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Numerical Discretization of Variational Phase Field Model for Phase Transitions in Ferroelectric Thin Films

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Abstract. Phase field methods have been widely used to study phase transitions and polarization switching in ferroelectric thin films. In this paper, we develop an efficient numerical scheme for the variational phase field model based on variational forms of the electrostatic energy and the relaxation dynamics of the polarization vector. The spatial discretization combines the Fourier spectral method with the finite difference method to handle three-dimensional mixed boundary conditions. It allows for an efficient semi-implicit discretization for the time integration of the relaxation dynamics. This method avoids explicitly solving the electrostatic equilibrium equation (a Poisson equation) and eliminates the use of associated Lagrange multipliers. We present several numerical examples including phase transitions and polarization switching processes to demonstrate the effectiveness of the proposed method.

AMS subject classifications: 37N15, 49S05, 65K10, 65N22, 65Z05, 74N99, 78M30

Key words: Ferroelectric, phase field, phase transition, polarization switching, minimum energy path.

1 Introduction

In recent two decades, ferroelectric thin films have been given much attention both theoretically and experimentally [1–7]. These functional materials possess a spontaneous polarization that can be switched between energetically equivalent states in a single crystal by an electric field. Ferroelectric phase transitions and polarization switching depend

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on not only the stability of domain structures but also the electrostatic interactions or the external electric field [8].

Phase field methods have been successfully applied to study a wide range of physical problems, such as nucleation in solid-state phase transformations [9], coarsening process via epitaxial thin film model [10], phase transitions and domain structures in ferroelectric thin films [11], etc. In the study of ferroelectric thin films, they characterize the detailed three-dimensional (3D) domain structures without any *a prior* assumptions with regard to the possible domain structures [12]. Phase field methods are able to predict not only the volume fractions of different orientation domains and the change of domain structures under the effect of applied external conditions, but also the temporal evolution of polarization during a ferroelectric phase transition [13, 14].

In the phase field approach to study the ferroelectric phase transitions, the polarization vector is commonly used to describe a domain structure as the primary phase field variable. The electric potential, another variable, is then used to account for the electrostatic contributions in the phase field model [15]. In the existing literature [13–16], the relation between the two variables is described by the electrostatic equilibrium equation (a Poisson equation), meaning that the electric potential is a function of the polarization vector and can be obtained by solving the electrostatic equilibrium equation when given a polarization distribution. While, the two variables are simply taken as independent ones when calculating the electrostatic driven force for the relaxation dynamics (electrostatic energy variation) of the polarization vector, ignoring their explicit relation given by the electrostatic equilibrium condition. Such treatment may simplify the calculation, but the effect of the electric field on phase transitions and polarization switching is underestimated.

In a recent study [17], new variational phase field formulations were proposed based on a hybrid representation in both real and Fourier variables in order to handle mixed electric boundary conditions (BCs) so that the coupling between the two phase field variables are directly incorporated. Indeed, the variational formulations allow a direct variational calculation of the phase field electrostatic energy and the driving force for the relaxation dynamics of the polarization vector. Furthermore, by utilizing the electrostatic equilibrium relation under the bound charge condition, the calculation of the electrostatic energy and its driving force can be done with respect to the polarization vector alone, thus simplifying the analytical derivations. Such variational forms can precisely and explicitly calculate the electrostatic energy and its corresponding driving force under different common-used electric BCs, e.g., the constant, open circuit, and tip-induced BCs.

In this paper, we develop an efficient numerical scheme for the variational phase field model to discretize the variational phase field formulations for effective 3D numerical simulations. It handles the 3D mixed BCs by combining the Fourier spectral method with the finite difference method for the spatial discretization. This in turn allows for the relaxation dynamics to be solved in a semi-implicit way. In particular, this numerical scheme implements the calculation of the electric potential and the electrostatic driving force as matrix and vector multiplications at the discrete level that avoids explicitly solv-