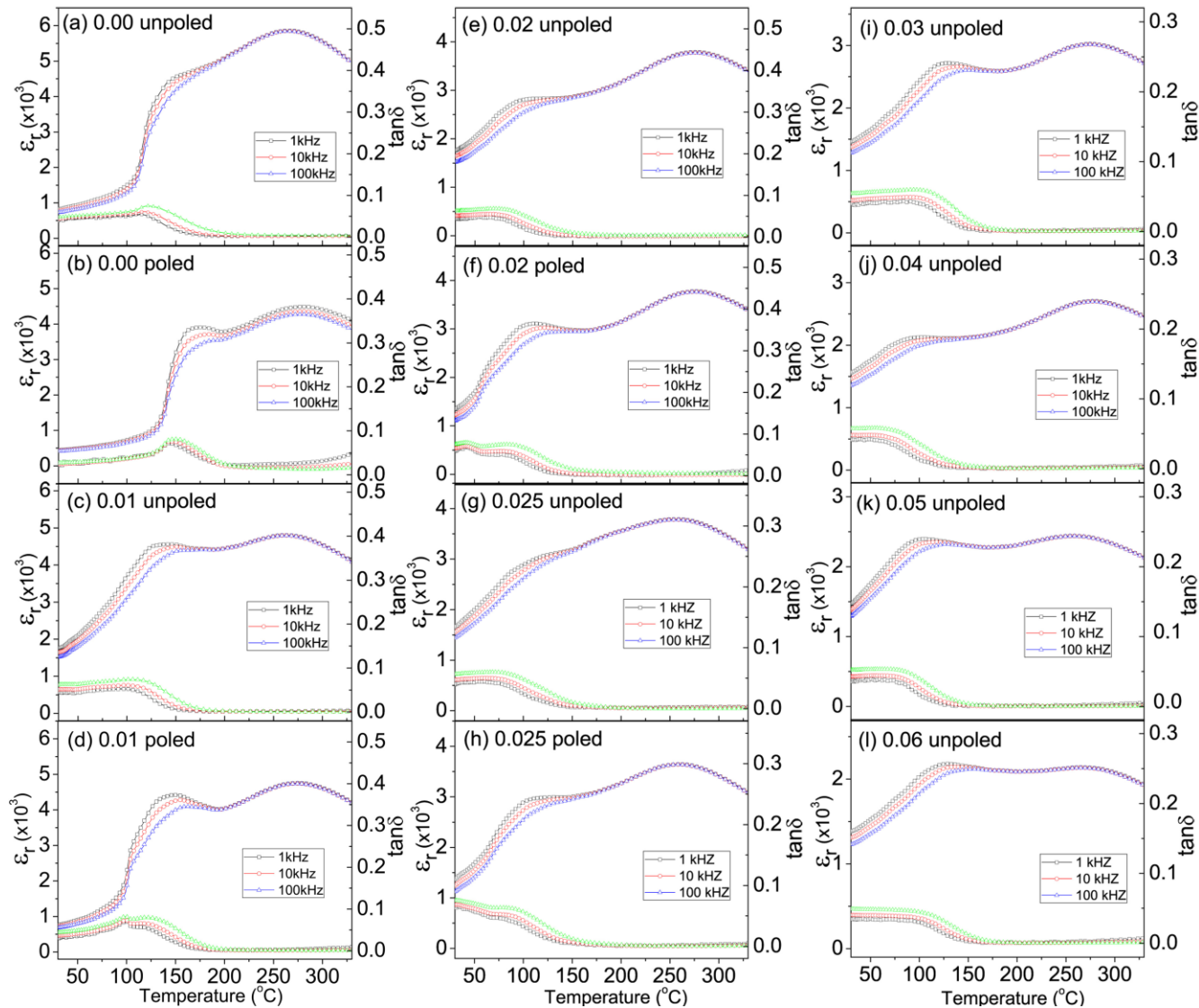
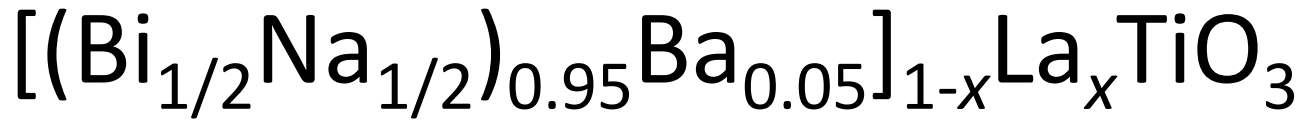


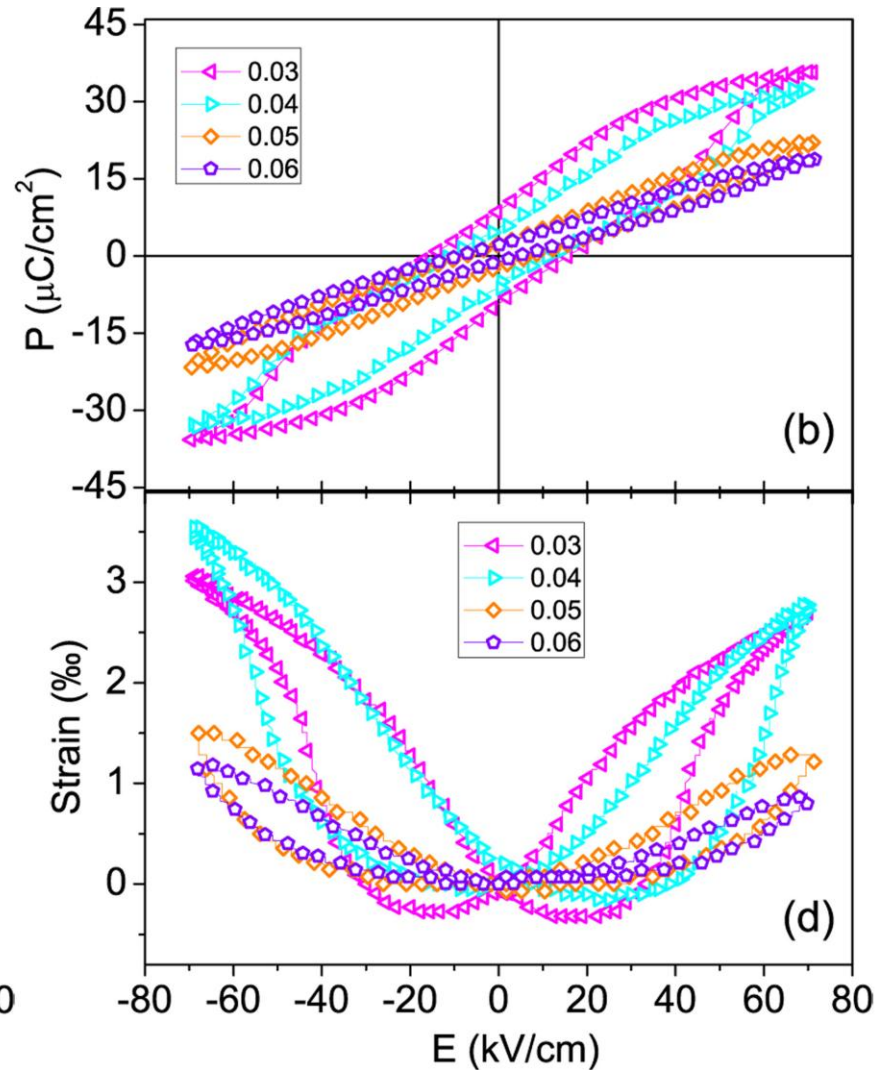
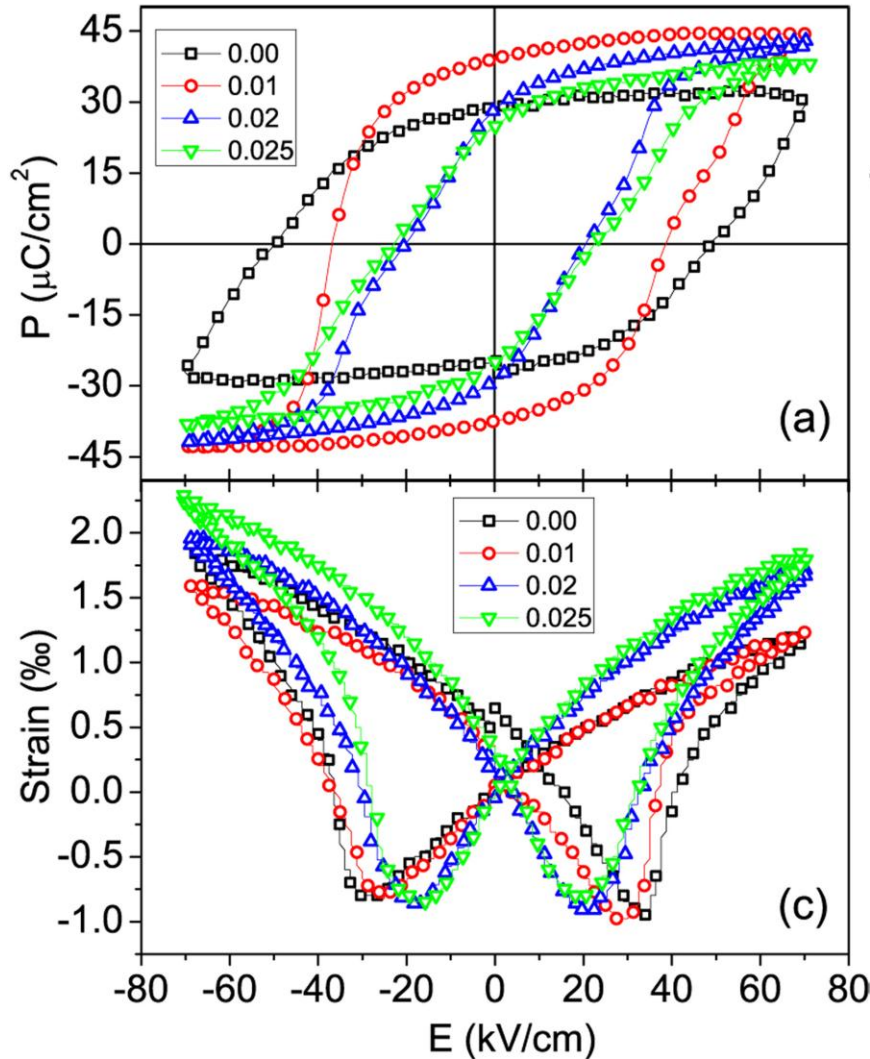
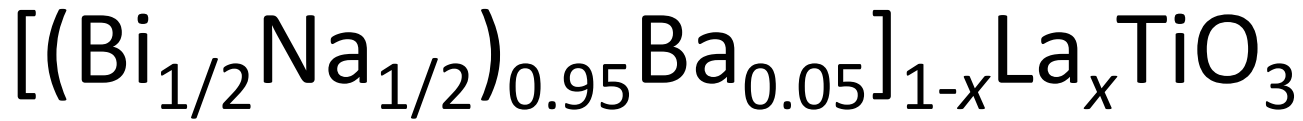
PNZST 43/8/2

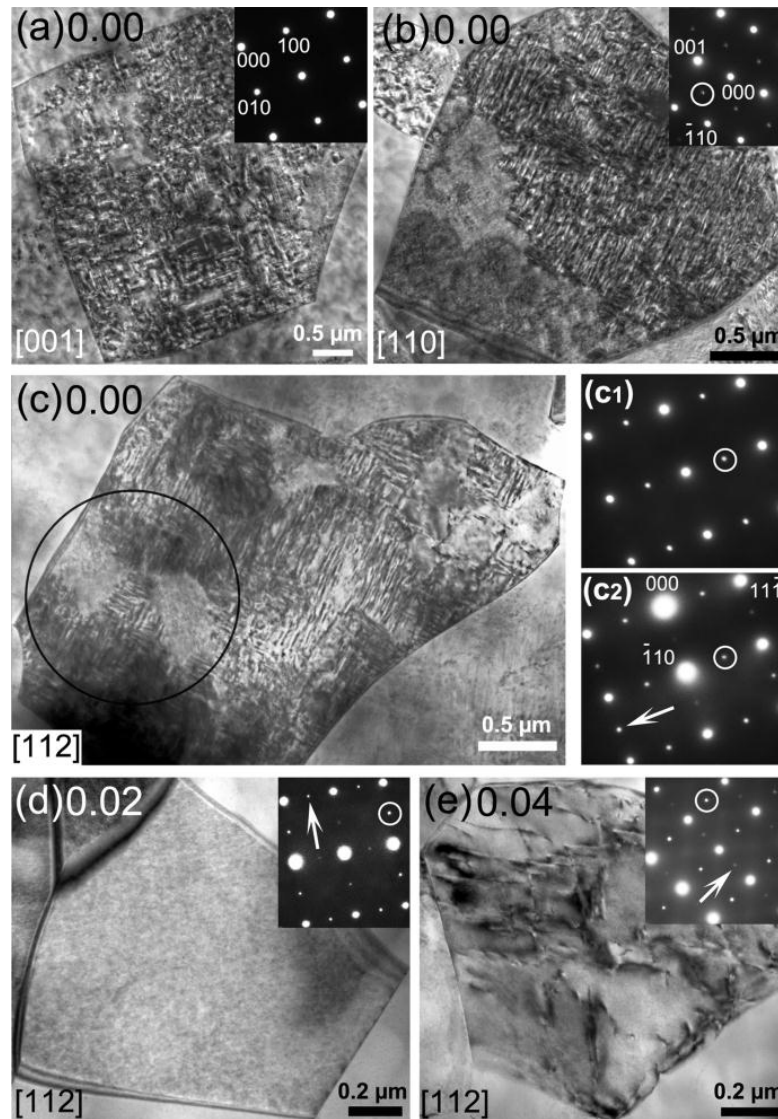
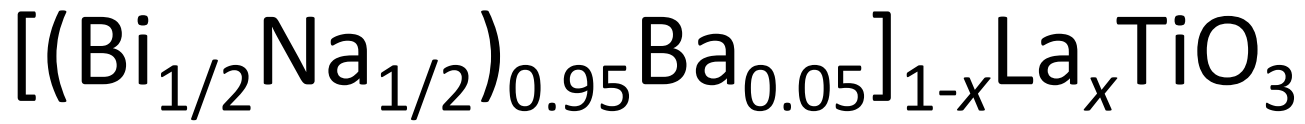


R.C. Miller and G. Weinreich, *Phys. Rev.* 117, 1460 (1960).

Y.H. Shin, I. Grinberg, I.W. Chen, and A.M. Rappe, *Nature* 449, 881 (2007).









# The **cycling** of ferroelectric polarization

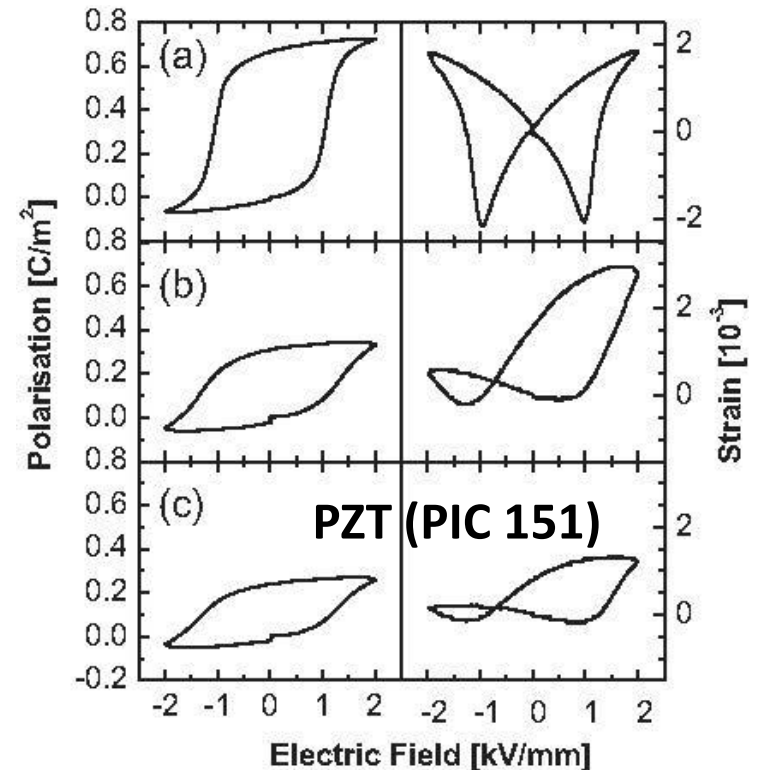
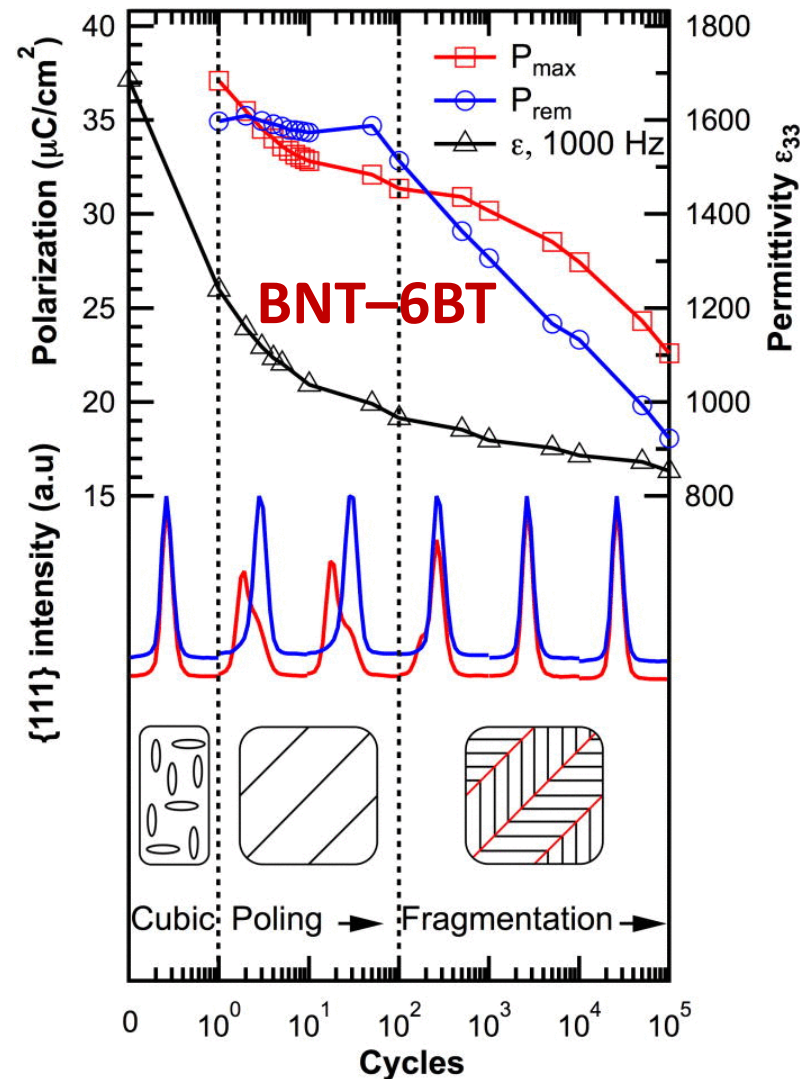
*J. Am. Ceram. Soc.*, 1 16 (2014)

DOI: 10.1111/jace.12811

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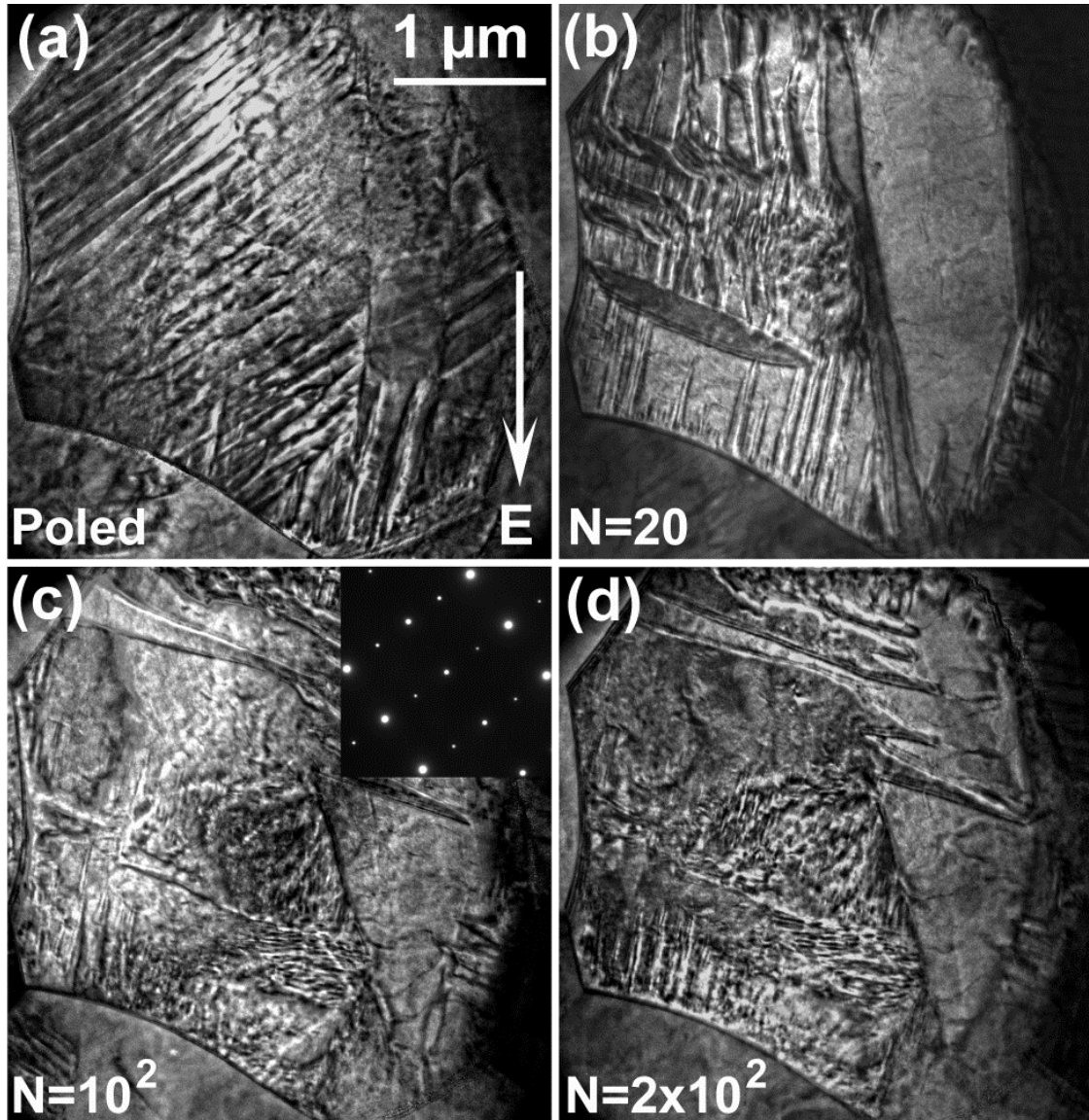
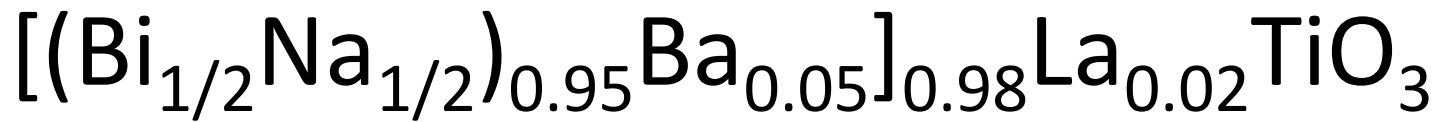
## Electric Fatigue of Lead-Free Piezoelectric Materials

Julia Glaum<sup>†</sup> and Mark Hoffman



D.C. Lupascu and J. Rödel, *Adv. Eng. Mater.* 7, 882 (2005).

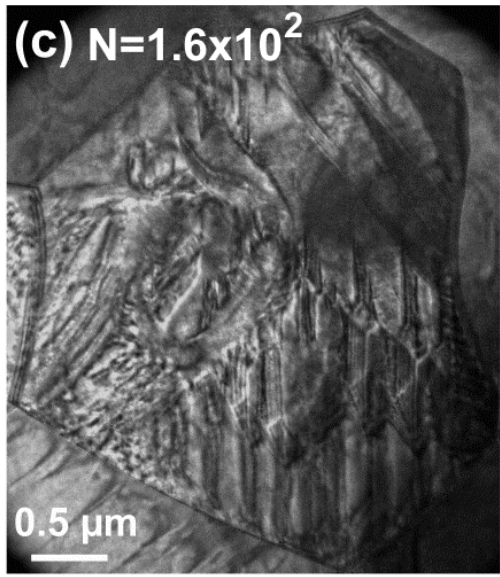
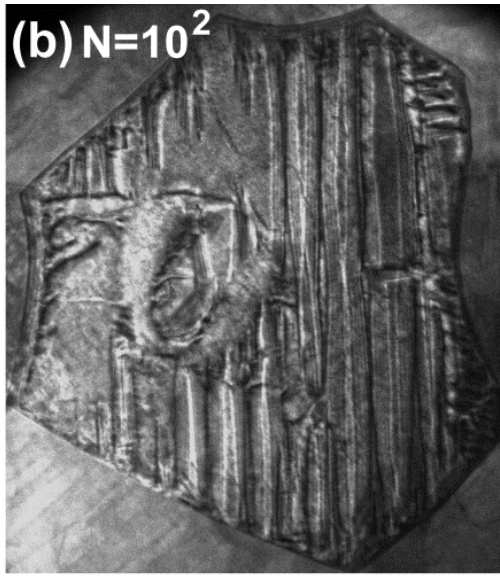
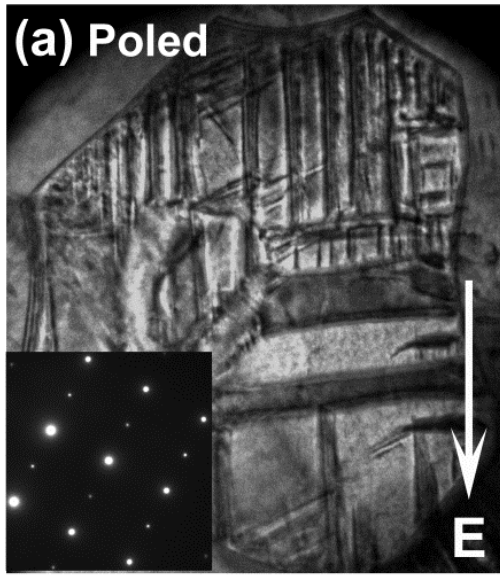
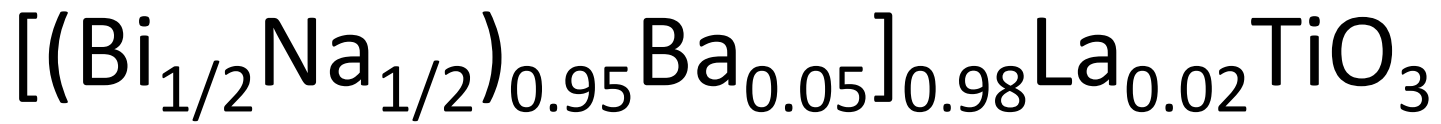
H. Simons, J. Glaum, J.E. Daniels, A.J. Studer, A. Liess, J. Rödel, and M. Hoffman, *J. Appl. Phys.* 112, 044101 (2012).



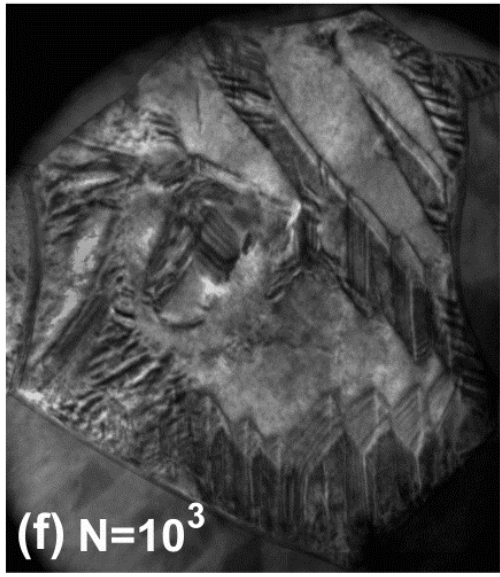
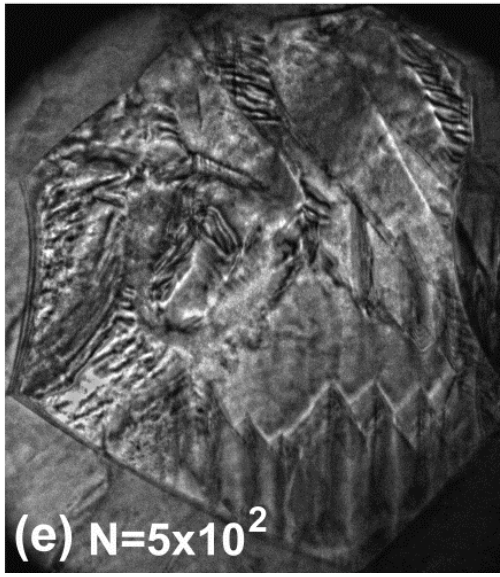
**ZA [112]**

**Cycling at  
 $\pm 30\text{kV/cm}$  at 1 Hz.**

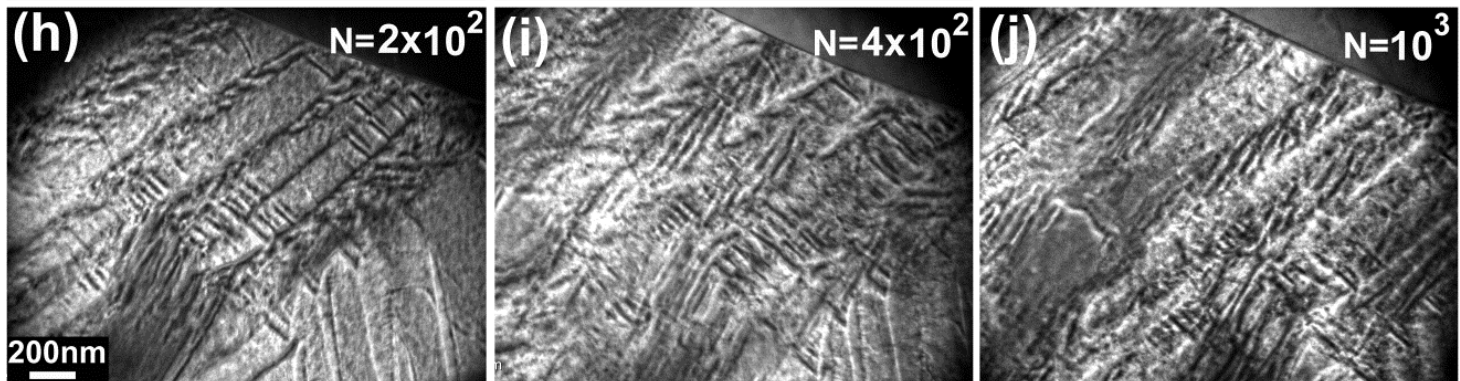
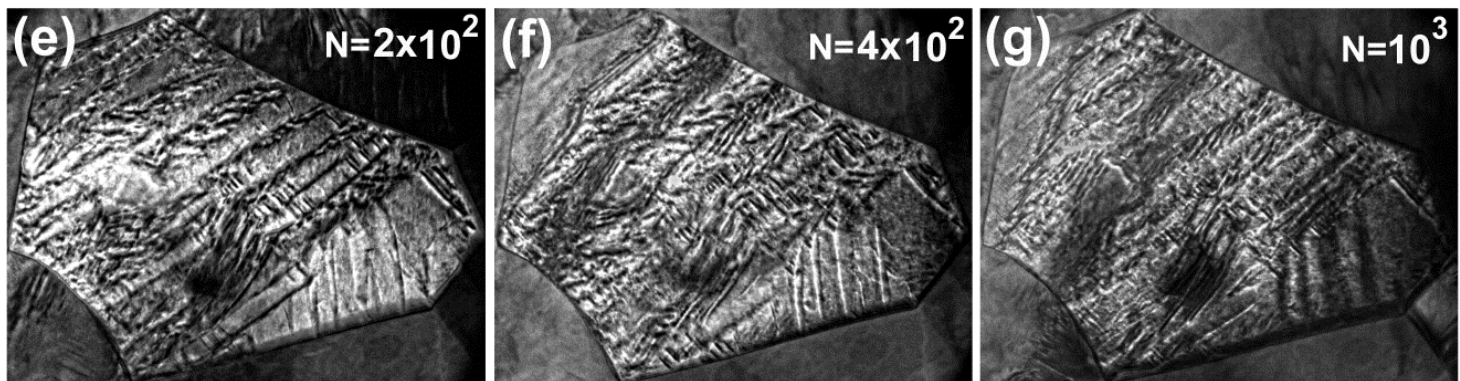
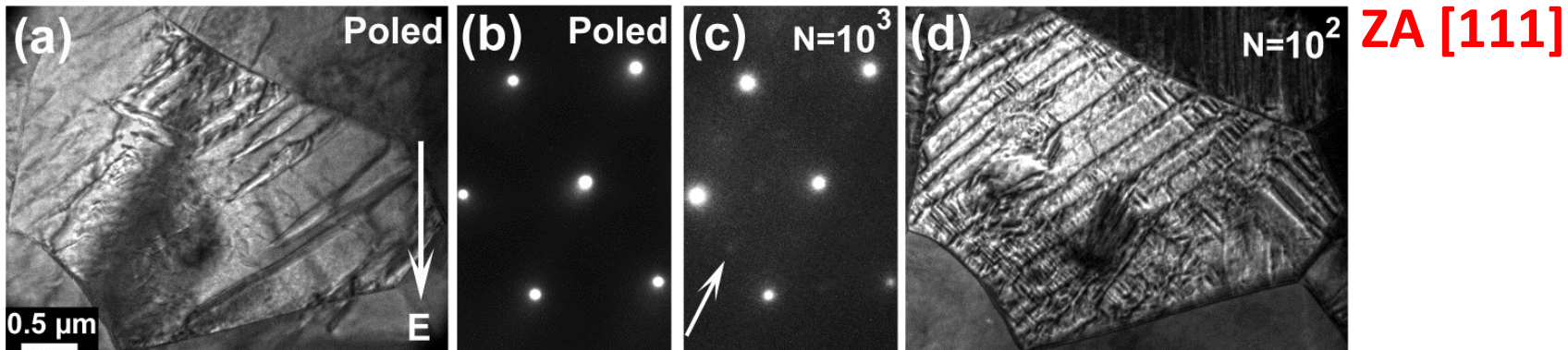
**Micrographs recorded  
at zero field.**



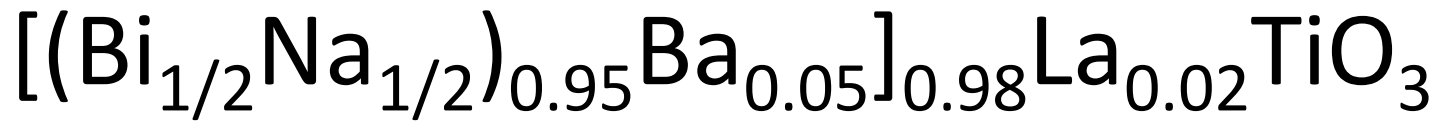
**ZA [112]**



**The same  
TEM  
specimen**

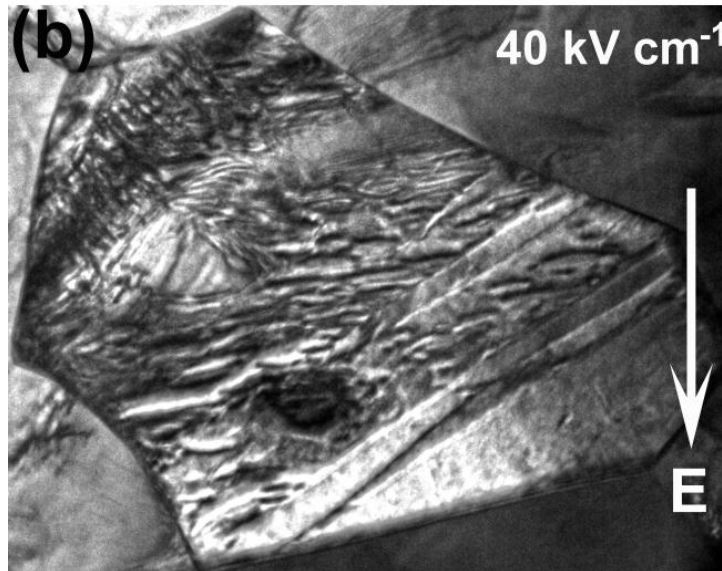


The same  
TEM  
specimen



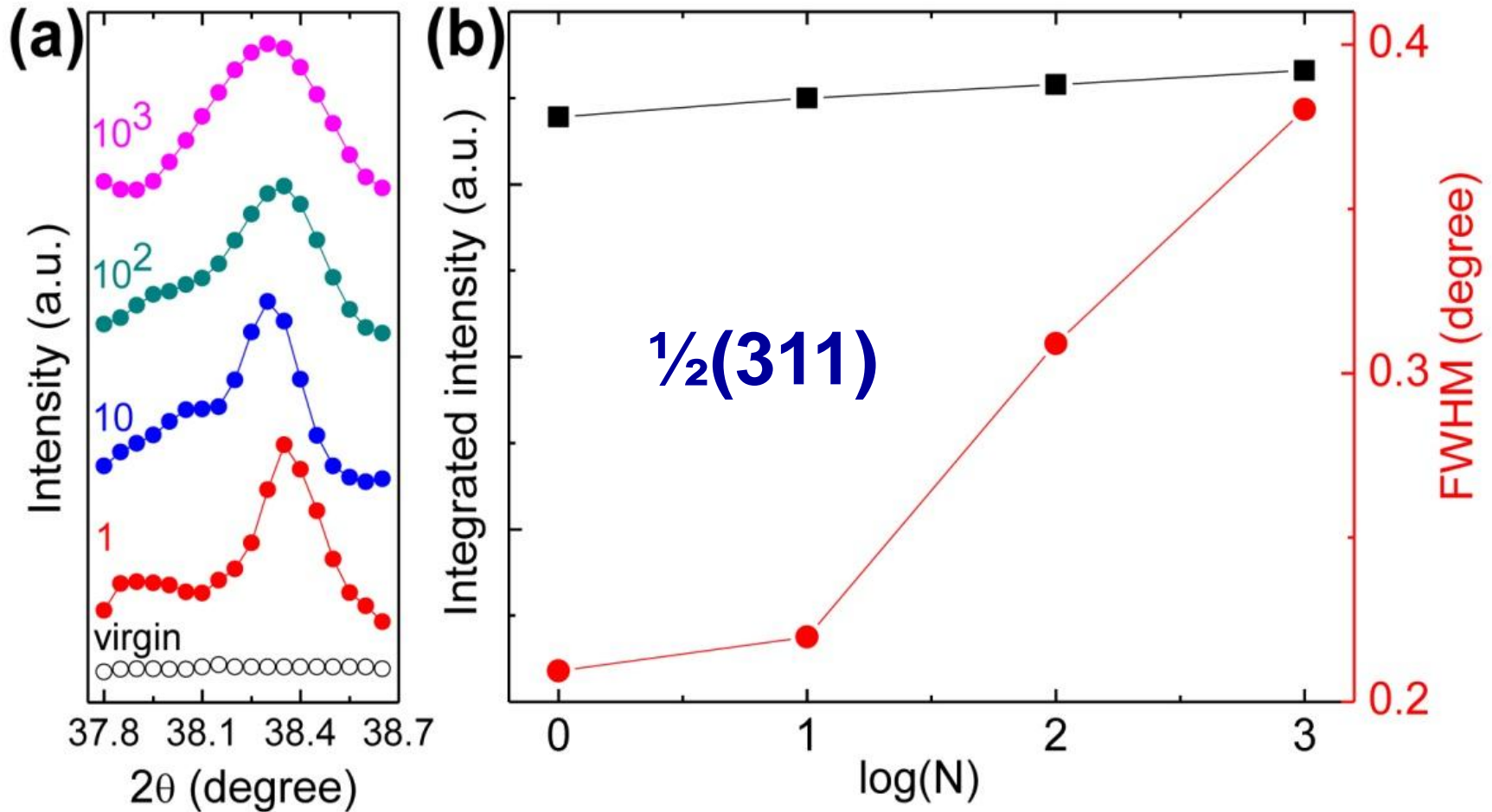
After initial poling  
at 30kV/cm

**ZA [111] (b)**



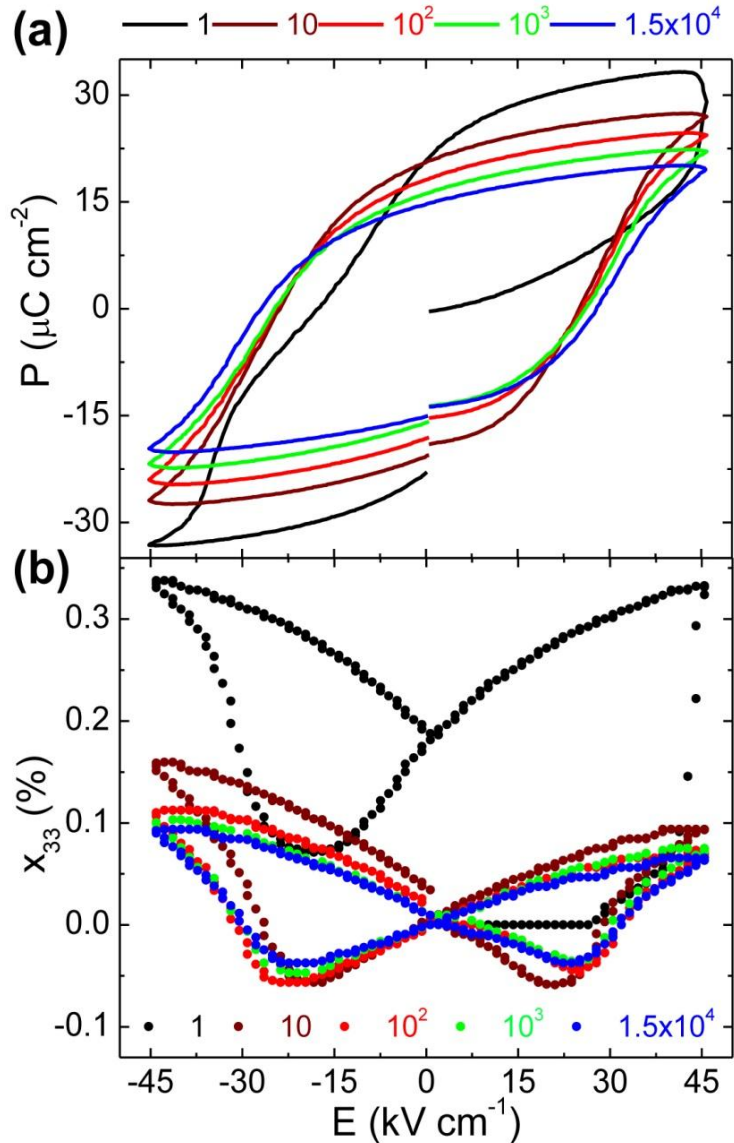
At 40kV/cm  
after  $10^3$  cycles  
at  $\pm 30\text{kV/cm}$

# X-ray diffraction on a bulk specimen

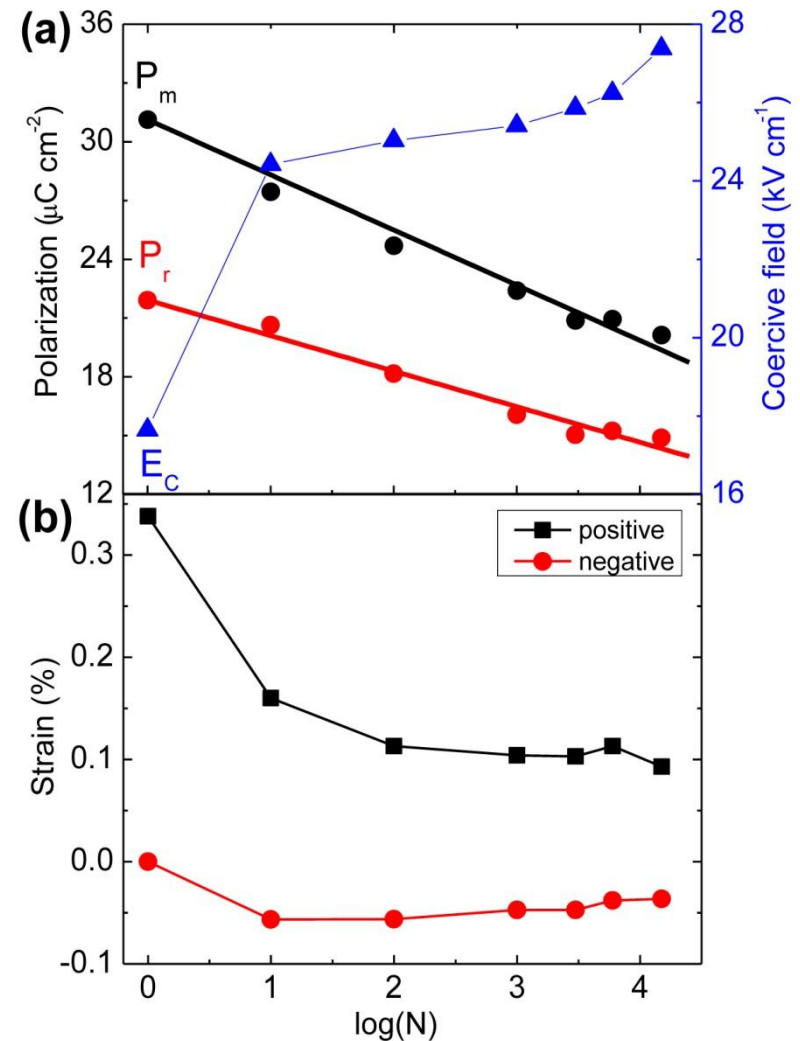


Estimated domain size reduces from 40 nm to 20 nm after  $10^3$  cycles ( $\pm 45$  kV/cm at 4Hz).

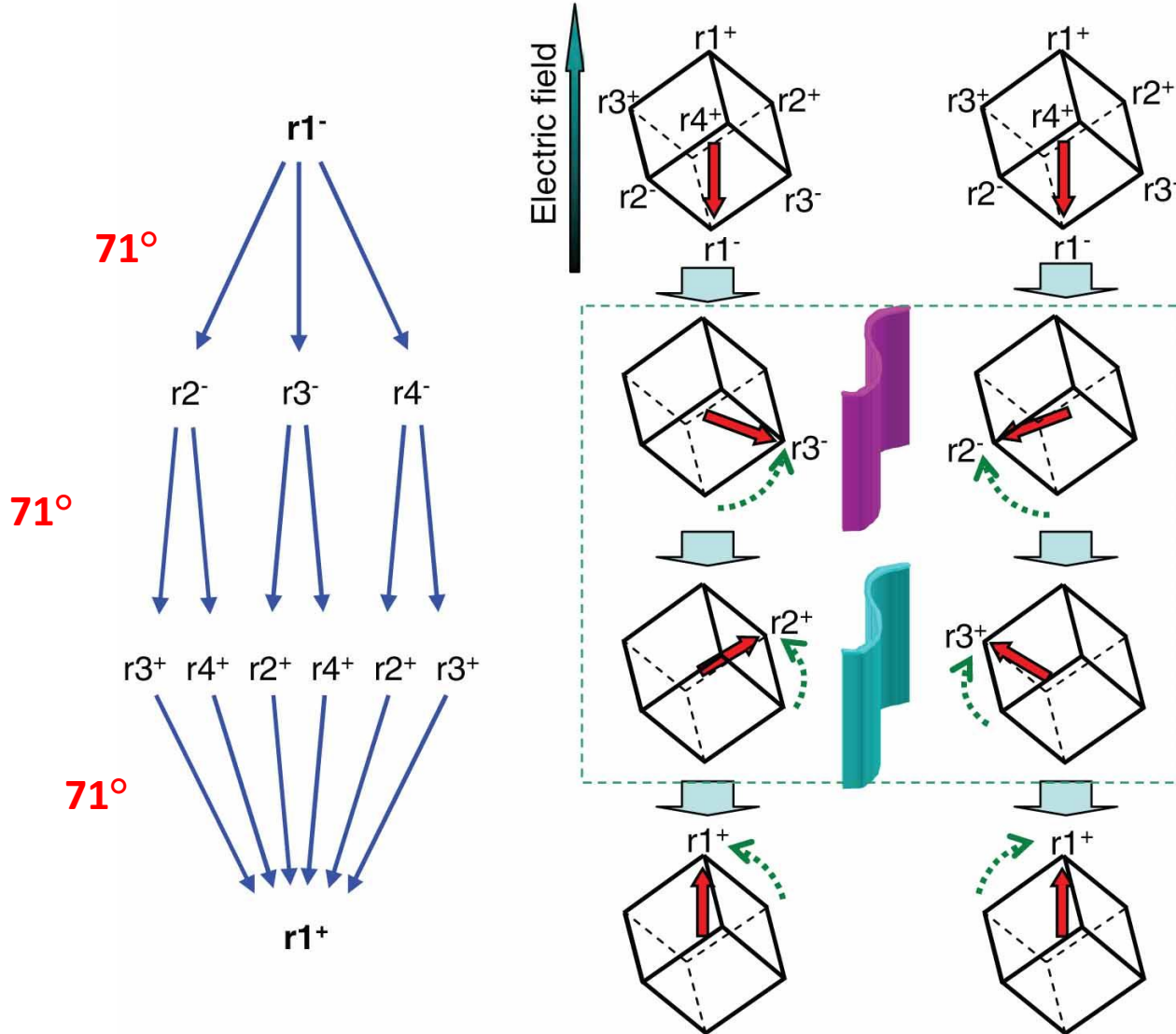
# Macroscopic behavior in bulk specimens



$$P = P_0 + A \log(N)$$



# Why it happens?



Kitanaka, Y. et al., *Phys. Rev. B* 89, 104104 (2014).

Baek, S.H. et al., *Adv. Mater.* 23, 1621 (2011).



# Conclusions

## $[(\text{Bi}_{1/2}\text{Na}_{1/2})_{1-x}\text{Ba}_x]\text{TiO}_3$ -based ceramics:

- Phase transitions occur during electrical **poling**.
- Phase transitions occur during polarization **reversing**.
- Domain fragmentation occurs during electrical **cycling**.

# Acknowledgements

- TEM access at Ames Laboratory, US-DOE.



- Financial support from National Science Foundation.



- Support and collaborations with Prof. Jürgen Rödel

