An exact formulation for exponential-logarithmic transformation stretches in a multiphase phase field approach to martensitic transformations

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Abstract
A general theoretical and computational procedure for dealing with an exponential-logarithmic kinematic model for transformation stretch tensor in a multiphase phase field approach to stress- and temperature-induced martensitic transformations with $N$ martensitic variants is developed for transformations between all possible crystal lattices. This kinematic model, where the natural logarithm of transformation stretch tensor is a linear combination of natural logarithm of the Bain tensors, yields isochoric variant–variant transformations for the entire transformation path. Such a condition is plausible and cannot be satisfied by the widely used kinematic model where the transformation stretch tensor is linear in Bain tensors. Earlier general multiphase phase field studies can handle commutative Bain tensors only. In the present treatment, the exact expressions for the first and second derivatives of the transformation stretch tensor with respect to the order parameters are obtained. Using these relations, the transformation work for austenite $\leftrightarrow$ martensite and variant $\leftrightarrow$ variant transformations is analyzed and the thermodynamic instability criteria for all homogeneous phases are expressed explicitly. The finite element procedure with an emphasis on the derivation of the tangent matrix for the phase field equations, which involves second derivatives of the transformation deformation gradients with respect to the order parameters, is developed. Change in anisotropic elastic properties during austenite–martensitic variants and variant–variant transformations is taken into account. The numerical results exhibiting twinned microstructures for cubic to orthorhombic and cubic to monoclinic-I transformations are presented.

Keywords
Martensitic phase transformations, multiphase phase field approach, exponential-logarithmic transformation rule, instability of solids, finite element method, twinning
1. Introduction

Martensitic transformations (MTs) play the central role in determining some extraordinary properties in solids such as pseudoelasticity in shape memory alloys (SMAs) [1], increased yield strength and hardness in ferrous and some other materials [2], etc. In this phenomenon a parent phase, called austenite, transforms into a product phase, called martensite, which usually has multiple crystallographically equivalent variants [1]. The materials during and after MTs are composed of very special and complex microstructures.

Landau introduced a phase field approach where a scalar, called the order parameter (internal variable), was used to describe an order (lower symmetry) ↔ disorder (higher symmetry) transformation [3, 4]. Similar approaches augmented by a gradient-based energy, introduced by Ginzburg to incorporate an interfacial energy, were later used to study phase transformations, microstructure evolution in materials, and several other physical phenomena in nature; see [4] for some examples. Various phase field approaches were developed to study the multivariant stress- and temperature-induced MTs [5–27]. In all phase field methods, the order parameters are considered to describe the phases. Concentration-based order parameters have been used in microscale phase field models [5–9], and transformation strains related order parameters have been used in nanoscale phase field models [10–17]. The evolution of the phases is governed by a system of Ginzburg–Landau equations coupled with the equations of elasticity theory. In concentration-related order parameters based models in [5–9], linear interpolation functions (mixture rule type) are used. On the other hand, the models having order parameters related to the transformation strains use nonlinear interpolation functions. The readers are referred to the authors’ paper [16] for a detailed comparative study of the models used in [5–25]. In [26, 27] a new nonlinear interpolation function was introduced to describe a new instability criteria related to silicon-I ↔ silicon-II phase transformations.

The large-strain formulations for multivariant MTs were developed and applied to solve various problems in [6, 13, 15, 16, 20–22, 24–26]. Various kinematic models (KMs) were used for the transformation deformation gradient $F_t$.

(i) KM-I: $F_t$ or its rate equation is motivated from the twinning equations in crystallographic theory of MTs [16, 20, 25, 28].

(ii) KM-II: $F_t$ is a linear combination of the Bain tensors multiplied with either linear [5, 6, 8, 21]. or nonlinear interpolation functions [10, 12, 15–17, 20, 22, 25–27].

(iii) KM-III: Recently, $F_t$ was considered to be equal to exponential of a linear combination of the natural logarithm of the Bain tensors multiplied with linear [5, 6] or nonlinear [16, 25] interpolation functions.

The KM-I, derived from the twinning equation, is consistent with the crystallographic theory and is volume preserving for the entire variant ↔ variant transformation path for any pair of variants. However, the problem is that not all the variants are in twin relations (see, e.g., [1, Chapter 5]), and hence this transformation rule cannot be generalized for all the MTs. In addition, the twinning equation is not applicable when incomplete martensite evolves with curved interfaces [16, 29]. The KM-II is very simple, easy to handle, and valid for any number of variants. However, it yields a non-isochoric variant ↔ variant transformation path. Notably, the isochoric nature of the variant ↔ variant transformations is considered to be a plausible condition, although the reality is not yet known to the best of the authors’ knowledge. On the other hand, KM-III can be used for any number of variants and it yields isochoric variant ↔ variant transformations along the entire transformation path; see [6, 16] for proofs. Therefore, KM-III can potentially be used for future studies of multiphase field models for MTs and the interaction between MTs and plasticity/fracture. In particular, KM-III was utilized to study microstructure evolutions during MTs with multiple variants [5, 6, 16, 25] and single variant [27].

The present authors developed a thermodynamically consistent multiphase phase field approach for $N$ martensitic variants in [16]. Here $N + 1$ order parameters $\eta_0, \eta_1, \eta_2, \ldots, \eta_N$ were introduced, where $\eta_0$ describes $A \leftrightarrow M$ transformations and $\eta_1, \eta_2, \ldots, \eta_N$ describe $N$ variants $M_1, M_2, \ldots, M_N$ such that

$$\sum_{i=1}^{N} \eta_i = 1,$$ (1)
where $\mathbf{A}$, $\mathbf{M}$, and $\mathbf{M}_i$ denote austenite, martensite, and $i$-th martensitic variant, respectively. We assume $\eta_0 = 0$ in $\mathbf{A}$ and $\eta_0 = 1$ in $\mathbf{M}_i$; $\eta_i = 1$ in $\mathbf{M}_i$ and $\eta_i = 0$ in $\mathbf{M}_j$ for all $j \neq i$. The KM-III used in [16] is

$$F_i = \mathbf{U}_i = \exp \left[ \varphi(a_i, \eta_0) \sum_{i=1}^{N} \phi_i(\eta_i) \ln \mathbf{U}_i \right],$$

where $\mathbf{U}_i$ is the transformation stretch tensor, $\mathbf{U}_i$ is the Bain stretch tensor for variant $\mathbf{M}_i$ for all $i = 1, \ldots, N$, and $\varphi(a_i, \eta_0)$ and $\phi_i(\eta_i)$ are the nonlinear interpolation functions for $\mathbf{A} \leftrightarrow \mathbf{M}$ and $\mathbf{M}_i \leftrightarrow \mathbf{M}_j$ transformations, respectively [16]:

$$\varphi(a_i, \eta_0) = a_i \eta_0^2 + (4 - 2a_i) \eta_0^3 + (a_i - 3) \eta_0^4 \quad \text{and} \quad \phi_i(\eta_i) = \eta_i^3(3 - 2\eta_i).$$

In (3), $a_i$ is a constant parameter such that $0 \leq a_i \leq 6$. The exponential and natural logarithm of an arbitrary second-order tensor $\mathbf{A}$ in a $d$-dimensional vector space are defined as [30, 31]

$$\exp \mathbf{A} = \sum_{k=0}^{\infty} \frac{\mathbf{A}^k}{k!} \quad \text{and} \quad \ln \mathbf{A} = \sum_{k=1}^{\infty} \frac{(-1)^{k+1}(\mathbf{A} - \mathbf{I})^k}{k},$$

respectively. The series in (4)1 is convergent for all $\mathbf{A}$. However, the series in (4)2 is absolutely convergent if and only if $|\mathbf{A} - \mathbf{I}| < 1$, where $|\mathbf{A}|$ denotes the Euclidean norm of $\mathbf{A}$ (see the end of this section for its definition). Notably, for all known MTs the Bain tensors satisfy $|\mathbf{U}_i - \mathbf{I}| < 1$ (see [1]) and, hence, the series in (4)2 is convergent for $\mathbf{A} = \mathbf{U}_i$ for all $i = 1, \ldots, N$. The interpolation functions in (3) satisfy the conditions $\varphi(a_i, 0) = 0$, $\varphi(a_0, 1) = 1$, and $\frac{\partial \varphi(a_i, 0)}{\partial \eta_0} = 0$; $\phi_i(0) = 0$, $\phi_i(1) = 1$, and $\frac{\partial \phi_i(\eta_i)}{\partial \eta_i} = 0$ for all $i = 1, 2, \ldots, N$, which were derived from the thermodynamic equilibrium conditions of all the phases [16]. In [5, 6], the model was used for a two-variant system only and the interpolation functions (i.e., $\varphi(a_i, \eta_0)$ and $\phi_i(\eta_i)$) used therein are linear, i.e., our conditions for zero derivatives for each phase are not satisfied. Thermodynamic equilibrium for all phases, however, is met owing to the imposing constraints of the type $0 \leq \eta_0 \leq 1$ and Eq. (1). Since the model in [5, 6] does not consider thermodynamic instability criteria, the analytical expression for second derivative was not required. Also, the derivatives of $F_i$ with respect to the order parameters appearing in the Ginzburg–Landau equations and the tangent matrix for the Newton’s iterations were treated numerically therein. Thus, the analytical expression for the linearization of the weak form [32] is not available.

On the other hand, the thermodynamically consistent model proposed by the authors in [16, 29] studies the transformation work terms and the thermodynamic instability criteria. Hence, the exact expressions for the first and second derivatives of $F_i$ is necessary. Notably, in [16, 29] the model was studied only for NiAl alloy which undergoes cubic to tetragonal transformations. The Bain tensors are commutative in that case, i.e., $\mathbf{U}_i \cdot \mathbf{U}_j = \mathbf{U}_j \cdot \mathbf{U}_i$ (for all $i, j = 1, 2, 3$ and $i \neq j$). One can verify that if $\mathbf{U}_i$ and $\mathbf{U}_j$ are commutative, $\ln \mathbf{U}_i$ and $\ln \mathbf{U}_j$ are also commutative in the following manner. Using (4)2 we have

$$\ln \mathbf{U}_i \cdot \ln \mathbf{U}_j = \left( \sum_{k=1}^{\infty} \frac{(-1)^{k+1}(\mathbf{U}_i - \mathbf{I})^k}{k} \right) \cdot \left( \sum_{l=1}^{\infty} \frac{(-1)^{l+1}(\mathbf{U}_j - \mathbf{I})^l}{l} \right).$$

Obviously, to show the commutativity of $\ln \mathbf{U}_i$ and $\ln \mathbf{U}_j$ one needs to show that $(\mathbf{U}_i - \mathbf{I})^m \cdot (\mathbf{U}_j - \mathbf{I})^n = (\mathbf{U}_j - \mathbf{I})^n \cdot (\mathbf{U}_i - \mathbf{I})^m$ for all $m, n = 1, 2, 3, \ldots$ when $m = n$ and $m \neq n$, which can be easily done using the method of induction. For commutative Bain tensors, determining the derivatives of $F_i$ with respect to all the order parameters (needed for obtaining transformation work, thermodynamic instability criteria, finite element formulations) is straightforward (see [16]). Note that the Bain tensors are not commutative for all other MTs such as cubic→orthorhombic, cubic→monoclinic, etc. [1]. However, the extension of the procedure shown in [16, 29] for exponential-logarithmic KM (i.e., KM-III) with non-commutative Bain tensors is non-trivial.

To understand the difficulty, let us consider a simple system which is fully martensite with two variants $\mathbf{M}_1$ and $\mathbf{M}_2$. Obviously, there is one independent order parameter for the variants due to the constraint given by (1). We denote it by $\eta_1$. In this case obviously $\varphi(a_1, 1) = 1$ and the functions $\phi_1$ and $\phi_2$ satisfy $\phi_1 + \phi_2 = 1$. $F_i$, given by (2), hence simplifies to [16, 29]

$$F_i = \exp [\ln \mathbf{U}_{12} + \phi_1(\eta_1)(\ln \mathbf{U}_{11} - \ln \mathbf{U}_{12})].$$
Following the approach of [16, 29], we use (4) to expand (5) as

\[ F_I = I + [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})] + \frac{1}{2!} [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})]^2 + \cdots. \]  

(6)

Let us obtain the derivative of, say, the third term in series of (6) with respect to \( \eta_1 \) as follows:

\[
\frac{\partial}{\partial \eta_1} \left[ [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})] \cdot [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})] \right]
\]

\[ = \frac{\partial \phi_1(\eta_1)}{\partial \eta_1} \left[ [\ln U_{11} - \ln U_{12}] \cdot [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})] \right]
\]

\[ + \left[ [\ln U_{12} + \phi_1(\eta_1)(\ln U_{11} - \ln U_{12})] \cdot (\ln U_{11} - \ln U_{12}) \right], \]

(7)

when \( \ln U_{11} \) and \( \ln U_{12} \) are non-commutative. Obtaining the derivatives of the higher-order terms from the series of (6) will be increasingly difficult. Thus, the exact expressions for \( \partial F_I / \partial \eta_1 \) and \( \partial^2 F_I / \partial \eta_1^2 \) using (6) is almost impossible to obtain when \( U_{11} \) and \( U_{12} \) are non-commutative. Notably, one can still compromise with an approximate expression by truncating the series after a finite number of terms. However, as shown through (7), the derivation for \( \partial F_I / \partial \eta_1 \) and \( \partial^2 F_I / \partial \eta_1^2 \) becomes increasingly cumbersome as the number of terms in the approximate expression increases and/or the number of variants increases, implying that the procedure is highly inefficient. However, determination of the exact expressions for the first two derivatives of \( F_I \) with respect to the order parameters \( \eta_i \) is utmost important for studying all the MTs using KM-III given by (2). The objective of this article is to present a general analytical and algorithmic (for finite element) treatment to deal with exponential-logarithmic transformation stretch tensor \( U_i \) for all MTs within a multiphase field approach developed by the authors in [16]. Furthermore, the numerical examples of twinning in CuNiAl and NiTi alloys, where the transformation stretch tensors for the variants are non-diagonal (hence, non-commutative), will be presented.

To this end, we use an alternative method from [30], which utilizes the exact expressions for the following fourth- and sixth-order tensors \( \partial(\exp A) / \partial A \) and \( \partial^2(\exp A) / \partial A^2 \) (where \( A \) is an arbitrary second-order tensor), respectively, to determine the necessary derivatives of \( F_I \) given by (2). Note that the present formulation is exact, elegant, and valid for both commutative and non-commutative Bain stretch tensors. Naturally, MTs between any crystal lattices can, hence, be studied. Using these relations we study the following.

1. **The transformation work in the Ginzburg–Landau equations.** We have proved that KM-III allows us to decouple the transformation work for \( A \leftrightarrow M \) transformations into parts due to volumetric part and deviatoric part of a generalized Cauchy stress. As far as the \( M_i \leftrightarrow M_j \) transformations are concerned, the volumetric work identically vanishes, which is a desired plausible condition.

2. **The thermodynamic instability criteria.** We have derived the thermodynamic instability criteria for all the phases. We have established that in the instability criteria for \( A \leftrightarrow M \) transformations the contribution of the volumetric and deviatoric parts of the generalized Cauchy stress can be decoupled, and in the instability criteria for \( M_i \leftrightarrow M_j \) transformations only the deviatoric part of the generalized Cauchy stress contributes, as desired. The instability criteria for \( A \leftrightarrow M \) transformations turn out to be the same for cubic to tetragonal transformation derived in [16]. However, the criteria for \( M_i \leftrightarrow M_j \) transformations derived here apply for all variant–variant transformations, and hence more general than those derived in [16].

3. **The weak forms of the Ginzburg–Landau equations and their linearizations in the finite element procedure.** We have solved the coupled phase field and elasticity equations using a finite element method, similar to that in [29]. But therein it was restricted to the commutative Bain tensors only. Note that a consistent expression for the tangent matrix is essential for good convergence for the iterative solvers. Here we have derived consistent expressions for the weak forms of all \( N \) independent Ginzburg–Landau equations and their linearizations that involve first and second derivatives of \( F_I \) with respect to the order parameters. Using the present theoretical framework and computational procedure, the twinned microstructures have been studied for CuAlNi (cubic to orthorhombic transformation) SMA and NiTi (cubic to monoclinic-I transformation) SMA in two-dimensional single crystal using a generalized plane strain approach [5, 6]. In both the cases, the Bain tensors are non-commutative [1]. A finite element code has been developed in the open-source FE package deal.ii [33]. The numerical results are compared with the analytical solutions taken from the crystallographic theory of MTs. The sample size effect on microstructures is also studied.
We have organized the article as follows: in Section 2 the essential system of coupled elasticity and phase field equations are enlisted; in Section 3 we derive the analytical expressions for the first and second derivatives of \( F_i \) with respect to all the order parameters; in Section 4 the analytical expressions for the transformation work and the thermodynamic instability criteria for MTs are derived and analyzed; in Section 5 some important relations from the crystallographic theory are discussed; in Section 6 the finite element procedure is established (also in the Appendix) and the numerical results are presented; in Section 7 we conclude the paper.

Notation. We denote the inner product and multiplication between two arbitrary second-order tensors as \( A \cdot B = A_{ij}B_{ij} \), respectively, where repeated indices denote Einstein’s summation, and \( A_{ij} \) and \( B_{ij} \) are the components of the tensors in a right-handed orthonormal Cartesian basis \( \{e_1, e_2, e_3\} \). The Euclidean norm of \( A \) is denoted as \( |A| = \sqrt{A : A^T} \); \( I \) denotes the second-order identity tensor; \( A^{-1}, A^T, \det A, \) and \( \tr A \) denote inversion, transposition, determinant, and trace of \( A \), respectively. The set of symmetric second-order tensors is denoted by \( \text{Sym} \), i.e., \( A = A^T \) if \( A \in \text{Sym} \). Dyadic product between two arbitrary vectors \( a \) and \( b \) is denoted as \( a \otimes b \) such that \( (a \otimes b)_{ij} = a_i b_j \). The reference, stress-free intermediate, and deformed configurations of the body are denoted by \( \Omega_0, \Omega_i \), and \( \Omega \), respectively. The symbols \( \nabla_0 \) and \( \nabla \) represent the gradient operators in \( \Omega_0 \) and \( \Omega \), respectively; \( \nabla_0^2 := \nabla_0 \cdot \nabla_0 \) and \( \nabla^2 := \nabla \cdot \nabla \) are the Laplacian operators in \( \Omega_0 \) and \( \Omega \), respectively. The symbol := stands for equality by definition. Deviatoric part of \( A \) is denoted by \( \text{dev}A \).

2. Governing coupled mechanics and phase field equations

In this section, we summarize the governing coupled elasticity and phase field equations from the recent multiphase phase field approach developed in [16]. We assume a set of \( N + 1 \) order parameters \( \eta^* = \{\eta_0, \eta_1, \ldots, \eta_i, \ldots, \eta_N\} \) with a subset \( \eta^*_M = \{\eta_1, \ldots, \eta_i, \ldots, \eta_N\} \). Since the order parameters \( \eta_1, \ldots, \eta_N \) satisfy the constraint \( \sum_{i=1}^{N} \eta_i = 1 \), there are \( N \) independent order parameters in our model. We denote the set of \( N \) independent order parameters, when \( \eta_N \) is eliminated using the constraint \( \sum_{i=1}^{N} \eta_i = 1 \), by \( \tilde{\eta} = \{\eta_0, \eta_1, \ldots, \eta_i, \ldots, \eta_{N-1}\} \) with a subset \( \tilde{\eta}_M = \{\eta_1, \ldots, \eta_i, \ldots, \eta_{N-1}\} \).

2.1. Kinematics

We consider the following multiplicative decomposition of the total deformation gradient \( F := \nabla_0 r \), decomposed into [23]

\[
F = F_e \cdot F_t = V_e \cdot R \cdot U_t,
\]

where \( r = r_0 + u(r_0,t) \) and \( r_0 \) are the position vectors of a particle in the deformed configuration \( \Omega \) and the reference configuration \( \Omega_0 \), respectively; \( u \) is the displacement vector; \( t \) denotes time; \( F_t \) and \( F_e \) are the transformation and elastic parts of \( F \), respectively; \( V_e, U_t, \) and \( R \) are the left elastic stretch tensor, right transformation stretch tensor, and the lattice rotation tensor, respectively. We define \( J = \det F := dV/dV_0, J_t = \det F_t := dV_t/dV_0, \) and \( J_e = \det F_e := dV_e/dV_0 \), where \( dV, dV_0, \) and \( dV_t \) are infinitesimal volume elements in \( \Omega, \Omega_0, \) and \( \Omega_t \), respectively. It is clear that \( J = J_t J_e \). The Lagrangian elastic and total strain tensors are defined as

\[
E_e := 0.5(C_e - I) \quad \text{and} \quad E := 0.5(C - I),
\]

respectively, where \( C := F^T \cdot F \) and \( C_e := F_e^T \cdot F_e \) are the right Cauchy–Green total strain and elastic strain tensors, respectively.

2.2. Kinematic model (KM-III) for \( U_t \)

We rewrite \( U_t \) given by (2) in a convenient form as (see also [16] for details)

\[
U_t = \exp W, \quad \text{where} \quad W(\eta^*) = \varphi(a, \eta_0)Z(\eta^*_M) \quad \text{and} \quad Z(\eta^*_M) = \sum_{i=1}^{N} \phi_i(\eta_i) \ln U_t.
\]
The expressions in (10) can also be equivalently expressed as functions of \( N \) independent order parameters \( \eta_0, \eta_1, \ldots, \eta_{N-1} \):

\[
\mathbf{U}_i = \exp \mathbf{W}, \quad \text{where } \mathbf{W}(\hat{\eta}) = \varphi(a, \eta_0)\mathbf{Z}(\hat{\eta}_M) \quad \text{and } \mathbf{Z}(\hat{\eta}_M) = \sum_{i=1}^{N-1} \phi_i(\eta_i) \ln \mathbf{U}_i + \phi_N(\hat{\eta}_M) \ln \mathbf{U}_N,
\]

where the function, say \( \mathcal{F}(\eta_0, \eta_1, \ldots, \eta_N) \) (which can be scalar, vector, or tensor) with an over-bar is expressed in terms of the \( N - 1 \) independent order parameters \( \eta_0, \eta_1, \ldots, \eta_{N-1} \) such that \( \mathcal{F}(\eta_0, \eta_1, \ldots, \eta_{N-1}) = \mathcal{F}(\eta_0, \eta_1, \ldots, 1 - \sum_{i=1}^{N-1} \eta_i) \).

### 2.3. Free energy

We assume the Helmholtz free energy per unit mass of the body as [16, 23]:

\[
\psi(F, \theta, \eta_0, \eta_1, \nabla \eta_0, \nabla \eta_1) = \frac{J_i}{\rho_0} \psi^e(F_i, \theta, \eta_0, \eta_1) + \tilde{\psi}^\theta(\theta, \eta_0, \eta_1) + J \tilde{\psi}^\phi(\theta, \eta_0, \eta_1) + \psi^p(\eta_0, \eta_1) + J \psi^\nabla(\eta_0, \nabla \eta_0, \nabla \eta_1)
\]

for all \( i = 1, 2, \ldots, N \),

where \( \psi^e \) is the elastic strain energy per unit volume of \( \Omega_i \), \( \tilde{\psi}^\theta \) is the thermal energy, \( \tilde{\psi}^\phi \) is the barrier energy related to \( A \leftrightarrow M \) and all the \( M_i \leftrightarrow M_j \) transformations, \( \psi^p \) penalizes the deviation of the variant–variant transformation paths from the straight lines, and \( \psi^\nabla \) is the interfacial energy [16]:

\[
\psi^e = \frac{1}{2} E_e : \hat{\mathbf{C}}_e(\eta_0, \eta_1) : E_e, \quad \text{where } \hat{\mathbf{C}}_e(\eta_0, \eta_1) = (1 - \varphi(a, \eta_0)) \hat{\mathbf{C}}_{(e)0} + \varphi(a, \eta_0) \sum_{i=1}^{N} \phi_i(\eta_i) \hat{\mathbf{C}}_{(e)i};
\]

\[
\tilde{\psi}^\theta = \tilde{\psi}^\theta(\theta) + \tilde{\psi}^\theta(3 - 2\eta_0) \Delta \psi^\theta(\theta), \quad \text{where } \Delta \psi^\theta = -\Delta s_{0M}(\theta - \theta_\epsilon); \quad (14)
\]

\[
\tilde{\psi}^\phi = [A_{0M}(\theta) + (a_\theta - 3) \Delta \psi^\phi(\theta)] \eta_0^2 (1 - \eta_0)^2 + \tilde{\phi}(a_\beta, \eta_0) \sum_{i=1}^{N} \sum_{j=i+1}^{N} \eta_i^2 \eta_j^2; \quad (15)
\]

\[
\psi^p = \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} K_{ij}(\eta_i + \eta_j - 1)^2 \eta_i^2 \eta_j^2, \quad \text{where } K_{ii} = 0; \quad (16)
\]

\[
\psi^\nabla = \frac{\beta_{0M}}{2 \rho_0} (\nabla \eta_0)^2 + \tilde{\phi}(\eta_0, a_\beta, a_\alpha) \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \beta_{ij} (|\nabla \eta_i|^2 - 2 \nabla \eta_i \cdot \nabla \eta_j + |\nabla \eta_j|^2), \quad \text{where } \beta_{ij} = 0; \quad (17)
\]

\[
\tilde{\phi}(a_\beta, a_\alpha, \eta_0) = a_\epsilon + a_\beta \eta_0^2 - 2(a_\beta - 2(1 - a_\epsilon)) \eta_0^3 + [a_\beta - 3(1 - a_\epsilon)] \eta_0^4. \quad (18)
\]

Let us define the symbols used in (13)–(18). Here \( \hat{\mathbf{C}}_{e}(\eta_0, \eta_1) \) is the fourth-order elastic moduli tensor at a material point; \( \hat{\mathbf{C}}_{(e)0} \) and \( \hat{\mathbf{C}}_{(e)i} \) are the elastic moduli tensors of \( A \) and \( M_i \), respectively; \( A_{0M} > 0 \) is the energy barrier height between \( A \) and \( M \); \( A > 0 \) is the energy barrier height between \( M_i \) and \( M_j \) for all \( i \neq j \); \( \tilde{\psi}^\theta \) is the thermal energy of \( A \); \( \Delta \psi^\phi = \psi_{\theta}^\phi - \tilde{\psi}_{\theta}^\phi \) is the thermal energy difference between \( A \) and \( M \); \( \Delta s_{0M} = s_M - s_0 \) is the change in entropy owing to \( A \) to \( M \) transformation \( (s_0 \) and \( s_M \) denoting the entropy of \( A \) and \( M \), respectively); \( \theta_\epsilon > 0 \) is the absolute temperature; \( \theta_\epsilon \) is the thermodynamic equilibrium temperature between \( A \) and \( M \); \( K_{ij} \geq 0 \) is a controlling parameter for penalizing the deviation of the transformation \( M_i \leftrightarrow M_j \) from the straight line \( \eta_i + \eta_j = 1 \) for all \( \eta_k = 0 \) and \( k \neq i, j \); \( \beta_{0M} > 0 \) and \( \beta_{ij} > 0 \) are the gradient energy coefficients for \( A \rightarrow M \) and \( M_i \rightarrow M_j \) interfaces, respectively; \( \rho_0 \) is the mass density of the solid in \( \Omega_0 \); and \( a_\alpha, a_\beta, a_\epsilon, a_\epsilon \) are the material parameters. As the elastic strains are small, the quadratic strain energy given by (13) can be accepted to be convex in \( E_e \). Note that here we did not penalize the triple and higher junctions between the phases. However, one can easily penalize them in a manner similar to [16, 29]. It should be noted that in (12) the barrier energy and the gradient energy is multiplied by \( J \) and the gradient of \( \eta_0 \) and \( \eta_1 \) is expressed in \( \Omega \). This yields the desired form of the structural stresses (here given by (25)); see also [16, 23, 29] for details.
Any material property \( B \) at each material point is determined using [16]

\[
B(\eta^*, \theta, F) = B_0(1 - \varphi(a, \eta_0)) + \varphi(a, \eta_0) \sum_{i=1}^{N} B_i \phi_i(\eta),
\]

where \( B_0 \) and \( B_i \) are the properties of the phases \( A \) and \( M_i \), respectively, \( \varphi(a, \eta_0) \) has the same functional form of \( \varphi(a_*, \eta_0) \) given by (3) when \( a_* \) is replaced by the constant parameter \( a \). Note that \( \varphi(a, \eta_0) \) also satisfies the conditions similar to those satisfied by \( \varphi(a_*, \eta_0) \); see Section 1. Evidently, the interpolation (19) reduces to the properties of the phases \( A \) and \( M_i \) for appropriate values of the order parameters corresponding to each phase.

2.4. Elastic moduli of the phases

The general form of the elastic moduli for all the crystalline solids is [34]

\[
\hat{C}_{(e)ABCD} = \sum_{n=1}^{3} \sum_{s=1}^{3} \left( D_{ns}(\delta_{An}\delta_{Bs}\delta_{Cs}\delta_{Ds} + \delta_{An}\delta_{Bs}\delta_{Cs}\delta_{Ds} + \delta_{An}\delta_{Bs}\delta_{Cn}\delta_{Ds} + \delta_{An}\delta_{Bs}\delta_{Cs}\delta_{Ds}) + M_{ns}(\delta_{An} \times \delta_{Bs}\delta_{Cs}\delta_{As} + \delta_{Cs}\delta_{Ad}\delta_{As}) + N_{ns}(\delta_{An}\delta_{Cs}\delta_{BD} + \delta_{Bs}\delta_{Cn}\delta_{AD} + \delta_{An}\delta_{As}\delta_{BC} + \delta_{Bs}\delta_{Cs}\delta_{AD}) \right),
\]

where \( \delta_{ij} \) is the Kronecker delta, the coefficient \( D_{ns} \) is not symmetric, i.e., \( D_{ns} \neq D_{ns} \) (hence, nine independent coefficients), and the coefficients \( M_{ns} \) and \( N_{ns} \) are symmetric, i.e., \( M_{ns} = M_{ns} \) and \( N_{ns} = N_{sn} \) (hence, six independent coefficients from each). Thus, there are total 21 elastic constants. Note that (20) represents the elastic moduli of the triclinic crystal which has the lowest symmetry. The coefficients \( D_{ns}, M_{ns}, \) and \( N_{sn} \) are related to the components of the fourth order elasticity tensor by

\[
\begin{align*}
D_{11} &= C_{11} + C_{23} + 2C_{44} - (C_{12} + C_{13} + 2C_{55} + 2C_{66}); \\
D_{22} &= C_{22} + C_{13} + 2C_{55} - (C_{12} + C_{23} + 2C_{44} + 2C_{66}); \\
D_{33} &= C_{33} + C_{12} + 2C_{55} - (C_{12} + C_{23} + 2C_{44} + 2C_{55}); \\
D_{23} &= C_{24} - C_{14} - 2C_{56}; \\
D_{33} &= C_{15} - C_{25} - 2C_{46}; \\
D_{32} &= C_{34} - C_{14} - 2C_{56}; \\
D_{31} &= C_{35} - C_{25} - 2C_{46}; \\
M_{11} &= 0.5(C_{12} + C_{13} - C_{23}); \\
M_{22} &= 0.5(C_{12} + C_{23} - C_{13}); \\
M_{33} &= 0.5(C_{13} + C_{23} - C_{12}); \\
M_{23} &= C_{14}, \\
M_{13} &= C_{25}; \\
M_{12} &= C_{36}; \\
N_{11} &= 0.5(C_{55} + C_{66} - C_{44}); \\
N_{22} &= 0.5(C_{44} + C_{55} - C_{66}); \\
N_{33} &= 0.5(C_{44} + C_{55} - C_{66}); \\
N_{23} &= C_{56}; \\
N_{13} &= C_{46}; \\
N_{12} &= C_{45};
\end{align*}
\]

where the elastic constants \( C_{AB} \) for \( A, B = 1, \ldots, 6 \) are expressed by substituting \( 11 \to 1, 22 \to 2, 33 \to 3, 23 \to 4, 13 \to 3, 13 \to 14, 23 \to 14 \), and \( 12 \) and \( 21 \) \to 6 in the fourth-order elasticity tensor \( \hat{C}_{(e)ABCD} \). For example, \( \hat{C}_{(e)1111} = C_{11}, \hat{C}_{(e)1112} = C_{16}, \) etc. For a monoclinic crystal there are 13 independent elastic constants and

\[
C_{14} = C_{15} = C_{24} = C_{25} = C_{34} = C_{35} = C_{46} = C_{56} = 0.
\]

For an orthorhombic crystal, in addition to the conditions in (22) we have \( C_{16} = C_{26} = C_{36} = C_{45} = 0, \) and hence there are 9 independent elastic constants. A tetragonal crystal further satisfies \( C_{22} = C_{11}, C_{23} = C_{13}, C_{33} = C_{44}, \) i.e., it has 6 independent constants. A cubic crystal has 3 independent constants and additionally satisfies \( C_{33} = C_{22} = C_{11}, C_{23} = C_{13}, C_{33} = C_{12}, \) and \( C_{33} = C_{44} = C_{55} \).

In this article, we considered a monoclinic crystal for which the elasticity tensor is obtained using (22) in (20) as

\[
\hat{C}_{(e)ABCD} = \sum_{n=1}^{3} \left[ \alpha_n (\delta_{An}\delta_{Bs}\delta_{Cs}\delta_{Ds} + \delta_{An}\delta_{Bs}\delta_{Cn}\delta_{Ds} + \delta_{An}\delta_{Bs}\delta_{Cs}\delta_{Ds}) + \mu_n (\delta_{An}\delta_{Cn}\delta_{BD} + \delta_{Bs}\delta_{Ds}\delta_{AC} + \delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD} + \delta_{Bs}\delta_{Cn}\delta_{BD} + \delta_{An}\delta_{Bs}\delta_{Cs}\delta_{AD} + \delta_{An}\delta_{Bs}\delta_{Cs}\delta_{AD}) \right] + \kappa_n (\delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD} + \delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD} + \delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD} + \delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD} + \delta_{As}\delta_{Bn}\delta_{Cn}\delta_{BD})
\]

(23)
where \( \alpha_n = D_{nn}, \mu_n = M_{nn}, \nu_n = N_{nn} \) (no sum on \( n \)), and \( \kappa_1 = M_{12}, \kappa_2 = B_{12}, \kappa_3 = D_{12}, \text{ and } \kappa_4 = D_{21} \). Obviously, for orthorhombic, tetragonal, and cubic crystals, \( \kappa_1 = \kappa_2 = \kappa_3 = \kappa_4 = 0 \) in (23).

### 2.5. Mechanical equilibrium equations and stresses

Neglecting the body forces and inertia we write the mechanical equilibrium equations as [16, 23]

\[
\nabla \cdot \sigma = 0 \quad \text{in } \Omega, \quad \text{where } \sigma = \sigma_e + \sigma_{st} \quad \text{and} \quad \sigma_e = J_e^{-1}F_e \cdot \frac{\partial \psi_e(E_e)}{\partial E_e} \cdot F_e^T
\]

(24)

are the total and elastic Cauchy stress tensors, respectively, and the structural part of \( \sigma \) (interfacial stresses or tension) is

\[
\sigma_{st} = \rho_0(\dot{\psi}^\sigma + \psi \hat{\nabla}J) - \beta_{0M} \nabla \eta_0 \otimes \nabla \eta_0 - \frac{\nu}{4} \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \beta_{ij} \left[ \nabla \eta_i \otimes \nabla \eta_i + \nabla \eta_j \otimes \nabla \eta_j - 2 \text{sym}(\nabla \eta_i \otimes \nabla \eta_j) \right].
\]

(25)

The physical meaning of \( \sigma_{st} \) can be found in [23, 29]. The external boundary \( S_0 \) consists of the traction boundary \( S_{0t} \) where the traction is specified and the displacement boundary \( S_{0u} \) where the displacements are specified. The exact boundary conditions used for the problems would be specified while discussing the results in Section 6.

### 2.6. \( N \) independent Ginzburg–Landau equations

In [16] we have derived \( N + 1 \) Ginzburg–Landau equations for all \( N + 1 \) order parameters that determine the evolution of the phases. Since \( N \) order parameters describing the martensitic variants are related through the order parameters describing the martensitic variants are related through the constraint \( \sum_{i=1}^{N} \eta_i = 1 \), only the following \( N \) independent Ginzburg–Landau equations should be solved to determine the evolution of the phases (see [29] for derivation):

\[
\begin{align*}
\dot{\eta}_0 &= \mathcal{L}_{0M} \bar{X}_0, \quad \text{and} \quad \dot{\eta}_i = \sum_{j=1, \neq i}^{N} \mathcal{L}_0(\bar{X}_i - \bar{X}_j) \quad \text{for } i = 1, 2, \ldots, N - 1, \tag{26}
\end{align*}
\]

where the conjugate forces \( \bar{X}_0 \) and \( \bar{X}_i \) (for all \( i = 1, 2, \ldots, N \)) are

\[
\begin{align*}
\bar{X}_0 &= -\rho_0 \frac{\partial \psi}{\partial \eta_0} + \nabla_0 \cdot \left( \rho_0 J \frac{\partial \psi}{\partial (\nabla \eta_0)} \right) \\
&= (J F_e^{-1} \cdot \sigma_e \cdot F - J \psi^e I) : \mathcal{U}_e^{-1} \cdot \frac{\partial \mathcal{U}_e}{\partial \eta_0} - J \frac{\partial \psi}{\partial \eta_0} \bigg| _{F_e} - \rho_0 (6 \eta_0 - 6 \eta_0^2) \Delta \psi^\theta - J \rho_0 \mathcal{A} \left( \sum_{i=1}^{N-1} \sum_{j=i+1}^{N} \eta_i \eta_j^2 + \sum_{i=1}^{N-2} \sum_{j=i+1}^{N-1} \eta_i \eta_j^2 \right) \\
&\quad - \rho_0 (a_0 + 3) \Delta \psi^\theta(\theta) [2 \eta_0 - 6 \eta_0^2 + 4 \eta_0^3] \frac{J \partial \psi}{\partial \eta_0} \cdot \frac{\partial \psi}{\partial \eta_0} \times \left( \sum_{i=1}^{N-2} \sum_{j=i+1}^{N-1} \beta_{ij} | \nabla \eta_i - \nabla \eta_j |^2 + \sum_{i=1}^{N-1} \beta_{in} | \nabla \eta_i + \sum_{j=1}^{N-1} \nabla \eta_j |^2 \right) + \nabla_0 \cdot (J \rho_{0M} F_e^{-1} \cdot \nabla \eta_0), \quad \text{and} \tag{27}
\end{align*}
\]
respectively. In (27) and (28), \( \eta_N = 1 - \sum_{m=1}^{N-1} \eta_m \). We consider \( L_{ij} = 0 \) in Eq. (26) when \( \eta_i = 0 \) or \( \eta_j = 0 \). Note that all the derivatives with an overbar, e.g., \( \overline{\frac{\partial \psi}{\partial \eta_i}} \) in (27) and (28) have the following interpretation: at first \( \overline{F_{ij}}(\eta_0, \ldots, \eta_{i-1}, \eta_j, \eta_{j+1}, \ldots, \eta_N) \) is differentiated with respect to \( \eta_j \) assuming all the \( N \) order parameters \( \eta_1, \ldots, \eta_{i-1}, \eta_j, \eta_{j+1}, \ldots, \eta_N \) to be mutually independent, and after that \( \eta_N \) is substituted by \( 1 - \sum_{k=1}^{N-1} \eta_k \); see [16] for derivation. We apply homogeneous Neumann boundary condition on the external boundary for all the order parameters, i.e., \( \nabla \eta_i \cdot \mathbf{n} = 0 \) on \( S \) for \( i = 0, 1, 2, \ldots, N \), which physically means that the energy of the surface remains constant during phase transformation [23].

3. Exact expressions for first and second derivatives of \( U_t \)

In this section, we derive the first and second derivatives of the transformation stretch tensor \( U_t = F_t \) given by (10) with respect to the order parameters \( \eta_0 \) and \( \eta_i \) for all \( i = 1, \ldots, N - 1 \). Notably, the first derivatives appear in the Ginzburg–Landau equations and the second derivatives are required for analyzing the instability criteria (see Section 4) and determining the tangent matrices (see the Appendix) for finite element formulation. In Section 3.1, we enlist the expressions for the exponential of a second-order tensor and its derivatives, and in Section 3.2 we determine the exact expressions for derivatives of \( U_t \) with respect to the order parameters (see (10) and (11)).

3.1. Exponential of a second-order tensor and its derivatives

We denote the eigenvalues of an arbitrary second-order tensor \( \Lambda \) in a \( d \)-dimensional vector space by \( \{\Lambda_1, \Lambda_2, \ldots, \Lambda_d\} \) and corresponding \( d \) eigenvectors (normalized) of \( \Lambda \) and \( \Lambda^T \) are denoted by \( \{i_1, i_2, \ldots, i_d\} \) and \( \{j_1, j_2, \ldots, j_d\} \), respectively. Obviously, if \( \Lambda \) is symmetric, \( i_a = j_a \) for all \( a = 1, \ldots, d \). The spectral representations of \( \Lambda \) and \( \exp \Lambda \) (defined by (29)) are, thus, [30]

\[
\Lambda = \sum_{a=1}^{d} \Lambda_a i_a \otimes j_a \quad \text{and} \quad \exp \Lambda = \sum_{a=1}^{d} \exp(\Lambda_a) i_a \otimes j_a, \tag{29}
\]

respectively. The first derivative of \( \exp \Lambda \) with respect to \( \Lambda \), which is a fourth-order tensor, is [30]

\[
\frac{d(\exp \Lambda)}{d\Lambda} = \sum_{a=1}^{d} \sum_{b=1}^{d} f(\Lambda_a, \Lambda_b) i_a \otimes j_b \otimes j_a \otimes i_b, \tag{30}
\]

where the function

\[
f(\Lambda_a, \Lambda_b) = \begin{cases} 
\frac{\exp(\Lambda_a) - \exp(\Lambda_b)}{\Lambda_a - \Lambda_b} & \text{if } \Lambda_a \neq \Lambda_b \\
\exp(\Lambda_a) & \text{if } \Lambda_a = \Lambda_b.
\end{cases} \tag{31}
\]
The expression in (30) can be expressed in the indicial notation in a Cartesian basis as

$$
\left( \frac{d(\exp A)}{dA} \right)_{IJKL} = \sum_{a=1}^{d} \sum_{b=1}^{d} \sum_{c=1}^{d} f(\Lambda_a, \Lambda_b, \Lambda_c) \left( i_a \otimes j_c \otimes j_b \otimes i_i + i_i \otimes j_c \otimes j_b \otimes i_a \right) \otimes (j_a \otimes i_b).
$$

(32)

The second derivative of $\exp A$ with respect to $A$, which is a sixth-order tensor, is given by [30]

$$
\frac{d^2(\exp A)}{dA^2} = \sum_{a=1}^{d} \sum_{b=1}^{d} \sum_{c=1}^{d} g(\Lambda_a, \Lambda_b, \Lambda_c) \left( i_a \otimes j_c \otimes j_b \otimes i_i + i_i \otimes j_c \otimes j_b \otimes i_a \right) \otimes (j_a \otimes i_b),
$$

(33)

where $\otimes$ in (33) denotes the tensorial product between fourth- and second-order tensors. For example, if $A$ and $A$ are fourth- and second-order tensors, respectively, the indicial notation for the tensor product is $(h \otimes A)_{IJKL} = h_{IJK}A_{LM}$. The function $g(\Lambda_a, \Lambda_b, \Lambda_c)$ in (33) is given by

$$
g(\Lambda_a, \Lambda_b, \Lambda_c) = \begin{cases} (\Lambda_a - \Lambda_c) \exp(\Lambda_a) + (\Lambda_c - \Lambda_a) \exp(\Lambda_b) + (\Lambda_a - \Lambda_b) \exp(\Lambda_c) & \text{if } \Lambda_a \neq \Lambda_b \neq \Lambda_c \\ -(\Lambda_a - \Lambda_b)(\Lambda_b - \Lambda_c)(\Lambda_c - \Lambda_a) & \text{if } \Lambda_a = \Lambda_b \neq \Lambda_c \\ 0.5 \exp(\Lambda_a) & \text{if } \Lambda_a = \Lambda_b = \Lambda_c. \end{cases}
$$

In indicial notation (in Cartesian basis), Equation (33) is expressed as

$$
\left( \frac{d^2(\exp A)}{dA^2} \right)_{IJKL} = \sum_{a=1}^{d} \sum_{b=1}^{d} \sum_{c=1}^{d} g(\Lambda_a, \Lambda_b, \Lambda_c) \left[ i_{(a)} \otimes j_{(c)} \otimes j_{(b)} \otimes \epsilon_{i,M} \right] j_{(a)M} \epsilon_{i,N}.
$$

(35)

### 3.2. First and second derivatives of $U_i$ with respect to order parameters

We now derive the exact expressions for the derivatives of $U_i$ given by (10) with respect to the order parameters $\eta_0$ and $\eta_i$.

#### 3.2.1. First derivatives of $U_i$ and some related terms

From the Ginzburg–Landau equations given by (26) we note that the derivatives $\partial U_i/\partial \eta_0$ and $\partial U_i/\partial \eta_i$ (for $i = 1, \ldots, N$) must be obtained considering all of the $N + 1$ order parameters to be independent. Note that the constraint (1) was taken into account while deriving the Ginzburg–Landau equations and driving forces given by (26)–(28) [16]. Differentiating (10) with respect to the order parameters and applying the chain rule for differentiation we obtain

$$
\frac{\partial U_i}{\partial \eta_0} = \frac{\partial [\exp W(\eta^*)]}{\partial \eta_0} = \frac{\partial (\exp W)}{\partial \eta_0} : \frac{\partial W(\eta^*)}{\partial \eta_0} = \frac{\partial \varphi(\eta, \eta_0)}{\partial \eta_0} K(Z),
$$

(36)

$$
\frac{\partial U_i}{\partial \eta_i} = \frac{\partial [\exp W(\eta^*)]}{\partial \eta_i} = \frac{\partial (\exp W)}{\partial \eta_i} : \frac{\partial W(\eta^*)}{\partial \eta_i} = \varphi(\eta, \eta_0) \frac{\partial \varphi_i}{\partial \eta_i} K(\ln U_i),
$$

(37)

where we have used (10) and the second-order tensor $K(A)$ for any $A \in \text{Sym}$ is given by

$$
K(A) = \frac{\partial [\exp W(\eta^*)]}{\partial W} : A.
$$

(38)

Using (36) and (37), we obtain the explicit expressions for $U_i^{-1} \cdot \frac{\partial U_i}{\partial \eta_0}$ and $U_i^{-1} \cdot \frac{\partial U_i}{\partial \eta_i}$ appearing (i) in the Ginzburg–Landau equations (26)–(28), (ii) in the derivatives of the conjugate forces $\lambda_0$ and $\lambda_i$ used to obtain
the thermodynamic instability criteria (see Section 4), and (iii) in the weak forms of the Ginzburg–Landau equations (see the Appendix). To this end, we determine for any \( A \in \text{Sym} \)

\[
H(A) := U_i^{-1} \cdot K(A) = \exp(-\overline{W}) \cdot \left( \frac{\partial (\exp \overline{W})}{\partial \overline{W}} : A \right)
\]

\[
= \left( \sum_{a=1}^{3} \exp(-\lambda_a)w_a \otimes w_a \right) \cdot \left( \sum_{b=1}^{3} \sum_{c=1}^{3} f(\lambda_b, \lambda_c)(w_b \otimes w_c \otimes w_b \otimes w_c) : A \right)
\]

\[
= \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a)\hat{f}_{ab}(A) w_a \otimes w_b,
\]

(39)

where \( \{\lambda_1, \lambda_2, \lambda_3\} \) and \( \{w_1, w_2, w_3\} \) are the set of eigenvalues and corresponding eigenvectors (unit vectors) of the symmetric tensor \( \overline{W}^{(\eta^*)} = \overline{W}(\bar{n}) \), we have used (exp\( A \))\(^{-1} = \exp(-A) \) (see, e.g., [31, Chapter 1]), and

\[
\hat{f}_{bc}(A) = f(\lambda_b, \lambda_c) w_b \otimes w_c : A.
\]

(40)

Since \( \overline{W}^{(\eta^*)} = \overline{W}(\bar{n}) \in \text{Sym} \), \( \{w_1, w_2, w_3\} \) forms an orthonormal basis [31]. Thus, \( A_{ab} = (A \cdot w_b) \cdot w_a \) is the \( ab \)-th component in \( \{w_1, w_2, w_3\} \) basis.

We now obtain the expressions for \( H(Z) \) and \( H(\ln U_i) \), which would be used for analyzing the transformation work and the instability criteria in Section 4 and finite element computations in the Appendix. Note that \( Z \) is given by (11)_3.

Using (40) we obtain \( \hat{f}_{ab} = 0 \) when \( \eta_0 = 0 \) (since \( \overline{W} = 0 \) in that case), and

\[
\hat{f}_{ab}(Z) = f(\lambda_a, \lambda_b) (w_a \otimes w_b : Z) = \frac{1}{\phi(a, \eta_0)} f(\lambda_a, \lambda_b) \overline{W}_{ab} \quad \text{for } 0 < \eta_0 \leq 1.
\]

(41)

Since \( \overline{W}_{ab} = 0 \) for \( a \neq b \), using (39), (41), and (11)_2,3 we obtain

\[
H(Z) = \exp(-\overline{W}) \cdot \left( \frac{\partial (\exp \overline{W})}{\partial \overline{W}} : Z \right) = \sum_{a=1}^{3} \frac{1}{\phi(a, \eta_0)} \lambda_a w_a \otimes w_a = Z \quad \text{for } 0 < \eta_0 \leq 1,
\]

(42)

where we have used the spectral form of \( \overline{W} \) and (31)_2. From (42) and (11)_3 we obtain the trace of \( H(Z) \) as

\[
\text{tr} \left[ H(Z) \right] = \sum_{i=1}^{N-1} \text{tr}(\phi_i \ln U_{ii}) + \text{tr}(\overline{\phi}_N \ln U_{NN})
\]

\[
= \sum_{i=1}^{N-1} \phi_i \ln J_{ii} + \overline{\phi}_N \ln J_{NN} = \ln J_{il} \left( \sum_{i=1}^{N-1} \phi_i + \overline{\phi}_N \right),
\]

(43)

where \( J_{ii} = \det U_{ii} \), and we have used [31]

\[
\text{tr}(\ln D) = \ln(\det D)
\]

(44)

for any positive-definite second-order tensor \( D \) and \( J_{ii} = J_j \) for all \( i \neq j \) and \( i = 1, \ldots, N \). We can verify from the interpolation function \( \phi_i(\eta_j) \) defined in Section 2.2 that for a system with two variants \( M_i \) and \( M_j, \phi_i + \phi_j = 1 \). Hence, utilizing (43) we obtain \( \text{tr} \left[ H(Z) \right] = \ln J_{il} \).

When \( A = \ln U_i \), using (40) we obtain \( \hat{f}_{ab}(\ln U_i) = f(\lambda_a, \lambda_b) w_a \otimes w_b : \ln U_i = f(\lambda_a, \lambda_b)(\ln U_i)_{ab} \). It can be easily shown by applying (39) that

\[
H(\ln U_i) = \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a) f(\lambda_a, \lambda_b)(\ln U_i)_{ab} w_a \otimes w_b.
\]

(45)
Employing (45) and (31) we demonstrate that

\[ \text{tr}[H(\ln U_i)] = \text{tr}(\ln U_i) = \ln J_i, \]  

where we have used the fact that \{w_1, w_2, w_3\} is an orthonormal basis. Applying (37), (39), and (45) we calculate

\[
\mathbf{U}^{-1}_i \cdot \left( \frac{\partial \mathbf{U}_i}{\partial \eta_i} - \frac{\partial \mathbf{U}_i}{\partial \eta_j} \right) = \psi(a_e, \eta_0) \left( \frac{\partial \phi_i}{\partial \eta_i} H(\ln U_i) - \frac{\partial \phi_i}{\partial \eta_j} H(\ln U_j) \right)
\]

\[ = \psi(a_e, \eta_0) \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a f(\lambda_a, \lambda_b)) \left( \frac{\partial \phi_i}{\partial \eta_i} \ln U_i - \frac{\partial \phi_j}{\partial \eta_j} \ln U_j \right) w_a \otimes w_b, \tag{47} \]

which will participate in calculating the driving forces in the Ginzburg–Landau equations; see (26), (28), and (91). The trace of the tensor in (47) is determined using (31) and (44) as

\[
\text{tr} \left[ \mathbf{U}^{-1}_i \cdot \left( \frac{\partial \mathbf{U}_i}{\partial \eta_i} - \frac{\partial \mathbf{U}_i}{\partial \eta_j} \right) \right] = \psi(a_e, \eta_0) \ln J_i \left( \frac{\partial \phi_i}{\partial \eta_i} - \frac{\partial \phi_j}{\partial \eta_j} \right), \tag{48} \]

where we have used \(J_i = J_j\) for all \(i \neq j\), and the fact that \(w_1, w_2,\) and \(w_3\) are mutually orthonormal.

When a system with austenite and two variants \(M_i\) and \(M_j\) is considered, we have \(\phi_i + \phi_j = 1\), \(\phi_i / \partial \eta_i = \partial \phi_j / \partial \eta_j\), (see Section 2.2) and, thus, from (47) we obtain

\[
\mathbf{U}^{-1}_i \cdot \left( \frac{\partial \mathbf{U}_i}{\partial \eta_i} - \frac{\partial \mathbf{U}_i}{\partial \eta_j} \right) = \psi(a_e, \eta_0) \ln J_i \left( \frac{\partial \phi_i}{\partial \eta_i} - \frac{\partial \phi_j}{\partial \eta_j} \right) w_a \otimes w_b, \tag{49} \]

and from (48) we obtain

\[
\text{tr} \left[ \mathbf{U}^{-1}_i \cdot \left( \frac{\partial \mathbf{U}_i}{\partial \eta_i} - \frac{\partial \mathbf{U}_i}{\partial \eta_j} \right) \right] = 0. \tag{50} \]

3.2.2. Second derivatives of \(U_i\) and some related terms. In a similar manner, we derive the second derivatives of \(U_i\) with respect to the order parameters using (36), (37), and (38). Note that the second derivatives are to be taken for the terms in \(N\) independent Ginzburg–Landau equations given by (26)–(28). Thus, we obtain

\[
\frac{\partial}{\partial \eta_0} \left( \frac{\partial \mathbf{U}_i}{\partial \eta_0} \right) = \frac{\partial^2 \psi(a_e, \eta_0)}{\partial \eta_0^2} \frac{\partial (\exp \mathbf{W})}{\partial \mathbf{W}} : \mathbf{Z}(\tilde{\eta}_M) + \frac{\partial \psi(a_e, \eta_0)}{\partial \eta_0} \frac{\partial}{\partial \eta_0} \left( \frac{\partial (\exp \mathbf{W}(\eta_0, \eta_M))}{\partial \mathbf{W}} \right) : \mathbf{Z}(\tilde{\eta}_M)
\]

\[ = \frac{\partial^2 \psi(a_e, \eta_0)}{\partial \eta_0^2} \mathbf{K}(\mathbf{Z}(\tilde{\eta}_M)) + \left( \frac{\partial \psi(a_e, \eta_0)}{\partial \eta_0} \right)^2 \mathbf{L}(\mathbf{Z}(\tilde{\eta}_M), \mathbf{Z}(\tilde{\eta}_M)), \tag{51} \]

and

\[
\frac{\partial}{\partial \eta_j} \left( \frac{\partial \mathbf{U}_i}{\partial \eta_i} \right) = \psi(a_e, \eta_0) \left[ \frac{\partial}{\partial \eta_i} \left( \frac{\partial \phi_i}{\partial \eta_i} \right) \mathbf{W} : \ln \mathbf{U}_i + \frac{\partial \phi_i}{\partial \eta_i} \frac{\partial \mathbf{W}}{\partial \eta_j} : \ln \mathbf{U}_i \right]
\]

\[ = \psi(a_e, \eta_0) \left[ \frac{\partial}{\partial \eta_i} \left( \frac{\partial \phi_i}{\partial \eta_i} \right) \mathbf{K}(\ln \mathbf{U}_i) + \frac{\partial \phi_i}{\partial \eta_i} \frac{\partial \mathbf{W}}{\partial \eta_j} : \ln \mathbf{U}_i \right] \]

for \(i = 1, \ldots, N\), and \(j = 1, \ldots, N - 1\),

\[ \tag{52} \]

respectively, where \(\mathbf{W} := \frac{\partial (\exp \mathbf{W})}{\partial \mathbf{W}} = \frac{\partial (\exp \mathbf{W})}{\partial \mathbf{W}}\) and the second-order tensor \(\mathbf{L}(\mathbf{A}, \mathbf{B})\) for any \(\mathbf{A}, \mathbf{B} \in \text{Sym}\) is given by

\[ \mathbf{L}(\mathbf{A}, \mathbf{B}) := \left( \frac{\partial \mathbf{W}}{\partial \mathbf{W'}} : \mathbf{A} \right) : \mathbf{B} = \left( \frac{\partial^2 [\exp \mathbf{W}(\eta_0, \tilde{\eta}_M)]}{\partial \mathbf{W'}^2} : \mathbf{A} \right) : \mathbf{B}. \tag{53} \]
Since $A$ and $B$ are symmetric, it can be easily verified that $L(A, B)$ given by (53) is also symmetric. Note that in (52), $\left(\frac{\partial}{\partial \eta_i} \left( \frac{\partial}{\partial \eta_j} \right) = 0 \right.$ for all $i = 1, \ldots, N - 1$ and is non-trivial for $i = N$ only (recall that we consider $\eta_N = 1 - \sum_{k=1}^{N-1} \eta_k$). Using (33) we rewrite (53) as

$$L(A, B) = \sum_{a=1}^{3} \sum_{b=1}^{3} \sum_{c=1}^{3} g(\lambda_a, \lambda_b, \lambda_c) [(w_a \otimes w_c)A_{bc} + (w_c \otimes w_b)A_{cb}]B_{ab}. \quad (54)$$

Using (54) the components of $L$ in $\{w_1, w_2, w_3\}$ basis are obtained as

$$L_{11} = 2g(\lambda_1, \lambda_1, \lambda_1)A_{11}B_{11} + 2g(\lambda_2, \lambda_1, \lambda_1)A_{12}B_{12} + 2g(\lambda_3, \lambda_1, \lambda_1)A_{13}B_{13},$$

$$L_{12} = g(\lambda_1, \lambda_1, \lambda_2)A_{11}B_{12} + g(\lambda_2, \lambda_2, \lambda_1)A_{12}B_{12} + g(\lambda_3, \lambda_2, \lambda_1)A_{13}B_{12}, \quad (55)$$

$$L_{13} = g(\lambda_1, \lambda_1, \lambda_3)A_{11}B_{13} + g(\lambda_2, \lambda_2, \lambda_3)A_{12}B_{13} + g(\lambda_3, \lambda_3, \lambda_1)A_{13}B_{13},$$

$$L_{22} = 2g(\lambda_2, \lambda_2, \lambda_2)A_{22}B_{22} + 2g(\lambda_2, \lambda_2, \lambda_1)A_{22}B_{12} + 2g(\lambda_3, \lambda_2, \lambda_2)A_{23}B_{22} + 2g(\lambda_3, \lambda_2, \lambda_1)A_{23}B_{12} + g(\lambda_2, \lambda_3, \lambda_2)A_{23}B_{23},$$

$$L_{23} = 2g(\lambda_3, \lambda_1, \lambda_3)A_{31}B_{13},$$

where using (34) we determine $g(\lambda_a, \lambda_a, \lambda_b) = g(\lambda_a, \lambda_b, \lambda_a) = g(\lambda_b, \lambda_a, \lambda_a)$ and

$$g(\lambda_1, \lambda_1, \lambda_1) = 0.5 \exp(\lambda_1), \quad g(\lambda_2, \lambda_2, \lambda_2) = 0.5 \exp(\lambda_2), \quad g(\lambda_3, \lambda_3, \lambda_3) = 0.5 \exp(\lambda_3),$$

$$g(\lambda_1, \lambda_1, \lambda_2) = g(\lambda_1, \lambda_2, \lambda_1) = \frac{\lambda_1 - \lambda_2 - 1}{(\lambda_1 - \lambda_2)^2} \exp(\lambda_1 + \exp(\lambda_2)), \quad (56)$$

$$g(\lambda_1, \lambda_2, \lambda_2) = \frac{\lambda_1 - \lambda_3 - 1}{(\lambda_1 - \lambda_3)^2} \exp(\lambda_1 + \exp(\lambda_3)),$$

$$g(\lambda_1, \lambda_3, \lambda_3) = \frac{\lambda_1 - \lambda_2 - 1}{(\lambda_1 - \lambda_2)^2} \exp(\lambda_3 + \exp(\lambda_2)), \quad (56)$$

Using (53) we obtain the following expression that would be used in (95) and (97)

$$N(A, B) := U^{-1} \cdot \left( \frac{\partial^2 (\exp W)}{\partial W^2} : A \right) : B = \exp(-W) \cdot \left( \frac{\partial^2 (\exp W)}{\partial W^2} : A \right) : B \quad (57)$$

Utilizing (57) we obtain $N(Z, Z)$ and its expression in terms of $\frac{\partial W}{\partial \eta_i}$, that will, in turn, be used to obtain some necessary expressions for the instability criteria in Section 4 and for finite element computations in the Appendix. Noting that $Z_{12} = Z_{23} = Z_{13} = 0$ in $(w_1, w_2, w_3)$ basis, we calculate using (57)

$$N(Z, Z) = \frac{1}{\varphi(a, \eta_0)^2} \exp(-W) \cdot \left( \frac{\partial^2 (\exp W)}{\partial W^2} : W \right) : W \quad \text{for} \ 0 < \eta_0 \leq 1$$

$$= \frac{1}{\varphi(a, \eta_0)^2} [\lambda_1^2 w_1 \otimes w_1 + \lambda_2^2 w_2 \otimes w_2 + \lambda_3^2 w_3 \otimes w_3] = \frac{1}{\varphi(a, \eta_0)^2} \tilde{W}^2 = \tilde{Z}^2. \quad (58)$$
Using (11) and (57) we calculate the components of the tensor $N\left( \frac{\partial W}{\partial \eta_j}, \ln U_\sigma \right)$ given by

$$N \left( \frac{\partial W}{\partial \eta_j}, \ln U_\sigma \right) = \exp(-W) \cdot \left[ \frac{\partial^2 (\exp W)}{\partial W^2} \cdot \left( \frac{\partial \phi_j(\eta_j)}{\partial \eta_j} \ln U_{ij} + \frac{\partial \phi_N(\eta_1, \ldots, \eta_N)}{\partial \eta_j} \ln U_{iN} \right) \right] : \ln U_\alpha, \quad (59)$$

where we have used the fact that $\phi_N(\eta_N) = \bar{\phi}_N(\eta_M)$ (recall the constraint (1)). Using (52) and (59) we finally determine

$$\frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) = \varphi(a_\sigma, \eta_0) \left[ \frac{\partial^2 \phi_j}{\partial \eta_i^2} K(\ln U_\sigma) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial \phi_j}{\partial \eta_j} \right) K(\ln U_\sigma) + \varphi(a_\sigma, \eta_0) \frac{\partial \phi_i}{\partial \eta_i} \times L \left( \frac{\partial W}{\partial \eta_i}, \ln U_\sigma \right) \right], \quad (60)$$

and

$$U_\sigma^{-1} \cdot \left[ \frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) \right] = \varphi(a_\sigma, \eta_0) \left[ \frac{\partial^2 \phi_j}{\partial \eta_i^2} H(\ln U_\sigma) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial \phi_j}{\partial \eta_j} \right) H(\ln U_\sigma) + \varphi(a_\sigma, \eta_0) \frac{\partial \phi_i}{\partial \eta_i} \times N \left( \frac{\partial W}{\partial \eta_i}, \ln U_\sigma \right) \right]. \quad (61)$$

Note that in (60) and (61) the term $\partial \phi_j/\partial \phi_i = 0$ for $j = 1, \ldots, N-1$ and it is non-trivial for $j = N$ only, owing to the constraint $\sum_{i=1}^N \eta_i = 1$.

For a two-variant system with $M_i$ and $M_j$ we have $\eta_i + \eta_j = 1$, $\phi_i + \phi_j = 1$, $\partial \phi_j/\partial \eta_i = \partial \phi_i/\partial \eta_j$, and $\bar{W}(\eta_0, \eta_0) = \varphi(a_\sigma, \eta_0)[\ln U_\sigma + \phi(\ln U_\sigma - \ln U_\sigma)]$. Hence, Equation (61) can be expressed as a function of $\eta_0$ and $\eta_j$ and simplified to

$$U_\sigma^{-1} \cdot \left[ \frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_j} \right) \right] = \varphi(a_\sigma, \eta_0) \left[ \frac{\partial^2 \phi_j}{\partial \eta_i^2} H(\ln U_\sigma) - \frac{\partial}{\partial \eta_i} \left( \frac{\partial \phi_j}{\partial \eta_j} \right) H(\ln U_\sigma) + \varphi(a_\sigma, \eta_0) \frac{\partial \phi_i}{\partial \eta_i} \times N \left( \frac{\partial W}{\partial \eta_i}, \ln U_\sigma \right) \right]. \quad (62)$$

### 4. Analysis of transformation work and instability criteria for the phases

In this section, we analyze the transformation works from the Ginzburg–Landau equations given by (26)–(28) in Section 4.1, and determine the thermodynamic instability criteria for the homogeneous phases in Section 4.2.

#### 4.1. Analysis of transformation work

The transformation work appearing in $X_0$ given by (27) can be rewritten as

$$W_0 = (J F^{-1} \cdot \sigma_\varepsilon \cdot F - J_i \psi^i I) : U_i^{-1} \cdot \frac{\partial U_i}{\partial \eta_0}$$

$$= (J \sigma_{00} - J_i \psi^i) \text{tr} \left( U_i^{-1} \cdot \frac{\partial U_i}{\partial \eta_0} \right) + J F^{-1} \cdot \text{dev} \sigma_\varepsilon \cdot F : \text{dev} \left( U_i^{-1} \cdot \frac{\partial U_i}{\partial \eta_0} \right), \quad (63)$$

where we have decomposed the Cauchy elastic stress $\sigma_\varepsilon$ as $\sigma_\varepsilon = \sigma_{00} I + \text{dev} \sigma_\varepsilon$, and $\sigma_{00}$ is the mean part of $\sigma_\varepsilon$.

In addition, we note that $F^{-1} \cdot \text{dev} \sigma_\varepsilon \cdot F$ is a deviatoric tensor, which we can prove by showing that its trace
identically vanishes: \( \text{tr}(F^{-1} \cdot \text{dev} \sigma_e \cdot F) = I : F^{-1} \cdot \text{dev} \sigma_e \cdot F = F \cdot F^{-1} : \text{dev} \sigma_e = \text{tr}(\text{dev} \sigma_e) = 0 \). Using (11), (42), and (43) we have \( W_0 = 0 \) when \( \eta_0 = 0 \) and

\[
W_0 = \frac{\partial \varphi(a_s, \eta_0)}{\partial \eta_0} \left[ (J_{\sigma_0} - J_i \psi^e) \text{tr} Z + J F^{-1} \cdot \text{dev} \sigma_e \cdot F : \text{dev} Z \right]
\]

\[
= \frac{\partial \varphi(a_s, \eta_0)}{\partial \eta_0} \left[ (J_{\sigma_0} - J_i \psi^e) \ln J_i \left( \sum_{i=1}^{N-1} \phi_i + \overline{\phi}_N \right) + J F^{-1} \cdot \text{dev} \sigma_e \cdot F : \text{dev} \left( \sum_{i=1}^{N-1} \phi_i \ln U_i + \overline{\phi}_N \ln U_N \right) \right]
\]

for \( 0 < \eta_0 \leq 1 \).

For a system with single variant \( M_i \), only \( \eta_0 \) is sufficient to describe the MT. In that case, we consider \( \eta_N = 1 \) and \( \eta_j = 0 \) for \( j \neq N \). Hence, Equation (64) simplifies to

\[
W_0 = \frac{\partial \varphi(a_s, \eta_0)}{\partial \eta_0} \left[ (J_{\sigma_0} - J_i \psi^e) \ln J_i + J F^{-1} \cdot \text{dev} \sigma_e \cdot F : \text{dev} \left( \ln U_i \right) \right].
\]  

(65)

From (65) it is obvious that for \( A \leftrightarrow M \) transformations, the transformation work \( W_0 \) can be decoupled into those due to volumetric parts of generalized elastic Cauchy stress \( J F^{-1} \cdot \sigma_e \cdot F - J_i \psi^e I \) and \( \ln U_i = \overline{W} \) (see (11)) and the corresponding deviatoric parts.

For the Ginzburg–Landau equations for \( \eta_i \) (see (26)2), the transformation work from the driving force \( \overline{X}_i - \overline{X}_j \) at any material point is given by

\[
W_{ij} = (J F^{-1} \cdot \sigma_e \cdot F - J_i \psi^e I) : \overline{U}^{-1} \cdot \left( \frac{\partial \overline{U}}{\partial \eta_i} - \frac{\partial \overline{U}}{\partial \eta_j} \right)
\]

\[
= (J_{\sigma_0} - J_i \psi^e) \text{tr} \left( \overline{U}^{-1} \cdot \frac{\partial \overline{U}}{\partial \eta_i} - \overline{U}^{-1} \cdot \frac{\partial \overline{U}}{\partial \eta_j} \right) + J F^{-1} \cdot \text{dev} \sigma_e \cdot F : \text{dev} \left( \overline{U}^{-1} \cdot \frac{\partial \overline{U}}{\partial \eta_i} - \overline{U}^{-1} \cdot \frac{\partial \overline{U}}{\partial \eta_j} \right)
\]

\[
= (J_{\sigma_0} - J_i \psi^e) \varphi(a_s, \eta_0) \ln J_i \left( \frac{\partial \phi_i}{\partial \eta_i} - \frac{\partial \phi_i}{\partial \eta_j} \right) + J \varphi(a_s, \eta_0) \frac{\partial \phi_i}{\partial \eta_i} F^{-1} \cdot \text{dev} \sigma_e \cdot F :
\]

\[
\text{dev} \left( \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a f(\lambda_a, \lambda_b) [(\ln U_{ia})_{ab} - (\ln U_{ib})_{ab}] \mathbf{w}_a \otimes \mathbf{w}_b \right),
\]

(66)

where we have used (47) and (48). For obtaining (66)1, we used the same approach as was used for obtaining (63). When a system with only two variants \( M_i \) and \( M_j \) is considered, \( W_{ij} \) in (66) simplifies to

\[
W_{ij} = J \varphi(a_s, \eta_0) \frac{\partial \phi_i}{\partial \eta_i} F^{-1} \cdot \text{dev} \sigma_e \cdot F : \left( \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a f(\lambda_a, \lambda_b) [(\ln U_{ia})_{ab} - (\ln U_{ib})_{ab}] \mathbf{w}_a \otimes \mathbf{w}_b \right),
\]

(67)

where we have used (50). We conclude from (67) that the transformation rule given by (10) yields a vanishing volumetric part of the transformation work during variant–variant transformations for any MT. This is a plausible condition, although the actual transformation path is not yet known. The result in (67) of vanishing volumetric transformation work during variant–variant transformation for the present KM is more general than that was proved in [16] for the commutative Bain tensors only, in particular, for cubic to tetragonal transformation.

### 4.2. Instability criteria for MTs

We now determine the criteria for instability of all the homogeneous phases under specified stresses and temperature. The instability criterion is as follows:

If for a thermodynamic equilibrium state \( (\hat{\eta}_j \text{ for } j = 0, 1, 2, \ldots, N) \) a spontaneous perturbation \( \Delta \eta \) of the order parameters is thermodynamically admissible under prescribed stresses and temperature, i.e., the dissipation rate is positive, then the equilibrium is unstable; see [14, 16, 22] for details. It was shown in [22] that
the instability criteria do not depend on which stress, e.g., the first Piola–Kirchhoff or Cauchy stress tensor, is prescribed. Using this definition one can show that the criteria for \( A \leftrightarrow M \) transformations are

\[
A \rightarrow M \quad \frac{\partial X_0(T, F_c(\eta_0 = 0), \eta_0 = 0, \theta)}{\partial \eta_0} \bigg|_T \geq 0, \quad M \rightarrow A \quad \frac{\partial X_0(T, F_c(\eta_0 = 1), \eta_0 = 1, \theta)}{\partial \eta_0} \bigg|_T \geq 0. \quad (68)
\]

In (68), \( T \) denotes the specified Piola-Kirchhoff or Cauchy stress tensor.

In order to obtain the criteria for variant\( \leftrightarrow \)variant transformations, we consider a fully martensitic system with variants \( M_i \) and \( M_j \) only. Thus, we set \( \eta_0 = 1 \) everywhere, and the only non-trivial order parameters \( \eta_i \) and \( \eta_j \) satisfy the constraint \( \eta_i + \eta_j = 1 \). The criteria for \( M_i \rightarrow M_j \) and \( M_j \rightarrow M_i \) transformations are therefore obtained using the above definition as

\[
M_i \rightarrow M_j \quad \frac{\partial X_{ij}(T, F_c(\hat{\eta}_i), \hat{\eta}_j, \theta)}{\partial \eta_i} \bigg|_T \geq 0, \quad \text{and} \quad M_j \rightarrow M_i \quad \frac{\partial X_{ji}(T, F_c(\hat{\eta}_j), \hat{\eta}_i, \theta)}{\partial \eta_j} \bigg|_T \geq 0, \quad (69)
\]

respectively, where recall that \( X_{ij} = \bar{X}_i - \bar{X}_j \) is to be calculated using Eq. (28).

### 4.2.1. Criteria for \( A \leftrightarrow M \) transformation

To obtain the explicit expressions for instability criteria from the inequalities (68), we determine with the help of (27) that

\[
\frac{\partial X_0}{\partial \eta_0} \bigg|_T = (J(\sigma_{\alpha 0} - J_i \psi^c) \text{tr} (U_i^{-1} \cdot \frac{\partial^2 U_i}{\partial \eta_0^2}) + JF^{-1} \cdot \text{dev}(\sigma_c) \cdot F : \text{dev}(U_i^{-1} \cdot \frac{\partial^2 U_i}{\partial \eta_0^2}) - (6 - 12 \eta_0) \rho_0 \Delta \psi^c(\theta) - 0.5J F_e : E_c - J \rho_0 [A(\theta) + (a_0 - 3) \Delta \psi^c(\theta)] \{2 - 12 \eta_0 + 12 \eta_0^2\} \quad \text{for} \ \eta_0 = 0, 1, \quad (70)
\]

where we have assumed \( \eta_0 = 1 \) and \( \eta_i = 0 \) for all \( j \neq i \), i.e., \( M = M_i \) without loss of generality. Using (70), (51), (42), and (43) in both inequalities given in (68) we finally establish the following instability criteria:

\[
A \rightarrow M : \quad a_i(J(\sigma_{\alpha 0} - J_i \psi^c) \text{tr} (\ln U_i) + a_i J_i F^{-1} \cdot \text{dev}(\sigma_c) \cdot F : \text{dev}(\ln U_i) - \rho_0 [3 + J_i(a_0 - 3)] \Delta \psi^c + 0.5a_i E_c : (C_0 - C_i) : E_c \geq \rho_0 J_i A(\theta); \quad (71)
\]

\[
M \rightarrow A : \quad (6 - a_i)(J(\sigma_{\alpha 0} - J_i \psi^c) \text{tr}(\ln U_i) + (6 - a_i)J F^{-1} \cdot \text{dev}(\sigma_c) \cdot F : \text{dev}(\ln U_i) + [J(a_0 - 3) - 3] \rho_0 \Delta \psi^c + 0.5(6 - a_i)J_i E_c : (C_0 - C_i) : E_c \leq -\rho_0 J_i A. \quad (71)
\]

Note that in inequalities (71)\(_1\) and (71)\(_2\), the volumetric and deviatoric parts of the transformation work related terms are decoupled.

### 4.2.2. Criteria for \( M_i \leftrightarrow M_j \) transformations

We obtain the explicit form of the instability criteria for variant\( \leftrightarrow \)variant transformations using inequalities (69). Using (28) we calculate

\[
\frac{\partial X_{ij}}{\partial \eta_i} \bigg|_T = (J(\sigma_{\alpha 0} - J_i \psi^c) \text{tr} \left[ U_i^{-1} \left( \frac{\partial U_i}{\partial \eta_i} - \frac{\partial U_i}{\partial \eta_j} \right) \right] + JF^{-1} \cdot \text{dev}(\sigma_c) \cdot F : \text{dev} \left[ U_i^{-1} \left( \frac{\partial U_i}{\partial \eta_i} - \frac{\partial U_i}{\partial \eta_j} \right) \right] - J_i \frac{\partial \psi^c}{\partial \eta_i} \left( \frac{\partial U_i}{\partial \eta_i} \right)_T - \rho_0 \tilde{A} J (2 - 12 \eta_i + 12 \eta_i^2) \quad \text{at} \ \eta_i = 0 \ \text{and} \ 1. \quad (72)
\]
Finally, using (72), (50), (49), and (46) in inequalities (69), the instability criteria for variant–variant transformations are obtained:

\[
\mathbf{M}_i \rightarrow \mathbf{M}_j : \quad JF^{-1} \cdot \text{dev} \sigma_c \cdot F : \left( \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a f(\lambda_a, \lambda_b)) ([\ln U_{ab}^{(i)}] - (\ln U_{ab}^{(j)}) \mathbf{w}_a \otimes \mathbf{w}_b \right) - 0.5J_i E_c : (\mathbf{C}_i - \mathbf{C}_j) : E_c \geq \frac{\rho_0 J_i A}{3};
\]

\[
\mathbf{M}_i \rightarrow \mathbf{M}_j : \quad JF^{-1} \cdot \text{dev} \sigma_c \cdot F : \left( \sum_{a=1}^{3} \sum_{b=1}^{3} \exp(-\lambda_a f(\lambda_a, \lambda_b)) ([\ln U_{ab}^{(i)}] - (\ln U_{ab}^{(j)}) \mathbf{w}_a \otimes \mathbf{w}_b \right) - 0.5J_i E_c : (\mathbf{C}_i - \mathbf{C}_j) : E_c \leq \frac{\rho_0 J_i A}{3}. \tag{73}
\]

Note that the volumetric part related to the transformation work does not contribute to the instability criteria for \(\mathbf{M}_i \leftrightarrow \mathbf{M}_j\) transformations, which is a plausible condition. In contrast, for a linear transformation rule, the volumetric part related to the transformation work is non-vanishing [16], which is not desired. As a special case, when the Bain stretch tensors are diagonal (hence, commutative), using (31) and using \(\text{tr}([\ln U_{ab}^{(i)}] = \text{tr}([\ln U_{ab}^{(j)}])\) or, equivalently, \(\ln J_i = \ln J_j\) (see (44) and [1]) the inequalities (73)\(_{1,2}\) are simplified to

\[
\mathbf{M}_j \rightarrow \mathbf{M}_i : \quad JF^{-1} \cdot \text{dev} \sigma_c \cdot F : \text{dev} ([\ln U_{ab}^{(i)}] - [\ln U_{ab}^{(j)}) \mathbf{C}_i - \mathbf{C}_j) : E_c \geq \frac{\rho_0 J_i A}{3};
\]

\[
\mathbf{M}_i \rightarrow \mathbf{M}_j : \quad JF^{-1} \cdot \text{dev} \sigma_c \cdot F : \text{dev} ([\ln U_{ab}^{(i)}] - [\ln U_{ab}^{(j)}) \mathbf{C}_i - \mathbf{C}_j) : E_c \leq \frac{\rho_0 J_i A}{3}. \tag{74}
\]

The inequalities (74)\(_{1,2}\) coincide with those derived for variant–variant transformations in tetragonal lattices in [16] where the Bain stretch tensors were diagonal. However, the criteria in (73)\(_{1,2}\) are general and apply for all kinds of variant–variant transformations.

5. Relations from crystallographic theory of twinned martensite

From the crystallographic theory of MTs [1], we know that austenite phase and a single martensite variant is not compatible for almost all the materials capable of undergoing MTs, and hence they cannot form sharp interfaces between them. We usually see microstructures with austenite and twinned martensite between two variants and a finite-width interface between \(A\) and twinned \(M\) (see the schematic in Figure 1(a) and also see [1]). Austenite phase and the alternative martensitic plates (twinned \(M\)) are shown. The \(A\)-twinned \(M\) interface is obviously of finite width.

Let us denote the unit normals to \(A\)-twinned \(M\) and twin boundary by \(\mathbf{m}\) and \(\mathbf{n}_t\), respectively, in a Cartesian basis whose axes are parallel to three perpendicular directions of a unit cell. The Hadamard’s compatibility relations for a twin boundary (between undeformed \(\mathbf{M}_i\) and \(\mathbf{M}_j\)) and the \(A\) (undeformed)-twinned \(M\) interface (see Figure 1(a)) are [1]

\[
\mathbf{Q}_1 : U_{ij} - U_{ij} = \mathbf{a} \otimes \mathbf{n}_t \quad \text{and} \quad \mathbf{Q}_2 : [\zeta \mathbf{Q}_1 \cdot U_{ij} + (1 - \zeta)U_{ij}] = \mathbf{I} + \mathbf{b}_t \otimes \mathbf{m}, \quad \tag{75}
\]

respectively, where \(\mathbf{Q}_1\) and \(\mathbf{Q}_2\) are rotations, \(\mathbf{a}\) and \(\mathbf{b}_t\) are two vectors related to twin boundary and \(A\)-\(M\) interfaces, and \(\zeta \in [0, 1]\) is the volume fraction of \(\mathbf{M}_i\) in \(\mathbf{M}_i\)-\(\mathbf{M}_j\) mixture. The average deformation gradient in a domain containing undeformed austenite and undeformed twinned martensite (with \(\mathbf{M}_i\) and \(\mathbf{M}_j\)) is given by [29]

\[
F_{av} = \zeta_0 M F_0 + (1 - \zeta_0 M) [\zeta F_j + (1 - \zeta) F_i] = \mathbf{I} + (1 - \zeta_0 M) \mathbf{b} \otimes \mathbf{m}, \quad \tag{76}
\]

where \(\zeta_0 M\) is the volume fraction of \(A\) within the sample, \(F_0 = \mathbf{I}, F_j = \mathbf{Q}_2 \cdot U_{ij},\) and \(F_j = \mathbf{Q}_2 \cdot \mathbf{Q}_1 \cdot U_{ij}\) are the deformation gradient tensors in undeformed \(A, M_i,\) and \(M_j,\) respectively. Note that \(U_{ij}\) and \(U_{ij}\) are known for a given material. Using them in (75)\(_1\) and (75)\(_2\) the analytical solutions for \(\mathbf{c}, \mathbf{b}, \mathbf{m}, \mathbf{Q}_1, \mathbf{Q}_2, \mathbf{n}_t,\) and \(\mathbf{a}\) can be determined; see, for example, [1, 35]. The solutions are customarily presented with respect to a Cartesian basis corresponding to the \(A\) unit cell.
In this article, we are interested in obtaining twinned microstructures by numerically solving the Ginzburg–Landau equations given by (26)\textsubscript{1,2} for CuAlNi and NiTi alloys. Since the twinned microstructures are very special microstructures, for numerical simulations proper boundary conditions have to be chosen. As we are interested in arresting the stationary microstructures, we will apply displacements at the boundaries of the sample at \( t = 0 \) and let the order parameters evolve until the stationary solution is reached. Such boundary displacements are to be obtained using

\[
\mathbf{u}|_{S_0} = (\nabla_0 \mathbf{u})_{av} \cdot \mathbf{r}_0 = (F_{av} - I) \cdot \mathbf{r}_0 \quad \forall \mathbf{r}_0 \in S_0,
\]

where \( F_{av} \) is calculated using (76), which involve the expressions for \( \mathbf{b} \) and \( \mathbf{m} \). If we use the expressions for \( \mathbf{b} \) and \( \mathbf{m} \) with respect to a Cartesian basis \( \{c_1, c_2, c_3\} \) whose axes are parallel to three perpendicular sides of cubic A unit cell, usually \((\nabla_0 \mathbf{u})_{am}\) have all nine non-trivial components (see Section 6 for specific examples).

Alternatively, we can choose a very special Cartesian coordinate system \( \{e_1, e_2, e_3\} \) as shown in the 3D austenite-twinned martensitic microstructure in Figure 1(a). The arrangement of the twin plates of variants \( \mathbf{M}_i \) and \( \mathbf{M}_j \) are shown. The A phase is shown in light blue colour. We have chosen this orientation of the microstructure such that the \( e_1-e_2 \) plane is coplanar with the plane formed by the unit normal to a twin boundary (denoted by \( n'_t \) when expressed in the \( \{e_1, e_2, e_3\} \) basis) and the unit normal to the A-twinned \( \mathbf{M} \) interface (denoted by \( m' \) when expressed in the \( \{e_1, e_2, e_3\} \) basis). The invariant planes corresponding to A–M interfaces are also shown by dashed lines. We have proved in Section 6 that if we transform \((\nabla_0 \mathbf{u})_{av}\) (expressed in \( \{e_1, e_2, e_3\} \) basis) to \((\nabla_0 \mathbf{u})_{av}'\), which is expressed in the \( \{e_1, e_2, e_3\} \) basis, the elements of the third column of \((\nabla_0 \mathbf{u})_{av}'\) are trivial. This implies that the microstructure is independent of the coordinate along \( e_3 \), thereby allowing us to simulate the twinned microstructure in a two-dimensional plane shown in Figure 1(b) using the generalized plane strain approach [5, 6]; see Section 6 for more detailed explanation. Note that the two-dimensional microstructure...
Figure 2. The unit cells of cubic A (dashed green lines) and orthorhombic CuAlNi variants M₁ (solid red lines) and M₃ (solid blue lines) for the twinning problem with respect to (a) the \( \{c_1, c_2, c_3\} \) basis where the axes are parallel to three perpendicular sides of A unit cell, and (b) the \( \{e_1, e_2, e_3\} \) basis as shown in Figure 1.

Figure 3. The unit cells of cubic A (dashed green lines) and monoclinic-I NiTi variants M₁ (solid red lines) and M₅ (solid blue lines) for the twinning problem (a) in the \( \{c_1, c_2, c_3\} \) basis, where the axes are parallel to three perpendicular sides of A unit cell, and (b) in the \( \{e_1, e_2, e_3\} \) basis as shown in Figure 1.

shown in Figure 1(b) is a projection of the three-dimensional microstructure (shown in Figure 1(a)) on the \( e_1-e_2 \) plane within the segment P₁P₂P₃P₄. The boundaries P₁P₂ and P₃P₄ are two invariant planes.

6. Numerical results

Using the formulation presented previously, in this section we show numerical examples of twinned microstructures:

(i) for CuAlNi SMA where A and the martensitic variants are cubic and orthorhombic crystals [1], respectively; and

(ii) for NiTi SMA where A and the martensitic variants are cubic and monoclinic-I crystals [1], respectively.

For CuAlNi and NiTi alloys there are 6 and 12 martensitic variants, respectively [1]. Without loss of generality, for CuAlNi we choose variants M₁ and M₃, which are in a twin relationship, and corresponding Bain tensors in the Cartesian basis \( \{c_1, c_2, c_3\} \) of cubic unit cell of A are (see [1, Chapter 4])

\[
U_{11} = \begin{bmatrix} 0.5(\alpha + \epsilon) & 0 & 0.5(\alpha - \epsilon) \\ 0 & \beta & 0 \\ 0.5(\alpha - \epsilon) & 0 & 0.5(\alpha + \epsilon) \end{bmatrix} \quad \text{and} \quad U_{13} = \begin{bmatrix} 0.5(\alpha + \epsilon) & 0.5(\alpha - \epsilon) & 0 \\ 0.5(\alpha - \epsilon) & 0.5(\alpha + \epsilon) & 0 \\ 0 & 0 & \beta \end{bmatrix},
\]  

\[ (78) \]
respectively, with \( \alpha = 1.0619 \), \( \epsilon = 1.0230 \), and \( \beta = 0.9178 \). For NiTi, we choose variants \( M_1 \) and \( M_5 \), which are in a twin relationship, and corresponding Bain tensors in \( \{e_1, e_2, e_3\} \) basis are (see [1, Chapter 4])

\[
U_{11} = \begin{bmatrix} \chi & \epsilon & \epsilon \\ \epsilon & \alpha & \beta \\ \epsilon & \beta & \alpha \end{bmatrix} \quad \text{and} \quad U_{15} = \begin{bmatrix} \alpha & \epsilon & \beta \\ \epsilon & \chi & \epsilon \\ \beta & \epsilon & \alpha \end{bmatrix},
\]

(79)

respectively, where \( \alpha = 1.0243 \), \( \epsilon = -0.0427 \), \( \beta = 0.0580 \), and \( \chi = 0.9563 \).

For both cases the Bain tensors are non-commutative. Thus, the procedure for determining the derivatives of \( U \), with respect to the order parameters presented in [16, 29] cannot be used, and the formulation presented in Sections 3 and 4 should be applied. The crystallographic solutions of \( n_t, a, \xi, b, \) and \( m \) for CuAlNi and NiTi alloys are listed in Tables 1 and 2, respectively. Obviously, for CuAlNi the twins are of Type-I [1, 5] and for NiTi the twins are of Type-II [1]. For our simulations we rotate the Bain tensors in such a way that the orientation of the unit normal to the twin boundary \( n_t \) is parallel to vector \( e_1 \) of the Cartesian basis \( \{e_1, e_2, e_3\} \) considered for the sample; see Figure 1 for schematic. Thus, the rotated twin boundary normal vector is \( n_t' = R' \cdot n_t = e_1 \), where the rotation \( R' \) for CuAlNi and NiTi are

\[
R'_{\text{CuAlNi}} = \begin{bmatrix} 0 & 0.7041 & -0.7041 \\ 0.6911 & 0.5137 & 0.5085 \\ 0.7228 & -0.4887 & -0.4887 \end{bmatrix}
\]

\[
R'_{\text{NiTi}} = \begin{bmatrix} -0.2954 & -0.2954 & -0.9006 \\ -0.9489 & -0.0055 & 0.3156 \\ 0.0989 & -0.9547 & 0.2807 \end{bmatrix},
\]

(80)

respectively, such that \( e_3 \) is perpendicular to the plane formed by \( n_t' \) and \( m' \). The average distortion tensors for the respective crystals in the rotated frame are

\[
(\nabla_0 u')_{\text{CuAlNi}}^{\text{av}} = (1 - \zeta_{0M}) \begin{bmatrix} -0.0329 & -0.0638 & 0 \\ 0.0110 & 0.0213 & 0 \\ 0.0324 & 0.0628 & 0 \end{bmatrix},
\]

\[
(\nabla_0 u')_{\text{NiTi}}^{\text{av}} = (1 - \zeta_{0M}) \begin{bmatrix} -0.0051 & -0.0534 & 0 \\ -0.0013 & -0.0138 & 0 \\ -0.0078 & -0.0819 & 0 \end{bmatrix},
\]

(81)

respectively, which we have calculated using (see (76)) \( (\nabla_0 u')_{\text{av}} = F'_{\text{av}} - I = (1 - \zeta_{0M}) R' \cdot b_i \otimes R' \cdot m \). We conclude from (81) that the microstructure in the plane made by \( m' \) and \( n' \) is independent of the coordinate along \( e_3 \) for both CuAlNi and NiTi alloys. Thus, we can perform calculations in a two-dimensional sample while taking all three displacements into consideration and without sacrificing the accuracy. Obviously, the displacement components are functions of \( r_{01} \) and \( r_{02} \) only, i.e., \( u_a = u_a(r_{01}, r_{02}) \) for \( a = 1, 2, 3 \). This is called the generalized plane strain approach [5, 6]. The transformation stretch tensor and the elasticity tensor at each material point are obtained in the coordinate system \( \{e_1, e_2, e_3\} \) using the standard transformation rules [31]:

\[
U'_{(1)AB} = R'_{AC} U_{(1)CD} R'_{BD} \quad \text{and} \quad C'_{(e)ABCD} = R'_{AE} R'_{BF} R'_{CG} R'_{DH} \hat{C}_{(e)EFGH},
\]

(82)

where \( U_{(1)CD} \) and \( \hat{C}_{(e)EFGH} \) are given by (11)_1 and (13)_2, respectively, and the rotations \( R' \) for both the crystals are given in (80).
Table 2. Crystallographic solution for NiTi alloy in Cartesian $\{c_1, c_2, c_3\}$ basis of cubic unit cell of A.

<table>
<thead>
<tr>
<th>$n_t$</th>
<th>$-0.2954 c_2 -0.2954 c_2 -0.9006 c_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>$-0.2146 c_1 + 0.2292 c_2 + 0.0216 c_3$</td>
</tr>
<tr>
<td>$\xi$</td>
<td>0.3231</td>
</tr>
<tr>
<td>$b_t$</td>
<td>$0.0278 c_1 + 0.1008 c_2 + 0.0375 c_3$</td>
</tr>
<tr>
<td>$m$</td>
<td>$-0.8891 c_1 + 0.0419 c_2 + 0.4558 c_3$</td>
</tr>
</tbody>
</table>

Figure 4. Twinned martensite (Type-I; see Table 1) in CuAlNi samples (shown in \(\Omega_1\)) with sizes (i) \(w = 15\) nm, \(l = 25\) nm, (ii) \(w = 20\) nm, \(l = 35\) nm, and (iii) \(w = 30\) nm, \(l = 50\) nm. The plots for \(\eta_{eq}\) are shown. Dark red with \(\eta_{eq} = 1\) denotes M\(_1\), dark blue with \(\eta_{eq} = -1\) denotes M\(_3\), and green with \(\eta_0 = 0\) denotes either A (for \(\eta_0 = 0\)) or points in M\(_1\)–M\(_3\) interfaces (for \(\eta_i = 0.5\)).

6.1. Boundary and initial conditions

We apply Dirichlet displacement boundary condition on all the external boundaries of the sample using

$$u|_{S_0} = (\nabla_0 u)^{\text{av}} \cdot r_0 \quad \forall r_0 \in S_0$$  \(83\)

at \(t = 0\), fix the boundary at that configuration for all \(t > 0\), and let the microstructure evolve, where \((\nabla_0 u)^{\text{av}}\) for respective sample is obtained using (81). Thus, we impose that A–M and twin boundaries correspond to the solution of crystallographic theory of the martensite [1]. Note that the faces \(P_1P_2\) and \(P_3P_4\) (in Figure 1) are the invariant planes and, hence, the twin plates would span over the entire sample where the twin boundaries are parallel to \(e_2\)-axis in \(\Omega_0\). Thus, while calculating the boundary displacements to be applied, we use \(\zeta_{0M} = 0\) in (83); see Figure 1(b). However, if one wants to have residual A in the sample as shown in Figure 1(a), a non-zero \(\zeta_{0M}\) must be used to calculate the boundary displacements and the regions of residual A must be constrained to \(\eta_0 = 0\). Here the initial conditions for the order parameters are taken as \(0 \leq \eta_0, \eta_i \leq 1\) distributed randomly in the entire sample. Recall that we are considering homogeneous Neumann boundary condition for all the order parameters as discussed in Section 2.6.

6.2. Material parameters

We assume the energy and width of A–M and M\(_i\)–M\(_j\) interfaces for both CuAlNi and NiTi sample to be \(\gamma_{0M} = 0.2\) N/m, \(\gamma_{ij} = 0.02\) N/m, \(\delta_{0M} = 2\) nm, and \(\delta_{ij} = 0.5\) nm; see [6] for the typical values. Using these constants and the analytical relations between interfacial energy and width with the parameters such as barrier heights (\(A_{0M}\) or \(A\)) and interfacial energy coefficients (\(\beta_{0M}\) or \(\beta_{ij}\)), given by \(\delta_{0M} = \sqrt{8\beta_{0M}/A_{0M}}\) and \(\beta_{0M} = \gamma_{0M}\delta_{0M}\) (see [16, 29]), we obtain \(A_{0M} = 1800\) MPa, \(A = 720\) MPa, \(\beta_{0M} = 4 \times 10^{-10}\) N, and \(\beta_{ij} = 1 \times 10^{-11}\) N. The temperature
Figure 5. The plots for $\eta_{eq}$ for twinned martensite (Type-II; see Table 2) in NiTi samples (shown in $\Omega_0$) with sizes (i) $w = 15$ nm, $l = 25$ nm, (ii) $w = 20$ nm, $l = 35$ nm, and (iii) $w = 30$ nm, $l = 50$ nm are presented.

Table 3. Elastic constants for CuAlNi in GPa [6].

<table>
<thead>
<tr>
<th>Phase</th>
<th>$C_{11}$</th>
<th>$C_{22}$</th>
<th>$C_{33}$</th>
<th>$C_{44}$</th>
<th>$C_{55}$</th>
<th>$C_{66}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{23}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>142</td>
<td>142</td>
<td>142</td>
<td>96</td>
<td>96</td>
<td>126</td>
<td>126</td>
<td>126</td>
<td></td>
</tr>
<tr>
<td>$M_1$</td>
<td>189</td>
<td>141</td>
<td>141</td>
<td>141</td>
<td>54.9</td>
<td>62.6</td>
<td>124</td>
<td>115</td>
<td></td>
</tr>
<tr>
<td>$M_3$</td>
<td>189</td>
<td>205</td>
<td>141</td>
<td>54.9</td>
<td>19.7</td>
<td>45.5</td>
<td>124</td>
<td>115</td>
<td></td>
</tr>
</tbody>
</table>

Table 4. Elastic constants for NiTi in GPa [9].

<table>
<thead>
<tr>
<th>Phase</th>
<th>$C_{11}$</th>
<th>$C_{22}$</th>
<th>$C_{33}$</th>
<th>$C_{44}$</th>
<th>$C_{55}$</th>
<th>$C_{66}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{23}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>169</td>
<td>169</td>
<td>169</td>
<td>40</td>
<td>40</td>
<td>138</td>
<td>138</td>
<td>138</td>
<td></td>
</tr>
<tr>
<td>$M_1$</td>
<td>223</td>
<td>241</td>
<td>200</td>
<td>45</td>
<td>40</td>
<td>129</td>
<td>125</td>
<td>125</td>
<td></td>
</tr>
<tr>
<td>$M_5$</td>
<td>241</td>
<td>223</td>
<td>200</td>
<td>45</td>
<td>90</td>
<td>90</td>
<td>90</td>
<td>90</td>
<td></td>
</tr>
</tbody>
</table>

is assumed to be constant in space and time and it is equal to the thermodynamic equilibrium temperature, i.e., $\theta = \theta_0$. The thermal energy is, thus, trivial in both cases; see (14). The mobilities of the interfaces are $L_{0M} = L_{ij} = 2600$ (Pa-s)$^{-1}$ [16]. The elastic constants for A and variant $M_1$ for both CuAlNi [6] and NiTi [9] alloys are listed in the first two rows of Tables 3 and 4, respectively. Note that the constants for the other variant $M_j$ are obtained using

$$\hat{C}_{(\alpha)ABCD} = R_{AE}^\alpha R_{BF}^\alpha R_{CG}^\alpha R_{DH}^\alpha \hat{C}_{(\alpha)EFGH},$$

(84)

where $R^\alpha$ is the symmetry-related rotation tensors such that $U_{ij} = R^\alpha \cdot U_{ij} \cdot R^{\top}$ [36]. The symmetry rotations for CuAlNi alloy (when $i = 1$ and $j = 3$) and NiTi alloy (when $i = 1$ and $j = 5$) are

$$R^\alpha_{CuAlNi} = \begin{bmatrix} 0 & -1 & 0 \\ -1 & 0 & 0 \\ 0 & 0 & -1 \end{bmatrix} \quad \text{and} \quad R^\alpha_{NiTi} = \begin{bmatrix} -1 & 0 & 0 \\ 0 & 0 & -1 \\ 0 & -1 & 0 \end{bmatrix},$$

(85)

respectively. The elastic moduli for $j$-th variants of CuAlNi and NiTi alloys are also listed in the last rows of Tables 3 and 4, respectively.
6.3. Numerical solution

For finite element computations we use the same computational algorithm as presented in [29], however, compute the derivatives of $U_t$ utilizing the procedure discussed in Section 3; see the Appendix for the list of finite element equations. The stationary twinned microstructures in CuAlNi and NiTi crystals are shown in Figures 4 and 5, respectively, within the reference configuration $\Omega_0$ in samples of varying size. A composite order parameter $\eta_{eq} = 2\eta_0(\eta_1 - 0.5)$ is plotted in these figures to display the resultant microstructures. Obviously, the dark red color with $\eta_{eq} = 1$ denotes variant $M_i$, the dark blue color with $\eta_{eq} = -1$ denotes variant $M_j$, and green color with $\eta_{eq} = 0$ denotes either A (due to $\eta_0 = 0$) or a particle from the $M_i$–$M_j$ interface (due to $\eta_1 = 0.5$). The solutions are shown in samples of increasing sizes. Almost the entire sample contains martensite and there is very small fraction of austenite, as desired. Laminated microstructures are obtained for both crystals where the normals to martensitic plates are aligned in $e_1$ direction in $\Omega_0$, and the maximum deviation was found to be less than $1^\circ$ for both the cases. We have also calculated the volume fraction of variant $M_i$ in the twinned microstructures by considering a line passing through the middle of the sample and using the relation $\xi_{FE} = \ell_2/(\ell_1 + \ell_2)$, where $\ell_1$ and $\ell_2$ are the length of the segments on that line where $0.95 \leq \eta_1 \leq 1$ (i.e., $M_i$) and $0 \leq \eta_1 \leq 0.05$ (i.e., $M_j$), respectively. For both the crystals $\xi_{FE}$ were calculated for the largest samples shown in Figures 4 and 5 and were obtained approximately as 0.26 and 0.29, respectively (compare them with the analytical solutions listed in Tables 1 and 2, respectively). The differences between the analytical and numerical solutions would vanish if we consider much larger samples. The number of twin plates $N_{tw}$ for both the crystals is approximately proportional to $\sqrt{w}$, i.e., $N_{tw} \sim \sqrt{w}$ (see [5, 6, 29, 37–39]). Similar twinned microstructures were presented in [29], but for a case where the Bain tensors are commutative (cubic A and tetragonal variants). Notably, nanoscale twin microstructures (Type-I and II) have been observed experimentally also both in CuAlNi [1, 40, 41] and in NiTi [42, 43] SMAs.

7. Conclusions

A theoretical and finite element procedure to deal with an exponential-logarithmic KM for transformation stretch tensor in a thermodynamically consistent multiphase phase field approach for MTs has been established. The present treatment is general and can be used for any kind of MT and for any number of variants. This transformation rule yields isochoric variant–variant transformations, which is a plausible condition from the point of view of crystallographic theory of MTs. The first and second derivatives of the transformation stretch tensor with respect to the order parameters have been determined, which are used to (i) analyze the transformation work in the Ginzburg–Landau equations, (ii) establish the thermodynamic instability criteria for homogeneous phases, and (iii) determine the weak forms of the Ginzburg–Landau equations and their linearizations. Change of anisotropic elastic properties during austenite–martensitic variants and variant–variant transformations has been taken into account. Using the present formulation, laminated twinned microstructures have been simulated using a finite element method for CuAlNi alloy (cubic to orthorhombic) and NiTi alloy (cubic to monoclinic-I). The microstructures are consistent with the crystallographic theory, which validates our treatment. Our theory can be extended for more complex exponential-logarithmic types of KMs for the transformation stretches. For example, Levitas [26] recently developed a more advanced phase field model for a two-phase system and the theory should be extended for a multivariant system. In that case, the present formulation can be used.

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The order parameters are then updated using \( \eta \) matrix corresponding to the order parameter.

\[ \eta_{\text{matrix corresponding to the order parameter}} \]

[39] Levitas, VI, and Javanbakht, M. Phase-field approach to martensitic phase transformations: Effect of martensite–martensite


[43] Chow, EGW. On the stress–strain relations for all crystal systems.


[47] Levitas, VI, and Javanbakht, M. Phase-field approach to martensitic phase transformations: Effect of martensite–martensite


for all \( k = 0, 1, \ldots, N - 1 \), where \( c_1, c_2, \) and \( c_3 \) are constants such that for backward time difference scheme of order one, \( c_1 = 1, c_2 = -1, \) and \( c_3 = 0 \), and for backward time difference scheme of order two, \( c_1 = 1.5, c_2 = -2, \) and \( c_3 = 0.5; \) \( \Delta t_n \) is the time step yielding convergence of all the order parameters; \( n_i \) is the number of degrees of freedom for \( \eta_i; \) \( n_{el} \) is the number of grid points for each finite element; \( n_{el} \) is the total number of finite elements in the entire \( \Omega_0 \) such that \( \Omega_0 \approx \cup_{\eta_{el}^0} \Omega_0^0 \) (see [32]); \( N_i \) is the shape function; \( L^0_i = L_{0,i}^0 \beta_{0,i} \), \( L^0_k = \sum_{m=1, \neq k}^{N} L_{k,m} \beta_{k,m}, \) \( h^0_k = 1, \) and \( \bar{\phi}(a_\beta, a_\alpha, \eta_0) \) for all \( k = 1, 2, \ldots, N - 1; \) gradient of the shape function \( \nabla N_i \) and the standard finite element \( \mathbf{B} \) are given by [32]

\[
\nabla N_i = \begin{bmatrix} N_{i,1} \\ N_{i,2} \\ N_{i,3} \end{bmatrix} \quad \text{and} \quad \mathbf{B}_i = \begin{bmatrix} N_{i,1} & 0 & 0 \\ 0 & N_{i,2} & 0 \\ 0 & 0 & N_{i,3} \end{bmatrix}, \quad (88)
\]

where the commas denote spatial derivatives with respect to the respective coordinates in \( \Omega \). The symbol \( \cup \) in (87) denotes the standard assembly operation. The functions \( f_0 \) and \( f_i \) (for \( i = 1, 2, \ldots, N \)) appearing in (87) are [29]

\[
f_0^m = L_{0,i}^0 \left[ - (J^n F^{-1} - \sigma^n e \cdot F^n - J^n \psi^{en} \mathbf{I}) : \mathbf{Y}_0 + J^n \frac{\partial \psi^n c}{\partial \eta_0} \right]_{F_e} + \rho_0 (6 \eta^n_0 - 6 \eta^n_{0,2}) \Delta \psi^n + J^n \rho_0 \bar{A} \times \\
\left( \sum_{i=1}^{\eta_1} \sum_{j=1}^{\eta_2} \eta_i^2 \eta_j^2 + \eta_N^2 \sum_{i=1}^{\eta_1} \eta_i^2 \right) \frac{\partial \phi(a_\beta, \eta_0^n)}{\partial \eta_0} + J^n \rho_0 (A_0 M (\theta) + (a_0 - 3) \Delta \psi^n (\theta)) (2 \eta^n_0 - 6 \eta^n_{0,2} + \\
4 \eta^n_0^3) + \frac{8}{J^n} \frac{\partial \bar{\phi}(a_\beta, a_\alpha, \eta_0^n)}{\partial \eta_0} \left( \sum_{i=1}^{\eta_1} \sum_{j=1, \neq i}^{\eta_2} \beta_j |\nabla \eta_i^n - \nabla \eta_j^n|^2 + \sum_{i=1}^{\eta_1} \beta_i |\nabla \eta_i^n - \nabla \eta_N^n|^2 \right), \quad (89)
\]

\[
f_i^m(\eta_j^n, \nabla \eta_j^n, \nabla^2 \eta_j^n) = - \sum_{m=1, \neq i}^{N} L_{im} (\bar{X}_{im}^{(loc)} + \bar{X}_{im}^{(1)}) \quad \text{for all} \ j = 0, 1, \ldots, N - 1; \quad (90)
\]

where

\[
\bar{X}_{im}^{(loc)} = (J^n F^{-1} - \sigma^n e \cdot F^n - J^n \psi^{en} \mathbf{I}) : (\mathbf{Y}_i^n - \mathbf{Y}_m^n) - J^n \left( \frac{\partial \psi^n c}{\partial \eta_i} \right)_{F_e} \left( - \frac{\partial \psi^{en}}{\partial \eta_m} \right)_{F_e} - 2 J^n \rho_0 \bar{A} \left( \sum_{j=1, \neq i}^{\eta_1} \eta_j^n \eta_j^n - \sum_{j=1, \neq i}^{\eta_1} \eta_j^n \eta_m^n + \eta_j^n \eta_j^n - \eta_N^n \eta_j^n \right) \psi(a_\beta, \eta_0^n) - 2 \rho_0 \sum_{j=1, \neq i}^{\eta_1} \psi(a_\beta, \eta_0^n) - \sum_{j=1, \neq m, \neq i}^{\eta_1} K_{ij}(\eta_j^n + \eta_N^n - 1)(2 \eta_j^n + \eta_j^n - 1) \times \eta_j^n \eta_j^n - \\
2 \rho_0 \sum_{j=1, \neq m, \neq i}^{\eta_1} K_{ij}(\eta_j^n + \eta_N^n - 1)(2 \eta_j^n + \eta_j^n - 1) \eta_j^n \eta_j^n - K_{ij}(\eta_j^n + \eta_N^n - 1)(2 \eta_j^n + \eta_j^n - 1) \eta_j^n \eta_j^n \eta_j^n - \\
2 \rho_0 \sum_{j=1, \neq m, \neq i}^{\eta_1} K_{ij}(\eta_j^n + \eta_N^n - 1)(2 \eta_j^n + \eta_j^n - 1) \eta_j^n - K_{ij}(\eta_j^n + \eta_N^n - 1)(2 \eta_j^n + \eta_j^n - 1) \eta_j^n \eta_j^n \eta_j^n; \quad (91)
\]

\[
\bar{X}_{im}^{(2)} = \bar{X}_{im}^{(2)} \quad \text{for all} \ i, m = 1, 2, \ldots, N; \quad (92)
\]
\[ \bar{X}^{(1)}_{im} = \nabla_0 \cdot \left[ \frac{\tilde{\omega}}{4 } F^{m-1} \cdot \left( \begin{array}{c} \beta_{jN} \sum_{j=1, j\neq i}^{N-1} \nabla \eta_j^m - \sum_{j=1, j\neq i}^{N-1} \beta_j \nabla \eta_j^m - \sum_{j=1, j\neq m, j}^{N-1} \beta_{mj} \nabla \eta_m^j + \sum_{j=1, j\neq m, j}^{N-1} \beta_{mj} \nabla \eta_j^m - \\ \beta_{mn} \nabla \eta_m^m - \beta_{mn} \sum_{j=1, j\neq i}^{N-1} \nabla \eta_j^m \end{array} \right) \right]; \] (93)

\[ \bar{X}^{(2)}_{im} = \nabla_0 \cdot \left[ \frac{\tilde{\omega}}{4 } F^{m-1} \cdot \nabla \eta_i^m \right] \quad \text{with} \quad \bar{b}_{im} = \sum_{j=1, j\neq i}^{N-1} \beta_{ij} + \beta_{mi} + 2 \beta_{IN} - \beta_{mN}; \] (94)

and \( Y^n_k = \frac{\partial \bar{U}^n_i}{\partial \eta^n_k} \) for all \( k = 0, 1, \ldots, N \). The expressions for \( \partial f^n_0 / \partial \eta^n_0 \) and \( \partial f^n_i / \partial \eta^n_i \) appearing in (87)_4 are given by

\[
\frac{\partial f^n_0}{\partial \eta^n_0} = \mathcal{L}_0 M \left[ - (J^n F^{m-1} \cdot \sigma_c \cdot F^n - J^n \psi^{en} I) \cdot \left( \bar{U}^{n-1} - \frac{\partial \bar{U}^n}{\partial \eta^n_0} - \frac{\partial \bar{P}^{en}}{\partial \eta^n_0} \right) \cdot F^n : Y^0^n \right. \\
\left. + J^n \bar{U}^n_i \psi^n_c \text{tr}(Y^n_i^2) + J^n \bar{U}^n_i \left( \frac{\partial \psi^n}{\partial \eta^n_0} \right) \text{tr}(Y^n_i) + \bar{U}^n_i \left( \frac{\partial \psi^n}{\partial \eta^n_0} \right) \text{tr}(Y^n_i F_i) \right) + \rho_0 (6 - 12 \eta^n_0) \Delta \psi^n_0 \\
\left. + J^n \rho_0 \tilde{A} \left( \sum_{i=1}^{N-2} \sum_{j=i+1}^{N-1} \eta^n_i^2 + \sum_{i=1}^{N-1} \eta^n_i \eta^n_j \right) + J^n \frac{\partial^2 \varphi(a_b, a_c, \eta^n_0)}{\partial \eta^n_0} \bar{U}^{n-1} \left( \sum_{i=1}^{N-2} \sum_{j=i+1}^{N-1} \beta_{ij} \nabla \eta_i^n - \nabla \eta_j^n \right)^2 \right. \\
\left. + \sum_{i=1}^{N-1} \beta_{IN} \nabla \eta_i^n + \sum_{j=1}^{N-1} \eta_j^n \right)^2 \right], \] (95)

and

\[
\frac{\partial f^n_i (\eta^n_0, \eta^n_1, \ldots, \eta^n_{N-1})}{\partial \eta^n_i} \bigg|_F = - \sum_{m=1, m\neq i}^{N} \mathcal{L}_{im} \frac{\partial \bar{X}^{\text{loc}}}{\partial \eta^n_i} \bigg|_F, \] (96)

where

\[
\frac{\partial \bar{X}^{\text{loc}}}{\partial \eta^n_i} = (J^n F^{n-1} \cdot \sigma_c \cdot F^n - J^n \psi^{en} I) : \bar{U}^{n-1} \left[ \frac{\partial \bar{U}^n}{\partial \eta^n_i} \left( \frac{\partial \bar{U}^n}{\partial \eta^n_i} - \frac{\partial \bar{U}^n_i}{\partial \eta^n_i} \right) \right] F_i + \frac{\partial \bar{P}^{en}}{\partial \eta^n_i} \cdot F_i \cdot Y^i_0 - Y^m_0 \right) \\
\left. + J^n \bar{U}^n_i \left( \frac{\partial \psi^n}{\partial \eta^n_i} \right) \text{tr}(Y^n_i F_i) \right) + \rho_0 (6 - 12 \eta^n_0) \Delta \psi^n_0 \\
\left. + \sum_{i=1}^{N-1} \beta_{IN} \nabla \eta_i^n + \sum_{j=1}^{N-1} \eta_j^n \right)^2 \right], \] (95)

and

\[
\frac{\partial f^n_i (\eta^n_0, \eta^n_1, \ldots, \eta^n_{N-1})}{\partial \eta^n_i} \bigg|_F = - \sum_{m=1, m\neq i}^{N} \mathcal{L}_{im} \frac{\partial \bar{X}^{\text{loc}}}{\partial \eta^n_i} \bigg|_F, \] (96)

where
\[-2\rho_0 \sum_{j=1, \neq i}^{N-1} \left\{ K_j (6\eta_j^n + 2\eta_j^n - 2\eta_j^n + 1) + K_m (4\eta_j^n + 4\eta_m^n) \right\} \]
\[+ 9\eta_i^n \eta_m^n - 6\eta_i^n - 6\eta_m^n - 2\eta_i^n + 2\eta_i^n - \eta_i^n \eta_m^n - K_{mN} (4\eta_i^n + 4\eta_m^n - 6\eta_i^n - 6\eta_m^n + 2) \eta_i^n \eta_m^n \eta_N^n\]
\[= K_{mN} (4\eta_i^n + 4\eta_m^n - 6\eta_i^n - 6\eta_m^n + 2) \eta_i^n \eta_m^n \eta_N^n. \quad (97)\]

The term $Y_0^n$ in (89) and (95) is determined using (36) and (42), and the terms $Y_k^n$ for $k = 1, \ldots, N$ in (91) and (97) are determined using (47). The term related to the second derivative of $U$, with respect to $\eta_0$ in (95) is determined using (51), (42), and (58), and the terms related to the second derivative of $U$, with respect to $\eta_i$ in (97) are calculated using (61), (45), and (59). For a two-variant system the simplified forms, i.e., (62) and (45) should be used. The expressions for some of the terms in (97) are given by (see [29])

\[
\frac{\partial P_{en}^T}{\partial \eta_i^n} \cdot F^n = \left[ J_{tr} \left( U_{i}^{n-1} \cdot \frac{\partial U_{i}^n}{\partial \eta_i^n} \right) \cdot \hat{S}_e \cdot U_{i}^{-T} - 2 \text{sym}(Z_e^n) \right] \\
+ J_{tr} \cdot \frac{\partial \hat{S}_e}{\partial \eta_i^n} \cdot \hat{S}_e \cdot U_{i}^{-T} \cdot C^n; \quad (98) \\
Z_i^n = S^n \cdot \frac{\partial U_{i}^n}{\partial \eta_i^n} \cdot U_{i}^{-T}; \quad \frac{\partial \hat{S}_e}{\partial \eta_i^n} \cdot \hat{S}_e \cdot \frac{\partial U_{i}^n}{\partial \eta_i^n} \cdot U_{i}^{-T} = \frac{\partial \hat{C}_e}{\partial \eta_i^n} \cdot E_e^n + \frac{\partial E_e^n}{\partial \eta_i^n} \cdot C_e^n; \quad (99) \\
\frac{\partial E_e^n}{\partial \eta_i^n} = - \text{sym} \left( C_e^n \cdot \frac{\partial \hat{U}_i^n}{\partial \eta_i^n} \cdot \hat{U}_i^{-1} \right); \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad (100) \\
\frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad (101) \\
\text{and} \quad \frac{\partial}{\partial \eta_k^n} \left( \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} \right) = 0.5E_e^n; \quad \frac{\partial}{\partial \eta_k^n} \left( \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} \right) = 0.5E_e^n; \quad \frac{\partial \psi^n}{\partial \eta_i^n} \bigg|_{F_e} = 0.5E_e^n; \quad (102)
\]

Note that the finite element procedure for phase field equations discussed in this appendix can be applied for any MT with any number of variants.