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SMR 1746 - 9

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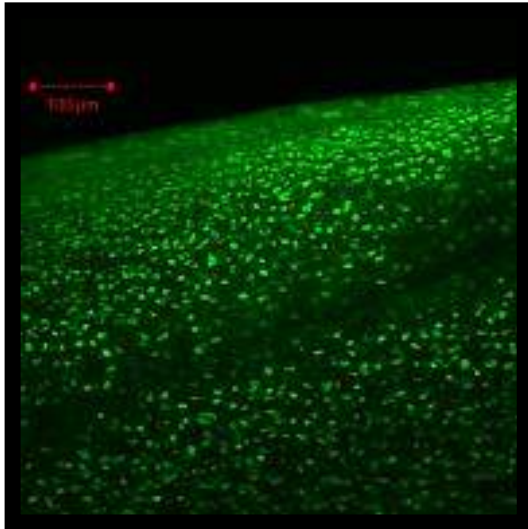
WORKSHOP ON DRIVEN STATES IN SOFT AND BIOLOGICAL MATTER  
18 - 28 April 2006

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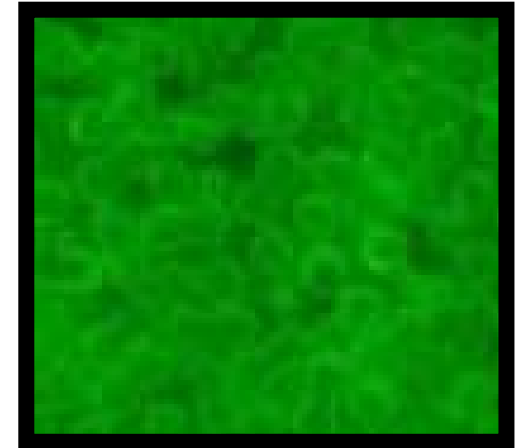
***Using Confocal Microscopy to Investigate Complex Matter Systems***  
*(for Talk on "Using Confocal Microscopy to Investigate Yielding in Colloidal Crystals")*

***Itai COHEN***  
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***Ithaca, NY 14853, U.S.A.***

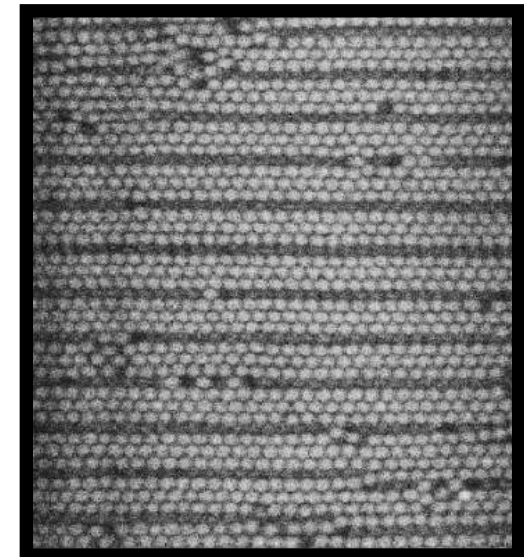
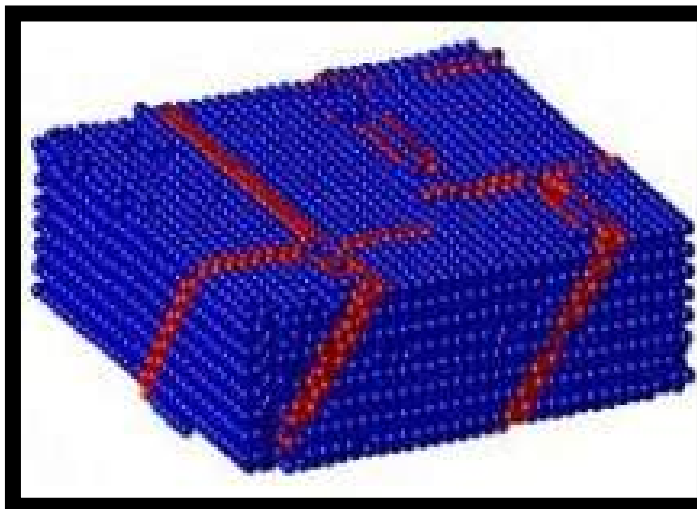
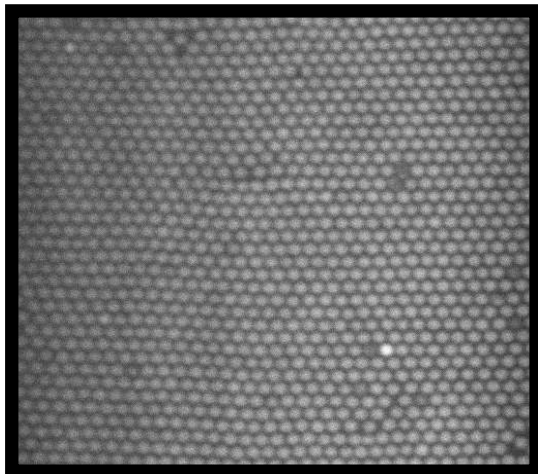
# Using Confocal Microscopy to Investigate Complex Matter Systems



Role of material  
micro-structure in  
determining  
macroscopic flows



**Itai Cohen**



# Why Colloids?

- **Industrially important**
  - Paints, coatings, lubrication, bio-rheology, electro-optical devices
- **Index and density matched to solvent**
- **Ising model of soft condensed matter:**
- **Rheology:**
  - **Shear thickening**
  - **Shear thinning**
  - **Shear banding**
  - **Etc.**
- **Models for atomic systems**

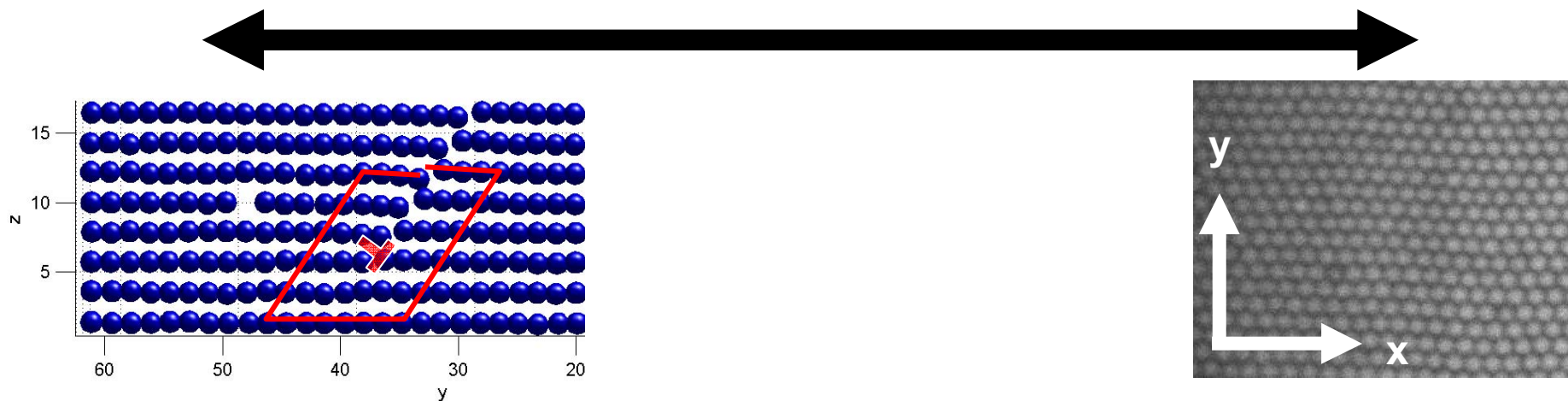
(E. R. Weeks, U. Gasser, D. A. Weitz, P. N. Pusey, and others)

# Yielding Changes With Time Scales

Slow

Shear Rate

Fast

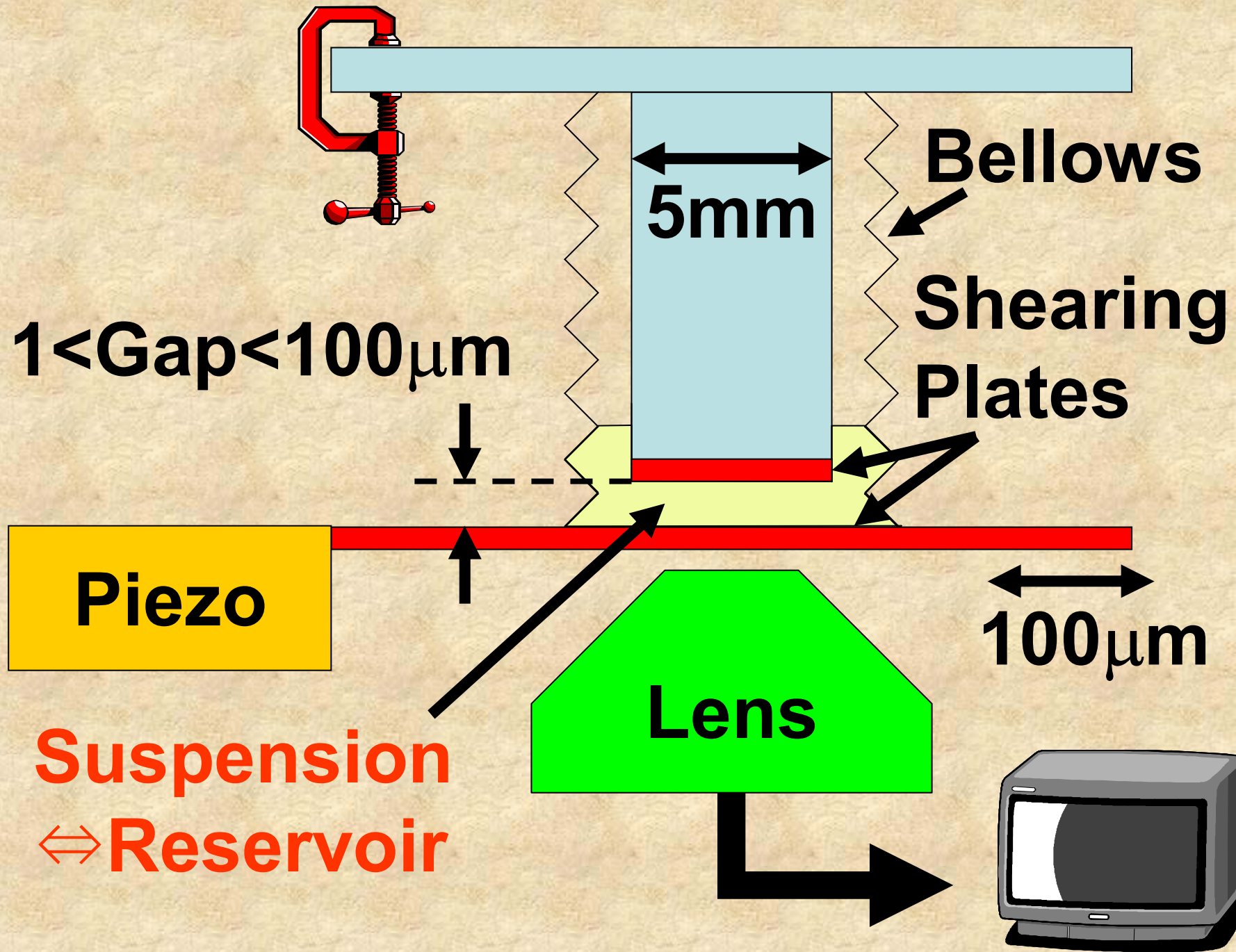


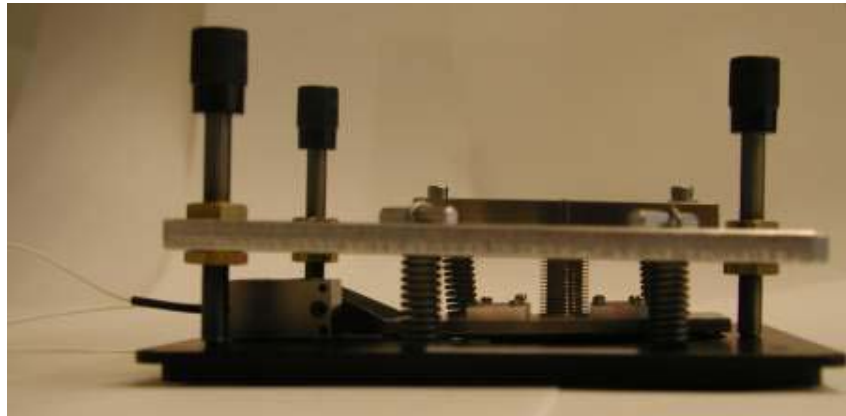
- Slow / Equilibrium: Brownian – Dislocations
- Fast / Non\_equilibrium: Fluidization hcp layers
- Study Equilibrium → Non-equilibrium

# Other Important Parameters

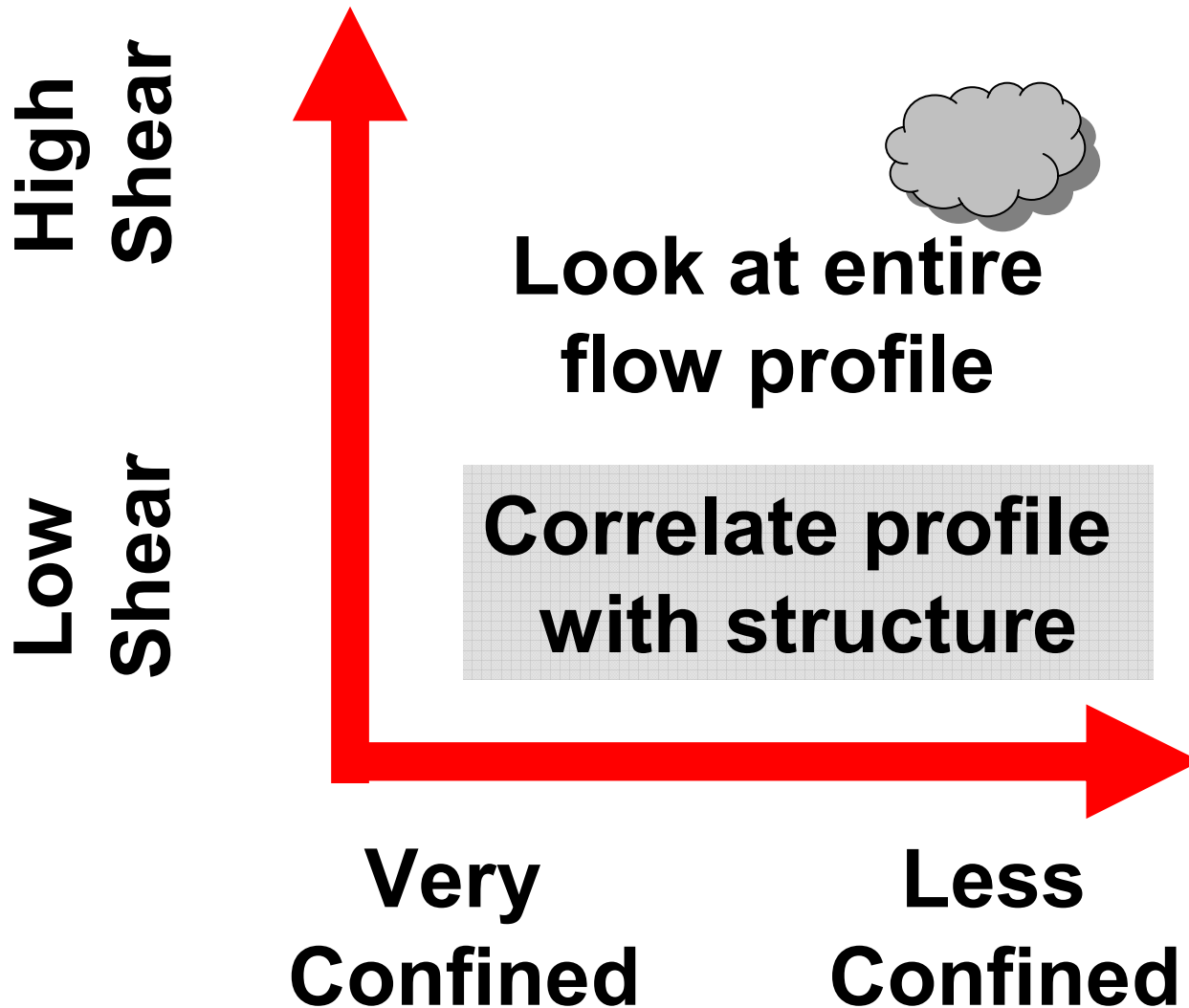
- **Particle Shape: Spheres, diameter = 1.4  $\mu\text{m}$**
- **Particle interaction: Slightly charged**
- **Volume fraction:  $\Phi = 61\%$**
- **Boundary conditions: Smooth plates**
  
- **Confinement**
  - **affects rheology of atomically thin films (Israelachvili, and others)**
  - **What about colloids?**







# High Shear & Less Confined



$5\text{Hz} < f < 30\text{Hz}$   
 $\text{Amp.} \cong 30\mu\text{m}$   
 $\text{Gap} \cong 70\mu\text{m}$

Diffusion:

$$D_0 = kT/6\pi\eta a$$

$$t_{a0} = \pi\eta a^3/kT$$

For  $a = 0.71\mu\text{m}$

$$t_{a0} \cong 0.7\text{ sec}$$

$$\text{Pe} = \gamma f t_{a0} \cong 10$$

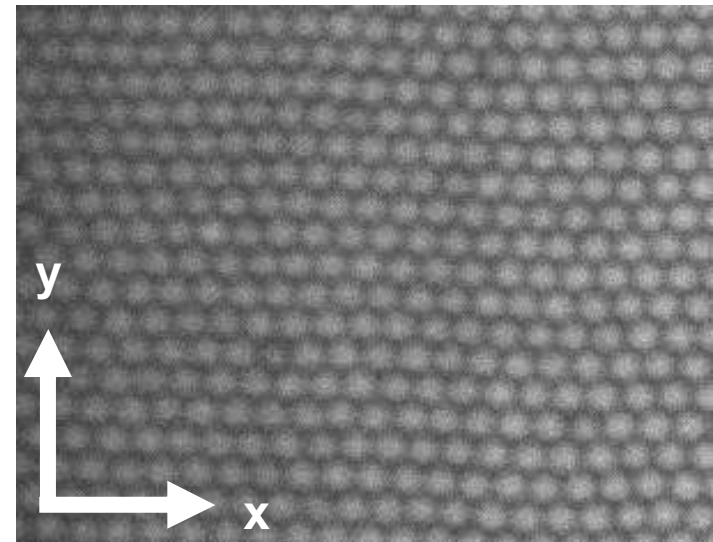
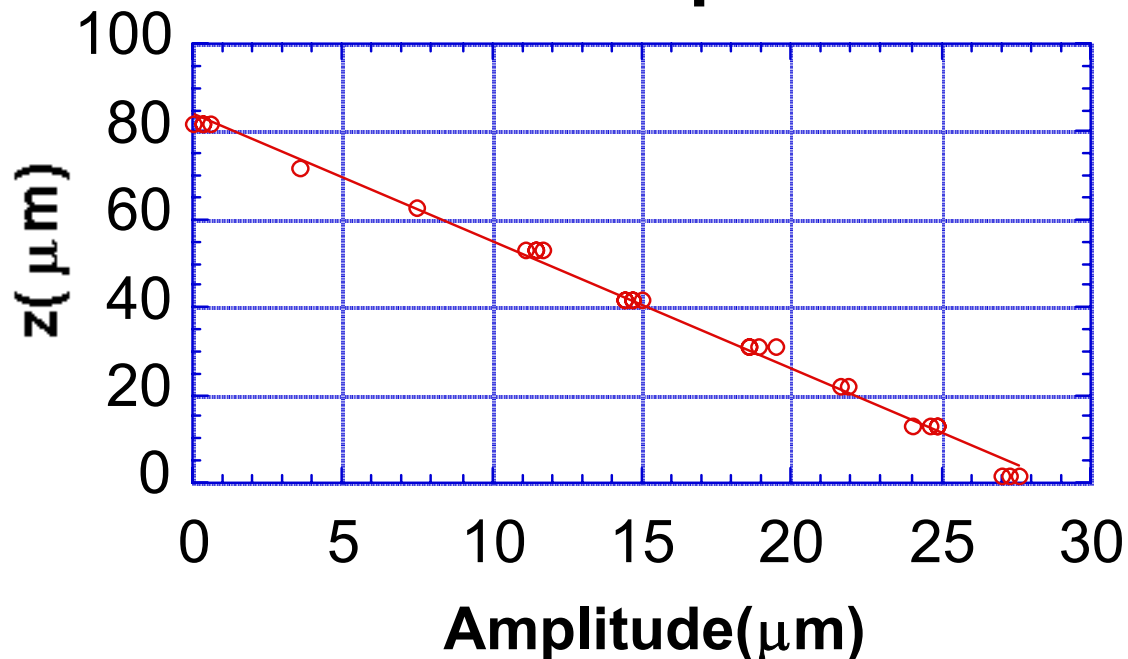


# Disordered Suspension $\rightarrow$ HCP Layers

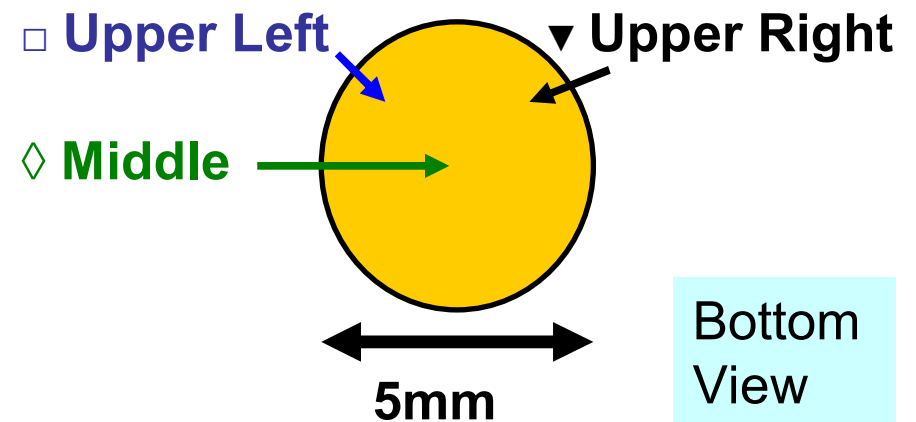
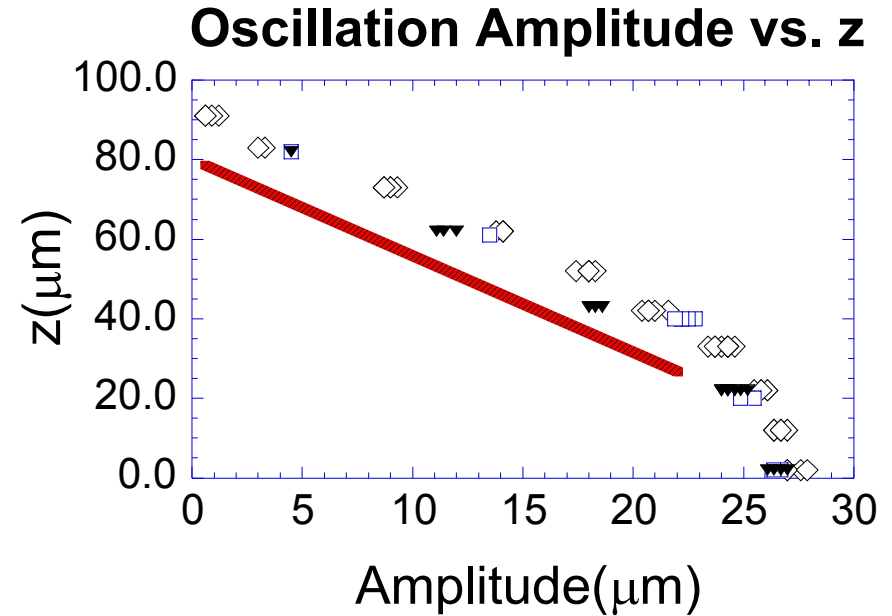
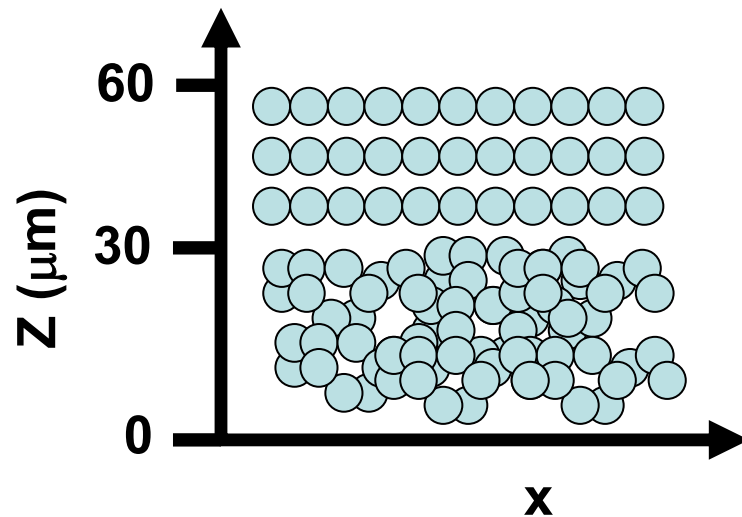
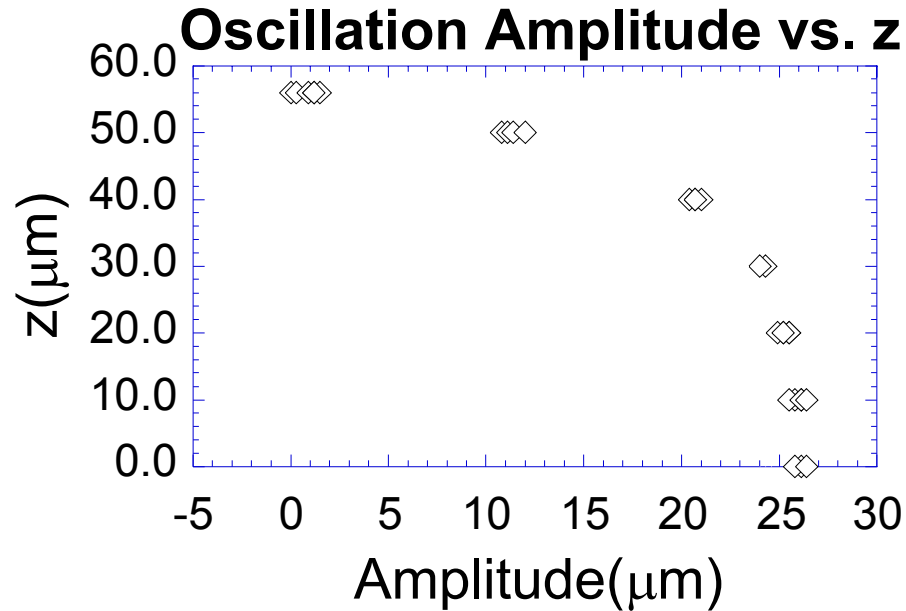
(Ackerson, Vermold and others)

Track particle motion at different heights,  $z$   
Observe linear shear gradient

Oscillation amplitude vs.  $z$



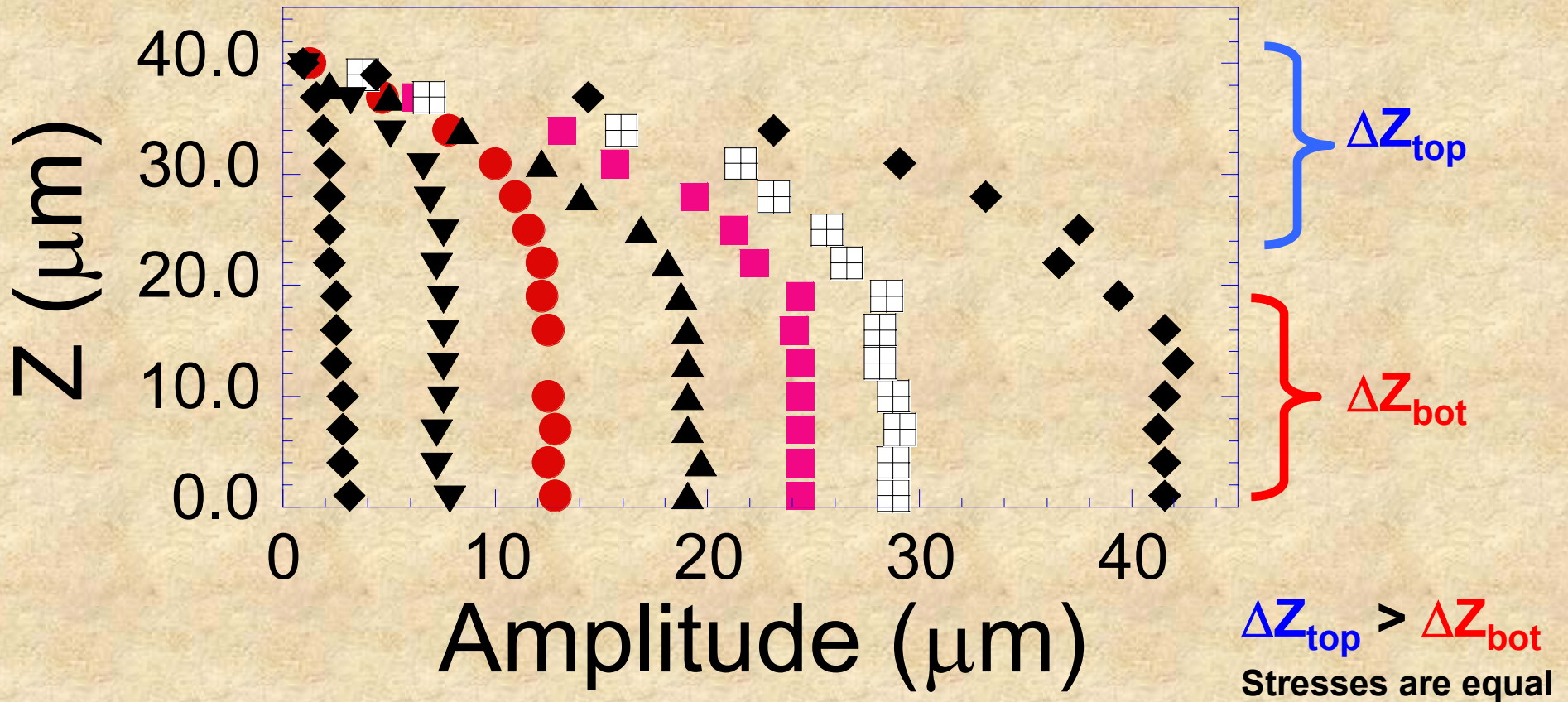
# Local Disorder → Global Coupling



# Shear Banding

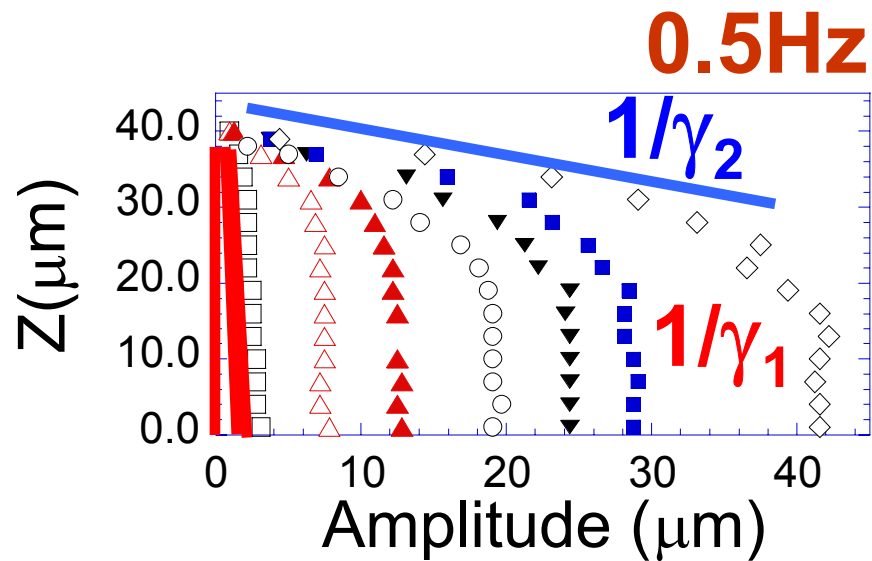
Fix frequency, increase strain  
Observe flow profile

0.5Hz

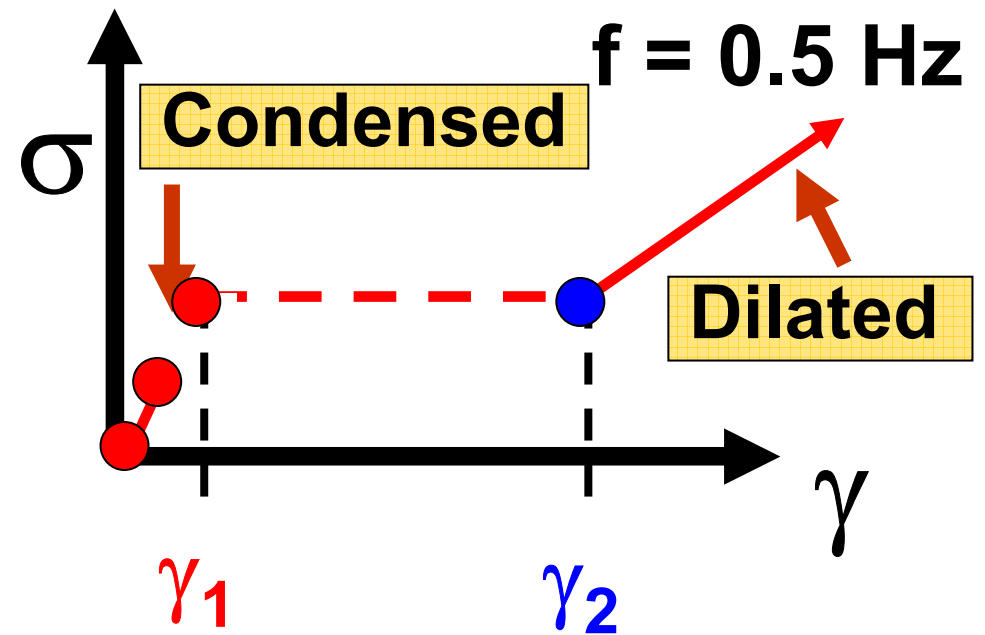


# Shear Thinning Rheology?

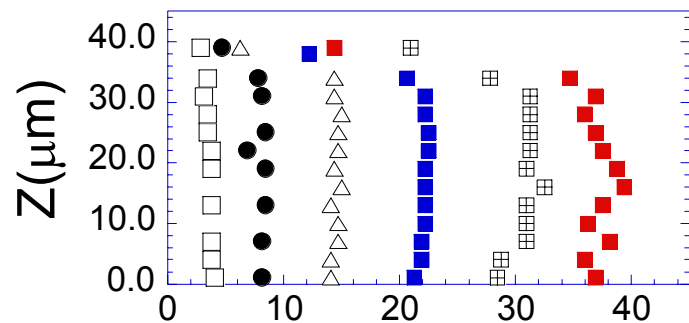
Data



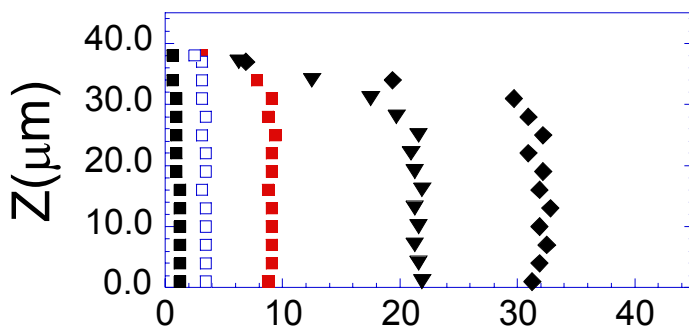
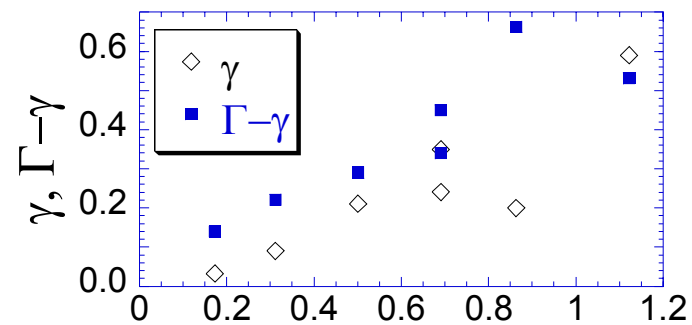
Schematic: stress vs. strain



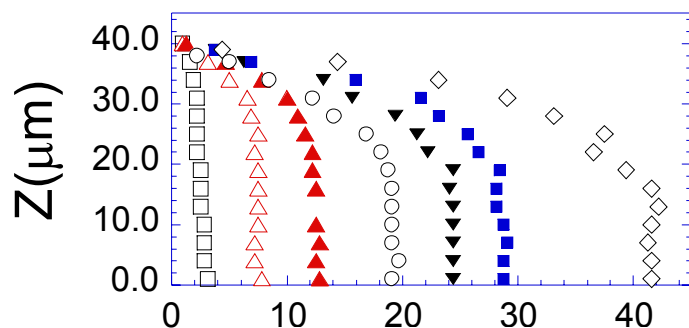
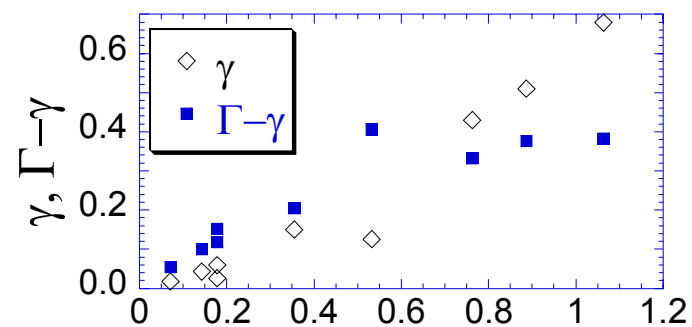
Shear bands tell us about material rheology



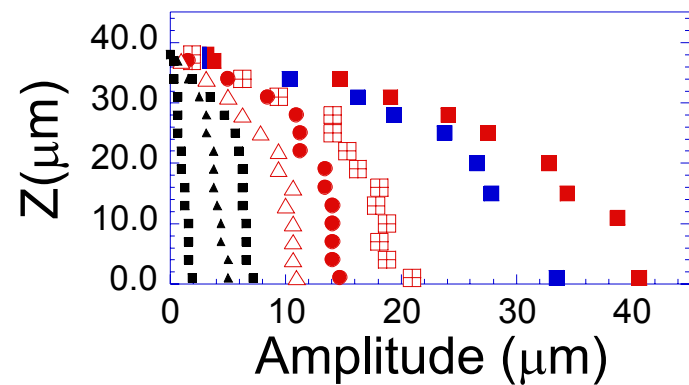
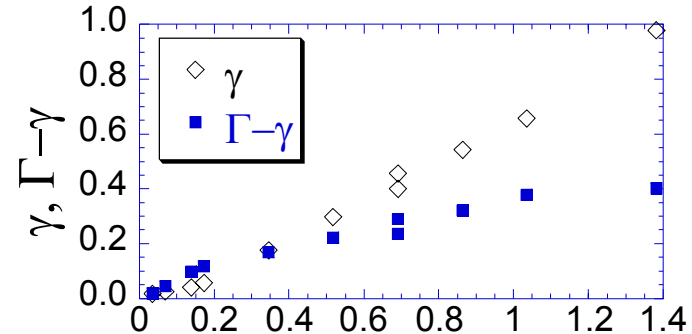
**0.02Hz**



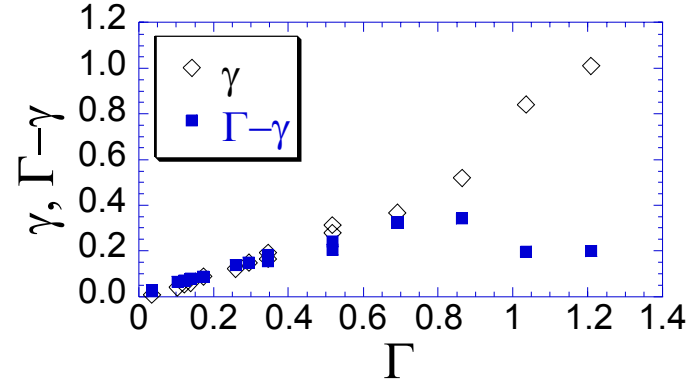
**0.11Hz**

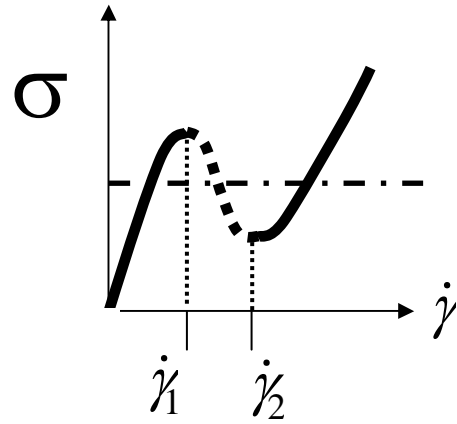


**0.5Hz**



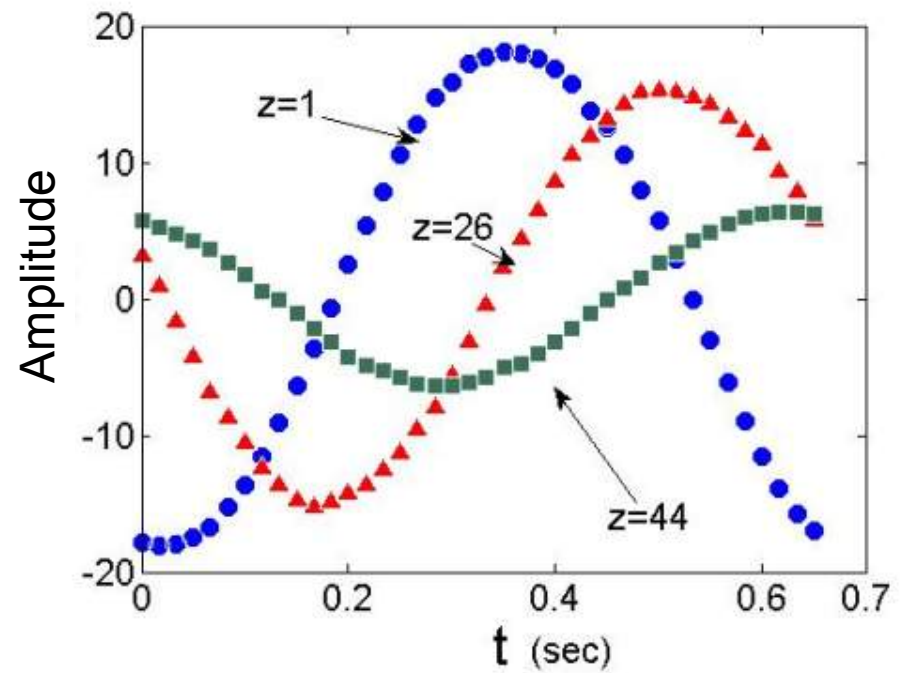
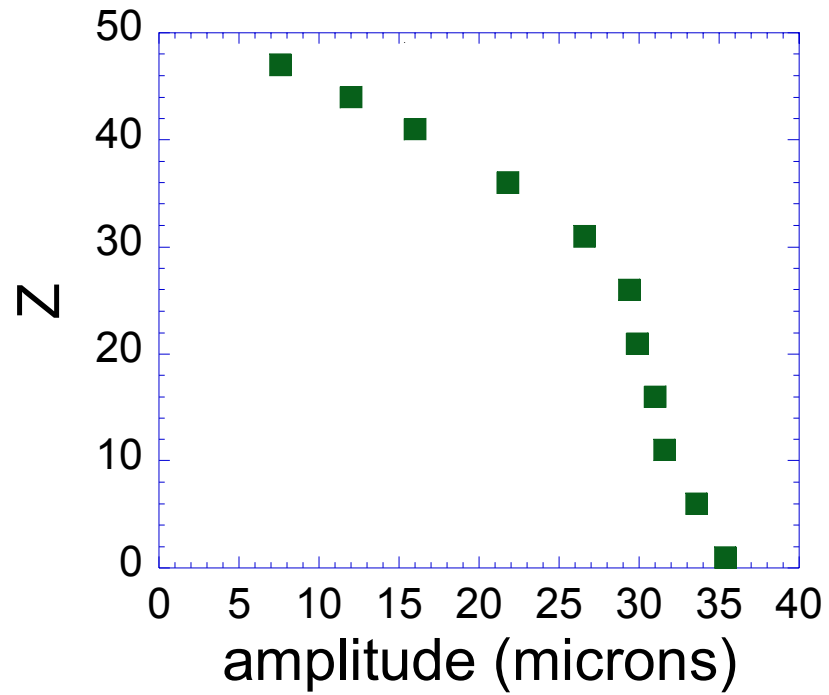
**1.5Hz**





## Beny Davidovich:

- Nonlinearities  $\rightarrow$  higher harmonics
- Discontinuous flows for oscillatory shear





# Stresses

For shear flow viscous and viscoelastic (Kelvin):

$$\sigma_s = G \underbrace{\partial u_s / \partial z}_\gamma + \eta_s \underbrace{\partial(\partial u_s / \partial t)}_{\dot{\gamma}} / \partial z$$

$$\sigma_f = \eta_f \underbrace{\partial(\partial u_f / \partial t)}_{\dot{\gamma}} / \partial z$$

# Linear Model

Solutions to  $\nabla \sigma = 0$  are of the form:

$$u_s = r_s(z) e^{i[\phi_s(z) + \omega t]}$$

$$u_f = r_f(z) e^{i[\phi_f(z) + \omega t]}$$

**Continuity at  $z=z^*$**

$$u_s(z = z^*) = u_f(z = z^*)$$

**Eq. (1,2)**

**Stress Balance**

$$G \partial u_s / \partial z + \eta_s \partial(\partial u_s / \partial t) / \partial z = \eta_f \partial(\partial u_f / \partial t) / \partial z$$

**Eq. (3,4)**

**Selection problem**

**Transition stress = yield stress ( $\delta$ ) Eq. (5) ?**

**Displacement**

$r_{s,f}(z)$  are at most linear in  $z$  (the usual)  
Generally,  $\phi$  is also a function of  $z$

**Five Unknowns:**

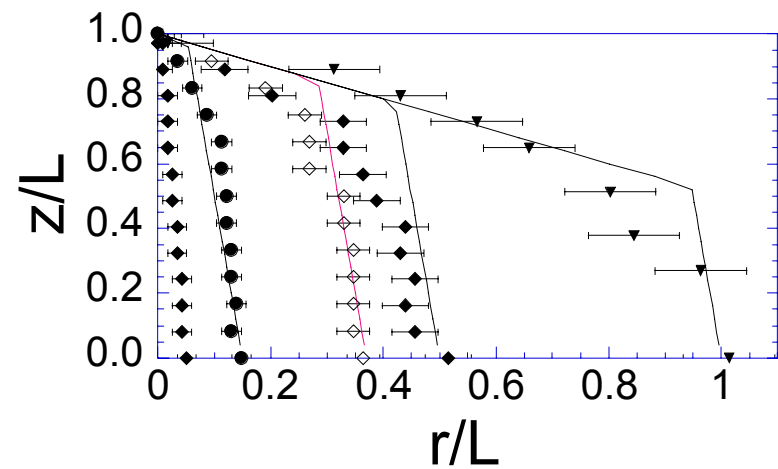
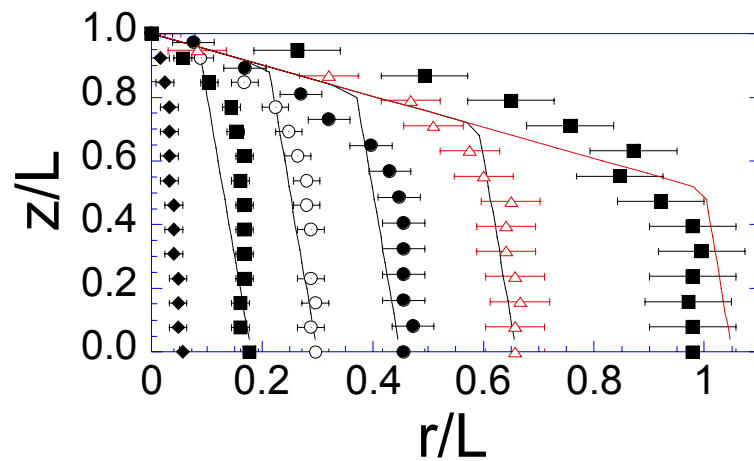
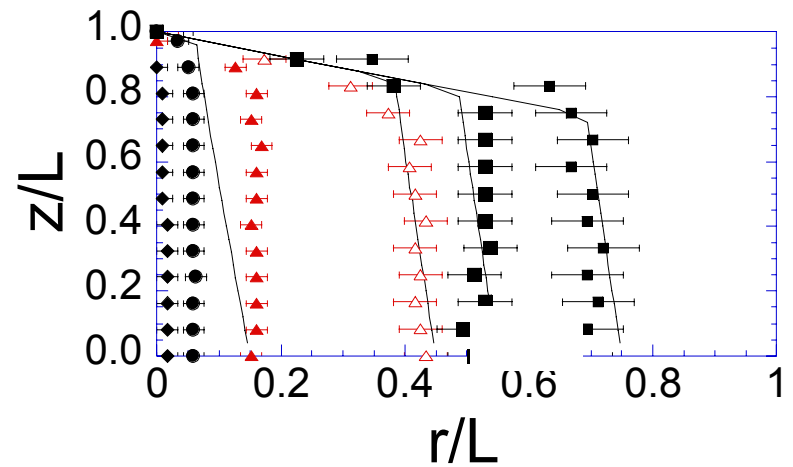
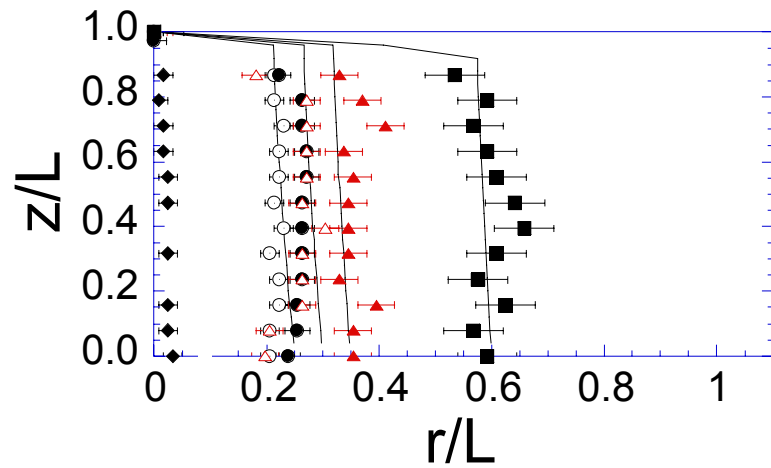
$r_s(z)$ ,  $\phi_s(z)$ ,  $r_f(z)$ ,  $\phi_f(z)$ ,  $z^*$

How well do we do?

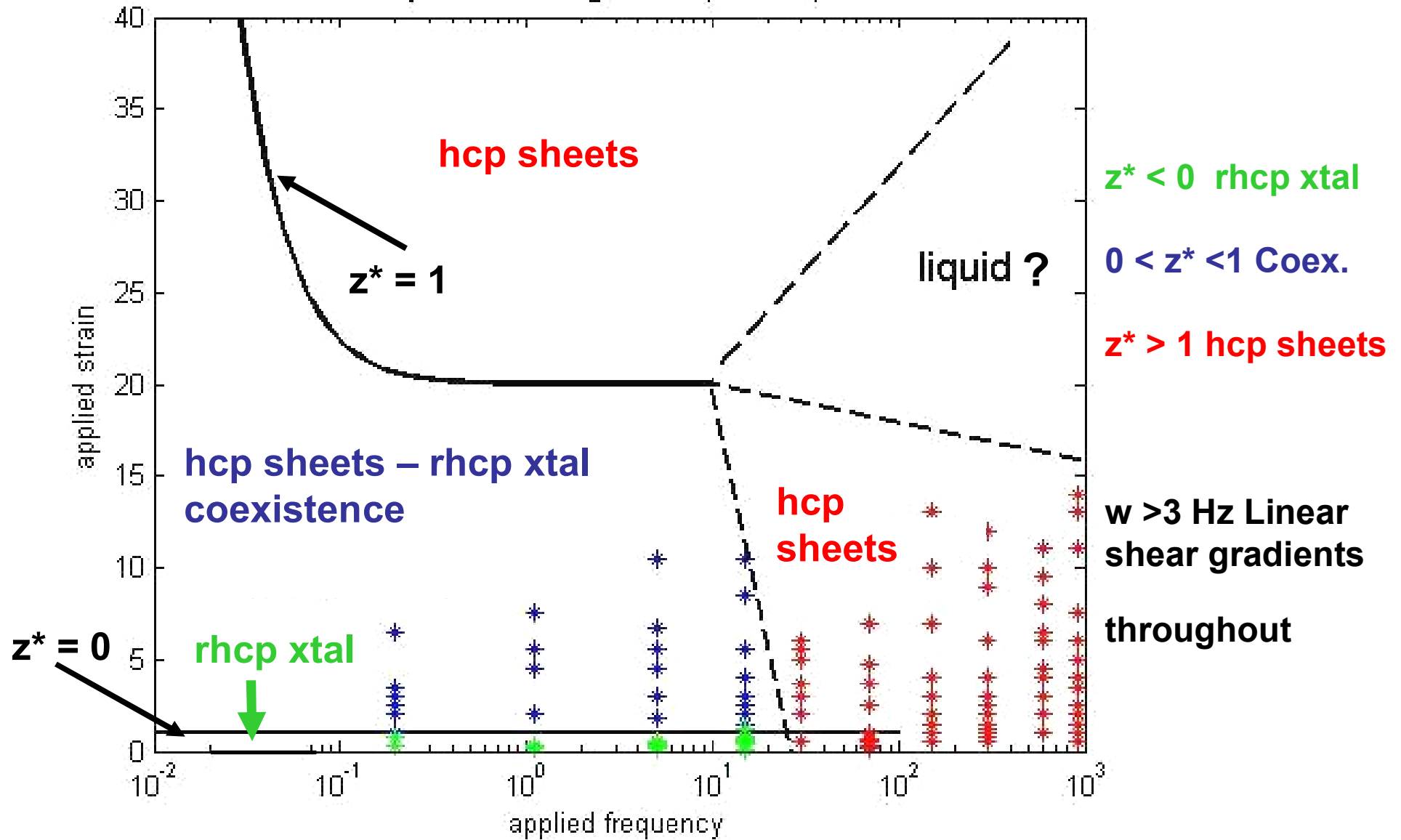
If we float  $\eta_s$ ,  $\eta_f$ ,  $\delta \rightarrow$   
too well

If we fix  $\eta_s$ ,  $\eta_f$ ,  $\delta$  to be  
constant throughout

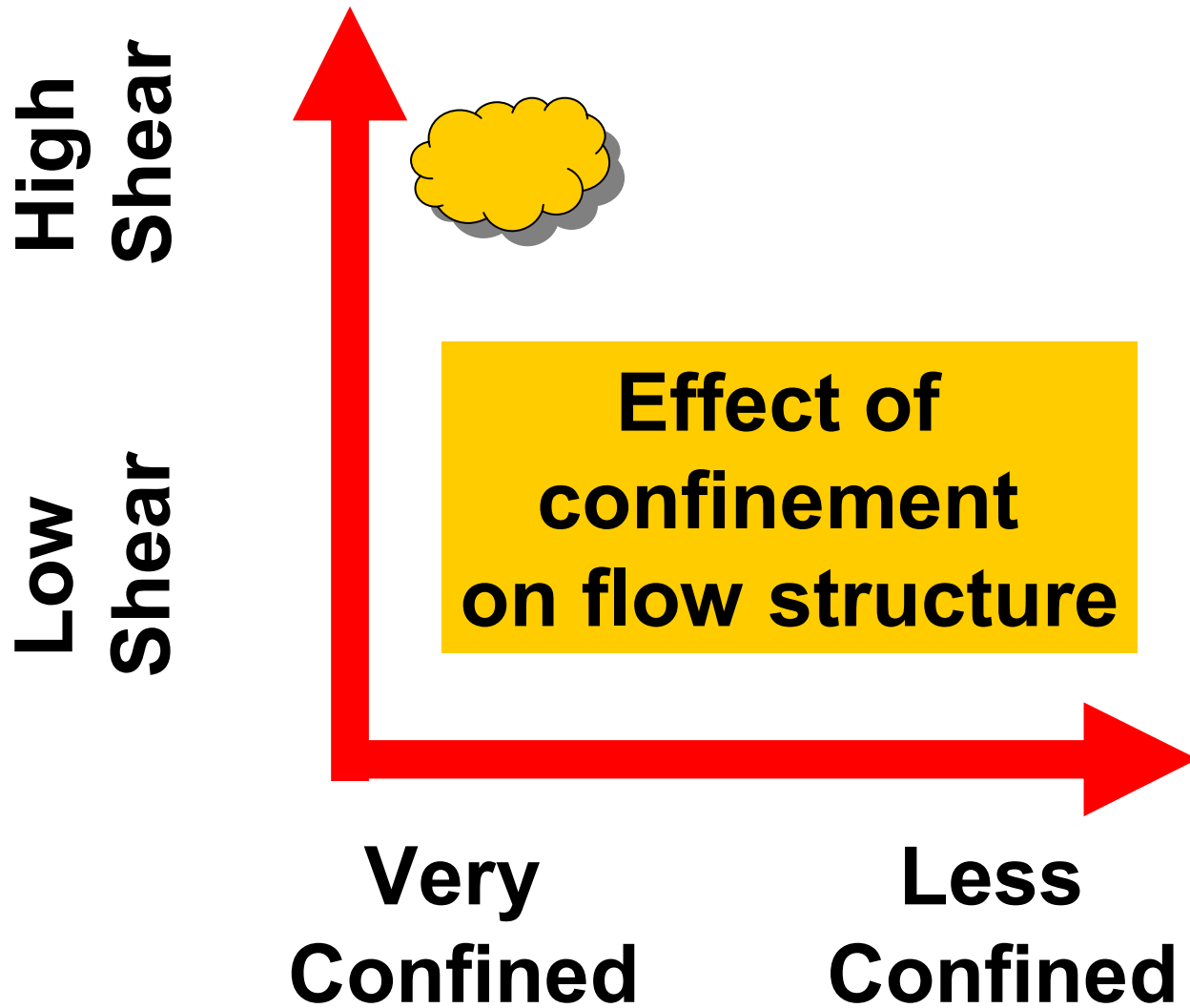
# Linear model fixed $\eta_s, \eta_f, \delta$



phase diagram (m=20)



# High Shear + Confinement



$\gamma > 0.3$   
 $f > 10 \text{ Hz}$   
Gap  $< 20 \mu\text{m}$

# Colloids Under Confinement

## Unconfined system:

Movie

5→4→3→2→1→0

0→1→2→3→4→5

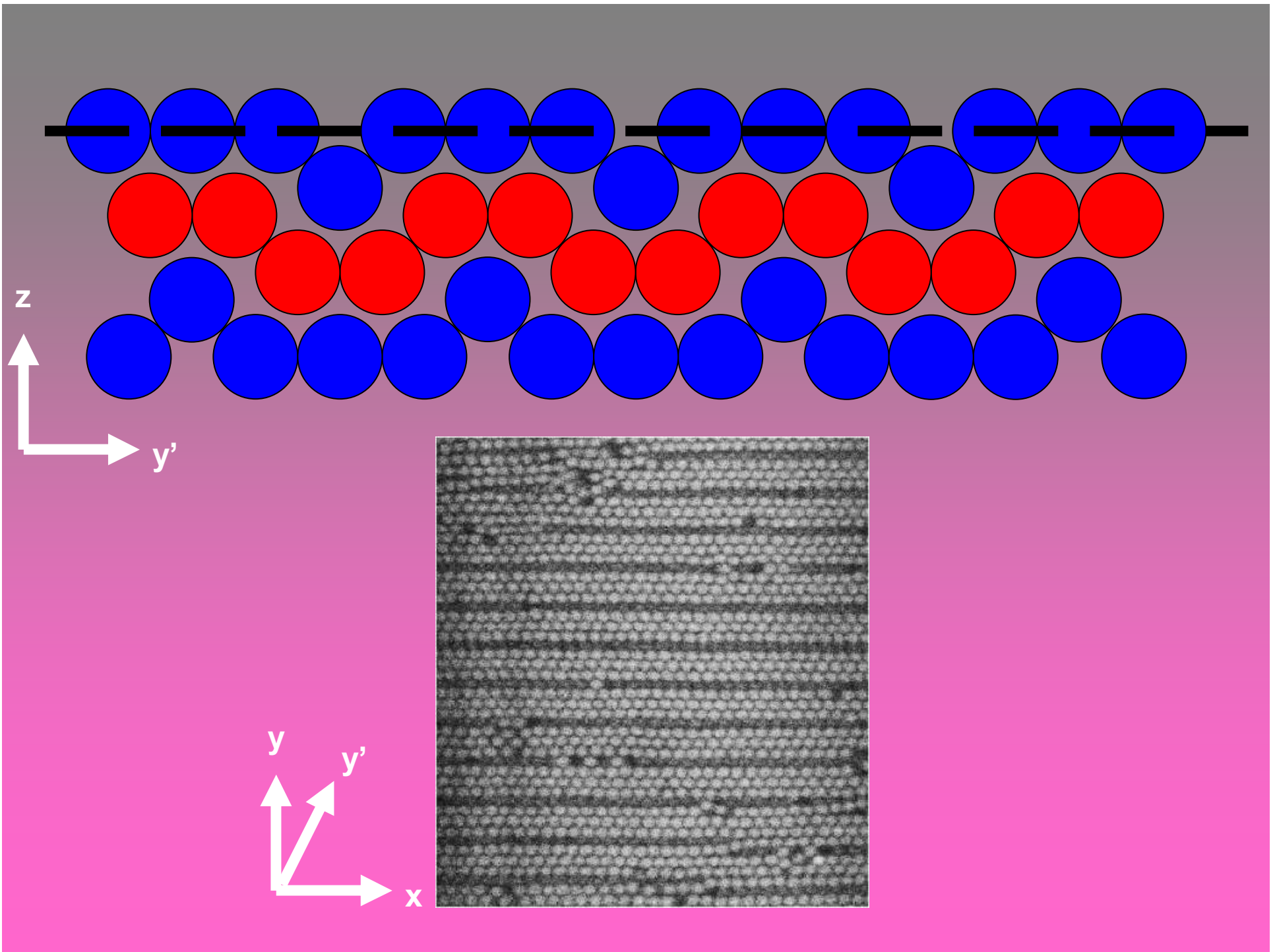
**Music by Vinay Prabhakar**  
**Movie by Cohen Studios**

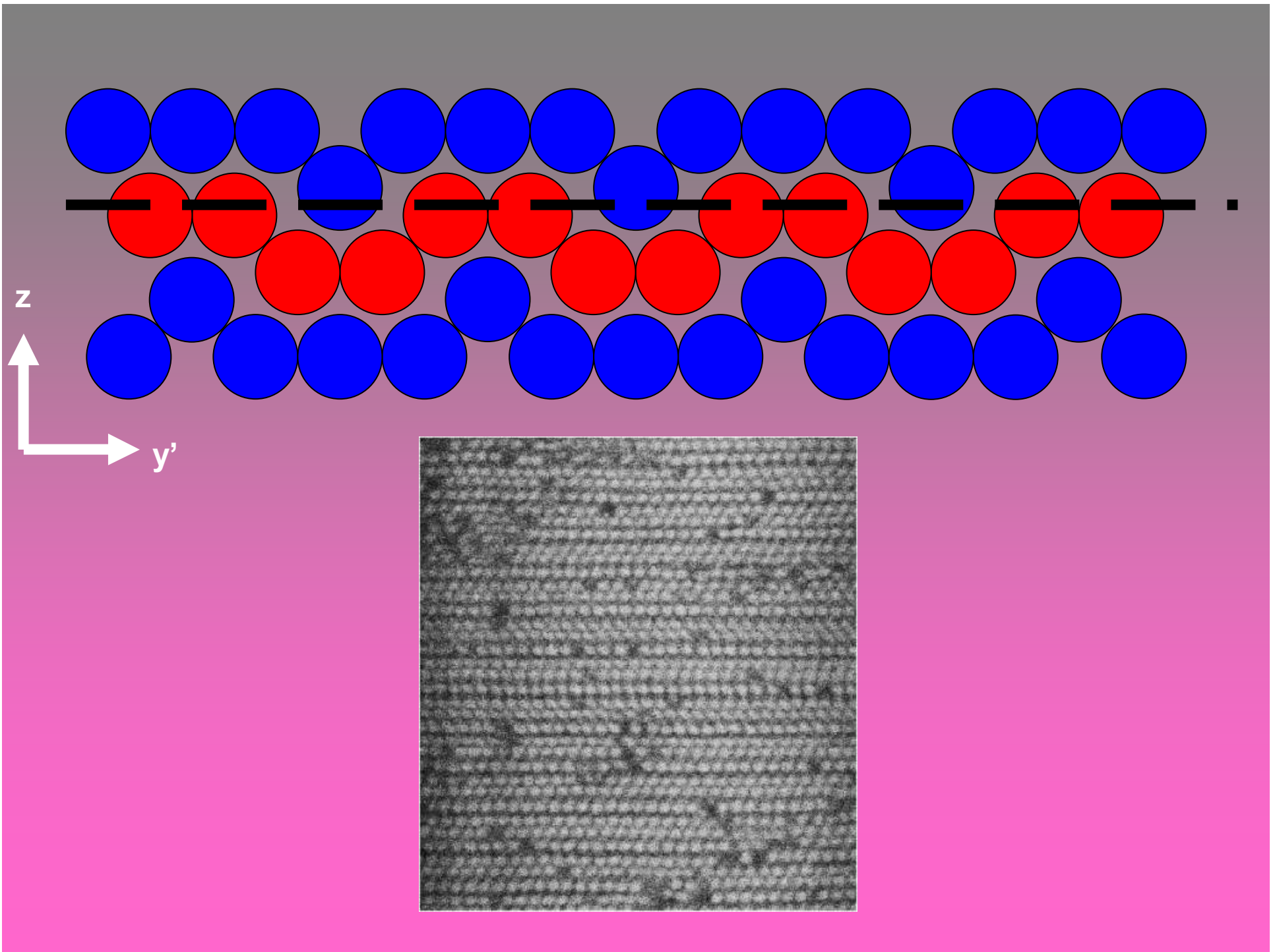
Next

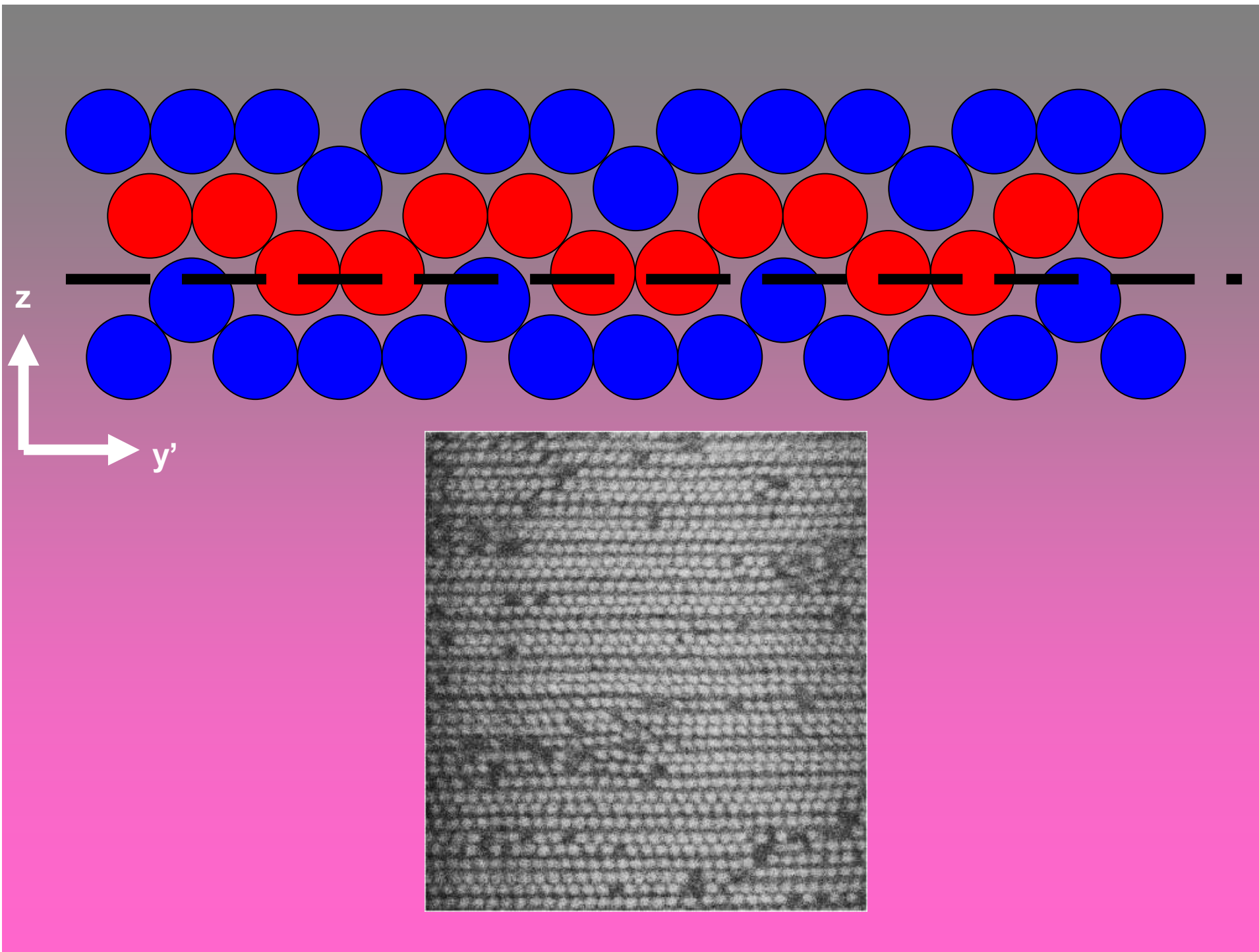
3→2→1→0

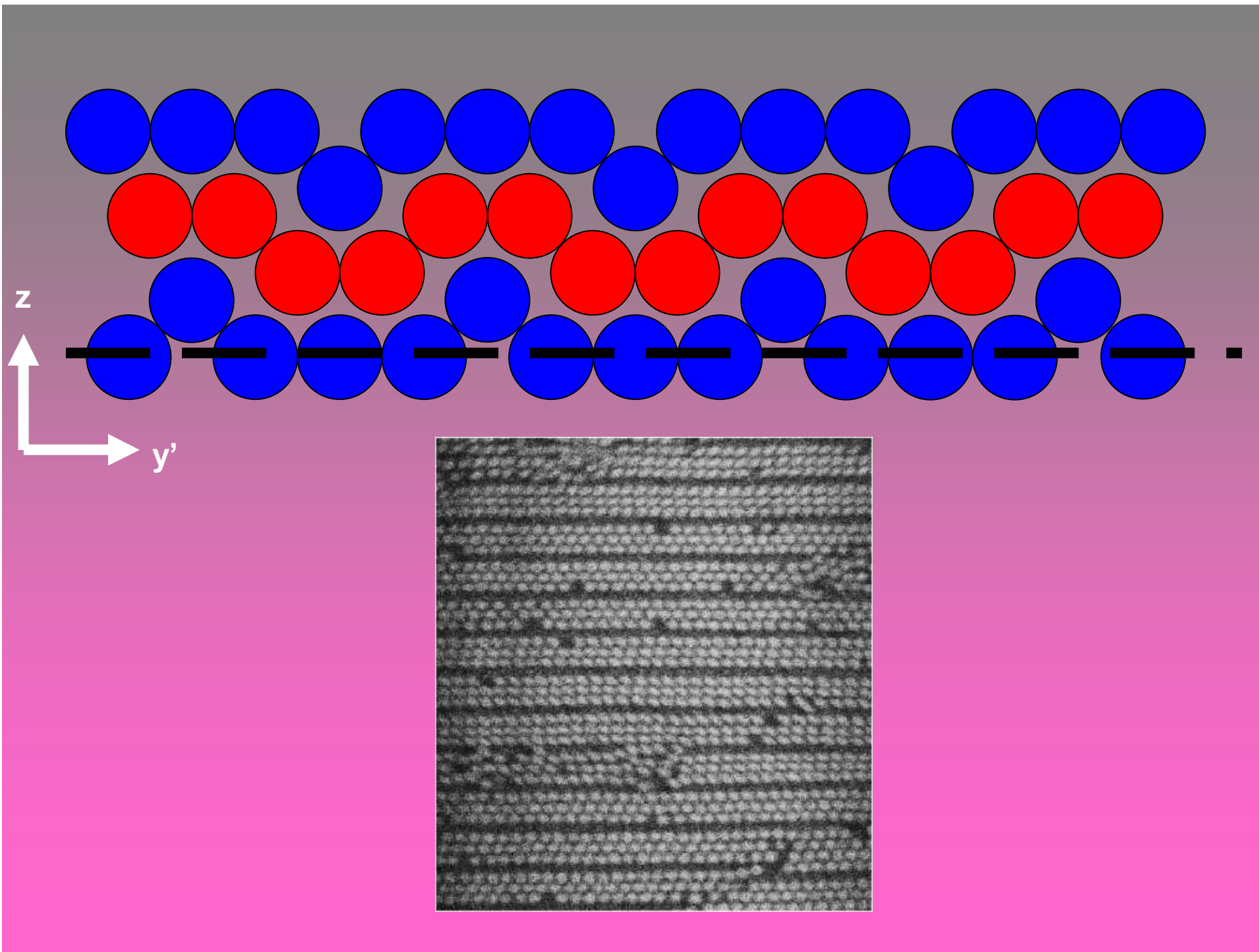
0→1→2→3

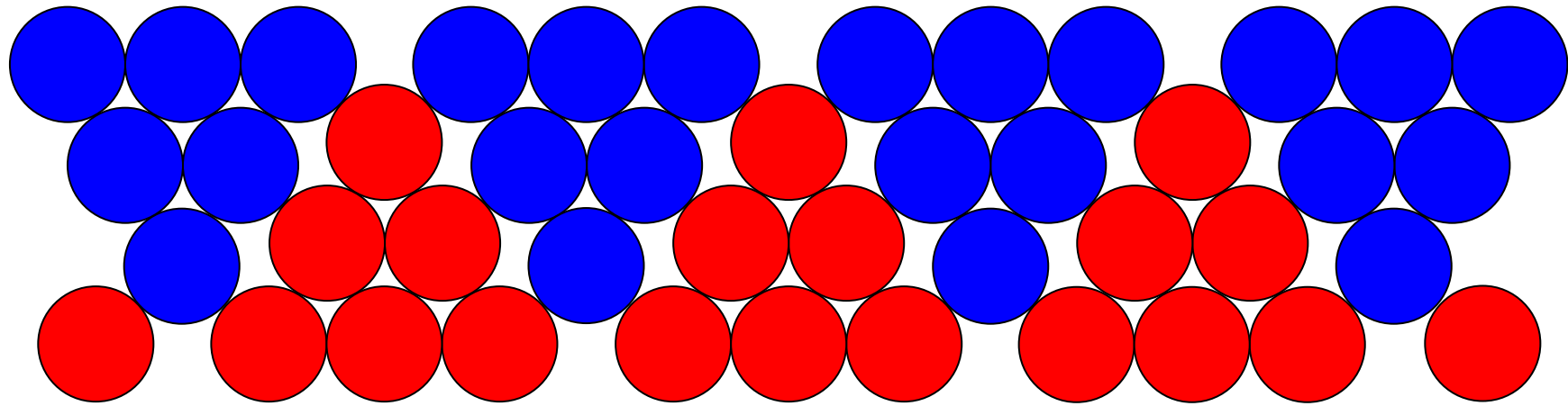










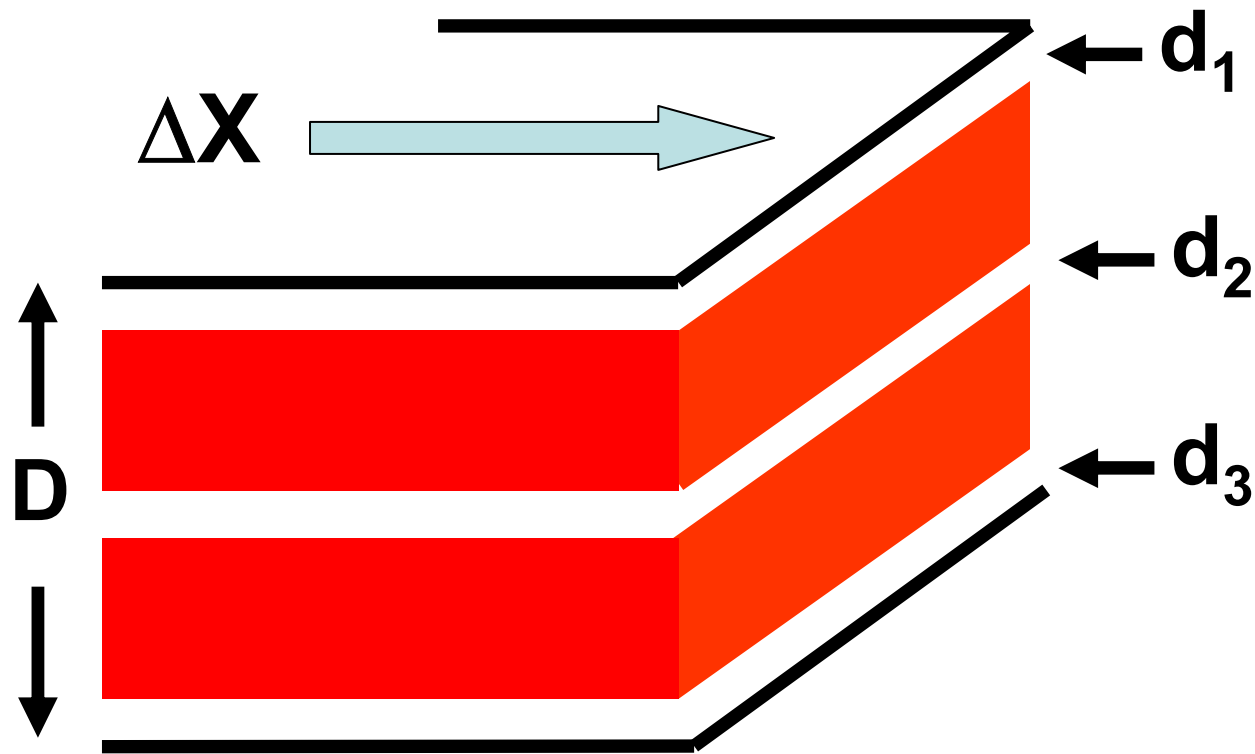


**Reservoir osmotic  
pressure important**

**Densest Incommensurate  
Packing in 2-D**

Piotr Pieranski & J. Finney, Acta Cryst **A35** 194-196 (1979)

# Osmotic Pressure ( $\Pi$ ) $\Leftrightarrow$ Shear Stress



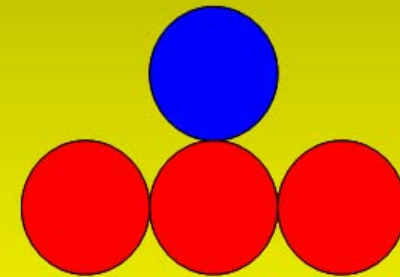
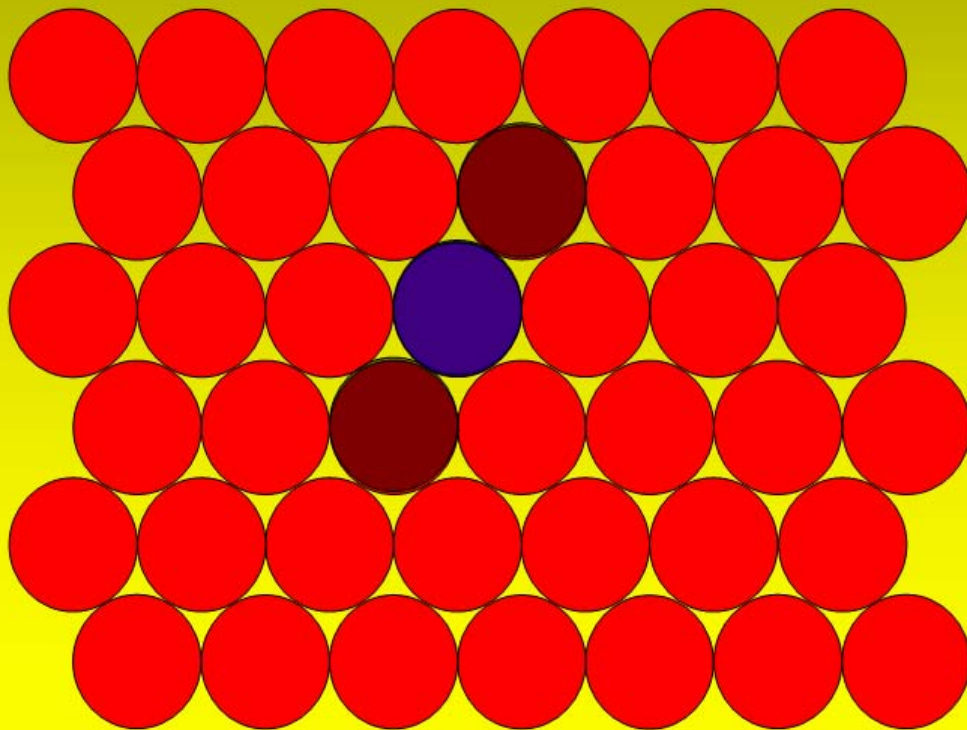
$$\eta_{\text{eff}} = \eta_0 * (D/(\sum d_i))$$



# Structure $\rightarrow$ Ambiguity

## Straight

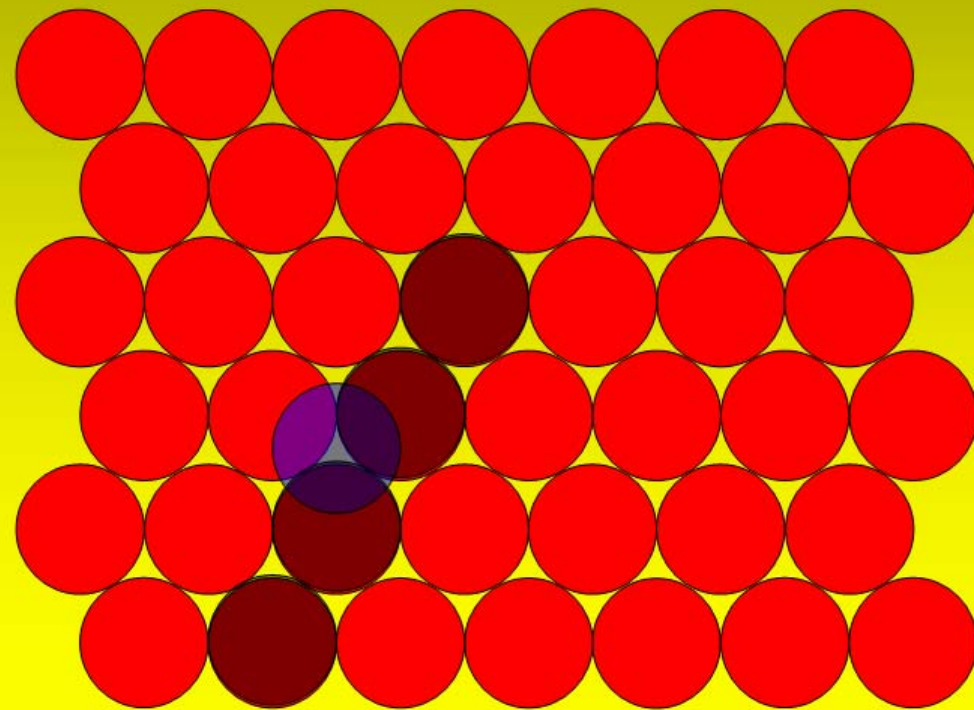
$$d_i = 0$$



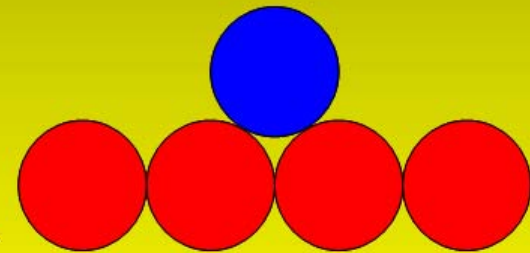
**Inter-layer  
Distance =  $2a$**

# Structure → Ambiguity

## Zigzag



$$d_i = 0$$



Inter-layer  
Distance =  $\sqrt{3} a$

# Estimate Shear Stress

$$\eta_{\text{eff}} \dot{\gamma} = \tau \text{ (stress)}$$

$\Pi(\phi) \rightarrow \text{Literature}$

$$\tau \sim 5 \Pi_{\text{HS}} \sim 6 \text{ dyn/cm}^2$$

# Building up a model

$$\Pi_{\tau} \cong \tau = \eta_{\text{eff}} \gamma f$$

$$(\cancel{\Pi_{\tau}} + \cancel{\Pi})_{\text{shear zone}} = (\cancel{\Pi_{\tau}} + \Pi)_{\text{reservoir}}$$

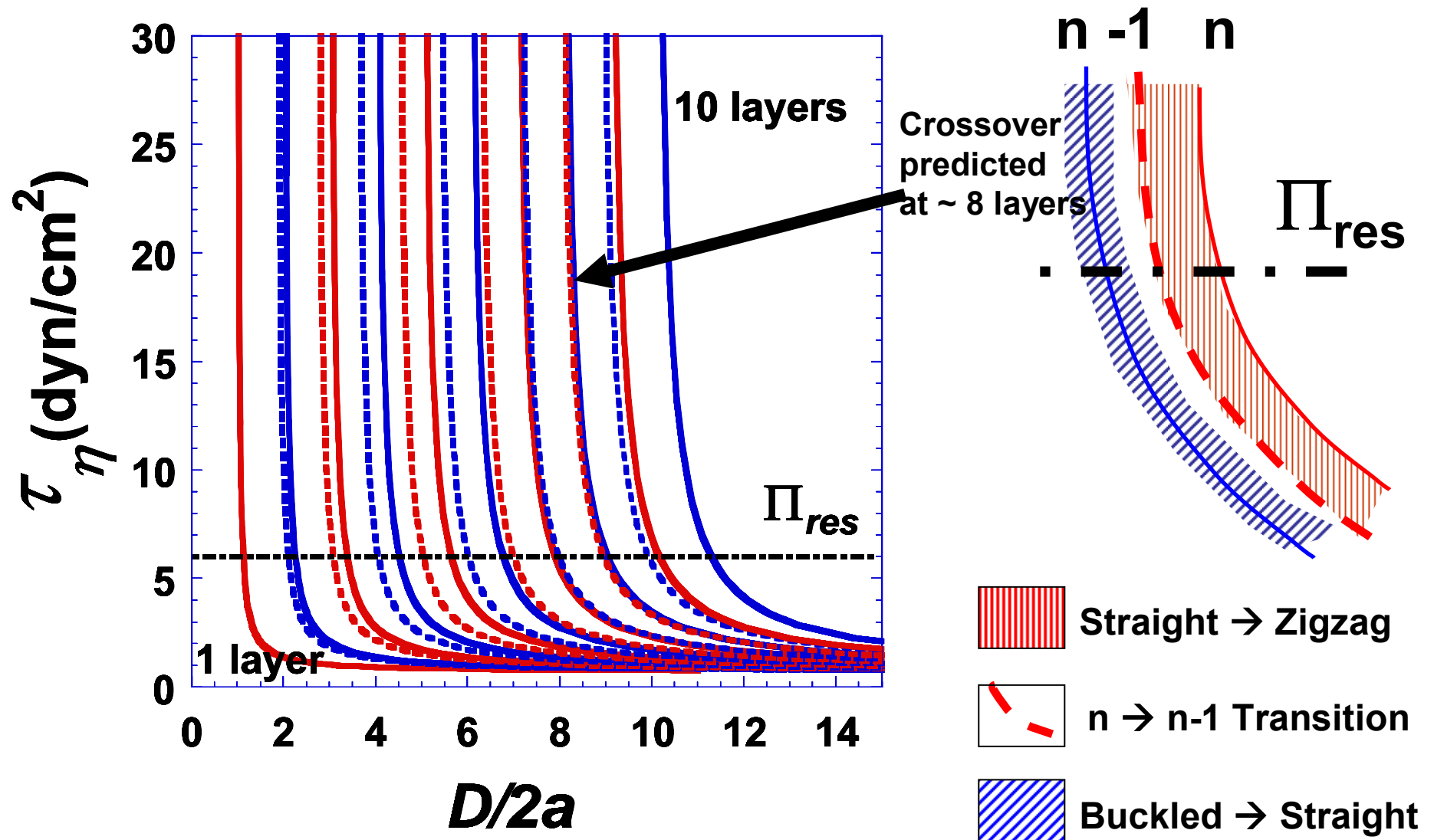
If gap  $\uparrow$   $\eta_{\text{eff}} \downarrow$  since  $d_i \uparrow$

Enforce  $\Pi_{\text{res}} = \Pi_{\tau}$ , &  $D \uparrow$ : **Sheets Buckle**

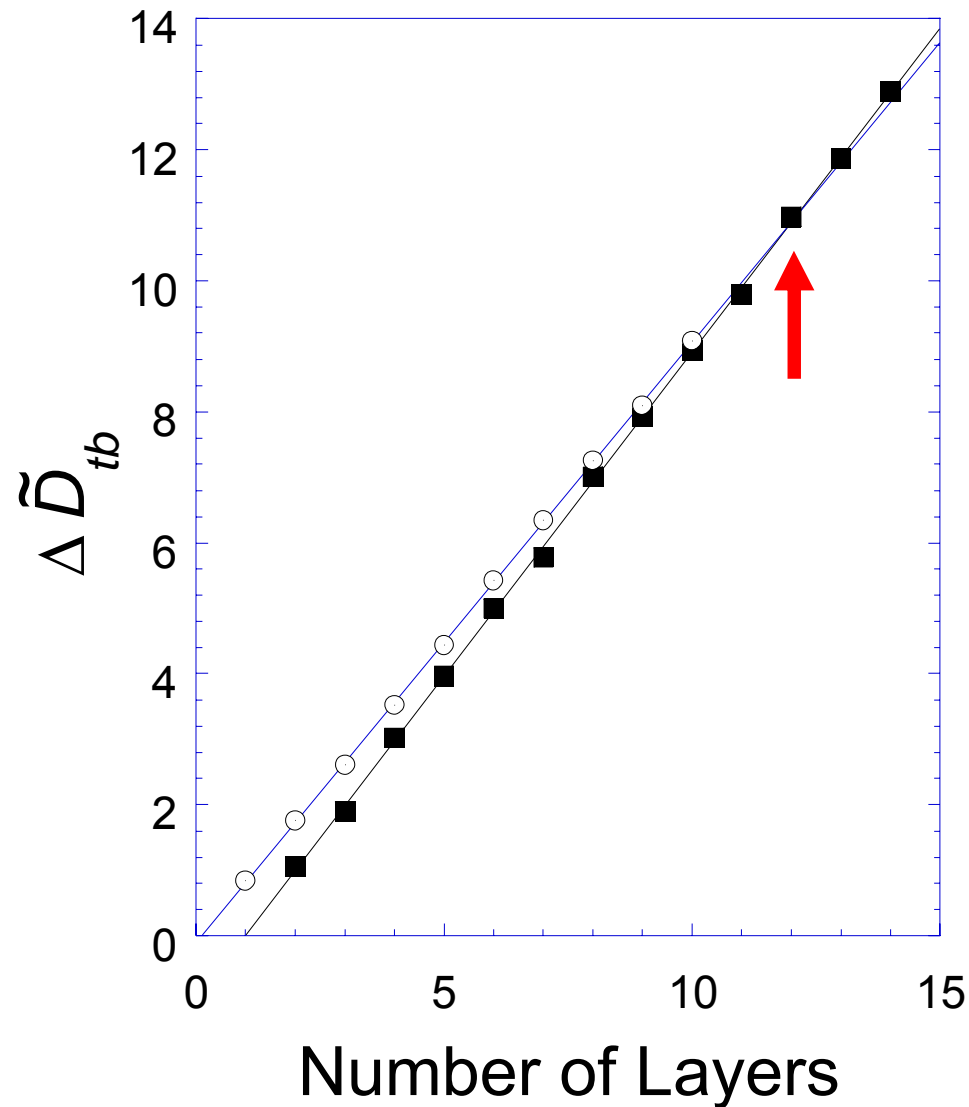
to keep  $d_i$  &  $\eta_{\text{eff}}$  constant

$\Pi_{\text{res}} = \Pi_{\tau}$  : given  $D, \gamma, f \rightarrow$  # of layers  
and transition mechanism

# Can predict # of layers



# Measurement of Crossover



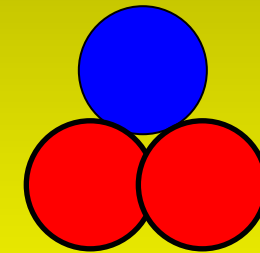
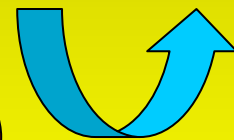
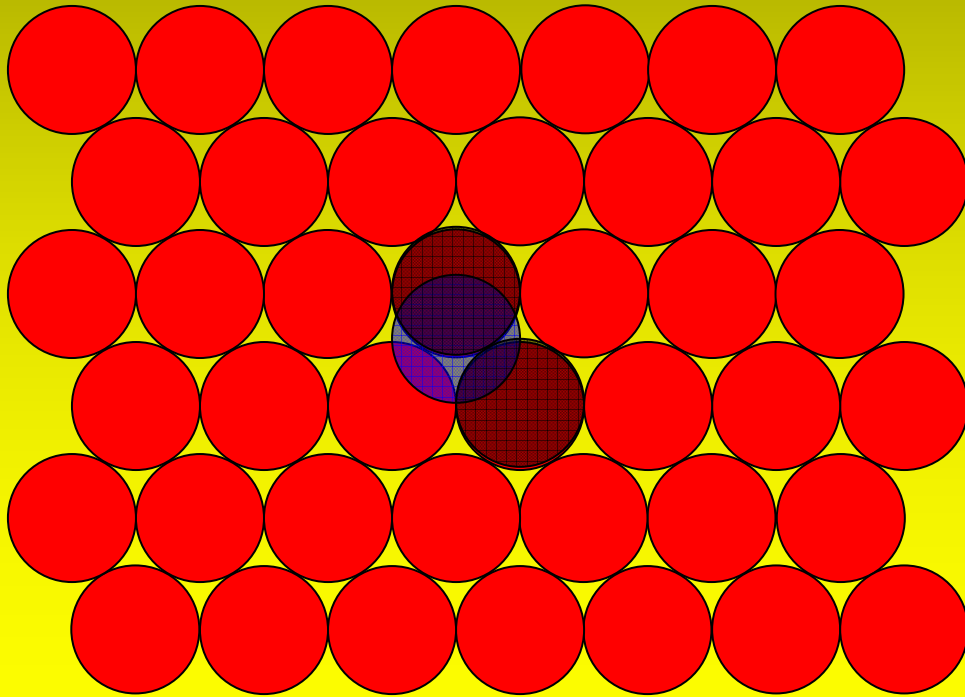
**Crossover occurs  
at ~ 12 layers**

**Why?**

# Straight But Registered

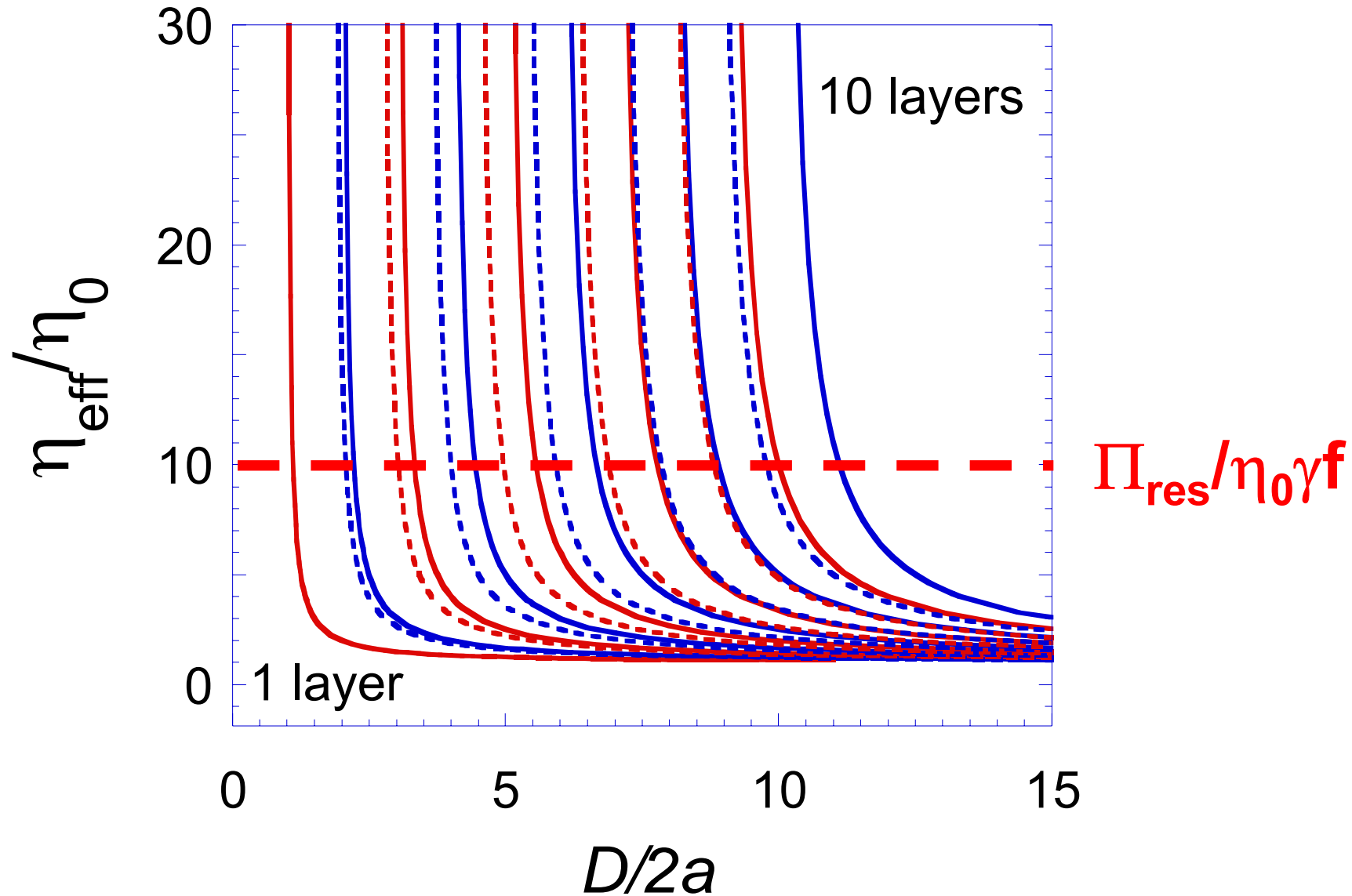
$$d_i = 0$$

Crossover occurs  
at  $\sim 13$  layers



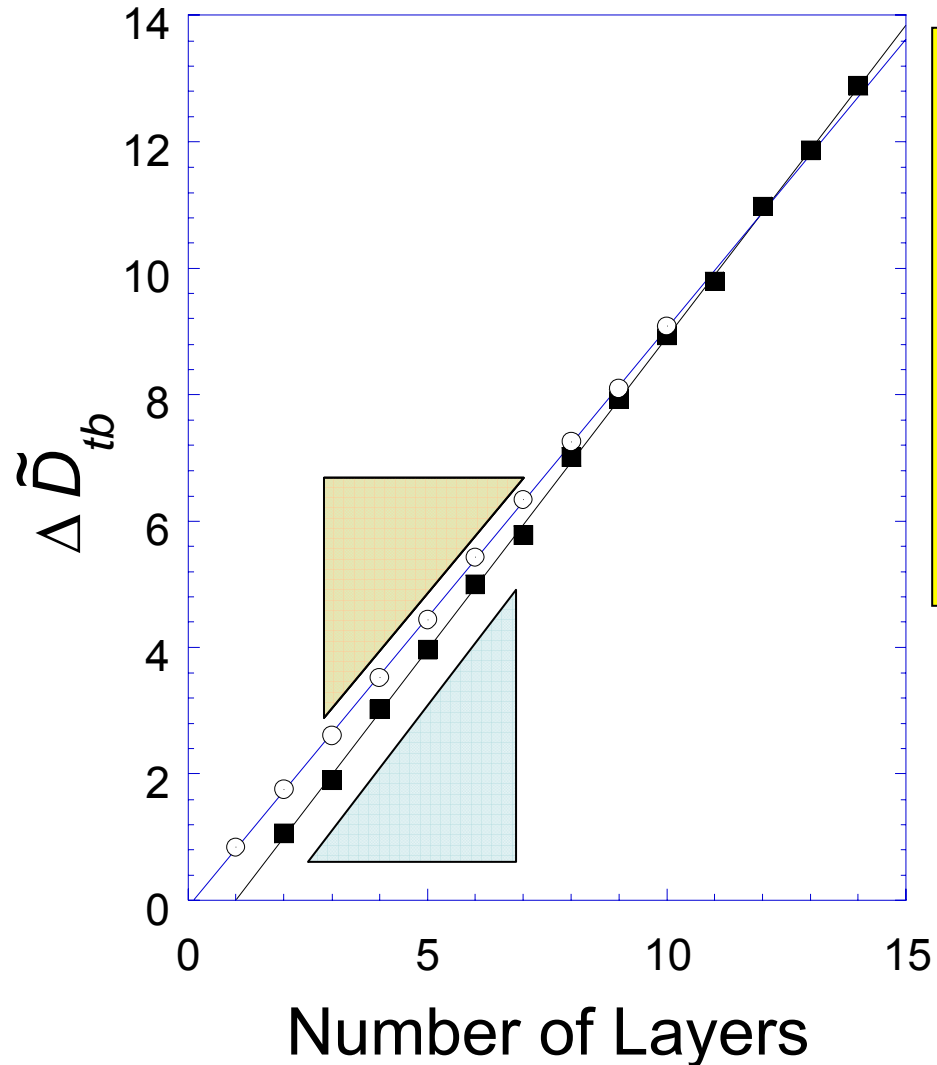
Inter-layer  
distance =  $1.88a$

# Predict Shear Rate Dependence





# Predict inter-layer distance



**Model:**  
 $\gamma f = 30 \text{s}^{-1}$   
**0.95**  
 $\gamma f = 0.30 \text{s}^{-1}$   
**0.87**

**Measured:**  
 $\gamma f = 30 \text{s}^{-1}$   
 **$0.92 \pm 0.05$**   
 $\gamma f = 0.30 \text{s}^{-1}$   
 **$0.89 \pm 0.05$**

$\gamma f$	Slope flat layers	Slope buckled layers
0.3	0.98	0.89
5.0	0.98	0.90
10.0	0.99	0.90
20.0	0.99	0.91
30.0	0.99	0.92

# Perspective

Three limits to colloid rheology:

1) Low Pe # in both shear zone & reservoirs

Osmotic pressures balance

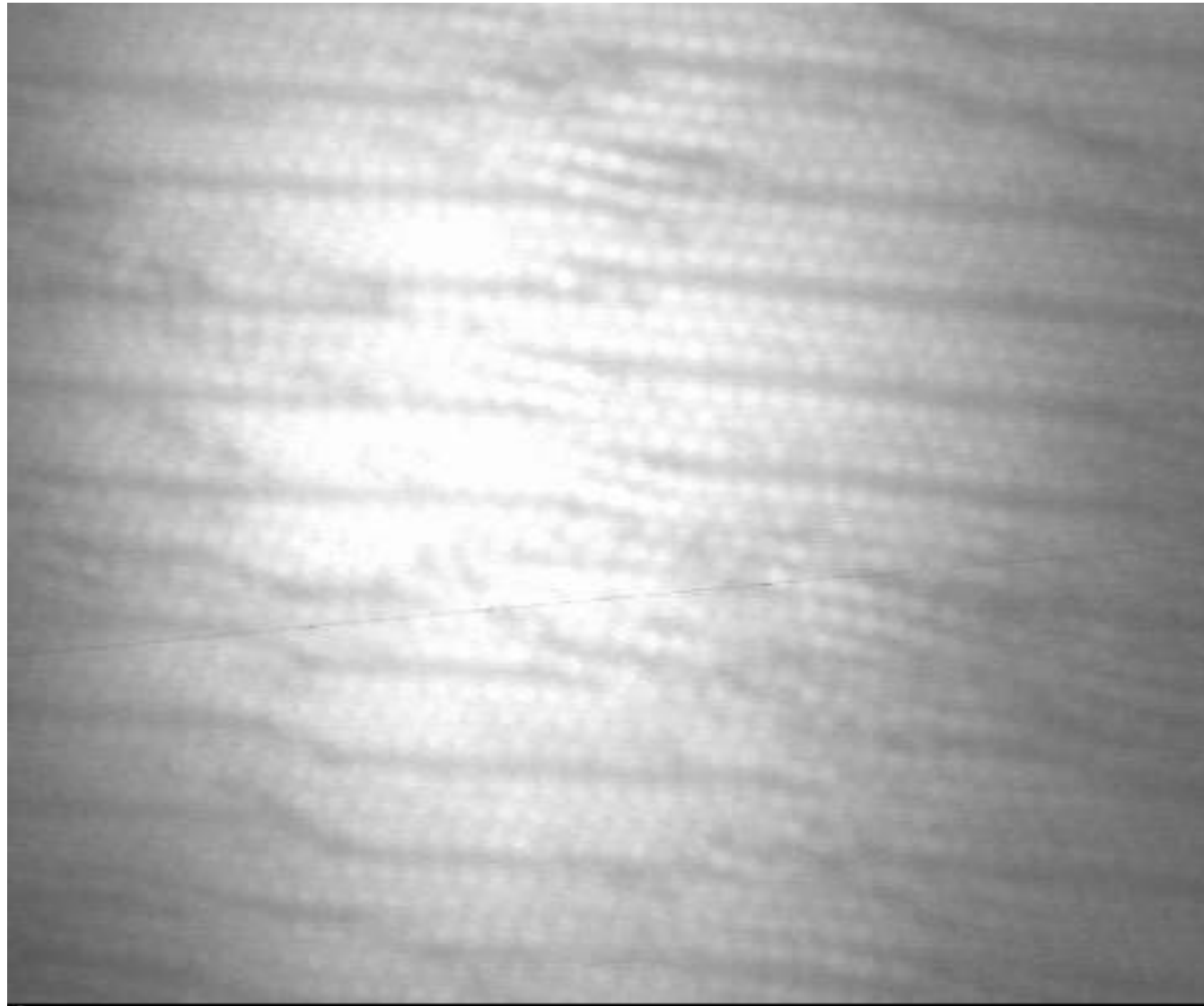
2) High Pe # in both shear zone & reservoir

$\Pi_\tau$  balances ??????

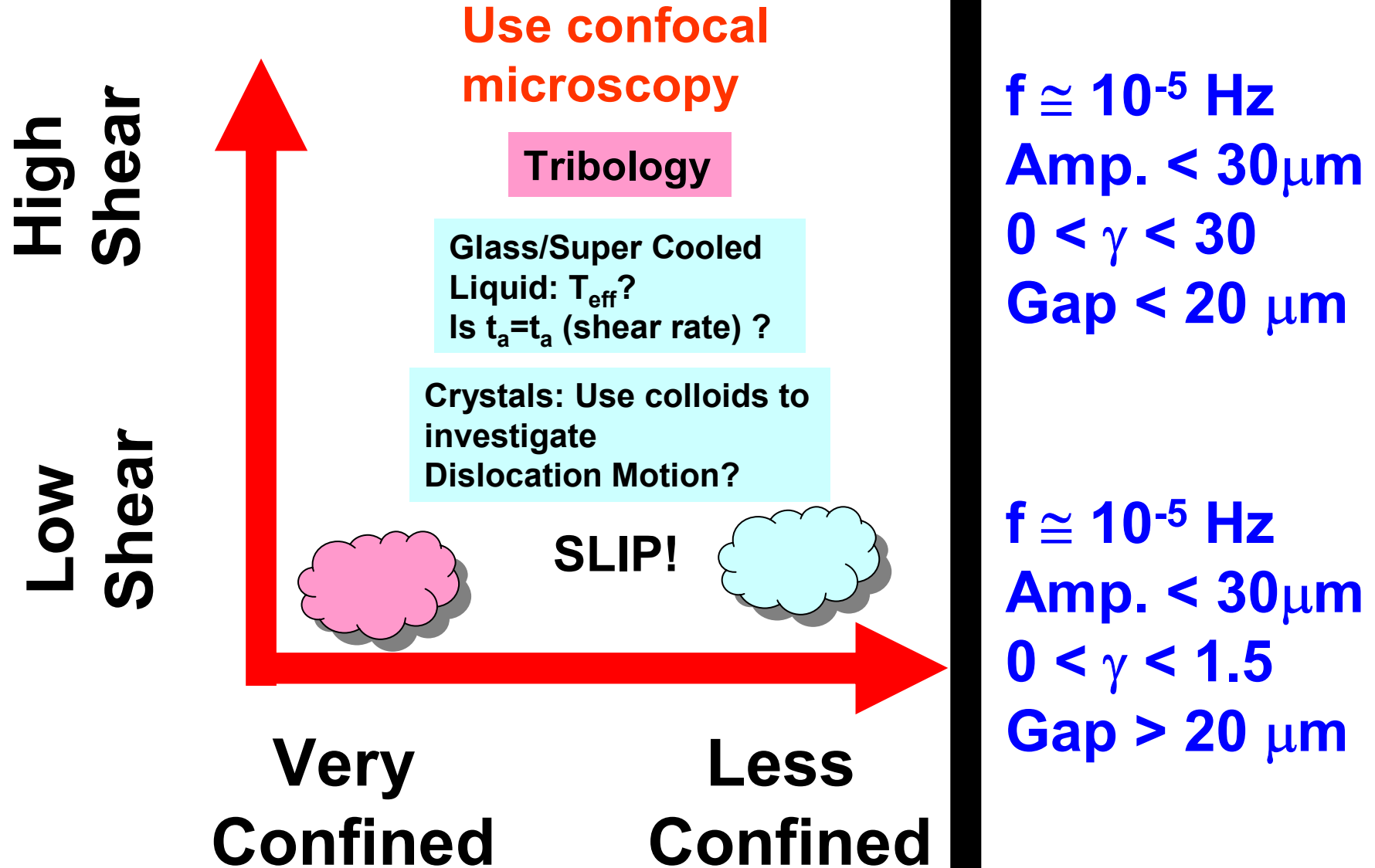
3) High Pe # in shear zone, low Pe # in reservoir

$\Pi_\tau$  balances  $\Pi_{res}$

# Grain Boundary Hydrodynamics

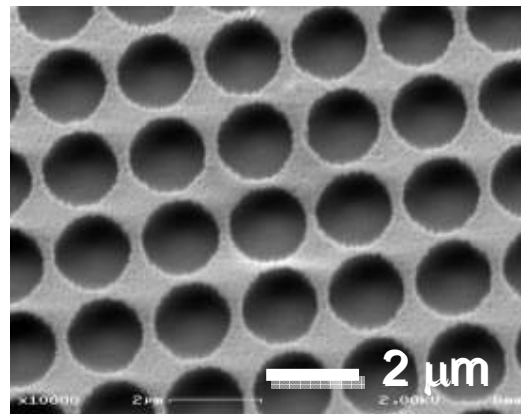
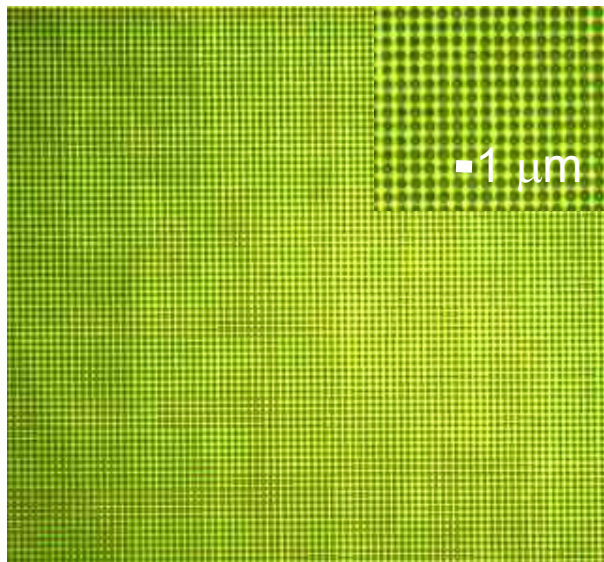


# Slow Shear

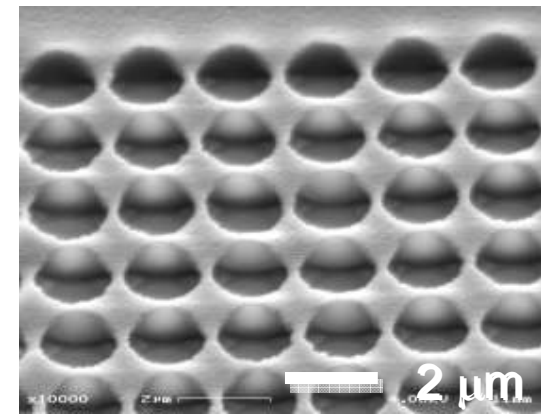


# Pattern Plates to Access Slow Shear

- Stamping (optical gratings)
- Photo Lithography → (5mm X 5mm region)



Top View



Isometric View

Tribology, Glasses, Dislocations

# Dislocations (Peter Schall)

- **Highly constrained systems**
  - **Nano: Smaller is harder/tougher**
  - **Cannot study dislocations in constrained atomic systems**
  - **Can we study them in colloids?**

- **Particle Shape: Spheres, diameter = 1.6  $\mu\text{m}$**

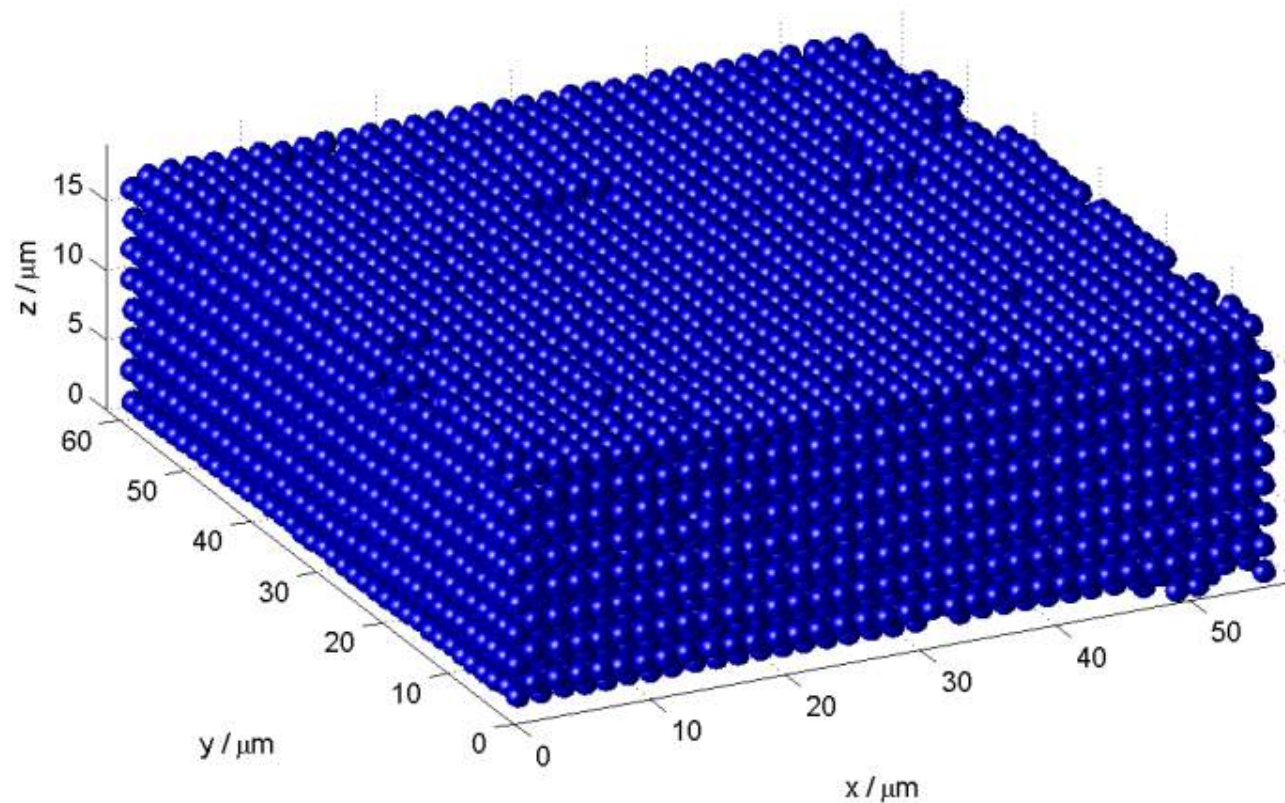
- **Particle interaction: Hard Spheres**

- **Volume fraction:  $\Phi \cong 70\%$**

- **Boundary conditions: Patterned surface**

# Grow Perfect FCC crystals

Grow crystals “epitaxially”  
by sedimentation



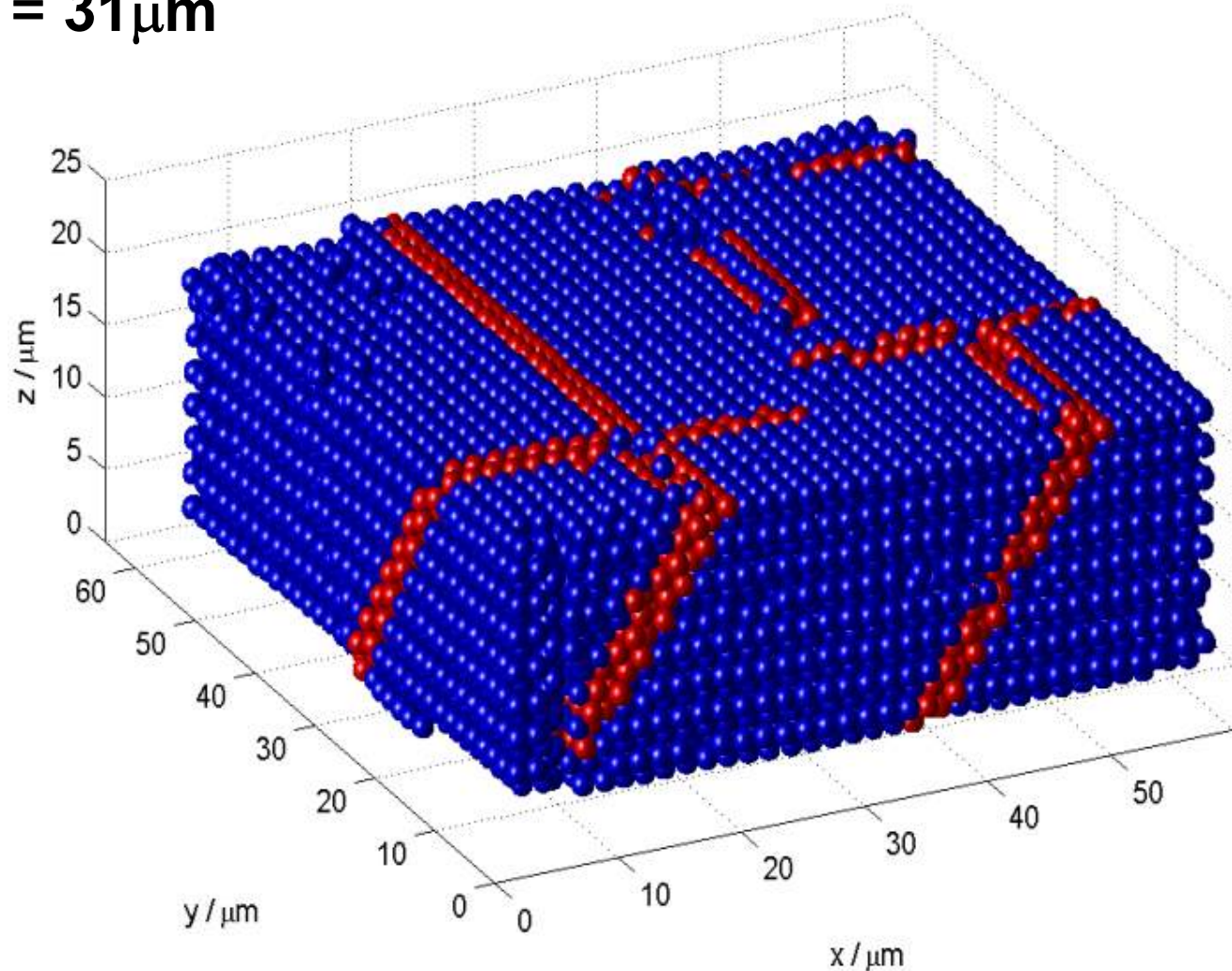
# Missmatched lattice → Misfit dislocations

**Grow crystal on mismatched lattice with  
 $\varepsilon_0=0.015$   
When crystal height,  $h < 22\mu\text{m}$   
Crystal is defect free**



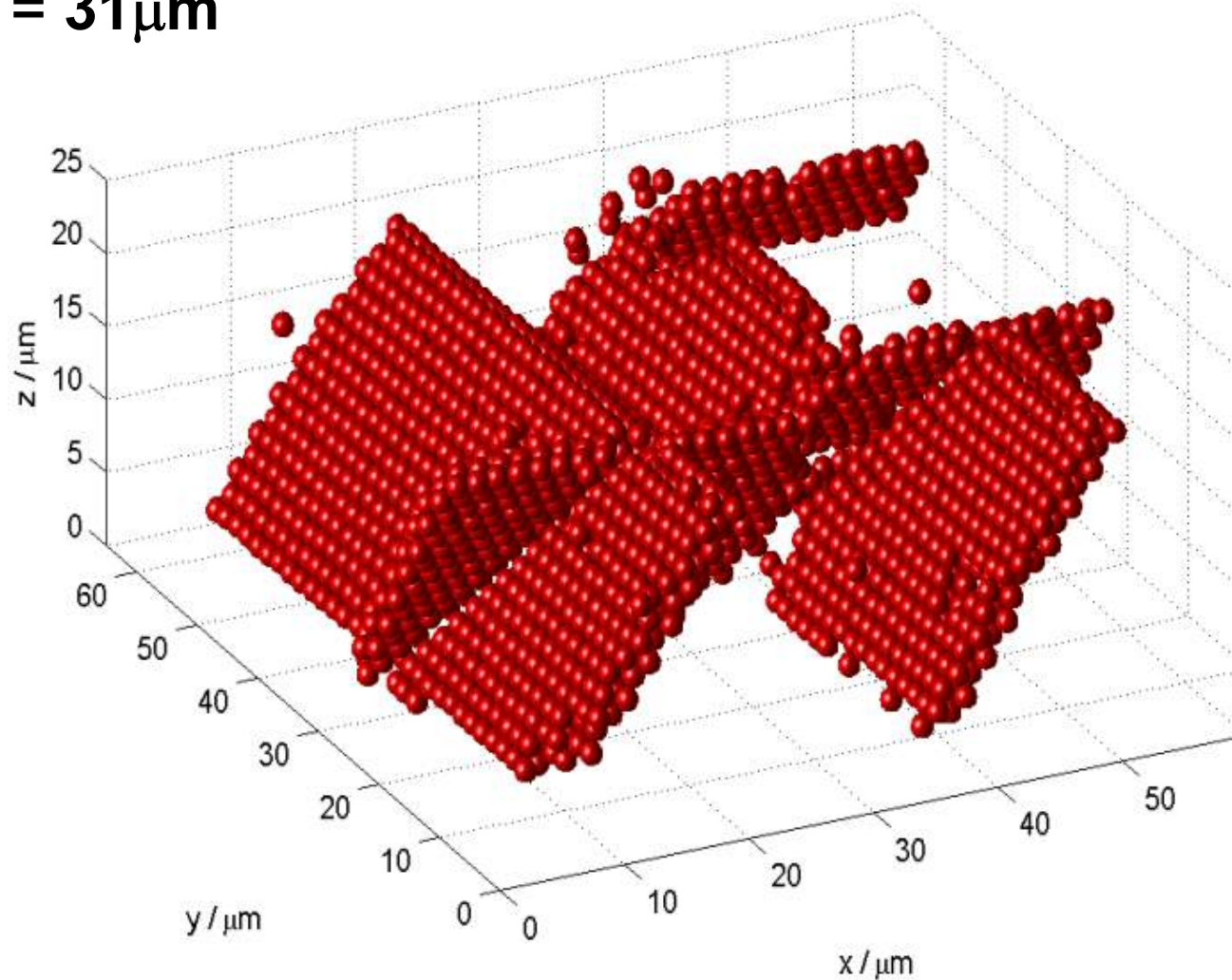
# Missmatched lattice → Misfit dislocations

When  $h = 31\mu\text{m}$

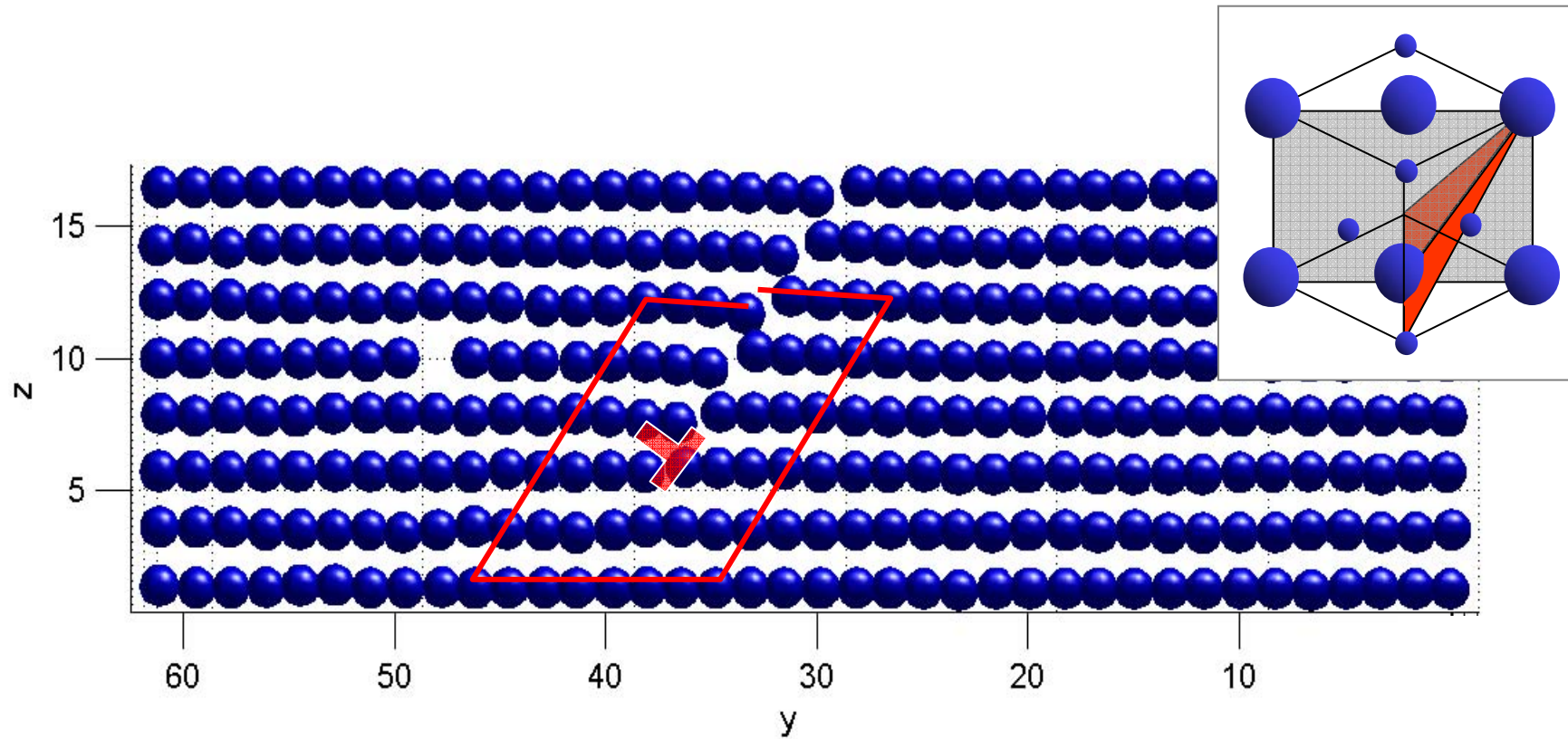


# Isolate stacking faults

When  $h = 31\mu\text{m}$



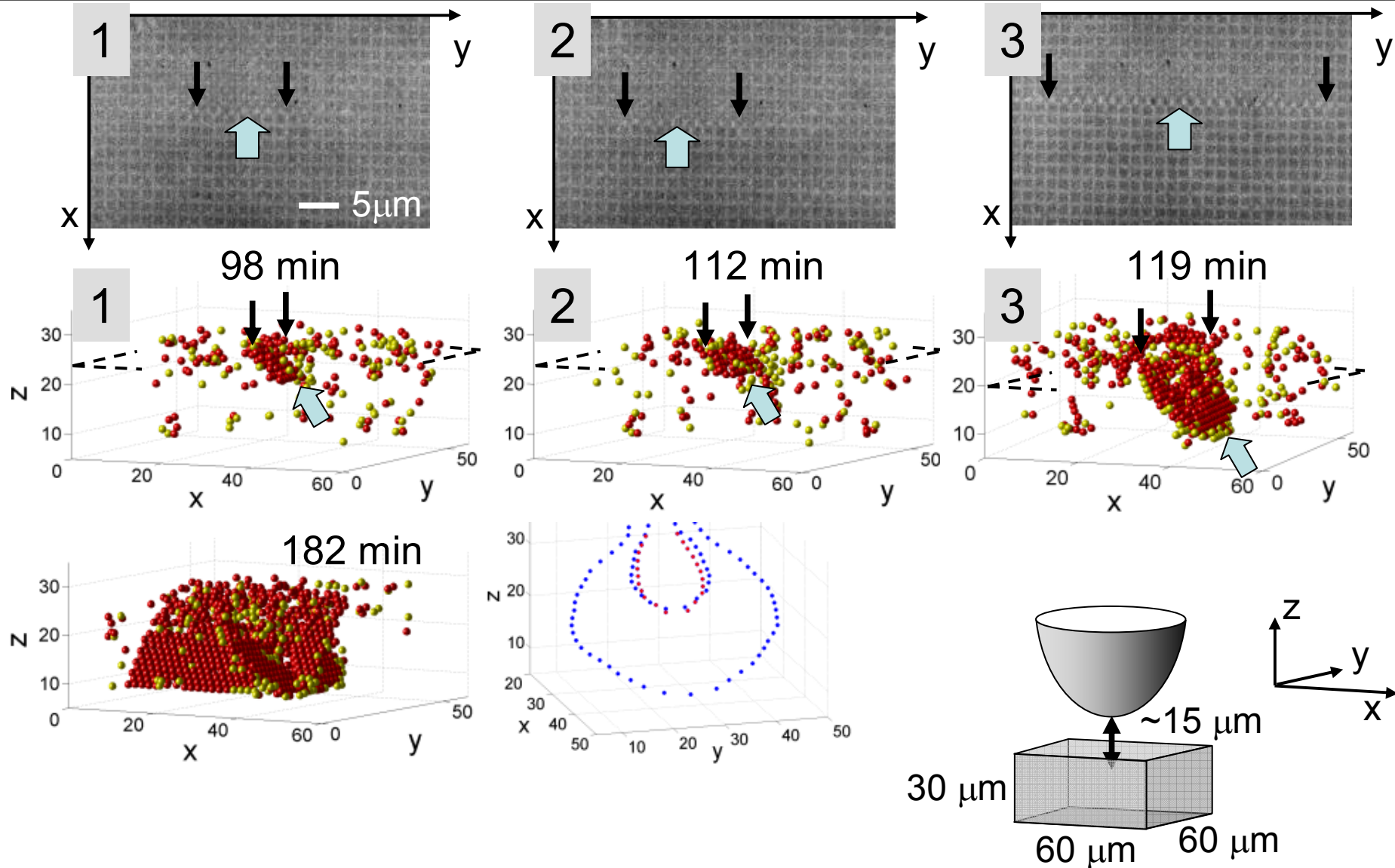
# Image a cut through the crystal



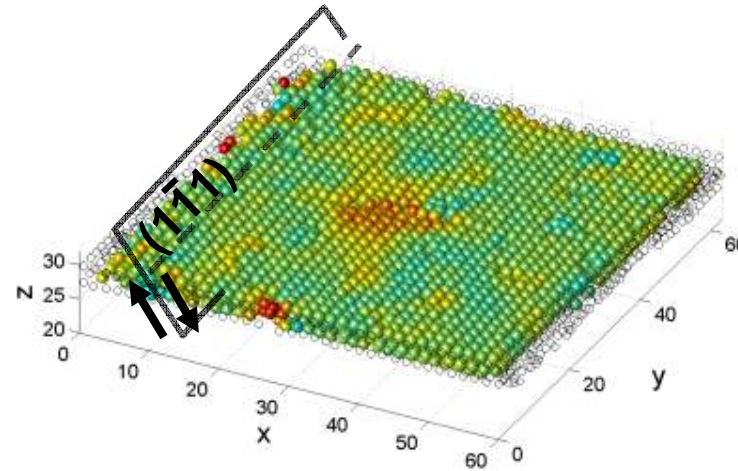
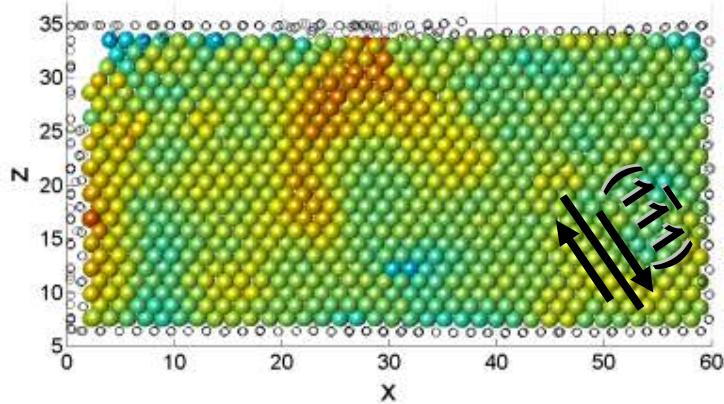
- Burger's vector:  $b = 0.94\mu\text{m}$  oriented  $\sim 54^\circ$  to  $y$  axis
- Shokley partial dislocation (usually paired in metals)



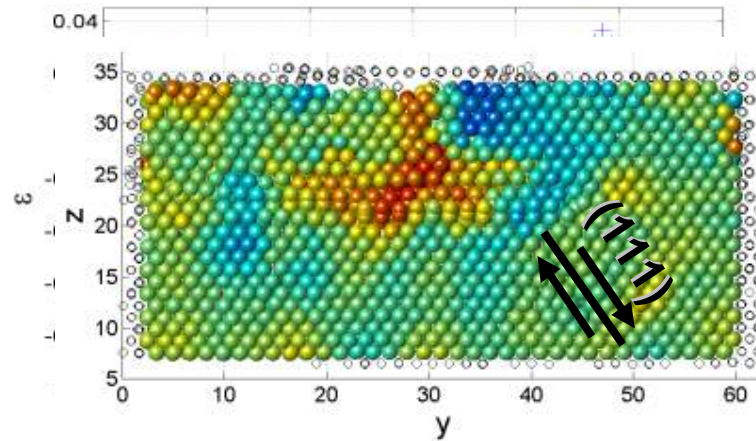
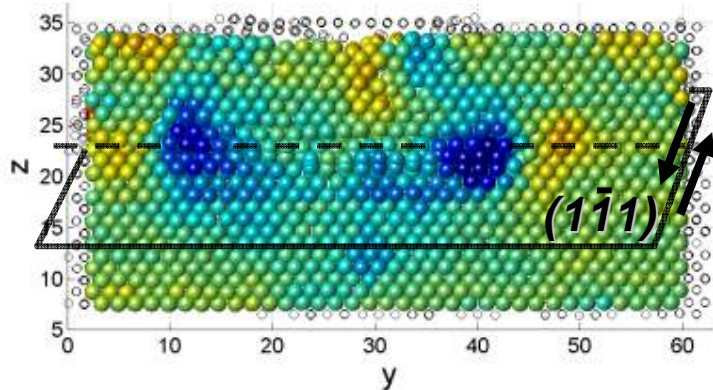
# Nanoindentation on micron scale



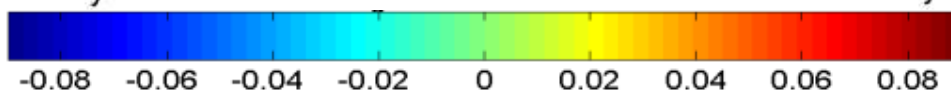
# Visualizing Strain



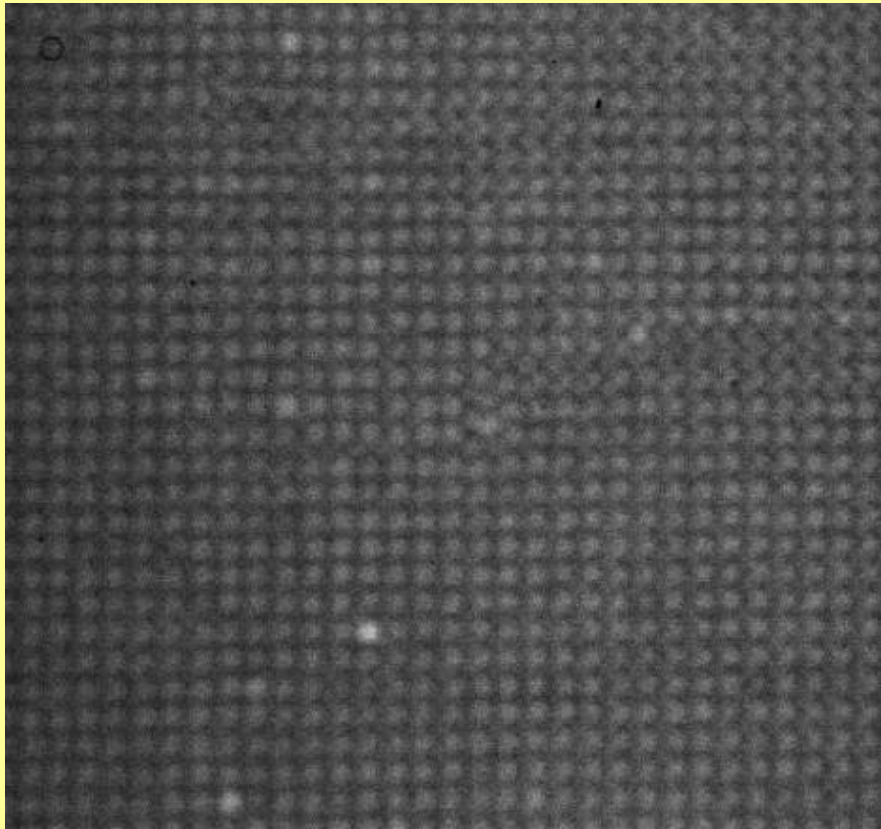
$$\begin{aligned} \varepsilon'_{ij} \\ x' (1,-1,-2) \\ z' (1,-1,1) \\ \gamma \equiv 2\varepsilon'_{x'z'} \end{aligned}$$



$\gamma'$

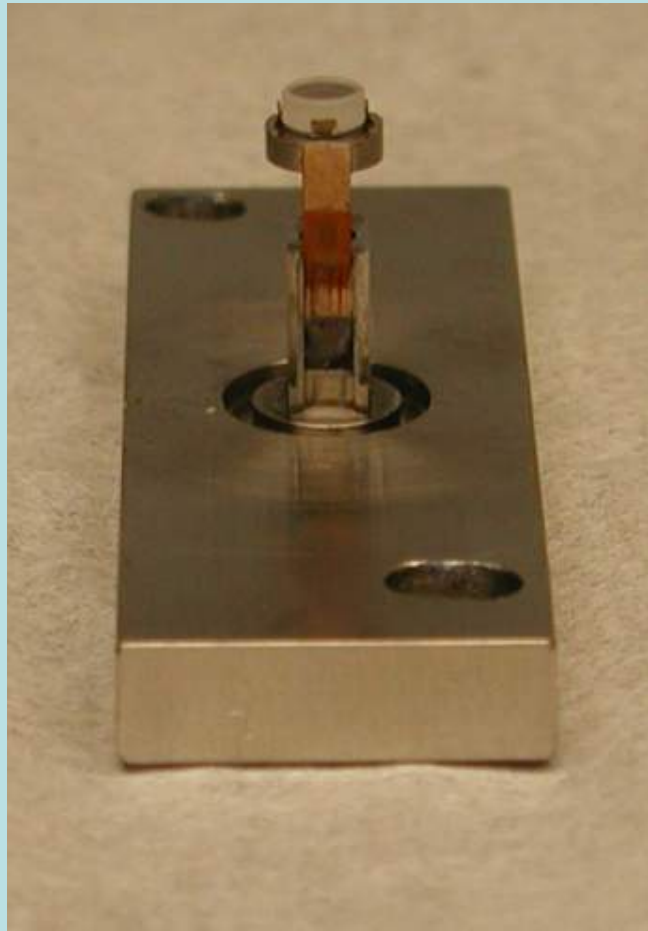


# Shearing Colloidal Crystal Films



Sharon Gerbode

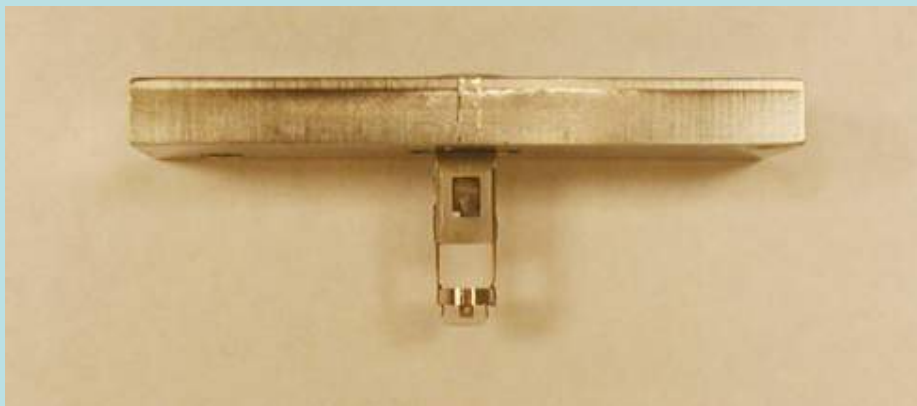




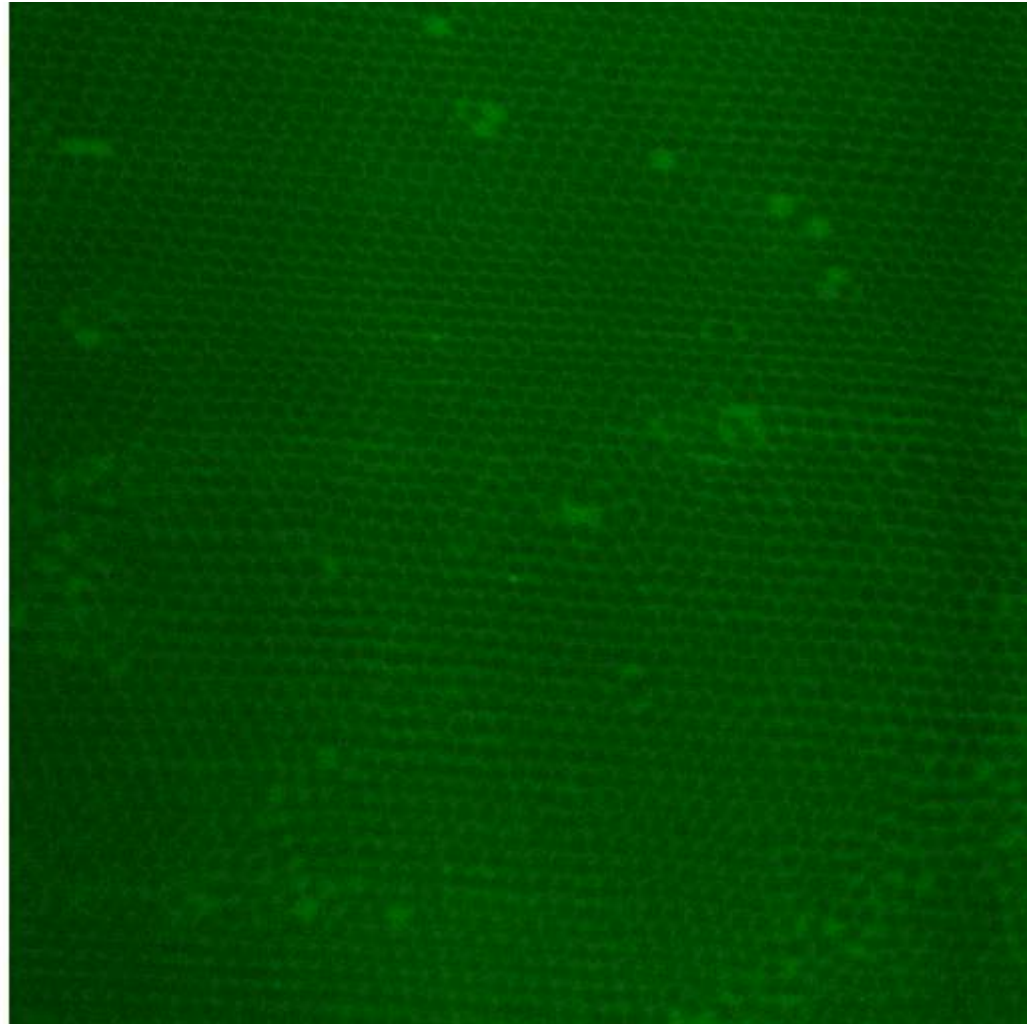
# Shear Thickening Flows



Mark Buckley



# Intermittent Flows





# The Future



- Particle shape (discs, dimers, tetramers,...)
- Particle interaction (depletion forces, E&M)
- Volume fraction (different phases)
- Boundary conditions (plate roughness)
- Shear rate / confinement parameter space

**Measure transmitted stress**

–(Jacob Israelachvili)

# Thanks:

David Weitz (Harvard)  
Thomas Mason (UCLA)  
Jacob Israelachvili (UCSB)  
Frans Spaepen (Harvard)  
Michael Brenner (Harvard)  
Peter Schall (Amsterdam)

Principal investigators

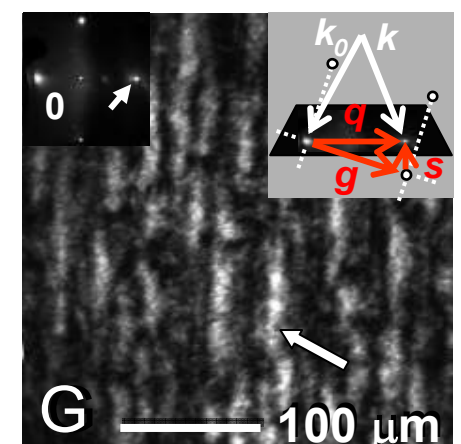
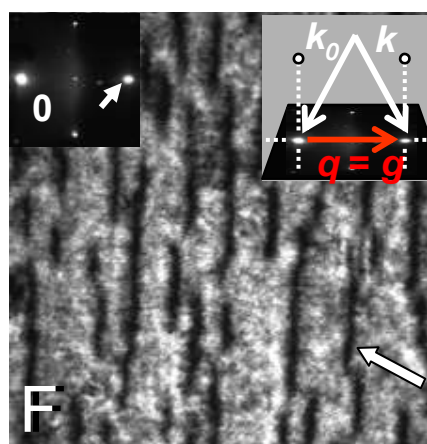
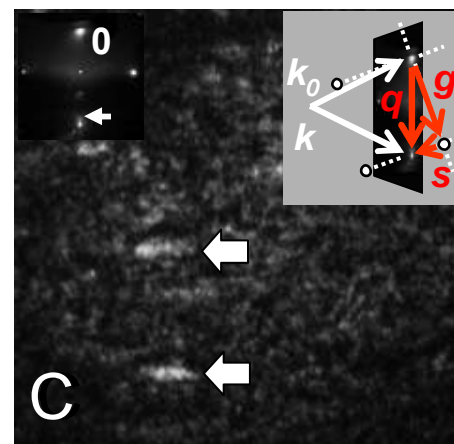
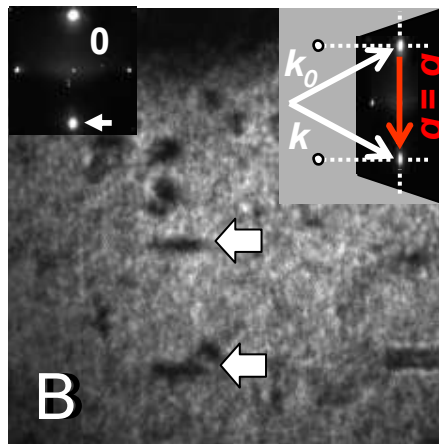
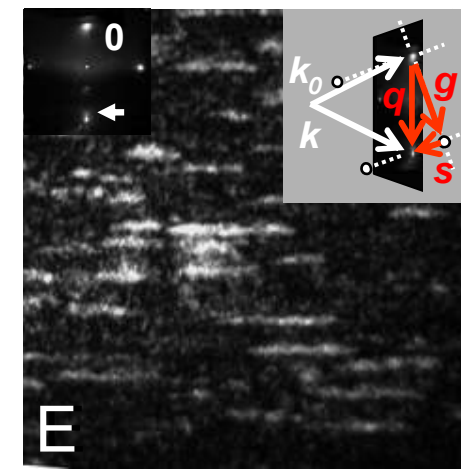
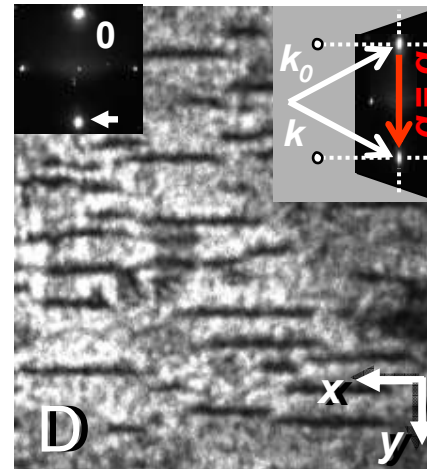
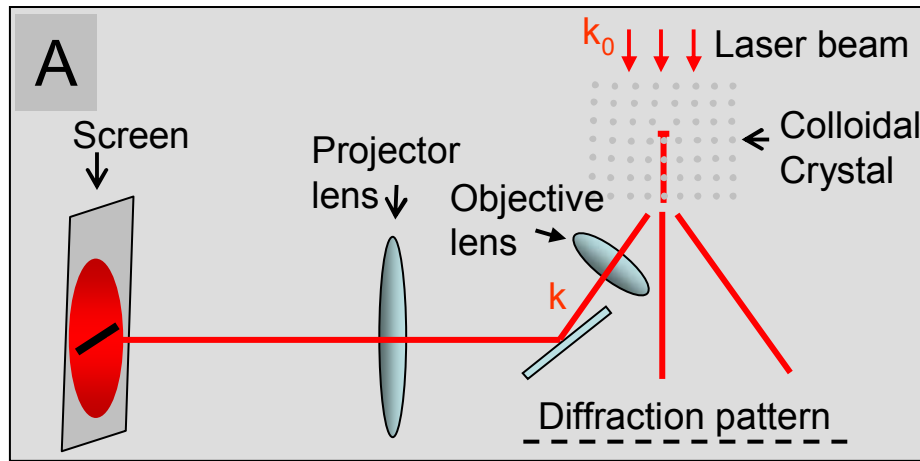
Benny Davidovich (Harvard)  
Dan Blair (Harvard)

Post. Docs

Mark Buckley (Cornell)  
Sharon Gerbode (Cornell)

Graduate students

# Laser Diffraction Microscopy (LDM)

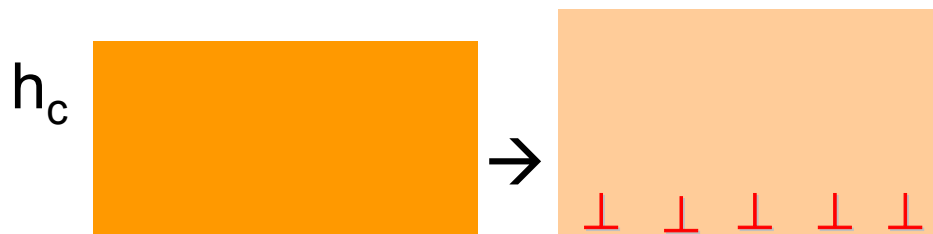


Nearly ideal lattice

Detection of lattice defects

# Test continuum theories

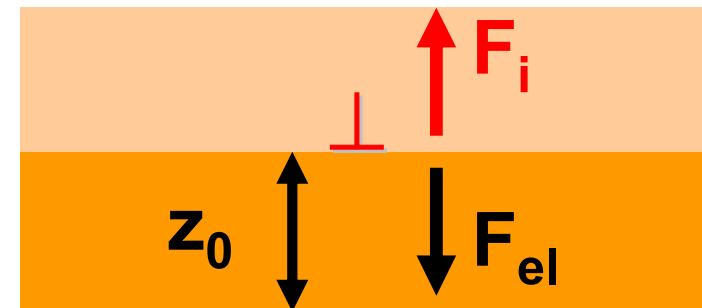
Prediction for  $h_c$   
1-d dislocation models:



$$U_{el} \propto h$$

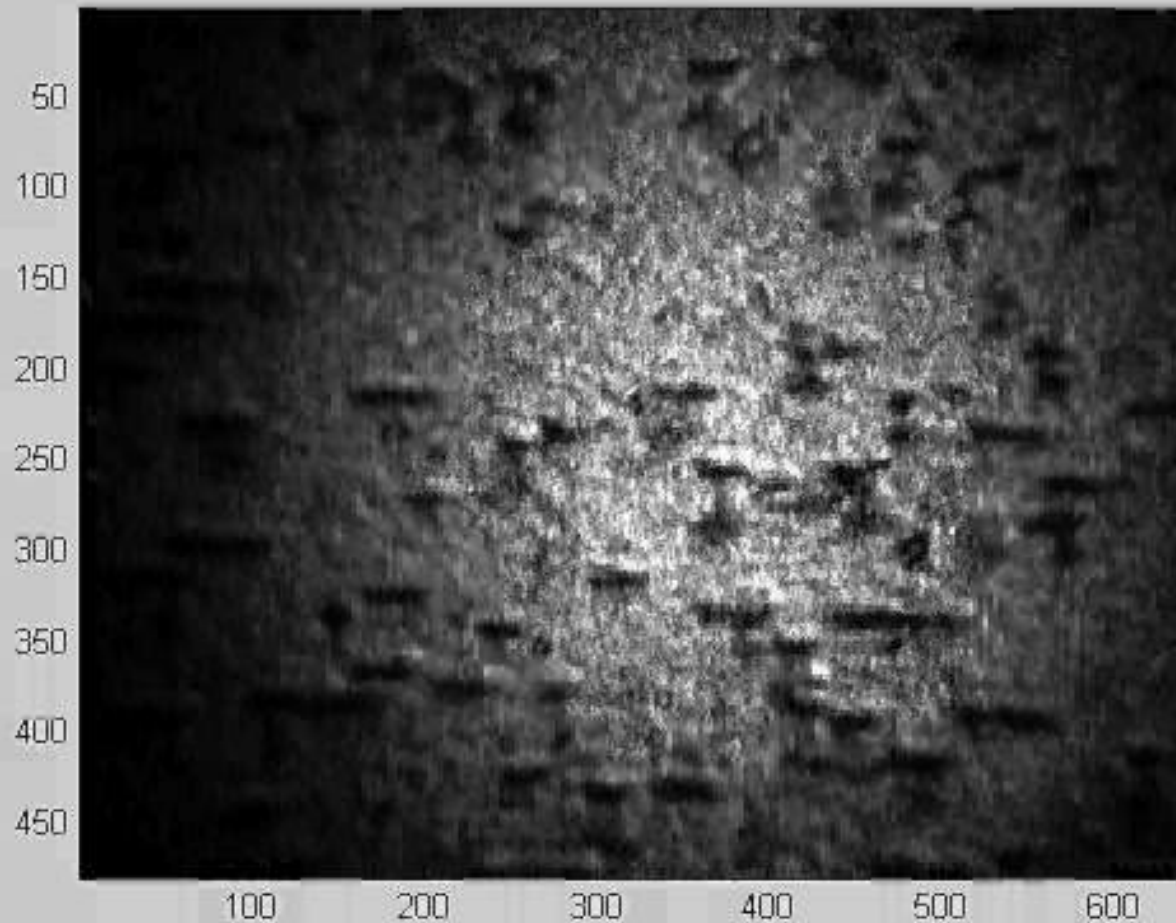
- Minimize  $U_{el} + U_l$  with respect to  $\rho$
- $h_c = b \ln(R/r_c) / 8\pi\epsilon_0(1-\nu^2)\cos\alpha$
- $h_c = 22\mu\text{m}$  (consistent)

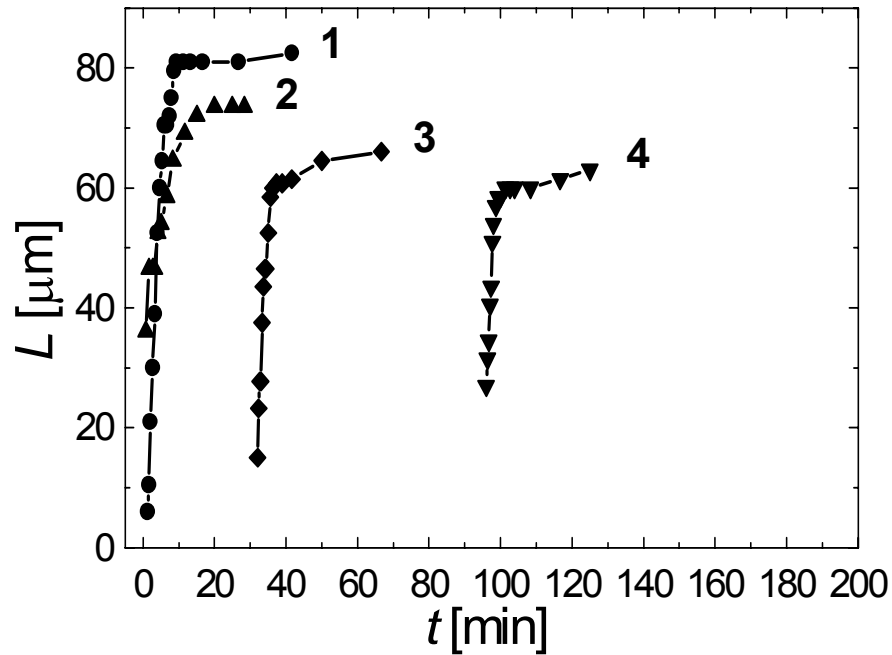
Prediction for dislocation  
resting height  $z_0$



- Balance  $F_{el}$  and  $F_i$
- $z_0 = (b\cos\alpha)^2\rho / 4\pi(1-\nu^2)\epsilon_0^2$
- $z_0 = 2.1\mu\text{m}$  (observe  $3\mu\text{m}$ )

# LDM movie of growing dislocation





## From Continuum Theory:

$F_{pk}$  = force due to elastic strain

$F_l$  = force due to line tension

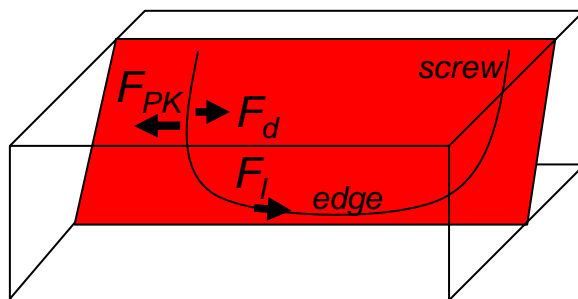
$F_d$  = drag force on dislocation

As dislocation length grows elastic strain,  $\epsilon$ , is relieved.

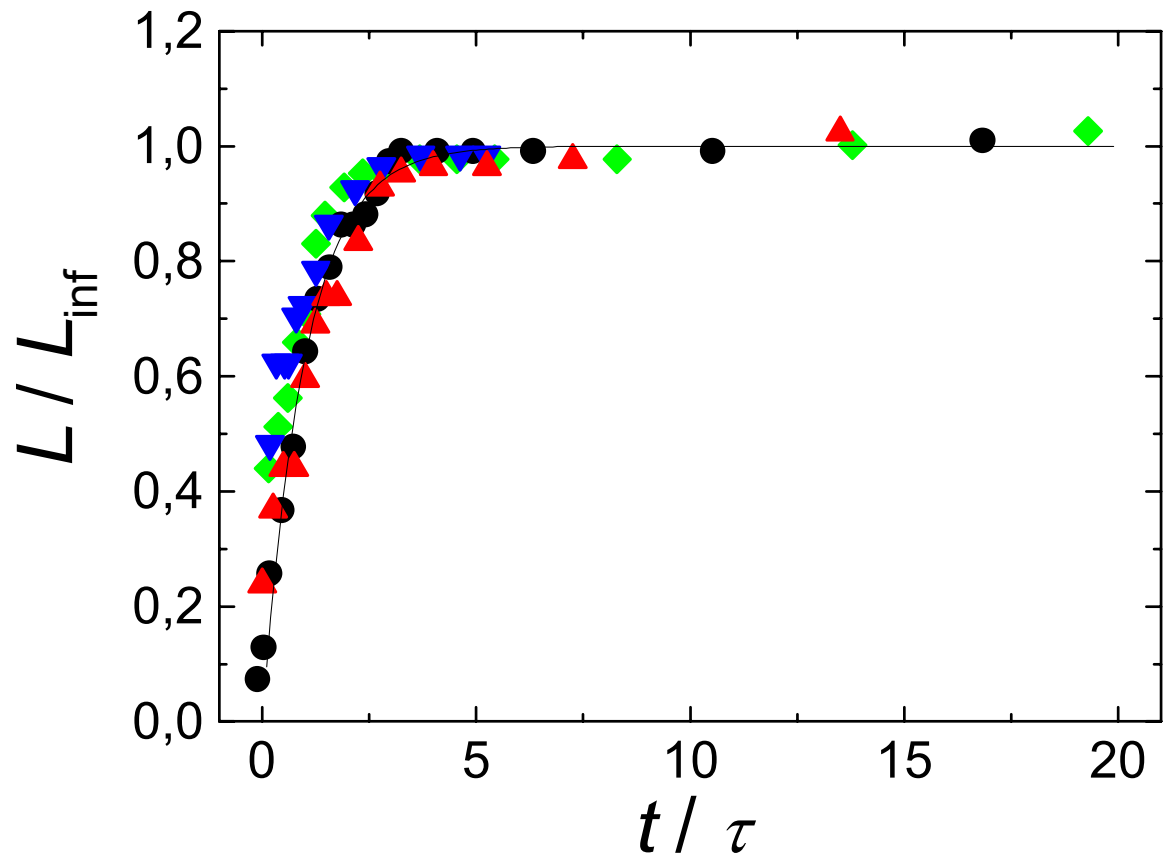
$$\epsilon(t) = \epsilon_{inf} (1 - \exp(-t/\tau))$$

since  $\epsilon \propto L$

$$L = L_{inf} (1 - \exp(-t/\tau))$$



Using continuum theory calculations to collapse data



$\tau = 130 \pm 40 \text{ s}$      $\rightarrow$     Elastic modulus  $\cong 0.3\text{Pa}$      $\checkmark$

Frenkel & Ladd, Phys. Rev. Lett. **59**, 1169 (1987)

# Nano indentation at the micron scale

