Modeling impacts of surface electromigration on stability and dynamical morphologies of a wetting, homoepitaxial solid film

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Key players: Surface electromigration and wetting/dewetting of the substrate by the film, I

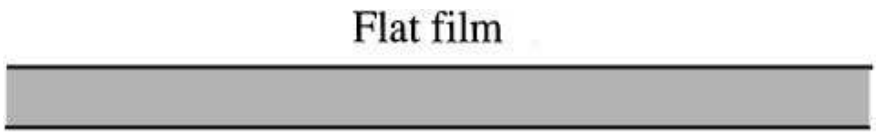
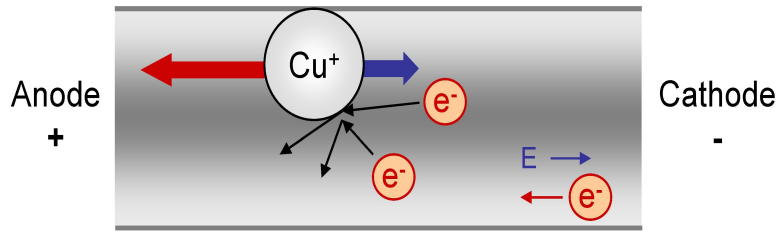


Figure : Homoepitaxial, single-crystal, metallic film grown on a substrate by MBE or CVD.

Surface electromigration: *Drift* of adatoms on **heated crystal surfaces** of metals in response to applied DC current, due to the *momentum transfer* from electrons to adatoms through scattering $\leftrightarrow$ "Electron wind"



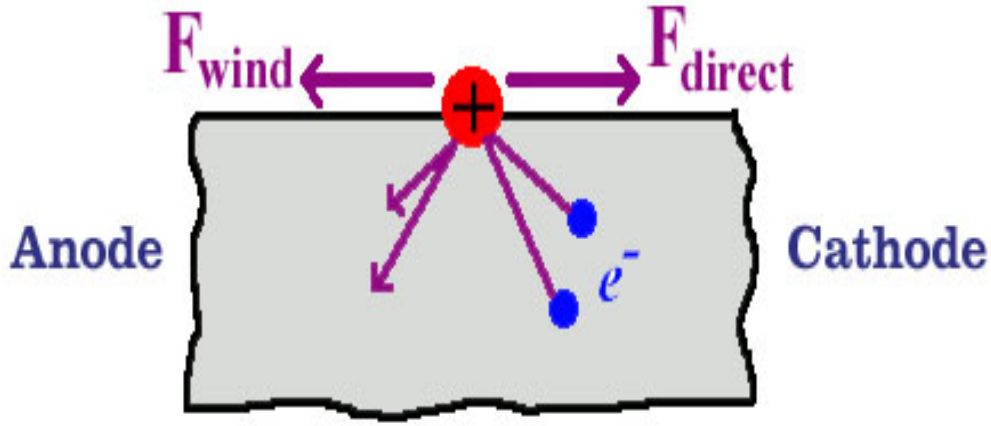
Adatom = adsorbed atom (mobile, is ionized via electrons scattering)
Homoepitaxial = absence of (or negligible) lattice mismatch stress at the film/substrate interface

Key players: Surface electromigration and wetting/dewetting of the substrate by the film, II

Driving forces for electromigration

- Direct Force: Direct action of the external field on the charge of the migrating ion
- Wind Force: Scattering of the conduction electrons by the metal adatom under consideration

$$F_{total} = F_{direct} + F_{wind} = qeE_0$$



Thin films are deposited on thick substrates acting as heat sinks, thus they can sustain huge current densities (up to several MA/cm^2) without melting

Key players: Surface electromigration and wetting/dewetting of the substrate by the film, III

Theory of surface electromigration on metals: application to self-electromigration on Cu(111)
P.J. Rous, T.L. Einstein, Ellen D. Williams

We present a calculation of the force, due to electron scattering, acting upon an adatom at the surface of a current-carrying metal, i.e., the so-called "wind" force in surface electromigration. The force is calculated from the Feynman-Hellmann theorem in which the electron scattering states are computed with full multiple scattering corrections by a layer-KKR method. ... At room temperature, the effective valence of the adatom is found to be ≈ -21. We conclude that surface electromigration of Cu on Cu(111) is dominated by the electron scattering force and that any direct force originating from charge transfer to the adatom is likely to be unimportant in this case. ...

Parametric formulation for motion of the surface, IV

The effective charge in surface electromigration

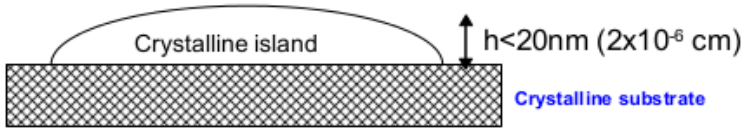
Elain S. Fu, D.-J. Liu, M.D. Johnson, J.D. Weeks, Ellen D. Williams

The rate of thermal decay of a metastable sawtooth morphology on Si(111) is greatly accelerated by the application of a bulk direct current in the "uphill" direction. STM measurements of the rate are compared with a mesoscopic theory of surface mass transport incorporating an effective surface electromigration force on the diffusing species. Quantitative agreement with the experimental observations is obtained for an effective charge ≤ 0.01 electron charge at 900°C.

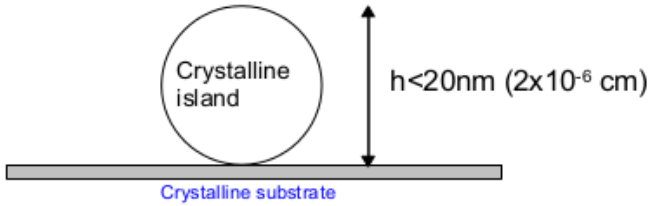
$\rightarrow \; q \sim 0.01$

Key players: Surface electromigration and wetting/dewetting of the substrate by the film, V

Wetting



``Wetting" island (solid 1 on substrate 1):
likes the substrate, tries to spread and maximize the contact area

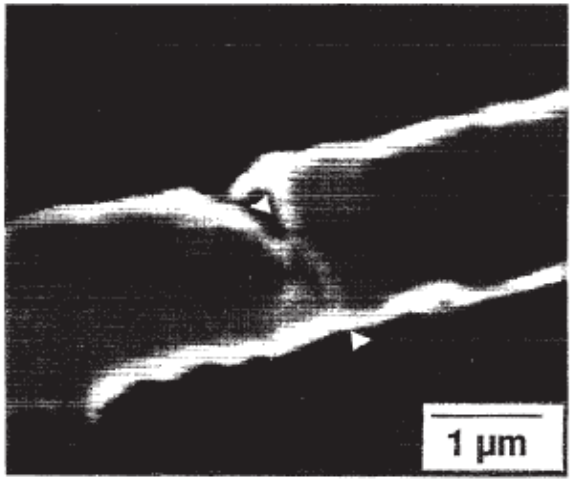


``Non-wetting" island (solid 1 on substrate 2):
hates the substrate, tries to contract and minimize the contact area

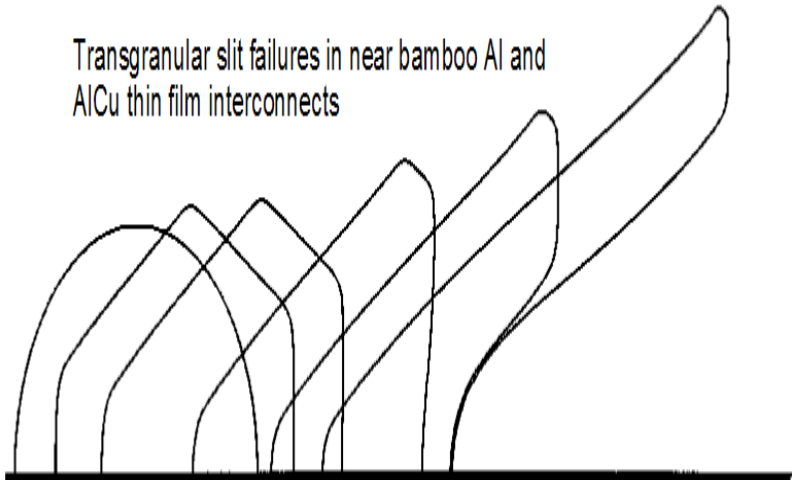
Continuous film (wetting): film surface energy is *smaller* than the substrate energy: $G = \gamma_S/\gamma_f > 1$

Particles (dewetting): film surface energy is *larger* than the substrate energy: $0 < G < 1$

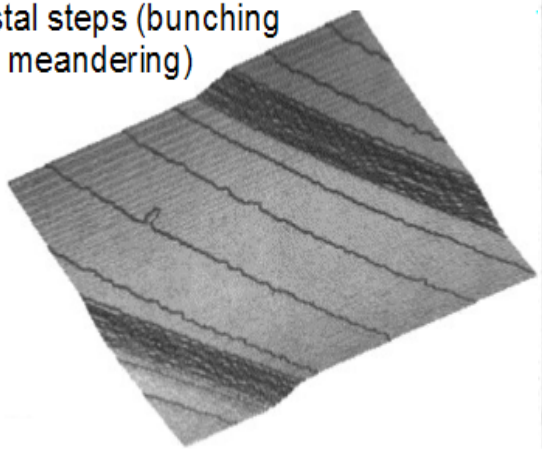
Surface electromigration in "thick" films: experiments

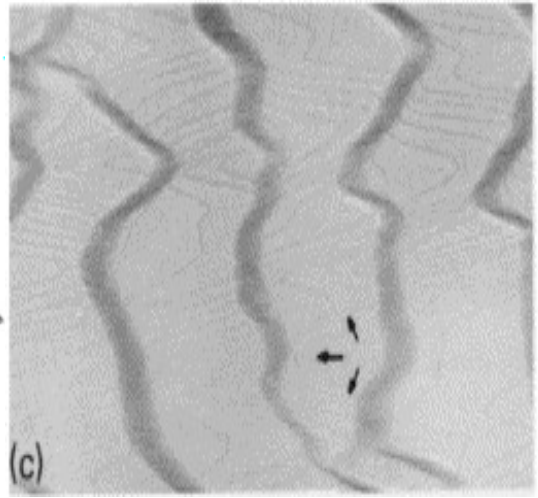


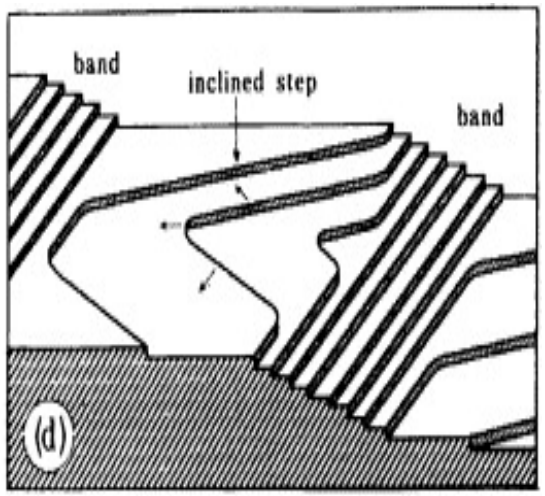
Transgranular slit failures in near bamboo Al and AlCu thin film interconnects



Instabilities of monoatomic crystal steps (bunching and meandering)

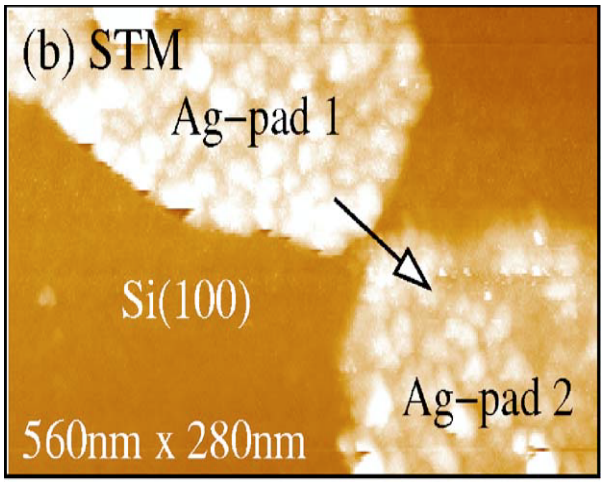






Surface electromigration in thin films: experiments

Another example: **Fabrication of sub-nanometer wide gaps (electrical contacts)**



A wetting ultrathin film: 10 ML (3 nm) thick Ag film on Si(001)
(from Appl. Phys. Lett. 89, 063120 (2006))

Open gap by applying electromigration current at 80 K

Close gap by enabling surface diffusion when heating to the room temperature

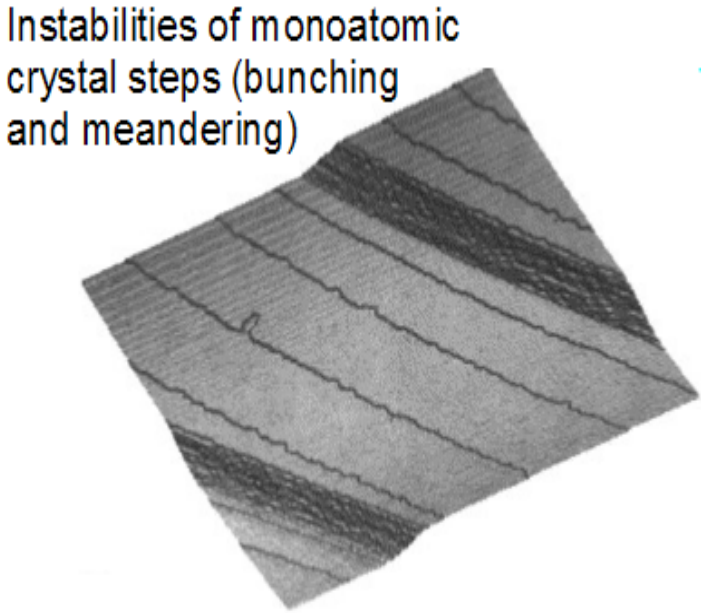
Broadly: What are the dynamical effects of substrate potential (wetting) and electromigration acting together on surface morphology of a *thin film - on length scales larger than interstep distance (continuum limit)* ?

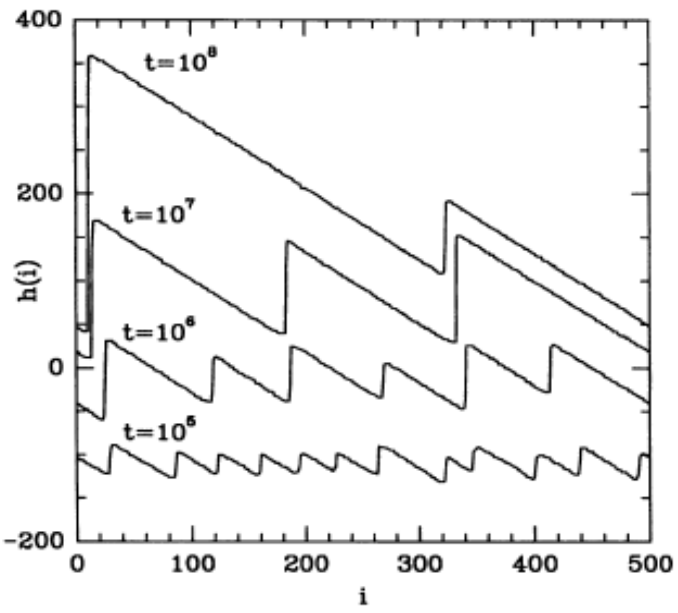
Current-driven surface faceting of THICK films

Continuum theory of surface dynamics on length scales much larger than the interstep distance: Krug & Dobbs'1994, Schimschak & Krug'1997

If the electric field is horizontal (along planar, unperturbed surface), then:

1. Facet orientations are first established locally,
2. Further evolution proceeds through a coarsening process





Facet size (hill-to-hill distance) $X \sim t^n$, where $n \approx 0.25$

Each facet = many monoatomic crystal steps

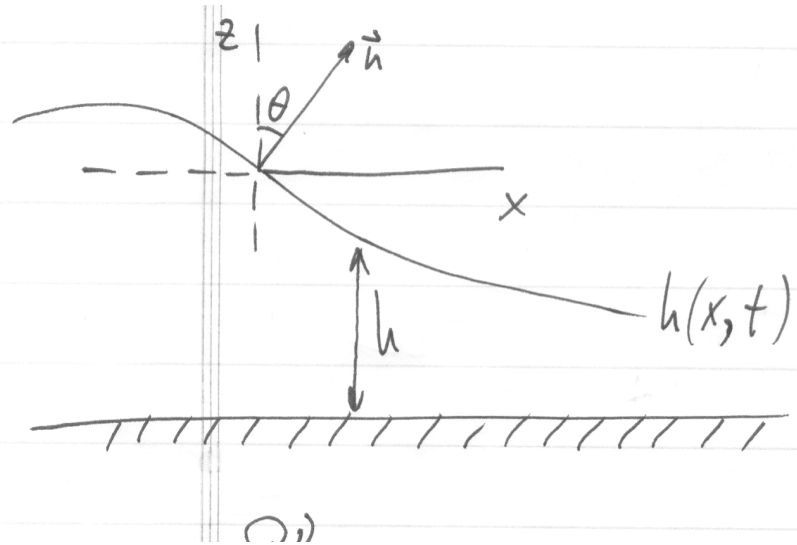
Thin films:

What are the effects on surface morphology of **both** wetting (the substrate potential) and electromigration - *on length scales much larger than interstep distance*, that is, in the continuum limit ?

Will there still be faceting and coarsening of facets ?

If yes, how will the rate of the coarsening process change ?

Idea for continuum modeling of 1D surface dynamics



$$h_t \cos\theta = -\Omega j_s \equiv V$$

Ω: atomic volume

s: arclength

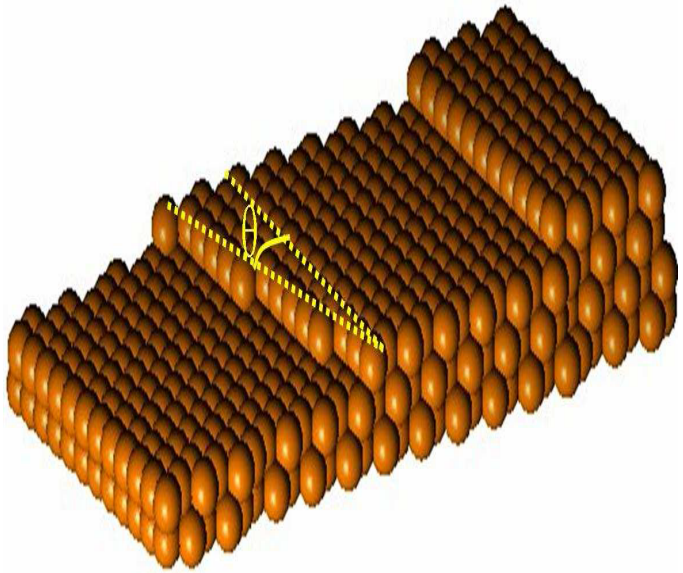
$$\frac{\partial}{\partial s} = (\cos\theta)\frac{\partial}{\partial x}\,;\quad \cos\theta = (1+h_x^2)^{-1/2}$$

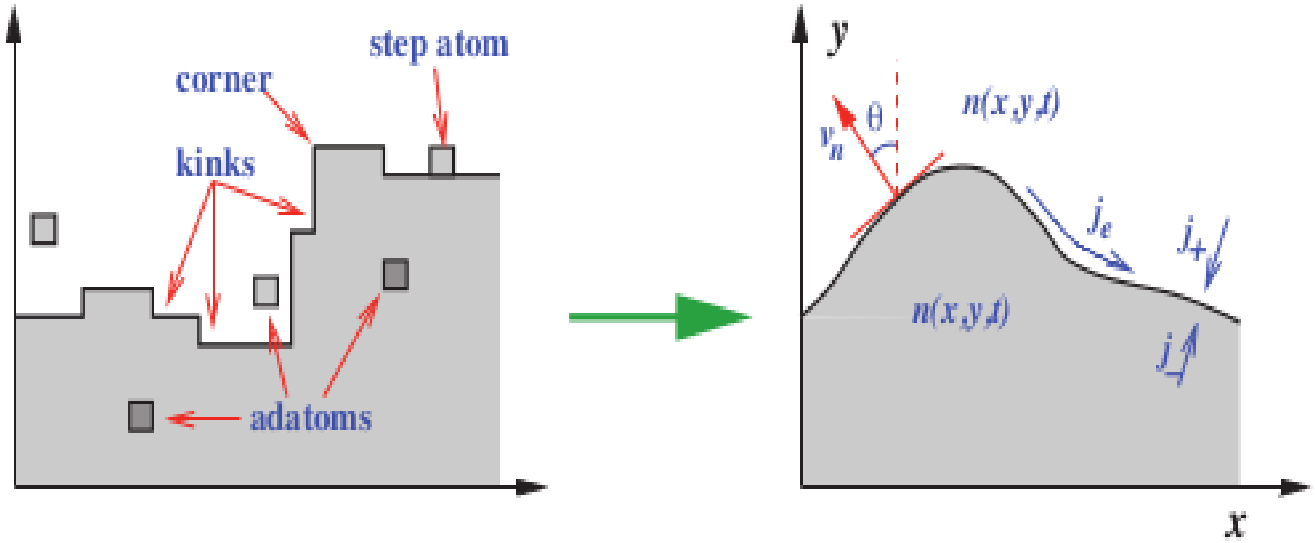
$$h_t = -\Omega j_x$$

j: adatoms flux on the film surface

Watching for non-graph (overhanging) surface shapes, transform the PDE into a parametric form (later)

Information: Continuum modeling of steps dynamics (NOT in this talk)





(From: J. Krug, arXiv:cond-mat/0405066)

Parametric formulation for motion of the surface, I

Let (x, z) be the components of the position vector of a point (a marker particle) on the evolving surface; then:

$$\begin{aligned} x_t &= V z_s = V \cos\theta = V z_u / g, \\ z_t &= -V x_s = -V x_u / g, \end{aligned}$$

$$g(u,t) = s_u = z_u / \cos\theta = \sqrt{x_u^2 + z_u^2} : \text{ the metric function}$$

$$V = \frac{D\Omega\nu}{kT} j_s = \left[M(\theta)\mu_s + (\alpha E_0 q) M(\theta) f(\theta) \right]_s ,$$

* u is the parameter along the surface, s is the arclength;
* $M(\theta)$ is the adatom mobility;
* $\mu(x, z, \theta, ...)$ is the surface chemical potential;
* E_0 is the strength of applied electric field;
* $f(\theta) = \cos\theta$ (when el. field is horizontal);
* q is the absolute value of **the effective charge** of (surface) diffusing ions;
* $\alpha = \pm 1$: electric field left/right.

First term in V: curvature driven surface diffusion; **second term in V:** surface diffusion enabled by electromigration (Mullins'63)

Parametric formulation for motion of the surface, II

The chemical potential: contributions from surface curvature (Mullins-like) and wetting interaction:

$$\mu = \Omega\left[\gamma\kappa + \gamma_{\mathbf{z}}\cos\theta\right], \quad \kappa = \theta_s = g^{-3}\left(z_{uu}x_u - x_{uu}z_u\right)$$

Use the two-layer interpolation model for the surface energy:

$$\gamma = \gamma(z) = \gamma_f + (\gamma_S - \gamma_f)\exp\left(-z/\ell\right),$$

where:
γ_S : the (constant !) energy of a bare substrate,
ℓ : the characteristic wetting length ($\sim 0.2nm$), and
γ_f : the (constant !) surface energy of a thick ($z \to \infty$) film.

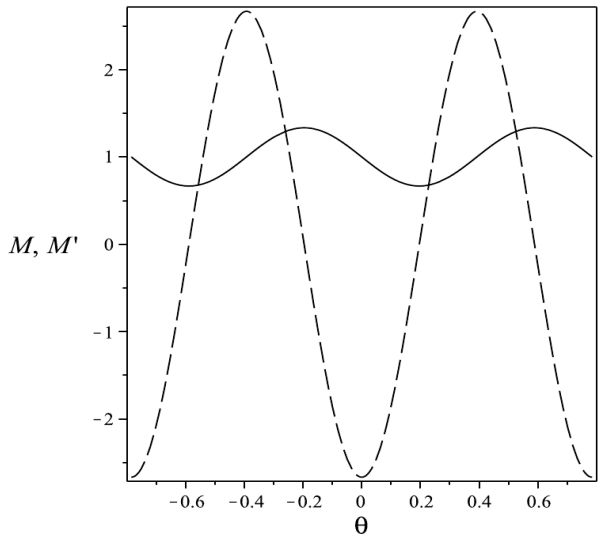
Constancy of γ_f is *the simplification*, since for crystal surfaces $\gamma_f = \gamma_f(\theta)$

Parametric formulation for motion of the surface, III

The anisotropic adatom mobility (Schimschak & Krug'1997):

$$M(\theta) = \frac{1 + \beta \cos^2 [N(\theta + \phi)]}{1 + \beta \cos^2 [N\phi]},$$

where:
N : the number of symmetry axes,
ϕ : the angle between a symmetry direction and the average surface orientation,
β : anisotropy strength.



$M(\theta)$ (solid line) and $M'(\theta)$ (dashed line) for $\beta = 1,\ N = 4$ and $\phi = \pi/16$

Dimensionless problem

ℓ : the length scale
ℓ^2/D : the time scale

$$\begin{aligned} x_t &= [BM(\theta)\mu_s + AM(\theta)f(\theta)]_s z_s \equiv V z_s, \\ z_t &= -V x_s, \\ \mu &= [1 + (G-1)\exp(-z)]\kappa + (1-G)\exp(-z) z_s. \end{aligned}$$

$f(\theta) = \cos\theta$: electric field is horizontal

$\kappa = \theta_s = g^{-3}(z_{uu}x_u - x_{uu}z_u)$

$B = \Omega^2\nu\gamma_f/(kT\ell^2)$, $A = \alpha\nu\Omega E_0 q/(kT) <> 0$, $G = \gamma_S/\gamma_f$
Wetting films : $G > 1$; Non-wetting films : $0 < G < 1$

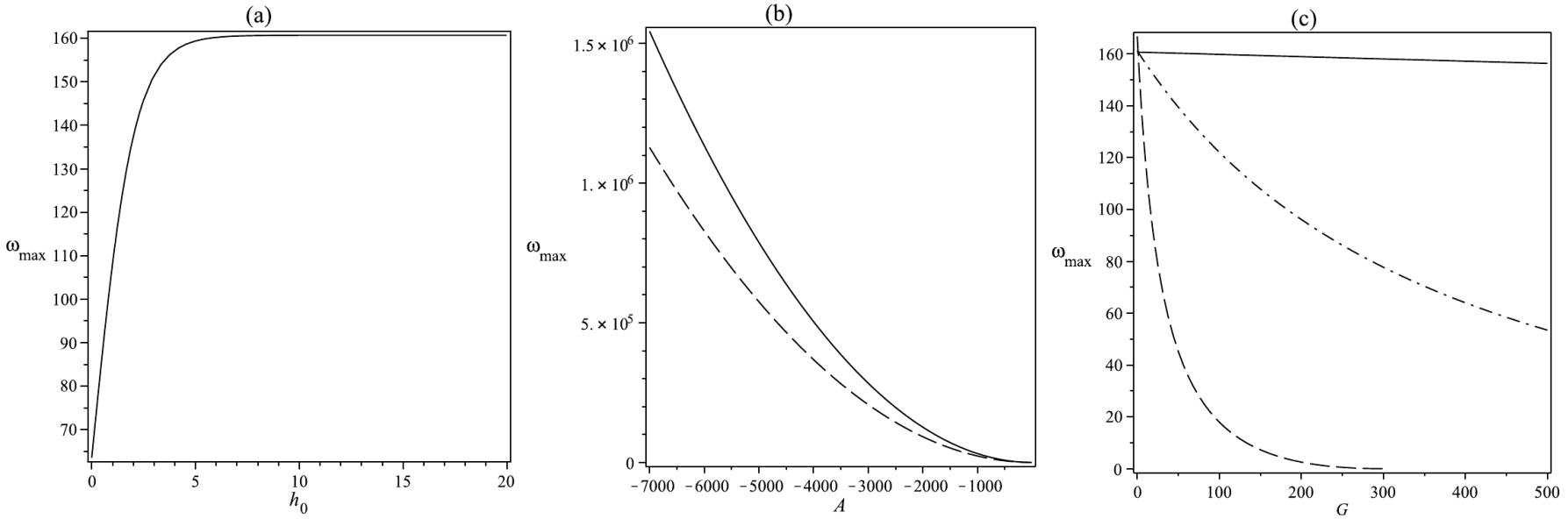
Typical values: $B = 8$, $71 \leq |A| \leq 7100$, $0.1 < G < 500$

Linear stability analysis for wetting films ($G > 1$)

$$\omega(k) = -B\left[1 + (G-1)\exp(-h_0)\right]k^4 - \left[B(G-1)\exp(-h_0) - AM'(0)\right]k^2$$

Long wave-unstable :

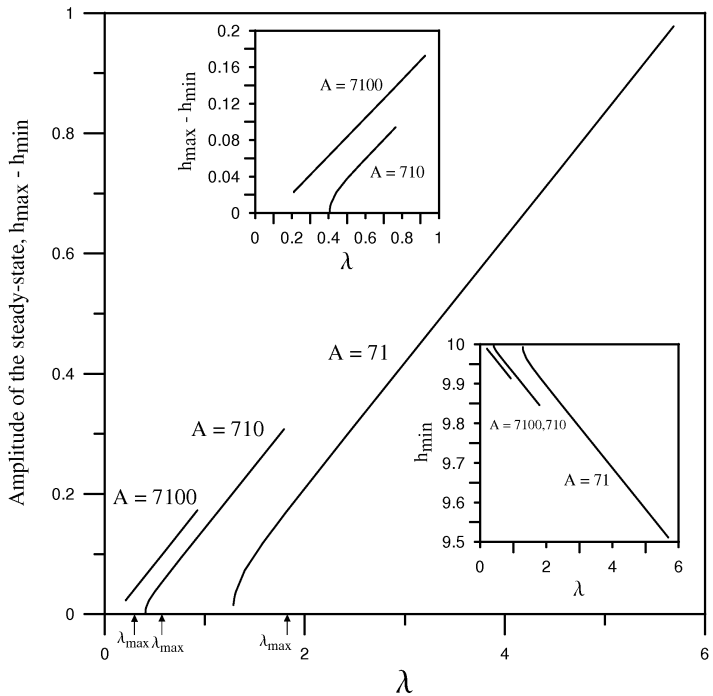
$AM'(0) > B(G-1)\exp(-h_0)$, or
$h_0 > h_{0c} = \ln\left(B|(1-G)/A|\right)$



(a) $A = -71$, $G = 2$;
(b) $h_0 = 10$ (solid line), $h_0 = 1$ (dashed line). $G = 2$;
(c) $h_0 = 10$ (solid line), $h_0 = 6$ (dash-dotted line), $h_0 = 3.5$ (dashed line). $A = -71$.

Nonlinear dynamics of unstable wetting films, I

Periodic steady-states from sinusoidal perturbations (λ: the wavelength)



$h_0 = 10, \;\; G = 2$

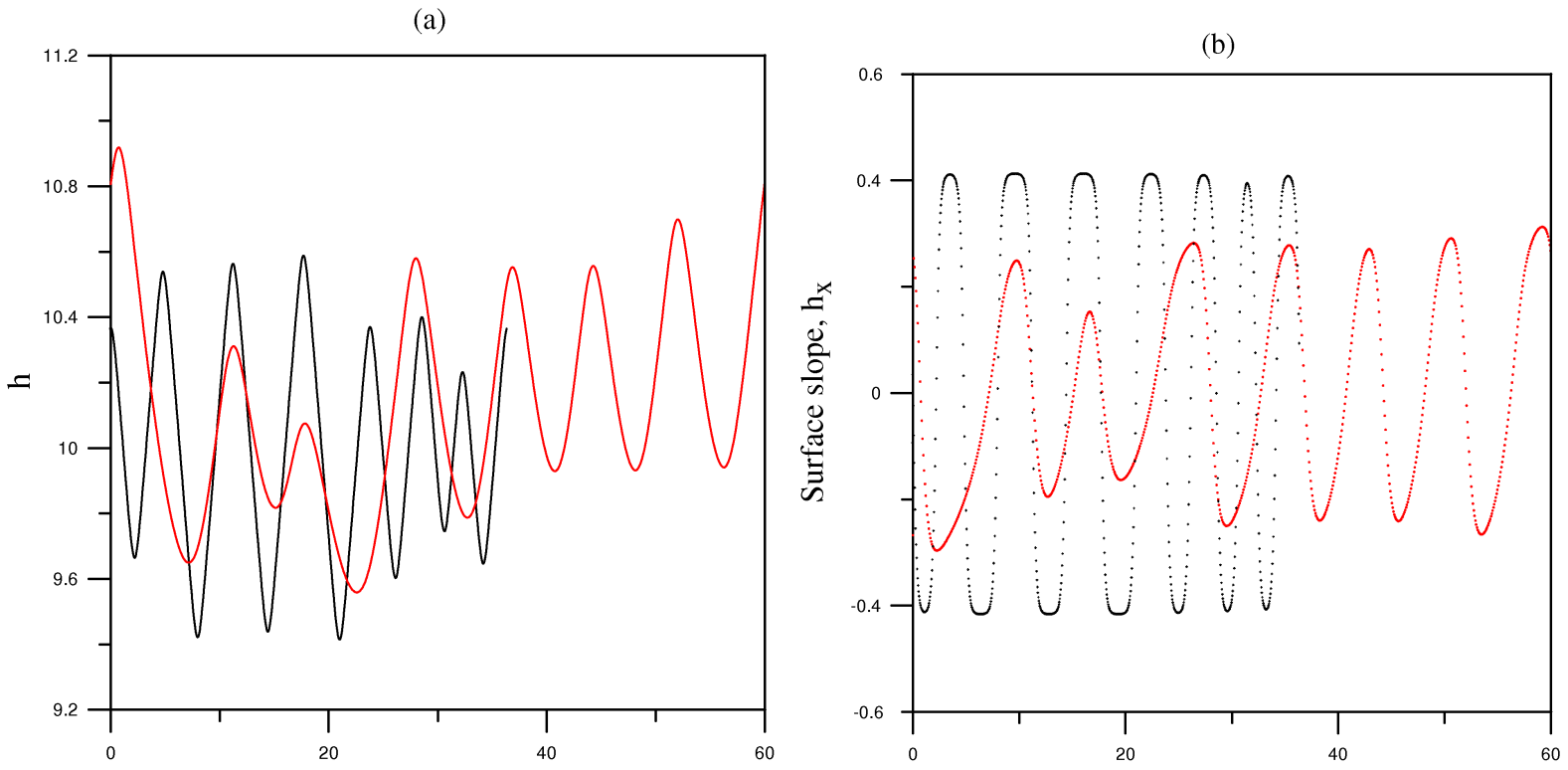
(i) *The* film does not dewet the substrate for all tested field strengths and all λ's (lower inset);

(ii) *Slope* of the curve (≈ 0.21) is insensitive to the field strength;

(iii) *Computed* steady-states are stable w/respect to random small- and large-amplitude perturbations (numerically tested).

Nonlinear dynamics of unstable wetting films, II

Snapshot of the surface: started from random perturbation (comp. domain : $0 \leq x \leq 20\lambda_{max}$, periodic b.c.'s))



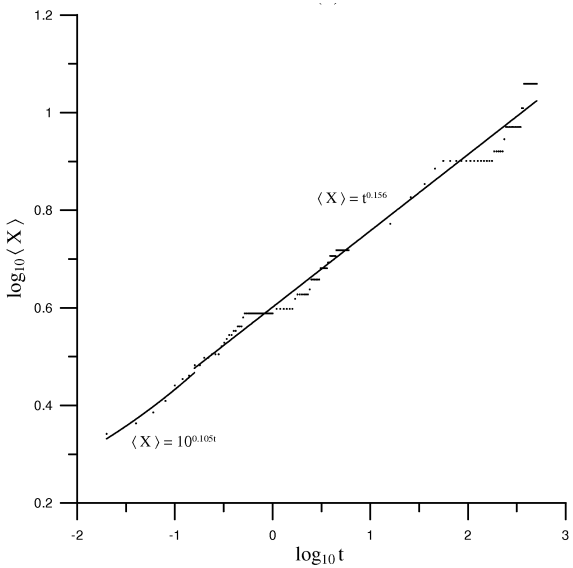
(a) Surface height. Black line: $A = 71,\ h_0 = 10,\ G = 2$, red line: $A = 71,\ h_0 = 6,\ G = 500$;

(b) Surface slope. Black crosses: $A = 71,\ h_0 = 10,\ G = 2$, red triangles: $A = 71,\ h_0 = 6,\ G = 500$.

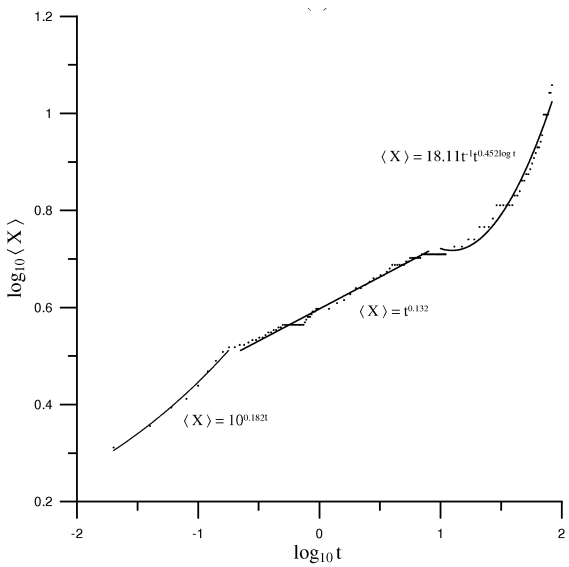
Nonlinear dynamics of unstable wetting films, III

Coarsening of random perturbation (comp. domain : $0 \leq x \leq 20\lambda_{max}$, periodic b.c.'s)

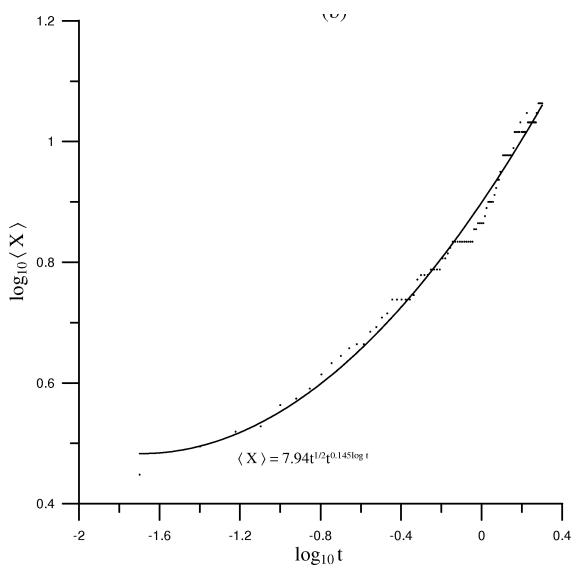
(i) *Hill* slope $= 24^\circ$ - constant during coarsening
(ii) *Stronger* field $\rightarrow$ faster coarsening
(iii) *Changes* from exponential law, to power law, to faster power law as wetting increases (h_0 decreases, G increases)



$A = 71,\ h_0 = 10,\ G = 2$



$A = 71,\ h_0 = 6,\ G = 2$



$A = 71,\ h_0 = 6,\ G = 500$

Summary[1]

- Wetting effect modifies strongly the stability properties of the film and the coarsening dynamics of electromigration-induced surface roughness
- Small-slope evolution equations are often inadequate for the description of strongly nonlinear phases of the dynamics
- Expect more complicated dynamics if:
  - *anisotropic* and *slope-dependent* surface energy, i.e. $\gamma = \gamma(h, \mathbf{h_x}, \theta)$: slope dependence has not been derived
  - non-local electric field (through the solution of the moving boundary value problem for the electric potential): traveling wave solutions (Schimschak & Krug'1997, Bradley'2002)

THANKS !

[1] To appear in the special issue "Nanoscale wetting of solids on solids" of the journal *Comptes Rendus Physique* (Olivier Pierre-Louis, Univ. Lyon, Editor)