Phase field modeling of domain evolution in ferroelectric materials

Introduction

Phase field modeling provides an efficient tool for the study of domain evolution in ferroelectric materials. Such models naturally introduce an inner length scale which represents the width of the interfaces between domains (domain walls). This inner length scale is of the order of a few unit cells, i.e. about 0.8 nm–2 nm. The focus of this presentation is on size effects in a) the switching behavior of ferroelectric thin films and b) the microstructure evolution in ferroelectric nanodots.

Phase field model

The phase field model contains the definitions for the strain and the electric field, as well as the mechanical and electrostatic balance equations (cf. [1]):

$$\boldsymbol{\varepsilon} = \operatorname{sym}(\nabla \boldsymbol{u}) \,, \qquad \boldsymbol{E} = -\nabla \varphi \,,$$
$$\operatorname{div} \boldsymbol{\sigma} = \boldsymbol{0} \,, \qquad \operatorname{div} \boldsymbol{D} = 0 \,.$$

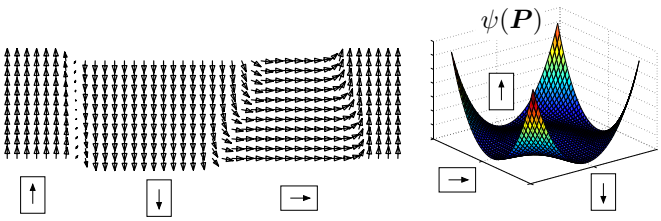
These are supplemented by the constitutive equations

$$\boldsymbol{\sigma} = \mathbb{C}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^0) - \boldsymbol{e}^T \boldsymbol{E} \,, \qquad \boldsymbol{D} = \boldsymbol{e}(\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}^0) + \boldsymbol{\epsilon} \boldsymbol{E} + \boldsymbol{P} \,,$$
$$\boldsymbol{\Sigma} = \kappa_{\mathrm{g}} \frac{\gamma \ell}{P_0^2} \nabla \boldsymbol{P} \,, \qquad \beta \dot{\boldsymbol{P}} = \operatorname{div} \boldsymbol{\Sigma} - \frac{\partial H}{\partial \boldsymbol{P}} \,.$$

Therein γ is the specific energy of a 180° interface, and ℓ is the inner length scale parameter which is calibrated to coincide with the width of that interface (see [2]). The quantity $\boldsymbol{P}$ is the order parameter of the model describing the state of spontaneous polarization. The electric enthalpy is given by

$$H = \tilde{H}^{\mathrm{bulk}}(\boldsymbol{\varepsilon}, \boldsymbol{E}, \boldsymbol{P}) + \kappa_{\mathrm{s}} \frac{\gamma}{\ell} \psi(\boldsymbol{P}) + \frac{1}{2} \kappa_{\mathrm{g}} \frac{\gamma \ell}{P_0^2} \|\nabla \boldsymbol{P}\|^2 \,. \qquad (1)$$

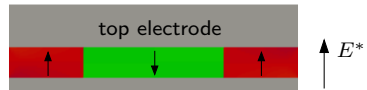
The non-convex polynomial $\psi(\boldsymbol{P})$ ensures phase separation and exhibits four minima in the 2d case:

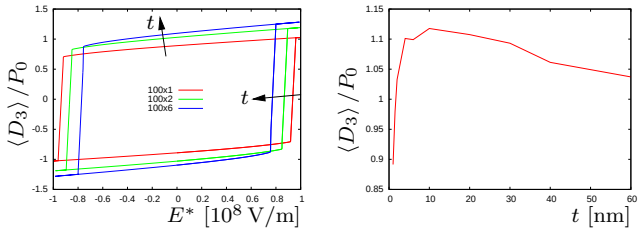


The dependency of the material tensors $\mathbb{C}$, $\boldsymbol{e}$, $\boldsymbol{\epsilon}$, and $\boldsymbol{\varepsilon}^0$ (the spontaneous strain) on the order parameter $\boldsymbol{P}$ can be obtained by employing an invariant formulation, see [3] for details. The model is implemented in a finite element framework (cf. [1]).

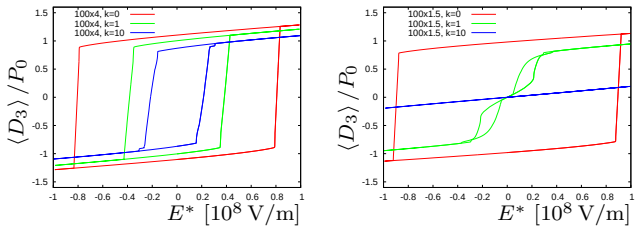
Poling behavior of $BaTiO_3$ (BTO) thin films

The poling behavior of BTO thin films is investigated by considering a BTO layer between two $SrRuO_3$ (SRO) electrodes (see screenshot). An electric field E^* is applied by prescribing a potential difference between the two electrodes. The misfit strain between the electrode and the active material allows for the poling process to be initiated at the lateral sides of the film. Poling is achieved by the movement of the two 180° domain walls. The resulting electrical hysteresis curves show that the switchable (or remanent) polarization decreases as the film thickness is reduced to 1 nm (while the coercive fields increase):



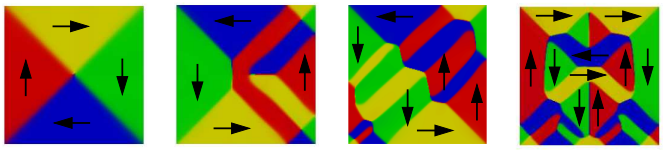


Experimental findings suggest that the remanent polarization can vanish if the film thickness is below about 2.5 nm. This effect can be captured by applying the Robin type boundary condition $\boldsymbol{\Sigma}\boldsymbol{n} - k\boldsymbol{P} = \boldsymbol{0}$ at the SRO-BTO interfaces. For a 100 nm×1.5 nm film the remanent polarization can be forced to vanish if the Robin parameter k is chosen sufficiently large:



Size effects in microstructure evolution

Size effects are also relevant for the microstructure evolution in ferroelectric nanodots. Using initially random polarizations, we let the microstructure of differently sized $BaTiO_3$ nanodots relax until thermodynamical equilibrium is reached. The boundary conditions are assumed as charge free and mechanically clamped, i.e. $\boldsymbol{u} = \boldsymbol{0}$. The final domain patterns vary in their complexity:



24 nm × 24 nm | 48 nm × 48 nm | 72 nm × 72 nm | 96 nm × 96 nm

References

[1] D. Schrade et al., *Comp. Meth. Appl. Mech. Eng.* 196:4365, 2007.
[2] D. Schrade et al., *Arch. Appl. Mech.* 83:1393–1413, 2013.
[3] D. Schrade et al., *Int. J. Sol. Struct.* 51:2144–2156, 2014.

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