Spatial dimensionality dependence of the glass and jamming transitions

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M. Adhikari, S karmakar, S. Sastry, Phys. Rev. Lett. 131 (16), 168202 (2023) [arXiv:2204,02936]

Outline

- Introductory: The glass transition and the jamming transition
- The relationship between the glass and jamming transitions
- The hard sphere glass transition from mean field theory
- Scaling analysis of dynamics in 3D (soft spheres) to estimate ideal glass transition density for hard spheres
- New (our) scaling analysis of dynamics in 3 – 8 dimensions
- Comparison with calculation for finite dimensions
- Summary

Introduction

The glass and jamming transitions: Two routes to the amorphous solid state.

Molecular liquids: Glass transition

Granular matter: Jamming transition

Colloidal suspensions: Both phenomena

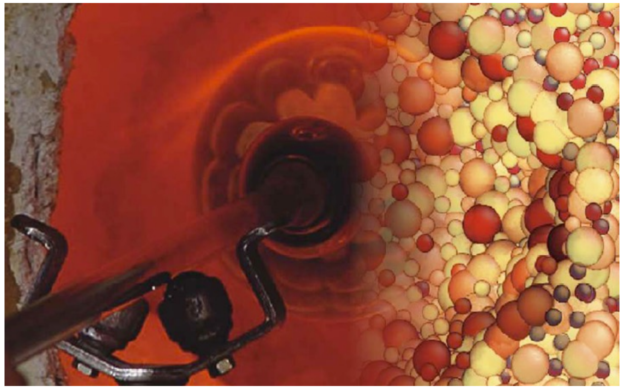
The glass transition

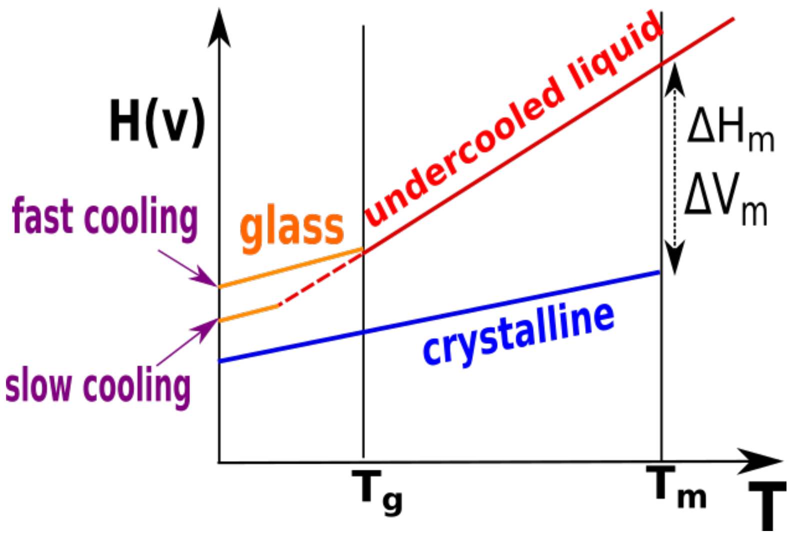
The glass transition is reached (typically) upon cooling a dense liquid.

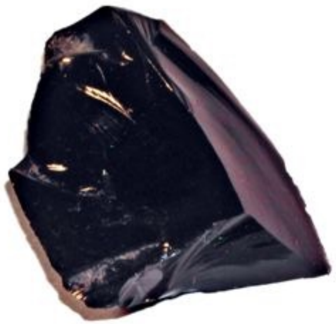
Divergence of relaxation times as the glass transition is approached.

Glass properties depend on the protocol of preparation, such as the cooling rate.

In the limit of infinitely slow cooling – The ideal glass transition/Kauzmann temperature (density).







The jamming transition

The jamming transition observed in assemblies of meso/macroscopic grains.

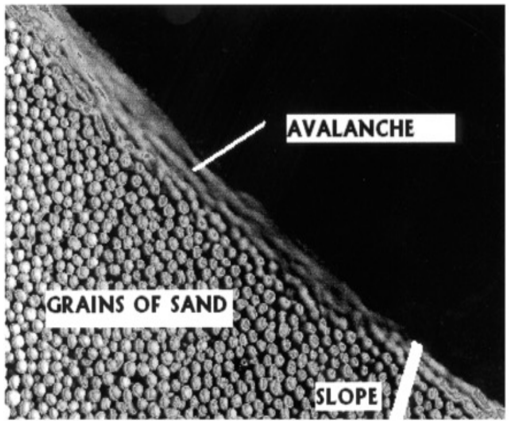
The control parameter is the density.

Rigidity of jammed packings determined by mechanical contacts.

Divergence of pressure for hard particle packings.

The hard sphere system has been employed to investigate both transitions.

Conventionally, sphere packings jam at a packing fraction of ~ 64% -- **Random Close Packing.**

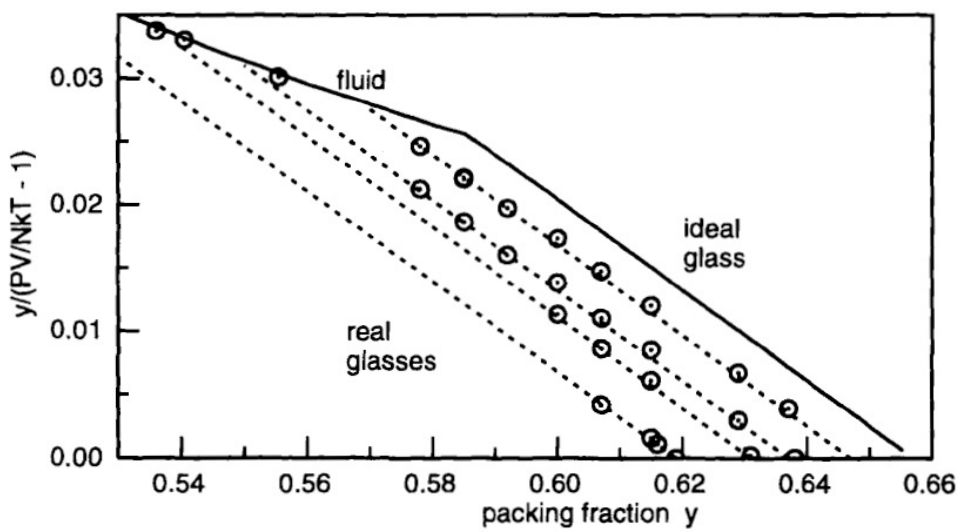


The hard sphere phase diagram

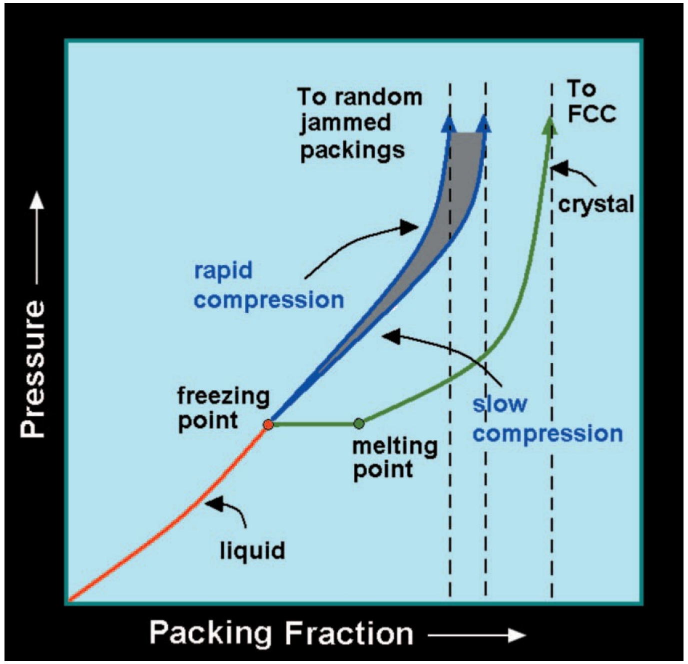
However, several studies discuss the non-uniqueness of the jamming transition.

Speedy – Falling out of equilibrium at different (glass transition) densities lead correspondingly to different jamming densities.

Torquato & co – Random jammed packing ambiguous. Protocol dependence, crystallization etc.



Robin Speedy Mol Phys 1998



From: Torquato Stillinger RMP 2010

Hard sphere jamming

Demonstration of the non-uniqueness of the jamming density – Chaudhuri et al

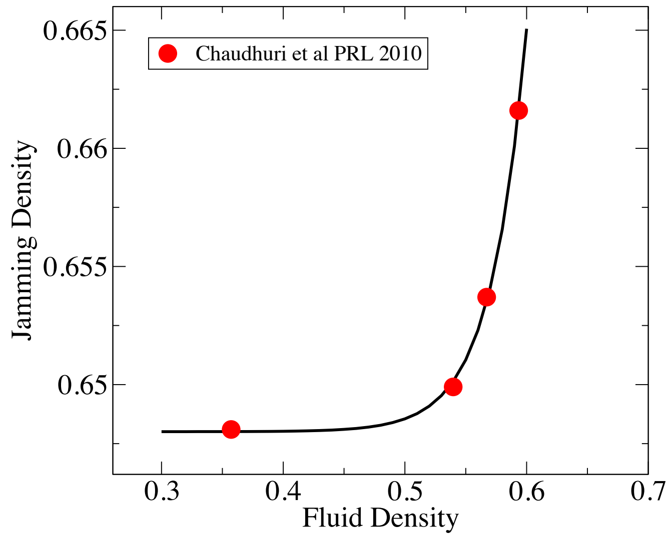
Consider ensembles of hard sphere fluid configurations at different starting densities.

Subject to the ***same*** jamming protocol.

Leads to an initial condition dependent jamming density.

The jamming line.

Random Close Packing/J point: The **lowest** jamming density.



Chaudhuri, Berthier, Sastry PRL 2010

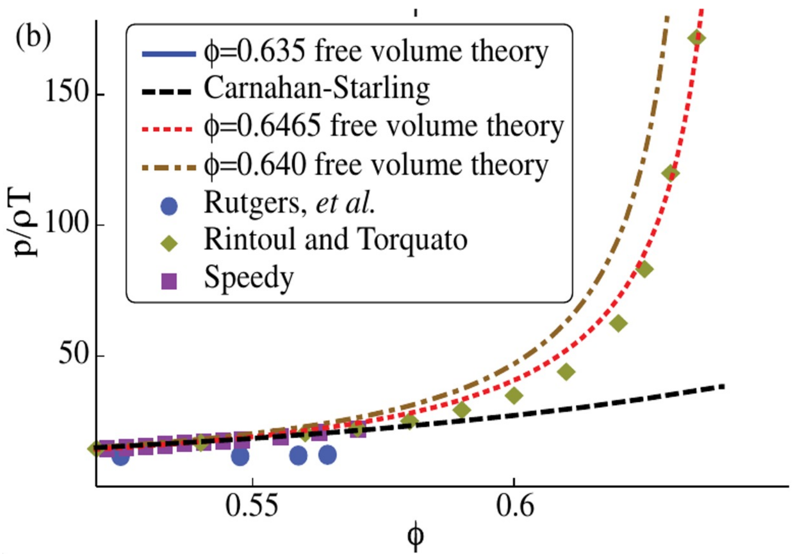
Glass transition and Jamming point

Not everybody agrees(d) – Based on cell/free volume theories, the finite dimensional expectation is that the Kauzmann density = (highest) Jamming density ~ 64.4%

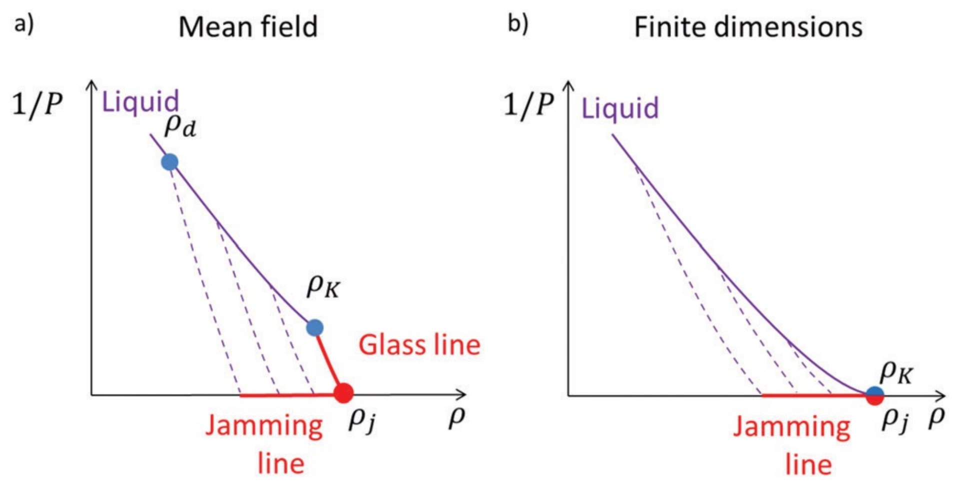
Not consistent with the mean field theory of the glass transition…

Why is Random Close Packing Reproducible?

Randall D. Kamien and Andrea J. Liu

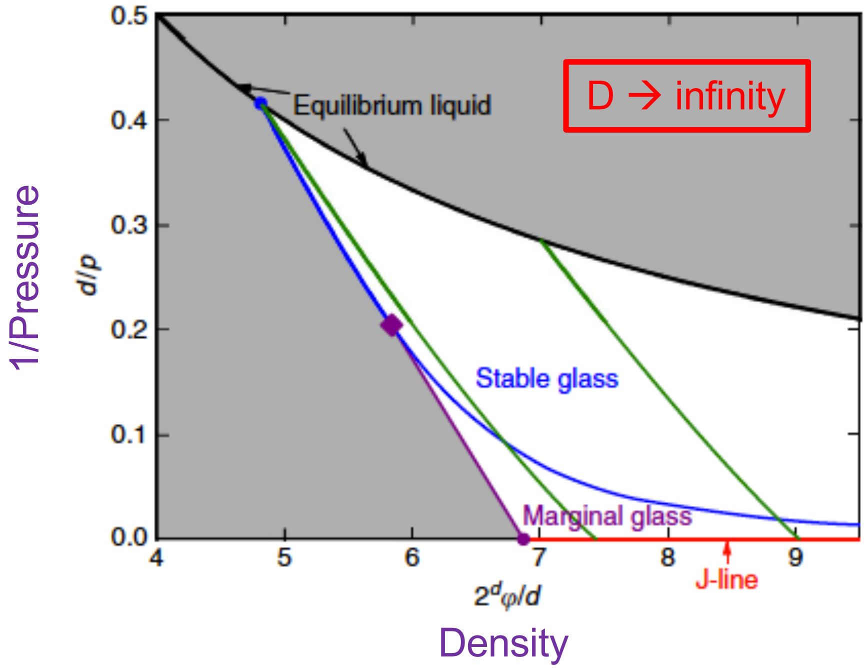


Kamien and Liu PRL, 2007

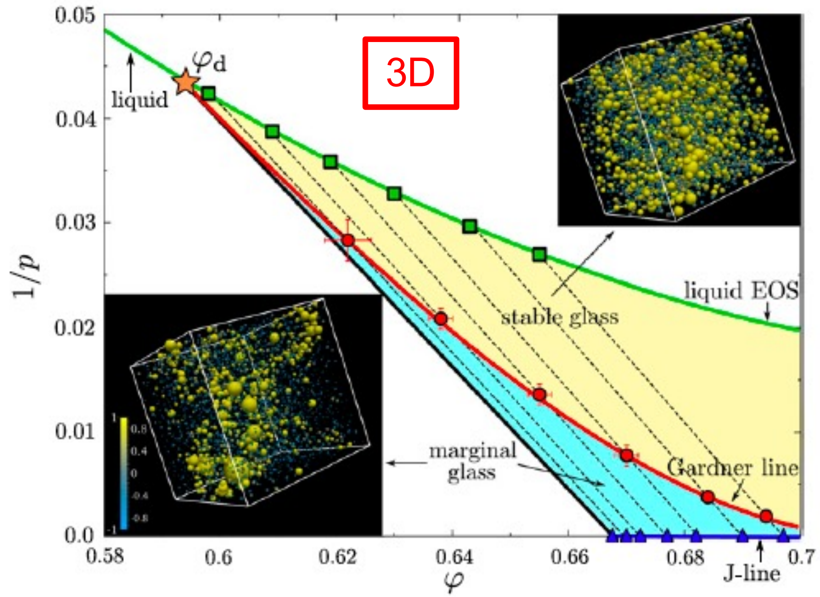


Coniglio et. al Soft matter (2017)

Infinite dimensional theory



Charbonneau et. al. Annual Rev. of Cond Matt Phys (2017)



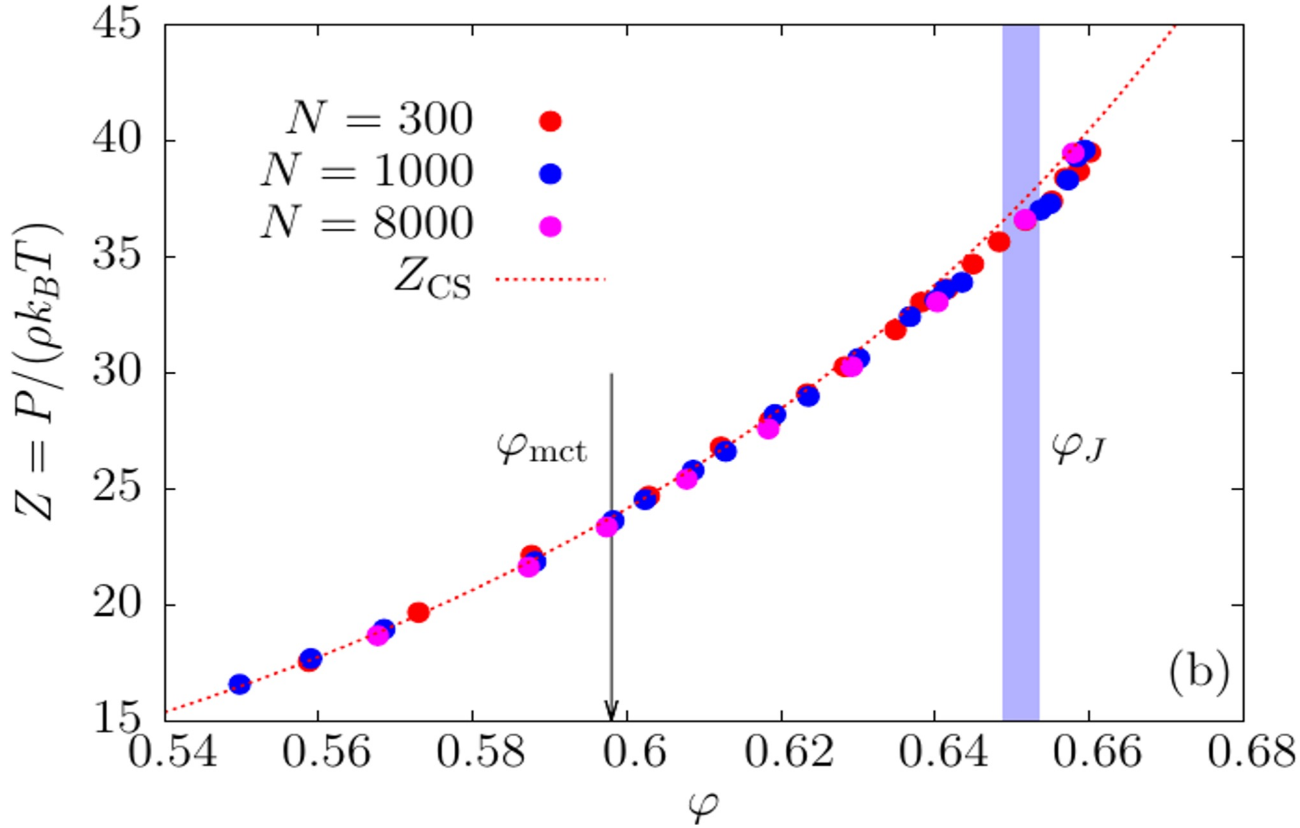
Berthier et al PNAS 2016

The infinite dimensional theory of the glass transition predicts the presence of a line of glass transitions and a corresponding line of jamming points – the J line.

Supporting evidence that this picture holds for D = 3.

But no precise identification of the jamming and Kauzmann densities.

3D Simulations



Using swap MC, pressure can be computed well above φ_J.

No divergence of pressure at $\varphi_{J.}$

Ideal glass transition and jamming densities (RCP) not related.

But no identification of the Kauzmann density in this work..

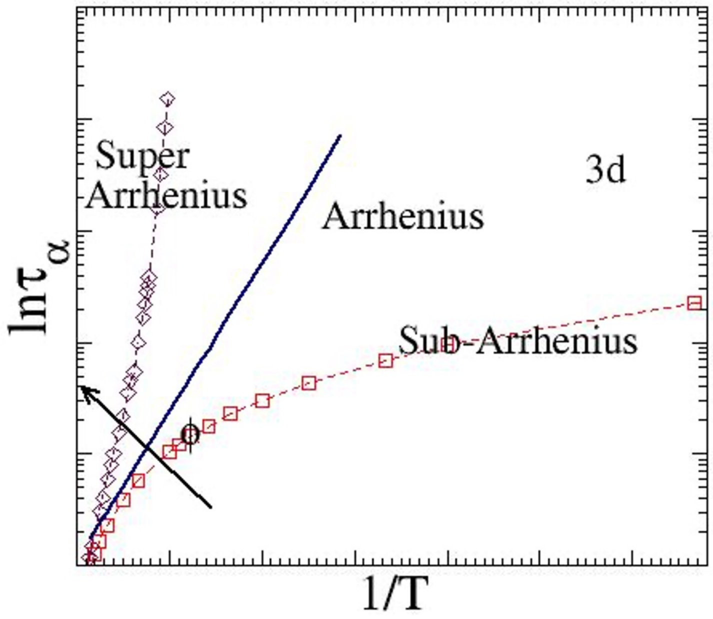
Berthier et al, PRL 2016

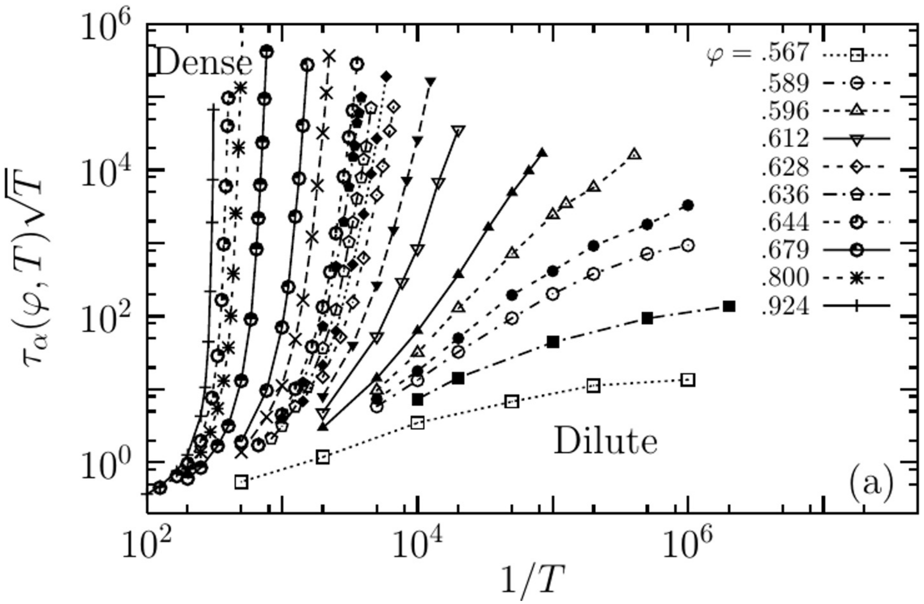
Soft Sphere Simulations: 3D

From simulation of soft sphere mixtures across densities, one observes a crossover from sub-Arrhenius to super-Arrhenius temperature dependence.

Arrhenius relaxation: $\tau_\alpha = \exp(A/k_B T)$

Sub- Arrhenius: τ_α increases slower than Arrhenius.
Super-Arrhenius: τ_α increases faster than Arrhenius.





Low densities: relaxation behavior sub-Arrhenius, High densities: Super-Arrhenius.

Berthier and Witten, EPL (2009)

Scaling analysis of dynamics

Scaling analysis of dynamics near the crossover between sub-Arrhenius to super Arrhenius to identify the ideal glass transition density (Berthier-Witten) in 3D.

Scaling form obtained based on two considerations:

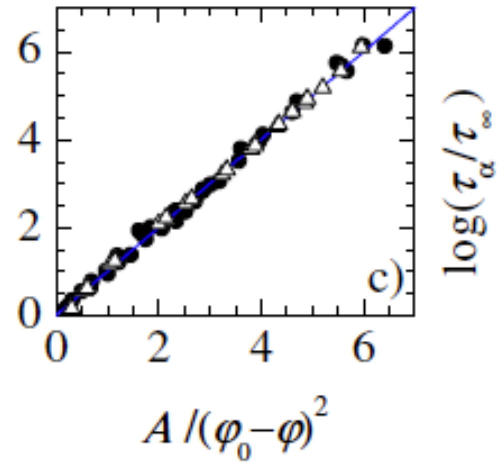
- Soft spheres at finite temperature treated as having effective diameters:

$$\phi_{eff} = \phi - aT^{\mu/2}$$

- The relaxation times for hard spheres obeys

$$\tau_\alpha(\phi) = \exp\left(\frac{A}{|\phi_0 - \phi|^\delta}\right)$$

with $\delta = 2$.



(Brambilla et al PRL 2009)

Berthier Witten (BW) scaling formula

$$\tau_\alpha(\phi, T) \sim \exp\left[\left(\frac{A}{|\phi_0 - \phi|^\delta}\right) F_\pm\left(\frac{|\phi_0 - \phi|^{\frac{2}{\mu}}}{T}\right)\right]$$

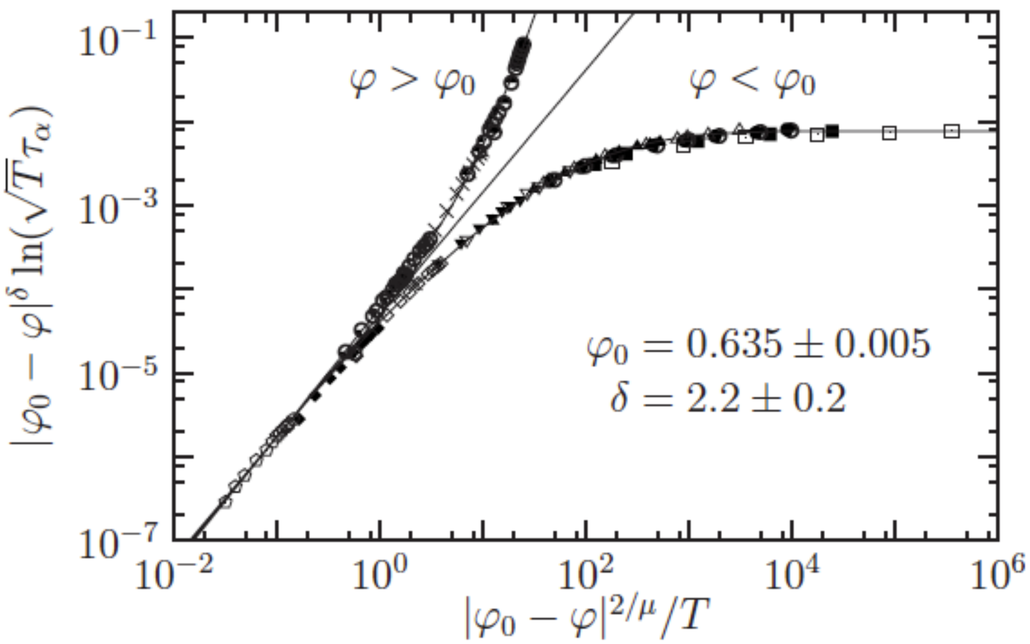
$$F_+(x \to 0) \sim F_-(x \to 0) \sim x^{\mu\delta/2}$$

Requires $\mu\delta/2 = 1$

For Arrhenius behavior at φ_0

Berthier-Witten scaling

For suitable choices of the parameters (φ_0 , μ, δ) all the data falls on a master curves.



Three parameters: φ_0,μ, δ

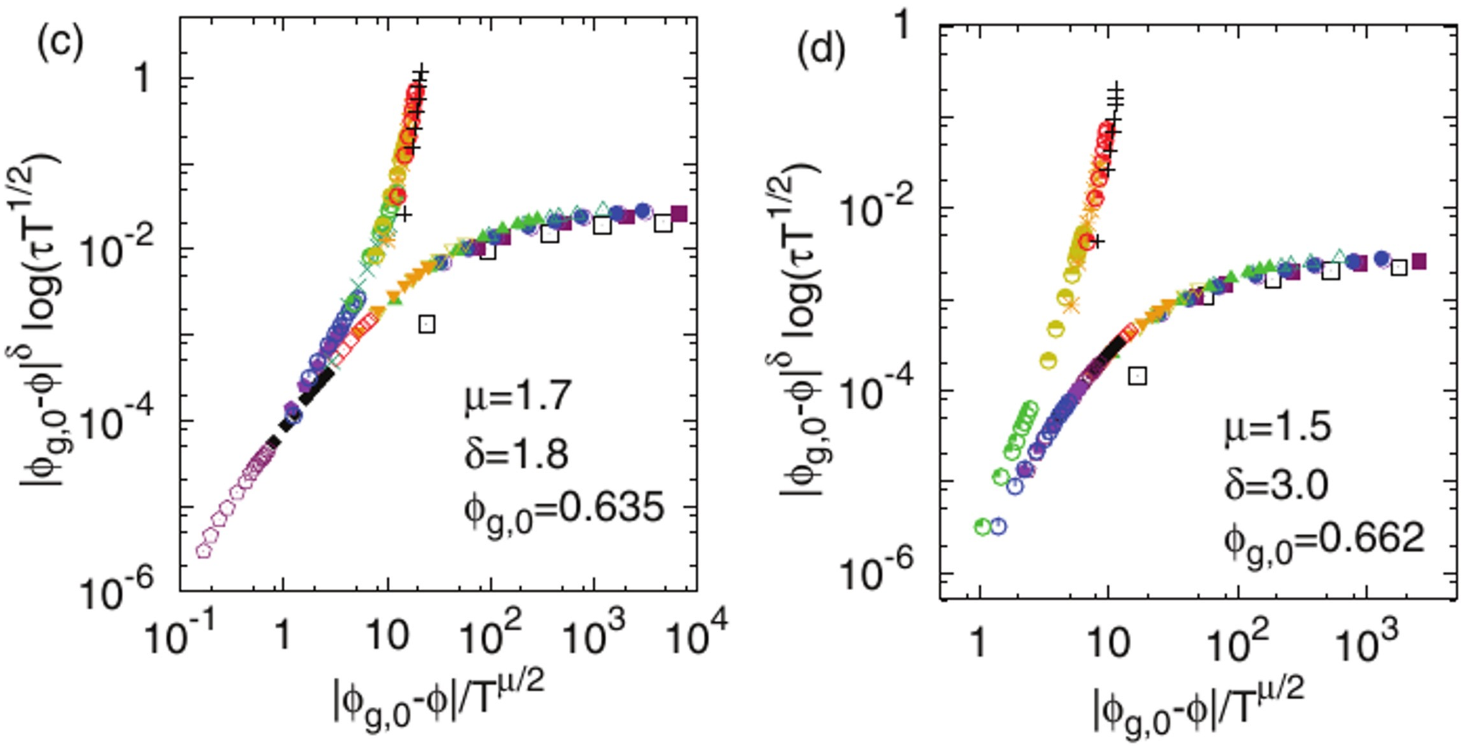
μ = 1.3
δ = 2.2 (not 2)
φ_0 = 0.635

φ_0 is very close to φ_J (d=3)

Berthier and Witten, PRE (2009)

Limitations

The same scaling analysis was revisited by Maiti and Schmiedeberg:



Equally good scaling collapse — completely different set of parameters.

Parameters used to obtain scaling collapse do not lead to an Arrhenius temperature dependence at $\varphi = \varphi_0$

How do you better estimate of φ_0?

Maiti and Schmiedeberg, J Phy: Cond. Matt (2019)

Our Simulation Study

Questions:

How do the estimates of ϕ_0 and ϕ_J depend on spatial dimensionality?

Can these densities be determined by a more robust procedure?

Do the results more convincingly distinguish ϕ_0 and ϕ_J ?

Can we compare our results satisfactorily with attempts to extend the infinite dimensional results to finite dimensions?

Our simulation investigation:

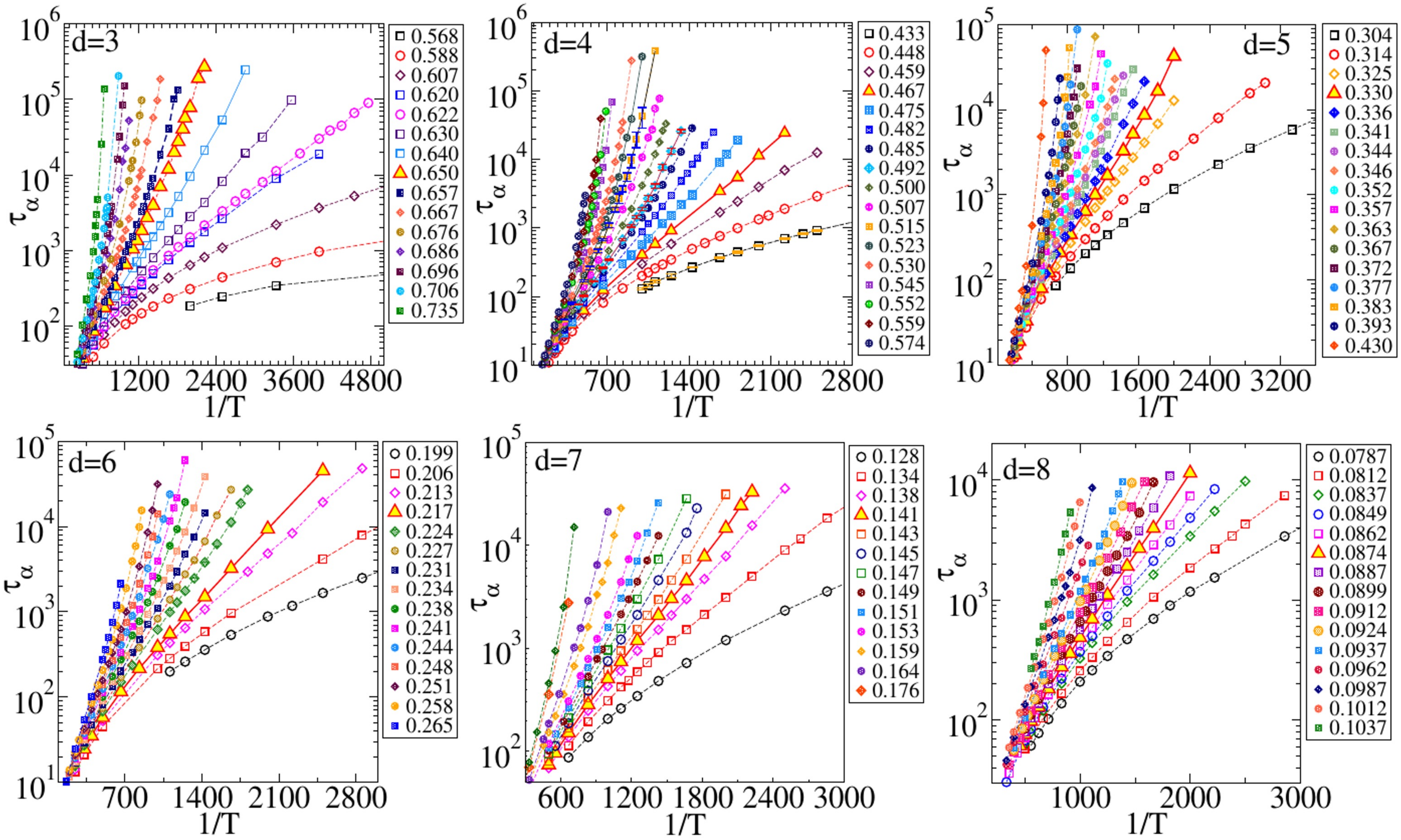
We study binary soft sphere mixtures for 3 – 8 dimensions.

Our model:

$$
\begin{aligned}
V_{\alpha\beta}(r) &= \frac{\epsilon_{\alpha\beta}}{2}\left(1-\frac{r}{\sigma_{\alpha\beta}}\right)^2, \qquad && r_{\alpha\beta} \leq \sigma_{\alpha\beta} \\
&= 0, && r_{\alpha\beta} > \sigma_{\alpha\beta}
\end{aligned}
$$

- Bi-disperse mixture: (50:50) $\sigma_{BB} = 1.4\sigma_{AA}$
- System size: 1000-5000
- density: 12-14 density around φ_J in each dimension.
- NVT MD simulations.
- Integration time step, dt = 0.01

Sub Arrhenius to super Arrhenius transition in all D



New Scaling Analysis

- We perform a scaling analysis similar to BW but with a newly proposed scaling function.

- We obtain the effective diameter of the soft spheres following Barker and Henderson.

$$\sigma_{eff} = \int_0^\sigma [1 - \exp(-u(r)/k_B T)]\, dr \qquad \sigma_{\text{eff}} \approx \sigma\left[1 - \tfrac{1}{2}\sqrt{\pi k_B T}\right]$$

φ_{eff} becomes, approximately:

$$\phi_{eff} \approx \phi\left(1 - a\sqrt{T} + b\, T^\beta\right)$$

Our scaling function:

$$\sqrt{T}\tau_\alpha(\phi, T) \sim \exp\left[\frac{A}{|\phi_0 - \phi|^\delta} F_\pm\left(\frac{|\frac{\phi_0}{\phi} - 1|}{a\sqrt{T} - bT^\beta}\right)\right].$$

Requires $\delta = 2$ to obtain Arrhenius behavior at $\phi = \phi_0$

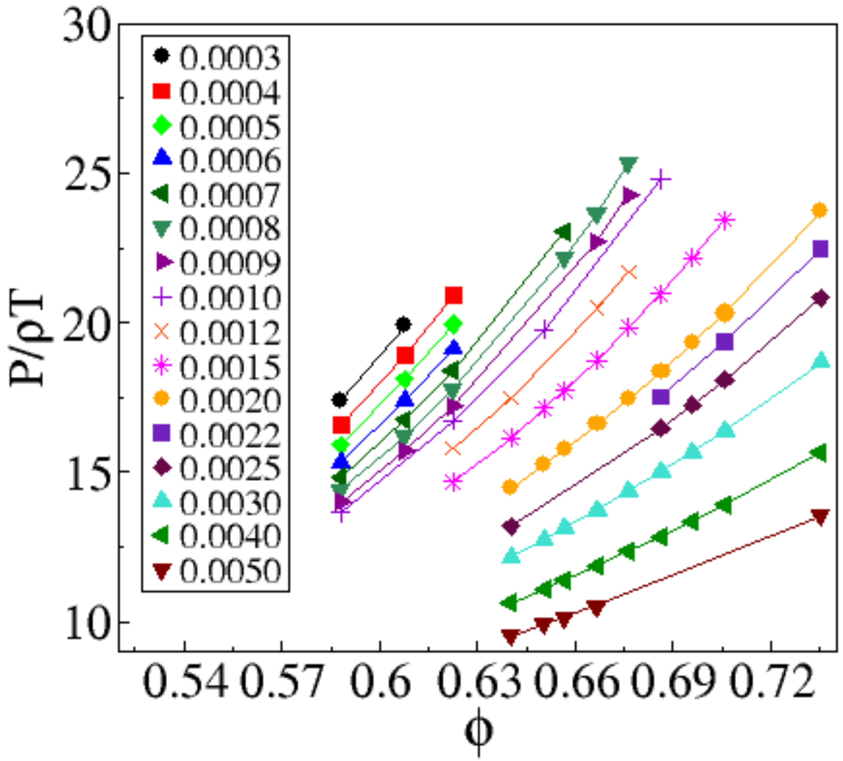
Barker and Henderson, JCP (1967)

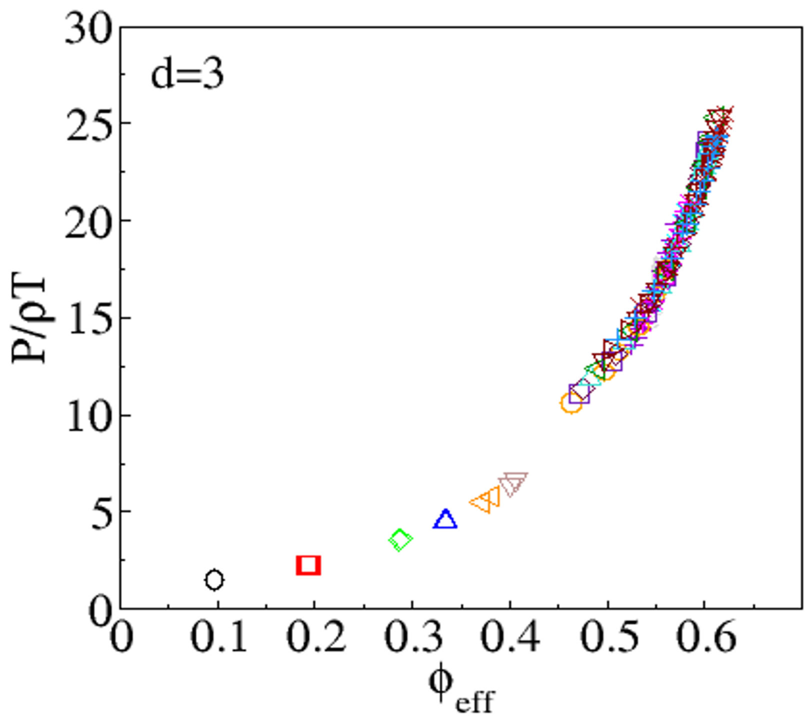
Equation of state to estimate b and β

We have four free parameters: b, β, δ and φ_0 !!!!

Rather than analyse dynamical data to estimate all of them, we employ the equation of state as the first step.

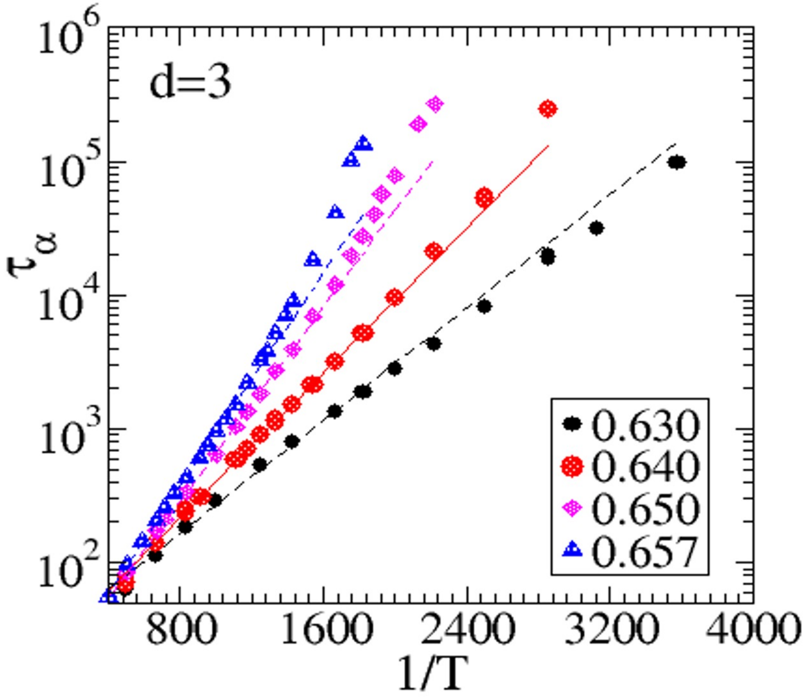
We choose b and β from pressure collapse. For a good choice of φ_{eff} pressure should collapse onto a master curve:

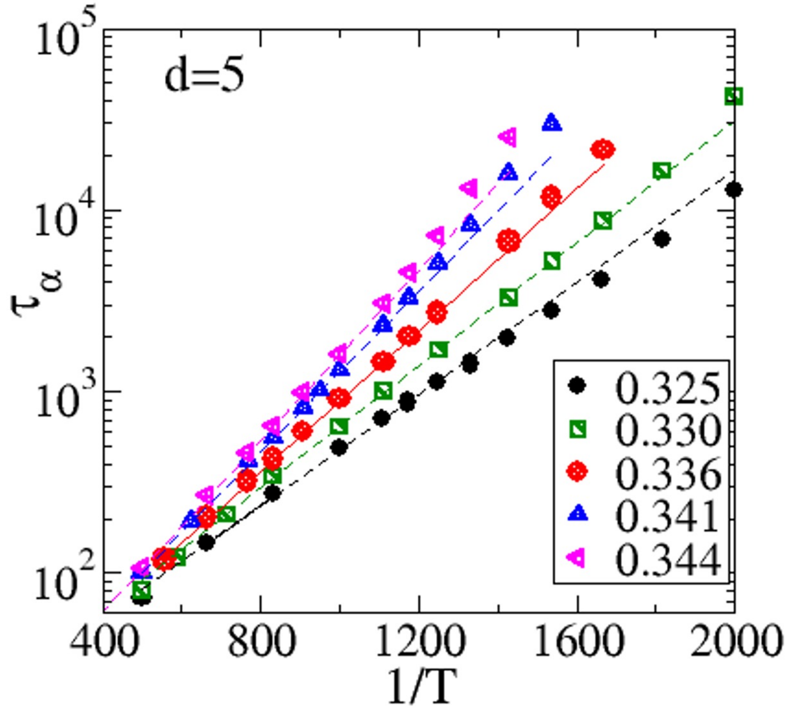




Choice of ϕ and δ

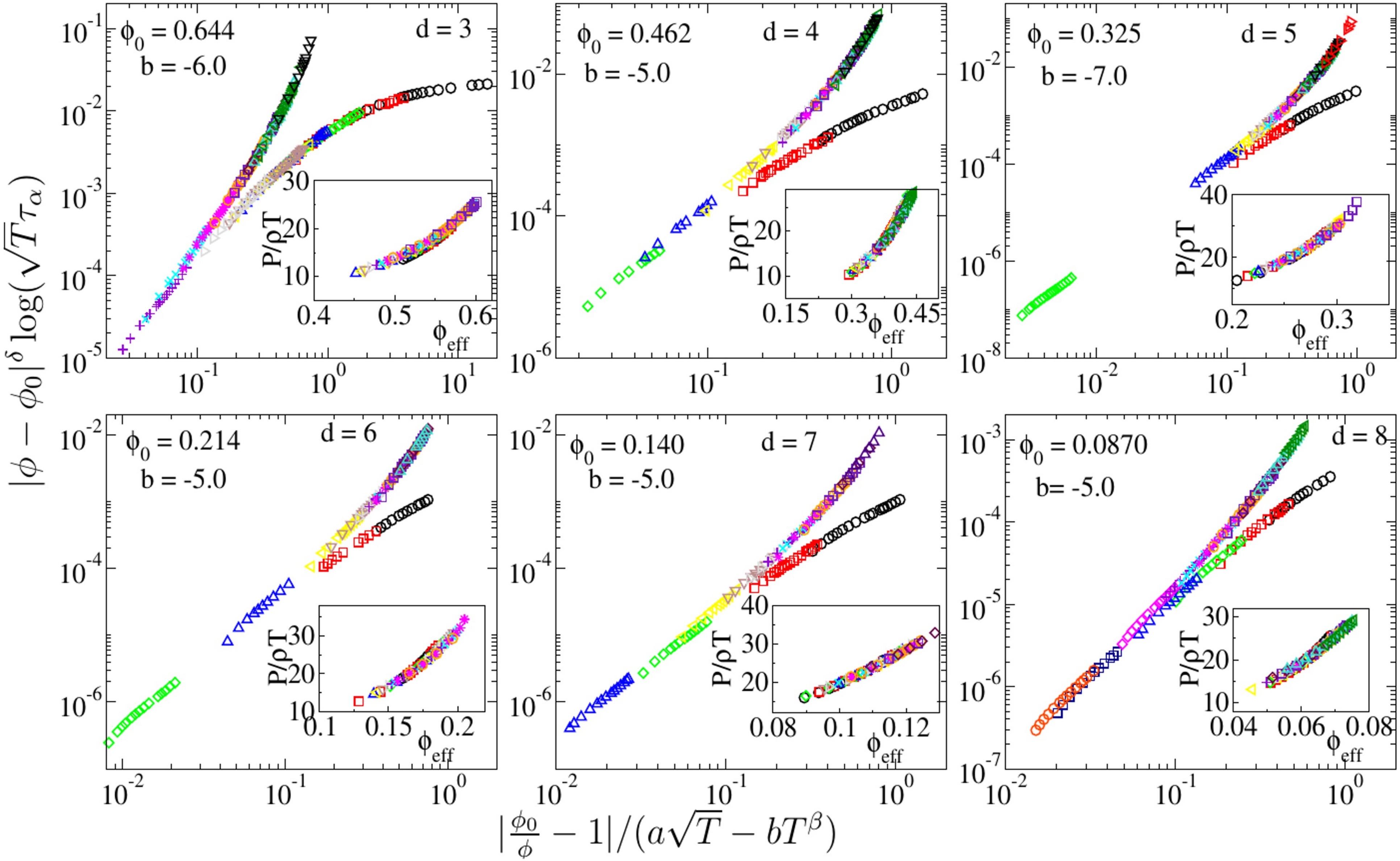
δ = 2.0 is well accepted value in literature. (We also have some understanding about this number.





Arrhenius fits -- demonstrates that the change from sub-Arrhenius to super-Arrhenius behavior occurs within the range of densities shown. Use this density range for initial guess of φ_0

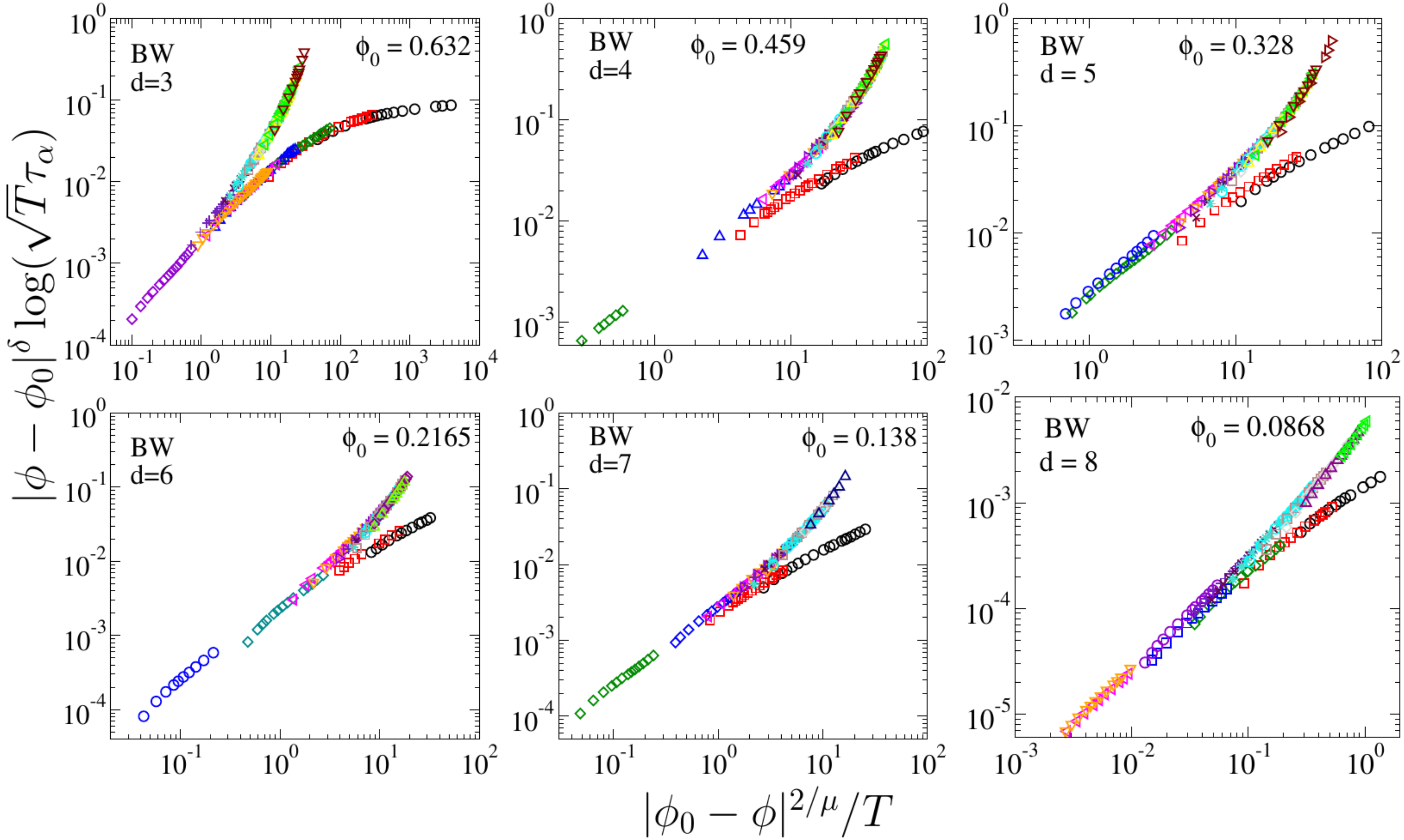
Scaling Collapse of τ: All dimensions



Very satisfactory scaling collapse.

Revisiting Berthier-Witten scaling

We impose: $\mu\delta = 2$, for Arrhenius behavior at $\varphi = \varphi_0$



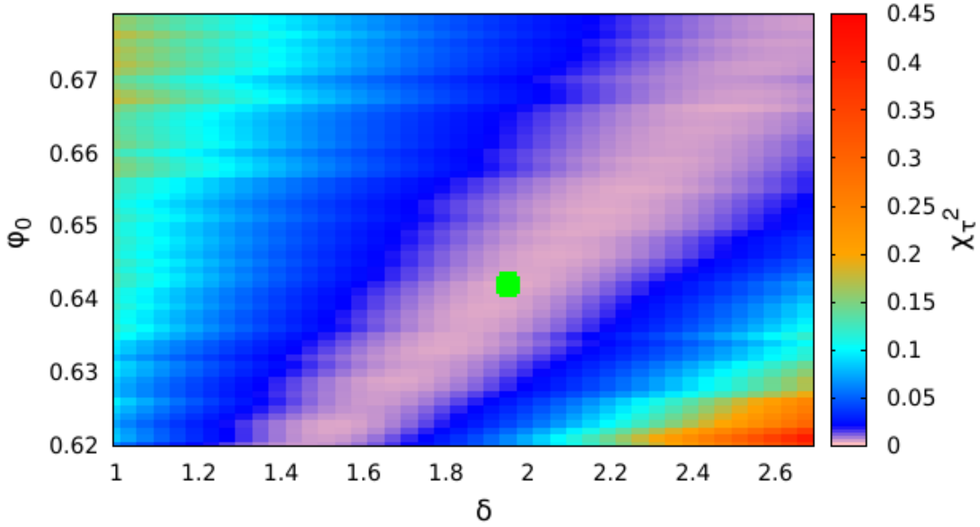
φ_0 values similar to our estimates.

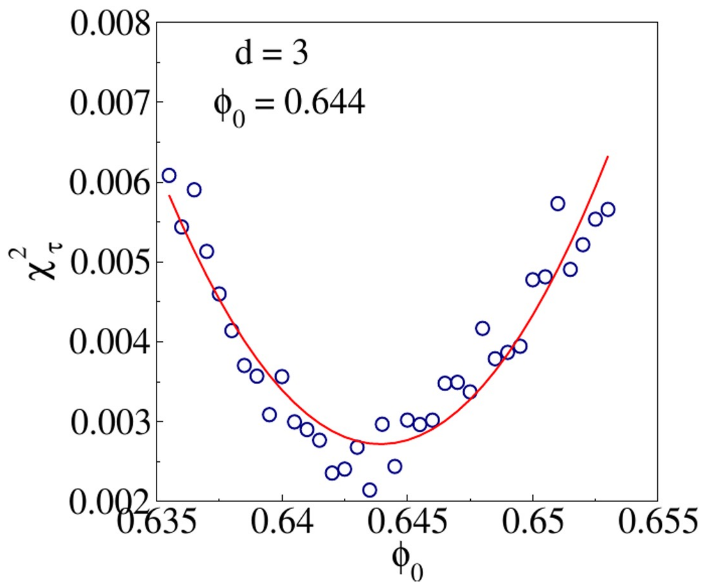
Quality of data collapse: Error analysis

For each subset of data, for each data point, we define an x value and a y value

$$x = \log\left(\frac{|\frac{\phi_0}{\phi} - 1|}{a\sqrt{T} - bT^{\beta}}\right) \quad y = \log\left[|\phi_0 - \phi|^{\delta} \log\left(\sqrt{T}\tau_\alpha(\phi, T)\right)\right] \quad \langle y_i^{\pm}\rangle = \frac{1}{n_i^{\pm}} \sum_{j=1}^{n_i^{\pm}} y_j^{\pm}$$

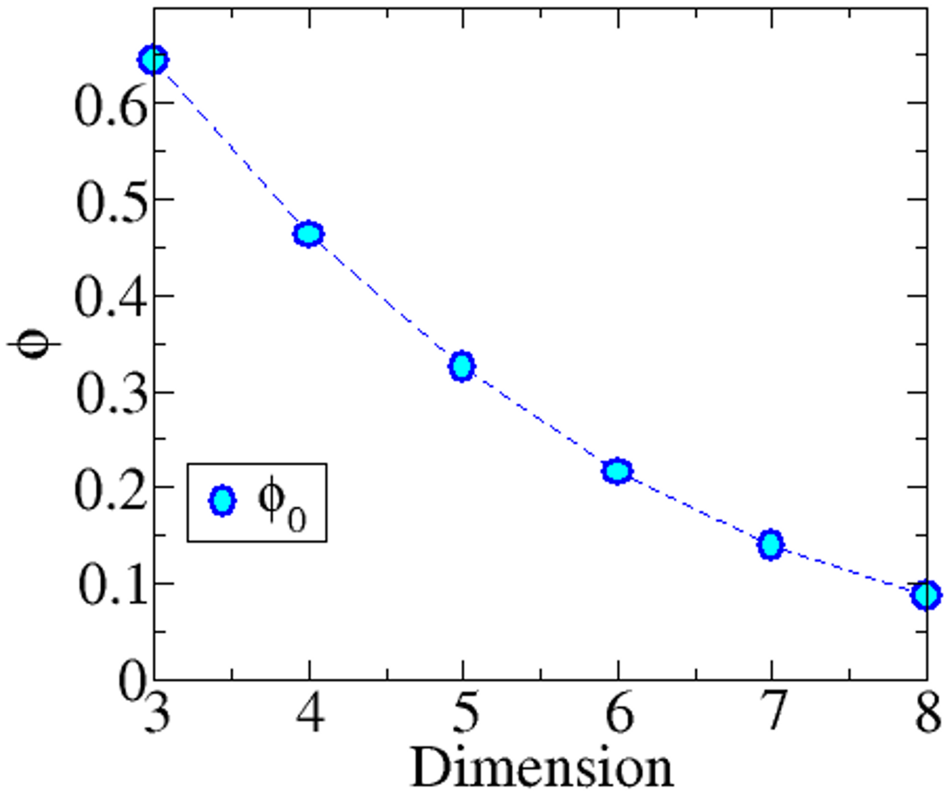
$$\chi_\tau^{\pm 2} = \frac{1}{tot_{bin}(n_i > 1)} \sum_{i(n_i^{\pm} > 1)} \frac{1}{n_i^{\pm}} \sum_{j=1}^{n_i^{\pm}} \left[y_i^{\pm}(j) - \langle y_i^{\pm}\rangle\right]^2$$





Minimum error around δ =2. Fixing δ =2, we can precisely compute φ_0.

φ_0: All dimensions



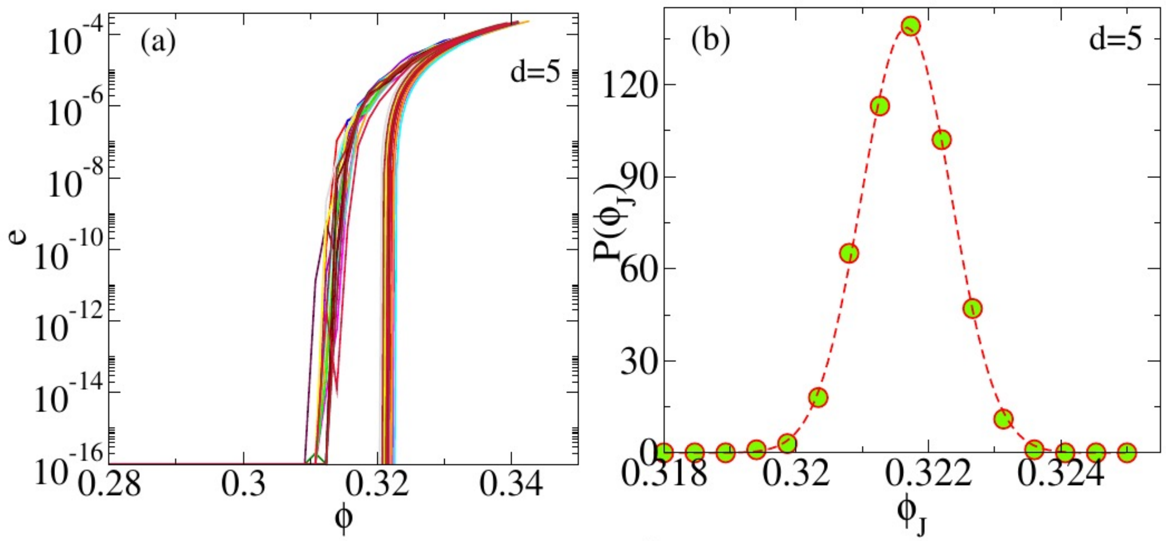
Dimension	ϕ_0
3	0.644
4	0.462
5	0.325
6	0.214
7	0.140
8	0.0870

φ_0 decreases with increasing dimension.

Jamming point

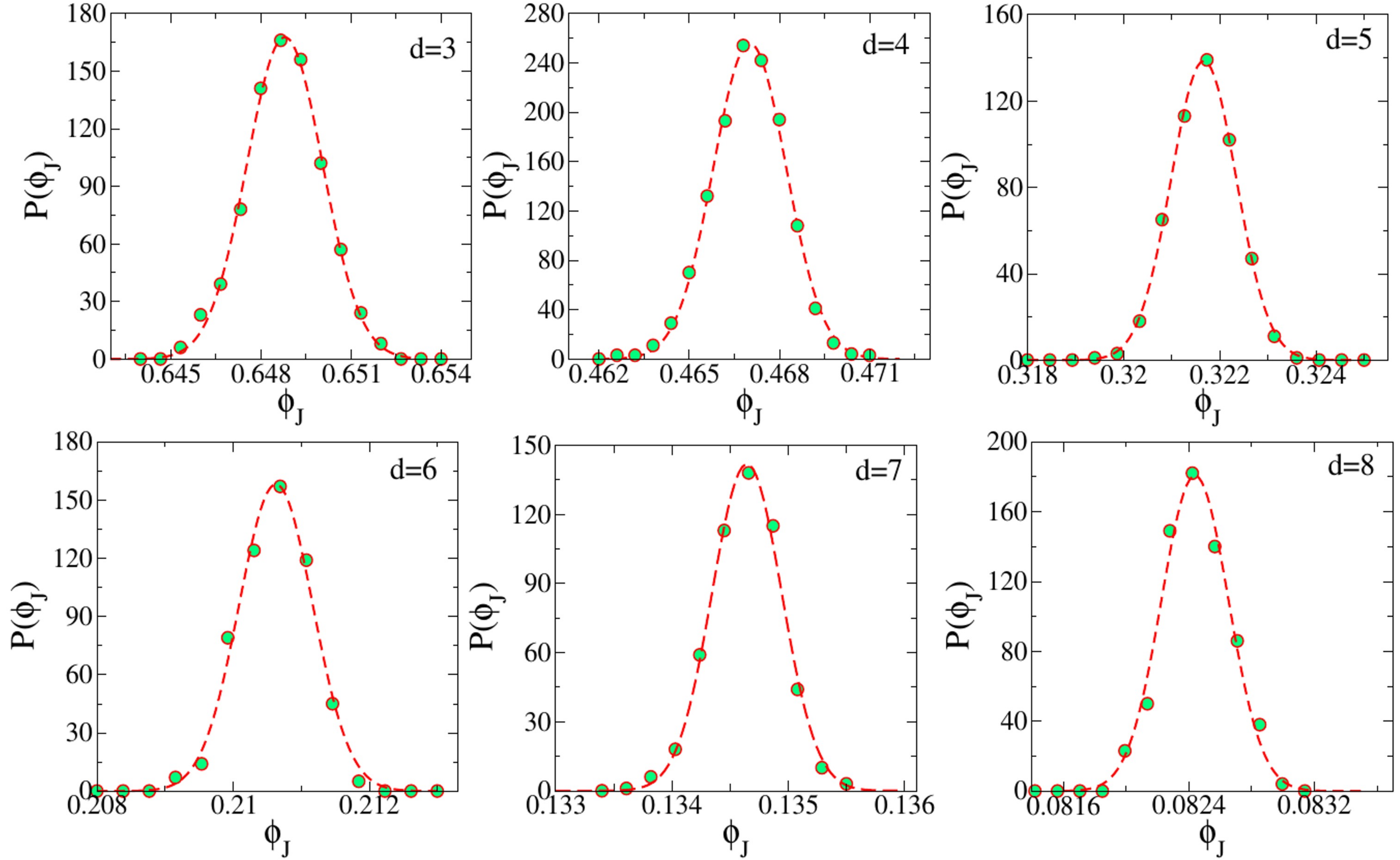
Protocol:

- Begin by random configuration of spheres.
- Apply compression uniformly by inflating the particle diameter, minimize the energy at each step of compression
- continue this process until the system reaches the energy of the order of 10^{-5} .
- Now, decompress the system minimizing energy at each step.
- The volume fraction at which energy becomes 10^{-16} is the jamming point.

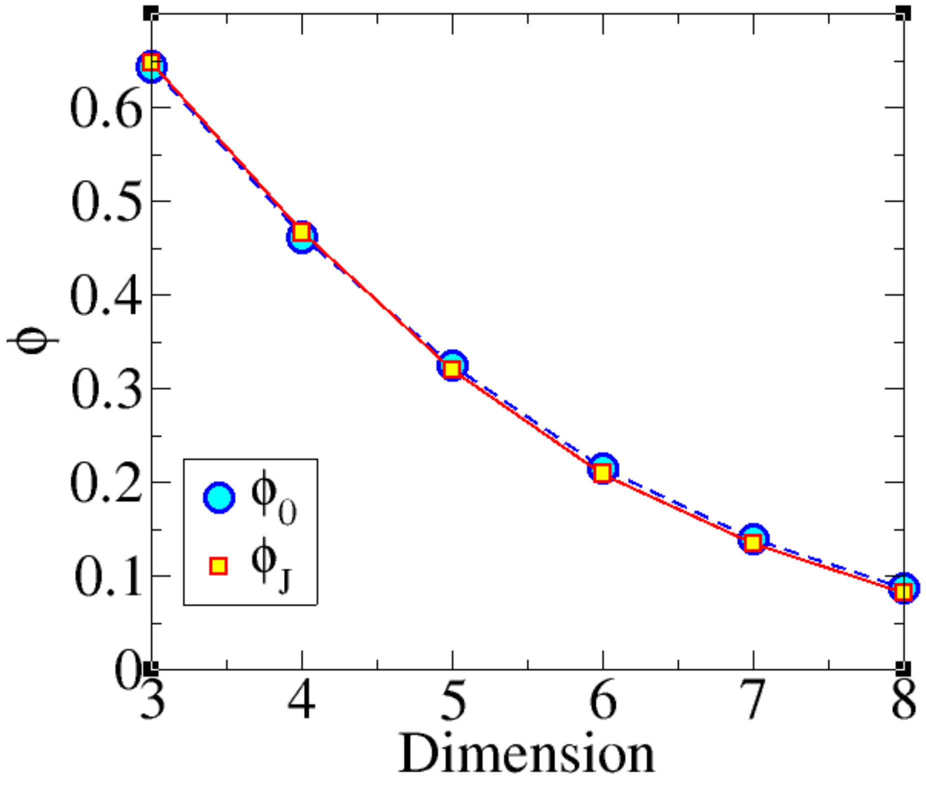


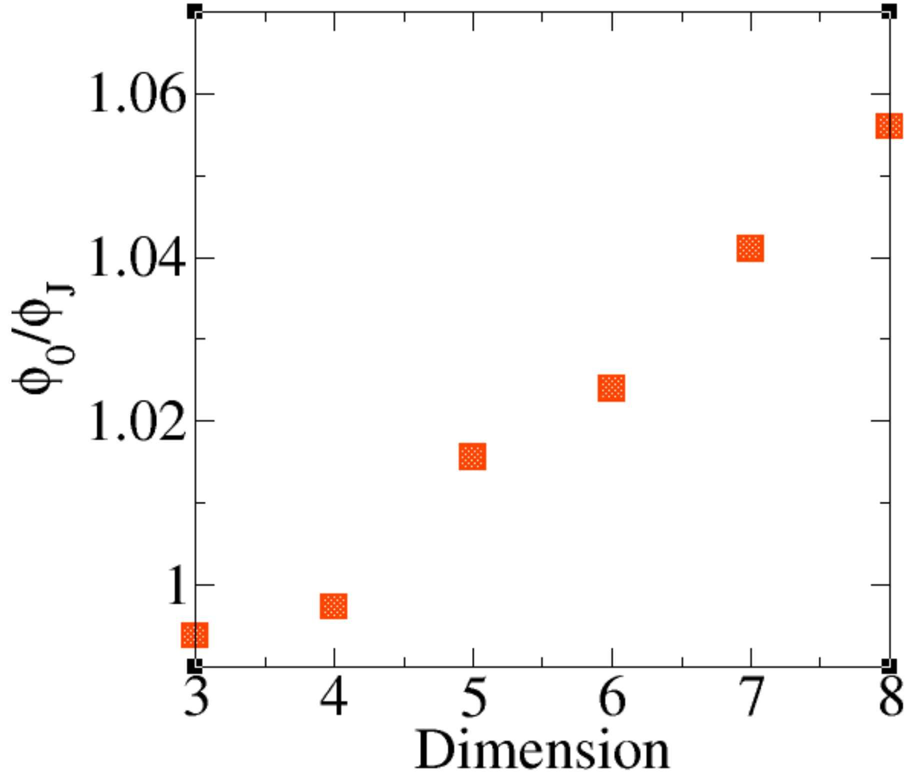
Obtain φ_J by averaging over a large number of initial samples

Jamming density: All dimensions



Glass transition and jamming densities





φ_J also decreases as spatial dimension becomes larger.

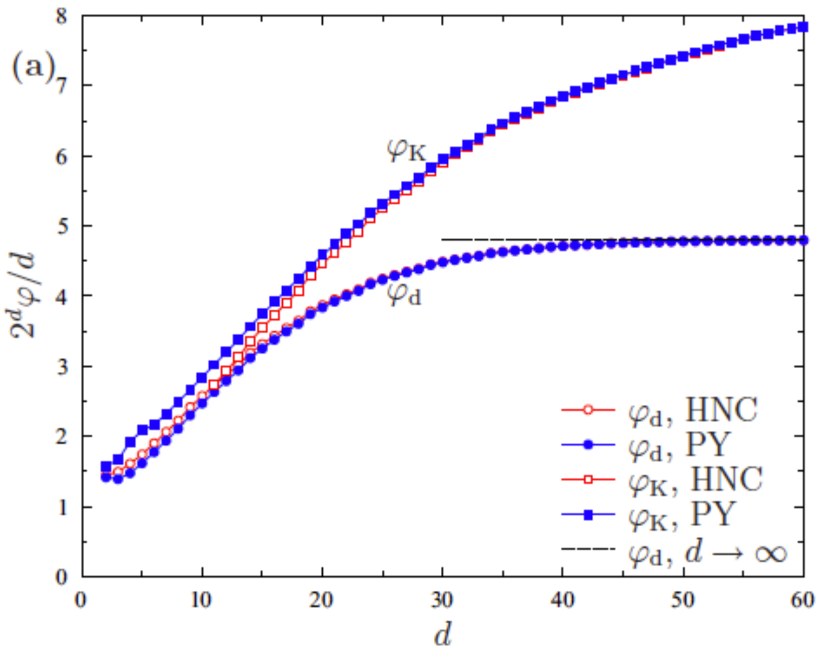
The ratio φ_0/φ_J increases with dimension with $\varphi_0 > \varphi_J$ for $d > 4$.

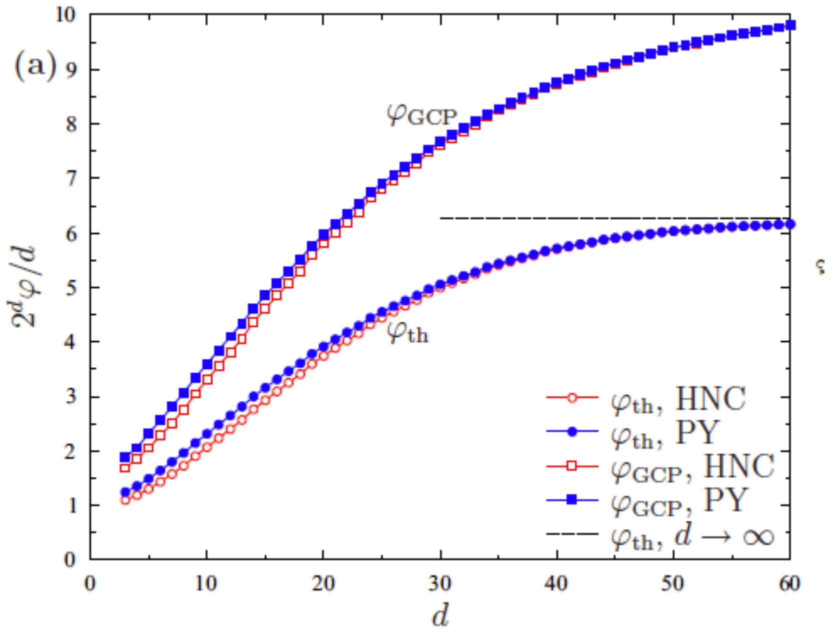
For $d = 3,4$, $\varphi_0 < \varphi_J$, with the two values being very close.

Theoretical results in finite dimension

Approximation scheme around the infinite dimensional solution – Mangeat and Zamponi (PRE 2016).

Theoretical prediction: $2^d\varphi_K/d \sim \log d$, $2^d\varphi_{th}/d \sim$ constant

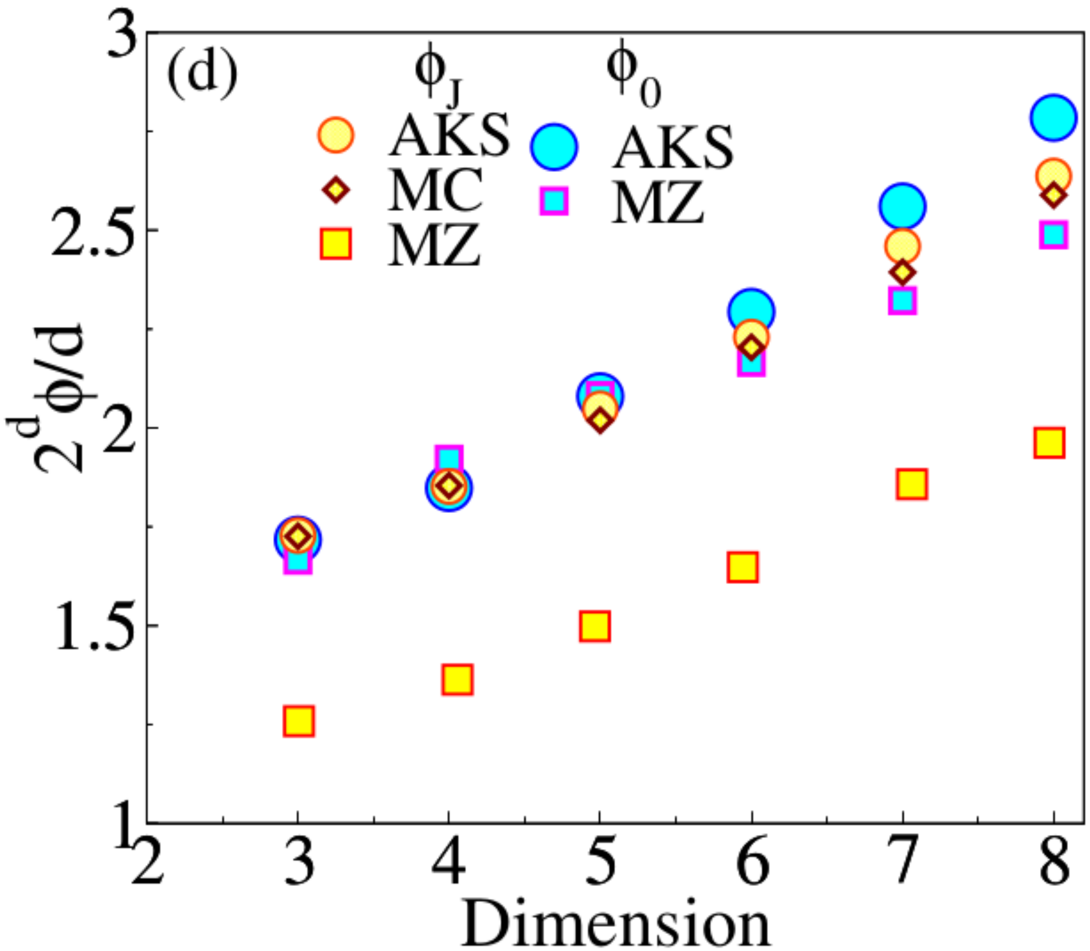




ϕ_{th} underestimates the jamming density.

Mangeat and Zamponi, Phys Rev E (2016).

Comparison with theoretical results

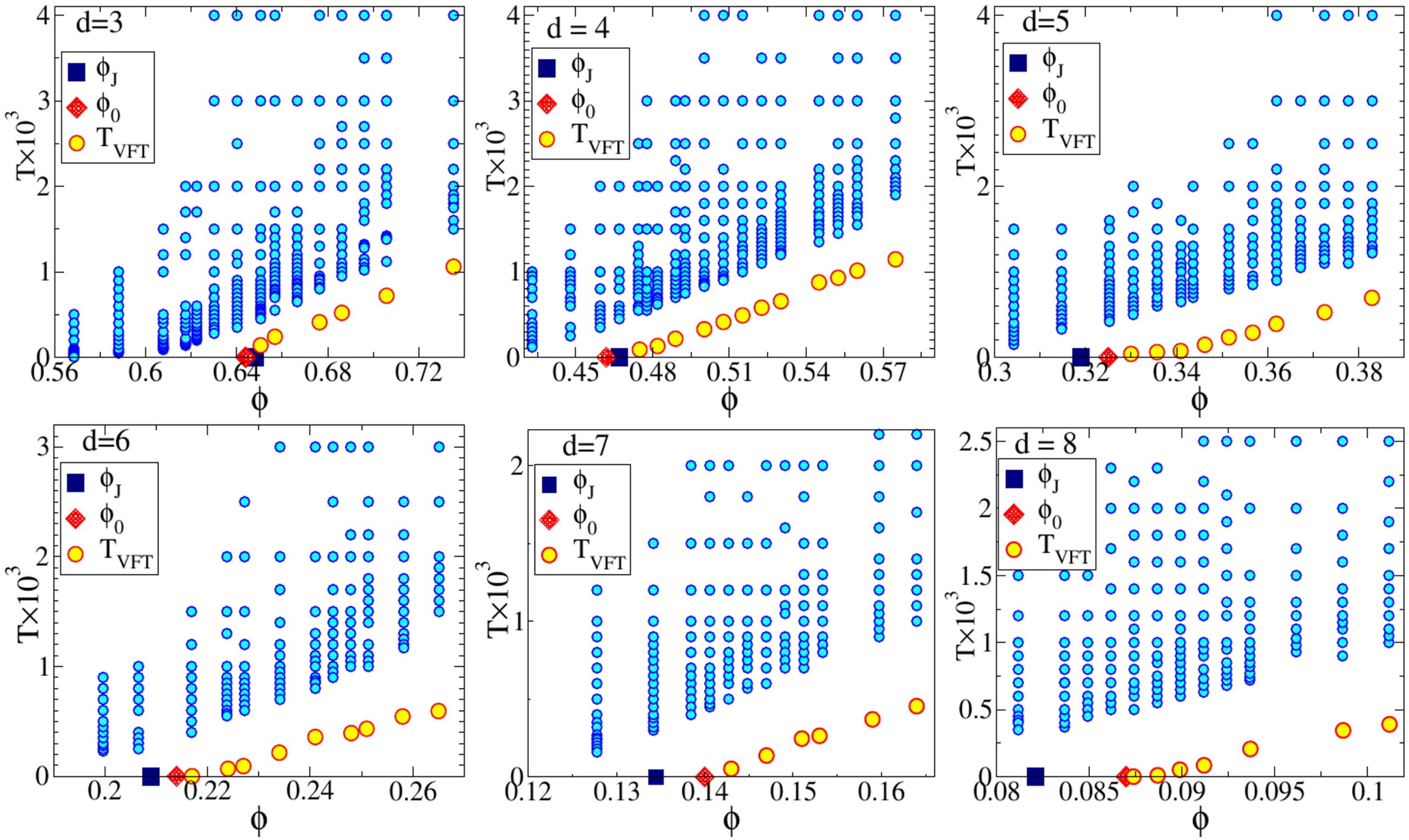


Although we are not able to verify quantitatively the prediction that ϕ_0 increases as log d, the values of ϕ_0 are in near quantitative agreement with theoretical results.

Density-Temperature diagram

we compute the temperature at which the relaxation times diverges by fitting the data at each density above φ_0 , to the VFT form:

$$\tau_\alpha = \tau_0 \exp\{1/K_{\rm VFT}[(T/T_{\rm VFT}) - 1]\}$$



density-temperature diagram shows φ_0 and φ_J, along with the density dependent T_{VFT}.

Summary

We perform a scaling analysis of the dynamics of soft sphere fluids in dimensions 3 to 8.

We propose and employ a new scaling procedure that employs the equation of state in addition to dynamical data.

We estimate the ideal glass transition and jamming densities, and show clearly that they are distinct, with a ratio that grows with spatial dimensionality.

The estimated glass transition densities are in near quantitative agreement with theoretical calculations.

Thank you for attention