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# Leakage currents in nanogenerator concepts in phase field simulations

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**Micro Abstract**
Efficient technologies for energy harvesting are in the focus of recent research. Our nanogenerator transforms parasitic mechanical oscillations into usable electric energy. When an electric field exists between electrodes, leakage currents appear if the ferroelectric ceramic is semiconducting. We focus on different formulations of leakage currents for semiconducting ceramics, usually given as scalar equations. We enhance our phase field model to account for these effects in space.

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## Introduction

The effect of different formulations of leakage current for energy harvesting on the nanoscale with ferroelectric barium titanate ($BaTiO_3$) is discussed. Engineered electric polarization domain topologies within the ferroelectric film enables the conversion of mechanical into electrical energy. A key feature is the alignment of the domain topology, resulting from mechanical oscillations and the structured top electrode.

Ambient parasitic vibrations serve as the energy source for this nanogenerator concept. These vibrations elongate or compress the $BaTiO_3$ ferroelectric film unidirectionally, which results in a switch of the emerging electric dipole. The ferroelectric film is deformed such that the electric polarization reorders and, consequently, causes a charge flow between locally separated electrodes. For the energy harvesting process, the electric polarization in the ferroelectric between the top electrode and the conductive seed layer needs to evolve from a specified initial state into a preferred final state.

Charging an electric storage medium implies that a gradient of electric potential exists between electrodes. Thus, leakage currents may appear between these electroded surfaces if the ferroelectric ceramic is not a perfect insulator. To optimize the nanogenerator concept, the prediction of leakage current is a highly debated topic.

In literature, e.g. [1, 2, 4, 5], different mechanisms for leakage current density $J$ in ceramics are discussed. These are as linear (Ohm's law, $OL$), quadratic (space-charge-limited current, $SCLC$) or exponential (Schottky emission, $SE$) relationships to the electric field. We consider these different descriptions to optimize our nanogenerator concept.

## 1 Phase field formulation for leakage currents for a nanogenerator concept

Our finite element phase field model is based on the Ginzburg-Landau free energy as suggested by Su and Landis [6] and extended by Münch and Krauss [3]. The mechanical displacements $u_i$, electric polarization $P_i$, and the electric potential $\varphi$ are the nodal degrees of freedom with $i$ declaring the number of dimensions. General elastic, piezoelectric and dielectric properties of $BaTiO_3$ fit the form of the free energy. Since the free energy depends also on an order parameter and its gradient, here, the order parameter is the electric polarization $P_i$ describing uniform polarized domains. Within the ferroelectric film $\mathcal{B}$, the dielectric displacement $D_i$ and the electric field $E_i$ are given by

$$D_i = P_i + \kappa_0 \, E_i \quad \text{and} \quad E_i = -\varphi_{,i} \quad \text{in} \quad \mathcal{B}, \tag{1}$$

with the permittivity of free space $\kappa_0$. The phase field model combines linear kinematics and quasi-static Maxwell's equations, such that

$$\varepsilon = \frac{1}{2}(u_{i,j} + u_{j,i}) \quad \text{and} \quad D_{i,i} - q = 0 \quad \text{in} \quad \mathcal{B}, \tag{2}$$

with the volume charge density $q$. The Ginzburg-Landau equation as well as the mechanical and electrical equilibrium equations can be expressed in a virtual work statement

$$\int_{\mathcal{B}} \left( \beta \dot{P}_i \delta P_i + \sigma_{ij} \, \delta\varepsilon_{ij} - D_i \, \delta E_i + \eta_i \, \delta P_i + \xi_{ij} \, \delta P_{i,j} \right) \mathrm{d}V \tag{3}$$

$$= \int_{\mathcal{B}} (b_i \delta u_i - q \delta\varphi) \, \mathrm{d}V + \int_{\partial\mathcal{B}} (t_i \delta u_i - \omega \delta\varphi + \xi_{ij} \, n_j \delta P_i) \, \mathrm{d}A \, . \tag{4}$$

To account for leakage current in the ferroelectric film, we consider the time integration of the conservation of charge equation, which enhances the finite element equation system by

$$K^{LC}_{\varphi\varphi} = \int_{\mathcal{B}} \int_t \Delta J_i \, \mathrm{d}t \, \delta\varphi_{,i} \, \mathrm{d}V, \quad \text{and} \tag{5}$$

$$R^{LC}_{\varphi} \int_{\mathcal{B}} \int_t J_i \, \mathrm{d}t \, \delta\varphi_{,i} \, \mathrm{d}V \, . \tag{6}$$

## 2 Constitutive laws for semiconductivity

### 2.1 Ohm's law

Ohm's law represents the linear relation between leakage and electric field

$$J_i = \Gamma \, E_i \, , \tag{7}$$

with the conductivity $\Gamma$. This conduction mechanism is observed at low electric fields in e.g. [1].

### 2.2 Space charge limited current

The space charge limited current (SCLC) relation assumes a quadratic dependency on the electric field. To account for also three-dimensional effects in leakage current with quadratic dependency on the electric field, the SCLC scalar formulation eq.(8) is extended to eq.(9)

$$J = \frac{9}{8} \mu \, \kappa_0 \, \varepsilon_r \, E^2 \, , \tag{8}$$

$$J_i = \frac{9}{8} \mu \, \kappa_0 \, \varepsilon_r \, E \, E_i \, , \tag{9}$$

with the carrier mobility $\mu$, the relative dielectric constant $\varepsilon_r$ and $E = \sqrt{E_i \, E_i}$. This mechanism is expected at higher values for the electric field.

### 2.3 Schottky emission

The Schottky emission is a formulation for the leakage current density with exponential dependency to the electric field. In scalar form it reads

$$J = A^* \, T^2 \exp\left[ \frac{-e \, \phi_0}{kT} \right] \left( \exp\left[ \frac{\sqrt{e/(4\pi \, \varepsilon_d \, \kappa_0)}}{kT} \sqrt{E} \right] - 1 \right) . \tag{10}$$

To account for three-dimensional problems, the formulation yields

$$J_i = A^* T^2 \exp\left[\frac{-e\,\phi_0}{kT}\right] \left(\exp\left[\frac{\sqrt{e/(4\pi\,\varepsilon_d\,\kappa_0)}}{kT}\sqrt{E}\right] - 1\right) \frac{E_i}{E}\,, \tag{11}$$

with the Richardson-Dushman constant $A^*$, the absolute temperature $T$, the Boltzmann constant $k$, the Schottky barrier height $e\,\phi_0$, and the optical dielectric constant $\varepsilon_d$, see e.g. [2]. This conduction mechanism is observed in experimental testing at even higher electric fields than SCLC, e.g. [4].

## 3 Numerical examples

The studied example is a rectangular region of $BaTiO_3$ with inhomogeneous electric potential on the boundary, see Fig. 1. First, the distance between adjacent electrodes is taken to be the same as the height of the ferroelectric film. Further, polarization and mechanical displacements are fixed at all nodes for these simulations.

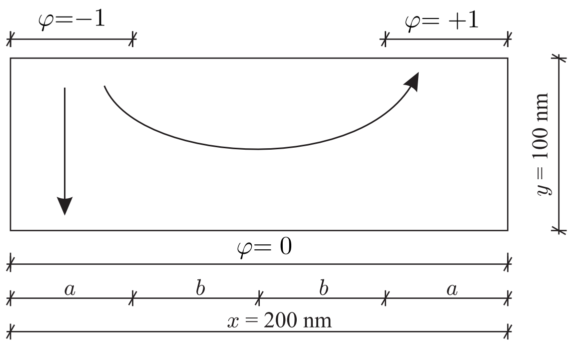


**Figure 1.** Ferroelectric film with inhomogeneous electroded boundary conditions.

Results from previous simulations [7] are explained briefly here. In particular, we use the following material parameters.

| Parameter | Value | Unity |
|---|---|---|
| Conductivity $\Gamma$ | $10^{-12}$ | $\frac{1}{\Omega\,m}$ |
| Permittivity of free space $\kappa_0$ | $8.854 \cdot 10^{-12}$ | $\frac{C}{V\,m}$ |
| Relative dielectric constant $\varepsilon_r$ | 100 | [-] |
| Carrier mobility $\mu$ | $1 \cdot 10^{-6}$ | $\frac{m^2}{V\,s}$ |

**Table 1.** Parameter for linear and quadratic leakage current density formulation

With these material parameters and comparing Ohm's law to the SCLC model, we found out that for Ohm's law $\approx$ 40% of the leakage current density flows directly in the bottom electrode and $\approx$ 60% to the adjacent top electrode. Whereas for the SCLC formulation, only 32% of the total current density flows to the bottom electrodes. Fig. 2 illustrates a comparison between linear (Ohm) and quadratic (SCLC) leakage current density. Results for the horizontal component of the leakage current density are displayed in normalized form for just one geometrical configuration. Our additional studies investigate the effects of polarization and mechanical displacements on the leakage current density as well as the Schottky emission model.

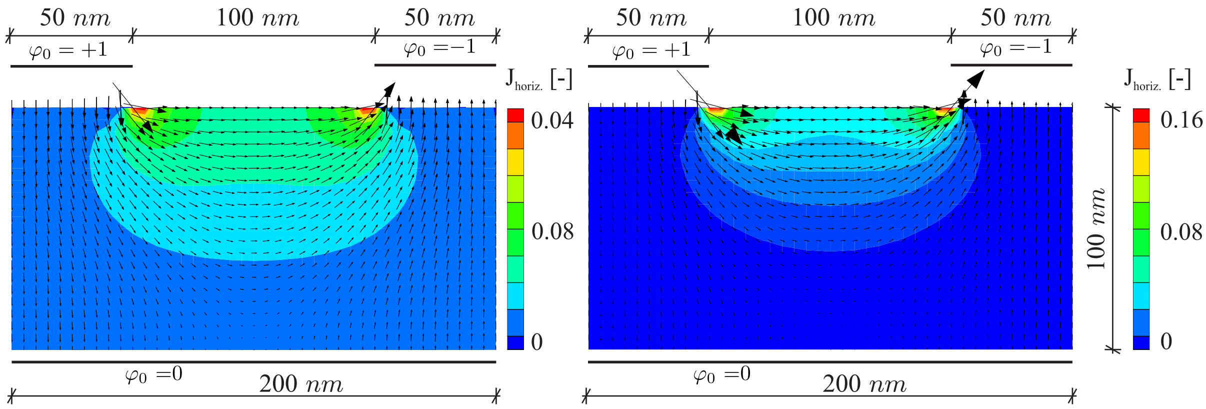


**Figure 2.** Resultant leakage current density for Ohm's law (left) and SCLC (right) displayed in vectors, horizontal leakage current in colors.

## Conclusions

We study different formulations for leakage current in ferroelectric nanogenerators. In our model, we generalize the conduction models to appropriate vectorial forms. This enables us to study how the distance between adjacent top electrodes needs to be varied for the optimization of the nanogenerator concept with respect to the leakage current density. Of primary interest is the effect of changing the ratio $a/h$ between the top electrode length $a$ and ferroelectric material height $h$.

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