Angoricity and compactivity describe the jamming transition in soft particulate matter

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Abstract – 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. For dissipative jammed systems—particulate grains or droplets—a key concept is to replace the energy ensemble describing conservative systems by the volume-stress ensemble. Here, we test the applicability of the volume-stress ensemble to describe the jamming transition by comparing the jammed configurations obtained by dynamics with those averaged over the ensemble as a probe of ergodicity. Agreement between both methods suggests the idea of “thermalization” at a given angoricity and compactivity. We elucidate the thermodynamic order of the jamming transition by showing the absence of critical fluctuations in static observables like pressure and volume. The approach allows to calculate observables such as the entropy, volume, pressure, coordination number and distribution of forces to characterize the scaling laws near the jamming transition from a statistical mechanics viewpoint.

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Introduction. – A granular system compresses into a mechanically stable configuration at a nonzero pressure in response to the application of an external strain [1–3]. This process is typically referred to as the jamming transition and occurs at a critical volume fraction \( \phi_c \) [3]. 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 \( V = N V_g / \phi \) (\( \phi \) 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, \( \langle Z \rangle \), or a second-order transition with the power-law scaling of the system’s pressure as the system approaches jamming with \( \phi - \phi_c \to 0^+ \) [4–7].

For a fixed number of grains, there exist many jammed states [11] 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 pressure-volume ensemble [8,12–14] was introduced for jammed matter. In the canonical ensemble the probability of a state is given by \( \exp[-W(\partial S / \partial V) - \Gamma(\partial S / \partial \Gamma)] \), where \( S \) is the entropy of the system, \( W \) 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) [9] 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 \( X = \partial V / \partial S \) [12] and the angoricity \( A = \partial \Gamma / \partial S \) [13,15–17].

In a recent paper [18] the compactivity was used to describe frictional 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 [19,20]. We demonstrate that the jamming transition can be probed thermodynamically by the angoricity \( A \) and the compactivity \( X \).
The calculation of jamming “heat” capacities characterizes the system fluctuations and shows the lack of critical fluctuations in the static quantities as the jamming transition point is approached from above \( \phi \to \phi_c^+ \). Thus, the thermodynamical viewpoint determines the order of the phase transition and allows one to calculate the physical observables near jamming.

Following the analogy with a statistical ensemble where angoricity in a granular system plays the role of temperature, our basic hypothesis would mean that the probability of finding a jammed state at a given \( (\Gamma, \phi) \) is known, then calculations of macroscopic observables, \( \phi \) function of \( \alpha \), would be proportional to \( \text{exp}(-\alpha \Gamma) \), where the inverse angoricity is defined as \( \alpha = 1/A = \partial S/\partial \Gamma \) and \( \Gamma \) is the internal virial of this state.

Thus, 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 \( (\Gamma, \phi) \) at \( A = \infty \)) is known, then calculations of macroscopic observables, like pressure \( p \) and average coordination number \( Z \) as a function of \( \phi \), can be performed by the canonical ensemble average [9,10] at a given volume:

\[
\langle p(\alpha, \phi) \rangle_{\text{ens}} = \frac{1}{Z} \int_0^\infty p \ g(\Gamma, \phi) \ e^{-\alpha \Gamma} \ d\Gamma, \tag{1}
\]

and

\[
\langle Z(\alpha, \phi) \rangle_{\text{ens}} = \frac{1}{Z} \int_0^\infty Z \ g(\Gamma, \phi) \ e^{-\alpha \Gamma} \ d\Gamma. \tag{2}
\]

In the integrands, \( p \) and \( Z \) refer to the average values of the corresponding quantities over all configurations with a given \( \Gamma \) value. The canonical partition function is \( Z = \int_0^\infty g(\Gamma, \phi) e^{-\alpha \Gamma} d\Gamma \) and the density of states is normalized as \( \int_0^\infty g(\Gamma, \phi) d\Gamma = 1 \). Although these forms follow from a typical statistical mechanics treatment of the problem, there is no a priori proof that each jammed state should be weighed according to the Boltzmann-type factor including the angoricity. In our work we demonstrate that these equations are indeed valid, by first exhaustively measuring all possible configurations and then assigning a probability to each configuration based on the value of angoricity. In this way, we effectively calculate the value of the above integrals. We then proceed to compare the above outcome with results from Molecular-Dynamics (MD) simulations under the same conditions.

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 [4]:

\[
(p)_{\text{dyn}} \sim (\phi - \phi_c)^a, \quad (Z)_{\text{dyn}} - Z_c \sim (\phi - \phi_c)^b, \tag{3}
\]

where \( a = 1.5 \) and \( b = 0.5 \) for Hertzian spheres and \( Z_c = 6 \) is the coordination number at the isostatic point (J-point) [4]. The average \( \langle \cdots \rangle_{\text{dyn}} \) indicates that these quantities are obtained by averaging over packings generated dynamically in either simulations or as opposed to the ensemble average over configurations \( \langle \cdots \rangle_{\text{ens}} \) of eqs. (1), (2). Comparing the ensemble calculations, eqs. (1), (2), with the direct dynamical measurements, eq. (3), 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 \( g(\Gamma, \phi) \). Since \( \alpha \) is still unknown we can only calculate the average \( \langle p(\alpha, \phi) \rangle_{\text{ens}} \) for a given \( \phi \) value as a function of \( \alpha \) using eq. (1). Then, we perform MD simulations at a given \( \phi \) value, where we can now directly calculate the average pressure \( \langle p \rangle_{\text{dyn}} \). This pressure is one point of the \( \langle p(\alpha, \phi) \rangle_{\text{ens}} \) vs. \( \alpha \) curve, so that by equating the two pressures \( \langle p(\alpha, \phi) \rangle_{\text{ens}} = \langle p \rangle_{\text{dyn}} \) we finally obtain \( \alpha(\phi) \) for a given \( \phi \) value. 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. Notice that the above procedure aims to identify a common \( \alpha \) value for a direct comparison of the exhaustive ensemble results with the MD data, and the two methods are independent of each other.

The systems used here for both, ensemble generation and molecular-dynamics simulation, are the same. They are composed of 30 spherical particles in a periodic three-dimensional boundary box. The particles have same radius \( r = 5 \mu m \) and interact via a Hertz normal repulsive force without friction. The interaction is defined as: \( F_n = \frac{2}{3} k_n r^{1/2} (\delta r)^3 \), where \( \delta r = (1/2) [2r - |\vec{r}_1 - \vec{r}_2|] > 0 \) is the normal overlap and \( k_n = 4G/(1 - \nu) \) is defined in terms of the shear modulus \( G \) and Poisson’s ratio \( \nu \) of the material of which the grains are made and \( \delta = 3/2 \). Here, we use \( G = 29 \) GPa and \( \nu = 0.2 \) for spherical particles and the density of the particles, \( \rho = 2 \times 10^3 \) kg/m$^3$.

**Ensemble calculations.** – The density of jammed states \( g(\Gamma, \phi) \) is calculated in the framework of the potential energy landscape (PEL) formulation introduced by Goldstein [21] and Stillinger-Weber [22] to describe supercooled liquids. In the case of frictionless jammed systems, the mechanically stable configurations are defined as the local minima of the potential energy surface (PES) of the system [4] (see fig. 1(a) for a schematic representation).

In order to enumerate the jammed states at a given volume fraction \( \phi \), 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. In order to find such a minimum, we apply the LBFGS algorithm provided by Nocedal and Liu [23]. The procedure is analogous to finding the inherent structures [24] of glassy systems. The LBFGS algorithm is also similar to the conjugate gradient method employed by O’Hern [4]. From fig. 1(b), we see that \( g(\Gamma, \phi) \) does not change significantly for different searching days, suggesting that enough ensemble packings have been obtained to capture the features.
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Fig. 1: (Color online) (a) A schematic two-dimensional potential energy landscape surface. The jammed states A and B are local minima (zero-order saddles) in the PES. (b) The density of states \( g(\Gamma, \phi) \) for 15 days searching at \( \phi = 0.625 \). Different color corresponds to the different day. (c) The density of states \( g(\Gamma, \phi) \) as a function of internal virial \( \Gamma \) for different volume fractions, \( \phi \), ranging from 0.610 to 0.670. (d) Dependence of the results on the system size. Both the average value of \( p \) and the distribution \( g(\Gamma, \phi) \) (inset) converge as early as \( N \approx 25 \) particles.

of \( g(\Gamma, \phi) \), since the initial packings are generated by a completely random protocol. Figure 1(c) shows \( g(\Gamma, \phi) \) vs. \( \Gamma \) for different volume fractions. The presented results seem to converge for \( N \approx 25 \) and above, fig. 1(d). 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 any \( N \) value.

MD calculations. – The pressure \( \langle p \rangle_{\text{dyn}} \) as a function of \( \phi \) is calculated by performing MD simulations. Packings are prepared by compressing a gas of particles from an initial (un jammed) low volume fraction to a final jammed state. This procedure simulates a dynamical packing preparation (see [18] for details). The compression rate is \( \Gamma_0 = 5.9t_0^{-1} \), where the time is in units of \( t_0 = R\sqrt{\rho}/G \). Here, 250 independent packings, having statistically independent random initial particle positions, are obtained for each fixed pressure (see fig. 2). \( \phi = \langle \phi \rangle_{\text{dyn}} \) and \( \langle Z \rangle_{\text{dyn}} \) are flat averages of these 250 packings. We obtain (fig. 2(a), inset): \( \langle p \rangle_{\text{dyn}} = p_0(\phi - \phi_c)^{1.05} \), where \( \phi_c = 0.6077 \) is the volume fraction corresponding to the isostatic point J [4] following eq. (3) and \( p_0 = 10.8 \text{MPa} \). This critical value and the exponent, \( a = 1.65 \), are slightly different from the values obtained for larger systems (\( a = 10 \) [4]). However, our purpose is to use the same system in the dynamical calculation and the exact enumeration for a proper comparison.

Fig. 2: (Color online) The cyan \( \circ \) is \( Z_{\text{dyn}} \) and \( \phi_{\text{dyn}} \) 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.

Calculation of angoricity. – For each \( \phi \) we use \( g(\Gamma, \phi) \) to calculate \( \langle p(\alpha) \rangle_{\text{ens}} \) by eq. (1). Then, we obtain \( \alpha(\phi) \) by setting \( \langle p(\alpha, \phi) \rangle_{\text{ens}} = \langle p \rangle_{\text{dyn}} \) for every \( \phi \). Since we obtain \( g(\Gamma, \phi) \) and \( \langle p \rangle_{\text{dyn}} \) for each volume fraction \( \phi \), we can calculate the inverse angoricity \( \alpha \) by eq. (1).

Figure 3(a) shows the result of the numerical integration of eq. (1) as a function of \( \alpha \) using the numerically obtained \( g(\Gamma, \phi) \) from fig. 1(c). To obtain the value of \( \alpha \) for each \( \phi \), we input the corresponding measure of the pressure obtained dynamically \( \langle p(\phi) \rangle_{\text{dyn}} \) and obtain the value of \( \alpha(\phi) \) (see the red \( \circ \) in fig. 3(a)).

We also check the inverse angoricity \( \alpha(\phi) \) using \( g(\Gamma, \phi) \) for different searching day (see fig. 3(b)) to ensure the accuracy and convergence to the proper value. We can see that \( \alpha(\phi) \) is stable due to the fact that the density of

\[ \langle p(\alpha, \phi) \rangle_{\text{ens}} = \langle p \rangle_{\text{dyn}} \]
state, \( g(\Gamma, \phi) \), does not change significantly. The resulting equation of state \( \alpha(\phi) \) is plotted in fig. 3(c). For a volume fraction much larger than \( \phi_c \), the system’s input pressure \( \left\langle p(\phi) \right\rangle_{\text{dyn}} \) reaches the plateau at low \( \alpha \) of the function \( \left\langle p(\alpha, \phi) \right\rangle_{\text{ens}} \) (see fig. 3(a)) 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 \( \phi \) becomes large. This might explain the plateau found in \( A \) when \( (\phi - \phi_c) > 2 \times 10^{-2} \) as shown in fig. 3(d). Figure 3(d) shows that the angoricity follows a power law, near \( \phi_c \), of the form:

\[
A \sim (\phi - \phi_c)^\gamma,
\]

with \( \gamma = 2.5 \). The result is consistent with \( \gamma = \delta + 1.0 \), suggesting that \( A \propto \Gamma \propto F_n r \). 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, \( \phi_c \), 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, \( \Lambda \). 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.

**Test of ergodicity.** – In principle, using the inverse angoricity, \( \alpha \), we can calculate any macroscopic statistical observable \( \langle B(\phi) \rangle_{\text{ens}} \) at a given volume by performing the ensemble average [10]:

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

We test the ergodic hypothesis in Edwards’ ensemble by comparing eq. (5) with the corresponding value obtained with MD simulations averaged over (250) sample packings, \( B_i \), generated dynamically:

\[
\langle B(\phi) \rangle_{\text{ens}} = \frac{1}{250} \sum_{i=1}^{250} B_i.
\]

The comparison is realized by measuring the average coordination number \( \langle Z \rangle \), the average force and the distribution of interparticle forces. Figures 4(a) and (b) show that the two independent estimations of the coordination number agree very well: \( \langle Z \rangle_{\text{ens}} \approx \langle Z \rangle_{\text{dyn}} \). We calculate \( \langle F \rangle_{\text{ens}} \) and \( \langle F \rangle_{\text{dyn}} \) and find that they coincide very closely (see fig. 4(c)). The full distribution of interparticle forces for jammed systems is also an important observable which has been extensively studied in previous works [4,25,26]. The force distribution is calculated in the ensemble \( P_{\text{ens}}(F/T) \) by averaging the force distribution for every configuration in the PES. Figure 4(d) shows the distribution functions. The peak of the distribution shown in fig. 4(d) indicates that the systems are jammed [4,25,26]. Besides the exact shape of the distribution, the similarity between the ensemble and the dynamical calculations shown in fig. 4(d) is significant. The study of \( \langle Z \rangle \), \( \langle T \rangle \) and \( P(F/T) \) 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.

**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 \( \phi \). 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 5 shows \( S = \ln(\Omega(p, \phi)) \) (\( \Omega \) is the number of states which is the unnormalized version of \( g(\Gamma, \phi) \)), which is the non-equilibrium entropy of the system at the given \( p, \phi \) in phase space. By the definition of angoricity \( A = \partial T/\partial S \) and compactivity \( X = \partial V/\partial S \), we have

\[
\frac{\partial S}{\partial \ln p} = \frac{\partial S}{\partial \phi} = \Gamma \frac{\partial S}{\partial \Gamma} = \frac{c_1}{\Lambda},
\]

\[
\frac{\partial S}{\partial \ln(\phi - \phi_c)} = \frac{\partial S}{\partial \phi} = (\phi - \phi_c) \frac{\partial V}{\partial \phi} \frac{1}{\Lambda} = -\frac{c_2}{\Lambda},
\]

where \( \phi = NV_0/V \), \( c_1 = \Gamma \) and \( c_2 = (\phi - \phi_c)(NV_0/\phi^2) \).

Figure 5 indicates that the jammed system always remains at the positions of maximal entropy \( \delta S = 0 \) in the direction \((-\sin \theta, \cos \theta)\), perpendicular to the jamming
Fig. 5: (Color online) The entropy surface $S(\ln(\phi - \phi_c), \ln p)$. The color bar indicates the value of the entropy. The superimposed blue $\circ$ is $(\phi(\phi))_{\text{dyn}}$ from MD calculations as in fig. 3(a). 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 $(\ln((\phi)_{\text{dyn}} - \phi_c), \ln(p)_{\text{dyn}})$, corroborating the maximum-entropy principle.

power-law curve. In order to further analyze this result, we plot the entropy distribution along the direction $(-\sin \theta, \cos \theta)$ in figs. 6(b)–(d). We see that the entropy of the corresponding jammed states remains at the peak of the distributions along $(-\sin \theta, \cos \theta)$, verifying the maximum-entropy principle in this particular direction. We notice that some deviations are found in the vicinity of $\phi_c$. The maximization of entropy is not on $\Gamma$ or $V$ alone, but on a combination of both. This means that the entropy $S(\ln((\phi)_{\text{dyn}} - \phi_c), \ln(p)_{\text{dyn}})$ is maximum along the direction of $(-\sin \theta, \cos \theta)$ and the slope of the entropy along this direction $(-\sin \theta, \cos \theta)$ is 0 (see fig. 6(a)), that is,

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

Thus, we verify the second law of thermodynamics for jammed systems: $\delta S = 0$ at $(\ln((\phi)_{\text{dyn}} - \phi_c), \ln(p)_{\text{dyn}})$. We can use this result to obtain a relation between angularity and compactivity. We write $\ln p = \ln p_0 + a \ln(\phi - \phi_c)$ where $a$ is the exponent in eq. (3), such that $S(\ln(\phi - \phi_c), \ln p)$ is maximized at the MD measures according to the direction of $(-\sin \theta, \cos \theta)$ ($\tan \theta = a$ is the slope of the power-law curve in the log-log plot in fig. 5). Since $\delta S = 0$ at $(\ln((\phi)_{\text{dyn}} - \phi_c), \ln(p)_{\text{dyn}})$ along $(-\sin \theta, \cos \theta)$, then $(\partial S/\partial \ln(p)\cos \theta - (\partial S/\partial \ln(\phi - \phi_c))\sin \theta = 0).$ We obtain $c_1 a + c_2 \beta = 0$ and the relation between $A$ and $X$ (see fig. 6(a)):

$$X = -a A(\phi - \phi_c)/p\phi.$$  

From eqs. (4) and (10) we obtain that $X \propto -(\phi - \phi_c)^{1+a-\gamma}/\phi$ and near $\phi_c$:

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

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 [27]: the system attains a larger volume (or magnetization in spins) at $\phi_c$ when $X \to 0$ and not $X \to +\infty$ (the bounds $\phi_c \leq \phi \leq 1$ imply that the jamming point at $X \to 0^-$ is “hotter” than $X \to +\infty$. At the same time $A \to 0^+$ since the pressure vanishes).

We conclude that $A$ and $X$ alone cannot play the role of temperature. Instead, there is an actual “jamming temperature” $T_j$ that determines the direction $(-\sin \theta, \cos \theta)$ in the log-log plot of fig. 5 along the jamming equation of state. By maximizing the entropy along this direction we obtain $T_j$ as a function of $A$ and $X$:

$$T_j = \sin \theta \Gamma = \frac{a}{\sqrt{1+a^2}} A \sim (\phi - \phi_c)^{-a}.$$  

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

$$C_T \equiv \partial\Gamma/\partial A \sim (\phi - \phi_c)^{-1} A^{-2/5},$$  

$$C_V \equiv \partial V/\partial X \sim (\phi - \phi_c)^{-1} |X|^{-1/2}.$$  

From eq. (13) and eq. (14), the jamming capacities diverge at the jamming transition as $A \to 0^+$ and $X \to 0^-$. However, this result does not imply that the transition is critical since from Einstein fluctuation theory applied to pressure and volume [27] we obtain (we consider $k_B = 1$ for simplicity)

$$\langle (\Delta \Gamma)^2 \rangle = A^2 C_T \sim A^{1.6},$$  

$$\langle (\Delta V)^2 \rangle = X^2 C_V \sim |X|^{-1.5}.$$  

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Thus, the pressure and volume fluctuations near the jamming transition do not diverge, but instead vanish as $A \rightarrow 0^+$ 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 can be found at the jamming point during isotropic compression. 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 [7,28]. Such a dynamic correlation length would not appear in a purely thermodynamic static treatment as developed here. We note though that responses to shear can be treated in the present formalism by allowing the inverse anisotropy to be tensorial [10]. The intensive jamming treated in the present formalism by allowing the inverse

treatment of the jamming transition. One possible function at the mean-field level. This treatment would

impose by a shear strain or in vibrating modes [7,28].

correlated the coupling between volume and coordination

tions when accounting for the full range of fluctuations

in the liquid to the jammed phase transition from below

Thus, a critical diverging length scale might still

appear as $\phi \rightarrow \phi_c^-$ [29,30]. Our results suggest an ensemble

treatment of the jamming transition. One possible

analytical route to use this formalism would be to incor-

porate the coupling between volume and coordination

number at the particle level found in [18] together with

similar dependence for the stress to solve the partition

function at the mean-field level. This treatment would

allow analytical solutions for the observables with the

goal of characterizing the scaling law near the jamming

transition.

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