Influence of computational domain size on the pattern formation of the phase field crystals

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Abstract. Modeling of crystallization process by the phase field crystal method (PFC) represents one of the important directions of modern computational materials science. This method makes it possible to research the formation of stable or metastable crystal structures. In this paper, we study the effect of computational domain size on the crystal pattern formation obtained as a result of computer simulation by the PFC method. In the current report, we show that if the size of a computational domain is changed, the result of modeling may be a structure in metastable phase instead of pure stable state. The authors present a possible theoretical justification for the observed effect and provide explanations on the possible modification of the PFC method to account for this phenomenon.

1. Introduction

The original PFC model has been developed in a parabolic form [1–3]. Then it has been modified to the extended form described by partial differential equation of a hyperbolic type. Such the modified or hyperbolic PFC model (MPFC model) includes faster degrees of freedom in a form of inertia and as such leads to the description of both fast and slow dynamics. Fast front dynamics proceeds when the driving force for the phase transition is large. This occurs when the free energy difference between the (meta)stable periodic solid and initially unstable phase is very large which in general occurs when a system is quenched far below a transition point, or in this case far below the equilibrium temperature of phase transition [4]. These conditions lead to a fast phase transition when the velocity of the front is comparable to the speed of atomic diffusion or the speed of local structural relaxation. The movement of a phase transition front at such fast velocities can lead to bulk phases that are not in a local structural or chemical equilibrium. As a result, the modified (hyperbolic) PFC-model includes the description of both fast and slow dynamics of transition [5–11] by taking into account diffusion and inertial effects [7].

Using the free-energy functional

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

related to the domain Ω, one can define the chemical potential as

$$\mu(\phi) = \frac{\delta \mathcal{F}}{\delta \phi} = \frac{df}{d\phi} + 2\nabla^2 \phi + \nabla^4 \phi, \qquad (2)$$

where ϕ describes conserved order parameter. The free energy density $f(\phi)$ represents the homogeneous part of energy and in the present work it is chosen in the Landau-de Ginnes form:

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

Here, $\epsilon = (T_c - T)/T_c$ is the dimensionless temperature T shows its deviation from T_c critical temperature of transition, and α is the measure of metastability. Finally, one can define the modified (hyperbolic) phase field crystal model, which describes a continuous and conserved field of atomic density $\phi(x,t)$ and it can be expressed by the following equation:

$$\tau \frac{\partial^2 \phi}{\partial t^2} + \frac{\partial \phi}{\partial t} = \nabla^2 \mu, \qquad (4)$$

where t is the time, τ is the relaxation time of the atomic flux to its stationary state. Using the chemical potential (2) with taking into account Eqs. (1) and (4), the PFC-equation (4) presents the second order in time and the six order in space equation. Its solution requires development special computational methods [11].

2. The results of simulations in computational domains of different size

In this section we present how the domain size influence on the steady solution of the MPFC task. This phenomenon can be illustrated by calculation results of the MPFC task with parameters of the average density $\phi = -0.34$ and the undercooling $\epsilon = 0.4$. This parameters correspond to the BCC crystalline pattern (Body Centered Crystal) from the three-dimension structure diagram of Jaatinen and Ala-Nissila [12]. The results were perfomed for the same task but for the cases of different cubic domains. Namely, consider domains with the size of the domain:

1) $25 \times 25 \times 25$,
2) $31 \times 31 \times 31$,
3) $32 \times 32 \times 32$,
4) $39 \times 39 \times 39$,
5) $45 \times 45 \times 45$.

All calculations are perfomed by the special computational method proposed in [13] which represented by the implicit finite element method (FEM) using regular hexagonal mesh. The computational efficiency of such algorithm was shown in [14, 15].

The ratio of the sizes of the computing domain and the mesh as shown in [16] should be taken into account for proper results. We have established this ratio to be equal to 0.78 for all tasks. Further in the section the figures with the visualization of stationary solutions of the described task are presented. The left side of each figure presents colored surface of computational domain in steady moment. The right side of each figure presents isocontours of the result into the computational domain it steady moment.

In figure 1, we can see pure BCC crystal pattern formed in the smallest domain 1. At the same time in domain 2 (figure 2) the stable solution is presented by curved rods or the "worms" pattern. Figure 3 presents that even small change of domain size can transform a stationary solution and instead of "worms" in the last case we can observe a mixed structure of domain 3. In figure 4, we can see pure BCC pattern again, this time in domain 4. But further increase
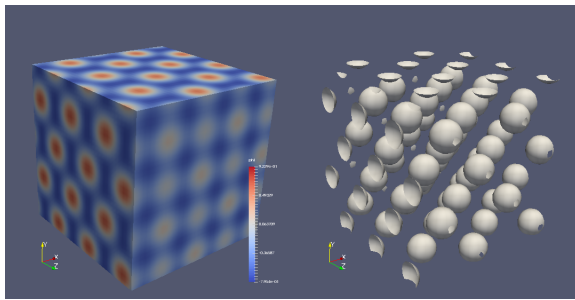
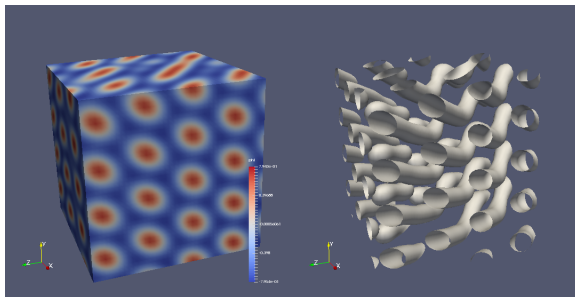**Figure 1.** Stable state formation prepared in simulation of the task in domain 1.

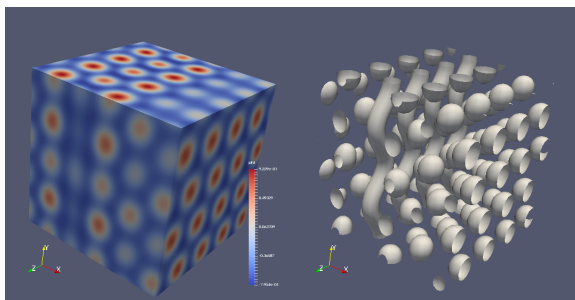**Figure 2.** Stable state formation prepared in simulation of the task in domain 2.

Figure 3. Stable state formation prepared in simulation of the task in domain 3.
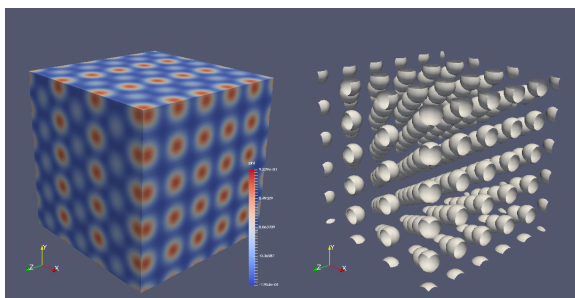

Figure 4. Stable state formation prepared in simulation of the task in domain 4.
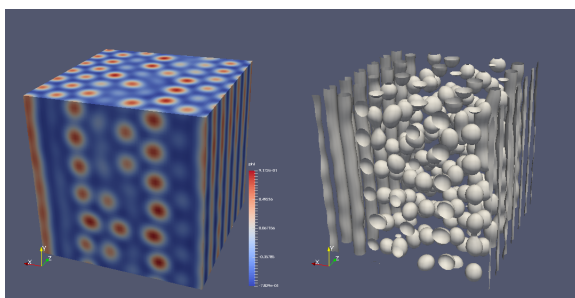

Figure 5. Stable state formation prepared in simulation of the task in domain 5.

in the size of the domain again leads to an alternative stationary solution of a mixed type. It's

presented in figure 5.

These results demonstrate the strong dependence of the stationary formation of the crystallization process on the sizes of the calculated domain. In two simulations the result is consistent with the expected BCC structure. In all other simulations the result could not be classified as known pattern type and probably is a metastable form. The reasons for this phenomenon may be a feature of the MPFC method. It consists in the fact that changes in the crystal structure occur due to minimization of the free energy functional (2). The local minima of the free energy functional can correspond to locally stable or metastable states of the crystal structure. With the increase in the size of the computational domain, the number of possible atomic density ϕ distributions in such a domain and in every moment of time is significantly increased. As a consequence, the probability of the system falling into the metastable region increases too. That, however, does not exclude the possibility of obtaining absolutely stable crystal formation.

3. Conclusions

The problem of crystal pattern formation in the Modified Phase Field Crystal Model has been discussed. To perform reliable computations, one should take into account the factors leading to instability. In the current report, we show that one of these factors is the size of computational domain. We believe that with increasing domain sizes, the probability of formation of metastable crystal patterns is substantially increased. In some cases, this can lead to a mismatch of the results with the predictions or to results that are unclassifiable. To improve the stability of the results we consider reasonable to add a stochastic vibrations in the atomic density field consistently with thermodynamic noise. It can be realized by modification of the PFC-model with taking into account the stochastic term. This should allow the system to come out of metastable states and increase the rate of formation of equilibrium and stable states consistent with diagrams of structures [12, 13].

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