MULTI-SCALE MATERIAL MODELLING OF GLASS-CERAMICS

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Fraunhofer IKTS in Profile
Research Fields

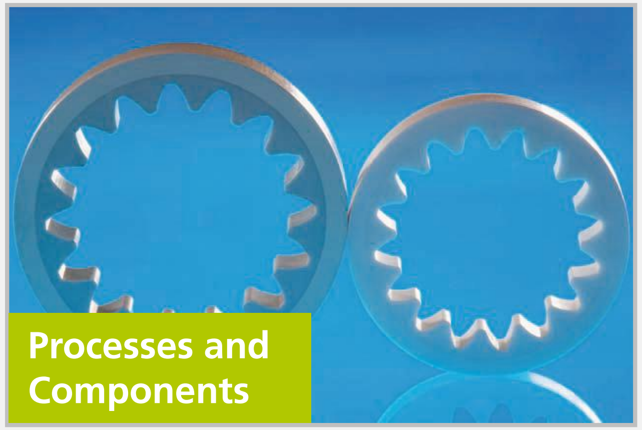
Structural ceramics

Functional ceramics

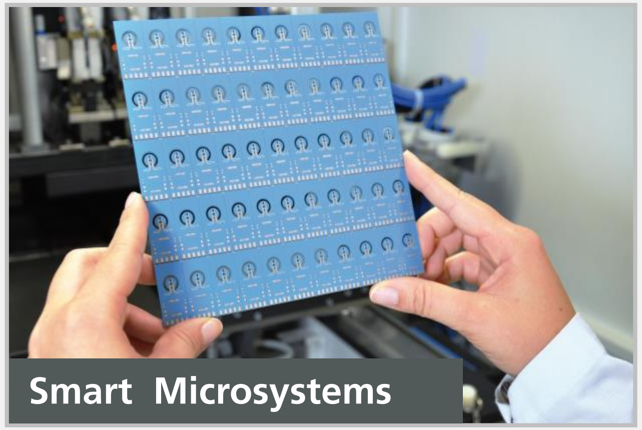

Materials


Sintering and Characterization


Energy Systems


Processes and Components


Environmental Engineering and Bioenergy


Smart Microsystems

Dresden

(1)

(2)

Dresden

(1) https://www.dresden.de/de/tourismus/sehen/sehenswuerdigkeiten/altstadt.php , Martin Dietrich

(2) http://www.lonelyplanet.com/maps/europe/germany/

CONTENT

- Motivation
- Basics
- Simulation
  - Multi-Scale Model
  - Material Description
  - Geometric Representation
- Results
- Fabrication of Glass-Ceramic
- Summary

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- **Motivation**
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Motivation

(1)

Exit from nuclear and fossil-fuel energy

Elevated demand for high voltage ceramic capacitors

Increase of dielectric medium

New material:

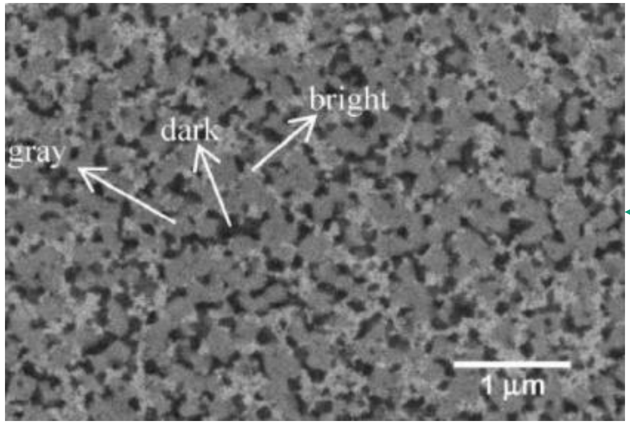
Glass-Ceramic

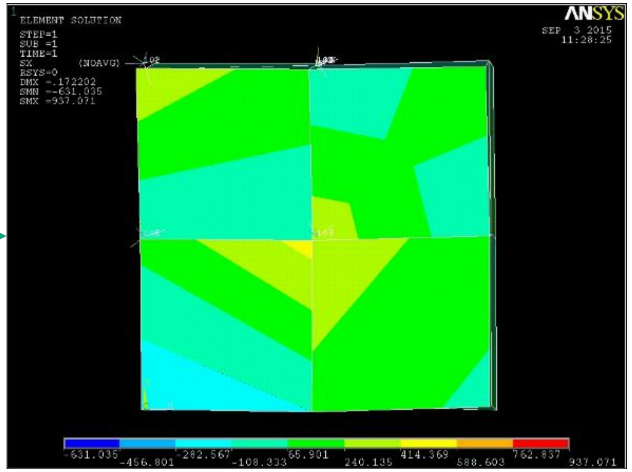
Understand microstructure-properties-correlation

Improve material properties

(2)

(3)



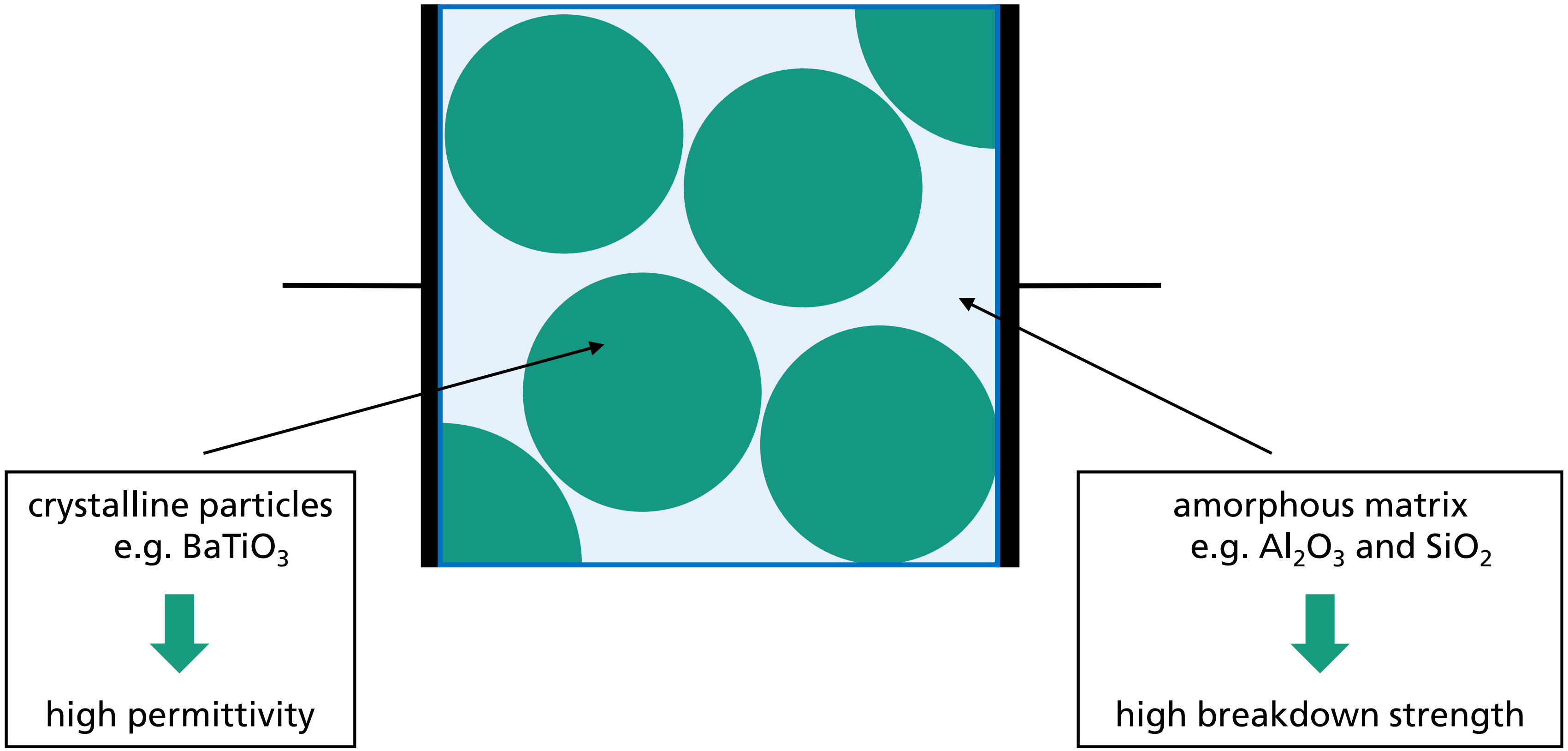


(1) http://www.fotocommunity.de/pc/pc/display/29499476 by hansen-net

(2) https://commons.wikimedia.org/wiki/File:HoheneckUmspannwerkTrafos.jpg

(3) Luo, J.; Du, J.; Tang, Q.; Mao, C. *IEEE Trans. Electron Devices* **2008,** *55* (12), 3549–3554.

Motivation: Glass-Ceramic

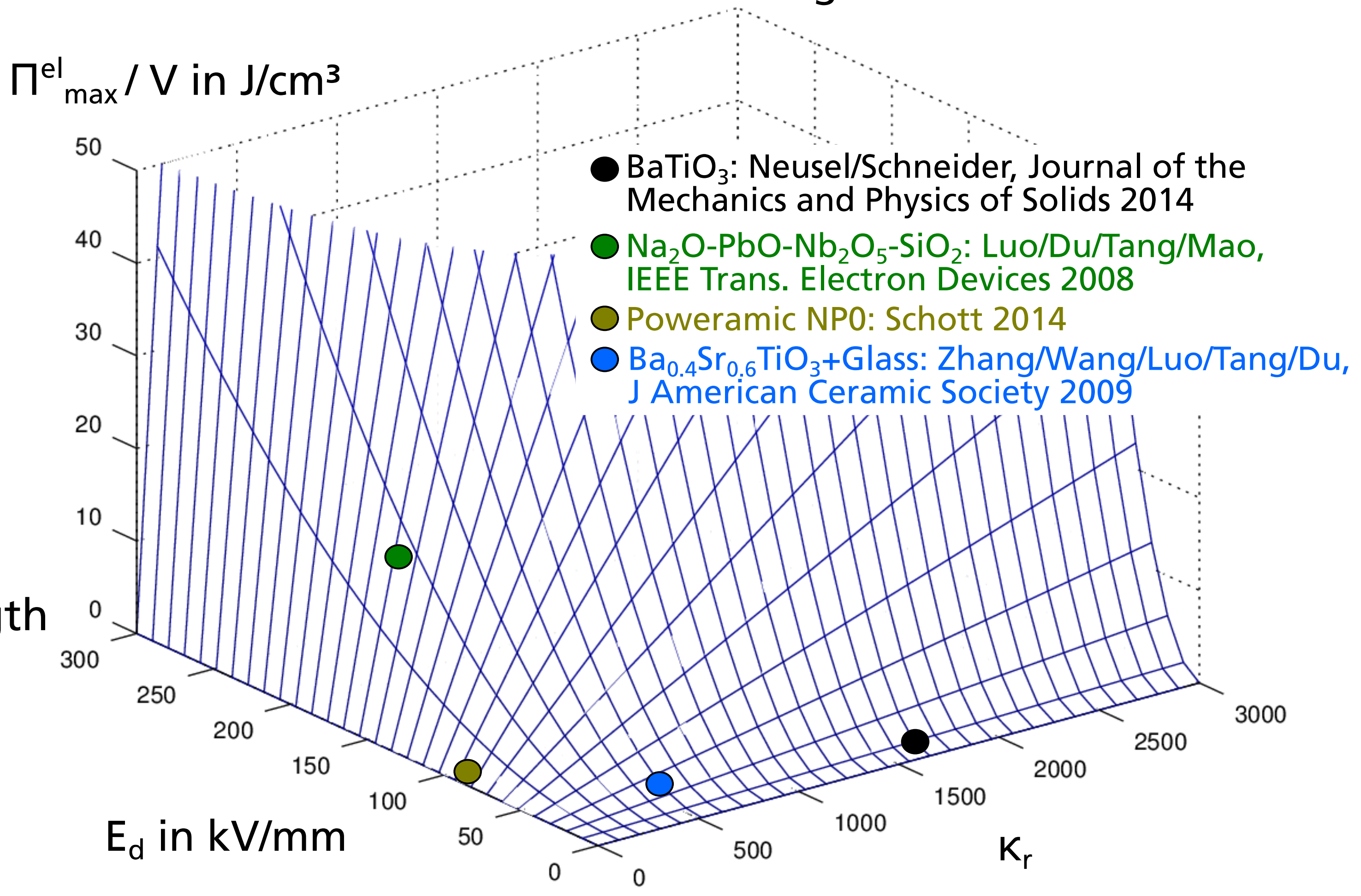


Motivation: High Energy Density and High Capacity per Volume

- $\frac{\Pi_{max}^{el}}{V} = \frac{1}{2}\,\kappa \cdot E_d^2$
- $\frac{C_{max} \cdot {U^a}^2}{V} = \kappa \cdot E_d^2$

Π_{max}^{el} = maximal energy
V = volume
κ = permittivity
E_d = dielectric breakdown strength
C_{max} = maximal capacity
U^a = applied voltage

Energy density as function of relative permittivity and dielectric breakdown strength



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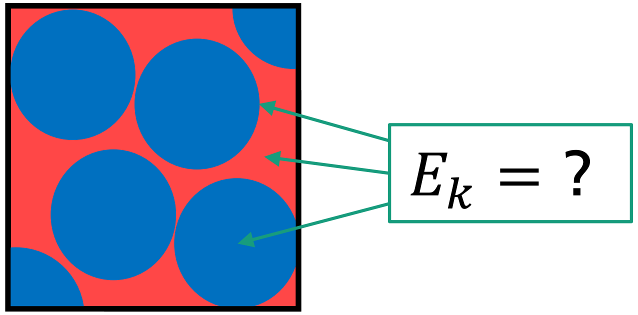
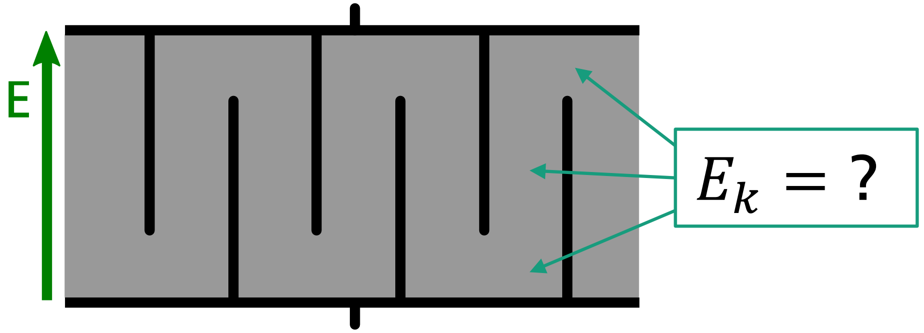
Motivation: Simulation of Glass-Ceramic

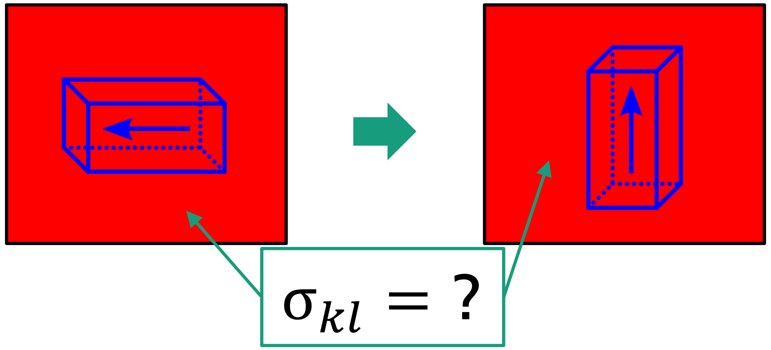
Distribution of electric field in microstructure



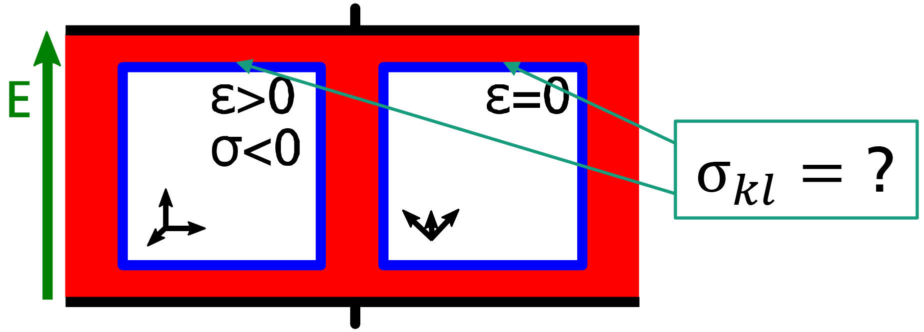
Inhomogeneous electric fields in component



Switching behavior: effect of matrix on particle



Mechanical tensions in microstructure caused by ferroelectric and piezoelectric grains



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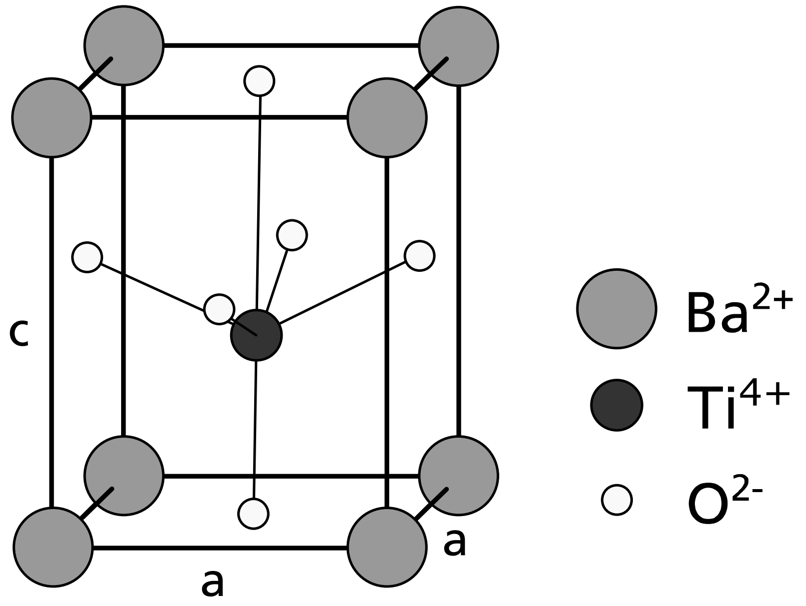
CONTENT

Basics: Barium Titanate

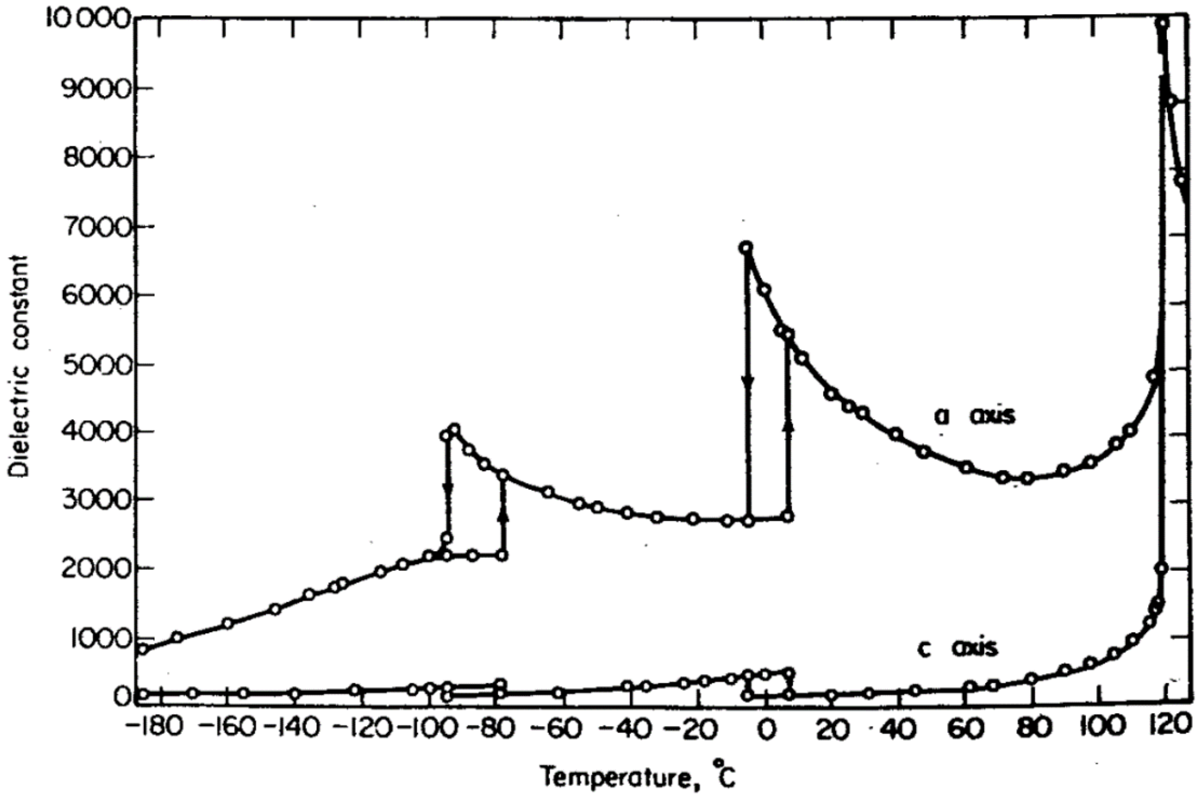
- Tetragonal unit cell below Curie-Temperature

  Perovskite structure



- High permittivity

(1)

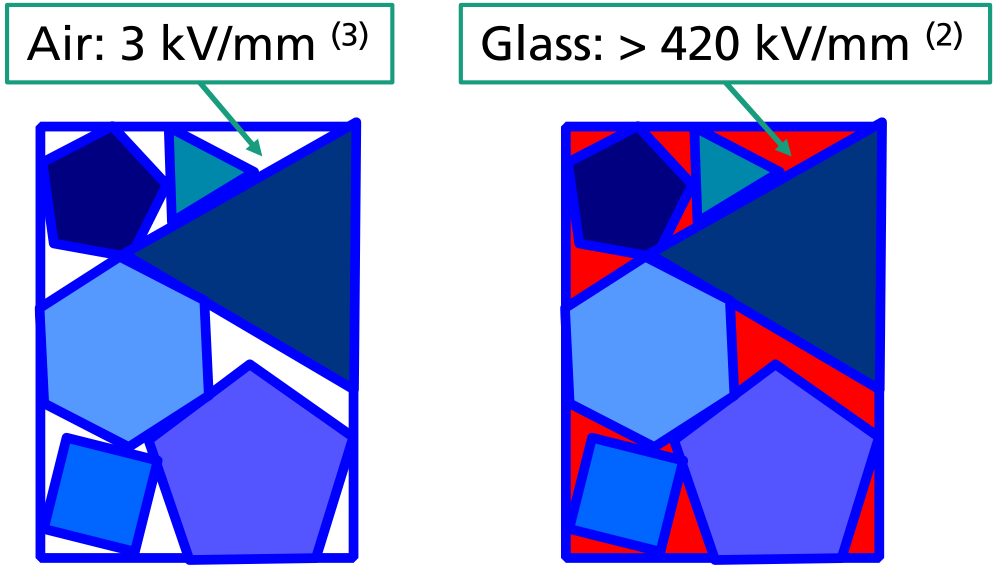


(1) Jaffe, B.; Cook, W. R.; Jaffe, H. L. *Piezoelectric ceramics*; Non-metallic solids v. 3; Academic Press: London, New York, 1971.

Basics: Advantages of Glass-Matrix

- Dielectric properties:

  Increase of breakdown strength



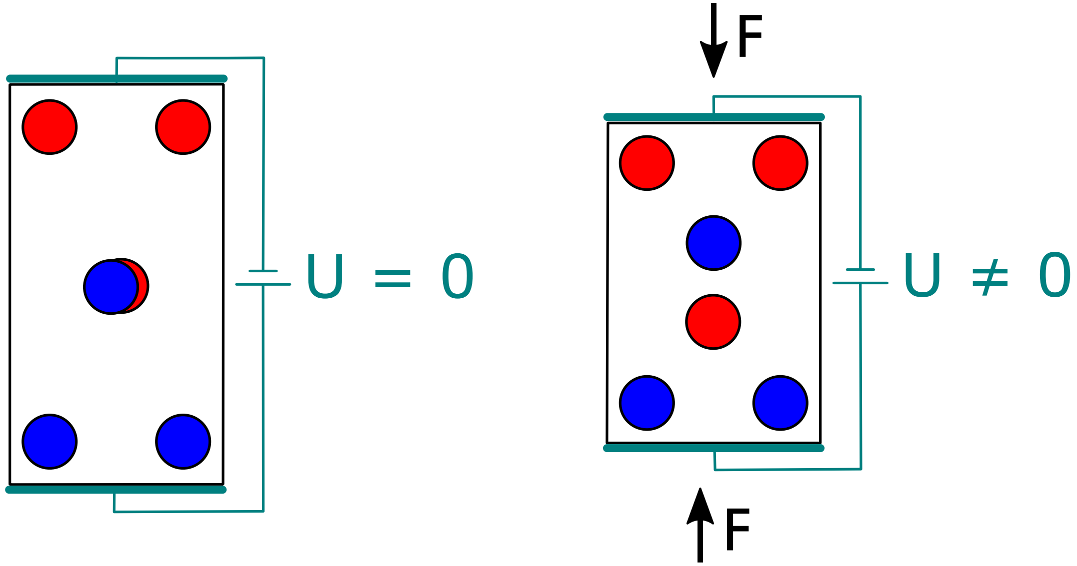
- Mechanical properties:
  - Decrease of micro cracks, voids and pinholes [1]
  - Reduction of residual stresses [1]
  - Increase of formability [1]

- Nanosized grains of glass-ceramic:
  - No switching
  - No hysteresis
  - Better mechanical strength

(1) Yao, K.; Zhu, W. *Thin Solid Films [Online]* **2002**, *408* (1–2), 11–14.
(2) Smith, N. J.; Rangarajan, B.; Lanagan, M. T.; Pantano, C. G. *Materials Letters [Online]* **2009**, *63* (15), 1245–1248.
(3) Küchler, A. *Hochspannungstechnik. Grundlagen - Technologie - Anwendungen*; VDI-Buch; Springer Berlin: Berlin, 2009.

Basics: Piezoelectric Effect

- Applying a force F on a piezoelectric material results in an electric tension U



- $\tilde{\varepsilon}_{ij} = \tilde{S}^E_{ijkl}\tilde{\sigma}_{kl} + \tilde{d}_{kij}\tilde{E}_k + \tilde{\varepsilon}^r_{ij}$
- $\tilde{D}_i = \tilde{d}_{ikl}\tilde{\sigma}_{kl} + \tilde{\kappa}^\sigma_{ik}\tilde{E}_k + \tilde{P}^r_i$
  - Macroscopic physical variables
    $\tilde{\sigma}_{kl}$ (mechanical tension), $\tilde{E}_k$ (electric field), $\tilde{D}_i$ (dielectric displacement)
  - Macroscopic material properties
    - $\tilde{\varepsilon}^r_{ij}$ (remanent strain), $\tilde{P}^r_i$ (remanent polarization)
    - $\tilde{S}^E_{ijkl}$ (compliance), $\tilde{d}_{kij}$ (piezoelectric coupling tensor), $\tilde{\kappa}^\sigma_{ik}$ (relative permittivity)

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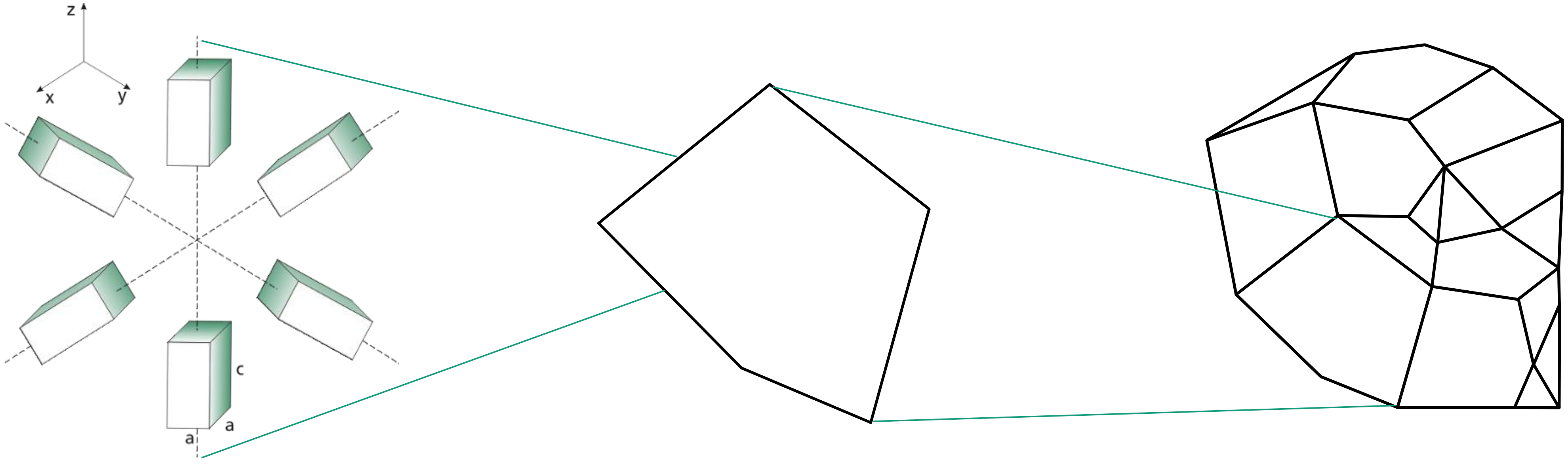
Simulation: Multi-Scale Model

- Micro scale
  Unit cell
  - Orientation
  - Properties
- Meso scale
  Grain
- Macro scale
  Polycrystal



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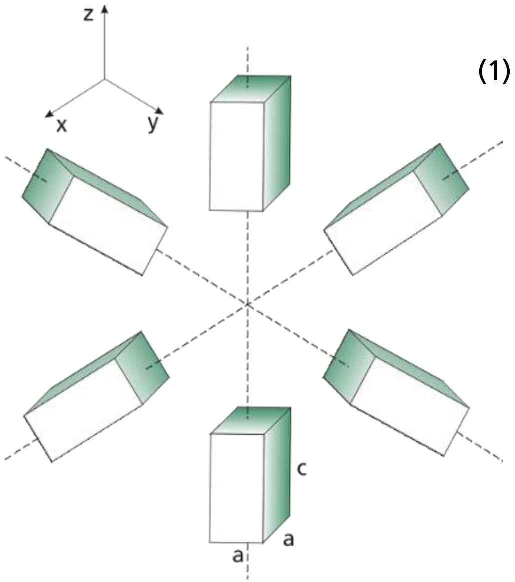
Simulation: Multi-Scale Model
Micro Scale - Unit Cell

- 6 possible poling directions
  - corresponding to volume fractions v^{α}
  - $\sum_{\alpha=1}^{6} v^{\alpha} = 1$
- Microscopic material properties



$$\hat{\varepsilon}^{S,+z} = \begin{pmatrix} -0.5 & 0 & 0 \\ 0 & -0.5 & 0 \\ 0 & 0 & 1 \end{pmatrix} \cdot \varepsilon^{S}, \quad \hat{P}^{S,+z} = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \cdot P^{S},$$

$$\hat{\kappa}^{\sigma,+z} = \begin{pmatrix} \kappa_{11} & 0 & 0 \\ 0 & \kappa_{11} & 0 \\ 0 & 0 & \kappa_{33} \end{pmatrix}, \quad \hat{d}^{+z,T} = \begin{pmatrix} 0 & 0 & d_{31} \\ 0 & 0 & d_{31} \\ 0 & 0 & d_{33} \\ 0 & d_{15} & 0 \\ d_{15} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \hat{S}^{E,+z} = \begin{pmatrix} s_{11} & s_{12} & s_{13} & 0 & 0 & 0 \\ s_{12} & s_{11} & s_{13} & 0 & 0 & 0 \\ s_{13} & s_{13} & s_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & s_{66} \end{pmatrix}$$

(1) after: Neumeister, P. *Mikromechanische Modellierung morphotroper PZT-Keramiken*. 2011.

Simulation: Multi-Scale Model
Meso Scale - Grain

- Mesoscopic physical variables

  σ_{kl}, E_k

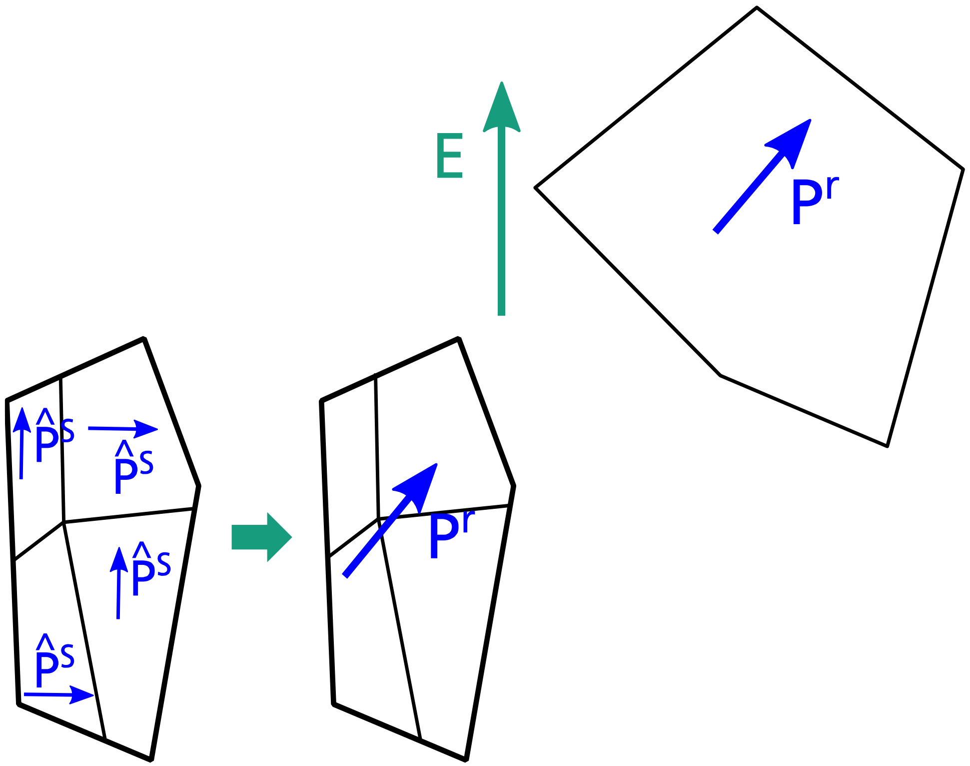
- Mesoscopic material properties

  Homogenization

  $P_i^r = \sum_{\alpha=1}^{6} \hat{P}_i^{S,\alpha} \cdot v^\alpha$

  (analog for ε_{ij}^r, κ_{ik}^σ, S_{ijkl}^E, d_{kij})

  $v^\alpha =$ volume fraction of domains polarized in directions +x, +y, +z, -x, -y, -z



Simulation: Multi-Scale Model
Macro Scale - Polycrystalline Compound

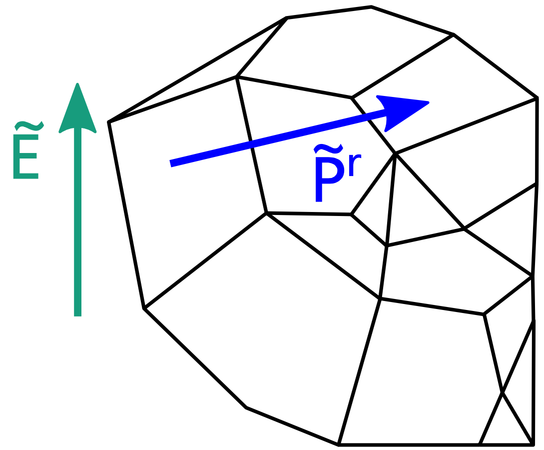
- Macroscopic physical variables

  $\tilde{\sigma}_{kl}, \tilde{E}_k$

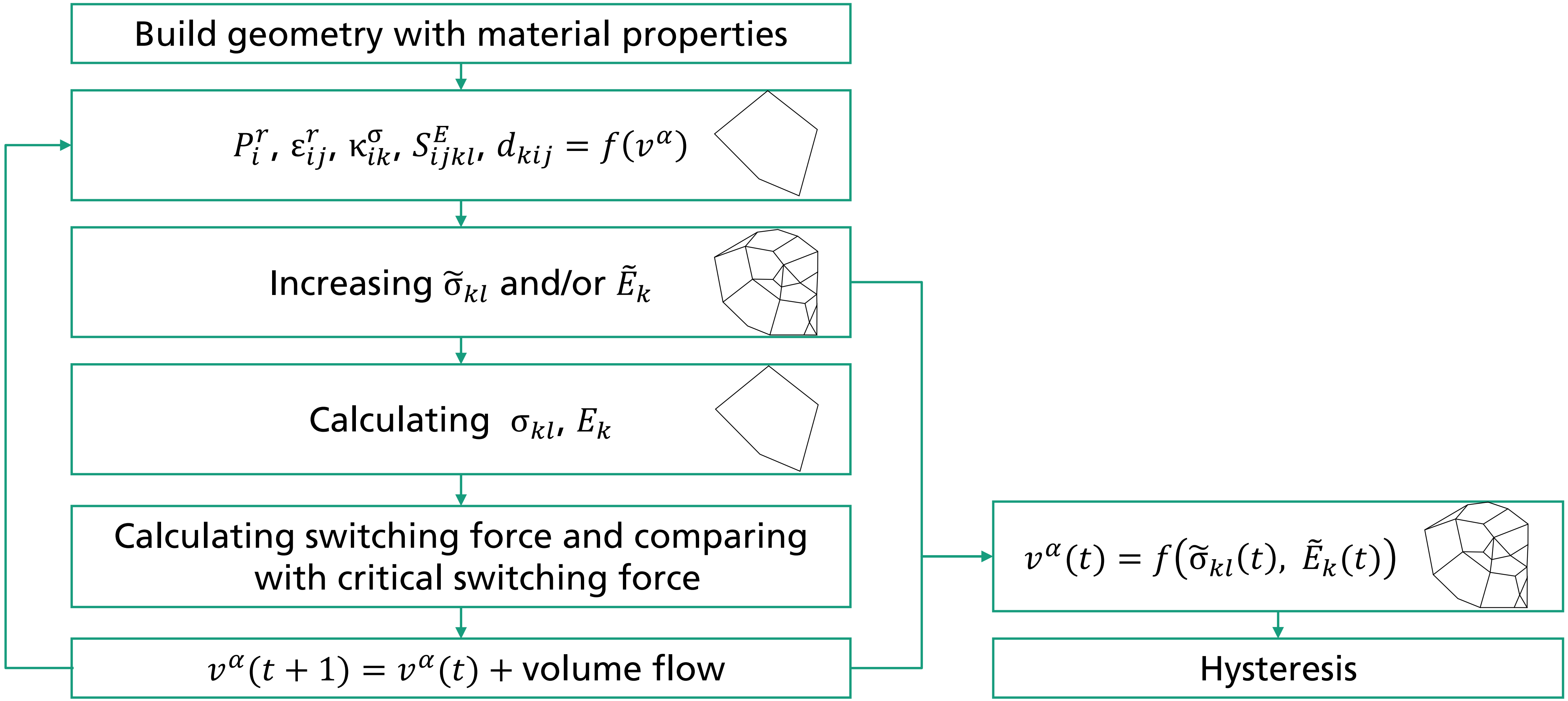
- Macroscopic material properties

  Homogenization

  - Equal sized grains: $\tilde{P}_i^r = \sum_i P_i^r$ (analog for $\tilde{\varepsilon}_i^r, \tilde{S}_{ijkl}^E, \tilde{d}_{kij}, \tilde{\kappa}_{ik}^{\sigma}$)
  - Variable sized grains: FEM



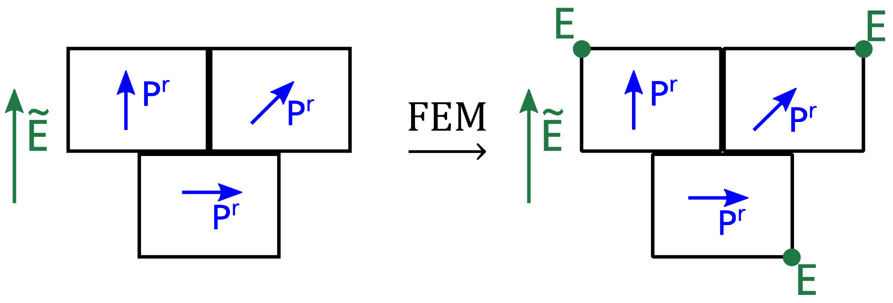
Simulation: Process

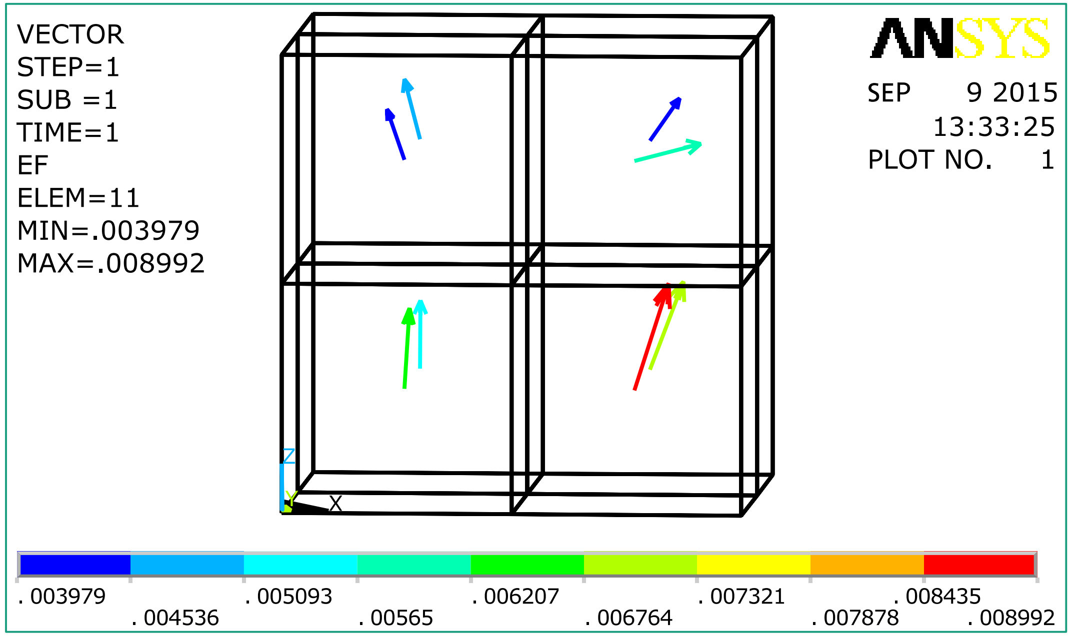


Simulation: Material Description
Mesoscopic Loads

Mesoscopic loads calculated out of macroscopic loads and mesoscopic material properties by FEM

$$\tilde{\sigma}_{kl},\ \tilde{E}_k,\ P_i^r,\ \varepsilon_{ij}^r,\ \kappa_{ik}^{\sigma},\ S_{ijkl}^E,\ d_{kij} \xrightarrow{\text{FEM}} \sigma_{kl},\ E_k$$





Simulation: Material Description
Volume Flows – Switching Criterion

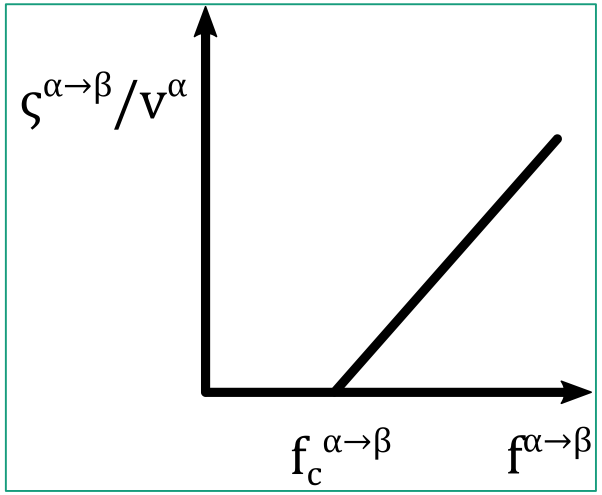
- $f^{\alpha\to\beta} = \sigma_{ij}\Delta\hat{\varepsilon}_{ij}^{S,\alpha\to\beta} + E_i\Delta\hat{P}_i^{S,\alpha\to\beta}$ [1, 2] driving force for switching system $\alpha \to \beta$

- Switching rule: $\varsigma^{\alpha\to\beta} = \begin{cases} k_s^{\alpha\to\beta}\left(\frac{f^{\alpha\to\beta}}{f_c^{\alpha\to\beta}} - 1\right) v^{\alpha} & \text{if } f^{\alpha\to\beta} \geq f_c^{\alpha\to\beta} \\ 0 & \text{else} \end{cases}$ [3]

$\varsigma^{\alpha\to\beta}$ = volume flow from domain orientation α to domain orientation β

$k_s^{\alpha\to\beta}$ = creep constant / inverse viscosity

$f_c^{\alpha\to\beta}$ = critical driving force



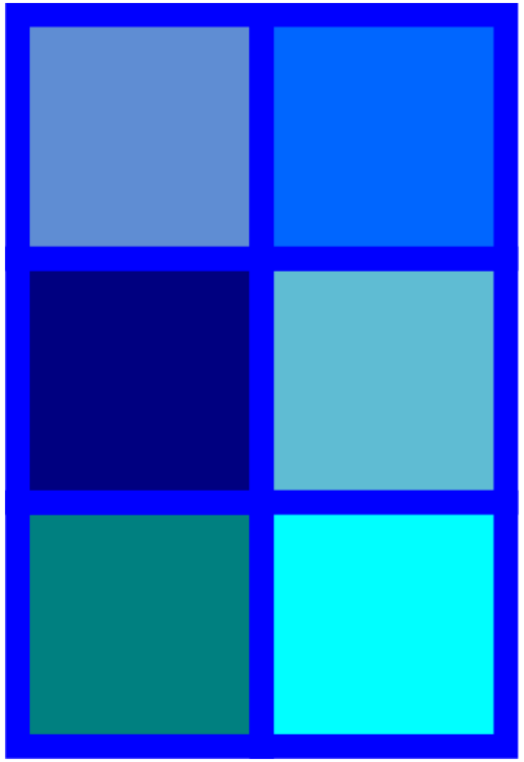
(1) Landis, C. M. *Current Opinion in Solid State and Materials Science* **2004,** *8* (1), 59–69.

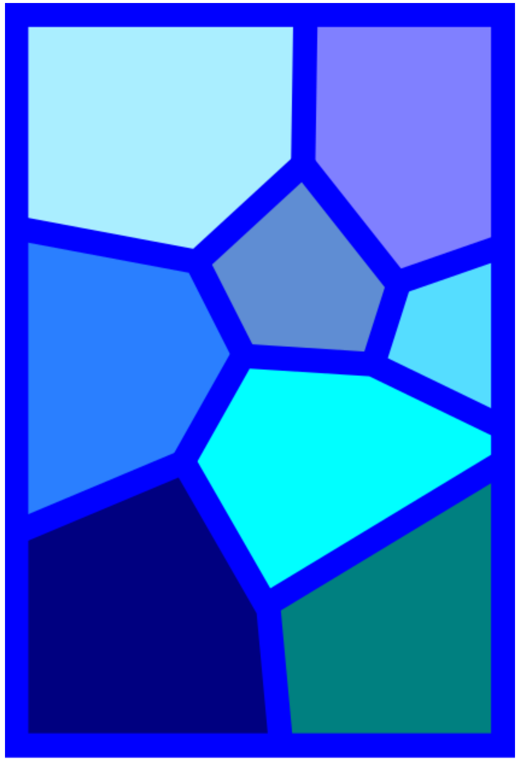
(2) Kessler, H.; Balke, H. *Journal of the Mechanics and Physics of Solids [Online]* **2001,** *49* (5), 953–978.

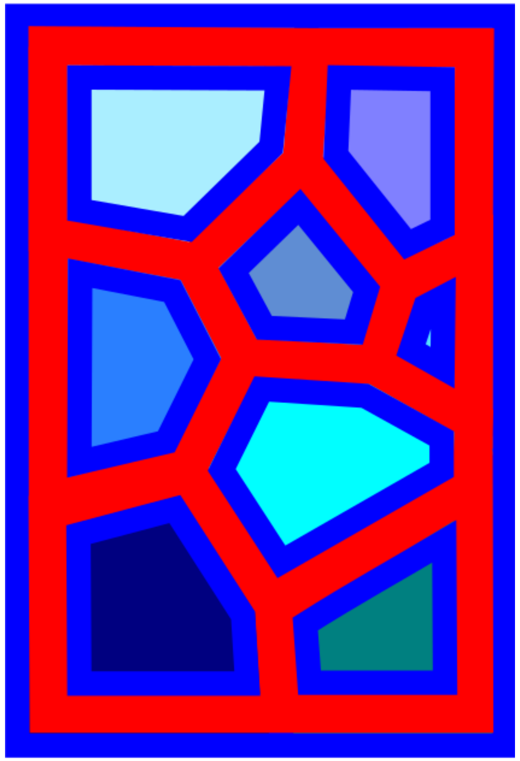
(3) Neumeister, P. *Mikromechanische Modellierung morphotroper PZT-Keramiken*. 2011

Simulation: Geometric Representation

- BaTiO3 polycrystal cubed
- BaTiO3 polycrystal voronoi
- Glass-ceramic voronoi







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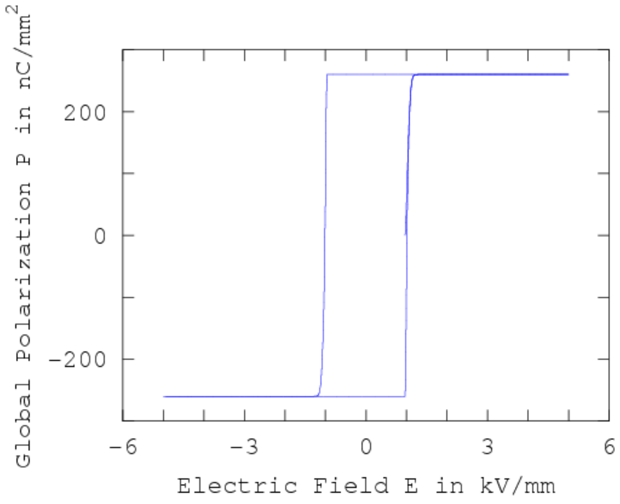
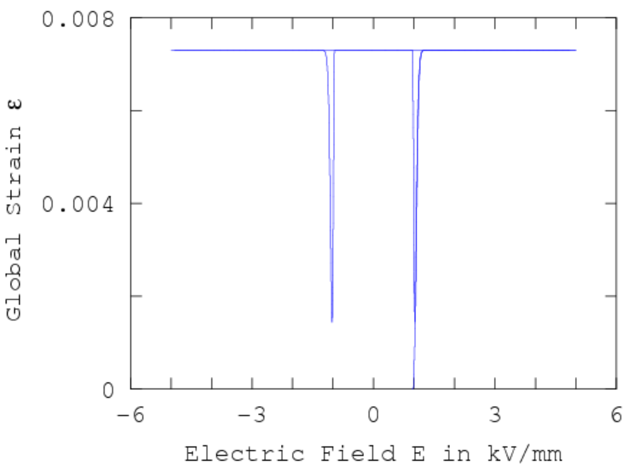
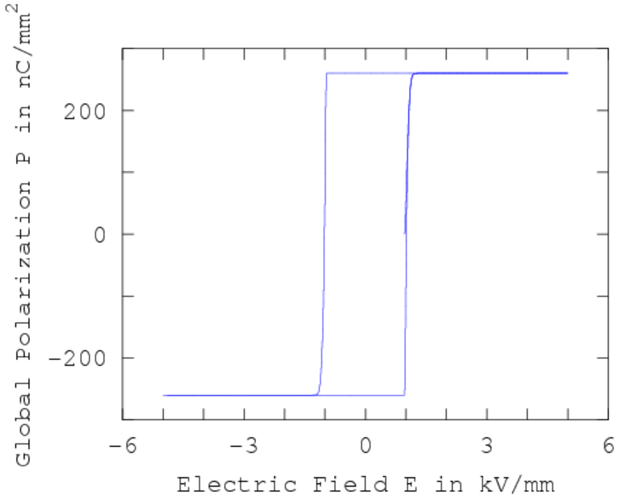
Results
Validation

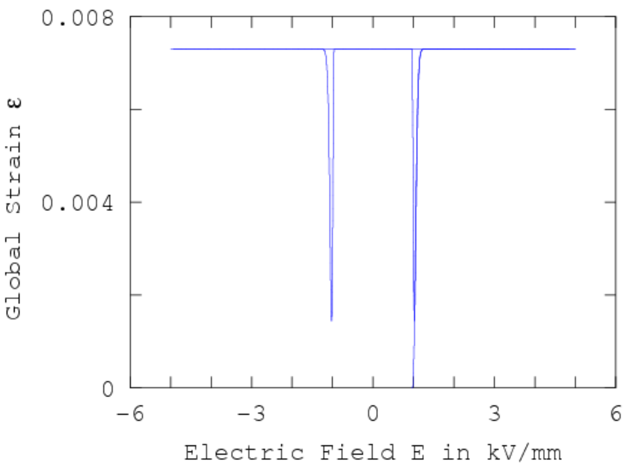
- Single crystal one element
  <100>-orientation





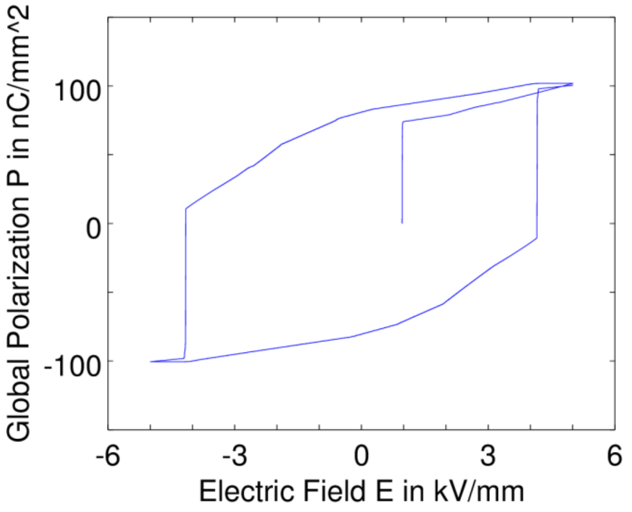
- Single crystal 8 elements
  All elements <100>-orientation

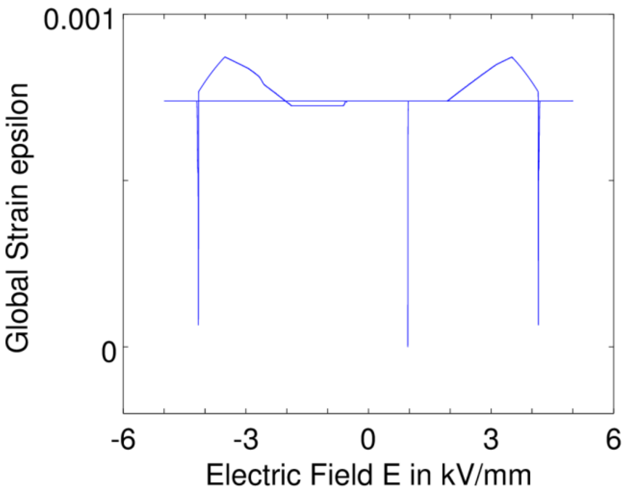




Results

- Polycrystal 8 elements
  Arbitrarily oriented elements





- Polycrystal 27 elements
  Arbitrarily oriented elements

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Fabrication of Glass-Ceramic: Composition and Procedure

- Glass: Composition and Melting [1]
  - [4]

Component	Mol%
Al_2O_3	10
BaO	38.2
SiO_2	20
TiO_2	31.8

  - Melting for 2 h at 1550 °C
  - Casting onto brass plate
  - Cooling from 650 °C (1 K/min)

- Glass-Ceramic Crystallized: [2]
  - Heating to 950 °C (1K/min)
  - Holding for 15 h in air
  - Cooling (5 K/min)

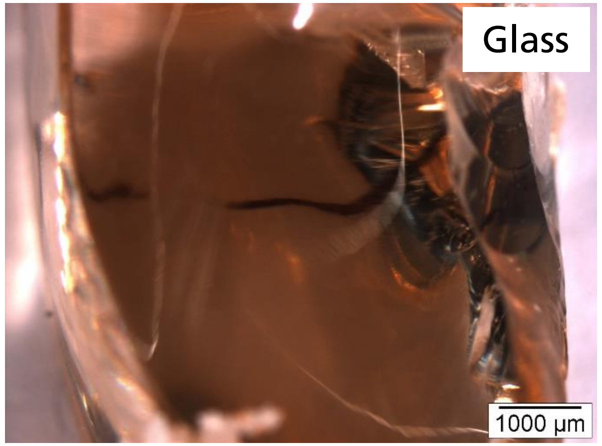
- Glass-Ceramic Sintered: [3]
  - Grinding to powder
  - Heating to 870 °C (3 K/min)
  - Holding for 30 min in air
  - Cooling (3 K/min)

(1,2,3) J. Schilm, K. Wätzig, I. Eichler

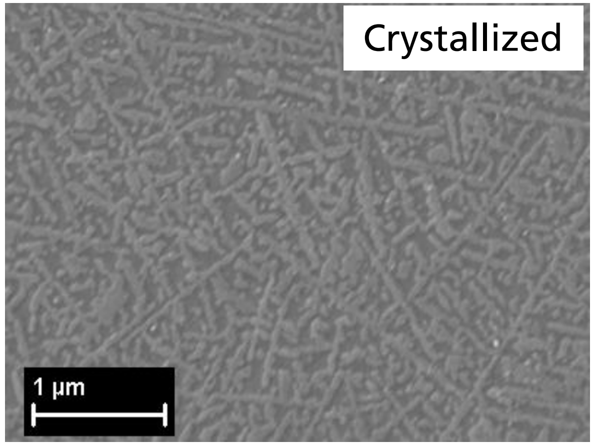
(4) McCauley, D.; Newnham, R. E.; Randall, C. A. *J American Ceramic Society* **1998,** *81* (4), 979–987

Fabrication of Glass-Ceramic:
Dielectric Properties and Microstructure

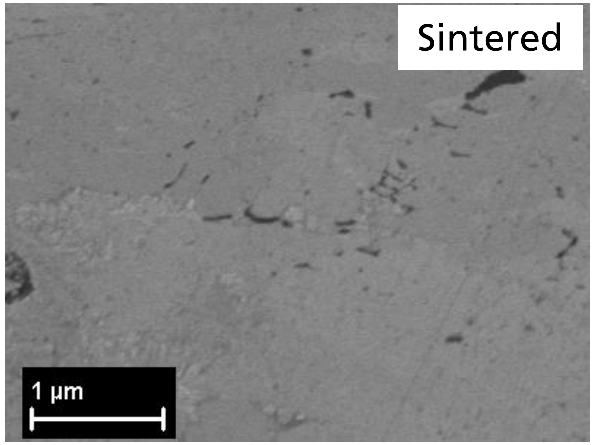
	Capacity	Loss Tangent	Relative Permittivity	Dielectric Breakdown Strength	Coercivity	Polarization
	[nF] (1kHz)	[1e-4]	[-]	[kV/mm]	[kV/mm]	[µC/cm²]
Crystallized	0.011	930	588	6.44	0.6152	1.0681
Sintered	0.001	9921	53	9.57	0.2610	0.1341



(1)



(2)



(3)

(1) Chr. Müller

(2,3) S. Höhn, S. Szokup

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CONTENT

Summary

- Glass-Ceramic Material

  Dielectric material for capacitor application
  - High dielectric breakdown strength
  - High energy density
- Simulation

  Energy based switching criterion
  - Validation of code on single crystal
  - First calculations polycrystal with 8 and 27 cubed elements
- Fabrication
  - Crystallized and Sintered Samples
  - Good relative permittivity, poor dielectric breakdown strength

Acknowledgement

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Attract 64-692 018

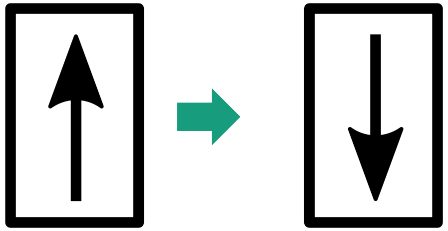
„Werkstoffmechanik der Funktionskeramiken:
Von der Mikrostruktur zum Bauteilverhalten“

Thank you for your attention!

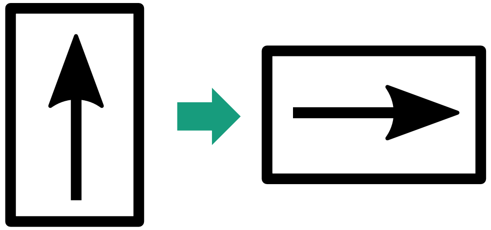
Basics: Ferroelectric Domains

- Regions with unit cells polarized in the same direction
- Domains separated by domain walls
- Movement of domain walls

  - 180° switching

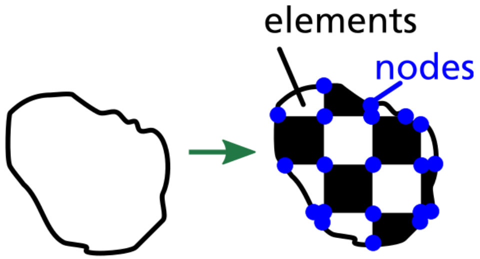


  - 90° switching



Basics: FEM (Finite Element Method)

- Numerical method for calculating physical fields
- Procedure:
  - Segmentation of desired volume into small elements

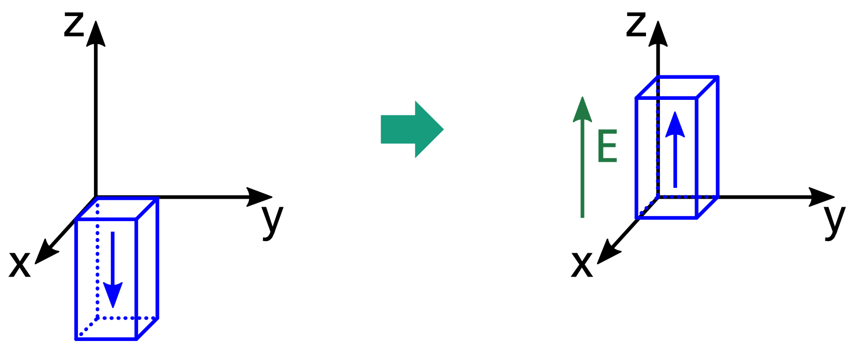


  - Approach for each element
  - Linear combination of element approaches
  - Insertion into differential equation
  - Solving equation system by aid of boundary, transition and initial conditions

Simulation: Material Description
Volume Flows – Switching Criterion I

- $f^{\alpha\rightarrow\beta} = \sigma_{ij}\Delta\hat{\varepsilon}_{ij}^{S,\,\alpha\rightarrow\beta} + E_i\Delta\hat{P}_i^{S,\,\alpha\rightarrow\beta}$ (1, 2) driving force for switching system $\alpha \rightarrow \beta$

  - $f^{\alpha\rightarrow\beta} = E_i\left(\hat{P}_i^{S,\,\beta} - \hat{P}_i^{S,\,\alpha}\right)$
  - $f^{\alpha\rightarrow\beta} = \begin{pmatrix}0\\0\\1\end{pmatrix} E \cdot \left(\begin{pmatrix}0\\0\\1\end{pmatrix} - \begin{pmatrix}0\\0\\-1\end{pmatrix}\right) P^S$



  - $f^{\alpha\rightarrow\beta} = \begin{pmatrix}0\\0\\1\end{pmatrix} E \cdot \left(\begin{pmatrix}0\\0\\1\end{pmatrix} - \begin{pmatrix}0\\-1\\0\end{pmatrix}\right) P^S$

(1) Landis, C. M. *Current Opinion in Solid State and Materials Science* **2004,** *8* (1), 59–69.
(2) Kessler, H.; Balke, H. *Journal of the Mechanics and Physics of Solids [Online]* **2001,** *49* (5), 953–978.