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Unjamming in models with analytic pairwise potentials

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Canonical models for studying the unjamming scenario in systems of soft repulsive particles assume pairwise potentials with a sharp cutoff in the interaction range. The sharp cutoff renders the potential nonanalytic but makes it possible to describe many properties of the solid in terms of the coordination number \( z \), which has an unambiguous definition in these cases. Pairwise potentials without a sharp cutoff in the interaction range have not been studied in this context, but should in fact be considered to understand the relevance of the unjamming phenomenology in systems where such a cutoff is not present. In this work we explore two systems with such interactions: an inverse power law and an exponentially decaying pairwise potential, with the control parameters being the exponent (of the inverse power law) for the former and the number density for the latter. Both systems are shown to exhibit the characteristic features of the unjamming transition, among which are the vanishing of the shear–to–bulk modulus ratio and the emergence of an excess of low-frequency vibrational modes. We establish a relation between the pressure–to–bulk modulus ratio and the distance to unjamming in each of our model systems. This allows us to predict the dependence of other key observables on the distance to unjamming. Our results provide the means for a quantitative estimation of the proximity of generic glass-forming models to the unjamming transition in the absence of a clear-cut definition of the coordination number and highlight the general irrelevance of nonaffine contributions to the bulk modulus.

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I. INTRODUCTION

The unjamming scenario describes the abrupt loss of solidity of gently compressed soft particles or of elastic networks that occurs when the coordination number \( z \) is reduced towards the isostatic point \( z_c = 2d \), where \( d \) is the spatial dimension. This is typically achieved in soft spheres or disks by decompressing the system such that their packing fraction \( \phi \) is reduced towards the random close packing fraction \( \phi_c \), or in elastic networks by removing interactions from the network. It is now well established that approaching the unjamming point is accompanied by the emergence of an excess of low-frequency vibrational modes [1,2], diverging correlation [3] and response [4–6] length scales, and the vanishing of elastic moduli [7]. Several claims have been made that the unjamming constitutes a nonequilibrium phase transition between disordered–solid and fluid phases of matter [8–10].

Many of the interesting phenomena associated with the unjamming transition are adequately explained by variational [11] and marginal stability [12,13] arguments and mean-field theories [14–20]. A common theme to many of these theoretical approaches is the underlying assumption that pairs of the constituent particles or degrees of freedom either interact or do not. In the vast majority of numerical investigations of the unjamming point, this assumption is embodied by the specific form of pairwise interaction potentials employed in canonical models; these are typically of the form

\[
\varphi(r) \propto \begin{cases} (r - \ell)^\alpha, & r \leq \ell \\ 0, & r > \ell \end{cases},
\]

where \( r \) is the distance between the centers of a pair of spherical particles, \( \ell \) is the sum of their radii, and \( \alpha \) is typically chosen to be 2 (harmonic interactions) or 5/2 (Hertzian interactions). Since potentials of this form have a cutoff at \( r = \ell \), the potentials are imperatively nonanalytic, but at the same time this facilitates the formulation of theoretical frameworks [2,14–17] in which contact number \( z \) plays a key role as the central order parameter.

Despite the central role of the interaction cutoff in theories of unjamming, substantial attention was drawn by the unjamming scenario following proposals that it can explain the origin of several fundamental but still elusive phenomena in atomic or molecular glassy systems, where such cutoffs in the interaction potential cannot be assumed. A few notable examples include the occurrence of the infamous boson peak—an excess of low-frequency vibrational modes over the Debye prediction—in glassy solids [16,21–23], transport properties and sound attenuation in glassy solids [16], and the fiercely debated origin of “fragility”—the degree to which relaxation is non-Arrhenius—of supercooled liquids [24,25]. This naturally leads to the questions we address in this work: (i) Can unjamming phenomena also be observed in systems with analytic pairwise potentials? (ii) Is there a general description of unjamming in such systems? (iii) How far are conventional models for glassy solids from the unjamming point?

In this work we address these questions by studying two different model systems of repulsive particles in two dimensions (2D) that interact via analytic pairwise potentials. We demonstrate that these models can be driven to the unjamming point, generally defined in systems of repulsive particles as the point at which the ratio of pressure-to-bulk modulus vanishes [16,26,27] by tuning the appropriate control parameter. In the first system, particles interact via a potential that decays exponentially with \( r \) (EXP). Similar models have been studied before, e.g., in the context of quasuniuniversality of simple liquids [28]. We find the surprising result that this system only unjams in the limit of vanishing density, and not at a finite density as the canonical models do. We also study a constant-volume system of pointlike particles interacting via...
an inverse power law $\varphi \propto r^{-\beta}$ (IPL), which is shown to unjam in the limit $\beta \to \infty$. Similar models were employed, e.g., in studies of supercooled liquids and the glass transition [29] and studies of the role of attractive forces in liquids [30]. We monitor a set of key observables as unjamming is approached: the ratio of shear-to-bulk moduli, the density of states, and characteristic vibrational frequencies. We further explain their measured scaling laws using the unjamming framework and the properties of the pairwise potentials employed.

This work is organized as follows; In Sec. II we provide details of the models investigated and of the numerical methods employed throughout our work. Section III describes the unjamming phenomenology observed in our model systems. In Section IV we provide arguments that explain the scaling laws observed approaching the unjamming point. Our work is summarized in Section V.

II. MODELS AND METHODS

As mentioned, we employ two different models of repulsive particles in 2D. In this section we spell out the details of these models and further discuss how key observables of interest are calculated. We end this section with an important discussion regarding the cutoff we introduced in the interaction range of the pairwise potentials and its role in the observed phenomena.

A. The exponential model

The first model, referred to as the exponential model, is a 50:50 binary mixture of “large” and “small” particles interacting via the pairwise potential

$$\varphi_{ij} = \begin{cases} \epsilon_{ij}(e^{-r_{ij}/l_0} + a_{ij}r_{ij} + b_{ij}), & r \leq r_c \\ 0, & r > r_c \end{cases} ,$$

where the constants $a_{ij}, b_{ij}$ are determined such that the potential and its first derivative vanish at a cutoff distance $r_c$, which was set separately for each density to be larger than the second coordination shell (see discussion at the end of this section). We discuss the importance of this cutoff and its role in the observed phenomena at the end of this section. The interaction strengths $\epsilon_{ij}$ are set to be $\epsilon_0, 1.64\epsilon_0$ and $3.05\epsilon_0$ for a small-small, small-large, or large-large interactions, respectively, where $\epsilon_0$ is our microscopic unit of energy. The interaction lengths $l_{ij}$ are set to be $l_0, 1.2l_0, \text{ and } 1.4l_0$ for small-small, small-large, and large-large interactions, respectively, where $l_0$ is our microscopic unit of length. The glass-forming ability of the EXP system is very sensitive to the particular choice of these parameters, as demonstrated in Fig. 1 and in the Supplemental Material [31]. The key control parameter in the IPL model is the exponent $\beta$, which we varied between 8 and 512.

B. The inverse power-law model

The second model employed is also a 50:50 binary mixture of “large” and “small” particles, this time interacting via the pairwise potential

$$\varphi_{ij} = \begin{cases} \epsilon_0 \left( \frac{l_{ij}}{\beta} \right)^{-\beta} + \sum_{k=0}^{3} c_{2k} \left( \frac{l_{ij}}{\beta} \right)^{2k}, & x_c \leq l_{ij} \\ 0, & l_{ij} > x_c \end{cases} ,$$

where $\epsilon_0$ is a microscopic energy scale, and the dimensionless cutoff length was set to $x_c = 1.9$, which guarantees that the first coordination shell is always within the interaction range. The interaction lengths $l_{ij}$ were set to $l_0, 1.18l_0, \text{ and } 1.4l_0$, respectively, where $l_0$ is a microscopic unit of length. The coefficients $c_{2k}$ are given by

$$c_{2k} = \frac{(-1)^{k+1}}{(6-2k)!!(2k)!!} \frac{(\beta + 6)!!}{(\beta - 2)!!(\beta + 2k)} r^{-\beta + 2k} ,$$

and ensure that the potential and three derivatives are continuous at $r_{ij}/l_{ij} = x_c$. We generated IPL solids under a fixed density of $\rho = 0.86$, which results in energies per particle of order unity, for commonly employed exponents $\beta \approx 10$ of the inverse power law. The key control parameter in the IPL model is the exponent $\beta$, which we varied between 8 and 512.

C. Interaction cutoff

In both models we introduce a cutoff in the pairwise potential for the sake of computational efficiency. This might appear to contradict the point of our work, which is to study the unjamming phenomena when such a cutoff is absent. We note here that the occurrence of unjamming phenomena in the canonical models begins to emerge when the first coordination shell starts to approach the cutoff distance of the interactions. This is never the case in our systems, as we always set the interaction cutoff such that the first coordination shell is well within the interaction range. In other words, the first coordination shell never probes the cut-off distance in any meaningful way in our numerical experiments, therefore any unjamming phenomena we observe is independent of the existence of this cutoff. We have indeed verified that eliminating the cutoff altogether has a quantitatively negligible effect on our results.
D. Sample generation

We created 1024 independent glassy samples of size \( N = 1600 \) for both the IPL and EXP systems. We verified that finite-size effects are negligible by simulating systems of \( N = 3249 \) as well, but most data are reported for \( N = 1600 \). For EXP we started by creating samples of density \( \rho = 0.56 \) by a quick quench to zero temperature from the melt and generated lower density configurations by decreasing the density by factors of \( 10^ {1/6} \), minimizing the potential energy after each such decrease. For densities lower than 2, \( \rho \), generated lower density configurations by decreasing the density by factors of \( 10^ {1} \) for the EXP system. The bare moduli are plotted in Fig. 2(a), we plot the ratio of the shear-to-bulk moduli, which is further discussed in Sec. IV. Below this crossover, \( \mu/B \) appears to vanish as \( \rho \rightarrow 0 \), which indicates the occurrence of an unjamming transition in the limit \( \rho \rightarrow 0 \). Omitting force terms in the calculation of the moduli causes the crossover to disappear altogether. The nonmonotonicity we find is reminiscent of the elastic behavior of highly compressed soft spheres, as observed in Ref. [35].

In Sec. IV we argue that in the EXP model \( \mu/B \) should scale as \( \rho^{1/4} \) as \( \rho \rightarrow 0 \); we therefore plot in Fig. 2(b) the rescaled ratio \( \eta \). The dash-dotted lines are guides to the eye, showing that the measured data are consistent with our

\[ \eta = \frac{\mu}{B} \]
prediction, although we do not yet cleanly observe this scaling in the accessible density range.

In Fig. 3 we display results for the shear and bulk moduli in the IPL system. Figure 3(a) shows the bare moduli, which appear to grow exponentially with increasing the exponent $\beta$ at fixed volume. This increase depends on our particular choice of density; upon decreasing the density, one expects eventually the opposite trend, i.e., that the moduli would decrease exponentially with $\beta$. However, since the IPL model possesses an intrinsic invariance to density changes, as previously shown in extensive work by Dyre et al. [28,36,37], the behavior of observables that are properly made dimensionless (and in particular of the ratio of shear-to-bulk moduli) does not depend on our choice of density. This point is further discussed in Sec. IV below.

Figure 3(b) shows the dependence of the ratio $\mu/B$ on the exponent $\beta$. Above a crossover at $\beta \approx 200$, we find the scaling $\mu/B \sim \rho^{-1/2}$, as predicted for the IPL system in Sec. IV. We observe the same behavior for the shadow system, albeit with an earlier crossover at around $\beta \approx 30$. Our data indicate that in the IPL system unjamming occurs in the limit $\beta \to \infty$, where $\mu/B$ presumably vanishes. We note that varying $\beta$ by a factor of 4, between $\beta = 8$ and $\beta = 32$, the shear-to-bulk modulus ratio changes by merely 20%, which is strong support of the quasiuniversality of the IPL model put forward by Dyre and coworkers [28,36,37], at least in the low-$\beta$ regime.

Interestingly, we find in both the EXP and IPL systems that the ratio of shear-to-bulk modulus is larger in the unstressed systems by a factor of $\approx 2$ on approaching the unjamming point, precisely as predicted by effective medium theory [16].

**B. Density of states**

Another hallmark of unjamming is the appearance of an excess of low-frequency vibrational modes in the density of states (DOS) as the unjamming point is approached. Here we test whether and how this observation manifests itself in our EXP and IPL model systems. In Fig. 4 we plot the DOS averaged over our ensemble of glassy samples of our two models, as a function of the rescaled frequency $\omega/\sqrt{B}$, for values of the control parameter as indicated by the legends. We note that $\sqrt{B}$ (which has the required units of frequency in two dimensions, recalling that our units of mass $m = 1$) is the natural high-frequency scale in the unjamming problem. This is because the conventional Debye frequency is defined in terms of the shear modulus, which exhibits anomalies close
DOS is observed. A pronounced enhancement of the low-frequency tails of the DOS appears as the exponent \( \beta \) in the support of the DOS as the exponent \( \beta \) approaches \( 1 \) and \( 2 \). Our data for the DOS does not allow us to reliably extract a characteristic low-frequency scale from our data of the DOS of both the EXP and IPL models. We resort therefore to extracting such a scale by different means; we follow Ref. [11] and calculate “trial modes” as the (normalized) response to a local dipolar force applied on a pair of interacting particles, as explained in Sec. II. We chose to perform this calculation on the shadow unstressed system.

Figure 5 displays our results; Fig. 5(a) shows the median \( \omega_\ast \) as a function of the control parameter. The continuous lines correspond to \( \omega_\ast / \sqrt{B} \sim \rho^{0.19} \) and \( \omega_\ast / \sqrt{B} \sim \beta^{-0.40} \) for the EXP and IPL systems, respectively.

### C. Characteristic frequency scale

As mentioned in the previous subsection, we are unable to reliably extract a characteristic low-frequency scale from our data of the DOS of both the EXP and IPL models. We resort therefore to extracting such a scale by different means; we follow Ref. [11] and calculate “trial modes” as the (normalized) response to a local dipolar force applied on a pair of interacting particles, as explained in Sec. II. We chose to perform this calculation on the shadow unstressed system.

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FIG. 4. (a) Density of states of the EXP system, at densities as indicated by the legend. (b) Density of states of the IPL system, calculated in states with exponents \( \beta \) as indicated by the legend. We left out of this plot \( D(\omega) \) obtained for the exponents \( \beta = 8 \) and \( \beta = 512 \) for visual clarity. We find that the same trend persists.

FIG. 5. Characteristic “unjamming” frequency scale \( \omega_\ast \) expressed in terms of \( \sqrt{B} \) for the EXP system (a) and the IPL system (b). Each data point represents the median calculated over 1600 responses. The insets display the bare medians of \( \omega_\ast \) vs. the control parameter. The continuous lines correspond to \( \omega_\ast / \sqrt{B} \sim \rho^{0.19} \) and \( \omega_\ast / \sqrt{B} \sim \beta^{-0.40} \) for the EXP and IPL systems, respectively.
IV. DISCUSSION

We begin with discussing the relation between the pressure–to–bulk modulus ratio \( p/B \) and the distance to the unjamming point. To this aim we spell out the expressions for \( p \) and \( B \) in the athermal limit [33], assuming the potential energy is expressed as a sum over radially symmetric pairwise interactions:

\[
p \equiv -\frac{1}{Vd} \sum_{i<j} \phi_i^r r_{ij},
\]

\[
B \equiv \frac{1}{V} \left( \sum_{i<j} \phi_i^{''} r_{ij}^2 - \mathbf{\tilde{z}}_k \cdot \mathbf{M}_{k\ell}^{-1} \cdot \mathbf{\tilde{z}}_\ell \right),
\]

where \( \mathbf{\tilde{z}}_k \equiv \sum_{i<j} \phi_i^{ij} r_{ij} \mathbf{a}^{ij}_k \) and the dipole vector \( \mathbf{a}^{ij}_k \) is defined in Eq. (11). Notice that \( g = 0 \) identically for the IPL model, which means that the second term on the right-hand side of Eq. (15), known as the “nonaffine” contribution to the bulk modulus, is identically zero in that system.

Let us focus first on the EXP system and express pairwise distances in terms of the density, namely \( \tilde{r} \equiv r / \sqrt{\rho} \). We now make the ansatz

\[
N^{-1} \sum_{i<j} \phi_i^{ij} \tilde{r}_{ij} \approx N^{-1} \sum_{i<j} \phi_i^{ij} \tilde{r}_{ij}^{2} \equiv g(\rho),
\]

where \( g(\rho) \) is an unknown function of the density. Using the ansatz in Eqs. (14) and (15), recalling that \( V \sim \rho^{-1} \), and neglecting for the moment the nonaffine term in Eq. (15), we write for the EXP system \( p = \sqrt{\rho/2} g(\rho) \) and \( B \approx g(\rho) \) (only valid in 2D with obvious generalization to 3D). From here we immediately see that

\[
p/B = \sqrt{\rho/2},
\]

as verified in Fig. 6(a), where it is shown that the scaling \( p/B \sim \sqrt{\rho/2} \) is predicted perfectly; however, the prefactor is off by roughly 15% due to the approximation made in relating \( p \) and \( B \) to the ansatz function \( g(\rho) \).

We learn from the good agreement of Eq. (17) with our numerics that neglecting the nonaffine contribution to the bulk modulus is a reasonable approximation close to unjamming. This can be justified as follows: Compare the vectors \( \mathbf{\tilde{z}}_k = \sum_{i<j} \phi_i^{ij} r_{ij} \mathbf{a}^{ij}_k \) and the net forces \( \tilde{F}_k = - \sum_{i<j} \phi_i^{ij} \mathbf{a}^{ij}_k \); the latter are identically zero due to mechanical equilibrium. Considering that in systems of purely repulsive interactions stiffnesses and forces are typically correlated, one would indeed expect that the vector \( \mathbf{\tilde{z}} \) would also be small in magnitude, resulting in a negligible nonaffine contribution to the bulk modulus.

The situation is more straightforward for the IPL system, where the nonaffine contribution to the bulk modulus vanishes identically. Here we make the ansatz

\[
V^{-1} \sum_{i<j} r_{ij}^{-\beta} \equiv f(\beta),
\]

where \( f(\beta) \) is an unknown function of the exponent \( \beta \). Using this ansatz, we write for the IPL system \( p = \frac{\beta}{2} f(\beta) \) and

\[
B = \beta(\beta + 1) f(\beta),
\]

and then we expect

\[
p/B = \frac{1}{2(\beta + 1)},
\]

as verified in Fig. 6(b).

In Fig. 7 we plot the ansatz functions \( g(\rho) \) and \( f(\beta) \), calculated using both the pressure and bulk modulus data for the EXP and IPL systems. As unjamming is approached, we find very good agreement between the two calculations for both functions, which are empirically found to fit very well the following functional forms:

\[
g(\rho) = e^{-\sqrt{0.72/\rho}},
\]

\[
f(\beta) = e^{\beta/13.6},
\]

which are represented by the continuous lines in Figs. 7(a) and 7(b), respectively.

The exponential dependence of \( g(\rho) \) on \( 1/\sqrt{\rho} \) arises naturally from the form of the interaction potential of the EXP system. The density scale \( \rho_0 \approx 0.72 \) is also consistent with the interaction length parameters \( l_i \), which were chosen to be equal or slightly larger than unity. We emphasize that the argumentation spelled out above is dimension dependent, and here we only focus on 2D.
The exponential form of $f(\beta)$ can be understood by differentiating the pressure or bulk modulus with respect to $\beta$; one finds then that the $f(\beta)$ should crucially depend on the density considered: For instance, at large densities one expects the bulk modulus and pressure to grow with increasing $\beta$, whereas for small densities the opposite behavior should occur. The scale that describes the exponential increase in $f(\beta)$, found to be approximately 14 in our system, is related to the (logarithm of) the characteristic ratio between typical pairwise distances and the interaction length parameters $l_{ij}$.

One well-known result from the unjamming literature [1,2] relates the coordination difference to the isostatic point $\delta_z \equiv z - 2d$ to the pressure–to–bulk modulus ratio as $\delta_z \sim \sqrt{\rho/B}$. We can use this relation to define an effective coordination in our systems (which lack a clear-cut definition of connectivity). For example, the canonical KABLJ system [38], in which pairwise interactions can be effectively described by a $r^{-18}$ law to a good approximation [37], would be assigned an effective $\delta_z$ of order unity.

We can further use the previously established results from the unjamming literature [1,2], $\mu/B \sim \delta_z \sim \sqrt{\rho/B}$ and $\omega_s/\sqrt{B} \sim \delta_z \sim \sqrt{\rho/B}$, to predict the dependence of the shear–to–bulk modulus ratio and the characteristic frequency scale on the distance to unjamming in our model systems. In particular, we expect

\[ \mu/B \sim \omega_s/\sqrt{B} \sim \rho^{1/4} \]  
\[ \mu/B \sim \omega_s/\sqrt{B} \sim \beta^{-1/2} \]

in the EXP system, and

in the IPL system, respectively. These relations are in good agreement with our numerical results for $\mu/B$ displayed in Figs. 2 and 3 and in reasonable consistency with our numerical results for $\omega_s/\sqrt{B}$ displayed in Fig. 5.

Finally, we comment on the correspondence between the physics of the models studied here and colloidal (Brownian) and inertial hard-sphere glasses; in the latter, the pressure and elastic moduli diverge as power laws in the distance to the jamming point [27]. In our IPL system, the pressure and elastic moduli grow exponentially with $\beta$, see, e.g., Fig. 3(a). However, there is no particular significance—in the context of unjamming—to their bare values, since the latter would depend on our particular choice of density (as discussed above), while none of the dimensionless observables we reported depend on density. We reiterate that choosing a lower density would result in exponentially decaying pressure and elastic moduli with increasing $\beta$, as observed for the EXP system on decreasing the density. For this reason, there is no direct correspondence between our IPL model and colloidal hard-sphere glasses at packing fractions approaching random close packing, in terms of the behavior of their bare pressure and elastic moduli.

It is nevertheless interesting to compare the properties of our models at their respective critical points ($\rho \to 0$ and $\beta \to \infty$) and jammed packings of hard spheres. This comparison should be made with care: Since the interaction potential we employ for the IPL model is nonadditive [39] (see model definitions in Sec. II B), and since the EXP model employs different energy parameters for different types of interactions (see model definitions in Sec. II A), we do not expect a perfect correspondence in terms of the geometry of configurations in the $\rho \to 0$ or $\beta \to \infty$ limits to jammed hard-sphere packings. These features of the interaction potentials we employed make it impossible to assign a single size to each particle and therefore would lead to configurations that do not have the precise geometry of a hard sphere packing.

V. SUMMARY

In this work we have studied the unjamming behavior of two computer model glass-forming systems of purely repulsive particles that interact via pairwise potentials with no sharp cutoffs in their respective interaction range. These models differ significantly from the canonical unjamming models, in which the sharp cutoff of the interaction range gives rise to unjamming once this cutoff probes the characteristic size of the first coordination shell of a particle (conventionally achieved by reducing the packing fraction or density). Despite the absence of a sharp cutoff in our models, we are still able to observe the hallmark phenomenology associated to the unjamming transition, in particular the vanishing of the shear–to–bulk moduli ratio, the emergence of excess low-frequency vibrational modes, and the vanishing of a characteristic frequency scale.

In the EXP model unjamming occurs in the limit $\rho \to 0$, and not at a finite density as in the canonical models. We find that the shear–to–bulk modulus ratio vanishes in good agreement with our scaling argument, which predicts $\mu/B \sim \rho^{1/4}$. We also find that the characteristic frequency scale $\omega_s/\sqrt{B}$ vanishes on unjamming as $\rho^{0.19}$, which is
close to, but not in perfect agreement with, our prediction $\omega_*/\sqrt{B} \sim \rho^{1/4}$.

In the IPL model unjamming occurs in the limit $\beta \to \infty$: We find that the shear–to–bulk modulus ratio vanishes as $\mu/B \sim \beta^{-1/2}$, in good agreement with our theoretical prediction. The characteristic frequency scale is found to follow $\omega_*/\sqrt{B} \sim \beta^{-0.40}$, not far from our prediction $\omega_*/\sqrt{B} \sim \beta^{-1/2}$.

Our predictions are based on a simple ansatz used to find the relation between the pressure–to–bulk modulus ratio and the distance to unjamming. Using a previously established result, this allows us to assign an effective excess coordination $\delta \zeta$ to each of our model systems, in which the connectivity cannot be cleanly defined. Once the dependence of the pressure–to–bulk modulus ratio is established, we use well-known results from the unjamming literature to predict the dependence of the shear–to–bulk modulus ratio and of the characteristic frequency scale (expressed in terms of the bulk modulus) on the distance to unjamming.

Our work highlights the importance of the pressure–to–bulk modulus ratio as a key dimensionless number that quantifies the distance to the unjamming point of any system with purely repulsive interactions and the generality of the insignificance of nonaffine contributions to the bulk modulus in such systems.

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[34] These derivatives should be understood as taken in the athermal limit, see Ref. [33] for further details.
[39] Non-additive interaction potentials are conventionally employed to avoid ordering and crystallization issues, see, for example, A. Widmer-Cooper and P. Harrowell, J. Chem. Phys. 135, 224515 (2011).