A VARIATIONAL APPROACH TO STRESS-INDUCED INSTABILITIES IN HETEROEPITAXIAL GROWTH

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ABSTRACT

We study a variational model for shape instabilities that occur in expitaxial growth of thin crystalline films. Such instabilities (also known as Asaro–Tiller–Grinfeld instabilities) result from lattice misfits between the film and the substrate. Equilibrium shapes of the film are defined variationally, as those that minimize a free energy functional, which combines elastic bulk energy and surface energy. Our model involves a scaling parameter K which multiplies the elastic bulk energy. In a 2–dimensionnal setting, we show that when K is smaller than a critical value, films with flat free surfaces are optimal. When K is large, however, corrugated surfaces are preferred.

1 INTRODUCTION

The shape of the free surface of a thin crystalline film growing on a susbtrate may show island–like patterns. These result from surface diffusion of atoms on the free surface, as film tries to relax elastic stresses that are induced by lattice mismatch with the substrate. Experimentally, one observes that films grow flat until a critical mean thickness is reached, after which corrugations or grooves appear. The control of such shape instabilities (also known as Asaro–Tiller–Grinfeld [1, 6]) in the growth process of heteroepitaxial systems (such as e.g. Si/Ge films) is a great challenge. The occurence of instabilities may or may not be desirable. For instance, this mechanism may be used to manufacture quantum devices, where tiny islands of a material spaced by a few hundreds nm are formed.

In most of the litterature on such instabilities (for a review see [5]) the free surface is sought in a parametric form as a function $h(x, y, t)$, as the solution to a PDE of the form

$$V_n \quad = \quad D\frac{\partial^2}{\partial s^2}\left(\gamma\kappa + \frac{1}{2}\,\sigma : e(u)_{/\,\Gamma}\right), \tag{1}$$

where V_n denotes the normal velocity of the free surface Γ, D is a diffusion constant, γ, the surface tension, κ, the surface curvature, and where u, $e(u)$ and σ and are respectively the elastic displacement and the (linearized) deformation and stress tensors. From the physical point of view, this equation expresses the evolution of the surface according to a chemical potential, sum of a (local) surface strain energy density and of a surface energy (surface tension times the curvature of the free surface in the reference state).

Linear stability around a uniform profile has been addressed for the steady-state and time-dependent problems in various configurations: when the film is modeled as a semi-infinite region, when the interaction between film and substrate (rigid or non-rigid) is taken into account. The stabilizing influence of kinetic (deposition) effects has also been studied with this model and a similar model has been investigated in the context of alloy thin films. [4, 5, 11] and the references therein.

At equilibrium, equation (1) reduces to a relation between the strain energy density along the free surface and the curvature

$$\gamma\kappa + \frac{1}{2}\,\sigma : e(u)_{/\Gamma} \quad = \quad \text{constant.} \tag{2}$$

We notice that the above equation is obtained under the hypothesis that the free surface and the elastic displacement are sufficiently smooth.

Instead of seeking the free surface as a solution to (2), we study a model (at the continuum level) where the instability result from the competition between two forms of energy : surface energy and bulk elastic energy. While surface energy promotes configurations with flat free surfaces, an asymptotic argument by M. Grinfeld [6] shows that corrugations may be preferable when minimizing the bulk elastic energy.

Specifically, we assume that the film can be modeled as a linear elastic material, whose Lamé tensor is denoted by A. Equilibrium shapes are defined as minimizers of a free energy functional $E(\Omega, u)$ that depends on the domain Ω occupied by the film and on the elastic displacement u

$$E(\Omega, u) \quad = \quad K \int_\Omega Ae(u) : e(u) + L(\Omega). \tag{3}$$

The term $L(\Omega)$ denotes surface energy. Its precise definition is given below. The parameter K results from the scalings from physical dimensions and is interpreted as the mean thickness of the film. In the minimization of the energy, admissible Ω's and u's are constrained to be periodic (in the directions of the plane of the substrate) to satisfy a volume constraint and boundary conditions that reflect the interaction between film and substrate.

This note is organized as follows : in the next Section, we explain how we define surface energy for very general surfaces, i.e., without *a priori* smoothness assumptions on the surface. In particular, our formulation allows free surfaces with vertical slopes or with cracks. Section 3 concerns approximation of the free energy using a phase field description of the free surface. In section 4, we give a stability result for flat free surfaces when the scaling parameter K is small.

2 THE ENERGY FUNCTIONAL IN 2 DIMENSIONS

Numerical approximations to solutions to (1) have proven very unstable, as the shape of free surfaces tend to become singular and tend to form cusps or deep grooves. In the variational context, Bonnetier et al [3] have analyzed a simplified 1–dimensionnal model where minimizing the energy (in the large K regime) produces a vertical crack in the film. From a mathematical point of view, these observations raise the question of the well–posedness of the minimization of a functional of the form (3) when the surface energy $L(\Omega)$ roughly measures the area of the free surface.

More precisely, let Q denote the x–periodic cylinder $\mathbf{R}/\mathbf{Z} \times \mathbf{R}$ and $Q^+ = \mathbf{R}/\mathbf{Z} \times (0, \infty)$. When the domain Ω of the film is smooth (*i.e.*, Ω lies between the $x-$axis and the graph of a smooth non–negative function $h(x), x \in (0, 1)$) one may define a free energy by

$$E^0(\Omega, u) \quad = \quad K \int_\Omega Ae(u) : e(u) \; + \; \sigma_c \mathcal{H}^1(\partial\overline{\Omega} \cap Q^+) \; + \; \sigma_s \mathcal{H}^1(\partial\overline{\Omega} \setminus Q^+), \tag{4}$$

where σ_c and σ_s denote the surface tensions of the film and of the substrate and where $\mathcal{H}^1$ denotes the one–dimensional Hausdorff (surface) measure. When one tries to minimize E^0

under the constraints

$$\left\{ \begin{array}{l} |\Omega| \;=\; V_0 \\ u(x,0) \;=\; (x,0) \quad \text{for } x \in (0,1) \\ u(1,y) - (1,0) \;=\; u(0,y) \quad \text{for } y \in (0,h(0)) \end{array} \right. \tag{5}$$

it may happen that minimizing sequences (Ω_n) converge to a domain Ω_∞ which is singular and the energy of which cannot be computed by (4).

The remedy is to perform a relaxation, *i.e.*, to generalize the set of admissible shapes in order to allow shapes with cusps and vertical cracks, and to extend accordingly the definition of the functional by computing the lower semi–continuous enveloppe of E^0. In 2 dimensions, Bonnetier and Chambolle [2] gave an explicit form of the relaxed functional when admissible shapes are subgraphs which we recall below:

Let $\mathcal{G}$ denote the set of subgraphs of positive lower semi–continuous functions defined on the torus $\mathbf{R}/\mathbf{Z}$

$$\begin{array}{rcl} \Omega \;\in\; \mathcal{G} & \Leftrightarrow & \Omega = \{(x,y) \in Q \;/\; \; y \le h(x) \;\}\,, \\ & & h \;\text{ l.s.c.}, \quad h \ge 0, \quad \displaystyle\int_0^1 h = V_0. \end{array}$$

For $\Omega \in \mathcal{G}$, let $\Omega^+ = \Omega \cap \{y > 0\}$ and define a set $X(\Omega)$ of admissible displacement by

$$X(\Omega) \;=\; \left\{ \begin{array}{l} u \in L^2_{loc}(\Omega) \quad e(u) \in L^2_{loc}(\Omega) \\ u(x,y) \;=\; (x,0) \quad \text{for } y < 0 \\ u - (x,0) \;\text{ is 1–}x\text{–periodic} \end{array} \right\}.$$

We say that a sequence $(\Omega_n) \subset \mathcal{G}$ converges to a set Ω, if the Hausdorff distance between the complements (Ω_n^c) and Ω^c tends to 0. In this topology, $\mathcal{G}$ is the closure of the set of Lipschitz graphs.

To define the relaxed free energy, let $\Omega \subset \mathcal{G}$. There exists 2 l.s.c. functions h and $\overline{h}$, such that

$$\Omega = \{(x,y) \in Q, \;\; y < h(x)\} \qquad \overset{\circ}{\overline{\Omega}} = \{(x,y) \in Q, \;\; y < \overline{h}(x)\}.$$

For $u \in X(\Omega)$, we set

$$E(\Omega,u) \;=\; K \int_{\Omega \cap Q^+} Ae(u)(x,y) {:} e(u)(x,y)\, dxdy \;+\; L(\Omega), \tag{6}$$

where

$$L(\Omega) \;=\; \sigma_c \mathcal{H}^1(\partial\overline{\Omega} \cap Q^+) \;+\; (\sigma_c \wedge \sigma_s)\mathcal{H}^1(\partial\overline{\Omega} \setminus Q^+) \;+\; 2\sigma_c \sum_{x \in \mathbf{S}^1} \left(\overline{h}(x) - h(x)\right). \tag{7}$$

Notice that the last term in (7) is the contribution of vertical cracks (twice their length). The middle term is the contribution of the parts of the substrate which are not covered by the film. This term is proportional to the lowest surface tension $\sigma_c \wedge \sigma_s$, which reflects the possible presence of a wetting layer, i.e. an infinitesimally thin layer of film.

Equilibrium shapes are defined as global minimizers of

$$\min\left\{ E(\Omega,u) \quad / \ \Omega\in\mathcal{G},\ \ u\in X(\Omega), \quad \text{such that } |\Omega\cap Q^+| = V_0\right\}, \tag{8}$$

and we obtain the following result ([2])

Theorem 1 *There exists an equilibrium state $\Omega\in\mathcal{G}, u\in X(\Omega)$.*

3 APPROXIMATE ENERGIES AND NUMERICAL COMPUTATIONS

Since subgraphs of *l.s.c.* functionals are rather delicate to manipulate, we introduce phase field approximations of the energy functional (6). From a physical point of view, this amounts to replacing the sharp interface between the film (or the substrate) and the degenerate phase above the film, by a diffuse interfacial zone of size ε, where the density of film varies continuously from 1 to 0 (see also Kassner and Misbah[7]). A phase function v, with value 1 in the film and 0 above it, keeps track of the geometry of the interface. The approximate energies are defined as follows: for $\varepsilon>0$ let

$$E_\varepsilon(v,u) \ = \ \int_{Q+} (v(x,y)+\eta_\varepsilon)\mathbf{A}e(u)(x,y){:}e(u)(x,y)\,dxdy \ + \ L_\varepsilon(v), \tag{9}$$

where

$$L_\varepsilon(v) \ = \ 2\sigma_c\left(\frac{4\varepsilon}{\pi^2}\int_{Q+}|\nabla v(x,y)|^2\,dxdy \ + \ \frac{1}{\varepsilon}\int_{Q+} v(x,y)(1-v(x,y))\,dxdy\right). \tag{10}$$

The Cahn–Hilliard energy L_ε is defined for $v\in H^1(Q^+)$ such that $0\le v(x,y)\le 1$, $\partial_y v(x,y)\le 0$ a.e. in Q^+, and $v\ge v_s$ on $S^1\times\{0\}$; and for $u\in H^1_{loc}(\mathbf{R}\times\mathbf{R}_+;\mathbf{R}^2)$ such that $u(x,y)-(x,0)$ is 1–periodic in x and vanishes on $\mathbf{R}\times\{0\}$. The constant $v_s\in(0,1]$ is given by

$$\int_0^{v_s}\sqrt{t(1-t)}\,dt \ = \ \frac{\sigma_c\wedge\sigma_s}{\sigma_c}\int_0^1\sqrt{t(1-t)}\,dt \ = \ \frac{\sigma_c\wedge\sigma_s}{\sigma_c}\frac{\pi}{8}. \tag{11}$$

If v,u do not satisfy these properties we set $E_\varepsilon(v,u)=+\infty$.

The functional L_ε is known to be an approximation, in the sense of Γ–convergence of the perimeter [9]. The following result is proven in [2]

Theorem 2 *If $(v_\varepsilon,u_\varepsilon)$ is a sequence of minimizers for E_ε, subject to $\int_{Q+}v_\varepsilon(x,y)dxdy=1$, then to each limit point u of (u_ε) corresponds a set $\Omega\in\mathcal{G}$, such that $|\Omega\cap Q^+|=1$ and*

$$E(\Omega,u) \ = \ \min_{\tilde\Omega\in\mathcal{G}}\ \min_{\tilde u\in X(\tilde\Omega)} E(\tilde\Omega,\tilde u).$$

From a computational point of view, the approximating energies (9) are easier to handle. For a fixed value of ε, one can write an Euler–Lagrange equation for both fields u_ε and v_ε. We solve these equations iteratively, starting from an initial configuration. At the nth step of the algorithm, the displacement is updated via the resolution of a problem of linear elasticity of the form

$$\operatorname{div}\left((v_n+\eta_\varepsilon)Ae(u_{n+1})\right) \ = \ 0.$$

The marker function v_ε is then upated by minimizing $E_\varepsilon(., u_{n+1})$ using a gradient flow method. Note that the corresponding Euler equation for v is linear:

$$\left\{ \begin{array}{c} \partial_t v \ = \ 2\sigma_c \left(\dfrac{8\varepsilon}{\pi^2} \Delta v - 1/\varepsilon (1-2v) \right) - Ae(u_{n+1}) : e(u_{n+1}) - \lambda \\ 0 \ \leq \ v \ \leq \ 1 \, , \ \ \partial_y v \ \leq \ 0 \, , \end{array} \right.$$

The bounds on v_ε and the constraint that v_ε should be a graph are imposed by truncation, while the volume constraint is imposed via a Lagrange multiplier.

4 A STABILITY RESULT

In this Section, we show the following stability result

Theorem 3 *There exists a constant $K_0 > 0$ such that if $0 < K < K_0$, the shape with flat free surface $\Omega = (0,1) \times (0, V_0)$ is a global minimizer of (8).*

This result shows that the variational problem (8) is consistent with the experimental observations that instabilities only occur after the film has reached a critical thickness.
Sketch of Proof: 1. For simplicity, we assume that $\sigma_s \geq \sigma_c = 1$ and that $V_0 = 1$. Let Ω_0 denote the domain $(0,1) \times (0,1)$ and let u_0 denote the linear displacement solution to

$$E(\Omega_0, u_0) \quad = \quad \inf_{u \in X(\Omega_0)} E(\Omega_0, u).$$

Denoting κ and μ the Lamé coefficients, one easily computes that $E(\Omega_0, u_0) = K \dfrac{4\kappa\mu}{\kappa + \mu} + 1$, which is smaller than 2, if K is sufficiently small. Assume that the statement of the Theorem is false: then, a sequence of numbers $K_j < 1/j$, of domains $\Omega_j \in \mathcal{G}$ and of displacements u_j could be found, such that

$$K_j \int_{\Omega_j} Ae(u_j) : e(u_j) \ + \ L(\Omega_j) \quad < \quad K_j \frac{4\kappa\mu}{\kappa + \mu} + 1. \tag{12}$$

By construction of the relaxed energy, we may assume that the sets Ω_j are smooth. Since $|\Omega_j| = 1$ and $L(\Omega_j) < K_j \dfrac{4\kappa\mu}{\kappa + \mu} + 1$, Golab's Theorem [10] implies that a subsequence (not relabelled) converges to some $\Omega_* \in \mathcal{G}$ for the topology of $\mathcal{G}$, which satisfies

$$|\Omega_*| \ = \ 1 \quad \text{and} \quad L(\Omega_*) \leq 1.$$

The isoperimetric inequality forces then $\Omega_* = \Omega_0$.
2. Given $\delta > 0$, the convergence of Ω_j to Ω_0 imply that when j is sufficiently large, the free surface $\partial\Omega_j \cap \{y > 0\}$ lies in the set $(0,1) \times (1-\delta, 1+\delta)$, *i.e.*, in a δ–neighborhood of the flat free surface. In particular, the term $L(\Omega_j)$ is then simply the length of the upper boundary of the smooth domain Ω_j. Using the same analysis as in Bonnetier et al. [3] (see estimate 3.3), one can easily show that

$$L(\Omega_j) \quad \geq \quad (1 + 8\delta_j^2)^{1/2},$$

where δ_j is the Hausdorff distance between Ω_j and Ω_0.

3. Since the sets Ω_j are smooth, the corresponding elastic energy can be estimated as in the computations of M. Grinfeld [6]

$$\int_{\Omega_j} Ae(u_j) : e(u_j) \quad = \quad \int_{\Omega_0} Ae(u_0) : e(u_0) - R_j\delta_j^2 + o(\delta_j^2),$$

where R_j is a positive term, bounded independently from j. It follows that

$$\begin{aligned} E(\Omega_j, u_j) &\geq \int_{\Omega_0} Ae(u_0) : e(u_0) - R_j\delta_j^2 + o(\delta_j^2) + (1 + 8\delta_j^2)^{1/2} \\ &= K_j \frac{4\kappa\mu}{\kappa+\mu} + 1 + \delta_j^2(4 - K_jR_j) + o(\delta_j^2), \end{aligned}$$

which contradicts (12).
4. When K is large, one easily finds examples of corrugated surfaces with a free energy lower than $E(\Omega_0, u_0)$.

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