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An energy-stable convex splitting for the phase-field crystal equation

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ABSTRACT

The phase-field crystal equation, a parabolic, sixth-order and nonlinear partial differential equation, has generated considerable interest as a possible solution to problems arising in molecular dynamics. Nonetheless, solving this equation is not a trivial task, as energy dissipation and mass conservation need to be verified for the numerical solution to be valid. This work addresses these issues, and proposes a novel algorithm that guarantees mass conservation, unconditional energy stability and second-order accuracy in time. Numerical results validating our proofs are presented, and two and three dimensional simulations involving crystal growth are shown, highlighting the robustness of the method.

1. Introduction

While the tight connection between material processing, structure and properties has been known for years, a microstructural model capable of accounting for the atomic scale features affecting the macroscale properties of a material has not yet been established. Progress has nonetheless been made in this direction, and this work tackles one of the solution strategies that has recently been proposed through the phase-field crystal equation (PFC). Developed as an extension to the phase-field formalism in which the fields take spatially uniform values at equilibrium [1,2], the free energy functional in the case of the PFC equation is minimized by periodic states. These periodic minima allow this particular phase-field model to represent crystalline lattices in two and three dimensions [3,4], and more importantly, to capture the interaction of defects that arise at the atomic scale without the use of additional fields, as is done in standard phase-field equations [5]. This model has also been shown to successfully cross time scales [6], thanks in part to the phase-field variable that describes a coarse grained temporal average (the number density of atoms). This difference in time scale with molecular dynamics, along with the periodic density states that naturally give rise to elasticity, multiple crystal orientations and the nucleation and motion of dislocations, are some of the reasons why this tool is being considered for quantitative modeling [7,8].

Several challenges are unfortunately faced while simulating the PFC numerically. It is a sixth-order, nonlinear, partial differential equation, where the solution should lead to a time-decreasing free energy functional. Recent work on this topic includes [9–14]. Inspired by the work presented for the Cahn–Hilliard equation in the context of tumor-growth [15], we developed a formulation capable of conserving mass, guaranteeing discrete energy stability while having second-order temporal accuracy. The numerical scheme achieves this through a convex splitting of the nonlinearity present in the equation, along with the addition of a stabilization term, while using a mixed form that segregates the partial differential equation into a system of three second order equations. This is similar in a sense to what was done in [12], where a mixed form is also used, but has the added advantage that the well-posedness of the variational form does not require globally $\mathcal{C}^1$-continuous basis functions. This presents an advantage in terms of computational cost [16–18] as linear, $\mathcal{C}^0$ finite elements can be used.

We provide mathematical proofs for mass conservation, energy stability and second-order accuracy, properties that the algorithm possesses, along with two-dimensional numerical evidence that corroborates our findings. We also present three dimensional results that showcase the effectiveness and robustness of our algorithm. The paper is structured as follows: In Section 2, we describe

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the phase field crystal equation. In Section 3, we present our numerical scheme. Section 4 presents numerical examples dealing with crystal growth in a supercooled liquid. We give concluding remarks in Section 5.

2. Phase-field crystal model

By using a free energy functional that is minimized by periodic density fields, the phase-field crystal equation is capable of representing crystalline lattices [1], and more importantly, capturing the interaction between material defects implicitly. The model is characterized by a conserved field related to the atomic number density, that is spatially periodic in the solid phase and constant in the liquid phase. It has been related successfully to other continuum field theories such as density-functional theory [6,19]. This work will show examples related to crystalline growth, as the PFC equation has found much of its success in modeling microstructural evolution [2,6,20–22], while it has also been used to model other physical phenomena such as foam dynamics [23], glass formation [24], liquid crystals [25], elasticity [1] and in the estimation of material properties [26].

Experimental and computational results can differ significantly, but work is nonetheless being done to reduce the mismatch [26–29]. The model that is considered in this work can be improved by increasing the number of critical wavelengths one considers in the free energy functional at the expense of computational cost, as the partial differential equation becomes harder to solve [8,28]. Also, molecular dynamics in a multi-scale setting can be used to estimate some of the parameters going into the phase-field crystal equation [30], and inverse formulations of the problem could be considered to validate the calculations [31]. Hopefully, these multi-scale approaches will allow for more complete studies on polycrystalline growth using the PFC equation, such as the ones presented in [32,33] in the setting of phase-field modeling.

2.1. Model formulation

The phase-field crystal equation was developed to study the evolution of microstructures, at atomic length scales and diffusive time scales, by considering a conservative description of the Rayleigh-Bénard convection problem [3]. The order parameter ϕ represents an atomistic density field in the model, which is periodic in the solid state and uniform in the liquid one. The free energy functional for the phase-field crystal equation in its dimensionless form is given by [2,4,12]

$$\mathcal{F}[\phi(\mathbf{x})] = \int_\Omega \left[\Psi(\phi) + \frac{1}{2}\left(\phi^2 - 2|\nabla\phi|^2 + (\Delta\phi)^2\right)\right] d\Omega, \tag{1}$$

where $\Omega \in \mathbb{R}^d$ represents an arbitrary open domain, with $d = 2$ or 3, and $\Psi(\phi) = -\frac{\epsilon}{2}\phi^2 + \frac{1}{4}\phi^4$. The parameter ϵ represents a critical transition variable, which in the case of crystal growth is associated to the degree of undercooling: the larger its value, the larger the undercooling is. The free energy functional presented in Eq. (1) is then minimized to achieve thermodynamical stability. To enforce this mathematically, one solves the Euler–Lagrange equation for the free energy, and takes its variational derivative with respect to ϕ. The variational derivative is given by

$$\frac{\delta\mathcal{F}}{\delta\phi} = \frac{\partial\mathcal{F}}{\partial\phi} - \nabla\cdot\frac{\partial\mathcal{F}}{\partial\nabla\phi} + \Delta\frac{\partial\mathcal{F}}{\partial\Delta\phi} = (1+\Delta)^2\phi + \Psi'(\phi), \tag{2}$$

where $\nabla\cdot, \nabla$ and Δ denote the divergence, gradient and Laplacian operators, respectively, and $\Psi'(\phi) = -\epsilon\phi + \phi^3$ with $(1+\Delta)^2 = 1 + 2\Delta + \Delta\Delta$. The partial differential equation, considering that the atomistic density field is a conserved quantity [2], is then formulated as

$$\frac{\partial\phi}{\partial t} = \nabla\cdot\left(M\nabla\frac{\delta\mathcal{F}}{\delta\phi}\right), \tag{3}$$

where $\phi \equiv \phi(\mathbf{x}, t)$ represents the phase field, $\mathbf{x}$ and t represent space and time, respectively, M is the mobility, and $\mathcal{F}$ is the free energy functional of the system. The partial differential equation, after substituting Eq. (2) into (3), becomes

$$\frac{\partial\phi}{\partial t} = \nabla\cdot\nabla\left[(1+\Delta)^2\phi + \Psi'(\phi)\right] = \Delta\left[(1+\Delta)^2\phi + \Psi'(\phi)\right],$$

where the mobility M is assumed equal to a constant of value one.

2.2. Phase-field crystal equation: strong form

The problem is stated as follows: over the spatial domain Ω and the time interval $]0, T[$, given $\phi_0 : \Omega \mapsto \mathbb{R}$, find $\phi : \Omega \times [0, T] \mapsto \mathbb{R}$ such that

$$\begin{cases} \frac{\partial\phi}{\partial t} = \Delta\left[(1+\Delta)^2\phi + \Psi'(\phi)\right] & \text{on } \Omega\times]0,T], \\ \phi(\mathbf{x}, 0) = \phi_0(\mathbf{x}) & \text{on } \Omega, \end{cases} \tag{4}$$

where $\phi_0(\mathbf{x})$ represents a function that approximates a crystalline nucleus, and periodic boundary conditions are imposed in all directions. We discuss the choices made to handle initial conditions in Section 4.

3. Stable time discretization for the phase-field crystal equation

The phase-field crystal equation is a sixth-order, parabolic partial differential equation. If an explicit time-stepping scheme were employed to solve it, a time step size Δt on the order of the sixth power of the grid size would be required. This restriction has motivated research in implicit algorithms [9–12] and adaptive algorithms [34]. On top of this, some properties need to be guaranteed while solving the equation, such as mass conservation, defined as

$$\int_\Omega \left(\frac{\partial\phi}{\partial t}\right) d\Omega = 0 \tag{5}$$

due to the fact that density is conserved, as well as strong energy stability [9], expressed as

$$\mathcal{F}[\phi(t_{n+1})] \leqslant \mathcal{F}[\phi(t_n)] \quad \forall n = 1, 2, \ldots, N, \tag{6}$$

which translates to having the free energy be monotonically decreasing. In this work, we develop an algorithm that extends the ideas presented in [11,15], guarantees the properties presented in Eqs. (5) and (6), while achieving second-order accuracy in time. The discretization in space is done using isogeometric analysis (IGA), a finite element method where NURBS are used as basis functions [35]. The method not only allows to control the spatial resolution of the mesh (h-refinement) and the polynomial degree of the basis (p-refinement), but also to increase their global continuity (k-refinement). Isogeometric analysis has successfully been applied to phase-field modeling [12,13,36–41]. The PFC model, being a nonlinear, sixth-order in space, first-order in time partial differential equation, allows for many choices in terms of discretizations and time stepping schemes. High-order, globally continuous basis functions can be easily generated within the IGA framework. This is the reason why it allows for the straightforward discretization of high-order partial differential equations. Alternatively, mixed formulations can be employed so as to reduce the continuity requirements down to standard $\mathcal{C}^0$ spaces used in traditional finite element methods. This work makes use of a mixed form, where the system that is solved involves a coupled system of three second-order equations.