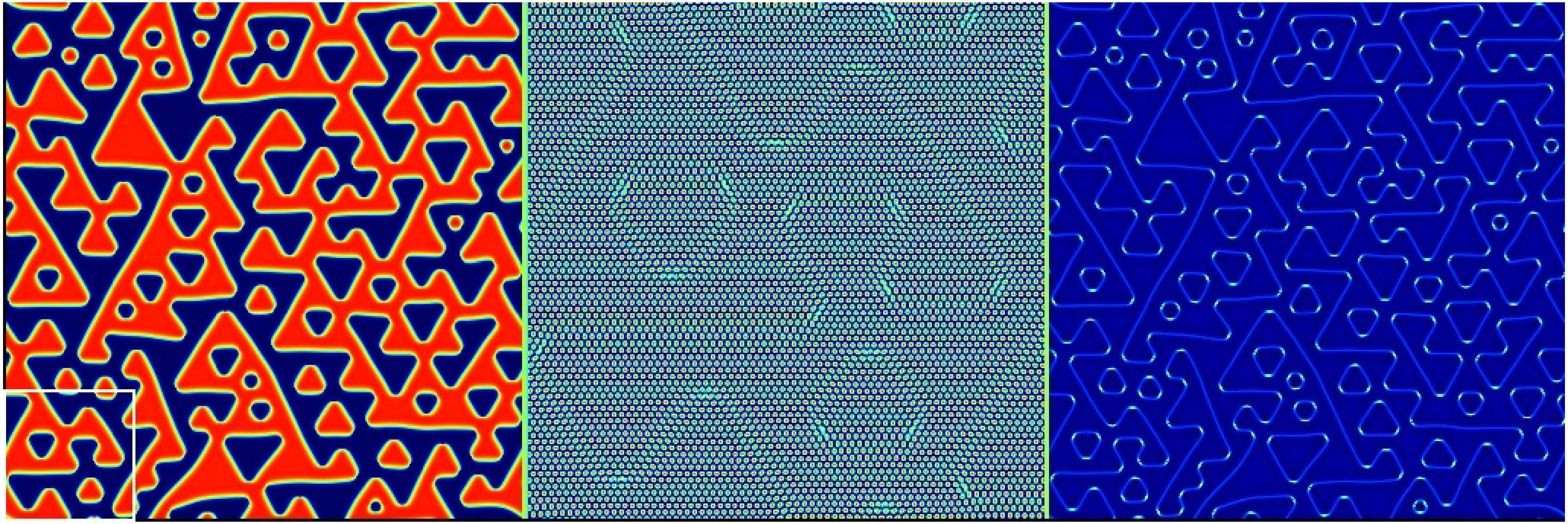
Patterning of Heteroepitaxial Overlayers from Nano to Micron Length Scales

Ken Elder



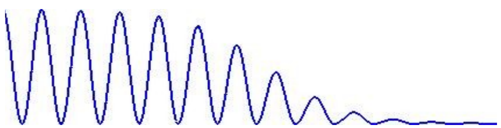
IPAM, October 22nd 2012

Overview

Part 1: Multiscale Modeling

mechanical properties - elasticity, dislocations,
grain boundaries, polycrystals, …

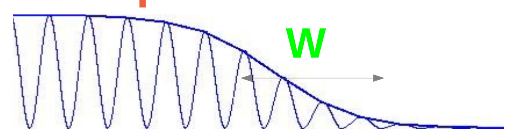
Phase field crystal



$\Delta x \sim a/10$

~0.1 Å

Amplitude



$\Delta x \sim W/10$

~1 Å

Continuum

$\Delta x \sim W/10$

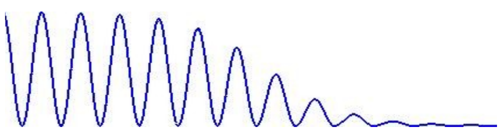
~1 Å

Overview

Part 1: Multiscale Modeling

mechanical properties - elasticity, dislocations, grain boundaries, polycrystals, …

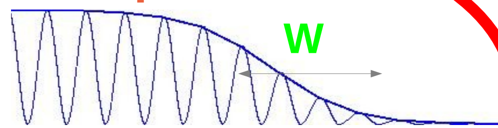
Phase field crystal



$\Delta x \sim a/10$

~0.1 Å

Amplitude



$\Delta x \sim W/10$

~1 Å

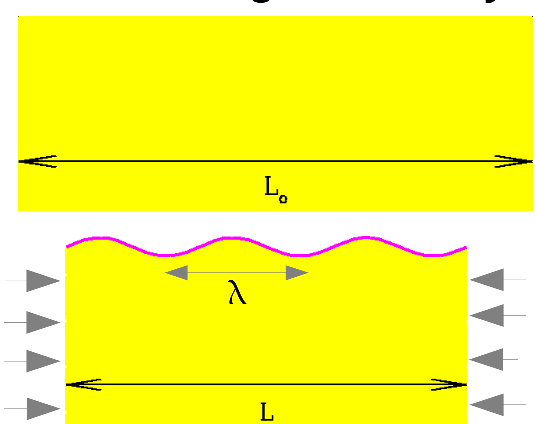
Continuum

$\Delta x \sim W/10$

~1 Å

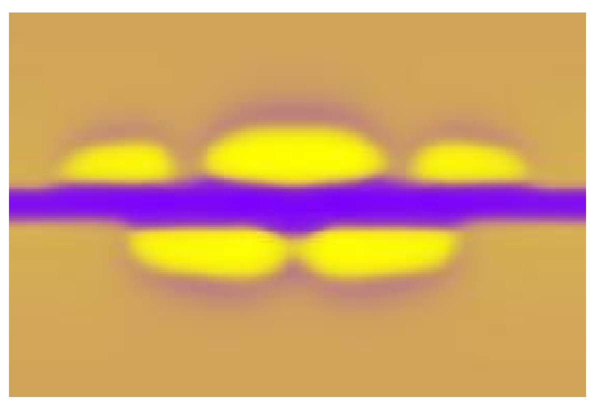
Part 2: Applications

Mounding instability



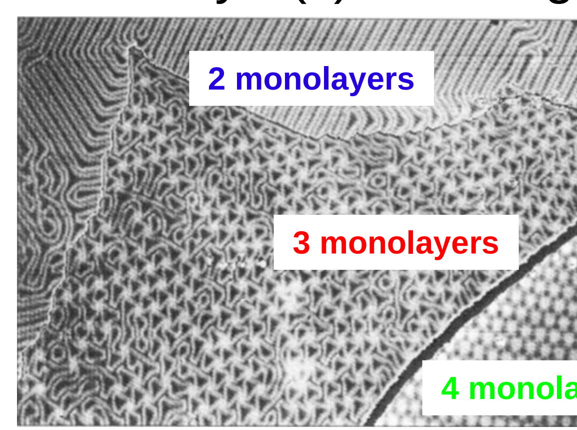
Wavelength Selection

Island formation on nano-membranes



Long range order?

Monolayer(s) Ordering



Pattern + Length Selection

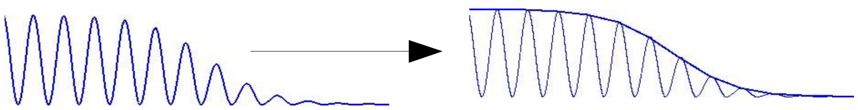
Oakland University

Collaborators

Multiscale modeling: Amplitude expansions

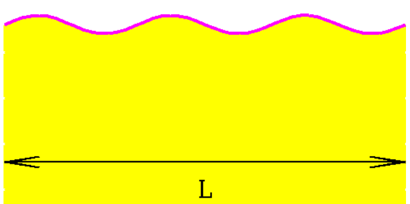
Zhi-Feng Huang[1], Nik Provatas[2]

Dong-Hee Yeon[3], Katsuyo Thornton[3]



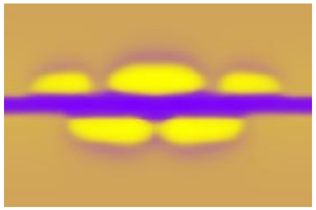
Applications:

mounding instabilities



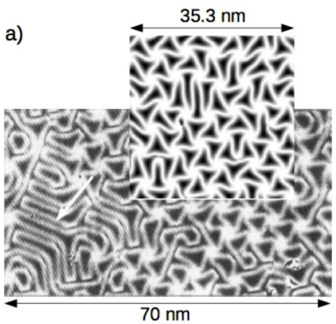
Zhi-Feng Huang[1]

quantum dots on nanomembranes



Zhi-Feng Huang[1]

Heteroepitaxial monolayers(s)



Giula Rossi[4], Pekka Kanerva[4], Fabio Sanches[5], See-Chen Ying[6], Enzo Granato[7], Christian Achim[4], Tapio Ala-Nissila[4,6]

1 Wayne State

2 McGill

3 Michigan

4 Aalto

5 Oakland

6 Brown

7 INPE

Funding

NSF

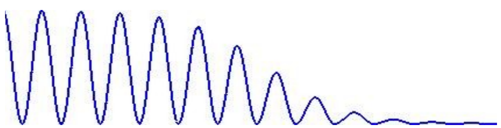
Division of Materials Research

Overview

Part 1: Multiscale modeling

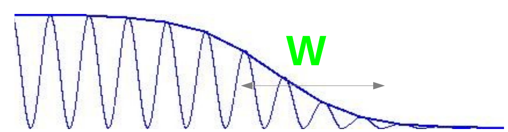
mechanical properties - elasticity, dislocations,
grain boundaries, polycrystals, …

Phase field crystal



Δx ~ a/10 **~0.1 Å**

Amplitude



Δx ~ W/10 **~1 Å**

Continuum

Δx ~ W/10 **~1 Å**

References

pure systems
Goldenfeld, Athreya, Dantzig, PRE (2005)
Athreya, Goldenfeld, Dantzig, PRE (2006)
Athreya, Goldenfeld, Dantzig, Greenwood, Provatas PRE (2007)
Chan, Goldenfeld PRE (2009)
Yeon, Huang, Elder, Thornton, Phil Mag (2010)
binary systems
Elder, Huang, Provatas PRE (2010)
Huang, Elder, Provatas PRE (2010)
Spatschek, Karma PRB (2010)

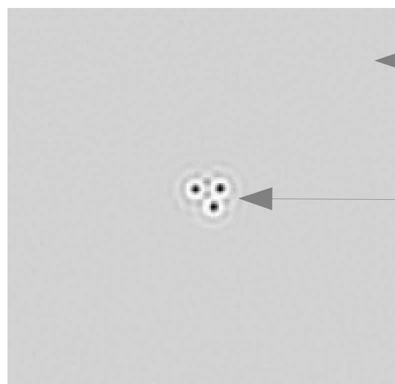
Part 1: Multiscale Modeling: PFC

$$\frac{\Delta \tilde{F}}{k_b T V \bar{\rho}} \approx \frac{R^d}{V} \int d\vec{r} \left[\frac{n}{2} \left(B^l + B^x \left(2\nabla^2 + \nabla^4 \right) \right) n - \frac{n^3}{6} + \frac{n^4}{12} \right]$$

Dissipative dynamics

$$\frac{\partial n}{\partial t} = \Gamma \nabla^2 \frac{\delta F}{\delta n} = \Gamma \nabla^2 \left[B^l + B^x (2\nabla^2 + \nabla^4) n - \frac{n^2}{2} + \frac{n^3}{3} \right]$$

Example – growth of a solid drop in supercooled liquid



supercooled liquid
n = constant

crystal seed
n = varying ~ lattice constant

Time Scales/computational speed

$$\frac{\text{vacancy diffusion time}}{\text{numerical time step}} \sim 10-100$$

Comparison: Molecular Dynamics Gold T = 800^{o}C

$$\frac{\text{vacancy diffusion time}}{\text{numerical time step}} \sim 10^{11}$$

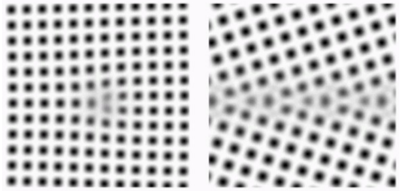
$$\frac{\Delta t_{MD}}{\Delta t_{PFC}} \sim 10^9$$

~ Billion times faster!

Part 1: Multiscale Modeling: PFC applications

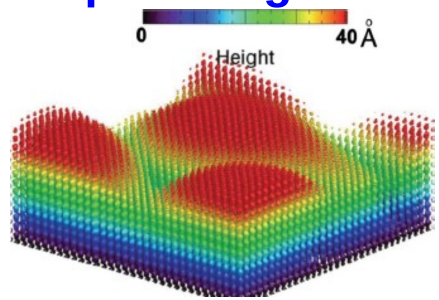
Elder Katakowski Haataja Grant PRL (2002) , Elder Grant PRE (2004)

Grain boundaries – energy, premelting



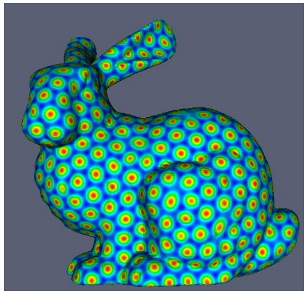
Berry Elder Grant PRB (2008),
Mellenthin Karma Plapp RPB (2008)
Jaatinen Achim Elder Ala-Nissila PRE (2009)

Strained films -epitaxial growth



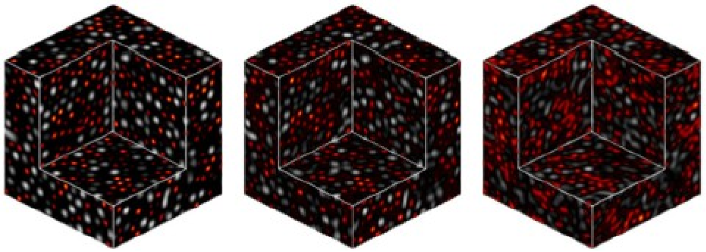
Huang Elder PRL (2008), PRB (2010)
Wu Voorhees PRB (2009)

Surface ordering



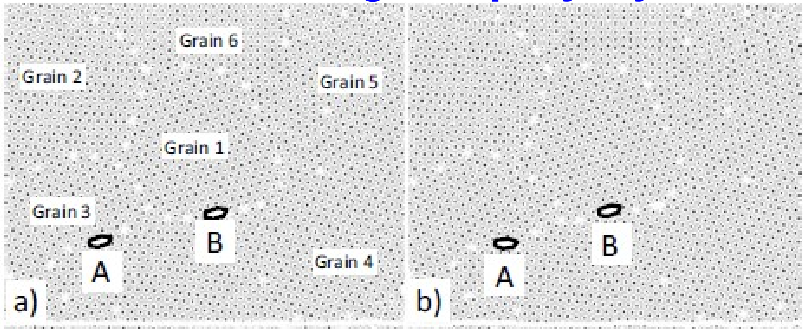
Achim Karttunen Elder Granato Ala-Nissila Ying PRE (2006)
Ramos Granato Achim Ying Elder Ala-Nissila PRE (2008), PRE (2009)
Backofen Voigt Witkowski PRE (2010)
Muralidharan Haataja PRL (2010)

Glass formation



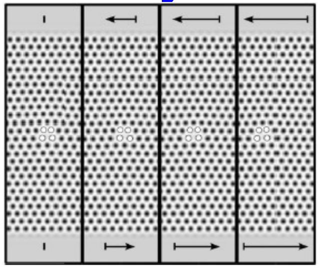
Berry Elder Grant PRE (2008), Berry Grant PRL (2011)

Material strength ~ polycrystals



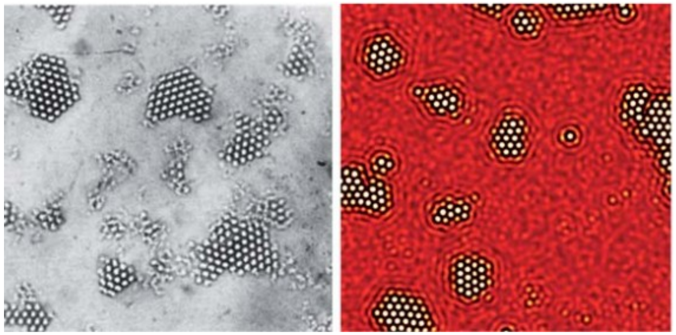
Hirouchi, Takaki, Tomita Comput Mater Sci (2009)
Stefanovic, Haataja, Provatas PRL (2006), PRE (2009)

Dislocation dynamics - plasticity



Berry Grant Elder PRE (2006)
Chan Tsekenis Dantzig
Dahmen Goldenfeld PRL (2010)

Solidification/nucleation/faceting



Backofen Ratz Voigt Phil Mag (2007)
Prieler Hubert Verleye Haberkern Emmerich J. Phys. Cond Mat (2009)
Galenko Danilov Lebedev PRE (2009)
Tegze Granasy Toth Douglas Pusztai Soft Matter (2010)
Tegze Toth Granasy PRL (2011),
Granasy Tegze Toth Pusztai (2011)

Part 1: Multiscale Modeling: PFC applications

Elder Katakowski Haataja Grant PRL (2002) , Elder Grant PRE (2004)

Glass formation

Angel Sci. (1995)

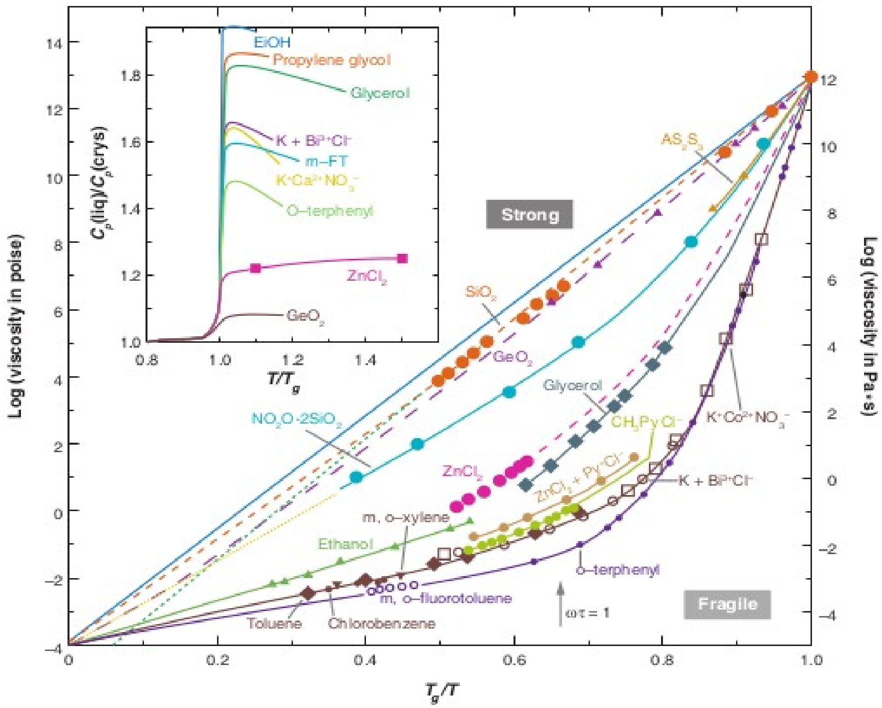
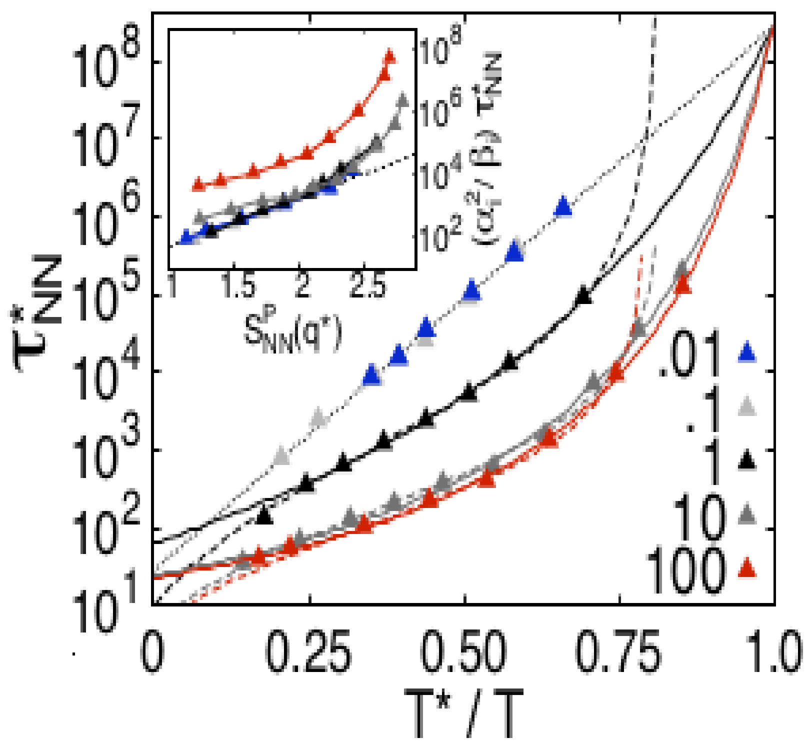
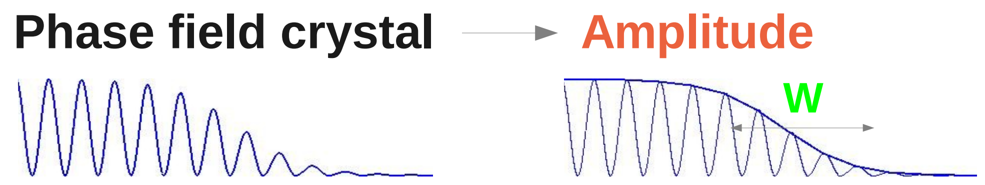


Figure 1.2: Experimental viscosity of glass forming liquids versus inverse temperature, from [1]. Time scales vary from $\sim 1ps$ at the lowest viscosities to $\sim 10^4 s$ at T_g. The inset shows the ratio of liquid and crystal specific heats.

Berry Grant PRL (2011)



PFC to Amplitude expansions



PFC free equation of motion

$$\frac{\partial n}{\partial t} = \Gamma \nabla^2 \left[\left(\Delta B + B^x \left(1 + \nabla^2\right)^2 \right) n - t\, n^2 + v\, n^3 \right]$$

Amplitude formulation:

$$n = \sum_{\vec{G}} \left(\eta_{\vec{G}}\, e^{i\vec{G}\cdot\vec{r}} + \eta^*_{\vec{G}}\, e^{-i\vec{G}\cdot\vec{r}} \right)$$

$$\vec{G} \equiv l\vec{q}_1 + m\vec{q}_2 + n\vec{q}_3$$

$$\left(\vec{q}_1, \vec{q}_2, \vec{q}_3\right) \equiv \text{principle reciprocal lattice vectors}$$

$$\left(l, m, n\right) \equiv \text{Miller indices}$$

Goal – derive $\dfrac{\partial \eta_{\vec{G}}}{\partial t} = \,?$

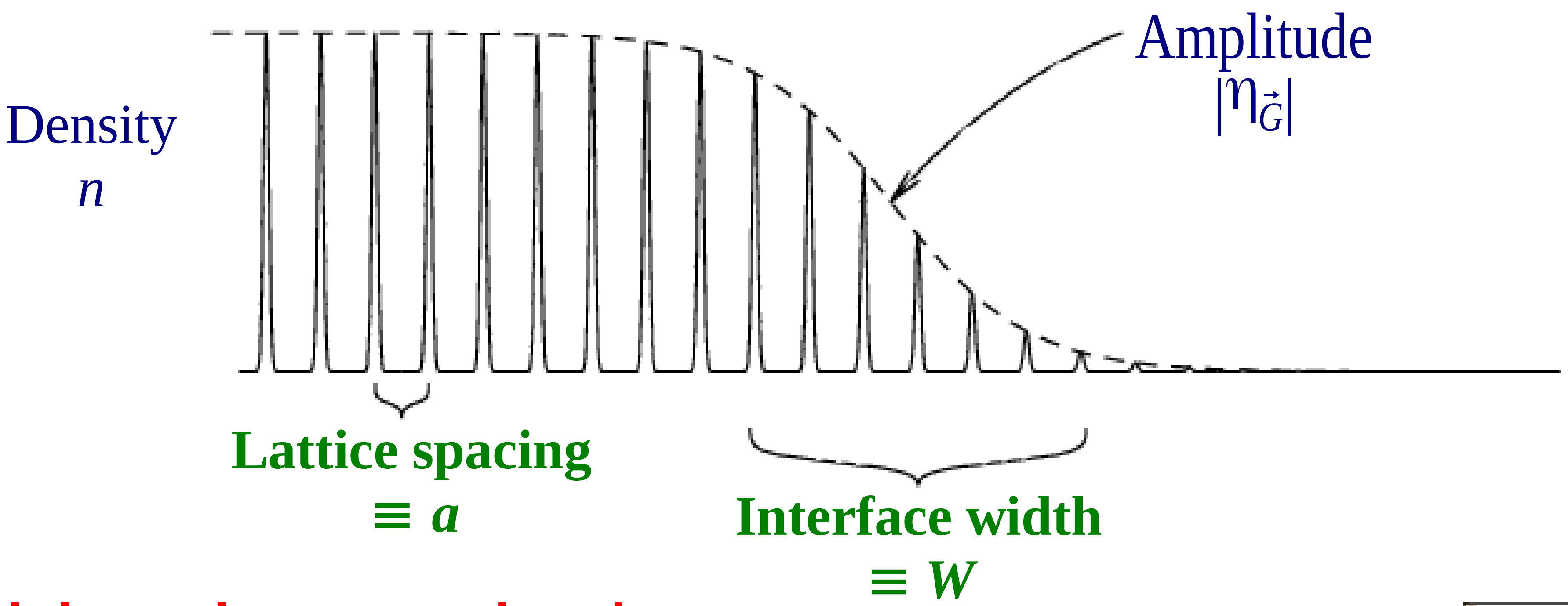
* Amplitude expansion: multiple scales approximation

$$n = \sum_{\vec{G}} \left(\eta_{\vec{G}}\, e^{i\vec{G}\cdot\vec{r}} + \eta^*_{\vec{G}}\, e^{-i\vec{G}\cdot\vec{r}} \right)$$

$$\frac{\partial \eta_{\vec{G}}}{\partial t} = ?$$

2 fields *(n, η_G)* – **2 length scales (*a,W)***

Consider schematic of liquid/solid interface



Multiple scales approximation

$W \gg a$ → Phase field limit

* Amplitude expansion: multiple scales approximation

$$n = \sum_{\vec{G}} \left(\eta_{\vec{G}}\, e^{i\vec{G}\cdot\vec{r}} + \eta^{*}_{\vec{G}}\, e^{-i\vec{G}\cdot\vec{r}} \right)$$

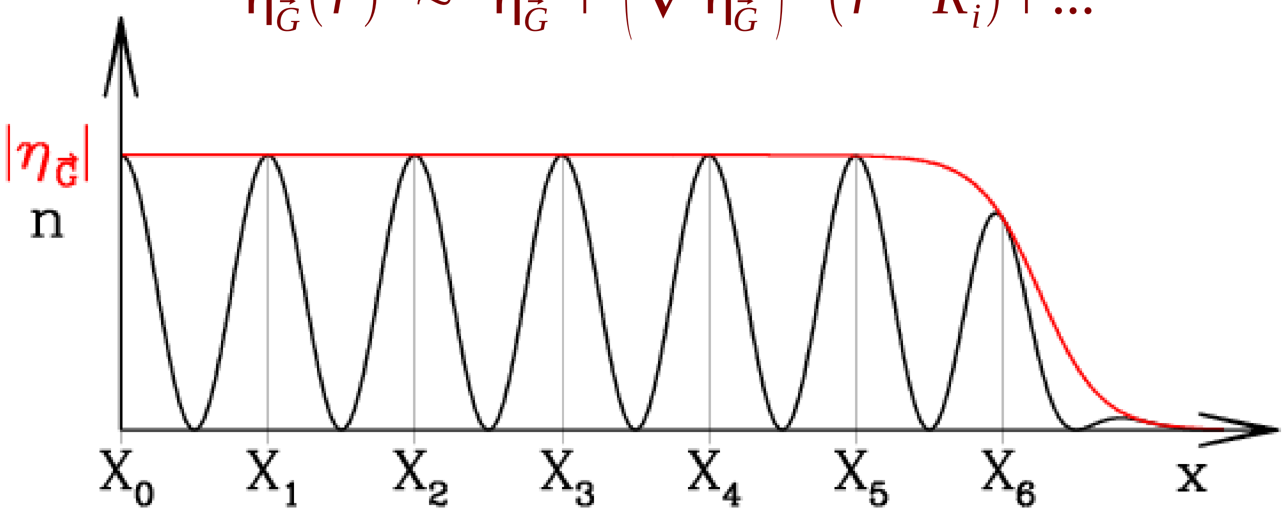
$$\frac{\partial \eta_{\vec{G}}}{\partial t} = ?$$

Multiple scales approximation

$W \gg a$ → Phase field limit

Expand amplitudes (η) around lattice sites, i.e.,

$$\eta_{\vec{G}}(\vec{r}) \approx \eta^{i}_{\vec{G}} + \left(\vec{\nabla} \eta^{i}_{\vec{G}} \right) \cdot (\vec{r} - \vec{R}_i) + \ldots$$



* Amplitude expansion: multiple scales approximation

$$n = \sum_{\vec{G}} \left(\eta_{\vec{G}} e^{i\vec{G}\cdot\vec{r}} + \eta^*_{\vec{G}} e^{-i\vec{G}\cdot\vec{r}} \right)$$

$$\frac{\partial \eta_{\vec{G}}}{\partial t} = ?$$

Expand amplitudes (η) around lattice sites, i.e.,

$$\eta_{\vec{G}}(\vec{r}) \approx \eta^i_{\vec{G}} + \left(\vec{\nabla}\eta^i_{\vec{G}}\right)\cdot(\vec{r}-\vec{R}_i)+\ldots$$

Multiply equation of motion by $e^{-i\vec{Q}_{\vec{G}}\cdot\vec{r}}$ and average over one unit cell eg., in one dimension

$$\frac{1}{a}\int_{x_i-a/2}^{x_i+a/2} dx\, e^{-iQ_G x}\left[\frac{\partial n}{\partial t} = \Gamma\nabla^2\left[\left(\Delta B + B^x\left(1+\nabla^2\right)^2\right)n - t\,n^2 + v\,n^3\right]\right]$$

Eg, left hand side

$$\frac{1}{a}\int_{x_i-a/2}^{x_i+a/2} dx\, e^{-iQx}\left[\frac{\partial}{\partial t}\left(\eta^i + \left(\vec{\nabla}\eta^i\right)\cdot(\vec{r}-\vec{R}_i)+\ldots\right)e^{iQx}\right]$$

$$\approx \frac{\partial \eta^i}{\partial t}\left(\frac{1}{a}\int_{x_i-a/2}^{x_i+a/2} dx\right) + \frac{\partial \vec{\nabla}\cdot\eta^i}{\partial t}\left(\frac{1}{a}\int_{x_i-a/2}^{x_i+a/2} dx\,(x-x_i)\right) + \ldots$$

Oakland UNIVERSITY

* Amplitude expansion: Two dimensions to lowest order

2d: triangular lattice, principle reciprocal lattice vectors ¿

$$\vec{q}_1 = -\frac{1}{2}\left(\sqrt{3}\,\hat{x} + \hat{y}\right)\ ;\ \vec{q}_2 = \hat{y}$$

$$\frac{\partial \eta_j}{\partial t} = \Im_j \frac{\delta F_{2d}}{\delta \eta_j^*} \approx -\left[\left(\Delta B + B^x \Im_j^2 + 3v\left(A^2 - |\eta_j|^2\right)\right)\eta_j - 2t\prod_{i\neq j}\eta_i^*\right]$$

$$F_{2d} = \int d\vec{r}\left[\frac{\Delta B}{2}A^2 + \frac{3v}{4}A^4 + \sum_{j=1}^{3}\left\{B^x|\Im_j \eta_j|^2 - \frac{3v}{2}|\eta_j|^4\right\} - 2t\left\{\prod_{j=1}^{3}\eta_j + c.c.\right\}\right]$$

where $A^2 \equiv 2\sum|\eta_j|^2$, $\Im_j \equiv \nabla^2 + 2i\,\vec{q}_j\cdot\vec{\nabla}$

→ **Now 6 equations (3 complex)**

→ **Still includes elasticity, dislocations, multiple crystal orientations**

* Amplitude expansion: applications

Polycrystals – (Φ,u) – adaptive mesh refinement

Athreya, Goldenfeld, Dantzig, Greenwood, Provatas, PRE **76**, 056706 (2007)

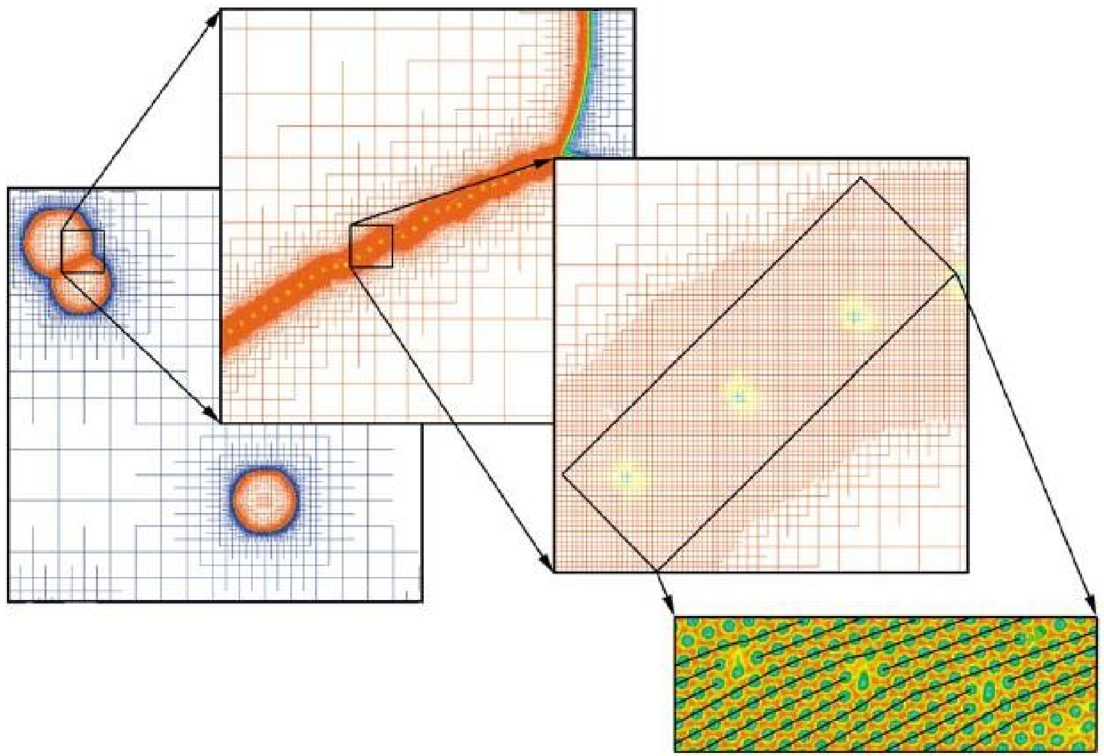


FIG. 12. (Color online) The above grid spans roughly three orders of magnitude in length scales, from a nanometer up to a micrometer. The leftmost box resolves the entire computational domain whereas the rightmost resolves dislocations at the atomic scale.

* Continuum limit of amplitude equations

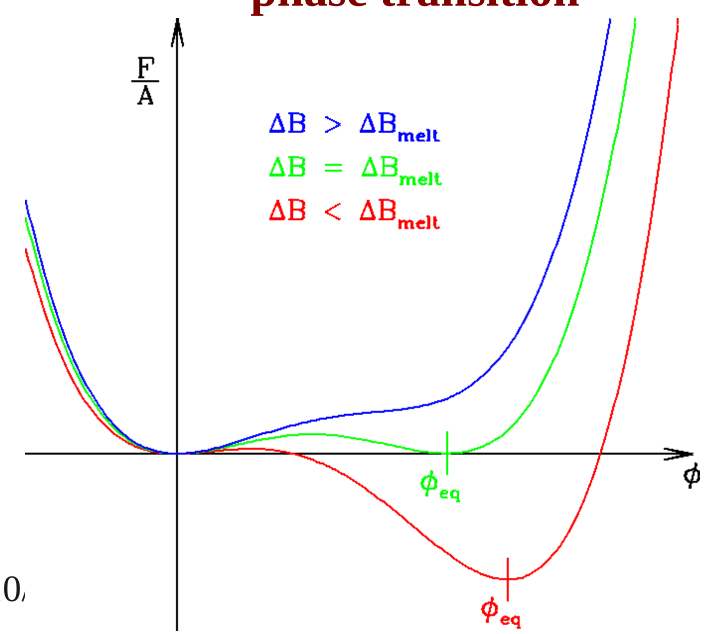
Limiting case: $\eta_j = \phi \, e^{i\vec{G}_j \cdot \vec{u}}$, where, $\vec{u}$ = displacement field

Small deformation limit

$\phi \rightarrow 1^{st}$ order liquid/solid transition, $\vec{u} \rightarrow$ continuum elasticity theory ¿

$$F_{2d} = \int d\vec{r} \left[3\Delta B \phi^2 - 4t\phi^3 + \frac{45}{2}\nu\phi^4 + 6B^x \left|\vec{\nabla}\phi\right|^2 + 3B^x\phi^2 \left\{ \frac{3}{2}\sum_{i=1}^{2} U_{ii}^2 + U_{xx}U_{yy} + 2U_{xy}^2 \right\} \right]$$

1st order Liquid/Solid phase transition — **surface energy** — **continuum elastic energy**



Where $U_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} + \frac{1}{2}\frac{\partial u_k}{\partial x_i}\frac{\partial u_k}{\partial x_j} \right)$

elastic constants

$$C_{11} = 9B^x\phi^2$$

$$C_{12} = C_{44} = C_{11}/3$$

Again note:

liquid: $\phi = 0$, elastic energy $= 0$

solid: $\phi \neq 0$, elastic energy > 0

* Continuum limit of amplitude equations

Limiting case: $\eta_j = \phi$ no deformations

$$F_{2d} = \int d\vec{r} \left[3\Delta B \phi^2 - 4t\phi^3 + \frac{45}{2} v\phi^4 + 6B^x \left|\vec{\nabla}\phi\right|^2 \right]$$

Dynamics

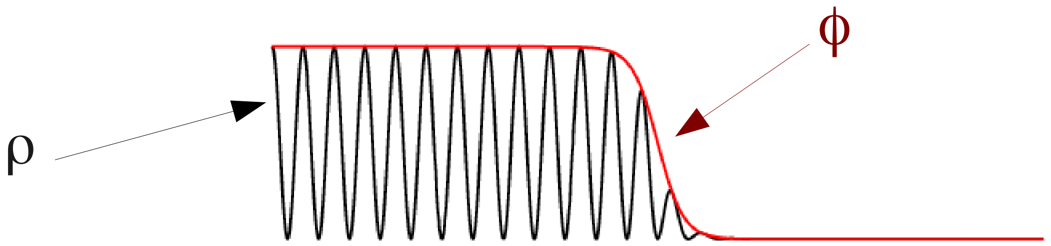
$$\frac{\partial \phi}{\partial t} = -\frac{M}{6}\frac{\delta F}{\delta \phi}$$

$$= M\left[2B^x \nabla^2 \phi - \Delta B \phi + 2t\phi^2 - 15 v \phi^3\right]$$

$\sim$ Model A

Phase field model of liquid/solid transition

order parameter - ϕ - amplitude of density fluctuations



* Continuum limit of amplitude equations

Repeat for binary alloy model:

- substitutional binary alloy A and B atoms, densities ρ_A and ρ_B define two fields,

$$\psi = 2c - 1\,,\; n = (\rho - \rho_l)/\rho_l, \text{ where } \rho \equiv \rho_A + \rho_B, \quad c \equiv \rho_A/\rho$$

- free energy (see Elder, Provatas, Berry, Stefanovic, Grant, PRB **75**, 064107 (2007))

$$\frac{\Delta F}{k_B T \rho_l} = \int d\vec{r} \left[\underbrace{\frac{B^l}{2} n^2 + B^x \frac{n}{2}\left(2R^2\nabla^2 + R^4\nabla^4\right) n - \frac{t}{3} n^3 + \frac{v}{4} n^4}_{\text{usual PFC model}} + \underbrace{\frac{\omega}{2}\psi^2 + \frac{u}{4}\psi^4 + \frac{K}{2}\left|\vec{\nabla}\psi\right|^2}_{\text{Model B/Cahn Hilliard}} \right]$$

$$\begin{aligned} \text{where } B^l &= B^l_0 + B^l_1\psi + B^l_2\psi^2 + \ldots &&\rightarrow \text{ eutectics phase diagrams etc.} \\ B^x &= B^x_0 + B^x_1\psi + B^x_2\psi^2 + \ldots &&\rightarrow \text{ elastic moduli} \sim \text{function of } \psi \\ R &= R_0 + R_1\psi + R_2\psi^2 + \ldots &&\rightarrow \text{ lattice constant} \sim \text{function of } \psi \end{aligned}$$

- dynamics, for mobilities M_A and M_B

$$\frac{\partial n}{\partial t} = M_1\nabla^2\frac{\delta F}{\delta n} + M_2\nabla^2\frac{\delta F}{\delta \psi}$$

$$\frac{\partial \psi}{\partial t} = M_2\nabla^2\frac{\delta F}{\delta n} + M_1\nabla^2\frac{\delta F}{\delta \psi}$$

where

$$M_1 \equiv (M_A + M_B)/\rho_l^2$$

$$M_2 \equiv (M_A - M_B)/\rho_l^2$$

* Amplitude expansion: Binary alloys, statics

Small deformation limit $\eta_j = \phi \exp(i \vec{G}_j \cdot \vec{u})$

$\psi \equiv$ concentration difference, $\phi \equiv$ liquid/solid order parameter

Elder, Huang, Provatas, PRE, **81**, 011602 (2010)

$$F_{2d} = \int d\vec{r} \Big[3\Delta B \phi^2 - 4t\phi^3 + \frac{45}{2} v \phi^4 + 6B^x |\vec{\nabla}\phi|^2 + 3B^x \phi^2 \Big\{ \frac{3}{2} \sum_{i=1}^{2} U_{ii}^2 + U_{xx}U_{yy} + 2U_{xy}^2 \Big\}$$
$$+ \left(\omega + 6B_2^l \phi^2\right) \frac{\psi^2}{2} + \frac{u}{4}\psi^4 + \frac{K}{2} |\vec{\nabla}\psi|^2 + 12\alpha B_0^x \Big(-\phi \nabla^2 \phi + \sum_{i=1} 2U_{ii}\phi^2 \Big) \psi \Big]$$

First Order Liquid/Solid transition with surface energy

Elastic energy

Phase Segregation, eutectic solidification, spindodal decomposition, etc.. with surface energy cost

Vegard's Law
$a = a_o(1+\alpha\Psi)$

Segregation at surfaces, dislocations, etc.

* Amplitude expansion: Binary alloys, dynamics

Small deformation limit $\eta_j = \phi \exp(i\vec{G}_j \cdot \vec{u})$

$\psi \equiv$ concentration difference, $\phi \equiv$ liquid/solid order parameter

Elder, Huang, Provatas, PRE, **81**, 011602 (2010)

$$F_{2d} = \int d\vec{r} \left[3\Delta B \phi^2 - 4t\phi^3 + \frac{45}{2} v \phi^4 + 6B^x |\vec{\nabla}\phi|^2 + 3B^x \phi^2 \left\{ \frac{3}{2} \sum_{i=1}^{2} U_{ii}^2 + U_{xx} U_{yy} + 2U_{xy}^2 \right\} \right.$$
$$\left. + \left(\omega + 6B_2^l \phi^2\right) \frac{\psi^2}{2} + \frac{u}{4}\psi^4 + \frac{K}{2} |\vec{\nabla}\psi|^2 + 12\alpha B_0^x \left(-\phi \nabla^2 \phi + \sum_{i=1} 2U_{ii} \phi^2 \right) \psi \right]$$

$$\frac{\partial \phi}{\partial t} = -\frac{\delta F}{\delta \phi}$$

Model **A**
Allen/Cahn

$$\frac{\partial \psi}{\partial t} = \nabla^2 \frac{\delta F}{\delta \psi}$$

Model **B**
Cahn/Hilliard
Hillert

Model **C** (Model A + Model B)

$$\sum_i \frac{\partial}{\partial x_i} \frac{\delta F}{\delta U_{ij}} \approx 0$$

Mechanical
Equilibrium

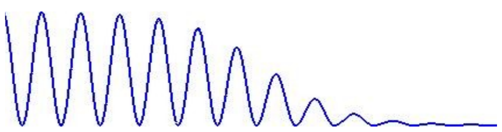
ABC's of pattern formation

Overview

Part 1: Multiscale Modeling

mechanical properties - elasticity, dislocations, grain boundaries, polycrystals, …

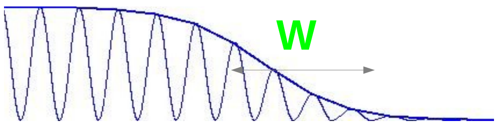
Phase field crystal



Δx ~ a/10

~0.1 Å

Amplitude



Δx ~ W/10

~1 Å

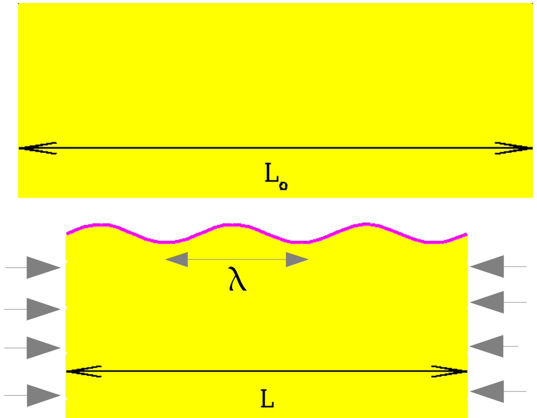
Continuum

Δx ~ W/10

~1 Å

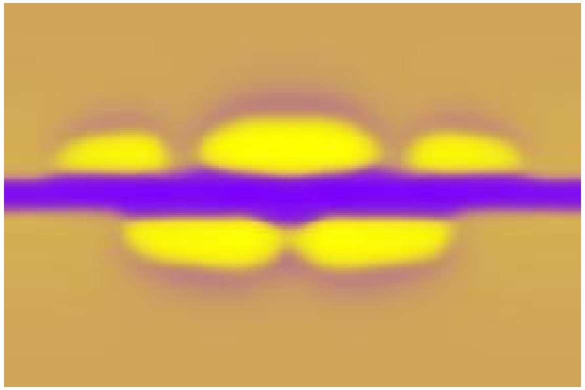
Part 2: Applications

Mounding instability



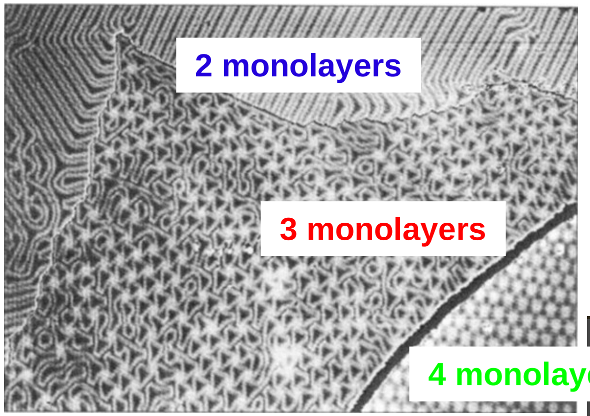
Wavelength Selection

Island formation on nano-membranes



Long range order?

Monolayer(s) Ordering

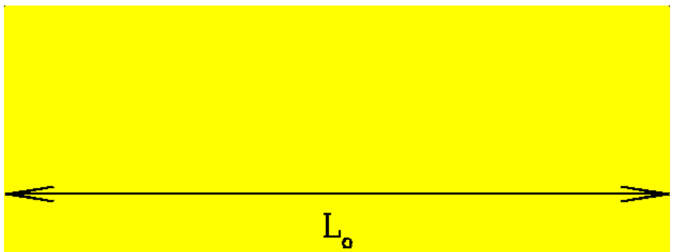


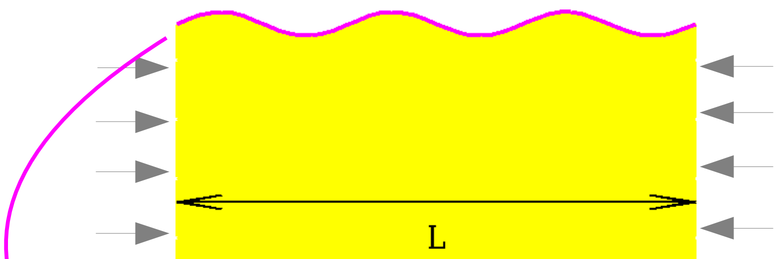
Pattern + Length Selection

* Amplitude expansion: applications

Mounding instability: stability of strained surface

Huang and Elder, PRL **101**, 158701 (2008), PRB **81**, 165421 (2010)

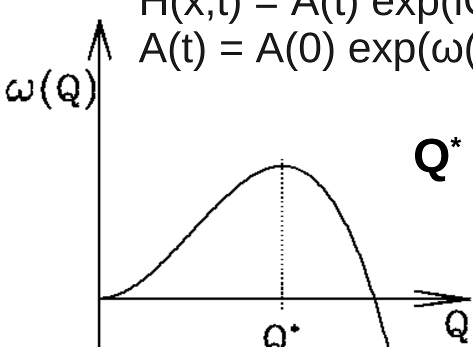




Misfit strain $\varepsilon_M \equiv (a_o - a)/a$

$a_o \equiv$ lattice constant of stress-free bulk film
$a \equiv$ lattice constant of strained film

Surface Profile: H(x,t)
H(x,t) = A(t) exp(iQx)
A(t) = A(0) exp(ω(Q)t)



Q* most unstable wavevector

1) Convenient to expand around strained state

$$n = \sum_j \eta_j e^{i\vec{G}_j\cdot\vec{r}} \rightarrow \sum_j \eta_j e^{i(1-\delta)\vec{G}_j\cdot\vec{r}}$$

where $\varepsilon_M = \delta/(1-\delta)$

2) Perturb around 1-d profile, $\eta_j^o(y)$

$$\eta_j(x,y,t) = \eta_j^o(y) + \delta\eta(y,t)e^{iQx}$$

3) Numerically solve for,

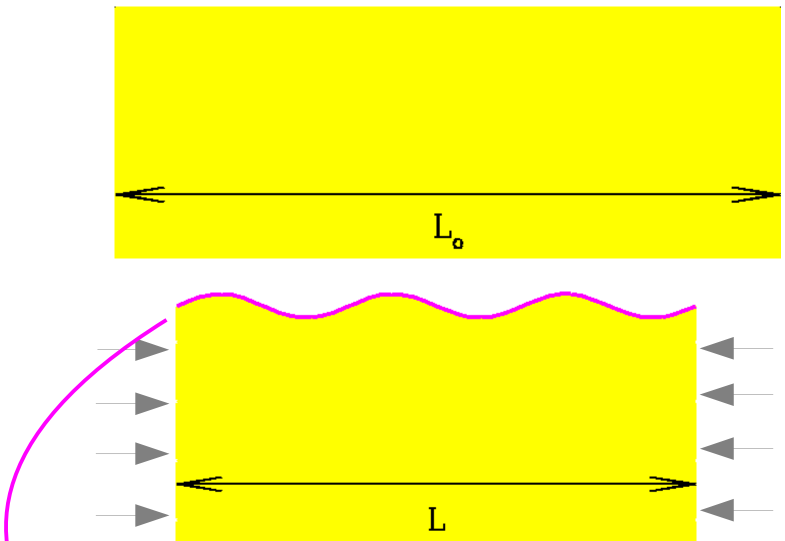
$$\delta\eta(y,t)$$

.
.
.
.

* Amplitude expansion: applications

Mounding instability: stability of strained surface

Huang and Elder, PRL **101**, 158701 (2008), PRB **81**, 165421 (2010)



Misfit strain $\varepsilon_M \equiv (a_o - a)/a$

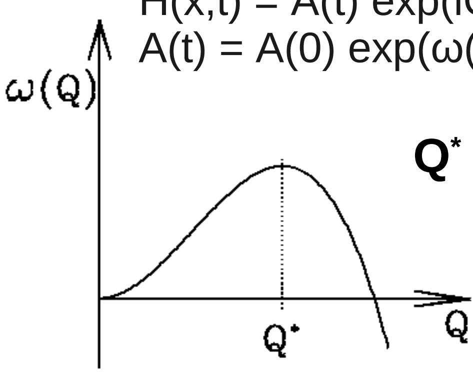
$a_o \equiv$ lattice constant of stress-free bulk film

$a \equiv$ lattice constant of strained film

Surface Profile: H(x,t)

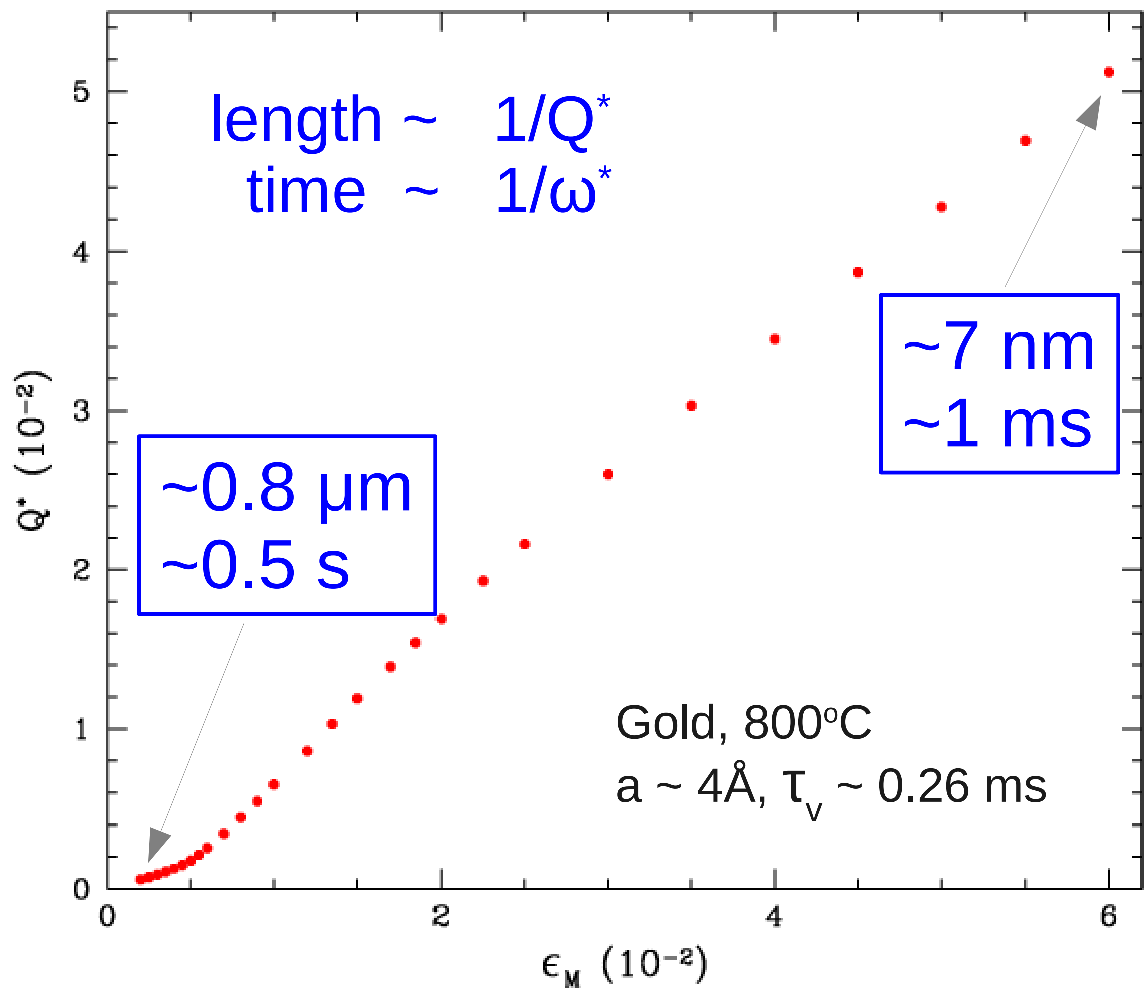
H(x,t) = A(t) exp(iQx)

A(t) = A(0) exp(ω(Q)t)



Q^* most unstable wavevector

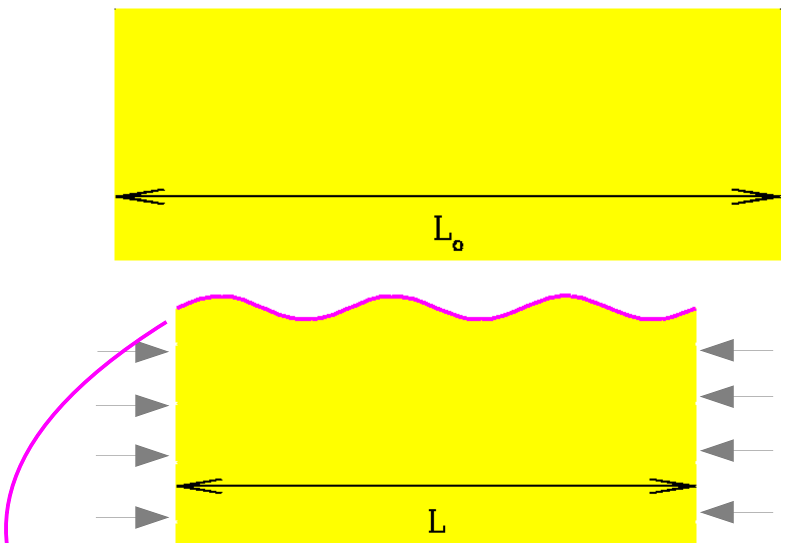
length, time scales



* Amplitude expansion: applications

Mounding instability: stability of strained surface

Huang and Elder, PRL **101**, 158701 (2008), PRB **81**, 165421 (2010)



Misfit strain $\varepsilon_M \equiv (a_o - a)/a$

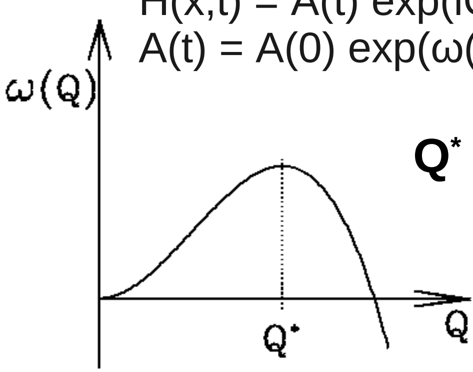
$a_o \equiv$ lattice constant of stress-free bulk film

$a \equiv$ lattice constant of strained film

Surface Profile: H(x,t)

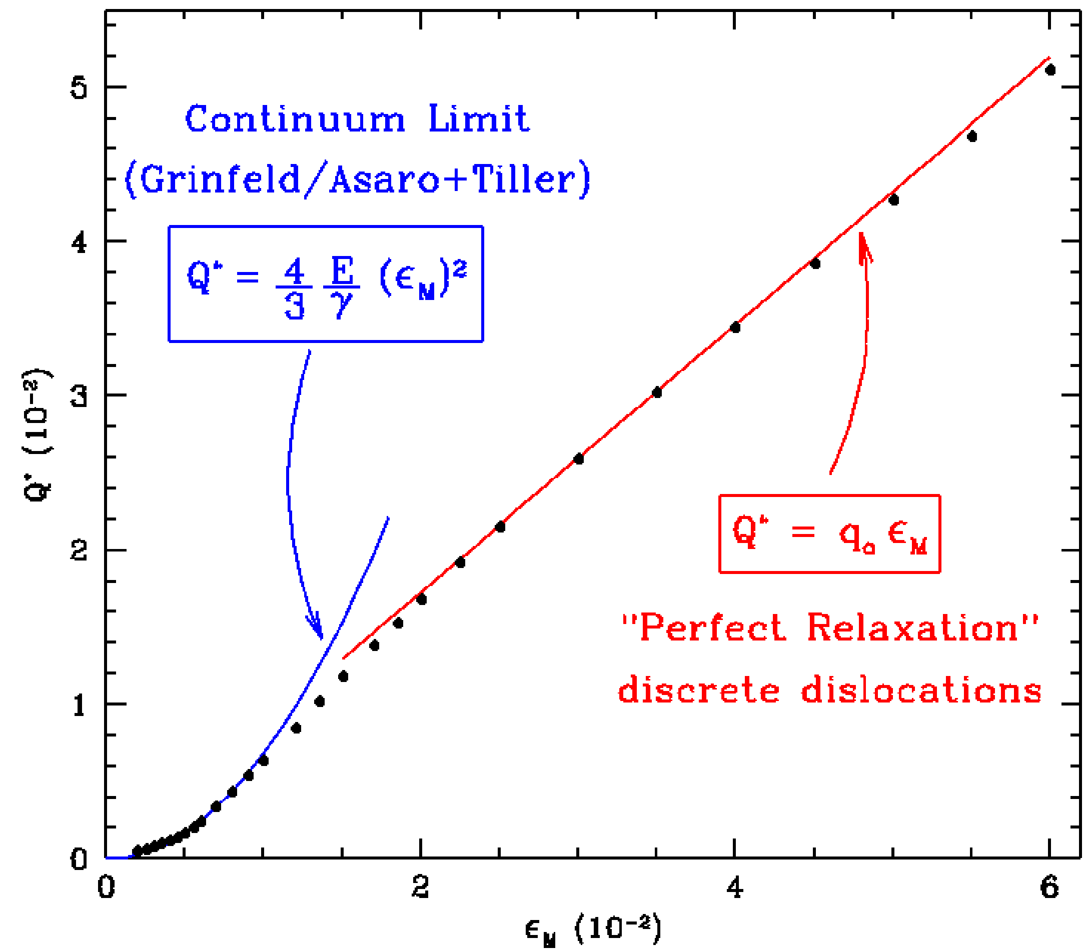
H(x,t) = A(t) exp(iQx)

A(t) = A(0) exp(ω(Q)t)



Q* most unstable wavevector

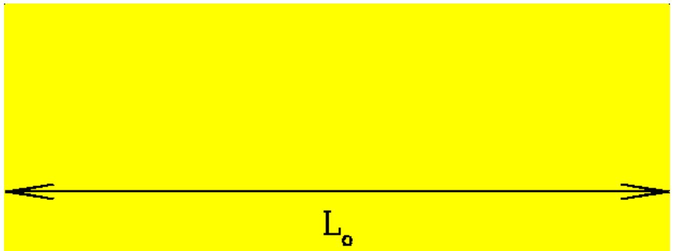
crossover : Continuum - Discrete

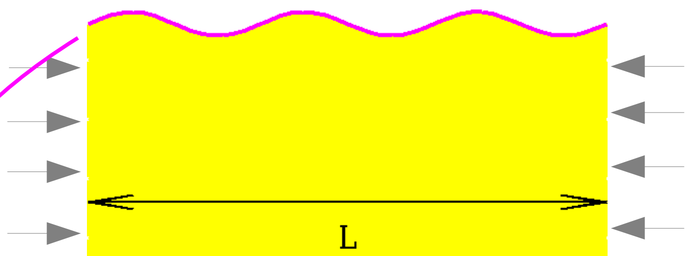


* Amplitude expansion: applications

Mounding instability: stability of strained surface

Huang and Elder, PRL **101**, 158701 (2008), PRB **81**, 165421 (2010)





Misfit strain $\varepsilon_M \equiv (a_o - a)/a$

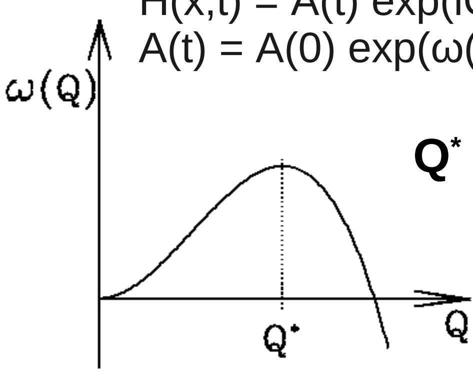
$a_o \equiv$ lattice constant of stress-free bulk film

$a \equiv$ lattice constant of strained film

Surface Profile: H(x,t)

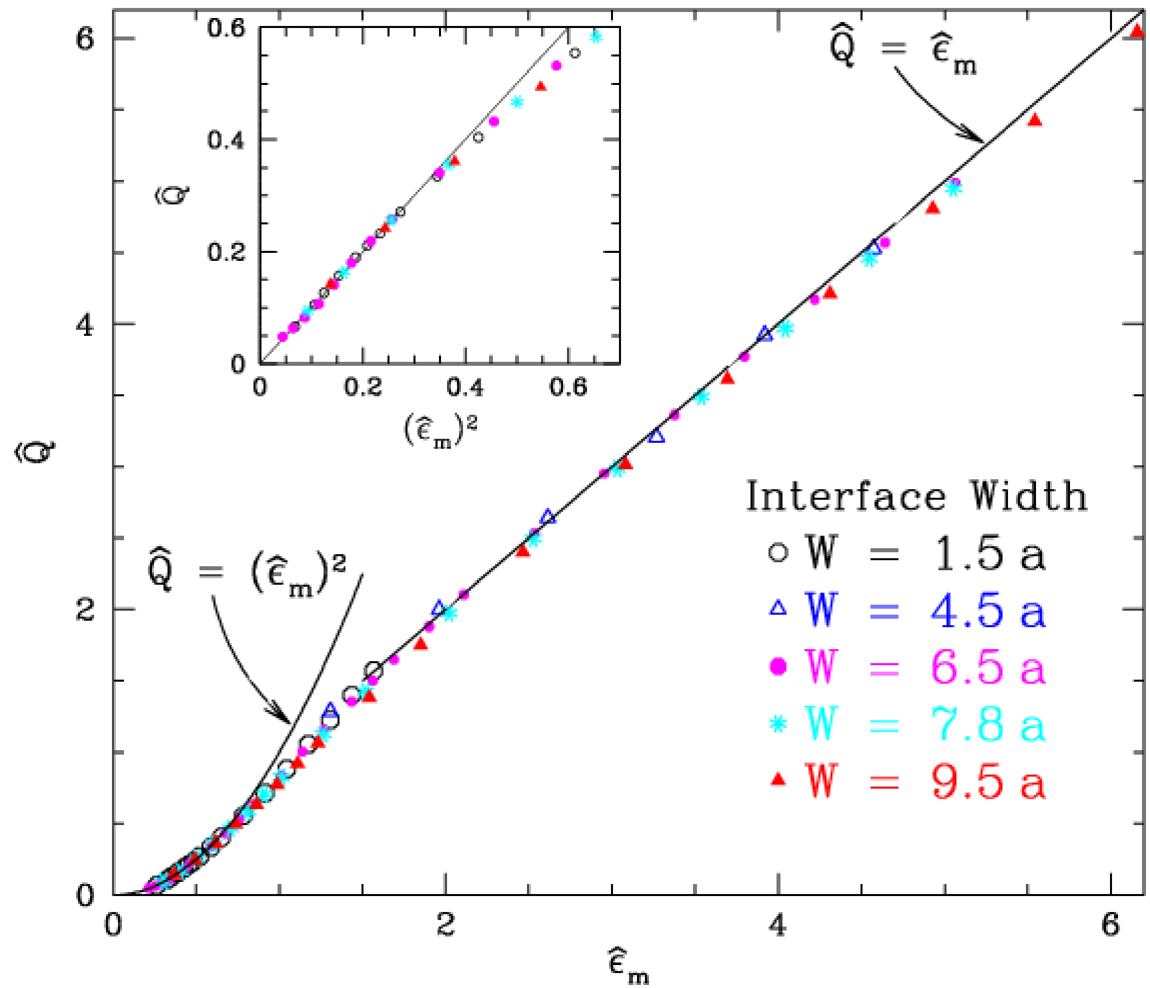
H(x,t) = A(t) exp(iQx)

A(t) = A(0) exp(ω(Q)t)



Q^* most unstable wavevector

universal scaling $\hat{Q} \equiv Q^*/Q^*_c,\ \hat{\varepsilon} \equiv \varepsilon_m/\varepsilon_m^c$



Epitaxial growth: stability of strained surface

$Si_{1-x}Ge_x$ grown on Si : back to back papers

Sutter/Lagally, PRL, **84**, 4637 (2000)

ment with all theoretical models for the morphological instability of strained layers. From Fig. 4 we deduce $\lambda \propto \varepsilon^{-(1.0\pm0.1)}$.

Our LEEM and AFM results are in obvious qualitative disagreement with simple models [8,18] that predict a functional form for the wavelength $\lambda(\varepsilon)$ of the instability

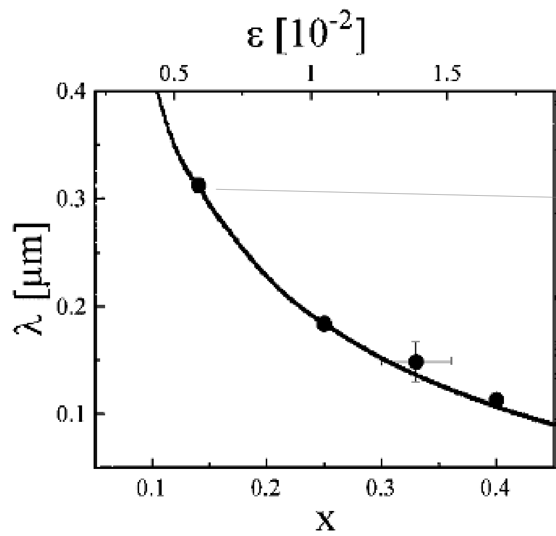
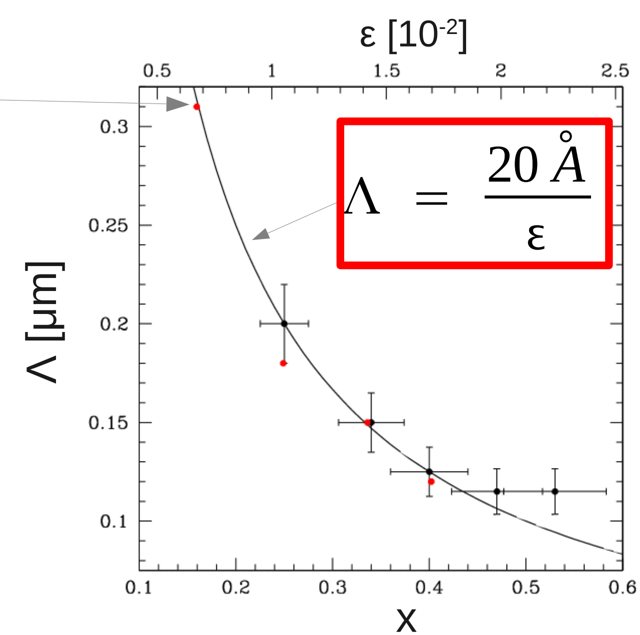


FIG. 4. Dependence of the wavelength λ of the initial cell pattern on Ge concentration, x, of the alloy film. Points denote values measured from LEEM images. The line is a calculation according to the model of Ref. [19], using a dimensionless growth rate $v = 10^{-3}$ and solute expansion coefficient $\eta^* = 0.5$.

Tromp/Ross/Reuter, PRL, **84**, 4641 (2000)

TABLE I. Roughening length Λ and roughening thickness T_R as a function of Ge concentration x. Experimental uncertainty in x is $\pm10\%$, in $\Lambda \pm 10\%$, in $T_R \pm 15\%$.

Ge Concentration x	Roughening length Λ	Roughening thickness T_R
0.25	200 nm	11.6 nm
0.34	150 nm	6.8 nm
0.40	125 nm	6.0 nm
0.47	115 nm	6.4 nm
0.53	115 nm	5.9 nm



perfect relaxation

$$\Lambda = \frac{5.5\,\mathring{A}}{\varepsilon}$$

Guyer + Voorhees, PRL **75**, 4031 (1995)

-- compositional inhomogeneities

* Amplitude expansion: applications

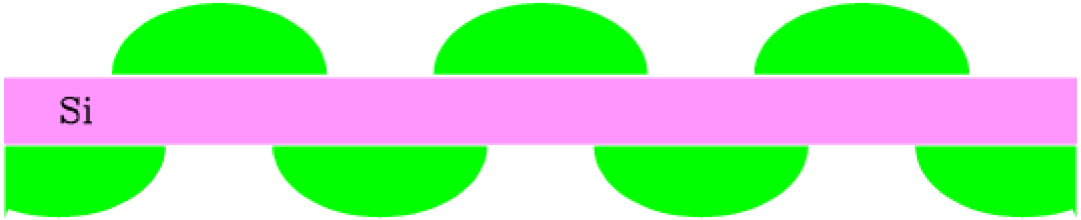
Island Formation on Nano-membranes

Elder, Huang J. Phys.: Condens. Matter. 22 , 364103 (2010).

Why on a nano-membrane?

- Maximum island size ~ 1/(nano-membrane thickness)

- Nanomembrane ~ induce island ordering

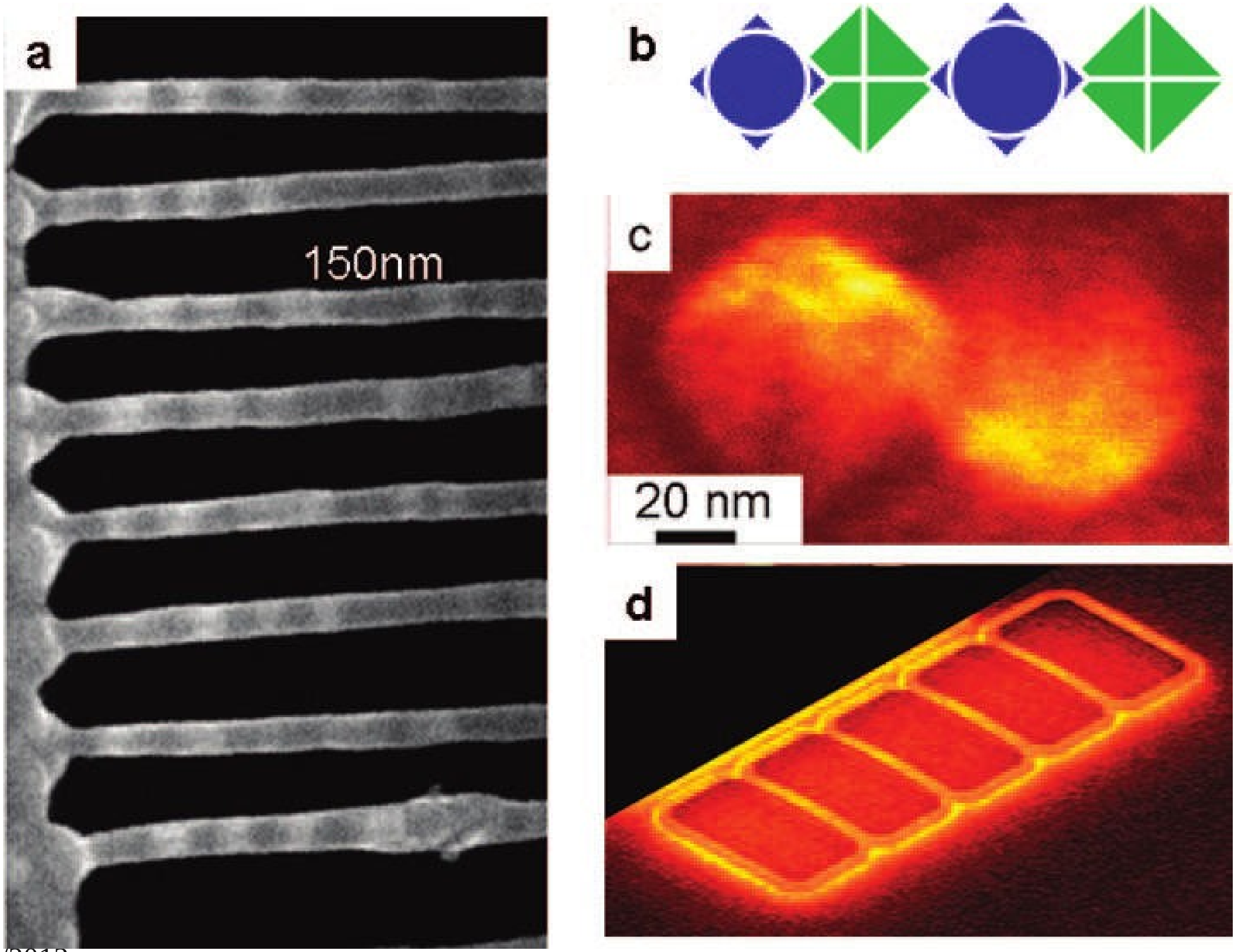


Ordering - modulated strain in Si nanomembrane
- modulated electronic band gap

Exploit – nanoscale photonic and electronic devices

Island Formation on Nano-membranes

Huang, Ritz, Novakovic, Yu, Zhang, Flack, Savage, Evans, Knezevic, Liu, Lagally, ACS Nano 3, 721 (2009)



Island Formation on Nano-membranes

Kim-Lee, Savage, Ritz, Lagally, Turner, PRL 102, 226103 (2009)

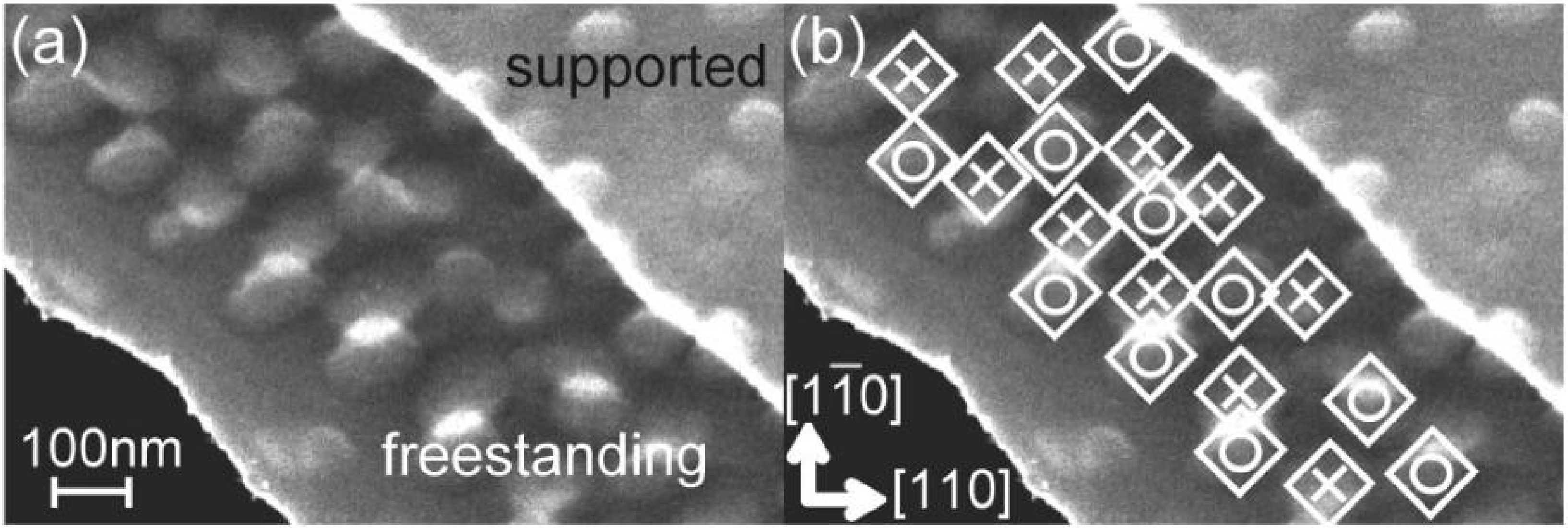
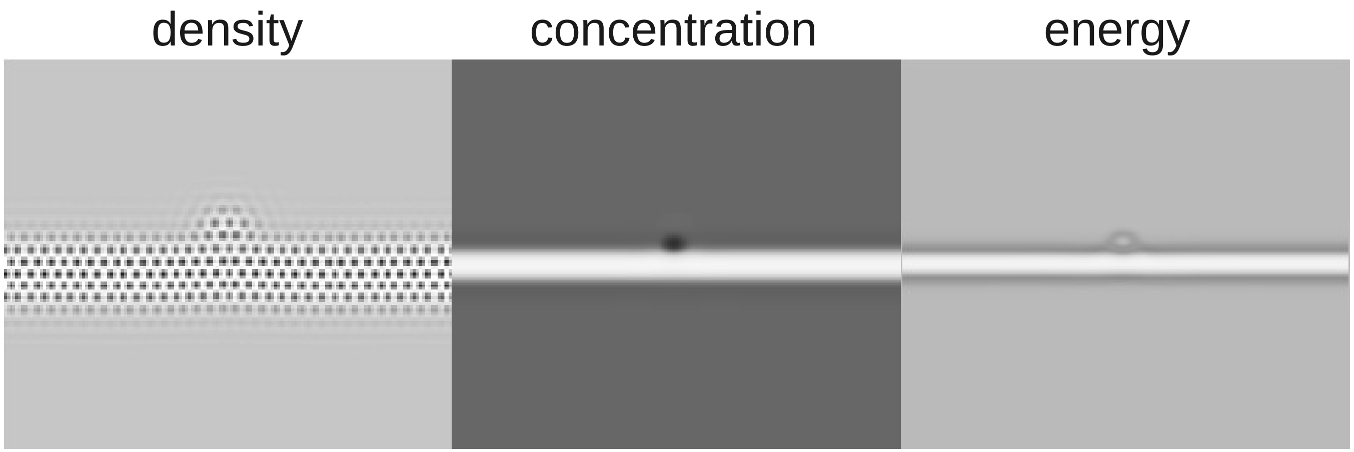


FIG. 1. (a) A Scanning electron micrograph of $Si_{0.36}Ge_{0.64}$ islands on a 23 nm thick Si membrane. The average hut size is about 70 nm, nearly three times the membrane thickness. The ordering is better on the freestanding region than the supported region. (b) The image in (a) with huts on the "top" and "bottom" of the freestanding region marked with O and X, respectively.

Island Formation on Nano-membranes

Binary Amplitude Model simulations



Film thickness
~ 6 layers

Island Formation on Nano-membranes

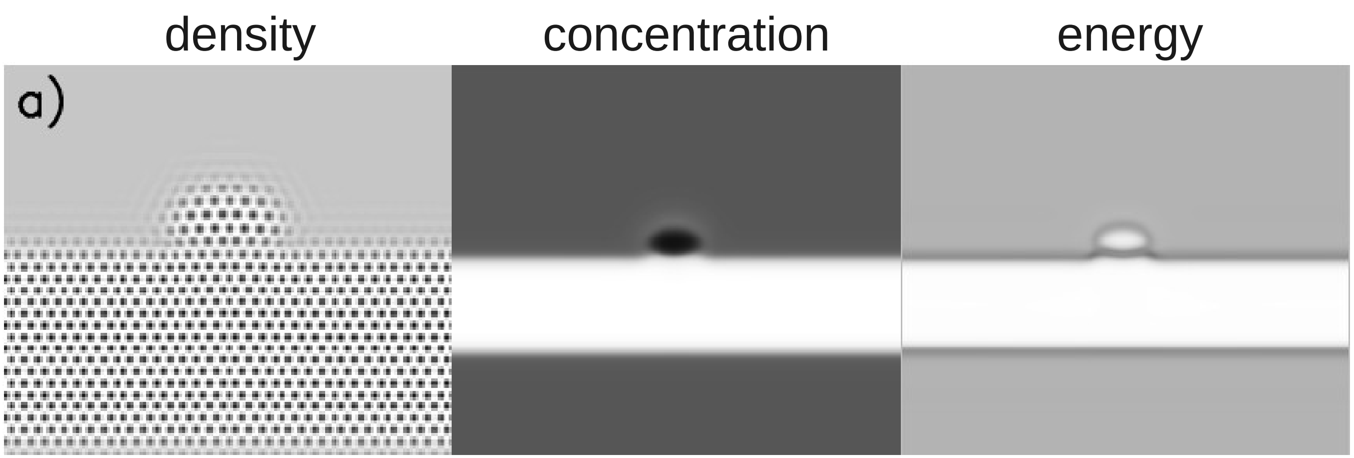
Binary Amplitude Model simulations

density concentration energy

Film thickness
~ 6 layers

Island Formation on Nano-membranes

Binary Amplitude Model simulations



Film thickness
~ 17 layers

Island Formation on Nano-membranes

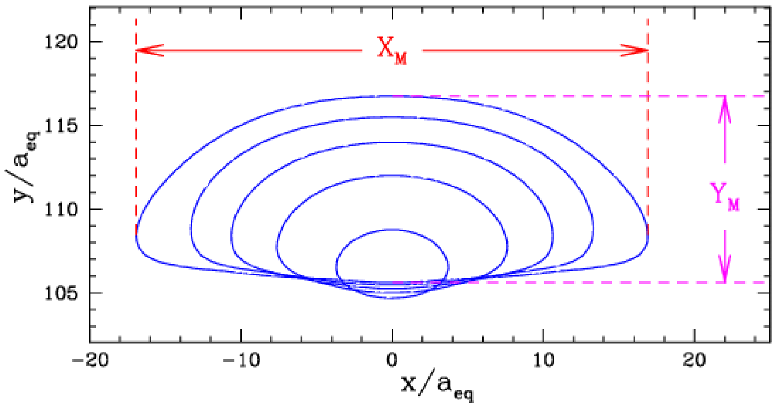
Binary Amplitude Model simulations

density concentration energy

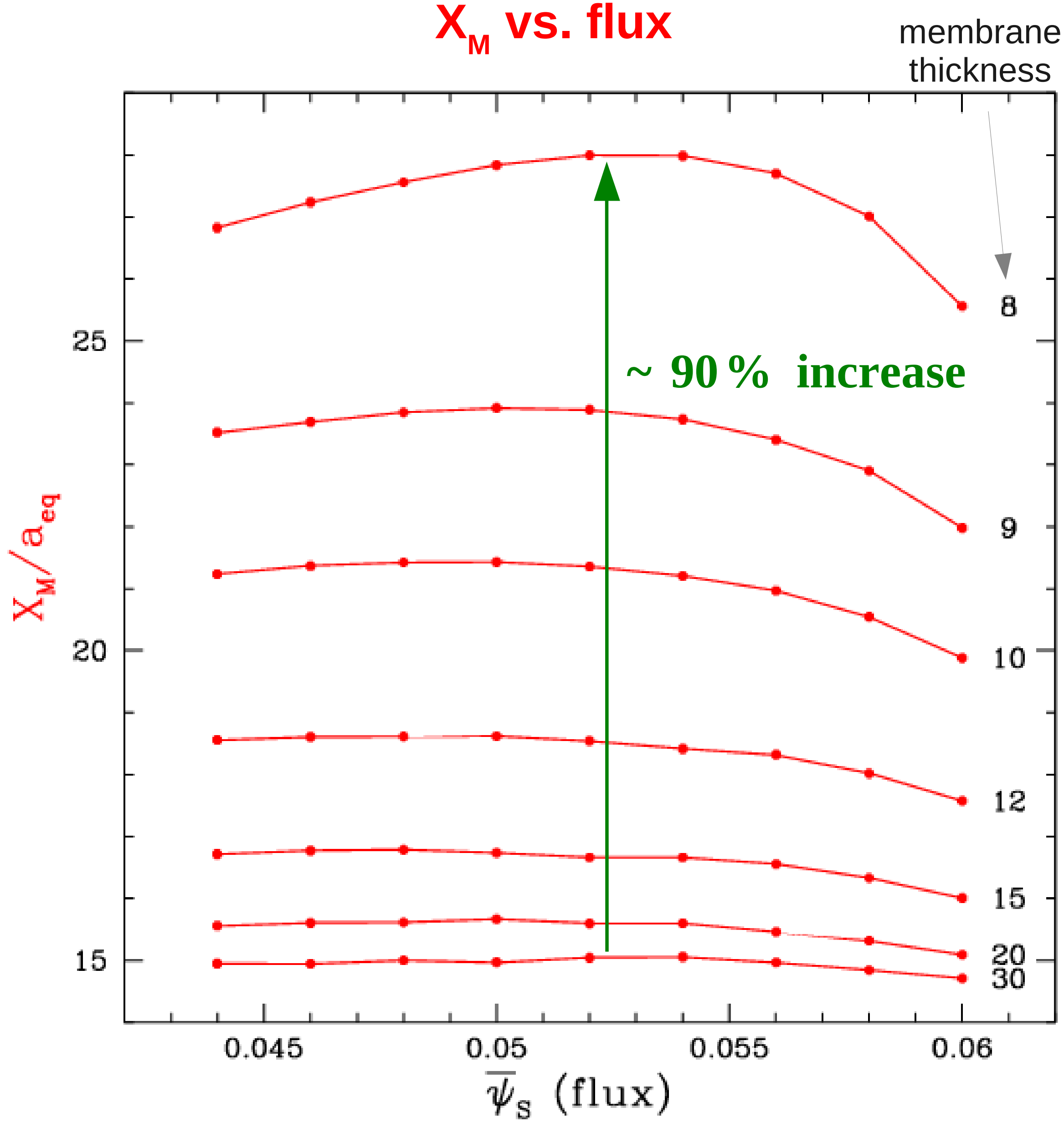
Film thickness
~ 17 layers

Island Formation on Nano-membranes: size

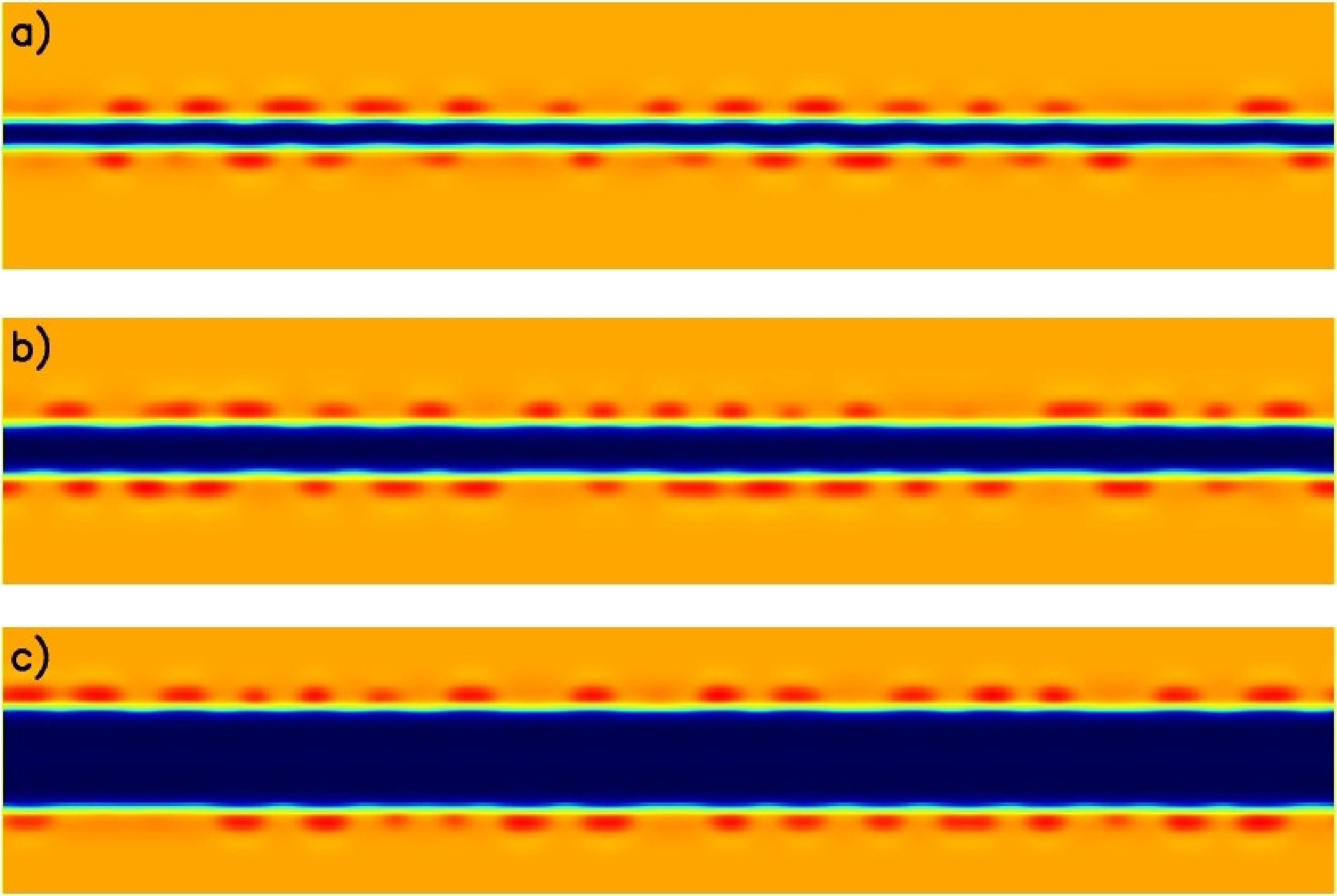
$X_M \equiv$ Maximum Island size before dislocation formation or membrane break



X_M vs. flux



Island Formation on Nano-membranes: ordering?

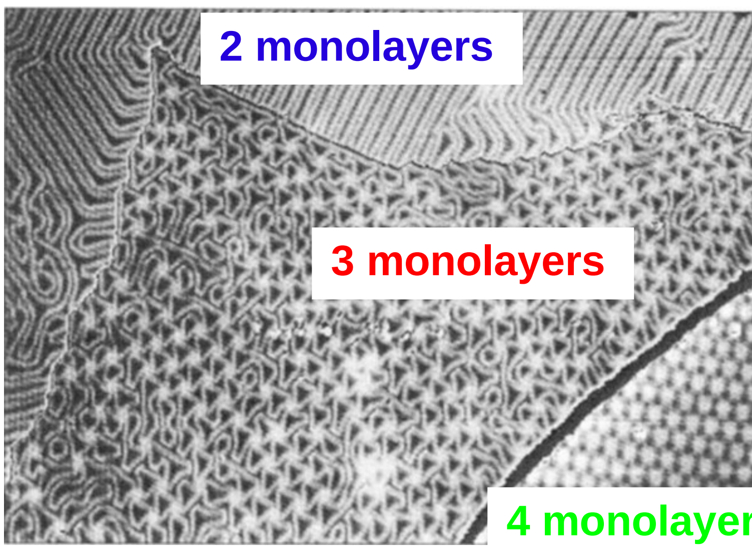


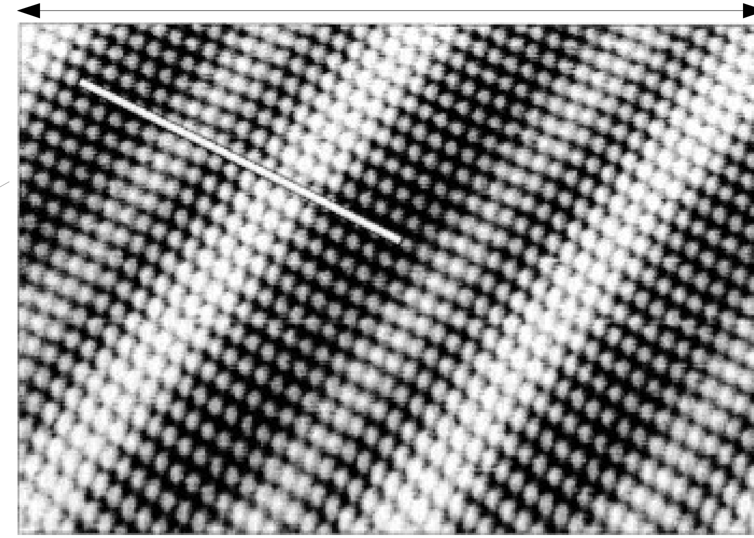
* Amplitude expansion: applications

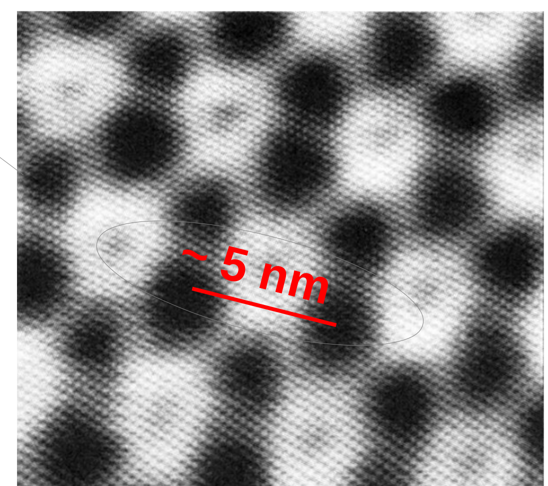
Patterning in Ultrathin Films

Example: **Cu on Ru (0001) surface**
Misfit strain 5.6%

top view





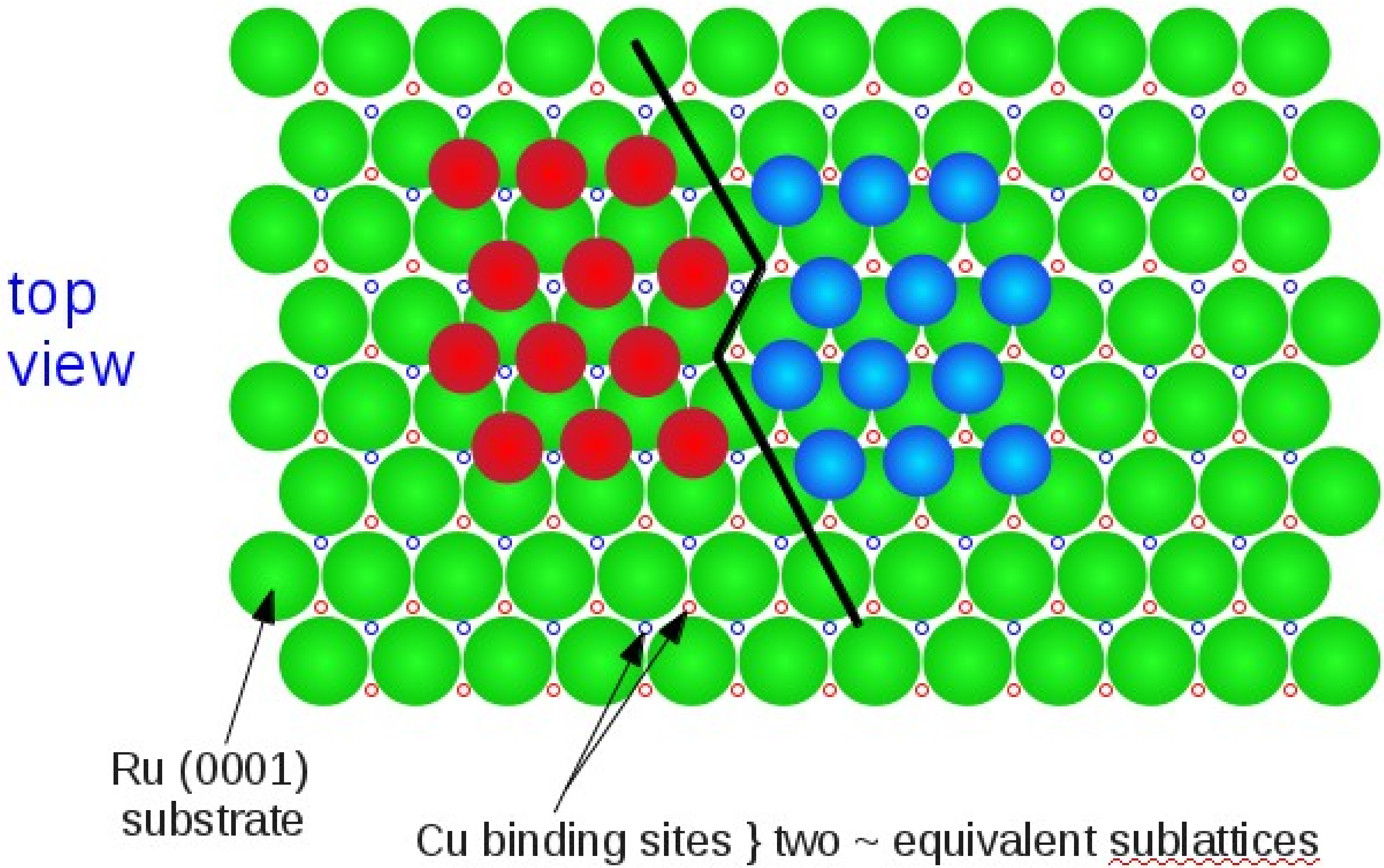


angstrom sized atoms forming **nanometer** sized objects ordering on **micron** scales

* Amplitude expansion: applications

Monolayer(s) ordering: Cu on Ru (0001)

Elder, Rossi, Kanerva, Sanches,Ying, Granato, Achim and Ala Nissila
PRL, **108**, 226102 (2012)



sublattice ordering
HCP sites
FCC sites

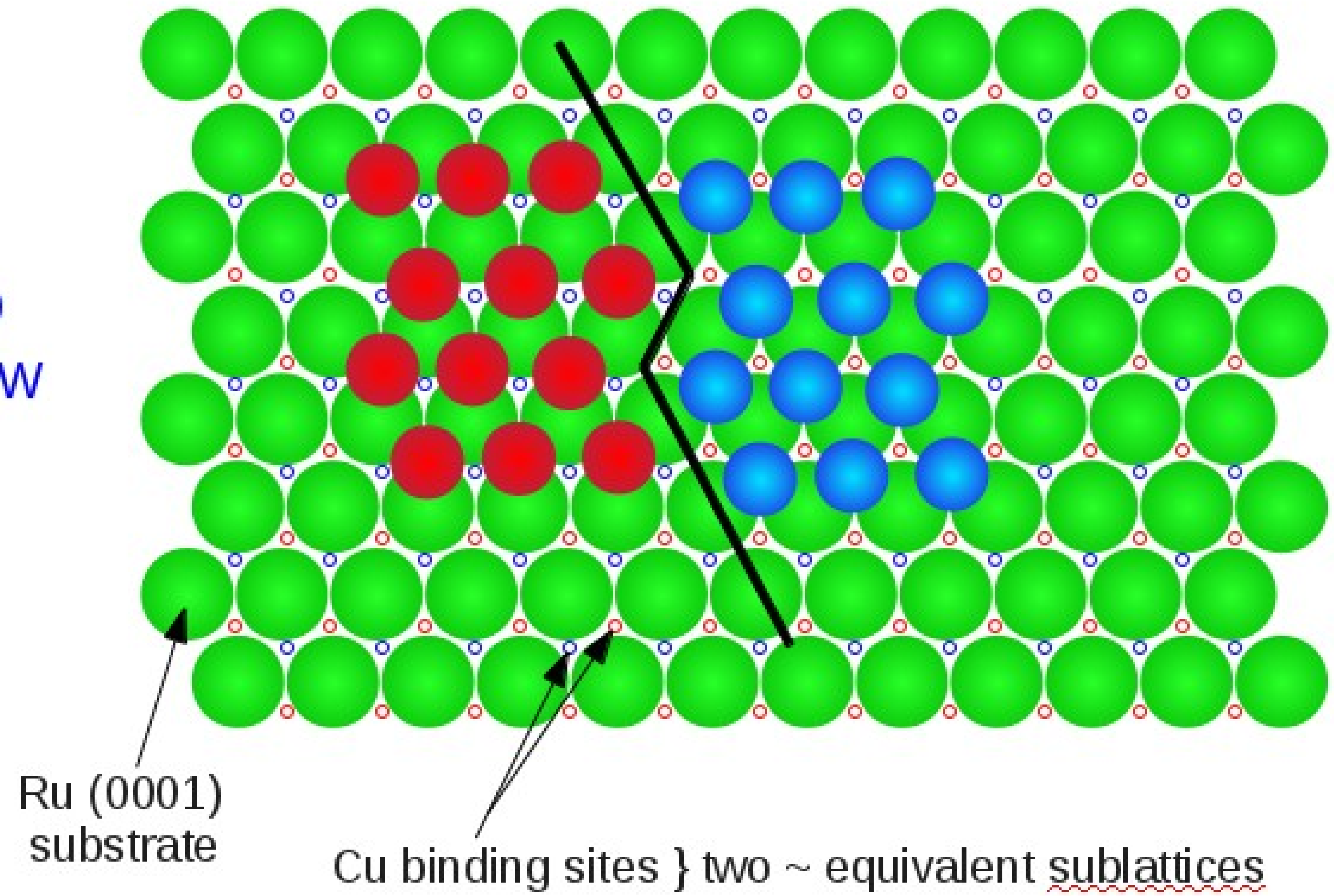
domain walls energy cost

* Amplitude expansion: applications

Monolayer(s) ordering: Cu on Ru (0001)

Elder, Rossi, Kanerva, Sanches,Ying, Granato, Achim and Ala Nissila
PRL, **108**, 226102 (2012)

top view



strain

a^{Ru} **= 2.70 Å**

a^{Cu} **= 2.55 Å**

ε = 5.6 %

domain walls relieve strain

Monolayer ordering: Fixed substrate potential

Approximate: substrate = **fixed potential** of the form

$$V = V_o \left[\sum_j e^{i \vec{q}_j^{Ru} \cdot \vec{r}} + c.c. \right]$$

Two important parameters
Misfit strain, ε
Substrate coupling, V_o

add to Free energy, i.e.,

$$F = \int d\vec{r} \left[\frac{B^l}{2} n^2 + \ldots + \mathbf{V} \; \mathbf{n} \right]$$

where $\vec{q}_j^{Ru} \equiv$ reciprocal lattice vectors for triangular lattice

New amplitude model

$$\frac{\partial \eta_j}{\partial t} = -\left[\left(\Delta B_o + B^x \Im_j^2 + 3v\left(A^2 - |\eta_j|^2\right)\right)\eta_j - 2t \prod_{i \neq j} \eta_i^* + \mathbf{V_o} \right]$$

where $\Im_j \equiv \nabla^2 + 2i\alpha \vec{q}_j^{Cu} \cdot \vec{\nabla} + 1 - \alpha^2$

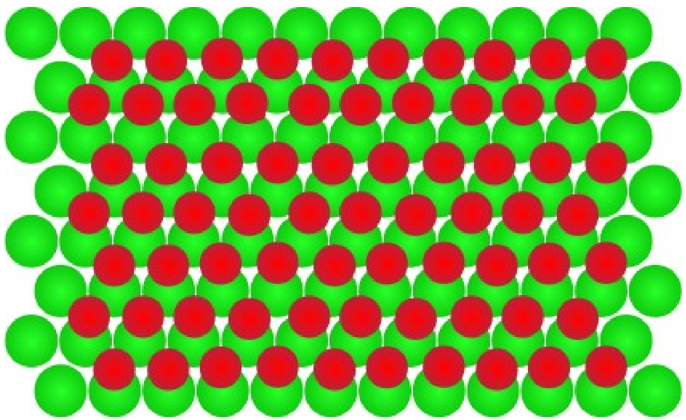
misfit strain $\varepsilon = 1 - \alpha$

Oakland UNIVERSITY

Equilibrium states ($\delta F/\delta \eta_j = 0$)

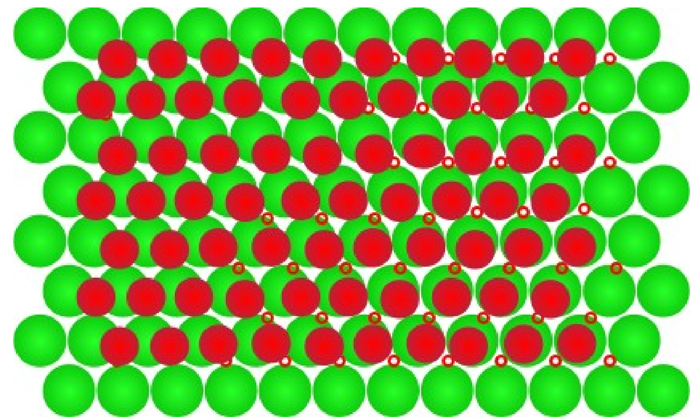
A) Uniform states

commensurate



large V_o

incommensurate



$V_o = 0$

Free energy difference

$$\Delta F_{ci} = F_c - F_i = \overbrace{-\,(3\,\Phi)\,V_o}^{\text{substrate coupling}} + \overbrace{(12\,B^x\,\Phi^2)\,\varepsilon^2}^{\text{elastic energy}}$$

where $\Phi \equiv$ amplitude of density fluctuations

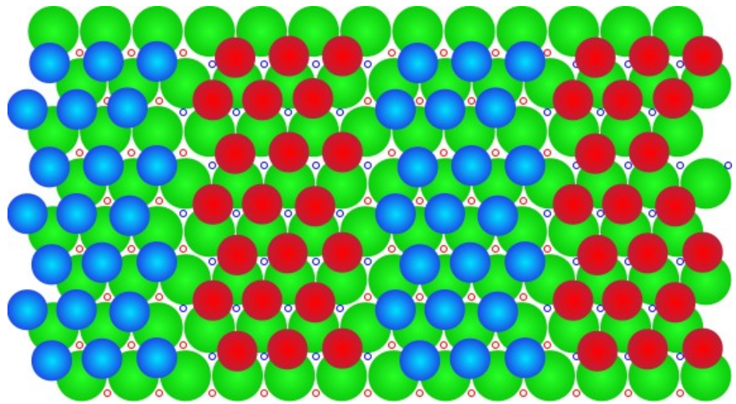
Commensurate lowest energy state if

$$V_o \geq V_{ci} = (4B^x\Phi)\,\varepsilon^2$$

Equilibrium states ($\delta F/\delta \eta_j = 0$)

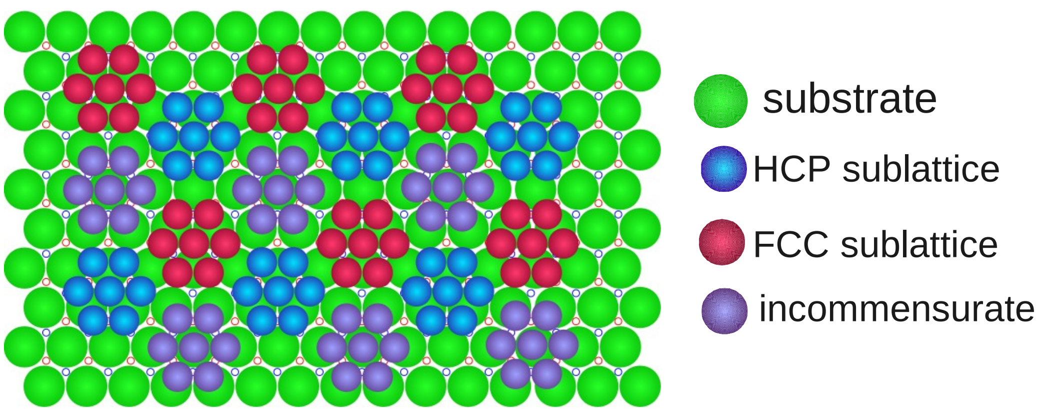
B) Periodic states

stripes



intermediate V_o

honeycomb



small V_o

Commensurate to Stripe transition (V_{cs})

simplify free energy → **Sine Gordon model**

$$V_{cs} \approx (9\pi^2 B^x/5)\, \varepsilon^2$$

Wavelength selection

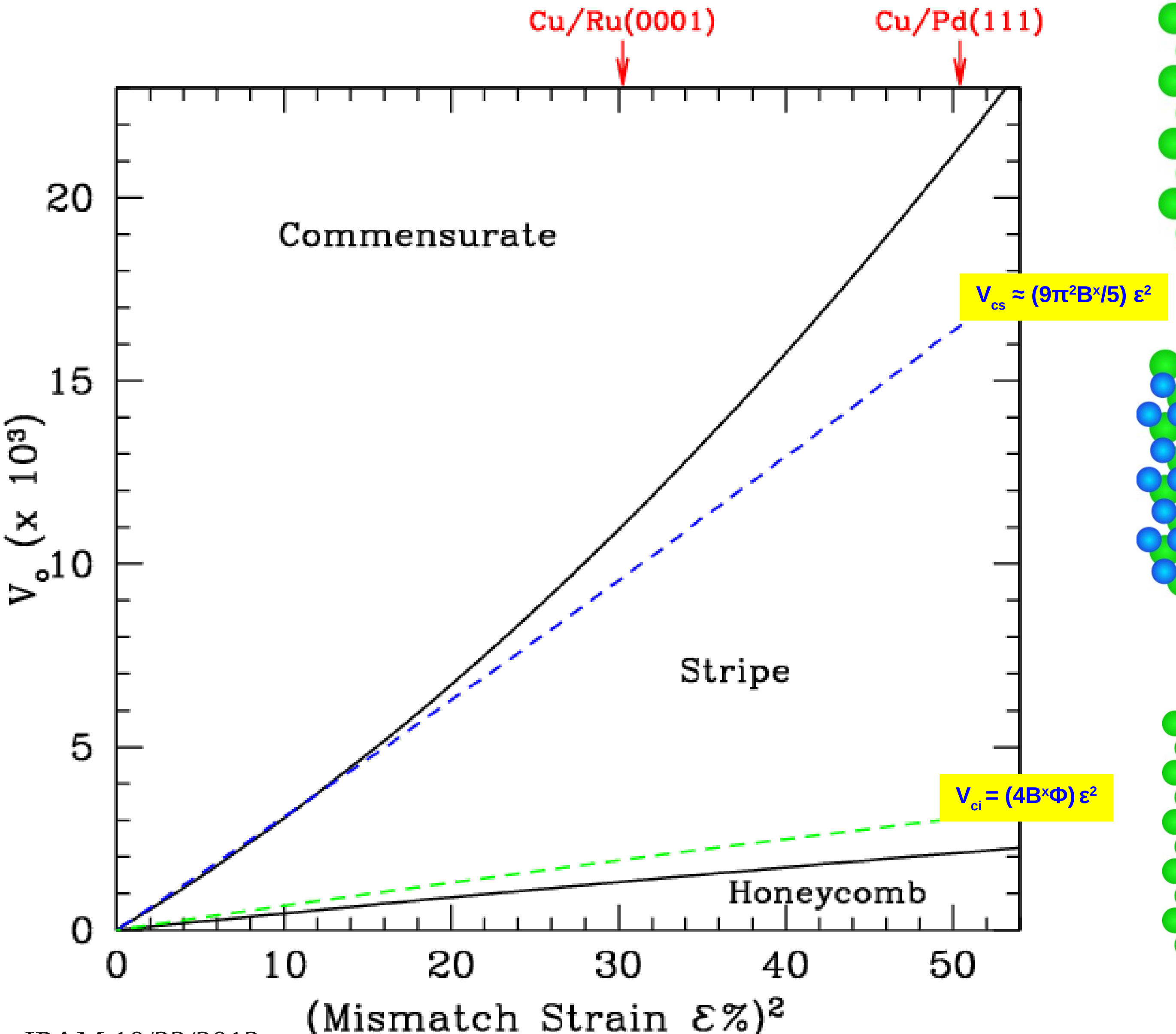
$$\lambda \approx -W \ln(\Delta V_o)$$

Where W ≡ domain walls width, $\Delta V_o \equiv (V_{cs} - V_o)/V_{cs}$

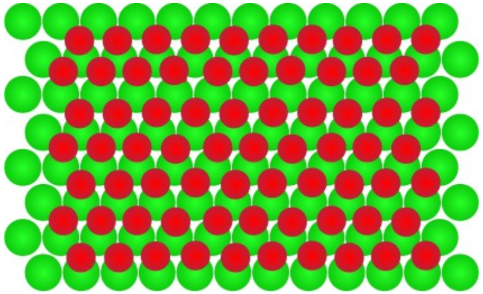
Stripe to Honeycomb transition (V_{sh})

numerical solution ....

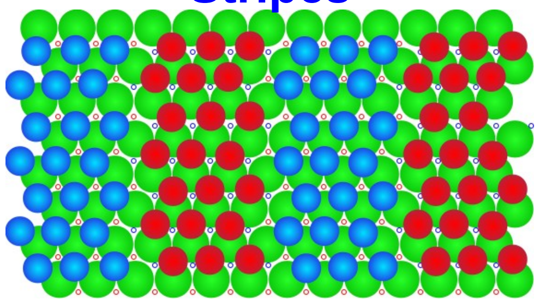
Phase Diagram



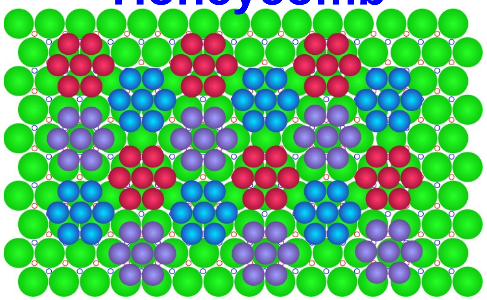
Commensurate



Stripes



Honeycomb



Oakland UNIVERSITY

Comparison with experiment

Pattern selection / Length scales

Theory: patterns controlled by V_o or ε, or

$$\text{Ratio} \sim \frac{E_{\text{int}}}{E_{str}} \sim \frac{V_o}{\varepsilon^2}$$

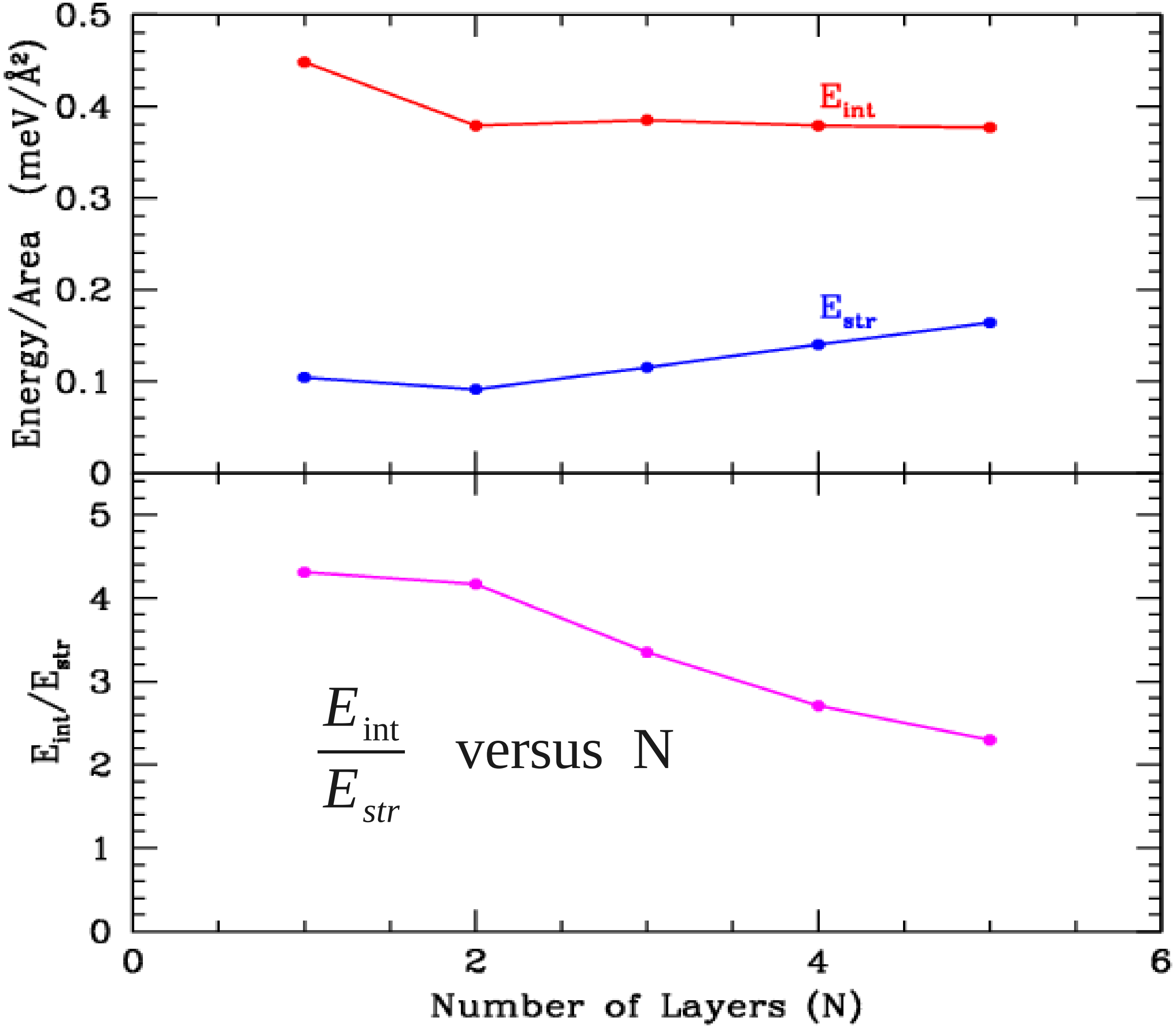
where, $E_{\text{int}} \equiv$ substate/film interaction energy

$E_{str} \equiv$ strain energy

Experiment controlled by number of layers (N)

$$\frac{E_{\text{int}}}{E_{str}} \text{ versus } N?$$

Cu/Pd semi-emperical calculations (G. Rossi)



Experiment
increasing layers
-> decreases ratio

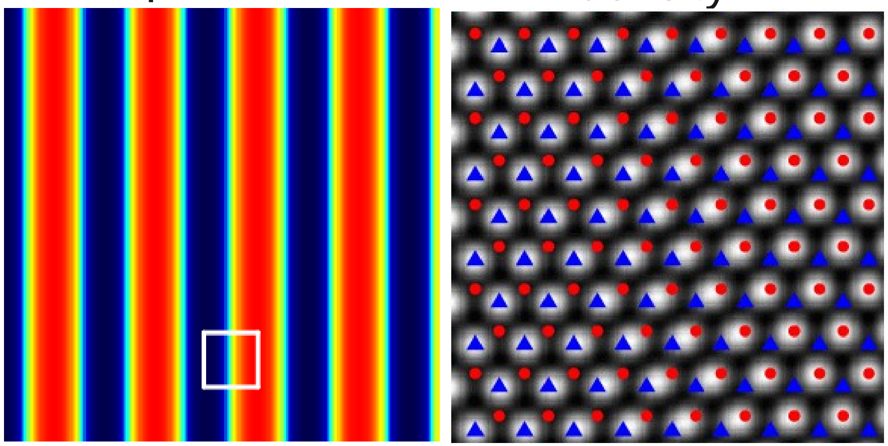
Theory
decreasing V_o
-> decreases ratio

decreasing V_o
~ increasing number of layers

Periodic equilibrium states

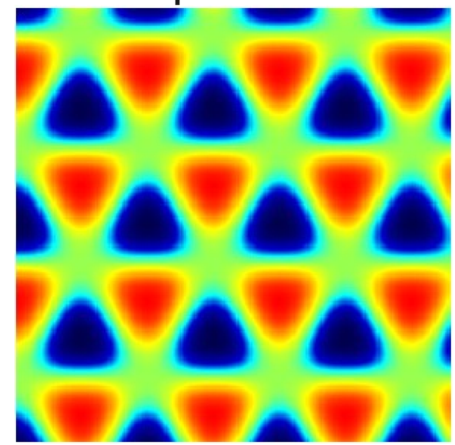
Stripe superlattice

phase density



Honeycomb superlattice

phase



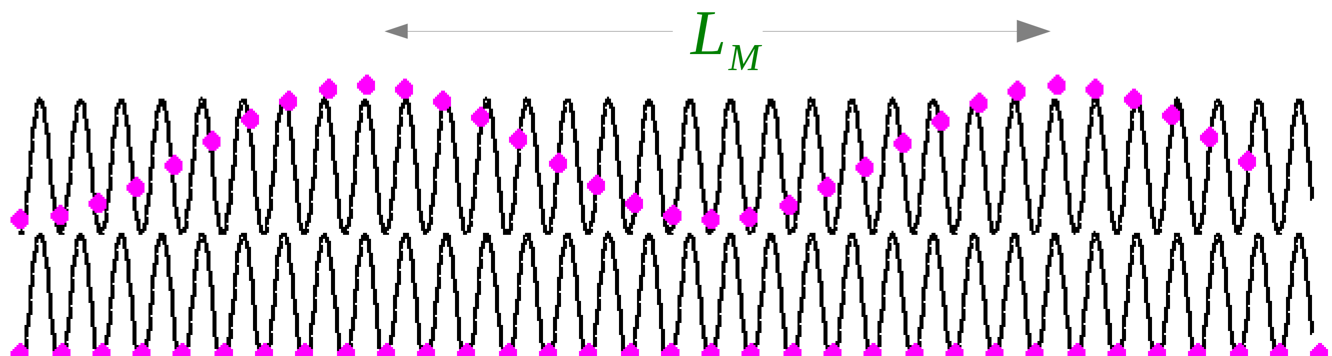
Length scale?

Natural length scale

$$L_M = \frac{2\pi}{q^{Ru} - q^{Cu}}$$

incommensurate

commensurate

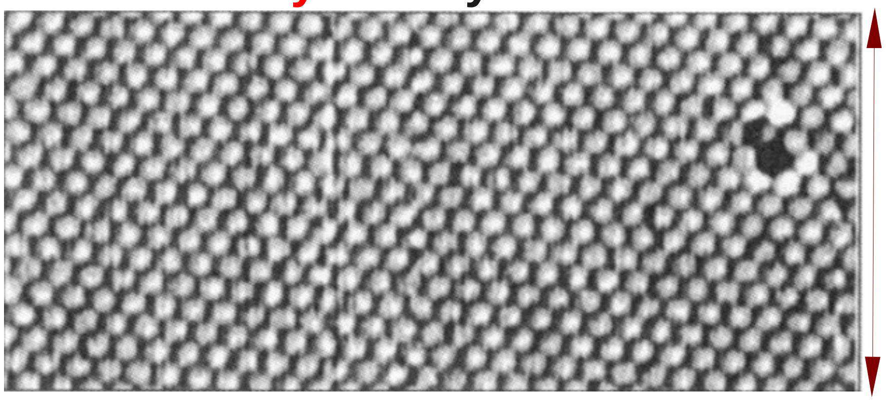


Monolayer ordering, Comparison with experiment

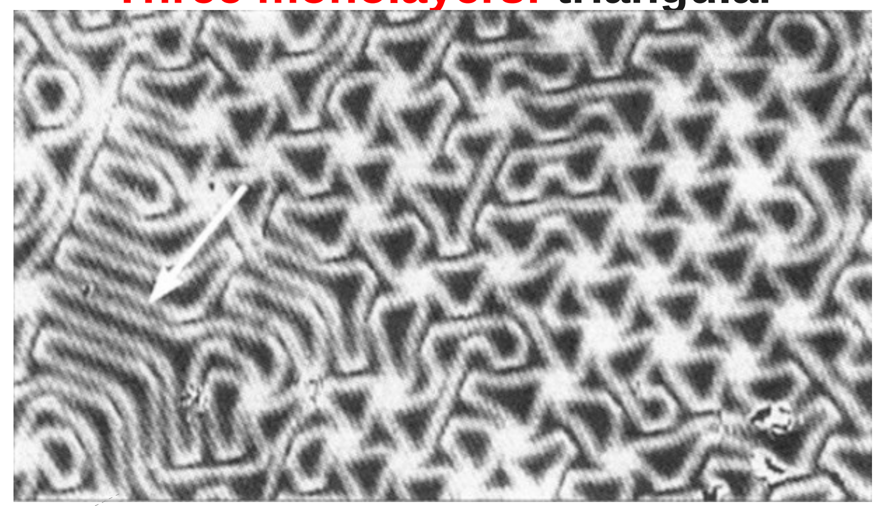
Günther, Vrijmoeth, Hwang and Behm, PRL **74**, 754 (1995): Cu/Ru(0001)

STM images

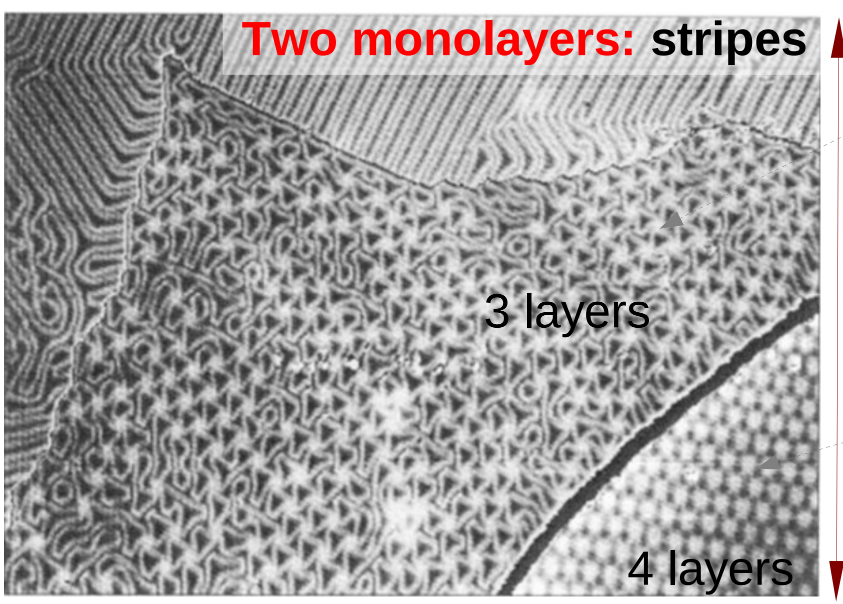
One monolayer: fully commensurate



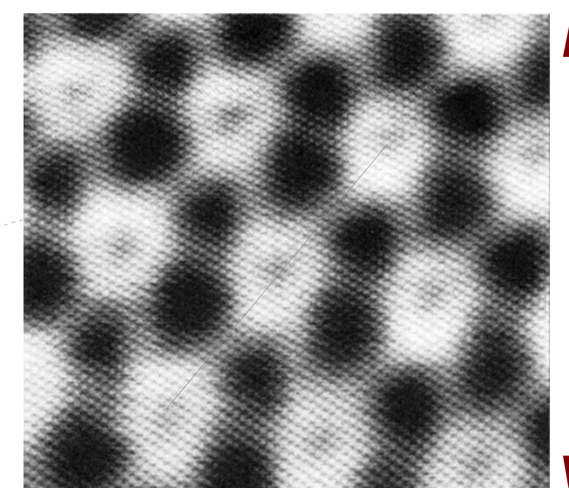
Three monolayers: triangular



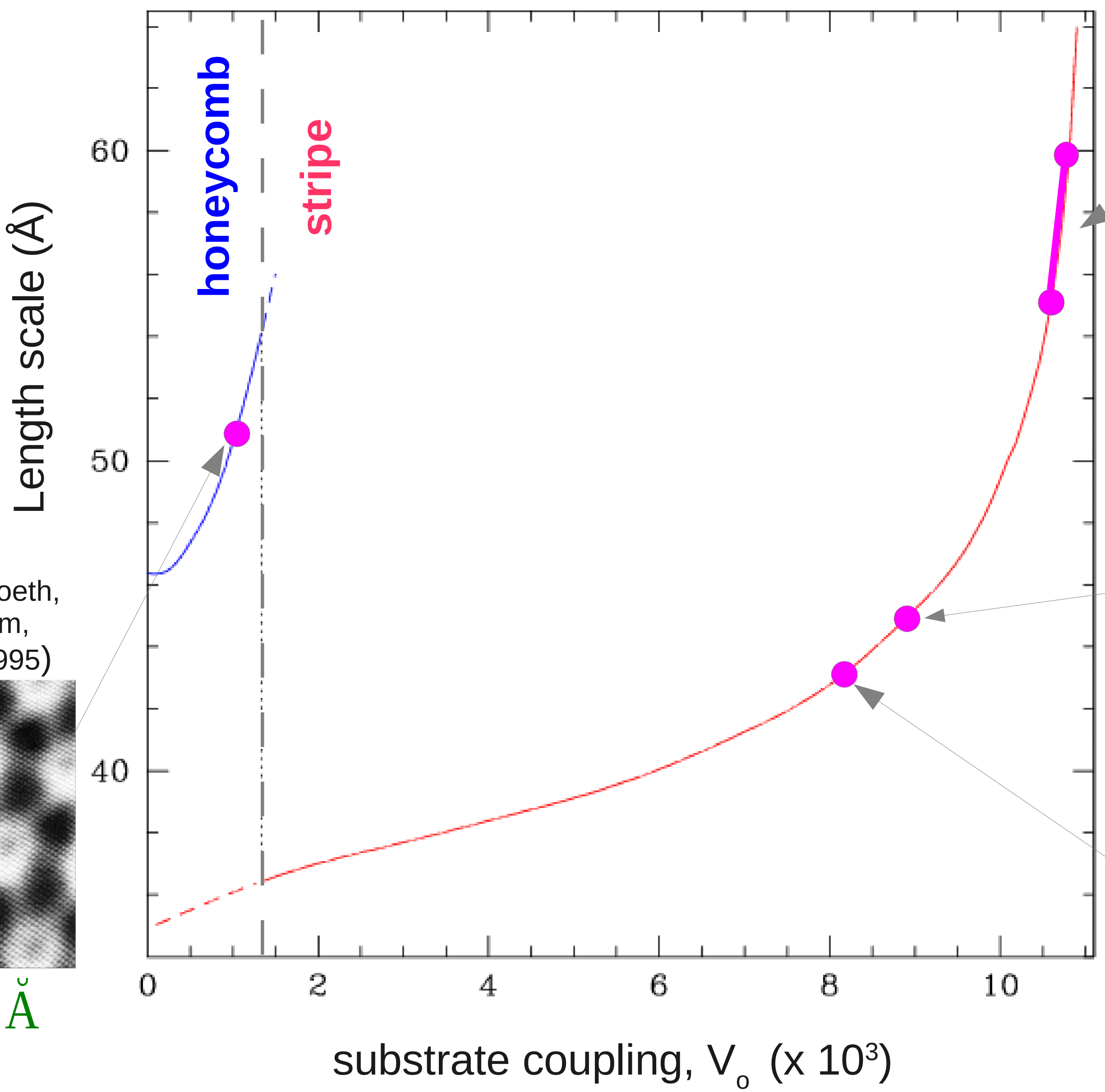
Two monolayers: stripes



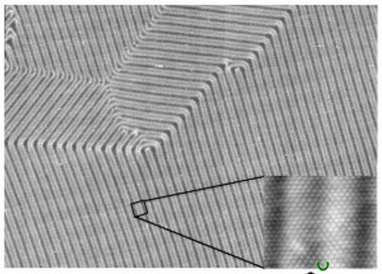
Four monolayers: honeycomb



Monolayer ordering, Length scales, Cu/Ru

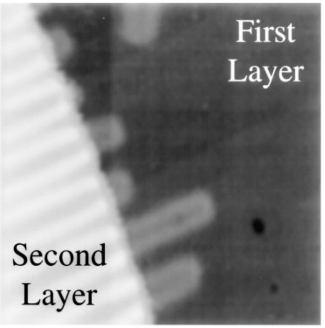


Figuera, Schmid, Bartelt, Pohl, Hwan
PRB **63**, 165431 (2001)



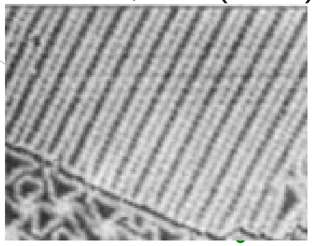
$\lambda \approx 55-60$ Å

Schmid, Bartelt, Hamilton, Carter, Hwang,
PRB **78**, 3507 (1997)



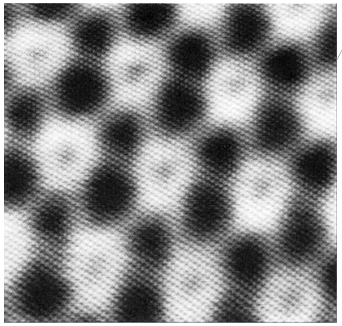
$\lambda \approx 45$ A

Günther, Vrijmoeth, Hwang ,Behm,
PRL **74**, 754 (1995)



$\lambda \approx 43$ Å

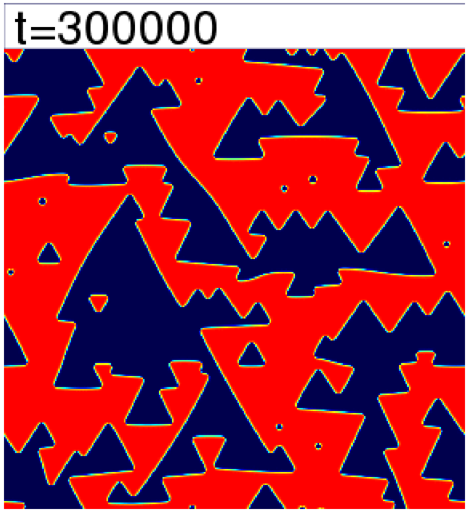
Günther, Vrijmoeth, Hwang ,Behm,
PRL **74**, 754 (1995)

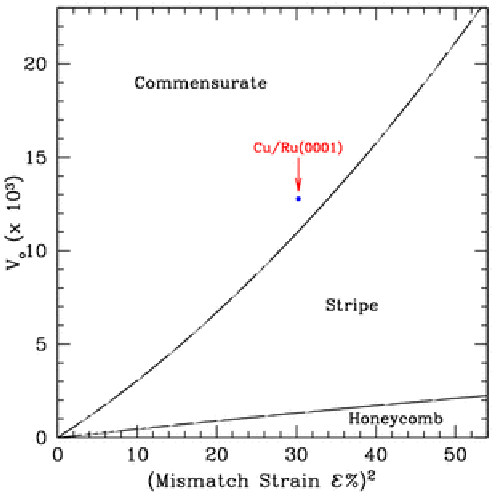


$a \approx 50.7$ Å

Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax in **Striped state**



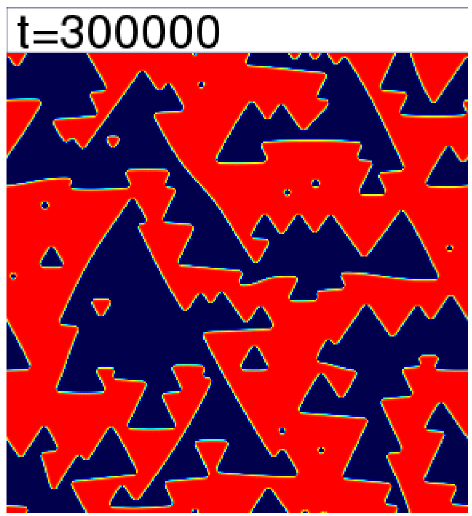


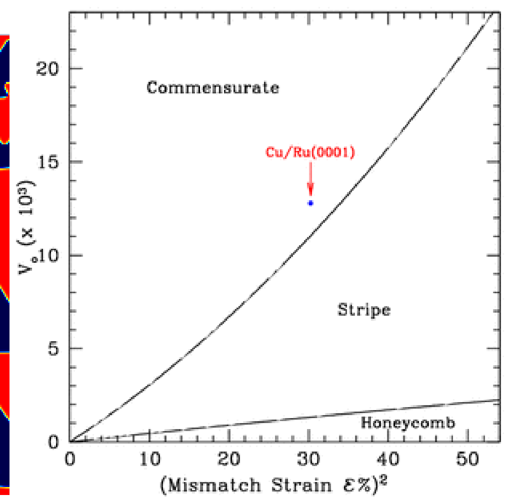
Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax in **Striped state**

Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax near **Stripe/Honeycomb transition**



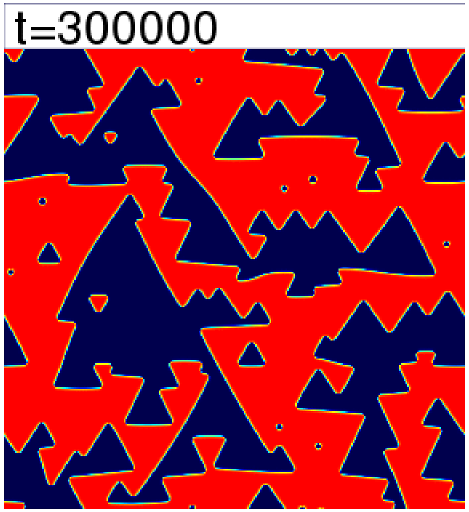


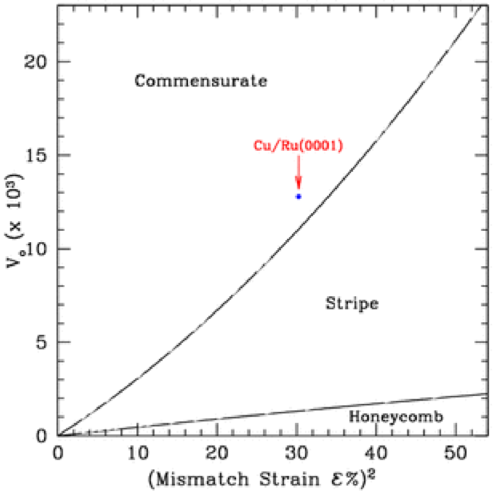
Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax near **Stripe/Honeycomb transition**

Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax in **Honeycomb state**



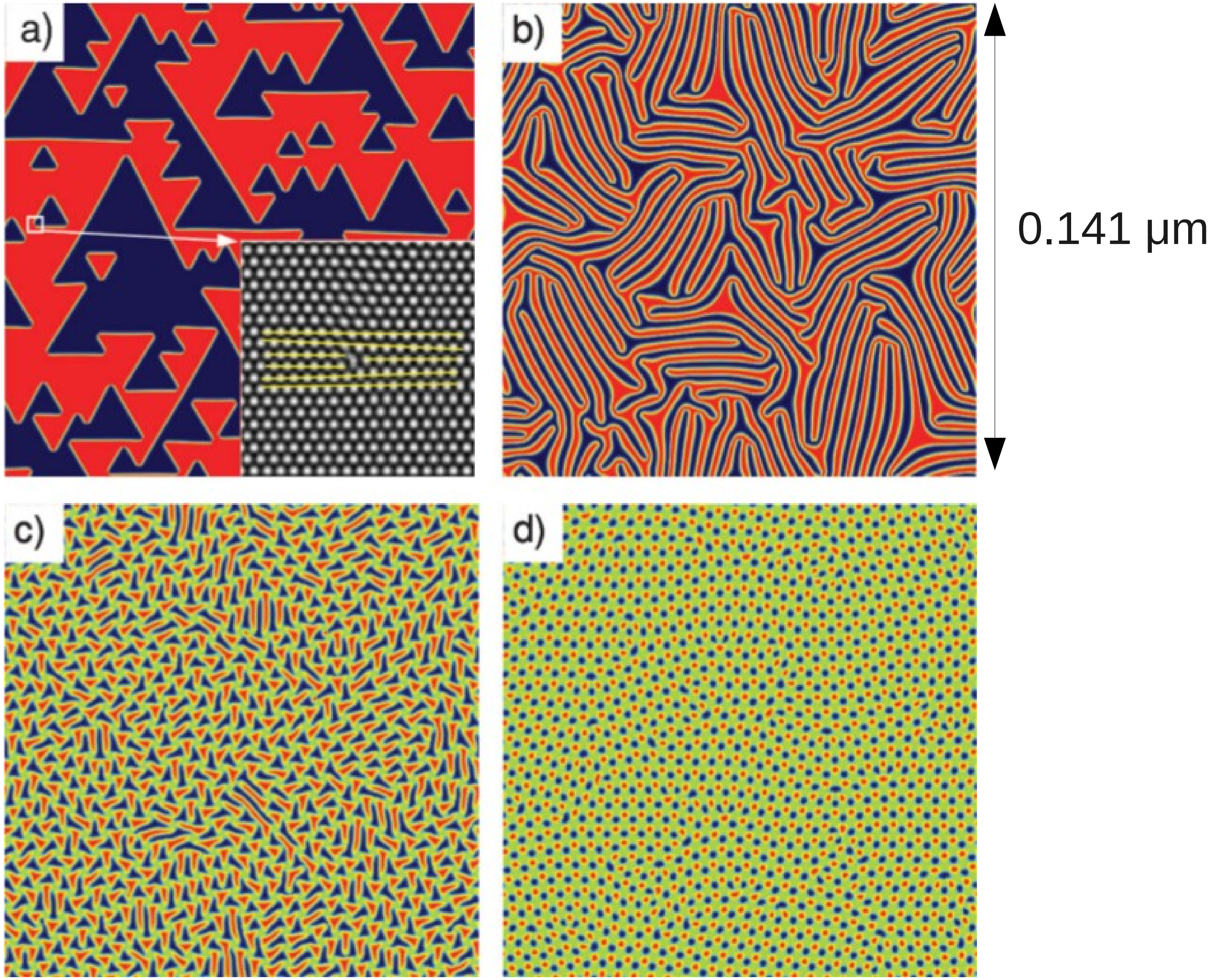


Monolayer ordering, Dynamics: Adding Layers

Add layers, stop/relax in **Honeycomb state**

Monolayer ordering, Dynamics: Adding Layers

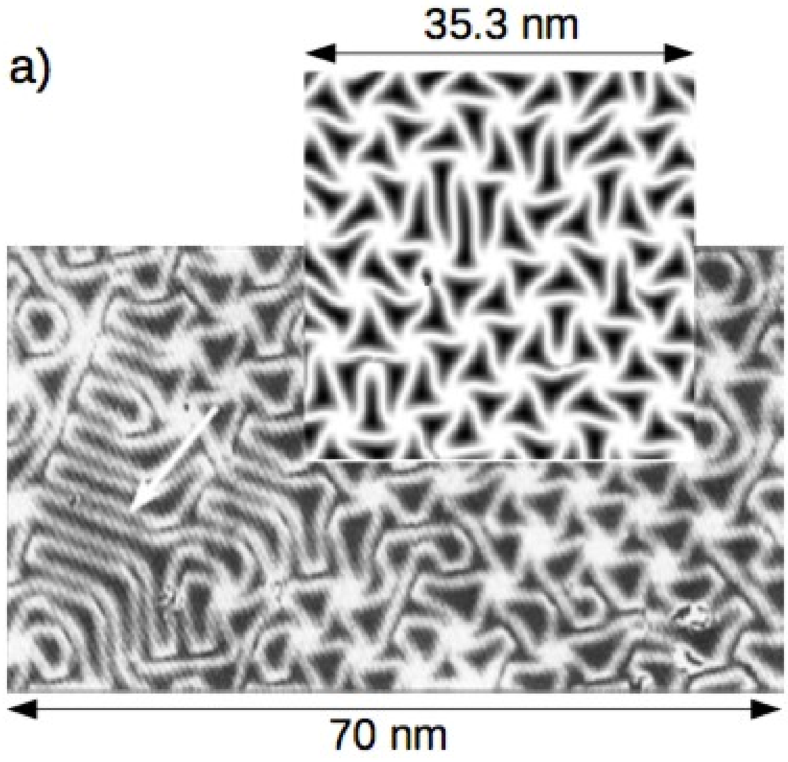
Adding layers, summary



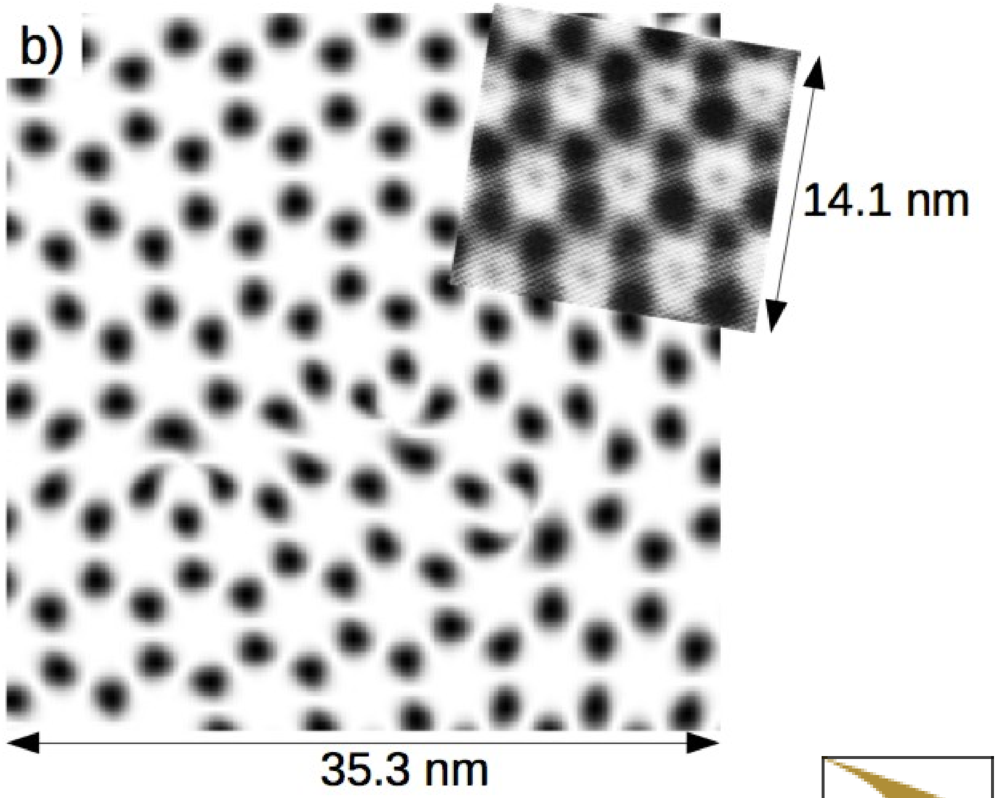
Monolayer ordering, Dynamics: Adding Layers

comparison with experiment

triangular

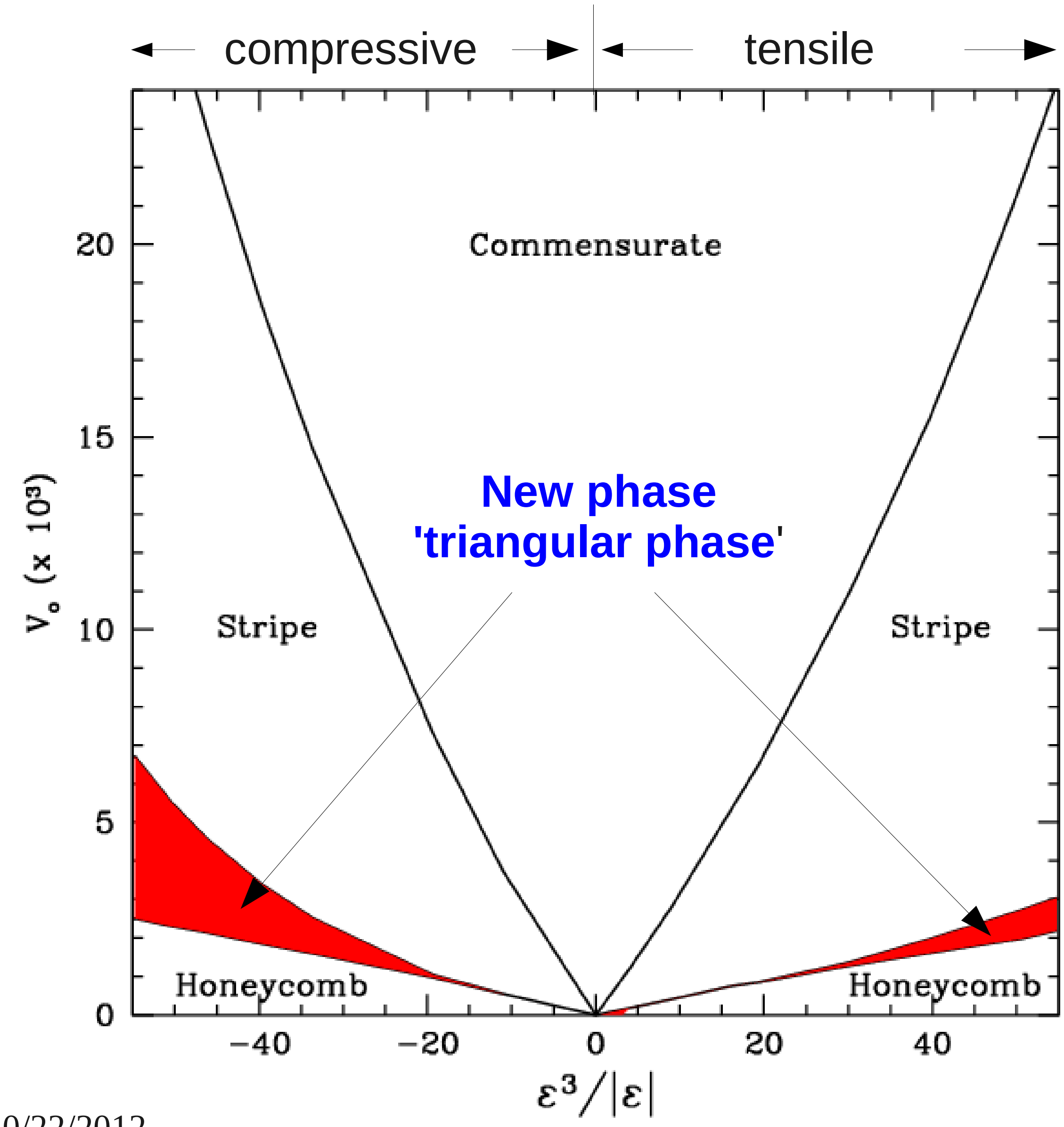


honeycomb



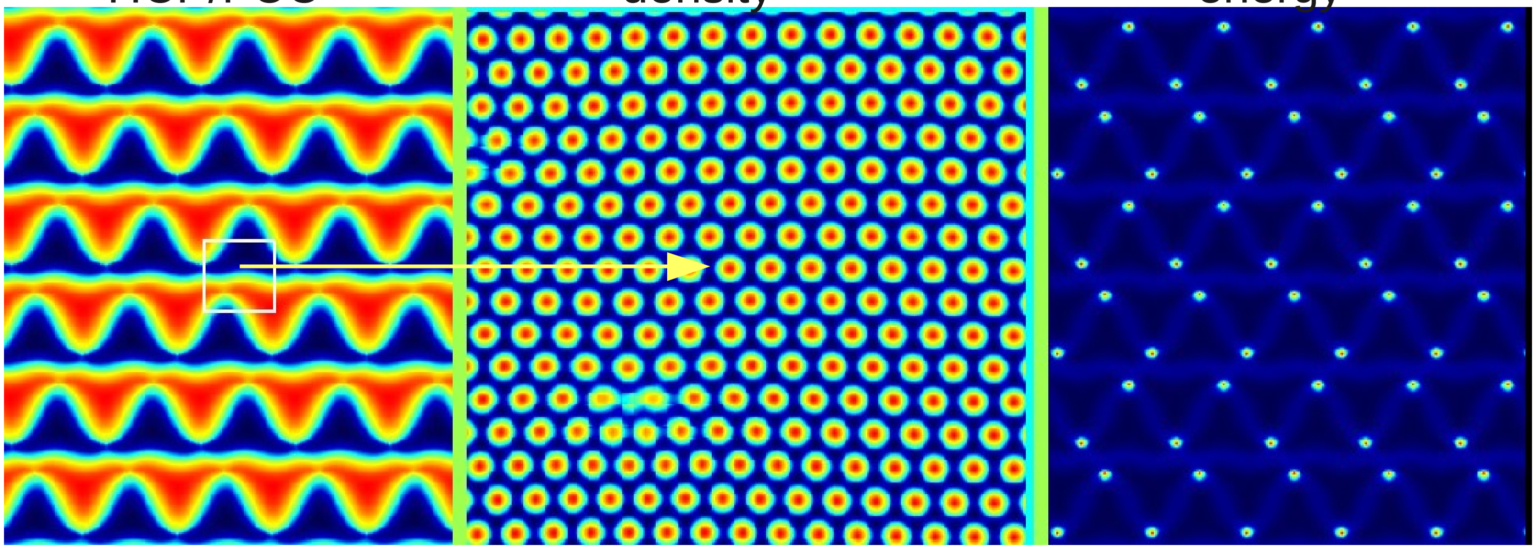
New results:
Compressive case

(tensile PRL, **108**, 226102, 2012)



tensile/compressive asymmetric

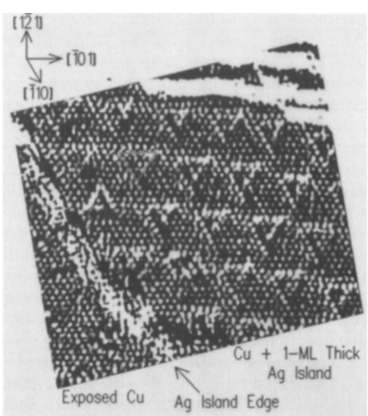
Triangular phase – contains dislocations (unlike other phases)



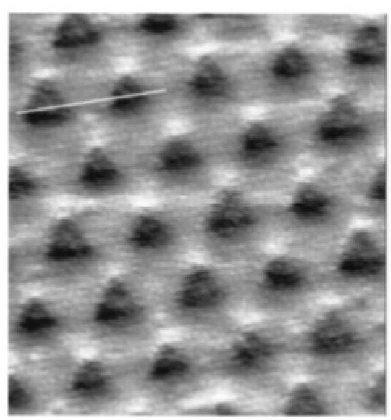
Triangular phase – experiments?

Ag on Cu

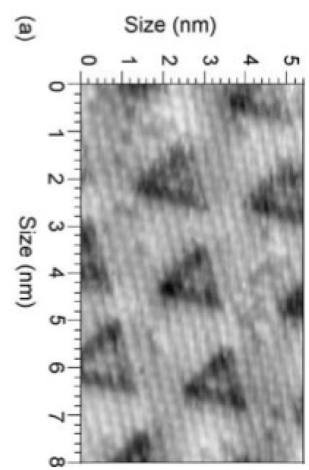
Au on Ni



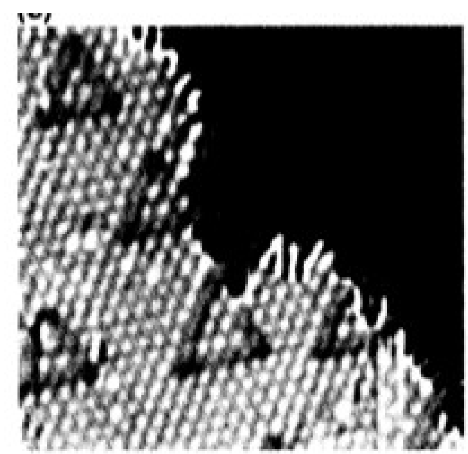
McMahon etal,
Surf Sci Lett, **279**,
L231 (1992)



Umezawa etal,
PRB **63**, 035402 (2000)



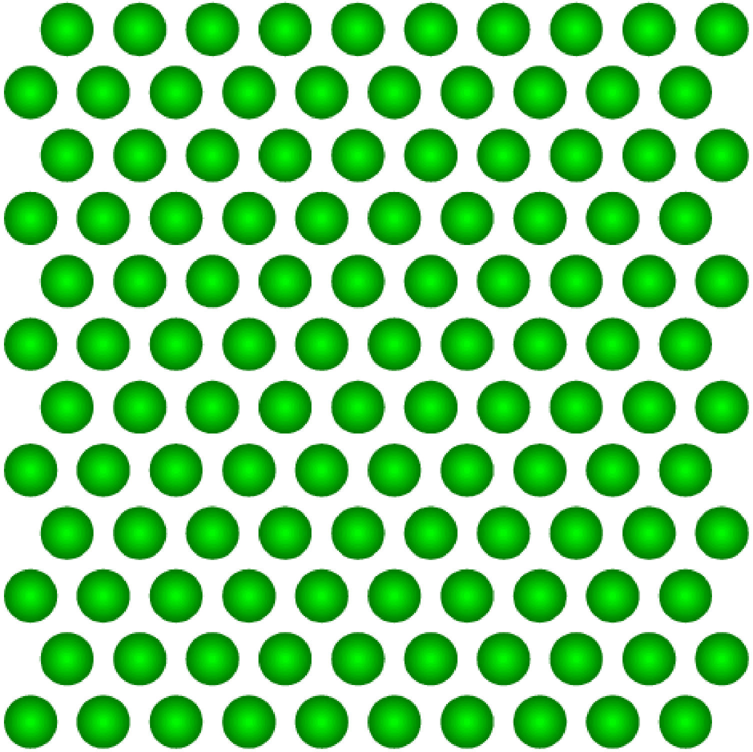
Bendounan etal
App Sur Sci, **212**,
33 (2003)



Jacobsen etal,
PRL, **75**, 489 (1995)

Monolayer ordering

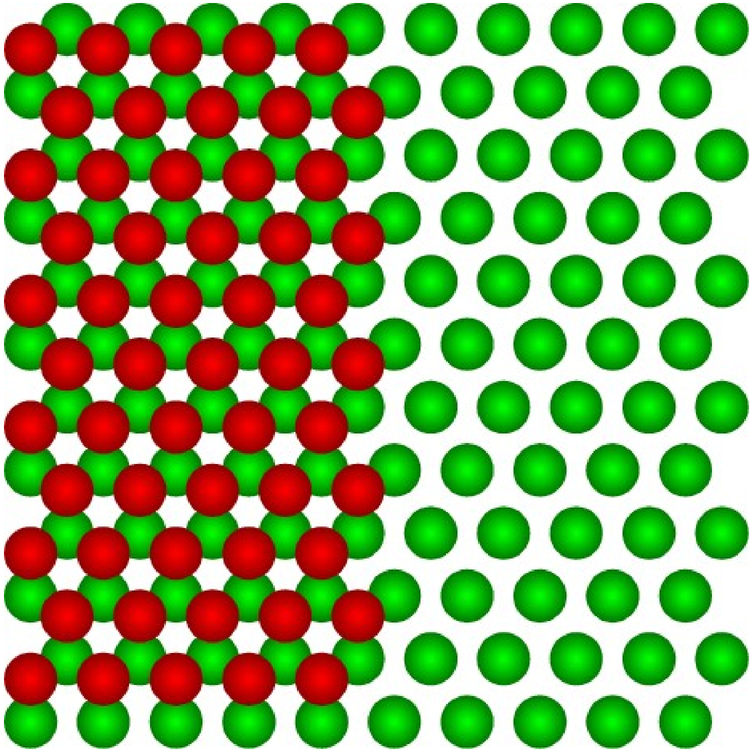
Two dimensional simulations
- no island/mound formation
- stacking ....



Monolayer ordering

three dimensional stacking

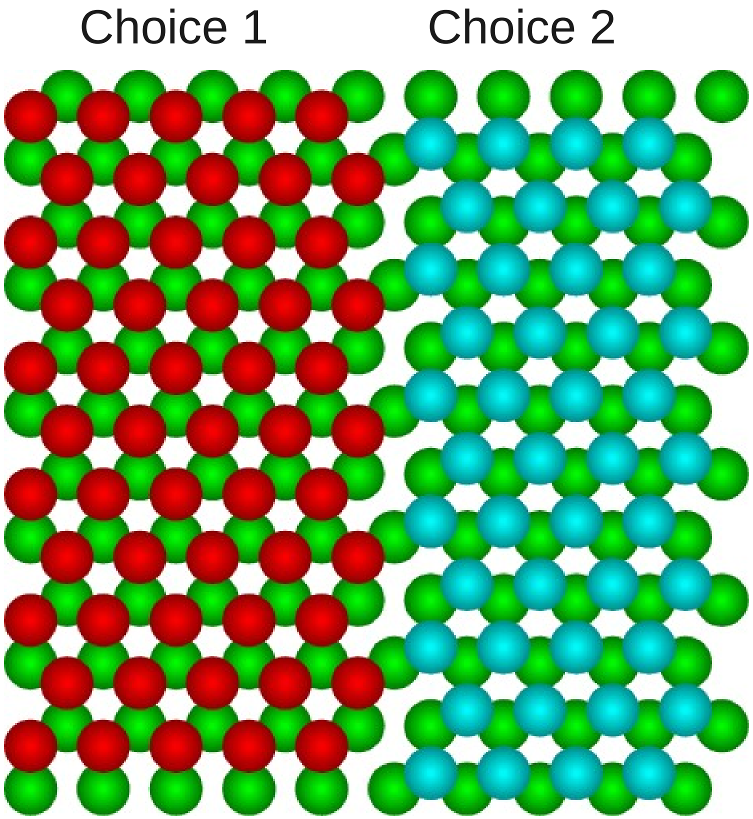
Choice 1



one monolayer
two choices

Monolayer ordering

three dimensional stacking

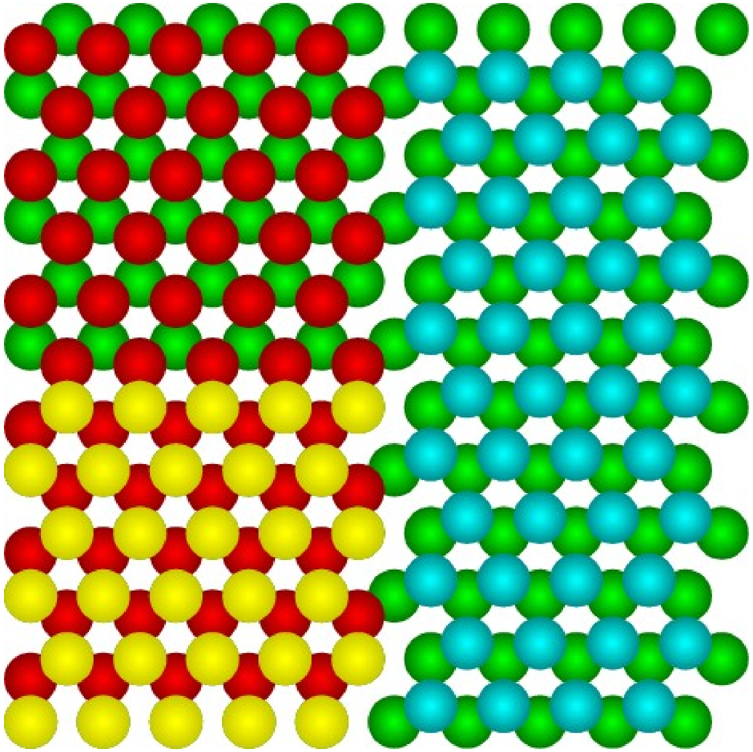


one monolayer
two choices

Monolayer ordering

three dimensional stacking

two monolayers
four choices



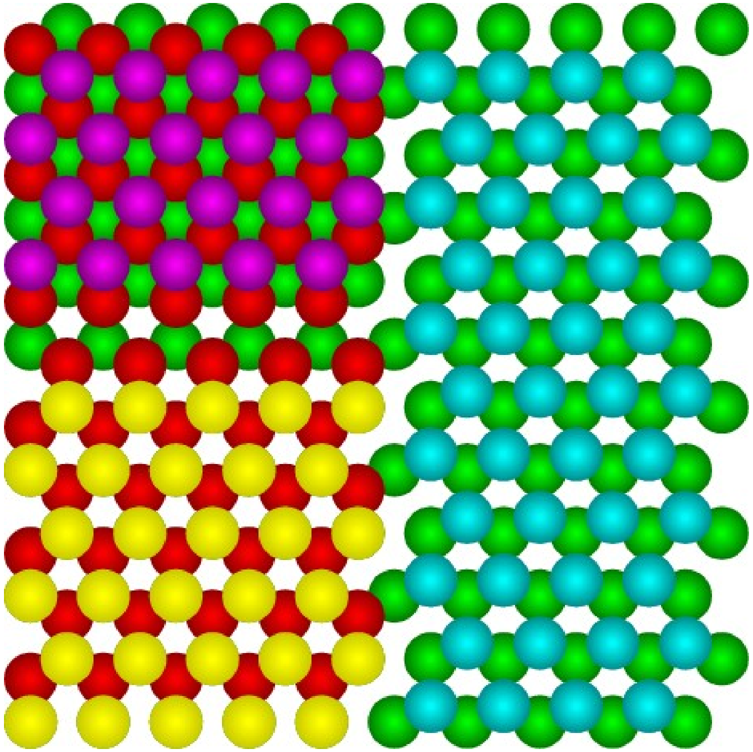
Choice 1

Monolayer ordering

three dimensional stacking

two monolayers
four choices

Choice 2



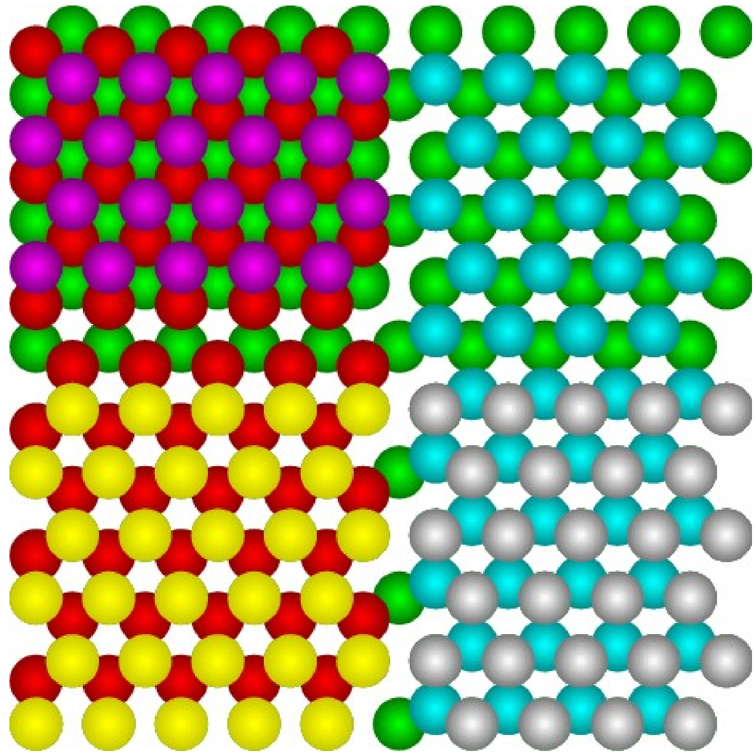
Choice 1

Monolayer ordering

three dimensional stacking

two monolayers
four choices

Choice 2

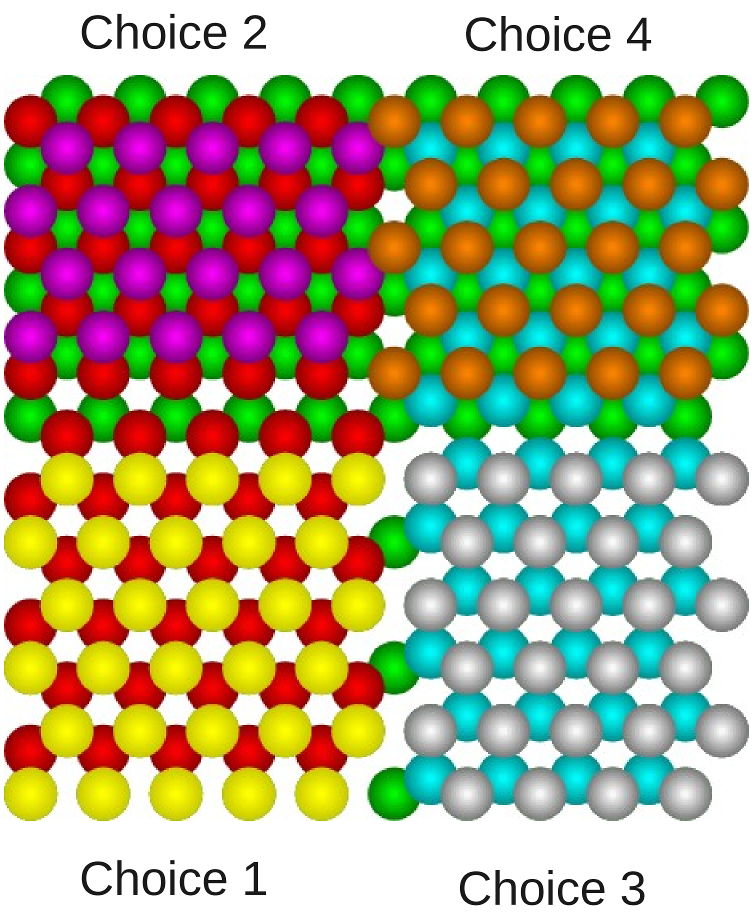


Choice 1 Choice 3

Monolayer ordering

three dimensional stacking

two monolayers
four choices



Final comment

amplitude description as reformulation of continuum elasticity theory

complex amplitudes $\left(\eta_{klm}\right)$ instead of vector, tensor fields $\left(\vec{u}, \vec{\vec{\sigma}}\right)$

$$\eta_{klm} \sim \phi \exp\left(i\theta\right)$$

allows for **phase slips** eg., **dislocations** (and surfaces, cracks, grain boundaries ...)

accounts for **displacements** (and rotations)