Computational Techniques for Modeling Time-Fractional Dynamics of Polarization Switching in Ferroelectrics ⋆ ⋆⋆

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Abstract. Ferroelectric polarization switching under external exposure is a complex phenomenon, the study of which can be enhanced by the application of mathematical modeling and computer simulation techniques. The paper extends the Landau–Ginzburg–Devonshire–Khalatnikov approach to simulate polarization switching in ferroelectrics as materials exhibiting time memory effects. The proposed mathematical model is expressed by a time-fractional semilinear partial differential equation. An implicit iterative finite difference scheme based on an approximation of the Caputo fractional derivative was constructed and then implemented in Matlab. The computational performance of derived algorithm is provided by a numerical analysis of a test-problem solution and demonstrated by results of computer simulations of polarization hysteresis in ferroelectrics under applied periodical field.

Keywords: Landau–Ginzburg–Devonshire–Khalatnikov model · Ferroelectric polarization switching · Time-fractional partial differential equation · Caputo fractional derivative · Implicit finite difference scheme.

1 Introduction

In recent years, numerous studies have been extensively conducted to explore physical systems, indicating irregularity, spatial scale invariance, self-similar behavior, hereditary properties and time memory effects. One of the theoretical frameworks describing complex physical phenomena is given by the fractional differential theory. Fractional differential equations can be used for the mathematical modeling of time-dependent responses in complex-structured media under non-equilibrium conditions. Such dynamics is also referred to as non-classical or anomalous processes [1, 3, 29]. In the present study, we consider ferroelectric materials as model objects examined with an application of the apparatus of fractional differential equations.

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Ferroelectrics are classified as a subclass of promising polar dielectrics, which have a wide range of applications in microelectronics, acoustics, radio engineering, optoelectronics, piezo- and pyrotechnics, etc. The most relevant applications of ferroelectrics in science and technology are associated with general mechanisms of polarization switching and domain structure dynamics stimulated by an external exposure [22, 31].

Significant attentions have been given over the years to theoretical investigations and experimental justifications of the domain boundaries moving, restructuring of domain configurations and kinetics of polarization switching in ferroelectrics. For instance, the Kolmogorov–Avrami model has been developed to model the polarization reversal processes in ferroelectrics based on statistical approach [20]. Also, the non-Kolmogorov–Avrami model has been proposed to simulate ferroelectric domain structure dynamics in polycrystalline disordered thin films [28]. A modification of the Kolmogorov–Avrami model applied to polarization reversal process stimulated by electron irradiation has been designed in our previous works [13, 18]. A wide spectrum of models constructed with approximation techniques have been reported in different studies, in particular, the numerically stable Preisach model [4].

For this study, it is of interest to model of polarization switching process in view of the importance that ferroelectrics possess self-similar domain structures and exhibit time memory effects in the process of registration of dynamic responses. Among the various models, we can emphasize models based on fractional differential approach used for description of polarization switching processes. Specifically, the study [17] has been reported a fractional differential modification of the Kolmogorov–Avrami model. Also, the fractional differential analogue of the model in the injection mode has been proposed in [18]. An approximation model of polarization-electric field hysteresis dependence has been presented in studies [10, 17] using computation of time-fractional derivative of polarization. In all these studies, fractional-differential models of complex domain dynamics and polarization switching have been considered, which are justified by indicating time memory effects, fractal properties and self-similar behavior of ferroelectrics. Furthermore, the simulation results based on fractional differential models have demonstrated a better agreement with the experimental data than the integer analogs.

In addition, a fundamental phenomenological theory of the Landau–Ginzburg–Devonshire can be applied for mathematical simulation of polarization switching processes in ferroelectrics [22, 26, 30]. Moreover, the Landau–Khalatnikov equation allows one to examine ferroelectric state and polarization dynamic behavior under external field [24]. The our previous study has been done to investigate the Landau–Ginzburg–Devonshire–Khalatnikov model from both theoretical and computational points of view [14]. Mathematically the generalized Landau–Khalatnikov model is governed by an initial-boundary value problem for a time-dependent nonlinear partial differential equation of reaction-diffusion type. To give some idea of the benefits of this approach the current study assumes the fractional differential modification of generalized Landau–Khalatnikov

model to describe time memory effects arising in ferroelectrics during polarization switching.

The mathematical formulation of this model represents a whole class of initial-boundary value problems for time-dependent nonlinear fractional partial differential equations of the reaction-diffusion type. As far as analytical methods can be applied for a sufficiently limited range of problems, numerical methods play a crucial role in mathematical modeling and computer simulation of anomalous reaction-diffusion systems. In particular, to solve numerically these problems, one can use computational schemes based on the finite difference approach [2, 5, 7, 12, 15, 16, 19, 21, 23, 25, 27, 32]. The application of finite difference schemes for solving fractional differential equations have some special features, which are attributed to providing a satisfactory approximation order and reducing rather intensive computations costs. This primarily depends on the chosen definition of the fractional derivative and on the method of approximation. In practice, the definitions of Grünwald–Letnikov [15, 16, 23] and Caputo [12, 23] are widely used. Therefore the construction and implementation of effective numerical schemes is particularly important and decisive.

Hence, the present study was undertaken to develop a model of polarization switching in ferroelectrics by means of constructing a time-fractional modification of the generalized Landau–Khalatnikov equation and a finite difference scheme for further computer simulations.

2 Mathematical Model

The phenomenological Landau–Ginzburg–Devonshire theory of ferroelectricity enables one to examine the relation between the polarization P and the external electric field E [22, 30]. In addition, the Landau–Khalatnikov approach can be used to formulate a time-dependent model of polarization dynamics in ferroelectrics. We will therefore apply the generalized Landau–Khalatnikov equation based on the Landau–Ginzburg–Devonshire–Khalatnikov theory to modeling polarization switching processes in ferroelectrics taking into account time memory effects. The fundamental thermodynamic model is described by a semilinear parabolic partial differential equation. By inductive assumption, we can introduce a time-fractional modification of the Landau–Khalatnikov equation to model complex polarization dynamics and polarization switching process.

To be definite, we also assume that an uniaxial ferroelectric crystal is under consideration and the polarization P is the order parameter describing its state. This implies that polarization P depends on one space variable x and sample surfaces are associated with the coordinates $x = 0$ and $x = L$ as illustrated in Fig. 1.

Also, suppose that the polarization reversal process in a ferroelectric crystal is realized due to the application of the sinusoidal electric field. Notice that the polarization and the intensity of applied electric field are vector quantities. In these terms, only two feasible states for polarization $\uparrow \mathbf{P}$ and $\downarrow \mathbf{P}$ are simulated

Fig. 1. The scheme of the 180° polarization reversal process in a ferroelectric uniaxial crystal and corresponding polarization states for the maximum value of applied field (a) and after the complete 180° switching event (b).

after the whole 180° switching event related to the orientation of applied field $\uparrow \mathbf{E}$ and $\downarrow \mathbf{E}$ respectively.

Whence, we can arrive at the mathematical problem statement described by an initial-boundary value problem for a time-fractional semilinear parabolic partial differential equation:

$$\frac{\partial^\alpha P}{\partial t^\alpha} = D\frac{\partial^2 P}{\partial x^2} + aP + bP^3 - cP^5 + E, \;\; 0 < x < L, \;\; 0 < t \le \frac{T}{T^*}, \tag{1}$$

$$P|_{t=0} = 0, \;\; 0 \le x \le L, \tag{2}$$

$$\left.\frac{\partial P}{\partial x}\right|_{x=0} = \frac{P}{\lambda}, \;\; \left.\frac{\partial P}{\partial x}\right|_{x=L} = -\frac{P}{\lambda}, \;\; 0 \le t \le \frac{T}{T^*}, \tag{3}$$

where α is the order of the time-fractional Caputo derivative (defined below), $\alpha \in (0,1)$; t is the dimensionless time; T^* is the characteristic time in s; T is the observation time in s; $P = P(x,t)$ is the polarization distribution in $\mathrm{C/m^2}$; $D = \delta k T^*$; k is the thermodynamic restoring force in $\mathrm{F/(m \cdot s)}$; δ is the gradient coefficient in $\mathrm{m^3/F}$; $E = \widetilde{E} k T^*$; $\widetilde{E}{=}E_0 \sin(\omega t)$ is the electric field applied along the polar axis in V/m; $a = -\widetilde{a} k T^*$, $b = -\widetilde{b} k T^*$ and $c = \widetilde{c} k T^*$; $\widetilde{a}$ in m/F, $\widetilde{b}$ in $\mathrm{m^5/(C^2 \cdot F)}$, $\widetilde{c}$ in $\mathrm{m^9/(C^4 \cdot F)}$ are the thermodynamic constants; λ is the extrapolation length in m.

By construction, note that the characteristic time T^* is introduced to the model to adjust the dimensions of time and distance (see, e.g., [19]). In addition, we consider the model of polarization reversal process in ferroelectrics with first order phase transitions. In the case of ferroelectrics with the second order phase transition, equation (1) can be reduced to a more simple form specified as a cubic time-fractional partial differential equation with parameter $b < 0$.

Since the proposed model is expressed by a nonlinear fractional-differential equation, the design of numerical schemes for such problems is of particular importance.

3 Outline of Algorithm

The accuracy of numerical solutions of fractional differential equations is directly related to the definition of the fractional derivative and the method of its approximation. A fractional derivative in contrast to a classical one, does not have an unambiguous definition. In our case, we consider the Caputo time-fractional derivative [23], which is defined as follows:

$$D_C^{\alpha} f(t) = \frac{1}{\Gamma(n-\alpha)} \int_0^t (t-\xi)^{n-\alpha-1} f^{(n)}(\xi) d\xi, \quad n-1 < \alpha \leq n, \tag{4}$$

where Γ is the Gamma function.

In recent decades, the theory of fractional differential problems has stimulated many investigations. Among the applications a special place is occupied by the numerical approaches for solving diffusion-type equations [2, 5, 12, 15, 16, 21, 25, 27, 32]. Numerous studies have been devoted to numerical solutions of the anomalous diffusion equation with a time-fractional derivative [2, 5, 12, 21, 25, 32] or with space-fractional derivatives [15, 16, 21, 27] or both of them [21]. In these studies numerical solutions of diffusion problems have been obtained using the definitions of Riemann–Liouville, Caputo, Grünwald–Letnikov, Ritz, etc.

The order of the approximation of fractional derivatives in most of the reported studies are less than the second. More accurate approximations have been derived for advection-diffusion equations (see [2, 12]) and time-fractional subdiffusion equation [5], as well as by constructing an algorithm for the numerical solution of the diffusion-wave equation [7].

In order to obtain a numerical solution of the problem (1)–(3) we derive a computational scheme based on the Caputo definition of fractional derivative (4), an implicit finite difference method and an iterative procedure. Here we apply the approach to the finite difference approximation of fractional derivative proposed in study [12], in which numerical solution of the advection-diffusion equation with a time-fractional derivative has been found, and also stability and convergence of the obtained difference scheme have been investigated.

Hence, for the Caputo fractional derivative of order $0 < \alpha < 1$ we can introduce an approximation on a finite-difference grid $\Omega^{\tau} = \left\{t^k = k\tau,\, k = \overline{0, N}\right\}$:

$$\frac{d^{\alpha} f\left(t^k\right)}{dt^{\alpha}} = \frac{\tau^{-\alpha}}{\Gamma(3-\alpha)} \sum_{j=0}^{k-1} \left[w^{k-j}\left(f^{j+1} - f^{j-1}\right) + s^{k-j}\left(f^{j+1} - 2f^{j} + f^{j-1}\right)\right] +$$

$$+ O\left(\tau^{3-\alpha}\right), \quad k = \overline{1, N}, \tag{5}$$

where τ is the time step; $f(t)$ is a differentiable function; the weight functions w and s are given as follows:

$$w^{k-j} = \frac{2-\alpha}{2}\left[(k-j)^{1-\alpha} - (k-j-1)^{1-\alpha}\right],$$

$$s^{k-j} = (k-j)^{2-\alpha} - (k-j-1)^{2-\alpha} - (2-\alpha)(k-j-1)^{1-\alpha}.$$

The fractional derivative (5) is approximated with the order of $O(\tau^{3-\alpha})$. However, if $j = 0$ in (5), we will have the function value f^{-1} at the dummy node, which is defined outside of the computational interval. We can approximate this value by following $f^{-1} = f(0) + O(\tau)$. As a result, the general scheme (called also $L1$-2 formula [8, 12]) is much more effective and more accurate than the $L1$ formula, which is derived using direct finite-difference approximation of the Caputo derivative [6].

Using the idea of constructing an implicit finite-difference scheme in conjunction with an approximation of the Caputo time-fractional derivative [12], and a finite difference approximation of the second-order space derivative, we can formulate a finite-difference analogue for the differential equation (1) on the space-time grid $\Omega_h^\tau = \{x_i = ih,\, i = \overline{0, M},\, t^k = k\tau,\, k = \overline{0, N}\}$:

$$\frac{\tau^{-\alpha}}{\Gamma(3-\alpha)} \sum_{j=0}^{k-1} \left[w^{k-j} \left(P_i^{j+1} - P_i^{j-1} \right) + s^{k-j} \left(P_i^{j+1} - 2P_i^j + P_i^{j-1} \right) \right] =$$

$$= \frac{D}{h^2} \left(P_{i+1}^k - 2P_i^k + P_{i-1}^k \right) + aP_i^k + b(P_i^k)^3 - c(P_i^k)^5 + E^k, \tag{6}$$

where $k = \overline{1, N}$, $i = \overline{1, M-1}$.

Since we solve the cubic-quintic partial differential equation, we arrive at a system of nonlinear difference equations on each time layer. In this way we can use of an iterative procedure, which will allow us to solve a system of linear algebraic equations. So that we form a sequence of approximations $P_i^{(q)}$ converging to P_i^k for each time moment t^k, $k = \overline{1, N}$, using the relations: $(P_i^{(s)})^3 \approx (P_i^{(q-1)})^2 P_i^{(q)}$, $(P_i^{(q)})^5 \approx (P_i^{(q-1)})^4 P_i^{(q)}$, where $q = 1, 2, ...$ is the number of iteration.

The iterative algorithm at the k time step starts by estimating an initial value of polarization using the value from the previous time step, that is $P_i^{(0)} = P_i^{k-1}$. The combination of finite difference schemes with an iterative procedure enables one to solve applied problems without losing the accuracy of the general computational scheme (see, e.g., [9, 11, 14]).

Also, we use the initial condition $P_i^0 = 0$, $i = \overline{0, M}$ and the asymmetric finite difference approximation for Robin boundary conditions (3) for $k = \overline{0, N}$:

$$\frac{-3P_0^k + 4P_1^k - P_2^k}{2h} = \frac{P_0^k}{\lambda}, \quad \frac{3P_M^k - 4P_{M-1}^k + P_{M-2}^k}{2h} = -\frac{P_M^k}{\lambda}. \tag{7}$$

The general system of algebraic equations is solved by the Gauss method, which guarantees solving the problem with the error determined by the accuracy of computations. The design of the computational algorithm emphasis the relation between the applied framework of fractional derivatives and the formalized long time memory physical process. The polarization value at a given time moment depends not only on the behavior of the polarization in the vicinity of this point, but also on the values from the entire range of the temporal variable. Hence we proposed the implicit iterative finite difference scheme based

on an approximation for the Caputo fractional derivative to solve semilinear time-fractional partial differential equation. This algorithm was implemented in Matlab software which allows high processing rates working with matrices.

4 Computer Simulation Results

4.1 Test Problem and Numerical Analysis

Let us first consider a simple test problem specified as a time-fractional diffusion equation with imposed initial and boundary conditions:

$$\frac{\partial^{0.85} u}{\partial t^{0.85}} = 0.5\frac{\partial^2 u}{\partial x^2} + \frac{\Gamma(4)}{\Gamma(3.15)}x^3 t^{2.15} - 3xt^3, \;\; 0 < x < 1, \;\; 0 < t \le 1, \tag{8}$$

$$u|_{t=0} = 0, \;\; 0 \le x \le 1, \tag{9}$$

$$u|_{x=0} = 0, \;\; u|_{x=1} = t^3, \;\; 0 \le t \le 1. \tag{10}$$

This problem has an analytical solution defined as $u(x,t) = x^3 t^3$. In order to verify this solution, one can use a simple relation for calculation of fractional-order derivative of a power function $v = t^p$ [15]:

$$\frac{d^\beta v}{dt^\beta} = \frac{\Gamma(p+1)}{\Gamma(p-\beta+1)}t^{p-\beta}.$$

Figure 2 shows the results of numerical solutions of the problem (8)–(10) in comparison with the analytical solution.

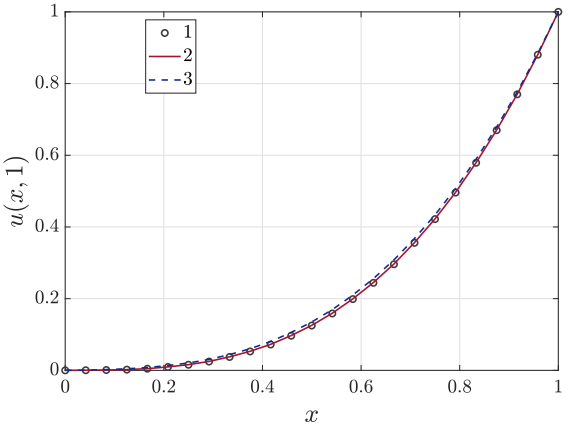

Fig. 2. The analytical – 1 and numerical solutions of the test problem (8)–(10) obtained using the Caputo definition of fractional derivative – 2 and the Grünwald–Letnikov formula – 3 at the time moment $t = 1$ and for space and time steps $h = \tau = 0.125$.

We conducted simulations based on the proposed numerical scheme (6), (7) and using an implicit finite difference scheme based on the Grünwald–Letnikov approximation of the fractional derivative [15, 16]. The latter is one of the most popular computational schemes used in different applications. The order of approximation of this scheme is estimated to be $O(\tau+h^2)$. The graphs are visualized as coordinate profiles of function $u(x,t)$ calculated at the last time moment of observation of the process.

In addition, we performed a numerical analysis of solutions of the problem (8)–(10). The accuracy of the results were estimated using the maximum norm $\delta = \|u - \widetilde{u}\|_\infty / \|u\|_\infty$, where $\widetilde{u}$ is the numerical solution, u is the exact solution calculated for the last time moment $t = 1$. Figure 3 demonstrates the values of the relative errors δ in the double log scale with the variation of the number of nodes M, N along the x and t axes, respectively.

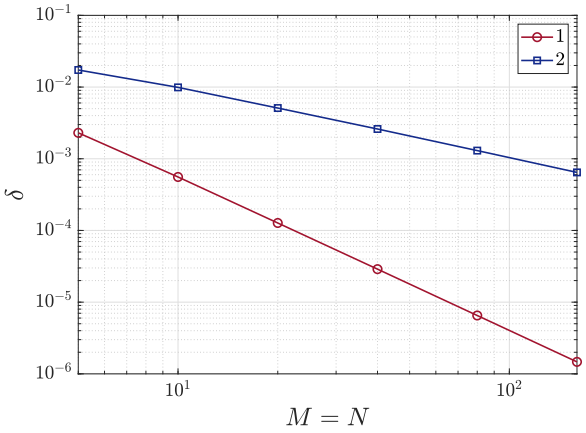

Fig. 3. Estimations of relative errors δ for numerical solutions of the problem (8)–(10) in the log-log scale with the variation of the number of nodes $M = N$: obtained using the Caputo fractional derivative – plot 1 and the Grünwald–Letnikov formula – plot 2.

A comparative analysis of the errors suggests that the described scheme provides a converged solution and an acceptable accuracy. These observations indicate that we can apply the derived computational scheme to get accurate numerical simulation for this class of problems. It should be pointed out that the algorithm can be characterized as resource-intensive and time-consuming. As an example, for number of nodes equal to $N = M = 160$ the accuracy corresponds to $\delta \approx 10^{-6}$ and the computation time approximately equals 2000 s.

4.2 Simulation of Polarization Hysteresis Loop

In this section, we perform numerical simulation of the hysteresis dependence between polarization and applied electric field arising in ferroelectrics. For instance, let us consider the following numerical example as a mathematical model describing a time-space distribution of polarization, which changes with the external sinusoidal electric field:

$$\frac{\partial^\alpha P}{\partial t^\alpha} = \frac{\partial^2 P}{\partial x^2} + 0.05P + 2.5P^3 - 30P^5 + 20\sin 5t, \;\; 0 < x < 1, \;\; 0 < t \le 1.5, \quad (11)$$

$$P|_{t=0} = 0, \;\; 0 \le x \le 1, \quad (12)$$

$$\left.\frac{\partial P}{\partial x}\right|_{x=0} = 100P, \;\; \left.\frac{\partial P}{\partial x}\right|_{x=1} = -100P, \;\; 0 \le t \le 1.5. \quad (13)$$

Here, a set of model parameters are normalized arbitrarily to promote comparability of simulated quantities (in particular, in view of the existence of the $P(E)$ hysteresis dependence) approximately corresponding to ferroelectric crystals with first-order phase transitions.

The result of computer implementation of the model (11)–(13) is presented in Fig. 4. The hysteresis dependencies $P(E)$ are plotted with varying the order of the time-fractional derivative α for values of space step $h = 0.0143$ and time step $\tau = 0.0214$. These findings suggest that a decrease in the order of the time-fractional derivative leads to a narrowing of the ferroelectric hysteresis loop while maintaining its shape. In other words, polarization hysteresis loop has a more narrow shape for crystals with a significant time-memory effect.

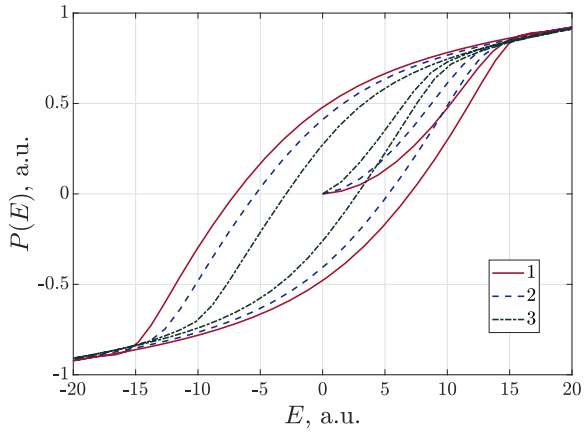

Fig. 4. Ferroelectric hysteresis loops $P(E)$ calculated with variation of the order of the time-fractional derivative: $\alpha = 0.99$ – plot 1; $\alpha = 0.85$ – plot 2; $\alpha = 0.6$ – plot 3.

Thus, the use of a time-fractional derivative in modeling polarization switching process potentially allows one to control the results of computations due to a variation of the order of fractional derivative α or adjust this parameter as a numerical characteristic of the time memory effect in ferroelectrics to provide the better agreement of simulations results with experimental data.

5 Conclusion

In this article we proposed a time-fractional modification of the thermodynamic model of polarization hysteresis in ferroelectrics and numerical scheme for its computer implementation.

The modification of the generalized Landau–Khalatnikov model was described by an initial boundary value problem for a time-fractional semilinear partial differential equation. We derived an implicit iterative finite difference scheme based on an approximation of the Caputo fractional derivative. The combination of finite difference schemes with an iterative procedure allowed applied problems to be solved without losing the accuracy of the general computational scheme. The formalized computational algorithm reveals the key peculiarities of simulation of long time-memory dynamic processes. The polarization value at a given time moment depends not only on the behavior of the polarization in the vicinity of this point, but also on the values from the entire range of the temporal variable. This algorithm was implemented in Matlab software and validated using test-problem.

We performed numerical simulations of the hysteresis dependence between polarization and applied electric field arising in ferroelectrics. Our findings indicate that the use of the fractional differential apparatus provides a more "flexible tool" due to a variation of the order of fractional derivative. This can be used to provide the better agreement of simulations results with experimental data.

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