Aging in Fe-doped $(1-x)(Bi_{1/2}Na_{1/2})TiO_3$-$xBaTiO_3$

TECHNISCHE UNIVERSITÄT DARMSTADT

SFB 595 –Project D1

International Symposium on Electrical Fatigue in Functional Materials

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Motivation

Lead-free piezoceramics
Environmentally friendly

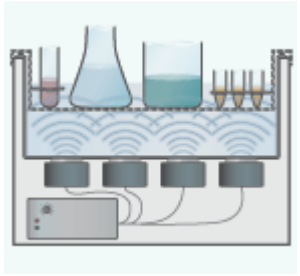
$E_{electrical} \leftrightarrow E_{mechanical}$

Actuators
Sensors

$(1\text{-}x)(Bi_{1/2}Na_{1/2})TiO_3\text{-}xBaTiO_3$

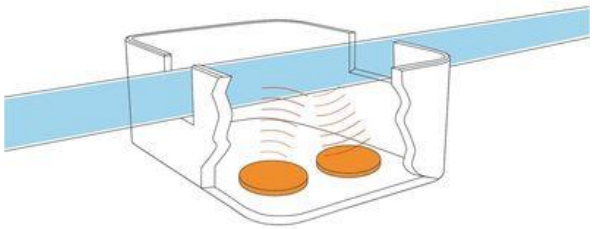
(BNT-100*x*BT)
Relaxor
Doping similar to PZT?

Ultrasonics



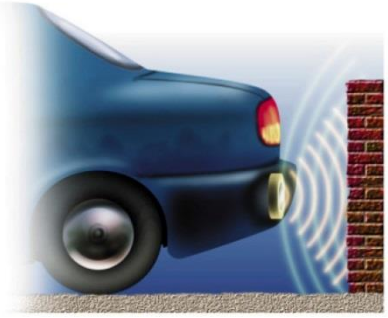
Jtt-ultraschall

Flow Meter



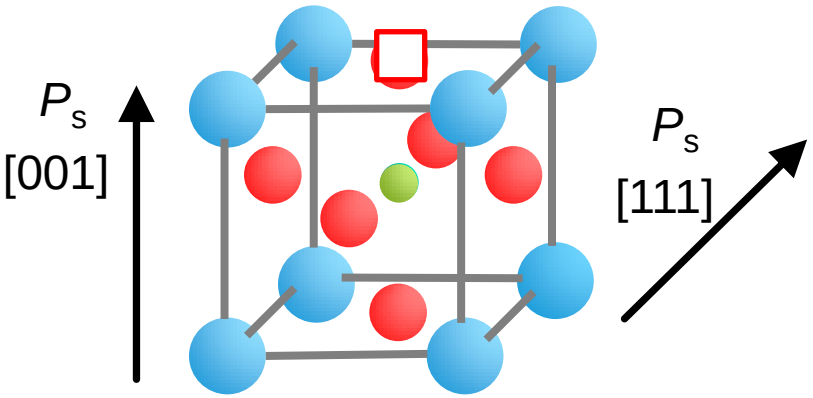
PIceramic

Distance control



Ceramtec

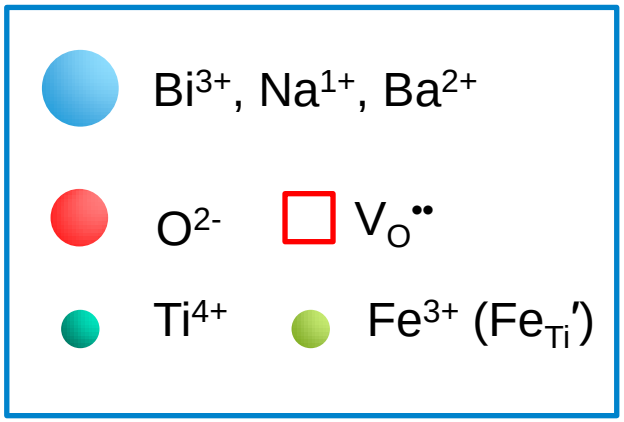
Fe-Doping in BNT-100*x*BT



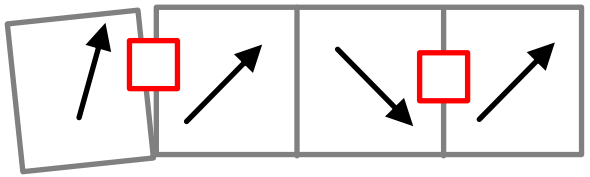
$$Fe_2O_3 \xrightarrow{TiO_2} 2Fe'_{Ti} + 3O_O^X + V_O^{\bullet\bullet}$$

$$[Fe'_{Ti}] \approx 2[V_O^{\bullet\bullet}]$$

Location of oxygen vacancies
→ *Aging model*

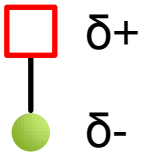


- Grain boundaries
- Domain walls

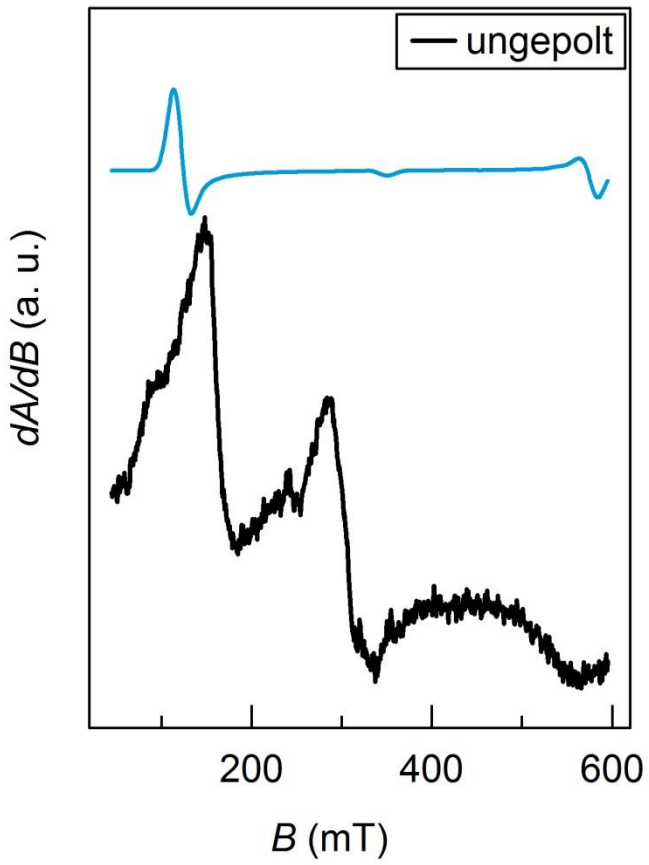


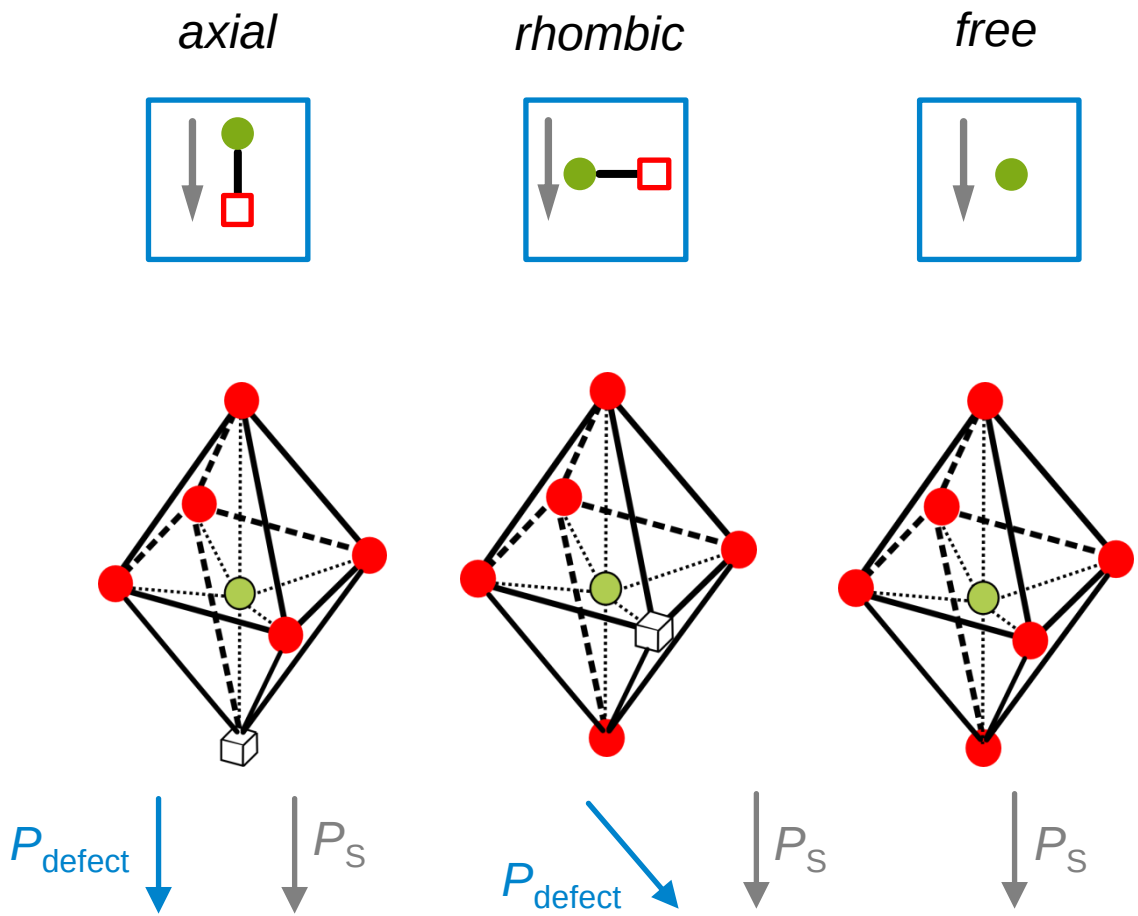
- Volume

Defect complex
($V_O^{\bullet\bullet}$ – Fe_{Ti}')$^{\bullet}$



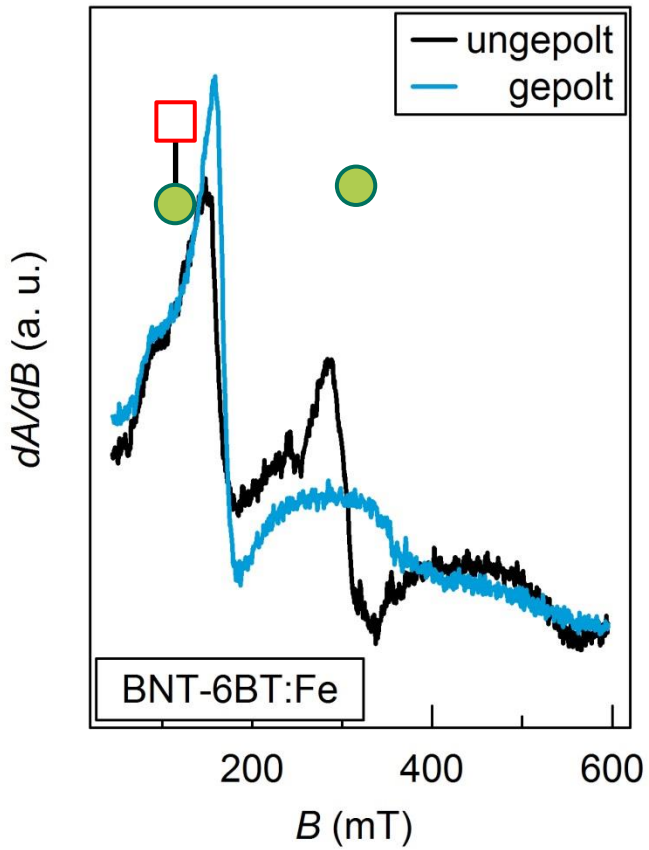
EPR Spectrum of BNT-6BT:Fe



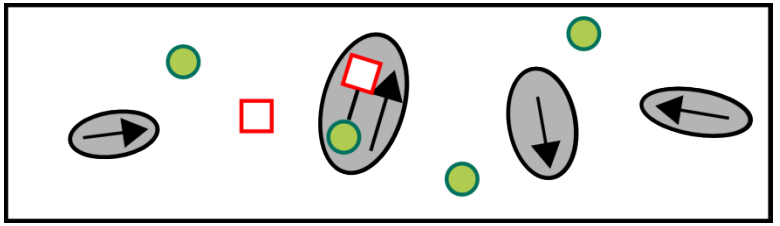


Orientations: Erhart *et al.,* Phys. Rev. B 88, 024107 (2013)
EPR: Sapper *et al., J. Appl. Phys.* **116** 104102 (2014)

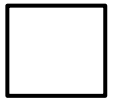
EPR Spectrum– Relaxor vs. Ferroelectric

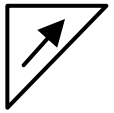


unpoled – Relaxor




PNR


Non-polar matrix

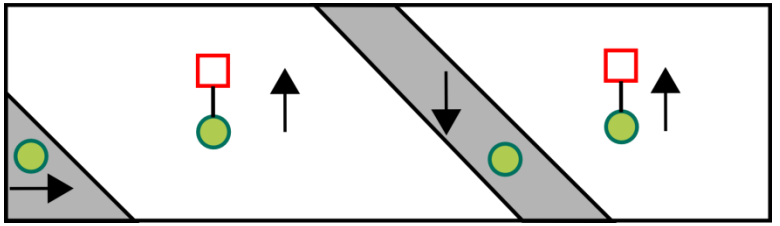

FE Domain


Fe_{Ti}'


$V_O^{\bullet\bullet}$


$(Fe_{Ti}' - V_O^{\bullet\bullet})^{\bullet}$
Defect complex

poled – Ferroelectric



EPR: Sapper *et al., J. Appl. Phys.* **116** 104102 (2014)

Aging Phenomena

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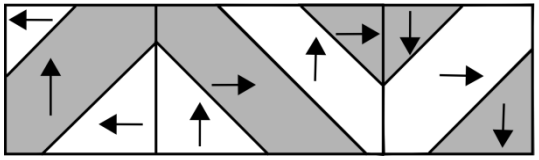
Spontaneous Polarization

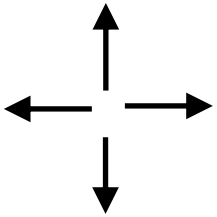
Defect complex Polarization

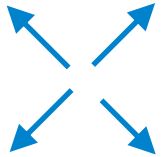
P

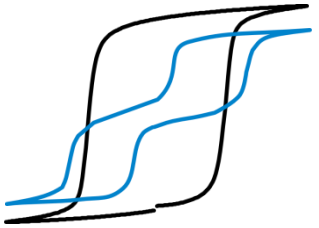
E

Ferroelectric, unpoled

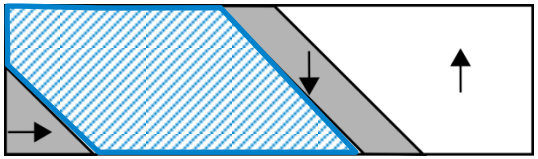


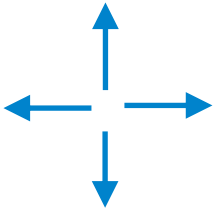




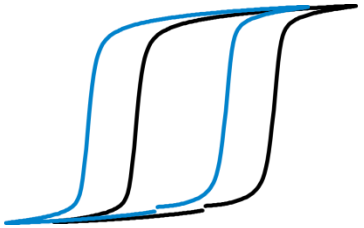


Ferroelectric, poled

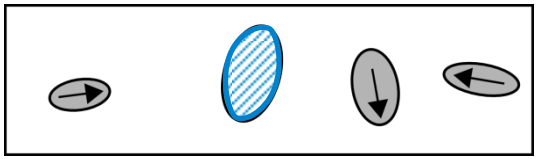


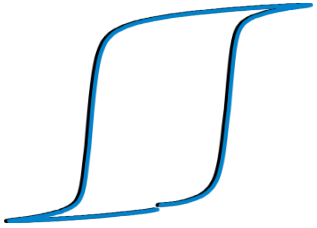


$$E_{\text{bias}} = -\frac{E_C^+ + E_C^-}{2}$$



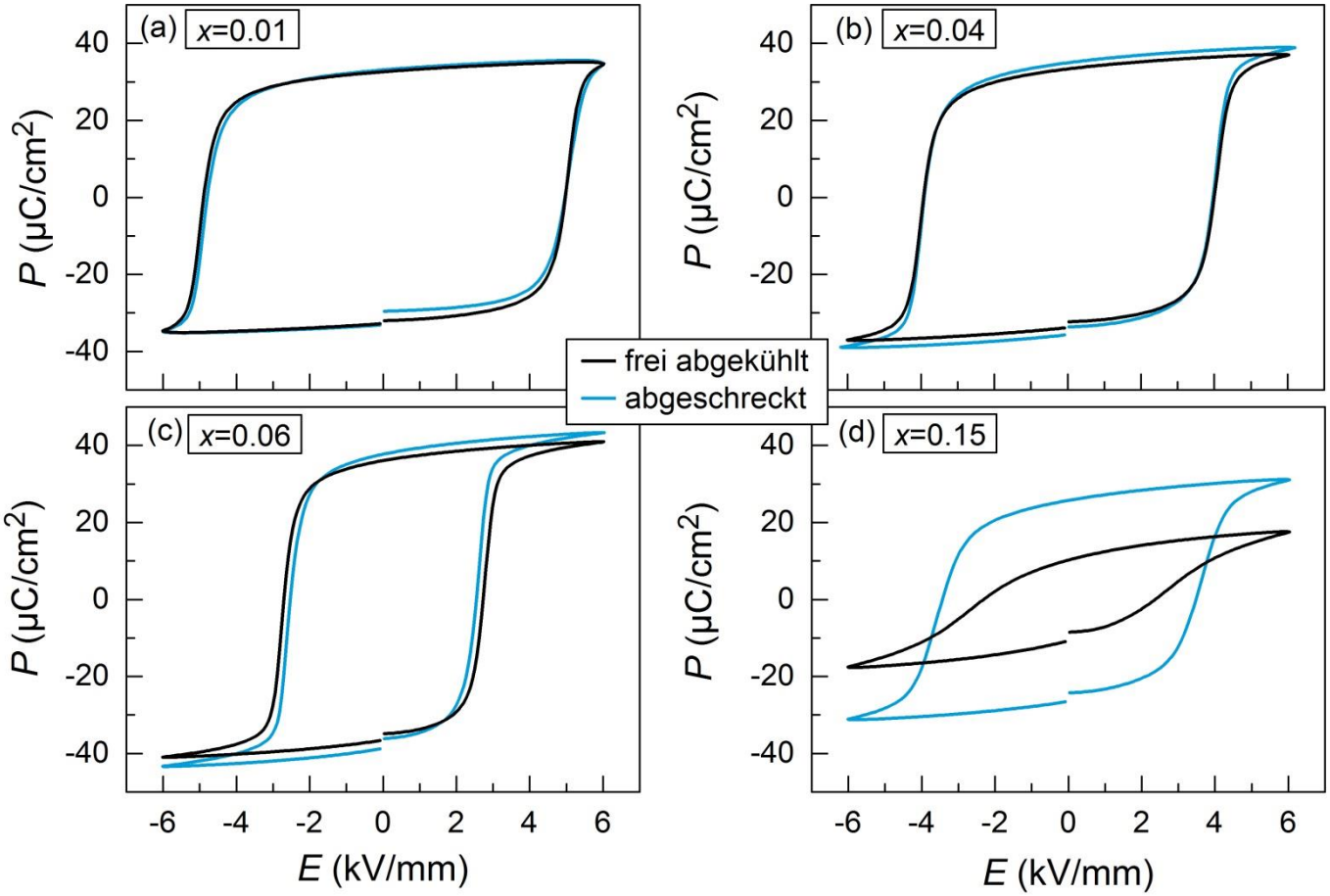
Relaxor

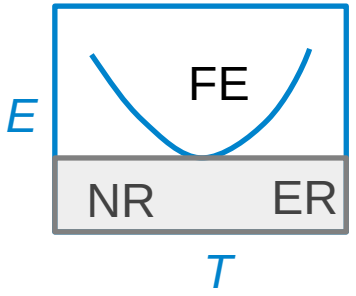




Aging in FE: Carl u. Härdtl, *Ferroelectrics*, 473-486, (1978)

Aging in BNT-100*x*BT:Fe (unpoled)





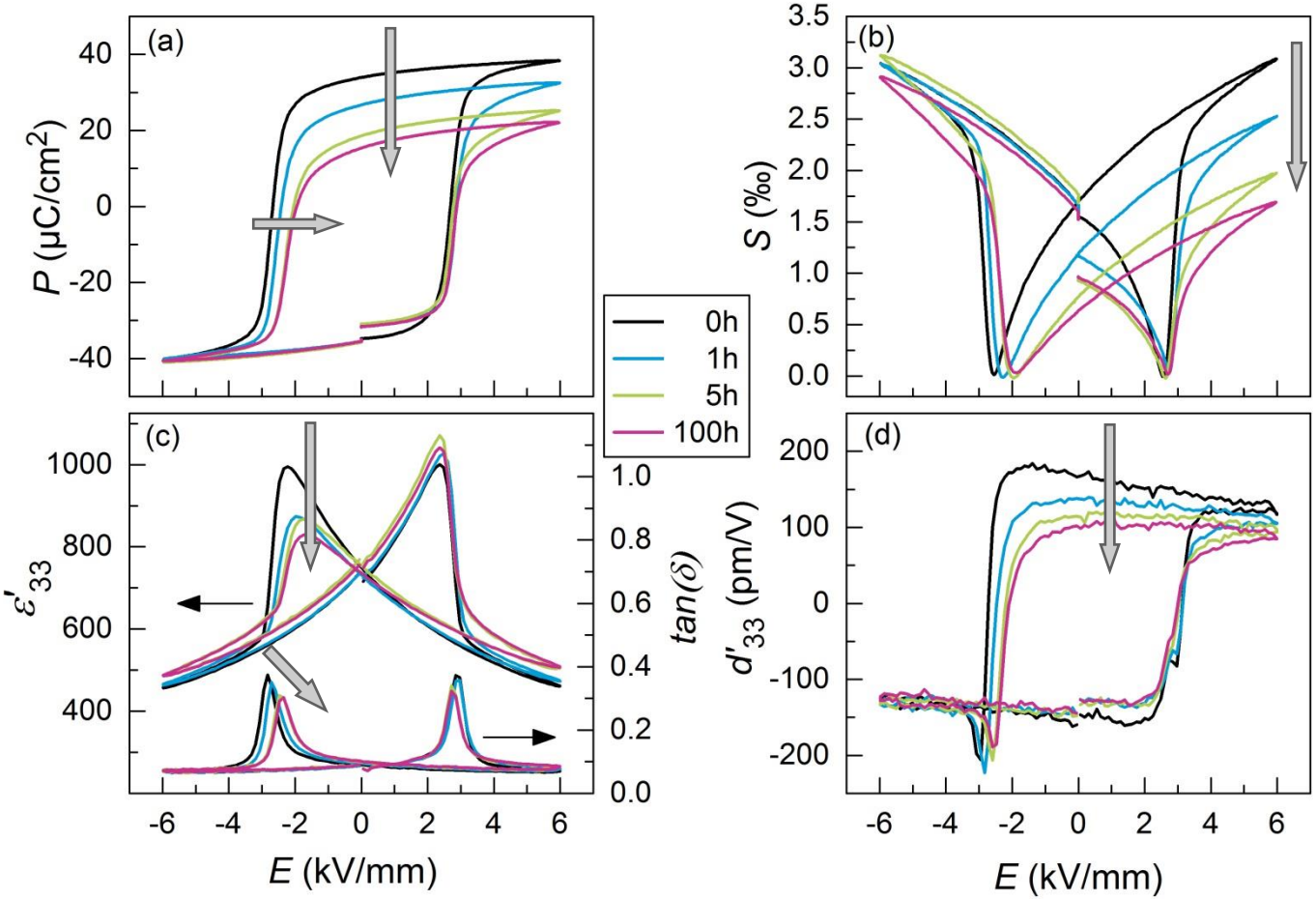
Measurement:
f =100 mHz
T= 25°C

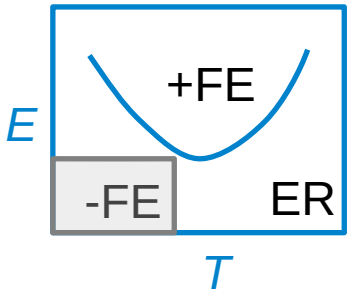
Annealing:
450°C

Quenching:
450°C → -196°C

Sapper *et al., J. Appl. Phys.* **116** 104102 (2014)

Aging in BNT-6BT:Fe (poled)



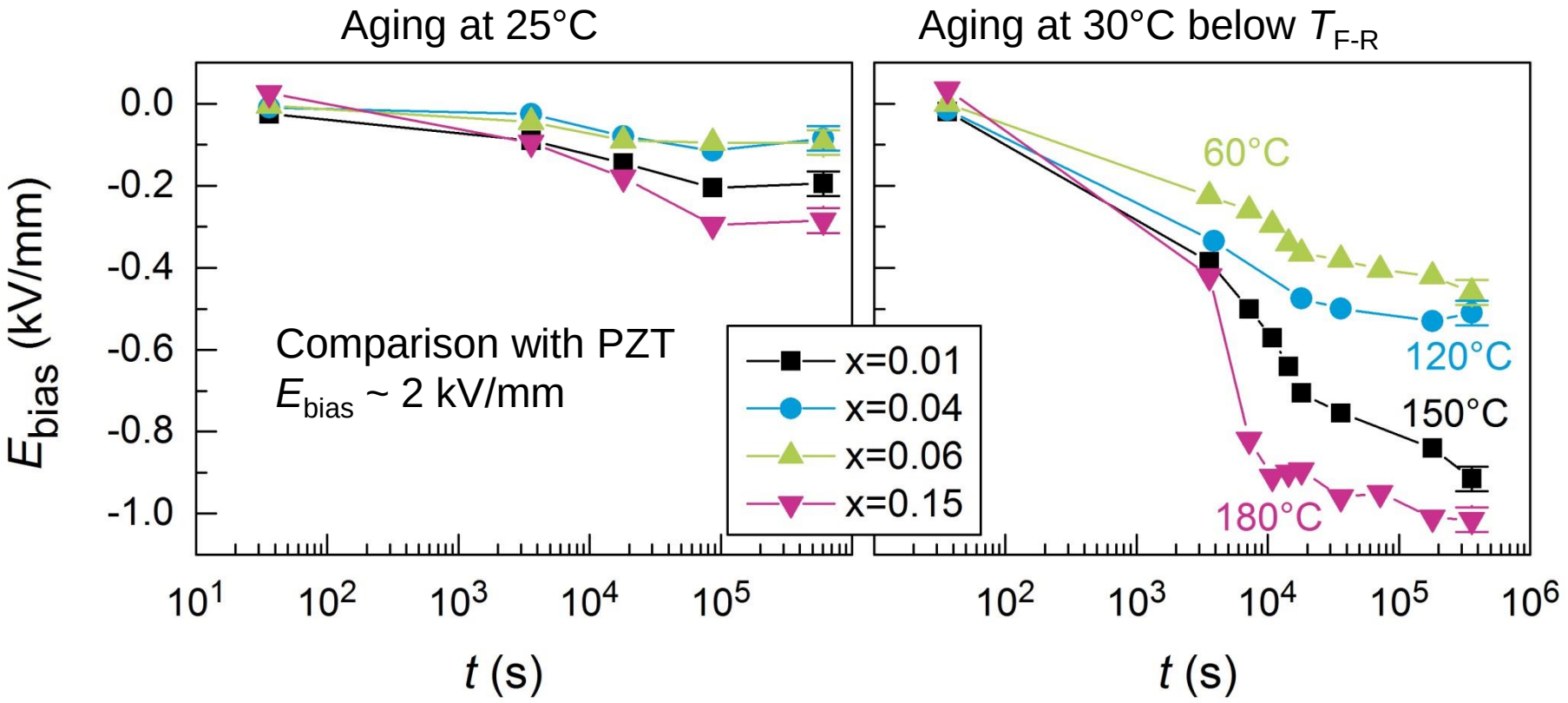


Measurement:
f_{GS}=100 mHz
f_{KS}=1 kHz
A_{KS}=10V/mm
T=25°C

Aging:
$-P_r$
T=60°C

Sapper *et al., J. Appl. Phys.* **116** 104102 (2014)

Internal Bias Field in BNT-100*x*BT:Fe



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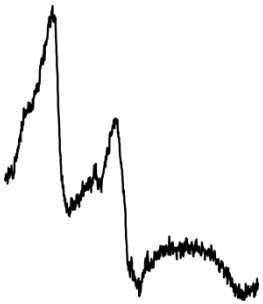
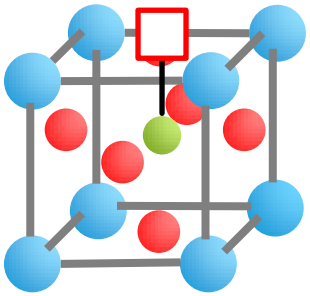
Summary

Acceptor Doping

$(1-x)(Bi_{1/2}Na_{1/2})(Ti,Fe)O_3$-$xBa(Ti,Fe)O_3$





EPR

→ defect complexes
+ free iron
→ ratio: poling conditions

Relaxor → ~~*pinching*~~

FE → $E_{bias}=f(t,T,x)$

- Hardening effect in unpoled BNT-100xBT with FE structure
- No hard-doping effect in unpoled BNT-100xBT with relaxor structure
- Aging occurs in poled samples in all compositions

→ Fe-doping can be utilized to tailor BNT-100xBT materials

Acknowledgments

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