Are some packings more equal than others? A direct test of the Edwards conjecture

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In the late 1980s, Sir Sam Edwards proposed a possible statistical-mechanical framework to describe the properties of disordered granular materials. A key assumption underlying the theory was that all jammed packings are equally likely. In the intervening years it has never been possible to test this bold hypothesis directly. Here we present simulations that provide direct evidence that at the unjamming point, all packings of soft repulsive particles are equally likely, even though generically, jammed packings are not. Typically, jammed granular systems are observed precisely at the unjamming point since grains are not very compressible. Our results therefore support Edwards' original conjecture. We also present evidence that at unjamming the configurational entropy of the system is maximal.

In science, most breakthroughs cannot be derived from known physical laws: they are based on inspired conjectures [1]. Comparison with experiment of the predictions based on such a hypothesis allows us to eliminate conjectures that are clearly wrong. However, there is a distinction between testing the consequences of a conjecture and testing the conjecture itself. A case in point is Edwards' theory of granular media. In the late 1980s, Edwards and Oakeshott [2] proposed that many of the physical properties of granular materials ('powders') could be predicted using a theoretical framework that was based on the assumption that all distinct packings of such a material are equally likely to be observed. The logarithm of the number of such packings was postulated to play the same role as entropy does in Gibbs' statistical-mechanical description of the thermodynamic properties of equilibrium systems. However, statistical-mechanical entropy and granular entropy are very different objects. Until now, the validity of Edwards' hypothesis could not be tested directly – mainly because the number of packings involved is so large that direct enumeration is utterly infeasible – and, as a consequence, the debate about the Edwards hypothesis has focused on its consequences, rather than on its assumptions. Here we present results that show that now, at last, it is possible to test Edwards' hypothesis directly by numerical simulation. Somewhat to our own surprise, we find that the hypothesis appears to be correct precisely at the point where a powder is just at the (un)jamming threshold. However, at higher densities, the hypothesis fails. At the unjamming transition, the configurational entropy of jammed states appears to be at a maximum.

The concept of 'ensembles' plays a key role in equilibrium statistical mechanics, as developed by J. Willard Gibbs, well over a century ago [3]. The crucial assumption that Gibbs made in order to arrive at a tractable theoretical framework to describe the equilibrium properties of gases, liquid and solids was that, at a fixed total energy, every state of the system is equally likely to be observed. The distinction between, say, a liquid at thermal equilibrium and a granular material is that in a liquid, atoms undergo thermal motion whereas in a granular medium (in the absence of outside perturbations) the system is trapped in one of many (very many) local potential energy minima. Gibbsian statistical mechanics cannot be used to describe such a system. The great insight of Edwards was to propose that the collection of all stable packings of a fixed number of particles in a fixed volume might also play the role of an 'ensemble' and that a statistical-mechanics like formalism would result if one assumed that all such packings were equally likely to be observed, once the system had settled into a mechanically stable 'jammed' state. The nature of this ensemble has been the focus of many studies [2, 4–6].

Jamming is ubiquitous and occurs in materials of practical importance, such as foams, colloids and grains when they solidify in the absence of thermal fluctuations. Decompressing such a solid to the point where it can no longer achieve mechanical equilibrium leads to unjamming. Studies of the unjamming transition in systems of particles interacting via soft, repulsive potentials have shown that this transition is characterised by power-law scaling of many physical properties [7–12]. However, both the exact nature of the ensemble of jammed states and the unjamming transition remains unclear.

In this letter, we report a *direct test* of the Edwards conjecture, using a numerical scheme for computing basin volumes of distinct jammed states (energy minima) of N polydisperse, frictionless disks held at a constant packing fraction ϕ. Uniquely, our numerical scheme allows us to compute Ω, the number of distinct jammed states, and the individual probabilities $p_{i\in\{1,\dots,\Omega\}}$ of each observed packing to occur. Fig. 1a shows a snapshot of a section of the system, consisting of particles with a hard core and a soft shell. We obtain jammed packings by equilibrating a hard disk fluid and inflating the particles instantaneously to obtain the desired soft-disk volume fractions (ϕ), followed by energy minimization (see SI). The minimization procedure finds individual stable packings with a probability p_i proportional to the volume v_i of their basin of attraction. Averages computed using this procedure, represented by

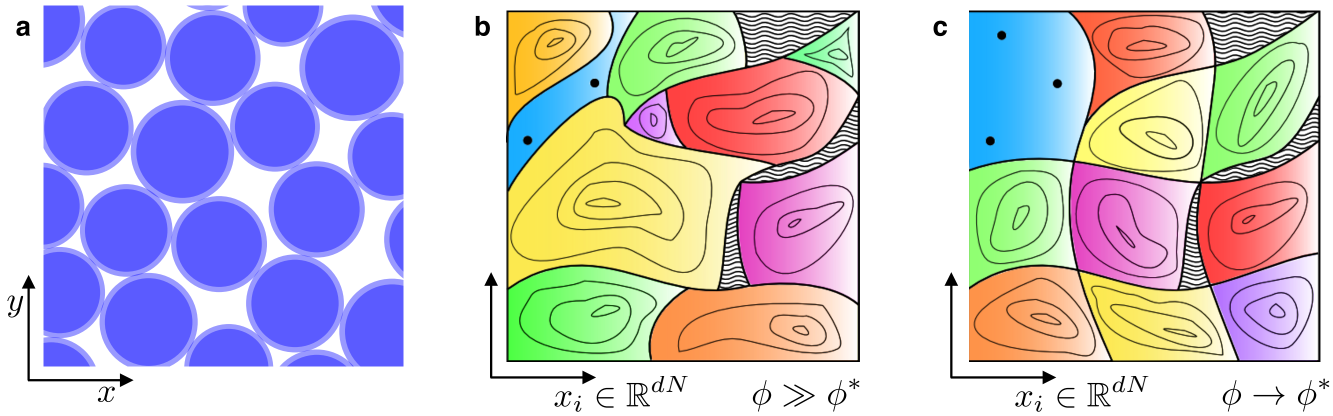

FIG. 1: (a) Snapshot of a jammed packing of disks with a hard core (dark shaded regions) plus soft repulsive corona (light shaded regions). (b)-(c) Illustration of configurational space for jammed packings. The dashed regions are inaccessible due to hard core overlaps. Single coloured regions with contour lines represent the basins of attraction of distinct minima. The dark blue region with solid dots indicates the coexisting unjammed fluid region and hypothetical marginally stable packings, respectively. The volume occupied by the fluid V_{unj} is significant only for finite size systems at or near unjamming. When $\phi \gg \phi^*$ (b) the distribution of basin volumes is broad but as $\phi \to \phi^*$ (c) the distribution of basin volumes approaches a delta function satisfying Edwards' hypothesis.

$\langle \dots \rangle_{\mathcal{B}}$, would then lead to a bias originating from the different v_i's. Recent advances in numerical methods [13, 15–17] have now enabled direct computation of v_i, and therefore, an unbiased characterization of the phase space. A summary of the technique is provided in SI.

We report a detailed analysis of the distribution of v_i for a fixed number of disks $N = 64$ (all maximum system sizes in our study were set by the current limits on computing power). We compute v_i using a thermodynamic integration scheme [13, 15–17], and compute the average basin volume $\langle v \rangle(\phi)$. The number of jammed states is, explicitly, $\Omega(\phi) = V_J(\phi)/\langle v \rangle(\phi)$, where $V_J(\phi)$ is the total available phase space volume at a given ϕ. A convenient way to check equiprobability is to compare the Boltzmann entropy $S_B = \ln \Omega - \ln N!$, which counts all packings with the same weight, and the Gibbs entropy $S_G = -\sum_i^{\Omega} p_i \ln p_i - \ln N!$ [18–20]. The Gibbs entropy satisfies $S_G \leq S_B$, saturating the bound when all p_i are equal: $p_{i \in \{1,\dots,\Omega\}} = 1/\Omega$. As shown in Fig. 2a, S_G approaches S_B from below as $\phi \to \phi^{*(S)}_{N=64} \approx 0.823$. Fig. 1b-c schematically illustrates the evolution of the basin volumes as the packing fraction is reduced.

To characterize the distribution of basin volumes, we analyse the statistics of v_i along with the pressure P_i of each packing. It is convenient to study $F_i \equiv -\ln v_i$ as a function of $\Lambda_i \equiv \ln P_i$. As shown in Fig. 2b, we observe a strong correlation between F_i and Λ_i which we quantify by fitting the data to a bivariate Gaussian distribution. The first principal component of this fit yields a linear relationship (denoted by solid lines in Fig. 2b) such that $\langle F \rangle_{\mathcal{B}}(\phi; \Lambda) \propto \lambda(\phi)\Lambda$, where $\langle F \rangle_{\mathcal{B}}(\phi; \Lambda)$ represents the average over all basins at a given Λ. Previous studies at higher packing fractions [13] indicate that this relationship is preserved in the thermodynamic limit. Defining $f = F/N$, we have (see SI for details):

$$\begin{aligned} \langle f \rangle_{\mathcal{B}}(\phi; \Lambda) =& \lambda(\phi)\Lambda + c(\phi) \\ =& \lambda(\phi)\Delta\Lambda + \langle f \rangle_{\mathcal{B}}(\phi) \ , \end{aligned} \tag{1}$$

where $\Delta\Lambda = \Lambda - \langle \Lambda \rangle_{\mathcal{B}}(\phi)$. For Edwards' hypothesis to be valid, we require that in the thermodynamic limit (i) the distribution of volumes approaches a Dirac delta, which follows immediately from the fact that the variance $\sigma_f^2 \sim 1/N$ [16] and (ii) F_i needs to be independent of Λ_i, as well as of all other structural observables, and therefore $\lambda(\phi)$ must necessarily vanish (see SI for a detailed discussion of these points). As can be seen from Fig. 2c-d, within the range of volume fractions studied, $\lambda(\phi)$ decreases but saturates to a minimum as $\phi \to \phi^{*(\lambda)}_{N=64}$. We argue below that the saturation is a finite size effect. An extrapolation using the linear regime in Fig. 2c indicates that $\lambda \to 0$ at packing fraction $\phi^{*(\lambda)}_{N=64} = 0.824 \pm 0.070$, remarkably close to where our extrapolation yields $S_G = S_B$. The analysis of basin volumes, therefore, strongly suggests that equiprobability is approached only at a characteristic packing fraction and that the vanishing of $\lambda(\phi)$ can be used to estimate the point of equiprobability.

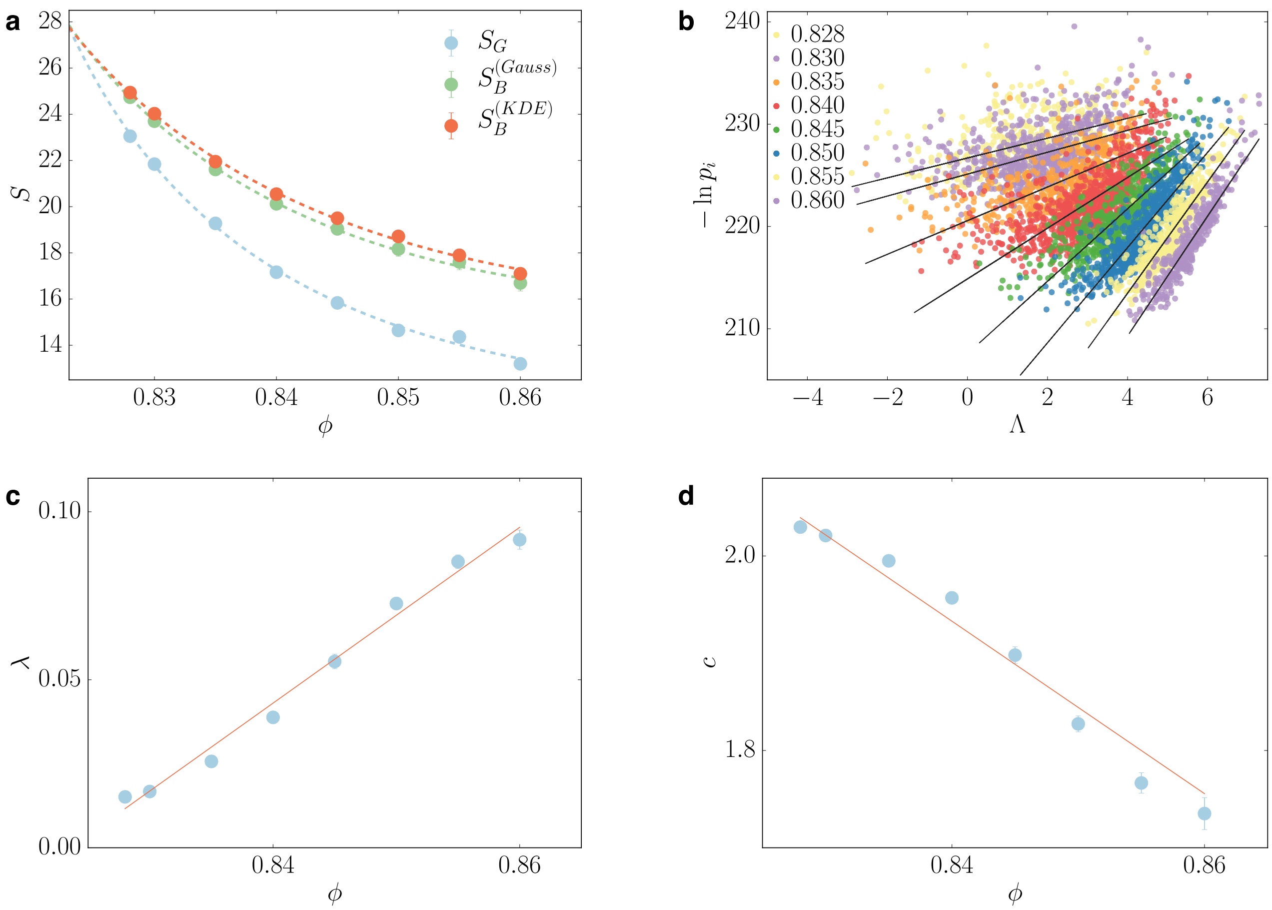

FIG. 2: (a) Gibbs entropy S_G and Boltzmann entropy S_B as a function of volume fraction. S_B is computed both parametrically by fitting $\mathcal{B}(f)$ with a generalised Gaussian function ('Gauss') and non-parametrically by computing a Kernel Density Estimate ('KDE') as in Ref. [13]. Dashed curves are a second order polynomial fit. (b) Scatter plot of the negative log-probability of observing a packing, $-\ln p_i = F_i + \ln V_J(\phi)$, where V_J is the accessible fraction of phase space (see SI) as a function of log-pressure, Λ. Black solid lines are lines of best fit computed by linear minimum mean square error using a robust covariance estimator and bootstrap (see SI). (c) Slopes $\lambda(\phi)$ and (d) intercepts $c(\phi)$ of linear fits for Eq. 1. Solid lines are lines of best fit and error bars refer to the standard error computed by bootstrap [14].

We next show that $\lambda(\phi)$ does indeed tend to zero in the thermodynamic limit. We use the fluctuations σ_f^2, σ_Λ^2, and the covariance $\sigma_{f\Lambda}^2$, obtained from the elements of the covariance matrix $\hat{\sigma} = ((\sigma_f^2, \sigma_{f\Lambda}^2), (\sigma_{f\Lambda}^2, \sigma_\Lambda^2))$ of the joint distribution of f and Λ (see SI for details), to define λ and c as:

$$\begin{aligned} \lambda(\phi) &\equiv \frac{\sigma_{f\Lambda}^2(\phi)}{\sigma_\Lambda^2(\phi)}, \\ c(\phi) &\equiv \langle f \rangle_{\mathcal{B}}(\phi) - \frac{\sigma_{f\Lambda}^2(\phi)}{\sigma_\Lambda^2(\phi)} \langle \Lambda \rangle_{\mathcal{B}}(\phi). \end{aligned} \tag{2}$$

From Fig. 2b we observe that the decrease of λ is driven by σ_Λ^2 increasing to a maximum, while σ_f^2 and $\sigma_{f\Lambda}^2$ decrease (see Fig. S2). We expect the main features of these distributions to be preserved as the system size N is increased [13], which suggests that for larger N, where basin volume calculations are still intractable for multiple densities, the maximum in σ_Λ^2 can be used to identify ϕ_N^*. We have directly measured $\chi_\Lambda = N\sigma_\Lambda^2$ using our sampling scheme – that samples packings with probability proportional to the volume of their basin of attraction – for systems of up to $N = 128$ disks (see inset of Fig. 3a) and finite size scaling indicates that χ_Λ diverges as $\phi \to \phi_{N\to\infty}^{*(\Lambda)} = 0.841(3)$ (see SI). The saturation of λ to a minimum as $\phi \to \phi_N^*$, for small N, is determined by the fact that χ_Λ only diverges in

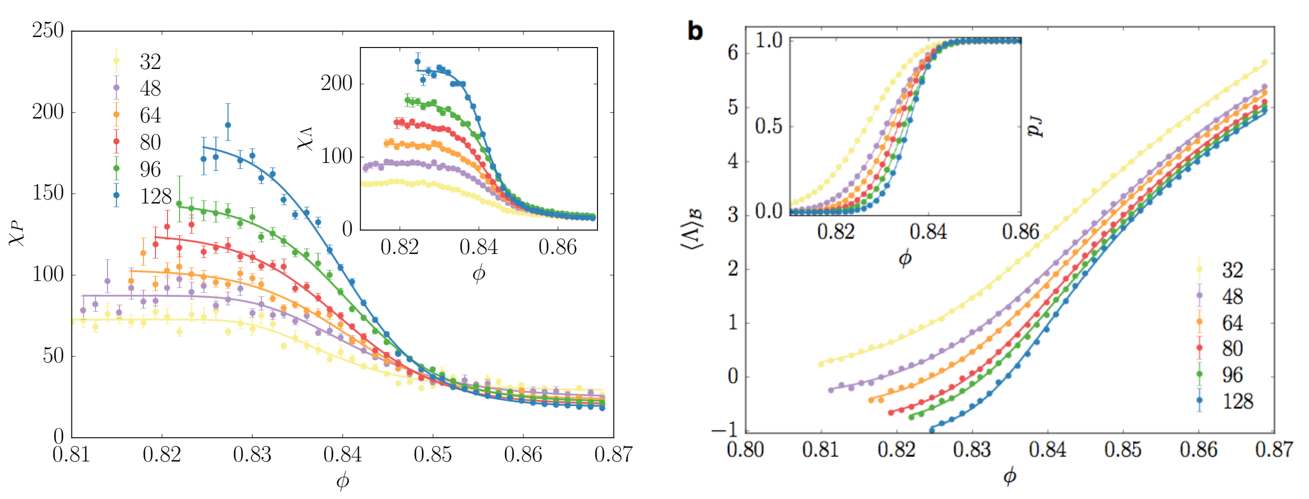

FIG. 3: (a) $\chi_P \equiv N\sigma^2(P/\langle P\rangle_{\mathcal{B}})$ and (inset) $\chi_\Lambda \equiv N\sigma^2_\Lambda$, plotted as a function of volume fraction ϕ. By finite size scaling (see SI) we show that the curves diverge in the thermodynamic limit as $\phi \to \phi^{J/*}$, implying $\phi^*_{N\to\infty} = \phi^J_{N\to\infty} = 0.841(3)$, see main text for discussion. For $\phi \gg \phi^J_N$, χ_P approaches a constant value indicating the absence of extensive correlations far from the transition. (b) Observed average log-pressure $\langle\Lambda\rangle_{\mathcal{B}}$ and (inset) probability of obtaining a jammed packings by our protocol, as a function of volume fraction ϕ. By finite size scaling (see SI) we show that $\langle\Lambda\rangle_{\mathcal{B}} \to -\infty$ as $\phi \to \phi^{J(\Lambda)}_{N\to\infty} = 0.841(3)$ and p_J collapses for $\phi \to \phi^{J(p_J)}_{N\to\infty} = 0.844(2)$, thus locating the unjamming point. Error bars, computed by BCa bootstrap [29], refer to 1σ confidence intervals. Solid lines are generalised sigmoid fits of the form $f(\phi) = a - (a-b)/(1+\exp(-w\Delta\phi))^{1/u}$. We only show values of ϕ where the probability of finding a jammed packing is at least 1%, so that the observables are computed over sufficiently large samples.

the thermodynamic limit, a detailed discussion is given in SI.

Interestingly, we find evidence that in the thermodynamic limit, the point of equiprobability $\phi^*_{N\to\infty}$, coincides with the point at which the system unjams, $\phi^J_{N\to\infty}$. We use two characteristics of the unjamming transition to locate $\phi^J_{N\to\infty}$ (i) the average pressure of the packings goes to zero, and therefore $\langle\Lambda\rangle \to -\infty$ (see Fig 3b) and (ii) the probability of finding jammed packings, p_J, goes to zero (see inset of Fig 3b). A scaling analysis indicates that $\langle\Lambda\rangle \to -\infty$ as $\phi^{J(\Lambda)}_{N\to\infty} = 0.841(3)$, and $p_J \to 0$ as $\phi^{J(p_J)}_{N\to\infty} = 0.844(2)$ (see SI). We thus find that $\phi^*_{N\to\infty} = \phi^J_{N\to\infty}$ within numerical error and up to corrections to finite size scaling [21]. Our simulations therefore lead to the surprising conclusion that the Edwards conjecture appears to hold precisely at the (un)jamming transition. We note that our earlier simulations at densities above jamming [13, 16] did not support the Edwards hypothesis. However, these simulations were carried out at densities well above jamming. The earlier simulations therefore do not contradict our current findings – although, we admit that, based on our earlier work, we did not expect to find equiprobability at the unjamming point

Why is χ_Λ related to the unjamming transition? As the particles interact via purely repulsive potentials, the pressure P is strictly positive, which implies that the fluctuations of P have a floor and go to zero at unjamming. The *relative* fluctuations $\chi_P \equiv N\sigma^2(P/\langle P\rangle_{\mathcal{B}})$, can be non-zero, and a diverging χ_P would then imply a diverging χ_Λ. Because of the bounded nature of P [22–24], however, χ_P can *only* diverge at the unjamming transition where $\langle P\rangle_{\mathcal{B}} \to 0$ (see SI). We find that χ_P does diverge (Fig. 3a) and finite size scaling yields $\phi^{*(P)}_{N\to\infty} = 0.841(3)$, in agreement with what has been found for χ_Λ. Returning to the $N = 64$ case that we have analysed using the basin volume statistics, we find that both χ_P and χ_Λ saturate to their maximum values over similar ranges of ϕ and our estimate $\phi^*_{N=64} \approx 0.824$ where $S_G = S_B$ and $\lambda \to 0$, falls in this region. In addition, the average number of contacts $\langle z\rangle_{\mathcal{B}}(\phi^*_{N=64}) = 4.050 \pm 0.24$ is close to the isostatic value $z^{(iso)}_{N=64} \equiv 2d - 2/64 \approx 3.97$ [10] (see SI).

Finally, we note that the states in the generalised Edwards ensemble [5, 25–27] characterised by ϕ and P have basin volumes that are similar, if not identical, over the full range of ϕ that we have explored (see scatter plot in Fig. 2b), indicating that equiprobability in the stress-volume ensemble [5, 25] is a more robust formulation of the Edwards hypothesis. This observation is consistent with recent experiments [28].

In conclusion, we have reported numerical evidence supporting the existence of a flat measure at unjamming for 2D soft repulsive sphere systems. Although, the equiprobability of jammed states at a given packing fraction was posited by Edwards for jammed packings of hard particles, our analysis shows that for soft particles, the Edwards hypothesis is valid only for the marginally jammed states at $\phi^*_{N\to\infty} = \phi^J_{N\to\infty}$, where the jamming probability vanishes, the entropy is maximised, and relative pressure fluctuations diverge. We have shown not only that there exist a practical 'Edwardsian' packing generation protocol, capable of sampling jammed states equiprobably, but we have uncovered an unexpected property of the energy landscape for this class of systems. At this stage, we cannot establish whether the same considerations are valid in 3D, although the already proven validity of Eq. 1 in 3D would suggest so [13]. The exact value of the entropy at unjamming, whether finite or not, also needs to be elucidated. The implications for 'soft' structural glasses is apparent: at ϕ^J the uniform size of the basins implies that the system, when thermalised, has the same probability of visiting all of its basins of attraction, hence there are no preferred inherent structures. This could be a signature of the hard-sphere transition occurring at the same point [30]. Our approach can, therefore, be extended to spin-glasses and related problems, and it would be clearly very exciting to explore the analogies and differences between 'jamming' in various systems for which the configuration space can break up into many distinct basins.

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[1] R. Laughlin, *A Different Universe: Reinventing Physics from the Bottom Down*, Basic Books, 2006.
[2] S. F. Edwards, R. B. S. Oakeshott, Physica A **157**, 180 (1991).
[3] J. W. Gibbs, *Elementary Principles of Statistical Mechanics*, Charles Scribner's sons, New York, Edward Arnold, London (1902).
[4] S. F. Edwards, D. V. Grinev, Adv. Phys. **51**, 1669 (2010).
[5] D. Bi, S. Henkes, K. E. Daniels, B. Chakraborty, Annu. Rev. Condens. Matter Phys. **6**, 63 (2015).
[6] A. Baule, F. Morone, H. Hermann, H. A. Makse, arXiv:1602.04369 (2016).
[7] P. Olsson, S. Teitel, Phys. Rev. Lett. **99**, 178001 (2007).
[8] M. Wyart, S. R. Nagel, T. A. Witten, EPL **72**, 486 (2005).
[9] L. E. Silbert, A. J. Liu, S. R. Nagel, Phys. Rev. E **73**, 041304 (2006).
[10] C. Goodrich, A. J. Liu, J. P. Sethna, Proc. Natl. Acad. Sci. **113**, 35 (2016).
[11] K. Ramola, B. Chakraborty, arXiv:1604.06148 (2016).
[12] K. Ramola, B. Chakraborty, arXiv:1609.07054, (2016).
[13] S. Martiniani, K. J. Schrenk, J. D. Stevenson, D. J. Wales, D. Frenkel, Phys. Rev. E **93**, 012906 (2016).
[14] B. Efron, Ann. Statist. **7**, 1 (1979).
[15] N. Xu, D. Frenkel, A. J. Liu, Phys. Rev. Lett. **106**, 245502 (2011).
[16] D. Asenjo, F. Paillusson, D. Frenkel, Phys. Rev. Lett. **112**, 098002 (2014).
[17] S. Martiniani, K. J. Schrenk, J. D. Stevenson, D. J. Wales, D. Frenkel, Phys. Rev. E **94**, 031301 (2016).
[18] R. H. Swendsen, Am. J. Phys. **74**, 187 (2006).
[19] D. Frenkel, Mol. Phys. **112**, 2325 (2014).
[20] M. E. Cates, V. N. Manoharan, Soft Matter **11**, 6538 (2015).
[21] D. Vagberg, D. Valdez-Balderas, M. A. Moore, P. Olsson, S. Teitel, Phys. Rev. E **83**, 030303 (2011).
[22] S. Henkes, B. Chakraborty, Phys. Rev. E **79**, 061301 (2009).
[23] G. Lois, J. Zhang, T. S. Majmudar, S. Henkes, B. Chakraborty, C. S. OHern, R. P. Behringer, Phys. Rev. E **80**, 060303 (2009).
[24] B. P. Tighe, *Force distributions and stress response in granular materials*, PhD thesis (2006).
[25] R. Blumenfeld, S. F. Edwards, J. Phys. Chem. B **113**, 3981 (2009).
[26] S. Henkes, C. S. O'Hern, B. Chakraborty, Phys. Rev. Lett. **99**, 038002 (2007).
[27] S. Henkes, B. Chakraborty, Phys. Rev. E **79**, 061301 (2009).
[28] J. G. Puckett, K. E. Daniels, Phys. Rev. Lett. **110**, 058001 (2013).
[29] B. Efron, J. Am. Stat. Assoc. **82**, 397 (1987).
[30] P. Charbonneau, J. Kurchan, G. Parisi, P. Urbani, F. Zamponi arXiv:1605.03008 (2016).