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Numerical simulation of the formation of spherulites in polycrystalline binary mixtures

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Abstract

Spherulites are growth patterns of average spherical form which may occur in the polycrystallization of binary mixtures due to misoriented angles at low grain boundaries. The dynamic growth of spherulites can be described by a phase field model where the underlying free energy depends on two phase field variables, namely the local degree of crystallinity and the orientation angle. For the solution of the phase field model we suggest a splitting scheme based on an implicit discretization in time which decouples the model and at each time step requires the successive solution of an evolutionary inclusion in the orientation angle and an evolutionary equation in the local degree of crystallinity. The discretization in space is done by piecewise linear Lagrangian finite elements. The fully discretized splitting scheme amounts to the solution of two systems of nonlinear algebraic equations. For the numerical solution we suggest a predictor-corrector continuation method with the discrete time as a parameter featuring constant continuation as a predictor and a semismooth Newton method for the first system and the classical Newton method for the second system as a corrector. This allows an adaptive choice of the time steps. Numerical results are given for the formation of a Category 1 spherulite.

1. Introduction

The morphosynthesis of polycrystalline thin films is of considerable importance in materials science. Due to their low surface roughness at the nanoscale and their thermodynamic stability, they are of interest for diffraction gratings, photonic band gap structures, and coatings based on structural colors instead of pigments. The morphosynthesis is a multistage process consisting of a polymer-induced liquid-precursor phase, the occurrence of spherulites due to surface nucleation processes, and the formation of a mosaic polycrystalline thin film. The polycrystallization sets in with the formation of objects of spherical shape, called spherulites, that spread across the substrate to form a uniform spherulitic structure. The final stage consists of a recrystallization of the spherulitic patterns into a mosaic polycrystalline thin film. Polycrystallization has been widely considered in the literature. We refer to the survey

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articles [9, 10] and the references therein.
According to experimental evidence [17] one distinguishes between Category 1 and Category 2 spherulites. The former grow radially from the nucleation site, whereas the latter grow as threadlike fibers. Both categories show crystalline branching which can be caused by a misorientation at low grain boundaries. In [8], the authors suggested a phase field model featuring an orientational free energy density f_{ori} in the phase field variables (local degree of crystallinity ϕ and orientation angle Θ) which takes care of misorientation. As a function of Θ, the orientational free energy density is related to the total variation of Θ. Consequently, from a mathematical point of view one has to consider Θ in the Banach space BV of functions of bounded variation as has been done in [13] for the two-field Kobayashi-Warren-Carter model [15] and in [14] for a three-field phase field model for polycrystalline growth in binary mixtures. As far as the minimization of functionals in terms of the total variation is concerned, there is a fundamental result stated in [16]. Roughly speaking, it says that if the integrand is a Carathéodory function, convex, and of linear growth at infinity, then the functional is lower semicontinuous with regard to the weak* topology in BV. However, the orientational free energy density f_{ori} as suggested in [8] is not convex. Here, we suggest a modification of f_{ori} whose convexification $f_{ori,c}$ makes the free energy functional weakly* lower semicontinuous in BV.

The paper is organized as follows: In section 2, we provide basic notations and preliminary results with emphasis on the Banach space BV of functions of bounded variation and weak* lower semicontinuity results for functionals in BV. The following section 3 is devoted to the phase field model in the local degree of crystallinity ϕ and the orientation angle Θ as phase field variables. In particular, we present a modification of the orientational free energy density f_{ori} from [8] in terms of a convexified function $f_{ori,c}$ which renders the free energy functional as a function of Θ to be weakly* lower semicontinuous in BV. Specifying mobilities M_ϕ and M_Θ, the dynamics of the polycrystallization process can be described by a coupled system of evolutionary initial-boundary value problems consisting of an evolution inclusion in Θ and an evolution equation in ϕ. Following the approach in [13] and [14], in section 4 we suggest a splitting scheme based on an implicit discretization in time which decouples the evolutionary problems such that at each time step the evolution inclusion in Θ is solved first followed by the evolution equation in ϕ. We show that the split system is related to two minimization problems for functionals in BV and the Sobolev space $W^{1,2}$ that both admit a solution as can be shown by tools from the calculus of variations. Section 5 deals with a further discretization in space by piecewise linear Lagrangian finite elements with respect to a uniform, geometrically conforming, simplicial triangulation of the computational domain. This results in two nonlinear algebraic systems for functions F_1 in Θ and F_2 in ϕ that have to be solved successively at each time step. Since F_1 only admits a generalized Jacobian, for the first system we have to resort to a semismooth Newton method. On the other hand, F_2 is differentiable in the standard sense so that the associated nonlinear system can be solved by the classical Newton method. However, the appropriate choice of the time steps is crucial for the convergence. To overcome that difficulty, we reformulate the systems as parameter dependent nonlinear systems with the discrete time as a parameter and suggest a predictor-corrector continuation strategy featuring constant continuation as a predictor and the semismooth and the classical Newton method as a corrector. The predictor-corrector continuation method allows an adaptive choice of the time steps. Finally, in section 6 we apply the suggested approach to the formation of a Category 1 spherulite and document the performance of the numerical solution method.

2. Notations and Basic Results

For an open or closed set $A \subset \mathbb{R}^d, d \in \mathbb{N}$, we denote by $C_0^m(A;\mathbb{R}^d), 0 \leq m < \infty$, the Banach space of m-times continuously differentiable vector-valued functions $\mathbf{q} = (q_1, \cdots, q_d)$ with compact support in A. In case $m = 0$ we write $C_0(A;\mathbb{R}^d)$ instead of $C_0^0(A;\mathbb{R}^d)$ and in case $d = 1$ we write $C_0^m(A)$ instead of $C_0^m(A;\mathbb{R}^1)$. We further refer to $C_0^\infty(A)$ as the linear space of infinitely smooth (scalar) functions with compact support in A and to $\mathcal{D}(A)$ as its dual space of distributional derivatives.
By $\mathcal{M}(A;\mathbb{R}^d), d \in \mathbb{N}$, we denote the Banach space of vector-valued bounded Radon measures $\boldsymbol{\mu} = (\mu_1, \cdots, \mu_d)$ equipped with the total variation norm

$$|\boldsymbol{\mu}|(A) := \sup \{\sum_{n=1}^{\infty} |\boldsymbol{\mu}(A_n)| \mid \Omega = \bigcup_{n=1}^{\infty} A_n, A_n \cap A_m = \emptyset \text{ for } n \neq m\} \tag{2.1}$$

and we refer to $\mathcal{M}^+(A;\mathbb{R}^d)$ as the set of positive Radon measures.
In view of the Riesz representation theorem $\mathcal{M}(A;\mathbb{R}^d)$ is the dual space of $C_0(A;\mathbb{R}^d)$ with the duality pairing

$$\langle \boldsymbol{\mu}, \mathbf{q}\rangle_{\mathcal{M},C_0} := \int\limits_\Omega \mathbf{q}\, d\boldsymbol{\mu} = \sum_{i=1}^d \int\limits_A q_i\, d\mu_i. \tag{2.2}$$

A sequence $\{\boldsymbol{\mu}_n\}_\mathbb{N}$ of Radon measures $\boldsymbol{\mu}_n \in \mathcal{M}(A;\mathbb{R}^d), n \in \mathbb{N}$, is said to converge weakly* to $\boldsymbol{\mu} \in \mathcal{M}(A;\mathbb{R}^d)$ ($\boldsymbol{\mu}_n \rightharpoonup^* \boldsymbol{\mu}\ (n\to\infty)$) if

$$\langle \boldsymbol{\mu}_n, \mathbf{q}\rangle_{\mathcal{M},C_0} \to \langle \boldsymbol{\mu}, \mathbf{q}\rangle_{\mathcal{M},C_0}\ (n\to\infty) \quad \text{for all } \mathbf{q}\in C_0^\infty(A;\mathbb{R}^d). \tag{2.3}$$

The following weak compactness result holds true:

Lemma 2.1. *Let $\{\boldsymbol{\mu}_n\}_\mathbb{N}$ be a bounded sequence of measures $\boldsymbol{\mu}_n \in \mathcal{M}(A;\mathbb{R}^d), n\in\mathbb{N}$. Then there exist a subsequence $\mathbb{N}'\subset\mathbb{N}$ and a measure $\boldsymbol{\mu}\in\mathcal{M}(A;\mathbb{R}^d)$ such that*

$$\boldsymbol{\mu}_n \rightharpoonup^* \boldsymbol{\mu}\ (\mathbb{N}'\ni n\to\infty) \quad \textit{in } \mathcal{M}(A;\mathbb{R}^d). \tag{2.4}$$

Proof. We refer to [7].

For a bounded Lipschitz domain $\Omega\subset\mathbb{R}^d, d\in\mathbb{N}$, with boundary $\Gamma=\partial\Omega$ we refer to $L^p(\Omega;\mathbb{R}^d), 1\le p<\infty$, as the Banach space of p-th power Lebesgue integrable vector-valued functions on Ω with norm $\|\cdot\|_{L^p(\Omega;\mathbb{R}^d)}$ and to $L^\infty(\Omega;\mathbb{R}^d)$ as the Banach space of essentially bounded vector-valued functions on Ω with norm $\|\cdot\|_{L^\infty(\Omega;\mathbb{R}^d)}$. In case $d=1$ we will write $L^p(\Omega)$ instead of $L^p(\Omega;\mathbb{R}^1)$. Further, we denote by $W^{1,p}(\Omega), 1\le p\le\infty$, the Sobolev spaces with norms $\|\cdot\|_{W^{s,p}(\Omega)}$ and by $W^{1,p}(\Omega), 1<p<\infty$, the closure of $C_0^\infty(\Omega)$ with respect to the $\|\cdot\|_{W^{1,p}}$ norm. We note that for $1<p<\infty$ the Sobolev space $W^{1,p}(\Omega)$ is reflexive with dual space $W^{1,q}(\Omega), 1/p+1/q=1$. For $p=1$ the dual space of the Sobolev space $W^{1,1}(\Omega)$ is $W^{1,\infty}(\Omega)$. However, the Sobolev space $W^{1,1}(\Omega)$ is not reflexive. We further note that for $p=2$ the spaces $L^2(\Omega;\mathbb{R}^d)$ and $W^{1,2}(\Omega)=H^1(\Omega)$ are Hilbert spaces with inner products $(\cdot,\cdot)_{L^2(\Omega;\mathbb{R}^d)}$ and $(\cdot,\cdot)_{W^{1,2}(\Omega)}$.
A sequence $\{u_n\}_\mathbb{N}$ of functions $u_n\in W^{1,p}(\Omega), n\in\mathbb{N}, 1<p<\infty$, is said to converge weakly to $u\in W^{1,p}(\Omega)$ ($u_n\rightharpoonup u\ (n\to\infty)$), if it holds

$$\langle v, u_n\rangle_{W^{1,q},W^{1,p}} \to \langle v, u\rangle_{W^{1,q},W^{1,p}}\ (n\to\infty) \quad \text{for all } v\in W^{1,q}(\Omega),\ 1/p+1/q=1.$$

Lemma 2.2. *Let $\{u_n\}_\mathbb{N}$ be a bounded sequence of functions $u_n\in W^{1,p}(\Omega), n\in\mathbb{N}, 1<p<\infty$. Then there exist a subsequence $\mathbb{N}'\subset\mathbb{N}$ and a function $u\in W^{1,p}(\Omega)$ such that*

$$u_n \rightharpoonup u\ (\mathbb{N}'\ni n\to\infty) \quad \textit{in } W^{1,p}(\Omega). \tag{2.5}$$

Proof. We refer to [7].

A functional $F: W^{1,p}(\Omega)\to\mathbb{R}, 1<p<\infty$, is said to be weakly sequential lower semicontinuous in $W^{1,p}(\Omega)$ if for every sequence $\{u_n\}_\mathbb{N}$ of functions $u_n\in W^{1,p}(\Omega), n\in\mathbb{N}$, such that $u_n\rightharpoonup u\ (n\to\infty)$ in $W^{1,p}(\Omega)$ for some $u\in W^{1,p}(\Omega)$ it holds

$$F(u)\le \liminf_{n\to\infty} F(u_n). \tag{2.6}$$

Due to the fact that $W^{1,1}(\Omega)$ is not reflexive, the situation for $p=1$ is more involved.
A sequence $\{u_n\}_\mathbb{N}$ of functions $u_n\in W^{1,1}(\Omega), n\in\mathbb{N}$, is said to be equiintegrable, if the following two conditions hold true:

(i) For any $\varepsilon > 0$ there exists a Lebesgue measurable set $A \subset \Omega$ such that

$$\int_{\Omega \setminus A} |u_n| + |\nabla u_n| \ dx < \varepsilon, \quad n \in \mathbb{N}. \tag{2.7a}$$

(ii) For any $\varepsilon > 0$ there exists $\delta > 0$ such that for any Lebesgue measurable set $E \subset \Omega$ with $|E| < \delta$ it holds

$$\int_{E} |u_n| + |\nabla u_n| \ dx < \varepsilon, \quad n \in \mathbb{N}. \tag{2.7b}$$

The following theorem by Dunford and Pettis gives a necessary and sufficient condition for weak compactness in $W^{1,1}(\Omega)$.

Theorem 2.1. *Let $\{u_n\}_{\mathbb{N}}$ be a sequence of functions $u_n \in W^{1,1}(\Omega), n \in \mathbb{N}$, that is uniformly bounded and equiintegrable. Then there exist a subsequence $\mathbb{N}' \subset \mathbb{N}$ and a function $u \in W^{1,1}(\Omega)$ such that*

$$u_n \rightharpoonup u \ (\mathbb{N}' \ni n \to \infty) \quad \text{in } W^{1,1}(\Omega).$$

Conversely, if the sequence $\{u_n\}_{\mathbb{N}}$ converges weakly to $u \in W^{1,1}(\Omega)$, then it is uniformly bounded and equiintegrable.

Proof. We refer to [7].

However, if equiintegrability does not apply, we have to resort to functions of bounded variation (cf., e.g., [1, 2]). A function $u \in L^1(\Omega)$ is said to be of bounded variation if its distributional derivative Du satisfies $Du \in \mathcal{M}(\Omega; \mathbb{R}^d)$, i.e., for all $\mathbf{q} \in C_0^1(\Omega; \mathbb{R}^d)$ we have

$$-\int_{\Omega} \nabla \cdot \mathbf{q} u \, dx = \int_{\Omega} \mathbf{q} \cdot dDu.$$

The total variation of u is defined as follows

$$|Du|(\Omega) := \sup \{-\int_{\Omega} \nabla \cdot \mathbf{q} u \, dx \mid \mathbf{q} \in C_0^1(\Omega; \mathbb{R}^d), |\mathbf{q}| \leq 1 \text{ in } \Omega\}. \tag{2.8}$$

We denote by $BV(\Omega)$ the Banach space of functions $u \in L^1(\Omega)$ such that $|Du|(\Omega) < \infty$ equipped with the norm

$$\|u\|_{BV(\Omega)} := \|u\|_{L^1(\Omega)} + |Du|(\Omega). \tag{2.9}$$

Clearly, we have $W^{1,1}(\Omega) \subset BV(\Omega)$ and $u \in W^{1,1}(\Omega)$ iff $u \in L^1(\Omega)$ and Du is absolutely continuous with respect to the Lebesgue measure. In particular, we have the Lebesgue-Radon-Nikodym decomposition

$$Du = \nabla u + D^s u, \tag{2.10}$$

where $\nabla u \in L^1(\Omega; \mathbb{R}^d)$ is called the approximate gradient of u and $D^s u \in \mathcal{M}(\Omega; \mathbb{R}^d)$ is said to be the singular part of the derivative.
Functions $u \in BV(\Omega)$ have a trace $u|_\Gamma \in L^1(\Gamma)$. The trace mapping $T : BV(\Omega) \to L^1(\Gamma)$ is linear, continuous from $BV(\Omega)$ endowed with the strict topology to $L^1(\Gamma)$ equipped with the strong topology (cf., e.g., Theorem 10.2.2 in [2]). The subspace $BV_0(\Omega)$ of $BV(\Omega)$ is the kernel of the trace mapping T. It is a Banach space equipped with the induced norm.

A sequence $\{u_n\}_{\mathbb{N}}$ of functions $u_n \in BV(\Omega), n \in \mathbb{N}$, is said to converge weakly* to $u \in BV(\Omega)$ if $u_n \to u$ in $L^1(\Omega)$ and $Du_n \rightharpoonup^* Du$ in $\mathcal{M}(\Omega; \mathbb{R}^d)$ as $\mathbb{N} \ni n \to \infty$.

Lemma 2.3. *Let $\{u_n\}_{\mathbb{N}}$ be a uniformly bounded sequence of functions $u_n \in BV(\Omega), n \in \mathbb{N}$, i.e.,*

$$\|u_n\|_{BV(\Omega)} \leq C, \quad n \in \mathbb{N},$$

for some $C > 0$. Then there exist a subsequence $\mathbb{N}' \subset \mathbb{N}$ and $u \in BV(\Omega)$ such that

$$u_n \rightharpoonup^* u \quad \text{in } BV(\Omega) \text{ as } \mathbb{N}' \ni n \to \infty.$$

Proof. We refer to [1] .

A functional $F : BV(\Omega) \to \mathbb{R}$ is said to be weakly* sequential lower semicontinuous in $BV(\Omega)$ if for every sequence $\{u_n\}_{\mathbb{N}}$ of functions $u_n \in BV(\Omega), n \in \mathbb{N}$, such that $u_n \rightharpoonup^* u$ $(n \to \infty)$ in $BV(\Omega)$ for some $u \in BV(\Omega)$ it holds

$$F(u) \leq \liminf_{n\to\infty} F(u_n). \tag{2.11}$$

A function $f : \bar{\Omega}\times\mathbb{R}^d \to \mathbb{R}$ is called a Carathéodory function, if f is measurable and the mapping $\mathbf{q} \mapsto f(x, \mathbf{q}), \mathbf{q} \in \mathbb{R}^d$, is continuous for almost all $x \in \bar{\Omega}$. It is said to have linear growth at infinity, if there exists $C_f > 0$ such that

$$|f(x, \mathbf{q})| \leq C_f(1 + |\mathbf{q}|) \quad \text{for almost all } x \in \bar{\Omega} \text{ and all } \mathbf{q} \in \mathbb{R}^d.$$

If the limit

$$f^\infty(x, \mathbf{q}) := \lim \{t^{-1} f(x', t\mathbf{q}') \mid x' \to x, \mathbf{q}' \to \mathbf{q}, t \to \infty\} \tag{2.12}$$

exists for all $x \in \bar{\Omega}$ and all $\mathbf{q} \in \mathbb{R}^d$, then the function $f^\infty : \bar{\Omega} \times \mathbb{R}^d \to \mathbb{R}$ is called the recession function of f.

Theorem 2.2. *Let $\Omega \subset \mathbb{R}^d$ be a bounded Lipschitz domain with boundary $\Gamma = \partial\Omega$ and inner boundary normal $\mathbf{n}_\Gamma$. Let $f : \bar{\Omega} \times \mathbb{R}^d \to \mathbb{R}$ be a Carathéodory function with linear growth at infinity such that the mapping $\mathbf{q} \mapsto f(x, \mathbf{q})$ is convex for each fixed $x \in \bar{\Omega}$. Further assume that the recession function $f^\infty \in C(\bar{\Omega} \times \mathbb{R}^d)$ exists, Then the functional*

$$\begin{aligned} F(u) := \int_\Omega f(x, \nabla u)\, dx + \int_\Omega f^\infty(x, \frac{D^s u}{|D^s u|})\, d|D^s u| + \\ \int_\Gamma f^\infty(x, \frac{u}{|u|} \otimes \mathbf{n}_\Gamma)\, d\mathcal{H}^{d-1}, \quad u \in BV(\Omega), \end{aligned} \tag{2.13}$$

is weakly sequential lower semicontinuous in $BV(\Omega)$, where $\mathcal{H}^{d-1}$ stands for the (d-1)-dimensional Hausdorff measure.*

Proof. We refer to Theorem 10 in [16].

3. The Phase Field Model

As a mathematical model for the growth of spherulites in polycrystalline binary mixtures we use a modification of a phase field model from [8] where the free energy depends on two phase field variables. These are a structural order parameter ϕ measuring the local degree of crystallinity (volume fraction of the crystalline phase) and an orientation field Θ which locally describes the crystallographic orientation. For a bounded domain $\Omega \subset \mathbb{R}^2$ with boundary $\Gamma = \partial\Omega$ the free energy reads as follows:

$$F(\phi, \Theta) = \int_\Omega \frac{1}{2} s(\nabla\phi, \Theta)^2\, |\nabla\phi|^2 + g(\phi) + \frac{H}{2\xi_0} f_{ori}(\omega(\phi), |\nabla\Theta|) \;\; dx. \tag{3.1}$$

Here, the functions $s = s(\nabla\phi, \Theta)$, $g(\phi)$, and $f_{ori}(\omega(\phi), \nabla\Theta)$ refer to an anisotropy function, a double-well potential, and an orientational free energy density. The function ω is a continuously differentiable interpolation function given by

$$\omega(\eta) = \begin{cases} \varepsilon_r \ , \eta \leq 0 \\ \varepsilon_r + 2(2 - 3\varepsilon_r)\eta^2 - 4(1 - \varepsilon_r)\eta^3 + \eta^4 \ , 0 \leq \eta \leq 1 \\ 1 - \varepsilon_r \ , \eta \geq 1 \end{cases} \ , \ \eta \in \mathbb{R}, \tag{3.2}$$

where $0 < \varepsilon_r \ll 1$. The function ω has the property

$$0 < \varepsilon_r \leq \omega(\eta) \leq 1 - \varepsilon_r, \quad \eta \in \mathbb{R}. \tag{3.3}$$

Moreover, the anisotropy function $s(\boldsymbol{\eta}, \gamma), \boldsymbol{\eta} = (\eta_1, \eta_2)^T \in \mathbb{R}^2, \gamma \in \mathbb{R}$, is given by

$$s(\boldsymbol{\eta}, \gamma) = 1 + s_0 \cos(m_S \vartheta - 2\pi\gamma), \tag{3.4a}$$

$$\vartheta := \begin{cases} \arctan(\eta_2/\eta_1) \ , \eta_1 \neq 0 \\ \operatorname{sign}(\eta_2) \ \frac{\pi}{2} \ , \eta_1 = 0 \end{cases} , \tag{3.4b}$$

where $0 \leq s_0 < 1$ is the amplitude of the anisotropy of the interfacial free energy and m_S is the symmetry index. We note that ϑ is related to the inclination of the normal vector of the interface in the laboratory frame. The function $g(\eta)$ is the quartic double-well function

$$g(\eta) = \frac{1}{4} \eta^2 (1 - \eta)^2 \tag{3.5}$$

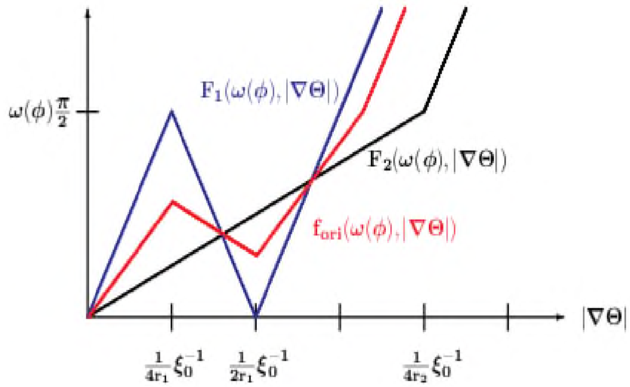

Figure 1. The functions $F_i(\omega(\phi), \nabla\Theta)$, $1 \leq i \leq 2$,, and $f_{ori}(\omega(\phi), \nabla\Theta)$.

The orientational free energy density f_{ori} as suggested in [8] is not convex. Here, in view of Theorem 2.2 we use a modification of f_{ori} from [8] which reads as follows

$$f_{ori}(\omega(\phi), \nabla\Theta) = \delta \ F_1(\omega(\phi), \nabla\Theta) + (1 - \delta) \ F_2(\omega(\phi), \nabla\Theta), \tag{3.6}$$

where $\delta \in (0, 1)$ and for $0 < r_2 < r_1$ the functions $F_i(\omega(\phi), \nabla\Theta)$, $1 \leq i \leq 2$, are given by

$$F_1(\omega(\phi), \nabla\Theta) = \begin{cases} 2\pi r_1 \xi_0 \omega(\phi)|\nabla\Theta| \ , \ \xi_0|\nabla\Theta| \leq \frac{1}{4r_1} \\ 2\pi r_1 \xi_0 \omega(\phi)(\frac{1}{4r_1}\xi_0^{-1} - |\nabla\Theta|) \ , \frac{1}{4r_1} \leq \xi_0|\nabla\Theta| \leq \frac{1}{2r_1} \\ 2\pi r_1 \xi_0 \omega(\phi)(|\nabla\Theta| - \frac{1}{2r_1}\xi_0^{-1}) \ , \ \xi_0|\nabla\Theta| \geq \frac{1}{2r_1} \end{cases} ,$$

$$F_2(\omega(\phi), \nabla\Theta) = \begin{cases} 2\pi r_2 \xi_0 \omega(\phi)|\nabla\Theta| \ , \ \xi_0|\nabla\Theta| \leq \frac{1}{4r_2} \\ \omega(\phi)\frac{\pi}{2} + 2\pi r_1 \xi_0 \omega(\phi)(|\nabla\Theta| - \frac{1}{4r_2}\xi_0^{-1}) \ , \ \xi_0|\nabla\Theta| \geq \frac{1}{4r_2} \end{cases} . \tag{3.7}$$

Here, the constants $H > 0$ and $\xi_0 > 0$ stand for the free energy of the low-grain boundaries and the correlation length of the orientational field (cf. Figure 1).

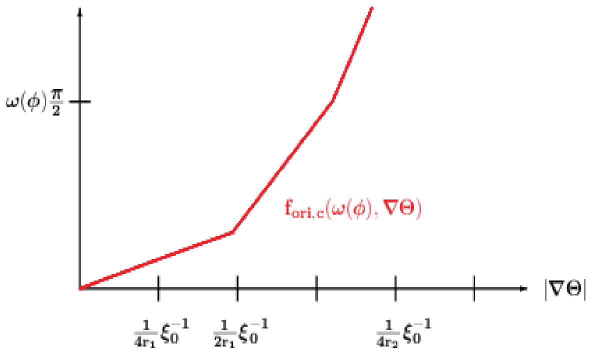

Figure 2. The convexified orientational free energy density $f_{ori,c}(\omega(\phi), \nabla\Theta)$.

The function f_{ori} as given by (3.6) is not convex. Its convexification $f_{ori,c}$ is given by (cf. Figure 2):

$$f_{ori,c}(\omega(\phi), \nabla\Theta) = \begin{cases} b_1\,|\nabla\Theta| \ , \ \xi_0|\nabla\Theta| \leq \frac{1}{2r_1} \\ a_2 + b_2\,(|\nabla\Theta| - \frac{1}{2r_1}\xi_0^{-1}) \ , \frac{1}{2r_1} \leq \xi_0|\nabla\Theta| \leq \frac{\delta+1/2}{2(\delta r_1+(1-\delta)r_2)} \\ a_3 + b_3(|\nabla\Theta| - \frac{\delta+1/2}{2(\delta r_1+(1-\delta)r_2)}\xi_0^{-1}) \ , \ \xi_0|\nabla\Theta| \geq \frac{\delta+1/2}{2(\delta r_1+(1-\delta)r_2)} \end{cases}, \tag{3.8}$$

where

$$\begin{aligned} &b_1 := 2(1-\delta)\pi\xi_0\omega(\phi)r_2, \\ &a_2 := (1-\delta)\pi\omega(\phi)\frac{r_2}{r_1}, \quad b_2 := 2\pi\xi_0\omega(\phi)\frac{(1/2-(1-\delta)r_2/r_1)r_1(\delta r_1+(1-\delta)r_2)}{r_1/2-(1-\delta)r_2}, \\ &a_3 := \pi\omega(\phi)/2, \quad b_3 := 2\pi r_1\xi_0\omega(\phi). \end{aligned}$$

The convexified orientational free energy density $f_{ori,c}(\omega(\phi), \nabla\Theta)$ is not differentiable in the classical sense, but admits a subdifferential $\partial_{\nabla\Theta} f_{ori,c}(\omega(\phi), \nabla\Theta)$ which is given by

$$\partial_{\nabla\Theta} f_{ori,c}(\omega(\phi), \nabla\Theta) = \begin{cases} b_1 \ , \ |\nabla\Theta| < \frac{1}{2r_1}\xi_0^{-1} \\ \left[b_1, b_2\right] \ , \ |\nabla\Theta| = \frac{1}{2r_1}\xi_0^{-1} \\ b_2 \ , \ \frac{1}{2r_1}\xi_0^{-1} < |\nabla\Theta| < \frac{1}{2r_1}\xi_0^{-1} \\ \left[b_2, b_3\right] \ , \ |\nabla\Theta| = \frac{1}{r_1+r_2}\xi_0^{-1} \\ b_3 \ , \ \frac{1}{r_1+r_2}\xi_0^{-1} < |\nabla\Theta| \end{cases}. \tag{3.9}$$

Likewise, the associated free energy $F(\phi, \Theta)$ is not Gâteaux differentiable in Θ. We set

$$\begin{aligned}
F_1(\Theta,\phi) &:= F_{11}(\Theta,\phi) + F_{12}(\Theta,\phi), \\
F_{11}(\Theta,\phi) &:= \frac{H}{2\xi_0}\int\limits_{\Omega} f_{ori,c}(\omega(\phi),\nabla\Theta)\,dx, \\
F_{12}(\Theta,\phi) &:= \frac{1}{2}\int\limits_{\Omega} s(\nabla\phi,\Theta)^2\,|\nabla\phi|^2\,dx.
\end{aligned} \tag{3.10}$$

We note that the functional $F_{12}(\Theta, \phi)$ is Gâteaux differentiable in Θ with Gâteaux derivative

$$\frac{\delta F_{12}(\Theta,\phi)}{\delta\Theta} = s(\nabla\phi,\Theta)\frac{\partial s(\nabla\phi,\Theta)}{\partial\Theta}|\nabla\phi|^2, \tag{3.11}$$

whereas the functional $F_{11}(\Theta, \phi)$ admits a subdifferential $\partial_\Theta F_{11}(\Theta, \phi)$ given by

$$\partial_\Theta F_{11}(\Theta,\phi) = \frac{H}{\xi_0}\Big\{ -\nabla\cdot\mathbf{q} \mid \mathbf{q} \in \begin{cases} \partial_{\nabla\Theta} f_{ori,c}(\omega(\phi),\nabla\Theta)\frac{\nabla\Theta}{|\nabla\Theta|} &, \text{ if } \nabla\Theta \neq 0 \\ s_1\,[-1,+1] &, \text{ if } \nabla\Theta = 0 \end{cases} \Big\}. \tag{3.12}$$

The functionals $F_{1i}(\Theta, \phi)$, $1 \le i \le 2$, are Gâteaux differentiable in ϕ with Gâteaux derivatives

$$\begin{aligned}
\frac{\delta F_{11}(\phi,\Theta)}{\delta\phi} &= g_\phi(\phi) + \frac{H}{\xi_0}\frac{\partial f_{ori,c}(\omega(\phi),\nabla\Theta)}{\partial\omega(\phi)}\omega_\phi(\phi), \\
\frac{\delta F_{12}(\phi,\Theta)}{\delta\phi} &= -\nabla\cdot(\mathbb{A}(\nabla\phi,\Theta)\nabla\phi),
\end{aligned} \tag{3.13}$$

where the 2×2 matrix $\mathbb{A}(\boldsymbol{\eta},\gamma) = (a_{ij}(\boldsymbol{\eta},\gamma))_{i,j=1}^2$, $\boldsymbol{\eta}\in\mathbb{R}^2, \gamma\in\mathbb{R}$, is given by

$$\begin{aligned}
a_{11}(\boldsymbol{\eta},\gamma) &= a_{22}(\boldsymbol{\eta},\gamma) = s(\boldsymbol{\eta},\gamma)^2, \\
a_{12}(\boldsymbol{\eta},\gamma) &= -a_{21}(\boldsymbol{\eta},\gamma) = -s(\boldsymbol{\eta},\gamma)\frac{\partial s(\boldsymbol{\eta},\gamma)}{\partial\vartheta},
\end{aligned} \tag{3.14}$$

and $g_\phi(\phi), h_\phi(\phi, c, T)$, and $\omega_\phi(\phi)$ stand for the derivatives of $g(\phi), h(\phi, c, T)$, and $\omega(\phi)$ with respect to ϕ.

Remark 3.1. *It is well known that a matrix $\mathbb{A}\in\mathbb{R}^{n\times n}$ is positive definite if and only if its symmetric part is positive definite. The symmetric part $\mathbb{A}_s(\boldsymbol{\eta},\gamma)$ of $\mathbb{A}(\boldsymbol{\eta},\gamma)$ is given by $\mathbb{A}_s(\boldsymbol{\eta},\gamma) = diag(s(\boldsymbol{\eta},\gamma)^2, s(\boldsymbol{\eta},\gamma)^2)$ and hence, $\mathbb{A}\in\mathbb{R}^{n\times n}$ is positive definite in $\boldsymbol{\eta}$ for any $0 \le s_0 < 1$. In particular, for such s_0 we have*

$$\boldsymbol{\eta}^T\mathbb{A}(\boldsymbol{\eta},\gamma)\boldsymbol{\eta} \ge (1-s_0)^2|\boldsymbol{\eta}|^2, \quad \boldsymbol{\eta}\in\mathbb{R}^2,\ \gamma\in\mathbb{R}. \tag{3.15}$$

Denoting by $M_\Theta > 0$ and $M_\phi > 0$ the mobilities with respect to Θ and ϕ and specifying initial and boundary conditions, the dynamics of the spherulitic growth are described by the coupled system of evolutionary processes

$$\frac{\partial\Theta}{\partial t} = M_\Theta\ \Big(-\partial_\Theta F_{11}(\Theta,\phi) + \frac{\delta F_{12}(\Theta,\phi)}{\delta\Theta}\Big), \quad \text{in } Q := \Omega\times(0,T), \tag{3.16a}$$

$$\Theta = 0 \quad \text{on } \Sigma := \Gamma\times(0,T), \tag{3.16b}$$

$$\Theta(0) = \Theta_0 \quad \text{in } \Omega, \tag{3.16c}$$

and

$$\frac{\partial\phi}{\partial t} = M_\phi\ \Big(-\frac{\delta F_{12}(\phi,\Theta)}{\delta\phi} + \frac{\delta F_{11}(\Theta,\phi)}{\delta\phi}\Big) \quad \text{in } Q := \Omega\times(0,T), \tag{3.17a}$$

$$\phi = 0 \quad \text{on } \Sigma := \Gamma\times(0,T), \tag{3.17b}$$

$$\phi(0) = \phi_0 \quad \text{in } \Omega, \tag{3.17c}$$

where Θ_0 and ϕ_0 are given initial configurations.

4. The splitting scheme

We consider a discretization in time with respect to a partition of the time interval $[0,T]$ into subintervals $[t_{m-1},t_m]$, $1 \leq m \leq M, M \in \mathbb{N}$, of length $\tau_m := t_m - t_{m-1}$. We denote by Θ^m and ϕ^m approximations of Θ and ϕ at time t_m and discretize the time derivatives in (3.16a) and (3.17a) by the backward difference quotient: Given Θ^{m-1} and $\phi^{m-1}, 1 \leq m < M$, compute Θ^m and ϕ^m such that

$$\Theta^m - \Theta^{m-1} \in \tau_m M_\Theta \ \ - \partial_\Theta F_{11}(\Theta^m,\phi^m) + \frac{\delta F_{12}(\Theta^m,\phi^m)}{\delta\Theta^m} \quad \text{in } \Omega, \tag{4.1a}$$

$$\Theta^m = 0 \quad \text{on } \Gamma, \tag{4.1b}$$

and

$$\phi^m - \phi^{m-1} = \tau_m M_\phi \ \ - \frac{\delta F_{12}(\phi^m,\Theta^m)}{\delta\phi^m} + \frac{\delta F_{11}(\Theta^m,\phi^m)}{\delta\phi^m} \quad \text{in } \Omega, \tag{4.2a}$$

$$\phi^m = 0 \quad \text{on } \Gamma. \tag{4.2b}$$

The splitting scheme is such that we decouple (4.1) and (4.2) as follows:
In $F_1(\Theta^m,\phi^m)$ we replace ϕ^m by ϕ^{m-1} and compute Θ^m as the solution of the second order elliptic inclusion

$$\Theta^m - \Theta^{m-1} \in \tau_m M_\Theta \ \ - \partial_\Theta F_{11}(\Theta^m,\phi^{m-1}) + \frac{\delta F_{12}(\Theta^m,\phi^{m-1})}{\delta\Theta^m} \quad \text{in } \Omega, \tag{4.3a}$$

$$\Theta^m = 0 \quad \text{on } \Gamma. \tag{4.3b}$$

We use the computed Θ^m in $F_1(\phi^m,\Theta^m)$ and compute $\phi^m \in W_0^{1,2}(\Omega)$ as the weak solution of the second order elliptic differential equation

$$\phi^m - \phi^{m-1} = \tau_m M_\phi \ \ - \frac{\delta F_{12}(\phi^m,\Theta^m)}{\delta\phi^m} + \frac{\delta F_{11}(\Theta^m,\phi^m)}{\delta\phi^m} \quad \text{in } \Omega, \tag{4.4a}$$

$$\phi^m = 0 \quad \text{on } \Gamma. \tag{4.4b}$$

In view of Theorem 2.2 we define the functional

$$\begin{aligned} F_1^{m,\tau_m}(\Theta) &:= \frac{1}{2}\|\Theta^m - \Theta^{m-1}\|^2_{L^2(\Omega)} + \tau_m \ F_{11}^{m,\tau_m}(\Theta) + F_{12}^{m,\tau_m}(\Theta) \ , \\ F_{11}^{m,\tau_m}(\Theta) &:= \frac{1}{2}\int_\Omega s(\nabla\phi^{m-1},\Theta)^2 \ |\nabla\phi^{m-1}|^2 \ dx, \\ F_{12}^{m,\tau_m}(\Theta) &:= \frac{H}{2\xi_0} \ \int_\Omega f_{ori,c}(\omega(\phi^{m-1}),\nabla\Theta) \ dx + \int_\Omega f^\infty_{ori,c}(\omega(\phi^{m-1}),\frac{D^s\Theta}{|D^s\Theta|}) \ d|D^s\Theta| \ , \end{aligned} \tag{4.5}$$

and consider the unconstrained minimization problem

$$F_1^{m,\tau_m}(\Theta^m) = \inf_{\Theta\in BV_0(\Omega)} F_1^{m,\tau_m}(\Theta). \tag{4.6}$$

We note that except for the second term on the right-hand side of $F_{12}^{m,\tau_m}(\Theta)$ the boundary value problem (4.4a), (4.4b) is the necessary and sufficient optimality condition for the unconstrained minimization problem (4.6).

Theorem 4.1. *The minimization problem* (4.6) *has a solution* $\Theta^m \in BV_0(\Omega)$.

Proof. Let $\{\Theta_n\}_\mathbb{N}, \Theta_n \in BV_0(\Omega), n \in \mathbb{N}$, be a minimizing sequence, i.e.,

$$F_1^{m,\tau_m}(\Theta_n) \to \inf_{\Theta\in BV(\Omega)} F_1^{m,\tau_m}(\Theta) \quad (n \to \infty). \tag{4.7}$$

Due to the coercivity of the first part of F_1^{m,τ_m} and of F_{11}^{m,τ_m} the sequence is bounded in $L^2(\Omega)$ and $BV_0(\Omega;\omega(\phi^{m-1}))$ and hence, according to section 2 there exist a subsequence $\mathbb{N}' \subset \mathbb{N}$ and $\Theta^m \in BV_0(\Omega)$ such that

$$\Theta_n \rightharpoonup \Theta^m \quad (\mathbb{N}' \ni n \to \infty) \text{ in } L^2(\Omega), \tag{4.8a}$$

$$\Theta_n \rightharpoonup^* \Theta^m \quad (\mathbb{N}' \ni n \to \infty) \text{ in } BV_0(\Omega). \tag{4.8b}$$

The weak lower semicontinuity of the convex functional $\frac{1}{2}\|\Theta - \Theta^{m-1}\|^2_{L^2(\Omega)}$ in Θ and (4.8a) imply

$$\frac{1}{2}\|\Theta^m - \Theta^{m-1}\|^2_{L^2(\Omega)} \leq \lim \inf_{\mathbb{N}'\ni n\to\infty} \frac{1}{2}\|\Theta_n - \Theta^{m-1}\|^2_{L^2(\Omega)}. \tag{4.9}$$

Moreover, the weak* lower semicontinuity of F_{11}^{m,τ_m} and (4.8b) in conjunction with Theorem 2.2 imply

$$F_{11}^{m,\tau_m}(\Theta^m) \leq \lim \inf_{\mathbb{N}'\ni n\to\infty} F_{11}^{m,\tau_m}(\Theta_n). \tag{4.10}$$

Finally, due to (4.8b) we have $\Theta_n \to \Theta^m$ in $L^1(\Omega)$ as $\mathbb{N}' \ni n \to \infty$. Passing to a subsequence $\mathbb{N}'' \subset \mathbb{N}'$, it follows that

$$\Theta_n \to \Theta^m \quad \text{almost everywhere in } \Omega \text{ as } \mathbb{N}'' \ni n \to \infty.$$

Due to the continuity of the anisotropy function s in Θ it follows that

$$s(\nabla\phi^{m-1}, \Theta_n)^2 \to s(\nabla\phi^{m-1}, \Theta^m) \quad \text{almost everywhere in } \Omega \text{ as } \mathbb{N}'' \ni n \to \infty.$$

The sequence $\{s(\nabla\phi^{m-1}, \Theta_n)^2|\nabla\phi^{m-1}|^2\}_{n\in\mathbb{N}''}$ is uniformly integrable and

$$s(\nabla\phi^{m-1}, \Theta^m)^2|\nabla\phi^{m-1}|^2 \in L^1(\Omega).$$

The Vitali convergence theorem (cf., e.g., [18]) yields

$$F_{12}(\Theta^m, \phi^{m-1}) = \lim_{\mathbb{N}''\ni n\to\infty} F_{12}(\Theta_n, \phi^{m-1}). \tag{4.11}$$

Combining (4.9), (4.10), and (4.11) we obtain

$$F_1(\Theta^m) \leq \lim \inf_{\mathbb{N}'\ni n\to\infty} F_1^{m,\tau_m}(\Theta_n)$$

which together with (4.7) shows that Θ^m satisfies (4.6).
Next, we consider the energy functional

$$F_2^{m,\tau_m}(\phi) := \frac{1}{2}\, \|\phi - \phi^{m-1}\|^2_{L^2(\Omega)} + \tau_m F_2(\phi, \Theta^m), \tag{4.12}$$

$$F_2(\phi, \Theta^m) := \frac{M_\phi}{2} \int_\Omega s(\nabla\phi, \Theta^m)^2\, |\nabla\phi|^2 g(\phi) + \frac{H}{2\xi_0}\, f_{ori,c}(\omega(\phi), \nabla\Theta^m)\;\; dx.$$

Theorem 4.2. *For sufficiently small $s_0 > 0$, the energy functional $F_2^{m,\tau_m} : W_0^{1,2}(\Omega) \to \mathbb{R}$ has a local minimizer $\phi^m \in W_0^{1,2}(\Omega)$, i.e.,*

$$F_2^{m,\tau_m}(\phi^m) = \inf_{\phi\in W_0^{1,2}(\Omega)} F_2^{m,\tau_m}(\phi). \tag{4.13}$$

Proof. We first show that the functional F_2^{m,τ_m} is coercive on $W^{1,2}(\Omega)$: By Young's inequality we find

$$\frac{1}{2}\, \|\phi - \phi^{m-1}\|^2_{0,\Omega} \geq \frac{1}{4}\, \|\phi\|^2_{0,\Omega} - \frac{1}{2}\, \|\phi^{m-1}\|^2_{0,\Omega}. \tag{4.14}$$

Further, we take advantage of (3.3) to conclude

$$F_2^{m,\tau_m}(\phi) \geq M_\phi \varepsilon_r (1-s_0)^2 \tau_m \|\nabla\phi\|_{0,\Omega}^2 + \frac{1}{4}\|\phi\|_{0,\Omega}^2 - \frac{1}{2}\ \|\phi^{m-1}\|_{0,\Omega}^2. \tag{4.15}$$

The functional F_2^{m,τ_m} is not convex in ϕ, but it can be split into a convex part $F_{2,1}^{m,\tau_m}$ and non-convex part $F_{2,2}^{m,\tau_m}$ according to

$$F_{2,1}^{m,\tau_m}(\phi) := \frac{1}{2}\ \|\phi - \phi^{m-1}\|_{0,\Omega}^2 + \tau_m M_\phi \int_\Omega s(\nabla\phi, \Theta^m)^2 |\nabla\phi|^2 \, dx,$$

$$F_{2,2}^{m,\tau_m}(\phi) := M_\phi \int_\Omega \ g(\phi) + \frac{H}{2\xi_0}\ f_{ori,c}(\omega(\phi), \nabla\Theta^m) \ \ dx.$$

The convexity of the first part $\|\phi - \phi^{m-1}\|_{0,\Omega}^2/2$ of $F_{2,1}^{m,\tau_m}$ in ϕ is obvious. For sufficiently small s_0 the convexity of the second part has been shown in [13].
In order to prove the existence of a local minimizer let $\{\phi_n\}_{\mathbb{N}}, \phi_n \in W_0^{1,2}(\Omega)$, be a minimizing sequence, i.e., it holds

$$F_2^{m,\tau_m}(\phi_n) \to \inf_{\phi \in W_0^{1,2}(\Omega)} F_2^{m,\tau_m}(\phi) \quad (n \to \infty). \tag{4.16}$$

Due to the coercivity of F_2^{m,τ_m} the sequence $\{\phi_n\}_{\mathbb{N}}$ is bounded in $W_0^{1,2}(\Omega)$. Hence, there exists a weakly convergent subsequence, i.e., there exist $\mathbb{N}' \subset \mathbb{N}$ and $\phi^m \in W_0^{1,2}(\Omega)$ such that $\phi_n \rightharpoonup \phi^m$ ($\mathbb{N}' \ni n \to \infty$) in $W_0^{1,2}(\Omega)$. The Rellich-Kondrachev theorem implies strong convergence in $L^p(\Omega)$ for any $1 \leq p < \infty$ and hence, for some subsequence $\mathbb{N}'' \subset \mathbb{N}'$ we have

$$\phi_n \to \phi^m \quad \text{almost everywhere in } \Omega \text{ as } \mathbb{N}'' \ni n \to \infty.$$

Due to the continuity of g and ω, we also have

$$g(\phi_n) \to g(\phi^m) \quad \text{almost everywhere in } \Omega \text{ as } \mathbb{N}'' \ni n \to \infty,$$
$$\omega(\phi_n) \to \omega(\phi^m) \quad \text{almost everywhere in } \Omega \text{ as } \mathbb{N}'' \ni n \to \infty.$$

Moreover, the sequence $\{M_\phi \ g(\phi_n) + \frac{H}{2\xi_0} f_{ori,c}(\omega(\phi_n), \nabla\Theta^m) \ \}_{n\in\mathbb{N}''}$ is uniformly integrable and

$$M_\phi \ g(\phi^m) + \frac{H}{2\xi_0} f_{ori,c}(\omega(\phi^m), \nabla\Theta^m) \ \in L^1(\Omega).$$

Again, the Vitali convergence theorem implies

$$F_{2,2}^{m,\tau_m}(\phi_n) \to F_{2,2}^{m,\tau_m}(\phi^m) \quad \text{as } \mathbb{N}' \ni n \to \infty. \tag{4.17}$$

Obviously, the functional $F_{2,1}^{m,\tau_m}$ is continuous on $W_0^{1,2}(\Omega)$ and thus lower semicontinuous. As we have shown before, it is convex and hence, it is weakly lower semicontinuous. This gives

$$F_{2,1}^{m,\tau_m}(\phi^m) \leq \lim \inf_{\mathbb{N}' \ni n \to \infty} F_{2,1}^{m,\tau_m}(\phi_n). \tag{4.18}$$

Now, (4.16), (4.17), and (4.18) imply that (4.13) holds true.

5. Discretization in space and numerical solution of the fully discretized system

For discretization in space of the implicitly in time discretized and split system (4.3), (4.4) we assume $\mathcal{T}_h(\Omega)$ to be a geometrically conforming, shape regular, simplicial triangulation of the computational domain Ω. Denoting by $P_k(K), k \in \mathbb{N}, K \in \mathcal{T}_h(\Omega)$, the linear space of polynomials of degree $\leq k$ on K, we refer to

$$V_h := \{v_h \in C_0(\Omega) \mid v_h|_K \in P_k(K), K \in \mathcal{T}_h(\Omega)\}$$

as the finite element space of continuous piecewise polynomial Lagrangian finite elements (cf., e.g., [3]). Moreover, in order to avoid dealing with a discrete variational inequality, we replace the multivalued subdifferential $\tilde{f} := \partial_{\nabla\Theta} f_{ori,c}$ of the convexified orientational free energy density by its single-valued Moreau-Yosida approximation $\tilde{f}_{\varepsilon_R}$ with regularization parameter $\varepsilon_R > 0$ (cf., e.g., [6]). Then, the finite element approximation of (4.3),(4.4) reads as follows: Given ϕ_h^{m-1}, find $\Theta_h^m, \phi_h^m \in V_h$ such that for all $v_h \in V_h$ and $w_h \in V_h$ it holds

$$\int_\Omega \Theta_h^m v_h \, dx + \tau_m \frac{H}{2\xi_0} M_\Theta \int_\Omega \tilde{f}_{\varepsilon_R}(\omega(\phi_h^{m-1}), \nabla\Theta_h^m) \frac{\nabla\Theta_h^m}{|\nabla\Theta_h^m|} \cdot \nabla v_h \, dx - \tag{5.1a}$$

$$\tau_m M_\Theta \int_\Omega s(\nabla\phi_h^{m-1}, \Theta_h^m) \frac{\partial s(\nabla\phi_h^{m-1}, \Theta_h^m)}{\partial\Theta} |\nabla\phi_h^{m-1}|^2 v_h \, dx = \int_\Omega \Theta_h^{m-1} v_h \, dx,$$

$$\int_\Omega \phi_h^m w_h \, dx + \tau_m M_\phi \int_\Omega a(\nabla\phi_h^m, \Theta_h^m) \nabla\phi_h^m \cdot \nabla w_h \, dx - \tag{5.1b}$$

$$\tau_m M_\phi \int_\Omega \left(g_\phi(\phi_h^m) + \frac{H}{2\xi_0} \frac{\partial f_{ori,c}(\phi_h^m, \nabla\Theta_h^m)}{\partial\omega(\phi)} \omega_\phi(\phi_h^m) \right) w_h \, dx = \int_\Omega \phi_h^{m-1} w_h \, dx.$$

By similar arguments as in the previous section it can be shown that (5.1a) and (5.1b) admit unique solutions $\Theta_h^m \in V_h$ and $\phi_h^m \in V_h$. The numerical solution of (5.1a) and (5.1b) amounts to the successive solution of two nonlinear algebraic systems. We assume $V_h = \text{span}\{\varphi_1, \cdots, \varphi_{N_h}\}, N_h \in \mathbb{N}$, such that

$$\Theta_h^m = \sum_{j=1}^{N_h} \Theta_j^m \varphi_j, \quad \phi_h^m = \sum_{j=1}^{N_h} \phi_j^m \varphi_j.$$

Setting $\mathbf{\Theta}^m := (\Theta_1^m, \cdots, \Theta_{N_h}^m)^T$ and $\mathbf{\Phi}^m := (\phi_1^m, \cdots, \phi_{N_h}^m)^T$, the algebraic formulation of (5.1a) and (5.1b) leads to the two nonlinear systems

$$\mathbf{F}_1(\mathbf{\Theta}^m, \mathbf{\Phi}^{m-1}, t_m) = \mathbf{0}, \tag{5.2a}$$

$$\mathbf{F}_2(\mathbf{\Theta}^m, \mathbf{\Phi}^m, t_m) = \mathbf{0}. \tag{5.2b}$$

Here, $\mathbf{F}_k : \mathbb{R}^{N_h} \times \mathbb{R}^{N_h} \times \mathbb{R}_+ \to \mathbb{R}^{N_h}$ and the components $\mathbf{F}_{k,i}$, $1 \le i \le N_h$, are given by

$$\mathbf{F}_{1,i}(\mathbf{\Theta}^m, \mathbf{\Phi}^{m-1}, t_m) = \sum_{j=1}^{N_h} \Theta_j^m \int_\Omega \varphi_i \varphi_j \, dx +$$

$$\tau_m \frac{H}{2\xi_0} M_\Theta \sum_{j=1}^{N_h} \Theta_j^m \int_\Omega \tilde{f}_{\varepsilon_R}(\omega(\mathbf{\Phi}^{m-1}), \sum_{k=1}^{N_h} \Theta_k^m \nabla\varphi_k) |\sum_{k=1}^{N_h} \Theta_k^m \nabla\varphi_k)|^{-1} \nabla\varphi_j \cdot \nabla\varphi_i \, dx$$

$$- \tau_m M_\Theta \int_\Omega s(\mathbf{\Phi}^{m-1}, \mathbf{\Theta}^m) s_\Theta(\mathbf{\Phi}^{m-1}, \mathbf{\Theta}^m) |\sum_{k=1}^{N_h} \phi_k^{m-1} \nabla\varphi_k|^2 \varphi_i \, dx -$$

$$\sum_{j=1}^{N_h} \Theta_j^{m-1} \int_\Omega \varphi_i \varphi_j \, dx$$

and

$$\mathbf{F}_{2,i}(\mathbf{\Theta}^m, \mathbf{\Phi}^m, t_m) = \sum_{j=1}^{N_h} \phi_j^m \int_\Omega \varphi_i \varphi_j \, dx +$$

$$\tau_m M\phi \sum_{j=1}^{N_h} \phi_j^m \int_\Omega a(\mathbf{\Phi}^m, \mathbf{\Theta}^m) \nabla \varphi_j \cdot \nabla \varphi_i \, dx -$$

$$\tau_m M_\phi \int_\Omega \; g_\phi(\mathbf{\Phi}^m) + \frac{H}{2\xi_0} \frac{\partial f_{ori,c}(\mathbf{\Phi}^m, \sum_{k=1}^{N_h} \Theta_k^m \nabla \varphi_k)}{\partial \omega(\phi)} \omega_\phi(\mathbf{\Phi}^m) \; \varphi_j \, dx -$$

$$\sum_{j=1}^{N_h} \phi_j^{m-1} \int_\Omega \varphi_i \varphi_j \, dx,$$

where

$$\omega(\mathbf{\Phi}^{m-1}) := \omega(\sum_{k=1}^{N_h} \phi_k^{m-1} \varphi_k), \; , \omega_\phi(\mathbf{\Phi}^m) := \omega_\phi(\sum_{k=1}^{N_h} \phi_k^m \varphi_k)$$

$$a(\mathbf{\Phi}^m, \mathbf{\Theta}^m) := a(\sum_{k=1}^{N_h} \phi_k^m \nabla \varphi_k, \sum_{k=1}^{N_h} \Theta_k^m \varphi_k),$$

$$s(\mathbf{\Phi}^{m-1}, \mathbf{\Theta}^m) := s(\sum_{k=1}^{N_h} \phi_k^{m-1} \varphi_k, \sum_{k=1}^{N_h} \Theta_k^m \varphi_k),$$

$$s_\Theta(\mathbf{\Phi}^{m-1}, \mathbf{\Theta}^m) := s_\Theta(\sum_{k=1}^{N_h} \phi_k^{m-1} \varphi_k, \sum_{k=1}^{N_h} \Theta_k^m \varphi_k).$$

We note that $\mathbf{F}_1$ is not differentiable in $\mathbf{\Theta}^m$ in the classical sense, but admits a generalized Jacobian $\partial_{\mathbf{\Theta}} \mathbf{F}_1$ in the sense of Clarke [4]. Hence, the nonlinear system (5.2a) can be solved by a semismooth Newton method (cf., e.g., [11]), whereas (5.2b) can be solved by the classical Newton method involving the Jacobian $\mathbf{F}_2'$. In both cases, the problem is the appropriate choice of the time step sizes $\tau_m, 1 \leq m \leq M$, in order to guarantee convergence. In fact, a uniform choice $\tau_m = T/M$ only works, if M is chosen sufficiently large which would require an unnecessary huge amount of time steps. An appropriate way to overcome this difficulty is to consider (5.2a), (5.2b) as parameter dependent nonlinear systems with the time as a parameter and to apply a predictor-corrector continuation strategy with an adaptive choice of the time steps (cf., e.g., [5, 12, 13, 14]). Given the pair $(\mathbf{\Theta}^{m-1}, \mathbf{\Phi}^{m-1})$, the time step size $\tau_{m-1,0} = \tau_{m-1}$, and setting $k = 0$, where k is a counter for the predictor-corrector steps, the predictor step for (5.2a) consists of constant continuation leading to the initial guesses

$$\mathbf{\Theta}^{(m,k)} = \mathbf{\Theta}^{m-1}, \quad t_m = t_{m-1} + \tau_{m-1,k}. \tag{5.3}$$

Setting $\nu_1 = 0$ and $\mathbf{\Theta}^{(m,k,\nu_1)} = \mathbf{\Theta}^{(m,k)}$, for $\nu_1 \leq \nu_{max}$, where $\nu_{max} > 0$ is a pre-specified maximal number, the semismooth Newton iteration

$$\begin{aligned} \partial_{\mathbf{\Theta}} \mathbf{F}_1(\mathbf{\Theta}^{(m,k,\nu_1)}, \mathbf{\Phi}^{m-1}, t_m) \Delta \mathbf{\Theta}^{(m,k,\nu_1)} &\ni -\mathbf{F}_1(\mathbf{\Theta}^{(m,k,\nu_1)}, \mathbf{\Phi}^{m-1}, t_m), \\ \mathbf{\Theta}^{(m,k,\nu_1+1)} &= \mathbf{\Theta}^{(m,k,\nu_1)} + \Delta \mathbf{\Theta}^{(m,k,\nu_1)}, \quad \nu_1 \geq 0, \end{aligned} \tag{5.4}$$

serves as a corrector whose convergence is monitored by the contraction factor

$$\Lambda_\Theta^{(m,k,\nu_1)} = \frac{\|\overline{\Delta \mathbf{\Theta}^{(m,k,\nu_1)}}\|}{\|\Delta \mathbf{\Theta}^{(m,k,\nu_1)}\|}, \tag{5.5}$$

where $\overline{\boldsymbol{\Delta\Theta}^{(m,k,\nu_1)}}$ is the solution of the auxiliary Newton step

$$\partial_{\boldsymbol{\Theta}}\mathbf{F}_1(\boldsymbol{\Theta}^{(m,k,\nu_1)},\boldsymbol{\Phi}^{m-1},t_m)\overline{\boldsymbol{\Delta\Theta}^{(m,k,\nu_1)}} \ni -\mathbf{F}_1(\boldsymbol{\Theta}^{(m,k,\nu_1+1)},\boldsymbol{\Phi}^{m-1},t_m). \tag{5.6}$$

If the contraction factor satisfies

$$\Lambda_\Theta^{(m,k,\nu_1)} < \frac{1}{2}, \tag{5.7}$$

we set $\nu_1 = \nu_1 + 1$. If $\nu_1 > \nu_{max}$, both the Newton iteration and the predictor-corrector continuation strategy are terminated indicating non-convergence. Otherwise, we continue the semismooth Newton iteration (5.4). If (5.7) does not hold true, we set $k = k + 1$ and the time step is reduced according to

$$\tau_{m,k} = \max\Big(\frac{\sqrt{2}-1}{\sqrt{4\Lambda_\Theta^{(m,k,\nu_1)}+1}-1}\,\tau_{m,k-1},\tau_{min}\Big), \tag{5.8}$$

where $\tau_{min} > 0$ is some pre-specified minimal time step. If $\tau_{m,k} > \tau_{min}$, we go back to the prediction step (5.3). Otherwise, the predictor-corrector strategy is stopped indicating non-convergence. The semismooth Newton iteration is terminated successfully, if for some $\nu_1^* > 0$ the relative error of two subsequent semismooth Newton iterates satisfies

$$\frac{\|\boldsymbol{\Theta}^{(m,k,\nu_1^*)}-\boldsymbol{\Theta}^{(m,k,\nu_1^*-1)}\|}{\|\boldsymbol{\Theta}^{(m,k,\nu_1^*)}\|} < \varepsilon \tag{5.9}$$

for some pre-specified accuracy $\varepsilon > 0$. In this case, we proceed with the prediction step (5.10) below.
The predictor step for (5.2b) also consists of constant continuation leading to the initial guesses

$$\boldsymbol{\Phi}^{(m,k)} = \boldsymbol{\Phi}^{m-1}, \quad t_m = t_{m-1} + \tau_{m-1,k}. \tag{5.10}$$

Setting $\nu_2 = 0$ and $\boldsymbol{\Phi}^{(m,k,\nu_2)} = \boldsymbol{\Phi}^{(m,k)}$, for $\nu_2 \le \nu_{max}$, the Newton iteration

$$\begin{aligned}\mathbf{F}_2'(\boldsymbol{\Theta}^{(m,k,\nu_1^*)},\boldsymbol{\Phi}^{m,k,\nu_2},t_m)\boldsymbol{\Delta\Phi}^{(m,k,\nu_2)} &= -\mathbf{F}_2(\boldsymbol{\Theta}^{(m,k,\nu_1^*)},\boldsymbol{\Phi}^{m,k,\nu_2},t_m),\\ \boldsymbol{\Phi}^{(m,k,\nu_2+1)} &= \boldsymbol{\Phi}^{(m,k,\nu_2)} + \boldsymbol{\Delta\Phi}^{(m,k,\nu_2)}, \quad \nu_2 \ge 0,\end{aligned} \tag{5.11}$$

again serves as the corrector with the convergence monitored by the contraction factor

$$\Lambda_\phi^{(m,k,\nu_2)} = \frac{\|\overline{\boldsymbol{\Delta\Phi}^{(m,k,\nu_2)}}\|}{\|\boldsymbol{\Delta\Phi}^{(m,k,\nu_2)}\|}, \tag{5.12}$$

where $\overline{\boldsymbol{\Delta\Phi}^{(m,k,\nu_2)}}$ is the solution of the auxiliary Newton step

$$\mathbf{F}_2'(\boldsymbol{\Theta}^{(m,k,\nu_1^*)},\boldsymbol{\Phi}^{m,k,\nu_2},t_m)\overline{\boldsymbol{\Delta\Phi}^{(m,k,\nu_2)}} = -\mathbf{F}_2(\boldsymbol{\Theta}^{(m,k,\nu_1^*)},\boldsymbol{\Phi}^{m,k,\nu_2+1},t_m). \tag{5.13}$$

If the contraction factor satisfies

$$\Lambda_\phi^{(m,k,\nu_2)} < \frac{1}{2}, \tag{5.14}$$

we set $\nu_2 = \nu_2 + 1$. If $\nu_2 > \nu_{max}$, both the Newton iteration and the predictor-corrector continuation strategy are terminated indicating non-convergence. Otherwise, we continue the Newton iteration (5.11). If (5.14) is not satisfied, we set $k = k + 1$ and the time step is reduced according to

$$\tau_{m,k} = \max\Big(\frac{\sqrt{2}-1}{\sqrt{4\Lambda_\phi^{(m,\nu_2)}+1}-1}\,\tau_{m,k-1},\tau_{min}\Big). \tag{5.15}$$

If $\tau_{m,k} > \tau_{min}$, we go back to the prediction step (5.3) for (5.2a). Otherwise, the predictor-corrector strategy is stopped indicating non-convergence. The Newton iteration is terminated successfully, if for some $\nu_2^* > 0$ the relative error of two subsequent Newton iterates satisfies

$$\frac{\|\boldsymbol{\Phi}^{(m,k,\nu_2^*)} - \boldsymbol{\Phi}^{(m,k,\nu_2^*-1)}\|}{\|\boldsymbol{\Theta}^{(m,k,\nu_2^*)}\|} < \varepsilon_T. \tag{5.16}$$

In this case, we set

$$\boldsymbol{\Theta}^m = \boldsymbol{\Theta}^{(m,k,\nu_1^*)}, \quad \boldsymbol{\Phi}^m = \boldsymbol{\Phi}^{(m,k,\nu_2^*)} \tag{5.17}$$

and predict a new time step according to

$$\tau_m = \min\left(\frac{(\sqrt{2}-1)\,\|\Delta\boldsymbol{\Theta}^{(m,k,0)}\|}{2\Lambda_\Theta^{(m,k,0)}\,\|\boldsymbol{\Theta}^{(m,k,0)} - \boldsymbol{\Theta}^m\|}, \frac{(\sqrt{2}-1)\,\|\Delta\boldsymbol{\Phi}^{(m,k,0)}\|}{2\Lambda_\phi^{(m,k,0)}\,\|\boldsymbol{\Phi}^{(m,k,0)} - \boldsymbol{\Phi}^m\|}, \text{amp}\;\; \tau_{m,k}\right), \tag{5.18}$$

where amp > 1 is a pre-specified amplification factor for the time step sizes. We set $m = m + 1$ and begin new predictor-corrector iterations for the time interval $[t_m, t_{m+1}]$.

6. Numerical results

We consider the formation of a Category 1 spherulite based on the phase field model specified in section 3 and the splitting method described in sections 4 and 5. The physical data for the phase field model are depicted in Table 1 and Table 2. In particular, the constants r_1 and r_2 for the orientational free energy density are chosen such that the angle of misorientation is 30^o leading to six preferred orientations.

M_Θ	M_ϕ	s_0	m_S	ε_r
$1.1 \cdot 10^1$	$1.5 \cdot 10^2$	0.2	2	$1.0 \cdot 10^{-3}$

Table 1. Physical data: Mobilities M_Θ, M_ϕ, modulus of anisotropy s_0, symmetry index m_S, parameter ε_r in the interpolation function ω.

H	ξ_0	r_1	r_2	δ
$1.0 \cdot 10^{-3}$	$2.1 \cdot 10^{-4}$	3.0	0.5	0.2

Table 2. Physical data: Free energy H of low grain boundaries, correlation length ξ_0 of the orientational field, constants r_1, r_2, δ determining the orientational free energy density f_{ori}.

h	ε_R	ν_{max}	τ_{min}	ε_T	amp
$5.68 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$	50	$1.0 \cdot 10^{-6}$	$1.0 \cdot 10^{-3}$	1.2

Table 3. Computational data for the spatial discretization and the predictor-corrector continuation strategy: mesh width h for the triangulation of the computational domain, regularization parameter ε_R for the Moreau-Yosida regularization of the orientational free energy density, maximum number ν_{max} of semismooth Newton iterations, minimum time step size τ_{min}, relative accuracy ε_T of semismooth Newton iterations, and amplification factor amp for new time step size.

The two-dimensional computational domain is $\Omega = (0\,\mu m, 6\,\mu m)^2$ which has been discretized by a uniform geometrically conforming simplicial triangulation with right isosceles of mesh width h. The computational data for the spatial discretization and the predictor-corrector continuation strategy are contained in Table 3.

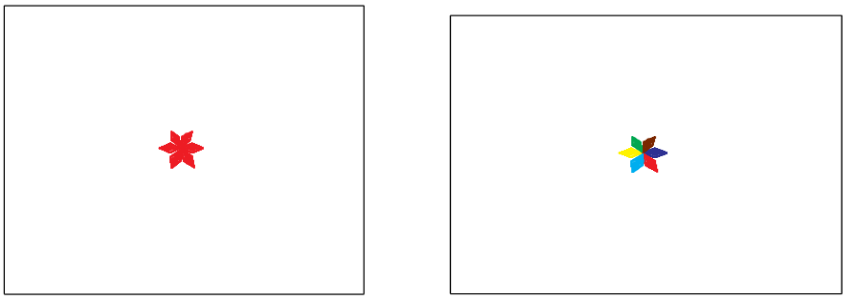

Figure 3. Formation of a spherulite at time $t = 4.3 \cdot 10^{-3}$ s. Left: Local degree of crystallinity ϕ_0 ('Red' indicates $\phi_0 = 1.0$). Right: Orientation angle Θ_0. The colors are explained in the narrative. The colors can be viewed in the online issue of the journal, which is available at the journal's website.

We consider the formation of a Category 1 spherulite from a nucleation site which is initially occupying a subdomain Ω_0 around the center of the computational domain Ω. The initial data are given by $\phi_0 = 1.0$ in Ω_0 and $\phi = 0.0$ elsewhere and by Θ_0 varying between 0.7π and 1.2π in Ω_0 and chosen randomly around 0.95π elsewhere. In particular, the assignment of the colors in Figures 3 and 4 is as follows: Blue (1.2π), Brown (1.0π), Cyan (0.9π), Green (0.8π), Red (1.1π), and Yellow (0.7π).

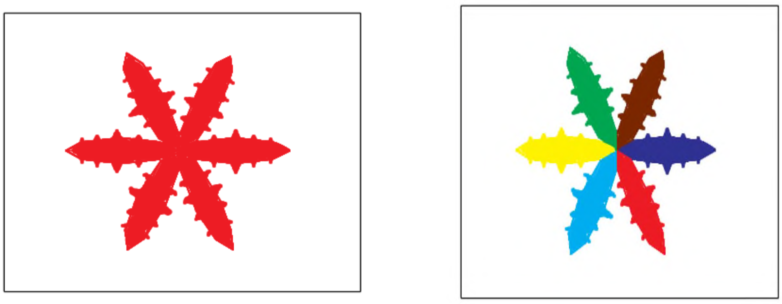

Figure 4. Formation of a spherulite at time $t = 1.3 \cdot 10^{-2}$ s. Left: Local degree of crystallinity. Right: Orientation angle. The colors are explained in the narrative. The colors can be viewed in the online issue of the journal, which is available at the journal's website.

The spherulite grows radially from the nucleation site and exhibits crystalline branching as can be seen from Figure 4 which shows the local degree of crystallinity and the orientation angle at time $t = 1.3 \cdot 10^{-2}$ s.
Finally, Figure 5 displays the history of the predictor-corrector strategy where the adaptively chosen time steps are shown as a function of the number of iterations. We observe large fluctuations in the time steps which are due the occurrence of very steep gradients at the growing front, particularly when crystalline branching takes place.

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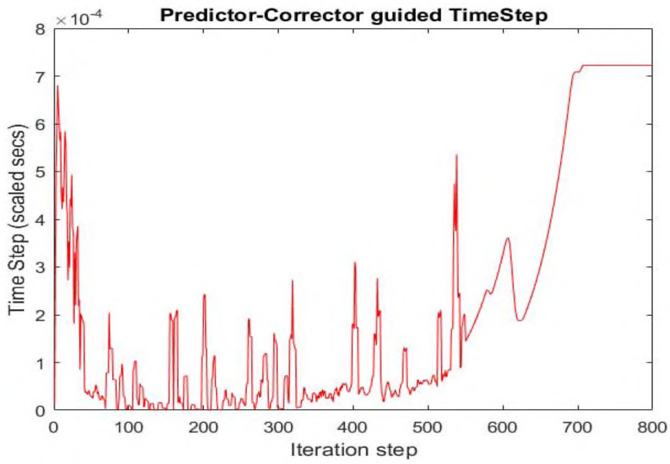

Figure 5. History of the predictor-corrector strategy with the adaptively chosen time steps as a function of the number of iterations.

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