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

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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.)  
Continuum film-growth model


- Assumptions:
  - $h_{,t}$ admits Taylor expansion in $\nabla h$, $\nabla \nabla h$, . . .
  - Invariance under $x \rightarrow -x$.
  - Decay rate no faster than $L^{-4}$.
  - Onsager reciprocity relations.

- Rate equation for surface profile:
  \[ h_{,t} = F(\nabla h) - V \left( \frac{h}{\delta} \right) e^{-h/\delta} + C \nabla \cdot [(k^{-2}|\nabla h|^2 - 1)\nabla h] - D \nabla^4 h \]

  \[
  F(\nabla h) \rightarrow \text{deposition flux} \quad -V \left( \frac{h}{\delta} \right) e^{-h/\delta} \rightarrow \text{interfacial energy}
  
  +C \nabla \cdot [(k^{-2}|\nabla h|^2 - 1)\nabla h] \rightarrow \text{evaporation/condensation}
  
  -D \nabla^4 h \rightarrow \text{capillarity} \quad k \rightarrow \text{magic slope} \]
Continuum film-growth model


- From Onsager reciprocity relations: \( h_{,t} = -\delta \frac{\partial \Phi[h]}{\partial h} \)
- Kinetic potential:

\[
\Phi[h] = \int_\Omega \left\{ -Fh - V(h + \delta)e^{-h/\delta} + \frac{C}{4k^2} (|\nabla h|^2 - k^2)^2 + \frac{D}{2}(\nabla^2 h)^2 \right\} d^2x
\]
Stability of flat films - island nucleation


- Linearization: \( h_0 \rightarrow h_0 + u \),

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

- Set: \( u = Ae^{-\lambda t}e^{-ik \cdot x} \),

\[
\lambda = Dk^4 - Ck^2 + C
\]

- Critical wavelength:

\[
k_c = \sqrt{C/2D}
\]

- \( h_c = 0 \Rightarrow \text{Volmer-Weber}. \)

- \( h_c > 0 \Rightarrow \text{Stranski-Krastanow}. \)

Critical thickness \( h_c \).
Surface-profile evolution


Level contours of surface height (nm).

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Systems with small capillarity


- $D \to 0$, sharp-interface approximation.
- Boundary layer width: $l = \sqrt{D/2C} \to 0$.
- Relaxation time: $\tau = D/2C^2 \to 0$.
- Limiting kinetic potential: $\Phi_0[h] = \int_S \sqrt{2CD} \left[ 2k_1^3(s)/3k \right] ds$, $S \equiv$ singular set of the profile.
- Limiting kinetics: $h_{\gamma t} = -\delta \Phi_0[h]/\delta h$. 
Coarsening construction


Evolution of singular set (ridges and grooves).
Coarsening construction


- Effective kinetic equation:

  \[
  h_{1,t} = \begin{cases} 
  \frac{80}{3} \frac{\sqrt{2CD\, k^4}}{12h_1^2 - 4ah_1k + 3a^2k^2}, & h_1 < ak \\
  \frac{16}{3} \frac{\sqrt{2CD\, k^4}}{-20h_1^2 + 60ah_1k - 29a^2k^2}, & h_1 > ak 
  \end{cases}
  \]

- 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:

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

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 \( \epsilon_{ij}^* \), misfit stresses \( \sigma_{ij}^* \), traction-free surface: \( \sigma_{3i}^* = 0 \).

- Perturb state of film by:
  - Roughening film profile: \( \langle h \rangle \rightarrow h(x_1, x_2) \).
  - Crystallographic slip \( \Rightarrow \) plastic strains \( \epsilon_{ij}^p \).

- Free energy:

\[
E = \int_{\Omega} \int_{0}^{h(x_1, x_2)} \frac{1}{2} c_{ijkl} (\epsilon_{ij} - \epsilon_{ij}^* - \epsilon_{ij}^p) (\epsilon_{kl} - \epsilon_{kl}^* - \epsilon_{kl}^p) \, d\Omega \, dx_3 \\
+ \int_{\Omega} \int_{0}^{h(x_1, x_2)} W^p \, d\Omega \, dx_3 + \int_{\Omega} \int_{-\infty}^{0} \frac{1}{2} c_{ijkl} u_{i,j} u_{k,l} \, d\Omega \, dx_3
\]

\( W^p \equiv \) stored energy of cold work.

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Crystallographic slip - Continuum theory

- Plastic strains: \( \epsilon_{\alpha 3}^p = \gamma_{\alpha}^p / 2 \)

- Density of screw dislocations:
  \( \rho = (\gamma_{2,1}^p - \gamma_{1,2}^p) / b \)

- Density of interfacial dislocations:
  \( \alpha_{31} = -\beta_{32}^p, \quad \alpha_{32} = \beta_{31}^p \quad \Rightarrow \quad \) prismatic loops.

- \( c \)-axis wobble:
  \( \theta^p = (1/2) \sqrt{(\gamma_1^p)^2 + (\gamma_2^p)^2} \)

- Yield condition: \( |\sigma_{\alpha 3}| = \tau_c \).

- Ideal plasticity: \( \tau_c = \text{constant} \).

Slip planes (a and b) and Burgers vector (c) in YBCO.
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 $\varepsilon \to 0$:
  \[
  \Delta E \sim - \int_{\Omega} \int_{\Omega} \frac{1}{2} G_{ik}(x - x') t_i(x) t_k(x') d\Omega d\Omega'
  \]
  where $G \equiv$ surface Green’s function for substrate.

- Effective tractions: $t_i \sim -\sigma^*_{i\beta} h,_{\beta} - (h c_{\beta kl} \epsilon^p_{kl}),_{\beta}$.
  1. $-\sigma^*_{i\beta} h,_{\beta} \Rightarrow$ effect of waviness (Gao, 1991).
  2. $(h c_{\beta kl} \epsilon^p_{kl}),_{\beta} \Rightarrow$ effect of crystallographic slip.
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: $u = A e^{\lambda t} e^{i k \cdot x}$
- Biaxial stress: $k_1 = k_2 = k$,
  \[ \lambda = -2C' k^2 + 4D k^4 - 4\sqrt{2}D k^3 \gamma^{-1} \sigma^* \sqrt{2} / E'_s \]
- Steady profile: $\lambda = 0$,
  \[ k \sim \sqrt{2} \sigma^* \sqrt{2} / \gamma E'_s, \quad \text{for } \sigma^* \text{ large.} \]
- Uniaxial stress: $\sigma^*_{11} = \sigma^*$, $k_1 = k$, $k_2 = 0$,
  \[ \lambda = -C' k^2 + D k^4 - 2D k^3 \gamma^{-1} \sigma^* \sqrt{2} / E'_s \]
- Steady profile: $\lambda = 0$,
  \[ k \sim 2 \sigma^* \sqrt{2} / \gamma E'_s, \quad \text{for } \sigma^* \text{ large.} \]
Elastic-plastic film - Biaxial stress

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

\[ x \]

\[ y \]

\[ h[\text{nm}] \]

- 34.0561
- 30.6817
- 27.3073
- 23.9328
- 20.5584
- 17.184
- 13.8096
- 10.4352
- 7.06073
- 3.68631
Interfacial prismatic dislocations
Strain-energy relaxation histories

Plastic, high misfit strain
periodic initial conditions

Elastic, high misfit strain
random initial conditions

Plastic, high misfit strain
random initial conditions

Plastic, low misfit strain

Elastic, low misfit strain
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) $t = 40 \text{ s}$
(b) $t = 132 \text{ s}$

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

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Strained elastic film growth

(c) $t = 12\,\text{s}$  

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

(d) $t = 42\,\text{s}$

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Comparison of growth models

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