

The Impact of Relaxor Properties on Aging and Fatigue in Lead- Free (1-x)BNT-xBT

Project D1



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Motivation

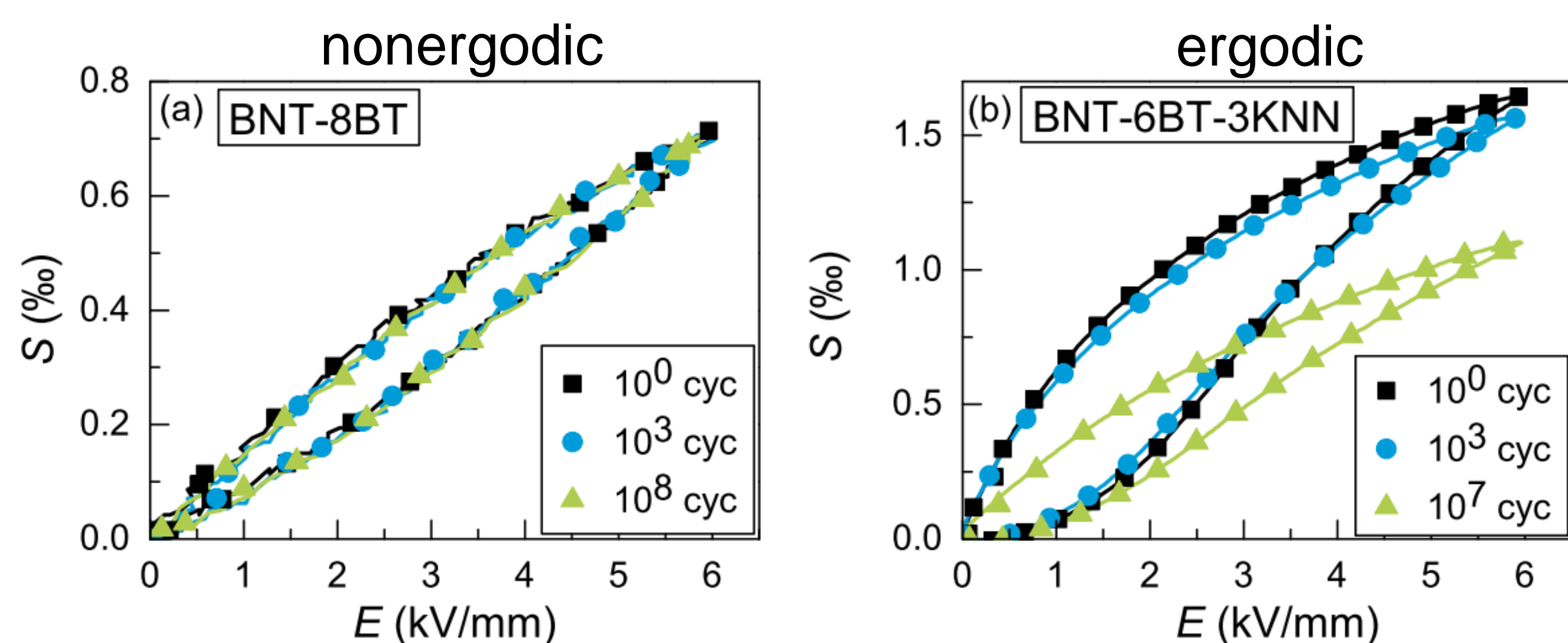
- Development of lead-free piezoceramics for actuator- and sensor-applications
- Identification and quantification of fatigue and aging mechanisms in lead-free piezoceramics
- Influence of the relaxor state on fatigue and aging
- Influence of doping: Possibility of tailoring the material?

→ *E-T* stability diagram

→ Acceptor-doping: aging and hardening effects

→ Fatigue characteristics in lead-free multilayer devices

Fatigue in Lead-Free Multilayer Actuators



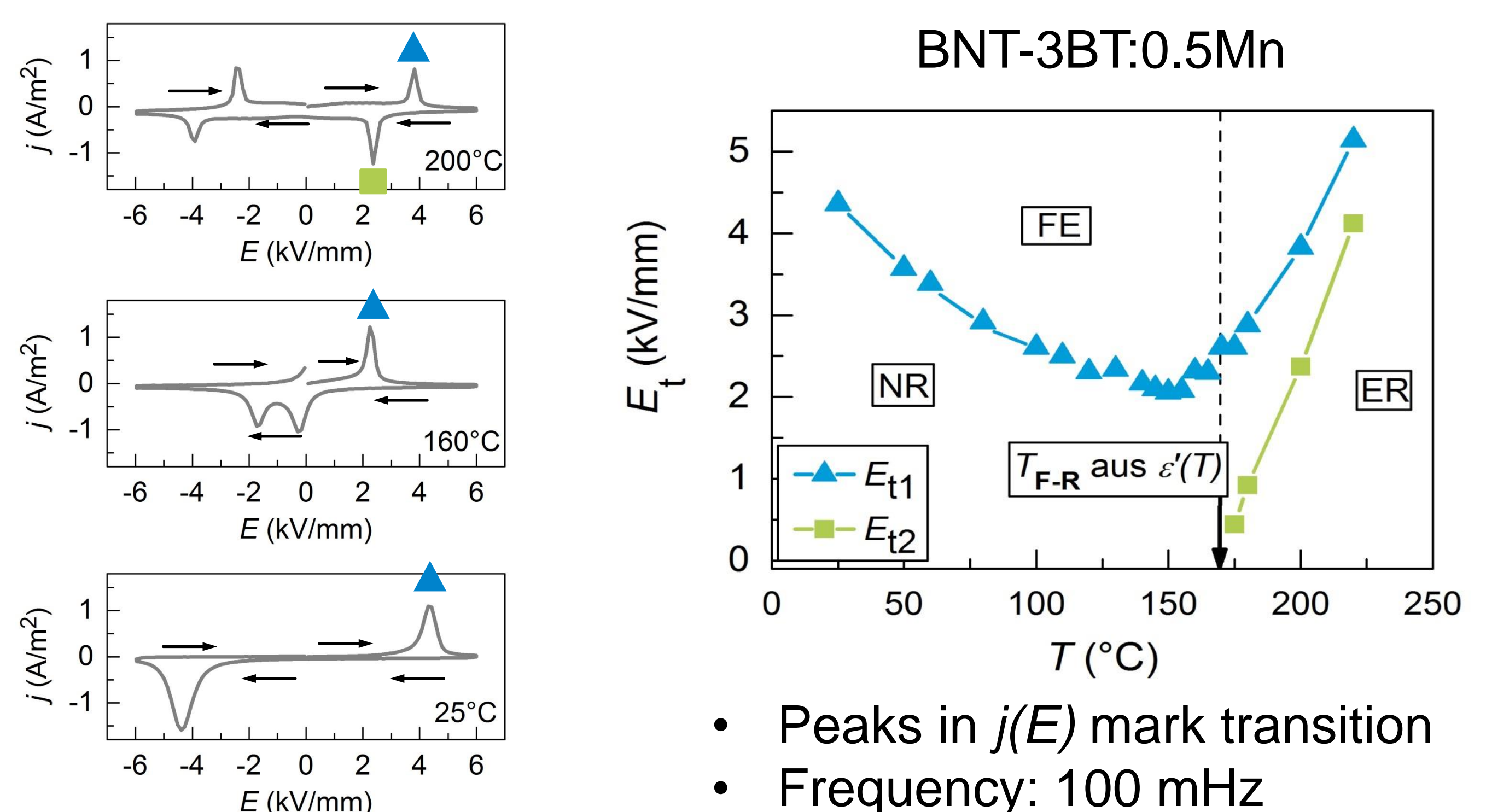
- Successful implementation of lead-free ceramics in multilayer devices
- Unipolar cycling at ambient temperature up to 10^8 cycles
- Nonergodic relaxor: low strain, high fatigue resistance
- Ergodic relaxor: high strain, low fatigue resistance

Publications last funding period

- 1) Sapper et al., *Aging in the relaxor and ferroelectric state of Fe-doped (1-x)(Bi_{1/2}Na_{1/2})TiO₃-xBaTiO₃ piezoelectric ceramics*, J. Appl. Phys., accepted (2014)
- 2) Sapper et al., *Electric-field-temperature phase diagram of the ferroelectric relaxor system (1-x)Bi_{1/2}Na_{1/2}TiO₃-xBaTiO₃ doped with manganese*, J. Appl. Phys., **115** (19) 194104 (2014)
- 3) Sapper et al., *Cycling stability of lead-free BNT-8BT and BNT-6BT-3KNN multilayer actuators and bulk ceramics*, J. Eur. Ceram. Soc., **34** (3) 653-661 (2014)
- 4) Glaum et al., *De-aging of Fe-doped lead-zirconate-titanate ceramics by electric field cycling: 180°- vs. non-180° domain wall processes*, J. Appl. Phys., **112** (3) 034103 (2012)
- 5) Jo et al., *Giant electric-field-induced strains in lead-free ceramics for actuator applications - status and perspective*, J. Electroceram., **29** 71-93 (2012)
- 6) Sapper et al., *Influence of electric fields on the depolarization temperature of Mn-doped (1-x)Bi_{1/2}Na_{1/2}TiO₃-xBaTiO₃*, J. Appl. Phys., **111** (1) 014105 (2012)
- 7) Luo et al., *Effect of Ferroelectric Long-Range Order on the Unipolar and Bipolar Electric Fatigue in Bi_{1/2}Na_{1/2}TiO₃-Based Lead-Free Piezoceramics*, J. Am. Ceram. Soc., **94** 3927-3933 (2011)
- 8) Glaum et al., *Temperature and driving field dependence of fatigue processes in PZT bulk ceramics*, Acta Mat., **59** 6083-6092 (2011)
- 9) Drahus et al., *Manganese-doped (1-x)BiScO₃-xPbTiO₃ high-temperature ferroelectrics: Defect structure and mechanism of enhanced electric resistivity*, Phys. Rev. B, **84** 064113 (2011)
- 10) Ehmke et al., *Stabilization of the Fatigue-Resistant Phase by CuO Addition in (Bi_{1/2}Na_{1/2})TiO₃-BaTiO₃*, J. Am. Ceram. Soc., **94** 2473 (2011)
- 11) Luo et al., *Bipolar and Unipolar Fatigue of Ferroelectric BNT-Based Lead-Free Piezoceramics*, J. Am. Ceram. Soc., **94** 529 (2011)

E-T Phase Diagram

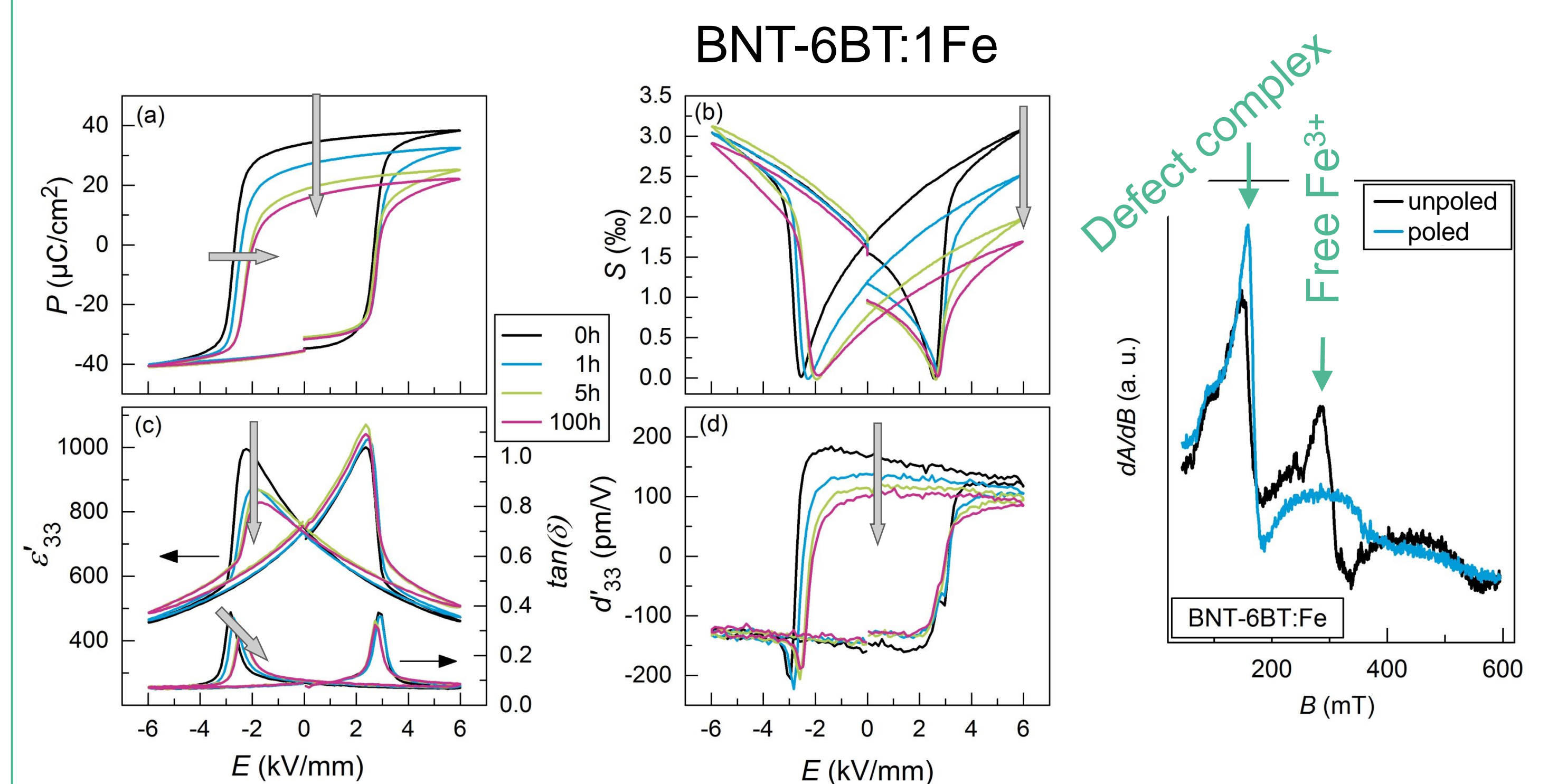
- Canonical relaxor: ergodic state, nonergodic state, field-induced ferroelectric state
- Stability regions depend on electric field and temperature
- Field-induced transitions visible in $\epsilon'(E)$ and $j(E)$



- Peaks in $j(E)$ mark transition
- Frequency: 100 mHz

Aging in Fe-doped Samples

- *Acceptor-doping*: hardening absent, aging absent in relaxor state
- Field-induced ferroelectric state: internal bias field develops
- EPR shows co-existence of defect complexes and free iron
- Ratio of defect complexes and free iron depends on poling state



5 Key Publications (2003-2014)

- 1) Balke et al., *Bipolar Fatigue Caused by Field Screening in Pb(Zr,Ti)O₃ Ceramics*, J. Am. Ceram. Soc., **90** 3869-3874 (2007)
- 2) Genenko et al., *Aging of poled ferroelectric ceramics due to relaxation of random depolarization fields by space-charge accumulation near grain boundaries*, Phys. Rev. B, **80** 224109 (2009)
- 3) Luo et al., *Effect of Ferroelectric Long-Range Order on the Unipolar and Bipolar Electric Fatigue in Bi_{1/2}Na_{1/2}TiO₃-Based Lead-Free Piezoceramics*, J. Am. Ceram. Soc., **94** 3927-3933 (2011)
- 4) Glaum et al., *Temperature and driving field dependence of fatigue processes in PZT bulk ceramics*, Acta Mat., **59** 6083-6092 (2011)
- 5) Sapper et al., *Electric-field-temperature phase diagram of the ferroelectric relaxor system (1-x)Bi_{1/2}Na_{1/2}TiO₃-xBaTiO₃ doped with manganese*, J. Appl. Phys., **115** (19) 194104 (2014)