

**DOE Grant DE-FG02-03ER15458: Studies of elasticity, sound
propagation and attenuation of acoustic modes in granular media**

FINAL REPORT

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Abstract

This is the final report describing the results of DOE Grant # DE-FG02-03ER15458 with original termination date of April 31, 2013, which has been extended to April 31, 2014. Section II describes the frequency-dependent attenuation and elasticity in unconsolidated earth materials via the normal mode decomposition of the effective mass and the effect of stress and damping. Section III investigates the pair fluctuation analysis of the elastic moduli of granular materials. Section IV explains the studies of microstructure of packings of particles ranging from spherical to non-spherical shapes, with adhesion, friction and in any dimensions. We conclude with a list of talks given under the support of this grant, a list of papers published and submitted, the list of personnel, and we attach all the papers published under this grant.

I. PROGRAM SCOPE

The goal of this project is to develop a theoretical and experimental understanding of sound propagation, elasticity and dissipation in granular materials. The topic is relevant for the efficient production of hydrocarbon and for identifying and characterizing the underground formation for storage of either CO₂ or nuclear waste material. Furthermore, understanding the basic properties of acoustic propagation in granular media is of importance not only to the energy industry, but also to the pharmaceutical, chemical and agricultural industries. We employ a set of experimental, theoretical and computational tools to develop a study of acoustics and dissipation in granular media. These include the concept effective mass of granular media, normal modes analysis, statistical mechanics frameworks and numerical simulations based on Discrete Element Methods. Effective mass measurements allow us to study the mechanisms of the elastic response and attenuation of acoustic modes in granular media. We perform experiments and simulations under varying conditions, including humidity and vacuum, and different interparticle force-laws to develop a fundamental understanding of the mechanisms of damping and acoustic propagation in granular media. A theoretical statistical approach studies the necessary phase space of configurations in pressure, volume fraction to classify granular materials.

We have developed a theory to predict the effective mass of granular media in terms the normal-mode spectrum of finite-sized granular systems. The theoretical formalism allows the interpretation of the effective mass experiments performed in our lab to perform a microscopic investigation of the origin of dissipation and elasticity properties in granular matter. The main goal is to derive expressions of the effective mass in terms of the complex valued normal modes of the confined granular medium. A pole-decomposition has been derived that allows us to obtain the normal mode spectrum from direct experimental measurement of the effective mass. The spectrum allows us to determine the precise dissipative properties of the medium. In a recent application we have shown a markedly increase of the damping rate of each normal mode with the humidity of the medium relevant to our measurements. We developed the theory for weakly and strongly damped medium to make contact with experimental results. This theory accurately predicts the frequencies, widths, and relative amplitudes of the various flexural mode resonances observed with rectangular bars, each having a cavity filled with loose tungsten granules. A set of computer simulations has been

performed to test the theory and finds agreement with theoretical and experimental results.

II. NORMAL MODE SPECTRUM OF GRANULAR SYSTEMS AND THE EFFECTIVE MASS

Below we explain the use of the newly developed concept of effective mass of granular matter to develop a theoretical, numerical and experimental understanding of sound propagation, elasticity and dissipation in granular materials.

A. Theory: normal mode analysis.

From the theoretical perspective [1], we have analyzed the acoustic response of a cavity filled with a loose granular material and derived expressions of the effective mass in terms of the complex valued normal modes of the confined granular medium. The theory accurately predicts the frequencies, widths, and relative amplitudes of the various mode resonances. We are able to decompose the effective mass into a sum over the contributions from each of the normal modes of the granular medium. Our results indicate that increasing either the viscosity or the humidity, as the case may be, does markedly increase the damping rate of each normal mode relevant to our measurements. However, there is appreciable damping even in the absence of any macroscopic film. With a notable exception in the case of large humidity, all the relevant modes are weakly damped in the sense of a microscopic theory based on damped contact forces between rigid particles.

B. Stress dependent normal modes of granular materials

We investigate the attenuation properties of granular materials by measuring the effective mass of a cavity filled with grains from a experimental and numerical approach using Discrete Element Methods to simulate a jammed packing of spherical particles [2]. A set of computer simulations are performed to test the normal mode theory and experimental results for the effective mass of a granular medium and finds agreement with theoretical and experimental results.

Our work shows the first experimental evidence of criticality at the jamming transition of

granular matter in terms of the scaling of normal modes from the effective mass of granular matter. Specifically, we show how the critical hypothesis at the jamming transition can be tested in a controlled laboratory experiment depicting a true thermodynamic transition characterizing jammed matter. Our experiment clearly shows that even though granular matter is athermal, i.e. there is no random thermal motion, the system displays the properties of a thermodynamic critical point at the jamming transition.

We developed a experiment in a realistic scenario involving frictional and dissipative granular materials [2]. To that end, we adapted an experimental technique to measure the normal modes of the granular medium: the stress and frequency-dependent effective mass. Previously, the normal modes were measured only numerically from diagonalization of the dynamical matrix. However, this technique was not successful at solving the experiment since the interparticle potential defining the matrix is not available in practice. The technical novelty of our work resides in the introduction of the effective mass technique which allows to investigate not only the normal modes of the system but also other mechanical properties of the jamming transition.

To that end, we developed a novel theoretical framework valid to extract the normal modes from a pole decomposition of the effective mass, which we show to be exact. From the pole decomposition, the characteristic normal mode frequency was extracted and then studied as a function of the external pressure. The results are plotted in Fig. 1 and show scaling behavior in terms of critical exponent Ω .

Our work shows strong evidence that the jamming transition (J-point) is indeed a critical point in this realistic experimental system involving friction and dissipative forces. An additional crucial result also emerges: the critical exponent Ω characterizing the jamming transition is significantly smaller than the prediction of theory for ideal frictionless spheres $\Omega = 1/2$. We interpret this result as a new universality class in frictional and dissipative jammed matter. Thus, our experiments not only demonstrate the critical hypothesis, but also open new theoretical challenges to explain the anomalous value of the critical exponent defining the jamming transition.

Our results have implications not only for granular matter but also for out-of-equilibrium systems ranging from structural glasses to spin-glasses, and other soft materials like colloids and compressed emulsions. All these systems present a jamming transition via structural arrest (or glass transition) which is believed to be controlled by the common critical point of

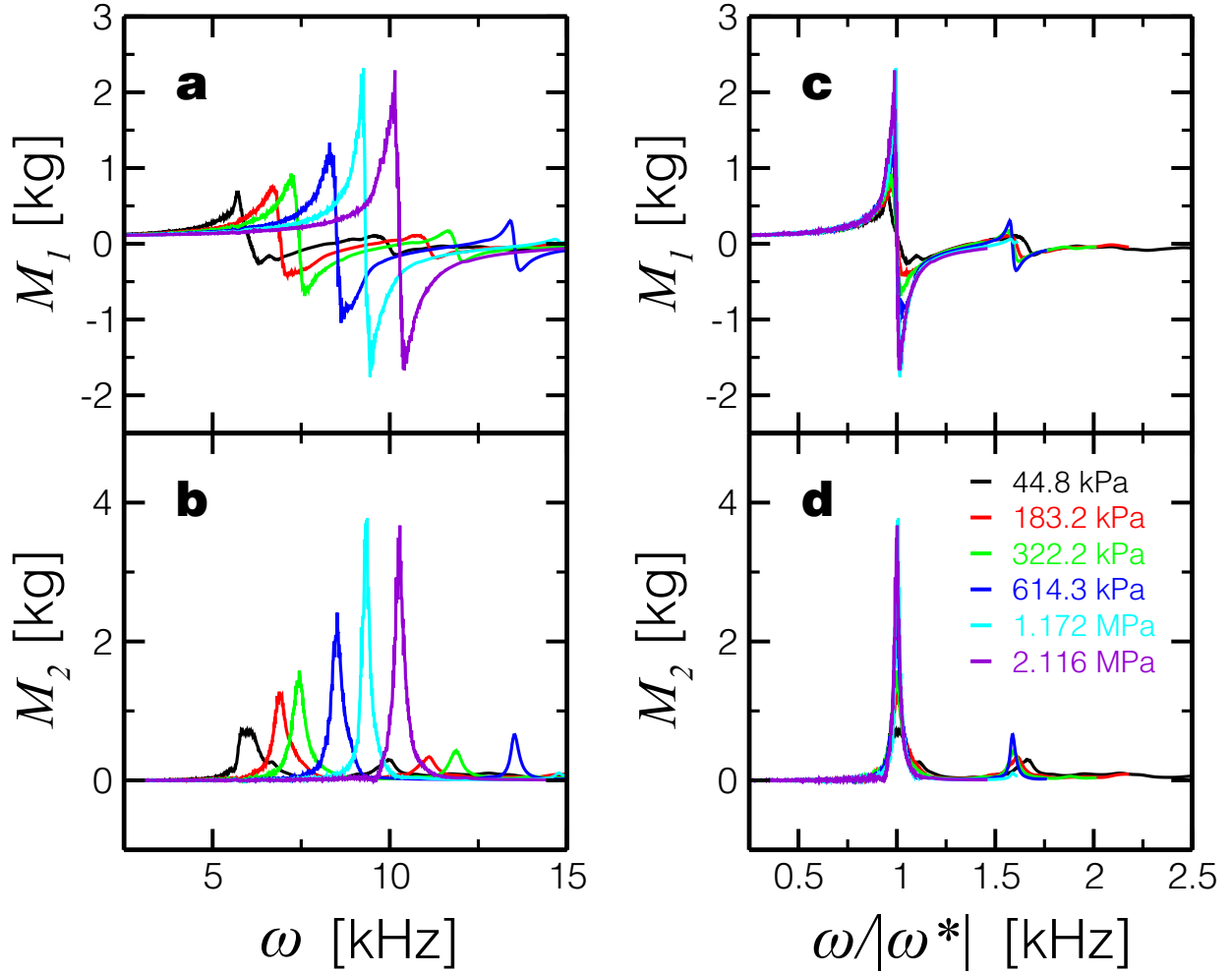


FIG. 1: Effective mass of tungsten particles in a cavity as a function of driving frequency: **(a)** Real part $M_1(\omega)$, and **(b)** Imaginary part, $M_2(\omega)$, plotted for the indicated external stresses. We calculate $\tilde{M}$ for 13 stresses between $\sigma = 44.8$ kPa and 6.39 MPa and show six curves as indicated for clarity. All datasets are available at <http://jamlab.org>. **(c)-(d)** Data collapse of $M_1(\omega)$ and $M_2(\omega)$ according to the scaling scenario.

jamming. The present effective mass technique and pole decomposition can be extended to investigate these generic jammed systems as well. Thus, our results will be of great interest to the broad experimental community of physicists interested in these non-equilibrium systems.

C. Frequency-dependent attenuation and elasticity in unconsolidated earth materials: effect of damping

We use the Discrete Element Method (DEM) to understand the underlying attenuation mechanism in granular media, with special applicability to the measurements of the so-called effective mass developed earlier. Results are reported in [3]. We consider that the particles interact via Hertz-Mindlin elastic contact forces and that the damping is describable as a force proportional to the velocity difference of contacting grains. We determine the behavior of the complex-valued normal mode frequencies using 1) DEM, 2) direct diagonalization of the relevant matrix, and 3) a numerical search for the zeros of the relevant determinant. All three methods are in strong agreement with each other. The real and the imaginary parts of each normal mode frequency characterize the elastic and the dissipative properties, respectively, of the granular medium. We demonstrate that, as the interparticle damping, ξ , increases, the normal modes exhibit nearly circular trajectories in the complex frequency plane and that for a given value of ξ they all lie on or near a circle of radius R centered on the point $-iR$ in the complex plane, where $R \propto 1/\xi$ (see Fig. 2). We show that each normal mode becomes critically damped at a value of the damping parameter $\xi \approx 1/\omega_n^0$, where ω_n^0 is the (real-valued) frequency when there is no damping. The strong indication is that these conclusions carry over to the properties of real granular media whose dissipation is dominated by the relative motion of contacting grains. For example, compressional or shear waves in unconsolidated dry sediments can be expected to become overdamped beyond a critical frequency, depending upon the strength of the intergranular damping constant.

D. Experiments

We have analyzed the acoustic response of a structure that contains a cavity filled with a loose granular material experimentally [4]. We have measured the effects of adsorbed films on the attenuative properties of loose granular media occupying a finite sized rigid container, which is open on the top. We measure the effective mass, $M(w)$, of loose tungsten particles prepared under two different sets of conditions: 1) We lightly coat tungsten grains with a fixed volume fraction of silicone oil (PDMS), where the liquid viscosity is varied for individual realizations. 2) In the other set of experiments we vary the humidity. Inasmuch as the

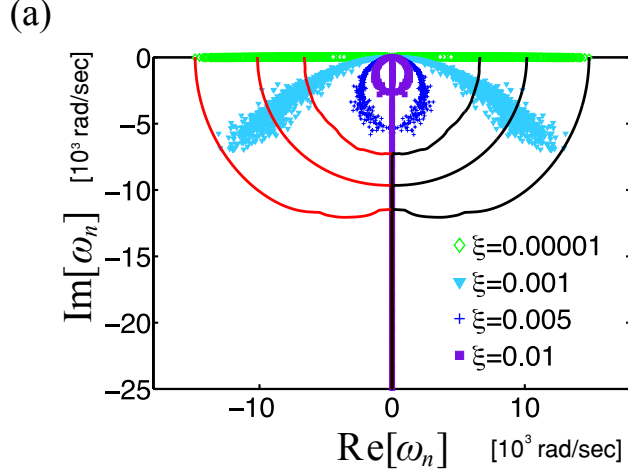


FIG. 2: Locus of all the complex-valued normal mode frequencies of our large system for four different values of the damping parameter, ξ . The normal modes are calculated via the solution of the dynamical matrix. Also shown are the trajectories of three of the normal mode frequencies as ξ is varied from low to high values. The frequencies approximately follow circular trajectories. The stated values for ξ are in millisecond.

dominant mechanism for damping is due to adsorbed water at the grain-grain contacts, the significant effects of humidity on both the effective mass of the granular medium as well as on the response of the grain-loaded bars are monitored. Here, depending upon the humidity and the preparation protocol, it is possible to observe one, two, or three distinct resonances in a wide frequency range (15 kHz) over which the empty bar has but one resonance. These effects are understood in terms of the theoretical framework, which may simplify in terms of perturbation theories.

III. IRREVERSIBLE INCREMENTAL BEHAVIOR IN A GRANULAR MATERIAL

In a collaboration with Prof. J. Jenkins from Department of Theoretical and Applied Mechanics, Cornell University, and Prof. Luigi La Ragione from Dipartimento di Ingegneria Civile e Ambientale, Politecnico di Bari, we are investigating the properties of isotropically compressed dense packings of spheres with the goal of capturing the relevant state variables to develop a theoretical framework for the effective bulk and shear moduli and dissipation in granular packings [5].

IV. STATISTICAL MECHANICS APPROACH TO CHARACTERIZE GRANULAR MEDIA AND THE RESPONSE FUNCTIONS

We have studied the structural properties determining the elastic response of granular media using a statistical mechanics approach based on the volume ensemble for granular materials.

A statistical theory of volume fluctuations has been developed based on the calculation of the Voronoi volume of a single particle in a packing [6] and a coarse-grained free volume fraction [7]. This allows the application of statistical mechanics to predict the structural properties of the granular aggregate. Our calculations reproduce the limiting densities of RCP and RLP [7], the entropy of the packing [8] as well as the distribution of volumes and coordination number [9]. The calculations have been extended to 2d packings [10]. We are investigating how the micro-structure described by the theory determines the elastic response and the vibrational density of states of the granular packing from the lowest densities of RLP up to crystallization at FCC [11]. The effect of the particles polydispersivity is investigated to understand the mechanical response of the granular material [12].

Analysis have been extended to investigate the pressure dependence of the structural properties. We have measured the so called, angoricity of the packings, a temperature-like variable that determine the stress distribution [13, 14].

A. A statistical theory of correlations in random packings of hard particles

A random packing of hard particles represents a fundamental model for granular matter. Despite its importance, analytical modeling of random packings remains difficult due to the existence of strong correlations which preclude the development of a simple theory. We take inspiration from liquid theories for the n -particle angular correlation function to develop a formalism of random packings of hard particles from the bottom-up. Papers published in [15, 16]. A progressive expansion into a shell of particles converges in the large layer limit under a Kirkwood-like approximation of higher-order correlations. We apply the formalism to hard disks and predict the density of two-dimensional random close packing (RCP), $\phi_{rcp} = 0.85 \pm 0.01$, and random loose packing (RLP), $\phi_{rlp} = 0.67 \pm 0.01$. Our theory also predicts a phase diagram and angular correlation functions that are in good agreement with

experimental and numerical data.

B. Force transmission in jammed disordered packings of hard particles

The force distribution of jammed disordered packings has always been considered a central object in the physics of granular materials. However, many of its features are poorly understood. In particular, analytic relations to other key macroscopic properties of jammed matter, such as the contact network and its coordination number, are still lacking. We have developed a mean-field theory for this problem, based on the consideration of the contact network as a random graph where the force transmission becomes a constraint satisfaction problem. We can thus use the cavity method developed in the past few decades within the statistical physics of spin glasses and hard computer science problems. Paper is published in [17]. This method allows us to compute the force distribution $P(f)$ for random packings of hard particles of any shape, with or without friction. We find a new signature of jamming in the small force behavior $P(f) \sim f^\theta$, whose exponent has attracted recent active interest. Furthermore, we relate the force distribution to a lower bound of the average coordination number $z_{min}(\mu)$ of jammed packings of frictional spheres with coefficient μ . This bridges the gap between the two known isostatic limits in zero and infinite dimensions by extending the naive Maxwell's counting argument to frictional spheres. The theoretical framework describes different types of systems, such as non-spherical objects in arbitrary dimensions, providing a common mean-field scenario to investigate force transmission, contact networks and coordination numbers of jammed disordered packings.

C. Statistical mechanics framework for microstructure of granular materials: from spherical to non-spherical particles with adhesion, friction and for any dimension.

Understanding random packings of objects of a particular shape is the first step towards a theory of elasticity of granular materials. However, such jammed matter states have eluded any systematic theoretical treatment due to the strong positional and orientational correlations involved. In recent years progress on a fundamental description of jammed matter could be made by starting from a constant volume ensemble in the spirit of conventional statistical mechanics. Our work has shown that this approach, first introduced by S. F.

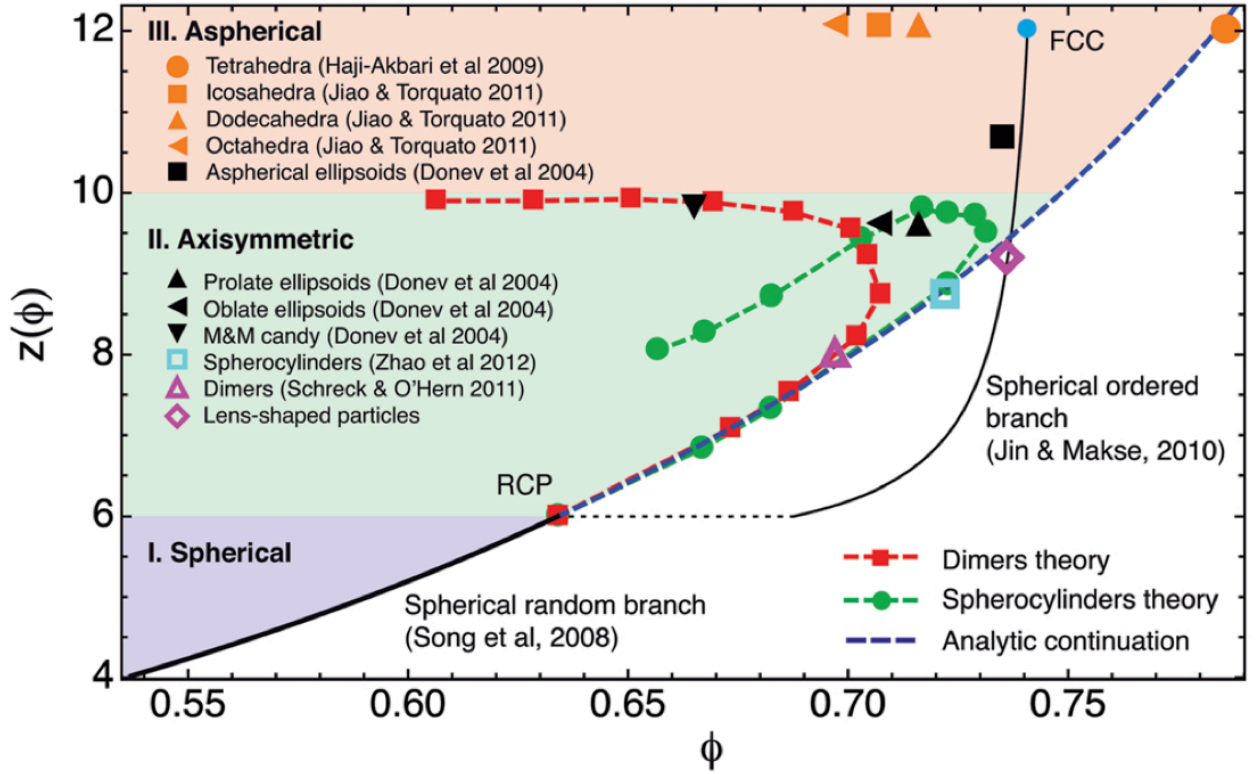


FIG. 3: Phase diagram for all shapes. By plotting z vs ϕ we obtain a phase diagram for smooth shapes. We observe that the spherical random branch ϕ_{sph} , which ends at the RCP point at $(0.634, 6)$, in fact continues smoothly upon deformation into dimers and spherocylinders as predicted by our theory. The spherocylinder continuation provides a boundary for all known packing states of rotationally symmetric shapes. The continuations from RCP. For a given value of z , the densest packing is achieved by spherocylinders, followed by dimers, prolate ellipsoids, and oblate ellipsoids. Note that the continuations for spherocylinders and dimers are almost identical.

Edwards more than two decades ago, can be cast into a predictive framework to calculate the packing fractions of both spherical and non-spherical hard particles, with or without adhesion and friction, and for any dimension. Papers published in [18–20].

In particular we have developed equations of states relating the volume fraction of a packing of non-spherical particles with the coordination number [18]. Finding the optimal random packing of non-spherical particles is an open problem with great significance in a

broad range of scientific and engineering fields. So far, this search has been performed only empirically on a case-by-case basis, in particular, for shapes like dimers, spherocylinders and ellipsoids of revolution. We have developed a mean-field formalism to estimate the packing density of axisymmetric non-spherical particles. We derive an analytic continuation from the sphere that provides a phase diagram (see Fig. 3) predicting that, for the same coordination number, the density of monodisperse random packings follows the sequence of increasing packing fractions: spheres $<$ oblate ellipsoids $<$ prolate ellipsoids $<$ dimers $<$ spherocylinders. We find the maximal packing densities of 73.1% for spherocylinders and 70.7% for dimers, in good agreement with the largest densities found in simulations. Moreover, we find a packing density of 73.6% for lens-shaped particles, representing the densest random packing of the axisymmetric objects studied so far. This theory allows us to characterize the microstructure of granular packings of any shape. The theory can be also extended to any dimension and we can also include the effects of adhesion forces which are crucial to understand packings of small particles.

V. TALKS GIVEN UNDER THE SUPPORT OF THE PRESENT GRANT

1. Complexity in Oil Industry, November 2011. International Center of Theoretical Physics, Natal, Brazil. “Nonlinear elasticity of granular materials”.
2. Frontier in Statistical Physics and Complex Systems, June 2-5, 2012, Catania, Italy. “Theory of packings”.
3. Yukawa Workshop on Glasses, July 16-19, 2013, Kyoto, Japan. “From Kepler to Ulam: searching for the optimal packing”.
4. STATPHYS 25, July 22-26, 2013, Seoul, South Korea. “Statistical Mechanics of Jammed Matter”.
5. Tsinghua University. Particle and Combustion Engineering Research Group, Department of Thermal Engineering. October 14, 2013. “From avalanches to packings: The physics of granular matter”.
6. Mechanics of Particulates, February 12, 2014, NYU School of Engineering, New York. “Packings on nonspherical particles”.

7. Jam Packed - Packing and Jamming of Particulate Systems, September 15-18, 2014, Erlangen, Germany. “Packings of spherical and nonspherical hard particles.”

VI. PERSONNEL SUPPORTED BY THIS GRANT.

Three graduate students from City College of New York have been supported by this grant: Yuliang Jin, Kun Wang and Lin Bo. Yuliang Jin has been developing theories of the strong correlations in packings of spherical hard spheres with the goal to understand its microstructure. Kun Wang developed a theoretical framework for thermodynamics of granular matter and Lin Bo developed a theoretical formulation of force transmissions in packings using the cavity method. A postdoc, Yanqing Hu is also supported. Dr. Hu has performed the normal mode analysis of the effective mass of granular media, including theoretical framework and simulations with Discrete Element Methods.

VII. PAPERS RESULTING FORM THIS GRANT:

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 - [2] Y. Hu, D. L. Johnson, J. J. Valenza, F. Santibanez, H. A. Makse. *Stress-dependent normal mode frequencies from the effective mass of granular matter*, Phys. Rev. E **89**, 062202 (2014).
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to non-spherical particles. Soft Matter **10**, 4415-4570 (2014).

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Angoricity and compactivity describe the jamming transition in soft particulate matter

doi: [10.1209/0295-5075/91/68001](https://doi.org/10.1209/0295-5075/91/68001)

From force distribution to average coordination number in frictional granular matter

doi: [10.1016/j.physa.2010.05.053](https://doi.org/10.1016/j.physa.2010.05.053)

Jamming II: Edwards' statistical mechanics of random packings of hard spheres

doi:[10.1016/j.physa.2010.10.017](https://doi.org/10.1016/j.physa.2010.10.017)

Jamming III: Characterizing randomness via the entropy of jammed matter

doi:[10.1016/j.physa.2010.05.054](https://doi.org/10.1016/j.physa.2010.05.054)

Jamming in two-dimensional packings

doi: [10.1016/j.physa.2010.07.030](https://doi.org/10.1016/j.physa.2010.07.030)

Jamming I: A volume function for jammed matter

doi:[10.1016/j.physa.2010.06.043](https://doi.org/10.1016/j.physa.2010.06.043)

Model of random packings of different size balls

[10.1103/PhysRevE.81.051303](https://doi.org/10.1103/PhysRevE.81.051303)

Cavity method for force transmission in jammed disordered packings of hard particles

DOI: [10.1039/c4sm00667d](https://doi.org/10.1039/c4sm00667d)

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[10.1103/PhysRevE.86.011305](https://doi.org/10.1103/PhysRevE.86.011305)

Frequency-dependent attenuation and elasticity in unconsolidated earth
materials: effect of damping

[10.1190/GEO2013-0459.1](https://doi.org/10.1190/GEO2013-0459.1)

Distribution of volumes and coordination number in jammed matter: mesoscopic ensemble

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Abstract

We investigate the distribution of the volume and coordination number associated to each particle in a jammed packing of monodisperse hard sphere using the mesoscopic ensemble developed in Nature **453**, 606 (2008). Theory predicts an exponential distribution of the orientational volumes for random close packings and random loose packings. A comparison with computer generated packings reveals deviations from the theoretical prediction in the volume distribution, which can be better modeled by a compressed exponential function. On the other hand, the average of the volumes is well reproduced by the theory leading to good predictions of the limiting densities of RCP and RLP. We discuss a more exact theory to capture the volume distribution in its entire range. The available data suggests a plausible order/disorder transition defining random close packings. Finally, we consider an extended ensemble to calculate the coordination number distribution which is shown to be of an exponential and inverse exponential form for coordinations larger and smaller than the average, respectively, in reasonable agreement with the simulated data.

1. Introduction

Jammed matter refers to a broad class of physical many-body systems ranging from granular matter to frictionless emulsions, and colloids. These systems share the property that their constitutive particles can be blocked in a configuration far from thermal equilibrium when undergoing a jamming transition. The statistical mechanical description of these materials is based on the volume fluctuations of the system [1] taken to be the conservative quantity instead of energy, as typically done in thermal system. Therefore, the probability distribution of the volume occupied by each jammed particle is of particular interest and many studies have been devoted to investigate them in detail [2, 3, 4, 5, 6, 7, 8].

Recently, a theory of volume fluctuations has been developed at the mesoscopic level providing a relation between the volume occupied by each particle

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and its number of contacts [9]. Thus, the distribution of particle volumes in the system is intimately related to the distribution of contacts per particle. In this paper, we calculate the distribution of (orientational) volumes occupied by each particle in a jammed system of monodisperse hard spheres and the distribution of coordination numbers by following the theoretical formalism of [9], which is in turn based on the Edwards statistical mechanics of jamming [1].

The distributions of volumes and contacts in real packings represent ensemble averages in the statistical mechanics sense. Therefore, the distributions depend on the state of the packings specified by their compactivity through a Boltzmann-like probability in the partition function. We show that in two limiting cases of zero and infinite compactivity (corresponding to the random close packing, RCP, and random loose packing, RLP, respectively) the distributions can be obtained in analytical form. Theory predicts that the distribution of orientational volumes is exponential with a mean volume varying with the average coordination number for RLP and constant for RCP. The theoretical predictions are compared with computer generated jammed packings of equal-size spheres for any friction coefficient. We find that the theory well reproduces certain features of the numerical distributions, but not all, in the entire range of volumes.

The mean value of the occupied volumes is well reproduced by the theory. However, we find important deviations between theory and simulations for the higher moments of the distribution. For instance, simulations show a plateau for small volumes while theory predicts an exponential. For intermediate values, the exponential shape seems to provide a good fit to the simulated data and the predicted dependence of the characteristic volume on the coordination number follows partially the theoretical prediction. However, a higher scrutiny shows deviations in the tail of the distribution which is found to decay faster than exponential, having a compressed exponential tail. We conclude that the full understanding of the distribution requires a more precise theory. We discuss how to obtain more exact solutions of the volume distribution which can capture the behavior in the entire ranges of volumes.

On the other hand, the distribution of coordination numbers provides fundamental information of the microscopic packing structures [3, 8, 10, 11, 12], as well as important characteristics of the mesoscopic volume ensemble. In this paper, we study the distribution of coordination numbers by generalizing the ensemble proposed in [9] to include fluctuations in the number of contacts. Theory predicts an exponential decay for large coordination number and an inverse exponential for small coordination number. Computer simulations well reproduce the predictions. Overall, the present paper serves as a critical assessment of the theoretical predictions of the mesoscopic theory towards the development of an exact formulation at the microscopic level that could capture the behavior in the entire range of volume and coordination number fluctuations of jammed matter.

2. Mesoscopic ensemble of jammed matter

In this section, we briefly review the statistical mechanics theory developed at the mesoscopic level in [9], which serves as the theoretical framework for the study of the distributions of volumes and coordination numbers. A theoretical formalism of the volume ensemble is the starting point for the statistical mechanics of jammed matter [13]. The role traditionally played by the energy in thermal systems is replaced by the volume, and a new parameter X , called “compactivity”, is introduced as an analogue of temperature. As a consequence, the canonical partition function can be written as:

$$\mathcal{Z}(X) = \int e^{-W/X} g(W) \Theta_W dW, \quad (1)$$

where W is the free volume function, $g(W)$ is the density of jammed states for a given volume W , and Θ_W imposes the jamming condition. It has been shown [9] that the free volume of coarse-grained “quasiparticles” in a monodisperse hard sphere packing has an inverse relation with their coordination number z :

$$W(z) = \frac{2\sqrt{3}}{z} V_g, \quad (2)$$

where V_g is the sphere volume. Since the quasiparticles are coarse-grained over a uniform background field produced by other particles, Eq.(2) should be understood as a mean-field result at the mesoscopic level. Assuming the quasiparticles are independent, we can simplify the partition function Eq.(1) by changing variables:

$$\mathcal{Z}_{\text{iso}}(X) = \int_Z^6 e^{-W(z)/X} g(z) dz. \quad (3)$$

The limit of integration here is given by the isostatic condition [14, 15, 16] over the mechanical coordination number, Z , counting the contacts with nonzero forces. The mechanical coordination number is different from the geometrical coordination number, z , which counts all contacts, even those with zero forces. By definition, it is easy to see that the geometrical coordination number z is always equal or greater than the mechanical coordination number Z , and in general we have $Z \leq z \leq 6$. The mechanical coordination, $Z(\mu)$, depends on the friction, μ , the interparticle friction coefficient, and varies between $Z(0) = 2d = 6$ and $Z(\infty) = d + 1 = 4$ in dimensions $d = 3$. The density of states $g(z)$ is assumed to have an exponential form, $g(z) = (h_z)^{z-2d} = e^{-(z-2d)/z^*}$, with the constant $h_z \ll 1$, representing the typical separation of the configurations in the phase space (analogous to the Planck constant in quantum mechanics). We have $z^* = -1/\ln h_z$. Equation (3) provides a useful tool to calculate the ensemble average of any physical quantity $f(z)$ since

$$f(X, Z) = \frac{1}{\mathcal{Z}_{\text{iso}}} \int_Z^6 f(z) e^{-W(z)/X} g(z) dz. \quad (4)$$

3. PDF of the orientational free volumes

To calculate the volume distribution using the established mesoscopic formalism, we start by introducing the different definitions of the volume associated with each particle necessary to understand the problem. The starting point is the volume of a Voronoi cell associated to each particle. The Voronoi tessellation tiles the entire packing is shown in [17, 9] to be a good candidate for the volume function of jammed matter. The volume function replaces the Hamiltonian in thermal systems, and describes the state of the jammed packings in the ensemble average in the partition function [1, 9]. The Voronoi volume for each particle $\mathcal{W}_i^{\text{vor}}$ gives rise to the total volume of the system $\mathcal{W} = \sum_{i=1}^N \mathcal{W}_i^{\text{vor}}$, when summed up over all the N particles. In terms of the relative coordinates of the particles, $\vec{r}_{ij}$, we have obtained in [17, 9] for monodisperse particles of radius R and volume V_g the following formula for the Voronoi volume:

$$\mathcal{W}_i^{\text{vor}} = \frac{1}{3} \oint \left(\min_{\hat{s} \cdot \hat{r}_{ij} > 0} \left(\frac{r_{ij}}{2\hat{s} \cdot \hat{r}_{ij}} \right) \right)^3 ds. \quad (5)$$

where the integration is performed over the direction $\hat{s}$ forming an angle θ_{ij} with $\vec{r}_{ij}$ as in Fig. 1, and $\cos \theta_{ij} = \hat{s} \cdot \hat{r}_{ij}$. Taking advantage of this integration we can define an orientational Voronoi volume, $\mathcal{W}_i^s$, for a fixed direction $\hat{s}$, satisfying:

$$\mathcal{W}_i^{\text{vor}} = \frac{1}{\oint ds} \oint \mathcal{W}_i^s ds = \langle \mathcal{W}_i^s \rangle_s, \quad (6)$$

from which we obtain:

$$\mathcal{W}_i^s \equiv V_g \left(\frac{1}{2R} \min_{\hat{s} \cdot \hat{r}_{ij} > 0} \frac{r_{ij}}{\hat{s} \cdot \hat{r}_{ij}} \right)^3. \quad (7)$$

$\mathcal{W}_i^s$ defines the orientational Voronoi volume which is obtained without the integration over $\hat{s}$.

The average of the orientational volume over $\hat{s}$ for a single particle, Eq. (7), is the Voronoi volume, $\langle \mathcal{W}_i^s \rangle_s = \mathcal{W}_i^{\text{vor}}$ and the average of the orientational volume over many particles for a fixed $\hat{s}$ is the same as the average of the Voronoi volume over the particles: $\langle \mathcal{W}_i^s \rangle_i = \langle \mathcal{W}_i^{\text{vor}} \rangle_i$, in the case of isotropic systems. This last property is useful since it allows the use of the orientational volume to define the ensemble average of the volume fraction without resorting to the use of the Voronoi volume which contains the average over $\hat{s}$ and therefore is more difficult to treat from a theoretical point of view. We therefore promote the use of the orientational volume function $\mathcal{W}_i^s$ as the fundamental quantity to characterize the state of jammed matter instead of $\mathcal{W}_i^{\text{vor}}$. It is important to note that the probability densities of $P(\mathcal{W}_i^s)$ and $P(\mathcal{W}_i^{\text{vor}})$ in general differ. For instance, as discussed in Fig. 1 the orientational free volume can be for instance zero while the Voronoi free volume cannot. The distribution of Voronoi volume $P(\mathcal{W}_i^{\text{vor}})$ can be fitted by a Gamma distribution [18], however, $P(\mathcal{W}_i^s)$ has a different form as shown below.

We define the reduced free orientational volume function as

$$w^s \equiv \frac{\mathcal{W}_i^s - V_g}{V_g}, \quad (8)$$

(we drop the subscript i in w^s for simplicity of notation).

In what follows, we provide a theory for the probability distribution function of the orientational free volume, $P(w^s)$, which is less complex than the full Voronoi volume distribution of Eq. (5). In [17], this distribution is obtained under assumption of uniformity in the packing, making the theory valid at a mesoscopic level of a few particles diameters. This approximation can be seen as defining quasiparticles of free volume w^s capturing the behavior at the mesoscopic distance. Under this approximation the inverse cumulative distribution $P_{>}$ is obtained (see Eq. (20) in [9] and Eq. (41) in [17]) from where the probability density can be calculated as, $P(w^s) = \frac{d(1-P_{>})}{dw^s}$, then

$$P(w^s) = \frac{1}{w} \exp\left(-\frac{w^s}{w}\right), \quad (9)$$

where the average value over the particles,

$$w \equiv \langle w^s \rangle_i = \int w^s P(w^s) dw^s, \quad (10)$$

was found to be directly related to the geometrical coordination number z (Eq. (2)) as :

$$w(z) = \frac{\kappa}{z}, \quad (11)$$

where $\kappa = 2\sqrt{3}$.

We note that

$$\langle w^s \rangle_i = \frac{\langle \mathcal{W}_i^s \rangle_i}{V_g} - 1 = \frac{\langle \mathcal{W}_i^{\text{vor}} \rangle_i}{V_g} - 1. \quad (12)$$

Therefore, the orientational volume w^s captures the behavior of the average volume function and thus can be used as the fundamental variable to define the microstates of the system instead of the more complicated Voronoi volume.

The distribution of Eq. (9) is not the distribution that one would obtain in real packings (generated either experimentally or numerically) corresponding to the ensemble average of Eq. (9). Therefore, further examination is required to derive the distribution of orientational volumes which can be directly compared with real packings; their states determined by the compactivity, X .

Under the volume ensemble point of view [1], the observables in real packings are ensemble average over the Boltzmann distribution function. Using Eq. (4), the probability distribution of volumes in the canonical volume ensemble for a single quasiparticle of orientational volume w is then:

$$\begin{aligned}
P(w^s|X, Z) &= \frac{1}{Z_{\text{iso}}} \int_Z^6 P(w^s) \exp\left[-\frac{w(z)}{X}\right] g(z) dz \\
&= \frac{1}{Z_{\text{iso}}} \int_Z^6 \frac{1}{w(z)} \exp\left[-\frac{w^s}{w(z)}\right] \exp\left[-\frac{w(z)}{X} + \frac{2\sqrt{3}}{w(z)} \ln h_z\right] dz,
\end{aligned} \tag{13}$$

where we have used the inverse relation Eq. (11) and the exponential density of states $g(z) \sim (h_z)^z$.

This equation cannot be solved analytically for a general (X, Z) . However, analytical forms can be obtained in the limiting cases of $X = 0$ (the ground state) and $X \rightarrow \infty$: the RCP and RLP lines in the terminology of [9] respectively (see Fig. 2). The advantage of studying these distributions is that they can be checked with simulations or experiments without the use of the compactivity as a fitting parameter.

From Eq. (13), we find along the RCP line, $P_{\text{RCP}}(w^s|Z) \equiv P(w^s|X = 0, Z)$:

$$P_{\text{RCP}}(w^s|Z) = \sqrt{3} \exp\left(-w^s \sqrt{3}\right), \quad Z \in [4, 6], \tag{14}$$

and for the RLP line, $P_{\text{RLP}}(w^s|Z) \equiv P(w^s|X \rightarrow \infty, Z)$:

$$P_{\text{RLP}}(w^s|Z) = \frac{Z}{2\sqrt{3}} \exp\left(-\frac{w^s Z}{2\sqrt{3}}\right), \quad Z \in [4, 6]. \tag{15}$$

We note that both limiting distributions coincide at $Z = 6$, the frictionless J-point.

In what follows, we test the above predictions with computer simulations. We generate packings at the jamming transition using the split algorithm explained in [9]. The packings consist of 10,000 spherical equal-size soft particles interacting via Hertz normal forces, Mindlin tangential forces and the Coulomb condition with friction coefficient μ . The mechanical coordination number Z versus the volume fraction ϕ of the generated packings are plotted in Fig. 2 in the framework of the phase diagram of [9]. We change friction from $\mu = 0$ to $\mu \rightarrow \infty$ to generate the packings along the RLP line as indicated in the figure with the corresponding change in the mechanical coordination number from $Z(0) = 6$ to $Z(\infty) = 4$. The RCP line is also generated by changing friction but the volume fraction remains constant, as seen in the figure, while the mechanical coordination varies from 6 to 4.

We focus on the calculation of the probability distribution function of w^s for the packings along the RCP-line to test Eq. (14) and along the RLP-line to test Eq. (15). Figure 3 shows the results. Along the RCP-line, shown in Fig. 3a, we find all distributions are the same, independent of friction and Z , as suggested by Eq. (14). On the other hand, the distribution along the RLP line, shown in Fig. 3b, depends on friction and therefore on $Z(\mu)$ as suggested by theory. The exponential dependence seems to be captured upon a first inspection of the data done in a semi-log graph of Figs. 3a and b (arguably better for the RCP

case), at least for intermediate values and the tail of the distribution. In the case of the RLP line (Fig. 3b), the slope of the semi-log plot of the exponential fit has the same dependence on Z as predicted by theory (that is the slope in the semi-log plot increases linearly with Z) but with twice the constant value as predicted by Eq. (15). We find that the exponential fit leads to a tail with characteristic volume $= Z/\sqrt{3}$, twice the value predicted by theory $Z/(2\sqrt{3})$, Eq. (15). However, the linear trend with Z is observed in the data.

On the other hand, the average of the distributions agrees very well with simulations (see below). But when we fix the mean according to theory the exponential tail is inaccurate. Theory either provides the exponential fit with the incorrect average or provides the correct average with the deviations from the exponential fit.

The same situation is observed in Fig. 3a for the RCP line. We force the fitting to be exponential, and then the average value has to be modified from $\sqrt{3} \rightarrow 2\sqrt{3}$. The reason for this discrepancy is that the theory does not capture the distribution in the full range of volumes. Figure 3 clearly show a plateau at small values of w^s deviating from the exponential behavior predicted by the theory.

Furthermore, a more strict scrutiny of the data seems to indicate that the exponential fit may not be sufficiently accurate as shown in Figs. 3a and b. While tempting to conclude that the exponential is a good fit to the data (for instance, the fitting in Fig. 3a looks convincing), further scrutiny reveals important deviations in the tail. To visualize the deviations, one should take the plot and look at it, not frontally, but from the side. It is evident that there is a slight curvature in the distributions deviating from the linearity in the semi-log plots. Indeed, the distributions decay slightly faster than the pure exponential decay predicted by theory. The largest evidence of this deviation is perhaps in the tail of the $\mu \rightarrow \infty$ data in the RLP, Fig. 3b.

A double log analysis of the data shown in Figs. 4a and 4b reveals that a compressed exponential behaviour might better capture the tail of the distributions above the average value:

$$P(w^s|Z) \sim A \exp \left[- \left(\frac{w^s}{w_c} \right)^{\beta_w} \right], \quad w^s > w, \quad (16)$$

where $\beta_w \approx 1.5$ is the compressed exponential exponent ($\beta_w = 1$ would be a pure exponential) valid for all the RCP and RLP packings according to Fig. 4a and 4b, w_c is a characteristic volume independent of Z in the RCP packings and depends on Z for the RLP packings, and A is a constant.

We want to stress the difficulties associated with a fit to a compressed (or stretched) exponential function like Eq. (16). A double log plot gives:

$$\ln \left(- \ln (P(w^s|Z)/A) \right) = \beta_w \ln(w^s) - \beta_w \ln(w_c), \quad (17)$$

providing a linear fit with slope β_w . Beyond the inherent subtleties associated to taking a double log of a function in a such a short range, a further complication arises because such a linear fit depends on the value of the constant A . Since

Eq. (16) covers only the tail of the distribution, A cannot be obtained from normalization, remaining as a fitting parameter. The result is a fitting of β_w dependent slightly on A , providing an extra level of difficulty.

Although the compressed exponential seems to fit the data better than the pure exponential, before more theory or numerical/experimental evidence in the limit $N \rightarrow \infty$ become available, we are inclined to conclude that the problem is not closed. We note that a similar dichotomy between exponential and compressed/stretched exponential behavior has plagued the study of the distribution of forces in jammed matter since the first studies on the subject [19]. Given the inherent difficulties in any numerical estimation, the dispute will have to eventually be settled when more exact theories become available.

Beyond the distributions of volumes, the theory reproduces very well the average value of the volumes, Eq. (11). Based on the properties of the averages expressed above, there is no need to calculate the full Voronoi volume to obtain the average volume fraction, since the average of the orientational w^s suffices. For instance in the frictionless packing we find $\langle w^s \rangle = 0.561$, which gives a volume fraction $\phi = 1/(1 + \langle w^s \rangle) = 0.641$, in agreement with the direct measurement of the volume fraction of the packing, 0.64.

A full comparison between theory and simulations is given in Fig. 5, where we study the dependence between average volume and coordination number. For each packing along the RLP line we calculate the average orientational volume focusing on the particles with a given z . We also calculate the average over all the particles for a given packing, plotted as the red dots in Fig 5. In practice, we do not measure the geometrical coordination number z but the mechanical coordination number Z . However, we know that for the packings along the RLP line $z \approx Z$ [9] (this is because $h_z \rightarrow 0$). Furthermore, the RLP line corresponds to $X \rightarrow \infty$ and therefore the prediction of the average volume function, Eq. (11) can be tested directly with these fully random numerical packings, extending this result, valid for a quasiparticle, to the entire packing. Thus, the packings along the RLP line reveal the approximate behaviour of quasiparticles of fixed coordination z . We notice that there could be still some subtleties when comparing Eq. (11), valid for quasiparticles, to the results in packings. Using $\phi^{-1} = w + 1$, we plot the volume fraction in Fig. 5b.

Figure 5a shows that the mean of the distribution of volumes, w , is well captured by the theory of Eq. (11) (see the black dashed line in comparison with the red dots in Fig. 5). This is why the theory provides very good fittings to the values of RCP and RLP in [9]. The agreement can be seen as well in the volume fraction in Fig. 5b, and exists despite the fact that the full distribution presents the deviations discussed above.

Figure 5 presents further interesting results. For a given packing along the RLP-line specified by a fixed friction, there are a variety of particles with varying coordination z , following a well-defined functional relation between the volume occupied by the particle and its coordination number (see for instance the red and blue dashed lines in Fig. 5a corresponding to fittings for the cases $\mu = 0$ and $\mu \rightarrow \infty$, respectively). While this plot does not tell us how many particles there are for a given coordination number (see next section) we see that for each

packing, there exists a variety of local volume functions with z ranging from $z = 0$ (since there are some rattlers) up to $z = 11$ (but not 12, interestingly, see below).

The assumptions for the limits of integration in the partition function in [9] or the ensemble average Eq. (3), $Z \leq z \leq 6$, seem to be violated here. However, we have to remember that the theory is mesoscopic and further coarse-graining is needed for these bounds to be more accurate. Regardless, even though the range in z extends further than the bounds, Fig. 5 corresponds to the average for a fixed z but does not tell how many states there are for every z . When these details are properly taken into account, the bounds are approximately satisfied, although fluctuations persist, bringing us to the next Section of this paper.

Focusing around the $z = 12$ point in the figures, we observe that an extrapolation of the fitting to the curve of frictionless packing seems to converge to the green dot in Figs. 5 at $z = 12$ which correspond to the free volume function of FCC, $w_{\text{FCC}} = 0.35135$ (Fig. 5a) and the FCC volume fraction $\phi_{\text{FCC}} = \pi/\sqrt{18} \approx 0.7402$ (Fig. 5b). An extrapolation of the fitting to the infinite friction data seems to pass through the volume fraction of the dodecahedron as indicated by the blue point in Fig. 5, which has also $z = 12$ but slightly larger volume fraction than FCC (the dodecahedron can't tile the space without leaving holes, so the best global packing is still the FCC). We observe that there are no particles in the packings with $z = 12$. Indeed the green and blue points at $z = 12$ in Figs. 5a and b were added by hand and the real curves stop shortly at $z = 11$. The absence of $z = 12$ states indicate the randomized state of the systems.

More importantly, we see that if we extend the theoretical result of Eq. (11) to the ordered region, examining z from $z = 6$ to $z = 12$, the theory does not fit the FCC value. Instead, we obtain $w(z = 12) = 2\sqrt{3}/12 = 0.2886$, below the FCC or dodecahedron value. In principle, this result is expected since the theory assumes random isotropic states while the FCC is an ordered anisotropic packing. However, the absence of a good fitting of the disordered branch through the FCC, together with the fact that the theory fits so well the disordered states, raises the interesting question of the existence of a phase transition between the RCP limit at $z = 6$ and the FCC at $z = 12$. Since packings cannot equilibrate above $z = 6$ without the formation of crystalline regions, we expect an ordered branch from the FCC point towards the RCP point. It seems plausible that there could be a discontinuity from the disordered branch to the ordered branch, the existence of which could determine whether there exist a disorder/order phase transition characterizing the RCP. This scenario has been confirmed by analysis of numerical packings in a recent study [20], which showed that RCP can be interpreted as a “freezing point” in a first-order phase transition between ordered and disordered phases.

To summarize this part of the study, while theory predicts an exponential behavior and approximates well some features of the distribution such as the average value, simulations indicate that a compressed exponential fitting could be also possible. We therefore require more refined theories to account for the full behavior of the volume distribution. The present approach suggests that study

of the ensemble of quasiparticles of fixed coordination number could provide clues to the behavior of the entire system through the Edwards ensemble approach. While new theoretical concepts are required, current attempts indicate that it might be possible to develop a theory of the volume distribution that is exact to, at least, a given coordination shell of particles. It appears that $P(w^s)$ for a fixed z -ensemble might be solved exactly by a brute force approach, since the range of a Voronoi cell is finite. Although, it may contain a large number of variables, the computer should handle such a computation. Such an analysis parallels the Hales proofs of the Kepler's conjecture [21].

4. PDF of the coordination number

Next, we analyze the distribution of coordination number by generalizing the theory of [9] to include fluctuations in z . While we have assumed [9] that every single quasiparticle satisfies the specified bounds: $Z \leq z \leq 6$, below we relax this constraint to extend the bounds to the geometrical coordination of the entire system by considering:

$$N z_{\min} \leq \sum_{i=1}^N z_i \leq N z_{\max}, \quad (18)$$

where in the following we set $z_{\min} = Z$ and $z_{\max} = 6$. This new condition implies that it is not possible to consider the single quasiparticle partition function, Eq. (3), and that the full N -particle partition function has to be considered:

$$\begin{aligned} \mathcal{Z} &= \int \dots \int_{N z_{\min} \leq \sum_{i=1}^N z_i \leq N z_{\max}} \prod_{i=1}^N e^{-w_i(z_i)} g_i(z_i) dz_i \\ &= \int \dots \int_{N z_{\min} \leq \sum_{i=1}^N z_i \leq N z_{\max}} \prod_{i=1}^N e^{-(z_i/z^* + \beta \kappa/z_i)} dz_i. \end{aligned} \quad (19)$$

We have $z^* = -1/\ln h_z$, the inverse compactivity $\beta = 1/X$ and the coupling constant

$$B = \frac{\beta \kappa}{z^*}. \quad (20)$$

Then, the ensemble average of the probability distribution function of the geometrical coordination number, $P(z)$, is

$$\begin{aligned} P(z) &\equiv \left\langle \frac{1}{N} \sum_{i=1}^N \delta(z - z_i) \right\rangle \\ &= \frac{1}{\mathcal{Z}} \int \dots \int_{(N z_{\min} - z) \leq \sum_{i=1}^{N-1} z_i \leq (N z_{\max} - z)} \prod_{i=1}^{N-1} e^{-(z_i/z^* + \beta \kappa/z_i)} dz_i, \end{aligned} \quad (21)$$

We obtain:

$$P(z) = \frac{\text{Erf}\left(\frac{\sqrt{N-1}(z'_{\max}-\mu)}{\sqrt{2}\sigma}\right) + \text{Erf}\left(\frac{\sqrt{N-1}(\mu-z'_{\min})}{\sqrt{2}\sigma}\right)}{\text{Erf}\left(\frac{\sqrt{N}(z_{\max}-\mu)}{\sqrt{2}\sigma}\right) + \text{Erf}\left(\frac{\sqrt{N}(\mu-z_{\min})}{\sqrt{2}\sigma}\right)} e^{-(z/z^* + \beta\kappa/z)}, \quad (22)$$

with the following constants:

$$\begin{aligned} z'_{\max} &\equiv (Nz_{\max} - z)/(N-1) \approx z_{\max} + (z_{\max} - z)/N, \\ z'_{\min} &\equiv (Nz_{\min} - z)/(N-1) \approx z_{\min} + (z_{\min} - z)/N, \end{aligned} \quad (23)$$

and $\text{Erf}(x)$ the Gauss error function.

The constants μ and σ in Eq. (22) are significant because they represent the mean and standard deviation of the a Gaussian expansion in a saddle-point approximation of the inverse Fourier transform of the partition function allowing the calculation of the free-volume density. They are:

$$\mu = z^* B^{1/2} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})}, \quad (24)$$

which is the same as the ensemble average of the coordination number, and

$$\sigma^2 \equiv z^{*2} B \left(\frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} - \frac{K_1(2B^{1/2})^2}{K_0(2B^{1/2})^2} \right), \quad (25)$$

where $K_n(a)$ is the modified Bessel function of the second kind:

$$\int_0^\infty x^n e^{-\frac{a}{2}(x+1/x)} dx = 2K_n(a). \quad (26)$$

Next, we consider the approximations of Eq. (22) for two cases: When $z_{\max} - \mu < \mu - z_{\min}$, then $\mu > (z_{\max} + z_{\min})/2$, and we obtain:

$$P(z) \sim \exp \left[-\frac{1}{z^*} \left(z(2 - \bar{w}/w_{\min}) + \frac{1}{z} \frac{\kappa^2}{\bar{w}^2} \right) \right], \quad (27)$$

or otherwise, we obtain:

$$P(z) \sim \exp \left[-\frac{1}{z^*} \left(z(2 - \bar{w}/w_{\max}) + \frac{1}{z} \frac{\kappa^2}{\bar{w}^2} \right) \right], \quad (28)$$

where $\bar{w}$ is the ensemble average of the volume function (which depends on β , or compactivity X), $w_{\min} = \kappa/z_{\max}$ and $w_{\max} = \kappa/z_{\min}$.

In the following we consider the distribution functions at two special points on the phase diagram and compare them to the numerical simulations. At the frictionless J-point and the infinitely frictional L-point (see Fig. 2), the distributions reduce to simple forms. For the RCP J-point, $X = 0$ and the system has the minimum average volume and maximum average coordination number, therefore, $\bar{w} \sim w_{\min}$ and $u \sim z_{\max}$. From Eq. (27) We find:

$$P_{\text{RCP}}(z) \sim \exp \left[-\frac{1}{z^*} \left(z + \frac{z_{\max}^2}{z} \right) \right]. \quad (29)$$

For the RLP L-point, $X \rightarrow \infty$ and the system has the maximum average volume and minimum average coordination number, therefore, $\bar{w} \sim w_{\max}$ and $u \sim z_{\min}$. From Eq. (28), we find:

$$P_{\text{RLP}}(z) \sim \exp \left[-\frac{1}{z^*} \left(z + \frac{z_{\min}^2}{z} \right) \right]. \quad (30)$$

We test these forms with the computer generated packings at the J-point and L-point. The constant z^* is treated as a fitting parameter since it determines the density of states and is difficult to know a priori. Figure 6 shows the result. The lin-lin plot of Fig. 6a shows that the distribution near the average value is well approximated by the theory for both points. To investigate the tails of the distributions, Fig. 6b plots a semi-log curve. While some deviations are observed, the fit is still reasonable except for the larger coordination number of J-point and smaller coordination number of L-point, which are very rare, about 10^{-2} less probable than the most probable value.

5. Conclusions

We have presented the predictions of the mesoscopic theory presented in [9] concerning the probability distribution of the orientational volumes in jammed matter. The theory captures very well the average volume and indeed gives rise to good predictions of the RCP and RLP volume fraction as shown in [9]. However, when comparing the full distribution we find important deviations. For instance, computer simulations are able to detect slight deviations from the pure exponential behaviour predicted by theory. This deviation could be better approximated by a compressed exponential behavior, although a more conclusive fitting necessitates a more precise theory or simulations in the large scale limit. The plateau observed in the volume distribution is not captured by the theory either. There is evidence that this plateau might originate from the spatial correlations between the first and second layer particles, which indicates that further progress could come from a systematic analysis of higher level coarse-graining. For example, by explicitly treating the second-layer neighbors, it is possible to improve the distribution functions predicted by the theory. This approach is particularly appropriate for two-dimensional systems, since the free variables are significantly reduced in the 2d case.

On the other hand, the behavior of the probability distribution of coordination number is captured well by the theory. Here, the mesoscopic theory of [9], considering restricted bounds for each quasiparticle, is extended to allow for coordination number fluctuations, by imposing the bounds to the entire system. Such a problem can be solved under several approximations and predicts a mixed exponential forms that are well reproduced by the simulations at the J and the L-point.

Why does the theory capture the contact distribution better than the volume distribution? The distribution of contacts is an ensemble average based on the states characterized by $w(z)$. Since this function is quite accurate, the contact

distribution follows. On the other hand, the volume distribution is based on the uniform approximations done on [17], and therefore is not quite exact. The full distribution of volumes is where more theoretical developments are required.

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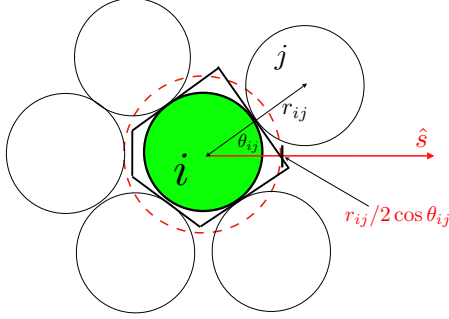


Figure 1: Schematics of the Voronoi volume and the orientational volume associated with particle i . The boundary of the Voronoi cell (shown in 2d for simplicity) corresponds to the irregular pentagon in black which defines $\mathcal{W}_i^{\text{vor}}$. The limit of the Voronoi cell of particle i in the direction $\hat{s}$ is the minimum of $r_{ij}/2 \cos \theta_{ij}$ over all the particles in the packing, as indicated. This defines the orientational volume $\mathcal{W}_i^s$ which is the volume of the sphere of radius $r_{ij}/2 \cos \theta_{ij}$ defined by the dash red circle in the figure. The Voronoi volume is the integration of the orientational volume over $\hat{s}$ as in Eq. (6). Notice that w^s , the orientational free volume associated with $\mathcal{W}_i^s$ defined in Eq. (8), ranges from zero (when the orientational volume coincides with the volume of the central ball, that is when the direction $\hat{s}$ coincides with a contact point) to in principle ∞ for an isolated ball, although the maximum w^s in a jammed system is, of course, bounded by the free space given by the first or second coordination shell. The Voronoi free volume, on the other hand, cannot be zero, by definition.

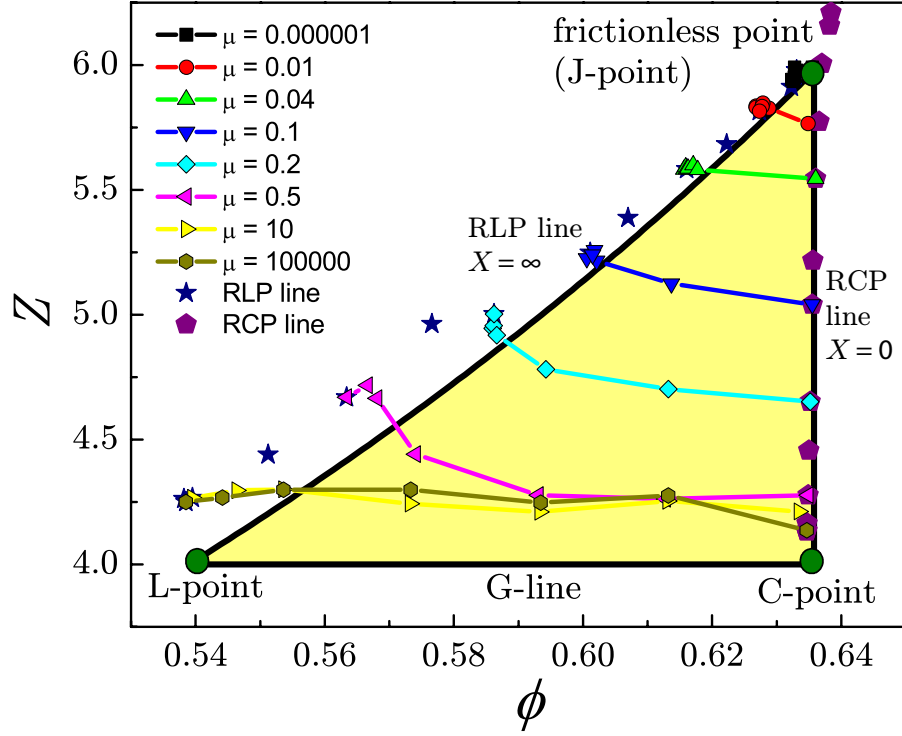


Figure 2: Computer generated packings arranged in the phase diagram of [9]. The packings are used to calculate the distribution of orientational volumes and coordination number. We concentrate our study on packings generated with different friction as indicated in the figure following the RLP line from the J point to the L point and the RCP line from the J point to the C point.

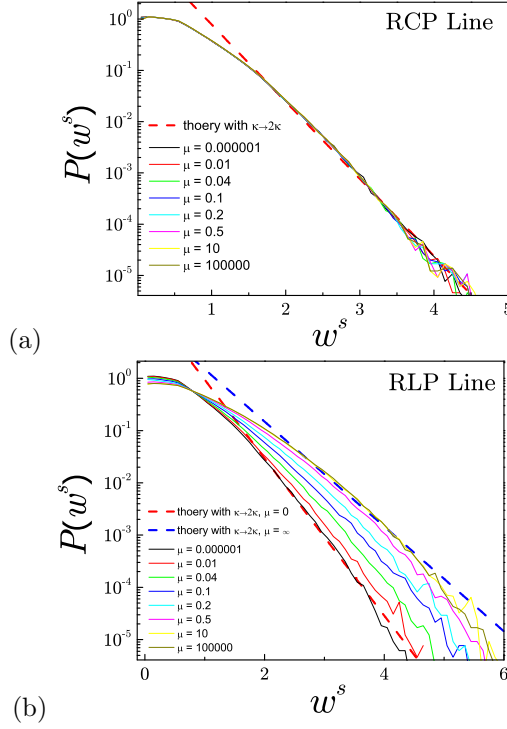


Figure 3: PDF of w^s . Exponential fit in a semilog plot. (a) We plot the results for the distribution of the packings along the RCP line with different values of friction, as indicated in the figure (see Fig 2). The red dashed line is a fit with the theoretical prediction $P_{\text{RCP}}(w^s)$, Eq. (14), but with the inverse characteristic volume or slope of $2\sqrt{3}$ instead of $\sqrt{3}$ as predicted by the theory. (b) Same as (a) but for the packings along the RLP line in Fig. 2 prepared with different μ . The red and blue dashed lines are fits to the theoretical prediction $P_{\text{RLP}}(w^s)$, Eq. (15), but with the inverse characteristic volume or slope replaced from $Z/(2\sqrt{3}) \rightarrow Z/\sqrt{3}$, twice as predicted by the theory like in (a). The red line is for $\mu = 0$ and has slope $2\sqrt{3}$. The blue line is for $\mu \rightarrow \infty$ and has slope $4/\sqrt{3}$.

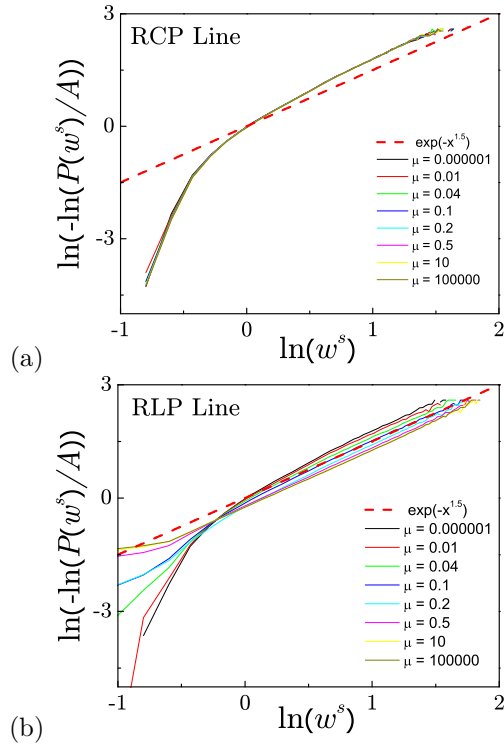


Figure 4: PDF of w^s . Compressed exponential fit in a double log plot. The results are the same as in Fig. 3 but now replotted in a double log plot to obtain the compressed exponential fitting exponent β which is extracted from the slope of such a plot, as explained in the text. (a) PDF for the packings along RLP-line. (b) PDF for the packings along the RLP-line.

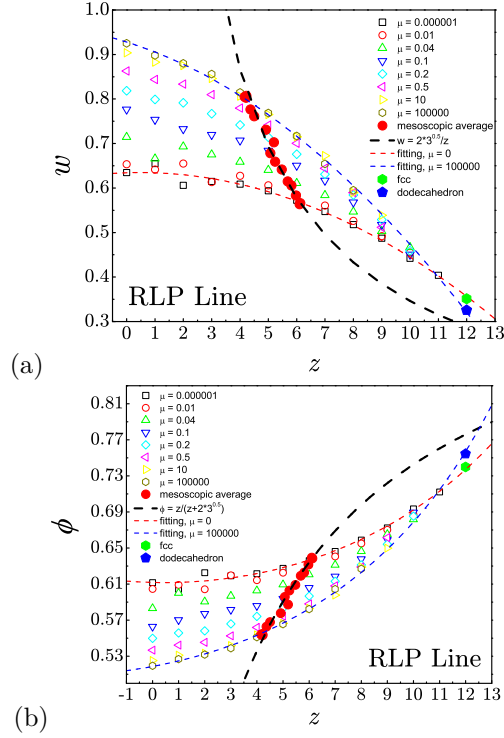


Figure 5: Relation between local volume per particle and its coordination number for (a) the volume function and (b) the volume fraction. We use the packings along the RLP line with the friction as indicated and we calculate the volume function binning the data by the coordination number of the particles. The red dots correspond to the average over all the particles in the packing of the volume function which fits the theory very well. The red and blue dashed lines are logarithmic-like fittings to the data and we add the value at $z = 12$ for the FCC and the dodecahedron. The data from the packings goes only up to $z = 11$, indicating the absence of ordered structures.

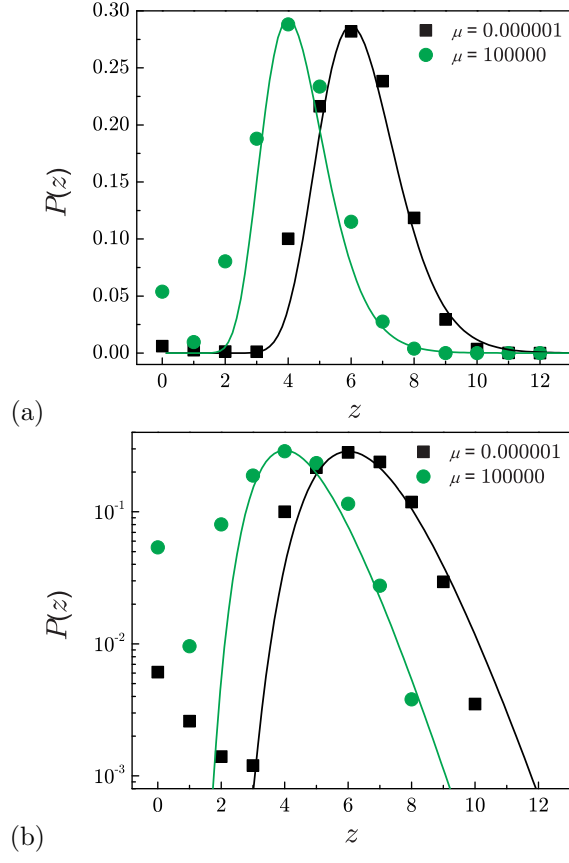


Figure 6: $P(z)$: comparison between theory and simulations in (a) a lin lin plot and (b) a semi-log plot to appreciate better the tail of the distribution. We plot the systems at the J-point ($\mu = 0$) and L-point ($\mu = \infty$) in Fig. 2. we use a fitting parameter $z^* = 0.5$ in Eqs. (29) and (30).

Irreversible Incremental Behavior in a Granular Material

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We test the elasticity of granular aggregates using increments of shear and volume strain in a numerical simulation. We find that the increment in volume strain is almost reversible, but the increment in shear strain is not. The strength of this irreversibility increases as the average number of contacts per particle (the coordination number) decreases. For increments of volume strain, an elastic model that includes both average and fluctuating motions between contacting particles reproduces well the numerical results over the entire range of coordination numbers. For increments of shear strain, the theory and simulations agree quite well for high values of the coordination number.

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Granular materials have received the attention of many researchers in the last decade because of unsolved problems with direct relevance to chemistry, physics and engineering. The behavior of a granular material can range between that of a gas and that of a solid, depending on the applied loading and the regime of deformation considered. Significant progresses have been made with the introduction of numerical tools (e.g. [1]) that permit the detailed analysis of an aggregate of particles. Although such simulations have provided information about inter-particle interactions, such as their elasticity, sliding, and deletion, and statistical measures of their cooperative behavior, such as induced anisotropy and force chains, it is still unclear how to incorporate these informations in a predictive theoretical model.

Attempts to do this have been made in the context of the effective medium theory (EMT) (e.g. [2, 3]) in which the contact displacements are given by the applied average strain. However, the predicted shear and bulk moduli are far from those measured in numerical simulations (e.g. [4, 5]). In order to improve the theoretical prediction, particle displacements are given by the sum of an average and the fluctuation components (e.g. [6, 7]). In particular, better predictions of the shear and bulk moduli have been obtained in a recent work that employs pair fluctuations [8, 9]. However, Agnolin and Roux [10] point out that such predictions fail when the coordination number, $\bar{Z}$, is low and close to the isostatic limit $\bar{Z}_{iso}$ at which the aggregate is statically determinate. They suggest that for low coordination number, more complicated models are needed that account for collective deformations among particles. That is, the assumption of pair fluctuations is not sufficient to capture the response of poorly coordinate aggregates of particles. Here, we address this issue and provide an alternative interpretation of the failure of an elastic description at low coordination numbers.

We carry out numerical simulations using a distinct element method and focus on the first incremental response of an isotropically compressed random aggregate that consists of identical, elastic, frictional spheres. We consider dense aggregates, with solid volume fractions ϕ near 0.64, that have different $\bar{Z}$. Previous work [10–12] have considered poorly coordinated aggregates and found that the shear modulus is proportional to $\bar{Z} - \bar{Z}_{iso}$, where $\bar{Z}_{iso}$ is equal to four for a packing of frictional spheres. Here, we find that when $\bar{Z}$ decreases, there is an irreversible behavior of the aggregate that involves local, coordinated, irreversible motions of the particles that are not resisted by forces. These motions result in a reduction of the apparent stiffness of the aggregate (e.g. [13]). That is, the initial configuration of a poorly coordinated aggregate can not sustain any incremental strain unless a change in the geometry of the packing occurs. When such irreversible changes are present, simple elastic theory can not reproduce the response of the aggregate and the utility of the elastic moduli is questionable.

We introduce measures of these irreversible deformations which vary with the coordination number. For increments in volume strain, the number of irreversible motions is so small that their effect is negligible and the response of the aggregate can be assumed to be elastic. More importantly, for shear increments, the strength of the irreversibility persists even at high $\bar{Z}$ and increases as $\bar{Z}$ decreases towards its isostatic value, indicating that elasticity does not describe the aggregate response.

Our numerical simulations consider 10,000 particles, each with diameter $d = 0.2$ mm, randomly generated in a periodic cubic cell. We employ material properties typical of glass spheres: a shear modulus $\mu = 29$ GPa and a Poisson's ratio, $\nu = 0.2$. The interaction between particles is a non-central contact force in which the normal component is the non-linear Hertz interaction and the tangential component is bilinear: an initial elastic

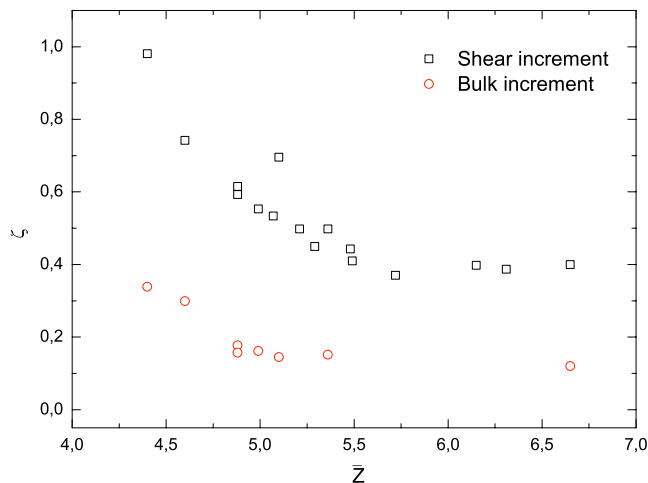


FIG. 1: Measurement of irreversibility in terms of displacements when increments of volume and shear strain are applied.

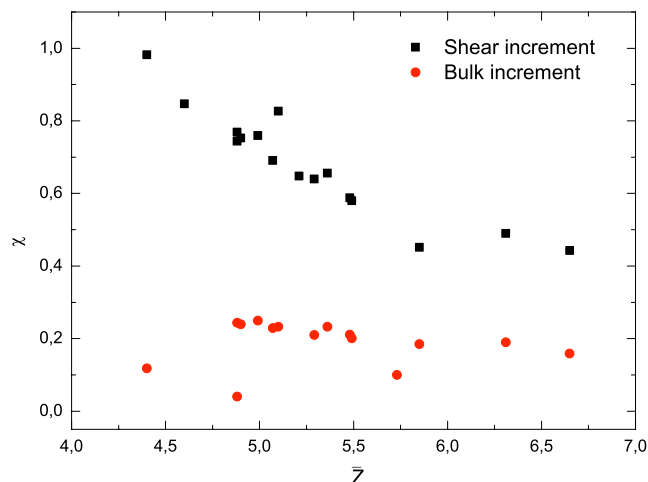


FIG. 2: Measurement of irreversibility in terms of contact forces when increments of volume and shear strain are applied.

displacement followed by Coulomb sliding (e.g.[5]). We create different initial isotropic states by varying the coefficient of friction between particles during the preparation (e.g. [14]); all initial states have a solid volume fraction $\phi \sim 0.64$ and a confining pressure that varies from 50KPa to 10MPa .

For all packings, we evaluate the response of the aggregate to homogeneous increments in volume and shear strain by setting the particle coefficient of friction high enough to prevent sliding. Because of the random positions of the particles and their different initial contact stiffnesses, the subsequent particle motions are the sum of the homogeneous applied strain and a fluctuation that relaxes the particles towards a new equilibrium state. When this relaxation involves a rearrangement of particles, the incremental response is irreversible.

We attempt to characterize the particle rearrangement and introduce two measures of the strength of the irreversibility: the first is related to contact displacement,

and the second is associated with contact forces. We apply a forward increment in strain followed by an identical backward strain in which the reference configuration should be recovered if the deformation were perfectly elastic.

We define the parameter ζ as the average over all contacts of the ratio of the absolute values of the contact displacements after and before the backward increment in strain. A parameter, χ , is similarly defined in terms of the contact forces. Both parameters would be zero for perfectly elastic behavior.

We measure ζ and χ in all of the packings for increments in both shear and volume strain. We apply increments of strain with magnitudes that depend on the confining pressure; the ratio of the associated volume strain with the confining pressure is constant at about 10^{-2} . The results are plotted in Figs. 1 and 2, displaying ζ and χ as functions of $\bar{Z}$.

Figs. 1 and 2 show small variations in ζ and χ associated with increments in volume strain. The strength of the irreversibility is almost negligible, with slight increases as $\bar{Z}_{iso}$ is approached. We conclude that an approximate elastic response for the aggregate is obtained for increments of volume strain, independent of the coordination number.

For increments in shear strain, both ζ and χ increase as $\bar{Z}_{iso}$ is approached. We believe that these irreversible motions are associated with the presence of local instability (e.g. [13]). Moreover, both ζ and χ are non-zero for high values of the coordination number, in contrast to what we find for increments in volume strain. The aggregate seems to experience rearrangements over the entire range of the coordination number, with the irreversibility becoming stronger as isostaticity is approached.

The condition of isostaticity has been an object of great interest for many researchers ([5, 11, 12, 15–17]). The contact forces in an aggregate are uniquely determined in terms of the applied loads, independent of the contact stiffness, when the aggregate is both statically and kinematically determinate (e.g. [18]). The condition for static determinacy, often referred to as Maxwells condition [19], insures the equality between the number of unknowns and the number of equilibrium equations. In a granular aggregate, this necessitates that $\bar{Z} = \bar{Z}_{iso}$. The condition for kinematic determinacy insures that there are no inextensional mechanisms in the aggregate; then a rigid aggregate is able to sustain any external self-equilibrated perturbation without changing the relative positions of its particle centers.

The kinematic condition is not often taken into account in recent work on granular aggregates (e.g. [20]) and sometimes has been emphasized in a different way. For example, Moukarzel [13], in his description of network rigidity, defines an isostatic system to be one in which the rank K of the rigidity matrix that relates the contact forces to the applied forces always equals to the number of equations, rather than one in which the simple Maxwell condition is satisfied. Then, when K is less than the

number of equations, the network is flexible; this can occur when $\bar{Z} = \bar{Z}_{iso}$.

The presence of irreversibility in all of our aggregates, even in the limit that $\bar{Z} = 4$ (see Figs. 1 and 2) indicates that the kinematic condition does not hold. Consequently, inextensional mechanisms and the associated soft or floppy vibrational modes are possible. This situation may occur whatever the value $\bar{Z}$, if particles location do not correspond to those of a rigid network. In particular, in packing characterized as isostatic by $\bar{Z} = \bar{Z}_{iso}$ (e.g. [11, 15, 16]), soft modes can occur as long as the kinematic condition is not satisfied. However, we should emphasize that the initial states that we employ are constructed in a much different way than those constructed to insure an initial, stable, elastic response (e.g. [21]).

We now turn from irreversibility to elasticity and report results from numerical simulation for the bulk modulus, $\bar{\Theta} = \frac{1}{3} \sum_{i=1}^3 \Delta\sigma_{ii}/\Delta V$, and the shear modulus, $\bar{G} = \Delta\sigma_{12}/\Delta\varepsilon_{12}$, where σ_{ij} is the average stress tensor, ε_{ij} the average strain tensor, and V is the volume of the aggregate. When a shear increment is applied to poorly coordinated systems, the irreversibility increases and the material response deviates from elasticity. That is, an elastic theory can only be considered as providing an upper bound on the shear modulus.

To make predictions of the moduli, we adopt the fluctuation theory developed by [9]. This theory improves upon the simplest average strain models [2, 3], because contacting particles are assumed to move with both the average deformation and fluctuations and because the statistics of the aggregate are taken in account. The theory employs force and moment equilibrium for a typical pair of particles to evaluate the fluctuations, and then uses them to determine the stress in the aggregate. However, at low values of the coordination number, the simple statistical model introduced to describe the variability of the neighborhood of contacting pairs of particles is probably too simple, (e.g. [22]) and the initial distribution contact forces (e.g.[23]) is not taken into account.

Here we repair the second deficiency and assume that the distribution, $w(P)$, of the normal component of the contact force, P is exponential (e.g.[4]):

$$w(P) = \frac{1}{\bar{P}} \exp\left(-\frac{P}{\bar{P}}\right), \quad (1)$$

where, by definition, $\bar{P} = \int_0^\infty P w(P) dP$.

From the effective medium theory (e.g.[24]), the confining pressure p_0 can be expressed as function of $\bar{P}$,

$$p_0 = \bar{Z}\phi\bar{P}/\pi d^2,$$

and the relation between the normal component of the contact displacement, δ , and $\bar{P}$ is

$$\delta = \left[\frac{3(1-\nu)}{2\mu d^{1/2}} \bar{P} \right]^{2/3}. \quad (2)$$

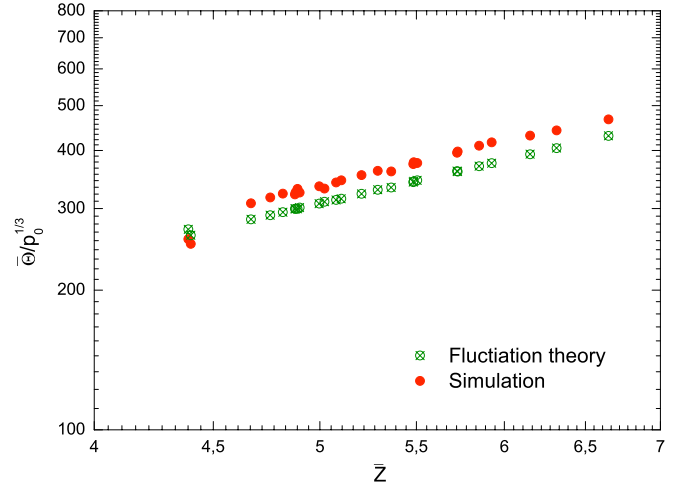


FIG. 3: Comparison between the numerical data and fluctuation theory for the normalized bulk modulus.

Using (1) in (2), we obtain

$$\begin{aligned} \bar{\delta}^{1/2} &= \left[\frac{3(1-\nu)}{2\mu d^{1/2}} \right]^{1/3} \int_0^\infty P^{1/3} w(P) dP \\ &= \left[\frac{3(1-\nu)}{2\mu d^{1/2}} \right]^{1/3} \bar{P}^{1/3} \Gamma\left(\frac{4}{3}\right), \end{aligned} \quad (3)$$

where Γ is the Gamma function.

So the average normal contact stiffness $\bar{K}_N = \mu d^{1/2} \bar{\delta}^{1/2} / (1-\nu)$ becomes

$$\bar{K}_N = d \left(\frac{1}{3} \right)^{1/2} \left[\frac{9\sqrt{3}\pi\mu^2}{2\bar{Z}(1-\nu)^2\phi} p_0 \right]^{1/3} \Gamma\left(\frac{4}{3}\right). \quad (4)$$

The average shear stiffness is $\bar{K}_T = 2\bar{K}_N(1-\nu)/(2-\nu)$.

Taking in account the initial distribution of forces in this way, we obtain a new solution for the fluctuations; with this, the resulting expressions for the bulk modulus $\bar{\Theta}$ and the shear modulus $\bar{G}$ are, respectively,

$$\bar{\Theta} = \frac{\phi\bar{K}_N}{5\pi d} \left[-2.8\bar{Z} - \frac{14.5}{\bar{Z}^3} + \frac{38}{\bar{Z}^2} - \frac{33.6}{\bar{Z}} - 12.7 \right] \quad (5)$$

$$\begin{aligned} \bar{G} &= \frac{\phi(\bar{K}_N - \bar{K}_T)}{5\pi d} \left[1.7\bar{Z} + \frac{8.7}{\bar{Z}^3} - \frac{22.8}{\bar{Z}^2} + \frac{20.2}{\bar{Z}} - 7.6 \right] \\ &\quad + \frac{\phi\bar{K}_T}{5\pi d} \left[6.6\bar{Z} + \frac{66.9}{\bar{Z}^3} - \frac{154.7}{\bar{Z}^2} + \frac{128.6}{\bar{Z}} - 46 \right]. \end{aligned} \quad (6)$$

where a relation between the rms value of the fluctuation in the number of contacts per particle and its average value has been adopted [14]. At the upper limit of validity of equations (5,6), $\bar{Z} = 22/3$, we recover the average strain prediction [2, 3]. This limit results from

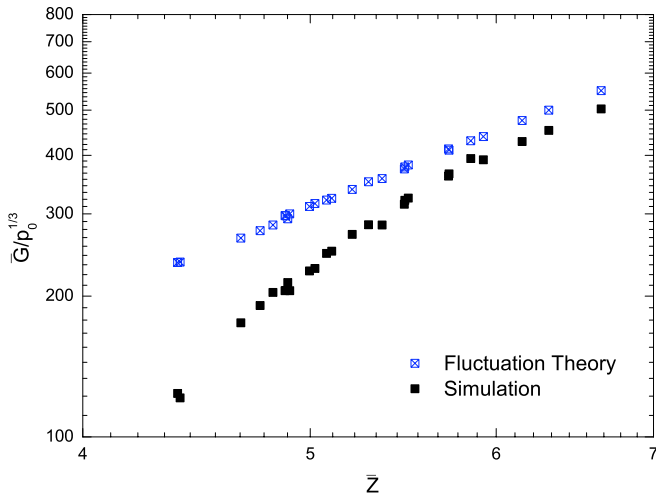


FIG. 4: Comparison between the numerical data and fluctuation theory for the normalized shear modulus.

the modeling of the statistical distribution of particles in the assembly.

Comparison between the predictions of the bulk and shear moduli and measurements in the numerical simulation are shown in Figs. 3 and 4. There is agreement at high values of the coordination number for the shear modulus; the slight difference may be attributed to the irreversibility. However, when the comparison is extended

to lower values of $\bar{Z}$, the prediction strongly deviate from the simulations, as also seen by others [10]. Here, we conclude that this discrepancy is due to the observed irreversible motions and that an elastic theory is not able to capture the behavior of the system for low values of $\bar{Z}$. For the bulk modulus, the theory works quite well over the entire range of $\bar{Z}$, because the irreversibility, measured by χ and ζ , can be neglected.

In conclusion, we have measured the irreversible motions in a granular aggregate subjected to incremental strains and determined their influence on the mechanical response of the material. We found that particles may experience rearrangements in their geometry and contact forces, even when small perturbations are applied and that these rearrangements are sensitive to the coordination number. The strength of this irreversibility is negligible for increments in volume strain, while it strongly increases for increments in shear strain as $\bar{Z}$ approaches $\bar{Z}_{iso}$. The presence of irreversible motions in the aggregates indicates a deviation from elastic behavior. That is, an elastic theory is appropriate to describe the material behavior only if mechanisms do not play an important role in the displacements of the particles.

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Edwards thermodynamics of the jamming transition for frictionless packings: ergodicity test and role of angoricity and compactivity

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This paper illustrates how the tools of equilibrium statistical mechanics can help to explain a far-from-equilibrium problem: the jamming transition in frictionless granular materials. Edwards ideas consist of proposing a statistical ensemble of volume and stress fluctuations through the thermodynamic notion of entropy, compactivity, X , and angoricity, A (two temperature-like variables). We find that Edwards thermodynamics is able to describe the jamming transition (J-point). Using the ensemble formalism we elucidate the following: (i) We test the combined volume-stress ensemble by comparing the statistical properties of jammed configurations obtained by dynamics with those averaged over the ensemble of minima in the potential energy landscape as a test of ergodicity. Agreement between both methods supports the idea of “thermalization” at a given angoricity and compactivity. (ii) A microcanonical ensemble analysis supports the idea of maximum entropy principle for grains. (iii) The intensive variables describe the approach to jamming through a series of scaling relations as $A \rightarrow 0^+$ and $X \rightarrow 0^-$. Due to the force-volume coupling, the jamming transition can be probed thermodynamically by a “jamming temperature” T_J comprised of contributions from A and X . (iv) The thermodynamic framework reveals the order of the jamming phase transition by showing the absence of critical fluctuations at jamming in observables like pressure and volume. (v) Finally, we elaborate on a comparison with relevant studies showing a breakdown of equiprobability of microstates.

The application of concepts from equilibrium statistical mechanics to out of equilibrium systems has a long history of describing diverse systems ranging from glasses to granular materials [1–3]. For dissipative jammed systems—particulate grains or droplets—the key concept proposed by Edwards is to replace the energy ensemble describing conservative systems by the volume ensemble [3]. However, this approach alone is not able to describe the jamming point (J-point) for deformable particles like emulsions and droplets [4–7], whose geometric configurations are influenced by the applied external stress. Therefore, the volume ensemble requires augmentation by the ensemble of stresses [8–11]. Just as volume fluctuations can be described by compactivity, the stress fluctuations give rise to an angoricity, another analogue of temperature in equilibrium systems.

In the past 20 years since the publication of Edwards work there has been many attempts to understand and test the foundations of the thermodynamics of powders and grains. Three approaches are relevant to the present study:

1. Experimental studies of reversibility.— Starting with the experiments of Chicago which were reproduced by other groups [12–15], a well-defined experimental protocol has been introduced to achieve reversible states in granular matter. These experiments indicate that systematically shaken granular materials show reversible behavior and therefore are amenable to a statistical mechanics approach, despite the frictional and dissipative character of the material. These results are com-

plemented by direct measurements of compactivity and effective temperatures in granular media [12, 14, 16–18].

2. Numerical test of ergodicity.— Numerical simulations compare the ensemble average of observables with those obtained from direct dynamical measures in granular matter and glasses. These studies [19–24] find general agreement between both measures and, together with the experimental studies of reversibility [12–15], suggest that ergodicity might work in granular media.

3. Numerical and experimental studies of equiprobability of jammed states.— Exhaustive searches of all jammed states are conducted in small systems to test the equiprobability of jammed states, as a foundation of the microcanonical ensemble of grains. Numerical simulations and experiments indicate that jammed states are not equiprobable [25–27]. These results suggest that a hidden extra variable [28] might be needed to describe jammed granular matter in contrast with the work in 1 and 2.

The current situation can be summarized as following: When directly tested or exploited in practical applications, Edwards ensemble seems to work well. These include studies where ensemble and dynamical measurements are directly compared, and recent applications of the formalism to predict random close packing of monodisperse spherical particles [29, 30], polydisperse systems [31], and two [32] and high dimensional systems

[33]. However, a direct count of microstates reveals problems at the foundation of the framework manifested in the breakdown of the flat average assumption in the microcanonical ensemble [25–28].

In this paper we investigate the Edwards ensemble of granular matter focusing on describing the jamming transition [4–7]. A short version of this study has been recently published in [34]. We employ a strategy that mixes the approaches 2 and 3 above. We first perform an exhaustive search of all jammed configurations in the Potential Energy Landscape (PEL) of small frictionless systems in the spirit of [25–28]. We then use this information to perform a direct test of ergodicity in the spirit of [19–24]. Our results indicate: (i) The dynamical and ensemble measurements of pressure, coordination number, volume, and distribution of forces agree well, supporting ergodicity. A microcanonical ensemble analysis supports also a maximum entropy principle for grains. (ii) Intensive variables like angoricity, A , and compactivity, X , describe the approach to jamming through a series of scaling relations. Due to the force-volume coupling, the jamming transition can be probed thermodynamically by a “jamming temperature” T_J comprised of contributions from A and X . (iii) These intensive variables elucidate the thermodynamic order of the jamming phase transition by showing the absence of critical fluctuations above jamming in static observables like pressure and volume. That is, the jamming transition is not critical and there is no critical correlation length arising from a thermodynamic n -point correlation function. We discuss other possible correlation lengths. (iv) Surprisingly, we reproduce the results of [25] regarding the failure of equiprobability of microstates while obtaining the correct dynamics measurements as in [19–24]. We then offer a possible solution to this conundrum to elucidate why the microstates seems to be not equiprobable while the ensemble averages produce the correct results.

The paper is organized as follows. Section I discusses the Edwards thermodynamics of the jamming transition. Section II describes the ensemble calculations in the Potential Energy Landscape formalism. Section III describes the Hertzian system to be studied. Section IV describes the ensemble measurements to be compared with the MD measures of Section V. Section VI explains how to calculate A from the data. The ergodicity test is made in Section VII. Section VIII describes the calculation in the microcanonical ensemble where the principle of maximum entropy is verified and the coupled jamming temperature is obtained. Section IX compares our results with those of O’Hern *et al.* [25] and Section X summarizes the work. Appendix A includes “de yapa” a study of coordination number fluctuations in the Edwards theory for random close packings of hard spheres.

I. EDWARDS THERMODYNAMICS AND THE JAMMING TRANSITION

The process typically referred to as the jamming transition occurs at a critical volume fraction ϕ_c where the granular system compresses into a mechanically stable configuration in response to the application of an external strain [1, 2, 4]. The application of a subsequent external pressure with the concomitant particle rearrangements and compression results in a set of configurations characterized by the system volume $V = NV_g/\phi$ (ϕ is the volume fraction of N particles of volume V_g) and applied external stress or pressure p (for simplicity we assume isotropic states).

It has been long argued whether the jamming transition is a first-order transition at the discontinuity in the average coordination number, Z , or a second-order transition with the power-law scaling of the system’s pressure as the system approaches jamming with $\phi - \phi_c \rightarrow 0^+$ [5–7, 35]. Previous work [11, 36, 37] has proposed to explain the jamming transition by a field theory in the pressure ensemble. Here, we use the idea of “thermalization” of an ensemble of mechanically stable granular materials at a given volume and pressure to study the jamming transition from a thermodynamic viewpoint.

For a fixed number of grains, there exist many jammed states [25, 26] confined by the external pressure p in a volume V . In an effort to describe the nature of this nonequilibrium system from a statistical mechanics perspective, a statistical ensemble [8, 10, 11] was introduced for jammed matter. In the canonical ensemble of pressure and volume, the probability of a state is given by $\exp[-\mathcal{W}(\partial S/\partial V) - \Gamma(\partial S/\partial \Gamma)]$, where S is the entropy of the system, $\mathcal{W}$ is the volume function measuring the volume of the system as a function of the particle coordinates and $\Gamma \equiv pV$ is the boundary stress (or internal virial) [36] of the system. Just as $\partial E/\partial S = T$ is the temperature in equilibrium system, the temperature-like variables in jammed systems are the compactivity [3]

$$X = \partial V/\partial S, \quad (1)$$

and the angoricity [8],

$$A = \partial \Gamma/\partial S. \quad (2)$$

In a recent series of papers [29–33] the compactivity was used to describe frictional and frictionless hard spheres in the volume ensemble. Here, we test the validity of the statistical approach in the combined pressure-volume ensemble to describe deformable, frictionless particles, such as emulsion systems jammed under osmotic pressure near the jamming transition [38, 39].

In general, if the density of states $g(\Gamma, \phi)$ in the space of jammed configurations (defined as the probability of finding a jammed state at a given (Γ, ϕ) at $A = \infty$) is known, then calculations of macroscopic observables, like pressure p and average coordination number Z as a function of ϕ , can be performed by the canonical ensemble

average [36, 37] at a given volume:

$$\langle p(\alpha, \phi) \rangle_{\text{ens}} = \frac{1}{\mathcal{Z}} \int_0^\infty p g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma, \quad (3)$$

and

$$\langle Z(\alpha, \phi) \rangle_{\text{ens}} = \frac{1}{\mathcal{Z}} \int_0^\infty Z g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma, \quad (4)$$

where the canonical partition function is

$$\mathcal{Z} = \int_0^\infty g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma, \quad (5)$$

and the density of states is normalized as $\int_0^\infty g(\Gamma, \phi) d\Gamma = 1$. The inverse angoricity is defined as

$$\alpha \equiv 1/A = \partial S / \partial \Gamma. \quad (6)$$

At the jamming transition the system reaches isostatic equilibrium, such that the stresses are exactly balanced in the resulting configuration, and there exists a unique solution to the interparticle force equations satisfying mechanical equilibrium. It is well known that observables present power-law scaling [5–7]:

$$\langle p \rangle_{\text{dyn}} \sim (\phi - \phi_c)^a, \quad (7)$$

$$\langle Z \rangle_{\text{dyn}} - Z_c \sim (\phi - \phi_c)^b, \quad (8)$$

where $a = 3/2$ and $b = 1/2$ for Hertzian spheres and $Z_c = 6$ is the coordination number at the frictionless isostatic point (J-point) [40]. The average $\langle \dots \rangle_{\text{dyn}}$ indicates that these quantities are obtained by averaging over packings generated dynamically in either simulations or experiments as opposed to the ensemble average over configurations $\langle \dots \rangle_{\text{ens}}$ of Eqs. (3)–(4). Comparing the ensemble calculations, Eqs. (3)–(4), with the direct dynamical measurements, Eqs. (7)–(8), provides a basic test of the ergodic hypothesis for the statistical ensemble.

Our approach is the following: We first perform an exhaustive enumeration of configurations to calculate $g(\Gamma, \phi)$ and obtain $\langle p(\alpha, \phi) \rangle_{\text{ens}}$ as a function of α for a given ϕ using Eq. (3). Then, we obtain the angoricity by comparing the pressure in the ensemble average with the one obtained following the dynamical evolution with Molecular Dynamics (MD) simulations. By setting $\langle p(\alpha, \phi) \rangle_{\text{ens}} = \langle p \rangle_{\text{dyn}}$, we obtain the angoricity as a function of ϕ . By virtue of obtaining $\alpha(\phi)$, all the other observables can be calculated in the ensemble formulation. The ultimate test of ergodicity is realized by comparing the remaining ensemble observables with the corresponding direct dynamical measures.

II. POTENTIAL ENERGY LANDSCAPE APPROACH: ENSEMBLE CALCULATIONS

A. Features of the Potential Energy Landscape

An appealing approach for understanding out-of-equilibrium systems is to study the properties of the

system’s “potential energy landscape” (PEL) [41], described by the $3N$ -coordinates of all particles in the multi-dimensional configuration space, or landscape, of the potential energy of the system (N is the number of particles). Characterizing such potential energy landscapes has become an important approach to study the behaviour of out-of-equilibrium systems. For example, this approach has provided important new insights into the origin of the unusual properties of supercooled liquids, such as the distinction between “strong” and “fragile” liquids [42].

In frictionless granular matter, the potential energy is well-defined and each jammed configuration corresponds to one local minimum in the PEL. For small systems ($N \lesssim 14$), it is possible to find all the minima with current computational power [25]. For somewhat larger systems $N \approx 30$, it is possible to obtain a representative ensemble, without exhaustively sampling all the states. Based on these stationary points, we test the combined volume-stress ensemble. The following work is only valid for frictionless systems where the potential energy of interaction is well defined. Frictional grains are path dependent due to Coulomb friction between particles and therefore not amenable to a PEL study since there is no well defined energy of interaction.

The formalism introduced by Goldstein [43] consists of partitioning the potential energy surface into a set of basins as illustrated in Fig. 1. The dynamics on the potential energy surface can be separated into two types: the vibrational motion inside each basin and the transitional motion between the local minima. Stillinger and coworkers [44] developed the method of inherent structure to characterize the PEL. In this method, a local minimum in the PEL is located by following the steepest-descent pathway from any point surrounding the minimum. The inherent structure formalism simplifies the energy landscape into local minima and ignores the vibrational motion around them. The dynamics between the inherent structures is introduced with the transition states identified with the saddle points in the PEL. The transition states are stationary points like the local minima but they have at least one maximum eigendirection.

B. Finding Stationary States

For the simplest system of N structureless frictionless particles possessing no internal orientational and vibrational degrees of freedom, the potential energy function of this N -body system is $E(r_1, \dots, r_N)$, where the vectors r_i comprise position coordinates. As mentioned above, the most interesting points of a potential energy surface are the stationary points, where the gradient vanishes. Here we explain how to locate these stationary points. The algorithm follows well established methods in computational chemistry [41]. The procedure is analogous to finding the inherent structures [45] of glassy systems. The algorithm employed, LBFGS algorithm, is also sim-

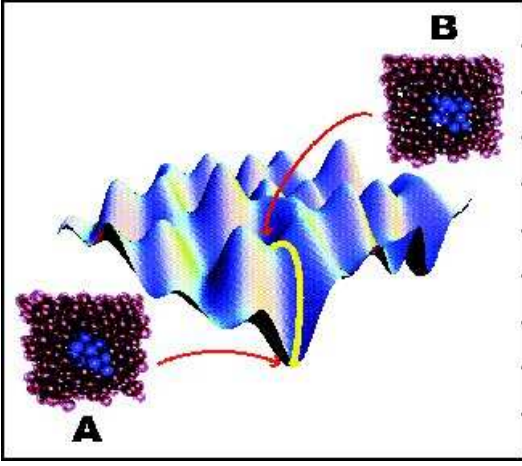


FIG. 1: A model two-dimensional potential energy surface. The energy landscape is divided into basins of attraction, where the minima are the jammed states connected by pathways through saddle points. States A and B are typical packing configurations of 30 particles (in blue) with their periodic boundary systems.

ilar to the conjugate gradient method employed by O' Hern [5, 25, 26], differing in the fact that it does not require the calculation of the Hessian matrix at every time step. We make the source code in C++ available at <http://www.jamlab.org> and free to use together with all the packings generated in this study. The algorithm has been used in the short version of this article [34] and in a study of the PEL in Lennard-Jones glasses to reconstruct a network of stationary states and apply a percolation picture of the glass transition [46].

C. General Method – Newton-Raphson Method

Consider the Taylor expansion of the potential energy, E , around a general point in configuration space, r ,

$$E(r+h) = E(r) + g^T h + \frac{1}{2} h^T H h + O(h^3), \quad (9)$$

where g is the gradient, $g_i = \partial_i E$, H is the Hessian matrix, $H_{ij} = \partial_i \partial_j E$, and h is a small step vector that gives the displacement away from r .

By Eq. (9), the calculation of energy difference for a given step h from the initial point r is complicated. By selecting the eigenvectors of the Hessian matrix e_α as our local coordinates, we can simplify the Taylor expansion of Eq. (9) as:

$$\Delta E = E(r+h) - E(r) \approx \sum_{\alpha} (g_{\alpha} h_{\alpha} + \frac{\lambda_{\alpha}}{2} h_{\alpha}^2), \quad (10)$$

where $g = \sum_{\alpha} g_{\alpha} e_{\alpha}$, $h = \sum_{\alpha} h_{\alpha} e_{\alpha}$, $H e_{\alpha} = \lambda_{\alpha} e_{\alpha}$, and λ_{α} is the eigenvalue of the Hessian matrix for component α .

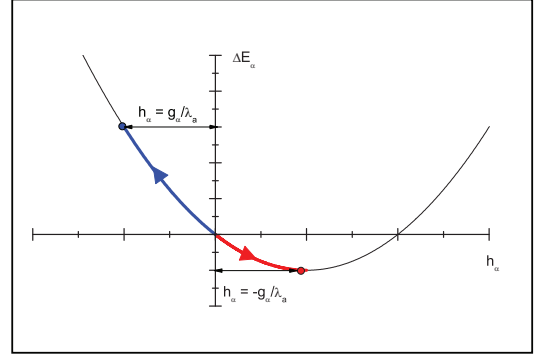


FIG. 2: A schematic energy change curve for one component with $\lambda_{\alpha} > 0$. We can select the downhill step as $h_{\alpha} = -\frac{g_{\alpha}}{\lambda_{\alpha}}$ to obtain a maximum energy change. The uphill step can not be too large since the Taylor expansion will not be accurate enough for the calculation. Here, the uphill step is chosen as $h_{\alpha} = \frac{g_{\alpha}}{\lambda_{\alpha}}$.

From Eq. (10), it is easy to see that the total change of energy could simply be the sum of the changes in each directions. This may help us to raise the energy in some directions and reduce the energy at others, and finally reach a stationary point. The length of each step components can be selected as the maximum change of energy:

$$h_{\alpha} = S_{\alpha} \frac{g_{\alpha}}{\lambda_{\alpha}}, \quad (11)$$

as shown in Fig.2. The sign $S_{\alpha} = \pm 1$ in this formula depends on the choice of uphill or downhill direction. In fact, for $\lambda_{\alpha} > 0$, it is possible to choose another step for the uphill case, since ΔE_{α} increases as $|h_{\alpha}|$, but for large steps, the Taylor expansion Eq. (9) may breakdown. Therefore, it is important to control the step length. For $\lambda_{\alpha} < 0$, we reach the opposite conclusion.

The stationary points can be separated into local minima and saddle points. Based on the eigenvalues of the Hessian matrix for the stationary point, the local minima are ordered as:

$$0 \leq \lambda_1 \leq \lambda_2 \leq \dots \leq \lambda_{3N}, \quad (12)$$

and for a saddle point of order α :

$$\lambda_1 \leq \dots \leq \lambda_{\alpha} \leq 0 \leq \lambda_{\alpha+1} \leq \dots \leq \lambda_{3N}. \quad (13)$$

Generally, this algorithm searches for the nearest stationary point on the surface by following the opposite ($\lambda_{\alpha} \geq 0$) and along ($\lambda_{\alpha} \leq 0$) the various gradient directions.

D. Finding local minima – LBFGS algorithm

It is much easier to locate local minima than saddle points because, for the first, we only need to search downhill in every direction. At present one of the most efficient methods to search the local minima for large system

is Nocedal's limited memory Broyden-Fletcher-Goldfarb-Shanno algorithm (LBFGS) [45, 47]. The LBFGS algorithm constructs an approximate inverse Hessian matrix from the gradients (first derivatives) which are calculated from previous points. Since it is only necessary to calculate the gradients at each searching step, the LBFGS algorithm increases the computational speed of the algorithm enormously.

In the Newton-Raphson method discussed above, the Hessian matrix of second derivatives is needed to be evaluated directly. Instead, the Hessian matrix used in LBFGS method is approximated using updates specified by gradient evaluations. The LBFGS algorithm code can be obtained from <http://www.netlib.org/opt/index.html>. Here we present a brief explanation of the algorithm

From an initial random point r_0 and an approximate Hessian matrix H_0 (in practice, H_0 can be initialized with $H_0 = I$), the following steps are repeated until r converges to the local minimum.

- Obtain a direction h_k by solving: $H_k h_k = -\nabla E(r_k)$.
- Perform a line search to find an acceptable step size γ_k in the direction found in the first step, then update $r_{k+1} = r_k + \gamma_k h_k$.
- Set $s_k = \alpha_k h_k$.
- Set $y_k = \nabla E(r_{k+1}) - \nabla E(r_k)$.
- Set the new Hessian, $H_{k+1} = H_k + \frac{y_k y_k^T}{y_k^T s_k} - \frac{H_k s_k (H_k s_k)^T}{s_k^T H_k s_k}$.

E. Finding saddles – Eigenvector following method

In the present study we do not make use of the saddle points. However, other studies using network theory to represent the PEL necessitate the links between minima through the saddle points [46]. For completeness, below we explain how to search for saddles. A particular powerful method for locating saddle points is the eigenvector following method [41].

The eigenvector-following method, developed by Cerjan, Miller and others [41, 48–52], consists of locating a saddle point from a local minimum. At each searching step towards a saddle point with α order, the directions are separated into two types: α uphill directions to maximization and $3N - \alpha$ downhill directions to minimization.

We follow the implementation of the eigenvector-following method by Grigera [49]. We give a general description: at each searching step, a step size h is calculated by the diagonalized Hessian matrix [49, 51, 52]:

$$h_\alpha = S_\alpha \frac{2g_\alpha}{|\lambda_\alpha| \left(1 + \sqrt{1 + 4g_\alpha^2/\lambda_\alpha^2}\right)}, \quad (14)$$

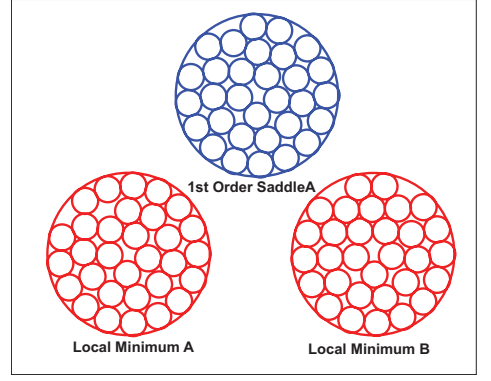


FIG. 3: A two dimensional 31 particle system in a circular boundary. Three different configurations in this system are generated with different algorithms. The LBFGS method is applied to locate minima A and B. For saddle C which connects A and B, the eigenvector following method is used.

where λ_α are the eigenvalues of the Hessian matrix and g_α are the components of the gradient in the diagonal base (h_α is set to 0 for the directions where $\lambda_\alpha = 0$). The sign $S_\alpha = \pm 1$ is chosen by the order of the saddle point. For a saddle point of order n , the algorithm will set $S_\alpha = -1$ for $1 \leq \alpha \leq n$ and $S_\alpha = 1$ for $\alpha > n$.

When $g_\alpha \rightarrow 0$, the step size h_α converges to the Newton-Raphson step as Eq. (11):

$$h_\alpha = S_\alpha \frac{g_\alpha}{\lambda_\alpha} + O(g_\alpha^2). \quad (15)$$

F. An Example

We generate a two dimensional soft-ball system in circular boundary, which contains 31 particles of equal radius, to illustrate the method of finding stationary and saddle points in the PEL. The interaction between particles (also the interaction between particles and wall) follows the Hertzian law [6]:

$$V(r_i, r_j) = \epsilon |r_i - r_j|^{\frac{5}{2}} \quad (16)$$

Here, ϵ is the interaction strength between particles i and j . The volume fraction is $\phi = 0.80$, which is closed to the jamming transition of $2d$ hard disks.

We first generate a random configuration, which is the initial point of the search of the minima. With the LBFGS method, we search the local minimum A nearby this initial point. After the minimum A is obtained, we apply the eigenvector following method to walk from the point A on the potential energy surface to locate the transition state C (here the transition state is a first order saddle). Finally, the minimum B is located by applying LBFGS method again. Figure 3 shows configurations of two local minima (marked as red) and the transition state (marked as blue) between them.

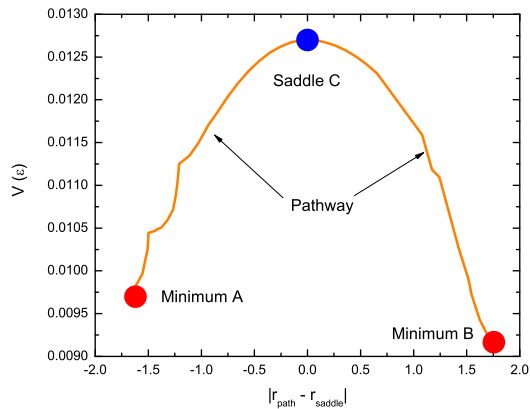


FIG. 4: The pathway from minimum A to minimum B, passing by the saddle C, the x-coordinate is the distance from saddle C, the y-coordinate is the potential energy of the packing.

The pathway from minimum A to minimum B, passing by transition state C, is shown in Fig. 4. The pathway distance is the Euclidean distance,

$$d = \sqrt{(r' - r)(r' - r)} = \sqrt{\sum_{i,\alpha} (r'_{i,\alpha} - r_{i,\alpha})^2}, \quad (17)$$

where $i = 1, 2, 3$, $\alpha = 1 \dots 3N$, r' is the coordinate of configuration passing along the searching method and r is the coordinate of saddle C .

The dynamics from minimum to minimum can be represented as a walk on a network whose nodes correspond to the minima and where edges link those minima which are directly connected by a transition state. The work of Doye [53] provides an illustration of such a landscape network for a LJ energy surface. To characterize the topology of the landscape network, Doye [53] study small Lennard-Jones clusters to locate nearly all the minima and transition states on the potential energy landscape. The inherent structure network of such a system has a scale-free and small-world properties. In a companion study [46] we repeated the main results as Doye studied. The numbers of minima and transition states are expected to increase roughly as $N_{min} \sim e^{\alpha N}$ and $N_{st} \sim N e^{\alpha N}$ respectively, where N is the number of atoms in the cluster. Therefore, the largest network that we are able to consider is for a 14-atom cluster for which we have located 4158 minima and 90 738 transition states in agreement with the results of Doye. In the next Section we apply the above formalism to find the stationary states for a 3d granular system of Hertz spheres in a periodic boundary.

III. SYSTEM INFORMATION. HERTZIAN SYSTEM OF SPHERES

Next we calculate the density of jammed states $g(\Gamma, \phi)$ in the framework of the PEL formulation for a system of

Hertz spheres. In the case of frictionless jammed systems, the mechanically stable configurations are defined as the local minima of the PEL [5, 26].

The systems used for both, ensemble generation and molecular dynamic simulation, are the same. They are composed of 30 spherical particles in a periodic boundary box. The particles have same radius $R = 5\mu m$ and interact via a Hertz normal repulsive force without friction. The normal force interaction is defined as [6, 35, 54]:

$$F_n = \frac{2}{3} k_n R^{1/2} (\delta r)^\delta, \quad (18)$$

where $\delta = 3/2$ is the Hertz exponent, $\delta r = (1/2)[2R - |\vec{x}_1 - \vec{x}_2|] > 0$ is the normal overlap between the spheres and $k_n = 4G/(1 - \nu)$ is defined in terms of the shear modulus G and the Poisson's ratio ν of the material from which the grains are made. We use typical values for glass: $G = 29$ GPa and $\nu = 0.2$ and the density of the particles, $\rho = 2 \times 10^3$ kg/m³ [6, 35]. The interparticle potential energy is

$$E = \frac{2}{3} \frac{k_n}{\delta + 1} R^{1/2} (\delta r)^{\delta+1}. \quad (19)$$

The Hertz potential is chosen for its general applicability to granular materials. The results are expected to be independent of the form of the potential. Below, we apply the LBFGS algorithms [45, 47] to find the local minima of the PES (zero-order saddles).

IV. ENSEMBLE GENERATION

In this section, we first explain the method to obtain geometrically distinct minima in the PEL to calculate the density of states. Then we show that the density of the states, $g(\Gamma, \phi)$, does not change significantly after sufficient searching time for the configurations.

In principle, if all local minima corresponding to the mechanically stable configurations of the PEL are obtained, the density of states $g(\Gamma, \phi)$ can be calculated. Such an exhaustive enumeration of all the jammed states requires that N not be too large due to computational limits. On the other hand, in order to obtain a precise average pressure in the MD simulation, $\langle p \rangle_{dyn}$, N cannot be too small such that boundary effects are minimized. Considering these constraints, we choose a 30 particle system.

In order to enumerate the jammed states at a given volume fraction ϕ , we start by generating initial unjammed packings (not mechanically stable) performing a Monte Carlo (MC) simulation at a high, fixed temperature. The MC part of the method applied to the initial packings assumes a flat exploration of the whole PEL. Every MC unjammed configuration is in the basin of attraction of a jammed state which is defined as a local minimum in the PES with a positive definite Hessian matrix, that is a zero-order saddle. In order to find such a minimum,

we apply the LBFGS algorithm provided by Nocedal and Liu [47] explained above. The PEL for each fixed ϕ likely includes millions of geometrically distinct minima by our simulation results. Therefore, an exhaustive search of configurations is computationally long; for a system of 30 particles it is impossible to find all the configurations with the current available computational power. However, we notice that it not crucial to find all the states, but rather a sufficiently accurate density of states. Therefore, we check that the number of found configurations has saturated after sufficient trials and that the density of states $g(\Gamma, \phi)$ has converged to a final shape under a prescribed approximation.

It is also important to determine if the local minima are distinct. Usually, the eigenvalues of the Hessian matrix at each local minimum can be used to distinguish these mechanically stable packings. Here, we follow this idea to compare minima to filter the symmetric packings. However, instead of calculating the eigenvalues of each packing, which is time consuming, we calculate a function of the distance between any two particles in the packing to improve search efficiency (for the LBFGS algorithm, we do not need to calculate the Hessian matrix). For each packing, we assign the function Q_i for each particle:

$$Q_i = \sum_{1 \leq j \leq N, j \neq i} \tan^2\left(\frac{\pi r_{ij}^2}{3L^2}\right), \quad (20)$$

where r_{ij} is the distance between particles i and j , L is the system size and $N = 30$. We list the Q_i for each packing from minimum to maximum $\{Q_i\} (1 \leq i \leq N)$. Since Q_i is a higher order nonlinear function, we can assume that two packings are the same if they have the same list. The tolerance is defined as:

$$T = \sqrt{\frac{\sum_{1 \leq i \leq N} (Q_i - Q'_i)^2}{N^2}}, \quad (21)$$

where Q_i and Q'_i are the corresponding values from the lists of two packings.

Figure 5 shows the distributions of the tolerance T for packings at different volume fractions. This figure suggests that two packings can be considered the same if $T \leq 10^{-1}$, which defines the noise level.

From Fig. 6, we see that after one week of searching, $g(\Gamma, \phi)$ does not change significantly, since the initial packings are generated by a completely random protocol. We also calculate the probability of finding new mechanically stable states for different searching days, defined as $N_{\text{new}}(i)/N_{\text{total}}(i)$, where $N_{\text{new}}(i)$ is the number of new configurations found on the i -th day and $N_{\text{total}}(i)$ is the total number of configurations found in i days. From Fig. 7, we see that, after one week searching, the probability of finding new configurations at different volume fractions seems to have converged in the linear plot. Figure 7b shows a detail of the actual number of new configurations found and $g(\Gamma, \phi)$ versus searching time in days suggesting convergence. However, the log-log plot of the inset in

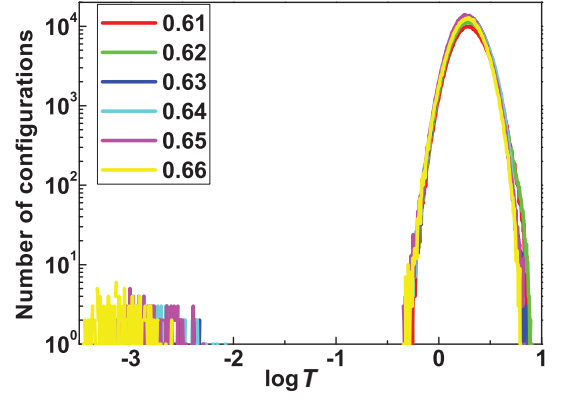


FIG. 5: The distribution of the tolerance T between any two packings at the given ϕ . From the graph, the value of T for which any two different packings are considered to be same is chosen to be 10^{-1} , which is above the noise threshold and below the distribution of T .

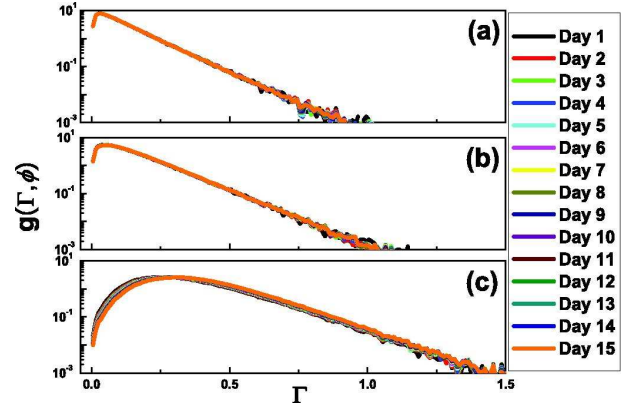


FIG. 6: Log-log plot of the distribution of $g(\Gamma, \phi)$ for 15 searching days (a) at $\phi = 0.609$, (b) at $\phi = 0.614$, (c) at $\phi = 0.625$. Different color in (a), (b), (c) corresponds to the different day. We find that after 15 days the distributions have converged.

Fig. 7a indicates that the algorithm is still searching for new configurations; the power-law relation in the inset suggesting a neverending story. However, the main question is whether the observables have converged. A further test of convergence is obtained below in Fig. 14 where the value of the inverse angoricity is measured as a function of the searching time in days. This plot suggests that enough ensemble packings have been obtained to capture the features of $g(\Gamma, \phi)$ that give rise to the correct observables. We conclude that we have obtained an accurate enough density of states for this particular system size. Regarding system size dependence, the presented results are still N dependent, although they started to converge for $N \sim 35$ and above, Fig. 8. More accurate calculations for large values of N remain computationally impossible, but in our treatment the exact choice of N is not as important as the consistency of the results between ensemble and MD, for a given N value.

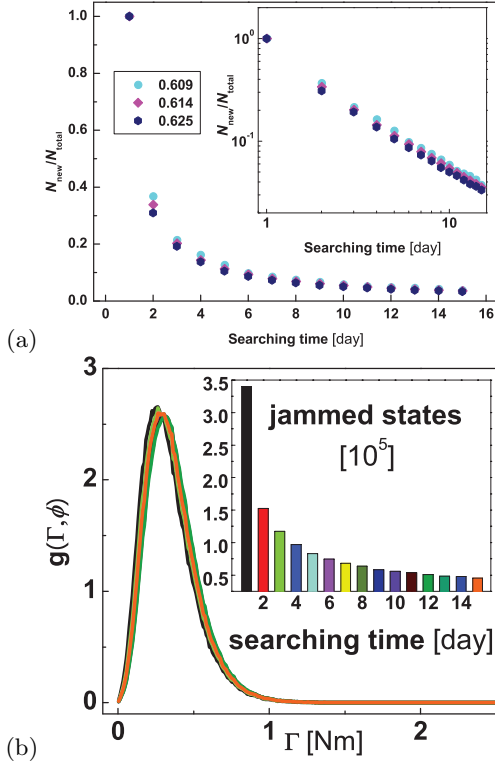


FIG. 7: (a) The probability to find new configurations as a function of searching time. (b) Linear plot of the density of states as a function of searching time. Different colors indicate different days according to the inset. Inset shows the actual number of new configurations.

Figure 9 shows $g(\Gamma, \phi)$ versus Γ for different volume fractions.

V. MD CALCULATIONS

In order to analyze numerical results, we perform MD simulations to obtain Z_{dyn} and ϕ_{dyn} , which are herein considered real dynamics. The algorithm is described in detail in [29, 35, 55]. Here, a general description is given: A gas of non-interacting particles at an initial volume fraction is generated in a periodically repeated cubic box. Then, an extremely slow isotropic compression is applied to the system. The compression rate is $\Gamma_0 = 5.9t_0^{-1}$, where the time is in units of $t_0 = R\sqrt{\rho/G}$. After obtaining a state for which the pressure p is a slightly higher than the prefixed pressure we choose, the compression is stopped and the system is allowed to relax to mechanical equilibrium following Newton's equations. Then the system is compressed and relaxed repeatedly until the system can be mechanically stable at the predetermined pressure. To obtain the static average of Z_{dyn} and ϕ_{dyn} , we repeat the simulation to get enough packing samples having statistically independent random initial particle positions. Here, 250 independent packings are obtained for each fixed pressure (see Fig. 10). $\phi = \langle \phi \rangle_{\text{dyn}}$ and

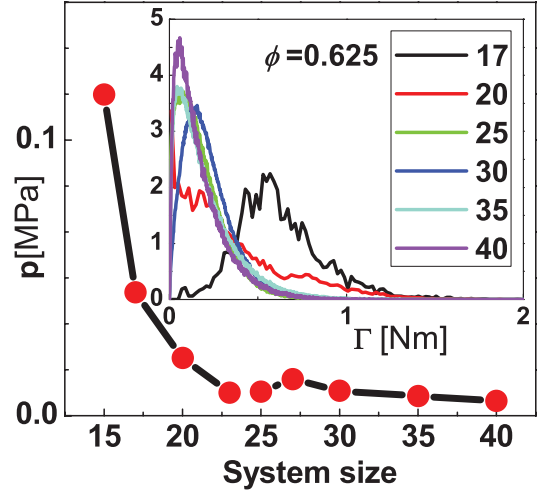


FIG. 8: Dependence of the results on the system size. The average value of p converges as early as $N \sim 25$ particles. The distribution $g(\Gamma, \phi)$ (inset) has not fully converged yet but its shape has converged after $N = 35$ and the first moment does not change as indicated by the average p .

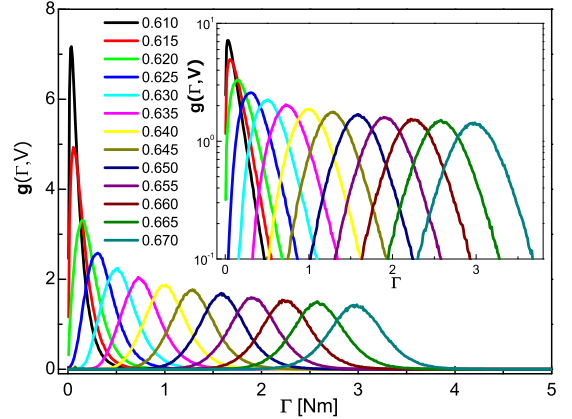


FIG. 9: The density of states $g(\Gamma, \phi)$ as a function of internal virial Γ for different volume fraction, ϕ , ranging from 0.610 to 0.670. The inset shows the logarithmic distribution of $g(\Gamma, \phi)$. At low volume fraction ($\phi \lesssim 0.625$), the distributions are sharp and the tails of the distributions are exponential. At high volume fraction ($\phi \gtrsim 0.640$), the distributions are much broader and the tails are Gaussian.

$\langle Z \rangle_{\text{dyn}}$ are flat averages of these 250 packings by

$$\langle \phi \rangle_{\text{dyn}} = \frac{\sum_{1 \leq i \leq 250} \phi_i}{250}, \quad (22)$$

and

$$\langle Z \rangle_{\text{dyn}} = \frac{\sum_{1 \leq i \leq 250} Z_i}{250}. \quad (23)$$

From previous studies, it has been observed the pressure p vanishes as power-law of ϕ when approaching the

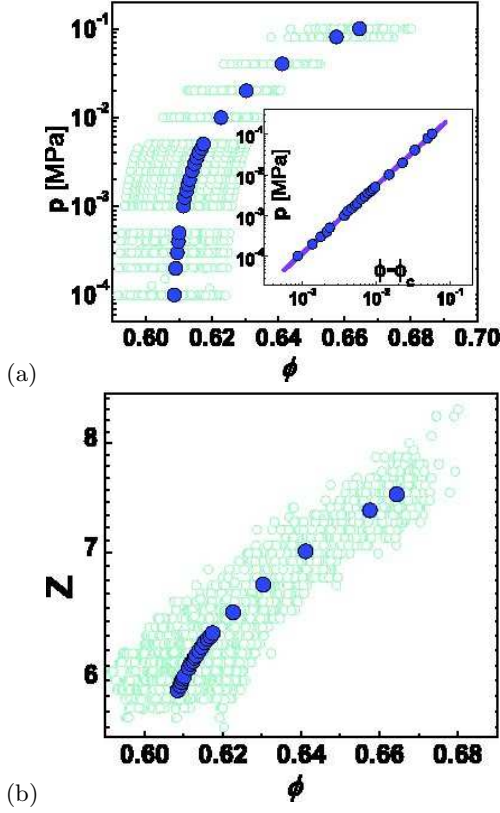


FIG. 10: The cyan $\circ$ is (a) ϕ_{dyn} and (b) Z_{dyn} for every single packing obtained with MD and the blue $\circ$ is $\langle\phi\rangle_{\text{dyn}}$ and $\langle Z\rangle_{\text{dyn}}$ average over the single packings for the system which are shown in the text of the paper.

jamming transition as seen in Eq. (7) [5, 6]. We obtain (Fig. 11)

$$\langle p \rangle_{\text{dyn}} = p_0 (\phi - \phi_c)^{1.65}, \quad (24)$$

where $\phi_c = 0.6077$ is the volume fraction corresponding to the isostatic point J [5, 6] following Eq. (8) and $p_0 = 10.8\text{MPa}$. This critical value ϕ_c and the exponent, $a = 1.65$, are slightly different from the values obtained for larger systems ($a = \delta$) [5, 6]. However, our purpose is to use the same system in the dynamical calculation and the exact enumeration for a proper comparison.

VI. ANGORITY CALCULATION

Since we obtain $g(\Gamma, \phi)$ and $\langle p \rangle_{\text{dyn}}$ for each volume fraction ϕ , we can calculate the inverse angoricity α by Eq. (3). The pressure $\langle p(\alpha, \phi) \rangle_{\text{ens}}$ for a given ϕ is a function depending on α as:

$$\langle p(\alpha, \phi) \rangle_{\text{ens}} = \frac{\int_0^\infty p g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma}{\int_0^\infty g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma} = \frac{\sum p e^{-\alpha \Gamma}}{\sum e^{-\alpha \Gamma}}. \quad (25)$$

Figure 12 shows the result of the numerical integration of Eq. (25) for a particular $\phi = 0.614$ as a function of

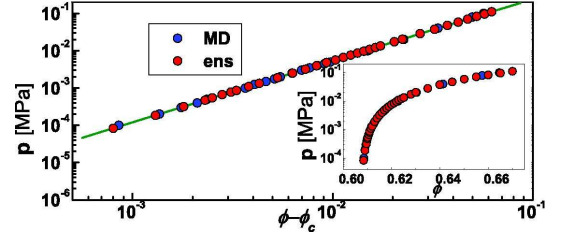


FIG. 11: Scaling of pressure. The blue $\circ$ shows the power-law relation for $\langle p \rangle_{\text{dyn}}$ vs $\langle \phi \rangle_{\text{dyn}} - \phi_c$ for the 30-particle system. Here, the pressure $\langle p \rangle_{\text{dyn}}$ are average values obtained by 250 independent MD simulations. The red $\circ$ is the pressure used to obtain the inverse angoricity α predicted by Eq. (24). The relatively small system size results in large fluctuations of the observables. In order to predict a precise relation for the system ($N = 30$), sufficient independent samples of the packings are generated to calculate the precise average for observables. We prepare 250 independent packings for each ϕ to get enough statistical samples to obtain $\langle p \rangle_{\text{dyn}}$ and $\langle Z \rangle_{\text{dyn}}$ by statistical average. The inset shows a semi-log plot.

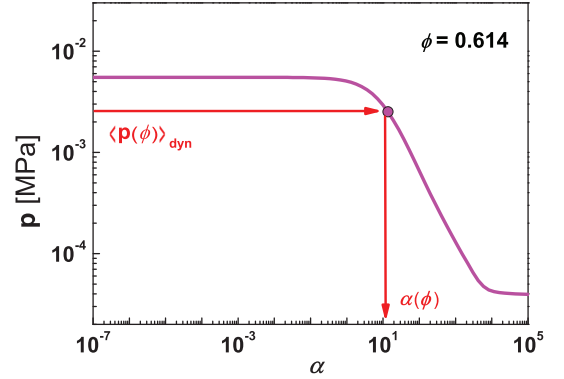


FIG. 12: The numerical integration of Eq. (25) for $\phi = 0.614$ is shown as the pink curve. We input the $\langle p \rangle_{\text{dyn}}$ (pink $\circ$ in the plot) and obtain the corresponding inverse angoricity α .

α using the numerically obtained $g(\Gamma, \phi)$ from Fig. 9. To obtain the value of α for this ϕ , we input the corresponding measure of the pressure obtained dynamically $\langle p(\phi) \rangle_{\text{dyn}}$ and obtain the value of α as schematically depicted in Fig. 12. The same procedure is followed for every ϕ (see Fig. 13) and the dependence $\alpha(\phi)$ is obtained.

We also check the inverse angoricity $\alpha(\phi)$ using $g(\Gamma, \phi)$ for different searching days. to ensure the accuracy and convergence to the proper value. From Fig. 14, we can see that, after 10 days searching, $\alpha(\phi)$ is stable due to the fact that the density of state, $g(\Gamma, \phi)$, does not change significantly.

For each ϕ we use $g(\Gamma, \phi)$ to calculate $\langle p(\alpha) \rangle_{\text{ens}}$ by Eq. (3). Then, we obtain $\alpha(\phi)$ by setting $\langle p(\alpha, \phi) \rangle_{\text{ens}} = \langle p \rangle_{\text{dyn}}$ for every ϕ . The resulting equation of state $\alpha(\phi)$ is plotted in Fig. 15 and shows that the angoricity follows a power-law, near ϕ_c , of the form:

$$A \propto (\phi - \phi_c)^\gamma, \quad (26)$$

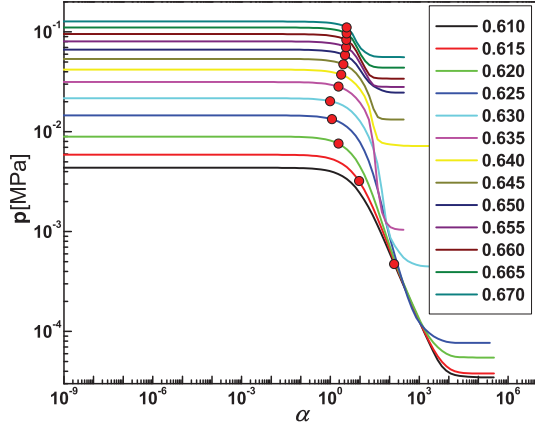


FIG. 13: Calculation of α for several volume fractions ϕ as explained in detail in Fig. 12

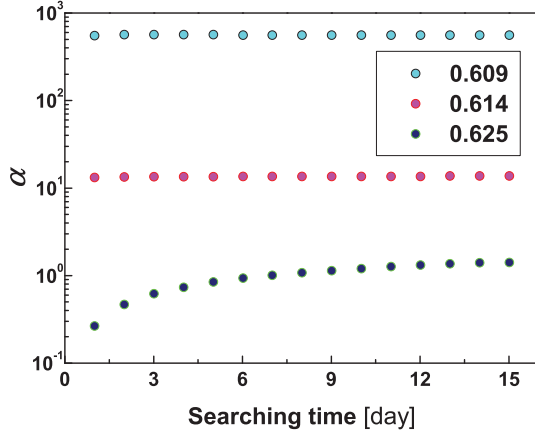


FIG. 14: Calculation of inverse angoricity α as a function of searching time.

with $\gamma = 2.5$. The result is consistent with $\gamma = \delta + 1.0$, suggesting that $A \propto \Gamma \propto F_n r$. For volume fraction much larger than ϕ_c , the system's input pressure $\langle p(\phi) \rangle_{\text{dyn}}$ reaches the plateau at low α of the function $\langle p(\alpha, \phi) \rangle_{\text{ens}}$ (see Fig. 13) and the corresponding $\alpha(\phi)$ becomes much smaller (the angoricity $A(\phi)$ becomes much larger), leading to large errors in the value of A as ϕ becomes large. This might explain the plateau found in A when $(\phi - \phi_c) > 2 \times 10^{-2}$ as shown in Fig. 15.

Angoricity is a measure of the number of ways the stress can be distributed in a given volume. Since the stresses have a unique solution for a given configuration at the isostatic point, ϕ_c , the corresponding angoricity vanishes. At higher pressure, the system is determined by multiple degrees of freedom satisfying mechanical equilibrium, leading to a higher stress temperature, A . The angoricity can also be viewed as a scale of stability for the system at different volume fractions. Systems jammed at larger volume fractions require higher angoricity (higher driving force) to rearrange.

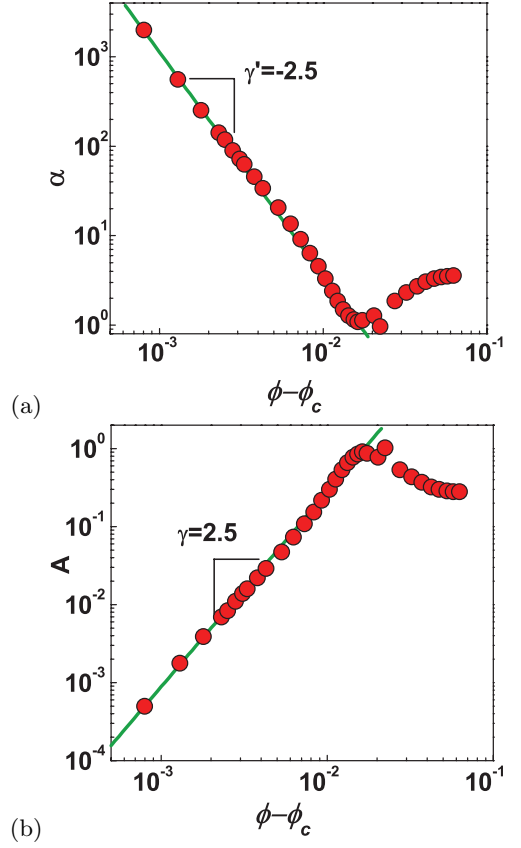


FIG. 15: (a) Inverse angoricity α as a function of $\phi - \phi_c$. We find a power-law relation for system's volume fraction ϕ near ϕ_c . The solid line has a slope of -2.5. (b) The angoricity $A (= 1/\alpha)$ vs $\phi - \phi_c$. To find A accurately for system's volume fraction ϕ much larger than ϕ_c , becomes difficult due to the large fluctuations and finite size effects. In principle, we expect that the plateau of A for large volume fraction ϕ might be related to the finite size of the sample. Indeed it is very difficult to estimate α since it falls in the plateau in Fig. 13.

VII. TEST OF ERGODICITY

In principle, using the inverse angoricity, α , from Eq. (26) we can calculate any macroscopic statistical observable $\langle B \rangle_{\text{ens}}$ at a given volume by performing the ensemble average [37]:

$$\langle B(\phi) \rangle_{\text{ens}} = \frac{1}{Z} \int_0^\infty B g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma. \quad (27)$$

We test the ergodic hypothesis in the Edwards's ensemble by comparing Eq. (27) with the corresponding value obtained with MD simulations averaged over (250) sample packings, B_i , generated dynamically:

$$\langle B(\phi) \rangle_{\text{dyn}} = \frac{1}{250} \sum_{i=1}^{250} B_i. \quad (28)$$

The comparison is realized by measuring the average coordination number, $\langle Z \rangle$, the average force and the distribution of interparticle forces. We calculate $\langle Z \rangle_{\text{ens}}$ by

Eq. (4) and $\langle Z \rangle_{\text{dyn}}$ as in Eq. (28). Using $\alpha(\phi)$ for each volume fraction, we calculate $\langle Z \rangle_{\text{ens}}$ by:

$$\langle Z(\phi) \rangle_{\text{ens}} = \frac{\int_0^\infty Z g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma}{\int_0^\infty g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma} = \frac{\sum Z e^{-\alpha\Gamma}}{\sum e^{-\alpha\Gamma}}. \quad (29)$$

The average force $\langle \bar{F} \rangle_{\text{ens}}$ is given by:

$$\langle \bar{F}(\phi) \rangle_{\text{ens}} = \frac{\int_0^\infty \bar{F} g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma}{\int_0^\infty g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma} = \frac{\sum \bar{F} e^{-\alpha\Gamma}}{\sum e^{-\alpha\Gamma}}, \quad (30)$$

where $\bar{F}$ is the average force for each ensemble packing. Finally, the force distribution $P_{\text{ens}}(F/\bar{F})$ is given by:

$$P_{\text{ens}}(F/\bar{F}) = \frac{\int_0^\infty P(F/\bar{F}) g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma}{\int_0^\infty g(\Gamma, \phi) e^{-\alpha\Gamma} d\Gamma} = \frac{\sum P(F/\bar{F}) e^{-\alpha\Gamma}}{\sum e^{-\alpha\Gamma}} \quad (31)$$

Equations (29)–(31) are then compared with the dynamical measures for a test of ergodicity in Figs. 16 and 17.

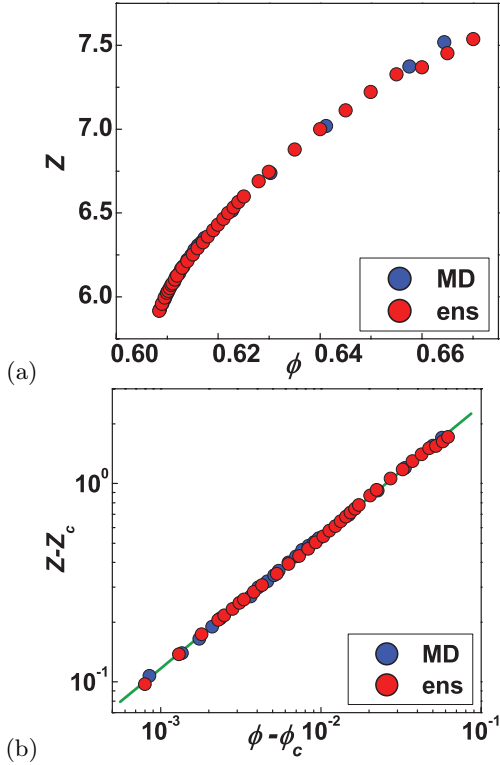


FIG. 16: **Test of ergodicity.** (a) The blue $\circ$ is the average coordination number $\langle Z \rangle_{\text{dyn}}$ obtained by 250 independent MD simulations. The red $\circ$ is the coordination number $\langle Z \rangle_{\text{ens}}$ calculated by the ensemble for different volume fractions. Agreement between both measures supports the concept of ergodicity in the system. (b) The same as (a) but in a log-log plot. The blue $\circ$ shows the power-law relations for $\langle Z \rangle_{\text{dyn}} - Z_c$ vs $\langle \phi \rangle_{\text{dyn}} - \phi_c$ for 30-particle system with $\phi_c = 0.6077$ and $Z_c = 5.82$.

Figure 16a and 16b show that the two independent estimations of the coordination number agree very well:

$\langle Z \rangle_{\text{ens}} = \langle Z \rangle_{\text{dyn}}$. The average inter-particle force $\bar{F}$ for a jammed packing is proportional to the pressure of the packing. We calculate $\langle \bar{F} \rangle_{\text{ens}}$ and $\langle \bar{F} \rangle_{\text{dyn}}$ and find that they coincide very closely (see Fig. 17a). The full distribution of inter-particle forces for jammed systems is also an important observable which has been extensively studied in previous works [5, 56, 57]. The force distribution is calculated in the ensemble $P_{\text{ens}}(F/\bar{F})$ by averaging the force distribution for every configuration in the PES. Figure 17b shows the distribution functions. The peak of the distribution shown in Fig. 17b indicates that the systems are jammed [5, 56, 57]. Besides the exact shape of the distribution, the similarity between the ensemble and the dynamical calculations shown in Fig. 17b is significant. The study of $\langle Z \rangle$, $\langle \bar{F} \rangle$ and $P(F/\bar{F})$ reveals that the statistical ensemble can predict the macroscopic observables obtained in MD. We conclude that the idea of “thermalization” at an angoricity is able to describe the jamming system very well.

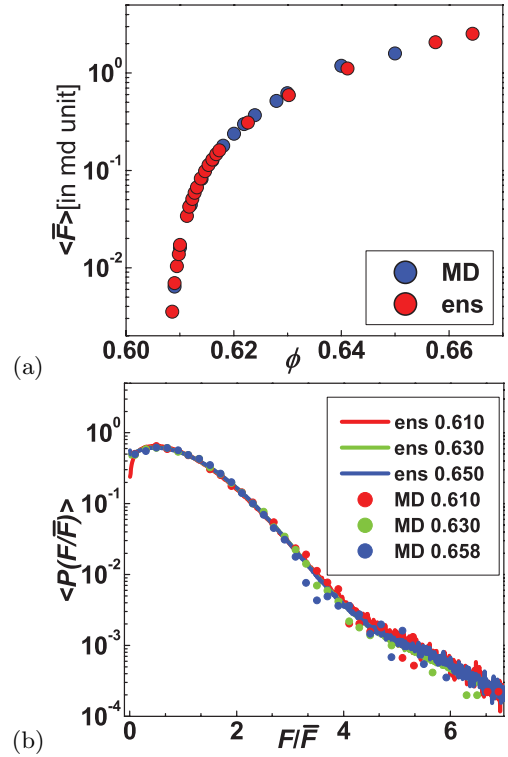


FIG. 17: **Test of ergodicity.** (a) Comparison of $\langle \bar{F} \rangle_{\text{dyn}}$ and $\langle \bar{F} \rangle_{\text{ens}}$ for different volume fractions. (b) The comparison of selected distribution of force $P_{\text{dyn}}(F/\bar{F})$ and $P_{\text{ens}}(F/\bar{F})$ for different volume fractions.

The MD simulations performed so far are at a predetermined pressure p . For this case there is no difference between the force distribution $P(F/\bar{F})$ and $P(F/\langle \bar{F} \rangle)$ [5]. On the other hand, a MD simulation at a given fixed volume fraction ϕ , gives rise to different distributions. For each system with fixed ϕ , the packings can have various pressure. This suggests that the force distribution for each packing scaled by the average force over all pack-

ings, $P(F/\langle\bar{F}\rangle)$, should be different from the force distribution scaled by the average force of that particular packing $P(F/\bar{F})$ [25]. We now proceed to investigate a constant volume MD, vMD simulation.

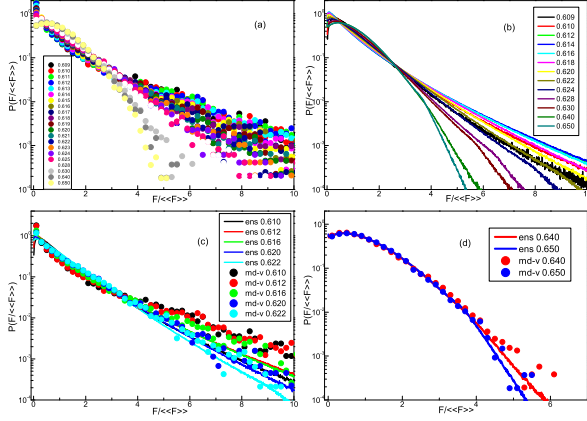


FIG. 18: (a) The distribution of force $P_{\text{vMD}}(F/\langle\bar{F}\rangle_{\text{vMD}})$. (b) The distribution of force $P_{\text{ens}}(F/\langle\bar{F}\rangle_{\text{ens}})$. (c) and (d) The comparison of selected $P(F/\langle\bar{F}\rangle)$ between vMD and ensemble predicted by angoricity.

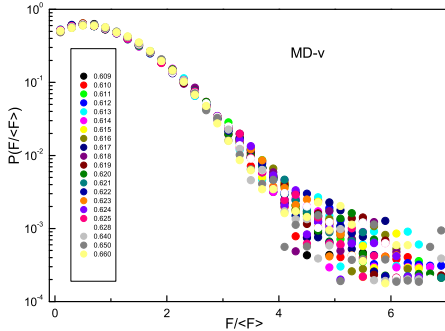


FIG. 19: The distribution of forces, $P(F/\langle F \rangle)_{\text{vMD}}$

The force distribution for vMD ensemble, $P_{\text{dyn}}(F/\langle\bar{F}\rangle_{\text{dyn}})$ is shown in Fig. 18a. From Fig. 18a, we find that the force distribution $P_{\text{dyn}}(F/\langle\bar{F}\rangle_{\text{dyn}})$ as a function of different volume fraction ϕ no longer collapse. At ϕ close to ϕ_c , the average system force $\bar{F}$ for each packing changes dramatically. While at ϕ is much above ϕ_c , the fluctuations of the average system force $\bar{F}$ decrease, then the force distribution $P_{\text{dyn}}(F/\langle\bar{F}\rangle_{\text{dyn}})$ changes continuously.

We can also calculate the force distribution

$P_{\text{ens}}(F/\langle\bar{F}\rangle_{\text{ens}})$ in the ensemble average:

$$P_{\text{ens}}(F/\langle\bar{F}\rangle_{\text{ens}}) = \frac{\int_0^\infty P(F/\langle\bar{F}\rangle_{\text{ens}})g(\Gamma, \phi)e^{-\alpha\Gamma}d\Gamma}{\int_0^\infty g(\Gamma, \phi)e^{-\alpha\Gamma}d\Gamma}, \quad (32)$$

where $\langle\bar{F}\rangle_{\text{ens}}$ is the overall average $\bar{F}$ of the ensemble.

From Fig. 18b, we find the same tendency as obtained in MD simulation. Furthermore, we check the distribution of force $P(F/\langle\bar{F}\rangle)$ for our vMD system (see Fig. 19). We see that $P(F/\langle\bar{F}\rangle)$ for different volume fraction ϕ collapses very well similarly to those obtained from the predetermined pressure system. This result suggests that $P(F/\langle\bar{F}\rangle)$ is a global quantity that can be used to verify if the system is jammed or not [25].

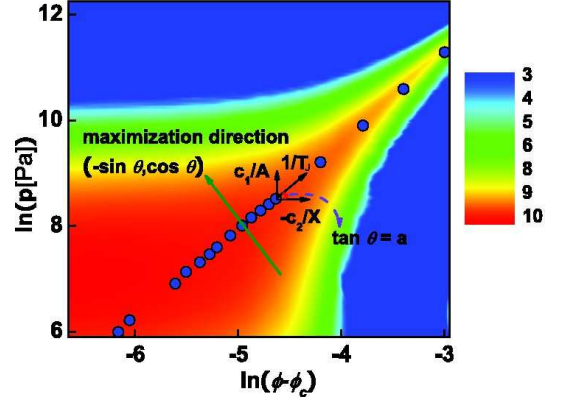


FIG. 20: Microcanonical calculations. The entropy surface $S(\ln(\phi - \phi_c), \ln p)$. The color bar indicates the value of the entropy. The superimposed blue $\bigcirc$ is $\langle p(\phi) \rangle_{\text{dyn}}$ from MD calculations as in Fig. 11. The olive arrow line indicates the maximization direction of the entropy $(-\sin \theta, \cos \theta)$. Following this direction, the entropy is maximum at the point $(\ln(\langle\phi\rangle_{\text{dyn}} - \phi_c), \ln\langle p \rangle_{\text{dyn}})$, corroborating the maximum entropy principle.

VIII. THERMODYNAMIC ANALYSIS OF THE JAMMING TRANSITION

So far we have considered how the angoricity determines the pressure fluctuations in a jammed packing at a fixed ϕ . The role of the compactivity in the jamming transition can be analyzed in terms of the entropy which is easily calculated in the microcanonical ensemble from the density of states. Figure 20 shows the entropy of the system as a function of (p, ϕ) in phase space:

$$S = \ln(\Omega(p, \phi)). \quad (33)$$

Here Ω is the number of states which is the unnormalized version of $g(\Gamma, \phi)$. It is important to note that Fig. 20 shows the non-equilibrium entropy, in the Edwards sense. At the Edwards equilibrium, the entropy is maximum respect to changes in ϕ and Γ . We will now see how the jammed system verifies the principle of maximum entropy.

We analyze the entropy surface $S(\ln(\phi - \phi_c), \ln p)$ plotted versus $(\ln(\phi - \phi_c), \ln p)$ in Fig. 20. When we plot superimposed the MD-obtained curve $\langle p(\phi) \rangle_{\text{dyn}}$ we see that the MD values pass along the maximum of the entropy surface constrained by the coupling between p and ϕ , Eq. (8) (such a curve is superimposed to the entropy surface in Fig. 20). Due to the coupling through the contact force law, the maximization of entropy is not on p or ϕ alone but on a combination of both. The entropy S reaches a maximum at the point $S(\ln(\langle \phi \rangle_{\text{dyn}} - \phi_c), \ln \langle p \rangle_{\text{dyn}})$ when we move along the direction perpendicular to the jamming curve $\langle p(\phi) \rangle_{\text{dyn}}$ (see the maximization direction in Fig. 20). This is a direct verification of the second-law of thermodynamics: the dynamical measures maximize the entropy of the system.

We can use this result to obtain a relation between angoricity and compactivity and show how a new “jamming temperature” T_J and the corresponding jamming “heat” capacity C_J can describe the jamming transition.

From the power-law relation $p = \Gamma/V \propto (\phi - \phi_c)^a$, we have:

$$\ln p = \ln p_0 + a \ln(\phi - \phi_c), \quad (34)$$

where p_0 is the constant depending on the system.

Figure 20 indicates that the jammed system always remain at the positions of maximal entropy,

$$\delta S = 0, \quad (35)$$

in the direction $(-\sin \theta, \cos \theta)$, perpendicular to the jamming power-law curve and the slope

$$\tan \theta = a. \quad (36)$$

In order to further analyze this result, we plot the entropy distribution along the direction $(-\sin \theta, \cos \theta)$ in Fig. 21. We see that the entropy of the corresponding jammed states remains at the peak of the distributions along $(-\sin \theta, \cos \theta)$. This is clear when we plot the value of (p, ϕ) from MD simulations in the plot of S in Fig. 21, blue dot. Except for volumes very close to jamming, the MD coincides with the maximum of S when taken along $(-\sin \theta, \cos \theta)$. We notice that the maximization is quite accurate for large volume fractions. For ϕ close to jamming deviations are seen. We cannot rule out that these deviations are finite size effects. The deviations for small ϕ (Fig. 21) remains to be studied. They could be due to finite size effects or due to the fact that the value of ϕ_c is different for the MD results and the microcanonical ensemble S due to the small size of the system. In general, this plot verifies the maximum entropy principle in this particular direction. An analogous plot where the entropy is shown as a function of ϕ but along the horizontal direction (or along the vertical direction, Γ) shows that the MD entropy is not maximal along these two directions.

Thus, the maximization of entropy is not on Γ or V alone, but on a combination of both. This means that the

entropy $S(\ln(\langle \phi \rangle_{\text{dyn}} - \phi_c), \ln \langle p \rangle_{\text{dyn}})$ is maximum along the direction of $(-\sin \theta, \cos \theta)$ and the slope for the entropy of the jamming power-law curve along this direction $(-\sin \theta, \cos \theta)$ is 0 (see Fig. 22), that is,

$$\frac{\partial S}{\partial \ln(\phi - \phi_c)} \sin \theta = \frac{\partial S}{\partial \ln p} \cos \theta. \quad (37)$$

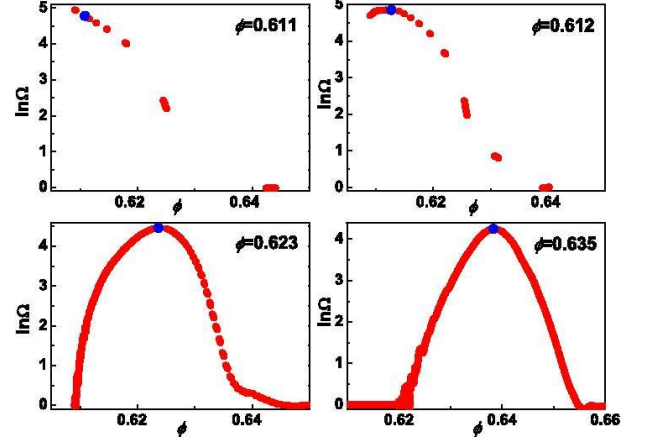


FIG. 21: The non-equilibrium entropy $S(\ln p, \ln(\phi - \phi_c))$ along the direction $(-\sin \theta, \cos \theta)$ for different jamming ensemble points. The blue $\circ$ represents the entropy of the jammed system obtained from MD. We see that closely follows the maximum of S for all the volume fractions except very close to the jamming point where the blue point does not coincide with the maximum of S . It remains to be studied if this deviation is a finite size effect, or it could be due to a different value of ϕ_c between simulations and microcanonical ensemble.

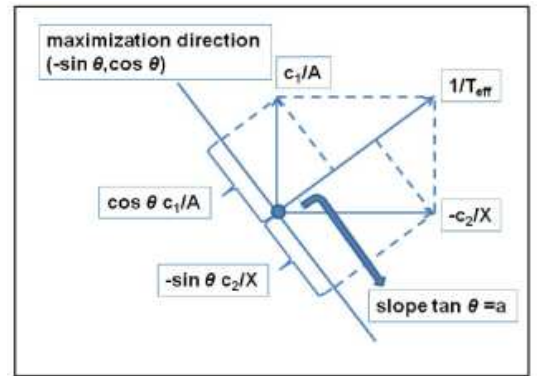


FIG. 22: The representation of the maximization analysis $\delta S = 0$ along the direction $(-\sin \theta, \cos \theta)$ for one point in the jamming power-law curve. Here $c_1 = \Gamma$ and $c_2 = (\phi - \phi_c)(NV_g/\phi^2)$.

By the definition of angoricity $A = \partial \Gamma / \partial S$ and compactivity $X = \partial V / \partial S$, we have:

$$\frac{\partial S}{\partial \ln p} = p \frac{\partial S}{\partial p} = \Gamma \frac{\partial S}{\partial \Gamma} = \frac{\Gamma}{A} = \frac{c_1}{A}, \quad (38)$$

$$\begin{aligned}\frac{\partial S}{\partial \ln(\phi - \phi_c)} &= (\phi - \phi_c) \frac{\partial S}{\partial \phi} = (\phi - \phi_c) \frac{\partial V}{\partial \phi} \frac{1}{X} = \\ &= -(\phi - \phi_c) \frac{NV_g}{\phi^2} \frac{1}{X} = -\frac{c_2}{X},\end{aligned}\quad (39)$$

where $\phi = NV_g/V$, $c_1 = \Gamma$ and $c_2 = (\phi - \phi_c)(NV_g/\phi^2)$.

By Eq. (38) and Eq. (39), we can simplify Eq. (37):

$$\frac{c_1}{A} + a \frac{c_2}{X} = 0. \quad (40)$$

The relation between X and A can be obtained then (Fig. 22):

$$X = -a \frac{c_2}{c_1} A = -a \frac{\phi - \phi_c}{p\phi} A. \quad (41)$$

From Eq. (41) we obtain that: $X \propto -(\phi - \phi_c)^{1+a-\gamma}/\phi$ and near ϕ_c :

$$X \sim -(\phi - \phi_c)^2. \quad (42)$$

We notice that the compactivity is negative near the jamming transition. A negative temperature is a general property of systems with bounded energy like spins [58]: the system attains the larger volume (or energy in spins) at ϕ_c when $X \rightarrow 0^-$ and not $X \rightarrow +\infty$ [The bounds $\phi_c \leq \phi \leq 1$ imply that the jamming point at $X \rightarrow 0^-$ is “hotter” than $X \rightarrow +\infty$. At the same time $A \rightarrow 0^+$ since the pressure vanishes].

We conclude that, A and X alone cannot play the role of temperature, but a combination of both determined by entropy maximization satisfying the coupling between stress and strain. Instead, there is an actual “jamming temperature” T_J that determines the direction $(-\sin\theta, \cos\theta)$ in the log-log plot of Fig. 20 along the jamming equation of state (see Fig. 22). By maximizing the entropy along this direction we obtain the “jamming temperature” T_J as a function of A and X :

$$\frac{1}{T_J} = \frac{c_1}{A} \sin\theta - \frac{c_2}{X} \cos\theta = \cos\theta \left(a \frac{c_1}{A} - \frac{c_2}{X} \right). \quad (43)$$

That is:

$$\begin{aligned}T_J &= \frac{A \sin\theta}{c_1} = -\frac{X \cos\theta}{c_2} = \frac{\sin\theta}{\Gamma} A = \\ &= \frac{a}{\sqrt{1+a^2}} \frac{A}{\Gamma} \sim (\phi - \phi_c)^{\gamma-a} \sim (\phi - \phi_c).\end{aligned}\quad (44)$$

Thus, the temperature vanishes at the jamming transition.

Furthermore, the “jamming energy” E_J , corresponding to the “jamming temperature” T_J in Eq. (43), has the

relation as below:

$$\begin{aligned}dE_J &= T_J dS \\ &= T_J \frac{\partial S}{\partial \ln(\phi - \phi_c)} d\ln(\phi - \phi_c) + T_J \frac{\partial S}{\partial \ln p} d\ln p \\ &= \left(-\frac{X \cos\theta}{c_2} \right) \left(-\frac{c_2}{X} \right) d\ln(\phi - \phi_c) + \frac{A \sin\theta}{c_1} \frac{c_1}{A} d\ln p \\ &= \cos\theta d\ln(\phi - \phi_c) + \sin\theta d\ln p \\ &= (\cos\theta + \sin\theta \tan\theta) d\ln(\phi - \phi_c) \\ &= \frac{d\ln(\phi - \phi_c)}{\cos\theta}.\end{aligned}\quad (45)$$

That is,

$$dE_J = \sqrt{a^2 + 1} d\ln(\phi - \phi_c), \quad (46)$$

and

$$E_J = (\sqrt{a^2 + 1}) \ln(\phi - \phi_c). \quad (47)$$

By the definition of “heat” capacity, we obtain two jamming capacities as the response to changes in A and X :

$$\begin{aligned}C_\Gamma &\equiv \partial\Gamma/\partial A \sim (\phi - \phi_c)^{-1} \sim A^{-2/5}, \\ C_V &\equiv \partial V/\partial X \sim (\phi - \phi_c)^{-1} \sim |X|^{-1/2}.\end{aligned}\quad (48)$$

The jamming capacity C_J can be obtained as:

$$C_J = T_J \frac{\partial S}{\partial T_J} = T_J \frac{\partial S}{\partial \ln p} \frac{\partial \ln p}{\partial T_J} + T_J \frac{\partial S}{\partial \ln(\phi - \phi_c)} \frac{\partial \ln(\phi - \phi_c)}{\partial T_J}. \quad (49)$$

Finally, with Eq. (37)–(39), the capacity C_J can be calculated:

$$C_J = T_J \left(\frac{c_1}{A} - \frac{c_2}{aX} \right) \frac{\partial \ln p}{\partial T_J} = T_J \frac{1+a^2}{a^2} \frac{c_1}{A} \frac{\partial \ln p}{\partial T_J}. \quad (50)$$

Since $T_J \sim (\phi - \phi_c)$ and $p \sim (\phi - \phi_c)^{1.5}$, we obtain

$$C_J \sim (\phi - \phi_c)^{-1}. \quad (51)$$

From Eq. (48), the jamming capacities diverge at the jamming transition as $A \rightarrow 0^+$ and $X \rightarrow 0^-$. However, this result does not imply that the transition is critical since from fluctuation theory of pressure and volume [58] we obtain:

$$\begin{aligned}\langle (\Delta\Gamma)^2 \rangle &= A^2 C_\Gamma \sim A^{1.6}, \\ \langle (\Delta V)^2 \rangle &= X^2 C_V \sim |X|^{1.5}.\end{aligned}\quad (52)$$

Thus, the pressure and volume fluctuations near the jamming transition do not diverge, but instead vanish when $A \rightarrow 0^+$ and $X \rightarrow 0^-$. From a thermodynamical point of view, the transition is not of second order due to the lack of critical fluctuations. As a consequence, no diverging static correlation length from a correlation function can be found at the jamming point. However, other correlation lengths of dynamic origin may still exist in the

response of the jammed system to perturbations, such as those imposed by a shear strain or in vibrating modes [7, 59]. Such a dynamic correlation length would not appear in a purely thermodynamic static treatment as developed here. We note that static anisotropic packings can be treated in the present formalism by allowing the inverse angoricity to be tensorial [37].

The intensive jamming temperature Eq. (44) gives use to a jamming effective energy E_J as the extensive variable satisfying $T_J = \partial E_J / \partial S$ and a full jamming capacity $C_J \sim (\phi - \phi_c)^{-1}$, which also diverges at jamming. However, the fluctuations of E_J defined as $\langle (\Delta E_J)^2 \rangle = T_J^2 C_J \sim T_J$ has the same behavior as the fluctuations of volume and pressure, vanishing at the jamming transition $T_J \rightarrow 0^+$ [$A \rightarrow 0^+$ in Eq. (44)].

IX. COMPARISON WITH O'HERN ET AL.

The results so far show a general agreement between MD and the ensemble average. These include the maximum entropy principle and ergodicity. We now turn to a comparison with similar simulations done by O'Hern *et al.* [25, 26]. These studies perform an exhaustive search of all configurations in the PEL of frictionless particles similarly as in the present paper. However, they find that the microstates are not equiprobable, i.e., microstates with the same pressure and volume fraction (pressure is fixed at zero since only hard sphere states are of interest) do not have the same probability when sampled by a given algorithm. Furthermore, experimental studies of equilibration between two systems [28], suggests that a hidden variable is necessary to describe the microstates, further supporting the results of [25]. The applicability of the microcanonical ensemble is based on the fact that the microstates are defined by (Γ, ϕ) . Thus, the fact that the states are not equiprobable implies that there must be an extra variable needed to describe their probabilities. Therefore, ergodicity and the maximum entropy principle, which are downstream from equiprobability, are not supposed to hold, in disagreement with the results shown in the present paper.

To investigate this situation, we repeat the same calculations as in [25] with our algorithms. We first rule out subtleties related to algorithmic dependent results in sampling the space of configurations. We use our 30 particles system and use $\phi = 0.61$ very close to jamming and $\Gamma = 0$ to look for the hard sphere packings. We search for the jammed configurations as above. We recall that the sampling of the space of configurations is not complete due to the relatively large system size but represent a good sampling as discussed above. Ref. [25] uses a different system of 14 particles in 2d for which 248,900 configurations are found exhaustively sampling the phase space (which is estimated to have $\sim 371,500$ states). These simulations correspond to a system with periodic boundary conditions for which a larger space is expected than the close boundary-system of Section II F.

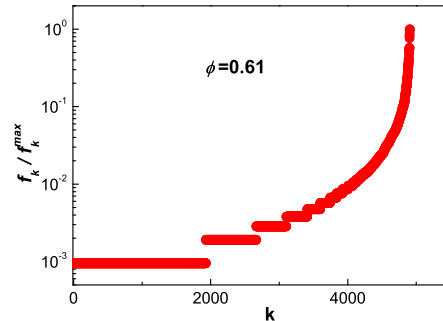


FIG. 23: Sampling probability of each microstate f_k identified by its rank k from low to high. Results are for a system of 30 particles at $\phi = 0.61$ and a narrow set of pressures around 0.

However, these differences do not affect the conclusions below.

We start by measuring f_k which is the probability to find a given microstate k as defined by [25]: each packing can be obtained many times during a search and therefore f_k measures the probability for which each packing occurs. The main result of [25] is that f_k differs by many orders of magnitude for states with fixed (Γ, ϕ) . Indeed, even configurations which are visually very similar can be 10^6 more frequent, see Fig. 1 of [25].

Figure 23 shows f_k sorted as a function of k , the rank, as in [25]. This plot reproduces the results of [25] in our system. For a fixed pressure and volume there are many states with a large difference in their probability. The least probable states are 10^{-3} less probable than the most probable state showing a breakdown of equiprobability. The question is how to interpret the results of ergodicity in the light of the failure of equiprobability and whether there is a need for an extra variable to describe the microstates.

We first mention the issue of the small system size. It is quite possible that the low probability states will completely disappear in the thermodynamic limit and the ones remaining are the most probable ones with equal probability. Indeed, the flat average assumption is only valid in the thermodynamic limit and simply says that even if there exists less probable states (10^{-3} less probable) then they will be irrelevant in the ensemble average, thus only the most probable and flat states are important.

We have done simulations with $N = 14$ particles and found that the least probable states are 10^{-5} less probable than the most probable states. Comparing with the factor 10^{-3} for $N = 30$, may indicate that the system size may take care of the non-equiprobability problem. However, calculations for larger system to fully test this assertion are out of the range of current and near future computational power.

Second, we notice that the coordination number is also

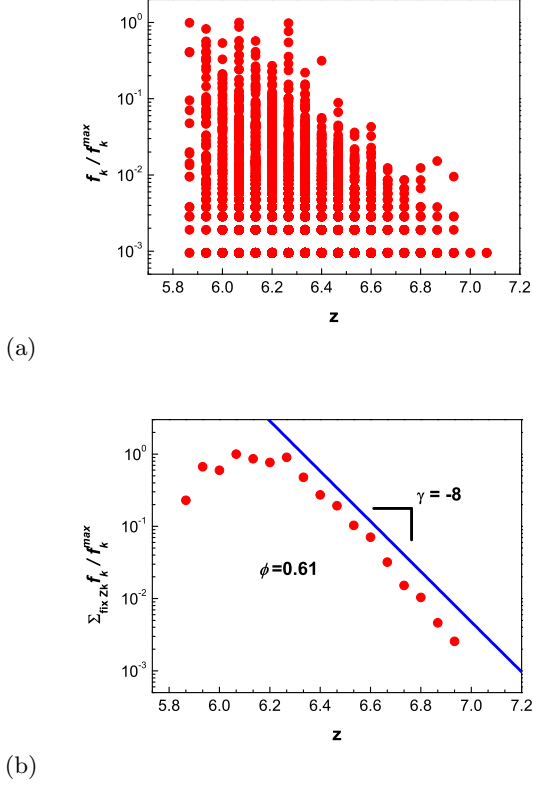


FIG. 24: (a) Sampling probability of each microstate f_k as a function of the coordination number Z_k of each microstate. (b) Plot of $\ln(\sum_{fix Z_k} f_k / f_k^{max})$ versus Z_k showing an exponential decay consistent with the density of states proposed in [29].

important to define the jammed states. Figure 24 plots the same states as Fig. 23 but as a function of Z_k , the coordination number of microstate k . The most probable states satisfy:

$$f_k(Z_k) \sim e^{-8Z_k}. \quad (53)$$

Furthermore, if we sum up all the states for a given Z_k and plot $\log(\sum_{fix Z_k} f_k)$ vs Z_k we obtain Eq. (53) as seen in Fig. 24b. This result does not mean that Z_k is the hidden variable but rather Eq. (53) provides the density of states proposed in [29] in the thermodynamics calculation of the random close packing of spheres. Indeed, we have predicted that the density of states $g(z) = h_z^z$, with h_z playing the role of a Planck constant defining the minimum size in the volume landscape. According to Eq. (53), this prediction is satisfied in average with $h_z = e^{-8}$ which is a small number as expected.

This result indicates that some variability in the probabilities of the microstates is expected from the fluctuations in the coordination number of each microstate. In Appendix A we elaborate an extension of the framework of [29] to incorporate fluctuations in Z that are neglected in [29]. The purpose is to test whether the RCP and jam-

ming transition are affected by these fluctuations. We find that the results are consistent with those found in [29].

We notice that for a fix Z_k there are still many marginal states with very small probabilities as seen in Fig. 24a. If these states do not completely disappear in the thermodynamic limit, then they need to be explained. We end this discussion by providing a possible explanation for the existence of these states.

The numerical breakdown of equiprobability might be related to the fact that the found packings are not indistinguishable. Indeed, we ignore the rotation and translation symmetries of the packing in order to make the numerical search possible. However, for the Edwards flat hypothesis, these packings should be assumed different. Once we breakdown the rotational symmetry, there would be many similar packings. The high degeneracy of the high symmetric packings may be responsible for the uneven distribution, which would be, in this case, simply artificial.

For instance, consider two packings with 4 particles: (a) a square packing with each particle on the corner and (b) a triangle with each particle in each corner plus one in the center.

For both packings there are $4! = 24$ different permutations, which should be considered as 24 different packings, in principle. However, since we can rotate the square packing by 90 degree and obtain the same one, there are only $24/4 = 6$ distinguishable packings. Similarly, for the triangle, there are $24/3 = 8$ distinguishable packings. The probability between (a) and (b) is uneven (6:8) if we assume that each distinguishable packing is equal-probable. Therefore, different symmetries of the packings may contribute to the unequal probabilities that we measure in the algorithms.

Therefore, if the Edwards assumption is correct, f_k should be proportional to S_k , where S_k is the order of the symmetry group (point group) of the packing k , since there are S_k degenerations (same packing if particles are identical). This conjecture needs extra evaluation of the symmetry of each packing. For instance, the translation invariance is important, and for cubic periodic boundary, it is also important to include the symmetry of cubic point group C_{3h} .

We do not investigate this conjecture but rather provide the codes and packings in <http://jamlab.org> to do that. Since the 3d case is complicated, one might try the 2d system first to easily visualize different packings. A simple question is: given two packings with different frequencies, how do they look like [25]? Would be the high symmetric one visited more, or inversely?

X. CONCLUSION

We have demonstrated that the concept of “thermalization” at a compactivity and angoricity in jammed systems is reasonable by the direct test of ergodicity. The

numerical results indicate that the full canonical ensemble of pressure and volume describes the observables near the jamming transition quite well. From a static thermodynamic viewpoint, the jamming phase transition does not present critical fluctuations characteristic of second-order transitions since the fluctuations of several observables vanish approaching jamming. The lack of critical fluctuations is respect to the angoricity and compactivity in the jammed phase $\phi \rightarrow \phi_c^+$, which does not preclude the existence of critical fluctuations when accounting for the full range of fluctuations in the liquid to jammed transition below ϕ_c . Thus, a critical diverging length scale might still appear as $\phi \rightarrow \phi_c^-$ [60], which has been recently observed by experiment [61].

In conclusion, our results suggest an ensemble treatment of the jamming transition. One possible analytical route to use this formalism would be to incorporate the coupling between volume and coordination number at the particle level found in [29, 62] together with similar dependence for the stress to solve the partition function. This treatment would allow analytical solutions for the observables with the goal of characterizing the scaling laws near the jamming transition.

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Appendix A: Microstates and Fluctuations in coordination number

Here, we develop a Z -ensemble for hard spheres in the limit of zero angoricity. In the main test we found that fluctuations in Z may account for certain variability in the probability of microstates. Here we investigate whether this variability affect the existence of RCP and the jamming point. We develop a partition function in Edwards ensemble to study the dependence of RCP on this type of fluctuations.

The partition function is

$$\mathcal{Z} = \int \dots \int_{N z_{min} < \sum z_i < N z_{max}} \prod_i e^{-(z_i/z^* + \beta\kappa/z_i)} dz_i, \quad (\text{A1})$$

where $z_{min} = Z$ and $z_{max} = 6$, $\beta = 1/X$, and $\kappa = 2\sqrt{3}$. We follow the notation and concepts from [29, 62–64]. We define $x = (\sum_i z_i)/N$, thus:

$$\mathcal{Z} = \int_{z_{min}}^{z_{max}} P(x) dx, \quad (\text{A2})$$

where

$$P(x) \equiv \int_0^\infty \dots \int_0^\infty \prod_i e^{-(z_i/z^* + \beta\kappa/z_i)} \delta\left(x - \frac{1}{N} \sum_i z_i\right) dz_i, \quad (\text{A3})$$

where $z^* = 1/8$ according to Fig. 24b. We consider the inverse Fourier transform of $P_x(f)$:

$$\begin{aligned} \mathcal{F}_f^{-1}[P_x(f)] &\equiv \int_{-\infty}^\infty e^{2\pi i f X} P(x) dx = \int_0^\infty \dots \int_0^\infty \prod_i e^{-(z_i/z^* + \beta\kappa/z_i)} e^{2\pi i f \sum z_i/N} dz_i = \\ &= \left[\int_0^\infty e^{-(z/z^* + \beta\kappa/z)} e^{2\pi i f z/N} dz \right]^N = \left\{ \int_0^\infty \left[1 + \left(\frac{2\pi i f z}{N} \right) + \frac{1}{2} \left(\frac{2\pi i f z}{N} \right)^2 + \dots \right] e^{-(z/z^* + \beta\kappa/z)} dz \right\}^N. \end{aligned} \quad (\text{A4})$$

Since

$$\int_0^\infty x^n e^{-\frac{a}{2}(x+1/x)} dx = 2K_n(a), \quad (\text{A5})$$

where $K_n(a)$ is the modified Bessel function of the second kind. By taking the coupling constant

$$\begin{aligned} B &\equiv \beta\kappa/z^*, \\ a &\equiv 2B^{1/2}, \\ z &= B^{1/2} z^* x. \end{aligned} \quad (\text{A6})$$

Then:

$$\int_0^\infty z^n e^{-(z/z^* + \beta\kappa/z)} dz = 2z^{*n+1} B^{(n+1)/2} K_n(2B^{1/2}). \quad (\text{A7})$$

Thus,

$$\begin{aligned} \mathcal{F}_f^{-1}[P_X(f)] &= (2z^*)^N \left[B^{1/2} K_0(2B^{1/2}) + \left(\frac{2\pi i f z^*}{N} \right) B K_1(2B^{1/2}) + \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \right)^2 B^{3/2} K_2(2B^{1/2}) + O(N^{-3}) \right]^N \\ &= (2z^*)^N \exp \left\{ N \ln \left[B^{1/2} K_0(2B^{1/2}) + \left(\frac{2\pi i f z^*}{N} \right) B K_1(2B^{1/2}) + \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \right)^2 B^{3/2} K_2(2B^{1/2}) + O(N^{-3}) \right] \right\} \\ &= (2z^* B^{1/2} K_0(2B^{1/2}))^N \exp \left\{ N \ln \left[1 + \left(\frac{2\pi i f z^*}{N} \right) \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} B^{1/2} + \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \right)^2 \frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} B + O(N^{-3}) \right] \right\} \end{aligned} \quad (\text{A8})$$

Now, we expand

$$\ln(1+x) = x - \frac{1}{2}x^2 + \frac{1}{3}x^3 + \dots \quad (\text{A9})$$

and

$$\begin{aligned}
& \exp \left\{ N \ln \left[1 + \left(\frac{2\pi i f z^*}{N} \right) \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} B^{1/2} + \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \right)^2 \frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} B + O(N^{-3}) \right] \right\} \\
&= \exp \left\{ N \left[\left(\frac{2\pi i f z^*}{N} \right) \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} B^{1/2} + \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \right)^2 \frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} B - \frac{1}{2} \left(\frac{2\pi i f z^*}{N} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} \right)^2 B + O(N^{-3}) \right] \right\} \\
&\approx \exp \left[2\pi i f \left(z^* B^{1/2} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} \right) - \frac{(2\pi f)^2}{2N} z^{*2} B \left(\frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} - \frac{K_1(2B^{1/2})^2}{K_0(2B^{1/2})^2} \right) \right],
\end{aligned} \tag{A10}$$

is just a Gaussian distribution with the mean

$$\mu = z^* B^{1/2} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})}, \tag{A11}$$

and the mean square deviation

$$\sigma_N = \frac{\sigma}{\sqrt{N}}, \tag{A12}$$

where

$$\sigma^2 \equiv z^{*2} B \left(\frac{K_2(2B^{1/2})}{K_0(2B^{1/2})} - \frac{K_1(2B^{1/2})^2}{K_0(2B^{1/2})^2} \right). \tag{A13}$$

Thus, by using the saddle point approximation, we obtain the free energy density f :

$$\begin{aligned}
\beta f \equiv & - \lim_{N \rightarrow \infty} \frac{\ln(\mathcal{Z})}{N} = \\
& - \ln(B^{1/2} K_0(2B^{1/2})) + \frac{1}{2\sigma^2} [(\mu - z_{max})^2 \Theta(\mu - z_{max}) + \\
& (z_{min} - \mu)^2 \Theta(z_{min} - \mu)].
\end{aligned} \tag{A14}$$

where $Z_{max} \equiv z_{max}/z^*$, $Z_{min} \equiv z_{min}/z^*$, and

$$L(B) \equiv B^{1/2} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})}, \tag{A18}$$

because

We also obtain the energy density, or volume density in the context of Edwards:

$$\begin{aligned}
\frac{z^*}{\kappa} w = \frac{d(\beta f)}{dB} = & -\frac{1}{2B} + B^{-1/2} \frac{K_1(2B^{1/2})}{K_0(2B^{1/2})} + \\
& \frac{1}{2} \frac{d}{dB} \left[\frac{(\mu - z_{max})^2}{\sigma^2} \right] \Theta(\mu - z_{max}) +
\end{aligned} \tag{A15}$$

$$\frac{dL(B)}{dB} = \frac{L(B)^2}{B} - 1. \tag{A19}$$

$$\text{Then, } \frac{1}{2} \frac{d}{dB} \left[\frac{(z_{min} - \mu)^2}{\sigma^2} \right] \Theta(z_{min} - \mu).$$

$$\frac{B}{2} \frac{d}{dB} \left[\frac{(\mu - z_{max})^2}{\sigma^2} \right] \Theta(\mu - z_{max}) = \left[\frac{1}{2} \left(\frac{L(B)(L(B) - Z_{max})}{B + L(B) - L(B)^2} \right)^2 - \frac{(B - Z_{max}L(B))(L(B) - Z_{max})}{B + L(B) - L(B)^2} \right] \Theta(L(B) - Z_{max}), \tag{A20}$$

and

$$\frac{B}{2} \frac{d}{dB} \left[\frac{(\mu - z_{min})^2}{\sigma^2} \right] \Theta(\mu - z_{min}) = \left[\frac{1}{2} \left(\frac{L(B)(L(B) - Z_{min})}{B + L(B) - L(B)^2} \right)^2 - \frac{(B - Z_{min}L(B))(L(B) - Z_{min})}{B + L(B) - L(B)^2} \right] \Theta(Z_{min} - L(B)). \tag{A21}$$

Thus,

$$\begin{aligned} \beta w = & -\frac{1}{2} + L(B) + \left[\frac{1}{2} \left(\frac{L(B)(L(B) - Z_{max})}{B + L(B) - L(B)^2} \right)^2 - \frac{(B - Z_{max}L(B))(L(B) - Z_{max})}{B + L(B) - L(B)^2} \right] \Theta(L(B) - Z_{max}) \\ & + \left[\frac{1}{2} \left(\frac{L(B)(L(B) - Z_{min})}{B + L(B) - L(B)^2} \right)^2 - \frac{(B - Z_{min}L(B))(L(B) - Z_{min})}{B + L(B) - L(B)^2} \right] \Theta(Z_{min} - L(B)), \end{aligned} \quad (\text{A22})$$

and the entropy density:

$$s = \beta(w - f). \quad (\text{A23})$$

There are two phase transitions at $L(B) = Z_{min}$ and $L(B) = Z_{max}$. For the jammed phase $Z_{min} < L(B) < Z_{max}$, we have $\beta w = L(B) - 1/2$. If z^* is a small value, $z^* = 1/8$ from Fig. 24, then B is relatively large. Thus, $L(B) \approx B^{1/2}$ and $w_{max} \approx L(B)/\beta = \kappa/z^*B^{-1/2} = \kappa/(z^*Z_{min}) = \kappa/z_{min}$. Similarly, $w_{min} \approx \kappa/z_{max}$, which is consistent with the boundaries of the phase diagram obtained in [29]. Furthermore, $f \approx 2B^{1/2}$ and $s \approx$

$s_0 - B^{1/2}$, where $s_0 = Z_{max}$. Or, $s = (z_{max} - \kappa/w)/z^*$. Thus, we have verified that the inclusion of fluctuations in the coordination number does not change the shape of the jamming phase diagram obtained in [29, 63]. These fluctuations may affect the probability of the microstates according to the density of states proposed in [29]. A further application of this generalized Z -ensemble is developed in [65] to calculate the probability of coordination numbers in packings, with good agreement with the numerical results for different packings in the phase diagram.

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HIGHLIGHT

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Fundamental challenges in packing problems: from spherical to non-spherical particles

Fundamental challenges in packing problems: from spherical to non-spherical particles

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Random packings of objects of a particular shape are ubiquitous in science and engineering. However, such jammed matter states have eluded any systematic theoretical treatment due to the strong positional and orientational correlations involved. In recent years progress on a fundamental description of jammed matter could be made by starting from a constant volume ensemble in the spirit of conventional statistical mechanics. Recent work has shown that this approach, first introduced by S. F. Edwards more than two decades ago, can be cast into a predictive framework to calculate the packing fractions of both spherical and non-spherical particles.

Introduction

In 1989 Edwards and Oakeshott made the remarkable proposal that the macroscopic properties of static granular matter can be calculated as ensemble averages over equiprobable jammed microstates controlled by the system volume.¹ In other words, granular matter is amenable to a statistical mechanical treatment, where the role of energy is played by the volume. Clearly, there is no *a priori* reason why such a treatment should be correct. Granular matter is profoundly out of equilibrium, since thermal fluctuations are essentially absent for the macroscopic

length scales considered. In particular, there is no equivalent of Liouville's theorem for equilibrium systems due to the strongly dissipative nature of granular assemblies, which are dominated by static frictional forces. Nevertheless, the Edwards' ensemble approach has proven exceedingly useful in characterizing the properties of these athermal states of matter and continues to intrigue both experimentalists and theoreticians alike.

The main statements of this approach are:^{1,2} (i) the distribution of jammed microstates is flat and independent of the compaction history leading to a natural definition of a configurational entropy $S = \ln \Omega_{\text{Edw}}$, where Ω_{Edw} is the number of jammed configurations. (ii) There is an equivalence between ensemble averages and time averages, if the system can explore its jammed configurations by some external drive (tapping or slow shearing). (iii) The compactness $X^{-1} = \partial S / \partial V$ characterizes the packing state analogous to the temperature in equilibrium systems. These strong assumptions

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have been scrutinized in various studies over recent years in order to obtain insight into the validity of Edwards' approach.³ Soft compaction experiments under continuous tapping have provided evidence for a reversible branch in the packing fraction for a variation of the tapping amplitude, indicating the existence of thermodynamic states.^{4–7} Simple models of such a compaction dynamics have confirmed ergodicity and have been connected to a slow relaxation dynamics akin to the relaxation in glasses.^{8–12} One signature of such a slow dynamics is the existence of a non-equilibrium fluctuation–dissipation relation.^{8,9,13} Indeed, the effective temperature appearing in FDRs under perturbations agrees with the configurational temperature $T_{\text{eff}}^{-1} = \partial S / \partial E$ in Edwards' framework.¹³

Ergodicity has also been demonstrated explicitly in more realistic simulations.^{14,15} The compactivity has been measured in simulations and experiments.^{16–20} On the other hand, results on the equiprobability of microstates are mixed. By evaluating the probabilities of jammed microstates in small clusters a break down of the flat distribution assumption has been demonstrated,^{21–24} which might be traced back to the packing protocol used.¹⁵ Recent studies have investigated the equilibration of granular subsystems in contact,^{15,25,26} providing further insight into the thermodynamic nature of granular matter.

Ultimately, the success of any statistical mechanical theory needs to be measured by the comparison with experiments. One key problem in Edwards' approach is to identify a suitable *volume function*, which parametrizes the total volume of the packing as a function of the particle configurations (positions and orientations), replacing the role of the Hamiltonian.² Here, different conventions can be employed to partition the total volume into cells associated with each particle,^{27,28} the simplest of which is the Voronoi tessellation.^{29,30} In 3D these exact volume functions are difficult to handle analytically, so that reduced representations are sought. The thermodynamic nature of packings suggests to use a coarse-grained description of the volume function in terms of observables such as the average number of contacts z (coordination number).^{31–34} In turn, z is determined by the force transmission in the contact network leading to a mechanically stable packing in which forces and torques on each particle balance.³⁵ For the force network ensemble approaches similar to the volume ensembles have been introduced in order to explain the observed force distribution³⁶ from an entropy maximization.^{37–41} The stress tensor has also been considered as a conserved quantity leading to a different class of ensembles.^{41,42}

Recently, the force transmission has been treated on a random graph under local mechanical stability constraints resulting in quantitative predictions for the force distribution and the value of z using a cavity method.⁴³ The problem of finding the densest random packing can be similarly formulated as a constraint optimization problem: random close packings appear as the ground state of the volume ensemble restricted to disordered packings as $X \rightarrow 0$ for a given z .^{31,32} This picture highlights that jamming falls into the class of NP optimization problems,⁴⁴ which can be tackled successfully with the methods of statistical mechanics such as cavity methods.⁴³ A full solution needs to combine the two approaches for the force

and volume ensembles, where the Hamiltonian that enforces jamming is a function of both the particle configurations and the contact forces on a random contact network. These ensemble approaches are thus similar in spirit to other recent works that consider jamming as the infinite pressure limit of metastable glass phases.^{44–51} Here, one considers instead of the Edwards entropy S , the “glass complexity” in order to obtain the statistics of the metastable basins as the pressure diverges. Treatments of this problem based on the random-first order transition picture and replica theory have been performed.⁵⁰

From spheres to non-spheres

Random packings of hard objects appear in a broad range of scientific and engineering fields like self-assembly of nanoparticles, liquid crystals, glass formation and bio-materials.⁵² In fact, the question of how densely objects of a particular shape can fill a given volume is probably one of the most ancient scientific problems and still of great practical importance for all industries involved in granular processing. The densest random packing has been extensively studied in experiments and simulations for spheres, which typically reach a maximal volume fraction of $\phi \approx 0.64$ in monodisperse assemblies.^{53,54} This value is quite robustly reproducible and commonly referred to as random close packing (RCP) density. However, much less is known about anisotropic shapes, despite the fact that all shapes in nature deviate from the ideal sphere. A theoretical investigation of the packing problem has proven notoriously difficult due to the strong positional and orientational correlations of dense packings. In fact, a mathematically rigorous treatment of random sphere packings has been the outstanding component in T.C. Hales' proof of Kepler's conjecture on the densest packing of spheres.⁵⁵

Recent empirical work has focused on packings of anisotropic shapes like ellipsoids, spherocylinders, and tetrahedra,

Table 1 Overview of maximal packing fractions found for disordered packings of a selection of regular shapes. We observe that spheres are the worst-packing objects among all shapes in 3d as conjectured by Ulam,⁸⁸ while tetrahedra achieve the densest disordered packing. We note that the tetrahedron is the only shape known that packs in a disordered arrangement denser than spheres in the FCC crystal ($\phi_{\text{FCC}} = 0.7405$). Ellipsoids and lens-shaped particles pack very close to this value

Shape	ϕ_{max} simulation	ϕ_{max} experiment	ϕ_{max} theory
Sphere	0.645 (ref. 54)	0.64 (ref. 53)	0.634 (ref. 31)
M&M candy		0.665 (ref. 58)	
Dimer	0.703 (ref. 91)		0.707 (ref. 34)
Oblate ellipsoid	0.707 (ref. 58)		
Prolate ellipsoid	0.716 (ref. 58)		
Spherocylinder	0.722 (ref. 68)		0.731 (ref. 34)
Lens-shaped particle			0.736 (ref. 34)
Octahedron	0.697 (ref. 67)		
Icosahedron	0.707 (ref. 67)		
Dodecahedron	0.716 (ref. 67)		
General ellipsoid	0.735 (ref. 58)	0.74 (ref. 59)	
Tetrahedron	0.7858 (ref. 63)	0.76 (ref. 64)	



which can achieve considerably denser volume fractions than the spherical RCP^{56–68} (see Table 1). In fact, a conjecture attributed to Ulam in the context of regular packings and recently also formulated for random packings⁶⁷ states that the sphere is, indeed, the worst packing object among all convex shapes. This suggests to improve packing fractions by searching in the space of object shapes, but in the absence of any general theory, this search could so far only be performed on a case-by-case basis using experiments and simulations. A caveat of such empirical studies is the strong protocol dependence of the final close packed state even for the same shape. While the range of achieved volume fractions is relatively small for spheres,⁵⁴ recent studies of spherocylinder packings, *e.g.*, exhibit a much greater variance depending on the algorithm used.^{56,57,60–62,65–68} Further theoretical insight is needed, which can be obtained by considering a coarse-grained distribution for the Voronoi volumes in the packing, as discussed next.

A mean-field theory for random close packings

In the Voronoi convention one associates with each particle the fraction of space that is closer to this particle than to any other one. This defines the Voronoi volume W_i of a particle i , which depends on the configurations of all remaining particles $\mathbf{x}_j = (\mathbf{r}_j, \hat{\mathbf{t}}_j)$, (including position $\mathbf{r}_j$ and orientation $\hat{\mathbf{t}}_j$). The total volume V occupied by N particles is $V = \sum_{i=1}^N W_i(\{\mathbf{x}_1, \dots, \mathbf{x}_N\})$, and the packing fraction of monodisperse particles of volume V_α is $\phi = NV_\alpha/V$. In order to determine W_i one has to know the Voronoi boundary (VB) between two particles i and j , which is the hypersurface that contains all points equidistant to the surfaces of both particles and thus depends on the particle shape and their relative configuration. The boundary of W_i then follows from a global minimization procedure over all pairwise VB.³² In order to take into account multi-particle correlations in the packing, we use a statistical treatment where the overall volume is expressed in terms of an average Voronoi volume: $V = N\bar{W}(z)$, so that $\phi = V_\alpha/\bar{W}(z)$. Instead of an exact description in terms of all configurations $\{\mathbf{x}_1, \dots, \mathbf{x}_N\}$, the average Voronoi volume is characterized by the coordination number z , which denotes the average number of contacting neighbours in the packing. We derive a self-consistent equation for the coarse-grained volume function $\bar{W}(z)$ of monodisperse particles:^{31,32,34}

$$\bar{W}(z) = \int d\mathbf{c} \exp \left\{ -\frac{V^*(\mathbf{c})}{\bar{W}(z) - V_\alpha} - \sigma(z)S^*(\mathbf{c}) \right\}. \quad (1)$$

Here, V_α is the volume of a single particle and $\sigma(z)$ is the average free-surface of particles at contact, which can be estimated from local configurations of z contacting particles. Formally, the integrand on the right hand side can be considered as the cumulative distribution function $P(\mathbf{c})$ containing the probability to find the boundary of the Voronoi volume in the direction $\hat{\mathbf{c}}$ at a value larger than c . This quantity can be interpreted geometrically as the probability to find all $N - 1$ particles outside a volume Ω centred at $\mathbf{c}$ from the

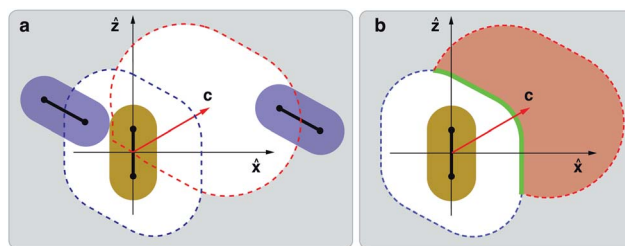


Fig. 1 The Voronoi excluded volume and surface for spherocylinders. (a) The volume Ω (red) is excluded for the remaining $N - 1$ particles in the packing because otherwise the Voronoi boundary would be found at a value smaller than c in the direction $\hat{\mathbf{c}}$. We draw the usual hard-core excluded volume $V_{\text{ex}}^{\text{70}}$ in blue. (b) The overlap of Ω and V_{ex} defines the Voronoi excluded volume V^* (red) and the Voronoi excluded surface S^* (green). Figure taken from ref. 34.

reference particle (see Fig. 1a). The particular form of $P(\mathbf{c})$ results from a factorization into bulk and contact terms, which are motivated from the dominant contributions in the radial distribution function.^{31,32,34,69}

The quantities V^* and S^* are the Voronoi excluded volume and surface, which extend the usual hard-core excluded volume of equilibrium systems $V_{\text{ex}}^{\text{70}}$ to packings. The volume V^* is the volume excluded by Ω for bulk particles and takes into account the overlap between Ω and V_{ex} : $V^* = \bar{\Omega} - \overline{\Omega \cap V_{\text{ex}}}$, where the bar denotes an orientational average. Likewise, S^* denotes the surface excluded by Ω for contacting particles: $S^* = \overline{\partial V_{\text{ex}} \cap \Omega}$. Plots of V^* and S^* for spherocylinders are shown in Fig. 1b. Analytical expressions for V^* and S^* can be derived in the spherical limit in closed form.^{31,32} For non-spherical shapes analytic expressions for the VB can be derived using a suitable decomposition of the shape into overlapping and/or intersecting spheres. This leads to exact expressions for V^* and S^* , which can be evaluated numerically.⁷¹ Interestingly, in the limit $\alpha \rightarrow 1$, eqn (1) admits an exact solution for spheres: $W(z) = 2\sqrt{3}V_1/z$. As a consequence, we obtain an equation of state for spherical packings^{31,32}

$$\phi(z) = \frac{z}{z + 2\sqrt{3}}, \quad (2)$$

which predicts the limiting values $\phi = 0.536$ and $\phi = 0.634$ under the isostatic conditions $z = 4$ and $z = 6$ for infinitely rough and frictionless spheres, respectively. Using the thermodynamic framework one can show that these two values are reached in the limits of infinite and zero compactivity, respectively.³¹ Therefore, the spherical equation of state leads to a statistical interpretation of RCP as the ground state of disordered sphere packings. The predictions for the limiting values are in good agreement with the values found in experiments and simulations for both random loose packings and RCP of spheres.

Under deformation from the sphere, higher packing fractions are typically reached, where the spherical limit appears as a singular point in the $\phi(\alpha)$ plane. Moreover, smooth shapes close to the sphere are not isostatic but hypostatic with $z < 2d_f$ due to redundancies in the force and torque balance equations.^{72,73} The variation $z(\alpha)$ is obtained by considering the average effective number of degrees of freedom $\tilde{d}_f$ defined as the



number of linearly independent force and torque balance equations: $z = \langle \bar{d}_f(\alpha) \rangle$.³⁴ Here, the probability of redundant configurations with $\bar{d}_f < d_f$ can be estimated by re-weighting all configurations by rotating into states of maximal redundancy.

The existence of redundant configurations explains the observed convergence in $z(\alpha)$ to values close to 8 for spherocylinders with large aspect ratios:^{62,68} for long spherocylinders the contacts are predominantly on the cylindrical part so that all normal forces are coplanar. As a consequence, the effective number of degrees of freedom is reduced by one, leading to $z = 8$.³⁴ The requirement of local force and torque balance can also be formulated as a constraint optimization problem on a factor graph, which describes the force transmission on a single particle.⁴³ Solving this problem with standard methods such as the cavity method predicts values of z in frictional packings and also allows for the computation of the distribution of contact forces in good agreement with experimental results.⁴³

Phase-diagram of jammed isotropic and anisotropic particles

The combination of the results for $\bar{W}(z)$ and $z(\alpha)$ leads to a complete theoretical prediction for the packing density $\phi(\alpha) = V_\alpha/\bar{W}(z(\alpha))$ of non-spherical particles without any adjustable parameters.³⁴ We estimate the maximum density of spherocylinders at $\alpha = 1.3$ with a density $\phi_{\max} = 0.731$ in good agreement with empirical data. The theory also reproduces well the density of dimers, estimating a maximum at $\alpha = 1.3$ with $\phi_{\max} \approx 0.707$. We have also calculated the packing fraction of lens-shaped particles, which can serve as approximations for oblate ellipsoidal shapes. Our theory yields $\phi_{\max} = 0.736$ for $\alpha = 0.8$. This shape represents the densest random packing of an axisymmetric shape known so far. The appearance of a maximum in ϕ for non-spherical shapes close to the sphere has been explained in a simple qualitative picture on the basis of the excluded volume V_{ex} .⁵⁶ For α close to 1, the ratio V_{ex}/V_α changes only slightly from the spherical value and a density increase results due to the additional orientational degrees of freedom, whereby the particles can fit into gaps by rotating, similar to the increase in packing efficiency due to polydispersity.⁷⁴ For larger α , V_{ex} exceeds V_α while z remains constant, so that the packing is dominated by the excluded volume and the packing fraction decreases. This argument can explain qualitatively the observed larger packing fraction of spherocylinders compared with dimers. The ratio V_{ex}/V_α is approximately equal for both shapes up to $\alpha \approx 1.2$, but for larger α the ratio for dimers increases beyond that of spherocylinders. The packing densities derived in our framework are interpreted as upper bounds to the empirically obtained densities and correspond to maximally random jammed states⁷⁵ by construction, since the distribution of contact angles in the first coordination shell is imposed to be uniform, avoiding any partial order.

By plotting $z(\alpha)$ against $\phi(\alpha)$ parametrically as a function of α , we obtain a phase diagram in the z - ϕ plane (Fig. 2). Surprisingly, we find that both dimer and spherocylinder packings appear as smooth continuations of spherical packings. The

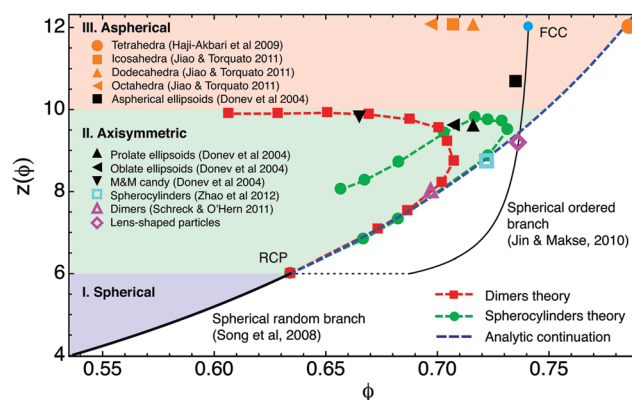


Fig. 2 Phase diagram of jammed matter. We plot our results for dimers and spherocylinders in the z - ϕ plane together with results from the literature for frictionless disordered packings of a selection of regular shapes. We have selected those shapes for which the z and ϕ values have been determined in the same simulation. The predicted spherical random branch eqn (2) (ref. 31) (thick black line) and a conjectured first order disorder–order transition at RCP for spheres⁷⁷ (dotted and thin black lines) are also indicated. We observe that the analytic continuation of RCP under deformation into dimers and spherocylinders provides an empirical bound to disordered packings in the phase diagram. The symmetry of the shape indicates the possible values of the coordination number z : (i) spheres have z between 4 (infinitely rough) and 6 (frictionless). (ii) Axisymmetric particles have z between 6 and 10. (iii) Fully aspherical particles have z between 10 and 12. Note that for polyhedra, z is associated with the total degrees of freedom blocked by the different types of contacts (face–face, face–vertex, vertex–vertex, face–edge).⁶⁷ The data point for lens-shaped particles is a theoretical prediction.³⁴

analytic form of this continuation from the spherical random branch can be derived (blue dashed line in Fig. 2).³⁴ A comparison of our theoretical results with empirical data for a large variety of shapes highlights that the analytic continuation provides a boundary line in the z - ϕ phase diagram. Maximally dense disordered packings appear to the left of this boundary, while the packings to the right of it are partially ordered. The spherical ordered branch provides another boundary, which separates tetrahedra from all other shapes: tetrahedra are the only shape that pack in a disordered way denser than spheres in a FCC crystal. We observe that the maximally dense packings of dimers, spherocylinders, lens-shaped particles and tetrahedra all lie surprisingly close to the analytic continuation of RCP. Whether there is any deeper meaning to this remains an open question.

The picture that emerges is that spherical packings can be generated between the RLP and RCP limits by variation of the inter-particle friction, since this leads to an increase in the coordination number under the isostatic condition from $z = 4$ to $z = 6$. Beyond RCP, the spherical equation of state can be continued smoothly by deforming the sphere into elongated shapes. Moreover, the spherical RCP is interpreted as the freezing point of disordered sphere packings, associated with a melting point at $\phi = 0.68$.^{76,77} The signature of this disorder–order transition is a discontinuity in the entropy density of jammed configurations as a function of the compactivity. This highlights the fact that beyond RCP, denser packing fractions of spheres can only be reached by partial crystallization up to the homogeneous FCC crystal phase.⁷⁵



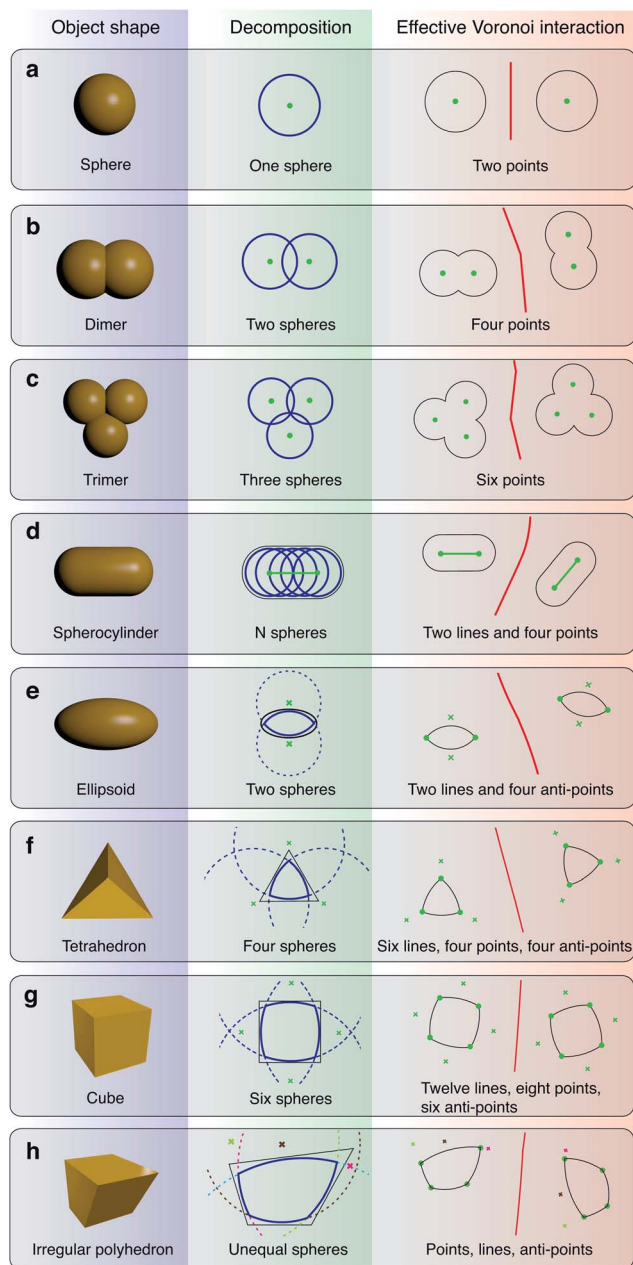


Fig. 3 Decomposition of various shapes to calculate the Voronoi boundary. The Voronoi boundary (VB) between two particles is defined as the hypersurface that contains all points equidistant to their surfaces. This implies that the VB between two equal spheres, *e.g.*, is that between two points at the centers of the spheres, so that the VB is generated effectively by the interaction of two points (a). Likewise, the VB between two spherocylinders is due to the effective interaction of two lines, since spherocylinders can be represented as dense overlaps of spheres (d). Arbitrary shapes can be decomposed into dense overlaps of spheres following certain design principles.⁹⁰ The VB between two such shapes can then be calculated following an exact algorithm that considers the effective Voronoi interactions between points and lines (a–d).³⁴ For shapes that are not naturally given as overlapping spheres (e–h), we propose alternatively an approximation in terms of a small number of intersecting spheres. In this way, two intersecting spheres (a lens-shaped particle) approximate an oblate ellipsoid and four intersecting spheres approximate a tetrahedron. The effective Voronoi interactions are then between points, lines, and anti-points (indicated by crosses).³⁴ Anti-points arise from the inversion of

Conclusions and outlook

The first-order transition of jammed spheres identified within Edwards' thermodynamics⁷⁷ is reminiscent of the entropy induced phase transition of equilibrium hard spheres, which is found at $\phi = 0.494$ and $\phi = 0.545$, respectively. However, it should be emphasized that the physical origin of these two transitions is fundamentally different: the equilibrium phase transition is a consequence of the maximization of the conventional entropy, while the transition at RCP of jammed spheres is driven by the competition between volume minimization and maximization of the entropy of jammed configurations S . For anisotropic particles at equilibrium, a disorder–order phase transition appears, *e.g.*, between isotropic and nematic phases of elongated shapes: for large α , Onsager's theory of equilibrium hard rods predicts a first order isotropic–nematic transition with freezing point at the rescaled density $\phi\alpha = 3.29$ and melting point at $\phi\alpha = 4.19$.⁷⁰ For colloidal suspensions of more complex shapes like polyhedra, both liquid crystal as well as plastic crystal and even quasicrystal phases have been found.^{78–80} By analogy with the case of jammed spheres, one might wonder whether packings of non-spherical particles exhibit similar transitions that might be characterized in the z – ϕ phase diagram. Packings of hard thin rods indeed satisfy a scaling law, where the RCP has been experimentally identified at $\phi\alpha \approx 5.4$.⁸¹

The Edwards' approach thus helps to elucidate how macroscopic properties of granular matter arise from the anisotropy of the constituents – one of the central questions in present day materials science.^{82,83} A better understanding of this problem will facilitate, *e.g.*, the engineering of new functional materials with particular mechanical responses by tuning the shape of the building blocks. A search in the space of object shapes for optimization can be performed by considering a small number of spheres and systematically exploring the different possible configurations.⁸⁴

Our approach eqn (1) can be applied to a large variety of both convex and non-convex shapes. The key is to parametrize the Voronoi boundary between two such shapes, which allows for the calculation of the Voronoi excluded volume and surface. In fact, analytical expressions for the Voronoi boundary can be derived following an exact algorithm for arbitrary shapes by decomposing the shape into overlapping and intersecting spheres (see Fig. 3). Therefore, a systematic search for maximally dense packings in the space of given object shapes can be performed using our framework. Extensions to mixtures and polydisperse packings can also be formulated. So far, exhaustive searches for dense packings have only been performed for

the effective interaction between the spheres in the decomposition. This is evident in the case of lens-shaped particles (e), where the interaction between the spheres is inverted compared to the case of dimers (b). The VB between two tetrahedra is then due to the interaction between the vertices (leading to four point interactions), the edges (leading to six line interactions), and the faces (leading to four anti-point interactions). This approach can be generalized to arbitrary polyhedra. Figure taken from ref. 34.



ordered packings using computer simulations^{85,86} and a combination of analytic and simulation techniques.⁸⁷ This has elucidated in particular the validity of Ulam's conjecture that the sphere is the worst packing object in 3d.⁸⁸ Analytical progress to prove this conjecture locally, that is, for shapes deformed from the sphere, has recently been made.⁸⁹

A more systematic investigation of disordered packings can shed light on the validity of a random variant of Ulam's conjecture, which so far has only been investigated in simulations.⁶⁷ Our analytic continuation from RCP highlights that this conjecture might hold more generally than previously assumed, containing not only convex shapes, but also a significant class of non-convex ones. Ultimately, our approach might lead to a more exhaustive theoretical investigation of Ulam's conjecture. Along the way one might be able to answer important questions such as if a shape that packs denser in a random configuration than in a regular one exists.⁷² Such objects could represent optimal glass formers with far reaching consequences for materials science.

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Mean-field theory of random close packings of axisymmetric particles

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Finding the optimal random packing of non-spherical particles is an open problem with great significance in a broad range of scientific and engineering fields. So far, this search has been performed only empirically on a case-by-case basis, in particular, for shapes like dimers, spherocylinders and ellipsoids of revolution. Here we present a mean-field formalism to estimate the packing density of axisymmetric non-spherical particles. We derive an analytic continuation from the sphere that provides a phase diagram predicting that, for the same coordination number, the density of monodisperse random packings follows the sequence of increasing packing fractions: spheres < oblate ellipsoids < prolate ellipsoids < dimers < spherocylinders. We find the maximal packing densities of 73.1% for spherocylinders and 70.7% for dimers, in good agreement with the largest densities found in simulations. Moreover, we find a packing density of 73.6% for lens-shaped particles, representing the densest random packing of the axisymmetric objects studied so far.

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Understanding the properties of assemblies of particles from the anisotropy of their building blocks is a central challenge in materials science^{1–3}. In particular, the shape that leads to the densest random packing has been systematically sought empirically^{4–17}, as it is expected to constitute a superior glass forming material¹. Despite the significance of random packings of anisotropic particles in a range of fields like self-assembly of nanoparticles, liquid crystals, glasses and granular processing¹⁸, there is yet no theoretical framework to estimate their packing density. Thus, random packings of anisotropic particles are typically investigated on a case-by-case basis using computer simulations, which have shown, for example, that elongated shapes like prolate ellipsoids and spherocylinders can pack considerably denser than the random close packing (RCP) fraction of spheres at $\phi_{\text{RCP}} \approx 0.64$. These shapes exhibit a maximum in the packing fraction for aspect ratios (length/width) close to the sphere^{4–6}.

Table 1 summarizes the empirical findings for maximal densities and highlights a further caveat of simulation and experimental studies: The protocol dependence of the final close-packed (or jammed) state leading to a large variance of the maximal packing fractions found for the same shape. This observation can be explained using the picture of a rugged energy landscape from theories of the glass phase¹⁹. Different algorithms get stuck in different metastable basins of the energy landscape, reaching different final packing states.

Here we present a mean-field approach to systematically study the packing fraction of a class of anisotropic shapes with rotational symmetry, which can therefore guide further empirical studies. Explicit results are obtained for axisymmetric particles like dimers, spherocylinders and lens-shaped particles, and we discuss generalizations to other shapes like tetrahedra, cubes and irregular polyhedra. Furthermore, we derive an analytic continuation of the spherical RCP which provides a phase diagram for these and other anisotropic particles like oblate and prolate ellipsoids. We first define the Voronoi volume of a non-spherical particle on which our calculation is based, and show that it can be calculated analytically for many different shapes by a decomposition of the shape into overlapping and intersecting spheres, which we organize into interactions between points, lines and anti-points. We then develop a statistical mean-field theory of the Voronoi volume to treat the particle correlations in the packing. This geometric mean-field approach is complemented by a quantitative estimation of the variation of the average contact number with the particle aspect ratio. The predicted packing density is interpreted as an upper bound of the empirically obtained packings.

Results

Voronoi boundary between non-spherical objects. We consider rotationally symmetric objects for which the aspect ratio α is defined as length/width, where the length is measured along the symmetry axis. In the following, we focus on the region $0 < \alpha < 2$, where the largest densities are found¹⁷. Our description of packings relies on a suitable tessellation of space into non-overlapping volumes²⁰. We use the standard Voronoi convention^{21,22}, where one associates with each particle the fraction of space that is closer to this particle than to any other one. This defines the Voronoi volume W_i of a particle i , which depends on the configurations $\mathbf{x} = (\mathbf{r}, \hat{\mathbf{t}})$ of all particles (including position $\mathbf{r}$ and orientation $\hat{\mathbf{t}}$). The total volume V occupied by N particles is $V = \sum_{i=1}^N W_i(\{\mathbf{x}_1, \dots, \mathbf{x}_N\})$, and the packing fraction of monodisperse particles of volume V_α and aspect ratio α follows as $\phi = NV_\alpha/V$. In order to determine W_i one has to know the Voronoi boundary (VB) between two particles i and j , which is the hypersurface that contains all points equidistant to both

Table 1 | Overview of packing fractions from simulations and experiments.

Shape	ϕ_{max}	Aspect ratio at ϕ_{max}	Reported z
Spherocylinder ⁵	0.653	1.5	9.8
M&M candy ⁶	0.665	0.5	
Spherocylinder ¹⁴	0.689		
Spherocylinder ⁸	0.694	1.4	8.6
Spherocylinder ⁴	0.695	1.4	
Dimer	0.697	1.4	
Dimer ¹²	0.703	1.4	8.0
Spherocylinder ¹⁵	0.703	1.5	9.6
Spherocylinder ⁹	0.704	1.4	
Oblate ellipsoid ⁶	0.707	0.6	
Dimer (theory)	0.707	1.3	8.74
Spherocylinder ¹⁰	0.708	1.5	9.1
Prolate ellipsoid ⁶	0.716	1.5	9.6
Spherocylinder ¹⁷	0.722	1.5	8.7
Spherocylinder (theory)	0.731	1.3	9.5
Lens-shaped particle (theory)	0.736	0.8	9.2
General ellipsoid ⁶	0.735		10.7
General ellipsoid ⁷	0.74		
Tetrahedron ¹³	0.76		
Tetrahedron ¹⁶	0.763		12
Tetrahedron ¹¹	0.7858		

The maximal packing fraction ϕ_{max} and reported coordination number z at ϕ_{max} of random packings of spherocylinders, dimers, ellipsoids and tetrahedra, determined from simulations and experiments. The aspect ratio is defined for rotationally symmetric objects. Some simulations do not report z . Results are separated by the symmetry of the object (rotationally symmetric and asymmetric) and ordered by packing fraction. From the available empirical data we cannot conclude whether spherocylinders pack better than dimers or ellipsoids of revolution, for instance.

particles (Fig. 1 for spherocylinders). The VB of the volume W_i along $\hat{\mathbf{c}}$, denoted by $l_i(\hat{\mathbf{c}})$, is the minimal one in this direction among all possible VBs of each particle j in the packing. It is formally obtained by the global minimization²⁰:

$$l_i(\hat{\mathbf{c}}) = \min_{j: s > 0} s(\mathbf{r}_j, \hat{\mathbf{t}}_j, \hat{\mathbf{c}}), \quad (1)$$

where $s(\mathbf{r}_j, \hat{\mathbf{t}}_j, \hat{\mathbf{c}})$ denotes the VB along $\hat{\mathbf{c}}$ between particles i and j with relative position $\mathbf{r}_j$ and orientation $\hat{\mathbf{t}}_j$ (Fig. 1). The Voronoi volume follows then exactly as the orientational integral,

$$W_i = \frac{1}{3} \oint d\hat{\mathbf{c}} l_i(\hat{\mathbf{c}})^3. \quad (2)$$

The VB between two equal spheres is identical to the VB between two points and is a flat plane perpendicular to the separation vector (Fig. 2a)²⁰. Finding the VB for more complicated shapes is a challenging problem in computational geometry, which is typically only solved numerically²³. We approach this problem analytically by considering a decomposition of the non-spherical shape into overlapping spheres. The VB is then determined as follows: Every segment of the VB arises due to the Voronoi interaction between a particular sphere on each of the two particles reducing the problem to identifying the correct spheres that interact. This identification follows an exact algorithm for a large class of shapes obtained by the union and intersection of spheres, which can be translated into an analytical expression of the VB as outlined in Fig. 3 for dimers, spherocylinders and lens-shaped particles.

For instance, a dimer is the union of a pair of spheres (Fig. 2b). The dimers VB is thus a composition of maximal four different surfaces depending on the relative orientation of the dimers defined by four points at the centre of each sphere (Fig. 3a).

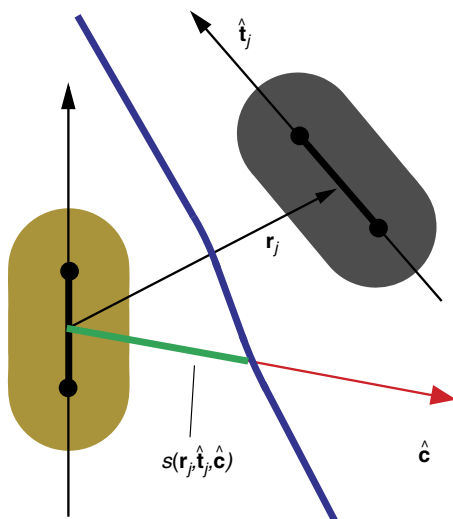


Figure 1 | Parametrization of the VB. The VB in blue, denoted by $s(\mathbf{r}_j, \hat{\mathbf{t}}_j, \hat{\mathbf{c}})$, along a direction $\hat{\mathbf{c}}$ between two spherocylinders of relative position $\mathbf{r}_j$ and orientation $\hat{\mathbf{t}}_j$.

The extension to trimers is straightforward (Fig. 2c). Likewise, n overlapping spheres lead to compositions of n surfaces. A spherocylinder is a dense overlap of spheres of equal radii and the VB interaction is identical to that between four points and two lines (Fig. 2d). The interactions then simplify into line–line, line–point, and point–point interactions, which generally lead to a curved VB for non-parallel orientations (Fig. 3b).

The Voronoi decomposition used for dimers and spherocylinders can be generalized to arbitrary shapes by using a dense filling of spheres with unequal radii²⁴. However, even if it is still algorithmically well defined, this procedure may become practically tedious for dense unions of polydisperse spheres. Alternatively one can apply specialized algorithms to compute numerical VBs between curved line segments²⁵. Here we propose an analytically tractable approach: Convex shapes can be approximated by intersections of a finite number of spheres. An oblate ellipsoid, for example, is well approximated by a lens-shaped particle, which consists of the intersection of two spheres; an intersection of four spheres is close to a tetrahedra, and six spheres can approximate a cube. This is illustrated in Fig. 2e–h, and the corresponding algorithms outlined in Fig. 3c. The main insight is that the effective Voronoi interaction of these shapes is governed by a symmetry: Points map to ‘anti-points’ (as the interactions between spheres is inverted; Fig. 3c). The VB of ellipsoid-like objects arises from the interaction between four anti-points and four points in two dimensions (Fig. 3c) or lines in three dimensions, and thus falls into the same class as spherocylinders. For cubes, the effective interaction is that of twelve lines, eight points and six anti-points (Fig. 2g). Analytic expressions of the VB for dimers and spherocylinders are calculated in the Supplementary Methods.

A statistical theory for Voronoi volume fluctuations. We turn the above formalism into a mean-field theory to calculate the volume fraction of a packing of monodisperse non-spherical objects. In order to take into account multi-particle correlations in the packing, we use a statistical mechanics treatment where the overall volume is expressed in terms of the average Voronoi volume $\bar{W}(z)$: $V = N\bar{W}(z)$ (ref. 20) characterized by the average coordination number z , which denotes the mean number of contacting neighbours in the packing. This approach is motivated by the observation that, as $N \rightarrow \infty$, packings exhibit reproducible

phase behaviour, which is characterized by only few observables such as ϕ and z (ref. 26). Our statistical mechanics framework is based on the Edwards ensemble approach, which considers the volume as a Hamiltonian of the system and attempts to find the minimum volume²⁷. Here $\bar{W}$ is given as the ensemble average of W_i over all particles in the packing: $\bar{W} = \langle W_i \rangle_i$. We obtain therefore from equation (2):

$$\begin{aligned} \bar{W} &= \left\langle \frac{1}{3} \oint d\hat{\mathbf{c}} l_i(\hat{\mathbf{c}})^3 \right\rangle_i = \frac{1}{3} \oint d\hat{\mathbf{c}} \langle l_i(\hat{\mathbf{c}})^3 \rangle_i \\ &= \frac{1}{3} \oint d\hat{\mathbf{c}} \int_{c^*(\hat{\mathbf{c}})}^{\infty} dc c^3 p(\mathbf{c}). \end{aligned} \quad (3)$$

In the last step, we have introduced the probability density $p(\mathbf{c})$, which contains the probability to find the VB at $\mathbf{c}$ in the direction $\hat{\mathbf{c}}$. The lower integration limit $c^*(\hat{\mathbf{c}})$ is the minimal value of the boundary along $\hat{\mathbf{c}}$, which corresponds to the hard-core boundary of the particle in that direction. We introduce the cumulative distribution function (CDF) $P(\mathbf{c})$ via the usual definition $p(\mathbf{c}) = -\frac{d}{dc}P(\mathbf{c})$. Substituting the CDF in equation (3) and performing an integration by parts leads to the volume integral

$$\bar{W}(z) = \int d\mathbf{c} P(\mathbf{c}, z), \quad (4)$$

where we indicate the dependence on z . In a geometric picture²⁰, $P(\mathbf{c}, z)$ is interpreted as the probability that $N-1$ particles are outside a volume Ω centred at $\mathbf{c}$ (see Fig. 4), as otherwise they would contribute a shorter VB. This leads to the definition

$$\Omega(\hat{\mathbf{c}}, \hat{\mathbf{t}}) = \int d\mathbf{r} \Theta(c - s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})) \Theta(s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})), \quad (5)$$

where $\Theta(x)$ denotes the usual Heavyside step. We refer to Ω as the Voronoi excluded volume, which extends the standard concept of the hard-core excluded volume V_{ex} considered by Onsager in his theory of elongated equilibrium rods²⁸ (Fig. 4).

The dependence of $P(\mathbf{c}, z)$ on Ω has been treated at a mean-field level in Song *et al.*²⁰ and has been derived from a theory of correlations using liquid state theory in²⁹ for high-dimensional sphere packings. In both cases it provides a Boltzmann-like exponential form $P(\mathbf{c}, z) \propto \exp\left\{-\int_{\Omega(\mathbf{c})} d\mathbf{r} \rho(\mathbf{r}, z)\right\}$ in the limit $N \rightarrow \infty$, where $\rho(\mathbf{r}, z)$ is the density of spheres at $\mathbf{r}$.

The crucial step is to generalize this result to anisotropic particles. Following Onsager²⁸, we treat particles of different orientations as belonging to different species. This is the key assumption to treat orientational correlations within a mean-field approach. Thus, the problem for non-spherical particles can be mapped to that of polydisperse spheres for which P factorizes into the contributions of the different radii³⁰. We thus obtain the factorized form:

$$P(\mathbf{c}, z) = \exp \left\{ - \int d\hat{\mathbf{t}} \int_{\Omega(\mathbf{c}, \hat{\mathbf{t}})} d\mathbf{r} \rho(\mathbf{r}, \hat{\mathbf{t}}, z) \right\}, \quad (6)$$

where $\rho(\mathbf{r}, \hat{\mathbf{t}}, z)$ is the density of particles with orientation $\hat{\mathbf{t}}$ at $\mathbf{r}$.

Next, we assume an approximation of this density in terms of contact and bulk contributions, which is motivated by the connection with the radial distribution function in spherical theories in both high and low dimensions^{20,29}. The contact contribution relies on the condition of contact between the two particles of a given relative position $\mathbf{r}$ and orientation $\hat{\mathbf{t}}$, which defines the contact radius $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$: r^* is the value of r for which the two particles are in contact without overlap. In the case of equal spheres the contact radius is simply $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}}) = 2a$. For non-spherical objects, $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$ depends on the object shape and the relative orientation (Supplementary Methods). Using $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$ we

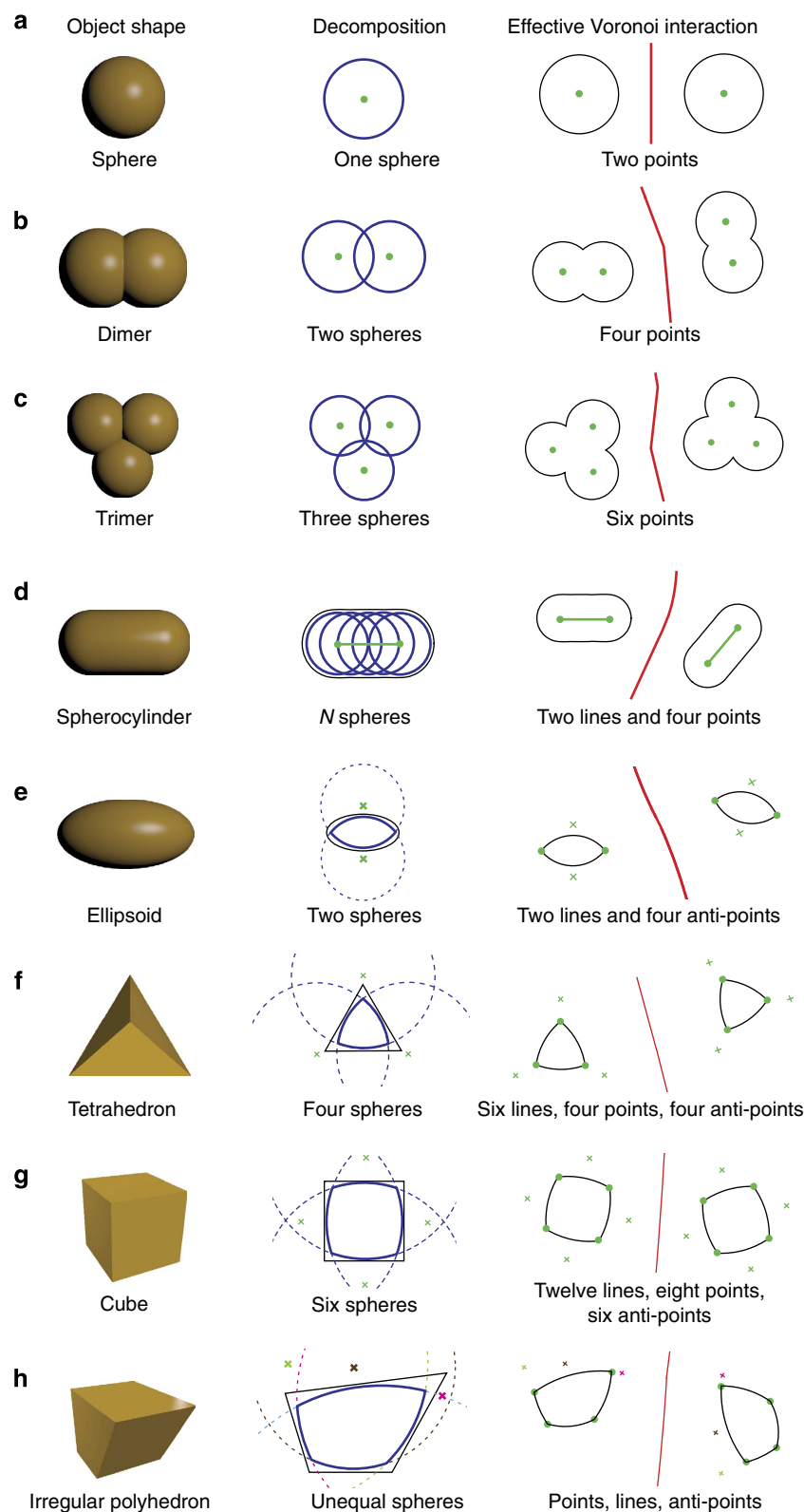


Figure 2 | Decomposition of various shapes and effective Voronoi interactions. Arbitrary object shapes can be decomposed into unions and intersections of spheres. **(a–d)** Union of spheres. The VB between two such objects is equivalent to the VB between the point multiplets at the centre of the spheres, as shown for four basic shapes. **(e–g)** Intersection of spheres. The VB between such intersections is equivalent to that between multiplets of ‘anti-points’ at the centre of the spheres, indicated by crosses, and, in addition, lines at the edges of the intersections, shown as points in **e–d**. The additional lines arise due to the positive curvature at the singular intersections, resulting in edges that point outwards from the particle rather than inwards. In the case of dimers and trimers shown in **b,c**, the curvature is negative and the edges do not influence the VB. The generalization to **(f)** tetrahedra-like, **(g)** cubes and **(h)** irregular polyhedra-like shapes is straightforward. Note that the VBs drawn in **e–h** are only qualitative.

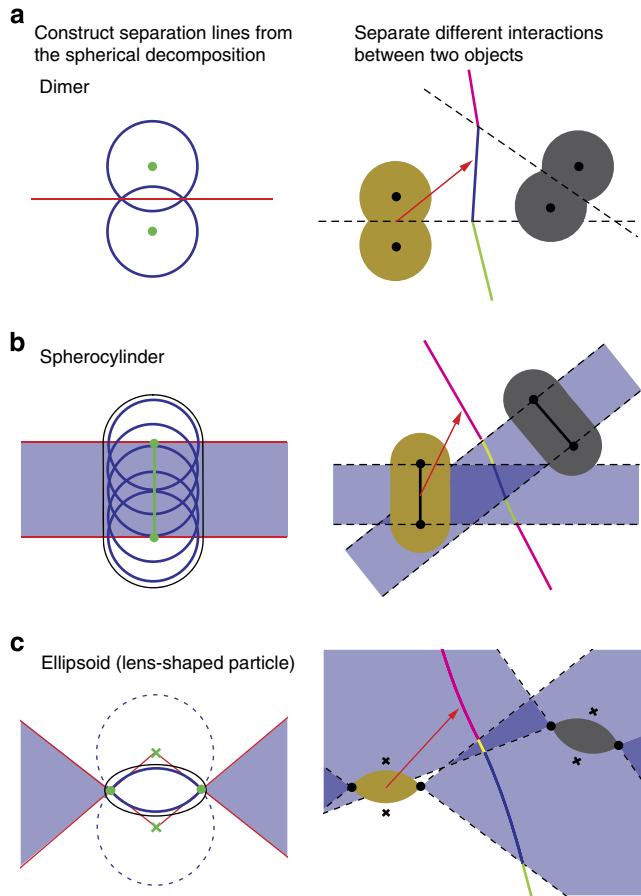


Figure 3 | Analytical solution to determine the VB for non-spherical objects. (a) The VB between the two objects of a given relative position and orientation consists of the VBs between particular spheres on each of the two objects. The spheres that interact are determined by separation lines given as the VBs between the spheres in the filling. For dimers, there is one separation line for each object, tessellating space into four areas, in which only one interaction is correct. The pink part in **a**, for example, is the VB between the two upper spheres. (b) The dense overlap of spheres in spherocylinders leads to a line as effective Voronoi interaction at the centre of the cylindrical part. This line interaction has to be separated from the point interactions due to the centres of the spherical caps as indicated. Overall, the two separation lines for each object lead to a tessellation of space into nine different areas, where only one of the possible line-line, line-point, point-line, and point-point interactions is possible. The yellow part in **b**, for example, is due to the upper point on spherocylinder 1 and the line of 2. Regions of line interactions are indicated by blue shades. (c) The spherical decomposition of ellipsoid-like shapes is analogous to dimers, only that now the opposite sphere centres interact. We indicate this inverted interaction by a cross at the centres of the spheres and refer to these points as ‘anti-points’. In addition, the positive curvature at the intersection point leads to an additional line interaction, which is a circle in 3D (a point in 2D) and indicated here by two points. The separation lines are then given by radial vectors through the intersection point/line. The Voronoi interaction between the two ellipsoids is thus given by two pairs of two anti-points and a line, which is the same class of interactions as spherocylinders. The different point and line interactions are separated analogous to spherocylinders, as shown.

can separate bulk and contact terms in $\rho(\mathbf{r}, \hat{\mathbf{t}}, z)$ as in^{20,29}:

$$\rho(\mathbf{r}, \hat{\mathbf{t}}, z) = \frac{1}{4\pi} [\bar{\rho} \Theta(r - r^*(\mathbf{r}, \hat{\mathbf{t}})) + \sigma(z) \delta(r - r^*(\mathbf{r}, \hat{\mathbf{t}}))]. \quad (7)$$

The prefactor $1/4\pi$ is the density of orientations, which we assume isotropic. The symbols $\bar{\rho}$ and $\sigma(z)$ stand for the average

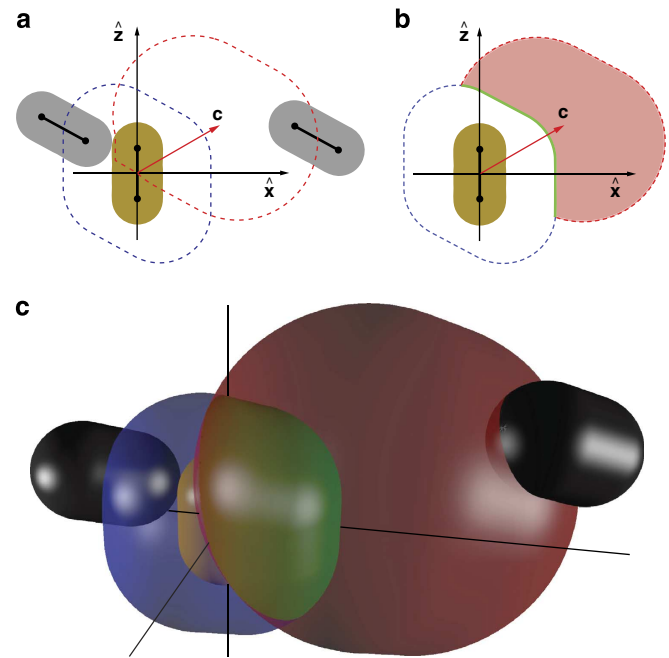


Figure 4 | The Voronoi excluded volume and surface. (a) The hard-core repulsion between the two objects defines the hard-core excluded volume V_{ex} (enclosed by a dashed blue line): This volume is excluded for the centre of mass of any other object. Packings of rods in the limit $\alpha \rightarrow \infty$ can be described by a simple random contact equation based on V_{ex} (ref. 51). We introduce the Voronoi excluded volume Ω (enclosed by a dashed red line), which is the basis of our statistical theory of the Voronoi volume. The volume Ω , equation (5), is excluded by the condition that no other particle should contribute a VB smaller than c in the direction $\hat{\mathbf{c}}$, which defines the CDF $P(\mathbf{c}, z)$. (b) Taking into account the hard-core exclusion leads to the effective Voronoi excluded volume V^* (indicated as red volume), which is excluded for bulk particles. Likewise, the overlap of V_{ex} and Ω excludes the surface S^* (thick green line) for all contacting particles. The volumes are shown here for a single orientation $\hat{\mathbf{t}}$. (c) The 3D plot corresponding to **b**: The central particle is in brown, V_{ex} is indicated in blue, V^* in red, and S^* in green.

free-volume of particles in the bulk and the average free-surface of particles at contact, respectively, which are discussed further below. The approximation equation (7) corresponds to considering a pair distribution function as a delta function modelling the contact particles plus a constant term modelling the particles in the bulk²⁹, which are thus considered as a uniform structure. These assumptions are further tested in the Methods section.

Substituting equation (7) into equation (6) leads to our final result for the CDF:

$$P(\mathbf{c}, z) = \exp\{-\bar{\rho}(\bar{W})V^*(\mathbf{c}) - \sigma(z)S^*(\mathbf{c})\}. \quad (8)$$

Here we have explicitly written the dependence of $\bar{\rho}$ on $\bar{W}$, which is important to interpret equation (4) as a self-consistent equation to obtain the volume fraction of the packing. The free-volume per particle in the bulk depends specifically on $\bar{W}(z)$ as $\bar{\rho} = 1/(\bar{W}(z) - V_{\alpha})$.

The CDF thus factorizes into two contributions: A contact term:

$$P_{\text{C}}(\mathbf{c}, z) = \exp\{-\sigma(z)S^*(\mathbf{c})\}, \quad (9)$$

and a bulk term:

$$P_{\text{B}}(\mathbf{c}) = \exp\{-\bar{\rho}(\bar{W})V^*(\mathbf{c})\}, \quad (10)$$

such that

$$P(\mathbf{c}, z) = P_C(\mathbf{c}, z) \times P_B(\mathbf{c}). \quad (11)$$

The volume V^* is the volume excluded by Ω for bulk particles and takes into account the overlap between Ω and the hard-core excluded volume V_{ex} : $V^* = \langle \Omega - \Omega \cap V_{\text{ex}} \rangle_{\hat{\mathbf{c}}}$, where $\langle \dots \rangle_{\hat{\mathbf{c}}}$ denotes an orientational average. Likewise, S^* is the surface excluded by Ω for contacting particles: $S^* = \langle \partial V_{\text{ex}} \cap \Omega \rangle_{\hat{\mathbf{c}}}$, where ∂V_{ex} denotes the boundary of V_{ex} . The volumes V_{ex} and Ω as well as the resulting V^* and S^* are calculated in the Supplementary Methods and shown in Fig. 4 for spherocylinders.

The surface density $\sigma(z)$ is a measure for the available surface for contacts when the packing is characterized by an average coordination number z . We evaluate this density by simulating random local configurations of one particle with z non-overlapping contacting particles and determining the average available free-surface. This surface is given by $S^*(\mathbf{c}_m)$, where $\mathbf{c}_m$ is the minimal contributed VB among the z contacts in the direction $\hat{\mathbf{c}}$. Averaging over many realizations with a uniform distribution of orientations and averaging also over all directions $\hat{\mathbf{c}}$ provides the surface density in the form,

$$\sigma(z) = \frac{1}{\langle \langle S^*(\mathbf{c}_m) \rangle \rangle_{\hat{\mathbf{c}}}}. \quad (12)$$

In this way, we can only calculate $\sigma(z)$ for integer values of z . For fractional z that are predicted from our evaluation of degenerate configurations in the next section, we use a linear interpolation to obtain $\bar{W}(z)$.

Equations (4) and (8) lead to a self-consistent equation for the average Voronoi volume $\bar{W}(z)$ in the form: $\bar{W}(z) = \mathcal{F}[\bar{W}(z)]$. Analytic expressions for V^* and S^* can be derived in the spherical limit in closed form, where also the self-consistency equation can be solved exactly²⁰. For non-spherical shapes we resort to a numerical integration to obtain V^* and S^* . Equation (4) can then be solved numerically, which yields $\bar{W}(z)$, and subsequently the equation of state for the volume fraction versus coordination number, $\phi(z, \alpha) = V_{\alpha}/\bar{W}(z)$, in numerical form (denoting explicitly the dependence on α).

Variation of the coordination number with aspect ratio. In this purely geometric theory of the average Voronoi volume, the packing fraction is given as $\phi(z, \alpha)$, with z and α free parameters, in principle. In practice, z is fixed by the symmetry properties of

the object shape, $z(\alpha)$, and the physical condition of mechanical stability, requiring force and torque balance on every particle. Under the assumption of minimal correlations, these conditions typically motivate the isostatic conjecture based on Maxwell's counting argument³¹: $z = 2d_f$, with d_f the number of degrees of freedom, giving $z = 6$ for fully symmetric objects (spheres), $z = 10$ for rotationally symmetric shapes like spherocylinders, dimers and ellipsoids of revolution⁶, and $z = 12$ for shapes with three different axes like aspherical ellipsoids and tetrahedra¹³. While the isostatic conjecture is well-satisfied for spheres, packings of non-spherical objects are in general hypoconstrained with $z < 2d_f$, where $z(\alpha)$ increases smoothly from the spherical value for $\alpha > 1$ (ref. 6). The fact that these packings are still in a mechanically stable state can be understood in terms of the occurrence of stable degenerate configurations (Fig. 5), which reduce the effective number of degrees of freedom³². However, the observed variation $z(\alpha)$ could not be explained quantitatively so far. Here we deduce the relation $z(\alpha)$ by evaluating the probability of finding these degenerate configurations to provide a prediction of $\phi(\alpha)$ in close form.

In a degenerate configuration, force balance already implies torque balance, as the net forces are aligned with the inner axis of the particle (Fig. 5). This implies that there is redundancy in the set of force and torque balance equations for mechanical equilibrium as force and torque balance equations are not linearly independent. Our evaluation of these degenerate configurations is based on the assumption that a particle is always found in an orientation such that the redundancy in the mechanical equilibrium conditions is maximal. This condition allows us to associate the number of linearly independent equations involved in mechanical equilibrium with the set of contact directions. Averaging over the possible sets of contact directions then yields the average effective number of degrees of freedom $\bar{d}_f(\alpha)$, from which the coordination number follows as $z(\alpha) = 2\bar{d}_f(\alpha)$ (Methods).

The results for $z(\alpha)$ are shown in Fig. 6a for prolate ellipsoids of revolution, spherocylinders, dimers and lens-shaped particles. We are able to recover the observed continuous transition as a function of α from the isostatic coordination number for spheres, $z = 6$ at $\alpha = 1$, to the isostatic value $z = 10$, for aspect ratios above ≈ 1.5 . The trend compares well to known data for ellipsoids⁶ and spherocylinders^{10,17}. In particular, our approach explains the decrease of z for higher aspect ratios observed in simulations of spherocylinders^{10,17}. For large α , the most probable case is to have

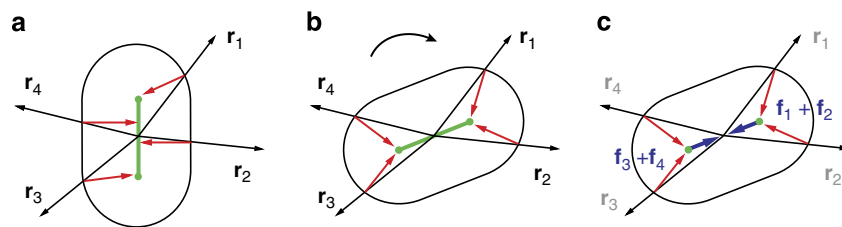


Figure 5 | Quantitative method to calculate $z(\alpha)$. (a) A 2D sketch of a spherocylinder with a random configuration of contact directions $\mathbf{r}_j$. The associated forces are along directions $\hat{\mathbf{n}}_j$ normal to the surface (indicated in red) and torques are along $\mathbf{r}_j \times \hat{\mathbf{n}}_j$. From these directions, one can determine if mechanical equilibrium has some redundancy, that is, if force and torque balance equations are not linearly independent. The configuration shown has no redundancy: The equivalent situation in three dimensions would show force and torque balance equations as five different constraints (the most general case for a 3D particle would be six constraints, but the torque along the axis of a spherocylinder is always vanishing, due to its rotational symmetry). (b) Here the spherocylinder is rotated. With the same contact directions $\mathbf{r}_j$ as in (a), the contact force directions $\hat{\mathbf{n}}_j$ are now modified. We explore the space of possible orientations for the spherocylinder, and try to find configurations which maximize redundancy in the mechanical equilibrium conditions. (c) As an example, this orientation exhibits some redundancy: All the contacts are on the spherical caps of the spherocylinder. Therefore, $\mathbf{f}_1 + \mathbf{f}_2$ and $\mathbf{f}_3 + \mathbf{f}_4$ are aligned with the spherocylinder axis and the condition of force balance automatically implies torque balance. If this is the orientation of the spherocylinder for which redundancy is maximal, we associate the number of linearly independent equations (that is, the effective number of degrees of freedom) from the mechanical equilibrium condition with the set of contact directions $\{\mathbf{r}_j\}$ and perform an average over the possible sets of $\{\mathbf{r}_j\}$. This yields the averaged effective number of degrees of freedom $\bar{d}_f(\alpha)$ for a spherocylinder having an aspect ratio α and the coordination number follows as $z(\alpha) = 2\bar{d}_f(\alpha)$. Note that for non-convex shapes like dimers, the resulting z is the number of contacting neighbours, not the number of contacts, which can exceed the former.

contacts only on the cylindrical part of the particle, so that all normal forces are coplanar reducing the effective number of degrees of freedom by one. Consequently, $z \rightarrow 8$ as $\alpha \rightarrow \infty$, as we obtain in Fig. 6a. This decrease is specific to spherocylinders, and not observed for dimers or ellipsoids, since the normal forces are not coplanar.

Phase diagram of non-spherical particles. Our calculation leads to a close theoretical prediction for the packing density $\phi(\alpha) = \phi(z(\alpha), \alpha)$ which does not contain any adjustable parameters. Figure 6b shows the prediction for dimers, spherocylinders and lens-shaped particles. For spherocylinders, results in the literature on $\phi(\alpha)$ vary greatly (Table 1), but all show a peak at around $\alpha \approx 1.3$ – 1.5 , which is captured by our formalism. We predict the maximum density of spherocylinders at $\alpha = 1.3$ with a density $\phi_{\max} = 0.731$ and that of dimers at $\alpha = 1.3$ with $\phi_{\max} = 0.707$. We have also calculated the packing fraction of the lens-shaped particles of Fig. 3c, which yields $\phi_{\max} = 0.736$ for $\alpha = 0.8$. This shape represents the densest random packing of an axisymmetric shape known so far.

We further investigate packings of non-spherical objects in the z - ϕ representation. This change in perspective allows us to characterize packings of differently shaped objects in a phase diagram. By plotting $z(\alpha)$ against $\phi(\alpha)$ parametrically as a function of α , we obtain a phase diagram for jammed anisotropic

particles in the z - ϕ plane (Fig. 6c). In the same diagram, we also plot the equation of state obtained with the present theory in the case of spheres in²⁰: $\phi_{\text{sph}}(z) = z/(z + 2\sqrt{3})$, which is valid between the two isostatic limits of frictionless spheres $z = 6$ and infinite frictional spheres at $z = 4$. Surprisingly, we find that both dimer and spherocylinder packings follow an analytical continuation of these spherical packings. This result highlights that the spherical random branch can be continued smoothly beyond the RCP in the z - ϕ plane.

The analytical continuation of RCP is derived by solving the self-consistent equation (4) close to the spherical limit (Supplementary Methods):

$$\phi(z) = \left(1 + \omega_1 \frac{1 + g_1(\omega_1) \left(\frac{z}{\bar{z}} - 1 \right) \frac{M_b}{M_z}}{\left[\frac{z}{\bar{z}} - g_2(\omega_1) \left(\frac{z}{\bar{z}} - 1 \right) \frac{M_b}{M_z} \right] \left[1 + \left(\frac{z}{\bar{z}} - 1 \right) \frac{M_v}{M_z} \right]} \right)^{-1} \quad (13)$$

here $\omega_1 = 1/\sqrt{3}$ denotes the spherical free-volume at RCP defined as $\omega_1 = 1/\phi_{\text{sph}} - 1$ evaluated at $z = 6$ as calculated in²⁰, $\bar{z} = 6$ is the spherical isostatic value, and the functions $g_{1,2}$ can be expressed in terms of exponential integrals. The dependence of equation (13) on the object shape is entirely contained in the geometrical parameters M_b , M_v , and M_z : M_b and M_v quantify the first-order deviation from the sphere at $\alpha = 1$ of the object's

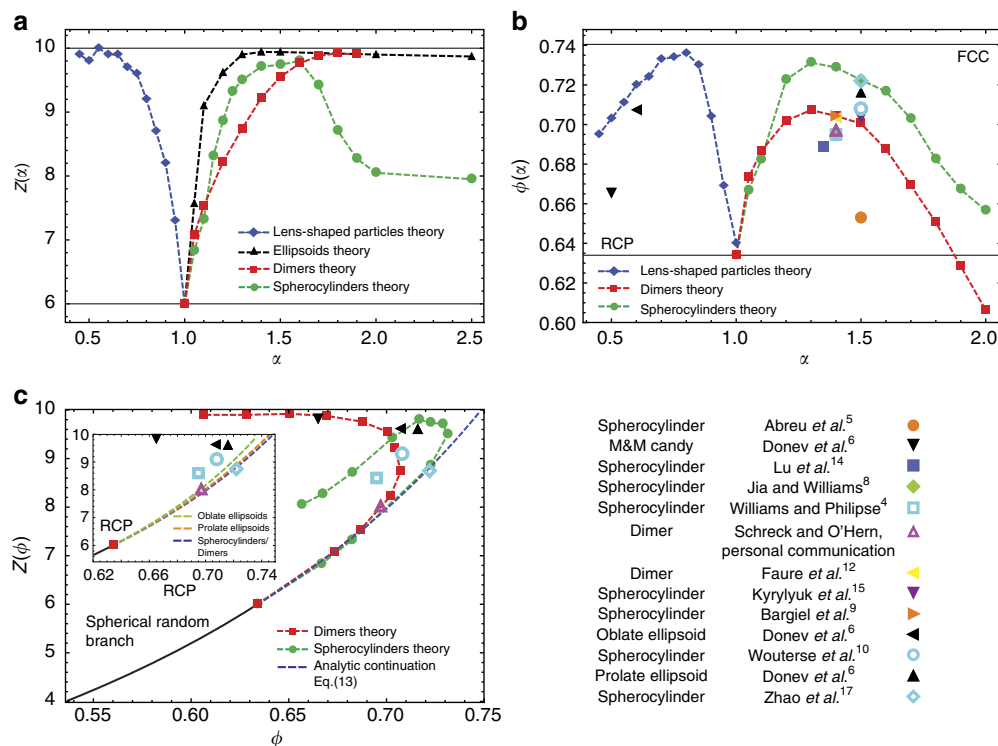


Figure 6 | Theoretical predictions for packings of particles with various shapes. (a) The function $z(\alpha)$ determined by evaluating the probability of degenerate configurations. Both spherocylinders and dimers increase up to just below the isostatic value $z = 10$. For dimers, $z(\alpha)$ is the number of contacting neighbours, not the number of contacts, as a single contacting particle can have more than one contacting point. For spherocylinders, z reduces to 8 for large α , as the forces acting on the cylindrical part are coplanar and reduce the effective degree of freedom. We also include the results from our method for prolate ellipsoids of revolution and lens-shaped particles. (b) The predicted packing fraction $\phi(\alpha)$ of spherocylinders, dimers and lens-shaped particles compared with simulation results of maximal densities from the literature. We predict the maximal packing fraction of spherocylinders $\phi_{\max} = 0.731$ at $\alpha = 1.3$ and of dimers $\phi_{\max} = 0.707$ at $\alpha = 1.3$, demonstrating that spherocylinders pack better than dimers. For the lens-shaped particles we obtain $\phi_{\max} = 0.736$ at $\alpha = 0.8$. (c) By plotting z vs ϕ we obtain a phase diagram for smooth shapes. We observe that the spherical random branch ϕ_{sph} , which ends at the RCP point at (0.634, 6) (ref. 20), in fact continues smoothly upon deformation into dimers and spherocylinders as predicted by our theory. The spherocylinder continuation provides a boundary for all known packing states of rotationally symmetric shapes. Inset: The continuations from RCP. For a given value of z , the densest packing is achieved by spherocylinders, followed by dimers, prolate ellipsoids and oblate ellipsoids. Note that the continuations for spherocylinders and dimers are almost identical.

hard-core boundary and its volume, respectively, while M_z measures the first-order change in the coordination number upon deformation of the sphere. The resulting continuations $z(\phi)$ obtained by inverting equation (13) for different object shapes are plotted in the inset of Fig. 6c.

For the smooth shapes considered, we find generally that denser packing states are reached for higher coordination numbers. For a given value of z , spherocylinders achieve the densest packing, followed by dimers, prolate ellipsoids and oblate ellipsoids, as seen in the inset of Fig. 6c. We observe that the densest packing states for dimers and spherocylinders found in simulations lie almost exactly on the continuation, while the one of the ellipsoids deviate considerably.

Comparison with empirical data. Table 1 indicates that there is a finite range of densities for random jammed packings according to the particular experimental or numerical protocol used (denoted as a J-line in the case of jammed spheres^{19,33}). On the other hand, our mean-field theory predicts a single density value and Fig. 6 indicates that our predictions are an upper bound of the empirical results. We interpret these results in terms of current views of the jamming problem developed in the limiting case of spheres, where the question of protocol-dependency of packings has been systematically investigated.

Random close packings can be considered as infinite-pressure limits of metastable glass states, which was shown theoretically in refs 19,34–36 and confirmed in computer simulations in ref. 37. Indeed, there exist a range of packing fractions named as $[\phi_{th}, \phi_{GCP}]$ following the notation of mean-field Replica Theory (RT)¹⁹. Here ϕ_{GCP} stands for the density of the ideal glass close packing and is the maximum density of disordered packings, while ϕ_{th} is the infinite-pressure limit of the least dense metastable states. In RT, the states $[\phi_{th}, \phi_{GCP}]$ are all isostatic.

From the point of view of simulations, the well-known Lubachevsky–Stillinger (LS) protocol³³ provides this range of packings for different compression rates. The densities $[\phi_{th}, \phi_{GCP}]$ are achieved by the corresponding compression rates (from large to small) $[\gamma_{th}, \gamma_{GCP} \rightarrow 0]$. Compression rates larger than γ_{th} all end to ϕ_{th} . The threshold value γ_{th} corresponds to the relaxation time $1/\gamma_{th}$ of the least dense metastable glass states. The denser states at GCP are unreachable by experimental or numerically generated packings, as it requires to equilibrate the system in the ideal glass phase, a region where the relaxation time is infinite. In general, large compression rates lead to lower packing fractions. This picture was investigated for sphere packings in^{33,38} and it is particularly valid for high-dimensional systems where crystallization is avoided¹⁹.

Random close packings are also known to display sharp structural changes^{39–43} signalling the onset of crystallization at a freezing point ϕ_c (ref. 18). All the (maximally random) jammed states along the segment $[\phi_{th}, \phi_{GCP}]$ can be made denser at the cost of introducing some partial crystalline order. Support for a order/disorder transition at ϕ_c is also obtained from the increase of polytetrahedral substructures up to RCP and its consequent decrease upon crystallization⁴⁴. In terms of protocol preparation like the LS algorithm, there exists a typical time scale t_c corresponding to crystallization. Crystallization appears in LS^{18,19,41} if the compression rate is smaller than $\gamma_c = 1/t_c$ around the freezing packing fraction⁴⁵. A possible path to avoid crystallization and obtain RCP in the segment $[\phi_{th}, \phi_{GCP}]$ is to equilibrate with $\gamma > \gamma_c$ to pass the freezing point, and eventually setting the compression rate in the range $[\gamma_{th}, \gamma_{GCP} \rightarrow 0]$ to achieve higher volume fraction.

As the present statistical mechanics framework is based on the Edwards ensemble approach²⁷, our prediction of the packing

density ϕ_{Edw} corresponds to the ensemble average over the configuration space of random states at a fixed coordination number. As the volume has the role of the Hamiltonian, the energy minimization in equilibrium statistical mechanics is replaced in our formalism by a volume minimization: The highest volume fraction for a given disordered system is achieved in the limit of zero compactivity. Therefore, the present framework provides a mean-field estimation of such a maximal volume fraction (minimum volume) of random packings with no crystallization. As we perform an ensemble average over all packings at a fix coordination number, the obtained volume fraction ϕ_{Edw} corresponds to the one with the largest entropy (called largest complexity in RT) along $[\phi_{th}, \phi_{GCP}]$. This point needs not to be ϕ_{th} , and in general it is a larger volume fraction. Thus, $\phi_{th} < \phi_{Edw} < \phi_{GCP}$.

The above discussion can be translated to the present case of non-spherical particles. In this case, unfortunately, there is no detailed study of the protocol-dependent packing density as done by^{19,33,38} for spheres. However, the survey of the available simulated data obtained by different groups (Table 1 and Fig. 6b,c) can be interpreted analogously as for spheres. In the case of spherocylinders, packings have been obtained in the range $[0.653, 0.722]$ (these minimum and maximum values have been obtained in ref. 5 and in ref. 17, respectively, see Table 1). Our predicted density is 0.731, representing an upper bound to the simulated results. In the case of dimers, there are two simulations giving a density of 0.697 (C. F. Schreck and C. S. O'Hern (2011) personal communication) and 0.703 (ref. 12), which are both smaller than and very close to our prediction 0.707. Thus, our prediction is interpreted as the upper limit in the range of packings observed with numerical algorithms. Under this scenario, which is consistent with analogous three-dimensional (3D) spherical results, packings may exist in the region $[\phi_{th}, \phi_{Edw}]$, and our theory is a mean-field estimation of ϕ_{Edw} . This region is very small for spheres but the above evidence indicates that non-spherical particles may pack randomly in a broader range of volumes. The present framework estimates the upper bound for such a range.

Discussion

We would like to stress that our analytic continuation is non-rigorous and appears as the solution of our mean-field theory for first-order deviations in α from the sphere using suitable approximations. The shapes of dimers, spherocylinders, ellipsoids are then all shown to increase the density of the random packing to first-order. In the case of regular (crystal) packings, recent mathematically rigorous work has shown in fact that for axisymmetric particles any small deformation from the sphere will lead to an increase in the optimal packing fraction of the crystal⁴⁶. This appears only in 3D and is related to Ulam's conjecture stating that the sphere is the worst case scenario for ordered packings in 3D (ref. 47). A full mathematical proof of this conjecture is still outstanding, but so far all computer simulations verify the conjecture. In particular, recent advances in simulation techniques allow to generate crystal packings of a large variety of convex and non-convex objects in an efficient manner^{48,49}. The extensive study of de Graaf *et al.*⁴⁸ has extended the verification of Ulam's conjecture to the first eight regular prisms and antiprisms, the 92 Johnson solids, and the 13 Catalan solids. The verification for regular n -prisms and n -antiprisms can be extended to arbitrary n using this method, providing an exhaustive empirical verification of the conjecture for these regular shapes. We remark that a random analogue of Ulam's packing conjecture has been proposed and verified for the Platonic solids (apart from the cube) in simulations¹⁶. The results presented here support the random version of Ulam's conjecture

and might help in investigating this conjecture further from a theoretical point of view.

We believe that our decomposition of various shapes into intersections and overlaps of spheres will be a useful starting point for a systematic investigation of this issue. Our approach can be systematically continued beyond the axisymmetric shapes considered here. For instance, in Fig. 2e–h, we have 2,3,6,n anti-points to describe ellipsoids and polyhedra of increasingly varying complexity. The challenge would be to implement our algorithm to calculate the resulting Voronoi excluded volumes that appear in our mean-field theory. For this, one might also consider a fully numerical evaluation using, for example, graphics hardware²⁵.

Methods

Quantitative method to calculate $z(\alpha)$. Mathematically, we can write the local mechanical equilibrium on a generic non-spherical frictionless particle having k contacts defined by their location $\mathbf{r}_j$, normal $\hat{\mathbf{n}}_j$ and force $f_j\hat{\mathbf{n}}_j$, as:

$$\begin{pmatrix} \hat{\mathbf{n}}_1 & \dots & \hat{\mathbf{n}}_k \\ \mathbf{r}_1 \times \hat{\mathbf{n}}_1 & \dots & \mathbf{r}_k \times \hat{\mathbf{n}}_k \end{pmatrix} \begin{pmatrix} f_1 \\ \vdots \\ f_k \end{pmatrix} \equiv \underline{\underline{N}} \underline{\underline{f}} = 0, \quad (14)$$

where $\underline{\underline{N}}$ is a $d_f \times k$ matrix. A local degenerate configuration has a matrix $\underline{\underline{N}}$ such that $\text{rank}(\underline{\underline{N}}) < \min(d_f, k)$. We base our evaluation on two assumptions: (i) Contact directions around a particle in the packing are uncorrelated, and (ii) Given one set of contact directions, a particle i is found in an orientation $\hat{\mathbf{t}}_i$ such that the

redundancy in the mechanical equilibrium conditions is maximal, that is, $\text{rank}(\underline{\underline{N}}_{\hat{\mathbf{t}}_i})$ is a minimum. Note that $\underline{\underline{N}}_{\hat{\mathbf{t}}_i}$ depends on $\hat{\mathbf{t}}_i$, as only the absolute direction of contact points are chosen, and thus rotating particle i affects the direction and normal of these contacts with respect to particle i . This situation is described in Fig. 5c, which includes a two-dimensional (2D) sketch of a 3D degenerate configuration that we observe often in our procedure. In this case, the rank is reduced by one unit, and the probability of occurrence of such a situation is large at small aspect ratio, as it just requires that there is no contact on the cylindrical part of the inner particle.

Within our assumptions, we explore the space of possible contact directions for one particle, given a local contact number k , and aspect ratio α . We then extract the average effective number of degrees of freedom $\bar{d}_f(\alpha, k)$, which is the average over the contact directions of the minimal value of $\text{rank}(\underline{\underline{N}}_{\hat{\mathbf{t}}_i})$: $\bar{d}_f(\alpha, k) = \langle \min_i(\text{rank}(\underline{\underline{N}}_{\hat{\mathbf{t}}_i})) \rangle_{\{\mathbf{r}_1, \dots, \mathbf{r}_k\}}$, where $\langle \dots \rangle_{\{\mathbf{r}_1, \dots, \mathbf{r}_k\}} = \mathcal{N}^{-1} \int_J \dots d\mathbf{r}_1 \dots d\mathbf{r}_k$ denotes the average over contact directions. This average is limited to a subset J of all possible $\{\mathbf{r}_1 \dots \mathbf{r}_k\}$ such that mechanical equilibrium (equation 14) is possible with positive forces, as expected for a packing of hard particles. This corresponds geometrically to sets $\{\mathbf{r}_1, \dots, \mathbf{r}_k\}$ which do not leave a hemisphere free on the unit sphere. Finally, the normalization $\mathcal{N}$ is the volume of J . For a packing with a coordination number distribution $Q_z(k)$, with average z , the effective $\bar{d}_f$ is: $\bar{d}_f(\alpha) = \sum_k Q_z(k) \bar{d}_f(\alpha, k)$, and the average z follows as $z(\alpha) = 2\bar{d}_f(\alpha)$. In our evaluation, we use a Gaussian distribution for $Q_z(k)$, with variance 1.2 and average z , consistent with simulations⁵⁰. Overall, $z(\alpha)$ is thus the solution of the following self-consistent relation:

$$z(\alpha) = 2 \sum_k Q_z(k) \left\langle \min_i(\text{rank}(\underline{\underline{N}}_{\hat{\mathbf{t}}_i})) \right\rangle_{\{\mathbf{r}_1, \dots, \mathbf{r}_k\}}. \quad (15)$$

The way we look for the orientation $\hat{\mathbf{t}}_i$ on the unit sphere showing the lowest rank is simply by sampling it randomly with a uniform distribution (10^6 samples).

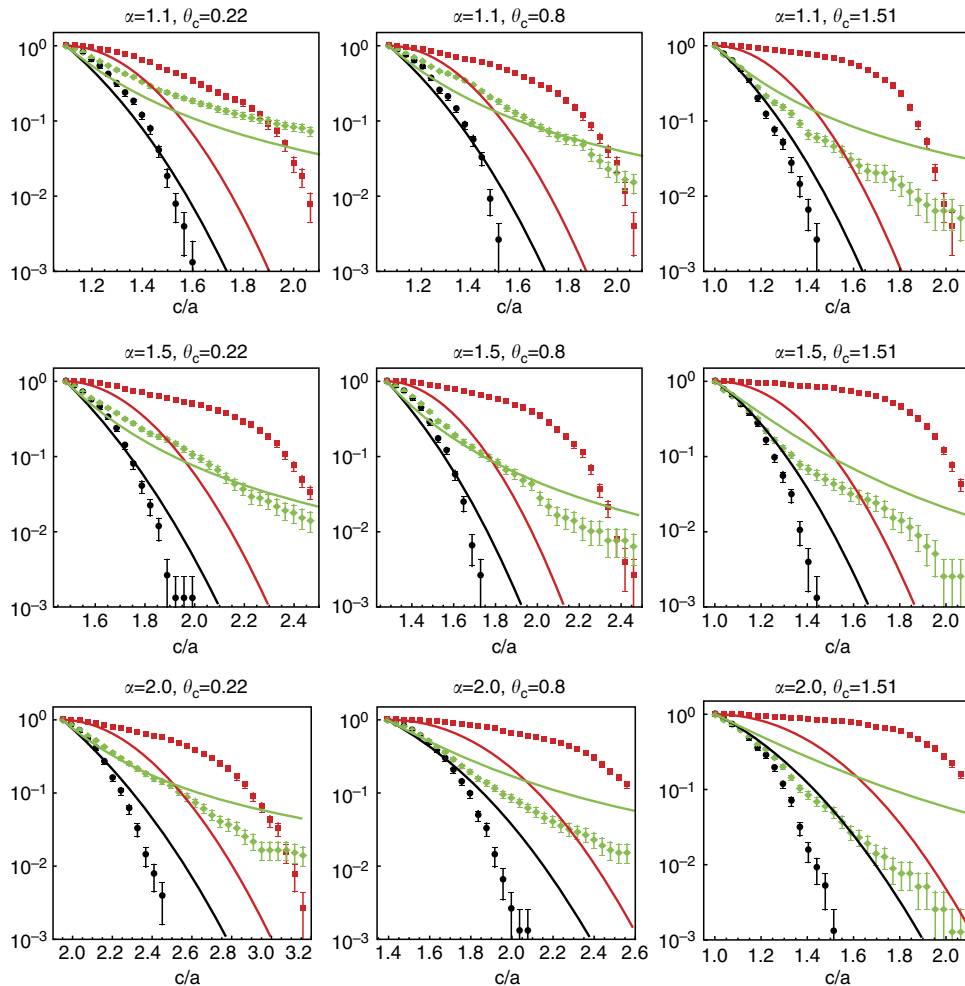


Figure 7 | Comparison of the CDF with simulation data. We plot the theoretical predictions (solid lines) for $P(\mathbf{c}, z)$ (black), $P_B(\mathbf{c})$ (red) and $P_C(\mathbf{c}, z)$ (green) with the corresponding CDFs sampled from simulated configurations (symbols) of spherocylinders. For each aspect ratio $\alpha = 1.1, 1.5, 2.0$, we plot results for three values of the polar angle $\theta_c \in [0, \pi/2]$. We generally observe that the three CDFs agree quite well in the regime of small c values, which provides the dominant contribution to the average Voronoi volume $\bar{W}(z)$. The same plots are shown on a linear scale in the Supplementary Fig. S1. The error bars denote the root mean square error of the finite-size sampling.

The computation of the rank is done via a standard Singular Value Decomposition of $\underline{N}_i$, which is here numerically accurate for $\alpha \geq 1.05$.

Test of the approximations of the theory. We perform a comprehensive test of the different approximations of the theory using computer simulations of spherocylinder packings (Supplementary Note 1). From the generated configurations at the jamming point we obtain the CDF $P(c, z)$, where z is also an observable of the simulation determined by the jamming condition. $P(c, z)$ contains the probability that the boundary of the Voronoi volume in the direction $\hat{c}$ is found at a value larger than c and is determined as follows. We select an orientation $\hat{c}$ relative to the orientation $\hat{t}_i$ of a chosen reference particle i . A large number of particles in the packing contribute a VB along $\hat{c}$ with particle i . We determine all these different VBs denoted by $s(\mathbf{r}_j, \hat{t}_j, \hat{c})$. The boundary of the Voronoi volume in the direction $\hat{c}$ is the minimum c_m of all positive VBs:

$$c_m = \min_{j: s > 0} s(\mathbf{r}_j, \hat{t}_j, \hat{c}), \quad (16)$$

where $\mathbf{r}_j$ and $\hat{t}_j$ are the relative position and orientation of particle j with respect to the reference particle i . Determining this minimal VB for all particles i in the packing yields a list of c_m values for a given $\hat{c}$ (which is always relative to the orientation $\hat{t}_i$). The CDF $P(c, z)$ simply follows by counting the number of values larger than a specified c .

Owing to the rotational symmetry of the spherocylinders, the orientational dependence of $P(c, z)$ is reduced to $P(c, \theta_c; z)$, where θ_c is the polar angle of the orientation $\hat{c}$ in spherical coordinates. Moreover, due to inversion symmetry it is sufficient to select only $\theta_c \in [0, \pi/2]$. Therefore, we choose three θ_c values to cover this range: $\theta_c = 0.22, 0.8, 1.51$. We also use the rotational symmetry to improve the sampling of $P(c, z)$: We fix θ_c to one of the three values, but select a number of azimuthal angles at random. As the packing is statistically isotropic for all azimuthal angles, the resulting c_m value for these directions can all be included in the same ensemble. We consider three different aspect ratios $\alpha = 1.1, 1.5, 2.0$ of the spherocylinders to capture a range of different shapes. The results are plotted in Fig. 7.

We test the two main approximations considered in the theory: (a) The derivation of $P(c, z)$ using a liquid like theory of correlations as done in Song *et al.*²⁰ Jin *et al.*²⁹ leading to the exponential form of equation (8). (b) The factorization of this CDF into contact and bulk contributions as in equation (11). This approximation neglects the correlations between the contacting particles and the bulk. In Fig. 7, we test these approximations by comparing theory and simulations for three different CDFs: $P(c, z)$, $P_B(c)$ and $P_C(c, z)$, equations (8)–(10). In order to determine the $P_B(c)$ from the simulation data we need to take the contact radius $r^*(\hat{r}_j, \hat{t}_j)$ between particle i and any particle j into account. The minimal VB, c_m , is determined from the contributed VBs of particles in the bulk only, that is, particles with $r_j > r^*(\hat{r}_j, \hat{t}_j)$. Likewise, $P_C(c)$ is determined from the simulation data by only considering VBs of contacting particles with $r_j = r^*(\hat{r}_j, \hat{t}_j)$.

Following this procedure, we have tested these approximations with the computer generated packings. We find (Fig. 7): (i) The contact term P_C is well approximated by the theory for the full range of c ; (ii) For small values of c the bulk distribution P_B is well approximated by the theory, and deviations are observed for larger c ; (iii) The full CDF $P(c)$ agrees well between the computer simulations and the theory, especially for small c . The small values of c provide the dominant contribution in the self-consistent equation to calculate the average Voronoi volume equation (4), and therefore to the main quantity of interest, the volume fraction of the packing. This can be seen by rewriting equation (4) as

$$\bar{W}(z) = V_\alpha + \oint \hat{c} \int_{c^*(\hat{c})}^{\infty} dc P(c, \hat{c}; z), \quad (17)$$

as the CDF is trivially unity for c values smaller than the hard-core boundary $c^*(\hat{c})$. The main contribution to the integral then comes from c values close to $c^*(\hat{c})$ due to the decay of the CDF.

Systematic deviations in our approximations arise in the bulk distribution P_B for larger values of c , but, interestingly, the slope of the decay still agrees with our theory. Overall, the comparison highlights the mean-field character of our theory: Correlations are captured well up to about the first coordination shell of particles, after which theory and simulations diverge, especially for the bulk term. The agreement is acceptable for the nearest neighbour-shell, but is incorrect for the second neighbours. Beyond this shell, bulk particles are affected in a finite range by correlations that we do not address, as we assume a uniform distribution of the density of these particles; this is a typical assumption in a mean-field theory. The additional unaccounted correlations lead to a slightly higher probability to observe the VB at intermediate c values in the simulation, compared with our theory. However, these deviations from simulations are small. For instance, Fig. 7 indicates that for a typical value $\alpha = 1.5$ and polar angle $\theta_s = 0.22$, the numerically measured CDF $P(c, z)$ at a relative large value $c/a = 2$ is of the order of 10^{-3} , while the theory predicts this probability at a slightly larger value of $c/a = 2.07$. This small discrepancy is not relevant, as such a value of the probability is negligibly small in the calculation of the volume fraction in equation (4). Thus, because of this small probability to find the VB with values larger than $c/a = 2$, the deviations expected from our approximations are small. These results indicate that, overall, the theory captures the distribution of VBs in the region of small c , which is the relevant region in the calculation of the volume fraction.

The neglected higher-order correlations in the upper coordination shells can only decrease the volume fraction in the calculation leading to smaller packing densities. Following this analysis, we interpret our predicted packing fractions as upper bounds for the empirically found ones, which is indeed observed in Fig. 6b,c.

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Author contributions

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Frequency-dependent attenuation and elasticity in unconsolidated earth materials: effect of damping

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Abstract

We use Discrete Element Methods (DEM) to test and further develop the theory framework for the effective mass of unconsolidated earth materials recently proposed by Valenza and Johnson, Phys. Rev. E **85**, 041302, (2012). First, we validate our DEM framework by comparing the computed effective mass of an unconsolidated granular medium to that determined theoretically. Then, we extract the low-frequency (≤ 15 kHz) normal modes and investigate the corresponding effect of different interparticle damping coefficients. We demonstrate that, as the interparticle damping increases, the normal modes exhibit roughly circular trajectories in the complex frequency plane. The complex valued normal modes characterize the dissipative and elastic properties of the granular material. Thus, our findings may be applied to interpret sonic logging data of near wellbore structures. Our numerical investigation yields two additional observations regarding the effective mass: 1) The individual normal modes are characterized by a unique critical damping parameter, above which the mode is overdamped. 2) The dissipative capacity of the system is largely derived from a few modes with a large imaginary component. Overall, we find that the DEM simulations support the theory of normal modes and effective mass of granular materials. The results are useful for optimizing the attenuation of sound in granular materials.

I. INTRODUCTION

An important challenge in the development of a hydrocarbon reservoir is optimizing well completion and production. After placing a well, sonic well logging techniques characterize the subsurface at high resolution in the immediate vicinity of a borehole. For a given strata, the acoustic properties depend on many characteristics (composition, structure, porosity) and conditions (stress, temperature, pore fluid). In the soft unconsolidated formations of interest in the present study, sonic measurements are further complicated by nonlinear effects arising from heterogeneities, inhomogeneous stress distributions, and dissipation due to fluid flow [2, 11, 14–16, 18, 19, 23, 29, 33, 44, 45, 48]. These nonlinearities have important effects on the dynamics of unconsolidated media as reviewed by [19] and [25]. Therefore, coupled with a representative model of the granular medium [2, 16, 18, 19, 33, 45], sonic wave propagation can be utilized to characterize subsurface conditions and elucidate active mechanisms. In turn, this information can be utilized during well completion and production. Another important effect of non-linearities is to cause dispersion in the system as studied in [19, 23].

Another issue for sonic logging tools is the interference between the signal from the formation and the acoustic energy propagating along the tool body. Wireline tools utilize various methods to attenuate the tool signal. The issue is more challenging for Logging While Drilling (LWD) acoustics [23, 35], because methods for attenuating structure borne sound must not diminish the mechanical integrity of the drill collar, or complicate the drilling process.

Loose granular media are capable of dissipating energy through the network of interparticle contacts and viscous dissipation through the surrounding medium [4, 5, 9, 12, 40]. In fact, it was previously demonstrated that loose grains damp acoustic modes very efficiently [5, 12] and they are routinely used as effective methods to optimize the damping of unwanted structure-borne acoustic signals [20, 41–43].

The motivation of the present study is to develop theoretical and numerical tools to study the acoustic and dissipative properties of unconsolidated granular materials. Our findings can then be employed to a) improve the interpretation of sonic logging measurements, and (b) to optimize granular media for the dissipation of acoustic energy. First, we utilize the Discrete Element Methods (DEM) to test the theory for the effective mass of discrete systems as outlined in [43]. We utilize this DEM and analytical framework to calculate the frequency-dependent effective mass of unconsolidated granular media held in a **cup**, and study the effect of interparticle damping on the

normal modes in the system.

Previous work on the normal modes of unconsolidated granular materials [1, 34, 37, 38, 46, 47] have studied the vibrational density of states of a granular system as the external applied pressure is diminished and the volume fraction of the system decreases towards the point of random close packing (RCP), i.e., when the granular medium is a fragile unconsolidated formation [30, 34]. However, these previous studies have not considered the important effect of attenuation on the normal modes. Attenuation plays a crucial role in governing the dynamic response of real system. Thus, interpretation of any experiment and acoustic logging needs to take into account the dissipative nature of granular matter.

In a previous study we have focused on the stress-dependent dissipative characteristics of the granular medium [21]. The effect of attenuation at the grain-grain contacts has been also studied experimentally and theoretically in [43]. Here, we test the theoretical developments presented in [43] with Discrete Element Methods (DEM) [13], with a particular focus on the low-frequency modes in the effective mass of granular materials. We further develop this theoretical basis for granular systems in order to investigate the dependence of the normal modes on the damping mechanism at the interparticle contact. An area of primary focus is the consequences of assuming the elastic and damping matrices commute.

The paper is organized as follows: Section II introduces the concept of the effective mass. Section III introduces previous analytical results, in particular, the relation between the effective mass and the normal modes in the system. Section IV investigates the trajectories of the normal modes frequencies as a function of damping. Section V outlines our DEM technique which is implemented to test the main implications of the theory in Section VI. Section VII develops the numerical methods to evaluate the trajectories of the normal modes in the frequency domain as a function of damping and the results are discussed in Section VIII. We summarize our findings in Section IX.

II. EFFECTIVE MASS OF A GRANULAR MEDIUM

The effective mass of the granular medium in a cup of mass M_c vibrating with frequency ω is defined as [20]:

$$\tilde{M}(\omega) = \frac{F(\omega)}{a(\omega)} - M_c. \quad (1)$$

For each frequency ω , $F(\omega)$ is determined by measuring the force necessary to displace the cup and, $a(\omega)$ is the associated acceleration. The system is schematically explained in Fig. 1. Experimentally, both quantities are recorded in the frequency domain after waiting a short stabilization period of a few seconds. Then, equation 1 is employed to construct $\tilde{M}(\omega)$. In general, $\tilde{M}(\omega) = M_1(\omega) + iM_2(\omega)$ is complex-valued and reflects the partially in-phase and out-of-phase motion of the individual grains relative to the cup motion. M_1 and M_2 characterize the ensemble averaged elastic and dissipative capacity of the material in a simplified continuum with negligible boundary effects (see for instance [49]). An ideal example is provided by a low viscosity liquid where the dimensions of the cup are much larger than the viscous skin depth across the experimental frequency band; In this case the effective mass resonates at odd-multiples of $1/4$ wavelength of the longitudinal wave in the medium [20]. In fact, the dominant low frequency normal modes in a granular medium exhibit similar scaling behavior. However, the scaling of the modes in a confined granular medium is effected by the preparation protocol, and boundary condition [49].

The imaginary part of the frequency dependent effective mass in a granular medium is roughly proportional to the frequency dependent attenuation in the system. This dissipation can be due to solid contact friction (asperities, material plasticity, etc.) or wetting dynamics from liquid bridges or films between grains. Figure 2 shows the results of a frequency sweep of the real (M_1) and imaginary (M_2) components of the effective mass obtained with DEM for packings under gravity with the indicated damping coefficients (DEM procedure outlined below). These results are qualitatively similar to those determined experimentally, and reported in [20] and [43]. Here, we adapt the DEM technique to obtain the low-frequency modes in the region of interest. In a manner analogous to that achieved experimentally in [43] we use DEM to test the effect of interparticle damping on the dynamics of discrete systems.

III. PREVIOUS THEORETICAL RESULTS

A. Hertz-Mindlin interparticle force law

We consider a granular medium made of spherical particles interacting via Hertz-Mindlin contact forces, Coulomb friction and dissipative forces at the contact point. The normal component of the contact force between any two contacting particles with radius R is the Hertz force [24, 27] defined as:

$$F_n = \frac{2}{3}k_n R^{1/2} x_{ij}^{3/2}, \quad (2)$$

where the normal deformation (1/2 the overlap between the spheres) between the neighboring grains is $x_{ij} = \frac{1}{2}[2R - |\mathbf{x}_i - \mathbf{x}_j|]$, $\mathbf{x}_{i,j}$ are the position vectors, and k_n is the normal spring constant. The latter is defined in terms of the corresponding material properties. The normal elastic constant is

$$k_n = 4G_g/(1 - \nu_g), \quad (3)$$

where G_g is the shear modulus, and ν_g is the Poisson's ratio of the material from which the grains are made.

The tangential Mindlin force between neighboring grains in contact is [36]:

$$\Delta F_t = k_t (R x_{ij})^{1/2} \Delta s, \quad (4)$$

where

$$k_t = 8G_g/(2 - \nu_g), \quad (5)$$

is the tangential spring constant, and the variable s is defined such that the relative shear displacement between the two grain centers is $2s$. Finally, Coulomb friction with interparticle friction coefficient μ imposes

$$F_t \leq \mu F_n, \quad (6)$$

at every contact.

The normal and tangential components of the damping force at the contact point between two particles have been calculated by [6] based on theory by [26] and linear viscoelastic theory by [27]. The dissipative force in the normal direction is:

$$F_{dis}^N = \xi k_n R^{1/2} x_{ij}^{1/2} \dot{x}_{ij}, \quad (7)$$

where ξ is the damping parameter related to the elastic and viscoelastic constant of the material from which the particles are made. From [27], we find:

$$\xi = \frac{1}{2} \frac{(3\eta_2 - \eta_1)^2}{(3\eta_2 + 2\eta_1)} \left[\frac{(1 + \nu_g)(1 - 2\nu_g)}{2G_g} \right], \quad (8)$$

where η_1 and η_2 are the viscous constants of the material of the particles (see Eq. (23) in [6]). The damping parameter ξ has dimension of time. The tangential component of the dissipative force is analogous to the normal component [6]:

$$F_{dis}^T = \xi k_t R^{1/2} x_{ij}^{1/2} \dot{s}. \quad (9)$$

We emphasize that the damping mechanism outlined by Eqs. (7 - 9) is governed by the properties of the materials that makes up the particles. However, this is not the only mechanism for dissipation that may contribute to attenuation in granular materials. For instance, when particles are coated with a viscous fluid, like oil, the dissipation is mainly due to the 'squirt flow' in liquid bridges at the particle contacts [43]. Given the fact that we consider infinitesimal perturbations this mechanism is accounted for in an identical manner as that outlined for viscoelastic deformation of the solid. The result is a variation of the above equations where the dissipative forces are independent of the static deformation at the contact, and are linearly dependent on the relative velocity. For example, set $k_{N,T}R^{(1/2)}x_{(ij)}^{(1/2)} = C$, in Eqs. (7) and (9), where C is a constant.

It could be mentioned that it was shown in [9] that while the above Hertz model qualitatively reproduce the specific stress-dependent viscous damping of granular matter, $(1/Q) P^{(1/3)}$, in terms of the Q-factor, yet, the Herzt model does not capture quantitatively the data due to the grain roughness, which is not captured by the Hertz force law.

Furthermore, frictional dissipation via the interparticle coefficient of friction μ may have implications for nonlinear effects arising from larger perturbations. However, we work in the linear response regime. The strain perturbations are infinitesimally small and they do not produce sliding at the contacts between the particles. Under this approximation, frictional dissipation will not affect the results. However, it will play a role as nonlinear effects and plastic arrangements arise for larger strain perturbations.

We are interested in the linear response of the granular system to infinitesimal perturbations. Therefore we consider the linearized forms of the above equations, ie, we use the Hertz-Mindlin force law linearized about the static value of the normal compression of contacts. The resulting elastic stiffness is the slope of the force law evaluated at x_{ij} (eg. Eqs. (7) and (9)):

$$k^N(x_{ij}) = k_n R^{1/2} x_{ij}^{1/2}, \quad (10)$$

and

$$k^T(x_{ij}) = k_t R^{1/2} x_{ij}^{1/2}. \quad (11)$$

For typical hard particles made of steel, glass or sand, the Poisson's ratio is small. For instance, for sand grains we have $\nu_g = 0.2$. Thus $k_n \simeq 4G_g$ and also $k_t \simeq 4G_g$. Therefore, this leads to $k_n \simeq k_t$ and consequently to

$$k^N(x_{ij}) \simeq k^T(x_{ij}). \quad (12)$$

B. Theory of normal modes

In a typical experiment or simulation, the grains are settled in a cup that moves sinusoidally with magnitude W , taken in the z -direction. We denote $\mathbf{W} = W\hat{\mathbf{z}}$ as the uniaxial displacement of the **cup**. In the analogous physical experiments [20], W is less than $1\mu\text{m}$, at least three orders of magnitude smaller than R . This assures that the external displacement is smaller than the static compression at the grain contacts. Therefore, we take all $\mathbf{u}_i$ to be infinitesimal, and utilize the linear equation of motion for the i -th particle with mass m to describe the system dynamics. The coupled equations of motion for the i -th particle in a system of N particles is written as (see [43] and [21] for details):

$$H_{ij}(\omega)u_j = K_{i\omega}W, \quad i, j = 1 : 6N. \quad (13)$$

The dynamical matrix $\mathbf{H}_{ij}(\omega)$ in the frequency domain can be written as,

$$H_{ij}(\omega) = -m_{ij}\omega^2 - i\omega B_{ij} + K_{ij}. \quad (14)$$

Here the mass matrix $\mathbf{M}$ has elements $m_{ij} = m\delta_{ij}$, with $\mathbf{I}$ the identity matrix if the particles interact via central forces only (frictionless), and it contains the moment of inertia for rotating particles interacting via tangential forces. Each term in H_{ij} accounts for the inertial term, the dissipative matrix $\mathbf{B}$, and elastic matrix $\mathbf{K}$, defined at the contact between particles i and j , respectively. The elements of the elastic matrix K_{ij} are

$$\mathbf{K}_{ij} = k^N(x_{ij})\hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij} + k^T(x_{ij})[\mathbf{I} - \hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij}], \quad (15)$$

where $\hat{\mathbf{d}}_{ij}$ denotes the direction of the normal displacement along the contact point and we use the dyadic notation. The damping matrix B_{ij} is

$$\mathbf{B}_{ij} = \gamma^N(x_{ij})\hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij} + \gamma^T(x_{ij})[\mathbf{I} - \hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij}]. \quad (16)$$

If the particles, i and j , are not in contact ($|\mathbf{x}_j - \mathbf{x}_i| > 2R$), we set both $\mathbf{K}_{ij} = 0$ $\mathbf{B}_{ij} = 0$. The vector $\{u_j\}$ accounts for the set of $3N$ particle displacements and $3N$ particle rotations, and $K_{i\omega}$ is the generalized spring constant connecting a particle to the walls of the cup which moves oscillatory in the z direction with amplitude W .

Inverting the matrix $\mathbf{H}$ we obtain the effective mass as has been shown in our previous work [43]:

$$\tilde{M}(\omega) = m[H^{-1}(\omega)]_{ij}K_{j\omega}. \quad (17)$$

This equation expresses the relation between the effective mass and the normal mode spectrum. The peaks observed in the effective mass (Fig. 2), $\tilde{M}(\omega)$, are due to the set of normal modes, e_j^n , that are a solution to Eq. (13) when there is no forcing by the cup, $W = 0$, i.e.,

$$H_{ij}(\omega_n)e_j^n = 0. \quad (18)$$

Thus, the normal modes, e_j^n , are those eigenvectors of $\mathbf{H}$ for which the corresponding eigenvalue is zero, and they occur at specific complex-valued frequencies, ω_n .

IV. EIGENVALUE TRAJECTORIES AS A FUNCTION OF DAMPING

While previous theoretical studies have considered the undamped modes of frictionless systems [46, 47], the interpretation of the effective mass data from experiments or sonic logging data necessitates a formalism that considers damping and rotational modes that are indigenous in to real granular matter. Our formalism thus generalizes previous results valid for frictionless undamped systems to more realistic granular materials consisting of dissipative interactions with tangential forces.

Equation (17) implies that ω_n is the resonance frequency when $\mathbf{H}(\omega)$ has at least one zero eigenvalue or the determinant of $\mathbf{H}(\omega)$ is equal to zero. When $\xi > 0$, the resonance frequency is a complex number and the imaginary part is relevant to the attenuative properties of the granular system. Here, we study the effect of interparticle damping, ξ , on the resonant modes ω_n .

We are interested in the complete set $\{e_j^n\}$ of complex valued normal modes that satisfy Eq. (18). For these modes, we can rewrite Eq. (18) in the frequency domain as (we follow [21] and utilize some results from [10]):

$$\left(-\mathbf{I}\omega_n^2 - i\omega_n\tilde{\mathbf{B}} + \tilde{\mathbf{K}}\right)\mathbf{q} = 0. \quad (19)$$

Here we have defined: $\mathbf{q} \equiv \mathbf{M}^{-1/2}\mathbf{e}$, $\tilde{\mathbf{B}} \equiv \mathbf{M}^{-1/2}\mathbf{B}\mathbf{M}^{-1/2}$ and $\tilde{\mathbf{K}} \equiv \mathbf{M}^{-1/2}\mathbf{K}\mathbf{M}^{-1/2}$, provided that the $\mathbf{M}$ matrix is positive definite, we can always find $\mathbf{M}^{-1/2}$.

In previous work [21] we have argued that $\tilde{\mathbf{B}}$ and $\tilde{\mathbf{K}}$ are proportional and commute. Indeed, under the approximation Eq. (12) we find that both damping constants, normal $\gamma^N(x_{ij})$ and tangential $\gamma^T(x_{ij})$, are proportional to the respective elastic constants in the elastic counterparts $k^N(x_{ij})$ and $k^T(x_{ij})$, as seen from Eqs. (7) and (9). Therefore,

$$\gamma^N(x_{ij}) = \xi k^N(x_{ij}), \quad (20)$$

and

$$\gamma^T(x_{ij}) = \xi k^T(x_{ij}). \quad (21)$$

We find:

$$\mathbf{B}_{ij} \simeq \xi \mathbf{K}_{ij}. \quad (22)$$

Equation (22) indicates that the matrices are approximately proportional and therefore commute $\mathbf{BK} \simeq \mathbf{KB}$. This result will be exploited next in the characterization of the normal modes [17].

An important implication of Eq. (22) is the set $\{q\}$ are the complete eigenvectors for both $\mathbf{K}$ and $\mathbf{B}$. The normal modes in the damped system are exactly the same as in the undamped case except that they now have complex-valued frequencies, due to the attenuation. Here, we study the effect of the commuting approximation on the behavior of the normal mode frequencies as ξ is varied.

Let $\omega_{n0}^2 > 0$ be the eigenvalues of $\tilde{\mathbf{K}}$ with $\mathbf{q}^n$ the corresponding eigenvectors. Thus, in the absence of attenuation $\tilde{\mathbf{B}} = 0$, $\pm\omega_{n0}$ is the undamped frequency of oscillation of this mode. If the matrices $\tilde{\mathbf{B}}$ and $\tilde{\mathbf{K}}$ exactly commute, then each mode exactly decouples and we can write:

$$\left[-\omega_n^2 - i\omega_n\xi\omega_{n0}^2 + \omega_{n0}^2 \right] \mathbf{q}^n = 0, \quad (23)$$

which is a quadratic equation in ω_n with roots:

$$\omega_n = -i\frac{\xi\omega_{n0}^2}{2} \pm \omega_{n0} \sqrt{1 - \left(\frac{\xi\omega_{n0}}{2}\right)^2}. \quad (24)$$

From this result, it is clear that the modes follow circular trajectories in the complex plane with a radius ω_{n0} . Equation (24) indicates that the position of the normal mode in the complex plane is a function of ξ as long as the damping is smaller than a critical damping given by

$$\xi_c = \frac{2}{\omega_{n0}}. \quad (25)$$

For large enough damping $\xi > \xi_c$, each normal mode becomes overdamped, and the corresponding frequencies are purely imaginary valued [3, 22]. For $\xi < \xi_c$, the modal frequencies are damped oscillators with

$$|\omega_n(\xi)| = \omega_{n0}. \quad (26)$$

That is, the trajectories of the modes in the plane $(\text{Re}[\omega_n(\xi)], \text{Im}[\omega_n(\xi)])$ are exactly circular as a function of ξ . For a fix ξ we find the following. According to Eq. (24), the plot of $\text{Im}[\omega_n]$ versus

$\text{Re}[\omega_n]$ would give a circular shape as long as $\xi < \xi_c$, following the equation:

$$\text{Re}[\omega_n]^2 + \left(\text{Im}[\omega_n] + \frac{1}{\xi}\right)^2 = \frac{1}{\xi^2}. \quad (27)$$

That is, the shape of the plot is a circle centered at $(\text{Re}[\omega_n], \text{Im}[\omega_n]) = (0, -1/\xi)$ and with radius $1/\xi$. If we take the low dissipation limit of Eq. (24), i.e. $\xi \rightarrow 0$ and expand the square root term, we see that the normal modes correspond to the purely elastic system ω_{n0} plus an imaginary part, proportional to the damping parameter:

$$\omega_n \simeq \omega_{n0} - i\frac{\xi\omega_{n0}^2}{2} + O(\xi^2). \quad (28)$$

In the same limit, we observe that the real and imaginary parts of the normal modes follow a parabola in the complex plane for a given small fixed ξ :

$$\text{Im}[\omega_n] \simeq -i\frac{\xi}{2}\text{Re}[\omega_n]^2 + O(\xi^2). \quad (29)$$

It is important to note that Eq. (25) is neither an exact result of the system, nor the result of a trivial dimensional analysis. Equation (25) is not an exact result from Eq. (19) since it involves a series of approximations which are not valid in general. Eq. (25) can only be derived from Eq. (19) when we assume that the damping matrix and the elastic matrix are proportional to each other as stated in Eq. (22). In turn, this approximation is only valid in the limit of small particle Poisson ratio $\nu_g \ll 1$. That is, assuming that the Poisson ratio of the particles is negligible then Eq. (12) is valid and then the matrices are proportional, from where we can derive Eq. (23) from Eq. (19), and then the scaling of Eq. (25) follows.

Then, Eq. (25) is only an approximation, since the Poisson ratio is not exactly zero, even though it is small $\nu_g = 0.2$ compared to one. Thus, the deviations in Fig. 7 from the scaling law arise from the fact that Eq. (25) is not exact, since the Poisson ratio of the particles is not exactly zero, but a small number.

Furthermore, we dimensionalize Eq. (23) and show that Eq. (25) remains a scaling law of the system. Eq. (23) is normalized by the characteristic time given by the constants defining the force-law. That is, $t_0 = R\sqrt{\rho/G}$. Then, normalizing the variables in Eq. (23) by this characteristic time: $\xi' = \xi/t_0$ and $\omega'_{n0} = \omega_{n0}t_0$, then Eq (25) transforms into $\xi'_c = 2/\omega'_{n0}$. Thus the scaling is still valid in this normalized form.

V. DEM SIMULATIONS

The primary motivation of this work is to test the predictions of Eqs. (25), (27) and (29) using the DEM. The discrete simulations consist of a monodisperse granular packing where the grains interact via the contact force laws defined in the previous sections. For all calculations the particles are homogeneous with radius $R = 1\text{mm}$, particle density $\rho = 10^3 \text{ kg m}^{-3}$, shear modulus $G_g = 161 \text{ GPa}$ and Poisson ratio $\nu_g = 0.2$. We prepare a granular packing and impose gravity $g = 9.8\text{m/s}^2$ in the direction perpendicular to the bottom wall. We impose with periodic boundary conditions in both the direction perpendicular and parallel to the surface of the page. Damping at the grain-grain contact is governed by Eqs. (7) and (9) with ξ the damping parameter. We measure the damping parameter in ms and the frequencies in kHz. The packings are composed of N particles and we vary N from small systems of 14 particles up to $N = 400$ to test different aspects of the theory. The small system of 14 particles is used only to test the calculation of the normal modes. Figure 1 shows a typical configuration of the packing under gravity. The preparation protocols used to generate packings are explained in [7, 8, 28–31, 39], where it is demonstrated that our approach generates stable jammed-packings.

VI. TEST OF THE THEORY WITH DEM SIMULATIONS

First we validate our DEM framework by comparing the dynamical effective mass to the analytical prediction of Eq. (17). The latter relates the effective mass of a packing, a dynamical measure, to the inverse of the dynamical matrix H_{ij} , which is given by the static packing structure. The test of equation 17 involves two steps: First we perform a direct dynamical measure of the effective mass by shaking a packing generated by computer simulations at a given frequency. For a given shaking frequency and amplitude $A = 1\mu\text{m}$, which is very small compared to the radii and overlap between particles, we record the equilibrium positions in the packing and shake the bottom wall for a time long enough to measure the force against the bottom wall. According to the time series of the force, we can compute [41] the effective mass corresponding to the frequency. For this calculation we consider a fixed value of damping parameter ξ . Thus, we measure the force and acceleration of the **cup** and extract the effective mass following Eq. (1). We hold the amplitude constant and vary the frequency therefore, the acceleration increases with the frequency.

The real and imaginary part of the DEM simulation are plotted as a function of ω in Fig. 2.

The calculated effective mass exhibits dominant resonance features at similar frequencies as that observed experimentally [20, 21, 41, 43]. Next, we calculate $H^{-1}(\omega)$ by using the static positions of the grains before the shaking in order to employ Eq. (17). Figure 2 shows that the DEM and analytical result (Eq. 17) are in agreement. We think it is important to point out that the analytical prediction (Eq. (17)) accounts for the complex behavior of dissipative granular media. Moreover, we demonstrate that the agreement is consistently good over an order of magnitude in damping parameter.

VII. METHOD TO EVALUATE THE TRAJECTORIES OF THE NORMAL MODES IN THE FREQUENCY SPACE

Given the good agreement demonstrated in Figure 2, we calculate the normal modes of the system to test equations (25), (27) and (29.) By using the static positions of the grains obtained from the packings, we can solve Eq. (18) to extract the normal modes by following the method derived by [32]. In the time domain, Eqs. (13) and (18) take the form:

$$\mathbf{M}\ddot{\mathbf{x}} + \mathbf{B}\dot{\mathbf{x}} + \mathbf{K}\mathbf{x} = 0. \quad (30)$$

If we utilize the state vector $\mathbf{y} = [x_1, \dot{x}_1, x_N, \dot{x}_N]^T$, Eq. (30) can be recast as an ordinary eigenvalue-eigenvector problem:

$$\dot{\mathbf{y}} = \mathbf{A}\mathbf{y}, \quad (31)$$

where

$$\mathbf{A} = \begin{pmatrix} \mathbf{0} & \mathbf{I} \\ -\mathbf{M}^{-1}\mathbf{K} & -\xi\mathbf{M}^{-1}\mathbf{B} \end{pmatrix}. \quad (32)$$

Here, the normal modes frequencies are

$$\omega_n = i\lambda_n, \quad (33)$$

where λ_n is the n-th eigenvalue of matrix $\mathbf{A}$.

VIII. EFFECT OF DAMPING ON THE NORMAL MODES

Since the results of our DEM simulations are consistent with theory and experiment, we proceed to study the effect of the damping parameter, ξ , on the normal mode frequencies. In particular

we investigate the trajectories of the normal modes in the complex plane as we change the damping from undamped to critically damped and beyond. We first calculate the normal modes of the undamped system. For any single trajectory, we calculate the starting point at the undamped frequency ω_{n0} . To obtain the trajectory of this particular normal mode we set an objective function $y = \min[|\lambda_1(\xi + \Delta\xi) - \lambda_1(\xi)|^2, \dots, |\lambda_n(\xi + \Delta\xi) - \lambda_n(\xi)|^2]$, where λ_i are given by Eq. (33). Then, we increase ξ by a small amount to $\xi + \Delta\xi$. Using the 'Eig' function in Matlab, we calculate y for a given damping. Next we employ the multidimensional unconstrained nonlinear minimization method ('Fminsearch' function in Matlab) with a starting point $\omega_n(\xi)$ to determine $\omega_n(\xi + \Delta\xi)$ corresponding to $y = 0$. It is important to note that using the starting point $\omega_n(\xi)$, implies that the solution will be in the vicinity of $\omega_n(\xi)$. This is the case when $\Delta\xi$ is small enough, which allows us to determine the exact trajectory of the normal mode in the complex plane as ξ is varied.

We first test the method of Eq. (32) to calculate the normal modes using a small system of 14 particles, for which the modes can be easily calculated. We compare the modes obtained from Eq. (32) with a direct numerical calculation of the eigenvalues of the dynamical matrix H_{ij} , ie, we calculate all the ω_n which satisfy $\det(\mathbf{H}) = 0$. We use a method derived from expanding the expression for the determinant of $\mathbf{H}$. The desired roots of the resulting polynomial in ω are equal to the eigenvalues of a simple matrix formed from the coefficients of that polynomial. The eigenvalues are calculated with the 'Eig' function in Matlab. A potential problem of this technique appears for large number of particles. Thus, we use this technique to validate the use of Eq. (33) in a small system of 14 particles, and later we will use Eq. (33) to calculate the normal modes for larger systems of 400 particles.

For instance, consider the results of 14 balls interacting via central forces. The number of normal modes is 6×14 and therefore the order of the polynomial to calculate the roots is 84. In our calculations, the different normal mode frequencies, ω_n , vary by a factor of 10 from the highest to lowest. If one were to evaluate the polynomial directly, the term ω^{84} would vary by a factor 10^{84} . This is well beyond the accuracy with which a computer stores numbers. So there may be a problem with the accuracy of the results obtained by a direct numerical evaluation of the normal modes via the eigenvalues of H_{ij} for large numbers of particles.

Figure 3 shows a comparison of both methods to calculate the normal modes as a function of ξ for a system of 14 particles. The system is small enough that we can plot the trajectories of all the 84 normal modes. We see that for this system, Eq. (33) gives the same result as the direct calculation of normal modes via eigenvectors of the dynamical matrix $\mathbf{H}$ (Eq. 18). We conclude

that the alternative method of [32], Eq. (33), is correct and then we proceed to use it for large system sizes.

Figure 4a shows the results of the trajectories of the normal modes as a function of ξ for a system of $N = 400$ particles. The normal modes are calculated via Eq. (33). We start with a specific undamped mode ω_{n0} and then follow its trajectory as a function of ξ in the $(\text{Re}[\omega_n(\xi)], \text{Im}[\omega_n(\xi)])$ -plane. We follow the trajectories of specific undamped modes by increasing the damping parameter ξ in small steps and using the method explained above. Since the system is large, we do not trace the trajectories of all 2400 modes. To indicate the general shape of the mode trajectories we trace that for three modes characterized by undamped frequencies $\omega_{n0} = 6.5, 10.2$ and 15kHz , as indicated by the solid red and black lines in the figure, respectively. In addition, we also plot all the modes for four values of ξ as indicated in the figure.

In general, the theoretical predictions Eqs. (25), (27) and (29) are in good agreement with the results in Figure 4. This plot shows that the modes follow a circular trajectory as we change ξ (the Menorah-shape traced in contiguous black and red lines) as predicted by Eq. (27). When $\xi \geq \xi_c$ the modes lie on the imaginary axis in agreement with the prediction of Eq. (25). For a constant ξ we find the parabolic shape predicted by Eq. (29). The latter prediction is valid for small ξ (light blue curve, $\xi = 0.08$). For greater ξ we find the circular plots predicted by Eq. (27) (see blue and purple curves for $\xi = 0.4$ and 0.8 , respectively). The center of the circles lie on the imaginary axis and they are characterized by a radius $\simeq \xi^{-1}$.

To further test the prediction of circular trajectories, we plot the evolution of the absolute value of selected modes for increasing values of ξ (Figure 4b). We find that the absolute value of the mode is constant, corresponding to the circular trajectory, Eq. (26) for $\xi < \xi_c(\omega_{n0})$, until the critical value of damping unique to the individual modes. When ξ approaches ξ_c , the absolute value of the normal mode frequency is no longer constant and the mode becomes overdamped, decreasing or increasing as a function of ξ along the imaginary axis.

Figure 5 shows the normal modes for a system with a fixed value of $\xi = 0.06$. In this case, the relative residue of each mode is indicated by the size of the data point. The residue is loosely indicative of the contribution of the mode to the effective mass [21, 43]; large residues correspond to modes with large damping contribution to the effective mass. The modes exhibit a parabolic dependence on $\text{Re}[\omega_n]$ consistent with Eq. (29), which is valid for this system characterized by small ξ . However, we also observe that several modes with large residue do not lie on the parabola. As previously noted, the large residue indicates that these outliers make an important contribution

to the effective mass.

Figure 6 shows the imaginary part of the effective mass, M_2 , calculated via DEM, for different values of the damping parameter ξ . As ξ increases the large amplitude modes spread out over a larger frequency band, and modes characterized by a small amplitude, disappear. That is, for small ξ we find a large number of peaks in the effective mass. As ξ increases, more and more peaks disappear; In general, the greater the amplitude the more difficult it is to dampen a mode. We also find a special low frequency mode smaller than 1 kHz with a very large resonance peak indicating that this frequency contributes a lot to the attenuation of the medium. When the damping is increased from $\xi = 8 \times 10^{-4}$ to $\xi = 8 \times 10^{-3}$, the peak almost disappears. Thus, even at small ξ modes that make important contributions to dissipation can be overdamped. **The strong attenuation of the first (low-frequency) peak is observed when increasing the damping parameter, compared to the high-frequency peaks. This effect is a consequence of Eq. (25), which states that the critical damping is inverse proportional to the frequency of the undamped mode. Thus, when the damping parameter increases ξ , then the attenuation of the low frequency modes appears first.**

Finally, we test the scaling of the critical damping ξ_c as a function of the frequency of the undamped mode ω_{n0} , as suggested by Eq. (25). Figure 7 shows good agreement between the numerical estimation of $\xi_c(\omega_{n0})$ and ω_{n0} , confirming the inverse scaling law of Eq. (25). This scaling law indicates that lower frequency modes are more resistant to becoming overdamped. Again, some high frequency modes do not obey the scaling behavior Eq. (25). These modes have a small ξ_c and therefore can be easily dampened. This insight is useful for optimizing granular media to attenuate structure borne sound.

IX. CONCLUSIONS

In summary, we explored the effect of interparticle damping on the normal modes in a granular medium. We find good agreement between the DEM simulations and the analytical predictions. As the interparticle damping is varied, the normal modes trajectories in the complex plane are roughly circular. This is a consequence of commutative properties of the damping and elastic matrices that govern the interparticle dynamics. It is plausible that the same commuting properties are realized in experimental systems. For the overwhelming majority of the normal modes, the corresponding critical damping obeys the inverse scaling law $\xi_c \propto \omega_{n0}^{-1}$.

A surprising observation is that there are some very special normal modes that do not obey this scaling law. The corresponding frequencies are easy to identify in trajectory plots and are characterized by a small critical damping parameter. These outliers make large contributions to dissipation of acoustic energy in the granular medium. Thus, this observation may have potential for application to dampen the tool signal in downhole sonic logging.

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FIG. 1. Schematic view of the simulation box and particles. The simulations consider periodic boundary conditions. The walls are made of fixed particles of the same features of the particles in the bulk. The figure shows a system prepared after the relaxation under gravity has finished. The walls are shaken during the dynamical calculation of the effective mass with an acceleration $a(\omega)$ and the force $F(\omega)$ is measured at the bottom of the cup to obtain the effective mass via Eq. (1).

FIG. 2. Comparison of theoretical estimation of the effective mass via Eq. (17) and the direct dynamical measurements using the numerically generated packings with DEM by shaking the packing at a given frequency ω . DEM results are shown by the symbols for two damping parameters $\xi = 0.08\text{ms}$ and $\xi = 0.8\text{ms}$ and separately for the real and imaginary part of the effective mass $\tilde{M}(\omega)$, as indicated. The DEM shaking simulation results are obtained from the frequency sweep with amplitude $A = 1\mu\text{m}$. The theory lines correspond to the solution of the right hand side of Eq. (17): $m[H^{-1}(\omega)]_{ij}K_{j\omega}$, where H^{-1} is calculated as discussed in the text. We observe a very good agreement between theory and DEM simulations validating the theoretical results.

FIG. 3. Trajectories of all the 84 resonance frequencies for a small packing of 14 particles. This plot shows the comparison between the trajectories of $\omega_n(\xi)$ calculated by the Meirovitch method of Eq. (32) (circles) with the direct determination of the determinant of the dynamical matrix $\mathbf{H}$ (line trajectories). We confirm that both methods gives the same result for the normal frequencies. This result indicates that Eq. (32) is an efficient way to calculate the normal modes, and it is used then in the calculations for larger system sizes in the rest of the paper. The trajectories follow the Menorah-like shapes which are further investigated in Fig. 4 for a larger system.

FIG. 4. (a) Menorah-like circular trajectories of the normal modes in the complex plane obtained for different modes with increasing values of ξ for a system of $N = 400$ particles. This result confirms the prediction of circular trajectories (red and black curves for three selected modes) as a function of ξ Eq. (26), for damping below the critical value ξ_c . The modes then become overdamped when $\xi > \xi_c$ and the frequency of the normal modes become purely imaginary, as shown in the figure. The normal modes are calculated via the solution of Eq. (32). We also plot the modes for fixed ξ as indicated. For small ξ the plot follows a parabola (green curve) as predicted by Eq. (29). For large damping (blue and purple plots) the shape is a circle following Eq. (27). (b) Mode amplitude for increasing values of ξ shown for a few selected modes obtained from (a). The constant value as a function of ξ corresponds to the circular trajectories in the complex plane which end at ξ_c where the mode becomes imaginary and decreases or increases as a function of ξ .

FIG. 5. Distribution of normal modes in the complex plane for a given fixed small damping

$\xi = 0.06\text{ms}$ for $N = 400$. The size of the symbol is proportional to the resonance mode residue. The shape of the curve confirms the parabolic prediction of Eq. (29) in average valid when the damping is small. However, we also find that some modes with large residues deviate considerably from the parabolic approximation. These modes are characterized by large residues and therefore contribute with large damping to the effective mass. They can be used to damp unwanted modes in the system.

FIG. 6. Imaginary effective mass calculated via the direct dynamical shaking of the medium for increasing values of ξ for $N = 400$. The plot shows the changes to $M_2(\omega)$ with ξ , as indicated. Normal modes with important contribution disappears with the increase of damping. When the damping is small, there are large number of peaks in the effective mass. With increasing damping the peaks gradually disappear. We find that the larger the peaks the more difficult to be damped. We also find a special low frequency mode less than 1 kHz with a very large resonance peak indicating that this frequency contributes much to the attenuation of the medium. When the damping is increased from $\xi = 8 \times 10^{-4}$ to $\xi = 8 \times 10^{-3}$, the peak almost disappears.

FIG. 7. Critical damping ξ_c as function of the normal mode frequency ω_{n0} of the undamped mode for $N = 400$. The theoretical prediction $\xi_c \propto \omega_{n0}^{-1}$ in Eq. (25) is in satisfactory agreement with the numerical results. However, we still find some modes that deviates from the inverse scaling law. These modes are indeed very easy to damp since they have very low ξ_c , which may lead to applications in damping acoustic noise in geomechanical tools.

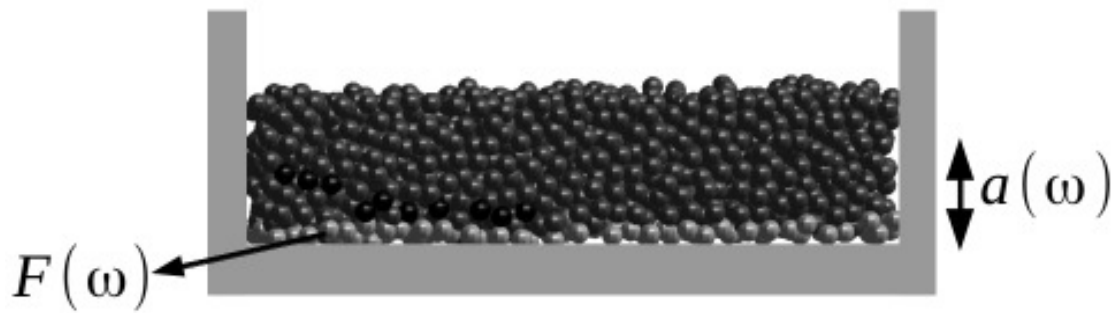


FIG. 1:

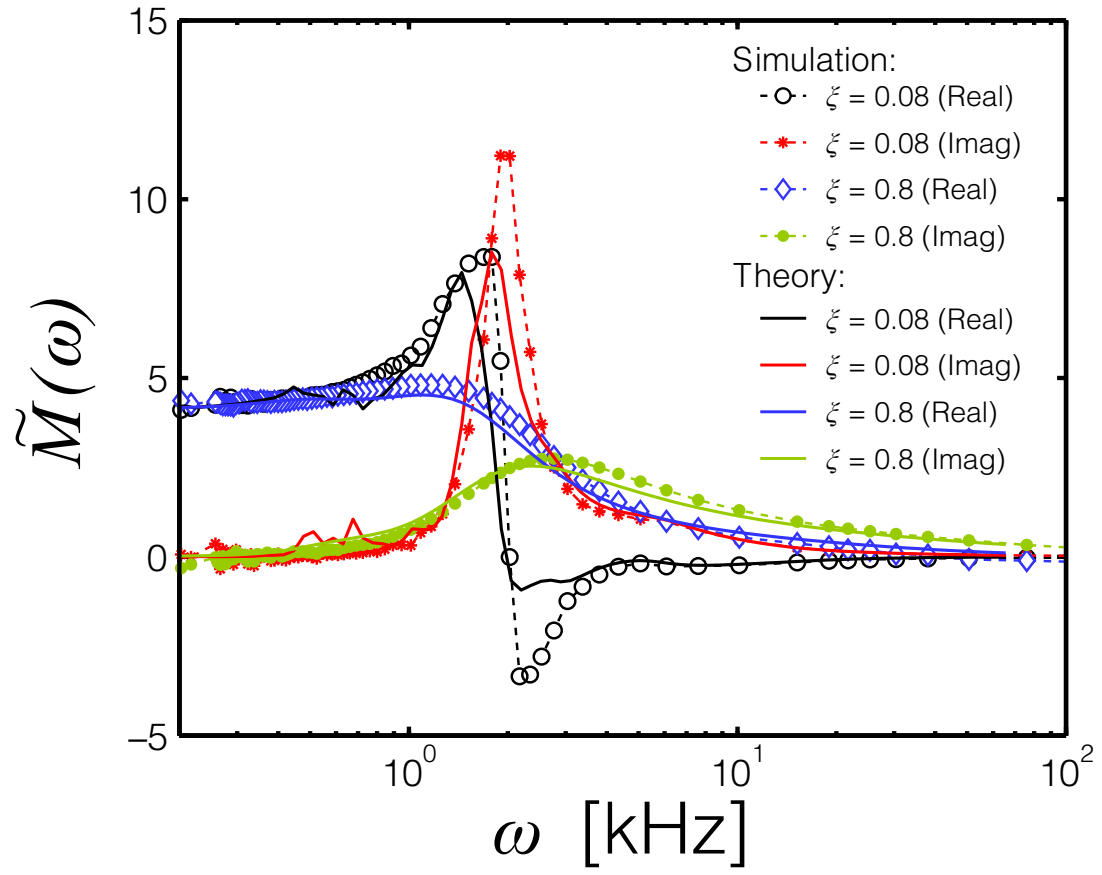


FIG. 2:

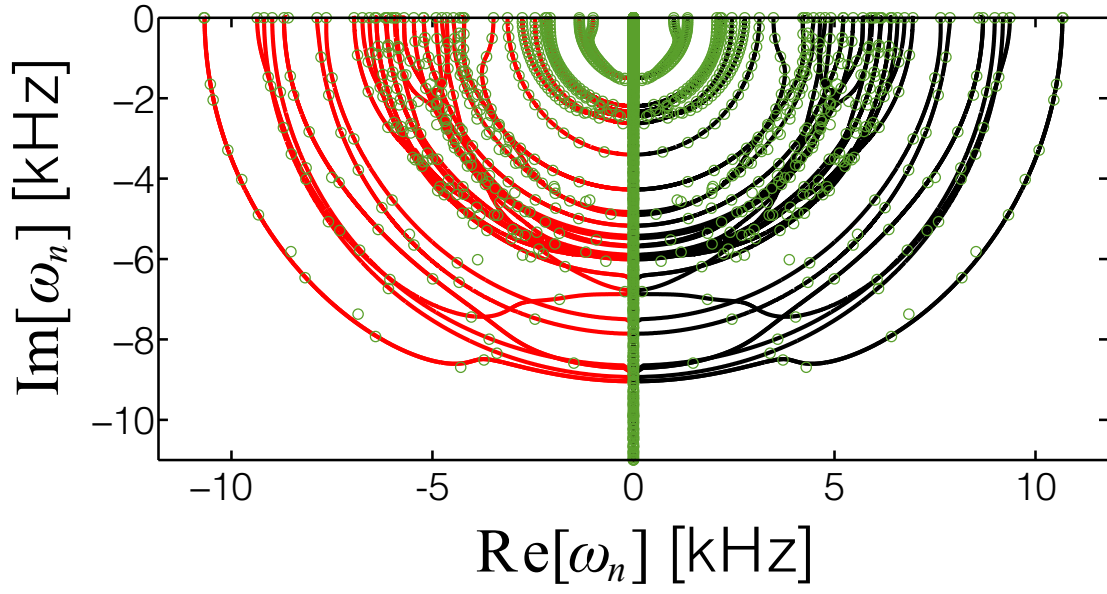


FIG. 3:

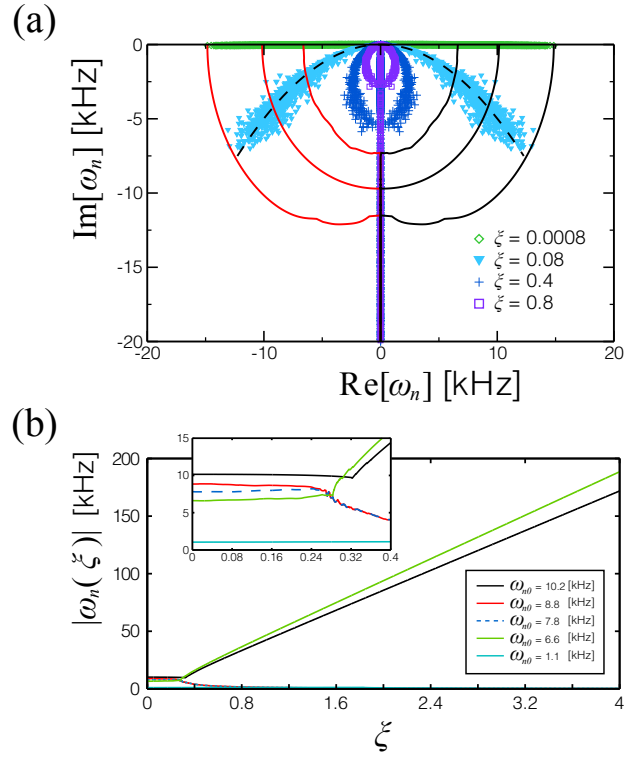


FIG. 4:

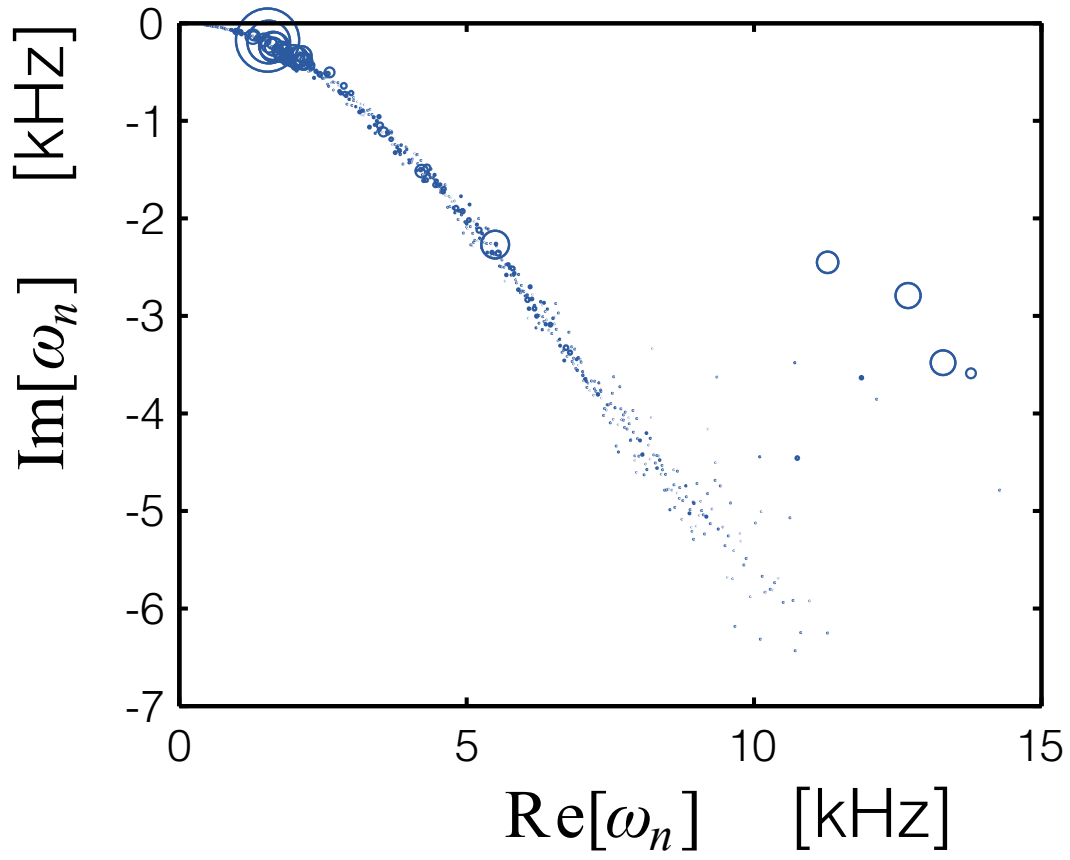


FIG. 5:

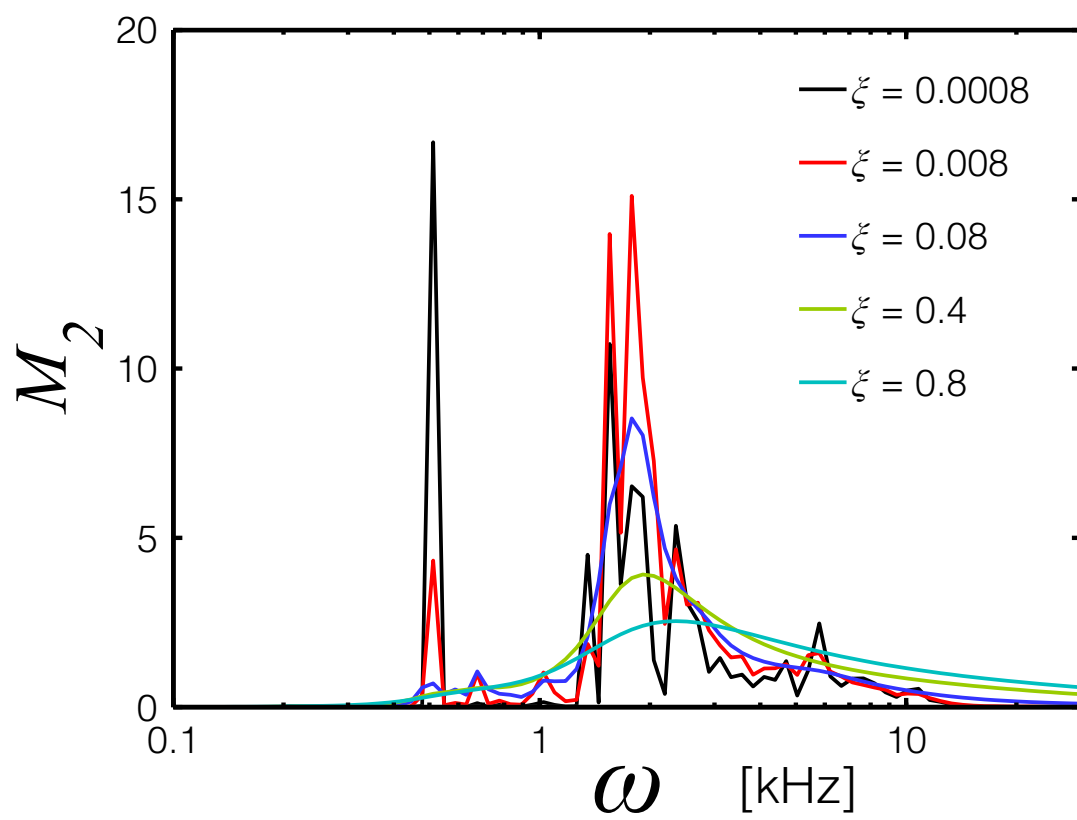


FIG. 6:

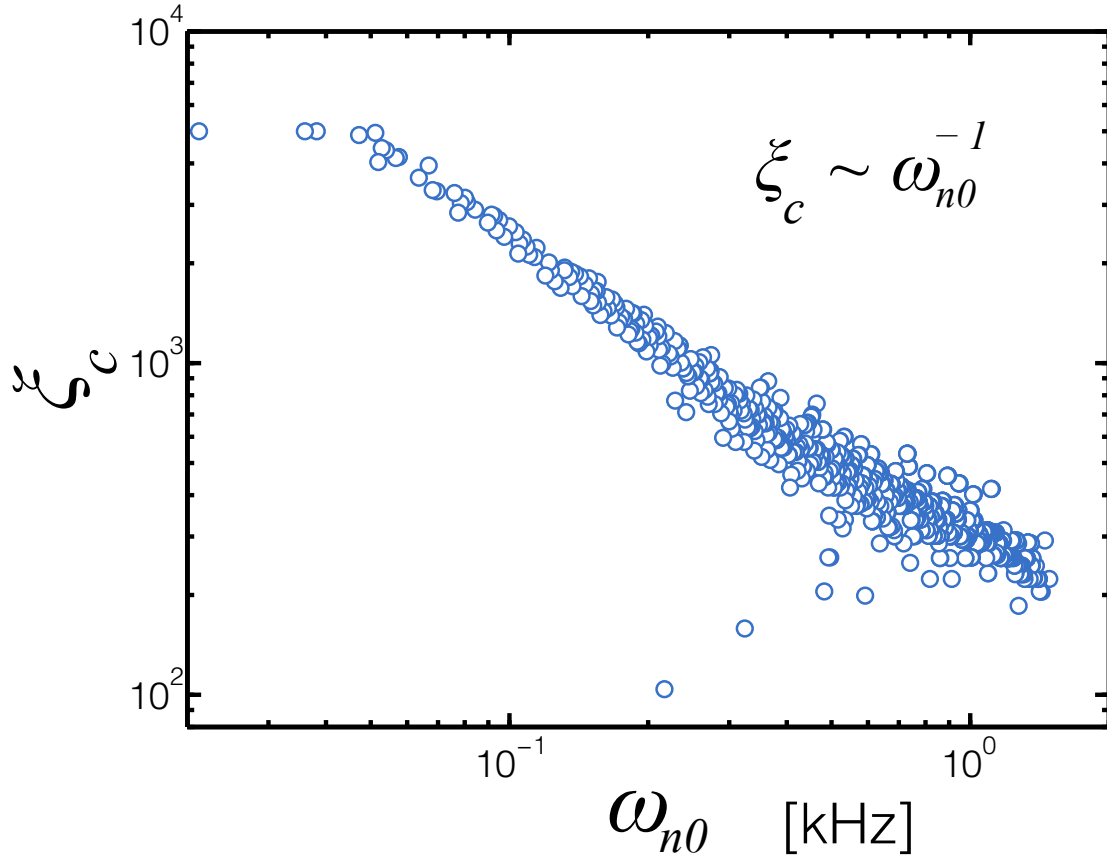
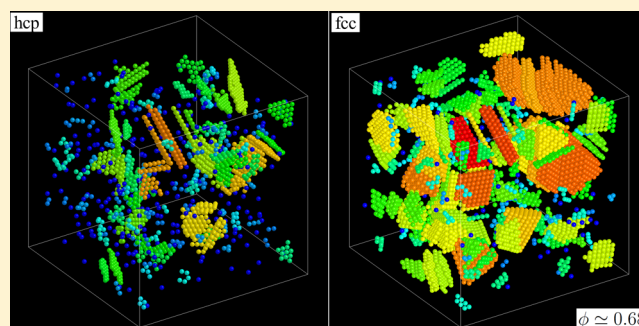


FIG. 7:

Structural Properties of Dense Hard Sphere Packings

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ABSTRACT: We numerically study structural properties of mechanically stable packings of hard spheres (HS), in a wide range of packing fractions $0.53 \leq \phi \leq 0.72$. Detailed structural information is obtained from the analysis of orientational order parameters, which clearly reveals a disorder–order phase transition at the random close packing (RCP) density, $\phi_c \simeq 0.64$. Above ϕ_c , the crystalline nuclei form 3D-like clusters, which upon further densification transform into alternating planar-like layers. We also find that particles with icosahedral symmetry survive only in a narrow density range in the vicinity of the RCP transition.



INTRODUCTION

As a fundamental model in condensed matter physics and material science, packings of hard spheres (HS) reproduce many essential structural properties of glassy and granular media.^{1–3} Structural changes of the HS system have been observed when a packing is densified above the density of random close packing (RCP⁴), $\phi_c \simeq 0.64$ (see, e.g., refs 5 and 6). A new order parameter, which is based on the cumulative distribution of the rotational invariant w_6 , was proposed in ref 5 to identify these structural changes. Random arrangement is transferred to partial crystallization across RCP, and two lattice types, the face centered cubic (fcc) and the hexagonal close-packed (hcp), are observed in the crystalline clusters. However, more detailed information has not been obtained due to insufficient system size. In this study, we aim to explore more detailed structural changes of the transition at RCP in comparison with previous studies,^{5,6} based on larger scale simulations.

We numerically generate a large set of HS packings composed of $N = 64 \times 10^3$ monodispersed spheres located in the cubic box with periodic boundary conditions, using the modified Lubachevsky–Stillinger (LS) algorithm.⁷

RESULTS

To examine the structural properties of jammed spheres, we first study their spatial correlations. Figure 1 shows the radial distribution function $g(r)$ of hard spheres for several different packing fractions in the range $[0.60, 0.68]$. The cumulative function $Z(r) \equiv 4\pi\rho \int_0^r r'^2 g(r') dr'$, which is the mean number of particles inside a sphere of radius r , is also plotted in Figure 1. When $\phi \leq \phi_c$, addition to the strong peaks at the contact distances $r = 1, 2, \dots$ (we set unit diameter), a weak secondary peak at $r = (3)^{1/2}$ is visible. This peak corresponds to a contact angle $\theta = 2\pi/3$ but not necessarily a lattice structure.⁸ Above ϕ_c ,

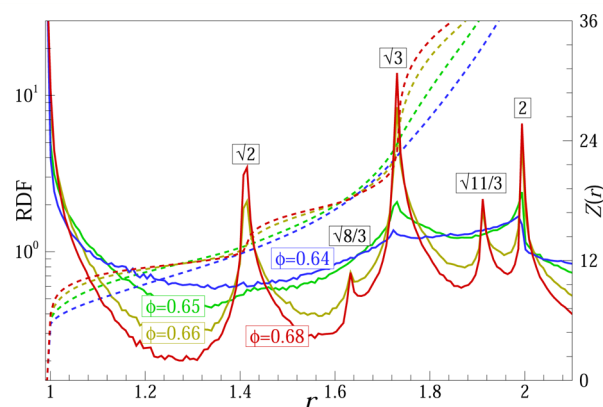


Figure 1. Radial distribution function $g(r)$ of hard spheres (solid lines) and its cumulative function $Z(r)$ (dashed lines) at different packing fractions ϕ . The values of ϕ , and the positions of peaks, are indicated on the plot.

additional peaks appear at $(2)^{1/2}$, $(8/3)^{1/2}$, and $(11/3)^{1/2}$, which are typical distances in the fcc and hcp lattices. The observation indicates the onset of crystallization when the packings are densified above ϕ_c .

More detailed structural information can be obtained from the well-known bond order parameter method,⁹ which is widely used to study the structural properties of condensed matter,^{9–11} hard spheres,^{12–19} Lennard-Jones systems,^{20–24} complex plasmas,^{25–34} colloidal and patchy systems,^{35–37} granular media,³⁸ metallic glasses,³⁹ repulsive shoulder systems,⁴⁰ etc. Each particle

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i is connected via vectors (bonds) with its $N_{\text{nn}}(i)$ nearest neighbors (NN), and the rotational invariants (RIs) of rank l of second $q_l(i)$ and third $w_l(i)$ orders are calculated as

$$q_l(i) = \left(\frac{4\pi}{(2l+1)} \sum_{m=-l}^l |q_{lm}(i)|^2 \right)^{1/2} \quad (1)$$

$$w_l(i) = \sum_{\substack{m_1, m_2, m_3 \\ m_1 + m_2 + m_3 = 0}} \begin{bmatrix} l & l & l \\ m_1 & m_2 & m_3 \end{bmatrix} q_{lm_1}(i) q_{lm_2}(i) q_{lm_3}(i) \quad (2)$$

where $q_{lm}(i) = N_{\text{nn}}(i)^{-1} \sum_{j=1}^{N_{\text{nn}}(i)} Y_{lm}(\mathbf{r}_{ij})$, Y_{lm} are the spherical harmonics, and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ are vectors connecting centers of particles i and j . In eq 2, $\begin{bmatrix} l & l & l \\ m_1 & m_2 & m_3 \end{bmatrix}$ are the Wigner 3j-symbols, and the summations performed over all the indexes $m_i = -l, \dots, l$ satisfying the condition $m_1 + m_2 + m_3 = 0$. As shown in the pioneer paper,⁹ the bond order parameters q_l and w_l can be used as measures, to characterize the local orientational order and the phase state of considered systems. Because each lattice type has a unique set of bond order parameters, the method of RIs can also be used to identify lattice structures in mixed systems. The values of q_l and w_l for a few common lattice types (including the liquid-like HS state) are presented in Table 1. Some of the values were

Table 1. Bond Order Parameters q_l and w_l ($l = 4, 6$) for Several Typical Lattices and Liquid-Like HS Random Packings (rp) at $\phi = 0.55$, Calculated via the Fixed Number of Nearest Neighbors (NN)

lattice type	q_4	q_6	w_4	w_6
hcp (12 NN)	0.097	0.485	0.134	−0.012
fcc (12 NN)	0.19	0.575	−0.159	−0.013
ico (12 NN)	1.4×10^{-4}	0.663	−0.159	−0.169
bcc (8 NN)	0.5	0.628	−0.159	0.013
bcc (14 NN)	0.036	0.51	0.159	0.013
rp at $\phi = 0.55$ (12 NN)	≈0.16	≈0.34	≈−0.019	≈−0.032

reported previously (see, e.g., refs 9 and 41–44). From eq 2, it is easy to see that $w_l \propto q_l^3$, and therefore, we expect w_l 's to be more sensitive measures compared to q_l 's.

In three dimensions (3D), the densest possible packings of identical hard spheres are the fcc and hcp lattices (with the densest packing fraction (pf) $\phi_{\text{fcc}} = \phi_{\text{hcp}} = (2)^{1/2}\pi/6 \simeq 0.74$). Dense HS systems may also include particles having the icosahedral (ico) type of symmetry. Icosahedral, fcc, and hcp spheres have 12 nearest neighbors (the spheres located in the first coordination shell).

To identify lattice-like particles, we calculate the bond order parameters q_l and w_l ($l = 4, 6$) for each particle with a fixed number of the nearest neighbors $N_{\text{nn}} = 12$.⁴⁵ A particle is called fcc-like (hcp-like, ico-like) if its coordinates in the four-dimensional space (q_4, q_6, w_4, w_6) are sufficiently close to those of the perfect fcc (hcp, ico) lattice type. An amorphous (liquid-like) particle is identified if its order parameters are sufficiently small, for example, $q_6^{\text{liq}} \simeq N_{\text{nn}}^{-1/2} \simeq 0.29 \ll q_6^{\text{fcc/hcp/ico}}$. Note that, by varying the NN number and rank l of parameters w_l and q_l , in principle, it is possible to find any lattice type. For instance, the first and second (next nearest neighbors) shells of body centered cubic (bcc) lattices correspond to $N_{\text{nn}} = 8$ and 14, respectively.

To quantify the local orientational order, it is convenient to use the probability distribution functions (PDFs) $P(q_l)$ and $P(w_l)$.

Figure 2 shows how the PDFs vary at different l ($l = 4, 6$) and ϕ (covering both amorphous and solid-like states). Densification of

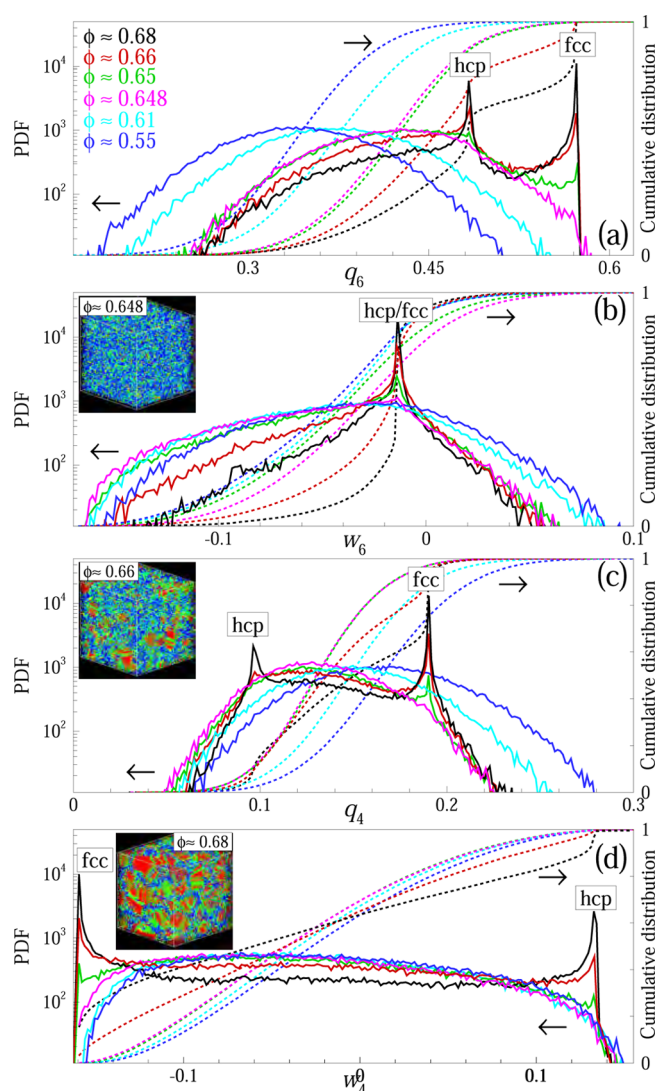


Figure 2. Probability distribution functions (PDFs) of the rotational invariants q_l (a and c) and w_l (b and d) at various packing fractions ϕ . Values of the bond order parameters for ideal lattice types (fcc and hcp) are also indicated. Cumulative distributions (normalized to unity) of these PDFs are shown by dashed lines, which are much less noisy than PDFs. By using these distributions, it is easy to estimate the abundance of crystalline and liquid-like particles. Insets (from top to bottom) show spatial distributions of $N = 64 \times 10^3$ HS in the cubic box at different packing fractions of $\phi \simeq 0.65, 0.66$, and 0.68 , respectively. Hard spheres are color-coded by their q_6 values, in order to visualize liquid-like (blue color) and crystalline hcp-like (green color) and fcc-like (red color) structures.

the HS, as clearly seen in Figure 2, results in appearance of the crystalline fcc-like and hcp-like spheres in the vicinity of the RCP. Corresponding cumulative distributions C_q^l and C_w^l (shown by dashed lines of the same color) are also plotted in Figure 2. For instance, the cumulative function C_q^l associated with the $P(q_l)$ is defined as

$$C_q^l(x) \equiv \int_{-\infty}^x P(q_l) dq_l \quad (3)$$

Evidently, $C_q^l(x)$ is the abundance of particles, having values of $q_i < x$ and $C_q^l(\infty) = 1$. Figure 3 shows two-dimensional PDFs—

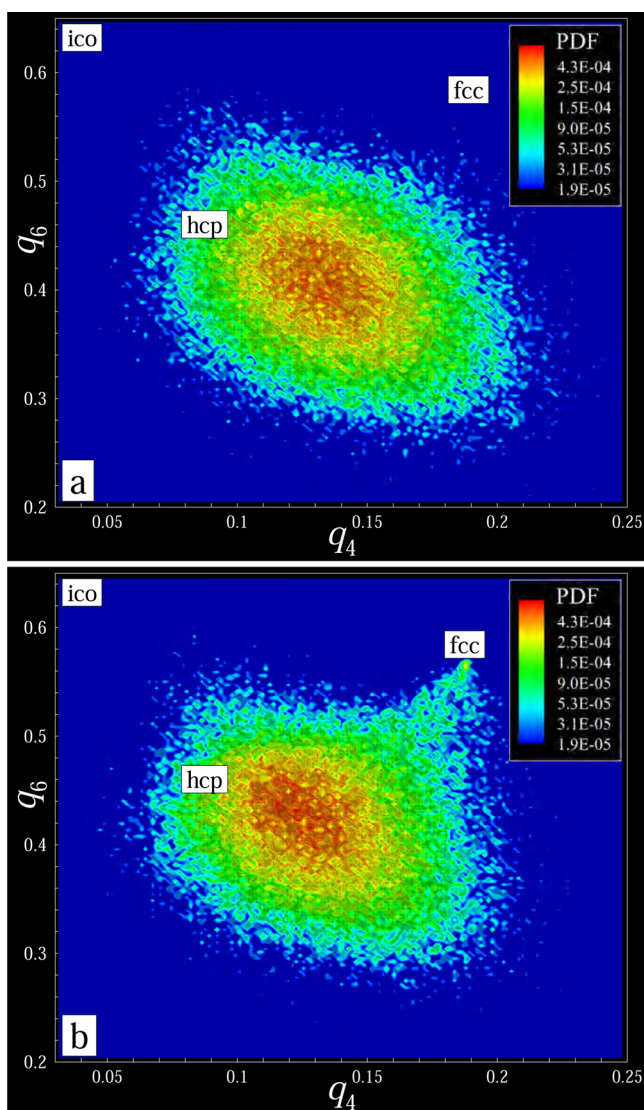


Figure 3. Probability distributions on the order parameter plane of q_4 – q_6 in the vicinity of the RCP at $\phi = 0.64$ (a) and $\phi = 0.65$ (b). Loci of the perfect fcc, hcp, and ico are also indicated on the plot. An fcc peak appears at $\phi = 0.65$. Note that the hcp lattice may have a large overlap with the liquid-like structure in this plot.

probability distributions on the plane q_4 – q_6 —in the vicinity of the RCP at $\phi \simeq 0.64$ and $\phi \simeq 0.65$. At $\phi \simeq 0.64$, structurally HS is amorphous, while at $\phi \simeq 0.65$ the formation of crystalline fcc-like spheres is clearly seen. The result suggests a disordered–ordered transition in the density range $[0.64, 0.65]$, consistent with previous studies.^{5,6,16}

The global order parameters, i.e., the cumulant W_6^c (defined as $C_w^l(W_6^c) = 1/2$) and the mean value of q_6 (defined from the global averaging $\langle q_6 \rangle = (1/N) \sum_{i=1}^N q_6(i)$), are shown in Figure 4. No significant finite size effects are observed between $N = 10^4$ and $N = 64 \times 10^3$ systems. For the purpose of detecting crystallization onset, we find W_6^c to be a more sensitive measure than $\langle q_6 \rangle$. This is because (1) liquid-like and lattice-like spheres have quite different values of w_6 and (2) w_6 is nearly the same for perfect fcc and hcp lattices (see Table 1). Because of this reason, the measure W_6^c has been recently used to describe the melting and

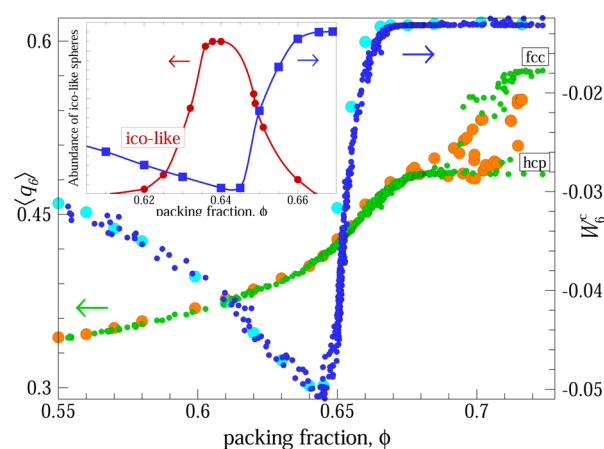


Figure 4. Cumulant W_6^c (blue and cyan circles) and $\langle q_6 \rangle$ (green and orange circles) versus packing fraction ϕ . Blue and green circles correspond to the HS system with $N = 10^4$ spheres; cyan and orange circles correspond to $N = 64 \times 10^3$ spheres. The minimum of W_6^c can be used to locate the RCP transition, $\phi_c \simeq 0.645$. The explosive-like growth of W_6^c above ϕ_c is a clear indicator of the appearance of ordered, crystalline-like (hcp-like and/or fcc-like) spheres. The growth of the $\langle q_6 \rangle$ value near ϕ_c is rather monotonous and relatively slow. The possibility for the dense HS system to be packed via different abundance of fcc-like and hcp-like particles is the key reason behind the fluctuation of $\langle q_6 \rangle$ when $\phi \gtrsim 0.68$. The inset shows the number of ico-like spheres versus ϕ : ico-like spheres survive only in the narrow range of parameters ϕ near the RCP transition.

freezing phase transitions of Lennard-Jones^{23,24} and Yukawa systems.^{32–34} On the other side, the density dependence of $\langle q_6 \rangle$ also reveals fascinating properties. The relatively slow increase of $\langle q_6 \rangle$ at $\phi \lesssim \phi_c$ can be attributed to single hcp-like particles,⁵ while at $\phi > \phi_c$ the bond order parameter $\langle q_6 \rangle$ increases more rapidly due to the emergence of crystalline clusters. Further densification of the HS leads to strong fluctuations (“oscillations”) of the global parameter $\langle q_6 \rangle$, which start at $\phi \simeq 0.68$ and suggest another transition.^{5,6} This oscillatory behavior is due to the fact that the hcp and fcc crystalline structures are characterized by quite different values of the bond order parameter q_6 (see Table 1), and at a given ϕ , dense partially crystallized HS packings can be realized with quite different abundances of hcp-like and fcc-like spheres. Another important parameter characterizing the local structure is the abundance N_{ico} of spheres having icosahedral-type (5-fold) symmetry (see Figure 4, inset). Here, ico-like spheres are defined as spheres having $q_6 \geq 0.61$. The density dependence of N_{ico} shows that ico-like spheres can exist only in a narrow range of pf, and its abundance maximizes at ϕ_c .

The above analysis provides detailed information about the local and global orders. Next, we study the abundance of crystalline particles and their spatial arrangement. We use the standard friends-of-friends algorithm⁴⁶ to find crystalline clusters. Figure 5 presents the spatial distribution and the shape of crystalline clusters composed of hcp- and fcc-like spheres, at several different ϕ . When $\phi < 0.65$ (Figure 5a,b), the crystalline particles are mostly isolated; increase of packing fraction ϕ leads to an appearance of several 3D (containing tens of spheres, both hcp- and fcc-type) clusters (Figure 5c,d at $\phi \approx 0.66$). Upon further densification, these 3D clusters transform into 2D layers spanning system-wide (see Figure 5e,f at $\phi \approx 0.68$).⁵

The abundances of hcp-like (n_{hcp}) and fcc-like (n_{fcc}) spheres are shown in Figure 6 as functions of packing fraction ϕ . At $\phi \leq$

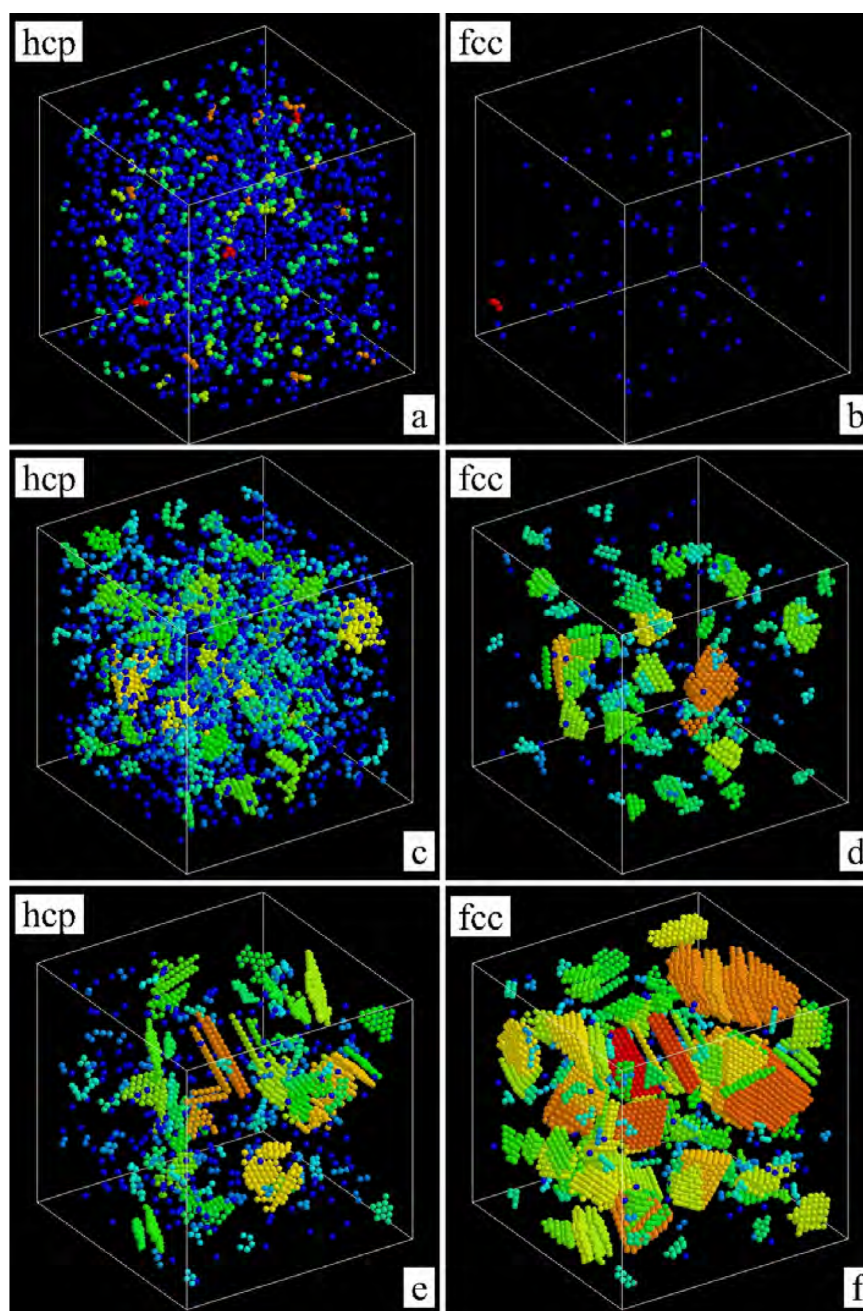


Figure 5. Distributions of hcp and fcc clusters over the space at different ϕ values: $\phi \simeq 0.65$ (a, b), $\phi \simeq 0.66$ (c, d), and $\phi \simeq 0.68$ (e, f). Particles are color-coded by the mass of the cluster (in the unit of the mass of a single sphere). At $\phi \simeq 0.65$, single hcp-like spheres dominate. At $\phi \simeq 0.65$, three-dimensional local clusters of both hcp- and fcc-like particles are observed. Further densification of HS results in a structural transition: the 3D local clusters transform into global and planar layers (e, f).

ϕ_c , the relatively slow increase of $n_{\text{tot}} = n_{\text{hcp}} + n_{\text{fcc}}$ is mainly due to the emergence of randomly distributed single hcp-like spheres. Around $\phi \simeq \phi_c$, the slopes of $n_{\text{hcp}}(\phi)$ and $n_{\text{fcc}}(\phi)$ increase significantly. At $\phi \simeq 0.66$, the abundance of the fcc-like spheres is equal to that of the hcp-like spheres ($n_{\text{fcc}} \approx n_{\text{hcp}}$). Additionally, the abundance of spheres having number of contacts $N_c = 12$ is plotted in Figure 6, as one more sensitive indicator of the crystallization of a dense HS. The relative cumulative spectrum of cluster mass is plotted in the inset of Figure 6, showing that this distribution becomes wider as the density increases.

DISCUSSION AND CONCLUSION

The above results illustrate a sharp first-order-like transition⁶ at ϕ_c between disordered (frictional) packings and partially ordered packings. Amorphous frictionless packings only exist at a single density ϕ_c : below ϕ_c the packings are unstable unless they are frictional (and the frictional packings are disordered);⁶ above ϕ_c the packings are partially ordered. On the other hand, multiple amorphous frictionless states have been found in other simulations,^{47–50} which supports the mean-field theoretical prediction that amorphous jammed packings should exist in the interval (so-called J-line) $\phi \in [\phi_{\text{th}}, \phi_{\text{GCP}}]$.¹ We stress that this apparent contrast is due to the presence of crystallization: If the nucleation rate is slow enough, such as in polydisperse⁴⁷ or large

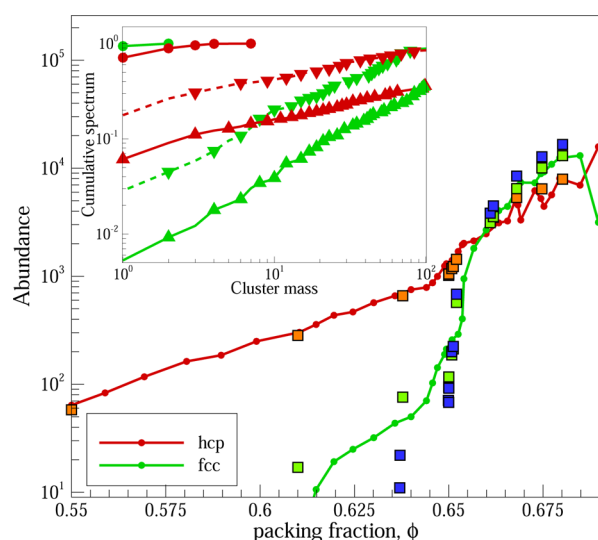


Figure 6. Abundance of hcp-like (red line points) and fcc-like (green line points) spheres versus the packing fraction ϕ , with $N = 10^4$. Results of larger scale simulations with $N = 64 \times 10^3$ spheres are shown by squares (orange and green symbols correspond to hcp- and fcc-like spheres). Additionally, the abundance of spheres having number of contacts $N_c = 12$ is plotted by blue squares. The inset shows the relative cumulative spectrum of hcp (fcc)-like (red and green lines, respectively) clusters versus the cluster mass at different ϕ values: $\phi \approx 0.68$ (solid, triangles), $\phi \approx 0.66$ (dashed, gradients), and $\phi \approx 0.65$ (solid, circles).

dimensional systems,⁴⁸ then crystallization is suppressed and dense amorphous packings can be obtained by slow compressions; while in 3D monodisperse systems, the nucleation process is so fast such that any slow compression would result in partial crystallization. Combined with the mean-field prediction, our results suggest two possible scenarios for 3D monodisperse packings: (i) $\phi_c < \phi_{th}$, therefore, dense amorphous packings $\phi > \phi_{th}$ are hidden by crystallization; (ii) $\phi_{th} \lesssim \phi_c$, however, the difference between ϕ_{th} and ϕ_c is very small, such that the amorphous packings in the range $[\phi_{th}, \phi_c]$ are difficult to detect on the basis of the present numerical accuracy. Further studies are required to test these two scenarios.

To conclude, we have studied structural properties of dense hard spheres in the vicinity of the Bernal limit. The spatial and bond order parameters and the abundances of hcp-, fcc-, and ico-like particles all indicate a sharp disorder–order phase transition at ϕ_c . Finally, we find that at packing fraction $\phi \approx 0.68$ an additional structural transition occurs: the 3D-like shape of solid-like crystalline clusters changes to a planar-like structure.

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Notes

The authors declare no competing financial interest.

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A statistical theory of correlations in random packings of hard particles

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A random packing of hard particles represents a fundamental model for granular matter. Despite its importance, analytical modeling of random packings remains difficult due to the existence of strong correlations which preclude the development of a simple theory. Here, we take inspiration from liquid theories for the n -particle angular correlation function to develop a formalism of random packings of hard particles from the bottom-up. A progressive expansion into a shell of particles converges in the large layer limit under a Kirkwood-like approximation of higher-order correlations. We apply the formalism to hard disks and predict the density of two-dimensional random close packing (RCP), $\phi_{\text{rcp}} = 0.85 \pm 0.01$, and random loose packing (RLP), $\phi_{\text{rlp}} = 0.67 \pm 0.01$. Our theory also predicts a phase diagram and angular correlation functions that are in good agreement with experimental and numerical data.

I. INTRODUCTION

In recent years, many important practical applications have been found for granular materials, which are commonly modeled by dense packings of hard spheres [1]. Sphere packing problems are equivalent to important problems in number theory and error-correcting coding [2], both of which are fundamental in computer science. Despite its importance, analytical developments in granular matter have lagged behind in comparison with other fields of condensed matter, like liquid theory. In the case of random packings [3], analytical results are still difficult to obtain. The theoretical difficulty arises due to (i) the absence of a first principle derivation of the statistical ensemble of packings (such as Liouville's theorem in ordinary liquids) that would lead to a proper definition of randomness [4], and (ii) the existence of correlations between the particle positions determining the properties of random packings.

In previous theories, these correlations have been neglected or treated using simple approximations. For instance, Gotoh and Finney [5] estimated the density of RCP based only on correlations among the contact neighbors. Another example is the ‘granocentric’ model [6], which considers the correlations between the central particle and nearest Voronoi neighbors. Beyond local correlations, the statistical treatment of Song *et al.* takes a mean-field approximation of the long-range correlations [7, 8]. Other mean-field approaches are developed based on liquid theories [9] and replica theory (RT) of the glass transition [10]. However, in low dimensions the effects of fluctuations are strong and mean-field approximations are insufficient. For example, in 2d, the coarse-grained approximation used in Ref. [7] works poorly [11], and therefore more sophisticated treatments of correlations become necessary.

In this paper, we aim to establish a framework for random packings that addresses the two problems stated above: (i) we define an ensemble of equiprobable graphs that satisfy the jamming conditions to represent the statistics of all possible contact networks, and (ii) we

take into account pair and higher-order particle correlations that are important to describe low-dimensional systems. Inspired by the more advanced liquid theories, our formulation is analogous to the Yvon-Born-Green (YBG) hierarchy [12] augmented to consider the contact network and local and global jamming conditions for packings. We develop a systematic layer-expansion within a Kirkwood-like superposition approximation [12] to provide a phase diagram and predictions of the volume fractions of jammed packings. The theoretical predictions on volume fractions and pair distribution functions agree well with experiments and computer simulations on two-dimensional frictional packings. We also discuss the relation between the present approach and glass theory frameworks in search of unifications of random packings and glasses [10, 13–15].

The present approach builds up on the Edwards mean-field theory of packings developed by Song *et al.* [7], by incorporating correlations between the particle positions. Previous theory [7] utilizes a mean-field assumption of uniformity of the particle density in the bulk as well as the particles in contact. The present theory is a bottom-up approach to take into account particle-particle correlations which were neglected in [7] in a systematic way. In the thermodynamic limit of infinite number of particles in the bulk and contacts, the theory recovers the results of Song *et al.* [7], namely the exponential form of the distribution of the excluded Voronoi volume which is the basic result to predict the volume fraction of the packing.

The paper is organized as follows: in Sec. II we develop a general theoretical formalism. The formalism is applied to 2d packings (Sec. III) which provides a phase diagram (Sec. IV). The theoretical predictions are tested with experiments and computer simulations in Sec. V. At the end, we conclude our paper with discussions (Sec. VI).

II. GENERAL FORMALISM

Within the context of Edwards statistical ensemble of packings [7, 8, 16–19], the volume associated to each par-

ticle plays the role of the Hamiltonian, since packings tend to minimize the occupied volume rather than energy. The fundamental quantity to describe the packing ensemble is the Voronoi volume surrounding each particle, which is defined as the volume of the Voronoi cell whose interior consists of the points that are closer to a given particle than to any other. The d -dimensional Voronoi volume W_i of particle i is an angular average of a function of the “Voronoi radius” ℓ [7] (see Fig. 1a):

$$W_i = \oint \int_0^\ell r^{d-1} d\hat{s} dr = \frac{S_d}{d} \langle \ell^d \rangle_s, \quad (1)$$

where S_d is the d -dimensional solid angle. By the definition of Voronoi cell, ℓ is the minimum of the projection of the distance $\vec{r}_{ij}$ (from particle i to any other particle j) along the direction $\hat{s}$, $\ell \equiv \min_{\hat{s} \cdot \vec{r}_{ij} > 0} \frac{r_{ij}}{2\hat{s} \cdot \vec{r}_{ij}}$, and $\hat{s} \cdot \vec{r}_{ij} > 0$ (Fig. 1b).

According to Eq. (1), the ensemble average of the Voronoi volume is:

$$\langle W \rangle_e = \frac{1}{N} \sum_i \langle W_i \rangle_e = \frac{S_d}{d} \langle \langle \ell^d \rangle_s \rangle_i, \quad (2)$$

where $\langle \dots \rangle_s$ is the average over direction $\hat{s}$, and $\langle \dots \rangle_i$ is the average over particle i . In the random ensemble of homogeneous and isotropic packings, each particle as well as each direction is equivalent. Thus ℓ is independent of particle i and direction $\hat{s}$:

$$\langle W \rangle_e = \frac{S_d}{d} \langle \ell^d \rangle_e. \quad (3)$$

Equation (3) shows that it is enough to consider the distributions of particle positions along any arbitrary direction around any arbitrary particle, and the result is representative for the global properties of the entire packing. This feature of random packings significantly simplifies the problem. Furthermore, this ensemble average can be calculated from distribution functions:

$$\langle W \rangle_e = \frac{S_d}{d} \int_0^\infty \ell^d p(\ell) d\ell = -\frac{S_d}{d} \int_0^\infty \ell^d dP(\ell), \quad (4)$$

and the packing fraction ϕ is the ratio between the volume of spheres

$$\phi = \frac{V_d}{\langle W \rangle_e}. \quad (5)$$

Here $p(\ell)$ is the probability distribution function of ℓ in the ensemble, and $P(\ell)$ is the inverse cumulative distribution function: $p(\ell) = -dP(\ell)/d\ell$. According to the definition, $P(\ell)$ is the probability that $\frac{r_{ij}}{2\hat{s} \cdot \vec{r}_{ij}} > \ell$ for all j -particles at a distance r_{ij} from i . Geometrically, $P(\ell)$ corresponds to the probability that all particles are outside a “Voronoi excluded volume”, $\Omega(\ell)$, which is a sphere of radius ℓ (Fig. 1b). The Voronoi excluded volume is a generalization of the excluded volume due to hard-core interactions, dating back to Onsager’s hard rods solution

[20]. The distribution function $P(\ell)$ is similar to the exclusion probability function in the scaled particle theory for liquids [21], and is related to the n -particle correlation functions g_n of all orders [8].

As shown in Fig. 1c, to determine $P(\ell)$ we need to consider Voronoi particles which are the only ones with possible contributions to the Voronoi radius ℓ . This means that in the condition $\frac{r_{ij}}{2\hat{s} \cdot \vec{r}_{ij}} > \ell$ for $P(\ell)$, we only need to consider particle j labelled as a Voronoi particle. In 2d, the Voronoi particles are located on the two closest branches to the direction $\hat{s}$, but in higher dimensions more branches should be considered. The positions of the Voronoi particles are described by the n -particle angular correlation function $G_n(\alpha_1, \alpha_2, \dots, \alpha_n)$ of exclusive angles.

However, to calculate G_n one needs to define a proper ensemble first. Here we use the principle of entropy maximization which corresponds to a statistical treatment of an ensemble of all jammed states, each of which has an equal probability [5, 7, 8, 16]. This ensemble can be represented by a set of contact networks satisfying the jamming condition, while for a given contact network, particle positions are allowed to fluctuate without destroying the contacts. Our approach defines a random packing as the typical state in a flat average over the ensemble of all possible graphs of contact network configurations constraint to a given average coordination number [7]. Mechanical force and torque balance is assured by the isostatic condition imposed on the coordination number [22].

The network representation is a unique feature of packings compared to unjammed liquid systems. For contacting neighbors, we only need to know the distribution of the surface angles: the original d -dimensional problem is mapped onto a $(d-1)$ -dimensional space. The theory is mathematically treatable in two limits: (i) In 2d, the one-dimensional surface space can be analyzed analytically; (ii) In large dimensions, the contacting neighbors on the surface can be approximated to the simple ideal gas [8]. Below we apply the general formalism to study 2d random packings, where correlations are more profound.

III. APPLICATION OF THE THEORY IN 2D

In this section, we apply the general formalism in 2d, and provide quantitative predictions which can be tested by experiments and computer simulations. The approach may be generalized to higher dimensions, although the calculations might become much more complicated.

A. Calculation of $P(\ell)$

By definition, $P(\ell)$ of the central particle $i = 1$ is the probability that the Voronoi excluded volume $\Omega(\ell)$ is empty of particles, or equivalently $\frac{r_{1j}}{2\hat{s} \cdot \vec{r}_{1j}} > \ell$ for any other particle j . It is sufficient to only consider “Voronoi

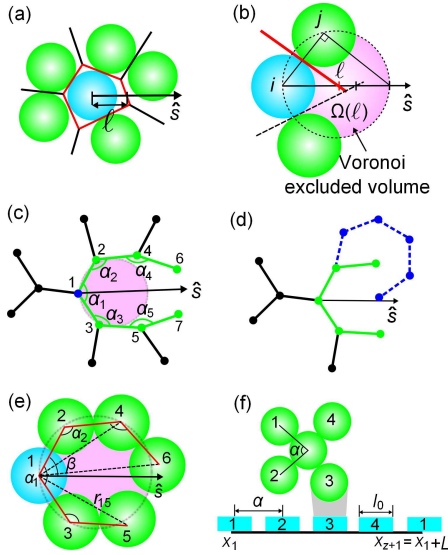


FIG. 1: (Color online) **Illustration of the theoretical formalism.** (a) A 2d illustration of the Voronoi volume (bounded by red lines) and the Voronoi radius ℓ in direction $\hat{s}$. (b) The Voronoi excluded volume $\Omega(\ell)$ (pink area). ℓ is determined by particle j because it minimizes $\frac{r_{ij}}{2\hat{s} \cdot \hat{r}_{ij}}$. (c) An illustration of the contact network and the Voronoi particles. Each dot represents a particle. The exclusive angle α_j is the angle between any two contact neighbors. No other contact particles are allowed to be inside this angle. (d) In 2d, the Voronoi radius ℓ is determined by the Voronoi particles on the two closest branches (green). Other particles may contribute only in an exceptional case such as shown by the dashed blue line. (e) An illustration of the geometrical quantities used in the calculation of $P(\ell)$. (f) Mapping monodisperse contact disks to 1d rods. The 2d exclusive angle α corresponds to the 1d gap.

particles” on the two closest branches except for the case shown in Fig. 1d. This exception disappears in the infinite expansion order limit $n \rightarrow \infty$. The condition that all Voronoi particles are outside $\Omega(\ell)$ requires that $\frac{r_{1j}}{2\cos\beta_j} > \ell$, where r_{1i} is the distance between the central particle $i = 1$ and the Voronoi particle j , and $\cos\beta_j = \hat{s} \cdot \hat{r}_{1j}$. We can write $P(\ell)$ as:

$$P(\ell) = \lim_{n' \rightarrow \infty} \int \cdots \int p(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1n'}) \times \prod_{j=2}^{n'} \Theta\left(\frac{r_{1j}}{2\hat{s} \cdot \hat{r}_{1j}} > \ell\right) d\vec{r}_{12} \cdots d\vec{r}_{1n'}, \quad (6)$$

where n' is the total number of Voronoi particles considered, and $p(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1n'})$ is the distribution function of the positions of Voronoi particles. The constraints $\Theta\left(\frac{r_{1j}}{2\hat{s} \cdot \hat{r}_{1j}} - \ell\right)$ impose the Voronoi exclusive conditions. For a given contact network, the positions $(\vec{r}_{12}, \vec{r}_{13}, \dots)$ can be transformed to the exclusive angles $(\alpha_1, \alpha_2, \dots)$ and the angle β of the direction $\hat{s}$, see Appendix A. Us-

ing this transformation, $P(\ell)$ becomes:

$$P(\ell) = \lim_{n \rightarrow \infty} \int \cdots \int p(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1,n+2}) \times \prod_{j=2}^{n+2} \Theta\left(\frac{r_{1j}}{2\hat{s} \cdot \hat{r}_{1j}} > \ell\right) \times \frac{\partial(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1,n+2})}{\partial(\beta, \alpha_1, \dots, \alpha_n)} d\beta d\alpha_1 \cdots d\alpha_n, \quad (7)$$

where we let $n' = n + 2$. **If the contact network is fixed, the degree of freedom of each particle is reduced from two to one.** Therefore the position variables $(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1,n+2})$ and angular variables $(\beta, \alpha_1, \dots, \alpha_n)$ have the same total $n + 1$ degrees of freedom.

Now the distribution of positions can be related to the distribution of angles:

$$p(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1,n+2}) \frac{\partial(\vec{r}_{12}, \vec{r}_{13}, \dots, \vec{r}_{1,n+2})}{\partial(\beta, \alpha_1, \dots, \alpha_n)} \sim G(\beta, \alpha_1, \dots, \alpha_n) \sim \Theta(\alpha_1 - \beta) G_n(\alpha_1, \dots, \alpha_n). \quad (8)$$

The Heavyside function $\Theta(\alpha_1 - \beta)$ means that the direction $\hat{s}$ is uniformly distributed and is bounded by the Voronoi particles ($\beta < \alpha_1$). Using Eq. (8), we rewrite Eq. (7) with the n -particle angular correlation function G_n :

$$P(\ell) = \lim_{n \rightarrow \infty} \frac{z}{L} \int \cdots \int \Theta(\alpha_1 - \beta) G_n(\alpha_1, \dots, \alpha_n) \times \prod_{j=2}^{n+2} \Theta\left(\frac{r_{1j}}{2\hat{s} \cdot \hat{r}_{1j}} - \ell\right) d\beta d\alpha_1 \cdots d\alpha_n, \quad (9)$$

where $L = 2\pi$, z is the average coordination number, and z/L is a normalization factor determined from the condition that $P(1/2) = 1$ (we set the particle diameter to be one). Equation (9) can be truncated at any value of n , and becomes exact in the limit $n \rightarrow \infty$. In this study, it is treated as an expansion of n or number of coordination layers (n corresponds to twice of the number of layers).

Equation (9) is similar to the YBG hierarchy [12] in liquid theories in the sense that it relates one distribution function, $P(\ell)$, to another, G_n . This similarity inspires us to bring the two approaches together to solve Eq. (9) within a closure approximation for G_n . In liquid theory, the 3-point correlation function g_3 is decomposed into the product of pair correlation functions g_2 , by the use of Kirkwood’s superposition approximation [23]. This provides a closure of the YBG hierarchy, which results in the non-linear integro-differential Born-Green equation [12]. Here we use a similar Kirkwood-like approximation to decompose G_n into the single-particle angular correlation

function $G(\alpha)$:

$$G_n(\alpha_1, \dots, \alpha_n) \approx \prod_{j=1}^n G(\alpha_j). \quad (10)$$

This approximation neglects higher-order correlations between particles that do not share any common neighbors (Appendix B).

B. Calculation of the single-particle angular correlation function $G(\alpha)$ from a 1d model

To find $G(\alpha)$, we map the contacting particles to a system of 1d rods with an effective potential. As shown in Fig. 1f, the contact particles in 2d can be mapped to a set of z interacting 1d hard rods at position x_i of length $l_0 = \pi/3$ and system size $L = 2\pi = 6l_0$, with a periodic boundary condition. The local jamming condition requires that each particle has at least $d + 1$ contacting neighbors, and not all of these neighbors are in the same “hemisphere”. In 2d, this means that $z \geq 3$ and there is no exclusive angle α that could be greater than π . In the equivalent 1d model, the latter condition requires that no two nearest neighbors are separated farther than $3l_0$. Thus, the jamming condition is equivalent to introducing an infinite square-well potential between two hard rods:

$$V(x) = \begin{cases} \infty, & \text{if } x/l_0 < 1 \text{ or } x/l_0 > a \\ 0, & \text{if } 1 < x/l_0 < a, \end{cases} \quad (11)$$

with potential parameter $a = 3$. The total potential is a sum of the pairwise potentials,

$$V(x_1, \dots, x_z) = V(L - x_z) + V(x_z - x_{z-1}) + \dots + V(x_2 - x_1). \quad (12)$$

To solve the 1d model, we first calculate the partition function $Q(L, z)$, which is

$$\begin{aligned} Q(L, z) &= \int \dots \int \exp[-V(x_1, \dots, x_z)] \prod_{i=2}^z dx_i \\ &= \int_0^L \exp[-V(L - x_z)] dx_z \\ &\quad \times \int_0^{x_z} \exp[-V(x_z - x_{z-1})] dx_{z-1} \dots \\ &\quad \times \int_0^{x_3} \exp[-V(x_3 - x_2)] \exp[-V(x_2)] dx_2, \end{aligned} \quad (13)$$

where we have used Eq. (12), and set the temperature to be unit since it is irrelevant for our system. This integral is a z -fold convolution for the Laplace transform of the function $\exp[-\beta V(x)]$ [26], which could be written as:

$$\begin{aligned} Q(L, z) &= \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{sL} q^z(s) ds, \\ q(s) &= \int_0^\infty \exp[-sx - V(x)] dx, \end{aligned} \quad (14)$$

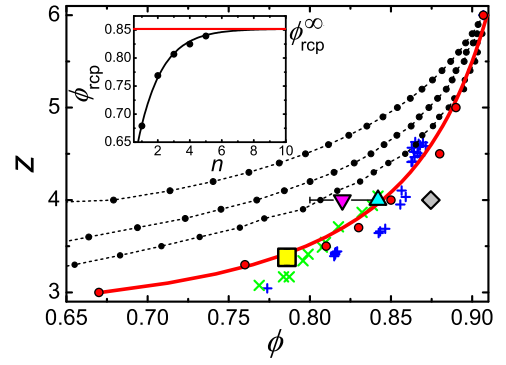


FIG. 2: (Color online) **Phase diagram of 2d packings.** Theoretical results for $n = 1, 2, 3$ (line-points, from left to right) and ϕ^∞ (red) are compared to (i) values in the literature: Berryman [9] (down triangle), RT [10] (diamond), and O’Hern *et al.* [24] (up triangle), (ii) simulations of 10,000 monodisperse disks (crosses), and polydisperse disks (pluses) with a discrete uniform distribution of radius in $[0.7, 1.0]$ (in unit of maximum radius), and (iii) experimental data of frictional disks (square). (inset) The theoretical RCP volume fraction $\phi_{\text{rcp}}^\infty(n)$ as a function of n . The points are fitted to a function $\phi_{\text{rcp}}^\infty(n) = \phi_{\text{rcp}}^\infty - k_1 e^{-k_2 n}$, where $k_1 = 0.34 \pm 0.02$, $k_2 = 0.67 \pm 0.06$, and $\phi_{\text{rcp}}^\infty = 0.85 \pm 0.01$ (blue dashed line). Other values of ϕ^∞ (with different z) are obtained in the same way.

where γ is greater than the real parts of all the singularities of $q(s)$. If we plug the potential $V(x)$ (Eq. (11)) in $q(s)$, we have (see Appendix C for details)

$$\begin{aligned} Q(L, z) &= \sum_{k=0}^{\lfloor L/l_0 - z \rfloor} (-1)^k \binom{z}{k} \frac{[L/l_0 - z - 2k]^{z-1}}{(z-1)!} \\ &\quad \times \Theta(L/l_0 - z) \Theta(3z - L/l_0), \end{aligned} \quad (15)$$

where $\lfloor x \rfloor$ is the integer part of x .

To provide an analytical form of the single-particle angular correlation function, we consider the distribution of gaps between 1d neighboring rods. For simplicity, we only consider the gap between rod 1 and 2 (its distribution is the same as that of other gaps due to translational invariance):

$$\begin{aligned} G(\alpha) &= \langle \delta(x_2 - x_1 - \alpha) \rangle \\ &= \frac{1}{Q(L, z)} \int \dots \int_{0=x_1 < x_2 < \dots < x_z < L} \prod_{i=2}^z dx_i \\ &\quad \times \exp[-\beta V(x_1, \dots, x_z)] \delta(x_2 - \alpha) \\ &= \frac{\exp[-\beta V(\alpha)]}{Q(L, z)} \int \dots \int_{\alpha=x_2 < x_3 < \dots < x_z < L} \prod_{i=3}^z dx_i \\ &\quad \times \exp[-\beta V(x_2, \dots, x_z)] \\ &= \frac{Q(\alpha, 1) Q(L - \alpha, z - 1)}{Q(L, z)}. \end{aligned} \quad (16)$$

If we set $a = \infty$ in the potential $V(x)$, the system

becomes a classical model – a one dimensional gas of hard rods (Tonks gas) [25]. In the thermodynamic limit ($L \rightarrow \infty$ and $z \rightarrow \infty$), the gap distribution is [25, 26]:

$$G_{\text{HR}}(\alpha) = \rho_f e^{-\rho_f(\alpha/l_0-1)}, \quad (17)$$

where $\rho_f = z/(L/l_0 - z)$ is the free density. This result is exact in 1d because the Kirkwood-like decomposition Eq. (10) is satisfied. Equation (17) is also consistent with the exponential form of the distribution of Voronoi excluded volume in Ref. [7], where the 1d hard rod model is used as a mean-field approximation for 3d packings.

IV. PHASE DIAGRAM OF 2D JAMMED PACKINGS

The strategy of our method to calculate the volume fraction for a fixed coordination number z is to first evaluate $G(\alpha)$ from Eq. (16), then plug it into Eqs. (10) and (9) to calculate $P(\ell)$ and eventually obtain $\langle W \rangle_e$ and ϕ via Eqs. (4) and (5). Equation (9) is a high-dimensional integration which is solved numerically by Monte Carlo method.

In the proof of the Kepler conjecture, Hales shows that considering a cluster of 50 spheres is sufficient in search for the optimal crystal packing [27]. Analogously, we expect that the volume fraction of random packings would converge quickly with n . We truncate the expansion Eq. (9) to a finite value of n , and extrapolate the finite behavior to the infinite limit. Indeed, our results show that $\phi(n)$ approaches the asymptotic value ϕ^∞ exponentially fast as $n \rightarrow \infty$ (Fig. 2 inset).

The results can be visualized into a 2d phase diagram in the $z - \phi$ plane. Figure 2 shows the equation of state $\phi^\infty(z)$ (see Appendix E for values) as well as the approach to this asymptotic value for small n . Our formalism reproduces the highest density in 2d packings obtained by Thue and Tóth [28] of hexagonal packing $\phi_{\text{hex}}^\infty = 0.91$ at $z = 6$. It also predicts the densities of isostatic packings with different friction coefficients. In order to have a mechanical stable packing, the isostatic counting argument [22, 29] requires that $z = 2d = 4$ for frictionless packings (RCP), and $z = d + 1 = 3$ for infinite frictional packings (RLP). Our theory asymptotically predicts in the two limiting cases: $\phi_{\text{rcp}}^\infty = 0.85 \pm 0.01$ and $\phi_{\text{rlp}}^\infty = 0.67 \pm 0.01$ for $z = 4$ and $z = 3$, respectively.

The 2d RCP density of monodisperse packings has been estimated theoretically by Berryman from a continuous extension of the liquid phase [9], which reports $\phi_{\text{rcp}} = 0.82 \pm 0.02$. However, this approach is questionable due to the existence of a glass transition between liquid and jammed phases as noted in [10]. Binary disk simulations (commonly used to suppress crystallization) obtain $\phi_{\text{rcp}} \sim 0.84$ [24] which is within the predicted ϕ_{rcp}^∞ . On the other hand, to our knowledge there is no reported density of 2d RLP.

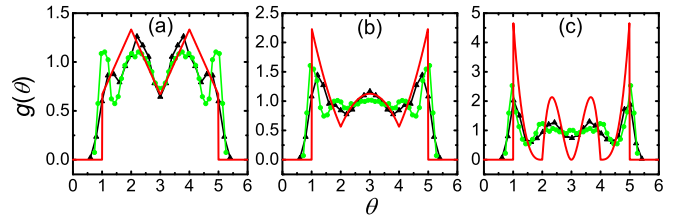


FIG. 3: (Color online) **Angular pair correlation function** $g(\theta)$. The theoretical $g(\theta)$ (red solid lines, rescaled by $\pi/3$) is compared to simulation (black triangle-lines) and experimental (green circle-lines) data, with local coordination number (a) $z_1 = 3$, (b) $z_1 = 4$, and (c) $z_1 = 5$ [30]. The simulation data are obtained from a polydisperse RCP packing in order to avoid crystallization. The subset of particles with local coordination number z_1 is used to evaluate $g(\theta)$.

More sophisticated theories use RT to solve for the density of hard spheres [10, 13], and predict that packings can exist in a range of volume fractions at the isostatic coordination number $z = 2d$ [10]. In the case of two-dimensional packings RT predicts isostatic packings in a range from the threshold density $\phi_{\text{th}} = 0.8165$ to the maximum density of glass close packing $\phi_{\text{GCP}} = 0.8745$ [10]. It is interesting to interpret our prediction of a single RCP point within the range predicted by RT [10]. The ensembles in our theory are characterized by the correlation functions like G_n or $P(\ell)$. This provides a systematic way to correlate ϕ to characteristic packing structures. If the isostatic packings could indeed have different correlations, which might be protocol-dependent in the experimental realizations, then our theory would also predict multiple packing fractions as in RT, based on proper characterizations of the correlations. This venue will test possible commonalities between Edwards statistical mechanics for jamming and the mean-field RT picture for glasses; a unification that has been sought after in the field [8, 13–15].

V. EXPERIMENTAL AND NUMERICAL TESTS

The experiments are conducted using a granular monolayer of photoelastic disks [31]. The data consist of 500 packings each containing 1004 bidisperse disks in a 1:1 concentration with diameters 11.0 mm and 14.4 mm, having an interparticle friction coefficient $\mu_B \approx 0.8$. Packings are isotropically compressed and recorded using separate images to measure the position of the disks and contact forces. This study presents data similar to Ref. [31], except that we consider only the majority particles in the bath with the same friction coefficient. More experimental details can be found in Ref. [31] and Appendix F. The average $\phi = 0.7859 \pm 0.0006$ and the average $z = 3.4 \pm 0.1$ agree well with the prediction of the theory as seen in Fig. 2.

Further test of the theory is obtained by comparing the correlations. For this purpose, we obtain the angular

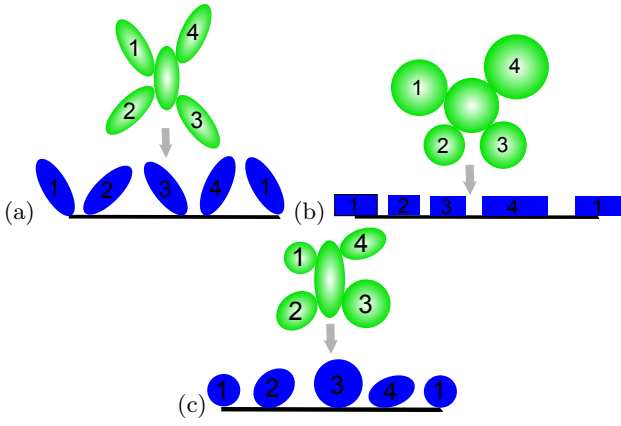


FIG. 4: (Color online) **Generalization of the model.** (a) Mapping contact ellipses to the Paris car parking model [40]. (b) Mapping polydisperse contact disks to polydisperse rods. (c) Mapping mixtures of disks and ellipses to a 1d model.

correlation function $g(\theta)$ [35] (equivalent to the pair correlation function of angles, where θ is the angle between any two surface particles) from the theory (Appendix D):

$$g(\theta) = \frac{L}{z} \sum_{m=1}^{z-1} \frac{Q(\theta, m)Q(l - \theta, z - m)}{Q(l, z)}, \quad (18)$$

which reproduces well the experimental data (Fig. 3). The theory deviates from data in the peak magnitudes but not locations when the local coordination number $z_1 = 5$. This might be due to the presence of the polydisperse effect in the experiments (which becomes more significant for larger coordination numbers), or the neglect of higher order correlations in the theory. The peak presented in the experimental data at $\theta \approx \pi/3$ (or $5\pi/3$) when $z_1 = 3$ is probably due to the remaining crystalline order in binary packings.

We also tested the theory with simulation packings generated by the Lubachevsky-Stillinger (LS) algorithm [32] and the “split” algorithm [7]. Using simulations we are able to test the full curve of $\phi(z)$. We prepare packings for both monodisperse and polydisperse disks in the random phase $3 < z < 4$ by changing the interparticle friction coefficient from zero ($z = 4$, RCP) to infinity ($z = 3$, RLP) [34]. We find a good agreement expect for small z , which suggests that for very loose packings, higher-order correlations beyond the Kirkwood decomposition Eq. (10) may be necessary. The numerical $g(\theta)$ is consistent with theories and experiments as seen in Fig. 3. Our results are in line with existing analysis [36].

VI. DISCUSSIONS AND CONCLUSIONS

In this paper, we construct a framework to study random packings. Our theory is based on a statistical approach, which assumes that each state can be visited with equal probability. The approach should be applied and

generalized with caution. For example, in Ref. [31], the authors studied the equilibrium of two subsystems with different frictions. They found that while each subsystem is equilibrated, only the angoricity (conjugate to the stress) but not the compactivity (conjugate to the volume) equilibrates between the two subsystems. In this case, one should appropriately integrate the stress ensemble with the volume ensemble. Moreover, in recent years, it is found that several protocols produce RCPs at densities different from the commonly observed values ($\phi_{\text{RCP}} \sim 0.64$ in 3d and $\phi_{\text{RCP}} \sim 0.84$ in 2d). The ensembles generated by these protocols are likely different from the Edwards ensemble, and the final states could depend on the dynamics of the protocols. In principle, one needs a dynamic theory for each of these protocols, and we leave the question open whether they can be described by static theories like the present approach.

Generalization of the formalism to other shapes of particles and polydisperse systems are possible. For instance, 2d packings of ellipses require a 1d model with orientations (Fig. 4a). The solution of such a model (named the “Paris car parking” problem [40]) will lead to a prediction of RCP and the optimal packing of elongated particles, an open theoretical problem with implication for self-assembly of nanoparticles and liquid crystal phases. It is also possible to generalize this model to polydisperse systems, by explicitly calculating the dependence of the local coordination numbers with the concentration of species [41], and mapping the problem to a “car parking” problem of polydisperse cars (Fig. 4b). Note that in our experiments and simulations, we have introduced a weak polydispersity to avoid crystalline order. Although one usually neglects the packing fraction corrections of weak polydispersities [36], we expect monodisperse theories to become insufficient for systems with strong polydispersities. Furthermore, the theory can be applied to mixtures of spherical and non-spherical objects in search of new phases of jammed matter (Fig. 4c).

Overall, the present formalism facilitates a systematic investigation of correlations in packings, and paves the path to a solvable model. The framework may be extended to predict the optimal ordered and disordered packings over a set of specified shapes, dimensions and friction properties.

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APPENDIX A: CALCULATION OF ANGLES IN $P(\ell)$

We need $n + 1$ angles, $(\beta, \alpha_1, \dots, \alpha_n)$, to determine the positions of Voronoi particles. According to the geomet-

rical relationships (Fig. 5), other angles and distances can be calculated from these integration variables recursively as:

$$\begin{aligned}
 \sigma_j &= \alpha_{j-2} - \tau_{j-2} \\
 r_{1j} &= \sqrt{r_{1,j-2}^2 + 1 - 2r_{1,j-2} \cos \sigma_j} \\
 \tau_j &= \arcsin \left(\frac{r_{1,j-2}}{r_{1j}} \sin \sigma_j \right) \\
 \eta_j &= \arcsin \left(\frac{1}{r_{1j}} \sin \sigma_j \right) \\
 \gamma_j &= \gamma_{j-2} + \eta_j,
 \end{aligned} \tag{A1}$$

and

$$\beta_j = \begin{cases} \beta_2 - \gamma_j, & \text{if } j = 4, 6, 8 \dots \\ \beta_3 - \gamma_j, & \text{if } j = 5, 7, 9 \dots \end{cases} \tag{A2}$$

with initial values

$$\begin{aligned}
 \sigma_2 &= \tau_2 = \eta_2 = \gamma_2 = 0, \\
 \beta_2 &= \beta, \\
 r_{12} &= 1, \\
 \sigma_3 &= \tau_3 = \eta_3 = \gamma_3 = 0, \\
 \beta_3 &= \alpha_1 - \beta_2, \\
 r_{13} &= 1.
 \end{aligned} \tag{A3}$$

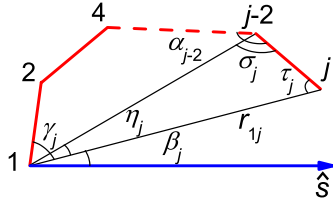


FIG. 5: An illustration of the geometrical relations between angles and distances.

APPENDIX B: A DISCUSSION ON THE KIRKWOOD-LIKE DECOMPOSITION OF THE n -PARTICLE ANGULAR CORRELATION FUNCTION

The Kirkwood-like decomposition Eq. (5) in the main text is an approximation of the n -particle angular correlation function $G_n(\alpha_1, \dots, \alpha_n)$, which neglects higher-order correlations between particles that do not share a common contact neighbor. To see this, let us look at the simplest case when $n = 2$. An expression of $G_2(\alpha_1, \alpha_2)$

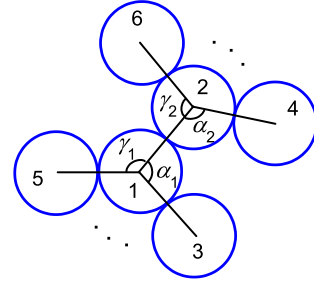


FIG. 6: An illustration of particles and angles in Eq. (B1). There are $z-3$ particles (not shown) between particles 3 and 5 (4 and 6).

is:

$$\begin{aligned}
 G_2(\alpha_1, \alpha_2) &\sim \int_{\gamma_1=0}^L \int_{\gamma_2=0}^L Q(\alpha_1, 1) Q(\gamma_1, 1) \\
 &\times Q(\alpha_2, 1) Q(\gamma_2, 1) \\
 &\times Q(L - \alpha_1 - \gamma_1, z - 2) \\
 &\times Q(L - \alpha_2 - \gamma_2, z - 2) \\
 &\times \Theta(r_{34} - 1) \Theta(r_{56} - 1) d\gamma_1 d\gamma_2,
 \end{aligned} \tag{B1}$$

where $L = 2\pi$, and $Q(L, z)$ is the partition function of 1d rods (see below). The particles and angles are indicated in Fig. 6. The Heaviside step functions impose the hard-sphere constraints between particles 3 and 4, and between 5 and 6, which are not in direct contact with any common neighbors (compared to “direct” particles such as particles 2 and 3, which share a common neighbor particle 1). If we neglect the hard-sphere constraints between these indirect particles, and only include correlations between direct particles, Eq. (B1) becomes

$$\begin{aligned}
 G_2(\alpha_1, \alpha_2) &\sim \int_{\gamma_1=0}^L \int_{\gamma_2=0}^L Q(\alpha_1, 1) Q(\gamma_1, 1) \\
 &\times Q(\alpha_2, 1) Q(\gamma_2, 1) \\
 &\times Q(L - \alpha_1 - \gamma_1, z - 2) \\
 &\times Q(L - \alpha_2 - \gamma_2, z - 2) d\gamma_1 d\gamma_2
 \end{aligned} \tag{B2}$$

Because

$$\int_{\gamma_1=0}^L \frac{Q(L - \alpha_1 - \gamma_1, z - 2) Q(\gamma_1, 1)}{Q(L - \alpha_1, z - 1)} d\gamma_1 = 1, \tag{B3}$$

(same for γ_2), Eq. (B2) can be further written as

$$\begin{aligned}
 G_2(\alpha_1, \alpha_2) &\sim Q(\alpha_1, 1) Q(L - \alpha_1, z - 1) \\
 &\times Q(\alpha_2, 1) Q(L - \alpha_2, z - 1) \\
 &\sim G(\alpha_1) G(\alpha_2).
 \end{aligned} \tag{B4}$$

The above derivation shows that the 2-particle angular correlation function $G_2(\alpha_1, \alpha_2)$ can be approximated as a product of single-particle angular correlation functions, if higher-order correlations are neglected. The same analysis can be extended to $G_n(\alpha_1, \dots, \alpha_n)$ when $n > 2$.

APPENDIX C: PARTITION FUNCTION OF 1D RODS

To simplify the notation, here we set the size of rods to be the unit, $l_0 = 1$. The full expressions (in the main text) are recovered by adding a proper scaling factor $1/l_0$ to the distance parameters, such as x and L . If we plug the potential Eq. (11) in $q(s)$ (Eq. (14)), we have

$$q(s) = \int_1^a e^{-sx} dx = \frac{e^{-s} - e^{-as}}{s}, \quad (C1)$$

and the partition function becomes

$$\begin{aligned} Q(L, z) &= \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} e^{sL} \left(\frac{e^{-s} - e^{-as}}{s} \right)^z ds \\ &= \sum_{k=0}^z (-1)^k \binom{z}{k} \left\{ \frac{1}{2\pi i} \int_{\gamma-i\infty}^{\gamma+i\infty} \frac{e^{s[L-z-k(a-1)]}}{s^z} \right\} \\ &= \sum_{k=0}^{\lfloor \frac{L-z}{a-1} \rfloor} (-1)^k \binom{z}{k} \frac{[L-z-k(a-1)]^{z-1}}{(z-1)!} \\ &\quad \times \Theta(L-z)\Theta(aL-L). \end{aligned} \quad (C2)$$

where we have used the binomial expansion of $\left(\frac{e^{-s} - e^{-as}}{s} \right)^z$.

APPENDIX D: ANGULAR PAIR CORRELATION FUNCTION $g(\theta)$

The 2d angular pair correlation function $g(\theta)$ is equivalent to the pair correlation function in the 1d model, which is the probability of finding a rod at a given distance θ from another rod. $g(\theta)$ is different from $G(\alpha)$ because other rods are allowed to be inside θ . Due to the translational invariance, we can choose any rod (rod 1 in this case) as the reference point:

$$\begin{aligned} \rho g(\theta) &= \left\langle \sum_{k=2}^z \delta(x_k - x_1 - \theta) \right\rangle \\ &= \frac{1}{Q(L, z)} \sum_{k=2}^z \int \cdots \int_{0=x_1 < x_2 < \cdots < x_z < L} \prod_{i=2}^z dx_i \\ &\quad \times \exp[-\beta V(x_1, \dots, x_z)] \delta(x_k - \theta) \\ &= \frac{1}{Q(L, z)} \sum_{k=2}^z \int \cdots \int_{0=x_1 < x_2 < \cdots < x_k = \theta} \prod_{i=2}^{k-1} dx_i \\ &\quad \times \exp[-\beta V(x_1, \dots, x_{k-1})] \\ &\quad \times \int \cdots \int_{\theta=x_k < x_{k+1} < \cdots < x_z < L} \prod_{i=k+1}^z dx_i \\ &\quad \times \exp[-\beta V(x_k, \dots, x_z)] \\ &= \sum_{k=2}^z \frac{Q(\theta, k-1)Q(L-\theta, z-k+1)}{Q(L, z)}, \end{aligned} \quad (D1)$$

where the number density $\rho = z/L$. From the last expression, the angular pair correlation function $g(\theta)$ can be written as

$$\begin{aligned} g(\theta) &= \frac{1}{\rho} \sum_{m=1}^{z-1} g_m(\theta), \\ g_m(\theta) &= \frac{Q(\theta, m)Q(L-\theta, z-m)}{Q(L, z)}. \end{aligned} \quad (D2)$$

The function $g_m(\theta)$ is the probability density of finding two contact particles at a relative angle θ , such that there is exactly $m-1$ contact particles between them. Equation (D2) is used to calculate the theoretical $g(\theta)$ in Fig. 3.

The normalization of $g(\theta)$ is conventional:

$$\int_0^L \rho g(\theta) d\theta = z - 1. \quad (D3)$$

APPENDIX E: THEORETICAL VALUES OF $\phi^\infty(z)$

In Table I, we list the extrapolated values of $\phi^\infty(z)$ evaluated from our theory (see Fig. 2).

z	3.0	3.3	3.5	3.7	4.0	4.5	5.0	6.0
ϕ^∞	0.67	0.76	0.81	0.83	0.85	0.88	0.89	0.91

TABLE I: Theoretical values of $\phi^\infty(z)$.

APPENDIX F: COLLECTION OF EXPERIMENTAL DATA

The experiments involve an assembly of 1004 bi-disperse, photoelastic disks having a diameter of 11.0 mm and 15.4 mm in equal concentration by number. Particles are composed of photoelastic material (Vishay PhotoStress PSM-4) and are birefringent under strain so that contact forces can be calculated. The granular monolayer rests on a nearly frictionless surface of an air table and is confined by two immovable walls and two pistons. The system is initially dilute and unjammed. Two pistons bi-axially compress the system through a series of small quasi-static steps with a size corresponding to $\Delta\Phi = 0.0009$. At each step, separate images are recorded to measure the displacement and contact forces. We use only data collected from jammed configurations over the range of $0.7836 < \phi < 0.7884$. After the system has reached the maximum desired ϕ , the pistons dilate and the system is mixed. This cycle is repeated ensuring generation of independent configurations. In this way, over 500 packings are obtained and analyzed. More details of the experimental apparatus and procedures are reported in a recent paper, Ref. [31].

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Calculation of the Voronoi boundary for lens-shaped particles and spherocylinders

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We have recently developed a mean-field theory to estimate the packing fraction of non-spherical particles [A. Baule *et al.*, *Nature Commun.* (2013)]. The central quantity in this framework is the Voronoi excluded volume, which generalizes the standard hard-core excluded volume appearing in Onsager's theory. The Voronoi excluded volume is defined from an exclusion condition for the Voronoi boundary between two particles, which is usually not tractable analytically. Here, we show how the technical difficulties in calculating the Voronoi boundary can be overcome for lens-shaped particles and spherocylinders, two standard prolate and oblate shapes with rotational symmetry. By decomposing these shapes into unions and intersections of spheres analytical expressions can be obtained.

I. INTRODUCTION

Packings of hard particles are ubiquitous in many fields in science and engineering [1]. Most studies have focused on particles with spherical symmetry, which in a disordered arrangement typically achieve packing fractions of $\approx 64\%$ volume fraction. By contrast, both prolate and oblate non-spherical shapes can achieve higher packing fractions, as found in simulations [2–4] and experiments [5]. The existence of strong positional and orientational correlations has so far prevented any systematic study apart from the limit of infinitely thin rods, which are treated in an equilibrium setting by Onsager's virial expansion of the free energy [6]. However, the densest packings are typically found in a regime close to the sphere [4], for which this expansion breaks down.

A statistical mechanical framework to treat jammed granular matter has been proposed more than two decades ago by S. F. Edwards, who postulated that the macroscopic properties of these systems can be calculated as ensemble averages similar to equilibrium systems [7]. These averages are taken over all jammed microstates at a fixed system volume, where all microstates are assumed as equiprobable. This means that the role of energy in thermal systems is replaced by the volume in granular systems, leading to an analogous statistical mechanical framework. The main task is then to obtain the volume function (analogue of the Hamiltonian), which describes the system volume as a function of the particles' positions and orientations. Here, different conventions can be employed to partition the total volume into cells associated with each particle [8, 9], the simplest of which is the Voronoi tessellation [10, 11]. However, in 3D these exact volume functions are difficult to handle analytically, requiring a suitable coarse-graining procedure.

We have recently followed such a mesoscopic approach in order to develop a mean-field theory of packings of both spheres and anisotropic particles [12–14]. The central quantity in our framework is the coarse-grained Voronoi volume $W(z)$ of a single particle, which, roughly speaking, contains

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the volume that is closer to this particle than to any other one on average. This approach is further motivated by the observation that, as the particle number $N \rightarrow \infty$, packings show reproducible phase behaviour and are characterized by only few observables such as the packing fraction and the average coordination number z [11, 15, 16]. $W(z)$ satisfies a self-consistent equation: [12–14]

$$W(z) = \int d\mathbf{c} \exp \left\{ -\frac{V^*(\mathbf{c})}{W - V_0} - \sigma(z)S^*(\mathbf{c}) \right\}, \quad (1)$$

where the key ingredients $V^*(\mathbf{c})$ and $S^*(\mathbf{c})$ are referred to as the Voronoi excluded volume and surface, which extend Onsager’s hard-core excluded volume to jammed packings. In Eq. (1), the quantity $\sigma(z)$ is the free surface density for a packing with z contacts and is obtained from simulations of local configurations. Once $W(z)$ is determined by solving the self-consistency equation, the packing fraction as a function of z follows simply from $\phi = V_0/W(z)$, where V_0 is the particle volume. This approach has to be complemented with a prescription for the value of z in the packing, which is fixed by the isostatic conjecture $z = 2d_f$ satisfied by spheres. However, anisotropic particles can have $z < 2d_f$ due to redundancy in the force and torque balance equations that define mechanical equilibrium. A quantitative theory for this effect has been developed in Ref. [14] and calculates z as an average $z = 2 \langle d_{\text{eff}} \rangle$ over effective number of degrees of freedom.

Therefore, in order to apply our framework the main task is to calculate $V^*(\mathbf{c})$ and $S^*(\mathbf{c})$ for a particular particle shape. Both quantities are defined from an exclusion condition on the Voronoi boundary (VB) between two particles, so that the calculation requires analytic expressions for the VB, which are typically difficult to obtain [10, 17]. In this paper we show how to calculate the VB of lens-shaped particles and spherocylinders, two model shapes for both oblate and prolate anisotropic particles. Regular crystal packings of these shapes have recently been investigated theoretically in Ref. [18]. For both shapes analytic expressions for the VB can be obtained, which is in contrast to, e.g., prolate and oblate ellipsoids. The underlying reason is that both can be decomposed into decompositions and intersections of spheres of equal radii, such that the VB is generated from simpler effective interactions, namely those between points, lines, and anti-points leading to an exact algorithm for the VB [14]. By comparison, the corresponding decomposition of prolate and oblate ellipsoids requires a dense set of spheres with continuously varying radii, which greatly complicates the problem. In the following we explicitly show how to calculate the VB for lens-shaped particles and spherocylinders following our algorithm. This will guide the calculation of the VB between more complicated shapes that can be decomposed similarly.

This paper is organized as follows. In the next section we first recapitulate how to calculate the VB between spheres before showing how to extend it to lens-shaped particles (Sec. II A) and how to calculate the contact radius between two such objects (Sec. II B). Then we review the calculation of both quantities for spherocylinders (Sec. II C and Sec. II D). We summarize how to use these results in order to evaluate the excluded volume and surface of these particles in Sec. II E. We finally conclude with a brief discussion of potential generalizations (Sec. III).

II. RESULTS

We set the centre of our coordinate system to the centre of mass of particle i and fix the orientation of this particle along $\hat{\mathbf{z}}$. Given a direction $\hat{\mathbf{c}}$, a point on the VB is found at $s\hat{\mathbf{c}}$, where s depends on the position $\mathbf{r}$ and orientation $\hat{\mathbf{t}}$ of particle j : $s = s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})$. The value of s is obtained from two conditions:

1. The point $s\hat{\mathbf{c}}$ has the minimal distance to each of the two objects along the direction $\hat{\mathbf{c}}$.

2. Both distances are the same.

The VB between two spheres of equal radii is the same as the VB between two points at the centres of the spheres. Therefore, condition 1 is trivially satisfied for every s and condition 2 translates into the equation

$$(s\hat{\mathbf{c}})^2 = (s\hat{\mathbf{c}} - \mathbf{r})^2, \quad (2)$$

leading to

$$s = \frac{r}{2\hat{\mathbf{c}}\hat{\mathbf{r}}}, \quad (3)$$

i.e., the VB is the plane perpendicular to the separation vector $\mathbf{r}$ at half the separation. Already for two spheres of unequal radii, the VB is a curved surface. Taking into account the different radii a_i and a_j , Eq. (2) becomes [19]

$$s - a_i = \sqrt{(s\hat{\mathbf{c}} - \mathbf{r})^2} - a_j, \quad (4)$$

which has the solution

$$s = \frac{1}{2} \frac{r^2 - (a_i - a_j)^2}{\hat{\mathbf{c}}\hat{\mathbf{r}} - (a_i - a_j)}. \quad (5)$$

Finding a solution for both conditions for general non-spherical objects is in general non-trivial. Next we show how to overcome the difficulties for lens-shaped particles.

A. The VB between two lens-shaped particles

A lens-shaped particle is made of two spherical parts and their circular junction that we call the “crown”. Here we use two spheres of the same radius R so that the gravity centre is the centre of the crown. L denotes its diameter of and l the thickness of the lens. The aspect ratio of a lens is then defined by $\alpha = l/L$ (see Fig. 1). We first wrote a short code (using Geogebra) to visualize qualitatively the VB between two lens-shaped particles in 2D for the most general case of an intersection of two spheres with different radii (Fig. 3).

We then attempted to follow the calculation explained above for spheres, with the same set of conditions to determine the VB. However, the spherical parts of the lens determine the VB only in specific regions. These are defined by cones as displayed in Fig. 1. When the VB is found out of these cones, we should consider the extremity of the lens that corresponds to the circle defined by all points belonging to both spheres (the “crown”) for the calculation. Indeed, a sphere is assimilated to its centre for the calculation because the centre, the point of the sphere that interacts and the VB are aligned. It is not valid anymore when being out of the cones for lenses. Thus if we fix two lenses in space, the VB along a given direction depends on these cones, and we have the following different types of interaction (Fig. 4):

1. *Sphere-Sphere* (4 interactions): The boundary falls in a cone of each lenses. We have the same calculation as in the sphere study.
2. *Sphere-Crown* and *Crown-Sphere* (4 interactions): The boundary falls in a cone of the first lens but does not fall in a cone of the second. The distances to equalize are between the surface of the interacting sphere and a precise point of the interacting crown.

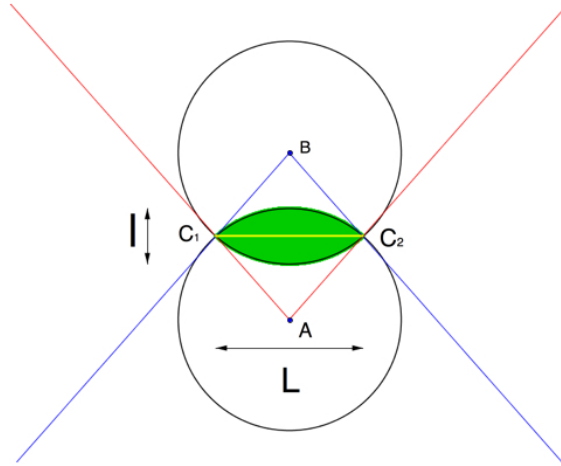


FIG. 1: A lens is made of two spheres centred in A and B. The lens is the green shape, the intersection of the spheres. The yellow line denotes the “crown”, whose extremities in the plane are marked C₁ and C₂.

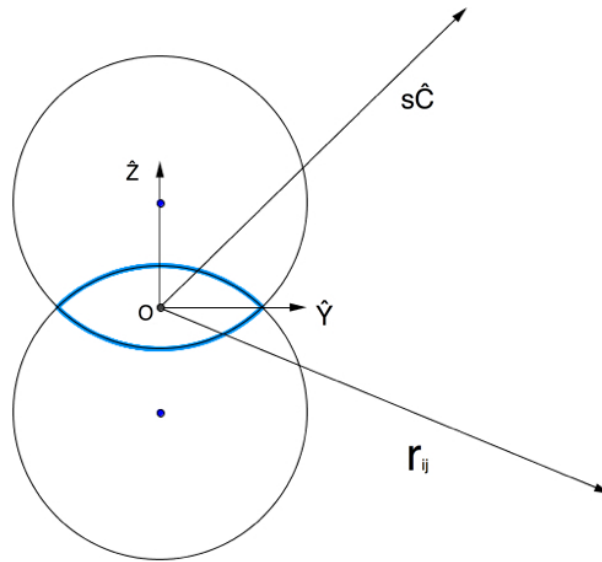


FIG. 2: Parametrization of lens *i*, blue points denote the centres of the spheres constituting the lens.

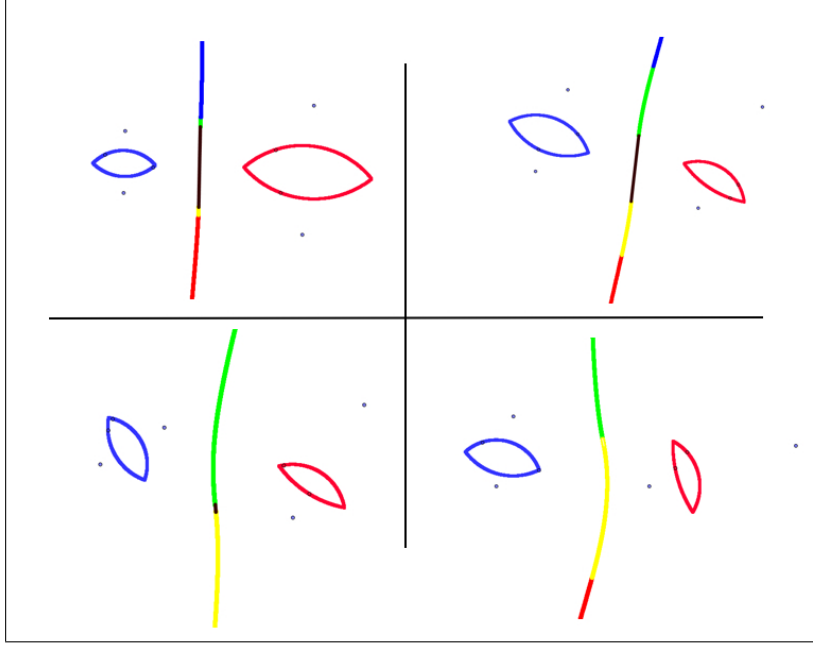


FIG. 3: VB of two lenses in the plane, coloured in function of the type of interaction. Sphere-Sphere interaction: red and blue lines; Sphere-Crown interaction: green and yellow lines; Crown-Crown: black line. The four points are the positions of the centres of the spheres constituting the lenses.

3. *Crown-Crown* (1 interaction): The boundary falls out of both cones for both lenses. The distances to equalize are between two precise points on the crowns.

The purpose of the following algorithm is to calculate each interaction one after the other and to evaluate if the VB effectively falls in the corresponding area and if it is the minimum of all interactions. If it does, then it is a valid solution. Some tests may allow one to avoid the evaluation of one or two cases.

1. Parametrization of the problem

We use the parameters displayed in Fig. 2. The origin of the coordinate system is set to the gravity centre of lens i . $r\hat{\mathbf{r}}_{ij}$ links the centres of lens i and j . The VB is to be found along vector $\hat{\mathbf{c}}$. We can set $\hat{\mathbf{c}}$ in the plane $(\hat{\mathbf{y}}, \hat{\mathbf{z}})$ because of the axisymmetry of the lens. The orientation of lens j in the space is given by the vector $\hat{\mathbf{z}}_j$. We are interested in the calculation of s along $\hat{\mathbf{c}}$, where s denotes the position of the VB between lens i and lens j . As mentioned before we have the same set of conditions as in the sphere study:

1. The point $s\hat{\mathbf{c}}$ has the minimal distance to each of the two objects along the direction $\hat{\mathbf{c}}$.
2. Both distances are the same.

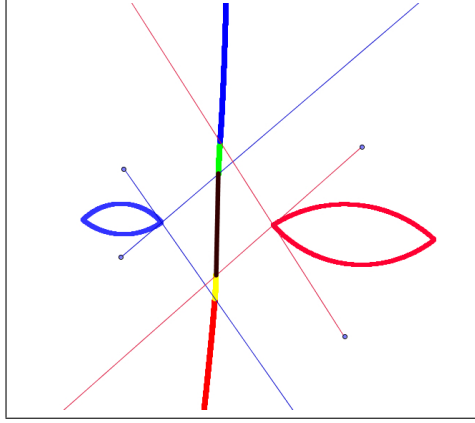


FIG. 4: Different interactions that generate the VB depending on the cone configurations

Depending on the direction of $\hat{\mathbf{c}}$, s may necessarily be in a cone of i , or not. It is determined by an angular test. If the direction of $\hat{\mathbf{c}}$ stays in a cone of lens i , the spherical part of lens i whose centre is the vertex of this cone, will interact. It is assimilated to the centre of this sphere. Thus, the square of the distance between $s\hat{\mathbf{c}}$ and the concerned sphere i is:

$$D_i^2 = (\pm(R - \frac{l}{2})\hat{\mathbf{z}} + s\hat{\mathbf{c}})^2. \quad (6)$$

If s may be found outside of both cones of lens i , depending of the orientation of $\hat{\mathbf{c}}$ relatively to the cones of lens i , one should also evaluate a crown interaction for i . In this case, the projection of $\hat{\mathbf{c}}$ on the crown i will interact since it is the closest point of lens i to s , changing Eq. (6) into:

$$D_i^2 = (-\frac{L}{2}\hat{\mathbf{y}} + s\hat{\mathbf{c}})^2, \quad (7)$$

We then compute all interactions with lens j and keep the minimum result that falls its good corresponding area. This is the purpose of the algorithm: We try to determine the interacting part of i at the beginning with an angular test and calculate each combination with j . For each interaction we determine if s falls inside the corresponding area of influence of j and is the minimum in comparison with the other cases. If it does then it is a valid solution, if not we calculate another interaction. Tests are based on an angular verification of the positions of s relatively to the cones of j , as well as we had for i . Thus, if s falls in a cone of j

$$D_j^2 = (\pm(R - \frac{l}{2})\hat{\mathbf{z}}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})^2. \quad (8)$$

If s falls outside of both cones j , the point of crown j that interacts is necessarily the one defined by the projection of the vector $-r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}}$ on the crown of lens j . It is given by $\mathbf{t}_j$:

$$\mathbf{t}_j = \frac{L}{2N}(\hat{\mathbf{z}}_j \times ((-r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}}) \times \hat{\mathbf{z}}_j)) \quad (9)$$

and

$$N^2 = r^2(\hat{\mathbf{z}}_j \times (\hat{\mathbf{r}}_{ij} \times \hat{\mathbf{z}}_j))^2 - 2rs(\hat{\mathbf{z}}_j \times (\hat{\mathbf{r}}_{ij} \times \hat{\mathbf{z}}_j))(\hat{\mathbf{z}}_j \times (\hat{\mathbf{c}} \times \hat{\mathbf{z}}_j)) + s^2(\hat{\mathbf{z}}_j \times (\hat{\mathbf{c}} \times \hat{\mathbf{z}}_j))^2 \quad (10)$$

Thus,

$$D_j^2 = (-\mathbf{t}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})^2, \quad (11)$$

Condition 2 requires $D_i^2 = D_j^2$. However, if a sphere is assimilated to its centre, a crown is not. Indeed if we assimilate the crown to its centre, we may have solutions on the sphere centred on the crown that are not on the plane of the crown. Thus we will have to subtract the radius of a sphere in one term of the equation $D_i^2 = D_j^2$ in the Sphere-Crown and Crown-Sphere interactions.

Condition 1 is always satisfied in each calculation because we work with spheres of the same radius, and because the interacting point of the crown is necessarily defined by the projections of the vectors calculated above. Now we show the results for the different interactions:

2. Sphere-Sphere interaction

This case arises if s falls in a cone of lens i and a cone of lens j . $D_i^2 = D_j^2$ leads to:

$$(\pm(R - \frac{l}{2})\hat{\mathbf{z}} + s\hat{\mathbf{c}})^2 = (\pm(R - \frac{l}{2})\hat{\mathbf{z}}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})^2 \quad (12)$$

Thus we obtain for s :

$$s = \frac{1}{2} \frac{r^2 \pm 2(R - l/2)r\hat{\mathbf{z}}_j\hat{\mathbf{r}}_{ij}}{\pm(R - l/2)\hat{\mathbf{z}}\hat{\mathbf{c}} \pm (R - l/2)\hat{\mathbf{z}}_j\hat{\mathbf{c}} + r\hat{\mathbf{r}}_{ij}\hat{\mathbf{c}}} \quad (13)$$

3. Crown-Sphere interaction

In this case s is outside of both cones of lens i . Thus, the distances to equalize are not between the centres of two spheres $S_i - S_j$ and s but between the crown of lens i and the spherical surface of lens j . The point of crown i that interacts is necessarily the one defined by the projection of $\hat{\mathbf{c}}$ on the crown of lens i , and we have to subtract the radius of the concerned sphere j ($\pm$) in the equation. Since all spheres do have the same radius we have:

$$(-\frac{L}{2}\hat{\mathbf{y}} + s\hat{\mathbf{c}})^2 + 2R\sqrt{(-\frac{L}{2}\hat{\mathbf{y}} + s\hat{\mathbf{c}})^2 + R^2} = (\pm(R - \frac{l}{2})\hat{\mathbf{z}}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})^2 \quad (14)$$

We then have an equation that is to be squared in order to obtain a polynomial of order 2 on s :

$$\begin{aligned} & r^2 + 2r(\pm(R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{r}}_{ij} - s\hat{\mathbf{r}}_{ij}\hat{\mathbf{c}}) \pm 2s(R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{c}} \\ & + sL\hat{\mathbf{y}}\hat{\mathbf{c}} + (R - \frac{l}{2})^2 - \frac{L^2}{4} - R^2 = 2R\sqrt{(-\frac{L}{2}\hat{\mathbf{y}} + s\hat{\mathbf{c}})^2} \end{aligned} \quad (15)$$

4. Sphere-Crown interaction

In this case, angular tests for the previous interactions show that s does not fall inside any of both cones of lens j . As in the Crown-Sphere interaction we subtract the radius of a sphere in one

term of the equation:

$$\left(\sqrt{\left(\pm\left(R - \frac{l}{2}\right)\hat{\mathbf{z}} + s\hat{\mathbf{c}}\right)^2 - R^2} - (-\mathbf{t}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})\right)^2 \quad (16)$$

Since the denominator N of $\mathbf{t}_j$ depends on the square root of terms in s^2, s , and constants, we square the previous equation to have a polynomial in s . It is a polynomial of order 6 that we resolve numerically.

5. Crown-Crown interaction

Now all the angular tests show that s is out of both cones of lens i and both cones of lens j . $D_i^2 = D_j^2$ leads to:

$$\left(-\frac{L}{2}\hat{\mathbf{y}} + s\hat{\mathbf{c}}\right)^2 = (-\mathbf{t}_j - r\hat{\mathbf{r}}_{ij} + s\hat{\mathbf{c}})^2 \quad (17)$$

We proceed as in Eq. (16) and obtain a polynomial of order 6 that we also resolve numerically.

B. Calculation of the contact radius of two lens-shaped particles

Still using the same parametrization, we now want to determinate the position r^* of the centre of lens j so that it is in contact with lens i along $\hat{\mathbf{r}}_{ij}$. So we just consider the vector $\hat{\mathbf{r}}_{ij}$, and $\hat{\mathbf{c}}$ has no influence in this calculus. We also have a test procedure and a choice to make for r^* .

It consists of calculating first a Sphere-Sphere contact and then determinate if the point of contact belongs to both lens. Indeed, it may happen that the point of contact is on a sphere constituting a lens, but not on the lens itself. This determines an upper limit to r^* . If the test is positive, it is a valid solution. If the test is negative, then one of the crowns necessarily is in contact.

Thus we calculate a Sphere-Crown contact and Crown-Sphere one. For each of these two new interactions we test if the point of contact belongs to both lenses. If the test is positive for both interactions the solution is valid and the same for both cases. If it is positive for only one interaction then it is a valid one. If it is negative for both interactions r^* is necessarily determined by a Crown-Crown contact. It may happen since the tests are exclusive and because the resolution of the equations is numerical for the Sphere-Crown and Crown-Sphere interactions. We may also have special cases where lenses i and j have the same orientation with their crown in the same plane, thus a Crown-Crown interaction is directly calculated. We note that there is one and only one Sphere-Sphere interaction to evaluate: The sphere from which $\hat{\mathbf{r}}_{ij}$ goes out for i and the one that it enters for j .

1. Sphere-Sphere

We directly obtain

$$\left(\pm\left(R - \frac{l}{2}\right)\hat{\mathbf{z}} + r^*\hat{\mathbf{r}}_{ij} \pm \left(R - \frac{l}{2}\right)\hat{\mathbf{z}}_j\right)^2 = 4R^2 \quad (18)$$

The signs ($\pm$) depend on which sphere interacts, and after developing we keep the positive root of the polynomial:

$$(r^*)^2 + 2r^*(R - l/2)(\pm \hat{\mathbf{z}} \pm \hat{\mathbf{z}}_j)\hat{\mathbf{r}}_{ij} + 2(R - \frac{l}{2})^2(1 \pm \hat{\mathbf{z}}\hat{\mathbf{z}}_j) - 4R^2 \quad (19)$$

2. Crown-Sphere and Sphere-Crown

These interactions are of course similar. We show here the Crown-Sphere interaction, and we follow the same procedure in a coordinate system fixed to lens j to calculate the Sphere-Crown interaction. We denote by (x, y, z) the point of contact. It belongs to the crown i , which simply means:

$$(S) \begin{cases} x^2 + y^2 = L^2/4 \\ z = 0 \end{cases}$$

It also belongs to the projection of the sphere j on the plane of crown i , thus we add to the previous system:

$$(r^*\hat{\mathbf{r}}_{ij}\hat{\mathbf{x}} \pm (R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{x}} - x)^2 + (r^*\hat{\mathbf{r}}_{ij}\hat{\mathbf{y}} \pm (R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{y}} - y)^2 = R^2 \quad (20)$$

Then we substitute x from (S) in Eq. (20), thus :

$$(r^*\hat{\mathbf{r}}_{ij}\hat{\mathbf{x}} \pm (R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{x}} - \sqrt{\frac{L^2}{4} - y^2})^2 + (r^*\hat{\mathbf{r}}_{ij}\hat{\mathbf{y}} \pm (R - \frac{l}{2})\hat{\mathbf{z}}_j\hat{\mathbf{y}} - y)^2 = R^2 \quad (21)$$

We develop and simplify the expression, which gives a polynomial of order 2 in y . The equations and the algorithm take into account that y and r^* must be positive since the signs in Eq. (20) depends on the interacting sphere ($\pm(R - l/2)\hat{\mathbf{z}}_j\hat{\mathbf{y}}$ and $\pm(R - l/2)\hat{\mathbf{z}}_j\hat{\mathbf{x}}$ terms).

Thus we do necessarily have one and only one solution for y so that $\Delta = 0$ for the polynomial of order 2 on y . This leads to a polynomial of order 6 in r^* that we resolve numerically. Thus we obtain y and an angular test allows one to obtain x :

$$x = \pm \sqrt{L^2/4 - y^2} \quad (22)$$

These coordinates x and y allow one to find now if the interaction is the good one with another angular test. In order to calculate the Sphere-Crown interaction we temporarily change the base that we set in lens j and do the same calculation in the plane of Crown j .

3. Crown-Crown

We denote by (x, y, z) the point of contact. It belongs to the crown i :

$$(S1) \begin{cases} x^2 + y^2 = L^2/4 \\ z = 0 \end{cases}$$

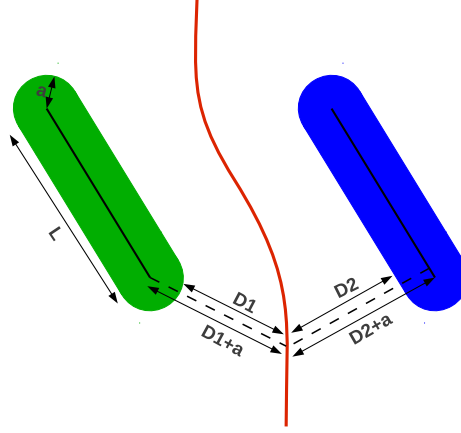


FIG. 5: Voronoi diagram equivalence between two spherocylinders and two line segments defining the spherocylinders. The VB is equivalent to that of two rods of vanishing width and length L , i.e., solving for the point where $D_1 = D_2$ is the same as solving $D_1 + a = D_2 + a$.

It also belongs to the crown j :

$$(S2) \begin{cases} (r^* \hat{\mathbf{r}}_{ij} \hat{\mathbf{x}} - x)^2 + (r^* \hat{\mathbf{r}}_{ij} \hat{\mathbf{y}} - y)^2 + r^{*2} \hat{\mathbf{r}}_{ij} \hat{\mathbf{z}} = L^2/4 \\ -x \hat{\mathbf{x}} \hat{\mathbf{z}}_j - y \hat{\mathbf{y}} \hat{\mathbf{z}}_j + r^* \hat{\mathbf{r}}_{ij} \hat{\mathbf{z}}_j = 0 \end{cases}$$

We substitute x in the first equation of $(S2)$ with its expression from the second equation of $(S2)$ to have an expression on y . Then we follow the exact same procedure as mentioned in the Sphere-Crown interaction.

C. The VB between two spherocylinders

The calculation of the Voronoi diagram of spherocylinders is comparatively simpler than the one for, e.g., ellipsoids thanks to the following property: The VB of two spherocylinders of length L (of the cylindrical part) and radius a (of the semi-spherical end-caps) is equivalent to the VB between two line segments of length L (see Fig. 5) at the centre of the cylindrical part. In the following we refer to these line segments as “rods”. This equivalence is analogous to the sphere–point equivalence of equal size spheres.

The radius a of the spherocylinders thus does not appear explicitly in the calculation of the VB as outlined in the next sections. However, the radius enters naturally as a limiting condition for the possible configurations of the spherocylinders and defines the contact radius $r^*(\hat{r}, \hat{t})$, which is discussed in more detail in Sec. IID.

The calculation of the VB and the contact radius between two spherocylinders has previously been presented in the supplementary material of Ref. [14]. In the following we use the convention that a vector $\mathbf{x}$ can be decomposed as $\mathbf{x} = x \hat{\mathbf{x}}$, where x denotes the absolute value and $\hat{\mathbf{x}}$ the unit direction. The product of two vectors $\mathbf{x} \mathbf{y}$ denotes the scalar product $\mathbf{x} \cdot \mathbf{y} = \sum_k x_k y_k$, where the sum is over all components.

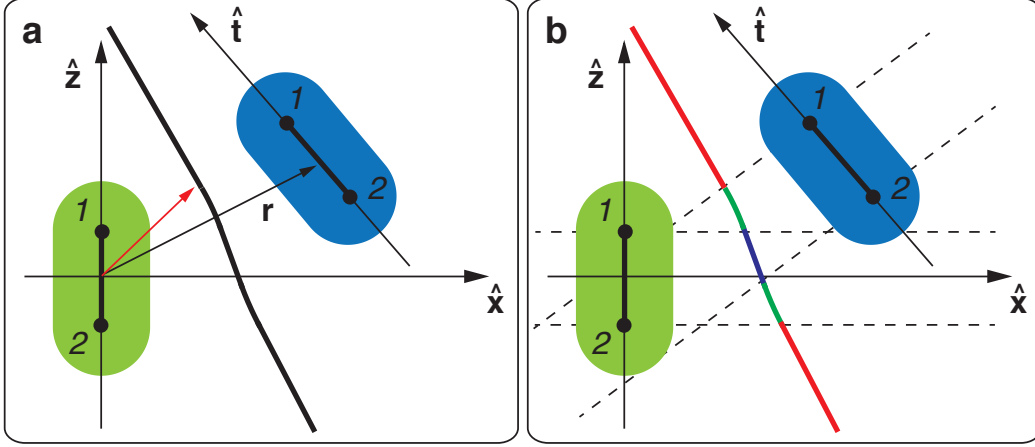


FIG. 6: **(a)** Parametrization of the VB between two spherocylinders of relative orientation $\hat{\mathbf{t}}$ and position $\mathbf{r}$. **(b)** The VB consists of the VBs due to the interaction of the four points and two lines (indicated in different colours), which are separated following our algorithm. The red part of the VB is due to the Point-Point interactions, the green part due to Line-Point and Point-Line, and the blue part due to the Line-Line.

We align rod i with the $\hat{\mathbf{z}}$ axis of our coordinate system, so that a point on it is parametrized by the vector $t_i \hat{\mathbf{z}}$ with $t_i \in [-L/2; L/2]$. Likewise, the orientation of rod j is given by $\hat{\mathbf{t}}$, so that a point on rod j is parameterized by $\mathbf{r} + t_j \hat{\mathbf{t}}$, where also $t_j \in [-L/2; L/2]$. See Fig. 6a for the setup of our coordinate system. In order to solve the two conditions that define the VB, we first find the distance between $s\hat{\mathbf{c}}$ and a point on rod i and j , denoted by D_i^2 and D_j^2 , respectively. We obtain

$$D_i^2 = (t_i \hat{\mathbf{z}} - s\hat{\mathbf{c}})^2, \quad (23)$$

$$D_j^2 = (\mathbf{r} + t_j \hat{\mathbf{t}} - s\hat{\mathbf{c}})^2. \quad (24)$$

Condition 1. (minimal distance) requires:

$$\frac{\partial D_i^2}{\partial t_i} = 0, \quad (25)$$

$$\frac{\partial D_j^2}{\partial t_j} = 0. \quad (26)$$

Solving these two conditions yields the minimal values

$$t_i^{\min} = s\hat{\mathbf{c}}\hat{\mathbf{z}} = s(\hat{\mathbf{c}}\hat{\mathbf{z}}), \quad (27)$$

$$t_j^{\min} = (s\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} = s(\hat{\mathbf{c}}\hat{\mathbf{t}}) - r. \quad (28)$$

Condition 2. (equal distances) requires:

$$D_i^{\min} = D_j^{\min}, \quad (29)$$

so that

$$(t_i^{\min} \hat{\mathbf{z}} - s\hat{\mathbf{c}})^2 = (t_j^{\min} \hat{\mathbf{t}} + \mathbf{r} - s\hat{\mathbf{c}})^2. \quad (30)$$

Here, it is important to note that $t_i^{\min}$ and $t_j^{\min}$ are only the correct minimal values when $t_i^{\min} \in [-L/2, L/2]$ and $t_j^{\min} \in [-L/2, L/2]$ due to the finite length of the rods. If $t_i^{\min}$ and/or $t_j^{\min}$ are not $\in [-L/2, L/2]$ interactions involving the end-points of the rods arise. Overall, one has to distinguish the cases:

1. *Line-Line* interaction: $t_i^{\min} \in [-L/2, L/2]$ and $t_j^{\min} \in [-L/2, L/2]$ (1 case).
2. *Line-Point* interaction between the segment i and an end-point of j : $t_i^{\min} \in [-L/2, L/2]$ and $t_j = \pm L/2$ (2 cases).
3. *Point-Line* interaction between the segment j and an end-point of i : $t_j^{\min} \in [-L/2, L/2]$ and $t_i = \pm L/2$ (2 cases).
4. *Point-Point* interaction between the end points of i and j : $t_i = \pm L/2$ and $t_j = \pm L/2$ (4 cases).

In the following the different Voronoi interactions are indicated by different subscripts, e.g., s_{ll} for Line-Line interaction, s_{lp} for Line-Point interaction, etc. Fig. 6b illustrates the separation of the different interactions.

1. Line-Line interaction

This interaction is valid if $t_i^{\min}$ and $t_j^{\min}$ fall inside the length of the segments. The conditions are thus:

$$t_i^{\min} \in [-L/2, L/2], \quad t_j^{\min} \in [-L/2, L/2]. \quad (31)$$

In this case $t_i^{\min}$ and $t_j^{\min}$ are given by Eqs. (27) and (28). Substituting these expressions into Eq. (30) yields a quadratic equation for the VB value $s = s_{ll}$:

$$\frac{s_{ll}^2}{r^2} [(\hat{\mathbf{c}}\hat{\mathbf{z}})^2 - (\hat{\mathbf{c}}\hat{\mathbf{t}})^2] + 2\frac{s_{ll}}{r} [(\hat{\mathbf{c}}\hat{\mathbf{t}})(\hat{\mathbf{r}}\hat{\mathbf{t}}) - \hat{\mathbf{r}}\hat{\mathbf{c}}] + 1 - (\hat{\mathbf{r}}\hat{\mathbf{t}})^2 = 0. \quad (32)$$

The correct solution of this equation is the real and positive one. We observe that the solution s_{ll} scales with the separation r . The conditions for Eqs. (31) to hold are

$$-L/2 \leq s_{ll}\hat{\mathbf{c}}\hat{\mathbf{z}} \leq L/2, \quad \text{and} \quad -L/2 \leq (s_{ll}\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} \leq L/2. \quad (33)$$

These two conditions define the separation lines for the Line-Line interaction visualized in Fig. 6b.

2. Line-Point interaction

In this case $t_j^{\min}$ is fixed at one of the end points of rod j and $t_i^{\min}$ is inside the line segment of rod i . We set the point to the top of $\mathbf{t}_j$, indicated by a subscript 1 and obtain:

$$t_i^{\min} \in [-L/2, L/2], \quad t_j^{\min} = L/2. \quad (34)$$

Substituting the Eq. (27) for $t_i^{\min}$ and $t_j^{\min} = L/2$ into Eq. (30) yields a quadratic equation for the VB value $s = s_{\text{lp}_1}$, where the index p_1 refers to the top point:

$$\frac{s_{\text{lp}_1}^2}{r^2}(\hat{\mathbf{c}}\hat{\mathbf{z}})^2 - 2\frac{s_{\text{lp}_1}}{r}\left[(\hat{\mathbf{r}}\hat{\mathbf{c}}) + \frac{L}{2r}(\hat{\mathbf{c}}\hat{\mathbf{t}})\right] + \left(\frac{L}{2r}\right)^2 + \frac{L}{r}(\hat{\mathbf{r}}\hat{\mathbf{t}}) + 1 = 0. \quad (35)$$

The VB generated by the bottom point s_{lp_2} , which is defined by $t_j^{\min} = -L/2$, follows straightforwardly by setting $L \rightarrow -L$ in Eq. (35). The conditions for the two Line-Point interactions are then (cf. Fig. 6b)

$$-L/2 \leq s_{\text{lp}_1}\hat{\mathbf{c}}\hat{\mathbf{z}} \leq L/2 \quad \text{and} \quad (s_{\text{lp}_1}\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} \geq L/2 \quad (36)$$

$$-L/2 \leq s_{\text{lp}_2}\hat{\mathbf{c}}\hat{\mathbf{z}} \leq L/2 \quad \text{and} \quad (s_{\text{lp}_2}\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} \leq -L/2. \quad (37)$$

3. Point-Line interaction

We can calculate this interaction in analogy to the Line-Point interaction. The conditions are:

$$t_i^{\min} = L/2, \quad t_j^{\min} \in [-L/2, L/2]. \quad (38)$$

Substituting $t_i^{\min} = L/2$ for the top point and Eq. (28) into Eq. (30) leads to

$$\begin{aligned} & \frac{s_{\text{p}_1}^2}{r^2}(\hat{\mathbf{c}}\hat{\mathbf{t}})^2 + 2\frac{s_{\text{p}_1}}{r}[(\hat{\mathbf{r}}\hat{\mathbf{c}}) - (\hat{\mathbf{c}}\hat{\mathbf{t}})(\hat{\mathbf{r}}\hat{\mathbf{t}})] \\ & - \frac{s_{\text{p}_1}}{r}\frac{L}{r}(\hat{\mathbf{c}}\hat{\mathbf{z}}) + \left(\frac{L}{2r}\right)^2 + (\hat{\mathbf{r}}\hat{\mathbf{t}})^2 - 1 = 0. \end{aligned} \quad (39)$$

Likewise for s_{p_2} . The conditions for the two point-line interactions are then

$$s_{\text{p}_1}\hat{\mathbf{c}}\hat{\mathbf{z}} \geq L/2, \quad \text{and} \quad -L/2 \leq (s_{\text{p}_1}\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} \leq L/2 \quad (40)$$

$$s_{\text{p}_2}\hat{\mathbf{c}}\hat{\mathbf{z}} \leq -L/2, \quad \text{and} \quad -L/2 \leq (s_{\text{p}_2}\hat{\mathbf{c}} - \mathbf{r})\hat{\mathbf{t}} \leq L/2. \quad (41)$$

4. Point-Point interaction

This interaction is obtained by fixing both $t_i^{\min}$ and $t_j^{\min}$ to $L/2$ or $-L/2$. We set

$$t_i^{\min} = L_i/2, \quad t_j^{\min} = L_j/2, \quad (42)$$

where $L_i = \pm L$ and $L_j = \pm L$ for the top and bottom points on each of the rods. The solution of Eq. (30) with Eqs. (42) is then:

$$s_{pp} = r \frac{1 + \frac{L_j}{r}(\hat{\mathbf{r}}\hat{\mathbf{t}})}{2(\hat{\mathbf{r}}\hat{\mathbf{c}}) + \frac{L_i}{r}(\hat{\mathbf{c}}\hat{\mathbf{t}}) - \frac{L_i}{r}(\hat{\mathbf{c}}\hat{\mathbf{z}})}. \quad (43)$$

The different Point-Point interactions are obtained by specifying the $L_{i,j}$. The VB value $s_{\text{p}_1\text{p}_1}$ due to the two top points $s_{\text{p}_1\text{p}_1}$, e.g., is obtained by setting $L_i = L_j = L$. Likewise for the other point

interactions. The conditions for the four different point-point Voronoi boundaries are

$$s_{p_1 p_1} \hat{\mathbf{c}} \hat{\mathbf{z}} \geq L/2, \quad \text{and} \quad (s_{p_1 p_1} \hat{\mathbf{c}} - \mathbf{r}) \hat{\mathbf{t}} \geq L/2, \quad (44)$$

$$s_{p_1 p_2} \hat{\mathbf{c}} \hat{\mathbf{z}} \geq L/2, \quad \text{and} \quad (s_{p_1 p_2} \hat{\mathbf{c}} - \mathbf{r}) \hat{\mathbf{t}} \leq -L/2, \quad (45)$$

$$s_{p_2 p_1} \hat{\mathbf{c}} \hat{\mathbf{z}} \leq -L/2, \quad \text{and} \quad (s_{p_2 p_1} \hat{\mathbf{c}} - \mathbf{r}) \hat{\mathbf{t}} \geq L/2, \quad (46)$$

$$s_{p_2 p_2} \hat{\mathbf{c}} \hat{\mathbf{z}} \leq -L/2, \quad \text{and} \quad (s_{p_2 p_2} \hat{\mathbf{c}} - \mathbf{r}) \hat{\mathbf{t}} \leq -L/2. \quad (47)$$

Eq. (43) reduces to the VB between two equal spheres, Eq. (3), in the limit $L/r \rightarrow 0$. Note that the VBs due to the four point-point interactions are flat surfaces, while interactions involving the line segment generate curved VBs.

5. Examples

As an example of the algorithm, we apply it to different situations in 2 dimensions, Fig. 7. We consider the rod i (with varying orientations) on the left and the rod j on the right. The top-left panel shows the Voronoi boundary in different colours corresponding to different interactions: A point-point interaction at the top of the boundary in red, then a point-line interaction in green, then a line-line interaction in blue, then another point-line interaction in green and so on. The other panels are analogous.

D. The contact radius of two spherocylinders

The contact radius $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$ is defined as the value of r such that two spherocylinders of relative orientation $\hat{\mathbf{t}}$ and position $r\hat{\mathbf{r}}$ are in contact. By comparison, two spheres are in contact if $r^* = 2a$, independent of $\hat{\mathbf{t}}$ and $\hat{\mathbf{r}}$. For spherocylinders we have to distinguish the possible contacts of the spherical endcaps and of the cylindrical segments (see Fig. 8). As before, we denote a point on rod i by $t_i \hat{\mathbf{z}}$ and a point on rod j by $\mathbf{r} + t_j \hat{\mathbf{t}}$. The squared distance between these two points is

$$\begin{aligned} D^2(\mathbf{r}, \hat{\mathbf{t}}, t_i, t_j) &= (t_i \hat{\mathbf{z}} - (\mathbf{r} + t_j \hat{\mathbf{t}}))^2 \\ &= t_i^2 + t_j^2 + r^2 + 2r(t_j(\hat{\mathbf{r}}\hat{\mathbf{t}}) - t_i(\hat{\mathbf{r}}\hat{\mathbf{z}})) - 2t_i t_j (\hat{\mathbf{z}}\hat{\mathbf{t}}). \end{aligned} \quad (48)$$

Two spherocylinders are in contact when the minimal D^2 equals the square of the diameter $(2a)^2$. We solve $\partial D^2 / \partial t_i = 0$ and $\partial D^2 / \partial t_j = 0$ in order to obtain the two minimal positions

$$t_i^* = r \frac{(\hat{\mathbf{r}}\hat{\mathbf{z}}) - (\hat{\mathbf{r}}\hat{\mathbf{t}})(\hat{\mathbf{z}}\hat{\mathbf{t}})}{1 - (\hat{\mathbf{z}}\hat{\mathbf{t}})^2} = r A_i \quad (49)$$

$$t_j^* = r \frac{(\hat{\mathbf{r}}\hat{\mathbf{z}})(\hat{\mathbf{z}}\hat{\mathbf{t}}) - (\hat{\mathbf{r}}\hat{\mathbf{t}})}{1 - (\hat{\mathbf{z}}\hat{\mathbf{t}})^2} = r A_j, \quad (50)$$

which define A_i and A_j . Substituting these expressions into Eq. (48) and solving for r under the condition $D^2 = 4a^2$ yields the contact radius

$$r_{\text{II}}^*(\hat{\mathbf{r}}, \hat{\mathbf{t}}) = \frac{2a}{\sqrt{1 + (A_i \hat{\mathbf{z}} - A_j \hat{\mathbf{t}})^2 + 2(A_j(\hat{\mathbf{r}}\hat{\mathbf{t}}) - A_i(\hat{\mathbf{r}}\hat{\mathbf{z}}))}}. \quad (51)$$

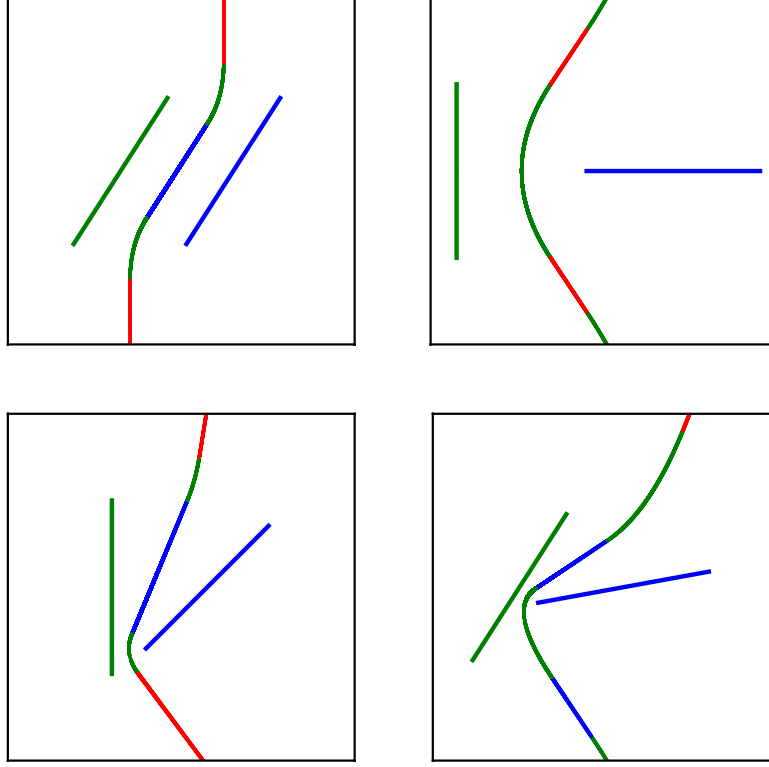


FIG. 7: Examples of the VB between two rods in 2d with different configurations. Each colour of the boundary represents a different type of interaction. Point-Point interaction: red; Line-Point and Point-Line: green; Line-Line: blue.

This contact radius is only valid for $t_i^* \in [-L/2, L/2]$ and $t_j^* \in [-L/2, L/2]$, since it does not take into account the finite length of the spherocylinders. Therefore, r_{ll}^* is the contact between the line segments (indicated by the ll subscript as before). In complete analogy to the Voronoi interactions we need to distinguish further the Line-Point, Point-Line and Line-Line contacts in addition to the Line-Line one.

For the Line-Point contact one has to consider $t_j = \pm L/2$. In order to find the minimal t_i^{*lp} one thus has to solve

$$\frac{\partial}{\partial t_i} D^2 \left(\mathbf{r}, \hat{\mathbf{t}}, t_i, \pm \frac{L}{2} \right) = 0. \quad (52)$$

Substituting this value back into D^2 and solving $D^2 = 4a^2$ for r yields the two Line-Point contact radii. These are valid when $t_i^{*lp} \in [-L/2, L/2]$. For the Point-Line contact one has to set $t_i = \pm L/2$, which determines the minimal t_j^{*pl} by the equation

$$\frac{\partial}{\partial t_j} D^2 \left(\mathbf{r}, \hat{\mathbf{t}}, \pm \frac{L}{2}, t_j \right) = 0. \quad (53)$$

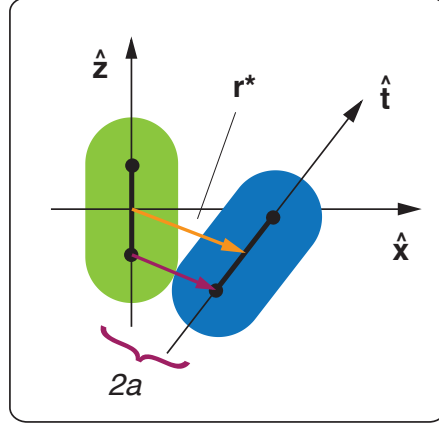


FIG. 8: The contact radius $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$ for two spherocylinders. Here, the contact is due to the spherical endcaps.

Substituting this value back into D^2 and solving $D^2 = 4a^2$ for r yields the two Point-Line contact radii. These are valid when $t_j^{\text{pl}} \in [-L/2, L/2]$. For the Point-Point contact one can solve directly

$$D^2 \left(\mathbf{r}, \hat{\mathbf{t}}, \pm \frac{L}{2}, \pm \frac{L}{2} \right) = 4a^2 \quad (54)$$

for r , which yields four different Point-Point contact radii.

Following this procedure, we obtain 9 possible values for the contact radius $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$, just like the 9 different VB values. Among these values correct radius is determined as the maximum of all positive and real ones.

E. Calculation of the excluded volume and surface

For completeness, we summarize here our method to calculate V^* and S^* for a given analytic form of the VB $s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})$. This method has previously been implemented in Ref. [14]. We define excluded Voronoi volume as $V^* = \langle \Omega - \Omega \cap V_{\text{ex}} \rangle_{\hat{\mathbf{t}}}$, which can be calculated as an orientational average over a volume integral:

$$V^*(\mathbf{c}) = \left\langle \int d\mathbf{r} \Theta(r - r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})) \Theta(c - s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})) \Theta(s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})) \right\rangle_{\hat{\mathbf{t}}}. \quad (55)$$

These integrals can be expressed in spherical coordinates. We denote with θ_r and β_r the polar and azimuthal angles, respectively, of the position. The corresponding orientational angles have a subscript t . Eq. (55) then corresponds to the multi-dimensional integral

$$V^*(c, \theta_c) = \frac{1}{2\pi} \int_0^\pi d\theta_r \int_{-\pi}^\pi d\beta_r \int_0^{\pi/2} d\theta_t \int_{-\pi}^\pi d\beta_t \int_{r^*(\theta_r, \beta_r, \theta_t, \beta_t)}^\infty dr r^2 \sin(\theta_t) \sin(\theta_r) \times \Theta[c - s(r, \theta_r, \beta_r, \theta_t, \beta_t, \theta_c)] \Theta[s(r, \theta_r, \beta_r, \theta_t, \beta_t, \theta_c)]. \quad (56)$$

The reduction of the full solid angle $\hat{\mathbf{c}}$ to θ_t and the integration limits of the θ_t integration take into account the symmetry of the spherocylinders. Clearly, Eq. (56) is a five dimensional integral, which be calculated numerically using, e.g., a Monte-Carlo scheme for a given $\mathbf{c}$.

We define the excluded Voronoi surface as $S^* = \langle \partial V_{\text{ex}} \cap \Omega \rangle_{\hat{\mathbf{t}}}$, which can be expressed as an orientational average over a surface integral:

$$S^*(\mathbf{c}) = \left\langle \oint d\hat{\mathbf{r}} \Theta(c - s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})) \Theta(s(\mathbf{r}, \hat{\mathbf{t}}, \hat{\mathbf{c}})) \Big|_{r=r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})} \right\rangle_{\hat{\mathbf{t}}}, \quad (57)$$

Here, the contact radius $r^*(\hat{\mathbf{r}}, \hat{\mathbf{t}})$ induces the surface element

$$d\hat{\mathbf{r}} = r^* \sqrt{\left(r^{*2} + \left(\frac{\partial r^*}{\partial \theta_r} \right)^2 \right) \sin^2(\theta_r) + \left(\frac{\partial r^*}{\partial \beta_r} \right)^2} d\theta_r d\beta_r, \quad (58)$$

which recovers the usual surface element $d\hat{\mathbf{r}} = r^{*2} \sin(\theta_r) d\theta_r d\beta_r$ for $r^* = \text{const.}$ Eq. (57) can thus be written in terms of the four-dimensional integral

$$S^*(c, \theta_c) = \frac{1}{2\pi} \int_0^\pi d\theta_r \int_{-\pi}^\pi d\beta_r \int_0^{\pi/2} d\theta_t \int_{-\pi}^\pi d\beta_t \sin(\theta_t) r^* \sqrt{\left(r^{*2} + \left(\frac{\partial r^*}{\partial \theta_r} \right)^2 \right) \sin^2(\theta_r) + \left(\frac{\partial r^*}{\partial \beta_r} \right)^2} \times \Theta[c - s(r^*, \theta_r, \beta_r, \theta_t, \beta_t, \theta_c)] \Theta[s(r^*, \theta_r, \beta_r, \theta_t, \beta_t, \theta_c)], \quad (59)$$

where $r^* = r^*(\theta_r, \beta_r, \theta_t, \beta_t)$. This expression can also be computed numerically.

III. DISCUSSION

The decomposition of a shape into compositions and intersections of spheres can be generalized to more complicated shapes. In particular, the VB between any shapes that are represented by a composition or intersection of a finite number of spheres, such as trimers, tetramers, etc. with varying degree of overlap, can be calculated in a straightforward way following this method. The main challenge is to develop a concrete and efficient procedure for a given shape, i.e. a methodic protocol that is valid for a given number n of overlapping spheres. The algorithm for lenses is already quite complicated. Indeed, even though the computational complexity to determine a single VB is not high, many cases have to be computed and kept in memory until the VB is identified that specifies the correct interaction. There are additional technical difficulties for the separation of the interactions. For lens-shaped particles, e.g., polynomials of order 6 have to be solved numerically.

Shapes of particular recent interest in theoretical materials science are polyhedra, which can self-assemble into structures much more complicated than spheres and have thus great potential for the assembly of new functional materials [20, 21]. In order to apply our framework to polyhedra,

one can consider a decomposition of the shape into a dense union of spheres with continuously varying radii. This kind of decomposition (also called filling) can be optimized following certain design principles [22]. Even though our method to determine the VB is still algorithmically well-defined for such a dense filling, it is difficult to implement in practice. A simpler approach would be to approximate a polyhedra as an intersection of a small finite number of spheres, similar to the approximation of an ellipsoid by a lens-shaped particle discussed here. A cube, e.g., can be approximated as the intersection of six spheres [14], such that the VB can be calculated following our method.

Even if an analytical expression for the VB of a particular shape can be obtained, the calculation of the excluded volume and surface are still computationally costly due to the high dimensional integrals. The calculation of the Jacobian provides additional difficulties. A direct numerical computation of the VB might provide an alternative approach, which can be optimized for speed using, e.g., graphics hardware [23].

The code used to generate the VB for lens-shaped particles and spherocylinders is available on www.jamlab.org.

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Stress-dependent normal mode frequencies from the effective mass of granular matter

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A zero-temperature critical point has been invoked to control the anomalous behavior of granular matter as it approaches jamming or mechanical arrest. Criticality manifests itself in an anomalous spectrum of low-frequency normal modes and scaling behavior near the jamming transition. The critical point may explain the peculiar mechanical properties of dissimilar systems such as glasses and granular materials. Here, we study the critical scenario via an experimental measurement of the normal modes frequencies of granular matter under stress from a pole decomposition analysis of the effective mass. We extract a complex-valued characteristic frequency which displays scaling $|\omega^*(\sigma)| \sim \sigma^{\Omega'}$ with vanishing stress σ for a variety of granular systems. The critical exponent is smaller than that predicted by mean-field theory opening new challenges to explain the exponent for frictional and dissipative granular matter. Our results shed light on the anomalous behavior of stress-dependent acoustics and attenuation in granular materials near the jamming transition.

I. INTRODUCTION

In granular media, a jammed system results if the particles pack together at high enough density such that all the particles are touching their neighbors [1]. As a result, the material undergoes a jamming transition above which the system remains arrested in a jammed state and is able to withstand a sufficiently small applied stress. The jamming transition occurs at a particularly subtle point where the particles have no redundant mechanical constraints, i.e. they are isostatic [2–5]. The system is a fragile solid that is highly susceptible to external perturbations. For instance, cutting a set of particle contacts creates floppy modes which necessitate global rearrangement to rebalance the system [2, 3, 6–8].

This critical point has important consequences for the mechanical properties of granular matter. In particular, there exists an excess of low-frequency modes above that expected from the Debye scaling of phonons in elastic solids [9, 10]. When the system is jammed, a band of excess modes exists for $\omega > \omega_D^*$, ω_D^* is a characteristic frequency that vanishes as the jamming transition is approached [10] [the subscript indicates that this frequency determines the limit of validity of Debye scaling in the density of states]. For $\omega < \omega_D^*$ one has the usual Debye density of states. The system is argued to be critical with a diverging correlation length ℓ^* [6] scaling as the inverse of the characteristic frequency. The excess modes distinguish an isostatic critical state at scales below ℓ^* from an elastic solid at scales larger than ℓ^* [6, 7]. The characteristic frequency follows scaling law [6–12]:

$$\omega_D^* \sim (\phi - \phi_c)^\Omega \sim \sigma^{\Omega'}, \quad (1)$$

with external stress, σ , or volume fraction, ϕ , measured with respect to the jamming point $\Delta\phi = \phi - \phi_c$ (eg random close packing or point J [4, 5, 9]).

The static properties of jammed systems are also characterized by scaling laws. In particular, the average coordination number scales as

$$Z - Z_c \sim (\phi - \phi_c)^{1/2}, \quad (2)$$

with scaling exponent approximately independent of friction [4, 5, 7–9, 11, 13]. Here, $\Delta Z = Z - Z_c$ measures the excess contacts from the minimal isostatic coordination number for frictionless grains $Z_c = 6$, or a critical value $4 \leq Z_c \leq 6$ for frictional grains in 3D [11, 13, 14]. The constitutive law between stress and volume fraction is

$$\sigma \sim (\phi - \phi_c)^\alpha, \quad (3)$$

where $\alpha = 3/2$ for Hertzian spheres and $\alpha = 1$ for linear springs. Thus, $\Omega = \Omega'\alpha$.

To compare across systems with different constitutive laws, we separate the trivial stress dependence of α from the non-trivial structural dependence characterized by exponent δ :

$$\omega_D^* \sim \Delta\phi^{(\alpha-1)/2} \Delta\phi^\delta, \quad (4)$$

with $\delta = \Omega + (1 - \alpha)/2$. Theory [6, 7] has found using a variational argument of boundary contact removal process for a system of particles with linear constitutive law ($\alpha = 1$)

$$\omega_D^*(\Delta Z) \sim \Delta Z, \quad (\alpha = 1, \text{mean-field scaling}), \quad (5)$$

yielding a prediction $\delta_{\text{th}} = 1/2$, using Eq. (2), which agrees with simulations of frictionless particles [10].

So far, the scaling behavior of Eq. (1) has not been tested by direct experimental measurements on zero-temperature granular matter. Indeed, experiments cannot calculate the density of states by directly measuring the interparticle potential as done in simulations [9, 10]. Here, we employ a novel *dynamical* measurement to measure the normal modes.

Previous experiments focused on thermally agitated colloidal glasses and supercooled liquid systems [15–17]. In contrast, granular materials are athermal (the energy necessary to displace a grain is much larger than the thermal energy $\sim 10^{14}$ times) and friction dominated. Moreover, Eq. (1) was derived for non-frictional non-dissipative systems [6, 7]. In the case of real granular media, dissipation plays a key role in governing the dynamics [20]. In this work, we develop an experimental study of the stress-dependent normal modes frequencies for the case of granular matter. We demonstrate that the normal mode spectrum of a finite sized granular system can be

determined through a pole decomposition of the frequency-dependent effective mass [18–20]. The results of this analysis supports the scaling hypothesis Eq. (1) in a realistic athermal and dissipative medium. However, the exponents characterizing this granular medium are smaller than those predicted by theory Eq. (5) [6, 7].

II. EXPERIMENTS

We employ a dynamical effective mass measurement to obtain the normal modes which complements other measures of the normal modes obtained from the Fourier transform of the velocity autocorrelation function and the eigenvalues of the displacement correlation matrix [8–12]. Specifically, we measure the frequency dependent effective mass, $\tilde{M}(\omega)$, introduced previously in [18–20], of a granular medium held in a cup under varying stress.

The full experimental program has been developed in [18–20] and it is described in detail in Appendix X. Here we refer to the most important aspects of the experiments and the corresponding set of predictions. The stress-dependent scaling behavior of the normal mode spectrum of the granular system can be determined through a pole decomposition of the frequency-dependent effective mass [18–20]. We use a variety of granular systems consisting of particles of different sizes (ranging from ~ 150 microns to millimeters), different shapes (irregular and spherical), different materials (tungsten, steel and glass), and different damping conditions (particles coated with silicone oil and uncoated).

An important factor in the dynamics of this system is the existence of dissipative modes. Accordingly, our initial experiments follow the procedure described in reference [20] to lightly coat the tungsten particles (nominal size $\sim 150 \mu\text{m}$) with silicone oil (PDMS), which yields a system with known dissipative properties— later, we will show results for uncoated particles. The weakly wet powder is poured into a cup, and the cup is tapped to encourage the powder to settle and yield a flat free surface, Fig. 1. Finally, a standard mechanical compaction protocol is followed [21–24] to prepare the system in a reversible state which yields a reproducible $\tilde{M}(\omega)$.

Once loaded, the cup is subjected to a vertical sinusoidal vibration at angular frequency ω . The force, $\tilde{F}(\omega)$, is measured by a force gauge mounted between the cup and the shaker, and the acceleration, $\tilde{a}(\omega)$, is taken as the average of that measured at two points on the opposite end of a single diameter. The effective mass of the granular medium is the causal response function [18–20]:

$$\tilde{M}(\omega) = \frac{\tilde{F}(\omega)}{\tilde{a}(\omega)} - M_c, \quad (6)$$

where M_c is the mass of the empty cup. Here, $\tilde{M}(\omega) = M_1(\omega) + iM_2(\omega)$ is complex-valued and reflects the partially in-phase and out-of-phase motion of the individual grains relative to the cup motion. M_1 is indicative of the elastic, and M_2 of the dissipative characteristics of the powder. Figures 2a and b show $M_1(\omega)$ and $M_2(\omega)$ for tungsten systems exposed to an incrementally increasing uniaxial confining stress,

σ . In all cases, the lowest characteristic frequency mode manifests itself as the sharpest resonance peak in $M_2(\omega)$. This peak and the other, lower amplitude, modes in the system are characterized by a complex-valued normal mode frequency, ω_n . Roughly speaking, $\text{Re}[\omega_n]$ is given by the position of the frequency mode, and $\text{Im}[\omega_n]$ is half of the full width at half max of the frequency mode.

To accurately infer ω_n from the data in Fig. 2 we analyze $\tilde{M}(\omega)$ within the context of a theory of damped and frictional contact forces [20], which generalizes the results of previous analyses [6, 7]. While previous theories have considered central force systems [6, 7], interpreting the dynamic response of our experiments necessitates a formalism that accounts for translational and rotational degree of freedom and damped modes. We develop such a formalism next.

III. THEORY

A. Interparticle force-law

In general the granular medium is modeled as a set of grains held in a rigid cup. The theory is valid for any linearized set of contact forces. Without loss of generality, below we define it for the specific case of Hertz-Mindlin contact forces. The normal force between any two contacting particles with radius R is [25, 26]:

$$F_n = \frac{2}{3}k_n R^{1/2} x_{ij}^{3/2}, \quad (7)$$

where the normal deformation (1/2 the overlap between the spheres) between the neighboring grains is $x_{ij} = \frac{1}{2}[2R - |\mathbf{x}_i - \mathbf{x}_j|]$, $\mathbf{x}_{i,j}$ are the position vectors, and k_n is the normal spring constant. The latter is defined in terms of the corresponding material properties. The normal elastic constant is

$$k_n = 4G_g/(1 - \nu_g), \quad (8)$$

where G_g is the shear modulus, and ν_g is the Poisson's ratio of the material from which the grains are made.

The tangential force between neighboring grains in contact is [26]:

$$\Delta F_t = k_t(Rx_{ij})^{1/2}\Delta s, \quad (9)$$

where

$$k_t = 8G_g/(2 - \nu_g), \quad (10)$$

is the tangential spring constant, and the variable s is defined such that the relative shear displacement between the two grain centers is $2s$. Finally, Coulomb friction with interparticle friction coefficient μ imposes $F_t \leq \mu F_n$ at every contact.

From the definition of particle interactions, the elastic spring constant tensor is written in terms of the normal (N) and transverse (T) stiffnesses as:

$$\mathbf{K}_{ij} = k^N(x_{ij})\hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij} + k^T(x_{ij})[\mathbf{I} - \hat{\mathbf{d}}_{ij}\hat{\mathbf{d}}_{ij}], \quad (11)$$

where $\hat{\mathbf{d}}_{ij}$ denotes the direction of the normal displacement and we use the dyadic notation. Since we are interested in infinitesimal displacements, without loss of generality we use a linearized version of the Hertz-Mindlin force law Eqs. (7) and (9) and define the elastic stiffnesses as:

$$k^N(x_{ij}) = k_n R^{1/2} x_{ij}^{1/2}, \quad (12)$$

and

$$k^T(x_{ij}) = k_t R^{1/2} x_{ij}^{1/2}, \quad (13)$$

where k_n is the normal spring constant defined in terms of the corresponding material properties above. That is, we use the Hertz-Mindlin force law linearized about the static value of the normal compression of contacts. The resulting elastic stiffness is simply the slope evaluated at x_{ij} . The tangential force between neighboring grains in contact is defined in terms of the tangential spring constant k_t above.

The dissipative properties of the particles are defined as follows. The definition of the damping tensor $\mathbf{B}_{ij}$ involves a normal damping and a tangential damping in analogy to the definition of the elastic matrix Eq. (11). A representation of the damping matrix in terms of the first order Taylor expansion gives the following form which is an extension of the original Hertz approach assuming the material to be viscoelastic instead of elastic. The form of the damping tensor $\mathbf{B}_{ij}$ has been calculated by Kuwabara and Kono [27] and Brilliantov *et al.* [25] (see also [26] for a review):

$$\mathbf{B}_{ij} = \gamma^N(x_{ij}) \hat{\mathbf{d}}_{ij} \hat{\mathbf{d}}_{ij} + \gamma^T(x_{ij}) [\mathbf{I} - \hat{\mathbf{d}}_{ij} \hat{\mathbf{d}}_{ij}]. \quad (14)$$

If the particles, i and j , are not in contact ($|\mathbf{x}_j - \mathbf{x}_i| > 2R$), we set both $\mathbf{K}_{ij} = 0$ $\mathbf{B}_{ij} = 0$.

Both damping constants, normal $\gamma^N(x_{ij})$ and tangential $\gamma^T(x_{ij})$, are proportional to the respective elastic constants in the elastic counterparts $k^N(x_{ij})$ and $k^T(x_{ij})$. This is seen by following the calculations of Brilliantov [25] for the dissipative force between two contacting particles. The total force acting between viscoelastic particles can be derived from the total stress tensor taking into account the elastic and dissipative parts (see Landau for details [28]):

$$\hat{\sigma} = \hat{\sigma}_{(el)} + \hat{\sigma}_{(dis)}. \quad (15)$$

The calculations are simplified since the elastic and dissipative parts of the stress tensor are related in the quasi-static limit [25, 28]:

$$\hat{\sigma}_{(dis)} = \dot{x}_{ij} \frac{\partial}{\partial x_{ij}} \hat{\sigma}_{(el)}. \quad (16)$$

This leads to a dissipative force of the form:

$$F_{dis}^N = \xi k_n R^{1/2} x_{ij}^{1/2} \dot{x}_{ij}, \quad (17)$$

where ξ is the damping parameter with unit of time and related to the elastic and viscoelastic constant of the material from which the particles are made. From [28] we have:

$$\xi = \frac{1}{2} \frac{(3\eta_2 - \eta_1)^2}{(3\eta_2 + 2\eta_1)} \left[\frac{(1 + \nu_g)(1 - 2\nu_g)}{2G_g} \right], \quad (18)$$

where η_1 and η_2 are the viscous constants of the material of the particles (see Eq. (23) in [25]). Comparing with the Hertzian counterpart Eq. (7) we have the formal relation for the total force in the normal direction:

$$F_{tot}^N = \text{const} \left(x_{ij}^{3/2} + \xi x_{ij}^{1/2} \dot{x}_{ij} \right). \quad (19)$$

A similar derivation holds for the tangential components, for which we have [25]:

$$F_{dis}^T = \xi k_t R^{1/2} x_{ij}^{1/2} \dot{x}_{ij}. \quad (20)$$

These considerations leads to Eq. (14) with

$$\gamma^N(x_{ij}) = \xi k^N(x_{ij}), \quad (21)$$

and

$$\gamma^T(x_{ij}) = \xi k^T(x_{ij}). \quad (22)$$

Combining these equations we arrive at:

$$\mathbf{B}_{ij} = \xi \mathbf{K}_{ij}, \quad (23)$$

which is used later to understand the trajectories of the normal mode frequencies in the complex plane, considered as functions of ξ : $\omega_n = \omega_n(\xi)$.

The constitutive contact laws expressed by the Hertz-Mindlin theory imply the trivial scaling between the stress and strain (or volume fraction) under compression [29]:

$$\sigma \sim \epsilon^\alpha, \quad (24)$$

with $\alpha = 3/2$ for Hertz contact force law, Eq. (7), σ is the uniaxial stress and ϵ is the strain. When the constitutive particles follow other force law, the exponent α is modified accordingly. In fact, $\alpha = 3/2$ is expected for a collection of monodisperse spherical Hertzian grains, which is not an accurate description for the system we study here. Therefore, we use a mechanical testing machine to measure the stress-strain relationship exhibited by the tungsten powder and the system of glass and steel beads confined in the cup. To perform this characterization, we sit a solid stainless steel plunger on top of the powder. As demonstrated in Fig. 3a, we find a weakly non-linear stress-strain response of the tungsten powder. For the powder we use in this study, the power law exponent always falls significantly below that predicted by Eq. (24), $\alpha = 1.15 \pm 0.01$. We also test the constitutive laws for the systems of glass beads and steel beads (Fig. 3b) obtaining the α exponent summarized in Table I.

B. Theory of normal modes in a dissipative medium

In what follows, $\mathbf{x}_i$, and $\mathbf{u}_i$ denote the equilibrium position, and displacement from equilibrium, respectively, of the i -th particle. The variables θ_i represent the librational angles of the i -th particle. In addition, $\mathbf{W} = W\hat{\mathbf{z}}$ represents the displacement of the cup wall in the z -direction. In the associated

experiment, $\mathbf{W} = W\hat{\mathbf{z}}$ is 1 micron, at least three orders of magnitude smaller than R , therefore, all $\mathbf{u}_i$ are infinitesimal and the linear equation of motion for the i -th particle with mass m is:

$$-m\omega^2\mathbf{u}_i = \mathbf{K}'_{i\omega}[\mathbf{u}_i + \theta_i \times \hat{\mathbf{d}}_{i\omega} - \mathbf{W}] + \sum_j \mathbf{K}'_{ij}[\mathbf{u}_j - \mathbf{u}_i + \theta_j \times \hat{\mathbf{d}}_{ji} - \theta_i \times \hat{\mathbf{d}}_{ij}], \quad (25)$$

where, $\mathbf{d}_{ij} = (1/2)(\mathbf{x}_i - \mathbf{x}_j)$ for the case when neighboring particles are identical spheres, and the subscript, w indicates a particle interacting with the cup wall. The equations of motion for the angular variables are:

$$-\omega^2\mathbf{I}_i\dot{\theta}_i = -\mathbf{d}_{i\omega} \times \mathbf{K}'_{i\omega}[\mathbf{u}_i + \theta_i \times \hat{\mathbf{d}}_{i\omega} - \mathbf{W}] + \sum_j \mathbf{d}_{ij} \times \mathbf{K}'_{ij}[\mathbf{u}_j - \mathbf{u}_i + \theta_j \times \hat{\mathbf{d}}_{ji} - \theta_i \times \hat{\mathbf{d}}_{ij}]. \quad (26)$$

The stiffness matrix $\mathbf{K}'_{ij}$ modulates the elastic and viscous interactions between neighboring particles and is defined in terms of the elastic matrix $\mathbf{K}_{ij}$ and the damping matrix $\mathbf{B}_{ij}$ as defined above:

$$\mathbf{K}'_{ij} = \mathbf{K}_{ij} - i\omega\mathbf{B}_{ij}. \quad (27)$$

Similarly, $\mathbf{K}'_{i\omega}$ describes the elastic and viscous interaction between a particle and the wall of the cup; it is zero for particles not in contact with the wall. Next we write a compact form of these equations of motion, Eqs. (25) and (26).

The linearized equation of motion for the i -th particle in the system of N particles with mass m can be succinctly written as:

$$H_{ij}(\omega)u_j = K_{i\omega}W, \quad i, j = 1 : 6N, \quad (28)$$

where the dynamical matrix is:

$$H_{ij}(\omega) = -m\delta_{ij}\omega^2 - i\omega B_{ij} + K_{ij}. \quad (29)$$

Each term in H_{ij} accounts for the inertial, dissipative and elastic interactions at the contact between particles i and j , respectively. The vector $\{u_j\}$ represents the set of $3N$ particle displacements and $3N$ particle rotations, and $K_{i\omega}$ is the generalized spring constant connecting a particle to the walls of the cup which moves oscillatory in the z direction with amplitude W . The effective mass is obtained by inverting the matrix $\mathbf{H}$ [20]:

$$\tilde{M}(\omega) = m[H^{-1}(\omega)]_{ij}K_{j\omega}. \quad (30)$$

This result formalizes the relation between the resonance peaks in the effective mass and the normal mode frequency spectrum. The peaks observed in $\tilde{M}(\omega)$ (Fig. 2) are due to the set of normal modes, e_j^n , that are a solution to Eq. (28) when there is no forcing by the cup, $W = 0$, i.e.,

$$H_{ij}(\omega_n)e_j^n = 0. \quad (31)$$

The normal modes are those eigenvectors of $\mathbf{H}$ for which the corresponding eigenvalue is zero, and they occur at the complex-valued frequencies, ω_n .

The normal mode frequencies, ω_n , are the non-trivial solutions of Eq. (31), in which the eigenvalue is 0. Regardless the properties of the matrices $\mathbf{K}$, $\mathbf{B}$ it is a rigorous result that for values of ξ below a critical and finite value, ξ_m , all the modes are underdamped [30], meaning $\Re(\omega_n) \neq 0 \forall n$. Similarly, there is another finite, critical value, ξ_M , such that when $\xi \geq \xi_M$ all the modes are overdamped [31]: $\Re(\omega_n) \equiv 0 \forall n$. As ξ is increased from ξ_m to ξ_M each branch becomes critically damped at the values ξ_c viz: $\omega_n(\xi_c) = i\lambda_n \quad n = 1, 2, \dots$. All normal modes have the same functional dependence on ξ in the vicinity of such a critical point. Let $D(\lambda, \xi) = \det\{H_{ij}(\omega = i\lambda, \xi)\}$. In the vicinity of the critical damping point $\lambda = \lambda_{nc}$, $\xi = \xi_{nc}$ we may expand D in a Taylors series:

$$D(\lambda, \xi) = a_n(\lambda - \lambda_{nc}) + b_n(\xi - \xi_{nc}) + d_n(\lambda - \lambda_{nc})^2 + e_n(\lambda - \lambda_{nc})(\xi - \xi_{nc}) + f_n(\xi - \xi_{nc})^2 + \dots, \quad (32)$$

where

$$\begin{aligned} a_n &= \frac{\partial D}{\partial \lambda} |_{(\lambda_{nc}, \xi_{nc})} & b_n &= \frac{\partial D}{\partial \xi} |_{(\lambda_{nc}, \xi_{nc})} \\ d_n &= (1/2) \frac{\partial^2 D}{\partial \lambda^2} |_{(\lambda_{nc}, \xi_{nc})} & e_n &= \frac{\partial^2 D}{\partial \lambda \partial \xi} |_{(\lambda_{nc}, \xi_{nc})} \\ f_n &= (1/2) \frac{\partial^2 D}{\partial \xi^2} |_{(\lambda_{nc}, \xi_{nc})} \end{aligned} \quad (33)$$

The coefficients $a_n, b_n, \dots, f_n$ are all real-valued because $\{a_i^j\}$, λ_{nc} and ξ_{nc} are all real-valued. λ_{nc} is a double root of D because it represents the coalescence of two distinct complex-conjugate roots in the limit $\xi \rightarrow \xi_{nc}^-$. Accordingly, $a_n \equiv 0$. One may solve for the roots of Eq. (32):

$$\omega_n = i\lambda_n = i\lambda_{nc} \pm ig_n(\xi - \xi_{nc})^{1/2} + O(\xi - \xi_{nc})^{+1}, \quad (34)$$

where $g_n = \sqrt{-b_n/d_n}$ is real-valued because λ_n is real-valued when $\xi > \xi_{nc}$.

Since $\mathbf{H}$ is a $6N \times 6N$ matrix, there are $12N$ normal modes in this model. In practice, the effective mass can be understood in terms of a subset of these normal modes, which correspond to the modes located in the complex region of interest $[-\omega_M \leq \Re[\omega_n] \leq +\omega_M : -\omega_M \leq \Im[\omega_n] \leq 0]$, where ω_M is the maximal frequency measured in the experiment. The frequency dependence of $\tilde{M}(\omega)$ can be expressed in terms of this subset of normal modes through a pole decomposition [20] as described in the next section.

Thus, this set of equations allow for an experimental measurement of the normal mode frequencies directly from the effective mass.

IV. POLE DECOMPOSITION

The matrix $\mathbf{H}$ is complex-valued, frequency-dependent and symmetric. A non-Hermitian matrix like $\mathbf{H}$ may lack, in general, the property that its eigenvectors form a complete orthonormal basis [32]. However, according to a recent theorem by Tzeng and Wu [33], there still exists $6N$ orthonormal vectors which satisfy the following modified eigenvector problem:

$$H_{ij}e_j^n = \lambda^n e_i^{n*}, \quad (35)$$

where the eigenvalues λ^n are complex valued and the asterisk denotes conjugation. A normal mode of the system is a solution to the set of Eqs. (25) and (26) in which there is no forcing by the cup: $W = 0$. The corresponding displacements or rotations are the eigenvectors of H whose eigenvalues are zero, i.e., $\lambda^n(\omega_n) = 0$. If we expand to first order: $\lambda^n(\omega) = \alpha^n(\omega - \omega_n) + O(\omega - \omega_n)^2$, and introduce this expansion into Eq. (30), we obtain the pole expansion:

$$\tilde{M}(\omega) = \sum_n \frac{A_n}{\omega - \omega_n}, \quad (36)$$

with the residues given by

$$A_n = \frac{me_i^n(\omega_n)e_j^n(\omega_n)K_{jv}(\omega_n)}{\alpha^n}. \quad (37)$$

In reference [20] it was demonstrated that the sum Eq. (36), is an exact expression for the effective mass. This allows us to use the rational function approximation, $\tilde{M}_{RF}$ discussed in the following section to extract the normal mode frequencies from the effective mass. The matrix $\mathbf{A}_n$ are the residues of the poles representing the strength of each resonance ω_n . Using this expansion, which can be proved to be exact [20], the set of normal modes and residues $\{\omega_n, A_n\}$ can be determined from the experimental data of Fig. 2 via a search of all zeros of the rational function M^{-1} analytically continued to the complex plane. We describe this procedure in the next Section V.

V. RATIONAL FUNCTION DECOMPOSITION TO EXTRACT NORMAL MODES

We use the measured effective mass to extract the normal modes via the pole decomposition described by Eq. (36). Here, we describe the numerical procedure for generating the rational function interpolation of our data $\tilde{M}_{RF}$, which we use to determine the poles and the residues in our system.

In the experiments, we measure the effective mass $\tilde{M}(\omega_i^e)$ at a series of discrete frequencies $e = 1 : 1490$ from 100 Hz to a maximum frequency of 15 kHz. Using the reflection property, $\tilde{M}(-\omega_i^{e*}) = \tilde{M}^*(\omega_i^e)$, we extend the experimental data to negative real-valued frequencies, and we set $\tilde{M}(0) = M_0$, the static mass of the powder. So we are working with 2981 data points on the real frequency axis.

To search for the normal mode frequencies we require a complex valued function that passes through all the data points on the real axis. To this end we employ the Bulirsch-Stoer algorithm [34] to determine a rational interpolation $R_{i(i+1) \dots (i+m)}(x)$ of our data $(x_i, y_i), \dots, (x_{i+m}, y_{i+m})$. Typically [35]:

$$R_{i(i+1) \dots (i+m)}(x) = \frac{p_0 + p_1(x) + \dots + p_\mu x^\mu}{q_0 + q_1(x) + \dots + q_\nu x^\nu}, \quad (38)$$

where the coefficients of the numerator and denominator can be complex numbers. We choose to use the rational function in the recurring form:

$$R_{i(i+1) \dots (i+m)}(x) = \frac{R_{(i+1) \dots (i+m)}(x) - R_{i \dots (i+m-1)}(x)}{\left(\frac{x-x_i}{x-x_{i+m}}\right)\left(1 - \frac{R_{(i+1) \dots (i+m)}(x) - R_{i \dots (i+m-1)}(x)}{R_{(i+1) \dots (i+m)}(x) - R_{i \dots (i+m-1)}(x)}\right)}, \quad (39)$$

where, the starting points are $R_i(x) = y_i$, $R_{i-1}(x) = 0$.

The poles ω_n correspond to $M_{RF}(\omega_n) = \infty$. Therefore, we identify the poles using the condition outlined by Eq. (41), $\frac{1}{M_{RF}(\omega_1)} = 0$. We employ Muller's method [35] to identify the poles in the complex plane. Once a normal mode frequency is identified, ω_1 , we use Eq. (36), to determine the corresponding residue A_1 , $A_1 = \lim_{\omega \rightarrow \omega_1} (\omega - \omega_1) M_{RF}(\omega)$. Using the reflection property, we obtain the corresponding symmetric pole and residue which are $-\omega_1^*$ and $-A_1^*$.

The main resonance peak in $\tilde{M}(\omega)$ is often very large compared to the rest of the peaks, which makes it simple to find the first pole, but increasingly difficult to find the remaining poles by Muller's method. Thus, to find the next pole we use the difference between the original effective mass and the effective mass given by Eq. (36). That is, we define a new set of data points $\{\omega_i^e, M_i^2\}$ by

$$M_i^2(\omega_i^e) = M_i(\omega_i^e) - \frac{A_1}{\omega_i^e - \omega_1} - \frac{-A_1^*}{\omega_i^e - (-\omega_1^*)}. \quad (40)$$

This iterative process is repeated to identify all of the poles that make a notable contribution to the measured effective mass. As a test of the accuracy of this approach, we compare the real data to that from Eq. (36) utilizing all of poles and residues as shown in Fig. 4. The difference between the data and the fitting is negligible indicating that the relevant poles in the frequency range of measurement have been identified. The other systems at different stress behave similarly.

As explained above, there are $12N$ normal modes. The modes that we observe in the effective mass are those with the largest residues in the pole decomposition as expressed by Eq. (36). While it is visually apparent that only a few modes appear in the experimental effective mass, like in Fig. 2, when we subtract the pole contribution of the principal mode, then a finer structure appears, as seen in Fig. 4. The curve called Remainder in Fig. 4 appears with many (small) peaks signaling the existence of many more modes in the system. The fact that these modes are visible only after subtracting the first modes, is because their residues are very small and therefore do not contribute much to the effective mass. Thus, the effective mass is sensitive to the most important extended modes in the system as given by the visible resonance peaks. These modes are extended and define the correlation length of the system. The remaining modes are still part of the effective mass but their contribution is small.

To locate the remaining modes, we repeat the process of subtracting the pole contribution from Eq. (36) after we locate the largest pole in the remainder signal. In this way, we keep locating all the modes in the system. We estimate that we have approximately 1 million particles in the tungsten system. While it is not realistic to expect to find all the 6 million modes by this method, the pole decomposition provides a large number of the most important modes that define the effective mass in the region of observation. Indeed, there could be other modes that are outside the frequency domain of measurement and cannot be measured. Beyond this, the only limitation to locate the modes is the resolution of the signal obtained as a remainder after subtracting each pole from Eq. (36).

VI. INTERPRETATION OF EXPERIMENTAL DATA

In practice, we fit a complex valued rational function to our experimental data, $\tilde{M}_{RF}(\omega)$ to determine the set of perceivable normal modes and residues $\{\omega_n, A_n\}$ (Fig. 2). Utilizing $\tilde{M}_{RF}(\omega)$ the normal modes are identified by the criteria of Section V:

$$\frac{1}{\tilde{M}_{RF}(\omega_n)} = 0. \quad (41)$$

The locations of the complex-valued normal modes in the ω -plane ($\text{Re}[\omega_n]$, $\text{Im}[\omega_n]$) are plotted in Fig. 5 for the tungsten system at $\sigma = 44.8$ kPa. The poles are located relatively close to the real axis, indicating that the system is underdamped, $\text{Im}[\omega_n] \ll \text{Re}[\omega_n]$, which is consistent with our previous experience [20]. Furthermore, the data of Fig. 5 of the identified normal modes frequencies follow, in average, a parabolic curve in the complex plane. We have confirmed that this parabolic shape also holds for the other stress levels, however the data is more noisy. This parabolic shape is also confirmed in numerical simulations done in [36].

This result provides the basis for interpreting the experimental data. Indeed, the parabolic curve can be shown to be the result of a weakly damped system with a commutative property of the dynamical matrices: if the damping and stiffness matrices commute, $\mathbf{KB} = \mathbf{BK}$, the trajectories can be approximated by parabolas for small damping. As an illustrative example of this, let us investigate a consequence of the approximation Eq. (23) between the elastic and the damping matrices.

An important implication of Eq. (23) is that the matrices $\mathbf{K}$ and $\mathbf{B}$ commute: $\mathbf{KB} = \mathbf{BK}$, and the set $\{e_j^n\}$ are the complete eigenvectors for both matrices. The normal modes in the damped system are exactly the same as in the undamped case except that they now have complex-valued frequencies, due to the attenuation. In the presence of damping, each mode exactly decouples.

Expanding Eq. (31) we have,

$$\left[-m\delta_{ij}\omega_n^2 - i\omega_n B_{ij} + K_{ij} \right] e_j^n = 0. \quad (42)$$

When $\xi > 0$ the normal mode frequencies are complex. If we substitute the approximation Eq. (23) into Eq. (42), we have:

$$\left[-m\delta_{ij}\omega_n^2 - i\omega_n \xi K_{ij} + K_{ij} \right] e_j^n = 0. \quad (43)$$

The approximation Eq. (23) implies that e_j^n is also an eigenvector of $\mathbf{B}$, which permits us to simplify Eq. (43):

$$-\omega_n^2 - i\xi\omega_n^2\omega_n + \omega_{n0}^2 = 0, \quad (44)$$

where ω_{n0} are the normal modes of the system without damping, $\xi = 0$. Equation (44) is a quadratic equation with roots:

$$\omega_n = -i\frac{\xi}{2}\omega_{n0}^2 \pm \omega_{n0} \sqrt{1 - \left(\frac{\xi\omega_{n0}}{2}\right)^2}. \quad (45)$$

For large enough damping, $\xi > \xi_c = 2/\omega_{n0}$, these normal modes are overdamped, and the corresponding frequencies are

purely imaginary. For weak damping, $\xi < \xi_c$, the modal frequencies are damped oscillators. In this case:

$$\xi < \xi_c : |\omega_n(\xi)| = \omega_{n0}. \quad (46)$$

Thus, when Eq. (23), the underdamped modes have the property that, for a fixed $\xi \ll \xi_c$, the modes lie approximately on a parabola given by:

$$\text{Im}[\omega_n] \approx -\frac{\xi}{2} \text{Re}[\omega_n]^2. \quad (47)$$

As seen in Fig. 5, the modes exhibit a parabolic shape in the ω_n -plane, which suggests the validity of Eq. (47) in average. We fit Eq. (47) to the modal frequencies obtained experimentally in Fig. 5, and find $\xi = (2.2 \pm 0.2) \times 10^{-5} \text{ s} \ll \xi_c = 2/\omega_{n0}$, confirming that the system is weakly damped. We conclude that the scaling of the undamped normal modes can be extracted via Eq. (46), i.e. by plotting the absolute value, $|\omega_n|$ against applied stress.

VII. LOCATION OF THE MISSING MODES

At this point we would like to show how the results of our analysis can give an indication of where, in the complex plane, the remaining normal mode frequencies are located. These missing normal mode frequencies correspond to residues which are negligibly small in the measured effective mass and are not visible in the effective mass decomposition. Consider Eq. (45) which may be re-written as

$$[\Re(\omega_n)]^2 + [\Im(\omega_n) + \xi^{-1}]^2 = \xi^{-2} \quad (48)$$

Thus, if the matrices $\mathbf{B}$ and $\mathbf{K}$ commute all the normal mode frequencies, for a fixed value of the damping parameter, ξ , lie on a circle of radius $1/\xi$ centered on the point $-i/\xi$, as discussed above. In a separate effort of ours [36] we have investigated the extent to which these results may hold true with computer simulations. We have performed molecular dynamic simulations of ensembles of spherical grains in which we take the spring constants, $\mathbf{K}$, to be given by the Hertz-Mindlin theory and we have taken the damping constants, $\mathbf{B}$, to be the same for every non-zero contact. Although $\mathbf{B}$ and $\mathbf{K}$ do not commute in this case, the computed normal mode frequencies are reasonably well described by the predictions of Eq. (45) as we show in Fig. 6 which is reproduced from [36]. Specifically, for each value of the damping parameter, ξ , the set $\{\omega_n\}$ approximately lies on a circle whose radius decreases as ξ is increased, Eq. (48). The matrices $\mathbf{B}$ and $\mathbf{K}$ are mostly zero-valued, except for grains which are actually in contact with each other. In this sense we may say that they approximately commute; hence Eqs. (46) and (48) are approximately true.

We suppose these features also apply to real data on real granular media as we have already seen in Fig. 5. In order to draw some quantitative conclusions from our data we replot, in Fig. 7, some of our previously published results [20] which were deduced from the measured effective mass of tungsten

granules lightly coated with viscous PDMS fluid. The difference between the system of coated tungsten particles used in Fig. 5 and Fig. 7 is that in the later we use larger amount of silicone oil to coat the particles (80 mg as indicated). Thus, the damping in the system is increased as shown in [20]. We have fit the data to Eq. (48) using the cost function

$$\chi(R) = \sum_n |A_n| r_n^2 [\theta_n - \theta(r_n; R)]^2. \quad (49)$$

Here, $R = 1/\xi$ is the radius of the circle and the resonant frequencies are written in polar coordinates: $\omega_n = r_n \exp(i\theta_n)$. A_n are the residues of the poles in the decomposition of the effective mass data, Eq. (36); their magnitudes are indicated by the sizes of the symbols in Fig. 7. $\theta(r; R) = -\arcsin(r/2R)$ is the representation of Eq. (48) in polar coordinates. The results are shown in Fig. 7(a).

Although the existing data cover only a small arc of the circle, we feel that based on the results of our numerical simulations, and the fact that the existing data do trend along the curve, Eq. (48), we may conclude that the underdamped resonance frequencies which we are not able to directly locate with our effective mass measurements lie roughly between the bounds of the two dashed curves in Fig. 7. The overdamped resonance frequencies will lie on the negative imaginary axis, of course.

VIII. STRESS-DEPENDENCE OF THE CHARACTERISTIC FREQUENCY

The pole with the largest residue contributing to the resonance frequency at the main peak of the effective mass defines the characteristic frequency $|\omega^*(\sigma)|$. By the absolute value we indicate that this is the characteristic frequency of the damped modes obtained via the effective mass to distinguish it from the undamped ω_D^* obtained from the Debye departure in the density of states in Eq. (1). Figure 8 shows $|\omega^*(\sigma)|$ indicating a power-law dependence on σ consistent with Eq. (1) for the tungsten powder. Using an OLS estimator, we find $\Omega' = 0.15 \pm 0.02$ ($R^2 = 0.9949$). More specifically, the fitting yields:

$$|\omega^*(\sigma)| = (3.27 \pm 0.03) \sigma^{\Omega'}, \quad (50)$$

with σ measured in kPa and the frequency in kHz.

Using the stress vs strain curve in Fig. 3a with $\alpha = 1.15 \pm 0.01$, we find $\Omega = 0.17 \pm 0.02$ and $\delta = 0.10 \pm 0.02$ for the tungsten powder.

We test further the results with packings constituted by different particle types. We use spherical particles made of glass beads (1mm diameter) and steel balls (2mm diameter) without coating. Figure 8 shows the scaling of $|\omega^*|$ with stress for these systems and the exponents ($\Omega', \Omega, \delta, \alpha$) are indicated in Table I (see Fig. 3b for constitutive law). We find for glass beads $\delta = 0.14 \pm 0.02$ and for steel balls $\delta = 0.13 \pm 0.02$.

The exponent of the tungsten particles is smaller than the rest. We hypothesize that irregularities in particle shape may add some complexity in the modes not seen in spherical particles. These particles were obtained by fusing two or three

irregular (approximately cubic) particles with facets and angularities that may introduce modes not seen in spherical particles coupling the rotational and translational modes in an uncontrolled way.

We also test the scaling of the other modes obtained from the effective mass by looking at the modes with smaller residue than the main characteristic frequency. We find that the other modes scale with similar exponent as the characteristic mode $|\omega^*|$ (Fig. 9). The lack of data in the second peak for large stress is due to the fact that they fall outside of our experimental range of observation for high enough pressures. This is also corroborated by the data collapse shown in Fig. 2c and 2d. The result of the characteristic frequency following a decreasing path in stress is plotted in Fig. 10 showing that the scaling of the characteristic frequency with stress yields similar exponent as the upward path in stress shown in Fig. 8.

IX. CONCLUSIONS

Our results suggest that granular materials near jamming behave critically with a characteristic frequency defined by a critical exponent δ . The experimental exponents δ are consistently smaller than $\delta_{th} = 1/2$ predicted by theory [6, 7]. Such an anomalous exponent $\delta < 1/2$ poses new theoretical challenges to explain frictional systems with translational and rotational degrees of freedom. It might be possible that the path followed in stress space may change the value of the exponents. The usual path followed in previous works is along the line of zero shear stress, as a function of packing fraction. The present experiments follow a line of changing uniaxial stress, which would place it along a path that excites bulk and shear modes. Thus, it is plausible that the exponents might be affected by the path taken to approach the jamming transition. Another interpretation is that the lowest of our frequencies may not be a proxy for the cutoff frequency ω_D^* , which separates the region of excess density of states from the Debye region. However, if some universality could be claimed at the jamming transition, both characteristic frequencies should scale with stress with the same universal exponent. In the same line, it is interesting to note that the lowest frequency estimated from the effective mass is directly obtained from the dynamical matrix as done in ω_D^* as well.

In general, other mechanical properties such as elastic constants (shear and bulk moduli), sound speeds, and attenuation can be obtained from the effective mass measurement [18, 20]. Furthermore, the theory can be extended to non-spherical particles as well [37]. Thus, the effective mass technique facilitates a systematic test of the scaling laws of the anomalous mechanical and acoustic properties of athermal and dissipative granular systems near the jamming transition. We hope that this technique may open new experimental tests of granular matter near the jamming point.

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Material	Size	Shape	Damping	Ω'	α	$\Omega = \alpha\Omega'$	δ
Tungsten powder	$\sim 150\mu\text{m}$	irregular	coating	0.15	1.15	0.17	0.10
Glass beads	1 mm	sphere	no coating	0.21	1.25	0.26	0.14
Steel balls	2 mm	sphere	no coating	0.195	1.21	0.24	0.13
Theory linear spring [6]	–	sphere	–	1/2	1	1/2	1/2
Theory Hertz [6]	–	sphere	–	1/2	3/2	3/4	1/2

TABLE I: Summary of the measured exponents and predicted by theory.

X. APPENDIX. EXPERIMENTAL SET UP

We characterize the normal mode spectrum of the granular medium by measuring the effective mass. The main results in the text are for a granular medium made of tungsten powder that is lightly coated with silicone oil (PDMS) of viscosity 894 cP. We also present results for spherical glass beads of 1 mm diameter and spherical steel balls of 2mm. Both systems are uncoated. The preparation protocol is the same for all cases. Below, we explain the experimental procedure for tungsten particles.

The tungsten powder [19] and coating procedure [20] are discussed elsewhere. In the latter reference we found that the light PDMS coating significantly dampened or eliminated the low amplitude modes in the system. This is advantageous for the purposes of this study because it permits us to unambiguously monitor the effect of pressure on the trajectory of the dominant large amplitude modes in the system. The purpose of using tungsten as a material for the particles is to achieve a large mass of the granular system in comparison with the mass of the cup, to maximize the difference between both quantities and obtain a reliable effective mass.

After coating, ~ 100 g of the weakly wet powder is poured into a cylindrical cup (Fig. 1). The cup has internal diameter=2.54 cm, and height=3.08 cm. The sides of the cup are tapped to encourage the powder to settle, and yield a flat free surface. Finally, the system is compacted using a cyclical stress imposed with an Inkstom press of sequentially increasing then decreasing amplitude as we have previously developed [21]. The maximum stress amplitude is 118.5 kPa. It was previously shown [19] that this handling procedure suitably produces a finite sized granular medium with a reproducible effective mass which is independent on the amplitude of oscillation. The mechanical compaction protocol is discussed in detail in reference [19].

The effective mass measurement is discussed in reference [18–20]. The cylindrical cup is subjected to a vertical, sinusoidal displacement where the vibrational frequency is varied in the range 0.1 - 15 kHz. We take frequency steps of 10 Hz, and at each frequency we measure the force on the bottom of the cup, and the acceleration at two points on opposite ends of a single diameter. The mass is determined by the difference between the ratio of the force to the average acceleration, and the static mass of the cup. The effective mass is a frequency dependent complex value, owing to the partial in phase partial out of phase motion of the individual grains.

The objective of this work is to observe the effect of a uniaxial confining pressure on the dominant normal modes in our granular system. To impose a sequentially increasing stress on the granular medium, we place a thin $\sim 5 - 8$ mm urethane plug on the free surface of the powder, and screw a cap on top of the cup. After determining the orientation of the cap corresponding to initial contact with the top of the urethane, we further tighten the cap to impose a uniaxial compressive stress. Each rotation of the cap corresponds to an axial displacement that is governed by the characteristics of the threads which support the cap. To convert this displacement to a stress, we use a mechanical testing machine to charac-

terize the stress-displacement relationship of the coupled urethane/tungsten powder system in the cup.

The results of the effective mass are shown in Fig. 2. It is visually apparent from Fig. 2 that only a few modes appear in the effective mass while 12N modes are expected. The effective mass is sensitive to the extended modes in the system corresponding to collective motion of the grains. For instance, the main mode from where we obtain the characteristic frequency, is an extended mode spanning the system size. The remaining modes are more localized, correspond to modes with smaller residues, or are heavily damped and are difficult to observe them in the shape of the effective mass. However, all the 12N modes appear in the effective mass measurement. The remaining modes, being more damped, do not show easily in the shape of the effective mass. Thus, the effective mass is most effective in finding the main extended modes in the system.

Thus, modes with small residues are difficult to see in the effective mass since they are overshadowed by the extended modes with larger residues appearing as large peaks in $\text{Im}(\omega)$. This picture is corroborated in the pole analysis of Fig. 4. When we subtract the main pole contribution of the characteristic frequency $|\omega^*|$, a finer structure with many small peaks (called Remainder in Fig. 4) appears. This finer structure corresponds to the remainder modes with smaller residues. By subtracting one by one the contribution of these modes from the effective mass, in principle, we could obtain the entire subset of the 12N modes that appear in the window of observation. In practice, we do it for only the few largest modes since we are interested in the extended modes that test the criticality of the system. However, the procedure can be extended to find more modes until a given resolution preset by the experimental measurement of the effective mass. That is, one can continue extracting modes from the pole decomposition as long as the mass measurement can resolve the peaks in the Remainder of the effective mass.

Thus, while nonlinearities are inherent in the system, still the 12N modes are part of the effective mass, at least those that are within the experimental window of observation. As long as the sought-after poles lie within the rectangle $[\pm 15 \text{ kHz}, \pm i 15 \text{ kHz}]$, our procedure will determine their properties with a reasonable accuracy.

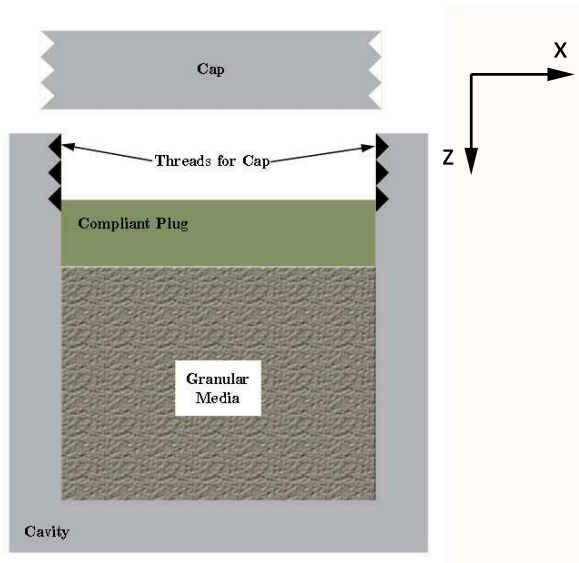


FIG. 1: (Color online) Schematic of cup used in effective mass experiments. A compliant plug is sandwiched between the tungsten powder and the cap. The uniaxial confining stress is increased by tightening the cap.

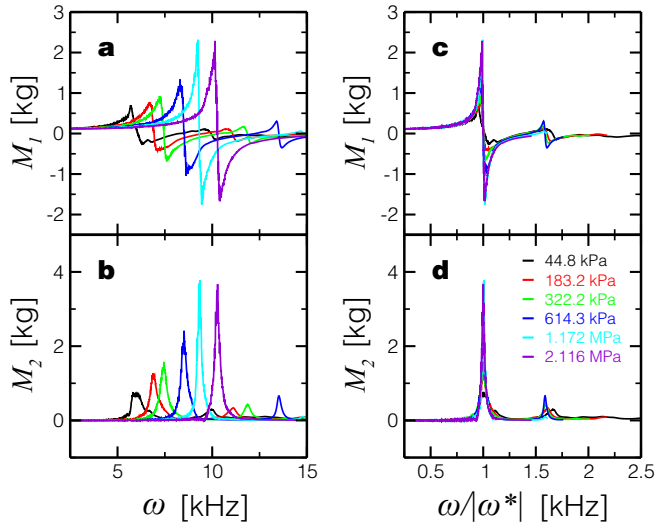


FIG. 2: (Color online) Effective mass of tungsten particles in a cup as a function of driving frequency: **(a)** Real part $M_1(\omega)$, and **(b)** Imaginary part, $M_2(\omega)$, plotted for the indicated external stresses. We calculate $\tilde{M}$ for 13 stresses between $\sigma = 44.8$ kPa and 6.39 MPa and show six curves as indicated for clarity. All datasets are available at <http://jamlab.org>. **(c)-(d)** Data collapse of $M_1(\omega)$ and $M_2(\omega)$ according to the scaling scenario Eq. (1).

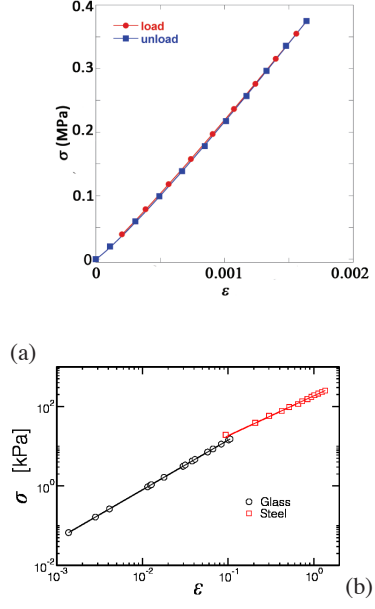


FIG. 3: (Color online) Force constitutive law. We measure the axial stress vs. strain response of (a) the tungsten powder confined in the cup and find an approximate power-law with exponent 1.15 ± 0.01 , averaged over load and unload, (b) the packing of glass beads and steel balls with exponents α given in Table I.

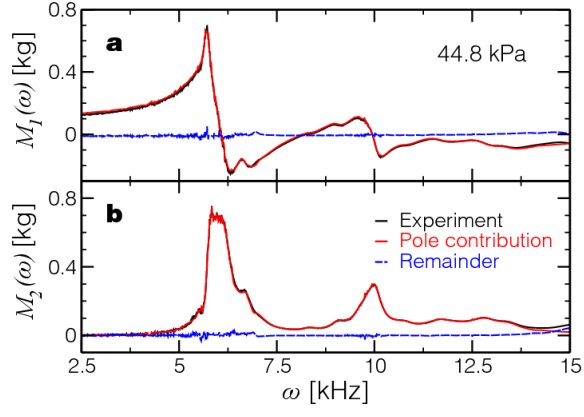


FIG. 4: (Color online) Pole decomposition. Comparison of pole decomposition using Eq. (36) and the obtained poles and experimental data at 44.8 kPa for the tungsten particles. We also plot the difference showing that many more nodes with small residues are still present in the effective mass.

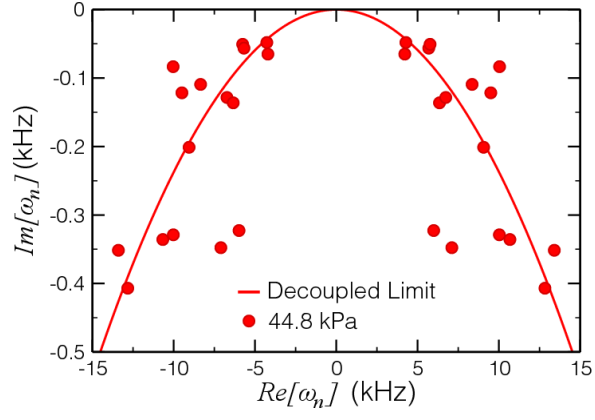


FIG. 5: (Color online) Location of the normal mode frequencies in the complex-value plane ($Re[\omega_n]$, $Im[\omega_n]$) which contribute to the pole decomposition Eq. (36) of the data plotted in Fig. 2 for $\sigma = 44.8$ kPa. Solid line represents a fit according to the parabolic Eq. (47). This equation neglects the fluctuations in the force distribution, which explains the scattering of the data around the parabolic fit. Other stresses behave similarly.

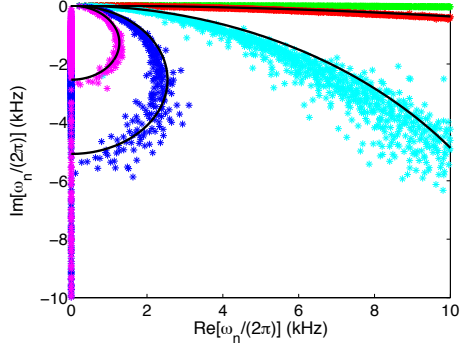


FIG. 6: (Color online) Computed complex-valued normal mode frequencies of a simulated system of 400 particles interacting via contact springs and dampers for 5 different values of the damping parameter. From Ref. [36]. Although the matrices $\mathbf{K}$ and $\mathbf{B}$ do not commute in this case, the normal mode frequencies approximately follow the predictions of Eq. (48), which is shown as a solid curve for different values of ξ .

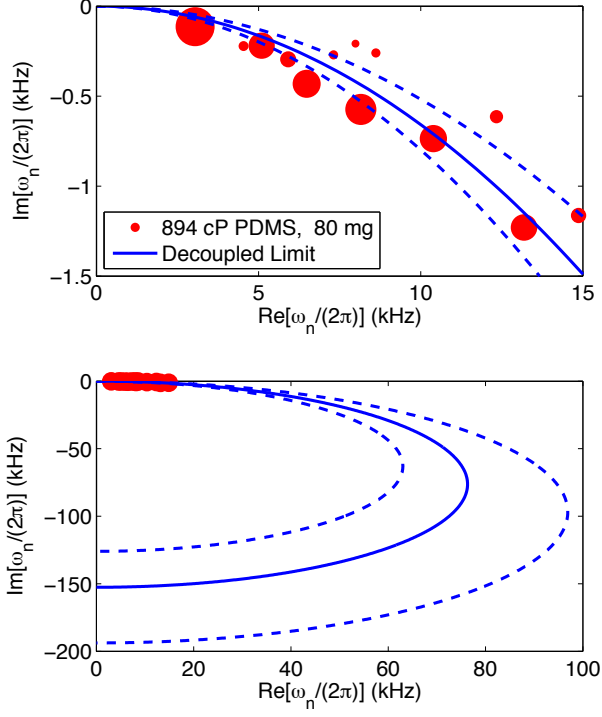


FIG. 7: (Color online) (a) Complex-valued normal mode frequencies determined from experimental data of loose granular tungsten particles, lightly coated with 80 mg of PDMS of viscosity 894 cP. From Ref [20]. This system is identical to the system of Fig. 5 except for larger amount of PDMS which increases the dissipation. The size of each symbol is proportional to the magnitude of the residue, A_n , in the decomposition of the effective mass data. The solid curve is a best fit of the data to Eq. (48). The dashed curves are for radius values corresponding to the full width at double the minimum of the cost function, Eq. (49). (b) Same as (a) but with an expanded scale. We expect the missing normal mode frequencies - those whose residues are negligibly small in the measured effective mass - to lie within these bands or on the negative imaginary axis.

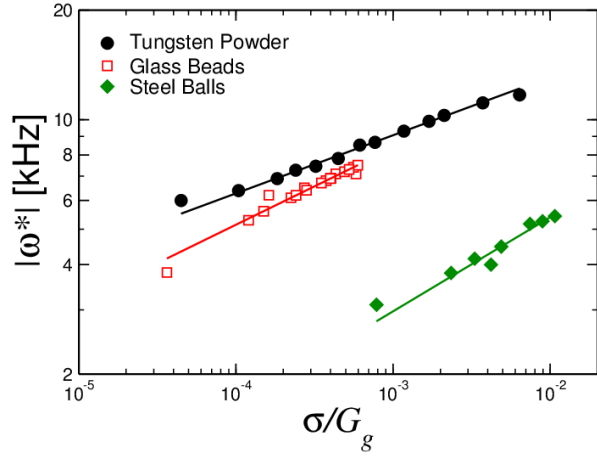


FIG. 8: (Color online) Absolute value of the characteristic frequency, $|\omega^*(\sigma)|$, corresponding to the main resonance peak of the effective mass as a function of stress σ rescaled by the shear modulus (G_g) of the material from which the particles are made. We plot results for experimental systems composed of tungsten irregular particles of $\sim 150\mu\text{m}$ coated with PDMS, uncoated spherical glass beads of 1mm, uncoated steel balls of 2mm. Solid lines are power-law fits with OLS estimator. Exponents are reported in Table I.

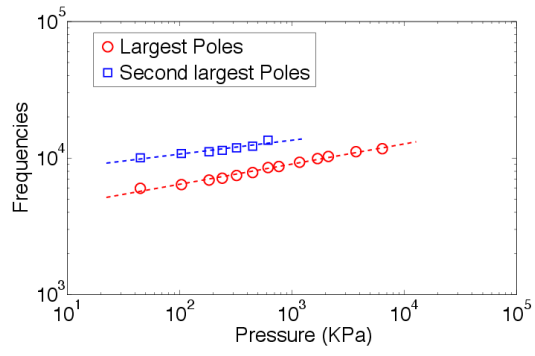


FIG. 9: (Color online) Scaling of the modes obtained from the first and second peak of the tungsten system showing similar scaling with pressure.

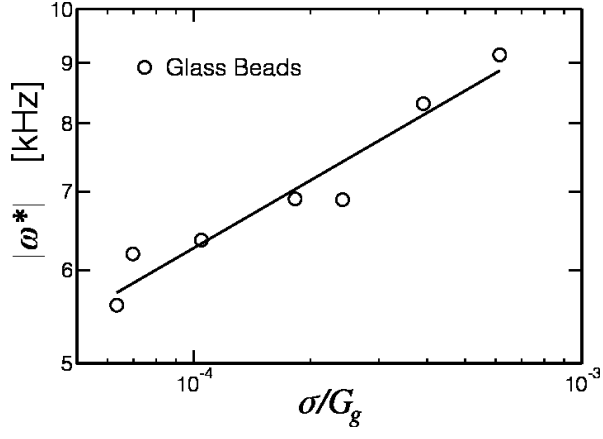


FIG. 10: Test of reversibility in the glass beads experimental system. We follow a decreasing path in pressure and obtain similar exponent $\Omega' = 0.20 \pm 0.02$ as the one obtained with the upward pressure path reported in Table I.