

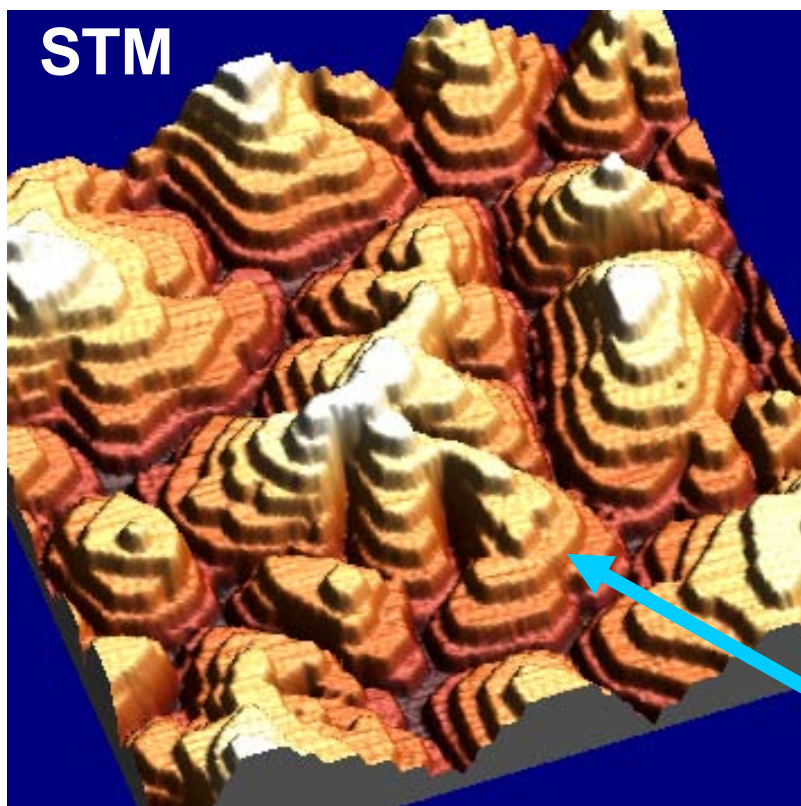
ATOMISTIC AND COARSE-GRAINED MODELING OF EPITAXIAL THIN FILM GROWTH ...INTEGRATED WITH STM EXPT.

Modeling: Yong Han, Maozhi Li¹, Kyle Caspersen², Da-Jiang Liu, Jim Evans

Expt: B. Unal, D. Jing, C. Yuen, T. Duguet, E. Cox, C. Stoldt³, T. Layson, C. Jenks, Patricia Thiel

Iowa State University \$\$\$ NSF Grant CHE-0809472

¹Renmin University, ²Lawrence Livermore NL, ³University of Colorado



Ag/Ag(111) film
3 Monolayers (ML)
Deposition @ 150K
Flux = 0.004 ML/s
STM image size:
125 x 125 nm²

Surface Science Reports **61** (2006) 1-128
J.W. Evans, P.A. Thiel, Maria C. Bartelt
M. Li et al. PRB **77** (2008) 033402; *JPCM* **21** (2009) 084216.

SINGLE-ATOM HIGH STEPS

OUTLINE

Basic concepts and modeling strategies:

Atomistic processes underlying epitaxial growth (LG models).

Far-from-equilibrium growth requires accurate description of kinetics

Modeling strategies: atomistic vs. step-dynamics vs. 3D continuum

Successes for predictive atomistic modeling:

Homoepitaxy (A on A): Ag/Ag(111); Ag/Ag(100).

Heteroepitaxy (A on B; A on BC): Ag/NiAl(110)

Homoepitaxy of alloys (A+B on AB): Ni + Al on NiAl(110)

Fundamental open issues for theory & modeling

Submonolayer island formation:

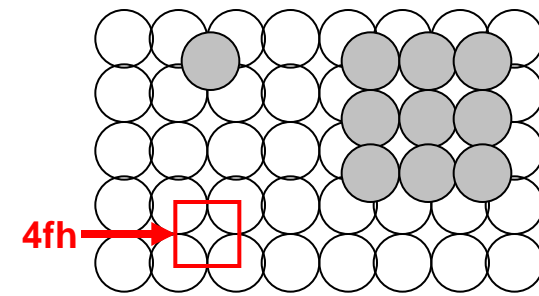
analytic theory, step dynamics and other modeling strategies

Multilayer kinetic roughening:

step dynamics modeling + coarse-graining to continuum PDE's

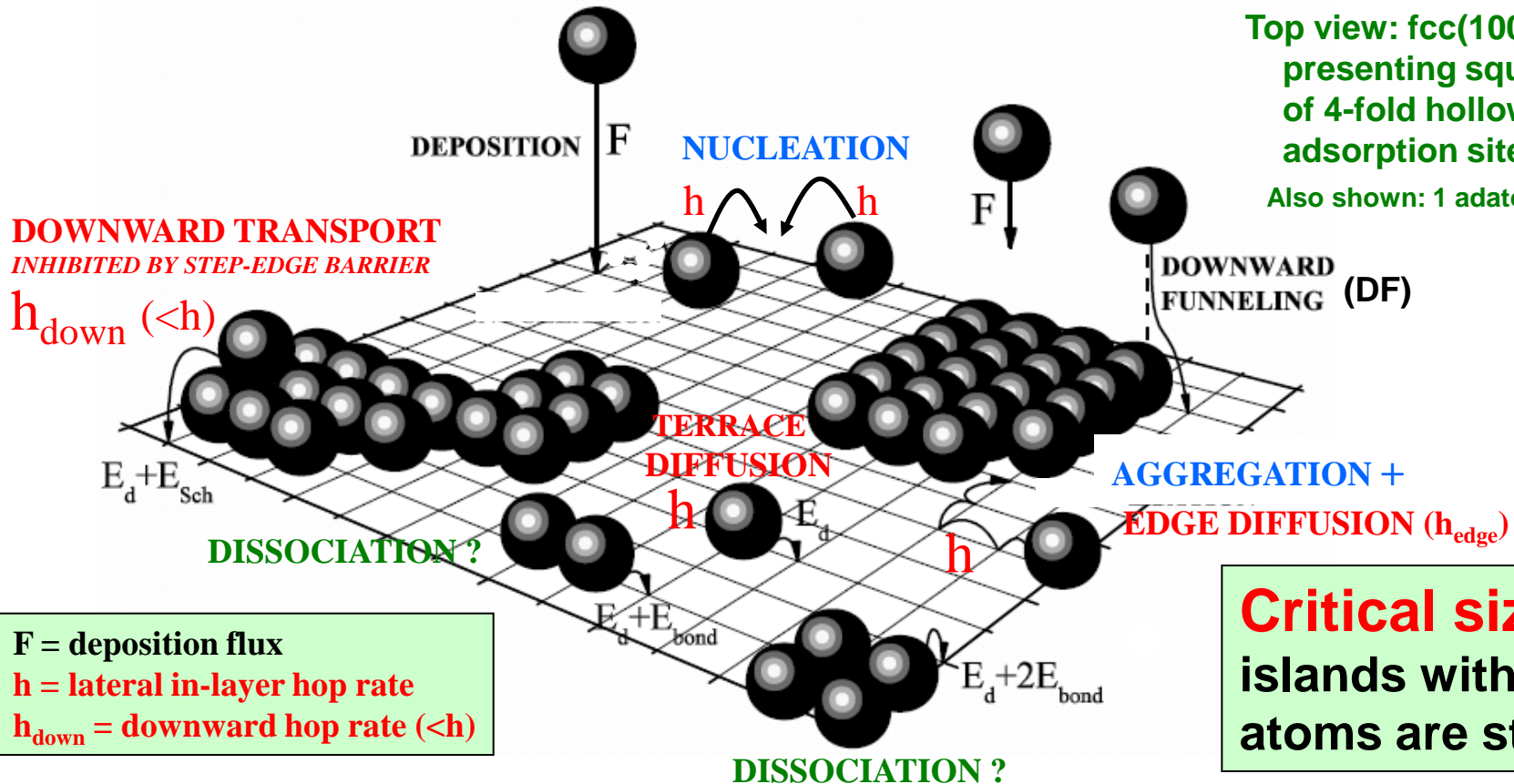
ATOMISTIC PICTURE OF EPITAXIAL FILM GROWTH AND OF LG MODELING

crystalline surface = period lattice of adsorption sites on which ad-atoms reside
 ⇒ LATTICE-GAS (LG) MODEL ...or discrete *Interacting Particle System*



Top view: fcc(100) surface presenting square array of 4-fold hollow (4fh) adsorption sites

Also shown: 1 adatom; 1 island



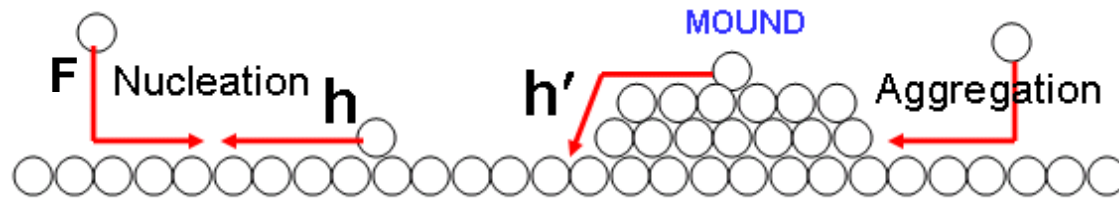
F = deposition flux
 h = lateral in-layer hop rate
 h_{down} = downward hop rate ($< h$)

Critical size i :
 islands with $> i$
 atoms are stable

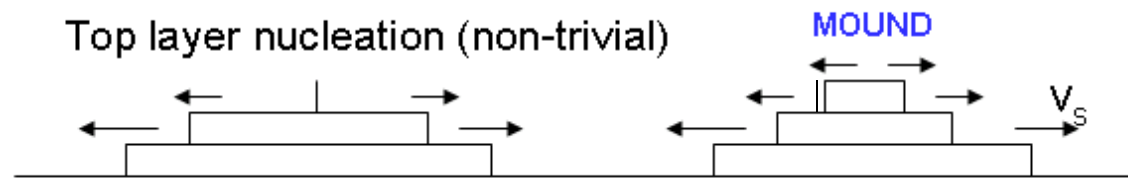
Arrhenius form of rates for hopping:
 $h = \nu \exp[-E_{\text{act}}/(kT)]$ etc E_{act} = activation barrier
 $\nu \approx 10^{12} \text{ s}^{-1}$ attempt frequency

KMC simulation:
 Stochastic implementation of processes with probabilities proportional to their rates

DIFFERENT MODELING FORMULATIONS

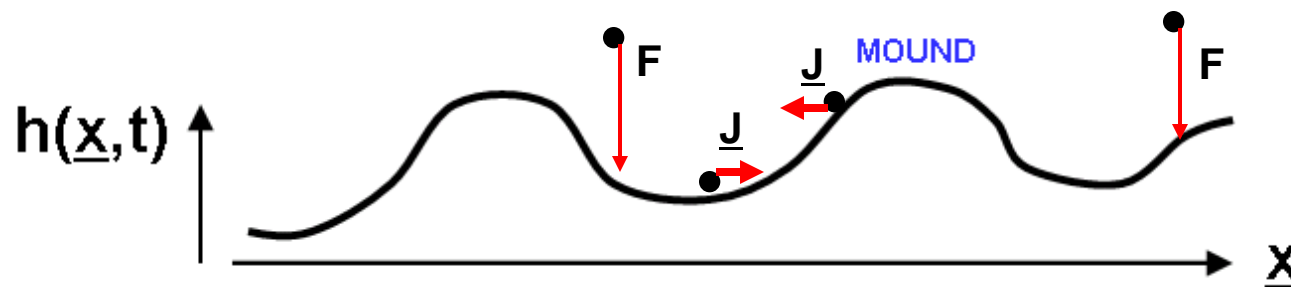


ATOMISTIC
(LATTICE-GAS MODELS)



2D CONTINUUM
(STEP-DYNAMICS MODELS)

Step velocities, v_s , from BCF. Topological complexity treated via level set or phase field.



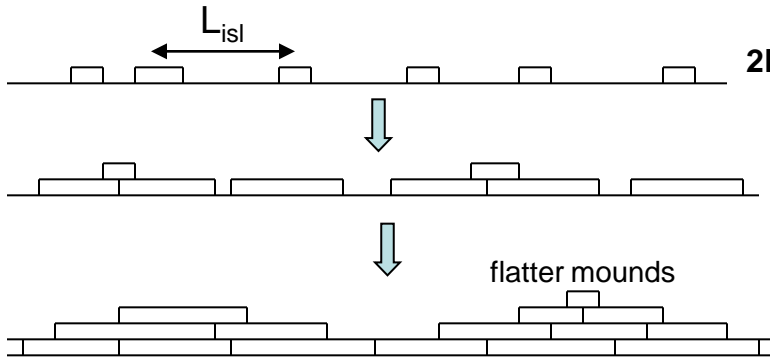
3D CONTINUUM
 $\partial h / \partial t = F - \nabla \cdot \underline{J} + \eta$
noise

HOMOEPI TAXY (A on A): UNSTABLE GROWTH = MOUNDING

INSTABILITY DUE TO AN EHRLICH-SCHWOEBEL (ES) BARRIER = STEP EDGE BARRIER (δ_{ES})

Michely & Krug, Islands, Mounds, and Atoms (Springer 2004) *JE, Thiel, Bartelt, Surf. Sci. Rep. 61 (2006)*

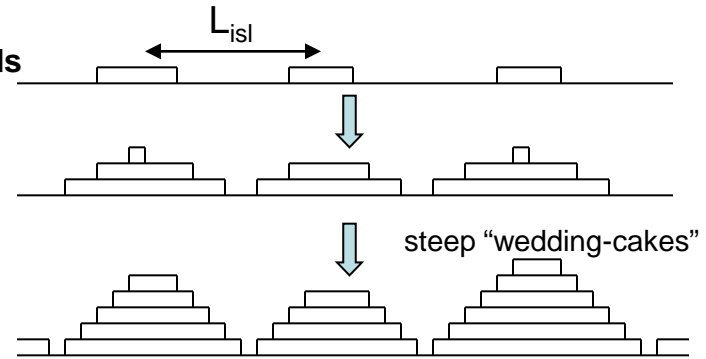
SMALL ES BARRIER ($L_{ES} \ll L_{isl}$)



2D islands

slope selection & significant mound coarsening

LARGE ES BARRIER ($L_{ES} \gg L_{isl}$)

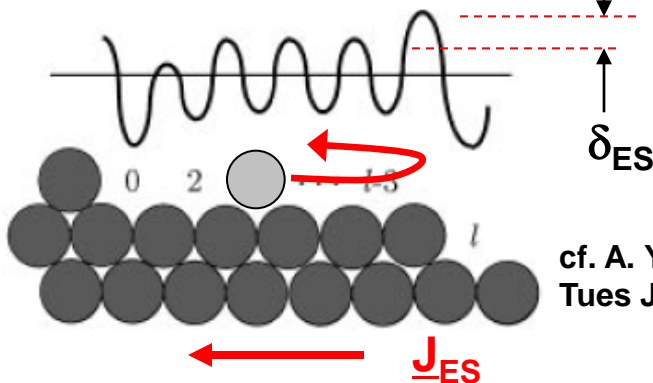


dramatic steepening & slow mound coarsening

$$L_{ES} \sim e^{\beta \delta_{ES}}$$

Growth instability due to ES barrier

Jacques Villain, J. Phys. I (1991)

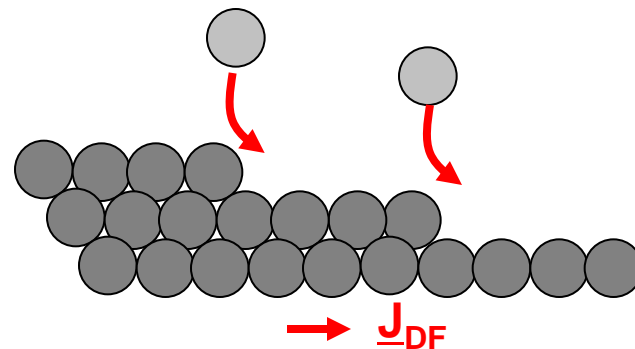


cf. A. Yagi
Tues July 27

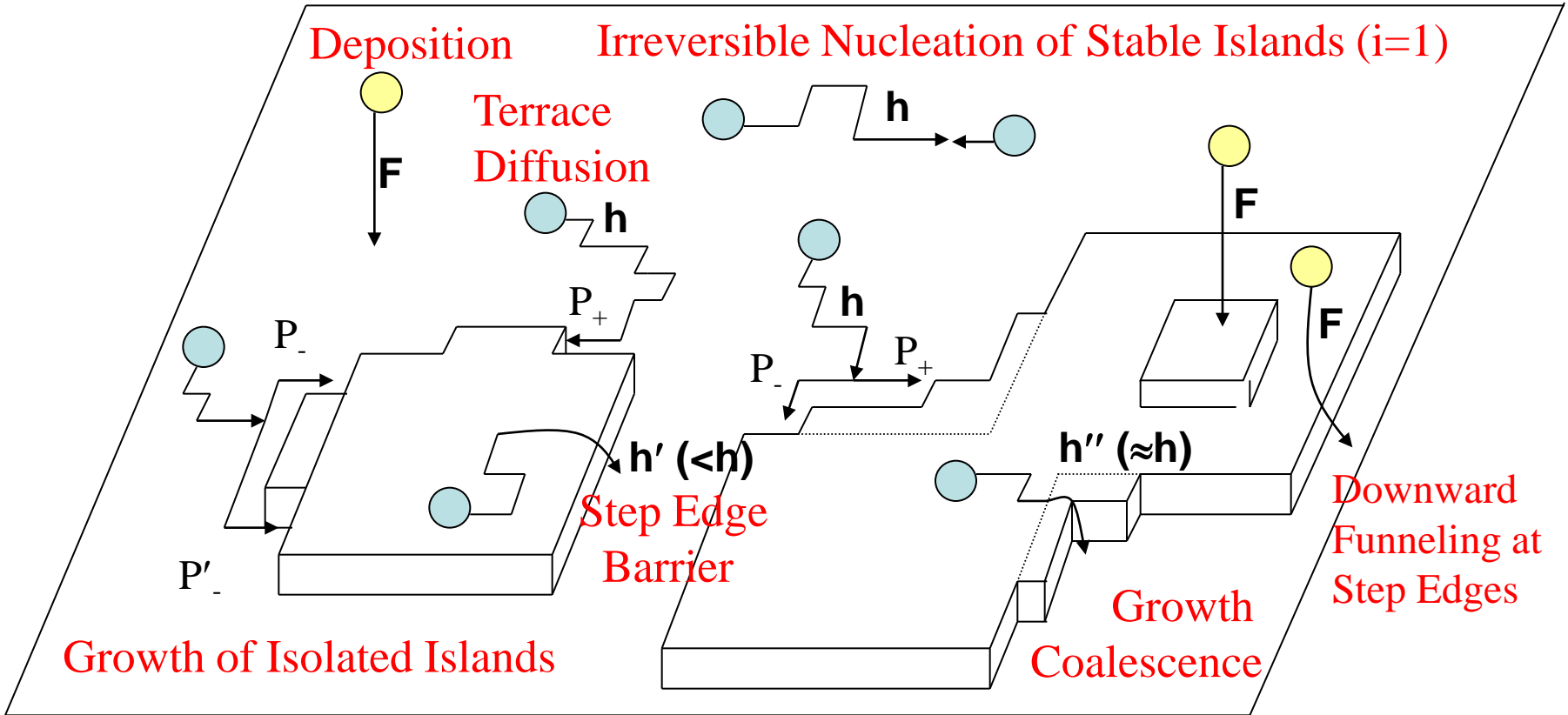
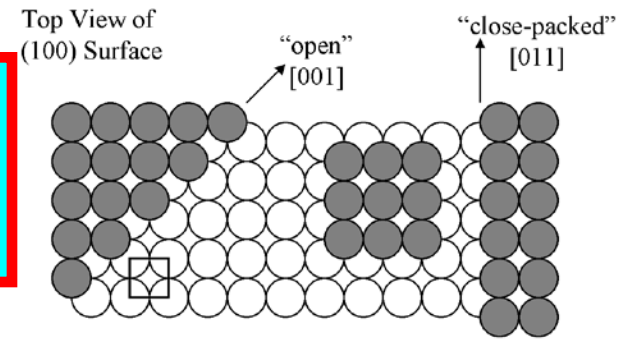
Downward funneling (DF) produces smooth growth at low T

and counterbalances the effect of ES barrier at higher T

JE, Sanders, Thiel, DePristo, Phys. Rev. B (1990)

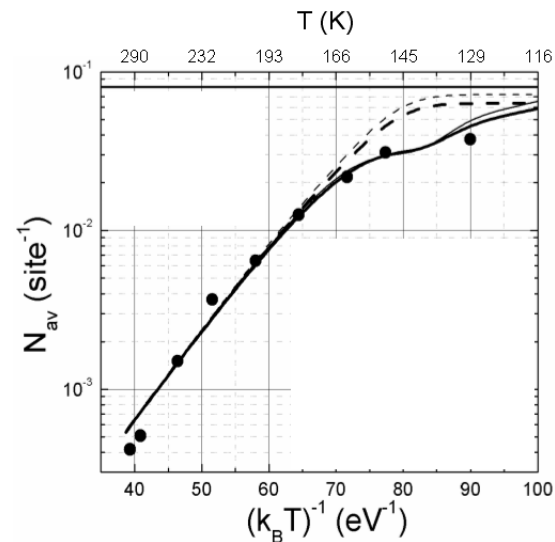
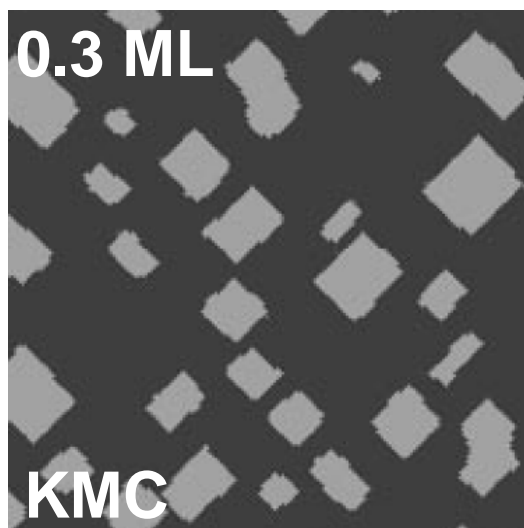
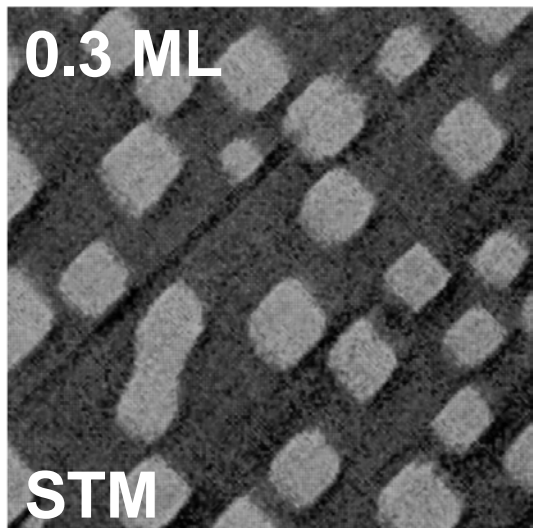


ATOMISTIC MODELING of Ag/Ag(100) GROWTH
...PROTOTYPE FOR SMOOTH GROWTH @ 300 K
DUE TO SMALL ES BARRIER (also *FAST EDGE DIFF^N*)

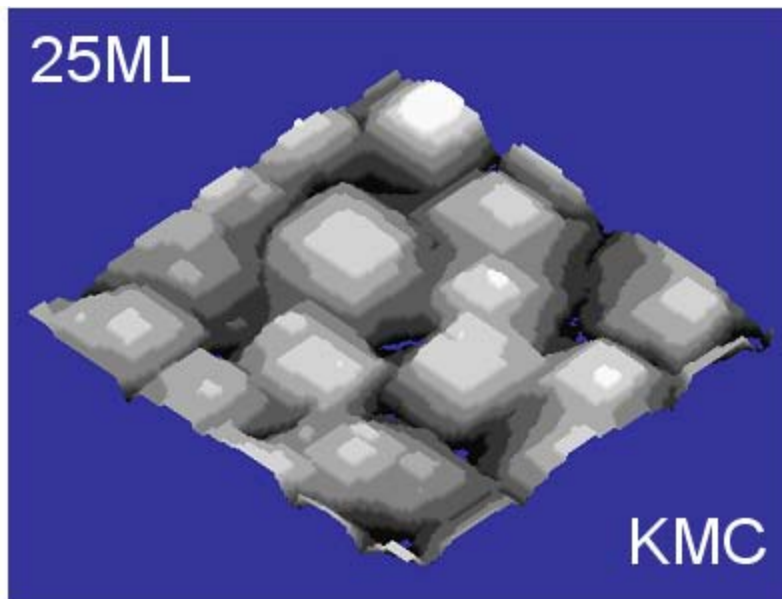
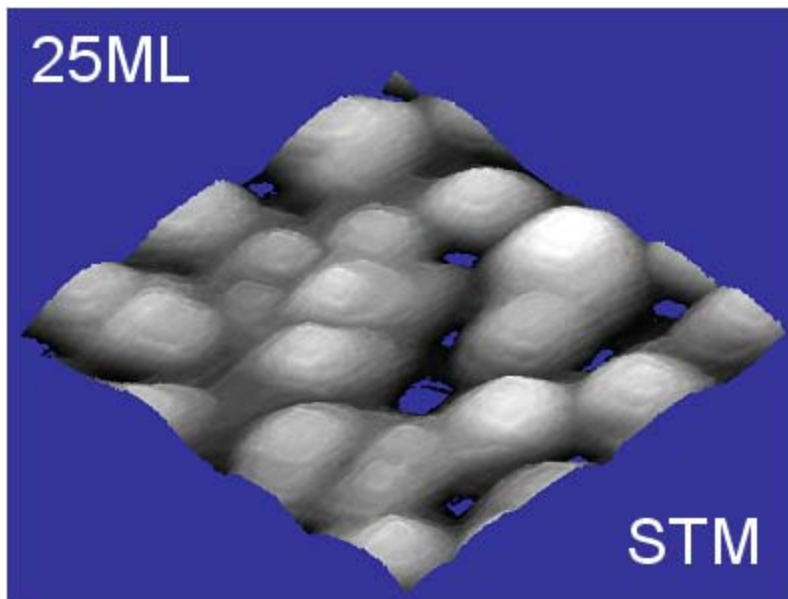


Metal(100) homoepitaxy: $i=1$ (irreversible island formation) typical for lower T; $i=3$ (stable tetramers) for some higher T

SUBMONOLAYER ISLAND FORMATION AT 300 K ($50 \times 50 \text{ nm}^2$) $E_d = 0.43 \text{ eV}$



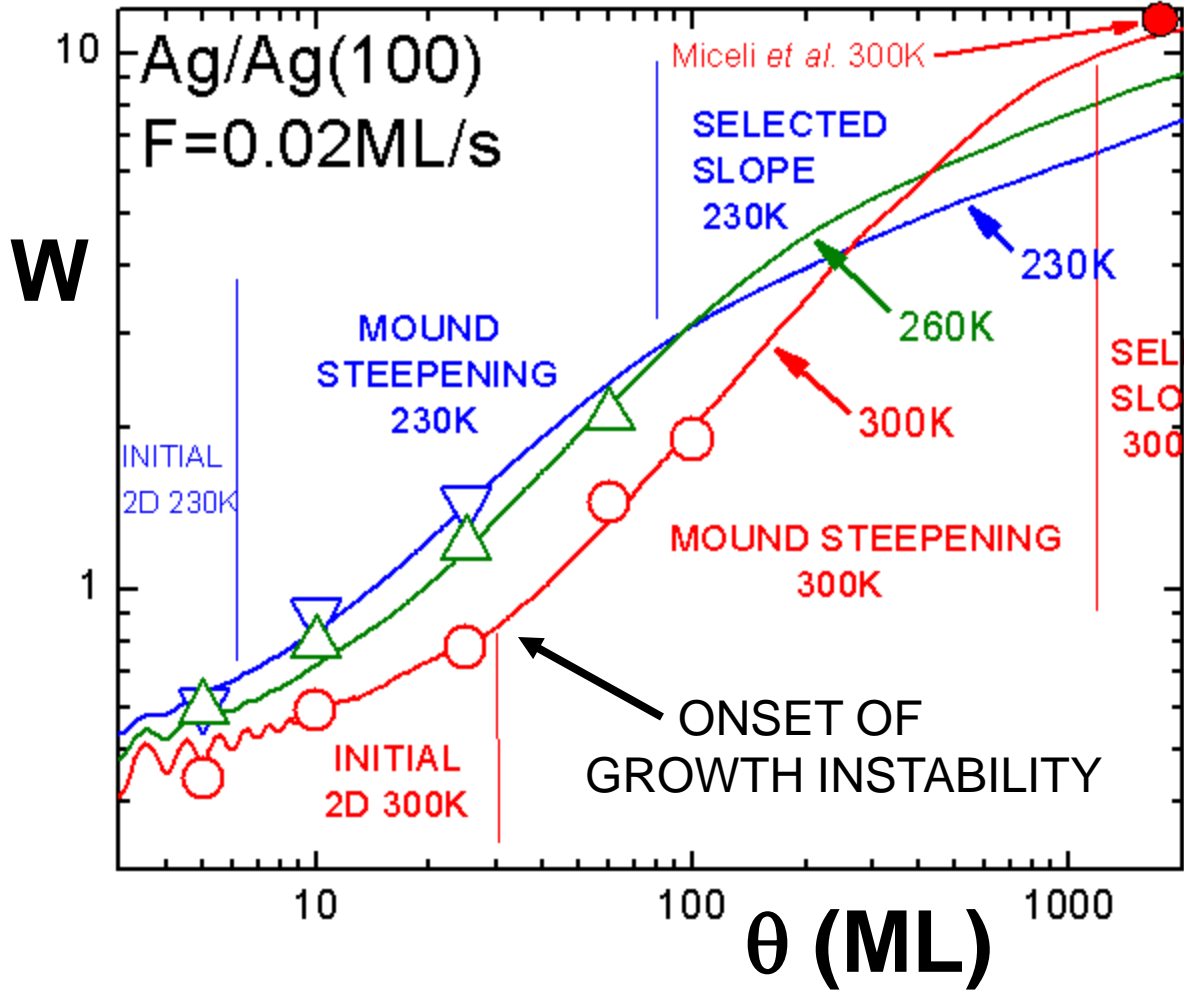
MULTILAYER MOUND FORMATION AT 230 K ($50 \times 50 \text{ nm}^2$) $\delta_{ES} = 0.07 \text{ eV}$



Ag/Ag(100) @ 300K: UNEXPECTED VERY ROUGH GROWTH FOR >30ML

Film Roughness W (in units of interlayer spacing)

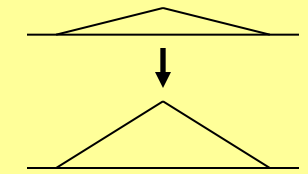
Elliott, Miceli, et al. PRB 54 (1996)
Surface X-ray Scattering Study



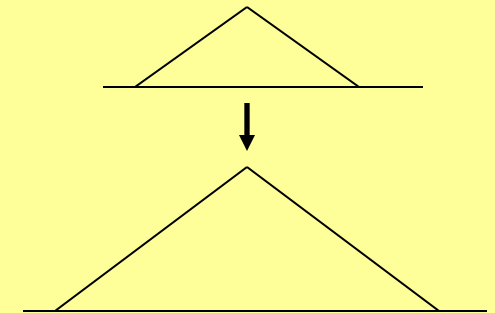
GROWTH REGIMES:

(i) INITIAL QUASI-LBL
...expected behavior

(ii) MOUNDS FORM &
SIDES STEEPEN



(iii) SELECTED SLOPE /
SHAPE + COARSENING



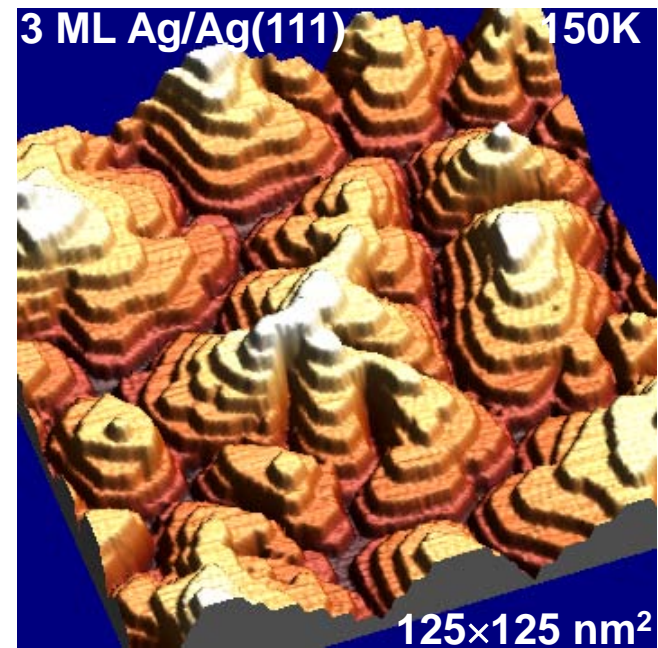
Modeling with small non-uniform step-edge barrier ($E_{act} = 0.07\text{eV}$)
Symbols = experimental data (STM); Curves = model predictions
Caspersen, Layson, Stoldt, Fournie, Thiel, JE, PRB 65 (2002) 193407

ATOMISTIC MODELING OF WEDDING-CAKE MORPHOLOGIES IN AG/AG(111) GROWTH ...PROTOTYPE FOR A LARGE ES-BARRIER

Maozhi Li, C.Chung, E.Cox, C.Jenks, P.Thiel, J.Evans

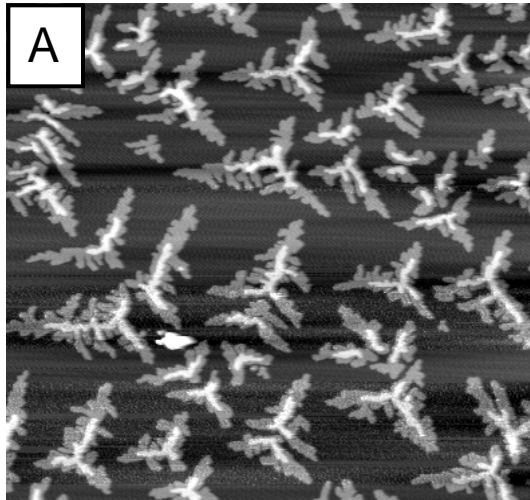
Phys. Rev. B 77 (2008) 033402; Surf. Sci. Rep. 61 (2006) 1

Goal: Develop predictive atomistic LG model for unstable growth in classic M/M(111) systems with large ES barrier:
(i) extract key energetics; (ii) predict morphologies versus T

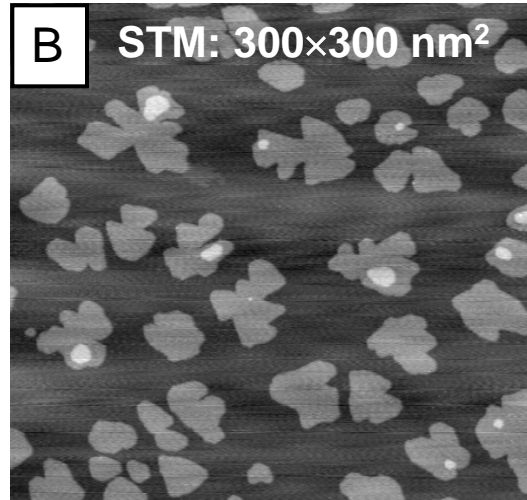


Step 1: describe submonolayer island shapes (slow edge diffⁿ, anisotropic corner diffⁿ)
Step 2: treatment of interlayer transport (non-uniformity and magnitude of ES barrier)

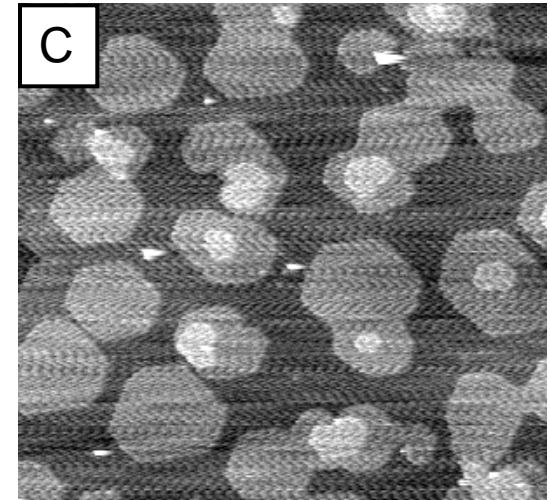
Cox et al. (ISU) PRB 71 (05)



135 K: Δ DENDRITES

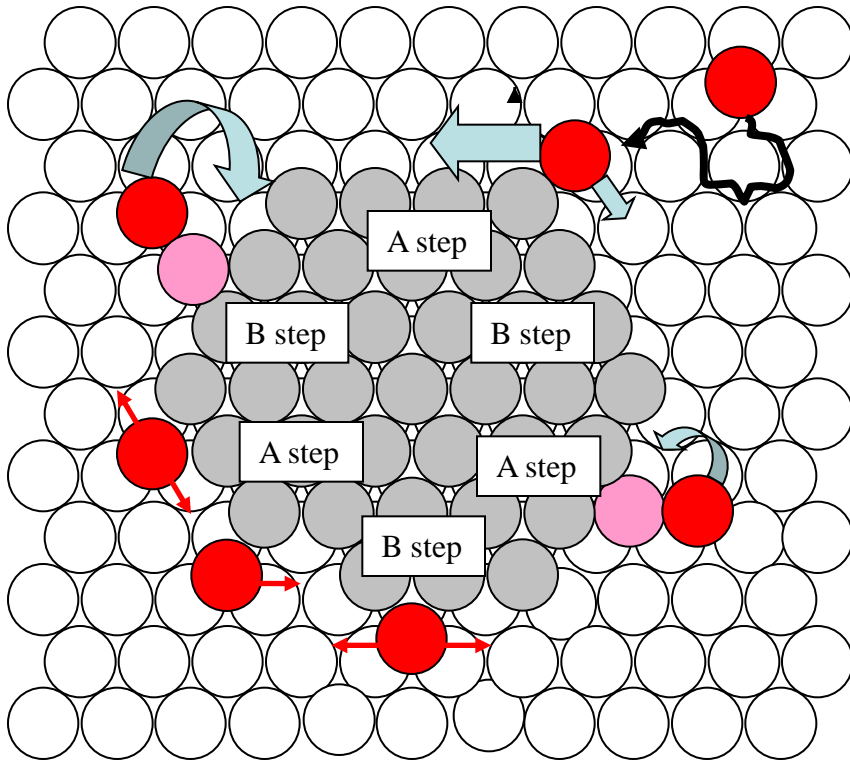


165K: FAT FRACTALS



200K: DISTORTED HEX.

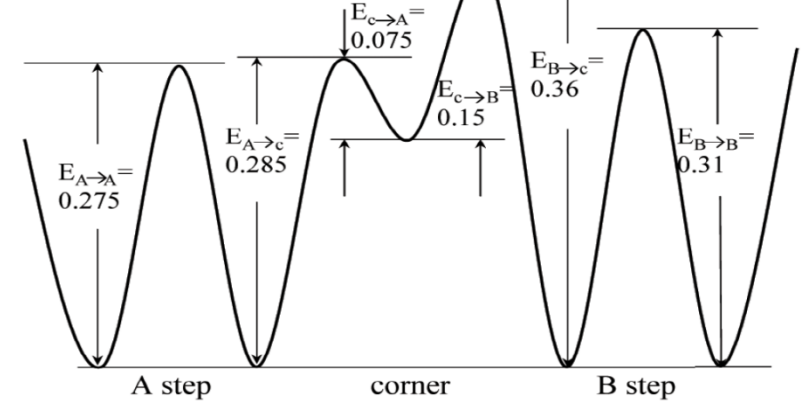
Ag/Ag(111): ATOMISTIC MODEL FOR ISLAND GROWTH SHAPES



CORNER DIFFUSION ANISOTROPY

cf. Hohage et al. Pt/Pt(111) PRL 76 (96); Ovesson et al. Al/Al(111) PRL 83 (99); Brune et al. Ag/Pt(111) SS 349 (96)

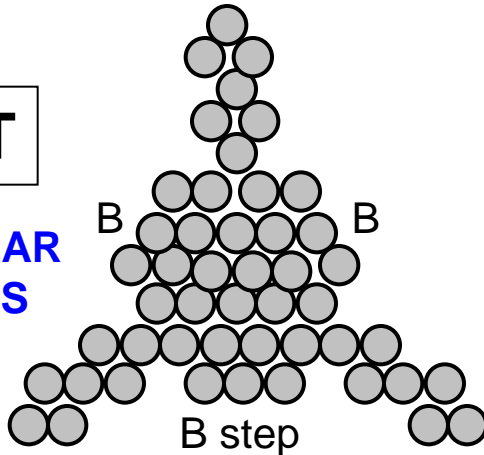
EAM PES



A- and B-steps energetically equivalent
 \Rightarrow hexagonal equilibrium shapes

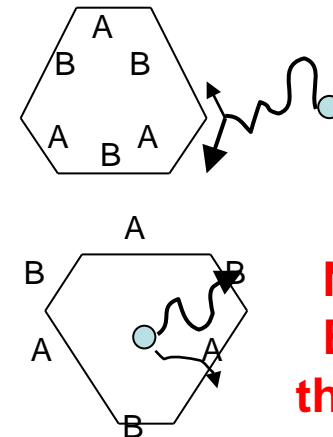
LOW T

TRIANGULAR DENDRITES



HIGH T

DISTORTED HEXAGONS

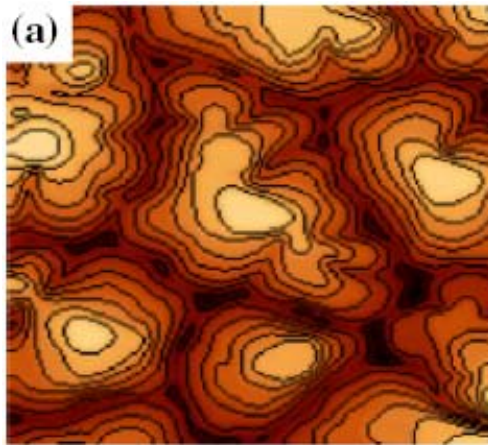


Corner Diffusion Anisotropy

Non-uniform ES Barrier... this effect wins

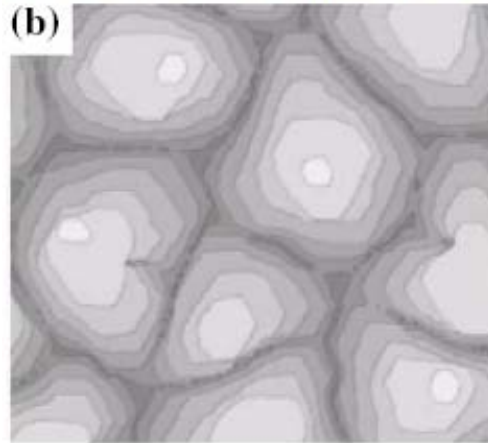
3ML Ag/Ag(111)
F=0.003ML/s
T=150K

STM image
86×86 nm²



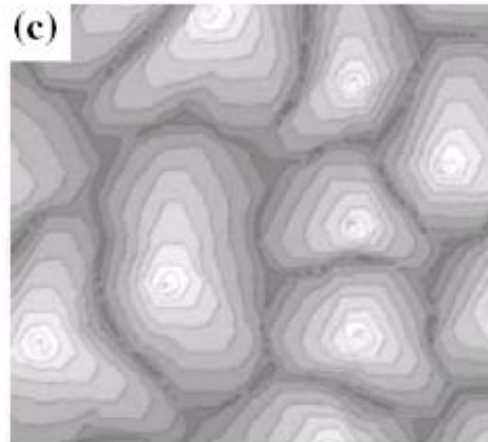
KMC simulation
ES barrier
=0.08 eV (B step)
=2×0.08 eV (A step)

...need non-uniform
step-edge barrier



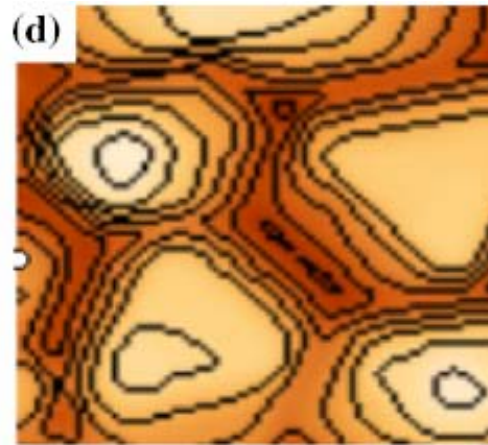
KMC simulation
ES barrier = ∞

Must accurately
treat nucleation
(i=2) for consistent
results at 150+180K

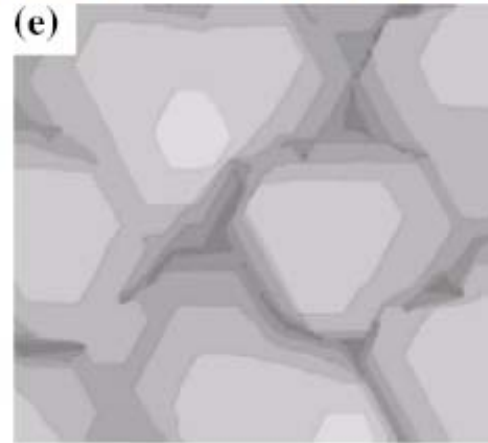


3ML Ag/Ag(111)
F=0.003ML/s
T=180K

STM image
140×140 nm²

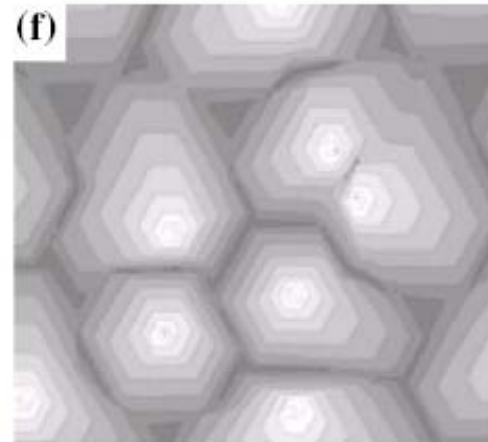


KMC simulation
ES barrier
=0.08 eV (B step)
=2×0.08 eV (A step)



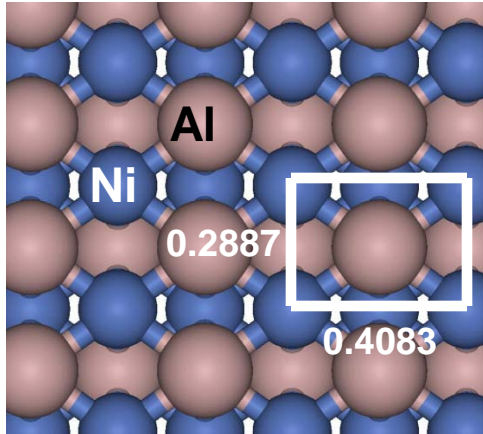
KMC simulation
ES barrier = ∞

cf. analytic work
by Krug, Politi,
Maass, et al.

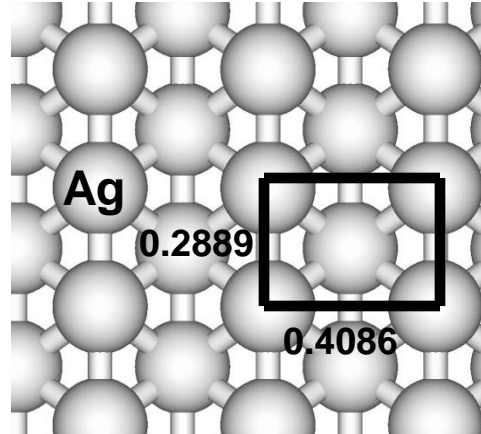


Ag on NiAl(110): BILAYER GROWTH DRIVEN BY QSE

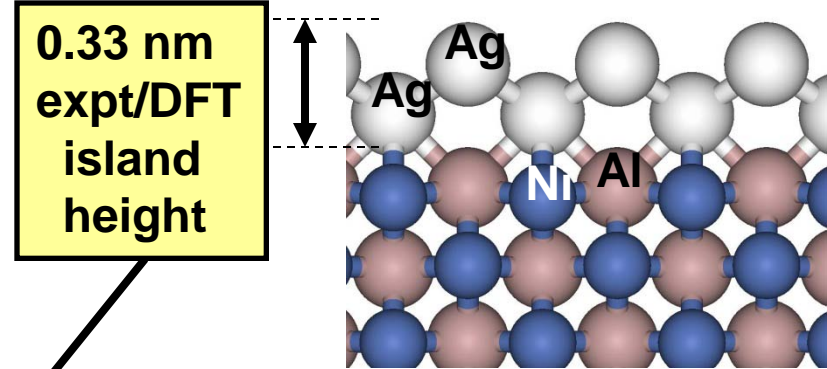
Top view: NiAl(110)



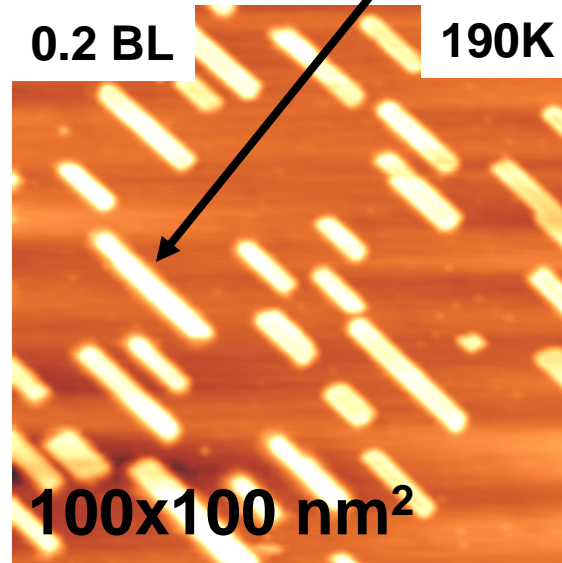
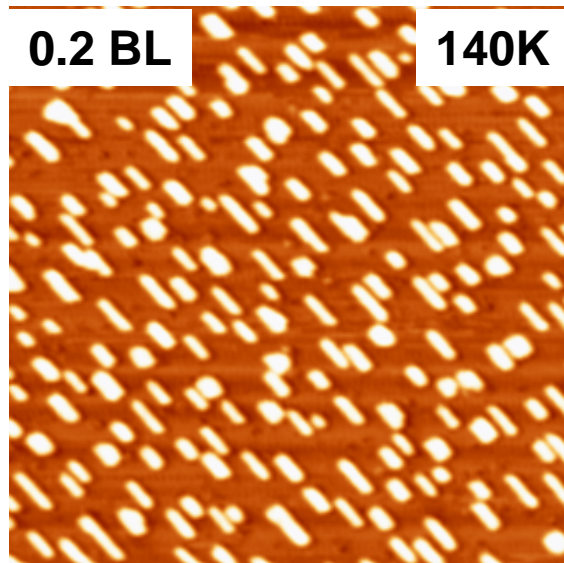
Top view: Ag(110)



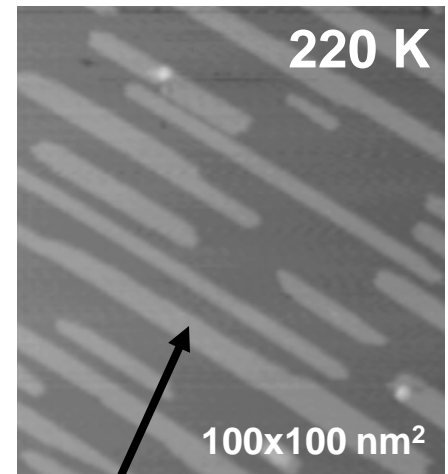
Side view: Ag(110) bilayer on NiAl(110)



DFT analysis of structure



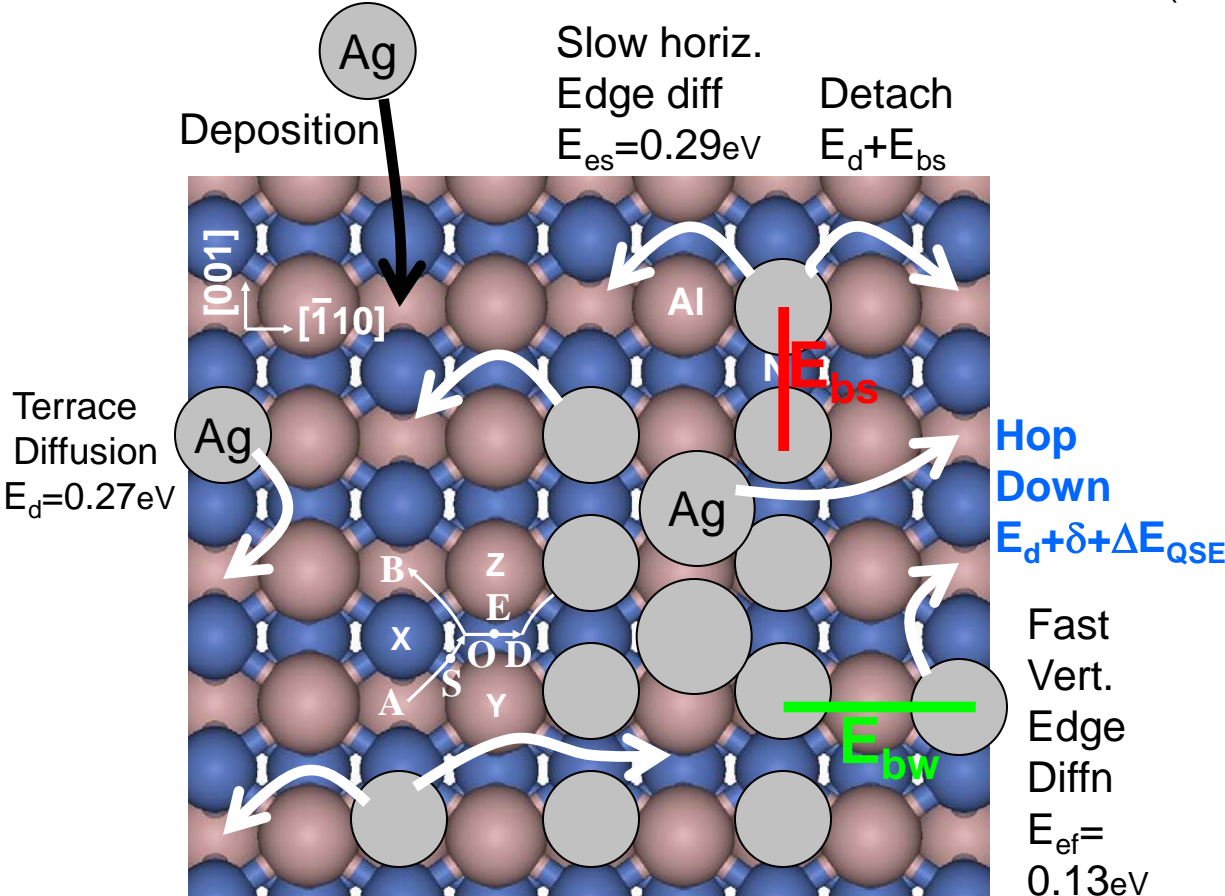
Ag/Ag(110) homoepitaxy



0.15 nm height (1 layer)

ATOMISTIC LG MODELING OF BILAYER ISLAND FORMATION

Han et al. PRL 100 (2008) 116105; PRB 81 (2010) 115642



$E_{bs} = 0.09\text{eV}$ (stronger attraction)

$E_{bw} = 0.03\text{eV}$ (WEAK attraction)

...all energies from DFT analysis

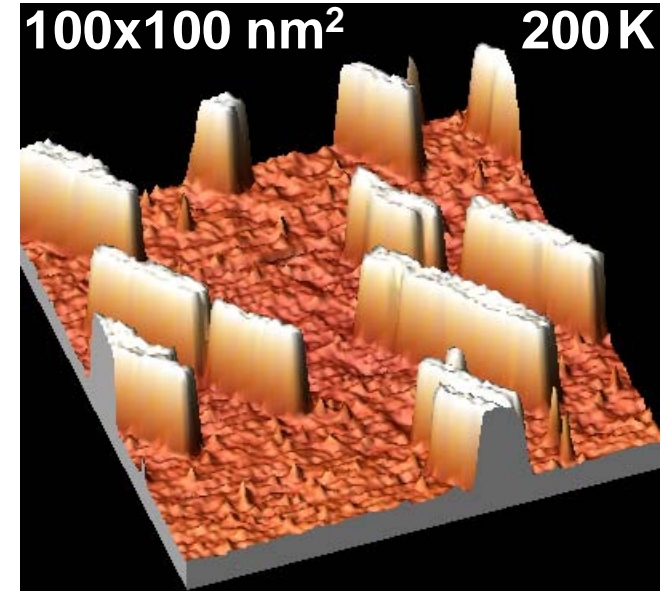
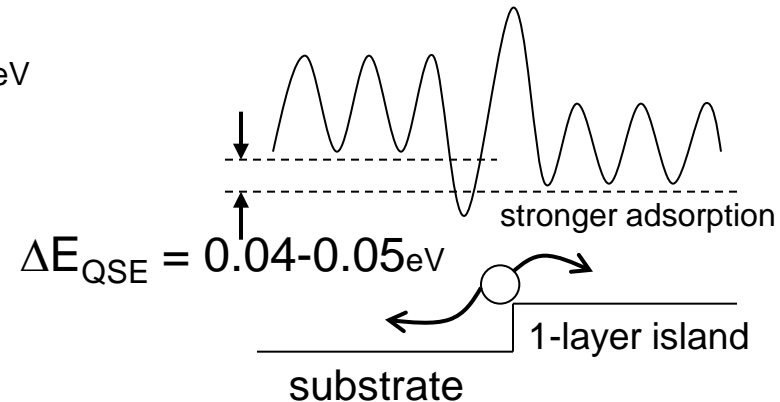


Image shown on NSF Chemistry web site

Ag/NiAl(110) 200 K 20% covered
Island Height: 0.33nm @ all T

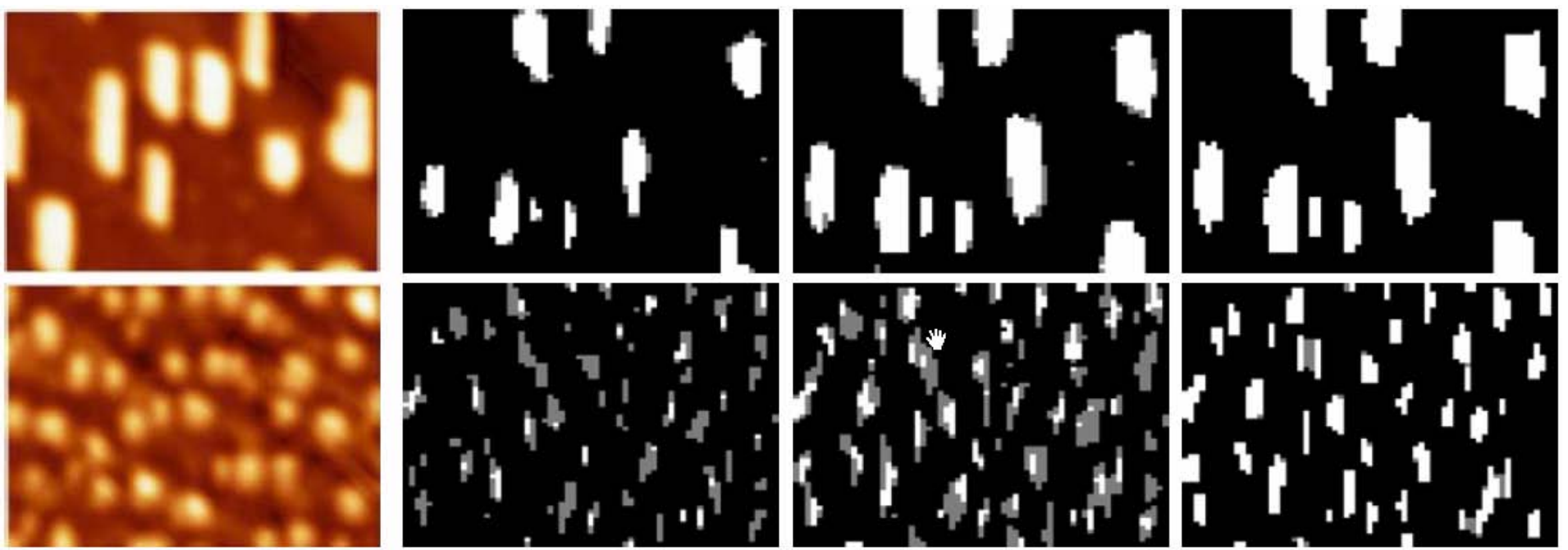


STM VS. KMC SIMULATION OF BILAYER ISLAND FORMATION

STM & KMC images of Ag islands on NiAl(110) are $27 \times 19 \text{ nm}^2$
KMC images: grey = 1st layer; white = 2nd layer Ag adatoms

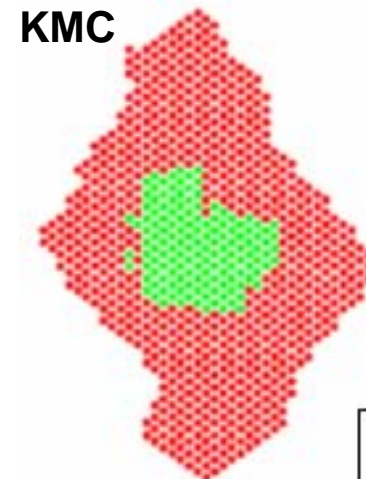
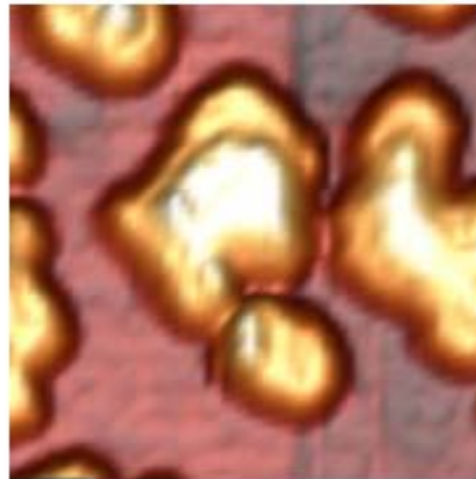
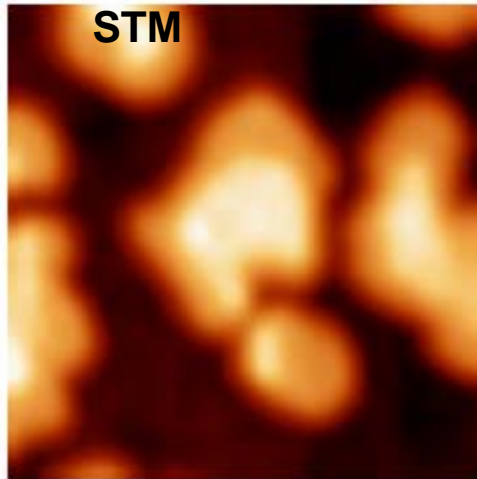
Han et al. PRL 100 (2008)
PRB 81 (2010)

TOP: DEPOSITION OF Ag ON NiAl(110) AT 140K WITH LOW F = 0.003BL/S
STM: 0.2BL KMC: 0.1BL KMC: 0.2BL KMC: 10 min later



STM: 0.14BL KMC: 0.07BL KMC: 0.14BL KMC: 10 min later
BOTTOM: DEP.N OF Ag ON NiAl(110) AT 130K WITH HIGH F = 0.03 BL/S

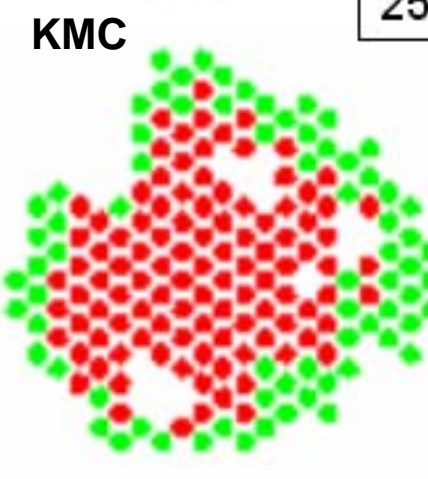
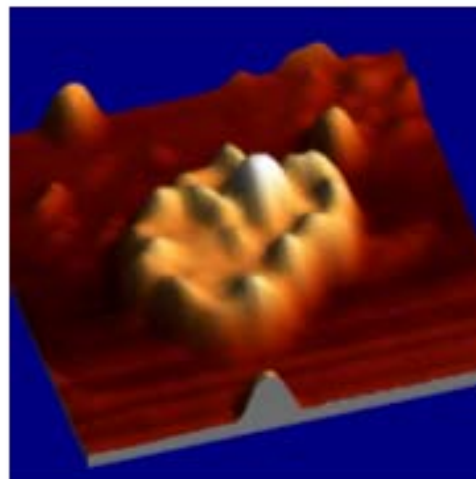
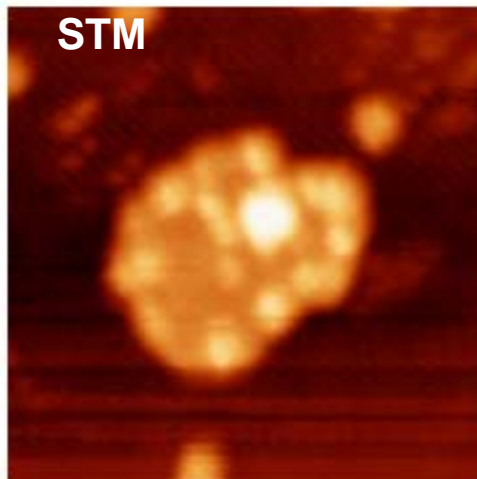
SEQUENTIAL CO-DEPOSITION OF Ni + Al ON NiAl(110) AT 300K



Al then Ni

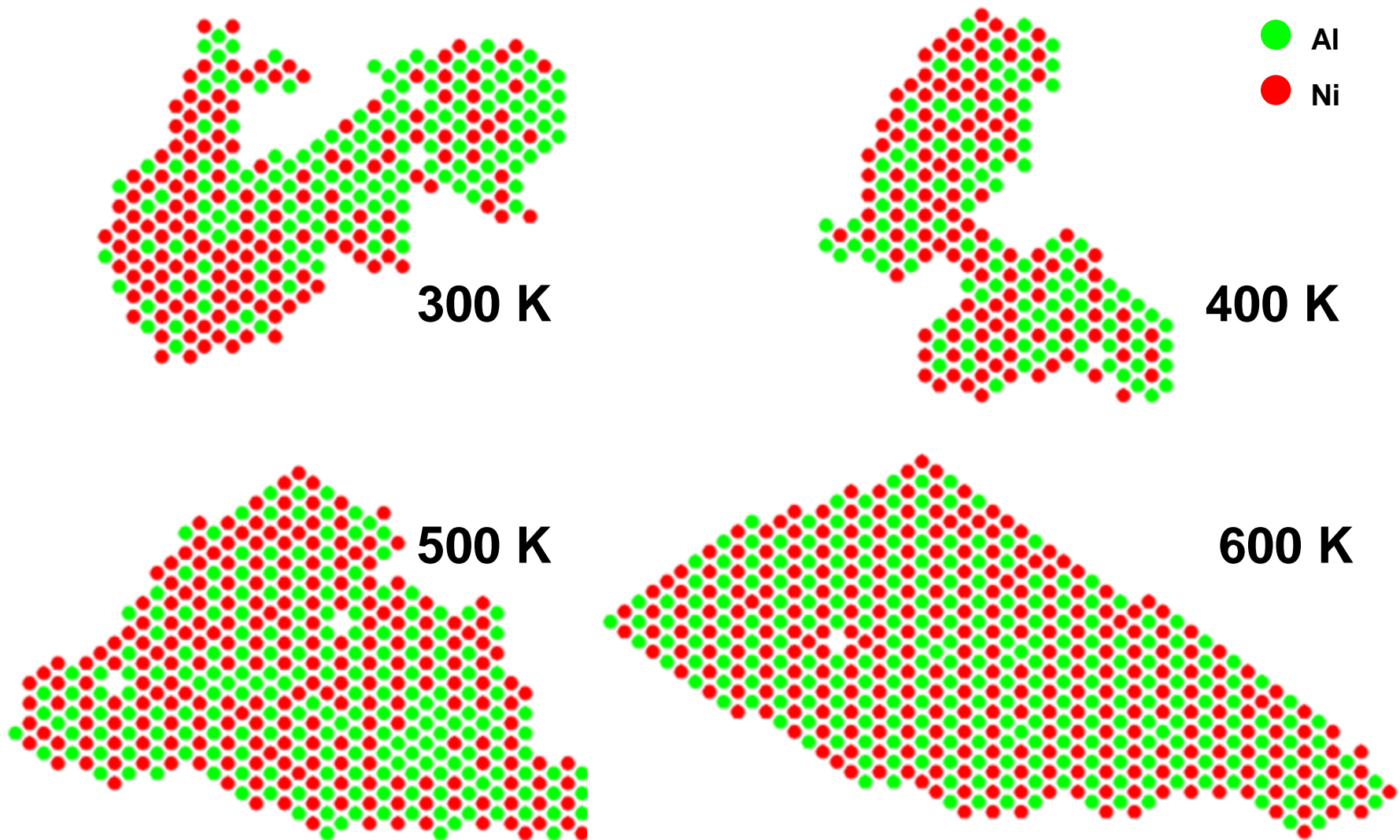
- Al
- Ni

STM images:
25 x 25 nm²



Ni then Al
(2nd layer
Al islands
not shown
In KMC)

SIMULTANEOUS CO-DEPOSITION OF Ni + Al ON NiAl(110)



SUBMONOLAYER ISLAND FORMATION

(NUCLEATION AND GROWTH OF 2D ISLANDS)

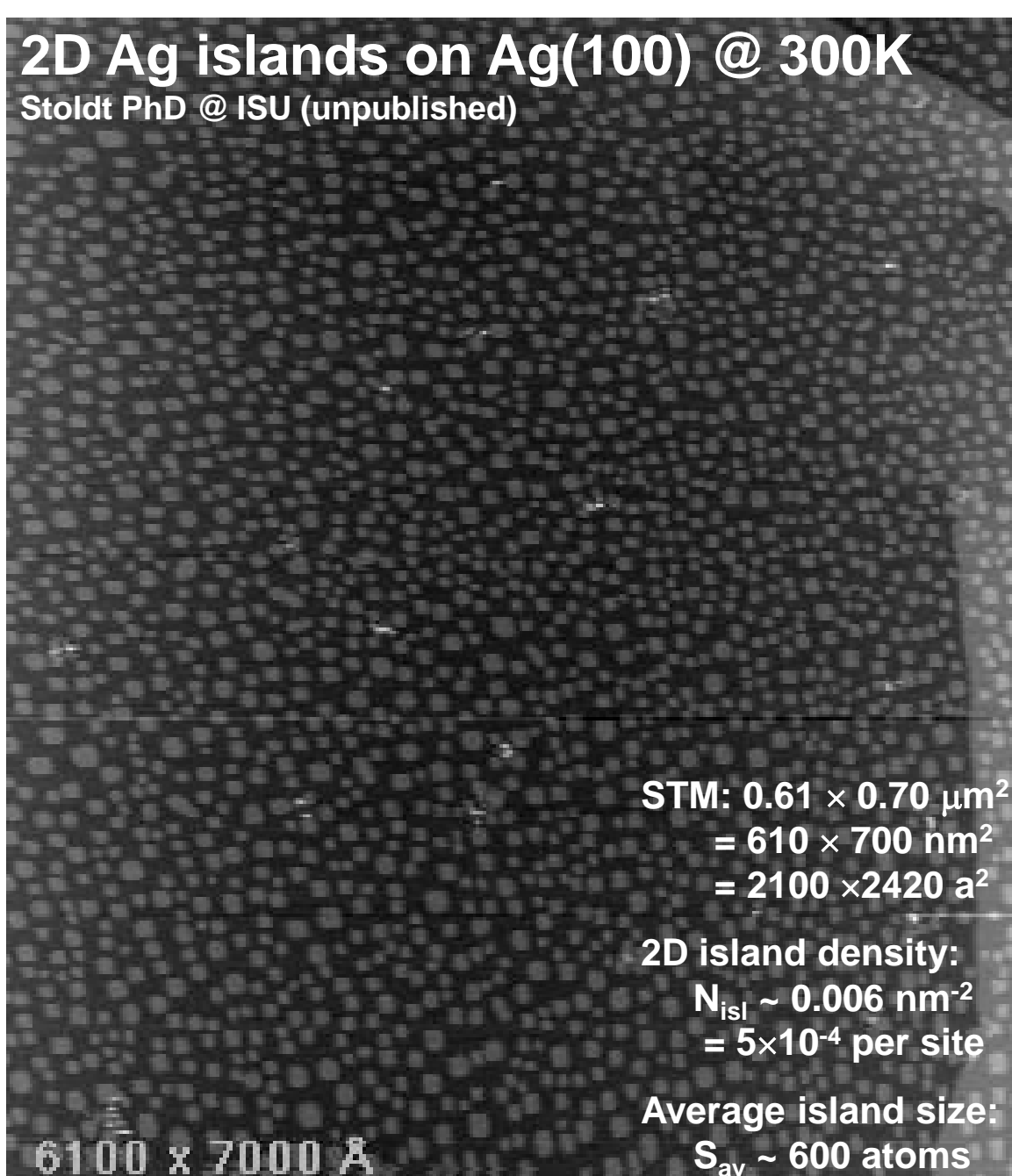
...analytic theory & step-dynamics + geometry-based simulation

Mean island size & density?

Island size distribution?

Island spatial distribution?

2D Ag islands on Ag(100) @ 300K
Stoldt PhD @ ISU (unpublished)



STM: $0.61 \times 0.70 \mu\text{m}^2$
 $= 610 \times 700 \text{ nm}^2$
 $= 2100 \times 2420 \text{ a}^2$

2D island density:
 $N_{\text{isl}} \sim 0.006 \text{ nm}^{-2}$
 $= 5 \times 10^{-4} \text{ per site}$

Average island size:
 $S_{\text{av}} \sim 600 \text{ atoms}$

6100 x 7000 Å

ANALYSIS OF SUBMONOLAYER ISLAND FORMATION

Rate Equation Theory (traditional mean-field theory 1960's- Zinsmeister, Venables,...)

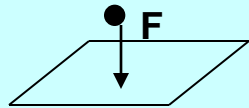
N_{isl} = TOTAL density of ALL stable islands (counting all sizes $s > i$ where **i = critical size**)

N_1 = density of adatoms

$$\frac{d}{dt} N_{isl} = \sigma_i h N_1 N_i (= K_{nuc}) \text{ where } N_i \approx \exp[E_i/(kT)] (N_1)^i \quad (1)$$



$$\frac{d}{dt} N_1 = F - \sigma_{isl} h N_1 N_{isl} - (i+1)K_{nuc} \quad (2)$$



where the "capture number" σ_i applies for islands of size i , and σ_{isl} applies for stable islands

Quasi-steady-state analysis:

Rough balance between adatom gain & loss: $\frac{d}{dt} N_1 \approx 0$ so $F \approx \sigma_{isl} h N_1 N_{isl}$ from (2) given small K_{nuc}

Substitute $N_1 \approx F/(\sigma_{isl} h N_{isl})$ into (1) and integrate:

Central result... $N_{isl} \sim \exp[-(i+2)^{-1}E_i/(kT)] (F/h)^{i/(i+2)}$

ANALYSIS OF SUBMONOLAYER ISLAND FORMATION

Rate Equation Theory (traditional mean-field theory 1960's- Zinsmeister, Venables,...)

N_s = density of stable islands of size $s > i$ where **i = critical size**; N_1 = density of adatoms

$$d/dt N_s = \sigma_{s-1} h N_1 N_{s-1} - \sigma_s h N_1 N_s \quad \dots \text{for } s \geq i+1$$



where σ_s = "capture number" for islands of size s (increases with s ...bigger islands capture better)

$$N_s \propto f(s/s_{av}); \sigma_s \propto c(s/s_{av}); s_{av} \sim t^z \Rightarrow (1-2z) f(x) - z \cdot x \frac{d}{dx} f(x) = - \frac{d}{dx} [c(x) \cdot f(x)]$$

$$\Rightarrow f(x) = f(0) \exp\left[\int_{0 < y < x} dy \{(2z-1) - c'(y)\}/\{c(y) - z \cdot y\}\right] \quad \text{Bartelt \& Evans (BE) PRB 54 (1996)}$$

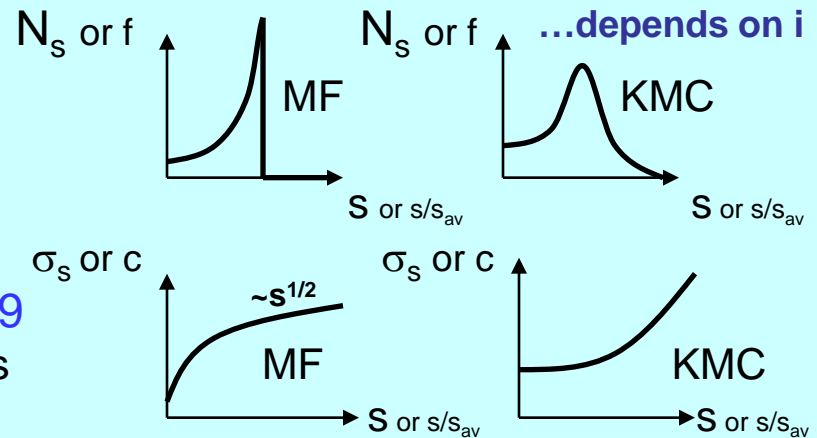
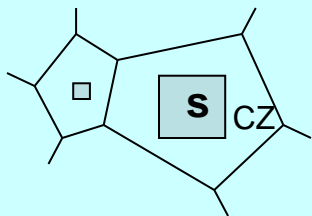
Successes of KMC & failure of MF theory

Precise KMC results for ISD showing failure of traditional Venables-type mean-field (MF) theory

BE PRB 46 (1992) 12675; SS (1993)

Explanation of MF failure BE PRB 54 (1996) 17359

KMC results for size-dependence of capture numbers



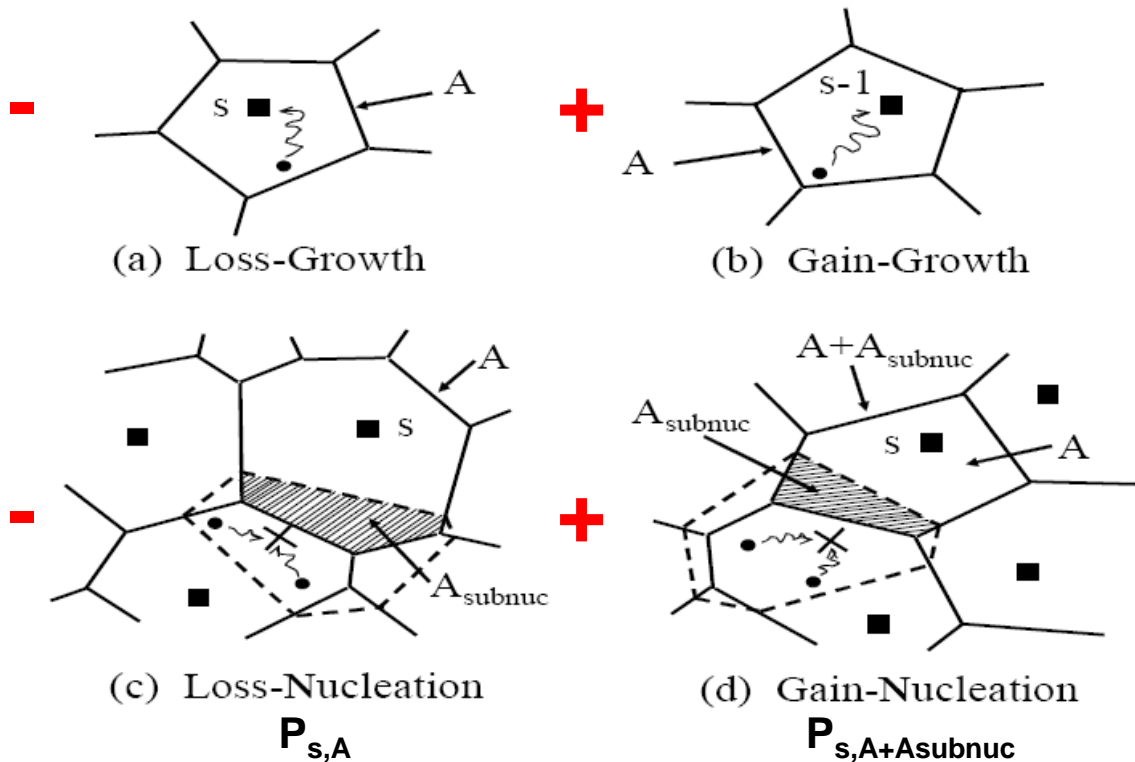
MF theory assumes all islands have same local environment
 ...but KMC shows bigger islands have bigger CAPTURE ZONES (CZ)

ANALYTICAL DEVELOPMENTS BEYOND-MF THEORY

JE & Bartelt, PRB 66 (2002) 235410; PRB 63 (2001) 235408; SSR (2006) ...motivated by Mulheran & Robbie EPL (2000)

$N_{s,A}$ = joint probability distribution (density) for islands of size s and CZ area A

$$\frac{d}{dt} N_{s,A} = - \underset{(a)}{F A N_{s,A}} + \underset{(b)}{F A N_{s-1,A}} - \underset{(c)}{P_{s,A} K_{nuc}} + \underset{(d)}{P_{s,A+A_{subnuc}} K_{nuc}} \quad \dots K_{nuc} = \text{nucleation rate}$$



Growth = easy

Rate \propto CZ area

Nucleation = tricky

Probabilities $P_{s,A+\dots}$ depends on spatial details of the nucleation process, especially location relative to CZ bndry

CAPTURE-ZONE AREA DISTRIBUTION: THEORY AND SIMULATION

PRL 99, 226102 (2007)

PHYSICAL REVIEW LETTERS

week ending
30 NOVEMBER 2007

Capture-Zone Scaling in Island Nucleation: Universal Fluctuation Behavior

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(Received 15 December 2006; published 27 November 2007)

In island nucleation and growth, the distribution of capture zones (in essence proximity cells) can be described by a simple expression generalizing the Wigner surmise (power-law rise, Gaussian decay) from random matrix theory that accounts for spacing distributions in a host of fluctuation phenomena. Its single adjustable parameter, the power-law exponent, can be simply related to the critical nucleus of growth models and the substrate dimensionality. We compare with extensive published kinetic Monte Carlo data and limited experimental data. A phenomenological theory elucidates the result.

Population of CZ's with area A

$$N_A \approx N_{\text{isl}}/A_{\text{av}} g(A/A_{\text{av}})$$

Heuristic theory (Generalized Wigner Surmise)

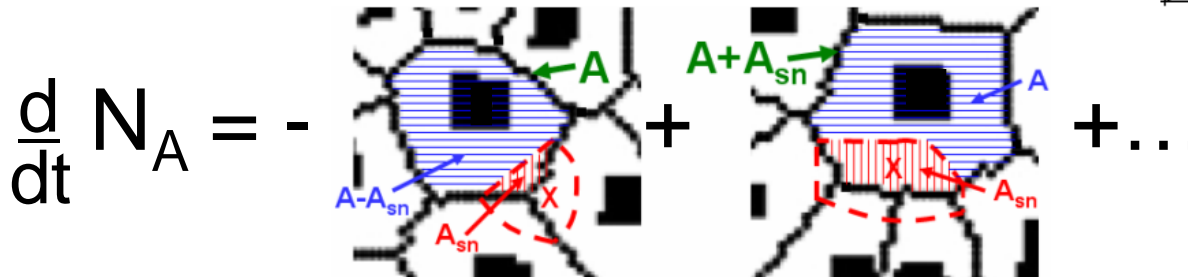
$$\text{GWS: } g(\alpha) \propto \alpha^\beta \exp(-b\alpha^2)$$

...with $\beta = i+1$ ($\beta=2$ for $i=1$)

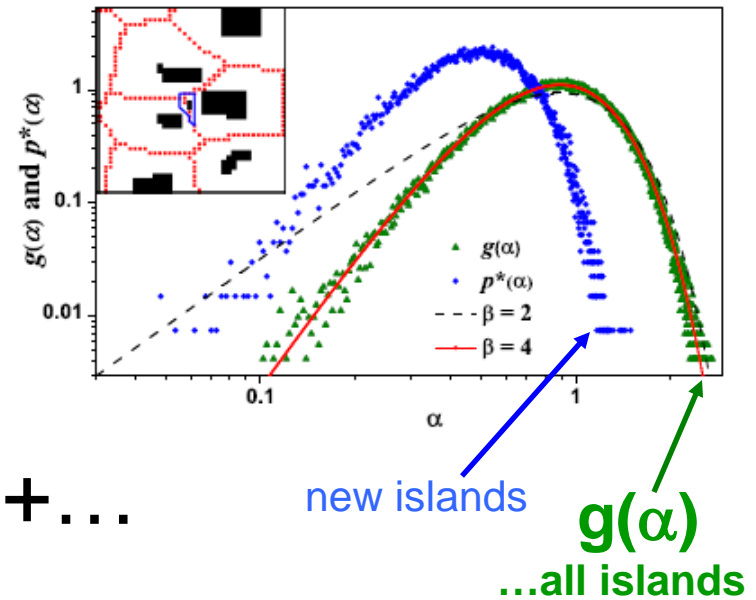
Fundamental Theory for N_A and g
incorporating spatial aspects of nucleation

$$g(\alpha) \sim \alpha^\beta \exp(-b\alpha^n) \text{ with } n \approx 1.5 \text{ for } i=1$$

$$\beta \approx 4 \text{ (KMC } i=1); \beta \approx 3 \text{ (KMC } i=0) \quad \beta \approx 3i/2 + 3$$

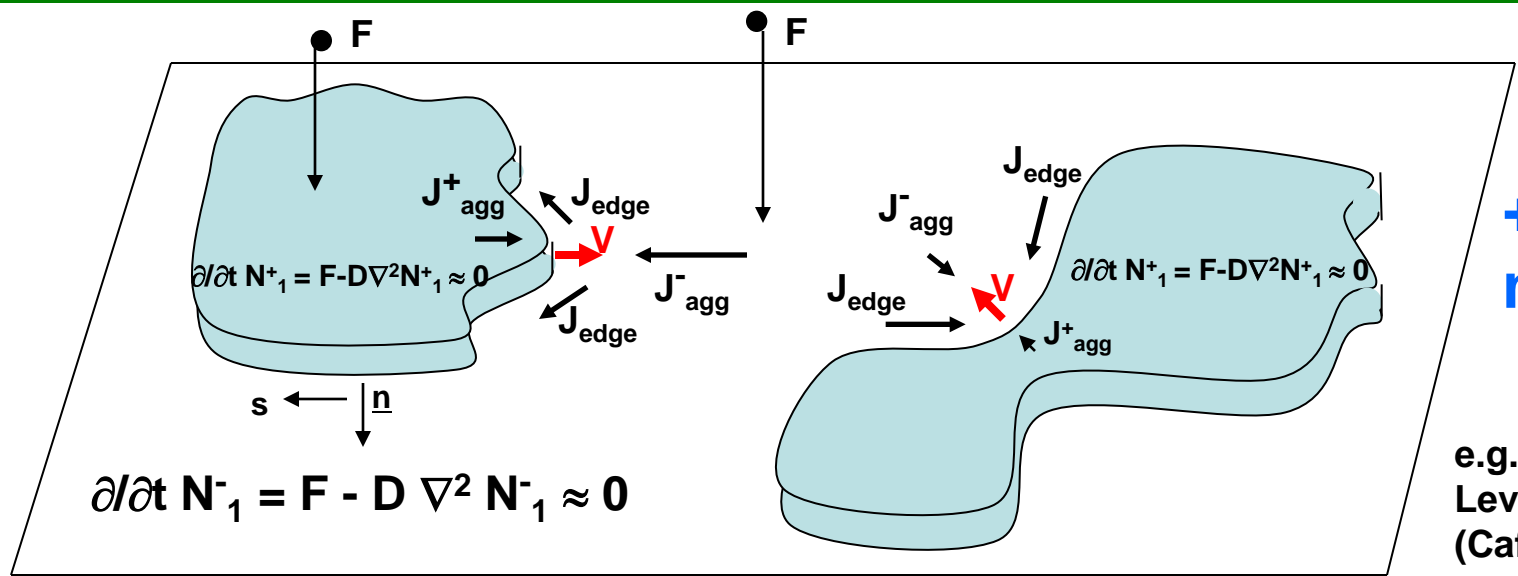


$$\Rightarrow 2g(\alpha) + \alpha \frac{d}{d\alpha} g(\alpha) = -Mq(\alpha)g(\alpha) + M\langle(1+\alpha'/\alpha)q(\alpha+\alpha')g(\alpha+\alpha')\rangle' +$$



Maozhi Li, Y. Han, JE,
PRL 104 (2010) 149601

STEP-DYNAMICS MODELING OF 2D ISLAND FORMATION



+ island nucleation

e.g., UCLA
Level-set approach
(Caflich, Ratsch,..)

N^{\pm}_1 = adatom density on upper/lower terraces

$J^{\pm}_{agg} = |D \nabla N^{\pm}_1|$ = aggregation fluxes; J_{edge} = step edge diffusion flux

Normal velocity of island edge: $V = J^-_{agg} + J^+_{agg} - \nabla_s J_{edge}$

BC for PDE: $\pm D \underline{n} \cdot \nabla N^{\pm}_1 = K^{\pm} (N^{\pm}_1 - N_{eq})$ + permeability term K^{\pm} = kinetic coefficients

Traditional BCF theory: $K^{\pm} = \infty$ if NO barrier to attach to step edge \Rightarrow Dirichlet BD $N^{\pm}_1 = N_{eq}$

But really K^{\pm} are finite & depend on step orientation etc. (Margetis & Caflich 2008; Ackerman & JE 2010)

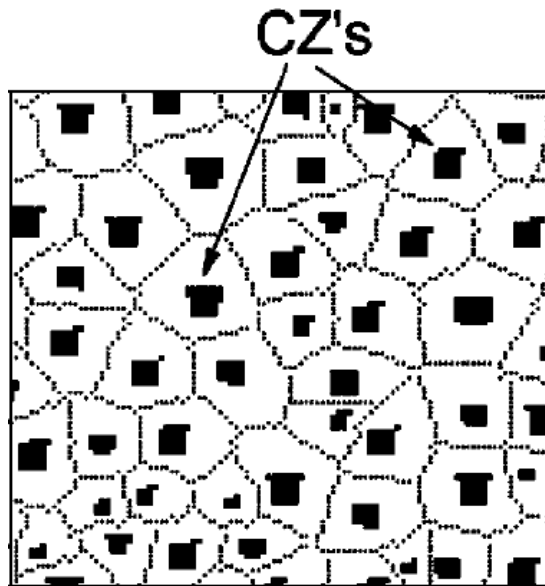
Edge diffusion: $J_{edge} = J_{equil} + J_{non-equil}$ where $J_{equil} = -\sigma_{step} \nabla \mu_{step}$ with $\mu_{step} \propto$ curvature

But $J_{non-equil} = \dots$ not fully understood (heuristic expressions available). **JE et al. Surf Sci Rep (2006)**

GEOMETRY-BASED SIMULATION OF ISLAND FORMATION

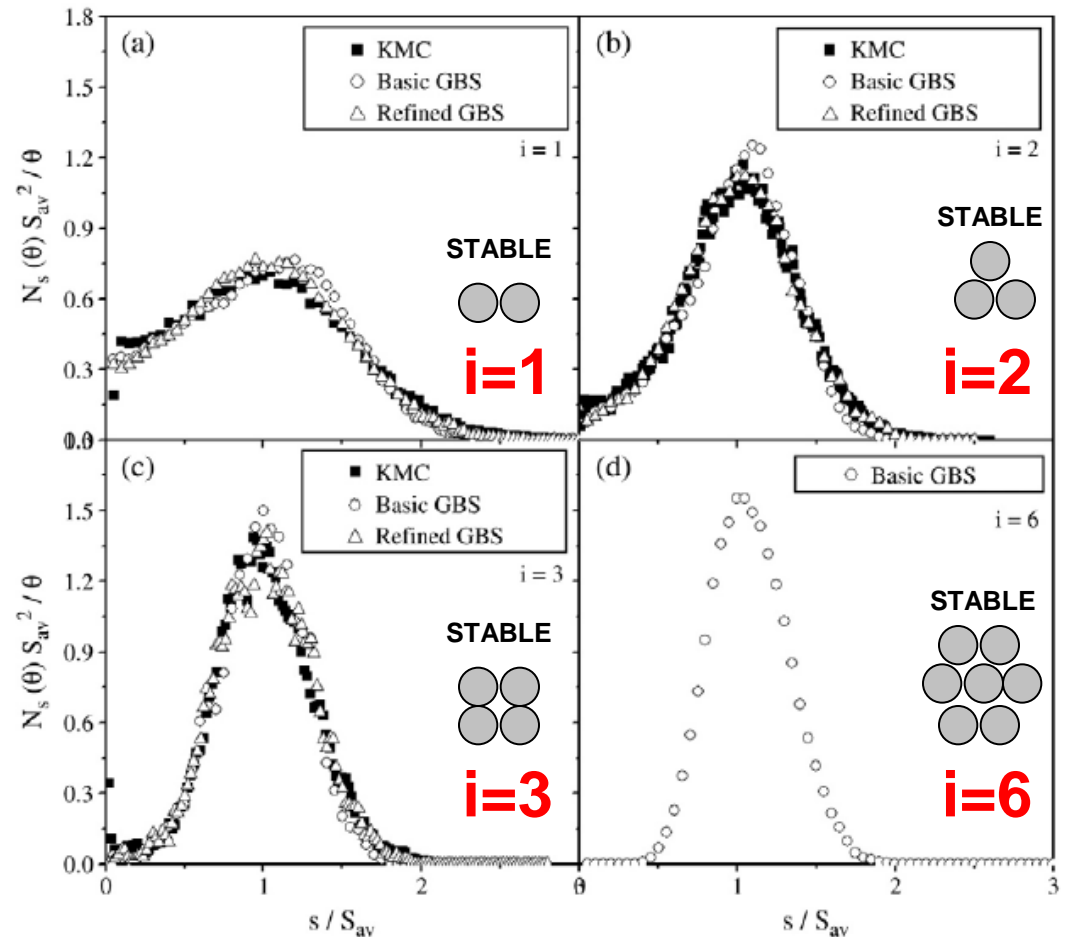
Maozhi Li et al. PRB 68 (2003) R121401; Surf Sci 546 (2003); SIAM MMS 3 (2005); Evans et al SSR 61 (2006)

GBS strategy: simulation based on CZ tessellation of surface (stochastic geometry) using CZ areas to determine island growth, and physics-guided nucleation near CZ boundaries



$i=1$

Li et al Surf Sci 546 (2003)
Evans et al Surf Sci Rep 61 (2006)

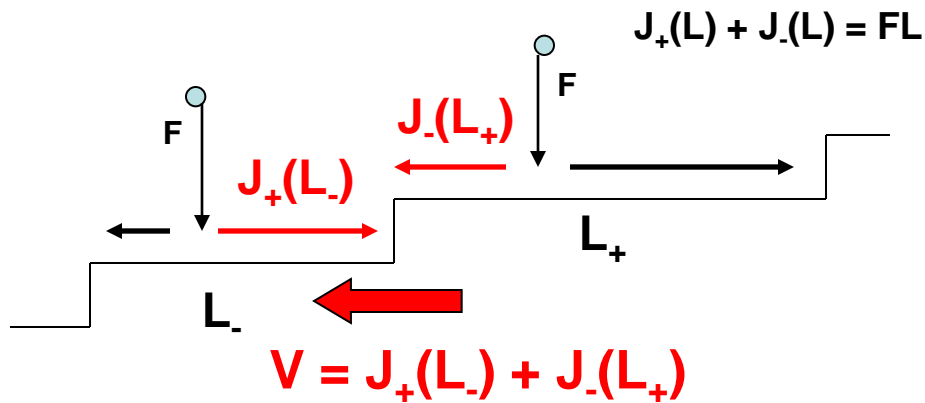


Key: realistic incorporation of island nucleation along CZ boundaries, probability weighted by distance from island edge

STEP-DYNAMICS MODELS FOR MULTILAYER GROWTH

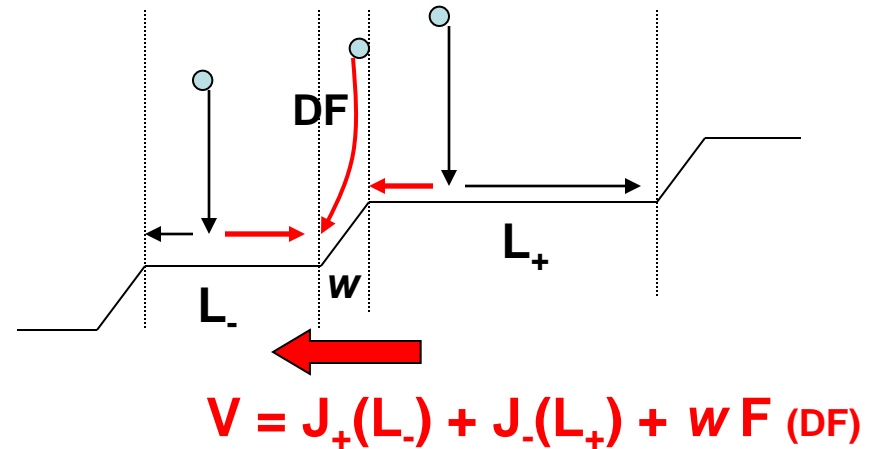
TRADITIONAL SD MODELING: NO DF

Politi & Villain, PRB 1996; Krug JSP 1997 etc.

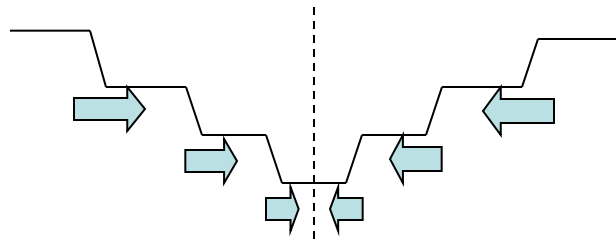


MODIFIED SD MODELING INCLUDING DF

Li & Evans, PRL 2005, PRB 2006, Surf Sci Rep 2006



Annihilation of steps at valleys between mounds unless $P_- = 0, c = 0$

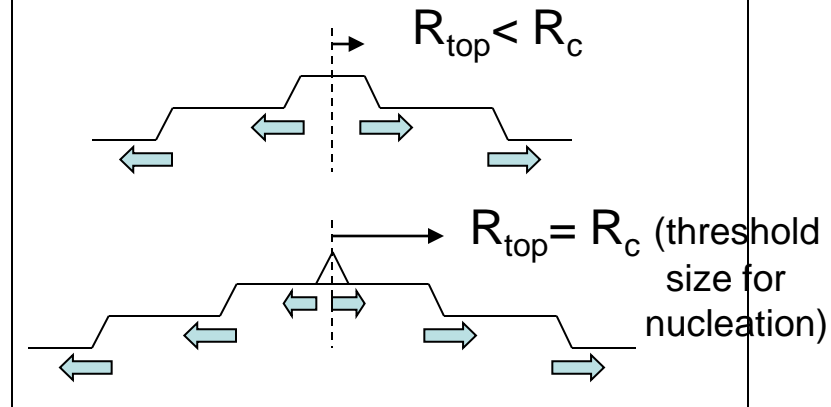


$P_+ = 1, P_- = 0, W = 0$
Zeno effect

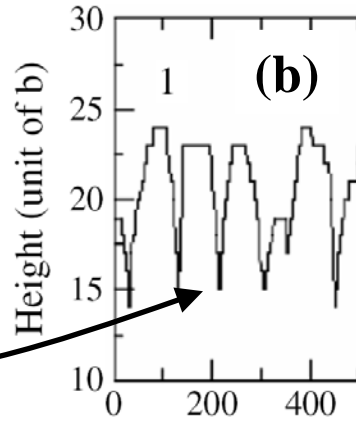
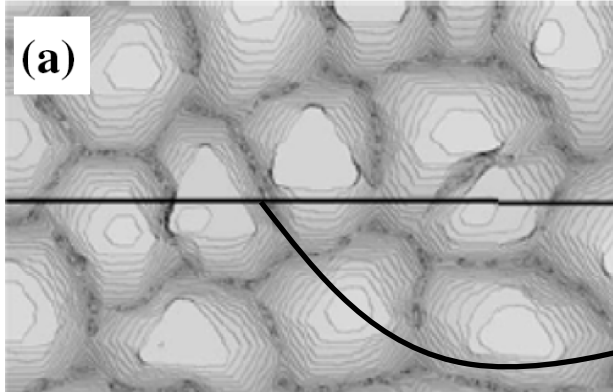
∞ ES barrier
no DF

$W > 0$
DF

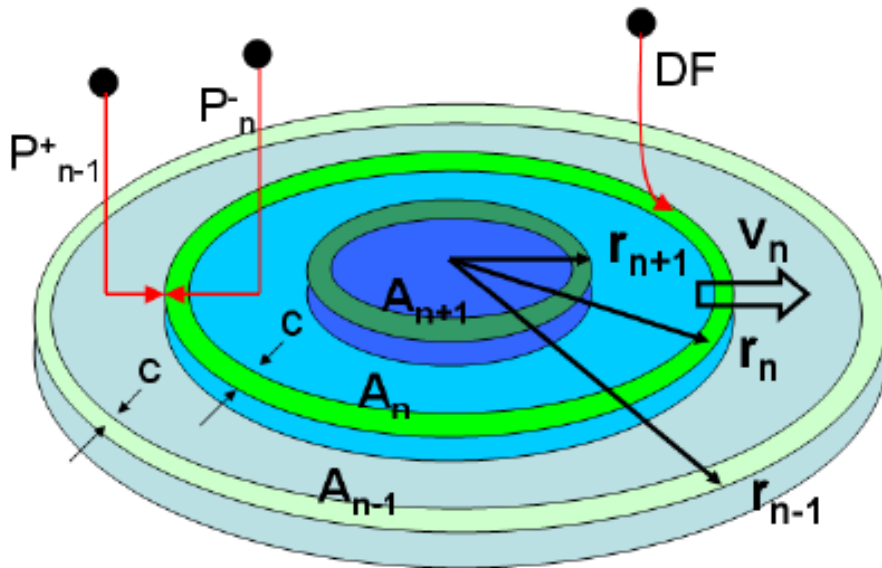
Nucleation of new islands at the peaks of mounds (Krug)



STEP DYNAMICS MODELING OF MOUND PROFILES FOR Ag/Ag(111)

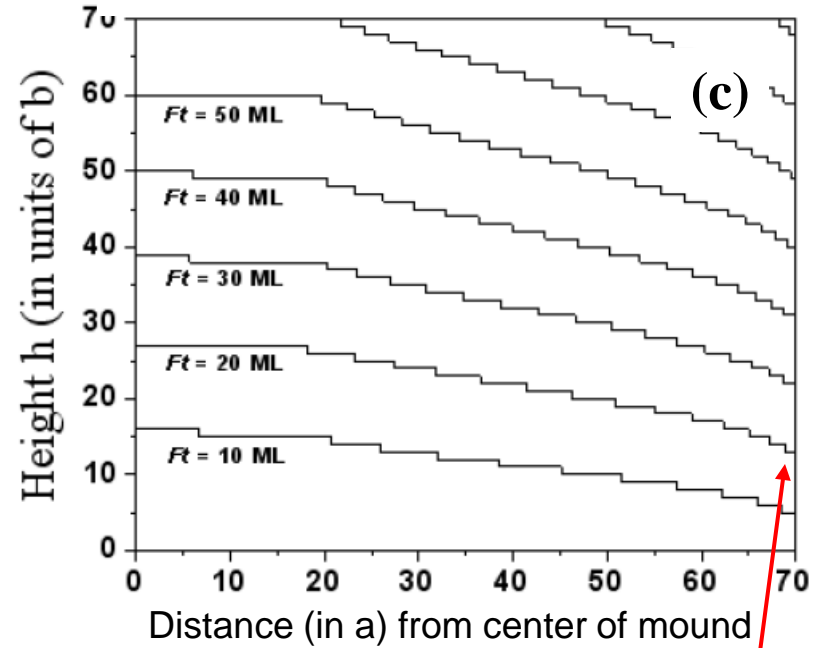


20 ML Ag/Ag(111) @ 150K



2+1D step dynamics with circular islands

STEP DYNAMICS SIMULATION



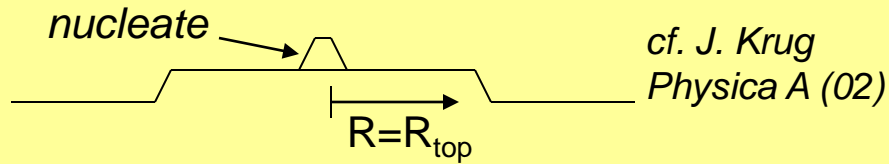
Height of valley between mounds
~ 13 layers above substrate @ 20 ML

Li, Han, JE, J. Phys.: Cond. Matt. (2009)

Alternative: use standard step-dynamics model with cut-off when bottom terrace width falls below 1 lattice constant (Krug)

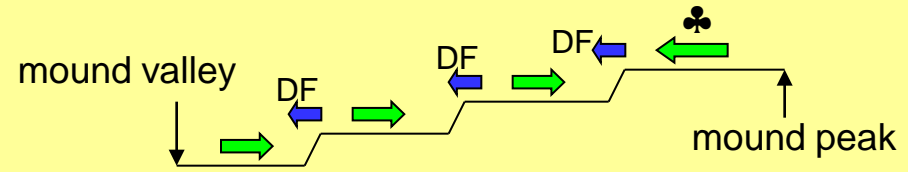
SD ANALYSIS OF SHAPE SELECTION FOR FINITE MOUNDS

Left: Selected Mound Shapes *versus* Threshold Radius for top layer nucleation



For small R_{top}/R : $m = m_s$ (infinite mound)

Right: (Micro) Net Step Attachment Flux = \sum fluxes from right - \sum fluxes from left



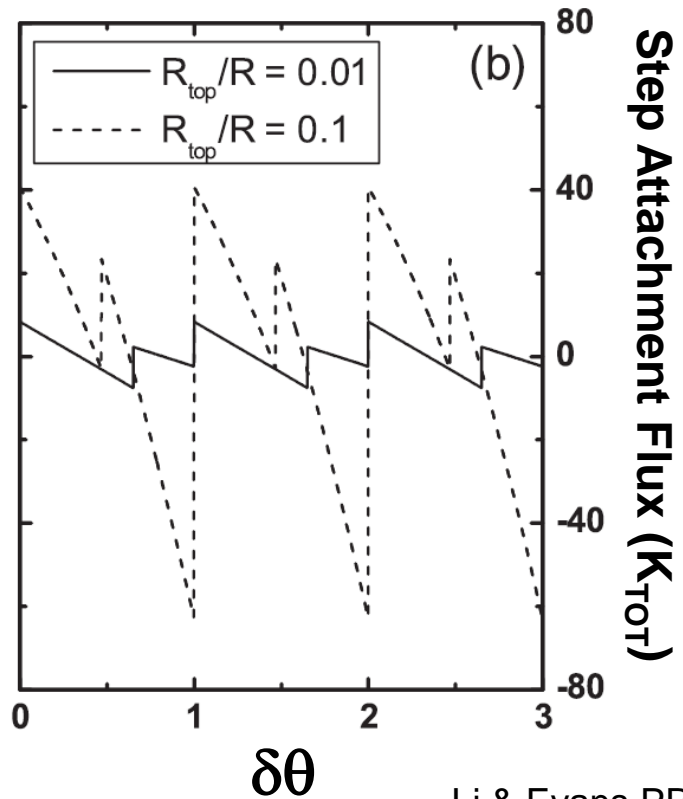
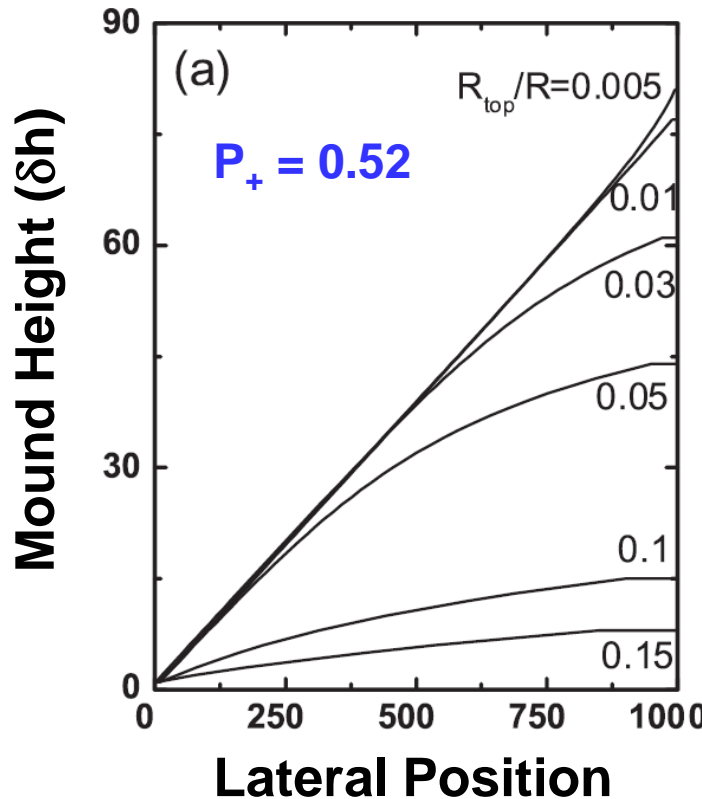
Key result: $\langle K_{tot} \rangle = 0$ (zero time-average)

SMALL $R_c = R_{top}$
(threshold size)

SELECTED
SLOPE
corresp. to
flux balance

LARGE $R_c = R_{top}$

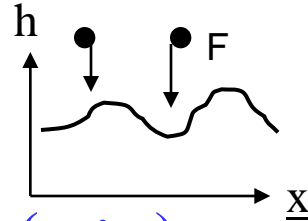
SELECTED
SHAPE with
side slope
smaller than
selected value



3D PHENOMENOLOGICAL CONTINUUM THEORY (PCT) FOR MOUND FORMATION IN MULTILAYER METAL HOMOEPITAXY

COARSE-GRAINED EVOLUTION EQUATION FOR FILM HEIGHT:

$h = h(\underline{x}, t)$ = film height at lateral position \underline{x} & time t ; $\underline{m} = \nabla h$ = local slope

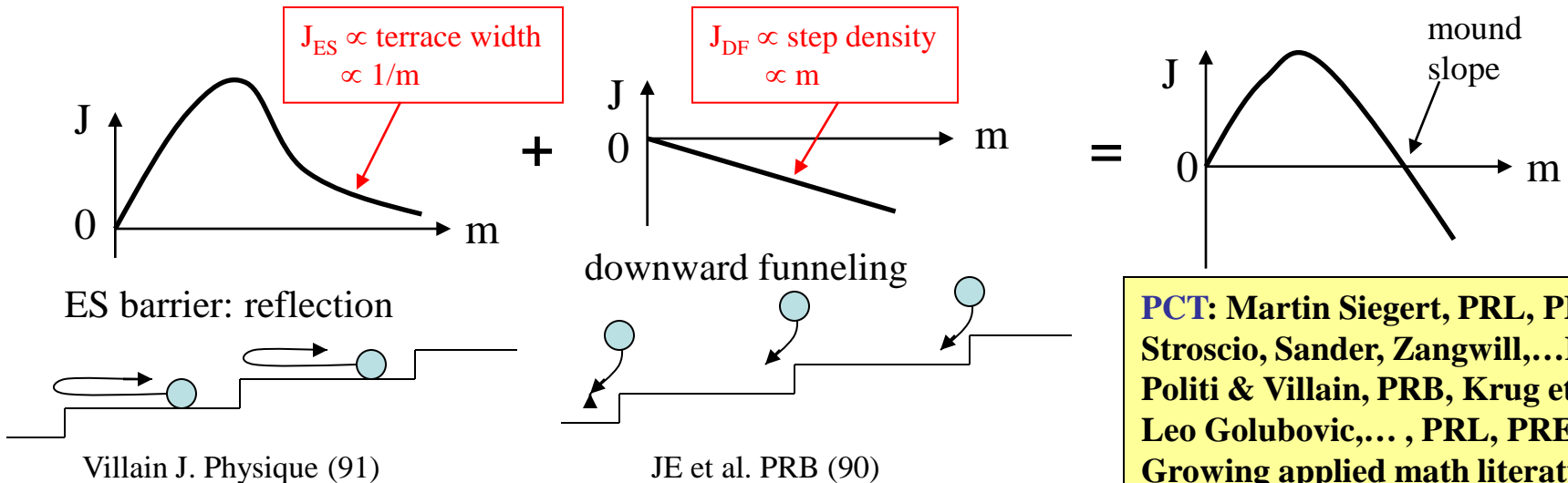


$$\frac{\partial}{\partial t} h = F \text{ (deposition flux)} - \nabla \cdot \underline{J}_{\text{PCT}} \text{ (lateral mass current)} + \eta \text{ (noise)}$$

where phenomenological $\underline{J}_{\text{PCT}} = \underline{J}_{\text{SLOPE}}(\underline{m}) + \underline{J}_{\text{RELAX}} + \underline{J}_{\text{SB}} + \dots$

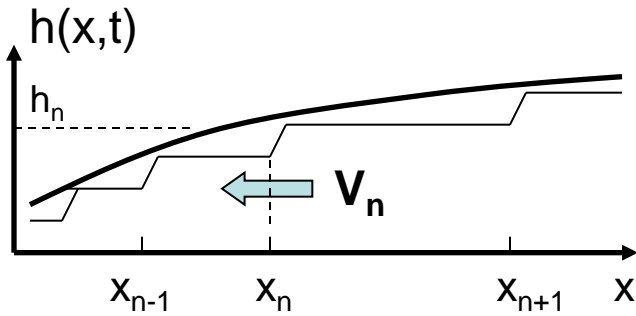
$v_k (-1)^k \nabla (\nabla^2)^k h$ $\nabla A(m^2) \leftarrow$ up-down symmetry breaking

$$\underline{J}_{\text{SLOPE}} = \underline{J}_{\text{ES}} \text{ (uphill-destabilizing)} + \underline{J}_{\text{DF}} \text{ (downhill-stabilizing)}$$



**PCT: Martin Siegert, PRL, PRB
Stroscio, Sander, Zangwill, ... PRL
Politi & Villain, PRB, Krug et al.
Leo Golubovic, ... , PRL, PRE
Growing applied math literature:
Bo Li, Kohn, Wise, Yagi, et al.**

COARSE-GRAIN SD \Rightarrow EXACT 3D CONTINUUM PDE



cf. W. E & Yip JSP 104 (2001)
Kohn et al. MRS Proc. (2002)

Fit smooth curve, $h(x,t)$, through SD configuration so $h_n = h(x_n,t)$ and $\partial/\partial t h(x_n,t) \approx -V_n \partial/\partial x h(x_n,t)$ and take small-slope continuum limit to obtain...

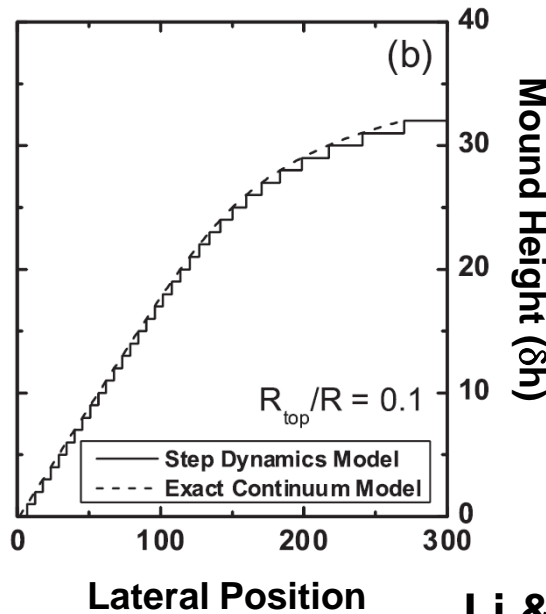
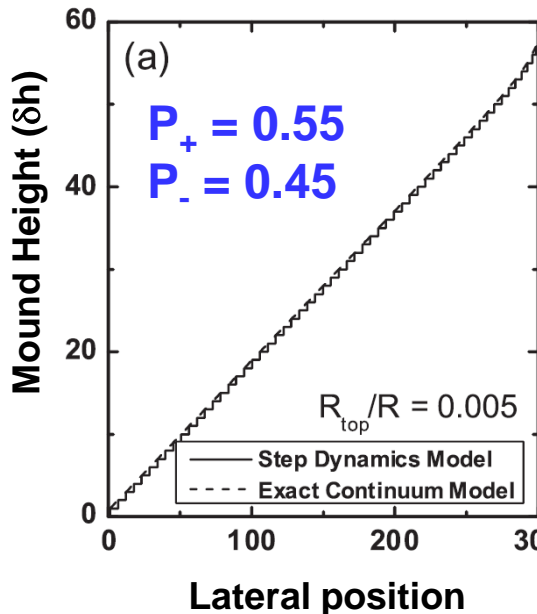
$$\partial/\partial t h(x,t) = Fb - \partial/\partial x J_{\text{EXACT}} \quad \text{where}$$

$$J_{\text{EXACT}} = [J_{\text{ES}} = Fb\Delta(1/m - ca)/2] + [J_{\text{SB}} = -Fbm^{-3}m_{xx}/6]$$

No J_{DF} or J_{RELAX} terms! *cf. Politi & Villain PRB (96)*

For selected mound shape:

Solve $J_{\text{EXACT}} = \text{const.}$ with suitable BC's at mound valley (selected slope) and peak



BC at valley:

Imposing selected slope accounts for effect of DF

BC at peak from $\langle K_{\text{tot}} \rangle = 0$:

$$\delta h = m_s R - f(\Delta) R_{\text{top}}$$

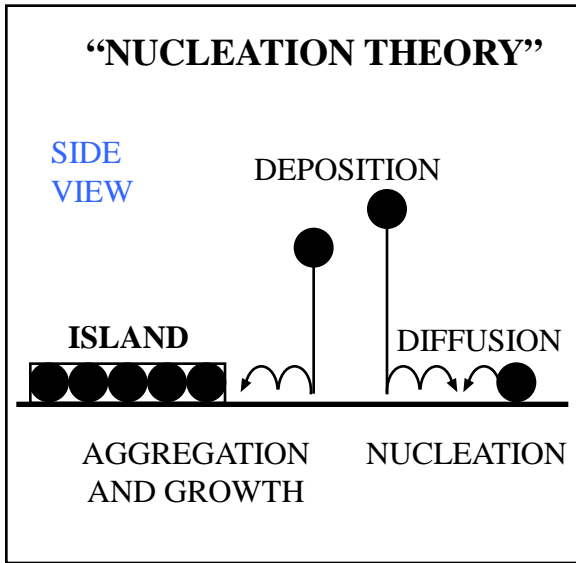
reflects prescription of nuc.
... "imposes relaxation"

CONCLUSIONS

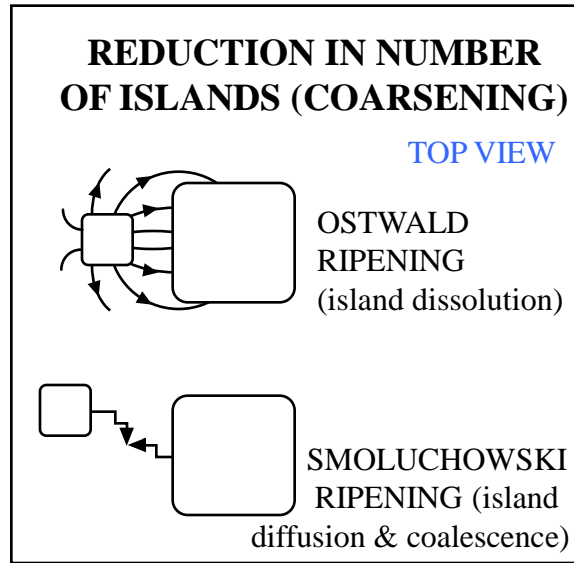
- ◆ **A on A Homoepitaxial Growth: simple systems, complex behavior**
Ag/Ag(111), Ag/Ag(100): Models developed with predictive capability describing diverse growth morphologies vs. T
- ◆ **A+B on AB Homoepitaxial Alloy Growth: ordering vs growth**
Ni+Al on NiAl(110): realistic models of alloy self-growth kinetics
- ◆ **Heteroepitaxial Growth: complex systems with novel behavior**
Many studies of strain effects (e.g., SK growth); we focus on lattice-matched systems
Ag on NiAl(110): growth mediated by Quantum Size Effects
Explain bilayer island growth with facile upward mass transport
- ◆ **Challenges for theory and modeling (homoepitaxy)**
Submonolayer island formation: analytic beyond-MF theory; efficient simulation
Multilayer kinetic roughening: rigorous coarse-grained PDE? stochastic models?

SUBMONOLAYER

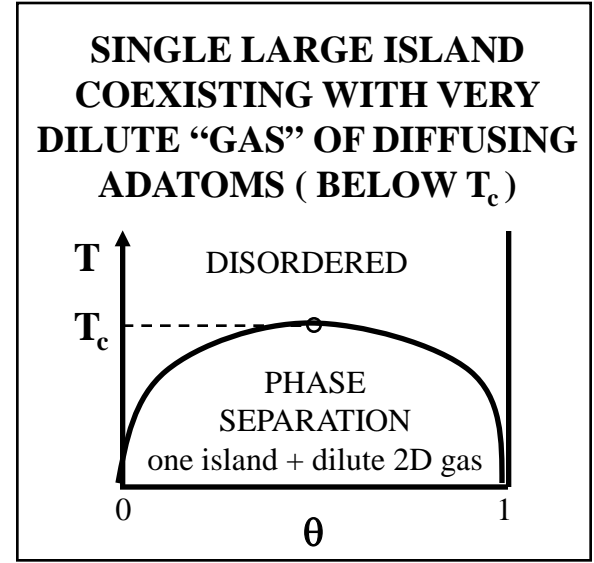
GROWTH (during deposition)



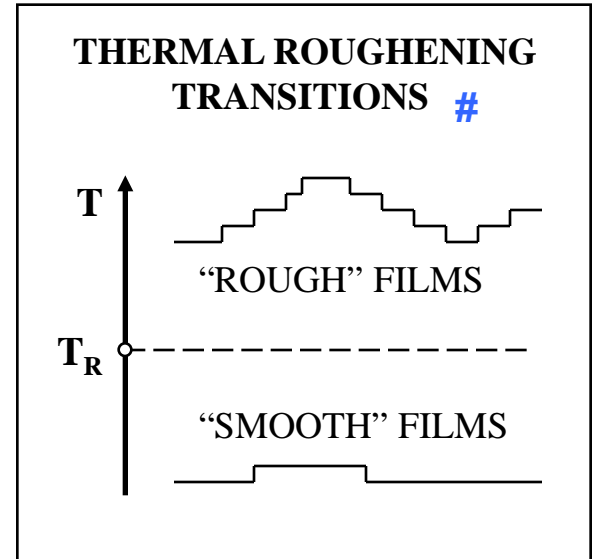
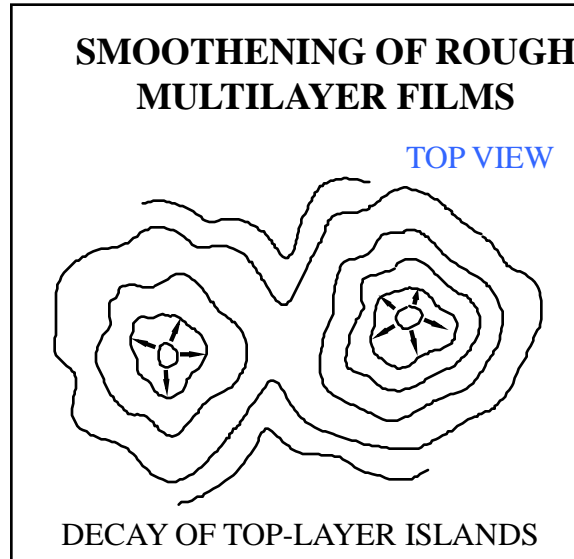
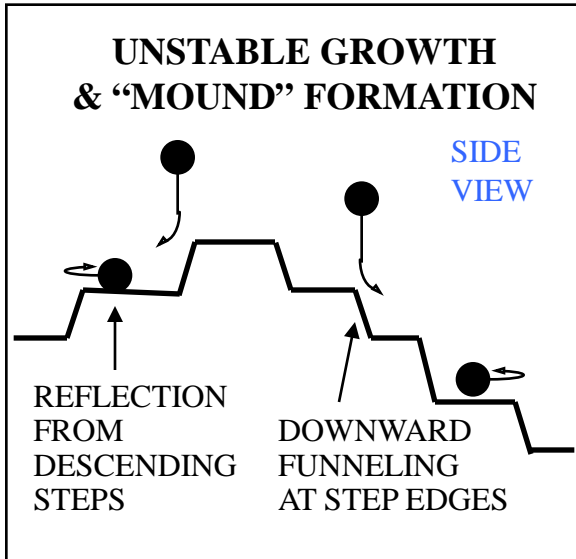
RELAXATION (post-deposition)



EQUILIBRIUM



MULTILAYER



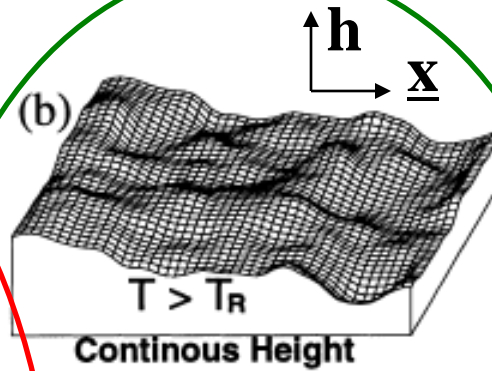
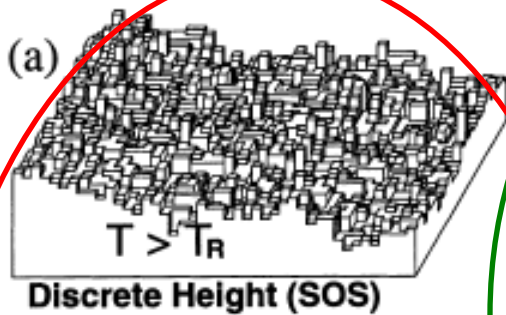
System driven far-from-equilibrium

System returns to equilibrium

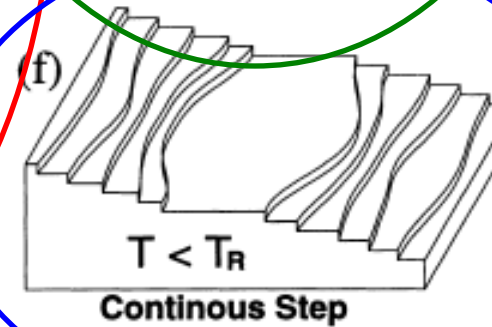
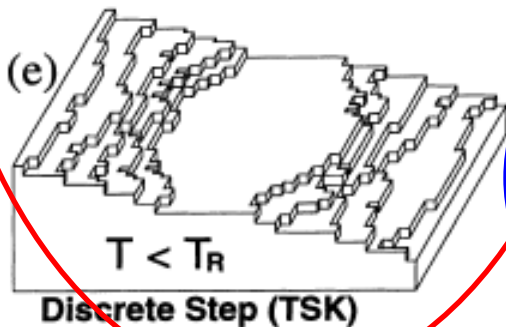
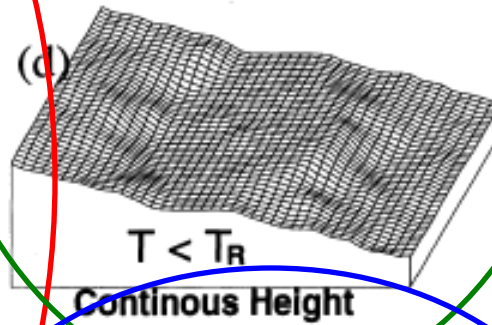
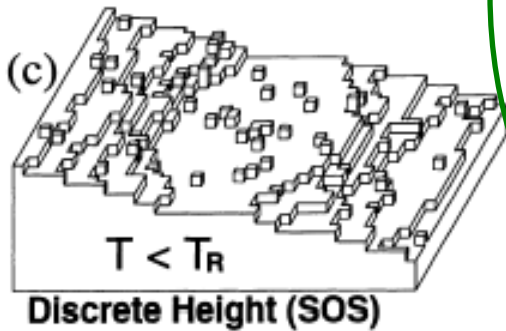
Simple equilibrium morphologies

ATOMISTIC VS. CONTINUUM MODELING STRATEGIES

H.-C. Jeong, E.D. Williams / *Surface Science Reports* 34 (1999) 171–294

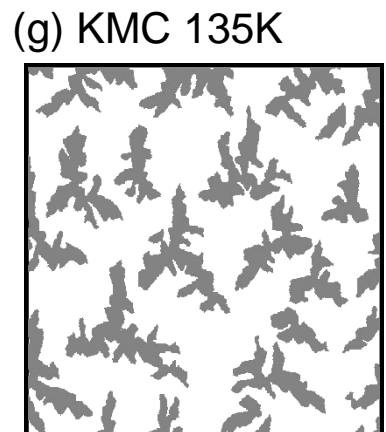
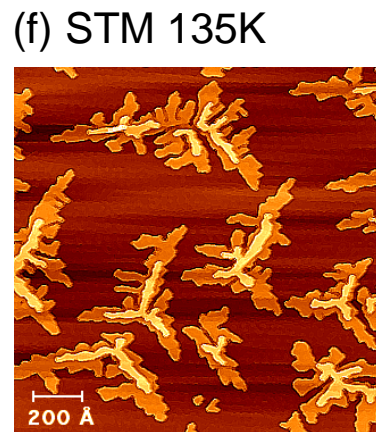
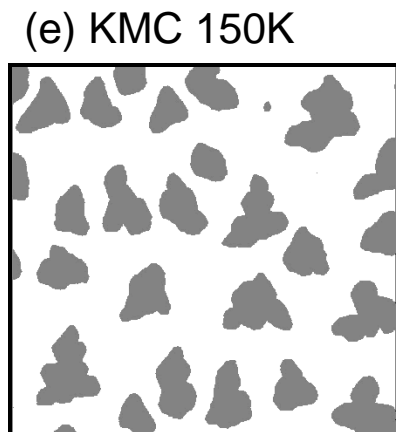
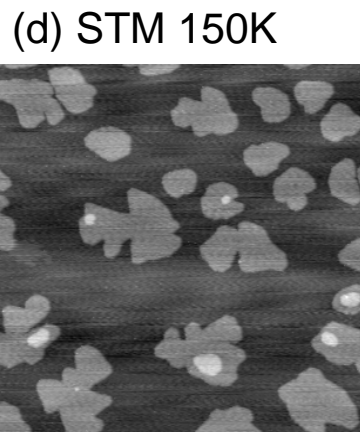
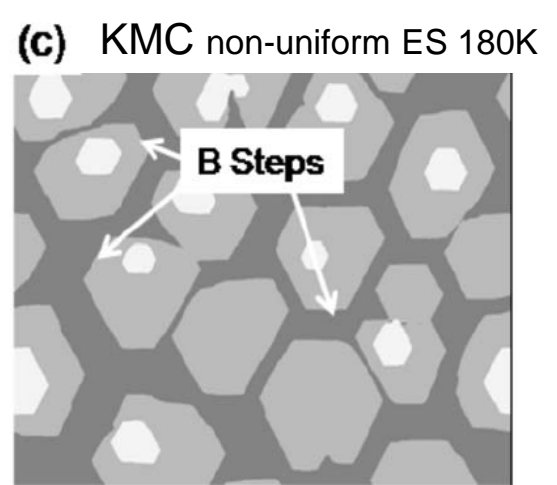
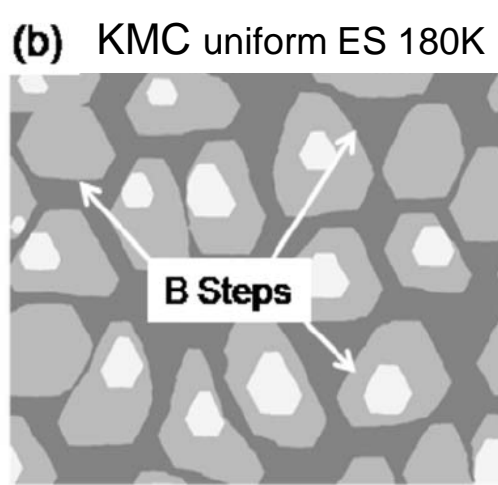
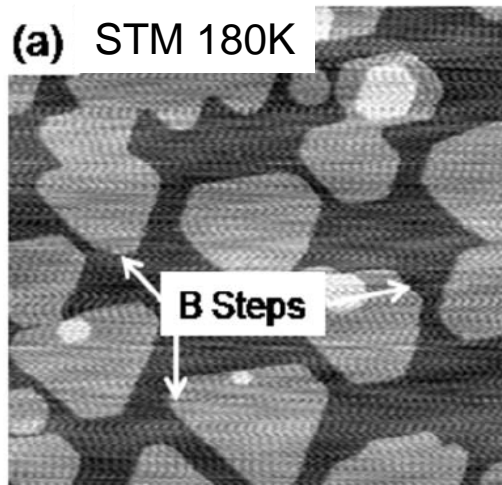


3D
CONTINUUM
 $\partial/\partial t h(\underline{x},t) = \dots$



2D
CONTINUUM:
(i) sharp steps
(e.g., level-set)
(ii) smeared steps
(i.e., phase-field)

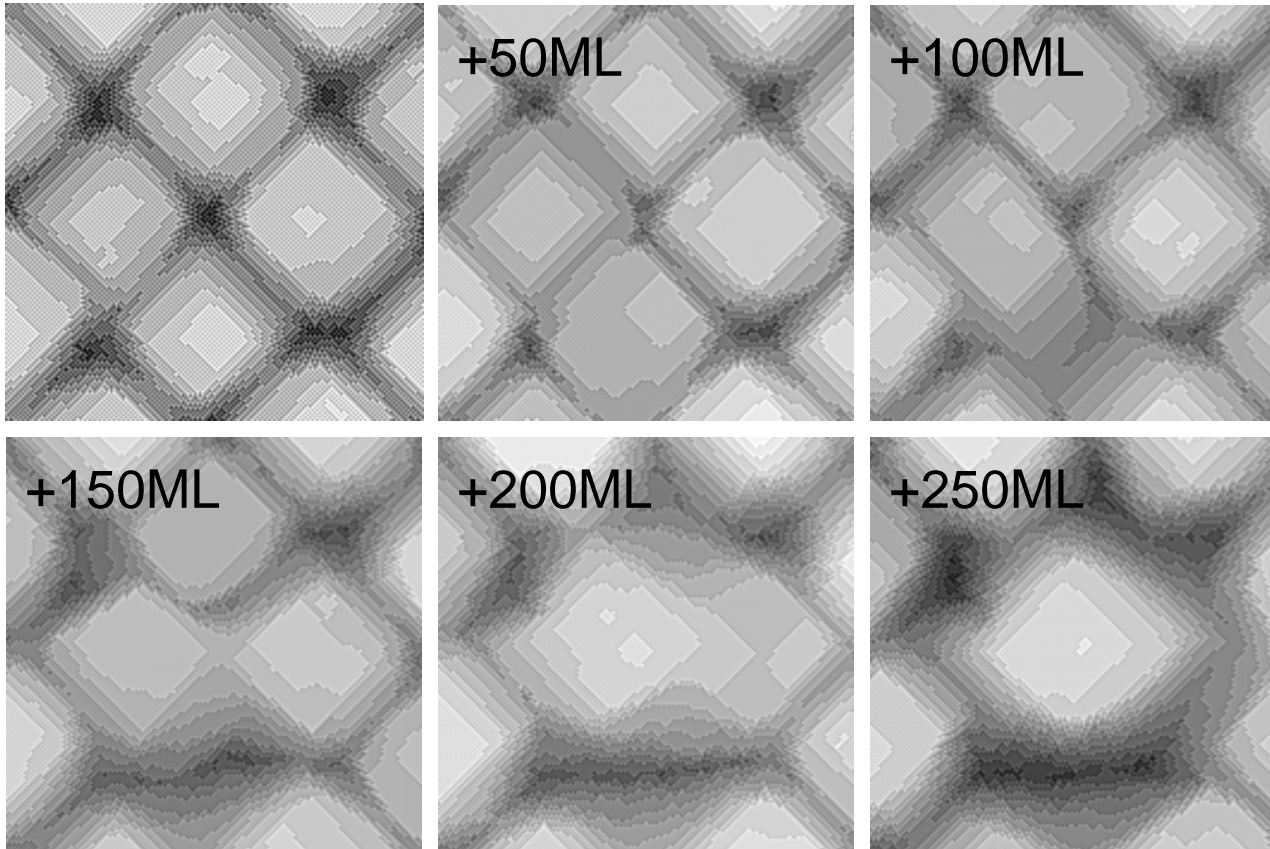
ATOMISTIC



FLUCTUATION-MEDIATED COARSENING OF MOUNDS

PREDICTIONS from REALISTIC ATOMISTIC MODEL ($i=1$; non-uniform ES barrier; DF, etc.)

- ◆ Mounds order into a 1×1 pattern for long times
- ◆ Strong up-down symmetry breaking (all upward no inverted pyramids)
- ◆ Fluctuations initiate mound disappearance, then concerted rearrangement



EXAMPLES: VAPOR PHASE DEPOSITION OF METAL FILMS

HOMOEPITAXY (crystal self-growth):

Ag films on Ag(111) etc. [A on A]

- Far-from-equilibrium morphologies (mounds) controlled by **Step-Edge (ES) Barriers** inhibiting downward transport
- Predictive atomistic models; coarse-graining challenge

Alloy self-growth: Ni+Al on NiAl(110) [B+C on BC]

- Explore interplay between film growth & alloy ordering
- Predictive atomistic modeling (of complex alloy system)

HETEROEPITAXY:

Ag on Pt(111); Fe on Cu(100); Pb on Cu(111);... [A on B]

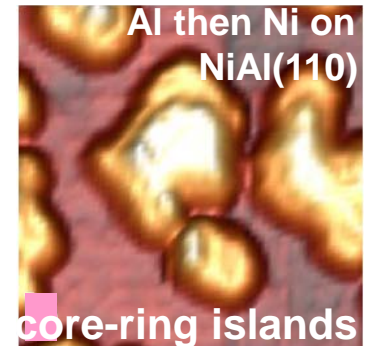
- Strain; exchange with substrate; quantum size effects;...

Ag films on binary alloy NiAl(110) [A on BC]

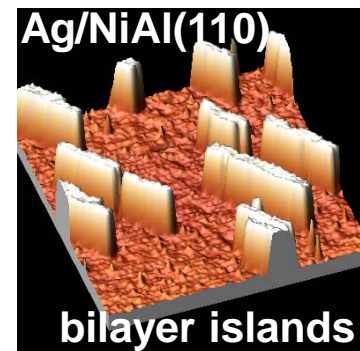
- No lateral mismatch \Rightarrow no lateral strain !
- **Quantum Size Effects** drive bilayer-by-bilayer growth
- atomistic modeling of facile upward transport at low T



PRB 77 (2008) 033402



PNAS (2010) Special Issue



PRL 100 (2008) 116105

ISU NSF CHE-0809472