New Conjectural Lower Bounds on the Optimal Density of Sphere Packings

S. Torquato and F. H. Stillinger

CONTENTS

Sphere packings in high dimensions interest mathematicians and physicists and have direct applications in communications theory. Remarkably, no one has been able to provide exponential improvement on a hundred-year-old lower bound on the maximal packing density due to Minkowski in d-dimensional Euclidean space $\mathbb{R}^d$. The asymptotic behavior of this bound is controlled by 2^{-d} in high dimensions. Using an optimization procedure that we introduced earlier [Torquato and Stillinger 02] and a conjecture concerning the existence of disordered sphere packings in $\mathbb{R}^d$, we obtain a conjectural lower bound on the density whose asymptotic behavior is controlled by $2^{-0.77865\ldots d}$, thus providing the putative exponential improvement of Minkowski's bound. The conjecture states that a hard-core nonnegative tempered distribution is a pair correlation function of a translationally invariant disordered sphere packing in $\mathbb{R}^d$ for asymptotically large d if and only if the Fourier transform of the autocovariance function is nonnegative. The conjecture is supported by two explicit analytically characterized disordered packings, numerical packing constructions in low dimensions, known necessary conditions that have relevance only in very low dimensions, and the fact that we can recover the forms of known rigorous lower bounds. A byproduct of our approach is an asymptotic conjectural lower bound on the average kissing number whose behavior is controlled by $2^{0.22134\ldots d}$, which is to be compared to the best known asymptotic lower bound on the individual kissing number of $2^{0.2075\ldots d}$. Interestingly, our optimization procedure is precisely the dual of a primal linear program devised by Cohn and Elkies [Cohn and Elkies 03] to obtain upper bounds on the density, and hence has implications for linear programming bounds. This connection proves that our density estimate can never exceed the Cohn–Elkies upper bound, regardless of the validity of our conjecture.

1. INTRODUCTION

A collection of congruent spheres in d-dimensional Euclidean space $\mathbb{R}^d$ is called a sphere packing P if no two of the spheres have an interior point in common. The *packing density* or simply density $\phi(P)$ of a sphere packing is

the fraction of space $\mathbb{R}^d$ covered by the spheres. We will call

$$\phi_{\max} = \sup_{P \subset \mathbb{R}^d} \phi(P)$$

the *maximal density*, where the supremum is taken over all packings in $\mathbb{R}^d$. The sphere-packing problem seeks to answer the following question: Among all packings of congruent spheres, what is the maximal packing density $\phi_{\max}$, i.e., largest fraction of $\mathbb{R}^d$ covered by the spheres, and what are the corresponding arrangements of the spheres [Rogers 64, Conway and Sloane 98]? The sphere-packing problem is of great fundamental and practical interest, and arises in a variety of contexts, including classical ground states of matter in low dimensions [Chaikin and Lubensky 95], the famous Kepler conjecture for $d = 3$ [Hales 05], error-correcting codes [Conway and Sloane 98], and spherical codes [Conway and Sloane 98].

For arbitrary d, the sphere-packing problem is notoriously difficult to solve. In the case of packings of congruent d-dimensional spheres, the exact solution is known for the first three space dimensions. For $d = 1$, the answer is trivial because the spheres tile the space so that $\phi_{\max} = 1$. In two dimensions, the optimal solution is the triangular lattice arrangement (also called the hexagonal packing) with $\phi_{\max} = \pi/\sqrt{12}$. In three dimensions, the Kepler conjecture that the face-centered cubic lattice arrangement provides the densest packing with $\phi_{\max} = \pi/\sqrt{18}$ was only recently proved by Hales [Hales 05]. For $3 < d < 10$, the densest known packings of congruent spheres are lattice packings (defined below). However, for sufficiently large d, lattice packings are likely not to be the densest. Indeed, this paper suggests that disordered sphere arrangements may be the densest packings as $d \to \infty$.

We review some basic definitions that we will subsequently employ. A packing is *saturated* if there is no space available to add another sphere without overlapping the existing particles. The set of lattice packings is a subset of the set of sphere packings in $\mathbb{R}^d$. A *lattice* Λ in $\mathbb{R}^d$ is a subgroup consisting of the integer linear combinations of vectors that constitute a basis for $\mathbb{R}^d$. A *lattice packing* P_L is one in which the centers of nonoverlapping spheres are located at the points of Λ. In a lattice packing, the space $\mathbb{R}^d$ can be geometrically divided into identical regions F called *fundamental cells*, each of which contains the center of just one sphere. Thus, the density of a lattice packing ϕ^L consisting of spheres of unit diameter is

given by

$$\phi^L = \frac{v_1(1/2)}{\text{Vol}(F)},$$

where

$$v_1(R) = \frac{\pi^{d/2}}{\Gamma(1 + d/2)} R^d \qquad (1\text{--}1)$$

is the volume of a d-dimensional sphere of radius R and $\text{Vol}(F)$ is the volume of a fundamental cell. We will call

$$\phi_{\max}^L = \sup_{P_L \subset \mathbb{R}^d} \phi(P_L)$$

the maximal density among all lattice packings in $\mathbb{R}^d$. For a general packing of spheres of unit diameter for which a density $\phi(P)$ exists, it is useful to introduce the *number* (or *center*) density ρ defined by

$$\rho = \frac{\phi(P)}{v_1(1/2)},$$

which therefore can be interpreted as the average number of sphere centers per unit volume.

Three distinct categories of packings have been distinguished, depending on their behavior with respect to nonoverlapping geometric constraints and/or externally imposed virtual displacements: *locally* jammed, *collectively* jammed, and *strictly* jammed [Torquato and Stillinger 01, Torquato et al. 03, Donev et al. 04]. Loosely speaking, these *jamming* categories, listed in order of increasing stringency, reflect the degree of mechanical stability of the packing. A packing is locally jammed if each particle in the system is locally trapped by its neighbors; i.e., it cannot be translated while the positions of all other particles are held fixed. Each sphere simply has to have at least $d + 1$ contacts with neighboring spheres, not all in the same d-dimensional hemisphere. A collectively jammed packing is any locally jammed configuration in which no finite subset of particles can simultaneously be continuously displaced so that its members move out of contact with one another and with the remainder set. A strictly jammed packing is any collectively jammed configuration that disallows all globally uniform volume-nonincreasing deformations of the system boundary. Importantly, the jamming category is generally dependent on the type of boundary conditions imposed (e.g., hard-wall or periodic boundary conditions) as well as the shape of the boundary. The range of possible densities for a given jamming category decreases with increasing stringency of the category. Whereas the lowest-density states of collectively and strictly jammed packings in two or three dimensions are currently unknown,

one can achieve locally jammed packings with vanishing density [Böröczky 64]. This classification of packings according to jamming criteria is closely related to the concepts of "rigid" and "stable" packings found in the mathematics literature [Connelly et al. 98].

In the next section, we summarize some previous upper and lower bounds on the maximal density. For large d, the asymptotic behavior of the well-known Minkowski lower bound [Minkowski 05] on the maximal density is controlled by 2^{-d}. Thus far, no one has been able to provide exponential improvement on this lower bound. Using an optimization procedure and a conjecture concerning the existence of disordered sphere packings in high dimensions, we obtain conjectural lower bounds that yield the long-sought asymptotic exponential improvement on Minkowski's bound. We believe that consideration of truly disordered packings is the key notion that will yield exponential improvement on Minkowski's lower bound. A byproduct of our approach is an asymptotic conjectural lower bound on the average kissing number that is superior to the best known asymptotic lower bound on the individual kissing number.

2. SOME PREVIOUS UPPER AND LOWER BOUNDS

The *nonconstructive* lower bounds of Minkowski [Minkowski 05] established the existence of reasonably dense lattice packings. He found that the maximal density $\phi_{\max}^L$ among all lattice packings for $d \geq 2$ satisfies

$$\phi_{\max}^L \geq \frac{\zeta(d)}{2^{d-1}}, \qquad (2\text{–}1)$$

where $\zeta(d) = \sum_{k=1}^{\infty} k^{-d}$ is the Riemann zeta function. Note that for large values of d, the asymptotic behavior of the Minkowski lower bound is controlled by 2^{-d}. Since 1905, many extensions and generalizations of equation (2–1) have been obtained [Davenport and Rogers 47, Ball 92, Conway and Sloane 98], but none of these investigations have been able to improve on the dominant exponential term 2^{-d}. It is useful to note that the density of a saturated packing of congruent spheres in $\mathbb{R}^d$ for all d satisfies

$$\phi \geq \frac{1}{2^d}. \qquad (2\text{–}2)$$

The proof is trivial. A saturated packing of congruent spheres of unit diameter and density ϕ in $\mathbb{R}^d$ has the property that each point in space lies within a unit distance from the center of some sphere. Thus, a covering of the space is achieved if each sphere center is encompassed by a sphere of unit radius and the density of this covering

$(2)2^{-d}$	Minkowski (1905)
$[\ln(\sqrt{2})d]2^{-d}$	Davenport and Rogers (1947)
$(2d)2^{-d}$	Ball (1992)

TABLE 1. Dominant asymptotic behavior of lower bounds on $\phi_{\max}^L$ for large d.

$(d/2)2^{-0.5d}$	Blichfeldt (1929)
$(d/e)2^{-0.5d}$	Rogers (1958)
$2^{-0.5990d}$	Kabatiansky and Levenshtein (1978)

TABLE 2. Dominant asymptotic behavior of upper bounds on $\phi_{\max}$ for large d.

is $2^d \phi \geq 1$. Thus, the bound (2–2), which is sometimes called the "greedy" lower bound, has the same dominant exponential term as (2–1). In Section 4.1, we show that there exists a construction of a disordered packing of congruent spheres that realizes the weaker lower bound of (2–2), i.e., $\phi = 2^{-d}$.

The best currently known lower bound on $\phi_{\max}^L$ was obtained by Ball [Ball 92]. He found that

$$\phi_{\max}^L \geq \frac{2(d-1)\zeta(d)}{2^d}. \qquad (2\text{–}3)$$

Table 1 gives the dominant asymptotic behavior of several lower bounds on $\phi_{\max}^L$ for large d.

Nontrivial upper bounds on the maximal density $\phi_{\max}$ for any sphere packing in $\mathbb{R}^d$ have been derived. Blichfeldt [Blichfeldt 29] showed that the maximal packing density for all d satisfies $\phi_{\max} \leq (d/2+1)2^{-d/2}$. This upper bound was improved by Rogers [Rogers 58, Rogers 64] by an analysis of the Voronoi cells. For large d, Rogers's upper bound asymptotically becomes $2^{-d/2}d/e$. Kabatiansky and Levenshtein [Kabatiansky and Levenshtein 78] found an even stronger bound, which in the limit $d \to \infty$ yields $\phi_{\max} \leq 2^{-0.5990d(1+o(1))}$. Cohn and Elkies [Cohn and Elkies 03] obtained and computed linear programming upper bounds, which provided improvement over Rogers's upper bound for dimensions 4 through 36. They also conjectured that their approach could be used to prove sharp bounds in 8 and 24 dimensions. Indeed, Cohn and Kumar [Cohn and Kumar 04] used these techniques to prove that the Leech lattice is the unique densest lattice in $\Re^{24}$. They also proved that no sphere packing in $\Re^{24}$ can exceed the density of the Leech lattice by a factor of more than $1 + 1.65 \times 10^{-30}$, and gave a new proof that E_8 is the unique densest lattice in $\Re^8$. Table

2 provides the dominant asymptotic behavior of several upper bounds on $\phi_{\max}$ for large d.

3.   REALIZABILITY OF POINT PROCESSES

As will be described in Section 5, our new approach to lower bounds on the density of sphere packings in $\mathbb{R}^d$ rests on whether certain one- and two-point correlation functions are realizable by sphere packings. As we will discuss, a sphere packing can be regarded as a special case of a point process and so a more general question concerns the necessary and sufficient conditions for the realizability of point processes in $\mathbb{R}^d$. Before discussing the realizability of point processes, it is useful to recall some basic results from the theory of spatial stochastic (or random) processes. Let $\mathbf{x} \equiv (x_1, x_2, \ldots, x_d)$ denote a vector position in $\mathbb{R}^d$. Consider a stochastic process $\{Y(\mathbf{x}; \omega) : \mathbf{x} \in \mathbb{R}^d; \omega \in \Omega\}$, where $Y(\mathbf{x}; \omega)$ is a real-valued random variable, ω is a realization generated by the stochastic process, and $(\Omega, \mathcal{F}, \mathcal{P})$ is a probability space (i.e., Ω is a sample space, $\mathcal{F}$ is a σ-algebra of measurable subsets of Ω, and $\mathcal{P}$ is a probability measure). For simplicity, we will often suppress the variable ω.

3.1   Ordinary Stochastic Processes

We will assume that the stochastic process is translationally invariant (i.e., statistically homogeneous in space). Let us further assume that the mean $\mu = \langle Y(\mathbf{x}) \rangle$ and autocovariance function

$$\chi(\mathbf{r}) = \langle [Y(\mathbf{x}) - \mu][Y(\mathbf{x} + \mathbf{r}) - \mu] \rangle \qquad (3\text{--}1)$$

exist, where angular brackets denote an expectation, i.e., an average over all realizations. The fact that the mean μ and autocovariance function $\chi(\mathbf{r})$ are independent of the variable $\mathbf{x}$ is a consequence of the translational invariance of the measure. Clearly,

$$\chi(0) = \langle Y^2 \rangle - \mu^2, \qquad (3\text{--}2)$$

and it follows from Schwarz's inequality that

$$|\chi(\mathbf{r})| \leq \langle Y^2 \rangle - \mu^2. \qquad (3\text{--}3)$$

It immediately follows [Loève 63] that the autocovariance function $\chi(\mathbf{r})$ must be positive semidefinite (nonnegative) in the sense that for any finite number of spatial locations $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_m$ in $\mathbb{R}^d$ and arbitrary real numbers $a_1, a_2, \ldots, a_m$,

$$\sum_{i=1}^{m} \sum_{j=1}^{m} a_i a_j \chi(\mathbf{r}_i - \mathbf{r}_j) \geq 0. \qquad (3\text{--}4)$$

It is clear that $\langle [Y(\mathbf{x} + \mathbf{r}) - Y(\mathbf{x})]^2 \rangle = 2[\chi(\mathbf{0}) - \chi(\mathbf{r})]$. Thus, if the autocovariance function $\chi(\mathbf{r})$ is continuous at the point $\mathbf{r} = \mathbf{0}$, the process $Y(\mathbf{x})$ on $\mathbb{R}^d$ will be *mean square continuous*, i.e., $\lim_{\mathbf{r} \to \mathbf{0}} \langle [Y(\mathbf{x} + \mathbf{r}) - Y(\mathbf{x})]^2 \rangle = 0$ for all $\mathbf{x}$. Stochastic processes that are continuous in the mean square sense will be called *ordinary*. It is simple to show that if $\chi(\mathbf{r})$ is continuous at $\mathbf{r} = \mathbf{0}$, it is continuous for all $\mathbf{r}$.

Does every continuous positive semidefinite function $f(\mathbf{r})$ correspond to a translationally invariant ordinary stochastic process with a continuous autocovariance $\chi(\mathbf{r})$? The answer is yes, and a proof is given in the book by Loève [Loève 63] for stochastic processes in time. Here we state without proof the generalization to stochastic processes in space.

Theorem 3.1. *A continuous function $f(\mathbf{r})$ on $\mathbb{R}^d$ is an autocovariance function of a translationally invariant ordinary stochastic process if and only if it is positive semidefinite.*

Remark 3.2. Assuming that $f(\mathbf{r})$ is positive semidefinite, one needs to show that there exists a random variable $Y(\mathbf{x})$ on $\mathbb{R}^d$ such that $\langle [Y(\mathbf{x}) - \mu][Y(\mathbf{x} + \mathbf{r}) - \mu] \rangle = f(\mathbf{r})$. This is done by demonstrating that there exists a Gaussian (normal) process for every autocovariance function [Loève 63]. A crucial property of a Gaussian process is that its full probability distribution is completely determined by its mean and autocovariance.

The nonnegativity condition (3–4) is difficult to check. It turns out that it is easier to establish the existence of an autocovariance function by appealing to its spectral representation. We denote the space of absolutely integrable functions on $\mathbb{R}^d$ by L^1. The Fourier transform of an L^1 function $f : \mathbb{R}^d \to \Re$ is defined by

$$\tilde{f}(\mathbf{k}) = \int_{\mathbb{R}^d} f(\mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}} d\mathbf{r}. \qquad (3\text{--}5)$$

If $f : \mathbb{R}^d \to \mathbb{R}$ is a radial function, i.e., f depends only on the modulus $r = |\mathbf{r}|$ of the vector $\mathbf{r}$, then its Fourier transform is given by

$$\tilde{f}(k) = (2\pi)^{\frac{d}{2}} \int_0^\infty r^{d-1} f(r) \frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}} dr, \qquad (3\text{--}6)$$

where k is the modulus of the wave vector $\mathbf{k}$ and $J_\nu(x)$ is the Bessel function of order ν. The Wiener–Khintchine theorem states that a necessary and sufficient condition for the existence of a continuous autocovariance function $\chi(\mathbf{r})$ of a translationally invariant stochastic process

$\{Y(\mathbf{x}) : \mathbf{x} \in \mathbb{R}^d\}$ is that its Fourier transform be non-negative everywhere, i.e., $\tilde{\chi}(\mathbf{k}) \geq 0$ for all $\mathbf{k}$ [Yaglom 87, Torquato 02]. The key "necessary" part of the proof of this theorem rests on a well-known theorem due to Bochner [Bochner 36], which states that any continuous function $f(\mathbf{r})$ is positive semidefinite in the sense of inequality (3–4) if and only if it has a Fourier–Stieltjes representation with a nonnegative bounded measure.

3.2   Generalized Stochastic Processes

The types of autocovariance functions that we are interested in must allow for generalized functions, such as Dirac delta functions. The Wiener–Khintchine theorem has been extended to autocovariances in the class of generalized functions called *tempered distributions*, i.e., continuous linear functionals on the space S of infinitely differentiable functions $\Phi(\mathbf{x})$ such that $\Phi(\mathbf{x})$ as well as all of its derivatives decays faster than polynomially. Nonnegative tempered distributions are nonnegative unbounded measures ν on $\mathbb{R}^d$ such that

$$\int_{\mathbb{R}^d} \frac{d\nu(\mathbf{r})}{(1 + |\mathbf{r}|)^n} < \infty$$

for some n. The interested reader is referred to the books by Gel'fand [Gel'fand and Vilenkin 64] and Yaglom [Yaglom 87] for details about generalized stochastic processes. It suffices to say here that $\{Y(\Phi(\mathbf{x})) : \mathbf{x} \in \mathbb{R}^d\}$ is a generalized stochastic process if for each $\Phi(\mathbf{x}) \in S$ we have a random variable $Y(\Phi(\mathbf{x}))$ that is linear and mean square continuous in Φ. Then the mean is the linear functional $\mu(\Phi(\mathbf{x})) = \langle Y(\Phi_1(\mathbf{x})) \rangle$ and the autocovariance function is the bilinear functional $\langle [Y(\Phi_1(\mathbf{x})) - \mu(\Phi(\mathbf{x}_1))][Y(\Phi_2(\mathbf{x}+\mathbf{r})) - \mu(\Phi(\mathbf{x}_2))] \rangle$, which we still denote by $\chi(\mathbf{r})$ for simplicity.

Theorem 3.3. *A necessary and sufficient condition for an autocovariance function $\chi(\mathbf{r})$ to come from a translationally invariant generalized stochastic process $\{Y(\Phi(\mathbf{x})) : \mathbf{x} \in \mathbb{R}^d\}$ is that its Fourier transform $\tilde{\chi}(\mathbf{k})$ be a nonnegative tempered distribution.*

Remark 3.4. We will call Theorem 3.3 the generalized Wiener–Khintchine theorem.

3.3   Stochastic Point Processes

Loosely speaking, a stochastic point process in $\mathbb{R}^d$ is defined as a mapping from a probability space to configurations of points $\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3, \ldots$ in $\mathbb{R}^d$. More precisely, let X denote the set of configurations such that each configuration $x \in X$ is a subset of $\mathbb{R}^d$ that satisfies two regularity conditions: (i) there are no multiple points ($\mathbf{x}_i \neq \mathbf{x}_j$ if $i \neq j$) and (ii) each bounded subset of $\mathbb{R}^d$ must contain only a finite number of points of x. We denote by $N(B)$ the number of points within $x \cap B$, $B \in \mathcal{B}$, where $\mathcal{B}$ is the ring of bounded Borel sets in R^d. Thus, we always have $N(B) < \infty$ for $B \in \mathcal{B}$ but the possibility $N(\mathbb{R}^d) = \infty$ is not excluded. We denote by $\mathcal{U}$ the minimal σ-algebra of subsets of X that renders all of the functions $N(B)$ measurable. Let $(\Omega, \mathcal{F}, \mathcal{P})$ be a probability space. Any measurable map $x(\omega) : \Omega \to X$, $\omega \in \Omega$, is called a stochastic point process [Stoyan 95]. Point processes belong to the class of generalized stochastic processes.

A particular realization of a point process in $\mathbb{R}^d$ can formally be characterized by the random variable

$$n(\mathbf{r}) = \sum_{i=1}^{\infty} \delta(\mathbf{r} - \mathbf{x}_i), \qquad (3\text{–}7)$$

called the "local" density at position $\mathbf{r}$, where $\delta(\mathbf{r})$ is a d-dimensional Dirac delta function. The "randomness" of the point process enters through the positions $\mathbf{x}_1, \mathbf{x}_2, \ldots$. Let us call

$$I_A(\mathbf{r}) = \begin{cases} 1, & \mathbf{r} \in A, \\ 0, & \mathbf{r} \notin A, \end{cases} \qquad (3\text{–}8)$$

the *indicator function* of a measurable set $A \subset \mathbb{R}^d$, which we also call a "window." For a particular realization, the number of points $N(A)$ within such a window is given by

$$\begin{aligned} N(A) &= \int_{\mathbb{R}^d} n(\mathbf{r}) I_A(\mathbf{r}) \, d\mathbf{r} \\ &= \sum_{i=1}^{\infty} \int_{\mathbb{R}^d} \delta(\mathbf{r} - \mathbf{x}_i) I_A(\mathbf{r}) \, d\mathbf{r} \\ &= \sum_{i \geq 1} I_A(\mathbf{x}_i). \qquad (3\text{–}9) \end{aligned}$$

Note that this random setting is quite general. It incorporates cases in which the locations of the points are deterministically known, such as a lattice. A packing of congruent spheres of unit diameter is simply a point process in which any pair of points cannot be closer than a unit distance from one another.

It is known that the probability measure on $(X, \mathcal{U})$ exists provided that the infinite set of n-point correlation functions ρ_n, $n = 1, 2, 3, \ldots$, meet certain conditions [Lennard 73, Lennard 75a, Lennard 75b]. The n-point correlation function $\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$ is the contribution to the expectation $\langle n(\mathbf{r}_1) n(\mathbf{r}_2) \cdots n(\mathbf{r}_n) \rangle$ when the indices

on the sums are not equal to one another, i.e.,

$$\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$$

$$= \left\langle \sum_{i_1 \neq i_2 \neq \cdots \neq i_n}^{\infty} \delta(\mathbf{r}_1 - \mathbf{x}_{i_1}) \delta(\mathbf{r}_2 - \mathbf{x}_{i_2}) \cdots \delta(\mathbf{r}_n - \mathbf{x}_{i_n}) \right\rangle.$$

Note that the distribution-valued function $\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$ also has a probabilistic interpretation: apart from trivial constants, it is the probability density function associated with finding n different points at positions $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n$. For this reason, ρ_n is also called the *n-particle density* and, for any n, has the nonnegativity property

$$\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n) \geq 0 \qquad \forall \mathbf{r}_i \in \mathbb{R}^d \quad (i = 1, 2, \ldots, n). \tag{3-10}$$

Translational invariance means that for every constant vector $\mathbf{y}$ in $\mathbb{R}^d$, $\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n) = \rho_n(\mathbf{r}_1 + \mathbf{y}, \ldots, \mathbf{r}_n + \mathbf{y})$, which implies that

$$\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n) = \rho^n g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}), \tag{3-11}$$

where ρ is the number (or center) density and $g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n})$ is the *n-particle correlation function*, which depends on the relative positions $\mathbf{r}_{12}, \mathbf{r}_{13}, \ldots$, where $\mathbf{r}_{ij} \equiv \mathbf{r}_j - \mathbf{r}_i$ and we have chosen the origin to be at $\mathbf{r}_1$.

For such point processes without *long-range order*, $g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}) \to 1$ when the points (or "particles") are mutually far from one another, i.e., as $|\mathbf{r}_{ij}| \to \infty$ $(1 \leq i < j < \infty)$, $\rho_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n) \to \rho^n$. Thus, the deviation of g_n from unity provides a measure of the degree of spatial correlation between the particles, with unity corresponding to no spatial correlation. Note that for a translationally invariant Poisson point process, g_n is unity for all values of its argument.

As we indicated in the beginning of this section, the first two correlation functions, $\rho_1(\mathbf{r}_1) = \rho$ and $\rho_2(\mathbf{r}_1, \mathbf{r}_2) = \rho^2 g_2(\mathbf{r})$, for translationally invariant point processes are of central concern in this paper. If the point process is also rotationally invariant (statistically isotropic), then g_2 depends on the radial distance $r = |\mathbf{r}|$ only, i.e.,

$$g_2(\mathbf{r}) = g_2(r), \tag{3-12}$$

and is referred to as the *radial distribution function*. Strictly speaking, one should use different notation for the left and right members of (3–12), but to conform to conventional statistical-mechanical usage, we invoke the common notation for both. Because $\rho_2(\mathbf{r}_1, \mathbf{r}_2)/\rho = \rho g_2(r)$ is a conditional joint probability density, then

$$Z(r_1, r_2) = \int_{r_1}^{r_2} \rho s_1(r) g_2(r) \, dr$$

is the expected number of points at radial distances between r_1 and r_2 from a randomly chosen point. Here $s_1(r)$ is the surface area of a d-dimensional sphere of radius r given by

$$s_1(r) = \frac{2\pi^{d/2} r^{d-1}}{\Gamma(d/2)}. \tag{3-13}$$

For a packing of congruent spheres of unit diameter, $g(r) = 0$ for $0 \leq r < 1$, i.e.,

$$\mathrm{supp}(g_2) \subseteq \{r : r \geq 1\}. \tag{3-14}$$

Note that the radial distribution function $g_2(r)$ (or any of the ρ_n) for a point process must be able to incorporate Dirac delta functions. We will specifically consider those radial distribution functions that are nonnegative distributions. For example, $g_2(r)$ for a lattice packing is the rotational symmetrization of the sum of delta functions at lattice points at a radial distance r from any lattice point [Torquato and Stillinger 03].

For a translationally invariant point process, the auto-covariance function $\chi(\mathbf{r})$ is related to the pair correlation function via

$$\chi(\mathbf{r}) = \rho \delta(\mathbf{r}) + \rho^2 h(\mathbf{r}),$$

where

$$h(\mathbf{r}) \equiv g_2(\mathbf{r}) - 1 \tag{3-15}$$

is the *total correlation function.* This relation is obtained using definitions (3–1) and (3–7) with $Y(\mathbf{x}) = n(\mathbf{x})$. Note that $\chi(\mathbf{r}) = \rho \delta(\mathbf{r})$ (i.e., $h = 0$) for a translationally invariant Poisson point process. *Positive and negative pair correlations* are defined as those instances in which h is positive (i.e., $g_2 > 1$) and h is negative (i.e., $g_2 < 1$), respectively. The Fourier transform of the distribution-valued function $\chi(\mathbf{r})$ is given by

$$\tilde{\chi}(\mathbf{k}) = \rho + \rho^2 \tilde{h}(\mathbf{k}), \tag{3-16}$$

where $\tilde{h}(\mathbf{k})$ is the Fourier transform of $h(\mathbf{r})$. It is common practice in statistical physics to deal with a function trivially related to the spectral function $\tilde{\chi}(\mathbf{k})$ called the structure factor $S(\mathbf{k})$, i.e.,

$$S(\mathbf{k}) \equiv \frac{\tilde{\chi}(\mathbf{k})}{\rho} = 1 + \rho \tilde{h}(\mathbf{k}). \tag{3-17}$$

A natural question to ask at this point is the following: Given a positive number density ρ and a pair correlation function $g_2(\mathbf{r})$, does there exist a translationally invariant point process in $\mathbb{R}^d$ with measure $\mathcal{P}$ for which ρ and g_2

are one-point and two-point correlation functions, respectively? Two obvious nonnegativity conditions [Torquato and Stillinger 02] that must be satisfied are the following:

$$g_2(\mathbf{r}) \geq 0 \qquad \text{for all} \quad \mathbf{r} \qquad (3\text{--}18)$$

and

$$S(\mathbf{k}) = 1 + \rho \tilde{h}(\mathbf{k}) \geq 0 \qquad \text{for all} \quad \mathbf{k}. \qquad (3\text{--}19)$$

The first condition is trivial and comes from (3–10) with $n = 2$. The second condition is nontrivial and derives from the generalized Wiener–Khintchine theorem (Theorem 3.3) using relations (3–16) and (3–17). However, for realizability of point processes in arbitrary dimension d, the two standard conditions (3–18) and (3–19) are only necessary, not necessary and sufficient. The same state of affairs applies to the theory of random sets [Torquato 02], where it is known that the Wiener–Khintchine theorem provides only a necessary condition on realizable autocovariance functions. The simplest example of a random set is one in which $\mathbb{R}^d$ is partitioned into two disjoint regions (phases) but with an interface that is known only in a probabilistic sense. (A packing can therefore be viewed as a special random set.) Thus, a random set is described by a random variable that is the indicator function for a particular phase, i.e., it is a binary stochastic process. The class of autocovariances that comes from a binary stochastic process is a subclass of the total class that comes from an ordinary process $\{Y(\mathbf{x}) : \mathbf{x} \in \mathbb{R}^d\}$ and meets the existence condition of Theorem 3.3. Similarly, the class of autocovariances that comes from a point process is a subset of of a generalized process $\{Y(\Phi(\mathbf{x})) : \mathbf{x} \in \mathbb{R}^d\}$ and therefore the existence condition of Theorem 3.3 is only necessary.

It has recently come to light that a positive g_2 for a positive ρ must satisfy an uncountable number of necessary and sufficient conditions for it to correspond to a realizable point process [Costin and Lebowitz 04]. However, these conditions are very difficult (or, more likely, impossible) to check for arbitrary dimension. In other words, given $\rho_1 = \rho$ and $\rho_2 = \rho^2 g_2$, it is difficult to ascertain whether there are some higher-order functions $\rho_3, \rho_4, \ldots$ for which these one- and two-point correlation functions hold. Thus, an important practical problem becomes the determination of a manageable number of necessary conditions that can be readily checked.

One such additional necessary condition, obtained by Yamada [Yamada 61], is concerned with $\sigma^2(A) \equiv \langle (N(A) - \langle N(A) \rangle)^2 \rangle$, the variance in the number of points $N(A)$ contained within a window $A \subset \mathbb{R}^d$. Specifically,

he showed that

$$\sigma^2(A) \geq \theta(1 - \theta), \qquad (3\text{--}20)$$

where θ is the fractional part of the expected number of points $\rho|A|$ contained in the window. This inequality is a consequence of the fact that the number of points $N(A)$ within a window at some fixed position is an integer, not a continuous variable, and sets a lower limit on the number variance. We note in passing that the determination of the number variance for lattice point patterns is an outstanding problem in number theory [Kendall 48, Kendall and Rankin 53, Sarnak and Strömbergsson 05]. The number variance for a specific choice of A is necessarily a positive number and generally related to the total pair correlation function $h(\mathbf{r})$ for a translationally invariant point process [Torquato and Stillinger 03]. In the special case of a spherical window of radius R in $\mathbb{R}^d$, it is explicitly given by

$$\sigma^2(R) = \rho v_1(R) \left[1 + \rho \int_{\mathbb{R}^d} h(\mathbf{r}) \alpha_2(\mathbf{r}; R) \, d\mathbf{r} \right] \geq \theta(1 - \theta),$$
$$(3\text{--}21)$$

where $\sigma^2(R)$ is the number variance for a spherical window of radius R in $\mathbb{R}^d$, $v_1(R)$ is the volume of the window, and $\alpha_2(r; R)$ is the volume common to two spherical windows of radius R whose centers are separated by a distance r divided by $v_1(R)$. We will call $\alpha_2(r; R)$ the *scaled intersection volume*. The lower bound (3–21) provides another integral condition on the pair correlation function.

For large R, it has been proved that $\sigma^2(R)$ cannot grow more slowly than γR^{d-1}, where γ is a positive constant [Beck 87]. This implies that the Yamada lower bound in (3–21) is always satisfied for sufficiently large R for any $d \geq 2$. In fact, we have not been able to construct any examples of a pair correlation function $g_2(\mathbf{r})$ at some number density ρ that satisfy the two nonnegativity conditions (3–18) and (3–19) and simultaneously violate the Yamada condition for any R and any $d \geq 2$. Thus, it appears that the Yamada condition is most relevant in one dimension, especially in those cases in which $\sigma^2(R)$ is bounded. We note that point processes (translationally invariant or not) for which $\sigma^2(R) \sim R^{d-1}$ for large R are examples of *hyperuniform* point patterns [Torquato and Stillinger 03]. This classification includes all lattices as well as aperiodic point patterns. Hyperuniformity implies that the structure factor $S(\mathbf{k})$ has the following small-$\mathbf{k}$ behavior:

$$\lim_{\mathbf{k} \to 0} S(\mathbf{k}) = 0. \qquad (3\text{--}22)$$

The scaled intersection volume $\alpha_2(r; R)$ appearing in (3–21) will play a prominent role in this paper. It has support in the interval $[0, 2R]$, range $[0, 1]$, and the following integral representation:

$$\alpha_2(r; R) = c(d) \int_0^{\cos^{-1}(r/(2R))} \sin^d(\theta)\, d\theta, \qquad (3\text{–}23)$$

where $c(d)$ is the d-dimensional constant given by

$$c(d) = \frac{2\Gamma(1 + d/2)}{\pi^{1/2}\Gamma((d+1)/2)}.$$

Following the analysis given by Torquato and Stillinger [Torquato and Stillinger 03] for low dimensions, we obtain the following *new* series representation of the scaled intersection volume $\alpha_2(r; R)$ for $r \leq 2R$ and for any d:

$$\alpha(r; R) = 1 - c(d)x \qquad (3\text{–}24)$$
$$+ c(d) \sum_{n=2}^{\infty} (-1)^n \frac{(d-1)(d-3)\cdots(d-2n+3)}{(2n-1)[2 \cdot 4 \cdot 6 \cdots (2n-2)]} x^{2n-1},$$

where $x = r/(2R)$. This is also easily proved, starting from (3–24), with the help of Maple. For even dimensions, relation (3–24) is an infinite series, but for odd dimensions, the series truncates such that $\alpha_2(r; R)$ is a univariate polynomial of degree d. Except for the first term of unity, all of the terms in relation (3–24) involve only odd powers of x. Figure 1 shows graphs of the scaled intersection volume $\alpha_2(r; R)$ as a function of r for the first five space dimensions. For any dimension, $\alpha(r; R)$ is a monotonically decreasing function of r. At a fixed value of r in the interval $(0, 2R)$, $\alpha_2(r; R)$ is a monotonically decreasing function of the dimension d. For large d, we will subsequently make use of the asymptotic result

$$\alpha_2(R; R) \sim \left(\frac{6}{\pi}\right)^{1/2} \left(\frac{3}{4}\right)^{d/2} \frac{1}{d^{1/2}}. \qquad (3\text{–}25)$$

Before closing this section, it is useful to note that there has been some recent work that demonstrates the existence of point processes for a specific ρ and g_2 provided that ρ and g_2 meet certain restrictions. For example, Ambartzumian and Sukiasian proved the existence of point processes that come from Gibbs measures for a special g_2 for sufficiently small ρ [Ambartzumian and Sukiasian 91]. Determinantal point processes have been considered by Soshnikov [Soshnikov 00] and Costin and Lebowitz [Costin and Lebowitz 04]. Costin and Lebowitz have also studied certain one-dimensional renewal point processes [Costin and Lebowitz 04]. Stillinger and Torquato [Stillinger and Torquato 04] discussed
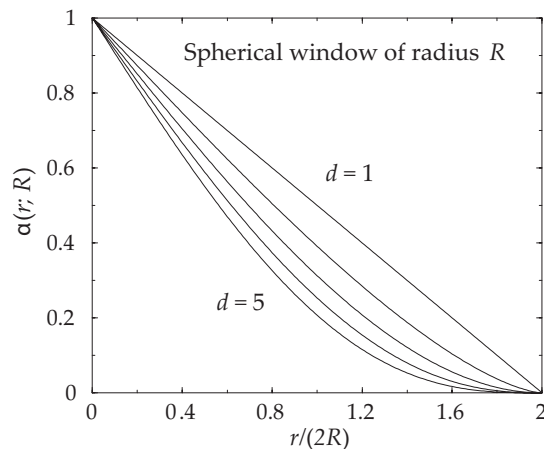

FIGURE 1. The scaled intersection volume $\alpha_2(r; R)$ for spherical windows of radius R as a function of r for the first five space dimensions. The uppermost curve is for $d = 1$ and lowermost curve is for $d = 5$.

the possible existence of a general interparticle pair potential (associated with a Gibbs measure) for a given ρ and g_2 using a cluster expansion procedure but did not address the issue of convergence of this expansion. Koralov [Koralov 05] indeed proves the existence of a pair potential on a lattice (with the restriction of single occupancy per lattice site) for which ρ is the density and g_2 is the pair correlation function for sufficiently small ρ and g_2. There is no reason to believe that Koralov's proof is not directly extendable to the case of a point process corresponding to a sphere packing in $\mathbb{R}^d$, where the nonoverlap condition is the analogue of single occupancy on the lattice. Thus, we expect that one can prove the existence of a pair potential in $\mathbb{R}^d$ corresponding to a sphere packing for a given ρ and g_2 provided that ρ and g_2 are sufficiently small.

4. DISORDERED PACKINGS IN HIGH DIMENSIONS AND THE DECORRELATION PRINCIPLE

In this section, we examine the asymptotic behavior of certain disordered packings in high dimensions and show that unconstrained spatial correlations vanish asymptotically, yielding a *decorrelation principle*. We define a *disordered packing* in $\mathbb{R}^d$ to be one in which the pair correlation function $g_2(\mathbf{r})$ decays to its long-range value of unity faster than $|\mathbf{r}|^{-d-\varepsilon}$ for some $\varepsilon > 0$. The decorrelation principle as well as a number of other results (which will be discussed in Section 5) motivate us to propose a conjecture in Section 5 that describes the circumstances in which the two standard nonnegativity conditions given

by (3–18) and (3–19) are necessary and sufficient to ensure the existence of a disordered sphere packing.

4.1   Example 1: Disordered Sequential Packings

First we show that there exists a disordered sphere packing that realizes the greedy lower bound (2–2) ($\phi = 1/2^d$) for all d. Then we study the asymptotic properties of the n-particle correlation functions in the large-dimension limit.

The disordered packing that achieves the greedy lower bound is a special case of a generalization of the so-called random sequential addition (RSA) process [Torquato 02]. This generalization, which we introduce here, is a subset of the Poisson point process. Specifically, the centers of "test" spheres of unit diameter arrive continually throughout $\mathbb{R}^d$ during time $t \geq 0$ according to a translationally invariant Poisson process of density per unit time η, i.e., η is the number of points per unit volume and time. Therefore, the expected number of centers in a region of volume Ω during time t is $\eta\Omega t$, and the probability that this region is empty of centers is $\exp(-\eta\Omega t)$. However, this Poisson distribution of test spheres is not a packing because the spheres can overlap. To create a packing from this point process, one must remove test spheres such that no sphere center can lie within a spherical region of unit radius from any sphere center. Without loss of generality, we will set $\eta = 1$.

There is a variety of ways of achieving this "thinning" process such that the subset of points corresponds to a sphere packing. One obvious rule is to retain a test sphere at time t only if it does not overlap a sphere that was successfully added to the packing at an earlier time. This criterion defines the well-known RSA process in $\mathbb{R}^d$ [Torquato 02], and is clearly a statistically homogeneous and isotropic sphere packing in $\mathbb{R}^d$ with a time-dependent density $\phi(t)$. In the limit $t \to \infty$, the RSA process corresponds to a saturated packing with a maximal or *saturation* density $\phi_s(\infty) \equiv \lim_{t\to\infty} \phi(t)$. In one dimension, the RSA process is commonly known as the "car-parking problem," which Reńyi showed has a saturation density $\phi_s(\infty) = 0.7476\dots$ [Reńyi 63]. For $2 \leq d < \infty$, an exact determination of $\phi_s(\infty)$ is not possible, but estimates for it have been obtained via computer experiments for low dimensions [Torquato 02]. However, as we will discuss below, the standard RSA process at small times (or, equivalently, small densities) can be analyzed exactly.

Another thinning criterion retains a test sphere centered at position $\mathbf{r}$ at time t if no other test sphere is within a unit radial distance from $\mathbf{r}$ for the time interval κt prior to t, where κ is a positive constant in the interval

$[0, 1]$. This packing is a subset of the RSA packing, and therefore we refer to it as the generalized RSA process. Note that when $\kappa = 0$, the standard RSA process is recovered, and when $\kappa = 1$, a relatively unknown model due to Matérn [Matérn 86] is recovered. The latter is amenable to exact analysis.

The time-dependent density $\phi(t)$ in the case of the generalized RSA process with $\kappa = 1$ is easily obtained. (Note that for any $0 < \kappa \leq 1$, the generalized RSA process is always an *unsaturated* packing.) In this packing, a test sphere at time t is accepted only if it does not overlap an existing sphere in the packing as well as any previously rejected test spheres (which we will call "ghost" spheres). An overlap cannot occur if a test sphere is outside a unit radius of any successfully added sphere or ghost sphere. Because of the underlying Poisson process, the probability that a trial sphere is retained at time t is given by $\exp(-v_1(1)t)$, where $v_1(1)$ is the volume of a sphere of unit radius having the same center as the retained sphere of radius $\frac{1}{2}$. Therefore, the expected number density $\rho(t)$ and packing density $\phi(t)$ at any time t are respectively given by

$$\rho(t) = \int_0^t \exp(-v_1(1)t')\, dt' = \frac{1}{v_1(1)}[1 - \exp(-v_1(1)t)]$$

and

$$\phi(t) = \rho(t)v_1(1/2) = \frac{1}{2^d}[1 - \exp(-v_1(1)t)]. \qquad (4\text{–}1)$$

We see that $\phi(t)$ is a monotonically increasing function of t. This result was first given by Matérn using a different approach and he also gave a formal expression for the time-dependent radial distribution function $g_2(r;t)$ (see Section 3). Here we present an explicit expression for $g_2(r;t)$ at time t for any dimension d:

$$g_2(r;t) = \frac{\Theta(r-1)}{2^{2d-1}[\beta_2(r;1)-1]\phi^2(t)} \qquad (4\text{–}2)$$

$$\times \left[2^d\phi(t) - \frac{1 - \exp[-2^d\beta_2(r;1)t]}{\beta_2(r;1)}\right].$$

Here

$$\Theta(x) = \begin{cases} 0, & x < 0, \\ 1, & x \geq 0, \end{cases} \qquad (4\text{–}3)$$

is the unit step function and

$$\beta_2(r;R) = 2 - \alpha_2(r;R)$$

is the union volume of two spheres of radius R (whose centers are separated by a distance r) divided by the volume of a sphere of radius R and $\alpha_2(r;R)$ is the scaled

intersection volume of two such spheres given by equation (3–23). Our approach for obtaining (4–2) is different from Matérn's and details are given elsewhere [Torquato and Stillinger 06].

It is useful to note that at small times or, equivalently, low densities, formula (4–1) yields the asymptotic expansion $\phi(t) = v_1(1)t/2^d - v_1^2(1)t^2/2^{d+1} + \mathcal{O}(t^3)$, which when inverted yields $t = 2^d\phi/v_1(1) + 2^{d-1}\phi^2 + \mathcal{O}(\phi^3)$. Substitution of this last result into (4–2) gives

$$g_2(r;\phi) = \Theta(r-1) + \mathcal{O}(\phi^3), \qquad (4\text{–}4)$$

which implies that $g_2(r;\phi)$ tends to the unit step function $\Theta(r-1)$ as $\phi \to 0$ for any finite d.

In the limit $t \to \infty$, the maximum density is given by

$$\phi(\infty) \equiv \lim_{t\to\infty} \phi(t) = \frac{1}{2^d}$$

and

$$g_2(r;\infty) \equiv \lim_{t\to\infty} g_2(r;t) = \frac{2\Theta(r-1)}{\beta_2(r;1)} = \frac{\Theta(r-1)}{1 - \alpha_2(r;1)/2}. \qquad (4\text{–}5)$$

We see that the greedy lower-bound limit on the density is achieved in the infinite-time limit for this sequential but unsaturated packing. This is the first time that such an observation has been made. Obviously, for any $0 \le \kappa < 1$, the maximum (infinite-time) density of the generalized RSA packing is bounded from below by $1/2^d$ (the maximum density for $\kappa = 1$). Note also that because $\beta_2(r;1)$ is equal to 2 for $r \ge 2$, $g_2(r;\infty) = 1$ for $r \ge 2$, i.e., spatial correlations vanish identically for all pair distances except those in the small interval $[0,2)$. Even the positive correlations exhibited for $1 < r < 2$ are rather weak and decrease with increasing dimension. The function $g_2(r;\infty)$ achieves its largest value at $r = 1^+$ in any dimension and for $d = 1$, $g_2(1^+;\infty) = \frac{4}{3}$. The radial distribution function $g_2(r;\infty)$ is plotted in Figure 2 for the first five space dimensions. Using the asymptotic result (3–25) and relation (4–5), it is seen that for large d,

$$g_2(1^+;\infty) \sim \frac{\Theta(r-1)}{1 - \left(\frac{3}{2\pi}\right)^{1/2}\left(\frac{3}{4}\right)^{d/2}\frac{1}{d^{1/2}}},$$

and thus $g_2(r;\infty)$ tends to the unit step function $\Theta(r-1)$ exponentially fast as $d \to \infty$ because the scaled intersection volume $\alpha_2(1;1)$ vanishes exponentially fast.

The higher-order correlation functions for this model have not been given previously. In another work [Torquato and Stillinger 06], we use an approach different from the one used by Matérn to obtain not only g_2 but an explicit formula for the general n-particle correlation
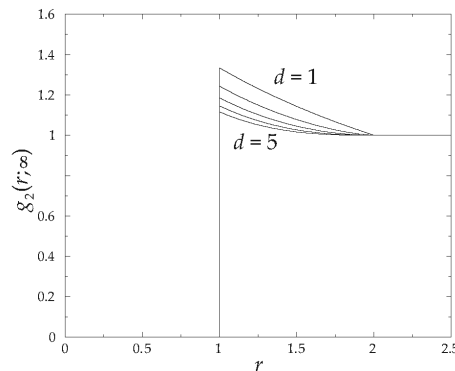

FIGURE 2. Radial distribution function for the first five space dimensions at the maximum density $\phi = 1/2^d$ for the generalized RSA model with $\kappa = 1$.

function g_n, defined by (3–11), for any time t and n and for arbitrary dimension d. To our knowledge, this represents the first exactly solvable disordered sphere-packing model for any d. These details are somewhat tangential to the present work and for our purposes it suffices to state the final result in the limit $t \to \infty$ for $n \ge 2$:

$$g_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\infty) = \frac{\prod_{i<j}^n \Theta(r_{ij}-1)}{\beta_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;1)}\left[\sum_{i=1}^n g_{n-1}(Q_i;\infty)\right], \qquad (4\text{–}6)$$

where the sum is over all the n distinguishable ways of choosing $n-1$ positions from n positions $\mathbf{r}_1,\ldots,\mathbf{r}_n$ and the arguments of g_{n-1} are the associated $n-1$ positions, which we denote by Q_i, and $g_1 \equiv 1$. Moreover, $\beta_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;R)$ is the union volume of n congruent spheres of radius R, whose centers are located at $\mathbf{r}_1,\ldots,\mathbf{r}_n$, where $r_{ij} = |\mathbf{r}_j - \mathbf{r}_i|$ for all $1 \le i < j \le n$, divided by the volume of a sphere of radius R.

Lemma 4.1. *In the limit $d \to \infty$, the n-particle correlation function $g_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\infty)$ approaches 1 uniformly in $(\mathbf{r}_1,\ldots,\mathbf{r}_n) \in \mathbb{R}^d$ such that $r_{ij} \ge 1$ for all $1 \le i < j \le n$. If $r_{ij} < 1$ for any pair of points $\mathbf{r}_i$ and $\mathbf{r}_j$, then $g_n(\mathbf{r}_1,\ldots,\mathbf{r}_n;\infty) = 0$.*

Proof: The second part of the lemma is the trivial requirement for a packing. Whenever $r_{ij} \ge 1$ for all $1 \le i < j \le n$, it is clear from (4–6) that we have the following upper and lower bounds on the n-particle correlation function:

$$\frac{n}{\beta_n} \le g_n \le \frac{ng_{n-1}^*}{\beta_n},$$

where g_{n-1}^* denotes the largest possible value of g_{n-1}. The scaled union volume β_n of n spheres obeys the

bounds

$$n - \sum_{i<j} \alpha_2(r_{ij}; 1) \leq \beta_n \leq n,$$

but since the scaled intersection volume of two spheres $\alpha_2(r; 1)$ attains its maximum value for $r \geq 1$ when $r = 1$, we also have

$$n - \frac{n(n-1)}{2}\alpha_2(1; 1) \leq \beta_n \leq n.$$

Use of this inequality and the recursive relation (4–6) yields the bounds

$$1 \leq g_n \leq \frac{1}{1 - \frac{1}{4}n(n-1)\alpha_2(1; 1) + \mathcal{O}(\alpha_2(1; 1)^2)}.$$

Using the asymptotic result (3–25), we see that the upper bound tends to the lower bound for any given n as $d \to \infty$, which proves the lemma. □

In summary, the lemma enables us to conclude that in the limit $d \to \infty$ and for $\phi = 1/2^d$,

$$g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}; \infty) \sim \prod_{i<j}^n g_2(r_{ij}; \infty),$$

where

$$g_2(r; \infty) \sim \Theta(r - 1). \qquad (4\text{–}7)$$

Importantly, we see that the asymptotic behavior of g_2 in the low-density limit $\phi \to 0$ for any d [cf. (4–4)] is the same as the high-dimensional limit $d \to \infty$ [cf. (4–7)], i.e., spatial correlations, which exist for positive densities at fixed d, vanish for pair distances beyond the hard-core diameter. Note also that g_n for $n \geq 3$ asymptotically factorizes into products involving only the pair correlation function g_2. Is the similarity between the low-density and high-dimensional limits for this model of a disordered packing a general characteristic of disordered packings? In what follows, we discuss another disordered packing that has this attribute and subsequently formulate what we refer to as a "decorrelation principle."

4.2 Example 2: The Classic Gibbsian Hard-Sphere Packing

The statistical mechanics of the classic Gibbsian hard-sphere packing is well established (see [Torquato 02] and references therein). The purpose of this subsection is simply to collect some results that motivate the decorrelation principle. Let $\Phi_N(\mathbf{r}^N)$ be the N-body interaction potential for a finite but large number of particles with configuration $\mathbf{r}^N \equiv \{\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N\}$ in a volume V in $\mathbb{R}^d$ at absolute temperature T. A large collection of such

systems in which N, V, and T are fixed but in which the particle configurations are otherwise free to vary is called the *Gibbs canonical ensemble*. Our interest is in the *thermodynamic limit*, i.e., the distinguished limit in which $N \to \infty$ and $V \to \infty$ such that the number density $\rho = N/V$ exists. For a Gibbs canonical ensemble, when the n-particle densities ρ_n (defined in Section 3) exist, they are entirely determined by the interaction potential $\Phi_N(\mathbf{r}^N)$. For a hard-sphere packing, the interaction potential is given by a sum of pairwise terms such that

$$\Phi_N(\mathbf{r}^N) = \sum_{i<j}^N u_2(|\mathbf{r}_j - \mathbf{r}_i|), \qquad (4\text{–}8)$$

where $u_2(r)$ is the pair potential defined by

$$u_2(r) = \begin{cases} +\infty, & r < 1, \\ 0, & r \geq 1. \end{cases} \qquad (4\text{–}9)$$

Thus, the particles do not interact for interparticle separation distances greater than or equal to unity but experience an infinite repulsive force for distances less than unity. The hard spheres have kinetic energy, and therefore a temperature, but the temperature enters in a trivial way because the configurational correlations between the spheres are independent of the temperature. We call this the *classic equilibrium* sphere packing, which is both translationally and rotationally invariant.

In one dimension, the n-particle densities ρ_n for such packings are known exactly in the thermodynamic limit. The density ϕ lies in the interval $[0, 1]$ but this one-dimensional packing is devoid of a discontinuous (first-order) transition from a disordered (liquid) phase to an ordered (solid) phase. Although a rigorous proof for the existence of a liquid-to-solid phase transition in two or three dimensions is not yet available, there is overwhelming numerical evidence (as obtained from computer simulations) that such a transformation takes place at sufficiently high densities. The maximal densities for equilibrium sphere packings in two and three dimensions are $\phi_{\max} = \pi/\sqrt{12}$ and $\phi_{\max} = \pi/\sqrt{18}$, respectively, i.e., they correspond to the density of the densest sphere packing in the respective dimension.

Figure 3 shows the three-dimensional radial distribution function as obtained from computer simulations for a density $\phi = 0.49$, which is near the maximum value for the stable disordered branch. It is seen that the packing exhibits *short-range* order (i.e., $g_2(r)$ has both positive and negative correlations for small r), but $g_2(r)$ decays to its long-range value exponentially fast after several diameters. By contrast, in the limit $d \to \infty$, it has been shown
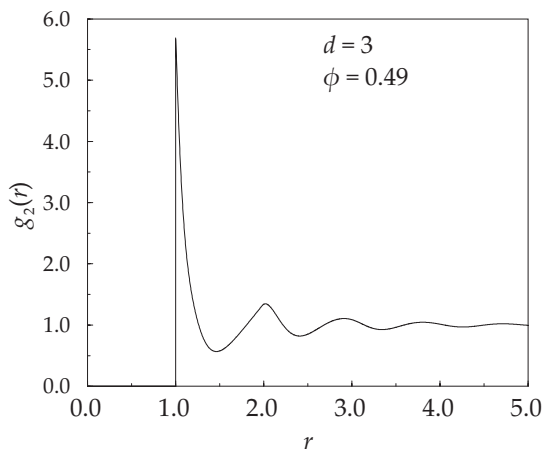
FIGURE 3. The radial distribution function for the classic three-dimensional equilibrium packing at $\phi = 0.49$ as obtained from molecular-dynamics computer simulations. The graph is adapted from Figure 3.15 of [Torquato 02].

that the "pressure" [Ruelle 99] of an equilibrium packing is exactly given by the first two terms of its asymptotic low-density expansion for some positive density interval $[0, \phi_0]$ [Wyler et al. 87, Frisch and Percus 99]. (Roughly speaking, the pressure is the average force per unit area acting on an "imaginary planar wall" in the packing due to collisions between the spheres and the wall.) Frisch and Percus [Frisch and Percus 99] have established, albeit not rigorously, that $\phi_0 = 1/2^d$. This result for the pressure implies that the leading-order term of the low-density expansion of the radial distribution function in arbitrary dimension [Torquato 02]

$$g_2(r) = \Theta(r-1)\Big[1 + 2^d \alpha_2(r;1)\phi + \mathcal{O}(\phi^2)\Big] \quad (4\text{–}10)$$

becomes asymptotically exact in the limit $d \to \infty$ in the same density interval. The presence of the unit step function $\Theta(r-1)$ in relation (4–10) means that the scaled intersection volume $\alpha_2(r;1)$ need be considered only for values of r in the interval $[1, 2]$. Since $\alpha_2(r;1)$ is largest when $r = 1$ for $1 \leq r \leq 2$ and $\alpha_2(1;1)$ has the asymptotic behavior (3–25), the product $2^d \alpha_2(1;1)\phi$ vanishes no more slowly than $(6/\pi)^{1/2}/[(4/3)^{d/2}d^{1/2}]$ in the limit $d \to \infty$ for $0 \leq \phi \leq 1/2^d$, and therefore $g_2(r)$ tends to $\Theta(r-1)$ exponentially fast. In summary, we see again that spatial correlations that exist in low dimensions for $r > 1$ completely vanish in the limit $d \to \infty$. Moreover, this is yet another disordered packing model in which the high-dimensional asymptotic behavior corresponds to the low-density asymptotic behavior.

The corresponding n-particle correlation function g_n, defined by (3–11), in the low-density limit [Salpeter 58]

is given by

$$g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n})$$
$$= \prod_{i<j}^{n} g_2(r_{ij})\Big[1 + 2^d \alpha_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}; 1)\phi + \mathcal{O}(\phi^2)\Big],$$

where $\alpha_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}; R)$ is the intersection volume of n congruent spheres of radius R (whose centers are located at $\mathbf{r}_1, \ldots, \mathbf{r}_n$, where $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ for all $1 \leq i < j \leq n$) divided by the volume of a sphere of radius R. The scaled intersection volume $\alpha_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}; R)/n$ has the range $[0, 1]$. Now since $\alpha_2(r_{ij}, 1) \geq \alpha_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}; 1)$ for any pair distance $r_{ij} = |\mathbf{r}_{ij}|$ such that $1 \leq i < j \leq n$, it follows from the analysis above that in the limit $d \to \infty$ for $0 \leq \phi \leq 1/2^d$,

$$g_2(r) \sim \Theta(r-1) \quad (4\text{–}11)$$

and

$$g_n(\mathbf{r}_{12}, \ldots, \mathbf{r}_{1n}) \sim \prod_{i<j}^{n} g_2(r_{ij}). \quad (4\text{–}12)$$

Again, as in the generalized RSA example with $\kappa = 1$, g_n factorizes into products involving only g_2's in the limit $d \to \infty$. Moreover, we should also note that the standard RSA process (generalized RSA process with $\kappa = 0$) has precisely the same asymptotic low-density behavior as the standard Gibbs hard-sphere model [Torquato 02]. More precisely, these two models share the same low-density expansions of the g_n through terms of order ϕ and therefore the same asymptotic expressions (4–11), (4–12).

4.3 Decorrelation Principle

The previous two examples illustrate two important and related asymptotic properties that are expected to apply to all disordered packings:

1. the high-dimensional asymptotic behavior of g_2 is the same as the asymptotic behavior in the low-density limit for any finite d, i.e., *unconstrained* spatial correlations, which exist for positive densities at fixed d, vanish asymptotically for pair distances beyond the hard-core diameter in the high-dimensional limit;

2. g_n for $n \geq 3$ asymptotically can be inferred from a knowledge of only the pair correlation function g_2 and number density ρ.

What is the explanation for these two related asymptotic properties? Because we know from the Kabatiansky-Levenshtein asymptotic upper bound on the maximal
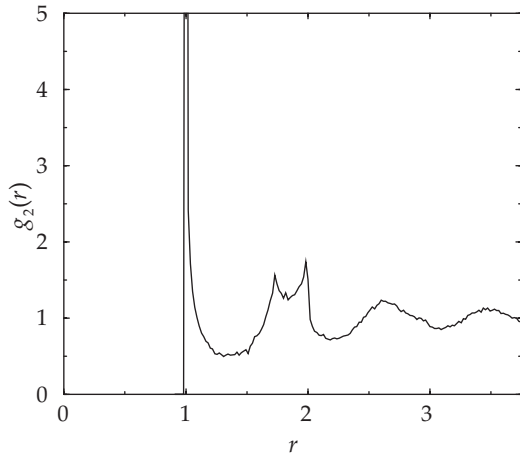
FIGURE 4.   The radial distribution function of a three-dimensional packing of spheres near the maximally random jammed state [Torquato et al. 00, Torquato 02] at a density $\phi = 0.64$ as obtained from computer simulations [Torquato and Stillinger 02]. The delta function contribution at $r = 1$ (of course, not explicitly shown) corresponds to an average kissing number of about six.

density that ϕ must go to zero at least as fast as $2^{-0.5990d}$ for large d [Kabatiansky and Levenshtein 78], unconstrained spatial correlations between spheres are expected to vanish, i.e., statistical independence is established. (An example of *constrained* spatial correlations is described below.) Such a decorrelation means that the g_n for $n \geq 3$ are determined entirely from a knowledge of the decorrelated pair correlation function g_2. In the specific examples that we considered, the g_n factorize into products involving only g_2's, but there may be other decompositions. For example, the g_n for $n \geq 3$ can be functionals that involve only ρ and g_2. We will call the two asymptotic properties the *decorrelation principle* for disordered packings. This principle as well as other results described in Section 5 leads us to a conjecture concerning the existence of disordered sphere packings in high dimensions, which we state in Section 5.1.

An example of constrained spatial correlations that would not vanish asymptotically is illustrated in Figure 4, where we show the pair correlation function $g_2(r)$ for a three-dimensional sphere packing near the so-called *maximally random jammed state* [Torquato et al. 00, Torquato 02]. A special feature of $g_2(r)$ for a maximally random jammed packing is a delta-function contribution at $r = 1$, which reflects the fact that the *average kissing number* (i.e., average number of contacting particles per particle) is effectively six for this collectively jammed packing, meaning that the packing is *isostatic*

[Donev et al. 05]. A positive average kissing number is required if the packing is constrained to be jammed, and in $\mathbb{R}^d$ this means that the average kissing number is $2d$ for either collective or strict jamming [Donev et al. 05]. Isostatic packings are jammed packings with the minimum number of contacts for a particular jamming category. According to the decorrelation principle, as d tends to infinity, g_2 for a maximally random jammed packing would retain this delta-function contribution but the unconstrained spatial correlations beyond $r = 1$ would vanish. Of course, the manner in which the g_2 shown in Figure 4 approaches the asymptotic limit of a step function $\Theta(r-1)$ plus a delta-function contribution at $r = 1$ is crucial. We note that maximally random jammed packings contain about 2%–3% *rattler* spheres, i.e., spheres trapped in a cage of jammed neighbors but free to move within the cage.

5.   A NEW APPROACH TO LOWER BOUNDS

The salient ideas behind our new approach to the derivation of lower bounds on $\phi_{\max}$ were actually laid out in our earlier work [Torquato and Stillinger 02]. The main objective of that work was to study sphere packings in three dimensions in which long-range order was suppressed and short-range order was controlled (i.e., disordered sphere packings in $\Re^3$) using so-called g_2-invariant processes. A g_2-*invariant process* is one in which a given nonnegative pair correlation $g_2(\mathbf{r})$ function remains invariant as the density varies, for all $\mathbf{r}$, over the range of densities

$$0 \leq \phi \leq \phi_*.$$

The terminal density ϕ_* is the maximum achievable density for the g_2-invariant process subject to satisfaction of the structure-factor inequality (3–19). A five-parameter test family of g_2's had been considered, which incorporated the known features of core exclusion, contact pairs, and damped oscillatory short-range order beyond contact. The problem of finding the maximal packing fraction ϕ_* was posed as an optimization problem: maximize ϕ over the set of parameters subject to the constraints (3–18) and (3–19). We noted in passing that when the damped-oscillatory contribution to g_2 was set equal to zero, the optimization problem could be solved analytically for all space dimensions, leading to a terminal density $\phi_* = (d+2)/2^{d+1}$. Under the assumption that such a g_2 was a realizable packing, we also observed that this ϕ_* was a lower bound on the maximal density for any sphere packing [i.e., $\phi_{\max} \geq (d+2)/2^{d+1}$] because the terminal density would have been higher by including the

damped-oscillatory contribution to g_2. This conjectural lower bound was noted to provide linear improvement over Minkowski's lower bound, but we were not aware of Ball's similar lower bound [Ball 92] at the time. Since our original 2002 paper, we also learned about other necessary conditions for the realizability of a point process for a given number density ρ and g_2, such as Yamada's condition (3–20). In any event, our brief remarks about lower bounds on sphere packings were not intended to be mathematically rigorous.

It is our intent here to make our optimization methodology to obtain lower bounds on $\phi_{\max}$ more mathematically precise, especially in light of recent developments and the considerations of the previous two sections. We then apply the optimization procedure to provide alternative derivations of previous lower bounds as well as a new bound.

We will consider those "test" $g_2(r)$'s that are distributions on $\mathbb{R}^d$ depending only on the radial distance r such that $h(r) = g_2(r) - 1$. For any test $g_2(r)$, we want to maximize the corresponding density ϕ satisfying the following three conditions:

(i.) $g_2(r) \geq 0$ for all r,

(ii.) $\mathrm{supp}(g_2) \subseteq \{r : r \geq 1\}$,

(iii.)

$$S(k) = 1 + \rho \, (2\pi)^{d/2} \int_0^\infty r^{d-1}[g_2(r) - 1]$$
$$\times \frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}} \, dr$$

is greater than or equal to zero for all k.

We will call the maximum density the *terminal density* and denote it by ϕ_*.

Remark 5.1. The conditions (i)–(iii) are just recapitulations of (3–14), (3–18), and (3–19) for this class of test functions. We will call condition (ii) the *hard-core* constraint.

Remark 5.2. When there exist sphere packings with g_2 satisfying conditions (i)–(iii) for ϕ in the interval $[0, \phi_*]$, then we have the lower bound on the maximal density given by

$$\phi_{\max} \geq \phi_*. \tag{5–1}$$

The best lower bound would be obtained if one could probe the entire class of test functions. In practice, we will consider here only a small subset of test functions and

in particular those that are amenable to exact asymptotic analysis. In some instances, we will associate with the terminal density ϕ_* an optimized *average kissing number* Z_*. Thus, whenever inequality (5–1) applies, the *maximal kissing number* $Z_{\max}$ is bounded from below by Z_*, i.e.,

$$Z_{\max} \geq Z_*. \tag{5–2}$$

In the next subsection, we put forth a conjecture that states when the conditions (i)–(iii) are necessary and sufficient for the existence of disordered sphere packings.

Remark 5.3. Remarkably, the optimization problem defined above is identical to one formulated by Cohn [Cohn 02]. In particular, it is the dual of the primal infinite-dimensional linear program that Cohn employed with Elkies [Cohn and Elkies 03] to obtain upper bounds on the maximal packing density. One need only replace $S(k)$ with $\hat{g} - c\delta(k)$, where c plays the role of number density, g is a tempered distribution, and $\hat{g}$ is its Fourier transform in Cohn's notation. Thus, even if there does not exist a sphere packing with g_2 satisfying conditions (i)–(iii), our formulation has implications for upper bounds on $\phi_{\max}$, which we discuss in Section 6. For finite-dimensional linear programs (and many infinite-dimensional ones) there is no "duality gap," i.e., the optima of the primal and dual programs are equal. However, in this infinite-dimensional setting, it is not clear how to prove that there is no duality gap [Cohn 02]. Therefore, it is rigorously true that the terminal density ϕ_* can never exceed the Cohn–Elkies upper bound, which is a desirable feature of our formulation, for otherwise, the terminal density could never correspond to a rigorous lower bound.

We will show that for the test radial distribution functions considered in this paper, Yamada's condition, inequality (3–20), is relevant in only one dimension, and even then in just some cases. A remark about the Yamada condition for sphere packings is in order here. In earlier work [Torquato and Stillinger 03], we observed that for any sphere packing of congruent spheres, the number variance for a spherical window of radius R defined by (3–21) obeys the lower bound

$$\sigma^2(R) \geq 2^d \phi R^d \left[1 - 2^d \phi R^d\right] \tag{5–3}$$

for any R. This is a tight bound for sufficiently small R and is exact for $R \leq \frac{1}{2}$. However, we note here that if $2^d \phi R^d \leq 1$, the Yamada lower bound (3–20) and lower bound (5–3) are identical. Thus, the Yamada lower bound for any sphere packing needs to be checked only

for $R > R_0$, where

$$R_0 = \frac{1}{2\phi^{1/d}}. \qquad (5\text{--}4)$$

5.1 Existence of Disordered Packings in High Dimensions

We have seen that a necessary condition for the existence of a translationally invariant point process with a specified positive ρ and nonnegative g_2 is that $S(\mathbf{k})$ be nonnegative [cf. (3–19)]. In other words, given ρ and g_2, it does not mean that there are some higher-order functions $g_3, g_4, \dots$ for which these one- and two-point correlation functions hold. The function g_2 specifies how frequently pair distances of a given length occur statistically in $\mathbb{R}^d$. The third-order function g_3 reveals how these pair separations are linked into triangles. This additional information generally cannot be inferred from the knowledge of ρ and g_2 alone, however. The fourth-order function g_4 controls the assembly of triangles into tetrahedra (and is the lowest-order correlation function that is sensitive to chirality) but g_4 cannot be determined by knowing only ρ, g_2, and g_3. In general, g_n for any $n \geq 3$ is not completely determined from a knowledge of the lower-order correlation functions alone. This is to be contrasted with general stochastic processes in which nonnegativity of first- and second-order statistics (mean and autocovariance) are necessary and sufficient to establish existence because one can always find a Gaussian process with such given first- and second-order statistics. For a Gaussian process, first- and second-order statistics determine all of the high-order statistics.

There are a number of results that suggest that it is reasonable to conclude that the generally necessary nonnegativity conditions for the existence of a disordered sphere packing become necessary and sufficient for sufficiently large d. First, the decorrelation principle of the previous section states that unconstrained correlations in disordered sphere packings vanish asymptotically in high dimensions and that the g_n for any $n \geq 3$ can be inferred entirely from a knowledge of ρ and g_2. Second, as we noted in Section 4, the necessary Yamada condition appears to have relevance only in very low dimensions. Third, we will demonstrate below that other new necessary conditions also seem to be germane only in very low dimensions. Fourth, we will describe numerical constructions of configurations of disordered sphere packings on the torus corresponding to certain test radial distribution functions in low dimensions for densities up to the terminal density. Finally, we will show that certain test radial distribution functions recover the asymptotic

forms of known rigorous bounds. In light of these results, we propose the following conjecture:

Conjecture 5.4. *For sufficiently large d, a hard-core nonnegative tempered distribution $g_2(\mathbf{r})$ that satisfies $g_2(\mathbf{r}) = 1 + \mathcal{O}(|\mathbf{r}|^{-d-\varepsilon})$ as $|\mathbf{r}| \to \infty$ for some $\varepsilon > 0$ is a pair correlation function of a translationally invariant disordered sphere packing in $\mathbb{R}^d$ at number density ρ if and only if $S(\mathbf{k}) \equiv 1 + \rho\tilde{h}(\mathbf{k}) \geq 0$. The maximum achievable density is the terminal density ϕ_*.*

Remark 5.5. A weaker form of this conjecture would replace the phrase "for sufficiently large d" with "in the limit $d \to \infty$."

Remark 5.6. Employing the aforementioned optimization procedure with a certain test function g_2 and this conjecture, we obtain in what follows conjectural lower bounds that yield the long-sought asymptotic exponential improvement on Minkowski's bound. Before obtaining this result, we first apply the procedure to two simpler test functions that we examined in the past.

5.2 Step Function

The simplest possible choice for a radial distribution function corresponding to a disordered packing is the following unit step function:

$$g_2(r) = \Theta(r - 1). \qquad (5\text{--}5)$$

This states that all pair distances beyond the hard-core diameter are equally probable, i.e., spatial correlations vanish identically. The corresponding structure factor [cf. condition (iii)] for this test function in any dimension d is given by [Torquato and Stillinger 02]

$$S(k) = 1 - \frac{\phi 2^{3\nu}\Gamma(1+\nu)}{k^\nu} J_\nu(k),$$

where $\nu = d/2$. Since there are no parameters to be optimized here, the terminal density ϕ_* is readily obtained by determining the highest density for which the condition (3–19) is satisfied, yielding

$$\phi_* = \frac{1}{2^d}. \qquad (5\text{--}6)$$

Now we show that the Yamada condition (3–20) is satisfied in any dimension for $0 \leq \phi \leq 2^{-d}$. Consider the more general class of radial distribution functions:

$$0 \leq g_2(r) \leq 1 \qquad \text{for } r > 1. \qquad (5\text{--}7)$$

The test function (5–5) belongs to this class. Note that for any dimension, the scaled intersection volume given by (3–24) obeys the inequality

$$\alpha_2(r; R) \leq 1 - \frac{r}{2R} \qquad \text{for } 0 \leq r \leq 2R, \qquad (5\text{–}8)$$

where the equality applies when $d = 1$. For $g_2(r)$ satisfying (5–7), relation (3–21) and inequality (5–8) yield the following lower bound for any d:

$$\sigma^2(R) \geq 2^d \phi R^d \left[1 + d2^d \phi \int_0^{2R} r^{d-1} h(r) \left[1 - \frac{r}{2R} \right] dr \right]. \qquad (5\text{–}9)$$

At $\phi = 1/2^d$, the lower bound (5–9) for the test function (5–5) is given by

$$\sigma^2(R) \geq \frac{d}{2(d+1)} R^{d-1}$$

and because $R_0 = 1$ [cf. (5–4)], we only need to consider $R > 1$. In particular, the right side of this inequality is smallest at $R = 1$, so that

$$\sigma^2(R) \geq \frac{d}{2(d+1)}.$$

Since $\sigma^2(R) \geq \frac{1}{4}$ for $d \geq 1$, Yamada's condition is satisfied for all R for the step function (5–5) at $\phi = 1/2^d$ as well as all $\phi < 1/2^d$.

We already established in Section 4 that there exist sphere packings that asymptotically have radial distribution functions given by the simple unit step function (5–5) for $\phi \leq 2^{-d}$. Nonetheless, invoking Conjecture 5.4 and terminal density specified by (5–6) implies that the asymptotic lower bound on the maximal density is given by

$$\phi_{\max} \geq \frac{1}{2^d},$$

which provides an alternative derivation of the elementary bound (2–2).

Using numerical simulations with a finite but large number of spheres on the torus, we have been able to construct particle configurations in which the radial distribution function (sampled at discretized pair distances) is given by the test function (5–5) in one, two, and three dimensions for densities up to the terminal density [Crawford et al. 03, Uche et al. 06]. The existence of such a discrete approximation to (5–5) of course is not conclusive proof of the existence of such packings in low dimensions, but it is suggestive that the standard nonnegativity conditions may be sufficient to establish existence in this case for densities up to ϕ_*.

5.3 Step Plus Delta Function

An important feature of any dense packing is that the particles form contacts with one another. Ideally, one would like to enforce strict jamming (see Section 1). The probability that a pair of particles form such contacts at the pair distance $r = 1$ for the test function (5–5) is strictly zero. Accordingly, let us now consider the test radial distribution function given by the previous test function plus a delta-function contribution as follows:

$$g_2(r) = \Theta(r - 1) + \frac{Z}{s_1(1)\rho} \delta(r - 1). \qquad (5\text{–}10)$$

Here $s_1(r)$ is the surface area of a d-dimensional sphere of radius r given by (3–13) and Z is a parameter, which is the average kissing number. Because we allow for interparticle contacts via the second term in (5–10), the terminal density is expected to be greater than 2^{-d}, which will indeed be the case. The corresponding structure factor [cf. (iii)] for this test function in any dimension d is given by [Torquato and Stillinger 02]

$$S(k) = 1 - \frac{\phi 2^{3\nu} \Gamma(1 + \nu)}{k^\nu} J_\nu(k) + \frac{Z 2^\nu \Gamma(1 + \nu)}{dk^{\nu-1}} J_{\nu-1}(k),$$

where $\nu = d/2$. The structure factor for small k can be expanded in a MacLaurin series as follows:

$$S(k) = 1 + (Z - 2^d \phi) + \left[\frac{2^{d-2}\phi}{1 + d/2} - \frac{Z}{2d} \right] k^2 + \mathcal{O}(k^4).$$

The last term changes sign if Z increases past $2^d \phi d/(d+2)$. At this crossover point,

$$S(k) = 1 - \frac{2^{d+1}}{d+2} \phi + \mathcal{O}(k^4).$$

Under the constraint that the minimum of $S(k)$ occurs at $k = 0$, we then have the exact results

$$\phi_* = \frac{d+2}{2^{d+1}}, \qquad Z_* = \frac{d}{2}. \qquad (5\text{–}11)$$

We see that at the terminal density, the average kissing number Z_* is equal to $d/2$, which does not even meet the local jamming criterion described in Section 1.

The Yamada condition (3–20) is violated only for $d = 1$ for the test function (5–10) at the terminal density specified by (5–11). It is easy to verify directly that the Yamada condition becomes less restrictive as the dimension increases from $d = 2$. Interestingly, we have also shown via numerical simulations that there exist sphere packings possessing radial distribution functions given by the test function (5–10) (in the discrete approximation)

in two and three dimensions for densities up to the terminal density [Uche et al. 06]. This is suggestive that Conjecture 5.4 for this test function may in fact be stronger than is required.

In the high-dimensional limit, we invoke Conjecture 5.4 and the terminal density given by (5–11), yielding the conjectural lower bound

$$\phi_{\max} \geq \frac{d+2}{2^{d+1}}. \qquad (5\text{--}12)$$

This lower bound provides the same type of linear improvement over Minkowski's lower bound as does Ball's lower bound [Ball 92].

5.4   Step Plus Delta Function with a Gap

The previous test function (5–10) provided an optimal average kissing number $Z_* = d/2$ that did not even meet the local jamming criterion. Experience with disordered jammed packings in low dimensions reveals that the kissing number as well as the density can be substantially increased if there is a low probability of finding noncontacting particles from a typical particle at radial distances just larger than the nearest-neighbor distance. This small-distance negative correlation is clearly manifested in the graph of $g_2(r)$ for the three-dimensional maximally random jammed packing (Figure 4) for values of r approximately between 1.1 and 1.5. We would like to idealize this small-distance negative correlation in such a way that it is amenable to exact asymptotic analysis. Accordingly, we consider a test radial distribution function that is similar to the previous one [cf. (5–10)] but one in which there is a gap between the location of the unit step function and the delta function at finite d, i.e.,

$$g_2(r) = \Theta(r - \sigma) + \frac{Z}{s_1(1)\rho}\delta(r - 1). \qquad (5\text{--}13)$$

The expression contains two adjustable parameters, $\sigma \geq 1$ and Z, which must obviously be constrained to be nonnegative. According to the decorrelation principle of Section 4, the location of the step function $r = \sigma$ must approach unity asymptotically, i.e., it must approach the previous test function (5–10). However, as we have emphasized, the manner in which the test function (5–13) approaches (5–10) is crucial. Indeed, we will see that the presence of a gap between the unit step function and delta function will indeed lead asymptotically to substantially higher terminal densities.

The structure factor is given by

$$S(k) = 1 - \frac{2^{3\nu}\phi\sigma^d\Gamma(1+\nu)}{(k\sigma)^\nu}J_\nu(k\sigma) + \frac{2^\nu Z\Gamma(1+\nu)}{dk^{\nu-1}}J_{\nu-1}(k). \qquad (5\text{--}14)$$

The goal now is to find the optimal values of the the adjustable nonnegative parameters Z and σ that maximize the density ϕ subject to the constraint (iii). This search in two-dimensional parameter space can be reduced by imposing the further condition that a minimum of the structure factor occur at $k = 0$. The MacLaurin expansion of expression (5–14) gives

$$S = 1 + [Z - (2\sigma)^d\phi] + \left[\frac{2^{d-1}\sigma^{d+2}\phi}{d+2} - \frac{Z}{2d}\right]k^2 + \mathcal{O}(k^4).$$

Requiring that a zero of $S(k)$ occur at the origin (hyperuniformity) such that the quadratic coefficient is nonnegative implies the restrictions

$$Z = (2\sigma)^d\phi - 1 \qquad (5\text{--}15)$$

and

$$(2\sigma)^d\phi[d\sigma^2 - (d+2)] + d + 2 \geq 0.$$

Combination of (5–14) and (5–15) yields the structure factor as

$$S(k) = 1 - c_1(d)\frac{J_\nu(k\sigma)}{(k\sigma)^\nu} + c_2(d)\frac{J_{\nu-1}(k)}{(k\sigma)^{\nu-1}}, \qquad (5\text{--}16)$$

where the d-dependent coefficients $c_1(d)$ and $c_2(d)$ are given by

$$c_1(d) = \phi\sigma^d 2^{3\nu}\Gamma(1+\nu) \qquad (5\text{--}17)$$

and

$$c_2(d) = [(2\sigma)^d\phi - 1]2^\nu\frac{\Gamma(1+\nu)}{d}. \qquad (5\text{--}18)$$

Now the problem reduces to finding the optimal value of the parameter $\sigma(d)$ as a function of the space dimension d that maximizes the density ϕ subject to the constraint (iii). It will be shown below that the optimal σ is of order unity and approaches unity in the limit $d \to \infty$. It immediately follows from (5–16) and the asymptotic properties of the Bessel functions of fixed order that $S(k) \to 1$ for $k \to \infty$.

In general, $S(k)$ will possess multiple minima, and thus we want to ensure that the values of $S(k)$ at each of these minima are all nonnegative. To find the minima of $S(k)$, we set its first derivative to zero, yielding the relation

$$\frac{c_1(d)}{\sigma^{\nu-1}}J_{\nu+1}(k\sigma) = c_2(d)kJ_\nu(k), \qquad (5\text{--}19)$$
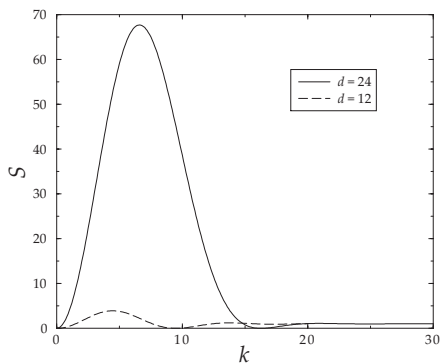
FIGURE 5. The optimized structure factor for $d = 12$ and $d = 24$.

where we have used the identity

$$\frac{d}{dx}\left[\frac{J_\nu(x)}{x^\nu}\right] = -\frac{J_{\nu+1}(x)}{x^\nu}.$$

For sufficiently small d ($d \leq 200$), the search procedure is carried out numerically using Maple, and is made more efficient by exploiting the fact that the minima of $S(k)$ occur at the real solutions of (5–19). Figure 5 shows the optimized structure factor for $d = 12$ and $d = 24$. Our numerical examination of $S(k)$ for a wide range of d values has consistently shown that the first minimum for positive k is the deepest one. Although we have not proven this rigorously, we assume that this is a general result.

We should note that the Yamada condition (3–20) is violated only for $d = 1$ for the test function (5–13) for the terminal density ϕ_* and associated optimized parameters σ_* and Z_* [calculated via (5–15)]. One can again verify directly that the Yamada condition becomes less restrictive as the dimension increases from $d = 2$. However, although the test function (5–13) for $d = 2$ with optimized parameters $\phi_* = 0.74803$, $\sigma_* = 1.2946$, and $Z_* = 4.0148$ satisfies the Yamada condition, it cannot correspond to a sphere packing because it violates local geometric constraints specified by σ_* and Z_*. Specifically, for an average kissing number of 4.0148, there must be particles that are in contact with at least five others. But no arrangement of the five exists that is consistent with the assumed pair correlation function (step plus delta function with a gap from 1 to 1.2946). Simple geometric considerations show that either some pairs of the five would be forced into the gap, or they would be restricted to fixed separations that would correspond to undesired delta functions beyond the gap. To our knowledge, this is the first example of a test radial distribution function that satisfies the

d	σ_*	Z_*	ϕ_*	$\frac{2^{d+1}\phi_*}{d+2}$
3	1.246997	7.932582	0.5758254	1.842641
4	1.212589	13.71016	0.4252472	2.267985
5	1.186929	21.97918	0.3048322	2.787037
6	1.167000	33.53884	0.2136444	3.418310
7	1.151106	49.42513	0.1471058	4.184343
8	1.137967	70.88348	0.09985085	5.112364
24	1.058992	5473.546	8.245251e-05	106.4095
36	1.041611	76521.15	2.566299e-07	928.1828
56	1.028036	4.248007e06	1.253255e-11	31140.19
60	1.026330	9.179315e06	1.674130e-12	62262.60
64	1.024823	1.968233e07	2.221414e-13	124175.32
80	1.020211	3.908042e08	6.521679e-17	1.922982e06
100	1.016421	1.478804e10	2.288485e-21	5.688234e08
125	1.013311	1.246172e12	5.610270e-27	3.758024e09
150	1.011214	9.698081e13	1.275632e-32	2.319290e11
175	1.009671	7.086019e15	2.745830e-38	1.485866e13
200	1.008510	4.959086e17	5.667098e-44	9.016510e14

TABLE 3. Optimized parameters σ_*, Z_*, and ϕ_*, and the ratio $\frac{2^{d+1}\phi_*}{d+2}$, which is the relative improvement of the terminal density over the gapless-test-function terminal density [cf. (5–11)].

two standard nonnegativity conditions (3–18) and (3–19) and the Yamada condition (3–20), but cannot correspond to a point process. Thus, there is at least one previously unarticulated necessary condition that has been violated in the low dimension $d = 2$.

In three dimensions one obtains $\phi_* = 0.5758254$, $\sigma_* = 1.246997$, and $Z_* = 7.932582$. The last of these requires that some nonzero fraction of the spheres have at least eight contacting neighbors. We have verified that valid arrangements of both eight and nine contacts are possible, thereby avoiding the analogue of the violation encountered in $d = 2$. As is the case with the Yamada condition (3–20), this additional necessary condition appears to lose relevance as d increases.

The terminal density ϕ_* and the associated optimized parameters σ_* and Z_* are listed in Table 3 for selected values of the space dimension between $d = 3$ and $d = 200$. Note that for $d \leq 56$, the terminal density lies below the density of the densest known packing. For $d = 56$, the densest arrangement is a lattice (designated by $L_{56,2}(M)$ [Nebe 98]) with density $\phi = 2.327670 \times 10^{-11}$, which is about twice as large as ϕ_*, as shown in the table. However, for $d > 56$, ϕ_* can be larger than the density of the densest known arrangement. For $d = 60$, the densest known packing is again a lattice (designated by $L_{56,2}(M)$ [Conway and Sloane 98]) with density $\phi = 2.966747 \times 10^{-13}$, which is about five times smaller than ϕ_*, as shown in the table. The next dimension for which data are available is $d = 64$, where

the densest known packing is the Ne_{64} lattice [Nebe 98] with density $\phi = 1.326615 \times 10^{-12}$, which is about six times larger than ϕ_*. The table also reveals exponential improvement of the terminal density ϕ_* over that for the gapless case, i.e., $\phi_* = (d+2)/2^{d+1}$. The crucial question is whether such exponential improvement persists in the high-dimensional limit.

To obtain an asymptotic expression for ϕ_* for large d, we use the fact that $(2\sigma)^d \phi \gg 1$, implying that $c_1(d)/c_2(d) \to d$ [cf. (5–17) and (5–18)]. Therefore, the minima of $S(k)$ for large d are the solutions of

$$\frac{J_{\nu+1}(k\sigma)}{\sigma^{\nu-1}} = \frac{k}{d}J_\nu(k). \qquad (5–20)$$

We see that the locations of the minima depend only on σ (not on ϕ). The deepest minimum of $S(k)$, after the one at $k = 0$, is a zero and occurs at the wave number $k = k_{\min}$. (This characteristic is true in any dimension; see Figure 5.) Therefore, $S(k = k_{\min}) = 0$, $c_2(d) = c_1(d)/d$, and relation (5–14) gives the condition

$$\frac{c_1(d)}{k_{\min}^\nu}\Delta_\nu(k_{\min}) = 1, \qquad (5–21)$$

where

$$\Delta_\nu(k_{\min}) = \frac{J_\nu(k_{\min}\sigma)}{\sigma^\nu} - k_{\min}\frac{J_{\nu-1}(k_{\min})}{d}. \qquad (5–22)$$

The solution of equation (5–21) produces the desired optimal values of σ_* and ϕ_*, where

$$\phi_* = \frac{k_{\min}^\nu}{2^{3\nu}\Gamma(1+\nu)\sigma_*^{2\nu}\Delta_\nu(k_{\min})}. \qquad (5–23)$$

We find the solutions of (5–20) by linearizing each Bessel function in (5–20) around its respective first positive zero, i.e.,

$$J_\nu(x) = \beta_1(x_0)(x - x_0) + \mathcal{O}\left((x - x_0)^2\right),$$
$$J_{\nu+1}(x) = \beta_2(y_0)(x - y_0) + \mathcal{O}\left((x - y_0)^2\right),$$

where

$$\beta_1(x_0) = \frac{1}{2}[J_{\nu-1}(x_0) - J_{\nu+1}(x_0)], \qquad (5–24)$$

$$\beta_2(y_0) = \frac{1}{2}[J_\nu(y_0) - J_{\nu+2}(y_0)], \qquad (5–25)$$

and x_0 and y_0 denote the locations of the first positive zeros of $J_\nu(z)$ and $J_{\nu+1}(z)$, respectively. Similarly, we employ the linearized form

$$xJ_\nu(x) = x_0\beta_1(x_0)(x - x_0) + \mathcal{O}\left((x - x_0)^2\right).$$

Use of these relations in (5–20) yields the following equation for $k_{\min}$:

$$k_{\min} \approx x_0 - \frac{d(y_0 - \sigma x_0)}{\frac{\beta_1}{\beta_2}\sigma^{\nu-1}x_0 - d\sigma}. \qquad (5–26)$$

This formula provides an excellent approximation for $k_{\min}$. For example, for $d = 200$ (or $\nu = 100$), substitution of the exact values $x_0 = 108.8361659$, $y_0 = 109.8640469$, and $\beta_1/\beta_2 = 1.003189733$ as well as the numerical search solution $\sigma_* = 1.008510$ into this formula predicts $k_{\min} = 108.4368917$. This value is to be compared to the numerical search solution of $k_{\min} = 108.4395$. This supports the fact that the higher-order terms in the aforementioned linearized forms of the Bessel functions are negligibly small. Indeed, we expect that this can be rigorously proved, but we shall not do so here. We will assume the validity of the linearized forms in the asymptotics displayed below.

For large $d = 2\nu$, we make use of the asymptotic formulas

$$x_0 = \nu + a_1\nu^{1/3} + \frac{a_2}{\nu^{1/3}} + \frac{a_3}{\nu} + \mathcal{O}\left(\frac{1}{\nu^{5/3}}\right),$$

$$y_0 = \nu + a_1\nu^{1/3} + 1 + \frac{a_2}{\nu^{1/3}} + \frac{a_2}{3\nu^{2/3}} + \frac{a_3}{\nu} + \mathcal{O}\left(\frac{1}{\nu^{4/3}}\right),$$

where the constants a_1, a_2, and a_3 are explicitly given in the appendix, Section 7. For $d = 200$, these formulas predict $x_0 = 108.8362067$ and $y_0 = 109.8640871$, which are in excellent agreement with the exact values reported in the preceding paragraph. Using the asymptotic results given in the appendix, we obtain that

$$\frac{\beta_1}{\beta_2} = 1 + \frac{2}{3\nu} - \frac{2C_2}{3C_1\nu^{5/3}} + \mathcal{O}\left(\frac{1}{\nu^2}\right),$$

where the constants C_1 and C_2 are given explicitly in terms of the constants a_1 and a_2 in the appendix. For $d = 200$, for example, this formula together with (7–7) provides the estimate $\beta_1/\beta_2 = 1.007122331$, which is to be compared to the exact result $\beta_1/\beta_2 = 1.006215695$.

The optimized asymptotic form for σ_* is obtained by taking the derivative of both sides of the zero condition (5–21) with respect to σ and solving for σ using relation (5–26) for $k_{\min}$. We obtain that

$$\sigma_* = 1 + \frac{q_1}{\nu} + \frac{q_2}{\nu^{5/3}} + \mathcal{O}\left(\frac{1}{\nu^2}\right), \qquad (5–27)$$

where

$$q_1 = 0.90763589355\ldots \qquad (5–28)$$

is the unique positive root of $xe^x + e^{2x} - 5e^x + 4 = 0$ and

$$q_2 = \frac{a_1(8e^{q_1} - 2q_1e^{q_1} - 10e^{2q_1} + 4 + e^{3q_1} + 4q_1e^{2q_1})}{3e^{q_1}(2q_1e^{q_1} - 2q_1 + 12 + 3e^{2q_1} - 13e^{q_1})}$$
$$= -1.279349474. \qquad (5\text{–}29)$$

Therefore, expression (5–26) for $k_{\min}$ has the asymptotic form

$$k_{\min} = \nu + a_1\nu^{1/3} + Q_1 + \frac{a_2}{\nu^{1/3}} + \mathcal{O}\left(\frac{1}{\nu^{2/3}}\right), \quad (5\text{–}30)$$

where

$$Q_1 = \frac{2(q_1 - 1)}{e^{q_1} - 2} = -0.3860921576. \qquad (5\text{–}31)$$

These formulas predict $\sigma_* = 1.008482538$ and $k_{\min} = 108.4501542$, which again are in excellent agreement with values reported above.

Linearizing each Bessel function appearing in (5–22) about its first positive zero and using the results of the appendix yields

$$\Delta_\nu(k_{\min}) \approx \frac{\beta_1(x_0)(k_{\min}\sigma_* - x_0)}{\sigma_*^\nu}$$
$$- \frac{\beta_3(z_0)k_{\min}(k_{\min} - z_0)}{2\nu},$$

where $\beta_1(x_0)$ is given by (5–24), $\beta_3(z_0) = [J_{\nu-2}(z_0) - J_\nu(z_0)]/2$, and z_0 is the first positive zero of J_{nu-1}. Using relations (5–27) and (5–30) and the results of the appendix yields the asymptotic expansion of $\Delta_\nu(k_{\min})$:

$$\Delta_\nu(k_{\min}) = \frac{D_1}{\nu^{2/3}} + \frac{D_2}{\nu^{4/3}} + \mathcal{O}\left(\frac{1}{\nu^{5/3}}\right),$$

where

$$D_1 = \frac{C_1(2 - e^{q_1})}{2e^{q_1}},$$
$$D_2 = \frac{C_1\left[a_1(2e^{q_1} + 6q_1e^{-q_1} - 7) + 3q_2(q_1 - 1)\right]}{3(2 - e^{q_1})} + \frac{C_2 D_1}{C_1}.$$
$$(5\text{–}32)$$

Note also that

$$\left(\frac{k_{\min}}{\nu}\right)^\nu = e^{a_1\nu^{1/3}+Q_1}\left[1 + \frac{E_1}{\nu^{1/3}} + \frac{E_2}{\nu^{2/3}} + \mathcal{O}\left(\frac{1}{\nu}\right)\right]$$

and

$$\sigma_*^{2\nu} = e^{2q_1}\left[1 + \frac{2q_2}{\nu^{2/3}} - \frac{q_1^2}{\nu} + \frac{2q_2^2}{\nu^{4/3}} + \mathcal{O}\left(\frac{1}{\nu^{5/3}}\right)\right],$$

where

$$E_1 = a_2 - \frac{a_1^2}{2}, \quad E_2 = -Q_1 a_1 + \frac{a_1^4 - 4a_1^2 a_2 + 4a_2^2}{8}, \qquad (5\text{–}33)$$

and Q_1 is given by (5–31). For $d = 200$, these formulas [together with the constants specified by equations (5–28), (5–29), (5–32), (5–33), (7–3), and (7–7)] predict $\Delta_\nu(k_{\min}) = 0.00567441932$, $(k_{\min}/\nu)^\nu = 3353.018128$, and $\sigma_*^{2\nu} = 5.405924156$. These values should be compared to the exact value of $\Delta_\nu(k_{\min}) = 0.00559813885$, $(k_{\min}/\nu)^\nu = 3301.799093$, and $\sigma_*^{2\nu} = 5.445550297$.

Thus, substituting the asymptotic relations above into the optimal expression (5–23) for the density and invoking Conjecture 5.4 yields the conjectural lower bound

$$\phi_{\max} \geq \phi_* \qquad (5\text{–}34)$$
$$= \frac{1}{2^{[3-\log_2(e)]\nu - \log_2(e)a_1\nu^{1/3} + (2q_1 - Q_1)\log_2(e)}}$$
$$\times \left[\frac{1}{2D_1}\sqrt{\frac{2}{\pi}}\left[\nu^{1/6} + \frac{E_1}{\nu^{1/6}} + \frac{E_2 - 2q_2 - D_2/D_1}{\nu^{1/2}}\right.\right.$$
$$\left.\left. + \mathcal{O}\left(\frac{1}{\nu^{5/6}}\right)\right]\right],$$

where we have used the asymptotic relation $\Gamma(1 + \nu) \sim \nu^\nu\sqrt{2\pi\nu}e^{-\nu}$. For $d = 200$, this asymptotic formula, together with the constants specified by equations (5–28), (5–29), (5–32), (5–33), (7–3), and (7–7), predicts $\phi_* = 5.626727001 \times 10^{-44}$, which is in good agreement with the numerical search solution of $\phi_* = 5.667098 \times 10^{-44}$. Note also that the formula (5–23) with $k_{\min}$ estimated from (5–26) yields $\phi_* = 5.666392126 \times 10^{-44}$, which is remarkably close to the numerical solution. For large d, result (5–34) yields the following dominant asymptotic formula for the conjectural lower bound on $\phi_{\max}$:

$$\phi_{\max} \geq \phi_* \sim \frac{d^{1/6}}{2^{2/3}D_1\sqrt{\pi}\,2^{[3-\log_2(e)]d/2}}$$
$$= \frac{3.276100896\,d^{1/6}}{2^{0.7786524795\ldots d}}. \qquad (5\text{–}35)$$

This putatively provides the long-sought exponential improvement on Minkowski's lower bound. Note that the constant $D_1 = 0.1084878572$ appearing in (5–35) is determined from the appropriate relation in (5–32) using the value for q_1 given by (5–28) and the more refined estimate of C_1 given by (7–9).

Substitution of the asymptotic expression (5–35) into (5–15) and use of (5–2) yields a conjectural lower bound

on the maximal kissing number

$$Z_{\max} \geq Z_* \sim \frac{2^{1/3} e^{2q_1} d^{1/6}}{D_1 \sqrt{\pi}} 2^{[\log_2(e)-1]d/2}$$
$$= 40.24850787 \, d^{1/6} \, 2^{0.2213475205\ldots d},$$

which applies for large d. This result is superior to the best known asymptotic lower bound on the maximal kissing number of $2^{0.2075\ldots d}$ [Wyner 65]. Note that such a disordered packing would be substantially *hyperstatic* (the average kissing number is greater than $2d$ [Donev et al. 05]) and therefore would be appreciably different from a maximally random jammed packing [Torquato et al. 00, Torquato 02], which is isostatic (see Section 4.3) and hence significantly smaller in density.

6. DISCUSSION

Our results have immediate implications for the linear programming bounds of Cohn and Elkies [Cohn and Elkies 03], regardless of the validity of Conjecture 5.4. As we noted earlier, our optimization procedure is precisely the dual of the their primal linear programming upper bound. The existence of our test functions (5–10) and (5–13), which satisfy conditions (i), (ii), (iii) of Section 5 for densities up to the terminal density ϕ_*, narrows the duality gap; cf. (5–11) and (5–34). In particular, inequality (5–34) provides the greatest lower bound known for the dual linear program. Moreover, the existence of the inequalities (5–12) and (5–34) proves that linear programming bounds cannot possibly match the Minkowski lower bound for any dimension d. Finally, this link to the Cohn–Elkies formulation proves that the terminal density ϕ_* can never exceed the Cohn–Elkies upper bound, which obviously must be true if the terminal density corresponds to a rigorous lower bound.

Conjecture 5.4, concerning the existence of disordered sphere packings, is plausible for a number of reasons: (i) the decorrelation principle of Section 4.3; (ii) the necessary Yamada condition appears to have relevance only in very low dimensions; (iii) other new necessary conditions described in Section 5.4 also seem to be germane only in very low dimensions; (iv) there are numerical constructions of configurations of disordered sphere packings on the torus corresponding to these test radial distribution functions in low dimensions for densities up to the terminal density [Crawford et al. 03, Uche et al. 06]; and (v) the test radial distribution functions (5–5) and (5–10) recover the asymptotic forms of known rigorous bounds. Concerning the latter point, if Conjecture 5.4 is false, it is certainly not revealed by the results produced by

the test functions (5–5) and (5–10) because the forms of known rigorous results, obtained using completely different techniques, are recovered. If Conjecture 5.4 is false, why would it suddenly be revealed by the introduction of a gap in the test radial distribution function (cf. (5–13)) relative to (5–10)? This would seem to be unlikely and lends credibility to the conjecture in our view.

Conjecture 5.4, the particular choice (5–13), and our optimization procedure lead to a lower bound on the maximal density that improves on the Minkowski bound by an exponential factor. Our results suggest that the densest packings in sufficiently high dimensions may be disordered rather than periodic, implying the existence of disordered classical ground states for some continuous potentials. A byproduct of our procedure is an associated lower bound on the maximal kissing number, which is superior to the currently best known result. By no means is the choice (5–13) optimal. For example, one may be able to improve our putative lower bound by allowing the test function to be some positive function smaller than unity for $1 \leq r \leq \sigma$. Of course, it would be desirable to choose test functions that make asymptotic analysis exactly rather than numerically tractable.

Our putative exponential improvement over Minkowksi's bound is striking and should provide some impetus to determine the validity of Conjecture 5.4. As a first step in this direction, it would be fruitful if one could show that for sufficiently small densities, the two standard nonnegativity conditions on the pair correlation function g_2 are sufficient to ensure the existence of a point process, whether it is a sphere packing or not. Another problem worth pursuing is the demonstration of the existence of a pair interaction potential in $\mathbb{R}^d$ corresponding to a sphere packing for a given ρ and g_2 provided that ρ and g_2 are sufficiently small. Such a proof may be possible following the methods that Koralov [Koralov 05] used for the lattice setting. It would also be profitable to pursue the construction of disordered sphere packings with densities that exceed $1/2^d$ for sufficiently large d.

7. APPENDIX

An asymptotic expression for $J_\nu(x)$ when ν is large and $x > \nu$ is given by [Watson 58]

$$J_\nu(x) = A_\nu(x) \left[\cos[\omega_\nu(x) - \frac{\pi}{4} + \mathcal{O}\left(\frac{3x^2 + 2\nu^2}{12(x^2 - \nu^2)} \right) \right],$$
$$(7\text{–}1)$$

where

$$A_\nu(x) = \left[\frac{2}{\pi\sqrt{x^2 - \nu^2}} \right]^{1/2}$$

and

$$\omega_\nu(x) = \sqrt{x^2 - \nu^2} - \nu\cos^{-1}(\nu/x).$$

The function $A_\nu(x)\cos[\omega_\nu(x) - \pi/4]$ in (7–1) actually represents the dominant term in the asymptotic expansion of [Watson 58, p. 244 , equation (4)] for $J_\nu(x)$ when ν is large and $x > \nu$, and $A_\nu(x)$ multiplied by the big-$\mathcal{O}$ term represents the largest absolute error when this dominant term is used to estimate $J_\nu(x)$. A problem of central concern is an estimate of $J_\nu(x)$ in the vicinity of its first positive zero x_0 when ν is large. The first positive zero has the asymptotic expansion [Olver 60]

$$x_0 = \nu + a_1\nu^{1/3} + \frac{a_2}{\nu^{1/3}} + \frac{a_3}{\nu} + \mathcal{O}\left(\frac{1}{\nu^{5/3}}\right), \qquad (7\text{–}2)$$

where

$$a_1 = 1.8557571\ldots, \quad a_2 = 1.033150\ldots,$$
$$a_3 = -0.003971\ldots. \qquad (7\text{–}3)$$

Expanding $J_\nu(x)$ in a Taylor series about $x = x_0$ and neglecting quadratic and higher-order terms gives the linear estimate

$$J_\nu(x) \approx \frac{1}{2}[J_{\nu-1}(x_0) - J_{\nu+1}(x_0)](x - x_0),$$

where we take the Bessel functions on the right side to be given by the asymptotic forms

$$J_{\nu+1}(x_0) = A_{\nu+1}(x_0)\left[\cos[\omega_{\nu+1}(x_0) - \pi/4] \right.$$
$$\left. + \mathcal{O}\left(\frac{3x^2 + 2\nu^2}{12(x^2 - \nu^2)}\right) \right]$$

and

$$J_{\nu-1}(x_0) = A_{\nu-1}(x_0)\left[\cos[\omega_{\nu-1}(x_0) - \pi/4] \right.$$
$$\left. + \mathcal{O}\left(\frac{3x^2 + 2\nu^2}{12(x^2 - \nu^2)}\right) \right].$$

We will also need to consider the related functions

$$J_{\nu+1}(x) \approx \frac{1}{2}[J_\nu(y_0) - J_{\nu+2}(y_0)](x - y_0),$$
$$J_{\nu-1}(x) \approx \frac{1}{2}[J_{\nu-2}(z_0) - J_\nu(z_0)](x - z_0),$$

where y_0 and z_0 are the first positive zeros of $J_{\nu+1}(x)$ and $J_{\nu-1}(x)$, respectively, which are asymptotically given by

$$y_0 = \nu + a_1\nu^{1/3} + 1 + \frac{a_2}{\nu^{1/3}} + \frac{a_2}{3\nu^{2/3}} + \frac{a_3}{\nu} + \mathcal{O}\left(\frac{1}{\nu^{4/3}}\right),$$
$$z_0 = \nu + a_1\nu^{1/3} - 1 + \frac{a_2}{\nu^{1/3}} - \frac{a_2}{3\nu^{2/3}} + \frac{a_3}{\nu} + \mathcal{O}\left(\frac{1}{\nu^{4/3}}\right).$$

Note that the asymptotic expressions for the zeros given here for $\nu = 100$ ($d = 200$) predict $x_0 = 108.8362071$, $y_0 = 109.8641774$, and $z_0 = 107.8082369$, which are in excellent agreement with exact results $x_0 = 108.8361659$, $y_0 = 109.8640469$, and $z_0 = 107.8081033$ obtained from Maple.

Using Maple and the results above, we obtain the following asymptotic expansions:

$$\frac{1}{2}[J_{\nu-1}(x_0) - J_{\nu+1}(x_0)] = \frac{C_1}{\nu^{2/3}} + \frac{C_2}{\nu^{4/3}}$$
$$+ \mathcal{O}\left(\frac{1}{\nu^2}\right),$$

$$\frac{1}{2}[J_\nu(y_0) - J_{\nu+2}(y_0)] = \frac{C_1}{\nu^{2/3}} + \frac{C_2}{\nu^{4/3}} - \frac{2C_1}{3\nu^{5/3}}$$
$$+ \mathcal{O}\left(\frac{1}{\nu^2}\right), \qquad (7\text{–}4)$$

$$\frac{1}{2}[J_{\nu-2}(z_0) - J_\nu(z_0)] = \frac{C_1}{\nu^{2/3}} + \frac{C_2}{\nu^{4/3}} + \frac{2C_1}{3\nu^{5/3}}$$
$$+ \mathcal{O}\left(\frac{1}{\nu^2}\right),$$

where

$$C_1 = -\frac{2^{1/4}\left[\sqrt{2}f_1(a_1) + 8a_1^{3/2}f_2(a_1) \right]}{8\sqrt{\pi}a_1^{5/4}}, \qquad (7\text{–}5)$$

$$C_2 = \frac{1}{3840\sqrt{\pi}a_1^{13/4}} \qquad (7\text{–}6)$$
$$\times \left[2^{3/4}\left[1152a_1^6 - 3840a_1^4a_2 - 180a_1^3 \right. \right.$$
$$\left. + 600a_1a_2 - 225\right]f_1(a_1)$$
$$\left. + 2^{1/4}\left[3072a_1^{9/2} - 200a_1^{3/2}\right]f_2(a_1) \right],$$

and

$$f_1(a_1) = \sin\left(\frac{(2a_1)^{3/2}}{3}\right) + \cos\left(\frac{(2a_1)^{3/2}}{3}\right),$$
$$f_2(a_1) = \sin\left(\frac{(2a_1)^{3/2}}{3}\right) - \cos\left(\frac{(2a_1)^{3/2}}{3}\right).$$

Thus, substitution of the values for the constants a_1 and a_2 into the expressions above yields the estimates

$$C_1 = -1.104938082, \qquad C_2 = 1.627074727. \qquad (7\text{–}7)$$

For $d = 200$ ($\nu = 100$), for example, the asymptotic expansions (7–4) predict -0.04778125640, -0.04743934518 and -0.04812316762, respectively. This is to compared to the corresponding exact results: -0.04829366129, -0.04799533693 and -0.04859672879. Note that although the estimates of the constants C_1 and C_2 given

by (7–7) involve a small error due to our use of only the dominant asymptotic term (7–1), the functional forms of the asymptotic expansions (7–4) are exact.

We can show that the exact expressions for the constants C_1 and C_2 appearing in (7–4) are rapidly converging asymptotic expansions in inverse powers of the constant a_1 appearing in (7–2). For example, using the expansion of [Watson 58, p. 244 , equation (4)], we find that the first three terms of the expansion of C_1 are given by

$$C_1 = \frac{C_{11}}{a_1^{5/4}} + \frac{C_{12}}{a_1^{11/4}} + \frac{C_{13}}{a_1^{17/4}} + \mathcal{O}\left(\frac{1}{a_1^{23/4}}\right), \qquad (7\text{–}8)$$

where

$$C_{11} = -\frac{2^{1/4}\left[\sqrt{2}f_1(a_1) + 8a_1^{3/2}f_2(a_1)\right]}{8\sqrt{\pi}},$$

$$C_{12} = \frac{5 \cdot 2^{1/4}\left[4\sqrt{2}f_1(a_1) - 7a_1^{3/2}f_2(a_1)\right]}{384\sqrt{\pi}},$$

$$C_{13} = -\frac{385 \cdot 2^{1/4}\left[13\sqrt{2}f_1(a_1) + 8a_1^{3/2}f_2(a_1)\right]}{221184\sqrt{\pi}}.$$

The first term of the expansion (7–8) is the dominant one and is identical to the estimate given in (7–5). The first term of (7–8) is about 66 times larger than the second term in absolute value and the second term is about 7 times larger than the third term in absolute value. Substitution of the constant a_1 into (7–8) yields the more refined estimate

$$C_1 = -1.123958144. \qquad (7\text{–}9)$$

This refined estimate differs from the dominant first-term estimate given in (7–7) in the third significant figure. One could continue correcting this estimate by including additional terms in the asymptotic expansion but this quickly becomes tedious and is not necessary because, as we show at the end of Section 5, the precise value of C_1 is not relevant for the putative exponential improvement of Minkowski's lower bound on the density, as specified by (5–35).

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S. Torquato, Department of Chemistry, Program in Applied and Computational Mathematics, Princeton Institute for the Science and Technology of Materials and Princeton Center for Theoretical Physics, Princeton University, Princeton, NJ 08544 (torquato@electron.princeton.edu)

F. H. Stillinger, Department of Chemistry, Princeton University, Princeton, NJ 08544