LECTURE 1 - Introduction to Granular Materials
Jamming, Random Close Packing, The Isostatic State

Granular materials

- particles only interact when they touch
  hard cores: rigid, incompressible, particles
  soft cores: deformable particles repel with a finite force when they overlap
- particles are *large* - thermal fluctuations can be ignored
- collisions inelastic, energy is dissipated and not conserved

Due to above properties, granular materials are effectively at $T = 0$. The various allowed configurations of grains are *not* sampled according to a thermal Gibbs ensemble. Different configurations can only get sampled via external perturbations such as stirring, shearing, vibrating, etc. Although granular materials are thus a *non-equilibrium* system, we nevertheless will try to describe them using methods borrowed from statistical mechanics. But it always remains a question what particular properties are general and what may vary with the particular way the material is prepared.

Jamming

Consider N particles (grains), each of a fixed volume v_0, all confined to a box of volume V. A key parameter of the system is the *packing fraction*, or volume density (or simply "density"), ϕ,

$$\phi \equiv N v_0 / V \tag{1}$$

which gives the fraction of the total volume of the box that is physically occupied by the particles. The *free volume* is $(1 - \phi)V$.

When ϕ is sufficiently small, particles do not touch each other and the pressure of the granular system is $p = 0$. If one agitates the particles via some external perturbation, the particles will bounce around like particles in a gas or liquid. If one orients the particles in a gravitational field they will flow like a liquid.

Now imagine increasing ϕ, for example by slowly pushing in a piston to decrease the volume V of the box while the number of particles N remains constant. As ϕ increases one will reach a value ϕ_J at which the particles touch and (except for isolated rattlers) lock into a rigid but disordered structure. Pushing on the piston to further increase ϕ the system will exert a a pressure $p > 0$ back on the piston. This is the *jamming transition*: a transition from a liquid-like state to a rigid but disordered solid state as the packing fraction ϕ is varied through a critical value ϕ_J. For a system with a finite number of particles N, the specific value of ϕ_J may depend somewhat on the initial state of the system as it is compressed. However

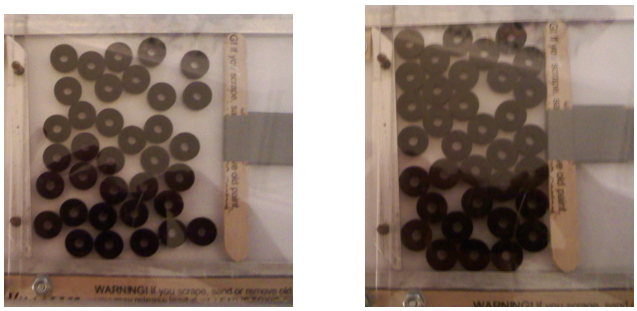


Figure 1: left: particles in a liquid-like state; right: particles in a jammed state

as $N \to \infty$, ϕ_J is believed to approach a unique value independent of initial configuration (however it may still be that the value of ϕ_J might depend slightly on the particular physical process that measures the response - more later on this).

The jammed state for $\phi \geq \phi_J$ is one in which each particle (except for isolated rattlers that are not locked into the rest of the structure) is in a mechanically stable equilibrium:

- Forces on each particle balance to zero - if displace a particle, forces from its neighbors will push it back.
- Torques on each particle balance to zero - if rotate a particle, torques from its neighbors will push it back

The nature of the jammed state and the value ϕ_J of the jamming transition depends on the dimensionality d of the system as well as the properties of the individual particles.

- frictionless vs frictional particles: When particles contact, there will be a repulsive normal force $\mathbf{F}_n$. If particle surfaces are rough, there can also be a tangential frictional force, $\mathbf{F}_t$, with $|\mathbf{F}_t| \leq \mu |\mathbf{F}_n|$, where μ is the coefficient of friction in a simple Coulomb model of static friction. If $\mu = 0$, we say the particles are frictionless and $\mathbf{F}_t$ always vanishes.
- spherical vs non-spherical particles

We will start our discussion by considering the specific case of spherical frictionless particles. Later we will have some comments about the more general cases. But in the subsequent two lectures we will return to consider only the simple case of frictionless spheres.

Random Close Packing

For spherical, frictionless, and *rigid* (incompressible, non-deformable) particles, the density at which particles jam is often called the *random close packing* density ϕ_{RCP}.

Closed packing means the particles are touching and packed in as tightly as possible. *Ordered* close packing Bravais lattices in two and three dimensions are the hexagonal and face centered cubic (fcc) lattices respectively.

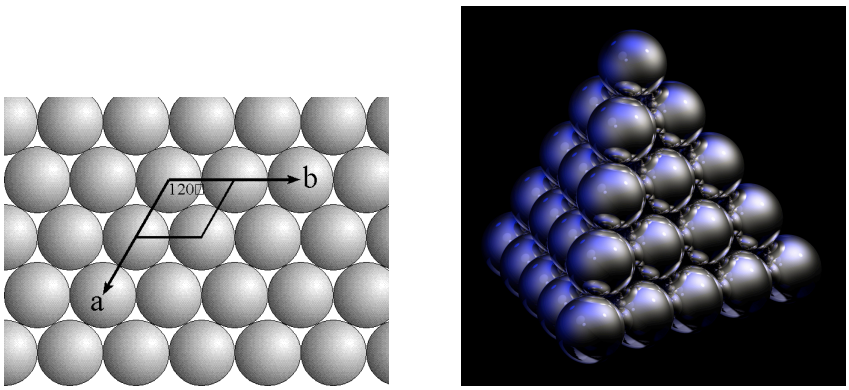


Figure 2: left: hexagonal lattice in two dimensions; right: face centered cubic lattice in three dimensions

These ordered lattices have packing fractions $\phi_{\rm hex} = \pi/(2\sqrt{3}) = 0.9069$ and $\phi_{\rm fcc} = \sqrt{2}\pi/6 = 0.7405$ and represent the densest packings of rigid spheres in 2D and 3D respectively.

In *random close packing*, the particles are put down as close as possible, but randomly. From numerous experiments and numerical simulations, one finds that for large number of particles N, the random close packing densities are:

- in 2D $\quad \phi_{\rm RCP} \simeq 0.84$
- in 3D $\quad \phi_{\rm RCP} \simeq 0.64$

Random close packing occurs at a *lower* density than ordered close packing. The randomly closed packed jammed state is therefore in principal only *meta-stable*. For large N in 3D, however, this meta-stability is extremely stable!

In 2D, even for large N, perturbing the randomly packed state of uniform disks will often result in its crystallization into the ordered hexagonal lattice. To study random packing in 2D one therefore usually uses a bidisperse, or polydisperse, mixture of disks of different radii. Such randomly packed non uniform mixtures are usually very stable against ordering [although even in this case the random packed state is only in principal meta-stable; the different size disks could in principal phase separate and then order]. The value of $\phi_{\rm RCP}$ seems rather robust against the details of the polydisperse distribution in 2D, it is always around 0.64.

Despite the many consistent experimental and numerical determinations of the values of $\phi_{\rm RCP}$, a clear mathematical definition of what is meant by RCP, and a precise analytical

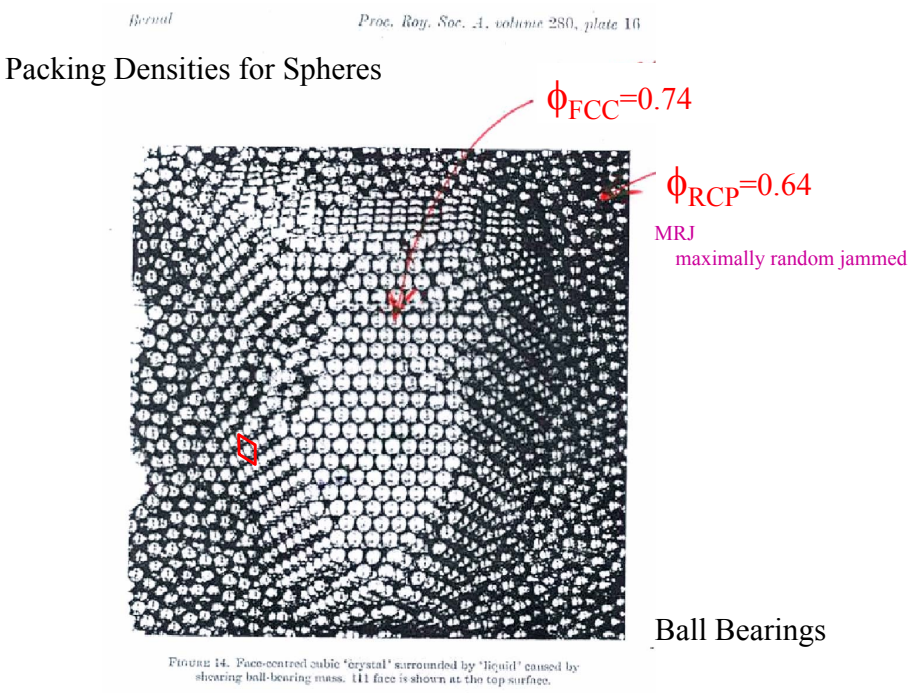


Figure 3: slide of 3D ball bearing packing, stolen from colloquium of Paul Chaikin

calculation of the values of $\phi_{\rm RCP}$ remain elusive. It remains debated even if there is any such clear mathematical definition!

A very simple model that gives a good estimate for the value of $\phi_{\rm RCP}$ in 2D was given by Williams in PRE 57, 7344 (1998) [note the relatively recent date!]. Consider a Voronoi tesselation of a configuration of disks, as shown below.

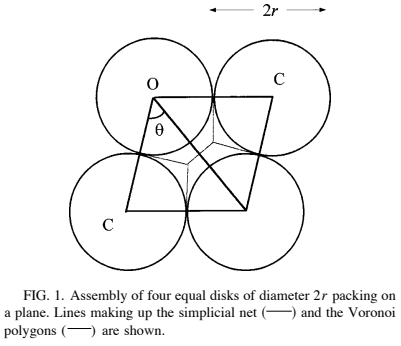


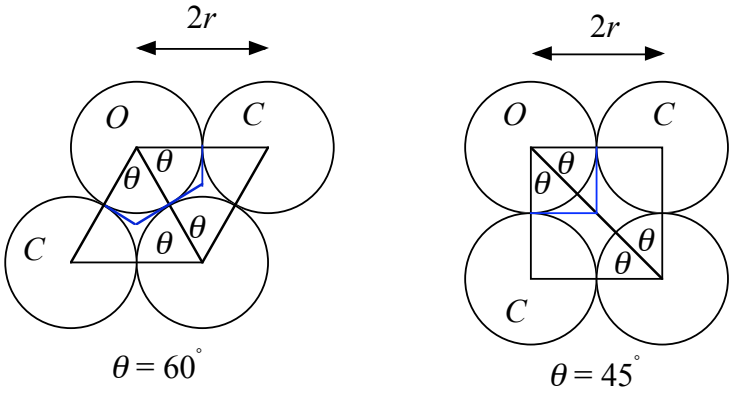
FIG. 1. Assembly of four equal disks of diameter $2r$ packing on a plane. Lines making up the simplicial net (——) and the Voronoi polygons (——) are shown.

Figure 4: from Williams, PRE 57, 7344 (1998)

To do a Voronoi tesselation about a particular disk, one draws lines from the center of that

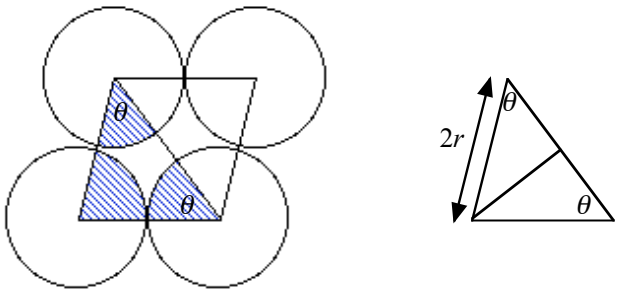
disk to the centers of all other disks. Then one draws the perpendicular bisectors of all these lines. The inner envelop of all these bisecting lines encloses the area that is closer to the center of the disk of interest than to that of any other disk. The lines bisected by this inner envelop define the nearest neighbors of the disk of interest. One can do the same for all disks to get a unique, non-overlapping, tiling of the total area.

Consider the four close packed (i.e. touching) nearest neighbor disks above. The four sides of the rhombus are all of equal length $2r$, the diameter of a disk. The largest the angle θ can be is $\theta = 60°$. In this case, the disk opposite disk "O" is actually touching "O" and the disks are as in an ordered hexagonal structure. The smallest value is $\theta = 45°$. In this case, the disk opposite "O" is no longer a nearest neighbor (the bond connecting its center to the center of "O" is no longer bisected in the tesselation of "O").



Williams then argues that in a random close packing, all possible angles $45° \leq \theta \leq 60°$ are equally likely. The average angle is thus $\bar{\theta} = 52.5°$.

Now the packing fraction associated with a particular angle θ is just the ratio of the area of the disks contained within a given triangle (the shaded area below) to the area of the triangle.



Since the angles of the triangle sum to 180°, the area of the disks contained in the tri-

angle is just half the area of one disk, $\frac{1}{2}\pi r^2$. The area of the isosceles triangle is just $(2r\cos\theta)(2r\sin\theta) = 4r^2\cos\theta\sin\theta = 2r^2\sin 2\theta$. So the packing fraction associated with a particular angle θ is

$$\phi(\theta) = \frac{\frac{1}{2}\pi r^2}{2r^2\sin 2\theta} = \frac{\pi}{4\sin 2\theta} \tag{2}$$

If we set $\theta = \bar{\theta} = 52.5°$, the average value, we then get Williams' value $\phi_{\rm RCP} \simeq 0.813$. We can do a little better by averaging $\phi(\theta)$ rather than evaluating $\phi(\bar{\theta})$, i.e.

$$\phi_{\rm RCP} \simeq \frac{3}{\pi}\int_{\pi/4}^{\pi/3} d\theta \frac{\pi}{4\sin 2\theta} = 0.824 \tag{3}$$

Despite the extreme simplicity of the calculation, the result is not too far from what is observed in simulations.

The extension of the above calculation to 3D was done by Jalali and Li, J. Chem. Phys. 120, 1138 (2004) [note the *very* recent date!]. They estimate in 3D $\phi_{\rm RCP} \simeq 0.6394$, which agrees very well with simulations.

Because the RCP state is only metastable, there always exist mechanically stable states of higher density (up to the ordered close packed density). One can always trade off density vs order: include a little ordering, to get to a higher density. In an experiment or simulation it is therefore possible that the exact value of $\phi_{\rm RCP}$ that one finds may depend somewhat on the protocol one is using to create the jammed state.

Torquato et al. (Torquato, Truskett, and Debenedetti, PRL 84, 2064 (2000)) have questioned whether the RCP is indeed a mathematically well defined concept. They propose instead a "MRJ", the *maximally random jammed* state. One defines some *ordering measure* ψ that measures how ordered the packing is ($\psi = 1$ is ordered close packed). One can then draw the region in the $\psi - \phi$ plane where stable jammed packing can occur. The MRJ is defined as the jammed state with the smallest possible value of ψ. Torquato et al.'s numerical simulations find $\phi_{\rm MRJ} \simeq 0.64$, in agreement with accepted values for $\phi_{\rm RCP}$.

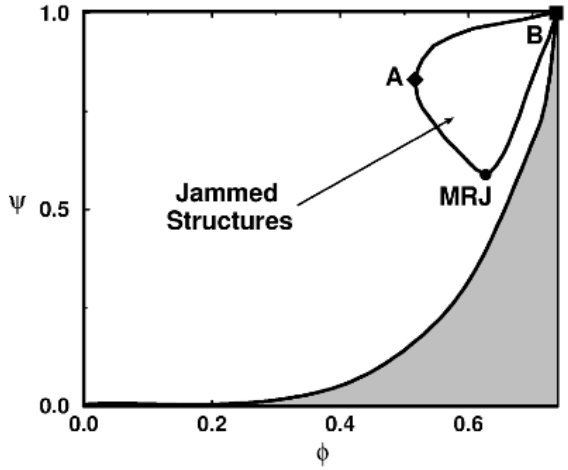


FIG. 1. A schematic plot of the order parameter c versus volume fraction f for a system of identical spheres with prescribed interactions. All structures at a given value of f must lie between the upper and lower bounds (white region); gray region is inaccessible. The boundary containing the subset of jammed structures is shown. The jammed structures are shown to be one connected set, although, in general, they may exist as multiply disconnected. Point A represents the jammed structure with the lowest density and point B represents the densest ordered jammed structure (e.g., close-packed fcc or hexagonal lattice for d 3, depending on the choice for c). The jammed structure which minimizes the order parameter c is the maximally random jammed state.

Figure 5: taken from Torquato et al. PRL 84, 2064 (2000)

Isostatic Packings

We return now to the more general case where the particles may be frictional and may have arbitrary shape. We now consider some aspects of the geometry of the jammed configuration, in particular the average contact number $\langle z \rangle$, where z is number of contacts that a particular grain has with other grains.

In a jammed state, each particle is in a state of stable mechanical equilibrium. Therefore the total force and the total torque of each particle i should vanish.

$$\text{force balance on particle } i: \qquad \sum_j{}' \mathbf{F}_{ij} = 0 \tag{4}$$

$$\text{torque balance on particle } i: \qquad \sum_j{}' [\mathbf{F}_{ij}\mathbf{d}_{ij} - \mathbf{d}_{ij}\mathbf{F}_{ij}] = 0 \tag{5}$$

Here the sum is over all particles j in contact with i, $\mathbf{F}_{ij}$ is the force on particle i due to particle j, and $\mathbf{d}_{ij}$ is the displacement from the center of particle i to the point of contact

with particle j. In expressing the condition of torque balance, we have made use of the force moment tensor $\mathbf{Fd} - \mathbf{dF}$ which is the generalization to any dimension of the three dimensional cross product $\mathbf{d} \times \mathbf{F}$.

For a given particle geometry (i.e. fixed particle positions and orientations) we can view the above force and torque balance equations as a set of linear equations for the unknown contact forces $\mathbf{F}_{ij}$. The number of such equations is determined as follows. Since force is a vector with d components in dimension d, the force balance gives d equations for each particle i, for a total of Nd equations. The force moment tensor is *antisymmetric*, hence in d dimensions it has $d(d-1)/2$ independent components. Thus torque balance gives $d(d-1)/2$ equations for each particle i, for a total of $Nd(d-1)/2$ equations. The total is therefore

$$Nd + Nd(d-1)/2 = Nd(d+1)/2 \tag{6}$$

linear equations to express the requirement of mechanical stability.

The number of contact forces $\mathbf{F}_{ij}$ is just given by the average number of contacts in the packing. If $\langle z \rangle$ is the average number of contacts per particle, the number of contact forces is then $N\langle z \rangle/2$.

If the number of linear equations is greater than the number of unknowns, the system is overconstrained, and in general there is no solution for the $\mathbf{F}_{ij}$, hence no jammed state. If the number of linear equations is less than the number of unknowns, there are many possible solutions for the forces (this fact gives rise to the idea of the force ensemble for $\phi > \phi_J$).

When the number of linear equations equals the number of unknowns, there is a unique solution for the forces. This is termed the *marginally stable* state, and is generally assumed to be the case exactly at the jamming transition ϕ_J. Removing one bond from the marginally stable state is generally believed to make the structure go *floppy*. The value of $\langle z \rangle$ that gives the marginally stable state is called the *isostatic* value z_{iso}.

Consider particles with a simple Coulomb frictional law at their surface. At a contact point between two particles, the tangential frictional force is related to the repulsive normal force by $|\mathbf{F}_t| \leq \mu |\mathbf{F}_n|$ where μ is the coefficient of friction. Consider the extreme limits of perfectly rough particles with $\mu = \infty$, and frictionless particles with $\mu = 0$.

- $\mu = \infty$, perfect frictional
  The tangential force $\mathbf{F}_t$ can be as large as one wishes, with no constraint on it. Therefore, in d dimensions, each contact force $\mathbf{F}_{ij}$ has d independent components. Since the number of contact forces is $\langle z \rangle N/2$, the number of "unknowns" in the linear equations for mechanical stability is thus $\langle z \rangle Nd/2$. Equating the number of equations with the number of unknowns give the isostatic value of z in this perfect frictional case.

$$\frac{Nd(d+1)}{2} = \frac{\langle z \rangle Nd}{2} \quad \Rightarrow \quad z_{iso} = d + 1 \qquad \text{(perfect frictional)}$$

- $\mu = 0$, frictionless
  Now the tangential force $\mathbf{F}_t$ vanishes, and $\mathbf{F}_{ij}$ always points in the direction normal to the surface at the point of contact. Each $\mathbf{F}_{ij}$ thus has only one independent component. The number of force unknowns is thus $\langle z \rangle N/2$. If we limit consideration to *spherical* particles, then $\mathbf{F}_{ij}$ is always radially outward and so can give no torque. We can therefore ignore the torque balance equations. Equating the number of force balance equations to the number of force unknowns, then determines z_{iso} for frictionless spherical particles.

  $$Nd = \frac{\langle z \rangle N}{2} \quad \Rightarrow \quad z_{iso} = 2d \qquad \text{(frictionless spheres)}$$

  For non-sperical particles we can write instead,

  $$Nd_f = \frac{\langle z \rangle N}{2} \quad \Rightarrow \quad z_{iso} = 2d_f \qquad \text{(frictionless)}$$

  where here d_f is the number of degrees of freedom as determined by the symmetry of the particles. For a generally shaped particle $d_f = d(d+1)/2$, but for more symmetric cases we have: in 2D, circular disks ($d_f = 2$, $z_{iso} = 4$), ellipses ($d_f = 3$, $z_{iso} = 6$); in 3D, spheres ($d_f = 3$, $z_{iso} = 6$), spheroids ($d_f = 5$, $z_{iso} = 10$), general ellipsoid ($d_f = 6$, $z_{iso} = 12$).

But there is a problem with this analysis for frictionless non-spherical particles. If $\langle z \rangle = a_{iso}$ at jamming, there would seem to be discontinuous behavior. If one just slightly distorted a spherical particle to make it only slightly ellipsoidal, d_f jumps discontinuously from 3 to 6 and so z_{iso} jumps discontinuously from 6 to 12. This seems unphysical. Numerical work (Donev, Connelly, Stillenger and Torquato, PRE 75, 051304 (2007)) shows that as one smoothly increases the aspect ratio to turn a sphere increasingly ellipsoidal, $\langle z \rangle$ at jamming *smoothly* increases (with no jumps or discontinuities) from its isostatic value of 6 for spheres to the isostatic value of 12 for ellipses, as the aspect ratio gets large. Thus in general, ellipsoidal particles are *hypostatic* (i.e. $\langle z \rangle < z_{iso}$) at jamming. See figure below from Donev et al.

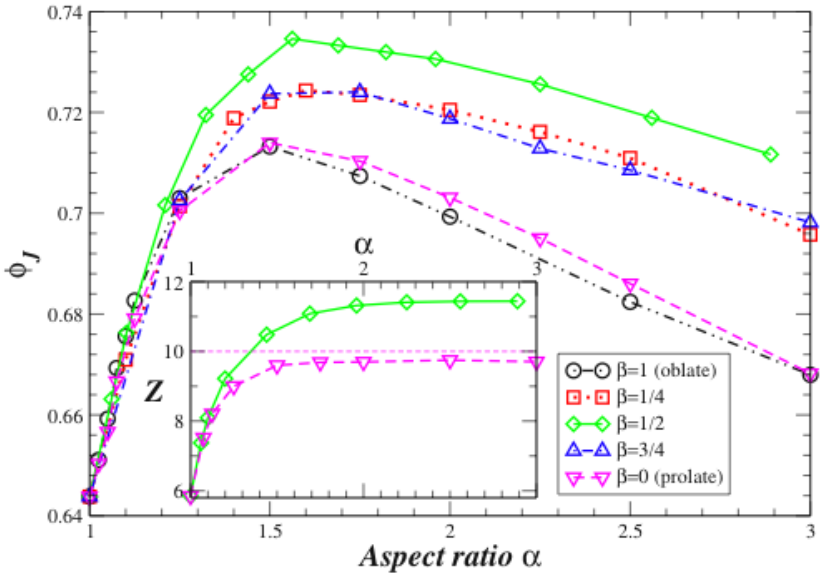


FIG. 1. (Color online) Jamming density and average contact number (inset) for packings of $N = 10\,000$ ellipsoids with ratios between the semi-axes of $1 : \alpha^{\beta} : \alpha$ (see Fig. 2 in Ref. [4]). The isoconstrained contact numbers of 10 and 12 are shown as a reference.

Figure 6: from Donev et al., PRE 75 051304 (2007)

Note, all the above counting arguments for z_{iso} only hold for *random* packings. One is assuming that if the particle positions are random, then the force and torque balance equations for each particle are linear independent of those of the other particles. This is not so if particle positions are correlated in some way to make some subset of the equations linearly dependent; this is the case with ordered packings.

Finally, we return to *spherical* particles. We had the two limits for jamming,

$$\langle z \rangle = d + 1 \qquad \mu = \infty \text{ (perfect frictional)} \tag{7}$$

$$\langle z \rangle = 2d \qquad \mu = 0 \text{ (frictionless)} \tag{8}$$

For finite friction $0 < \mu < \infty$, as μ decrease from ∞ to zero it is believed that $\langle z \rangle$ at jamming goes from one limit above to the other, $d + 1 < \langle z \rangle < 2d$. For frictional particles, where generally $\phi_J < \phi_{\rm RCP}$, one sometimes refers to ϕ_J as *random loose packing* RLP.

Song, Wang and Makse in Nature 453, 629 (2008), consider frictional spheres in 3D and using approximate analytic arguments and simulations map out the region in the $\langle z \rangle - \phi$ plane where one can have jamming occur, as μ varies. The parameter X in their phase diagram below is the *compactivity*, which we will discuss in the next lecture.

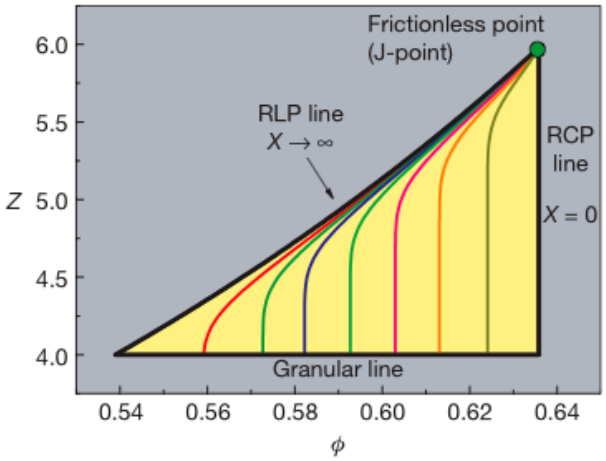


Figure 1 | Phase diagram of jamming: theory. Theoretical prediction of the statistical theory. All disordered packings lie within the yellow triangle demarcated by the RCP line, RLP line and granular line. Lines of uniform finite compactivity are in colour. Packings are forbidden in the grey area.

Figure 7: from Song, Wang and Makse, Nature 453, 629 (2008)