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Ginzburg-Landau theory of microstructures: Stability, transient dynamics, and functionally graded nanophases

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Abstract. – The stability, transient dynamics, and physical interpretation of microstructures obtained from a Ginzburg-Landau theory of first-order phase transformations are studied. The Jacobi condition for stability fails numerically, thus an alternative exact stability criterion, based on critical (most destabilizing) fluctuations, is developed. The degree-of-stability parameter is introduced to quantify the physical stability of long-lived unstable microstructures. For nanofilms, the existence of functionally graded nanophases is demonstrated. Numerical simulations indicate that graded nanophases can be produced by dissolving material from both surfaces of a nanofilm. Stability under finite fluctuations and post-bifurcation microstructure evolution are investigated numerically.

Introduction. – The Ginzburg-Landau (GL) equation (1) is used for the description of a wide class of first-order phase transformations (PTs), including martensitic PTs in shape memory alloys [1, 2], some reconstructive PTs [3, 4], ferroelastic, magnetoelastic, and ferroelectric PTs. The GL framework also describes dislocation generation [5], stress-induced martensitic PTs, twinning and dislocation generation (with an emphasis on accounting for typical experimental observations [6], in contrast to previous approaches). Several types of periodic analytic solutions of the static GL equation (microstructures) have been found [1, 4], but the stability of these microstructures, and consequently their physical interpretation, as well as their transient dynamics were not thoroughly analyzed. This is in contrast to microstructures associated with other physical processes (described by equations other than GL or by other types of GL equation) for which the stability of static and dynamic solutions has been treated extensively [7]. In these papers the stability of static solutions is studied by carrying out linear stability analyses, which are essentially eigenvalue problems, with ansatzes for the forms of the fluctuations. Such analyses, however, do not guarantee that all unstable solutions are found because the most destabilizing fluctuations are not obtained by these methods.

In this letter we present a new *exact* method for analyzing the stability of a static microstructure described by the GL equation. We seek the critical (most destabilizing) fluctuation h_c that (by definition) minimizes the second variation of the energy, $\delta^2 e$, at a critical point of the energy. If $\delta^2 e$ is negative (or positive) for such a fluctuation, then the microstructure under study is unstable (or stable). Using this approach, we comprehensively study all single- and multi-interface microstructures in one dimension for a specific GL energy. We find several types of unstable microstructures, and we numerically solve the time-dependent GL (TDGL) equation to elucidate the physical meaning of these microstructures. Some are found to be

critical nuclei (martensitic, austenitic and martensite-martensite). It is found that for other microstructures (equilibrium austenite-martensite or martensite-martensite) there are critical sample lengths below which they are unstable; explicit expressions for the critical lengths are determined. These unstable microstructures may be long-lived; some initial configurations evolve to these unstable microstructures, which is unexpected and nontrivial. We introduce the degree of stability parameter to characterize the physical stability of microstructures that are formally (mathematically) unstable. We show that the n -interface microstructures have the same degree of stability as the single-interface microstructure, which is unexpected because their energy is $\sim n$. Also, for nanofilms, the existence of functionally graded nanophases is demonstrated. Numerical simulations indicate that graded nanophases can be produced by dissolving material from both surfaces of a nanofilm. Microstructural stability under finite fluctuations and post-bifurcation microstructure evolution are investigated numerically.

In one dimension, the dimensionless GL energy is of the form $g_{GL} = g(\xi) + \xi'^2$, where ξ is the order parameter and $\xi' = \partial\xi/\partial x$. The corresponding TDGL equation reads

$$\partial\xi/\partial t = -dg/d\xi + 2\xi''; \quad \xi'(\pm l/2) = 0. \quad (1)$$

In this paper we investigate the solutions of (1) for the Landau potential $g(\xi) = B\xi^2 - \xi^4 + \xi^6$ in a sample of length l . For $\partial\xi/\partial t = 0$, $x = \int (g - g_0)^{-1/2} d\xi$ and the boundary conditions become $g(\pm l/2) = g_0$, where g_0 is a constant and $g \geq g_0$ [1, 6]. The potential g has three minima: austenite, A ($\xi(A) = 0$), and two martensitic variants, M_- and M_+ ($-\xi(M_-) = \xi(M_+) = \xi_M$). These are just convenient names, because our results are not limited to martensitic PTs.

Stability of static microstructures I: Jacobi method. – A static solution $\xi_s(x)$ of the GL equation is stable if it minimizes the energy $e[\xi] = \int_{-l/2}^{l/2} g_{GL}(\xi) dx$. The solution $\xi_s(x)$ yields a minimum of the energy if the second variation of the energy

$$\delta^2 e[h] = 2 \int_{-l/2}^{l/2} (C(x)h(x)^2 + h'(x)^2) dx, \quad C(x) := \frac{1}{2} \frac{d^2 g(\xi_s(x))}{d\xi^2}, \quad (2)$$

is positive for all admissible fluctuations $h(x)$ ($h'(\pm l/2) = 0$). The exact Jacobi condition that $\xi_s(x)$ minimizes $e[\xi]$ [8] is that the solution of the corresponding Jacobi equation

$$h''(x) = C(x)h(x) \quad (3)$$

for $h(-l/2) = 0$ does not have a zero in the interval $(-l/2, l/2)$ [8]. The Jacobi condition is usually formulated for extrema with fixed values at the boundaries [8]. However, we found that the Jacobi condition is also valid for $\xi'(\pm l/2) = 0$.

A-M interfaces: equilibrium functionally graded nanophases. – For $0 < B < 1/3$ and $g_0 > 0$, solutions of the TDGL equation represent periodic A-M microstructures [1, 4, 6]:

$$\xi^{AM}(x) = \xi_1 / \sqrt{1 - (1 - \xi_1^2/\xi_2^2) sn^2(\xi_2 p x, \xi_3 s/(\xi_2 p))}, \quad (4)$$

where sn is the Jacobi elliptic function; $s := \sqrt{\xi_2^2 - \xi_1^2}$ and $p := \sqrt{\xi_3^2 - \xi_1^2}$, and $g = g_0$ has three pairs of real roots, $\pm\xi_1$, $\pm\xi_2$, and $\pm\xi_3$, for which we assume $0 < \xi_1 \leq \xi(y) \leq \xi_2 < \xi_3$. Application of the boundary conditions gives $l = nK(\xi_3 s/(\xi_2 p))/(\xi_2 p)$, where K is the complete elliptic integral of the first kind and n is the number of interfaces. As all roots ξ_i are functions of B and g_0 , the last equation allows us to determine g_0 for prescribed B , n and l , and thereby complete the solution. The period of the microstructure is $T = 2l/n$. Both $\xi^{AM}(x)$ and $g(\xi)$ for A and M in thermodynamic equilibrium ($B = 1/4$), $n = 1$, and several l are shown in fig. 1a.

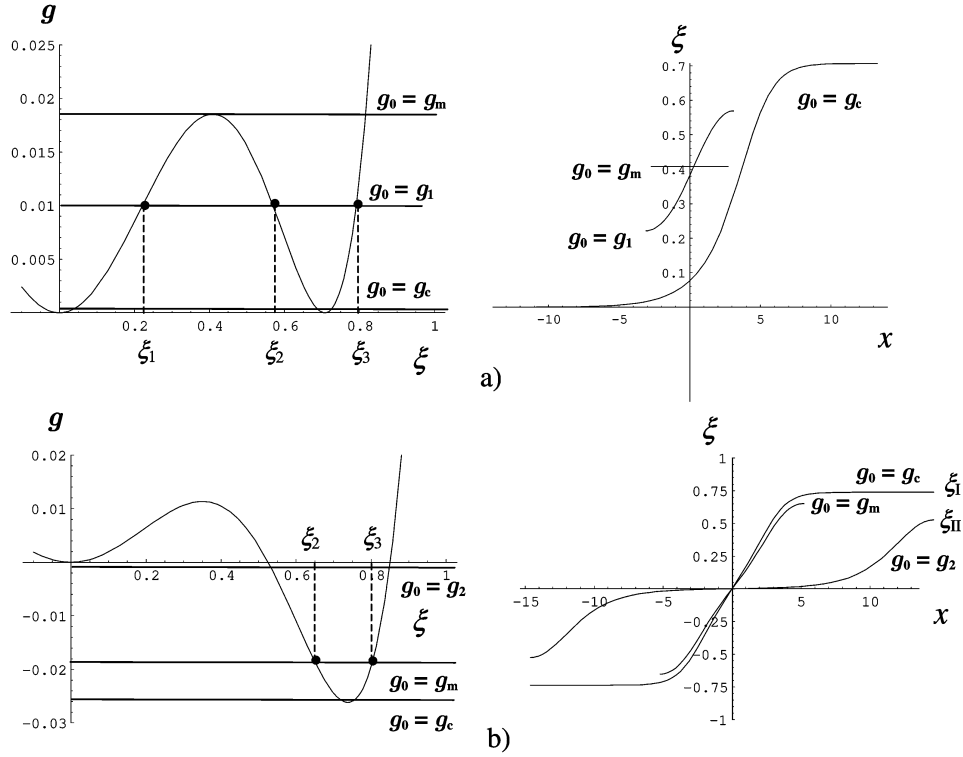


Fig. 1 – Landau potentials (symmetric with respect to ξ) and static microstructures $\xi_s(x)$ for (a) A-M and (b) M₋-M₊ interfaces; g_m corresponds to the minimal sample length l_m and g_c is for the critical length l_c .

If A and M are not in thermodynamic equilibrium ($B \neq 1/4$), then the Jacobi method indicates that ξ^{AM} is unstable for any n and l . Numerical analysis of the TDGL eq. (1) demonstrates that *these microstructures are critical M nuclei* for $B < 1/4$ (in the region of stability of M). Thus the initial fluctuation $k \xi^{AM}(x)$ with $k > 1$ ($k < 1$) evolves into M (A). Similarly, the *microstructures for $B > 1/4$ are critical A nuclei*. Their energies [1] are the activation energies for nucleation. Since the energy $\sim n$ [1], it is *very improbable that critical nuclei with $n > 2-3$ would be observed*

Although the formal solution (4) was known, its physical interpretation for a nanoscale sample was lacking. The solution ξ^{AM} is a constant for l/n below a minimal length l_m that can be determined from the condition $\xi_1 = \xi_2$ (fig. 1a): $l_m = \sqrt{3}\pi / (2\sqrt{1 - \sqrt{1 - 3B}}(1 - 3B)^{1/4})$. This homogeneous phase corresponds to a *maximum of the Landau energy* rather than a minimum! For $B = 1/4$ (thermodynamic equilibrium between A and M), $\xi_h^{AM} = \sqrt{1/6}$ represents a *new homogeneous phase*. Numerical analysis confirms that it is unstable: both homogeneous and asymmetric fluctuations induce the transformation of this phase into either A or M. However, ξ_h^{AM} plus large initial sinusoidal fluctuations still evolves toward ξ_h^{AM} , but eventually the specimen transforms to pure A or M (fig. 2). If we start from ξ^{AM} for $B = 1/4$ and $l = l_c$ (minimal length for stability; see below) and repeatedly reduce l by small equal decrements from both ends, then the solution of the TDGL equation for each length of the

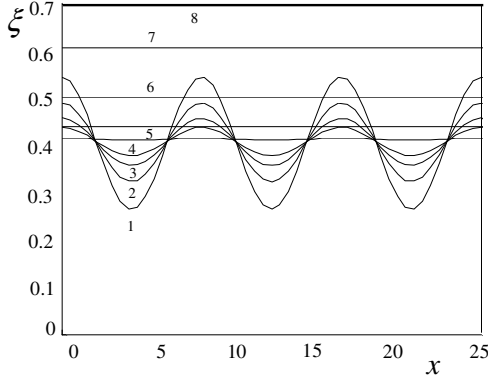


Fig. 2

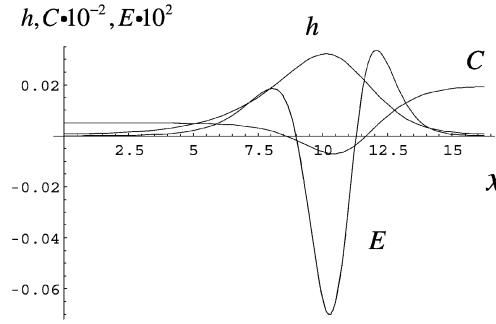


Fig. 3

Fig. 2 – Evolution of ξ_h^{AM} plus a large sinusoidal fluctuation to ξ_h^{AM} followed by transformation to M. The numbers show the time sequence.

Fig. 3 – Functions $C(x)$, the critical fluctuation $h_c(x)$, and $E = 2[C(x)h(x)^2 + h'(x)^2]$ for the equilibrium A-M microstructure.

specimen evolves to the corresponding unstable static microstructure, including $\xi_h^{\text{AM}} = \sqrt{1/6}$ for $l = l_m$. This procedure suggests that this *new phase could be produced by dissolving material from both surfaces of a nanofilm (or nanotube of large radius) at the same rate. This phase is highly energetic and its energy would be released during its transformation to A or M.*

When l is greater than l_m but smaller than the interface width, the microstructure consists of continuously varied phases $\xi(x)$, $\xi_1 \leq \xi \leq \xi_2$, with continuously varying properties across the sample (fig. 1a). In three dimensions, the crystal structures of these phases may differ from those of both A and M. Since the interface width is typically in the nanometer range, the solution predicts the *existence of functionally graded nanophases* in nanofilms or nanotubes of large radius. Only for $l \gg l_m$ ($g_0 \simeq 0$) does the microstructure (4) consist of regions of A ($\xi = 0$) and M ($\xi = \xi_M$) divided by narrow diffuse interfaces.

M₋-M₊ interfaces. – Periodic microstructures for $0 < B < 1/4$ and $g_0 < 0$ are [1]:

$$\xi^{\text{MM}}(x) = \frac{\bar{\xi}_1 \xi_2 \text{sn}(s\xi_3 x, p\xi_2/(s\xi_3))}{\sqrt{\xi_2^2 \text{cn}^2(s\xi_3 x, p\xi_2/(s\xi_3)) - \xi_1^2}}, \quad l = \frac{2nK(p\xi_2/(s\xi_3))}{\xi_3 s}; \quad (5)$$

cn is a Jacobi elliptic function. The equation $g = g_0$ has one pair of imaginary roots $\pm\xi_1 = \pm i\bar{\xi}_1$ and two pairs of real roots, $(\pm\xi_2, \pm\xi_3)$. The microstructure (5) does not exist for l/n below a minimal length l_m , which can be found from the condition $\partial l/\partial g_0 = 0$. For each admissible l/n there are two possible microstructures, $\xi_I^{\text{MM}}(x)$ and $\xi_{II}^{\text{MM}}(x)$ (fig. 1b). $\xi_I^{\text{MM}}(x)$ has the smaller energy and varies from M₋ to M₊ in a narrow diffuse interface for relatively large l/n ; it corresponds to $\xi_2 \simeq \xi_3$ and $g_0 \simeq g(\xi_2)$. For relatively large l/n , $\xi_{II}^{\text{MM}}(x)$ has a large A region between M₋ and M₊; it corresponds to $0 > g_0 \simeq 0$. These solutions converge toward each other with decreasing l/n and coincide at $l/n = l_m$, which provides a physical interpretation of l_m . For small l , the microstructure (5) represents a functionally graded nanophase.

The Jacobi method shows that the microstructure $\xi_{II}^{\text{MM}}(x)$ with the higher energy (fig. 1b) is unstable for all l and B . It was shown by numerical solution of the TDGL equation (1) that $\xi_{II}^{\text{MM}}(x)$ represents a *critical M₋-M₊ nucleus*: the initial fluctuation $k\xi_{II}^{\text{MM}}(x)$ evolves into the low energy microstructure $\xi_I^{\text{MM}}(x)$ for $k > 1$, while for $k < 1$ it evolves into A.

Stability of static microstructures II: critical fluctuations. – For A-M interfaces in thermodynamic equilibrium and for the M_-M_+ microstructure $\xi_1^{MM}(x)$, the *Jacobi method breaks down*: $h(l/2)$ oscillates around zero with a magnitude of order the numerical error. The problem is clearly illustrated by comparing analytic and numerical solutions of the Jacobi equation for the homogeneous (most unstable) nanophase $\xi_h^{AM} = \sqrt{1/6}$ for which $C = -\pi^2 l_m^{-2}$. The exact solution of the Jacobi eq. (3) for $h(-l_m/2) = 0$ and $h'(-l_m/2) = \mu$ is $h(x) = l_m \mu \cos(\pi x/l_m)$, thus $h(l_m/2) = 0$. However, numerical solutions of eq. (3) yield small positive or negative values of $h(l_m/2)$ with equal probability, independent of numerical accuracy.

We have developed an alternative to the exact Jacobi method in which we seek the critical (most destabilizing) fluctuation h_c ($h'_c(\pm l/2) = 0$) that minimizes $\delta^2 e$ at a critical point of $g_{GL}(\xi)$, *i.e.*, at $\xi_s(x)$. If $\delta^2 e[h_c] > 0$ ($\delta^2 e[h_c] < 0$), then the static microstructure $\xi_s(x)$ is stable (unstable). However, finding h_c is not straightforward because the proper problem formulation has to be determined. The Euler-Lagrange equation for $\delta^2 e \rightarrow \min$ is the Jacobi equation (3), which is homogeneous in h ; thus, if h_c is a solution then $k h_c$ is also a solution. Then $\delta^2 e[k h_c] = k^2 \delta^2 e[h_c]$, which is less than $\delta^2 e[h_c]$ for $|k| < 1$. Hence, a critical fluctuation cannot be a solution of the Jacobi equation. Multiplying eq. (3) by $h(x)$, integrating over $[-l/2; l/2]$, and taking the boundary conditions into account, one obtains $\delta^2 e = 0$ for any solution h . However, for any constant fluctuation $h = \alpha$ one has $\delta^2 e = a^2 J < 0$ if $J := \int_{-l/2}^{l/2} C dx < 0$.

Determination of h_c is a well-posed problem only if the freedom to rescale h_c , which can give $|\delta^2 e|$ any value on $[0, \infty)$, is eliminated from the problem formulation. It is sufficient to fix the normalization of h on $[-l/2, l/2]$: $K := \int_{-l/2}^{l/2} h(x) dx = N$. This constraint changes the Euler-Lagrange equation from eq. (3) to the inhomogeneous linear equation

$$h'' = C(x)h(x) + \lambda, \quad (6)$$

where λ is the Lagrange multiplier. As we now show, the solution to eq. (6) is still not a critical fluctuation. Adding a constant α to the solution h of (6) changes $\delta^2 e[h]$ to $\delta^2 e[h + \alpha] = \delta^2 e[h] + 2\alpha^2 J + 4\alpha A[h]$, where $A[h] := \int_{-l/2}^{l/2} C(x)h(x) dx$ (see eq. (2)). Only the case $J > 0$ is of interest (for $J \leq 0$, the microstructure $\xi_s(x)$ is unstable for $h = \text{const}$). The functional $\delta^2 e[h + \alpha]$ has a minimum for $\alpha = -A[h]/J$. Thus $A[h] = 0$ is a necessary condition for h to be a critical fluctuation, while integration of (6) shows that $A[h] = -\lambda l \neq 0$. Consequently, $A[h] = 0$ must be imposed as an additional constraint to find a critical fluctuation. Our final problem formulation is $\delta^2 e \rightarrow \min$, $K = N$, and $A = 0$, which results in the equation

$$h''(x) = C(x)[h(x) - \alpha] + \lambda. \quad (7)$$

Integrating eq. (7) over $[-l/2; l/2]$, one obtains $\alpha = \lambda l/J$. The solution to eq. (7) and consequently N scale with λ . Since our stability analysis based on $\delta^2 e$ is valid for arbitrary infinitesimal perturbations, the magnitude of λ can have any infinitesimal value. This property is physically relevant: only the shape of the critical fluctuation is determined. In practice we solve eq. (6) and then simply add $\lambda l/J$ to $h(x)$ to give $h_c(x)$. For the microstructures ξ_s corresponding to critical A, M, and M-M nuclei, the solution to (6) gives $\delta^2 e > 0$ even though the ξ_s are unstable, but after adding $\lambda l/J$, we obtain $\delta^2 e < 0$, as expected.

Critical fluctuations were calculated for the equilibrium A-M interface ($B = 1/4$) and the M_-M_+ microstructure $\xi_1^{MM}(x)$ for $n = 1$ and several values of B as a function of l . This numerical analysis shows that there is a critical length l_c above (below) which each microstructure is stable (unstable) for a given value of B . The critical lengths are approximated by $l_c = 21.626 - 3.795B + 248.115B^2$ for M_-M_+ interfaces and $l_c = 26.6$ for the equilibrium A-M interface. Similar calculations for the potential $g_4 = B\xi^2 - \xi^3 + \xi^4$ resulted in $l_c = 44.12$ for the A-M interface for $B = 1/4$. However, the weak dependence of $\delta^2 e$ on l in the vicinity of l_c precludes

the accurate determination of l_c . The small value of $\delta^2 e$ near l_c is the difference of two large numbers, the integrals of $\delta^2 g_{GL}$ over the regions where $\delta^2 g_{GL}$ is positive and negative (fig. 3).

In addition to unavoidable numerical inaccuracy, there is also a conceptual problem. For $l \leq l_c$, where $\delta^2 e[h_c] < 0$, the probability of appearance of the specific fluctuation h_c may be small while deviations from h_c for which $\delta^2 e > 0$ may be frequent; therefore, the static microstructure $\xi_s(x)$ could be long-lived and physically stable. We seek a physical measure of stability that ascribes a high (low) degree of stability to long-lived (short-lived) microstructures which are formally (mathematically) unstable. Accordingly, we introduce the *degree of stability of the static microstructure* $\xi_s(x)$ with corresponding critical fluctuation $h_c(x)$:

$$DS(l, B) := -\log_{10}(-\delta^2 e[h_c] / \int_{-l/2}^{l/2} |\delta^2 g_{GL}(h_c)| dx). \quad (8)$$

For $C(x) < 0$, $DS = 0$, which is the smallest possible value of DS . A logarithmic scale is chosen so that DS amplifies the small values of $\delta^2 e$ near the critical length, so only the order of magnitude is important. DS increases with l at fixed B and decreases with B at fixed l . As $l \rightarrow l_c$ from below, $\delta^2 e \rightarrow 0^-$, resulting in a logarithmic singularity in DS . For $l > l_c$, where the microstructures are stable, that is, where $\delta^2 e[h_c] > 0$, DS is not defined. The results of the numerical calculations can be approximated for $l < l_c$ ($\delta^2 e < 0$) by $DS(l, B) = (-3.54 - 3.23B) + (0.72 - 1.06B)l$ ($0 \leq B \leq 0.2$) for $M_- - M_+$ interfaces, $DS(l, 1/4) = -1.62 + 0.29l$ for the equilibrium A-M interface, and $DS = -2.02 + 0.22l$ for the g_4 potential for an equilibrium A-M interface. DS can be used to compare the relative stabilities of various microstructures; larger values of DS correspond to more stable microstructures. Thus, DS grows with increasing l or decreasing B or both. Multi-interface microstructures correspond to periodically continued $C(x)$ and $h_c(x)$; the period of $C(x)$ is $T/2 = l/n$. Consequently, $\delta^2 e(n) = n\delta^2 e(1)$; the critical length for n interfaces is nl_c and $DS(n) = DS(1)$, both of which are *unexpected because the energy of a n -interface microstructure is $\sim n$* .

Note that the general form of the integral constraint for which the Euler-Lagrange equation is homogeneous in h and has nonzero solution, must be linear in h , *i.e.*, $\int_{-l/2}^{l/2} h(x)f(x)dx = M$ with arbitrary f . Our formulation has the physical advantage that it does not include functions unrelated to the Landau potential. Note that the shape of h_c can be estimated for any $C(x)$: $h_c(x)$ is close to zero in the regions where $C > 0$ and has a peak in the region $C < 0$ (fig. 3); the maximum of $|h'|$ is controlled by the gradient term in $\delta^2 g_{GL}$. All of our solutions h_c satisfy these requirements. Even if the critical length is increased from l_c to \tilde{l}_c for some f , there would be no practical consequences since $DS(l_c)$ would still be large and the corresponding $\xi_s(x)$ would be long-lived.

Transient microstructure dynamics and concluding remarks. – Numerous simulations of the evolution of static microstructures under finite fluctuations h were performed. The evolution depends on the specific form of the fluctuation. For example, for $B = 0.1$, $l = 16.4 < l_c$, $a = 0.004$, and $w = \sin(2\pi x/l)$, the unstable microstructure $\xi_{II}^{MM}(x)$ evolves to: $\xi_I^{MM}(x)$ under $h = aw$; metastable A under $h = -aw$; M_+ under $h = aw^2$; and M_- under $h = -aw^2$. For $B = 0.1$, $l = 7.8 < l_c$, and $a = 0.006$, $\xi_I^{MM}(x)$, which is generally unstable, transforms to M_+ under $h = aw^2$, M_- under $h = -aw^2$, and it is stable under $h = \pm aw$. For $B = 0.1$ and $l = 16.4 < l_c$, an A initial state is stable under $h = 0.1w$ but transforms to $\xi_I^{MM}(x)$ under the larger fluctuation $h = 0.4w$. Interestingly, instability of $\xi_s(y)$ with respect to some fluctuations does not preclude the possibility that an initial configuration that differs significantly from $\xi_s(y)$ nevertheless evolves toward $\xi_s(y)$; see fig. 2. If a stable n -interface A-M or $M_- - M_+$ static microstructure is continuously embedded into a single-variant microstructure, it remains static —*there is no tendency to decrease n and consequently the*

energy of the system by the lateral growth of one variant. These microstructures are also stable under $h = 0.1 \xi_M w$. For a sinusoidal initial state (not a solution of the GL equation) with $l/n > l_m$ ($l/n < l_m$), the solution evolves to a static microstructure with the same n (with $\tilde{n} < n$ satisfying $l/\tilde{n} > l_m$) or to pure A or M. The stability of multiple M_- - M_+ interfaces is important because in three dimensions such a twinned M structure plays a *key role in the minimization of elastic energy through the formation of A-twinned M habit planes*.

To summarize, it was found that the Jacobi condition for stability fails numerically for certain microstructures. A new exact approach to analyzing the stability of static microstructures, which includes determination of critical (most destabilizing) fluctuations, was developed. The DS parameter was introduced to quantify the physical stability of long-lived unstable microstructures. Single and periodic n -interface A-M microstructures are unstable for any l if the A and M are not in thermodynamic equilibrium ($B \neq 1/4$). These microstructures represent critical A and M nuclei and their energies are the activation energies for nucleation. Since the energy is $\sim n$ [1], it is improbable that microstructures with $n > 2-3$ can be formed. For a given l/n , there are both low-energy ($\xi_I^{MM}(x)$) and high-energy ($\xi_{II}^{MM}(x)$) M_- - M_+ microstructures; $\xi_{II}^{MM}(x)$ is a M_- - M_+ critical nucleus. For the equilibrium A-M interface and $\xi_I^{MM}(x)$ for $n = 1$, there is critical specimen length l_c above which the microstructure is stable under all infinitesimal fluctuations. The critical length for n interfaces is nl_c , despite the fact that their energy $\sim n$.

The GL equation predicts the existence of A-M and M_- - M_+ functionally graded nanophases in specimens whose lengths are greater than l_m but less than the interface width. They could be formed by dissolving material from both sides of a nanofilm or from both the inner and outer surfaces of a nanotube of large radius. When $l = l_m$, then $\xi_s = 1/\sqrt{6}$, which is neither A nor M, but a novel homogeneous nanophase. It corresponds to a maximum of the Landau potential instead of a minimum. These highly energetic nanophases would release energy during a PT to A or M. We believe that our approach can be applied to other GL-type models [7], as well as be extended to higher dimensions.

* * *

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