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A phase field study of morphological instabilities in multilayer thin films

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Abstract

We studied the microstructural evolution of multiple layers of elastically stiff films embedded in an elastically soft matrix using a phase field model. The coherent and planar film/matrix interfaces are rendered unstable by the elastic stresses due to a lattice parameter mismatch between the film and matrix phases, resulting in the break-up of the films into particles. With an increasing volume fraction of the stiff phase, the elastic interactions between neighbouring layers lead to: (i) interlayer correlations from an early stage; (ii) a longer wavelength for the maximally growing wave; and therefore (iii) a delayed break-up. Further, they promote a crossover in the mode of instability from a predominantly anti-symmetric (in phase) one to a symmetric (out of phase) one. We have computed a stability diagram for the most probable mode of break-up in terms of elastic modulus mismatch and volume fraction. We rationalize our results in terms of the initial driving force for destabilization, and corroborate our conclusions using simulations in elastically anisotropic systems.

Keywords: Phase field modelling; Thin films; Asaro–Tiller–Grinfeld instability; Microstructural evolution; Elastically stressed solids

1. Introduction

It is well known that the planar interface of a non-hydrostatically stressed solid in equilibrium with its melt or vapour is unstable with respect to perturbations [1–3]: this instability is known as Asaro–Tiller–Grinfeld (ATG) instability. Based on elastic and interfacial energy considerations alone, Grinfeld showed that, in the absence of interfacial energy, a planar boundary between a solid and its melt or vapour is unstable with respect to perturbations of any wavelength; the interfacial energy sets the lower wavelength limit of this instability [4].

The literature on ATG instabilities is too vast and varied to be summarized here; we refer the interested reader to the excellent monographs of Nozières [5, chapter 1], Pimpinelli and Villain [6] and Freund and Suresh [7], and the reviews (and references therein) of Shchukin and Bimberg [8], Gao and Nix [9], Stangl et al. [10], Johnson and Voorhees [11] and Balibar et al [12] for a summary of the experimental and theoretical studies.

Sridhar et al. [13,14] (hereafter SRS) have shown the crucial role played by an elastic modulus mismatch in promoting ATG instabilities; more specifically, for a misfitting thin film layer embedded in a matrix (both in the presence and absence of externally applied stresses), the planar film/matrix interface is unstable with respect to perturbations as long as the film is elastically stiffer than the matrix. In the case of a non-misfitting film embedded in a matrix under an externally applied stress, the interface is unstable as long as the film and matrix have different elastic constants.

Using a linear stability analysis that accounts for both curvature (or interfacial) and elastic contributions, SRS identify two dominant modes of instabilities that a film/matrix interface can undergo. Depending on whether the undulations of the upper and lower interfaces of a film are out of phase or in phase (see their schematic in Fig. 8

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of Ref. [14]), these modes are known as symmetric and anti-symmetric, respectively. In addition, their stability diagrams show that the anti-symmetric mode is promoted at higher values of the driving force for film destabilization.

While a linear stability analysis is ideal for the study of onset of instability, it cannot accurately predict the evolution, break-up and coarsening of the microstructure. Further, for multilayer films, interlayer correlations are far too complicated to be included in such an analysis; thus, for example, the multilayer part of the study by SRS was carried out for a particular kind of interlayer correlation in which the undulations of the upper interfaces of all the films are in phase.

There are several simulation studies on the elastic stress-induced morphological instabilities in thin films [15–35]; while most of these studies are based on the phase field method [15–27], the rest are based on the phase field crystal [28,29], continuum, sharp-interface [30–33] and atomistic [34,35] models.

All the phase field studies that we are aware of are concerned with the evolution of multilayer films in the presence of the film/vapour (or film/melt) interface. In these studies, the microstructural evolution is influenced by both the multilayer setting and the film/vapour interface, leading to different behaviours of individual layers, depending on their distance from the film/vapour interface. Our study, on the other hand, focuses on a "pure" multilayer geometry. It allows us to determine the effects that arise from the multilayers alone. It also allows us to explore quantitatively the role of system parameters in determining the mode of onset of instability and the maximally unstable wavelength.

This paper is organized as follows: we present a brief outline of the formulation in Section 2; in Section 3 we present our numerical simulation results, and discuss the key features of multilayer evolution in Section 4; we conclude the paper with a short summary.

2. Formulation

In this section, we outline our phase field model for systems that are elastically inhomogeneous and anisotropic. The model is based on the Cahn–Hilliard equation [36]; details of its formulation and numerical implementation can be found in Ref. [37,38].

We consider a binary alloy consisting of two phases, namely, p and m, whose scaled equilibrium compositions in the absence of elastic stresses are unity and zero, respectively. Our phase field formulation starts with the following expression for the total free energy F of a system with spatial variations in composition c:

$$F = N_v \int [Ac^2(1-c)^2 + \kappa(\nabla c)^2] dV + \frac{1}{2}\int \sigma^{el} : \epsilon^{el} dV \qquad (1)$$

where N_v is the number of atoms per unit volume, κ is the (positive) gradient energy coefficient, A is the free energy barrier between the two phases, ϵ^{el} and σ^{el} are the elastic strain and stress fields, respectively, and ':' denotes the tensor inner product. The microstructural evolution in the system is described by the Cahn–Hilliard equation:

$$\frac{\partial c}{\partial t} = \nabla \cdot M \nabla \mu \qquad (2)$$

where μ is the chemical potential, defined as the variational derivative of the free energy with respect to composition:

$$\mu = \frac{\delta(F/N_v)}{\delta c} \qquad (3)$$

The stress, σ^{el}, is obtained by assuming Hooke's law (that is, the phases m and p are linear elastic):

$$\sigma^{el}_{ij} = C_{ijkl}\epsilon^{el}_{kl} \qquad (4)$$

where we have used Einstein's convention of summation over repeated indices. C_{ijkl} is the elastic modulus tensor and ϵ^{el}_{ij} is the elastic strain, given by

$$\epsilon^{el}_{ij} = \epsilon_{ij} - \epsilon^0_{ij} \qquad (5)$$

where ϵ^0_{ij} is the eigenstrain and ϵ_{ij} is the total strain compatible with the displacement u_i:

$$\epsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial r_j} + \frac{\partial u_j}{\partial r_i}\right) \qquad (6)$$

The displacement field is obtained by solving the equation of mechanical equilibrium:

$$\frac{\partial \sigma^{el}_{ij}}{\partial r_j} = 0 \qquad (7)$$

We assume the following composition dependences for the elastic moduli and the eigenstrain:

$$C_{ijkl}(c) = C^{eff}_{ijkl} + \alpha(c)\Delta C_{ijkl} \qquad (8)$$

$$\epsilon^0_{ij}(c) = \beta(c)\epsilon^{\mathrm{T}}\delta_{ij} \qquad (9)$$

where $\alpha(c)$ and $\beta(c)$ are scalar interpolating functions of composition, $C^{eff}_{ijkl} = (C^p_{ijkl} + C^m_{ijkl})/2$ is the arithmetic average of the elastic moduli of the two phases, $\Delta C_{ijkl} = C^p_{ijkl} - C^m_{ijkl}$ is the difference between the elastic modulus of the p phase and that of m phase, ϵ^{T} denotes the strength of the eigenstrain and δ_{ij} is the Kronecker delta.

2.1. Simulation details and parameters

We use a semi-implicit Fourier spectral technique to solve the evolution equation for composition (Eq. 2) in two dimensions, with periodic boundary conditions. The evaluation of the right-hand side (RHS) of Eq. 2 involves not only the composition and its gradient, but also the elastic stress and strain fields. For a given composition field, we obtain the elastic stress and strain fields by solving the equation of mechanical equilibrium (Eq. 7). The solution of the equation of mechanical equilibrium is also obtained using a Fourier-based iterative technique, assuming prescribed displacement conditions (in other words, the homogeneous strain is zero); see Refs. [37–39] for details. Once we obtain the stress and strain fields, the RHS of Eq. 2 can be evaluated, which is then used for time stepping