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Universality of polarization response in virgin and fatigued ferroelectrics

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Project B7

Motivation

Polarization switching dynamics in virgin and fatigued ferroelectrics
(PZT, lead-free ferroelectric ceramics: BNT-BT, BZT-BCT and single crystals)

- switching characteristics for time interval from 10^{-6}s to 10^{3}s and temperature range from -50^0C to $+200^0$C: τ - switching time, E_a - activation field, switched and back-switched polarization versus time and applied field
- influence of bipolar fatigue on switching parameters (defects, pinning centers, crack formation, dead-layer), fatigue regime: 50Hz, $2E_c$, cycles number up to 10^8
- new models for polarization reversal (Inhomogeneous Field Mechanism – IFM model (2010)): distribution of switching times $g(\tau)$, local field distribution $f(E/E_m)$

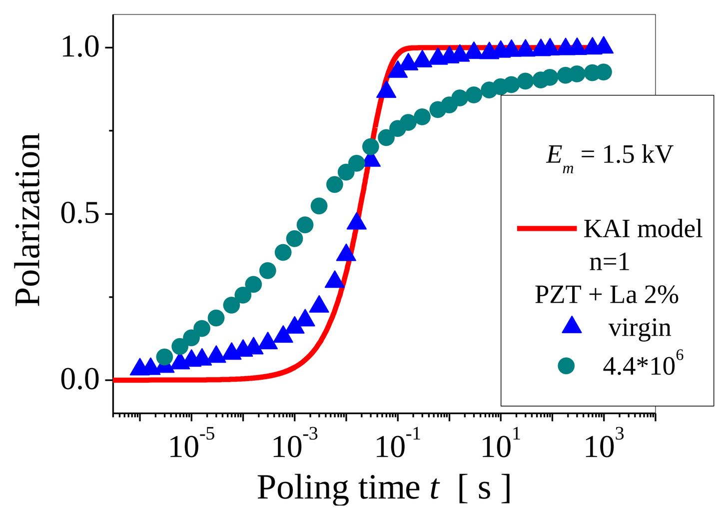
Models with single τ

Classical model of nucleation and growth: Kolmogorov-Avrami-Ishibashi (KAI)

$$\Delta P(t) = 2P_s\left\{1-\exp\left[-\left(\frac{t}{\tau}\right)^n\right]\right\}$$
$$n = 3;\ 2;\ 1$$

Classical KAI model cannot properly describes the polarization reversal in fatigued ceramics

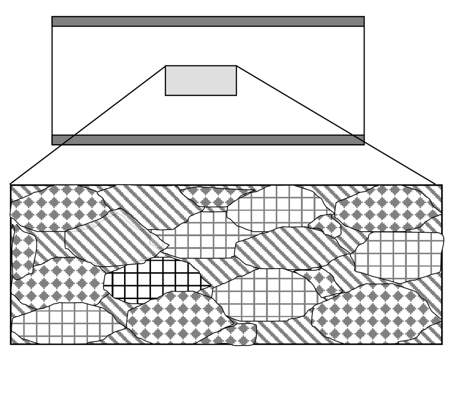
KAI model vs. experiment



Models with spectrum of τ

Switching volume is represented as an ensemble of many regions with independent dynamics

Total polarization of the system results from the summation of the local polarizations $p(t,\tau)$ and can be represented as



$$\Delta P(E_m,t) = \int_0^\infty d\tau\, g(\tau)\, p(t,\tau)$$

where $g(\tau)$ is the distribution of the switching times in the system

How to obtain g(τ) from experiment ?

- Nucleation-limited-switching (NLS) model by Tagantsev et al., 2002
- Lorentzian distribution for g(log τ) as a result of the local field distribution due to pinning sites (Jo et al. 2007, Nautial et al. 2010, Dabra 2010)

Results

Inhomogeneous Field Mechanism Model (IFM -Model) for polarization reversal

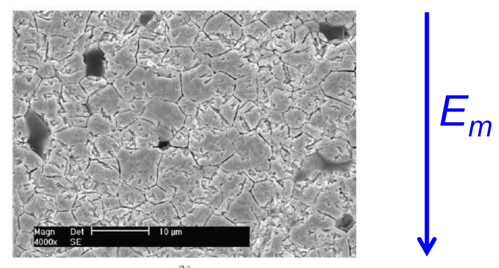
$$\Delta P(E_m,t) \cong \Delta P_{max} \int_0^{E_m/E_{max}(t)} \Phi(u)\frac{du}{u}$$

$$\Phi(E_m/E_{max}) = \frac{1}{\Delta P_{max}}\frac{\partial \Delta P}{\partial(\ln E_m)}$$

$$E_{max}(t) \Leftrightarrow \tau(E_m)$$

E_m – applied field, ΔP – switched polarization

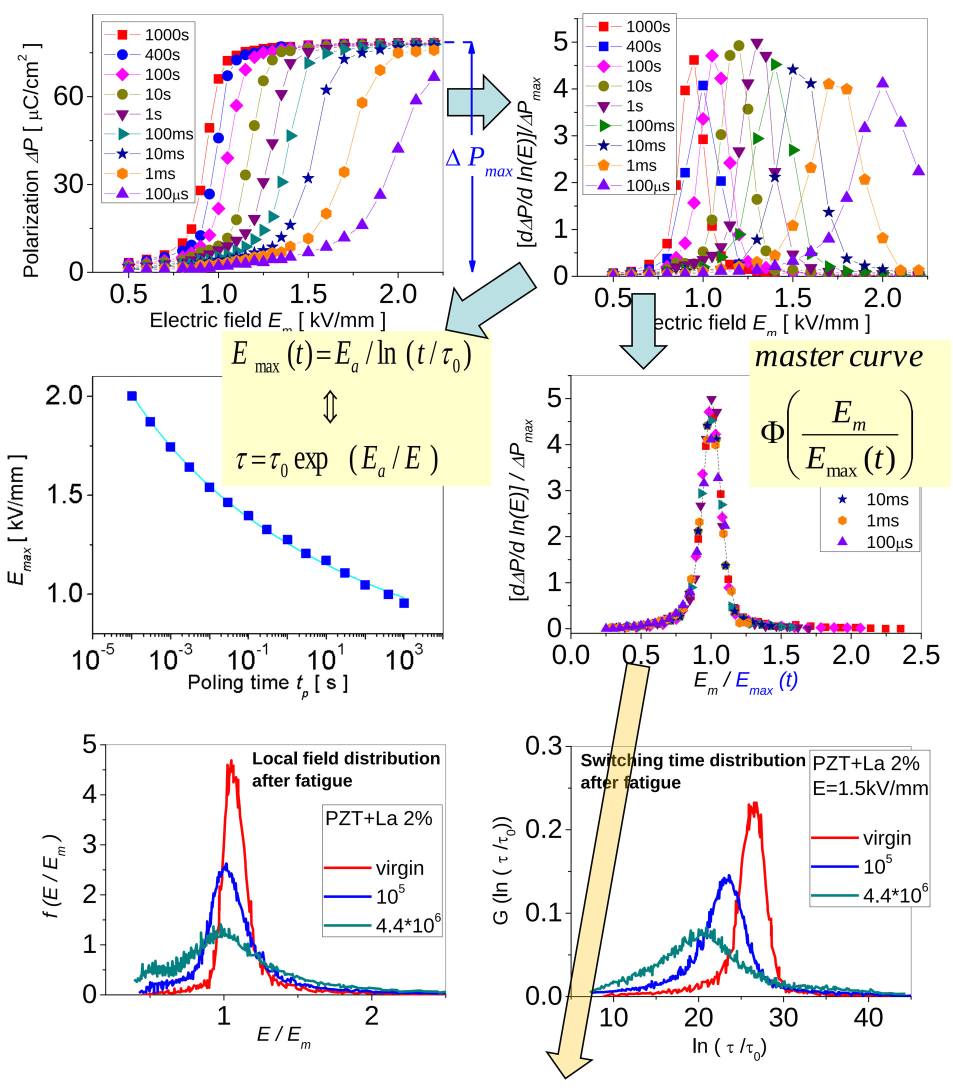
Local field distribution $Z(E/E_m)$ in polycrystalline ceramic



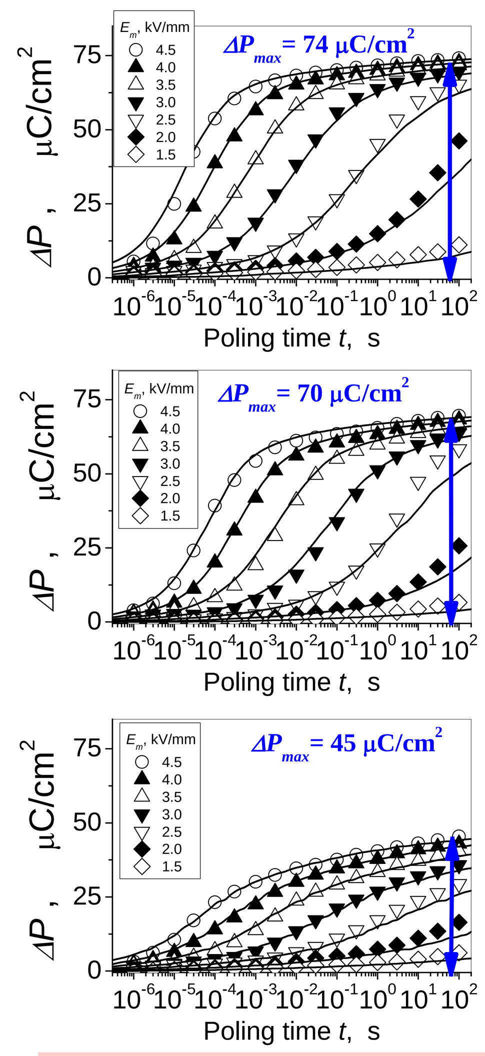
Factors determined the local field distribution in polycrystalline ceramic at the mesoscopic scale

- ✓Grain boundaries and triple points
- ✓Pores and cracks
- ✓Polar defects and pinning sites
- ✓Spatial variation in dielectric permittivity tensor

Scaling properties of the normalized logarithmic derivative $\{\partial P/\partial(\ln E)\}$ / ΔP_{max} - virgin and fatigued samples of $Pb(Zr_{0,525}Ti_{0,475})O_3$ + 2% La



Polarization kinetics in Lead-free ceramic in virgin and fatigued states: BNT-6BT+Cu



Fatigue resistance comparable to some commercial PZT ceramics

Scaling properties in different classes of well disordered ferroelectrics

PZT polycrystalline bulk ceramics (virgin and fatigued)

Lead-free ceramics BNT-6BT + Cu; BZT-BCT (virgin and fatigued)

Organic ferroelectrics: PVDF, p(VDT-TrFE)

The polarization reversal is primarily controlled by the statistical characteristics of disorder (local field distribution) rather than by a temporal law of the local polarization switching.

Universal Switching Behavior

Publications last funding period

1) S. Zhukov, H. Kungl, Yu.A. Genenko and H. von Seggern, *"Statistical electric field and switching time distributions in PZT 1Nb2Sr ceramics: Crystal- and microstructure effects,"* J. Appl. Phys. 2014, 115, 014103.

2) S. Zhukov, Yu.A. Genenko, M. Acosta, H. Humburg, W. Jo, J. Rödel and H. von Seggern, *"Polarization dynamics across the morphotropic phase boundary in $Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ ferroelectrics,"* Appl. Phys. Lett. 2013, 103, 152904.

3) Y.A. Genenko, S. Zhukov, S.V. Yampolskii, J. Schütrumpf , R. Dittmer, W. Jo, H. Kungl, M.J. Hoffmann and H. von Seggern, *"Universal polarization switching behavior of disordered ferroelectrics,"* Adv. Funct. Mater. 2012, 22, 2058–2066.

4) J. Schütrumpf, S. Zhukov, Y. A. Genenko and H von Seggern, *"Polarization switching dynamics by inhomogeneous field mechanism in ferroelectric polymers,"* J. Phys. D: Appl. Phys. 2012, 45, 165301.

5) Yu.A. Genenko, J. Wehner and H. von Seggern, *"Self-consistent model of polarization switching kinetics in disordered ferroelectrics,"* J. Appl. Phys. 2013, 114, 084101.

6) Y. Zuo, Yu.A. Genenko, A. Klein, P. Stein and B. Xu, *"Domain wall stability in ferroelectrics with space charges,"* J. Appl. Phys. 2014, 115, 084110.

7) S. Li, J. Morasch, A. Klein, C. Chirila, L. Pintilie, L. Jia, K. Ellmer, M. Naderer, K. Reichmann, M. Gröting, and K. Albe, *"Influence of orbital contributions to the valence band alignment of Bi2O3, Fe2O3, BiFeO3, and Bi0.5Na0.5TiO3,"* Phys. Rev. B 2013, 88, 045428.

8) F. Chen and A. Klein, *"Polarization dependence of Schottky barrier heights at inter-faces of ferroelectrics determined by photoelectron spectroscopy,"* Phys. Rev. B 2012, 86, 094105.

9) A. Klein, *"Energy band alignment at interfaces of semiconducting oxides: A review of experimental determination using photoelectron spectroscopy and comparison with theoretical predictions by the electron affinity rule, charge neutrality levels, and the common anion rule,"* Thin Solid Films 2012, 520, pp.3721–3728.

10) R. Schafranek, S. Li, F. Chen, W. Wu, and A. Klein, *"$PbTiO_3/SrTiO_3$ interface: Energy band alignment and its relation to the limits of Fermi level variation,"* Phys. Rev. B 2011, 84, 045317.

11) F. Chen, R. Schafranek, W. Wu, and A. Klein, *"Reduction-induced Fermi level pinning at the interfaces between $Pb(Zr,Ti)O_3$ and Pt, Cu and Ag metal electrodes,"* J. Phys. D: Appl. Phys. 2011, 44, 255301.

5 Key Publications (2003-2014)

1) S. Zhukov, Yu.A. Genenko, O. Hirsch, J. Glaum, T. Granzow, and H. von Seggern, *"Dynamics of polarization reversal in virgin and fatigued ferroelectric ceramics by inhomogeneous field mechanism,"* Phys. Rev. B 2010, 82, 014109.

2) Y.A. Genenko, S. Zhukov, S.V. Yampolskii, J. Schütrumpf , R. Dittmer, W. Jo, H. Kungl, M.J. Hoffmann and H. von Seggern, *"Universal polarization switching behavior of disordered ferroelectrics,"* Adv. Funct. Mater. 2012, 22, 2058–2066.

3) S. Zhukov, H. Kungl, Yu.A. Genenko and H. von Seggern, *"Statistical electric field and switching time distributions in PZT 1Nb2Sr ceramics: Crystal- and microstructure effects,"* J. Appl. Phys. 2014, 115, 014103.

4) F. Chen and A. Klein, *"Polarization dependence of Schottky barrier heights at inter-faces of ferroelectrics determined by photoelectron spectroscopy,"* Phys. Rev. B 2012, 86, 094105.

5) R. Schafranek, S. Li, F. Chen, W. Wu, and A. Klein, *"$PbTiO_3/SrTiO_3$ interface: Energy band alignment and its relation to the limits of Fermi level variation,"* Phys. Rev. B 2011, 84, 045317.