Phase field approach with anisotropic interface energy and interface stresses: large strain formulation

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Abstract

A thermodynamically consistent, large-strain, multi-phase field approach (with consequent interface stresses) is generalized for the case with anisotropic interface (gradient) energy (e.g. an energy density that depends both on the magnitude and direction of the gradients in the phase fields). Such a generalization, if done in the “usual” manner, yields a theory that can be shown to be manifestly unphysical. These theories consider the gradient energy as anisotropic in the *deformed* configuration, and, due to this supposition, several fundamental contradictions arise. First, the Cauchy stress tensor is non-symmetric and, consequently, violates the moment of momentum principle, in essence the Herring (thermodynamic) torque is imparting an unphysical angular momentum to the system. In addition, this non-symmetric stress implies a violation of the principle of material objectivity. These problems in the formulation can be resolved by insisting that the gradient energy is an isotropic function of the gradient of the order parameters in the *deformed* configuration, but depends on the direction of the gradient of the order parameters (is anisotropic) in the *undeformed* configuration. We find that for a propagating nonequilibrium interface, the structural part of the interfacial Cauchy stress is symmetric and reduces to a biaxial tension with the magnitude

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equal to the temperature- and orientation-dependent interface energy. Ginzburg-Landau equations for the evolution of the order parameters and temperature evolution equation, as well as the boundary conditions for the order parameters are derived. Small strain simplifications are presented. Remarkably, this anisotropy yields a *first order* correction in the Ginzburg-Landau equation for small strains, which has been neglected in prior works. The next strain-related term is third order. For concreteness, specific orientation dependencies of the gradient energy coefficients are examined, using published molecular dynamics studies of cubic crystals. In order to consider a fully specified system, a typical sixth order polynomial phase field model is considered. Analytical solutions for the propagating interface and critical nucleus are found, accounting for the influence of the anisotropic gradient energy and elucidating the distribution of components of interface stresses. The orientation-dependence of the nonequilibrium interface energy is first suitably defined and explicitly determined analytically, and the associated width is also found. The developed formalism is applicable to melting/solidification and crystal-amorphous transformation and can be generalized for martensitic and diffusive phase transformations, twinning, fracture, and grain growth, for which interface energy depends on interface orientation of crystals from either side.

Keywords:

Phase field approach, Phase transformation, Large strains, Anisotropic interface energy and interface stresses

1. Introduction

Phase field approach. Ginzburg-Landau, or phase field, approaches are routinely used to simulate various structural changes, including first-order solid-solid phase transformations (PTs) (Artemev and Khachaturyan (2001); Chen (2002); Finel et al. (2010); Jin et al. (2001a); Levitas et al. (2004); Levitas and Lee (2007); Vedantam and Abeyaratne (2005)), melting (Anderson et al. (2001); Slutsker et al. (2006); Wheeler and McFadden (1997)), and

also the evolution of multigrain structures (Kobayashi et al. (1998)), as well as twinning (Clayton and Knap (2011a,b); Hildebrand and Miehe (2012); Levitas et al. (2009, 2013); Levitas and Roy (2015)). There are a number of books on the phase field approach (Provatas and Elder (2010); Salje (1991); Toledano and Dmitriev (1996); Toledano and Toledano (1998); Umantsev (2012)), but these, however, do not include any substantial treatment of mechanics. In phase field modeling, the central concept is the introduction of order parameters η_i that describe material instabilities during PTs in a continuous way. The energy density of the system depends on the strain tensor, temperature, the order parameters, and their gradients, which provides an energy penalty for the formation of interfaces. For a given strain and temperature, the energy density has as many minima in the order parameter space as there are accessible phases or structural states of system. The time evolution of the order parameters describes the evolution of a multi-connected microstructure. This evolution is obtained by the solution of the Ginzburg-Landau equations, which represent linear relationships between $\dot{\eta}_i$ and the generalized forces X_i thermodynamically conjugated to $\dot{\eta}_i$, and coupled to all equations derived through continuum thermomechanics. The phase field approach is computationally attractive because the interfaces between phases appear and evolve automatically as a result of the solution to the above mentioned equations obviating the need to develop special methods for tracking them. The main theoretical advantage of the phase field approach, in comparison with sharp interface approach, is that it contains information about all intermediate states between phases and corresponding energy barriers, as well as including stationary heterogeneous intermediate states such as critical nuclei. Before examining phase field models in detail, however, we now explore the classical ideas that frame the analysis of interface motion in systems under stress.

Interface energy and generalized forces. Classically, a sharp interface between two phases has a total interface energy $\Gamma = \int \gamma dA$ or, for a homogeneous γ , $\Gamma = \gamma A$, where A is the interface area in the deformed state at a point in time and γ is the interface energy per

Figure 1: A representation of the generalized forces acting at an interface. (a) Interface stresses, which represent biaxial tension and contribute directly to mechanics, i.e., to the momentum equation; (b) the driving force for the translational interface propagation X_Σ (the Eshelby driving force, shown conditionally as a vector) and for the interface reorientation $\mathbf{X}_k$ (Herring torque). Both X_Σ and $\mathbf{X}_k$ are thermodynamic (configurational) forces, which do not contribute directly to the mechanics of the system, namely, momentum balance and the moment of momentum principle. (c) Artificial nonsymmetric interface shear stresses, which appear in the previous theories and are eliminated in the current paper. They produce torque, which violates the moment of momentum principle.

unit area. For liquid-liquid and liquid-gas interfaces, γ is independent of the change in interface area and consequently interface strain. For interfaces that involve solid phases, γ may depend on the change in interface area due to interface strain. For an anisotropic interface energy we write $\gamma = \gamma(\mathbf{k})$, where $\mathbf{k}$ is the unit normal to the interface. An interface is subjected to the following generalized forces related to three different processes of its evolution.

(a) Generalized forces preventing interface contraction called interface stresses (Gibbs (1948)), which represent the biaxial tension within interface (Fig. (1a)). In the simplest case of small strain and isotropic stresses within interface, the magnitude $\bar{\sigma}^S$ of the interface stress is defined by equation,

$$\bar{\sigma}^S dA = d\Gamma = d(\gamma A) = (\gamma + A \partial \gamma / \partial A) dA \Rightarrow \bar{\sigma}^S = \gamma + A \partial \gamma / \partial A = \gamma + \partial \gamma / \partial \varepsilon_s = \bar{\sigma}_{st} + \bar{\sigma}_e^S, \quad (1)$$

where $d\varepsilon_s = dA/A$ is the increment of the interface strain and subscript st indicates the structural part of the interface stresses. This is the Shuttleworth equation (Shuttleworth (1950)), see also review by Fischer et. al. (2008). The interface stress consists of two parts, one, $\bar{\sigma}_{st} = \gamma$, is the same as for a liquid-liquid interface, and another, $\bar{\sigma}_e^S$, is due to elastic deformation of an interface. The later can be anisotropic, i.e., tensorial, in more general case. Note that the expressions with a “bar” above them, such as $\bar{\sigma}^S$, have units of force per unit length, and the notation is used to distinguish these quantities from actual stresses (i.e., force per area), which can be singular at the interface in the sharp interface limit.

This generalized force is mechanical in nature, i.e., it contributes to the momentum balance equation or, for statics, the mechanical equilibrium equation. If one considers a liquid film on a rectangular frame with one movable side of the length l , one has to apply mechanical force equal to $\bar{\sigma}_{st}l$ to keep it in equilibrium; otherwise, such an interface would shrink to zero size. In continuum theories with sharp interfaces, the singular part of the stress tensor corresponding to biaxial interface stresses is introduced at each point of the

interface. This quantity is taken into account in solution of mechanical problem and results, in particular, in accounting for the jump in normal stress across an interface.

(b) A driving force for translational interface motion is associated with the quantity

$$X_\Sigma = -\Delta G - 2\gamma\kappa_{av}, \quad (2)$$

where ΔG is the jump in the Gibbs energy across the interface and κ_{av} is the mean interface curvature. For simplicity, we assume here isotropic γ , which is sufficient for our goal. This is a thermodynamic driving force, which does not contribute explicitly to the equations of mechanics. In particular, if $X_\Sigma \neq 0$, mechanical force cannot equilibrate an interface and it will propagate through material. X_Σ is also called the configurational force acting on the interface, or Eshelby driving force (Eshelby (1951, 1956, 1970); Gurtin (2000); Maugin (1993, 1995); Šilhavý (1997)).

(c) A driving force for the interface reorientation emerges from the dependence of the interface energy on orientation, that is we define

$$\mathbf{X}_k = -\frac{\partial\gamma}{\partial\mathbf{k}}, \quad (3)$$

which is called the Herring torque (Herring (1951)). Similar to X_Σ , the Herring torque, despite the name, is also a thermodynamic (configurational) driving force, which does not affect directly mechanical equations. In particular, if $\mathbf{X}_k \neq 0$, a mechanical torque cannot equilibrate an interface and it will change its orientation with respect to the material until it reaches stationary orientation corresponding to the local energy minimum with respect to $\mathbf{k}$.

The key point here is that the Herring torque is *not a mechanical torque* localized at the interface, and thus has no influence on the arguments made developing the moment of momentum equations. This is similar to the fact that X_Σ is not a mechanical body force localized at the interface that should be introduced in momentum balance equations.

Interface stresses. For sharp interfaces, there have been significant efforts in formulating balance laws and constitutive equations for elastic interfaces, when interface energy depends on strains, see Cahn (1979); Gurtin and Murdoch (1975); Gurtin and Struthers (1990); Javili and Steinmann (2010); Nix and Gao (1998); Podstrigach and Povstenko (1985); Povstenko (1991); Šilhavý (2011), and review articles by Cammarata (2009); Duan et al. (2009); Fischer et al. (2008). Most of these studies do not consider PTs. The central challenge in sharp interface modeling of systems with PTs is whether the strong heterogeneities in the fields of properties, transformation and total strains, and stresses that manifest across an interface can be realistically modeled through simple constitutive equations in terms of parameters averaged across an interface, and neglecting their gradients. Complicating this effort are the large number of material parameters that are unknown for interfaces.

The phase field approach provides a significant physical finesse to the above challenges. Our paper (Levitas (2014b)) provides a reasonable starting point for the current work, as it develops the large strain phase field approach for multivariant martensitic PTs, twinning, and melting, accounting for thermodynamically consistent interface stresses. The thermodynamic potential in this theory is quite general, satisfying a large number of physical requirements, as is detailed in Levitas and Preston (2002a,b); Levitas et al. (2003); Levitas (2013a), in contrast to all other theories (Artemev and Khachaturyan (2001); Chen (2002); Clayton and Knap (2011a,b); Denoual et al. (2010); Finel et al. (2010); Hildebrand and Miehe (2012); Jin et al. (2001a); Levitas et al. (2004); Levitas and Lee (2007); Salje (1991); Vedantam and Abeyaratne (2005)). Specifically, these requirements allow for consistent and noncontradictory description of the thermodynamic equilibrium states, stress-strain curves, and all thermomechanical properties. Also, PTs criteria follow directly from consideration of thermomechanical instability conditions. The approach described in Levitas (2014b) generalizes earlier large strain theory and simulations for multivariant martensitic PTs (Levin et al. (2013); Levitas et al. (2009); Levitas (2013a)), both without interface stresses, and small

strain theory with interface stresses for martensitic PTs (Levitas and Javanbakht (2010, 2011a); Levitas (2013a,b)), as well as theories for melting (Levitas and Samani (2011a,b)). Additionally, the position of the Gibbs dividing surface, which is required for a strict definition of the nonequilibrium interface energy, is defined in Levitas (2014a,b). A more detailed literature review can be found in Levitas (2014b) of the influence of interface stresses and large strains on this wide range of PTs, as well as an analysis of the drawbacks of some previous approaches (Anderson et al. (2001); Fried and Grach (1997); Lowengrub and Truskinovsky (1998); Slutsker et al. (2006); Wheeler and McFadden (1997)). We explicitly mention large strains and interface stresses together, as it shown in Levitas and Javanbakht (2010); Levitas (2013a,b); Levitas (2014b) that in order to introduce interface stresses that represent biaxial tension of the same magnitude as the interface energy, one must introduce geometrically nonlinear features *even when strains are infinitesimally small*.

Approach in Levitas and Javanbakht (2010); Levitas (2013a,b); Levitas (2014b) utilizes gradient of the order parameters in the current (deformed) configuration $\nabla\eta_i$, which is one the of conditions to introduce correct interface stresses. This approach improves some earlier works (Anderson et al. (2000, 2001); Lowengrub and Truskinovsky (1998)) in which phase field is coupled to mechanics. In these treatments of liquid-liquid, liquid-vapor, and solid-melt PTs, the reference (undeformed) configuration is not considered, and all equations are naturally presented in the actual (deformed) configuration. As the interface energy depends on the gradient of the order parameter in the deformed state, $\nabla\eta$, some (but not all) contributions to the interface stresses appeared automatically. Solid was treated as a very viscous liquid in studying melting (Anderson et al. (2000, 2001)). **The same expression for interface stresses like in Anderson et al. (2000, 2001) was obtained earlier in Wheeler and McFadden (1997) for the case without straining, by applying Noether's theorem.** For a conserved order parameter (which will not be considered here), the elements of the stress

tensor related to the gradient of the mass density (order parameter) are a realization of the so-called the “Korteweg stress” (Truesdall and Noll (1965)).

For PTs in solids and twinning (Artemev and Khachaturyan (2001); Chen (2002); Clayton and Knap (2011a,b); Denoual et al. (2010); Finel et al. (2010); Hildebrand and Miehe (2012); Jin et al. (2001a); Levin et al. (2013); Levitas et al. (2004); Levitas and Lee (2007); Levitas (2013a); Salje (1991); Vedantam and Abeyaratne (2005)), interface stresses have largely been ignored until very recently. In most prior work, the gradient of the order parameter was evaluated either in the undeformed state or the difference between reference and actual configuration was not mentioned let alone taken into account.

Interface stresses have been included in such models in Levitas and Javanbakht (2010, 2011a); Levitas (2013a,b); Levitas (2014b). It was demonstrated in Levitas and Javanbakht (2010, 2011a) that in circumstances where the size of microstructural units (e.g., martensitic plates or twins) is particularly small (a few nanometers), as well as at plate tips, interface stresses can make an important contribution to the nucleation and evolution of nanostructures, the emergence of elastic stresses, and consequently, to PT thermodynamics, kinetics, and morphology.

A different approach to the interface stresses in solids was suggested in Fried and Grach (1997). That treatment is performed in the reference configuration and the gradient energy depends on the gradient of the order parameter in the undeformed state, $\nabla_0 \eta$. However, the gradient energy in Fried and Grach (1997) also depends on the strain along the interface, which results in additional gradient-dependent interface stresses. There were no attempts in Fried and Grach (1997) to make this theory consistent in the limit case with liquids, i.e., to obtain biaxial interface true stresses with a magnitude equal to the interface energy. Thus, the interface stresses do not have a structure shown in Eq.(1).

Also, we note that the prior literature does not acknowledge that even where no stresses are introduced through $\nabla \eta$ or a strain-dependence of the gradient energy, the heteroge-

neous distribution of the transformation strain and elastic moduli across a finite-width interface (it is omitted in Fried and Grach (1997)) generates large elastic interface stresses, see Levitas and Samani (2011a,b); Slutsker et al. (2006) for a solid-melt interface. The magnitude of these stresses is much higher than those found in molecular dynamics simulations (Frolov and Mishin (2010a,b)). These internal elastic stresses suppress the melting of nanoparticles even above the bulk melting temperature, contradicting experiments on the size dependence of the melting temperature for Al nanoparticles (Levitas and Samani (2011a,b)). To eliminate this contradiction, it was found that an extra equation for stress relaxation at the interface could be formulated (Levitas and Samani (2011a,b)), obviating, at least for models of melting, the need to introduce additional elastic interface stresses. Ideally, the necessity of introducing additional elastic interface stresses for various solid-solid PTs and twinning (usually in a post hoc manner) should be validated through comparison with atomistic simulations or experiment. Since this has not yet been done, our approach in Levitas and Samani (2011a,b) for melting and in Levitas and Javanbakht (2010); Levitas (2013a,b); Levitas (2014b) for martensitic PTs and twinning is based on a assumption that *the elastic contribution to the interface stresses arises from the solution of the Ginzburg-Landau and mechanics equations for a PT problem* without any additional modeling assumptions. The main challenge lay in the determination of the structural contribution to the interface stresses σ_{st} (see Fig. 1), i.e., like for liquid-liquid or liquid-gas interfaces.¹

In the case of an isotropic interface energy, a rational formulation and solutions for inter-

¹ **Finite element simulations in Levitas and Javanbakht (2010, 2011a) demonstrated two important points: (a) Elastic interface stresses in some cases produce not only force but bending couple as well, i.e., more sophisticated gradient-type sharp-interface theories (e.g., like in Rubin and Benveniste (2004)) are required. (b) Elastic stresses are not generally localized within interface and they spread in bulk away from the interface; thus, it is not even conceptually clear how to define excess interface elastic stresses.**

face stresses for the general large strain case have been proposed in Levitas (2014b). Phase field equations are formulated therein, the Ginzburg-Landau equation is shown to admit a solution with a propagating interface with unit normal $\mathbf{k}$ in the deformed configuration, with no elastic and viscous interface stresses, where the structural Cauchy (true) stress tensor within the finite-width interface is found to be

$$\boldsymbol{\sigma}_{st} = \sigma_{st}(\eta, \nabla\eta)(\mathbf{I} - \mathbf{k} \otimes \mathbf{k}); \quad \mathbf{k} = \nabla\eta/|\nabla\eta|. \quad (4)$$

Since $\mathbf{I}$ and $\mathbf{I} - \mathbf{k} \otimes \mathbf{k}$ are the unit tensor and the two-dimensional unit tensor within a diffuse interface (i.e., it is orthogonal to $\mathbf{k}$), respectively, the tensor $\boldsymbol{\sigma}_{st}$ is the biaxial interface stress with a magnitude σ_{st} . Such stresses are localized within the interface and there are no other components of structural stress within an interface. The force per unit interface length

$$\bar{\sigma} := \int_{-\infty}^{\infty} \sigma_{st} d\varsigma = \gamma, \quad (5)$$

is equal to the nonequilibrium interface energy γ , where ς is the coordinate along the $\mathbf{k}$. It was found (Levitas (2013a,b); Levitas (2014b)) that in order to introduce such an interface stresses, the gradient energy per unit mass should have a form $\psi^\nabla = \frac{\beta}{2\rho} |\nabla\eta|^2$ and one of the local energy contributions should be proportional to $J = \rho_0/\rho$, where β is the gradient energy coefficient, and ρ and ρ_0 are the mass density in the deformed and undeformed states, respectively. It was also proved that if the free energy is an isotropic function of the gradients of the (multiple) order parameters in the deformed state, then the Cauchy stress is symmetric.

Some designations. For clarity we indicate the mathematical conventions used here. We designate contractions of tensors $\mathbf{A} = A_{ij}\mathbf{e}_i \otimes \mathbf{e}_j$ and $\mathbf{B} = B_{ij}\mathbf{e}_i \otimes \mathbf{e}_j$ over one and two indices as $\mathbf{A} \cdot \mathbf{B} = A_{ik}B_{kj}\mathbf{e}_i \otimes \mathbf{e}_j$ and $\mathbf{A} : \mathbf{B} = A_{ij}B_{ji}$, respectively; A_{ij} and B_{ij} are the components of tensors in Cartesian unit basis vectors $\mathbf{e}_i$, $\otimes$ designates a dyadic product, and summation is assumed over the repeated indices, unless otherwise stated. The superscript T indicates the

transpose of the tensor; subscripts s and a designate symmetric and skew-symmetric parts of a second-rank tensor; subscripts e , t , and θ mean elastic, transformational, and thermal strains; subscript 0 means that the parameter is defined in the reference configuration Ω_0 ; $\nabla_0 = \frac{\partial}{\partial \mathbf{r}_0}$ and $\nabla = \frac{\partial}{\partial \mathbf{r}}$ are the gradient operators in the reference configuration Ω_0 and the actual configuration Ω , respectively; in particular, for the second-rank tensor $\mathbf{B}$ we can write its gradient as $\frac{\partial \mathbf{B}}{\partial \mathbf{r}_0} = \frac{\partial B_{bc}}{\partial r_{0i}} \mathbf{e}_b \otimes \mathbf{e}_c \otimes \mathbf{e}_i$; $\mathbf{I}$ is the second-rank unit tensor with components δ_{ij} , the Kronecker delta; the Laplacians in the undeformed and deformed states are respectively $\nabla_0^2 := \nabla_0 \cdot \nabla_0 = \frac{\partial}{\partial \mathbf{r}_0} \cdot \frac{\partial}{\partial \mathbf{r}_0}$ and $\nabla^2 := \nabla \cdot \nabla = \frac{\partial}{\partial \mathbf{r}} \cdot \frac{\partial}{\partial \mathbf{r}}$. The expression $:=$ is used to indicate equality by definition.

Anisotropic interface energy. Anisotropic interface energies **for the case without mechanics** have been considered in a large number of publications (Anderson et al. (2000); Braun et al. (1997); Cahn and Hoffman (1974); Debierre et al. (2003); Hoffman-Cahn (1972); Kobayashi (1993); Taylor and Cahn (1998, 1994); Warren-Boettinger (1995); Wheeler and McFadden (1997)). They include derivations of the general phase field equations, their specification, calculation of sharp-interface limits, and solution of specific problems. Anisotropy of the interface energy is an essential ingredient in descriptions of dendritic growth, faceting (i.e., substitution of a smooth interface with a piece-wise smooth interface, containing faces with orientation corresponding to small interface energy), and realistic interface kinetics. **In these works, the free energy is postulated to be an anisotropic function of the gradient of the order parameters.**

For the case with mechanics (Anderson et al. (2000, 2001)), the free energy is accepted as a similar anisotropic function of the gradient of the order parameter in the deformed configuration $\psi^\nabla(\nabla\eta)$, because description of viscous liquid or solid does not involve the undeformed configuration. This assumption results in a generally nonsymmetric contribution to the Cauchy stress proportional to

$\nabla\eta_i \otimes \frac{\partial\psi^\nabla}{\partial\nabla\eta_i}$ (Anderson et al. (2000, 2001)). The same contribution for generally nonsymmetric interface stresses was obtained in Wheeler and McFadden (1997) for neglected strains by using the Noether's theorem and in Levitas (2013a); Levitas (2014b) for small and large strains. This nonsymmetry of the Cauchy stress in turn leads to two non-physical consequences:

- The stress power $\boldsymbol{\sigma}^T:\nabla\mathbf{v}$ is not invariant under superposition of the rigid-body rotation in the actual configuration, i.e., resultant theory is not objective. We are not aware of any prior discussion or study of this flaw in prior theories.
- The moment of momentum principle (which requires symmetry of the true stress) is violated.

Note that these problems do not exist in an approach by Fried and Grach (1997) based on the treatment in the reference configuration and the gradient energy $\psi^\nabla(\nabla_0\eta, \mathbf{F}) = \tilde{\psi}^\nabla(\nabla_0\eta, \mathbf{U})$ that depends on the gradient of the order parameter in the undeformed state and deformation gradient $\mathbf{F}$. Application of the principle of material frame-indifference (Levitas (1996); Lurie (1990); Šilhavý (1997); Truesdall and Noll (1965)) reduces $\mathbf{F}$ to the right-stretch tensor $\mathbf{U}$. Due to dependence on $\mathbf{U}$, gradient-dependent symmetric interface stresses appear. However, they are not reduced in the limit case of melting/solidification to biaxial interface true stresses with a magnitude equal to the interface energy. Also, gradient energy was not presented in the form that reduces for strain-free case to the known expressions used in Anderson et al. (2000); Braun et al. (1997); Cahn and Hoffman (1974); Debierre et al. (2003); Hoffman-Cahn (1972); Kobayashi (1993); Taylor and Cahn (1998, 1994); Warren-Boettinger (1995); Wheeler and McFadden (1997). This approach was not used for analysis of the interface

stresses and simulations of PTs. Similar approach in the reference configuration was presented in Clayton and Knap (2011a); Hildebrand and Miehe (2012). However, for specific models for anisotropic interface energy and simulations, U was omitted from ψ^∇ and interface stresses did not appear.

Thus, we have two choices: either eliminate contradictions of the models with anisotropic $\psi^\nabla(\nabla\eta)$ (which are broadly used in simulations) when strains are taken into account or transform objective formal model $\psi^\nabla(\nabla_0\eta, U)$ to the form that is consistent with known strain-free models with $\psi^\nabla(\nabla\eta)$ and reproduces correct expression for interface stresses. The first approach looks more straightforward and physical, especially since we have already solved the problem for isotropic interface energy $\psi^\nabla(\nabla\eta)$ for large strains (Levitas (2014b)). The main *goal* of the current paper is to generalize results in Levitas (2014b) for an anisotropic interface energy $\gamma(\mathbf{k})$ and, consequently, anisotropic $\psi^\nabla(\nabla\eta_i)$, in a consistent manner.

Note that both above problems could be in principle resolved by considering more general micropolar theory (see Appendix 2) with some microstructure, which rotates under action of some body couples and couple stresses. In fact, it is stated in Wheeler and McFadden (1997) that nonsymmetric part of the stress tensor generates an associated body couple distribution within finite-width interface. This statement has not been further elaborated and to our best knowledge none of known works on phase field approach treated the moment of momentum balance in this respect. However, this statement contradicts our previous, physically motivated, conclusion that the Herring torque is not a mechanical torque localized at the interface, that is, this choice of anisotropy of the interface energy produces configurational forces that rotate an interface with respect to the embedding material but these forces cannot directly influence the momentum equation. To reiterate, *a physically correct formulation of an anisotropic interface energy cannot change the momentum equation* and,

consequently, the *Cauchy stress tensor cannot be non-symmetric*.

This internal contradiction suggests that the initial assumption that the free energy is an anisotropic function of the gradient of the order parameters in the deformed configuration $\psi^\nabla(\nabla\eta_i)$ is incorrect. It will be shown in Section 4 that, in a the sharp-interface model, the proper approach to representing the interface energy anisotropy is through a dependency $\gamma(\mathbf{k}_0)$, where $\mathbf{k}_0$ is the unit normal to the interface in the *undeformed* state, and similarly, for a phase field approach the gradient energy should have a hybrid form $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0)$. That is, the anisotropy should be described with respect to the reference configuration (for which crystal lattice symmetry group is known) but the magnitude of the gradient of the order parameter $|\nabla\eta|$ is evaluated for the gradient in the deformed configuration (otherwise, interface stresses will not appear at all). Since $\mathbf{k}_0$ is independent of the deformation gradient $\mathbf{F}$, its inclusion does not change the expression for the Cauchy stresses, which remain symmetric. **Because $|\nabla\eta| = (\nabla\eta \cdot \nabla\eta)^{0.5} = (\nabla_0\eta \cdot \mathbf{F}^{-1} \cdot \mathbf{F}^{-1T} \cdot \nabla_0\eta)^{0.5} = (\nabla_0\eta \cdot \mathbf{U}^{-2} \cdot \nabla_0\eta)^{0.5}$, the function $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0) = \tilde{\psi}^\nabla(\nabla_0\eta, \mathbf{U})$ has the same objective form as postulated in Fried and Grach (1997) (see also Appendix 1). However, in contrast to Fried and Grach (1997), the function $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0)$ describes strain-independent in the actual configuration anisotropic interface energy (like for melting/solidification), easily generalizes anisotropic functions $\psi^\nabla(\nabla\eta)$ used for strain-free case, and will be specified in the way that it produces correct interfaces stresses. For multiple parameters, the gradient energy should have a form $\psi^\nabla(\nabla\eta_i \cdot \nabla\eta_j, \mathbf{k}_{0i})$, which also results in symmetric Cauchy stresses. This resolves the problem with description of anisotropy of the gradient and interface energies. This idea is realized within non-strict small strain formulation in Levitas and Warren (2015).**

Note that many of steps in derivation of the equations for anisotropic interface energy coincide with those in Levitas (2014b) for isotropic interface. We will repeat them as concisely as possible to maintain an internally complete presentation and refer to Levitas (2014b) for

detail. Further information about geometrically nonlinear formulations of continuum mechanics can be found in Levitas (1996, 1998); Lurie (1990); Malvern (1977); Šilhavý (1997); Truesdall and Noll (1965).

In Section 2, the kinematic, thermodynamic, and constitutive equations are developed, including the generalized Ginzburg-Landau equations for evolution of the order parameters. In Section 3, the structure of the Helmholtz free energy and expressions for the first Piola-Kirchhoff and Cauchy stresses are derived. In Section 4, the main contradictions of the existing theories based on the anisotropic gradient energy in the form $\psi^\nabla(\nabla\eta_i)$ are found, and a solution is proposed: that the gradient energy in the form $\psi^\nabla(\nabla\eta_i \cdot \nabla\eta_j, \mathbf{k}_{0i})$ is justified, which results in symmetric Cauchy stresses. In Section 5, explicit expression for Ginzburg-Landau equations are derived in the reference configuration. Simplifications for small strains are performed. Remarkably, this anisotropy yields a *first order* correction in the Ginzburg-Landau equation for small strains, which has been neglected in prior work. The next non-zero term containing the transformation work is third order in the strain. Boundary conditions for the order parameters are formulated in the reference configuration in Section 6. In Section 7, previous expressions for nonsymmetric interface stresses are analyzed. In Section 8, regularization for the case with the strong anisotropy is performed. In Section 9, we simplify the general treatment, specifying the Helmholtz energy and Ginzburg-Landau equation for a single order parameter. Readers interested in getting a general feel for the results might examine the single order parameter results first. In Section 10, an analytical solution for the propagating interface, which includes the anisotropic gradient energy and distribution of components of interface stresses, has been found using a sixth-degree polynomial in the phase field. Also, the splitting of the thermal parts of the free energy into a part that contributes to the interface stresses, and another that does not, is presented. In Section 11, the orientation-dependence of the, suitably defined, *nonequilibrium* interface energy and width is found. In Section 12, a detailed analytical study of the energy, width, entropy excess, and stresses for

a nonequilibrium interface is performed. Interface stresses for a critical martensitic nucleus are determined in Section 13. A complete system of equations for fifth- and sixth-degree polynomials forms of the free energy is presented in the reference configurations in Section 13. Section 14 contains concluding remarks. Equations for entropy and temperature evolution as well as possible micropolar theory are presented in the Appendix.

2. Kinematics, Thermodynamics, and Constitutive Equations

Kinematics. Let a continuous vector function $\mathbf{r} = \mathbf{r}(\mathbf{r}_0, t)$, where $\mathbf{r}_0$ and $\mathbf{r}$ are the positions of points in the reference (undeformed) Ω_0 and the actual (deformed) Ω configurations, respectively, and t is the time, describe the motion of the elastic material with PTs. Assume that at time t_0 the material is in the high symmetry phase $\mathbf{H}$ and it may be transformed into a number of lower symmetry phases $\mathbf{L}_i$, which may include martensitic variants. Each of the PTs $\mathbf{H} \leftrightarrow \mathbf{L}_i$ is described by corresponding order parameter $\eta_i \geq 0$ with $\eta_i = 0 \ \forall i$ for $\mathbf{H}$ and $\eta_k = 1, \eta_i = 0 \ \forall i \neq k$ for $\mathbf{L}_k$. Let us define the deformation gradient $\mathbf{F} = \frac{\partial \mathbf{r}}{\partial \mathbf{r}_0} = \nabla_0 \mathbf{r} = \frac{\partial r_i}{\partial r_0^j} \mathbf{e}_i \otimes \mathbf{e}_j$, where r_i and r_0^j are the components of the vectors $\mathbf{r}$ and $\mathbf{r}_0$ in the Cartesian unit basis vectors $\mathbf{e}_i$. We will utilize the multiplicative decomposition of the deformation gradient,

$$\mathbf{F} = \mathbf{F}_e \cdot \mathbf{U}_\theta \cdot \mathbf{U}_t; \quad \mathbf{U}_\theta = \mathbf{U}_\theta^T; \quad \mathbf{U}_t = \mathbf{U}_t^T \quad (6)$$

into elastic, rotation-free thermal, and rotation-free transformational contributions, see Levitas (2014b) for details and Fig. 2 for designation of corresponding configurations. The following Jacobians of deformation gradients will be introduced in terms of ratios of volumes V and mass densities ρ in the corresponding configurations:

$$\begin{aligned} J &:= \frac{dV}{dV_0} = \frac{\rho_0}{\rho} = \det \mathbf{F}; & J_t &:= \frac{dV_t}{dV_0} = \frac{\rho_0}{\rho_t} = \det \mathbf{U}_t; & J_\theta &:= \frac{dV_\theta}{dV_t} = \frac{\rho_t}{\rho_\theta} = \det \mathbf{U}_\theta; \\ J_e &:= \frac{dV_e}{dV_\theta} = \frac{\rho_\theta}{\rho_e} = \det \mathbf{F}_e; & J_{t\theta} &:= \frac{dV_\theta}{dV_0} = \frac{\rho_0}{\rho_\theta} = \det(\mathbf{U}_\theta \cdot \mathbf{U}_t) = J_t J_\theta; & J &= J_t J_\theta J_e. \end{aligned} \quad (7)$$

Figure 2: Multiplicative decomposition of the deformation gradient into elastic, thermal, and transformational parts and corresponding configurations: Ω_0 and Ω are the reference and current configurations, respectively. The intermediate transformed configuration Ω_t represents the crystal lattice of the lower symmetry phase L_i at the reference temperature θ_0 , i.e., the reference configuration for L_i . The intermediate unloaded configuration Ω_θ is obtained from Ω after a local release of stresses to zero at current temperature θ_0 .

For small strains and rotations, one has $\mathbf{F} \simeq \mathbf{I} + \boldsymbol{\varepsilon} + \boldsymbol{\omega}$, $\mathbf{F}_e \simeq \mathbf{I} + \boldsymbol{\varepsilon}_e + \boldsymbol{\omega}_e$, $\mathbf{U}_t \simeq \mathbf{I} + \boldsymbol{\varepsilon}_t$, and $\mathbf{U}_\theta \simeq \mathbf{I} + \boldsymbol{\varepsilon}_\theta$, where $\boldsymbol{\varepsilon}$ and $\boldsymbol{\omega}$ are the small symmetric strain tensor and the small antisymmetric rotation tensor, respectively, with components that are small in comparison with unity. Neglecting the product of small terms the multiplicative decomposition (6) reduces to the additive decomposition of small strains:

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_e + \boldsymbol{\varepsilon}_t + \boldsymbol{\varepsilon}_\theta. \quad (8)$$

Due to zero thermal and transformational rotations, $\boldsymbol{\omega} = \boldsymbol{\omega}_e$. A double contraction of Eq.(8) with the unit tensor (the trace) leads to an additive decomposition of volumetric strains:

$$\varepsilon_0 = \boldsymbol{\varepsilon} : \mathbf{I} = \varepsilon_{e0} + \varepsilon_{t0} + \varepsilon_{\theta 0}; \quad J \simeq 1 + \varepsilon_0. \quad (9)$$

Thermodynamics laws. The first and second laws of thermodynamics represent the global energy balance equation

$$\int_{S_0} (\mathbf{p} \cdot \mathbf{v} - \mathbf{h}_0 \cdot \mathbf{n}_0) dS_0 + \int_{S_0} \mathbf{Q}_{0i}^\eta \dot{\eta}_i \cdot \mathbf{n}_0 dS_0 + \int_{V_0} \rho_0 (\mathbf{f} \cdot \mathbf{v} + r) dV_0 = \frac{d}{dt} \int_{V_0} \rho_0 (U + 0.5 \mathbf{v} \cdot \mathbf{v}) dV_0 \quad (10)$$

and the global entropy balance equation combined with the Clausius-Duhem inequality:

$$S_i := \frac{d}{dt} \int_{V_0} \rho_0 s dV_0 - \int_{V_0} \rho_0 \frac{r}{\theta} dV_0 + \int_{S_0} \frac{\mathbf{h}_0}{\theta} \cdot \mathbf{n}_0 dS_0 \geq 0, \quad (11)$$

Here $\mathbf{p} = \mathbf{P} \cdot \mathbf{n}_0$ is the traction vector, $\mathbf{P}$ is the first Piola–Kirchhoff stress tensor, $\mathbf{v} = \dot{\mathbf{r}}$ is the material velocity, $\mathbf{h}_0$ is the heat flux, $\mathbf{n}_0$ is the unit outer normal to S_0 , U and s are the specific internal energy and entropy, $\mathbf{f}$ is the body force and r is the specific volumetric heat supply rate, S_i is the total entropy production rate, and $\theta \geq 0$ is the temperature. All specific functions are defined per unit mass. The generalized surface forces $\mathbf{Q}_{0i}^\eta \cdot \mathbf{n}_0$ conjugated with $\dot{\eta}_i$ are introduced in order to balance terms due to dependence of the thermodynamic potential on $\nabla_0 \eta_i$, which makes it possible to apply Eq.(10) for an arbitrary volume. The

energy of the external surface will not be included in the laws of thermodynamics because the volume V_0 can be arbitrarily chosen within an actual body and its surface S_0 is not an external surface.

Utilizing the Gauss theorem to transform surface integrals into integrals over the volume and momentum balance equation, as well as taking into account an arbitrariness of the volume, we obtain a local form of the thermodynamics laws:

$$\mathbf{P}:\dot{\mathbf{F}}^T - \rho_0\dot{U} - \nabla_0 \cdot \mathbf{h}_0 + \rho_0 r + \nabla_0 \cdot (\mathbf{Q}_{0i}^\eta \dot{\eta}_i) = 0; \quad (12)$$

$$\rho \tilde{S}_i := \rho_0 \dot{s} - \rho_0 \frac{r}{\theta} + \nabla_0 \cdot \frac{\mathbf{h}_0}{\theta} = \rho_0 \dot{s} - \rho_0 \frac{r}{\theta} + \frac{1}{\theta} \nabla_0 \cdot \mathbf{h}_0 - \frac{\nabla_0 \theta}{\theta^2} \cdot \mathbf{h}_0 \geq 0, \quad (13)$$

where $\tilde{S}_i$ is the local entropy production rate. Introducing the local dissipation rate, $\bar{\mathcal{D}} := \theta \tilde{S}_i$ and the specific Helmholtz free energy $\psi = U - \theta s$ (i.e., substituting $U = \psi + \theta s$), and resolving Eq.(12) for the expression $\nabla_0 \cdot \mathbf{h}_0 - \rho_0 r$, and substituting it in the equation for $\bar{\mathcal{D}}$, we obtain the following dissipation inequality

$$\rho_0 \bar{\mathcal{D}} := \mathbf{P}:\dot{\mathbf{F}}^T - \rho_0 \dot{\psi} - \rho_0 s \dot{\theta} + \nabla_0 \cdot (\mathbf{Q}_{0i}^\eta \dot{\eta}_i) - \frac{\nabla_0 \theta}{\theta} \cdot \mathbf{h}_0 = \rho_0 \mathcal{D} - \frac{\nabla_0 \theta}{\theta} \cdot \mathbf{h}_0 \geq 0. \quad (14)$$

It is usually assumed that the heat conduction and other thermomechanical processes are mutually independent. Then Eq.(14) is equivalent to two stronger inequalities: the Fourier's inequality $-\nabla_0 \theta \cdot \mathbf{h}_0 \geq 0$ and the mechanical dissipation inequality:

$$\rho_0 \mathcal{D} = \mathbf{P}:\dot{\mathbf{F}}^T - \rho_0 \dot{\psi} - \rho_0 s \dot{\theta} + \nabla_0 \cdot (\mathbf{Q}_{0i}^\eta \dot{\eta}_i) \geq 0. \quad (15)$$

We will need the Cauchy (true) stress tensor $\boldsymbol{\sigma}$ and velocity gradient in the deformed configuration $\mathbf{l}$,

$$\boldsymbol{\sigma} := \frac{\rho}{\rho_0} \mathbf{P} \cdot \mathbf{F}^T; \quad \mathbf{P} = \frac{\rho_0}{\rho} \boldsymbol{\sigma} \cdot \mathbf{F}^{T-1}; \quad \mathbf{l} := \nabla \mathbf{v} = \dot{\mathbf{F}} \cdot \mathbf{F}^{-1}. \quad (16)$$

With their help the stress power can be presented in terms of tensors in the actual configuration

$$\mathbf{P}:\dot{\mathbf{F}}^T = \frac{\rho_0}{\rho} \boldsymbol{\sigma}:\mathbf{F}^{T-1}:\dot{\mathbf{F}}^T = \frac{\rho_0}{\rho} \boldsymbol{\sigma}:(\dot{\mathbf{F}} \cdot \mathbf{F}^{-1})^T = \frac{\rho_0}{\rho} \boldsymbol{\sigma}^T:(\dot{\mathbf{F}} \cdot \mathbf{F}^{-1}) = \frac{\rho_0}{\rho} \boldsymbol{\sigma}^T:\mathbf{l}, \quad (17)$$

which will be utilized below. For a symmetric Cauchy stress tensor the stress power simplifies to $\frac{\rho_0}{\rho} \boldsymbol{\sigma}:\mathbf{d}$, where $\mathbf{d} = (\mathbf{l})_s$ is the deformation rate.

Constitutive equations. Accepting $\psi = \bar{\psi}(\mathbf{F}, \eta_i, \theta, \nabla_0 \eta_i)$ and substituting $\dot{\psi}$ into Eq.(15) results in

$$\begin{aligned} \rho_0 \mathcal{D} = & \left(\mathbf{P} - \rho_0 \frac{\partial \bar{\psi}}{\partial \mathbf{F}} \right) : \dot{\mathbf{F}}^T - \rho_0 \left(s + \frac{\partial \bar{\psi}}{\partial \theta} \right) \dot{\theta} \\ & - \left(\rho_0 \frac{\partial \bar{\psi}}{\partial \eta_i} - \nabla_0 \cdot \mathbf{Q}_{0i}^\eta \right) \dot{\eta}_i + \left(\mathbf{Q}_{0i}^\eta - \rho_0 \frac{\partial \bar{\psi}}{\partial \nabla_0 \eta_i} \right) \cdot \dot{\overline{\nabla_0 \eta_i}} \geq 0. \end{aligned} \quad (18)$$

We took into account the following identity:

$$\nabla_0 \cdot (\mathbf{Q}_{0i}^\eta \dot{\eta}_i) = (\nabla_0 \cdot \mathbf{Q}_{0i}^\eta) \dot{\eta}_i + \mathbf{Q}_{0i}^\eta \cdot \nabla_0 \dot{\eta}_i = (\nabla_0 \cdot \mathbf{Q}_{0i}^\eta) \dot{\eta}_i + \mathbf{Q}_{0i}^\eta \cdot \dot{\overline{\nabla_0 \eta_i}}. \quad (19)$$

Assuming independence of the dissipation rate of $\dot{\theta}$ and $\dot{\overline{\nabla_0 \eta_i}}$ implies the constitutive equation for entropy and the definition of the thermodynamic forces $\mathbf{Q}_{0i}^\eta$:

$$s = -\frac{\partial \bar{\psi}}{\partial \theta}; \quad \mathbf{Q}_{0i}^\eta = \rho_0 \frac{\partial \bar{\psi}}{\partial \nabla_0 \eta_i}. \quad (20)$$

The remaining inequality (18) can be presented in the form

$$\rho_0 \mathcal{D} = \mathbf{P}_v : \dot{\mathbf{F}}^T + \rho_0 X_i \dot{\eta}_i \geq 0; \quad \mathbf{P}_v := \mathbf{P} - \rho_0 \frac{\partial \bar{\psi}}{\partial \mathbf{F}}; \quad \rho_0 X_i := \rho_0 \frac{\partial \bar{\psi}}{\partial \eta_i} - \nabla_0 \cdot \mathbf{Q}_{0i}^\eta. \quad (21)$$

where the generalized viscous stresses $\mathbf{P}_v$ and dissipative forces X_i work-conjugated to $\dot{\eta}_i$ were introduced. The assumption that the processes described by $\dot{\mathbf{F}}$ and $\dot{\eta}_i$ are thermodynamically independent leads to two stricter inequalities:

$$\mathbf{P}_v : \dot{\mathbf{F}}^T \geq 0; \quad X_i \dot{\eta}_i \geq 0. \quad (22)$$

To satisfy them, one has to assume that $\mathbf{P}_v = \mathbf{P}_v(\dot{\mathbf{F}}^T)$ and $X_i = X_i(\dot{\eta}_j)$, otherwise, these inequalities can be easily violated. In combination with Eq.(21) these assumptions lead to constitutive equations for the first Piola–Kirchhoff stress and the evolution equation for η_i

$$\mathbf{P} = \rho_0 \frac{\partial \bar{\psi}}{\partial \mathbf{F}} + \mathbf{P}_v; \quad \rho_0 X_i = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta_i} + \nabla_0 \cdot \left(\rho_0 \frac{\partial \bar{\psi}}{\partial \nabla_0 \eta_i} \right). \quad (23)$$

For linear relationships $\dot{\eta}_i = L_{ij} X_j$, Eq.(23) transforms to the generalized Ginzburg-Landau equation

$$\dot{\eta}_j = L_{ji} \left(-\frac{\partial \bar{\psi}}{\partial \eta_i} + \frac{1}{\rho_0} \nabla_0 \cdot \left(\rho_0 \frac{\partial \bar{\psi}}{\partial \nabla_0 \eta_i} \right) \right), \quad (24)$$

where L_{ij} are positive definite kinetic coefficients satisfying the Onsager reciprocal relationships $L_{ij} = L_{ji}$. For initially homogeneous material ρ_0 does not vary in space and disappears from Eq.(24).

3. Structure of the Helmholtz free energy and expression for stresses

3.1. Structure of the Helmholtz free energy

We will use the same structure of the free energy per unit mass as in Levitas (2014b)

$$\begin{aligned} \bar{\psi}(\mathbf{F}, \eta_i, \theta, \nabla \eta_i, \nabla_0 \eta_i) &= \frac{J_{t\theta}}{\rho_0} \psi^e(\mathbf{F} \cdot \mathbf{U}_t^{-1}(\eta_i) \cdot \mathbf{U}_\theta^{-1}(\theta, \eta_i), \eta_i, \theta) \\ &+ J\check{\psi}^\theta(\theta, \eta_i) + \tilde{\psi}^\theta(\theta, \eta_i) + J\psi^\nabla(\nabla \eta_i, \nabla_0 \eta_i), \end{aligned} \quad (25)$$

but in order to describe anisotropy of the interface energy, the gradient energy $\psi^\nabla(\nabla \eta_i, \nabla_0 \eta_i) = \psi^\nabla(\nabla_0 \eta_i \cdot \mathbf{F}^{-1}, \nabla_0 \eta_i)$ depends on the gradients of the order parameters both in the reference and current configuration. This leads to dependence of ψ^∇ on $\mathbf{F}$ in a specific way. An additional effect of $\mathbf{F}$ can be included as in Fried and Grach (1997), if it is proven to be required (see Introduction). In Eq.(25) ψ^e is the elastic energy, which depends on $\mathbf{F}_e = \mathbf{F} \cdot \mathbf{U}_t^{-1} \cdot \mathbf{U}_\theta^{-1}$, η_i , and θ ; $\check{\psi}^\theta$ is the thermal (chemical) energy localized at the interfaces, which is equal to

zero in the bulk (i.e., when all order parameters are 0 or one of them is equal to 1) and $\tilde{\psi}^\theta$ is the thermal energy related to the thermal driving force for H - L_i PT (i.e., to the difference between the thermal parts of the free energies of L_i and H, ΔG^θ).

It was demonstrated in Levitas (2013a) for small strains and in Levitas (2014b) for large strains that considering the gradient $\nabla\eta_i$ in the deformed configuration as an argument of ψ^∇ and multiplying $\tilde{\psi}^\theta$ and ψ^∇ by the Jacobian J will lead (with the proper choice of $\tilde{\psi}^\theta$) to the desired expression for the interface stresses, at least for a single martensitic variant and isotropic function $\psi^\nabla(\nabla\eta_i)$, i.e., isotropic interface energy. The terms $\tilde{\psi}^e$ and $\tilde{\psi}^\theta$, which are not multiplied by J , do not contribute to the interface stresses σ_{st} . The multiplier $\frac{J_{t\theta}}{\rho_0}$ for the elastic energy is introduced to assure that the elasticity rule is defined in the unloaded configuration Ω_θ , in which it is determined experimentally or in atomistic simulations. These geometric nonlinearities should be kept even in small strain formulation (Levitas (2013a)).

3.2. Constitutive equations for stresses

Using Eq.(23) and the same procedure as in Levitas (2014b) we derive the following expression for the first Piola–Kirchhoff stress tensor

$$\begin{aligned} \mathbf{P} &= \mathbf{P}_e + \mathbf{P}_{st} + \mathbf{P}_v(\dot{\mathbf{F}}); \quad \mathbf{P}_e = J_{t\theta} \mathbf{F}_e \cdot \frac{\partial \psi^e}{\partial \mathbf{E}_e} \cdot \mathbf{F}_e^T \cdot \mathbf{F}^{T-1}; \\ \mathbf{P}_{st} &= \mathbf{P}^J + \mathbf{P}^\nabla = \rho_0 J (\tilde{\psi}^\theta + \psi^\nabla) \mathbf{F}^{T-1} - \rho_0 J \nabla\eta_i \otimes \frac{\partial \psi^\nabla}{\partial \nabla\eta_i} \cdot \mathbf{F}^{T-1}, \end{aligned} \quad (26)$$

and for the Cauchy stress tensor

$$\begin{aligned} \sigma &:= \frac{\rho}{\rho_0} \mathbf{P} \cdot \mathbf{F}^T = \sigma_e + \sigma_{st} + \sigma_v(\mathbf{d}); \quad \sigma_e = J_e^{-1} \mathbf{F}_e \cdot \frac{\partial \psi^e}{\partial \mathbf{E}_e} \cdot \mathbf{F}_e^T; \\ \sigma_{st} &= \sigma^J + \sigma^\nabla = \rho_0 (\tilde{\psi}^\theta + \psi^\nabla) \mathbf{I} - \rho_0 \nabla\eta_i \otimes \frac{\partial \psi^\nabla}{\partial \nabla\eta_i}, \end{aligned} \quad (27)$$

where $\mathbf{E}_e = 0.5(\mathbf{F}_e^T \cdot \mathbf{F}_e - \mathbf{I})$ is the Lagrangian elastic strain. Here $\mathbf{P}_e$ and σ_e are elastic stresses, which are related to elastic strain-dependence of elastic energy ψ^e ; $\mathbf{P}^J$ and σ^J

are stresses due to multiplication of $\check{\psi}^\theta$ and ψ^∇ in Eq.(25) by a Jacobian J , and $\mathbf{P}^\nabla$ and $\boldsymbol{\sigma}^\nabla$ are due to dependence of the gradient energy on the deformation gradient $\mathbf{F}$. Stresses $\boldsymbol{\sigma}^J$ and $\boldsymbol{\sigma}^\nabla$ (i.e., non-elastic stresses) are combined in $\boldsymbol{\sigma}_{st}$ (similar for the Piola-Kirchhoff stress), which are called structural stresses at the interface. Adding $\nabla_0 \eta_k$ as an argument of ψ^∇ in comparison with Levitas (2014b) does not change the expression for stresses since it is independent of $\mathbf{F}$. It is shown in Levitas (2014b) for isotropic gradient energy and shown below for anisotropic gradient energy that for the nonequilibrium interface, structural stresses are localized at the interface and for a single order parameter $\boldsymbol{\sigma}_{st}$ reduces to biaxial tension with the magnitude of force per unit length equal to the interface energy. For a geometrically linear case, we have $\mathbf{P}_{st} = \boldsymbol{\sigma}_{st}$ and the expression for $\boldsymbol{\sigma}_{st}$ does not change.

For true stress, the elastic and viscous parts as well as part due to the Jacobian are symmetric tensors. However, stresses due to gradient energy in general possess a skew-symmetric part:

$$\boldsymbol{\sigma}_a = -\rho_0 \left(\nabla \eta_i \otimes \frac{\partial \psi^\nabla}{\partial \nabla \eta_i} \right)_a = \rho_0 \left(\frac{\partial \psi^\nabla}{\partial \nabla \eta_i} \otimes \nabla \eta_i \right)_a. \quad (28)$$

Isotropic interface energy. For the material with isotropic interface energy, ψ^∇ is an isotropic function of $\boldsymbol{\zeta}_i = \nabla \eta_i$, i.e., it depends on invariants $a_{jk} = \boldsymbol{\zeta}_j \cdot \boldsymbol{\zeta}_k = a_{kj}$ for all k and j , $\psi^\nabla = \psi^\nabla(a_{jk})$. It was proven in Levitas (2014b) that in this case the Cauchy stress is symmetric, i.e.,

$$\left(\frac{\partial \psi^\nabla}{\partial \nabla \eta_i} \otimes \nabla \eta_i \right)_a = 0. \quad (29)$$

Simple manipulations confirm this:

$$\frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_i} \otimes \boldsymbol{\zeta}_i = \frac{\partial \psi^\nabla}{\partial a_{jk}} \frac{\partial a_{jk}}{\partial \boldsymbol{\zeta}_i} \otimes \boldsymbol{\zeta}_i = \frac{\partial \psi^\nabla}{\partial a_{jk}} (\boldsymbol{\zeta}_k \delta_{ji} \otimes \boldsymbol{\zeta}_i + \boldsymbol{\zeta}_j \delta_{ik} \otimes \boldsymbol{\zeta}_i) = \frac{\partial \psi^\nabla}{\partial a_{jk}} (\boldsymbol{\zeta}_k \otimes \boldsymbol{\zeta}_j + \boldsymbol{\zeta}_j \otimes \boldsymbol{\zeta}_k), \quad (30)$$

because invariants a_{jk} are symmetric.

4. Structure of expression for anisotropic gradient energy

Main contradictions. The main problem in developing a PFA with an anisotropic interface (gradient) energy is that the interface Cauchy stress $\boldsymbol{\sigma}$ becomes non-symmetric in general. In this case the application of traditional continuum mechanics treatment is *contradictory*.

(a) First, stress power (see Eq.(17)) $\boldsymbol{\sigma}^T : \nabla \mathbf{v}$ is not invariant with respect to rigid body rotation $\mathbf{r}^* = \mathbf{Q} \cdot \mathbf{r}$ in the actual configuration, where $\mathbf{Q}$ is an arbitrary proper orthogonal rotation tensor. Indeed, transformation rules of tensors under rigid body rotation are

$$\boldsymbol{\sigma}^* = \mathbf{Q} \cdot \boldsymbol{\sigma} \cdot \mathbf{Q}^T; \quad \boldsymbol{\sigma}^{T*} = \mathbf{Q} \cdot \boldsymbol{\sigma}^T \cdot \mathbf{Q}^T; \quad \nabla \mathbf{v}^* = \mathbf{Q} \cdot \nabla \mathbf{v} \cdot \mathbf{Q}^T + \dot{\mathbf{Q}} \cdot \mathbf{Q}^T, \quad (31)$$

where $\boldsymbol{\Omega} := \dot{\mathbf{Q}} \cdot \mathbf{Q}^T$ is the antisymmetric spin tensor associated with $\mathbf{Q}$. Then

$$\boldsymbol{\sigma}^{T*} : \nabla \mathbf{v}^* = \mathbf{Q} \cdot \boldsymbol{\sigma}^T \cdot \mathbf{Q}^T : \mathbf{Q} \cdot \nabla \mathbf{v} \cdot \mathbf{Q}^T + \mathbf{Q} \cdot \boldsymbol{\sigma}^T \cdot \mathbf{Q}^T : \dot{\mathbf{Q}} \cdot \mathbf{Q}^T = \boldsymbol{\sigma}^T : \nabla \mathbf{v} + \boldsymbol{\sigma}^T : \mathbf{Q}^T \cdot \dot{\mathbf{Q}}, \quad (32)$$

where $\mathbf{Q}^T \cdot \dot{\mathbf{Q}} = -\dot{\mathbf{Q}}^T \cdot \mathbf{Q}$ is the antisymmetric spin tensor associated with $\mathbf{Q}^T$. If stress $\boldsymbol{\sigma}$ is the symmetric tensor, then $\boldsymbol{\sigma} : \mathbf{Q}^T \cdot \dot{\mathbf{Q}} = 0$ and stress power $\boldsymbol{\sigma}^{T*} : \nabla \mathbf{v}^* = \boldsymbol{\sigma}^T : \nabla \mathbf{v}$ is independent of the rigid body rotations. However, for non-symmetric stress $\boldsymbol{\sigma} : \mathbf{Q}^T \cdot \dot{\mathbf{Q}} \neq 0$ and stress power explicitly depends on the rigid body rotations, i.e., it is non-objective.

(b) The second problem is, since the moment of momentum balance requires symmetry of the Cauchy stress, it is violated for nonsymmetric Cauchy stress. As it was discussed in Introduction, an anisotropy of the interface energy produces a configurational force (Herring torque) that rotates the interface with respect to material but does not produce a mechanical torque that contributes to the moment of momentum equations. That is why anisotropy of the interface energy does not change the moment of momentum equations and, consequently, the Cauchy stress tensor cannot be non-symmetric.

This implies that the initial assumption that the free energy is an anisotropic function of the gradient of the order parameters in the deformed configuration $\psi^\nabla(\nabla \eta_i)$ is *contradictory*.

Sharp-interface approach. To get an indication on how this can be done, we first answer the following question: for a sharp interface with an orientation-dependent interface energy, should γ depend on the unit normal $\mathbf{k}$ in the deformed state or on the unit normal $\mathbf{k}_0$ in the undeformed state? The crystal lattice and symmetry are known in the undeformed state, so function $\gamma(\mathbf{k}_0)$ should be invariant with respect to the symmetry group of the undeformed lattice. Thus, γ should depend on the joint invariants of constant tensors $\mathbf{K}_j$ that describe symmetry of the undeformed lattice and $\mathbf{k}_0$. Symmetry of the deformed lattice depends (in addition to the symmetry group of the undeformed lattice) on the deformation gradient $\mathbf{F}$, consequently, $\gamma = \gamma(\mathbf{k}, \mathbf{F}, \mathbf{K}_{Rj})$ should depend on the joint invariants of tensors $\mathbf{K}_{Rj}$ (if we want them to be expressed in the current configuration), $\mathbf{k}_0$, and $\mathbf{F}$. Thus, the primary function is $\gamma(\mathbf{k}_0)$, which can be transformed to the form of $\gamma = \gamma(\mathbf{k}, \mathbf{F}, \mathbf{K}_{Rj})$.

Phase field approach. The same is true for the phase field approach. It is known that for single order parameter an isotropic interface energy depends on $|\nabla\eta|$ and reproduces the correct expression for the interface stresses. For an anisotropic interface energy, a priori one may have $\psi^\nabla(|\nabla\eta|, \mathbf{k}) = \tilde{\psi}^\nabla(\nabla\eta)$ or $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0)$. The first function is inadmissible because it leads to nonsymmetric Cauchy stress and should additionally in any case depend on $\mathbf{F}$ to take the change in lattice symmetry during deformation into account. The function $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0)$ corresponds to $\gamma(\mathbf{k}_0)$ and (as it will be shown below) does not violate the symmetry of the Cauchy stress. We do not consider function $\psi^\nabla(|\nabla_0\eta|, \mathbf{k}_0)$, because it results in a total lack of structural interface stresses.

Thus, we will describe *anisotropy of the interface energy in terms of vector $\mathbf{k}_0$ or (for multiple order parameters) $\mathbf{k}_{0i}$* , which leads us to a function

$$\psi^\nabla = \psi^\nabla(\nabla\eta_k \cdot \nabla\eta_j, \mathbf{k}_{0i}), \quad (33)$$

which is essentially different from $\psi^\nabla(\nabla\eta_i)$ used in the previous works. Deformation gradient $\mathbf{F}$ also can be included in this dependence.

Expression for structural stresses. An expression for gradient-related structural stresses $\mathbf{P}^\nabla$ and $\boldsymbol{\sigma}^\nabla$ for the isotropic gradient energy $\psi^\nabla(\nabla\eta_k \cdot \nabla\eta_j)$ can be obtained by combining Eqs.(27) and (30). For anisotropic gradient energy we added arguments $\mathbf{k}_{0i} = \nabla_0\eta_i/|\nabla_0\eta_i|$, which are independent of the deformation gradient $\mathbf{F}$, so there is no additional contribution to the expression for stresses $\mathbf{P}^\nabla$ and $\boldsymbol{\sigma}^\nabla$. Thus, for anisotropic gradient energy function $\psi^\nabla(\nabla\eta_k \cdot \nabla\eta_j, \mathbf{k}_{0i})$ one obtains from Eqs.(27) and (30) for the first Piola-Kirchhoff stress

$$\mathbf{P}^\nabla = -\rho_0 J \nabla\eta_k \otimes \frac{\partial\psi^\nabla}{\partial\nabla\eta_k} \cdot \mathbf{F}^{T-1} = -\rho_0 J \frac{\partial\psi^\nabla}{\partial a_{jk}} (\nabla\eta_k \otimes \nabla\eta_j + \nabla\eta_j \otimes \nabla\eta_k) \cdot \mathbf{F}^{T-1} \quad (34)$$

and the corresponding Cauchy stress

$$\boldsymbol{\sigma}^\nabla = J^{-1} \mathbf{P}^\nabla \cdot \mathbf{F}^T = -\rho_0 \frac{\partial\psi^\nabla}{\partial a_{jk}} (\nabla\eta_k \otimes \nabla\eta_j + \nabla\eta_j \otimes \nabla\eta_k), \quad (35)$$

where, as before, $a_{jk} = \nabla\eta_j \cdot \nabla\eta_k = a_{kj}$. *Symmetry of this tensor* is evident, which resolves our problem. In particular, for

$$\rho_0 \psi^\nabla = 0.5\beta^2(\mathbf{k}_0)|\nabla\eta|^2 \quad (36)$$

one obtains

$$\boldsymbol{\sigma}^\nabla = -\beta^2(\mathbf{k}_0)\nabla\eta \otimes \nabla\eta, \quad (37)$$

and from Eq.(27)

$$\begin{aligned} \boldsymbol{\sigma}_{st} &= (\rho_0 \check{\psi}^\theta + 0.5\beta^2(\mathbf{k}_0)|\nabla\eta|^2)\mathbf{I} - \beta^2(\mathbf{k}_0)\nabla\eta \otimes \nabla\eta = \\ &\beta^2(\mathbf{k}_0)|\nabla\eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho_0 \check{\psi}^\theta - 0.5\beta^2(\mathbf{k}_0)|\nabla\eta|^2)\mathbf{I}. \end{aligned} \quad (38)$$

As it will be shown below, function $\check{\psi}^\theta$ can be defined in the way that the last term disappears for a propagating interface and interface stress reduces to a biaxial tension

$$\boldsymbol{\sigma}_{st} = \sigma_{st}(\mathbf{I} - \mathbf{k} \otimes \mathbf{k}); \quad \sigma_{st} = \beta^2(\mathbf{k}_0)|\nabla\eta|^2 \quad (39)$$

with the magnitude equal to the double interface energy. For solutions that do not represent a propagating interface (e.g., a critical nuclei or any intermediate nonstationary structures), Eq.(38) should be used.

Note that after equations are derived for models with strain, the interface stresses in Eq.(38) do not disappear for zero strains. Indeed, if free energy includes the term $\mathbf{A}:\boldsymbol{\varepsilon}$ with strain-independent $\mathbf{A}$, stress $\boldsymbol{\sigma}_{st}$ includes the term $\mathbf{A}$, which is not zero even for zero strains. Apparently, these strain-free Cauchy stresses can be obtained without involving strains, e.g., by utilizing the Noether's theorem (see Wheeler and McFadden (1997)). Still, there is a difference: stress in Wheeler and McFadden (1997) includes $\rho\psi$, which includes the bulk part that can be set zero in one phase but is nonzero in another one (Levitas and Javanbakht (2010); Levitas and Samani (2011a); Levitas (2014b)), which is contradictory. We eliminated this problem by excluding $\tilde{\psi}^\theta$ from the expression for $\boldsymbol{\sigma}_{st}$ by not multiplying it by J in Eq.(25). We do not see how to do the same in strain-free approach.

5. Ginzburg-Landau equation

5.1. Previous expressions

Let us first describe how gradient energy was presented in the previous works, i.e., when $\rho_0\psi^\nabla = 0.5\beta^2(\mathbf{k})|\nabla\eta|^2$. It is proven that $\beta(\mathbf{k}) = m\gamma(\mathbf{k})$, where $\gamma(\mathbf{k})$ is the energy of an equilibrium interface and m is a constant, which is completely determined by the double-well barrier (Taylor and Cahn (1998, 1994)). That is why we prefer to use a coefficient of the form β^2 instead of β , which we used before (Levitas and Javanbakht (2010); Levitas (2013a,b); Levitas (2014b)). It is traditional (Taylor and Cahn (1998, 1994)) to define the homogeneous degree one function $\beta(\nabla\eta) = |\nabla\eta|\beta(\mathbf{k})$; then $\rho_0\psi^\nabla = 0.5\beta^2(\nabla\eta) = 0.5\beta^2(\boldsymbol{\zeta})$.

One can define the so-called $\boldsymbol{\xi}$ -vector (introduced in Cahn and Hoffman (1974); Hoffman-Cahn (1972) for the sharp interfaces and in Wheeler and McFadden (1997) for the phase field approach) as

$$\boldsymbol{\xi} = \frac{\partial \beta(\nabla \eta)}{\partial \nabla \eta} = \frac{\partial \beta(\boldsymbol{\zeta})}{\partial \boldsymbol{\zeta}}. \quad (40)$$

Based on property of the homogeneous degree one functions, one can present $\beta(\nabla \eta) = \boldsymbol{\xi} \cdot \nabla \eta = \boldsymbol{\xi} \cdot \boldsymbol{\zeta}$. Also,

$$\rho_0 \frac{\partial \psi^\nabla}{\partial \nabla \eta} = \beta(\nabla \eta) \frac{\partial \beta(\nabla \eta)}{\partial \nabla \eta} = \beta(\boldsymbol{\zeta}) \frac{\partial \beta(\boldsymbol{\zeta})}{\partial \boldsymbol{\zeta}} = \beta(\boldsymbol{\zeta}) \boldsymbol{\xi}. \quad (41)$$

Then the counterpart of the Ginzburg-Landau equation (24) in the actual configuration and for single order parameter takes the form Taylor and Cahn (1998)

$$\begin{aligned} \dot{\eta} &= L \left(-\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho} \nabla \cdot \left(\rho \frac{\partial \bar{\psi}}{\partial \nabla \eta} \right) \right) = L \left(-\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho} \left(\frac{\partial \beta}{\partial \zeta_k} \frac{\partial \beta}{\partial \zeta_i} + \beta \frac{\partial^2 \beta}{\partial \zeta_k \partial \zeta_i} \right) \frac{\partial^2 \eta}{\partial r_k \partial r_i} \right) = \\ &= L \left(-\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho} \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta} \partial \boldsymbol{\zeta}} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r} \partial \mathbf{r}} \right). \end{aligned} \quad (42)$$

5.2. Current theory

With the function $\beta(\nabla_0 \eta) = |\nabla_0 \eta| \beta(\mathbf{k}_0)$ we present the gradient energy (36) in the form

$$\begin{aligned} \rho_0 \psi^\nabla &= 0.5 (\beta(\mathbf{k}_0) |\nabla \eta|)^2 = 0.5 \left(\beta(\nabla_0 \eta) \frac{|\nabla \eta|}{|\nabla_0 \eta|} \right)^2 = 0.5 \left(\beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}|}{|\boldsymbol{\zeta}_0|} \right)^2 \\ &= 0.5 \left(\beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|} \right)^2 = 0.5 \hat{\beta}^2(\boldsymbol{\zeta}_0); \quad \hat{\beta}(\boldsymbol{\zeta}_0) := \beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|} = \beta(\boldsymbol{\zeta}_0) \frac{(\boldsymbol{\zeta}_0 \cdot \mathbf{B} \cdot \boldsymbol{\zeta}_0)^{0.5}}{|\boldsymbol{\zeta}_0|}, \end{aligned} \quad (43)$$

where symmetric tensor $\mathbf{B} = \mathbf{F}^{-1} \cdot \mathbf{F}^{-1T} = (\mathbf{F}^T \cdot \mathbf{F})^{-1}$ is introduced, which is the strain measure inverse to the Cauchy-Green strain measure. Thus, in the reference configuration the expression for a new gradient energy function differs from the previous one by a scalar, which depends on $\boldsymbol{\zeta}_0$. If $\mathbf{F} = \mathbf{R} \cdot \mathbf{U}$ or $\mathbf{F} = \mathbf{R}_e \cdot \mathbf{U}_e \cdot \mathbf{U}_\theta \cdot \mathbf{U}_t$ (see Eq. (6) with $\mathbf{F}_e = \mathbf{R}_e \cdot \mathbf{U}_e$, where $\mathbf{R}_e$ is the proper orthogonal elastic rotation and $\mathbf{U}_e$ is the right-stretch elastic tensor),

then $\mathbf{B} = (\mathbf{U} \cdot \mathbf{U})^{-1}$ or $\mathbf{B} = (\mathbf{U}_t \cdot \mathbf{U}_\theta \cdot \mathbf{U}_e \cdot \mathbf{U}_e \cdot \mathbf{U}_\theta \cdot \mathbf{U}_t)^{-1}$. Thus, a correcting scalar and the entire theory in the reference configuration are independent of any rigid-body rotation in the actual configuration. One can introduce the $\boldsymbol{\xi}_0$ -vector in the reference configuration

$$\boldsymbol{\xi}_0 = \frac{\partial \hat{\beta}(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} = \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|} + \beta(\boldsymbol{\zeta}_0) \left[\frac{\mathbf{B} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0| |\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|} - \frac{\boldsymbol{\zeta}_0 |\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|^3} \right]. \quad (44)$$

Then

$$\begin{aligned} \rho_0 \frac{\partial \psi^\nabla}{\partial \nabla_0 \eta} &= \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_0} = \hat{\beta}(\boldsymbol{\zeta}_0) \boldsymbol{\xi}_0 = \beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|} \boldsymbol{\xi}_0 = \\ &= \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|^2}{|\boldsymbol{\zeta}_0|^2} + \beta^2(\boldsymbol{\zeta}_0) \left[\frac{\mathbf{B} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} - \frac{\boldsymbol{\zeta}_0 |\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|^2}{|\boldsymbol{\zeta}_0|^4} \right]. \end{aligned} \quad (45)$$

Note that if $\mathbf{F} = \mathbf{R}$, where $\mathbf{R}$ is an arbitrary orthogonal tensor, which does not change the length of the unit normal $\mathbf{k}_0$, then all ψ^∇ -related equations reduce to the same form as in the previous theories but in the reference configuration:

$$\begin{aligned} \rho_0 \psi^\nabla &= 0.5 \beta^2(\nabla_0 \eta) = 0.5 \beta^2(\boldsymbol{\zeta}_0); \quad \boldsymbol{\xi}_0 = \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0}; \\ \rho_0 \frac{\partial \psi^\nabla}{\partial \nabla_0 \eta} &= \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_0} = \beta(\boldsymbol{\zeta}_0) \boldsymbol{\xi}_0 = \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0}. \end{aligned} \quad (46)$$

For convenience of the evaluation of the divergence term in the Ginzburg-Landau equation we present Eq.(45) in the component form in the Cartesian coordinate system

$$\rho_0 \frac{\partial \psi^\nabla}{\partial (\partial \eta / \partial r_{0i})} = \rho_0 \frac{\partial \psi^\nabla}{\partial \zeta_{0i}} = \beta \frac{\partial \beta}{\partial \zeta_{0i}} \frac{\mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} + \beta^2 \left[\frac{\mathcal{B}_{ib} \zeta_{0b}}{\zeta_{0a} \zeta_{0a}} - \frac{\zeta_{0i} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{(\zeta_{0a} \zeta_{0a})^2} \right], \quad (47)$$

where for compactness we designate $\beta = \beta(\boldsymbol{\zeta}_0)$ (rather than $\beta = \beta(\mathbf{k}_0)$). Then we obtain

$$\begin{aligned} \frac{\partial}{\partial r_{0i}} \rho_0 \frac{\partial \psi^\nabla}{\partial \zeta_{0i}} &= \left(\frac{\partial \beta}{\partial \zeta_{0k}} \frac{\partial \beta}{\partial \zeta_{0i}} + \beta \frac{\partial^2 \beta}{\partial \zeta_{0k} \partial \zeta_{0i}} \right) \frac{\partial^2 \eta}{\partial r_{0k} \partial r_{0i}} \frac{\mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} + \\ &+ \beta \frac{\partial \beta}{\partial \zeta_{0i}} \left(\frac{\frac{\partial \mathcal{B}_{bc}}{\partial r_{0i}} \zeta_{0b} \zeta_{0c} + 2 \mathcal{B}_{bc} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} - \frac{2 \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\ &+ 2 \beta \frac{\partial \beta}{\partial \zeta_{0k}} \frac{\partial^2 \eta}{\partial \zeta_{0k} \partial \zeta_{0i}} \left(\frac{\mathcal{B}_{ib} \zeta_{0b}}{\zeta_{0a} \zeta_{0a}} - \frac{\zeta_{0i} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{(\zeta_{0a} \zeta_{0a})^2} \right) \end{aligned} \quad (48)$$

$$\begin{aligned}
& +\beta^2 \left(\frac{\frac{\partial \mathcal{B}_{ib}}{\partial r_{0i}} \zeta_{0b} + \mathcal{B}_{ib} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}}}{\zeta_{0a} \zeta_{0a}} - \frac{2 \mathcal{B}_{ib} \zeta_{0b} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\
& +\beta^2 \left(-\frac{\frac{\partial \mathcal{B}_{bc}}{\partial r_{0i}} \zeta_{0i} \zeta_{0b} \zeta_{0c} + \frac{\partial^2 \eta}{\partial r_{0i} \partial r_{0i}} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c} + 2 \zeta_{0i} \mathcal{B}_{bc} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}} \zeta_{0c}}{(\zeta_{0a} \zeta_{0a})^2} + 4 \frac{\zeta_{0i} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^3} \right),
\end{aligned}$$

where we explicitly show the second derivative of η . After some simplifications the Ginzburg-Landau equation (24) in the reference configuration takes the form

$$\begin{aligned}
\frac{\rho_0 \dot{\eta}}{L} = & -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_{0k}} \frac{\partial \beta}{\partial \zeta_{0i}} + \beta \frac{\partial^2 \beta}{\partial \zeta_{0k} \partial \zeta_{0i}} \right) \frac{\partial^2 \eta}{\partial r_{0k} \partial r_{0i}} \frac{\mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} + \\
& \beta \frac{\partial \beta}{\partial \zeta_{0i}} \left(\frac{\frac{\partial \mathcal{B}_{bc}}{\partial r_{0i}} \zeta_{0b} \zeta_{0c} + 4 \mathcal{B}_{bc} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} - \frac{4 \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\
& +\beta^2 \left(\frac{\frac{\partial \mathcal{B}_{ib}}{\partial r_{0i}} \zeta_{0b} + \mathcal{B}_{ib} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}}}{\zeta_{0a} \zeta_{0a}} - \frac{4 \mathcal{B}_{ib} \zeta_{0b} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\
& -\beta^2 \left(\frac{\frac{\partial \mathcal{B}_{bc}}{\partial r_{0i}} \zeta_{0i} \zeta_{0b} \zeta_{0c} + \frac{\partial^2 \eta}{\partial r_{0i} \partial r_{0i}} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c}}{(\zeta_{0a} \zeta_{0a})^2} - 4 \frac{\zeta_{0i} \mathcal{B}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^3} \right).
\end{aligned} \tag{49}$$

Eq.(49) can be written in the direct tensor notations:

$$\begin{aligned}
\frac{\rho_0 \dot{\eta}}{L} = & -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_0} \otimes \frac{\partial \beta}{\partial \zeta_0} + \beta \frac{\partial^2 \beta}{\partial \zeta_0 \partial \zeta_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \frac{\zeta_0 \cdot \mathbf{B} \cdot \zeta_0}{|\zeta_0|^2} + \\
& \beta \frac{\partial \beta}{\partial \zeta_0} \cdot \left(\frac{\zeta_0 \otimes \zeta_0 : \frac{\partial \mathbf{B}}{\partial \mathbf{r}_0} + 4 \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \cdot \mathbf{B} \cdot \zeta_0}{|\zeta_0|^2} - \frac{4 \zeta_0 \cdot \mathbf{B} \cdot \zeta_0 \zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^4} \right) \\
& +\beta^2 \left(\frac{\zeta_0 \cdot \frac{\partial \mathbf{B}}{\partial \mathbf{r}_0} : \mathbf{I} + \mathbf{B} : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^2} - \frac{4 (\mathbf{B} \cdot \zeta_0) \cdot \left(\zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \right)}{|\zeta_0|^4} \right) \\
& -\beta^2 \left(\frac{\zeta_0 \otimes \zeta_0 : \frac{\partial \mathbf{B}}{\partial \mathbf{r}_0} \cdot \zeta_0 + \nabla_0^2 \eta \zeta_0 \cdot \mathbf{B} \cdot \zeta_0}{|\zeta_0|^4} - 4 \frac{\zeta_0 \cdot \mathbf{B} \cdot \zeta_0 \zeta_0 \otimes \zeta_0 : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^6} \right).
\end{aligned} \tag{50}$$

Below, we present a more detailed expression for the local term in the Ginzburg-Landau equation obtained in Levitas (2014b):

$$-\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} = \mathbf{P}_e \cdot \mathbf{U}_t : \frac{\partial \mathbf{U}_\theta(\theta, \eta_k)}{\partial \eta} \cdot \mathbf{F}_e^T + \mathbf{P}_e : \frac{\partial \mathbf{U}_t(\eta_k)}{\partial \eta} \cdot \mathbf{U}_\theta \cdot \mathbf{F}_e^T - J_{t\theta} \frac{\partial \psi^e}{\partial \eta} \Big|_{\mathbf{F}_e} \tag{51}$$

$$-J_{t\theta}\psi^e \left(\mathbf{U}_t^{-1} : \frac{\partial \mathbf{U}_t}{\partial \eta} + \mathbf{U}_\theta^{-1} : \frac{\partial \mathbf{U}_\theta}{\partial \eta} \right) - J\rho_0 \frac{\partial \check{\psi}^\theta}{\partial \eta} - \rho_0 \frac{\partial \tilde{\psi}^\theta}{\partial \eta}.$$

It is clear that the interface stresses do not directly contribute to the Ginzburg-Landau equation. However, they change the distribution of the elastic stresses and affect the Ginzburg-Landau equation through them.

5.3. Some simplifications

Let us introduce the strain tensor associated with $\mathbf{B}$, namely $\mathbf{\mathcal{E}} = 0.5(\mathbf{I} - \mathbf{B})$. When deformations and rotations are absent, i.e., $\mathbf{F} = \mathbf{I}$, then $\mathbf{B} = \mathbf{I}$ and $\mathbf{\mathcal{E}} = \mathbf{0}$. Substituting $\mathbf{B} = \mathbf{I} - 2\mathbf{\mathcal{E}}$ in Eqs.(49)-(50) will allow us to eliminate some $\mathbf{B}$ -related terms which are not related to strain $\mathbf{\mathcal{E}}$ and also find expressions for small strain approximation. Indeed, after such a substitution, the Ginzburg-Landau equation (49) in the reference configuration is simplified to

$$\begin{aligned} \frac{\rho_0 \dot{\eta}}{L} = & -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_{0k}} \frac{\partial \beta}{\partial \zeta_{0i}} + \beta \frac{\partial^2 \beta}{\partial \zeta_{0k} \partial \zeta_{0i}} \right) \frac{\partial^2 \eta}{\partial r_{0k} \partial r_{0i}} \frac{(\delta_{bc} - 2\mathcal{E}_{bc}) \zeta_{0b} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} + \\ & 2\beta \frac{\partial \beta}{\partial \zeta_{0i}} \left(-\frac{\frac{\partial \mathcal{E}_{bc}}{\partial r_{0i}} \zeta_{0b} \zeta_{0c} + 4\mathcal{E}_{bc} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}} \zeta_{0c}}{\zeta_{0a} \zeta_{0a}} + \frac{4\mathcal{E}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\ & - 2\beta^2 \left(\frac{\frac{\partial \mathcal{E}_{ib}}{\partial r_{0i}} \zeta_{0b} + \mathcal{E}_{ib} \frac{\partial^2 \eta}{\partial r_{0b} \partial r_{0i}}}{\zeta_{0a} \zeta_{0a}} - \frac{4\mathcal{E}_{ib} \zeta_{0b} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^2} \right) \\ & + 2\beta^2 \left(\frac{\frac{\partial \mathcal{E}_{bc}}{\partial r_{0i}} \zeta_{0i} \zeta_{0b} \zeta_{0c} + \frac{\partial^2 \eta}{\partial r_{0i} \partial r_{0i}} \mathcal{E}_{bc} \zeta_{0b} \zeta_{0c}}{(\zeta_{0a} \zeta_{0a})^2} - 4 \frac{\zeta_{0i} \mathcal{E}_{bc} \zeta_{0b} \zeta_{0c} \zeta_{0a} \frac{\partial^2 \eta}{\partial r_{0a} \partial r_{0i}}}{(\zeta_{0a} \zeta_{0a})^3} \right). \end{aligned} \quad (52)$$

Eq.(52) can be presented in the direct tensor notations:

$$\begin{aligned} \frac{\rho_0 \dot{\eta}}{L} = & -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \frac{\boldsymbol{\zeta}_0 \cdot (\mathbf{I} - 2\mathbf{\mathcal{E}}) \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \\ & 2\beta \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \cdot \left(-\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \mathbf{\mathcal{E}}}{\partial \mathbf{r}_0} + 4 \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \cdot \mathbf{\mathcal{E}} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \frac{4\boldsymbol{\zeta}_0 \cdot \mathbf{\mathcal{E}} \cdot \boldsymbol{\zeta}_0 \zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^4} \right) \end{aligned}$$

$$\begin{aligned}
& -2\beta^2 \left(\frac{\boldsymbol{\zeta}_0 \cdot \frac{\partial \boldsymbol{\mathcal{E}}}{\partial \mathbf{r}_0} : \mathbf{I} + \boldsymbol{\mathcal{E}} : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^2} - \frac{4(\boldsymbol{\mathcal{E}} \cdot \boldsymbol{\zeta}_0) \cdot \left(\boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \right)}{|\boldsymbol{\zeta}_0|^4} \right) \\
& + 2\beta^2 \left(\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\mathcal{E}}}{\partial \mathbf{r}_0} \cdot \boldsymbol{\zeta}_0 + \nabla_0^2 \eta \boldsymbol{\zeta}_0 \cdot \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^4} - 4 \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^6} \right). \quad (53)
\end{aligned}$$

Now it is clear that at $\boldsymbol{\mathcal{E}} = \mathbf{0}$ and coinciding reference and actual configurations, Eqs. (52)-(53) reduce to Eq. (42). If $\mathbf{F} = a\mathbf{I}$ is a volumetric expansion/contraction, then $\mathbf{B} = a^{-2}\mathbf{I}$, $\boldsymbol{\mathcal{E}} = 0.5(1 - a^{-2})\mathbf{I}$, and Eqs. (43)-(45) and (52)-(53) simplify:

$$\rho_0 \psi^\nabla = 0.5 (\beta(\nabla_0 \eta) a^{-1})^2; \quad \boldsymbol{\xi}_0 = \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} a^{-1}; \quad \rho_0 \frac{\partial \psi^\nabla}{\partial \nabla_0 \eta} = \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} a^{-2}. \quad (54)$$

$$\frac{\rho_0 \dot{\eta}}{L} = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_{0k}} \frac{\partial \beta}{\partial \zeta_{0i}} + \beta \frac{\partial^2 \beta}{\partial \zeta_{0k} \partial \zeta_{0i}} \right) \frac{\partial^2 \eta}{\partial r_{0k} \partial r_{0i}} a^{-2} + \beta \frac{\partial \beta}{\partial \zeta_{0i}} \frac{\partial a^{-2}}{\partial r_{0i}}; \quad (55)$$

$$\frac{\rho_0 \dot{\eta}}{L} = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} a^{-2} + \beta \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \cdot \frac{\partial a^{-2}}{\partial \mathbf{r}_0}. \quad (56)$$

All terms in Eqs.(52)-(53) that disappeared for pure dilatational strain are proportional to a tensor characterizing change in shape. For small changes in shape, they are proportional to the deviatoric strain.

5.4. Small strain and rotation approximation

Using the same approximation for small strains and rotations as in Section 2, i.e., $\mathbf{F} \simeq \mathbf{I} + \boldsymbol{\varepsilon} + \boldsymbol{\omega}$, one evaluates $\mathbf{F}^{-1} \simeq \mathbf{I} - \boldsymbol{\varepsilon} - \boldsymbol{\omega}$, $\mathbf{F}^{-1T} \simeq \mathbf{I} - \boldsymbol{\varepsilon} + \boldsymbol{\omega}$, $\mathbf{B} \simeq \mathbf{I} - 2\boldsymbol{\varepsilon}$ and $\boldsymbol{\mathcal{E}} \simeq \boldsymbol{\varepsilon}$, where all products of small tensors are neglected. The tensor $\mathbf{B}$ is independent of rotation, as it is expected from the finite strain theory. Some simplifications can be found in the definitions. Indeed, for such an approximation, we obtain

$$\rho_0 \psi^\nabla = 0.5(\beta(\boldsymbol{\zeta}_0))^2 - (\beta(\boldsymbol{\zeta}_0))^2 \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0}. \quad (57)$$

To evaluate vector $\boldsymbol{\xi}_0$, we utilize the Taylor expansion

$$\begin{aligned} |\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}| &= (\boldsymbol{\zeta}_0 \cdot \mathbf{B} \cdot \boldsymbol{\zeta}_0)^{0.5} \simeq (\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)^{0.5} - \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{(\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)^{0.5}}; \\ |\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|^{-1} &= (\boldsymbol{\zeta}_0 \cdot \mathbf{B} \cdot \boldsymbol{\zeta}_0)^{-0.5} \simeq (\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)^{-0.5} + \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{(\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)^{1.5}}. \end{aligned} \quad (58)$$

Then

$$\boldsymbol{\xi}_0 = \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} - 2 \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \left(\frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} \right)^{0.5} - 2 \beta(\boldsymbol{\zeta}_0) \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon}}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0}. \quad (59)$$

and

$$\begin{aligned} \rho_0 \frac{\partial \psi^\nabla}{\partial \nabla_0 \eta} &= \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_0} = \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \\ &- 2 \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} + 2 (\beta(\boldsymbol{\zeta}_0))^2 \frac{\boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} \cdot (\mathbf{I} \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} - \boldsymbol{\varepsilon}). \end{aligned} \quad (60)$$

It is clear that the Ginzburg-Landau equations at small strains can be trivially obtained from Eqs.(52)-(53) by substituting the finite strain $\boldsymbol{\mathcal{E}}$ with the small strain $\boldsymbol{\varepsilon}$ and neglecting $\boldsymbol{\varepsilon}$ in comparison with $\mathbf{I}$:

$$\begin{aligned} \frac{\rho_0 \dot{\eta}}{L} &= -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} + \\ &2 \beta \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \cdot \left(-\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\mathcal{E}}}{\partial \mathbf{r}_0} + 4 \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \frac{4 \boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^4} \right) \\ &- 2 \beta^2 \left(\frac{\boldsymbol{\zeta}_0 \cdot \frac{\partial \boldsymbol{\mathcal{E}}}{\partial \mathbf{r}_0} : \mathbf{I} + \boldsymbol{\varepsilon} : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^2} - \frac{4 (\boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0) \cdot \left(\boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \right)}{|\boldsymbol{\zeta}_0|^4} \right) \\ &+ 2 \beta^2 \left(\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\mathcal{E}}}{\partial \mathbf{r}_0} \cdot \boldsymbol{\zeta}_0 + \nabla_0^2 \eta \boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^4} - 4 \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^6} \right). \end{aligned} \quad (61)$$

For small strains, $\mathbf{P} \simeq \boldsymbol{\sigma}$, $\rho \simeq \rho_0$ and the local term (51) in the Ginzburg-Landau equation simplifies to

$$-\rho \frac{\partial \bar{\psi}}{\partial \eta} = \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_\theta(\theta, \eta_k)}{\partial \eta} + \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_t(\eta_k)}{\partial \eta} - \frac{\partial \psi^e}{\partial \eta} \Big|_{\boldsymbol{\varepsilon}_e} - \psi^e \left(\frac{\partial \boldsymbol{\varepsilon}_{t0}}{\partial \eta} + \frac{\partial \boldsymbol{\varepsilon}_{\theta 0}}{\partial \eta} \right) - \rho \frac{\partial \check{\psi}^\theta}{\partial \eta} - \rho \frac{\partial \tilde{\psi}^\theta}{\partial \eta}. \quad (62)$$

In the simplest quadratic approximation one has

$$\rho\psi^e = 0.5\boldsymbol{\varepsilon}_e:\mathbf{C}(\eta):\boldsymbol{\varepsilon}_e; \quad \boldsymbol{\sigma}_e = \mathbf{C}(\eta):\boldsymbol{\varepsilon}_e, \quad (63)$$

where $\mathbf{C}$ is the fourth-rank tensor of elastic moduli. Substituting this expression in Eq.(61) allows us to analyze the importance of each strain-related term to the driving force for change in η . Thus, the contribution in Eq.(61) due to anisotropy of the interface energy (the last three terms) is a linear function of the small magnitude of the strain tensor. The first three terms in Eq.(62) (the work of stresses on change in transformation and thermal strains and change in elastic energy due to change in elastic moduli) are quadratic in the small magnitude of the strain tensor. And finally, the fourth term in Eq.(62) proportional to elastic energy is cubic in the magnitude of the strain tensor. Thus, surprisingly, the first order correction in the Ginzburg-Landau equation due to strains is related to the anisotropy of the interface energy (which was completely neglected) rather than being due to mechanical work. Also, one has to take into account that usually elastic and thermal strains are at least an order of magnitude lower than the transformation and total strains, i.e., $\boldsymbol{\varepsilon}_e \sim \boldsymbol{\varepsilon}_\theta \sim \boldsymbol{\varepsilon}_t^2 \sim \boldsymbol{\varepsilon}^2$. Then

(a) the terms due to anisotropy of the interface energy are the first order of smallness for small total strains, i.e., $\mathbf{O}(\boldsymbol{\varepsilon})$;

(b) the term with the transformation strain is the third order of smallness, $\mathbf{O}(\boldsymbol{\varepsilon}^3)$;

(c) the stress work on the thermal strain and the term related to change in elastic moduli are $\mathbf{O}(\boldsymbol{\varepsilon}^4)$,

(d) and the contribution proportional to ψ^e is $\mathbf{O}(\boldsymbol{\varepsilon}^5)$ for the term with the transformation strain and $\mathbf{O}(\boldsymbol{\varepsilon}^6)$ for the term containing the thermal strain. The magnitude of each of these terms of course depends on the corresponding coefficients, which may change our conclusions for small but not infinitesimal strain. Thus, for small anisotropy but high stresses the importance of the contributions due to anisotropy of the interface energy and transformation work may change.

When all terms with strains related to anisotropy of the gradient energy are neglected, the Ginzburg-Landau equation simplifies to Eq.(42) with $\frac{\partial \bar{\psi}}{\partial \eta}$ given by Eq.(62).

5.5. Specification for cubic crystals

As an example, we consider the orientation dependence of the interface energy $\gamma(\mathbf{k}_0)$ as suggested for crystal growth in Qin and Bhadeshia (2009), which describes results of molecular dynamics simulations for two dozen cubic metals with reasonable accuracy. Since we will find below (see Eq.(97)) that function $\beta(\mathbf{k}_0) = Z\gamma(\mathbf{k}_0)$, where Z is a factor defined by Eq.(97), we will use the same function for $\beta(\mathbf{k}_0)$:

$$\beta(\mathbf{k}_0) = \alpha_0 + \alpha_1 \frac{\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2}{|\boldsymbol{\zeta}_0|^4} + \alpha_2 \frac{\zeta_{0i}^2 \zeta_{0j}^2 \zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^6} + \alpha_3 \frac{(\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)^2}{|\boldsymbol{\zeta}_0|^8}. \quad (64)$$

Parameters α_i can be determined based on similar parameters for $\gamma(\mathbf{k}_0)$ given in Qin and Bhadeshia (2009); here and below $i \neq j \neq k$ and there is no summation over these indices. Then

$$\beta(\boldsymbol{\zeta}_0) = \alpha_0 |\boldsymbol{\zeta}_0| + \alpha_1 \frac{\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2}{|\boldsymbol{\zeta}_0|^3} + \alpha_2 \frac{\zeta_{0i}^2 \zeta_{0j}^2 \zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^5} + \alpha_3 \frac{(\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)^2}{|\boldsymbol{\zeta}_0|^7}, \quad (65)$$

The components of $\boldsymbol{\zeta}_0$ can be considered as Millers indices of crystallographic planes. Derivatives of β -related terms in Eqs. (52)-(53) are given in Appendix. For a two-dimensional case one uses $\zeta_{0j} = 0$; the terms with α_2 in this case disappears.

An example of function $\gamma(\mathbf{k}_0) = \beta(\mathbf{k}_0)/Z$ for Li for which $\alpha_0 = 0.443$, $\alpha_1 = 1.164$, $\alpha_2 = 4.555$, and $\alpha_3 = 1.123$ in Eq.(64) (see Qin and Bhadeshia (2009)), all in J/m^2 , is presented in Fig. 3a. Its cross sections within [110] and [100] planes are shown in Figs. 4a and 5a. These figures also contain plots $\gamma^2(\mathbf{k}_0) = \beta^2(\mathbf{k}_0)/Z^2 = 2\psi^\nabla/(Z^2|\boldsymbol{\zeta}|^2) = \sigma_{st}/(Z^2|\boldsymbol{\zeta}|^2)$, where σ_{st} is the magnitude of biaxial interface tension according to Eq.(39). All functions show strong and nontrivial anisotropy, and the anisotropy of interface stress is substantial.

Figure 3: a) Orientation dependence of the function $\gamma(\mathbf{k}_0) = \beta(\mathbf{k}_0)/Z$ for Li, which is similar to that for interface width $\delta(\mathbf{k}_0)$. b) Orientation dependence of the function $1/\gamma(\mathbf{k}_0)$ that exhibits concave regions, which should be regularized with the planes. c) Orientation dependence of the function $\gamma^2(\mathbf{k}_0) = \beta^2(\mathbf{k}_0)/Z^2$, which is equal to the gradient energy $\psi^\nabla(\mathbf{k}_0)$ and magnitude of the biaxial interface tension $\sigma_{st}(\mathbf{k}_0)$ normalized by $0.5Z^2|\zeta|^2$ and $Z^2|\zeta|^2$, respectively.

6. Boundary conditions for order parameters

One of the possible boundary conditions, Neumann, is obtained through the prescription of the normal to the external surface component of the generalized force $\mathbf{Q}_{0i}^\eta$, which was introduced at the external surface (see Eq.(20)₂), similar to the heat or diffusion flux:

$$\mathbf{n}_0 \cdot \mathbf{Q}_{0i}^\eta = \mathbf{n}_0 \cdot \rho_0 \frac{\partial \psi}{\partial \nabla_0 \eta_i} = H_{0i}, \quad (66)$$

where H_{0i} are some given functions. Substituting $\frac{\partial \psi}{\partial \nabla_0 \eta_i}$ from Eq.(45), we obtain a more explicit form:

$$\mathbf{n}_0 \cdot \rho_0 \left[\beta(\zeta_0) \frac{\partial \beta(\zeta_0)}{\partial \zeta_0} \frac{\zeta_0 \cdot \mathbf{B} \cdot \zeta_0}{|\zeta_0|^2} + (\beta(\zeta_0))^2 \left(\frac{\mathbf{B} \cdot \zeta_0}{|\zeta_0|^2} - \frac{\zeta_0 \zeta_0 \cdot \mathbf{B} \cdot \zeta_0}{|\zeta_0|^4} \right) \right] = H_0. \quad (67)$$

Utilizing $\mathbf{B} = \mathbf{I} - 2\mathbf{\mathcal{E}}$, we transform

$$\mathbf{n}_0 \cdot \rho_0 \left[\beta(\zeta_0) \frac{\partial \beta(\zeta_0)}{\partial \zeta_0} \frac{\zeta_0 \cdot (\mathbf{I} - 2\mathbf{\mathcal{E}}) \cdot \zeta_0}{|\zeta_0|^2} - 2(\beta(\zeta_0))^2 \left(\frac{\mathbf{\mathcal{E}} \cdot \zeta_0}{|\zeta_0|^2} - \frac{\zeta_0 \zeta_0 \cdot \mathbf{\mathcal{E}} \cdot \zeta_0}{|\zeta_0|^4} \right) \right] = H_0. \quad (68)$$

Figure 4: Orientation dependence of some functions for Li within $\{110\}$ plane. a) Function $\gamma(\mathbf{k}_0) = \beta(\mathbf{k}_0)/Z$, which is similar to that for interface width $\delta(\mathbf{k}_0)$; b) function $\gamma^2(\mathbf{k}_0) = \beta^2(\mathbf{k}_0)/Z^2$, which is equal to the normalized gradient energy $2\psi^\nabla(\mathbf{k}_0)/(Z^2|\boldsymbol{\zeta}|^2)$ and magnitude of the biaxial interface tension $\sigma_{st}(\mathbf{k}_0)/(Z^2|\boldsymbol{\zeta}|^2)$; c) function $1/\gamma(\mathbf{k}_0)$ that exhibits concave regions, which should be regularized with the planes, and d) artificial normalized shear stress $\bar{\tau}_{31} = \tau_{31}/(Z^2|\boldsymbol{\zeta}|^2)$, which is present in previous theories and is eliminated here.

Figure 5: Orientation dependence of some functions for Li within $\{100\}$ plane. a) Function $\gamma(\mathbf{k}_0) = \beta(\mathbf{k}_0)/Z$, which is similar to that interface width $\delta(\mathbf{k}_0)$; b) function $\gamma^2(\mathbf{k}_0) = \beta^2(\mathbf{k}_0)/Z^2 = 2\psi^\nabla(\mathbf{k}_0)/(Z^2|\boldsymbol{\zeta}|^2) = \sigma_{st}(\mathbf{k}_0)/(Z^2|\boldsymbol{\zeta}|^2)$; c) function $1/\gamma(\mathbf{k}_0)$ that exhibits concave regions, which should be regularized with the planes, and d) artificial shear normalized shear stress $\bar{\tau}_{31} = \tau_{31}/(Z^2|\boldsymbol{\zeta}|^2)$, which is present in previous theories and is eliminated in the current work.

If $\mathbf{F} = a\mathbf{I}$, then $\mathbf{B} = a^{-2}\mathbf{I}$, $\boldsymbol{\mathcal{E}} = 0.5(1 - a^{-2})\mathbf{I}$, and

$$\mathbf{n}_0 \cdot \rho_0 \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} a^{-2} = H_0. \quad (69)$$

If strains $\boldsymbol{\mathcal{E}}$ are negligible with unity, Eq.(68) simplifies to the following form:

$$\mathbf{n}_0 \cdot \rho_0 \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} = H_0. \quad (70)$$

Alternatively, one can prescribe $\eta_i = \text{const}_i$ (in particular, 0 or 1) or periodic boundary conditions for η_i , or mixed boundary conditions. A common model choice is $H_{0i} = -\frac{\partial \gamma_{0s}(\eta_i)}{\partial \eta_i}$ (Levitas and Javanbakht (2010); Levitas and Samani (2011a,b); Levitas (2014b); Lipowsky (1982); Pluis et al. (1990)), where $\gamma_{0s}(\eta_i)$ is the surface energy per unit reference area. Dependence of the surface energy on the order parameter is due to different surface energies of the parent and product phases. This leads to surface-induced pre-transformations and barrierless nucleation (Levitas et al. (2006b); Levitas and Javanbakht (2010); Levitas

and Samani (2011a,b); Lipowsky (1982); Pluis et al. (1990)), which have been studied either without deformation or for small strains. A more detailed approach is related to the introduction of the finite-width external surface with the help of an additional order parameter that describes a stationary solid-exterior interface (Levitas and Janbakht (2011b); Levitas and Samani (2014)). This leads to various nontrivial scale and mechanics effects and morphological transitions in surface-induced PTs.

7. Previous expression for structural stresses

Previous theories (Anderson et al. (2000); Debierre et al. (2003); Kobayashi (1993); Taylor and Cahn (1998, 1994); Warren-Boettinger (1995); Wheeler and McFadden (1997)) posited that

$$\rho_0 \psi^\nabla = 0.5 \beta^2(\mathbf{k}) |\nabla \eta|^2 = 0.5 \beta^2(\nabla \eta) = 0.5 \beta^2(\zeta). \quad (71)$$

According to Eq. (27) this leads to the following expression for structural stresses:

$$\begin{aligned} \sigma_{st}^p &= (\rho_0 \check{\psi}^\theta + 0.5 \beta^2(\mathbf{k}) |\nabla \eta|^2) \mathbf{I} - \beta(\nabla \eta) \nabla \eta \otimes \frac{\partial \beta(\nabla \eta)}{\partial \nabla \eta} = \\ &\beta^2(\nabla \eta) (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho_0 \check{\psi}^\theta - 0.5 \beta^2(\nabla \eta)) \mathbf{I} - \sigma_{st}^{dif}, \end{aligned} \quad (72)$$

where superscript p is for previous and

$$\sigma_{st}^{dif} := \beta(\nabla \eta) \nabla \eta \otimes \frac{\partial \beta(\nabla \eta)}{\partial \nabla \eta} - \beta^2(\mathbf{k}) \nabla \eta \otimes \nabla \eta = \beta(\zeta) \zeta \otimes \frac{\partial \beta(\zeta)}{\partial \zeta} - \beta^2(\mathbf{k}) \zeta \otimes \zeta \quad (73)$$

is the difference between the previous theory and the more correct result given in Eq.(38) in the limit of infinitesimal strains. Let us choose the local coordinate system in which axis 3 coincides with the normal $\mathbf{k}$ and mutually orthogonal axes 1 and 2 with unit vectors $\mathbf{t}$ and $\mathbf{p}$ are orthogonal to 3 and located within interface (Fig. 1a). As it will be shown below, function $\check{\psi}^\theta$ can be chosen in a way that the term $\rho_0 \check{\psi}^\theta - 0.5 \beta^2(\nabla \eta) = 0$ at the

moving interface and without σ_{st}^{dif} the interface stress σ_{st} represents equal biaxial tension in directions 1 and 2. Let us evaluate σ_{st}^{dif} . The term $\beta^2(\mathbf{k})\zeta \otimes \zeta$ has only one component 33 and the term $\beta(\zeta)\zeta \otimes \frac{\partial\beta(\zeta)}{\partial\zeta}$ possesses three components: normal component 33 and shear stresses τ_{31} and τ_{32} , which are located at planes within the interface (i.e., orthogonal to axes 1 and 2) and directed along the axis 3. Multiplying Eq.(73) by ζ from the right we obtain

$$\sigma_{st}^{dif} \cdot \zeta = \beta(\zeta)\zeta \frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \zeta - \beta^2(\mathbf{k})\zeta|\zeta|^2 = \beta^2(\zeta)\zeta = \beta^2(\zeta)\zeta = \mathbf{0}, \quad (74)$$

where we take into account that $\frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \zeta = \beta(\zeta)$. Thus, the 33 component of σ_{st}^{dif} is zero and only extra shear stresses,

$$\tau_{31} = \mathbf{k} \cdot \beta(\zeta)\zeta \frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \mathbf{t} = \beta(\zeta)|\zeta| \frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \mathbf{t}; \quad \tau_{32} = \mathbf{k} \cdot \beta(\zeta)\zeta \frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \mathbf{p} = \beta(\zeta)|\zeta| \frac{\partial\beta(\zeta)}{\partial\zeta} \cdot \mathbf{p}, \quad (75)$$

appear (Fig. 1c). Since $\tau_{13} = \tau_{23} = 0$, according to Eq.(151) the shear stress τ_{31} produces a moment about axis 2 and shear stress τ_{32} produces a moment about axis 1. Orientation dependence of the magnitude of the biaxial tension for Li is presented in Fig. 3 for 3D and in Figs. 4b and 5b for $\{110\}$ and $\{100\}$ planes, respectively. To plot Figs. 3 for 3D and Figs. 3-5 for 2D, the unit vector $\mathbf{k}$ first was expressed in a spherical coordinate system, in which

$$k_{01} = \sin \Theta \cos \Phi; \quad k_{02} = \sin \Theta \sin \Phi; \quad k_{03} = \cos \Theta, \quad (76)$$

where Θ and Φ are angles between the Cartesian axes x_3 and x_1 along the cubic directions, respectively. For $\{110\}$ plane $\Phi = \pi/4$ and for $\{100\}$ plane $\Phi = 0$. In these planes the unit vector $\mathbf{t}$ is inclined under the angle $\pi/2 - \Theta$ to the axis x_3 . Then

$$\tau_{31}^{\{110\}} = |\zeta|^2 \beta(\mathbf{k}) \left(\left(\frac{\partial\beta(\zeta)}{\partial\zeta_1} + \frac{\partial\beta(\zeta)}{\partial\zeta_2} \right) \frac{\cos \Theta}{\sqrt{2}} - \frac{\partial\beta(\zeta)}{\partial\zeta_3} \sin \Theta \right) \quad (77)$$

for $\{110\}$ plane and

$$\tau_{31}^{\{100\}} = |\zeta|^2 \beta(\mathbf{k}) \left(\frac{\partial\beta(\zeta)}{\partial\zeta_2} \cos \Theta - \frac{\partial\beta(\zeta)}{\partial\zeta_3} \sin \Theta \right) \quad (78)$$

for $\{100\}$ plane. Utilizing Eqs.(64) and (152), normalized shear stresses are plotted in Figs. 4d and 5d. The maximum magnitude of the shear stress is quite large, equal to 0.75 of the maximum interface tension.

8. Regularization of $\beta(\zeta)$ for strong anisotropy

Figure 6: Schematics of the functions $\beta(\mathbf{k}_0)$ (a) and $1/\beta(\mathbf{k}_0)$ (b) within a plane. As the Ginzburg-Landau equation becomes ill-posed when the function $1/\beta(\mathbf{k}_0)$ is nonconvex for some set of $\mathbf{k}_0$, this part of the function $1/\beta(\mathbf{k}_0)$ is regularized with a plane. The concave part of $1/\beta(\mathbf{k}_0)$ corresponds to the points in $\beta(\mathbf{k}_0)$, which cannot be touched by a circle (a sphere) $\mathbf{B} \cdot \mathbf{k}_0$ plotted on some vector $\mathbf{B}$ as on a diameter, without intersecting $\beta(\mathbf{k}_0)$ at other points. The regularizing plane in $1/\beta(\mathbf{k}_0)$ corresponds to the circle $\beta_c(\mathbf{k}_0) = \mathbf{g} \cdot \mathbf{k}_0$, plotted on the constant vector $\mathbf{g}$ as on diameter, where $\mathbf{g}$ is orthogonal to the regularizing plane.

For strong anisotropy, some orientations with high interface energy are not present in the equilibrium shape of the transformed phase. Formally, the Ginzburg-Landau equation becomes ill-posed when the inverse function $1/\beta(\mathbf{k}_0)$ becomes nonconvex for some set of $\mathbf{k}_0$ (Eggleston et al. (2001); Lin et al. (2011); Taylor and Cahn (1998)) (Fig. 6b). That is why we included plots $1/\beta(\mathbf{k}_0) = 1/(Z\gamma(\mathbf{k}_0))$ in Figs. 3-5. For convex $1/\gamma(\mathbf{k}_0)$ a sphere $\mathbf{B} \cdot \mathbf{k}_0$, plotted on some vector $\mathbf{B}$ as on a diameter, can touch the surface $\gamma(\mathbf{k}_0)$ for any $\mathbf{k}_0$ without

intersecting $\gamma(\mathbf{k}_0)$ at other points. For nonconvex $1/\gamma(\mathbf{k}_0)$, a sphere $\mathbf{B} \cdot \mathbf{k}_0$ cannot touch the surface $\gamma(\mathbf{k}_0)$ for those $\mathbf{k}_0$ that correspond to the nonconvex part of $1/\gamma(\mathbf{k}_0)$ without intersecting $\gamma(\mathbf{k}_0)$ at other points (Fig. 6a). To regularize the problem for vectors $\mathbf{k}_0$ for which $1/\gamma(\mathbf{k}_0)$ is nonconvex it is substituted with a common tangent plane $\frac{1}{\gamma_c(\mathbf{k}_0)} = \frac{A}{\mathbf{s} \cdot \mathbf{k}_0}$, where $\mathbf{s}$ is the unit vector orthogonal to this plane and A is the distance from the origin to this plane (Fig. 6b), and corresponding $\beta_c(\mathbf{k}_0)$. This plane corresponds to a sphere $\gamma_c(\mathbf{k}_0) = \bar{\mathbf{g}} \cdot \mathbf{k}_0$ plotted on the constant vector $\bar{\mathbf{g}} := \mathbf{s}/A$ as on a diameter, which substitutes $\gamma(\mathbf{k}_0)$ for those $\mathbf{k}_0$ for which this sphere cannot touch $\gamma(\mathbf{k}_0)$ without intersecting it at other points (Fig. 6a). For such a $\gamma_c(\mathbf{k}_0) = \bar{\mathbf{g}} \cdot \mathbf{k}_0$ and corresponding $\beta_c(\mathbf{k}_0) = \mathbf{g} \cdot \mathbf{k}_0$ with $\mathbf{g} = Z\bar{\mathbf{g}}$, all equations are simplified, because

$$\beta_c(\boldsymbol{\zeta}_0) = \mathbf{g} \cdot \boldsymbol{\zeta}_0 = |\mathbf{g}||\boldsymbol{\zeta}_0| \cos \vartheta; \quad \frac{\partial \beta_c}{\partial \boldsymbol{\zeta}_0} = \mathbf{g}; \quad \frac{\partial^2 \beta_c}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} = \mathbf{0}, \quad (79)$$

where ϑ is the angle between the vectors $\mathbf{g}$ and $\boldsymbol{\zeta}_0$. Then

$$\begin{aligned} \tau_{31} &= \beta_c(\boldsymbol{\zeta}_0) |\boldsymbol{\zeta}_0| \mathbf{g} \cdot \mathbf{t} = |\mathbf{g}||\boldsymbol{\zeta}_0|^2 \cos \vartheta |\mathbf{g}| \cos(\pi/2 + \vartheta) = -0.5 |\mathbf{g}|^2 |\boldsymbol{\zeta}_0|^2 \sin 2\vartheta; \\ \sigma_{st} &= \beta_c(\boldsymbol{\zeta}_0)^2 = |\mathbf{g}|^2 |\boldsymbol{\zeta}_0|^2 \cos^2 \vartheta; \quad \frac{|\tau_{31}|}{\sigma_{st}} = \tan \vartheta, \end{aligned} \quad (80)$$

where we consider that the normal $\mathbf{t}$ to the plane at which shear stress τ_{31} acts is coplanar with vectors $\mathbf{g}$ and $\boldsymbol{\zeta}_0$. Thus, τ_{31} is zero for the direction of collinear $\mathbf{g}$ and $\boldsymbol{\zeta}_0$, and the ratio $|\tau_{31}|/\sigma_{st}$ increases with increasing ϑ . Maximum ϑ is determined by the points in which the surface $1/\gamma(\mathbf{k}_0)$ and the regularizing plane touch. For the vertical regularizing line in Fig. 6b, the maximum value of ϑ is defined by equation $d(\cos \vartheta / \beta(\vartheta)) = 0$. For Li $\vartheta_{max} = 0.642$ and $\tan \vartheta_{max} = 0.75$ for $\{100\}$ plane and $\vartheta_{max} = 0.476$ and $\tan \vartheta_{max} = 0.52$ for $\{110\}$ plane.

9. Specification for a single order parameter

All previous results for multiple order parameters can be simplified for a single order parameter just by dropping the subscripts for η_i and L_{ij} . Here we present a specific 2 – 4 – 6

polynomial for the Helmholtz free energy with the general structure in Eq.(25):

$$\bar{\psi}(\mathbf{F}, \eta, \theta, \nabla\eta) = \frac{J_{t\theta}}{\rho_0} \psi^e(\mathbf{F}_e, \eta, \theta) + J\check{\psi}^\theta + \tilde{\psi}^\theta + J\psi^\nabla(\nabla\eta, \nabla_0\eta), \quad (81)$$

where $\psi^\nabla(\nabla\eta, \nabla_0\eta)$ is defined by Eq.(36). When surface stress is neglected, i.e., $J = 1$, the thermal part of free energy should reduce to that in Levitas et al. (2003); Levitas (2013a):

$$\check{\psi}^\theta + \tilde{\psi}^\theta = f(\theta, \eta) = G_H^\theta(\theta) + \Delta G^\theta(\theta)\eta^4(3 - 2\eta^2) + 0.5A(\theta)\eta^2(1 - \eta^2)^2; \quad 0 \leq \eta \leq 1. \quad (82)$$

Here, the terms $\Delta G^\theta(\theta)\eta^4(3 - 2\eta^2)$ and $0.5A(\theta)\eta^2(1 - \eta^2)^2$ are parts of the thermal (chemical) energy $f(\theta, \eta)$ related to the thermal driving force for the PT and the double-well barrier, respectively. Function $G_H^\theta(\theta)$ is the thermal part of the free energy of **H**, which is important for the determination of entropy, specific heat, and temperature evolution in **H**. However, for our purposes below we can set $G_H^\theta(\theta) = 0$ without loss of generality, which is equivalent to operating with an excess energy $f(\theta, \eta) - G_A^\theta(\theta)$. The importance of the proper division of $f(\theta, \eta)$ into two functions, $\check{\psi}^\theta$ and $\tilde{\psi}^\theta$, is related to the fact that $\check{\psi}^\theta$ contributes to the interface stresses Eq.(27) but $\tilde{\psi}^\theta$ does not. Eq.(38) for stresses reduces to

$$\boldsymbol{\sigma}_{st} = (\beta(\mathbf{k}_0))^2 |\nabla\eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho_0 \check{\psi}^\theta - 0.5(\beta(\mathbf{k}_0))^2 |\nabla\eta|^2) \mathbf{I}. \quad (83)$$

To complete the model and obtain desired biaxial surface tension the term $\check{\psi}^\theta(\theta, \eta)$ should be defined in a way that it eliminates the second terms in Eq.(83) for the propagating interface. It is independent of strains, so we will treat the simplest version of the Ginzburg-Landau equation Eq.(42) for *neglected stresses and elastic strain*:

$$\dot{\eta} = \bar{L}(\mathbf{k}) \left(-\rho_0 \frac{\partial f}{\partial \eta} + \left(\frac{\partial \beta}{\partial \nabla\eta} \otimes \frac{\partial \beta}{\partial \nabla\eta} + \beta \frac{\partial^2 \beta}{\partial \nabla\eta \partial \nabla\eta} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r} \partial \mathbf{r}} \right), \quad (84)$$

where $\bar{L} = L/\rho_0$ is the kinetic coefficient. Let us consider an interface propagating in an arbitrary chosen direction of the interface normal $\mathbf{k}$ and direct axis x of the Cartesian coordinate system along $\mathbf{k}$. If $\mathbf{k}$ does not correspond to the local minimum of the interface energy,

then the interface will rotate toward to the close minimum energy orientation. However, we will forbid interface rotation by fixing $\mathbf{k}$ in time and assuming that η depends on x only. The main point is to find interface parameters for an arbitrary $\mathbf{k}$. The same parameters can be found for different $\mathbf{k}$ using atomistic simulations by constraining interface orientation through periodic boundary conditions, which can be used for calibrating interface properties (e.g., like in Qin and Bhadeshia (2009)). For $\eta = \eta(x)$ and fixed $\mathbf{k}$, one has

$$\rho_0 \psi^\nabla = 0.5 \left(\beta(\mathbf{k}) \frac{d\eta}{dx} \right)^2; \quad \dot{\eta} = \bar{L}(\mathbf{k}) \left(-\rho_0 \frac{\partial f}{\partial \eta} + (\beta(\mathbf{k}))^2 \frac{\partial^2 \eta}{\partial x^2} \right). \quad (85)$$

10. Explicit expression for free energy and interface stresses

10.1. Stationary interface and arbitrary thermal energy

For the stationary interface, the Ginzburg-Landau equation can be transformed to $\frac{\partial f}{\partial \eta} = \frac{d\psi^\nabla}{d\eta}$ and its integral form:

$$\psi^\nabla = 0.5 \beta^2(\mathbf{k}) \left(\frac{d\eta}{dx} \right)^2 = \rho_0 (f(\theta, \eta) - f_0), \quad (86)$$

where f_0 is the integration constant (Levitas (2014b); Umantsev (2012)). Thus, for any stationary solution, the gradient energy is equal to the excess of the local energy at each point. Let us consider a stationary plane interface, when one has $\mathbf{H}$ as $x \rightarrow -\infty$ and $\mathbf{L}$ as $x \rightarrow \infty$. Then $\frac{d\eta}{dx} = 0$ for $x \rightarrow \pm\infty$, and $f(-\infty) = f(\mathbf{H}) = f_0 = 0$ (i.e., $f_0 = 0$) and $f(\infty) = f(\mathbf{L}) = f_0 = 0$. Since $f(\mathbf{H}) = f(\mathbf{L}) = f(1) = \Delta G^\theta = 0$, then $\mathbf{H}$ and $\mathbf{L}$ are in thermodynamic equilibrium, and $\theta = \theta_e$ (which is consistent with $\Delta G^\theta(\theta_e) = 0$). Thus, for a stationary interface Eq.(86) simplifies to

$$0.5 \beta^2(\mathbf{k}) \left(\frac{d\eta_{in}}{dx} \right)^2 = \rho_0 f(\theta_e, \eta). \quad (87)$$

Here subscript *in* designates that solution η_{in} is for an interface. According to Eq.(87), for any point x and given $\eta_{in}(x)$, the gradient $(d\eta_{in}/dx)^2$ is determined by the chosen direction

$\mathbf{k}$. To obtain biaxial interface stresses in Eq.(83) for the stationary interface one has to define for the general case (i.e. for an arbitrary distribution of η):

$$\check{\psi}^\theta := \psi^\nabla = \frac{\beta^2(\mathbf{k})}{2\rho_0} |\nabla \eta_{in}|^2 = f(\theta_e, \eta). \quad (88)$$

Then for an interface the last term in Eq.(83) disappears and surface stress represents biaxial tension within the interface. The magnitude of the biaxial interface tension in Eq.(83) is equal to $2\rho_0\psi^\nabla(\mathbf{k}) = 2\rho_0f(\theta_e, \eta) = \rho_0\psi^\nabla(\mathbf{k}) + \rho_0f(\theta_e, \eta)$, i.e., to the total equilibrium interface energy at each point. Integrating it over the x , we obtain that the magnitude of the force per unit interface length is equal to the equilibrium interface energy γ_e , as required.

10.2. Propagating interface

The solution to the Ginzburg-Landau Eq.(85) for the specific function f in Eq.(82) for the nonequilibrium plane interface is (Levitas et al. (2010); Levitas (2014a)):

$$\eta_{in} = (1 + e^{-\zeta})^{-0.5} \quad \text{with} \\ \zeta = k(\mathbf{k})(x - ct); \quad k(\mathbf{k}) = 2 \frac{\sqrt{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}}{\beta(\mathbf{k})}; \quad c = \frac{8\bar{L}(\mathbf{k})\rho_0\Delta G^\theta(\theta)}{k(\mathbf{k})}, \quad (89)$$

where k is inversely proportional to the interface width (see below) and c is the interface velocity, and we added $\mathbf{k}$ -dependence of parameters. The most important property of solution Eq.(89) for us is

$$\frac{d\eta_{in}}{d\zeta} = 0.5\eta_{in}(1 - \eta_{in}^2). \quad (90)$$

Using it and the definition of k in Eq.(89), we obtain a key relationship for points of a propagating interface:

$$\psi^\nabla = \frac{\beta^2(\mathbf{k})}{2\rho_0} \left(\frac{d\eta_{in}}{dx} \right)^2 = \frac{(\beta(\mathbf{k})k(\mathbf{k}))^2}{2\rho_0} \left(\frac{d\eta_{in}}{d\zeta} \right)^2 = 0.5(A(\theta) - 4\Delta G^\theta(\theta))\eta_{in}^2(1 - \eta_{in}^2)^2. \quad (91)$$

For phase equilibrium ($\Delta G^\theta(\theta) = 0$), this relationship reduces to Eq.(87). To obtain biaxial interface stress in Eq.(83) for the propagating interface one must define for the general case (i.e. for an arbitrary distribution of η rather than for η_{in}):

$$\check{\psi}^\theta := \psi^\nabla = \frac{\beta^2(\mathbf{k})}{2\rho_0} \left(\frac{d\eta_{in}}{dx} \right)^2 = 0.5(A(\theta) - 4\Delta G^\theta(\theta))\eta^2(1 - \eta^2)^2, \quad (92)$$

where Eq.(91) was used. It is clear that $\check{\psi}^\theta$ is localized at the diffuse interface, as required. Then,

$$\tilde{\psi}^\theta = f(\theta, \eta) - \check{\psi}^\theta = \Delta G^\theta(\theta)\eta^2(2 - \eta^2). \quad (93)$$

Substituting Eq.(92) in the general expression for the interface stress (83), we obtain for a propagating interface

$$\boldsymbol{\sigma}_{st} = \beta^2(\mathbf{k})|\nabla\eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) = 2\rho_0\check{\psi}^\theta (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) = \sigma_{st} (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}). \quad (94)$$

Still, it is necessary to check whether the magnitude of the force per unit interface length due to σ_{st} is equal to the interface energy γ .

11. Nonequilibrium interface energy and width

Gibbsian dividing surface and interface energy. We employ a suitable definition of the interface energy under nonequilibrium conditions (see, e.g., Gibbs (1948); Kaplan et al. (2013); Levitas and Samani (2011a); Levitas (2014a)), that represents the excess energy with respect to H in the H region $\varsigma \leq \varsigma_{ds}$ and with respect to L in the L region $\varsigma > \varsigma_{ds}$:

$$\gamma := \int_{-\infty}^{\varsigma_{ds}} \rho_0(\psi - \psi_H) d\varsigma + \int_{\varsigma_{ds}}^{\infty} \rho_0(\psi - \psi_L) d\varsigma, \quad (95)$$

where $\varsigma := x - ct = \zeta/k$ moves together with the interface with velocity c , ς_{ds} is the sharp interface or Gibbsian dividing surface position. The position of the Gibbsian dividing surface

for isotropic interface energy has been determined in Levitas (2014a,b) using the principle of static equivalence. Namely, stresses σ_{st} distributed through the finite interface width is equivalent to the concentrated load with the magnitude γ and zero moment applied to the sharp interface, i.e., the dividing surface:

$$T = \int_{-\infty}^{\infty} \sigma_{st}(\zeta) d\zeta = \gamma, \quad M = \int_{-\infty}^{\infty} \sigma_{st}(\zeta)(\zeta - \zeta_{ds}) d\zeta = 0. \quad (96)$$

While in general these are two independent conditions for a single position of the dividing surface ζ_{ds} , which can give different results, it is shown in Levitas (2014a,b) that both equations give the same result. Namely, at the dividing surface $\eta = \bar{\eta} = \frac{1}{\sqrt{1+e}} = 0.5186$ (where $e = 2.71828$ is the Euler's number), which is independent of material parameters. Exactly the same derivations can be repeated for anisotropic interface energy with the same result. Thus, we obtain

$$\begin{aligned} \gamma(\mathbf{k}, \theta) &= \Psi^l + \Psi^\nabla = 2\Psi^l = 2\Psi^\nabla = \frac{\beta(\mathbf{k}, \theta) \sqrt{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}}{4} = \frac{\beta(\mathbf{k}, \theta)}{Z} \\ &= \frac{k(\mathbf{k}, \theta) \beta^2(\mathbf{k}, \theta)}{8} = \frac{\rho_0(A - 4\Delta G^\theta)}{2k(\mathbf{k}, \theta)}, \quad Z = \frac{4}{\sqrt{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}}, \end{aligned} \quad (97)$$

where

$$\Psi^l = \int_{-\infty}^{\zeta_{ds}} \rho_0(\psi^l - \psi_A) d\zeta + \int_{\zeta_{ds}}^{\infty} \rho_0(\psi^l - \psi_M) d\zeta, \quad \Psi^\nabla := \int_{-\infty}^{\infty} \rho_0 \psi^\nabla dx \quad (98)$$

are the contribution to the total interface energy due to local (ψ^l) and gradient parts, which appeared to be equal for the nonequilibrium interface. Thus, orientational dependence of the energy of the nonequilibrium interface is proportional to the orientational dependence of the β . Previously (Qin and Bhadeshia (2009); Taylor and Cahn (1998); Wheeler and McFadden (1997)), this result was obtained for the equilibrium interface only.

Interface width. The nonequilibrium interface width is defined as

$$\delta(\mathbf{k}, \theta) := \frac{10}{k(\mathbf{k}, \theta)} = \frac{5\beta(\mathbf{k}, \theta)}{\sqrt{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}} = \frac{5}{4} \frac{\beta^2(\mathbf{k}, \theta)}{\gamma(\mathbf{k}, \theta)} \quad (99)$$

and its orientational dependence is also proportional to the orientational dependence of the β . The factor of 10 is an approximate width of the interface $\eta_{in}(\zeta)$ in terms of ζ in Eq.(89). Alternative definitions of the interface width results in a factor different from 10 (Anderson et al. (2001); Levitas et al. (2003, 2010); Levitas and Samani (2011a); Svoboda et al. (2012)).

Toward experimental definition of material functions. The ratio

$$\frac{\gamma(\mathbf{k}, \theta)}{\delta(\mathbf{k}, \theta)} = \frac{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}{20}, \quad (100)$$

is independent of the interface orientation. For an equilibrium interface at $\theta = \theta_e$ ($\Delta G^\theta(\theta_e) = 0$) one has

$$\gamma(\mathbf{k}, \theta_e) = \frac{\beta(\mathbf{k}, \theta_e)\sqrt{\rho_0 A(\theta_e)}}{4}; \quad \delta(\mathbf{k}, \theta_e) = \frac{5\beta(\mathbf{k}, \theta_e)}{\sqrt{\rho_0 A(\theta_e)}}, \quad \frac{\gamma(\mathbf{k}, \theta_e)}{\delta(\mathbf{k}, \theta_e)} = \frac{\rho_0 A(\theta_e)}{20}. \quad (101)$$

We assume that function $\Delta G^\theta(\theta)$ is known from thermodynamic functions of phases; orientational dependence of the interface energy $\gamma(\mathbf{k}, \theta)$ is known from the molecular dynamic simulations (Qin and Bhadeshia (2009)) (at least for $\theta = \theta_e$). If one determines from the molecular dynamics simulations for $\{1, 0, 0\}$ orientation $\beta_0(\theta) := \beta((1, 0, 0), \theta)$, $\gamma_0(\theta) := \gamma((1, 0, 0), \theta)$, and $\delta_0(\theta) = \delta((1, 0, 0), \theta)$, then

$$\frac{\gamma(\mathbf{k}, \theta)}{\delta(\mathbf{k}, \theta)} = \frac{\rho_0(A(\theta) - 4\Delta G^\theta(\theta))}{20} = \frac{\gamma_0(\theta)}{\delta_0(\theta)}, \quad (102)$$

which determines function $A(\theta)$. Then Eqs.(97) and (99) can be presented in the form

$$\gamma(\mathbf{k}, \theta) = \beta(\mathbf{k}, \theta)\sqrt{\frac{5\gamma_0(\theta)}{4\delta_0(\theta)}}; \quad \delta(\mathbf{k}, \theta) = \beta(\mathbf{k}, \theta)\sqrt{\frac{5\delta_0(\theta)}{4\gamma_0(\theta)}}. \quad (103)$$

Thus, due to direct proportionality between $\gamma(\mathbf{k}, \theta)$ and $\beta(\mathbf{k}, \theta)$, all coefficients α_i in $\beta(\mathbf{k}, \theta)$ in Eq.(64) are determined from the corresponding coefficients for $\gamma(\mathbf{k}, \theta)$ from Qin and Bhadeshia (2009) by dividing by $\sqrt{(5\gamma_0(\theta))/(4\delta_0(\theta))}$. This differs from the relationships in Qin and Bhadeshia (2009) even at the phase equilibrium temperature, because in Qin

and Bhadeshia (2009) it was overlooked that the interface width is also orientation dependent. According to Eq.(103)

$$\gamma(\mathbf{k}, \theta) \delta(\mathbf{k}, \theta) = \frac{5}{4} \beta^2(\mathbf{k}, \theta), \quad (104)$$

i.e., the temperature-dependence of the product $\gamma(\mathbf{k}, \theta) \delta(\mathbf{k}, \theta)$ is fully determined by the temperature-dependence of function β .

12. Energy, width, entropy excess, and stresses for a nonequilibrium interface: specification

We will consider the same specification as in Levitas (2014b) with the only difference being that $\beta(\theta)$ from prior work is replaced here with $\beta^2(\mathbf{k}, \theta)$, which also results in k to be substituted with $k(\mathbf{k}, \theta)$. That is why we skip most of the intermediate steps and present the final equations only.

Energy and entropy excess. It is traditionally accepted (Levitas and Preston (2002a); Levitas et al. (2003)) that

$$A = A_0 (\theta - \theta_c), A_0 > 0; \quad \Delta G^\theta(\theta) = -\Delta s_0 (\theta - \theta_e), \quad \Delta s_0 < 0, \quad (105)$$

where Δs_0 is the entropy jump between L and H phases at the phase equilibrium temperature θ_e , and θ_c is the critical temperature for the loss of the thermodynamic stability of the H at zero stresses. We introduce the dimensionless temperature, $\bar{\theta}$, and other parameters by the following equations:

$$\bar{\theta} := \frac{\theta - \theta_e}{\theta_e - \theta_c}; \quad \varpi := -\frac{4\Delta s_0}{A_0} > \frac{2}{3}; \quad \Upsilon := \bar{\theta}(1 - \varpi) + 1 \geq 0; \quad \tilde{A} := \rho_0 A_0 (\theta_e - \theta_c). \quad (106)$$

The expression (97) for the interface energy takes the form

$$\begin{aligned} \gamma &= \frac{\tilde{A} \Upsilon}{2k(\mathbf{k}, \theta)} = \frac{\beta(\mathbf{k}, \theta) \sqrt{\rho_0 A_0 [(\theta - \theta_e)(1 - \varpi) + (\theta_e - \theta_c)]}}{4} = \\ &= \frac{\beta(\mathbf{k}, \theta) \sqrt{\rho_0 A_0 (\theta_e - \theta_c)}}{4} \sqrt{\bar{\theta}(1 - \varpi) + 1} = \frac{\beta(\mathbf{k}, \theta) \sqrt{\tilde{A}}}{4} \sqrt{\Upsilon}. \end{aligned} \quad (107)$$

At $\theta = \theta_e$, we obtain $\Upsilon = 1$ and

$$\gamma_e = \frac{\beta(\mathbf{k}, \theta_e) \sqrt{\tilde{A}}}{4}; \quad \gamma_0(\theta_e) = \frac{\beta_0(\theta_e) \sqrt{\tilde{A}}}{4}. \quad (108)$$

The excess entropy at an interface is

$$s_i = -\frac{\partial \gamma}{\partial \theta} = \frac{\beta(\mathbf{k}, \theta) \sqrt{\rho_0 A_0}}{8\sqrt{\theta_e - \theta_c}} \frac{\varpi - 1}{\sqrt{\tilde{A}}} + \frac{\partial \beta(\mathbf{k}, \theta)}{\partial \theta} \frac{\sqrt{\rho_0 A_0 (\theta_e - \theta_c)}}{4} \sqrt{\Upsilon}. \quad (109)$$

Interface width. The parameter k can be determined either based on the definition Eq.(89) or on the relation to the interface energy Eq.(97):

$$\begin{aligned} k(\mathbf{k}, \theta) &= \frac{8\gamma(\mathbf{k}, \theta)}{\beta^2(\mathbf{k}, \theta)} = 2 \frac{\sqrt{\rho_0 A_0 ((\theta - \theta_e)(1 - \varpi) + (\theta_e - \theta_c))}}{\beta(\mathbf{k}, \theta)} \\ &= 2 \frac{\sqrt{\tilde{A} [\bar{\theta}(1 - \varpi) + 1]}}{\beta(\mathbf{k}, \theta)} = 2 \frac{\sqrt{\tilde{A} \Upsilon}}{\beta(\mathbf{k}, \theta)}. \end{aligned} \quad (110)$$

Then the diffuse interface width at arbitrary and equilibrium temperatures is defined as

$$\delta(\mathbf{k}, \theta) := \frac{10}{k(\mathbf{k}, \theta)} = 5 \frac{\beta(\mathbf{k}, \theta)}{\sqrt{\tilde{A} [\bar{\theta}(1 - \varpi) + 1]}} = \frac{5}{4} \frac{\beta^2(\mathbf{k}, \theta)}{\gamma(\mathbf{k}, \theta)}; \quad \delta(\mathbf{k}, \theta_e) = 5 \frac{\beta(\mathbf{k}, \theta_e)}{\sqrt{\tilde{A}}}. \quad (111)$$

The factor of 10 is an approximate width of the diffuse interface $\eta_{in}(\zeta)$ in terms of ζ in Eq.(89). We define the dimensionless interface width $\tilde{\delta}$ normalized by the width at the equilibrium temperature θ_e and the $\langle 100 \rangle$ direction:

$$\tilde{\delta} := \frac{\delta(\mathbf{k}, \theta)}{\delta_0(\theta_e)} = \frac{\beta(\mathbf{k}, \theta)}{\beta_0(\theta_e) \sqrt{[\bar{\theta}(1 - \varpi) + 1]}} = \frac{\beta(\mathbf{k}, \theta)}{\beta_0(\theta_e) \sqrt{\Upsilon}}. \quad (112)$$

Interface stress. According to Eqs.(92) and (94), the magnitude of the biaxial interface stresses is defined as:

$$\sigma_{st} = 2\rho_0 \check{\psi}^\theta = \rho_0 A_0 [(\theta - \theta_e)(1 - \varpi) + (\theta_e - \theta_c)] \eta_{in}^2 (1 - \eta_{in}^2)^2 = \tilde{A} \Upsilon \eta_{in}^2 (1 - \eta_{in}^2)^2. \quad (113)$$

Since the interface profile is time-independent, we can chose time and perform simple algebraic manipulations as in Levitas (2014b):

$$e^{-\zeta} = e^{-k(\mathbf{k},\theta)x + \ln 3} = 3e^{-10x/\delta((\mathbf{k},\theta))} = 3e^{-\frac{10x}{\delta_0(\theta_e)} \frac{\delta_0(\theta_e)}{\delta((\mathbf{k},\theta))}} = 3e^{-10y/\tilde{\delta}(\mathbf{k},\theta)}; \quad y := \frac{x}{\delta_0(\theta_e)}, \quad (114)$$

where with the dimensionless coordinate y the width of interface $\simeq 1$ in the $< 100 >$ direction at $\theta = \theta_e$ and $\eta_{in}(y = 0) = 0.5$. Then, utilizing Eqs.(89) and (113), we obtain the interface stress distribution and its dimensionless counterpart $\bar{\sigma}_{st}$:

$$\sigma_{st} = \tilde{A}\Upsilon \frac{9e^{-20y/\tilde{\delta}(\mathbf{k},\theta)}}{\left(1 + 3e^{-10y/\tilde{\delta}(\mathbf{k},\theta)}\right)^3}; \quad \bar{\sigma}_{st} := \frac{\sigma_{st}}{\tilde{A}\Upsilon} = \frac{9e^{-20y/\tilde{\delta}(\mathbf{k},\theta)}}{\left(1 + 3e^{-10y/\tilde{\delta}(\mathbf{k},\theta)}\right)^3}. \quad (115)$$

Remarkably, the maximum dimensionless interface stress is independent of $\mathbf{k}$ and equal to $\frac{4}{27}$ at $y = \ln(\frac{3}{2})\frac{\tilde{\delta}(\mathbf{k},\theta)}{10}$, which corresponds to $\tilde{\eta} = 1/\sqrt{3} = 0.577$. A plot of $\bar{\sigma}_{st}(y)$ for $\theta = \theta_e$ and several crystallographic orientations is presented in Fig. 7 for bcc Li and Fe. Both the shift of the maximum and the area below the curve (which is an interface energy) are proportional to $\tilde{\delta}$ and β . For $y = 0$ the stress is also independent of $\tilde{\delta}$ and equal to 0.141. **Since sixth-degree potential at $\theta = \theta_e$ is asymmetric with respect to $\eta = 0.5$, the interface profile η_{in} and interface stresses are asymmetric with respect to $\eta = 0.5$ (i.e., $y = 0$) as well, in contrast to the fourth-degree potential used in Levitas (2013a,b).**

13. Interface stresses for a critical nucleus

All results for a critical L nucleus in Levitas (2014b) can be repeated here with adding the $\mathbf{k}$ -dependence of β and, consequently, the width of the critical nucleus l . Thus, profile of the critical nucleus is described by equation

$$\eta_c = 2 \left[4 - P + \sqrt{P^2 - 8P/3} \cosh \left(20\sqrt{\bar{\theta} + 1} x/l(\mathbf{k},\theta) \right) \right]^{-1/2};$$

$$P := 3\varpi\bar{\theta}/(\bar{\theta} + 1); \quad l(\mathbf{k},\theta) := 10\beta(\mathbf{k},\theta)/\sqrt{\tilde{A}} \quad (116)$$

Figure 7: (a) Distribution of the dimensionless biaxial interface stress $\bar{\sigma}_{st}(y)$ for $\theta = \theta_e$ and several crystallographic directions for bcc Li and Fe. The area below the plots is proportional to the interface energy $\gamma(\mathbf{k}, \theta_e)$, which has the same orientation dependence as $\tilde{\delta}$ and β .

or in terms of dimensionless coordinate

$$\eta_c = 2 \left[4 - P + \sqrt{P^2 - 8P/3} \cosh \left(20\sqrt{\bar{\theta} + 1} y / \tilde{l}(\mathbf{k}, \theta) \right) \right]^{-1/2};$$

$$y := x/l((1, 0, 0), \theta); \quad \tilde{l}(\mathbf{k}, \theta) := l(\mathbf{k}, \theta)/l((1, 0, 0), \theta). \quad (117)$$

and shown in Fig. 8 for several orientations of Li crystal for $\varpi = 1$, and two temperatures. This solution exists for $\theta \leq \theta_e$, and satisfies the boundary conditions $\eta_c(\pm\infty) = 0$. Since $\frac{d\eta_c}{dx}(\pm\infty) = 0$ and $f(\theta, 0) = 0$, Eq.(86) results in $f_0 = 0$ and

$$\psi^\nabla(\mathbf{k}, \theta, \eta_c) = f(\theta, \eta_c), \quad \psi(\mathbf{k}, \theta, \eta_c) = \psi^\nabla(\mathbf{k}, \theta, \eta_c) + f(\theta, \eta_c) = 2\psi^\nabla(\mathbf{k}, \theta, \eta_c) = 2f(\theta, \eta_c) \quad (118)$$

Equation (83) for the interface stress tensor allowing for Eq.(118) leads to

$$\boldsymbol{\sigma}_{st} = \rho_0 \psi(\theta, \eta_c) (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) - \rho_0 \tilde{\psi}^\theta(\theta, \eta_c) \mathbf{I}. \quad (119)$$

It is evident that in addition to the tensile biaxial interface stress the tensile mean stress (negative pressure) $-\rho_0 \tilde{\psi}^\theta > 0$ acts at each point of a critical nucleus. Since the magnitude

of the biaxial tension is equal at each point to the local total free energy per unit volume $\rho_0\psi(\theta, \eta_c)$, then the total force is also equal to the total energy of a critical nucleus. The dimensionless magnitude of the biaxial tension and the tensile mean stress

$$\bar{\sigma}_{st} := \sigma_{st}/\tilde{A} = 0.5\varpi\bar{\theta}\eta_c^4(3 - 2\eta_c^2) + (\bar{\theta} + 1)\eta_c^2(1 - \eta_c^2)^2 > 0; \quad (120)$$

$$p := -4\rho_0\tilde{\psi}^\theta/\tilde{A} = -\varpi\bar{\theta}\eta_c^2(2 - \eta_c^2) > 0 \quad (121)$$

is plotted in Fig. 8 for the same conditions as for η_c . The $\mathbf{k}$ -dependence of stresses comes solely from the solution η_c through the width l of the nucleus.

Figure 8: Profile of the critical martensitic nucleus $\eta_c(y)$, and distribution of the dimensionless biaxial surface stress $\bar{\sigma}_{st}$ and mean stress p for several orientations of Li crystal, $\varpi = 1$, and two temperatures, $\bar{\theta} = -0.01$ (top) and $\bar{\theta} = -0.3$ (bottom).

With increasing overcooling, the magnitude of η_c and biaxial tension (and consequently, nucleus energy) reduces, while mean stress increases and essentially becomes larger than

tension. This is because the nucleus profile more significantly deviates from the complete nucleus (with maximum $\eta_c = 1$) with two equilibrium interfaces.

14. Complete system of equations

Below we present the complete system of equations for a single order parameter and isothermal processes, similar to that in Levitas (2014b) but for anisotropic gradient energy. **We also included small strain approximation, in which we keep linear in strain ϵ terms, which components are small in comparison with unity, and infinitesimal strain approximation, when we neglect these terms as well in comparison with unity.**

1. Kinematics

1.1. Decomposition of the deformation gradient $\mathbf{F}$

$$\mathbf{F} = \nabla_0 \mathbf{r} = \mathbf{F}_e \cdot \mathbf{U}_\theta \cdot \mathbf{U}_t. \quad (122)$$

1.2. Jacobians

$$J := \frac{\rho_0}{\rho} = \det \mathbf{F}; \quad J_t := \frac{\rho_0}{\rho_t} = \det \mathbf{U}_t; \quad J_\theta := \frac{\rho_t}{\rho_\theta} = \det \mathbf{U}_\theta; \quad J_e := \frac{\rho_\theta}{\rho_e} = \det \mathbf{F}_e; \quad (123)$$

$$J = J_t J_\theta J_e; \quad J_{t\theta} := \frac{\rho_0}{\rho_\theta} = \det(\mathbf{U}_\theta \cdot \mathbf{U}_t) = J_t J_\theta. \quad (124)$$

1.3. Transformation $\mathbf{U}_t$ and thermal $\mathbf{U}_\theta$ deformation gradients

$$\begin{aligned} \mathbf{U}_t(\eta) &= \mathbf{I} + \boldsymbol{\epsilon}_t = \mathbf{I} + \boldsymbol{\epsilon}_{tL} \varphi(a_t, \eta); \\ \mathbf{U}_\theta(\theta, \eta) &= \mathbf{I} + \boldsymbol{\epsilon}_\theta = \mathbf{I} + \boldsymbol{\epsilon}_{\theta H} + (\boldsymbol{\epsilon}_{\theta L} - \boldsymbol{\epsilon}_{\theta H}) \varphi(a_\theta, \eta); \\ \varphi_6(a, \eta) &:= a\eta^2/2 + (3-a)\eta^4 + (a-4)\eta^6/2; \quad 0 < a < 6; \\ \varphi_4(a, \eta) &:= a\eta^2(1-\eta)^2 + (4\eta^3 - 3\eta^4); \quad 0 < a < 6. \end{aligned} \quad (125)$$

1.4. Small strains

$$\boldsymbol{\varepsilon} = (\nabla_0 \mathbf{u})_s; \quad \boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_e + \boldsymbol{\varepsilon}_t(\eta) + \boldsymbol{\varepsilon}_\theta(\theta, \eta); \quad J = \frac{\rho_0}{\rho} = 1 + \varepsilon_0; \quad \varepsilon_0 = \boldsymbol{\varepsilon} : \mathbf{I} = \varepsilon_{0e} + \varepsilon_{0t} + \varepsilon_{0\theta} \quad (126)$$

2. Gradient energy

2.1. Finite strains

$$\begin{aligned} \rho_0 \psi^\nabla &= 0.5 \beta^2(\mathbf{k}_0) |\nabla \eta|^2 = 0.5 (\beta(\mathbf{k}_0) |\nabla \eta|)^2 = 0.5 \left(\beta(\nabla_0 \eta) \frac{|\nabla \eta|}{|\nabla_0 \eta|} \right)^2 \\ &= 0.5 \left(\beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}|}{|\boldsymbol{\zeta}_0|} \right)^2 = 0.5 \left(\beta(\boldsymbol{\zeta}_0) \frac{|\boldsymbol{\zeta}_0 \cdot \mathbf{F}^{-1}|}{|\boldsymbol{\zeta}_0|} \right)^2 = 0.5 \beta^2(\boldsymbol{\zeta}_0) \frac{(\boldsymbol{\zeta}_0 \cdot \mathbf{B} \cdot \boldsymbol{\zeta}_0)}{(\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)}; \\ \boldsymbol{\zeta}_0 &:= \nabla_0 \eta; \quad \boldsymbol{\zeta} := \nabla \eta; \quad \mathbf{B} := \mathbf{F}^{-1} \cdot \mathbf{F}^{-1T} = (\mathbf{F}^T \cdot \mathbf{F})^{-1}. \end{aligned} \quad (127)$$

2.2. Small strains

$$\rho_0 \psi^\nabla = 0.5 (\beta(\mathbf{k}_0) |\nabla \eta|)^2 = 0.5 \left(\beta(\nabla_0 \eta) \frac{|\nabla \eta|}{|\nabla_0 \eta|} \right)^2 = 0.5 \beta^2(\boldsymbol{\zeta}_0) \frac{(\boldsymbol{\zeta}_0 \cdot (\mathbf{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_0)}{(\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0)} \quad (128)$$

2.3. Infinitesimal strains

$$\rho \psi^\nabla = 0.5 \beta^2(\mathbf{k}) |\nabla \eta|^2 = 0.5 (\beta(\mathbf{k}) |\nabla \eta|)^2 = 0.5 (\beta(\nabla \eta))^2. \quad (129)$$

3. Helmholtz free energy per unit mass and its contributions

$$\bar{\psi}(\mathbf{F}, \eta, \theta, \nabla \eta, \nabla_0 \eta) = \frac{J_{t\theta}}{\rho_0} \psi^e(\mathbf{F}_e, \eta, \theta) + J \check{\psi}^\theta(\eta, \theta) + \tilde{\psi}^\theta(\eta, \theta) + J \psi^\nabla(|\nabla \eta|, \nabla_0 \eta); \quad (130)$$

$$\begin{aligned} \check{\psi}_4^\theta &= (A(\theta) - 3\Delta G^\theta(\theta)) \eta^2 (1 - \eta)^2; \quad \tilde{\psi}_4^\theta = \Delta G^\theta(\theta) \eta^2 (3 - 2\eta); \\ \check{\psi}_6^\theta &= 0.5 (A(\theta) - 4\Delta G^\theta(\theta)) \eta^2 (1 - \eta^2)^2; \quad \tilde{\psi}_6^\theta = \Delta G^\theta(\theta) \eta^2 (2 - \eta^2); \\ \psi^e &= \frac{1}{2} \mathbf{E}_e : \mathbf{C}(\eta) : \mathbf{E}_e; \quad \mathbf{C}(\eta) = \mathbf{C}_H + (\mathbf{C}_L - \mathbf{C}_H) \varphi(a_C, \eta). \end{aligned} \quad (131)$$

4. First Piola–Kirchhoff and Cauchy stress tensors

4.1. Finite strains

$$\mathbf{P} = \mathbf{P}_e + \mathbf{P}_{st} + \mathbf{P}_d; \quad \boldsymbol{\sigma} = \boldsymbol{\sigma}_e + \boldsymbol{\sigma}_{st} + \boldsymbol{\sigma}_d; \quad (132)$$

$$\mathbf{P}_e = J_{t\theta} \mathbf{F}_e \cdot \frac{\partial \psi^e(\mathbf{E}_e, \eta)}{\partial \mathbf{E}_e} \cdot \mathbf{F}_e^T \cdot \mathbf{F}^{T-1} = J_{t\theta} \mathbf{F}_e \cdot (\mathbf{C} : \mathbf{E}_e) \cdot \mathbf{F}_e^T \cdot \mathbf{F}^{T-1}; \quad (133)$$

$$\boldsymbol{\sigma}_e = J_e^{-1} \mathbf{F}_e \cdot \frac{\partial \psi^e}{\partial \mathbf{E}_e} \cdot \mathbf{F}_e^T = J_e^{-1} \mathbf{F}_e \cdot (\mathbf{C} : \mathbf{E}_e) \cdot \mathbf{F}_e^T, \quad (134)$$

$$\begin{aligned} \boldsymbol{\sigma}_{st} &= \rho_0(\check{\psi}^\theta + \psi^\nabla) \mathbf{I} - \rho_0 \nabla \eta \otimes \frac{\partial \psi^\nabla}{\partial \nabla \eta} = (\rho_0 \check{\psi}^\theta + 0.5 \beta^2(\mathbf{k}_0) |\nabla \eta|^2) \mathbf{I} - \beta^2(\mathbf{k}_0) \nabla \eta \otimes \nabla \eta \\ &= \beta^2(\mathbf{k}_0) |\nabla \eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho_0 \check{\psi}^\theta - 0.5 \beta^2(\mathbf{k}_0) |\nabla \eta|^2) \mathbf{I}. \end{aligned} \quad (135)$$

$$\begin{aligned} \mathbf{P}_{st} &= \rho_0 J \left((\check{\psi}^\theta + \psi^\nabla) \mathbf{I} - \nabla \eta \otimes \frac{\partial \psi^\nabla}{\partial \nabla \eta} \right) \cdot \mathbf{F}^{T-1} = \\ &\rho_0 J \left[\beta^2(\mathbf{k}_0) |\nabla \eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho_0 \check{\psi}^\theta - 0.5 \beta^2(\mathbf{k}_0) |\nabla \eta|^2) \mathbf{I} \right] \cdot \mathbf{F}^{T-1}; \\ \boldsymbol{\sigma}_v &= \boldsymbol{\sigma}_v(\mathbf{d}); \quad \mathbf{P}_v = J \boldsymbol{\sigma}_v(\mathbf{d}) \cdot \mathbf{F}^{T-1}. \end{aligned} \quad (136)$$

4.2. Infinitesimal strains

$$\begin{aligned} \mathbf{P} &= \boldsymbol{\sigma} = \boldsymbol{\sigma}_e + \boldsymbol{\sigma}_{st} + \boldsymbol{\sigma}_d; \quad \boldsymbol{\sigma}_e = \frac{\partial \psi^e}{\partial \boldsymbol{\varepsilon}_e} = \mathbf{C} : \boldsymbol{\varepsilon}_e; \\ \boldsymbol{\sigma}_{st} &= (\rho \check{\psi}^\theta + 0.5 \beta^2(\mathbf{k}) |\nabla \eta|^2) \mathbf{I} - \beta^2(\mathbf{k}) \nabla \eta \otimes \nabla \eta \\ &= \beta^2(\mathbf{k}) |\nabla \eta|^2 (\mathbf{I} - \mathbf{k} \otimes \mathbf{k}) + (\rho \check{\psi}^\theta - 0.5 \beta^2(\mathbf{k}) |\nabla \eta|^2) \mathbf{I}. \end{aligned} \quad (137)$$

5. Ginzburg–Landau equations

5.1. Compact form

$$\dot{\eta}(\mathbf{r}_0, t) = L \left(-\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho_0} \nabla_0 \cdot \left(\rho_0 \frac{\partial \bar{\psi}}{\partial \nabla_0 \eta} \right) \right). \quad (138)$$

5.2. Detailed form for large strains

$$\begin{aligned} \frac{\rho_0 \dot{\eta}}{L} &= -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \frac{\boldsymbol{\zeta}_0 \cdot (\mathbf{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \\ &2\beta \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \cdot \left(-\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} + 4 \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \frac{4 \boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\boldsymbol{\zeta}_0|^4} \right) \end{aligned}$$

$$\begin{aligned}
& -2\beta^2 \left(\frac{\zeta_0 \cdot \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} : \mathbf{I} + \boldsymbol{\varepsilon} : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^2} - \frac{4(\boldsymbol{\varepsilon} \cdot \zeta_0) \cdot \left(\zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \right)}{|\zeta_0|^4} \right) \\
& + 2\beta^2 \left(\frac{\zeta_0 \otimes \zeta_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} \cdot \zeta_0 + \nabla_0^2 \eta \zeta_0 \cdot \boldsymbol{\varepsilon} \cdot \zeta_0}{|\zeta_0|^4} - 4 \frac{\zeta_0 \cdot \boldsymbol{\varepsilon} \cdot \zeta_0 \zeta_0 \otimes \zeta_0 : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^6} \right). \quad (139)
\end{aligned}$$

$$\begin{aligned}
& -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} = \mathbf{P}_e \cdot \mathbf{U}_t : \frac{\partial \mathbf{U}_\theta(\theta, \eta)}{\partial \eta} \cdot \mathbf{F}_e^T + \mathbf{P}_e : \frac{\partial \mathbf{U}_t(\eta)}{\partial \eta} \cdot \mathbf{U}_\theta \cdot \mathbf{F}_e^T - J_{t\theta} \frac{\partial \psi^e}{\partial \eta} \Big|_{\mathbf{E}_e} \\
& - J_{t\theta} \psi^e \left(\mathbf{U}_t^{-1} : \frac{\partial \mathbf{U}_t}{\partial \eta} + \mathbf{U}_\theta^{-1} : \frac{\partial \mathbf{U}_\theta}{\partial \eta} \right) - J \frac{\partial \tilde{\psi}^\theta}{\partial \eta} - \frac{\partial \tilde{\psi}^\theta}{\partial \eta}. \quad (140)
\end{aligned}$$

5.3. Small strains

$$\begin{aligned}
& \frac{\rho_0 \dot{\eta}}{L} = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_0} \otimes \frac{\partial \beta}{\partial \zeta_0} + \beta \frac{\partial^2 \beta}{\partial \zeta_0 \partial \zeta_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} + \\
& 2\beta \frac{\partial \beta}{\partial \zeta_0} \cdot \left(-\frac{\zeta_0 \otimes \zeta_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} + 4 \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \cdot \boldsymbol{\varepsilon} \cdot \zeta_0}{|\zeta_0|^2} + \frac{4 \zeta_0 \cdot \boldsymbol{\varepsilon} \cdot \zeta_0 \zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^4} \right) \\
& - 2\beta^2 \left(\frac{\zeta_0 \cdot \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} : \mathbf{I} + \boldsymbol{\varepsilon} : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^2} - \frac{4(\boldsymbol{\varepsilon} \cdot \zeta_0) \cdot \left(\zeta_0 \cdot \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} \right)}{|\zeta_0|^4} \right) \\
& + 2\beta^2 \left(\frac{\zeta_0 \otimes \zeta_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \mathbf{r}_0} \cdot \zeta_0 + \nabla_0^2 \eta \zeta_0 \cdot \boldsymbol{\varepsilon} \cdot \zeta_0}{|\zeta_0|^4} - 4 \frac{\zeta_0 \cdot \boldsymbol{\varepsilon} \cdot \zeta_0 \zeta_0 \otimes \zeta_0 : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0}}{|\zeta_0|^6} \right). \quad (141)
\end{aligned}$$

$$-\rho \frac{\partial \bar{\psi}}{\partial \eta} = \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_\theta(\theta, \eta_k)}{\partial \eta_i} + \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_t(\eta_k)}{\partial \eta_i} - \frac{\partial \psi^e}{\partial \eta_i} \Big|_{\boldsymbol{\varepsilon}_e} - \psi^e \left(\frac{\partial \boldsymbol{\varepsilon}_{t0}}{\partial \eta_i} + \frac{\partial \boldsymbol{\varepsilon}_{\theta 0}}{\partial \eta_i} \right) - \rho \frac{\partial \tilde{\psi}^\theta}{\partial \eta_i} - \rho \frac{\partial \tilde{\psi}^\theta}{\partial \eta_i}. \quad (142)$$

5.4. Finite volumetric strain and infinitesimal change in shape

$$\frac{\rho_0 \dot{\eta}}{L} = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \zeta_0} \otimes \frac{\partial \beta}{\partial \zeta_0} + \beta \frac{\partial^2 \beta}{\partial \zeta_0 \partial \zeta_0} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r}_0 \partial \mathbf{r}_0} a^{-2} + \beta \frac{\partial \beta}{\partial \zeta_0} \cdot \frac{\partial a^{-2}}{\partial \mathbf{r}_0}. \quad (143)$$

5.5. Infinitesimal strains

$$\frac{\rho \dot{\eta}}{L} = -\rho \frac{\partial \bar{\psi}}{\partial \eta} + \left(\frac{\partial \beta}{\partial \boldsymbol{\zeta}} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta} \partial \boldsymbol{\zeta}} \right) : \frac{\partial^2 \eta}{\partial \mathbf{r} \partial \mathbf{r}}. \quad (144)$$

Examples of the homogeneous degree on function $\beta(\boldsymbol{\zeta}_0)$ for the cubic symmetry and its derivative that participate in Eq.(139) are presented in Eqs.(65)-(154).

6. Momentum balance equation

6.1 Finite strains

$$\nabla_0 \cdot \mathbf{P} + \rho_0 \mathbf{f} = \rho_0 \dot{\mathbf{v}}. \quad (145)$$

6.2. Small strains

$$\nabla \cdot \boldsymbol{\sigma} + \rho \mathbf{f} = \rho \dot{\mathbf{v}}. \quad (146)$$

7. Boundary conditions for the order parameter

7.1 Finite strains

$$\mathbf{n}_0 \cdot \rho_0 \left[\beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \frac{\boldsymbol{\zeta}_0 \cdot (\mathbf{I} - 2\boldsymbol{\mathcal{E}}) \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} - 2(\beta(\boldsymbol{\zeta}_0))^2 \left(\frac{\boldsymbol{\mathcal{E}} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} - \frac{\boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \cdot \boldsymbol{\mathcal{E}} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^4} \right) \right] = H_0. \quad (147)$$

$$H_0 = -\frac{\partial \gamma_s(\eta)}{\partial \eta}; \quad \gamma_s(\eta) = \gamma_s^A + (\gamma_s^M - \gamma_s^A) \varphi(a_s, \eta). \quad (148)$$

7.2. Infinitesimal strains

$$\mathbf{n} \cdot \rho \beta(\boldsymbol{\zeta}) \frac{\partial \beta(\boldsymbol{\zeta})}{\partial \boldsymbol{\zeta}} = H. \quad (149)$$

Subscripts 4 and 6 designate the $2 - 3 - 4$ and $2 - 4 - 6$ polynomials, see Eqs. (125) and (131). Monotonous functions $\varphi(a, \eta)$ with different parameters a smoothly connect any material properties of A and M and possess the following properties:

$$\varphi(a, 0) = 0; \quad \varphi(a, 1) = 1; \quad \frac{\partial \varphi(a, 0)}{\partial \eta} = \frac{\partial \varphi(a, 1)}{\partial \eta} = 0. \quad (150)$$

The last condition is required in order to have $\eta = 0$ and $\eta = 1$ as solutions of thermodynamic equilibrium conditions $X = 0$ for any stresses tensor and temperature. For further generality, the elastic energy (131) and elasticity rule (134) can be substituted with those for a nonlinear elastic rule (Levitas (2013a)).

15. Concluding remarks and future directions

In this paper a general thermodynamically consistent, large strain phase field approach for multiple order parameters developed in Levitas (2014b) is generalized for an anisotropic interface energy. While it seems to be a trivial task, by simply using anisotropic gradient energy $\psi^\nabla(\nabla\eta_i)$ as in Anderson et al. (2000); Braun et al. (1997); Cahn and Hoffman (1974); Hoffman-Cahn (1972); Kobayashi (1993); Taylor and Cahn (1998, 1994); Warren-Boettinger (1995); Wheeler and McFadden (1997), it appeared to be a very nontrivial basic problem. Indeed, strict continuum mechanical treatment was lacking in the above publications, and the fact that the Cauchy stress is non-symmetric in these theories was not viewed as problematic. We demonstrated that the non-symmetric Cauchy stress violates the moment of the momentum balance and the principle of material objectivity. This is because the Herring torque (orientational derivative of the interface energy) is a configurational thermodynamic force, which does not contribute to the moment of momentum balance. It is found in the paper that to eliminate these problems the gradient energy should have a form $\psi^\nabla(|\nabla\eta|, \mathbf{k}_0)$ for a single and $\psi^\nabla(\nabla\eta_i \cdot \nabla\eta_j, \mathbf{k}_{0i})$ for multiple order parameters. Thus, anisotropy (i.e., dependence on the unit normal to the interface $\mathbf{k}_0 = \nabla\eta_i/|\nabla\eta_i|$) should be described with respect to the reference configuration (for which the crystal lattice symmetry group is known) but the magnitude of the gradient of the order parameter $|\nabla\eta|$ or invariants $\nabla\eta_i \cdot \nabla\eta_j$ are evaluated for the gradient in the deformed configuration (otherwise, interface stresses will not appear at all). For such a theory, since $\mathbf{k}_0$ is independent of the deformation gradient $\mathbf{F}$, its inclusion does not change the general form of the expression for the Cauchy stresses (which remain symmetric) and only slightly changes specific expression for them. However, the Ginzburg-Landau equation gains many additional terms in comparison with previous theories.

Here we continue the important point made in Levitas and Javanbakht (2010); Levitas

and Samani (2011a,b); Levitas (2013a,b); Levitas (2014b) that a physical phenomenon like interface stresses is introduced with the help of geometric nonlinearity, i.e., large strain contributions, which are retained even in the infinitesimal strain approximation. Thus, the correct expression for the anisotropic gradient energy requires keeping the difference between gradients in the reference and actual configuration even at infinitesimal strains. Indeed, if we assume that $\psi^\nabla = \psi^\nabla(|\nabla_0\eta|, \mathbf{k}_0)$ because of small strains, then the interface stresses will not appear at all. If we assume that $\psi^\nabla = \psi^\nabla(|\nabla\eta|, \mathbf{k})$ because of small strains, then the stress tensor is non-symmetric and the moment of momentum balance is violated. Thus, even for infinitesimal strains all derivations should be performed assuming finite strains and small-strain terms can be neglected in the final equations only. Fortunately, with such an approach, we were able to easily generalize an analytical solution for a propagating interface and an expression for the interface energy, width, and stresses, as well as the position of the Gibbsian dividing surface, obtained for the isotropic interface energy for the case with anisotropic interface energy. Surprisingly, the first order correction in the Ginzburg-Landau equation due to small strains is related to the anisotropy of the interface energy, which was completely neglected before; the next term with the transformation work is the third order of smallness in strain only, but was always taken into account in coupled Ginzburg-Landau and mechanics studies.

The developed formalism is applicable to melting/solidification (Anderson et al. (2000, 2001); Debierre et al. (2003); Kobayashi (1993); Levitas and Samani (2011a,b); Slutsker et al. (2006); Warren-Boettinger (1995)) and the crystal-amorphous transformation. Similar formalism can be applied to fracture (Jin et al. (2001b)). It can be generalized for martensitic and diffusive phase transformations (Artemev and Khachuaturyan (2001); Chen (2002); Finel et al. (2010); Jin et al. (2001a); Levitas et al. (2004); Levitas and Lee (2007); Vedantam and Abeyaratne (2005)), twinning (Clayton and Knap (2011a,b); Hildebrand and Miehe (2012); Levitas et al. (2013); Levitas and Roy (2015)), grain growth (Kobayashi et al.

(1998)), and interaction of cracks and dislocations with phase transformations (Boulbitch and Toledano (1998); Boulbitch and Korzhenevskii (2011); Idesman et al. (2000); Javanbakht and Levitas (2015); Levitas et al. (1998); Levitas (2000); Levitas and Javanbakht (2012, 2013, 2014); Levitas and Javanbakht (2015)), for which interface energy depends on the interface orientation of crystals from both its sides. This is, however, a topic for a separate study.

Reorientation of an interface, even for the case of isotropic interface energy, may occur under action of the stress tensor. The thermodynamic driving force for such a reorientation was determined in Levitas and Ozsoy (2009a,b). In PFA one does not need additional efforts to obtain such an effect provided that the PFA and mechanics equations are coupled. Also, anisotropy can be introduced in the expression for energy of the external surface for a sharp (Levitas and Javanbakht (2010); Levitas and Samani (2011a); Lipowsky (1982)) and finite-width (Levitas and Janbakht (2011b); Levitas and Samani (2014)) treatment of the external boundaries. It may lead to reshaping and faceting of nanowires (Levitas et al. (2012)) and other nanoobjects. It also can be included for melting within grain boundaries (Lobkovsky and Warren (2002)) and at the interfaces between two solid phases (Levitas (2005); Levitas et al. (2012); Levitas and Momeni (2014); Luo and Chiang (2008); Momeni and Levitas (2014); Momeni et al. (2015)).

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nor is it intended to imply that the materials or software identified are necessarily the best available for the purpose.

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Appendix

1. *Principle of material objectivity for anisotropic $\psi^\nabla(\nabla\eta_i)$.* The previous theories (Anderson et al. (2000, 2001)) that consider the free energy as an anisotropic function of the gradient of the order parameters in the deformed configuration $\psi^\nabla(\nabla\eta_i)$ require examination from the point of view of the principle of material objectivity. Since $\nabla\eta_i$ transform under superposed rigid-body rotations in the actual configuration, $\nabla\eta_i^* = \mathbf{Q} \cdot \nabla\eta_i^*$, then function $\psi^\nabla(\nabla\eta_i^*)$ can be invariant only if supplemented by an explicit introduction of tensors $\mathbf{K}_{Rj} = \mathbf{R}_e \times \mathbf{K}_j$ that describe material anisotropy in the actual configuration and also rotate together with a body in the actual configuration; here $\mathbf{K}_j$ are constant tensors and $\mathbf{R}_e \times$ means that each basis vector of the tensor $\mathbf{K}_j$ is contracted with the rotation tensor $\mathbf{R}_e$. Then $\psi^\nabla(\nabla\eta_i, \mathbf{K}_j) = \psi^\nabla(\nabla\eta_i^*, \mathbf{K}_j^*) = \psi^\nabla(\mathbf{Q} \cdot \nabla\eta_i, \mathbf{Q} \cdot \mathbf{K}_{Rj})$ and by choosing $\mathbf{Q} = \mathbf{R}_e^T$, one obtains the objective expression $\psi^\nabla(\mathbf{R}_e^T \cdot \nabla\eta_i, \mathbf{K}_j) = \tilde{\psi}^\nabla(\mathbf{R}_e^T \cdot \nabla\eta_i)$, where constant tensors $\mathbf{K}_j$ are excluded from the list of explicit arguments. Thus, while differentiating function ψ^∇ one has to take into account its explicit dependence on $\mathbf{R}_e^T$ and appearance of corresponding additional terms, which was not done before.

However, this is irrelevant for our treatment and our main goal is to change a contradictory assumption $\psi^\nabla = \psi^\nabla(\nabla\eta_i)$ with the physically correct one.

2. *Micropolar theory.* Both problems could be in principle resolved by considering more general micropolar theory with some microstructure (Eringen and Kafadar (1976); Kafadar and Eringen (1971); Mindlin (1964); Toupin (1964)), which rotates with the angular velocity $\tilde{\omega}$ under action of some body couples $\mathbf{Y}$ and couple stress tensor $\boldsymbol{\mu}$. In this theory, the moment of momentum balance equation takes form

$$-2\sigma_a : \boldsymbol{\epsilon} + \nabla \cdot \boldsymbol{\mu} + \rho \mathbf{Y} = \rho \mathbf{J} \cdot \dot{\tilde{\omega}}; \quad \epsilon_{imn} \sigma_{mn} + \nabla_m \mu_{mi} + \rho Y_i = \rho J_{ij} \dot{\tilde{\omega}}_j. \quad (151)$$

Here $\mathbf{J}$ is the symmetric moment of microinertia tensor, the Levi-Civita symbols ϵ_{ilk} are equal to 1 or -1 if ilk is an even or odd permutation of 123, and 0 for any equal indices i, l , or k ; corresponding third-rank tensor is designated as $\boldsymbol{\epsilon}$. Such a theory, which is objective, is essentially more sophisticated. It requires proper physical interpretation of the additional microstructure and finding additional constitutive equations for the body couples and couple stresses, and material parameters. As examples of such a microstructure at the scale of an interface width ($\sim 1nm$) one can consider orientation of macromolecules of polymers or

director vector of liquid crystals. For solid-liquid metal interface, we are unable to identify any reasonable microstructure. Thus ω should be associated with rotation of the normal to interface $\mathbf{k}$ and this kinematic constrain should be incorporated in a thermomechanical theory. While such a theory can be in principle elaborated, it still will have a conceptual contradiction. It is based on introduction in the moment of momentum equation the term σ_a related to orientation dependence of the energy (i.e., the counterpart of the Herring torque). This contradicts to our conclusion in Introduction that the Herring torque is not a mechanical torque localized at the interface. That is why anisotropy of the interface energy does not change momentum of momentum equations and, consequently, the Cauchy stress tensor cannot be non-symmetric.

3. Let evaluate derivatives of $\beta(\zeta_0)$ (Eq. (65)) that participate in Eqs. (52)-(53):

$$\begin{aligned} \frac{\partial \beta}{\partial \zeta_{0i}} = & \alpha_0 \frac{\zeta_{0i}}{|\zeta_0|} + \alpha_1 \left(2 \frac{\zeta_{0i} (\zeta_{0j}^2 + \zeta_{0k}^2)}{|\zeta_0|^3} - 3 \frac{\zeta_{0i} (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)}{|\zeta_0|^5} \right) + \\ & \alpha_2 \left(\frac{2 \zeta_{0i} \zeta_{0j}^2 \zeta_{0k}^2}{|\zeta_0|^5} - \frac{5 \zeta_{0i}^3 \zeta_{0j}^2 \zeta_{0k}^2}{|\zeta_0|^7} \right) + \\ & \alpha_3 \left(\frac{4 \zeta_{0i} (\zeta_{0j}^2 + \zeta_{0k}^2) (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)}{|\zeta_0|^7} - \frac{7 \zeta_{0i} (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)^2}{|\zeta_0|^9} \right) \end{aligned} \quad (152)$$

$$\begin{aligned} \frac{\partial^2 \beta}{\partial \zeta_{0i} \partial \zeta_{0k}} = & -\alpha_0 \frac{\zeta_{0i} \zeta_{0k}}{|\zeta_0|^3} - \alpha_1 \left(\frac{2 \zeta_{0i} \zeta_{0k} (\zeta_{0i}^2 + \zeta_{0k}^2 + 4 \zeta_{0j}^2)}{|\zeta_0|^5} - \frac{15 \zeta_{0i} \zeta_{0k} (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)}{|\zeta_0|^7} \right) \\ & - \alpha_2 \frac{\zeta_{0i} \zeta_{0j}^2 \zeta_{0k} (6 \zeta_{0i}^4 - 4 \zeta_{0j}^4 + 6 \zeta_{0k}^4 + 2 \zeta_{0j}^2 \zeta_{0k}^2 + 2 \zeta_{0i}^2 \zeta_{0j}^2 - 23 \zeta_{0i}^2 \zeta_{0k}^2)}{|\zeta_0|^9} + \\ & \alpha_3 \left(\frac{8 \zeta_{0i} \zeta_{0k} (\zeta_{0j}^4 + 2 \zeta_{0j}^2 \zeta_{0k}^2 + 2 \zeta_{0i}^2 (\zeta_{0k}^2 + \zeta_{0j}^2))}{|\zeta_0|^7} + \frac{63 \zeta_{0i} \zeta_{0k} (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)^2}{|\zeta_0|^{11}} - \right. \\ & \left. \frac{28 \zeta_{0i} \zeta_{0k} (\zeta_{0i}^4 (\zeta_{0k}^2 + \zeta_{0j}^2) + \zeta_{0k}^2 \zeta_{0j}^2 (\zeta_{0k}^2 + 2 \zeta_{0j}^2) + \zeta_{0i}^2 (2 \zeta_{0j}^4 + 4 \zeta_{0j}^2 \zeta_{0k}^2 + \zeta_{0k}^4))}{|\zeta_0|^9} \right), \end{aligned} \quad (153)$$

and

$$\frac{\partial^2 \beta}{\partial \zeta_{0i}^2} = \alpha_0 \left(\frac{1}{|\zeta_0|} - \frac{\zeta_{0i}^2}{|\zeta_0|^3} \right) + \alpha_1 \left(\frac{2 (\zeta_{0j}^2 + \zeta_{0k}^2)}{|\zeta_0|^3} - \frac{3 (3 \zeta_{0i}^2 \zeta_{0j}^2 + 3 \zeta_{0i}^2 \zeta_{0k}^2 - \zeta_{0k}^2 \zeta_{0j}^2)}{|\zeta_0|^5} + \right.$$

$$\begin{aligned}
& \frac{15\zeta_{0i}^2 (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)}{|\zeta_0|^7} \Bigg) + \alpha_2 \frac{\zeta_{0j}^2 \zeta_{0k}^2 (12\zeta_{0i}^4 - 21\zeta_{0i}^2 (\zeta_{0j}^2 + \zeta_{0k}^2) + 2 (\zeta_{0j}^2 + \zeta_{0k}^2)^2)}{|\zeta_0|^9} + \\
& \alpha_3 \left(\frac{4 (\zeta_{0k}^2 + \zeta_{0j}^2) (\zeta_{0j}^2 \zeta_{0k}^2 + 3\zeta_{0i}^2 (\zeta_{0k}^2 + \zeta_{0j}^2))}{|\zeta_0|^7} + \frac{63\zeta_{0i}^2 (\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2)^2}{|\zeta_0|^{11}} - \right. \\
& \left. \frac{7 (\zeta_{0j}^4 \zeta_{0k}^4 + 10\zeta_{0i}^2 \zeta_{0j}^2 \zeta_{0k}^2 (\zeta_{0k}^2 + \zeta_{0j}^2) + 9\zeta_{0i}^4 (\zeta_{0k}^2 + \zeta_{0j}^2)^2)}{|\zeta_0|^9} \right). \tag{154}
\end{aligned}$$

4. *Equations for entropy and temperature evolution.* After elaborating all terms in Eq.(20)₁ for the entropy in a way similar to that in Levitas (2014b) we obtain

$$s = \frac{1}{\rho_0} \mathbf{P}_e : \mathbf{U}_t \cdot \frac{\partial \mathbf{U}_\theta}{\partial \theta} \cdot \mathbf{F}_e^T - \frac{J_{t\theta}}{\rho_0} \frac{\partial \psi^e}{\partial \theta} \Big|_{\mathbf{F}_e} - \frac{J_t}{\rho_0} \psi^e \frac{\partial J_\theta}{\partial \theta} - J \frac{\partial \check{\psi}^\theta}{\partial \theta} - \frac{\partial \tilde{\psi}^\theta}{\partial \theta} - \frac{\partial (\beta(\theta, \mathbf{k}_0))^2}{\partial \theta} \frac{J}{2\rho_0} |\nabla \eta_i|^2; \tag{155}$$

$$s = \frac{1}{\rho} \boldsymbol{\sigma}_e : \mathbf{F}_e \cdot \frac{\partial \mathbf{U}_\theta}{\partial \theta} \cdot \mathbf{U}_\theta^{-1} \cdot \mathbf{F}_e^{-1} - \frac{J_e^{-1}}{\rho} \frac{\partial \psi^e}{\partial \theta} \Big|_{\mathbf{E}_e} - \frac{J_e^{-1} J_\theta^{-1}}{\rho} \psi^e \frac{\partial J_\theta}{\partial \theta} - J \frac{\partial \check{\psi}^\theta}{\partial \theta} - \frac{\partial \tilde{\psi}^\theta}{\partial \theta} - \frac{\partial (\beta(\theta, \mathbf{k}_0))^2}{\partial \theta} \frac{1}{2\rho} |\nabla \eta_i|^2. \tag{156}$$

If interface energy depends on temperature through parameter β (i.e., through α_i), then the entropy obtains contribution from the gradient of the order parameters. Similar to the Ginzburg-Landau equation, elastic stresses affect entropy, while interface and viscous stresses do not. For the small strains and rotations, any of the Eqs.(155) or (156) reduce to

$$s = \frac{1}{\rho_0} \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_\theta}{\partial \theta} - \frac{1}{\rho_0} \frac{\partial \psi^e}{\partial \theta} \Big|_{\boldsymbol{\varepsilon}_e} - \frac{1}{\rho_0} \psi^e \frac{\partial J_\theta}{\partial \theta} - \frac{\partial \check{\psi}^\theta}{\partial \theta} - \frac{\partial \tilde{\psi}^\theta}{\partial \theta} - \frac{\partial (\beta(\theta, \mathbf{k}_0))^2}{\partial \theta} \frac{1}{2\rho_0} |\nabla \eta_i|^2. \tag{157}$$

If one derives equation for entropy starting with small strain formulation, the third term in Eq.(156) will be missed; it may be, however, comparable with the second term.

To derive the temperature evolution equation, we combine equations $\bar{\mathcal{D}} := \theta \tilde{S}_i$, (13), (14), and (21):

$$\theta \dot{s} = \mathcal{D} + r - \rho_0^{-1} \nabla_0 \cdot \mathbf{h}_0 = \mathbf{P}_v : \dot{\mathbf{F}}^T + X_i \dot{\eta}_i + r - \rho_0^{-1} \nabla_0 \cdot \mathbf{h}_0. \tag{158}$$

Based on Eq.(155), we present $s = s(\theta, \mathbf{P}_e, \eta_i, \nabla_0 \eta_i)$. In this equation, we express $\nabla \eta_i$ via $\nabla_0 \eta_i$ and $\mathbf{F}$; deformation gradient $\mathbf{F}$ will be substituted with its multiplicative decomposition

(6); $\mathbf{F}_t$ and $\mathbf{F}_\theta$ will be expressed in terms of η_i and θ , and $\mathbf{F}_e$ is excluded utilizing the elasticity rule. After substitution $s = s(\theta, \mathbf{P}_e, \eta_i, \nabla_0 \eta_i)$ in the entropy evolution equation (158) and definition of the specific heat at constant elastic Piola-Kirchhoff stress, $c_{\mathbf{p}} := \theta \frac{\partial s}{\partial \theta}$, and resolving for the temperature rate, we obtain the temperature evolution equation

$$c_{\mathbf{p}} \dot{\theta} = \mathbf{P}_v : \dot{\mathbf{F}}^T + \left(X_i - \theta \frac{\partial s}{\partial \eta_i} \right) \dot{\eta}_i + r - \rho_0^{-1} \nabla_0 \cdot \mathbf{h}_0 - \theta \frac{\partial s}{\partial \mathbf{P}_e} : \dot{\mathbf{P}}_e^T - \theta \frac{\partial s}{\partial \nabla_0 \eta_i} : \dot{\nabla}_0 \eta_i. \quad (159)$$

Utilizing the Fourier thermal conductivity law in the reference configuration, $\mathbf{h}_0 = -\mathbf{\Lambda}_0 \cdot \nabla_0 \theta$ ($\mathbf{\Lambda}_0$ is the thermal conductivity tensor), Eq.(159) specifies to

$$c_{\mathbf{p}} \dot{\theta} = \mathbf{P}_v : \dot{\mathbf{F}}^T + \left(X_i - \theta \frac{\partial s}{\partial \eta_i} \right) \dot{\eta}_i + r + \rho_0^{-1} \nabla_0 \cdot (\mathbf{\Lambda}_0 \cdot \nabla_0 \theta) - \theta \frac{\partial s}{\partial \mathbf{P}_e} : \dot{\mathbf{P}}_e^T - \theta \frac{\partial s}{\partial \nabla_0 \eta_i} : \dot{\nabla}_0 \eta_i. \quad (160)$$