Three dimensional structures predicted by the modified phase field crystal equation

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Abstract

We present the first numerical results on three dimensional structures predicted by the modified phase field crystal equation. The computations are performed using parallel algorithms based on isogeometric analysis, a generalization of the finite element method. The evolution of crystal structures to their steady equilibrium state is predicted for the various atomic density and temperatures. These steady structures are consistent with the structure diagram predicted earlier using one-mode approximation of analytical solutions of the phase-field-type equations.

Keywords: Phase field, crystal, isogeometric analysis, computer simulation

The phase field crystal (PFC) model has been proposed to incorporate the physics naturally embedded on atomic length scales (elasticity, dislocation, etc.) and on diffusive time scales [1, 2]. The PFC model describes a field that is related to the local atomic number density, such that it is spatially periodic in the solid and constant in the liquid.

Originally formulated in a parabolic form for description of pure dissipative dynamics, the PFC model has also been extended to include faster degrees of freedom consistent with inertia due to propagative regimes of phase transformation. In particular, a hyperbolic or modified PFC model was introduced which includes an inertial term, and thus allows for the description of both fast and slow processes in phase transformations [3, 4, 5, 6, 7, 8, 9]. Although essential progress has been made in modeling of parabolic PFC-equation [10, 11, 12] interpretation of results obtained in modeling of the modified (hyperbolic) PFC-equation required special efforts due to higher time order of the equation [7, 8, 9]. Moreover, physically reasonable and practically important

results of this equation are currently absent for the case of three spatial dimensions. To the author's knowledge, only mathematical analysis on stability and existing of attractors in solutions of the three dimensional modified PFC-equation were made [13, 14, 15]. In the numerical modeling, a transition from two dimensional to three dimensional crystal lattice leads to higher variety of patterns. For these reasons, prime goals of the present article are (*i*) to analyze first results of the numerical solution and (*ii*) to predict the structures modeled by the three dimensional modified PFC-equation. To reach these goals an efficient, parallel numerical algorithm is formulated and the numerical scheme for modeling of patters is developed.

The modified phase field crystal equation describes a continuous atomic density field $\phi(\boldsymbol{x}, t)$ and it is expressed by the following sixth order in space and second order in time equation:

$$\tau \frac{\partial^2 \phi}{\partial t^2} + \frac{\partial \phi}{\partial t} = \nabla^2 \mu, \tag{1}$$

with the chemical potential

$$\mu(\phi) = \frac{\delta \mathcal{F}}{\delta \phi} = \phi^3 + \alpha \phi^2 + (1 - \epsilon)\phi + 2\nabla^2 \phi + \nabla^4 \phi, \tag{2}$$

free energy functional

$$\mathcal{F}[\phi, \nabla\phi, \nabla^2\phi] = \int_\Omega \left[f(\phi) - |\nabla \phi|^2 + \frac{1}{2}(\nabla^2 \phi)^2 \right] \mathrm{d}\Omega \tag{3}$$

and the homogeneous part of the free energy density

$$f(\phi) = \frac{1 - \epsilon}{2}\phi^2 + \frac{\alpha}{3}\phi^3 + \frac{1}{4}\phi^4. \tag{4}$$

Here t is the time, Ω is the computational domain, τ is the relaxation time of the atomic flux to its stationary state, $\epsilon = (T_c - T)/T_c$ is the undercooling, T and T_c are the temperature and critical temperature of transition, α is a coefficient which is a measure of metastability. Equation (1) modifies the traditional parabolic phase field crystal (PFC) equation by including an inertial term $\tau \partial^2 \phi / \partial t^2$ that plays principal role in description of fast phase transitions [5].

In the numerical formulation of the modified PFC equation (1) we use isogeometric analysis (IGA) [16, 17] for the spatial discretization, which allows us to generate the $\mathcal{C}^2$-continuous functions needed for the discretization of the sixth-order spatial derivatives in primal form. We use a weak form of the problem which assumes periodic boundary conditions in all directions. Then, we define a trial solution functional space $\mathcal{X} = \mathcal{X}(\Omega)$, and a weighting function space $\mathcal{Y} = \mathcal{Y}(\Omega)$ which are supposed to be identical. The variational formulation can be stated as follows: Find $\phi \in \mathcal{X}$ such that $\forall w \in \mathcal{Y}$,

$$B(w, \phi) = 0, \tag{5}$$

where

$$B(w,\phi) = \int_\Omega w\left(\tau\frac{\partial^2\phi}{\partial t^2} + \frac{\partial\phi}{\partial t}\right)\mathrm{d}\Omega + \int_\Omega \nabla w\cdot\nabla\phi F''(\phi)\mathrm{d}\Omega$$
$$- \int_\Omega 2\nabla^2 w\nabla^2\phi\mathrm{d}\Omega + \int_\Omega \nabla\left(\nabla^2 w\right)\cdot\nabla\left(\nabla^2\phi\right)\mathrm{d}\Omega. \tag{6}$$

Using the Non-Uniform Rational Bezier Spline-based (NURBS-based) isogeometric analysis, the spatial discretization of the coupled problem is made by finite-dimensional approximations of the functional and weighting spaces. Namely, we define $\mathcal{X}_h$ and $\mathcal{Y}_h$ such that $\mathcal{X}_h \subset \mathcal{X}$ and $\mathcal{Y}_h \subset \mathcal{Y}$. In such case, (5) can be approximated by the following variational problem over the finite element spaces: find $\phi_h \in \mathcal{X}_h$ such that $\forall w_h \in \mathcal{Y}_h$,

$$B(w_h,\phi_h) = 0, \tag{7}$$

where

$$\phi_h(\boldsymbol{x},t) = \sum_{A\in I}\phi_A(t)N_A(\boldsymbol{x}), \quad w_h(\boldsymbol{x}) = \sum_{A\in I} w_A N_A(\boldsymbol{x}). \tag{8}$$

In Eqs. (8), the N_A's are a set of basis functions defined on Ω and I is their global-index set.

Integration in time of the modified PFC equation is made by the generalized α-method proposed by Chung and Hulbert [18] for the equations of structural dynamics. Let us denote $\boldsymbol{\Phi} = \{\phi_A\}$ the vector containing the global degrees of freedom associated to ϕ_h. The first and second time derivatives of $\boldsymbol{\Phi}$ are denoted, respectively, by $\dot{\boldsymbol{\Phi}}$ and $\ddot{\boldsymbol{\Phi}}$. We will also make use of the following residual vector

$$\boldsymbol{R} = \{R_A\}, \quad \text{where} \quad R_A = B(N_A,\phi_h). \tag{9}$$

Then, the time stepping scheme can be defined by the discrete approximation to the global vectors of control variables at time t_n. Namely, defining $\boldsymbol{\Phi}_n$, $\dot{\boldsymbol{\Phi}}_n$, $\ddot{\boldsymbol{\Phi}}_n$ and the current time step $\Delta t = t_{n+1} - t_n$ one gets $\boldsymbol{\Phi}_{n+1}$, $\dot{\boldsymbol{\Phi}}_{n+1}$, $\ddot{\boldsymbol{\Phi}}_{n+1}$ such that

$$\boldsymbol{R}(\boldsymbol{\Phi}_{n+\alpha_f}, \dot{\boldsymbol{\Phi}}_{n+\alpha_f}, \ddot{\boldsymbol{\Phi}}_{n+\alpha_m}) = 0, \tag{10}$$

where

$$\boldsymbol{\Phi}_{n+\alpha_f} = \boldsymbol{\Phi}_n + \alpha_f\left(\boldsymbol{\Phi}_{n+1} - \boldsymbol{\Phi}_n\right), \tag{11a}$$
$$\dot{\boldsymbol{\Phi}}_{n+\alpha_m} = \dot{\boldsymbol{\Phi}}_n + \alpha_m\left(\dot{\boldsymbol{\Phi}}_{n+1} - \dot{\boldsymbol{\Phi}}_n\right). \tag{11b}$$

The solutions (10) are updated using the formulas

$$\dot{\boldsymbol{\Phi}}_{n+1} = \dot{\boldsymbol{\Phi}}_n + \Delta t((1-\gamma)\ddot{\boldsymbol{\Phi}}_n + \gamma\ddot{\boldsymbol{\Phi}}_{n+1}), \tag{12a}$$
$$\boldsymbol{\Phi}_{n+1} = \boldsymbol{\Phi}_n + \Delta t\dot{\boldsymbol{\Phi}}_n + \frac{\Delta t^2}{2}\left((1-2\beta)\ddot{\boldsymbol{\Phi}}_n + 2\beta\ddot{\boldsymbol{\Phi}}_{n+1}\right), \tag{12b}$$

Note that although $\boldsymbol{\Phi}_{n+1}$, $\dot{\boldsymbol{\Phi}}_{n+1}$, $\ddot{\boldsymbol{\Phi}}_{n+1}$ are treated separately in the algorithm, they are not independent because they have to satisfy to Eqs. (12). The above

parameters α_f, α_m, γ and β are selected using accuracy and stability criteria. Note that Eq. (10) constitutes a nonlinear system of algebraic equations at each time step. We linearized this nonlinear system using Newton's method, and solved the resulting linear systems using preconditioned GMRES.

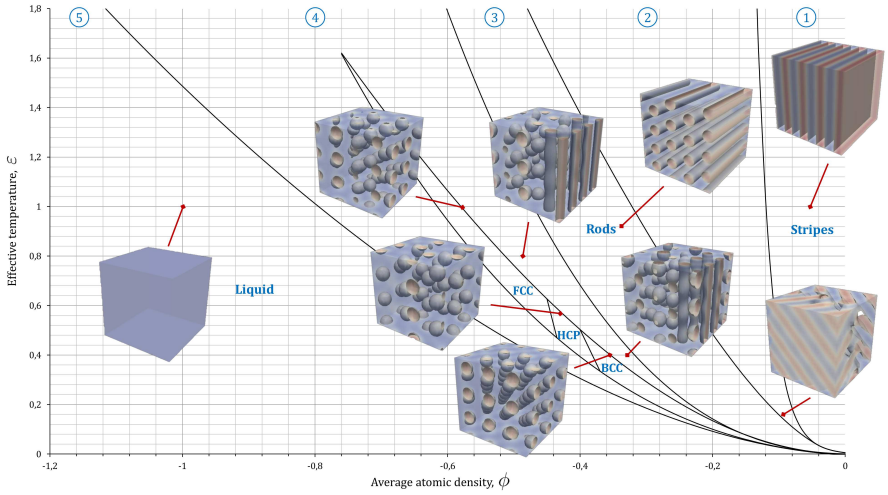

Figure 1: Computed structures in comparison with the regions of diagram of Jaatinen and Ala-Nissila [19]. Regions of existing structures are: (1) Stripes; (2) Stripes and Rods; (3) Rods; (4) mixed structures with the existence of pure body centered cubic (BCC), pure hexagonal close packed (HCP), and pure face centered cubic (FCC) crystalline patterns; (5) homogeneous phase (liquid).

The numerical simulations presented herein have been performed by a C-language code based on a widely used library called PETIGA [20]. This software implements a NURBS-based IGA [17] which can be understood as a NURBS-based Galerkin finite element method (FEM). The computational domain of the simulations was assumed to be the box $\Omega = [0, 50]^3$ and the spatial mesh was composed of 64^3 C^2-quadratic elements. For each crystal pattern modeled by the parallel algorithm we employed 150 processors AMD Opteron (TM) 6212 with 2.6 GHz, which led to five-hour simulations on average. In all cases, the value for parameters α and τ from Eqs. (1)-(4) was $\alpha = 0$ and $\tau = 0.1$. Initial distribution with a small spatial gradient of the atomic density ϕ and periodic boundary conditions have been chosen.

The initial state of the atomic density has been taken for the homogeneous phase (liquid) with the given average value of ϕ quenched by the parameter ϵ into the chosen region of structure diagram. The diagram was previously calculated by Jaatinen and Ala-Nissila [19] for three dimensional structures given by the PFC-equation in one-mode approximation that is shown as solid lines in Fig. 1. As it can be seen in this figure, the present modeling gives the following structures: (1) stripes, (2) mixture of stripes and rods, (3) rods, (4) multiple patterns highly sensitive to values of ϵ and ϕ, (5) liquid. All of

these structures are consistent with the regions of previously predicted structure diagram [19]. This exhibits an efficiency of the presently developed algorithm for the numerical solution of the modified three dimensional PFC-equation. Note that, in our computations we have provided independence of results from the computational grid. However, several important questions are still to be resolved for the modeling of concrete crystal patterns. Among them: (i) influence of the minimal size of computational domain on the type of crystal structure and (ii) effect of order of approximation of the crystal lattice on the crystal symmetry modeled in the complicated region 4 of Fig. 1. Resolving these questions will allow us to carry out practically important numerical simulations.

As a final result, we have predicted the evolution of three dimensional crystal structures to their steady equilibrium state for the various atomic density and temperatures given by the modified PFC-equation. It is known that the evolution given by the modified hyperbolic equation differs from that predicted by the parabolic purely dissipative transition, especially, during the first stages of transformation under consideration. In this case, patterns frozen during their evolution to equilibrium state have a metastable nature [21]. In this work, have clearly shown that the final steady structures given by the numeric solution of the modified (hyperbolic) PFC-equation are consistent with the structure diagram predicted earlier using one-mode approximation of analytical solutions of the traditionally parabolic phase-field-type equations (see Fig. [19]).

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