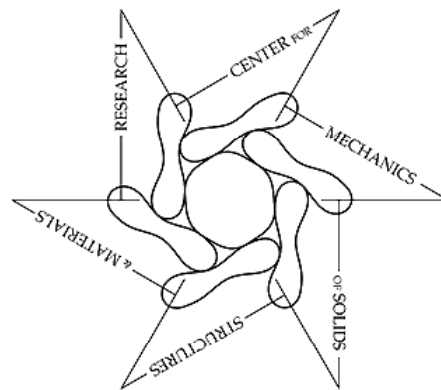




Nonlinear Dynamics of Wrinkle Growth and Pattern Formation in Stressed Elastic Thin Films

Se Hyuk Im and Rui Huang

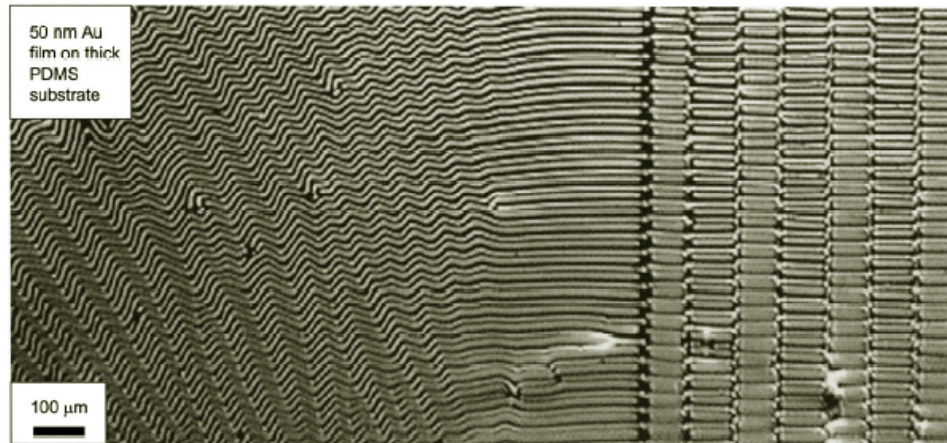
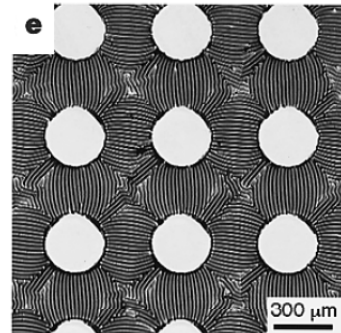
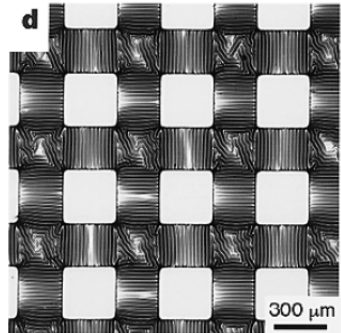
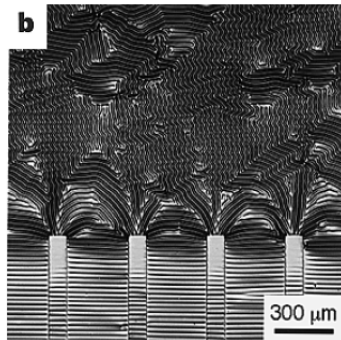
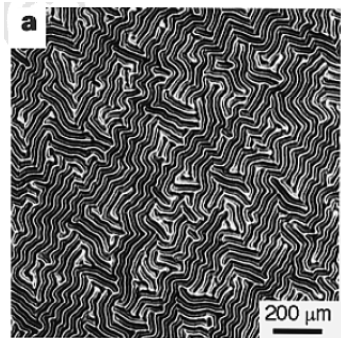
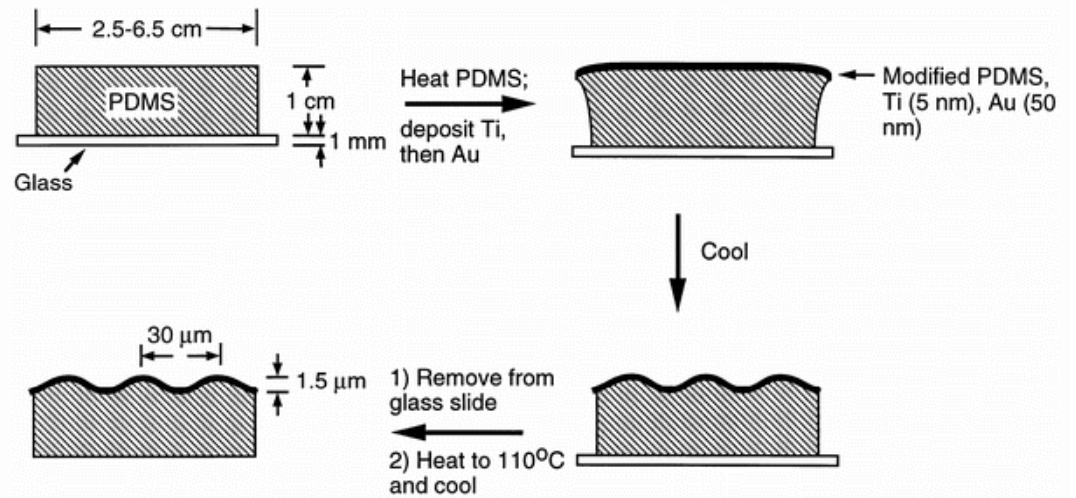
Center for Mechanics of Solids, Structures and Materials
Department of Aerospace Engineering and Engineering Mechanics
The University of Texas at Austin



----- Work supported by NSF CAREER Award (CMS-0547409) -----

Au film on PDMS: *thermoelastic wrinkling*

- Bowden et al., 1998 & 1999
- Huck et al., 1999
- Efimenko et al., 2005



SiGe film on BPSG: *viscous wrinkling*

- Hobart et al., 2000
- Yin et al., 2002 & 2003
- Peterson et al., 2006
- Yu et al., 2005 & 2006

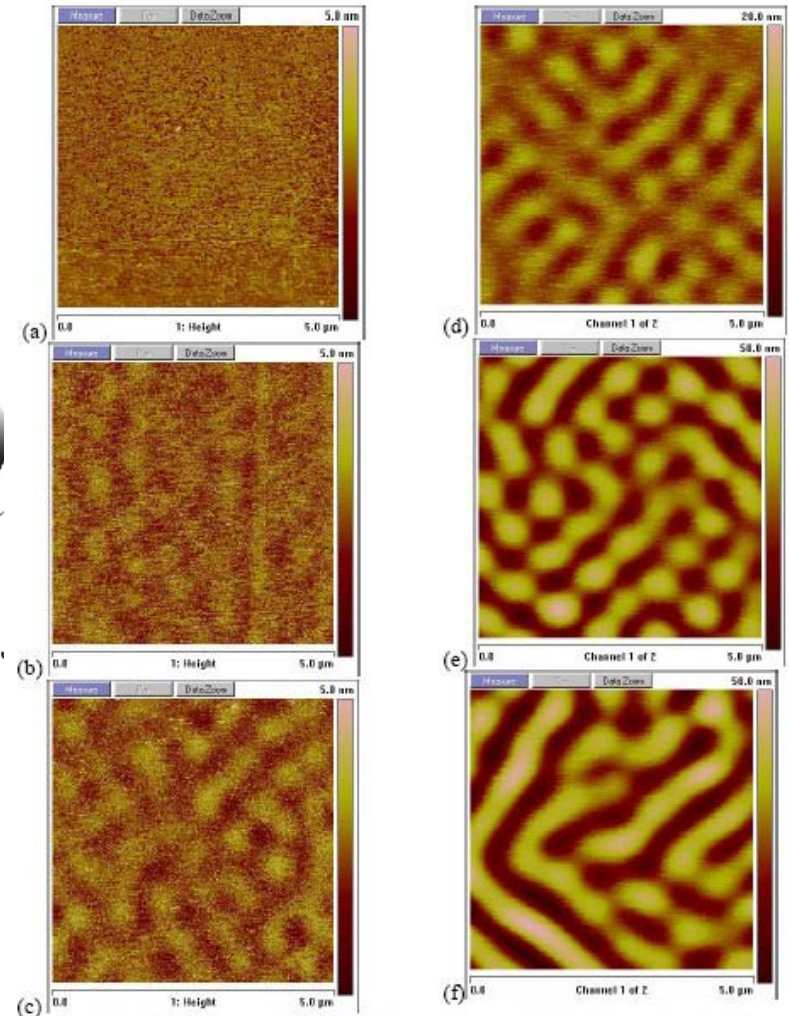
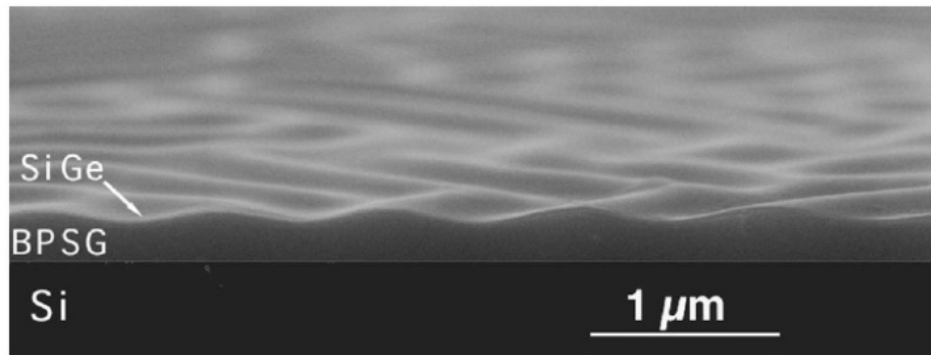
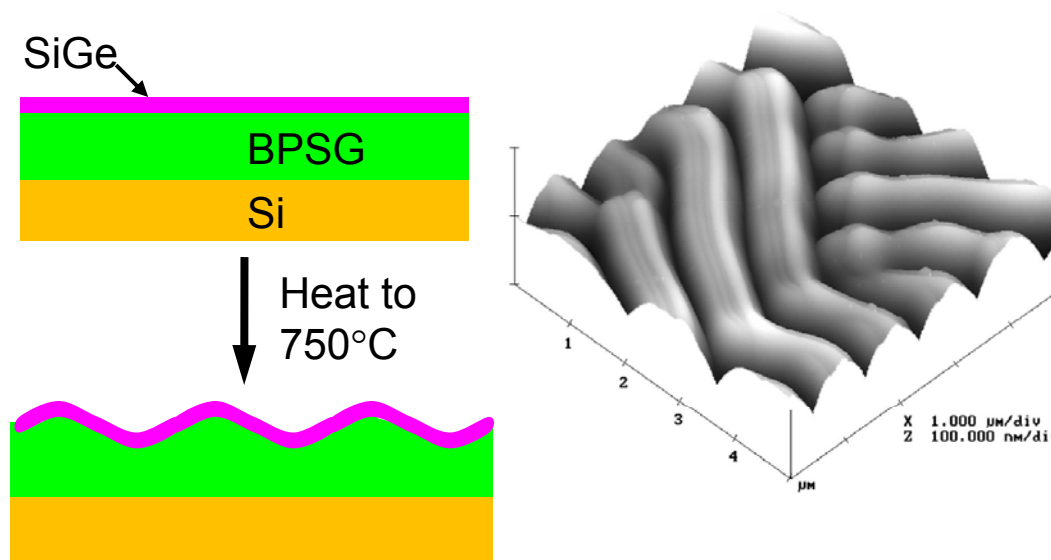


Figure 6.13: AFM images of buckled SiGe on 235-nm BPSG at the center of 150- μm x 150- μm islands after 750°C anneals of various lengths: (a) no anneal, (b) 25 min, (c) 50 min, (d) 105 min, (e) 253 min, and (f) 600 min. The AFM scan size is 5 μm x 5 μm , and the z-axis scale is indicated for each image. The scan edge is along a $\langle 110 \rangle$ crystal direction; buckles are along $\langle 100 \rangle$, as before.

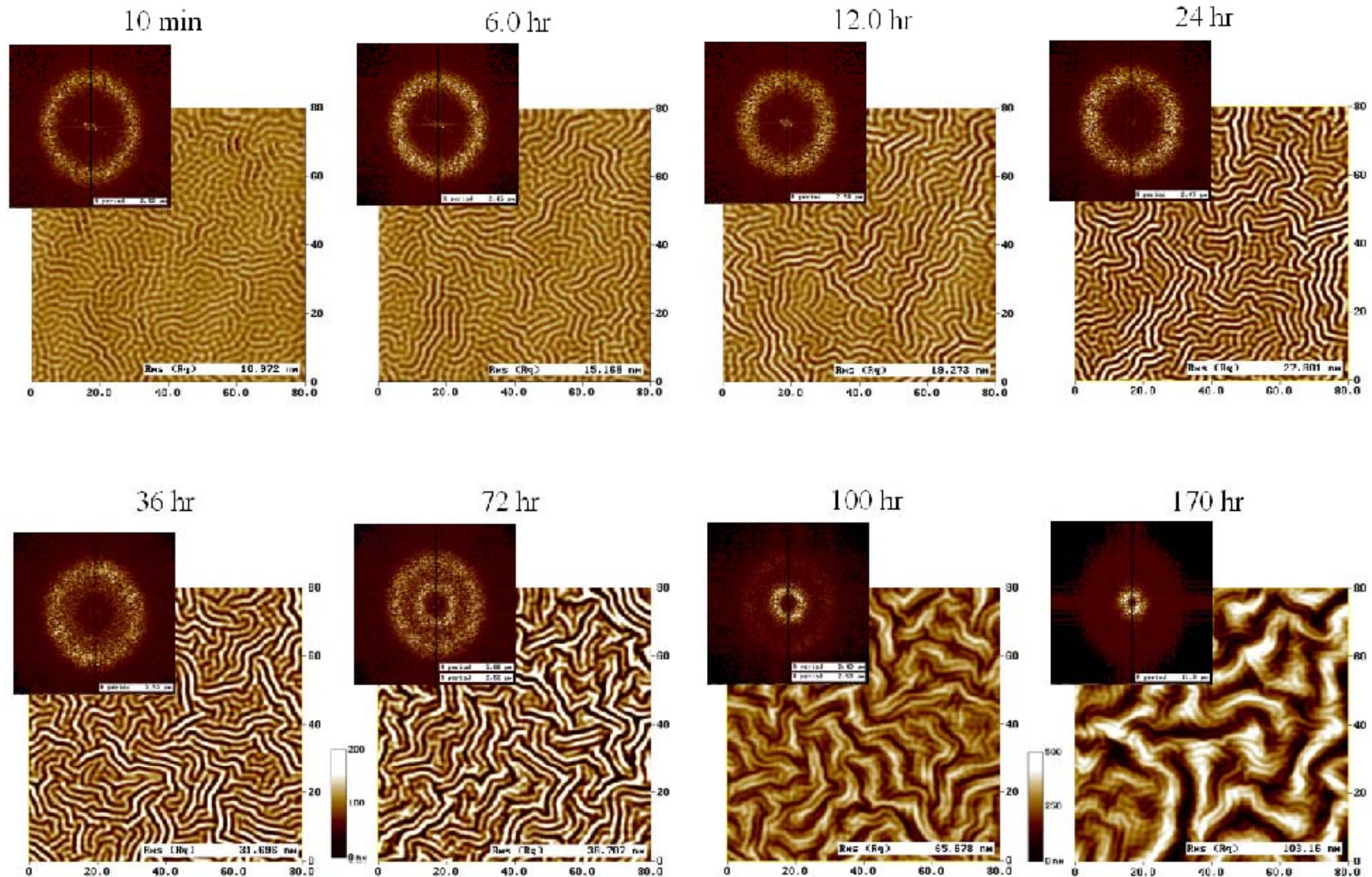
Elastic wrinkling vs ***Viscous wrinkling***

film/substrate materials	elastic/elastic	elastic/viscous
Onset of wrinkling	Critical stress or strain depends on the elastic modulus and thickness of substrate.	Unstable with any compressive stress in a thin film ($L/h \gg 1$).
Wrinkle amplitude	Equilibrium amplitude increases with the stress/strain.	Amplitude grows over time.
Wrinkle wavelength	Equilibrium wavelength depends on the elastic modulus and thickness of substrate.	The fastest growing wavelength dominates the initial growth, then coarsening over time.
Energetics	Spontaneous energy minimization.	Energy dissipation by viscous flow.

Al film on PS: *viscoelastic wrinkling*

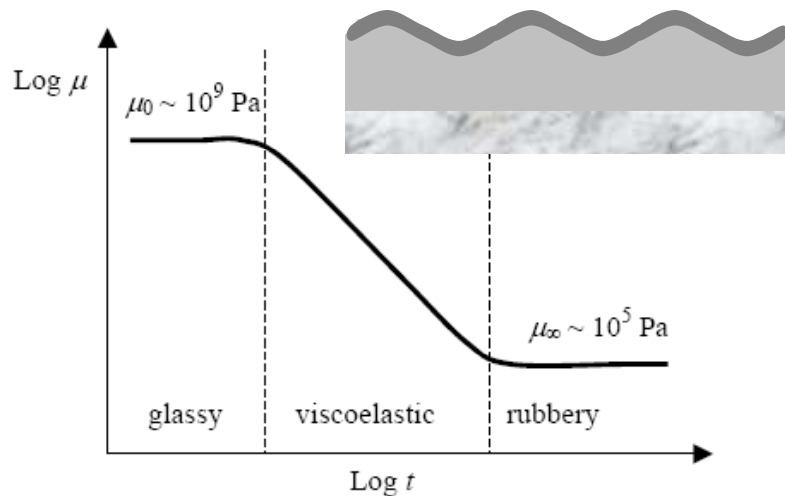
(Yoo and Lee, 2003-2005)

PS 400 nm (MW 1,340,000) Al 40nm, 145 °C



A model for viscoelastic wrinkling

Viscoelastic relaxation modulus

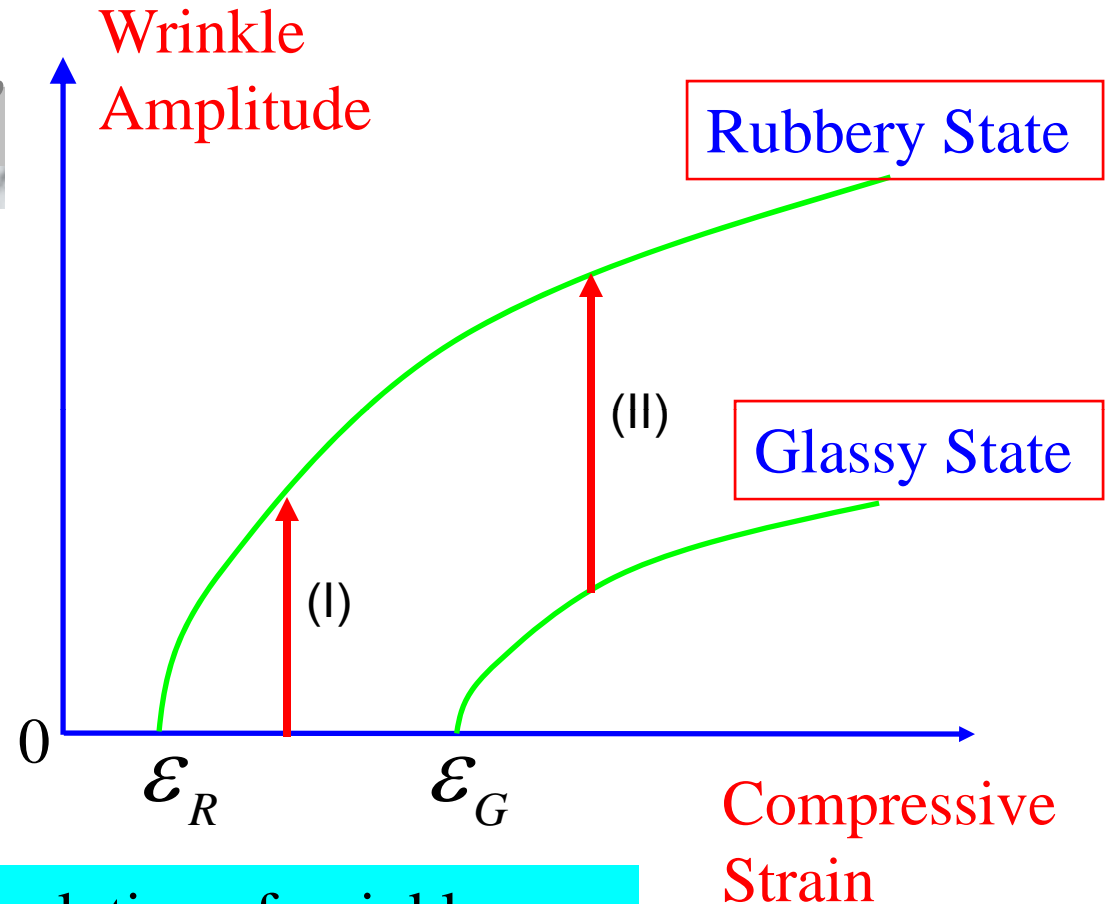


Equilibrium states at the elastic limits (thick substrate):

$$\varepsilon_c = -\frac{1}{4} \left(\frac{3\bar{E}_s}{\bar{E}_f} \right)^{2/3}$$

$$A_{eq} = h_f \left(\frac{\varepsilon}{\varepsilon_c} - 1 \right)^{1/2}$$

$$\lambda_{eq} = 2\pi h_f \left(\frac{\bar{E}_f}{3\bar{E}_s} \right)^{1/3}$$



Evolution of wrinkles:

- (I) Viscous to Rubbery
- (II) Glassy to Rubbery

Scaling of evolution equation

$$\frac{\partial w}{\partial t} = -K \nabla^2 \nabla^2 w + F \nabla \cdot (\boldsymbol{\sigma} \cdot \nabla w) - R w$$

$$K = \frac{(1-2\nu_s)\mu_f h_f^3 h_s}{12(1-\nu_s)(1-\nu_f)\eta_s} \quad F = \frac{1-2\nu_s}{2(1-\nu_s)} \frac{h_s h_f}{\eta_s} \quad R = \frac{\mu_R}{\eta_s}$$



Fastest growing mode:

$$\lambda_m = 2\sqrt{2}\pi L_1 \quad A = A_0 \exp\left(\frac{t}{4\tau_1}\right)$$

Length and time scales: $L_1 = \sqrt{-\frac{K}{F\sigma_0}} \quad \tau_1 = \frac{K}{(F\sigma_0)^2}$

critical stress:

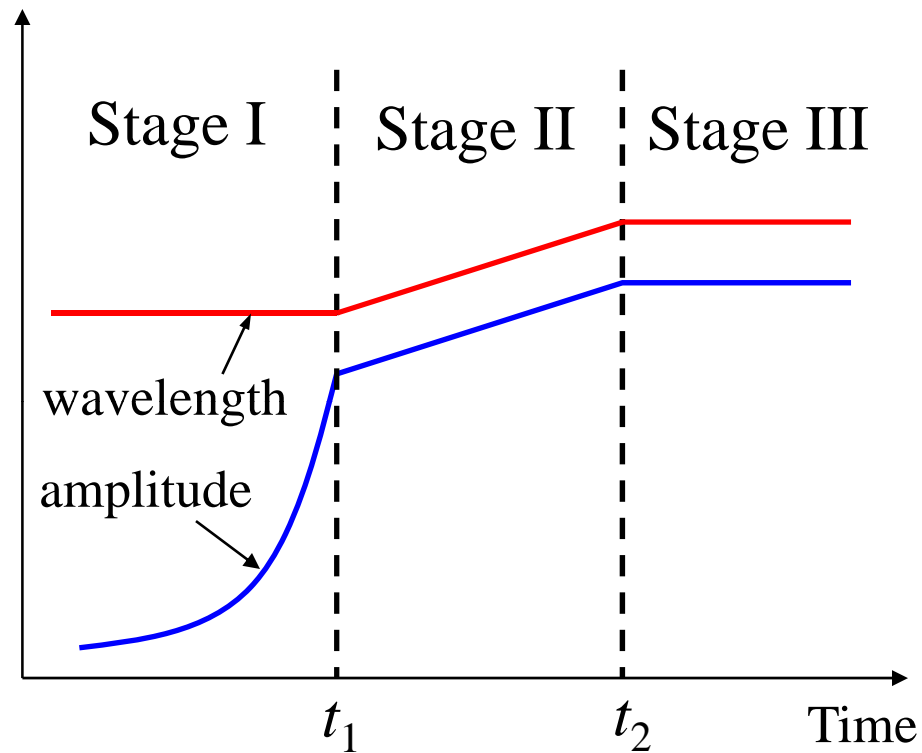
$$\sigma_c = -\sqrt{\frac{4KR}{F^2}}$$

Equilibrium state:

$$(\sigma_0 < \sigma_c)$$

$$\lambda_{eq} = 2\pi \left(\frac{K}{R}\right)^{1/4} > \lambda_m \quad A_{eq} = h_f \sqrt{\frac{2}{3} \left(\frac{\sigma_0}{\sigma_c} - 1\right)}$$

Three-Stage Viscoelastic Wrinkle Evolution

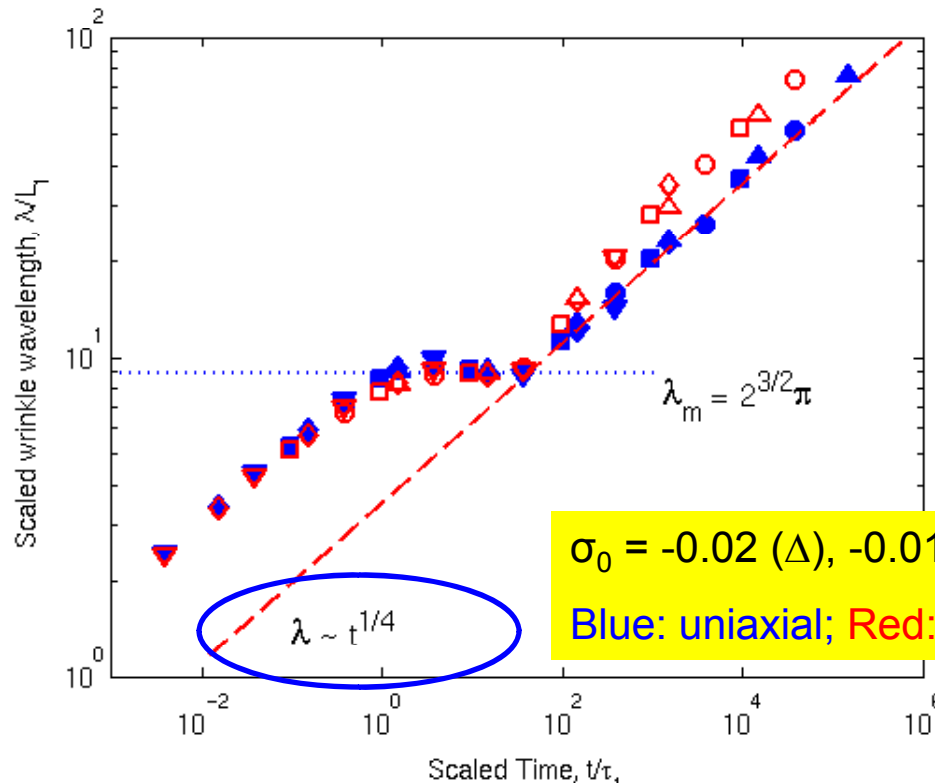


I: Exponential growth, dominated by the fastest growing mode

II: Coarsening ($\lambda_m \rightarrow \lambda_{eq}$, $A \uparrow$)

III: Nearly equilibrium (with local ordering)

Power-law Coarsening



Length and time scaling:

$$L_1 = \sqrt{-\frac{K}{F\sigma_0}}$$

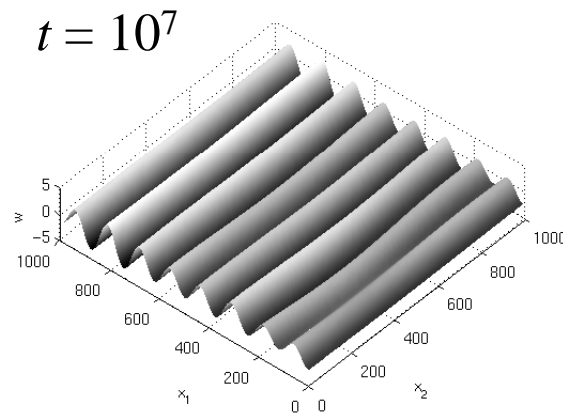
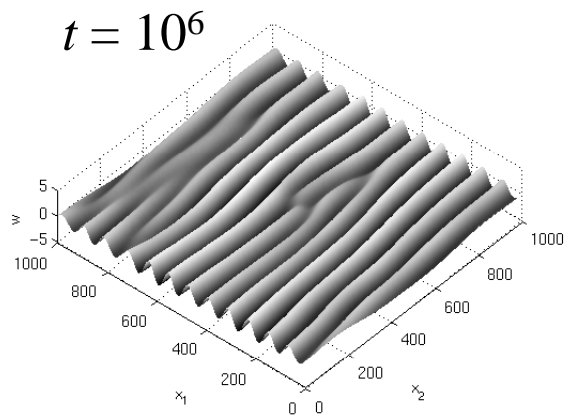
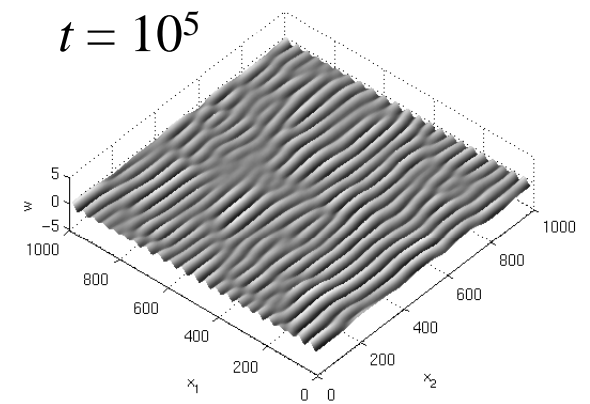
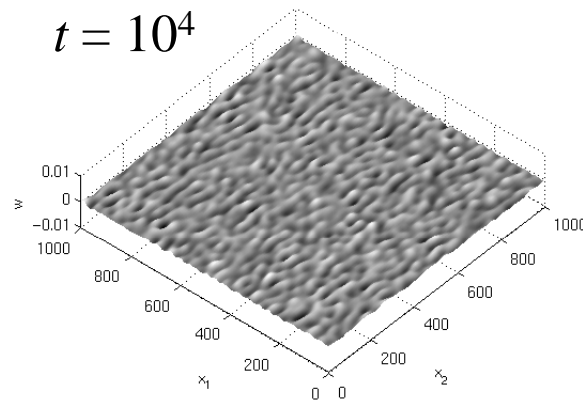
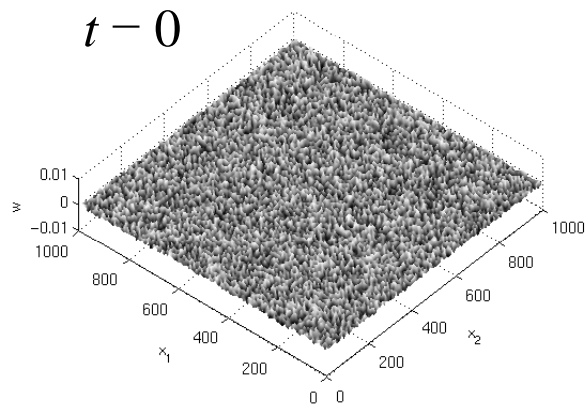
$$\tau_1 = \frac{K}{(F\sigma_0)^2}$$

$\sigma_0 = -0.02$ (Δ), -0.01 (\circ), -0.005 (\square), -0.002 (\diamond), -0.001 (∇)

Blue: uniaxial; Red: equi-biaxial

- On a compressible, thin viscoelastic substrate: $\lambda \sim t^{1/4}$;
- On an incompressible, thin viscoelastic substrate, $\lambda \sim t^{1/6}$;
- On a thick VE substrate, $\lambda \sim t^{1/3}$.
- The substrate elasticity slows down coarsening as it approaches the equilibrium state.

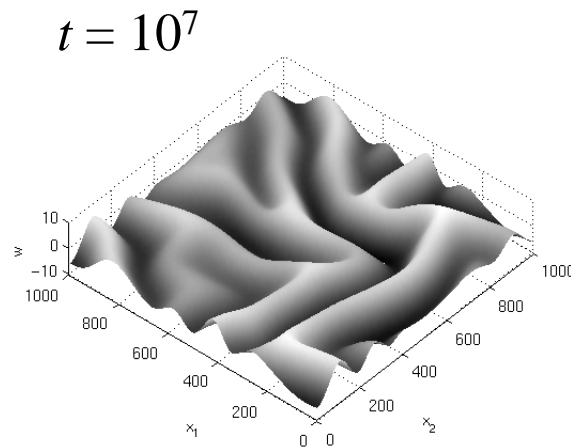
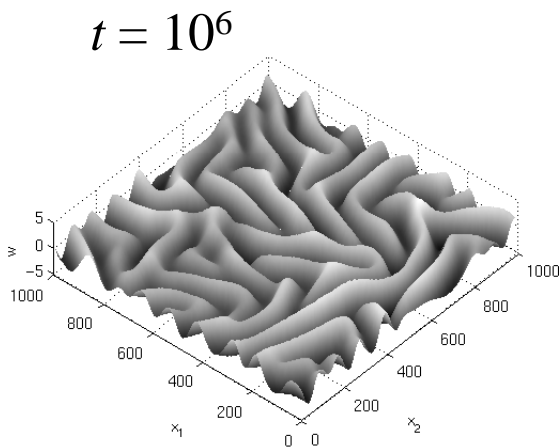
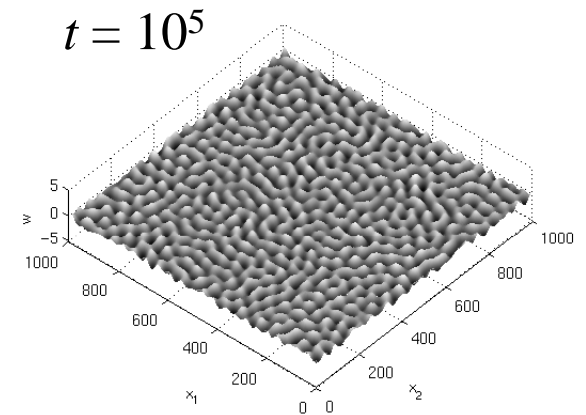
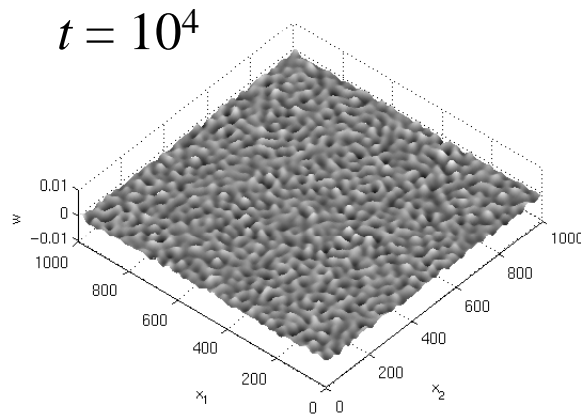
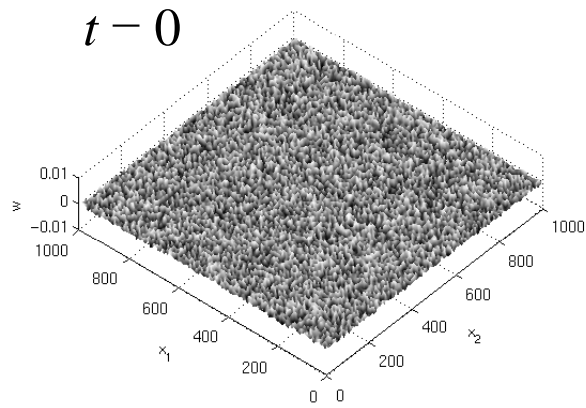
Numerical simulation: uniaxial wrinkling



$$\sigma_{11}^{(0)} = -0.01, \sigma_{22}^{(0)} = 0$$

$$\frac{\mu_R}{\mu_f} = 0$$

Numerical simulation: equi-biaxial wrinkling



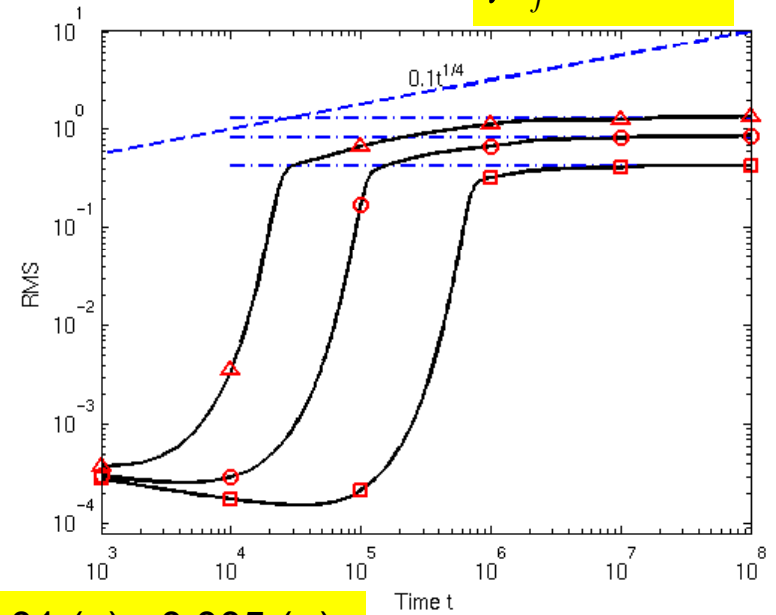
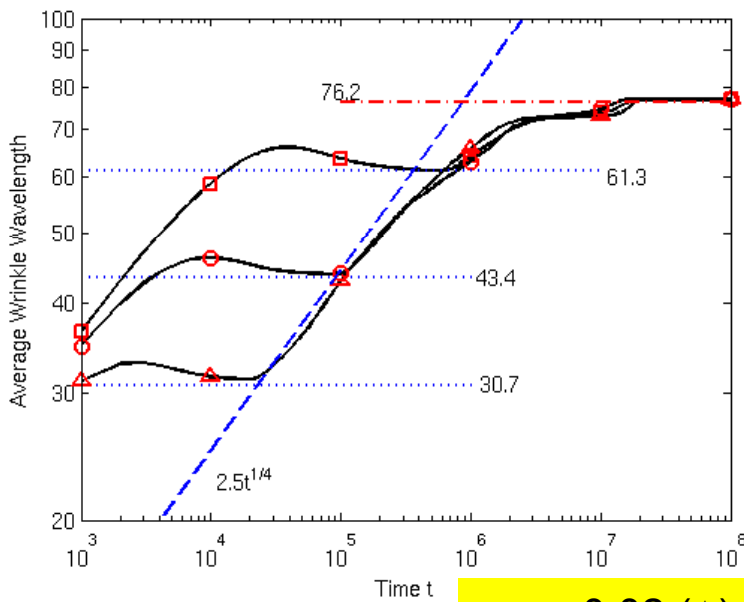
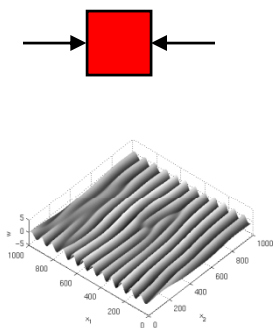
$$\sigma_{11}^{(0)} = \sigma_{22}^{(0)} = -0.01$$

$$\frac{\mu_R}{\mu_f} = 0$$

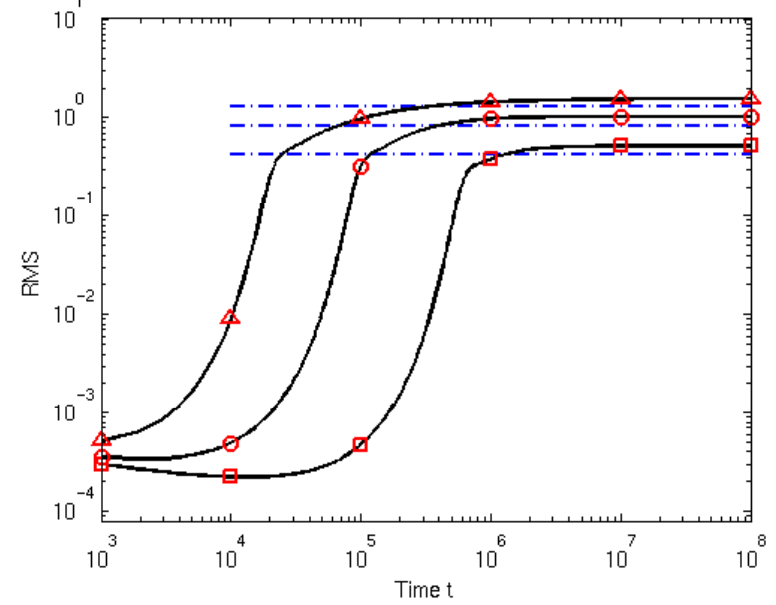
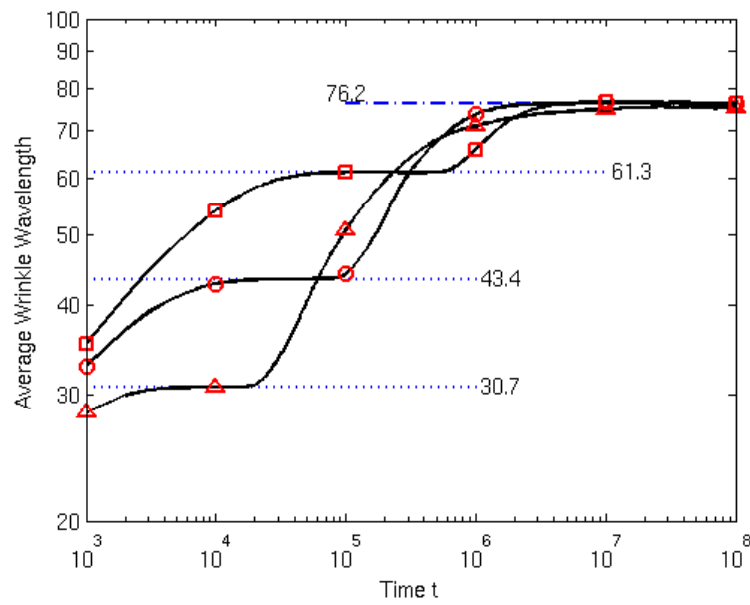
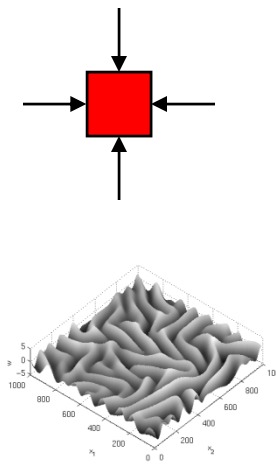
Huang and Im, PRE 2006.

Effect of Substrate Elasticity

$$\frac{\mu_R}{\mu_f} = 10^{-5}$$

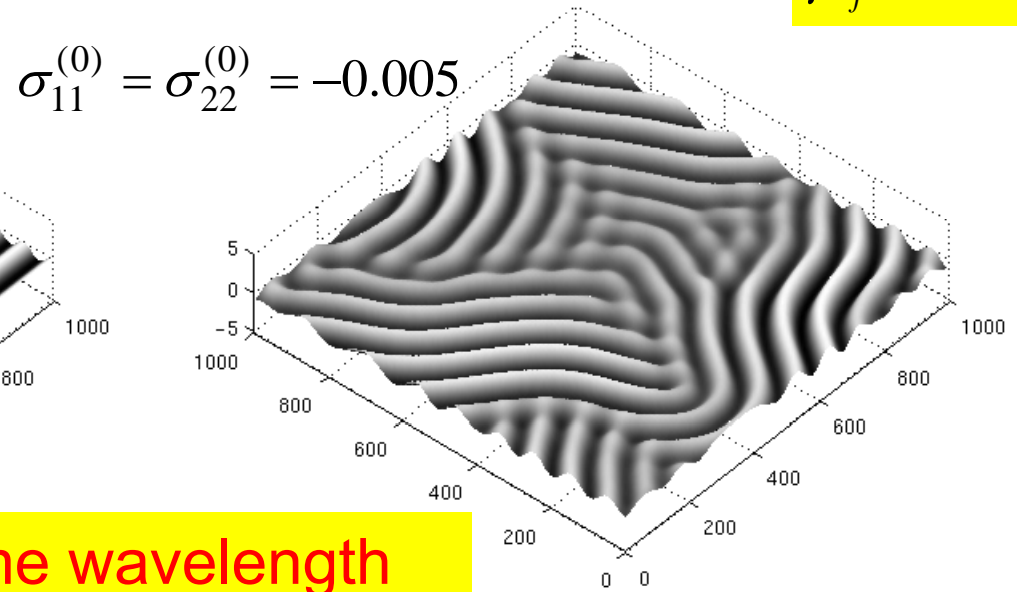
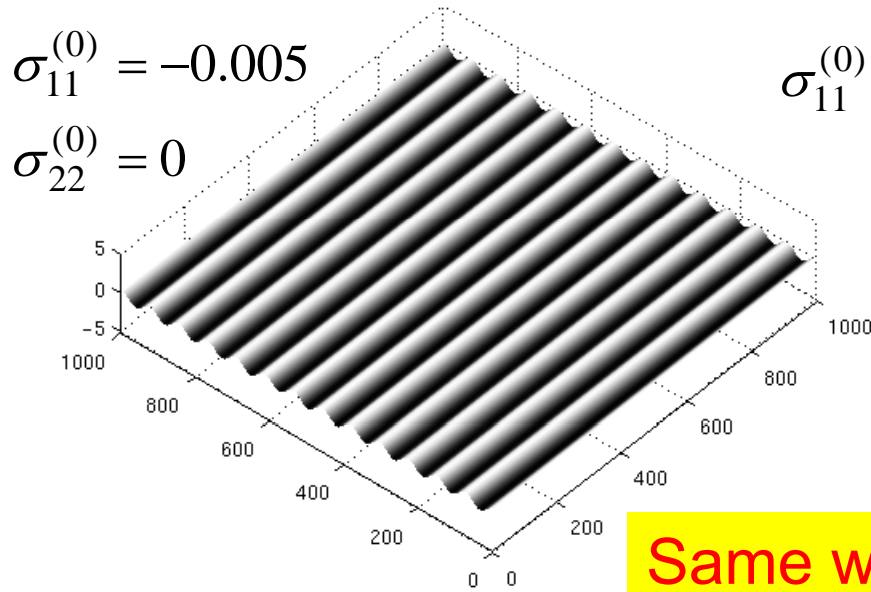


$\sigma_0 = -0.02$ (Δ), -0.01 (\circ), -0.005 (\square)

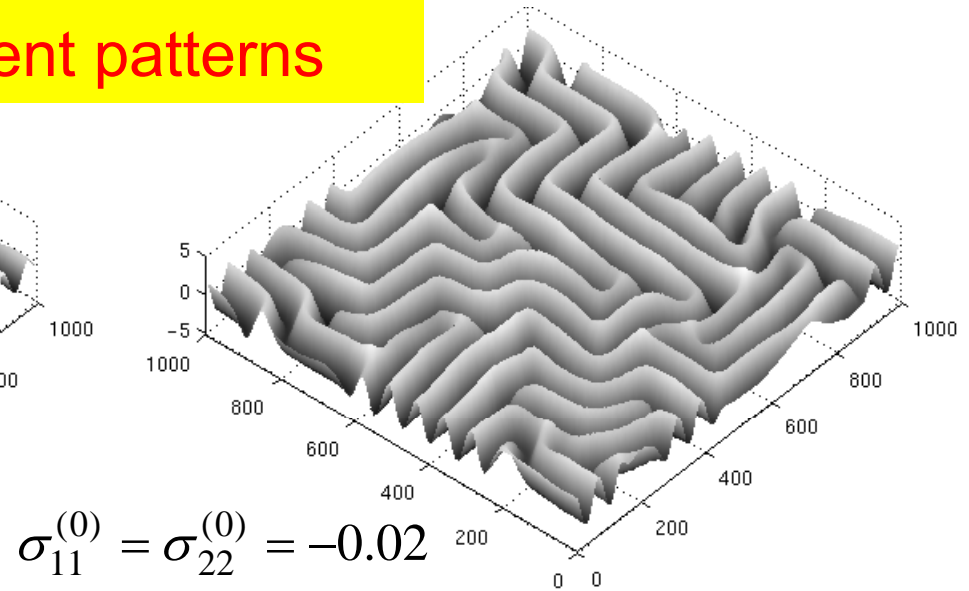
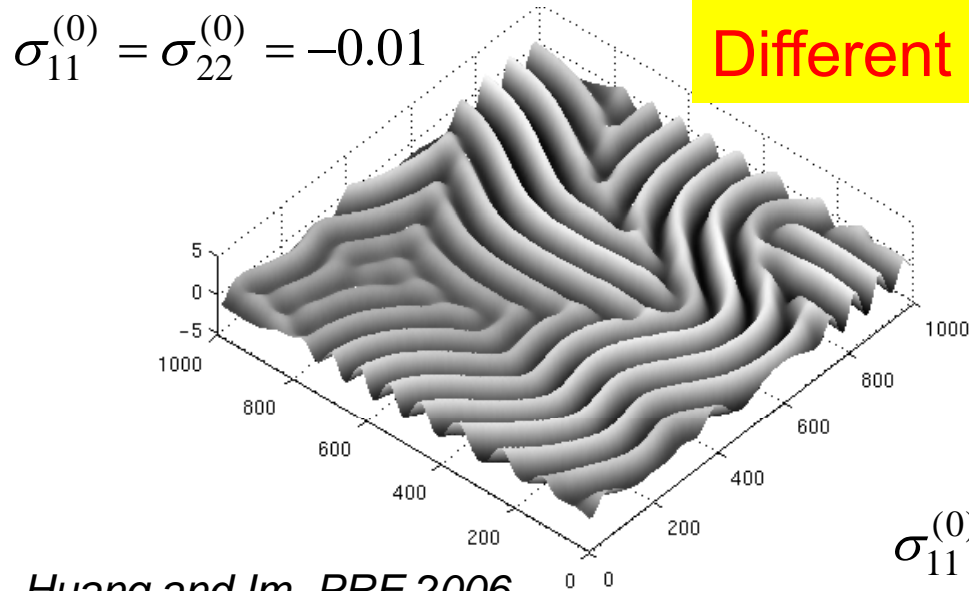


Near-Equilibrium Patterns

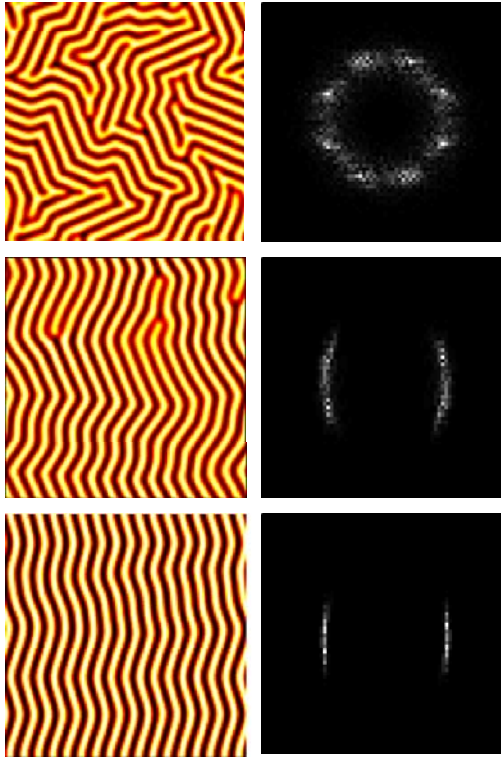
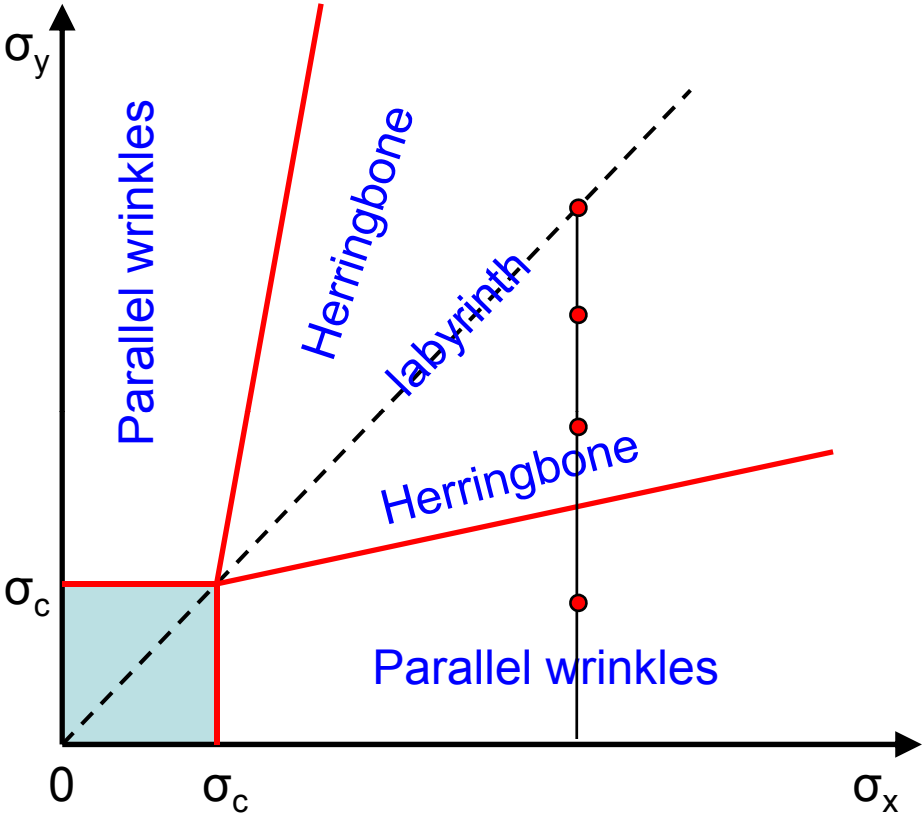
$$\frac{\mu_R}{\mu_f} = 10^{-5}$$



Same wavelength
Different patterns

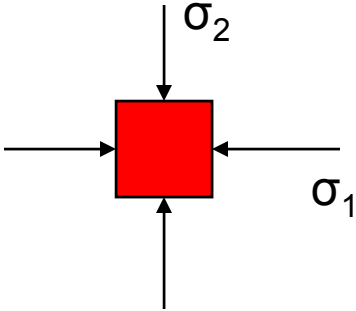
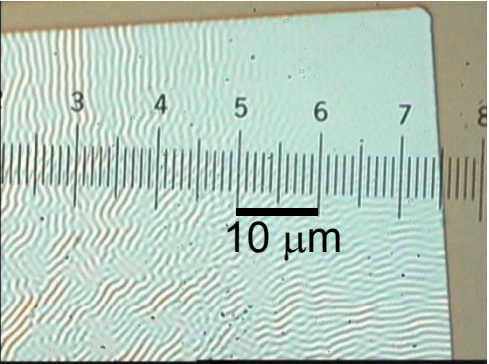
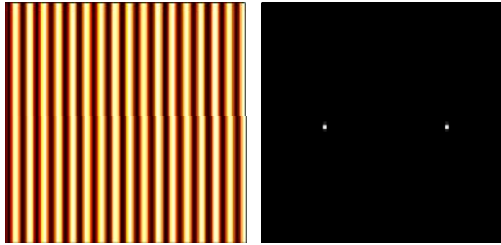


Wrinkle patterns under biaxial stresses

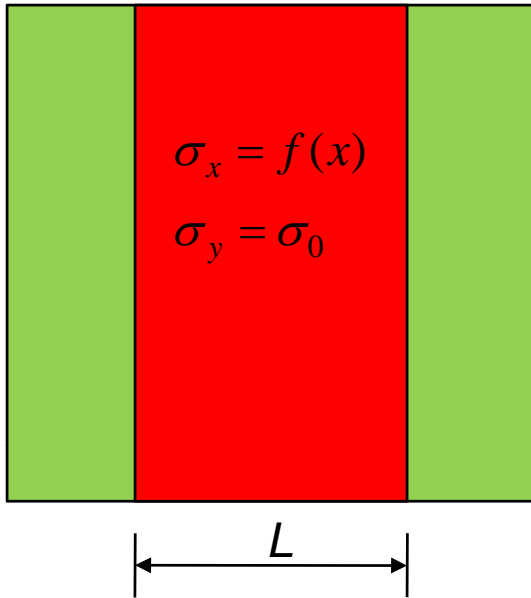


↑ Transition stress:

$$\frac{\sigma_y - \sigma_c}{\sigma_x - \sigma_c} = v_f$$



Wrinkle patterns under non-uniform stresses

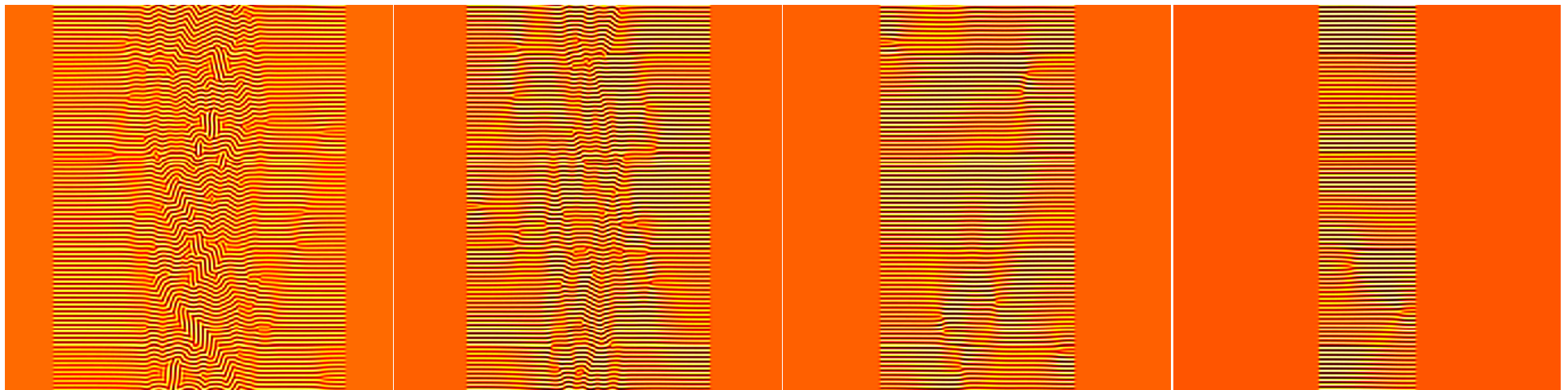
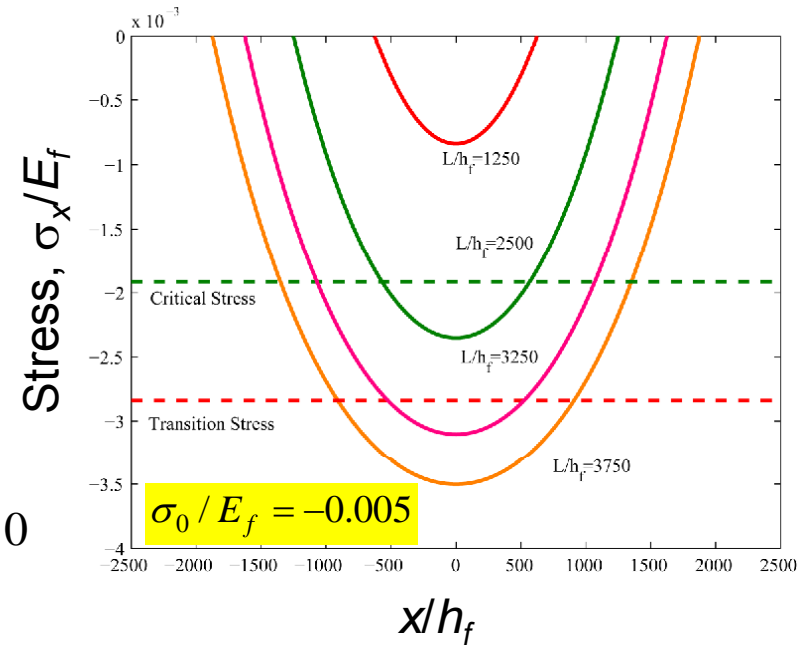


1D shear-lag model:

$$\sigma_x = \sigma_0 \left[1 - \frac{\cosh(x/a)}{\cosh(L/2a)} \right]$$

$$a = \sqrt{\frac{\bar{E}_f h_s h_f}{\mu_R}}$$

outside (green): $\sigma_x = \sigma_y = 0$



$L = 3750 h_f$

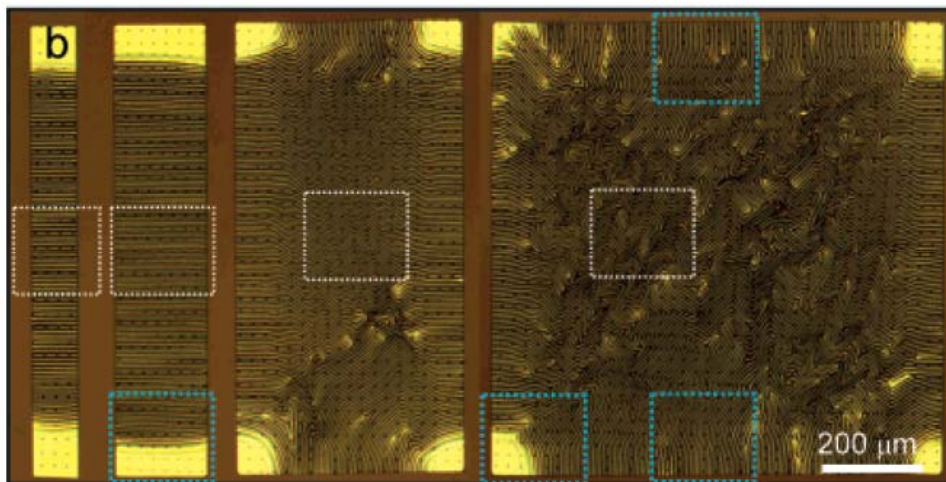
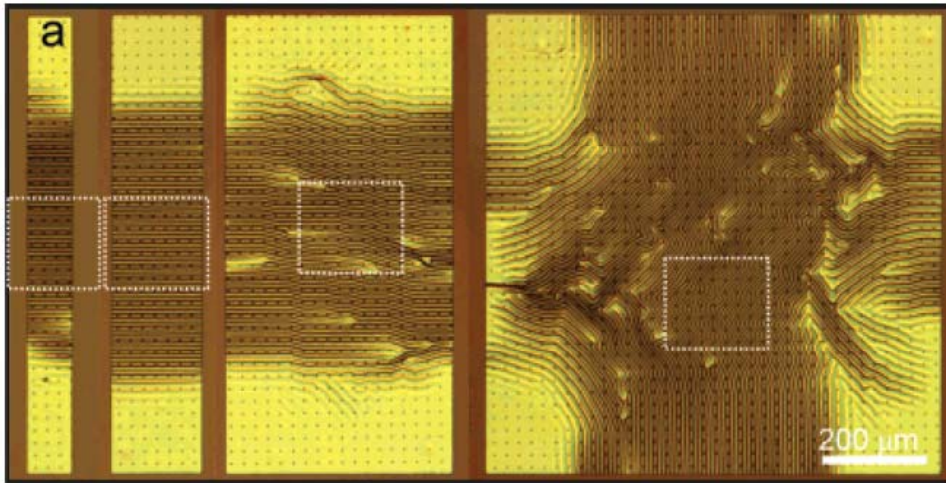
$L = 3250 h_f$

$L = 2500 h_f$

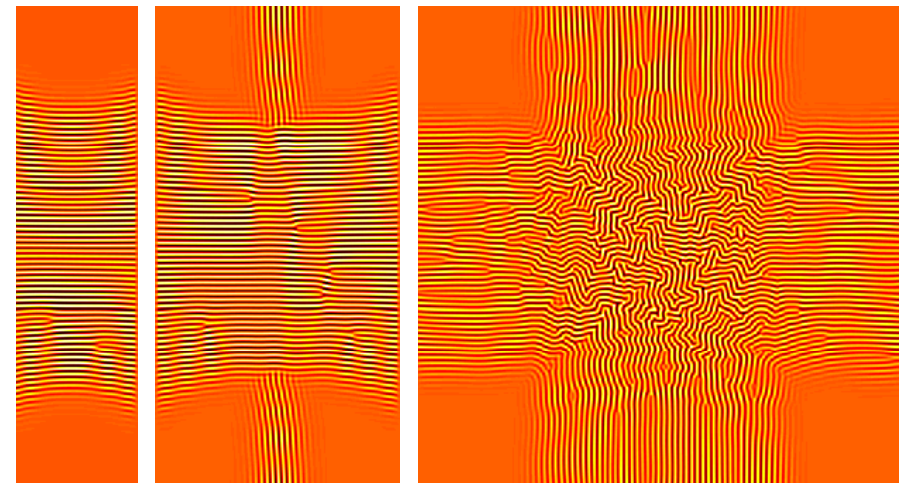
$L = 1250 h_f$

Wrinkling of rectangular films

Experiments (Choi et al., 2007):



Simulations (Im and Huang, 2009):



1250

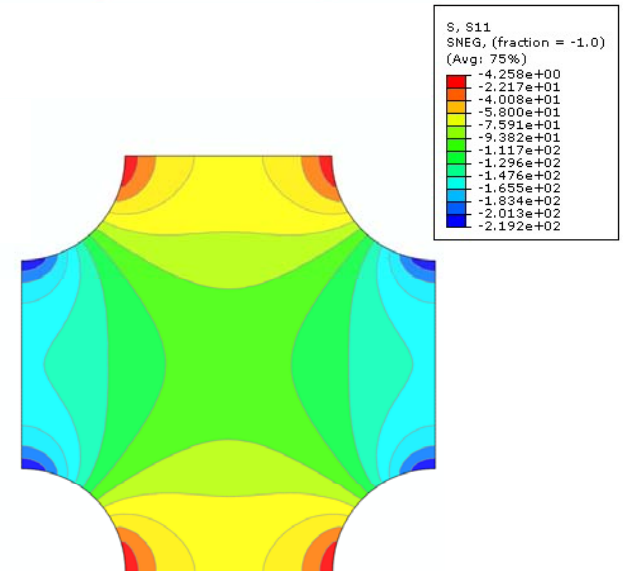
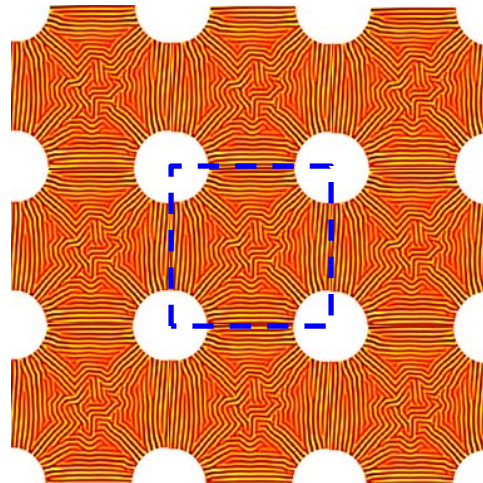
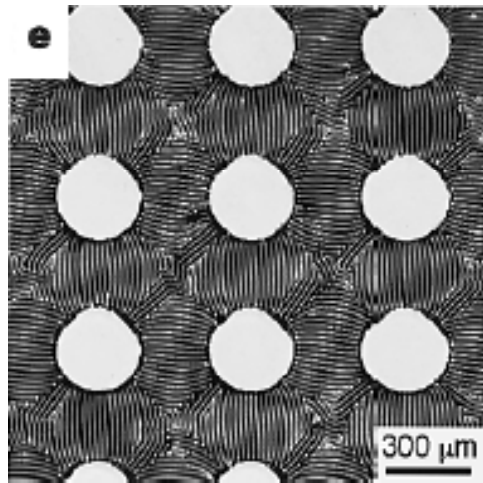
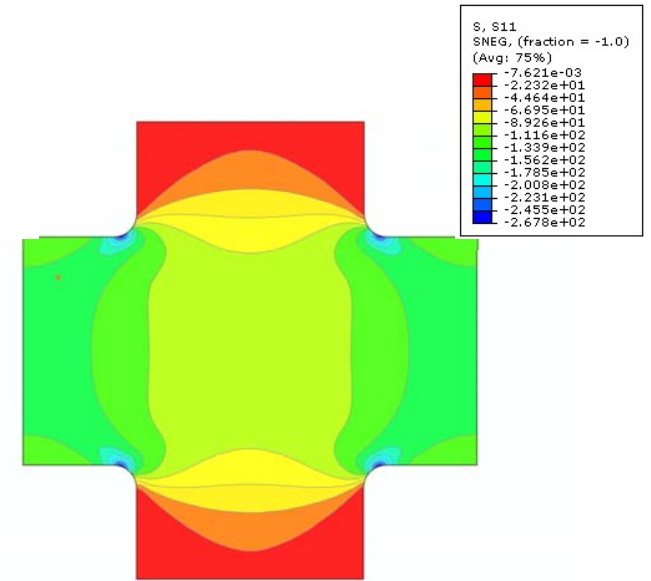
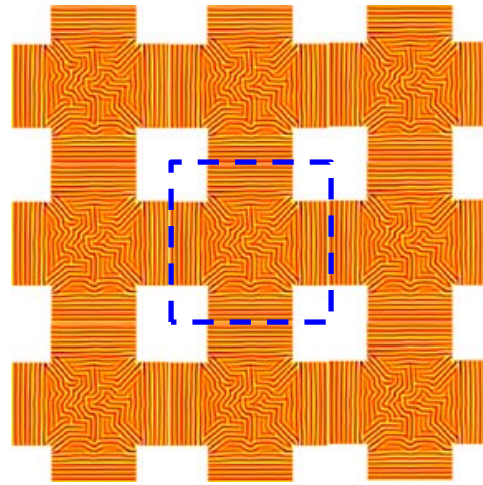
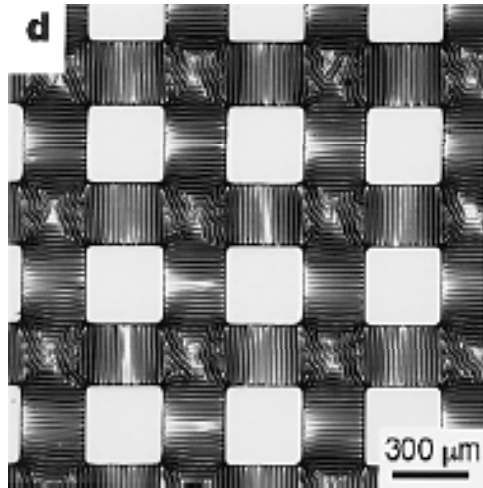
2500

5000

- Stress relaxation in both x and y directions from the edges.
- The length of edge relaxation depends on the elastic modulus of the substrate.
- No wrinkles at the corners, parallel wrinkles along the edges, and transition to zigzag and labyrinth in large films.

Wrinkling of periodically patterned films

Experiments (Bowden et al., 1998):



Simulations (Im and Huang, 2009)

Wrinkling of Single-Crystal Thin Films

30 nm SiGe film on BPSG at 750°C
 (Hobart et al., 2000; Yin et al., 2002 & 2003;
 R.L Peterson, PhD thesis, Princeton, 2006)

100 nm Si on PDMS (Song et al., 2008)

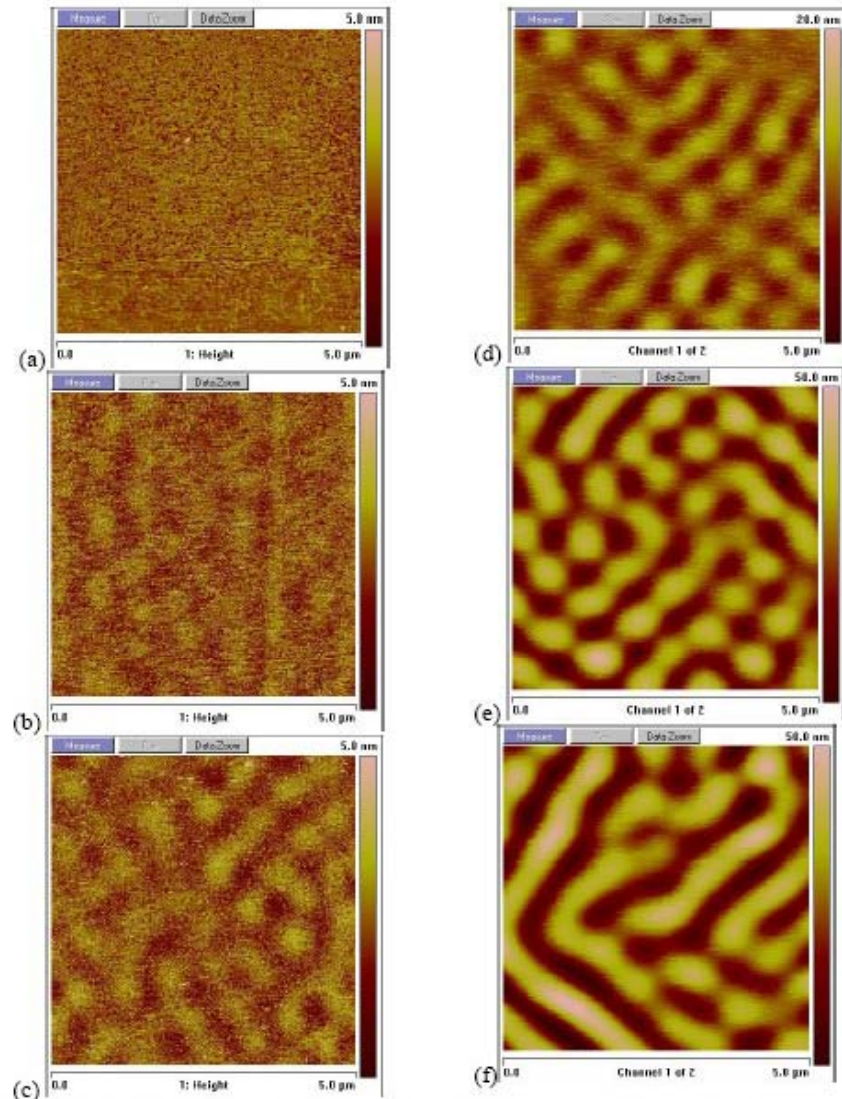
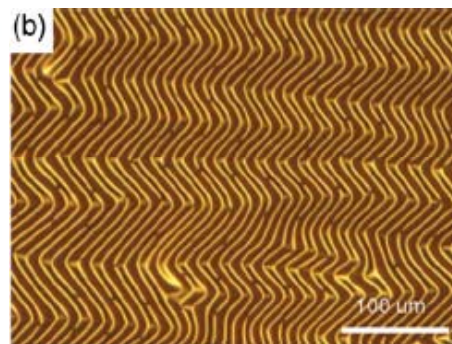
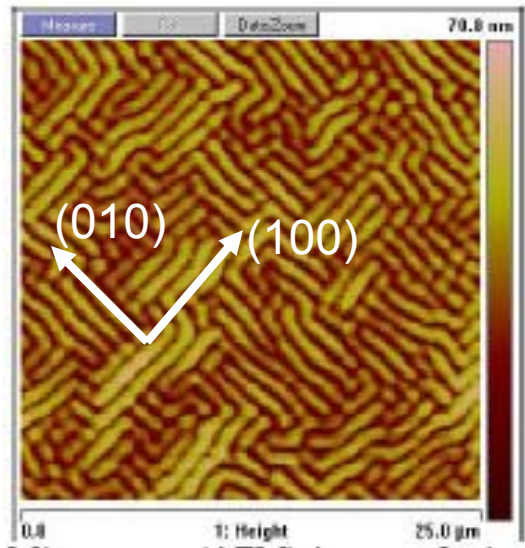
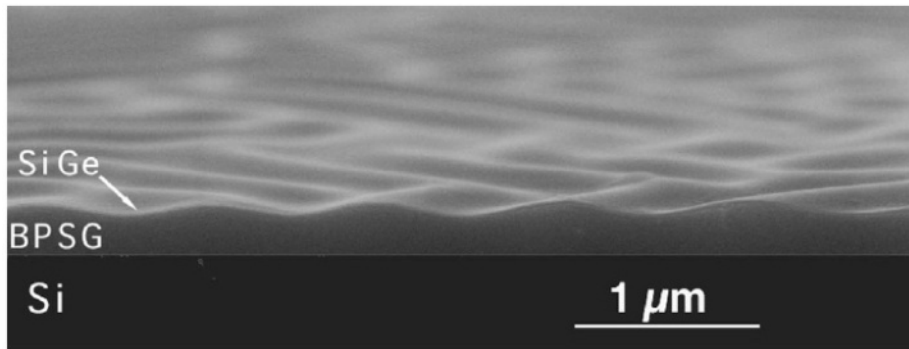
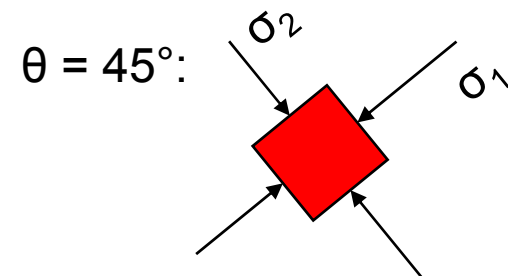
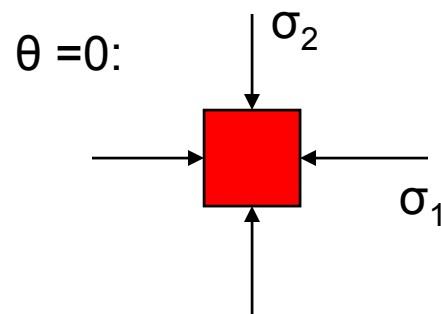
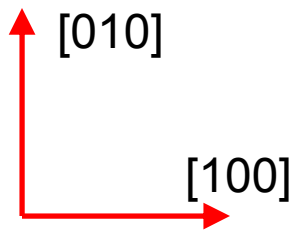
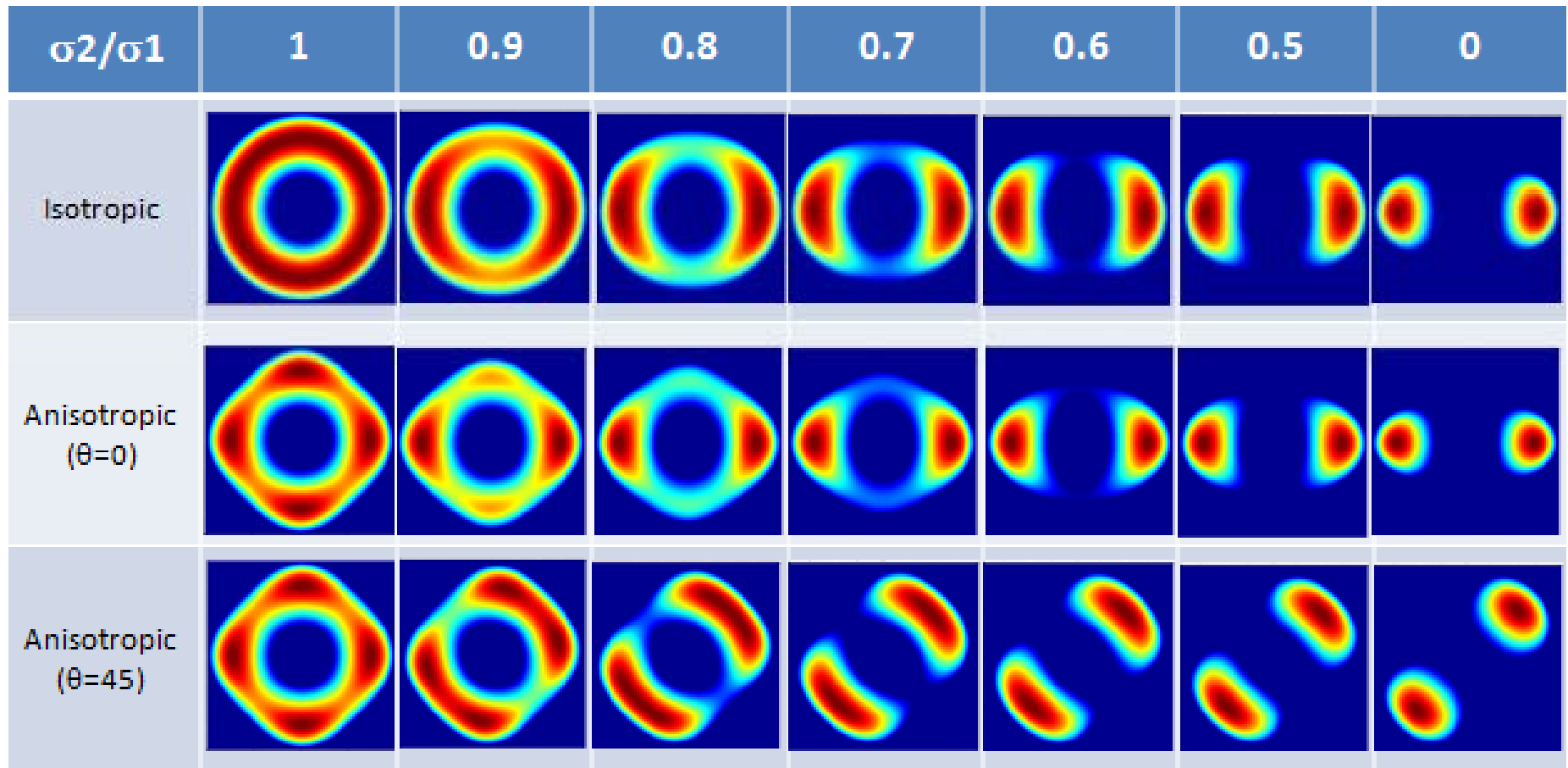


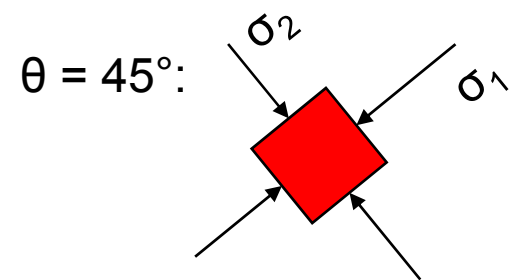
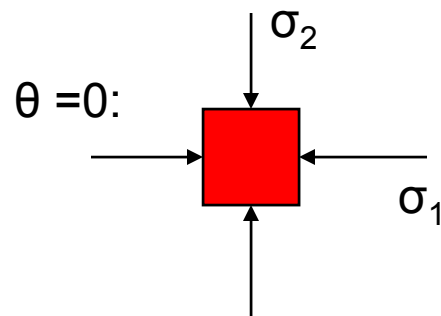
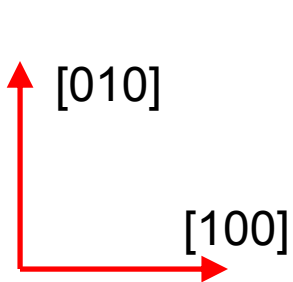
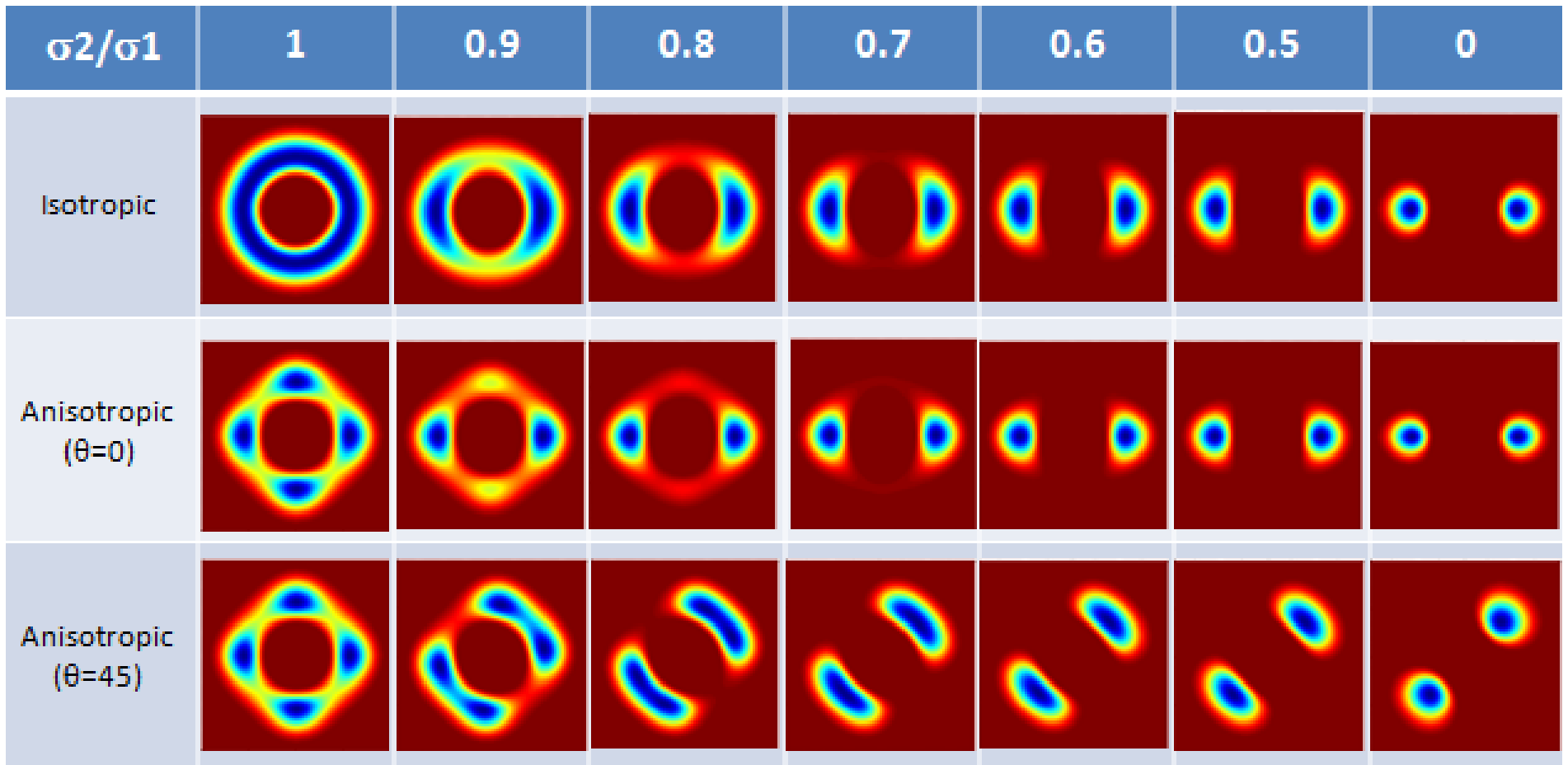
Figure 6.13: AFM images of buckled SiGe on 235-nm BPSG at the center of 150- μm x 150- μm islands after 750°C anneals of various lengths: (a) no anneal, (b) 25 min, (c) 50 min, (d) 105 min, (e) 253 min, and (f) 600 min. The AFM scan size is 5 μm x 5 μm , and the z-axis scale is indicated for each image. The scan edge is along a $\langle 110 \rangle$ crystal direction; buckles are along $\langle 100 \rangle$, as before.

Spectra of initial growth rate



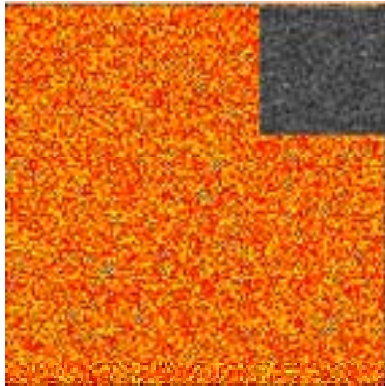
Im and Huang, JMPS 2008.

Energy Spectra of Equilibrium Wrinkles

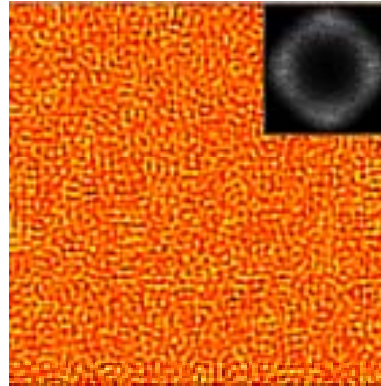


Evolution of orthogonal wrinkle patterns

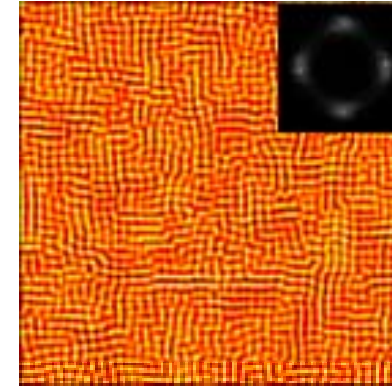
$$\sigma_1 = \sigma_2 = -0.003$$



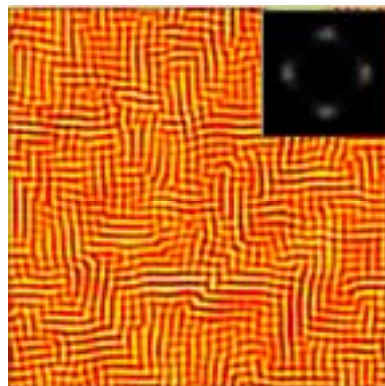
$t = 0:$
RMS = 0.0057
 $\lambda_{\text{ave}} = 38.8$



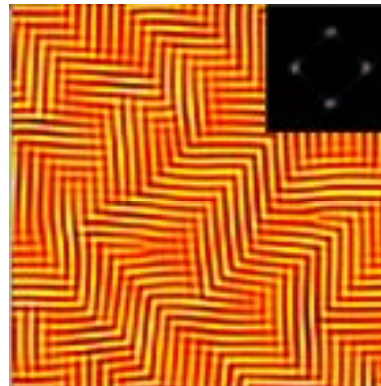
$t = 10^5:$
RMS = 0.0165
 $\lambda_{\text{ave}} = 44.8$



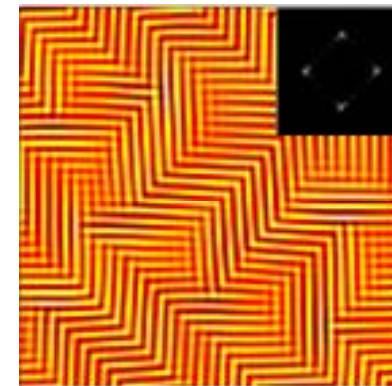
$t = 5 \times 10^5:$
RMS = 0.4185
 $\lambda_{\text{ave}} = 47.1$



$t = 10^6:$
RMS = 0.4676
 $\lambda_{\text{ave}} = 50.8$



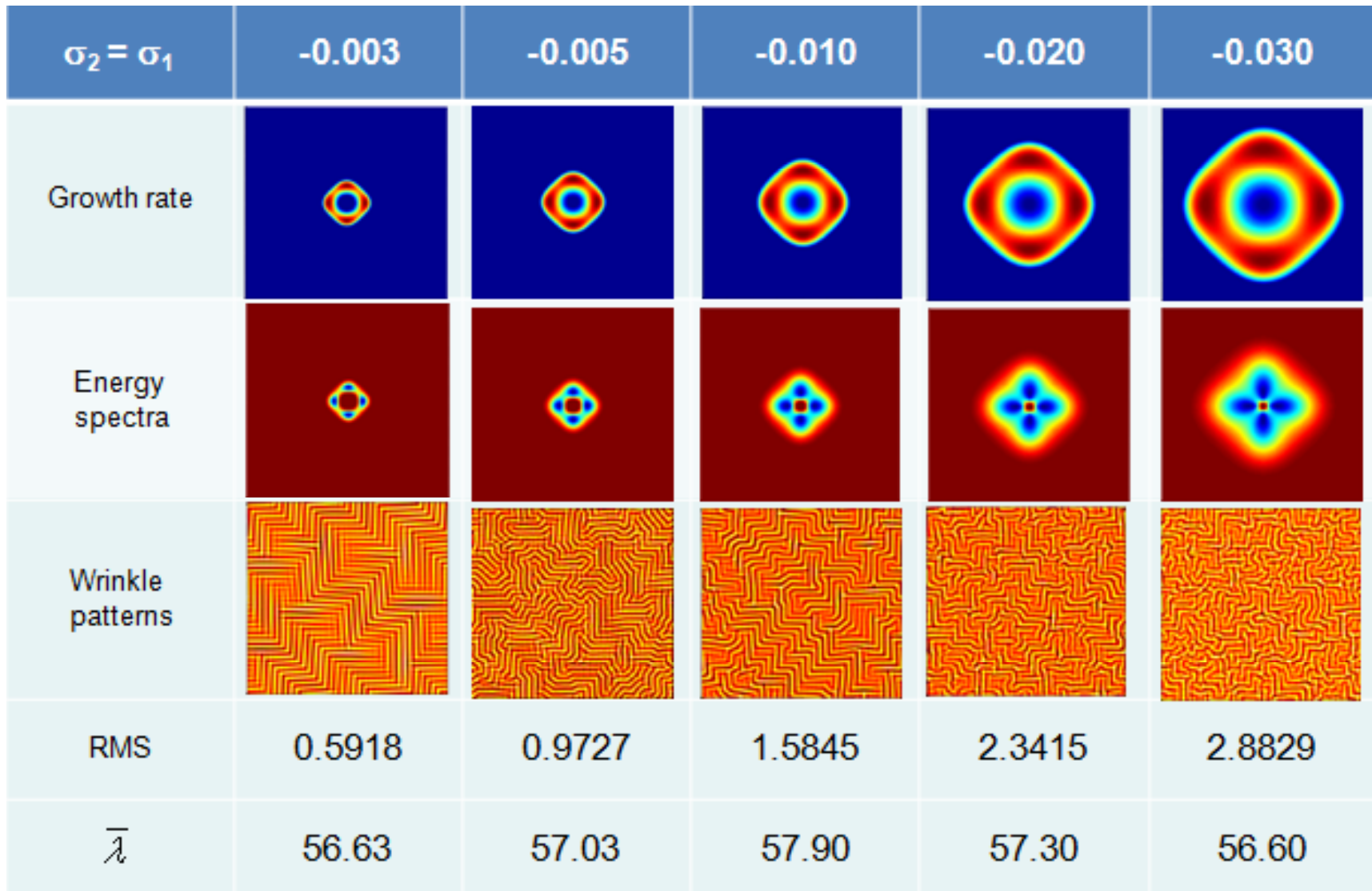
$t = 10^7:$
RMS = 0.5876
 $\lambda_{\text{ave}} = 56.4$



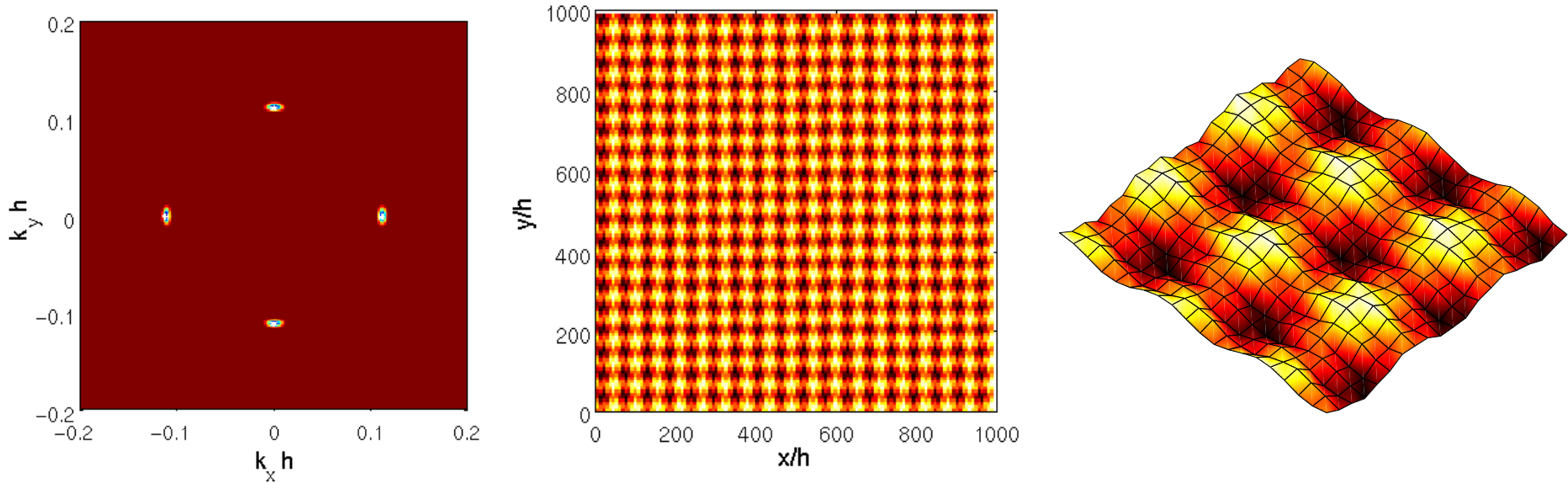
$t = 10^8:$
RMS = 0.5918
 $\lambda_{\text{ave}} = 56.6$

Im and Huang, JMPS 2008.

Effect of stress magnitude



Checkerboard wrinkle pattern

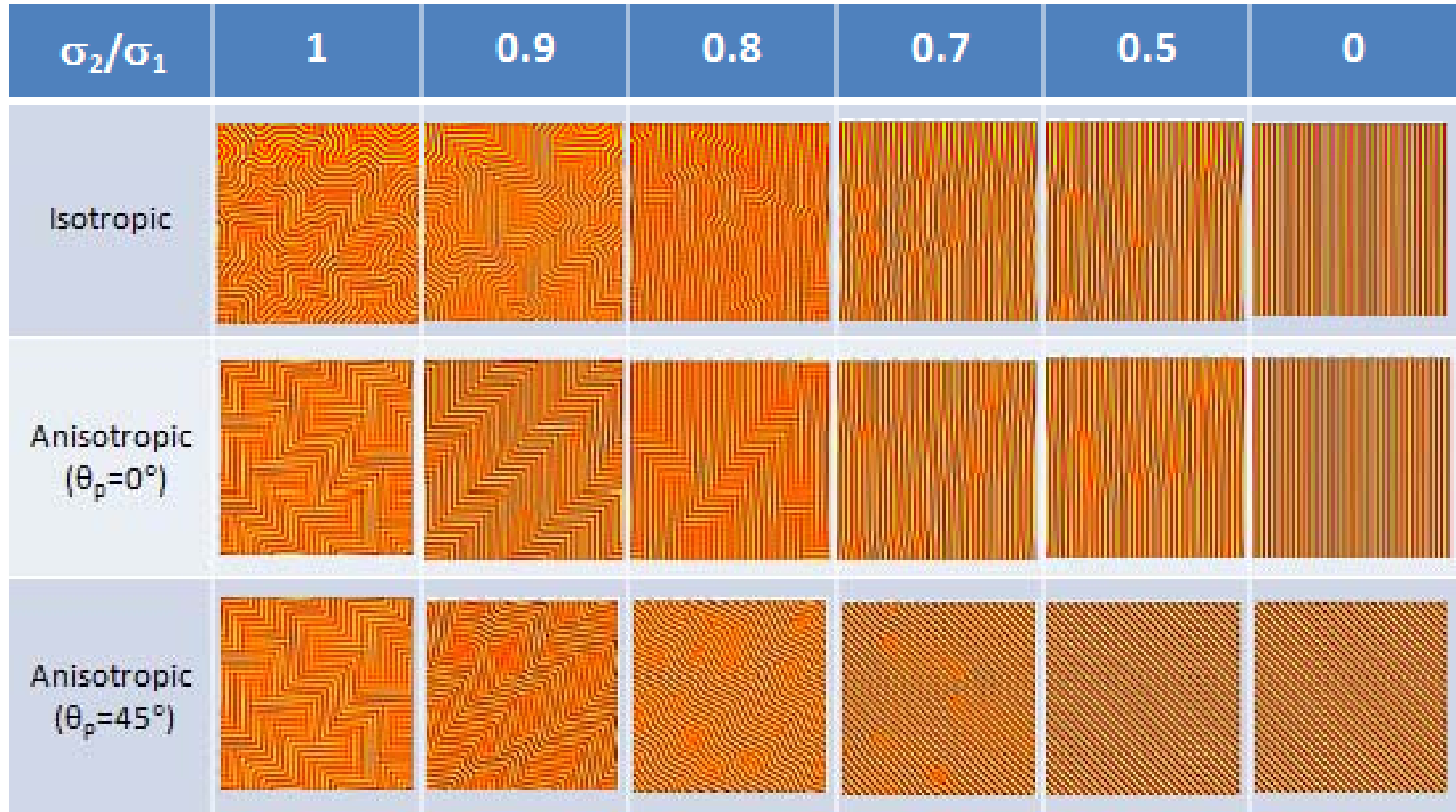


$$\sigma_1 = \sigma_2 = -0.00178$$

$$\text{RMS} = 0.05286 \text{ and } \lambda_{\text{ave}} = 55.6$$

- ❖ Transition of local buckling mode from spherical to cylindrical as the compressive stress/strain increases and/or as the wrinkle amplitude increases.

Transition of wrinkle patterns



Summary

- **Viscoelastic wrinkling**
 - Three-stage evolution
 - Power-law coarsening
 - Near-equilibrium patterns
- **Diverse wrinkle patterns**
 - Anisotropic patterns under biaxial stresses
 - Nonuniform patterns
 - Wrinkling of anisotropic crystal thin films