Jamming criticality of near-crystals

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Abstract – Amorphous and crystalline solids have long been considered as two distinct kinds of rigidity at the opposite ends of the order-disorder continuum. Crystals are usually treated in equilibrium with defects arising as perturbations or excitations (Ashcroft N. W. and Mermin N. D., Solid State Physics (Thomson Brooks/Cole) 1976). Amorphous solids are frustrated and out of equilibrium where preparation protocol can be important. Nevertheless the onset of rigidity of athermal amorphous matter (Liu A. J. and Nagel S. R., Nature, 396 (1998) 21) has been established as a critical point with extended universality. The universal scaling behavior that characterizes jamming at high amorphisation has been demonstrated by a wealth of simulations and models, including an infinite-dimensional mean field theory (Charbonneau P. et al., Annu. Rev. Condens. Matter Phys., 8 (2017) 265). At the other end, the crystal of minimal disorder has not been shown to display such universal behavior. Here we provide numerical evidence that slightly polydisperse crystals can become critically jammed at packing fractions extremely close to the maximum close-packed density. At the near-crystal jamming point some of the characteristic scalings are universal and agree with maximally amorphous jamming, others are novel. The set of scaling results we provide establishes jamming criticality of maximum-packing crystals in 2 and 3 dimensions.

Introduction. – Jamming, prototypically, concerns the problem where a number of particles must fit in a certain volume, touch minimally without overlapping, and be globally rigid. At the jamming critical point there is no way to improve space coverage without violating at least one of the non-overlap constraints while one contact less makes the system loose. It represents a model for the rigidity transition of athermal matter [1–4]. Jamming criticality has been found in continuous constraint satisfaction problems (CCSP) such as the single-layer perceptron [5,6] and multilayer supervised learning models [7] (see also [8]). A transition point separates a SAT phase where all constraints are satisfied, from an unsatisfiable (UNSAT) phase. At the isostatic SAT-UNSAT point all variables are exactly constrained with the number of constraints equal to the number of variables. At the packing jamming point the inter-particle contact network is isostatic as it can maintain mechanical equilibrium marginally. Randomness is necessary for isostaticity to arise, since the crystal is also a packing solution of the jamming CCSP at a higher packing fraction\(^1\) than the maximally random jammed state [3,11–13].

Jamming has been recently connected with hard sphere glasses through an infinite-dimensional mean field theory (MFT) [14,15]. Hard sphere glasses exist in two phases [16]. Stable glasses exhibit caging dynamics as they experience the width of an isolated energy metabasin. Marginally stable glasses, at higher pressure and density, display aging dynamics as they explore the fractal bottom of the metabasin. The marginal phase, also called Gardner phase, terminates at infinite pressure on a line of jamming points. Packings at the jamming line exhibit criticality with anomalous scaling exponents for the force and gap distribution [15,17–23], flat vibrational density of states (VDOS) [5,18,24–26] and characteristic normal modes that can exhibit quasi-localization [25,27]. Even though the problem is out of equilibrium where preparation protocol matters strongly [4,17] a jamming universality

\(^1\)For identical spheres the hexagonal lattice (HEX) is the maximum packing fraction configuration in 2 dimensions [9]. The face-centered cubic (FCC) lattice sets the maximum packing fraction in 3 dimensions [10].
has emerged [14,15] with an upper critical dimension $d_{uc} = 2$ [28].

Monodisperse or polydisperse packings have been brought towards a jammed state with crystalline order either through energy minimization schemes of soft spheres [29–32] or as hard spheres with molecular dynamics [3,11] and linear programming optimization protocols [12,13]. For the highly symmetric soft sphere crystals such as the hexagonal lattice (HEX) in 2d and the face-centered cubic (FCC) lattice in 3d with continuous polydispersity there is evidence of a phase of disordered crystals above the closed-packed density [31]. Some of the mechanical properties of disordered crystals approaching jamming, such as deformation moduli and contact number, exhibit numerical measurable scaling [31] that appears qualitatively consistent with an infinite-dimensional model [33].

In this work we measure the critical jamming properties of slightly polydisperse crystals that correspond to the maximum-packing periodic structures in 2d and 3d, namely HEX and FCC. At their isostatic mechanical equilibrium these highly ordered near-crystals are described by characteristic scaling exponents and functions, a subset of which are in agreement with the corresponding quantities of amorphous packings in $d \geq 2$ [23] and the amorphous ($d = \infty$) MFT description [14,15]. Another

2For details on methodology and further figures regarding the jammed near-crystal packings, the dipolar floppy modes, the buckler/non-buckler classification of forces, the gaps distribution, the normal modes, the FCC bond-orientational order, and the pair correlation function, see the Supplementary Material Supplementarymaterial.pdf (SM) [23,34,35].
results over \(N_{\text{runs}} = 9\) (18) different runs of \(N = 4096\) (2048) particles in each run.

**Near-crystal jamming properties.** – We calculate the inter-particle forces of the packing at the isostatic jamming point following the \(S\)-matrix construction (more details in [23] and the supplementary information therein). The \(S\)-matrix has dimensions \(N_c\) by \(N'd\) and is comprised of the contact unit vectors between all particles that are touching. It contains no rattlers since they are not in contact with any particle. The symmetric matrix \(N = SS'\) at isostaticity has a unique zero eigenvalue whose eigenvector contains the magnitudes of the inter-particle forces which comprise the random force network at mechanical equilibrium. The distributions of the force magnitudes exhibit power-law tails at small values (eqs. (1), (2)) a feature also seen in maximally amorphous packings [6,15,18,21,23,36]. In fig. 1 and fig. S1 of the SM one can see that the force network of isostatic near-crystals is disordered even though the underlying structure may be highly ordered. We use the dipolar floppy modes [37] to separate the forces between those whose contact is related to an extended as opposed to a localized dipolar excitation (more details in [23,32] and the supplementary information therein). An infinitesimal opening of a contact with a force dipole will induce an elementary excitation in the packing, which, if all other contacts stay the same, will not change the energy of the system. Such a dipolar floppy mode is termed extended (localized) if a large (small) fraction of particles moves appreciably [37]. In fig. S3 of the SM we plot the distribution of the median over the mean of particle displacements, \(\delta r_{ij}\), of the dipolar floppy mode of each contact. Localized (extended) forces correspond to dipolar floppy modes with median \(\{\delta r_{ij}\}\)/mean \(\{\delta r_{ij}\}\) below (above) a threshold value (see fig. S3 of the SM) and their cumulative distribution is shown in fig. 2(a) and fig. 2(b)\(^3\). The distributions of extended forces, fig. 2(a), in minimally disordered crystals for all \(s\) and both \(d = 2, 3\) are in agreement with the \(d = \infty\) MFT result [14,15]. However an exponent that decreases with decreasing polydispersity is revealed for the localized-force distributions, fig. 2(b), which tends to the MFT values for high enough amorphisation.

The distribution of the gaps, \(h = r_{ij}/\sigma_{ij} - 1\), between neighboring non-touching particles, is power-law for small forces they have power-law tails with scaling exponents that increase with polydispersity reaching the MF value of \(1 + \theta_{oc}^{MF} = 1 + 0.17462\) (continuous line) at maximum amorphisation. The probability distribution of the same data is shown in the SM, fig. S5.

\[C(f_{\text{ext}}) \sim f_{\text{ext}}^{1+\theta_{oc}},\]
\[C(f_{\text{loc}}) \sim f_{\text{loc}}^{1+\theta_{oc}},\]
\[C(h) \sim h^{1-\gamma}.\]

The exponent relations that signify mechanical stability for amorphous solids, \(1/(2 + \theta_{oc}) \leq \gamma\) and \(1 - \theta_{oc} \leq 2\gamma\) [6,15,20,21,37], appear to be violated for isostatic disordered crystals. A similar violation was observed in more complicated jammed polydisperse crystals [32]. Long-range structural correlations may need to be taken into account in a new set of inequalities for the marginal mechanical stability of jammed near-crystals.

The Hessian is the matrix of curvature in configurational space at the isostatic energy minima (inherent structures) that our packings arrive. In general it can be calculated

\(^3\)The non-buckler vs. buckler characterization of forces as in maximally amorphous packings [23] gives similar results, shown in fig. S4 in the SM.
in the Euclidean space as the matrix of second spatial derivatives of the potential energy of the packing

$$ H_{ij} = \frac{\partial U^2}{\partial x_i \partial x_j} = (S^T S)_{ij}. \quad (4) $$

At the jamming point the Hessian can be expressed through the $S$-matrix of contact vectors which is commonly called a geometric construction and is identical to the above expression at isostaticity for the harmonic interaction. The eigenvalues $\lambda$ and eigenvectors $\vec{u}(\lambda)$ of $H$ give us the frequency $\omega = \sqrt{\lambda}$ and normal modes of the packing [5,24,25,38]. The d zero-frequency modes correspond to the trivial global translations of the packing and are ignored in this analysis. The density of vibrational states for the near-crystal packing appears flat (fig. 3(a)) in both $d = 2, 3$ and clearly distinct from Debye $D(\omega) \sim \omega^{d-1}$ [29,31,32].

In order to gauge the amount of localization the Hessian eigenvectors exhibit we calculate the inverse participation ratio

$$ Y(\omega) = \frac{\sum_i N_i(\omega)^4}{\sum_i N_i(\omega)^2} \quad (5) $$

This construction for the IPR has the property $Y \sim 1/N$ for extended eigenvectors and $Y \sim 1$ for localized eigenvectors [25,27,39–42]. In studies of amorphous jamming [25] it has been found that for small frequencies the IPR of maximally amorphous solids exhibits more localization at $\varphi$ higher than $\varphi_J$ while the smallest localization can be seen near the jamming point. Our results, fig. 3(b), show that at the jamming point a more amorphised near-crystal exhibits less localization for small frequencies in accordance with the behavior of amorphous packings [25]. However, for highly ordered jammed near-crystals a dual behavior is revealed: a small number of eigenvectors appears increasingly localized as $\omega \to 0$ alongside a significantly delocalized majority which itself displays a dimension-dependent scaling with frequency, fig. 3(b). In figs. S6, S7 of the SM we plot the smallest- and largest-IPR normal mode for each polydispersity in $d = 2, 3$.

**Near-crystal crystalline properties.** — We quantify crystalline order with measures of translational and bond-orientational order appropriately chosen for HEX and FCC [31,43,44]. The translational order for both structures is measured by

$$ Q_T = \frac{1}{N} \left| \sum_{i=1}^{N} e^{i\vec{G}\cdot\vec{r}_i} \right|, \quad (6) $$

where $\vec{G}$ is a reciprocal lattice vector and $\vec{r}_i$ is the location of particle $i$. The sum is over all $N$ particles including rattlers. We perform this sum for a total of $d$ distinct reciprocal lattice vectors and average the results. A common measure of the bond-orientational order for HEX in $d = 2$ is

$$ Q_b = \frac{1}{N_b} \left| \sum_{i=1}^{N_b} e^{i\theta_i} \right|, \quad (7) $$

where $\theta_i$ is the angle the $i$-th bond makes with the $x$-axis and $N_b$ is the total number of distinct pairs of neighbors in the packing even if they are not in contact including rattlers.

In $d = 3$ a bond direction can be characterized by its overlap with the spherical harmonic $Y_{\ell m}(\theta, \phi)$,

$$ Q_{Y(\ell)} = \left[ \frac{4\pi}{2\ell + 1} \sum_{m=-\ell}^{\ell} \frac{N_b}{N_b} \sum_{i=1}^{N_b} Y_{\ell m}(\theta_i, \phi_i) \right]^{1/2}, \quad (8) $$
shown for FCC in fig. S8 of the SM [43]. In the absence of polydispersity all measures of translational and bond-orientational order match the corresponding crystal values. As polydispersity is increased all measures of order decay parabolically with $s$ [31],

$$Q_x(s) \sim Q_x(0)(1 - s^2),$$

where $x = \{T, 6, Y(\ell)\}$, see fig. 4(b) (bottom). In fig. 4(b) (top) we show that the packing fraction at jamming relates linearly with polydispersity

$$\varphi_J(s) \sim \varphi_{cp}(1 - s),$$

where $\varphi_{cp}$ is the maximum packing fraction of the perfect crystal. Combining eq. (10) with the decay of amorphisation, eq. (9), we arrive at a parabolic relation of order

$$\varphi_J(s) = \varphi_J(0)/\varphi_{cp}(0) \text{ for } x = \{T, 6, Y(\ell)\} \text{ and } \varphi_J = \varphi_J(s)/\varphi_{cp}.$$
more thoroughly [50,52] in order to be incorporated in a complete theory [33].

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