The Effect of Elastic Stresses and Crystallographic Slip on Island Growth in Thin Films

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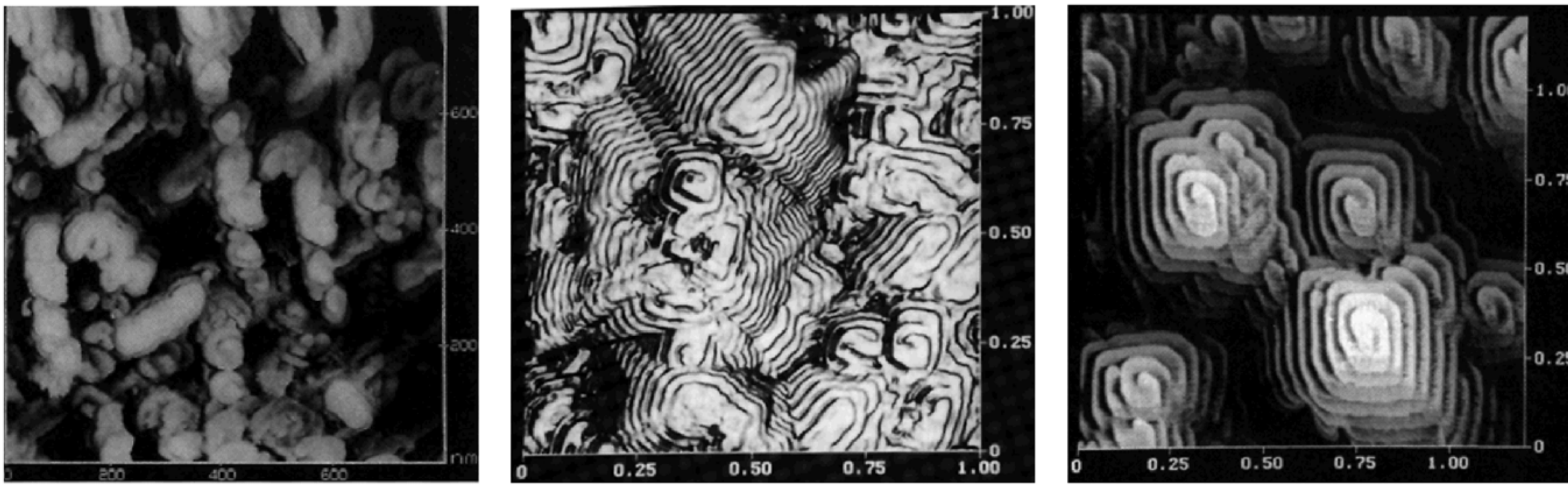
Third SIAM Conference
on
Mathematical Aspects of Materials Science
Philadelphia, PA, May 24, 2000

Sputtered YBCO Film on MgO substrate



(a) $h = 10nm$ (b) $h = 160nm$ (c) $h = 500nm$

I. D. Raistrick and M. Hawley, in: S. L. Shindé and D. A. Rudman (eds.) *Interfaces in High T_c Superconducting Systems*, Springer-Verlag, 1994.

Continuum film-growth model

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.

- Assumptions:
  - $h_{,t}$ admits Taylor expansion in $\nabla h, \nabla\nabla h, \dots$
  - Invariance under $\mathbf{x} \to -\mathbf{x}$.
  - Decay rate no faster than L^{-4}.
  - Onsager reciprocity relations.
- Rate equation for surface profile:

$$\boxed{h_{,t} = F(\nabla h) - V\,(h/\delta)\,\mathrm{e}^{-h/\delta} + C\,\nabla\cdot[(k^{-2}|\nabla h|^2 - 1)\nabla h] - D\nabla^4 h}$$

$F(\nabla h) \to$ deposition flux $\quad -V\,(h/\delta)\,\mathrm{e}^{-h/\delta} \to$ interfacial energy

$+C\,\nabla\cdot[(k^{-2}|\nabla h|^2 - 1)\nabla h] \to$ evaporation/condensation

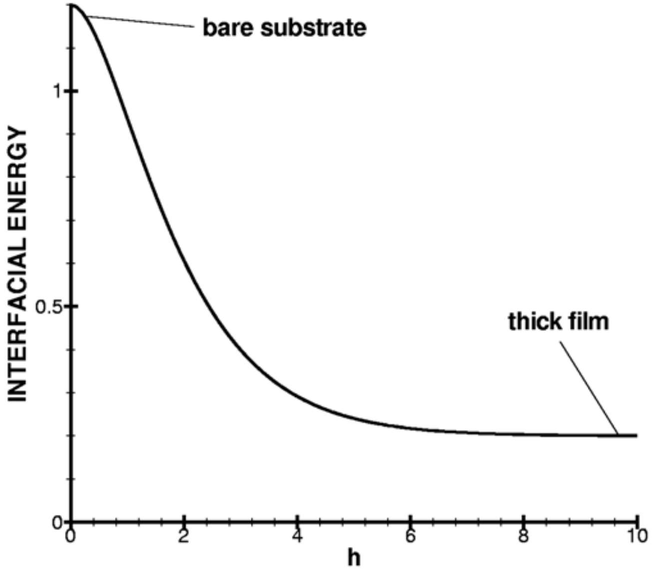
$-D\nabla^4 h \to$ capillarity $\quad k \to$ magic slope

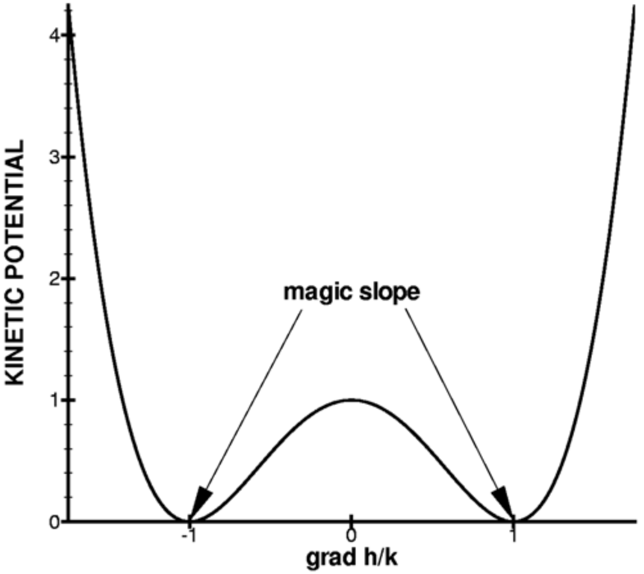
Continuum film-growth model

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.

- From Onsager reciprocity relations: $h_{,t} = -\delta\Phi[h]/\delta h$
- Kinetic potential:

$$\Phi[h] = \int_\Omega \{-Fh - V\,(h+\delta)\mathrm{e}^{-h/\delta} + \frac{C}{4k^2}(|\nabla h|^2 - k^2)^2 + \frac{D}{2}(\nabla^2 h)^2\}\, d^2x$$





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Stability of flat films - island nucleation

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.

- Linearization: $h_0 \to h_0 + u$,

$$u_{,t} = -Bu - C\nabla^2 u - D\nabla^4 u$$

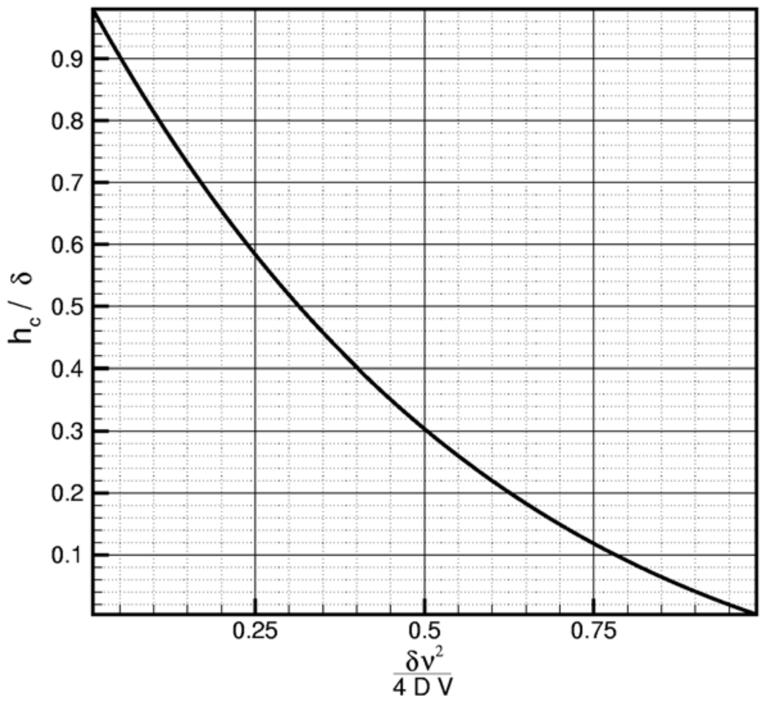
- Set: $u = A\mathrm{e}^{-\lambda t}\mathrm{e}^{-i\boldsymbol{k}\cdot\boldsymbol{x}}$,

$$\lambda = Dk^4 - Ck^2 + C$$

- Critical wavelength:

$$k_c = \sqrt{C/2D}$$

- $h_c = 0 \Rightarrow$ Volmer-Weber.
- $h_c > 0 \Rightarrow$ Stranski-Krastanow.



Critical thickness h_c.

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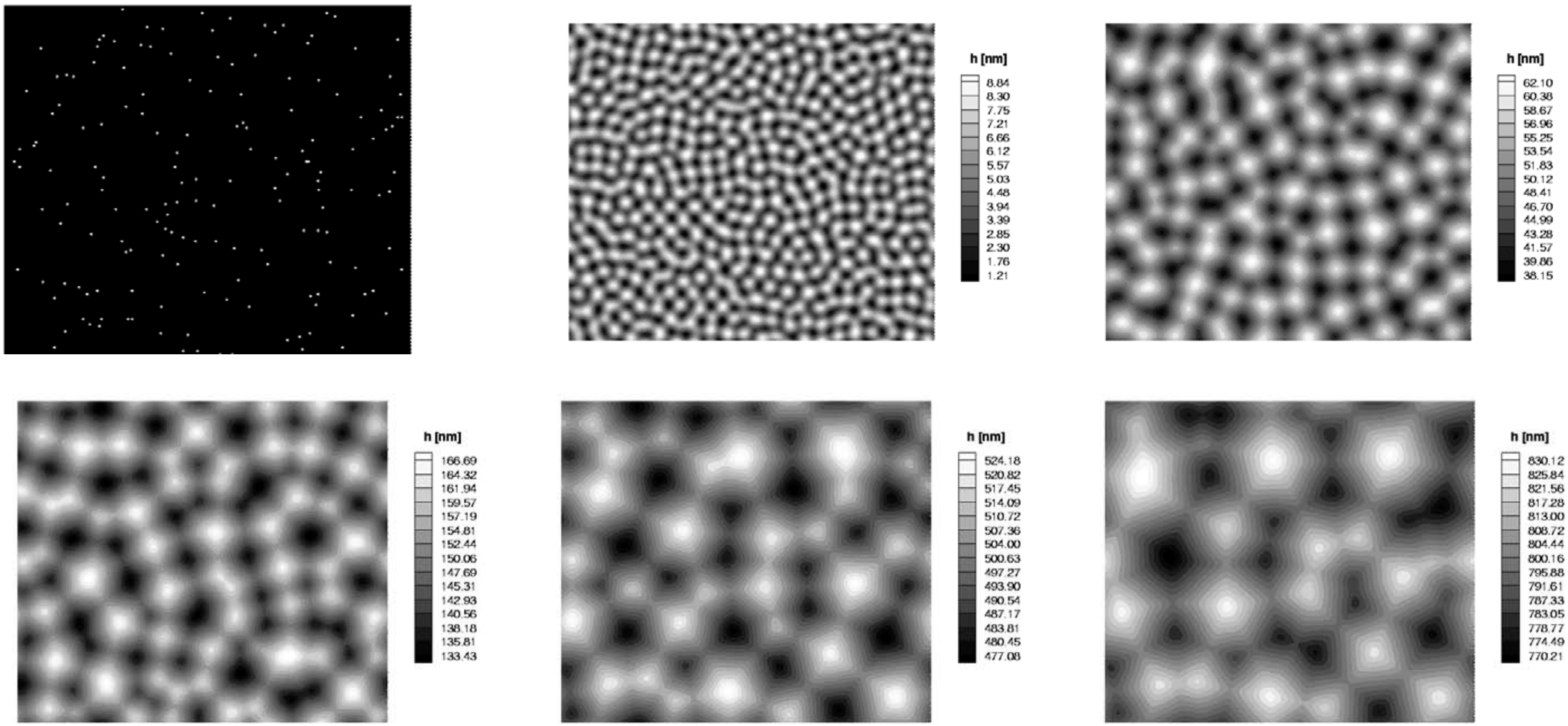
Surface-profile evolution

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.



Level contours of surface height (nm).

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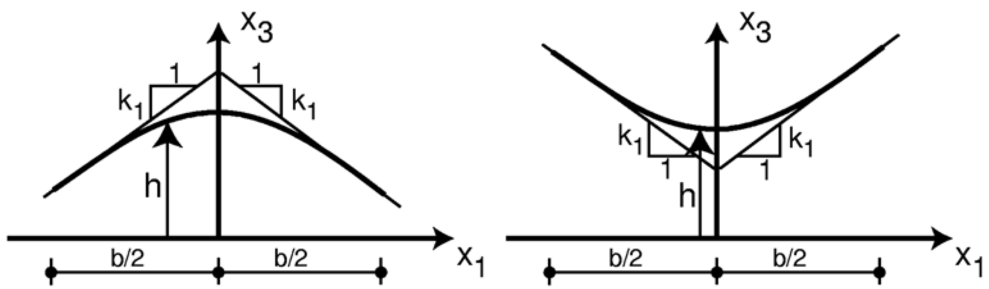
Systems with small capillarity

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.



- $D \to 0$, sharp-interface approximation.
- Boundary layer width: $l = \sqrt{D/2C} \to 0$.
- Relaxation time: $\tau = D/2C^2 \to 0$.
- Limiting kinetic potential: $\Phi_0[h] = \int_{\mathcal{S}} \sqrt{2CD}\,[2k_1^3(s)/3k]\,ds$,

  $\mathcal{S} \equiv$ singular set of the profile.
- Limiting kinetics: $h_{,t} = -\delta\Phi_0[h]/\delta h$.

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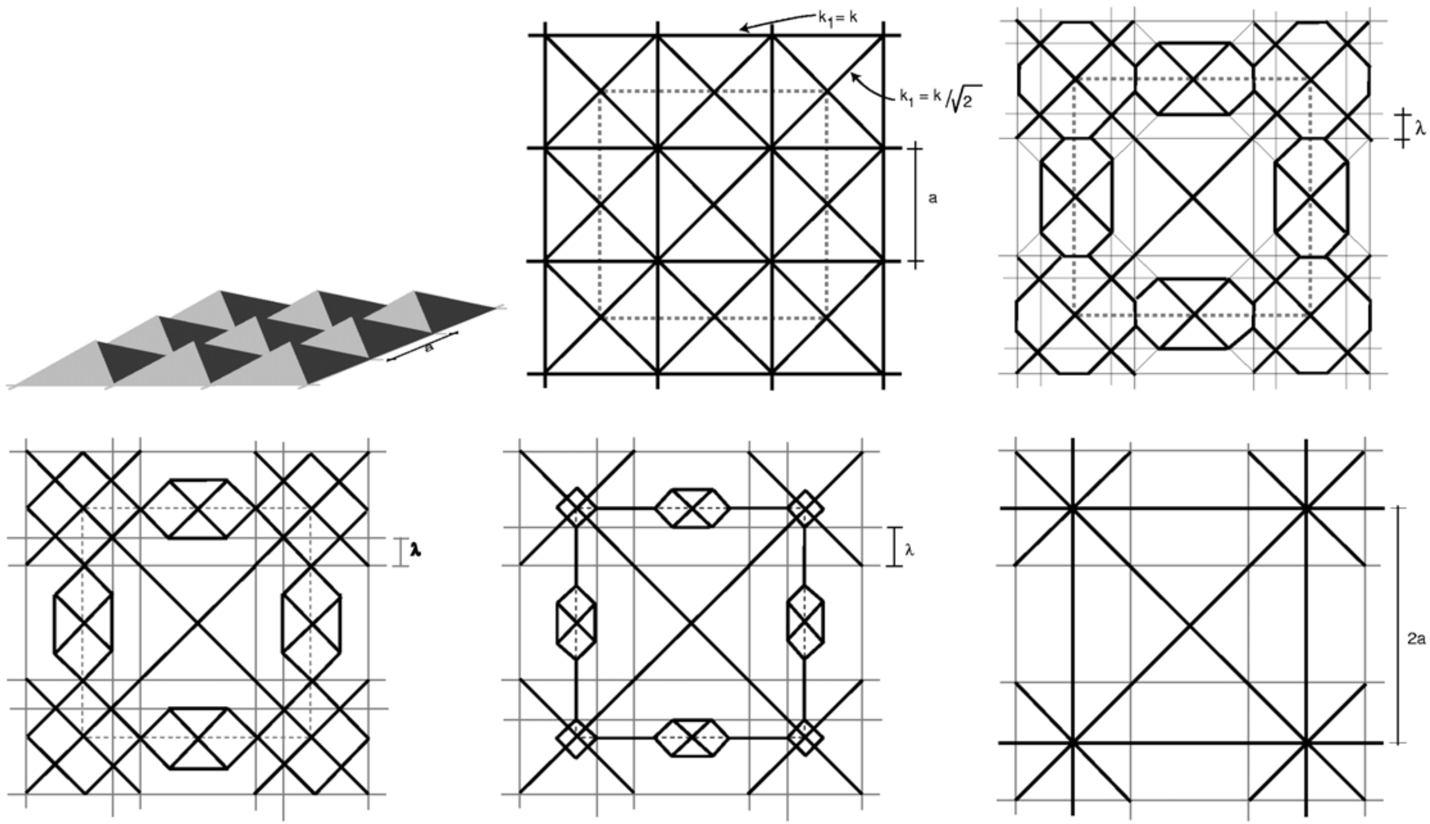
Coarsening construction

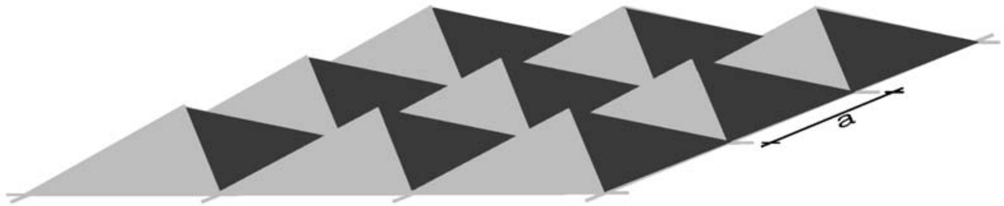
M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.



Evolution of singular set (ridges and grooves).

Coarsening construction

M. Ortiz, E. A. Repetto and H. Si, *J. Mech. Phys. Solids*, **47** (1999) 697.



- Effective kinetic equation:

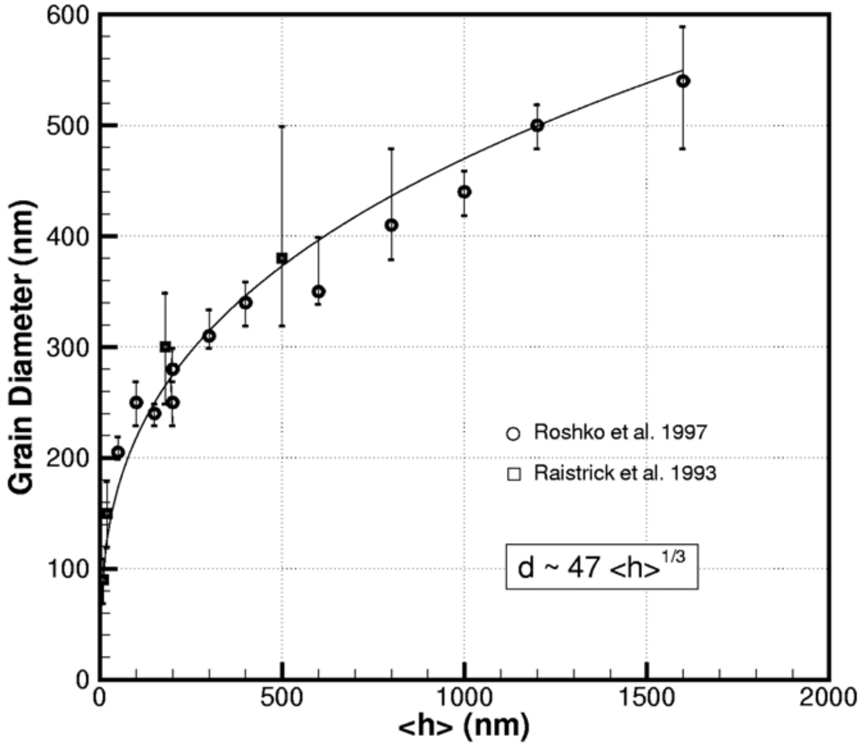
$$h_{1,t} = \frac{80}{3}\frac{\sqrt{2CD}\,k^4}{12h_1^2 - 4ah_1k + 3a^2k^2}, \qquad h_1 < ak$$

$$h_{1,t} = \frac{16}{3}\frac{\sqrt{2CD}\,k^4}{-20h_1^2 + 60ah_1k - 29a^2k^2}, \quad h_1 > ak$$

- Grain-size growth law:

$$d = \left(\frac{280}{59}\frac{\sqrt{2CD}\,k}{F}\langle h\rangle\right)^{1/3} \sim \langle h\rangle^{1/3}$$

Grain-size evolution



Grain diameter d *vs.* average film thickness $\langle h \rangle$. Theoretical fit to data of Raistrick and Hawley (1993) and Roshko *et al.* (1997).

Strained-film growth

- Rate equation for surface profile:

$$\boxed{h_{,t} = F(\nabla h) + C\,\nabla\cdot[(k^{-2}|\nabla h|^2 - 1)\nabla h] - D\nabla^2(\nabla^2 h - \gamma^{-1} U)}$$

where $U(x_1, x_2)$ is the volume free-energy density at $x_3 = h(x_1, x_2)$.

- First-order perturbation formula:

$$\delta E = \int_\Omega U\,\delta h\,d\Omega \quad \Rightarrow \quad U = \frac{\delta E}{\delta h}$$

- Goals of the analysis:
  - Derive asymptotic formulae for free energy E as a functional of h in the limit of small film thickness and shallow waviness.
  - Analyze the effect of elastic energy on island size and morphology.

Strained-film growth

- Reference configuration: Film of uniform thickness $\langle h \rangle$, misfit strains ϵ^*_{ij}, misfit stresses σ^*_{ij}, traction-free surface: $\sigma^*_{3i} = 0$.
- Perturb state of film by:
  - Roughening film profile: $\langle h \rangle \to h(x_1, x_2)$.
  - Crystallographic slip $\Rightarrow$ plastic strains ϵ^p_{ij}.
- Free energy:

$$
\begin{aligned}
E &= \int_\Omega \int_0^{h(x_1,x_2)} \frac{1}{2} c_{ijkl} (u_{i,j} - \epsilon^*_{ij} - \epsilon^p_{ij})(u_{k,l} - \epsilon^*_{kl} - \epsilon^p_{kl}) \, d\Omega \, dx_3 \\
&+ \int_\Omega \int_0^{h(x_1,x_2)} W^p \, d\Omega \, dx_3 \; + \; \int_\Omega \int_{-\infty}^0 \frac{1}{2} c_{ijkl} u_{i,j} u_{k,l} \, d\Omega \, dx_3
\end{aligned}
$$

$W^p \equiv$ stored energy of cold work.

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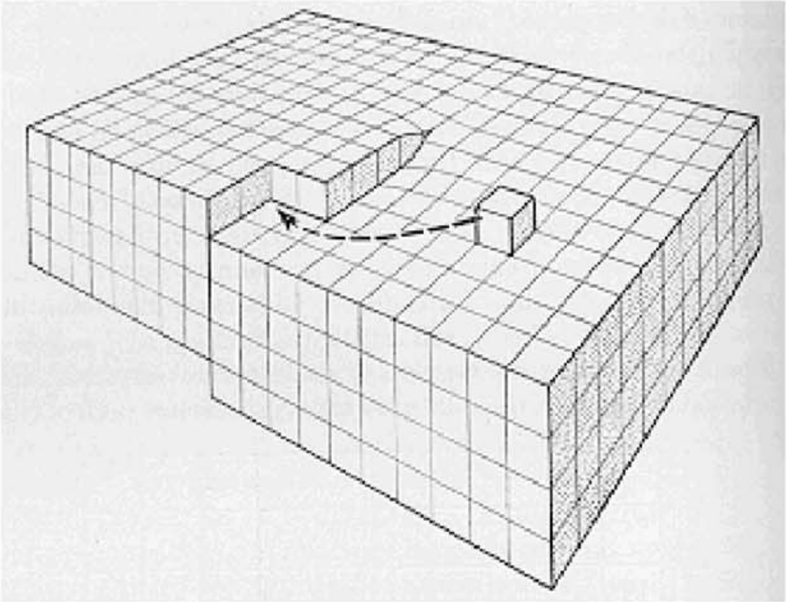
Crystallographic slip - Continuum theory

- Plastic strains: $\epsilon^p_{\alpha 3} = \gamma^p_\alpha / 2$
- Density of screw dislocations:
  $\rho = (\gamma^p_{2,1} - \gamma^p_{1,2})/b$
- Density of interfacial dislocations:
  $\alpha_{31} = -\beta^p_{32}, \quad \alpha_{32} = \beta^p_{31} \quad \Rightarrow$
  prismatic loops.
- c-axis wobble:
  $\theta^p = (1/2)\sqrt{(\gamma^p_1)^2 + (\gamma^p_2)^2}$
- Yield condition: $|\sigma_{\alpha 3}| = \tau_c$.
- Ideal plasticity: $\tau_c =$ constant.



Slip planes (a and b) and Burgers vector (c) in YBCO.

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Asymptotic analysis

- Assume:
  - Shallow waviness: $|\nabla h| < \varepsilon \ll 1$.
  - Small thickness: $\langle h \rangle / d < \varepsilon \ll 1$, $d \equiv$ in-plane dimension.
- Asymptotic form of the elastic energy as $\varepsilon \to 0$:

$$\triangle E \sim -\int_\Omega \int_\Omega \frac{1}{2} G_{ik}(\boldsymbol{x} - \boldsymbol{x}') t_i(\boldsymbol{x}) t_k(\boldsymbol{x}') d\Omega d\Omega'$$

where $G \equiv$ surface Green's function for substrate.

- Effective tractions: $t_i \sim -\sigma^*_{i\beta} h,_\beta - (h\, c_{i\beta kl} \epsilon^p_{kl}),_\beta$.
  1. $-\sigma^*_{i\beta} h,_\beta \Rightarrow$ effect of waviness (Gao, 1991).
  2. $(h\, c_{i\beta kl} \epsilon^p_{kl}),_\beta \Rightarrow$ effect of crystallographic slip.

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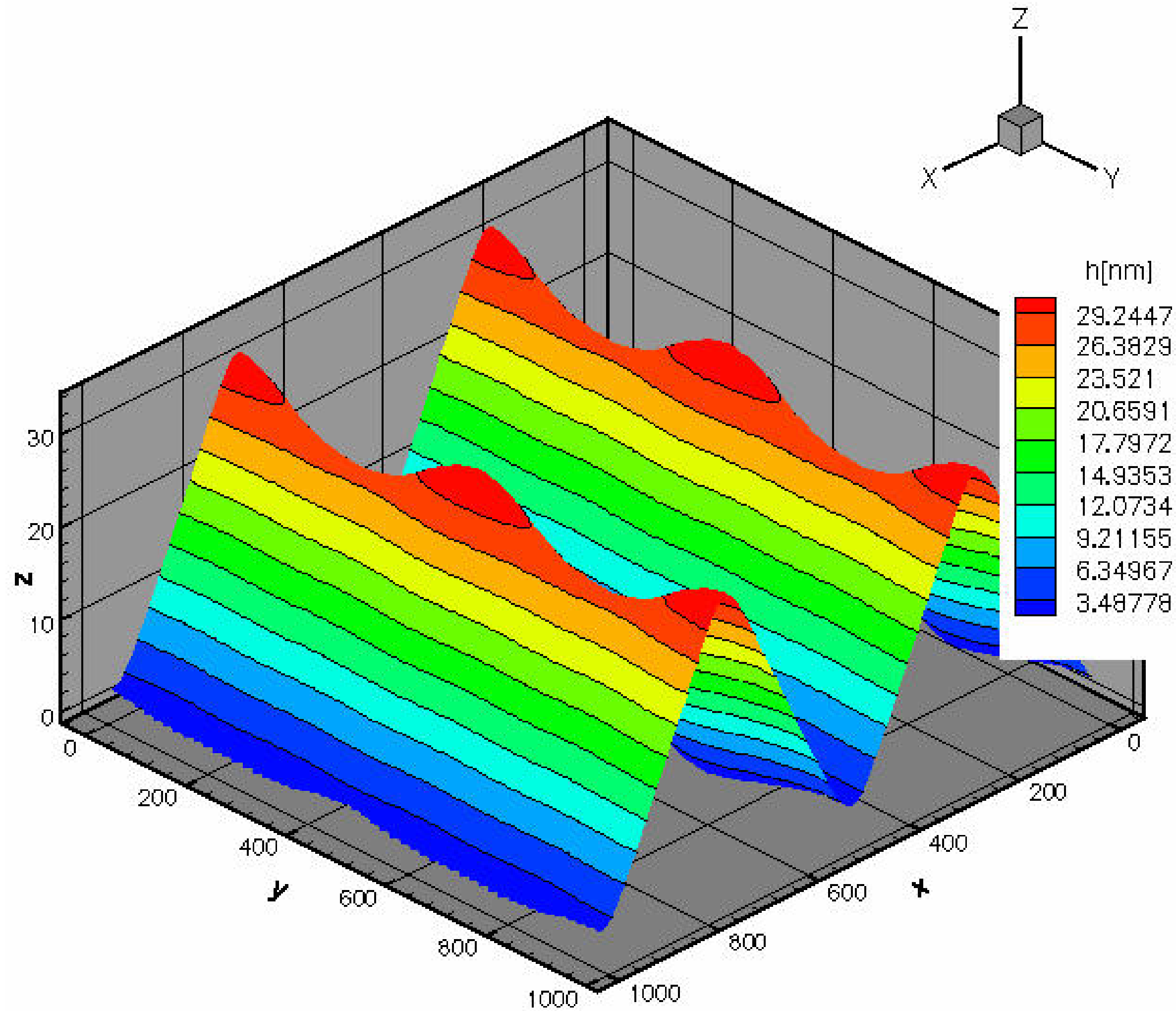
Elastic film - Biaxial stress



Surface profile snapshots,
t = 0, 200, 300, 400, 500, 600 s

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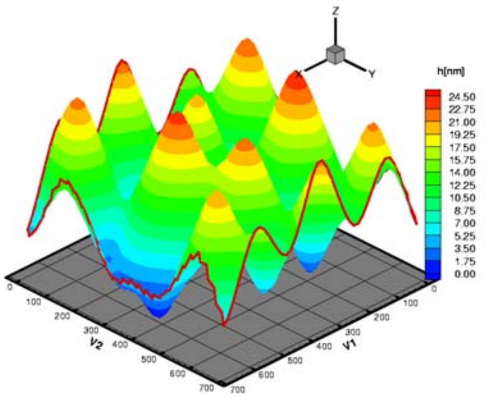
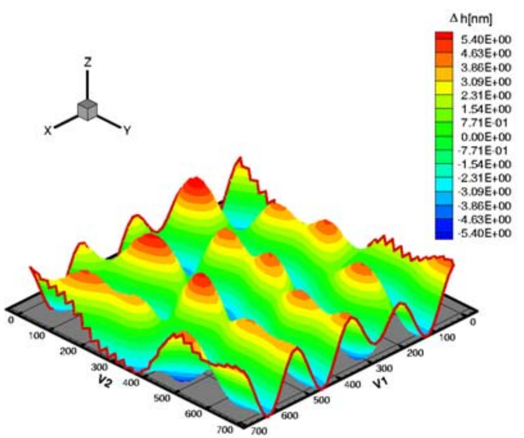
Elastic film - Linear stability analysis

- Elastic film, $h = \langle h \rangle + u$, linearize:
  $u_{,t} = -C\nabla^2 u - D\nabla^2(\nabla^2 u - \gamma^{-1} U)$
- Set: $u = A\mathrm{e}^{\lambda t}\mathrm{e}^{i\boldsymbol{k}\cdot\boldsymbol{x}}$
- Biaxial stress: $k_1 = k_2 = k$,
  $\lambda = -2Ck^2 + 4Dk^4 - 4\sqrt{2}Dk^3\gamma^{-1}{\sigma^*}^2/E'_s$
- Steady profile: $\lambda = 0$,
  $k \sim \sqrt{2}{\sigma^*}^2/\gamma E'_s$, for σ^* large.
- Uniaxial stress: $\sigma^*_{11} = \sigma^*$, $k_1 = k$, $k_2 = 0$,
  $\lambda = -Ck^2 + Dk^4 - 2Dk^3\gamma^{-1}{\sigma^*}^2/E'_s$
- Steady profile: $\lambda = 0$,
  $k \sim 2{\sigma^*}^2/\gamma E'_s$, for σ^* large.





Biaxial and uniaxial stress

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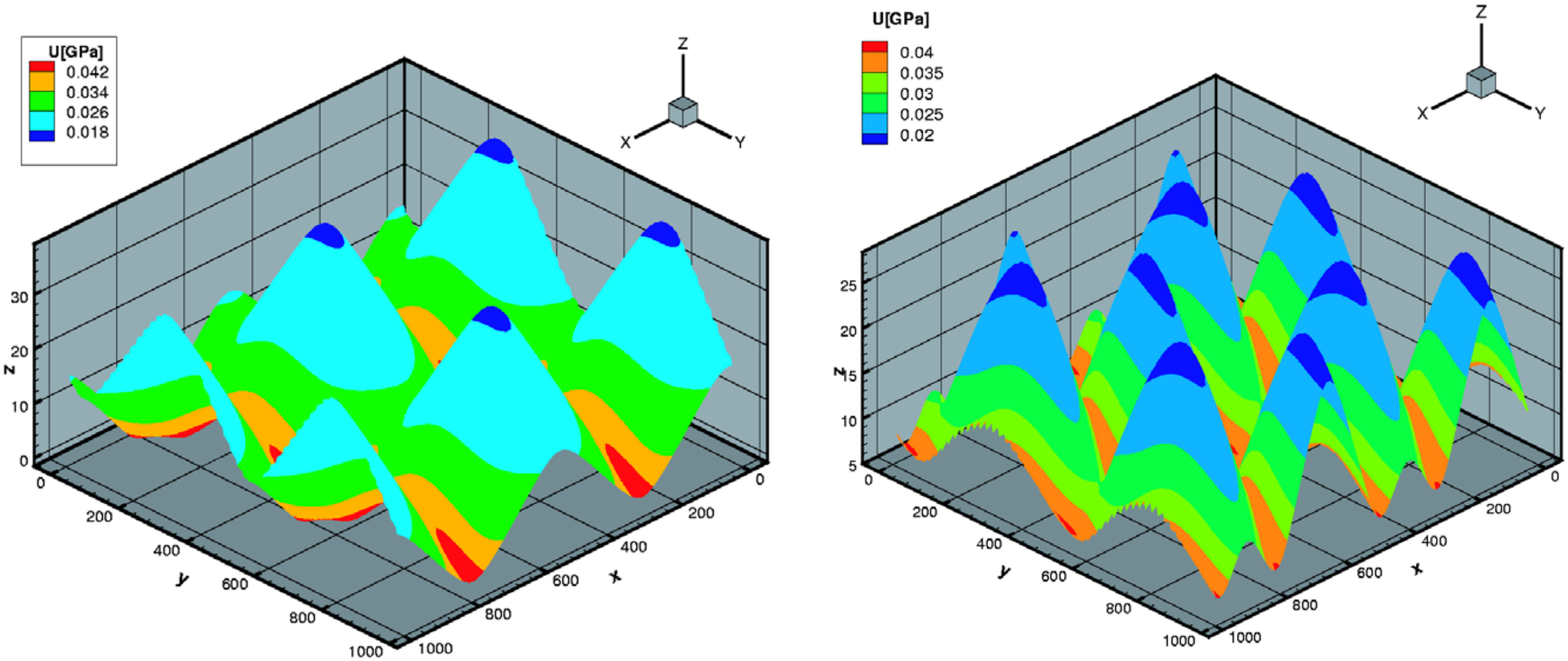
Elastic-plastic film - Biaxial stress



(a) (b)

Surface profile and free energies for elastic-plastic film.
a) Low misfit strain; b) High misfit strain.

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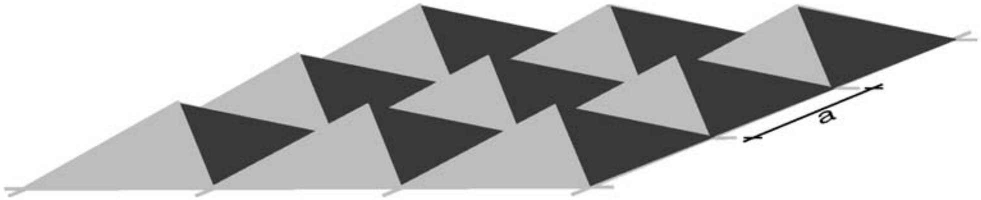
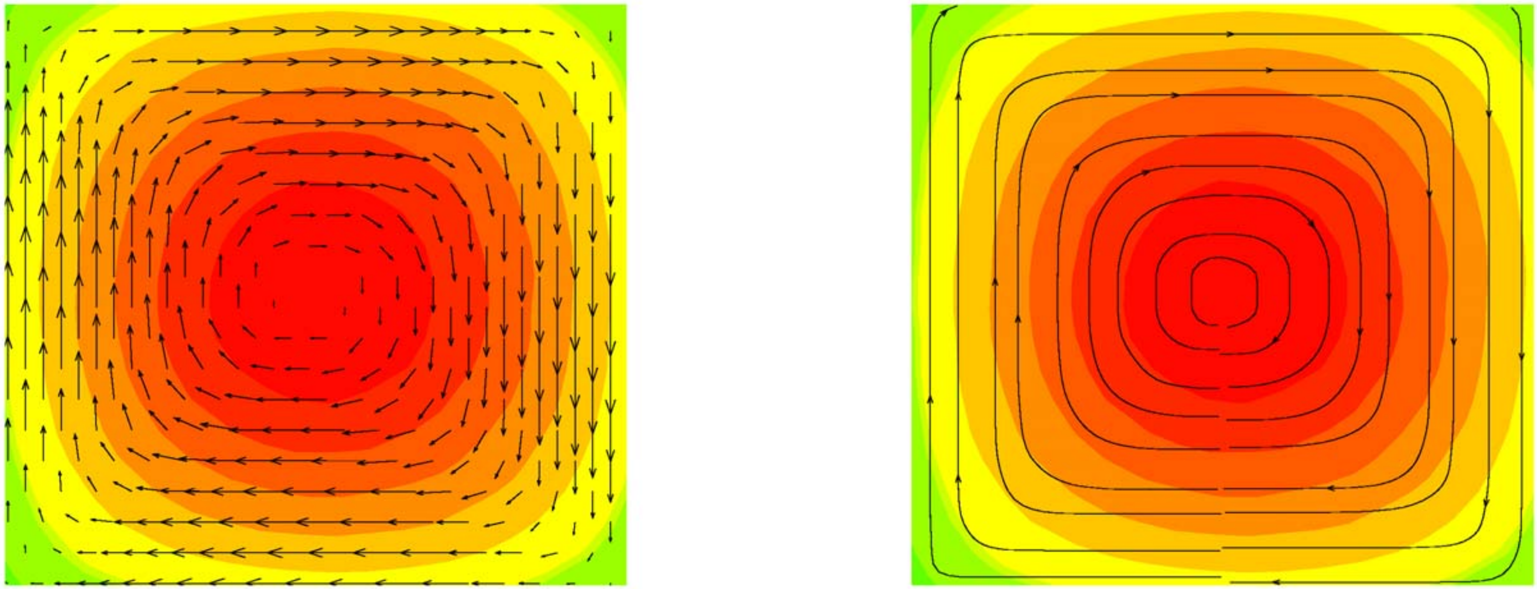
Interfacial prismatic dislocations





Interfacial dislocation density and prismatic dislocation loops.

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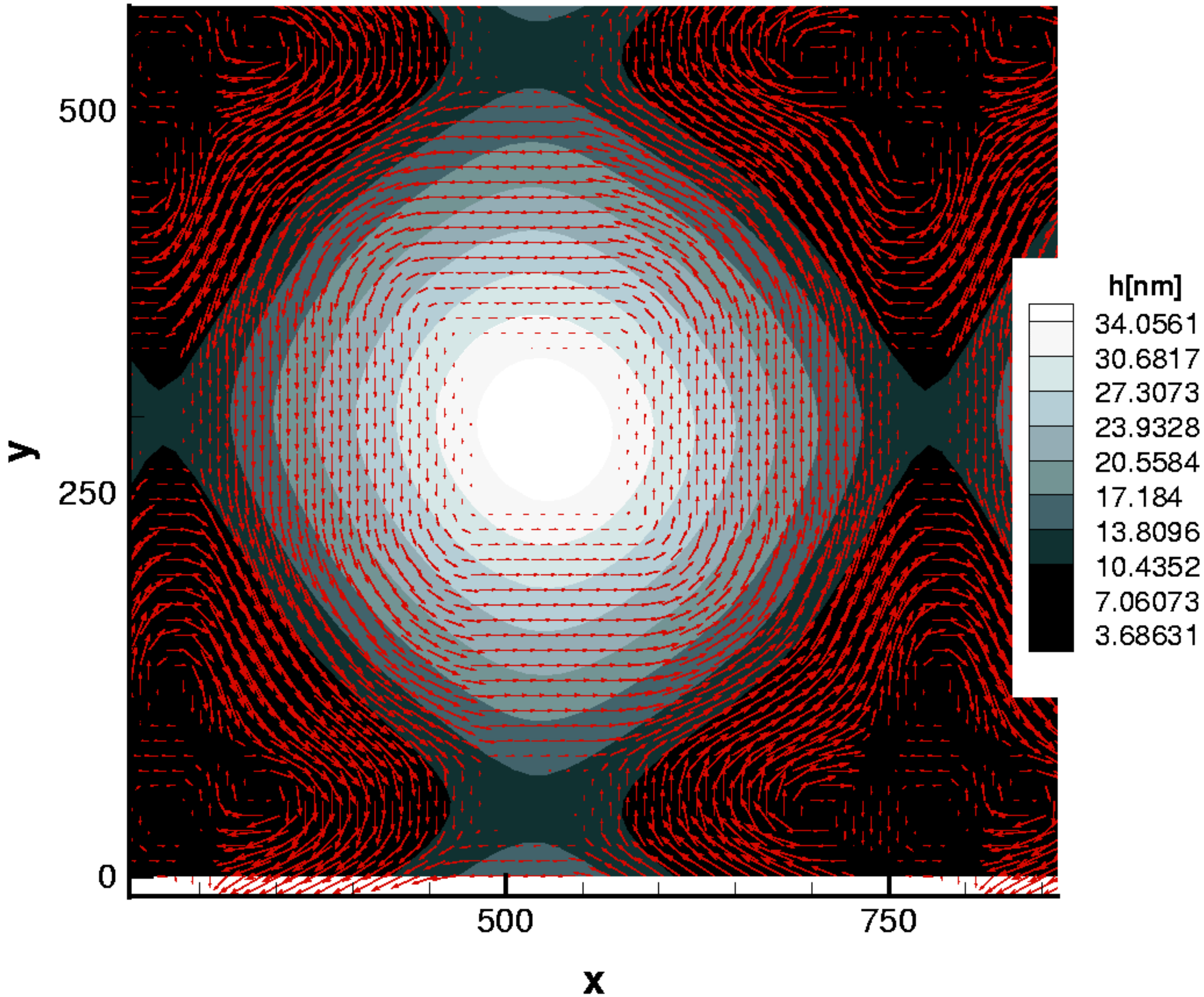
Interfacial prismatic dislocations



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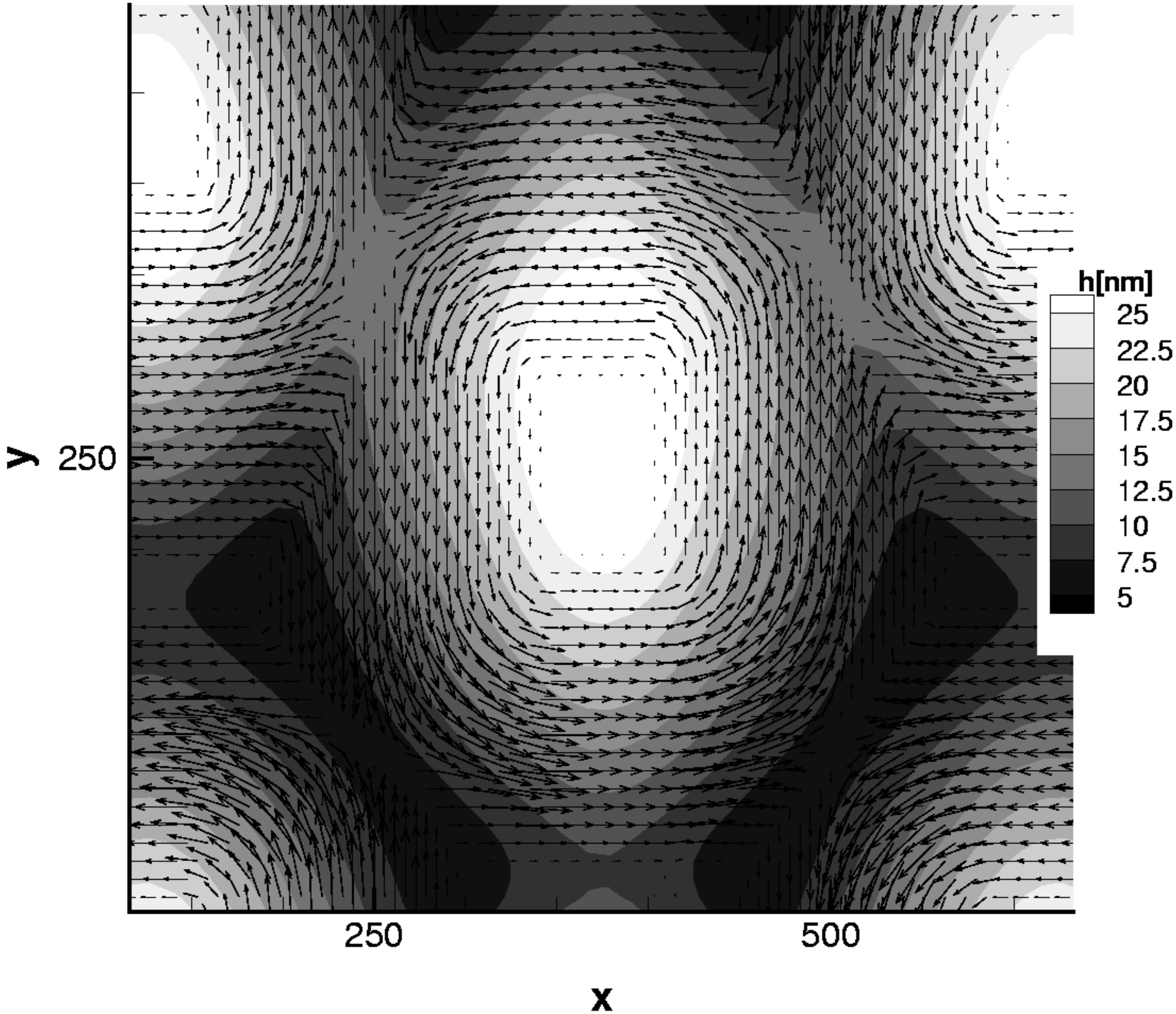
Interfacial prismatic dislocations



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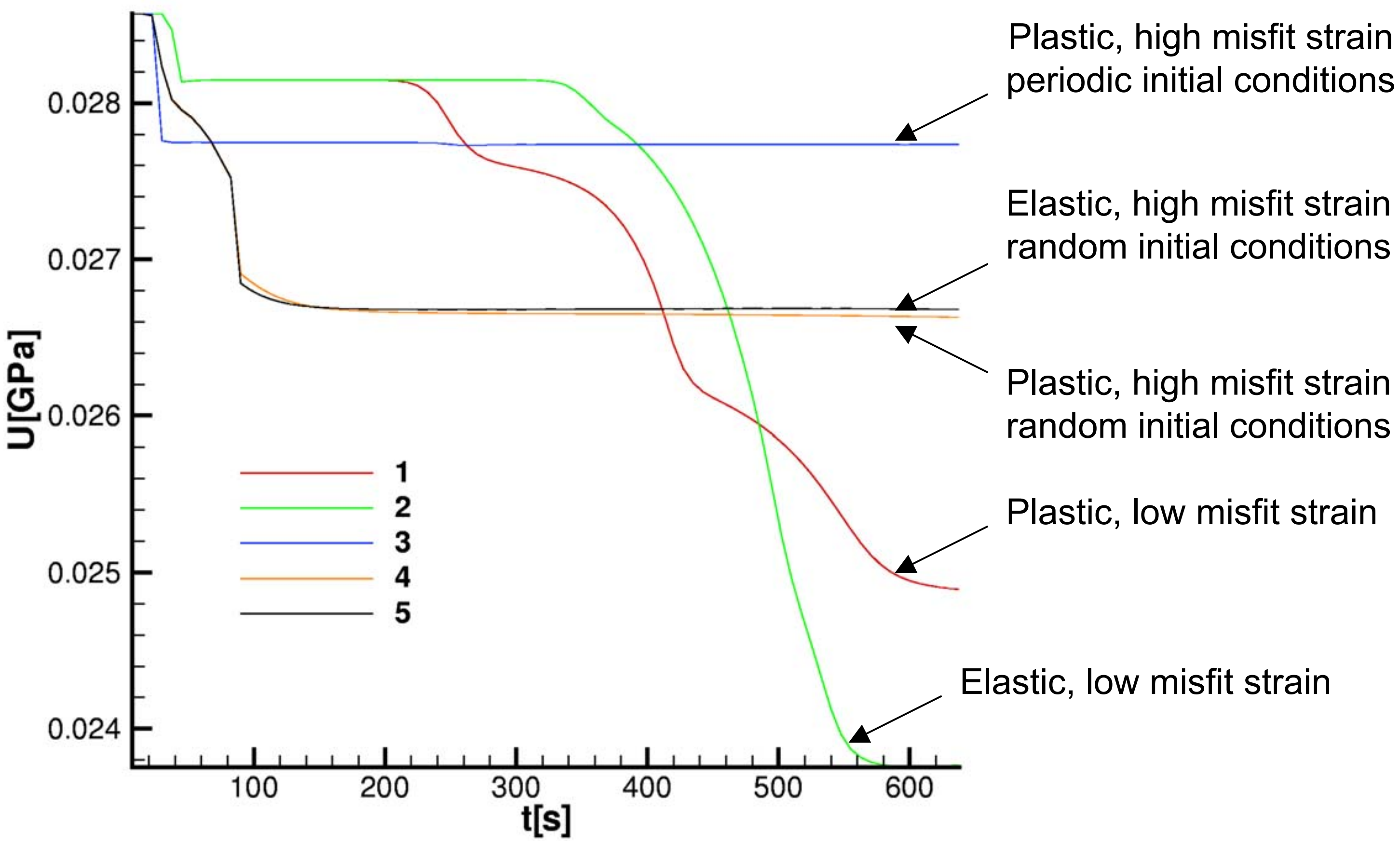
Strain-energy relaxation histories



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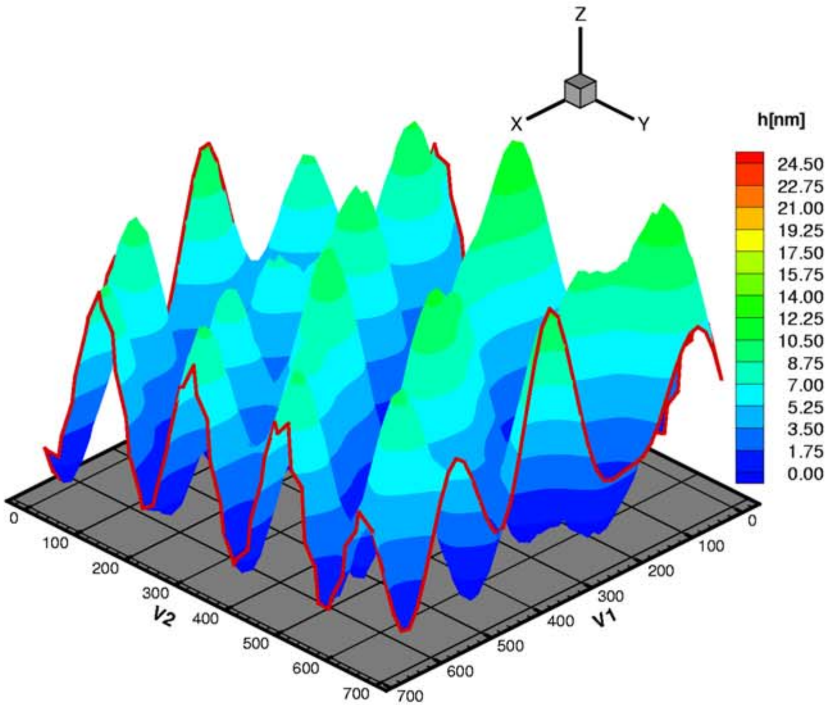
Summary and conclusions

- Continuum model of YBCO film growth predicts:
  - Initial Volmer-Weber island growth.
  - 'Magic slope'.
  - $t^{1/3}$ coarsening law.
- Asymptotic expansion leads to simple expression for elastic energy in the limit of small thickness, shallow profile.
- The effects of elastic strain are:
  - Islands become more 'dome-like'.
  - Islands tend to elongate and to align into lattices.
  - Coarsening rate is slowed down.
  - A stationary island size becomes possible.

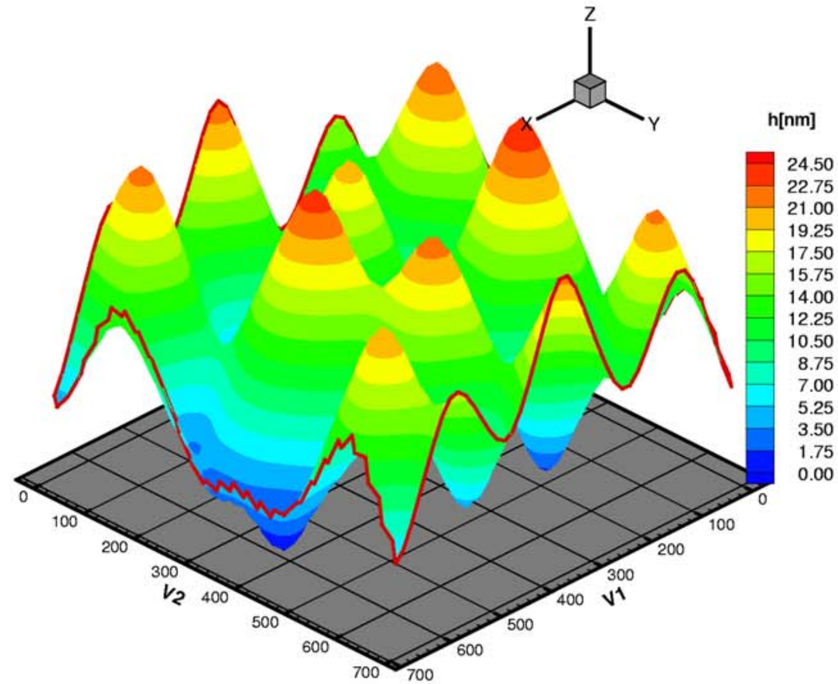
Summary and conclusions

- Crystallographic slip may be taken into account in a 'mean-field' sense by recourse to single-crystal plasticity.
- The elastic field induced by a rough strained film is relaxed by screw dislocations with Burgers vector normal to the substrate.
- The effects of crystallographic slip are:
  - Mitigates the effects of elasticity.
  - Results in a distribution of interfacial prismatic loops.

Strained elastic film growth



(a) t = 40 s



(b) t = 132 s

Growth of film with large biaxial misfit stresses, no crystallographic slip.

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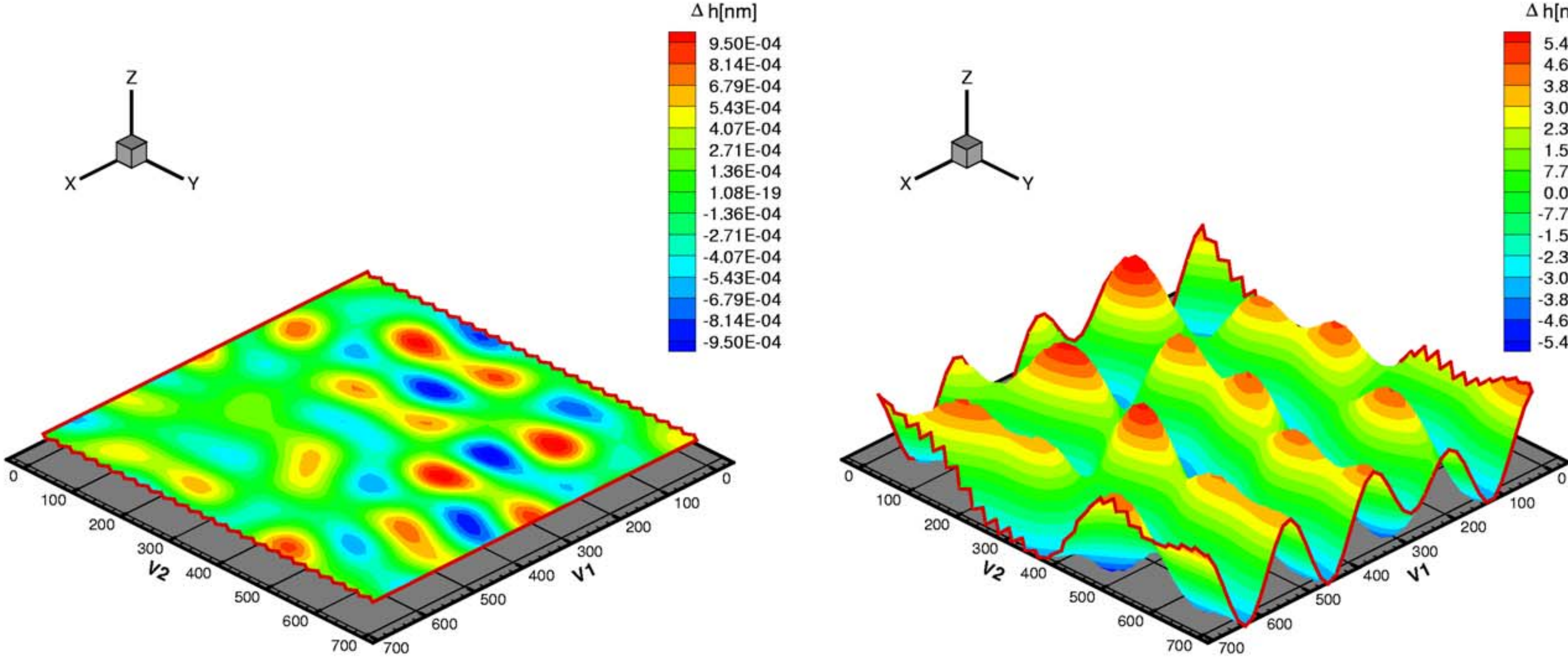
Strained elastic film growth



(c) t = 12 s

(d) t = 42 s

Growth of film with large uniaxial misfit stresses, no crystallographic slip.

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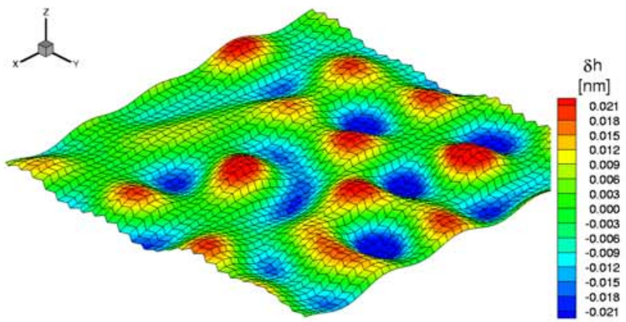
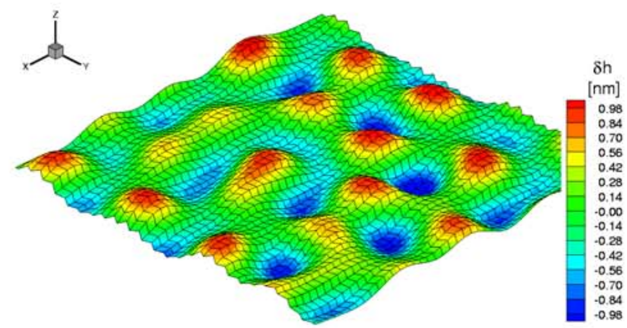
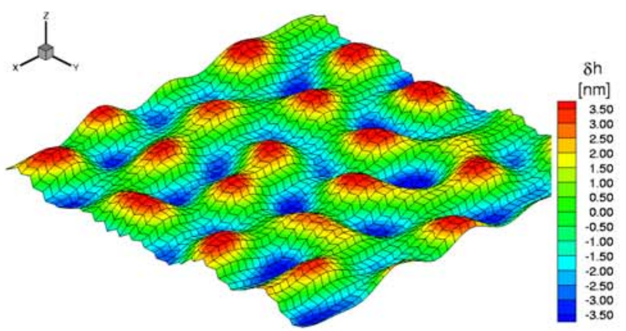
Comparison of growth models



(a)



(c)



(b)

Film profiles at 21.5 s. a) Diffusion only; b) Diffusion and elasticity; c) Diffusion, elasticity and crystallographic slip.