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Analysis and computer implementation of the mathematical model of 180° domain structures formation in ferroelectrics

The study is devoted to the theoretical analysis and numerical implementation of the 2D mathematical model of 180° ferroelectric domain structures formation within the framework of the Landau – Ginzburg – Devonshire thermodynamic approach supplemented by the Landau – Khalatnikov equation to express the polarization dynamics. The mathematical problem statement is formalized as an initial-boundary value problem for semilinear parabolic partial differential equation. A finite element implementation of the model is performed with the use of COMSOL Multiphysics platform. A series of computational experiments were conducted to visualize various configurations of ferroelectric domain structures.

Key words: *reaction-diffusion system, Landau – Ginzburg – Devonshire – Khalatnikov model, finite elements method, ferroelectric domain structure.*

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Introduction

In general, reaction-diffusion models are applied to explore space-time structures in nonlinear media arising in various subject areas such as ecology, medicine, biology, chemistry, physics, etc. [1]. The study the patterns formation in living organisms and in non-living objects is one of the actual problems of mathematical modelling of a wide class of systems.

In physics, the thermodynamic Landau – Ginzburg theory is used to describe a large class of bifurcations and nonlinear problems in spatially extended systems. The Landau – Ginzburg approach has been used to analyze the properties of ferroelectrics, model ferroelectric polarization switching and hysteresis loops (see [2–6] and references therein).

Mathematically, the spatial-temporal distribution of polarization can be described on the basis of the Landau – Ginsburg – Devonshire – Khalatnikov thermodynamic model in the formulation of an initial-boundary value problem for a reaction-diffusion partial

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differential equation [5–9]. Visualization of the polarization distribution is attributed to the imaging domain structures in ferroelectrics or ferroelectric patterns.

In spite of the various number of studies that are devoted to mathematical problems for the general cubic-quintic time-dependent Ginzburg – Landau equation, theoretical analysis of the Landau – Khalatnikov model awaits further investigations to develop the mathematical basis for the thermodynamic theory of ferroelectricity. In our previous study [8], theoretical and numerical analysis of the initial-boundary value problem for the generalized 1D Landau – Khalatnikov model was performed. The existence and uniqueness of the solution are proved. The present study is a continuation of that work and focuses on the model justification as well as modelling the ferroelectric domain patterns in the framework of the Landau – Ginzburg – Devonshire – Khalatnikov model using tools of the COMSOL Multiphysics software.

1 Governing equations

By the above, we assume that the mathematical model of ferroelectric domain pattern formation can be formalized with the use of the thermodynamic Landau – Ginzburg – Devonshire theory. In addition, we will take into account the Landau – Khalatnikov approach to theoretically describe nonstationary polarization changes. By classification, this model can be referred to as deterministic and expressed using differential equations. For ferroelectrics with 180° domain structures, polarization reorientation is realized along one of its components. Hence, the mathematical model is governed by the initial-boundary value problem for a time-dependent cubic-quintic Landau – Khalatnikov equation:

$$\frac{\partial P}{\partial t} = D\Delta P + aP + bP^3 - cP^5 + E, \quad 0 < x < L, \ 0 < y < L, \ 0 < t \le t_b, \tag{1}$$

$$P|_{t=0} = P_0(x, y), \quad 0 < x < L, \ 0 < y < L, \tag{2}$$

$$q\frac{\partial P}{\partial \mathbf{n}}|_{\Gamma} + wP|_{\Gamma} = g, \quad 0 < t \le t_b, \tag{3}$$

where $P(x, y, t)$ is the space-time distribution of the polarization; Γ is the boundary of the solution domain that is a square with a linear size of L; D is the positive thermodynamic parameter; E is the applied field; a, b, c, q, w, g are the thermodynamic constants; t_b is the observation time.

The thermodynamic constant a is positive for all known ferroelectrics [3]; $c > 0$; $b > 0$ for ferroelectrics with the first order phase transition and $b < 0$ for ferroelectrics with the second order phase transition (in this case the model equation can be reduced to cubic partial differential equation). Generally, the applied field E is defined as a temporal periodical function in modelling of induced polarization switching and set to be zero in modelling of initial ferroelectric domain state. Notably, to perform simulations, the particular expressions of boundary conditions are determined due to specific electrical conditions of physical experiments.

2 The brief theoretical foundations for the analysis of the model

Following [8], where a similar one-dimensional model is considered, we reduce the problem (1)–(3) to the Cauchy problem for an equation with an operator coefficient.

Let $\Omega = (0, L) \times (0, L)$, $Q = \Omega \times (0, T)$. By L^p, $1 \le p \le \infty$, we denote the Lebesgue space, and by H^s the Sobolev space W_2^s.

The space $L^s(0, T; X)$ (respectively $C([0, T]; X)$) consists of class functions L^s, $s \ge 1$, defined on $(0, T)$ (respectively continuous on $[0, T]$) with values in a Banach space X. Let $H = L^2(\Omega)$, $V = H^1(\Omega)$. By V' we denote the dual space of V, $V \subset H = H' \subset V'$.

We define the operator $A\colon V \to V'$ and the functional $g_b \in V'$ so that

$$(AP, v) = D \int\limits_{\Omega} \nabla P \nabla v \, dxdy + \frac{Dw}{q} \int\limits_{\Gamma} Pv \, d\Gamma, \quad (g_b, v) = \frac{D}{q} \int\limits_{\Gamma} gv \, d\Gamma \quad \forall P, v \in V.$$

The initial-boundary value problem (1)–(3) is written in the standard way in the following form. Find $P \in L^2(0, T; V)$,

$$P' + AP = aP + bP^3 - cP^5 \text{ a.e. on } (0, T), \quad P|_{t=0} = P_0. \tag{4}$$

The following result is valid, which is proved similarly to the case of an initial-boundary value problem with homogeneous Dirichlet conditions [10, Th. 1.1].

Theorem 1. *For $P_0 \in H$, there exists a unique solution P of (4) which satisfies the following conditions: $P \in C([0, T]; H) \cap L^6(Q)$, $P' \in L^2(0, T; V') + L^{6/5}(Q)$.*

3 Computational experiments and discussion

In order to perform computer modelling, we apply the COMSOL Multiphysics software (license agreement No 20/15/230). First, we demonstrate the dynamics of formation of a 180° natural domain structure from zero polar orientation at the start moment. To conduct simulations, we initialize a set of model parameters using arbitrary units: $L = 40$, $D = 1$, $P_0 = 0$, the thermodynamic constants a, b, c are set equal to unity. Here we define zero flux of polarization at $y = 0$, $y = L$ for $0 < x < L$ and $P|_{x=0} = -P_s$, $P|_{x=L} = P_s$, for $0 < y < L$, where $P_s = 1$ is the spontaneous polarization. By model construction, the positive value of polarization corresponds to the "head up" direction $\uparrow \mathbf{P}$, whereas the negative value means "head down" direction of polarization $\downarrow \mathbf{P}$.

Figure 1 shows two frames of a 180° two-domain structure formation. One of the key questions in mathematical modelling of ferroelectric domain patterns is the setting of initial and boundary conditions due to their influence on the simulation results, especially in change-of-gradient region. To be precise, we have a quasi 2D model with the polarization changes only along the coordinate x due to the Dirichlet conditions at the boundaries of the sample $x = 0$, $x = L$ as defined above. For $y = 0$ and $y = L$ at $0 < x < L$, the application of the zero-flux boundary conditions as well as Robin boundary conditions $\partial P/\partial \mathbf{n} = -P/\lambda$, where $\lambda = 1$, leads to the similar results. The simulated space-time distribution of polarization is presented in Figure 2. This observation suggests that the physical system comes to the equilibrium state very quickly compared to the Fourier time $t_{F_o} \approx 10^3$ for the given experimental conditions. This time is estimated to be $t \approx 20$.

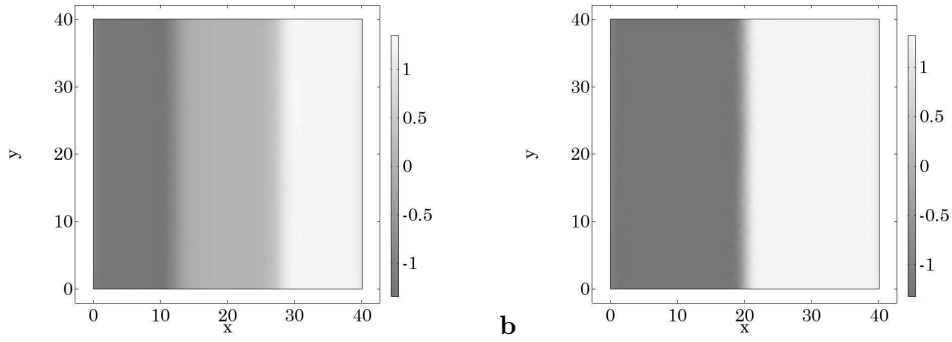

Fig. 1: The successive stages of 180° two-domain pattern formation at moments $t = 7$ — a and $t = 20$ — b.

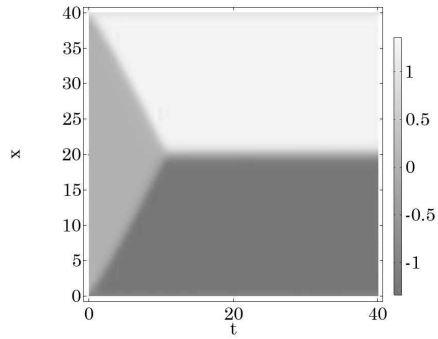

Fig. 2: The space-time distribution of polarization at $y = L/2$.

Further computations illustrate the main peculiarities of 180° multi-domain pattern formation. Zero value of initial polarization can be associated with the artificial creation of a non-polarized state of ferroelectric stimulated by an applied external field. This implies that it would be more appropriate for a natural state of domain structures to specify the initial condition with a random distribution of polarization. We initialize $P_0(x, y)$ as the uniform random distribution ranging from 0 to 1 with a zero mean. Here we set zero-flux boundary conditions for all boundaries of the solution domain.

In Figure 3 we visualize the sequential stages of 180° domain pattern dynamics for fixed times. Figure 3 d corresponds to the image of domain boundaries due to calculation of the the absolute value of polarization (the similar way is usually used in scanning electron microscopy and atomic force microscopy). We can observe unstable multi-domain pattern of ferroelectric sample. The system relaxes after a time $t \approx 200$ to a single-domain state. The application of the Robin boundary conditions or zero-flux boundary conditions leads to similar results. Note also that obtained simulation data refer to uniaxial ferroelectric crystals with first-order phase transitions (e.g., lithium niobate, lithium tantalate) [11]. The similar types of 180° ferroelectric domain patterns can be simulated for ferroelectrics with the second-order of phase transitions.

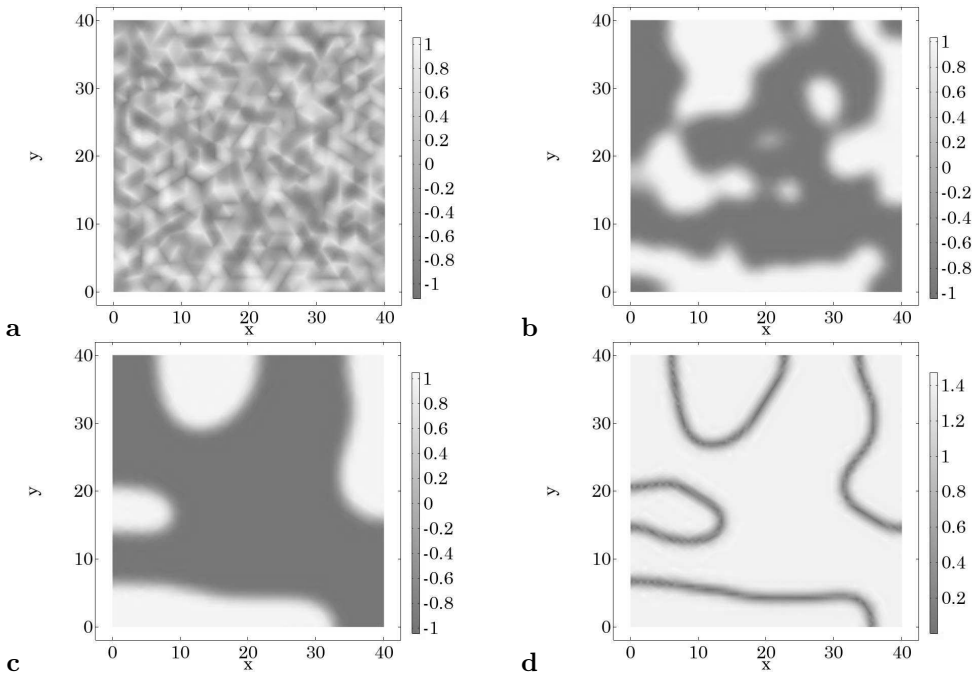

Fig. 3: The sequential stages of 180° multi-domain pattern formation at moments $t = 0$ — a, $t = 3$ — b, $t = 20$ — c and visualization of domain boundaries $t = 20$ — d.

Conclusion

Thus, the study has contributed to the development of mathematical basis for 2D thermodynamics model of 180° domain pattern formation in ferroelectrics. The model formalized as an initial-boundary value problem for time-dependent cubic-quintic Landau – Khalatnikov equation is examined from the theoretical and the numerical points of view. The existence and uniqueness of a weak solution are discussed. Finite element implementation of the model with COMSOL Multiphysics software allowed us to establish the features of the development of various domain patterns depending on the setting of boundary and initial conditions.

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АННОТАЦИЯ

Работа посвящена теоретическому анализу и численной реализации 2D модели формирования 180° сегнетоэлектрических доменных структур в рамках термодинамического подхода Ландау – Гинзбурга – Девоншира, дополненного уравнением Ландау – Халатникова для выражения динамики поляризации. Модель формализована в виде начально-краевой задачи для полулинейного параболического уравнения в частных производных. Обсуждается вопрос существования и единственности слабого решения. Конечно-элементная реализация модели выполнена с использованием платформы COMSOL Multiphysics. Проведена серия вычислительных экспериментов для визуализации различных конфигураций сегнетоэлектрических доменных структур.

Ключевые слова: *реакционно-диффузионная система, модель Ландау – Гинзбурга – Девоншира – Халатникова, метод конечных элементов, сегнетоэлектрическая доменная структура.*