Exactly solvable disordered sphere-packing model in arbitrary-dimensional Euclidean spaces

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We introduce a generalization of the well-known random sequential addition (RSA) process for hard spheres in d-dimensional Euclidean space $\mathbb{R}^d$. We show that all of the n-particle correlation functions of this nonequilibrium model, in a certain limit called the "ghost" RSA packing, can be obtained analytically for all allowable densities and in any dimension. This represents the first exactly solvable disordered sphere-packing model in an arbitrary dimension. The fact that the maximal density $\phi(\infty)=1/2^d$ of the ghost RSA packing implies that there may be disordered sphere packings in sufficiently high d whose density exceeds Minkowski's lower bound for Bravais lattices, the dominant asymptotic term of which is $1/2^d$. Indeed, we report on a conjectural lower bound on the density whose asymptotic behavior is controlled by $2^{-(0.778\,65\ldots)d}$, thus providing the putative exponential improvement on Minkowski's 100-year-old bound. 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.

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INTRODUCTION

A collection of congruent spheres in d-dimensional Euclidean space $\mathbb{R}^d$ is called a sphere packing if no two spheres overlap. The *packing density* or simply density ϕ of a sphere packing is the fraction of space $\mathbb{R}^d$ covered by the spheres. Hard-sphere packings have been used to model a variety of systems, including liquids [1], amorphous and granular media [2], and crystals [3]. Nonetheless, there is great interest in understanding sphere packings in high dimensions in various fields. For example, it is known that the optimal way of sending digital signals over noisy channels correspond to the densest sphere packing in a high-dimensional space [4]. These "error-correcting" codes underlie a variety of systems in digital communications and storage, including compact disks, cell phones, and the Internet. Physicists have studied hard-sphere packings in high dimensions to gain insight into ground and glassy states of matter as well as phase behavior in lower dimensions [5,6]. The determination of the densest packings in arbitrary dimensions is a problem of long-standing interest in discrete geometry [4].

It is instructive to note that upper and lower bounds on the *maximal density*

$$\phi_{\max} = \sup_{P\subset\mathbb{R}^d} \phi(P) \tag{1}$$

exist in all dimensions [4], where the supremum is taken over all packings P in $\mathbb{R}^d$. For example, Minkowski [7] proved that the maximal density $\phi^L_{\max}$ among all Bravais lattice packings for $d\geqslant 2$ satisfies the lower bound

$$\phi^L_{\max} \geqslant \frac{\zeta(d)}{2^{d-1}}, \tag{2}$$

where $\zeta(d)=\Sigma_{k=1}^{\infty} k^{-d}$ is the Riemann zeta function. One observes that for large values of d, the asymptotic behavior of the *nonconstructive* Minkowski lower bound is controlled by 2^{-d}. Interestingly, the density of a *saturated* packing of congruent spheres in $\mathbb{R}^d$ for all d satisfies

$$\phi \geqslant \frac{1}{2^d}. \tag{3}$$

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 center is encompassed by a sphere of unit radius and the density of this covering is $2^d\phi\geqslant 1$, which proves the so-called *greedy* lower bound (3) Note that it has the same dominant exponential term as (2).

A statistically homogeneous (i.e., translationally invariant) packing is completely configurationally characterized by specifying all of the n-particle correlation functions. For such packings in $\mathbb{R}^d$, these correlation functions are defined so that $\rho^n g_n(\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_n)$ is proportional to the probability density for simultaneously finding n particles at locations $\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_n$ within the system, where ρ is the number density. Thus, each g_n approaches unity when all particle positions become widely separated within $\mathbb{R}^d$, indicating no spatial correlations. To date, an exact determination of all of the n-particle correlation functions for a packing has only been possible for $d=1$ in the special case of an equilibrium ensemble of such particles [8]. Observe that in the limit d

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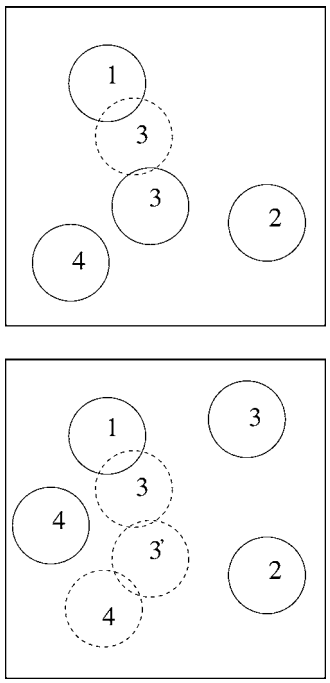

FIG. 1. The addition of four successfully added particles (in the numerical order indicated) in the generalized RSA process at the two extremes of $\kappa=0$ (top panel) and $\kappa=1$ (bottom panel). In both cases, the rejected particles have dashed boundaries. For the case $\kappa=1$, a test sphere cannot overlap a ghost sphere. Here $3'$ represents the second attempt to add a third sphere.

$\rightarrow\infty$, it is known that the pressure of an equilibrium hard-sphere fluid is exactly given by the low-density expansion up to the second-virial level for a positive range of densities [5], which implies a simplified form for all of the correlation functions [9].

We present in Sec. II a generalization of the well-known random sequential addition (RSA) process of hard particles [2,10]. In a particular limit of this nonequilibrium model that we call the "ghost" RSA process, we are able to obtain the g_n for all allowable densities exactly for any n and dimension d. The key geometric quantity that determines g_n is the union volume of n overlapping exclusion spheres of radius equal to the sphere diameter. We show that this construction of a disordered but *unsaturated* packing realizes the greedy lower bound (3). This implies that there may be disordered sphere packings in sufficiently high d whose density exceeds Minkowski's lower bound (2). Indeed, in Sec. III, we report on a conjectural lower bound on the density whose asymptotic behavior is controlled by $2^{(-0.778\,65\ldots)d}$, thus providing the putative exponential improvement on Minkowski's 100-year-old bound. Our results lead to the counterintuitive possibility that optimal packings in sufficiently high dimensions may be disordered and thus have implications for our fundamental understanding of classical ground states of matter.

GENERALIZED RANDOM SEQUENTIAL ADDITION MODEL

We introduce a disordered sphere-packing model in $\mathbb{R}^d$ that is a subset of the Poisson point process and is a generalization of the standard random RSA process. The centers of "test" spheres of the unit diameter arrive continually throughout $\mathbb{R}^d$ during time $t\geqslant 0$ according to a translationally invariant Poisson process of density η per unit time, 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 the 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 correspond 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 standard RSA process in $\mathbb{R}^d$ [2,10], which generates a homogeneous and isotropic sphere packing in $\mathbb{R}^d$ with a time-dependent density $\phi(t)$. In the limit $t\rightarrow\infty$, the RSA process corresponds to a saturated packing with a maximal or *saturation* density $\phi_s(\infty)\equiv\lim_{t\rightarrow\infty}\phi(t)$. In one dimension, the RSA process is commonly known as the "car parking problem," which Reńyi showed has a saturation density $\phi_s(\infty)=0.7476\ldots$ [10]. For $2\leqslant d<\infty$, an exact determination of $\phi_s(\infty)$ is not possible, but estimates for it have been obtained via computer experiments for low dimensions [2].

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 hence 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 model due to Matérn [11] is recovered [12]. The latter is amenable to exact analysis and is the main focus of this paper. For any $0<\kappa\leqslant 1$, the generalized RSA process is always an *unsaturated* packing. Figure 1 illustrates the differences between the generalized RSA process at the two extremes of $\kappa=0$ and $\kappa=1$. In the remainder of this section, we will focus on the case of $\kappa=1$.

The time-dependent density $\phi(t)$ in the case of the generalized RSA process with $\kappa=1$ is easily obtained. In this packing, a test sphere at time t is retained only if does not overlap an existing sphere in the packing as well as any previously rejected test sphere, which we will call "ghost" spheres. The model itself will be referred to as the ghost RSA process. 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 $v_1(1)=\pi^{d/2}/\Gamma(1+d/2)$ is the volume of a sphere of the unit radius. Therefore, the expected time-dependent number density $\rho(t)$ and packing density $\phi(t)=\rho(t)v_1(\frac{1}{2})$ at any time t are given by

$$\rho(t) = \int_0^t \exp[-v_1(1)t']dt' = \frac{1-\exp[-v_1(1)t]}{v_1(1)},$$
$$\phi(t) = \frac{1-\exp[-v_1(1)t]}{2^d}. \tag{4}$$

In the limit $t\to\infty$, we therefore have that

$$\rho(\infty) \equiv \lim_{t\to\infty}\rho(t) = \frac{1}{v_1(1)}, \quad \phi(\infty) \equiv \lim_{t\to\infty}\phi(t) = \frac{1}{2^d}. \tag{5}$$

Observe that the greedy lower bound (3) on the density is achieved in the infinite-time limit for this sequential but *unsaturated* packing, which was pointed out only recently [9]. Although the limiting packing density $\phi(\infty)=1/2^d$ is far from optimal in low dimensions, it is relatively large in high dimensions, as discussed in our concluding remarks. Obviously, for any $0\leqslant\kappa<1$, the maximum (infinite-time) density of the generalized RSA packing is bounded from below by $1/2^d$ (i.e., the maximum density for $\kappa=1$). Henceforth, we write $v_1\equiv v_1(1)$.

The derivation of the expression of $g_2(r;t)$ is actually a simple extension of the aforementioned one for $\rho(t)$. Two test spheres that arrive at times t_1 and t_2 and whose centers are separated by a distance r can only be retained if no other test spheres arrived before t_1 and t_2, respectively (see Fig. 2). Thus, the key geometrical object is the union volume $v_2(r)$ of two spheres of unit radius whose centers are separated by a distance r, which can be expressed in terms of the intersection volume $v_2^{int}(r)$ [13] between two such spheres via the relation

$$v_2(r) = 2v_1 - v_2^{int}(r).$$

For $r\geqslant 2$, there is no volume common to two such spheres $[v_2^{int}(r)=0]$ and, therefore, $g_2(r;t)=1$, i.e., pair correlations vanish. However, if $1\leqslant r\leqslant 2$, the two spheres have a common volume and

$$\begin{aligned}\rho^2(t)g_2(r;t) &= \int_0^t\int_0^t \exp\{-t_1[v_1-v_2^{int}(r)]-t_2[v_1-v_2^{int}(r)] \\ &\quad -\max(t_1,t_2)v_2^{int}(r)\}dt_1dt_2 \\ &= 2\int_0^t dt_2\exp[-t_2v_1]\int_0^{t_2}dt_1 \\ &\quad \times\exp\{-t_1[v_1-v_2^{int}(r)]\} \\ &= \frac{2}{v_2(r)-v_1}\left[\frac{1-e^{-tv_1}}{v_1}-\frac{1-e^{-tv_2(r)}}{v_2(r)}\right].\end{aligned} \tag{6}$$

In relation (6) the terms within the first three brackets are the distinct volumes of the regions labeled 1, 2, and 12 in the top panel of Fig. 2. Therefore, the time-dependent pair correlation function for all r and t is given by

$$\rho^2(t)g_2(r;t) = \frac{2\Theta(r-1)}{v_2(r)-v_1}\left[\frac{1-e^{-tv_1}}{v_1}-\frac{1-e^{-tv_2(r)}}{v_2(r)}\right], \tag{7}$$

where $\Theta(x)$ is the unit step function, equal to zero for $x<0$ and unity for $x\geqslant 1$. It is useful to note that at small times or, equivalently, low densities, formula (4) yields the asymptotic expansion $\phi(t)=t-2^{d-1}t^2+O(t^3)$, which when inverted, yields $t=\phi+2^{d-1}\phi^2+O(\phi^3)$. Substitution of this last result into (7) gives

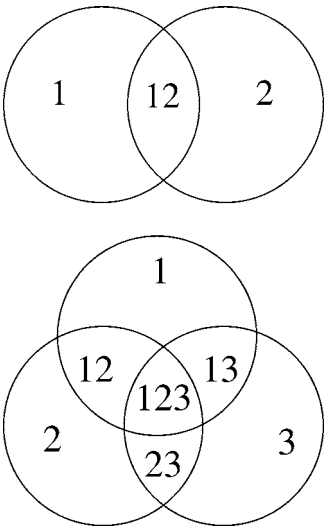

FIG. 2. Top panel: Relevant subvolumes for two overlapping spheres of unit radius associated with the arrivals of two test spheres. The labels refer to distinct, nonoverlapping regions. Bottom panel: Relevant subvolumes for three overlapping spheres of unit radius associated with the arrivals of three test spheres.

$$g_2(r;\phi) = \Theta(r-1) + O(\phi^3), \tag{8}$$

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

In the limit $t\to\infty$, we have from (7) that $\rho^2(\infty)g_2(r;\infty)=2\Theta(r-1)/[v_1v_2(r)]$ or, using (3),

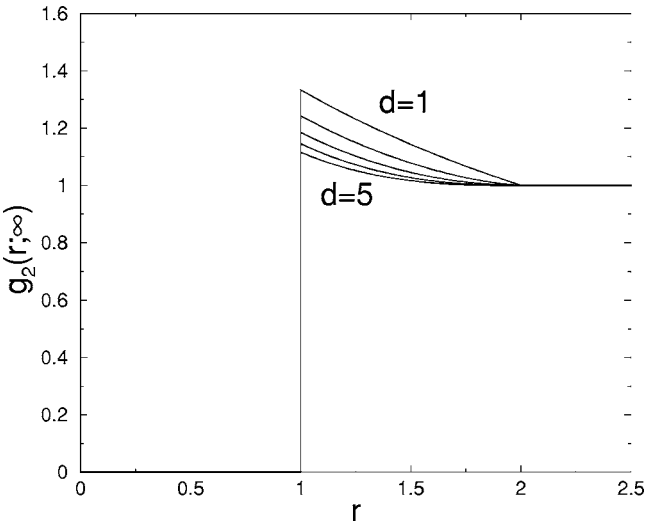

FIG. 3. Radial distribution function for the first five space dimensions at the maximum density $\phi=1/2^d$ for the generalized RSA model with $\kappa=1$, i.e., the "ghost RSA process."

$$g_2(r;\infty) = \frac{2\Theta(r-1)}{\beta_2(r)}, \tag{9}$$

where $\beta_2(r) = v_2(r)/v_1$. The radial distribution function $g_2(r;\infty)$ is plotted in Fig. 3 for the first five space dimensions. Because $\beta_2(r)$ is equal to 2 for $r \geqslant 2$, $g_2(r;\infty) = 1$ for $r \geqslant 2$, 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 exponentially fast with increasing dimension [9], i.e., $g_2(r;\infty)$ tends to the unit step function as $d \to \infty$, i.e., beyond the hard core (a constrained correlation), spatial correlations vanish.

Matérn originally gave an expression for the time-dependent density $\phi(t)$ and a formal expression (as opposed to explicit expression for any d) for the time-dependent radial distribution function $g_2(r;t)$ when $\kappa = 1$ using a completely different approach. However, he did not consider obtaining any of the higher-order correlation functions.

Let us now derive the time-dependent triplet correlation function $g_3(\mathbf{r}_{12}, \mathbf{r}_{13};\infty)$. Here the relevant geometrical object is the union volume $v_3(r_{12}, r_{13}, r_{23})$ of three spheres of unit radius whose centers are separated by the distances r_{12}, r_{13}, and r_{23}, which can be expressed in terms of the intersection volume $v_3^{int}(r_{12}, r_{13}, r_{23})$ between three such spheres via the relation

$$v_3(r_{12}, r_{13}, r_{23}) = 3v_1 - v_2^{int}(r_{12}) - v_2^{int}(r_{13}) - v_2^{int}(r_{23}) + v_3^{int}(r_{12}, r_{13}, r_{23}). \tag{10}$$

Whenever there is no overlap between the three spheres, $g_3 = 1$, i.e., triplet correlations vanish. On the other hand, whenever the spheres overlap such that each pair distance is greater than or equal to unity, there are triplet correlations. In such situations, it is convenient to introduce the time-dependent triplet function

$$\begin{aligned} F(r_{12}, r_{13}, r_{23}; t_1, t_2, t_3) = &- t_1[v_1 - v_2^{int}(r_{12}) - v_2^{int}(r_{13}) + v_3^{int}(r_{12}, r_{13}, r_{23})] - t_2[v_1 - v_2^{int}(r_{12}) - v_2^{int}(r_{23}) + v_3^{int}(r_{12}, r_{13}, r_{23})] \\ &- t_3[v_1 - v_2^{int}(r_{13}) - v_2^{int}(r_{23}) + v_3^{int}(r_{12}, r_{13}, r_{23})] - \max(t_1, t_2)[v_2^{int}(r_{12}) - v_3^{int}(r_{12}, r_{13}, r_{23})] \\ &- \max(t_1, t_3)[v_2^{int}(r_{13}) - v_3^{int}(r_{12}, r_{13}, r_{23})] - \max(t_2, t_3)[v_2^{int}(r_{23}) - v_3^{int}(r_{12}, r_{13}, r_{23})] \\ &- \max(t_1, t_2, t_3) v_3^{int}(r_{12}, r_{13}, r_{23}). \end{aligned} \tag{11}$$

The terms within the first three brackets are the volumes of the regions labeled 1, 2, and 3 in the bottom panel of Fig. 2. The terms within the fourth through sixth brackets are the volumes labeled 12, 13, and 23 in the bottom panel of Fig. 2. Of course, the region labeled 123 denotes the intersection volume of three spheres. The triplet correlation function at time t is given by

$$\rho^3(t) g_3(r_{12}, r_{13}, r_{23}; t) = \int_0^t \int_0^t \int_0^t \exp\left[-F(r_{12}, r_{13}, r_{23}; t_1, t_2, t_3)\right] dt_1 dt_2 dt_3$$

and therefore, at infinitely large times, we have, using (5), (9), and (13), that

$$\begin{aligned} &\rho^3(\infty) g_3(r_{12}, r_{13}, r_{23};\infty) \\ &= 2\int_0^\infty dt_3 \exp[-t_3 v_1] \int_0^{t_3} dt_2 \exp\{-t_2[v_1 - v_2^{int}(r_{23})]\} \int_0^{t_2} dt_1 \exp\{-t_1[v_1 - v_2^{int}(r_{12}) - v_2^{int}(r_{13}) + v_3^{int}(r_{12}, r_{13}, r_{23})]\} \\ &\quad + 2\int_0^\infty dt_1 \exp[-t_1 v_1] \int_0^{t_1} dt_3 \exp\{-t_3[v_1 - v_2^{int}(r_{13})]\} \int_0^{t_3} dt_2 \exp\{-t_2[v_1 - v_2^{int}(r_{12}) - v_2^{int}(r_{23}) + v_3^{int}(r_{12}, r_{13}, r_{23})]\} \\ &\quad + 2\int_0^\infty dt_2 \exp[-t_2 v_1] \int_0^{t_2} dt_1 \exp\{-t_1[v_1 - v_2^{int}(r_{12})]\} \int_0^{t_1} dt_3 \exp\{-t_3[v_1 - v_2^{int}(r_{13}) - v_2^{int}(r_{23}) + v_3^{int}(r_{12}, r_{13}, r_{23})]\} \\ &= \frac{2}{v_1 v_3(r_{12}, r_{13}, r_{23})}\left[\frac{1}{v_2(r_{12})} + \frac{1}{v_2(r_{13})} + \frac{1}{v_2(r_{23})}\right]. \end{aligned} \tag{12}$$

Combination of (5), (9), and (13) yields the following expression for the triplet correlation function for arbitrary positions at infinitely large times:

$$g_3(\mathbf{r}_{12}, \mathbf{r}_{13};\infty) = \frac{\Theta(r_{12}-1)\Theta(r_{13}-1)\Theta(r_{23}-1)}{\beta_3(r_{12}, r_{13}, r_{23})}\left[g_2(r_{12};\infty) + g_2(r_{13};\infty) + g_2(r_{23};\infty)\right], \tag{13}$$

where $\beta_3(r_{12},r_{13},r_{23})=v_3(r_{12},r_{13},r_{23})/v_1$ and $g_3(\mathbf{r}_{12},\mathbf{r}_{13};\infty) \equiv g_3(r_{12},r_{13},r_{23};\infty)$.

A similar analysis reveals that the four-particle correlation function in the limit $t\to\infty$ is given by

$$g_4(\mathbf{r}_{12},\mathbf{r}_{13},\mathbf{r}_{14};\infty) = \frac{\prod_{i<j}^{4}\Theta(r_{ij}-1)}{\beta_4(\mathbf{r}_{12},\mathbf{r}_{13},\mathbf{r}_{14};\infty)}\big[g_3(\mathbf{r}_{12},\mathbf{r}_{13};\infty)+g_3(\mathbf{r}_{12},\mathbf{r}_{14};\infty) + g_3(\mathbf{r}_{13},\mathbf{r}_{14};\infty)+g_3(\mathbf{r}_{23},\mathbf{r}_{24};\infty)\big]. \quad (14)$$

By induction, the n-particle correlation function for arbitrary positions at infinitely large times is given by

$$g_n(\mathbf{r}_1,\ldots,\mathbf{r}_\mathbf{n};\infty)=\frac{\prod_{i<j}^{n}\Theta(r_{ij}-1)}{\beta_n(\mathbf{r}_1,\ldots,\mathbf{r}_n)}\left[\sum_{i=1}^{n} g_{n-1}(Q_i;\infty)\right], \quad (15)$$

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. Moreover, $\beta_n(\mathbf{r}_{12},\mathbf{r}_{13},\ldots,\mathbf{r}_{1n}) =v_n(\mathbf{r}_{12},\mathbf{r}_{13},\ldots,\mathbf{r}_{1n})/v_1$, where $v_n(\mathbf{r}_{12},\mathbf{r}_{13},\ldots,\mathbf{r}_{1n})$ is the union volume of n congruent spheres of a unit radius whose centers are located at $\mathbf{r}_1,\ldots,\mathbf{r}_\mathbf{n},\mathbf{r}_{ij}=\mathbf{r}_j-\mathbf{r}_i$ for all $1\leqslant i<j\leqslant n$ and $r_{ij}=|\mathbf{r}_{ij}|$.

It can be shown [9] that in the limit $d\to\infty$ and for $\phi=1/2^d$

$$g_n(\mathbf{r}_{12},\ldots,\mathbf{r}_{1n};\infty)\sim\prod_{i<j}^{n} g_2(r_{ij};\infty), \quad (16)$$

where $g_2(r;\infty)\sim\Theta(r-1)$. We see that unconstrained spatial correlations vanish asymptotically. Specifically, (i) the high-dimensional asymptotic behavior of g_2 is the same as the asymptotic behavior in the low-density limit for any d [cf. (8)], 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; and (ii) g_n for $n\geqslant 3$ asymptotically can be inferred from a knowledge of only the pair correlation function g_2 and number density ρ. These two asymptotic properties, which we have called the *decorrelation principle* [9], apply more generally to any disordered packing, as discussed in Ref. [9]. Asymptotically, unconstrained correlations vanish (i.e., statistical independence is established) because we know from the Kabatiansky and Levenshtein asymptotic upper bound on the maximal density $\phi_{\max}$ of any sphere packing that the density must go to zero at least as fast as $2^{-0.5990d}$ for large d [14].

DISCUSSION

The fact that the maximal density $\phi(\infty)=1/2^d$ of the ghost RSA packing coincides with the greedy lower bound (3) strongly suggests that there are saturated disordered packings that have larger densities, i.e., the greedy lower bound is a weak bound for saturated packings [15]. This implies that there may be disordered sphere packings in sufficiently high d whose density exceeds Minkowski's lower bound (2) for Bravais lattices, the dominant asymptotic term of which is $1/2^d$. Our results already give insight into this fascinating possibility. For example, consider the so-called checkerboard lattice D_d in d dimensions [4], which is a d-dimensional generalization of the optimal (densest) face-centered cubic lattice in three dimensions, and thought to be the optimal packing for $d=4$ and 5. Its packing density $\phi=\pi^{d/2}/[\Gamma(1+d/2)2^{(d+2)/2}]$ exponentially decreases with increasing d (because it quickly becomes unsaturated) and falls below the ghost-RSA-process value of $1/2^d$ for the first time at $d=28$ [16]. The ratio of densities of the ghost RSA process to the checkerboard at $d=100$ is given by $\phi_{ghost}/\phi_{checker}\approx 7.5\times 10^{25}$. Although both packings are unsaturated in such high dimensions, the fact that $g_2(r)$ for the ghost RSA process is effectively uniform (unity) for all $r>1$ but for the checkerboard lattice involves Dirac δ functions of *weak* strength at *widely spaced* discrete distances explains why the former is enormously denser than the latter.

Over the last century, many extensions and generalizations of Minkowski's lower bound (2) have been obtained [4], but none of these investigations have been able to improve upon the dominant exponential term 2^{-d}. In another work [9], we will present comprehensive rigorous evidence that this exponential improvement may be provided by considering specific disordered sphere packings. Here we simply sketch the procedure leading to this putative improvement over Minkowski's lower bound. The basic ideas underlying our recent approach to the derivation of lower bounds on $\phi_{\max}$ were actually described in our earlier work [17] in which we studied so-called g_2-invariant processes. A *g_2-invariant process* is one in which a given non-negative pair correlation $g_2(\mathbf{r})$ function remains invariant for all $\mathbf{r}$ over the range of densities

$$0\leqslant\phi\leqslant\phi_*. \quad (17)$$

The terminal density ϕ_* is the maximum achievable density for the g_2-invariant process subject to satisfaction of certain necessary conditions on the pair correlation. In particular, we considered those "test" $g_2(r)$'s that are distributions on $\mathbb{R}^d$ depending only on the radial distance r. For any test $g_2(r)$, we want to maximize the corresponding density ϕ satisfying the following three conditions:

$$g_2(r)\geqslant 0 \text{ for all } r, \quad \text{(i)}$$

$$g_2(r)=0 \text{ for } r<1, \quad \text{(ii)}$$

$$S(k)=1+\rho(2\pi)^{d/2}\int_0^\infty r^{d-1}h(r)\frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}}dr\geqslant 0 \text{ for all } k, \quad \text{(iii)}$$

where $h(r)=g_2(r)-1$ is the total correlation function. Condition (i) is a trivial consequence of the fact that g_2 is a probability density function. Condition (ii) is just the *hard-core* constraint for spheres of unit diameter. Condition (iii) states that the structure factor $S(k)$ in d dimensions must be non-

negative for all k. 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

$$\phi_{\max} \geqslant \phi_*. \tag{18}$$

It is rather remarkable that the optimization problem defined above is identical to one formulated by Cohn [18]. Specifically, it is the *dual* of the *primal* infinite-dimensional linear program that Cohn employed with Elkies [19] to obtain upper bounds on the maximal packing density. Thus, even if there does not exist a sphere packing with g_2 satisfying conditions (i)–(iii), the terminal density ϕ_* can never exceed the Cohn-Elkies upper bound and, more generally, our formulation has implications for upper bounds on $\phi_{\max}$.

In addition to the structure factor condition, there are generally many other conditions that a pair correlation function corresponding to a point process must obey [20]. One such additional necessary condition, obtained by Yamada [21], is concerned with the variance $\sigma^2(\Omega) \equiv \langle [N(\Omega)^2 - \langle N(\Omega)\rangle]^2\rangle$, in the number $N(\Omega)$ of particle centers contained within a region or "window" $\Omega \subset \mathbb{R}^d$:

$$\sigma^2(\Omega) = \rho|\Omega|\left[1 + \rho\int_\Omega h(\mathbf{r})d\mathbf{r}\right] \geqslant \theta(1-\theta), \tag{19}$$

where θ is the fractional part of the expected number of points $\rho|\Omega|$ contained in the window. This is a consequence of the fact that the number of particles in any window must be an integer.

In Ref. [17], 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 that are features intended to describe *disordered* jammed sphere packings for $d=3$. However, because of the functional complexity of this test g_2, the terminal density could only be determined numerically. The general optimization procedure outlined above was employed in Ref. [9] to obtain analytical estimates of the terminal density in high dimensions that together with the following conjecture provide the putative exponential improvement on Minkowski's lower bound on $\phi_{\max}$:

Conjecture 1. A hard-core non-negative tempered distribution $g_2(\mathbf{r})$ is a pair correlation function of a translationally invariant disordered sphere packing in $\mathbb{R}^d$ at number density ρ for sufficiently large d if and only if $S(\mathbf{k}) \geqslant 0$. The maximum achievable density is the terminal density ϕ_*.

In other words, for a given $g_2(\mathbf{r})$ that meets the conditions (i)–(iii), at or above a critical dimension d_c, packings exist with such a g_2. A *disordered packing* in $\mathbb{R}^d$ is defined in Ref. [9] to be one in which the pair correlation function $g_2(\mathbf{r})$ decays to its long range value of unity faster than $|\mathbf{r}|^{-d+\epsilon}$ for some $\epsilon > 0$. Employing the aforementioned optimization procedure with a certain test function g_2 and *Conjecture 1*, we obtain in what follows provisional lower bounds that yield the long-sought asymptotic exponential improvement on Minkowski's bound. An important feature of any dense packing is that the particles form contacts with one another. Experience with disordered jammed packings in low dimensions reveals that the contact or 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. It is desired to idealize this small-distance negative correlation (relative to the uncorrelated value of unity) in such a way that it is amenable to exact asymptotic analysis. Accordingly, a test radial distribution function was considered in Ref. [9] in which there is a gap between the location of a unit step function and the δ function at finite d, i.e.,

$$g_2(r) = \Theta(r-\sigma) + \frac{Z}{s_1(1)\rho}\delta(r-1), \tag{20}$$

where $s(r)$ is the surface area of a d-dimensional sphere of radius r and Z is a parameter, which is the average contact or kissing number, and unity is the sphere diameter. The expression contains two adjustable parameters, $\sigma \geqslant 1$ and Z, which must obviously be constrained to be non-negative.

Before reporting the main results of this optimization, it is instructive to examine the test function (20) for two special cases: (1) one in which $\sigma=1$ and $Z=0$ and (2) the other in which $\sigma=1$ and $Z>0$ (which were considered in Ref. [17]). In the first special instance, there are no parameters to be optimized here, and the terminal density ϕ_* is given by $\phi_* = 1/2^d$. It is simple to show that the Yamada condition (19) is satisfied in any dimension for $0 \leqslant \phi \leqslant 2^{-d}$. We already established in the previous section that there exist sphere packings that asymptotically have radial distribution functions given by the simple unit step function for $\phi \leqslant 2^{-d}$. Nonetheless, invoking *Conjecture 1* and the obtained terminal density, implies the asymptotic lower bound on the maximal density is given by

$$\phi_{\max} \geqslant \frac{1}{2^d}, \tag{21}$$

which provides an alternate derivation of the elementary bound (3). 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 is given by the test function (20) with $\sigma=1$ and $Z=0$ in one, two, and three dimensions for densities up to the terminal density [22,23]. The existence of such a discrete approximation to this test g_2 is suggestive that the standard non-negativity conditions may be sufficient to establish existence in this case for densities up to ϕ_*. In the second special case ($\sigma=1$ and $Z>0$) and under the constraint that the minimum of $S(k)$ occurs at $k=0$ then we have the exact results $\phi_* = (d+2)/2^{d+1}$ and $Z_* = d/2$ where Z_* is the optimized average kissing number. The Yamada condition (19) is violated here only for $d=1$ and 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 this test function in two and three dimensions for densities up to the terminal density [23]. This is suggestive that *Conjecture 1* for this test function may in fact be stronger than is required. In the high-dimensional limit, invoking *Conjecture*

1 and the obtained terminal density, yields the conjectural lower bound

$$\phi_{\max} \geqslant \frac{d+2}{2^{d+1}}. \tag{22}$$

This lower bound provides the same type of linear improvement over Minkowski's lower bound as does Ball's rigorous lower bound [24] obtained using a completely different approach.

Now let us consider the problem when both σ and Z in (20) must be optimized. The presence of a gap between the unit step function and δ function will indeed lead asymptotically to substantially higher terminal densities. For sufficiently small d ($d \leqslant 200$), the optimization procedure is carried out numerically [9]. The Yamada condition (19) is violated only for $d=1$ for the test function (20) for the terminal density ϕ_* and associated optimized parameters σ_* and $Z_*=(2\sigma_*\phi_*)^d-1$. One can again verify directly that the Yamada condition becomes less restrictive as the dimension increases from $d=2$. However, although the test function (20) 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_* [9]. To our knowledge, this is the first example of a test radial distribution function that satisfies the two standard non-negativity conditions (i) and (iii) and the Yamada condition (19), 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 $d=2$. As is the case with the Yamada condition (19), this additional necessary condition appears to lose relevance in low dimensions because we have shown that there is no analogous local geometric constraint violation for $d \geqslant 3$. For $d \leqslant 56$, the terminal density lies below the density of the densest known packing (a Bravais lattice) [4]. However, for $d>56, \phi_*$ can be larger than the density of the densest known arrangements, which are ordered. Our numerical results for d between 3 and 200, reveal exponential improvement of the terminal density ϕ_* over the one for the gapless case, where $\phi_*=(d+2)/2^{d+1}$.

For large d, an exact (but nontrivial) asymptotic analysis can be performed [9], yielding the optimal terminal density. This result in conjunction with *Conjecture 1* yields the conjectural asymptotic lower bound

$$\phi_* \sim \frac{3.276\,100\,896 d^{1/6}}{2^{[3-\log_2(e)]d/2}} = \frac{3.276\,100\,896 d^{1/6}}{2^{(0.778\,652\,479\,5\ldots)d}}. \tag{23}$$

This putatively provides the long-sought exponential improvement on Minkowski's lower bound. We call this is a conjectural lower bound because it relies on *Conjecture 1* being true, which a number of results support. First, the decorrelation principle states that unconstrained correlations in disordered sphere packings vanish asymptotically in high dimensions and that the g_n for any $n \geqslant 3$ can be inferred entirely from a knowledge of ρ and g_2. Second, the necessary Yamada condition appears to only have relevance in very low dimensions. Third, we have demonstrated that other new necessary conditions also seem to be germane only in very low dimensions. Fourth, we recover the form of known rigorous bounds [cf. (21) and (22)] in special cases of the test radial distributions function (20) when we invoke *Conjecture 1*. Finally, in these two instances, configurations of disordered sphere packings on the torus have been numerically constructed with such g_2 in low dimensions for densities up to the terminal density [22,23].

A byproduct of the bound (23) is the conjectural asymptotic lower bound on the maximal kissing number [9]

$$\begin{aligned} Z_{\max} \geqslant Z_* &\sim 40.24\,850\,787 d^{1/6} 2^{[\log_2(e)-1]d/2} \\ &= 40.24\,850\,787 d^{1/6} 2^{(0.221\,347\,520\,5\ldots)d}. \end{aligned} \tag{24}$$

This result is superior to the best known asymptotic lower bound on the maximal kissing number of $2^{0.2075,\ldots,d}$ [25].

The work described above suggests 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. In fact, there is no fundamental reason why disordered ground states are prohibited in low dimensions [26]. A case in point are the "pinwheel" tilings of the plane, which possess both statistical translational and rotational invariance [27].

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[1] J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic Press, New York, 1986).
[2] S. Torquato, *Random Heterogeneous Materials: Microstructure and Macroscopic Properties* (Springer-Verlag, New York, 2002).
[3] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics*. (Cambridge University Press, New York, 1995).
[4] J. H. Conway and N. J. A. Sloane, *Sphere Packings, Lattices and Groups* (Springer-Verlag, New York, 1993).
[5] H. L. Frisch and J. K. Percus, Phys. Rev. E **60**, 2942 (1999).
[6] G. Parisi and F. Slanina, Phys. Rev. E **62**, 6554 (2000).
[7] H. Minkowski, J. Reine Angew. Math. **129**, 220 (1905).
[8] Z. W. Salsburg, R. W. Zwanzig, and J. G. Kirkwood, J. Chem. Phys. **21**, 1098 (1953).
[9] S. Torquato and F. H. Stillinger, Experimental Math. (to be published). See also e-print math.MG/0508381 for the preprint version of the paper.
[10] A. Reńyi, Probab. Math. Stat. **4**, 203 (1963); B. Widom, J. Chem. Phys. **44**, 3888 (1966); J. Feder, J. Theor. Biol. **87** 237 (1980); R. M. Ziff and R. D. Vigil, J. Phys. A **23**, 5103 (1990); P. Viot, G. Tarjus, and J. Talbot, Phys. Rev. E **48**, 480 (1993).
[11] B. Matérn, *Spatial Variation*, Lecture Notes in Statistics Vol. 36 (Springer-Verlag, New York, 1986).

[12] Interestingly, this model can be mapped exactly to a irreversible multilayer adsorption problem in which spheres are deposited from the $(d+1)$-th dimension onto a d-dimensional substrate. See P. R. Van Tasell and P. Viot, Europhys. Lett. **40**, 293 (1997).

[13] S. Torquato and F. H. Stillinger, Phys. Rev. E **68**, 041113 (2003); **68**, 069901 (2003). This paper gives an explicit formula for the intersection volume $v_2^{\text{int}}(r)$ in any d. See also Ref. [9] for another exact representation of this intersection volume for any d.

[14] G. A. Kabatiansky and V. I. Levenshtein, Probl. Inf. Transm. **14**, 1 (1978).

[15] Indeed, we will show in a future work that the standard RSA packing ($\kappa=0$) has a substantially higher density than $1/2^d$ in high dimensions.

[16] In such dimensions, denser Bravais lattice packings exist, but it is conjectured that in sufficiently high dimensions all Bravais lattices become unsaturated (see Ref. [4]) and therefore inefficient packings.

[17] S. Torquato and F. H. Stillinger. J. Phys. Chem. B **106**, 8354 (2002); **106**, 11406 (2002)

[18] H. Cohn, Geom. Topol. **6**, 329 (2002).

[19] H. Cohn and N. Elkies, Ann. Math. **157**, 689 (2003).

[20] O. Costin and J. Lebowitz, J. Phys. B **108**, 19614 (2004).

[21] M. Yamada, Prog. Theor. Phys. **25**, 579 (1961).

[22] J. R. Crawford, S. Torquato, and F. H. Stillinger, J. Chem. Phys. **119**, 7065 (2003).

[23] O. Uche, F. H. Stillinger, and S. Torquato. Physica A **360**, 21 (2006).

[24] K. Ball, Duke Math. J. **68**, 217 (1992).

[25] A. D. Wyner, Bell Syst. Tech. J. **44**, 1061 (1965).

[26] D. Ruelle, Physica A **113**, 619 (1982); J. Miekisz and C. Radin, Phys. Rev. B **39**, 1950 (1989).

[27] C. Radin, Ann. Math. **139**, 661 (1994).