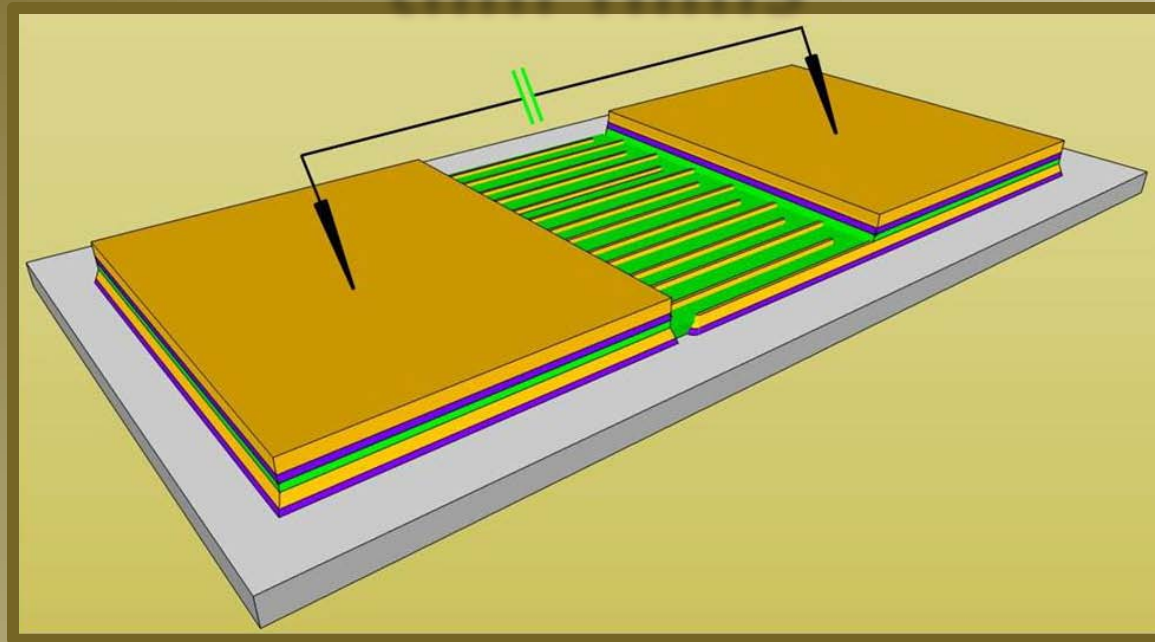


# Growth and characterization of multiferroic $\text{BiFeO}_3$ thin films



*Taimur Ahmed*

Chalmers University of Technology, Sweden

# Outline

- ▣ Background
  - Ferromagnetics
  - Ferroelectrics
  - Multiferroics
    - ▣  $\text{BiFeO}_3$  (BFO)
- ▣ Design & modeling of dielectric response
- ▣ Device fabrication
- ▣ Characterization & Analysis
- ▣ Conclusion
- ▣ Future outlook

# Background – Ferromagnetics

- ▣ Ferromagnetic materials
  - Materials retain their magnetic polarization
  - Curie-Weiss law (susceptibility is inverse dependent on Curie temperature)

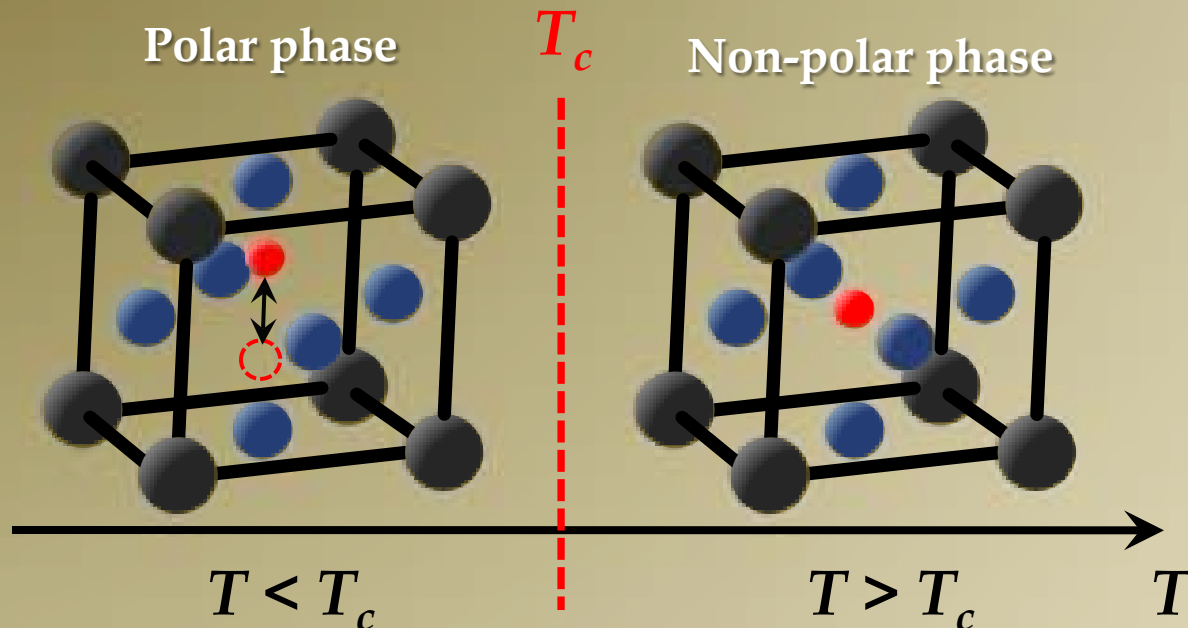
$$\chi = \frac{C}{(T - T_c)}$$

*C = Curie constant of material*  
*T = Absolute temperature*  
*T<sub>c</sub> = Curie temperature*

- ▣  $T > T_c$ : Ferromagnetics changes to paramagnetics
- ▣  $T = T_c$ : Material is in phase transition

# Background – Ferroelectrics

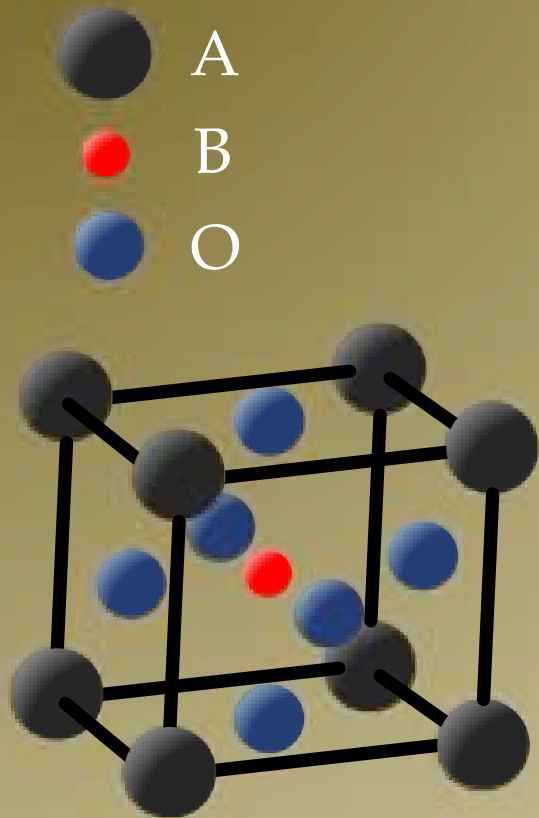
- ▣ Ferroelectric materials
  - Materials retain their electric (dipole) polarization
  - Curie temperature defines
    - ▣ Polar (ferroelectric) and non-polar (paraelectric) phase



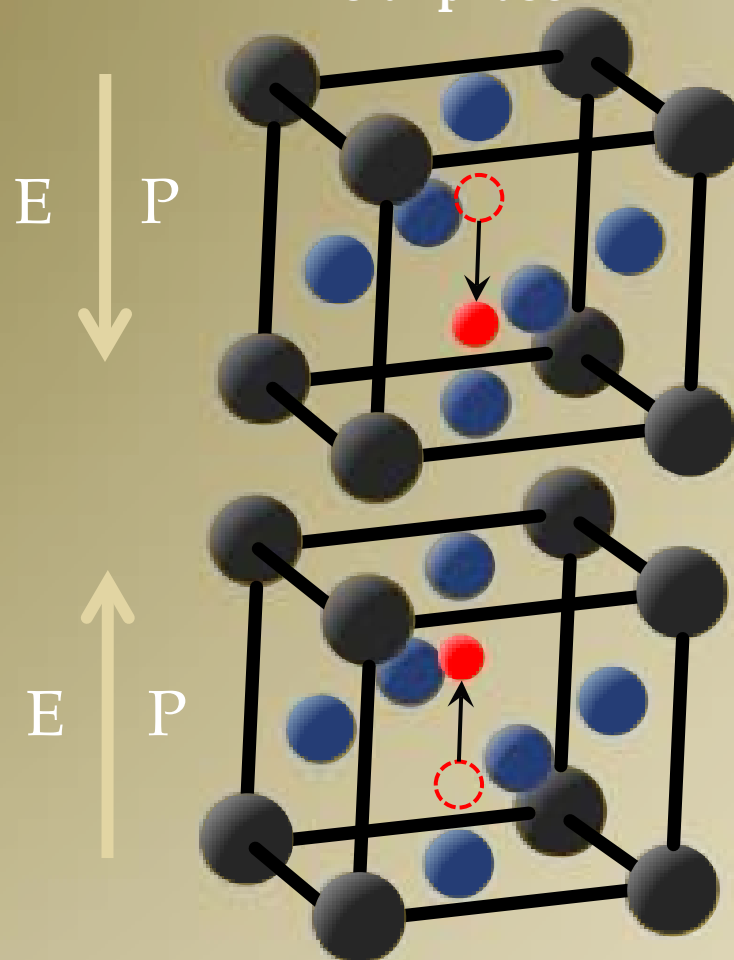
# Background – Ferroelectrics

## Perovskite - $\text{ABO}_3$

Non-polar phase



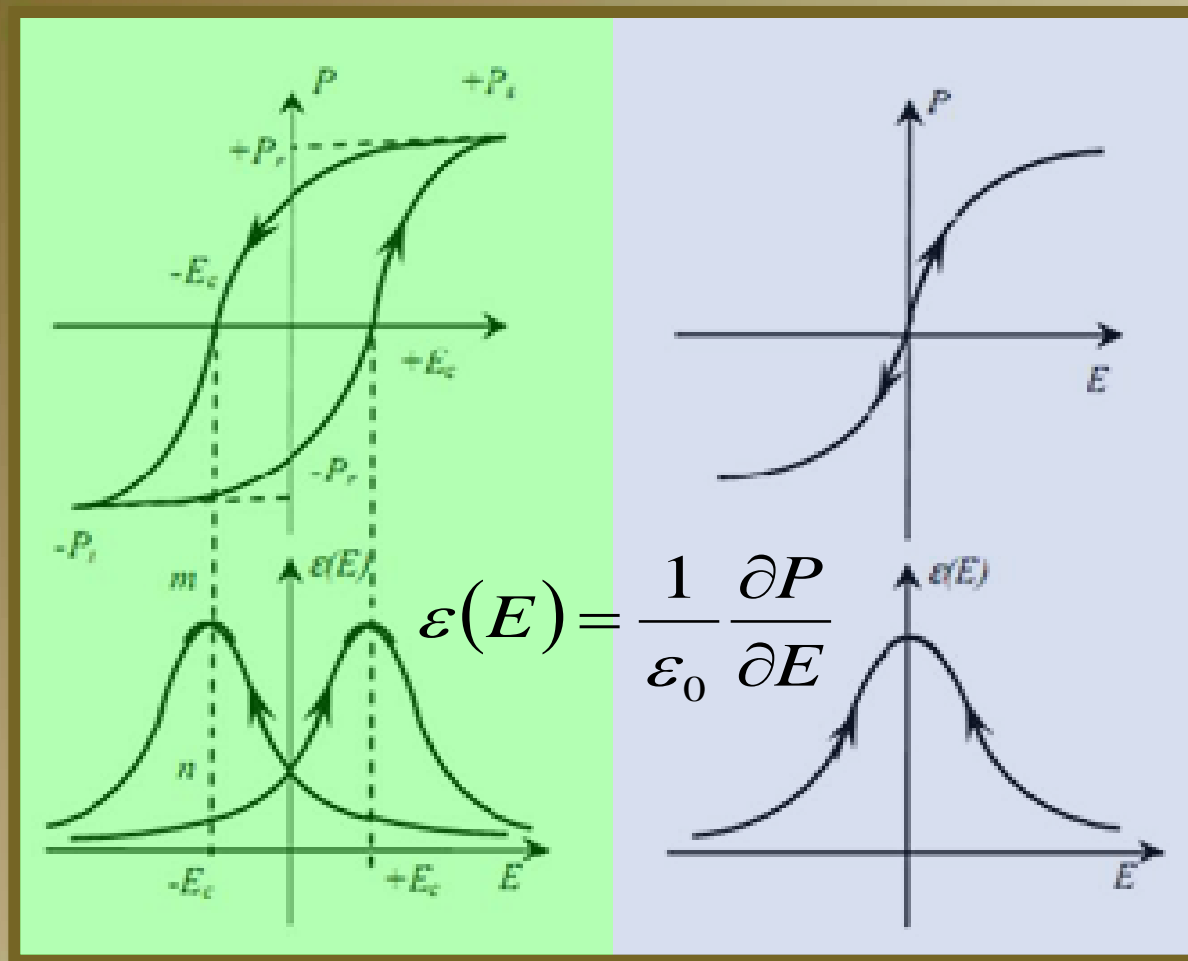
Polar phase



# Background – Ferroelectrics

Polar phase

Non-polar phase

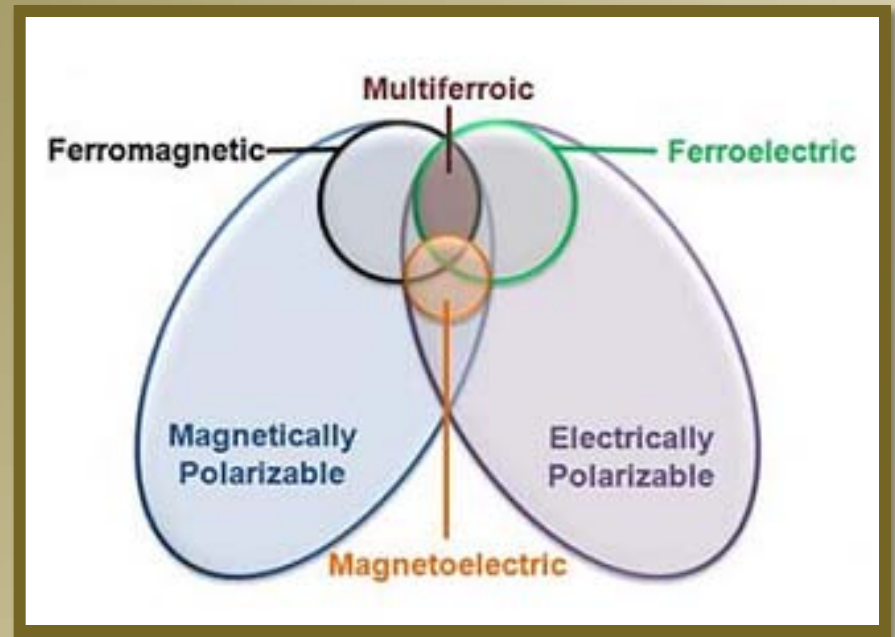


# Background – Multiferroics

- ▣ Hans Schmid (1990):  
“A material that combines two (or more) of the primary ferroic orders in one phase”

ME = Ferromagnetic  
+  
Ferroelectric

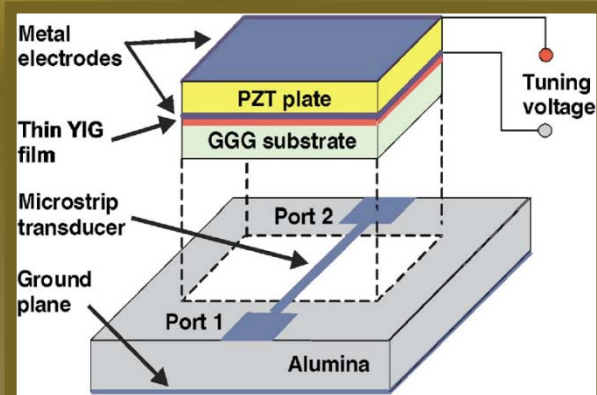
- ▣ In practice often:  
Multiferroic = Magnetoelectric



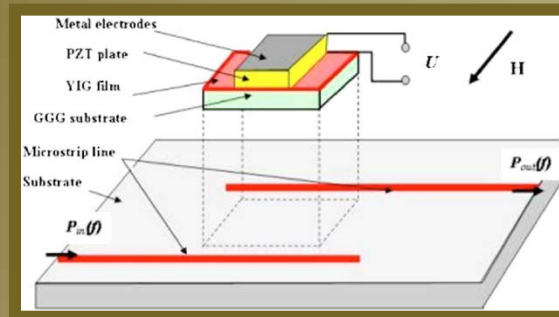


# Device Level Applications

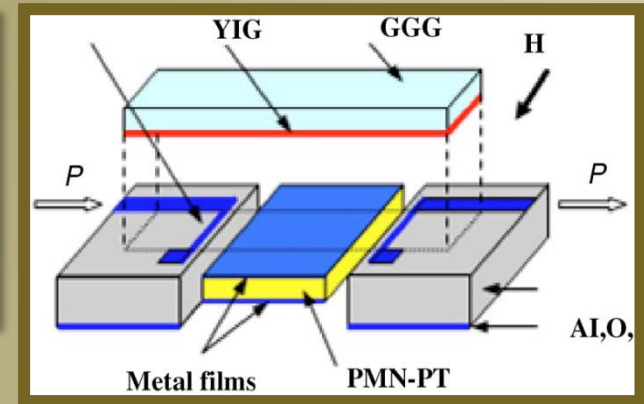
## Tunable resonator



## Tunable filter



## Tunable phase shifter/delay line

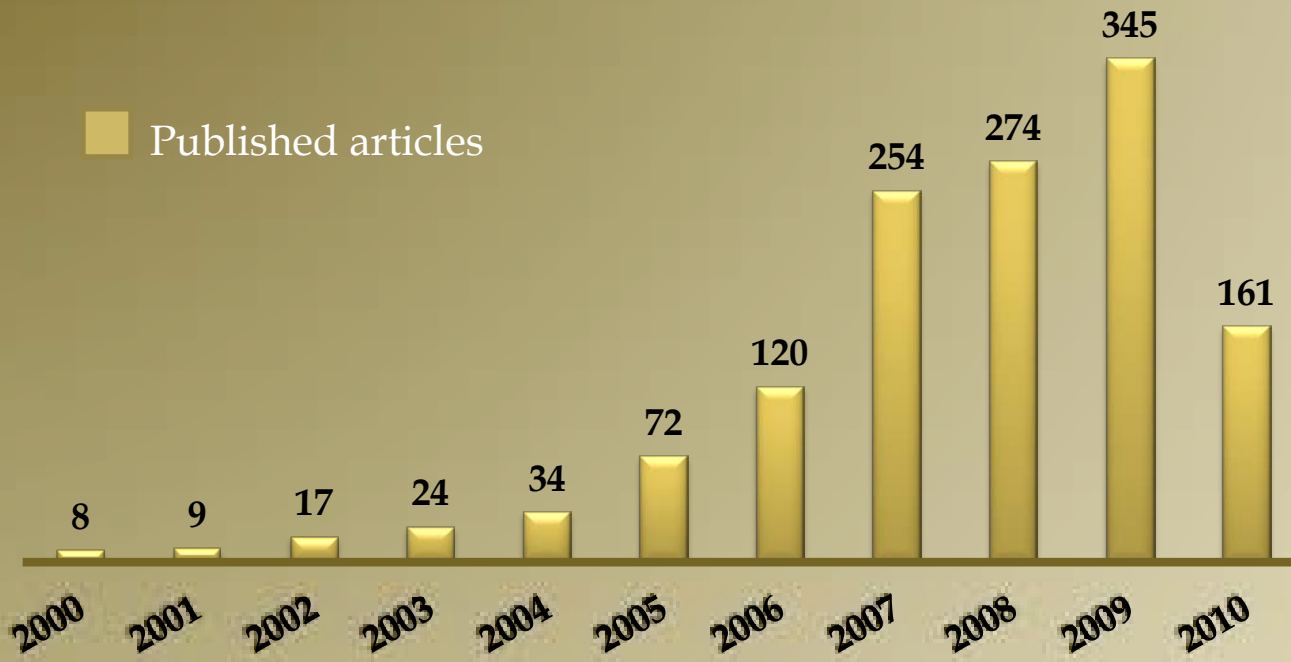


- ▣ Magnetoelectric memories
- ▣ Magneto-mechanical actuators
- ▣ Tunable microwave devices
- ▣ Sensors



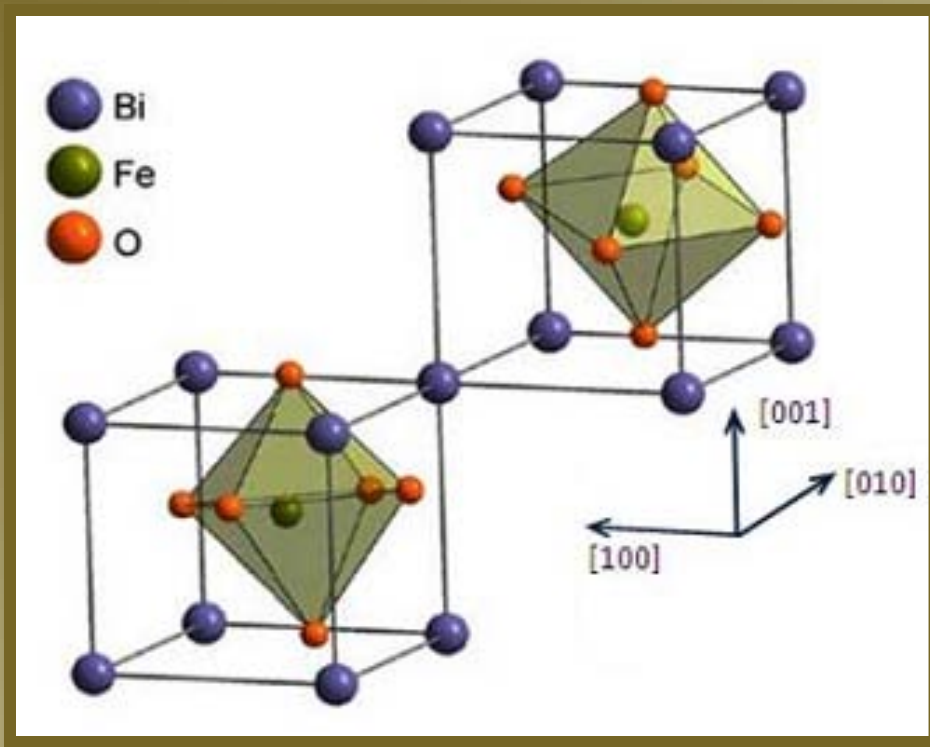
# Background – BiFeO<sub>3</sub>

- ▣ Bismuth ferrite – BiFeO<sub>3</sub> (BFO)
  - High Curie temperature,  $T_C$  (850°C)

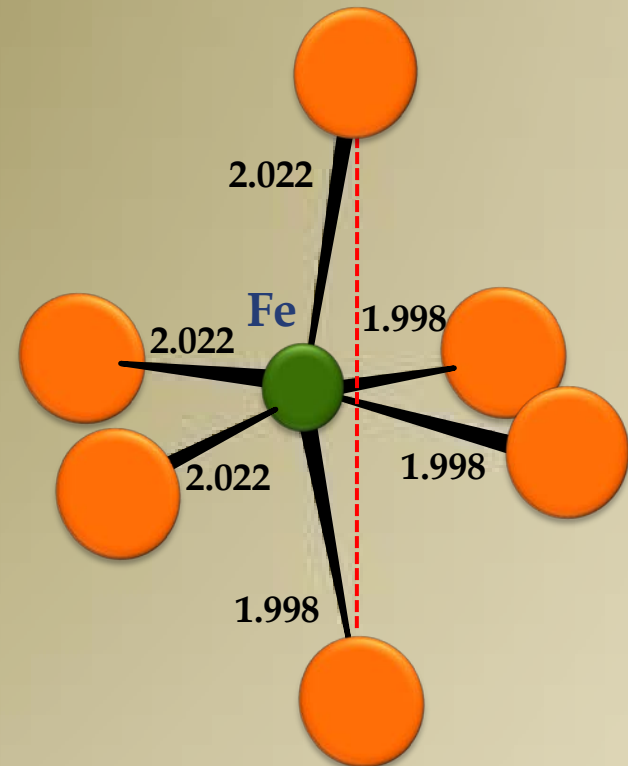


# BiFeO<sub>3</sub>: Crystal structure

Distorted Rhombohedral Perovskite



Oxygen octahedron



# BiFeO<sub>3</sub> films: Challenges

- ▣ Leakage current
  - Non-stoichiometric thin films (Bi depletion)
  - Microstructural defects (grain boundaries *etc.*)
- ▣ Weak magnetoelectric coupling

# Objective

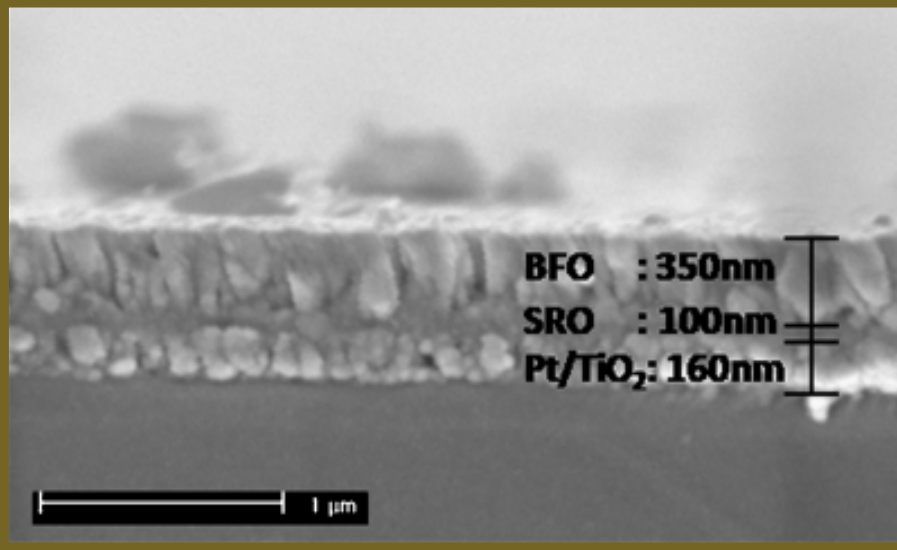
- ▣ Optimization of growth temperature of  $\text{BiFeO}_3$  (BFO) thin films
  - To have stoichiometric thin films
  - To have low leakage current

# Outline

- ▣ Background
- ▣ Design & modeling
  - Design configurations
    - ▣ Grain structure of  $\text{BiFeO}_3$  films
    - ▣ Parallel plate configuration
    - ▣ Co-planar configuration
    - ▣ Co-planar configuration with buried ID electrode
  - Permittivity models
- ▣ Device fabrication
- ▣ Characterization & Analysis
- ▣ Conclusion

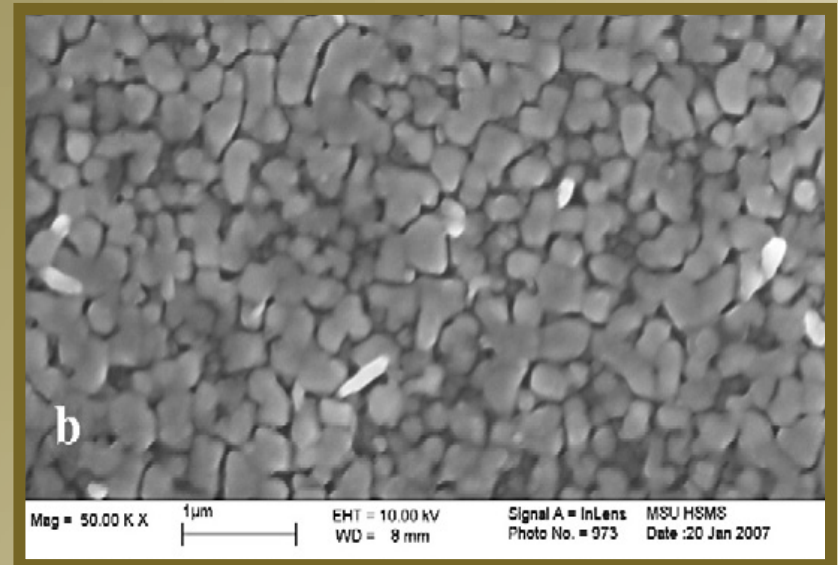
# Grain structure of $\text{BiFeO}_3$ films

## RF sputtering



R. Zheng et al. *J. Am. Ceram. Soc.* 91 (2008) 463.

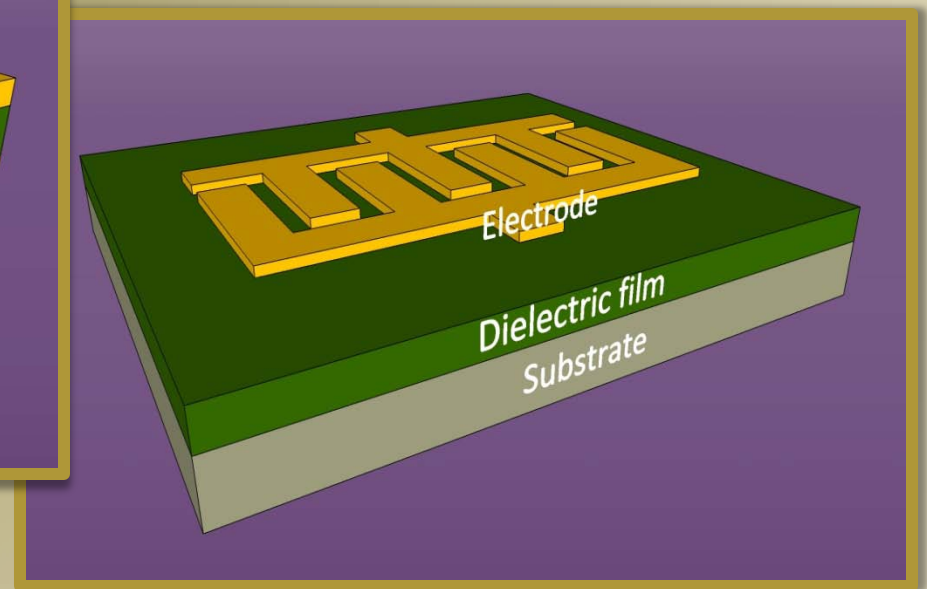
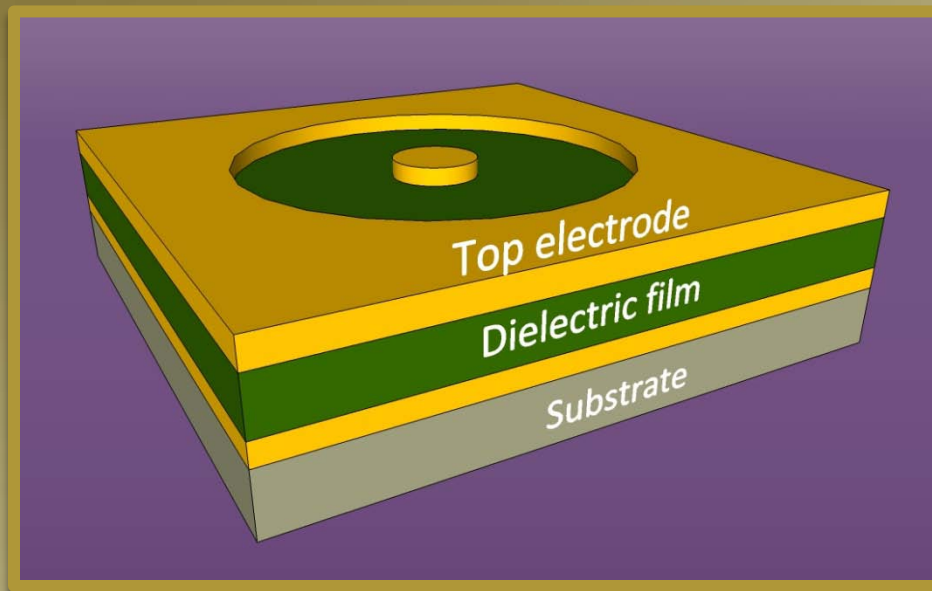
## MOCVD



M.S. Kartavtseva et al. *Surface and Coatings Technology* 201 (2007) 9149.

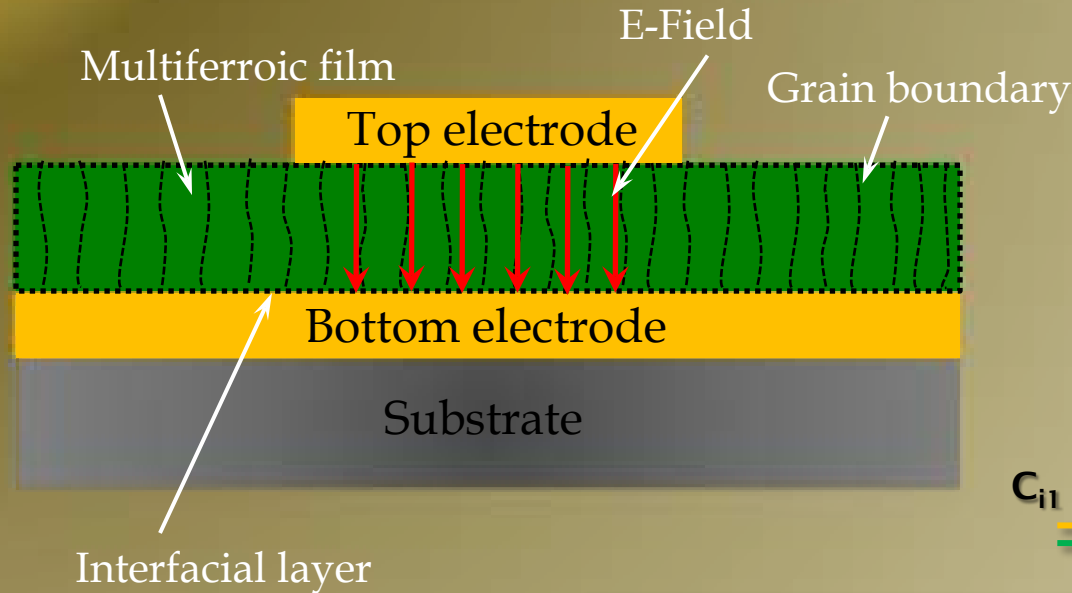
# Design & modeling of dielectric response

- ▣ Design configurations
  - Parallel plate configuration
  - Co-planar inter digital (ID) configuration





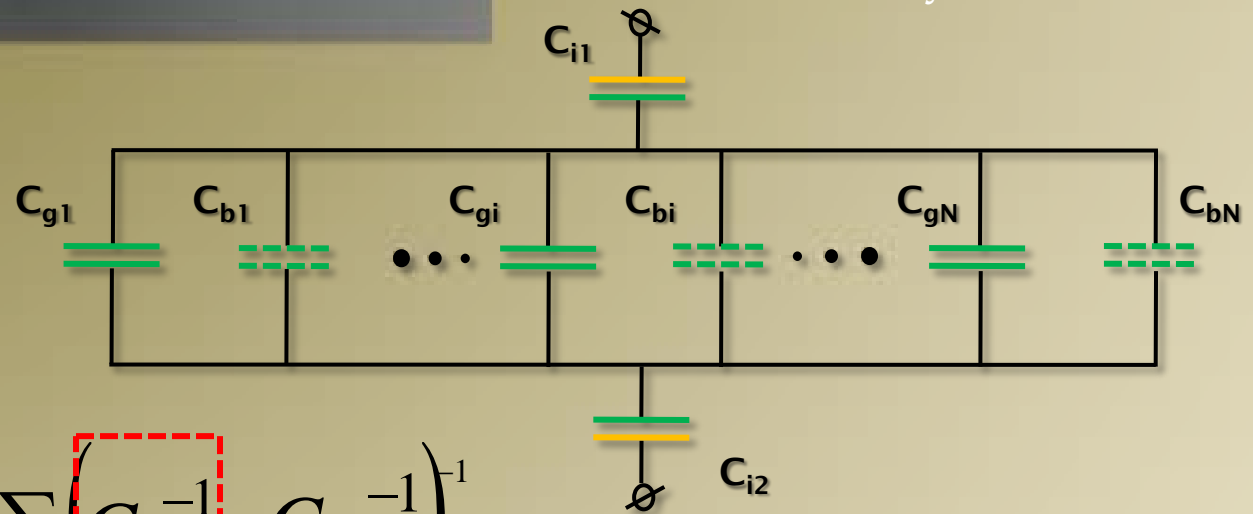
# Parallel plate configuration



$C_b$  = Capacitance of grain boundaries

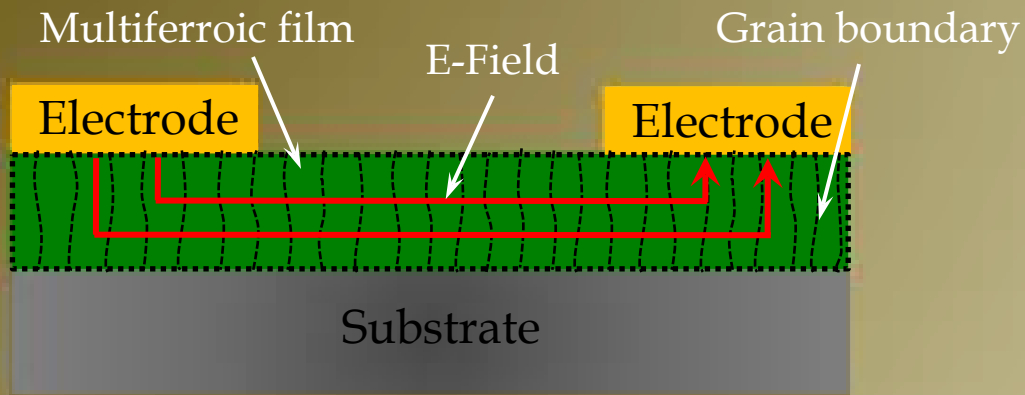
$C_g$  = Capacitance of grains

$C_i$  = Capacitance of interface layers



$$C_{eq} = \sum C_b + \sum \left( C_i^{-1} + C_g^{-1} \right)^{-1}$$

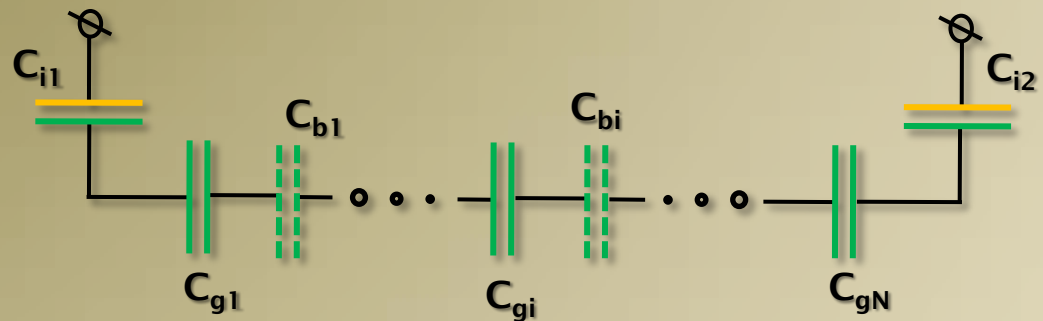
# Co-planar configuration



$C_b$  = Capacitance of grain boundaries

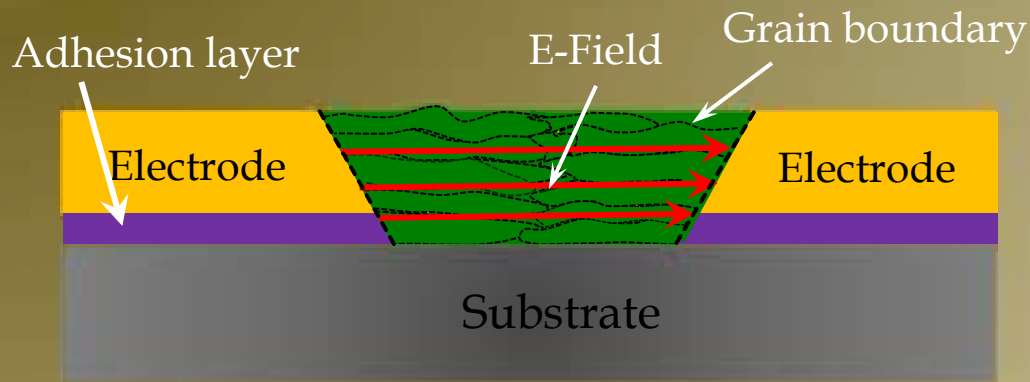
$C_g$  = Capacitance of grains

$C_i$  = Capacitance of interface layers



$$C_{eq}^{-1} = \left[ \sum C_b^{-1} + \sum C_i^{-1} \right] + \sum C_g^{-1}$$

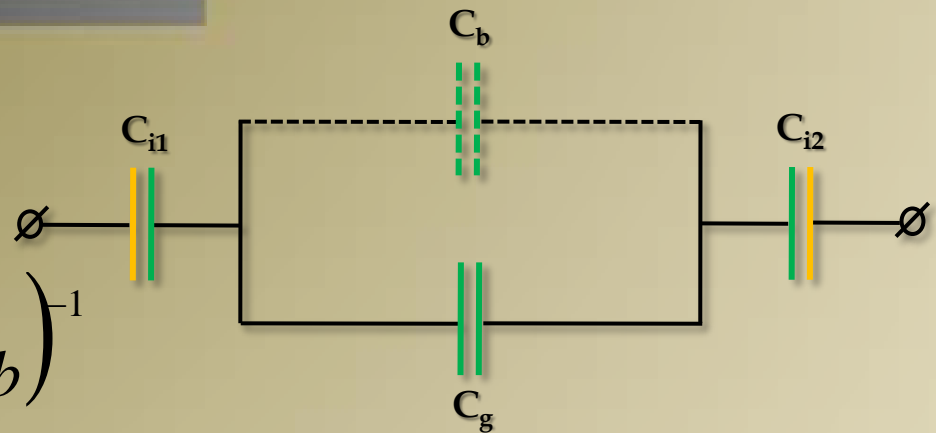
# Co-planar configuration: buried electrodes



$C_b$  = Capacitance of grain boundaries

$C_g$  = Capacitance of grains

$C_i$  = Capacitance of interface layers



$$C_{eq}^{-1} = \sum C_i^{-1} + \sum (C_{g+b})^{-1}$$

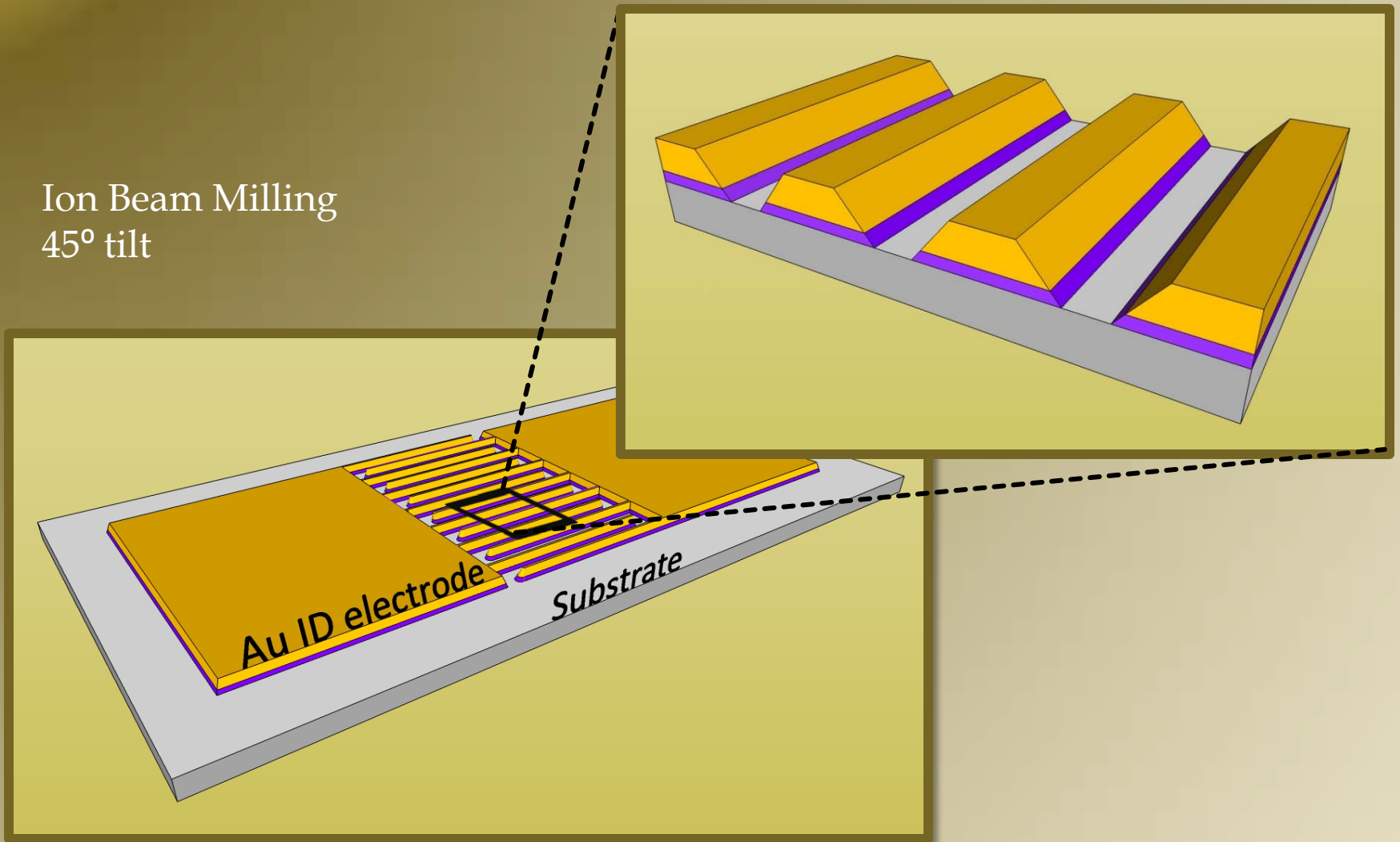
$$C_{eq}^{-1} \approx \sum C_g^{-1}$$

# Outline

- ▣ Objective
- ▣ Background
- ▣ Design & modeling of dielectric response
- ▣ Device micro fabrication
  - Patterning of IDC electrodes
  - BFO thin film growth (PLD)
  - Contact pads deposition
- ▣ Characterization & Analysis
- ▣ Conclusion
- ▣ Future outlook

# ID electrode patterning

Ion Beam Milling  
45° tilt



# BiFeO<sub>3</sub> thin film growth

## ▣ Pulsed Laser Deposition (PLD)

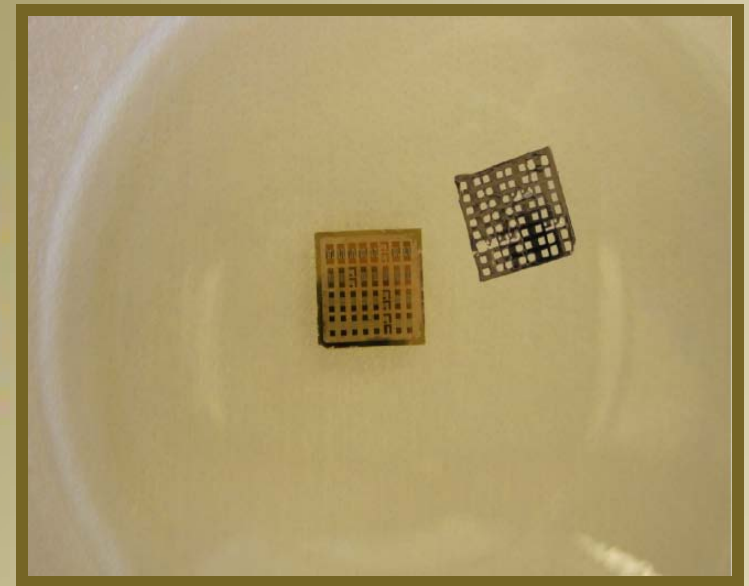
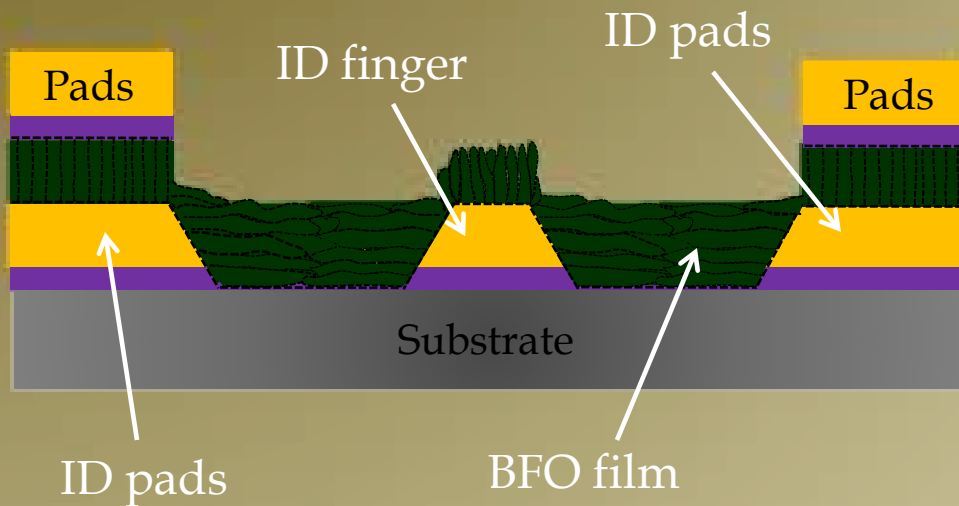
Parameters	Comments
Laser source	KrF
Laser wavelength	248 nm
Energy	1.5 mJ/cm <sup>2</sup>
Target	Bi <sub>1.1</sub> FeO <sub>3</sub>
Oxygen pressure	1 x 10 <sup>-2</sup> mbar
Repetition rate	10 Hz
Substrate	Au/Ti/SiO <sub>2</sub>
<b>Substrate temperature</b>	<b>500°C -750°C</b>
Substrate-target distance	6cm

# Contact pad deposition & lift-off

E-beam evaporator

Ti: 50 nm

Au: 500 nm



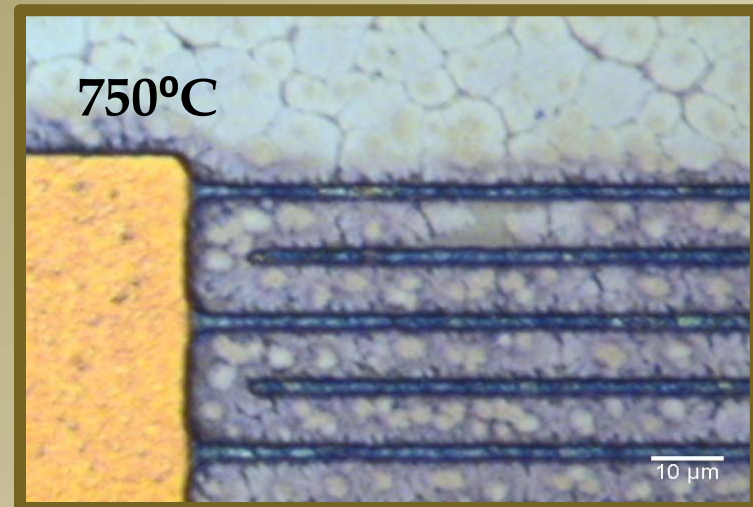
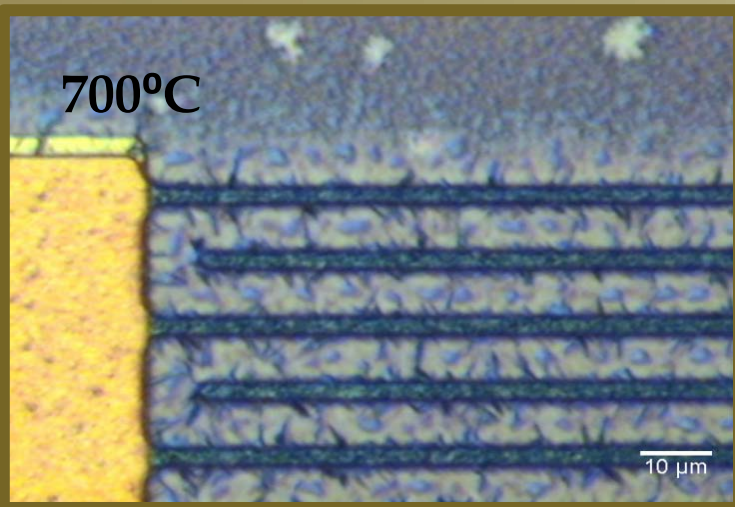
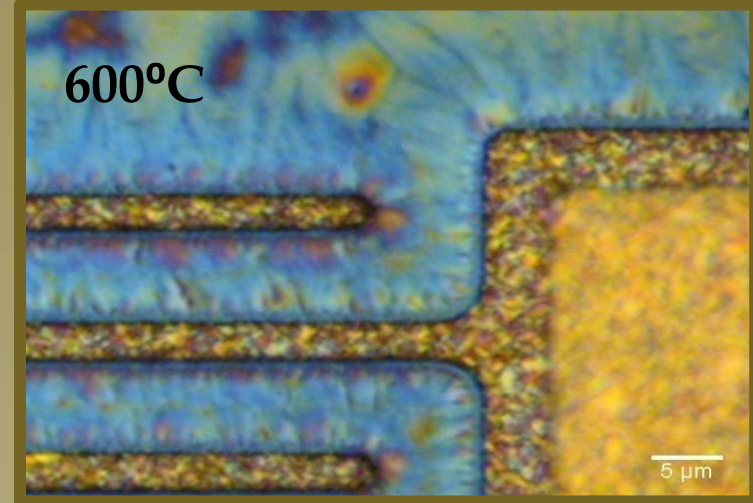
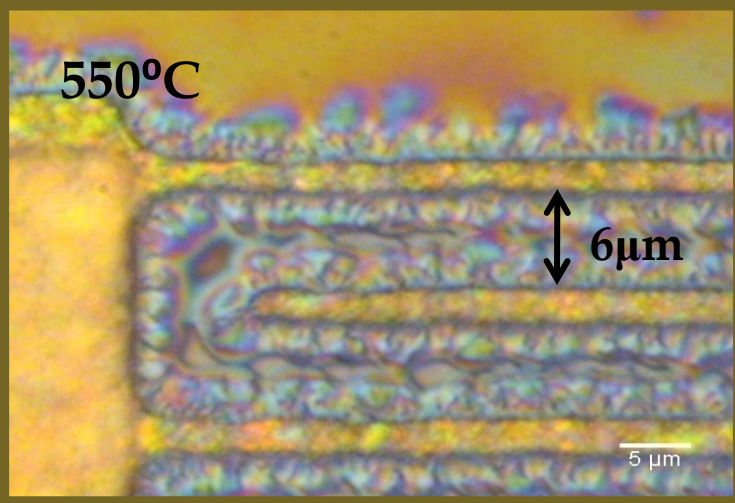


# Outline

- ▣ Background
- ▣ Design & modeling of dielectric response
- ▣ Device fabrication
- ▣ Characterization & Analysis
  - Microstructure of deposited thin films
  - Dielectric response
  - Magnetoelectric response
- ▣ Conclusion

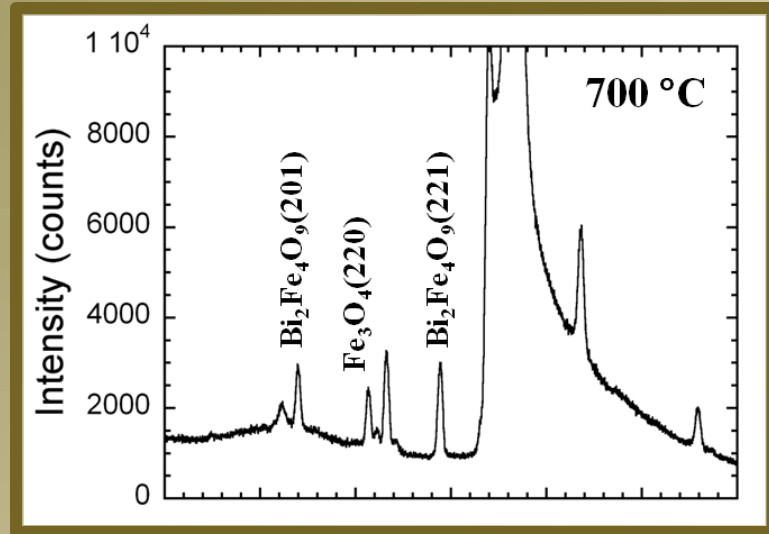
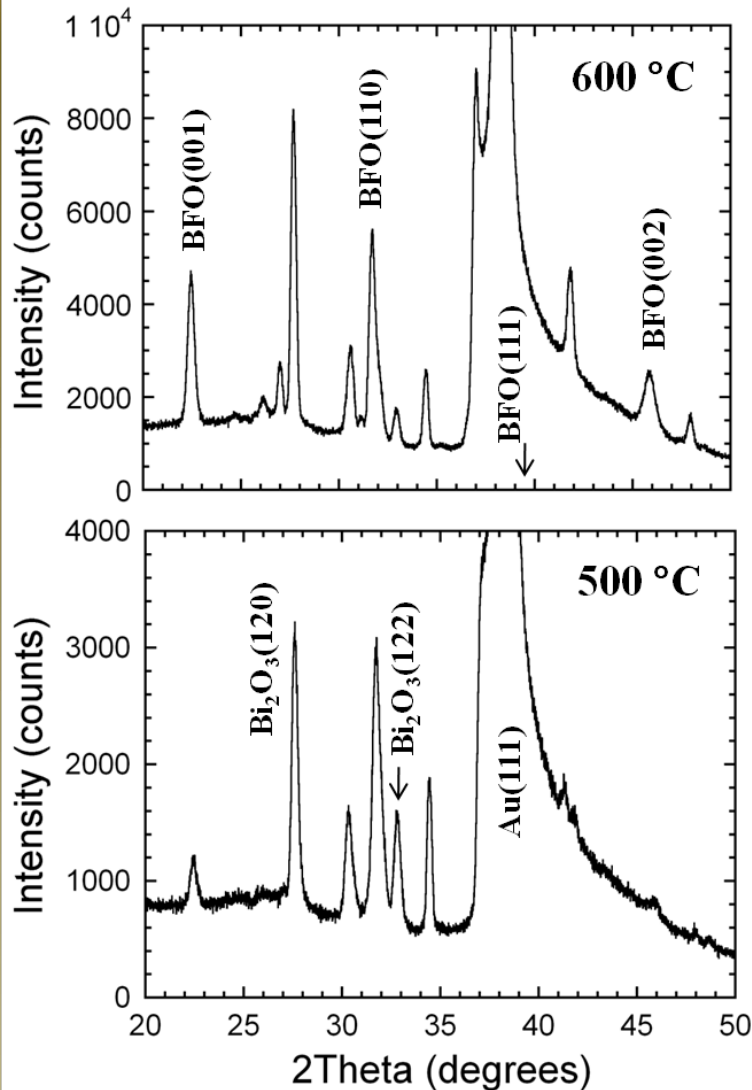
# Microstructure

## As-deposited films



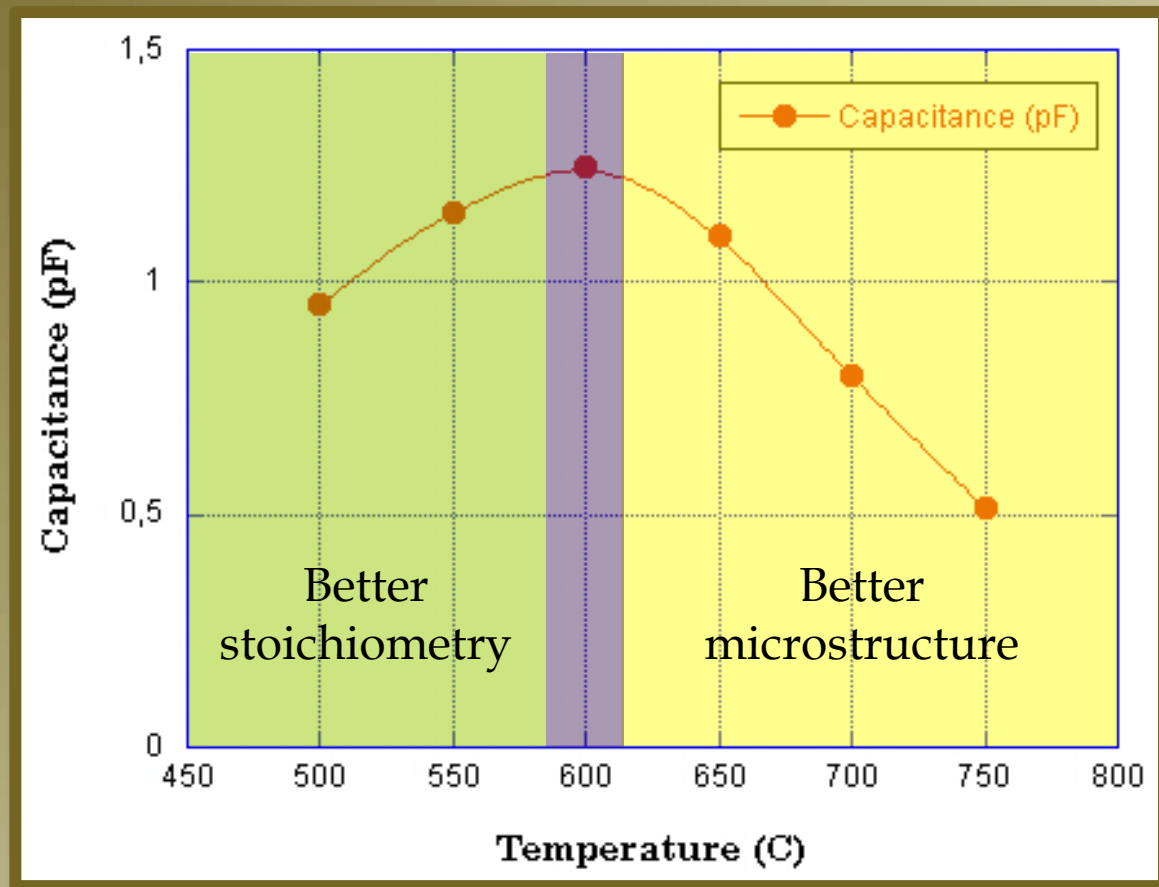
# Stoichiometry As-deposited films

## XRD Spectra



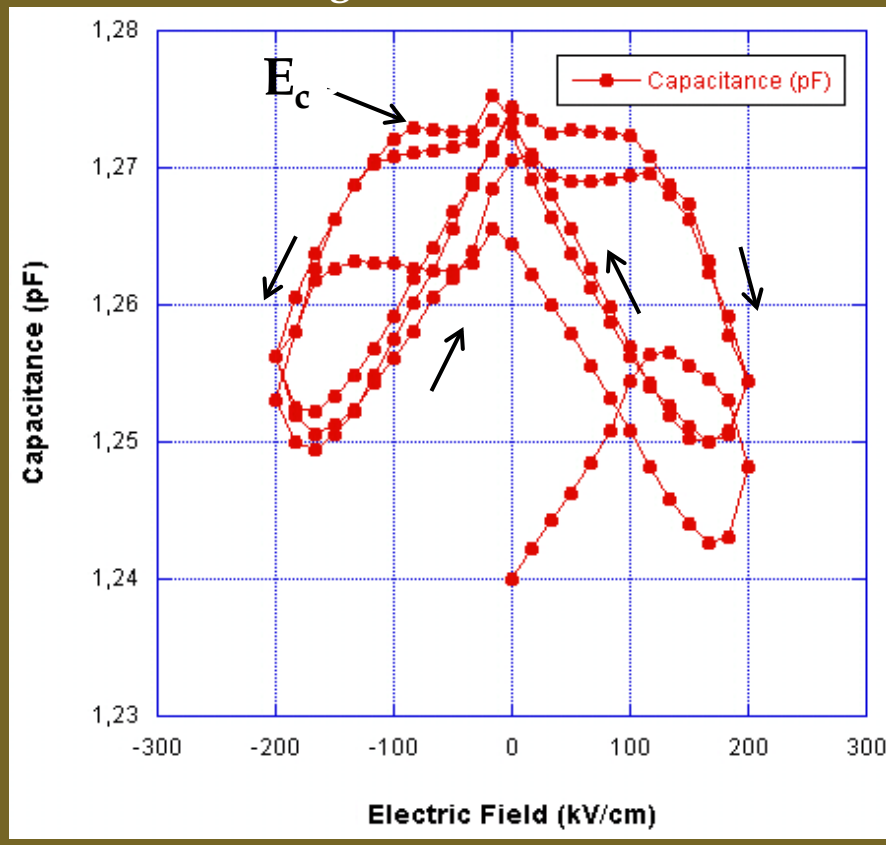
# Dielectric Response

As – deposited BFO thin films

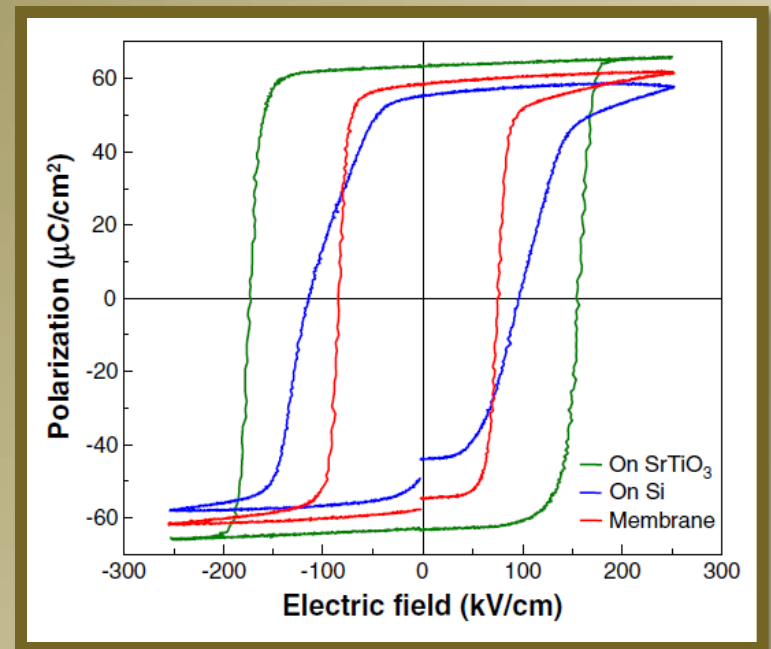


# C(V) response

$$T_g = 600^{\circ}\text{C}$$



## Epitaxial BiFeO<sub>3</sub> thin films



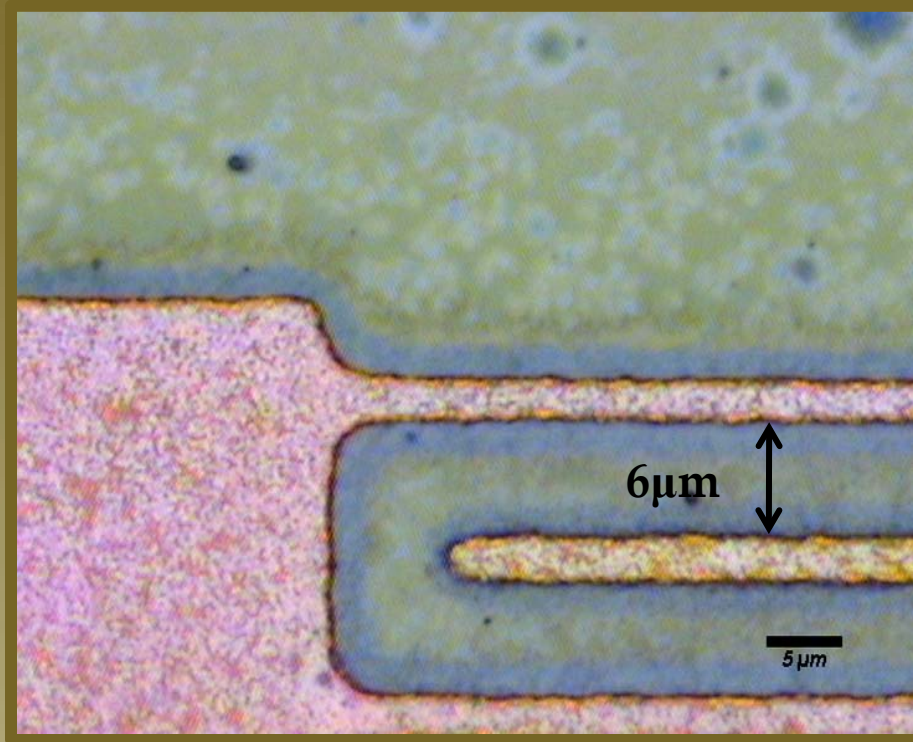
H. W. Jang et al. Phys. Rev. Lett. 01  
(2008) 107602-1



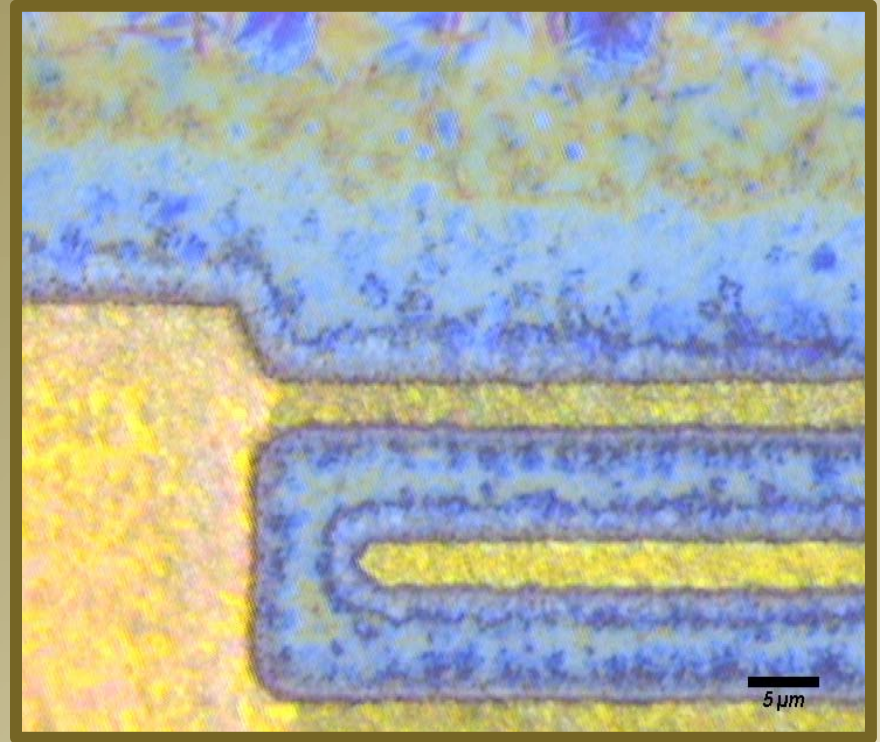
# Microstructure

## Ex-situ annealed films

As deposited  $T_g = 500^{\circ}\text{C}$

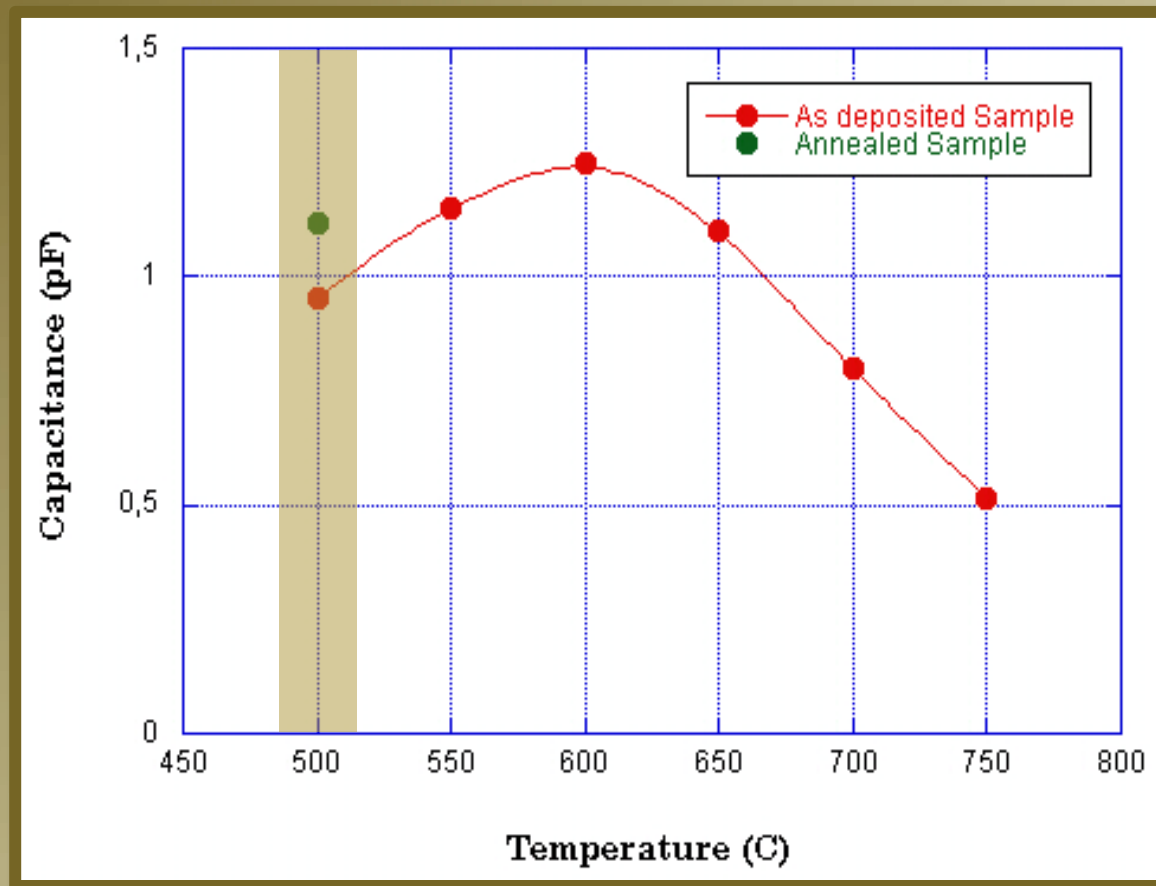


$T_g = 500^{\circ}\text{C}$  & Annealed at  $700^{\circ}\text{C}$



# Dielectric Response

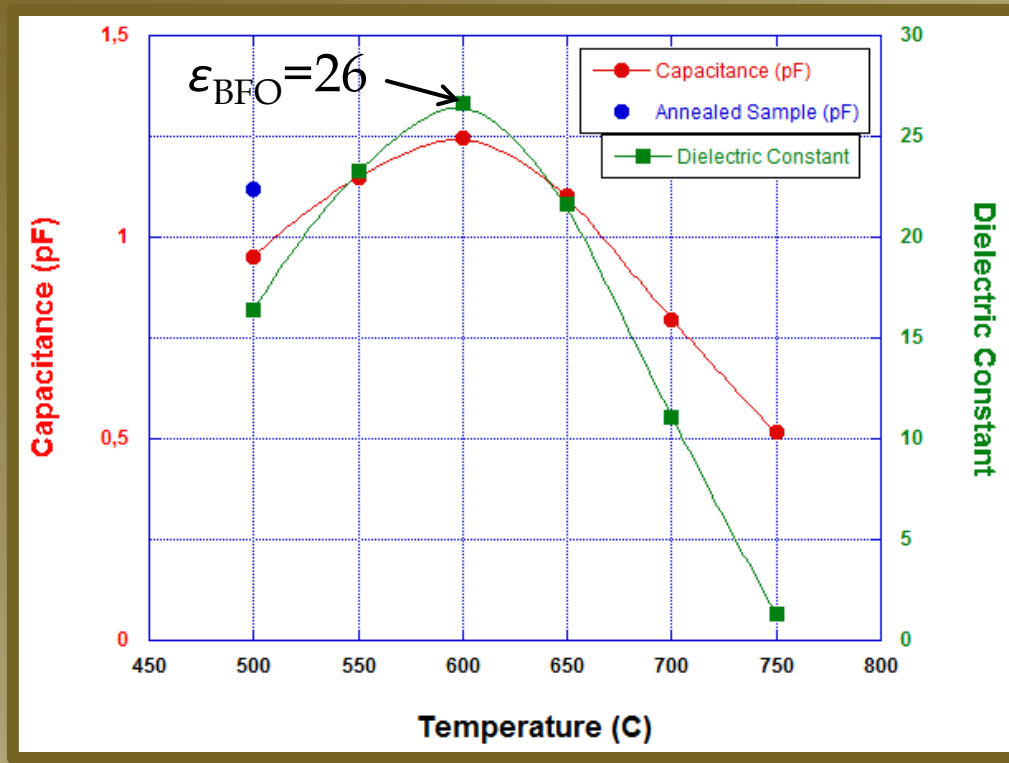
Ex – situ annealed (700°C) BFO thin film



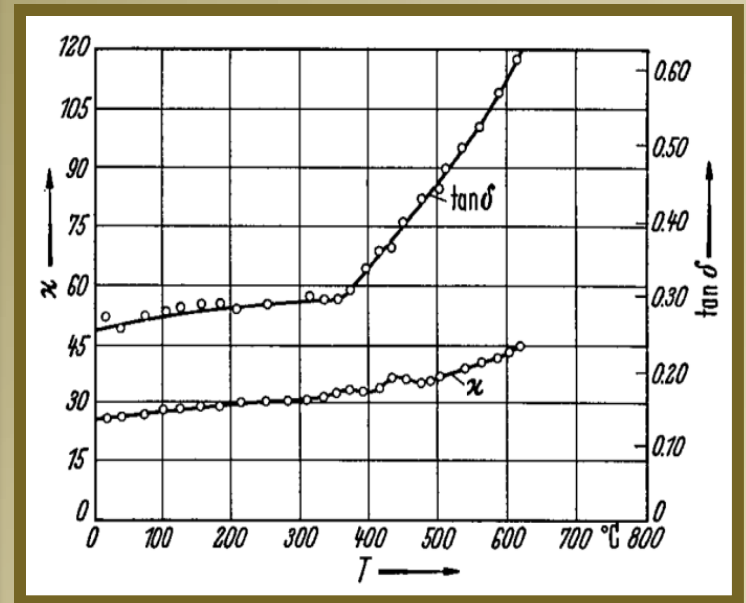


# Dielectric Response

Dielectric constant (Farnell's model)



Bulk BiFeO<sub>3</sub> ceramic

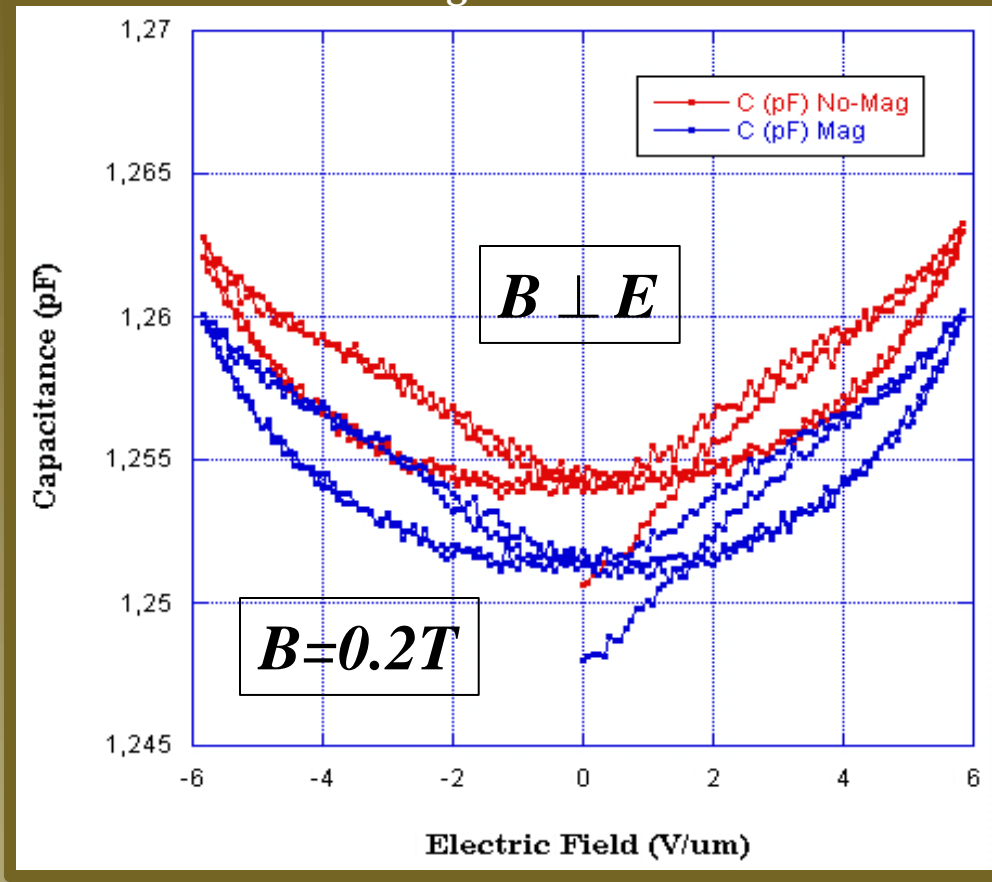


Yu.E. Roginskaya, et al. Sov.

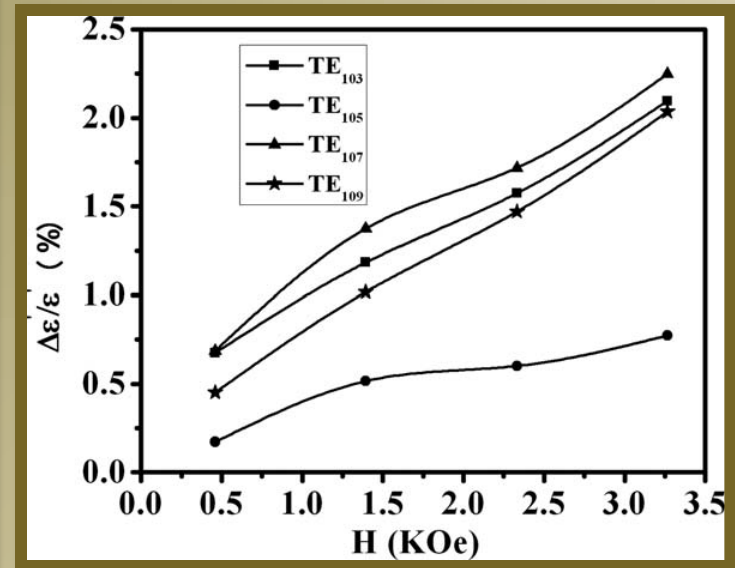
Phys. JETP, 23 (1966) 47

# Magnetoelectric response

$$T_g = 600^\circ\text{C}$$



BiFeO<sub>3</sub>/quartz substrate



F. B. A. Ahad, *J. Appl. Phys.* 105, 07D912 (2009)

$$\text{ME tunability} = \Delta\epsilon/\epsilon = 0.21\%$$

# Outline

- ▣ Background
- ▣ Objective
- ▣ Design & modeling of dielectric response
- ▣ Device fabrication
- ▣ Characterization & Analysis
- ▣ Conclusion

# Conclusions

- ▣ Effect of  $T_g$  on BFO films growth
  - Strong dependence on film's microstructure and stoichiometry
- ▣ BFO films grown over Au IDC electrodes
  - Improved microstructure/large in-plane grains
  - Reduced parasitics/less grain boundaries
- ▣ Permittivity (26),  $E_c$  (80kV/cm) and ME tunability of 0.21 % by 0.2 T @  $T_g=600^\circ\text{C}$  are close to bulk BFO

# Conclusions

- ▣ Post deposition ex-situ annealing results higher permittivity than as-deposited films
  - Improved microstructure/Increased grain size
  - Simultaneously preserving stoichiometry

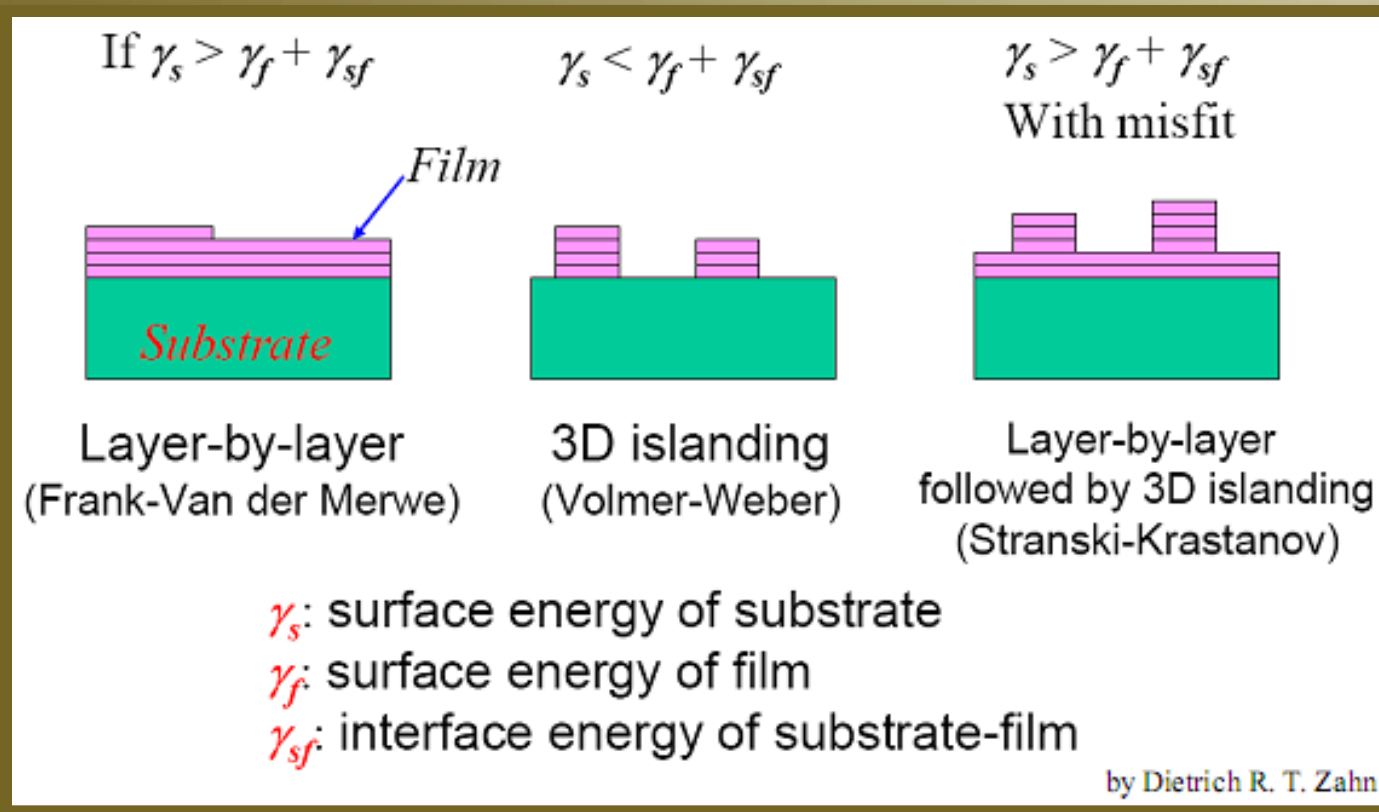
# Journal Publications

- ▣ **Taimur Ahmed**, A. Vorobiev, S. Gevorgian, “*Growth temperature dependent dielectric properties of BiFeO<sub>3</sub> thin films deposited on silica glass substrates*” Thin Solid Films, 520 (13) pp. 4470-4474 (2012).
- ▣ A. Vorobiev, **Taimur Ahmed**, S. Gevorgian, “*Microwave response of BiFeO<sub>3</sub> films in parallel-plate capacitors*” Integrated Ferroelectrics, 134 pp. 111-117 (2010).

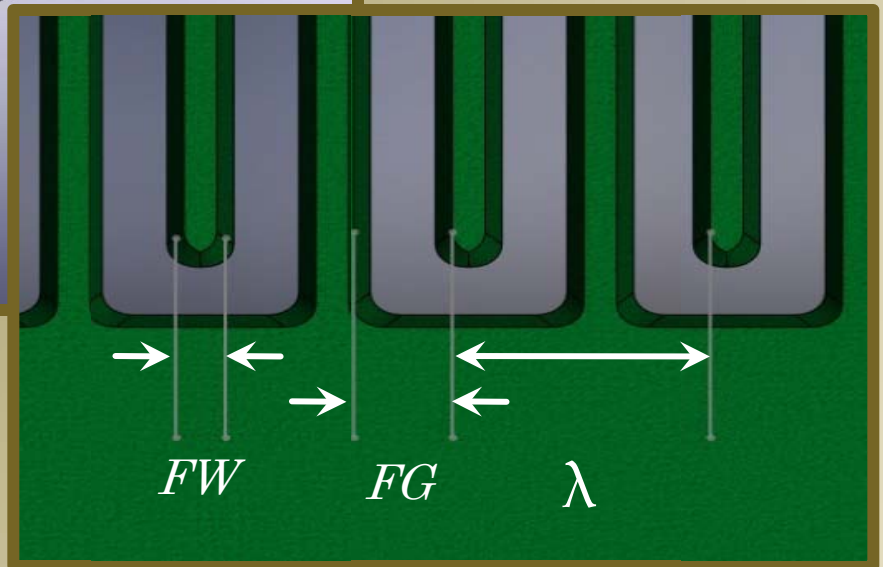
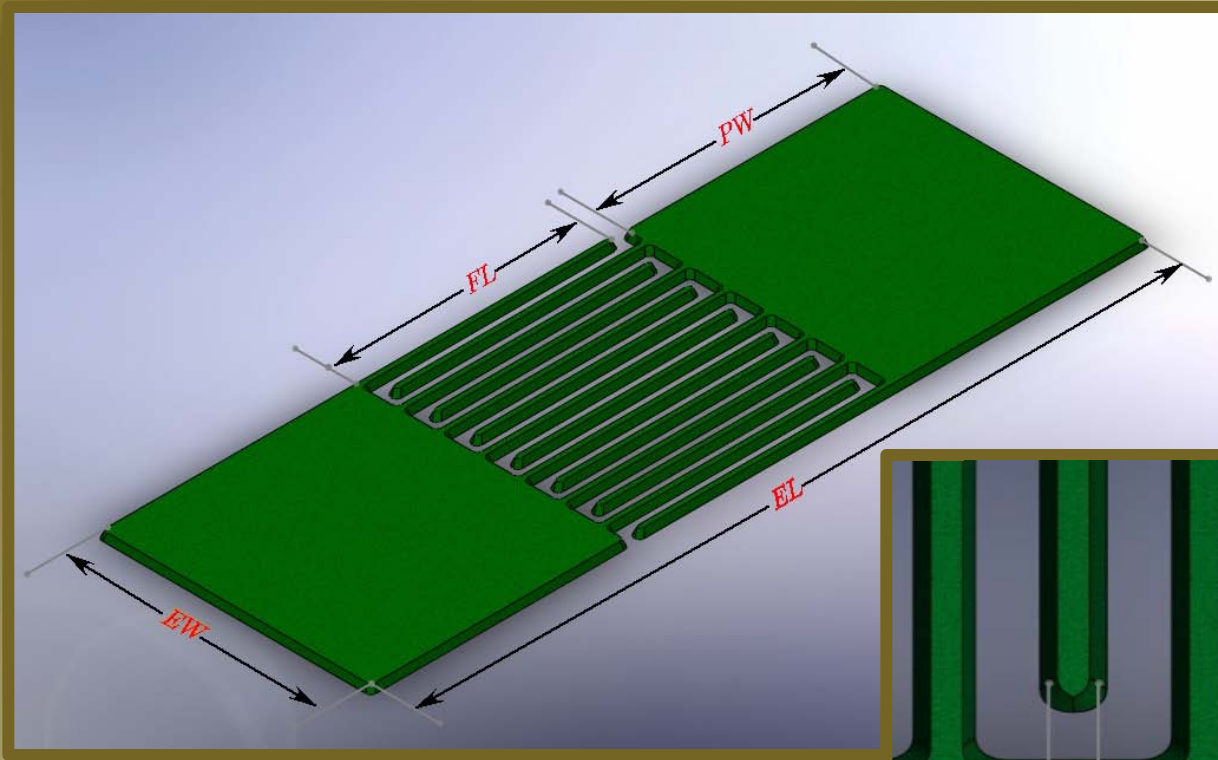




# PLD: Film nucleation and growth



# Permittivity model



# Permittivity model

## ▣ Farnell's model

$$\epsilon_{BFO} = \frac{(C - K \epsilon_s)}{K + \left\{ 0.29 \left( \frac{h}{L} \right)^{-1.5} - 1 \right\}}$$

$$K = 6.5 \left( \frac{FW}{L} \right)^2 + 1.08 \left( \frac{FW}{L} \right) + 2.37$$

C = Measured capacitance

$\epsilon_s$  = Dielectric constant of substrate (SiO<sub>2</sub>)

L = (FW + FG)

Parameters		Dimensions ( $\mu\text{m}$ )
FL	Finger length	700
FW	Finger width	3
FG	Finger gap	6
h	Finger/electrode thickness	0.55
P	Number of finger pairs	25