Structure and Rigidity in Maximally Random Jammed Packings of Hard Particles

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Abstract

Packings of hard particles have served as a powerful, yet simple model for a wide variety of physical systems. One particularly interesting subset of these packings are so-called maximally random jammed (MRJ) packings, which constitute the most disordered packings that exist subject to the constraint of jamming (mechanical stability). In this dissertation, we first investigate the consequences of recently-discovered sequential linear programming (SLP) techniques to present previously-unknown possibilities for MRJ packings of two- and three-dimensional hard disks and spheres, respectively. We then turn our focus away from the limit of jamming and identify some structural signatures accompanying various compression processes towards jammed states that are indicative of an incipient rigid structure.

In Chapter 2, we utilize the Torquato-Jiao SLP algorithm to construct MRJ packings of equal-sized spheres in three dimensions that possess substantial qualitative differences from previous putative MRJ states. We turn towards two dimensions in Chapter 3 and establish the existence of highly disordered, jammed packings of equal-sized disks that were previously thought not to exist. We discuss the implications that these findings have for our understanding of disorder in packing problems.

In Chapter 4, we utilize a novel SLP algorithm we call the “pop test” to scrutinize the conjectured link between jamming and hyperuniformity. We uncover deficiencies in standard protocols’ abilities to construct truly-jammed states accompanied by a correlated deficiency in exact hyperuniform behavior, suggesting that precise jamming is a particularly subtle matter in probing this connection. In Chapter 5, we consider the direct correlation function as a means of identifying various static signatures of jamming as we compress packings towards both ordered and disordered jammed states with particular attention paid to the growing suppression of long-ranged density fluctuations (“hyperuniformity”). In Chapter 6, we continue this investigation by studying our packings as they approach jamming through the lens of so-called “ge-
ometric” and “force” percolation problems, tuning the relevant parameters to study the configurations in the vicinity of their percolation thresholds and look for signs of an incipient contact network.
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This dissertation carries the number T-3324 in the records of the Department of Mechanical and Aerospace Engineering.
To my mom and dad, Lisa and Kevin Atkinson.

I hope this explains what I’ve been up to.
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6.5 The average coordination $z$ of the particles in the percolating cluster as a function of the hard-core packing fraction $\phi_{HS}$ for the FCC equilibrium crystal and disordered packings compressed using the LS and TJ algorithms at their respective percolation thresholds for geometric percolation. Error bars correspond to a 95% confidence interval. 

6.6 The two-point cluster function $C_2(r)$ for packings produced by the LS algorithm at various hard-core packing fractions $\phi_{HS}$ and their percolation thresholds. 

6.7 The two-point cluster function $C_2(r)$ for packings produced by the TJ algorithm at various hard-core packing fractions $\phi_{HS}$ and their percolation thresholds. 

6.8 Two-point cluster function $C_2(r)$ for ensembles of MRJ-like packings created by the LS algorithm for system sizes $N = 10^3 - 10^4$ for force and geometric percolation. The infinite-system limiting behavior is shown with thick black dashed lines. 

6.9 Log-log plots of the mean cluster size $S$ taken at the percolation threshold for various systems for geometric and force percolation. Error bars correspond to a 95% confidence interval, computed using the Clopper-Pearson method [8]. Lines show least-squares fits to the data. 

6.10 Bond correlation function measured for a variety of systems under both geometric and force percolation.
Chapter 1

Introduction

Our lives are filled with examples of “packings.” In the morning, we spoon out piles of coffee grounds into our coffee maker, possibly adding in a spoonful of sugar, found similarly as a heap of small grains. At the grocer, one sees stands of produce, and displays of fruits are often ordered in an efficient, stable manner. At the beach, we walk on the sand and feel it moving under our feet as we step. The excluded-volume interaction in colloids leads to a rough description using packing principles. Particulate composites such as solid propellants often feature highly complex microstructures which can be understood as a dense particle packing. In low-temperature materials, where repulsions become the most salient feature of interatomic reactions, one may consider related packings of hard particles to discern organizing principles. Very recently with the recent explosion of 3D printing technology, the understanding of this additive manufacturing process as a packing problem of small droplets is apt and has already resulted in significant breakthroughs [9, 10].

What we observe in these examples seems very plain at first: collections of closely-packed particles, arranged in a random, yet familiar fashion. Expressing this familiarity, one of the forefathers of modern packing theory, JD Bernal, once quoted a comment on random packings which Saint Luke attributes to Jesus: “Give and it will
be given unto you; good measure, pressed down, and shaken together, and running over . . . For by your standard of measure it will be measured to you in return.” [11]. However, upon a careful physics-based consideration, we realize that something remarkable is happening. In contrast to the thermal behavior that one might see at the molecular scale which allows, for example, a cup of water to remain in equilibrium and readily crystallize into ice upon cooling, we notice that the dynamics by which packings of macroscopic particles explore their environment is greatly hindered, resulting in the emergence of rich nonequilibrium behavior of considerable diversity. For that reason, it is of great interest to both characterize these systems, and to understand the organizing principles that govern over packings in this “athermal” regime (i.e., where the thermal energy in a grain of sand is wholly insufficient to disturb it).

Despite the apparent simplicity in posing packing problems, it is remarkably difficult to solve these problems. For that reason, it is expedient to begin by considering as simple a system as possible, so long as the desired behaviors are reproduced faithfully. Accordingly, the venerable model of frictionless hard spheres in three-dimensional Euclidean space has served as a fruitful starting point from which considerable progress has been made to explain substances such as liquids, crystalline and glassy solids, low-temperature states of matter, colloids, particulate composites, and biological systems, to name a few [12, 13, 14, 15, 4, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35]. In two dimensions, the ordering principles illuminated by packings of hard disks have not only served to increase our understanding of adsorption, monolayer films, and epithelial cell patterns [36, 37, 38, 39, 40, 41], but they have also provided a means to understanding more fundamental questions concerning the effect of dimensionality as it pertains to organizing principles and critical theories of jamming [42, 43, 29].

The subset of jammed packings possess a special importance because of their direct applicability to understanding questions of structure and mechanical properties
of ordered and disordered solids. Despite the intuitive nature of jamming, a rigorous understanding of many fundamental concepts (including jamming itself!) remained unestablished for decades following the seminal work of Bernal [44]. For example, a formal geometric definition that may be used to inform research in packing problems came only as recently as last decade, where Torquato et al. defined rigorous, hierarchical jamming categories, making our notion of mechanical stability in hard-particle packings concrete [45, 4, 16].

One of the most familiar topics within the realm of packing problems centers around understanding the seemingly-intuitive concept of “random close packing” (RCP), typically thought of as the densest configuration that a disordered packing might be able to achieve [44, 12]. However, upon closer investigation, one finds that the notion of “randomness” is not so black and white [45]. Rather, one might slightly increase the density of a packing by paying the price of a slight decrease in disorder. Importantly, this partial ordering may be very subtle and not involve crystallization (an obvious example of ordering). To illustrate this, Fig. 1.1 shows examples of packings at slightly different densities, with correspondingly-varying degrees of disorder. Thus, the concepts of randomness and close packing are at odds with each other, and one must make some subjective assessment as to what combination of “randomness” and “close packing” is intended by the idea of RCP. As it stands, the concept is ill-defined.

Torquato et al. resolved this issue by introducing the notion of the maximally random jammed (MRJ) state, which is defined as the packing which minimizes some order metric subject to the constraint that it be jammed [45]. By defining order metrics that quantify one’s intuition of disorder, it is possible in theory to enumerate and rank-order jammed packings according to the degree of order present according to these standard order metrics. In fact, the concept of an order metric predates the concept of the MRJ state; for example, Steinhardt et al. used a bond-orientational
Figure 1.1: Examples of packings of equal-sized spheres with packing fractions of $\phi = 0.637$ (a), 0.635 (b), 0.644 (c), and 0.656 (d). Note that the packing shown in (a) is not jammed, and the packings shown in (b) through (d) are positively correlated with regard to their packing fractions and degrees of order.

Hand in hand with this definition is the paradigm of the geometric-structure approach, in which one considers packings on an individual basis, in contrast to the notion of an ensemble which is fundamental to statistical thermodynamics. The advantage to such an approach is that one may put aside the issue of how frequently a packing (or others like it) is likely to be observed. The rationale behind this is that disordered packings are fundamentally nonequilibrium in nature [47]—the thermody-
namically favored configuration of hard spheres at a high density is believed to be the face-centered cubic (FCC) crystal structure [47, 48]. Indeed, at sufficiently high density, no packings other than FCC and its stacking variants (“Barlow packings”) can exist [49].

There is a practical challenge associated with the concept of the MRJ state: it is impossible to enumerate all of the jammed packings that exist even for small numbers of particles [22]. In fact, it is thought that the number of ways to pack $N$ spheres is exponential in $N$, meaning that one faces a list of packings longer than the number of atoms in the universe in systems of far fewer than 100 particles [50]. Thus, one must necessarily rely on suggestions constructed by some sort of (experimental or numerical) packing protocol. The Lubachevsky-Stillinger (LS) molecular dynamics algorithm is one such protocol which, since its discovery, has been regarded as a “gold standard” in hard-particle packing, and has spurred many variations which expand upon it while keeping the same underlying physics [51, 52, 53, 54, 55]. The introduction of the LS algorithm was a boon for the study of packing problems, and has motivated countless investigations.

One such discovery was the observation that compressing hard-sphere packings towards jamming at the MRJ state was accompanied by an anomalous suppression of density fluctuations on large length scales—a property now known as “hyperuniformity” [17, 56]. Such a discovery necessarily calls for the consideration of incredibly large packings; for example, Donev et al. used packings of 1,000,000 spheres in order to investigate length scales on the order of 100 sphere diameters [56], and subsequent investigations necessarily rely on similarly large packings [47]. However, even with modern computing resources, it remains a considerable computational challenge to study such large packings, and the need to supplement such efforts with efficient algorithms is evident. Presently, there is a conjectured link between hyperuniformity and jamming in hard-particle packings which is the subject of intense attention.
With the advent of the Torquato-Jiao (TJ) sphere-packing algorithm, which harnesses the power of sequential linear programming to pose the packing problem for hard-spheres as a rigorous optimization problem, we find ourselves at a particularly exciting time to delve deeply into questions regarding the nature of the MRJ state [61]. By virtue of its design, the TJ algorithm is adept at efficiently generating a diversity of jammed states that are guaranteed by design to be strictly jammed, i.e., stable against compression and shear. Moreover, the use of linear programming to pose packing problems rigorously as optimization problems shows incredible promise, particularly with regards to questions within the context of the geometric-structure approach [62]. It is within this context that this dissertation expands our understanding of the structure and rigidity of disordered hard-particle packings.

This dissertation focuses on a variety of structural signatures that arise in jammed packings as they are compressed towards jammed states. As outlined below, the Chapters that follow each explore different avenues within this paradigm, and there are commonalities in the themes investigated therein. However, each Chapter has been written in a style that allows it to stand on its own. This benefits the reader who may be interested in specific aspects of the work; however, it does result in the repetition of certain preliminaries and definitions.

In Chapter 2, we use the TJ algorithm to generate disordered 3D packings of hard spheres with previously-unattained numerical precision. We find that the packings which TJ creates are closer to the true MRJ state than those produced by other standard protocols, as shown using bond-orientational and translational order metrics. In addition to being more disordered and having a lower density, these putative MRJ packings possess almost half as many “rattler” particles. These findings paint an enticing picture that suggests that the TJ algorithm stands to revise our notion of disorder in the context of jammed particle packings. The work in Chapter 2 has been
published previously as:


They have also been presented in a short talk at the 111th Statistical Mechanics Conference at Rutgers University in May, 2014.

Motivated by the success of the TJ algorithm in redefining the nature of the MRJ state in 3D, we proceed in Chapter 3 by investigating packings of equal-sized disks in 2D. The two-dimensional problem is notoriously difficult, as the congruence of the densest local packing of disks (i.e. a triangle) with the densest global packing (i.e. the triangular lattice) results in the fact that it is very difficult to thwart spontaneous ordering. Thus, observing highly disordered packings similar to what can be found in 3D has proven elusive. Using the TJ algorithm, we demonstrate by construction the existence of relatively large maximally random jammed (MRJ) packings with exactly isostatic jammed backbones and a packing fraction (including rattlers) of $\phi = 0.826$. By contrast, the concept of random close packing (RCP) that identifies the most probable packings as the most disordered misleadingly identifies highly ordered disk packings as RCP in 2D. Fundamental structural descriptors such as the pair correlation function, structure factor, and Voronoi statistics show a strong contrast between the MRJ state and the typical hyperstatic, polycrystalline packings with $\phi \approx 0.88$ that are more commonly obtained using standard packing protocols. Establishing that the MRJ state for monodisperse hard-disks is isostatic and qualitatively distinct from commonly-observed polycrystalline packings contradicts conventional wisdom that such a disordered, isostatic packing does not exist due to a lack of geometrical frustration and sheds light on the nature of disorder. The work in Chapter 3 has been published previously as:

- S. Atkinson, F. H. Stillinger, and S. Torquato, *Existence of Isostatic, Maxi-

They have also been presented in a short talk at the 112th Statistical Mechanics Conference at Rutgers University in December, 2014.

In Chapter 4, we take a close look at the conjectured link between strict jamming (mechanical rigidity) and (effective or exact) hyperuniformity in frictionless hard-particle packings. In doing so, one must necessarily study very large packings in order to access the long-ranged behavior and to ensure that the packings are truly jammed. To this end, we present a rigorous test that uses linear programming techniques in order to test for jamming in putatively collectively and strictly jammed packings of hard-disks in 2D. We show that this rigorous jamming test is superior to standard ways to ascertain jamming, including the so-called “pressure-leak” test. We find that various standard packing protocols struggle to reliably create packings that are jammed for even modest system sizes of $N \approx 10^3$ bidisperse disks in two dimensions; importantly, these packings have a high reduced pressure that persists over extended amounts of time, meaning that they appear to be jammed by conventional tests, though rigorous jamming tests reveal that they are not. We present evidence that suggests that deviations from hyperuniformity in disordered, putatively jammed packings can in part be explained by a shortcoming of the numerical protocols to generate exactly-jammed configurations as a result of a type of “critical slowing down” as the packing’s collective rearrangements in configuration space become locally confined by high-dimensional “bottlenecks” from which escape is a rare event. Additionally, various protocols are able to produce packings exhibiting hyperuniformity to different extents, but this is because certain protocols are better able to approach exactly-jammed configurations. Nonetheless, while one should not expect exact hyperuniformity for disordered packings with rattlers, we find that when jamming is ensured, our packings are very nearly hyperuniform, and deviations from
hyperuniformity correlate with an inability to ensure jamming, suggesting that strict jamming and hyperuniformity are indeed linked. The work in Chapter 4 has been published previously as:


They have also been presented in a short talk at the 115th Statistical Mechanics Conference at Rutgers University in May, 2016.

In Chapter 5, we consider the process of compressing hard-sphere packings towards both ordered and disordered jammed configurations and the role of the direct correlation function \( c(r) \) as a sensitive means of observing structural signatures of the incipient jammed network. While dynamical signatures are known to precede jamming, the task of identifying static structural signatures indicating the onset of jamming have proven more elusive. The observation that compressing hard-particle packings towards jamming manifests an increasing degree of hyperuniformity has paved the way for the analysis of jamming as an “inverted critical point” in which the direct correlation function \( c(r) \), rather than the total correlation function \( h(r) \) diverges. We expand on the notion that \( c(r) \) provides both universal and protocol-specific information as packings approach jamming. We identify a short-ranged scaling \( c(r) \propto -1/r \) that accompanies the formation of the delta function at \( c(D) \), where \( D \) is the diameter of a sphere, that indicates the formation of contacts in all cases, and argue that this is conveying information regarding the long-ranged behavior of \( c(r) \). At densities in the vicinity of the freezing density, we find striking qualitative differences in the structure factor \( S(k) \) as well as \( c(r) \) between TJ- and LS-generated configurations, including the early formation of a delta function at \( c(D) \) in the TJ algorithm’s packings, indicating the early formation of clusters of particles in near-contact. Both algorithms yield structure factors that tend towards zero in the low-wave number limit as jam-
ming is approached. Correspondingly, we observe an emergent power-law decay in $c(r)$ suggesting that it is becoming long-ranged, as predicted by theoretical considerations. These findings advance the notion that static signatures are exhibited by hard-particle packings as they approach jamming and underscores the utility of the direct correlation function as a sensitive means of monitoring for the appearance of an incipient rigid network. The results in Chapter 5 have been accepted for publication and will appear as:


In Chapter 6, we continue our investigation of static signatures that precede jamming by considering the problems of geometric and force percolation as applied to these systems of ordered and disordered sphere packings near jamming. Using geometric percolation, we find a number of indicators that point to the formation of an incipient rigid backbone as the packings are compressed toward ordered and disordered jammed states. We also observe qualitative differences between the percolation behavior of packings generated using the LS and TJ algorithms and connect these observations to underlying differences in how these protocols explore configuration space. We measure the critical exponents associated with the two percolation problems and our various systems and determine that the geometric and force percolation and geometric models are in different universality classes and that they do not depend upon the degree of disorder in the system. We also remark on the singular nature of jamming and its consequences for these percolation problems as they approach this singular point. Our results highlight the utility of percolation techniques to identify static signatures of jamming and infer the presence of an incipient structure in nearly-jammed hard-particle packings.

As it stands, the broad topic of understanding the structure and organizing prin-
cles at play in MRJ-like and other disordered packings remains full of unanswered questions. In Chapter 7, we review the advances that have been discussed in this dissertation, and we also consider a range of interesting questions, the answers to which seem to be reachable in the near future.
Chapter 2

Detailed Characterization of Rattlers in Exactly Isostatic, Strictly Jammed Sphere Packings

2.1 Introduction

A packing in $d$-dimensional Euclidean space $\mathbb{R}^d$ is defined as a collection of particles that do not overlap with one another. The packing density, $\phi$, is the fraction of $\mathbb{R}^d$ covered by the particles. In three dimensions (3D), considerable attention has been given to characterizing packings of monodisperse hard spheres since they serve as simple, yet powerful models of many-particle systems such as liquids, glasses, colloids, particulate composites, and biological systems, to name a few [12, 14, 20, 24, 31]. In particular, considerable effort has been put into studying the subset of hard-sphere packings that are jammed (roughly speaking, packings that are mechanically stable). It is known that jammed packings of hard spheres can take on a wide range of densities as high as $\phi = \pi/\sqrt{18} = 0.74048 \ldots$ for the fcc crystal and as low as $\phi = \pi\sqrt{2}/9 = 0.49365 \ldots$ for the tunneled crystal [2]. In addition to this considerable range in
density, jammed sphere packings may also exhibit a wide continuum of order to disorder from perfect crystals to packings that exhibit no crystalline order whatsoever. Torquato et al. have proposed “order maps”, one of which is shown in Fig. 2.1, in which all sphere packing configurations can be characterized according to their density, some order metric $\psi$ (subject typically to the normalization $0 \leq \psi \leq 1$), and whether or not they are jammed [45, 63, 1, 62]. Importantly, the frequency of occurrence of a particular configuration is irrelevant insofar as the order map is concerned. In other words, the order map emphasizes a geometric-structure approach to packing by characterizing single configurations, regardless of how they were generated (e.g., whether through some physical dynamical process or otherwise) or their occurrence probability [1].

Among all jammed sphere packings, the maximally random jammed (MRJ) state is the one that minimizes $\psi$ among all statistically homogeneous and isotropic jammed packings [45, 4, 1]. The MRJ state is a well-defined minimum in an order map in that for a particular choice of jamming category and order metric it can be identified unambiguously, making mathematically precise the familiar notion of random close packing (RCP). In order to study the MRJ state, it is necessary to provide a precise definition of jamming. To this end, Torquato and Stillinger have defined the following rigorous hierarchical jamming categories [4]: a locally jammed packing is one in which no particle may be moved while holding all other particles fixed. A collectively jammed packing is any locally jammed configuration in which no subset of particles can be collectively displaced with a globally-nondeformable boundary. A strictly jammed packing is any collectively jammed configuration that disallows all globally uniform volume-nonincreasing deformations of the system boundary. Note that these definitions imply that collectively-jammed packings are stable to uniform compression, and strictly jammed packings are additionally stable against shear deformations. Rigorous methods have been devised to test whether a packing is collectively
or strictly jammed [64]. In this work, we will restrict our considerations to strictly jammed packings. Note that it is not uncommon to find that some subset of spheres is jammed (the “backbone”) while the remainder are not jammed but are locally imprisoned by their neighbors (the “rattlers”). If there is no jammed backbone, then the packing is considered “unjammed”.

The literature on disordered sphere packings is in agreement with regards to sev-
eral key properties of the MRJ state for 3D monodisperse hard spheres in the infinite-system limit. For example, a variety of sensible, positively correlated order metrics produce an MRJ state with $\phi \approx 0.64$ [63, 1, 62]; and there is very strong evidence that it has an isostatic backbone [65, 19, 61, 62], meaning that it has the minimum number of interparticle contacts required by the strict jamming constraint [1, 66].

While previous efforts have successfully produced jammed packings that possess no crystalline order [66, 67, 68], it has been a challenge for previous protocols to produce hard-sphere packings that are both guaranteed to be strictly jammed and exactly isostatic—a difficulty which we address in this present work.

The difficulty in creating isostatic packings of either hard or soft spheres stems from two factors. The first is the choice of preparation protocol. It is reasonable to expect that different methods will sample different ensembles of jammed states. One example of this is that the Lubachevsky-Stillinger (LS) hard-sphere molecular dynamics algorithm can be tuned to sample jammed states with a variety of densities between 0.64 and 0.74 by varying the rate at which the spheres grow in size. Note as well that density does not uniquely characterize an ensemble of jammed packings and it follows that a density of $\phi \approx 0.64$ is not sufficient to identify a disordered packing as MRJ, as the order map in Fig. 2.1 makes clear: a collection of packings exist along the vertical line starting at the MRJ point; a partially-diluted fcc crystal, for example, exists on this line at some (presumably-high) value of $\psi$ [63, 62]. As we will demonstrate, other more subtle, yet macroscopic differences can exist between jammed, disordered packings that are all at the MRJ density, calling into question whether the traditional methods have been producing the true MRJ state.

The second difficulty in making an isostatic packing stems from a deficiency in numerical precision in obtaining a truly-jammed state within the practical constraint

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1We distinguish between hard-sphere and soft-sphere protocols because the former perform a search through available space (in which particles never overlap) and the latter search through unavailable space (in which particles can overlap until the final state). It has not been theoretically established that the two techniques approach the same limit.
of computational time. At this time, this is not due to insufficient machine precision (e.g. using quadruple-precision arithmetic instead of double-precision). All of the current (simulation) protocols used in creating disordered, jammed sphere packings rely on some sort of iterative refinement which asymptotically approaches the final jammed state, and so some judgment must be made when the algorithm has converged sufficiently; in theory, an infinite number of iterations is required to reach the final jammed state. Because a diagnosis of isostaticity relies on one’s ability to reliably identify interparticle contacts, it is crucial that a protocol be capable of producing a packing of high numerical fidelity (i.e. converging in an efficient manner) such that interparticle contacts are clearly identifiable. Distinguishing between true contacts and near-contacts is nontrivial since a disordered sphere packing will typically contain pairs of nearly-contacting spheres with very small interparticle gaps [66]; we will elaborate on this point later on.

One symptom of this challenge is manifested in the identification of rattlers. Since rattlers were first identified in disordered hard-sphere packings, different authors have offered a variety of estimates of the overall rattler fraction [51, 66, 1, 69, 29]. The considerable variability that exists among many of the reported rattler fractions can be attributed to these aforementioned difficulties (preparation protocol and simulational truncation). Clearly, a reliable characterization of the rattler population requires that the ideal jammed state be created as precisely as possible. Moreover, a more detailed characterization of the rattler population has not been carried out, leaving many assumptions about the spatial and topological characteristics of rattlers and their surroundings untested; we address several of these in this Chapter.

In spite of these mathematical imprecisions, the preponderance of (both simulational and experimental) evidence strongly suggests that rattlers are an intrinsic aspect of MRJ packings of monodisperse hard spheres. In any strictly jammed packing, rattlers owe their existence to local geometric frustration that interrupts attainment
of the maximum packing efficiency illustrated by fcc and hcp crystals. The mean-field theory of Edwards has been used to predict some of the macroscopic properties of jammed particle packings [70, 71]. However, it fails to predict the existence of rattlers altogether, further emphasizing the fact that the rattler population has been an under-studied facet of the MRJ state and underscoring the need for a more detailed theory of jamming. Furthermore, the behavior of rattlers in hard-sphere packings has unexplored applications with regards to local interaction “weak spots” in real amorphous insulator solids [72].

The Torquato-Jiao (TJ) sphere packing algorithm is well-suited to study the MRJ state—especially when one is not concerned with any dynamics or history in getting to that end state—because it is capable of producing inherent structures (mechanically stable local density maxima) possessing backbones that are guaranteed to be strictly jammed and are often highly disordered [61]. To accomplish this, the algorithm poses the packing problem as an optimization problem—to maximize $\phi$ (locally or globally [61, 73]) subject to linearized nonoverlap constraints between spheres in a deformable periodic box. The algorithm solves a sequence of linear programs (LPs) using particle translations and the symmetric strain tensor of the deformable periodic box as design variables. As a consequence, when no incremental solution to the LP exists, the system admits no further collective motions coupled with box deformations—the packing is, by definition, strictly jammed. In addition, the linear program’s solution directly encodes a contact network within which rattlers can be clearly identified and packing backbones are exactly isostatic with high probability.

Since its introduction [61], the TJ algorithm has been used to identify the densest known packings of binary sphere systems for a variety of size and number ratios [73] and to generate exactly isostatic, strictly jammed MRJ packings of bidisperse spheres [9]. Nonetheless, the algorithm is still in its infancy and its consequences are still far from being fully understood.
In this work, we show that the TJ algorithm can be used to generate disordered packings with unsurpassed numerical fidelity. This ability, combined with the TJ algorithm’s natural ability to generate inherent structures simultaneously addresses both of the aforementioned difficulties and allows one to have the clearest view of the MRJ state yet. In particular, we quantify the numerical tolerance to which our packings are prepared and demonstrate a difference of several orders of magnitude between the most distant true contact and the closest near-contact. In studying the consequences of this remarkable numerical tolerance, we present rattler fraction probability distributions for our packings as a function of system size and extrapolate an infinite-system rattler fraction that is substantially lower than previous estimates. This appears to be the result of the fact that the TJ algorithm is producing the true MRJ state with heretofore unattained accuracy in the sense that, in the large-$N$ limit, it is converging toward a generic isostatic state [74] without any added correlations beyond those required by the strict jamming constraint. We support this conclusion using the standard bond-orientational order metric and a $g_2$-based translational order metric in Sec. 2.6.

Motivated by this observation, we proceed to consider the rattler population in detail, showing significant spatial correlations through the rattler-rattler pair correlation function, contrary to conventional wisdom which assumes that they be randomly distributed. In particular, we identify clusters of rattlers sharing common cages or otherwise interacting through pair collisions which we call polyrattlers, simultaneously identifying a “bottleneck” geometry in the backbone which allows pairs of rattlers to undergo pair collisions despite being imprisoned in distinct cages. Using this information, we decompose the rattler pair correlation function’s contributions from interacting and non-interacting rattlers and find that the former are almost exclusively responsible for the sharp increase in the pair correlation function as contact is approached. Clusters of up to 5 rattlers are contained within our packings, show-
ing that MRJ packings of monodisperse spheres can contain large rattler cages while still obeying the strict jamming criterion. We have included graphs which describe all of the polyrattler topologies which we have observed as well as images of several polyrattler configurations that show a variety of notable features. Finally, we observe that the backbone spheres that form rattler cages are significantly hypostatic as a whole (i.e. possessing fewer than 6 contacts per sphere), implying that there must be other regions within the packing that are hyperstatic (possessing more contacts than the isostatic number). Importantly, since the rattler fraction is lower in our packings compared to other protocols [54, 66, 55, 61], the occurrence of such hyperstatic regions should also be suppressed. Conversely, by controlling the occurrence of spheres with high coordination numbers, one might be able to tune the rattler fraction of a sphere packing.

The rest of this Chapter is organized as follows: In Sec. 2.2, we provide an overview of the TJ algorithm and supply the relevant simulational parameters we used in this study; In Sec. 2.3, we present the results which we have mentioned in the previous paragraph; in Sec. 2.4, we offer conclusions, discuss the significance of our results, and propose ways in which our work might be extended.

2.2 Method

We use the TJ algorithm to generate strictly jammed MRJ packings of monodisperse spheres with periodic boundary conditions for system sizes $N = 100, 200, 500, 1000,$ and 2000 spheres per packing; the details regarding the implementation of this algorithm can be found in [61], but we provide a brief sketch of the algorithm here for the sake of completeness.

The TJ algorithm accepts as input any hard-sphere packing; we consider here packings generated via random sequential addition (RSA) within a fundamental cell.
with periodic boundary conditions. The densification process is an iterative procedure driven by solving linear programs (LPs). Since the goal of the protocol is to maximize \( \phi \) (or, in the spirit of the energy landscape picture [61], minimize \(-\phi\)), we may pose an objective function in terms of a strain tensor acting on the fundamental cell. A linearization gives the following objective function for the LP:

\[
\text{minimize } Tr(\epsilon) = \epsilon_{11} + \epsilon_{22} + \epsilon_{33} + \cdots + \epsilon_{dd},
\]

where \( \epsilon \) is a strain tensor that deforms the fundamental cell described by a generating matrix \( \Lambda \). This implies that the components of the strain tensor \( \epsilon \) are design variables in the LP. The other design variables are the displacements for each sphere (in the lattice coordinate system), \( \Delta x_1^\lambda, \Delta x_2^\lambda, \Delta x_3^\lambda, \ldots, \Delta x_N^\lambda \); the superscript lambda denotes that the vectors are expressed in terms of \( \Lambda \). Since no two spheres can overlap in a hard-sphere packing, our LP’s constraints must reflect that \( r_{ij}^G \), the (global) distance between the centroids of spheres \( i \) and \( j \), with diameters \( D_i \) and \( D_j \), respectively; be \( r_{ij}^g \geq (D_i + D_j)/2 \). Expressing this in terms of the spheres’ lattice coordinates, and taking into account the spheres’ displacements and deformable fundamental cell, we have

\[
\sqrt{\Lambda \cdot r_{ij}^\lambda \cdot (1 + \epsilon)^2 \cdot \Lambda \cdot r_{ij}^\lambda} \geq (D_i + D_j)/2,
\]

the relative displacement \( r_{ij}^\lambda = (x_j^\lambda + \Delta x_j^\lambda) - (x_i^\lambda + \Delta x_i^\lambda) \). Linearizing this in the sphere displacements and the strain tensor gives

\[
\Lambda \cdot r_{ji}^\lambda \cdot \epsilon \cdot \Lambda \cdot r_{ji}^\lambda + \Delta x_i^\lambda \cdot G \cdot r_{ji}^\lambda + \Delta x_j^\lambda \cdot G \cdot r_{ij}^\lambda \geq \frac{1}{2} \left((D_i + D_j)/2 - r_{ji}^\lambda \cdot G \cdot r_{ji}^\lambda\right) + R
\]

where \( G = \Lambda^T \cdot \Lambda \) is the Gram matrix of the lattice \( \Lambda \) and \( R \) contains all of the higher-order terms; in practice, it is acceptable to let \( R = 0 \). This constraint must be posed for all pairs of spheres that are close to each other and therefore at risk of overlapping; to do this, an “influence sphere” of radius \( \gamma \) is defined such that a
constraint will be included in the LP for any pair of spheres whose centroids are separated by less than \((D_i + D_j)/2 + \gamma\). Since Eq. (2.2) is a local linearization of a quadratic constraint, we impose an artificial limit on the extent of the design variables in each iteration to preserve the accuracy of the linearization.

Having defined an objective function and constraints, we solve the LP to determine how the packing will be rearranged and densified. After applying the sphere displacements and lattice deformation, the LP is reformulated using the new sphere positions and fundamental cell tensor. The process is iterated until the solution converges and the packing does not change by more than some termination threshold. We have found that the most effective threshold is the fundamental cell volume; when the cell volume fails to decrease by an appreciable amount, the packing is jammed to a corresponding precision.

We use initial conditions generated by random sequential addition (RSA) at an initial density \(\phi_{\text{init}} = 0.1\) in a fundamental cell with unit volume. We have found that, for densities significantly under the RSA saturation density, the resulting packings are largely insensitive to \(\phi_{\text{init}}\). Packings are compressed using the TJ algorithm with an “influence sphere” of radius \(\gamma = D/40\) [61], where \(D\) is the sphere diameter. For sufficiently large system sizes (e.g. \(N \geq 100\)), the size of the influence sphere does not affect the final state of the packings to any detectable extent. For a single LP iteration, box deformations (both normal and shear movements) are limited in magnitude to less than \(D/200\) and sphere translations are limited to \(|\Delta r_i| \leq D/200\). The algorithm is terminated when two successive compressions fail to decrease the lattice volume by \(V_k - V_{k-2} < 3.0 \times 10^{-12}\), where \(V_k\) is the volume of the fundamental cell on iteration \(k\). We chose to limit ourselves to \(N = 2000\) so that we can generate 1000 packings per system size for accurate statistics\(^2\).

\(^2\)The average time required to generate one packing with \(N = 2000\) to the tolerance used in this paper was about 6.3 hours on a single thread running at 2.40 GHz using the Gurobi linear programming solver.
2.3 Results

We use the protocol described in the previous section to generate 1000 packings of system sizes, \( N = 100, 200, 500, 1000, \) and \( 2000 \), yielding 5000 packings in total. The following details subsections characterize various aspects of the packings.

2.3.1 Isostaticity

Before presenting packing statistics, we first establish that the \( N \)-particle packings are indeed strictly jammed and isostatic using the exact relations given in [64]. For frictionless spheres, there are \( d(N_B - 1) \) degrees of freedom associated with translating the spheres (up to uniform translations of the whole packing under periodic boundary conditions), where \( N_B \) is the number of (jammed) backbone spheres. The simulation box is allowed to deform for strict jamming and thus there are \( d(d + 1)/2 \) additional degrees of freedom associated with straining the unit cell, totaling \( F_s = d(N_B - 1) + d(d + 1)/2 \) degrees of freedom which must be constrained. Since the nonoverlap constraints are inequality constraints, \( F_s + 1 \) of them are required. Since the system volume cannot increase, the first constraint is \( \text{Tr}(\epsilon) \leq 0 \). Therefore, the number of other constraints—contact pairs—is equal to the number of degrees of freedom; for \( d = 3 \), this corresponds to \( 3N_B + 3 \) contact pairs [66], or an average contact number of \( \bar{z} = 6 + 6/N_B \).\(^{34} \) Our packings prepared using the TJ algorithm have precisely this many contact pairs.

\(^3\)If we consider collective jamming, we drop the degrees of freedom associated with straining the box, and we also drop the trace constraint with it, yielding \( 3N_B - 2 \) contact pairs, or \( \bar{z} = 6 - 4/N_B \).

\(^4\)Both results for collective and strict jamming differ by one contact pair from those given in [43] and [30]. This means that, for both collective and strict jamming criteria, Dagois-Bohy et al. have created isostatic packings through their soft-sphere protocol. The discrepancies stem from their assumption that the number of constraints ought to be equal to the number of degrees of freedom; this is only true if the constraints are equality constraints; since spheres in contact are not “stuck” together, this is not the case.
2.3.2 High-Fidelity Strictly Jammed Packings

First, we quantify the precision to which our packings are made by considering the contact tolerance \( \delta \) (the numerical separation distance of particles that are actually touching). When \( \delta = 0 \), no spheres are in contact, and the packing is unjammed. Increasing \( \delta \) introduces interparticle contacts, and the packing becomes jammed at some minimum tolerance \( \delta_{\text{min}} \). A feature of the TJ algorithm is that the feasible tolerance with which the linear programs are solved is intimately related to \( \delta_{\text{min}} \). The packing first becomes strictly jammed (i.e. \( \delta_{\text{min}} \) is found) when the isostatic number of contacts are formed. The contact tolerance at which the first excess contact is introduced is \( \delta_{\text{max}} \), and corresponds to the point at which a near-contact is mislabeled as a true contact. It is therefore imperative that the numerical fidelity of the packing allow for a significant difference between \( \delta_{\text{min}} \) and \( \delta_{\text{max}} \). If not, then there is a risk of confusing near-contacts with true contacts, thereby finding the wrong contact network and mislabeling rattlers and backbone spheres. For our 1000 packings generated with \( N = 2000 \), \( \delta_{\text{max}} - \delta_{\text{min}} \) is between 2 and 7 decades, with a median of over 4 decades; the mean for \( \delta_{\text{min}} \) is \( 10^{-11} D \), with most instances falling within one order of magnitude, and \( \delta_{\text{max}} \) is typically, at most, \( 10^{-6} D \). \( \delta_{\text{max}} \) decreases as \( N \) increases since near-contacts may be found at arbitrarily small separations in the infinite-system limit [66]. In addition to the high numerical fidelity which the TJ algorithm allows us to attain, the linear programming method possess a unique safeguard in that, by investigating the active and nonactive constraints in the final LP solution, one may directly identify interparticle contacts and so check the result obtained by choosing a contact tolerance. Therefore, the packings generated with TJ are guaranteed to have accurate backbone information. Further details are given about our packings' numerical fidelity in Sec. 2.5.

\[5\] In linear programming, an active constraint is one that is satisfied as an equality; a nonactive constraint is satisfied as a strict inequality.
2.3.3 Rattler Fraction Distribution Function

Having created packings with robust isostatic contact networks, we now consider the behavior of the rattler fraction as a function of system size. To do this we compute the probability distribution of the rattler fraction $N_R/N$ for each system size; the result is plotted in Fig. 2.2. These distributions are very closely fit by appropriately-scaled binomial distributions. For system sizes $N < 500$, we have observed packings that do not have any rattlers in them, though this probability decreases with increasing system size, owing to the narrowing of the distribution (the standard deviation of the distribution scales as $N^{-1/2}$); the probability that a packing with $N = 500$ has no rattlers is estimated to be about one in two thousand.

Our results show a mean rattler fraction that asymptotically approaches a sharply-defined infinite-system limit of $\lim_{N \to \infty} N_R/N = 0.015$. This value is significantly lower than the estimated 0.025 to 0.030 characteristic of the Lubachevsky-Stillinger (LS) algorithm [54, 66, 55, 61]. Because the packings created by the TJ algorithm are exactly isostatic inherent structures by design (unlike those generated by LS), we conclude that the TJ algorithm is generating packings closer to the true MRJ state than ever before in the sense described in the Introduction. We have confirmed this claim by computing bond-orientational and translational order metrics on both the configurations generated by TJ and some generated by LS; see Sec. 2.6 for details. This implies that the MRJ state has a substantially smaller rattler fraction than what was previously thought. The packing density approaches an infinite-system limit of $\lim_{N \to \infty} \phi(N) = 0.639$, with fluctuations decaying as $N^{-1/2}$, in agreement with past results for monodisperse hard-sphere systems [62].

2.3.4 Rattler Pair Correlation Function

Next, we extract higher-order statistical spatial information about the rattlers and backbone through computing the pair correlation function for rattler-rattler and
Figure 2.2: Probability density functions for the rattler fraction $N_R/N$ plotted for various $N$. The inset shows the mean rattler fraction as a function of $N$ with vertical bars denoting fluctuations of two standard deviations (error bars are too small to be seen). Note that $N_R/N$ asymptotically approaches $\lim_{N \to \infty} N_R/N = 0.015$ with fluctuations narrowing proportionally to $N^{-1/2}$.

Both the backbone and rattlers exhibit similar oscillatory behavior, but $g^R_2(r)$ behaves more smoothly, lacking the sharpness in the peaks at $r/D = \sqrt{3}$ and $r/D = 2$ that are found in the backbone pair correlation function, which usually correspond to the presence of coplanar double-triangles of particles and three collinear particles, respectively [66, 75]. The rattlers’ marked spatial pair correlations dispel the traditional notion that they are randomly distributed in a packing [76]. Rather, our work
suggests that the correlations between rattlers owe directly to the strong influence of the surrounding backbone.

In addition, it is significant that the oscillations in $g^R_2$ are delayed in phase compared to those found in $g^B_2$. This might be linked to the extra available space which a cage allows around a rattler. Two rattlers separated by a similar geometric configuration as found in the backbone would have this extra distance due to the “buffer” created by the cages. Importantly, $g^R_2$ rises dramatically near contact, and conceivably could be consistent with a divergence\textsuperscript{6}.

![Figure 2.3: Rattler-rattler and backbone-backbone pair correlation functions as a function of dimensionless separation $r/D - 1$ averaged over 1000 packings for $N = 2000$. The rattlers’ spatial distribution shows strongly-correlated behavior including the existence of $n$-rattlers ($n \geq 2$), as evidenced by the very steep increase near contact. $g^B_2$ rises dramatically as the contact value is approached; the left most point (not shown) is $g^B_2(0.025) = 14.79$.]

\textsuperscript{6}The possibility of a divergence is based upon the observation that $g^R_2(1/40) = 14.79$ which is greater than than that of $g^B_2$, which is known to diverge at contact. Further exploration of the bin size effect must be done to substantiate this possibility.
2.3.5 Polyrattlers

The sharp increase in $g_2^R$ near contact implies the existence of “polyrattlers”—rattler clusters that are either imprisoned within the same cage or otherwise interact through pair collisions. Motivated by this finding, we proceed to identify and characterize the polyrattlers inside our jammed packings inside which rattlers may interact directly through pair collisions. The single-particle available space, $a_i$, of (rattler) sphere $i$ is defined as the locus of all positions covered by the rattler sphere under continuous displacements while obeying the nonoverlap constraints with the backbone spheres. Note that all other rattlers are disregarded when determining a rattler’s single-particle available space.

The only way for a polyrattler to exist in two dimensions (2D) is for both rattlers to be in the same cage (and therefore have identical $a_i$’s). However, in three dimensions (3D), a distinct geometry is possible in which rattlers may pair-collide, but occupy different cages. In this case, rattlers are separated into different, yet partially-overlapping cages by the presence of “bottlenecks” in the jammed backbone. Two examples of 2-rattlers (polyrattlers of cluster size 2)—one within a single common cage, and one with a bottleneck—are shown in Figures 2.4a and b, respectively. The “bottleneck” geometry is very significant when considering the nature of rattler clusters in 3D: 2-rattlers (polyrattlers with cluster size 2) are almost twenty times more likely to be separated by a bottleneck than be in the same cage. Moreover, 2-rattlers separated by a bottleneck have a mean separation distance of $1.1 \times 10^{-2}D$, whereas those in the same cage have a mean separation of $2.4 \times 10^{-3}D$.

By classifying all pairs of rattlers as either (1) not interacting, (2) interacting through a bottleneck, or (3) in the same cage, we can decompose the pair correlation function $g_2^R$. Doing this, we find that the major contributor to the sharp increase near contact indeed comes from polyrattlers. This decomposition also shows that the polyrattler contribution to $g_2^R$ becomes small quickly beyond contact.
Figure 2.4: (a) A 2-rattler where both rattlers (solid red spheres) are in the same cage (translucent spheres). (b) A 2-rattler with a bottleneck. The rattlers (light red and dark blue solid spheres). The cage spheres (translucent) are colored according to which rattler they enclose (either light red or dark blue), and the two “bottleneck” spheres (translucent green spheres marked with solid dots in their centers) contribute to the cage of both rattlers. See the Supplemental Material for [3] for rotating animations of these Figures.

Cages do not typically allow much space for rattlers to move. For monorattlers, the mean distance between the rattler and a cage sphere is about $0.01D$, and the probability that a cage sphere is some distance from the rattler decays rapidly with increasing distance from contact. In addition, our packings are saturated (no void exists which is large enough to allow for the insertion of an additional sphere), reinforcing previous investigations using LS [56].

Having established the existence of polyrattlers, we identify and enumerate all of the polyrattlers that we observe in our packings up to a system size of $N = 2000$. Figure 2.6 shows the rattler fraction contributions according to the cluster size, $n$, of the polyrattler. The contributions decay approximately exponentially in cluster size, and do not depend strongly on system size. The insets in Fig. 2.6 show the topologies of the polyrattlers that we find: nodes correspond to rattlers, and interacting pairs (i.e., with overlapping $a_i$’s) are connected by edges. The discovery of polyrattlers with cluster size up to 5 is significant because it shows, surprisingly, that 3D MRJ monodisperse packings can contain large rattler cages while still obeying strict jam-
Figure 2.5: Rattler-rattler pair correlation function averaged over 1000 packings for $N = 2000$, separated according to the three different types of interactions: non-interacting pairs (black curve), pairs interacting through a bottleneck (green squares), and pairs sharing a common cage (red filled circles). This decomposition shows that the near-contact regime is dominated by interacting pairs, and that pairs interacting through bottlenecks can be found at comparatively large separations. Note that the ordinate in this Figure is expressed in a log scale unlike Fig. 2.3.

Figure 2.7 shows one such 5-rattler which we identified; it is composed of three rattlers in a common cage, plus two additional rattlers separated into their own cages by bottlenecks. How large a polyrattler can be found in an MRJ packing in the infinite-system limit? If there is an upper limit on polyrattler size, we believe that it may be substantially larger than what we have observed so far via simulations or has been appreciated in the literature.

\(^7\)It is considerably easier to form rattlers in 2D bisperse systems [19] both because of the lower dimensionality and the particles' size dispersity.
Figure 2.6: Rattler fraction contributions according to polyrattler cluster size $n$ and system size $N$. Diagrams show examples of $n$-rattler topologies. Nodes represent rattlers, and edges connect rattlers with overlapping $a_i$‘s. Chain-like 3-rattlers occur about twice as frequently as the triangular variety; due to the large diversity and rare appearance of 4- and 5-rattlers, we do not comment here on their occurrence frequency.

### 2.3.6 Rattler Cage Coordination

The backbone cage spheres surrounding rattlers are typically significantly hypostatic, with a mean coordination number of $\bar{z} = 5.76$. This suggests that rattlers arise in regions within the packing where the local coordination structure becomes so sparse that spheres cease to be sufficiently supported to be jammed. A contact distribution for the cage spheres (generated from our $N = 2000$ packings) is shown in Fig. 2.8. The shape of this contact distribution is significantly different than that of the whole packing, demonstrating a concrete geometrical feature which is characteristic of the neighborhood near rattlers.

In order for the backbone to be isostatic as a whole, there must be other locally-hyperstatic regions in the packing in order to compensate for these hypostatic regions.
Figure 2.7: A 5-rattler with three rattlers occupying the same cage (solid light pink spheres). The other two rattlers (solid red spheres) interact with the 3-rattler cage through bottlenecks, and do not interact with each other directly. The cage spheres are translucent.

If one assumes that areas with increased coordination are undesirable in an MRJ packing\(^8\), then, by limiting the number of rattlers in a packing, one may also limit the occurrence of these locally-hyperstatic regions. This reinforces the notion that the suppressed occurrence of rattlers in packings generated by TJ allows it to come closer to the MRJ state than ever before. As a corollary, controlling the occurrence of highly-coordinated spheres may be one method by which one may, in turn, control the rattler fraction in a packing.

\(^8\)This is reasonable since there is generally a positive correlation between coordination and order in jammed packings [77].
Figure 2.8: The contact distribution for backbone spheres that make up rattler cages in our $N = 2000$ ensemble of packings. The mean contact number is $\bar{z} = 5.76$ and the shape of the distribution is significantly different from that of the whole backbone (shown for comparison); the inset quantifies the difference between the two distributions as a function of $z$.

2.4 Conclusions and Discussion

The TJ algorithm allows one to accurately identify the contact network of jammed, disordered sphere packings. We have used this capability to prepare thousands of strictly jammed, exactly isostatic packings of monodisperse spheres of high numerical fidelity. From these packings, we have shown the probability distribution of rattler fractions as a function of system size and have extrapolated from our data an infinite-system limit value of $\lim_{N \to \infty} N_R / N = 0.015$, which differs significantly from previous estimates. This is because the TJ algorithm is coming closer to the true MRJ state than ever before, as shown by standard order metrics (See Sec. 2.6). Motivated by this finding, we investigated the geometrical and topological particulars of the rattlers in our MRJ systems. The rattler population displays significant spatial correlations.
similar to the backbone, prompting us to investigate the cages they inhabit. Rattler clusters are formed through rattler that occupy a common cage as well as by rattlers that interact through a “bottleneck” configuration that does not occur in 2D; these interacting rattlers constitute the majority of rattler pairs with a small pair distance, as demonstrated by a decomposed rattler pair correlation function. In addition, we found polyrattlers with surprisingly large cluster sizes within our strictly jammed packings. Lastly, the backbone spheres which encage rattler spheres are often significantly hypostatic, implying a possible connection between rattlers and areas of highly-coordinated spheres in exactly-isostatic monodisperse hard-sphere packings.

It is interesting that the TJ algorithm generates packings that have significantly fewer rattlers than the LS algorithm. The fundamental reason for this difference of the fact that TJ and LS utilize completely different dynamics. The biggest of these consequences is that packings generated using TJ are guaranteed to be strictly jammed; the same is not necessarily true for packings generated with LS, even though the pressure may diverge. In fact, it has been known for a long time that if the expansion rate is not carefully monitored during the LS algorithm, the pressure will diverge prematurely and the packing will be hypostatic; subsequent equilibration reveals the unjammed nature of such packings. This challenge becomes more prominent as the system size increases—a smaller final expansion rate must be used, and an unjamming motion takes longer to show up (if it exists) [66].

We have found that substantial computational time must be put into the final minute rearrangements in a packing’s structure to cause jamming; because LS searches these rearrangements using the dynamically-indirect heuristic of random collisions from molecular dynamics, it becomes increasingly challenging to generate exactly jammed packings as the system size becomes large [66]. By contrast, the ability of TJ to use linear programming to directly search for optimal rearrangements near the jamming limit makes it ideally-suited at the final approach to jamming. In addition,
TJ can also quickly identify unjamming motions in seemingly-jammed packings prepared by other protocols. Therefore, it will be instructive to investigate the result when packings that are initially prepared through other protocols are subsequently given to TJ for final densification and jamming.

Moreover, TJ and LS tend toward different configurations due to the dynamics at play. On one hand, the TJ algorithm seeks to maximize the density of the packing within a local neighborhood, and is free to choose any collective displacement of spheres in order to achieve this goal, meaning that the resulting configurations are always local density maxima\(^9\). By contrast, the molecular dynamics used in LS are constantly equilibrating the packing during the slow compression, which will cause it to tend to avoid local density maxima. The result is that packings generated with LS tend towards the global density maximum, and one must take special care to divert it from achieving this goal when preparing disordered packings. Therefore, while it is certainly possible to create disordered packings using LS, TJ is, by contrast, naturally suited to generate maximally random packings. While both protocols may create disordered packings with similar density, one should clearly expect that the packings be fundamentally different; the pronounced difference in rattler fraction validates this expectation.

What does the lower rattler fraction tell us about the jammed states that TJ is accessing? The qualitative difference between TJ and LS packings points out an ambiguity which causes one to call into question whether or not the standard protocols have been truly producing the MRJ state, as has been taken for granted. As we have shown for our packings, the cages around rattlers are noticeably hypostatic. Because the whole backbone is known to be exactly isostatic, one should expect that there are other regions in the packing which are significantly hyperstatic. It is already known that any departure from isostaticity in a jammed packing must increase the packing’s

\(^9\)The linearization of the packing problem in TJ is sufficient in practice to limit the algorithm’s access to regions of configuration space that lead to crystallization.
order, regardless of whether that increase in coordination comes with an increase or decrease in density [45, 63, 1, 62]. Therefore, it is not unreasonable to expect that the suppression of rattlers in packings generated by TJ comes hand in hand with a suppression of hyperstatic subregions which would bring the packings away from the MRJ state. In addition, the small amount of available space in the rattler cages implies that the amount of ordering required to create a rigid cage is minimized.

It is all the more surprising that the rattlers made by TJ, though more dilute, are still strongly correlated, as opposed to what conventional wisdom dictates. More still, that one can have clusters of rattlers as large as we have shown while obeying the strict jamming criterion is remarkable. A key mechanism for this is the bottleneck geometry which allows a packing to place cages adjacent to one another. Therefore, rattler clusters which use the bottleneck geometry avoid the alternative, substantial challenge—to form a single cage capable of fitting a large number of rattlers. Therefore, while one certainly might observe substantially larger polyrattlers than the 5-rattlers we have found here thanks to the assistance of bottlenecks, it is much less clear that MRJ packings will exhibit similarly large single cages; the largest such cage we have found here held three rattlers inside. Clearly, we see that there is rich behavior to be found amidst the rattler-backbone interactions in disordered, jammed sphere packings, highlighting the need for a statistical mechanical theory that is capable of accounting for such behavior.

It is conjectured that all strictly jammed, saturated packings are hyperuniform [1]. Interestingly, removing rattlers from MRJ packings of monodisperse spheres results in a deviation in the structure factor in the limit \( \lim_{k \to 0} S(k) \) [56], and it was thought that the magnitude of this deviation would be accurately predicted from the assumption that the rattlers be Poisson-distributed. However, our work has shown that there is significant spatial ordering of the rattlers, and so we predict that the actual deviation from hyperuniformity should be markedly smaller than this prediction. A related
question is whether or not $g^R_2$ exhibits the same quasi-long-range behavior as the whole packing, i.e., $g^R_2 - 1$ decaying as $-1/r^4$. To show this, one must construct very large MRJ packings with very high numerical fidelity—a nontrivial task, as we have shown even for modest system sizes. In the future, we will consider issues of hyperuniformity in considerably larger packings to this end.

It is already known that the existence of rattlers in packings is sensitive to the particle shape; MRJ packings of monodisperse superballs, convex polyhedra, and even some ellipsoids do not exhibit rattlers at all [78, 79, 74]. It is still unclear what makes spheres “special” in that rattlers are such a prominent feature in their MRJ state. Therefore, it is an interesting question whether the rattler fraction can be incorporated as a tunable parameter in sphere packing protocols; as we have pointed out, controlling the occurrence of hyperstatic regions is a good place to start. Moreover, in the same way that there is a maximum packing density that can be achieved, there must be some upper limit to the rattler fraction in a jammed packing which is nontrivial to identify; “tunneled crystals” (which contain chains of vacancies that permeate the structures) present a starting upper bound, since they have the lowest known density among strictly jammed packings [2]. On the other hand, it would be interesting if the rattler fraction could be tuned to decrease towards zero. If so, one might be able to answer the larger question of how rattlers affect the large-scale properties of a packing and offer additional insight into the MRJ state.

The situation of geometric frustration which produces rattlers in hard sphere packings appears to be connected to an analogous feature arising in real amorphous insulator solids. Specifically, the latter incorporate local interaction frustration “weak spots” compared to the crystalline forms. This gives rise to low-temperature heat capacity and thermal conductivity anomalies due to quantum tunneling in two-level localized degrees of freedom [72, 80, 81, 82, 83]. The rattlers examined herein evidently represent the outcome for amorphous solids as continuous, realistic interactions
pass to the discontinuous hard-sphere limit. Therefore, we suggest that it would be important to explore the evolution of weak spots in amorphous materials as one deforms the potential function continuously from the model potential used to describe them to the hard-sphere model in which rattlers are observed.
2.5 Appendix A: Numerical Fidelity and Contact Distributions of MRJ Sphere Packings Generated Using the Sequential Linear Programming Method

2.5.1 Contact Network Fidelity

Figure 2.9 shows the probability density functions and cumulative distribution functions for the minimum and maximum contact tolerances $\delta_{\text{min}}$ and $\delta_{\text{max}}$ for our $N = 2000$ packings. We also present this information by percentile in Table 2.1. Not only do individual packings show a good separation between $\delta_{\text{min}}$ and $\delta_{\text{max}}$ (as seen in Fig. 2.10, which present this data as a scatter plot for all of our system sizes), but we have prepared our packings to such a high degree of fidelity that there is no overlap between the two probability density functions. The sharp rise at the end of $pdf(\delta_{\text{min}})$ is due to the truncation criterion for our simulations.

Table 2.1: Percentiles for $\delta_{\text{min}}$ and $\delta_{\text{max}}$ for 1000 MRJ configurations with $N = 2000$.

<table>
<thead>
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</table>
Figure 2.9: Log-weighted probability density function and cumulative density function for $\delta_{\min}$ and $\delta_{\max}$. Notice that there is no overlap between the two distributions, indicating a generally-robust separation between true contacts and near-contacts.

### 2.5.2 Contact Distribution

Because of both the high fidelity of our packings and the high precision to which contacts are identified, we are able to present a contact distribution for the backbone spheres in our packings which exhibits a system size dependence on $Pr(z)$ for any given $z$. This arises as a direct consequence of the fact that all of our packings are exactly isostatic; the finite system size contribution to the isostatic number which makes the average coordination number slightly above 6 for finite systems is responsible for the increase in $Pr(z)$ when $z < 6$ and decrease when $z > 6$ as $N$ increases towards infinity. Given this physical intuition, one might expect that the contact distribution for the MRJ state have a functional form of $Pr(z; N) - \lim_{N \to \infty} Pr(z; N) = cN^{-\alpha}$ with a critical exponent $\alpha = 1$ owing to the nature of the finite size correction to the isostatic number. We present the contact distribution for $N = 2000$ in Table 2.2
Figure 2.10: A scatter plot of $\delta_{\text{min}}$ and $\delta_{\text{max}}$ for the packings generated for various $N$. The red shaded region illustrates where $\delta_{\text{min}} \geq \delta_{\text{max}}$, which is disallowed by definition. Packings which are far from this region clearly differentiate between physically-justified near-contacts and true contacts corresponding to the numerical precision of the TJ algorithm. In the best cases for $N = 2000$, the first near contact has a pair separation seven orders of magnitude larger than the biggest contact, whereas the average packing has a separation of about four orders of magnitude. This gap generally increases for the smaller system sizes.

since these values should be reasonably close to those of the infinite system limit\textsuperscript{10}. We also point out that one sphere with $z = 12$ was found in each of our ensembles of packings with $N = 100, 500, \text{and } 1000$. The coordination structure about these spheres is always approximately icosahedral but with some distortions. Since a twelve-fold coordination scenario does not necessarily correspond to a crystalline arrangement such as fcc or hcp, we do not regard these observations as necessarily

\textsuperscript{10}The values for $Pr(z, N = 2000)$ are all within the 95% confidence interval of an extrapolation towards the infinite system limit of the form mentioned in the text. Studying larger system sizes is essential for refining the extrapolation.
being incompatible with the MRJ state. Our results are in agreement with previous estimates of the contact distribution [66].

Figure 2.11: The contact distribution for the backbone spheres in our packings, averaged over 1000 isostatic configurations with $N = 100, 200, 500, 1000, \text{ and } 2000$. Vertical bars represent two standard deviations (error bars corresponding to a 95% confidence interval are smaller than the markers). The probabilities for $z = 11$ are very small but nonzero. One sphere with $z = 12$ was found in each ensemble of packings with $N = 100, 500, \text{ and } 1000$.

2.6 Appendix B: Order Metric Calculations for Disordered Packings

In order to quantify the order in our packings, we consider the well-known bond-orientational order metric $Q_6$ [46], which is normalized so that the FCC crystal yields
Table 2.2: The contact probability distribution for MRJ monodisperse hard spheres for a system size $N = 2000$.

<table>
<thead>
<tr>
<th>$z$</th>
<th>$Pr(z)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>0.1252 ± 3.6 × 10^{-4}</td>
</tr>
<tr>
<td>5</td>
<td>0.2423 ± 5.2 × 10^{-4}</td>
</tr>
<tr>
<td>6</td>
<td>0.2895 ± 5.8 × 10^{-4}</td>
</tr>
<tr>
<td>7</td>
<td>0.2182 ± 4.8 × 10^{-4}</td>
</tr>
<tr>
<td>8</td>
<td>0.0992 ± 3.3 × 10^{-4}</td>
</tr>
<tr>
<td>9</td>
<td>0.0233 ± 1.9 × 10^{-4}</td>
</tr>
<tr>
<td>10</td>
<td>0.0022 ± 6.4 × 10^{-5}</td>
</tr>
<tr>
<td>11</td>
<td>5.480 × 10^{-5} ± 1.0 × 10^{-5}</td>
</tr>
</tbody>
</table>

An order metric of unity; defined as

\[
Q_l = \left[ \frac{4\pi}{2l + 1} \sum_{m=-l}^{l} |Y_{lm}(\theta, \phi)|^2 \right]^{1/2},
\]

where $Y_{lm}$ is the spherical harmonic function whose average is computed over all contacting pairs of spheres. Figure 2.12 shows the probability density function for $Q_6$ for 1000 packings created using LS with $N = 2000$ under rapid compression as well as our 1000 packings created using TJ with $N = 2000$. While the packings generated with TJ are clearly more disordered on average, our findings demonstrate that $Q_6$ is a poor order metric when discerning differences between highly-disordered packings. This is reasonable because $Q_6$ can be thought of as measuring to what extent a packing deviates from a perfect crystal; far from this reference state, it becomes less discriminating. It is therefore all the more remarkable that a difference between the two protocols is discernible using $Q_6$.

Motivated by the discrepancy that is nonetheless evident for $Q_6$, we compute the $g_2$-based order metric $T^*$ [84], which makes no assumption about a most-ordered reference state; defined as

\[
T^* = \frac{\int_{\xi_c}^{\xi_c} \left| g_2(\xi) - 1 \right| \, d\xi}{\xi_c - Dp^{1/3}}.
\]

42
Figure 2.12: Probability density functions for the dimensionless order metric $Q_6$ for packings generated with LS and TJ with system size $N = 2000$.

where $\xi = r\rho^{1/3}$, $\rho = N/V$ is the number density, and $\xi_c$ is a cutoff value, chosen here to be 3.5. The qualitative behavior of $T^*$ tends to be independent of $\xi_c$ as long as the first several coordination shells are included within the integration domain. Figure 2.13 shows the probability density function for $T^*$ for the same collections of configurations. $T^*$ is more sensitive with these disordered configurations, and there is a clear distinction between the two protocols. This can be explained by noting that $T^*$ can be thought of as a “disorder metric” in that its reference state is the Poisson process, for which $T^* \equiv 0$. MRJ-like packings, being more similar to a Poisson process than a regular crystal, are therefore more easily distinguished by the $T^*$ order metric.

While we have only considered two of many order metrics here, the evidence we present is sufficient to demonstrate a statistically-significant distinction between LS and TJ packings with regards to order. Moreover, the distributions presented here tend to narrow with increasing system size as $1/\sqrt{N}$ (data not shown), and so we
expect that these distributions will approach delta functions in the infinite system limit. Examining the full picture with regards to other order metrics is a subject for future work.
Chapter 3

Existence of isostatic, maximally random jammed monodisperse hard-disk packings

3.1 Significance

Disordered particle packings are ubiquitous in all areas of science. While disordered jammed packings of hard-spheres are readily observed in 3D, the story is quite different for disks. Most 2D packing protocols tend to produce highly ordered disk arrangements, suggesting that truly disordered jammed disk packings might not exist in 2D. The maximally random jammed (MRJ) configurations we observe have a complete lack of crystallinity and are distinct from the typical jammed configuration that is most probable upon rapid compression. A protocol-independent geometric-structure approach allows us to create, identify, and analyze these previously-elusive packings. Our results shed new light on the nature of randomness, which is an issue that arises across the physical, mathematical, and biological sciences.
3.2 Introduction

A packing in $d$-dimensional Euclidean space $\mathbb{R}^d$ is defined as a collection of particles that do not overlap with one another. In three dimensions (3D), hard particle packings have served as simple, yet powerful models for a wide variety of condensed matter systems including liquids, glasses, colloids, particulate composites, and biological systems, to name a few [12, 14, 20, 24, 31]. In two dimensions (2D), they have been used to model systems such as the molecular structure of monolayer films [38, 39], adsorption of molecules on substrates [36, 37], and the organization of epithelial cells [40, 41]. Moreover, particular interest has been devoted towards packings that are jammed (roughly speaking, packings that are mechanically stable) [4, 19, 22, 26, 1, 29, 30].

Jammed packings of monodisperse spheres in 3D exist over a wide range of packing fractions from $\phi = \frac{\pi}{\sqrt{18}} \approx 0.74048 \ldots$ to $\phi = \frac{\pi\sqrt{2}}{9} \approx 0.49365 \ldots$, where the former corresponds to the fcc lattice, and the latter corresponds to the “tunneled crystals” [2]. In addition, jammed packings exist with intermediate packing fractions and a wide variety of order, including packings that are fully noncrystalline. Of particular interest is the “maximally random jammed” (MRJ) state, defined as the packing that minimizes some scalar order metric $\psi$ subject to the jamming constraint, replacing the familiar notion of random close packing (RCP) [45], originally defined as the densest configuration that a “random” packing could attain without ever defining “randomness.” The concept of the MRJ state is a natural outcome of the geometric-structure approach, in which packings are analyzed primarily on an individual basis.

The situation for disordered monodisperse disk packings in 2D is very different from the 3D counterpart because the former lacks geometrical frustration. Specifically, the globally-densest packing of disks (the triangular lattice) is compatible with the densest local packing (a triangle). However, in 3D, there is an incompatibility between the globally-densest packing of spheres (the FCC lattice) and the densest local packing (an icosahedral arrangement). As a result, packing protocols have struggled
to generate disordered, jammed packings of monodisperse disks, suggesting that the most disordered packing is a polycrystalline arrangement—a dubious proposition for the MRJ state. Nonetheless, a truly disordered, jammed disk packing has remained elusive. Here we show by construction, using the geometric-structure approach, that the MRJ state for monodisperse disks is not polycrystalline; rather, it is isostatic and significantly more disordered as measured by bond-orientational and translational order metrics, as well as its vivid visual impression.

In order to formalize the concept of jamming, Torquato and Stillinger have provided rigorous definitions for local, collective, and strict jamming [4]. Since collectively jammed packings are stable to uniform compression, and strictly jammed packings are additionally stable against shear deformations, we will restrict ourselves to considering only these two categories of jamming for the purposes of our current work. It is not uncommon to find that some subset of particles is jammed (the backbone) while the remainder are not jammed but are locally imprisoned by their neighbors (the rattlers). If there is no jammed backbone, then the packing is unjammed. Note that, unless specified otherwise, packings are typically characterized (e.g., in order maps) while including rattlers.

The family of jammed packings is conveniently described via “order maps” that classify packings according to their packing fraction, order metric, and whether or not they are jammed. Figure 3.2 provides a schematic order map for 3D frictionless monodisperse sphere packings. We first contrast a schematic order map for the 2D case in Figure 2 that has some important distinctions. In 3D, the densest packing is the fcc lattice or its stacking variants (line $B-B'$ in Figure 3.2). By contrast, in 2D, the densest packing, the triangular lattice, is unique; therefore, it is represented by a single point $B$ in Figure 2. In a similar manner, the least dense jammed 3D sphere packing, conjectured to be the “tunneled crystal” [2], is not unique, and has stacking variants; these populate the line $A-A'$ in Figure 3.2. Similarly in 2D, the analogous
reinforced kagomé lattice packings (conjectured to be the least dense jammed disk packings [85]) populate the line $A-A'$ in Figure 2, and are generated by altering the way in which vacancies are selected from the triangular lattice. In both 2D and 3D, intermediate packings may be generated by filling in the vacancies in these packings (curves $A-B$ and $A'-B'$ in 3D, and $A-B$ and $A'-B$ in 2D).

![3D Monodisperse Spheres]

Figure 3.1: Schematic order map in the density-order ($\phi-\psi$) plane for 3D strictly jammed, frictionless, monodisperse hard-sphere packings, adapted from [1]. White and blue regions contain the attainable packings, the blue region represents the jammed subset of packings, and the dark shaded region contains no packings. The locus of points $A-A'$ corresponds to the lowest-density jammed packings, conjectured to be tunneled crystals with $\phi = \pi \sqrt{2}/9$ [2]. The locus of points $B-B'$ corresponds to the densest jammed packings (stacking variants of the fcc lattice). Packings along the curves joining these extrema can be generated by randomly inserting spheres into the vacancies of the tunneled crystal until the corresponding fcc variant is obtained. The point MRJ represents the maximally random jammed state, i.e., the most disordered state subject to the jamming constraint.

It is important to note that the order map paradigm is protocol-independent and does not conflate existence with frequency of observation. Just as the reinforced
Figure 3.2: Schematic order map in the density-order ($\phi$-$\psi$) plane for 2D strictly jammed, frictionless, monodisperse hard-disk packings in the infinite-system limit. White, blue and dark-shaded regions are the same as in Fig. 1. The locus of points $A-A'$ corresponds to the lowest-density jammed packings, conjectured to be the reinforced kagomé lattice, reinforced rectangular kagomé lattice, and other combinations, all with $\phi = \pi \sqrt{3}/8$ [4]. The point $B$ corresponds to the triangular lattice. Packings along the curves joining these extrema can be generated by randomly inserting spheres into the vacancies of the packings found along $A-A'$ lattice until the triangular lattice is obtained. The point $MRJ$ represents the maximally random jammed state. The entropically-defined location of RCP is labeled to stress the difference between using ensemble-based and geometric-structure approaches.

kagomé lattice is an interesting structure that is elusive to typical packing protocols, the MRJ state is interesting in its own right, regardless of how (often) it is observed. A characterization of the MRJ state by construction is thus of general interest.

Similarly, the MRJ state is protocol-independent and quantifies the order of an individual packing, regardless of its frequency of occurrence, i.e., using a geometric-structure approach [1]. This is to be contrasted to the current prevailing definition of RCP, in which an entropic measure is used to define randomness, i.e., that RCP pack-
nings are an ensemble of jammed configurations that are the most probable outcome upon uniformly densifying a random initial configuration to the point of jamming [19, 22, 26, 86, 87]. Such an ensemble-based definition implies that the most probable configurations represent the most disordered state. This is a reasonable postulate since entropy is the standard way of thinking about randomness in thermodynamic systems, and indeed the distinction is subtle for spheres in 3D because the packing fraction of the MRJ state seems to be coincident with such a location in phase space based on information gathered from many standard protocols [19, 45, 88, 89, 54, 3]. However, many other properties, such as the degree of order (as measured by the standard scalar order metrics) and rattler attributes have been found to vary in disordered packings at the MRJ packing fraction depending upon the protocol used to generate them. In other words, packing fraction alone is not sufficient to characterize a disordered jammed packing [3, 63, 62].

The distinction between the geometric-structure and ensemble-based approaches is even more critical in 2D, especially for monodisperse disk packings. In particular, the lack of “frustration” [90, 91] in 2D analogs of 3D computational and experimental protocols that lead to putative RCP states result in highly crystalline packings, forming rather large triangular coordination domains (grains) [85, 54, 51]. Because such highly ordered packings are the most probable outcomes for these typical protocols, “entropic measures” of disorder misleadingly identify these as the most disordered. This has even caused some to hypothesize that RCP does not exist at all for monodisperse disks [92]. An appropriate order metric, on the other hand, is capable of identifying a particular configuration (not an ensemble of configurations) that is consistent with one’s intuitive notion of maximal disorder. However, typical packing protocols do not generate such large disordered disk configurations due to their inherent implicit bias toward undiluted crystallization. It has been suggested that jammed packings with a significantly lower \( \phi \) and \( \psi \) exist [1, 93]; the geometric-
structure approach involved in the definition of the MRJ state is necessary to identify such packings, even if rare with respect to most protocols, as highly disordered.

Before the advent of these aforementioned rigorous definitions of jamming and the MRJ state, numerous attempts were made to generate RCP packings of monodisperse disks [38, 89, 94, 95, 96, 97]. Upon devising a rigorous jamming test [64], these results were revisited, and it was found that these packings were not even collectively jammed [85].

The Torquato-Jiao (TJ) linear programming algorithm [61] can be used to generate sphere packings in an arbitrary dimension that are guaranteed a priori to be strictly jammed. In addition, the sphere packings that it produces in $d \geq 3$ are exactly isostatic with high probability. It has been used recently to provide new details to the nature of the MRJ state in 3D [3]. Given its promising abilities in generating disordered, jammed sphere packings in 3D, we now turn our attention towards the 2D problem to see if it might provide new insights.

We use the TJ algorithm to produce collectively and strictly jammed packings of monodisperse disks that are exactly isostatic and lack any sixfold-coordinated disks. These packings are characterized according to their packing fraction and order using bond-orientational and translational order metrics to determine the candidates for the MRJ state. We also present pair correlation function and Voronoi statistics. The packings that we have generated demonstrate the existence of an isostatic MRJ state in 2D that is fundamentally different from the polycrystalline packings produced by typical protocols.

\footnote{An isostatic packing is one that is jammed and has the minimum number of contacts required for mechanical stability. In 2D, this is $z = 4 - 2/N_B$ for collective jamming and $z = 4 + 2/N_B$ for strict jamming, where $N_B$ is the number of backbone disks [66].}
3.3 Packing Generation

Here, we use the TJ algorithm to generate frictionless jammed packings of monodisperse disks within a fundamental cell (FC) with periodic boundary conditions. The process may be broken into two steps: generating initial configurations, and densifying said configurations to the point of jamming.

Since TJ is very sensitive to the initial configuration, we explored a variety of initial configurations. The first of these was the usual random sequential addition (RSA), carried out at a low packing fraction (typically 0.10) so as to approximate a Poisson process. We also explored several FC shapes including the usual square FC as well as slightly distorted FCs to aid in causing geometric frustration. We also tried a rhombic FC with interior angle $\pi/3$, which is compatible with the triangular lattice if the number of disks is a square integer.

In order to explore disordered packings at higher initial packing fractions, we used the serial algorithm for generating packings with controlled orientational order introduced in [98] and the version in which no orientational preference is indicated (i.e. the “Eden” model [99] adapted to hard disks).

The final initial condition ensemble that we considered was created by generating bidisperse disk packings with number ratio 0.5 and diameter ratio 1.4. Initial conditions for the bidisperse packing were generated via RSA at low packing fraction; the packing was subsequently jammed using TJ. The large disks in the resulting jammed configurations were then shrunk so that the packings became monodisperse; these packings were then fed back to TJ to yield jammed monodisperse disk packings.

The TJ algorithm [61] uses an iterative process to densify packings of hard-spheres in an arbitrary dimension using a linear optimization scheme to maximize $\phi$ to first order by shrinking the FC. The optimization variables are the $N$ displacement vectors for the spheres in the system ($Nd$ variables) as well as the symmetric strain tensor that deforms and shrinks the FC ($d(d + 1)/2$ variables). The constraints are that
no pair of spheres may overlap (to a locally linear approximation). By iteratively solving this linear program (LP), one obtains a local packing fraction maximum of the initial configuration. Furthermore, when the LP cannot find any further densification up to some numerical threshold, the packing is guaranteed to be strictly jammed to a corresponding tolerance, and is therefore a local packing fraction maximum. Furthermore, if the FC is disallowed from deforming (i.e. its shape is preserved), the packing is instead guaranteed to be collectively jammed. For mathematical details, see the Materials and Methods section.

3.4 Results

We generated at least $10^4$ packings per system size ($10 \leq N \leq 200$) per initial condition for both collective and strict jamming. The main result of this Chapter is that we demonstrate by construction the existence of jammed packings that are significantly more disordered than what was previously thought possible. Examples of such packings are shown in Fig. 3.3. The following subsections characterize various statistical and geometrical details of the packings.

3.4.1 Order Maps

We begin by presenting order maps of the packings that we have generated. Fig. 3.4a and b show scatter plots of collectively and strictly jammed packings, respectively. The packings were generated with (a) $N = 150$ and (b) $N = 110$ from RSA initial conditions in a rhombic FC with interior angle $2\pi/5$. Isostatic packings are shown as green squares. The order metric $\psi$ used here is the pair correlation function-based order metric $T^*$ [84], defined as

$$T^* = \frac{\int_{D\rho^{1/d}}^{\xi_c} |g_2(\xi) - 1| d\xi}{\xi_c - D\rho^{1/d}},$$

53
Figure 3.3: Examples of exactly isostatic, (left) collectively jammed and (right) strictly jammed monodisperse disk packings with $N = 150$ and 110 disks, respectively. Disks are colored to indicate their (backbone) coordination as follows: dark blue = 3 contacts, green = 4 contacts, orange = 5 contacts, and white = rattler (0 contacts). The fundamental cell is outlined in black.

where $D$ is the disk diameter, $\xi = r\rho^{1/d}$ is a dimensionless distance, $g_2(\xi)$ is the isotropic pair correlation function, $\rho = N/V$ is the number density, and $\xi_c$ is a dimensionless cutoff value, chosen here to be 3. For most monodisperse disk packings, this corresponds to a pair distance that is slightly less than 3 diameters. The quantity $T^*$ may be thought of as a “disorder metric” in that it quantifies the amount by which a packing differs from a Poisson point process, for which $g_2 = 1$ everywhere. Interestingly, the MRJ state for monodisperse disks is nearly as disordered (as quantified by $T^*$) as typical disordered packings of bidisperse disks with diameter ratio between unity and 1/1.4, which more readily exhibit disordered, isostatic jammed states; see Sec. 3.7 for further details. One may also consider other order metrics aside from $T^*$; we discuss this as well in Sec. 3.7.

We expand the traditional order map paradigm here by adding a third axis: the average backbone coordination number, $z$ [1]. Projections of this three-axis ($\phi$-$\psi$-$z$) order map onto the $\phi$-$z$ and $\psi$-$z$ planes are shown in Figures 3.5 and 3.6, respectively;
again, we chose to use $T^*$ as our order metric. This expanded picture shows that, as the degree of order decreases and the MRJ state is approached, the range of $z$ narrows towards isostaticity, implying that the MRJ state for monodisperse disks has an isostatic backbone, just as is the case for MRJ packings of hard-spheres in 3D [3].

Figure 3.4: Order maps of disk packings that are (a) collectively jammed and (b) strictly jammed. Initial configurations were generated from RSA with $\phi_{\text{init}} = 0.10$. Isostatic packings are shown as green squares.

Figure 3.5: $\phi$–$z$ projection of the 3-axis order map for 1000 (a) collectively jammed and (b) strictly jammed packings generated from RSA with $\phi_{\text{init}} = 0.10$. Isostatic packings are shown as green squares.

For collective jamming, the MRJ state has a packing fraction of $0.826 \pm 0.001$ and a rattler fraction of $N_R/N = 0.035 \pm 0.002$; for strict jamming these values are
Figure 3.6: $\psi-z$ projection of the 3-axis order map for 1000 (a) collectively jammed and (b) strictly jammed packings generated from RSA with $\phi_{\text{init}} = 0.10$; the order metric used here is the $g_2$-based order metric $T^*$. Isostatic packings are shown as green squares. As $T^*$ decreases, the distribution of packings in terms of coordination number narrows towards isostaticity.

$\phi = 0.826 \pm 0.002$ and $N_R/N = 0.034 \pm 0.004$. The rattler fraction is significantly higher than that for the MRJ state for 3D [3] and obeys the general trend that the rattler fraction tends to decrease as $d$ increases [55]. We reiterate that our packings are fundamentally different from previously-obtained disordered disk packings in that our packings are collectively or strictly jammed, whereas previous attempts failed to produce packings that were even collectively jammed (and, in some cases, locally jammed\(^2\)). Thus, we are demonstrating the existence of mechanically stable packings that are significantly disordered when compared to the usual polycrystalline packings generated by most protocols. Importantly, these packings demonstrate an isostatic MRJ state for 2D (shown in Fig. 3.3) that is distinct from the polycrystalline state that standard jamming protocols tend to produce. An example of the latter is shown in Fig. 3.7.

Several isostatic, collectively jammed packings were found with relatively high $\phi$ and $\psi$ (see Fig. 3.4a), which to our knowledge are also outcomes unique to the

\(^2\)A locally jammed packing is one in which no particle may be moved while holding all other particles fixed. This jamming category is insufficient to guarantee mechanical stability of any sort.
3.4.2 Success Rate

Using TJ, we have found that the probability of producing a packing with an isostatic backbone decreases in a roughly exponential manner as $N$ increases; while the choice
of initial configuration and FC shape cannot be disregarded, in general, the probability for \( N = 100 \) is approximately \( 3 \times 10^{-3} \) for collective jamming and \( 2 \times 10^{-4} \) for strict jamming; this probability decreases by a factor of 10 for every 40 additional disks. Details are available in Sec. 3.7. While this exponential decay may be the case for the present algorithm, there is no apparent reason why isostatic, MRJ packings would not exist for arbitrarily large system sizes; it is of interest to devise a protocol that increases their frequency of occurrence.

### 3.4.3 Pair Correlation Function

We have produced 16 packings with \( N = 150 \) and 177 packings with \( N = 100 \) that are exactly isostatic and representative of the MRJ state for collective jamming; for strict jamming, we found 28 packings with \( N = 110 \). To contrast these packings with the typical results (“ensemble average”) generated by the TJ protocol, we randomly selected 1000 collectively jammed packings with \( N = 150 \) and 1000 strictly jammed packings with \( N = 110 \).

Figure 3.8 shows the pair correlation function \( g_2(r) \) for (a) collectively and (b) strictly jammed packings. The peaks that correspond to the triangular lattice geometry are considerably suppressed among the MRJ packings when compared to the ensemble averages. This stresses the significant qualitative differences between the MRJ state and the most common states. The corresponding structure factors are given in Sec. 3.7.

### 3.4.4 Hyperuniformity

We have found that number density fluctuations in our MRJ packings grow more slowly than the area of a (randomly-placed) observation window, implying that the MRJ state is hyperuniform (infinite-wavelength density fluctuations vanish [17]) in two dimensions; see Sec. 3.7. This is to be contrasted with typical 2D disordered
systems in which the variance grows in proportion to the window area. 3D MRJ packings have also been shown to be hyperuniform with quasi-long-range order [56].

### 3.4.5 Voronoi Statistics

Voronoi diagrams were created using the point patterns of the disk centers. Figure 3.9 shows the probability distribution of the number of sides per Voronoi cell for isostatic packings as well as for a full ensemble average. While most cells are hexagons in both cases, it is clear that the isostatic packings exhibit a much larger variability. In addition, while most hexagons in polycrystalline packings are regular, corresponding to a locally-close-packed configuration, this is far from the case for the isostatic packings. Distributions of the local packing density of the Voronoi cells are shown in Sec. 3.7.

### 3.5 Discussion

We have used the TJ algorithm to produce collectively and strictly jammed packings of monodisperse disks that are exactly isostatic with system sizes up to $N = 150$ and
Figure 3.9: Probability distribution for the number of edges per Voronoi cell for (a) collectively and (b) strictly jammed disk packings. In addition to the MRJ packings and the ensemble average, data for the equilibrium hard-disk fluid with packing fraction $\phi = 0.67$ is included for the sake of comparison.

100, respectively. These isostatic packings are hyperuniform, have an average packing fraction of $\phi = 0.826$, and are significantly more disordered than the polycrystalline packings generated by typical protocols. In fact, these packings are candidates for the MRJ state according to bond-orientational and translational order metrics. The pair correlation function includes spikes corresponding to some of the local geometries found in the triangular lattice, but they are strongly suppressed in comparison to the ensemble average. In a similar manner, the presence of 6-sided Voronoi cells corresponding to hexagonal close-packing is strongly suppressed, and a variety of other geometries have arisen, including the increased presence of non-hexagonal Voronoi cells.

It is well-known that the MRJ state for monodisperse spheres in 3D is isostatic. In addition, polydisperse disk packings in 2D have been a popular way to elicit disordered packings, which are often isostatic [19, 22, 29, 61]. However, an isostatic packing of monodisperse disks has proved to be elusive until now. While they remain difficult to obtain, we have shown that they do in fact exist, and that some of them have
properties that are consistent with what one should expect from the MRJ state.

It is critical to point out that the 2D MRJ packings that we have shown are not entropically favored. This is problematic for a picture of RCP that relies on one’s choice of protocol as well as an ensemble-based, entropic definition. Assuming that one chooses an algorithm that is capable of producing disordered, isostatic packings, one still risks being misled by the likelihood that most packings will be highly ordered, hyperstatic polycrystalline packings, concluding that RCP is a rather unsatisfying concept in 2D. Rather, one must employ a geometric-structure approach and apply some order metric that is well defined for any single packing. This method not only allows one to unambiguously identify disorder in packings without relying on a particular packing protocol, but is also able to identify disorder in a manner that is more consistent with one’s intuition. Our findings in this work call into question the merits of studying RCP by means of an entropic, ensemble-based methodology because of the misleading picture it presents for 2D monodisperse disks.

In 3D, the least dense jammed packing known, the tunneled crystal, is hyperstatic: every sphere is sevenfold-coordinated, i.e., \( z = 7 \). By contrast, the reinforced kagomé (the least dense packing in 2D) lattice is isostatic in the infinite-system limit with average coordination number \( z = 4 \). What happens between these extremal points and the MRJ points in the jammed subsection of the order map? While some transition from \( z = 6 \) to 7 is expected in 3D, it is unclear whether there is an isostatic continuum of packings connecting the MRJ state and reinforced kagomé lattice in 2D. One possible picture is that the triangular lattice and reinforced kagomé lattice both represent states with an infinitely-large crystal; by reducing the grain size, disorder increases until the MRJ point is reached. Then, as the continuum from the MRJ point to the triangular lattice may be spanned by slowly growing crystalline grains within the packing, the continuum from MRJ to reinforced kagomé may be spanned by growing diluted crystals.
Demonstrating the MRJ state for monodisperse disks by construction is important conceptually in a similar way to the discovery of the reinforced kagomé lattice in that both are extremal packings that are very interesting though hard to observe via traditional packing protocols. This reflects a persisting fundamental lack of knowledge that we still have with regards to designing packing protocols; nevertheless, the MRJ state for disks is an interesting state because it represents an extreme state within the confines of the jamming constraint. This prompts an algorithmic question: can one devise a packing protocol that favors MRJ-like states for hard-disks? Remarkable examples of biological processes that suppress crystallization have already been found in nature such as thermal hysteresis antifreeze proteins, found in both overwintering insects and polar marine fishes [100, 101], and may be used to inspire the design of specialized packing protocols and, in turn, materials synthesis techniques.

The question, “What is randomness?” is an extremely fundamental, ubiquitous question arising not only in physics and chemistry, but mathematics and biology as well. By uncovering the MRJ state for hard disks, we identify a serious challenge to the traditional notion of disorder—that the most probable distribution is correlated with randomness. We have shown here such a distribution that is not correlated with disorder at all; the most disordered jammed configuration is not the one that shows up the most frequently in any known protocol. On the other hand, we confirm that certain order metrics are capable of accurately characterizing the order of packings on an individual basis even in this challenging case. Moreover, these revelations concerning randomness are not limited to hard-particle packings: what does the MRJ state look like for real-world systems such as water molecules or polymers? The geometric-structure approach appears to provide the proper groundwork for considering such questions.
3.6 Materials and Methods

The densification process that the TJ algorithm performs is an iterative procedure driven by solving linear programs (LPs). The protocol goal is to maximize $\phi$ (or, in the energy landscape picture [61], minimize $-\phi$), and so we may pose an objective function in terms of a strain tensor $\epsilon$ acting on the fundamental cell’s generating matrix $\Lambda$. A linearization gives the following objective function for the LP:

$$\min \text{Tr}(\epsilon) = \epsilon_{11} + \epsilon_{22} + \epsilon_{33} + \cdots + \epsilon_{dd}. \quad (3.1)$$

This implies that the components of the strain tensor $\epsilon$ are design variables; the other variables are the displacements for each sphere (in the lattice coordinate system), $\Delta x^\lambda_1, \Delta x^\lambda_2, \Delta x^\lambda_3, \ldots, \Delta x^\lambda_N$, all of which are $d$-dimensional vectors, where lambda denotes that the vectors are expressed in terms of $\Lambda$. Since no two spheres can overlap, our LP’s constraints must reflect that $r^G_{mn}$, the (global) distance between the centroids of spheres $m$ and $n$; with diameters $D_m$ and $D_n$, respectively; be $r^g_{mn} \geq 1/2(D_m + D_n)$. Expressing this in terms of the spheres’ lattice coordinates, and taking into account the spheres’ displacements and deformable fundamental cell, we have

$$\sqrt{\Lambda \cdot r^{\lambda}_{mn} \cdot \Lambda \cdot (1 + \epsilon) \cdot \Lambda \cdot r^{\lambda}_{mn}} \geq (D_i + D_j)/2,$$

where the relative displacement $r^{\lambda}_{mn} = (x^\lambda_n + \Delta x^\lambda_n) - (x^\lambda_m + \Delta x^\lambda_m)$. Linearizing this gives

$$\Lambda \cdot r^{\lambda}_{nm} \cdot \epsilon \cdot \Lambda \cdot r^{\lambda}_{nm} + \Delta x^\lambda_m \cdot G \cdot r^{\lambda}_{nm} + \Delta x^\lambda_n \cdot G \cdot r^{\lambda}_{nm} \geq \frac{1}{2} \left( (D_m + D_n)/2 - r^{\lambda}_{nm} \cdot G \cdot r^{\lambda}_{nm} \right) + \mathcal{R} \quad (3.2)$$

where $G = \Lambda^T \cdot \Lambda$ is the Gram matrix of the lattice $\Lambda$ and $\mathcal{R}$ contains all of the higher-order terms; in practice, it is acceptable to let $\mathcal{R} = 0$. An “influence sphere” of radius $\gamma$ is defined such that a constraint will be included in the LP for any pair of spheres whose centroids are separated by less than $(D_m + D_n)/2 + \gamma$. Since Eq.
(3.2) is a local linearization of a quadratic constraint, we impose an artificial limit on the extent of the design variables in each iteration to preserve the accuracy of the linearization.

If we replace the strain tensor $\epsilon$ with a scalar, we constrain the shape of the fundamental cell to remain constant. In such a way, the TJ algorithm can be quickly modified to produce collectively jammed packings instead of strictly jammed packings.

This LP is solved to determine how the packing will be rearranged and densified. After applying the sphere displacements and lattice deformation, the LP is reformulated using the new sphere positions and fundamental cell's generating matrix. The process is iterated until the solution converges and the packing does not change by more than some termination threshold. We have found that the most effective threshold is the fundamental cell volume; when the cell volume fails to decrease by an appreciable amount, the packing is jammed to a corresponding precision.

Packings are compressed using the TJ algorithm with an “influence sphere” of diameter $\gamma = D/40$ [61], where $D$ is the diameter of a sphere. For a single LP iteration, box deformations (both normal and shear movements) are limited in magnitude to less than $D/200$ and sphere translations are limited to $|\Delta r_i| \leq D/200$. The algorithm is terminated when two successive compressions fail to decrease the lattice volume by $V_k - V_{k-2} < 2.0 \times 10^{-12}$, where $V_k$ is the volume of the fundamental cell on iteration $k$. 
3.7 Appendix

3.7.1 Order Metric Correlation

The bond-orientational order metric $\langle \psi_{6,k} \rangle$ is generally positively-correlated with the translational order metric $T^*$; Figure 3.10 shows scatter plots of (a) collectively and (b) strictly jammed packings from Fig. 3 in the main text. We have found $T^*$ to be a more discerning order metric than $\langle \psi_{6,k} \rangle$ since the latter relies exclusively on local information. In addition, $T^*$ is more likely to unambiguously identify isostatic packings as MRJ-like, in agreement with the intuition that isostaticity allows for the most random configurations possible because it disallows any redundancies with regards to mechanical stability.

![Figure 3.10: Correlation of the order metrics $T^*$ and $\langle \psi_{6,k} \rangle$ for (a) collectively jammed packings with $N = 150$ and (b) strictly jammed packings with $N = 110$ generated from RSA at $\phi_{init} = 0.10$. Isostatic packings are indicated as green squares.](image)

3.7.2 Success Rate

Figure 3.11 shows the probability that an initial configuration, once compressed using TJ, will be isostatic as a function of system size for some initial condition protocols and the constraint of collective jamming; error bars represent a 95% confidence interval using the Clopper-Pearson method. Once finite-size effects become small, it is more
difficult to obtain an isostatic jammed configuration as \( N \) increases; as a general rule of thumb, adding 40 disks to the system reduces the chances of finding an isostatic packing by approximately 10. Clearly, it is of interest to design a method of generating initial conditions which favor isostatic jammed states.

Figure 3.11: The probability that a packing of monodisperse disks will be exactly isostatic (i.e. the number of backbone contacts, \( Z \), is equal to the isostatic number of backbone contacts, \( Z_{iso} \)) when jammed by TJ for collective jamming. In general, the probability decays at an exponential rate as the system size increases; a fit of the data gives \( Pr(Z = Z_{iso}; N) = 10^{\alpha N + \beta} \) where (a) \( \alpha = -0.023 \) and \( \beta = -0.25 \) (shown as a thick dashed line). Error bars correspond to a 95% confidence interval using the Clopper-Pearson method.

The probability of finding isostatic packings is lower when strict jamming is required. For most system sizes we tried, the success rate was on the order of \( 10^{-4} \); 1 million or more packings might be needed per system size in order to infer success rates accurately enough to differentiate between system sizes.
### 3.7.3 Structure Factor

Figure 3.12 shows the structure factor for (a) collectively and (b) strictly jammed packings. These were computed using the same datasets used for the pair correlation function in Figs. 7a and 7b of the main text: for collective jamming, the MRJ packings are 16 packings with $N = 150$ and 177 packings with $N = 100$ and the ensemble average is computed from 1000 randomly-selected packings with $N = 150$; for strict jamming, the MRJ packings are 28 packings with $N = 110$ and the ensemble average is computed from 1000 randomly-selected packings with $N = 110$. In a similar manner to the pair correlation function, we found that the spikes that are prominent in polycrystalline packings are considerably suppressed, indicating that the order that is characteristic of close packing in two dimensions has been significantly disrupted. The data is truncated at $k = 0.37$ because our systems are too small and few to resolve accurate statistics for small $k$.

![Figure 3.12: Structure factor for exactly isostatic, MRJ packings (black solid lines) and the ensemble average (dotted green lines) for (a) collective and (b) strict jamming. As with the pair correlation function, harmonics that correspond to features of the triangular lattice that are readily found in polycrystalline packings are considerably suppressed in the MRJ state.](image)
3.7.4 Number Density Fluctuations

Our MRJ packings are not large enough to directly observe the long-wavelength behavior of the structure factor. However, we were able to determine that the MRJ state is hyperuniform by measuring the number density fluctuations. To do this, we repeatedly threw a circular window of radius $R$ into our packings and counted the number of disk centers that fell within the window, $\langle N(R) \rangle$ as well as the the square of this quantity, $\langle N(R)^2 \rangle$. If the number variance $\sigma(R) = \langle N(R)^2 \rangle - \langle N(R) \rangle^2$ grows more slowly than the window area $\pi R^2$, then the system is hyperuniform. Figure 3.13 shows $\sigma(R)$ for (a) collectively and (b) strictly jammed packings. Because this quantity grows more slowly than $R^2$, we conclude that the number density fluctuations are suppressed in the large-system limit in a manner consistent with hyperuniformity. Data for the equilibrium fluid at $\phi = 0.60$ (for which $\sigma(R)$ scales as $R^2$) is shown for comparison.

Figure 3.13: Number variance $\sigma(R)$ for (a) collectively and (b) strictly jammed packings. Because $\sigma(R)$ seems to grow more slowly for the MRJ packings than for the equilibrium fluid, number density fluctuations are suppressed in the large-wavelength limit, suggesting that the MRJ state is hyperuniform.
3.7.5 Pentagonal and Heptagonal Voronoi Cells

As mentioned in the main text, the supremum of the local packing fraction among all heptagonal Voronoi cells seems to be \( \phi_L = \pi / (2 + \sqrt{3}) = 0.8418 \ldots \). This is accomplished by inserting two infinitesimally-small edges into the pentagonal Voronoi cell shown in Fig. 3.14.

![Diagram of Voronoi cells](image)

Figure 3.14: Two common pentagonal Voronoi cells found in isostatic disk packings with local density \( \phi_L = \pi / (2 + \sqrt{3}) = 0.8418 \ldots \). Small distortions in the disk packing can cause this Voronoi cell to gain an edge and become a hexagon, as shown in the zoom bubble, with a very small change in \( \phi_L \). Doing the same on the other similar corner creates the smallest heptagonal Voronoi cell.
Chapter 4

Critical slowing down and hyperuniformity on approach to jamming

4.1 Introduction

Dense packings of hard (nonoverlapping) spheres in $d$-dimensional Euclidean space $\mathbb{R}^d$ have been a source of fascination to scientists across the physical and mathematical sciences. Particle packings have served as simple, yet powerful models for a wide variety of condensed matter systems including liquids, glasses, colloids, particulate composites, granular materials, and biological systems, to name a few [12, 14, 15, 4, 16, 17, 19, 20, 21, 22, 102, 23, 24, 25, 26, 27, 28, 29, 30, 31, 103, 32, 33, 34, 35, 104]. Of particular interest are mechanically stable or jammed packings [4, 19, 22, 26, 1, 29, 30]. In order to make the notion of jamming rigorous, Torquato et al. introduced the following rigorous hierarchical jamming categories for frictionless spheres in $\mathbb{R}^d$ [4, 16]: a locally jammed packing is one in which no particle may be displaced while all others are fixed in place. A collectively jammed packing is one in
which no subset of particles may be displaced while fixing the shape of the system boundary. A strictly jammed packing is a packing in which no subset of particles may be displaced while allowing volume-preserving deformations of the system boundary [16]. Thus, all strictly jammed packings are collectively jammed, and all collectively jammed packings are locally jammed (each particle is locally trapped by at least \(d+1\) contacting spheres not all in the same hemisphere). Collectively jammed packings are stable against uniform compression, i.e., their bulk modulus is positive; and strictly jammed packings are additionally stable against shear, implying they also have a positive shear modulus. In the limit of exact strict jamming, the bulk and shear moduli of hard-particle packings both diverge to infinity [16].

Torquato and Stillinger have conjectured that any strictly jammed saturated infinite packing of identical spheres is hyperuniform\(^1\) [17]. A saturated packing of hard spheres is one in which there is no space available to add another sphere. Any hyperuniform point pattern is poised at a “critical point” because it is characterized by an anomalously large suppression of large-scale density fluctuations such that the direct correlation function is long-ranged [17], which is manifested by a local number variance \(\sigma^2(R)\) that grows more slowly than \(R^d\) for a spherical observation window of radius \(R\) or, equivalently, a structure factor \(S(k)\) that tends to zero as the wave number \(k = |k|\) tends to zero. More generally, for two-phase media, hyperuniformity is manifested by a local volume-fraction variance that decays more rapidly than \(R^{-d}\) or, equivalently, by a spectral density \(\tilde{\chi}(k)^2\) [91] that tends to zero in the limit \(k \to 0\) [18]. To date, there is no known counterexample to this conjecture, notwithstanding a recent study that calls into question the link between jamming and hyperuniformity

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1\(^1\)Infinite packings include any finite packing configuration in a fundamental (unit) cell that is periodically replicated throughout all of Euclidean space as well as statistically homogeneous (i.e., translationally invariant) packing configurations in \(\mathbb{R}^d\) that are also ergodic, i.e., the result of averaging over all configurations of the ensemble is equivalent to averaging over the volume for one configuration in the infinite-volume limit [91].

2\(^2\)The spectral density \(\tilde{\chi}(k)\) of a two-phase medium is the Fourier transform of the autocovariance function, which is trivially related to the probability of finding the end points of a vector \(\mathbf{r}\) inside one of the phases.
What is the rationale for such a conjecture? First, we note that a packing can be hyperuniform without being strictly jammed. For example, a honeycomb lattice packing of identical circular disks in two dimensions is only locally jammed but is hyperuniform with positive bulk modulus and zero shear modulus. This example stresses the importance of the strict jamming constraint. Indeed, appropriate deformations and compressions of a packing that is only locally or collectively jammed (and hence hypostatic with respect to strict jamming) can lead to a denser strictly jammed packing that is isostatic or hyperstatic [85], filling space more uniformly. We also know that there are infinite periodic packings, such as the triangular lattice of identical circular disks in $\mathbb{R}^2$ and face-centered-cubic lattice in $\mathbb{R}^3$ that are rigorously known to be strictly jammed [16] under periodic or hard-wall boundary conditions, and hyperuniform. In such situations, randomly removing a finite fraction of particles such that there are no “divacancies” in the two-dimensional example and no “trivacancies” in the three-dimensional example [105] while maintaining strict jamming results in nonhyperuniform packings, i.e., $S(0) \neq 0$. This example illustrates vividly that hyperuniformity is degraded by “defects”—an issue that we will discuss in more detail in the conclusions. Therefore, the conjecture includes the saturation condition. Moreover, we know that collisions in equilibrium hard-sphere configurations on the approach to jammed ordered states are not strictly hyperuniform due to vibrational fluctuations and only become exactly hyperuniform when the ideal jammed state without any defects is attained. We expect this to be the case on the approach to disordered jammed states. Thus, based on these considerations, one expects that statistically homogeneous disordered strictly jammed saturated packings of identical spheres are hyperuniform.

Importantly, the conjecture eliminates packings that may have a rigid backbone but possess “rattlers” (particles that not locally jammed but are free to move about
a confining cage) because a strictly jammed packing cannot contain rattlers [66, 1]. Typical packing protocols that have generated disordered jammed packings tend to contain a small concentration of rattlers; because of these particles, one cannot say that the whole (saturated) packing is “jammed.” Therefore, the conjecture cannot apply to these packings—a subtle point that has not been fully appreciated. Nonetheless, it is an open question what effect the rattlers have on hyperuniformity in the context of strict jamming and whether there exists a maximally random jammed (MRJ) state with no rattlers in the infinite-volume limit that is exactly hyperuniform.

Donev et al. [56] set out to see to what extent relatively large MRJ-like sphere packings (with system sizes of up to $N = 10^6$ particles) in $\mathbb{R}^3$ were hyperuniform, even though there was a small concentration of rattlers (about 2.5%), precluding them from the conjecture as noted above. Nonetheless, they found that a packing of $10^6$ particles that included the rattlers was nearly hyperuniform with $\lim_{k \to 0} S(k) = 6.1 \times 10^{-4}$ and first peak value $S_{\text{max}} = 4.1^3$. (When the rattlers were removed, the structure factor at the origin had a substantially larger value, showing that the backbone alone is far from hyperuniform.) This numerical finding supporting the link between effective hyperuniformity of an isostatic disordered packing to its mechanical rigidity spurred a number of subsequent numerical and experimental investigations that reached similar conclusions [76, 106, 79, 58, 107, 47, 108]. In all cases, effective or near hyperuniformity is conferred because the majority of the particles are contained in the strictly jammed backbone and there are few rattlers. Indeed, it has been systematically shown that as a hard-sphere system, substantially away from a jammed state, is driven toward strict jamming through densification, $S(0)$ monotonically decreases until effective hyperuniformity is achieved at the putative MRJ state. Specifically,

\[^{3}\text{While what one considers to be “effective” or “near” hyperuniformity is ultimately subjective, an empirical operational definition for such behavior is that the first peak value of the structure factor } S(k) \ [\text{or spectral density } \tilde{\chi}(k)] \text{ relative to its value at the origin is roughly } 10^4 \text{ or larger. In the case of a perfect periodic crystal, this ratio diverges to infinity.}\]
$S(0)$ was found to approach zero approximately linearly as a function of density from 93% to 99% of jamming density, where extrapolating the linear trend in $S(0)$ to jamming density yielded $S(0) = -1 \times 10^{-4}$ [47]. This study clearly establishes a correlation between distance to jamming and hyperuniformity, and additionally introduces a “nonequilibrium index” describing the interplay between hyperuniformity and a dynamic measure of distance to jamming.

In $\mathbb{R}^2$, disordered, MRJ-like packings of equal-sized disks are very hard to observe, and it has only recently been shown that highly disordered, isostatic jammed states exist at all [109]. Therefore, it is common to introduce a size dispersity in order to induce geometrical frustration and increase the degree of disorder in the resulting packings. However, examining the point configurations derived from the centers of such polydisperse packings could lead one to incorrectly conclude that the packings were not hyperuniform. Zachary et al. demonstrated [106] that the proper means of investigating hyperuniformity in this case is through a packing’s spectral density $\tilde{\chi}(k)$; that is, making an extrapolation towards the origin to estimate $\lim_{k \to 0} \tilde{\chi}(k) = 0$. They found that MRJ-like binary systems of disks with size ratio $\alpha = 1.4$ and small disk mole fraction $x = 0.75$ exhibited near hyperuniform behavior with $\lim_{k \to 0} \tilde{\chi}(k) = 1.0 \times 10^{-5}$. Thus, even though polydispersity is not part of the original conjecture [17], effective hyperuniformity can be observed in polydisperse packings as well, provided that the size distribution is suitably constrained. It is even possible that the conjecture can be extended to polydisperse strictly jammed saturated packings; however, necessary and sufficient conditions for this criterion are highly nontrivial. Nonetheless, jamming is again a crucial necessary property to attain near-hyperuniformity in $\mathbb{R}^2$ as it was in $\mathbb{R}^3$.

A fascinating open question remains as to whether putative MRJ packings can

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4The nonequilibrium index $X = S(0)/(\rho k_B T \kappa_T) - 1$ provides a means of quantitatively measuring how a system has deviated from equilibrium; a recent investigation [59] found that some equilibrium principles can be modified to understand certain behaviors found in these fundamentally nonequilibrium states.
be made to be even more hyperuniform than established to date or exactly hyperuniform with numerical protocols as the system size is made large enough. This is an extremely delicate question to answer because one must be able to ensure that true jamming is achieved to within a controlled tolerance as the system size increases without bound. The latter condition is required to ascertain the infinite-wavelength hyperuniformity property and yet any packing algorithm necessarily must treat a finite system and hence the smallest accessible positive wave number at which $S(k)$ or $\tilde{\chi}(k)$ can be measured is of the order of $2\pi/N^{1/d}$, where $N$ is the number of particles. The situation is further complicated by noise at the smallest wave numbers, numerical and protocol-dependent errors, and the reliance on extrapolations of such uncertain data to the zero-wave-number limit. To make matters even more complex, we will present evidence that current packing algorithms stop short of hyperuniformity—and jamming—because requisite collective rearrangements of the particles become practically impossible as criticality is approached, i.e., a type of “critical slowing down” [110, 111].

In this regard, it is noteworthy that general nearly hyperuniform point configurations can be made to be exactly hyperuniform by very tiny collective displacements via the collective-coordinate approach [5], which by construction enables the structure factor to be constrained to take exact targeted values at a range of wave vectors, as shown recently in Ref. [6]. Figure 4.1 vividly illustrates this point using an initial nearly hyperuniform configuration in which $S(0) = 1 \times 10^{-4}$ (which is comparable to the value obtained in MRJ-like states) and then collectively displacing the particles by tiny amounts until the structure factor $S(k)$ vanishes linearly with $k$ in the limit $k \to 0$, as in the case of disordered jammed packings. While these particles are not jammed, this example serves to emphasize that it only requires very tiny displacements to make a nearly hyperuniform system exactly hyperuniform. Thus, a critical slowing down implies that it becomes increasingly difficult numerically to drive the
value of $S(0)$ down to its lowest possible value if a true jammed critical state could be attained.

Figure 4.1: A disordered nonhyperuniform configuration with $S(0) = 1 \times 10^{-4}$ (left panel) and a disordered hyperuniform configuration in which the structure factor $S(k)$ vanishes linearly with $k$ in the limit $k \to 0$ (right panel). The configuration on the right is obtained by very small collective displacements of the particles on the left via the collective-coordinate methods described in Ref. [5]. Visually, these configurations look very similar to one another, vividly revealing the “hidden order” that can characterize disordered hyperuniform systems [6]. Indeed, each particle in the left panel on average moves a root-mean-square distance that is about 0.2% of the mean-nearest-neighbor distance as measured by the configuration proximity metric [7] to produce the configuration in the right panel.

In this Chapter, we will investigate the role of jamming and present evidence that suggests that deviations from hyperuniformity in MRJ-like packings can in part be explained by a shortcoming of the numerical protocols to generate exactly jammed configurations as a result of the type of critical slowing down mentioned above. In order to attempt to observe jammed states, we will utilize a variety of standard hard-sphere packing protocols including the Lubachevsky-Stillinger (LS) event-driven molecular dynamics algorithm [51, 54] and the Torquato-Jiao (TJ) sequential linear programming algorithm [61] to obtain putatively collectively jammed and strictly
jammed MRJ packings\textsuperscript{5}, respectively.

We will focus on frictionless binary disk packings in two dimensions because it allows us to study the behavior at smaller wave numbers than three dimensions, assuming $N$ is held constant. In addition, it is computationally easier to ensure proper jamming at a given system size, further increasing our ability to query the long-wavelength behavior of the MRJ state. Studying such systems will enable us to make contact with the recent investigation of Wu et al. [60] who examined binary packings of soft disks above the jamming transition. They found that at finite positive pressures the spectral density exhibited a local minimum at a finite wave number and proceeded to grow for smaller wave numbers, calling into question whether hyperuniformity is observed when approaching the jamming transition from above. However, they also recognized that the presence of hyperuniformity at jamming may be sensitive to the specific protocol used to construct the jammed configurations.

The rest of the Chapter is organized as follows: in Sec. 4.2, we present a variety of methods to test whether a packing, ordered or not, is truly jammed. These tests include a modification of the rigorous linear programming method of Donev et al. [64] and so-called “pressure-leak” tests. We then apply them to our packings and find that standard protocols fail to produce jammed packings at surprisingly low system sizes. In Sec. 4.4, we investigate the subtleties related to system size and numerical protocol that affect one’s ability to observe hyperuniform configurations in putatively jammed disordered packings using computer simulations. Discussion and concluding remarks are given in Sec. 4.5.

\textsuperscript{5}The MRJ state is a protocol-independent state in that it represents the most disordered state among all sphere packings subject to the jamming condition. Importantly, we are not concerned with precisely pinpointing MRJ states for binary disk packings, which would require an exhaustive study beyond the scope of the present paper. However, we know from experience that the Lubachevsky-Stillinger and Torquato-Jiao hard-sphere packing protocols are able to yield MRJ-like states.
4.2 Methods to test for jamming

A variety of methods have been used to test for jamming in hard-particle packings in the past; we will begin by reviewing some common methods, then introduce the linear programming method that we use in this work to rigorously test packings for jamming. We will show that even relatively small 2D packings with a high reduced pressure $P = pV/(Nk_B T)$ may not be jammed, even though some methods may imply otherwise.

4.2.1 Pressure leak method

An effective heuristic means of testing for collective jamming is a so-called “pressure leak” test [51, 54, 4, 64], in which the spheres are subjected to standard molecular dynamics for some relatively large time\(^6\). If the system pressure begins to drop substantially, then one may conclude that the packing was not collectively jammed; the pressure leak indicates that the particles have discovered an unjamming motion. While this test is effective for packings that are not well jammed or have a large interparticle gap, it struggles with packings that are at a high reduced pressure and packings that are nearly jammed, but for which an unjamming motion requires the cooperative motion of many spheres. In configuration space, this scenario is analogous to the $(Nd)$-dimensional configuration point being locally confined to a high-dimensional “bottleneck” from which escape may only occur in very specific directions. In such cases, the algorithm may require to process prohibitively many collisions per particle\(^7\).

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\(^6\)If the simulation box is allowed to deform so as to relieve shear stresses, then this test may be used to test for strict jamming.

\(^7\)Animations demonstrating the pressure test on an ordered and a disordered configuration are included in the Supplemental Material of [112].
4.2.2 Linear Programming-Based Approach

Donev et al. introduced a method that uses randomized sequential linear programming to rigorously test for collective or strict jamming in packings of frictionless spheres [64, 85]. Speaking physically, the algorithm applies random body forces to the spheres in the packing and seeks to maximize the work due to those forces by displacing them in the direction of their applied forces while obeying the constraint that no spheres overlap. Spheres that displace as a result of the optimization can be identified as rattlers; if every sphere is a rattler, then the packing is unjammed.

This is implemented using sequential linear programming techniques. Let \( \mathbf{R} = (\mathbf{r}_1, \ldots, \mathbf{r}_N)^T \) describe the position of the \( N \) spheres at the beginning of an iteration, and let \( \Delta \mathbf{R} = (\Delta \mathbf{r}_1, \ldots, \Delta \mathbf{r}_N)^T \) be a vector of design variables describing how the spheres displace. The vector \( \mathbf{B} \in \mathbb{R}^{Nd} \) contains the body forces which will attempt to displace the spheres; the scalar objective function \( Z = \mathbf{B}^T \Delta \mathbf{R} \) to be maximized is, physically speaking, the work performed on the packing due to \( \mathbf{B} \).

For two spheres \( i \) and \( j \) not to overlap, we require \( |(\mathbf{r}_i + \Delta \mathbf{r}_i) - (\mathbf{r}_j + \Delta \mathbf{r}_j)| \geq D_{ij} \), where \( D_{ij} = (D_i + D_j)/2 \) is the additive diameter between spheres \( i \) and \( j \) with diameters \( D_i \) and \( D_j \). Linearizing this gives the following linear program (LP):

\[
\text{maximize } Z = \mathbf{B}^T \Delta \mathbf{R}
\]

subject to \( \Delta \mathbf{r}_i - \Delta \mathbf{r}_j \leq r_{ij} - D_{ij} \forall i, j \neq i \), \( \mathbf{r}_{ij} = |\mathbf{r}_i - \mathbf{r}_j| \).

where \( d(d + 1)/2 \) strain variables to deform the fundamental cell and a constraint that the fundamental cell volume \( V \) does not increase. For a packing with periodic boundary conditions (as we consider throughout the current work), these variables
enter the constraints through pairs of spheres interacting through periodic boundary conditions; in the case where one is considering a packing with hard walls, these variables would show up in constraint terms involving the boundary.

When dealing with nonideal packings (i.e., packings thought to be very close to exact jamming but with $\phi_c - \phi > 0$ where $\phi_c$ is the jamming density), one must provide for the fact that even backbone spheres will be able to move by some small amount. Therefore, one is forced to relax the criterion that any sphere that moves must be a rattler. This was done by introducing a tolerance, i.e., any sphere that moves more than $\Delta_{tol}$ is a rattler. In addition, one must now ask not whether the packing is exactly jammed or not, but whether or not it is confined to a jamming basin. Given a jammed configuration $R_J$ at packing fraction $\phi_c$, a jamming basin $J(R_J, \phi_c)$ is defined as the set of points in configuration space for which the only accessible local packing fraction maximum under continuous displacements corresponds to $R_J$ (modulo rattlers). The density $\phi^* < \phi_c$ is defined as the highest density at which $R$ may be continuously displaced to arrive at at least one other local maximum; the quantity $\phi_c - \phi^*$ is the “depth” of the jamming basin.

It can be difficult in practice to pick a value for $\Delta_{tol}$, or to answer the “jamming basin” question definitively. This is because the available configuration space to a packing is, in general, very complicated, and the impression one obtains of it through this algorithm is dependent on the particular choice of $B$. Traditionally, $B$ is generated randomly, and the LP is solved iteratively several times in order to begin exploring in the direction of $B$; thus, one might obtain a sense of the distance over which spheres in the packing might displace despite being in a very closely packed configuration [85]. In addition, the variations in the geometry of various jamming basins (even those corresponding to an ensemble of similar packings) are considerable. Between these two factors, it is very difficult in practice to answer the binary question of whether or not a packing is truly within a jamming basin using the standard LP
jamming test.

We overcome this difficulty by choosing $B$ in a special manner designed to elucidate a particular local rearrangement that we call a “pop.” We pick a backbone sphere $i$ and a $d$ combination of backbone spheres contacting it, $C = \{j_1, \ldots, j_d\}$. We then determine the plane containing the spheres in $C$ and $b$, the unit vector orthogonal to this plane facing away from sphere $i$. The load applied to the packing is

$$B = \begin{cases} 
  b & \text{for sphere } i \\
  -b/d & \text{for spheres in } C \\
  0 & \text{otherwise.}
\end{cases}$$

This is illustrated in Fig. 4.2. Physically, we are asking for sphere $i$ to “pop” through the plane described by $C$, thus leaving the current jamming basin and entering a new one with a distinct contact network. It is important that the body forces sum to zero so that trivial uniform translations of the packing are not favored; such movements can obscure whether progress is made in realizing a “pop.”

It is important to note that while $B$ is nonzero for only a small number of spheres, the linear program is free to displace all of the spheres in the packing as it carries out its optimization, meaning that global rearrangements are being considered. In other words, while the salient characteristic of a pop might be a local rearrangement, very complicated collective movements of many particles may take place in effecting it.

We solve the linear program in Eq. (4.1) iteratively until either sphere $i$ pops through the plane or the packing stops rearranging. The latter may be inferred by monitoring the objective value associated with an optimization iteration. In the former case, we have found an unjamming motion, and the packing is unjammed; this may be confirmed by compressing the packing from this new configuration and

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8Animations demonstrating the pop test on an ordered and a disordered configuration are included in the Supplemental Material of [112].
Figure 4.2: Illustration of the body forces (arrows) chosen for a single iteration of the pop test. The green dashed line connects the two blue (dark gray when viewed in black and white) spheres that make up $C$. The forces are chosen such that the red (light gray) sphere “pops” through the two blue spheres; the sum of the forces on the packing is zero so that uniform translations of the entire packing are not favored. If the center of the red sphere crosses the green dashed line, then the packing is not confined to a jamming basin.
comparing the ensuing contact network to the original network. If a pop is not found, we continue by picking another combination of contacts or another sphere until all possibilities are exhausted, at which point we conclude that the packing must be jammed. By focusing on particle displacements instead of density increases, this test “inverts” the jamming problem and is able to efficiently determine if a given packing is within a jamming basin, even when improving the packing density is computationally difficult or the “pressure test” fails to find an unjamming motion even after considerable computational time.

4.3 Comparison of Jamming Tests

In order to establish a benchmark for the pressure test’s performance, we investigated the case of the square lattice and found that when the interparticle gap is on the order of $10^{-12}$ disk diameters, evidence of a developing pressure leak might only become visible after up to $10^6$ collisions per sphere; see Appendix 4.6 for details.

In order to illustrate the difference between the pressure test and pop test, we consider putatively collectively jammed disordered bidisperse disk packings made using a standard compression schedule with the Lubachevsky-Stillinger (LS) algorithm within a square box with periodic boundary conditions. The LS algorithm is capable of creating packings that are collectively jammed, but only if one is careful to design a compression schedule that is slow enough to allow the packing to escape unstable mechanical equilibria by discovering the requisite collective particle rearrangements. On the other hand, compression rates that are too slow allow the system to equilibrate towards less disordered states, which is at odds with our desire to probe the MRJ state [84]. To accomplish this, we start from initial conditions produced by random sequential addition at a density of $\phi_0 = 0.40$. We use an initial dimensionless expansion rate of $\gamma = dD_{\text{max}}/dt \sqrt{m/(k_B T)} = 10^{-3}$ where $D_{\text{max}}$ is the diameter of
the largest disk and $m$ is the mass of a disk and proceed until the reduced pressure exceeds $P = 10^6$; at that point, the expansion rate is reduced to $\gamma = 10^{-6}$ and the procedure continues until the reduced pressure reaches at least $P = 10^8$. We considered system sizes of $N = 10^2 - 10^4$, and we produce between $10^2$ and $10^4$ packings for each system size as is needed for reliable statistics. To encourage the protocol to generate disordered configurations, we consider binary packings with a size ratio $\alpha = 1.4$ and number ratio $x = 0.5$. An example is shown in Fig. 4.3. At $N = 10000$, the mean density of our packings is $\bar{\phi} = 0.8474$ with a standard deviation of $\sigma(\phi) = 1.8 \times 10^{-4}$ [113].

To provide a basis for comparison, we also used the Torquato-Jiao (TJ) sequential linear programming method to generate collectively jammed MRJ packings with the same size ratio, number ratio, and system size. Details of the algorithm, which solves the “adaptive shrinking-cell” optimization problem, can be found in Ref. [61]. For our current work, we use an influence sphere of radius $\gamma_{ij} = D_{max}/10$, translation limit $|\Delta x| \leq D_{max}/20$, and global strain limit $|\epsilon_{ij}| \leq D_{max}/20$. Initial conditions are created using random sequential addition at a density of $\phi = 0.40$ inside a square fundamental cell of unit volume with periodic boundary conditions. The packings are compressed while holding the box shape fixed until the volume changes by less than $10^{-12}$ over two successive iterations. To control for distance to jamming, the density of the terminal configurations is decreased so that its short-time reduced pressure (measured over 1000 collisions per disk) is $P = 10^9$.

We subject our packings to both a pressure test lasting for $10^6$ collisions per disk as well as the pop test outlined above. Importantly, nearly all of the LS and TJ packings that we produced pass the pressure test. Figure 4.4 shows the probability that a packing generated using this compression schedule passes the pop test. A curve is also included for the TJ algorithm’s pass rate for the pressure test. Our results are consistent with the intuition that jammed packings are more difficult to produce as
Figure 4.3: A collectively jammed bidisperse packing produced by the LS algorithm under the compression schedule specified in Sec. 4.2. The size ratio of the disks is $\alpha = 1/1.4$, and the number ratio is $x = 0.5$. Disks are colored according to their size: large disks are colored red, and small disks are colored blue.
\( N \) increases. However, our results illustrate vividly that standard methods to assess jamming can give misleading results even for modest system sizes, i.e., as the system size becomes on the order of one thousand disks.

![Graph](image)

Figure 4.4: The probability that a packing of a given system size \( N \), created using the LS and TJ algorithms, will be jammed, as determined by the pop and pressure tests. Error bars correspond to a 95% confidence interval as calculated using the Clopper-Pearson method [8].

Our results show that one must be careful in assuming that a given protocol is producing packings that are (nearly) truly jammed. Observing a high reduced pressure that persists for an extended period of time is not a reliable means of demonstrating that a packing is jammed. The test we demonstrate here is an efficient means of determining collective motions that unjam packings, revealing that even packings of modest size which were previously thought to be jammed may not actually be so. As the system size increases, this becomes an increasingly subtle yet crucial point to which traditional methods like the pressure test are not sensitive. Given this, one must be careful when relying on numerical results when drawing conclusions about the nature of the MRJ state. Our results also suggest that previous studies of large,
disordered, putatively jammed packings were not carried out on truly jammed configurations.

4.4 Considerations that prevent numerical packings from being exactly hyperuniform

In the following subsections, we will examine the effect of system size as well as the packing protocol used when measuring hyperuniformity in nearly jammed, finite packings of disks. To do this, we quantify density fluctuations in real and reciprocal space using the local volume fraction variance $\sigma^2_\tau(R)$ and isotropic spectral density $\tilde{\chi}(k)$, respectively.

In packings of equal-sized spheres, one may investigate the presence of hyperuniformity by considering the sphere centers and computing either their local number density variance $\sigma^2(R)$ or structure factor $S(k)$. However, these approaches fail to take into account the effect of polydispersity and have been shown [76, 106, 79] to incorrectly suggest that MRJ packings of polydisperse or anisotropic particles are not hyperuniform, whereas $\sigma^2_\tau(R)$ and $\tilde{\chi}(k)$, which properly account for these particle characteristics, show otherwise.

We begin by reviewing the procedure for computing the spectral density $\tilde{\chi}(k)$ [91] and local volume fraction variance $\sigma^2_\tau(R)$ [114, 18] for a packing of polydisperse spheres in $\mathbb{R}^d$. For complete derivations, see Ref. [106]. For a system of hard spheres with periodic boundary conditions, the spectral density may be defined via discrete Fourier transform as

$$\tilde{\chi}(k) = \left| \frac{\sum_{j=1}^{N} \exp(-i k \cdot r_j) \tilde{m}(k; D_j/2)}{V} \right|^2 \quad (k \neq 0), \quad (4.2)$$
where
\[
\tilde{m}(k; R) = \left(\frac{2\pi}{kR}\right)^{d/2} R^d J_{d/2}(kR)
\] (4.3)
is the Fourier transform of the indicator function for a \(d\)-dimensional sphere of radius \(R\) [17]; \(J_\nu(x)\) is the Bessel function of the first kind of order \(\nu\). The vectors \(k\) at which this may be evaluated are integer combinations of the reciprocal basis vectors, defined as the columns of the matrix \(\Lambda_R = [(2\pi)\Lambda^{-1}]^T\), where the columns of \(\Lambda \in \mathbb{R}^{d \times d}\) span the fundamental cell of our simulation box.

To investigate the nature of density fluctuations in real space, one may consider the variance of the local volume fraction, defined as [114]
\[
\sigma^2_\tau(R) = \frac{1}{v_1(R)} \int_{\mathbb{R}^d} \chi(r) \alpha(r; R) dr,
\] (4.4)
where \(v_1(R)\) is the volume of a \(d\)-dimensional sphere of radius \(R\), \(\chi(r)\) is the autocovariance function, and \(\alpha(r; R)\) is the scaled intersection volume, which is the intersection volume of two spheres of radius \(R\) separated by a distance \(r\) divided by \(v_1(R)\). In practice, \(\sigma^2_\tau\) may be computed by randomly placing a sufficiently large number of spherical windows of radius \(R\) within the packing.

### 4.4.1 Hyperuniformity and system size

To study the relation between jamming and hyperuniformity in disordered packings that are putatively jammed, we use the LS algorithm with the compression schedule described above to produce packings of binary disks with system sizes up to \(N = 2 \times 10^4\). In particular, we will establish that not only does jamming become less common as \(N\) increases (as shown above), but that hyperuniformity is concomitantly lost to a “saturation” in \(\tilde{\chi}(k)\) at small wave numbers. The spectral densities of our packings are plotted in Fig. 4.5; the curves drawn are ensemble averages with 1000 packings per curve; data are binned according to wave number \(k\) with a bin width of
\[ \Delta k = 0.01. \]

There are significant variations within any ensemble from packing to packing; this variation and its effect on determining hyperuniformity will be discussed for a set of 1000 packings of \( N = 500 \) binary disks produced using the LS protocol in this section. To quantify this, we fit the unbinned spectral density of each packing individually with a polynomial of order \( n \) [i.e. \( f(|\mathbf{k}|; a_0, \ldots, a_n) = \sum_{j=0}^{n} a_j |\mathbf{k}|^j \)] for all wave numbers within \( 0 < |\mathbf{k}| \langle D \rangle / 2\pi \leq k_{\text{max}} \), where \( \langle D \rangle = 1/N \sum_{i=1}^{N} D_i \) is the number-averaged diameter. For \( n = 1 \) and 2, we use \( k_{\text{max}} = 0.11 \) to investigate the behavior below the kink seen in Fig. 4.5. For \( n = 3 \), we use \( k_{\text{max}} = 0.40 \) for more complete data. We pick this particular value of \( k_{\text{max}} \) for \( n = 3 \) because the standard deviation of the value of the fit’s intercept is minimized for this range. To illustrate the goodness of fit typical of our fits, the inset of Fig. 4.5 shows binned data for the \( N = 20000 \) ensemble along with its fitted cubic polynomial.

For our ensemble of \( N = 500 \) fitted with a cubic polynomial, the intercept at the origin \( a_0 \) has a mean of \( 6.5 \times 10^{-5} \) and a standard deviation of \( 1.8 \times 10^{-3} \), meaning that these packings can be considered effectively hyperuniform in the sense that if one were to consider a single packing from this ensemble and ask whether is is hyperuniform, then the data imply that the random variations from packing to packing are large enough that one could conclude that the answer is “yes” within this noise. Interestingly, the volume fraction fluctuations in direct space strongly corroborate this conclusion, suggesting that it is of considerable utility when diagnosing hyperuniformity in small systems; we will look at this presently. The mean extrapolated values of \( \tilde{\chi}(0) \) for the other ensembles of packings shown in Fig. 4.5 are presented in Table 4.1 for various \( n \) and \( k_{\text{max}} \).

As the system size grows, a deviation from hyperuniformity becomes increasingly apparent in the spectral density curves, i.e., a “saturation” appears at low wave numbers; the curvature of the trend inflects at about \( k \langle D \rangle / (2\pi) = 0.05 \). Interestingly,
Figure 4.5: Spectral density for LS packings with various system sizes. Curves shown are binned ensemble averages with 1000 packings per curve and bin width $\Delta k = 0.01$. While ensembles of smaller system sizes seem to be hyperuniform, a saturation in $\tilde{\chi}(k)$ is observed as the system size increases and jamming is no longer ensured. Error bars are shown for a 95% confidence interval. The inset shows the cubic fit reported in Table 4.1 (orange dashed line) on top of the binned data for $N = 20000$. 
Table 4.1: Extrapolated spectral densities at $k = 0$ for ensembles of $N_p$ packings created with the LS algorithm with system size $N$, fitted using a polynomial of order $n$ for data at wave numbers $0 < k\langle D \rangle/2\pi \leq k_{\text{max}}$. Reported values are the mean $\bar{a}_0$ and standard deviation $\sigma(a_0)$ for ensembles of 1000 packings for $N \leq 2000$ and 100 packings for $N = 20000$.

<table>
<thead>
<tr>
<th>$N$</th>
<th>$N_p$</th>
<th>$n$</th>
<th>$k_{\text{max}}$</th>
<th>$\bar{a}_0$</th>
<th>$\sigma(a_0)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>1000</td>
<td>3</td>
<td>0.40</td>
<td>$-1.2 \times 10^{-4}$</td>
<td>$4.2 \times 10^{-5}$</td>
</tr>
<tr>
<td>500</td>
<td>1000</td>
<td>3</td>
<td>0.40</td>
<td>$6.5 \times 10^{-5}$</td>
<td>$1.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>1000</td>
<td>1000</td>
<td>1</td>
<td>0.11</td>
<td>$8.1 \times 10^{-5}$</td>
<td>$6.5 \times 10^{-4}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2</td>
<td>0.11</td>
<td>$1.6 \times 10^{-4}$</td>
<td>$1.7 \times 10^{-3}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>0.40</td>
<td>$1.0 \times 10^{-5}$</td>
<td>$1.1 \times 10^{-3}$</td>
</tr>
<tr>
<td>2000</td>
<td>1000</td>
<td>1</td>
<td>0.11</td>
<td>$8.3 \times 10^{-6}$</td>
<td>$4.1 \times 10^{-4}$</td>
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<td></td>
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<td>2</td>
<td>0.11</td>
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<td></td>
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<td>3</td>
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<td>20000</td>
<td>100</td>
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<td></td>
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<td>3</td>
<td>0.40</td>
<td>$6.5 \times 10^{-5}$</td>
<td>$2.2 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

This wave number roughly corresponds to the largest wavelength that is typically observable in a jammed system of 500 disks. At the same time, this system size lies approximately at the point where a given packing will be jammed with a likelihood of about 50%.

This prompts us to suggest that the physical origin of the “saturating” behavior may be fundamentally linked to the ability of the packing protocol to resolve collective rearrangements on a corresponding length scale that may be necessary to reach a truly-jammed state. A point in configuration space may become locally trapped within a “bottleneck” from which escape may only happen in very few directions. The result in practice is a “critical slowing down”[110] in which exact jamming takes increasingly long to resolve, as we elaborate in Sec. 4.5.

We have also found that this “saturating” behavior can be misrepresented when care is not taken in extrapolating the effective value of $\tilde{\chi}(0)$ from the $\tilde{\chi}(k)$ for which data are available. Specifically, the effect of binning data to obtain an ensemble average can artificially increase or decrease the perceived value of $\tilde{\chi}(0)$, where we
have observed mostly increases in $\tilde{\chi}(0)$ when bin size is substantially larger than the smallest wave number. Moreover, the spectral density of individual packings tend to vary by a significant amount from that of the ensemble average. This begs the question: how close to zero must the extrapolated spectral density at $k = 0$ be in order to be considered “effectively hyperuniform”? We address these questions in detail in Appendix 4.7.

Our estimates for a single packing in the $N = 2000$ ensemble described in Appendix 4.7 indicate that $a_0 = \tilde{\chi}(0)$ will be randomly distributed about zero with a standard deviation of about $4 \times 10^{-4}$. Averaging over an entire 100 packing set yields a better estimate of the mean value $\overline{a_0}$ of the average of the set, and tighter zero-hypothesis confidence intervals. Our methods show that with about 50% probability, a mean value of $\overline{a_0}$ such that $-4 \times 10^{-5} \leq \overline{a_0} \leq 4 \times 10^{-5}$ indicates effective hyperuniformity, given the noise inherent in the calculation. Referencing Table 4.1 values for the $N = 2000$ packings and applying Student’s $t$ distribution to the standard deviation reported, we see that with 68% probability the mean is within $6.1 \times 10^{-5}$ of $4.6 \times 10^{-5}$. This is on the higher side of the range $[-4 \times 10^{-5}, 4 \times 10^{-5}]$, indicating that the packings together could be hyperuniform within the error, but that individually many of them are not. This result corresponds well with the prediction that jamming and hyperuniformity are linked, since the majority of the $N = 2000$ packings are not jammed according to the pop test, yet they are close to jamming since they all pass the pressure test.

We also compute the local packing fraction variance $\sigma_r^2$ for these ensembles with various system sizes, as shown in Fig. 4.6. For hyperuniform packings, $\sigma_r^2(R)$ will scale towards zero more quickly than $R^{-2}$ as $R$ tends towards infinity. Equivalently, $R^2 \sigma_r^2(R)$ will be a decreasing quantity with growing $R$. This is clearly the case with our data, indicating that these ensembles are hyperuniform by this metric. We see a sudden decrease for sufficiently large $R$ for each $N$, but our studies on larger
samples imply that this is a finite-size effect similar to that observed in Ref. [108]. However, note that the hyperuniformity trends are apparent even for the smaller system sizes. These results indicate that there is a maximum length scale $R_{\text{max}}$ (smaller than the half width of the simulation box) that can be considered when diagnosing hyperuniformity. In addition, while the spectral density calculation may require hundreds or even thousands of packings to converge to well-resolved curves, the direct-space curves converge much more quickly. It has been noted before that if one must ascertain hyperuniformity from either a small system or an ensemble with a limited population, the direct-space computation is particularly effective at diagnosing hyperuniformity [108].

![Figure 4.6](image)

Figure 4.6: Window packing fraction variance scaled by the window volume $(R/\langle D \rangle)^2$ for LS packings for various $N$. Curves shown are binned ensemble averages. If the ordinate scales towards zero, then the systems are hyperuniform. The sudden decrease at the highest values of $R$ for each curve is due to finite-size effects, but hyperuniformity is apparent even for the smaller system sizes. The uncertainty in the curves is very small, on the order of the linewidth.
4.4.2 Hyperuniformity and protocol dependence

We now consider the effect that one’s choice of packing protocol has on the degree of hyperuniformity. In particular, we will present evidence that different protocols, which come near to exact jamming to different degrees, create packings with correspondingly different degrees of hyperuniformity. Importantly, while all of the protocols considered have the capacity to generate jammed packings given arbitrarily high numerical precision and computing time, practical considerations like computational cost and the rate of convergence force one to terminate any algorithm before exact jamming is attained. It is this distance to jamming that we would like to focus on. To do this, we begin with our LS-generated binary disk packings as well as packings made using a standard soft-sphere protocol following the procedure in [60]. For the latter algorithm, particles interact through the standard harmonic pair potential [19]

\[
\psi_{ij}(r_{ij}) = \begin{cases} 
\frac{1}{2} \left(1 - \frac{r_{ij}}{D_{ij}}\right)^2 & \text{for } r_{ij} < D_{ij} \\
0 & \text{for } r_{ij} \geq D_{ij}
\end{cases}
\] (4.5)

where \(D_{ij} = (D_i + D_j)/2\) and \(D_i\) is the diameter of particle \(i\). An enthalpy-like function \(H = \Gamma_N \ln(V) + \sum_{i,j} \psi_{ij}\) is minimized for \(\Gamma_N/N = 1.373 \times 10^{-4}\) using a conjugate gradient method, terminating when the gradient of the objective function falls below \(10^{-13}\) or its value remains constant (as expressed in double precision) over ten optimization steps. Initial configurations are Poisson point processes at reduced density \(\sum_i v_i/V = 0.84\), where \(v_i\) is the volume of particle \(i\).

We input the resulting packings from these two protocols as initial conditions for the TJ algorithm, and proceed to generate putatively strictly jammed packings\(^9\). Since TJ seeks out local packing fraction maxima, the threshold state being ap-

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\(^9\)The diameters of the disks in the soft-sphere packings are all reduced by a constant factor such that no pairs of disks are overlapping and that the closest pair of disks are in contact.

\(^{10}\)An animation demonstrating this combination protocol using the soft-sphere algorithm followed by the TJ algorithm is provided in the Supplemental Material of [112].
approached in the soft-sphere protocol is the same state that is being approached using TJ. While it is beyond the scope of this work to investigate the similarities and differences in the approach to the jamming transition from these two protocols, the limiting configuration is essentially unchanged when the packings are given to TJ.

Figure 4.7 shows the ensemble-averaged spectral density of ensembles of 100 packings of system size $N = 10^4$ for both protocols before and after being input to TJ; the corresponding direct-space measurement of the window volume fraction variance $\sigma_\tau^2(R)$ is shown in Fig. 4.8. The LS-generated configurations do not change by a significant amount upon being fed to the TJ algorithm, suggesting that the difference between collective and strict jamming is minor at sufficiently large system sizes [85, 66].

Of particular importance is that the sharp increase in $\tilde{\chi}$ near the origin that is observed in soft-sphere packings at a positive pressure [60] vanishes upon subsequent packing using the TJ algorithm. This is seen in the direct-space calculations as well, since the nonhyperuniform $R^{-2}$ scaling in $\sigma_\tau^2(R)$ seen with the soft-sphere protocol at length scales beyond $R = 7\langle D \rangle$ becomes hyperuniform upon subsequent packing with TJ. This ability to obtain length scales over which hyperuniformity is observed is one of the main strengths of the direct-space calculation [108]. TJ is allowing us to get several orders of magnitude closer to exact jamming and more accurately discern its spectral density: for TJ, $\phi_J - \phi \approx 10^{-12}$, whereas in the soft-sphere case, $\phi - \phi_J \approx 10^{-3}$. This reinforces the idea that bringing a packing closer to exact jamming will improve its hyperuniformity. Additionally, this result again emphasizes the subtleties in characterizing the long-wavelength nature of the MRJ state and underscores the need to be mindful of the nature of the numerical insensitivities in packing protocols—particularly as they approach highly disordered states. We point out in passing that we don’t expect the spectral density of the soft-sphere packings to match that of the packings generated with the LS algorithm, even after further
Figure 4.7: The spectral density of disordered binary disk packings created by a variety of protocols. Curves shown are binned ensemble averages with bin width $\Delta k = 0.01$. Feeding the packings produced by the soft-sphere protocol (identified as “SS” in the legend) into the TJ algorithm increases the hyperuniformity of the packings significantly as they are brought closer to exact jamming. The putatively collectively jammed packings generated by LS are almost unchanged upon being given to TJ for strict jamming, implying that the difference between collective and strict jamming is small at large system sizes. Error bars are shown for a 95% confidence interval.
Figure 4.8: Window number variance scaled by the window volume \((R/\langle D \rangle)^2\) for binary disk packings with system size \(N = 10^4\) created by a variety of protocols. Feeding the packings produced by the soft-sphere protocol (identified as “SS” in the legend) into the TJ algorithm increases the range over which hyperuniform scaling behavior is observed as the packings are brought closer to exact jamming. The LS packings exhibit hyperuniform scaling over a significant range of length scales (i.e., the ordinate scales towards zero as \(R\) increases). The uncertainty in the curves is on the order of the linewidth.
packing with TJ, because the two packing protocols utilize qualitatively different dynamics, meaning that they are expected to sample from the ensemble of jammed packings with different weightings. Thus, the packings that one might expect to see with each protocol are expected to possess different structures as well.

4.5 Conclusions and Discussion

We have introduced a method that uses sequential linear programming to test for jamming in packings of frictionless hard spheres. We applied this algorithm to disordered packings of bidisperse disks and found that standard protocols struggle to create packings that are truly confined to a jamming basin when the system size grows to be on the order of 1000 disks or more. Importantly, heuristic tests like the pressure test fail to find these unjamming motions. We then examined the spectral density of the packings we generated for a variety of system sizes and found an inflection at a wave number corresponding to the largest accessible length scale in a typical packing with system size \( N = 500 \)—the same system size for which the probability of producing a truly jammed configuration is approximately 50%. Given this, we conclude that our inability to observe exactly hyperuniform configurations at larger system sizes is directly linked to the difficulty of producing exactly jammed configurations.

We also found that, by bringing soft-sphere packings closer to their jamming transition point by using the TJ algorithm, the degree of hyperuniformity increased by several orders of magnitude as measured by the volume fraction fluctuations in direct space. This ability to obtain length scales over which hyperuniformity is observed is one of the main strengths of the direct-space calculation [108]. This finding also suggests that the break from hyperuniformity at large length scales, previously thought to be inherent to packings above the jamming transition, is in reality due to an excessive distance from the jamming transition, and that subsequent resolution makes this
feature disappear. Because of our careful consideration of packings’ distance to jamming and the corresponding evolution of their structure, we conclude that one cannot rely solely on current technology to say that there is no connection between jamming in disordered packings and hyperuniformity since jamming cannot be ensured.

In both of the cases studied above, we point out an evident “critical slowing down” in that the collective particle movements required to reach an exactly jammed state take longer to resolve as the system size grows. A point in configurational space may become locally trapped within a “bottleneck” from which escape may only happen in very few directions, corresponding to the collective rearrangements that the packing must undergo. For an event-driven MD protocol such as LS, it may take many collisions per particle before this escape is discovered. As the system size increases, the dimensionality of configuration space increases as well and this “escape” becomes an increasingly rare event. The result in practice is a “critical slowing down” in which exact jamming takes increasingly long to resolve. This dynamic critical behavior is well known in other physical systems, the most well known of which is perhaps the kinetic Ising model [110]. The fact that it is also observable in packing contributes additional evidence to support the idea that the MRJ state lies at a special type of critical point, namely, one in which the direct correlation function $c(r)$, rather than the total correlation function $h(r)$, is long-ranged due to the fact that the appropriate spectral function is zero at $k = 0$ [17]. This is to be contrasted with a thermal critical point in which density fluctuations diverge because $h(r)$ is long-ranged.

It is important to notice that the protocols that we are aware of tend to produce configurations possessing a positive rattler fraction. As these particles do not contribute to the rigidity of the backbone, they might be regarded as “defects” within the disordered configuration. Their location is also not uniquely specified, in contrast to the positions of the backbone particles. Therefore, we expect that any packing containing rattlers cannot necessarily be exactly hyperuniform due to the freedom
the rattlers possess. However, one must also be aware that the backbone configurations that give rise to rattler cages are also interesting in that the cages surrounding the rattlers tend to have significantly different local structures from that of the rest of the packing [3]. Therefore, it is not enough to expect that “optimizing” the rattlers’ positions will necessarily yield a hyperuniform configuration. In general, one should not expect that a packing possessing a jammed backbone will be hyperuniform unless it is saturated and does not possess any rattlers. It has been observed previously [3] that the TJ algorithm produces packings of equal-sized spheres in three dimensions that are simultaneously more disordered and have significantly fewer rattlers than other known protocols (e.g., [54]), leaving open the possibility that packings that are more disordered may have even fewer rattlers still. This raises the possibility that the ideal MRJ (most disordered, strictly jammed) packings of identical spheres have no rattlers. Indeed, if this is true, then one might reasonably expect that this would also hold for two-dimensional systems of identical particles [109] and certain polydisperse packings in two and three dimensions (given qualifications on the distribution of particle sizes). This also would imply that the most disordered jammed states are those that contain no defects. In other words, defects increase the degree of order. Devising algorithms that produce large rattler-free disordered jammed packings is an outstanding, challenging task. According to the Torquato-Stillinger conjecture, any MRJ-like strict jammed packing without any rattlers would be hyperuniform in the infinite-volume limit [17].

It has been found that the average rattler fraction $N_R/N$ observed in a disordered packing is dependent upon the protocol being used; in three dimensions, putative MRJ packings produced with the LS algorithm tend to have a rattler fraction of approximately $N_R/N \approx 0.025$ [56]. On the other hand, the TJ algorithm produces packings with $N_R/N \approx 0.015$ [3]. For the binary systems considered in this work with $(\alpha, x) = (1.4, 0.5)$, the LS algorithm produces an average rattler fraction of $N_R/N =$
0.063 ± 0.001, whereas the TJ algorithm produces a mean of $N_R/N = 0.048 ± 0.001$, mirroring the story in three dimensions. Given that the packings produced by TJ are significantly more disordered as measured by standard order metrics [3], we ask whether the true MRJ state has no rattlers. The existence of such a jammed state remains an open question, and addressing it would presumably require a novel packing protocol.

For $d = 3$, similar difficulties to those observed in this work exist in producing truly jammed packings. For example, previous investigations have suggested that it is difficult to produce disordered packings of $N = 10^4$ spheres using the LS algorithm that can pass even a pressure-leak test [66]. Simulations using TJ take increasingly long amounts of time in producing jammed packings at comparable system sizes. Thus, we point out that there is an issue that is practical in nature associated with producing jammed packings of monodisperse spheres at large system sizes. An apparent “saturation” was observed in the structure factor of disordered soft-sphere packings in which the trend $S(k) \propto k$ breaks down and becomes constant and positive for smaller wave numbers, implying that jamming and hyperuniformity may not be connected [59]. The wave number associated with this “turnover” corresponds to wavelengths on the order of approximately 20 spheres, which mirrors the “critical” system size above which it seems to be difficult to generate truly jammed packings. This parallels the observations we have made in our current study. We suggest, therefore, that this observed departure from exact hyperuniformity may be due to an inability to resolve the particle displacements necessary to approach the jamming threshold—particularly given the system sizes that were considered ($N = 5 \times 10^5$).

Our results demonstrate the particular difficulty of producing systems of either hard or soft particles that fall precisely at the jamming transition. Moreover, ensuring that a packing is truly in a jamming basin is a highly nontrivial task. Is there hope for producing improved algorithms to yield higher quality MRJ-like states? Of
critical importance is that the algorithm be efficient at determining and resolving the collective movements that allow the packing to escape configurations that are not true local density maxima. Evidently, some of these motions may involve all of the particles in the system, and may achieve very small immediate changes in density. Thus, an intelligent way of identifying and carrying out these displacements is warranted.

We have suggested that reaching an exactly jammed state requires particles to “slide” by each other in very specific ways when the packing is barely inside a jamming basin. Nonetheless, we point out that near hyperuniformity may be readily observed and quantified in a number of meaningful ways. One is to measure the ratio between the value of the structure factor at the origin (e.g., as obtained by scattering experiments) and at the first peak\textsuperscript{11}. A second technique is to measure the volume fraction fluctuations in real space as a function of observation window size and look for the appropriate scaling relation as above [108, 115]. This latter technique has indicated the existence of real-world systems that are hyperuniform over length scales spanning as much as four orders of magnitude [115]. Thus, it is of interest to investigate the physical consequences of near hyperuniformity as observed in such systems.

\textsuperscript{11}See footnote 3
4.6 Appendix: Jamming tests on the square lattice

In order to assess the reliability of the pop test for discovering unjamming motions, we consider the case of a square lattice in two dimensions, using both (a) a monodisperse disk packing and (b) a binary disk packing with size ratio $(\alpha, x) = (1.4, 0.5)$; examples of the starting configurations are shown in Fig. 4.9.

![Figure 4.9: Square lattice packings of (a) monodisperse and (b) binary disks.](image)

We considered system sizes of $N = 10^2$ and $10^4$ and size ratios $\alpha = 1$ and 1.4 (the former referring to the monodisperse limit). We perform pressure pressure tests for $\phi_c^* - \phi = 10^{-8}$ and $10^{-11}$, where $\phi_c^*(\alpha) = \left[\pi(1 + \alpha^2)\right]/[2(1 + \alpha)^2]$ is the close-packing density of the lattice\textsuperscript{12}. The reduced pressure is plotted as a function of the number of events per sphere in Fig. 4.10. It is important to note that the results of the pressure test will vary with different starting velocities, so we include several representative runs for each case. As expected, the pressure drops quickly after some amount of time, indicating that these packings are not jammed and that an unjamming motion

\textsuperscript{12}This is valid for $\alpha \in [\sqrt{2} - 1, \sqrt{2} + 1]$. 

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has been discovered. Note that more events per sphere are required to observe a pressure “leak” when $\phi_c^\ast - \phi$ is smaller. In addition, the pressure leak takes slightly longer to show up in the bidisperse case, presumably due to the lessened degree of symmetry in the packing and correspondingly smaller degeneracy of its unjamming motions.

The pop test was also able to demonstrate the existence of “popping” motions for all of the aforementioned square lattices. We also note that the pop test did not perform differently for different jamming gaps, suggesting that it is robust against differences in interparticle distance.

4.7 Appendix: Considerations regarding ensemble-averaging spectral density data

Great care must be taken when numerically extrapolating $\tilde{\chi}(k)$ values to $\tilde{\chi}(0)$. Additionally, claims about hyperuniformity for small sets of packings with extrapolated $\tilde{\chi}(0)$ values near zero must be accompanied by reasonable estimates of confidence intervals in order to be valid. In this appendix, we show one way in which this might be accomplished.

We mentioned above that the effect of binning data to obtain an ensemble average can give misleading results about the perceived value of $\tilde{\chi}(k)$, particularly when $\tilde{\chi}$ is small. This can be the case when (i) binning strategies are used to average data from a group of packings produced using the same protocol (but different initial conditions), then (ii) a curve is fit to the binned $\tilde{\chi}(k)$ values, and finally (iii) the curve is extrapolated to $k = 0$ to derive $\tilde{\chi}(0)$. The choice of bin width has a significant effect on the appearance of the ensemble averaged curve.

For example, considering a set of 100 packings of $N = 2000$ binary disks produced using the LS protocol, binning the $\tilde{\chi}(k)$ values with bin width equal to $\Delta k = 0.01$ and
Figure 4.10: Pressure tests for a square lattice of (a) monodisperse and (b) binary spheres with \((\alpha, x) = (1.4, 0.5)\) with \(N = 10^2\) disks at putative jamming gaps of \(\phi_J - \phi = 10^{-8}\) (lower curves in each subfigure) and \(10^{-11}\) (upper curves). As the jamming gap approaches zero, longer simulations are required to observe unjamming. For systems with \(P \approx 10^8\), a simulation of one million collisions per particle is usually long enough to observe an unjamming motion.
then fitting a third order polynomial for $0 \leq k\langle D \rangle/2\pi \leq 0.4$ yields an extrapolated $\tilde{\chi}(0)$ value of $-4.6 \times 10^{-5}$. However, fitting individual 3rd order polynomials to each packing’s $\tilde{\chi}(k)$ and then averaging the 100 values of $a_0$ yields $\overline{a_0} = 4.6 \times 10^{-5}$.

This difference between a single extrapolated $\tilde{\chi}(0)$ value and the average $\overline{a_0}$ of individual fits is small but significant. It begs the question, how close to zero must $a_0$ or $\overline{a_0}$ be to be “effectively hyperuniform”? To this end, we begin by considering the probability distribution associated with $\tilde{\chi}(k)$ as a function of the wave number $k$. We take an ensemble of 100 packings of $N = 2000$ bidisperse disks and consider the set of spectral densities observed at each wave number within the range $0 \leq k/2\pi \leq 0.5$. From the data at these wave numbers (normalized by their respective means), probability density functions are obtained; these are shown in Fig. 4.11 as sets of filled circles. The solid black line is found by aggregating the data together to obtain an average over all wave numbers. The data are well fitted by an exponential distribution, indicating that for wave numbers $k$ that are near one another for all of the packings studied, the expected value of any single $\tilde{\chi}(k)$ is equal to the standard deviation of the distribution.

To determine an estimate for the standard deviation of any extrapolated value $a_0 = \tilde{\chi}(0)$ for a single packing, we considered the following toy problem: suppose that for a hypothetical $\tilde{\chi}(k)$ curve, the spectral density corresponding to each wave number $0 \leq k/2\pi \leq 0.4$ for a hypothetical packing of side length $45\langle D \rangle$ is chosen from an exponential distribution with a mean given by $\overline{\tilde{\chi}}(k) = \beta k$, where $\beta$ is chosen empirically to reflect the data from MRJ bidisperse packings. The standard deviation of the constant term in a linear fit to this hypothetical $\tilde{\chi}(k)$ curve over the range $0 \leq k/2\pi \leq 0.4$ is $\sigma(a_0) = 4.2 \times 10^{-4}$. This provides an estimate for the standard deviation of an $a_0$ value extrapolated from the spectral density of any single packing.

\footnote{Generally speaking, no two packings within an ensemble of MRJ-like packings will have the same fundamental cell. Therefore, the wave numbers at which the spectral density can be measured will vary accordingly by a small amount.}
Figure 4.11: Probability density functions of spectral density values observed in nearly jammed packings of $N = 2000$ bidisperse disks. One dataset is shown (as a series of filled circles) for each wave number, and the thick black line shows the average over all wave numbers. Normalizing the distributions with respect to their means makes them collapse to a single master curve, showing that $\tilde{\chi}(k)$ across an ensemble of packings is approximately exponentially distributed at any given wave number.

in the aforementioned set of 100, assuming that the packings in the set exhibit roughly linear behavior in $\tilde{\chi}(k)$ for small $k$ with a near-zero extrapolated $\tilde{\chi}(0)$.

However, the $\overline{a_0}$ value of an ensemble should still converge toward zero as the population of the ensemble grows towards infinity, provided that all of the packings were in fact effectively hyperuniform. To establish to what extent this convergence would occur, we consider a second toy problem in which we generate 20 sets of 100 such hypothetical $\tilde{\chi}(k)$ curves with each $\tilde{\chi}(k)$ for each curve derived from an exponential distribution as just described. We calculate linear fits and determine $a_0$ for each curve. We average these values to obtain $\overline{a_0}$ for each set, then compute the mean and standard deviation of these 20 measurements. We find values of $1 \times 10^{-6}$ and $5 \times 10^{-5}$,
respectively. This suggests that a good estimate for a $\overline{a_0}$ of a set of 100 packings exhibiting linear and hyperuniform spectral density would be within $\pm 5 \times 10^{-5}$ of zero about 68% of the time, within $\pm 1.0 \times 10^{-4}$ about 95% of the time, and within $\pm 1.5 \times 10^{-4}$ about 99% of the time.

For our 100 packings with $N = 2000$, $\overline{a_0} = 4.6 \times 10^{-5} \pm 6.1 \times 10^{-4}$, falling at about the 64th percentile of the distribution we obtained in the above toy problem (in which the ensemble was exactly hyperuniform by construction), indicating that with about 64% probability, not all the packings in the set are hyperuniform. This conclusion suggests that the numerical precision to which we can determine $\tilde{\chi}(0)$ for this set of 100 packings is not sufficient to rule out effective hyperuniformity with reasonable certainty. A larger sample set might yield more certainty. However, this conclusion supports the notion that jamming is associated with hyperuniformity: only one of the packings in the set passed the pop test and is therefore jammed.

In summary, using the distribution of $\tilde{\chi}(k)$ values near a given wave number $k$, we are able to provide an estimate, for a system of $N = 2000$ binary disks produced as described, of a range of extrapolated $\tilde{\chi}(0) = a_0$ values that might be considered hyperuniform. That range is $0.0 \pm 4.2 \times 10^{-4}$, within one standard deviation. Using this previously described method, estimates for confidence intervals over which a set of such packings might be considered effectively hyperuniform can be derived. For 100 packings of $N = 2000$ such disks, an $\overline{a_0}$ value of $0.0 \pm 1.5 \times 10^{-4}$ could be considered hyperuniform. Estimates for larger and smaller sets could be performed as well.

These findings, based on the binning study and the study of the distribution of $\tilde{\chi}(k)$ values, lead us to suggest that great care must be taken when numerically extrapolating $\tilde{\chi}(k)$ values to $\tilde{\chi}(0)$. Additionally, claims about hyperuniformity for small sets of packings with extrapolated $\tilde{\chi}(0)$ values near zero must be accompanied by reasonable estimates of confidence intervals, perhaps derived from the method described in this appendix, in order to be valid.
Chapter 5

Static Structural Signatures of Nearly Jammed Disordered and Ordered Hard-Sphere Packings: I. Direct Correlation Function

5.1 Introduction

Packings of hard particles in \( d \)-dimensional Euclidean space \( \mathbb{R}^d \) have been used ubiquitously as a powerful model to describe many-body systems such as liquids, glasses, colloids, granular materials, particulate composites, and biological systems, among others [12, 14, 15, 4, 16, 17, 18, 19, 20, 21, 22, 102, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 104]. In three dimensions, the venerable hard-sphere model is particularly useful, owing to its mathematical simplicity and the rich diversity of equilibrium and non-equilibrium behavior that it exhibits.

It has been shown that bringing hard-particle packings towards jamming (roughly speaking, mechanical stability) is accompanied by an anomalous suppression of long-
range density fluctuations [57, 106, 79, 47, 116, 112]—a phenomenon known as “hyperuniformity” [17, 18]. A many-particle system is hyperuniform if the structure factor $S(k)$ (trivially related to the Fourier transform of pair statistics in direct space) tends to zero in the limit that the wave number $|k|$ tends to zero. Hyperuniformity may be conceptualized as an “inverted critical point” in which the direct correlation function $c(r)$, which is defined through the Ornstein-Zernike integral equation for a system with number density $\rho$:

\[ h(r_{12}) = c(r_{12}) + \rho \int_{\mathbb{R}^d} h(r_{23})c(r_{13})dr_3, \]  

becomes long-ranged, i.e., its volume integral diverges [17]. This is to be contrasted with the usual thermal critical point (e.g., liquid-vapor or Curie critical points) in which the total correlation function $h(r)$ [rather than $c(r)$] becomes long-ranged. Accordingly, a static length scale, obtained from the Fourier transform of $c(r)$, $\xi = [\tilde{c}(k = 0)]^{1/d}$, grows as a system approaches a hyperuniform state and ultimately diverges at this critical state. In hard-particle packings, this occurs as jamming is approached [47], and a similar analysis can be used to obtain meaningful information about the nature of glassy states of particles with soft interaction potentials [117]. Because of this, it has been an intriguing prospect to investigate disordered hyperuniform systems by adapting standard tools used to investigate critical phenomena.

At the same time, the direct correlation function has proven to be a fruitful starting point for efforts to characterize the structure of disordered systems such as simple liquids since the pair statistics may be obtained through Eq. (5.1) [118]. Physically, the equation suggests that pair statistics between any two particles may be decomposed into a “direct” contribution encoded in $c(r)$ as well as an “indirect” contribution mediated through “chains” of particles, expressed mathematically through the con-
volution between $h(r)$ and $c(r)$. For systems with suitably well-behaved interactions, one may equivalently think of $c(r)$ as describing the linear response of a system to a perturbation in an externally applied potential field [118].

It has been observed that maximally random jammed (MRJ) hard-sphere packings, which constitute the most disordered configurations as measured by some scalar order metric subject to the constraint of jamming and isostaticity [45, 1], are hyperuniform and exhibit a nonanalytic linear behavior in the structure factor for low $k$, namely, $S \propto k$ [56]. Hopkins et al. [47] studied the behavior of very large ($N = 10^6$) sphere packings produced by the Lubachevsky-Stillinger event-driven molecular dynamics algorithm [55] under rapid compression so as to study the approach to the MRJ-like states at densities well above the freezing density and close to jamming. They found evidence that the nonanalytic linear behavior in $S(k)$ was evident considerably in advance of jamming, and that upon further compression, the extrapolated value at the origin tended towards zero, implying that corresponding long-ranged behavior in $c(r)$ might be observable for this protocol. Their computations for the Fourier-transformed direct correlation function showed $\tilde{c}$ extending towards negative infinity near the origin as the packings were compressed, supporting this prediction.

In the current work, we further develop the view that one may find static, structural precursors to jamming in hard-particle systems. Because $c(r)$ is known to generally possess a qualitatively simpler functional form while still encoding the complete pair statistics of the system, we will focus primarily on the signatures therein, paying particular attention to features that point towards the development of an incipient contact network and hyperuniform density fluctuations [i.e. long-rangedness in $c(r)$]. Moreover, $g_2(r)$ is known to possess various singularities at jamming (e.g., a Dirac delta function at contact, discontinuity at a distance of two diameters), and we determine to what extent these features are inherited by $c(r)$.

However, one should also bear in mind that different packing protocols will tend
to produce different ensembles of disordered jammed states [62]. In this paper, we will also bring attention to qualitative differences in protocols’ approach to their jammed states. Additionally, it is nontrivial to ensure that standard protocols approach properly-jammed states and avoid becoming stuck in unstable mechanical equilibria. This has been found to be related to a “critical slowing down” that becomes of practical concern for large systems [112]. Therefore, we carry out our current investigation as if our systems are indeed jammed and hyperuniform with the important caveat that this is more difficult to do with high precision in practice than previously thought. If one had a protocol to produce better-jammed packings, we expect that the packings would possess stronger structural signatures consistent with hyperuniformity.

To this end, we analyze computer-generated packings of monodisperse hard-spheres of diameter $D$ created by the LS algorithm as well as the Torquato-Jiao (TJ) sequential linear programming algorithm [61]. We consider these two algorithms since (i) they are both known to generate highly disordered packings under suitable conditions, but also because (ii) the jammed states they produce possess considerable differences in their macroscopic properties, including density, rattler fraction, and degree of order as measured by various standard order metrics [3]. By investigating multiple protocols that differ considerably, we seek to discern what features are protocol-dependent, and which are in common to a diversity of MRJ-like states. We also briefly consider the behavior of the hard-sphere FCC crystal, conjectured to be the equilibrium phase [48, 119] at packing fractions $\phi \in [0.55, \phi_{FCC})$ along the solid branch ending at close packing with a packing fraction of $\phi_{FCC} = \pi/\sqrt{18}$. This case provides valuable information about jamming under an arguably more well-behaved setting, where one need not worry about metastability, and hyperuniformity may be approached to arbitrary numerical precision with minimal practical issues.

We observe that the TJ and LS protocols exhibit markedly different qualitative
behavior in \( c(r) \), even at packing fractions far from jamming. Specifically, we find that the direct correlation of packings produced by the TJ algorithm exhibit signs of a delta function at \( r = D \) at packing fractions below the freezing density \( \phi \approx 0.494 \), whereas features in \( c(r) \) exhibited by LS for \( r > D \) are substantially more subtle up until much higher densities. With the development of the expected delta function at \( c(r = D) \), we observe a concomitant development of a dominant \( -1/r \) scaling for \( c(r < D) \), and we demonstrate that these two features are causally linked to each other. Interestingly, we observe power-law scaling in \( c(r) \) for large \( r \) that is predicted by the linear trend in \( S(k) \) for small \( k \). Observing this behavior is difficult in practice because it requires an accurate measurement of \( S(k) \) for small wave numbers, which requires that one consider large packings. Our work advances the notion that static signatures are exhibited by hard-particle packings as they approach jamming and underscores the utility of the direct correlation function as a sensitive means of monitoring for the appearance of an incipient rigid network.

The remainder of the paper is organized as follows: in Sec. 5.2, we discuss some relevant analytical results pertaining to the structure of disordered hard-sphere packings through \( c(r) \) and the Ornstein-Zernike equation. In Sec. 5.3, we discuss quantitatively the manner by which \( c(r) \) inherits singularities from \( g_2(r) \). In Sec. 5.4, we review some known facts regarding the critical scaling behavior expected for systems approaching hyperuniformity. In Sec. 5.5, we review the protocols that we use to generate nearly-jammed hard-sphere packings. In Sec. 5.6, we present the structure factor and direct correlation functions of our ordered and disordered packings as they approach jamming and point out emergent static structural features exhibited by each. Conclusions and discussion are presented in Sec. 5.7.
5.2 Ornstein-Zernike Equation and Jamming

We begin by reviewing a number of relationships between the standard pair statistical descriptors of point processes with the goal of relating the direct correlation function to other familiar statistical descriptors. We then proceed by reviewing theoretical progress that has been made in obtaining an accurate description of disordered hard-sphere systems by use of the direct correlation function, including shortfalls that persist with the current state of the art which point to the necessity for our present numerical investigations.

The structure factor is defined for a translationally-invariant system in $\mathbb{R}^d$ as

$$ S(k) = 1 + \rho \tilde{h}(k), \quad (5.2) $$

where $\tilde{h}(k)$ is the Fourier transform of the total correlation function. This is related to the scattering intensity $S(k)$, defined for a single system of $N$ particles within a fundamental cell under periodic boundary conditions as

$$ S(k) = \frac{1}{N} \left| \sum_{j=1}^{N} e^{-ik \cdot r_j} \right|^2, \quad (5.3) $$

which includes forward scattering, i.e., $S(0) \equiv N$. This is to be contrasted with the definition of Eq. (5.2), in which $S(0)$ is related to the volume integral of $h(r)$. Except at $k = 0$, the scattering intensity is identical to the structure factor for a single configuration. For an ensemble of such periodic point configurations (e.g. derived from the particle centers of our packings), the ensemble average of $S(k)$ is directly related to the structure factor $S(k)$ via

$$ \lim_{N,v \to \infty} \langle S(k) \rangle = (2\pi)^d \delta(k) + S(k), \quad (5.4) $$

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where $\delta(k)$ is the Dirac delta function and the limit being taken on $N$ and the fundamental cell volume $v_F$ are such that the relevant physical system (e.g. unjammed packings at some constant density or at some given distance to the jamming density $\phi_c$) is preserved. In practice, we directly compute $S(k)$ from our simulation data and average the data from a large number of packings with the same system size to approximate $S(k)$ for the ensemble of packings generated by a given protocol, keeping in mind that finite-system artifacts are expected to persist to some degree. In all cases, the quantity $S(0)$ must be inferred through extrapolation to the origin. Using this ensemble average, one may combine the Fourier transform of Eq. (5.1) with Eq. (5.2) in order to express the Fourier transform of the direct correlation function as

$$\tilde{c}(k) = S(k) - \frac{1}{\rho S(k)},$$

(5.5)

from which $c(r)$ may be computed by inverse Fourier transform.

A number of closure relations have been proposed to offer approximate solutions to Eq. (5.1). In particular, the Percus-Yevick (PY) closure demands that $h(r) = -1$ for $0 \leq r < D$ and $c(r) = 0$ for $r > D$ for monodisperse spheres of unit diameter. Physically, these requirements specify that no two spheres may overlap, and that direct interactions [in the sense of Eq. (5.1)] are absent beyond the particles’ hard cores. We would like the former criterion to apply in any solution for hard-spheres, but the latter assumption is increasingly violated as the packing fraction $\phi = \pi \rho/6$ increases. The cubic polynomial form of $c(r)$ produced by the PY closure that solves Eq. (5.1) accurately describes much of the equilibrium liquid branch of the hard-sphere system with good quantitative agreement for $\phi < 0.40$ and qualitative agreement for $\phi < 0.49$. However, it possesses various shortcomings at higher densities still within the liquid branch that one might like to improve: it underpredicts $g_2(1^+)$ (and thus the

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1To be mathematically precise, the statistical functions $g_2, h, S, \text{and } c$ are all functions which map $\mathbb{R}^d \rightarrow \mathbb{R}$; however, since these functions are often isotropic, we often express them in terms of $r = |r|$ or $k = |k|$ as appropriate.
pressure), and oscillations in the pair correlation function are out of phase and decay too slowly with increasing $r$ [91]. At higher densities, $g_2(r)$ fails basic satisfiability criteria such as nonnegativity and hence ceases to be physical.

To address this shortcoming, a variety of adjustments have been made to the PY approximation to improve the range of densities over which it may apply. A classic approach is to introduce a Yukawa term beyond the core [120, 121, 122], i.e., $c(r > D) = Ke^{-z(r-D)}/r$. This improves the degree to which the approximation matches qualitative features in $c(r)$ and provides additional fitting parameters to allow for a quantitative match of additional system properties. To this end, the recent work of Jadrich and Schweizer [123] used a two-Yukawa generalized mean spherical approximation which allowed them to match the system’s compressibility as well as $g_2(D^+)$ and its first derivative in an attempt to accurately describe the behavior of the hard-sphere system along some metastable branch leading towards a disordered jammed state. By allowing $z$ and $K$ to approach infinity, this model may capture the appearance of a delta function at $c(r = D)$, and predicts a functional form for $c(r)$ inside the core that departs from the solution to the Percus-Yevick approximation.

For a single-Yukawa form with $d = 3$, one finds [120]

$$c(x) = -a - bx - \phi ax^3/2 - \nu \frac{1 - e^{-zx}}{zx} - \nu^2 \frac{\cosh(zx) - 1}{2K^2e^z}$$  \hspace{1cm} (5.6)$$

for $x < 1$, where $x = r/D$, $a = 1 - 24\phi \int_0^\infty c(x)x^2dx$, $\nu = 24\phi \int_1^\infty xe^{-z(x-1)}g_2(x)dx$, $b$ satisfies

$$24\phi y_0^2 = -4b + 2\nu z - \frac{\nu^2}{Ke^z}$$  \hspace{1cm} (5.7)$$

$$24\phi (y_1^2 - 2y_0y_2) = 24\phi a - 2\nu z^3 + \frac{\nu^2 z^2}{Ke^z}$$  \hspace{1cm} (5.8)$$

and $y_i = (d^i/dx^i)[xg_2(x)]_{D+}$. Interestingly, upon taking $k$ and $z$ towards infinity so as to obtain a delta function at $c(r = D)$, we see that the fourth term in Eq. (5.6)
scales as $-1/r$ for $r_z < r$, where $r_z \approx 1/z$. Thus, as $z$ is taken to infinity, $r_z$ goes to zero. For $r < r_z$, the term saturates to a constant. Note that, in order for this scaling behavior to imply that $c(r)$ diverges at the origin, one must have $\nu \to \infty$. This is not necessarily implied by this analysis.

While this approach improves upon these features, it still lacks the correct long-$r$ scaling behavior for disordered packings. Accordingly, the small-$k$ behavior of the structure factor is not in qualitative agreement either. As a consequence, one would expect that $c(r)$ decays as a power-law, whereas the Yukawa form predicts exponential decay. In addition to this, several salient features in the pair correlation function, including (i) the power-law divergence as $r \to D^+$, (ii) the cusp at $r/D = \sqrt{3}$, and (iii) the correct step discontinuity at $r/D = 2$, remain elusive.

### 5.3 Inheritance of features in $c(r)$ from $g_2(r)$

Here, we present a derivation for the magnitude of the step discontinuity in $g_2(2)$ in terms of the information in $c(r)$ for the specific case of disordered, jammed hard-sphere packings in three dimensions. Critically, we assume throughout this analysis that (i) $g_2$ and $c$ are isotropic (radial) functions and that (ii) the packings are isostatic, meaning that the average contact number for backbone spheres in the packing is $z = 6 + \mathcal{O}(1/N_B)$, where $N_B = (1 - f_r)N$ is the number of backbone spheres in a packing with rattler fraction $f_r$. It is estimated that $f_r = 0.015$ for TJ and $f_r = 0.025$ for LS [3]. The particular form of the $\mathcal{O}(1/N_B)$ term depends upon whether one is considering collective or strict jamming, but becomes irrelevant in the infinite-system limit [66]. The pivotal observation from which this analysis follows is that a singularity of a given order in $c$ may not contribute to a singularity in $g_2$ of lower order. For example, a step discontinuity in $c$ cannot cause a delta function to appear in $g_2$. This assumption is justified by recursively inserting the form of $h(r)$ into the
Ornstein-Zernike relation to obtain

\[
  h(r_{12}) = c(r_{12}) + \rho \int_{\mathbb{R}^3} c(r_{13})c(r_{23})dr_3 \\
  + \rho^2 \int_{\mathbb{R}^3} c(r_{13})c(r_{34})c(r_{24})dr_3dr_4 + \ldots
\]

(5.9)

and noting that each successive convolution ought to increase the order of the differentiability class of the term by one.

We begin with the delta function at \( g_2(1) \). By Eq. (5.9), we see immediately that the only contribution to this must come from a delta function in \( c(1) \), and that the two must be of equal magnitude. The number of particles that are separated at a distance \( r \) is given by

\[
z(r) = \lim_{\epsilon \to 0} \int_{r-\epsilon}^{r+\epsilon} 4\pi x^2 \rho g_2(x)dx.
\]

We substitute \( z(1) = 6f_b \), where \( f_b = 1 - f_r \) is the backbone fraction, and obtain the result that the strength of the delta function in \( g_2(1) \) is equal to \( f_b/4\phi \).

We now proceed to identify the source of the jump discontinuity found at \( g_2(r = 2) \). We begin by decomposing \( c(r) \) into three parts:

\[
c(r) = c_0(r) + c_\delta(r) + c_\Theta(r),
\]

(5.10)

where \( c_\delta(r) = f_b \delta(r - 1)/4\phi \) is the delta function contribution from above; \( c_\Theta(r) = A(1 - \Theta(r - 2)) \), where \( \Theta(x) \) is the Heaviside step function, captures the step discontinuity predicted in \( c \) at \( r = 2 \), and \( c_o(r) \) captures the remainder of the direct correlation and is assumed to be at least continuous at \( r = 2 \). Figure 5.1 shows an illustrative example where the same sort of decomposition has been applied to the direct correlation function given by the Percus-Yevick approximation, where there is no delta function, but there is a step discontinuity at \( r = 1 \).
Figure 5.1: A decomposition of the direct correlation function following the approach of Eq. (5.10). Here, \( c(r) \) is given by the solution under the Percus-Yevick approximation at a density of \( \phi = 0.42 \). Because there is no delta function in \( c(r) \), \( c_\delta(r) = 0 \) everywhere and therefore is not shown.

Substituting this into Eq. (5.9) gives

\[
\begin{align*}
  h(r_{12}) &= c_\circ(r_{12}) + c_\delta(r_{12}) + c_\Theta(r_{12}) \\
  &+ \rho \int_{\mathbb{R}^3} [(c_\circ(r_{13}) + c_\delta(r_{13}) + c_\Theta(r_{13})) \\
  &+ (c_\circ(r_{23}) + c_\delta(r_{23}) + c_\Theta(r_{23}))] \, dr_3 + \ldots
\end{align*}
\tag{5.11}
\]

We may first evaluate the right hand side of Eq. (5.11) with \( r_{12} = 2 + \epsilon \), then evaluate it with \( r_{12} = 2 - \epsilon \). Subtracting the former from the latter while letting \( \epsilon \to 0 \), we
obtain
\[ \Delta_c h(2) = \Delta_c c_0(2) + \Delta_c c_\delta(2) + \Delta_c c_\Theta(2) + \rho \Delta_c I(2), \quad (5.12) \]

where we have used the shorthand \( \Delta_c f(x) = \lim_{\epsilon \to 0} f(x + \epsilon) - f(x - \epsilon) \) and defined

\[ I(r_{12}) = \int_{\mathbb{R}^3} c(r_{13}) c(r_{23}) \, dr_3. \quad (5.13) \]

We notice that \( \Delta_c c_\delta(2) = \Delta_c c_0(2) = 0 \), meaning that the only surviving term of the first three is \( \Delta_c c_\Theta(2) = -A \). Furthermore, by considering the decomposition of Eq. (5.10) as applied to Eq. (5.13), we can see that the term with the differentiability class of the lowest degree is given by

\[ I_\delta(r_{12}) = \int_{\mathbb{R}^3} c_\delta(r_{13}) c_\delta(r_{23}) \, dr_3, \quad (5.14) \]

and that \( \Delta_c I_\delta(2) \) should be nonzero so long as the delta function represented by \( c_\delta \) has a nonzero amplitude (i.e. \( c_\delta \) is not trivially zero everywhere). All other terms contributing to \( I(r) \) are continuous at \( r = 2 \), as are all higher-order convolutions. Thus, we are left with

\[ \Delta_c h(2) = \rho \Delta_c I_\delta(2) - A. \quad (5.15) \]

To evaluate the first term, we define \( \bar{r} = (r_1 + r_2)/2 \), so that

\[ I_\delta(r_{12}) = \int_{\mathbb{R}^3} c_\delta \left( r_3 - \bar{r} + \frac{r_{12}}{2} \right) c_\delta \left( r_3 - \bar{r} - \frac{r_{12}}{2} \right) \, dr_3. \quad (5.16) \]

We invoke the convolution theorem to write

\[ \tilde{I}_\delta(k) = \tilde{c}_\delta(k)^2, \]

where we have invoked translational invariance to remove \( \bar{r} \) and the symmetry of the two terms within the integrand with respect to \( r_{12} \). Carrying out the inverse Fourier
transform gives the result

\[
\rho I_\delta (r_{12}) = \frac{\rho}{2\pi^2 r} \int_0^\infty \tilde{c}_3^2(k) k \sin kr \, dk
\]

\[
= \frac{\rho}{2\pi^2 r} \int_0^\infty \left( \frac{\pi}{\phi k} \sin k \right)^2 k \sin kr \, dk
\]

\[
= \frac{3}{\pi \phi r} \int_0^\infty \frac{1}{k} \sin^2 k \sin kr \, dk
\]

\[
= \frac{3}{4\phi r} (1 - \Theta(r - 2)).
\]

This result is plotted in Fig. 5.2 using \( \phi = 0.64 \), as is typical for MRJ-like packings.

Since \( I_\delta \) represents the “sharpest” contribution from the single convolution term (i.e. the convolutions of all other pairs of terms will be continuous at \( r = 2 \)), we can
conclude immediately that no other terms within the first convolution term (and no further convolution terms) will contribute to the quantity \( \lim_{\epsilon \to 0} h(2 + \epsilon) - h(2 - \epsilon) \) since they will be too smooth (i.e. they do not retain a step discontinuity following convolution). Thus, we arrive at the result that the magnitude of the step discontinuity in the total correlation function (and, equivalently, the pair correlation function) at \( r = 2 \) is

\[
\Delta_{\epsilon} h(2) = \Delta_{\epsilon} c(2) - \frac{3}{8\phi}.
\]  

(5.18)

By extending this analysis, we can claim that even in the absence of any additional nonanalytic features in \( c(r) \), \( g_2(r) \) is expected to have discontinuities in successively higher derivatives at further integer values of \( r \). For example, there ought to be a discontinuity in the first derivative of \( g_2 \) at \( r = 3D \), a discontinuity in the second derivative at \( r = 4D \), and so on.

### 5.4 Scaling relations for systems in the vicinity of hyperuniformity

In this section, we recall various scaling behaviors for various pair statistics in direct and Fourier spaces for ordered and disordered packings of hard spheres in the vicinity of jamming that are particularly germane to this paper.

Torquato and Stillinger have shown [17] that a hyperuniform system with a structure factor that scales as

\[
S(k) \propto k^{2-\eta}, \quad k \to 0
\]

(5.19)

may be thought of as an “inverted critical point.” At this point, the direct correlation becomes long-ranged, scaling as

\[
c(r) \propto -r^{2-d-\eta}, \quad r \to \infty
\]

(5.20)
in dimension $d$, where $\eta$ is a critical exponent such that $2 - d < \eta < 2$. Additionally, the inverse of the structure factor at the origin exhibits critical scaling behavior in the vicinity of its critical density, i.e.,

$$S^{-1}(0) \propto (1 - \phi/\phi_c)^{-\gamma} \quad (5.21)$$

for densities close to, but below $\phi_c$ [17].

In the case of the equilibrium crystal branch, we may exploit the compressibility relation relating the structure factor to the isothermal compressibility $\kappa_T$ at temperature $T$

$$S(0) = \rho k_B T \kappa_T \quad (5.22)$$

along with the free-volume equation of state [124] which predicts that the pressure $p$ behaves as

$$\frac{p}{\rho k_B T} = \frac{d}{1 - \phi/\phi_J}, \quad (5.23)$$

to obtain the result that $\kappa_T \propto (1 - \phi/\phi_c)^2$ in the vicinity of jamming. From this, it follows that

$$\gamma = 2 \text{ (equilibrium crystal).} \quad (5.24)$$

Note that this result is independent of dimension. One may also define a correlation length $\xi$ with the critical behavior

$$\xi \propto (1 - \phi/\phi_c)^{-\nu} \quad (5.25)$$

which may be related to the previous critical exponents through

$$\gamma = (2 - \eta)\nu. \quad (5.26)$$

As mentioned before, one such length scale can be defined by the volume integral of
the direct correlation function: \( \xi = (-\int_{\mathbb{R}^3} c(r)dr)^{1/d} \).

For packings along glassy metastable branches leading to disordered jammed states, one cannot use the compressibility relation because the states are nonequilibrium in nature [47]. However, one may reconcile the differing pictures presented by \( S(0) \) and \( \kappa_T \) by introducing a “nonequilibrium index”, defined as

\[
X \equiv \frac{S(0)}{\rho k_B T \kappa_T} - 1. \tag{5.27}
\]

Hopkins et al. studied the behavior of \( X \) under rapid compression toward jamming for disordered packings prepared by the LS algorithm for system sizes up to \( N = 10^6 \) and found that, as \( \phi \) increased toward jamming, \( X \propto (1 - \phi/\phi_c)^{-1} \) [47].

By combining this result with the observation that the pressure in disordered packings again diverges according to the free-volume equation of state, we get the result that

\[
\gamma = 1 \text{ (MRJ packings).} \tag{5.28}
\]

By Eq. (5.26), this implies that \( \nu = 1 \) for MRJ packings as well. This is a noteworthy result because it tells us that while both disordered and ordered packings become increasingly hyperuniform as they are compressed, they are different universality classes with respect to the critical exponents \( \eta \) and \( \gamma \) associated with hyperuniformity.

### 5.5 Simulation Methods

To study the behavior of the equilibrium hard-sphere FCC crystal for densities between \( \phi = 0.55 \) and the close-packing density \( \phi_{FCC} = \pi/\sqrt{18} \), we used standard event-driven molecular dynamics [55]. Configurations of \( N = 4M^3 \) spheres with \( 4 \leq M \leq 63 \) were placed on their lattice sites and allowed to equilibrate at fixed packing fraction within a cubic fundamental cell with periodic boundary conditions.
for $10^5$ collisions per sphere before taking statistics. Measurements of the structure factor were made every $10^3$ collisions per sphere to verify that equilibrium had been attained.

To generate disordered, strictly jammed sphere packings in three dimensions, we begin with initial conditions produced by random sequential addition (RSA) at an initial packing fraction of $\phi = 0.10$. We use the Torquato-Jiao (TJ) sequential linear programming method [61] with system sizes of up to $N = 10^4$ using the same parameters as those used to study the MRJ state in Ref. [3]. The final mean density of these packings is $\phi = 0.6352 \pm 2.6 \times 10^{-4}$ for a system size of $N = 2000$. After the algorithm terminates and a putatively jammed state is reached, the packing is equilibrated within its jamming basin using event-driven molecular dynamics at fixed density. We also compare these packings to those generated using the well-known event-driven Lubachevsky-Stillinger (LS) molecular dynamics algorithm [51]. For LS, we use an initial dimensionless growth rate of $\Gamma = dD/dt \sqrt{m/(k_B T)} = 10^{-2}$ until packings reach a dimensionless pressure of $P = pV/Nk_B T = 10^4$, at which point the expansion rate is slowed to $\gamma = 10^{-5}$, and packing continues until $P = 10^8$. The mean density of the final packings as prepared under this protocol is $\phi = 0.6434 \pm 1.0 \times 10^{-4}$ at a system size of $N = 10000$.

### 5.6 Results

In this section, we present results pertaining to our computer-generated hard-sphere packings as they approach both ordered and disordered jammed states. We will present our analysis assuming that the particle diameter $D$ is unity unless otherwise specified.
5.6.1 FCC

We begin by examining the behavior of the hard-sphere FCC crystal because the behavior exhibited by the crystal undergoing thermal motion away from jamming serves as an interesting starting point from which we can make several observations to guide our subsequent investigation of disordered jammed systems. We will investigate the crystal’s approach to close-packing with attention to the fluctuating component of the structure factor as well as the implications it has for the qualitative form of \( c(r) \).

Figure 5.3 shows plots of the radially-averaged structure factor \( S(k) \) for the equilibrated FCC crystal for a variety of densities along the solid branch; our computation follows the collective coordinate formulation of Eq. (5.3). Curves are averaged over ensembles of 100 packings with \( N = 2048 \). Figure 5.4 shows the corresponding radially-averaged direct correlation function evaluated numerically using discrete Fourier transform techniques following Eq. (5.5).

In addition to the Bragg peaks that arise from the crystal geometry from the “frozen-in” structure of the packings, the curves possess a background contribution derived from thermal fluctuations which scales as \( k^2 \) starting at a sufficiently-high wave number (on the order of unity) and saturates to \( S = 1 \). While the domains of interest in the scaling descriptions of Eqs. (5.19) and (5.20) as written are \( k \to 0 \) and \( r \to \infty \), one might reasonably suspect that it is possible to invert the two limits so as to infer the scaling behavior of \( c(r \to 0) \) from \( S(k \to \infty) \). In the case of our FCC data, we see that this implies that the direct correlation function ought to scale as \( r^{-1} \) for \( r < 1 \). Looking at Fig. 5.4, this seems to be the case. We note in passing that this is also consistent with the prediction given by the analysis of the Yukawa form mentioned in Sec. 5.4.

At sufficiently low wave numbers, \( S(k) \) converges to a constant, given to a good approximation as \( S(0) = (1 - (\phi/\phi_{FCC})^{1/3})^2 \), confirming that the critical exponent
Figure 5.3: The structure factor of the hard-sphere FCC crystal at various densities and system size $N = 2048$. Dashed lines showing $S = (1 - (\phi/\phi_{FCC})^{1/d})^2$ highlighting that the critical behavior of $S(0)$ is included.

$\gamma = 2$ as predicted in Eq. (5.24). Importantly, at the point of exact jamming, the FCC crystal is trivially stealthy (its structure factor is identically zero up to some positive wave number), meaning that the critical exponent $\eta$ may be thought of as being infinite. This discontinuous change from the equilibrium behavior at even vanishingly small distances to jamming highlights the singular nature of jamming and underscores the need to be careful when considering limiting behavior.

It is important to note that a structure factor that scales as $k^2$ can be obtained by applying random, uncorrelated displacements to each particle in the crystal [125]. Given the inherent anharmonicity of the system owing to the singular nature of the hard-sphere interaction potential, we are motivated to ask whether the probability
distribution of the pair separation of nearest-neighbor spheres in the crystal might be, to a good approximation, statistically independent from pair to pair on certain length scales. On larger length scales (lower wave numbers), the exact form of $S(k)$ is in excellent agreement with a normal mode analysis based on the fictitious interparticle potential derived in [126], suggesting underlying correlated displacements on corresponding length scales. In the next Chapter, we investigate the consequences of this by analyzing the percolation properties of an intimately-related “cherrypit model” [127].
5.6.2 Disordered Packings and the MRJ state

We now turn our attention to characterizing the approach to jamming in disordered packings produced by the LS and TJ algorithms as described in Sec. 5.5. Unlike the case for the FCC crystal, MRJ-like packings do not strongly indicate signs of an incipient jammed structure at densities far below that of jamming. Furthermore, there is a strong protocol dependence yielding qualitative differences in various packing protocols’ approaches towards disordered, jammed states.

Figure 5.5 shows the ensemble-averaged structure factor of our packings for a variety of densities and protocols. As jamming is approached, \( \lim_{k \to 0} S(k) \) approaches zero, implying that the jammed state is hyperuniform, in agreement with previous investigations [56, 47]. Because the data are presented on a log-log scale, the vertical offset between the nearly-jammed TJ and LS configurations corresponds to a difference in slope in their respective linear behavior in the vicinity of the origin. Interestingly, the packings produced by the TJ algorithm display anomalous behavior well before jamming is reached, including a structure factor that increases as the wave number approaches zero for intermediate densities. By contrast, \( S(k) \) for LS-generated packings seems to monotonically decrease for \( kD/2\pi < 1 \) at all packing fractions leading up to jamming. This suggests that the intermediate configurations that TJ creates on its way to jamming are highly nonequilibrium in nature—even at packing fractions below the freezing density \( \phi_f \approx 0.494 \) [128]. For configurations that are close to jamming at a density of \( \phi_c \), we group packings according to the quantity \( 1 - \phi/\phi_c \) rather than \( \phi \).

The TJ algorithm seems to find configurations that are consistently more disordered than those visited by LS starting at intermediate densities and continuing up
Figure 5.5: The structure factor for ensembles of packings created using TJ (solid lines) and LS (dashed lines) at various packing fractions $\phi$ and distances to jamming $1 - \phi/\phi_c$.

to jamming\(^2\). One may consider the order metric

$$\tau = \frac{1}{(2\pi)^d} \int_{|k|<k_{\text{max}}} (S(k) - 1)^2 dk,$$

which may be thought of as quantifying extent to which a given configuration differs from a Poisson process (for which $S = 1$ for all $k$). This order metric was used in [6] with $k_{\text{max}} \to \infty$. The order metric $\tau$ is also reminiscent of the direct-space order metric $T^*$ that we have utilized before [3], which measures deviations in the pair correlation function from unity, as well as the two-body excess entropy $s^{(2)}$ [84,\(^2\)for very low densities, the RSA configurations that are given to TJ as initial conditions are rather similar to the equilibrium fluid.}
Here, we keep $k_{\text{max}}$ finite to prevent $\tau$ from diverging due to the contributions of Dirac delta functions in the corresponding direct-space statistics characteristic of jammed packings; a similar issue arises in the aforementioned order metrics as discussed in [84].

Looking at Fig. 5.5, one can see immediately that $S(k)$ for TJ-generated packings remains much closer to unity at intermediate densities than for LS. This difference persists up to jamming. Figure 5.6 shows the quantity $\tau$ computed for ensembles of 1000 packings created by the TJ and LS algorithms corresponding to the densities used for Fig. 5.5 as a function of $k_{\text{max}}$. Consistent with other order metrics including $T^*$ as well as the standard bond-orientational order metric $Q_6$, $\tau$ demonstrates that the packings generated by TJ are more disordered than those produced by LS [3]. However, unlike $T^*$, which focuses on short-range order, $\tau$ focuses primarily on long-range order. Thus, it provides complimentary information to $T^*$.

Near jamming and at sufficiently high wave numbers, the behavior of the integrand in Eq. (5.29) is dominated by the contribution arising from the delta function in $c$ at $r = D$. This causes $\tau$ to diverge toward infinity at a rate that is linear in $k_{\text{max}}$. The thick black line in Fig. 5.6 illustrates the slope associated with this behavior. If the delta function is not sharp (because of a spread in nearest-neighbor distances), then $\tau$ will saturate to a constant. This is seen for the LS and TJ packings for $\phi = 0.40$ and 0.50 as well as for LS at $\phi = 0.60$. This suggests that, while near-contacts accumulate as TJ densifies its packings, these are not necessarily true contacts; $\tau(k_{\text{max}})$ demonstrates by its saturation that the nearest neighbors in the packings are spread out over a range of pair distances.

Because of the noise in measuring the structure factor numerically (due to both a finite number of packings in the ensemble as well as finite system sizes), the decaying oscillations converging to $S = 1$ will eventually saturate to white noise. Therefore, $\tau$ will begin to grow with increasing cutoff as $k_{\text{max}}^d$. The beginning of this trend is
Figure 5.6: The order metric $\tau$ for ensembles of packings created using TJ (solid lines) and LS (dashed lines) at various packing fractions $\phi$ and distances to jamming $1 - \phi/\phi_c$ for different cutoffs $k_{\text{max}}$. Visible in Fig. 5.6 for our ensembles at lower densities.

Figure 5.7 shows the corresponding direct correlation functions for these ensembles of packings. The first salient feature is that the direct correlation function for the packings produced by the TJ algorithm exhibits a prominent peak at $r = 1$ that is clearly visible at packing fractions as low as $\phi = 0.50$. This is accompanied by a steep decrease in $c(r)$ for $r < 1$ that is dominated by a $-1/r$ scaling. As mentioned in Sec. 5.2, the analysis of a Yukawa-like $c(r)$ beyond the core as $z$ goes to infinity [120, 132, 121, 122, 123] provides the rationalization for the appearance of a scaling behavior of this form. However, it only becomes dominant if the volume integral of $c(r)$ outside the core is sufficiently large. That is, the short-ranged behavior of $c(r)$...
seems to be, fascinatingly, communicating its growing long-rangedness in the sense of the theoretical considerations of Sec. 5.4.

The early appearance of a delta function in $c$ at $r = 1$ leads us to suggest that cluster formation in TJ occurs long before the packing is confined to a jamming basin. This result is likely related to the observation of Shen et al. [133] that athermal packings of spheres compressed from low densities in the presence of a viscous background exhibit a “contact percolation” which is accompanied by the emergence of a nontrivial mechanical response to applied stress at densities significantly below that of jamming. Because of the manner in which the sequential linear programming algorithm searches for local optimizations in packing fraction, which require little reconfiguration at low densities, there is reason to believe that the TJ algorithm explores available configuration space in a similar fashion to the procedure of Shen et al. for low to intermediate densities.

We also note in passing that the direct correlation functions exhibit both a cusp at $r = \sqrt{3}$ and a mild step discontinuity at $r = 2$, mirroring the features found in the pair correlation function. We have noticed that the step discontinuity observed in $g_2$ of jammed packings is consistently larger than what is produced as an effect of the delta function at $c(r = 1)$; this is explained in light of the result contained in Eq. (5.18). Note also that the particular form of Eq. (5.18) relies on the assumption that the packing is isostatic, meaning that there are the minimum number of backbone contact pairs necessary to ensure jamming. In the infinite system limit, this corresponds to a mean backbone coordination number of $z = 6 + \mathcal{O}(1/N)$, where the vanishing term reflects the difference between collective or strict jamming [66].

Figure 5.8 shows $-c(r)$ plotted on a log scale. As jamming is approached, we observe that $c \propto -1/r^2$ for large $r$, confirming numerically the prediction of Eq. (5.20) for $\eta = 1$ in the case of disordered packings. This scaling behavior is difficult to obtain numerically since one must accurately obtain $S(k)$ data for low wave numbers.
Figure 5.7: Direct correlation functions computed for packings generated by (a) LS and (b) TJ for various densities.
in order to extract the large-$r$ behavior of $c$. In particular, one must necessarily extrapolate $S(0)$, as a direct computation using Eq. (5.3) contains a forward scattering contribution which must be omitted. Additionally, to improve numerical stability, our $\tilde{c}(k)$ data were multiplied by the Fourier transform of a narrow triangular window so that the real-space data is smoothed accordingly via convolution. Details of the procedure are given in Appendix 5.8.

The difference in the protocols’ approaches to jamming may be readily traced back to differences in the dynamics involved: on one hand, the constant thermal motion inherent to LS acts to equilibrate packing and avoid metastable branches terminating at low-density jammed states; an aggressive expansion rate works against this, though one must worry about the algorithm becoming trapped in an unstable mechanical equilibrium (which is, by definition, not a jammed state). The possible displacements obtained by TJ are highly degenerate since, in general, there are many different displacements which allow for the same increase in the packing fraction (which is limited to a small value so that the linear approximations in the LPs’ formulation remain reasonably accurate). Therefore, TJ tends to displace spheres in a “lazy” fashion, only moving what is necessary to increase the packing fraction and no more.

Evidence of this aforementioned qualitative difference may be observed directly; Figure 5.9 shows snapshots of two-dimensional packings of monodisperse disks created by TJ and LS (using a rapid compression rate) at a packing fraction of $\phi = 0.55$. In two dimensions just as in three, we can see that the structures of the packings are qualitatively different. In particular, the TJ algorithm exhibits clustering of particles that might be quickly dispersed through thermal motion; in the absence of this, the clusters continue to combine and aggregate as jamming is approached. We point out that particles within these clusters do not necessarily contact one another; some separation is expected to remain between particles owing to the nonlinearities that are not captured in the TJ algorithm’s linear approximation to the packing
Figure 5.8: Log-log representation of $-c(r)$ for (a) LS and (b) TJ packings at system sizes of $N = 10000$ and $N = 2000$, respectively. Data for positive $c$ is shown using dashed lines of the same color in (b). The thick black dashed guide line shows a slope of $-1/r^2$. 

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problem. In LS, on the other hand, particles tend to space themselves out more uniformly through their thermal motion. We note in passing that, while this difference does not prevent the LS algorithm from discovering MRJ-like states in 3D, the two-dimensional case was recently shown to be considerably more subtle—the difference in how TJ creates jammed packings has led to the first observations of MRJ-like packings of monodisperse disks in two dimensions, whereas the LS algorithm and other standard protocols are unable to observe them, finding significantly more ordered, polycrystalline structures even under rapid compression [109].

Figure 5.9: Two-dimensional packings of monodisperse disks created by the (a) LS and (b) TJ algorithms at a packing fraction of $\phi = 0.55$.

5.7 Conclusions and Discussion

In this work, we have compared and contrasted the approach of both ordered and disordered hard-sphere packings towards jammed states through considering the behavior of their structure factors and direct correlation functions. By considering the degree and position of singularities in $c(r)$ as well as how they are changed by the convolutions found in Eq. (5.1), we have established quantitative statements about the structure of the direct correlation function with regards to features it inherits from $g_2(r)$. These relations provide a concrete means of identifying what features must be
expressed in \(c(r)\) if one hopes to reproduce various details in \(g_2(r)\) accurately.

Moreover, we found that the LS and TJ protocols approach their respective jammed states in markedly different manners, as shown by various pair statistics. Specifically, the structure factor of TJ-generated packings shows anomalous increasing behavior for small \(k\) at intermediate densities, and generally remains closer to \(S = 1\) at all densities leading up to jamming when compared to LS. The order metric \(\tau\) compares a configuration’s pair statistics to that of an uncorrelated (Poisson) point process, which may be thought of as a maximally disordered reference state. In this sense, \(\tau\) may be thought of as a “disorder metric”. At low to intermediate densities, \(\tau\) suggests that packings created by TJ are more disordered on large length scales, but more ordered on short length scales as evidenced by the crossover as the truncation in the integration domain \(k_{\text{max}}\) is made increasingly large. This is consistent with the intuition that TJ does not disturb the packings as they are compressed as much as LS does, leaving the large-scale characteristics similar to the initial conditions obtained from low-density RSA. On the other hand, the formation of near-contacts well before jamming may be interpreted as a sort of “ordering” that LS avoids through equilibration; therefore, LS yields configurations that are more disordered locally at densities far from jamming.

TJ shows signs of particles in close proximity at surprisingly low densities as evidenced by the appearance of a clear precursor to the delta function at \(c(1)\) and corresponding \(-1/r\) scaling within the core. By evaluating \(\tau\), we see that these near-contacts are distributed across a range of pair separations, and the delta function’s precursor is not “sharp” until higher densities. This is to be expected because of the linear approximations that TJ makes as the packing is compressed; the inaccuracies due to nonlinear contributions are largest when large changes in the system configuration (particle translations and box deformations) are made. This is the case at densities far from jamming, where the linear approximations to the packing problem
still leave a large amount of configuration space accessible. Nonetheless, this feature in the intermediate-density structures produced by the TJ algorithm suggests that it possesses important qualitative commonalities with the physical process of compressing hard-spheres embedded in a dampening background, providing a conceptual physical analog to the algorithm as witnessed in practice.

It has been suggested previously [17, 47] that the hyperuniform, linear nonanalytic behavior of $S(k)$ for MRJ-like packings ought to give rise to a long-ranged direct correlation function which exhibits a power-law decay of $c \propto -1/r^2$. We have confirmed this numerically using simulated packings of hard-spheres generated by two very different protocols, adding to the evidence to the conjectured link between jamming and hyperuniformity [56, 1] and supporting the idea that the emergence of large-$r$ scaling behavior consistent with hyperuniformity may be regarded as a structural precursor to jamming. It would be interesting to consider new semi-empirical forms for $c(r)$ incorporating this long-range behavior in order to gain understanding regarding its structural consequences.

It is interesting that the structure factor of the FCC crystal exhibits scaling that is constant for low $k$, but gives way to $k^2$ beginning at wave numbers on the order of unity, extending to an increasingly large maximum wave number as jamming is approached. We noted above that this would imply that the average pair distance between any given pair of nearest-neighbor particles might be spatially uncorrelated to a good approximation. It has been observed elsewhere [59] that the fluctuating component of the structure factor in disordered packings of thermally-excited soft-spheres exhibits a similar quadratic scaling at densities slightly above the jamming transition density. We have noticed that this behavior is also exhibited for disordered hard-sphere packings at packing fractions below, but close to jamming, suggesting similarly that the pair separations between nearest-neighbor particles may fluctuate in an uncorrelated manner to a good approximation.
We suspect that the aforementioned differences between the LS and TJ algorithms should be evident in other ways. In the next Chapter, we will study our hard-sphere systems in the context of two different percolation problems. In the first, we decorate the hard cores with a perfectly penetrable shell (this is known as the “cherrypit model” [91]) and tuning the size of this shell for various configurations to explore percolation criticality. Based on our findings here, one would expect for TJ that the critical shell thickness would become very small rather quickly, whereas it might decrease more steadily towards zero for LS as jamming is approached. Moreover, one might expect to find structural differences in the percolating clusters between these two algorithms. Second, we investigate percolation by measuring the time-averaged magnitudes of pair forces in nearly-jammed, structurally-arrested configurations as a function of a minimum “threshold” force. This approach serves to “average out” fluctuations in particle position, and thus provides insight into the role of fluctuations in the incipient structure of disordered, jammed systems. Both approaches provide insight into the jamming process using static structural features, expanding upon the work presented here.
5.8 Appendix: Numerical procedure for obtaining $c(r)$ from $S(k)$ data

In this appendix, we provide procedural details regarding our numerical computation of the direct correlation function used to ascertain the large-$r$ behavior.

We begin by measuring $S(k)$ at wave vectors that are integer combinations of the columns of the reciprocal matrix $\Lambda_R$ defined as $\Lambda_R = [(2\pi)\Lambda^{-1}]^T$, where the columns of $\Lambda \in \mathbb{R}^{d \times d}$ span the fundamental cell of our system in direct space. Measurements are then binned and averaged according to their wave number with a bin width of $\Delta k/2\pi = 0.01$, so that we have reported measurements at wave numbers given by $k_n = (\Delta k/2\pi)(1/2 + n)$ for $n = 0, 1, 2, \ldots$. Because the density of wave vectors scales as $k^{d-1}$, we randomly select wave vectors with a probability proportional to the inverse of this density such that, for higher wave numbers, the expected number of measurements per bin is $E(n_s) = 10^5$. At smaller wave numbers, the structure factor is measured at every available wave vector.

Once we have obtained data for all of our packings, we perform an ensemble average. If there are empty bins, then we linearly interpolate a value for those bins, expecting that this is representative of the large-system limit. We also linearly extrapolate $S(k)$ down to zero if any bins are missing data; our results are qualitatively robust against small variations in this extrapolation.

Our data is then converted via Eq. (5.5) to give us $\tilde{c}(k)$. Because of the asymptotic behavior of $\tilde{c}(k)$, we find that it is necessary to apply a convolution in order to eliminate artifacts caused by difficulties associated with numerically integrating this high-frequency behavior. We do this by multiplying $\tilde{c}(k)$ with the Fourier transform of a triangular window in direct space given by $w(r) = (3/\pi r_c^3)(1-r/r_c)(1-\Theta(r-r_c))$.  

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The Fourier transform of this radial function is

\[
\tilde{w}(k) = \begin{cases} 
\frac{12}{(kr_c)^4} (2 - 2\cos(kr_c) - kr_c\sin(kr_c)), & k > 0 \\
1, & k = 0 
\end{cases} \tag{5.30}
\]

For a general three-dimensional, radial function \( f(\mathbf{r}) \equiv f(|\mathbf{r}|) \), the Fourier transform and its inverse may be expressed \([91]\) as

\[
\tilde{f}(k) = \begin{cases} 
\frac{4\pi}{k} \int_0^\infty r f(r) \sin(kr) dr, & k > 0 \\
\int_{\mathbb{R}^3} f(\mathbf{r}) d\mathbf{r}, & k = 0 
\end{cases} \tag{5.31}
\]

\[
f(\mathbf{r}) = \begin{cases} 
\frac{1}{2\pi^2 r} \int_0^\infty k \tilde{f}(k) \sin(kr) dk, & r > 0 \\
\frac{1}{(2\pi)^3} \int_{\mathbb{R}^3} \tilde{f}(k) dk, & r = 0 
\end{cases} \tag{5.32}
\]

Note that both the forward and inverse transforms are equivalent up to their scaling coefficients. To take advantage of the usual one-dimensional fast Fourier transform algorithm for our three-dimensional, radial \( \tilde{c}(k) \), we take our discrete \( C_n = \tilde{c}(n\Delta k) \) for \( n = 0, 1, 2, \ldots, l \) and compute

\[
C'_n = \begin{cases} 
(n-l)\Delta k C_{l-n-1}, & n = 0, 1, \ldots, l \\
0, & n = l + 1 \\
(n-l-1)\Delta k C_{n-l-1}, & n = l + 2, \ldots, 2l + 1 
\end{cases} \tag{5.33}
\]

We then compute the usual one-dimensional inverse FFT on this data, defined here as

\[
c'_m = \mathcal{F}^{-1}[C'_n, m] = \frac{\Delta k}{2l + 1} \sum_{n=0}^{2l+1} C'_n e^{2\pi i nm/(2l+1)} \tag{5.34}
\]
where $m = 1, \ldots, 2l + 1$. We then apply the prefactor and a phase correction to account for the fact that the index $n = l + 1$ corresponds to $k = 0$ to obtain

$$c_m = \frac{c'_m}{2\pi^2 r_m} e^{-\frac{2\pi iml}{2l+1}}$$

(5.35)

where $r_m = m\Delta r$ and $\Delta r = 2\pi/\Delta k(2l + 1)$. Through analogy with Eq. (5.32), one expects that $c_m$ is completely imaginary, while one expects the Fourier transform to be completely real-valued. This is reconciled by dropping the imaginary unit from $c_m$; doing so is justified since the imaginary prefactor is expected if one applies Euler’s formula to the exponential term in Eq. (5.34), but is missing as a prefactor to the sine term in Eq. (5.32). Finally, the value for $c_0$ corresponding to $c(r = 0)$ is obtained by integrating $C_n$ numerically according to Eq. (5.32).

Once the direct space $c(r)$ has been found (represented discretely through $c_m$), one must then be sure to truncate the data at $r_{max} = L_{max}/2$ where $L_{max}$ is the width of the simulation box; data beyond this point is subject to finite-size artifacts.
Chapter 6

Static Structural Signatures of Nearly Jammed Disordered and Ordered Hard-Sphere Packings: II. Force Percolation

6.1 Introduction

In the previous Chapter, we studied the structural evolution of packings of hard spheres as they approached ordered (i.e. FCC crystal) and disordered jammed (i.e. mechanically stable [16]) states, looking to the direct correlation function \( c(r) \) so as to determine static signatures that indicated the presence of an incipient jammed backbone. In this Chapter, we probe the structures of the same packings using both techniques of geometric percolation [134, 14, 135, 136, 137, 138] as well as force percolation [139] in an attempt to identify indications of their incipient structure prior to jamming. In the former problem, “bonds” between particles are formed if the pair of particles are separated by less than some threshold distance; equivalently,
one may decorate the hard spheres with a perfectly penetrable outer layer of thickness $t$ (the “cherrypit” model) and look at the percolation of overlapping particles [135]. Thus, the connectivity associated with geometric percolation problem is related to the proximity of the particles to each other. In the latter, bonds are counted when particles share an interparticle pair force whose magnitude exceeds some minimum threshold force $f_{\text{min}}$. Thus, the connectivity associated with force percolation is connected to a a mechanical aspect of the packing. In both cases, percolation is said to have occurred when an infinite cluster appears. Force percolation, as we define it in this Chapter, should not be confused with so-called “central-force percolation” or “rigidity percolation” as studied elsewhere [140, 141, 142]. The elimination of a single pair contact in a jammed, isostatic system of hard particles is sufficient to destroy the rigidity of the entire system [143], making rigidity percolation ill-suited to address jamming in the hard-particle systems that we consider here.

It is known that compressing packings of hard particles towards jammed states is accompanied by an anomalous suppression of density fluctuations on large length scales consistent with hyperuniformity [17, 56, 57, 58, 107, 47]. Mathematically speaking, this means that the structure factor $S(k)$ of the packings tends towards zero in the limit that the wave number $k = |k|$ approaches zero [17]. Importantly, hyperuniformity may be viewed as a sort of “inverted critical point” in which $c(r)$, rather than the total correlation function $h(r)$, becomes long-ranged [17]. Thus, it is natural to ask if percolation, which exhibits critical behavior similar to ordinary thermal critical points [144], might also yield insight regarding packings’ compression trajectories towards jammed states.

Additionally, we noticed in Chapter 5 that the fluctuating component of the structure factor $S(k)$ for equilibrated packings along the equilibrium crystal (FCC) branch exhibited a quadratic scaling behavior in the wave number $k$ over an increasingly large range as jamming was approached. By decomposing the structure factor of the disor-
dered packings into a static and a fluctuating component, a similar underlying trend was identified, consistent with the results found in [59]. As mentioned there, this suggests that, the displacements of the particles from their “frozen-in”, jammed positions in these packings might be, to some degree, uncorrelated. Here, we ask to what extent this is the case.

In this Chapter, we utilize geometric and force percolation techniques to search for structural precursors to jamming. Using the Lubachevsky-Stillinger (LS) and Torquato-Jiao (TJ) hard-sphere packing algorithms, we compress packings towards disordered jammed states in order to gain insight about the maximally random jammed (MRJ) state [45]. We also consider the equilibrium branch where packings are compressed towards the FCC crystal [48, 119]. For geometric percolation, we measure the critical shell thickness \( t_c \) as a function of the hard-sphere packing density \( \phi_{HS} = Nv_1(D)/V \), where \( N \) is the number of spheres (with diameter \( D \) and volume \( v_1(D) \)) inside a fundamental cell of volume \( V \) with periodic boundary conditions. We also monitor the coordination of the percolating cluster at the percolation threshold as a means of obtaining rough information regarding its structure so as to compare the different packing protocols that we use. Additionally, we measure the two-point cluster function \( C_2 \) at the percolation threshold, which is known to exhibit power-law scaling of the form \( C_2(r) \propto r^{2-d-\eta} \), where \( \eta \) is a critical exponent. For nearly-jammed packings, we determine the cluster size distribution and compute the mean cluster size \( S \) as a function of \( t_c \) so as to obtain critical exponents for our systems to determine their universality classes.

For force percolation, we are limited to studying metastable packings in which a force network persists over time scales long enough to resolve the pair forces in the packing. As with geometric percolation, we measure \( S \) as a function of \( f_{\text{min}} \) and obtain critical exponents for the case of force percolation. We also look at \( C_2 \) using force percolation data and compare the data with that of geometric percolation so as
to discern differences in the structure of our jammed packings as presented by these two problems.

Our results show that geometric percolation can be used to detect structural signatures of the incipient jammed structure. The packing fractions at which these signatures can be observed are in rough agreement with the points during the packing process that the signatures discussed in Chapter 5 appear, adding to the growing evidence suggesting that an incipient contact network might be discernible prior to jamming. Furthermore, we find that geometric percolation and force percolation occupy different universality classes, mirroring the story in two dimensions [139].

The remainder of this Chapter is organized out as follows: in Sec. 6.2, we review the protocols that we used to generate nearly-jammed hard-sphere packings. In Sec. 6.3, we present the results of our percolation analysis. Conclusions and discussion are presented in Sec. 6.4.

6.2 Methods

To study the behavior of the equilibrium hard-sphere FCC crystal for densities between $\phi = 0.55$ and the close-packing density $\phi_{FCC} = \pi / \sqrt{18}$, we used standard event-driven molecular dynamics [55], following the procedure outlined in Chapter 5. We also use the Lubachevsky-Stillinger (LS) event-driven molecular dynamics and Torquato-Jiao (TJ) sequential linear programming algorithms to study two different ways of approaching highly disordered, MRJ-like states. We use the same densification schedules as those used in Chapter 5. All of our simulations are carried out in a cubic simulation box with periodic boundary conditions.

For geometric percolation, a perfectly penetrable shell of thickness $t$ is added to the hard sphere configurations. Bonds are drawn between hard spheres whose penetrable shells overlap. This is analogous to the “cherriypit” model [135, 91] in which an
impenetrability parameter $\lambda$ describes the ratio of the diameters of the penetrable shell and hard core. Specifically, $\lambda = D / (D + 2t)$. For any given packing, percolation occurs at a critical shell thickness $t_c$ where a system-spanning cluster first appears.

To study force percolation, a packing is equilibrated using molecular dynamics for a set number of collisions per sphere. During this run, pair collisions and their associated momentum exchanges are tracked. From this information, the time-averaged pair force $f_{ij}$ on particle $i$ due to collisions with particle $j$ may be computed as

$$f_{ij} = \frac{1}{t_{sim}} \sum_k p_{ij,k}, \quad (6.1)$$

where $p_{ij,k}$ is the change in momentum in particle $i$ due to its $k$-th collision with particle $j$, and $t_{sim}$ is the time duration of the simulation. By convention, we normalize the forces in a packing with respect to the mean force. Bonds are drawn between pairs of particles whose pair force is greater than some threshold force; the percolation threshold occurs once the threshold has been decreased low enough to give rise to a system-spanning cluster. It is necessary that the packing not undergo any collective rearrangements while the pair forces are being measured. Thus, we only examine force percolation for packings which are close to their putative jammed states.

To test for a system-spanning cluster, we create periodic images of the simulation box in all directions, yielding a system of $3^d N$ spheres. We then enumerate all of the clusters that are formed according to the given percolation problem and threshold. If a cluster contains both a sphere $i$ as well as one of $i$’s periodic images, then it is a system-spanning cluster. Furthermore, we are able to determine the direction in which the cluster is system-spanning. Generally, we find that the largest cluster will span the system in a single direction first then subsequently span it in other directions as the threshold is further adjusted to recognize more bonds. The percolation threshold is defined as the threshold value at which a cluster first spans the system in at least...
one direction. Figures 6.1 and 6.2 show an illustrative example of force percolation in two dimensions with a disordered packing of bidisperse disks with size ratio $\alpha = 1.4$ and number ratio $x = 0.5$. In Fig. 6.1, all interacting pairs are connected with a line; the thickness and color denote the relative magnitude of the pair forces across a continuum: the weakest forces are denoted with thin, blue lines; and increasingly strong forces are indicated with lines that are increasingly thick and red. The visible voids are occupied by rattlers, which have a vanishingly small pair force with their enclosing cage relative to the forces present in the backbone as jamming is approached. Figure 6.2 shows the effect of thresholding near the percolation threshold such that weak pair forces are eliminated. Figure 6.3 shows geometric percolation for this same system; lines are drawn between pairs with overlapping penetrable shells; here, the thickness $t$ of the shells was chosen so that the system is close to its geometric percolation threshold. Note that the circles denote the disk centers and do not reflect the diameter of the disks. We emphasize that we have only chosen to use two-dimensional systems here for the purposes of illustration; by contrast the ensuing analysis is carried out on three-dimensional packings of equal-sized spheres.

6.3 Results

In this section, we discuss the results regarding the application of both the geometric and force percolation percolation problems to our packings in an effort to identify signs of an incipient structural network. We begin by focusing on the former, then continue with the latter.

Figure 6.4 shows the critical shell thickness $t_c$ as a function of $\phi_{HS}$ for the FCC equilibrium crystal and disordered packings compressed using the LS and TJ algorithms towards MRJ-like states. We used a system size of $N = 2048$ for the equilibrium packings so that the FCC crystal is commensurate with the cubic simulation
Figure 6.1: The force network for a putatively jammed system of $N = 1000$ bidisperse disks with size ratio $\alpha = 1.4$ and number ratio $x = 0.50$. Nodes denote disk centers and do not reflect the actual size of the disks, and edges denote contact pairs that transmit a force. The color and thickness of the lines interpolate from weak forces (thin, blue lines) through strong forces (thick, red lines).

box. For the LS and TJ protocols, we used a system size of $N = 2000$. For the equilibrium crystal branch as well as the LS packings at sufficiently low distance to jamming, we see that the data fall into a simple trend in which $t_c$ scales linearly with the distance to jamming in a manner analogous to the free-volume equation of state [124]. Namely, for our data, we have

$$t_c = A(1 - \phi_{HS}/\phi_c),$$

(6.2)

where $A = 0.092$ is a proportionality constant. This trend is shown as a black dashed line in Fig. 6.4. We have confirmed that this behavior does not change appreciably
upon increasing the system size to $N = 10^4$. Interestingly, we find that $A$ is consistent across ensembles and does not depend upon whether the system is ordered or disordered. Thus, we suggest that the observation of this trend is evidence for the existence of a packing’s incipient backbone prior to jamming. For the FCC crystal, the incipient jammed structure may be inferred along the entire crystal branch, as we would expect. For the LS protocol, evidence of the incipient jammed structure may be inferred from our data starting at a relatively small jamming gap.

For the TJ protocol, $t_c$ quickly approaches zero and remains less than that of the packings produced by LS at an equivalent distance to jamming. This may be explained by noting that the TJ algorithm explores configuration space in a substantially different way from molecular dynamics, as we described in Chapter 5. Because an optimal solution to the LP at any given step does not require substantial movement of the spheres, the TJ algorithm does not tend to displace spheres far from contacting. If a packing compressed by TJ to a sufficiently high density is subsequently
Figure 6.3: The same packing as in Fig. 6.1, analyzed through geometric percolation with $t = 5.0 \times 10^{-9}$. The reduced pressure is $P = 10^8$. Nodes denote disk centers and do not reflect the actual size of the disks, and edges connect overlapping pairs of disks. Again, the color and thickness of the lines correspond to the pair separation between the disks: smaller pair separations are indicated with thick, red lines, and greater pair separations are indicated with thin, blue lines.

provided as an initial condition for the LS algorithm, then it will assume the linear trend seen in the other procedures. Before the measurement at the smallest distance to jamming, the TJ packings are equilibrated briefly through molecular dynamics, and we see that this is indeed the case.

Figure 6.5 shows the mean coordination of the packings at their percolation threshold when compressed using the three protocols as a function of distance to jamming. Again, we see that, at sufficiently small distances to jamming, the FCC and LS packings converge to a constant mean coordination, supplying evidence of an incipient
jammed structure. By contrast, TJ again displays qualitatively different behavior. For packing fractions below $\phi_{HS} = 0.55$, the percolating cluster remains sparsely coordinated. Beyond this, we see a dramatic increase in coordination which is sustained until the final step (leftmost point on the curve) in which the packing is equilibrated using molecular dynamics. We noticed that rapid changes in the packings’ structure factor and direct correlation function seem to occur at a similar point in the compression process, as noted previously in Chapter 5.

To obtain a more nuanced picture of the structure of the packings that we observe, we turn to the two-point cluster function $C_2(r)$, which describes that probability that a pair of randomly-placed points will fall in the same cluster of connected particles. Figure 6.6 shows the radially-averaged $C_2(r)$ for ensembles of packings created by LS at various hard-sphere packing fractions at their percolation threshold. At the percolation threshold, the $C_2(r)$ will exhibit a power-law decay at large values of $r$: $C_2 \propto r^{2-d-\eta}$, where $\eta$ is a critical exponent. When viewed on a log-log plot, we find that the power-law decay exhibited by the ensemble seems to possess the same expo-
Figure 6.5: The average coordination $z$ of the particles in the percolating cluster as a function of the hard-core packing fraction $\phi_{HS}$ for the FCC equilibrium crystal and disordered packings compressed using the LS and TJ algorithms at their respective percolation thresholds for geometric percolation. Error bars correspond to a 95% confidence interval.

Densification across all of the packing densities, suggesting that the universality class of this percolation problem does not change as jamming is approached. However, the prefactor associated with the power-law decay changes as a function of packing fraction. In particular, we notice that its maximum among the packing fractions that we considered occurs at $\phi = 0.60$, then falls abruptly upon further compression, converging to a steady value. This indicates that the structure of the packing is exhibiting signs of an incipient backbone and that further compression will not substantially alter the structure that is being approached.

Figure 6.7 shows the same analysis performed on packings created with the TJ algorithm. In stark contrast to the trend shown with the LS-generated packings, the prefactor decreases up to $\phi = 0.55$, then proceeds to grow quickly as jamming is approached, suggesting a substantial qualitative change in the structure of the packings. We suggest that the densification process that TJ undergoes as its packings approach
MRJ-like states may be broken down into three qualitatively distinct “stages.” First, at low densities, spheres are mostly undisturbed as the packing fraction increases and spheres come into close proximity to form localized clusters; these clusters then merge with one another and a system-spanning “cluster” composed of particles in close proximity emerges. Second, at intermediate densities, substantial collective rearrangements start to become necessary in order to allow for further densification, at which point the packing begins to fill the simulation box more uniformly. Finally, the packing is in the vicinity of its jammed state, and further densification is generally accompanied by minimal macroscopic rearrangements. Our data imply that a transition from the first to the second stage tends to happen between $\phi_{HS} = 0.55$ and 0.60 and the second transition happens between $\phi_{HS} = 0.60$ and $1 - \phi_{HS}/\phi_c = 10^{-3}$. 

Figure 6.6: The two-point cluster function $C_2(r)$ for packings produced by the LS algorithm at various hard-core packing fractions $\phi_{HS}$ and their percolation thresholds.
At sufficiently high pressures, we are able to equilibrate the packings using molecular dynamics without observing any significant rearrangements, allowing us to measure the average interparticle forces in the incipient jammed structures. This allows us to define a critical threshold force $f_{\text{min}}$ that describes the force percolation threshold. Figure 6.8 shows $C_2(r)$ for both geometric and force percolation for ensembles of packings generated by the LS algorithm at a reduced pressure of $P = 10^8$ and system sizes $N = 10^2 - 10^4$. We observe a system size dependence for the critical exponent $\eta$, reflected in the data as a different slope for each system size. Interestingly, we notice that the limiting behavior that the geometric and force percolation problems approach does not seem to be the same, implying that geometric and force percolation are not in the same universality class.
To more accurately address this possibility, we perform an analysis of the mean cluster to determine more information about the critical exponents for this system. First, we compute the mean cluster size

\[ S = \frac{\sum_k k^2 n_k}{\sum_k k n_k} \]  

(6.3)

as a function of \( t_c \) and \( f_{\min, c} \) for geometric and force percolation, respectively, where \( n_k \) is the number of clusters of size \( k \).

Following the standard procedure, we plot the logarithm of \( S \) as a function of the logarithm of \( N \) and note that the data follow a linear trend; the slope of this line is equal to \( \gamma / d \nu \), giving us the ratio between these critical exponents. The results of
this analysis are shown in Fig. 6.9 for geometric percolation applied to LS-generated packings both far from jamming ($\phi_{HS} = 0.40$) and near jamming ($P = 10^8$), TJ-generated packings near jamming ($P = 10^8$); and force percolation applied to LS- and TJ-generated packings near jamming. Table 6.1 lists the values measured from the analysis of Fig. 6.9.

![Log-log plots of the mean cluster size $S$ taken at the percolation threshold for various systems for geometric and force percolation. Error bars correspond to a 95% confidence interval, computed using the Clopper-Pearson method [8]. Lines show least-squares fits to the data.](image)

Figure 6.9: Log-log plots of the mean cluster size $S$ taken at the percolation threshold for various systems for geometric and force percolation. Error bars correspond to a 95% confidence interval, computed using the Clopper-Pearson method [8]. Lines show least-squares fits to the data.

Our data imply that the various systems that we studied using geometric percolation seem to be in the same universality class; by contrast, the critical exponents computed for LS packings under force percolation are substantially different, implying
Table 6.1: Critical exponents measured for various protocols for both geometric and force percolation using system sizes $N_{\text{min}} \leq N \leq N_{\text{max}}$. Uncertainties reported correspond to a 95% confidence interval.

<table>
<thead>
<tr>
<th>System</th>
<th>$N_{\text{min}}$</th>
<th>$N_{\text{max}}$</th>
<th>$\gamma/d\nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Poisson spheres, geometric percolation</td>
<td>1000</td>
<td>10000</td>
<td>0.701 ± 2.0 × 10^{-2}</td>
</tr>
<tr>
<td>FCC, geometric percolation, $\phi_{\text{FCC}} - \phi = 10^{-4}$</td>
<td>864</td>
<td>10976</td>
<td>0.684 ± 1.5 × 10^{-2}</td>
</tr>
<tr>
<td>LS, geometric percolation, $\phi = 0.40$</td>
<td>1000</td>
<td>10000</td>
<td>0.675 ± 2.3 × 10^{-2}</td>
</tr>
<tr>
<td>LS, geometric percolation, $P = 10^8$</td>
<td>1000</td>
<td>10000</td>
<td>0.663 ± 2.5 × 10^{-2}</td>
</tr>
<tr>
<td>LS, force percolation, $P = 10^8$</td>
<td>1000</td>
<td>10000</td>
<td>0.641 ± 2.5 × 10^{-2}</td>
</tr>
</tbody>
</table>

that force percolation is likely part of a different universality class. The rationale that geometric and force percolation occupy different universality classes is that there are significant spatial correlations between pair forces in jammed packings, manifested in the so-called “force chains” that are known to exist in marginally stable packings [145].

It has been proposed [135] that geometric percolation of randomly-overlapping particles is in the same universality class as ordinary lattice percolation. The rationale behind this claim is that correlations between bonds in the former case are short-ranged and thus do not cause the large-system behavior to differ from that of lattice percolation, where bonds are removed at random. To quantify this claim, we have defined a correlation function which measures spatial correlations between bonds in the two percolation problems. For a pair of equal-sized spheres $i$ and $j$ located at $r_i$ and $r_j$, respectively, that undergo pair collisions over some sufficiently long interval in time (i.e., having a positive mean interparticle force), we assign a position to their bond as $r_{b,ij} = (r_i + r_j)/2$. We may define a “bond pair correlation function” in a manner analogous to the standard pair correlation function

$$g_{2,\text{bond}}(r_1, r_2) = \rho_{\text{bond}}^{-2} \frac{N_{\text{bond}}}{(N_{\text{bond}} - 2)!} \int P_{N_{\text{bond}}}(r^{N_{\text{bond}}}) dr^{N_{\text{bond}}-2}, \quad (6.4)$$

where $\rho_{\text{bond}} = N_{\text{bond}}/V$ is the bond number density and $P_N(r^N)$ is the probability of
observing a set of points (here, the positions assigned to the bonds as described above) positioned at \( r^N = (r_1, \ldots, r_N) \). After thresholding according to either \( t \) or \( f_{\text{min}} \), some number of bonds \( N'_{\text{bond}} \leq N_{\text{bond}} \) remain. We can then compute \( g'_{2,\text{bond}}(r_1, r_2) \) in an analogous manner on the remaining bonds. In all cases, we assume translational invariance and express these as a function of \( r = r_2 - r_1 \). Finally, we define

\[
H(r)dr = \frac{g'_{2,\text{bond}}(r)}{g_{2,\text{bond}}(r)} - 1. \tag{6.5}
\]

The case in which \( H(r) = 0 \) corresponds to random (spatially uncorrelated) removals of bonds; this is realized for ordinary lattice bond percolation. Thus, by examining \( H(r) \), we can examine the spatial correlations in the bonds and quantitatively assess the premise of bond correlations. Note that \( H(r) \) is only defined at values of \( r \) where \( g_{2,\text{bond}}(r) \) is nonzero.

Figure 6.10 shows the radially-averaged \( H(r) \) for the cases of geometric percolation on both the FCC crystal at a density of \( \phi_{\text{HS}} = \phi_{\text{FCC}} - 10^{-10} \) and nearly-jammed MRJ-like packings produced using the LS and TJ algorithms, as well as force percolation for the LS and TJ ensembles. We also include data for the case in which small random, uncorrelated displacements are applied to the FCC lattice from its perfect crystal configuration so as to evaluate the claim made in Chapter 5. Threshold values are chosen in all cases so that we are at the percolation threshold.

As expected, the force percolation problem exhibits markedly larger spatial correlations than the geometric percolation problem, and the case of uncorrelated displacements for the FCC lattice displays no correlations beyond a pair distance of \( r/D = 1 \), as expected. We expect spatial correlations inside of this distance because displacing a sphere from its mean position towards one of its nearest neighbors moves it away from any spheres in the opposite direction.
Figure 6.10: Bond correlation function measured for a variety of systems under both geometric and force percolation.

### 6.4 Conclusions and Discussion

In this Chapter, we applied the problems of geometric and force percolation to three-dimensional packings of monodisperse hard spheres in an effort to obtain structural indicators of an incipient rigid backbone as the packings were compressed toward ordered (FCC) and disordered (MRJ-like) jammed states. For geometric percolation, we measured the critical thickness $t$ of the perfectly-penetrable shell as a function of the hard-sphere packing fraction, and found asymptotic behavior analogous to the free-volume equation of state in the vicinity of jamming. Like the free-volume equation of state, the asymptotic behavior we observed does not change depending upon whether the system is ordered or disordered. We also measured the mean coordi-
nation of the system-spanning cluster at the percolation threshold as the systems approached jamming and found that this converges to a constant that depends upon the structural details of the packing. The two-point cluster function $C_2(r)$ revealed power-law scaling at the percolation threshold for all of the configurations that we studied; for disordered packings created using both TJ and LS, the prefactor for this scaling relation was found to change as the packings were compressed toward jamming, eventually converging towards a constant at a distance to jamming that is consistent with the previous metrics. To determine the critical exponents associated with our systems’ universality classes, we measured the mean cluster size at the percolation threshold and determined the exponents that characterize its system size dependence. Lastly, we devised a correlation function that allowed us to study the spatial correlations of the bonds in geometric and force percolation, and found that the spatial correlations are markedly more pronounced for force percolation, providing a rationale for why geometric and force percolation might occupy separate universality classes.

The concept of an incipient jammed structure is important because it implies that one might obtain useful information regarding the terminal state that a packing protocol is approaching. Here, we have shown evidence suggesting that this regime of incipient rigidity in hard-sphere packings may extend to packing fractions on the order of $10^{-2}$ lower than that of the jamming point. Thus, one might glean important information about the terminal structure even when one is far away from it. However, this does not rule out the possibility that one may continuously displace a configuration so as to approach a different terminal state at sufficiently high distances to jamming. Thus, the dynamics of how a configuration might “hop” among the neighborhoods of various jammed states is a subject of ongoing interest.

It is interesting to note that, when a configuration is trapped in the neighborhood of a jammed configuration, the problems of geometric and force percolation
may be brought together by noticing that the measurement of pair forces involves a time average that “averages out” positional fluctuations that are fully present in geometric percolation. When a system is driven by molecular dynamics (as is the case for LS, but not TJ), one might expect that particles will “vibrate” about their jammed positions. When one considers a snapshot of a configuration for geometric percolation, one captures information about both the “frozen-in” configuration as well as instantaneous fluctuations about that configuration. By contrast, when one measures average pair forces over some interval in time, one might seek to have that interval to be as large as is practically possible so as to obtain reliable measurements for the pair forces in the packing. Similarly, one might also think of performing a time average of the positions that the spheres occupy as they vibrate around their frozen-in positions. If one were to study the geometric percolation properties of this time-averaged configuration, one might reasonably expect to obtain results that are analogous to the force percolation problem. We note that this would be consistent with the finding that the time-averaged pair force can be related to the average pair separation of interacting particles [66, 126]. Thus, one might think of the length of the interval of time as a “tuning parameter” that can take one continuously from geometric percolation to force percolation. It remains to be seen how the universality class of the problem might change as the time interval is continuously increased.

While our investigations of geometric and force percolation are carried out on nearly-jammed packings, it is worth noting that the point of exact jamming represents a “singular” point at which all pair separations become exactly zero. For geometric percolation, the approach of defining some threshold that identifies some but not all nearest neighbor pairs breaks down, and one sees a discontinuous jump when crossing the percolation threshold, making it impossible to probe from critical behavior. It is unclear how one might go about treating such a limit case theoretically. However, since not all pair contacts in a packing are necessarily force-bearing, the appearance of
such pathological behavior under force percolation (i.e., in which all pair forces diverge towards infinity upon jamming) may depend upon the structure of the packing (e.g., whether it is isostatic or not). Thus, one must bear in mind that geometric and force percolation possess this important distinction that may have interesting consequences when dealing with systems that are not isostatic such as the FCC crystal.

Finally, while our study has been performed using insights obtained from the LS and TJ algorithms, we point out that there are still many other known packing protocols that might display different behavior upon approaching jammed states. It has been pointed out previously that different protocols can have a marked effect on the sort of configurations that one might observe [45, 62, 3, 109]. In a similar way, we point out here that one might expect to find differences in the way in which various protocols approach jamming. Our work suggests that percolation problems might serve as a useful frame of reference for understanding these differences.
Chapter 7

Conclusions and Future Work

In this dissertation, we investigated the structure of disordered packings that represent the putative maximally random jammed (MRJ) state. In doing so we have uncovered insights regarding the role of numerical packing protocols in informing our perception of the MRJ state and uncovered interesting avenues along which one might extend our investigations.

In Chapter 2, we used the Torquato-Jiao (TJ) linear programming algorithm to produce thousands of strictly jammed, exactly isostatic packings of monodisperse spheres of high numerical fidelity. We found that the TJ algorithm is coming closer to the true MRJ state than ever before, as shown by standard order metrics. We also found differences in other macroscopic packing properties such as packing fraction and rattler fraction compared to previous estimates of the MRJ state. The probability distribution of rattler fractions tends towards an infinite-system limit value of $\lim_{N \to \infty} N_R/N = 0.015$, which differs significantly from previous estimates for the MRJ state. The qualitative difference between TJ and LS packings points out an ambiguity which causes one to call into question whether or not the standard protocols have been truly producing the MRJ state, as has been taken for granted.

In Chapter 3, we used the TJ algorithm to demonstrate by construction the
existence of the isostatic MRJ state in two dimensions for packings of equal-sized hard-disks by producing collectively and strictly jammed packings of monodisperse disks that are exactly isostatic with system sizes up to \( N = 150 \) and 100, respectively. These isostatic packings are hyperuniform, have an average packing fraction of \( \phi = 0.826 \), and, again, are significantly more disordered than the polycrystalline packings generated by typical protocols. In fact, these packings are candidates for the MRJ state according to bond-orientational and translational order metrics. Other standard structural descriptors suggest further similarities between these packings and the MRJ state in 3D. It is critical to point out that the 2D MRJ packings that we have shown are not entropically favored. This is problematic for a picture of RCP that relies on one’s choice of protocol as well as an ensemble-based, entropic definition. Demonstrating the MRJ state for monodisperse disks by construction is important conceptually in a similar way to the discovery of the reinforced kagomé lattice in that both are extremal packings that are very interesting though hard to observe via traditional packing protocols. This reflects a persisting fundamental lack of knowledge that we still have with regards to designing packing protocols.

In Chapter 4, we introduced a method that uses sequential linear programming to test for jamming in packings of frictionless hard-spheres and found that standard protocols struggle to create packings that are truly confined to a jamming basin for what might be regarded as modest system sizes by current standards. Importantly, heuristic tests like the pressure test fail to find these unjamming motions. We then examined the spectral density of the packings we generated for a variety of system sizes and found evidence that our inability to observe exactly-hyperuniform configurations at larger system sizes is directly linked to the difficulty of producing exactly-jammed configurations. We also found that as packings are brought closer to their jamming transition point, the degree of hyperuniformity increases correspondingly, further supporting the notion that an absence of exact hyperuniformity is linked to an absence.
of exact jamming.

In Chapter 5, we compared and contrasted the approach of both ordered and disordered hard-sphere packings towards jammed states through considering the behavior of their structure factors and direct correlation functions. We found that the LS and TJ protocols approach their respective jammed states in markedly different manners, as shown by various pair statistics. Specifically, the structure factor of TJ-generated packings shows anomalously increasing behavior for small wave numbers at intermediate densities, and generally remains closer to unity for low and intermediate wave numbers at all packing fractions leading up to jamming when compared to LS, suggesting that the configurations that TJ visits in approaching its jammed states are substantially more disordered. Additionally, TJ shows signs of particles in close proximity at surprisingly low densities as evidenced by the appearance of a clear precursor to the delta function in the direct correlation function at a distance of one sphere diameter and corresponding $-1/r$ scaling within the core. We have noted that these features in the intermediate-density structures produced by the TJ algorithm suggest that it possesses important qualitative commonalities with the physical process of compressing hard-spheres embedded in a dampening background, providing a conceptual physical analog to the algorithm as witnessed in practice.

In Chapter 6, we applied the problems of geometric and force percolation to three-dimensional packings of monodisperse hard spheres in an effort to obtain structural indicators of an incipient rigid backbone as the packings were compressed toward ordered (FCC) and disordered (MRJ-like) jammed states. For geometric percolation, we measured the critical thickness $t$ of the perfectly-penetrable shell as a function of the hard-sphere packing fraction, and found asymptotic behavior analogous to the free-volume equation of state in the vicinity of jamming. Like the free-volume equation of state, the asymptotic behavior we observed does not change depending upon whether the system is ordered or disordered. We also measured the mean coordi-
nation of the system-spanning cluster at the percolation threshold as the systems approached jamming and found that this converges to a constant that depends upon the structural details of the packing, reinforcing the observations made in Chapter 5 that the LS and TJ protocols possess striking qualitative differences as they explore configuration space on their way towards producing MRJ-like states. The two-point cluster function \( C_2(\mathbf{r}) \) revealed power-law scaling at the percolation threshold for all of the configurations that we studied; for disordered packings created using both the TJ and LS protocols, the prefactor for this scaling relation was found to change as the packings were compressed toward jamming, eventually converging towards a constant at a distance to jamming that is consistent with the previous metrics. To determine the critical exponents associated with our systems’ universality classes, we measured the mean cluster size at the percolation threshold and determined the exponents that characterize its system size dependence. Lastly, we devised a correlation function that allowed us to study the spatial correlations of the bonds in geometric and force percolation, and found that the spatial correlations are markedly more pronounced for force percolation, providing a rationale for why geometric and force percolation might occupy separate universality classes.

Despite the progress that we have made in this dissertation, a number of fascinating questions remain. Here, we review some of the questions that are of particular importance and the answers to which may be within our grasp in the near future.

One outstanding question concerns the physical origin of the linear nonanalytic behavior of the structure factor in MRJ hard-sphere packings. While some hypotheses have been offered [57], this remains a fascinating question, the answer to which is guaranteed to provide profound insight with regards to the macroscopic organizing principles behind MRJ-like packings. At the same time, offering such an explanation is a daunting task because it seems tantamount to explaining the origins of the MRJ state itself.
Another question intimately related to the MRJ state concerns the degree to which one can expect to find these states in physical experiments. While numerical simulations offer an attractive setting for exploring these systems under ideal conditions, the fact remains that perfectly monodisperse, frictionless spheres, jammed in precisely the same way as we have come to expect when conducting detailed investigations into the nature of the MRJ state, cannot be found in real life. Thus, a critical question of practical importance is to what degree the properties of the MRJ state persist in experiments. The first part of answering this question is to explore to what degree structural signatures such as hyperuniformity persist; one cannot reasonably expect to observe perfectly hyperuniform systems in real life, though systems displaying considerable closeness have been readily observed [146, 108, 115], suggesting that near-hyperuniformity is a concept that is highly relevant and warrants attention.

Another matter that has shaped our understanding of jamming has been the necessary role of protocol dependence, whether one conducts numerical or experiments investigations. The recent past has seen our understanding of disorder and jamming evolve substantially, from the discovery that random close packing is ill-defined [45] to the introduction of a multitude of packing protocols, all which seem to give slightly-differing pictures regarding the variety of disordered jammed packings that exist. It should be clear that we do not expect that our journey is over in this regard. In this dissertation, we have presented a diversity of disordered packings, many of which were unexpected, and some of which were simply thought not to exist. We have asked if one might be able to devise algorithms that produce jammed packings with different macroscopic properties, but, as we have noted, our understanding of packing algorithms and how their dynamics are related to their final jammed states remains crude. While we have some intuition over how the packing fraction might be altered (e.g. by tuning the expansion rate in the LS algorithm), we are aware that our intuition has limits; for example, we cannot use these algorithms to observe
low-density jammed states such as tunneled crystals, which are nonetheless known
to exist and can be constructed in experiments [2]. We also have very little idea
how to deliberately control the rattler fraction in a predictable way apart from our
knowledge of the statistical behavior of our current protocols; the underlying physical
reasons for such behavior remain elusive.

The pop test introduced in Chapter 4 shows promise as a tool to broach some of
these questions of protocol dependence. While we have already demonstrated that
it may be used to identify unjamming motions that are difficult to find otherwise,
one might reasonably believe that this procedure may be adapted so as to become
a general “saddle traversal” method in which narrow bottlenecks in configuration
space might be identified and explored in an efficient manner. Such a method then
allows for the systematic exploration of potential jammed states, including those that
current standard protocols are not designed to encounter. In particular, one might
be able to use this procedure in combination with an optimization scheme such as a
Markov chain Monte Carlo method to target packings with unusual densities, rattler
fractions, or other macroscopic quantities of interest, as well as to establish a range
of intermediate structures that span these points of interest. The discovery of such
packings stands to increase our knowledge regarding the diversity of jammed states
that exist and to enhance our understanding of the interplay of these and other
macroscopic structural features.
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of binary disks that we consider in our work, i.e., \((\alpha, x, p) = (1.4, 0.4, 1)\), the aforementioned formula predicts \(\phi_{MRJ}\) very accurately and is in agreement with our simulations.


