Continuum Theory of Thin-Film Growth

SFU, Sep. 98

1. Modeling deposition, surface diffusion using stochastic differential equations

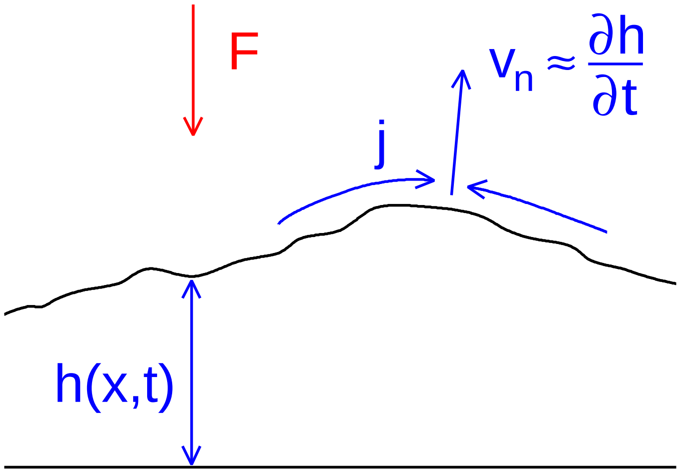
2. Numerical Methods
   - finite difference schemes
   - spectral methods

3. The coarsening puzzle
   - what drives it?
   - scaling?

   $\rightarrow$ tail of the structure factor (Porod's law)
   $\rightarrow$ growth laws
   $\rightarrow$ role of anisotropies

4. Conclusion

Introduction: Diffusion Currents in MBE



- particle deposition:
  flux F, beam fluctuations η
- no overhangs, no defects in the growing film
- no desorption,
  relaxation through surface diffusion exclusively
- sticking coefficient $= 1$

$\Rightarrow$ conservation law: mean velocity $\equiv F$

$$\frac{\partial h}{\partial t} = -\nabla \cdot \mathbf{j} + F + \eta$$

$$\mathbf{j}[h] = \mathbf{j}_{\text{eq}} + \mathbf{j}_{\text{neq}}$$

Surface Diffusion Currents

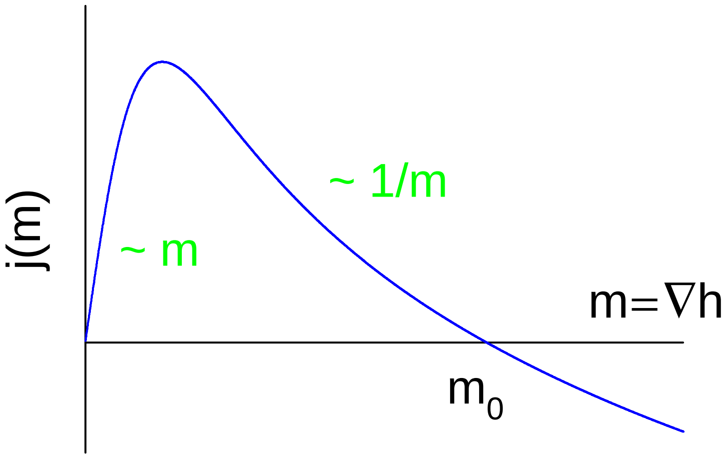
height evolution:

$$\frac{\partial h}{\partial t} = -D\Delta\Delta h - \nabla \cdot \mathbf{j}_{\text{neq}}[\nabla h] + F + \eta$$

Mullins 1957	Ehrlich 1966	flux	noise
Politi, Villain 1996	Schwoebel 1966		
	Villain 1991		

$$\langle \eta(\mathbf{x},t)\rangle = 0,\ \langle \eta(\mathbf{x},t)\eta(\mathbf{x}',t')\rangle = 2\epsilon\delta(\mathbf{x}-\mathbf{x}')\delta(t-t')$$

typical form of $\mathbf{j}_{\text{neq}}[\mathbf{m}],\ \mathbf{m} = \nabla h$:



2d-generalization? $j_{x,y}[m_x, m_y] = ?$

Surface Diffusion

Equilibrium ($F = 0$)

$\mathbf{j}_{\mathrm{eq}} \sim -\nabla\mu$ Fick's law

μ chemical potential

$\mu = \tilde{\sigma}\kappa$ Herring's equation

$\tilde{\sigma} = \sigma(\vartheta) + \sigma''(\vartheta)$ surface stiffness

$\kappa \simeq \nabla^2 h$ curvature

$$\left[\quad \mu = -\frac{\delta\mathcal{F}}{\delta h}\,, \quad \mathcal{F} = \int dA\,\sigma(\nabla h) \quad \right]$$

$$\Rightarrow \qquad \frac{\partial h}{\partial t} = -D\nabla^2\nabla^2 h$$

$+$ nonlinear terms $+$ thermal noise

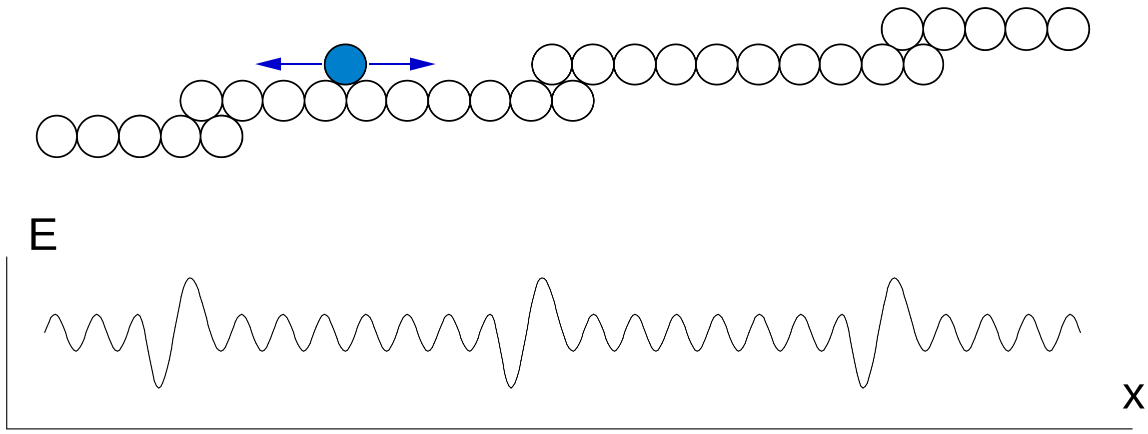
(Mullins 1957)

Surface Diffusion

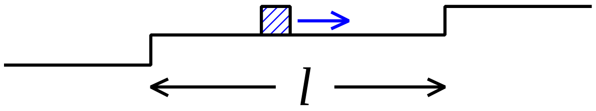
Nonequilibrium: $F \neq 0$

Slope dependent diffusion currents:

Lattice potential for an adatom on a vicinal surface:

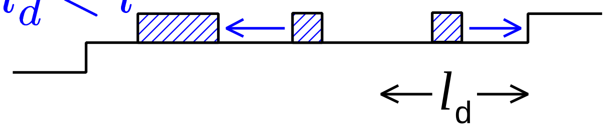


a) diffusion length $l_d > l$



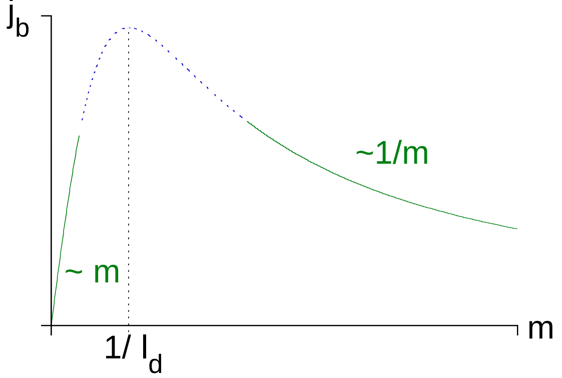
$\Rightarrow \quad j_b \sim l = 1/m$

b) $l_d < l$



$\Rightarrow \quad j_b \sim l_d \times l_d/l = m\, l_d^2$

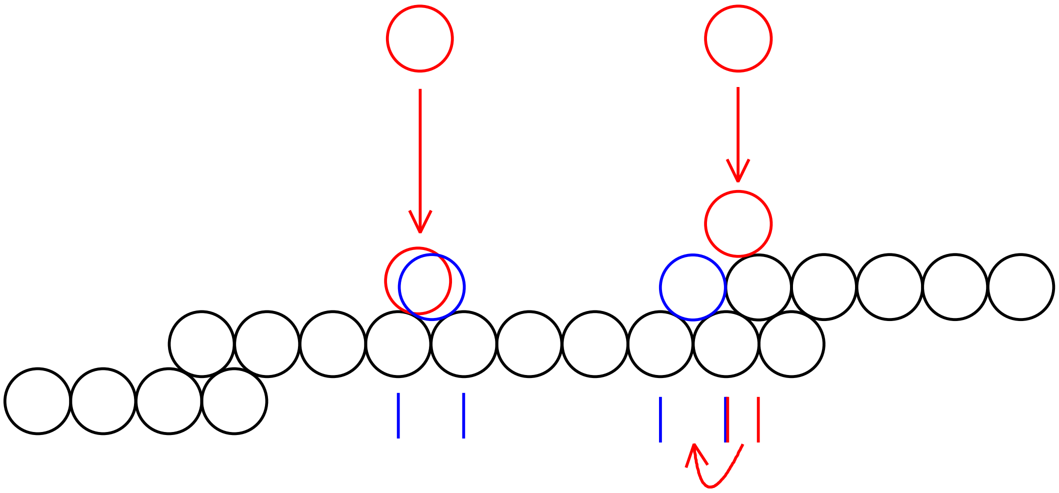
Interpolation:



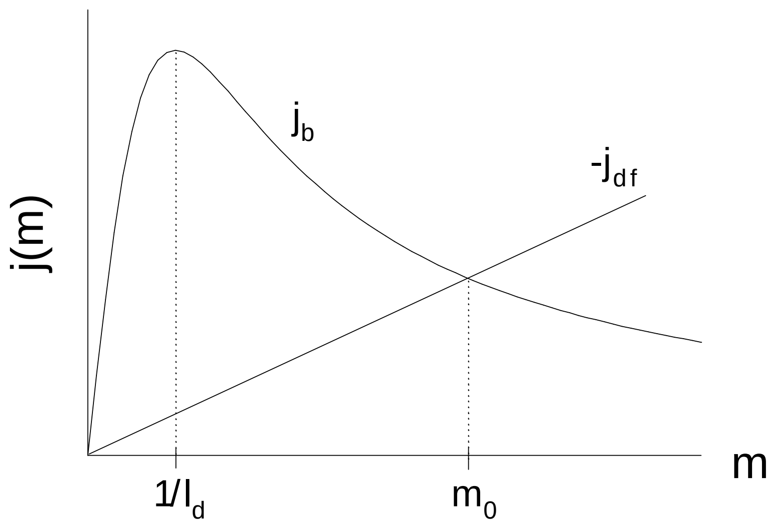
$j'(m = 0) > 0 \;\Rightarrow\;$ $m = 0$ surface becomes unstable

Downward Funneling

[M. Bartelt, J. Evans]



$$j_{df} = -F \times \frac{a}{2\ell} \times \frac{3}{4}a = -\frac{3}{8}F a\, m$$



2d Generalization

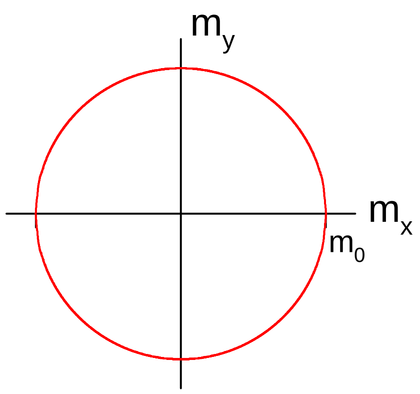
simplest choice: $\mathbf{j} \parallel \mathbf{m}$

$$\mathbf{j} = \mathbf{m} f(|\mathbf{m}|)$$

$$f(m_0) = 0$$

stable

orientations



$\rightarrow$ unrealistic

$\rightarrow$ does not describe coarsening correctly

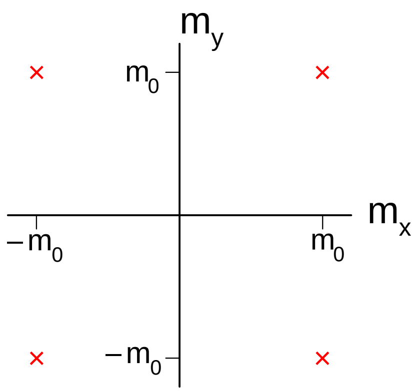
quadratic symmetry, (001) substrates

$$j_x = m_x f(m_x) \ , \quad j_y = m_y f(m_y)$$

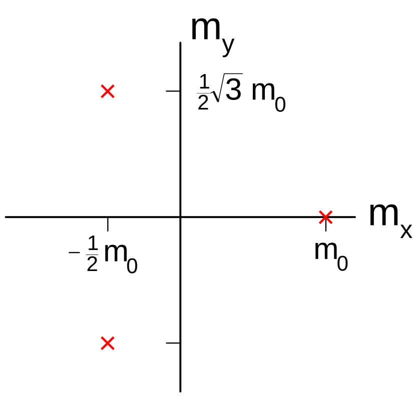
$$f(m_0) = 0$$

stable

orientations

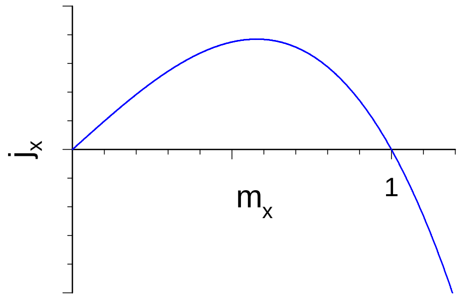


triangular symmetry, (111) substrates



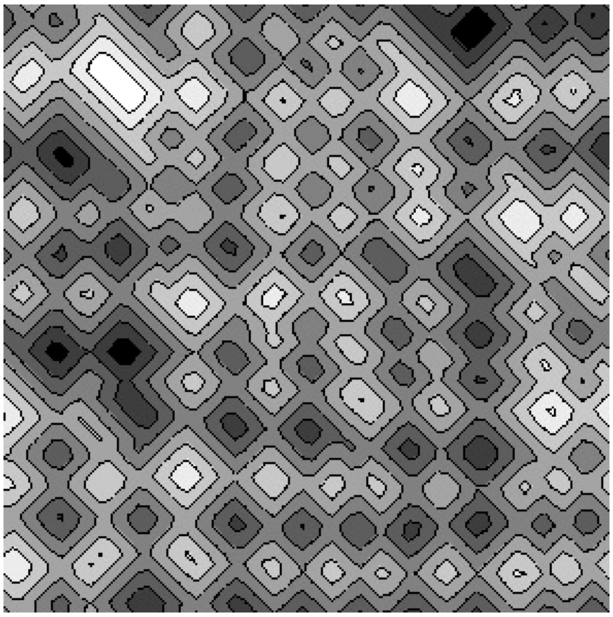
quadratic symmetry

$$\partial_t h = -\Delta\Delta h - \Delta h + \partial_x(\partial_x h)^3 + \partial_y(\partial_y h)^3 + \eta$$

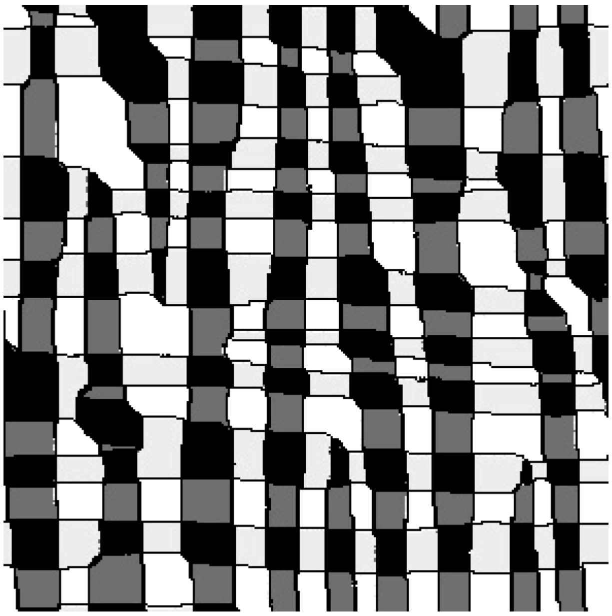


$j_x = m_x - m_x^3$

numerical solution



height



slope

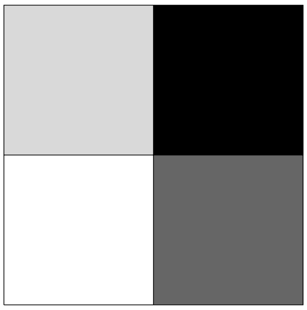
morphology coarsens:

average pyramid size $R(t) \sim t^n$

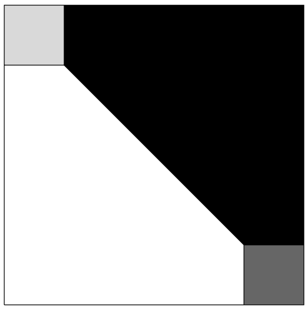
- why?
- $n =?$

two types of domain walls (line defects)

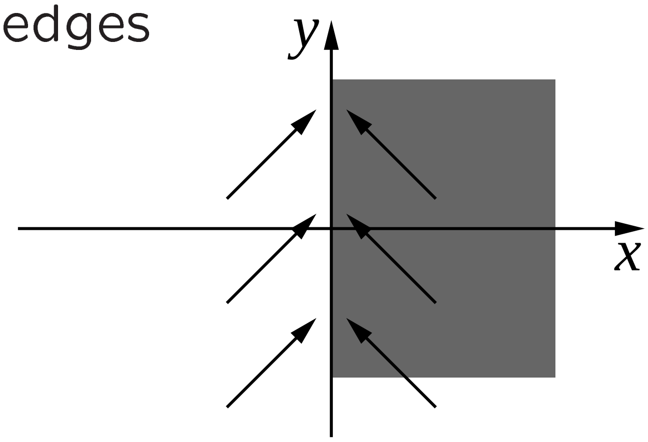
pyramids edges



roof tops



shape of pyramid edges



$$\partial_t h = -D\Delta\Delta h - a\left[m_0^2\Delta h - \partial_x(\partial_x h)^3 - \partial_y(\partial_y h)^3\right] + \eta$$

$\partial_t h = 0,\ m_y = m_0,\ \eta = 0$

$$\Rightarrow \qquad D\partial_x^2 m_x + a m_x(m_0^2 - m_x^2) = 0$$

solution: $m_x = \pm m_0 \tanh\dfrac{x}{\xi}$ with $\xi = \sqrt{\dfrac{2D}{a m_0^2}}$

$\xi \equiv$ domain wall width: intrinsic length scale

Numerical Methods: Finite Differences

$$h(\mathbf{r},t+\Delta t) = h(\mathbf{r},t) + \zeta(\mathbf{r},t) - \int_t^{t+\Delta t} d\tau \Big\{ D\Delta\Delta h(\mathbf{r},\tau) - \nabla\cdot\mathbf{j}[h(\mathbf{r},\tau)] \Big\}$$

with

$$\zeta(\mathbf{r},t) = \int_t^{t+\Delta t} d\tau\, \eta(\mathbf{r},\tau) \ , \quad \langle \zeta(\mathbf{x},t)\zeta(\mathbf{x}',t')\rangle = 2\epsilon\Delta t \delta_{t,t'}\delta(\mathbf{x}-\mathbf{x}')$$

Finite differences:

$\mathbf{r} = (x,y) = \Delta x(i,j),\ i,j = 1,2,\ldots,N$
pbc ($L = N\Delta x$ system size)

$$\zeta(\mathbf{x},t) = \left[\frac{2\epsilon\Delta t}{\Delta x^2}\right]^{1/2} \nu(\mathbf{x},t) \quad \text{with} \quad \langle \nu(\mathbf{x},t)\nu(\mathbf{x}',t')\rangle = \delta_{t,t'}\delta_{\mathbf{r},\mathbf{r}'}$$

choice of Δx: as large as possible

$\Rightarrow \quad \Delta x = 1.5 \quad (\xi = \sqrt{2})$

Euler method: largest time step $\Delta t = 0.125$

How to improve on the Euler Method?

(i) implicit methods (Crank-Nicholson)

difficulty: solving nonlinear equations

$\rightarrow$ possible only for small time steps

alternative: use implicit method only for linear terms

$\rightarrow$ stability not very good

(ii) predictor-corrector methods

disadvantage: two evaluations of right-hand side per time step

e.g., $\partial_t y = f[y]$

predictor AB(2)

$$\tilde{y}(t+\Delta t) = y(t) + \frac{\Delta t}{2}\Big\{3f[y(t)] - f[y(t-\Delta t)]\Big\}$$

corrector AM(3)

$$y(t+\Delta t) = y(t) + \frac{\Delta t}{12}\Big\{5f[\tilde{y}(t+\Delta t)] + 8f[y(t)] - f[y(t-\Delta t)]\Big\}$$

advantages: more stable: $\Delta t = 0.2$

higher accuracy

Spectral Methods

$\widehat{h}(\mathbf{k},t) = \int d^2x\, e^{i\mathbf{k}\,\mathbf{x}}\, h(\mathbf{x},t)$ Fourier transform

$$\mathbf{k} = (k_x, k_y)\ , \qquad k_{x,y} = \frac{2\pi}{L} n\ , \qquad n = 1, 2, \ldots$$

$$\Rightarrow \quad \partial_t \widehat{h} = -Dk^4 \widehat{h} - \widehat{\nabla \cdot \mathbf{j}} + \widehat{\eta}$$

or

$$\widehat{h}(\mathbf{k}, t + \Delta t) = \widehat{h}(\mathbf{k}, t) e^{-Dk^4 \Delta t} + \widehat{\zeta}(\mathbf{k}, t) - \int_t^{t+\Delta t} d\tau\, e^{-Dk^4(t+\Delta t - \tau)} \widehat{\nabla \cdot \mathbf{j}}(\mathbf{k}, \tau)$$

with

$$\widehat{\zeta}(\mathbf{k}, t) = \left[\frac{\epsilon L^2}{Dk^4} \left(1 - e^{-2Dk^4 \Delta t} \right) \right]^{1/2} \widehat{\nu}(\mathbf{k}, t)$$

and

$$\langle \widehat{\nu}(\mathbf{k}, t) \widehat{\nu}(\mathbf{k}', t') \rangle = \delta_{t,t'} \delta_{\mathbf{k},-\mathbf{k}'}$$

Euler method is unconditionally stable ($\Delta t = 4$)!

accuracy lousy

$\Rightarrow$ reduce time step: $\Delta t \simeq 0.2 \quad \rightarrow \quad$ slow

<u>ABM</u>

$$\int_t^{t+\Delta t} d\tau \, e^{-Dk^4(t+\Delta t-\tau)} \widehat{\nabla \cdot} \mathbf{j}(\mathbf{k},\tau)$$
$$\simeq \frac{\Delta t}{12} i\mathbf{k} \cdot \Big\{ 5\hat{\mathbf{j}}[\tilde{h}(t)] + 8\hat{\mathbf{j}}[h(t)]e^{-Dk^4\Delta t} - \hat{\mathbf{j}}[h(t-\Delta t)]e^{-2Dk^4\Delta t} \Big\}$$

with

$$\hat{\tilde{h}}(\mathbf{k},t) = \hat{h}(\mathbf{k},t)e^{-Dk^4\Delta t} + \hat{\zeta}(\mathbf{k},t) - \frac{\Delta t}{2} e^{-Dk^4\Delta t} i\mathbf{k} \cdot \Big\{ 3\hat{\mathbf{j}}[h(t)] - \hat{\mathbf{j}}[h(t-\Delta t)]e^{-Dk^4\Delta t} \Big\}$$

no longer unconditionally stable ($\Delta t \simeq 1$)

accuracy good for $\Delta t \simeq 0.5$

Coarsening: what is the driving mechanism?

deterministic dynamics: $\eta = 0$

Correlation functions:

– height-height and slope-slope correlation functions

$$G(\mathbf{r},t) = \frac{1}{L^2}\sum_{\mathbf{x}}\langle h(\mathbf{r}+\mathbf{x},t)h(\mathbf{x},t)\rangle$$

$$C(\mathbf{r},t) = \frac{1}{L^2}\sum_{\mathbf{x}}\langle \mathbf{m}(\mathbf{r}+\mathbf{x},t)\mathbf{m}(\mathbf{x},t)\rangle$$

– structure factor

$$S(\mathbf{k},t) = \langle \widehat{h}(\mathbf{k},t)\widehat{h}(-\mathbf{k},t)\rangle$$

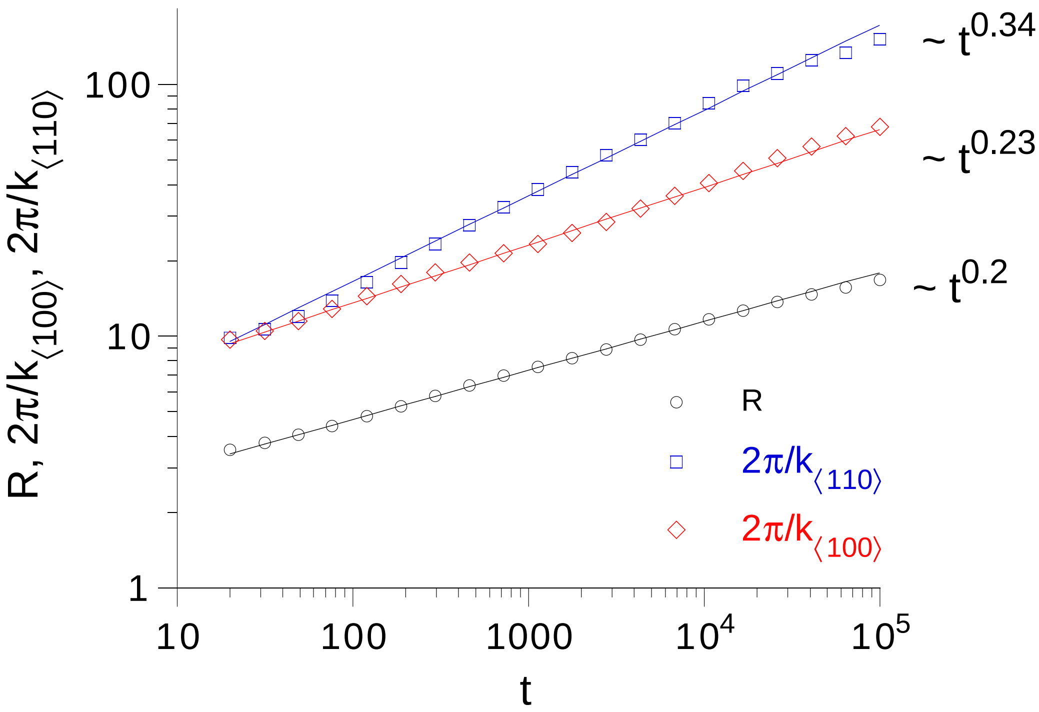
characteristic length scale:

- $R(t)$ first zero of $C(\mathbf{r},t)$ in the $\langle 110\rangle$ direction
- $k_{\langle 100\rangle}(t) = \frac{\sum_{k_x} k_x S(\mathbf{k}=(k_x,0),t)}{\sum_{k_x} S(\mathbf{k}=(k_x,0),t)}$, first moment of $S(\mathbf{k},t)$
- $k_{\langle 110\rangle}(t) = \frac{\sum_{k} k S(\mathbf{k}=(k,k),t)}{\sum_{k} S(\mathbf{k}=(k,k),t)}$

same time dependence?

scaling?

Result $(\eta = 0)$



$\Rightarrow$ two different length scales (at least)

where do they come from?

$R(t) \sim$ pyramid size

$k_{\langle 100 \rangle}(t)$, $k_{\langle 110 \rangle}(t)$?

Tail of the Structure Factor

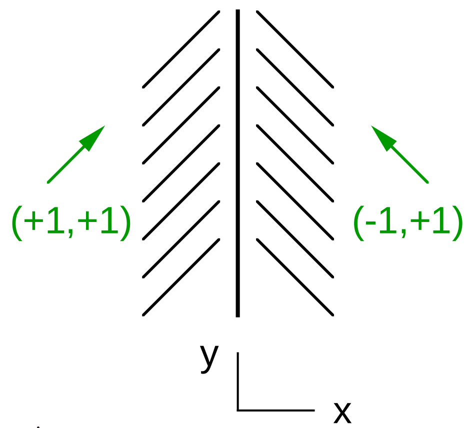
Calculate $S(\mathbf{k},t)$ for $k \gg 2\pi/R(t)$:

assume $S(\mathbf{k},t) \sim k^{-\gamma}$

$\Rightarrow$ origin: singular part of $G(\mathbf{r},t)$ for $r \ll R(t)$

$\xi \ll r \ll R(t)$: study single edge:

$$h(x,y) = -|x| + y$$



analogously for edges in x and $\langle 110\rangle$ directions :

$$\begin{aligned} C(\mathbf{r},t) &= -2(|x|+|y|)\rho_{\langle 100\rangle}(t) \\ &\quad -2\sqrt{2}(|x+y|+|x-y|)\rho_{\langle 110\rangle}(t) \end{aligned}$$

density of edges in the

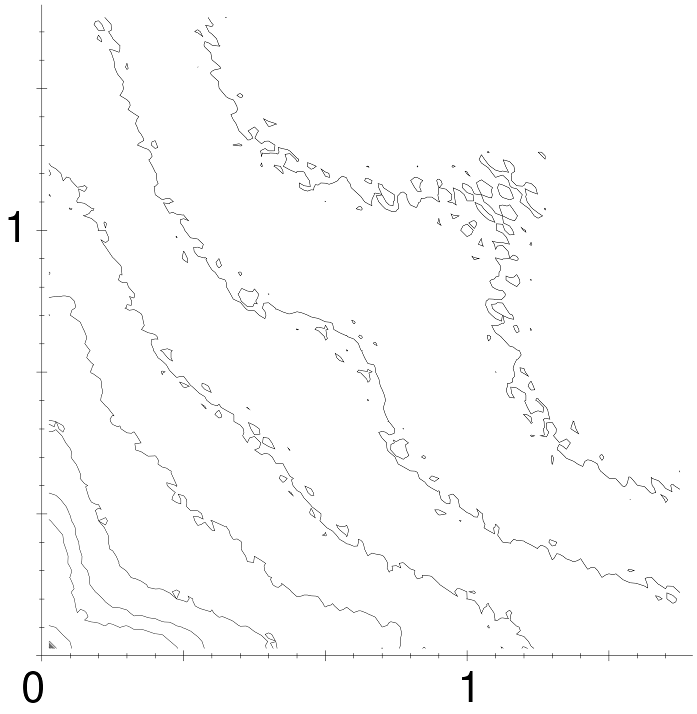
$\langle 100\rangle$ and $\langle 110\rangle$ directions

$+$ analytic terms

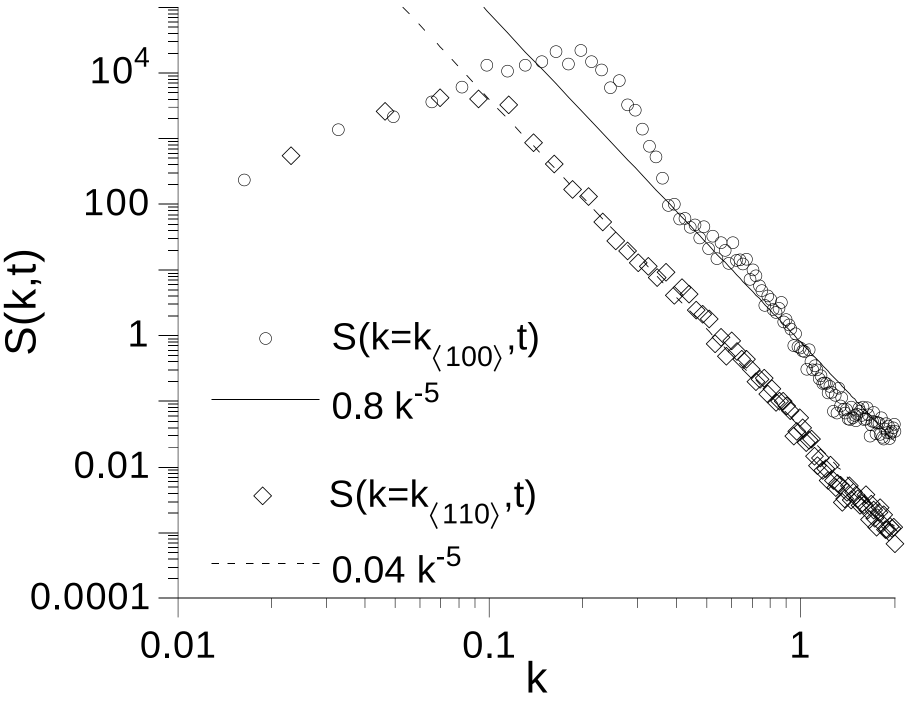
$2\pi/\xi \gg k \gg 2\pi/R(t)$, $\mathbf{k} = k(\cos\varphi, \sin\varphi)$

$$\Rightarrow \quad S(\mathbf{k},t) \sim \frac{1}{k^5}\Big[\rho_{\langle 100\rangle}(t)\sum_{\nu=0}^{3}\delta(\varphi-\nu\frac{\pi}{2}) + 2\rho_{\langle 110\rangle}(t)\sum_{\nu}\delta(\varphi-\frac{\pi}{4}-\nu\frac{\pi}{2})\Big] + \ldots$$

contourplot of $\log[S(\mathbf{k}, t)]$



$S(\mathbf{k}_{\langle 100\rangle}, t),\ S(\mathbf{k}_{\langle 110\rangle}, t)$

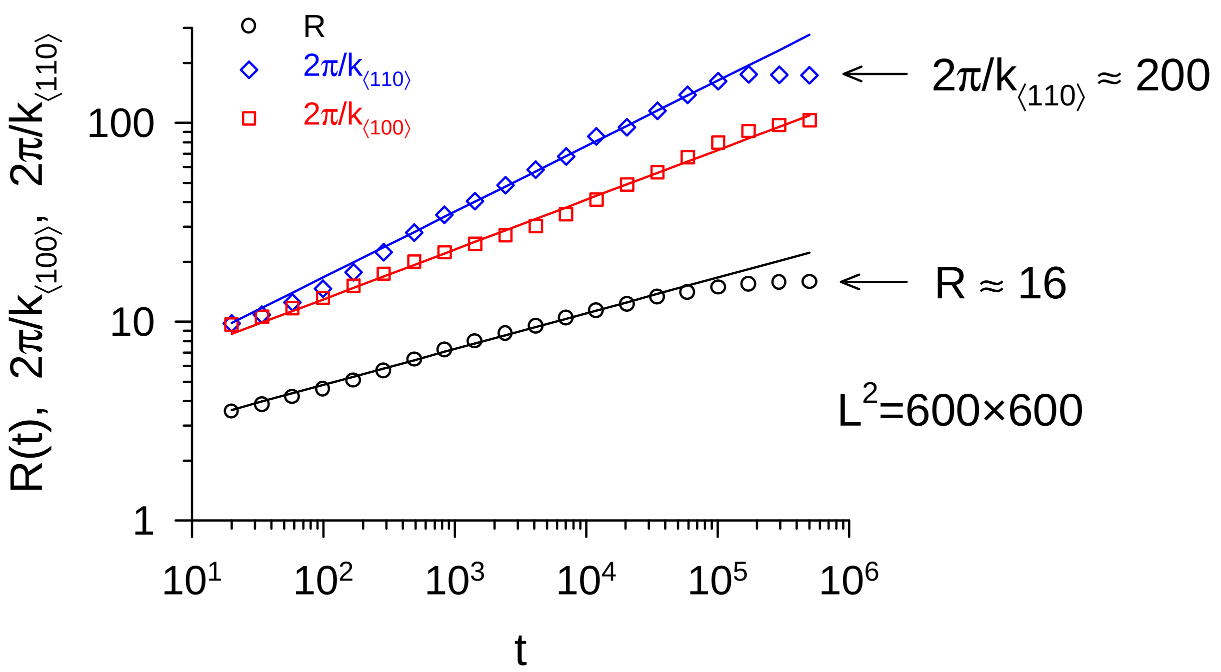


Coarsening Mechanism

- a configuration without "roof tops" does not coarsen ($\eta = 0$)

  $\Rightarrow$ metastable states

- system orders by eliminating diagonal edges: roof tops $\leftrightarrow$ "defects" in pyramid lattice



Why pyramids and not roof tops?

$$j_x = m_x - m_x^3 - b m_x m_y^2$$

$$j_y = m_y - m_y^3 - b m_y m_x^2$$

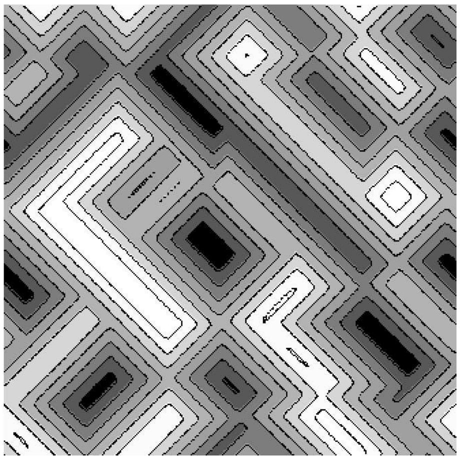
up to now: $b = 0$

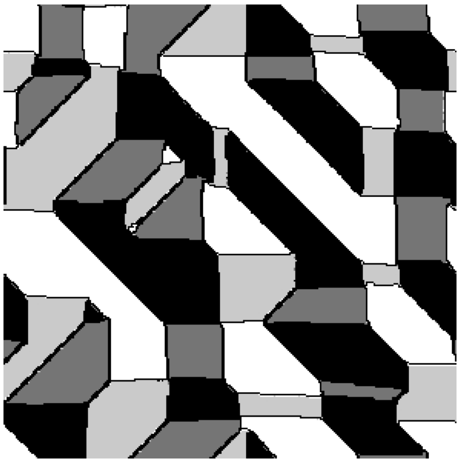
$b \neq 0$ $(-1 < b \leq 1)$

$b > -3/4$: $\Rightarrow$ pyramids

$-1 < b < -3/4$: $\Rightarrow$ roof tops

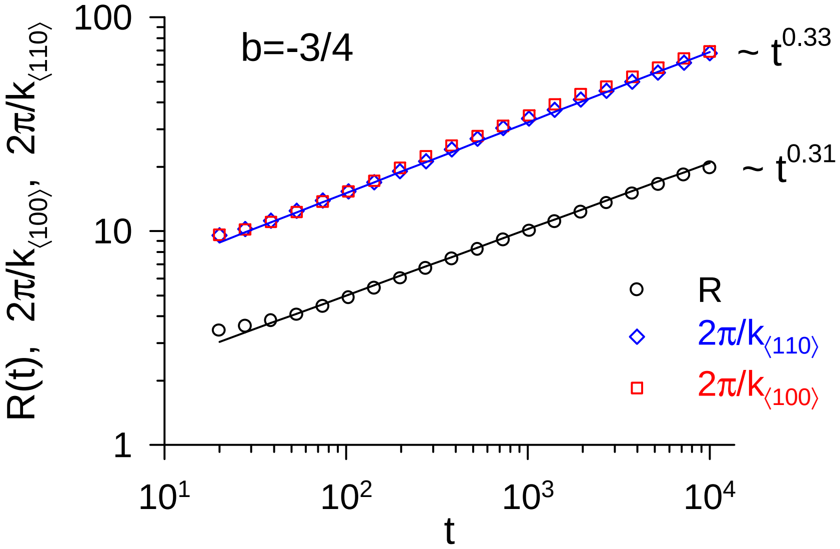
$b = -3/4$





- both types of edges are equally favoured

  $\Rightarrow$ fast coarsening

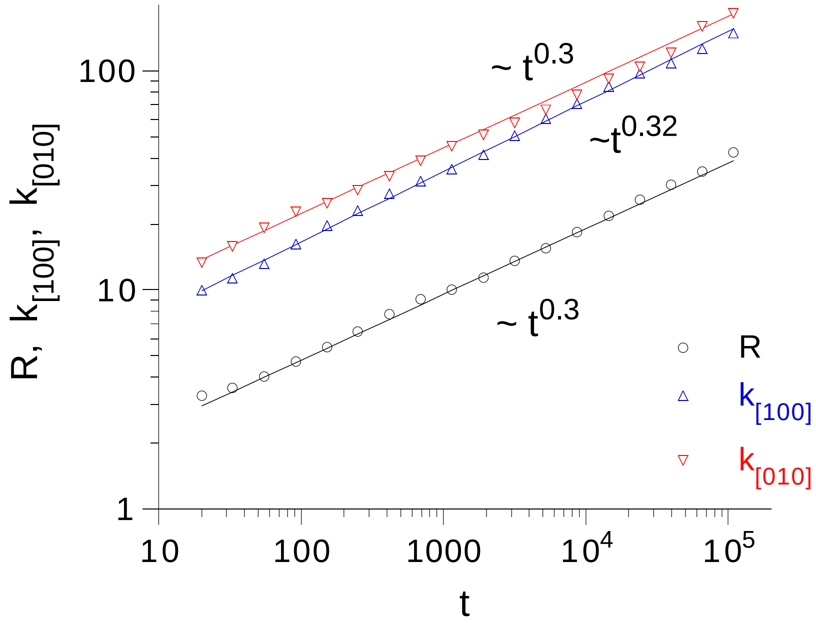


Triangular Symmetry

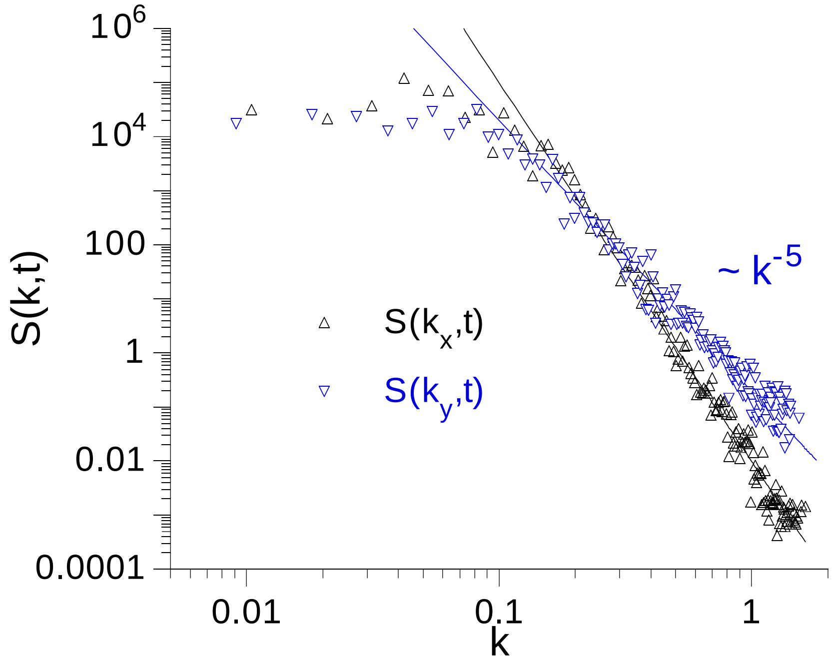
- all edges are equivalent (no roof tops)

  no metastable states

  $\Rightarrow$ fast coarsening



$S(\mathbf{k}, t)$ anisotropic



Conclusion

- Theory of coarsening must include anisotropies

  quadratic symmetry

  - two length scales with different time dependencies
  - standard scaling assumption not valid
  - metastable states

  triangular symmetry

  - no metastable states $\rightarrow$ fast coarsening
  - correlation functions still anisotropic

- Numerical Methods:
  - It is difficult to improve on the Euler method suggestions?
  - Predictor-Corrector methods work fairly well

- open problems:
  - time dependence of $k_{\langle 110 \rangle}$
  - microscopic explanation of the "b" parameter $\rightarrow$ $2d$ generalization of BCF theory