Diversity of order and densities in jammed hard-particle packings

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Recently the conventional notion of random close packing has been supplanted by the more appropriate concept of the maximally random jammed (MRJ) state. This inevitably leads to the necessity of distinguishing the MRJ state among the entire collection of jammed packings. While the ideal method of addressing this question would be to enumerate and classify all possible jammed hard-sphere configurations, practical limitations prevent such a method from being employed. Instead, we generate numerically a large number of representative jammed hard-sphere configurations (primarily relying on a slight modification of the Lubachevsky-Stillinger algorithm to do so) and evaluate several commonly employed order metrics for each of these packings. Our investigation shows that, even in the large-system limit, jammed systems of hard spheres can be generated with a wide range of packing fractions from $\phi \approx 0.52$ to the fcc limit ($\phi \approx 0.74$). Moreover, at a fixed packing fraction, the variation in the order can be substantial, indicating that the density alone does not uniquely characterize a packing. Interestingly, each order metric evaluated yielded a relatively consistent estimate for the packing fraction of the maximally random jammed state ($\phi_{\mathrm{MRJ}} \approx 0.63$). This estimate, however, is compromised by the weaknesses in the order metrics available, and we propose several guiding principles for future efforts to define more broadly applicable metrics.

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I. INTRODUCTION

Hard-sphere systems have served as useful starting points to study the structure of diverse systems such as liquids, living cells, granular media, glasses, and powders [1–5]. Despite its apparent simplicity, however, the hard-sphere system offers many conundrums. One of the most notable examples of such a conundrum is the recent demonstration that the venerable notion of the *random close packed* (RCP) ''state'' of hard-sphere systems is ill defined and must be abandoned [6]. This explains why, despite many attempts, there has never been a rigorous prediction of the RCP density. Consistent with the ill-defined nature of the RCP state is the fact that the putative RCP density is clearly dependent on the protocol used to generate the packing, even when this density is reproducible within a specific protocol. To replace the RCP notion, Torquato, Truskett, and Debenedetti [6] introduced a new concept called the *maximally random jammed* (MRJ) state, defined to be the configuration that maximizes disorder among all jammed hard-sphere arrangements. This definition lays the groundwork for studying randomness in packings of particles (and in condensed-phase systems in general) and initiates the challenging search for the MRJ state in a quantitative fashion.

One method for identifying the MRJ state would be to generate the ensemble of all possible jammed configurations (or a statistical sample of this ensemble) and then calculate an ''entropy'' as a function of packing fraction based on these configurations. An example of a possible entropy definition is given by Edwards [7]. However, there are several practical hurdles to the use of an entropy measure for finding the MRJ state. Most challenging among these is the necessity of generating all possible jammed states in an unbiased fashion using a ''universal'' protocol in the large-system limit. Even if such a protocol could be developed, however, the issue of weighting the resulting configurations remains. Some regions of configuration space are likely to be much more densely populated with jammed states than other regions. This suggests that configurations in those regions should be accorded modified weight, but to what degree is unclear. Consequently, at least in the near term, a different approach is necessary.

We seek to identify the MRJ state by generating a large database of representative jammed configurations and relying on commonly employed *order metrics* (such as those discussed in Ref. [8]) to identify the most random among them. In principle, the disorder in a packing is completely characterized by the many-body configurational probability density function. In practice, however, such complete information is never available and one must settle for reduced information. For example, from functionals of lower-order correlation functions, one can extract a set of scalar order metrics. Although there is no uniquely qualified order metric to apply to the identification of the MRJ state, one anticipates that it is possible to develop a set of reasonable and distinct

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order metrics. Ideally, each order metric in this set will identify the same MRJ state, so the most sensitive among them can be chosen to identify the MRJ density precisely.

To create a representative database of packings, we primarily rely on the well-known Lubachevsky-Stillinger (LS) algorithm [9], but we also include a small number of configurations based on the method of Zinchenko [10] or using the protocol of Speedy [11]. The details of these methods are contained in Sec. II. We then evaluate the order of each configuration generated based on several order metrics (defined in Sec. III). We attempt to use this information to estimate the density of the MRJ state in Sec. IV. In doing so, we identify several areas in which the order metrics we have employed fail and, relying on this experience, we conclude in Sec. V with a discussion of guidelines for designing and evaluating new order metrics.

II. GENERATION OF SPHERE PACKINGS

In the results described here, we have employed systems of 500 identical hard spheres. We have chosen 500 spheres to minimize the computational costs of generating each configuration. Several larger systems of up to 10 000 spheres were also generated. As the system size became larger, the packing fractions for a given set of parameters become more narrowly distributed. Importantly, however, by varying the parameters employed in the LS algorithm, a wide range of packing fractions can be generated, i.e., approximately from $\phi=0.62$ to the fcc limit ($\phi\approx 0.74$). Indeed, the narrowing of the distribution of packing fractions means that for sufficiently large systems the packing fraction can be specified quite precisely *a priori* anywhere in this wide range of possible densities. Note that other protocols, as discussed below, can achieve an even broader range of densities (with packing fractions as low as $\phi=0.52$).

The simulation box was a cube with periodic boundary conditions. We employed the LS algorithm [9] for the generation of most of our hard-sphere packings. This algorithm is essentially a molecular dynamics simulation in which the spheres grow over time. Once the initial conditions (sphere positions and velocities) are fixed, the system evolves deterministically. Intuitively, the most random configurations should result from initializing the sphere centers as a Poisson process with diameters $\sigma\rightarrow 0$ (i.e., as an ideal gas). Using more dense initial configurations like the random sequential addition (RSA) algorithm [12] or a low-density equilibrium liquid, however, produces very similar final packings but saves significant computational time. We use RSA configurations to set the initial positions of the sphere centers. In the RSA method, sphere centers are placed uniformly in space one at a time, rejecting any placement that would result in an overlap. This allows us to start our simulations at packing fractions of $\phi=0.30$ rather than the very low-density ideal gas state.

The LS algorithm has a single parameter Γ, which represents the sphere growth rate relative to the mean sphere speed. As the spheres grow larger, the collision frequency increases and a maximum packing fraction is asymptotically approached. In monodisperse systems, Torquato, Truskett, and Debenedetti [6] have shown that this maximum packing fraction is dependent on the growth rate of the spheres. Roughly speaking, by choosing a high growth rate, the structure of the initial configuration is preserved to some extent, leading to a more random final configuration. A slower growth rate allows the spheres more time to equilibrate and so yields more dense, but somewhat more ordered, final systems.

While the final configurations that can be generated from the LS algorithm have been shown to span a substantial range of volume fractions essentially independent of system size [6], it is reasonable to expect that jammed configurations may exist outside the range that has been generated previously. To extend this range, however, it is necessary to modify the protocol employed very slightly. Specifically, rather than choosing a single growth rate Γ, the growth rate of the spheres is decreased during the generation of a packing. This permits the structure of the original packing to be retained to the greatest degree possible. We have chosen to employ discrete drops in the growth rate, in which the growth rate is decreased by a constant factor at each step. Other schedules for the growth rate are certainly feasible.

As recently noted by Torquato and Stillinger [13], there are several possible definitions of ''jammed.'' Running the LS algorithm until the collision rate diverges ensures that the spheres are *locally jammed* (except for the possible presence of caged but movable particles, or ''rattlers''). Local jamming requires that no single sphere in the system can be translated while holding fixed the positions of all other spheres in the system. Recent work has shown that the LS protocol generates packings that are strictly jammed in almost all cases [14]. Strict jamming requires as a prerequisite that there can be no collective motion of any subset of the spheres, holding the positions of the remaining spheres fixed. This definition of jamming also requires that attempted deformations of the simulation box (e.g., shearing the box) not generate any sphere motions. We restrict our interest here to strictly jammed systems. Typically a small percentage (2–3 %) of rattler particles are present in our packings. The remainder of the particles, however, satisfy the strictly jammed condition. We retain the rattlers to remain in accord with experimental packings. In restricting ourselves to strictly jammed packings, we eliminate some common lattices from our consideration, most notably the simple cubic lattice. We have tested a representative sample of the random configurations generated for strict jamming and found all configurations tested to meet this criterion.

In addition to the packings generated using the LS algorithm, we also include a small number of packings generated using the Zinchenko protocol [10] or based on the Speedy protocol [11]. The original packings that result from the Speedy protocol, however, are not even collectively jammed and so cannot be directly compared to strictly jammed structures. We can use the LS algorithm to compress the Speedy configurations further, producing strictly jammed packings. This is possible because the LS algorithm can be initialized with any sphere configuration that does not include overlaps. Thus, we use the Speedy configurations as initial configurations for the LS algorithm, yielding strictly jammed packings.

III. DEFINITION OF ORDER METRICS

The bond-orientational order metric Q_6 defined by Steinhardt, Nelson, and Ronchetti [1] provides a *global* measure of crystallinity in the system. For each sphere, a set of bonds is defined connecting its center to the centers of its nearest neighbor spheres. For this purpose it is necessary to choose a method for designating two spheres as nearest neighbors. A number of methods for doing this is possible and will be discussed below in greater detail, but for the present definition it is sufficient to assume that nearest neighbor spheres can be identified. We can then calculate Q_6 based on the spherical harmonics $Y_{lm}(\theta,\varphi)$ as

$$Q_6 \equiv \left(\frac{4\pi}{13} \sum_{m=-6}^{6} \left| \frac{1}{N_b} \sum_{i=1}^{N_b} Y_{6m}(\theta_i,\varphi_i) \right|^2 \right)^{1/2}, \qquad (1)$$

where N_b is to the number of nearest neighbor bonds in the system and θ_i and φ_i are the polar and azimuthal angles of bond i. Note that because we are using Q_6 only the Y_{6m} are used, though in the original Steinhardt, Nelson, and Ronchetti definition any spherical harmonic could be used (i.e., Q_8 could be calculated using Y_{8m}). Q_6 is chosen here because it reaches it maximum value for the perfect FCC crystal (though the exact value varies slightly depending on the definition of nearest neighbors).

A more local measure of orientational order, $Q_{6,\mathrm{local}}$, can be obtained by evaluating the bond-orientational order at each sphere individually, and then averaging over all spheres. More precisely, $Q_{6,\mathrm{local}}$ may be calculated as

$$Q_{6,\mathrm{local}} \equiv \sum_{j=1}^{N} \left(\frac{4\pi}{13} \sum_{m=-6}^{6} \left| \frac{1}{n_b} \sum_{i=1}^{n_b^j} Y_{6m}(\theta_i,\varphi_i) \right|^2 \right)^{1/2}, \qquad (2)$$

where n_b^j is the number of nearest neighbors of sphere j. $Q_{6,\mathrm{local}}$ is analogous to the two-dimensional definition of local bond-orientational order in Kansal, Truskett, and Torquato [15]. As noted in that work, a local measure of order is more sensitive to small crystalline regions within a packing than is a global measure. In addition, using a local measure avoids the possibility of destructive interference between different crystalline regions.

In addition to bond-orientational order, sphere systems may also be characterized by the presence of translational order. Using the fcc lattice as a reference, Truskett, Torquato, and Debenedetti [8] have defined a translational order metric T. In this metric, the mean occupation of thin shells concentric with each individual sphere in a system is compared to the mean occupation of the same shells in the open fcc lattice and the ideal gas at the same number density. The translational order T is calculated as

$$T \equiv \left| \frac{\sum_{i=1}^{N_{\mathrm{shells}}} (n_i - n_i^{\mathrm{ideal}})}{\sum_{i=1}^{N_{\mathrm{shells}}} (n_i^{\mathrm{fcc}} - n_i^{\mathrm{ideal}})} \right|, \qquad (3)$$

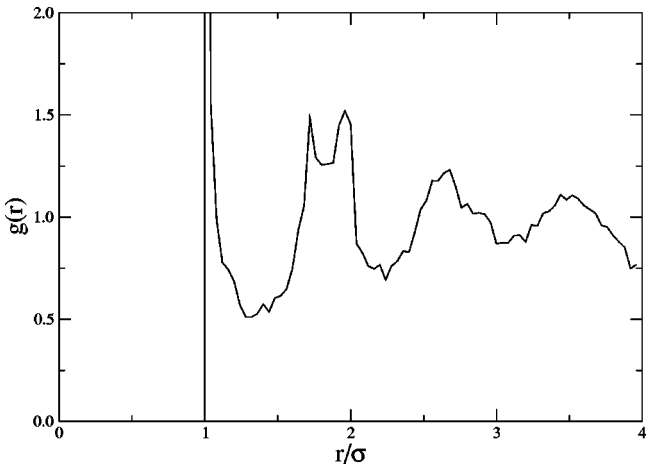

FIG. 1. Radial distribution function of a single 500 sphere packing ($\phi = 0.637$). Note that there are sizable fluctuations in the area of the first minimum, making a precise determination of the position of the minimum impossible.

in which n_i is the average occupancy of the ith shell and the superscripts ''fcc'' and ''ideal'' refer to the reference open fcc lattice and ideal gas systems, respectively. There are several parameters that must be specified to make use of this order metric. In particular, N_{shells} is the number of shells over which the summation is carried out. In addition, it is necessary to specify the position of the shells and their width. In order to ensure that the fcc lattice maximizes the value of T it is necessary to fix the shell positions such that they correspond to the successive neighbor shells of the open fcc lattice. In addition, it is useful to define the shell width such that the maximum amount of space is covered, but that no two shells overlap. In other words, the shell width can be defined as the minimum separation between coordination shells in the open fcc lattice among the first N_{shells} shells.

IV. CALCULATION OF ORDER METRICS

A total of 2600 sphere packings were generated using the LS algorithm with different growth rates and initial configurations. In addition, 60 packings based on the Speedy protocol were analyzed, as were four packings using the Zinchenko protocol. For each of these configurations, the translational order T and the local bond-orientational order $Q_{6,\mathrm{local}}$ were calculated.

Before discussing the results of this analysis, it is necessary to specify the method employed for identifying nearest neighbors for use in evaluating bond-orientational order. Truskett, Torquato, and Debenedetti [8] employed a definition of nearest neighbors based on the radial distribution function of a packing. In particular, a cutoff distance r_{max} is defined by the position of the first minimum in the radial distribution function (see Fig. 1). All spheres within a distance r_{max} are then considered to be nearest neighbors of one another. While this definition works well under some conditions, it is difficult to implement with consistency. In particular, except for lattices or for very large packings, the radial distribution function of a finite packing will contain some

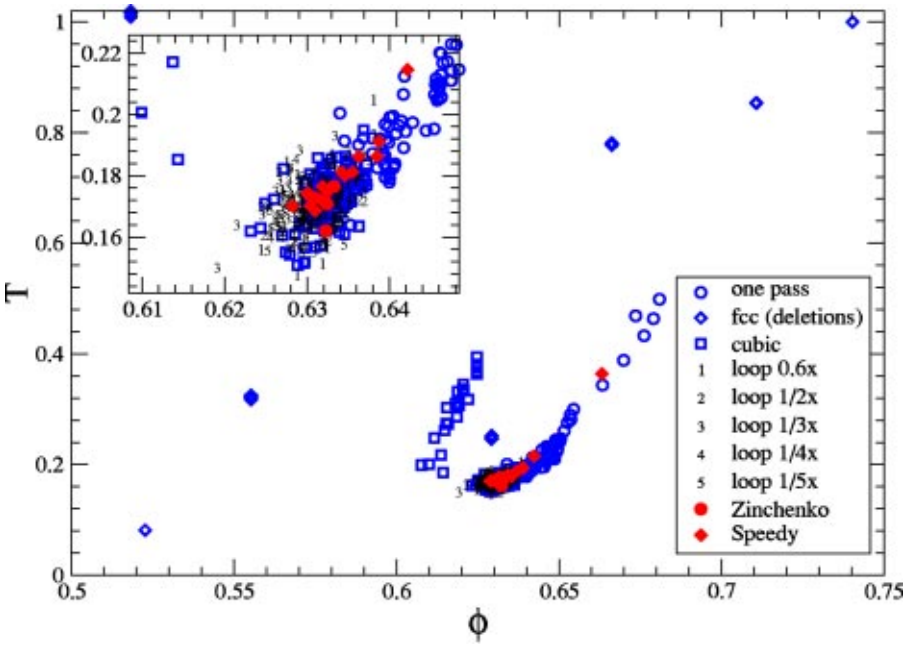

FIG. 2. (Color online) Plot of translational order (T) versus packing fraction (ϕ) for jammed configurations. A description of the parameters used in generating each set of points is contained in the text. Inset is a magnification of the neighborhood near a packing fraction of 0.64. Shown here and in the subsequent figures is a representative subset of all configurations generated.

fluctuations. Consequently, the position of the first minimum will necessarily be subject to some variation. The local bond-orientational order, however, is very sensitive to the value of $r_{\max}$. Reasonable estimates of $r_{\max}$ can lead to differences in $Q_{6,\mathrm{local}}$ of over 5%. These variations are unacceptably large for identifying the MRJ state, so a less subjective definition of nearest neighbors is required. We use the Delaunay triangulation, which is suitable for identifying nearest neighbors uniquely and unambiguously for almost any point set [16]. The exception comes for cases in which five spheres are all equidistant from any single point in space. While such degeneracies are extremely unlikely in random packings, they are present in the simple cubic lattice, the bcc lattice, and the fcc lattice. In these cases, we have perturbed each sphere very slightly from its original location by moving it a distance of $10^{-5}\times\sigma$ in a random direction (where σ is the sphere diameter). This will remove any degeneracy and allow nearest neighbors to be defined.

Figures 2 and 3 show ordering phase diagrams of T and $Q_{6,\mathrm{local}}$ versus packing fraction, ϕ, respectively. The different symbols in the plots correspond to variations in the sphere growth rate and initial condition used in the LS algorithm. The configurations labeled ''one pass'' were generated using a constant growth rate for each configuration. The dimensionless growth rates used were in the range $0.001\leqslant\Gamma\leqslant0.04$ for these configurations. The configurations labeled

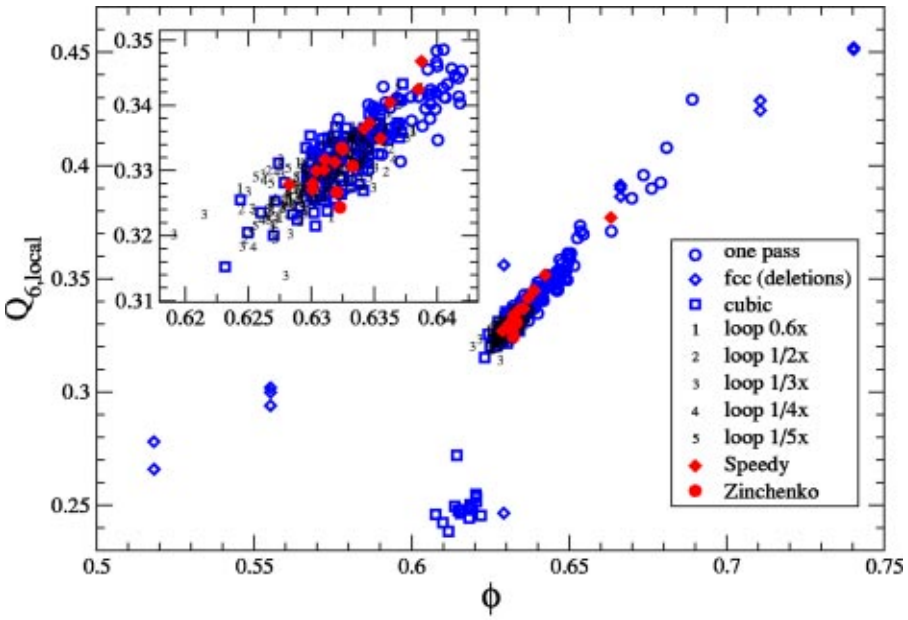

FIG. 3. (Color online) Plot of local bond-orientational order ($Q_{6,\mathrm{local}}$) versus packing fraction (ϕ) for jammed configurations. The same configurations used in Figure 2 are shown here. Inset is a magnification of the neighborhood near a packing fraction of 0.64.

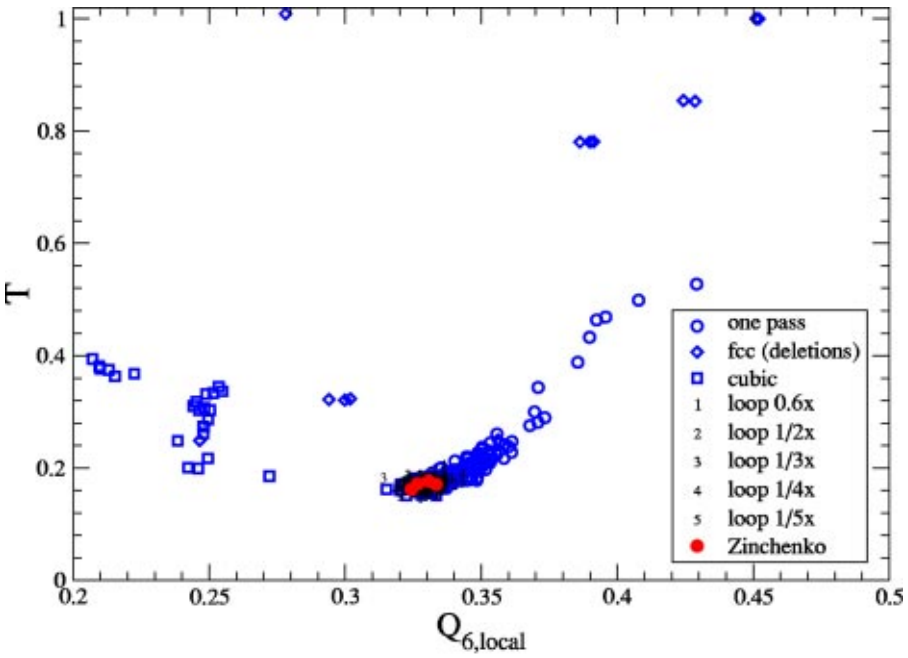

FIG. 4. (Color online) Plot of translational order (T) versus local bond-orientational order ($Q_{6,\mathrm{local}}$) for a representative subset of all configurations generated. Note that it is possible to increase translational order while decreasing orientational order and vice versa.

''loop'' were generated by decreasing the growth rate in a stepwise fashion over the course of generating a single packing. The factor by which the growth rate was decreased at each step is indicated in the legend. For each of these configurations, the initial growth rate was $\Gamma=1.0$, which drops to $\Gamma=0.004$ over the course of the simulation. Points labeled ''cubic'' were generated using the same stepwise decreasing growth rate protocol, but were initialized with the spheres arrayed as a simple cubic lattice. Configurations generated based on the Speedy or Zinchenko protocols are labeled ''Speedy'' and ''Zinchenko,'' respectively, for a representative subset of all (approximately 2700) configurations generated. Note that the Speedy configurations were compressed using the LS algorithm until the collision rate diverged. To retain as much of the original character of the packings as possible, we used a growth rate that started at a value of $\Gamma=1.0$ and dropped by a factor of 3 at each step. In addition, the Speedy configurations each contained 4000 spheres versus the 500 sphere present in all other configurations. For local measures of order, however, the larger system size should not play a significant role. Finally, a set of configurations based on the fcc lattice was also generated and is labeled ''fcc.'' These configurations were generated by randomly deleting spheres from the full fcc lattice such that the final configuration was still strictly jammed, yielding lower-density structures. Using this protocol it proves feasible to reduce the packing fraction to the neighborhood of $\phi=0.52$.

Comparing Fig. 2 and Fig. 3 yields a number of interesting observations. Perhaps the first point worth mentioning regards the neighborhood in which $\phi\approx0.64$ (i.e., near the traditional packing fraction of random close packing). Independent of the choice of order metric, this neighborhood does not indicate any unique point in the ordering ''phase'' diagram. Increases in packing fraction can be obtained at the cost of increasing order and decreases in order can be obtained at slightly lower packing fractions. This shows that the arguments of Torquato, Truskett, and Debenedetti [6] can be applied to packing fractions below $\phi=0.64$ along with those above this value as had been shown previously. Importantly, this demonstrates that there is a relatively broad range of packing fractions over which amorphous hard-sphere systems can be generated. A closely related observation is that for the set of configurations at any given packing fraction there is a range of values for each order metric. Even among amorphous systems at the same packing fraction, some configurations are less ordered than others. This clearly shows that the packing fraction alone does not uniquely characterize an ''amorphous'' packing. In contrast to the RCP state, the MRJ state is defined independently of density and its hallmarks are specified by a collection of relevant structural properties that serve to distinguish it from among the entire diverse collection of jammed packings.

Focusing on the region in which $\phi\approx0.63$, an apparent minimum in both order metrics can be seen. The identification of this region as the MRJ state, however, is problematic for a number of reasons. Foremost, it is clear that for either metric it is possible to generate configurations with a lower value of the metric than even the lowest-order configurations with $\phi\approx0.63$. The types of configurations that result in the lowest values of each order metric are instructive. $Q_{6,\mathrm{local}}$ is minimized for the configurations that resulted from initializing the sphere positions according to the simple cubic lattice. This indication of disorder, however, is spurious, as can be seen from the high value of T for the same set of configurations. Similarly, configurations that result from simply deleting spheres from the fcc lattice can produce extremely low order as measured by T. Again, however, this small amount of order indicated is contradicted by the significant amount of orientational order revealed by the $Q_{6,\mathrm{local}}$ values for these configurations. In fact, these extreme cases are not the only ones in which translational and bond-orientational order are

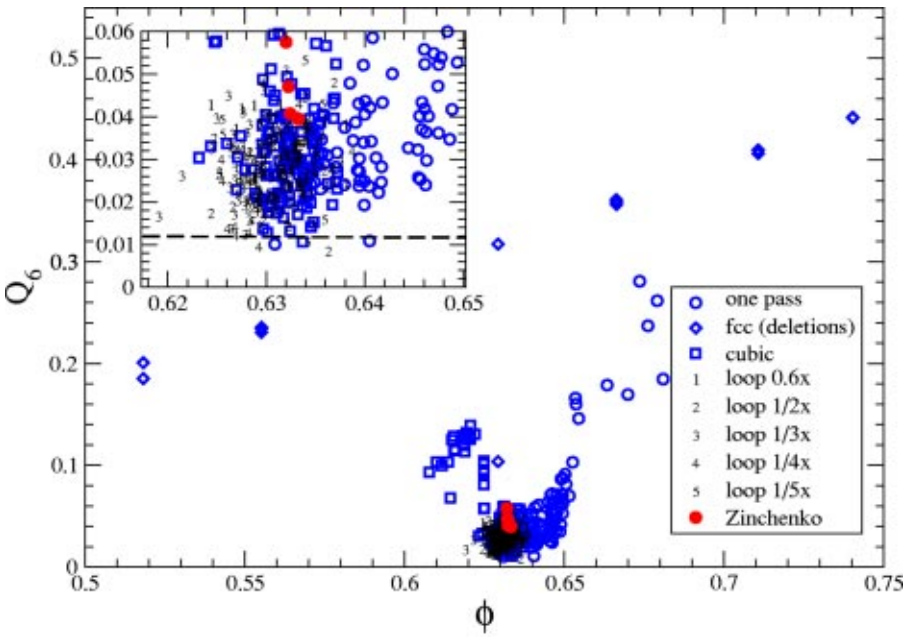

FIG. 5. (Color online) Plot of global bond-orientational order (Q_6) versus packing fraction (ϕ) for a representative subset of all configurations generated. Inset is a magnification of the region near a packing fraction of 0.64. The dashed line indicates the Q_6 value of an ideal gas of 500 particles. Because Q_6 decreases with increasing system size, the packings based on the Speedy configurations are not included in this figure.

not directly related. Shown in Fig. 4 is a plot of T versus $Q_{6,\mathrm{local}}$ for the packings. Note that, while there is a general trend in which both of these order metrics are directly related, there are several exceptions. Most readily visible is the inverse relation between the order metrics for packings that were based on the simple cubic lattice. In addition, it can be observed that there is a nonunique relationship between T and $Q_{6,\mathrm{local}}$ for the fcc lattice with different numbers of deletions. The lack of a monotonic relationship between the different order metrics also makes identifying the MRJ state problematic. In particular, we have not identified any configuration that minimizes both T *and* $Q_{6,\mathrm{local}}$. In other words, configurations with the least order as measured by $Q_{6,\mathrm{local}}$ are not the same as the configurations with the least order measured by T. Thus, among the configurations shown here, it is not possible to uniquely identify a single state as having being the most random based on these order metrics.

In addition to these local measures of order, we have also calculated the global bond-orientational order Q_6 for each configuration. Figure 5 shows a plot of Q_6 versus packing fraction. Because Q_6 tends to decrease with increasing system size (going roughly as $1/N^2$), the 4000 sphere packings based on the Speedy configurations are not included. A few features of this plot are particularly notable. The first is that the structures based on both the simple cubic lattice (compressed to meet the strict jamming criteria) and the fcc lattice with deletions are properly recognized as having high degrees of order. While this suggests that Q_6 may be the most broadly applicable order metric for the configurations considered here, its inability to properly identify order in polycrystalline systems excludes it as a universal metric. In addition, the global Q_6 measure is relatively insensitive between different random systems. While the range of packing fractions that produce the lowest values of Q_6 gives evidence for this observation, a comparison with ideal gas systems offers a more concrete illustration. The average Q_6 value for ideal gas systems of 500 particles is shown as a dashed line in Fig. 5. Note that a number of jammed systems have Q_6 values below that of the ideal gas. Naturally, it is not possible for a system with hard-sphere constraints to be as disordered as the ideal gas, so we can conclude that at low values Q_6 is not sensitive to changes in order. However, the consistency of the estimate of the packing fraction of the maximally random jammed state (excluding those packings that are clearly indicated as ordered by any metric) is worth noting and gives hope that this state can be conclusively identified using an improved order metric.

V. DISCUSSION AND CONCLUSIONS

We have generated a large database of strictly jammed configurations and calculated several order metrics for each packing intended to categorize this diverse set. In doing so, we have shown that there is a wide range of densities over which strictly jammed systems can be created, namely, from $\phi \approx 0.52$ to the fcc limit ($\phi \approx 0.74$). Importantly, this range is at most weakly dependent on system size. Thus, jammed hard-sphere packings can be generated at will in this allowable density range, independent of the system size. Furthermore, we have demonstrated that at a fixed packing fraction, the variation in the order can be substantial, indicating that the density alone does not uniquely characterize a packing. This proves once again that the RCP state is not a meaningful concept. On the other hand, this weakness is not shared by the MRJ state, which is defined independently of density and is specified by a collection of relevant structural properties that serve to distinguish it from among the entire diverse collection of jammed packings. It is noteworthy that each order metric evaluated yielded a relatively consistent estimate for the packing fraction of the maximally random jammed state ($\phi_{\mathrm{MRJ}} \approx 0.63$).

The difficulties encountered in applying any of the commonly employed order metrics to our jammed configurations, however, suggest that the problem of creating a metric that can be applied to a broad range of configurations (or even one that is effective for the much smaller set of jammed configurations) is extremely challenging. Based on the failings of the commonly employed order metrics that we have evaluated, however, it is possible to begin to construct some requirements that any order metric should meet and a set of guidelines for developing better metrics:

We begin with some fundamental mathematical properties that define an order metric, ψ.

(1) The order metric ψ is a well-defined scalar function of the coordinates $\boldsymbol{r}_1, \ldots, \boldsymbol{r}_N$ for any N-particle system.

(2) ψ is subject to the normalization $0 \leqslant \psi \leqslant 1$. For any two states A and B, $\psi(A) > \psi(B)$ implies that state A is to be considered as more ordered than state B.

(3) ψ is invariant to spatial reflections and to translation or rotation of the system as a whole.

Naturally there is an enormous family of scalar functions that possess these three properties. While by our definition all such functions could be considered order metrics, they are not necessarily good order metrics. For example, a function that is minimized for the simple cubic lattice should be recognized as a poor description of order. To distinguish between good order metrics and poor ones, some additional properties of good metrics must be specified. Based on our experiences with bond-orientational and translational order metrics, some of these properties are listed below.

(1) A good order metric should be sensitive to any type of ordering in a system and should not be biased toward any reference system.

(2) A good order metric should reflect the hierarchy of ordering between prototypical systems given by common physical intuition.

(3) Order at any length scale should be detected.

(4) Both the variety of local coordination patterns and the spatial distribution of such patterns should affect the amount of order measured in a system.

In considering the first criterion listed above, it is useful to begin by noting that the failures in the translation order parameter and in the local bond-orientational order parameter are related to the use of a reference system in their design. The bond-orientational order metrics are designed to find signatures of icosahedral order in a packing, but are insensitive to the presence of other types of order. This is manifested most dramatically in the failure of $Q_{6,\mathrm{local}}$ to identify structures which resemble the simple cubic lattice as ordered. A more fundamental problem with the order metrics discussed previously in this work is that each is sensitive only to translational or orientational order; none picks up signatures of both types of order. Thus we can conclude that a good order metric should not be biased toward any single reference structure or type of order.

Although using any specific configuration as a reference state is unlikely to result in a universally useful metric, the output of a good metric should still generally conform to our intuitive notions of order. Specifically, it should be able to reproduce an intuitive hierarchy of order for systems such as perfect crystals, crystals with defects, dense amorphous system, ideal gas. Within any of these broad categories there may be many types of systems, each with different degrees of order. Though it is tempting to create a more stringent test by expanding the hierarchy in each category (e.g., demanding that the fcc lattice be identified as more ordered than the simple cubic lattice), it is difficult to do so without biasing the test toward a specific type of order or reference structure. It is important, however, to ensure that all structures that fall within a given category are properly identified by the order metric. For example, a good metric should be able to identify both crystals with point defects and polycrystals as less ordered than any perfect crystal, but more ordered than any amorphous system. The global bond-orientational order metric fails to meet this criterion in (at least) two ways. First, it cannot distinguish many jammed configurations from the ideal gas. Second, it incorrectly identifies polycrystalline systems as highly disordered.

The example of polycrystalline systems is useful to discuss the third criterion for good order metrics. Specifically, Q_6 fails to identify this system as ordered because it is only sensitive to order at the length scale of the system. A good order metric should be sensitive to order at any length scale. Meeting this condition is likely to require that an order metric use correlation information from all length scales. This is in contrast to the bond-orientational order metrics, which only rely on nearest neighbor information, or the translational order metric, which only uses a fixed number of coordination shells. Of course, to produce a single scalar from such information will require that it be integrated in some fashion. The related fourth criterion is that an order metric should be sensitive to the different local coordinations present in a system (i.e., the number of different arrangements of a sphere and its first coordination shell). In particular, a good order metric should reflect the variety of local coordinations present as well as the manner in which they are distributed throughout the system. Any spatial correlations between distinguishable ''phases'' (i.e., pattern segregation) should increase the order of the system.

Along with these criteria that a good, broadly applicable order metric must meet, there are several features that the set of order metrics useful for identifying the MRJ state should additionally satisfy. Because any search for the MRJ state will be based on finite systems, ψ should be effective for such packings. The identification of the MRJ state requires that for any jammed particle packing, $0 < \psi_{\mathrm{min}} \leqslant \psi$, where ψ_{min} is independent of the size of the configuration. The minimum value ψ_{min} should be realized at only a single statistical state for each class of jamming. The state that realizes this minimum is the MRJ state. Ideally, the same statistical state will realize the minimum value for all good metrics.

Although our efforts indicate that it is necessary that an order metric conform to all of the above conditions to be useful in identifying the MRJ state, they do not speak to the sufficiency of this list. It is an open question whether other criteria on order metrics are important in the search for the MRJ state. In addition, the actual design and application of an order metric that follows these guidelines remains to be demonstrated. Although it may not be possible to define a

metric that meets all of the above conditions in a closed mathematical form, an acceptably close approximation in closed form may be attainable. Finding such a metric, however, is a necessary step in the effort to understand the fundamental character of ''randomness'' in hard-particle systems.

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