# Jamming and the Glass Transition

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# The Jamming Scenario



- Focus on foams/emulsions or frictionless granular material
  - soft, repulsive, finite-range spherically-symmetric potentials
- Such systems have T=0 1st/2nd-order phase transition

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## How we study Point J

- Generate configurations near J
  - Start with some initial config

$$V(r) = \begin{cases} \frac{\varepsilon}{\alpha} \left(1 - \frac{r}{\sigma}\right)^{\alpha} & r \le \sigma \\ 0 & r > \sigma \end{cases}$$

- Conjugate gradient energy minimization (inherent structures, Stillinger & Weber)
- Classify resulting configurations



#### Onset of Overlap is Onset of Jamming



D=2 D=3

•Pressures for different states collapse on a single curve

•Shear & bulk moduli, G & B, vanish at same  $\phi_c$ 

D. J. Durian, PRL 75, 4780 (1995);

C. S. O'Hern, S. A. Langer, A. J. Liu, S. R. Nagel, PRL 88, 075507 (2002).

#### Onset of Overlap has 1st-Order Character



Durian, PRL **75**, 4780 (1995).

O'Hern, Langer, Liu, Nagel, PRL 88, 075507 (2002).

Majmudar, Sperl, Luding, Behringer, PRL 98, 058001 (2007).

- What is the minimum number of interparticle contacts needed for mechanical equilibrium?
- No friction, spherical particles, D dimensions
  Match

unknowns (number of interparticle normal forces)

- equations (force balance for mechanical <sub>J</sub> stability)
- -Number of unknowns per particle=Z/2
- -Number of equations per particle = D

Phillips, Thorpe, Boolchand, Edwards, Ball, Blumenfield



James Clerk Maxwell

Z = 2D

#### Isostaticity and Diverging Length Scale

M. Wyart, S.R. Nagel, T.A. Witten, EPL 72, 486 (05)

•For system at  $\phi_c$ , Z=2d

 Removal of one bond makes entire system unstable by adding one soft mode

-This implies diverging length as  $\varphi \text{--} \diamond_c \ ^+$ 



For  $\phi > \phi_c$ , cut bonds at boundary of circle of size L Count number of soft modes within circle

$$N_s \approx L^{d-1} - (Z - Z_c)L^d$$

Define length scale at which soft modes just appear

$$\ell \approx \frac{1}{Z - Z_c} \approx \left(\phi - \phi_c\right)^{-0.5}$$

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# Consequences of Diverging Length Scale

- Solids are all alike at low T or  $\omega$ :
  - density of vibrational states  $D(\omega) \sim \omega^2$  in d=3
  - vibrational heat capacity  $C(T) \sim T^3$
  - thermal conductivity  $\kappa$  (T)~Cv  $\ell$  ~T<sup>3</sup>
- Why?

Low-frequency excitations are sound modes. At long length scales all solids look elastic

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BUT at Point J, there is a diverging length scale  $\ell^*$ 

So what happens?

## Vibrations in Marginally Jammed Solids

L. E. Silbert, A. J. Liu, S. R. Nagel, PRL 95, 098301 ('05)



- More and more modes in excess of Debye prediction as  $\phi \rightarrow \phi_c$  (boson peak)
- New class of excitations distinct from sound modes originates from soft modes at Point J M. Wyart, S.R. Nagel, T.A. Witten, EPL 72, 486 (05)
- Robust for systems near isostaticity Souslov, Liu, Lubensky, PRL 103, 205503 (2009); Mao, Xu, Lubensky arXiv: 09092616

#### Vibrational Modes Predict Soft Spots

- Adams-Gibbs Cooperatively-Rearranging Regions?
- Shear Transformation Zones?
  - M. Lisa Manning's talk next week



## Summary of Point J

$$V(r) = \begin{cases} \frac{\varepsilon}{\alpha} \left( 1 - \frac{r}{\sigma} \right)^{\alpha} & r \le \sigma \\ 0 & r > \sigma \end{cases}$$

• Mixed first-order/second-order transition (RFOT)

1

• Number of overlapping neighbors per particle

$$Z = \begin{cases} 0 & \phi < \phi_c \\ Z_c + z_0 \left(\phi - \phi_c\right)^{\beta \equiv 1/2} & \phi \ge \phi_c \end{cases}$$

• Static shear modulus/bulk modulus

$$G / B \sim \left(\phi - \phi_c\right)^{\gamma \equiv 1/2}$$

• Two diverging length scales

$$\ell^* \sim \left(\phi - \phi_c\right)^{-\nu \equiv -1/2}$$
$$\ell^\dagger \sim \left(\phi - \phi_c\right)^{-\nu^\dagger \equiv -1/4}$$

# Similarity to Other Models

- This behavior has only been found in a few models, all in mean-field limit
  - 1-RSB p-spin interaction spin glass Kirkpatrick, Thirumalai, Wolynes
  - compressible frustrated Ising antiferromagnet Yin, Chakraborty
  - kinetically-constrained Ising models Sellitto, Toninelli, Biroli, Fisher
  - k-core percolation and variants Schwarz, Liu, Chayes, Toninelli, Biroli, Fisher, Harris, Jeng
  - Mode-coupling approximation of glasses Biroli, Bouchaud
  - 1-RSB solution of hard spheres Zamponi, Parisi
- These other models all exhibit glassy dynamics!!

First hint of quantitative connection between sphere packings and glass transition

## Point J and the Glass Transition



• How do ideal spheres behave at nonzero temperature?

#### Dramatic Increase of Relaxation Time



#### Similar fitting functions. WHY??

L.-M. Martinez and C. A. Angell, Nature **410**, 663 (2001).

0.4

0.6

 $T_q/T$ 

0.8

1.0

0.2

# Very Different Underlying Pictures

#### Colloidal glass transition free volume



Map of Lake O'Hara - YellowMaps To Gidlass transition

#### complex energy landscape

8/5/09 4·25 PM



Pressure is most important It governs amount of free volume



# Very Different Underlying Pictures

#### Colloidal glass transition free volume



Map of Lake O'Hara - YellowMaps To Gillass transition

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Pressure is most important It governs amount of free volume



energy barriers

There are systems for which these two transitions are the same phenomenon

## **Relaxation Time**

- Look at relaxation time along different trajectories
  - Fix p, lower T
  - Fix T, raise p



### **Relaxation Time**

- Look at relaxation
  time along different
  trajectories
  - Fix p, lower T
  - Fix T, raise p





#### **Relaxation Time**



### Data Collapse!





Data for different (p, T) collapse on to single scaling curve!

 Recall interaction potential  $V(r) = \begin{cases} \frac{\varepsilon}{2} \left( 1 - \frac{r}{\sigma} \right)^2 & r \le \sigma \\ 0 & r > \sigma \end{cases}$  particle mass We have 3 dimensional parameters in model:  $\varepsilon$ , m,  $\sigma$ interaction energy Relaxation  $\tau \sqrt{\frac{\varepsilon}{m\sigma^2}} = h\left(\frac{T}{\varepsilon}, \frac{p\sigma^3}{\varepsilon}\right)$ particle diameter

or equivalently

$$\tau \sqrt{\frac{p\sigma}{m}} = g\left(\frac{T}{p\sigma^3}, \frac{p\sigma^3}{\varepsilon}\right)$$

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$$\tau \sqrt{\frac{p\sigma}{m}} = g\left(\frac{T}{p\sigma^3}, \frac{p\sigma^3}{\varepsilon}\right)^0$$



- In low p limit, the relaxation time depends only on T/p
- Data collapse for different trajectories

# Data Collapse for Different Potentials

- pσ<sup>3</sup>/ε->O corresponds to low p limit for soft spheres AND to the hard sphere limit
- Should see collapse for any  $\alpha \ge 0$  including  $\alpha = 0$  (hard spheres)

$$V(r) = \begin{cases} \frac{\varepsilon}{\alpha} \left( 1 - \frac{r}{\sigma} \right)^{\alpha} & r \le \sigma \\ 0 & r > \sigma \end{cases}$$

# Data Collapse for Different Potentials

- $p\sigma^3/\epsilon$ ->0 corresponds to low p limit for soft spheres AND to the hard sphere limit
- Should see collapse for any  $\alpha \ge 0$  including  $\alpha = 0$  (hard spheres) **YES!** 10<sup>-</sup>

 $\lim_{T/\varepsilon \to 0} \tau \sqrt{\frac{p\sigma}{m}} = f\left(\frac{T}{p\sigma^3}\right)$ 

 $T/p\sigma^3$ 

0.2

 $10^{3}$ 

 $10^{0}$ 

O

 $10^{-10}$  $10^{2}$  $10^{2}$  $10^{2}$  $10^{1}$ 



T does work against pressure to open up free volume

```
Glass transition equivalent to colloidal glass transition
                          as p\sigma^3/\epsilon \rightarrow 0
```

0.4

Hard spheres

# Relaxation times at high pressure



- As spheres soften, relaxation time decreases (Weitz/Reichman)
- New mechanism of relaxation controlled by  $p\sigma^{3}/\epsilon$  emerges

#### Soft spheres have smaller equivalent hard sphere diameter



Rowlinson, Barker-Henderson, Andersen-Weeks-Chandler

# Strategy for choosing $\sigma_{eff}$

Andersen, Weeks, Chandler J. Chem. Phys. 54, 5237 (1971)

- Taylor expand free energy around hard-sphere potential
- Choose  $\sigma_{\rm eff}$  so that first-order functional derivative of free energy with respect to exp(-V(r)/T) vanishes



- Can calculate  $\sigma_{\rm eff}$  from the soft-sphere potential and hard-sphere properties alone!
- This approximation reproduces captures static quantities beautifully

# **Relaxation time**

- Start with soft spheres at arbitrary pressure
- Use ACW to calculate effective hard sphere diameter  $\sigma_{\text{eff}}$
- Obtain new packing fraction for effective hard spheres  $\phi_{\text{eff}}$

# Relaxation time

- Start with soft spheres at arbitrary pressure
- + Use ACW to calculate effective hard sphere diameter  $\sigma_{\text{eff}}$
- Obtain new packing fraction for effective hard spheres  $\phi_{\text{eff}}$



ACW approx rocks!! τ collapses onto hard-sphere curve for all finite-ranged repulsive potentials and pressures studied

#### Recall Hard-Sphere Curve



#### Universal Hard-Sphere Master Curve



- Given
  - hard-sphere master curve
  - pair interaction potential of soft-sphere system
- can calculate relaxation time of soft-sphere system!
- Can also use soft spheres to extend master curve

## Two Mechanisms of Relaxation in Soft Spheres

• Temperature opens up free volume against the pressure

$$T = p\Delta V$$

- hard spheres are fragile glassformers (super-Arrhenius increase of relaxation time)
- Temperature allows soft spheres to overlap so they behave as hard spheres with smaller diameter (less super-Arrhenius with increasing overlap)
- In energy landscape, canyons do not become deeper but become narrower and more convoluted as p  $\uparrow$  or T  $\downarrow$

## Connection to Point J

- Point J corresponds to double limit T/p $\sigma^3 \rightarrow 0$ ,  $p\sigma^3/\epsilon \rightarrow 0$
- What is

$$\lim_{T/p\sigma^3\to 0} \lim_{p\sigma^3/\varepsilon\to 0} \tau \sqrt{\frac{p\sigma}{m}} = \lim_{T/p\sigma^3\to 0} \lim_{p\sigma^3/\varepsilon\to 0} g\left(\frac{T}{p\sigma^3}, \frac{p\sigma^3}{\varepsilon}\right) = \lim_{T/p\sigma^3\to 0} f\left(\frac{T}{p\sigma^3}\right)$$

- Does  $\tau$  diverge at Point J? Or does it diverge at T/p $\sigma^3$  > 0?
- Does Pt J control glass transition? Or is there an underlying thermodynamic glass transition?



# Fitting Forms

- Vogel-Fulcher form:  $\tau/\tau_0 = \exp\left(\frac{A}{T-T_0}\right)$
- Elmatad-Chandler-Garrahan form:



## Form of Scaling Function



• Hard spheres are very fragile!

- Can't distinguish between V-F form and E-C-G form
- Can't tell if there is a thermodynamic glass transition or not
- But if not, then Point J controls dynamical glass transition

# Conclusions

- Point J is a special point
- Hint of connection to glass transition in exponents for jamming transition
- Similarity in form of slow down in dynamics due to equivalence of
  - hard sphere glass transition



- Hard spheres tell us everything about soft spheres
- Point J controls dynamical glass transition of hard spheres if thermodynamic glass transition does not exist
- Still ahead: attractions



## Thanks to

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J. M. Schwarz	Syracuse
Lincoln Chayes	UCLA

Vincenzo Vitelli Leiden Matthieu Wyart Princeton Anton Souslov UPenn Tom Lubensky UPenn Lynn Daniels UPenn Doug Durian UPenn Zexin Zhang UPenn Ke Chen UPenn Arjun Yodh UPenn Randy Kamien UPenn

Bread for Jam:

DOE DE-FG02-03ER46087 German Academic Exchange Service (DAAD) Effective diameter



- As  $p\sigma^{3}/\epsilon$  increases,  $\sigma_{eff}$  decreases
- As T/p $\sigma^3$  increases,  $\sigma_{eff}$  decreases

 $V(r) = \begin{cases} \frac{\varepsilon}{\alpha} \left( 1 - \frac{r}{\sigma} \right)^{\alpha} & r \le \sigma \\ 0 & r > \sigma \end{cases}$ 

#### Jamming vs. Glass Transition



# BUT: Real Liquids have Attractions

Point J only exists for repulsive, finite-range potentials

 $(T_x, \phi_x)$ 

 Attractions can lead to vapor-liquid transition which preempts Point J

#### But there is hope:

Attractions serve to hold system at high enough density that repulsions come into play (WCA) Behavior of liquids is controlled by repulsions, and attractions are perturbation



#### Similar Behavior in Jammed State



- Behavior of amorphous solids is
  - very different from that of crystals
  - same in all amorphous solids
  - still not understood

# Marginally Jammed Solid

L. E. Silbert, A. J. Liu, S. R. Nagel, PRL 95, 098301 ('05) Density of Vibrational Modes



- Density of states is not Debye-like at low  $\omega$
- Result of isostaticity M. Wyart, S.R. Nagel, T.A. Witten, EPL 72, 486 (05)
- Scaling of ω\* is robust to systems near isostaticity Souslov, Liu, Lubensky, PRL 103, 205503 (2009); Mao, Xu, Lubensky arXiv: 09092616

### **Boson** Peak



- Excess in density of states is tied to peak in heat capacity  $C(T)/T^3$
- This frequency/temperature can be tuned in jammed packings by varying  $\phi$   $\phi_{\rm c}$

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#### Thermal Conductivity





Kubo formulation

$$d_{i} = \frac{\pi}{3\hbar^{2}\omega_{i}^{2}} \sum_{i \neq j} \left| S_{ij} \right|^{2} \delta\left(\omega_{i} - \omega_{j}\right)$$

Kittel's 1949 hypothesis: rise in  $\kappa$  above plateau due to regime of freq-independent diffusivity

#### Ioffe-Regel Crossover



•Crossover from weak to strong scattering at  $\omega^*$ •At Point J, the diffusivity is flat down to  $\omega=0$ •Freq-indep diffusivity originates from Point J

N. Xu, V. Vitelli, M. Wyart, A. J. Liu, S. R. Nagel, PRL **102**, 038001 (2008). V. Vitelli, N. Xu, M. Wyart, A. J. Liu, S. R. Nagel, arXiv:0908.2176

### Consequences for Thermal Conductivity



#### Consequences for Thermal Conductivity



boson peak in C end of plateau in  $\kappa$ 

tied together through  $\omega^*$ 





N. Xu, Vi Vitelli, A. J. Liu, S. R. Nagel, arXiv:0909.3710



N. Xu, Vi Vitelli, A. J. Liu, S. R. Nagel, arXiv:0909.3710



Modes become quasilocalized near Ioffe-Regel crossover

N. Xu, Vi Vitelli, A. J. Liu, S. R. Nagel, arXiv:0909.3710



- Modes become quasilocalized near Ioffe-Regel crossover
- High displacements occur in low-coordination regions

N. Xu, Vi Vitelli, A. J. Liu, S. R. Nagel, arXiv:0909.3710

## Anharmonicity



- The low-frequency quasi-localized modes have the lowest energy barriers to rearrangement
  - two-level systems?
  - STZ's? N. Xu, Vi Vitelli, A. J. Liu, S. R. Nagel, arXiv:0909.3710

- Culling process
  - Occupied sites with fewer than k occupied neighbors become vacant
- Repeat culling process until no more can be removed
- Remaining occupied sites called the k-core



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# Jamming vs k-Core (Bootstrap) Percolation

J. M. Schwarz, A. J. Liu, L. Chayes, EPL **73**, 560 (2006).

- Jammed configs at T=0 are mechanically stable
- For particle to be locally stable, it must have at least d
   +1 overlapping neighbors in d
   dimensions
- Each of its overlapping nbrs must have at least d+1 overlapping nbrs, etc.
- At \$\phi > \$\phi\_c\$ all particles in loadbearing network have at least d+1 neighbors

- Consider lattice with coord. # Z<sub>max</sub> with sites indpendently occupied with probability p
- For site to be part of "k-core", it must be occupied and have at least k=d+1 occupied neighbors
- Each of its occ. nbrs must have at least k occ. nbrs,etc
- Look for percolation of k-core

# Long-Ranged Interactions/Attractions

- Point J only exists for repulsive, finite-range potentials
- Real liquids have attractions

Attractions serve to hold system at high enough density that repulsions come into play (WCA)



- Excess vibrational modes (boson peak) believed responsible for unusual low temp properties of glasses
- These modes derive from the excess modes near Point J



N. Xu, M. Wyart, A. J. Liu, S. R. Nagel, PRL **98**, 175502 (2007).

# Effect of Particle Shape



- Introduce new rotational band but onset of translational band still scales as for spheres
- Ellipsoids controlled by Point J for spheres

#### What happens at $\Sigma$ >0?



Glasses & colloidal glasses shear thin--is there a connection?





Stress is indep of potential for small T, p

$$\lim_{p\sigma^2/\varepsilon\to 0}\frac{\Sigma}{p}=F\left(\frac{T}{p\sigma^2},I\equiv\dot{\gamma}\sqrt{\frac{m}{p}}\right)$$

#### Data Collapse for Rheology



#### Consequences of Rheology Collapse



describes jamming surface: universal near Point J!