Non-affine fields in solid–solid transformations: the structure and stability of a product droplet

Arya Paul 1, Surajit Sengupta 2,3 and Madan Rao 4,5

1 Indian National Centre for Ocean Information Sciences, ‘Ocean Valley’, Pragathi Nagar, Nizampet, Hyderabad-500090, India
2 TIFR Centre for Interdisciplinary Sciences, 21 Brundavan Colony, Narsingi, Hyderabad-500075, India
3 Centre for Advanced Materials, Indian Association for the Cultivation of Science, 2A & 2B Raja S C Mullick Road, Jadavpur, Kolkata-700032, India
4 Raman Research Institute, C V Raman Avenue, Sadashivnagar, Bangalore-560080, India
5 National Centre for Biological Sciences, Tata Institute of Fundamental Research, Bellary Road, Bangalore 560065, India

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Abstract
We describe the microstructure, morphology, and dynamics of growth of a droplet of martensite nucleating in a parent austenite during a solid–solid transformation, using a Landau theory written in terms of both conventional affine elastic deformations and non-affine deformations. Non-affineness, \( \phi \), serves as a source of strain incompatibility and screens long-ranged elastic interactions. It is produced wherever the local stress exceeds a threshold and anneals diffusively thereafter. Using a variational calculation, we find three types of stable solution (labeled I, II, and III) for the structure of the product droplet, depending on the stress threshold and the scaled mobilities of \( \phi \) parallel and perpendicular to the parent–product interface. The profile of the non-affine field \( \phi \) is different in these three solutions: I is characterized by a vanishingly small \( \phi \), II admits large values of \( \phi \) localized in regions of high stress within the parent–product interface, and III is a structure in which \( \phi \) completely wets the parent–product interface. The width \( l \) and size \( W \) of the twins follow the relation \( l \propto \sqrt{W} \) in solution I; this relation does not hold for II or III. We obtain a dynamical phase diagram featuring these solutions, and argue that they represent specific solid-state microstructures.

(Some figures may appear in colour only in the online journal)

1. Introduction

Unlike solidification [1], where a crystal nucleates and grows from a melt, nucleation of a solid within another solid may encounter specific complications arising from the crystallographic incompatibility of the parent and product solids. In most cases, it is geometrically impossible to fit a nucleus of the homogeneous product within the parent (austenite) matrix without gaps. To maintain continuity, therefore, the system needs to adapt, which it does using a variety of viable strategies. A common strategy is the simultaneous nucleation of several crystallographically equivalent and degenerate variants of the product in the form of a twinned martensite [2], whose microstructure contains an alternate array of elastically coupled variants or twins. At the martensite–austenite interface, crystallographic incompatibility is accommodated on the average. This is just one of the many possibilities of accommodating incompatibility, and there is an extensive literature on experiments [1–3], microscopic and coarse-grained theory [4–12], and coarse-grained and atomistic simulations [9, 13–17] of such martensites.

On the other hand, there exist other kinds of martensitic microstructures that incorporate defects, such as dislocation arrays [18], that also go to make the interface compatible on the average. The incompatibility problem may also...
be resolved by apparent diffusion of particles, as is the case in ferritic microstructures which involve no twins and are produced at higher temperatures where particle trajectories during transformation are disorderly and resemble diffusion [1, 19]. In a realistic situation, multiple modes of accommodating interfacial strain compatibility must be present. Though there have been attempts to include the effect of isolated defects within an otherwise strain-only approach [20], a complete theory of microstructure, capable of describing dislocated and twinned as well as ferritic microstructures, has not yet been realized. Possibly, one needs to incorporate additional degrees of freedom, other than strains, in order to describe the multiple microstructures within a single framework. What are these non-elastic variables?

In [21], we show that atomic displacements, when coarse grained, can naturally be projected onto mutually orthogonal subspaces corresponding to affine and non-affine deformations of a reference lattice. The affine subspace represents the elastic deformation of the reference volume, while the non-affine subspace contains atomic displacements within the reference volume corresponding to short-ranged atomic ‘shuffles’ which cannot be described as strains. We parameterize these displacements by a phenomenological field \( \phi \), whose dynamics determines microstructure selection. Indeed, our detailed molecular dynamics simulations of early time nucleation in a solid undergoing a (square \( \rightarrow \) rhombic) structural transition [14, 17] show that (1) non-affine strains are generated during the transition and are localized in regions we call non-affine zones (NAZs), (2) the transformation is driven by a small number of active particles [17] undergoing rearrangements within NAZs, (3) the NAZs lie close to regions where the non-order parameter (NOP) strains are larger than a threshold, and (4) the dynamics of the NAZs determine microstructure selection such that a martensite forms when the NAZs are localized at the interface while a ferritic microstructure forms when the NAZs spread over large scales, as shown in figure 1.

We have previously described microstructure selection in solid-state transformations, using a Landau theory that goes beyond strain-only approaches by including both elastic and non-affine degrees of freedom, \( \phi \), that explicitly violates compatibility [11], at least locally. We showed that our theory could describe both martensitic and ferritic microstructures, and derived conditions when one or the other obtained. In this paper, we describe a droplet theory [7, 12] to study the structure and growth of a small region of martensite embedded in the matrix of the parent austenite phase. A simple but realistic variational ansatz for the order parameter and non-affine fields enables us to derive the optimized free energy of this droplet and to show that there are multiple solutions within specific ranges of system-dependent parameters, representing possible morphologies.

The plan of the paper is as follows. In section 2, we discuss the dynamical equations for the elastic and non-affine variables. In section 3, we show that the equations of motion admit a Lyapunov functional, which we use to explore the space of solutions. In section 4, we describe the droplet variational ansatz of martensite embedded in the austenite phase. In section 5, we show that in the space of our system-dependent external parameters the Lyapunov functional has multiple minima which may be related to structurally and dynamically distinct microstructures. We discuss our results in section 6 in the light of known results, and conclude.

2. Dynamical equations for elastic and non-affine variables

In this section, we will paraphrase our earlier derivation [11] of the dynamical equations for the elastic strains and \( \phi \) during a structural transition. While our theory is general, motivated by our earlier work [11, 14, 17], we describe a formulation based on a square to rhombic transformation. The square to rhombic transition is a special case of the more general transition from square to oblique two-dimensional Bravais lattices. The affine shear strain \( e_3 = \partial u_x/\partial y + \partial u_y/\partial x \) and the affine deviatoric strain \( e_2 = \partial u_x/\partial x - \partial u_y/\partial y \) are the order parameters (OPs) for the square to oblique transition [11, 14], where \( u_i (i = x, y) \) is the displacement field describing deformations from the (square) reference lattice. The equilibrium value of \( e_2 \) for the rhombic lattice is zero. The volumetric affine strain \( e_1^a = \partial u_x/\partial x + \partial u_y/\partial y \), which does not lead to a change of local symmetry, is a non-order parameter (NOP) strain [8, 10, 11, 14], and it relaxes to its local steady-state value faster than the OP strains. We will therefore ignore the dynamics of the NOP strain, replacing it by its steady-state value determined by (or ‘slaved’ to) the instantaneous values of the OP strains [11].
We incorporate non-affine fields in our description by assuming that the total NOP strain may be decomposed into an affine part $e_i^A$ and a non-affine part $\phi$, i.e., $e_i = e_i^A + \phi$. This decomposition is analogous (though not equivalent!) to the decomposition of total strain into elastic and plastic parts, routinely made in the continuum theories of plasticity [23, 24]. In contrast to most plasticity theories, however, $\phi$ is generated in the volumetric sector, an unusual requirement, because deformation in solids is known to remain elastic for large compressional or dilatational stresses, with plasticity being produced mainly under shear deformation, $e_3$ [21]. This requirement can be readily understood if we remind ourselves of the following: (1) the shear strain $e_3$, which is the broken-symmetry OP strain, is large within the martensite droplet and decays to zero in the parent phase, (2) gradients of $e_3$ generate large NOP ($e_i^A$) strain at the interface in order to satisfy elastic compatibility, and (3) the combined presence of $e_3$ and $e_1$ makes the interfacial region susceptible to local rearrangements of particles; this generates NAZs [14] in regions of high NOP strain present at the moving parent–product front. The topological rearrangements, now parameterized by $\phi$, occur when the local conjugate stress $\sigma_1$($e_3$) exceeds an appropriate local threshold $\sigma_{1c}$($e_3$) or yield criterion. Note that both the conjugate stress and the threshold which determine $\phi$ are finally **functions** of $e_3$.

The Lagrangian is given by

$$\mathcal{L} = \int \left( \frac{\rho}{2} \left[ (\dot{x}_1^2 + \dot{y}_1^2) \right] \right) \mathrm{d}x \mathrm{d}y - \mathcal{F},$$

where the first term is the kinetic energy with $\rho$ the mass density, which may be set to unity without loss of generality. The free energy functional $\mathcal{F}$ depends on the structural transition being considered, and for the two-dimensional square to rhombic transition one may set

$$\mathcal{F} = \frac{1}{2} \int \mathrm{d}x \mathrm{d}y \left[ a_1 (e_1^A + \phi)^2 + a_2 e_2^2 + a_3 e_3^2 - \frac{1}{2} b_3 e_3^4 \right]$$

$$+ \frac{1}{2} d_3 e_3^6 + c_1 (\nabla e_1^A)^2 + c_2 (\nabla e_2)^2 + c_3 (\nabla e_3)^2].$$

(2)

The parameters $a_1$, $a_2$, and $a_3$ are elastic constants appropriate for the square (parent) phase, and $c_1$, $c_2$, and $c_3$ are related to strain correlation lengths. For the range of parameters considered, $\mathcal{F}$ has three minima at $e_3 = 0$ representing the square, parent, or austenite, and $e_3 = \pm e_0 = \pm (b_3/2d_3 + \sqrt{b_3^2 - 4a_3d_3/2d_3})^{1/2}$, the two degenerate variants of the product rhombic phase. The connection with external control parameters such as temperature ($T$) is, as usual, through the temperature dependence of the coefficients.

To obtain the equation of motion in the displacement fields, we need to solve the Euler Lagrange equation,

$$\frac{\partial \mathcal{L}}{\partial \dot{u}_i} - \frac{\partial \mathcal{L}}{\partial u_i} = - \frac{\partial \mathcal{R}}{\partial u_i}. \quad (3)$$

The Rayleigh dissipation functional [25] $\mathcal{R} = \frac{1}{2} \int \left[ \gamma_1(e_1^A)^2 + \gamma_2 e_2^2 + \gamma_3 e_3^2 \right] \mathrm{d}x \mathrm{d}y$, where the coefficients $\gamma_1$, $\gamma_2$, and $\gamma_3$ are the corresponding ‘viscosities’.

We take $e_i^A$ to be a slaved variable which reaches the steady state much faster than the OP strains. The equations of motion for the components of the strain can then be written as

$$\nabla^2 e_1^A = \frac{q_{13}}{\rho} \frac{\partial^2 e_3}{\partial x \partial y} - \nabla^2 \phi, \quad (4)$$

$$\dot{e}_3 = \nabla^2 \left( a_3 e_3 - b_3 e_3^3 + d_3 e_3^5 - c_3 \nabla^2 e_3 \right) - 2c_1 \frac{\partial^2 e_1^A}{\partial x \partial y} + \gamma_3 e_3 \right) + \frac{2 \partial^2}{\partial x \partial y} \left[ a_1 (e_1^A + \phi) \right]. \quad (5)$$

where $q_{13} = (a_2 - a_3)/(a_1 + a_2)$. The OP strain $e_2$ has been eliminated using a modified St Venant condition,

$$\nabla^2 e_1^A - \frac{\partial^2}{\partial x^2} - \frac{\partial^2}{\partial y^2} e_2 - 2\frac{\partial^2 e_3}{\partial x \partial y} = - \nabla^2 \phi, \quad (6)$$

where the strain incompatibility is completely determined by gradients of $\phi$. To obtain a closed set of equations, we need to prescribe a dynamics for the non-affine field $\phi$ for which we propose the following **phenomenological** equation of motion:

$$\dot{\phi} = - \int_{-\infty}^{t'} \frac{\sigma_1}{h_1} H(\sigma_1 - \sigma_{1c}) K(t, t') \mathrm{d}t'$$

$$+ c_1 (\nabla^2 \phi) + c_2 (\nabla^2 \phi). \quad (7)$$

The dynamics (7) incorporates two dynamical processes. The first term accounts for the fact that $\phi$ is produced at a rate given by $h_1^{-1}$, in response to the stress $\sigma_1 = \partial \mathcal{F}/\partial e_1^A = a_1 (e_1^A + \phi)$, locally exceeding a fixed threshold $\sigma_{1c}$ independent of any state variable (figure 2(b)); $H(x)$ is the unit step function. In general, this response may be delayed, through a response kernel $K(t, t')$, where $t$, $t'$ are times. This is the source for NAZs (figure 2(b)), and it goes towards reducing the local stress $\sigma_1$. Second, as the transformation proceeds, $\phi$ produced at regions of large $\sigma_1$ (figure 2(c)) at the austenite–martensite interface needs to anneal, which is accounted for by the next two ‘diffusive’ terms. Since the nucleation of the martensite droplet spontaneously defines an interface or habit plane, the diffusion coefficients $c_1$, $c_2$, and $c_3$ in (7) may, in principle, be obtained from careful computer simulations of detailed atomistic models.

3. The Lyapunov functional

Equations (4) and (5) together with (7) describe the dynamics of $\epsilon_i$ ($i = 1, 3$) and $\phi$ completely. We are interested in the steady-state solutions of these equations. While the equations of motion of the strains are derived from the free energy functional (2), the non-affine field follows a phenomenological dynamics (7) not derivable from this free energy. In this section, we show that, nevertheless, one may be able to define a Lyapunov functional whose minimizers correspond to the steady states of (4), (5), and (7), thereby reducing a dynamical problem to one of optimization.

We begin by introducing the simplification $K(t, t') = \delta(t - t')$ in (7), which amounts to assuming that the system
respects to a stress overshoot by instantaneously generating a \( \phi \) field, and obtain
\[
\dot{\phi} = \frac{\sigma_1}{h_1} H (|\sigma_1| - \sigma_{1c}) + c_x (\frac{\partial^2 \phi}{\partial x^2}) + c_y (\frac{\partial^2 \phi}{\partial y^2}).
\] (8)
Equation (8) needs to be solved using the initial/boundary conditions \( \phi(t = 0) = 0 \) and \( \phi \rightarrow 0 \) at large distances and reduces to two steady-state situations depending on the magnitude of \( \sigma_1 \). For \( |\sigma_1| > \sigma_{1c} \), we have
\[
-\frac{a_1 (e_A^1 + \phi)}{h_1} = c_x (\frac{\partial^2 \phi}{\partial x^2}) + c_y (\frac{\partial^2 \phi}{\partial y^2}),
\] (9)
while for \( |\sigma_1| < \sigma_{1c} \) we obtain the anisotropic diffusion equation,
\[
c_x (\frac{\partial^2 \phi}{\partial x^2}) + c_y (\frac{\partial^2 \phi}{\partial y^2}) = 0.
\] (10)
These may be combined in the form of a single equation,
\[
c_x (\frac{\partial^2 \phi}{\partial x^2}) + c_y (\frac{\partial^2 \phi}{\partial y^2}) + a_1 (e_A^1 + \phi)f(\sigma_{1c}, \sigma_1) = 0,
\] (11)
where \( f(\sigma_{1c}, \sigma_1) \) interpolates smoothly between the two limits (9) and (10). A convenient choice is \( f = [1 - \exp(-|\sigma_1|/\sigma_{1c})] \); thus
\[
c_x (\frac{\partial^2 \phi}{\partial x^2}) + c_y (\frac{\partial^2 \phi}{\partial y^2}) + a_1 (e_A^1 + \phi) \int \left\{ 1 - \exp \left( -\frac{|\sigma_1|}{\sigma_{1c}} \right) \right\} = 0,
\] (12)
where we have absorbed the constant \( h_1 \) into the definition of the dynamical parameters \( c_x \) and \( c_y \) quantifying the ratios of the times it takes for \( \phi \) to form to that it takes to spread in the \( x \) and \( y \) directions, respectively. One can show by direct plotting that the chosen representation for the step function, though strictly accurate for small \( \sigma_{1c} \), reproduces the expected qualitative behavior even if \( \sigma_{1c} \) is larger. We emphasize that this choice is not a small-\( \sigma_{1c} \) approximation; in principle, we could have chosen a different smooth representation for the step function, accurate over a larger range of \( \sigma_{1c} \).

We now seek a Lyapunov functional whose minimizers would automatically generate the steady-state solutions for \( \epsilon_i \) and \( \phi \). It is straightforward to show that this corresponds to
\[
\mathcal{F}_L = \frac{1}{2} \int \left\{ a_1 (\phi^2 + 2e_A^1 \phi) + 2 \left( \frac{\sigma_{1c}}{a_1} \right) (\sigma_{1c} + \sigma_1) e^{-|\sigma_1|/\sigma_{1c}} \right\} dx dy + \cdots,
\] (13)
where the dots represent terms which are independent of \( \phi \). Collecting terms, we obtain
\[
\mathcal{F}_L = \mathcal{F} + \frac{1}{2} \int \left\{ \frac{2\sigma_{1c}}{a_1} (\sigma_{1c} + \sigma_1) e^{-|\sigma_1|/\sigma_{1c}} \right\} dx dy.
\] (14)

One can check that \( \mathcal{F}_L \) is, indeed, locally positive definite, and that the system relaxes to the local minima of \( \mathcal{F}_L \), the basic requirements of a Lyapunov functional [26, 27]. We reiterate that this Lyapunov functional is not unique. Indeed, in a closed system, the irreversible entropy production rate may itself serve as an appropriate Lyapunov functional [28].

We can now readily verify that a functional minimization of \( \mathcal{F}_L \) obtains the required steady-state solutions for strains and \( \phi \). Finally, note that, in (14), the first term within the integral is a purely elastic contribution, whereas the second term is non-affine. Elasticity favors the production of \( \phi \) (with appropriate sign) once the threshold criterion is satisfied, since the elastic energy is reduced through stress relaxation. On the other hand, large \( c_x \) and/or \( c_y \) inhibits the formation of (non-uniform) \( \phi \) at the NAZ, where the stress is large. The balance of these two tendencies optimizes the magnitude and the sign of \( \phi \) and, consequently, the nature of the microstructure.

4. The droplet ansatz

Our variational ansatz for \( \epsilon_1(x, y) \) is shown in figure 3(a). We consider a rectangular droplet of the martensitic (twinned rhombic) phase occupying a region \( R_3 \) of dimension \( L \times W \) and consisting of a linear array of \( N \) twins with alternating values of the order parameter \( \epsilon_1 = \pm \epsilon_0 \) represented by

\[\text{Figure 2.} \quad \text{(a) Schematic illustration of a typical local rearrangement producing non-affine displacements. Note that such a transformation produces five and seven coordinated neighborhoods which may be interpreted as a pair of tightly packed dislocations with overlapping cores.} \quad \text{(b) Illustration of the threshold dynamics for the non-affine field given in (7).} \quad \text{(c) Schematic illustration of NAZs produced when the stress } \sigma_1(x) \text{ crosses the threshold } \sigma_{1c}. \text{ Horizontal lines show three values for } \sigma_{1c}. \text{ For large } \sigma_{1c} > \sigma_1(x) \text{(thin dashed line) everywhere, NAZs are never produced. As } \sigma_{1c} \text{ decreases, small isolated NAZs begin to form—the regime described in our present calculation (bold dashed line). For } \sigma_{1c}, \text{ which is too small (dotted line), NAZs form everywhere, and our droplet ansatz eventually breaks down.}\]
alternating strips of black and white. The droplet is oriented such that the habit planes or the parent–product interfaces coincide with the lines \( x = \pm W/2 \) and the twin interfaces \( y_k = -L/2 + kLN', \) for \( k = 0 \cdots N' \). We assume periodic boundary conditions in both directions. All interfaces are assumed to be flat, which we later show to be a reasonable assumption for most cases. Region R1 represents the parent austenite (square) phase \( e_3 = 0 \) which is unaffected by the presence of the martensite droplet. Within region R2, of size \( L/N', \) the order parameter \( e_3 \) still vanishes, but the presence of the droplet produces stress fields \( \sigma_1 \) which determine the structure and dynamics of the interface. In figure 3(b), we have shown a typical interface of width \( \eta \) between twins with \( e_3 = +e_0 \) and \( e_3 = -e_0 \). The points are from a numerical solution of the dynamical equations (4) [11], and the line is a linear interpolation which we use for our droplet ansatz here. The interface between the parent and the product where the order parameter \( e_3 \) intergrades between \( \pm e_0 \) and 0 is similarly represented by a linear ansatz with interfacial width \( \xi \). Within region R2, the order parameter \( e_3 \) vanishes, and hence nonlinearities in the free energy functional may be neglected.

Since the volumetric strain is not a broken-symmetry variable, it relaxes rapidly to its steady-state value determined by the configuration of \( \phi \). We determine \( e_i^A \) by solving the partial differential equation (4) in region R2. Equation (4) is essentially a Poisson equation with ‘electric potential’ \( e_i^A \) being produced by a ‘charge density’ \( \partial^2 e_3 / \partial x \partial y \) with the boundary condition \( e_i^A = 0 \) at distances large from the droplet, i.e., region R1. In region R2, \( \partial^2 e_3 / \partial x \partial y = 0 \) everywhere except at the triple junctions where twin interfaces meet the martensite–austenite interface (shown in figures 3(a) and (c)). This analogy is expected to be valid in the presence of small or moderate amounts of \( \phi \), which reduces the effect of the anisotropic long-range elastic kernel. This corresponds to a situation when the threshold stress is relatively large together with small diffusion times so that \( \phi \) accumulated at the interface is never too large (figure 2(c)). When, on the other hand, generous amounts of \( \phi \) are produced, this relation no longer holds. Therefore, in the limit of small interfacial width, \( e_i^A \) is the electric potential in two dimensions due to a configuration of point charges of alternate sign \( \pm \sqrt{2\pi \lambda} \) centered at the triple junctions with

\[
\lambda = \frac{1}{\sqrt{2\pi\pi}} q \sin(e_0) \left( \frac{1}{\xi} + \frac{1}{\eta} \right) \sqrt{\xi^2 + \eta^2}. \tag{15}
\]

For large \( N' \), the potential (\( e_i^A \)) at any point \((x, y)\) within R2 and R3 due to this charge configuration in the absence of \( \phi \) may be written as

\[
e_i^A(x, y) = \lambda \cos(qy)(e^{-q|x+W/2|} - e^{-q|x-W/2|}), \tag{16}
\]

where \( q = \pi N'/L, \) and we have used a Fourier decomposition limiting ourselves to the most significant term.

Initially, the stress \( \sigma_1 = a_1 e_3^A \) is also largest at these triple junctions and is most likely to cross the threshold \( \sigma_{1c} \) there, generating \( \phi \) (which decreases \( \sigma_1 \)). Accordingly, we supplant our ansatz for the OP field with one for \( \phi \), namely

\[
\phi = \phi_0 \left[ e^{-q(x+W/2^2)/2\beta_1} - e^{-q(x-W/2^2)/2\beta_1} \right] \times \sum_{k=-N'}^N e^{-q(kL/2N'/2\beta_1)} \cos(qy), \tag{17}
\]

\[
\text{Figure 3.} \quad (a) \text{Schematic showing our ansatz for } e_3 \text{ corresponding to the martensite droplet. Periodic boundary conditions are assumed in both the } x \text{ and } y \text{ directions. There are three regions: R1, R2, and R3. Region R1 contains the parent austenite (} e_3 = 0). \text{ All strain and associated stress fields are zero in this region. Region R3 is of size } L \times W \text{, and it contains } N' \text{ twins (six shown). The twins consist of regions where } e_3 = \pm e_0, \text{ designated by white and black, which are degenerate in free energy. The gray regions are interfaces with widths } \eta \text{ and } \xi. \text{ Region R2 has no } e_3 \text{ but contains a non-zero non-order parameter strain } e_3^A \text{ and associated stress } \sigma_1 \text{ which is produced due to the strain inhomogeneity created at the droplet. } \sigma_1, \text{ which alternates in sign, reaches maximum absolute values at the blue and red colored circles marking regions of maximum strain inhomogeneity. (b) The ansatz } e_3(y) \text{ versus } y \text{ (solid line) and the numerical solution of the dynamical equation (filled squares). The interfacial width } \eta \text{ is shown. (c) A close up of the triple junction where the two twin variants (white and black) meet at a austenite–martensite interface. The red ellipse with widths } \beta_x \text{ and } \beta_y \text{ represents a typical NAZ.}
\]
which peaks at every triple junction with anisotropic widths \( \beta_x \) and \( \beta_y \) in the \( x \) and \( y \) directions, respectively, and an amplitude which changes sign following that of \( e_6^A \). The form of the ansatz (shown schematically in figure 3(c)) interpolates between a \( \phi \) which may either be strongly localized (small \( \beta_x, \beta_y \)) or one which completely wets the austenite–martensite interface and is modulated with a period matching that of \( e_6^A \) and \( e_3 \).

Substituting the OP and slaved NOP strains and \( \phi \) in \( \mathcal{F}_L \), we obtain the droplet free energy:

\[
\mathcal{F}_L = I_0 L W + 2\Sigma_{pp} L + \Sigma_{in} W N + 2\pi a_1 \lambda^2 \frac{L^2}{N} + F_\phi(\phi_0, \beta_x, \beta_y) N, \tag{18}
\]

where the constant \( I_0 = a_3 e_6^A / 2 - b_3 e_6^A / 4 + d_3 e_6^B / 6 \) multiplies the first term representing the contribution from the bulk of the droplet. The second and the third terms with coefficients \( \Sigma_{pp} = c_3 \int_{-\infty}^{\infty} dx (\partial e_3 / \partial x)^2 = 2 e_0 \sqrt{c_3 I_0} \), the surface tension due to the parent–product and \( \Sigma_{in} = \frac{\pi}{2} \int_{-\infty}^{\infty} dy (\partial e_3 / \partial y)^2 = 4 e_0 \sqrt{c_3 I_0} \) for each twin interface with \( I_0 = a_3 e_6^A / 3 - b_3 e_6^A / 5 + d_3 e_6^B / 7 \), represent the energies of the parent–product and twin interfaces. The fourth term gives the contribution of the fringing NOP field \( e_1^A \). The function \( F_\phi(\phi_0, \beta_x, \beta_y) \) in the fifth term is the contribution of the non-affine field, \( \phi \), to the Lyapunov functional \( \mathcal{F}_L \), which we had to obtain by numerical integration, checking our results against analytical expressions obtained in the pathologic but instructive limit \( \sigma_{1c} \to 0 \). \( F_\phi(\phi_0, \beta_x, \beta_y) \) contains terms which are linear and quadratic in \( \phi_0 \). Unimportant contributions from gradients of \( e_1^A \) have also been ignored, for simplicity.

Equation (18) is minimized with respect to \( \beta_x, \beta_y, N, \) and \( \phi_0 \) as well as the interfacial widths \( \xi \) and \( \eta \) for a fixed choice of all the parameters and size of the droplet \( L \) and \( W \).

5. Steady-state solutions and the dynamical phase diagram

Minimization of \( \mathcal{F}_L \) with respect to the interfacial widths immediately yields \( \eta = \xi = e_0 \sqrt{c_3 / I_0} \), giving \( \lambda = 2 q_{13} e_0 / \sqrt{\pi} \).

Since we know that interfacial widths in solids are typically of the order of a lattice spacing [2], this correspondence sets the microscopic length scale of our continuum model. Using these values, we minimize \( \mathcal{F}_L \) with respect to the rest of the free parameters, namely, \( \beta_x, \beta_y, \phi_0, \) and \( N \), for a range of values for \( c_x, c_y, \) and \( \sigma_{1c} \). Our main results may be summarized as follows.

(i) We obtain three distinct solutions or dynamical phases, labeled I, II, and III, which correspond to unique local minima of \( \mathcal{F}_L \) in particular regions of parameter space.

(ii) The three dynamical phases show a wide variation in physical properties. While solution I is essentially affine, solutions II and III involve non-zero values of \( \phi \).

(iii) \( \phi \), if present, is always strongly localized perpendicular to the habit plane, i.e. along \( x \) such that \( \beta_x < \beta_y \ll W \) for all three solutions. Twin interfaces, therefore, do not ever contain any \( \phi \).

(iv) In solution II, \( \phi \) is localized at regions of high stress concentration at the habit plane, while in III, \( \phi \) completely ‘wets’ the habit plane.

(v) Individual twins in I are elastically coupled, while they are decoupled in II and III.

(vi) While I is identified as twinned ferro-elastic martensite, we propose that II and III are dislocated martensites.

We now explain each of these results in detail. The magnitudes of \( \phi_0 \) and \( \beta_y \) are controlled by the dynamical constants \( c_x \) and \( c_y \), and \( \sigma_{1c} \). For fixed \( \sigma_{1c} \), solution I, which exists for large \( c_x \) and \( c_y \), has negligible non-affineness \( (\phi_0 \approx 0) \) except at a small region of size \( \sim \xi \) at the triple junctions. In this case, a suitable coarse graining of the interface taking \( \xi \to 0 \) may be used to derive a re-normalized strain-only theory without \( \phi \) [11]. Therefore, this solution most resembles ferro-elastic martensite domains describable completely within strain-only approaches [8, 10, 12].

To compare the nature of the two remaining solutions, we have plotted \( \beta_y \) and \( \phi_0 \) for both II and III as a function of \( c_x \) for fixed \( c_y \) over a typical range of parameters in figure 4(a).
The sign of $\phi_0$ in both solutions II and III is negative, showing that the non-affine field effectively decreases the NOP stress ($\propto \lambda$). Solution II comprises localized NAZs of large $|\phi_0|$ at the triple junctions, while III has smaller $|\phi_0|$ which completely wets the habit plane. As $c_y$ increases, $|\phi_0| \to 0$ for solution III, though, before it vanishes altogether, a first-order dynamical transition to I intervenes. The width of the $\phi$ distribution along the habit plane, $\beta_y$, is relatively insensitive over the entire range of $c_y$ over which these solutions are stable.

Increasing $N$ leads to smaller NOP fringing fields $c_y^4$ at the austenite–martensite interface and consequently lowers the contribution of the elastic energy due to elastic coupling between the twins. However, large $N$ introduces more twin interfaces and hence increases the interfacial energy of the droplet. For vanishing $\phi$, the interplay between these two factors alone decides the optimum number of twins, $N_c$. For example, we obtain

$$\frac{L}{N_c} = \sqrt{\frac{2\pi \Sigma_{1w} W}{a_1 \lambda^2}}.$$  \hfill (19)

This relation between the size of the twin $L/N_c$ and the width $W$ has been verified experimentally, and has been obtained in earlier calculations [6, 7, 12]. Substituting the minimized $N$ in (18), one obtains $F_1$, as a sum of bulk ($\propto LW$) and surface ($\propto L + W$) terms with an effective surface tension given by

$$\Sigma_{eff} = \Sigma_{pp} + \sqrt{a_1 \lambda^2 \Sigma_{1w} W/2\pi}.$$

Including $\phi$ changes the relation between the twin size and the width depending on the resulting optimum value for $\phi_0$, since $\phi$ changes both the numerator and the denominator of (19), namely

$$\frac{L}{N_c} = \sqrt{\frac{2\pi (\Sigma_{1w} W + A\phi_0 + B\phi_0^2)}{a_1 \lambda^2 + 2a_1 \lambda \phi_0}},$$  \hfill (20)

where $A$ and $B$ are positive definite functions of $\beta_x$, $\beta_x$, and $L/N_c$. For $\phi \to 0$, namely for solution I, we can neglect the term quadratic in $\phi_y$; the small linear contributions to both the numerator and the denominator in (20) then keeps the scaling relation unaltered. This is shown in figure 4(b), where we have plotted $L/N$ versus $W/\xi$ in scaled coordinates by changing $W$ in our ansatz for a fixed $L$ and for sets of parameters where each of the phases are stable. As $|\phi_0|$ increases, the solution for $L/N$ is not straightforward, because the numerator is itself a (complicated) function of $L/N$ and one needs to solve (20) self-consistently and numerically. The result, also shown in figure 4(b), reveals that $\phi$ decreases the stress, since non-affineness screens the elastic interactions between twins. For both solutions II and III, the twins appear to be completely decoupled elastically, suggesting that $\phi_0 \sim W$, canceling the $W$ dependence of the numerator, making the twin width $L/N$ almost independent of $W$.

We obtain a dynamical phase diagrams showing regions of stability (lowest $F_1$) for solutions I, II, and III in figures 5(a) and (b) in the scaled parameters $c_y/W\xi$ and $c_y/c_x$ for fixed $\sigma_{ic}$. Remarkably, in these variables, the dependence on $W$, the size of the droplet, may be scaled away. For large $c_x$ and $c_y$, the system finds it increasingly difficult to generate non-affinity, and there is a transition from structures II or III (large $|\phi_0|$) to I (small $|\phi_0|$). Solids which form only twinned martensite microstructures are expected to have dynamical parameters corresponding to this region of our phase diagram. For lower $c_x$ and $c_y$, on the other hand, $\phi$ is produced and solutions II and III are stable. In this region, as $c_y$ is increased, there is a jump in the values of both the localization parameter $2\beta_x N/L$ and $\phi_0$ as the system switches from solution II with localized $\phi$ with large amplitude to solution III with a relatively delocalized broad (in $y$) $\phi$ distribution, albeit with a smaller amplitude. During this transition from II to III, $\beta_x$ also decreases, though this change is not as much as in $\beta_y$.

Finally, if $\sigma_{ic}$ is increased, predictably, non-affineness decreases stabilizing the ‘strain-only’ I solution. In this case, the triple point in the phase diagram shifts to lower $c_x$, values, and the phase boundary between the I and II phases shifts to the left (figure 5(b)) and eventually tends to unphysical negative values for $c_x$ and $c_y$. For large $\sigma_{ic}$, again, one recovers the results of strain-only theories where twinned martensite with $\phi = 0$, i.e. solution I, is the only stable dynamical phase.
While the interpretation of solution I as a strain-only ferro-elastic twinned martensitic droplet is clear, the nature of solutions II and III still needs to be established. Our calculation shows that $\phi$ decreases the strong elastic coupling between the twins so that they are, in effect, individual grains of the product phase surrounded by an interface containing regions of large non-affinity where the St Venant incompatibility $\nabla^2 \phi \neq 0$. Solution III, where the NAZ completely surrounds the product droplet, is very similar to the ferrite nuclei observed by us in our MD simulations [9, 14, 17]. On the other hand, solution II, which retains characteristics of both martensites, with a well-defined twinned structure, together with some localized $\phi$ as in ferrite, may correspond to a generic intermediate microstructure [2, 3, 18], many examples of which are observed in real materials like steel.

6. Discussion and conclusion

In this paper, we have introduced a coarse-grained continuum model which extends strain-only theories of martensites by including non-elastic degrees of freedom capable of describing localized regions called NAZs within which particles are known to have non-affine displacements. Before we end, we discuss our present results, taking up a couple of issues which examine our assumptions, illustrate the relevance of our findings, and point out avenues for further work.

First, our ansatz for the shape of the martensite droplet is rectangular, implying flat twin and parent–product interfaces. How good is this assumption? While twin interfaces are usually flat because they minimize elastic energy, moving parent–product interfaces during growth of the product phase may undergo dynamical (Mullins–Sekerka) instabilities, producing a variety of interesting structures and shapes, e.g. dendrites [30]. On the other hand, austenite–martensite interfaces in ferro-elastic materials like steel may correspond to a generic intermediate microstructure [2, 3, 18], many examples of which are observed in real materials like steel.

In order to investigate the validity of our ansatz for solutions II and III, we calculate $F_L$ numerically after imposing a modulation $\sim \Lambda_m \sin(q_m y)$ (figure 6(a)) on the habit plane. In figure 6(b), we show that the curvature of the free energy $\partial^2 F_L / \partial \Lambda_m^2 \sim q_m^{-2} > 0$, proving that our ansatz is stable against such modulations. Nevertheless, dynamical instabilities of the Mullins–Sekerka type can still arise, since $\phi$ screens elastic interactions leading to a less than $\sqrt{W}$ dependence of the effective surface tension. Indeed austenite–ferrite interfaces which contain many defects [19] are hardly planar. This has also been observed in our own MD simulations [14] and elasto-plastic theory [11].

Are there direct experimental signatures for the dynamics of the $\phi$? Since the velocity of martensitic interfaces is usually quite large, resolution of the dynamics $\phi$ relative to the moving interface is notoriously difficult. Nevertheless, slow dynamics of non-elastic degrees of freedom such as atomic shuffles are routinely observed in complex pseudo-martensites such as the olivine to spinel transition in fayalite [31] using in situ time-resolved x-ray diffraction. We believe that similar in situ techniques, modified to observe local interfacial dynamics [32], may be able to capture some of the phenomena discussed in our paper.

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