Nonuniversality of density and disorder in jammed sphere packings

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We show for the first time that collectively jammed disordered packings of three-dimensional monodisperse frictionless hard spheres can be produced and tuned using a novel numerical protocol with packing density ϕ as low as 0.6. This is well below the value of 0.64 associated with the maximally random jammed state and entirely unrelated to the ill-defined "random loose packing" state density. Specifically, collectively jammed packings are generated with a very narrow distribution centered at any density ϕ over a wide density range $\phi \in (0.6, 0.740\,48\ldots)$ with variable disorder. Our results support the view that there is no universal jamming point that is distinguishable based on the packing density and frequency of occurrence. Our jammed packings are mapped onto a density-order-metric plane, which provides a broader characterization of packings than density alone. Other packing characteristics, such as the pair correlation function, average contact number, and fraction of rattlers are quantified and discussed. © *2011 American Institute of Physics*.
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I. INTRODUCTION

The fundamental study of disordered jammed packings has a long history dating back at least to the pioneering work of Bernal[1,2] on the so-called random-close-packing (RCP) state. That was traditionally thought to correspond to the densest "random" packing with a unique packing fraction (or density) $\phi \approx 0.64$ for frictionless monodisperse three-dimensional (3D) spheres. That work spawned substantial research on disordered jammed packings.[3–12] About a decade ago,[7] it was argued that the RCP state is ill-defined for various reasons, including the fact that "randomness" was never quantified and the idea that a "random" packing can ever achieve a maximal density is not meaningful because one can infinitesimally increase the density of the putative RCP state, which indeed is dependent on the packing protocol and container boundaries, with imperceptible change in the amorphous pair correlation function. It was suggested that the RCP state should be replaced by the maximally random jammed (MRJ) state,[7] which is the one that minimizes a scalar order metric ψ subject to the condition of the degree of jamming.[12,13] Studies of different order metrics for 3D frictionless spheres have consistently led to a minimum at approximately the same density $\phi \approx 0.64$,[7,8,14] for collectively and strictly jammed packings in the ϕ-ψ plane (i.e., the "order map").[13] This consistency among the different order metrics speaks to the utility of the order-metric concept, even if a perfect order metric has not yet been identified. The fact that this jammed state is epitomized by maximal disorder has been advocated by other groups.[9] Bernal originally studied such jammed packings to understand the structure of liquids but it is now known that 3D MRJ monodisperse sphere packings possess quasilong-range pair correlations.[10] This property is markedly different from typical liquids with short-range interactions, which possess pair correlations decaying exponentially fast. Thus, MRJ sphere packings can be regarded to be prototypical glasses in that they possess the highest degree of disorder among all jammed packings with diverging elastic moduli.[15]

The definition of the MRJ state implies that the density ϕ alone is not sufficient to characterize jammed packings, since any packing configuration, jammed or not, is a point in the ϕ-ψ plane. This two-parameter description is but a very small subset of the relevant parameters that are necessary to fully characterize a configuration but it nonetheless enables one to draw important conclusions.[12] The frequency of occurrence of a particular configuration is irrelevant insofar as the order map is concerned, i.e., the order map emphasizes a "geometric-structure" approach to analyze packings by characterizing individual configurations, regardless of their occurrence probability.[12]

One might argue that the maximum of an appropriate "entropic" metric (based on the frequency of occurrence of the packings) would be an ideal way to characterize the randomness of a packing, and therefore, the MRJ state. However, as pointed out by Ref. 8, a substantial hurdle to overcome in implementing such an order metric is the necessity to generate all possible jammed states in an unbiased fashion using a "universal" protocol in the large-system limit, which is an intractable problem. Even if such a universal protocol could be developed, the issue of what weights to assign the resulting configurations remains. Moreover, there are other fundamental problems with entropic measures. It is well known that the lack of "frustration"[12] in two-dimensional analogs of 3D computational and experimental protocols that

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lead to putative MRJ states result in packings of monodisperse circular disks that are highly crystalline, forming rather large triangular coordination domains. Because such highly ordered packings are the most probable outcomes for these typical protocols, "entropic measures" of disorder would identify these as the most disordered, which is clearly a misleading conclusion.

On the other hand, an appropriate geometric-structure order metric is capable of identifying a particular configuration (not an ensemble of configurations) of considerably lower density. For example, a jammed vacancy-diluted triangular lattice packing (and its multidomain variant) or a jammed packing containing many small crystalline regions and "grain" boundaries that is consistent with our intuitive notions of maximal disorder possesses small scalar order metrics. However, typical packing protocols would almost never generate such disordered disk configurations because of their inherent implicit bias toward undiluted crystallization. The readers are referred to Ref. 12 for further discussion. Thus we seek to devise order metrics that can be applied to single jammed configurations, as prescribed by the geometric-structure point of view. The geometric-structure approach incorporates not only maximally dense packings (e.g., Kepler's conjecture) and random "Bernal" packings, but an infinite class of other significant jammed states not previously recognized, including "tunneled" crystals that are putatively at the jamming threshold with $\phi \approx 0.493\ 65\ldots$.[15]

Furthermore, the geometric-structure approach naturally incorporates the algorithmic variability of different packing protocols that leads to a diversity of density and disorder in jammed sphere packings.[12] In a typical numerical packing protocol, either the particle growth or system compression leads to an increase in ϕ,[3,4,16,17] which makes the particle-pair nonoverlap constraints consume larger and larger portions of the configuration space.[12] Further increase in ϕ causes the available configuration space to fracture, generating isolated "islands" that each eventually collapse into final jammed states with distinct density and structure. Presumably, any protocol would sample the disconnected regions of the configuration space with fixed probability, leading to well-defined average of any packing characteristic of interest.[12] Unless chosen to be highly restrictive, a typical packing protocol applied to a system with N particles could produce a large number of geometrically distinguishable packings with some dispersion in their ϕ and ψ values. A narrowing of the distributions of the packing characteristics with increasing N can be expected due to operation of a central limit theorem. The particular values to which the distributions individually converge are protocol-specific, meaning that these values can be controlled by choosing appropriate protocols or tuning the parameters of a protocol. Indeed, jammed packings with a diversity of density and disorder have been produced via a variety of protocols,[4,7,8,14,15,18,19] including $\phi \approx 0.64$ which is empirically the outcome of a considerable large number of laboratory experiments and numerical simulations for identical frictionless spheres. Since the jammed packings with $\phi \approx 0.64$ are apparently the "most probable states," significance has been attached to the so-called unique J (jamming) point, i.e., $\phi \approx 0.64$, which is suggested to correspond to the onset of collective jamming in soft-sphere systems.[9] However, the wide spectrum of density values [e.g., $\phi \in (0.63, 0.740\ 48\ldots)$] that has been achieved clearly suggests that conclusions drawn from any particular protocol are highly specific rather than general and thus claims of uniqueness of packing states based on their frequency of occurrence overlook the wide variability of packing algorithms and the distribution of configurations that they generate. We will elaborate on this issue in the conclusions and discussion (Sec. IV).

In this paper, we show explicitly that exploring algorithmic variability of packing protocols can lead to a diversity of density and disorder of jammed sphere packings, which is consistent with the geometric-structure approach. In particular, we demonstrate that collectively jammed packings can be generated with a narrow distribution centered at any density ϕ over a wide range $\phi \in [0.6, 0.740\ 48\ldots]$. A novel sequential linear programming (SLP) packing algorithm[20] is used to produce jammed disordered packings with ϕ as low as 0.6 for the first time, i.e., the *onset* of disordered jamming occurs well below the MRJ density of about 0.64. The Lubachevsky–Stillinger (LS) packing algorithm[3] is employed to produce jammed packings with ϕ spanning continuously from that of MRJ state ($\phi \approx 0.64$) all the way up to the face-centered-cubic (fcc) close-packed density ($\phi = 0.740\ 48\ldots$).[7,8,14] However, the standard LS algorithm as well as all previously used numerical protocols do not produce disordered collectively jammed states with ϕ well below 0.64. We control the jamming density by tuning certain parameters of the packing protocols. The jammed packings with a diversity of disorder are mapped onto a density-order-metric plane. Our results strongly support the view that there is no universal jamming point that is distinguishable based on the packing density and its frequency of occurrence. Other packing characteristics, such as the pair correlation function g_2, average contact number Z, and fraction of rattlers f_r are quantified and discussed.

II. PACKING PROTOCOLS AND CONTROL PARAMETERS

A. LS algorithm

The LS algorithm is an event-driven molecular dynamics in which particles can grow in size at a certain expansion rate $\gamma = \frac{1}{2} dD/dt$ (D is the diameter of the sphere) in addition to their thermal motion.[3] Note that γ is a key parameter that controls the jamming density.[7,10] Sufficiently small γ enables the system almost to be in equilibrium during the densification, which will finally crystallize in three dimensions into a fcc packing. For large γ, the system will quickly fall out of equilibrium and reaches a jammed state with an amorphous structure. Intermediate γ will result in various degrees of partial crystallization in the system, which leads to a continuous range of jamming densities. It is noteworthy that a very small value of γ should be used toward the jamming limit such that a true particle contact network can be formed for both crystalline and disordered packings.[12,21] We use a modified version of the LS algorithm[22] to generate jammed packings for $\phi \geq 0.64$.

B. SLP algorithm

A recently devised SLP algorithm[20] is used here to produce disordered jammed sphere packings with $\phi < 0.64$. This algorithm solves an optimization problem called the adaptive shrinking cell scheme:[16,17] jammed particle packings are generated by maximizing the packing density subject to interparticle nonoverlapping constraints. The optimization variables include particle positions as well as shape and size of the simulation box. Starting from an initial configuration, a new configuration is obtained by (locally) maximizing the density via both individual particle motions and collective motions induced by the deformation/shrinkage of the simulation box. For spheres, the objective function and constraints can be linearized for a given packing configuration and the SLP method is used to solve the optimization problem. The final packings produced by the SLP are at least collectively jammed due to its incorporation of inherent collective particle motions. Details of this algorithm and its applications for generating both disordered and maximally dense packings in high dimensional Euclidean space are given in Ref. 20.

By removing spheres from the fcc packing and its stacking variants without destroying the rigidity of the contact network, Torquato and Stillinger[15] have constructed strictly jammed "tunneled crystal" packings with ϕ as low as 0.49 365…. Similar removal procedures can be applied to MRJ packings. However the contact network of the remaining packing is generally not rigid anymore. The removed particles are required to have a large coordination number and to be mutually separated by at least a few sphere diameters. Compressing the remaining packing using the SLP algorithm leads to a rejammed configuration, with a minimum degree of structural relaxation (i.e., significant particle reconfigurations are localized around the cavities).[20] The rejammed configurations generally possess $\phi < 0.64$ and the removal-compression procedure can be repeated several times until a lower limit of ϕ is reached. The control parameters of the SLP algorithm include the initial MRJ configurations and the number of removed particles.

III. JAMMED PACKINGS WITH VARIABLE DENSITY AND DISORDER

A. Histograms for jammed packings with $\phi > 0.64$

We employ the LS algorithm to generate a large number of jammed packings of monodisperse spheres in three dimensions for $\phi \geqslant 0.64$ with $N=250$, 500, 1000, 2500, 5000, and 10 000, and a wide range of initial expansion rate $\gamma \in [10^{-2}, 10^{-6}]$. Results are verified to be at least collectively jammed,[21] using either a linear programming protocol[21] or by monitoring the instantaneous pressure of the systems for long time periods;[10] and rattlers are included to compute the reported densities. By tuning γ, the density at which the systems jam can be controlled. Figures 1(b)–1(d) contrast distributions of ϕ for two distinctly different system sizes ($N=250$ and 2500) converging onto $\phi \approx 0.64$, 0.68, and 0.72, respectively. [The case of $\phi \approx 0.60$ shown in Fig. 1(a) will be discussed separately below.] It is clear that as N increases the ϕ distributions narrow for all three mean density values. We

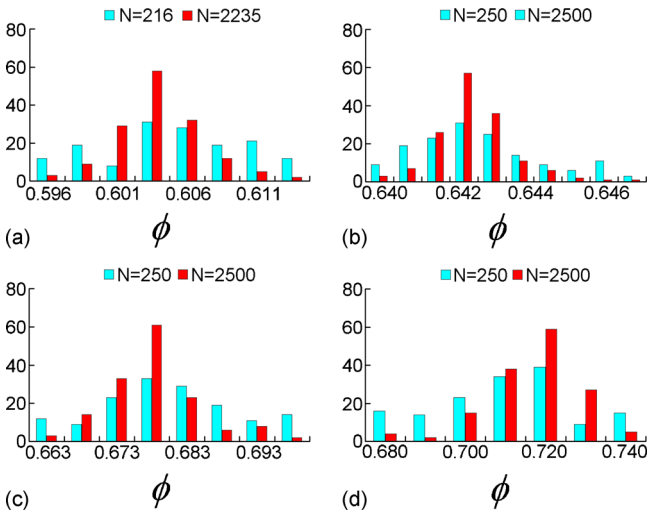

FIG. 1. (Color online) Histograms of jammed sphere packings that are centered around different mean densities. Panel (a) shows packings generated using the SLP algorithm with $\phi \approx 0.6$. Panels (b)–(d) show packings generated using the LS algorithm with $\phi \approx 0.64$, 0.68 and 0.72, respectively. The distributions become narrower as the system size increases.

also find that such narrowing becomes even more significant for $N=5000$ and 10 000. Specifically, Fig. 2 shows how the standard deviation σ of the density distribution varies with system size N for the case in which the mean $\phi \approx 0.66$. Observe that σ is a monotonically decreasing function of N and σ approximately scales $N^{-1/2}$ for large N. A similar narrowing of the ϕ distribution occurs for $\phi \approx 0.60$ shown in Fig. 1(a), and this case will be discussed separately below. Thus, one can expect that in the "thermodynamic" limit (i.e., $N \to \infty$) the jamming density will converge to a well-defined value anywhere over the interval $\phi \in [0.60, 0.740\,48\ldots]$. These results imply that any inclination to select a specific ϕ value as uniquely significant (e.g., $\phi \approx 0.64$) is primarily based on inadequate sampling of the full range of algorithmic richness and diversity that is available at least in the underlying mathematical theory of jamming.

B. Histograms for jammed packings with $\phi < 0.64$

We produce the majority of jammed packings with $\phi < 0.64$ using the SLP algorithm. MRJ packings generated via

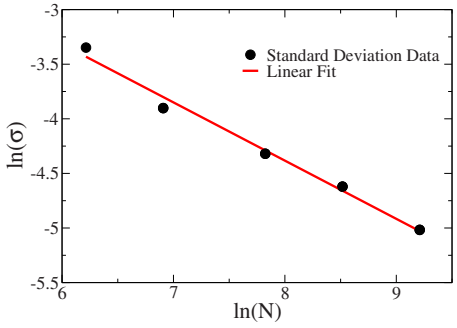

FIG. 2. (Color online) The standard deviation σ of the density distribution with mean $\phi \approx 0.66$ as a function of system size N. System sizes $N=500$, 1000, 2500, 5000, and 10 000 are used here. The linear fit for ln σ vs ln N gives a slope $k \approx -0.533$, which indicates that σ approximately scales as $N^{-1/2}$ for large N. Such a σ-N relation is also observed for density distributions with the other mean values studies here.

the LS algorithm are used as initial configurations. Each time approximately $f_s=0.1\%-2.5\%$ of the spheres are removed from the initial packing and the remaining spheres are compressed to a jammed state using the SLP algorithm. Such a procedure is repeated $n_r=5-10$ times before a lower limit on ϕ is reached. The fraction f_s of removed spheres is decreased as the limit is approached. We stress that our packings with $\phi<0.64$ are not so-called random loose packings,[23] which are not even collectively jammed.[21] A few packings with $\phi\approx0.62$ are generated using the LS algorithm with open simple-cubic lattice packings as initial configurations.[8]

Figure 1(a) contrasts ϕ distributions for two distinctly different system sizes ($N=216$ and 2235) converging onto $\phi\approx0.60$. It can be seen that as N increases the ϕ distributions narrow. Such narrowing is also observed for other convergence densities within the interval (0.6, 0.64). We note that it is very difficult to produce jammed packings with ϕ significantly lower than 0.6 using the SLP algorithm (though removing the rattlers in the packing results in a slightly lower density $\sim$0.595), while it has been rigorously shown that strictly jammed sphere packings (e.g., tunneled crystals and the associated stacking variants) can possess ϕ as low as $\sqrt{2}\pi/9=0.493\,65\ldots$.[15] However, jammed packings that combine the tunneled crystals and MRJ packings can be constructed. In particular, stackings of layers of honeycomb-lattice packings of spheres stabilized with triangular-lattice layers on top and bottom are inserted into the MRJ packings. These "layered" packings are then compressed to jamming using the SLP algorithm. This construction enables one to obtain jammed packings with variable disorder within the density range $\phi\in(0.49,0.64)$.

C. Order metrics and other packing characteristics for jammed packings with $\phi\in[0.49,0.74]$

It has been established that a variety of different useful order metrics are positively correlated[7,8,14] and hence any one of them can be used to characterize the packings. To quantify the order of the packings, we compute the translational order metric $\mathfrak{T}$,[7] defined as

$$\mathfrak{T}=\left|\sum_{i}^{N_s}(n_i-n_i^{\text{ideal}})/\sum_{i}^{N_s}(n_i^{\text{FCC}}-n_i^{\text{ideal}})\right|, \tag{1}$$

where n_i is the average occupation number for the shell i centered at a distance from a reference sphere that equals the ith nearest-neighbor separation for the open FCC lattice at that density and N_s is the total number of shells for the summation ($N_s=45$ is used here for $N\sim2500$); n_i^{ideal} and n_i^{FCC} are the corresponding shell occupation numbers for an ideal gas (spatially uncorrelated spheres) and the open FCC lattice. For a completely disordered system (e.g., a Poisson distribution of points) $\mathfrak{T}=0$, whereas $\mathfrak{T}=1$ for the FCC lattice.

Figure 3 shows the ϕ-$\mathfrak{T}$ plane on which representative jammed packings with $N\sim2500$ and $\phi\in[0.49,0.74]$ are mapped. The packings generated using the LS algorithm are shown as black circles. Note that for these packings each ϕ is associated with a range of $\mathfrak{T}$ values. Moreover, increasing ϕ from the MRJ state can be achieved at the cost of decreasing the degree of disorder, as pointed out in Ref. 7. Further

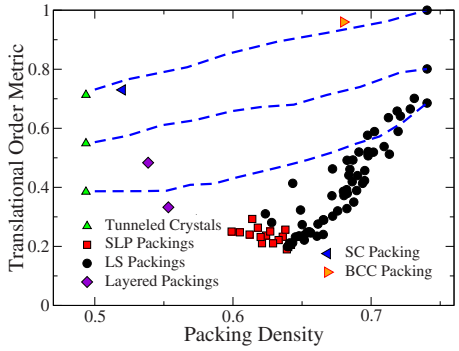

FIG. 3. (Color online) Order map for jammed sphere packings with $N\sim2500$: translational order metric $\mathfrak{T}$ vs packing density ϕ. The dashed blue lines (which are consistent with the qualitative trends indicated in the order maps discussed in Ref. 12) show the spectrum of packings generated by randomly filling the vacancies in the corresponding tunneled crystal packings, which leads to perfect FCC, HCP, and Barlow packings at the maximal density. The simple-cubic (SC) and body-centered-cubic (BCC) packings (not jammed) are also shown.

increase in ϕ is associated with partial crystallization of the packings, as indicated by the sharp peaks of the pair correlation function g_2 of the packings (see Fig. 4). The fraction of "rattlers" (i.e., locally unjammed individual particles that can move freely within cages of jammed neighbors) decreases (from $\sim$2.8% with $\phi\approx0.64$ to 0% with $\phi=0.740\,48\ldots$) and the average contact number per particle Z increases (from 6 to 12) as the density increases due to the (partial) crystallization of the packings, and we have $Z\approx6.00$, 6.45, and 7.96, respectively, for densities $\phi\approx0.64$, 0.68, and 0.72 (rattlers are excluded when computing Z).

Several representative packings obtained via the SLP algorithm are also mapped onto the ϕ-$\mathfrak{T}$ plane (red squares in Fig. 3). It can be seen that jamming at $\phi<0.64$ is necessarily associated with an increase in the degree of order, which is also indicated by the increase in average contact number per particle (i.e., $Z\approx6.37$ for $\phi\approx0.6$) and the decrease in the fraction of rattlers (i.e., $\sim$1.1% for $\phi\approx0.6$).[24] The pair correlation function g_2 of a representative packing configuration (with $N=2235$) is shown in Fig. 4.

The "tunneled crystal" packings are mapped onto the ϕ-$\mathfrak{T}$ plane (green up-triangles). The FCC tunneled crystal

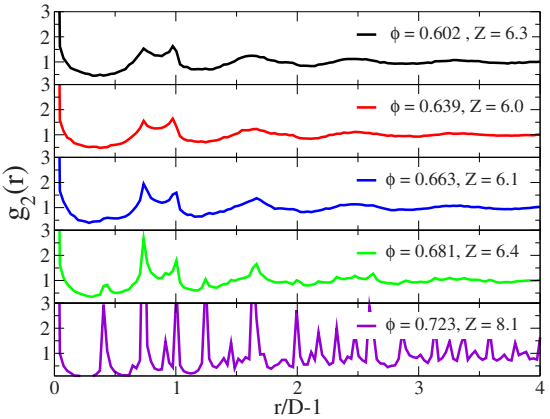

FIG. 4. (Color online) Pair correlation function g_2 and average contact number Z of representative jammed sphere packings at different densities, where D is the sphere diameter.

possesses the highest translational order metric $\mathfrak{T}$, the "disordered" Barlow tunneled crystal (a random stacking of layers of honeycomb-lattice sphere packings) possesses the lowest $\mathfrak{T}$, and the "zig-zag" tunneled crystal (analog of the hexagonal-close sphere packing)[15] is in between. The dashed blue lines show the spectrum of packings generated by randomly filling the vacancies in the corresponding "tunneled crystal" packings, leading to perfect FCC, HCP, and "disordered" Barlow packings at the maximal density.[12] Several representative layered packings are also mapped onto the ϕ-$\mathfrak{T}$ plane (purple diamonds) in Fig. 3. The simple-cubic (ϕ=0.5235…, blue left-triangle) and body-centered-cubic (ϕ=0.6801…, orange right-triangle) packings are also shown in Fig. 3. Note the SC and BCC packings are not collectively jammed.

IV. CONCLUSIONS AND DISCUSSION

In summary, we have shown that jammed sphere packings can be produced with a narrow distribution centered at any ϕ over a wide range $\phi \in [0.6, 0.740\,48\ldots]$ with a diversity of disorder, by exploring the algorithmic variability of packing protocols. This suggests that any temptation to select a specific ϕ value as uniquely significant or universal (e.g., $\phi \approx 0.64$) based on its frequency of occurrence is primarily due to an inadequate sampling of the full range of algorithmic richness and variability of packing protocols. Our packings are characterized by a "geometric-structure" approach, i.e., they are mapped onto the ϕ-$\mathfrak{T}$ plane; and we have shown that moving away from the MRJ density in both directions (i.e., lower or higher ϕ) leads to a higher degree of order and a larger average contact number.

It is claimed that the protocol (e.g., the athermal relaxation method used in Ref. 9) in which the jammed states of soft spheres are weighted by the volume of their basins of attraction has a clear interpretation in terms of the energy landscape, e.g., an ensemble of jammed states is considered. In fact, the conceptual framework of such a protocol (e.g., the energy landscape picture and the ensemble of inherent structures) was introduced by one of us over 40 years ago.[25–27] Although this approach is well-defined and theoretically possible, it is inevitable that all protocols sample the energy landscape in their own biased fashion. Therefore, there is no compelling mathematical criterion for selecting one protocol over the others. For example, from completely random initial configurations (i.e., Poisson distribution of points), it is found the jamming density sharply peaks at $\phi \approx 0.64$.[9] However, it was shown nearly a decade ago that a much wider density range can be achieved for both monodisperse and polydisperse sphere packings using the LS packing algorithm;[7,8,14,28] and recently similar results for polydisperse spheres have been obtained[18] using the same athermal protocol as in Ref. 9. More recently, we have devised a novel (athermal) linear-programming packing protocol that enables one to obtain a spectrum of inherent structure ranging from disordered jammed packings up to the maximal density packings starting from random initial configurations.[20] All of these results suggest that there is no "universal" protocol that can generate all possible jammed states in an unbiased fashion.

Moreover, it is not clear how the jammed states should be weighted in an ensemble. It has been suggested that the volume of the basins of attraction for systems starting from Poisson distributions of interacting points[9] should be used for the weighting. However, in reality virtually no jammed systems are experimentally produced using a Poisson distribution as an initial configuration. In addition, the jammed packings produced either numerically or experimentally always contain a small but different fraction of rattlers. Strictly speaking, these jammed states are not single points on the energy landscape but bounded regions with dimensions equal to the number of degrees of freedom of the rattlers. The dimension of the jamming basins are different, and therefore, it is ambiguous to consider their volumes for weighting and to characterize them on the same footing. Ideally, each jamming basin should be characterized individually as emphasized in the "geometric-structure" approach. Removing the rattlers will not affect the jamming nature of the packings, but leads to the lower jamming density. This further adds ambiguity to a density-alone characterization of jammed packings, which is employed in the "ensemble" approach.

We also would like to note that as exercised the "ensemble" approach has focused on protocols that disproportionately generate disordered packings, presumably because such packings have a high frequency of occurrence in typical experiments or simulations. The specific protocols employed discriminate against the ordered structures such as the FCC packing (and its stacking variants) and the tunneled crystals[15] and result in an inappropriate narrow vision of the set of all possible collectively jammed packings. Thus, the physical relevance of jammed packings should not be determined based on their frequency of occurrence in experiments or simulations, which again are protocol dependent.

From our point of view, the "ensemble" and "geometric-structure" approaches do not conflict with each other but rather are complementary. For example, the "geometric-structure" approach characterizes individual packing configurations drawn from well-defined ensembles. However, in our experience, the "geometric-structure" approach can always provide nontrivial solutions when the "ensemble" approach breaks down, such as the possible identification of disordered jammed two-dimensional packings discussed in the Introduction. Therefore, we believe that the "geometric-structure" approach, when utilized in the broad context of a full set of available protocols, has a capability to discover, integrate and characterize packing structures that go well beyond the disordered set. We emphasize that all of the aforementioned issues have been addressed in the literature.[7,8,12,14,29] In this paper, we have examined the applications of the "geometric-structure" approach in a broader context by amplifying and quantifying some of these issues.

An interesting open question that naturally arises is whether $\phi \approx 0.60$ is the lower limit on the density of jammed amorphous sphere packings? We note that the distribution of density for $N \sim 2500$ shown in Fig. 1(a) can be fitted with a Gaussian with mean ϕ=0.602 and standard deviation σ

$=0.002$. This strongly indicates that there is a small but finite probability of finding jammed packings with even lower densities. Given enough number of trials, such packings could be obtained in principle. It is noteworthy that the density $\phi = \sqrt{2}\pi/9 = 0.493\,65\ldots$ associated with the "tunneled crystals" is likely to be the threshold (i.e., lowest possible) density for strictly jammed sphere packings in three dimensions.[15] However, neither the LS nor the SLP algorithms as normally implemented is able to produce such packings, presumably because both algorithms tend to densify the packing and jamming is a consequence of their "compression" nature. Lower ϕ is attainable via the SLP algorithm because jamming is achieved through local reconfigurations. However, it is currently not designed to find highly unsaturated jammed packings, such as the tunneled crystals. Therefore, we see no reason that jammed amorphous sphere packings with even lower density cannot be produced via carefully designed protocols.

A limitation of current protocols designed to produce jammed packings is that they invariably lead to packings with high densities. It is highly desirable to devise protocols that explicitly take into account the requirement of jamming as well as other packing characteristics (e.g., ϕ and Z). One possible approach is to delineate the conditions for jamming (and other characteristics) in a quantitative way and to include them as constraints of an optimization problem. We have suggested possible solutions to the problem of producing low-density jammed amorphous packings in Ref. 20. In future research, we will focus on the development of such packing protocols, which is a highly nontrivial challenge.

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[1]J. D. Bernal and J. Mason, Nature (London) **188**, 910 (1960).
[2]J. D. Bernal, in *Liquids: Structure, Properties, Solid Interactions*, edited by T. J. Hughel (Elsevier, New York, 1965), pp. 25–50.
[3]B. D. Lubachevsky and F. H. Stillinger, J. Stat. Phys. **60**, 561 (1990).
[4]R. J. Speedy, J. Chem. Phys. **100**, 6684 (1994).
[5]S. F. Edwards, in *Granular Matter*, edited by A. Mehta (Springer-Verlag, New York, 1994).
[6]A. J. Liu and S. R. Nagel, Nature (London) **396**, 21 (1998).
[7]S. Torquato, T. M. Truskett, and P. G. Debenedetti, Phys. Rev. Lett. **84**, 2064 (2000).
[8]A. R. Kansal, S. Torquato, and F. H. Stillinger, Phys. Rev. E **66**, 041109 (2002).
[9]C. S. O'Hern, L. E. Silbert, A. J. Liu, and S. R. Nagel, Phys. Rev. E **68**, 011306 (2003).
[10]A. Donev, F. H. Stillinger, and S. Torquato, Phys. Rev. Lett. **95**, 090604 (2005).
[11]G. F. Parisi and F. Zamponi, Rev. Mod. Phys. **82**, 789 (2010).
[12]S. Torquato and F. H. Stillinger, Rev. Mod. Phys. **82**, 2633 (2010).
[13]S. Torquato and F. H. Stillinger, J. Phys. Chem. B **105**, 11849 (2001).
[14]T. M. Truskett, S. Torquato, and P. G. Debenedetti, Phys. Rev. E **62**, 993 (2000).
[15]S. Torquato and F. H. Stillinger, J. Appl. Phys. **102**, 093511 (2007).
[16]S. Torquato and Y. Jiao, Nature (London) **460**, 876 (2009).
[17]S. Torquato and Y. Jiao, Phys. Rev. E **80**, 041104 (2009).
[18]P. Chaudhuri, L. Berthier, and S. Sastry, Phys. Rev. Lett. **104**, 165701 (2010). We note that these authors have explicitly shown that disordered 3D binary soft-sphere packings can jam within a density range [0.646, 0.662], by tuning the density of initial configurations used to produce the jammed packings. This is a relatively narrow density range but is consistent with our results and those reported in Ref. 8.
[19]M. Hermes and M. Dijkstra, EPL **89**, 38005 (2010).
[20]S. Torquato and Y. Jiao, Phys. Rev. E **82**, 061302 (2010).
[21]A. Donev, S. Torquato, F. H. Stillinger, and R. Connelly, J. Appl. Phys. **95**, 989 (2004).
[22]A. Donev, S. Torquato, and F. H. Stillinger, J. Comput. Phys. **202**, 737 (2005).
[23]G. Y. Onoda and E. G. Liniger, Phys. Rev. Lett. **64**, 2727 (1990).
[24]Note that we have also computed the "crystal-independent" translational order metric T^* defined in Ref. 14 (i.e., one that does not assume a reference crystal state as the most ordered) for typical packings for $0.6 \leq \phi \leq 0.64$. As expected, these results for T^* are positively correlated with those for the aforementioned "crystal-dependent" translational order metric $\mathfrak{T}$ and hence these two translational order metrics are consistent with one another.
[25]F. H. Stillinger, E. A. DiMarzio, and R. L. Kornegay, J. Chem. Phys. **40**, 1564 (1964).
[26]F. H. Stillinger and T. A. Weber, Phys. Rev. A **25**, 978 (1982); F. H. Stillinger and T. A. Weber, Science **225**, 983 (1984) (In these papers, the concept of inherent structures was formalized for many-particle system interacting via continuous soft potentials. The set of configurational points that map to the same mechanically stable local energy minimum via a steepest-descent trajectory define uniquely a basin associated with the local minimum called an inherent structure).
[27]F. H. Stillinger and T. A. Weber, J. Chem. Phys. **83**, 4767 (1985) (In this paper, the concept of inherent structures was rigorously generalized to hard-sphere systems by considering the infinite-n limit of purely repulsive power-law pair potentials $1/r^n$).
[28]A. R. Kansal, S. Torquato, and F. H. Stillinger, J. Chem. Phys. **117**, 8212 (2002).
[29]A. Donev, S. Torquato, F. H. Stillinger, and R. Connelly, Phys. Rev. E **70**, 043301 (2004).