Statistical Mechanics of Granular Materials

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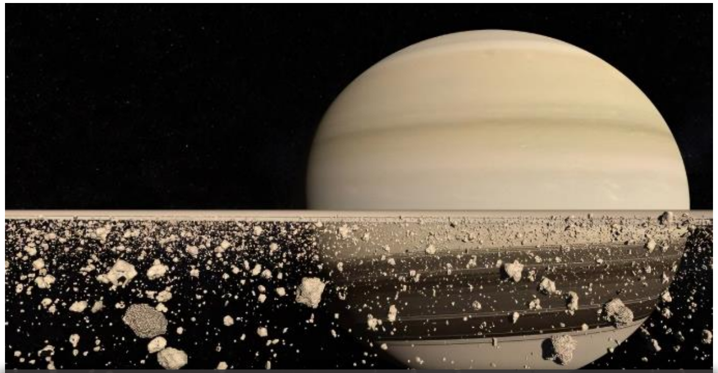
Frontiers in Non-equilibrium Physics, 2024.07

Granular Materials

- Discrete system consisting of a large number of particles of macroscopic size (>1 μm).
- These materials are closely related to many important industrial applications, and are also the carriers for many geological processes like earthquake, avalanches and etc.


Food industry


Mining industry


Aerospace industry


Desert


Earthquake


Avalanche

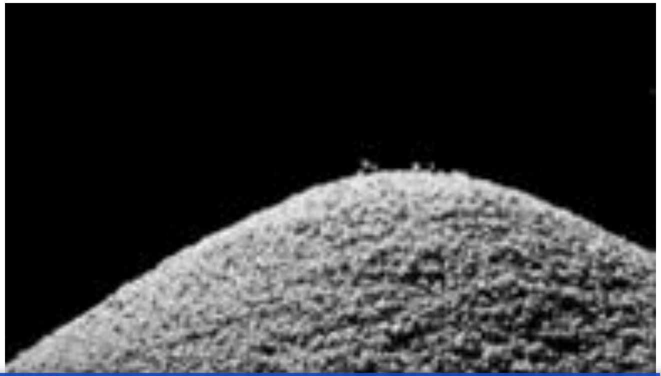
The phase behaviors of granular materials

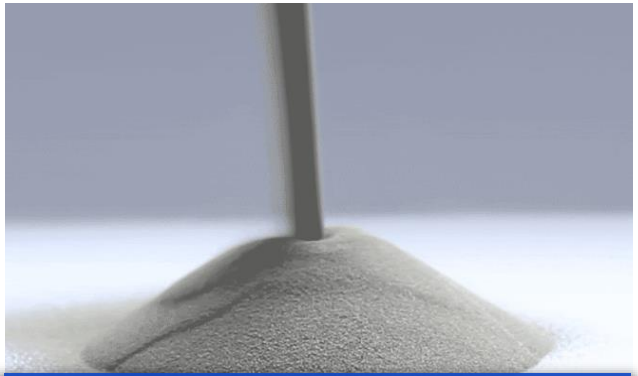
- Granular materials are by nature out-of-equilibrium systems due to the macroscopic size of the particles and irrelevancy of thermal motion.
- However, under external driving forces, they can still display gas, liquid, and and solid phases similar to those of molecular and atomic systems.

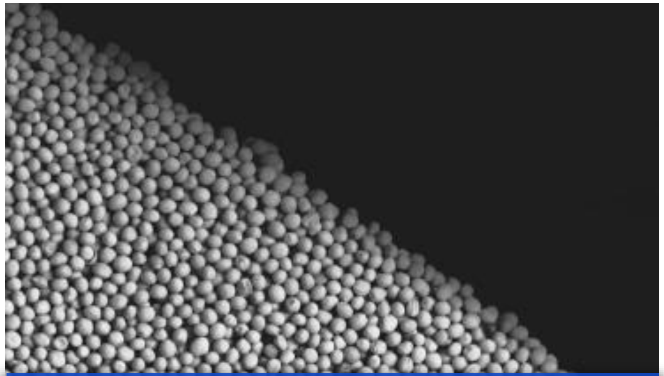

Gas


Liquid


Solid


Granular gas

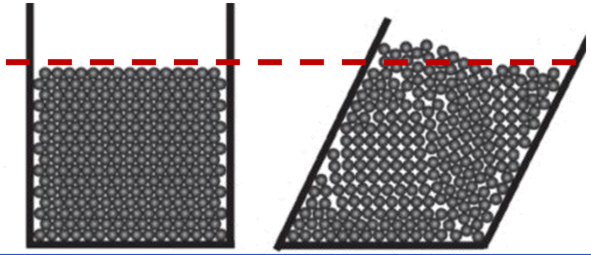

Granular liquid


Granular solid

Complex properties

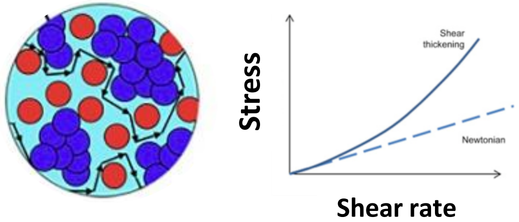
- Unlike ordinary Newtonian fluids or elastic solids, granular materials display **complex rheological properties** like shear dilatancy, shear thickening, and arching effect, etc.

Shear dilatancy

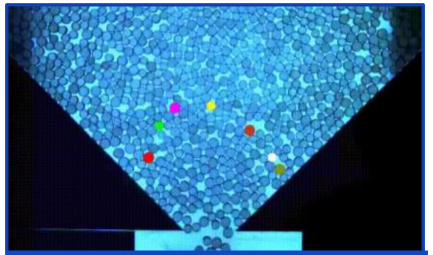


Volume fraction increases under shear

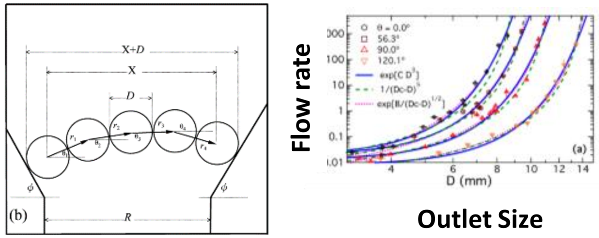
Shear thickening



Viscosity increases under rapid shear



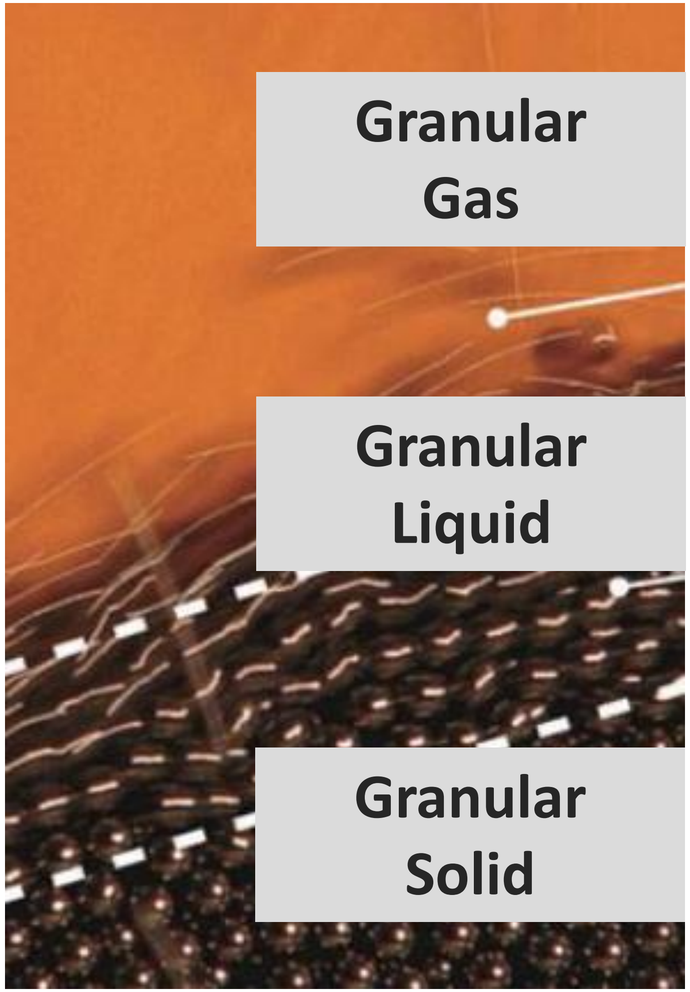
Arch effect

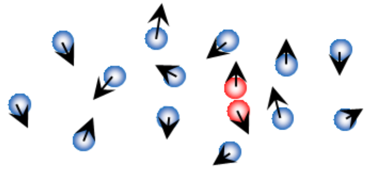
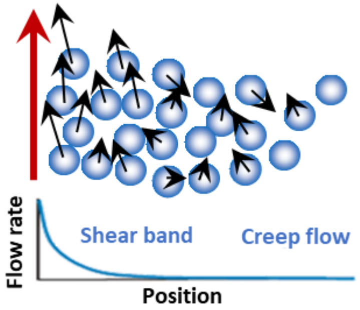
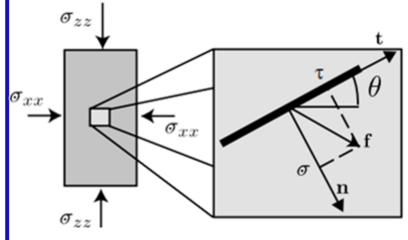


Mechanical stable structures form at the outlet

Empirical Constitutive Theories

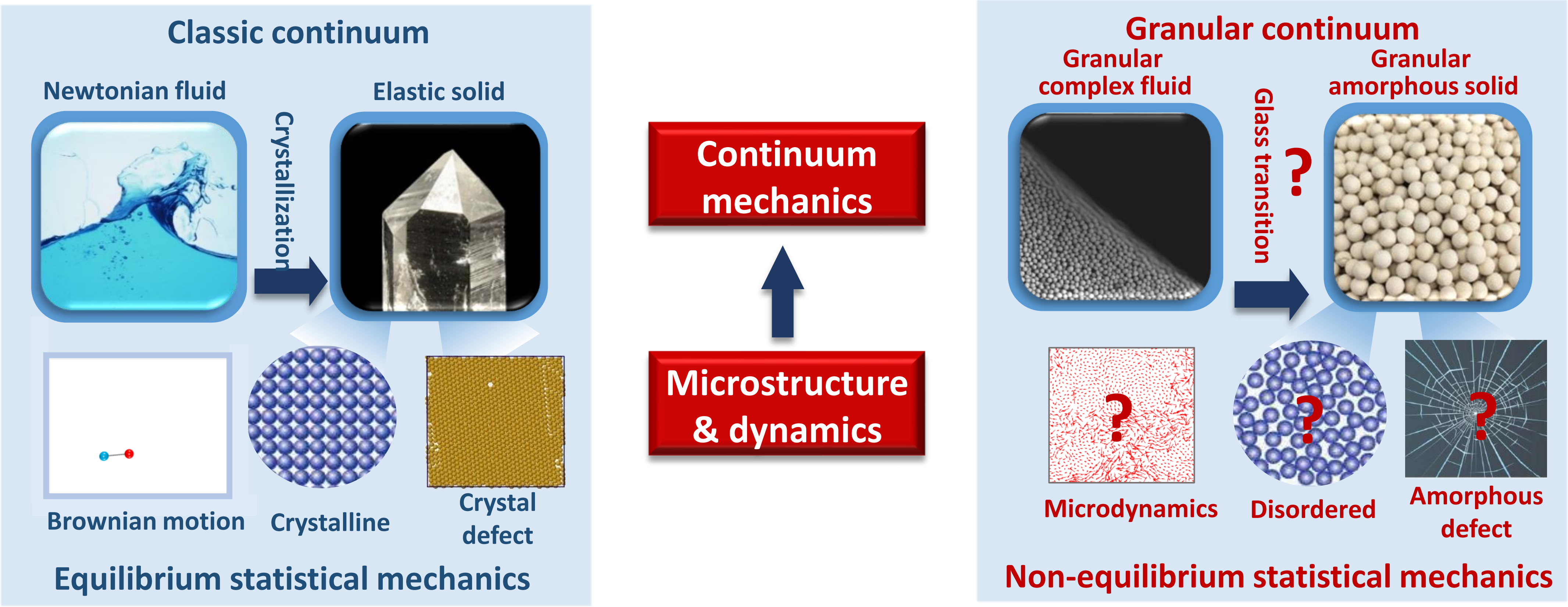
- Historically, empirical constitutive theories are normally employed to understand the behaviors of granular materials.
- However, these theories lack microscopic mechanisms, and encountered many difficulties.

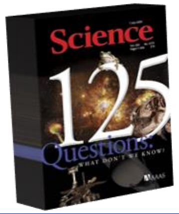


 Dissipative inelastic collision	$P \propto \rho_p d^2 \dot{\gamma}^2$ $\tau \propto \rho_p d^2 \dot{\gamma}^2$	Bagnold's Scaling (Dimensional analysis)
	$\mu = \tau / \sigma \quad I = \frac{\dot{\gamma} d}{\sqrt{\sigma / \rho}}$ $\mu(I) = \mu_1 + \frac{\mu_2 - \mu_1}{I_0 / I + 1}$	$\mu(I)$ rheology (viscoplastic)
 Mohr-Coulomb Model	$\sin\delta = \frac{r}{\sigma_0} = \frac{\sigma_{zz} - \sigma_{xx}}{\sigma_{xx} + \sigma_{zz}}$	Mohr-Coulomb Model (rigid-plastic)

Methods

■ To develop a continuum theory for granular materials, we need to establish the connections between the micro and the macro within **a statistical framework for out-of-equilibrium granular systems**.





Science
125 questions

43. Can we develop a general theory of the dynamics of turbulent flows and the motion of granular materials?

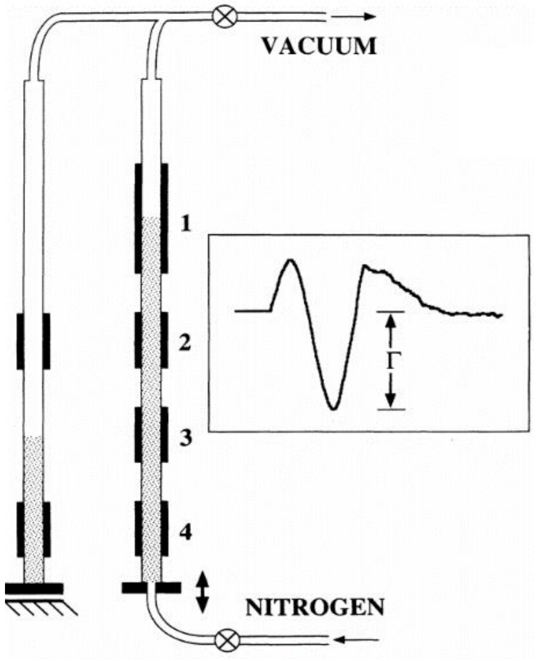
Statistical mechanics for granular materials?

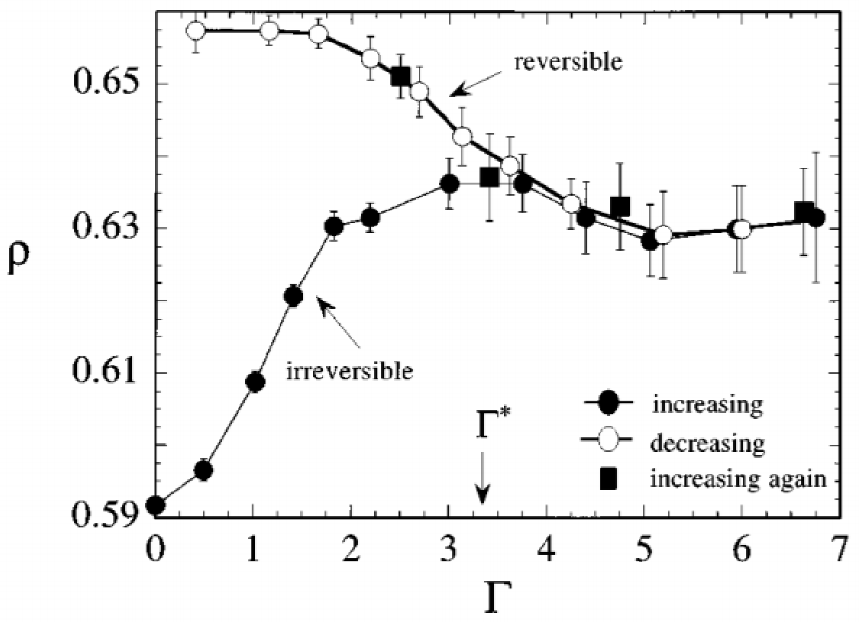
Edwards ensemble

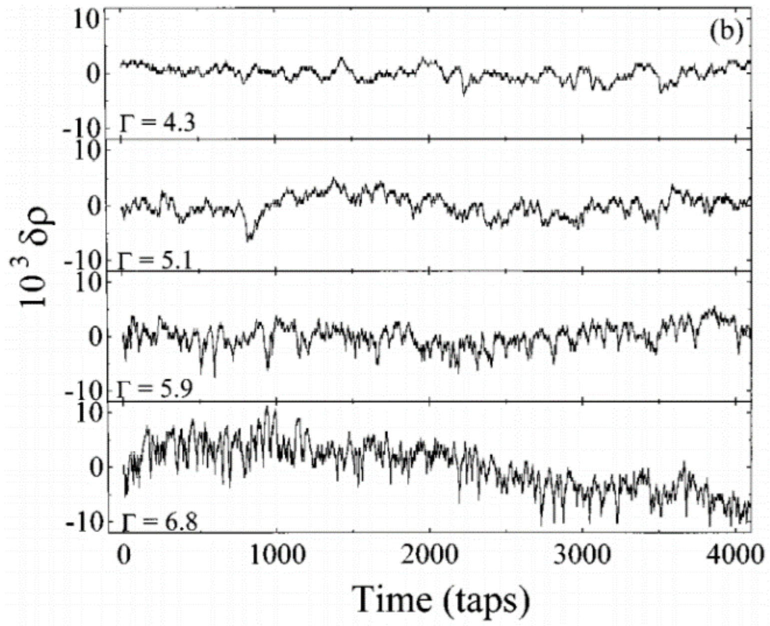
- **Granular materials are by nature out-of-equilibrium, as thermal motions of the particles are negligible. Without external perturbation, granular materials will remain in stable packing state.**
- **Edwards and coworkers introduced a statistical mechanical framework—Edwards ensemble for granular packings.**

Edwards and Oakeshott, *Physica A*, 1989.

Reproduceable macroscopic observables







Granular packings under consecutive tapping can evolve into history-independent stationary states with fairly constant volume fractions, and it fluctuates around its mean value. This resemble the behavior of the energy of a thermal system at constant temperature.

Knight *et al.*, *PRE*, 1995.
Nowak *et al.*, *PRE*, 1998.

Edwards statistical ensemble for granular materials

S. F. Edwards

"We assume that when *N* grains occupy a volume *V* they do so in such a way that **all configurations are equally weighted**. It is the analog of the **ergodic hypothesis of conventional thermal physics**."

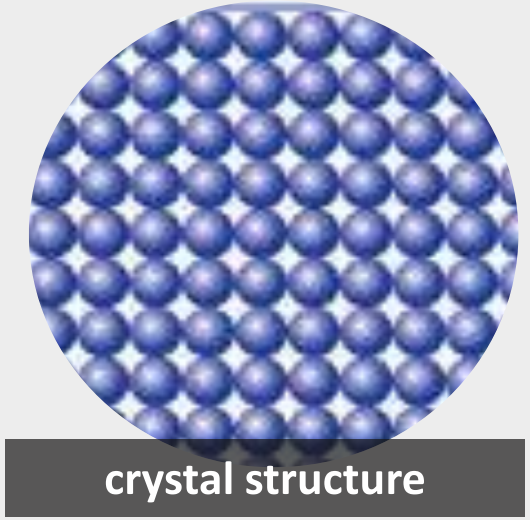
	Boltzmann	Edwards
Conserved quantity	Energy, E	Volume, V
Number of valid configurations	$\Omega_B(E)$	$\Omega(V)$
Entropy	$S = k_B \ln \Omega_B$	$S = \lambda \ln \Omega$
Equilibrating quantity	Temperature	Compactivity
	$\frac{1}{T} = \frac{\partial S(E)}{\partial E}$	$\frac{1}{X} = \frac{\partial S(V)}{\partial V}$
Distribution	$e^{-\frac{E}{k_B T}}$	$e^{-\frac{V}{X}}$

The number of mechanical stable states of granular packing (refers to $\Omega(V)$)

Bi et al., *Annu. Rev. Condens. Matter Phys.*, 2015.   Martiniani et al. *Nat. Phys.*, 2017.

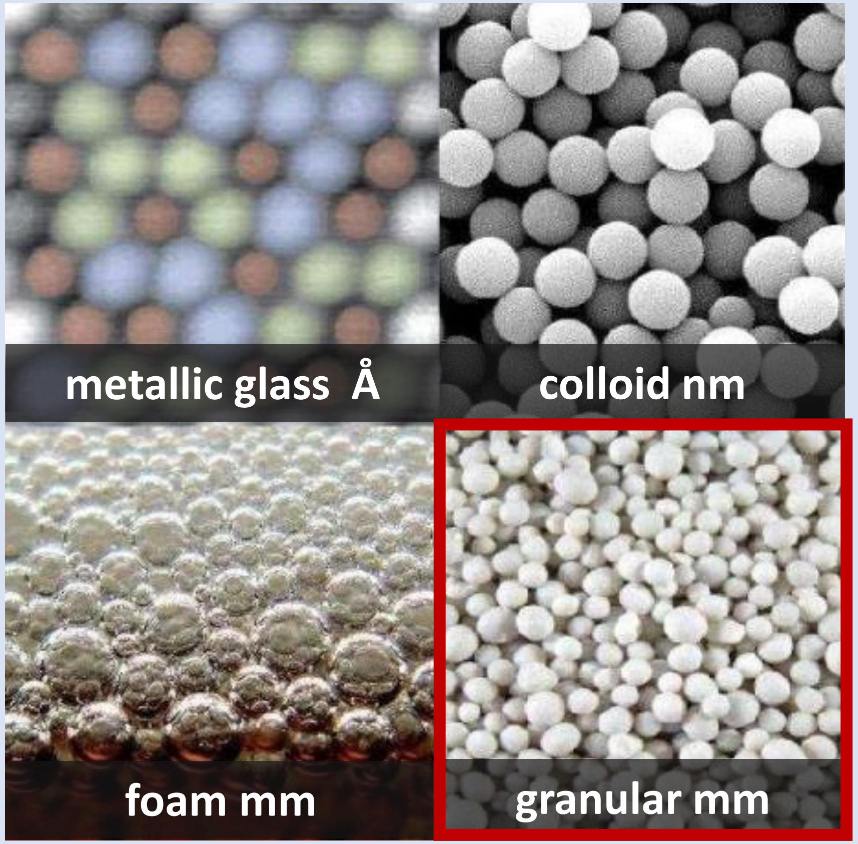
Granular packings as a class of disordered materials

➢Crystal



➢Disordered materials

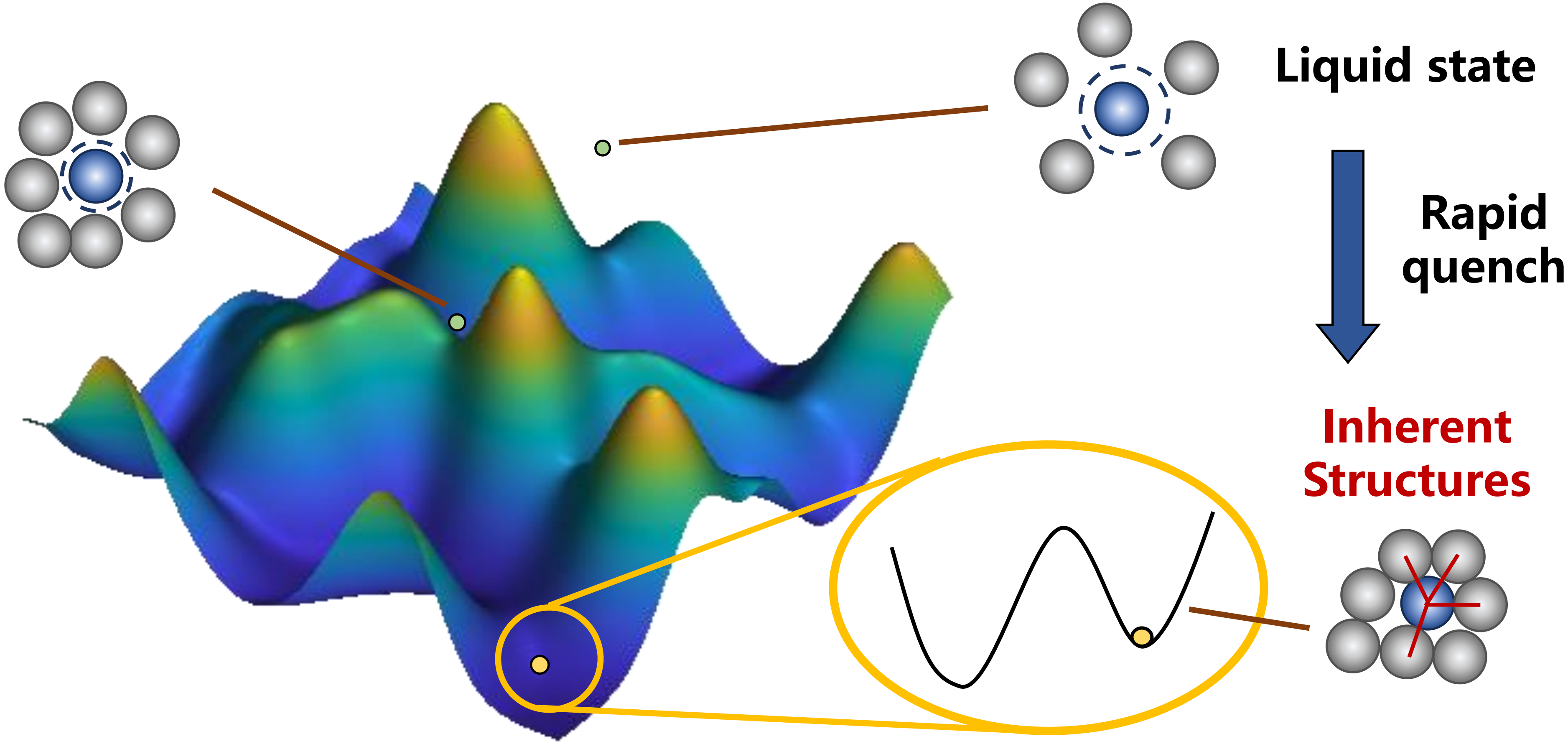
granular materials are one class of disordered systems.



Berthier and Biroli, *RMP*, 2011

Landscape of disordered materials

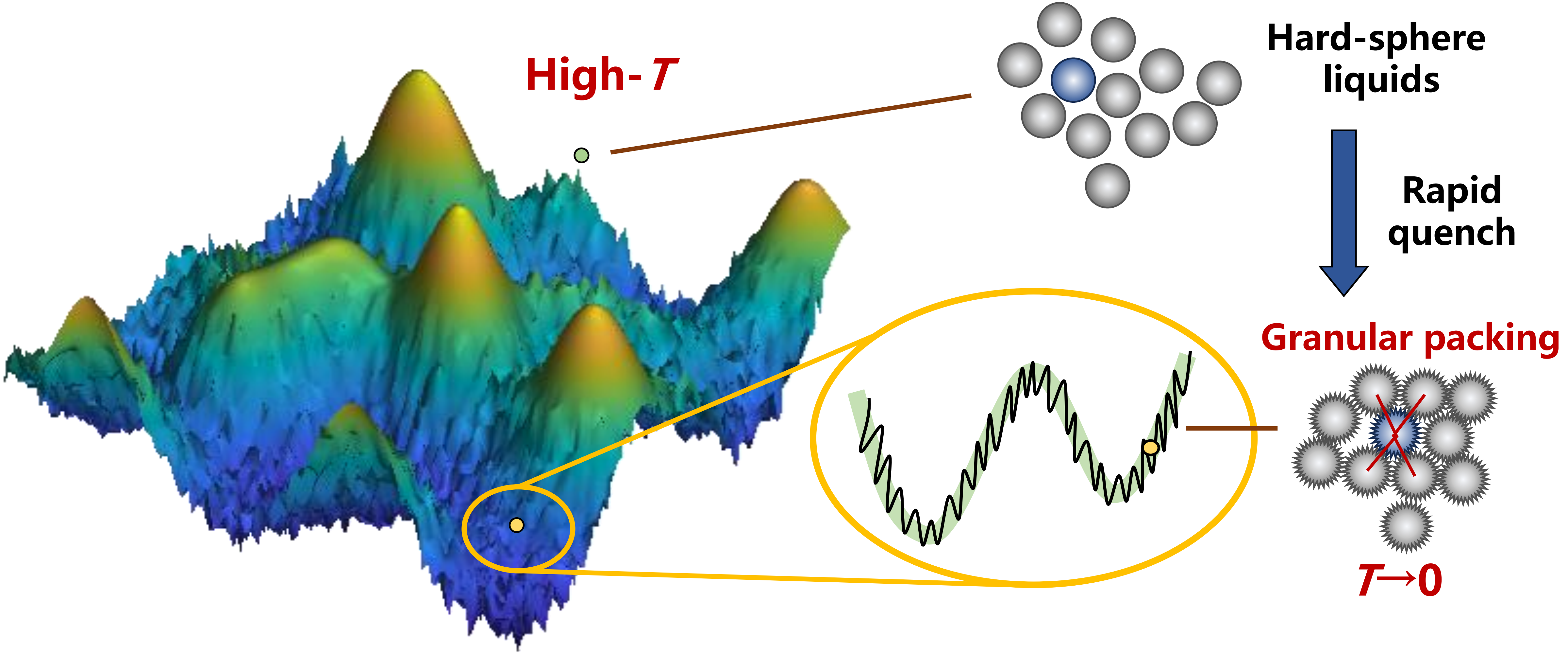
- In order to describe the disordered configuration of the amorphous solids, the concept of the energy landscape is introduced, where each local energy minimum represents a microstate of the system.



Charbonneau et al. *Nat. Commun.*, 2014.

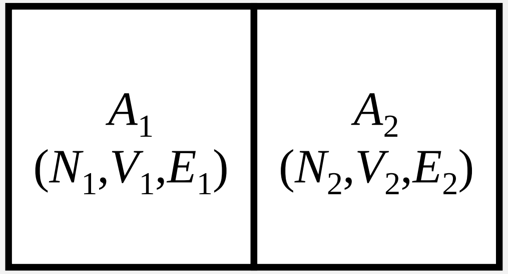
Landscape of granular materials

- **For granular materials, due to the presence of friction, the landscapes become very rugged because many highly distorted configurations can be stablized, and each of the local minima corresponds to a microstate of the Edwards ensemble.**



Entropy and Compactivity

➢ Thermal system



Number of microstates Ω

Energy E

$$\Omega(E_1+E_2)=\Omega_1(E_1)\Omega(E_2)$$

$$\frac{\partial\Omega}{\partial E_1}=\frac{\partial\Omega_1(E_1)}{\partial E_1}\Omega_2(E_2)+\Omega_1(E_1)\frac{\partial\Omega_2(E_2)}{\partial E_2}\frac{\partial E_2}{\partial E_1}=0$$

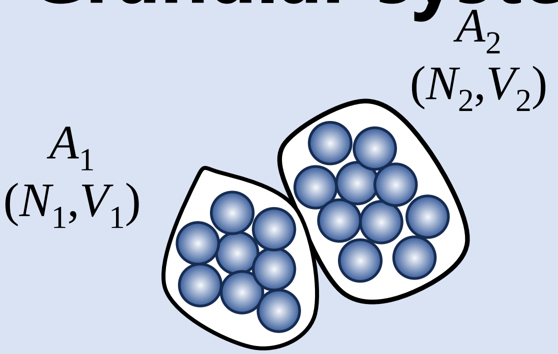
Maximum Entropy Principle

With $\frac{\partial E_2}{\partial E_1}=-1$ **Conservation of energy**

$$\frac{\partial\ln\Omega_1(E_1)}{\partial E_1}=\beta_1=\beta_2=\frac{\partial\ln\Omega_2(E_2)}{\partial E_2}$$

$$S=k\ln\Omega(E)\qquad\beta=\frac{1}{k_BT}$$ **Temperature**

➢ Granular system



Two system separated by a "soft wall", so that the volume become variable

$$\Omega(V_1+V_2)=\Omega_1(V_1)\Omega(V_2)$$

$$\frac{\partial\Omega}{\partial V_1}=\frac{\partial\Omega_1(V_1)}{\partial V_1}\Omega_2(V_2)+\Omega_1(V_1)\frac{\partial\Omega_2(V_2)}{\partial V_2}\frac{\partial V_2}{\partial V_1}=0$$

With $\frac{\partial V_2}{\partial V_1}=-1$ **Conservation of Volume**

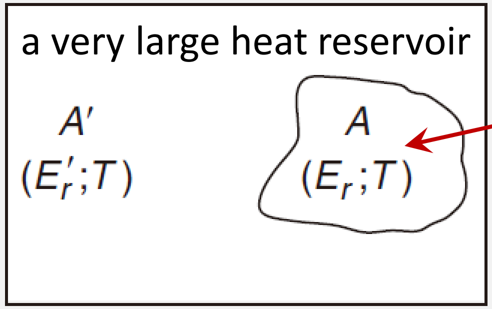
$$\frac{\partial\ln\Omega_1(V_1)}{\partial V_1}\equiv\beta_1=\beta_2\equiv\frac{\partial\ln\Omega_2(V_2)}{\partial V_2}$$

$$S=\ln\Omega(V)\qquad\beta=\frac{1}{\chi}$$ **Compactivity**

Pathria and Beale, Statistical Mechanics (Third Edition)

The Canonical Ensemble

➢ Thermal system



A given subsystem

$$E_r + E'_r = E^{(0)} = const. \quad \text{and} \quad \frac{E'_r}{E^{(0)}} = \left(1 - \frac{E_r}{E^{(0)}}\right) \simeq 1$$

$$\begin{cases} P_r \propto \boldsymbol{\Omega}'(E'_r) \equiv \boldsymbol{\Omega}'(E^{(0)} - E_r) \\ \ln \boldsymbol{\Omega}'(E'_r) = \ln \boldsymbol{\Omega}'(E^{(0)}) + \left(\dfrac{\partial \ln \boldsymbol{\Omega}'}{\partial E'}\right)_{E'_r = E^{(0)}} (E'_r - E^{(0)}) + \cdots \\ \qquad \simeq \text{const.} - E_r / k_B T \end{cases}$$

Thus $P_r \propto \exp(-E_r / k_B T)$

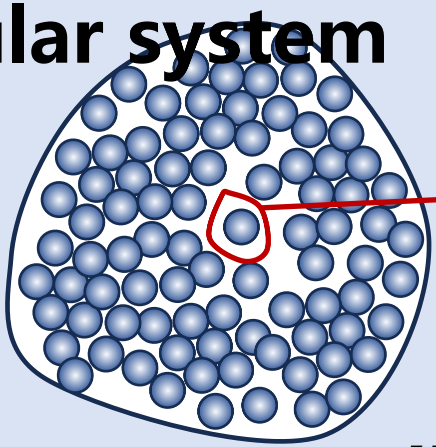
$$P_r = \frac{\exp(-E_r / k_B T)}{\sum_r \exp(-E_r / k_B T)}$$

Consider degenerate energy level

$$P_r = \frac{\Omega(E_r)\exp(-\beta E)}{\sum_r \Omega(E_r)\exp(-\beta E)}$$

Density of states (DOS)

➢ Granular system



A given subsystem in a very large packing

$$V_r + V'_r = V^{(0)} = const. \quad \text{and} \quad \frac{V'_r}{V^{(0)}} = \left(1 - \frac{V_r}{V^{(0)}}\right) \simeq 1$$

$$\begin{cases} P_r \propto \boldsymbol{\Omega}'(V'_r) \equiv \boldsymbol{\Omega}'(V^{(0)} - V_r) \\ \ln \boldsymbol{\Omega}'(V'_r) = \ln \boldsymbol{\Omega}'(V^{(0)}) + \left(\dfrac{\partial \ln \boldsymbol{\Omega}'}{\partial V'}\right)_{V'_r = V^{(0)}} (V'_r - V^{(0)}) + \cdots \\ \qquad \simeq \text{const.} - V_r / \chi'_r \end{cases}$$

Thus $P_r \propto \exp(-V_r / \chi)$

$$P_r = \frac{\exp(-V_r / \chi)}{\sum_r \exp(-V_r / \chi)}$$

The DOS of jammed microstates

$$P_r = \frac{\Omega(V_r)\exp(-V_r / \chi)}{\sum_r \Omega(V_r)\exp(-V_r / \chi)}$$

Pathria and Beale, Statistical Mechanics (Third Edition)

Motivation

- **Validity of the equal probability hypothesis of the Edwards ensemble.**

Density of State (DOS)

$$P_\mu(V) = \frac{\Omega_\mu(V)}{\int \Omega_\mu(V) e^{-V/\chi} dV} e^{-V/\chi} = \frac{\Omega_\mu(V)}{Z(\chi)} e^{-V/\chi}$$

Boltzmann factor

Boltzmann distribution

Partition function

- **How friction influences granular statistical mechanics, including both the density of states and the compactivity.**

Tapping experiment

➢ Particles

Particles (6mm)

- **ABS** plastic beads
- 3D-printed (**3DP**)
- bumpy surface (**BUMP**)

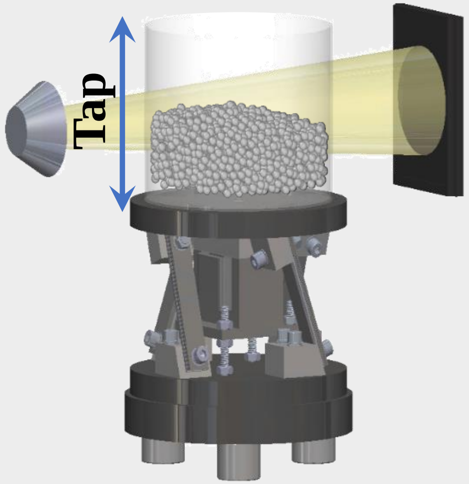
Friction Coefficient

(Measured by repose angle)

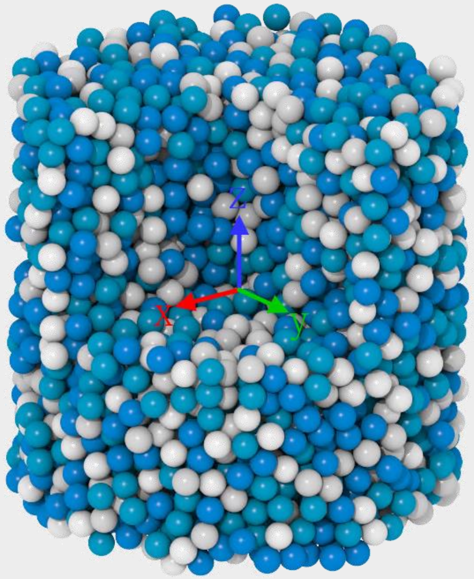
$$\mu_{BUMP} > \mu_{3DP} > \mu_{ABS}$$

0.81 0.67 0.61

➢ Packing



X-ray imaging

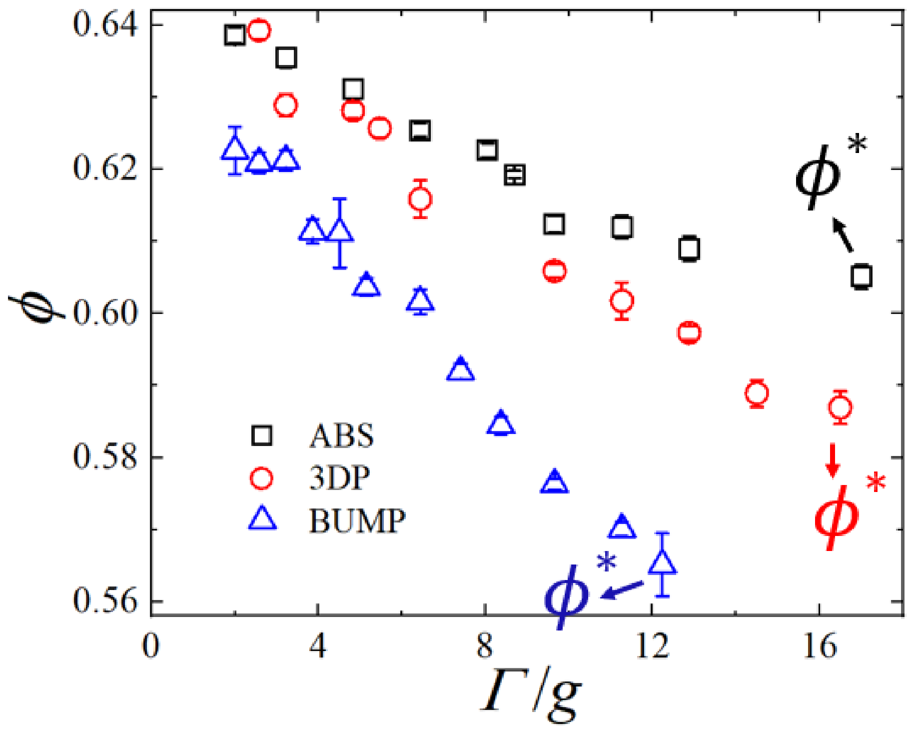


Tap intensity

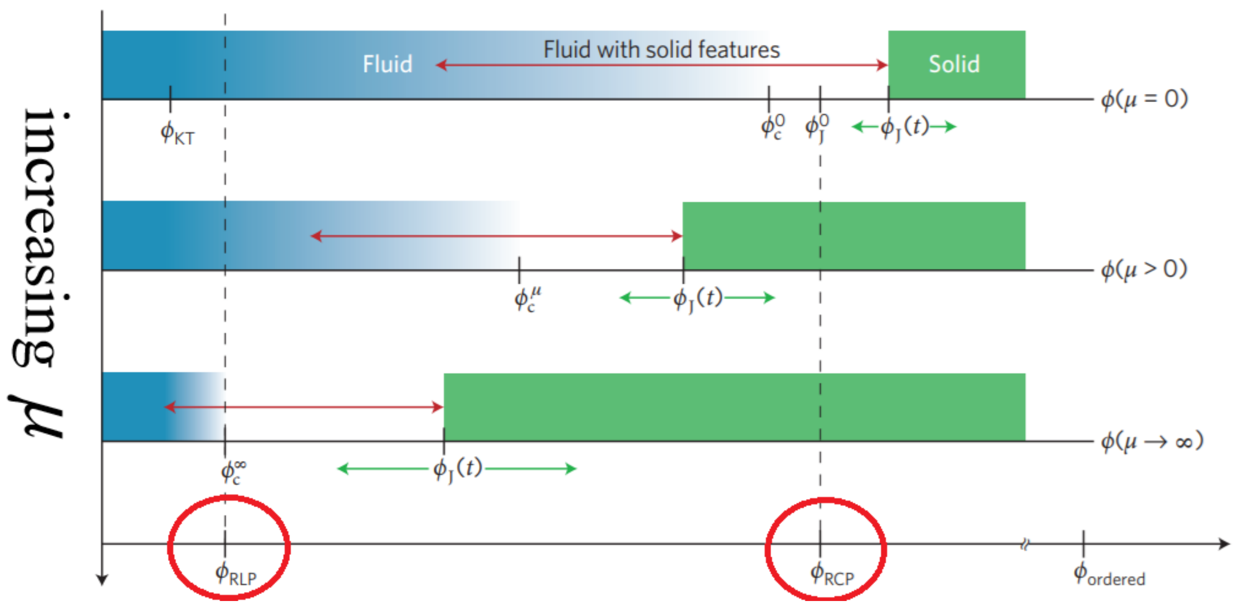
$\Gamma = 2g \sim 16g$

Structure reconstruction

Range of Packing Fraction



Yuan *et al*, *PRL* **127**, 018002 (2021)



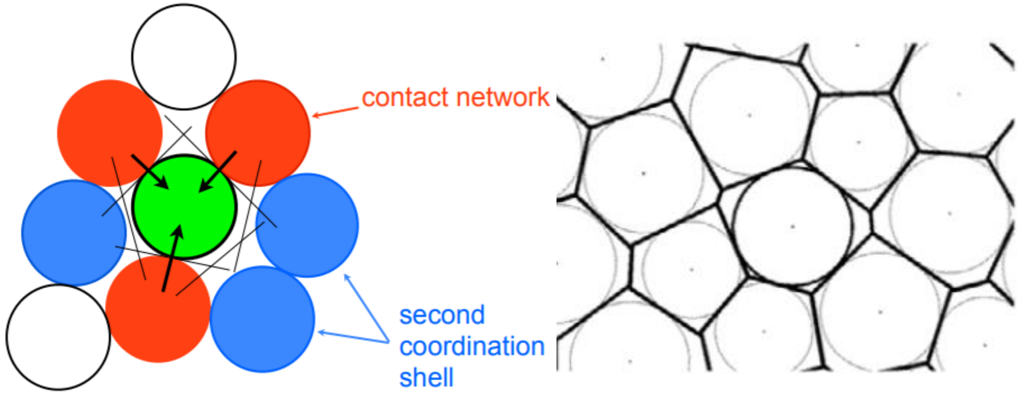
$\phi_{RLP} \approx 0.56$ $\phi_{RCP} \approx 0.64$

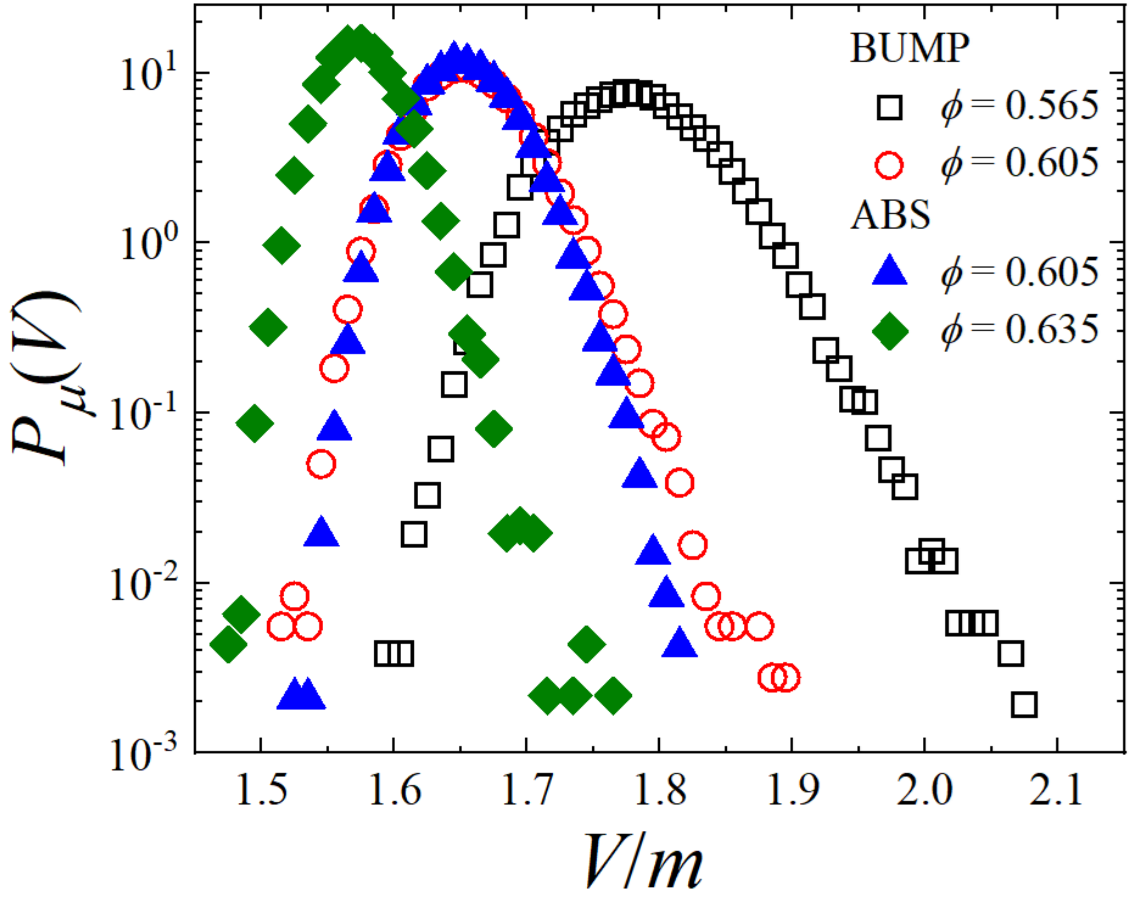
Luding, *Nat. Phys.*, 2016.

- Samples fully cover all **mechanically stable packings** between **random loose packing** (**RLP**) and **random close packing** (**RCP**)
- Γ vs. ϕ is not universal for different beads

Volume distribution

➢ **Voronoi cell**

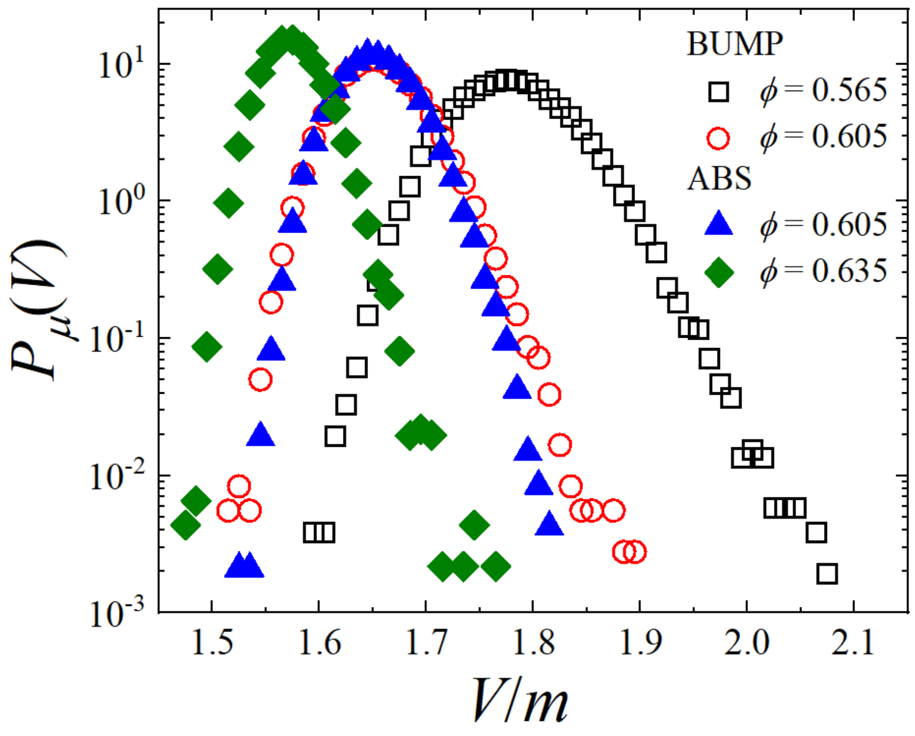


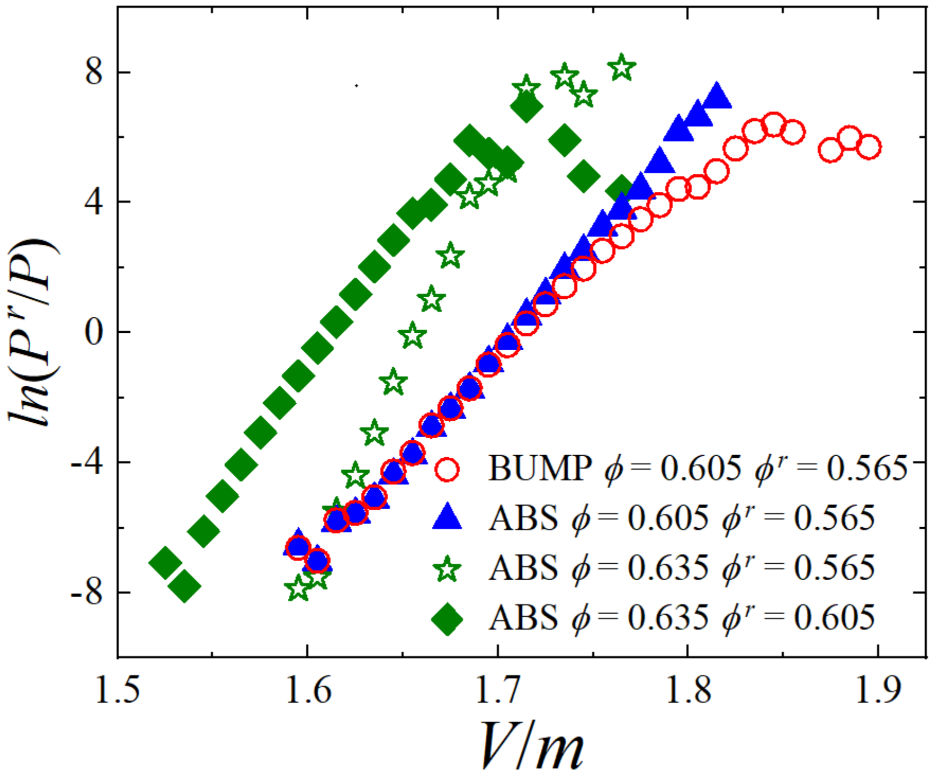


Boltzmann-like distribution of volume

$$P_\mu(V) = \frac{\Omega_\mu(V)}{Z(\chi)} e^{-V/\chi}$$

Overlapping histogram method





Boltzmann distribution of volume

$$P_\mu(V) = \frac{\Omega_\mu(V)}{Z_\mu(\chi)} e^{-V/\chi}$$

$$\frac{P_\mu^r(V)}{P_\mu(V)} = \frac{Z_\mu(\chi)}{Z_\mu(\chi^r)} e^{\left(\frac{1}{\chi} - \frac{1}{\chi^r}\right)V}$$

Volume-independent Partition function

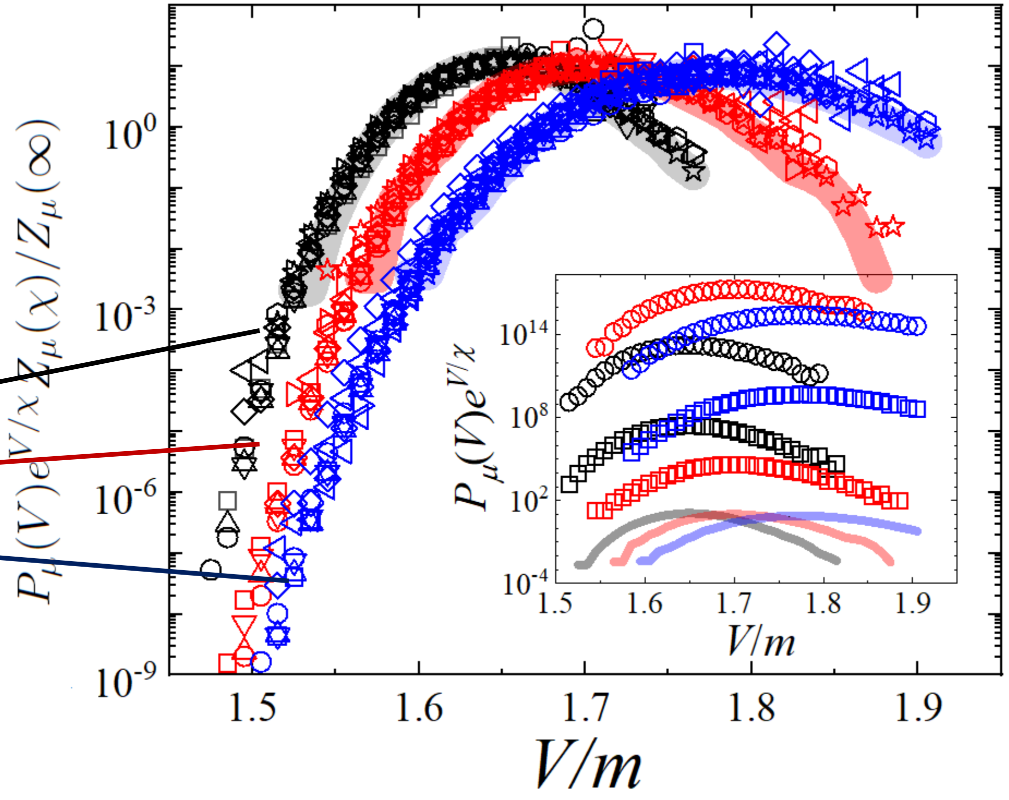
Boltzmann-like distribution

$$P_\mu(V) = \frac{\Omega_\mu(V)}{Z(\chi)} e^{-V/\chi}$$

➢ **Partition function**

$$\Omega_\mu(V) = P_\mu(V) e^{V/\chi} Z(\chi) = P_\mu(\infty) Z(\infty)$$

DOS for packings of three kinds of beads collapse on three curves



Partition function is independent of the volume

Free energy

$$P_\mu(V) = \frac{\Omega_\mu(V)}{\boxed{Z(\chi)}} e^{-V/\chi}$$

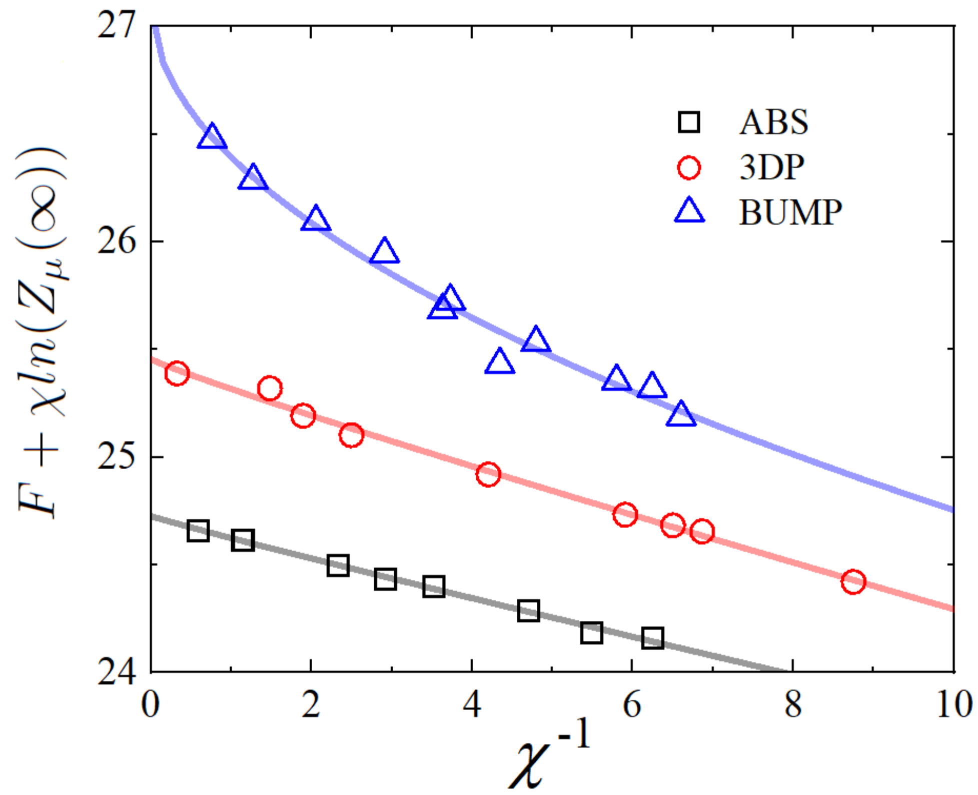
- **Free energy**

($F = -k_B T \ln Z$ **for thermal system**)

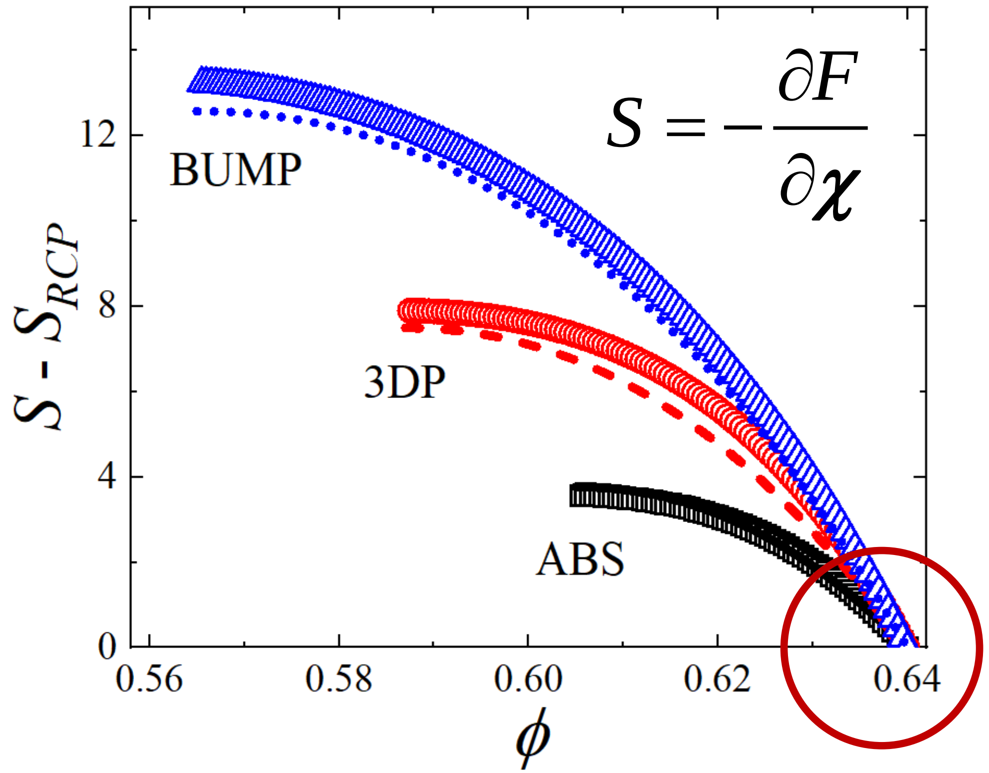
$$F = -\chi \ln\left[Z(\chi)/Z(\infty)\right] - \boxed{\chi \ln Z(\infty)}$$

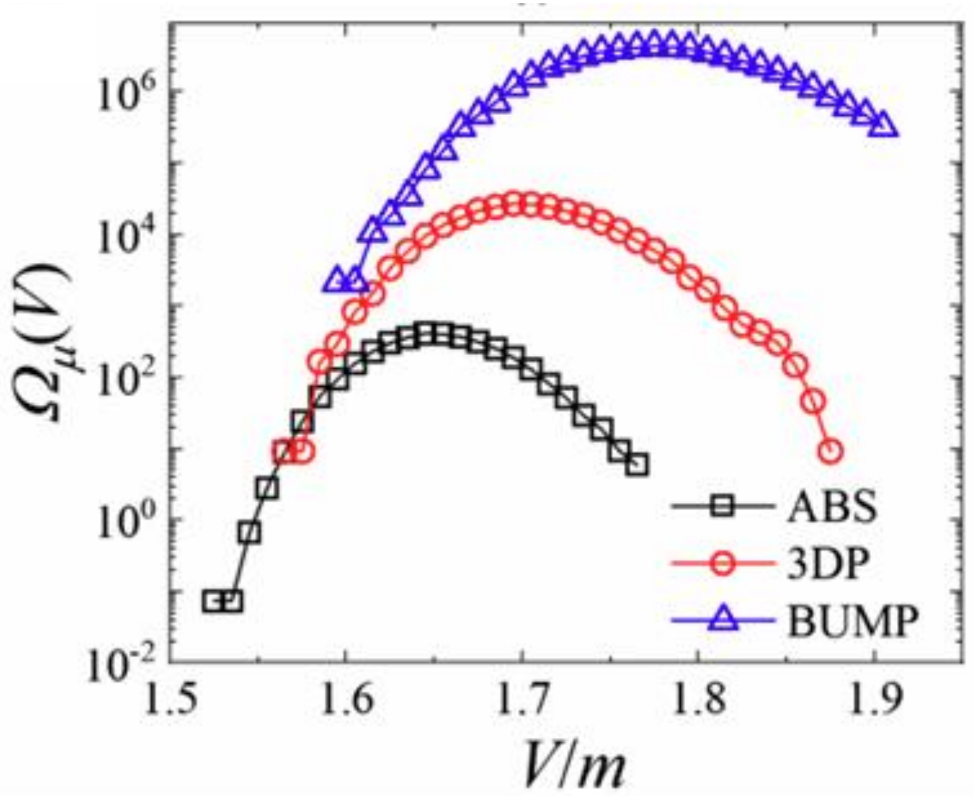
Undetermined Additive constant

where $$\frac{Z(\chi)}{Z(\infty)} = \frac{P^{RLP}(V)}{P(V)} \exp\left[-\frac{V}{\chi}\right]$$



Density of States



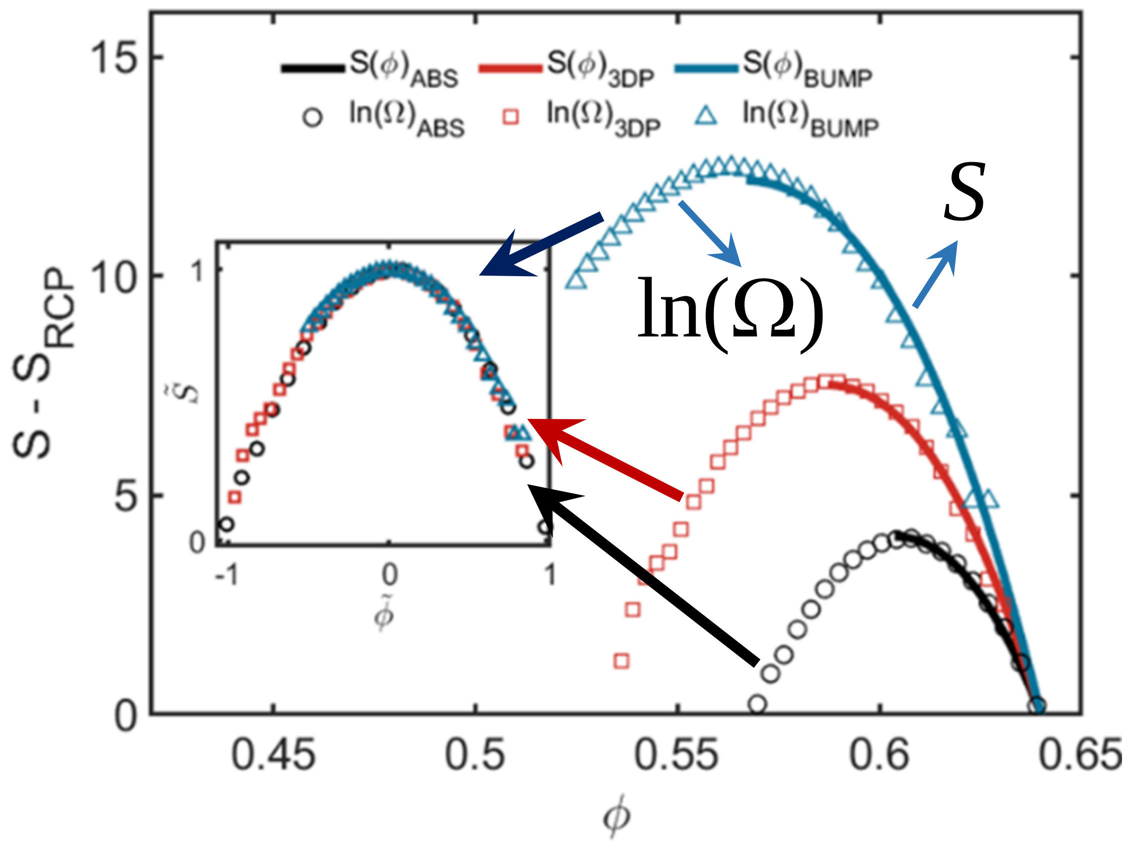


We postulate that $S_{RCP} = 0$ for all three systems, from which $Z_\mu(\infty)$ can be determined.

$$\Omega_\mu(V) = P_\mu^{\chi \to \infty}(V) Z(\infty)$$

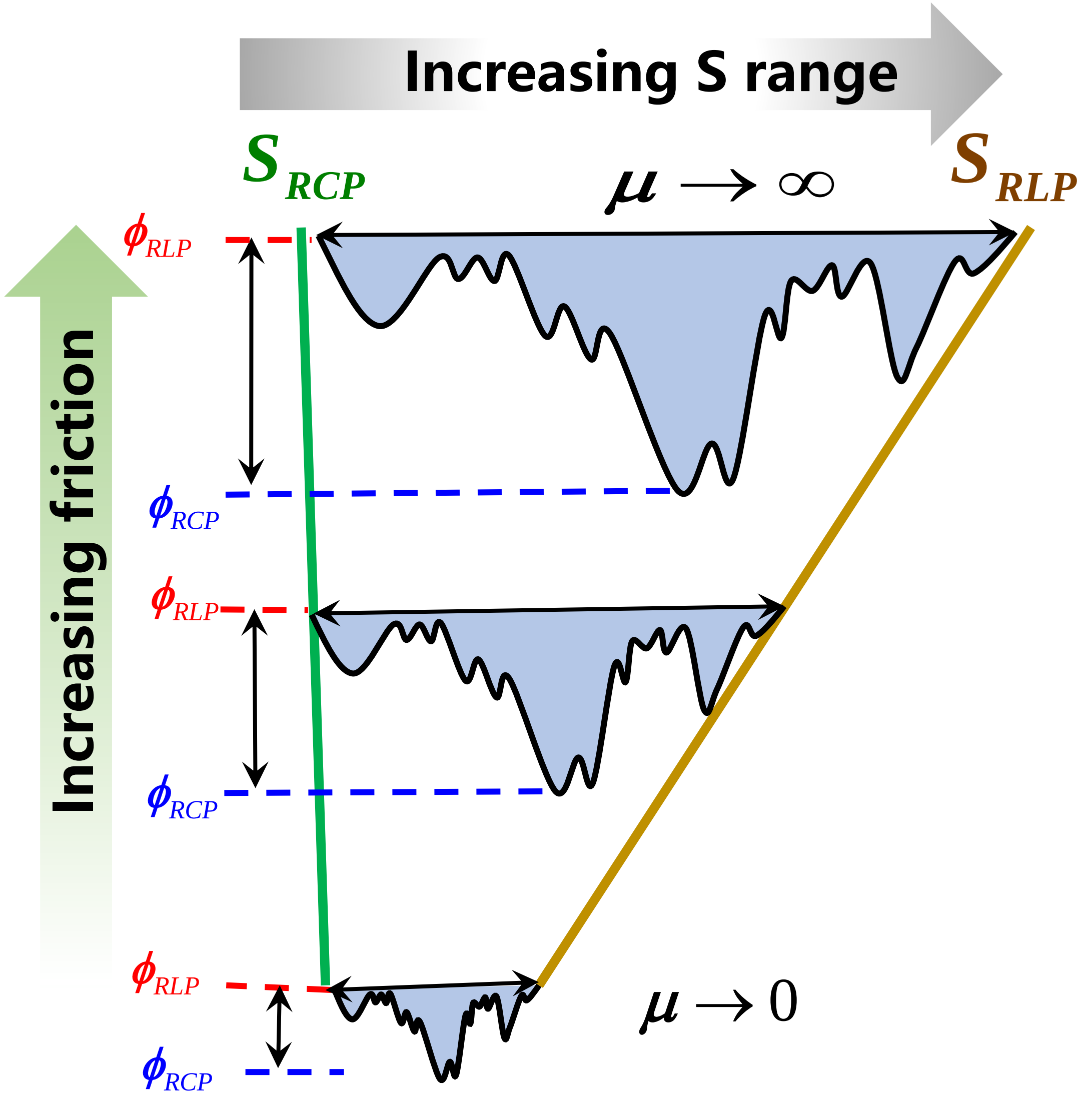
$$S_{RCP} = \frac{\partial F_{RCP}}{\partial \chi_{RCP}} = \ln\left[\frac{Z(\chi_{RCP})}{Z(\infty)}\right] - \ln\left(Z(\infty)\right) = 0$$

Scaling of DOS for different friction systems



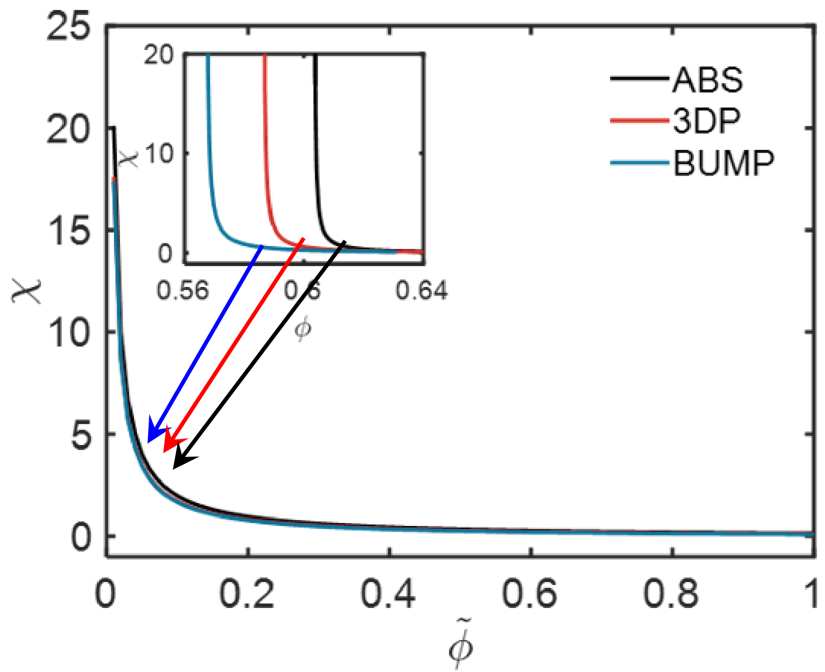
$$\tilde{\phi} = (\phi - \phi_{RLP}) / (\phi_{RCP} - \phi_{RLP})$$

$$S = (S - S_{RCP}) / (S_{RLP} - S_{RCP})$$



mainly due to length scale separation between structure and force

Scaling of state equations of different friction systems



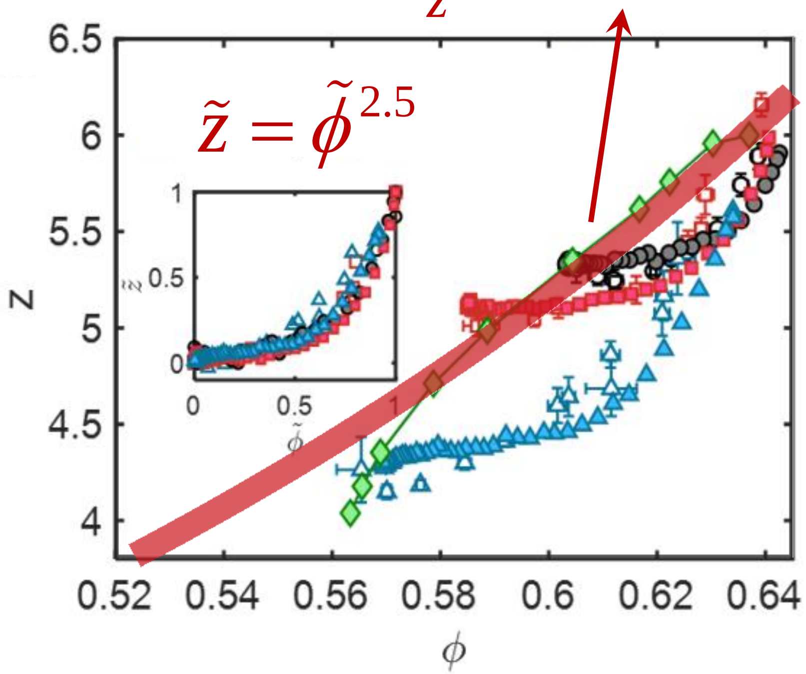
$$\tilde{\phi} = (\phi - \phi_{RLP}) / (\phi_{RCP} - \phi_{RLP})$$

(One-to-one relationship between volume and χ)

ϕ-z relation for isostatic packings

$$V(z) = \frac{z + 2\sqrt{3}}{z} V_g$$

Song, Wang and Makse, *Nature*, 2008



$$\tilde{z} = (z - z_{RLP}) / (z_{RCP} - z_{RLP})$$

Contact number

(Different from previous study where z only depends on ϕ)

Granular and frictionless hard sphere system

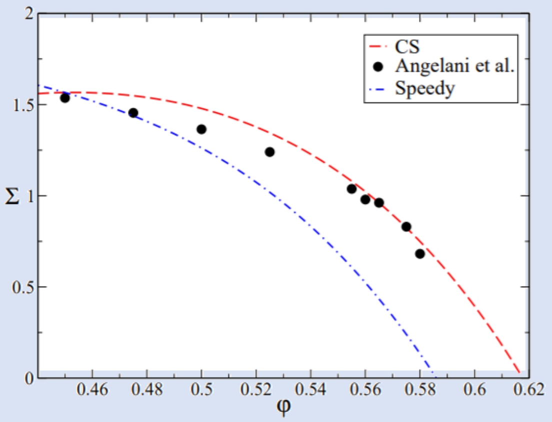
Frictionless hard sphere packing

Frictional granular packing

T_{on} ← ϕ_{RLP}

T_d or T_c ← ϕ_{RCP}

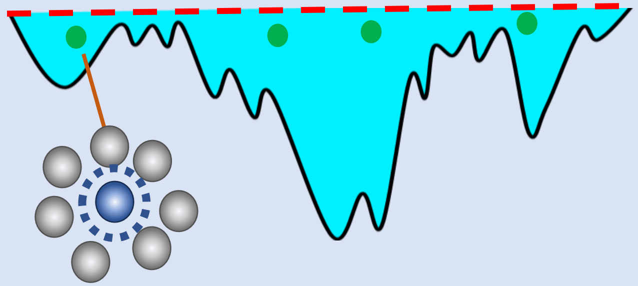
Configurational entropy



Energy landscape

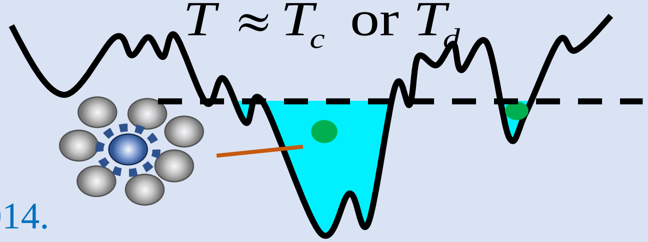
Glass onset temperature

$T \approx T_{on}$



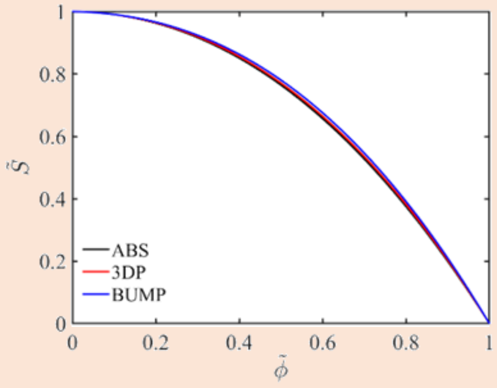
Glass/dynamical transition temperature

$T \approx T_c$ or T_d



Barrat *et al*, *PRL*, 2000.
Parisi *et al*, *RMP*, 2010.
Charbonneau *et al*, *Nat. Commun.*, 2014.

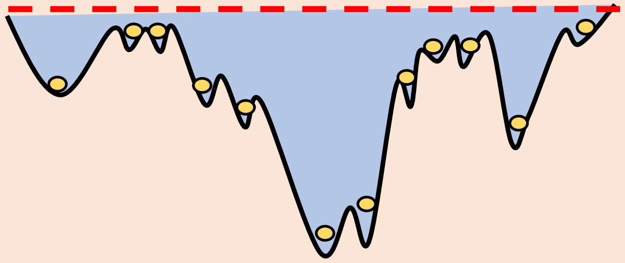
Edwards entropy



Energy landscape

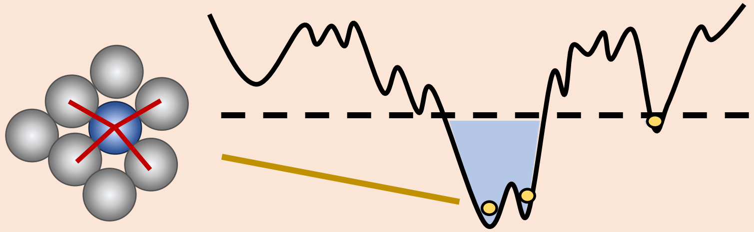
RLP

ϕ_{RLP} corresponds T_{on}



RCP

ϕ_{RCP} corresponds T_d or T_c



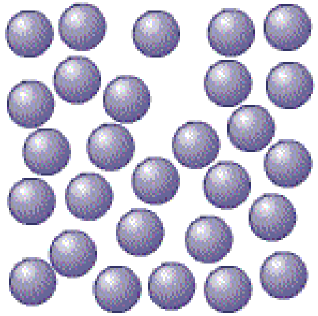
Equation of state

- The Carnahan-Starling equation of state for the fluid phase of the hard sphere model.

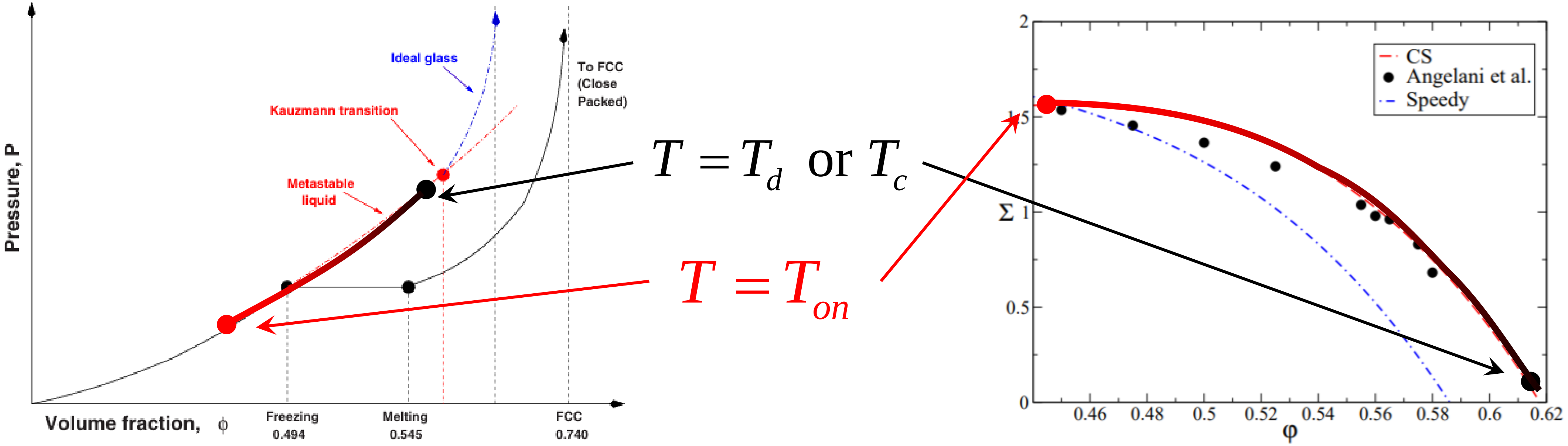
Compressibility factor

$$Z = \frac{V}{Nk_B}\boxed{\frac{p}{T}} = \frac{1+\phi+\phi^2-\phi^3}{(1-\phi)^3}$$

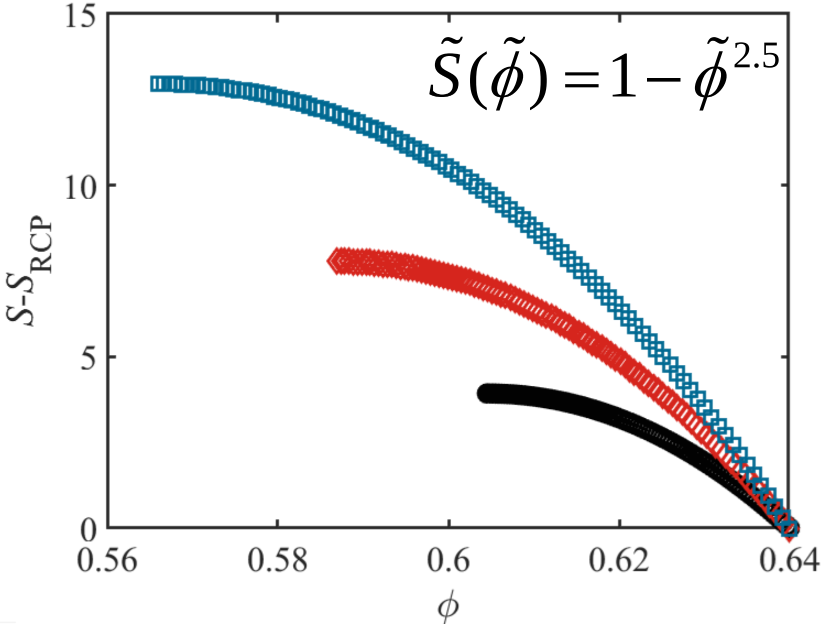
$= \chi$ in Edwards ensemble



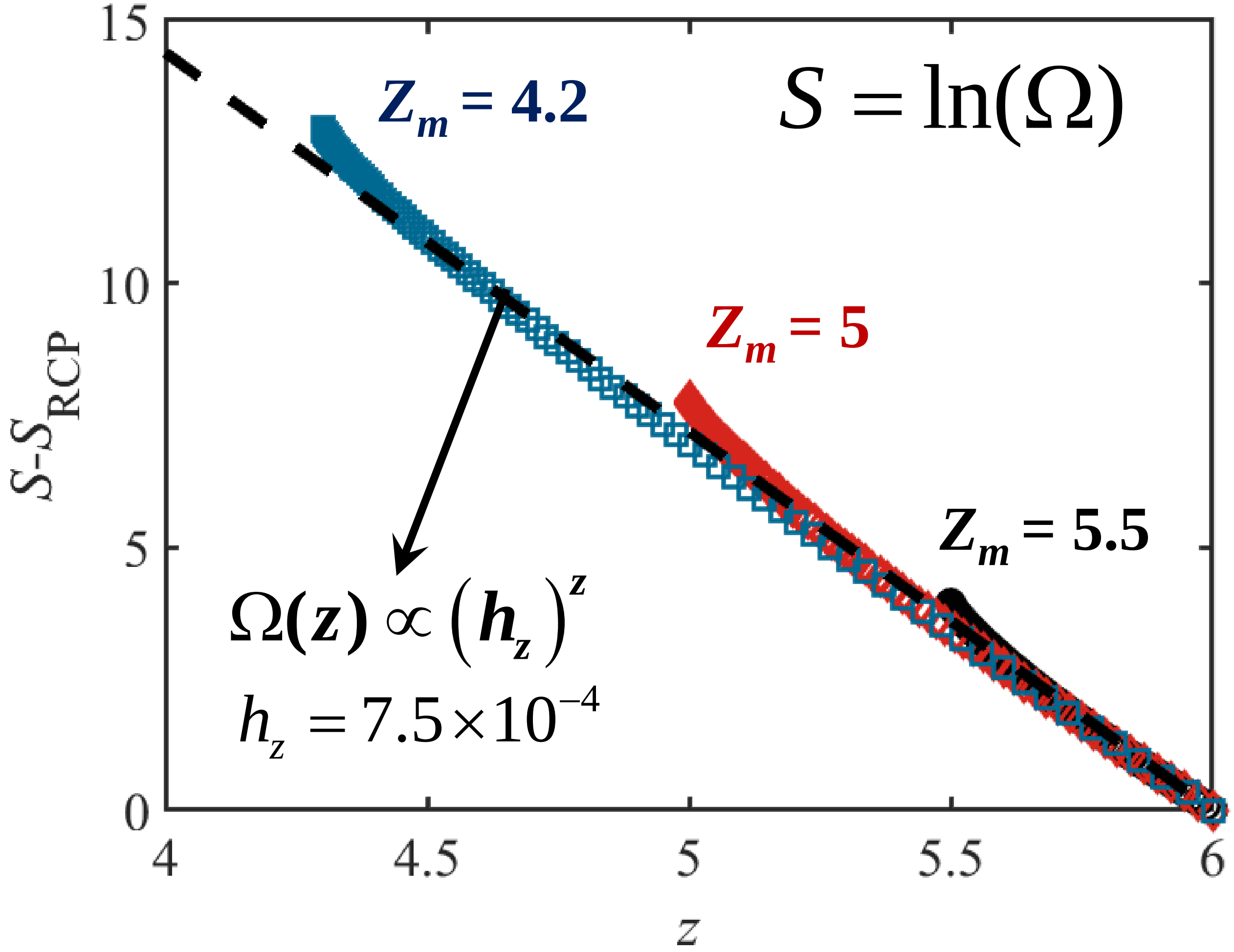
Hard sphere liquid



Density of State of different friction systems



$\tilde{z} = \tilde{\phi}^{2.5}$



$$Q(\chi, Z) = \int_{Z_m}^{6} \Omega(z) e^{-V(z)/\chi} dz$$

$$\Omega(z) = \frac{1}{(h_z)^{D-z}} \propto (h_z)^z = A\exp(z)$$

h_z: typical distance between jammed configurations in the space, $h_z << 1$

Song et al., *Nature*, 2008.

Entropy depends almost linearly on contact number z, which supports that scaling factor is controlled by friction

Comparison of the partition functions

➢ Partition function for jammed states

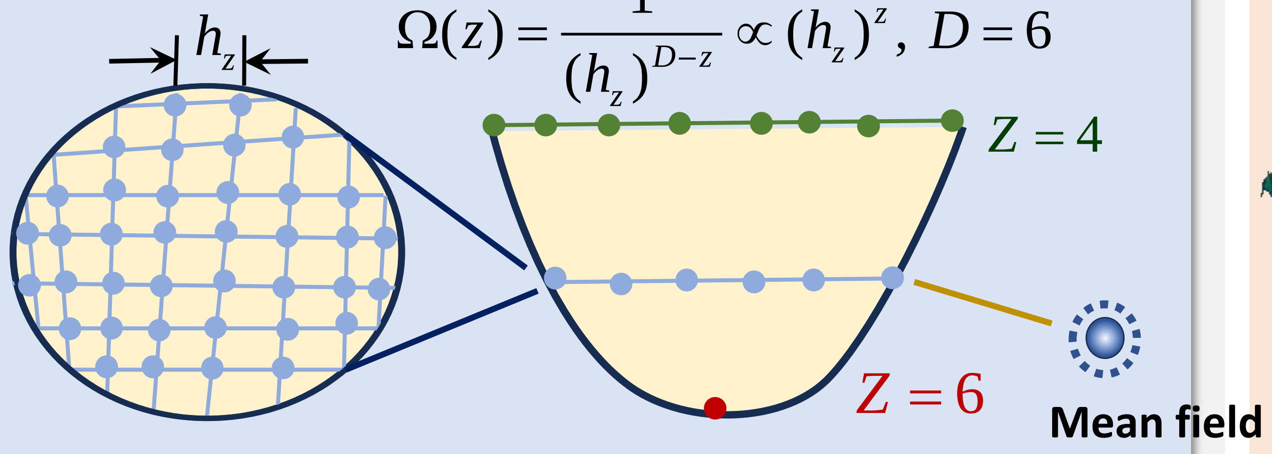
$$Z = \int dV \Omega(V) e^{-V/\chi} \Theta_{jam}$$

0 or 1, condition of jamming

Consider the contact number associated with mechanical stability

$$Z = \int \Omega(z) e^{-V(z)/\chi} dz \qquad V(z) = \frac{z + 2\sqrt{3}}{z} V_g$$

$$\Omega(z) = \frac{1}{(h_z)^{D-z}} \propto (h_z)^z, \; D = 6$$



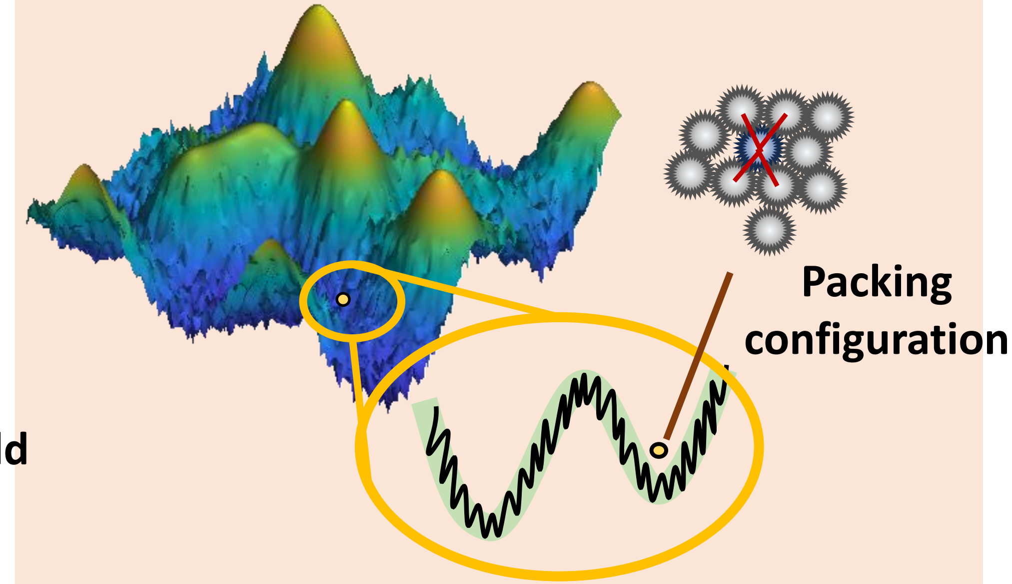
$h_z \ll 1$ A small constant analogous to Planck constant

Song, Wang and Makse, *Nature*, 2008

➢ Partition function in our work

$$Z(\chi) = \int \Omega_{\mu}(V) e^{-V/\chi} dV$$

DOS of jammed packings which inherit the configuration of high-T hard-sphere liquid

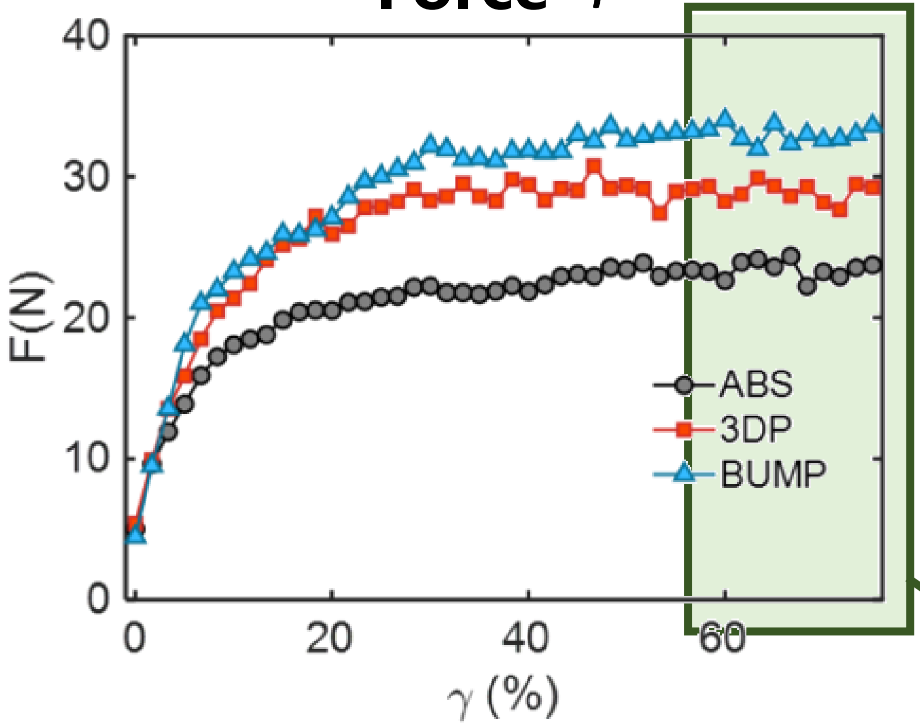


Origin of critical state

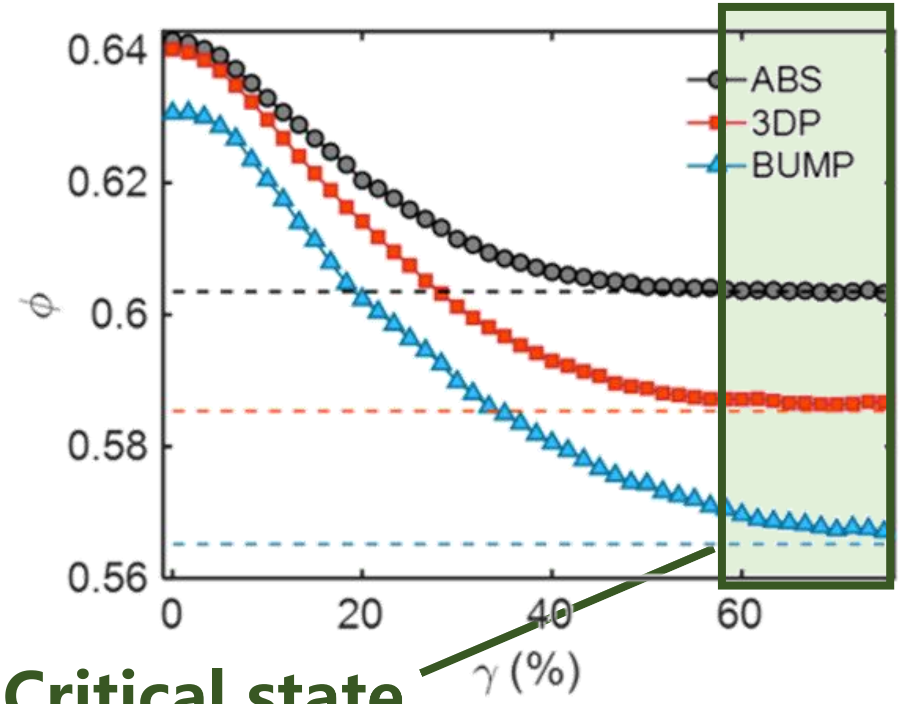
Critical state

Granular materials, if continuously sheared, **flow as a frictional fluid**, would eventually reach a well-defined **critical state.** At the critical state, the shear stress and the volume fraction reach steady state values.

Force F



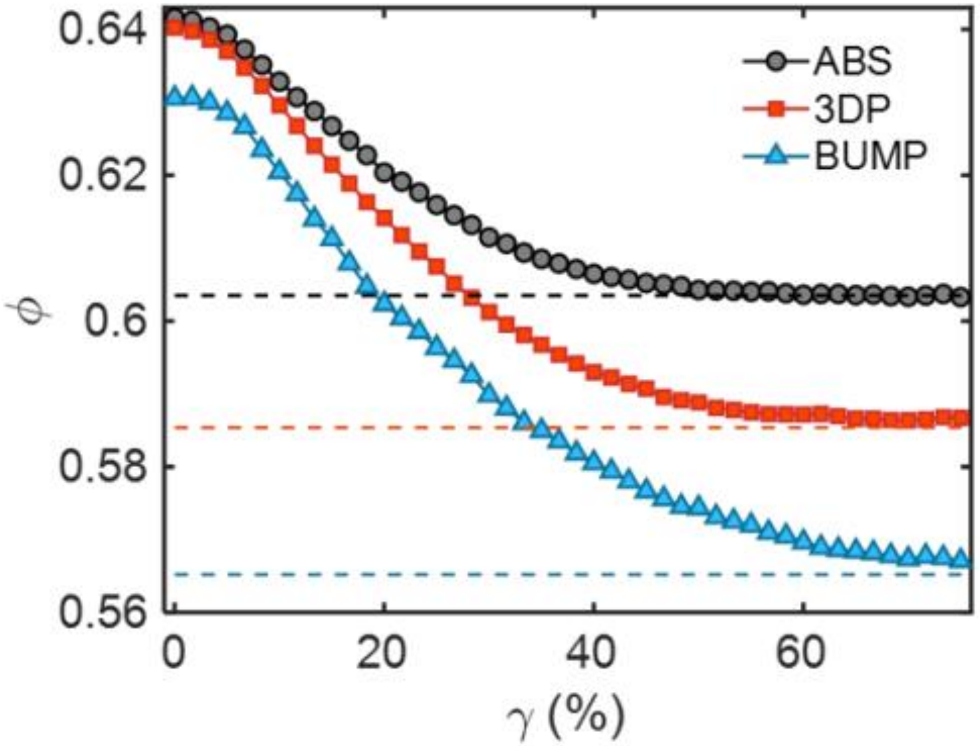
Volume fraction ϕ



Critical state

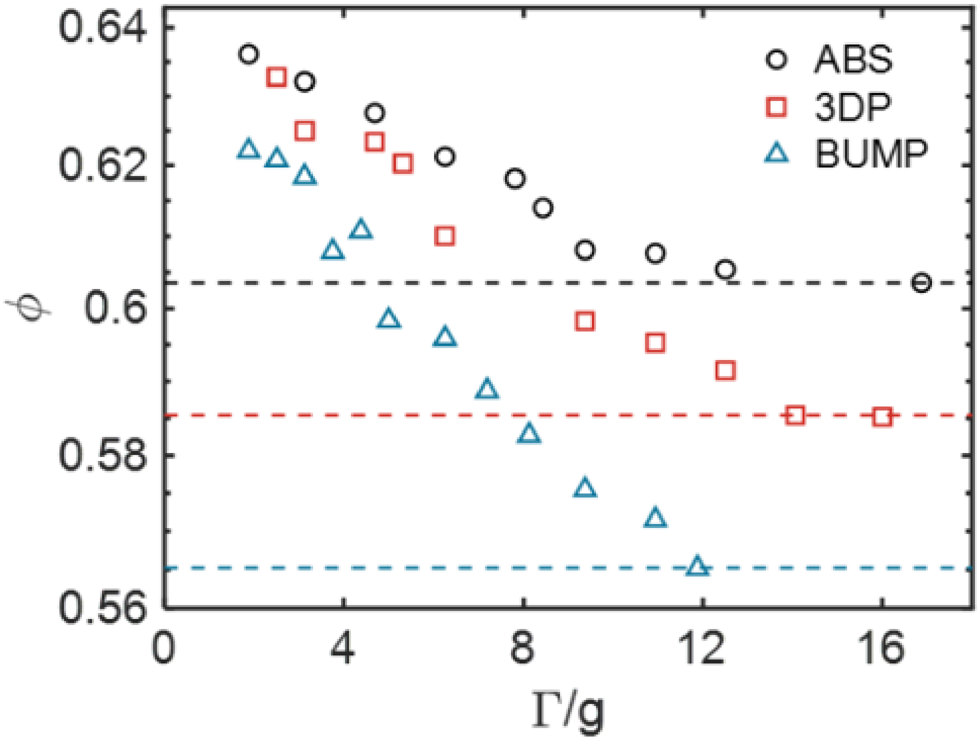
Critical state and RLP

Critical state of sheared system



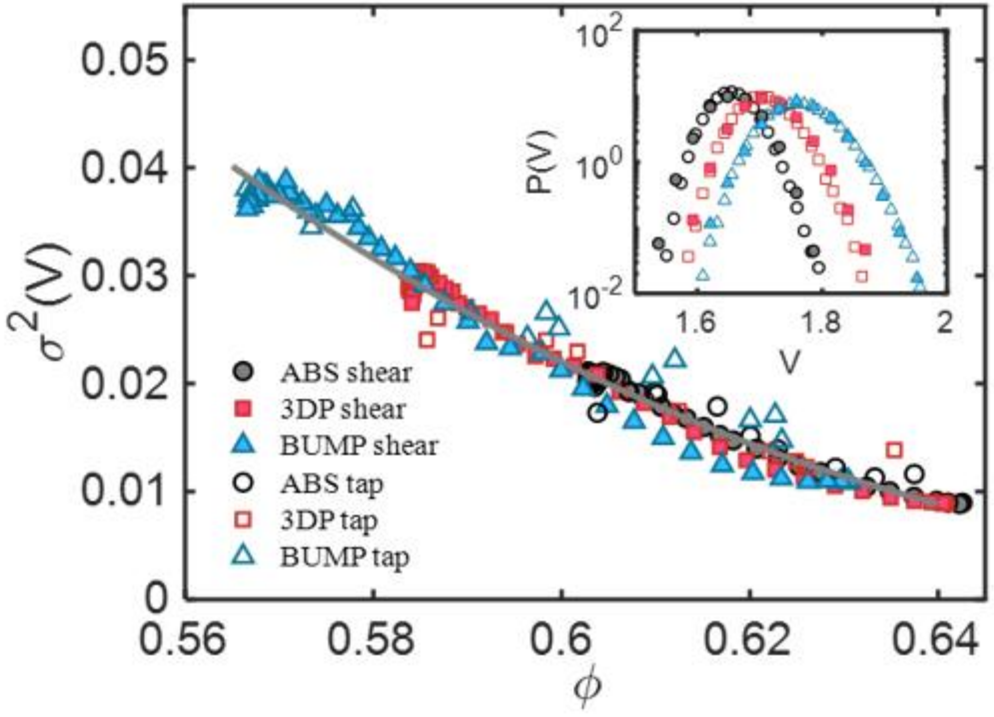
The same ϕ

RLP of same system prepared by tapping



Volume fraction of the critical state of the sheared system are exactly the same as the volume fraction of the random loose packing of the same system prepared under tapping.

Thermodynamic understanding of critical state



Identical volume distributions

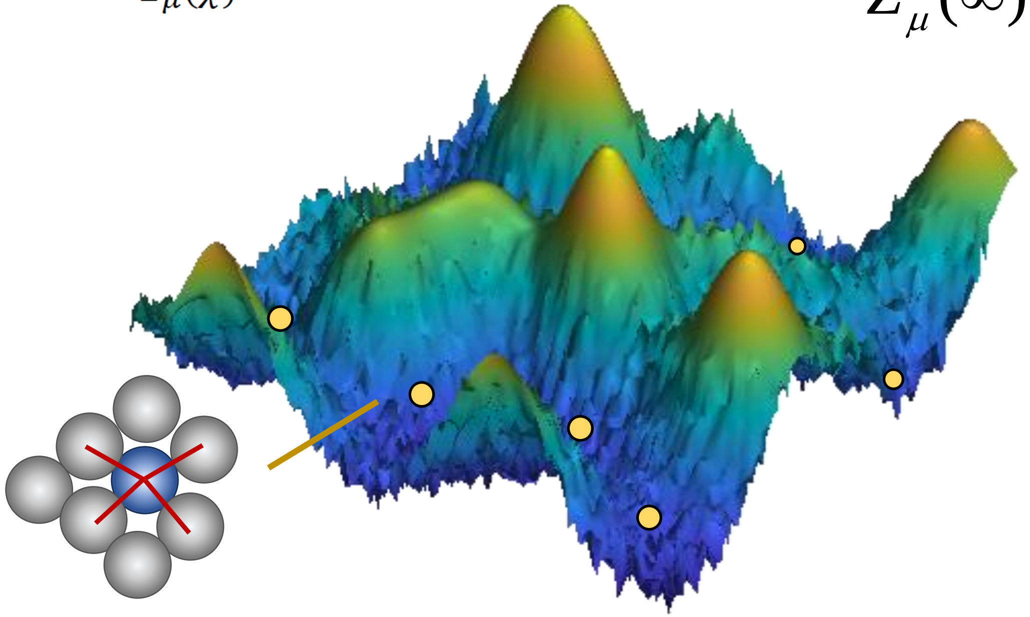
equivalence of thermodynamic states

Critical state = RLP

RLP & Critical state:

$\chi \to \infty$

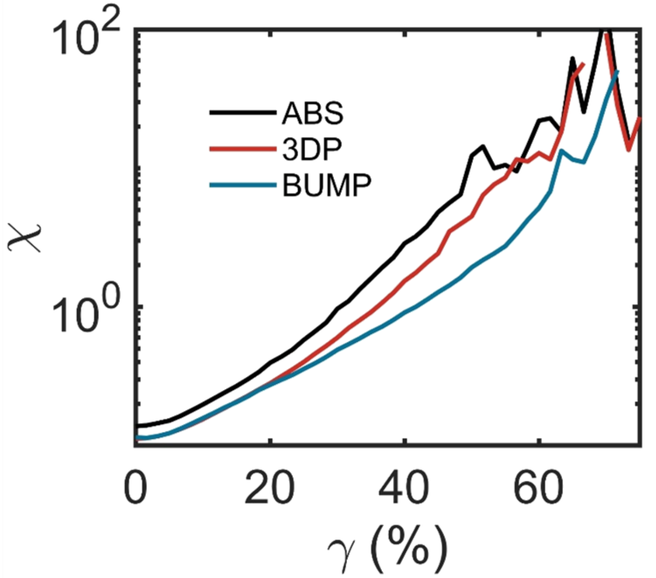
$$P_\mu(V) = \frac{\Omega_\mu(V)}{Z_\mu(\chi)} e^{-V/\chi} \quad \Rightarrow \quad P_\mu(V) = \frac{\Omega_\mu(V)}{Z_\mu(\infty)}$$

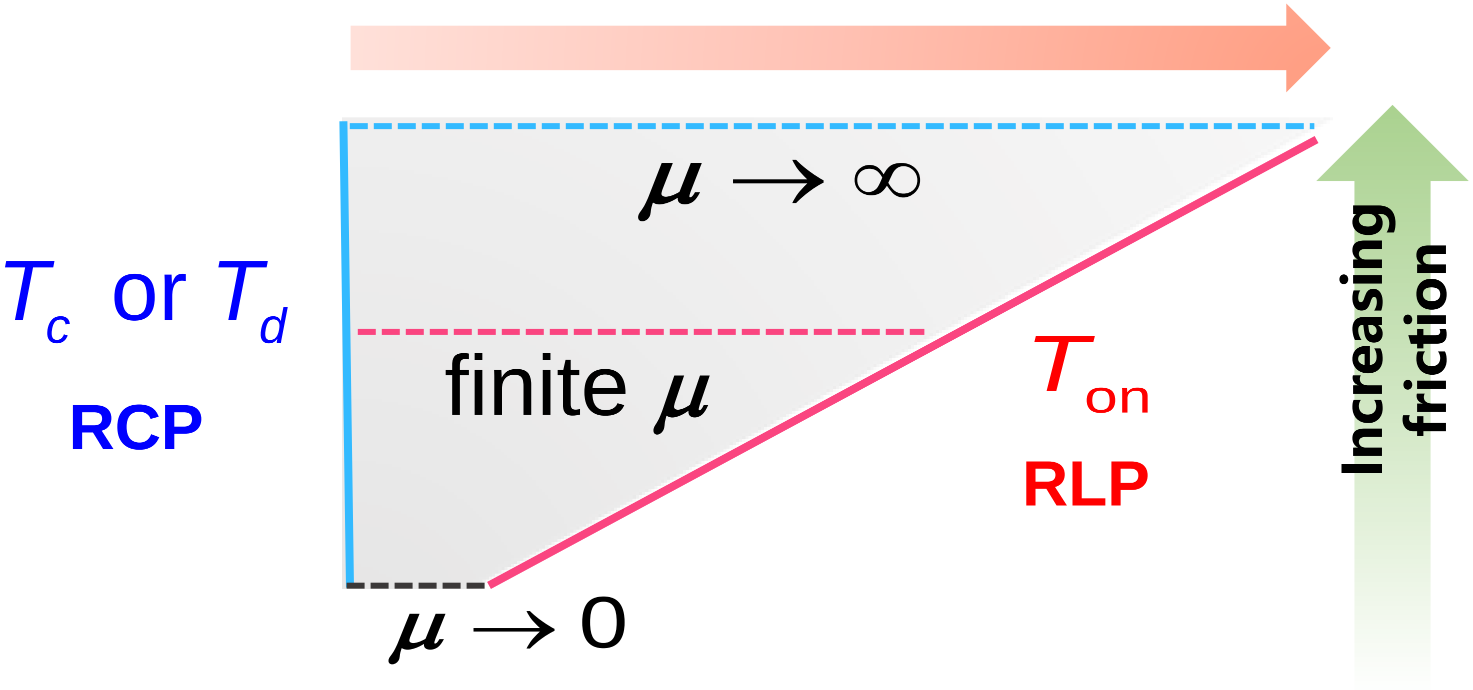


All mechanically stable states are sampled with equal probability

Origin of shear dilatancy of granular materials

Shear induced rejuvenation of the glass

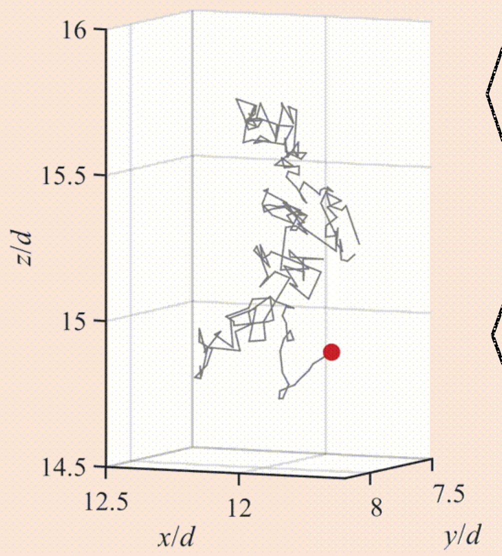




For **large friction systems**, the difference between ϕ_{RCP} and ϕ_{RLP} is larger, which leads to **more significant shear dilatancy effect**, which should be very **small** for **frictionless systems**.

Fluctuation-Dissipation temperature

➢ Fluctuation-Dissipation



$$\left\langle [x(t)-x(0)]^2 \right\rangle = 2Dt$$

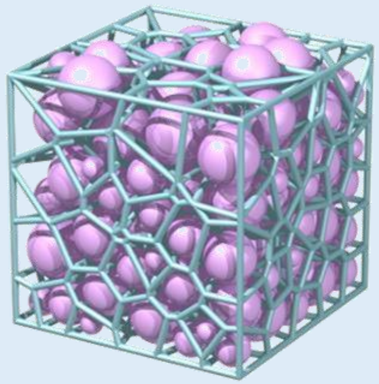
Diffusion

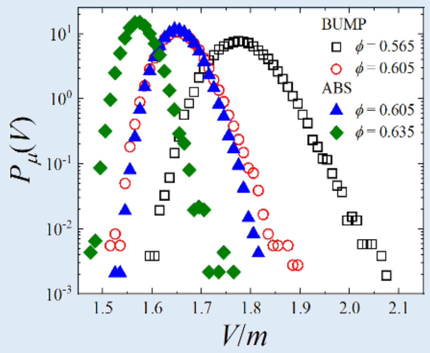
$$\left\langle x(t)-x(0) \right\rangle = BFt$$

Directed motion

$$T_{FDT} = \frac{D}{B}$$

➢ Edwards Ensemble





$$\frac{P_\mu^r(V)}{P_\mu(V)} = \frac{Z_\mu(\chi)}{Z_\mu(\chi^r)} e^{\left(\frac{1}{\chi}-\frac{1}{\chi^r}\right)V}$$

$$\chi$$

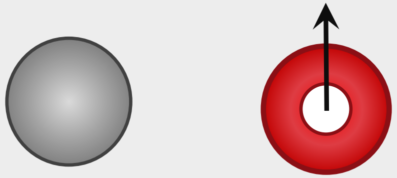
Relationship：？

Experimental setup

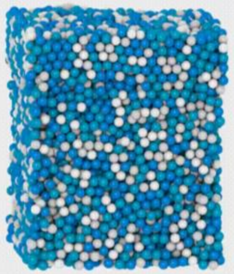
- **We 3D print solid background particles and hollow tracers.**

"Buoyancy"

$F \sim \Delta mg$

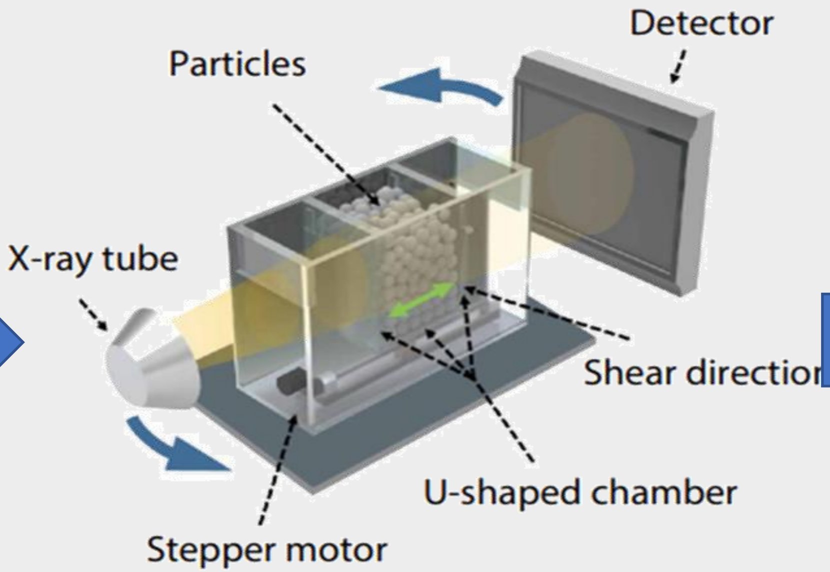


Background particle   Hollow tracer

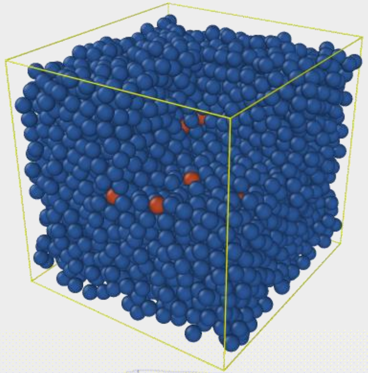


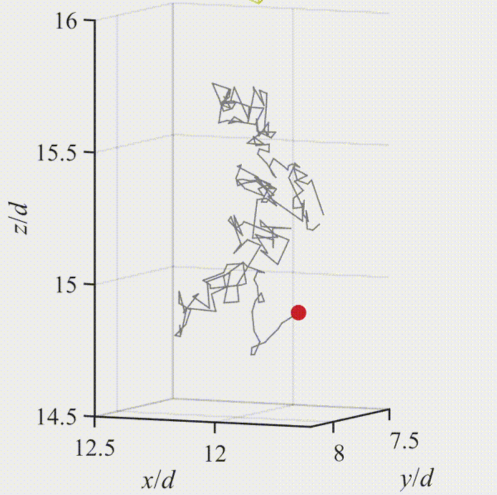
Cyclic shear as a "heat bath"

- **One CT scan after every 10 shear cycles**

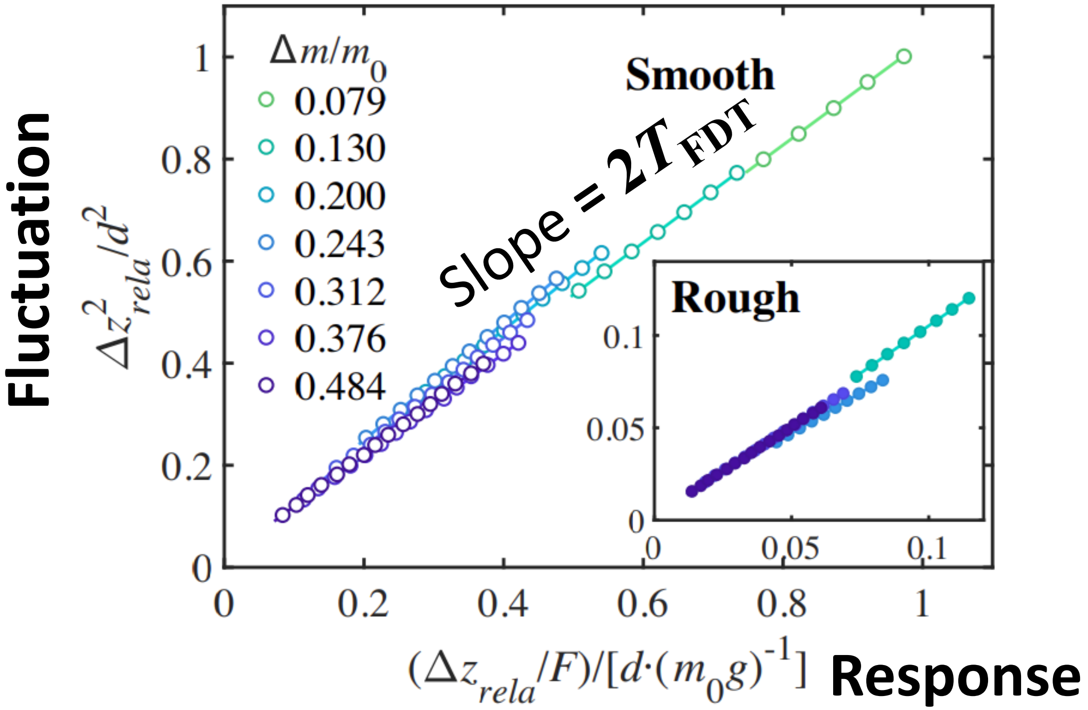


- **Obtaining** **tracers' trajectories** **and packing** **structures**





Fluctuation-Dissipation Temperature

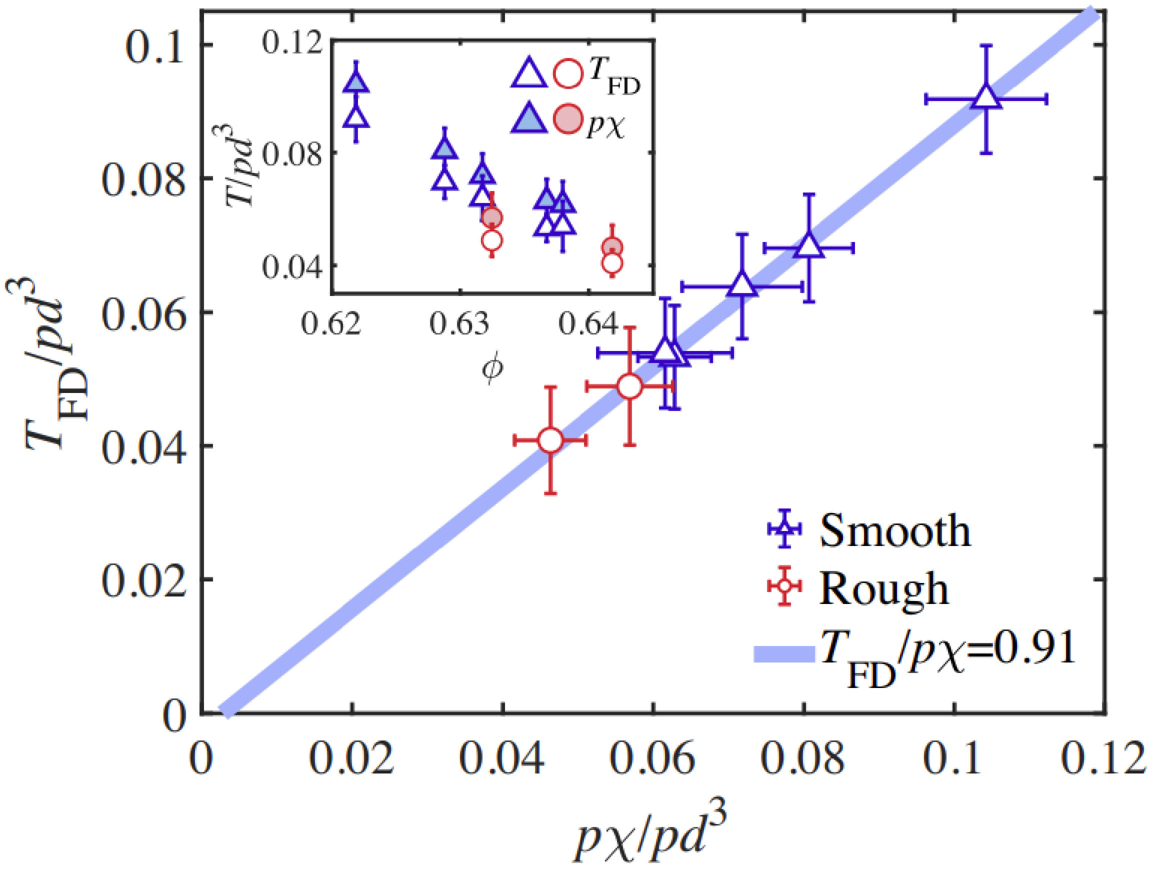


$$T_{FD} = \frac{D}{B} = \frac{\mathrm{MSD}(\Delta t)}{2\Delta z(\Delta t)/F}$$

For different tracers, we always obtain the same fluctuation-dissipation temperature, which suggest that it is an intrinsic property of the entire granular system, instead of the tracer used.

Comparison between T_{FDT} and χ

In all cases, Edwards temperature equals the fluctuation-dissipation temperature, within an error of 10%.



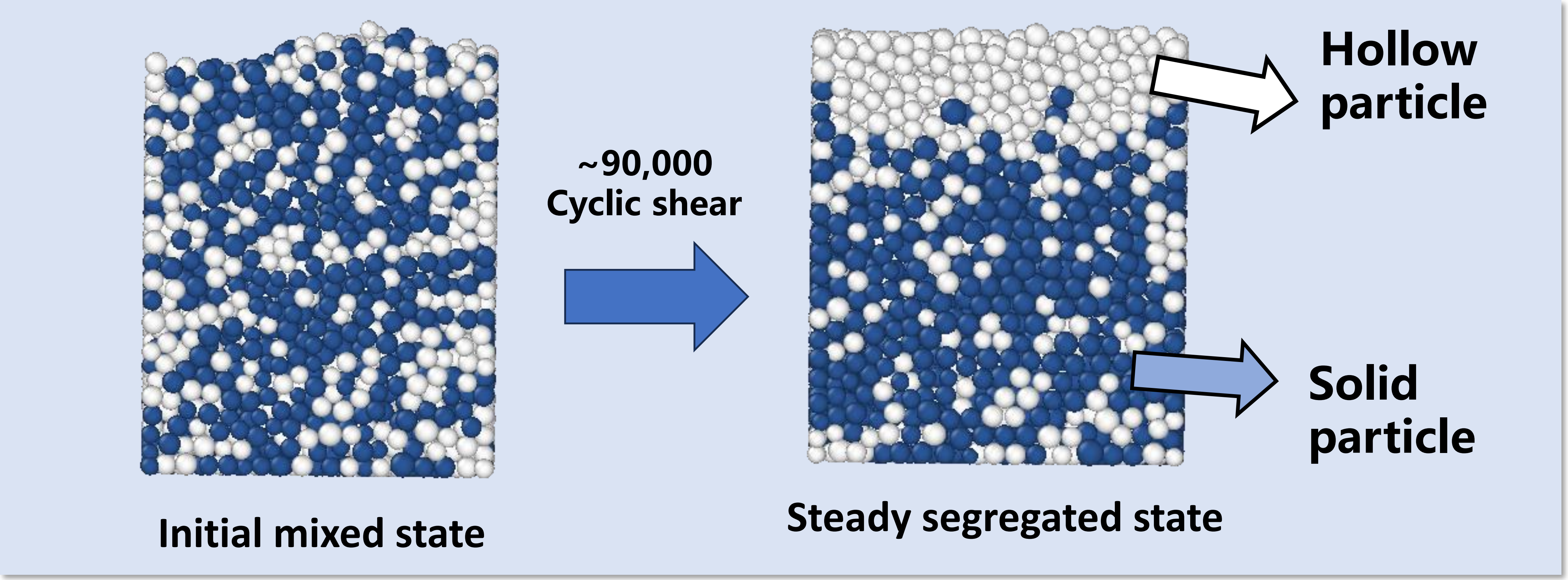
Edwards Compactivity χ

$$T_{FD} / p\chi = 0.91 \pm 0.12$$

FD Temperature T_{FDT}

Segregation process of different densities particles

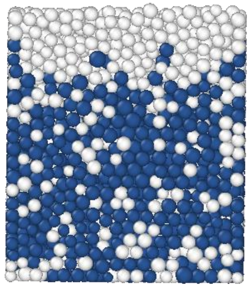
- A 50:50 mixture of hollow particles (HP) and solid particles (SP) ($\Delta m/m_{sol} = 0.5$) of the same diameter (*D*=7mm) and polydisperse (8%).



Dynamics of density segregation

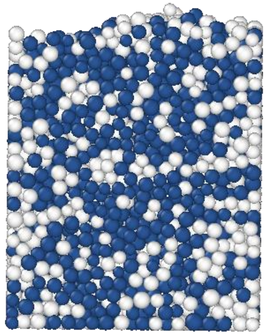
Degree of segregation:

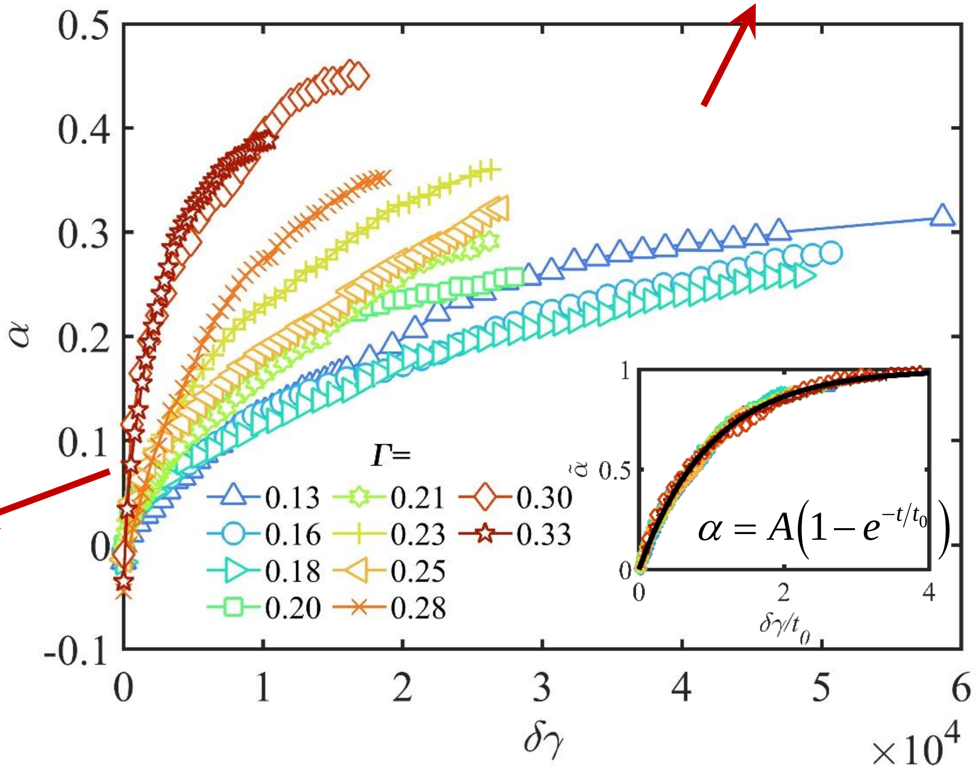
$$\alpha = \frac{2\left(\bar{H}_{\mathrm{h}} - \bar{H}_{\mathrm{s}}\right)}{H_{max}}$$



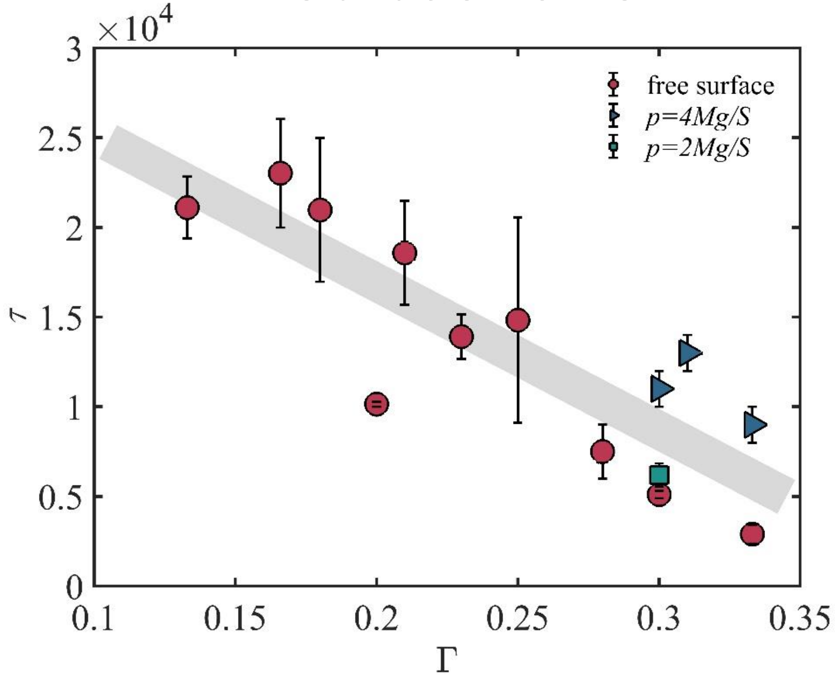
Steady state

Initial state





Relaxation time



Height distribution of particles

➢ **Thermodynamics**

Minimizing the free energy

$$\delta F = \delta E - \delta(T_{seg} S) \equiv 0$$

Constraint condition $\delta n_s(H) = -\delta n_h(H)$

$$\frac{n_h(H)}{n_s(H)} = \exp\left(-\frac{\Delta mgH}{2T_{seg}}\right)$$

Particle number $N_h = \int n_h(H)dH \quad N_s = \int n_s(H)dH$

Particle energy $E = \int m_h gHn_h(H)dH + \int m_s gHn_s(H)dH$

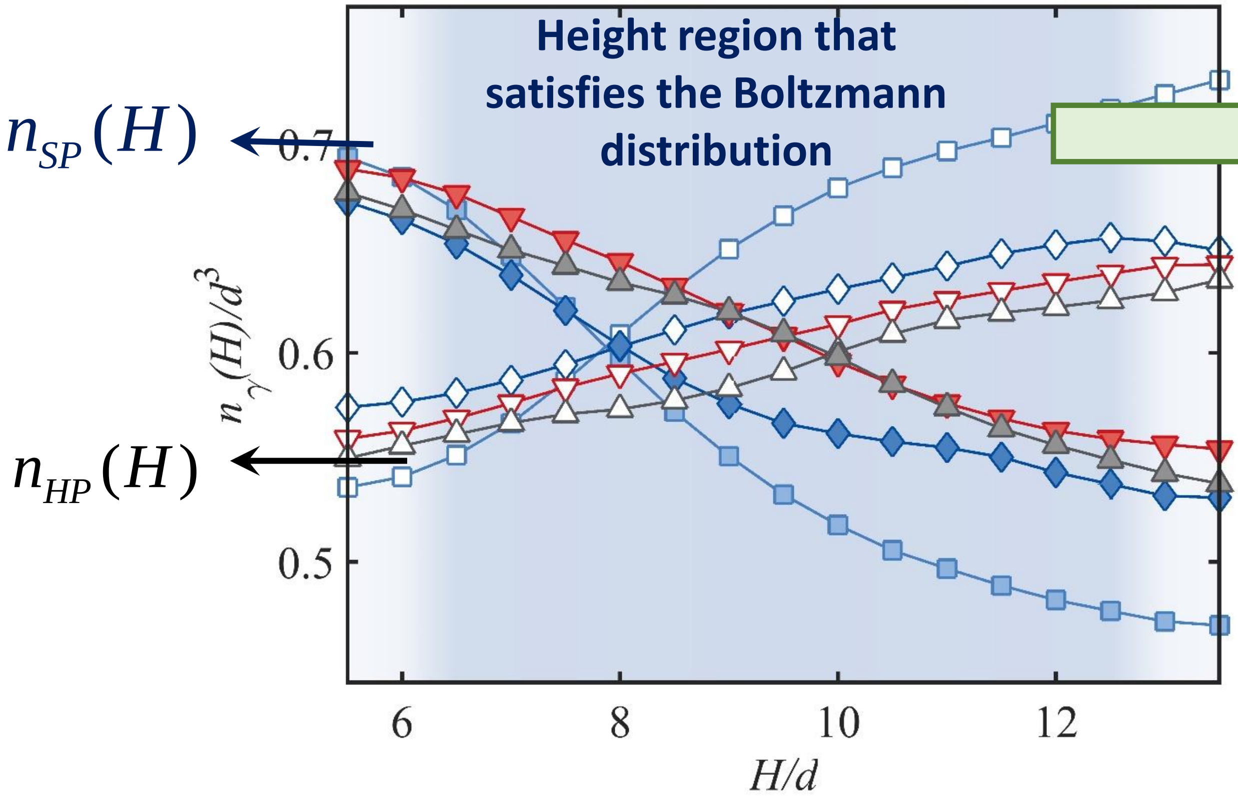
Entropy $S = -2\int n_s(H)\ln n_s(H)dH - 2\int n_h(H)\ln n_h(H)dH$

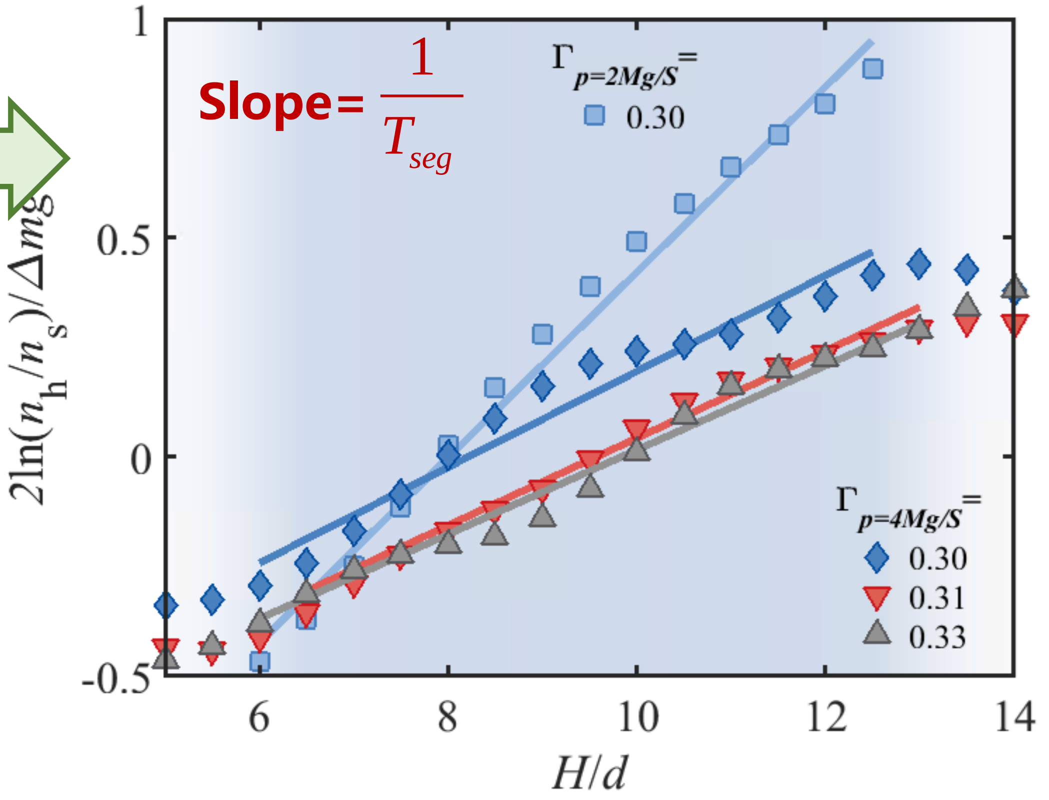
Height distribution of particles

➢ **Confined system covered with lid: uniform pressure p inside**

$n_{SP}(H)$、$n_{HP}(H)$: **The number distributions of hollow and solid beads along different heights.**

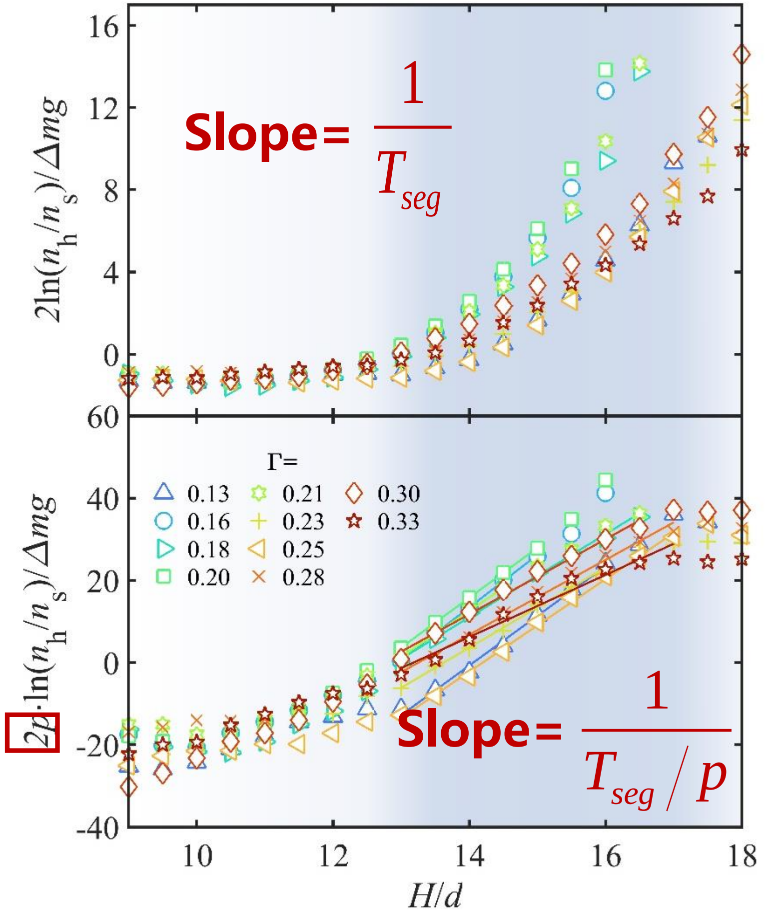
$$\frac{\Delta mgH}{2T_{seg}} = \ln\left(\frac{n_h(H)}{n_s(H)}\right)$$





Height distribution of particles

- **Free surface system: hydrostatic pressure** $p \propto (H_{max} - H)$



No linear relationship

$$\frac{\Delta mgH}{2T_{seg}} = \ln\left(\frac{n_h(H)}{n_s(H)}\right)$$

Different T_{seg} at different height

linear regime

$$\frac{\Delta mgH}{2T_{seg}/p} = p \cdot \ln\left(\frac{n_h(H)}{n_s(H)}\right)$$

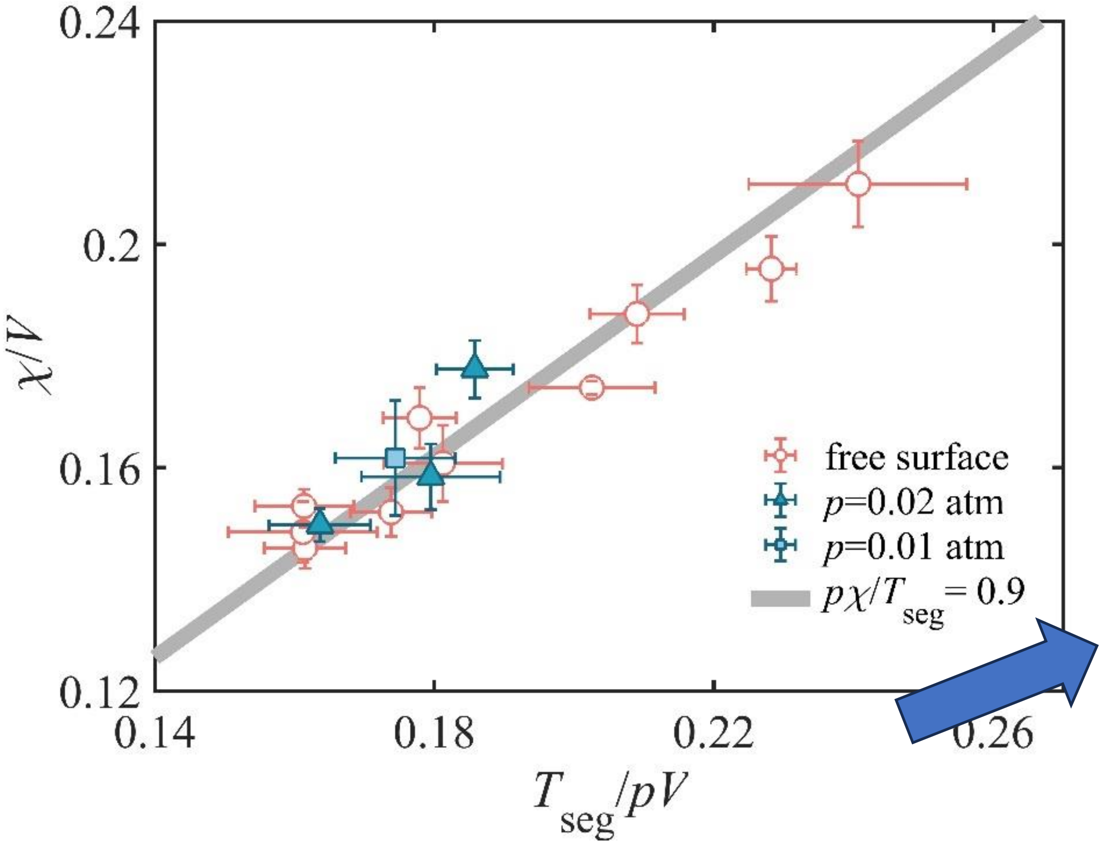
Identical T_{seg}/p at different height

In Boltzmann regime, different heights of the system have identical T_{seg}/p instead of T_{seg}.

Comparison of the two temperature

χ

From the volume fluctuation



T_{seg}/p

From the height distribution

Thermodynamic free energy for granular materials

- As in free surface systems, different heights of the system equilibrate at identical T_{seg}/p or χ .

- Free energy in the traditional thermodynamic framework

$$\delta F = \delta E - \delta(T_{seg} S) \equiv 0$$

$$F = E + pV - TS$$

- Free energy in granular thermodynamic framework

$$\delta F = \delta \frac{E}{p} - \delta(\chi_{seg} S) \equiv 0$$

$$F = E/p + V - \chi S$$

- Edwards thermodynamics with real energy term works, It is reasonable to anticipate that additional terms, e.g., elastic energy and chemical potential, can be included in this general granular thermodynamic framework.

Short Summary

- For 3D granular packings under mechanical tapping, we experimentally test the validity of Edwards volume ensemble.

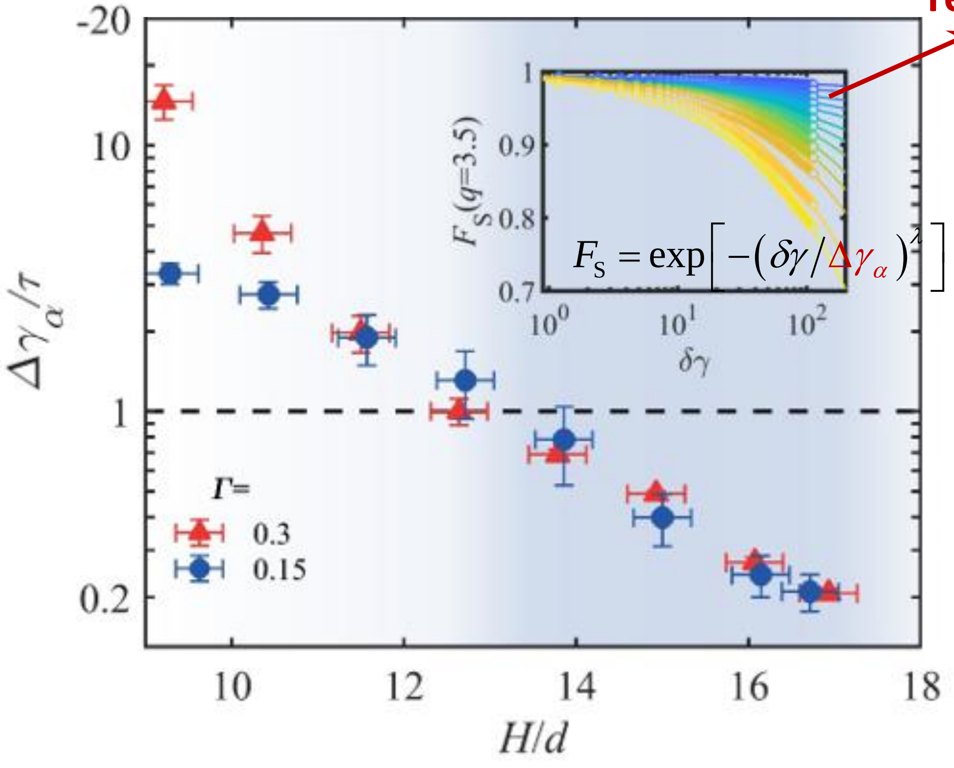
- We give the thermodynamics understanding of critical state and shear dilatancy for sheared granular materials and indicate the unification of frictional granular and frictionless hard-sphere systems.

- Finally, for a 3D granular system under cyclic shear, we experimentally calculated the effective temperature via fluctuation-dissipation theorem, which agrees with compactivity based on Edwards volume ensemble. The density segregation process of granular mixtures can also be understood within the Edwards thermodynamic framework.

Boltzmann regime

➤ **Boltzmann regime:** **segregation timescale** $\tau < \Delta\gamma_\alpha$ **Structural relaxation time**

Free surface systems



Structural relaxation time

Confined systems