

How Glasses Relax and Go With the Flow

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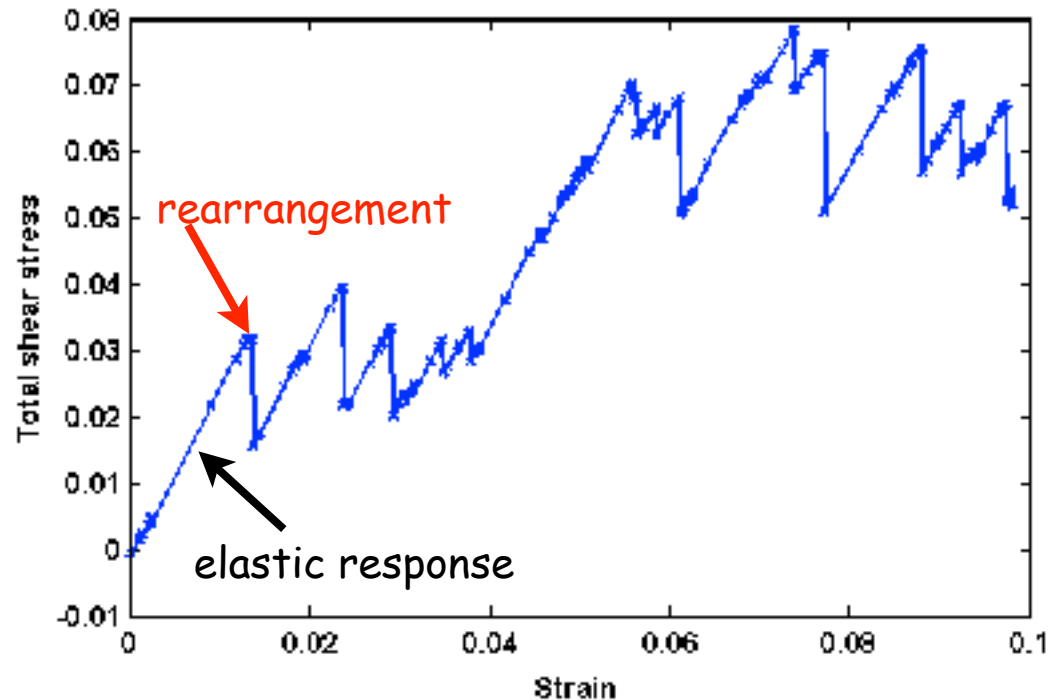
Tim Kaxiras

Harvard

IRG3 of UPenn MRSEC

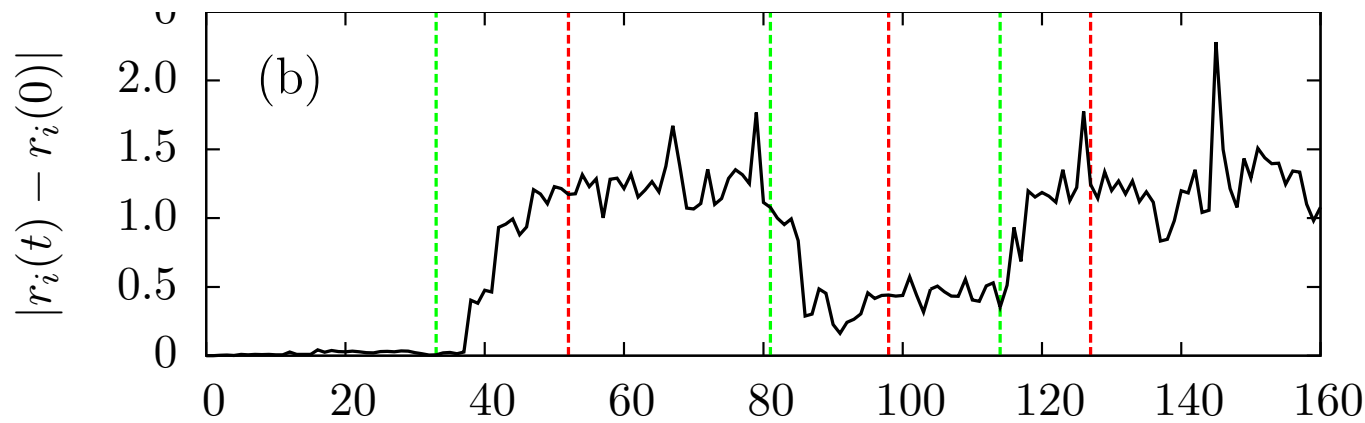
How Solids Flow

- When **ordinary liquids** flow, any particle can rearrange
- Consider solids: they initially respond elastically but above yield strain they begin to
- **Crystals**
 - Particles at dislocations far more likely to rearrange
- **Glasses**
 - Can we identify particles with high propensity to rearrange?
 - Challenge to identify such particles from structure is nearly 50-year-old problem (Argon 1970's)

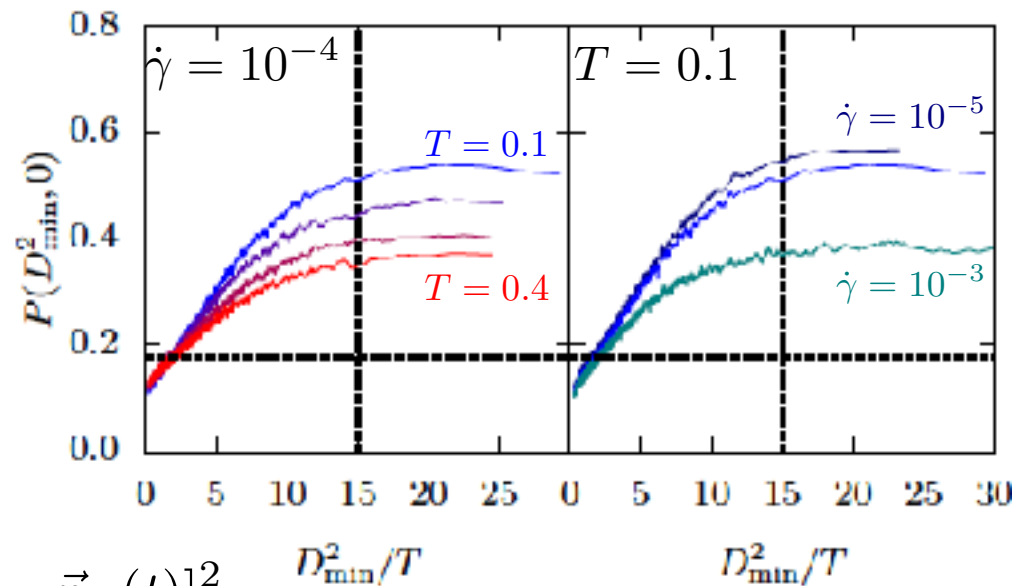


How to Tell if a Particle is Rearranging

- Characterize rearrangements in terms of P_{hop}

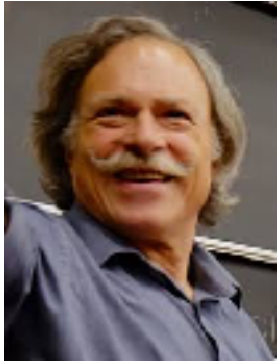


- or D_{min}^2



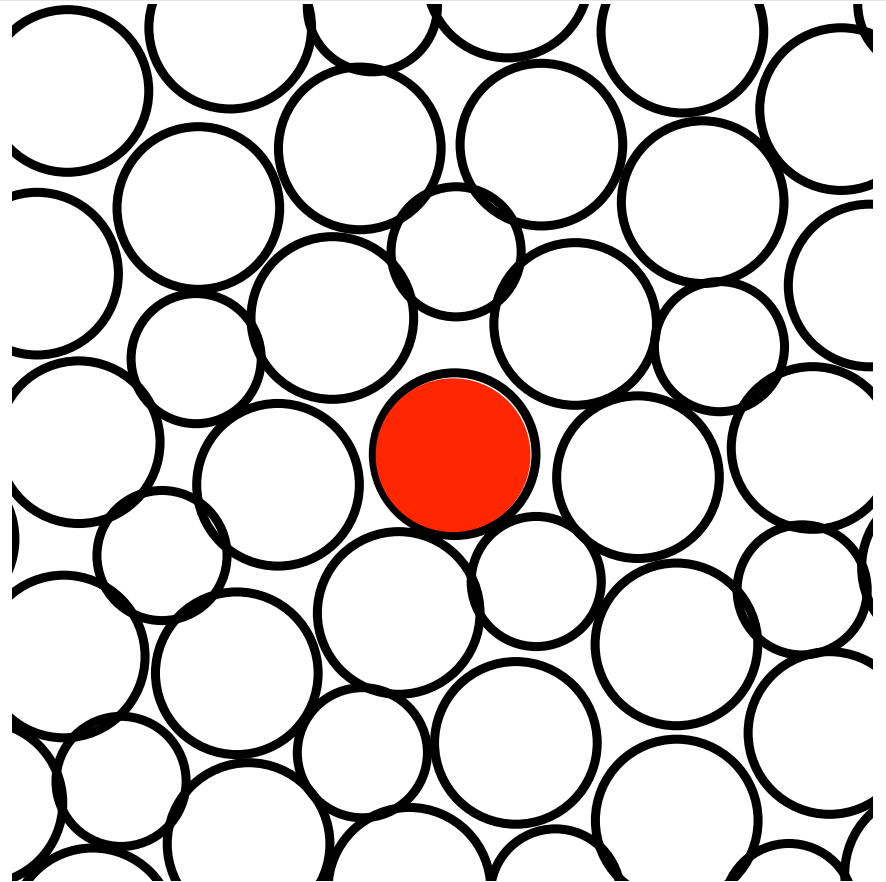
$$D_{\text{min}}^2(i; t) = \frac{1}{z} \sum_j [\vec{r}_{ij}(t + \Delta t) - \Lambda_{\text{min}} \vec{r}_{ij}(t)]^2$$

Machine Learning to Identify Particles Likely to Rearrange



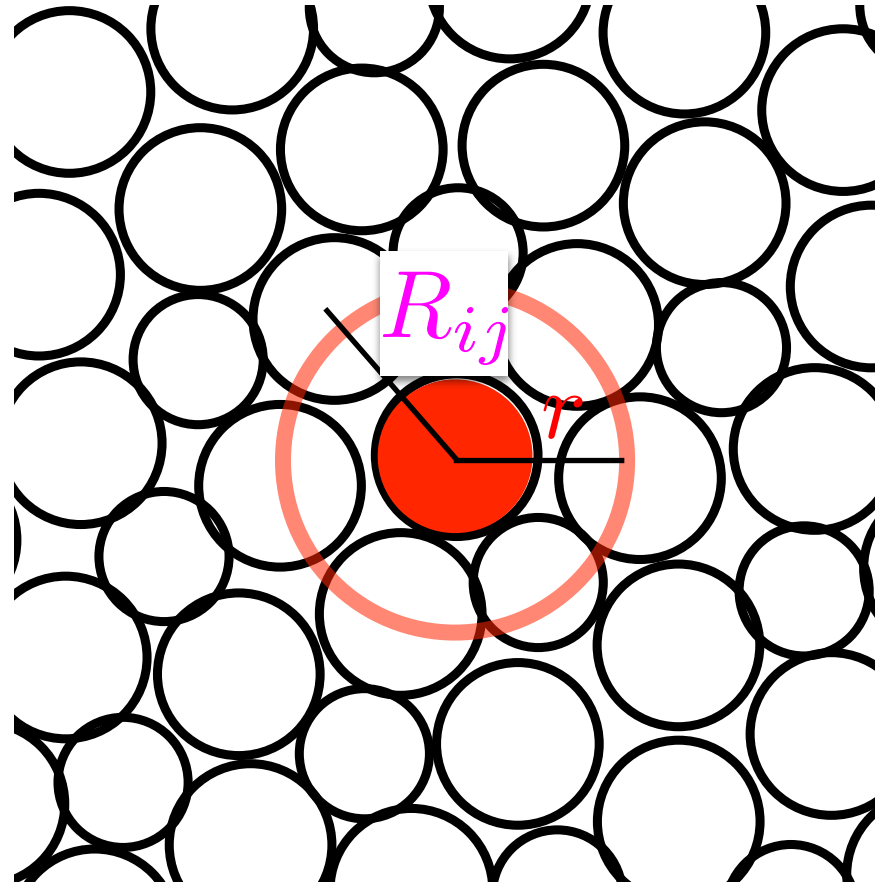
How does iPhoto figure out this is Paul Chaikin?

- Don't just use one quantity to characterize structure
- Use **MANY**
- Use small subset of data to infer linear combination of quantities that correlates most strongly with particle rearrangements



Machine Learning Approach

- Don't just use one quantity to characterize structure
- Use **MANY**
- Use small subset of data to infer linear combination of quantities that correlates most strongly with particle rearrangements
- Gain physical intuition *a posteriori*



$$G_X(i; r, \sigma) = \frac{1}{\sqrt{2\pi}} \sum_{j \in X} e^{-\frac{1}{2\sigma^2} (R_{ij} - r)^2}$$

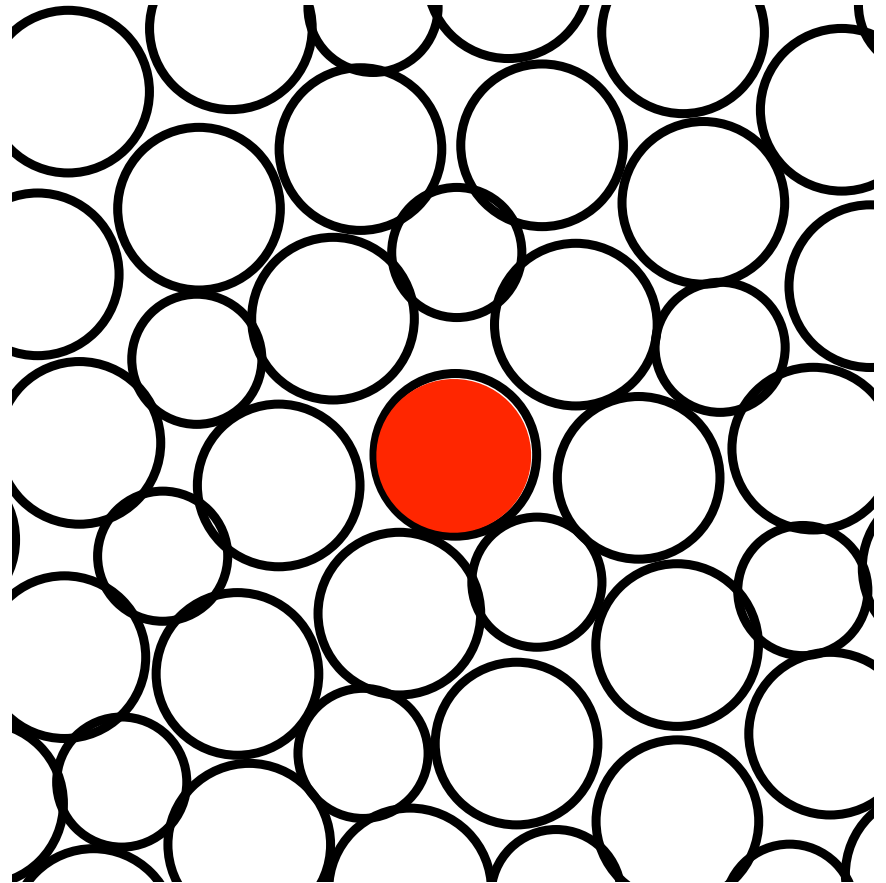
species

radius to probe

distance between particle i and j

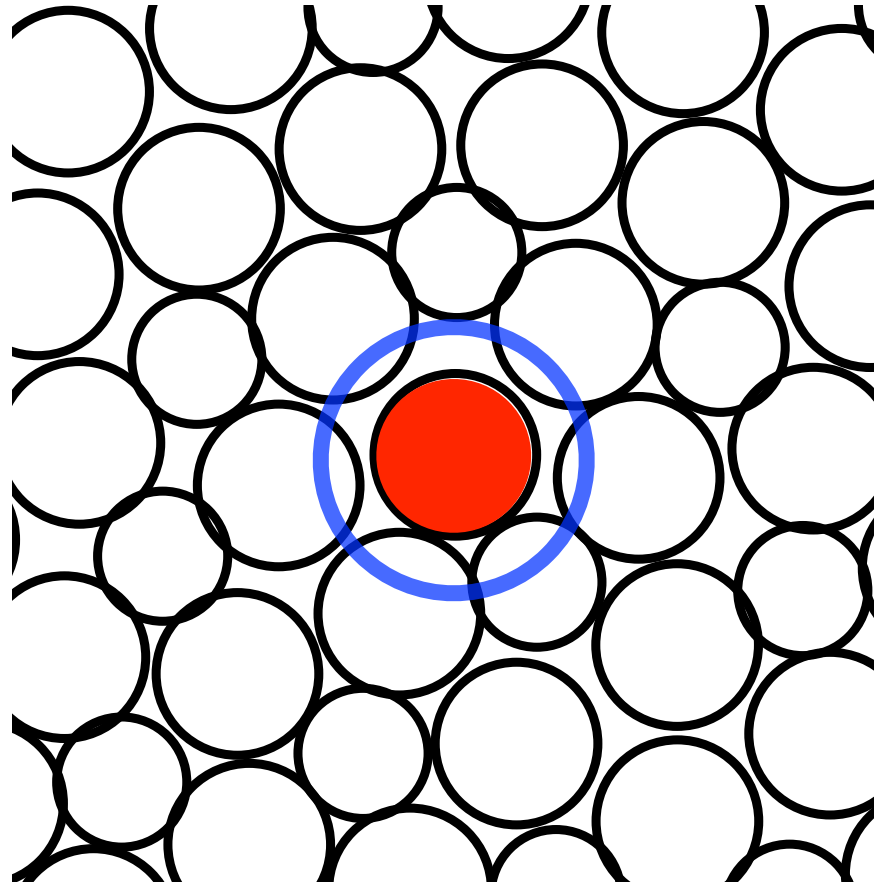
Machine Learning Approach

- Don't just use one quantity to characterize structure
- Use **M** quantities eg $g(r)$ at many r
- Use small subset of data to infer linear combination of quantities that correlates most strongly with particle rearrangements
- Construct M -dim vector whose components are values of $g(r)$ at different r



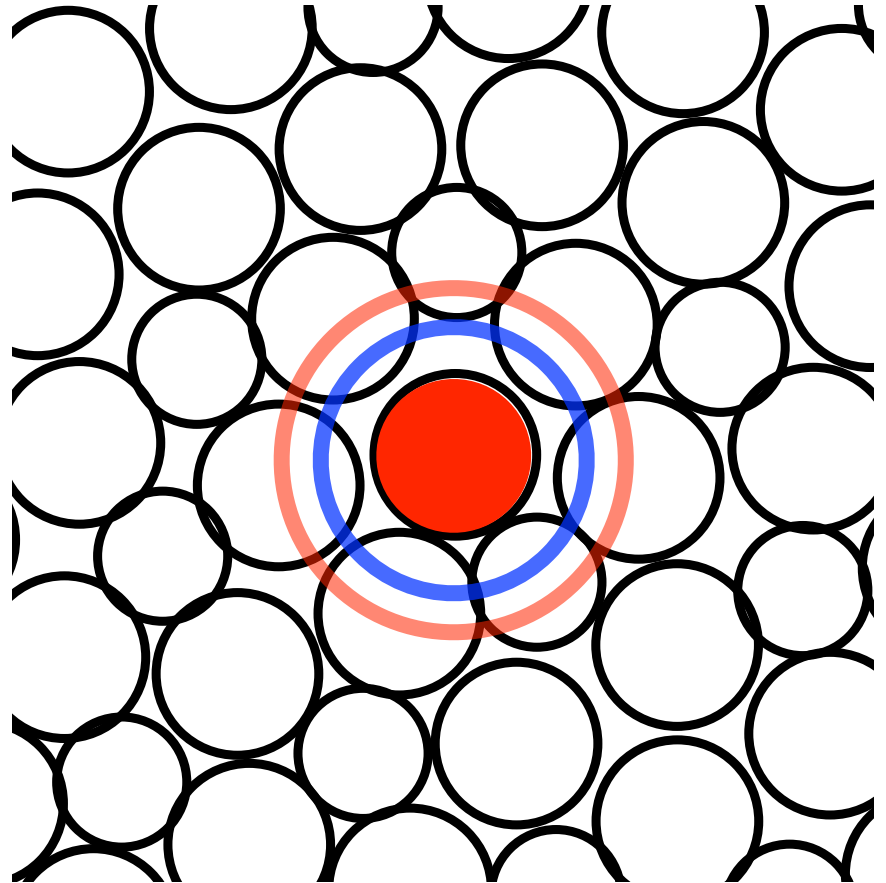
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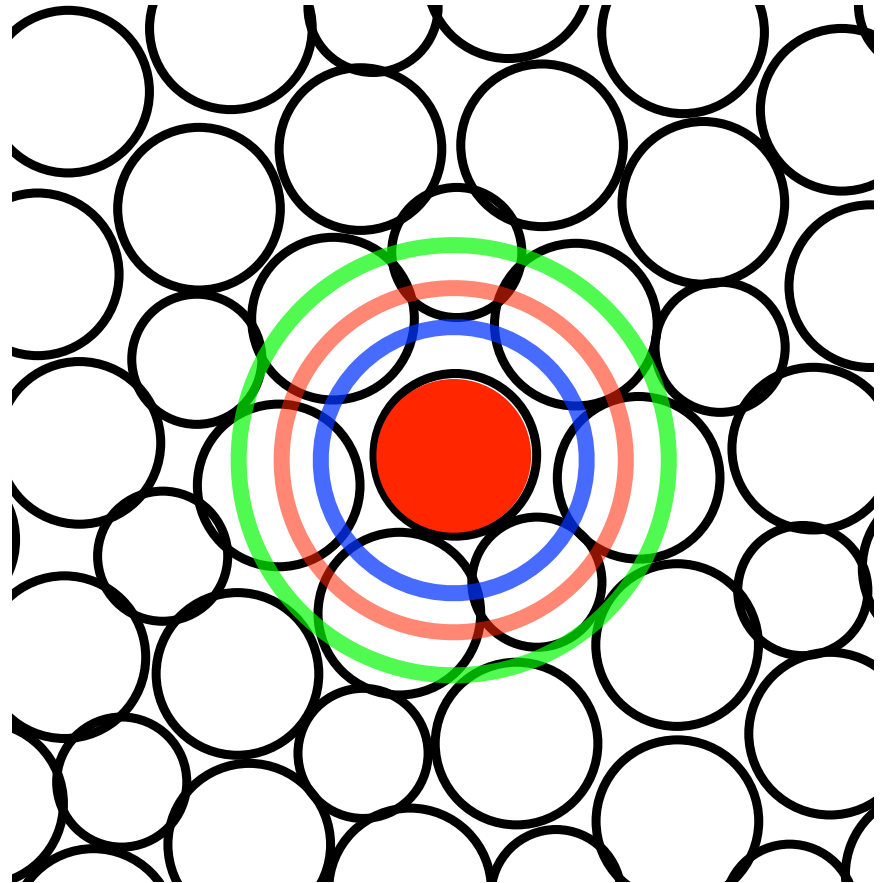
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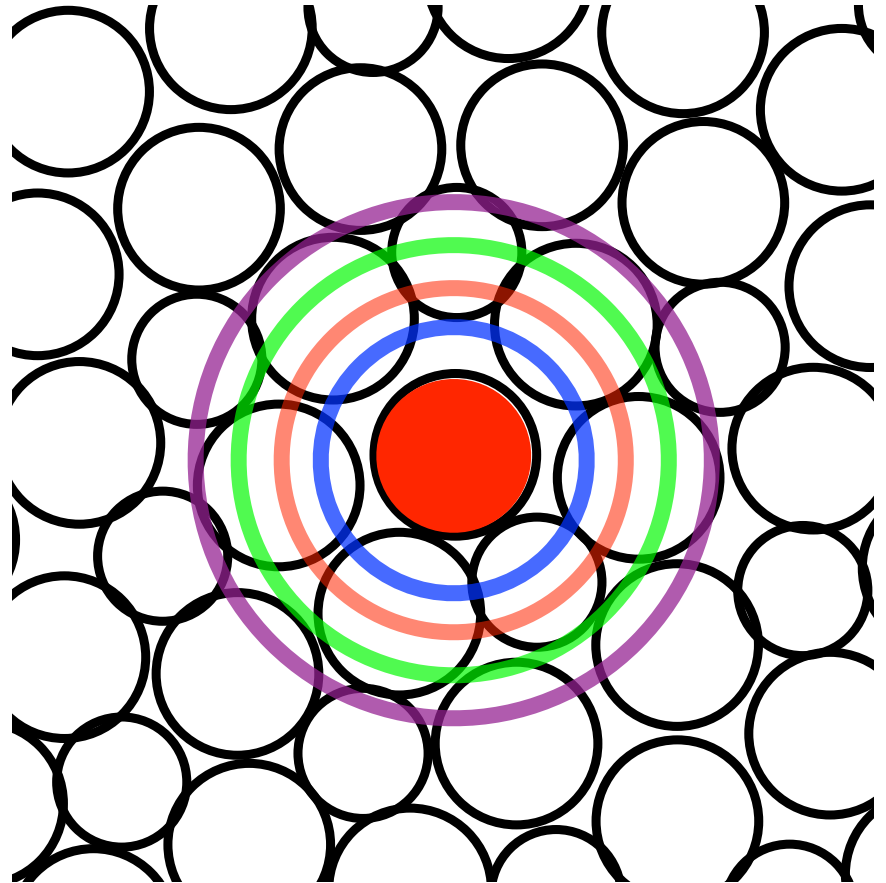
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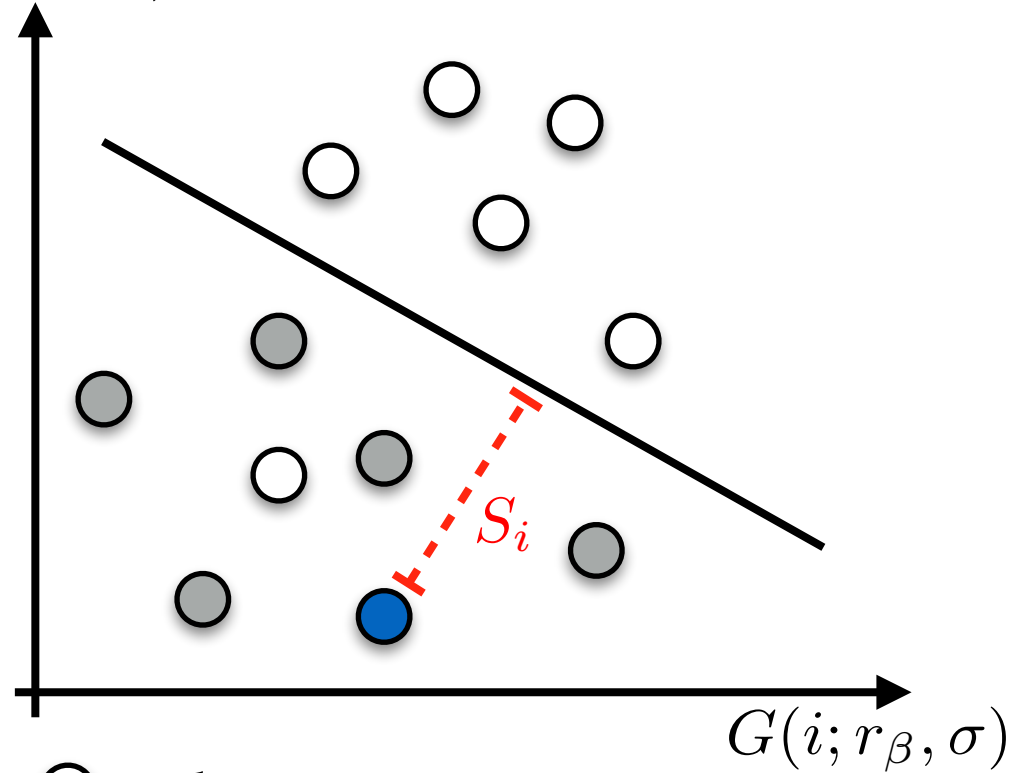
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Training Set

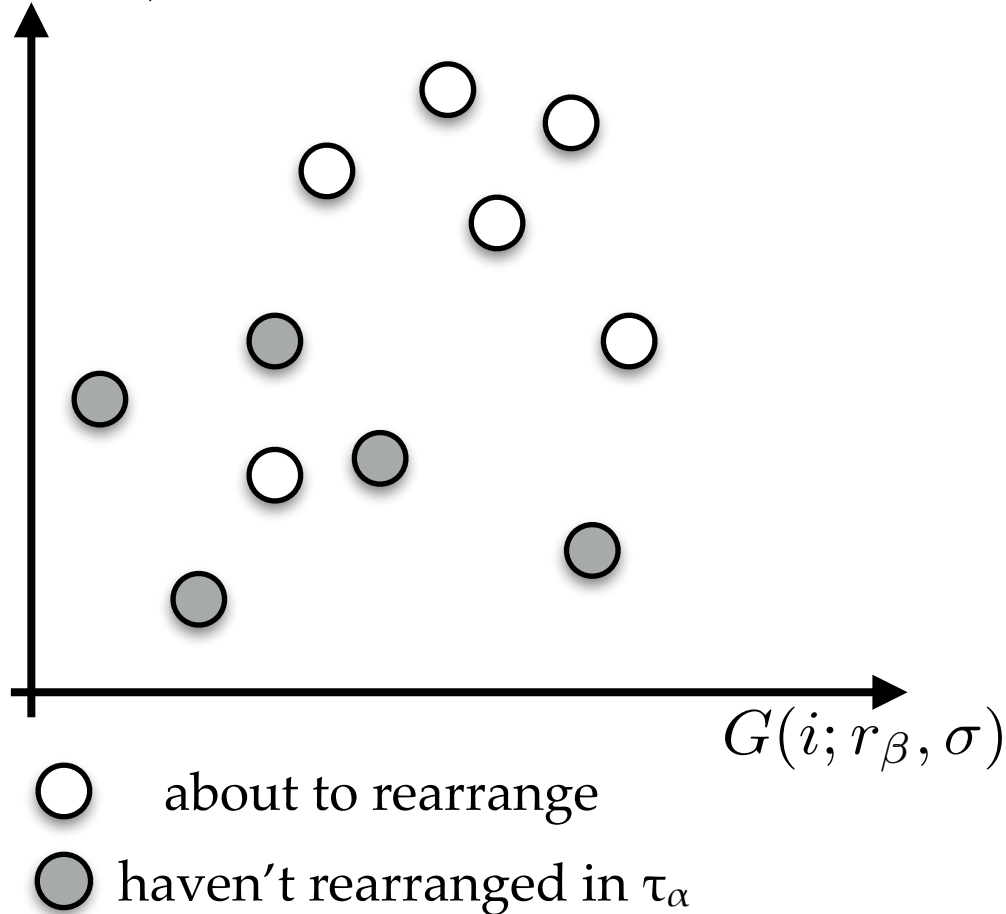
- Identify small number of $G(i; r_\alpha, \sigma)$
 - particles that are about to rearrange
 - particles that haven't rearranged in time τ_α
- construct **hyperplane** that best separates two groups
- For new data, **softness** is signed distance from hyperplane



- about to rearrange
- haven't rearranged in τ_α
- new data
- S_i softness value

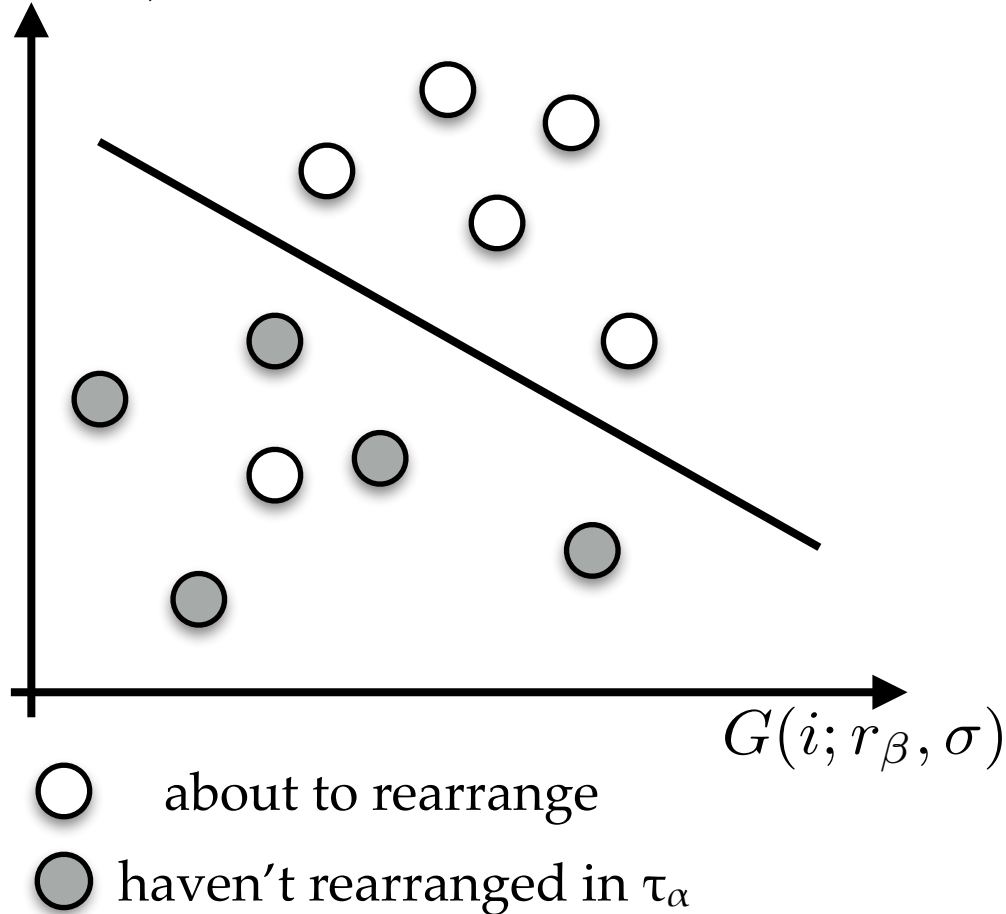
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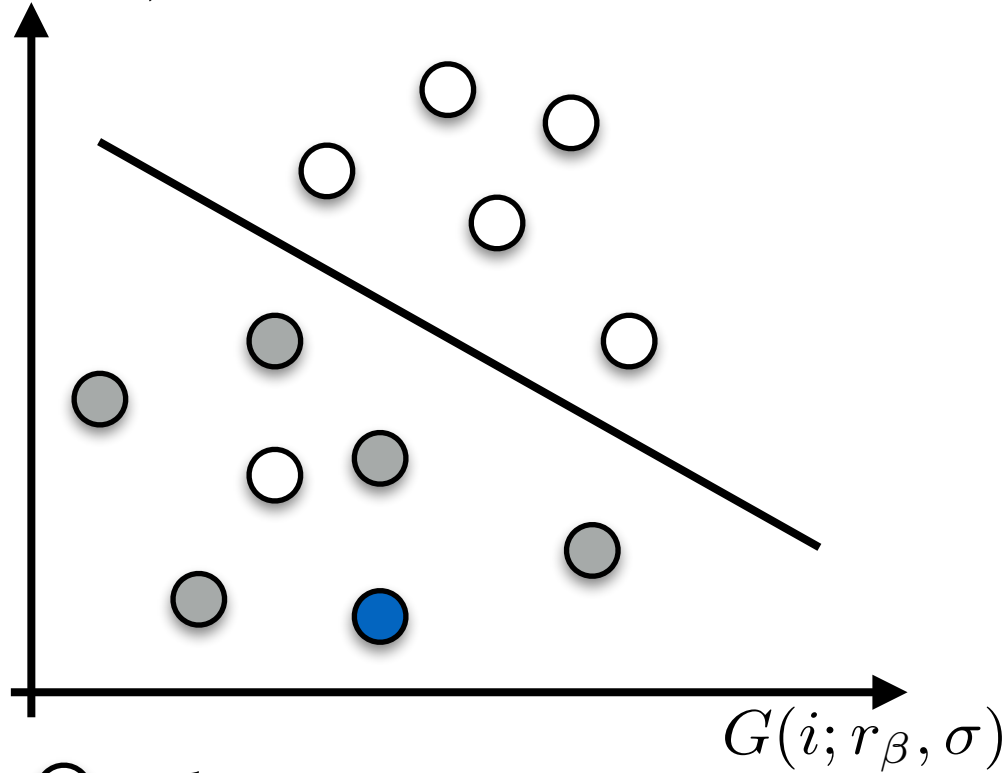
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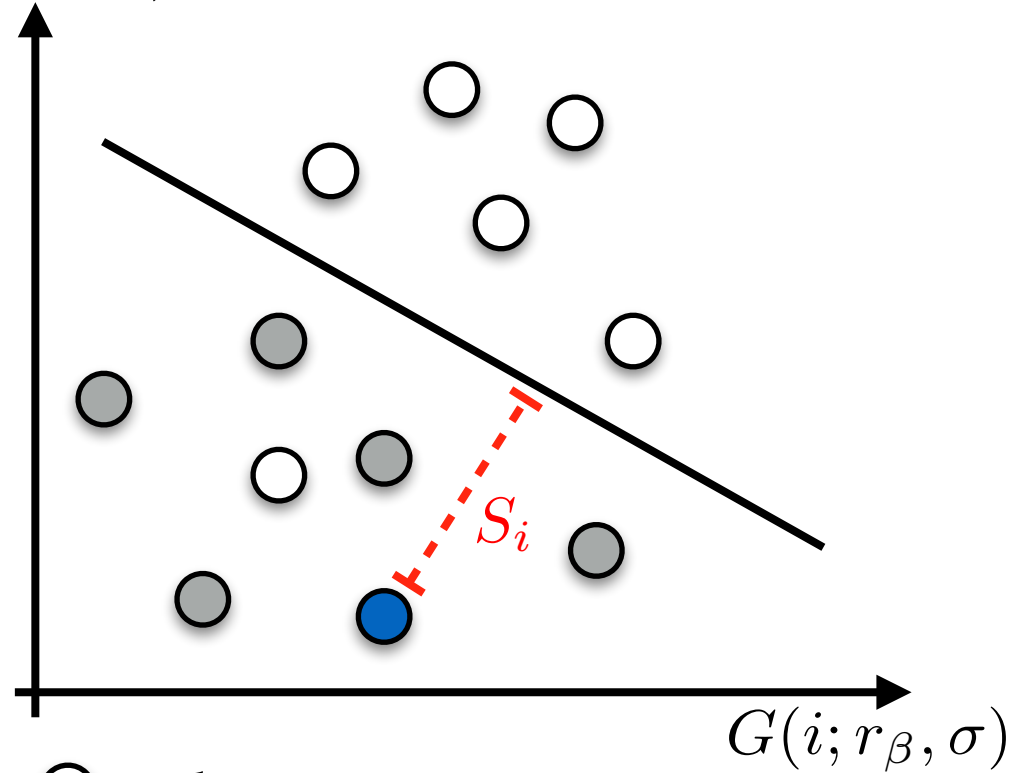
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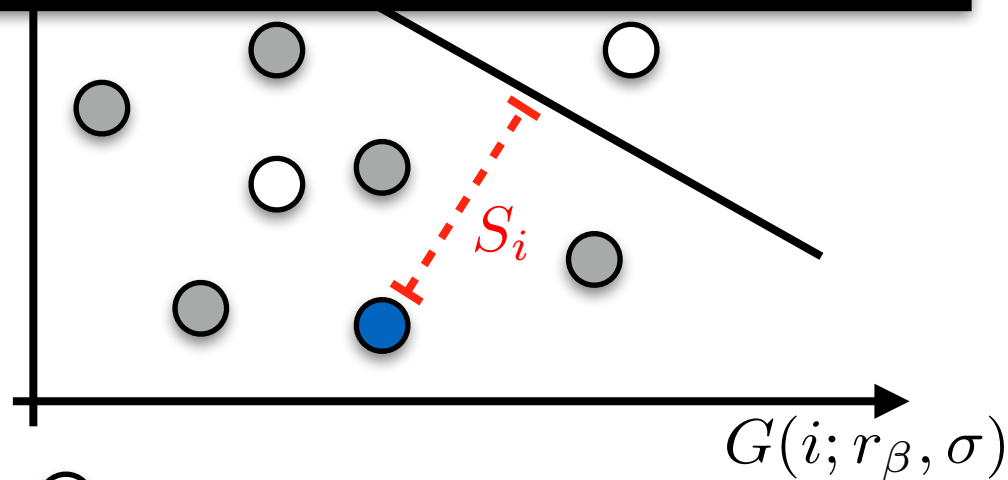
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Hyperplane encodes relation between **structure** and **dynamics** and can be interpreted physically *a posteriori*

rearranged in time τ_α

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Training Set

Hyperplane encodes relation between **structure** and **dynamics** and can be interpreted physically *a posteriori*

rearranged in time τ_α

Hyperplane is different for each different system so unlike dislocations, **softness is not universal**

signed distance from hyperplane

$G(i; r_\beta, \sigma)$

- about to rearrange
- haven't rearranged in τ_α
- new data
- S_i softness value

Cubuk, Schoenholz, Rieser, Malone,
Rottler, Durian, Kaxiras, AJL PRL (2015)
Cubuk, Schoenholz, Kaxiras, AJL Nat Phys
(2016)

What is Softness?

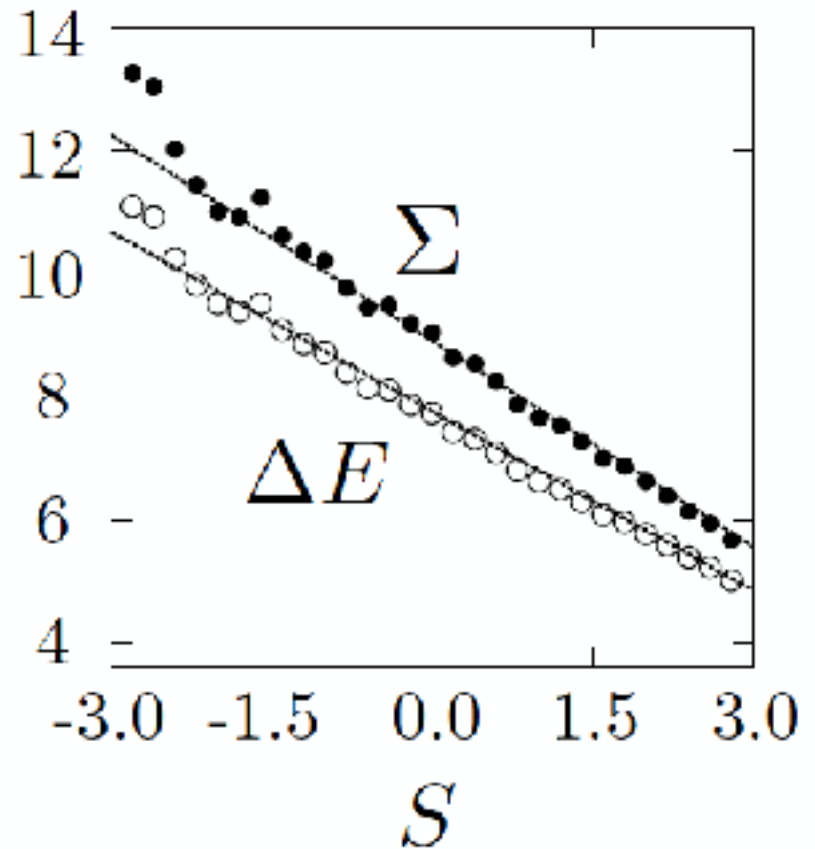
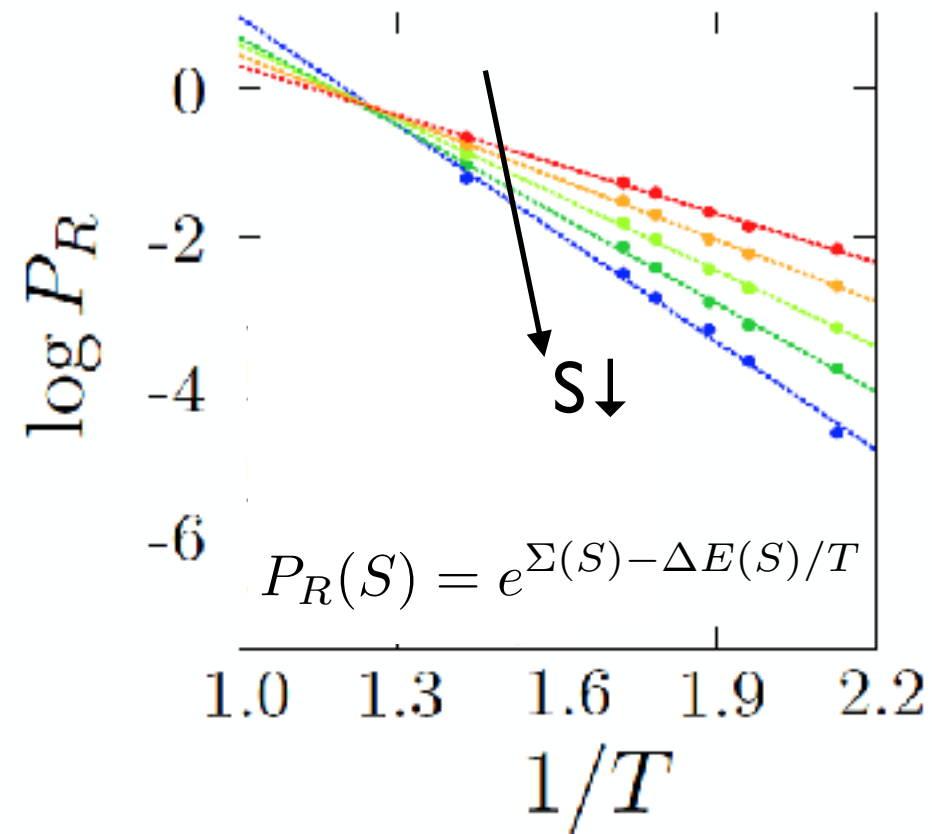
- More generally, we use two families of structural variables

$$G_X(r, i, \sigma) = \frac{1}{2\pi} \sum_{j \in X} e^{-(R_{ij}-r)^2/2\sigma^2}$$

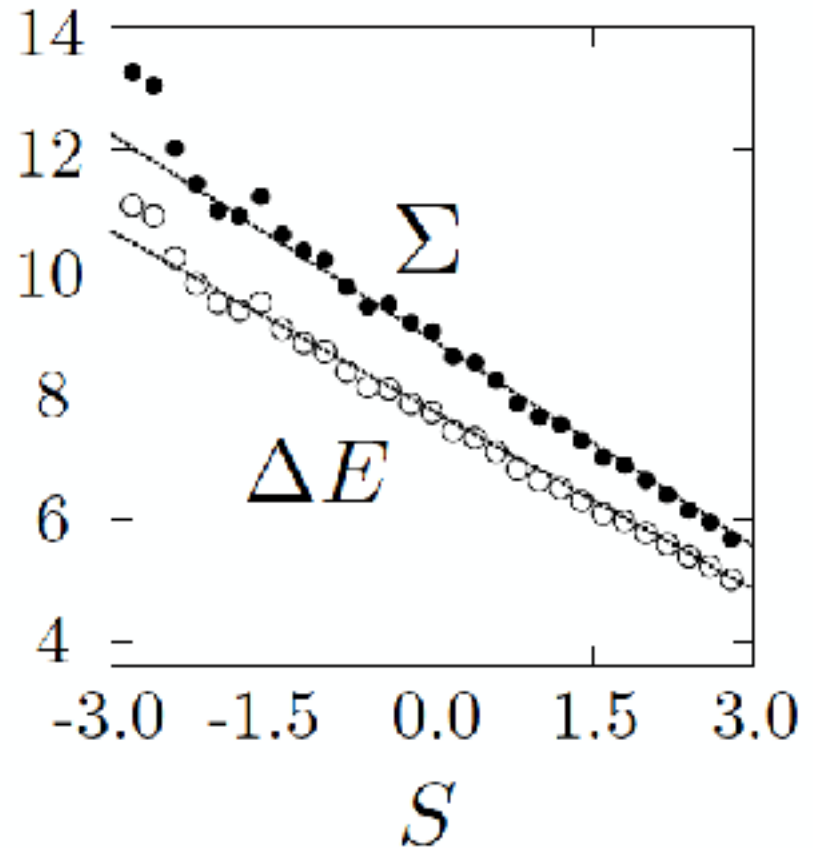
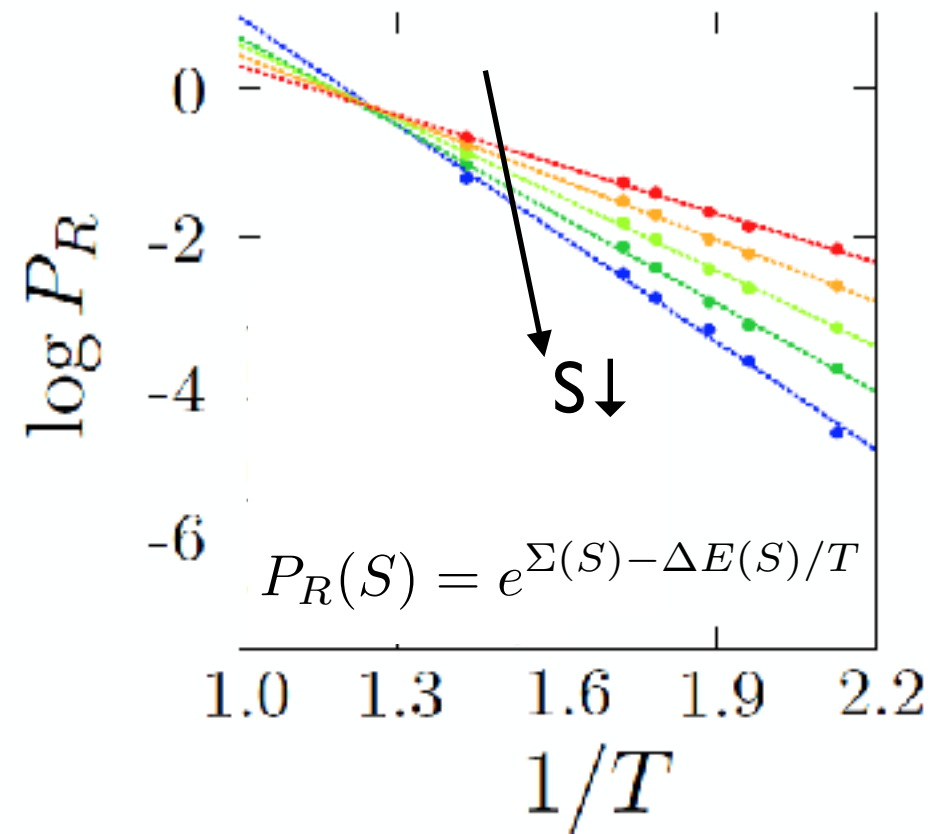
$$Q_{YZ}^X(i; \xi, \lambda, \zeta) = \sum_j \sum_k e^{-(R_{ij}^2 + R_{ik}^2 + R_{jk}^2)/\xi^2} (1 + \lambda \cos \theta_{ijk})^\zeta$$

- Q measures relative bond orientation
- For Kob-Andersen Lennard-Jones glass
 - 90% accuracy using 100 radial functions and 22 angular functions
 - 88% accuracy using 100 radial functions alone
 - For central A particle, 77% accuracy using just the height of 1st peak of g_{AA} , g_{AB}

Rearrangement Probability for fixed S is Arrhenius!



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There is a characteristic energy barrier associated with particles of given softness

Distribution of S /energy barriers leads to heterogeneous dynamics

Look at Softness and Plasticity in Range of Systems

- Constituent particles:
 - atoms (Sim)
 - Lennard-Jones
 - BKS silica
 - oligomer molecules (Sim) ([Riggleman](#))
 - colloids (Expt)
 - PNIPAM ([Yodh](#))
 - PMMA ([Gianola](#))
 - grains (Expt) ([Durian](#))

72-95% of rearrangements occur at particles with $S_i > 0$

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Interactions:

van der Waals attractions

Covalent attractions

Screened electrostatic repulsions

Hard core repulsions

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Source of Rearrangements:

thermal fluctuations
mechanical load

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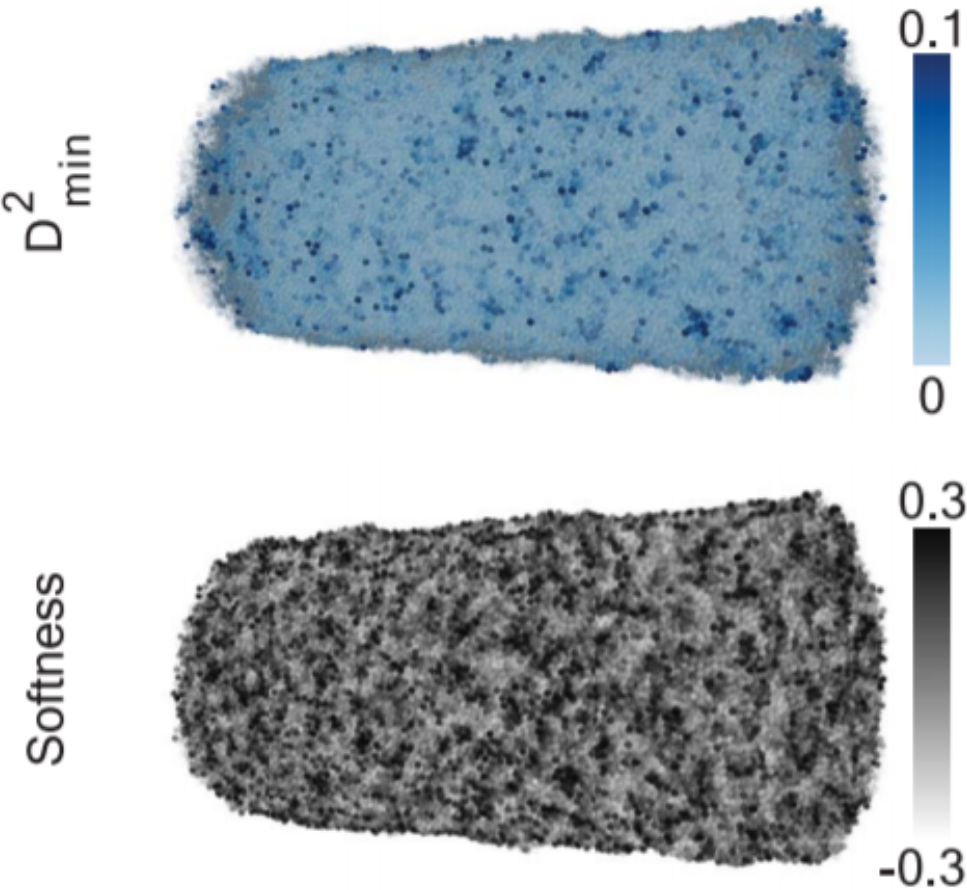
thermal fluctuations
mechanical load

Systems span over 7 orders of magnitude in particle size and
13 orders of magnitude in elastic modulus

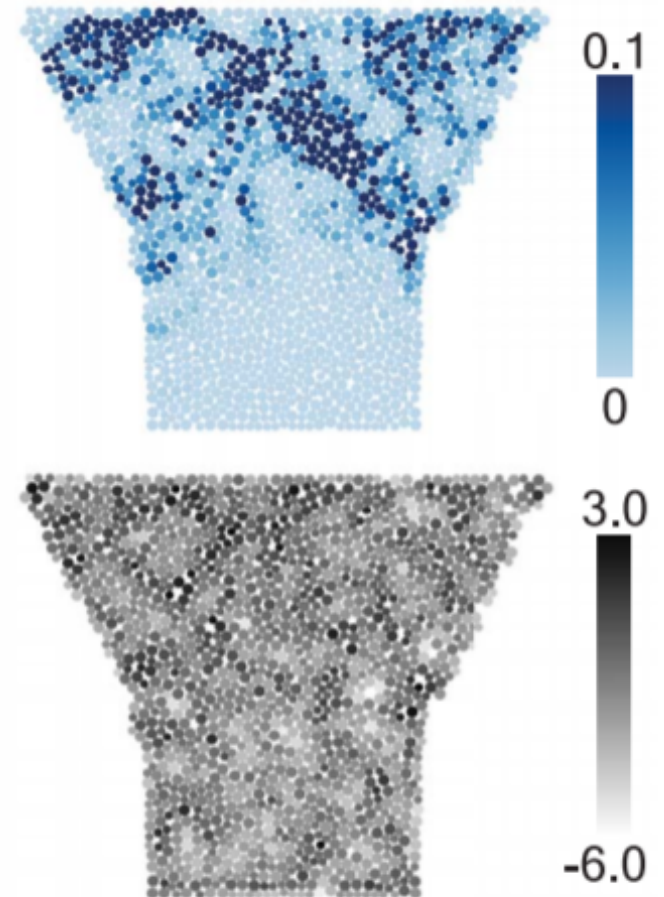
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Link Between Structure and Rearrangements

Oligomer Pillar Simulations

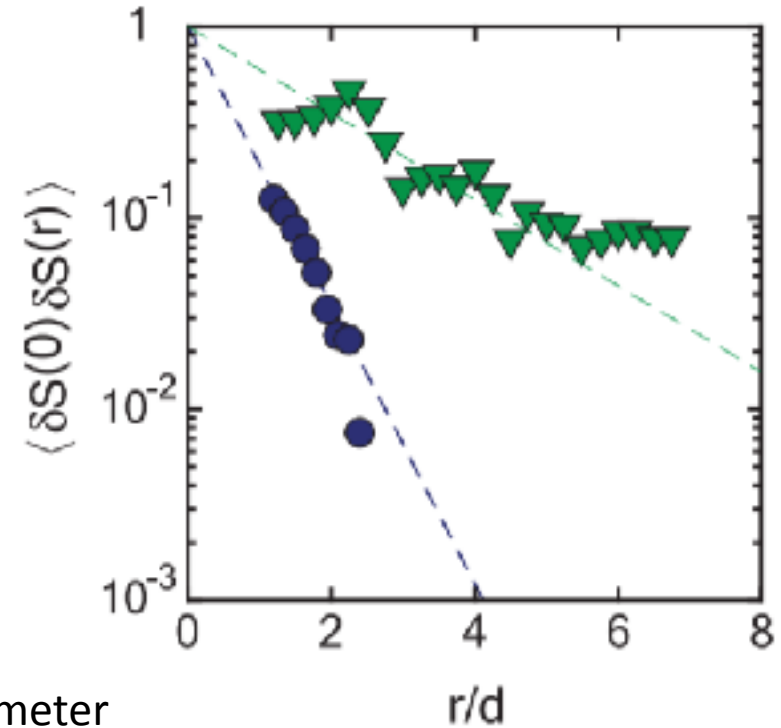
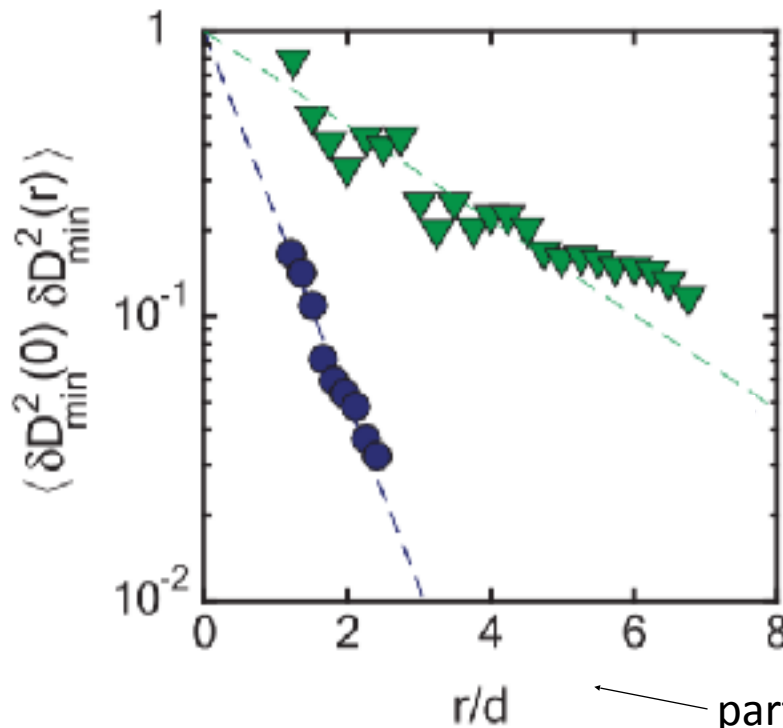


2D Granular Pillar Experiments



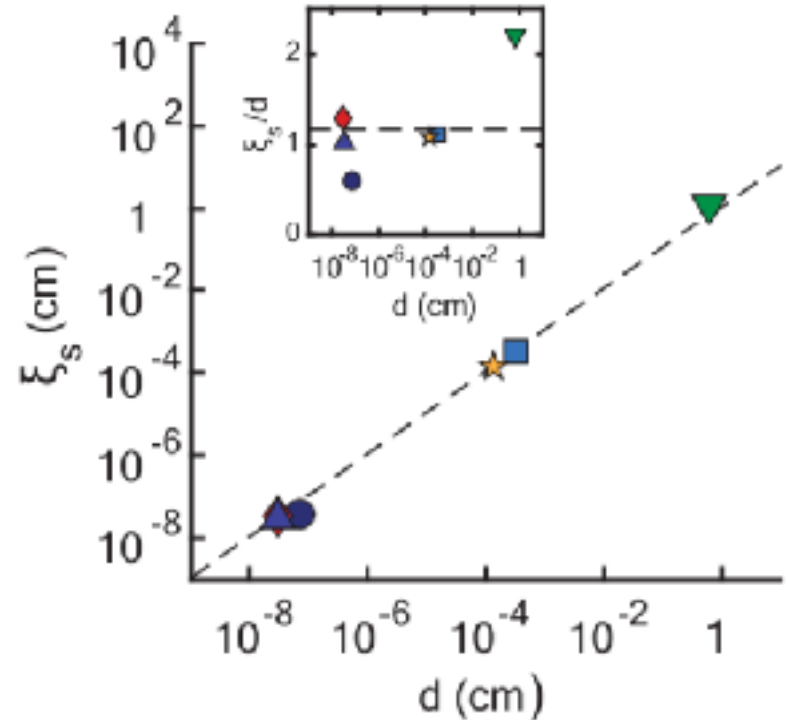
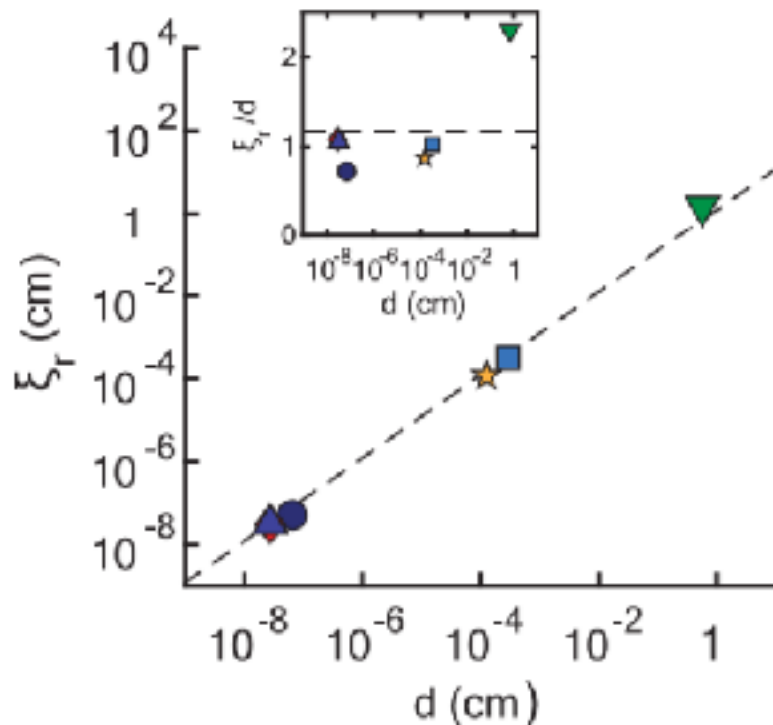
- Small fraction of particles are rearranging; 10-30% of particles are soft

Spatial Correlations for $D_{\min}^2$ and softness are similar



Exponential fit to measure size of rearrangement or soft region

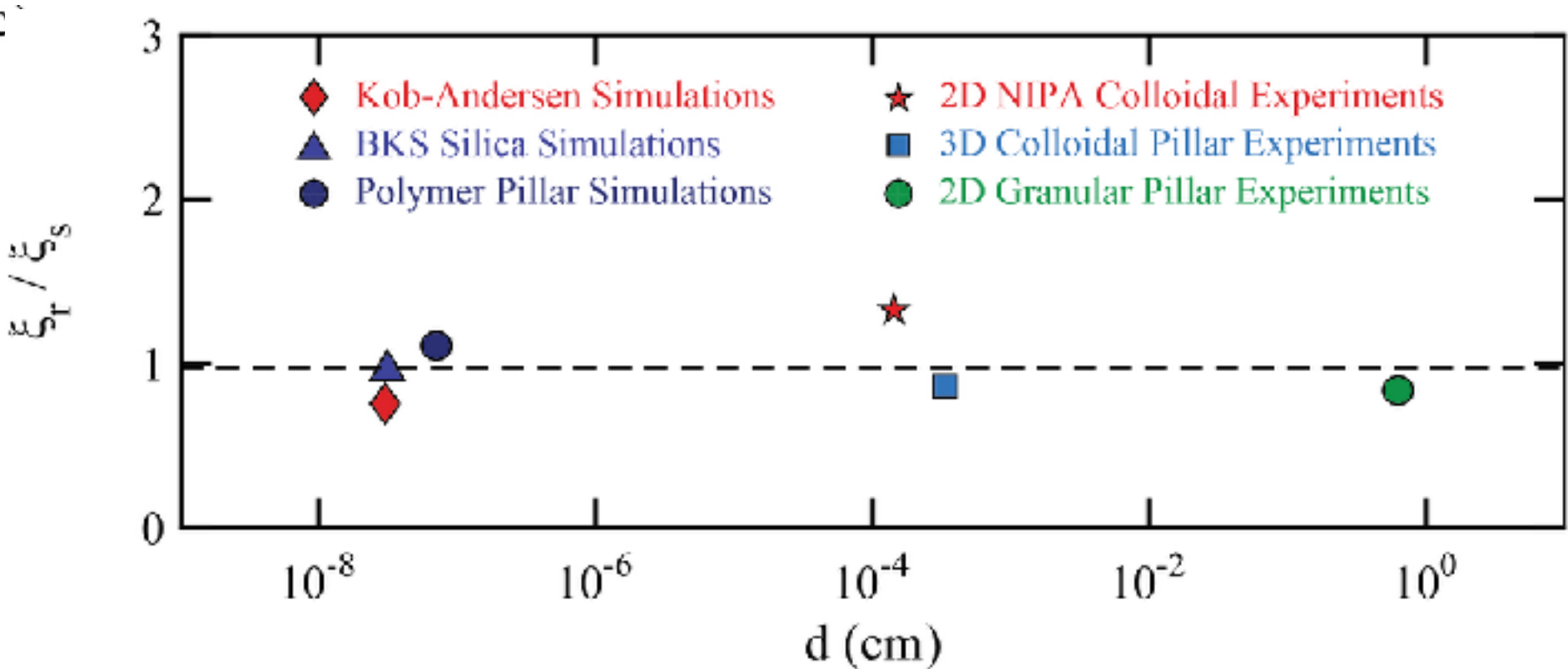
Common Rearrangement and Soft Spot Size



- ◆ Kob-Andersen Simulations
- ▲ BKS Silica Simulations
- Oligomer Pillar Simulations
- ★ 2D PNIPAM Colloidal Experiments
- 3D PMMA Colloidal Experiments
- ▼ 2D Granular Pillar Experiments

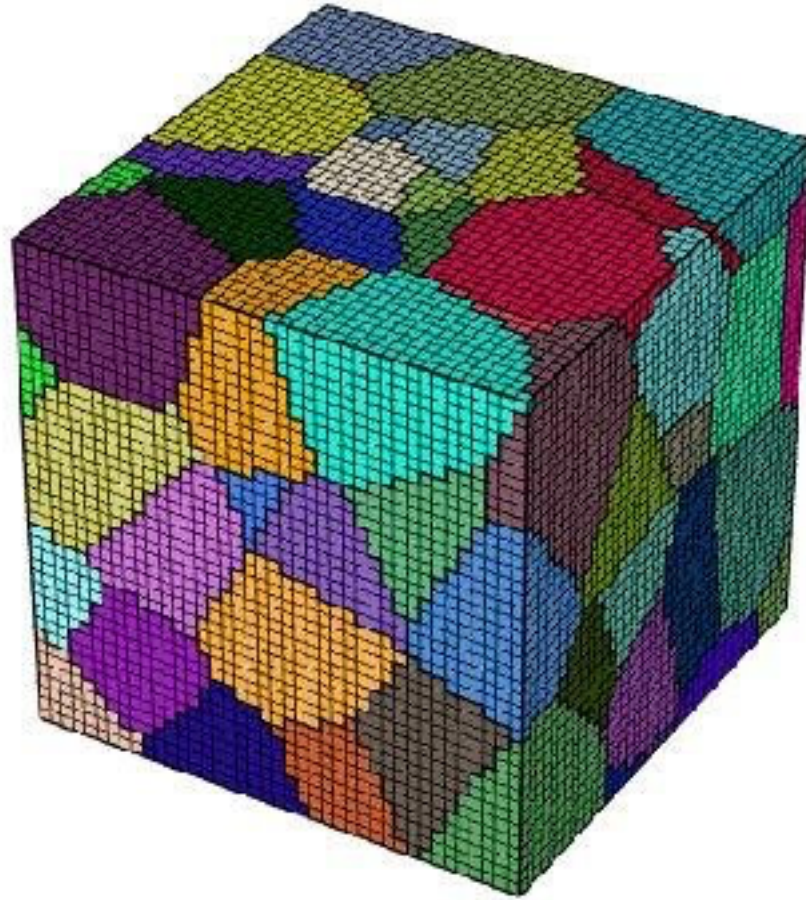
$\xi/d \approx 1$
for both $D_{\min}^2$ and S

Rearrangement Size is Softness Correlation Length



- We can rationalize the common size of rearrangements as a reflection of the common range of softness correlations

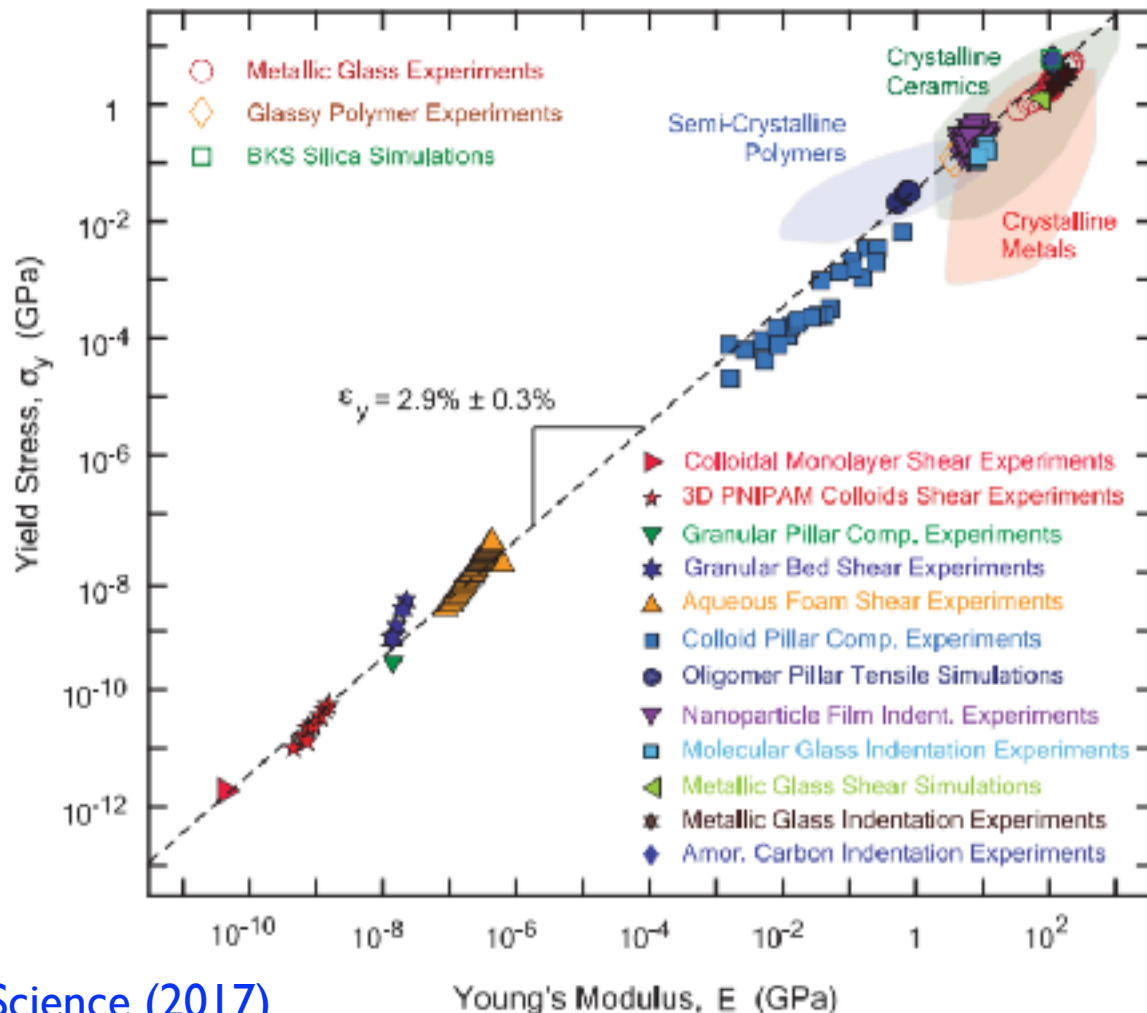
What About Crystals?



- Softness correlation length will reflect size of grain boundaries, etc—**not universal for crystals**
- Size of rearrangements can be extended—**not universal for crystals**

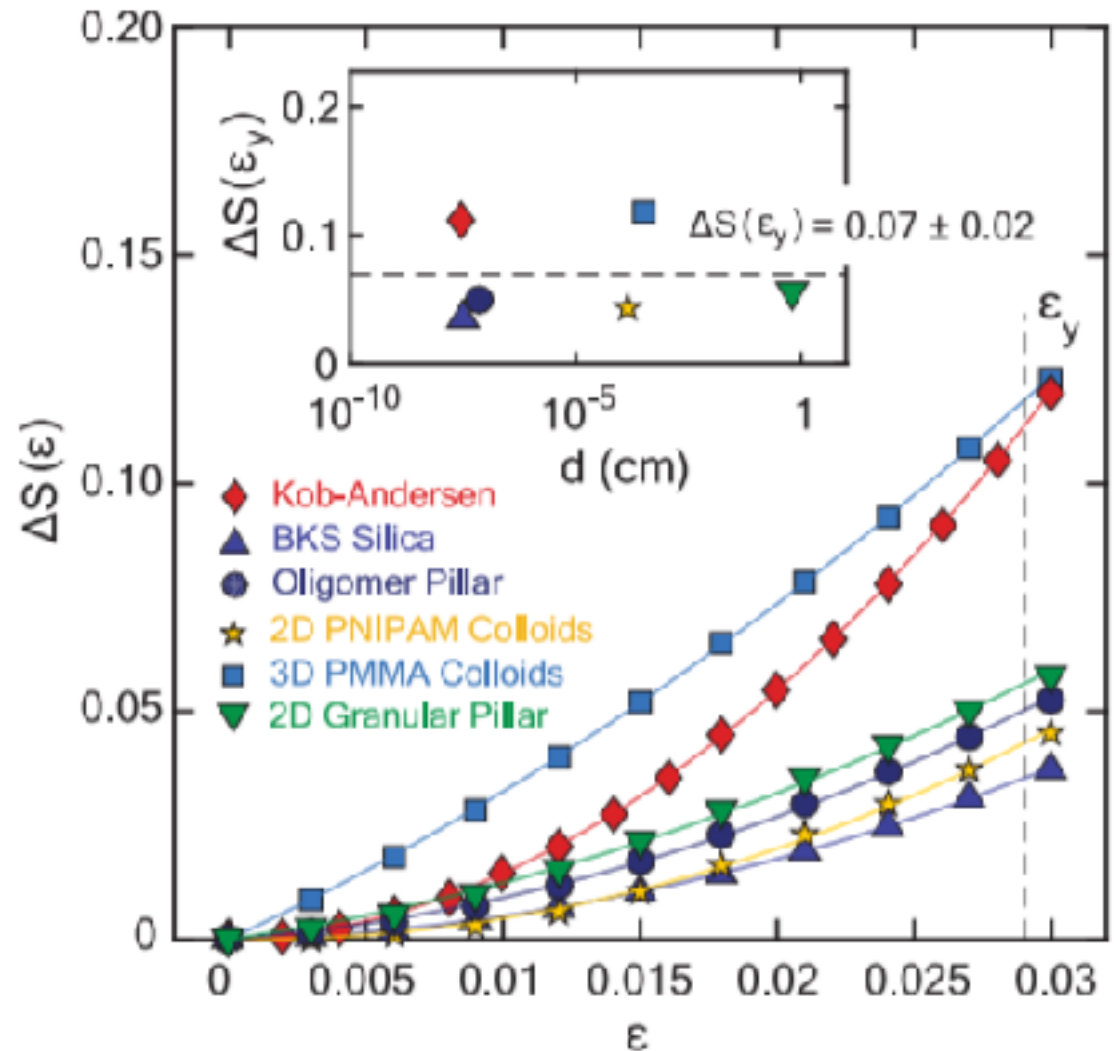
Commonality in the Onset of Rearrangements with Strain

- There is a **common yield strain** for systems spanning >13 orders of magnitude in loading modulus in glassy systems—but **not crystals**



Can we rationalize common yield strain in terms of softness?

- Apply affine strain to neighborhood of each particle
- Use hyperplane to recompute softness of particle as function of strain
- Compare shift of softness with strain to width of softness distribution
- There is a **common response of softness to strain**



Softness and Plasticity

- Softness does not have universal definition—it depends on particle interactions, etc
- BUT emergent properties ARE UNIVERSAL
 - softness correlation length is on scale of particle diameter
 - softness response to strain is about the same
- This is true for systems with interactions that are repulsive or that have van der Waals, covalent or metallic bonding
 - range of system sizes $> 10^7$
 - range of loading moduli $> 10^{13}$
- Crystals are the opposite—there is a universal definition of dislocation, but emergent properties are NONUNIVERSAL

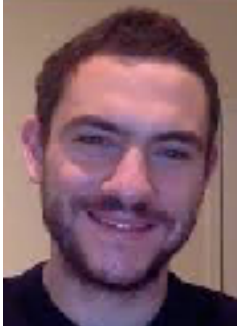
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Perhaps we will someday understand plasticity in disordered solids better than plasticity in crystals

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Thanks to



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