Jamming and hard sphere glasses

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Outline

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Introduction

The sphere packing problem

Hard spheres are ubiquitous in condensed matter ($d = 2, 3$)...

- Liquids
- Solids: crystals and glasses
- Colloids
- Granulars
- Powders
- Binary mixtures, alloys...

...and in computer science!

- Digitalization of signals ($d \to \infty$)
- Error correcting codes (spheres on the hypercube)
- Constraint satisfaction problem (CSP)

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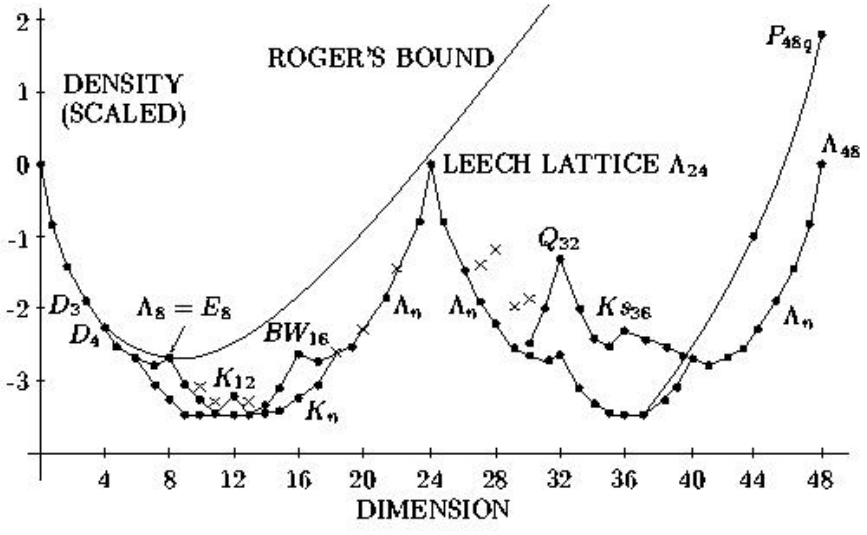
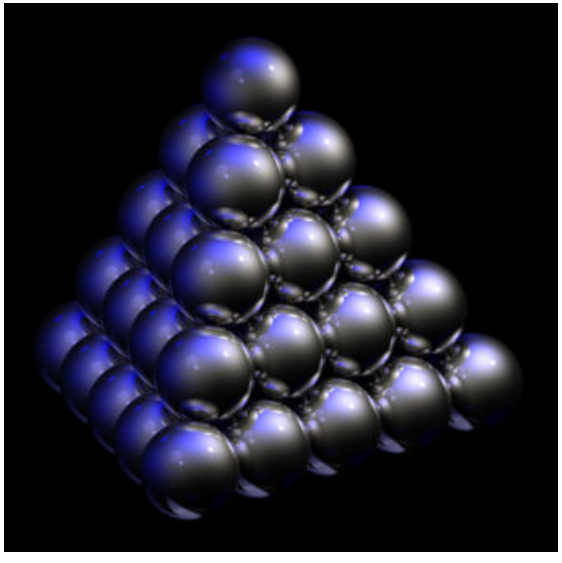
Mathematicians have studied in great detail the problem of finding the densest packing in $\mathbb{R}^d$ (*http://www.research.att.com/∼njas/*)





In $d = 3$ the densest packing is *a simple lattice*...

...but in some dimensions (e.g. $d = 10$) the best known packing is a complicated lattice with a very large fundamental cell!

No exact results for $d \to \infty$ (see G.Parisi, arXiv:0710.0882)

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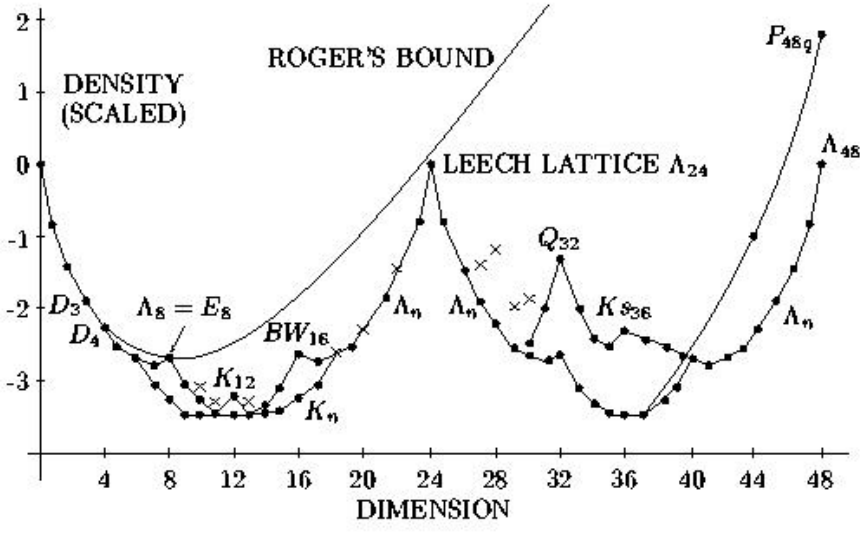

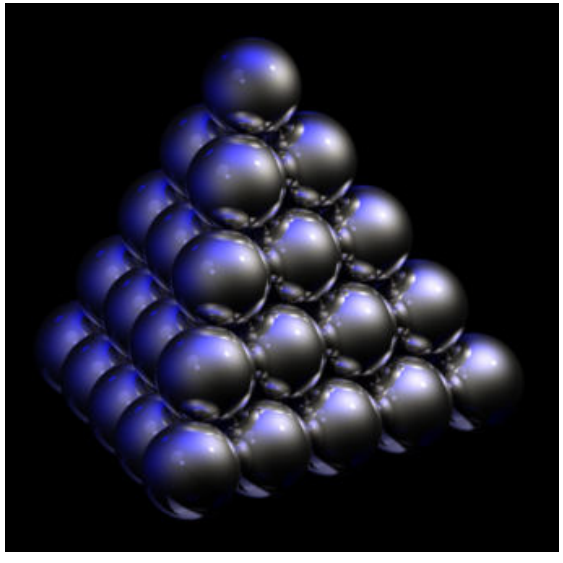

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The random close packing density

Amorphous packings are interesting: in colloids and granulars the crystal is not reached for kinetic reasons (*glass transition*, *jamming*)

Nonequilibrium states: they are prepared using dynamical protocols (algorithms)

- Throw spheres at random in a box and shake/tap the box
- Randomly deposit spheres around a disordered seed cluster
- Inflate the spheres during a molecular dynamics run
- Use a soft potential and inflate the spheres while minimizing the energy

Repeat until *jamming*: particles cannot move anymore

Universality of random close packing?

In $d = 3$ all these procedures give a final density $\varphi \sim 0.64$

Still a small dependence on the protocol is observed

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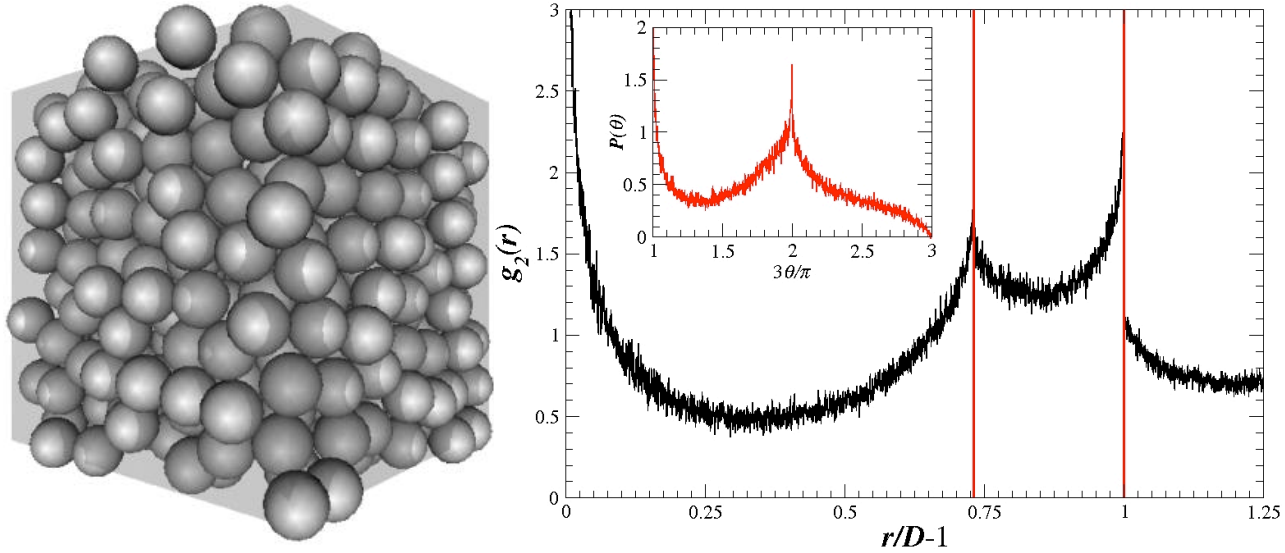


Universal structural properties of amorphous packings:

- Isostatic: $z = 2d$ + anomalous vibrational spectrum (excess of soft modes)
- Square root singularity $g(r) \sim (r-1)^{-0.5}$
- Peak in $r = \sqrt{3}$ and jump in $r = 2$
- Long range correlations, $g(r) - 1 \sim r^{-4}$

Close to $\varphi \sim 0.64$ there is a pocket of amorphous states with similar structural properties.

Introduction

Entropy and surface tension

(At least) three different approaches to the sphere packing problem:

1. "Geometry of phase space": classify all configurations
2. "Equilibrium stat.mech.": look to the flat measure over all configurations and study its structure (equilibrium and metastable states)
3. "Algorithmic": choose a (*non-equilibrium*) dynamical protocol to generate an ensemble of final packings. Study the occurrence frequencies of packings.

The relation between the three is not at all trivial, already in simple mean-field models!

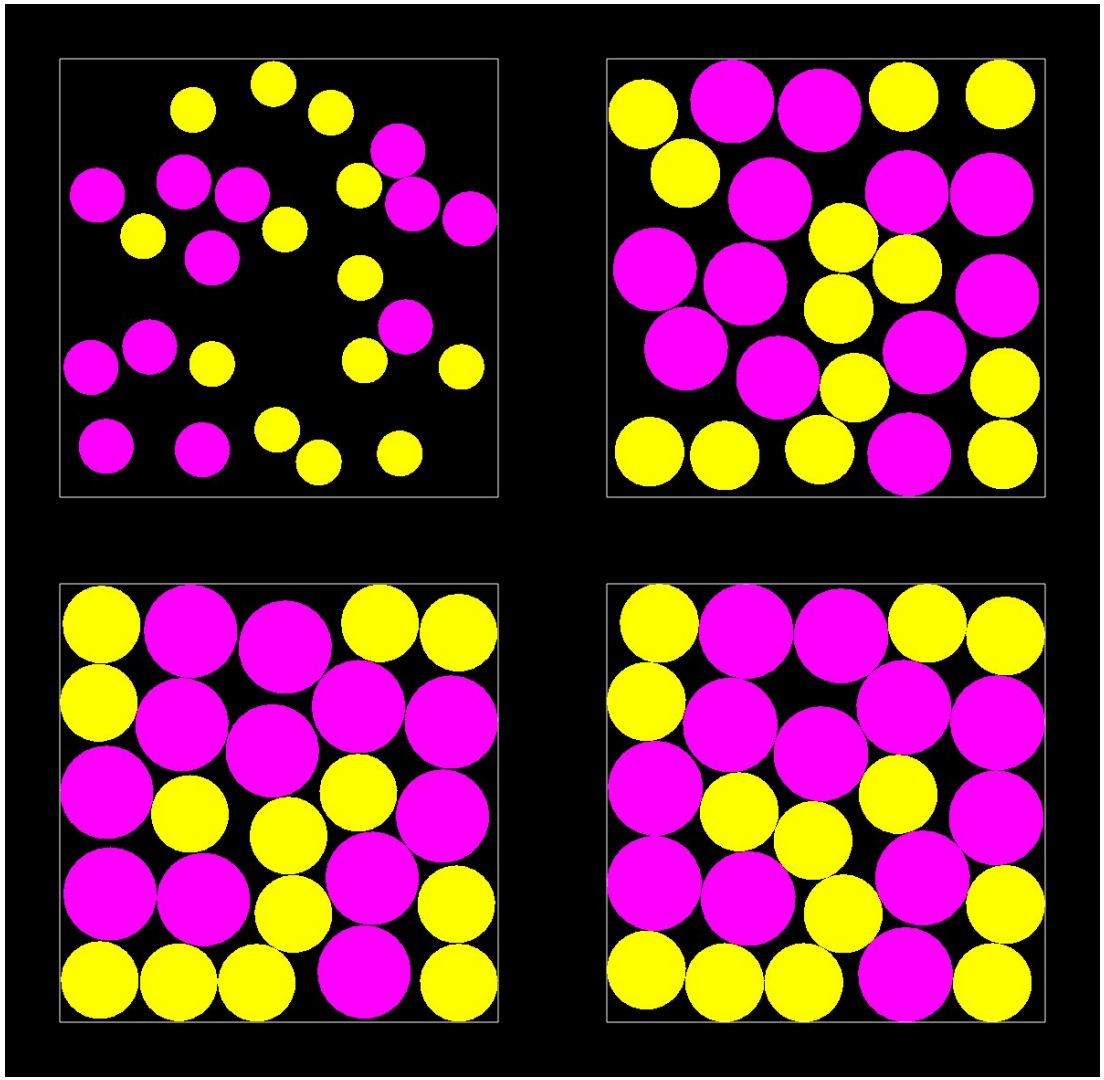
Here we will focus on equilibrium statistical mechanics and identify packings with infinite pressure limit of metastable states:
a central role will be played by entropy and (entropic) surface tension.
$\Rightarrow$ a classical example: first order liquid-crystal transition

The study of surface tension in disordered systems is very difficult and only recently numerical results appeared for glasses
(Cavagna et al. arXiv:0805.4427; arXiv:0903.4264; arXiv:0904.1522).

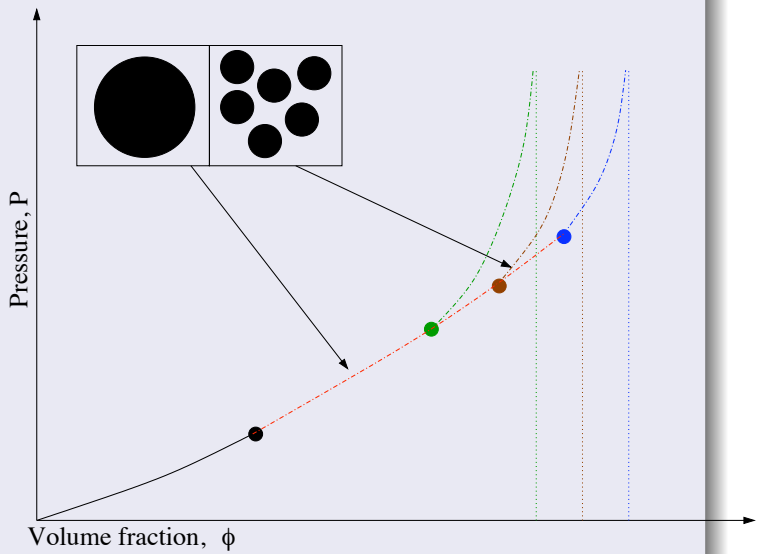
Outline

A simple example: 25 particles in two dimensions

Fast compression starting from a random configuration



"Phase diagram"



Woodcock and Angell, 1981
Stoessel and Wolynes, 1984
Speedy, 1994, 1998
Krzakala and Kurchan, 2007
Parisi and Zamponi, 2008

Questions, and some answers

In the thermodynamic limit many questions arise...

Q There are special configurations (crystals). What is their role? Can one separate them from amorphous configurations?

A This is a very tricky question. Let's discuss it later...

Q Are different configurations really disconnected?

A No, unless $P = \infty$. The self-diffusion coefficient is always finite at finite pressure [Osada, 1998].

Q In what sense they *might* be disconnected?

A1 the time needed to go from one to the other diverges exponentially, $\tau \sim \exp(N)$ [metastability].

A2 If we add an infinitesimal external potential that favors one configuration, the system will stay forever close to that one.

Q Is the jamming density φ_j unique in the thermodynamic limit?

A1 $\varphi_j = \varphi_j^{(\infty)} + O(1/\sqrt{N})$

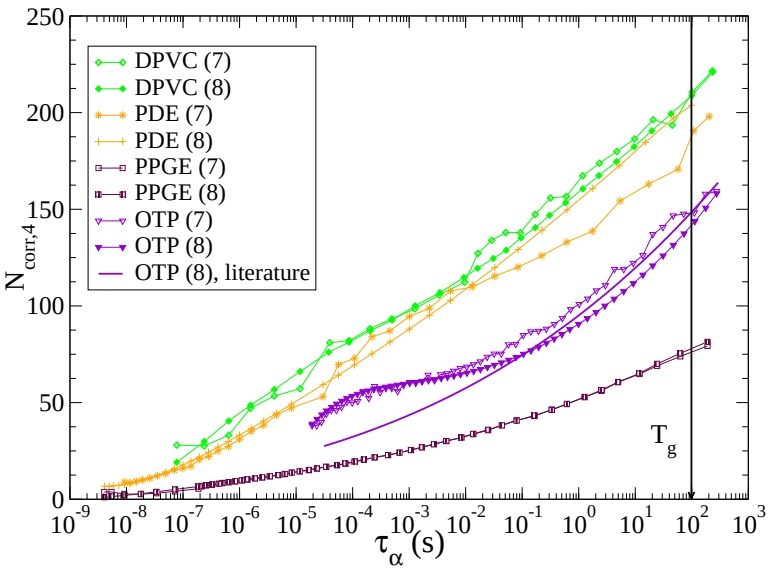
A2 $\varphi_j \in [\varphi_{th}, \varphi_{GCP}]$

1000 particles are interesting

τ_0 microscopic scale (relaxation time at very low density)

Accessible time and length scales in some disordered particle systems

- Numerical simulations: $N \sim 1000 \div 10000$ and $\tau/\tau_0 \sim 10^7$
- Colloids: $D \sim 200$ nm, $L \sim 1$ mm, $N \sim (L/D)^3 = 10^{11}$; $\tau_0 \sim 10^{-3}$ s and $\tau/\tau_0 \sim 10^7$
- Granulars: $D \sim 1$ cm, $L \sim 1$ m, $N \sim 10^6$; $\tau_0 \sim 0.1$ s and $\tau/\tau_0 \sim 10^5$
- Glass forming liquids: $D \sim 1$ A, $N \sim N_A \sim 10^{23}$; $\tau_0 \sim 1$ ps, $\tau/\tau_0 \sim 10^{11}$



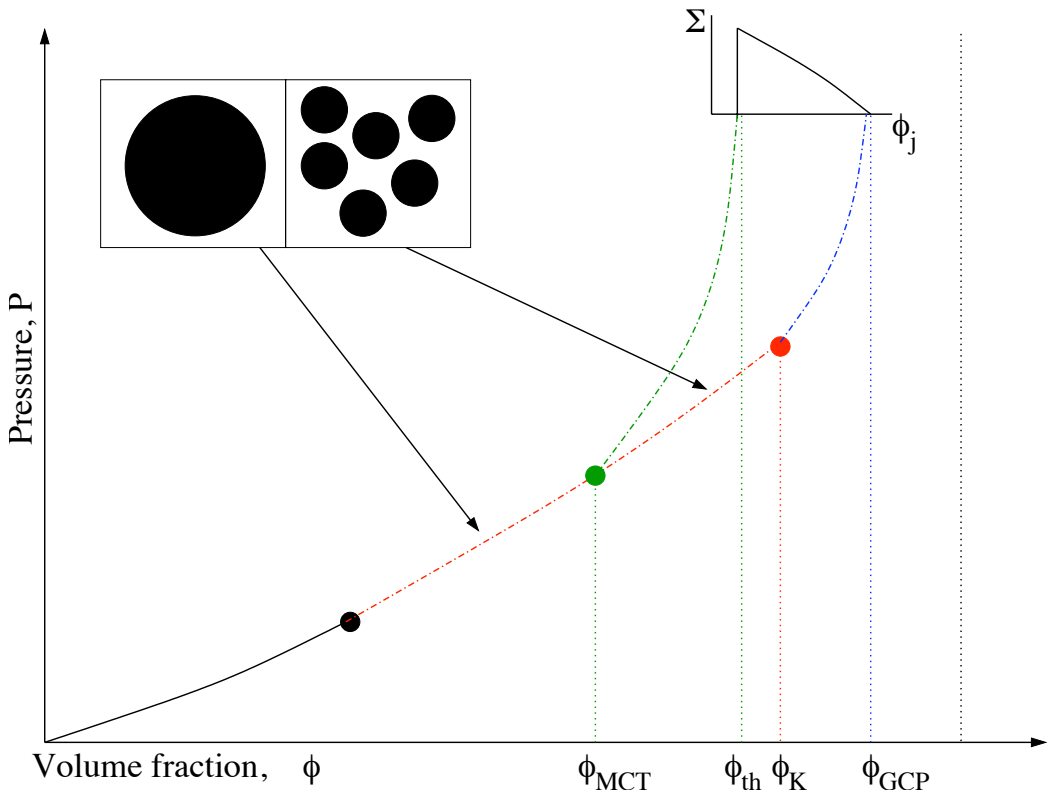
Slow dynamics of disordered systems:
Observing a rearrangement of N_0 particles needs a time $\tau \sim \exp(N_0^\alpha)$
[Glass theory: Adam-Gibbs, RFOT]

Having $N \gg N_0$ does not help much if one cannot wait for a long enough time...
...so $N \sim 1000$ seems already enough to be relevant for colloids and granulars!

On relatively small length and time scales, mean field theory is a good approximation

The mean field theory of glasses: a long story (1980-2009...)

Density Functional Theory: Stoessel and Wolynes, 1984
Mode-Coupling Theory: Bengtzelius, Gotze, Sjolander, 1984
Replicas: Cardenas, Franz, Parisi, 1998 + Parisi and Zamponi, 2005
''Energy'' Landscape: Krzakala and Kurchan, 2007
Random lattice: Mari, Krzakala, Kurchan, 2008



Nonequilibrium glasses
⇕
Metastable states with very large life time

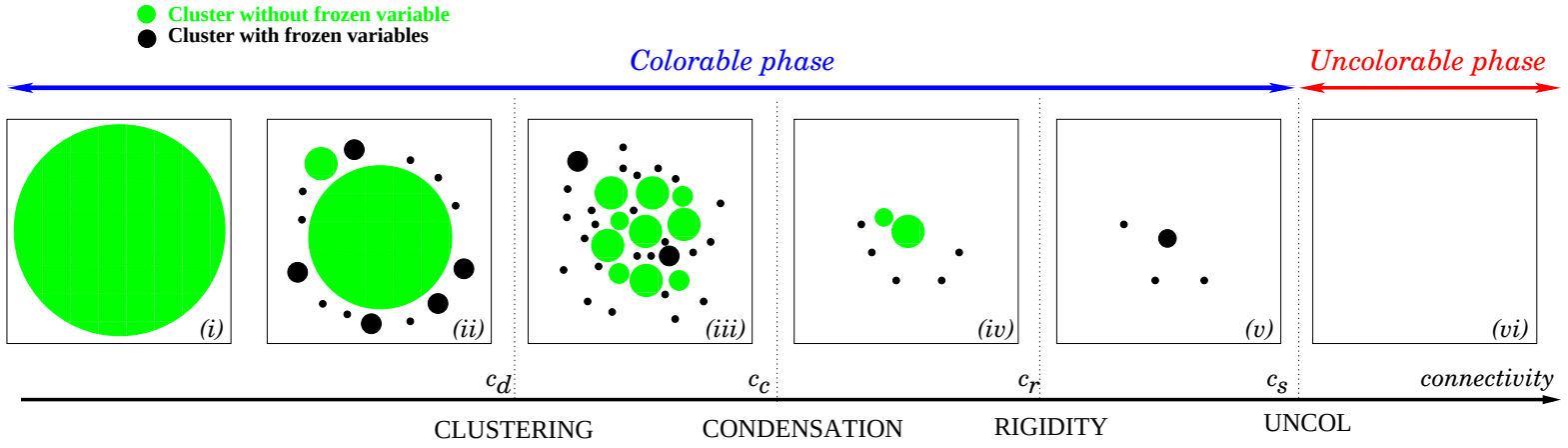
Jammed states
⇕
$P \to \infty$ limit of metastable states

φ_j is a function of φ_{in}
(protocol dependence)

Note: no crystal here, by definition

Phase transitions in Random CSP

Structure of the configuration space:



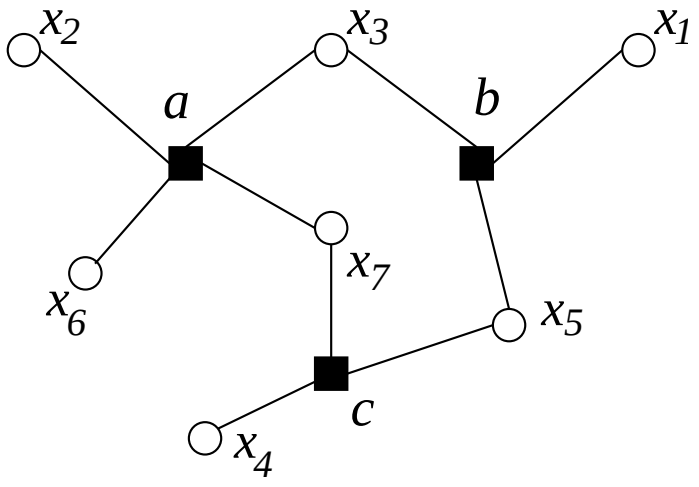
Phase transitions in Random CSP:

- $c < c_d$: Most of the configurations form a unique cluster
- $c_d < c < c_K$: The configurations form many ($\sim e^{N\Sigma}$) clusters
- $c_K < c < c_0$: A small number of clusters dominate
- $c > c_0$: No configurations (UNSAT)

c_d, c_K, c_0: Discontinuous jump of an "order parameter"

A "mean-field" model

Site: $x_i \in [0,1]^d$ PBC
Box: $\prod_{i<j} e^{-\varphi(x_i - x_j)}$



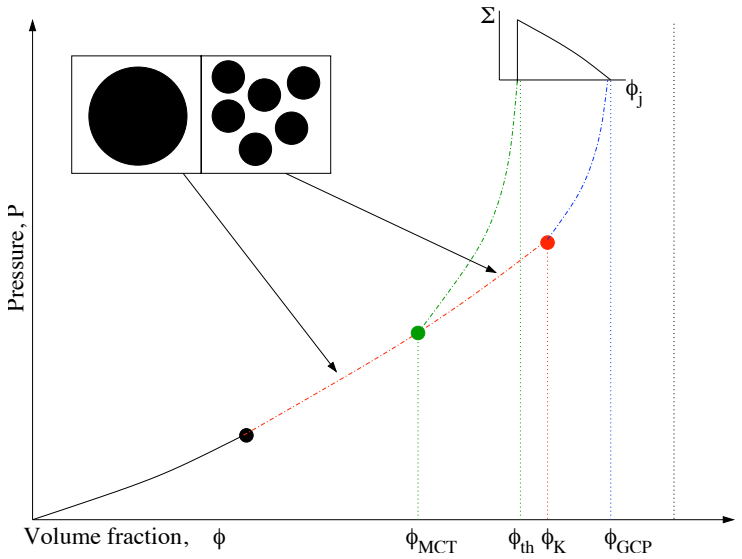
- N variables
- Each variable connected to z boxes
- Each box connected to p variables
- Pick the graph at random with these constraints

Introduced by `Mari, Krzakala and Kurchan (PRL, 2009)`;
Same qualitative phase diagram
Compression at given rate leads to isostatic packings with soft modes etc.

Outline

The replica method

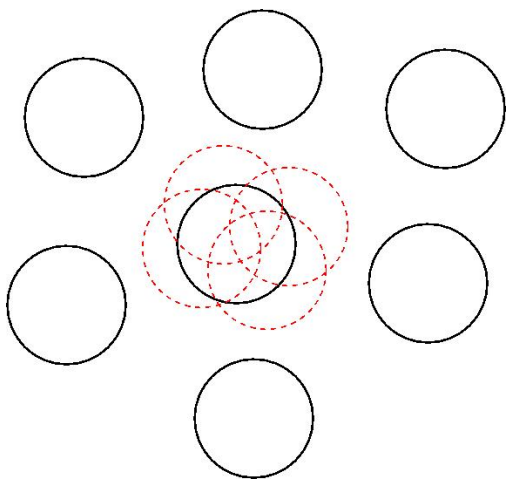
How to compute the distribution of entropies of the states?



$$\mathcal{N}(s,\varphi) = e^{N\Sigma(s,\varphi)}$$

$$Z(\varphi) = \sum_\alpha e^{Ns_\alpha} = \int ds\, e^{N[\Sigma(s,\varphi)+s]}$$

$$Z_m(\varphi) = \sum_\alpha e^{Nms_\alpha} = \int ds\, e^{N[\Sigma(s,\varphi)+ms]}$$



$Z_m(\varphi)$ = partition function of m copies of the system constrained to be all in the same cluster

The replica method

$$Z_m = \sum_\alpha e^{N m s_\alpha} = \int ds\, e^{N[\Sigma(s,\varphi)+ms]} \quad \Rightarrow \quad \mathcal{S}(m,\varphi) = \max_s[\Sigma(s,\varphi)+ms]$$

$$s(m,\varphi) = \frac{\partial\, \mathcal{S}(m,\varphi)}{\partial m}$$
$$\Sigma(m,\varphi) = -m^2 \frac{\partial\,[m^{-1}\mathcal{S}(m,\varphi)]}{\partial m} \quad \Rightarrow \quad \Sigma(s,\varphi) \quad \text{(at constant } \varphi\text{)}$$

To constrain the replicas in the same state we add a coupling:

$$Z_m(\epsilon) = \int_V \frac{d^N x_1 \cdots d^N x_m}{N!}\, e^{-\sum_{i<j}\sum_a v(x_{ai}-x_{aj}) - \frac{\epsilon}{m}\sum_i \sum_{a<b}(x_{ai}-x_{bi})^2}$$

We use standard liquid theory for the replicated liquid:

- Integral equations, HNC, PY... (Franz, Parisi)
- Large coupling (small cage) expansion (Parisi, Mézard)
- Resummations $\rightarrow$ mapping on a Baxter adhesive model

The crystal is automatically excluded by assuming that the replicated system is liquid, i.e. translationally invariant

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- Each replica is a different "chemical" species α: $\rho_\alpha(x)$ its density, $g_{\alpha\beta}(x,y)$ pair correlation. Write $S[\rho_\alpha(x), g_{\alpha\beta}(x,y)]$ using HNC approximation for mixtures.
- Advantage: full access to $g(r)$ and non-ergodicity factor
- Disadvantage: does not work at $m < 1$ and in general when the cage radius is small (e.g. $d \to \infty$).

Large coupling (small cage) expansion (Parisi, Mézard)

- Assume "molecules" composed of one atom per molecule: $(x_1, \cdots, x_m) = \underline{x}$. Write $S[\rho(\underline{x})]$ using virial series. Assume $\rho(\underline{x})$ is a Gaussian of width A. Expand the entropy in powers of $\sqrt{A}$.
- Advantage: very good for small A (large pressure, large dimension, etc.)
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Resummations $\to$ mapping on a Baxter adhesive model

- In the "molecular" framework $S[\rho(\underline{x})]$, integrate over replicas 2 to m. Obtain effective potentials for replica 1. Study the liquid of replica 1 with standard techniques.
- Very good results in $d = \infty$, very poor in $d = 3$ (for the moment)

In the comparison with numerics, all the theoretical results come from a first order small cage expansion

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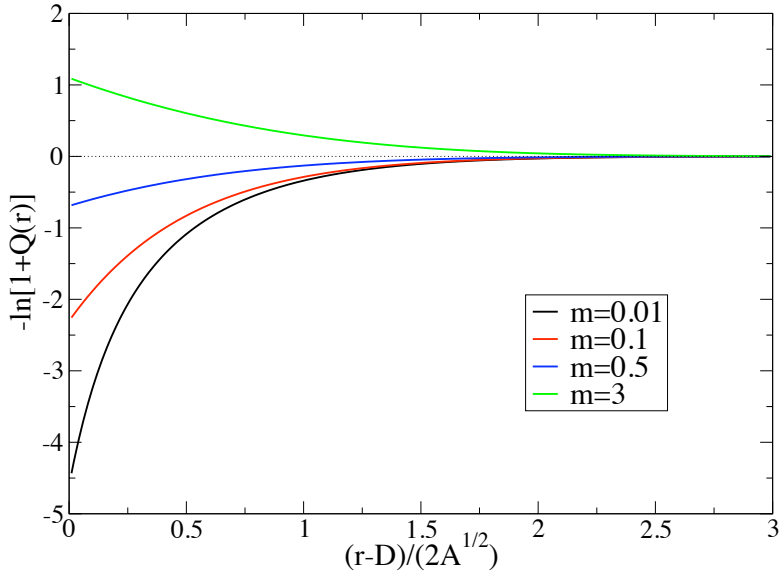
Baxter resummation

It is possible to rewrite the replicated liquid as an atomic liquid with an effective potential $v_{eff}(r) = v_{HS}(r) + \delta v(r)$

$\delta v(r)$ has short range ($\sim$ the amplitude of vibration) and is attractive



This approximation should be very effective for small cage radius

Infinite dimension

- Exact solution: all the phase diagram obtained within the same approximation
- $\varphi_0, \varphi_K \sim 2^{-d} d \log d$, $\varphi_d \sim 2^{-d} d$
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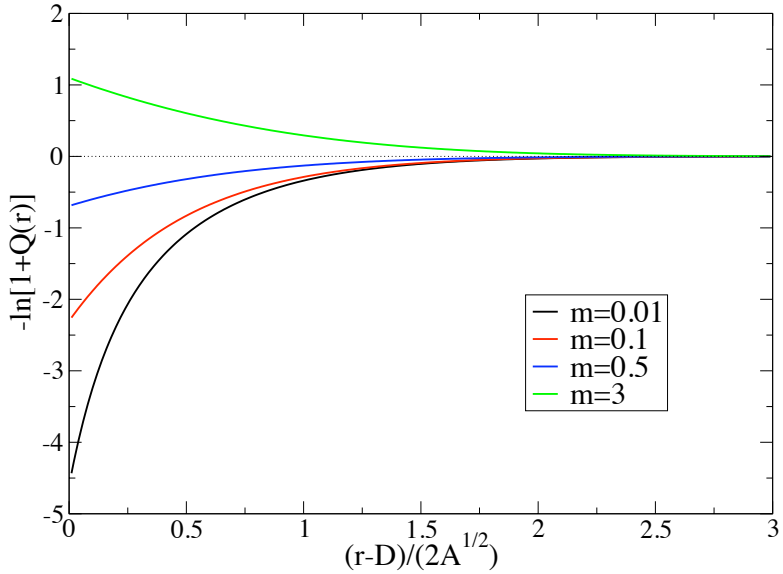

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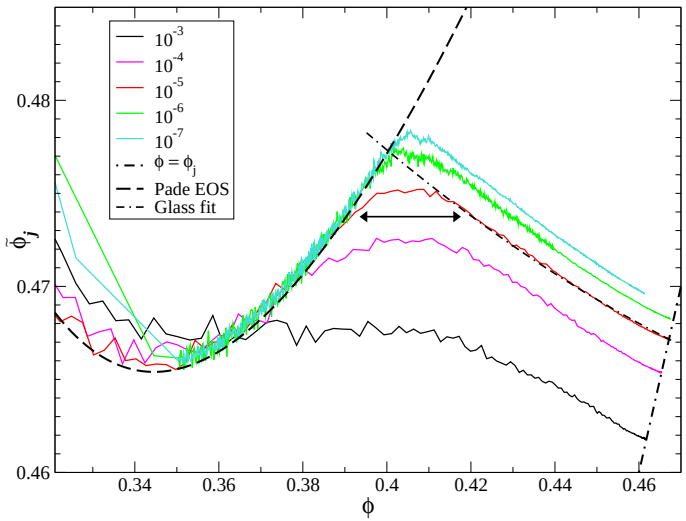
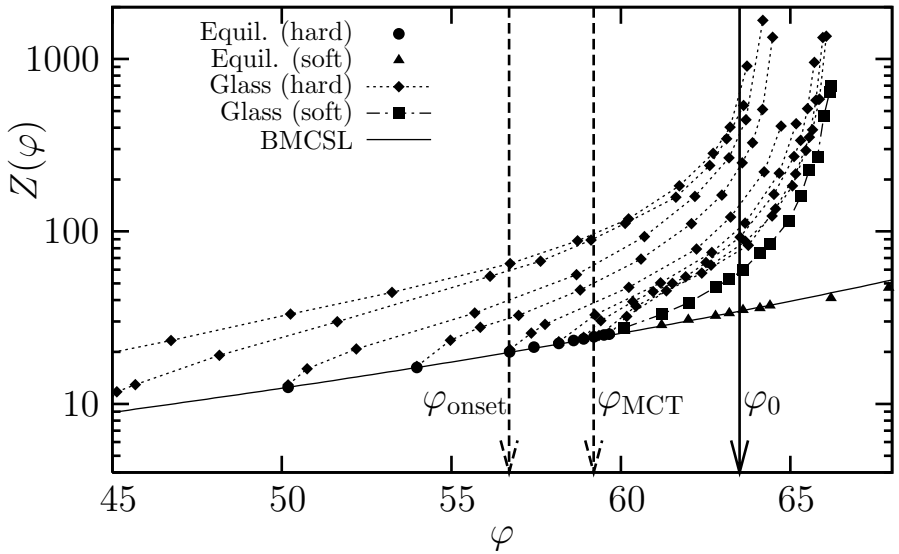
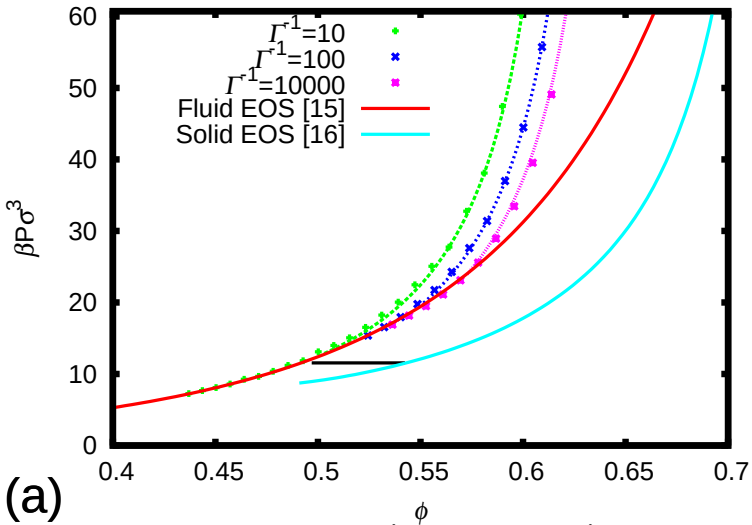
Numerical results: many glasses

Skoge, Donev, Torquato, Stillinger ($d = 4$)

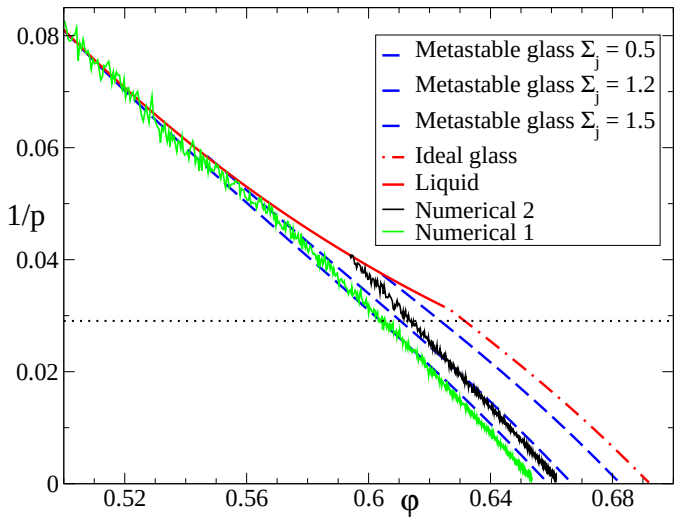


Berthier and Witten (binary $d = 3$)





Hermes and Dijkstra (binary $d = 3$)

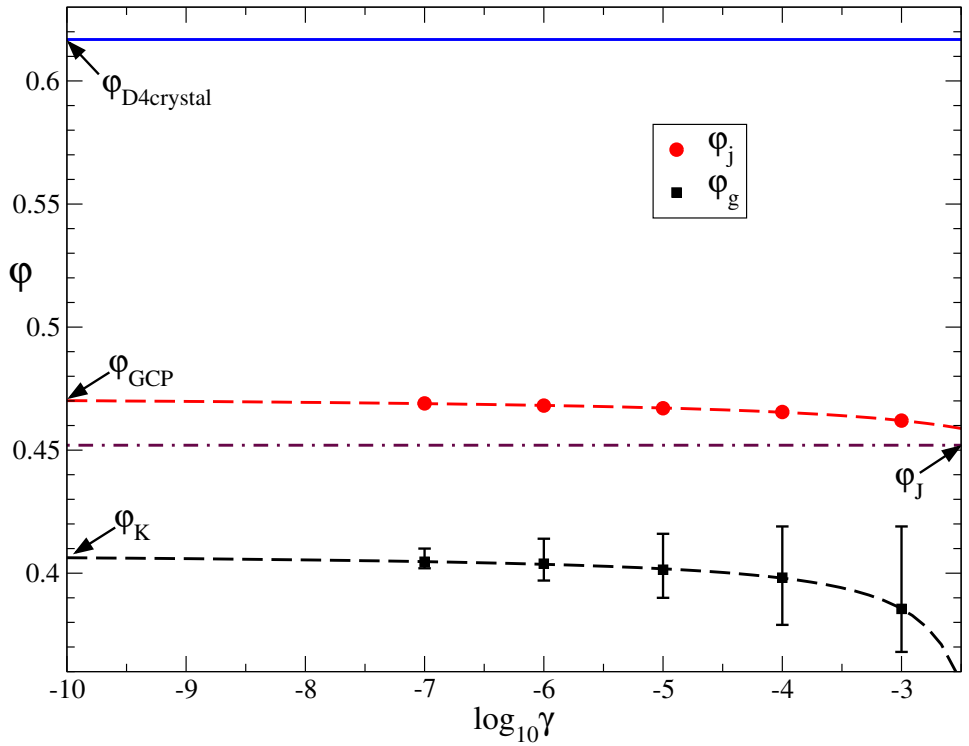


Biazzo, Caltagirone, Parisi, Zamponi (binary $d = 3$)

+ O'Hern, Liu et al. + Pica Ciamarra et al.: All packings have similar structural properties (e.g. isostaticity)

Numerical results: compression rate dependence

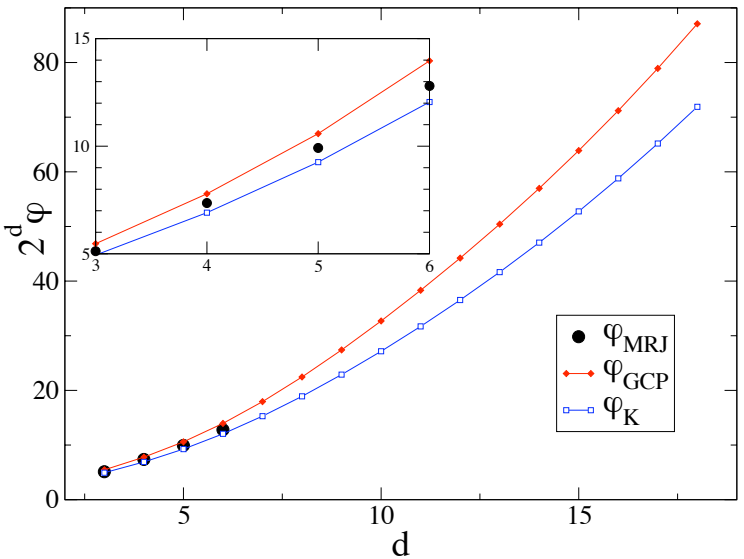
Focus on $d = 4$ where crystallization is very unlikely (Charbonneau et al. 2009)
Data from Skoge et al. (2006); J-point from Schreck and O'Hern, unpublished



Note: extrapolation might be non-sense.
But we are interested in the behavior on intermediate time scales, $\gamma \gtrsim 10^{-7}$.

More precise results on φ_K obtained by Berthier and Witten (2009)

Theory vs numerics: transition densities

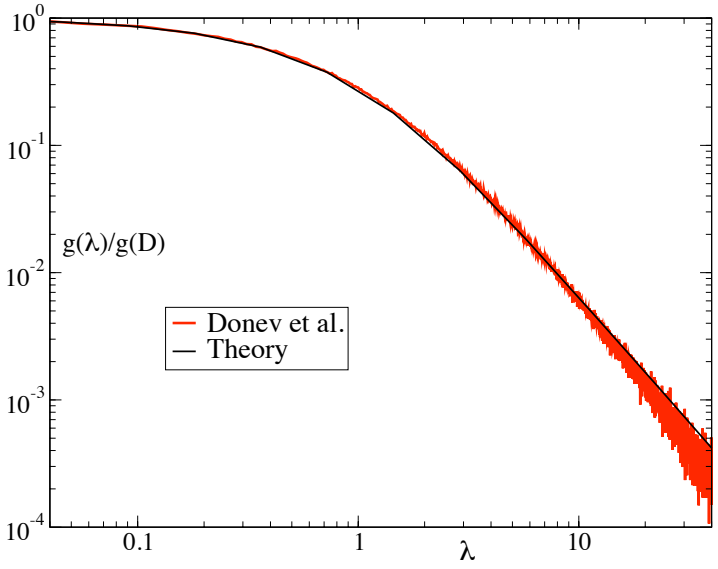


d	φK (theory)	φGCP (theory)	φJ (num.)†	φMRJ (num.)*	φK (extr.)	φGCP (extr.)
2	0.8165	0.8745	—	0.84	—	—
3	0.6175	0.6836	0.640	0.64	—	—
4	0.4319	0.4869	0.452	0.46	0.409	0.473
5	0.2894	0.3307	—	0.31	—	—
6	0.1883	0.2182	—	0.20	—	—
7	0.1194	0.1402	—	—	—	—
8	0.0739	0.0877	—	—	—	—
∞	$2^{-d} d \ln d$	$2^{-d} d \ln d$	—	$2^{-d} d$?	—	—

† O'Hern et al. obtained using energy minimization for soft spheres

* Skoge et al. obtained using the inflating algorithm with high compression rate

Theory vs numerics: scaling functions of $g(r)$



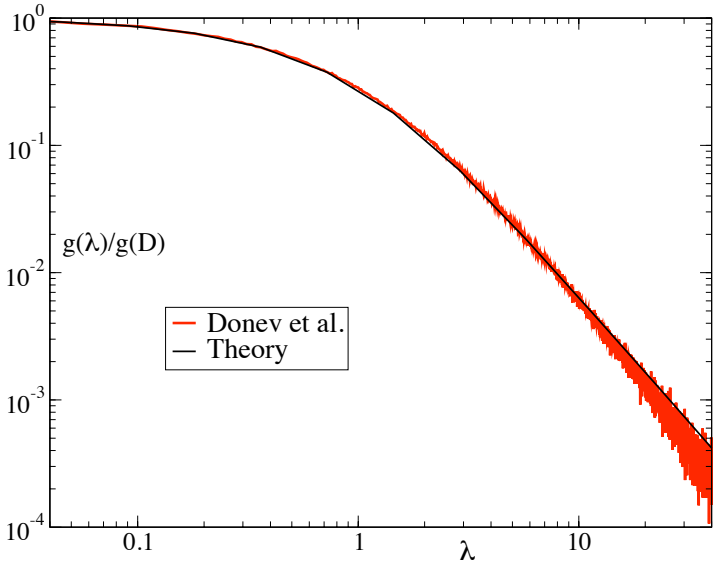
Scaling at infinite pressure: $p \sim g(D) \sim (\varphi_j - \varphi)^{-1}$

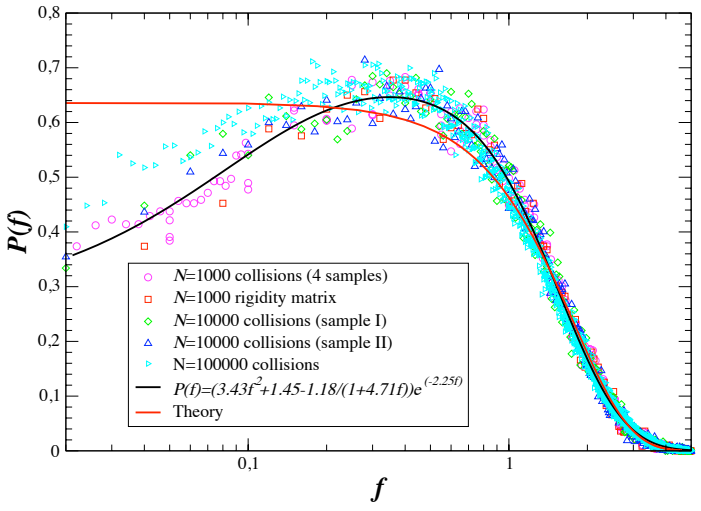
$\frac{g(r)}{g(D)} = \Delta\left(\frac{r-D}{D}p\right) = \Delta(\lambda)$ $z = 2d$: isostatic packings

$\Delta(\lambda) = \int_0^\infty df\ f\ P(f)e^{-\lambda f} \quad \Rightarrow \quad P(f) = \frac{2}{\pi}e^{-f^2/\pi}$ (where $f = F/\langle F\rangle$)

Obtained via the first-order small cage expansion
($g(r)$ *far from contact cannot be computed*)

Theory vs numerics: scaling functions of $g(r)$





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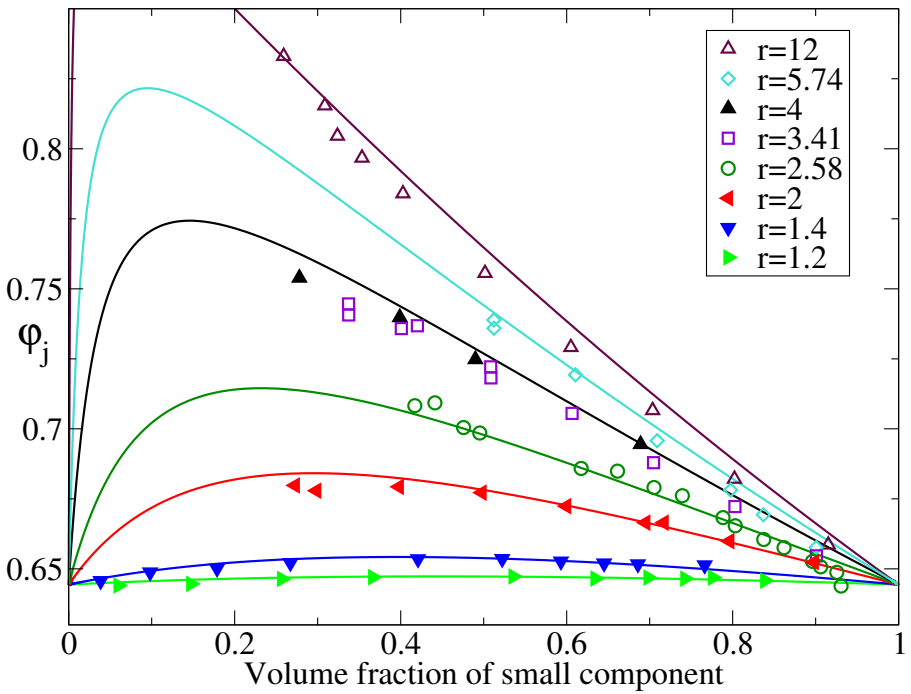
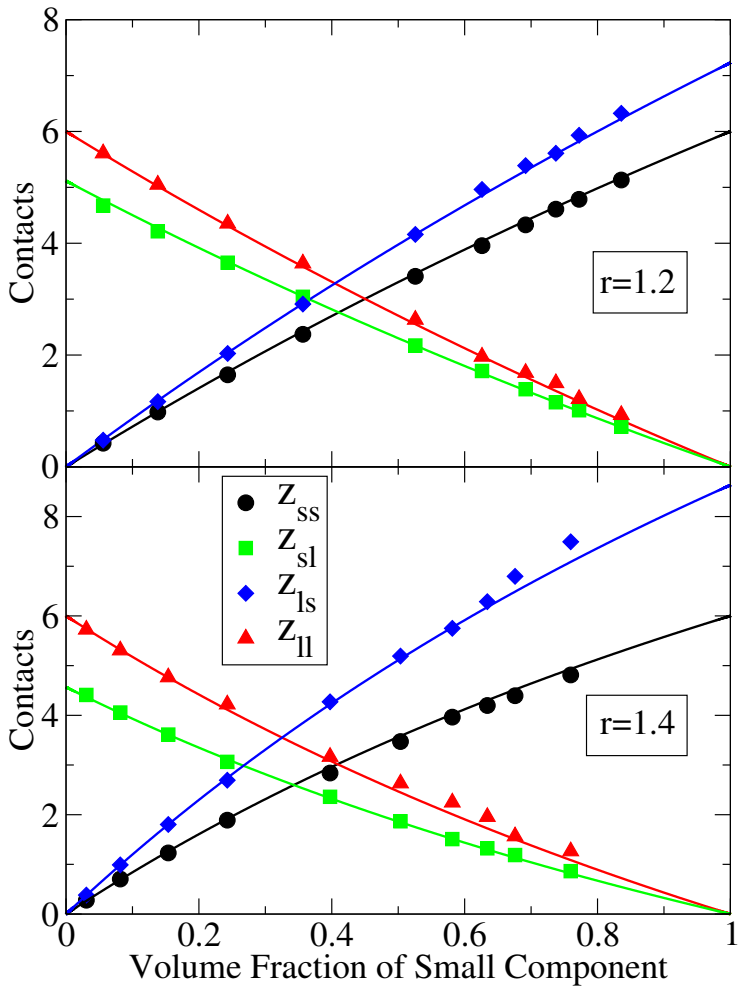
Theory vs numerics: packing geometry

Focus on binary mixture: jamming density and interparticle contacts
(Biazzo, Caltagirone, Parisi, Zamponi, PRL 2009)





$r = D_A/D_B$

All packings are predicted to be globally isostatic.
Partial contact numbers are almost independent of φ_j.

Outline

Our main assumption

Amorphous packing created by complicated nonequilibrium dynamical processes (tapping, shaking, inflating, ...) are metastable glassy states at infinite pressure

Then using replica theory and numerics one can show that:

Mean field *really* holds at moderate length/time scales $N \sim 1000$, $\tau/\tau_0 \sim 10^7$; these scales are explored in numerical simulations, in granulars, and in colloids:

- φ_K "exists" (*in the sense that the relaxation time behaves as if it existed*)
- There are amorphous packings in a finite range of densities around "random close packing": $\varphi_j \in [\varphi_{th}, \varphi_{GCP}]$ with common structural properties
- $\varphi_j = \varphi_j^\infty + O(1/\sqrt{N})$ *for a given protocol*
- the equation of state of the glass is similar to what is obtained during slow compressions
- structural quantities [$S(q)$, non-ergodic factor, coordination, etc.] can be measured and compared with analytic [replica] computations, with good agreement
- Theory predicts that amorphous packings are isostatic, $z = 2d$
- We find a consistent solution in $d \to \infty$ that gives non-trivial predictions for the scaling of the random close packing density, $\varphi \sim 2^{-d} d \log d$

...with some big open problems:

- Better understanding of metastability:
  - Number of states $\mathcal{N} \sim \exp(N\Sigma)$, and Σ vanishes at φ_K: to be confirmed [preliminary results by Speedy 1998, Angelani and Foffi 2005]
  - A complete theory of the surface tension in glassy systems is still missing
  - A theory of the glass transition should explain why mean field holds on such scales (Ginzburg criterion)
  - What happens at larger length/time scales? Nucleation arguments, RFOT, point-to-set correlations
- Nonlinear susceptibilities, soft modes, J-point criticality
- How friction modifies all this picture?

...and some technical open problems:

- Better investigation of the Baxter resummation in $d = 3$: hope to describe all the phase diagram consistently within the same approximation
- Study the equilibrium dynamics (Mode-Coupling theory) in $d \to \infty$: same value for φ_d? (nice work in $d = 4$ by Charbonneau et al. 2009)
- Generalize to potentials for colloids: hard spheres + attractive tail. Reentrance of the glass transition line?

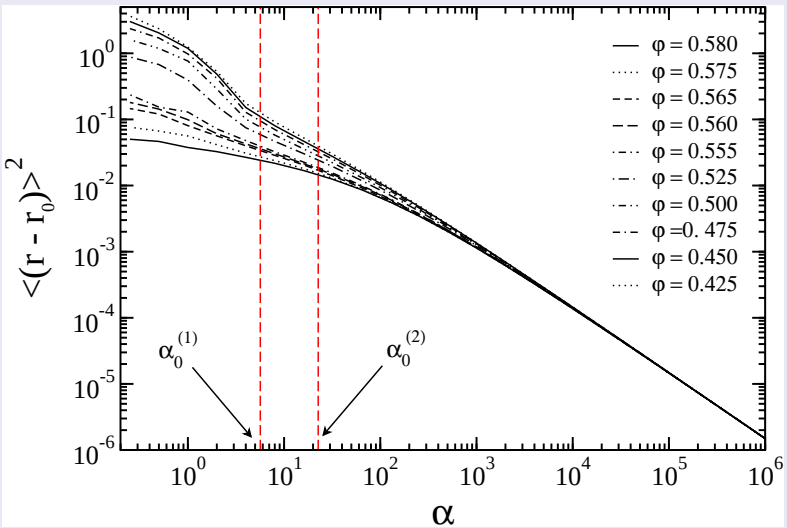
On the definition of complexity

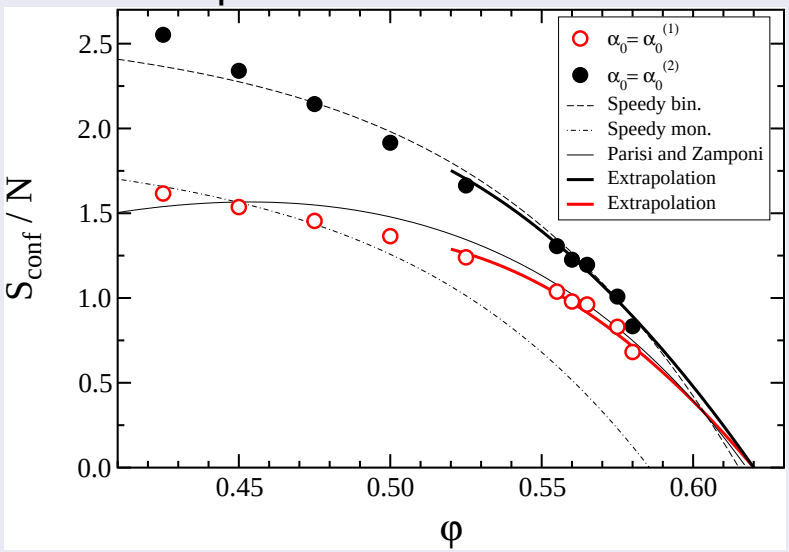
- The definition and numerical computation of the complexity (number of metastable states) is the biggest problem of the mean field scenario.
- Many amorphous states; to select one, we must impose an external potential but we do not know it

Solution 1: couple the system to a reference equilibrium configuration.

[Parisi, Coluzzi, Verrocchio; Angelani, Foffi]

- Self-consistent external potential: $V(r_1, \cdots, r_N) = \alpha \sum_i (r_i - x_i)^2$.
- However, in finite dimension, an infinitesimal potential is enough *only* above φ_K; below φ_K the system escapes from the metastable state at low enough α [via a first order transition]
- Extrapolate to $\alpha = 0$; it is difficult, need for more precise simulations.





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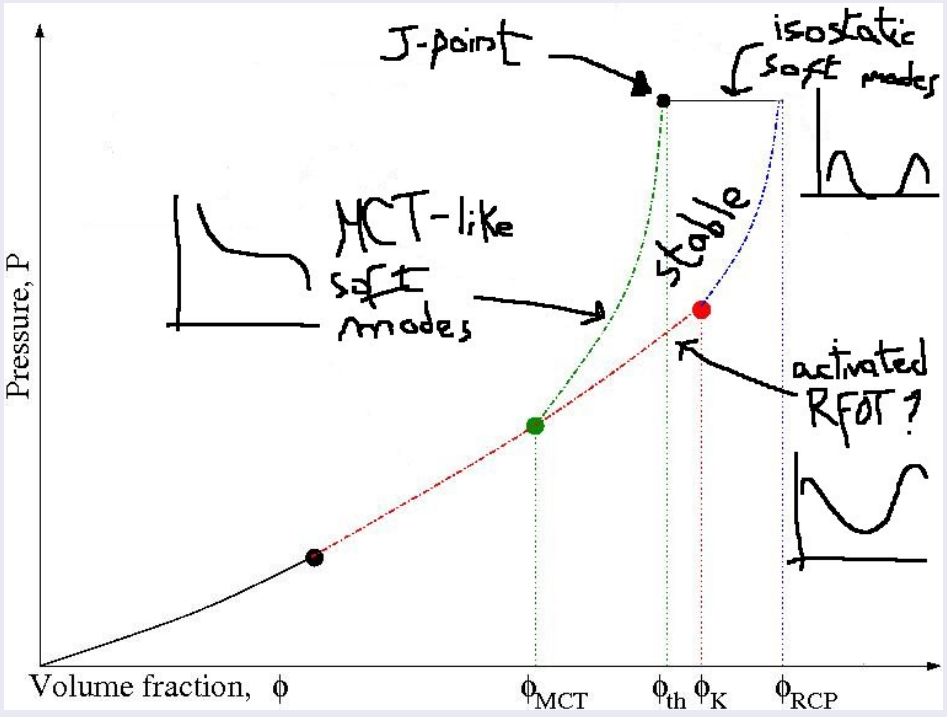
Solution 2: thermodynamics of a bubble.
[Biroli, Bouchaud; Franz; Cavagna, Grigera, Verrocchio

- Consider a bubble of radius R whose boundary is obtained by freezing a larger equilibrium configuration.
- Compute the entropy $s_{int}(R, \varphi)$ of the bubble (via thermodynamic integration)
- Define the complexity as $\Sigma(R, \varphi) = S(\varphi) - s_{int}(R, \varphi)$
- Study the behavior of $\Sigma(R, \varphi)$ as a function of R. It should vanish for $R > \xi$ where ξ is the *point-to-set* correlation length
- Advantage: no need for extrapolation, well defined quantities, direct access to the behavior as a function of the length scale
- Disadvantage: numerically heavy, and in finite dimension, no single state inside the bubble [BBCGV], is this a problem?

Nonlinear susceptibilities and soft modes

A set of jammed states ($= P \to \infty$ limit of metastable states) in a range $[\varphi_{th}, \varphi_{rcp}]$.

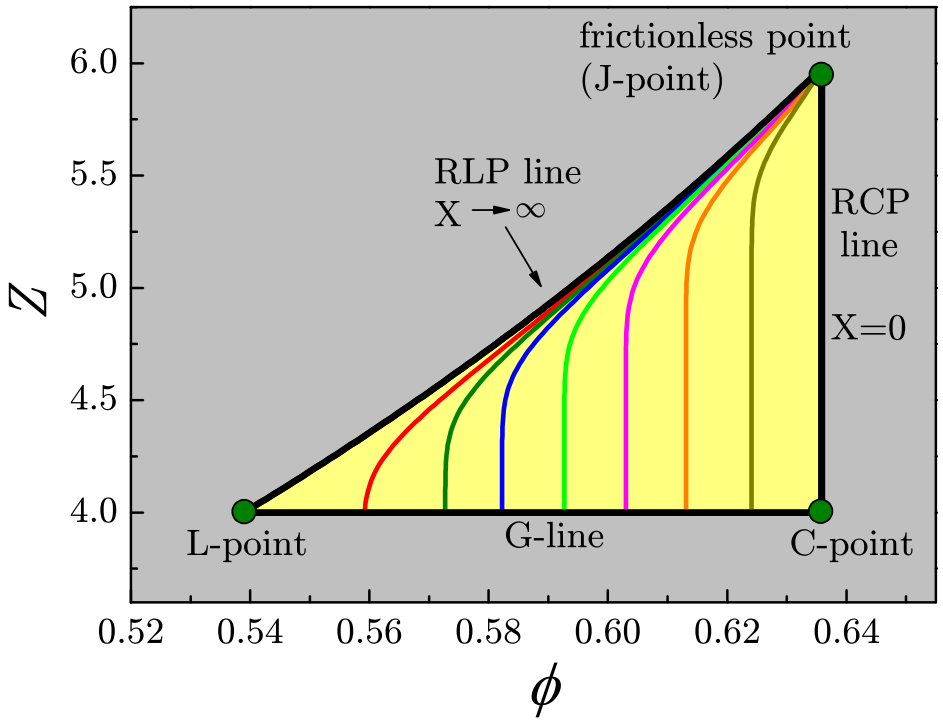


- Do these states have similar structural properties (isostaticity, soft-modes, square-root singularity, hyperuniformity, ...)?
- Is there anything special in the J point (soft both in the "isostatic" and in the "MCT-like" sense) or it just corresponds to a particular procedure?

- Numerical simulations
- MCT in a metastable state
- Soft modes in the replica approach?

Friction complicates the picture

Recent proposals for a schematic phase diagram of jammed matter [Makse et al., van Hecke et al.]



- φ_J is not a point: what consequences on this phase diagram?
- What happens at finite pressure?