FERROSIM: a software package for modeling the influence of internal fields and heat treatment on the polarization of ferroelectrics

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Abstract

In this paper, a software package is proposed for modeling the behavior of ferroelectric materials polarization characteristics depending on processing modes and composition, including gradient structures. Examples of calculated dielectric hysteresis loops for barium strontium titanate with homogeneous and gradient composition are given. The software package can also be used as a platform for training and development of new models in materials science.

Keywords: computer modeling, ferroelectric materials, polarization of ferroelectrics, unipolarity, internal fields.

Introduction

Currently, the development of methods of multi-scale computer modeling with the placement of software on computing clusters of hybrid architecture makes it possible to solve problems of creating new materials in the field of microelectronics. Widely used are methods of predictive computer modeling based on the application of multiscale approaches, which are directly related to the calculations at different scale levels, often in parallel modes and requiring increasing computational costs.

Ferroelectrics are nonlinear dielectrics that have spontaneous polarization in a certain temperature range, which changes significantly under the influence of external influences. Among the most studied and used in practice are barium titanate, triglycine sulfate, and so on. Ceramics based on solid solutions of barium-strontium titanate ($Ba_{1-x}Sr_xTiO_3$, BST) are one of the most widely studied objects in the field of ferroelectric materials science. High dielectric characteristics of such materials and the ability to control their parameters using external influences (in particular, an electric field) determine their widespread use in memory elements, capacitors, and microwave technology [1]. A distinctive property of BST is that its dielectric and ferroelectric characteristics change monotonically with a change in the Ba/Sr ratio. The phase transition temperature (Curie temperature) varies from $T_c \approx 120$°C for pure $BaTiO_3$ (x=0) to room temperature at x=0.35. The disadvantages of BST include a strong temperature dependence of its parameters in the Curie temperature region, which hinders its practical use. Gradient structures are of great interest in terms of obtaining materials with a diffuse phase transition [2]. The blurring parameters can be controlled by changing the magnitude of the composition gradient. This approach allows obtaining materials with high temperature stability of characteristics. The overwhelming majority of works on this topic, completed to date, are devoted to the study of structures obtained using various thin-film technologies.

However, the complexity of predicting the behavior of these materials, including the hysteresis dynamics, the influence of pyroelectric effects and the consequences of heat treatment, requires in-depth modeling. The lack of accurate computational tools that take into account the interaction of structural defects, domain dynamics and external fields limits the possibility of creating materials with specified characteristics for operation in extreme conditions.

The aim of the work is to create an open software platform for multi-method modeling and comparative analysis of hysteresis phenomena in ferroelectrics, which allows for a quantitative assessment of the influence of crystal lattice defects, temperature conditions and heat treatment modes on the polarization dynamics.

Experimental part

For numerical modeling, a multi-method approach was used that combines the following: a modified Landau-Khalatnikov equation; the Monte Carlo method for a 2D spin lattice (L×L); the Giles-Atherton model, separating the polarization into reversible (Prev) and irreversible (Pirr) terms, and the annealing model with parameterization of unipolarity k.

In the developed FerroSim program, this phenomenon is modeled by a modified Landau-Khalatnikov equation:

$$\frac{dP}{dt} = -\Gamma\left(\alpha P + \beta P^3 + \gamma P^5 - (E + E_{int})\right), \quad (1)$$

where E_{int} simulates local fields from defects. For example, at E_{int}=2.0 V/m, the coercive field increases by 25%, which is consistent with experiments for $BaTiO_3$ with Cr^{3+} impurities [2]. The polarization asymmetry caused by the unipolarity k is modeled in FerroSim by adding the parameter k to the equation:

$$P_{obs} = P + k, \quad (2)$$

where k depends on the annealing mode. For homogeneous samples:

$$k = k_0 \cdot e^{-n\cdot\lambda}, \quad (3)$$

and for inhomogeneous samples — by a gradient change in k.

Barium titanate ($BaTiO_3$), a model ferroelectric with pronounced nonlinear properties and a typical hysteresis loop, was chosen as the object of study. The hysteresis loop of ferroelectrics such as barium titanate describes the dependence of the polarization P on the external electric field E. In an ideal crystal lattice, the loop is symmetrical with respect to the P and E axes. However, in real conditions, its shape and position depend significantly on structural defects, temperature effects, and heat treatment. The presence of internal fields E_{int} caused by defects (oxygen vacancies, impurity ions) leads to a shift of the loop along the E axis.

Results and discussions

Modeling in FerroSim revealed:

1. For homogeneous samples: decrease in k by 50% after 10 annealing cycles (λ=0.3); decrease in residual polarization Pr from 0.28 to 0.15 C/m²;
2. For inhomogeneous samples: stabilization of k after 3 cycles (Δk=0.05); preservation of loop asymmetry due to fixed impurity gradients.

The developed FerroSim application is a universal platform that combines theoretical models and practical tools for studying ferroelectrics. Its key feature is the integration of four methods - from the classical Landau-Khalatnikov approach to statistical modeling using the Monte Carlo method, which allows researchers to analyze hysteresis phenomena within a single interface. The flexibility of setting parameters such as internal fields (E_{int}), temperature (T_c) and unipolarity (k) is combined with an intuitive graphical interface developed in PyQt5 [3,4]. This allows for quick comparison of results from different methods without the need to switch between separate programs. The FerroSim interface (Figure 1) includes specialized tabs for each algorithm, a progress bar for tracking calculations, and Matplotlib-based illustrations that support customization of plot styles in real time [4].

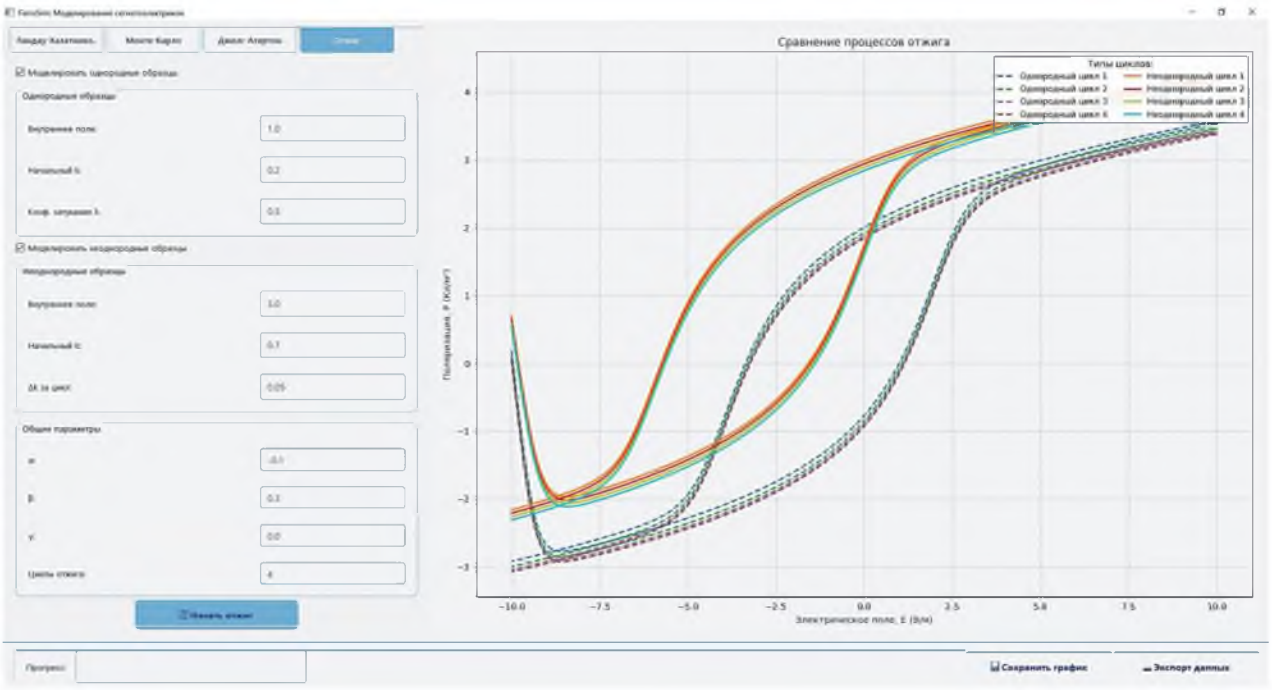

Figure 1.Interface of the FerroSim software

Thanks to the implementation of multithreading (QThread), resource-intensive calculations, such as modeling large spin lattices, are performed in the background, without blocking interaction with the program. To ensure reproducibility of results, data export functions are provided: hysteresis loops are saved in vector and raster formats (PDF, SVG, PNG) for publications, and calculated data are saved in CSV for further analysis in third-party packages. Open source code (GPLv3) and built-in examples of settings for barium titanate and other materials make FerroSim not only a calculation tool, but also a platform for training and developing new models. Data results export functions are provided: hysteresis loops are saved in vector and raster formats.

The experimentally obtained hysteresis loops for homogeneous samples had a standard form and were well approximated by the expression :

$$P = \pm P_s \,\mathrm{th}\left(\frac{\pm E - E_c}{2\delta}\right) \pm P_r(1-a)\ , \qquad (4)$$

where Ps is the saturation polarization; Pr is the residual polarization; Ec is the coercive field (the upper sign corresponds to the ascending branch of the loop, the lower sign to the descending branch).

It can be noted that the calculations performed showed, for example, that the difference in hysteresis loops for gradient samples with different composition configurations (x = 0–0.3 and x = 0–0.3–0) was insignificant (Figure 2). It should be especially noted that no shift in the hysteresis loops (either along the P axis or along the E axis) was detected in the materials studied.

There are various theoretical approaches to describing the features of the polarization behavior of gradient (multilayer) ferroelectric structures. In most of them, these features (in particular, the shift of the hysteresis loop along the P axis) were associated with the spatial dependence of the polarization P(z). Thus, in a series of works by J.V. Mantese and S.P. Alpay (summarized in the monograph [5]), it was shown that the magnitude of the loop shift is due to the dependence on the coordinate of the thermodynamic coefficients and taking into account the gradient term ~ (dP/dz)2 in the Landau–Ginzburg–Devonshire expansion for the free energy.

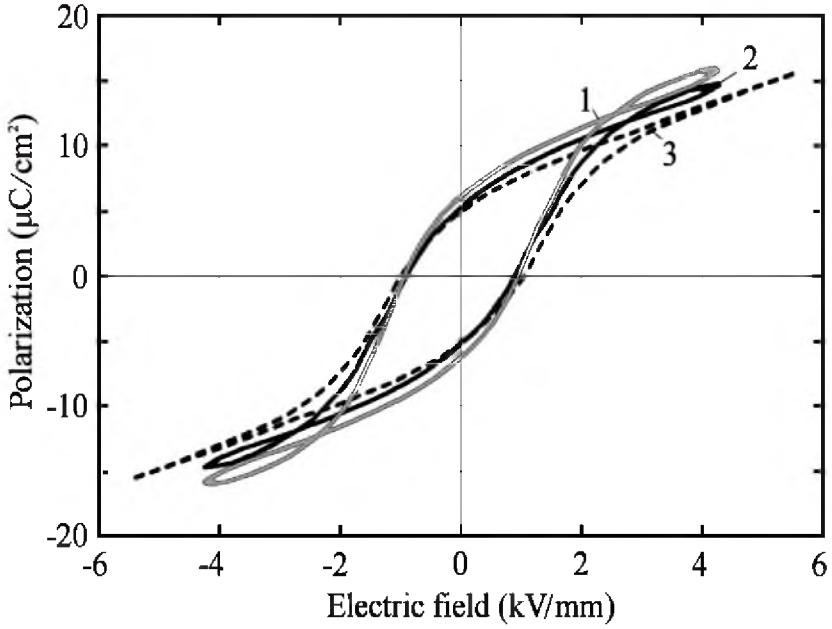

Figure 2. Hysteresis loops of gradient BST ceramic samples:
1) x = 0–0.3–0; 2) x = 0–0.3; 3) calculation loop for x = 0–0.3

In the future, it is planned to take into account the specified theoretical calculations for modeling the processes of repolarization of thin-film gradient ferroelectric materials.

Conclusion

As a result, a multifunctional software platform FerroSim was developed, integrating four methods for modeling the hysteresis of ferroelectric materials. It was found that internal fields and unipolarity significantly affect the asymmetry and shift of the hysteresis loop, and heat treatment reduces k in homogeneous samples, leading to symmetrization. The developed platform, due to the support of a multi-method approach and an open architecture, can be adapted for studying a wide class of ferroelectrics and multiferroics. FerroSim can serve as a tool for fundamental research and a platform for engineering design of materials with controlled hysteresis characteristics.

References:

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