

Robust algorithm to generate a diverse class of dense disordered and ordered sphere packings via linear programming

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We have formulated the problem of generating dense packings of nonoverlapping, nontiling nonspherical particles within an adaptive fundamental cell subject to periodic boundary conditions as an optimization problem called the adaptive-shrinking cell (ASC) formulation [S. Torquato and Y. Jiao, *Phys. Rev. E* **80**, 041104 (2009)]. Because the objective function and impenetrability constraints can be exactly linearized for sphere packings with a size distribution in d -dimensional Euclidean space \mathbb{R}^d , it is most suitable and natural to solve the corresponding ASC optimization problem using sequential-linear-programming (SLP) techniques. We implement an SLP solution to produce robustly a wide spectrum of jammed sphere packings in \mathbb{R}^d for $d=2, 3, 4, 5$, and 6 with a diversity of disorder and densities up to the respective maximal densities. A novel feature of this deterministic algorithm is that it can produce a broad range of inherent structures (locally maximally dense and mechanically stable packings), besides the usual disordered ones (such as the maximally random jammed state), with very small computational cost compared to that of the best known packing algorithms by tuning the radius of the *influence sphere*. For example, in three dimensions, we show that it can produce with high probability a variety of strictly jammed packings with a packing density anywhere in the wide range $[0.6, 0.7408\dots]$, where $\pi/\sqrt{18}=0.7408\dots$ corresponds to the density of the densest packing. We also apply the algorithm to generate various disordered packings as well as the maximally dense packings for $d=2, 4, 5$, and 6 . Our jammed sphere packings are characterized and compared to the corresponding packings generated by the well-known Lubachevsky-Stillinger (LS) molecular-dynamics packing algorithm. Compared to the LS procedure, our SLP protocol is able to ensure that the final packings are truly jammed, produces disordered jammed packings with anomalously low densities, and is appreciably more robust and computationally faster at generating maximally dense packings, especially as the space dimension increases.

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I. INTRODUCTION

Hard-particle packings have provided a rich source of outstanding theoretical problems and served as useful starting points to model the structure of liquids [1,2], glasses [3–5], crystals [7–9], colloids [2,10], granular media [11–14], living cells [6,15], random media [6,16–18], and polymeric systems [19,20]. We focus our attention in this paper on the venerable idealized hard-sphere model in d -dimensional Euclidean space \mathbb{R}^d in which the only interparticle interaction is an infinite repulsion for overlapping particles, which can be thought of as the “Ising model” for hard spheres [14].

There has been resurgent interest in hard-sphere packings in dimensions greater than three in both the physical and mathematical sciences. For example, it is known that the optimal way of sending digital signals over noisy channels corresponds to the densest sphere packing in a high-dimensional space [21,22]. 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 [23–28]. The determination of the densest packings in arbitrary dimension is a problem of longstanding interest in discrete geometry and number theory [22,29,30].

The hard-sphere Ising model enables us to be precise about the important concept of “jamming.” The generation and characterization of jammed packings are the main concerns of this paper. A jammed packing is one in which the particle positions are fixed by the impenetrability constraints and boundary conditions (e.g., hard-wall or periodic boundary conditions). For sphere packings, one can define hierarchical jamming categories beginning from the least restrictive to the most stringent one [13]. In particular, a packing is *locally* jammed if no particle in the system can be translated while fixing the positions of all other particles, requiring that each sphere in the packing be in contact with at least $d+1$ spheres not all in the same hemisphere. A *collectively* jammed packing is a locally jammed packing such that no subset of spheres can simultaneously be continuously displaced so that its members move out of contact with one another and with the remainder set. A packing is *strictly* jammed if it is collectively jammed and all globally uniform volume nonincreasing deformations of the system boundary are disallowed by the impenetrability constraints. The reader is referred to Refs. [13,14] for further details. Our main concern in this paper will be packings that are at least collectively jammed.

The packing density ϕ , the fraction of \mathbb{R}^d covered by the spheres, is the simplest characteristic of a jammed packing. However, such a characterization is clearly not sufficient in

order to distinguish between ordered and disordered packings. In fact, jammed packings may be produced with variable degrees of disorder/order. It has been suggested that a scalar order metric ψ be employed to measure the degree of order in a packing, such that $\psi=1$ corresponds to fully ordered [e.g., the perfect face-centered cubic (fcc) crystal in three dimensions] and $\psi=0$ corresponds to perfectly a disordered Poisson distribution of sphere centers [4,31]. Thus, very large jammed packings have been classified based on their locations in the density-order ϕ - ψ plane, called the *order map*. The points on the boundary of the jammed domain in the order map constitute optimal packings. For example, in \mathbb{R}^3 , the maximally dense fcc lattice sphere packing ($\phi = \pi/\sqrt{18} \approx 0.7048\dots$) [32] also has the highest order metric ($\psi=1$) when appropriately defined. Another extremal state of special interest is the maximally random jammed (MRJ) state, which is the one that minimizes a scalar order metric ψ subject to the condition of the degree of jamming [4,14]. Studies of different order metrics [33] for three-dimensional frictionless spheres have consistently led to a minimum at approximately the same density $\phi \approx 0.64$ for collective and strict jamming in the order map [4,31]. Such MRJ packings have also been shown to be hyperuniform, i.e., infinite-wavelength density fluctuations vanish [34]. The lowest density strictly jammed states in \mathbb{R}^3 , thought to be tunneled crystals [8], are a fascinating set of extremal loci in the jamming region of the order map [14]. The frequency of occurrence of a particular configuration is irrelevant insofar as the order map is concerned, i.e., the order map emphasizes a “geometric-structure” approach to analyze packings by characterizing individual configurations, regardless of their occurrence probability [14].

During the last two decades, the Lubachevsky-Stillinger (LS) algorithm [35] has been the premier workhorse to generate a wide spectrum of dense jammed sphere packings with variable disorder in both two and three dimensions [4,31,33]. This is an event-driven (or collision-driven) molecular-dynamics algorithm in which an initial configuration of spheres of a given size within a periodic box are given initial random velocities and the motion of the spheres are followed as they collide elastically and also expand uniformly until the spheres can no longer expand. This algorithm has been generalized by Donev, Torquato, and Stillinger [36] to generate jammed packings of smoothly shaped nonspherical particles, including ellipsoids [37], superdisks [38], and superballs [39].

Not surprisingly, this packing protocol is not without some inadequacies. Event-driven packing protocols with growing particles do not guarantee jamming of the final packing configuration, since jamming is not explicitly incorporated as a termination criterion. To produce jammed random packings, for example, a large expansion rate is necessary in the early stages of the simulation to suppress crystallization, but using a high expansion rate is highly undesirable toward the end of the simulation, which often leads to unjammed configurations. Thus either a variable expansion rate must be used (which decreases as a function time in some arbitrary fashion) or, if a uniform expansion is employed, the spheres of the terminal packing must be shrunk by some arbitrary small amount and then this initial packing

configuration must be redensified using a very small expansion rate. Maximally dense jammed packings are highly computationally expensive to generate, especially in high dimensions, because a very small expansion rate is necessary (on the order of 10^{-6} to 10^{-10} [40], dependent on dimension). We will see that in five and six dimensions, even an expansion rate of 10^{-10} fails to produce the maximal-density packings. Moreover, a large number of total collisions per particle is required; for MRJ packings and the densest known maximally dense packings, on the order of 10^5 and 10^7 collisions per particle are required, respectively, the latter of which is computationally very costly. Finally, such event-driven packing protocols evolve stochastic velocity initializations, which makes one have less control of the final packings via the initial configuration.

The next generation packing protocol to generate jammed sphere packings should retain the versatility and advantages of the LS algorithm while correcting its imperfections, including improving computational speed. We show that our proposed sequential-linear-programming (SLP) solution of the adaptive-shrinking cell (ASC) optimization problem formulated elsewhere for general particle shapes (including polyhedra) [43] indeed has all of these desirable features in so far as jammed sphere packings with a size distribution are concerned. Because the design variables (including periodic simulation box shape and size) and impenetrability constraints can be exactly linearized for spheres packings, the deterministic SLP solution in principle always leads to jammed packings (up to a high numerical tolerance) with a wider range of densities and degree of disorder than packings produced by the LS algorithm. Each linear-programming solution step starting from some initial particle configuration involves a deterministic collective motion of the entire particle configuration to a higher density and, because the periodic simulation box can simultaneously deform and shrink, the final state is guaranteed to be at least collectively jammed in principle. Whereas the LS algorithm requires between 10^5 and 10^7 collisions per particle without ensuring jammed final states, the deterministic SLP solution, which is easy to implement, requires only 10 to 100 linear-programming steps to achieve jammed packings, depending on the desired density, which can be controlled by tuning what we call the size of the *influence sphere*. Thus, the SLP algorithm is computationally very efficient in generating the maximally dense packings, even in high space dimensions. By appropriately choosing initial conditions, one can achieve strictly jammed disordered sphere packings with anomalously low densities, e.g., $\phi \approx 0.6$ in three dimensions [41,42]. Indeed, a novel feature of the algorithm is that it can produce a broad range of *inherent structures* (locally maximally dense and mechanically stable packings), such as the maximally random jammed states as well as the globally maximally dense inherent structures, with very small computational cost.

The rest of the paper is organized as follows. In Sec. II, we provide basic definitions for packing problems. In Sec. III we present the mathematical formulation and algorithmic details of our sequential-linear-programming procedure to solve the adaptive-shrinking-cell optimization problem to generate jammed sphere packings. In Sec. IV, we discuss the energy landscape (negative of the packing density) for

jammed sphere packings (inherent structures) and show that our SLP solution procedure is able to lead to both the mechanically stable local minima (i.e., local density maxima) and the global minima (maximal-density packings). In Sec. V, we employ the SLP algorithm to produce a diverse class of disordered jammed packings, including MRJ states, as well as maximal-density packings of hard spheres in \mathbb{R}^d for $d=2, 3, 4, 5$, and 6. The characteristics of these packings are compared to those generated using the LS algorithm. In Sec. VI, we employ the SLP algorithm to produce jammed packings in \mathbb{R}^3 with varying degrees of disorder and packing densities anywhere in the wide range $[0.6, 0.7408\dots]$. In Sec. VII, we discuss the ramification of our results and make concluding remarks.

II. DEFINITIONS

A lattice Λ in \mathbb{R}^d is a subgroup consisting of integer linear combinations of vectors $\boldsymbol{\lambda}_i$ that constitute a basis for \mathbb{R}^d . In a lattice, the space \mathbb{R}^d can be geometrically divided into identical regions F called *fundamental cells*, each of which contains one lattice site specified by the *lattice vector*

$$\mathbf{p} = n_1\boldsymbol{\lambda}_1 + n_2\boldsymbol{\lambda}_2 + \dots + n_{d-1}\boldsymbol{\lambda}_{d-1} + n_d\boldsymbol{\lambda}_d, \quad (1)$$

where $\boldsymbol{\lambda}_i$ ($i=1, 2, \dots, d$) are the *basis vectors* and n_i spans all the integers for $i=1, 2, \dots, d$. The *generator matrix* $\mathbf{M}_\Lambda = \{\boldsymbol{\lambda}_1, \boldsymbol{\lambda}_2, \dots, \boldsymbol{\lambda}_d\}$ of a lattice Λ is a matrix with the basis vectors $\boldsymbol{\lambda}_i$ as columns and, thus, contains d^2 elements. In the physical sciences and engineering, a lattice Λ is referred to as a *Bravais lattice*. Unless otherwise stated, the term ‘‘lattice’’ will refer here to a Bravais lattice only. A *lattice packing* of congruent spheres P_L is one in which the centers of the nonoverlapping identical spheres are located at the points of Λ and hence each fundamental cell contains within it exactly one sphere. Thus, the density of a lattice packing of spheres of radius R is given by

$$\phi = \frac{v_1(R)}{v_F}, \quad (2)$$

where v_F is the volume of the fundamental cell and

$$v_1(R) = \frac{\pi^{d/2} R^d}{\Gamma(1 + d/2)}, \quad (3)$$

is the volume of a d -dimensional spherical particle of radius R and $\Gamma(x)$ is the Euler gamma function.

A more general notion than a lattice packing is a periodic packing of spheres (not necessarily identical in size), which is obtained by placing a fixed configuration of N spheres (where $N \geq 1$) of radii R_1, R_2, \dots, R_N in one fundamental cell of a lattice Λ , which is then periodically replicated without overlaps. Thus, the packing is still periodic under translations by Λ , but the N spheres can occur anywhere in the chosen fundamental cell subject to the overall nonoverlap condition. The packing density of a periodic packing of spheres is given by

$$\phi = \frac{\sum_{i=1}^N v_1(R_i)}{v_F} = \rho \langle v_1(R) \rangle, \quad (4)$$

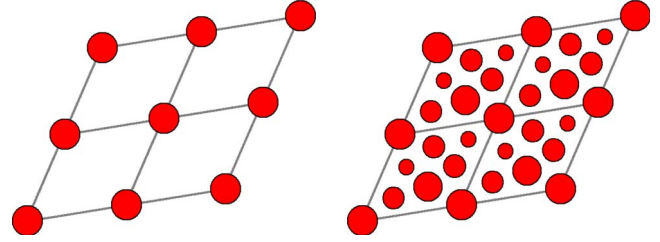


FIG. 1. (Color online) Illustrations of two basic sphere packings in \mathbb{R}^2 : (a) lattice packing of circular hard disks in \mathbb{R}^2 , which necessarily can only involve congruent circles, and (b) periodic packing of circles of different sizes.

where $\rho = N/v_F$ is the *number density*, i.e., the number of particles per unit volume, and

$$\langle v_1(R) \rangle = \frac{1}{N} \sum_{i=1}^N v_1(R_i) \quad (5)$$

expected volume of a sphere. Figure 1 illustrates in \mathbb{R}^2 a lattice packing of spheres (which by definition requires that they be identical in size) and a periodic packing of spheres in which the spheres have a size distribution.

III. MATHEMATICAL AND ALGORITHMIC DETAILS

We have formulated the problem of generating dense packings of nonoverlapping, nonspherical particles within a fundamental cell in \mathbb{R}^3 subject to periodic boundary conditions as an optimization problem called the adaptive-shrinking cell scheme [43]. The objective function is taken to be the negative of the packing density ϕ . Starting from an initial unsaturated packing configuration of particles of fixed size in the fundamental cell, the positions and orientations of the particles are design variables for the optimization. We also allow the boundary of the fundamental cell to deform macroscopically as well as compress or expand (while keeping the particles fixed in size) such that there is a net compression (increase of the density of the packing) in the final state. Therefore, the deformation and compression/expansion of the cell boundary, called the *adaptive fundamental cell*, are also design variables.

The ASC optimization problem can be solved using various techniques, depending on the shapes of the particles. For example, for polyhedron particles the nonoverlap condition is highly nonlinear, which makes it inefficient to solve even using nonlinear-programming methods, and hence Monte Carlo techniques provide an efficient means of solving the problem [43]. However, for spheres, one can exactly linearize the objective function, design variables and constraints, enabling one to exploit efficient linear-programming techniques.

We first explicitly formulate the ASC scheme for noncongruent sphere packings, i.e., spheres with a *size distribution* in \mathbb{R}^d . (Obviously, for sphere packings, particle rotations are irrelevant.) Then we describe the solution of the ASC optimization problem using sequential-linear-programming techniques.

A. Adaptive shrinking cell scheme for spheres

Consider a periodic packing of N spheres with diameters D_1, D_2, \dots, D_N in a fundamental cell F of a lattice Λ in \mathbb{R}^d . As noted in Sec. II, the fundamental cell is specified by the generator matrix $\mathbf{M}_\Lambda = \{\boldsymbol{\lambda}_1, \boldsymbol{\lambda}_2, \dots, \boldsymbol{\lambda}_d\}$, where $\boldsymbol{\lambda}_i$ are the lattice basis vectors. The design variables of the ASC scheme includes both particle displacements, and deformation and volume changes of the fundamental cell F . Let the *global* position of the center of sphere n (referred to some arbitrary set of nonorthogonal basis vectors) be denoted by \mathbf{x}_n^g . For simplicity, it is useful to choose orthogonal bases vectors or a Cartesian system. Denote by $\Delta \mathbf{x}_n^g$ a translational displacement of this particle center. Thus, the new position of the center of particle n is given by

$$\tilde{\mathbf{x}}_n^g = \mathbf{x}_n^g + \Delta \mathbf{x}_n^g. \quad (6)$$

However, since the fundamental cell is adaptive, we also need to consider the positions of the particle centers in terms of the lattice basis vectors $\boldsymbol{\lambda}_i$ ($i=1, 2, \dots, d$). For the center of particle n , this relative position \mathbf{x}_n^λ is related to the global position \mathbf{x}_n^g by the transformation [44]

$$\mathbf{x}_n^\lambda = \mathbf{M}_\Lambda^{-1} \cdot \mathbf{x}_n^g. \quad (7)$$

The adaptive fundamental cell allows for a small change in the fundamental cell

$$\Delta \mathbf{M}_\Lambda = \boldsymbol{\varepsilon} \cdot \mathbf{M}_\Lambda, \quad (8)$$

including both volume and shape changes, where $\boldsymbol{\varepsilon}$ is a symmetric strain tensor, i.e.,

$$\boldsymbol{\varepsilon} = \begin{bmatrix} \epsilon_{11} & \epsilon_{12} & \dots & \epsilon_{1d} \\ \epsilon_{21} & \epsilon_{22} & \dots & \epsilon_{2d} \\ \vdots & \vdots & \ddots & \vdots \\ \epsilon_{d1} & \epsilon_{d2} & \dots & \epsilon_{dd} \end{bmatrix}, \quad (9)$$

The new lattice is specified by the new matrix generator

$$\tilde{\mathbf{M}}_\Lambda = \mathbf{M}_\Lambda + \Delta \mathbf{M}_\Lambda. \quad (10)$$

Substituting the above equation into Eq. (7) yields

$$\tilde{\mathbf{x}}_n^g = \tilde{\mathbf{M}}_\Lambda \cdot \mathbf{x}_n^\lambda = \mathbf{x}_n^g + \Delta \mathbf{M}_\Lambda \cdot \mathbf{x}_n^\lambda. \quad (11)$$

Straining the fundamental cell corresponds to nontrivial *collective* motions of the particle centers. In general, the translational motions of the particles contain contributions from a direct part [given by Eq. (6)] and the collective motion imposed by the adaptive fundamental cell. It is this collective motion that enables the algorithm to explore the configuration space more efficiently and to produce highly dense packings.

The displacement \mathbf{r}_{mn}^λ pointing from the centroid of sphere m to that of sphere n in terms lattice basis vectors $\boldsymbol{\lambda}_i$ is given by

$$\mathbf{r}_{mn}^\lambda = \mathbf{x}_n^\lambda - \mathbf{x}_m^\lambda, \quad (12)$$

and the counterpart global displacement vector \mathbf{r}_{mn}^g is

$$\mathbf{r}_{mn}^g = \mathbf{x}_n^g - \mathbf{x}_m^g. \quad (13)$$

The Euclidean distance r_{mn}^g between the centroids is then given by

$$r_{mn}^g = |\mathbf{r}_{mn}^g| = \sqrt{\mathbf{r}_{mn}^g \cdot \mathbf{r}_{mn}^g} = \sqrt{\mathbf{r}_{mn}^\lambda \cdot \mathbf{G} \cdot \mathbf{r}_{mn}^\lambda}, \quad (14)$$

where $\mathbf{G} = \mathbf{M}_\Lambda^T \cdot \mathbf{M}_\Lambda$ is the *Gram matrix* of the lattice Λ . Thus, the general mathematical formulation of the our ASC optimization scheme for hard sphere packings in d -dimensional Euclidean space \mathbb{R}^d is

$$\text{minimize } -\phi(\mathbf{x}_1^\lambda, \dots, \mathbf{x}_N^\lambda; \mathbf{M}_\Lambda),$$

$$\text{subject to: } r_{mn}^g \geq \bar{D}_{mn}, \quad \text{for all neighbor pairs } (m, n) \text{ of interest,} \quad (15)$$

where $\bar{D}_{mn} = (D_m + D_n)/2$.

It should be emphasized that the *neighbor* pairs here do not necessarily mean nearest neighbors. Instead, they are determined by a distance γ_{mn} , i.e., two spheres m and n are neighbors of one another if their pair distance $r_{mn}^g < \gamma_{mn}$. Thus, by *near neighbors* of a given sphere we mean all of the spheres within some radius of that given sphere. Here, we choose $\gamma_{mn} = \alpha \bar{D}_{mn}$ and α is a positive in the interval $[1, L/2\bar{D}_{mn}]$, where L is the length of the shortest lattice vector associated with the fundamental cell. For monodisperse packings, γ_{mn} is identical for all particle pairs (m, n) . For polydisperse packings, γ_{mn} is generally different for each pair (m, n) . We call γ_{mn} the radius of *influence sphere* associated with each pair (m, n) . As we will see, these radii are crucial in determining the density and degree of disorder of the final jammed states.

B. Solving the ASC formulation using sequential linear programming

The formulated problem for the ASC scheme can be solved by considering an equivalent *sequential-linear-programming* problem. Suppose that we only allow the fundamental cell to change by a small amount from its original size and shape, and only allow small particle displacements; then both the impenetrability constraints and the objective function can be linearized. Due to the linearization, one needs now to explicitly places bounds on the strain components and the particle displacements, which do not appear in the original problem. Once the linearized problem is solved via the linear-programming method, a new many-particle configuration can be generated using the resulting particle displacements and fundamental cell. Then this new configuration is used as an initial configuration, based on which a new LP problem is formulated and solved. This sequential-linear-programming procedure is repeated until the increase of the packing density is smaller than some prescribed small tolerance value, implying the system is jammed up to high numerical accuracy.

Consider an initial packing configuration of N spheres within a fundamental cell of volume v_F^0 . According to relation (4), the density of the packing is given by

$$\phi_0 = \frac{\sum_{i=1}^N v_1(R_i)}{v_F^0}, \quad (16)$$

where $v_1(R_i)$ is the volume of sphere i with radius R_i . Now consider a small change of the fundamental cell that leads to a cell volume change Δv , as well as a change of the positions of the centers of every sphere that obey the impenetrability constraint. The density of the new configuration in the new fundamental cell is given by

$$\phi = \frac{\sum_{i=1}^N v_1(R_i)}{v_F^0 + \Delta v}. \quad (17)$$

If Δv is sufficiently small, we can expand ϕ as a Taylor series in Δv , keeping only the linear terms, i.e.,

$$\phi = \frac{\sum_{i=1}^N v_1(R_i)}{v_F^0 + \Delta v} \approx \frac{\sum_{i=1}^N v_1(R_i)}{v_F^0} \left[1 - \frac{\Delta v}{v_F^0} \right] = \phi_0 [1 - \text{tr}(\boldsymbol{\varepsilon})], \quad (18)$$

where we have used the relation $\Delta v/v_F^0 = \text{tr}(\boldsymbol{\varepsilon})$, i.e., the relative small volume change is given by the trace of the strain tensor. We see from the above equation that maximizing the packing density ϕ is equivalent to minimizing the *trace of the strain tensor* $\text{tr}(\boldsymbol{\varepsilon})$.

The nonoverlapping conditions can be linearized in a similar way, which leads to the following sequential-linear-programming formulation for the ASC procedure:

$$\text{minimize } \text{tr}(\boldsymbol{\varepsilon}) = \varepsilon_{11} + \varepsilon_{22} + \dots + \varepsilon_{dd}$$

subject to

$$\begin{aligned} & \mathbf{M}_\Lambda \cdot \mathbf{r}_{nm}^\lambda \cdot \boldsymbol{\varepsilon} \cdot \mathbf{M}_\Lambda \cdot \mathbf{r}_{nm}^\lambda + \Delta \mathbf{x}_m^\lambda \cdot \mathbf{G} \cdot \mathbf{r}_{nm}^\lambda + \Delta \mathbf{x}_n^\lambda \cdot \mathbf{G} \cdot \mathbf{r}_{nm}^\lambda \\ & \geq \frac{1}{2} (\bar{D}_{mn}^2 - \mathbf{r}_{nm}^\lambda \cdot \mathbf{G} \cdot \mathbf{r}_{nm}^\lambda) + \mathcal{R}, \end{aligned} \quad (19)$$

for all neighbor pairs (m, n) of interest

$$\Delta \mathbf{x}_n^{\lambda, \text{lower}} \leq \Delta \mathbf{x}_n^\lambda \leq \Delta \mathbf{x}_n^{\lambda, \text{upper}}, \quad \text{for all } n = (1, \dots, N),$$

$$\boldsymbol{\varepsilon}^{\text{lower}} \leq \boldsymbol{\varepsilon} \leq \boldsymbol{\varepsilon}^{\text{upper}}.$$

Note that the tensor/vector inequalities in Eq. (19) apply to the corresponding components and $\mathbf{r}_{nm}^\lambda = -\mathbf{r}_{mn}^\lambda = \mathbf{x}_m^\lambda - \mathbf{x}_n^\lambda$ is the displacement vector between spheres m and n in the initial configuration. The scalar quantity $\mathcal{R} > 0$ is a relaxation variable, which accounts for the effects of higher-order terms that would be ignored in the purely linearized problem. The value of \mathcal{R} is determined by the bounds on the design variables, i.e.,

$$\mathcal{R} = \max_{(m,n)} \{-\Delta \mathbf{M}_\Lambda \cdot \Delta \mathbf{x}_{mn}^\lambda\}. \quad (20)$$

Introducing \mathcal{R} has several advantages. First, it makes the linearized problem rigorously equivalent to the original (un-

linearized) optimization problem. Second, it enables us to set practically large bounds on the design variables (e.g., the strain components and the particle displacements), which in turn enables the algorithm to explore a larger region of the configuration space around the initial point and makes it more efficient to generate dense packings. In practice, however, we find that as long as the bounds on the design variables are chosen to be sufficiently small, one can safely set $\mathcal{R}=0$ and check the generated packing to ensure that no impenetrability constraints are violated. If some of them are violated due to the deformable fundamental cell, i.e., leading to a non-positive-definite quadratic term associated with the strain tensor in these conditions, the bound widths are reduced to one half of their original value and the linearized problem is re-solved.

Since the objective function and constraints in the new problem are linear functions of the design variables, the new problem [Eq. (19)] is solved using standard linear-programming methods (e.g., simplex method, interior-point method, etc.). Starting from an initial packing configuration, we solve the linearized problem and find a new packing, which is denser and slightly different from the starting configuration. Then the new packing is used as the starting configuration, and a new linearized problem is solved. By repeating this process, one actually solves a sequence of linear programming problems to generate a denser packing from the previous configuration.

The choice of the bounds for the design variables is also important in practice. For example, different bound widths for the principle strain components and shear strain components correspond to different compression and deformation rates for the packing. Choosing the bound widths carefully can dramatically improve the ability of the algorithm to generate dense packings. We also emphasize that the choice of neighbor pairs (i.e., the value of the influence sphere radius γ_{mn}) is also nontrivial, which we will elucidate in the ensuing discussion.

C. Ensuring jamming

Finally, we note that our SLP solution procedure guarantees (up to numerical precision) that the final locally or globally maximally dense states are indeed strictly jammed under periodic boundary conditions. In general, each impenetrability (nonoverlapping) condition defines a curved hypersurface in the configuration space that separates accessible and inaccessible regions [14,45]. Linearization of the impenetrability conditions corresponds to replacing the curved hypersurfaces with hyperplanes, which further reduces the accessible region (i.e., the linearized conditions are even stronger than the original conditions). However, near the jamming point, the available configuration space for the spheres asymptotically approaches a closed convex polytope which is exactly determined from the linearized impenetrability conditions [14,45]. In other words, toward the jamming point, the linearized conditions become exact nonoverlapping conditions asymptotically. Therefore, the SLP algorithm guarantees jamming of the final packing configurations.

In addition, Our SLP algorithm near the final steps of the process is intimately related to a linear-programming (LP)

protocol to test for jamming for hard-sphere packings that we previously developed [46]. In the LP jamming-test protocol, the interparticle gaps are maximized subject to the nonoverlapping conditions. If large gaps can be opened, the packing under consideration is not jammed; otherwise it is jammed. For our SLP algorithm, the packing density is driven to a local or global maximum, which is not only collectively jammed (since as mentioned earlier, the descent to maxima necessarily involve collective configurational motions) but strictly jammed, since the fundamental box is deforming and shrinking (on average). In general, if a packing is not jammed, there exist collective particle motions and boundary deformations that can lead to a higher density. All of the sphere packings in \mathbb{R}^2 and \mathbb{R}^3 produced by our SLP algorithm are tested using the LP jamming-test protocol [46]. The packings with a deformable/shrinking fundamental cell were found to be strictly jammed and those with an isotropically shrinking fundamental cell were found to be collectively jammed, which verifies the robustness of our algorithm in producing jammed packings.

IV. ENERGY LANDSCAPE, INHERENT STRUCTURES (LOCAL MINIMA) AND GLOBAL MINIMA

A. Energy landscape picture

From a statistical-mechanical point of view, the solution of the ASC formulation using a SLP amounts to searching nearby local minima of the *energy landscape*. The energy landscape, defined by the energy (objective function), equal to the negative of the trace of the strain tensor associated with the fundamental cell F of the packing or, equivalently, the negative of the density ϕ , is a surface embedded in a $(dN+1)$ -dimensional space, where d is the number of degrees of freedom of a single particle and N is the number of particles in F . A given initial configuration has an energy that corresponds to a point on the landscape. Starting from this point in this multidimensional space, our SLP algorithm finds a nearby point, which can be reached from the initial point by a single linear multidimensional displacement with a lower energy (higher density) subjected to the limits on the size and directions of the displacement as well as the impenetrability conditions. This procedure is repeated until a jammed state with a high density is reached, which can either be a local energy minimum (local density maximum) or a global energy minimum (global density maximum) depending on the bound widths on the design variables, as well as the influence sphere radii γ_{mn} for near-neighbor determination.

As indicated in Sec. III A, for monodisperse sphere packings, the radius γ_{mn} of the influence sphere for pair (m,n) is identical for each pair of particles. We call this the *influence sphere* for each particle (see Fig. 2) because only the particles whose centers are within the influence sphere will affect the central particle as far as the nonoverlapping conditions are concerned. For polydisperse packings, γ_{mn} is generally different for each particle pair. However, loosely speaking, one can still imagine an effective influence sphere around each particle, which determines the neighbors that affect the central particle. If the influence sphere (i.e., γ_{mn}) is

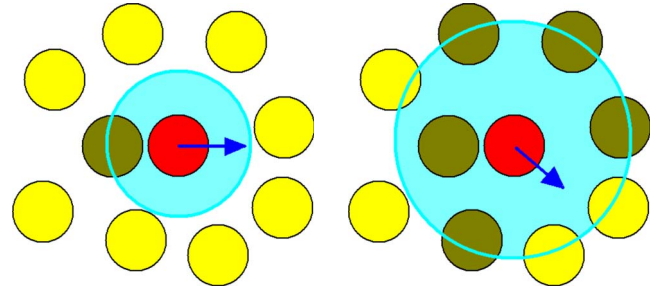


FIG. 2. (Color online) Influence spheres (i.e., effective interaction range for the central particle in so far as the nonoverlapping condition is concerned) and the directions of motion of the particle. (a) Only the nearest neighbor of the central (red or darkest gray) particle is included in the influence sphere. The central sphere will move away from its neighbor along a line connecting their centers, which allows a maximal shrinkage of the fundamental cell. Such a shrinkage would lead to a local maximum in the packing density. (b) More near neighbors are included in the influence sphere. The central particle moves along a direction that maximizes its distance from *all* of the neighbors within the influence sphere. In other words, the movement of the central particle is coupled with all of the near neighbors within the influence sphere and allows a maximal shrinkage of the fundamental cell, which after a sufficient number of iterations leads to a global maximum of the packing density.

sufficiently small such that only pairs with minimal separation distance are considered, particle motions are controlled by the local environment [see Fig. 2(a)], i.e., they move in opposite directions along the line connecting their centroids. If only particle pairs (or triplets, etc.) with the minimal separation distance are considered at each stage of the SLP procedure, the system rigorously follows the steepest-descent trajectory and evolves to the associated inherent structure. As the influence sphere (γ_{mn}) becomes larger, successively larger numbers of spheres must respect the nonoverlap condition, all of which affect the motion of the central particle [see Fig. 2(b)].

Consequently, the influence sphere radius γ_{mn} can be considered to be an effective interaction range. For each near-neighbor pair, a nonoverlapping condition for that pair is included in the SLP formulation, which in general will lead to a different jammed-packing solution from one in which that pair of particles are not considered neighbors of one another. Therefore, the neighbor pairs effectively interact with each other through the nonoverlapping condition. A larger γ_{mn} corresponds to a longer interaction range, which enables more directions in the energy landscape to be explored beyond the one that leads to a local density increase (i.e., a steepest descent in the energy landscape). We will see in the ensuing discussion that this feature of the SLP procedure enables it to find both local and global energy minima (density maxima). We note that the number of neighbor pairs n_{pair} is a monotonically increasing function of γ_{mn} , namely, $n_{pair} \sim \gamma_{mn}^d$ for any jammed packings in \mathbb{R}^d . Although a large value of γ_{mn} is necessary to produce maximally dense packings, one generally only needs an associated small number of particles in the fundamental cell. Therefore, the total computational cost when a large value of γ_{mn} is employed to generate maximally dense packings is usually much smaller than

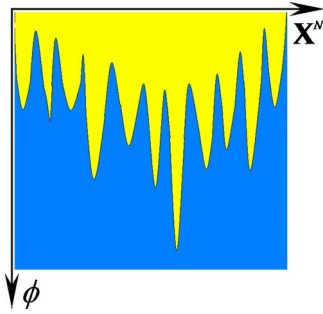


FIG. 3. (Color online) A schematic illustrating the idea of inherent structures for a system composed of N hard spheres as adapted from Ref. [49], though the term “inherent structure” was not explicitly used there. The horizontal axis labeled \mathbf{X}^N stands for the entire set of centroid positions, and the density ϕ increases downward. The jagged curve is the boundary between accessible (upper) configurations (shown in yellow or light gray in the print version) and inaccessible (lower) configurations (shown in blue or dark gray in the print version). The deepest point of the accessible configurations corresponds to the maximal-density packings of hard spheres.

that associated with a small value of γ_{mn} to produce disordered packings with a large number of particles per fundamental cell. Empirically, we find that when $\gamma_{mn} > 4\bar{D}_{mn}$, the SLP algorithm generally leads to the maximally dense packings, where $\bar{D}_{mn} = (D_m + D_n)/2$ is the average diameter of two spheres.

Moreover, one can view this SLP compression process physically as a compression of a hard-sphere system in a *superviscous liquid* subjected to periodic boundary conditions in the absence of a gravitational field. Although the immersion of the hard spheres in the superviscous liquid precludes any collisions between the particles, there is a dynamical interaction with the system boundary. Thus, the only “dynamical” parameters of this physical compression are the deformation parameters of the fundamental cell. For this SLP “compression” process, the system moves efficiently by collective particle motions to nearby denser configurations.

B. Inherent structures—mechanically stable local minima

When the influence sphere radius γ_{mn} is sufficiently small (e.g., $\gamma_{mn} \sim 1.5\bar{D}_{mn}$) and the bound widths are taken to be sufficiently large (e.g., $|\epsilon_{ij}| \sim 0.1$ and $|\Delta\mathbf{x}^\lambda| \sim 0.5\bar{D}$, where \bar{D} is the averaged diameter of all the spheres in the packing), the SLP solution of the ASC scheme from some initial configurations leads to a mechanically stable local energy minimum (local density maximum), which in principle is the *inherent structure* associated with the starting initial many-particle configuration [47,48]; see Fig. 3. All initial configurations that compress to the same jammed structure, excluding distinctions between states that differ by the interchange of identical particles, belong to the same inherent structure. In two and three dimensions, the number of distinct inherent structures scales with the number of particles N in the fundamental cell, i.e., $\exp(\alpha N)$, where α is dimensional-dependent constant of order unity [47].

Stillinger, DiMarzio, and Kornegay were the first to introduce the idea of inherent structures for two-dimensional hard circular disks [49]. However, at that time they did not explicitly use the terminology “inherent structure.” Nonetheless, they indeed proposed a conceptual procedure to produce inherent structures for hard-sphere systems and applied it to hard disks in two dimensions. They considered a dilute configuration of N spheres centered at $\mathbf{x}_1, \dots, \mathbf{x}_N$, and subsequently expanded the spheres. During such an expansion, there will occur a first contact between the pair of spheres with the minimal original pair separation distance. As the spheres are further expanded, one should move this pair apart (each member at an equal rate along the line joining the centers) just to maintain the contact. Other pairs in the system similarly will come into contact, and will be rearranged by the same prescription. As the process proceeds, triplets, and larger sets of spheres will touch, and subsequently should be moved by a generalization of the pair procedure. These authors proposed that this procedure can be mapped into a steepest-descent problem. Namely, if one has a set of n spheres in contact at $\mathbf{x}_1, \dots, \mathbf{x}_n$, the displacements $d\mathbf{x}_1, \dots, d\mathbf{x}_n$, which maintain contacts under the diameter increase ΔD , should be selected to minimize the positive definite form

$$\sum_{i=1}^n \left(\frac{\Delta \mathbf{x}_i}{\Delta D} \right)^2 \quad (21)$$

subject to those constraints. In particular, these authors commented that “for very large N , the chance of selecting (at random) a set of initial positions that would lead to jamming in the regular close-packed array becomes very small.” In modern language, this statement means that random initial configurations for the case of identical spheres would lead to maximally random jammed states for $d \geq 3$, which are isostatic configurations [50], with very high probability. However, for two-dimensional monodisperse circular disk packings, it is well known that random initial configurations lead to highly ordered crystalline packings with very high probability [14]. If an entropic measure of disorder was employed, such two-dimensional crystalline packings would be designated to be the most disordered packings or MRJ states, which is clearly incorrect. This is one of the many reasons that an entropic (i.e., occurrence frequency of the same configurations) measure of disorder can be misleading [14].

It is not difficult to see that our SLP algorithm when provided with the appropriate parameters closely follows the aforementioned conceptual steepest-descent procedure for obtaining inherent structures. Specifically, if the nonoverlapping conditions are only specified for nearest-neighbor pairs, triplets etc., i.e., a small influence sphere radius γ_{mn} is used (see Fig. 2), the SLP algorithm will only move apart those spheres that are explicitly considered in the nonoverlapping conditions such that the largest possible compression of the system (consistent with the nonoverlapping conditions and the bounds) can be performed (through applying the solved strain tensor). In other words, starting from an initial configuration, the SLP algorithm can lead to a maximal-density increase subject to the constraints imposed by closest neigh-

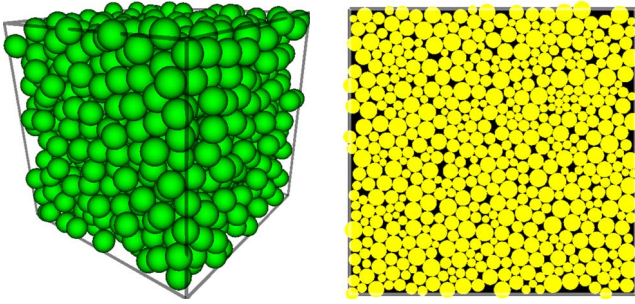


FIG. 4. (Color online) Maximally random jammed packings in low dimensions via SLP algorithm. (a) A MRJ packing of 1000 monodisperse spheres in three dimensions. (b) A MRJ packing of 500 polydisperse circular disks in two dimensions.

bors, following the trajectory of steepest descent in the energy landscape. This is an equivalent reformulation of the Stillinger-DiMarzio-Kornegay steepest-descent procedure. Importantly, the SLP algorithm is not limited to steepest-descent mappings. We will see in the next section that by using relatively large influence spheres, one can access unusual inherent structures from random initial configurations.

We note that Zinchenko [51] proposed and implemented a numerical algorithm conceptually equivalent to the Stillinger-DiMarzio-Kornegay steepest-descent procedure to produce disordered jammed spheres packings in three dimensions. However, the Zinchenko algorithm involves the numerical solution of many different differential equations (dN of them) as well as the solutions to dN algebraic equations that prescribe the densification process that allows the particles to swell while retaining sphere contacts to the extent possible. This makes the algorithm both computationally expensive and incapable of generating ordered packings. Furthermore, O’Hern *et al.* employed a conjugate-gradient (CG) method to produce jammed packings of soft spheres interacting with short-ranged repulsive power-law pair potentials [52]. Starting from a dilute initial configuration, the spheres are allowed to grow, which causes an increase of the total energy of the system. Then CG method is used to relax the system to zero energy. The procedure is repeated until the total energy cannot be relaxed to zero, and the system is considered jammed. Although not explicitly indicated, this algorithm in general produces inherent structures of such soft-sphere systems.

C. Energy minima—density maxima

Though our SLP algorithm can produce inherent structures, it is crucial to emphasize that our algorithm is much more general than the aforementioned conceptual steepest-descent procedure [49], the Zinchenko protocol [51] and the conjugate-gradient method [52]. In particular, including more neighbors associated with any given sphere in the set of nonoverlapping conditions, i.e., increasing the radius of the influence sphere γ_{mn} , effectively introduces “long-range interactions” (i.e., $\gamma_{mn} \sim 3.5\bar{D}_{mn}$). Together with smaller bound widths (e.g., $|\epsilon_{ij}| \sim 0.01$ and $|\Delta \mathbf{x}^\lambda| \sim 0.05\bar{D}$, where \bar{D} is the effective diameter of all of the spheres in the packing),

the SLP solution of the ASC scheme from random initial configurations can lead to unusual inherent structures, such as a mechanically stable global energy minimum (global density maximum).

Recall that when the radius influence sphere (γ_{mn}) becomes larger, successively larger numbers of particles must respect the nonoverlap condition, all of which affect the motion of the central particle. A sufficiently large γ_{mn} implies that the particles experience effectively long-range “interactions” via the influence spheres, which enable more directions in the energy landscape to be explored besides the steepest-descent direction. In other words, multiple minima can now be explored; and instead of moving to a local energy minimum (local density maximum) by a steepest-descent trajectory, the system is able to evolve toward the deepest minimum available until it is in the basin of the global minimum.

D. Unusual inherent structures—low-density jammed packings

From its definition, it is clear that an inherent structure is highly dependent on its associated initial configuration. Therefore, an innovative procedure to generate unusual initial configurations in principle enables one to obtain unusual inherent structures. Moreover, there is no reason to limit oneself to random initial configurations alone. For example, in Ref. [41] diluted MRJ packings are employed as initial configurations to produce low-density jammed packings in \mathbb{R}^3 .

V. JAMMED DISORDERED AND ORDERED PACKINGS

In this section, we employ our SLP algorithm to produce maximally random jammed (MRJ) packings as well as maximally dense packings of hard spheres in d -dimensional Euclidean space \mathbb{R}^d for $d=2, 3, 4, 5$, and 6 . In \mathbb{R}^1 , there is only a single jammed state, namely, the integer-lattice packing with unit density. Therefore, we will not consider this trivial case here. The characteristics of the packings produced by the SLP algorithm are compared to those obtained using the LS algorithm, which verifies the robustness and low-computational cost of our SLP algorithm. Moreover, we show that the SLP algorithm is superior to the LS algorithm in several aspects, especially in producing maximal dense packings.

A. Maximally random jammed packings

Random dilute packings with a density $\phi=0.05$ are used as initial configurations to produce MRJ packings for $d=2, 3, 4, 5$, and 6 using the SLP algorithm. In two dimensions, polydisperse disks with diameters uniformly distributed within $[D, 2D]$ are used to generate MRJ packings because monodisperse circular disks have an inevitable propensity to form highly ordered packings in the jamming limit, as discussed in Sec. IV B. In higher dimensions, monodisperse spheres are used. As indicated in the previous section, to produce MRJ packings (which are the maximally disordered inherent structures), a small influence sphere radius γ_{mn} is used such that evolution of the packing in the energy land-

TABLE I. Characteristics of MRJ packings produced using the SLP and LS algorithms. ϕ is the density of final jammed packing and f_r is the fraction of the rattlers. t_s is the total simulation time. For LS algorithm, t_s includes the time of simulations with both the initial large expansion rate and the fine-tuning expansion rate.

	LS algorithm	SLP algorithm
$d=3$	$\phi=0.642 \pm 0.005$ $f_r=0.030 \pm 0.003$ $t_s=1.5$ h	$\phi=0.640 \pm 0.004$ $f_r=0.028 \pm 0.003$ $t_s=10$ min
$d=4$	$\phi=0.460 \pm 0.006$ $f_r=0.012 \pm 0.002$ $t_s=4.8$ h	$\phi=0.465 \pm 0.005$ $f_r=0.014 \pm 0.002$ $t_s=46$ min
$d=5$	$\phi=0.315 \pm 0.005$ $f_r=0.008 \pm 0.002$ $t_s=14$ h	$\phi=0.310 \pm 0.005$ $f_r=0.006 \pm 0.002$ $t_s=3.2$ h
$d=6$	$\phi=0.201 \pm 0.005$ $f_r=0.005 \pm 0.001$ $t_s=193.5$ h	$\phi=0.199 \pm 0.005$ $f_r=0.005 \pm 0.001$ $t_s=8.3$ h

scape follows a path of steepest descent (e.g., $\gamma_{mn} \sim 1.5\bar{D}_{mn}$, $|\epsilon_{ij}| \sim 0.1$, and $|\Delta\mathbf{x}^\lambda| \sim 0.5\bar{D}$). The number of particles in the fundamental cell are $N=500, 2000, 4000, 8000, 12\,000$, respectively, for $d=2, 3, 4, 5, 6$. The packing is considered jammed and the simulation is terminated if the increase of the packing density is less than 10^{-8} . Figure 4 shows typical packing configurations in two and three dimensions.

The characteristics of the monodisperse packings are compared to those obtained using the LS algorithm [25], which has been verified to be a robust protocol to produce MRJ packings. A two-step procedure is used for the LS algorithm to generate jammed disordered packings: a large initial expansion rate of ~ 0.01 and a small fine-tuning expansion rate of $10^{-(3+d)}$ (where d is the spatial dimension). A comparison of the packing density and fraction of rattlers (i.e., movable particles caged by their jamming neighbors) is given in Table I. Each density ϕ and the fraction of rattlers f_r are obtained by averaging over five configurations. We note that the MRJ densities obtained via the LS and SLP algorithms are very close to each other. We expect that in the infinite-system limit, the MRJ densities generated via the two algorithms should be essentially identical to one another. Figure 5 shows the pair correlation functions g_2 of the packings obtained using both the SLP and LS algorithm [55]. It can be seen clearly that the MRJ packings produced by our SLP algorithm are consistent with those produced by the LS protocol across dimensions. Specifically, in high dimensions, the split-second peak in g_2 , present for $d=3$, gets dramatically diminished and oscillations in g_2 get significantly dampened. These findings are consistent with a recently proposed “decorrelation principle” [27] that states that unconstrained spatial correlations in disordered packings should vanish asymptotically in the high-dimensional limit. The two-dimensional polydisperse packing possesses a density $\phi=0.846$ and approximately 3% of rattlers.

We note that in producing MRJ packings using the LS algorithm, a large expansion rate is necessary in the beginning of the simulation to drive the system out of equilibrium but it is undesirable toward the end, which may lead to unjammed configurations. Therefore, a variable expansion rate is needed and must be very small at the end of the simulation to ensure jamming, as we have done here. In our SLP algorithm, the control parameters are single-valued. Moreover,

the SLP algorithm naturally leads to jammed packings, as we discussed in Sec. III B, which makes it superior to the LS algorithm in producing jammed packings.

B. Maximal-density packings

In order to further test the SLP algorithm, we apply to see whether it can recover the densest packings of congruent spheres in two and three dimensions and densest known congruent sphere packings in \mathbb{R}^4 , \mathbb{R}^5 , and \mathbb{R}^6 . Recall that we do not consider the case in \mathbb{R}^1 , because that corresponds to the trivial solution of the integer-lattice packing. In \mathbb{R}^2 and \mathbb{R}^3 , the densest packings of congruent spheres have been proved to be the triangular lattice [53] and fcc lattice [32], respectively. One of the generalizations of the three-dimensional fcc lattice to higher dimensions is the d -dimensional checkerboard lattice D_d , defined by taking a hypercubic lattice and placing spheres on every site at which the sum of the lattice indices is even, i.e., every other site [22]. The four- and five-dimensional checkerboard lattices D_4 and D_5 with densities $\phi=\pi/16=0.6168\dots$ and $\phi=\sqrt{2}\pi^2/30=0.4652\dots$, respectively, are believed to be the densest sphere packings in those dimensions. For $d=6$, the densest sphere packing is conjectured to be the “root” lattice E_6 with density $\phi=\sqrt{3}\pi^3/144=0.3729\dots$ [22].

It is important to note that in order to generate perfect crystalline jammed packings using a finite simulation cell with periodic boundary conditions, the number of particles in the fundamental cell must be consistent with its shape. For example, if a fixed cubic fundamental cell or rhombohedral fundamental cell (associated with the fcc lattice) in three dimensions is used, this “magic” number of spheres should be $N=4m^3$ and $N=m^3$ ($m=1, 2, 3, \dots$), respectively. In most cases, the fundamental cell associated with the densest packing is not known *a priori* and hence one needs to explore a variety of different particle numbers (by carrying out many simulations) to obtain the maximal-density packing with typical protocols that use a fixed cell. Therefore, it is highly desirable to explore systematically the shape of the appropriate fundamental cell by employing a single simulation with variable particle numbers, which our SLP algorithm enables one to do. In particular, because our SLP algorithm does not simulate the real many-particle dynamics, a very small value of N can be used, which can significantly reduce the simula-

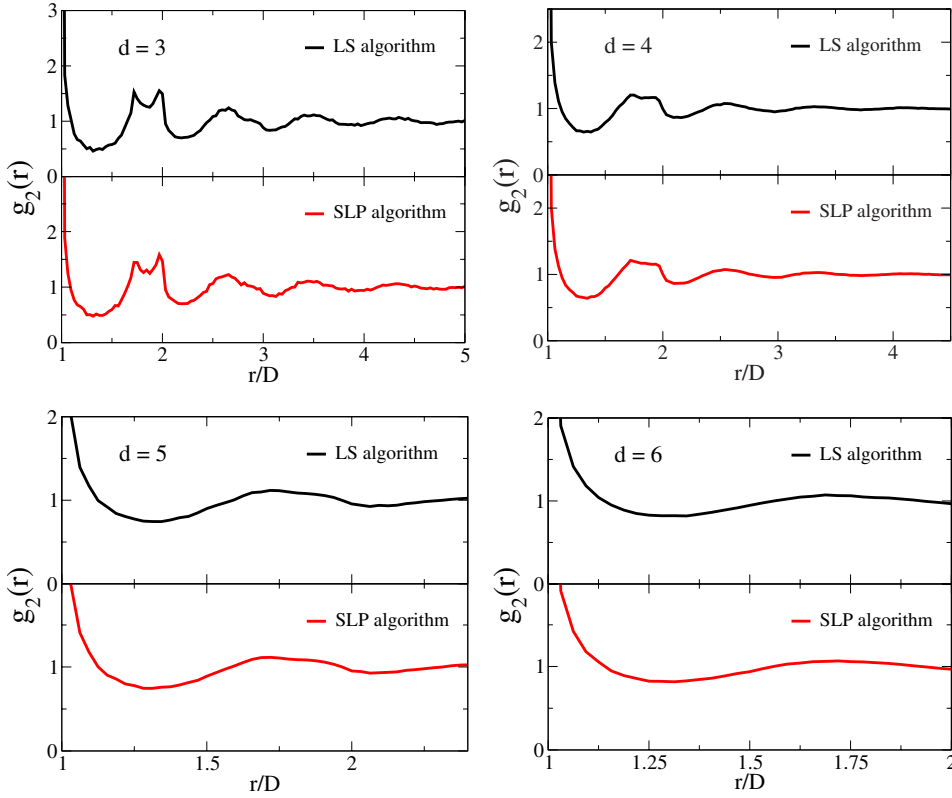


FIG. 5. (Color online) Pair correlation function g_2 of maximally random jammed packings of monodisperse hyperspheres in selected dimensions via the SLP and LS algorithm.

tion time. For example, one can choose N to be as small as one, which reduces the SLP algorithm to a lattice-optimization algorithm. For the LS protocol, using a very small value of N will introduce systematic errors due to lack of collisions between the particles within the fundamental cell or because collisions are not properly treated.

To produce maximal-density packings in \mathbb{R}^d , initial packing configurations with $N=3^d$ spheres in (hyper)cubic fundamental cells possessing a density lower than the “equilibrium” hard-sphere freezing density (Ref. [26]) for each dimension (i.e., $\phi=0.45, 0.4, 0.3, 0.15, 0.06$ for $d=2, 3, 4, 5, 6$, respectively) are used for both the LS and SLP algorithm. We note that for the current implementation of the LS algorithm, a deformable fundamental cell program is available for $d=2$ and 3 [36]; for $d \geq 4$, only a fixed cubic fundamental cell is available [26]. The sphere expansion rate used to generate a maximally dense packing in \mathbb{R}^d is $10^{-(3+d)}$. For the SLP algorithm, a deformable fundamental cell is used in all dimensions. The influence sphere radius is chosen to be $\gamma_{mn} \sim 3.5D$, and $|\epsilon_{ij}| \sim 0.01$ and $|\Delta \mathbf{x}^\lambda| \sim 0.05D$ are used. The final densities produced by the two algorithms and the corresponding computational times to produce the jammed states are compared in Table II. It can be seen that the SLP algorithm successfully reproduces the densest known packings in all dimensions; while our implementation of the LS algorithm only was able to reproduce the maximal-density packings in \mathbb{R}^2 and \mathbb{R}^3 . Although the LS algorithm does not generate perfect crystals for $d=4, 5$, and 6, the high densities, it produces suggest that these packings contain very large crystallized regions. We have also used $N=128$ in \mathbb{R}^4 and $N=512$ in \mathbb{R}^5 for the LS algorithm, which are the magic numbers for cubic fundamental cells in four and five

dimensions, respectively, as well as an expansion rate of 10^{-10} . In \mathbb{R}^4 , the packing converges to the D_4 -lattice packing with $\phi=0.6168\dots$; while in \mathbb{R}^5 , the final packing is partially crystallized with $\phi=0.4392\dots$

These results clearly demonstrate the advantage of our SLP algorithm in producing maximally dense sphere packings, i.e., the SLP algorithm is both much more computationally efficient and more robust in finding the densest known sphere packings than the LS algorithm. In particular, the adaptive fundamental cell is crucial for obtaining perfect maximal-density packing structures. In addition, we have tried a variety of particle numbers in the fundamental cell (e.g., $1 \leq N \leq 2^d$ for \mathbb{R}^d) in four, five, and six dimensions and have not obtained any packings possessing a larger density than the densest known packings. This provides numerical support that the densest known packing in these dimensions may indeed be the conjectured maximal-density packings.

We note that very recently Kallus, Elser, and Gravel [54] applied a periodic “divide” and “concur” (PDC) algorithm to generate maximally dense packings of hard particles. In this variant of the PDC algorithm, they follow Ref. [43] by allowing the fundamental cell to deform and change shape during the simulation. Interparticle impenetrability constraints are relaxed in the beginning of the simulation such that overlapping intermediate configurations are allowed. Toward the end of the simulation, all overlaps are systematically removed. Among other applications, these authors applied the PDC algorithm to successfully reproduce the densest known lattice sphere packings (not necessarily the densest packings) and the best known lattice kissing arrangements in up to 14 and 11 dimensions, respectively, since they were limited to using only a small number of particles per

TABLE II. Characteristics of densest jammed packings produced using the SLP and LS algorithms. Here ϕ is the density of final jammed packing, N is the number of particles in the fundamental cell, and t_s is the total simulation time.

	LS algorithm	SLP algorithm
$d=2$	$N=9$ $\phi=0.9068\dots$ $t_s=0.1$ min	$N=9$ $\phi=0.9068\dots$ $t_s=0.1$ min
$d=3$	$N=27$ $\phi=0.7408\dots$ $t_s=6.5$ min	$N=27$ $\phi=0.7408\dots$ $t_s=1.5$ min
$d=4$	$N=81$ $\phi=0.5608\dots$ $t_s=1.6$ h	$N=81$ $\phi=0.6168\dots$ $t_s=4.5$ min
$d=5$	$N=243$ $\phi=0.4154\dots$ $t_s=47.5$ h	$N=243$ $\phi=0.4652\dots$ $t_s=11$ min
$d=6$	$N=729$ $\phi=0.3287\dots$ $t_s=283.5$ h	$N=729$ $\phi=0.3729\dots$ $t_s=27$ min

fundamental cell. Although the PDC algorithm is efficient in producing dense lattice sphere packings, it does not appear to be capable of generating *jammed* sphere packings with a diversity of disorder and density that can be produced via either the SLP or LS algorithm.

VI. ORDER MAP

Besides the capability of generating MRJ packings and maximally dense jammed packings efficiently and robustly, the SLP algorithm has the capacity to produce jammed sphere packings of with a wide range of densities and degrees of disorder/order, i.e., a diverse set of inherent structures. Here we illustrate this capability by focusing on identical spheres in three dimensions. By tuning the bound widths and the influence sphere radius γ_{mn} , the density at which the packing is jammed and its degree of disorder can be controlled to a great degree. In particular, the ranges of the parameter values used are as follows: $\gamma_{mn} \in (1.5D_{mn}, 3.5D_{mn})$, $|\epsilon_{ij}| \in (0.01, 0.1)$, and $|\Delta \mathbf{x}^\lambda| \in (0.05D, 0.5D)$ (where D is the diameter of the spheres in the packing). In general, a larger value of γ_{mn} and smaller bound widths allow for a larger number of particles to be effectively coupled to one another, which in turn leads to a higher jammed-packing density. By continuously varying the values of the control parameters, a spectrum of packing density $\phi \in [0.64, 0.74048\dots]$ can be obtained when random dilute packings are used as initial configurations. This density range is also achievable by the LS algorithm, but we shall see that disordered jammed packings with densities significantly lower than 0.64 can be generated using the SLP algorithm.

Typical configurations of such packings with $\phi \in [0.64, 0.74048\dots]$ and $N \approx 1000$ are mapped onto a density-order-metric diagram (black circles). The diagram or *order map* emphasizes a “geometric-structure” approach to analyze packings by characterizing individual configurations, regardless of their occurrence probability [14]. Here the translational order metric \mathfrak{T} [4] is used to quantify the order

of the packings (other order metrics yield equivalent results [4,31]). The translational order metric \mathfrak{T} [4] is defined as

$$\mathfrak{T} = \left| \frac{\sum_i^{N_s} (n_i - n_i^{ideal})}{\sum_i^{N_s} (n_i^{FCC} - n_i^{ideal})} \right|, \quad (22)$$

where n_i is the average occupation number for the shell i centered at a distance from a reference sphere that equals the i th nearest-neighbor separation for the open FCC lattice at that density and N_s is the total number of shells for the summation; n_i^{ideal} and n_i^{FCC} are the corresponding shell occupation numbers for an ideal gas (spatially uncorrelated spheres) and the open FCC lattice. For a completely disordered system (e.g., a Poisson distribution of points) $\mathfrak{T}=0$, whereas $\mathfrak{T}=1$ for the FCC lattice.

Moreover, we have produced jammed disordered packings with densities as low as 0.595, which is an anomalously low value, using the SLP algorithm from special initial configurations. Specifically, a small fraction ($f_s=0.1-2.5\%$) of particles are removed from MRJ packings which are obtained using the SLP algorithm to produce unjammed initial configurations. The remaining unjammed spheres are then compressed to a jammed state using the SLP algorithm. Such a procedure is repeated $n_r=5-10$ times before a lower limit on the final jammed-packing density ϕ is reached. Low-density jammed packing with densities in the range $\phi \in [0.6, 0.64]$ have been produced (removing the rattlers in the packing results in a slightly lower density, $\phi \approx 0.595$). These packings are mapped onto the ϕ - \mathfrak{T} diagram (red squares). We note that a similar procedure was employed in Ref. [41] to produce low-density jammed packings. However, there the LS algorithm instead of the SLP algorithm was used to generate the initial configurations. Moreover, we would like to emphasize that the low-density jammed packings produced by the SLP algorithm are completely unrelated to the so-called ill-defined “random loose packings” of hard spheres, which have been shown to be not even collectively jammed [46].

“Tunneled” crystal packings (green triangles) [8] and maximal-density packings (fcc, hexagonal close-packed, and their stacking variants called the Barlow packings) are also shown on the order map. The blue dashed curves show the spectrum of packings generated by randomly filling the vacancies in the tunneled crystal packings, which leads to the corresponding maximal-density packings. These dashed curves were first reported in Ref. [41]. The tunneled crystals have density $\phi = \sqrt{2\pi}/9 = 0.49365\dots$ are currently the best candidates for the lowest density strictly jammed packings in three dimensions. Each sphere in any of the tunneled crystals contacts exactly seven others and therefore such packings are *hyperstatic*.

We note that for comparison to existing literature, the data presented in Fig. 6 for the order map include the rattlers, which constitute a small percent of spheres near the MRJ state (e.g., $2-3\%$). The fraction of rattlers decreases as the packing density increases or decreases from MRJ state along the lower boundary of jammed states. In addition, consistent with the results reported in Ref. [41], we find that the average contact number Z per particle increases from the isostatic

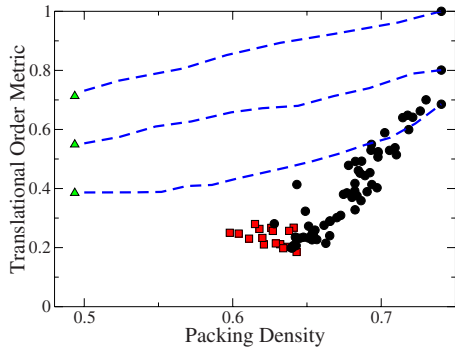


FIG. 6. (Color online) Order map for strictly jammed packings of congruent spheres in three dimensions with $N \sim 1000$ generated using the SLP algorithm: translational order metric \mathcal{T} versus packing density ϕ . The black circles are data produced via the SLP algorithm using random initial conditions. While the density in this data set ranges essentially continuously from $\phi \approx 0.64$, which coincides with that of the MRJ state, up to the maximal density of $\phi = \pi/\sqrt{18} = 0.7408\dots$, the degree of order at fixed density is variable. The red squares are data corresponding to disordered jammed packings with anomalously low densities, down to density as low as $\phi \approx 0.595$, which were generated via the SLP protocol using special initial conditions involving diluted MRJ packings, as described in the text. The dashed blue curves represent the spectrum of packings generated by randomly filling the vacancies in three of the tunneled crystal packings [8] with density $\phi = \sqrt{2}\pi/9 = 0.49365\dots$ (green triangles), each of which leads to the corresponding maximal-density Barlow packing (with $\phi = \pi/\sqrt{18} = 0.7408\dots$).

value of 6 at the MRJ state as the density increases or decreases from the MRJ-state value along the lower boundary of jammed states. In particular, at $\phi \approx 0.6$ the average contact number $Z \approx 6.3$. It is expected that Z will increase to a contact value per particle of 7 as ϕ is decreased to the tunneled-crystal value $\phi = 0.49365\dots$. On the other hand, as ϕ is increased from the MRJ value, the average contact number Z continuously increases from 6 all the way up to 12 for the hyperstatic Barlow packings with a density of $0.74048\dots$. Thus, moving off the MRJ state along the lower boundary of jammed states (in either direction in density) is associated with an increase in the average contact number Z and the degree of order.

VII. DISCUSSION AND CONCLUSIONS

In this paper, we have proposed and implemented a sequential-linear-programming procedure to solve the adaptive-shrinking-cell optimization problem to generate jammed sphere packings. The SLP procedure is particularly suitable and natural for sphere packings with a size distribution because the objective function and impenetrability constraints can be exactly linearized and the final state is ensured to be jammed in principle. We have shown that the SLP algorithm can produce robustly a wide spectrum of jammed sphere packings in \mathbb{R}^d for $d=2, 3, 4, 5$, and 6 with varying degrees of disorder and densities. In particular, we applied the algorithm to generate various disordered packings as well as the maximally dense packings for $d=2, 4, 5$, and 6. More-

over, we showed that our SLP algorithm can produce with high probability a variety of strictly jammed packings with a packing density anywhere in the wide range $[0.6, 0.7408\dots]$ in three dimensions, which supports the view that there is no universal jamming point that is distinguishable based on the packing density alone [14,31,41]. Our jammed sphere packings are characterized and compared to the corresponding packings generated by the well-known and versatile Lubachevsky-Stillinger molecular-dynamics packing algorithm. Compared to the LS procedure, our SLP protocol is able to ensure that the final packings are truly jammed, produces disordered jammed packings with anomalously low densities, and is appreciably more robust and computationally faster at generating maximally dense packings, especially as the space dimension increases. Therefore, the SLP algorithm for producing jammed sphere packings retains the versatility and advantages of the LS algorithm while improving upon its imperfections.

An important feature of the SLP algorithm is that it can produce a broad range of inherent structures (locally maximally dense and mechanically stable packings), besides the usual disordered ones (such as the maximally random jammed state) with very small computational cost compared to those of the best known packing algorithms by tuning the radius of the *influence sphere*. This is to be contrasted with other packing protocols, such as the conjugate-gradient method for soft three-dimensional spheres [52] and the Zinchenko algorithm for three-dimensional hard spheres [51], which can only lead with high probability to the MRJ-like inherent structures from random initial configurations. These protocols are limited in the sense that the trajectory from one point to another on the energy landscape is strongly limited, i.e., only those directions corresponding to local steepest descents can be chosen. This makes the final packings strongly dependent on the random initial configurations. As we have shown, exploring other directions in the energy landscape can lead to a variety of jammed-packing structures with variable disorder and density even if the same random initial configuration is used. In addition, it is not clear that these other packing protocols would lead to inherent structures with the same level of order/disorder, since no order metrics have been explicitly measured for the resulting jammed packings. This emphasizes a major point made in Refs. [14,31,41], namely, although the diversity of packing protocols have yet to be fully explored, there is already substantial evidence that available algorithms, including the one reported in this paper, can produce jammed packings at the same density but with a wide range of order/disorder.

A straightforward generalization of the current SLP procedure for hard-sphere packings in Euclidean space \mathbb{R}^d is to devise a linear-programming protocol for packing spheres in curved spaces, such as the surface of a sphere. In such cases, additional constraints on the sphere displacements are necessary to keep them on the spherical surface, but these constraints are simple to incorporate.

One advantage that the LS packing protocol has over any SLP algorithm is in producing jammed packings of smoothly shaped nonspherical particles (e.g., ellipsoids and superballs) because the nonoverlap functions are nonlinear [36–39] and linearization of the constraints is no longer rigorous and

hence jamming cannot be guaranteed. However, in such instances, the ASC scheme can be solved using Monte Carlo methods, as was done for hard-polyhedron packings [43], or by using nonlinear-programming methods [56]. When the particle shape deviates only slightly from a perfect sphere, a LP solution could still be possible. Moreover, if one is interested in the true dynamics that leads to jamming, the LS algorithm is clearly preferred over the SLP protocol, which is a deterministic algorithm that is incapable of capturing the real dynamics. However, if one is simply interested in generating jammed packings without regard to history, which the geometric-structure point of view advocates [4,14,41], then the SLP protocol is preferable.

Finally, we note that it is highly desirable to devise packing protocols that targets the density or other packing characteristics while ensuring jamming. It is natural to formulate such an inverse problem as an optimization problem [57] and

to solve it for sphere packings using a LP solution procedure. In future work, we will generalize the current SLP algorithm to create even lower density jammed states of hard spheres than have been able to produced thus far. In three dimensions, this could enable the large gap without any jammed states between densities ~ 0.49 and ~ 0.6 with relatively low order metric values (see Fig. 6) to be filled in.

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- [1] J. P. Hansen and I. R. McDonald, *Theory of Simple Liquids* (Academic, New York, 1986).
- [2] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, New York, 2000).
- [3] R. Zallen, *The Physics of Amorphous Solids* (Wiley, New York, 1983).
- [4] S. Torquato, T. M. Truskett, and P. G. Debenedetti, *Phys. Rev. Lett.* **84**, 2064 (2000).
- [5] G. Parisi and F. Zamponi, *Rev. Mod. Phys.* **82**, 789 (2010).
- [6] S. Torquato, *Random Heterogeneous Materials: Microstructure and Macroscopic Properties* (Springer, New York, 2002).
- [7] Z. W. Salsburg and W. W. Wood, *J. Chem. Phys.* **37**, 798 (1962).
- [8] S. Torquato and F. H. Stillinger, *J. Appl. Phys.* **102**, 093511 (2007); **103**, 129902 (2008).
- [9] T. Aste and D. Weaire, *The Pursuit of Perfect Packing* (Taylor & Francis, New York, 2008).
- [10] W. B. Russel, D. A. Saville, and W. R. Schowalter, *Colloidal Dispersions* (Cambridge University Press, Cambridge, England, 1989).
- [11] *Granular Matter*, edited by A. Mehta (Springer, New York, 1994).
- [12] S. F. Edwards and D. V. Grinev, *Phys. Rev. Lett.* **82**, 5397 (1999); *Chem. Eng. Sci.* **56**, 5451 (2001).
- [13] S. Torquato and F. H. Stillinger, *J. Phys. Chem. B* **105**, 11849 (2001).
- [14] S. Torquato and F. H. Stillinger, *Rev. Mod. Phys.* **82**, 2633 (2010).
- [15] J. L. Gevertz and S. Torquato, *PLOS Comput. Biol.* **4**, e1000152 (2008).
- [16] T. I. Zohdi, *Mech. Mater.* **38**, 969 (2006).
- [17] A. Mejdoubi and C. Brosseau, *J. Appl. Phys.* **101**, 084109 (2007).
- [18] S. Torquato, *Annu. Rev. Mater. Res.* **40**, 101 (2010).
- [19] N. Ch. Karayiannis and M. Laso, *Phys. Rev. Lett.* **100**, 050602 (2008).
- [20] K. Foteinopoulou, N. Ch. Karayiannis, M. Laso, M. Kroger, and M. L. Mansfield, *Phys. Rev. Lett.* **101**, 265702 (2008).
- [21] C. E. Shannon, *Bell Syst. Tech. J.* **27**, 379 (1948); **27**, 623 (1948).
- [22] J. H. Conway and N. J. A. Sloane, *Sphere Packings, Lattices and Groups* (Springer-Verlag, New York, 1998).
- [23] H. L. Frisch and J. K. Percus, *Phys. Rev. E* **60**, 2942 (1999).
- [24] G. Parisi and F. Slanina, *Phys. Rev. E* **62**, 6554 (2000).
- [25] S. Torquato and F. H. Stillinger, *Phys. Rev. E* **73**, 031106 (2006).
- [26] M. Skoge, A. Donev, F. H. Stillinger, and S. Torquato, *Phys. Rev. E* **74**, 041127 (2006).
- [27] S. Torquato and F. H. Stillinger, *Exp. Math.* **15**, 307 (2006).
- [28] J. A. van Meel, D. Frenkel, and P. Charbonneau, *Phys. Rev. E* **79**, 030201 (2009).
- [29] H. Cohn and N. Elkies, *Ann. Math.* **157**, 689 (2003).
- [30] H. Cohn and A. Kumar, *Ann. Math.* **170**, 1003 (2009).
- [31] A. R. Kansal, S. Torquato, and F. H. Stillinger, *Phys. Rev. E* **66**, 041109 (2002).
- [32] T. C. Hales, *Ann. Math.* **162**, 1065 (2005).
- [33] T. M. Truskett, S. Torquato, and P. G. Debenedetti, *Phys. Rev. E* **62**, 993 (2000).
- [34] A. Donev, F. H. Stillinger, and S. Torquato, *Phys. Rev. Lett.* **95**, 090604 (2005).
- [35] B. D. Lubachevsky and F. H. Stillinger, *J. Stat. Phys.* **60**, 561 (1990); B. D. Lubachevsky, F. H. Stillinger, and E. N. Pinson, *ibid.* **64**, 501 (1991).
- [36] A. Donev, S. Torquato, and F. H. Stillinger, *J. Comput. Phys.* **202**, 737 (2005); **202**, 765 (2005).
- [37] A. Donev, R. Connelly, F. H. Stillinger, and S. Torquato, *Phys. Rev. E* **75**, 051304 (2007).
- [38] Y. Jiao, F. H. Stillinger, and S. Torquato, *Phys. Rev. Lett.* **100**, 245504 (2008).
- [39] Y. Jiao, F. H. Stillinger, and S. Torquato, *Phys. Rev. E* **79**, 041309 (2009).
- [40] The sphere expansion rate γ is defined as $\gamma = \frac{1}{D} \frac{dD}{dt}$, where D is the diameter of the sphere.
- [41] Y. Jiao, F. H. Stillinger, and S. Torquato, *J. Appl. Phys.* (to be published).

- [42] In Ref. [41], it was shown that jammed packings with a very narrow density distribution centered at any ϕ over a wide range $\phi \in [0.6, 0.7408 \dots]$ with variable disorder can be generated in three dimensions. These results as well as those reported here support the view that there is no universal jamming point that is distinguishable based on the packing density alone and occurrence frequency. These results stress the importance of the “geometric-structure” approach to analyzing packing problems [14]. In Ref. [41], the SLP algorithm was employed to produce disordered jammed sphere packings in three dimensions with anomalously low densities. However, the algorithmic details of the SLP solution were not provided in that paper. Here we also use the SLP algorithm to generate disordered jammed sphere packings with anomalously low densities but using only initial conditions produced by the SLP algorithm. Additional details are provided in Sec. VI.
- [43] S. Torquato and Y. Jiao, *Phys. Rev. E* **80**, 041104 (2009); *Nature (London)* **460**, 876 (2009).
- [44] In practice, the relative coordinates (rather than the global coordinates) are used in the algorithm due to the changing sphere positions with respect to the adaptive fundamental cell. Note that the terminology “global” coordinate was not explicitly used in Ref. [43], even though the equations for the global positions of particle centers were explicitly given.
- [45] A. Donev, S. Torquato, and F. H. Stillinger, *Phys. Rev. E* **71**, 011105 (2005).
- [46] A. Donev, S. Torquato, F. H. Stillinger, and R. Connelly, *J. Appl. Phys.* **95**, 989 (2004); *J. Comput. Phys.* **197**, 139 (2004).
- [47] F. H. Stillinger and T. A. Weber, *Phys. Rev. A* **25**, 978 (1982); *Science* **225**, 983 (1984). In these papers, the concept of inherent structures was formalized for many-particle system interacting via continuous soft potentials. The set of configurational points that map to the same mechanically stable local energy minimum via a steepest-descent trajectory define uniquely a basin associated with the local minimum called an inherent structure.
- [48] F. H. Stillinger and T. A. Weber, *J. Chem. Phys.* **83**, 4767 (1985). In this paper, the concept of inherent structures was rigorously generalized to hard-sphere systems by considering the infinite- n limit of purely repulsive power-law pair potentials $1/r^n$.
- [49] F. H. Stillinger, E. A. DiMarzio, and R. L. Kornegay, *J. Chem. Phys.* **40**, 1564 (1964).
- [50] A packing is isostatic if it possesses the minimal number of contacts for a particular jamming category. In large-packing limit, this implies that the average contact per particle in a strictly jammed packing is equal to $2d$, where d is the spatial dimension [14,45].
- [51] A. Zinchenko, *J. Comput. Phys.* **114**, 298 (1994).
- [52] C. S. O’Hern, L. E. Silbert, A. J. Liu, and S. R. Nagel, *Phys. Rev. E* **68**, 011306 (2003).
- [53] L. Fejes Tóth, *Regular Figures* (Macmillan, New York, 1964).
- [54] Y. Kallus and V. Elser, and S. Gravel, *Phys. Rev. E* **82**, 056707 (2010).
- [55] We note that there is a slight difference between the pair correlation functions for the three-dimensional MRJ packings produced by the LS and LSP algorithms, which appears to be primarily due to the statistical noise. The corresponding translational order metrics of the packings are measured to be $\mathfrak{T}_{LS}=0.197$ and $\mathfrak{T}_{SLP}=0.194$, which indicate that the packings virtually have the same degree of disorder.
- [56] A. Ruszczyński, *Nonlinear Optimization* (Princeton University Press, Princeton, NJ, 2006).
- [57] S. Torquato, *Soft Matter* **5**, 1157 (2009).