

High-dimensional generalizations of the kagomé and diamond crystals and the decorrelation principle for periodic sphere packings

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Received 19 August 2011

Accepted 22 September 2011

Published 17 October 2011

Online at stacks.iop.org/JSTAT/2011/P10017

[doi:10.1088/1742-5468/2011/10/P10017](https://doi.org/10.1088/1742-5468/2011/10/P10017)

Abstract. In this paper, we introduce constructions of the high-dimensional generalizations of the kagomé and diamond crystals. The two-dimensional kagomé crystal and its three-dimensional counterpart, the pyrochlore crystal, have been extensively studied in the context of geometric frustration in antiferromagnetic materials. Similarly, the polymorphs of elemental carbon include the diamond crystal and the corresponding two-dimensional honeycomb structure, adopted by graphene. The kagomé crystal in d Euclidean dimensions consists of vertex-sharing d -dimensional simplices in which all of the points are topologically equivalent. The d -dimensional generalization of the diamond crystal can then be obtained from the centroids of each of the simplices, and we show that this natural construction of the diamond crystal is distinct from the D_d^+ family of crystals for all dimensions $d \neq 3$. We analyze the structural properties of these high-dimensional crystals, including the packing densities, coordination numbers, void exclusion probability functions, covering radii and quantizer errors. Our results demonstrate that the so-called decorrelation principle, which formally states that unconstrained correlations vanish in asymptotically high dimensions, remarkably applies to the case of periodic point patterns with inherent long-range order. We argue that the decorrelation principle is already exhibited in periodic crystals in low dimensions via a ‘smoothed’ pair correlation function

obtained by convolution with a Gaussian kernel. These observations support the universality of the decorrelation principle for any point pattern in high dimensions, whether disordered or not. This universal property in turn suggests that the best conjectural lower bound on the maximal sphere-packing density in high Euclidean dimensions derived by Torquato and Stillinger (2006 *Expt. Math.* **15** 307) is, in fact, optimal.

Keywords: correlation functions, disordered systems (theory), fluctuations (theory)

ArXiv ePrint: 1108.3792

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1. Introduction

There has been substantial recent interest in the physics of high-dimensional systems with applications to ground-state problems, packing problems, number theory and phase behaviors of many-particle systems [1]–[8]. The problem of identifying the densest sphere packings in high-dimensional Euclidean space \mathbb{R}^d is an open and fundamental problem in discrete geometry and number theory with important applications to communications theory [9]. In particular, Shannon showed that the optimal method of sending digital signals over noisy channels corresponds to the densest packing in a high-dimensional space [10]. Although the densest packings in dimensions two and three are known to be Bravais lattice packings (the triangular and FCC lattices, respectively [11]), in sufficiently high dimensions, non-Bravais lattices almost surely are the densest packings. In addition to providing putative exponential improvement on Minkowski's lower bound for the maximal sphere-packing density, Torquato and Stillinger presented strong arguments to suggest that the densest packings in high dimensions are, in fact, disordered [12, 13].

Their methods rely on the so-called decorrelation principle for disordered sphere packings, which states that, as the dimension d increases, all unconstrained correlations asymptotically vanish and any higher-order correlation functions $g_n(\mathbf{r}^n)$ may be expressed in terms of the number density ρ and the pair correlation function g_2 [12]. Since its introduction, additional work has shown that the decorrelation principle is remarkably robust, meaning that it is already manifested in low dimensions and applies also to certain soft-matter systems [14] and quantum many-particle distributions [15]. Furthermore, detailed numerical studies of saturated maximally random jammed hard-sphere packings, which are the most disordered packings of spheres that are rigorously incompressible and nonshearable, have demonstrated that unconstrained correlations beyond the hard-sphere diameter asymptotically vanish even in relatively low dimensions $d = 1-6$ [4]. Similar results have also been observed for the exactly-solvable 'ghost' random sequential addition (RSA) process [16] along with the usual RSA process [17]. All evidence to date supports the notion that the decorrelation principle applies fundamentally to disordered many-particle systems. In this paper, we provide evidence that the decorrelation principle applies more generally to any periodic crystal, which has implications for the densest sphere packings in high dimensions and sheds light on the reasons why it is robust in low dimensions.

The properties of periodic crystal structures are fundamental to the physical and mathematical sciences. Experience in two- and three-dimensional systems suggests that crystals are prototypical ground states of matter [18], obtained by slow annealing of an interacting many-particle system to absolute zero temperature. Unlike disordered states of matter, including gases and liquids, crystals possess complete long-range correlations and translational symmetry. As such, periodic crystals can be specified by translational copies of a single fundamental cell containing one (in the case of a Bravais lattice) or more particles.

Elemental carbon is known to adopt numerous polymorphs of fundamental significance. Its four-electron valence structure implies that it can be tetrahedrally, covalently bonded with itself to form the diamond crystal. Certain 'superdense' polymorphs of carbon involving different packings of carbon tetrahedra have also recently been reported in the literature [19]. The two-dimensional analog of the diamond crystal is

the so-called honeycomb crystal, in which points are placed at the vertices of hexagons that tile the plane. This polymorph of carbon is the graphene structure, variations of which have gained substantial interest as nanomaterials [20]. Each point in the honeycomb crystal is coordinated with three other points of the structure, and by placing particles at the midpoints of each of the ‘bonds’, one obtains the kagomé crystal. The kagomé crystal and its three-dimensional counterpart, the pyrochlore crystal, have been used in models of spin-frustrated antiferromagnetic materials [21]. This type of geometric frustration in so-called ‘spin ice’ induces a nonvanishing residual entropy in the ground state, equivalent to behavior identified in liquid water [22].

Recently, Torquato has reformulated the covering and quantizer problems from discrete geometry as ground-state problems involving many-body interactions with one-body, two-body, three-body and higher-body potentials [1]. Formally, the covering problem seeks the point configuration that minimizes the radius of overlapping spheres circumscribed around each of the points required to cover \mathbb{R}^d [9, 1]. The quantizer problem involves finding the point configuration in \mathbb{R}^d that minimizes a Euclidean ‘distance error’ associated with replacing a randomly placed point in \mathbb{R}^d with the nearest point of the point process [9, 1]. Closely related is the so-called number variance problem, which aims to identify the distribution of points that minimize fluctuations in the local number density over large length scales [2, 3]. This problem can also be interpreted as the determination of the ground state for a particular soft, bounded pair interaction and, for the special case of Bravais lattices, is equivalent to identifying the minimizer of the so-called Epstein zeta function [23]. Note that the number variance of a point pattern has been suggested to quantify structural order over large length scales [2, 3].

Studies of many-body fluids and amorphous packings have attempted to glean new information about low-dimensional physical properties, including the equation of state, radius of convergence of the virial series, phase transitions and structure, from high-dimensional models. Frisch and Percus have shown that, for repulsive interactions, Mayer cluster expansions of the free energy are dominated by ring diagrams at each order in particle density ρ [24]. This result was extended by Zachary, Stillinger and Torquato to show that the so-called mean-field approximation for soft, bounded pair interactions becomes exact in the high-dimensional limit [14]. Parisi and Zamponi have utilized the HNC approximation to the pair correlation function and mean-field theory to understand hard-sphere glasses and jamming in high dimensions [5], and Rohrmann and Santos have generalized results from liquid-state theory to study fluids of hard spheres in high dimensions [6]. Michels and Trappeniers [25], Skoge *et al* [4], van Meel *et al* [7] and Lue *et al* [8] have numerically studied the effect of dimensionality of the disorder–order transition in equilibrium hard-sphere systems in up to dimension six. Additionally, Doren and Herschbach have developed a dimensionally dependent perturbation theory for quantum systems to draw conclusions about the energy eigenvalues in low dimensions [26].

In this paper, we generalize the kagomé and diamond crystals for high-dimensional Euclidean space \mathbb{R}^d . We are motivated by the observation that there are $d + 1$ particles within the fundamental cell of the kagomé crystal, which grows with the dimension. The high-dimensional kagomé crystal thus possesses a large basis of particles and approximates the case of a (possibly irregular) N -particle many-particle distribution subject to periodic boundary conditions for N large. The d -dimensional

kagomé crystal provides an intriguing structure for which to test the applicability of the decorrelation principle for periodic point patterns. Since such periodic crystals possess full long-range order, it is highly nonintuitive that the decorrelation principle should apply, and yet we provide indirect and direct evidence that it continues to hold in this general setting. Furthermore, by analyzing the structural properties of the high-dimensional diamond and kagomé crystals, we show that certain ‘disordered’ packings can be quantitatively more ordered with respect to local fluctuations in the number density than periodic crystals, even in relatively low dimensions. Our results therefore have important implications for the low- and high-dimensional problems outlined above.

Our major results are summarized as follows:

- (i) We develop constructions of high-dimensional generalizations of the kagomé and diamond crystals using the geometry of the fundamental cell for the A_d Bravais lattice (defined below). Our results suggest a natural method for constructing a large class of ‘kagomé-like’ crystals in high dimensions.
- (ii) We examine the behavior of structural features of the kagomé and diamond crystals, including the packing densities, coordination numbers, covering radii and quantizer errors. In particular, we show that the kagomé crystal possesses a lower packing fraction than the diamond crystal for all $d \geq 4$, a larger covering radius for all $d \geq 2$ and a larger quantizer error for all $d \geq 3$.
- (iii) We relate these structural features to the distribution of the void space external to the particles in the fundamental cell via numerical calculation of the void exclusion probability function E_V (defined below). As the spatial dimension increases, the fundamental cell of the kagomé lattice develops substantially large holes, thereby skewing the bulk of the void-space distribution such that large holes are less rare than in the uncorrelated Poisson point pattern. The kagomé crystal therefore lies above Zador’s upper bound on the minimal quantizer error in sufficiently high dimensions.
- (iv) We calculate the number variance coefficients governing asymptotic surface area fluctuations in the local number density for the kagomé and diamond crystals. The kagomé crystal for all $d \geq 3$ possesses a larger number variance coefficient than a certain correlated disordered packing corresponding to a so-called g_2 -invariant process [12, 27, 28], providing indirect evidence for a decorrelation principle of high-dimensional periodic point patterns.
- (v) We provide direct evidence for a decorrelation principle of periodic structures by examining a ‘smoothed’ pair correlation function for the d -dimensional kagomé crystal. Our analysis also applies to Bravais lattices as shown by corresponding calculations for the hypercubic lattice \mathbb{Z}^d , establishing the universality of the decorrelation effect. These results suggest that pair correlations alone are sufficient to completely characterize a sphere packing for large dimension d and that the best conjectural lower bound on the maximal density of sphere packings provided by Torquato and Stillinger [12] may, in fact, be optimal in high dimensions. This statement suggests that the densest sphere packings in high dimensions are, in fact, disordered.

2. Definitions

2.1. Crystals and correlation functions

A d -dimensional Bravais lattice is a periodic structure defined by integer linear combinations of a set of basis vectors $\{\mathbf{e}_j\}$ for \mathbb{R}^d , i.e.

$$\mathbf{p} = \sum_{j=1}^d n_j \mathbf{e}_j \equiv M_\Lambda \mathbf{n} \quad (n_j \in \mathbb{Z} \forall j) \quad (1)$$

for all points \mathbf{p} of the Bravais lattice [9], where we have defined the generator matrix M_Λ of the Bravais lattice Λ with columns given by the basis vectors. The basis vectors define a fundamental cell for the Bravais lattice containing only one lattice point. This concept can be naturally generalized to include multiple points within the fundamental cell, defining a periodic crystal or non-Bravais lattice³. Specifically, a non-Bravais lattice consists of the union of a Bravais lattice with one or more translates of itself; it can therefore be defined by specifying the generator matrix M_Λ for the Bravais lattice along with a set of translate vectors $\{\boldsymbol{\nu}_j\}$. Note that the special case of a single zero translate vector $\mathbf{0}$ defines a Bravais lattice.

Every Bravais lattice Λ possesses a dual Bravais lattice Λ^* with lattice points \mathbf{q} defined by $\mathbf{p} \cdot \mathbf{q} = m \in \mathbb{Z}$.⁴ The generator matrix for the dual Bravais lattice is given by [9]

$$M_{\Lambda^*} = (M_\Lambda^T)^{-1}, \quad (2)$$

where B^T denotes the transpose of a matrix B . A Bravais lattice and its dual obey the Poisson summation formula⁵ for any Schwarz function [18, 29]. In general, a crystal, containing more than one particle per fundamental cell, does not possess a dual structure in the same sense as for a Bravais lattice.

A many-particle distribution is determined by its number density ρ , equal to the number of particles per unit volume, and the set of n -particle correlation functions g_n , proportional to the probability density of finding a configuration \mathbf{r}^n of any n particles within the system. Of particular importance is the pair correlation function $g_2(r)$, which for an isotropic and statistically homogeneous point pattern is a function only of the magnitude r of pair separations between particles. For any periodic crystal consisting of topologically equivalent particles, the angularly averaged pair correlation function has the form

$$\rho s(r) g_2(r) = \sum_{k=1}^{+\infty} Z_k \delta(r - r_k), \quad (3)$$

³ Physicists and crystallographers also refer to non-Bravais lattices as lattices ‘with a basis’. Mathematicians reserve the term lattice only to refer to a Bravais lattice. Here, we will use the term lattice to refer only to a Bravais lattice; our usage of the term crystal refers generally to any periodic point pattern.

⁴ Physicists also define a dual Bravais lattice with the condition $\mathbf{p} \cdot \mathbf{q} = 2\pi m$ for some $m \in \mathbb{Z}$, corresponding to a different scaling for the Fourier transform.

⁵ The Poisson summation formula generalizes the Fourier series of an appropriately well-behaved function f that is periodic with respect to the fundamental cell of a Bravais lattice Λ . Namely, for every vector $\mathbf{v} \in \mathbb{R}^d$, $\sum_{\mathbf{p} \in \Lambda} f(\mathbf{p} + \mathbf{v}) = [v_F(\Lambda)]^{-1} \sum_{\mathbf{q} \in \Lambda^*} \exp(-2\pi i \mathbf{q} \cdot \mathbf{v}) \hat{f}(\mathbf{q})$, where $v_F(\Lambda)$ is the volume of the fundamental cell for Λ and \hat{f} is the unitary Fourier transform of the function f . See [38] (see footnote 7) for an application of Poisson summation to maximally dense sphere packings.

where Z_k is the number of points at a radial distance r_k away from a reference particle of the lattice and $s(r)$ is the surface area of a d -dimensional sphere of radius r . The cumulative coordination number $Z(R)$, the total number of particles within a radial distance R from a reference particle, is therefore given by

$$Z(R) = \rho \int_0^R s(r)g_2(r) dr; \quad (4)$$

for a periodic crystal. This identity simplifies to $Z(R) = \sum_{k=1}^K Z_k$, where K denotes the highest index for which $r_K \leq R$.

2.2. Hyperuniformity and the number variance problem

Torquato and Stillinger have characterized fluctuations in the local number density of a many-particle distribution [2] and have shown that these fluctuations behave differently for periodic crystals and uncorrelated systems. Define the random variable $N(\mathbf{x}_0; R)$ to be the number of particles within a spherical observation window of radius R centered at position \mathbf{x}_0 . By definition, $\langle N(\mathbf{x}_0; R) \rangle = \rho v(R)$, where $v(R)$ is the volume of a d -dimensional sphere of radius R . For a Poisson point pattern in which there are no correlations between particles, the underlying Poisson counting measure also implies that

$$\sigma^2(R) = \langle N^2(\mathbf{x}_0; R) \rangle - \langle N(\mathbf{x}_0; R) \rangle^2 = \langle N(\mathbf{x}_0; R) \rangle = \rho v(R), \quad (5)$$

meaning that fluctuations in the local number density of the observation window scale with the window volume.

However, this scaling is not a general feature of all point patterns. In the general case of correlated point patterns, the local number variance is given by [2]

$$\sigma^2(R) = \rho v(R) \left\{ 1 + \rho \int [g_2(\mathbf{r}) - 1] \alpha(r; R) d\mathbf{r} \right\}, \quad (6)$$

where $\alpha(r; R)$ is the so-called scaled intersection volume, defined geometrically as the volume of the intersection of two d -dimensional spheres of radius R with centers separated by a distance r , normalized by the volume $v(R)$ of a sphere. Explicit expressions for the scaled intersection volume in various dimensions have been given by Torquato and Stillinger [2, 12].

Introducing a length scale D (e.g. the mean nearest-neighbor distance between points) and a corresponding reduced density $\eta = \rho v(D/2)$, one can show that the asymptotic behavior of the number variance for large observation windows is [2]

$$\sigma^2(R) = 2^d \eta \{ A(R/D)^d + B(R/D)^{d-1} + o[(R/D)^{d-1}] \}, \quad (7)$$

where $o(x)$ denotes terms of order less than x . The coefficients A and B are given by

$$A = 1 + \rho \int [g_2(\mathbf{r}) - 1] d\mathbf{r} = \lim_{\|\mathbf{k}\| \rightarrow 0} S(\mathbf{k}) \quad (8)$$

$$B = \frac{-\eta \Gamma(1 + d/2)}{D v(D/2) \Gamma[(d + 1)/2] \Gamma(1/2)} \int \|\mathbf{r}\| [g_2(\mathbf{r}) - 1] d\mathbf{r}, \quad (9)$$

where $S(\mathbf{k}) = 1 + \rho \mathfrak{F}\{g_2(\mathbf{r}) - 1\}(\mathbf{k})$, with \mathfrak{F} denoting the Fourier transform, is the structure factor. It follows that the number variance of any point pattern for which $A = 0$

grows more slowly than the volume of the observation window, implying that the point pattern is effectively homogeneous even on local length scales. Such systems are known as hyperuniform [2] or superhomogeneous point patterns [30]. Examples of hyperuniform point patterns include all Bravais and non-Bravais lattices, quasicrystals possessing Bragg peaks, and certain disordered point patterns with pair correlations decaying exponentially fast.

It has also been suggested that the coefficient B quantifies large-scale order in a hyperuniform point pattern [2, 3]. The issue of identifying the point pattern that minimizes this coefficient defines the so-called number variance problem [1, 2]. It has recently been proved that the integer lattice is the unique number variance minimizer in one dimension among all hyperuniform point patterns [2]. Numerical results strongly suggest that the triangular lattice minimizes the number variance in two dimensions [2, 3]. However, contrary to the expectation that the densest lattice packing should also minimize the number variance, it has been shown in three dimensions that the BCC lattice possesses a lower asymptotic number variance coefficient B than the FCC lattice [2]. It is worth mentioning in this regard that the BCC lattice is the dual of FCC.

2.3. Jamming in hard-sphere packings

A sphere packing is obtained from a point pattern in d -dimensional Euclidean space by decorating each of the points with a sphere of radius R_P such that no spheres overlap after the decoration; the parameter R_P is the packing radius. It is an open and nontrivial problem to quantify the extent of randomness (equivalently, of order) in a sphere packing, which reflects nontrivial structural information about the system. Research in this area is aimed at identifying sets of order metrics ψ [31] that align with physical intuitions of order, at least in relatively low dimensions, and are positively correlated. It has recently been proposed that hyperuniformity is itself a measure of order over large length scales [2, 3].

Torquato and Stillinger have introduced a classification of sphere packings in terms of the extent to which they are jammed [32, 13]. In particular, they have provided a mathematically precise hierarchy of jammed sphere packings, distinguished depending on the nature of their mechanical stability [32, 13]:

- (i) *Local jamming*: each particle in the packing is locally trapped by at least $d + 1$ contacting neighbors, not all in the same hemisphere. Locally jammed particles cannot be translated while fixing the positions of all other particles.
- (ii) *Collective jamming*: any locally jammed configuration is collectively jammed if no subset of particles can simultaneously be displaced so that its members move out of contact with each other and with the remainder set.
- (iii) *Strict jamming*: any collectively jammed configuration is strictly jammed if it disallows all uniform volume-nonincreasing strains of the system boundary.

These categories certainly do not include all possible distinctions of jammed configurations, but they span a reasonable spectrum of possibilities. Importantly, jamming depends explicitly on the boundary conditions for the packing [32, 13].

Isostatic packings are jammed packings with the minimal number of contacts M for a given jamming category under the specified boundary conditions [13]. Under periodic boundary conditions, for collective jamming $M = 2N - 1$ and $3N - 2$ for $d = 2$ and

$d = 3$, respectively, and for strict jamming $M = 2N + 1$ and $3N + 3$ for $d = 2$ and $d = 3$, respectively [13, 33]. In this case, the relative differences between isostatic collective and strictly jammed packings diminishes for large N , and an isostatic packing in d dimensions has a mean contact number per particle $Z = 2d$, known as the isostatic condition. Note, however, that packings for which $Z = 2d$ are not necessarily collectively or strictly jammed; the two-dimensional square lattice and three-dimensional simple-cubic lattice are simple counterexamples in dimensions $d = 2$ and $d = 3$, respectively. Another interesting example is the two-dimensional kagomé crystal, which is locally jammed but neither collectively nor strictly jammed under periodic boundary conditions and possesses a nearest-neighbor contact number per particle $Z = 4$ [32, 33]. However, this structure can be made strictly jammed by ‘reinforcing’ it with an extra ‘row’ and ‘column’ of disks [33].

2.4. The covering problem

Consider a distribution of particles at unit number density. The covering radius for the point process is defined by decorating each of the particles with a sphere of radius R and identifying the minimal radius R_C necessary to cover the space completely. More precisely, for any choice of R , we can define the volume fraction of space ϕ_P occupied by the spheres; the volume fraction occupied by the void space external to the spheres is then $\phi_V = 1 - \phi_P$. The covering radius R_C is then defined as the minimal value of R for which $\phi_P = 1$ and $\phi_V = 0$.

The volume fraction ϕ_V of the void space external to a set of spheres of radius R is equivalent to the probability of inserting a ‘test’ sphere of radius R into the system and finding it contained entirely in the void space. This latter quantity is known as the void exclusion probability function $E_V(R)$ and can be expressed in terms of the n -particle correlation functions for the underlying point pattern [35, 34]:

$$E_V(R) = 1 + \sum_{k=1}^{+\infty} \frac{(-\rho)^k}{\Gamma(k+1)} \int g_k(\mathbf{r}^k) \prod_{j=1}^k \Theta(R - \|\mathbf{x} - \mathbf{r}_j\|) d\mathbf{r}_j, \quad (10)$$

where $\Theta(x)$ is the Heaviside step function. This expression can also be rewritten for a statistically homogeneous point pattern in terms of intersection volumes of spheres [1]:

$$E_V(R) = 1 + \sum_{k=1}^{+\infty} \frac{(-\rho)^k}{\Gamma(k+1)} \int g_k(\mathbf{r}_{12}, \dots, \mathbf{r}_{1k}) v_{\text{int}}^{(k)}(\mathbf{r}^k; R) d\mathbf{r}^k, \quad (11)$$

where $v_{\text{int}}^{(k)}(\mathbf{r}^k; R)$ is the intersection volume of k spheres of radius R and centers at \mathbf{r}^k :

$$v_{\text{int}}^{(k)}(\mathbf{r}^k; R) = \int d\mathbf{x} \prod_{j=1}^k \Theta(R - \|\mathbf{x} - \mathbf{r}_j\|). \quad (12)$$

The expression (11) implies that $E_V(R)$ can be interpreted as a total energy per particle associated with a many-particle interaction involving one-body, two-body, three-body and higher-body potential energies [1]. For a single realization of N points in a volume $V \subset \mathbb{R}^d$ [1]

$$E_V(R) = 1 - \rho v(R) + \frac{1}{V} \sum_{i < j} v_{\text{int}}^{(2)}(r_{ij}; R) - \frac{1}{V} \sum_{i < j < k} v_{\text{int}}^{(3)}(r_{ij}, r_{ik}, r_{jk}; R) + \dots \quad (13)$$

We remark that truncating the series expression (11) at order k provides an upper bound to E_V when k is even and a lower bound when k is odd [36].

The covering problem concerns identifying the point pattern with the minimal covering radius at unit number density. In particular, one attempts to identify the point pattern that minimizes the one-dimensional Lebesgue measure of the interval of compact support $[0, R_C]$ of the void exclusion probability function $E_V(R)$. A lower bound on the minimal covering radius can be obtained by truncating the series representation (11) for E_V at first order, implying at unit number density that

$$E_V(R) \geq [1 - v(R)]\Theta[1 - v(R)]. \quad (14)$$

This lower bound has a zero at $R^* = \Gamma^{1/d}(1 + d/2)/\sqrt{\pi}$, which increases as \sqrt{d} for large d .

2.5. The quantizer problem

A d -dimensional quantizer is a device that takes as an input a point at position \mathbf{x} in \mathbb{R}^d generated from some probability density function $p(\mathbf{x})$ and outputs the nearest point \mathbf{r}_j of a known point pattern to \mathbf{x} [9]. The quantizer problem is then to choose the point pattern to minimize the scaled dimensionless error $\mathcal{G} = \langle R^2 \rangle / d$, where $\langle R^2 \rangle$ is the second moment of the nearest-neighbor distribution function for the void space external to the particles in the point process. Specifically, we define the void nearest-neighbor density function $H_V(R)$ such that $H_V(R)dR$ is the probability of finding the nearest particle of a point pattern with respect to an arbitrary point \mathbf{x} of the void space within a radial distance $R + dR$ from \mathbf{x} .

The void exclusion probability function $E_V(R)$ is then the complementary cumulative distribution function associated with $H_V(R)$ [34]:

$$E_V(R) = 1 - \int_0^R H_V(r) dr. \quad (15)$$

Using integration by parts, one can then show that

$$\mathcal{G} = \frac{1}{d} \int_0^{+\infty} R^2 H_V(R) dR \quad (16)$$

$$= \frac{2}{d} \int_0^{+\infty} R E_V(R) dR. \quad (17)$$

The quantizer error therefore depends sensitively on the shape of the void-space distribution. This situation is distinct from the covering problem, which is concerned only with the compact support of E_V .

Using upper and lower bounds on E_V , Torquato has been able to re-derive Zador's bounds for the minimum scaled dimensionless error [1]:

$$\frac{\Gamma^{2/d}(1 + d/2)}{\pi(d + 2)} \leq \mathcal{G}_{\min} \leq \frac{\Gamma^{2/d}(1 + d/2)\Gamma(1 + 2/d)}{\pi d}. \quad (18)$$

These bounds converge in asymptotically high dimensions, implying

$$\mathcal{G}_{\min} \rightarrow (2\pi e)^{-1} \quad (d \rightarrow +\infty). \quad (19)$$

This convergence implies that lattices and disordered point patterns are equally good quantizers in asymptotically high dimensions. Using known results for sphere packings, Torquato has also presented an improved upper bound to the minimal quantizer error [1], which is generally appreciably tighter than Zador's upper bound for low to moderately high dimensions and converges to the exact asymptotic result (19) in high dimensions.

2.6. Comparison of the packing, number variance, covering and quantizer problems

In his study of the best solutions of the covering and quantizer problems in up to 24 dimensions, Torquato [1] compared these results to the best known solutions for the sphere packing and number variance problems. In \mathbb{R} and \mathbb{R}^2 , it is well known that the integer lattice \mathbb{Z} and the triangular lattice, respectively, possess simultaneously the maximal packing density, the minimal asymptotic number variance, the minimal covering radius and the minimal quantizer error [9, 1]. However, the solutions to these problems are no longer the same in as low as three dimensions. Although the FCC lattice generates the densest sphere packing in three dimensions [11], its dual lattice BCC minimizes the three-dimensional covering radius, quantizer error and asymptotic number variance. To understand these differences, Torquato has shown that, while the number variance, covering and quantizer problems are described by soft, bounded interactions, the packing problem is described by a short-ranged pair potential that is zero whenever two spheres do not overlap and infinite when they do [1]. Furthermore, although the number variance problem can be interpreted as the determination of the ground state of a short-ranged soft pair interaction [2, 1], the covering and quantizer problems involve one-body, two-body, three-body and higher-order interactions [1]. Therefore, for $d \geq 4$, the solutions for each of these problems are not necessarily the same. One notable exception occurs in \mathbb{R}^{24} , where the Leech lattice Λ_{24} [9] likely provides the globally optimal solution for all four problems [1]. It is currently unknown whether such globally optimal solutions exist for dimensions other than $d = 1, 2$ and 24. It was shown [1] that disordered saturated sphere packings provide both good coverings and quantizers in relatively low dimensions and may even surpass the best known lattice coverings and quantizers in these dimensions. We shall return to this point in section 4.

3. High-dimensional generalizations of the kagomé and diamond crystals

Our constructions of the d -dimensional generalizations of the kagomé and diamond crystals will involve an underlying A_d Bravais lattice structure. All angles between the basis vectors for the A_d lattice are $\pi/3$ radians, implying that $\mathbf{e}_j \cdot \mathbf{e}_k = a^2/2$ for all $j \neq k$, where a is the magnitude of each basis vector \mathbf{e}_j .⁶ It is therefore possible to identify a coordinate system in which the generator matrix M_{A_d} is triangular. The two-dimensional A_2 lattice is the usual triangular lattice, which is the known densest packing in \mathbb{R}^2 . Similarly, the A_3 lattice is one representation for the FCC lattice, which is the densest packing in three

⁶ Using these properties, one can recursively write down the basis vectors \mathbf{e}_j for the A_d lattice. For example, assuming $a = 1$, we have in two dimensions $\mathbf{e}_1 = (1, 0)^T$ and $\mathbf{e}_2 = (1/2, \sqrt{3}/2)^T$. The basis vectors for $d = 3$ are $\mathbf{e}_1 = (1, 0, 0)^T$, $\mathbf{e}_2 = (1/2, \sqrt{3}/2, 0)^T$ and $\mathbf{e}_3 = (1/2, \sqrt{3}/6, \sqrt{6}/3)^T$. The $d = 4$ vectors are $\mathbf{e}_1 = (1, 0, 0, 0)^T$, $\mathbf{e}_2 = (1/2, \sqrt{3}/2, 0, 0)^T$, $\mathbf{e}_3 = (1/2, \sqrt{3}/6, \sqrt{6}/3, 0)^T$ and $\mathbf{e}_4 = (1/2, \sqrt{3}/6, \sqrt{6}/12, \sqrt{10}/4)^T$.

dimensions [11]. However, for $d \geq 4$, the A_d is no longer optimally dense, even among Bravais lattices.

3.1. The d -dimensional diamond crystal

The fundamental cell for the A_d lattice is a regular rhombotope, the d -dimensional generalization of the two-dimensional rhombus and three-dimensional rhombohedron. Therefore, the points $\{\mathbf{0}\} \cup \{\mathbf{e}_j\}_{j=1}^d$, where \mathbf{e}_j denotes a basis vector of the A_d lattice, are situated at the vertices of a regular d -dimensional simplex. The d -dimensional diamond crystal can therefore be obtained by including in the fundamental cell the centroid of this simplex:

$$\boldsymbol{\nu} = \frac{1}{d+1} \sum_{j=1}^d \mathbf{e}_j, \quad (20)$$

resulting in a periodic crystal with two points per fundamental cell. By construction, the number of nearest neighbors to each point in the d -dimensional diamond crystal is $d+1$, corresponding to one neighbor for each vertex of the regular simplex. One can verify by translation of the fundamental cell that all points of the d -dimensional diamond crystal are topologically equivalent. Note that the two-dimensional diamond crystal is the usual honeycomb lattice, in which each point is at the vertex of a regular hexagon.

We mention that our construction of the diamond crystal is distinct for all $d \neq 3$ from the D_d^+ structure mentioned by Conway and Sloane [9]. The D_d lattice is obtained by placing points using a ‘checkerboard’ pattern in \mathbb{R}^d [9]:

$$D_d = \left\{ (x_1, \dots, x_d) \in \mathbb{Z}^d : \sum_{j=1}^d x_j = 2m \text{ for some } m \in \mathbb{Z} \right\}. \quad (21)$$

The structure D_d^+ is then obtained by including the translate vector $\boldsymbol{\nu} = (1/2, 1/2, \dots, 1/2)$ in the fundamental cell. Although in three dimensions the D_d^+ structure does provide an equivalent construction of the diamond crystal, the relationship to our structure does not hold for any other dimension. Indeed, D_d^+ is a Bravais lattice for all even dimensions, which is not true for our construction of the d -dimensional diamond crystal. For example, in two dimensions, D_2^+ is equivalent to a rectangular lattice with generator matrix

$$M_{D_2^+} = \begin{pmatrix} a/2 & 0 \\ 0 & a \end{pmatrix}, \quad (22)$$

where a determines the fundamental cell size. Each point in this structure possesses two nearest neighbors and is therefore distinct from the honeycomb crystal, in which the coordination number of each particle is three.

3.2. A d -dimensional kagomé crystal

The two-dimensional kagomé crystal is obtained by placing points at the midpoints of each nearest-neighbor bond in the honeycomb crystal, resulting in a non-Bravais lattice with three particles per fundamental cell. Similarly, the three-dimensional kagomé crystal,

also known as the pyrochlore crystal [37], can be constructed by placing points at the midpoints of each nearest-neighbor bond in the three-dimensional diamond crystal. We therefore generalize the kagomé crystal to higher dimensions using the aforementioned construction of the d -dimensional diamond crystal, placing points at the midpoints of each nearest-neighbor bond. With respect to the underlying A_d Bravais lattice structure, these points are located at

$$\mathbf{x}_0 = \boldsymbol{\nu}/2 \quad (23)$$

$$\mathbf{x}_j = \boldsymbol{\nu} + \boldsymbol{\eta}_j/2 \quad (j = 1, \dots, d), \quad (24)$$

where

$$\boldsymbol{\eta}_j = \mathbf{e}_j - \boldsymbol{\nu} \quad (25)$$

denotes a ‘bond vector’ of the d -dimensional diamond crystal. By translating the fundamental cell such that the origin is at \mathbf{x}_0 , we can also represent the d -dimensional kagomé crystal as $A_d \oplus \{\mathbf{v}_j\}$, where

$$\mathbf{v}_j = \mathbf{e}_j/2 \quad (j = 1, \dots, d). \quad (26)$$

The d -dimensional kagomé crystal therefore has $d+1$ points per fundamental cell, growing linearly with dimension. Each point of the d -dimensional kagomé crystal is at the vertex of a regular simplex obtained by connecting all nearest neighbors in the structure, implying that each point possesses $2d$ nearest neighbors in d Euclidean dimensions [38]⁷. We illustrate our constructions of the two-dimensional kagomé and diamond (honeycomb) crystals in figure 1.

3.3. Other high-dimensional kagomé crystals

Our simple construction of the d -dimensional kagomé crystal suggests that there exists a large family of ‘kagomé-like’ crystals obtained by including the midpoints of the basis vectors for a Bravais lattice within the fundamental cell. A simple example is to include basis-vector midpoints into the d -dimensional integer lattice \mathbb{Z}^d . A more interesting example is obtained in \mathbb{R}^4 by including the midpoints of the basis vectors for the D_4 lattice, the densest known packing in four dimensions, into the fundamental cell. The packing density of the resulting structure is $\phi' = 5\pi^2/256 \approx 0.1928$, which should be compared to the density of the four-dimensional kagomé crystal $\phi = \sqrt{5}\pi^2/128 \approx 0.1724$. Note, however, that this kagomé-like structure does not possess the same relationship to the d -dimensional diamond crystal as our construction above, and we therefore focus the remainder of the discussion on the more natural generalization of the kagomé crystal in terms of vertex-sharing simplices in Euclidean space \mathbb{R}^d .

⁷ Our constructions of the d -dimensional kagomé and diamond lattices were suggested in the article by Torquato and Stillinger and are equivalent to, but somewhat simpler than, similar constructions given by O’Keeffe. However, while Torquato and Stillinger were interested in the mathematically precise notion of jamming in hard-sphere packings, O’Keeffe’s work makes reference only to so-called ‘rare’ sphere packings, which are distinct from jammed packings. Neither paper considers the covering or quantizer problems for these structures nor references a decorrelation principle for periodic lattices.

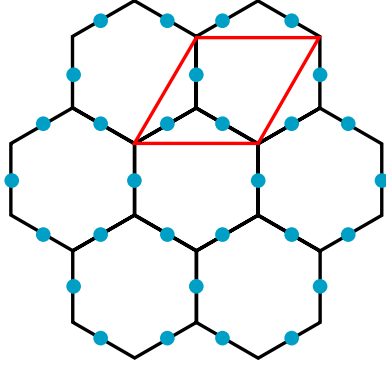


Figure 1. Portion of the honeycomb crystal (two-dimensional diamond) with the A_2 fundamental cell (rhombus). The points of the honeycomb crystal are the vertices of the regular hexagons. The kagomé crystal (circular points) is then constructed from the midpoints of the bonds between nearest neighbors in the honeycomb crystal. The kagomé crystal consists of vertex-sharing simplices, the centroids of which recover the honeycomb crystal.

4. Structural properties of the high-dimensional kagomé and diamond crystals

4.1. Packing densities and coordination numbers

The packing density associated with a periodic point pattern is the maximal fraction of space that can be occupied by decorating each of the points with a sphere of radius R_P , where R_P is the packing radius, defined as the maximal value of R for which $E_V(R)$ exactly obtains its one-point lower bound at unit number density ρ :

$$R_P \equiv \sup_{\rho=1} \{R : E_V(R) = 1 - v(R)\}. \quad (27)$$

Note that this definition is consistent with our discussion of the packing radius in section 2.3, involving decorating each of the points in a point pattern with a sphere of maximal radius R_P such that none of the resulting spheres overlap. However, the definition (27) helps to elucidate the remarkable connections among the packing, covering, quantizer and number variance problems. For lattices, this formulation is equivalent to identifying the minimal lattice vector at unit number density, which can be obtained from the in-radius of the Voronoi cell for a given lattice point [9]. Note that we have the following weak upper bound on the packing radius for any Euclidean dimension d :

$$R_P \leq \Gamma^{1/d}(1 + d/2)/\sqrt{\pi} \leq R_C, \quad (28)$$

where R_C is the covering radius. Substantially improved upper bounds [39] and conjectural lower bounds [12] have been provided for the packing radii of the densest sphere packings in any Euclidean dimension d .

To calculate the packing density of the d -dimensional kagomé crystal, we first consider the A_d Bravais lattice at unit number density, which has a known packing density [9]

$$\phi_{A_d} = v(R_P^{(A_d)}) = \frac{\pi^{d/2}}{2^{d/2}\Gamma(1 + d/2)\sqrt{d+1}}, \quad (29)$$

where $R_{\text{P}}^{(A_d)}$ is the corresponding packing radius:

$$2R_{\text{P}}^{(A_d)} = \left(\frac{2^{d/2}}{\sqrt{d+1}} \right)^{1/d}. \quad (30)$$

By construction, the d -dimensional kagomé crystal has the same fundamental cell with $d+1$ particles, and the associated packing radius is

$$R_{\text{P}}^{(\text{Kag}_d)} = (2^{d/2} \sqrt{d+1})^{1/d} / 4 = (d+1)^{1/d} R_{\text{P}}^{(A_d)} / 2. \quad (31)$$

Therefore

$$\phi_{\text{Kag}_d} = v(R_{\text{P}}^{(\text{Kag}_d)}) = (d+1) \phi_{A_d} / 2^d \quad (32)$$

$$= \frac{\pi^{d/2} \sqrt{d+1}}{2^{3d/2} \Gamma(1+d/2)}. \quad (33)$$

The packing density of the d -dimensional diamond crystal can be calculated similarly. In particular, the packing radius of the diamond crystal in d Euclidean dimensions is

$$R_{\text{P}}^{(\text{Dia}_d)} = \|\boldsymbol{\nu}\|/2 = 2^{1/d} \sqrt{\frac{d}{2(d+1)}} R_{\text{P}}^{(A_d)}, \quad (34)$$

where $\boldsymbol{\nu}$ is the translate vector (20) corresponding to the centroid of the regular simplex formed by the basis vectors for the A_d Bravais lattice. The norm of this translate vector can be evaluated by induction and the recursion relation

$$K_d = \left\| \sum_{j=1}^d \mathbf{e}_j \right\| = \sqrt{K_{d-1}^2 + d}. \quad (35)$$

It follows that the packing density of the d -dimensional diamond crystal is

$$\phi_{\text{Dia}_d} = 2 \left(\frac{d}{2(d+1)} \right)^{d/2} \phi_{A_d} \quad (36)$$

$$= \frac{(\pi d)^{d/2}}{2^{d-1} (d+1)^{(d+1)/2} \Gamma(1+d/2)}. \quad (37)$$

Figure 2 compares the packing densities of the d -dimensional kagomé and diamond crystals for increasing dimension d . It is interesting to note that, for $d \leq 3$, the kagomé crystal is a denser packing than the diamond crystal; however, this trend reverses for all $d \geq 4$. We will argue in the following sections that this behavior is related to the distribution of the void space external to the particles in the lattices. Specifically, the d -dimensional kagomé structure has increasingly large holes within the fundamental cell, skewing the void exclusion probability function $E_{\text{V}}(R)$ to higher values of R . This behavior implies that the kagomé crystal is effectively ‘filamentary’ in asymptotically high dimensions, in the sense that the net of bonds between nearest neighbors consists of strands that branch at each point of the crystal but are separated by increasingly large holes within the fundamental cell. While this argument should also hold for the

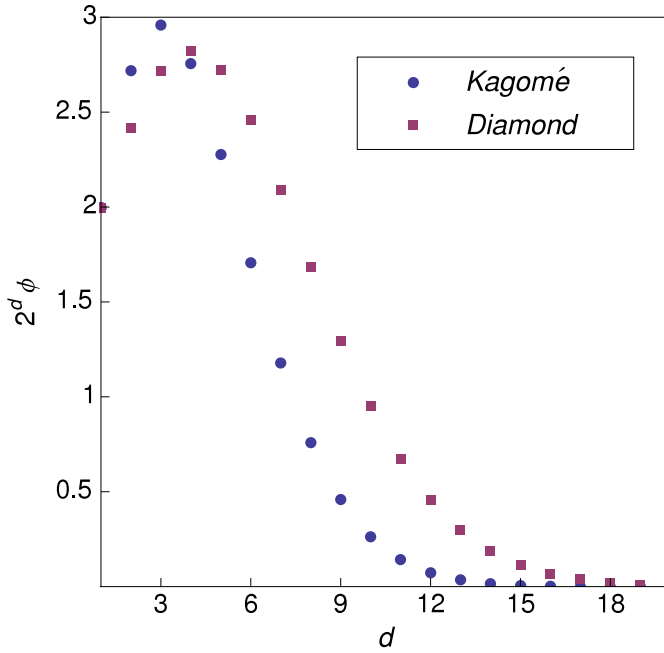


Figure 2. Scaled packing densities $2^d \phi$ of the d -dimensional kagomé and diamond crystals.

d -dimensional diamond crystal, the placement of a particle at the centroid of the regular simplex formed by the basis vectors for the fundamental cell apparently prevents the lattice holes from growing more rapidly than in the kagomé structure.

We have also determined the coordination numbers for both the d -dimensional diamond and kagomé crystals up to at least the first 100 coordination shells. Such calculations are helpful in the evaluation of lattice sums for these structures and provide insight into the coordination structure of the crystals [2, 38] (see footnote 7). Table 1 provides abridged results up to $d = 5$. Note that, although both structures possess nearest-neighbor coordination numbers growing linearly with dimension, the kagomé crystal in high Euclidean dimensions has a much larger number of nearest neighbors than the diamond crystal. This observation implies that the kagomé crystal is a much more ‘branched’ structure (in the sense defined above) than the diamond crystal with highly coordinated particles separated over increasingly large length scales by holes in the fundamental cell. This observation also has implications for the number variance of the kagomé structure and the decorrelation principle for periodic point patterns, which we discuss in further detail in subsequent sections.

4.2. Void exclusion probabilities, covering radii and quantizer errors

As previously mentioned, the covering radius R_C and scaled dimensionless quantizer error \mathcal{G} can be determined from knowledge of the void exclusion probability function $E_V(R)$ of a point pattern, which contains information about the distribution of the void space external to the particles. This connection to E_V was first explicitly mentioned recently by Torquato [1] and we have been unable to find studies of this function for any periodic

Table 1. Coordination numbers for the d -dimensional diamond (Dia_d) and kagomé (Kag_d) crystals. The square-coordination distance r_k^2 is given in parentheses, followed by the number of neighboring particles Z_k at the distance r_k . The nearest-neighbor distance determines the length scale for each structure.

Shell number	Dia_2	Kag_2	Dia_3	Kag_3	Dia_4	Kag_4	Dia_5	Kag_5
1	(1) 3	(1) 4	(3) 4	(4) 6	(2) 5	(1) 8	(5) 6	(1) 10
2	(3) 6	(3) 4	(8) 12	(12) 12	(5) 20	(3) 24	(12) 30	(3) 40
3	(4) 3	(4) 6	(11) 12	(16) 12	(7) 30	(4) 20	(17) 60	(4) 30
4	(7) 6	(7) 8	(16) 6	(20) 12	(10) 30	(5) 48	(24) 90	(5) 120
5	(9) 6	(9) 4	(19) 12	(28) 24	(12) 30	(7) 72	(29) 90	(7) 200
6	(12) 6	(12) 6	(24) 24	(32) 6	(15) 60	(8) 30	(36) 140	(8) 90
7	(13) 6	(13) 8	(27) 16	(36) 18	(17) 80	(9) 56	(41) 240	(9) 190
8	(16) 3	(16) 6	(32) 12	(44) 12	(20) 60	(11) 96	(48) 270	(11) 360
9	(19) 6	(19) 8	(35) 24	(48) 24	(22) 60	(12) 60	(53) 210	(12) 140
10	(21) 12	(21) 8	(40) 24	(52) 36	(25) 120	(13) 144	(60) 360	(13) 520

crystal in the literature. Here, we determine E_V for the d -dimensional kagomé and diamond crystals and use our results to provide estimates for the covering radii and quantizer errors for these systems.

Our calculations involve Monte Carlo sampling of the void space within the fundamental cell for the underlying A_d Bravais lattice. Periodicity of the point pattern implies that E_V must have compact support, and it is therefore sufficient only to sample within a single fundamental cell, subject to periodic boundary conditions, to obtain the full distribution E_V . Noting that any point \mathbf{r} within the fundamental cell can be expressed as an appropriate linear combination of the Bravais lattice basis vectors:

$$\mathbf{r} = M_{A_d} \mathbf{x}, \quad (38)$$

where $\mathbf{x} = (x_j)_{j=1}^d$ with $0 \leq x_j \leq 1$ for all j , we can efficiently sample the void space by placing points randomly and uniformly in the d -dimensional unit cube and then mapping those points to the fundamental cell with the generator matrix M_{A_d} as in (38). The void exclusion probability function is then obtained by measuring nearest-neighbor distances between the sampling points and the particles of the crystal. Note that this calculation of the void exclusion probability function is more efficient than direct calculation of the Voronoi tessellation for the crystals in high dimensions, thereby providing a facile means of obtaining estimates for R_C and \mathcal{G} .

Our results are shown in figure 3. Table 2 summarizes our results for the covering radii and quantizer errors of the diamond and kagomé crystals. The d -dimensional kagomé crystal possesses relatively large covering radius in each dimension, implying that the covering of Euclidean space with the kagomé crystal involves much more than pairwise overlap potentials, even in two dimensions. This behavior follows directly from the increasing sizes of holes within the fundamental cell in high dimensions. Since all of the particles in the kagomé crystal are relegated to the boundary of the fundamental cell, the majority of the space in the fundamental cell is void space, thereby increasing the value of the covering radius relative to the A_d Bravais lattice.

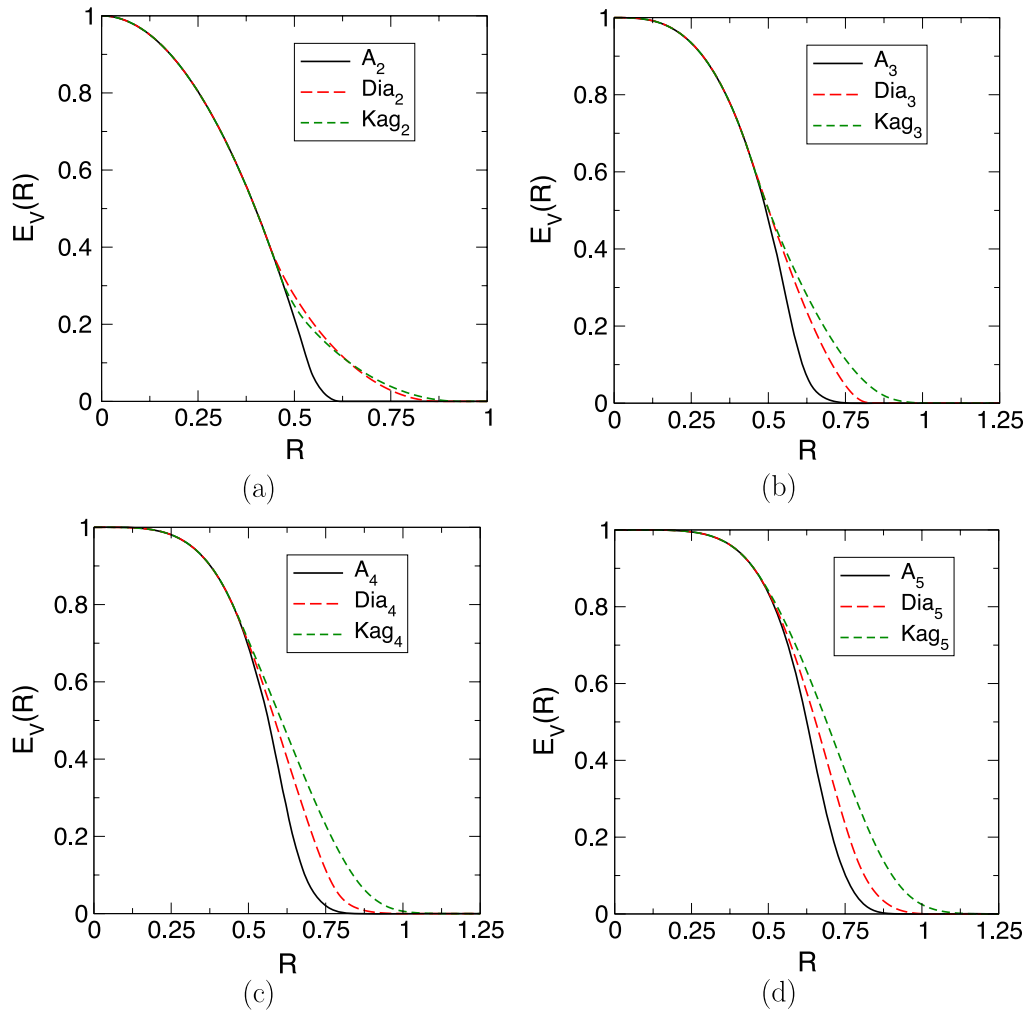


Figure 3. Void exclusion probability functions $E_V(R)$ for the A_d , d -dimensional diamond (Dia_d) and d -dimensional kagomé (Kag_d) crystals at unit number density: (a) $d = 2$. (b) $d = 3$, (c) $d = 4$ and (d) $d = 5$.

Table 2. Estimates of the covering radius R_C and quantizer error \mathcal{G} for the A_d , d -dimensional diamond Dia_d and d -dimensional kagomé Kag_d lattices. Errors for the calculations are ± 0.0004 for the covering radii and ± 0.00004 for the quantizer errors, as estimated by comparison with exact results for the A_d lattice in two and three dimensions. The covering radii for the A_2 , A_3 , Dia_2 and Kag_2 lattices are known exactly [9, 1], and these exact results are reported here; also reported are the exact values for the quantizer errors of the A_2 and A_3 lattices [1].

	$d = 2$		$d = 3$		$d = 4$		$d = 5$	
	R_C	\mathcal{G}	R_C	\mathcal{G}	R_C	\mathcal{G}	R_C	\mathcal{G}
A_d	0.6204	0.080 18	0.7937	0.078 75	0.8816	0.077 80	0.9984	0.077 69
Dia_d	0.8774	0.096 27	0.8640	0.091 12	1.0472	0.088 25	1.0776	0.086 49
Kag_d	0.9306	0.096 15	1.0384	0.099 25	1.2048	0.099 73	1.2824	0.099 39

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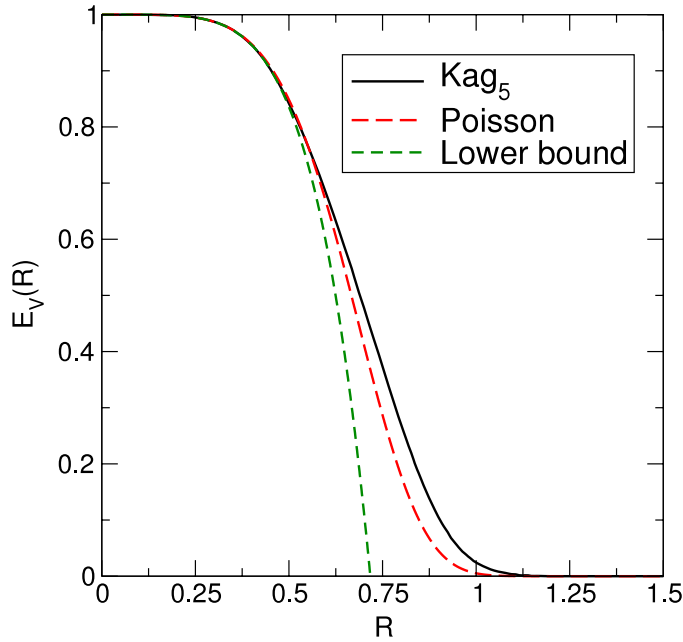


Figure 4. Void exclusion probability functions $E_V(R)$ at unit number density for the five-dimensional kagomé crystal and a disordered, uncorrelated Poisson point process. Also included is the one-point series lower bound $1 - v(R)$.

However, it is interesting to note that the quantizer error for the two-dimensional kagomé crystal is actually smaller than the associated error for the honeycomb (two-dimensional diamond) crystal. Indeed, we recall that the kagomé crystal generates a denser sphere packing in two dimensions than the honeycomb crystal, implying by definition that $E_V(R) = 1 - v(R)$ at unit density for a larger range in R . The void exclusion probability of the kagomé crystal is therefore relatively ‘tight’ compared to the honeycomb crystal in such a way that the longer tail does not substantially affect the first moment of the distribution. The two-dimensional kagomé crystal therefore provides an interesting example of how increasing the complexity of a crystal structure can conceivably improve the quantizer error; ‘simpler’ structures are not always better quantizers, even in low dimensions.

This behavior changes drastically in higher dimensions, where the quantizer error for the d -dimensional kagomé structure is unusually high relative to the d -dimensional diamond and A_d structures. Indeed, the bulk distribution of the void space for the five-dimensional kagomé crystal is seen to be larger than the corresponding curve for a Poisson-distributed point pattern (see figure 4), consisting of uncorrelated random points in Euclidean space. This unusual property implies that the quantizer error for the five-dimensional kagomé crystal is larger even than Zador’s upper bound (18) for the minimal quantizer error. It is highly counterintuitive that a disordered point pattern should be a better quantizer than a periodic crystal with relatively low complexity and in relatively low dimensions. Nevertheless, this observation is consistent with the prevalence of large void regions in the high-dimensional kagomé crystal and supports our description of this

system as being effectively ‘filamentary’ in high dimensions⁸. This result also suggests the onset of a decorrelation principle for the d -dimensional kagomé crystal, an issue we explore in more detail in section 5.

4.3. Number variance coefficients

As previously mentioned, the asymptotic scaling of the number variance provides a quantitative metric for the extent of order within a structure over asymptotically large length scales with respect to the mean nearest-neighbor separation between points [2, 3]. Since we are utilizing the d -dimensional kagomé crystal to probe the applicability of the decorrelation principle to periodic structures, it is therefore of interest to calculate the surface area coefficient B (cf, (9)) governing surface area fluctuations in the local number density. Note that periodicity of the fundamental cell implies the presence of full long-range order in both the d -dimensional kagomé and diamond crystals, which is sufficient to induce hyperuniformity.

Unfortunately, this long-range order also implies that the integral (9) diverges; however, Torquato and Stillinger have reformulated this expression using a convergence ‘trick’ [40] to ensure a properly convergent expression for periodic crystals [2]. Specifically, we rewrite the expression (9) for the coefficient B as

$$B = \lim_{\beta \rightarrow 0^+} \frac{-\rho\kappa(d)}{D} \int \exp(-\beta r^2) r [g_2(\mathbf{r}) - 1] \, \mathrm{d}\mathbf{r}, \quad (39)$$

where $\kappa(d) = \Gamma(1 + d/2)/\{\Gamma[(d + 1)/2]\Gamma(1/2)\}$ and $r = \|\mathbf{r}\|$. Expanding this integral implies that

$$B = \frac{\rho d \pi^{(d-1)/2}}{2D\beta^{(d+1)/2}} - \frac{\rho\kappa(d)}{D} \int \exp(-\beta r^2) r g_2(\mathbf{r}) \, \mathrm{d}\mathbf{r} \quad (\beta \rightarrow 0^+), \quad (40)$$

and the remaining integral involving the pair correlation function can be interpreted as the average pair sum for the pair interaction $v(r) = \exp(-\beta r^2)r$ over the underlying crystal structure, which is convergent for all $\beta > 0$. Writing the average pair sum explicitly, we find

$$B = \frac{\rho d \pi^{(d-1)/2}}{2D\beta^{(d+1)/2}} - \frac{\kappa(d)}{ND} \sum'_{j,\ell,\mathbf{p}} \exp(-\beta \|\mathbf{p} + \boldsymbol{\nu}_j - \boldsymbol{\nu}_\ell\|^2) \|\mathbf{p} + \boldsymbol{\nu}_j - \boldsymbol{\nu}_\ell\| \quad (\beta \rightarrow 0^+), \quad (41)$$

where the prime on the summation means that the vector $\mathbf{p} = \mathbf{0}$ is excluded when $\boldsymbol{\nu}_j = \boldsymbol{\nu}_\ell$. To remove the dependence of B on the length scale D , we report the scaled coefficient $\eta^{1/d}B$, where $\eta = \rho v(D/2)$, as has previously been done in the literature [2, 3].

Table 3 reports our results for the number variance coefficients of the d -dimensional diamond and kagomé crystals. It is helpful to compare these results to similar calculations performed for certain so-called g_2 -invariant processes [28]. A g_2 -invariant process involves constraining a chosen non-negative form for the pair correlation function g_2 to remain invariant over a nonvanishing density range while keeping all other relevant macroscopic

⁸ Note that E_V for a Poisson point pattern decays exponentially as $E_V(R) = \exp[-\rho v(R)]$, implying that this structure possesses an infinite covering radius. However, the long tail of this function does not contribute substantially enough for the quantizer error to be larger than the kagomé lattice in sufficiently high dimensions.

Table 3. Number variance coefficients $\eta^{1/d}B$ for the A_d , d -dimensional kagomé Kag_d and d -dimensional diamond Dia_d crystals. Here, we have taken η to be the packing density of the structure. The last two entries correspond to g_2 -invariant processes as discussed in the text. The approximate error for each entry is ± 0.00005 by comparison with previously reported results [2, 3].

d	A_d	Kag_d	Dia_d	Step function	Step + delta-function
2	0.12709	0.14675	0.14176	0.21221	0.15005
3	0.15569	0.20740	0.17737	0.28125	0.19086
4	0.17734	0.27330	0.20555	0.33953	0.22342
5	0.19579	0.35412	0.23144	0.39063	0.25092

variables fixed [28]. We consider the following two examples of g_2 -invariant processes: the so-called ‘step function g_2 ’, in which the pair correlation function has the form

$$g_2(r) = \Theta(r - D) \quad (42)$$

for some length scale D and the ‘step + delta function g_2 ’, given by

$$g_2(r) = \Theta(r - D) + \frac{Z}{\rho s(D)} \delta(r - D), \quad (43)$$

where Z can be interpreted as an average contact coordination number [2]. Both of these processes correspond to disordered point patterns that are hyperuniform at the critical densities

$$\eta_c = 1/2^d \quad (\text{step-function}) \quad (44)$$

$$\eta_c = (d + 1)/2^{d+1} \quad (\text{step + delta-function}). \quad (45)$$

Strong numerical evidence has been presented to suggest that these pair correlation functions are indeed realizable as point processes at the critical densities [41]. Torquato and Stillinger have used g_2 -invariant processes to define an optimization procedure that places lower bounds on the maximal sphere-packing density in d Euclidean dimensions [12, 28].

Torquato and Stillinger have analytically evaluated the number variance coefficients for these g_2 -invariant processes:

$$B_{\text{step}} = \frac{d^2 \Gamma(d/2)}{4 \Gamma[(d + 3)/2] \Gamma(1/2)} \quad (46)$$

$$2^d \eta_c B_{\text{delta+step}} = \frac{d^2 (d + 2) \Gamma(d/2)}{16 \Gamma[(d + 3)/2] \Gamma(1/2)}, \quad (47)$$

and these results are included in table 3. One notices that, for all $d \geq 3$, the d -dimensional kagomé crystal possesses a higher number variance coefficient than the step + delta-function process, suggesting that there exists a disordered configuration of points in high dimensions that is more ordered over asymptotically large length scales than this periodic structure. This result is surprising since points in the step + delta-function point

pattern are completely decorrelated from each other beyond the constrained hard-particle diameter. Furthermore, the average contact coordination number for this process is [2]

$$Z = d/2, \quad (48)$$

which is, for all dimensions d , less than the nearest-neighbor coordination number of the d -dimensional kagomé crystal, $Z_{\text{Kag}_d} = 2d$.

To understand this behavior, we first note that the packing density (45) is less than the corresponding density (33) for the d -dimensional kagomé crystal for all $d \leq 4$; however, for $d \geq 5$, the step + delta function process possesses a higher packing density than the kagomé crystal. This observation implies that the local ordering between points induced by the delta-function contribution to the pair correlation function of the step + delta-function process is sufficient to regularize the void space in such a way that the packing radius R_P remains relatively high compared to the kagomé structure. In particular, the large holes within the kagomé fundamental cell control the structural properties of the point pattern in high Euclidean dimensions and it is these holes that increase the asymptotic number variance coefficient in such a way that the point pattern can no longer be distinguished from correlated but disordered point patterns. This behavior is in accordance with an effective decorrelation between the points of the kagomé structure over large length scales and supports the presence of a decorrelation principle for this system.

It is important to note that the increasing nearest-neighbor coordination number of the d -dimensional kagomé crystal implies that correlations between nearest neighbors are increasing with increasing dimension. For this reason, the number variance coefficient governing surface area fluctuations is always smaller than the corresponding coefficient for the simple step-function process in any dimension; these constrained correlations are never removed by the dimensionality of the system. However, correlations over several nearest-neighbor distances apparently diminish in an effective manner, which we make more precise in section 5, and it is this type of decorrelation that we claim is responsible for unusually large asymptotic local-number-density fluctuations in the kagomé structure. Note also that these results are consistent with our analysis of the quantizer errors for the d -dimensional kagomé crystals.

5. The decorrelation principle for periodic point patterns

5.1. Universality of decorrelation in high dimensions

The decorrelation principle [12] states that unconstrained asymptotic n -particle correlations vanish in sufficiently high dimensions, and all higher-order ($n \geq 3$) correlation functions can be expressed in terms of the pair correlation function within some small error. Although originally stated in the context of hard-sphere packings, certain ‘soft’ many-particle distributions, including points interacting in the Gaussian core model [14] and noninteracting spin-polarized ground-state fermions [15], are also known to exhibit this effect, even in relatively low dimensions $d = 1$ –6. No rigorous proof for this principle has been found to date, but it is based on strong theoretical arguments and has been shown to be remarkably robust in theoretical and numerical studies.

Does the decorrelation principle apply in some generalized sense to periodic crystals? It is not trivial to extend the decorrelation principle to periodic crystals, which inherently

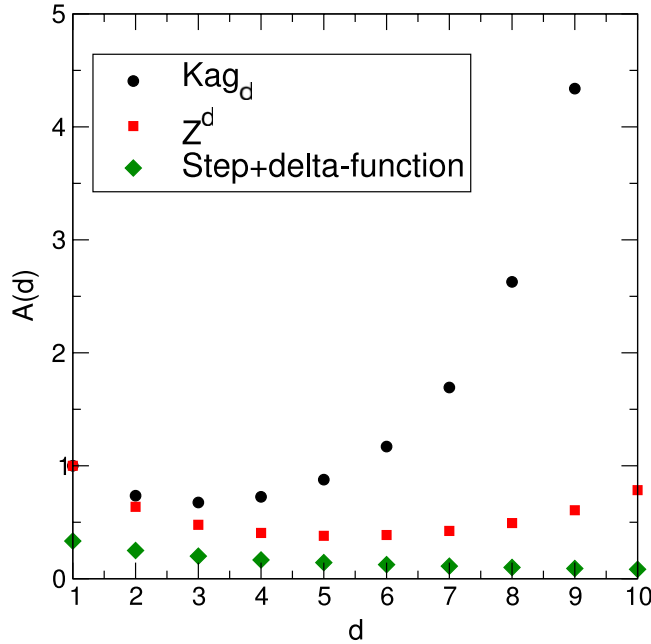


Figure 5. Intensity $A(d) = Z_1(d)/(2^d d \phi)$ associated with the first δ -function peaks of the pair correlation functions for the d -dimensional kagomé and hypercubic \mathbb{Z}^d crystals. Also shown for comparison is the result for the g_2 -invariant step + delta-function process (43).

possess long-range order owing to the regular arrangement of points within a lattice structure. This full long-range order induces deterministic correlations as manifested by Bragg peaks in the power spectrum. In particular, we recall from (3) that the angularly averaged pair correlation function consists of consecutive Bragg peaks at each coordination shell; it is convenient to express this relation in terms of the packing fraction ϕ and associated packing diameter D :

$$g_2(r) = \sum_{k=1}^{+\infty} \frac{Z_k}{2^d d \phi} \left(\frac{D}{r_k} \right)^{d-1} \delta[(r - r_k)/D]. \quad (49)$$

The intensity of each peak in the pair correlation function is therefore determined by the coordination number Z_k of the k th coordination shell, the packing density ϕ and the distance r_k to the k th coordination shell.

It is interesting to examine the behavior of the intensity

$$A(d) = Z_1(d)/(2^d d \phi) \quad (50)$$

associated with the first peak of the pair correlation function for the d -dimensional kagomé and hypercubic \mathbb{Z}^d crystals, shown in figure 5. Note that both of these crystals possess a nearest-neighbor contact number $Z_1(d) = 2d$, equivalent to the isostatic condition⁹.

⁹ Isostaticity, where $Z = 2d_f$ with d_f equal to the number of degrees of freedom per particle, has been closely linked to the notions of ‘generic’ or ‘random’ jammed sphere packings. However, for packings with high symmetry, including the hypercubic \mathbb{Z}^d and kagomé crystals, the isostatic condition does not imply collective or strict jamming. See [13] for a more complete discussion of this fundamentally important point.

After an initial drop in relatively low dimensions, the intensity $A(d)$ increases without bound for both of the periodic systems. Furthermore, the d -dimensional kagomé crystal possesses a first-shell intensity that grows much more rapidly with dimensions than even the hypercubic lattice \mathbb{Z}^d , which is a direct consequence of the exponentially diminishing packing density and the prevalence of large holes in the fundamental cell. In both cases, nearest-neighbor correlations asymptotically increase with dimension d and it is therefore unclear whether a decorrelation principle should hold for periodic crystals. This behavior should be contrasted with corresponding results for the disordered g_2 -invariant step + delta-function process (43), where the first-peak intensity $A(d) = 1/(d+2)$ diminishes for all dimensions d .

Nevertheless, one can consider a disordered point pattern to be a realization of a non-Bravais lattice with a large number of particles randomly distributed in the fundamental cell. This observation suggests that periodic crystals with an M -particle basis growing with dimension may exhibit the same decorrelation properties as a disordered many-particle distribution. If this notion is true, then the effects of decorrelation should then be readily observed even in relatively low dimensions as with disordered packings [12, 16, 17, 14, 15, 4]. It is therefore intriguing to test the decorrelation principle for the d -dimensional kagomé lattice which, as previously mentioned, possesses $d + 1$ particles per fundamental cell.

The deterministic long-range order of a periodic crystal implies that the decorrelation principle, if it applies, cannot be directly observed from the pair correlation function (49) itself, but rather from some smoothed form of g_2 . Instead, the pair correlation function of a crystal must be interpreted in the sense of distributions [42]; it gains physical meaning only when integrated with an admissible function. Therefore, the appropriate function to consider is the ‘smoothed’ pair correlation function:

$$g_2^{(a)}(r) = \sum_{k=1}^{+\infty} \frac{Z_k D}{2^d d \phi a \sqrt{\pi}} \left(\frac{D}{r_k} \right)^{d-1} \exp\{-[(r - r_k)/a]^2\}, \quad (51)$$

corresponding to a convolution of (49) with a Gaussian kernel [42, 43]. Note that $g_2^{(a)}(r) \rightarrow g_2(r)$ for $r \in [0, +\infty)$ (in the sense of distributions) as $a \rightarrow 0$.

Figure 6 compares the smoothed pair correlation functions for the $d = 2$ and 4 kagomé lattices. Remarkably, asymptotic pair correlations are observed to diminish even in the relatively low dimensions shown (as with disordered point patterns [12, 14, 15, 4, 16, 17]), implying that the pair correlation function approaches its asymptotic value of unity in sufficiently high dimensions. Importantly, this effect at large pair separations is observed for any nonzero choice of the smoothing parameter a with only qualitative differences in the pair correlation function, corresponding to localization of the δ -function peaks. Our results therefore suggest that the decorrelation principle applies to the d -dimensional kagomé crystal in the sense that any delocalization of the local density field is sufficient to cause asymptotic pair correlations to diminish with respect to increasing dimension. Note that these observations are consistent with our calculations for the asymptotic number variance coefficient for the kagomé crystal, which is higher than the corresponding result for the disordered step + delta-function process even in low dimensions.

Additional calculations suggest that this approach to the decorrelation principle for periodic structures is applicable even to crystals with M -particle bases that do not increase with dimension. Figure 7 provides calculations of the smoothed pair correlation function

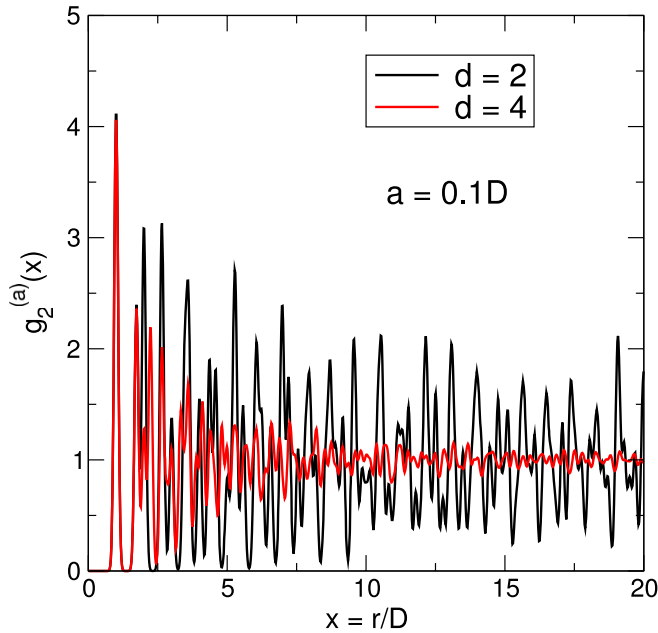


Figure 6. Smoothed pair correlation function for the kagomé crystal in dimensions $d = 2$ and 4 . The smoothing parameter $a = 0.1D$ (cf (51)).

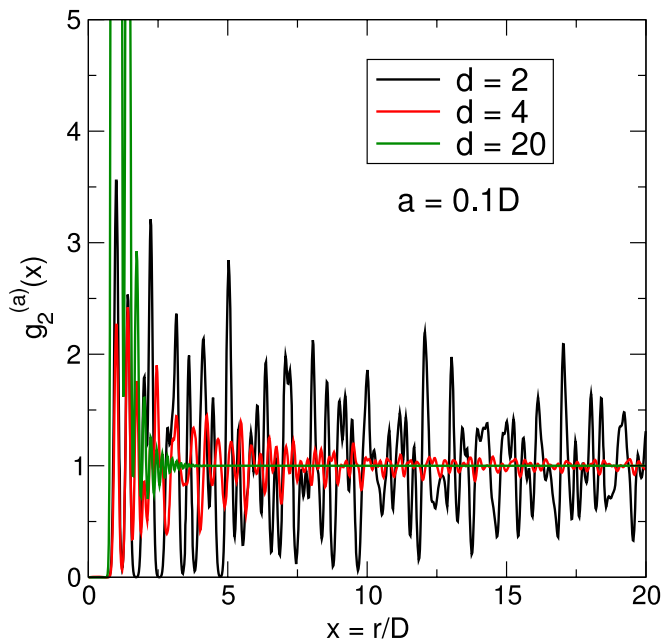


Figure 7. Smoothed pair correlation function for the hypercubic Bravais lattice \mathbb{Z}^d in dimensions $d = 2, 4$ and 20 . The smoothing parameter $a = 0.1D$.

for the hypercubic Bravais lattice \mathbb{Z}^d . Like the kagomé crystal, decorrelation is readily apparent even in low dimensions, and upon reaching $d = 20$ the system is essentially completely decorrelated beyond a few nearest-neighbor distances. Since any point pattern, disordered or not, can be modeled as a periodic point pattern, potentially with a

large number of points per fundamental cell, these observations support the remarkable statement that the decorrelation principle is a universal feature of high-dimensional point patterns, including those distributions associated with sphere packings. In particular, the principle should apply not only to disordered point patterns as originally discussed by Torquato and Stillinger [12] but also to periodic crystals.

The smoothing operation that we have introduced for the pair correlation function allows us to observe the effects of decorrelation in periodic crystals in relatively low dimensions. In asymptotically high dimensions, the widths of the Gaussians can be made arbitrarily small since consecutive coordination shells are tightly clustered. Decorrelation is therefore a fundamental feature of the pair correlation function itself of a high-dimensional periodic point pattern, whether it is a simple Bravais lattice or a crystal with many points per fundamental cell. This principle supports the claim that higher-order correlation functions do not provide additional information beyond that contained in g_2 , meaning that the pair correlations alone completely determine the packing in high dimensions.

5.2. Implications for the maximal density of sphere packings

The onset of decorrelation in high dimensions for periodic crystals has important implications for optimal lower bounds on the maximal sphere-packing density. Minkowski provided a nonconstructive proof that the asymptotic behavior of the maximal density of sphere packings is bounded from below by [12, 44]

$$\phi_{\max} \gtrsim \frac{1}{2^d} \quad (d \rightarrow +\infty). \quad (52)$$

This scaling is quite distinct from the Kabatiansky–Levenshtein upper bound on the maximal sphere-packing density [45]

$$\phi_{\max} \leq \frac{1}{20.5990^d} \quad (d \rightarrow +\infty). \quad (53)$$

Utilizing the decorrelation principle for disordered sphere packings, Torquato and Stillinger derived a conjectural lower bound on the maximal sphere-packing density that provides putative exponential improvement over Minkowski’s bound [12]:

$$\phi_{\max} \gtrsim \frac{d^{1/6}}{20.77865\dots^d} \quad (d \rightarrow +\infty). \quad (54)$$

This bound was derived using the aforementioned g_2 -invariant optimization procedure for a ‘test’ pair correlation function that in the high-dimensional limit becomes a step + delta function. It is a conjectural bound because it has yet to be shown that such a pair correlation function with packing density (54) is realizable by a point process, an issue to which we will return. The gap between the Kabatiansky–Levenshtein upper bound and the Torquato–Stillinger lower bound remains relatively large in high dimensions, and it is therefore an open problem to determine which bound provides the ‘correct’ asymptotic scaling.

To gain some insight into this issue, we note that in sufficiently high dimensions the distances between subsequent coordination shells become increasingly small, implying that the smoothing parameter a used to observe the decorrelation effect in the pair

correlation function does not need to be chosen very large. In the asymptotic dimensional limit, it follows that any choice of the smoothing parameter is sufficient to ‘collapse’ the pair correlation function onto its asymptotic value of unity with the exception of nearest-neighbor correlations, which are dominant in high dimensions. We therefore emphasize that the smoothing operation we have employed in this work is only a convenient tool that allows us to observe the decorrelation principle in even relatively low dimensions. The decorrelation principle itself is apparently a fundamental and universal phenomenon of any high-dimensional point pattern, ordered or not, manifested in the pair correlation function since higher-order correlation functions do not introduce additional information [12]. In summary, decorrelation suggests that the pair correlation functions of general periodic point patterns tend to the step + delta-function form, which is precisely the same asymptotic form as the test function that Torquato and Stillinger used to obtain the lower bound (54) [12, 46]. The appendix contains an analytical demonstration of the decorrelation effect for the previously mentioned g_2 -invariant step-function and step + delta-function processes.

However, the asymptotic scaling of the packing density for a sphere packing will depend inherently on the manner in which the pair correlation function approaches this asymptotic form. Since the dominant correlations in asymptotically high dimensions will be from nearest neighbors in a sphere packing, owing to the well-defined exclusion region in the pair correlation function, the decorrelation principle suggests that all sphere packings in high dimensions possess pair correlation functions of an effective step + delta-function form, which is precisely the same asymptotic form as the test function that Torquato and Stillinger used to obtain the lower bound (54) [12, 46]. The intensity $A(d)$ of the associated delta-function peak is given by (50). Whether this intensity increases (as with the d -dimensional kagomé and hypercubic \mathbb{Z}^d crystals) or diminishes (as with the g_2 -invariant step + delta-function process at its critical density (45)) in asymptotically high dimensions therefore depends on the relative scalings of $Z(d)$ and $\phi(d)$.

Using the same linear programming techniques introduced in [12], Scardicchio, Stillinger and Torquato have numerically explored the $Z(d)$ – $\phi(d)$ parameter space associated with the step + delta-function process when hyperuniformity is not enforced *a priori* (as it is at the critical density (45)) [46]. Their results provide the same exponential improvement on Minkowski’s lower bound for the maximal sphere-packing density as the Torquato–Stillinger lower bound (54). Additionally, this scaling is robust in the sense that it is recovered for test pair correlation functions containing any number of delta-function peaks [46]. This latter observation implies that next-nearest-neighbor correlations, even if they persist in high dimensions, do not provide additional exponential improvement over the Torquato–Stillinger lower bound (54) for the maximal sphere-packing density.

Since the g_2 -optimization procedure identifies the maximal packing density obtainable with the step + delta-function form, which is also obtained by high-dimensional periodic sphere packings by the decorrelation principle, our results support the remarkable possibility that the Torquato–Stillinger lower bounds may, in fact, be optimal in asymptotically high dimensions. If confirmed, this result would imply that the Kabatiansky–Levenshtein upper bound (53) therefore provides a suboptimal high-dimensional estimate. This conclusion is consistent with similar arguments put forth in [45]. If this claim is true, it is interesting to note that the results of Scardicchio, Stillinger and Torquato suggest that the intensity of the nearest-neighbor peak in the

pair correlation function of maximally dense sphere packings in high dimensions should diminish with increasing d [46], implying that the full pair correlation function completely decorrelates to a step-function form. It follows that certain periodic point patterns such as the d -dimensional kagomé and hypercubic \mathbb{Z}^d crystals cannot be maximally dense in high dimensions, thereby providing direct evidence that the manner in which the pair correlation function approaches the asymptotic step + delta-function form reflects the high-dimensional asymptotic scaling of the packing density. Indeed, the maximally dense sphere packings in high dimensions may therefore likely be disordered (i.e. with a pair correlation function decaying to unity sufficiently fast in the infinite-volume limit [12]) as first suggested by Torquato and Stillinger [12].

6. Concluding remarks

We have provided constructions of the high-dimensional generalizations of the kagomé and diamond crystals. The d -dimensional diamond crystal is obtained by including in the A_d fundamental cell the centroid of the regular simplex formed by the lattice basis vectors. The d -dimensional kagomé crystal can then be constructed by placing points at the midpoints of the ‘bonds’ in the diamond crystal. The kagomé crystal possesses a nearest-neighbor contact number $Z = 2d$ in d Euclidean dimensions, which is equivalent to the isostatic condition for jammed sphere packings. In two dimensions, the kagomé crystal is locally but neither collectively nor strictly jammed [47] under periodic boundary conditions; however, it can be reinforced to obtain the lowest-density strictly jammed subpacking of the triangular lattice. In three dimensions, the pyrochlore crystal has clustered equilateral-triangle vacancies. In contrast to $d = 2$, the d -dimensional kagomé crystal is therefore not strictly jammed for any $d \geq 3$ [38] (see footnote 7).

Using these constructions, we have derived analytically the packing densities of these structures and have shown that, while the kagomé crystal generates a denser sphere packing for $d = 2$ and 3, the diamond crystal is denser for all $d \geq 4$, at which point the holes in the kagomé lattice become substantially large. These observations are supported by numerical calculations for the void exclusion probabilities of the kagomé and diamond crystals. Surprisingly, the bulk of the void-space distribution for the kagomé lattice in dimensions $d \geq 5$ is larger than the corresponding result for the disordered, fully uncorrelated Poisson point pattern.

Our results have implications for the quantizer errors and covering radii of these structures in high dimensions. The diamond crystal provides a thinner covering of space than the kagomé crystal for all $d \geq 2$, even though the kagomé crystal is a better quantizer in two dimensions. However, the large holes in the fundamental cell for the kagomé lattice rapidly increase its quantizer error in high dimensions such that it even exceeds Zador’s upper bound and, therefore, also Torquato’s improved upper bound [1], in as low as $d = 5$. This observation implies that disordered point patterns can be better quantizers than certain periodic structures even in relatively low dimensions, which is consistent with the properties of certain disordered point patterns reported in [1].

We have also calculated the asymptotic surface area coefficients for the number variance of the kagomé and diamond crystals. Interestingly, the d -dimensional kagomé lattice possesses a larger asymptotic number variance coefficient even than the disordered step + delta-function g_2 -invariant process for all $d \geq 3$. Since the number variance

coefficient provides a quantitative measure of structural order over large length scales [2, 3], this result counterintuitively suggests that periodic crystals may possess less long-range structural order than prototypical ‘disordered’ point patterns in sufficiently high dimensions, which is consistent with a generalized decorrelation principle for periodic structures. By calculating a ‘smoothed’ pair correlation function for the d -dimensional kagomé crystal, we have provided direct evidence for the decorrelation principle in periodic point patterns. Indeed, the decorrelation principle appears to be universal, applying also to Bravais lattices as shown by corresponding calculations for the hypercubic lattice \mathbb{Z}^d in high dimensions.

Our work has important implications for the maximal sphere-packing density in high Euclidean dimensions. In particular, the suggested universality of the decorrelation principle for both disordered and periodic sphere packings suggests that the putative exponential improvement obtained by Torquato and Stillinger [12] on Minkowski’s lower bound for the maximal packing density is, in fact, optimal, which is consistent with previously reported results in the literature [46]. The pair correlation functions of high-dimensional sphere packings apparently possess a general step + delta-function form and optimization of the packing structure through the Z - ϕ parameter space [46], where Z is the mean nearest-neighbor contact number and ϕ is the packing density, suggests that maximally dense packings undergo a complete decorrelation in high Euclidean dimensions. In particular, the intensity of the nearest-neighbor peak in the pair correlation function diminishes in high dimensions, which should be contrasted with the corresponding behaviors for the d -dimensional kagomé and hypercubic \mathbb{Z}^d crystals. These latter structures therefore cannot be maximally dense in high dimensions, which is in accordance with the notion that the densest packings for asymptotically large d are, in fact, disordered [12]. Importantly, this work provides the foundation for a rigorous proof of the Torquato–Stillinger lower bound on the maximal-sphere packing density and its optimality in high dimensions. Future work is also warranted to explore the implications of the decorrelation principle for the covering, quantizer and number variance problems.

Acknowledgment

This work was supported by the Materials Research Science and Engineering Center (MRSEC) Program of the National Science Foundation under grant no. DMR-0820341.

Appendix. Analytical demonstration of the decorrelation principle for a simple example

Here we examine the step-function and step + delta-function g_2 -invariant processes at their critical densities (cf (42)–(45)). Our goal is to probe the manifestation of the decorrelation principle in delta-function contributions to the pair correlation function. As mentioned in the text, the step + delta-function g_2 -invariant process is expected to provide a good approximation to the effective pair correlation function of periodic crystals in asymptotically high Euclidean dimensions. Consider the linear average of g_2 over some radial length scale L :

$$G(L) = \frac{1}{L} \int_0^L g_2(r) dr. \quad (\text{A.1})$$

For the step-function process, this linear average is given by

$$G_{\text{step}}(L) = \left(1 - \frac{D}{L}\right) \Theta(L - D) \quad (\text{A.2})$$

for all dimensions. By contrast, for the step + delta-function process, we find

$$G_{\text{step}+\text{delta}}(L) = \left[1 - \left(\frac{d+1}{d+2}\right) \left(\frac{D}{L}\right)\right] \Theta(L - D), \quad (\text{A.3})$$

which converges to $G_{\text{step}}(L)$ as $d \rightarrow +\infty$. This is precisely the statement that ‘effective’ pair correlations in the step + delta-function process asymptotically vanish in high dimensions, which is the decorrelation principle as we have presented it for periodic structures.

We can also show that the cumulative coordination number $Z(R)$ (cf (4)), a particular smoothing operation on the pair correlation function, can be insensitive to decorrelation in disordered point patterns and is therefore not the appropriate quantity to examine for periodic structures. Direct calculation of $Z(R)$ for the g_2 -invariant processes above shows that the cumulative coordination numbers for both systems are identical and are given by

$$Z(R) = [\rho v(R) - 1] \Theta(R - D), \quad (\text{A.4})$$

which is equivalent to the hyperuniformity condition $Z(R) + 1 = \rho v(R)$ as $R \rightarrow +\infty$. Therefore, although $Z(R)$ can distinguish hyperuniform and non-hyperuniform point patterns, it provides little information about the behavior of asymptotic correlations in high dimensions.

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