# Thermodynamically consistent phase field theory of phase transformations with anisotropic interface energies and stresses

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The main focus of this paper is to introduce, in a thermodynamically consistent manner, an anisotropic interface energy into a phase field theory for phase transformations. Here we use a small strain formulation for simplicity, but we retain some geometric nonlinearities, which are necessary for introducing correct interface stresses. Previous theories have assumed the free energy density (i.e., gradient energy) is an anisotropic function of the gradient of the order parameters in the current (deformed) state, which yields a nonsymmetric Cauchy stress tensor. This violates two fundamental principles: the angular momentum equation and the principle of material objectivity. Here, it is justified that for a noncontradictory theory the gradient energy must be an isotropic function of the gradient of the order parameters in the current state, which also depends anisotropically on the direction of the gradient of the order parameters in the reference state. A complete system of thermodynamically consistent equations is presented. We find that the main contribution to the Ginzburg-Landau equation resulting from small strains arises from the anisotropy of the interface energy, which was neglected before. The explicit expression for the free energy is justified. An analytical solution for the nonequilibrium interface and critical nucleus has been found and a parametric study is performed for orientation dependence of the interface energy and width as well as the distribution of interface stresses.

## I. INTRODUCTION

The phase field approach (PFA) is routinely utilized for the simulation of various first-order phase transformations (PTs), including martensitic PTs [1–8], melting [9–14], twinning [15,16], and grain growth [17]. In PFA, the energy density of the system depends on the so-called order parameters $\eta_i$, $i = 1, 2, \ldots, n$, and their gradients, in addition to the strain tensor and temperature. In most cases the order parameters represent internal variables (the exceptions are components of the strain tensor for martensitic PTs, e.g., [3,8]), which describe material instabilities during structural changes in a continuous way. The energy of the system for each strain tensor and temperature has multiwell structure, i.e., it has a multiple local minima separated by energy barriers. Each minimum corresponds to a separate phase or structural state. Gradients of the order parameters are localized at the interfaces between phases and penalize the interface energy. The evolution of the microstructure is described by the Ginzburg-Landau equations, which are obtained as linear relationships between $\dot{\eta}_i$ and their conjugates, the thermodynamic forces $X_i$, together with a complete set of equations of continuum thermomechanics. In contrast to the sharp-interface approach, the solution exhibits finite-width interfaces, within which order parameters vary smoothly between the values corresponding to the local energy minima. Unlike sharp interface approaches, all one has to do is solve the above system of equations, there is no need for computational efforts to track interfaces.

Since the strain tensor is one of the thermodynamic parameters that governs a PT, the PFA is combined with the strict description of the deformation process, see, e.g.,

text book on continuum mechanics [18]. The motion of a material will be described by a continuous function $\boldsymbol{r} = \boldsymbol{r}(\boldsymbol{r}_0, t)$, where $\boldsymbol{r}_0$ and $\boldsymbol{r}$ are the positions of points in the reference (undeformed) $\Omega_0$ and the actual (deformed) $\Omega$ states (configurations), respectively, and $t$ is the time. In general, deformation may or may not cause PTs.

Recently, significant efforts have been devoted to introducing interface stresses in the PFA. Initially, liquid-liquid and liquid-solid interfaces were treated, for which interface stresses play a key role. For a liquid-liquid interface the interface stresses represent biaxial tension with a magnitude equal to the interface energy $\gamma$ [Fig. 1(a)]. Since, at the nanoscales and even microscales, interface stresses are important for solid-liquid and solid-solid interfaces, and have been broadly studied within the sharp-interface approach [19–27] as well molecular dynamics [28,29], the interface stresses have been introduced in PFA as well, see Refs. [11,12,14,30] for melting and [31–35] for solid-solid PTs. Here we will follow the most advanced theories at small strains [33] and large strains [35], where a detailed literature review is presented with critical analysis of the previous approaches. As the sharp-interface counterpart, we will start with the Shuttleworth equation [20,27] for the magnitude $\bar{\sigma}^S$ of the interface stress,

$$\bar{\sigma}^S = \gamma + \partial\gamma/\partial\varepsilon_s = \bar{\sigma}_{st} + \bar{\sigma}_e^S, \qquad (1)$$

where $\varepsilon_s$ is the interface strain and subscript $st$ means the structural part of the interface stresses. The interface stress consists of two contributions: (i) what we called the structural part, $\bar{\sigma}_{st} = \gamma$, as for a liquid-liquid interface, and (ii) the part $\bar{\sigma}_e^S$ due to elastic deformation of an interface. We use a bar above the symbol $\sigma$ for these stresses, because stresses in Eq. (1) are localized at the zero-width interface and have dimensions of force per unit interface length rather than area.
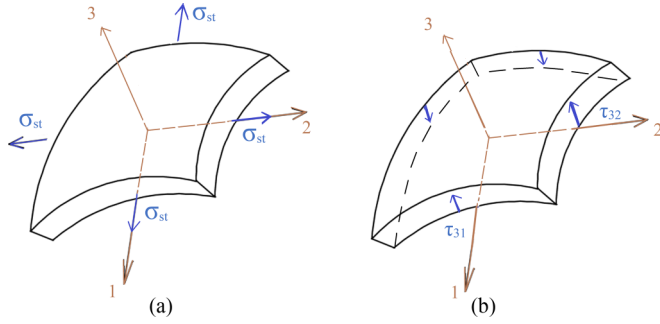
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FIG. 1. (Color online) Interface stresses. (a) Biaxial tension with the resultant force equal to the orientation-dependent nonequilibrium interface energy that appears in the current theory; (b) Additional artificial interface shear stresses, which are present in the previous theories and are absent in the current approach. Because shear stresses are nonsymmetric they produce artificial equilibrated torque.

Using the PFA, it is significant that the elastic contribution to the surface stresses comes directly from the solution of the Ginzburg-Landau and elasticity equations for a PT problem without any additional conditions [12,14,30]. These contributions appear due to a heterogeneous distribution of the transformation strain and elastic moduli across a finite-width interface. Even for a solid-melt interface, elastic stresses in PFA are much higher than those obtained by molecular dynamics [28,29], which leads to contradictions with the experimental data on the size dependence of the melting temperature for Al nanoparticles [14,30]. To remedy this discrepancy, additional relaxation equations for the elastic interface stresses are suggested in Refs. [14,30] to obtain correspondence with experiments. Thus, even for melting, when shear modulus tends to zero across an interface, introducing extra elastic interface stresses would be harmful. That is why our main hypothesis in Refs. [14,30,32–35] is that elastic interface stresses are completely defined from the solution of the Ginzburg-Landau and mechanics equations for a PT problem. Thus, the main problem reduces to introducing the structural contribution to the interface stresses $\sigma_{st}$ (see Fig. 1 with $\bar{\sigma}_{st} = \gamma$), i.e., as for liquid-liquid or liquid-gas interfaces.

This problem has been solved in Refs. [33,34,36] for small strain approximation and in Ref. [35] for general large strain formulation, but for the case of isotropic interface energy and, consequently, isotropic gradient energy $\psi^\nabla(\nabla \eta_i)$. It is important to note that in order to introduce interface stresses (i.e., physical phenomenon) that represent biaxial tension with a magnitude equal to the nonequilibrium interface energy, it is necessary to introduce some geometrically nonlinear features even when strains are infinitesimally small. In particular, the gradient of the order parameters, $\nabla \eta_i$, should be evaluated with respect to the deformed configuration. The goal of the current paper is to generalize PFA presented in Ref. [33] for anisotropic interface energy $\gamma(\boldsymbol{k})$, i.e., for anisotropic function $\psi^\nabla(\nabla \eta_i)$, where $\boldsymbol{k} := \nabla \eta_i / |\nabla \eta_i|$ is the unit normal to the interface. Anisotropy of the interface energy plays a very important part in the solidification and growth of dendrites. Anisotropy determines morphology and kinetics of crystal growth, and is important for crystal-crystal phase transformation, fracture, and grain growth as well.

An anisotropic interface energy has been treated in numerous publications [9,10,13,37–43] by considering an anisotropic function of $\nabla \eta_i$ in the deformed state, i.e., $\psi^\nabla(\nabla \eta_i)$. However, the anisotropic function $\psi^\nabla(\nabla \eta_i)$ results in a nonsymmetric contribution to the true (Cauchy) stress $\nabla \eta_i \otimes \frac{\partial \psi^\nabla}{\partial \nabla \eta_i}$ [33,35,37]. As we will discuss in Sec. II this leads to a violation of the angular momentum balance, which requires symmetry of the true stress. It is also possible to show that the stress power of the nonsymmetric stress is not invariant under superposition of the rigid-body rotation, which violates the principle of material objectivity. However, these basic contradictions have never been mentioned in the previous publications. We will justify that in order to noncontradictorily describe anisotropic interface energy, the gradient energy should be an isotropic function of the gradient of the order parameters in the deformed state, $\nabla \eta_i$, and in addition should depend on the direction of the normal to the interface in the reference (undeformed) state, $\boldsymbol{k}_0 := \nabla_0 \eta_i / |\nabla_0 \eta_i|$. This is consistent with the description of the anisotropic energy of the sharp interface in the undeformed state, $\gamma(\boldsymbol{k}_0)$, for which the crystal lattice symmetry group is known, rather than in the deformed state, $\gamma(\boldsymbol{k})$, for which the lattice symmetry also depends of the deformation gradient $\boldsymbol{F}$. For such a formulation, the true stress remains symmetric.

We would like to mention that there are various generic steps in some equation derivations for anisotropic interface energy that are the same or similar to those in Ref. [33] for isotropic energy. They will be repeated as briefly as possible and we will refer to Ref. [33] for detail.

The paper is organized as follows. In Sec. II the main problem formulation is justified, namely that the gradient energy should assume the form in $\psi^\nabla(|\nabla \eta|, \boldsymbol{k}_0)$ for a single order parameter and $\psi^\nabla = \psi^\nabla(\nabla \eta_k \cdot \nabla \eta_j, \boldsymbol{k}_{0i})$ for multiple order parameters, see Eq. (4). A thermodynamic treatment is performed in Sec. III, including the derivation of the general structure of the constitutive equations. The expression for the free energy that results in the desired structure for the interface stress tensor, including its symmetry, is specified in Sec. IV. An explicit expression for the Ginzburg-Landau equations is analyzed in detail in Sec. V. Anisotropy of the gradient energy produces many extra terms in these equations. Surprisingly, all of them are of the first degree of smallness for small strains, while the next term (transformation work) is of the third degree of smallness in strains. A complete system of equations is summarized in Sec. VI. In Sec. VII an analytical solution for the nonequilibrium interface propagating in an arbitrary direction $\boldsymbol{k}$ is presented, and the temperature and orientation dependence of the interface energy and width is determined. Artificial shear stresses and moments in the previous theories are analyzed in Sec. VIII. The case of strong anisotropy is elaborated in Sec. IX. Explicit results for a specific model are obtained in Sec. X. The orientation dependence of the distribution of the interface stresses for a critical nucleus is given in Sec. XI. Section XII contains concluding remarks.

We designate the contractions of tensors $\boldsymbol{A} = \{A_{ij}\}$ and $\boldsymbol{B} = \{B_{ji}\}$ over one and two indices as $\boldsymbol{A} \cdot \boldsymbol{B} = \{A_{ij} B_{jk}\}$ and $\boldsymbol{A}:\boldsymbol{B} = A_{ij} B_{ji}$, respectively. The subscripts $s$ and $a$ designate the symmetric and the skew-symmetric part of a second-rank tensor; subscripts $e$, $t$, and $\theta$ mean elastic, transformational,

and thermal strains; $\boldsymbol{I}$ is the unit tensor; $\delta_{ij}$ is the Kronecker $\delta$; parameters in the undeformed state will be designated with subscript 0 and in the deformed state will not have any subscript; in particular, $\boldsymbol{\nabla}$ and $\boldsymbol{\nabla}_0$ are the gradient operators in the deformed and undeformed states, respectively; and $\otimes$ designates a dyadic product and $:=$ is equal by definition.

## II. PROBLEM FORMULATION

### A. Drawbacks of existing approaches

*Sharp-interface approach.* There are two driving forces for interface motion: (i) the Eshelby driving force for the translational interface motion [44–47]

$$X_\Sigma = -\Delta G - 2\gamma \kappa_{av}, \tag{2}$$

and (ii) the Herring torque [48] for interface reorientation

$$\boldsymbol{X}_k = -\frac{\partial \gamma}{\partial \boldsymbol{k}}. \tag{3}$$

Here $\Delta G$ is the jump in the Gibbs energy across the interface and $\kappa_{av}$ is the averaged interface curvature. While in Eq. (3) anisotropy of $\gamma$ is the key to the existence of $X_\Sigma$, we keep isotropic $\gamma$ in Eq. (2) for simplicity because here it is sufficient for our descriptive purpose. Both $X_\Sigma$ and $\boldsymbol{X}_k$ are so-called thermodynamic configurational forces [44–47], which do not contribute explicitly to the equations of mechanics, namely, linear momentum balance and angular momentum balance. They do not describe the motion of material points, rather they describe the motion of interfaces with respect to the material. The confusion is often related to the fact that when one determines a thermodynamically equilibrium position of interfaces and their junctions, the conditions are applied so that the resultant force and torque (or their work) are zero, i.e., like in mechanics. However, this is mechanics of configurational forces, which is independent of the linear and angular momentum balances. This is in contrast to the interface stresses, which do contribute to the momentum balance equation.

*Phase field approach.* An anisotropic interface energy within PFA was broadly studied in numerous publications [9,10,13,37–43], in which the gradient energy is an anisotropic function of $\boldsymbol{\nabla}\eta_i$ in the current (deformed) state, i.e., $\psi^\nabla = \psi^\nabla(\boldsymbol{\nabla}\eta_i)$. However, there is a conceptual contradiction with the main principles of continuum mechanics in all these theories. Thus, the anisotropic function $\psi^\nabla(\boldsymbol{\nabla}\eta_i)$ results in a generally nonsymmetric contribution to the true (Cauchy) stress $\boldsymbol{\sigma}$ (i.e., force per current unit area) $\boldsymbol{\nabla}\eta_i \otimes \frac{\partial \psi^\nabla}{\partial \boldsymbol{\nabla}\eta_i}$ [10,33,35,37]. This, however, violates the angular momentum balance, which requires symmetry of the true stress (see any textbook on continuum mechanics, e.g., Ref. [18]). It is also easy to show that the nonsymmetric stress tensor $\boldsymbol{\sigma}$ produces stress power $\boldsymbol{\sigma}{:}\boldsymbol{\nabla}\boldsymbol{v}^t$, where $\boldsymbol{v}$ is the particle velocity, that is not invariant under superposition of the rigid-body rotation in the deformed state, i.e., it contradicts the principle of material objectivity. Each of these contradictions makes such a theory inadmissible from the point of view of traditional continuum mechanics. This, however, was completely overlooked in the previous theories.

*Micropolar theory.* Note that nonsymmetric stress is routinely used in a more general micropolar theory with some mi-

crostructure [49,50], which rotates with respect to a continuum with some angular velocity. In this theory some body couples, the couple stress tensor, and the rotational moment of inertia equilibrate the antisymmetric part of the stress in the angular momentum balance. However, apart from the unjustified complexity, there are two reasons why this theory is not applicable for our case. First, changing the angular momentum equation due to orientation dependence of the energy (i.e., the counterpart of the Herring torque) in PFA contradicts our statement that the Herring torque does not contribute to the moment of the momentum balance. Indeed, the driving force to interface rotation in the PFA due to anisotropy of the gradient energy also should not change the angular momentum balance, similar to the fact that the driving force for change in order parameters (i.e., interface translational motion) does not change the momentum equation. Both are configurational forces that cause translational and rotational interface motion with respect to the material but do not produce mechanical torque and force that contribute to the angular momentum balance and momentum equation. That is why the true stress tensor in PFA cannot be nonsymmetric due to anisotropy of the interface energy and $\psi^\nabla$ cannot be an anisotropic function of $\boldsymbol{\nabla}\eta_i$. Second, we are unable to identify any meaningful rotating microstructure within a nm-thick interface that is not present in a bulk.

We remark here that many finite strain theories for melting, martensitic PT, twinning, and fracture utilize gradient of the order parameter in the reference configuration [15,31,51–54] and include anisotropy of the interface energy. We do not consider such an approach here because it results in $\boldsymbol{\sigma}_{\text{st}} = 0$.

### B. Problem formulation

*Sharp-interface approach.* An important question is: Should $\gamma$ depend on the unit normal $\boldsymbol{k}$ in the deformed state or on the unit normal $\boldsymbol{k}_0$ in the undeformed state? Since the symmetry group of the crystal lattice is well defined in the undeformed state only, the interface energy $\gamma$ can be presented as a function $\gamma(\boldsymbol{k}_0)$, which is invariant with respect to the symmetry group of the undeformed lattice. Since deformation of the lattice described by the deformation gradient $\boldsymbol{F}$ changes its symmetry, the interface energy cannot be a function of $\boldsymbol{k}$ only but must also depend on $\boldsymbol{F}$, i.e., $\gamma = \gamma(\boldsymbol{k},\boldsymbol{F})$. The only way to determine this function is to find the $\gamma(\boldsymbol{k}_0)$ that corresponds to the symmetry of the undeformed lattice and express $\boldsymbol{k}_0$ in terms of $\boldsymbol{k}$ and $\boldsymbol{F}$. This is similar to the formulation of strain energy for an anisotropic elastic material: it is formulated in the undeformed state and, if desired, is expressed in terms of strain measures defined in the deformed state.

*Phase field approach.* Similar consideration will be applied here. For an isotropic interface energy the correct expression for the gradient energy that results in the desired expression for the interface stresses is $\psi^\nabla(|\boldsymbol{\nabla}\eta|)$ for a single order parameter and $\psi^\nabla = \psi^\nabla(\boldsymbol{\nabla}\eta_k \cdot \boldsymbol{\nabla}\eta_j)$ for the multiple order parameters. For a single order parameter and anisotropic interface energy, the options are $\psi^\nabla(|\boldsymbol{\nabla}\eta|,\boldsymbol{k}_0)$ or $\psi^\nabla(|\boldsymbol{\nabla}\eta|,\boldsymbol{k}) = \tilde{\psi}^\nabla(\boldsymbol{\nabla}\eta)$. The second option has already been explored [10,33,35,37] and results in a nonsymmetric stress tensor, which, as we found, is forbidden. The first option, since the second argument, $\boldsymbol{k}_0$, is strain-independent it does not change the general expression

for stress compared to the isotropic case, i.e., the stress tensor remains symmetric (see Sec. IV). The function $\psi^\nabla(|\boldsymbol{\nabla}\eta|, \boldsymbol{k}_0)$ also corresponds to $\gamma(\boldsymbol{k}_0)$ for a sharp interface and can be made invariant with respect to a known symmetry group of the undeformed crystal lattice. Similarly, for multiple order parameters, the anisotropy should be described in terms of $\boldsymbol{k}_{0i}$. Consequently, we will develop a theory below based on the symmetric stress tensor and gradient energy of the form

$$\psi^\nabla = \psi^\nabla(\boldsymbol{\nabla}\eta_k \cdot \boldsymbol{\nabla}\eta_j, \boldsymbol{k}_{0i}) \quad \text{or} \quad \psi^\nabla(|\boldsymbol{\nabla}\eta|, \boldsymbol{k}_0). \quad (4)$$

## III. THERMODYNAMIC TREATMENT

Due to the necessity of distinguishing between deformed and undeformed states, allowing for anisotropy of the interface energy in PFA can be done strictly in the framework of fully large strain formulation only, i.e., by generalizing results obtained in Ref. [35]. This is what we did, and then we simplified the final results for small strains to obtain consistent linearization. However, to simplify the presentation and to broaden our audience, we started with a geometrically linear formulation and kept only those geometrically nonlinear terms, which we found by simplifying the strict theory.

Let small distortions of a material be described by a continuous function $\boldsymbol{r} = \boldsymbol{r}(\boldsymbol{r}_0, t)$, where $\boldsymbol{r}_0$ and $\boldsymbol{r}$ are the positions of points in the reference (undeformed) $\Omega_0$ and the actual (deformed) $\Omega$ states (configurations), respectively, and $t$ is the time. Assume that at time $t_0$ the material is in the high symmetry phase H and it may be transformed into a number of lower symmetry phases $L_i$, which may include martensitic variants. Each of the PTs $H \leftrightarrow L_i$ are described by a corresponding order parameter $\eta_i$ with $\eta_i = 0 \ \forall i$ for H and $\eta_k = 1$, $\eta_i = 0 \ \forall i \neq k$ for $L_k$. The deformation gradient is $\boldsymbol{F} = \frac{\partial \boldsymbol{r}}{\partial \boldsymbol{r}_0} = \boldsymbol{\nabla}_0 \boldsymbol{u} + \boldsymbol{I} \simeq \boldsymbol{I} + \boldsymbol{\varepsilon} + \boldsymbol{\omega}$, where $\boldsymbol{\varepsilon} = (\boldsymbol{\nabla}_0 \boldsymbol{u})_s$ and $\boldsymbol{\omega} = (\boldsymbol{\nabla}_0 \boldsymbol{u})_a$ are small in comparison with unity symmetric strain and antisymmetric rotation tensors, respectively, and $\boldsymbol{u}$ is the displacement vector. The inverse deformation gradient is $\boldsymbol{F}^{-1} \simeq \boldsymbol{I} - \boldsymbol{\varepsilon} - \boldsymbol{\omega}$, which is easy to check: $\boldsymbol{F} \cdot \boldsymbol{F}^{-1} = \boldsymbol{I} - \boldsymbol{\varepsilon} \cdot \boldsymbol{\varepsilon} - \boldsymbol{\omega} \cdot \boldsymbol{\omega} - \boldsymbol{\omega} \cdot \boldsymbol{\varepsilon} - \boldsymbol{\varepsilon} \cdot \boldsymbol{\omega} \simeq \boldsymbol{I}$. We employ an additive decomposition of the strain tensor $\boldsymbol{\varepsilon}$

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_e + \boldsymbol{\varepsilon}_t(\eta_i) + \boldsymbol{\varepsilon}_\theta(\theta, \eta_i) \quad (5)$$

into elastic $\boldsymbol{\varepsilon}_e$, transformational $\boldsymbol{\varepsilon}_t$, and thermal $\boldsymbol{\varepsilon}_\theta$ parts.

Since we retain a symmetric stress tensor we can repeat the same thermodynamic treatment as in Ref. [33] and we arrive at the following dissipative inequality (see Eq. (10) in Ref. [33]):

$$\rho_0 \mathcal{D} = \boldsymbol{\sigma} : \dot{\boldsymbol{\varepsilon}} - \rho_0 \dot{\psi} - \rho_0 s \dot{\theta} + \boldsymbol{\nabla}_0 \cdot (\boldsymbol{Q}_i^\eta \dot{\eta}_i) \geqslant 0. \quad (6)$$

Here, $\mathcal{D}$ is the rate of dissipation per unit mass, $\rho_0$ is the mass density per unit undeformed volume, $\psi$ and $s$ are the specific Helmholtz free energy and the entropy, both per unit mass, $\theta$ is the temperature, and $\boldsymbol{Q}_i^\eta$ are generalized forces conjugate to $\dot{\eta}_i$ at the surface of a sample, which are introduced in order to balance terms due to the dependence of the thermodynamic potential on $\boldsymbol{\nabla}_0 \eta_i$. While in Ref. [33] this equation was written in terms of parameters (mass densities and gradient operator) determined for the deformed state, it will be more straightforward to perform derivations for anisotropic gradient energy if we use parameters determined per unit undeformed state. Because of the small strain approximation, both approaches

are equivalent, and we will change some parameters (e.g., $\rho$) from undeformed to deformed value for convenience without additional discussion.

Let $\psi = \psi(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}_0 \eta_i, \boldsymbol{\nabla}\eta_i) = \psi(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}_0 \eta_i, \boldsymbol{\nabla}_0 \eta_i \cdot \boldsymbol{F}^{-1}) = \psi(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}_0 \eta_i, \boldsymbol{\nabla}_0 \eta_i \cdot [\boldsymbol{I} - \boldsymbol{\varepsilon} - \boldsymbol{\omega}])$, where we used $\boldsymbol{\nabla}\eta_i = \boldsymbol{\nabla}_0 \eta_i \cdot \boldsymbol{F}^{-1}$. Since $\psi$ must be invariant with respect to the superposed rigid-body rotation, it cannot depend on $\boldsymbol{\omega}$. Thus, the objective form of $\psi$ is $\psi = \psi(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}_0 \eta_i, \boldsymbol{\nabla}_0 \eta_i \cdot (\boldsymbol{I} - \boldsymbol{\varepsilon})) = \bar{\psi}(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}_0 \eta_i)$. We will also need $\boldsymbol{\nabla}_0 \cdot (\boldsymbol{Q}_i^\eta \dot{\eta}_i) = (\boldsymbol{\nabla}_0 \cdot \boldsymbol{Q}_i^\eta) \dot{\eta}_i + \boldsymbol{Q}_i^\eta \cdot \boldsymbol{\nabla}_0 \dot{\eta}_i$. Substituting $\dot{\psi}$ and the above equation into Eq. (6), we obtain

$$\rho_0 \mathcal{D} = \left( \boldsymbol{\sigma} - \rho_0 \frac{\partial \bar{\psi}}{\partial \boldsymbol{\varepsilon}} \right) : \dot{\boldsymbol{\varepsilon}} - \rho_0 \left( s + \frac{\partial \bar{\psi}}{\partial \theta} \right) \dot{\theta}$$
$$- \left( \rho_0 \frac{\partial \bar{\psi}}{\partial \eta_i} - \boldsymbol{\nabla}_0 \cdot \boldsymbol{Q}_i^\eta \right) \dot{\eta}_i$$
$$+ \left( \boldsymbol{Q}_i^\eta - \rho_0 \frac{\partial \bar{\psi}}{\partial \boldsymbol{\nabla}_0 \eta_i} \right) \cdot \boldsymbol{\nabla}_0 \dot{\eta}_i \geqslant 0. \quad (7)$$

The usual assumption that the dissipation rate is independent of $\dot{\theta}$ and $\boldsymbol{\nabla}_0 \dot{\eta}_i$ leads to the constitutive equations for the entropy and generalized thermodynamic forces $\boldsymbol{Q}_i^\eta$:

$$s = -\frac{\partial \bar{\psi}}{\partial \theta}; \quad \boldsymbol{Q}_i^\eta = \rho_0 \frac{\partial \bar{\psi}}{\partial \boldsymbol{\nabla}_0 \eta_i}. \quad (8)$$

The residual dissipative inequality is

$$\rho_0 \mathcal{D} = \boldsymbol{\sigma}_d : \dot{\boldsymbol{\varepsilon}} + \rho_0 X_i \dot{\eta}_i \geqslant 0, \quad (9)$$

where the dissipative stress $\boldsymbol{\sigma}_d$ is equal to the parenthesis in front of $\dot{\boldsymbol{\varepsilon}}$ and the dissipative force $X_i$ is equal to the parenthesis in front of $\dot{\eta}_i$. This results in constitutive equations for the stress tensor and an evolution equation for $\eta_i$

$$\boldsymbol{\sigma} = \rho_0 \frac{\partial \bar{\psi}}{\partial \boldsymbol{\varepsilon}} + \boldsymbol{\sigma}_d; \quad X_i = -\frac{\partial \bar{\psi}}{\partial \eta_i} + \frac{1}{\rho_0} \boldsymbol{\nabla} \cdot \left( \rho_0 \frac{\partial \bar{\psi}}{\partial \boldsymbol{\nabla}\eta_i} \right), \quad (10)$$

assuming that constitutive equations for $\boldsymbol{\sigma}_d$ and $X_i$ are given. For initially homogeneous material, which we will consider below, $\rho_0$ disappears from Eq. (10) for $X_i$. The assumption about thermodynamic independence of processes described by $\dot{\boldsymbol{\varepsilon}}$ and $\dot{\eta}_i$ leads to two separate inequalities, $\boldsymbol{\sigma}_d : \dot{\boldsymbol{\varepsilon}} \geqslant 0$ and $X_i \dot{\eta}_i \geqslant 0$. They can be satisfied if at least $\boldsymbol{\sigma}_d = \boldsymbol{\sigma}_d(\dot{\boldsymbol{\varepsilon}})$ and $X_i = X_i(\dot{\eta}_j)$.

## IV. STRUCTURE OF THE HELMHOLTZ FREE ENERGY AND EXPRESSION FOR STRESSES

### A. Structure of the Helmholtz free energy

The structure of the Helmholtz energy will be used similarly to that in Ref. [33]

$$\bar{\psi}(\boldsymbol{\varepsilon}, \eta_i, \theta, \boldsymbol{\nabla}\eta_i, \boldsymbol{\nabla}_0 \eta_i) = \frac{J_{t\theta}}{\rho_0} \psi^e(\underbrace{\boldsymbol{\varepsilon} - \boldsymbol{\varepsilon}_t(\eta_i) - \boldsymbol{\varepsilon}_\theta(\theta, \eta_i)}_{\boldsymbol{\varepsilon}_e}, \eta_i, \theta)$$
$$+ J \check{\psi}^\theta(\theta, \eta_i) + \tilde{\psi}^\theta(\theta, \eta_i)$$
$$+ J \psi^\nabla(\boldsymbol{\nabla}\eta_k \cdot \boldsymbol{\nabla}\eta_j, \boldsymbol{k}_{0i}); \quad (11)$$
$$J = \det \boldsymbol{F} = 1 + \boldsymbol{I} : \boldsymbol{\varepsilon} = 1 + \varepsilon_0;$$
$$J_{t\theta} = 1 + \varepsilon_{0\theta} + \varepsilon_{0t},$$

but with two distinctions: (i) the expression given by Eq. (4) for the gradient energy is used in order to correctly describe anisotropy of the interface energy; (ii) the elastic energy $\psi^e$ is multiplied by $J_{t\theta}$ in order to obtain elasticity consistent in the limit with large strain formulation, see Ref. [35]. Here, $\breve{\psi}^\theta$ is the thermal (chemical) energy localized at the interfaces, which is equal to zero in the bulk and $\tilde{\psi}^\theta$ is the thermal energy, which is related to the difference between the thermal parts of the free energies of two phases contacting across an interface; $\varepsilon_0$ is the volumetric strain and $\varepsilon_{0\theta}$ and $\varepsilon_{0t}$ are its thermal and transformational parts. The Jacobian $J$ serves the same purpose as in Refs. [33,35]: it produces (with the correct choice of $\breve{\psi}^\theta$) the desired contribution to the spherical part of the structural stresses.

While for small strains $J \simeq J_{t\theta} \simeq 1$ and $\boldsymbol{\nabla}\eta_k \simeq \boldsymbol{\nabla}_0\eta_k$, these geometric nonlinearities must be retained even for infinitesimal strains in order to receive the proper expression for the interface stresses [33]. For example, even for negligible strains, according to Eq. (11) $dJ/d\boldsymbol{\varepsilon} = \boldsymbol{I}$, which will make the proper contribution to the interface stresses. If one would neglect small terms from the beginning rather than in the final result and use $J \simeq 1$, then $dJ/d\boldsymbol{\varepsilon} = \boldsymbol{0}$, and interface stresses $\boldsymbol{\sigma}_{st} = 0$. To evaluate stresses according to the first Eq. (10), we will use that $dJ/d\boldsymbol{\varepsilon} = \boldsymbol{I}$, $dJ_{t\theta}/d\boldsymbol{\varepsilon} = \boldsymbol{0}$ (since derivative is evaluated at fixed $\theta$ and $\eta_i$), and

$$\frac{\partial \psi^e}{\partial \boldsymbol{\varepsilon}} = \frac{\partial \psi^e}{\partial \boldsymbol{\varepsilon}_e} : \frac{\partial \boldsymbol{\varepsilon}_e}{\partial \boldsymbol{\varepsilon}} = \frac{\partial \psi^e}{\partial \boldsymbol{\varepsilon}_e}. \tag{12}$$

Let us designate $\boldsymbol{\zeta}_i = \boldsymbol{\nabla}\eta_i$ and $\boldsymbol{\zeta}_{0i} = \boldsymbol{\nabla}_0\eta_i$, and $a_{jk} = \boldsymbol{\zeta}_j \cdot \boldsymbol{\zeta}_k = a_{kj}$ for all $k$ and $j$. Then since $\boldsymbol{\zeta}_k \cdot \boldsymbol{\zeta}_j = \boldsymbol{\zeta}_{0k} \cdot \boldsymbol{F}^{-1} \cdot \boldsymbol{F}^{-1t} \cdot \boldsymbol{\zeta}_{0i}$ and $\boldsymbol{F}^{-1} \cdot \boldsymbol{F}^{-1t} = (\boldsymbol{I} - \boldsymbol{\varepsilon} - \boldsymbol{\omega}) \cdot (\boldsymbol{I} - \boldsymbol{\varepsilon} + \boldsymbol{\omega}) \simeq \boldsymbol{I} - 2\boldsymbol{\varepsilon}$, where all products of small tensors are neglected, then $a_{jk} = \boldsymbol{\zeta}_{0j} \cdot (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_{0k} = (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) : \boldsymbol{\zeta}_{0j} \otimes \boldsymbol{\zeta}_{0k} = (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) : \boldsymbol{\zeta}_{0k} \otimes \boldsymbol{\zeta}_{0j} = 0.5(\boldsymbol{I} - 2\boldsymbol{\varepsilon}) : (\boldsymbol{\zeta}_{0j} \otimes \boldsymbol{\zeta}_{0k} + \boldsymbol{\zeta}_{0k} \otimes \boldsymbol{\zeta}_{0j})$. Then

$$\frac{\partial \psi^\nabla}{\partial \boldsymbol{\varepsilon}} = \frac{\partial \psi^\nabla}{\partial a_{jk}} \frac{\partial a_{jk}}{\partial \boldsymbol{\varepsilon}} = -\frac{\partial \psi^\nabla}{\partial a_{jk}}(\boldsymbol{\zeta}_{0j} \otimes \boldsymbol{\zeta}_{0k} + \boldsymbol{\zeta}_{0k} \otimes \boldsymbol{\zeta}_{0j})$$

$$= -\frac{\partial \psi^\nabla}{\partial a_{jk}}(\boldsymbol{\zeta}_j \otimes \boldsymbol{\zeta}_k + \boldsymbol{\zeta}_k \otimes \boldsymbol{\zeta}_j). \tag{13}$$

The last transition is eligible because at small strains $\boldsymbol{\nabla}\eta_i \simeq \boldsymbol{\nabla}_0\eta_i$ and is performed here to make our results compatible with the large strain theory for the limit case of isotropic interface energy [35]. Collecting all parts, we obtain

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_e + \boldsymbol{\sigma}_{st} + \boldsymbol{\sigma}_d; \quad \boldsymbol{\sigma}_e = \rho_0 \frac{\partial \psi^e}{\partial \boldsymbol{\varepsilon}_e}; \tag{14}$$

$$\boldsymbol{\sigma}_{st} = \rho_0(\breve{\psi}^\theta + \psi^\nabla)\boldsymbol{I} - \rho_0 \frac{\partial \psi^\nabla}{\partial a_{jk}}(\boldsymbol{\zeta}_j \otimes \boldsymbol{\zeta}_k + \boldsymbol{\zeta}_k \otimes \boldsymbol{\zeta}_j). \tag{15}$$

Because of the symmetry of $a_{jk}$, the interface stress $\boldsymbol{\sigma}_{st}$ and consequently the total stress are symmetric, as required. Since normals $\boldsymbol{k}_{0i}$ are independent of strain, anisotropy of the gradient energy does not produce any additional contribution to the interface stresses. However, it will significantly complicate the Ginzburg-Landau equations. It is evident

that $\boldsymbol{F}^{-1} \cdot \boldsymbol{F}^{-1t}$ and $\psi^\nabla$ are independent of a rigid-body rotation.

For a single order parameter one has $\psi^\nabla = \psi^\nabla(\boldsymbol{\zeta} \cdot \boldsymbol{\zeta}, \boldsymbol{k}_0)$. We will use below the most popular expression

$$\rho_0 \psi^\nabla = 0.5\beta^2(\boldsymbol{k}_0)|\boldsymbol{\nabla}\eta|^2 \tag{16}$$

and obtain from Eq. (15)

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

$$= \beta^2(\boldsymbol{k}_0)|\boldsymbol{\nabla}\eta|^2(\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k}) + (\rho_0 \breve{\psi}^\theta - 0.5\beta^2(\boldsymbol{k}_0)|\boldsymbol{\nabla}\eta|^2)\boldsymbol{I}. \tag{17}$$

The only difference between Eq. (17) and its isotropic counterpart is the dependence of the gradient coefficient $\beta^2$ on the interface orientation $\boldsymbol{k}_0$ in the undeformed state. That is why we will pursue exactly the same approach as in Refs. [33,35] to prove that one can choose function $\breve{\psi}^\theta$ in a way that for the nonequilibrium interface the last term in Eq. (17) disappears and interface stresses represent the biaxial tension [Fig. 1(a)] with the resultant force equal to the interface energy.

## V. GINZBURG-LANDAU EQUATION

### A. General expression

The usual linear relationships $\dot{\eta}_i = L_{ij} X_j$ with positive definite kinetic coefficients $L_{ij}$, for which the Onsager reciprocal relationships $L_{ij} = L_{ji}$ are met, and together with the second Eq. (10) lead to the generalized Ginzburg-Landau equation

$$\dot{\eta}_j = L_{ji}\left(-\frac{\partial \bar{\psi}}{\partial \eta_i} + \boldsymbol{\nabla}_0 \cdot \frac{\partial \bar{\psi}}{\partial \boldsymbol{\nabla}_0 \eta_i}\right). \tag{18}$$

The local term in the driving force in Eq. (18) was evaluated in Ref. [35]:

$$-\frac{\partial \bar{\psi}}{\partial \eta_i} = \frac{\boldsymbol{\sigma}_e}{\rho_0} : \frac{\partial \boldsymbol{\varepsilon}_\theta(\theta,\eta_k)}{\partial \eta_i} + \frac{\boldsymbol{\sigma}_e}{\rho_0} : \frac{\partial \boldsymbol{\varepsilon}_t(\eta_k)}{\partial \eta_i} - \frac{1}{\rho_0}\frac{\partial \psi^e}{\partial \eta_i}\bigg|_{\boldsymbol{\varepsilon}_e}$$

$$- \frac{\psi^e}{\rho_0}\left(\frac{\partial \boldsymbol{\varepsilon}_{t0}}{\partial \eta_i} + \frac{\partial \boldsymbol{\varepsilon}_{\theta 0}}{\partial \eta_i}\right) - \frac{\partial \breve{\psi}^\theta}{\partial \eta_i} - \frac{\partial \tilde{\psi}^\theta}{\partial \eta_i}. \tag{19}$$

Allowing for the Jacobian $J_{t\theta}$ in front of the elastic energy in Eq. (11) resulted in an additional term in Eq. (19), which was absent in the small strain formulation in Ref. [33]. The interface stresses do not explicitly appear in the Ginzburg-Landau equation. However, they affect the distribution of the elastic stresses and indirectly contribute to Eq. (19).

The most commonly used boundary condition for the order parameter is [33,35]

$$\boldsymbol{n} \cdot \boldsymbol{Q}_i^\eta = \boldsymbol{n} \cdot \rho_0 \frac{\partial \psi}{\partial \boldsymbol{\nabla}\eta_i} = H_i, \tag{20}$$

where $H_i$ are given functions, in particular, those related to change in the surface energy during PT, and $\boldsymbol{n}$ is the unit normal to the external surface. Alternatively, one can use periodic boundary conditions, or a prescribed value of $\eta_i$, or consider a finite-width external surface [55,56], which results in a number of interesting scale and mechanics effects.

### B. Previous theory

Below we will consider a single order parameter for simplicity. The most common choice for the gradient en-

ergy (e.g., in Refs. [38,39]) is $\rho_0 \psi^\nabla = 0.5\beta^2(\boldsymbol{k})|\boldsymbol{\nabla}\eta|^2 = 0.5\beta^2(\boldsymbol{\nabla}\eta) = 0.5\beta^2(\boldsymbol{\zeta})$, where the homogeneous degree one function $\beta(\boldsymbol{\nabla}\eta) = |\boldsymbol{\nabla}\eta|\beta(\boldsymbol{k})$. Then

$$\rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\nabla}\eta} = \beta(\boldsymbol{\nabla}\eta)\frac{\partial \beta(\boldsymbol{\nabla}\eta)}{\partial \boldsymbol{\nabla}\eta} = \beta(\boldsymbol{\zeta})\frac{\partial \beta(\boldsymbol{\zeta})}{\partial \boldsymbol{\zeta}} \tag{21}$$

and the analog of the Ginzburg-Landau equation (18) in the deformed state is

$$\dot{\eta} = L\left[ -\frac{\partial \bar{\psi}}{\partial \eta} + \boldsymbol{\nabla} \cdot \left( \frac{\partial \bar{\psi}}{\partial \boldsymbol{\nabla}\eta} \right) \right] = L\left[ -\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho}\left( \frac{\partial \beta}{\partial \zeta_k}\frac{\partial \beta}{\partial \zeta_i} + \beta\frac{\partial^2 \beta}{\partial \zeta_k \partial \zeta_i} \right)\frac{\partial^2 \eta}{\partial r_k \partial r_i} \right]$$

$$= L\left[ -\frac{\partial \bar{\psi}}{\partial \eta} + \frac{1}{\rho}\left( \frac{\partial \beta}{\partial \boldsymbol{\zeta}} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}} + \beta\frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}\partial \boldsymbol{\zeta}} \right) : \frac{\partial^2 \eta}{\partial \boldsymbol{r}\partial \boldsymbol{r}} \right]. \tag{22}$$

### C. Current theory

Using the function $\beta(\boldsymbol{\nabla}_0\eta) = |\boldsymbol{\nabla}_0\eta|\beta(\boldsymbol{k}_0)$, we present the gradient energy (16) in the form

$$\rho_0 \psi^\nabla = 0.5(\beta(\boldsymbol{k}_0)|\boldsymbol{\zeta}|)^2 = 0.5\left( \beta(\boldsymbol{\zeta}_0)\frac{|\boldsymbol{\zeta}|}{|\boldsymbol{\zeta}_0|} \right)^2$$

$$= 0.5\beta^2(\boldsymbol{\zeta}_0)\frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{F}^{-1} \cdot \boldsymbol{F}^{-1T} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} = 0.5\beta^2(\boldsymbol{\zeta}_0)\frac{\boldsymbol{\zeta}_0 \cdot (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0}. \tag{23}$$

Then we evaluate

$$\boldsymbol{Q}^\eta = \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\nabla}_0\eta} = \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_0} = \beta(\boldsymbol{\zeta}_0)\frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} - 2\beta(\boldsymbol{\zeta}_0)\frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0}\frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} + 2\beta^2(\boldsymbol{\zeta}_0)\frac{\boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} \cdot \left( \boldsymbol{I}\frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} - \boldsymbol{\varepsilon} \right), \tag{24}$$

or in the component form in the Cartesian coordinate system:

$$Q_i^\eta = \rho_0 \frac{\partial \psi^\nabla}{\partial(\partial \eta/\partial r_{0i})} = \rho_0 \frac{\partial \psi^\nabla}{\partial \zeta_{0i}} = \beta(\boldsymbol{\zeta}_0)\frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \zeta_{0i}} - 2\beta(\boldsymbol{\zeta}_0)\frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \zeta_{0i}}\frac{\varepsilon_{bc}\zeta_{0b}\zeta_{0c}}{\zeta_{0a}\zeta_{0a}} - 2\beta^2(\boldsymbol{\zeta}_0)\frac{\zeta_{0b}}{\zeta_{0a}\zeta_{0a}}\left[ \varepsilon_{ib} - \delta_{ib}\frac{\varepsilon_{sc}\zeta_{0s}\zeta_{0c}}{\zeta_{0a}\zeta_{0a}} \right]. \tag{25}$$

These expressions for $\boldsymbol{Q}^\eta$ should be used in the boundary conditions (20). The explicit expression for the Ginzburg-Landau equation can be presented in the component form

$$\frac{\rho_0 \dot{\eta}}{L} = -\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} + \left( \frac{\partial \beta}{\partial \zeta_{0k}}\frac{\partial \beta}{\partial \zeta_{0i}} + \beta\frac{\partial^2 \beta}{\partial \zeta_{0k}\partial \zeta_{0i}} \right)\frac{\partial^2 \eta}{\partial r_{0k}\partial r_{0i}}\frac{(\delta_{bc} - 2\varepsilon_{bc})\zeta_{0b}\zeta_{0c}}{\zeta_{0a}\zeta_{0a}}$$

$$+ 2\beta\frac{\partial \beta}{\partial \zeta_{0i}}\left( -\frac{\frac{\partial \varepsilon_{bc}}{\partial r_{0i}}\zeta_{0b}\zeta_{0c} + 4\varepsilon_{bc}\frac{\partial^2 \eta}{\partial r_{0b}\partial r_{0i}}\zeta_{0c}}{\zeta_{0a}\zeta_{0a}} + \frac{4\varepsilon_{bc}\zeta_{0b}\zeta_{0c}\zeta_{0a}\frac{\partial^2 \eta}{\partial r_{0a}\partial r_{0i}}}{(\zeta_{0a}\zeta_{0a})^2} \right) - 2\beta^2\left( \frac{\frac{\partial \varepsilon_{ib}}{\partial r_{0i}}\zeta_{0b} + \varepsilon_{ib}\frac{\partial^2 \eta}{\partial r_{0b}\partial r_{0i}}}{\zeta_{0a}\zeta_{0a}} - \frac{4\varepsilon_{ib}\zeta_{0b}\zeta_{0a}\frac{\partial^2 \eta}{\partial r_{0a}\partial r_{0i}}}{(\zeta_{0a}\zeta_{0a})^2} \right)$$

$$+ 2\beta^2\left( \frac{\frac{\partial \varepsilon_{bc}}{\partial r_{0i}}\zeta_{0i}\zeta_{0b}\zeta_{0c} + \frac{\partial^2 \eta}{\partial r_{0i}\partial r_{0i}}\varepsilon_{bc}\zeta_{0b}\zeta_{0c}}{(\zeta_{0a}\zeta_{0a})^2} - 4\frac{\zeta_{0i}\varepsilon_{bc}\zeta_{0b}\zeta_{0c}\zeta_{0a}\frac{\partial^2 \eta}{\partial r_{0a}\partial r_{0i}}}{(\zeta_{0a}\zeta_{0a})^3} \right), \tag{26}$$

or in direct tensor notations

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

$$+ 2\beta\frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \cdot \left( -\frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \boldsymbol{r}_0} + 4\frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0} \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \frac{4\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0}}{|\boldsymbol{\zeta}_0|^4} \right)$$

$$- 2\beta^2\left( \frac{\boldsymbol{\zeta}_0 \cdot \frac{\partial \boldsymbol{\varepsilon}}{\partial \boldsymbol{r}_0} : \boldsymbol{I} + \boldsymbol{\varepsilon} : \frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0}}{|\boldsymbol{\zeta}_0|^2} - \frac{4(\boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0) \cdot (\boldsymbol{\zeta}_0 \cdot \frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0})}{|\boldsymbol{\zeta}_0|^4} \right)$$

$$+ 2\beta^2\left( \frac{\boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial \boldsymbol{\varepsilon}}{\partial \boldsymbol{r}_0} \cdot \boldsymbol{\zeta}_0 + \nabla_0^2 \eta \boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^4} - 4\frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0 \boldsymbol{\zeta}_0 \otimes \boldsymbol{\zeta}_0 : \frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0}}{|\zeta_0|^6} \right). \tag{27}$$

In these equations $\beta$ is considered as a homogeneous degree one function of $\boldsymbol{\zeta}_0$ rather than of unit normal $\boldsymbol{k}_0$. One can also neglect $2\boldsymbol{\varepsilon}$ in comparison with $\boldsymbol{I}$.

For a linear elastic material we set

$$\rho_0 \psi^e = 0.5\boldsymbol{\varepsilon}_e{:}\boldsymbol{C}(\eta){:}\boldsymbol{\varepsilon}_e; \quad \boldsymbol{\sigma}_e = \boldsymbol{C}(\eta){:}\boldsymbol{\varepsilon}_e, \qquad (28)$$

i.e., Hooke's law, where $\boldsymbol{C}$ is the fourth-rank tensor of elastic moduli, which is different in different phases. We substitute Hooke's law in Eqs. (19) and (27) and will evaluate the degree of smallness of each strain-related term in the right-hand side of the Ginzburg-Landau equation. We will do this under the assumption that usually elastic and thermal strains are at least an order of magnitude lower than the transformation and total strains, i.e., $\boldsymbol{\varepsilon}_e \sim \boldsymbol{\varepsilon}_\theta \sim \boldsymbol{\varepsilon}_t^2 \sim \boldsymbol{\varepsilon}^2$. In this case we find that (i) the terms due to anisotropy of the interface energy are proportional to $\boldsymbol{\varepsilon}$, i.e. they are of the first order of smallness for a small magnitude of $\boldsymbol{\varepsilon}$; (ii) the work of stresses on the change in the transformation strain is proportional to $\boldsymbol{\varepsilon}^3$; (iii) the stress work on the thermal strain and the term related to change in elastic moduli are proportional to $\boldsymbol{\varepsilon}^4$; and (iv) the term proportional to the elastic energy $\psi^e$ is proportional to $\boldsymbol{\varepsilon}_t^5$ and $\boldsymbol{\varepsilon}_\theta^6$.

Thus, unexpectedly, the first-order correction in the Ginzburg-Landau equation due to infinitesimal strains is related to our correction due to the anisotropic gradient energy rather than due to traditional transformation work. Such an analysis is strict for infinitesimal $\boldsymbol{\varepsilon}$. For small but finite $\boldsymbol{\varepsilon}$ the magnitude of each of this term depends on the corresponding coefficients, which may alter our conclusions. If one neglects all the terms with strains that appeared due to anisotropy of the gradient energy, Eqs. (26) and (27) reduce to Eq. (22).

## VI. COMPLETE SYSTEM OF EQUATIONS FOR A SINGLE ORDER PARAMETER

Below we collect the complete system of equations for a single order parameter and the anisotropic interface energy. Some functions and equations (e.g., $\check{\psi}^\theta$ and $\tilde{\psi}^\theta$) are taken from Ref. [33]. It will be shown in the next section that they are consistent with the analytical solution for a propagating interface and biaxial interface stresses, as in Ref. [33] for isotropic interface energy.

### A. Kinematics

#### 1. Decomposition of the strain tensor $\varepsilon$; volumetric strains

$$\boldsymbol{\varepsilon} = (\boldsymbol{\nabla}_0\boldsymbol{u})_s; \quad \boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_e + \boldsymbol{\varepsilon}_t(\eta) + \boldsymbol{\varepsilon}_\theta(\theta,\eta);$$

$$J = det\boldsymbol{F} = \frac{\rho_0}{\rho} = 1 + \boldsymbol{I}{:}\boldsymbol{\varepsilon} = 1 + \varepsilon_0;$$

$$J_{t\theta} = 1 + \varepsilon_{0\theta} + \varepsilon_{0t}. \qquad (29)$$

#### 2. Transformation $\varepsilon_t$ and thermal $\varepsilon_\theta$ strains

$$\boldsymbol{\varepsilon}_t = \bar{\boldsymbol{\varepsilon}}_t\varphi(a,\eta); \qquad \boldsymbol{\varepsilon}_\theta = \boldsymbol{\varepsilon}_{\theta H} + (\boldsymbol{\varepsilon}_{\theta L} - \boldsymbol{\varepsilon}_{\theta H})\varphi(a_\theta,\eta);$$

$$\varphi(a,\eta) = a\eta^2(1-\eta)^2 + (4\eta^3 - 3\eta^4). \qquad (30)$$

### B. Helmholtz free energy per unit mass and its contributions

$$\bar{\psi}(\boldsymbol{\varepsilon},\eta,\theta,\boldsymbol{\zeta},\boldsymbol{\zeta}_0) = \psi^e + J\check{\psi}^\theta + \tilde{\psi}^\theta + J\psi^\nabla; \quad \boldsymbol{\zeta}_0 = \boldsymbol{\nabla}_0\eta;$$

$$\boldsymbol{\zeta} = \boldsymbol{\nabla}\eta; \qquad (31)$$

$$\check{\psi}^\theta = (A(\theta) - 3\Delta G^\theta(\theta))\eta^2(1-\eta)^2;$$

$$\tilde{\psi}^\theta = \Delta G^\theta(\theta)\eta^2(3 - 2\eta);$$

$$\psi^e = \frac{1}{2\rho_0}\boldsymbol{\varepsilon}_e{:}\boldsymbol{C}(\eta){:}\boldsymbol{\varepsilon}_e; \qquad (32)$$

$$\boldsymbol{C}(\eta) = \boldsymbol{C}_H + (\boldsymbol{C}_L - \boldsymbol{C}_H)\varphi(a_C,\eta).$$

$$\rho_0\psi^\nabla = 0.5(\beta(\boldsymbol{k}_0)|\boldsymbol{\zeta}|)^2 = 0.5\left(\beta(\boldsymbol{\zeta}_0)\frac{|\boldsymbol{\zeta}|}{|\boldsymbol{\zeta}_0|}\right)^2$$

$$= 0.5\beta^2(\boldsymbol{\zeta}_0)\frac{\boldsymbol{\zeta}_0 \cdot (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0}. \qquad (33)$$

### C. Stress tensor

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_e + \boldsymbol{\sigma}_{st} + \boldsymbol{\sigma}_d; \qquad (34)$$

$$\boldsymbol{\sigma}_e = \rho_0\frac{\partial\psi^e}{\partial\boldsymbol{\varepsilon}_e} = \boldsymbol{C}(\eta){:}\boldsymbol{\varepsilon}_e; \quad \boldsymbol{\sigma}_d = \boldsymbol{B}{:}\dot{\boldsymbol{\varepsilon}};$$

$$\boldsymbol{\sigma}_{st} = \beta^2(\boldsymbol{k}_0)|\boldsymbol{\nabla}\eta|^2(\boldsymbol{I} - \boldsymbol{k}\otimes\boldsymbol{k}) + (\rho_0\check{\psi}^\theta - 0.5\beta^2(\boldsymbol{k}_0)|\boldsymbol{\nabla}\eta|^2)\boldsymbol{I}. \qquad (35)$$

### D. Ginzburg-Landau equation

#### 1. Coupled with mechanics

$$\frac{\rho_0\dot{\eta}}{L(\boldsymbol{k}_0)} = -\rho_0\frac{\partial\bar{\psi}}{\partial\eta} + \left(\frac{\partial\beta}{\partial\boldsymbol{\zeta}_0}\otimes\frac{\partial\beta}{\partial\boldsymbol{\zeta}_0} + \beta\frac{\partial^2\beta}{\partial\boldsymbol{\zeta}_0\partial\boldsymbol{\zeta}_0}\right){:}\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0}\frac{\boldsymbol{\zeta}_0 \cdot (\boldsymbol{I} - 2\boldsymbol{\varepsilon}) \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0}$$

$$+ 2\beta\frac{\partial\beta}{\partial\boldsymbol{\zeta}_0}\cdot\left(-\frac{\boldsymbol{\zeta}_0\otimes\boldsymbol{\zeta}_0{:}\frac{\partial\boldsymbol{\varepsilon}}{\partial\boldsymbol{r}_0} + 4\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0}\cdot\boldsymbol{\varepsilon}\cdot\boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^2} + \frac{4\boldsymbol{\zeta}_0\cdot\boldsymbol{\varepsilon}\cdot\boldsymbol{\zeta}_0\boldsymbol{\zeta}_0\cdot\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0}}{|\boldsymbol{\zeta}_0|^4}\right)$$

$$- 2\beta^2\left(\frac{\boldsymbol{\zeta}_0\cdot\frac{\partial\boldsymbol{\varepsilon}}{\partial\boldsymbol{r}_0}{:}\boldsymbol{I} + \boldsymbol{\varepsilon}{:}\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0}}{|\boldsymbol{\zeta}_0|^2} - \frac{4(\boldsymbol{\varepsilon}\cdot\boldsymbol{\zeta}_0)\cdot(\boldsymbol{\zeta}_0\cdot\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0})}{|\boldsymbol{\zeta}_0|^4}\right)$$

$$+ 2\beta^2\left(\frac{\boldsymbol{\zeta}_0\otimes\boldsymbol{\zeta}_0{:}\frac{\partial\boldsymbol{\varepsilon}}{\partial\boldsymbol{r}_0}\cdot\boldsymbol{\zeta}_0 + \nabla_0^2\eta\boldsymbol{\zeta}_0\cdot\boldsymbol{\varepsilon}\cdot\boldsymbol{\zeta}_0}{|\boldsymbol{\zeta}_0|^4} - 4\frac{\boldsymbol{\zeta}_0\cdot\boldsymbol{\varepsilon}\cdot\boldsymbol{\zeta}_0\boldsymbol{\zeta}_0\otimes\boldsymbol{\zeta}_0{:}\frac{\partial^2\eta}{\partial\boldsymbol{r}_0\partial\boldsymbol{r}_0}}{|\zeta_0|^6}\right). \qquad (36)$$

$$-\rho_0 \frac{\partial \bar{\psi}}{\partial \eta} = \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_\theta(\theta, \eta_k)}{\partial \eta} + \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_t(\eta)}{\partial \eta} - \frac{\partial \psi^e}{\partial \eta}\bigg|_{\boldsymbol{\varepsilon}_e} - \psi^e \left( \frac{\partial \boldsymbol{\varepsilon}_{t0}}{\partial \eta} + \frac{\partial \boldsymbol{\varepsilon}_{\theta 0}}{\partial \eta} \right) - \rho_0 \frac{\partial \check{\psi}^\theta}{\partial \eta} - \rho_0 \frac{\partial \tilde{\psi}^\theta}{\partial \eta}. \tag{37}$$

#### 2. Without strains and elastic stresses

$$\frac{\rho_0 \dot{\eta}}{L(\boldsymbol{k})} = -\rho_0 \frac{\partial (\check{\psi}^\theta + \tilde{\psi}^\theta)}{\partial \eta} + \left( \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} \otimes \frac{\partial \beta}{\partial \boldsymbol{\zeta}_0} + \beta \frac{\partial^2 \beta}{\partial \boldsymbol{\zeta}_0 \partial \boldsymbol{\zeta}_0} \right) : \frac{\partial^2 \eta}{\partial \boldsymbol{r}_0 \partial \boldsymbol{r}_0}. \tag{38}$$

#### 3. For interface propagating along the fixed normal **k**

$$\rho_0 \psi^\nabla = 0.5 \left( \beta(\boldsymbol{k}) \frac{d\eta}{dx} \right)^2; \quad \dot{\eta} = \bar{L}(\boldsymbol{k}) \left( -\rho_0 \frac{\partial(\check{\psi}^\theta + \tilde{\psi}^\theta)}{\partial \eta} + \beta^2(\boldsymbol{k}) \frac{\partial^2 \eta}{\partial x^2} \right); \quad \bar{L} = L/\rho_0. \tag{39}$$

#### E. Gradient energy coefficient for cubic crystals

$$\beta(\boldsymbol{k}_0) = \alpha_0 + \alpha_1 \frac{\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2}{|\boldsymbol{\zeta}_0|^4} + \alpha_2 \frac{\zeta_{0i}^2 \zeta_{0j}^2 \zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^6} + \alpha_3 \frac{\left(\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2\right)^2}{|\boldsymbol{\zeta}_0|^8}; \tag{40}$$

$$\beta(\boldsymbol{\zeta}_0) = \alpha_0 |\boldsymbol{\zeta}_0| + \alpha_1 \frac{\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2}{|\boldsymbol{\zeta}_0|^3} + \alpha_2 \frac{\zeta_{0i}^2 \zeta_{0j}^2 \zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^5} + \alpha_3 \frac{\left(\zeta_{0i}^2 \zeta_{0j}^2 + \zeta_{0i}^2 \zeta_{0k}^2 + \zeta_{0k}^2 \zeta_{0j}^2\right)^2}{|\boldsymbol{\zeta}_0|^7}. \tag{41}$$

#### F. Linear momentum balance equation

$$\boldsymbol{\nabla} \cdot \boldsymbol{\sigma} = \rho_0 \dot{\boldsymbol{v}}. \tag{42}$$

#### G. Boundary conditions for the order parameter

$$\boldsymbol{n} \cdot \rho_0 \frac{\partial \psi}{\partial \boldsymbol{\nabla} \eta} = H; \quad \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\nabla}_0 \eta} = \rho_0 \frac{\partial \psi^\nabla}{\partial \boldsymbol{\zeta}_0} = \beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} - 2\beta(\boldsymbol{\zeta}_0) \frac{\partial \beta(\boldsymbol{\zeta}_0)}{\partial \boldsymbol{\zeta}_0} \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} + 2\beta^2(\boldsymbol{\zeta}_0) \frac{\boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} \cdot \left( \boldsymbol{I} \frac{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\varepsilon} \cdot \boldsymbol{\zeta}_0}{\boldsymbol{\zeta}_0 \cdot \boldsymbol{\zeta}_0} - \boldsymbol{\varepsilon} \right).$$

#### H. Expression for entropy

$$s = \frac{1}{\rho_0} \boldsymbol{\sigma}_e : \frac{\partial \boldsymbol{\varepsilon}_\theta}{\partial \theta} - \frac{1}{\rho_0} \frac{\partial \psi^e}{\partial \theta}\bigg|_{\boldsymbol{\varepsilon}_e} - \frac{1}{\rho_0} \psi^e \frac{\partial J_\theta}{\partial \theta} - \frac{\partial \check{\psi}^\theta}{\partial \theta} - \frac{\partial \tilde{\psi}^\theta}{\partial \theta} - \frac{\partial \beta^2(\theta, \boldsymbol{k}_0)}{\partial \theta} \frac{1}{2\rho_0} |\boldsymbol{\nabla} \eta|^2. \tag{43}$$

The function $\beta(\boldsymbol{k}_0)$ in Eq. (40) is obtained based on the proportionality $\beta(\boldsymbol{k}_0) = \gamma(\boldsymbol{k}_0)Z$ [see Eq. (49) below where a factor $Z$ is defined] and the function $\gamma(\boldsymbol{k}_0)$ suggested in Ref. [57]. Specific parameters $\alpha_i$ were calibrated in Ref. [57] for two dozen cubic metals using molecular dynamics simulations. In Eqs. (40) and (41) $i \neq j \neq k$ and there is no summation over these indices. The components of $\boldsymbol{k}_0$ or $\boldsymbol{\zeta}_0$ can be treated as Miller indices of crystallographic planes. Derivatives of $\beta(\boldsymbol{\zeta}_0)$ are given in the Appendix. Equation (39) is written in order to study an interface propagating in an arbitrary chosen direction of the interface normal $\boldsymbol{k}$. The axis $x$ of the Cartesian coordinate system is directed along $\boldsymbol{k}$ and the problem is one dimensional and without mechanics. However, interface stresses [which do not affect Eq. (39)] will be determined. An interface rotation toward the interface energy minimum is forbidden by fixing $\boldsymbol{k}$. This will allow us to find the interface parameters for an arbitrary $\boldsymbol{k}$ and calibrate interface properties by comparing them with corresponding molecular dynamic simulations in Ref. [57]. Equation (43) is obtained similarly to that in Refs. [33,35] but for anisotropic $\beta$. The third term was absent in Ref. [33] but appeared in Ref. [35], where large strain formulation was simplified for small strains.

## VII. PROPAGATING INTERFACE: STRUCTURE, ENERGY, WIDTH, AND STRESSES

Our goal here is to show that all results obtained in Ref. [33] for isotropic interface energy (an analytical solution for the propagating interface and critical nucleus, an expression for the parts $\check{\psi}^\theta$ and $\tilde{\psi}^\theta$ of the free energy that result in biaxial interface tension with the resultant force equal to the nonequilibrium interface energy, as well as expressions for the interface energy and width) can be easily generalized for the anisotropic gradient energy Eq. (16). In particular, functions $\check{\psi}^\theta$ and $\tilde{\psi}^\theta$ remain the same as for isotropic interface energy. That is why we will take them from Ref. [33] and prove that they are correct for anisotropic interface energy rather than derive them as in Ref. [33]. At the same time, our result will generalize some known results [37,38,57]

for the anisotropic equilibrium interface for the anisotropic propagating interface, in particular, they determine orientation dependence of the interface energy and width in terms of $\beta(\boldsymbol{k}_0)$ [due to negligible strains, $\beta(\boldsymbol{k}_0) \simeq \beta(\boldsymbol{k})$]. Obtained equations allow one to calibrate orientation-dependent material functions in our model in terms of measurable (or calculated in atomistic simulations) orientation-dependent interface energy, width, and mobility.

*Analytical solution.* The structure of a propagating plane interface is described by the same analytical solution to Eq. (39) as in Refs. [33,58]:

$$\eta_{\mathrm{in}} = (1 + e^{-\zeta})^{-1}; \quad \zeta = k(\boldsymbol{k})(x - ct) \quad k(\boldsymbol{k}) = \sqrt{2B}/\beta(\boldsymbol{k});$$
$$B := \rho_0(A(\theta) - 3\Delta G^{\theta}(\theta)) \tag{44}$$

but with parameters depending on propagation direction $\boldsymbol{k}$. The interface velocity, $c$, and width, $\delta$, are:

$$c(\boldsymbol{k}) = 6\bar{L}\rho_0 \Delta G^{\theta}(\theta)/k(\boldsymbol{k}); \quad \delta(\boldsymbol{k}) = 10/k(\boldsymbol{k}). \tag{45}$$

An important property of the solution Eq. (44), $d\eta_{\mathrm{in}}/d\zeta = \eta_{\mathrm{in}}(1 - \eta_{\mathrm{in}})$, combined with the definition of $k(\boldsymbol{k})$ in Eq. (44) yields the key relationship for the points of a propagating interface:

$$\psi^{\nabla} = \frac{\beta^2(\boldsymbol{k})}{2\rho_0}|\boldsymbol{\nabla}\eta_{\mathrm{in}}|^2 = \frac{\beta^2(\boldsymbol{k})k^2(\boldsymbol{k})}{2\rho_0}\left(\frac{d\eta_{\mathrm{in}}}{d\zeta}\right)^2$$
$$= (A(\theta) - 3\Delta G^{\theta}(\theta))\eta_{\mathrm{in}}^2(1 - \eta_{\mathrm{in}})^2 = \breve{\psi}^{\theta}, \tag{46}$$

where the definition for $\breve{\psi}^{\theta}$ from Eq. (32) was used. Substitution of this identity in Eq. (35) for the interface stresses eliminates the last term and results in a biaxial tension [Fig. 1(a)]:

$$\boldsymbol{\sigma}_{\mathrm{st}} = \sigma_{\mathrm{st}}(\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k}); \quad \sigma_{\mathrm{st}} = \beta^2(\boldsymbol{k})|\boldsymbol{\nabla}\eta|^2 = 2\rho_0\breve{\psi}^{\theta} \tag{47}$$

with $\sigma_{\mathrm{st}}$ for the magnitude of the interface stress. This confirms correctness of the definition of $\breve{\psi}^{\theta}$ in Eq. (32).

*Nonequilibrium interface energy and width.* By the definition of the interface energy under the nonequilibrium condition (see, e.g., Ref. [19]), it is equal to the excess energy with respect to H in the region with H phase $x \leqslant x_i$ and with respect to L in the region with L phase $x > x_i$:

$$\gamma := \int_{-\infty}^{x_i} \rho_0(\psi - \psi_H)dx + \int_{x_i}^{\infty} \rho_0(\psi - \psi_L)dx. \tag{48}$$

Here $x_i$ is the position of the Gibbs dividing surface (sharp interface), which was determined in Refs. [35,36] using the principle of static equivalence. For the chosen fourth-degree thermodynamic potential, it was determined that the dividing surface corresponds to $\eta = 0.5$, which remains true for anisotropic gradient energy. Repeating the same derivations as in Ref. [33] but for anisotropic interface energy, we

obtain

$$\gamma(\boldsymbol{k},\theta) = \Psi^l + \Psi^{\nabla} = 2\Psi^l = 2\Psi^{\nabla} = \frac{\beta(\boldsymbol{k},\theta)\sqrt{2B}}{6}$$
$$= \frac{k(\boldsymbol{k},\theta)\beta^2(\boldsymbol{k},\theta)}{6} = \frac{B}{3k(\boldsymbol{k},\theta)} = \frac{\rho_0(A - 3\Delta G^{\theta})}{3k(\boldsymbol{k},\theta)}, \tag{49}$$

where $\Psi^l$ and $\Psi^{\nabla}$ are integrals of local and gradient energy, respectively. Thus, similar to the equilibrium interface, for the nonequilibrium interface the total energy is the doubled gradient energy. Since the magnitude of the interface stress is equal to the double gradient energy at each point [Eq. (47)], then the total force is equal to the double total gradient energy, which is $\gamma(\boldsymbol{k},\theta)$. Thus, the resultant force for the interface stresses is equal to the nonequilibrium interface energy, as desired.

For the equilibrium interface, substituting phase equilibrium temperature $\theta_e$, for which $\Delta G^{\theta}(\theta_e) = 0$ in Eq. (49), we simplify

$$\gamma_e(\boldsymbol{k},\theta_e) = \frac{\beta(\boldsymbol{k},\theta_e)\sqrt{2\rho_0 A(\theta_e)}}{6} = \frac{k(\boldsymbol{k},\theta_e)\beta^2(\boldsymbol{k},\theta_e)}{6}$$
$$= \frac{\rho_0 A(\theta_e)}{3k(\boldsymbol{k},\theta_e)}. \tag{50}$$

The nonequilibrium and equilibrium interface width are defined as

$$\delta(\boldsymbol{k},\theta) := \frac{10}{k(\boldsymbol{k},\theta)} = \frac{10\beta(\boldsymbol{k},\theta)}{\sqrt{2B}} = \frac{5}{3}\frac{\beta^2(\boldsymbol{k},\theta)}{\gamma(\boldsymbol{k},\theta)};$$
$$\delta_e(\boldsymbol{k},\theta_e) = \frac{10\beta(\boldsymbol{k},\theta)}{\sqrt{2\rho_0 A(\theta_e)}}. \tag{51}$$

Thus, the orientational dependence of both interface energy and width is proportional to the orientational dependence of the $\beta$, which, for cubic crystals, is given in Eq. (40). This differs from relationships in Ref. [57] for $\gamma_e(\boldsymbol{k})$ (nonequilibrium interfaces have not been considered in Ref. [57]), because it is overlooked that the width of the interface depends on the orientation as well. That is why the ratios

$$\frac{\gamma(\boldsymbol{k},\theta)}{\delta(\boldsymbol{k},\theta)} = \frac{\rho_0(A(\theta) - 3\Delta G^{\theta}(\theta))}{30}; \quad \frac{\gamma(\boldsymbol{k},\theta_e)}{\delta(\boldsymbol{k},\theta_e)} = \frac{\rho_0 A(\theta_e)}{30} \tag{52}$$

are independent of the interface orientation. Also, the temperature dependence of the product

$$\gamma(\boldsymbol{k},\theta)\delta(\boldsymbol{k},\theta) = \tfrac{5}{3}\beta^2(\boldsymbol{k},\theta), \tag{53}$$

is independent of $B(\theta)$.

*Examples for* Na. Function $\gamma(\boldsymbol{k}_0) = \beta(\boldsymbol{k}_0)/Z$ [$Z = 6/\sqrt{2B}$, see Eq. (49)] for Na body-centric cubic crystal in contact with its melt, for which $\alpha_0 = 0.295$, $\alpha_1 = -0.579$, $\alpha_2 = 1.915$, and $\alpha_3 = 0.477$, all in $J/m^2$, are taken from Ref. [57]), is shown in Fig. 2(a). In Fig. 2(b), spherical plots of function $\gamma^2(\boldsymbol{k}_0) = \beta^2(\boldsymbol{k}_0)/Z^2 = 2\psi^{\nabla}/(Z^2|\boldsymbol{\zeta}|^2) = \sigma_{\mathrm{st}}/(Z^2|\boldsymbol{\zeta}|^2)$ are presented. It is clear that the anisotropy of the biaxial tension is much more pronounced than anisotropy of the interface energy and width. Two-dimensional (2D) polar
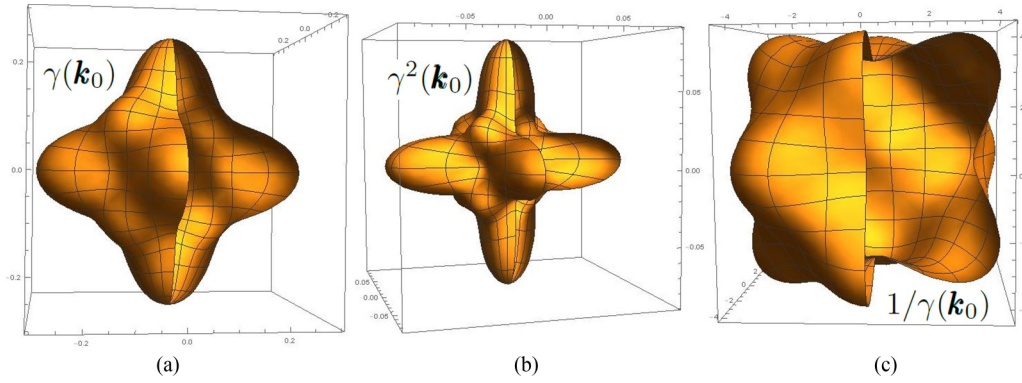
FIG. 2. (Color online) (a) Orientation dependence of the gradient energy coefficient $\gamma(\boldsymbol{k}_0) = \beta(\boldsymbol{k}_0)/Z$ for Na; interface width $\delta(\boldsymbol{k}_0)$ has the same orientation dependence [Eq. (51)]. (b) Spherical plot of the function $\gamma^2(\boldsymbol{k}_0)$, which has same orientation dependence as the magnitude of $\beta^2(\boldsymbol{k}_0)$, the biaxial interface stress $\sigma_{\mathrm{st}}(\boldsymbol{k}_0)$, and the gradient energy $\psi^\nabla(\boldsymbol{k}_0)$. (c) Orientation dependence of the function $1/\gamma(\boldsymbol{k}_0)$, possessing multiple concave parts that should be regularized with the planes.

plots of these functions for {110} and {100} planes are shown in Figs. 3(a), 3(c) and 4(a), 4(b).

## VIII. ARTIFICIAL SHEAR STRESSES AND MOMENTS IN THE PREVIOUS THEORIES

The gradient energy

$$\rho_0 \psi^\nabla = 0.5\beta^2(\boldsymbol{k})|\boldsymbol{\nabla}\eta|^2 = 0.5\beta^2(\boldsymbol{\zeta}) \tag{54}$$

utilized in the previous theories [9,10,13,37–39,43], according to equation

$$\boldsymbol{\sigma}_{\mathrm{st}} = \rho_0(\check{\psi}^\theta + \psi^\nabla)\boldsymbol{I} - \rho_0\boldsymbol{\zeta} \otimes \frac{\partial\psi^\nabla}{\partial\boldsymbol{\zeta}} \tag{55}$$

(see Ref. [35]) leads to the following expression for structural stresses:

$$\boldsymbol{\sigma}_{\mathrm{st}}^p = (\rho_0\check{\psi}^\theta + 0.5\beta^2(\boldsymbol{k})|\boldsymbol{\zeta}|^2)\boldsymbol{I} - \beta(\boldsymbol{\zeta})\boldsymbol{\zeta} \otimes \frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}}$$
$$= \beta^2(\boldsymbol{\zeta})(\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k}) - \boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}}, \tag{56}$$

where superscript $p$ is for previous, and

$$\boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}} := \beta(\boldsymbol{\zeta})\boldsymbol{\zeta} \otimes \frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} - \beta^2(\boldsymbol{k})\boldsymbol{\zeta} \otimes \boldsymbol{\zeta} \tag{57}$$

is the difference between the previous theory and the correct result in Eq. (47), and the equality (46) is taken into account. In the local Cartesian system of coordinates, in which axis 3 is along the normal $\boldsymbol{k}$ and axes 1 and 2 with unit vectors $\boldsymbol{t}$ and $\boldsymbol{p}$ are within an interface [Fig. 1(a)], the interface stress $\boldsymbol{\sigma}_{\mathrm{st}}$ in our theory represents equal biaxial tension in directions 1 and 2. In evaluating $\boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}}$, we recognize that while the term $\beta^2(\boldsymbol{k})\boldsymbol{\zeta} \otimes \boldsymbol{\zeta}$ has the only component 33, the term $\beta(\boldsymbol{\zeta})\boldsymbol{\zeta} \otimes \frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}}$ also possesses shear stresses $\tau_{31}$ and $\tau_{32}$, which are directed along the axis 3 and act at planes orthogonal to axes 1 and 2. Utilizing Eq. (57) and $\frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{\zeta} = \beta(\boldsymbol{\zeta})$, one has

$$\boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}} \cdot \boldsymbol{\zeta} = \beta(\boldsymbol{\zeta})\boldsymbol{\zeta} \frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{\zeta} - \beta^2(\boldsymbol{k})\boldsymbol{\zeta}|\boldsymbol{\zeta}|^2$$
$$= \beta^2(\boldsymbol{\zeta})\boldsymbol{\zeta} = \beta^2(\boldsymbol{\zeta})\boldsymbol{\zeta} = \boldsymbol{0}. \tag{58}$$
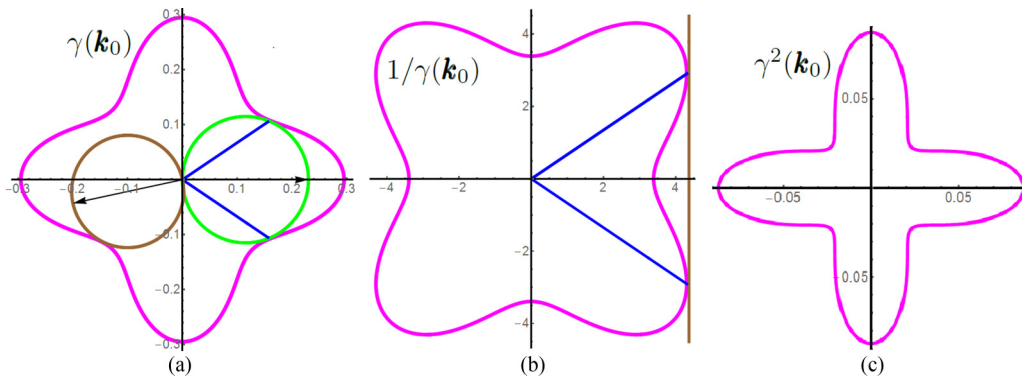


FIG. 3. (Color online) Polar plots for {110} plane of Na for (a) interface energy $\gamma(\boldsymbol{k}_0)$, which is proportional to the function $\beta(\boldsymbol{k}_0)$ and interface width $\delta(\boldsymbol{k}_0)$, (b) function $1/\gamma(\boldsymbol{k}_0)$, and (c) $\gamma^2(\boldsymbol{k}_0) = \beta^2(\boldsymbol{k}_0)/Z^2 = 2\psi^\nabla/(Z^2|\boldsymbol{\zeta}|^2) = \sigma_{\mathrm{st}}/(Z^2|\boldsymbol{\zeta}|^2)$. Convexification of a nonconvex part of function $1/\gamma(\boldsymbol{k}_0)$ with the straight line is shown in (b). It corresponds to the substitution of part of the curve $\gamma(\boldsymbol{k}_0)$ in (a), which cannot be touched by a circle $\boldsymbol{g} \cdot \boldsymbol{k}_0$ plotted on the vector $\boldsymbol{g}$ (which is orthogonal to the regularizing line) as on a diameter, without intersecting $\gamma(\boldsymbol{k}_0)$ at other points, with the circle $\gamma_c(\boldsymbol{k}_0) = \boldsymbol{g} \cdot \boldsymbol{k}_0$.
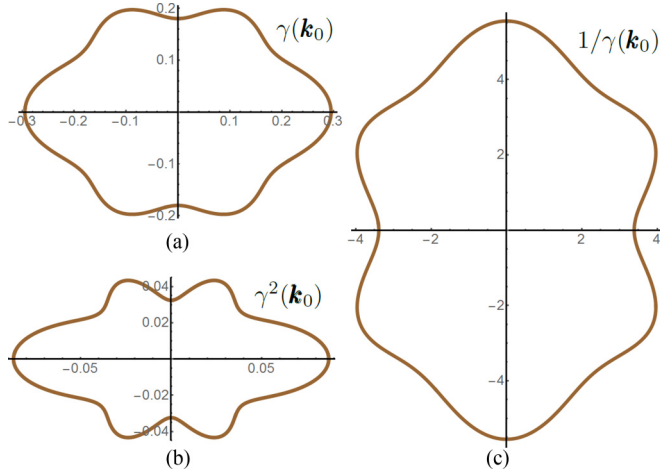
FIG. 4. (Color online) Polar plots for $\{100\}$ plane of Na for interface energy function (a) $\gamma(\boldsymbol{k}_0)$, which is similar to the $\beta(\boldsymbol{k}_0)$ and interface width $\delta(\boldsymbol{k}_0)$, (b) function $\gamma^2(\boldsymbol{k}_0) = \beta^2(\boldsymbol{k}_0)/Z^2 = 2\psi^\nabla/(Z^2|\boldsymbol{\zeta}|^2) = \sigma_{\mathrm{st}}/(Z^2|\boldsymbol{\zeta}|^2)$, and (c) $1/\gamma(\boldsymbol{k}_0)$.

Consequently, component 33 of $\boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}}$ vanishes. Thus, the only nonzero components of $\boldsymbol{\sigma}_{\mathrm{st}}^{\mathrm{dif}}$ are shear stresses:

$$\tau_{31} = \boldsymbol{k} \cdot \beta(\boldsymbol{\zeta})\boldsymbol{\zeta}\,\frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{t} = \beta(\boldsymbol{\zeta})|\boldsymbol{\zeta}|\frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{t};$$
$$\tau_{32} = \boldsymbol{k} \cdot \beta(\boldsymbol{\zeta})\boldsymbol{\zeta}\,\frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{p} = \beta(\boldsymbol{\zeta})|\boldsymbol{\zeta}|\frac{\partial\beta(\boldsymbol{\zeta})}{\partial\boldsymbol{\zeta}} \cdot \boldsymbol{p}, \tag{59}$$

see Fig. 1(b). Since $\tau_{13} = \tau_{23} = 0$, according to the angular momentum equation, shear stresses $\tau_{31}$ and $\tau_{32}$ produce moments about axes 2 and 1, respectively.

The orientation dependence of the normalized biaxial tension for Na is presented in Figs. 2–4. Normalized shear stresses for $\{110\}$ and $\{100\}$ planes are shown in Fig. 5. The maximum magnitude of shear stress reaches about 70% of the maximum $\sigma_{\mathrm{st}}$.

## IX. STRONG ANISOTROPY AND CONVEXIFICATION OF $\beta(\boldsymbol{\zeta})$

Plots of $1/\gamma(\boldsymbol{k}_0) = Z/\beta(\boldsymbol{k}_0)$ are included in Figs. 2–4 because if they are concave, the Ginzburg-Landau equation is ill posed and orientations with high interface energy are not present in the equilibrium microstructure [38,59,60]. For regularization of the problem [38,59,60], the nonconvex regions of $1/\gamma(\boldsymbol{k}_0)$ are substituted with the common tangent

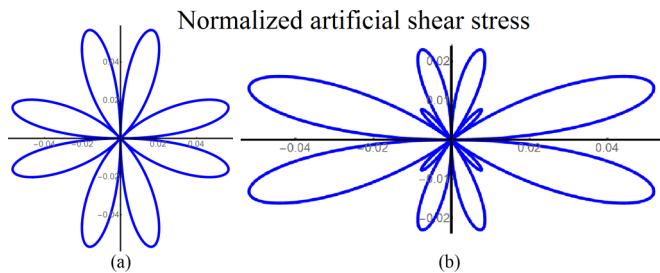### Normalized artificial shear stress



FIG. 5. (Color online) Polar plots of the artificial shear stresses normalized by $Z^2|\boldsymbol{\zeta}|^2$ for Na for (a) $\{100\}$ plane and (b) $\{110\}$ plane.

plane $\frac{1}{\gamma_c(\boldsymbol{k}_0)} = \frac{A}{\boldsymbol{s}\cdot\boldsymbol{k}_0}$, where $\boldsymbol{s}$ is the unit normal to the plane and $A$ is the distance to the plane from the origin [Fig. 3(b)]. The corresponding function $\gamma_c(\boldsymbol{k}_0) = \bar{\boldsymbol{g}} \cdot \boldsymbol{k}_0$ ($\bar{\boldsymbol{g}} := \boldsymbol{s}/A$) represents a sphere plotted on the vector $\bar{\boldsymbol{g}}$ as on the diameter [Fig. 3(a)]. It substitutes $\gamma(\boldsymbol{k}_0)$ for those directions $\boldsymbol{k}_0$, for which it cannot touch $\gamma(\boldsymbol{k}_0)$ without intersecting $\gamma(\boldsymbol{k}_0)$ at other points. All points in convex regions of $1/\gamma(\boldsymbol{k}_0)$ can be touched by a sphere $\boldsymbol{B} \cdot \boldsymbol{k}_0$ for some vector-diameter $\boldsymbol{B}$ [Fig. 3(a)]. For such a $\gamma_c(\boldsymbol{k}_0) = \bar{\boldsymbol{g}} \cdot \boldsymbol{k}_0$ and corresponding $\beta_c(\boldsymbol{k}_0) = \boldsymbol{g} \cdot \boldsymbol{k}_0$ with $\boldsymbol{g} = Z\bar{\boldsymbol{g}}$, all equations become simpler:

$$\beta_c(\boldsymbol{\zeta}_0) = \boldsymbol{g} \cdot \boldsymbol{\zeta}_0 = |\boldsymbol{g}||\boldsymbol{\zeta}_0|\cos\vartheta;\;\frac{\partial\beta_c}{\partial\boldsymbol{\zeta}_0} = \boldsymbol{g};\quad \frac{\partial^2\beta_c}{\partial\boldsymbol{\zeta}_0\partial\boldsymbol{\zeta}_0} = \boldsymbol{0}. \tag{60}$$

Here $\vartheta$ is the angle between the vectors $\boldsymbol{g}$ and $\boldsymbol{\zeta}_0$. Then for coplanar vectors $\boldsymbol{t}$, $\boldsymbol{g}$, and $\boldsymbol{\zeta}_0$ we obtain

$$\tau_{31} = \beta_c(\boldsymbol{\zeta}_0)|\boldsymbol{\zeta}_0|\boldsymbol{g} \cdot \boldsymbol{t} = |\boldsymbol{g}||\boldsymbol{\zeta}_0|^2\cos\vartheta|\boldsymbol{g}|\cos(\pi/2 + \vartheta)$$
$$= -0.5|\boldsymbol{g}|^2|\boldsymbol{\zeta}_0|^2\sin 2\vartheta; \tag{61}$$

$$\sigma_{\mathrm{st}} = \beta_c(\boldsymbol{\zeta}_0)^2 = |\boldsymbol{g}|^2|\boldsymbol{\zeta}_0|^2\cos^2\vartheta;\quad \frac{|\tau_{31}|}{\sigma_{\mathrm{st}}} = \tan\vartheta.$$

The ratio $|\tau_{31}|/\sigma_{\mathrm{st}}$ is a growing function $\vartheta$. Maximum $\vartheta$ is determined by the points in which the sphere $\gamma_c(\boldsymbol{k}_0) = \bar{\boldsymbol{g}} \cdot \boldsymbol{k}_0 = |\boldsymbol{g}|\cos\vartheta$ and $\gamma(\boldsymbol{k}_0)$ touch. The touching is described by equations $|\bar{\boldsymbol{g}}|\cos\vartheta = \gamma(\boldsymbol{k}_0)$ and $d|\bar{\boldsymbol{g}}|\cos\vartheta = d\gamma(\boldsymbol{k}_0)$. For a 2D case and horizontal $\bar{\boldsymbol{g}}$ [Fig. 3(a)], we obtain $\tan\vartheta = -\frac{1}{\gamma}\frac{d\gamma}{d\vartheta}$. For Na $\vartheta_{\max} = 0.593$ and $\tan\vartheta_{\max} = 0.674$ for $\{100\}$ plane and $\vartheta_{\max} = 0.477$ and $\tan\vartheta_{\max} = 0.517$ for $\{110\}$ plane.

## X. RESULTS FOR A SPECIFIC MODEL

We will perform the same specification as in Ref. [33] but for $\beta(\theta)$ substituted with $\beta^2(\boldsymbol{k},\theta)$ (since here we use the expression for the gradient energy typical for papers on anisotropic interface energy, i.e., with $\beta^2$ instead of $\beta$) and $k$ substituted with $k(\boldsymbol{k},\theta)$. That is why we will focus on the final equations since the derivations are very similar to those in Ref. [33].

*Energy and entropy excess.* It is routinely accepted [33]

$$A = A_0(\theta - \theta_c), \quad A_0 > 0;$$
$$\Delta G^\theta(\theta) = -\Delta s_0(\theta - \theta_e), \quad \Delta s_0 < 0, \tag{62}$$

where $\theta_c$ is the critical temperature at which H loses its thermodynamic stability and $\Delta s_0$ is the jump in entropy between L and H. Below we utilize the dimensionless temperature, $\bar{\theta}$, and other parameters:

$$\bar{\theta} := \frac{\theta - \theta_e}{\theta_e - \theta_c};\quad \varpi := -\frac{3\Delta s_0}{A_0} > 0.5;$$
$$\Upsilon := \bar{\theta}(1 - \varpi) + 1 \geqslant 0;\quad \tilde{A} := 2\rho_0 A_0(\theta_e - \theta_c). \tag{63}$$

The interface energy in Eq. (49) is expressed as

$$\gamma = \frac{\beta(\boldsymbol{k},\theta)\sqrt{2\rho_0 A_0(\theta_e - \theta_c)}}{6}\sqrt{\bar{\theta}(1 - \varpi) + 1}$$
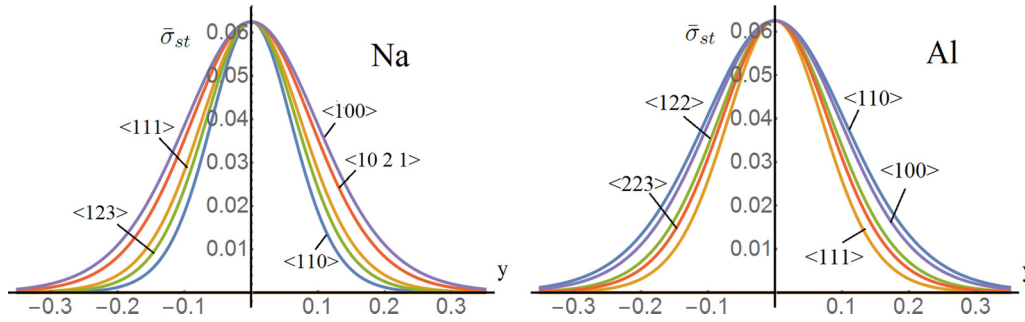$$= \frac{\beta(\boldsymbol{k},\theta)\sqrt{\tilde{A}\Upsilon}}{6};$$

FIG. 6. (Color online) Distribution of the dimensionless biaxial surface tension $\bar{\sigma}_{st}(y)$ for an equilibrium interface for several directions $\boldsymbol{k}$ for bcc Na and fcc Al.

$$\gamma_e = \frac{\beta(\boldsymbol{k},\theta)\sqrt{\tilde{A}}}{6}. \tag{64}$$

Excess of an interface entropy is evaluated as

$$s_i = -\frac{\partial \gamma}{\partial \theta} = \frac{\beta(\boldsymbol{k},\theta)\sqrt{2\rho_0 A_0}}{12\sqrt{\theta_e - \theta_c}} \frac{\varpi - 1}{\sqrt{\bar{\theta}(1-\varpi)+1}}$$
$$+ \frac{\partial \beta(\boldsymbol{k},\theta)}{\partial \theta} \frac{\sqrt{\tilde{A}}\Upsilon}{6}. \tag{65}$$

*Interface width.* Parameter $k$ can be expressed with the help of Eqs. (44) or (49):

$$k(\boldsymbol{k},\theta) = \frac{\sqrt{\tilde{A}[\bar{\theta}(1-\varpi)+1]}}{\beta(\boldsymbol{k},\theta)} = \frac{6\gamma(\boldsymbol{k},\theta)}{\beta^2(\boldsymbol{k},\theta)}. \tag{66}$$

Then the interface width at temperature $\theta$ and $\theta_e$ is:

$$\delta(\boldsymbol{k},\theta) := \frac{10}{k(\boldsymbol{k},\theta)} = \frac{10\beta(\boldsymbol{k},\theta)}{\sqrt{\tilde{A}[\bar{\theta}(1-\varpi)+1]}} = \frac{5\beta^2(\boldsymbol{k},\theta)}{3\gamma(\boldsymbol{k},\theta)};$$
$$\delta(\theta_e) = 10\frac{\beta(\boldsymbol{k},\theta_e)}{\sqrt{\tilde{A}}}. \tag{67}$$

Let us define $\beta_0(\theta)$ and the interface width $\delta_0(\theta)$ in the direction $\langle 100\rangle$. Below we will use the dimensionless interface width $\tilde{\delta}$ normalized by the interface width $\delta_0(\theta_e)$:

$$\tilde{\delta} := \frac{\delta(\boldsymbol{k},\theta)}{\delta_0(\theta_e)} = \frac{\beta(\boldsymbol{k},\theta)}{\beta_0(\theta)\sqrt{[\bar{\theta}(1-\varpi)+1]}}. \tag{68}$$

*Interface stress.* The magnitude of the biaxial interface stress is:

$$\sigma_{st} = 2\rho_0\breve{\psi}^\theta = 2\rho_0 A_0[(\theta-\theta_e)(1-\varpi)+(\theta_e-\theta_c)]\eta_{in}^2(1-\eta_{in})^2$$
$$= \tilde{A}\Upsilon\eta_{in}^2(1-\eta_{in})^2. \tag{69}$$

Since the interface profile is the same for any $t$, we can consider $t = 0$ without loss of generality. It is convenient to transform

$$e^{-\zeta} = e^{-k(\boldsymbol{k},\theta)x} = e^{-10x/\delta(\boldsymbol{k},\theta)} = e^{-\frac{10x}{\delta_0(\theta_e)}\frac{\delta_0(\theta_e)}{\delta((\boldsymbol{k},\theta))}} = e^{-10y/\tilde{\delta}(\boldsymbol{k},\theta)};$$
$$\tag{70}$$
$$y := \frac{x}{\delta_0(\theta_e)},$$

where the factor of 10 is an approximate width of the interface $\eta_{in}(\zeta)$ and the dimensionless coordinate $y$ is introduced in which the interface width $\simeq 1$ at $\theta = \theta_e$ in $\langle 100\rangle$ direction. Then Eqs. (44) and (69) result in the distribution of the

magnitude of the interface stresses and their dimensionless analog $\bar{\sigma}_{st}$:

$$\sigma_{st} = \tilde{A}\Upsilon \frac{e^{-20y/\tilde{\delta}(\boldsymbol{k},\theta)}}{(1+e^{-10y/\tilde{\delta}(\boldsymbol{k},\theta)})^4}$$
$$\bar{\sigma}_{st} := \frac{\sigma_{st}}{\tilde{A}\Upsilon} = \frac{e^{-20y/\tilde{\delta}(\boldsymbol{k},\theta)}}{(1+e^{-10y/\tilde{\delta}(\boldsymbol{k},\theta)})^4}. \tag{71}$$

The maximum dimensionless surface stress is independent of direction $\boldsymbol{k}$ and is 1/16 at $y = 0$. A plot of $\bar{\sigma}_{st}(y)$ for $\theta = \theta_e$ and several directions $\boldsymbol{k}$ is shown in Fig. 6 for fcc Al and bcc Na. The area below the plots is proportional to the interface energy $\gamma(\boldsymbol{k},\theta_e)$, which has the same $\boldsymbol{k}$ dependence as $\tilde{\delta}$ and $\beta$. Different crystallographic directions for maximum and minimum width of the interface for Al and Na exhibits different types of anisotropy for these crystals.

## XI. INTERFACE STRESSES FOR CRITICAL NUCLEUS

All results for a critical nucleus in Ref. [33] are valid here provided we add the $\boldsymbol{k}$ dependence of $\beta$ and width of the critical nucleus $l$. Thus, the stationary solution of the Ginzburg-Landau equation for the critical nucleus

$$\eta_c = 6[6-P+\sqrt{P^2-3P}\cosh(20\sqrt{\bar{\theta}+1}\,y/\tilde{l}(\boldsymbol{k},\theta))]^{-1};$$
$$P := 4\varpi\bar{\theta}/(\bar{\theta}+1);$$
$$l(\boldsymbol{k},\theta) := 10\sqrt{2}\beta(\boldsymbol{k},\theta)/\sqrt{\tilde{A}}; \quad y := x/l[(1,0,0),\theta];$$
$$\tilde{l}(\boldsymbol{k},\theta) := l(\boldsymbol{k},\theta)/l[(1,0,0),\theta] \tag{72}$$

is expressed in terms of a more convenient dimensionless coordinate $y$. It is plotted in Fig. 7 for Na for $\varpi = 1$, $\bar{\theta} = -0.01$, and three crystallographic directions. For each point of the nucleus

$$\psi^\nabla(\boldsymbol{k},\theta,\eta_c) = \breve{\psi}^\theta(\theta,\eta_c) + \tilde{\psi}^\theta(\theta,\eta_c);$$
$$\psi(\boldsymbol{k},\theta,\eta_c) = \psi^\nabla(\boldsymbol{k},\theta,\eta_c) + \breve{\psi}^\theta(\theta,\eta_c) + \tilde{\psi}^\theta(\theta,\eta_c)$$
$$= 2\psi^\nabla(\boldsymbol{k},\theta,\eta_c) = 2[\breve{\psi}^\theta(\theta,\eta_c) + \tilde{\psi}^\theta(\theta,\eta_c)]. \tag{73}$$

Substituting Eq. (73) into Eq. (35) for the interface stresses, one obtains

$$\boldsymbol{\sigma}_{st} = \rho_0\psi(\boldsymbol{k},\theta,\eta_c)(\boldsymbol{I} - \boldsymbol{k}\otimes\boldsymbol{k}) - \rho_0\tilde{\psi}^\theta(\theta,\eta_c)\boldsymbol{I}. \tag{74}$$

Thus, the magnitude of the tensile biaxial interface stress is equal at each point to the local total free energy per unit volume
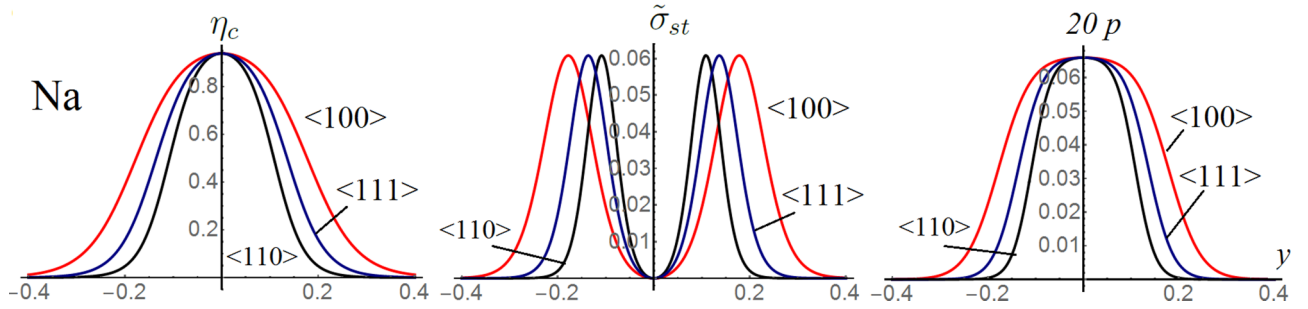
FIG. 7. (Color online) Profile of the critical nucleus $\eta_c(y)$, and distribution of the dimensionless biaxial surface tension $\tilde{\sigma}_{\mathrm{st}}$ and dimensionless mean stress $p$ for three different orientations of Na crystal for $\varpi = 1$ and $\bar{\theta} = -0.01$.

$\rho_0 \psi(\boldsymbol{k}, \theta, \eta_c)$. Consequently, the total interface force is also equal to the total free energy of a critical nucleus. In addition, the tensile mean stress $-\rho_0 \tilde{\psi}^\theta > 0$ is acting at each point of a nucleus. It is also orientation dependent because of the orientation dependence of the solution $\eta_c$.

The dimensionless magnitude of the biaxial surface stress $\tilde{\sigma}_{\mathrm{st}}(y) := \frac{\psi(\theta, \eta_c)}{A_0(\theta_e - \theta_c)}$ and dimensionless mean stress $p(y) := -\frac{\check{\psi}^\theta(\theta, \eta_c)}{A_0(\theta_e - \theta_c)}$ are shown in Fig. 7 for three crystal orientations. One can see that the surface tension is concentrated at the incomplete interfaces and is negligible at the center of a nucleus. Since the driving force for transformation is relatively small, the maximum value of $\eta_c$ is close to unity and the structure of the nucleus is close to two almost complete separated interfaces. That is why the magnitude of a biaxial tension is close to that in Fig. 6 and tensile mean stress is smaller by a factor of 20. The area below the $\tilde{\sigma}_{\mathrm{st}}(x/l)$ curve represents total force and, consequently, the energy of the critical nucleus normalized by $A_0(\theta_e - \theta_c)$. All fields in Fig. 7 depend on orientation through the width of a nucleus and the maximum of all fields is orientation independent.

## XII. CONCLUDING REMARKS

In this work we generalized a PFA developed in Ref. [33] for anisotropic interface energy and stresses. Previous papers on this topic overlooked that the nonsymmetric stress tensor, which is the consequence of anisotropic dependence of the gradient energy on $\boldsymbol{\nabla}\eta_i$, violates the angular momentum balance and principle of material objectivity. In the theory developed here, this problem was overcome by assuming that the gradient energy is an isotropic function of the gradient of the order parameters in the current state, which also depends on the direction of the gradient of the order parameters in the reference state. Thus, some elements of finite strain formulation should be included in the current small strain theory. This leads to a symmetric stress tensor that transforms to the biaxial tension with the magnitude equal to the orientation- and temperature-dependent interface energy for the nonequilibrium interface. The derived Ginzburg-Landau equations have many extra terms due to anisotropy of the interface energy. They are all of the first order of smallness for small strains, which is, surprisingly, a more important strain-related contribution than the next significant term, which comes from mechanics. Indeed, the largest mechanical contribution, which is the transformation work, is cubic in

small strains. The analytical study of the propagating interface and critical nucleus is not much more complicated than for an isotropic interface: one just has to substitute gradient energy and the kinetic coefficient with their orientational dependence. The analytical relationship for such dependence for the gradient energy coefficient is obtained in Ref. [57] using molecular dynamics.

The developed PFA is applicable to melting or solidification [9–14,30,43], sublimation, amorphization, and can be generalized for solid-solid PTs [1–6,8,32,61], twinning [15,16], grain growth [17], fracture [54,62], and interaction of cracks and dislocations with PTs [63–72], for which the interface energy depends on interface orientation of crystals from both its sides. It also has to be generalized for fully large strain formulation [35] and multivariant martensitic transformations and multiphase materials [73].

Similarly, orientation dependence can be introduced in the expression for energy of the external surface for a sharp [14,32,74] and finite-width [55,56] treatment of the external surfaces. It may lead to reshaping and faceting of nanowires [75] and other nanoobjects. It also can be included for melting within grain boundaries [76] and at the interfaces between two solid phases [75,77–81]. Note that reorientation of an interface may occur due to applied stresses and corresponding thermodynamic driving force is found in Refs. [82,83]. In PFA, such a reorientation will occur automatically.

# APPENDIX

Below, the derivatives of all $\beta$-related terms in Eqs. (36) and (26) are presented in the component form:

$$\frac{\partial \beta}{\partial \zeta_{0i}} = \alpha_0 \frac{\zeta_{0i}}{|\boldsymbol{\zeta}_0|} + \alpha_1 \left( 2\frac{\zeta_{0i}(\zeta_{0j}^2 + \zeta_{0k}^2)}{|\boldsymbol{\zeta}_0|^3} - 3\frac{\zeta_{0i}(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^5} \right) + \alpha_2 \left( \frac{2\zeta_{0i}\zeta_{0j}^2\zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^5} - \frac{5\zeta_{0i}^3\zeta_{0j}^2\zeta_{0k}^2}{|\boldsymbol{\zeta}_0|^7} \right)$$

$$+ \alpha_3 \left( \frac{4\zeta_{0i}(\zeta_{0j}^2 + \zeta_{0k}^2)(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^7} - \frac{7\zeta_{0i}(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)^2}{|\boldsymbol{\zeta}_0|^9} \right); \quad \text{(A1)}$$

$$\frac{\partial^2 \beta}{\partial \zeta_{0i} \partial \zeta_{0k}} = -\alpha_0 \frac{\zeta_{0i}\zeta_{0k}}{|\boldsymbol{\zeta}_0|^3} - \alpha_1 \left( \frac{2\zeta_{0i}\zeta_{0k}(\zeta_{0i}^2 + \zeta_{0k}^2 + 4\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^5} - \frac{15\zeta_{0i}\zeta_{0k}(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^7} \right)$$

$$- \alpha_2 \frac{\zeta_{0i}\zeta_{0j}^2\zeta_{0k}(6\zeta_{0i}^4 - 4\zeta_{0j}^4 + 6\zeta_{0k}^4 + 2\zeta_{0j}^2\zeta_{0k}^2 + 2\zeta_{0i}^2\zeta_{0j}^2 - 23\zeta_{0i}^2\zeta_{0k}^2)}{|\boldsymbol{\zeta}_0|^9}$$

$$+ \alpha_3 \left( \frac{8\zeta_{0i}\zeta_{0k}[\zeta_{0j}^4 + 2\zeta_{0j}^2\zeta_{0k}^2 + 2\zeta_{0i}^2(\zeta_{0k}^2 + \zeta_{0j}^2)]}{|\boldsymbol{\zeta}_0|^7} + \frac{63\zeta_{0i}\zeta_{0k}(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)^2}{|\boldsymbol{\zeta}_0|^{11}} \right.$$

$$\left. - \frac{28\zeta_{0i}\zeta_{0k}[\zeta_{0i}^4(\zeta_{0k}^2 + \zeta_{0j}^2) + \zeta_{0k}^2\zeta_{0j}^2(\zeta_{0k}^2 + 2\zeta_{0j}^2) + \zeta_{0i}^2(2\zeta_{0j}^4 + 4\zeta_{0j}^2\zeta_{0k}^2 + \zeta_{0k}^4)]}{|\boldsymbol{\zeta}_0|^9} \right), \quad \text{(A2)}$$

and

$$\frac{\partial^2 \beta}{\partial \zeta_{0i}^2} = \alpha_0 \left( \frac{1}{|\boldsymbol{\zeta}_0|} - \frac{\zeta_{0i}^2}{|\boldsymbol{\zeta}_0|^3} \right) + \alpha_1 \left( \frac{2(\zeta_{0j}^2 + \zeta_{0k}^2)}{|\boldsymbol{\zeta}_0|^3} - \frac{3(3\zeta_{0i}^2\zeta_{0j}^2 + 3\zeta_{0i}^2\zeta_{0k}^2 - \zeta_{0k}^2\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^5} \right.$$

$$\left. + \frac{15\zeta_{0i}^2(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)}{|\boldsymbol{\zeta}_0|^7} \right) + \alpha_2 \frac{\zeta_{0j}^2\zeta_{0k}^2(12\zeta_{0i}^4 - 21\zeta_{0i}^2(\zeta_{0j}^2 + \zeta_{0k}^2) + 2(\zeta_{0j}^2 + \zeta_{0k}^2)^2)}{|\boldsymbol{\zeta}_0|^9}$$

$$+ \alpha_3 \left( \frac{4(\zeta_{0k}^2 + \zeta_{0j}^2)[\zeta_{0j}^2\zeta_{0k}^2 + 3\zeta_{0i}^2(\zeta_{0k}^2 + \zeta_{0j}^2)]}{|\boldsymbol{\zeta}_0|^7} + \frac{63\zeta_{0i}^2(\zeta_{0i}^2\zeta_{0j}^2 + \zeta_{0i}^2\zeta_{0k}^2 + \zeta_{0k}^2\zeta_{0j}^2)^2}{|\boldsymbol{\zeta}_0|^{11}} \right.$$

$$\left. - \frac{7(\zeta_{0j}^4\zeta_{0k}^4 + 10\zeta_{0i}^2\zeta_{0j}^2\zeta_{0k}^2(\zeta_{0k}^2 + \zeta_{0j}^2) + 9\zeta_{0i}^4(\zeta_{0k}^2 + \zeta_{0j}^2)^2)}{|\boldsymbol{\zeta}_0|^9} \right). \quad \text{(A3)}$$

For a two-dimensional case one has to put $\zeta_{0j} = 0$; the terms with $\alpha_2$ in this case disappear.

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