Surface Relaxation versus the Ehrlich-Schwoebel Effect in Thin-Film Growth

Bo Li
Department of Mathematics
University of California, San Diego

Research supported in part by DOE and NSF

References

[1] B. Li and J.-G. Liu, European J. Applied Math, 14, 713-743, 2003.

[2] B. Li and J.-G. Liu, J. Nonlinear Sci., 14, 429-451, 2004.

[3] B. Li, SIAM J. Math. Anal., 38, 16-36, 2006.

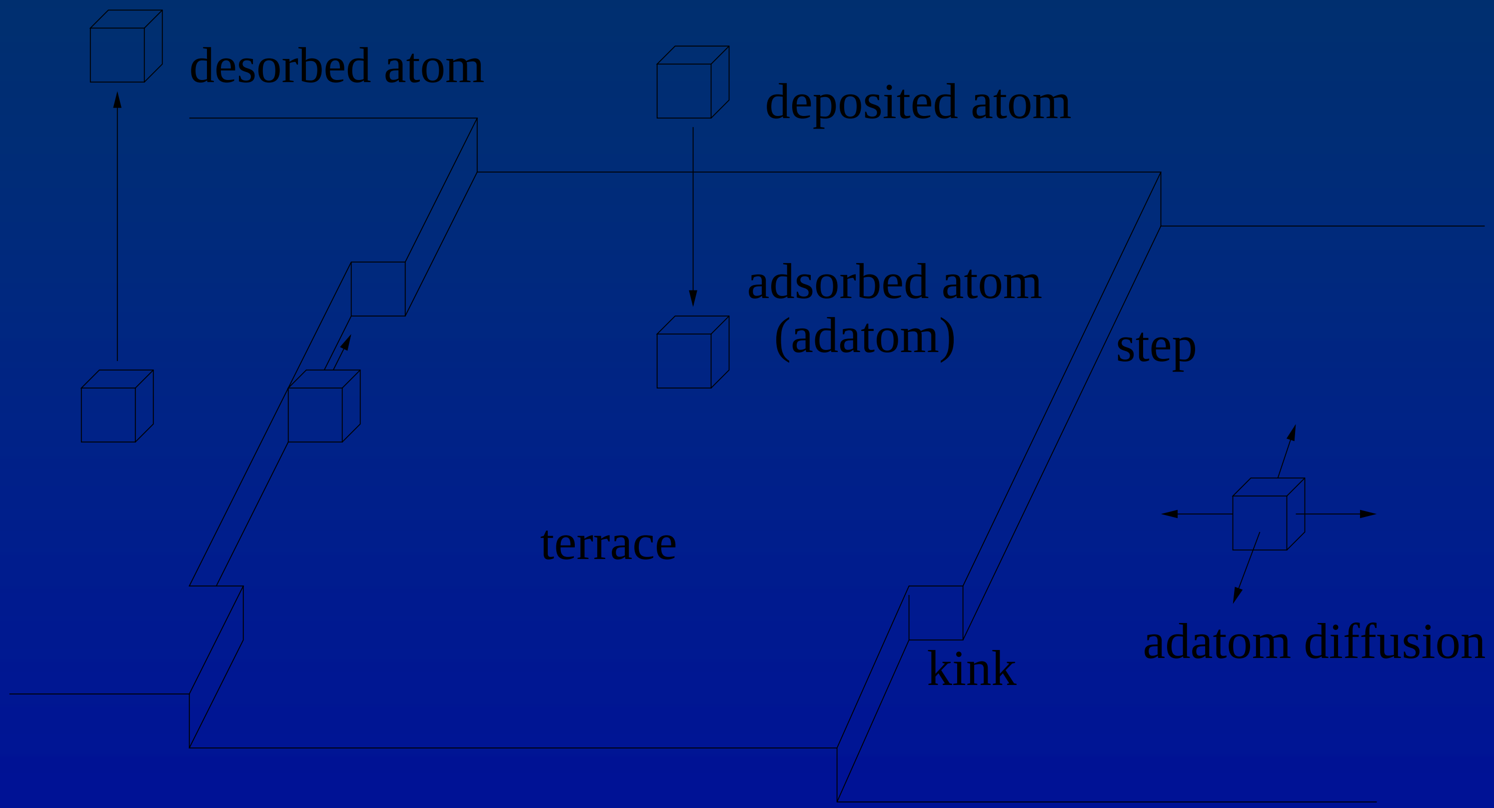
[4] B. Li, Nonlinerity, 19, 2581-2603, 2006.

OUTLINE

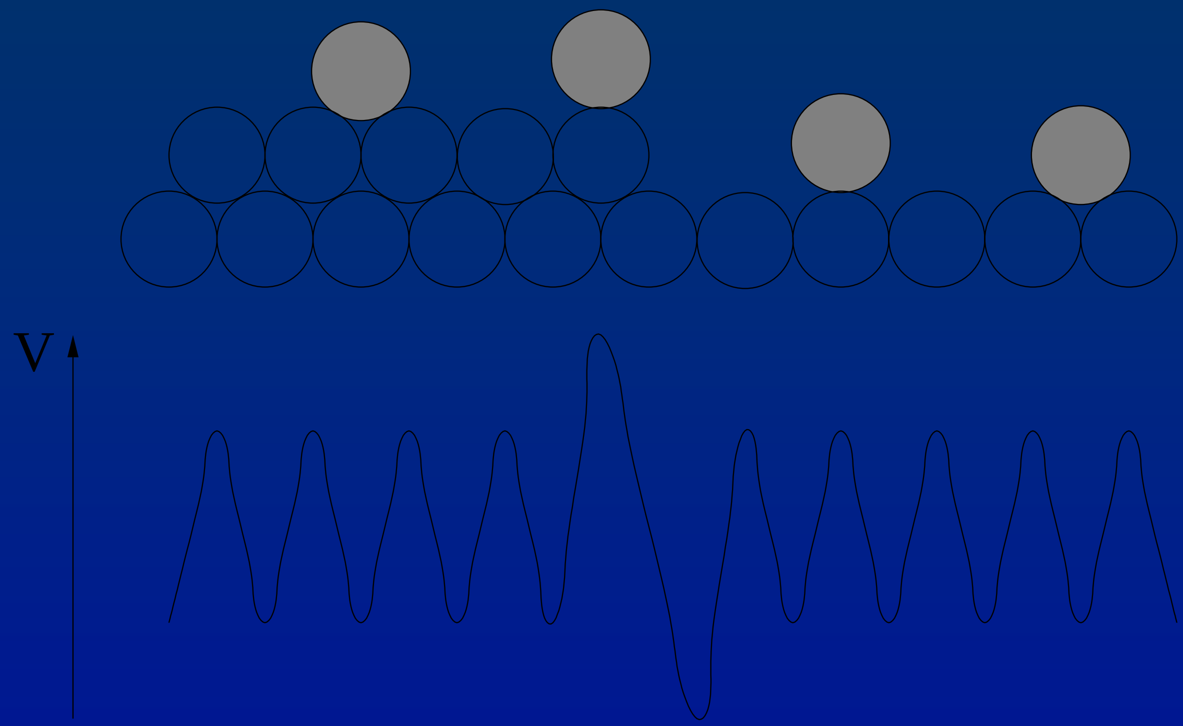
1. Introduction
2. Continuum Models
3. Energy Minimization
4. Scaling Laws
5. Conclusions

1. Introduction

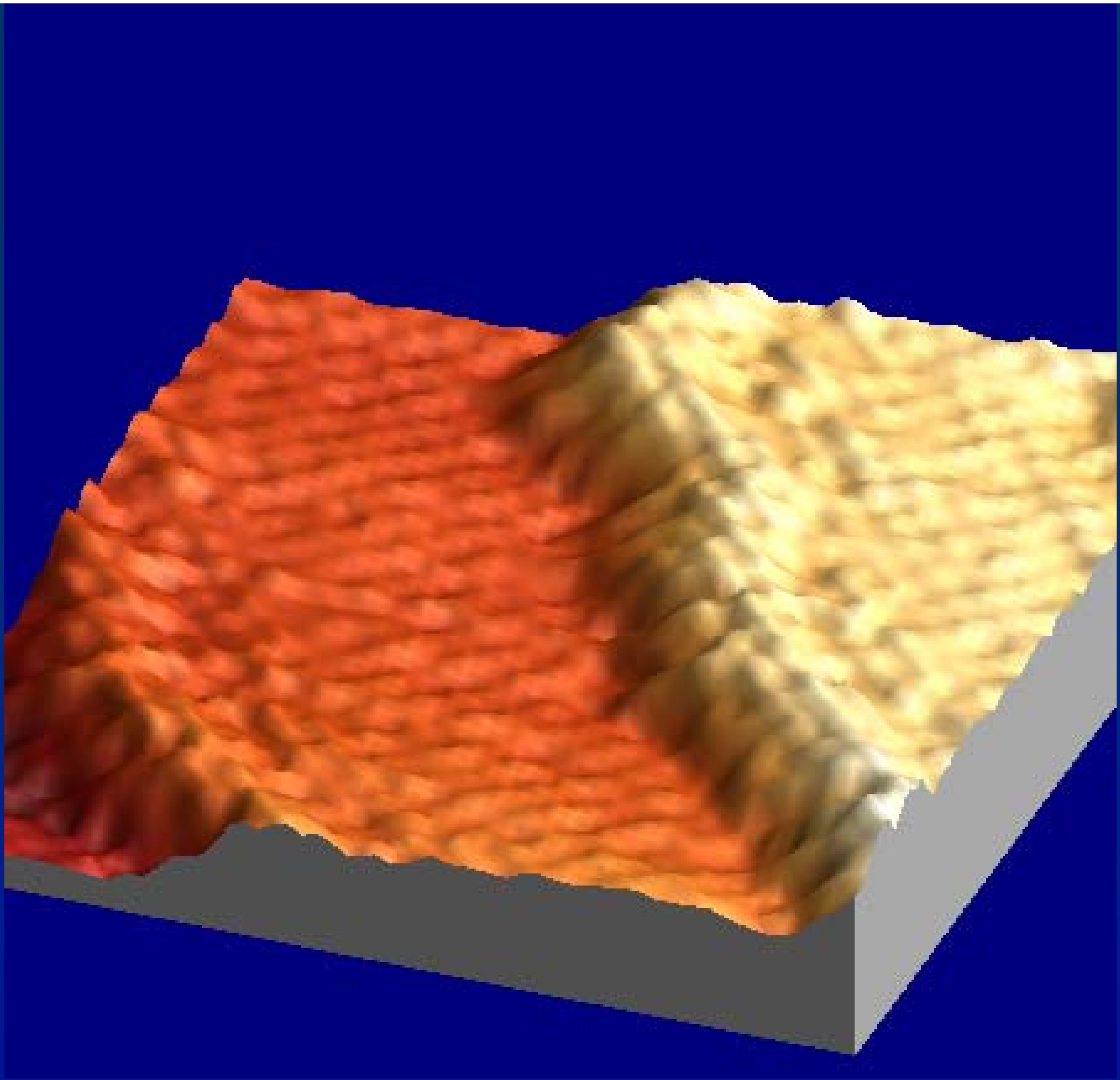
Microscopic processes in thinfilm epitaxy



The Ehrlich-Schwoebel (ES) barrier (1966, 1969)



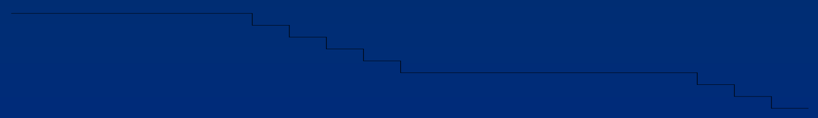
- Low temperature
- Metals and semiconductors



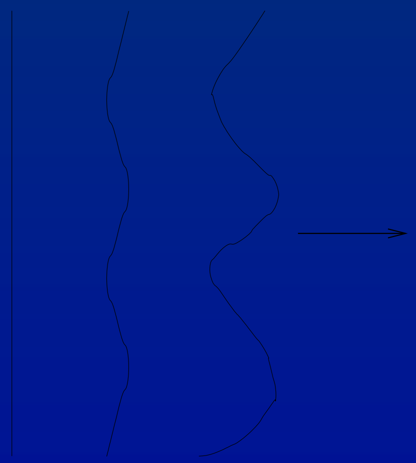
Monatomic Fe chains on Cu(111) vicinal surface, by Jiandong Gao (cf. Phys. Rev. B, 73, 193405, 2006).

Consequences of the ES effect

- Uphill current destabilizing nominal surfaces, but stabilizing vicinal surfaces with large slope, preventing step bunching.



- The Bales-Zangwill instability.

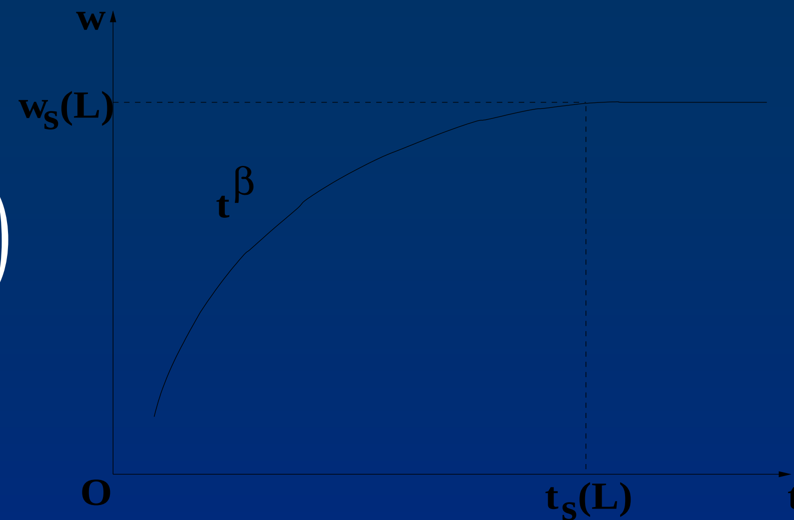


- Kinetic roughening of film surface: 2D to 3D growth, mound formation, etc.

Scaling laws

$$\langle h(x,t)h(0,t)\rangle = [w(t)]^2 g\left(\frac{|x|}{\lambda(t)}\right)$$

$$w(t) = L^{\alpha} f(t/L^{z})$$



- Interface width

$$w(t) \sim t^{\beta}$$

β : growth exponent

- Mound lateral size

$$\lambda(t) \sim t^{n}$$

n : coarsening exponent

- Saturation width

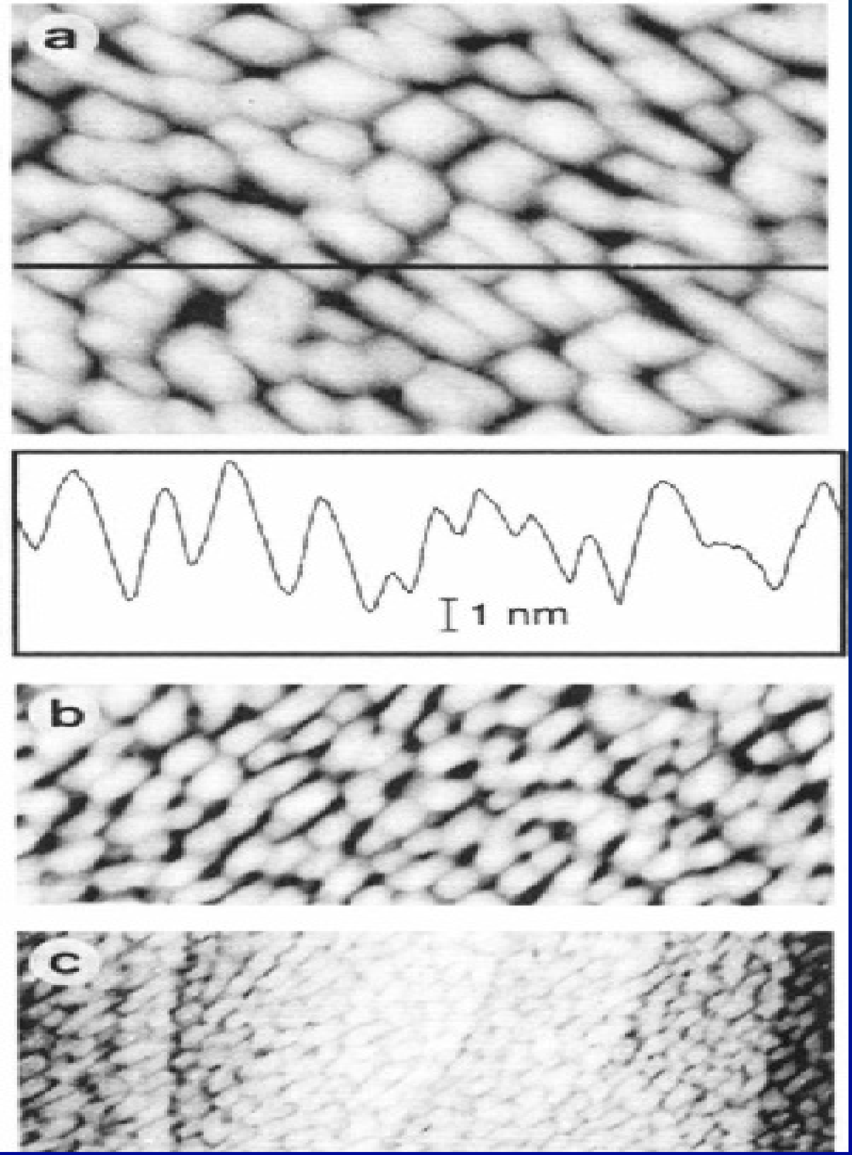
$$w_s(L) \sim L^{\alpha}$$

α : roughness exponent

- Saturation time

$$t_s(L) \sim L^{z}$$

$z = \alpha / \beta$: dynamic exponent



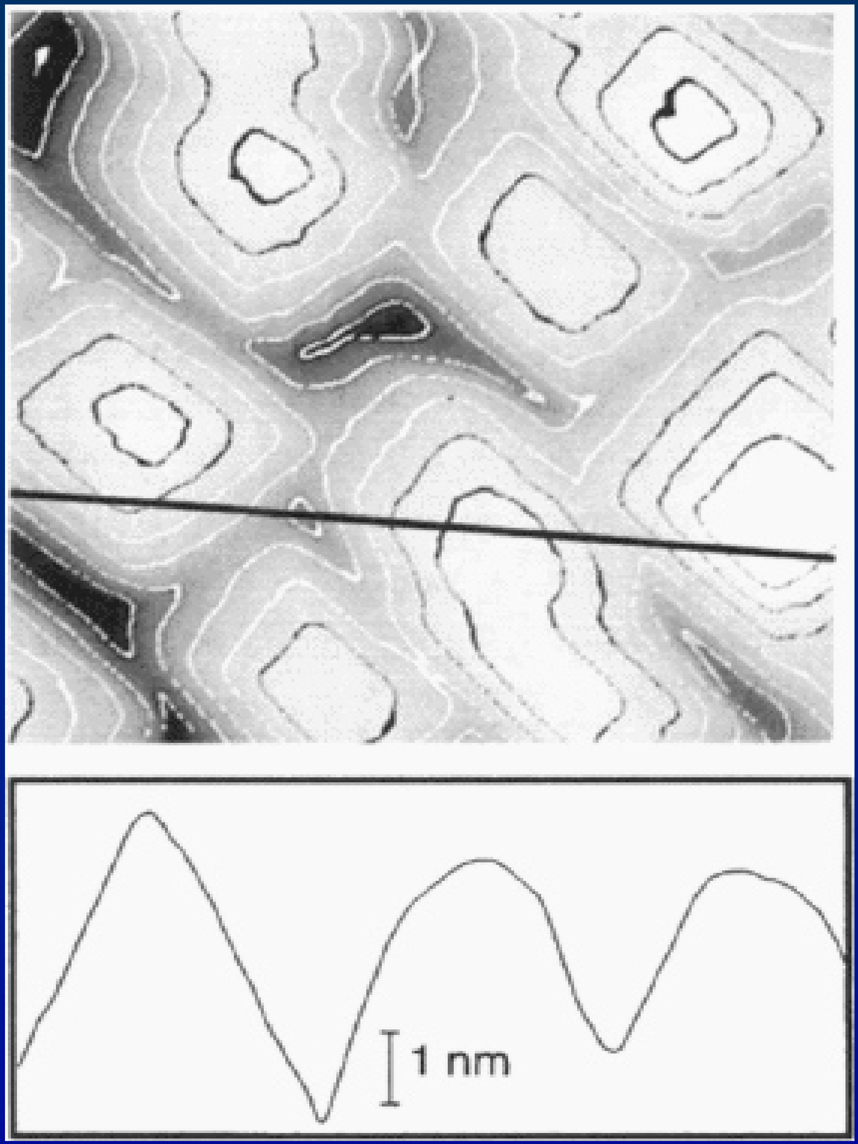
Fe(001) /Mg(001)

(a) Area = 200 nm x 160 nm
Thickness = 300 nm

(b) Area = 200 nm x 80 nm
Thickness = 50 nm

(c) Area = 300 nm x 120 nm
Thickness = 11 nm

Thuermer *et al*., PRL, 75, 1767,1995



- Top

  Fe(001)/Mg(001) film with contours of equal height separated by 1 nm.

  Area = 65 nm x 65 nm, thickness = 300 nm.

- Bottom

  Scan along the marked line in the top view.

Thuermer *et al*., PRL, 75, 1767,1995

surface relaxation + the ES effect

⇓

scaling ?

2. Continuum Models

h: coarse-grained height

- Mass conservation $$\partial_t h + \nabla \cdot j = F$$
- Surface relaxation $$j_{RE} = (-1)^m M_m \nabla \Delta^{m-1} h$$

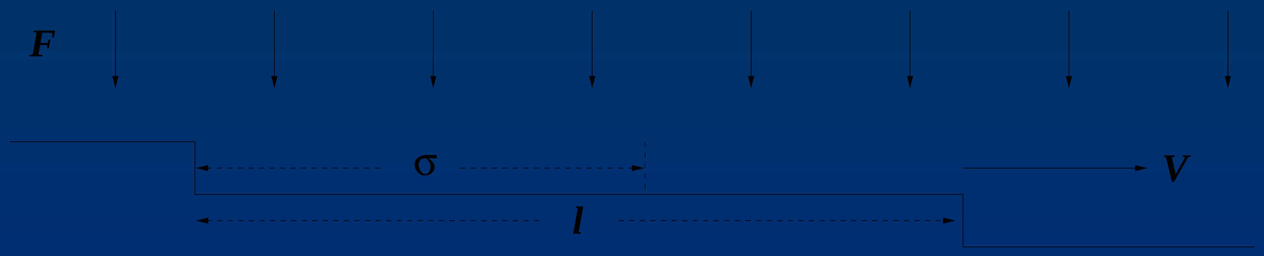
$m = 2$: surface diffusion (Herring-Mullins 1951,1957)

$m = 3$: Fe(001) homoepitaxy (Stroscio *et al*. 1995)

$m \geq 3$: anisotropic surface energy approximation (Stewart-Goldenfeld 1992, Liu-Metiu 1993)

- The ES effect $$j_{ES} = \begin{cases} \dfrac{F \nabla h}{|\nabla h|^2} & \text{infinite ES} \\ \dfrac{FS\sigma^2 \nabla h}{1 + \alpha^2 \sigma^2 |\nabla h|^2} & \text{finite ES} \end{cases}$$

Derivation (Johnson *et al*., PRL, 72, 116, 1994)



$s = |\nabla h| = 1/l$: macroscopic slope

σ: adatom diffusion distance

- Infinite ES barrier
  - Case 1: $s \ll 1$ and $\sigma < l$.

$$j_{ES} = (F\sigma)\times(\sigma/l) = F\sigma^2\nabla h$$

  - Case 2: $s \gg 1$ and $\sigma > l$.

$$j_{ES} = F\sigma^2 f(\sigma/l)\nabla h$$

$$f(x) \sim 1/x^2 \Rightarrow j_{ES} \sim (F/s^2)\nabla h$$

- Finite ES barrier: interpolation!

Infinite ES barrier: Villain's model

$$\partial_t h = (-1)^{m-1} M_m \Delta^m h - \nabla \cdot \left(\frac{F \nabla h}{|\nabla h|^2}\right)$$

$$E(h) = \int [\tfrac{M_m}{2} |\partial^m h|^2 - F \log |\nabla h|] dx$$

Finite ES barrier: the Michigan model

$$\partial_t h = (-1)^{m-1} M_m \Delta^m h - \nabla \cdot \left(\frac{FS\sigma^2 \nabla h}{1+\alpha^2\sigma^2 |\nabla h|^2}\right)$$

$$E(h) = \int [\tfrac{M_m}{2} |\partial^m h|^2 - \tfrac{FS}{2\alpha^2} \log(1+\alpha^2\sigma^2 |\nabla h|^2)] dx$$

- Co-moving frame: $h \to h - Ft$
- Periodical boundary conditions
- ``Energy-driven'' system: $\partial_t h = -\delta E(h)$
- Notation: $|\partial^m h|^2 = \begin{cases} |\Delta^{m/2} h|^2 & m = \text{even} \\ |\nabla \Delta^{(m-1)/2} h|^2 & m = \text{odd} \end{cases}$

Rescale: $h \to \eta h,\ x \to \xi x,\ t \to \zeta t,\ E \to eE$

Infinite ES barrier

$$\xi = \left(\tfrac{F}{M_m}\right)^{1/2m},\quad \eta = 1,\quad \zeta = M_m \xi^{2m},\quad e = \tfrac{1}{F}$$

$$\partial_t h = (-1)^{m-1} \Delta^m h - \nabla \cdot \left(\frac{\nabla h}{|\nabla h|^2} \right)$$

$$E(h) = \int [\tfrac{1}{2} |\partial^m h|^2 - \log |\nabla h|] dx$$

Finite ES barrier

$$\xi = \left(\tfrac{FS\sigma^2}{M_m}\right)^{1/(2m-2)},\ \eta = \alpha\sigma\xi,\quad \zeta = M_m \xi^{2m},\ e = \tfrac{\alpha^2}{FS}$$

$$\partial_t h = (-1)^{m-1} \Delta^m h - \nabla \cdot \left(\frac{\nabla h}{1+|\nabla h|^2} \right)$$

$$E(h) = \int \tfrac{1}{2} [|\partial^m h|^2 - \log(1 + |\nabla h|^2)] dx$$

Instabilities (Villain 91, Rost-Krug 94, Li-Liu 03)

$$\partial_t h = \frac{|\nabla h|^2 - 1}{(1+|\nabla h|^2)^2}\partial_{\|}^2 h - \frac{|\nabla h|}{1+|\nabla h|^2}\kappa - (-1)^{m-1}\Delta^m h_1$$

$$h = h_0 + \varepsilon h_1 + \varepsilon^2 h_2 + \varepsilon^3 h_3 + \cdots$$

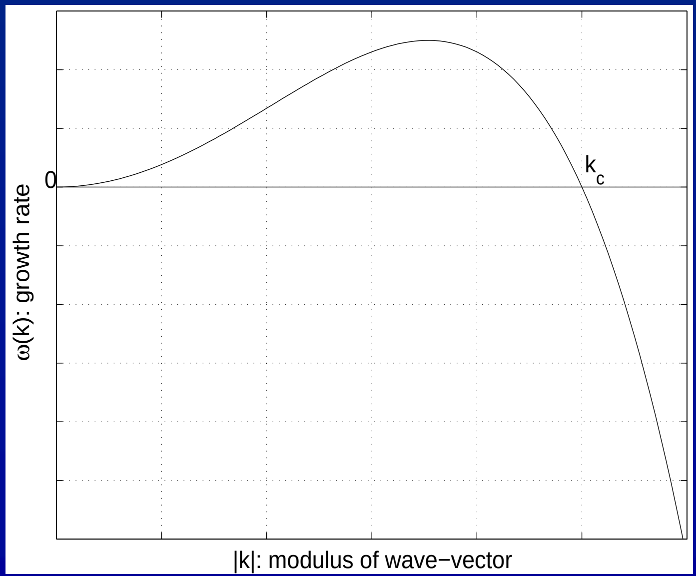
$$h_0(x) = b \cdot x$$

- Linear theory

$$\partial_t h_1 = \frac{|b|^2 - 1}{(1+|b|^2)^2}\partial_{\|}^2 h_1 - \frac{1}{1+|b|^2}\partial_{\perp}^2 h_1 - (-1)^{m-1}\Delta^m h_1$$

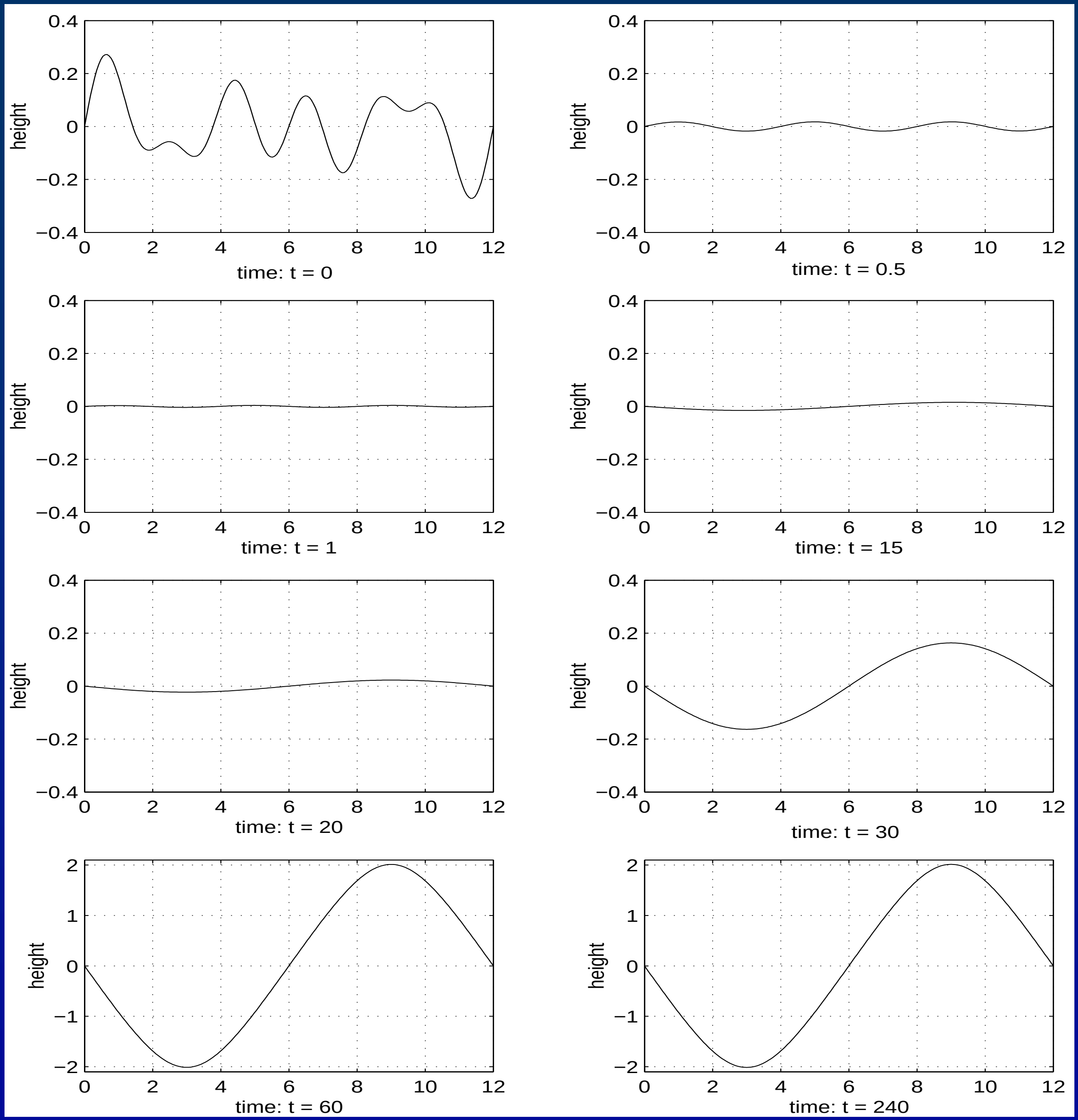
$$b = 0: \quad \omega(k) = k^2 - k^{2m}$$

$$[k_c = (\frac{FS\sigma^2}{M_m})^{1/(2m-2)}]$$

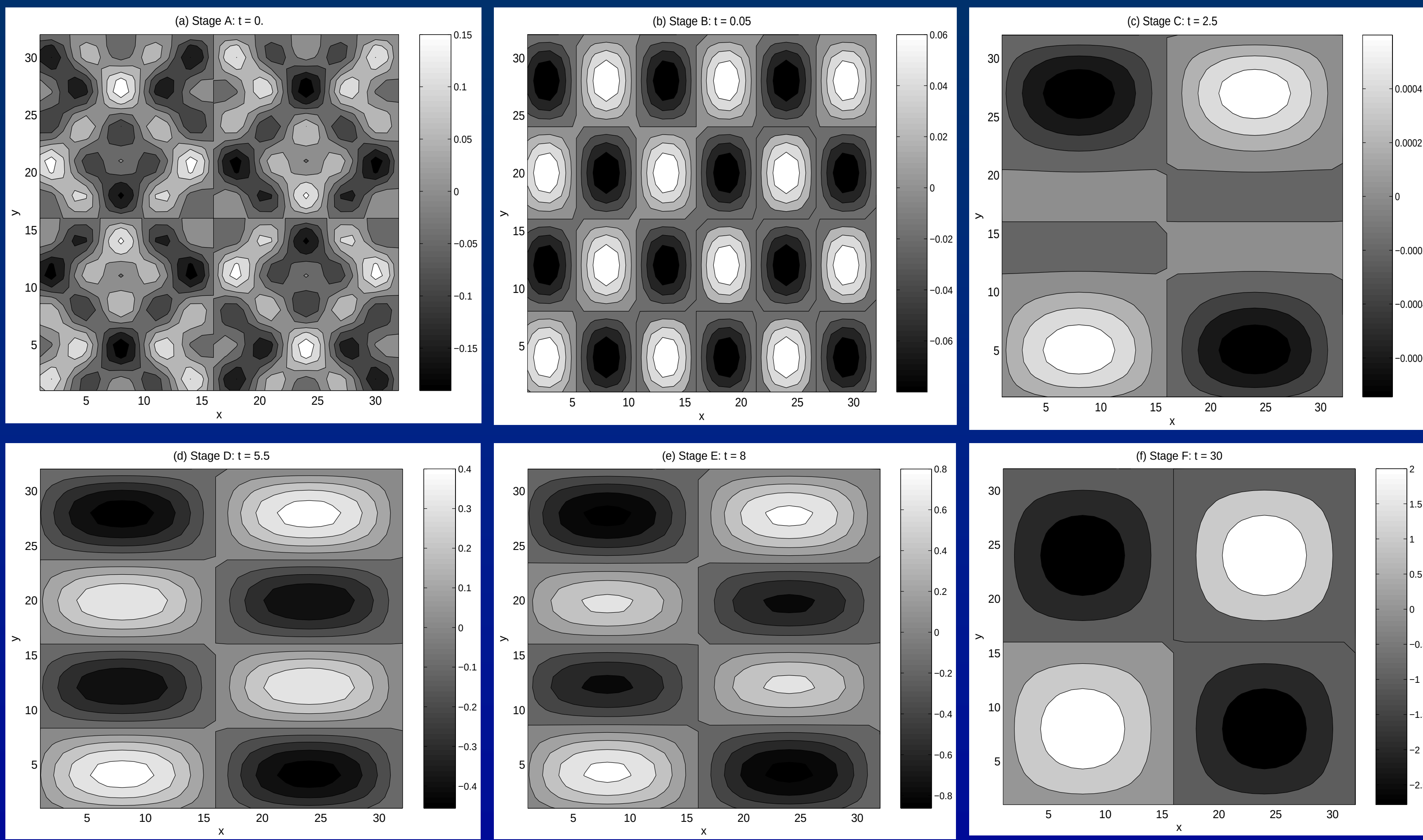


- Weakly nonlinear theory

  rough-smooth-rough pattern (Gyure *et al.* 98, Li-Liu 03)


time: t = 0
time: t = 0.5
time: t = 1
time: t = 15
time: t = 20
time: t = 30
time: t = 60
time: t = 240
height

Numerical solution: surface contours (Li-Liu 03)



3. Energy Minimization

``Energy-driven'' roughening and coarsening

$$\partial_t h = -\delta E(h)$$

- λ-minimizers: λ-periodical profiles of the least energy among all λ-periodical profiles.
- A simple scenario
  - There exist equilibrium λ_q-minimizers h_q with

$$E(h_N) > \cdots > E(h_n),$$
$$w(h_N) < \cdots < w(h_n),$$
$$\lambda_N < \cdots < \lambda_n.$$

  - The systems stays near such equilibriums, roughening and coarsening to reduce its (effective) energy.
  - The system saturates near a global minimizer.

Strategies

- Global minimizers: large-system asymptotics
- λ-minimizers
- Relation between the two models

Singularly perturbed energy

infinite ES: $E_\varepsilon(h) = \int_Q [\frac{1}{2}\varepsilon^{2m-2} |\partial^m h|^2 - \log |\nabla h|] dx$

finite ES: $E_\varepsilon(h) = \int_Q \frac{1}{2}[\varepsilon^{2m-2} |\partial^m h|^2 - \log(1+ |\nabla h|^2)] dx$

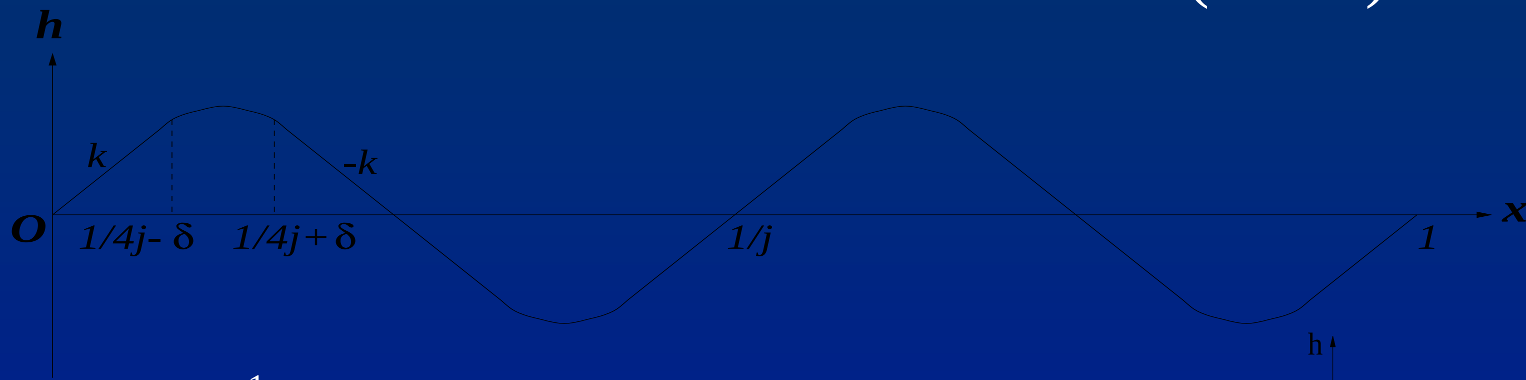
$x \to x/\varepsilon, \quad h \to h/\varepsilon,$

$\varepsilon = 1/L, \quad L =$ linear size of substrate,

$Q = (0,1) \times (0,1).$

Global minimization:
large-system asymptotics $(\varepsilon \to 0)$

- Heuristics: approximate global minimizer $(m=2)$

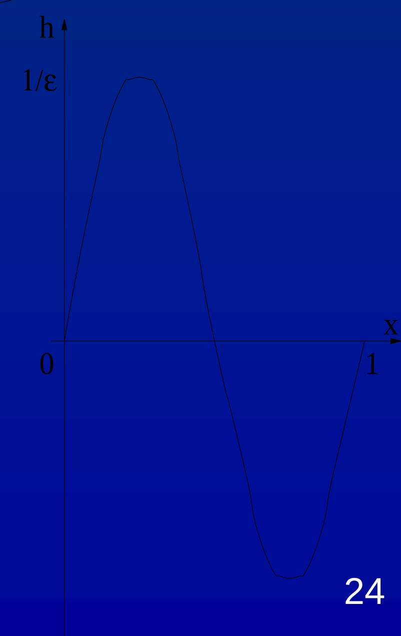


$$E_\varepsilon(h)=\int_0^1 \tfrac{1}{2}[\varepsilon^2\,|h''|^2-\log(1+|h'|^2)]dx = E_\varepsilon(\delta,k,j)$$

$$E_\varepsilon(k,j)=\min_\delta E_\varepsilon(\delta,k,j)$$

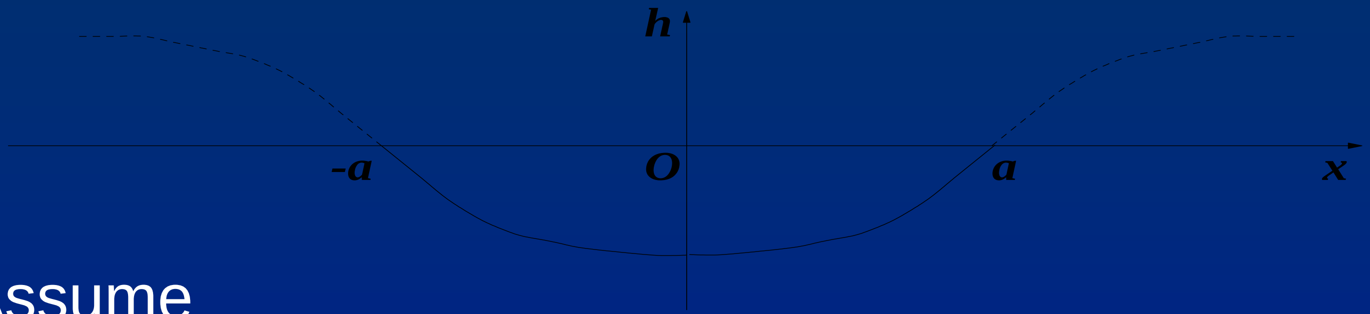
$$E_\varepsilon(j)=\min_k E_\varepsilon(k,j)$$

$k=O(1/\,j\varepsilon)$, $\delta=O(1/\,j)$, $E_\varepsilon(j)\sim\log(j\varepsilon)$.



- Heuristics: equilibrium profile $(m=2)$

$$\varepsilon^2 h^{(4)} + \left(\frac{h'}{1+h'^2}\right)' = 0$$



Assume

- h is even and convex in $[-a,a]$
- $h'(\pm a) = \pm k$ and $h(\pm a) = h''(\pm a) = 0$

Findings

- size of transition region $= O(k\varepsilon)$
- minimum energy $\sim -\log k$

Theorem

- Global energy-minimizers exist. Moreover,

$$C_1 + (m-1)\log\varepsilon \le \min_H E_\varepsilon \le C_2 + (m-1)\log\varepsilon.$$

- For any global minimizer $h \in H$,

$$C_3\varepsilon^{1-m} \le \| \nabla^k h \| \le C_4\varepsilon^{1-m} \quad (0 \le k \le m).$$

Notation

$H :=$ the closure of $\left\{ h \in C^\infty_{per}(Q) : \int_Q h\,dx = 0 \right\}$ in $H^m(Q)$

$$\| \nabla^k h \| = \sqrt{\sum_{|\alpha|=k} \int_Q | \nabla^\alpha h(x) |^2 \, dx}$$

Singularly perturbed energy: finite ES

$$E_\varepsilon(h) = \int_Q \tfrac{1}{2}[\varepsilon^{2m-2} |\partial^m h|^2 - \log(1+|\nabla h|^2)]dx$$

Proof

- Existence by direct methods.
- Upper bound of energy by construction.
- Lower bound of energy by Jensen's inequality

$$\min E_\varepsilon = E_\varepsilon(h) \geq -\tfrac{1}{2}\log\left(1+\|\nabla h\|^2\right)$$

and upper bound of gradients.

- Upper bound of gradients by

$$\delta E_\varepsilon(h)h = \int_Q \left(\varepsilon^{2m-2} |\partial^m h|^2 - \frac{|\nabla h|^2}{1+|\nabla h|^2} \right) dx = 0$$

  and the Poincare inequality.

- Lower bound of gradients by

$$\|\nabla h\|^2 = -\int_Q h \cdot \Delta h dx \leq \|h\| \cdot \|\Delta h\| \leq C_4 \varepsilon^{1-m} \|h\|,$$

$$C_2 + (m-1)\log \varepsilon \geq E_\varepsilon(h) \geq -\tfrac{1}{2}\log\left(1+\|\nabla h\|^2\right),$$

  and the Poincre inequality.

The Michigan model for a finite ES barrier

$$E(h) = \int [\tfrac{M_m}{2} |\partial^m h|^2 - \tfrac{FS}{2\alpha^2} \log(1 + \alpha^2 \sigma^2 |\nabla h|^2)] dx$$

Corollary

Let h be a global minimizer. Then, for $L >> 1$,

$$\min E \sim -\frac{FS}{\alpha^2} \log\left(\sqrt{\frac{FS\,\sigma^2}{M_m}} L^{m-1}\right),$$

$$\sqrt{\frac{1}{L^2}\int_{(0,L)^2} |\nabla^k h(x)|^2\, dx} \sim \frac{1}{\alpha}\sqrt{\frac{FS}{M_m}} L^{m-k}$$

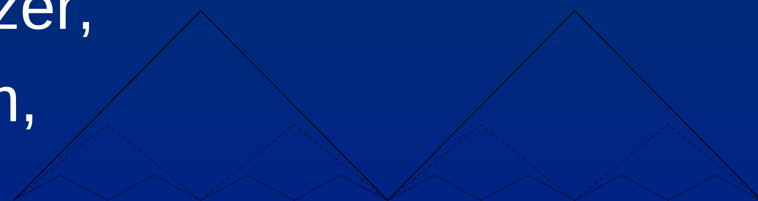
$$(0 \le k \le m).$$

λ-minimizers

Theorem

For any integer j, there exists a profile h_j such that

- h_j is an L/j-minimizer,
- h_j is an equilibrium,
- for $j << L$,



$$\min E \sim -\frac{FS}{\alpha^2}\log\left(\sqrt{\frac{FS\sigma^2}{M_m}}\left(\frac{L}{j}\right)^{m-1}\right),$$

$$\sqrt{\frac{1}{L^2}\int_{(0,L)^2}|\nabla^k h(x)|^2\,dx} \sim \frac{1}{\alpha}\sqrt{\frac{FS}{M_m}}\left(\frac{L}{j}\right)^{m-k}$$

$$(0 \le k \le m).$$

From finite to infinite ES barriers

Renormalized energies

$$\tilde{E}_\varepsilon(g) = E_\varepsilon\left(\tfrac{g}{\varepsilon}\right) - (m-1)\log\varepsilon$$

$$\tilde{E}_\varepsilon^{FES}(g) = \int \tfrac{1}{2}[\,|\partial^m g|^2 - \log(\varepsilon^{2(m-1)} + |\nabla g|^2)]dx$$

$$\tilde{E}^{IES}(g) = \int [\tfrac{1}{2}|\partial^m g|^2 - \log|\nabla g|]dx$$

Theorem $\quad \tilde{E}_\varepsilon^{FES} \xrightarrow{\Gamma} \tilde{E}^{IES} \quad (\varepsilon \to 0)$

large system $\Longrightarrow$ large slope

$\Longrightarrow$ $\dfrac{\nabla h}{1+|\nabla h|^2} \approx \dfrac{\nabla h}{|\nabla h|^2}$

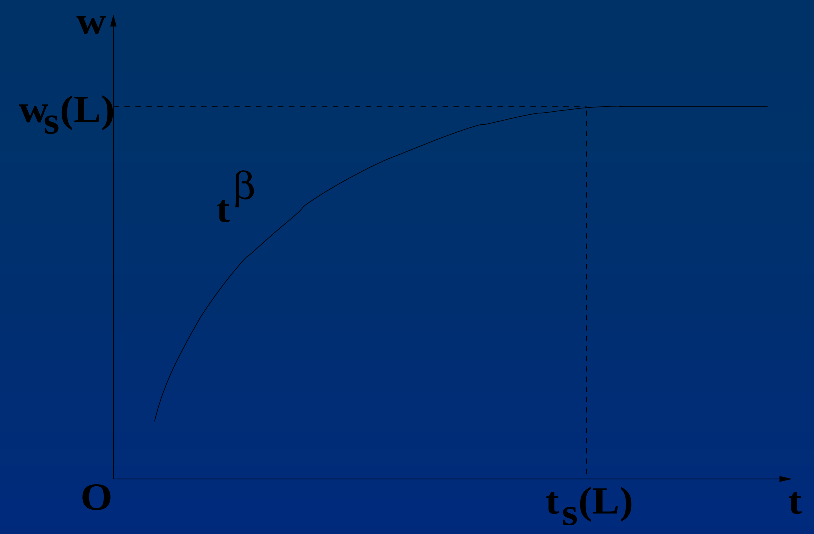
$\Longrightarrow$ enhancement of the ES effect

4. Scaling Laws

Scaling laws

$$\langle h(x,t)h(0,t)\rangle = [w(t)]^2 g\left(\frac{|x|}{\lambda(t)}\right)$$

$$w(t) = L^{\alpha} f(t/L^{z})$$



- Interface width

  $$w(t) \sim t^{\beta}$$

  β : growth exponent

- Mound lateral size

  $$\lambda(t) \sim t^{n}$$

  n: coarsening exponent

- Saturation width

  $$w_s(L) \sim L^{\alpha}$$

  α: roughness exponent

- Saturation time

  $$t_s(L) \sim L^{z}$$

  $z = \alpha/\beta$: dynamic exponent

Scaling prediction by energy minimization

$$w_s(L) \sim w(h_{\min}) \sim \frac{1}{\alpha}\sqrt{\frac{FS}{M_m}} L^m$$

$$\lambda(t_s) \sim t_s^n \text{ and } \lambda(t_s) \sim L \Longrightarrow t_s \sim L^{1/n}$$

$$\alpha = m, \qquad n = \beta / m, \qquad z = 1/n$$

- Agree with experiments ($m = 2, 3$) and previous theories: Ernst *et al.* 94, Johnson *et al.* 94, Stroscio *et al*. 95, Thuermer *et al.* 95, Rost & Krug 97, Golubovic 97.
- Precise prefector: competition of the two mechanisms.
- Previously known: $\beta = 1/2$ for all m. Not predicted by energy minimization.

Rigorous bounds for scaling laws

Finite ES: $$\partial_t h = (-1)^{m-1} M_m \Delta^m h - \nabla \cdot \left(\frac{FS\sigma^2 \nabla h}{1+\alpha^2\sigma^2|\nabla h|^2} \right)$$

Periodical boundary condition

Theorem

$$w(t) = \sqrt{\frac{1}{L^2}\int_{(0,L)^2} |h(x)|^2\, dx} \le \sqrt{\frac{2FS}{\alpha^2}}\, t^{1/2}$$

$$\sqrt{\frac{1}{(t-t_0)L^2} \iint_{[t_0,t]\times(0,L)^2} |\partial^k h(x,\tau)|^2\, dx d\tau} \le \frac{\sqrt{FS}}{\alpha M_m^{k/(2m)}}\, t^{(m-k)/(2m)} \quad (1 \le k \le m)$$

$$\frac{1}{t-t_0}\int_{t_0}^{t} E(h(\tau))\, d\tau \ge -\frac{FS}{\alpha^2}\log\left(\frac{FS\sigma^2}{M_m^{1/m}}\, t^{(m-1)/m} \right)$$

- Only one-sided bounds
- Bounds on coarsening rate not available

Finite ES: $\partial_t h = (-1)^{m-1}\Delta^m h - \nabla\cdot\left(\frac{\nabla h}{1+|\nabla h|^2}\right)$

Proof

- Energy method to bound the interface width and the highest-order gradients

$$\frac{1}{2}\frac{d}{dt}[w(t)]^2 + \int_{(0,L)^2} |\partial^m h|^2\, dx = \int_{(0,L)^2} \frac{|\nabla h|^2}{1+|\nabla h|^2} dx \le L^2$$

- Bound other gradients by the Cauchy-Schwarz inequality and

Lemma $A_k^2 \le A_{k-p}A_{k+p} \Longrightarrow A_k^n \le A_0^{n-k}A_n^k$

- Jensen's inequality for the lower bound of energy
- Scaling: put back parameters

5. Conclusions

Accomplishments

- A simple scenario of energy-driven roughening and coarsening: equilibrium λ-minimizers
- Energy minimization
  - large-system asymptotics: no-slope selection
  - λ-minimizers
  - Γ-convergence: enhanced ES effect
- Scaling prediction and bounds: precise prefectors

$$\beta = 1/2, \quad \alpha = m, \quad n = 1/(2m), \quad z = 2m$$

Questions

- Feedback to experiment: measurement of mobility and ES barrier?
- Limitations: gradient order parameters and periodical boundary conditions?
- Application to other models of energy-drive dynamics?

Current and future work

- Improve the theory
  - Stability of λ-minimizers
  - Upper bound for the coarsening rate
    - - approach of Golubovic?
    - - approach of Kohn-Otto and Kohn-Yan?
  - Optimality of bounds
- Scaling crossover

$$-\log\left(1+|\nabla h|^2\right)=\tfrac{1}{2}\left(|\nabla h|^2-1\right)^2-\tfrac{1}{2}+O\left(|\nabla h|^6\right)$$

- More effects: up-down asymmetry, downward funneling, elasticity, etc.

Thank you!