Scaling law and reduced models for epitaxially strained crystalline films

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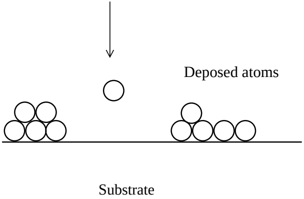
Joint work with P. Bella and B. Zwicknagl

Introduction

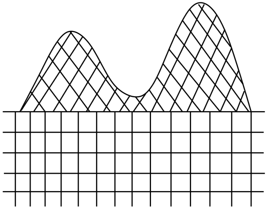
Epitaxially strained crystalline films are obtained by deposing thin layers on a thick substrate



Example : In-GaAs/GaAs or SiGe/Si.

Governing mechanism

There is a mismatch between the lattice parameters of the two crystals



The deposit layer is strained and the atoms try to rearrange for releasing elastic energy but this migration is also energetically expensive

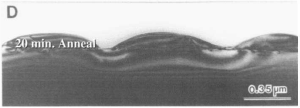
$\Longrightarrow$ interaction between bulk and surface energy.

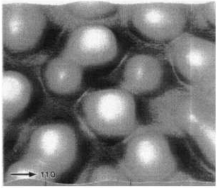
Numerical and experimental observations

Existence of several regimes depending on the volume of the thin layer and of the mismatch

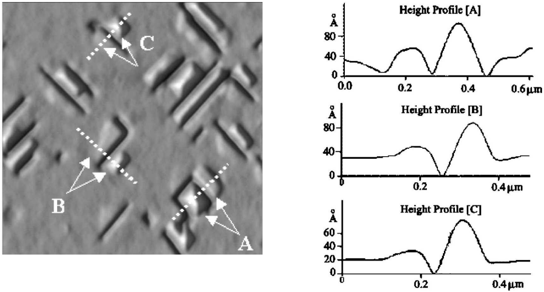
- For small volumes, the flat configuration is favored
- Above a certain threshold, the flat configuration is not stable anymore and the film develops corrugations
- For higher values of the volume/mismatch, there is formation of isolated islands

Goal: Understand these different regimes.



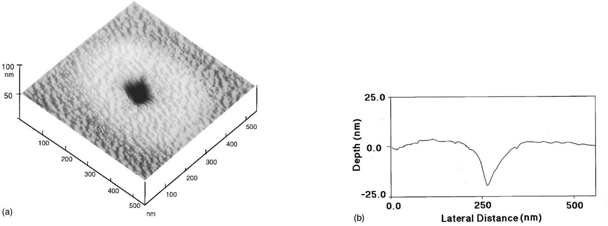


Surface roughening in SiGe/Si, images from Gao, Nix, *Surface roughening of heteroepitaxial thin films*, 1999.



Formation of islands, images from Gray, Hull and Floro *Formation of one-dimensional surface grooves from pit instabilities in annealed SiGe/Si(100) epitaxial films*, 2004.

Experimental results



Example of cusps, images from Chen, Jesson, Pennycook, Thundat, and Warmack, *Cuspidal pit formation during the growth of SixGe1-x strained films*, 1995

Numerical simulations

Numerical simulations from Bonnetier and Chambolle, *Computing the Equilibrium Configuration of Epitaxially Strained Crystalline Films*, 2002.

See also the numerical simulations of Smereka and of the University of Cambridge, DoITPoMS, http://www.doitpoms.ac.uk/tlplib/epitaxial-growth/index.php

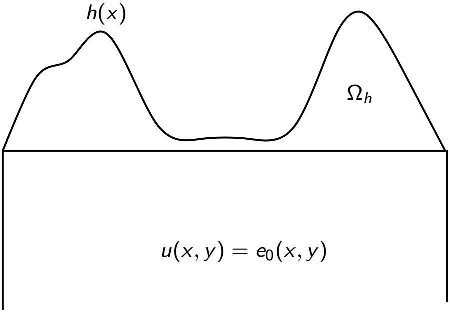
Applications

These epitaxially grown thin films are used for

- Optical and optoelectric devices (quantum dot laser).
- Semiconductors.
- Information storage.
- Nanotechnology.

The Mathematical Model

The film is taken to be the subgraph Ω_h of a function $h : [0,1] \to \mathbb{R}^+$



The substrate is considered as rigid hence in the substrate, the deformation is equal to $e_0(x,y)$ where e_0 is the mismatch.

The energy

Let $W : \mathbb{R}^4 \to \mathbb{R}^+$ be the stored elastic energy then we consider the variational problem:

$$F_{d,e_0}(u,h) := \int_{\Omega_h} W(\nabla u) + \int_0^1 \sqrt{1 + |h'|^2}$$

under the conditions that

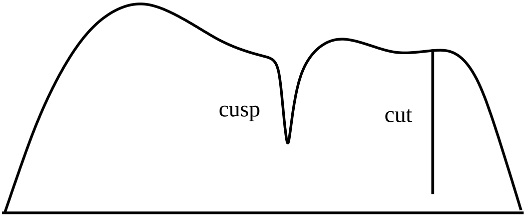
$$u(x,0) = e_0(x,0) \qquad \text{and} \qquad \int_0^1 h = d$$

Remark: most of the works consider energies W depending only on the symmetric part of the gradient.

Contributions of each term in the energy

- Due to the mismatch, there are no stress free configurations.
- In order to release elastic energy, the bulk term favors creation of singularities.
- On the other hand, the surface term tends to avoid too many oscillations.

Regularity results (in the geometrically linear setting)



Theorem [Chambolle-Larsen 03, Fonseca-Fusco-Leoni-Morini 07]

The profile h is regular out of a finite number of cusps and cuts. Moreover the film satisfies the zero angle condition.

Regularity results continued

Theorem [Fusco-Morini 12]

- For small mismatch, the flat configuration is minimizing (no matter how big is d).
- For greater mismatch, the following holds:
  1. for $d \leq d_0$, the flat configuration is minimizing
  2. for $d \leq d_1$ the flat configuration is locally minimizing
  3. for $d \leq d_2$, the flat configuration is not locally minimizing but every minimizer is smooth

Other results in the litterature

- Physical and engineering: Spencer-Meiron 94, Spencer-Tersoff 10, Gao-Nix 99.
- Regularity, relaxation and approximation: Bonnetier-Chambolle 02, Chambolle-Larsen 03, Fonseca-Fusco-Leoni-Morini 07, Chambolle-Solci 07, Fusco-Morini 12, Bonacini 14.
- Time evolution: Fonseca-Fusco-Leoni-Morini 12, Piovano 12.

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No rigourous result on the formation of the islands!

The main result

We will assume that

Hypothesis

(H1) $W \geq 0$

(H2) there exists $C > 0$ and $p > 1$ such that

$$C(|A|^p + 1) \geq W(A) \geq \frac{1}{C}(|A|^p - 1) \qquad \forall A \in \mathbb{R}^{2\times 2}.$$

Theorem

Under these assumptions, for every $e_0 > 0$ and $d > 0$ there holds

$$\min_{u,h} F_{e_0,d}(u,h) \simeq \max(1, d, e_0^{p/3} d^{2/3}).$$

<u>Remark:</u>

- Thanks to (H2), it is enough considering $W(\nabla u) = |\nabla u|^p$.
- Works also in the geometrically linear setting.

Heuristic explanation of the scaling

We consider for simplicity here $p = 2$ so that

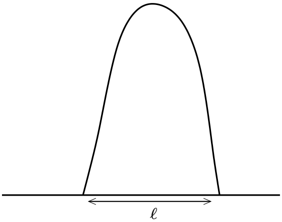
$$F_{e_0,d}(u,h) = \int_{\Omega_h} |\nabla u|^2 + \int_0^1 \sqrt{1+|h'|^2}$$

If $\Omega_h \cap \{y=0\} = [a, a+\ell]$ then since $|\Omega_h| = d$,

$$\int_0^1 \sqrt{1+|h'|^2} \geq \frac{d}{\ell}.$$

On the other hand

$$\min_{u(x,0)=e_0(x,0)} \int_{\Omega_h} |\nabla u|^2 \simeq e_0^2 |u|^2_{H^{1/2}(a,a+\ell)} \simeq e_0^2 \ell^2$$



Putting these together we find that

$$F_{e_0,d}(u,h) \gtrsim e_0^2\ell^2 + \frac{d}{\ell}$$

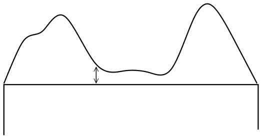
Optimizing in ℓ, we find that $\ell_{\min} \simeq \min(1, \left(\frac{d}{e_0^2}\right)^{1/3})$. So that two regimes appear:

- If $\left(\frac{d}{e_0^2}\right)^{1/3} \leq 1$, we have $\ell_{\min} = \left(\frac{d}{e_0^2}\right)^{1/3}$ and $\min F_{e_0,d} \simeq e_0^{2/3} d^{2/3}$.
- If $\left(\frac{d}{e_0^2}\right)^{1/3} \geq 1$, the flat configuration is favored and $\min F_{e_0,d} \simeq e_0^2 + d \simeq d$.

Difficulty:

when $h(x) \ll 1$, the constant in

the trace inequality degenerate i.e.



$$\min_{u(x,0)=e_0(x,0)} \int_{\Omega_h} |\nabla u|^2 \not\gtrsim e_0^2 |u|^2_{H^{1/2}(a,a+\ell)}$$

Reduced models

In order to study the asymptotic behavior of the energy, we rescale the domains and set

- $\widetilde{h} := h/d$
- $\Omega_{\tilde{h}} := \{(x, y) \ : \ (x, dy) \in \Omega_h\}$
- $\widetilde{u}(x, y) = u(x, dy)$

Dropping the tildes, the energy now reads

$$F_{d,e_0}(u, h) = d\left[\int_{\Omega_h} W\left(\frac{\partial u}{\partial x}, \frac{1}{d}\frac{\partial u}{\partial y}\right) dxdy + \int_0^1 \sqrt{\frac{1}{d^2} + |h'|^2} dx\right]$$

for (u, h) such that $\int_0^1 h dx = 1$, and $u \in W^{1,p}(\Omega_h)$ with

$u(x, 0) = e_0(x, 0)$.

Γ-convergence

Definition

We say that a sequence of functionals F_n Γ-converges to F if

$\forall u_n$ with $\sup F_n(u_n) < +\infty$, $\exists u$ such that $u_n \to u$ (up to a subsequence) and

$$\liminf F_n(u_n) \geq F(u)$$

$\forall u$, $\exists u_n \to u$ with

$$\limsup F_n(u_n) \leq F(u)$$

The surface dominant regime $F_{d,e_0} \simeq d$

We divide the energy by d and obtain the rescaled energy

$$F_d(u,h) := \int_{\Omega_h} W\left(\frac{\partial u}{\partial x}, \frac{1}{d}\frac{\partial u}{\partial y}\right) dxdy + \int_0^1 \sqrt{\frac{1}{d^2} + |h'|^2} dx.$$

In this regime, the surface energy is the dominating term, and the limit functional is given by

$$\bar{F}(h) := \int_0^1 |h'| + 2\mathcal{H}^1(\Gamma_{cuts}).$$

The minimizer of $\bar{F}$ is the flat configuration $h \equiv 1$

The elastic dominant regime, $F_{d,e_0} \simeq e_0^{p/3} d^{2/3}$

The relevant parameter is $\eta := \left(\frac{d}{e_0^p}\right)^{1/3} \to 0$, so that the energy scales like $\frac{d}{\eta}$. We thus consider the normalized energy:

$$F_\eta(u,h) := \eta \left[\int_{\Omega_h} W\left(\frac{\partial u}{\partial x}, \frac{1}{d}\frac{\partial u}{\partial y}\right) dxdy + \int_0^1 \sqrt{\frac{1}{d^2} + |h'|^2} dx \right].$$

Notice that in this case, no bound on the total variation of h is available and we expect that the configuration will get more and more irregular.

The convergence result

Let $\mu := h\ dx$ and

$$F_\eta(\mu) := \min_{u(x,0)=(x,0)} F_\eta(u,h).$$

Theorem

The following Γ-convergence holds :

$$\Gamma - \lim_{\eta \to 0} F_\eta = G$$

where

$$G(\mu) = \begin{cases} 3C_W^{1/3} \sum_{i=1}^{+\infty} d_i^{2/3} & \text{if } \mu = \sum_{i=1}^{+\infty} d_i \delta_{c_i} \quad \sum_{i=1}^{+\infty} d_i = 1 \\ +\infty & \text{elsewhere.} \end{cases}$$

Remark: The minimizer of the limit functional, i.e.

$$\min \left\{ \sum_{i=1}^{+\infty} d_i^{2/3} \ : \ \sum_{i=1}^{+\infty} d_i = 1 \right\}$$

is given by a single Dirac mass, i.e. $d_1 = 1$ and $d_i = 0$ for $i > 1$.

Few issues

(i) This analysis seems to heavily relies on the fact that we are on a bounded domain.

(ii) One would like to understand also the asymptotic shape of the islands.

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(i) This analysis seems to heavily relies on the fact that we are on a bounded domain.

(ii) One would like to understand also the asymptotic shape of the islands.

To understand these issues we study the functional

$$F(e_0, d) = \inf \left\{ \int_{\Omega_h} |\nabla u|^2 + \int_{\mathbb{R}} \sqrt{1 + |h'|^2} - 1 : \int_{\mathbb{R}} h = d,\ u(x, 0) = e_0 x \right\}.$$

The small slope approximation

We first study the small slope approximation $\sqrt{1+x^2} \approx 1 + \frac{x^2}{2}$

$$F^s(e_0, d) = \inf\left\{ \int_{\Omega_h} |\nabla u|^2 + \int_{\mathbb{R}} |h'|^2 : \int_{\mathbb{R}} h = d,\ u(x,0) = e_0 x \right\}.$$

Theorem

We have the following scaling law

$$F^s(e_0, d) \simeq \min\left\{e_0^2 d,\ d^{4/5} e_0^{6/5}\right\}.$$

Remark: for $e_0^4 d \gtrsim 1$, the typical island is of height $e_0^{2/5} d^{3/5}$ and of width $\left(\frac{d}{e_0}\right)^{2/5}$.

Existence vs nonexistence of minimizers

Theorem

There exists V_0 such that:

(i) if $e_0^4 d < V_0$ then $F^s(e_0, d) = e_0^2 d$ and there exists no minimizer of $F^s(e_0, d)$,

(ii) if $e_0^4 d > V_0$ then $F^s(e_0, d) < e_0^2 d$ and there exists a (smooth) minimizer of $F^s(e_0, d)$ which has connected and compact support.

Moreover, for $e_0^4 d = V_0$ there also exists a minimizer.

The asymptotic shape

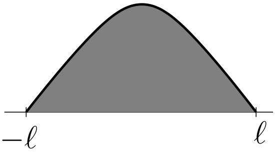
Using Γ-convergence techniques, we obtain

Theorem

Let $V = e_0^4 d$ and let h_V be a minimizer of $F^s(e_0, d)$ and let $\tilde{h}_V(x) = e_0^{-2/5} d^{-3/5} h_V \left(\left(\frac{d}{e_0} \right)^{2/5} x \right)$ then

$$\lim_{V \to +\infty} \tilde{h}_V = \frac{3}{2} \ell^{-3} \left(\ell^2 - x^2 \right) \quad x \in [-\ell, \ell]$$

where $\ell := \left(\frac{9}{16 C_W} \right)^{1/5}$.



The large slope approximation

We can similarly investigate the large slope approximation $\sqrt{1+x^2} \approx |x|$

$$F^L(e_0, d) = \inf \left\{ \int_{\Omega_h} |\nabla u|^2 + \int_{\mathbb{R}} |h'| : \int_{\mathbb{R}} h = d,\ u(x,0) = e_0 x \right\}.$$

Theorem

We have the following scaling law

$$F^L(e_0, d) \simeq \min\left\{e_0^2 d,\ d^{2/3} e_0^{2/3}\right\}.$$

Remark: for $e_0^4 d \gtrsim 1$, the typical island is of height $e_0^{2/3} d^{2/3}$ and of width $\left(\frac{d}{e_0^2}\right)^{1/3}$ as in the compact case.

Existence vs nonexistence of minimizers

In the large slope approximation we can only prove the weaker statement

Theorem

(i) If $F^L(e_0, d) < e_0^2 d$ and there exists a minimizer of $F^L(e_0, d)$ which has connected and compact support.

(ii)
$$\lim_{e_0^4 d \to 0} \frac{F^L(e_0, d)}{e_0^2 d} = 1$$

The asymptotic shape

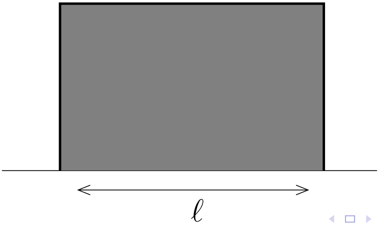
Using Γ-convergence techniques, we obtain

Theorem

Let $V = e_0^4 d$ and let h_V be a minimizer of $F^L(e_0, d)$ and let $\tilde{h}_V(x) = (e_0 d)^{-2/3} h_V\left(\left(\frac{d}{e_0^2}\right)^{1/3} x\right)$ then

$$\lim_{V \to +\infty} \tilde{h}_V = \frac{1}{\ell}\chi_{[0,\ell]}$$

where $\ell := \left(\frac{1}{2C_W}\right)^{1/3}$.



Walfrido 'Morning in the Tropic'

Thank you for your attention!