Breakdown of Elasticity Theory for Jammed Hard-Particle Packings: Conical Nonlinear Constitutive Theory

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Abstract

Hard-particle packings have provided a rich source of outstanding theoretical problems and served as useful starting points to model the structure of granular media, liquids, living cells, glasses, and random media. The nature of "jammed" hard-particle packings is a current subject of keen interest. We demonstrate that the response of jammed hard-particle packings to global deformations cannot be described by linear elasticity (even for small particle displacements) but involves a "conical" nonlinear constitutive theory. It is the singular nature of the hard-particle potential that leads to the breakdown of linear elasticity. Interestingly, a nonlinear theory arises because the feasible particle displacements (leading to unjamming) depend critically on the local spatial arrangement of the particles, implying a directionality in the feasible strains that is absent in particle systems with soft potentials. Mathematically, the set of feasible strains has a conical structure, i.e., components of the imposed strain tensor generally obey linear inequalities. The nature of the nonlinear behavior is illustrated by analyzing several specific packings. Finally, we examine the conditions under which a packing can be considered to "incompressible" in the traditional sense.

1 Introduction

Bernal (1965) has remarked that the problem of packing particles into a container or vessel is one of the oldest problems known to man. Hard-particle packings are an enduring source of many challenging theoretical problems and have served as useful starting points to study the structure of diverse many-body systems such as granular media, liquids, living cells, glasses, and random media (Bernal 1965, Mehta and Barker 1991, Edwards 1994, Reiss, Frisch and Lebowitz 1959, Hansen and McDonald 1986, Zallen 1983, Torquato 2002). The utility of hard-particle packings as models for dense many-body systems derives from the fact that repulsive forces are primarily responsible for determining their structure.

Many of the most difficult open questions involve classification and enumeration of "random" (i.e., irregular) disk and sphere packings. Indeed, it was recently shown that the venerable notion of the "random close packing" RCP state is in fact mathematically ill-defined and must be replaced by a new notion called the *maximally random jammed* (MRJ) state, which can be made precise (Torquato, Truskett and Debenedetti 2000). The identification of the MRJ state rests on the development of metrics for order (or disorder), a very challenging problem in condensed-matter theory, and a precise definition for the term "jammed." A preliminary study (Kansal, Torquato and Stillinger 2002) has attempted to identify the MRJ state (see Fig. 1). Besides the MRJ state one may wish to know the lowest density jammed structure(s), which is an open problem in two and three dimensions for nontrivial jamming categories described below.

Jammed configurations of hard particles are of great fundamental and practical interest. Three distinct categories of packings have been distinguished, depending on their behavior with respect to nonoverlap geometric constraints and/or externally imposed virtual displacements: locally jammed, collectively jammed, and strictly jammed (Torquato and Stillinger 2001). These categories are defined precisely in Section II. Roughly speaking, these jamming categories, listed in order of increasing stringency, reflect the degree of "rigidity" of the packing.

The purpose of this paper is to show that the response of jammed hard-particle packings to global deformations cannot generally be described by linear elasticity, but involves a well-defined alternative formalism. [1] Although we specialize to hard circular disks in two dimensions and hard spheres in three dimensions, the general conclusions of this paper apply as well to hard particles of arbitrary shape. Hard particles interact with each other only when they touch, and then with an infinite repulsion reflecting their impenetrable physical volume. Thus, the pair potential $\varphi(r)$ for hard disks or spheres of diameter D is specified by

$$\varphi(r) = \begin{cases} +\infty, & r \leq D, \\ 0, & r > D, \end{cases} \tag{1}$$

where r is the interparticle separation distance. It is the singular nature of this potential which leads to the general failure of linear elasticity in characterizing the stress-strain behavior of jammed hard-particle packings. A nonlinear constitutive theory arises due to the fact that feasible or allowable nonoverlapping sphere displacements depend critically on the local geometry (spatial arrangement of the particles). A manifestation of this dependence on the local geometry is the fact that jammed hard-particle packings cannot withstand expansions of the system but may withstand compressions. This directionality results in a nonlinear constitutive law for small particle displacements. By contrast, systems of particles with a "soft" potential, even if nonlinear, will result in a linear stress-strain law for sufficiently small displacements. Furthermore, even without linearizing the potential in this nonlinear case, one can still introduce "moduli" that depend on the state or history of the system. This is not true in the case of jammed hard-particle packings. Mathematically, the set of feasible strains is a polyhedral cone for such packings, i.e., components of the imposed strain tensor generally obey linear inequalities.

We should emphasize that the physics of hard-particle many-body phenomena includes a broad spectrum of conditions and responses, only a subset of which we consider in the present paper. We focus exclusively on idealized hard-sphere interactions: a model which has generated an enormous literature. That is, apart from perfectly rigid hard-core interactions, no other interactions are considered. This is in contrast to some studies of granular materials

[1] We note that in contrast to the problem of central concern in this paper, the study of the elastic moduli of hard-sphere systems in equilibrium is a well-trodden area of research. The latter systems are never jammed by definition. We refer the reader to the paper by B.N. Miller [*J. Chem. Phys.* **50**, 2733 (1969)] for literature references. This author discusses the elastic moduli of systems of particles with a power-law pair potential in the limit of infinite-negative exponent (hard-sphere limit) and its ramifications to the theory.

that consider non-spherical and deformable particles, friction, dynamics, and other effects. For a discussion of these other considerations, the reader should consult the recent paper by Roux (2000) and references contained therein.

In the subsequent section, we introduce basic definitions. In Section III, we show via a number of examples how linear elasticity breaks down for jammed hard-particle packings and obtain the appropriate conical nonlinear constitutive relations. We also investigate the conditions under which a packing can be considered to "incompressible" in the traditional sense. In Section IV, we discuss conical nonlinear constitutive theory for general packings, which include the important cases of non-crystalline packings, and packings containing distinct particle species.

2 Definitions and Jamming Categories

A *sphere packing* is a collection of spheres in Euclidean d-dimensional space $\Re^d$ such that the interiors of no two spheres overlap. A sphere packing $P(\mathbf{r}^N)$ of N spheres is characterized by the position vectors of the sphere centers $\mathbf{r}^N \equiv \mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N$. The *packing fraction* ϕ is defined to be the fraction of space covered by the spheres. Next we repeat, with slight modifications, the definitions of several hierarchical jamming categories as taken from Torquato and Stillinger (2001):

Locally jammed: Each particle in the system is locally trapped by its neighbors, i.e., it cannot be translated while fixing the positions of all other particles.

Collectively jammed: Any locally jammed configuration in which no subset of particles can simultaneously be displaced so that its members move out of contact with one another and with the remainder set.

Strictly jammed: Any collectively jammed configuration that disallows all globally uniform volume-nonincreasing deformations of the system boundary.

It is clear that the jamming categories are listed here in increasing order of stringency. Importantly, the jamming category is generally dependent on the type of boundary conditions imposed (e.g., hard wall or periodic boundary conditions) as well as the shape of the boundary. For local jamming each sphere has to have at least $d+1$ contacts with neighboring spheres (3 for disks, 4 for spheres), not all in the same d-dimensional hemisphere. The determination of whether a general packing is collectively and strictly jammed is a considerably more challenging problem. An efficient computational algorithm to test for collective and strict jamming has recently been devised by Donev, Torquato, Stillinger and Connelly (2002).

To begin, it is useful and instructive to classify simple ordered packings of disks or spheres into the above jamming categories. One can create such packings by placing disks or spheres at the sites of simple lattices within an appropriate hard-wall container. This is illustrated for three simple two-dimensional lattices within rectangular in Figure 2. Table 1 classifies common two and three-dimensional lattice packings into the three jamming categories for hard-wall boundaries (Torquato and Stillinger 2001), and Table 2 does the same for periodic boundary conditions (Donev et al. 2002). Figure 3 illustrates why the honeycomb lattice is not collectively jammed with hard-wall boundary conditions and why the Kagomé lattice is not jammed with periodic boundary conditions by showing a possible collective continuous displacement of the particles (Donev et al. 2002). Strict jamming also considers volume-nonincreasing deformations (including shearing) of the boundary and is discussed for periodic boundary conditions in the next section. We note the interesting fact that the Kagomé disk packing, which has an appreciably lower density than the triangular packing, can be made strictly jammed with periodic boundary conditions by suitably reinforcing it (Stillinger, Sakai and Torquato 2003, Donev et al. 2002), as illustrated in Fig. 4.

Note that these definitions of jamming prohibit the presence of "rattlers" (i.e., movable but caged particles) in the system. However, experimental or computational protocols that generate packings commonly contain a small concentration of such rattler particles. Nevertheless, it is the overwhelming majority of spheres that compose the underlying "jammed" network that confers "rigidity" to the particle packing and, in any case, the "rattlers" could be removed without disrupting the jammed remainder. As a final point, we note that whereas the lowest-density states of collectively and strictly jammed systems in two or three dimensions are currently unknown, one can achieve locally jammed packings with an arbitrarily low density (Boroczky 1964). The reader is referred to the paper by Donev et al. (2002) for additional mathematical details regarding these jamming categories as well as their relationships to other definitions used in the mathematical literature.

3 Breakdown of Linear Elasticity

It is important to emphasize that the aforementioned jamming definitions are purely kinematic, i.e., they do not refer to force loads or stresses acting on the system. Nonetheless, one could choose to relate the concomitant stresses to deformations (strains) via appropriate constitutive relations. It is obvious that a strictly jammed system can withstand any imposed non-expansive loading or deformation, implying that the "moduli" (if they exist) are infinite. As noted earlier, examples of strictly jammed systems include the two-dimensional triangular lattice and the three-dimensional FCC lattice (Torquato and Stillinger 2001).

This then raises that natural question as to what less stringent jamming states imply about the resistance to external loadings or deformations. Consider a packing that lies between strict and collective jamming, such as the square lattice in two dimensions and the simple cubic lattice in three dimensions with hard walls (Torquato and Stillinger 2001). For the square lattice, the only allowable non-expansive symmetric strains are pure shears oriented along the rows or columns of particles (see Fig. 5); all shear strains with other orientations are resisted.

For general jammed hard-particle packings, for a given imposed strain, the induced stresses will either be zero or infinity; in other words, some strains are allowed (lead to unjamming), implying *vanishing* "moduli," while others are forbidden (fully resisted), implying *infinitely* large moduli. We will call unjamming strains *feasible* or *allowable* strains. It is important to emphasize that the nonlinearity in the resulting constitutive relation here is quite distinct from usual nonlinearities in which the moduli depend, for example, on the magnitudes or rates of strain components. For jammed hard-particle systems, the "moduli" depend on the orientation of the strain (local geometry). In fact, the set of feasible strains is a *polyhedral cone* in the space of strain components. [2]

[2] A cone C in the mathematical sense is a subset of a vector space such that for all vectors $\mathbf{x} \in C$, $\beta\mathbf{x} \in C$ for all scalar $\beta \geq 0$. A polyhedral cone is a set of vectors that obey a homogeneous system of linear inequalities, $C = \{\mathbf{x} | \mathbf{a}_i \cdot \mathbf{x} \geq 0 \forall i = 1, \ldots, m\}$.

We now show by way of an illustration the conical nature of the feasible strains in jammed hard-particle systems. A good example of a two-dimensional lattice under periodic boundary conditions that has a nontrivial set of feasible symmetric strains is the rhombical lattice. This is a special *periodic packing*. A periodic packing $\widehat{P}(\mathbf{r}^N)$ is generated by a replicating a finite *generating packing* $P(\mathbf{r}^N)$ on a lattice $\boldsymbol{\Lambda} = \{\boldsymbol{\lambda}_1, \ldots, \boldsymbol{\lambda}_d\}$, where $\boldsymbol{\lambda}_i$ are linearly independent *lattice vectors* and d is the space dimension. Note that $\boldsymbol{\Lambda}$ is a matrix containing d^2 elements. A portion of the rhombical lattice and its primitive cell are shown in Fig. 6. The rhombus angle α is a parameter that varies between $\pi/3$ and $\pi/2$. Thus, at the extreme values of α one recovers the common triangular and square lattices. For the rhombical lattice, we choose, for simplicity, the generating packing to be a single particle, in which case we have that (for unit diameter)

$$\boldsymbol{\Lambda} = \begin{bmatrix} 1 & \cos(\alpha) \\ 0 & \sin(\alpha) \end{bmatrix}. \tag{2}$$

Let us focus for the moment on volume-preserving (shear) deformations, i.e., traceless symmetric strains of the form

$$\boldsymbol{\epsilon} = \begin{bmatrix} x & y \\ y & -x \end{bmatrix}. \tag{3}$$

The lattice points of a general lattice are described by position vectors of the form $\mathbf{r} = \boldsymbol{\Lambda} \cdot \mathbf{n}$, where the components of the vector $\mathbf{n}$ span the integers. Thus, if we allow a small distortion of the lattice $\Delta\boldsymbol{\Lambda}$, then the change in the position vectors is $\Delta\mathbf{r} = \Delta\boldsymbol{\Lambda} \cdot \mathbf{n}$ and the corresponding strain is $\boldsymbol{\epsilon} = \Delta\boldsymbol{\Lambda} \cdot \boldsymbol{\Lambda}^{-1}$. Combining these results with the matrix (2) and the imposed strain (3) yields

$$\Delta\boldsymbol{\Lambda} = \begin{bmatrix} x & x\cos(\alpha) + y\sin(\alpha) \\ y & y\cos(\alpha) - x\sin(\alpha) \end{bmatrix}. \tag{4}$$

Therefore, the change in the two lattice vectors are $\Delta\boldsymbol{\lambda}_1 = \big(x, y\big)$ and $\Delta\boldsymbol{\lambda}_2 = \big(x\cos(\alpha) + y\sin(\alpha), y\cos(\alpha) - x\sin(\alpha)\big)$. The impenetrability constraints for the sphere contacts along the sides of the rhombus sides forming the angle α (see Fig. 6) are given by $\Delta\boldsymbol{\lambda}_1 \cdot \boldsymbol{\lambda}_1 \geq 0$

and $\Delta\boldsymbol{\lambda}_2 \cdot \boldsymbol{\lambda}_2 \geq 0$ (Donev et al. 2002). [3] These relations lead to the inequality conditions

$$x \geq 0, \qquad y \geq \frac{1 - 2\cos^2(\alpha)}{\sin(2\alpha)} x. \tag{5}$$

This analysis illustrates the conical nature of the feasible strains for the 1x1 (primitive) unit cell. Figure 7 shows the cone of the components of the feasible strains. We have verified that this result is valid for arbitrarily large unit cells using the algorithms of Ref. 13. Note that the rhombical lattice actually lies between the local and collective jamming categories.

Now let us consider compressive loads or, equivalently, volume-decreasing deformations of the form

$$\boldsymbol{\epsilon} = \begin{bmatrix} x & y \\ y & -(x+\delta) \end{bmatrix}. \tag{6}$$

where δ is non-negative constant. Note this strain has a non-positive trace, namely, its trace is equal to $-\delta$. Here we have a more complicated situation with three parameters, i.e., a three-dimensional cone of feasible strains (whose description we will omit). Note that the usual definition of the bulk modulus involves the unit compressive strain

$$\boldsymbol{\epsilon} = \begin{bmatrix} -1 & 0 \\ 0 & -1 \end{bmatrix}. \tag{7}$$

i.e., uniform shrinkage of the boundary. If a unit compressive strain is not allowed, usual convention says that such a system is *incompressible*, i.e., the bulk modulus is infinite. If a unit compressive strain is allowed (i.e., the packing unjams), then it is common to refer to such a system as *compressible*. However, for the rhombical packing under the deformation specified by (6), we can achieve volume-decreasing deformations (positive δ) only by allowing off-diagonal strain components (nonzero y). Following the same derivation as before, we find that the largest decrease in volume of the system (if y is fixed, say $y = 1$) is achieved when

[3] Although there are four pairs of contacting particles in the primitive cell, only two are independent for $\pi/3 < \alpha \leq \pi/2$. The special case of the triangular lattice ($\alpha = \pi/3$), which is strictly jammed, is not included in this analysis because there are three independent pairs of contacting particles in its primitive cell. Note as the square lattice is approached ($\alpha \to \pi/2$), the only nontrivial feasible strain corresponds to $x \to 0$ and $y \to$ constant, i.e., the pure shear illustrated in Fig. 1.

$x = 0$ and $\delta = -2\cot(\alpha)$.

If we restrict ourselves to the usual definition of incompressibilty, namely, that the bulk modulus is infinite, then we can show that all collectively jammed structures are incompressible. This result follows as a simple corollary of a Theorem due to Connelly (1988). Paraphrasing, his theorem states that a packing is "locally maximally dense" if there is a "rigid" subpacking. The term "rigid" subpacking is equivalent to our definition of collectively jammed. Roughly speaking, a "locally maximally dense" packing is one in which the particles cannot grow uniformly in size to increase the density of the system. This implies that the container cannot undergo a uniform shape-preserving shrinkage. Thus, it follows that a collectively jammed packing is incompressible.

Clearly, incompressibility (in the sense of an infinite bulk modulus) does not necessarily imply a locally jammed configuration. For example, a packing composed of linear chains of spheres that exactly span the length of a square container is incompressible but is not locally jammed (see Fig. 8). Moreover, although there is a variety of locally jammed packings that are incompressible (e.g., honeycomb lattice and diamond lattice), it is not true that all locally jammed configurations are incompressible. More precisely, any locally jammed packing that has a non-vanishing self-stress [4] (i.e., possesses chains of particles that can support a load normal to the boundary) is incompressible. However, not all locally jammed packings have self-stresses and therefore are not all incompressible. For instance, Fig. 9 shows an example of a locally jammed packing that has a vanishing self-stress and hence is compressible.

4 Conical Nonlinear Theory for General Packings

In this paper, we illustrated the conical nature of the set of feasible strains for hard-sphere packings using simple examples, such as the periodic rhombical packing with a primitive lattice generator. Although the same kind of reasoning applies to more general packings, such as packings in hard-wall containers and/or large random packings, the analytical derivation becomes highly nontrivial, and in fact in most cases one cannot obtain closed analytical

[4] A self-stress is a state in which the contacting forces are in equilibrium without any applied internal loads. See, for example, the work by Moukarzel (1998).

descriptions of the cone of feasible strains.

The derivation, in principle, would involve the following steps. First, one writes down the impenetrability constraints between all pairs of contacting particles in terms of dN particle displacements as a system of linear inequalities. Then one expresses the macroscopic (imposed) strain as a linear transformation of the microscopic displacements and reduces the impenetrability constraints to only the $d(d+1)/2$ macroscopic strain components. This last reduction though is not possible analytically, since one cannot map from macroscopic strain tensors to microscopic particle displacements for general packings. Mathematically, one is projecting a (high dimensional) cone of feasible microscopic displacements onto the (low dimensional) space of macroscopic strains to obtain the equivalent cone of feasible strains. This in general requires solving a linear program, which cannot be done analytically because the solution is dependent on the exact positions of the spheres, i.e. on the local geometry of the packing.

We are currently working on investigating numerical and analytical approaches to the characterization of stress-strain behavior for large random and periodic packings of perfectly-hard-sphere and almost-perfectly-hard-sphere packings. This more comprehensive study of the nonlinear behavior of jammed hard-particle packings will be reported in a future work.

5 Acknowledgments

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Table 1: Classification of some of the common jammed ordered lattices of equi-sized disks in two and spheres three dimensions, where Z denotes the coordination number and ϕ is the packing fraction for the infinite lattice. Here hard boundaries are applicable: in two dimensions we use commensurate rectangular boundaries and in three dimensions we use a cubical boundary, with the exception of the hexagonal close-packed lattice in which the natural choice is a hexagonal prism (Torquato and Stillinger 2001). Note that with appropriately placed regular triangular- or hexagonal-shaped boundaries, the Kagomé lattice is locally, collectively and strictly jammed.

Lattice	Locally jammed	Collectively jammed	Strictly jammed
Honeycomb ($Z = 3$, $\phi \approx 0.605$)	yes	no	no
Kagomé ($Z = 4$, $\phi \approx 0.680$)	no	no	no
Square ($Z = 4$, $\phi \approx 0.785$)	yes	yes	no
Triangular ($Z = 6$, $\phi \approx 0.907$)	yes	yes	yes
Diamond ($Z = 4$, $\phi \approx 0.340$)	yes	no	no
Simple cubic ($Z = 6$, $\phi \approx 0.524$)	yes	yes	no
Body-centered cubic ($Z = 8$, $\phi \approx 0.680$)	yes	yes	no
Face-centered cubic ($Z = 12$, $\phi \approx 0.741$)	yes	yes	yes
Hexagonal close-packed ($Z = 12$, $\phi \approx 0.741$)	yes	yes	yes

Table 2: The analog of Table 1 for periodic boundary conditions. The results shown here do not depend on the choice of the unit cell so long as it contains sufficiently many spheres (Donev et al. 2002).

Lattice	Locally jammed	Collectively jammed	Strictly jammed
Honeycomb ($Z = 3$, $\phi \approx 0.605$)	yes	no	no
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Face-centered cubic ($Z = 12$, $\phi \approx 0.741$)	yes	yes	yes
Hexagonal close-packed ($Z = 12$, $\phi \approx 0.741$)	yes	yes	yes

Figure Captions

Figure 1: A strictly jammed packing of 500 spheres (with periodic boundary conditions) near the MRJ state at a packing fraction $\varphi \approx 0.64$.

Figure 2: The triangular, Kagomé and honeycomb lattice packings in rectangular hard-wall containers. The Kagomé is not even locally jammed, the honeycomb is not collectively jammed, and the triangular is strictly jammed (Torquato and Stillinger 2001).

Figure 3: *Left:* A collective unjamming mechanism for the honeycomb lattice inside a hard-wall rectangular container (Donev et al. 2002). The arrows in the figures given here show possible directions of motion of the disks, scaled by some arbitrary constant to enhance the figure. *Right:* Collective unjamming mechanism for the Kagomé lattice with periodic boundary conditions.

Figure 4: The reinforced Kagomé lattice is obtained by adding an extra "row" and "column" of disks to the Kagomé lattice and thus has the same density in the thermodynamic limit, namely, it has every 4th disk removed from the triangular packing. However, the reinforced packing *is* strictly jammed with periodic boundary conditions.

Figure 5: The top figure shows scaled particle displacements for the square lattice as dictated by the pure shear strain imposed. The bottom figure shows the deformed lattice at a later time.

Figure 6: A portion of the rhombical lattice (top). The primitive cell and the rhombus angle α (bottom). Here $\alpha = 75^o$.

Figure 7: The feasible strain components in the x-y plane [c.f. relation (3)] shown as the shaded region (cone). Here $\alpha = 75^o$.

Figure 8: A packing of linear chains of spheres that exactly span the length of a square container. Although the packing is not locally jammed, it is incompressible in the standard sense.

Figure 9: A locally jammed configuration of disks in a rectangular container is subjected to an isotropic compressive strain; arrows indicate scaled particle displacements (top). This applied deformation leads to uniform shrinkage of the container and thus to unjamming motions (bottom).

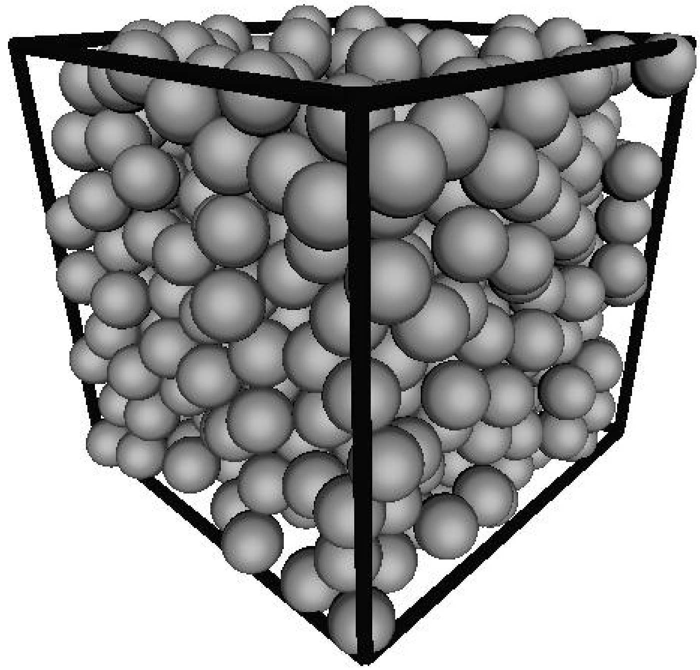
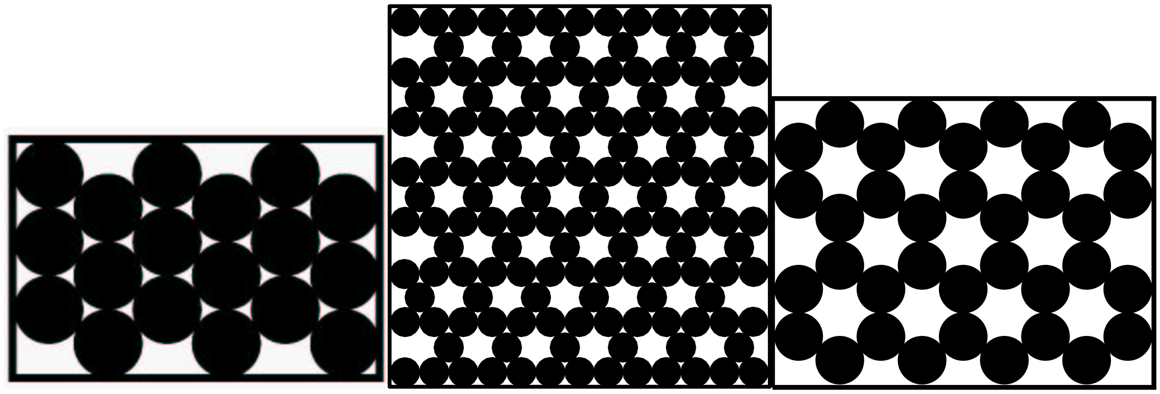


Figure 1: Torquato et al.



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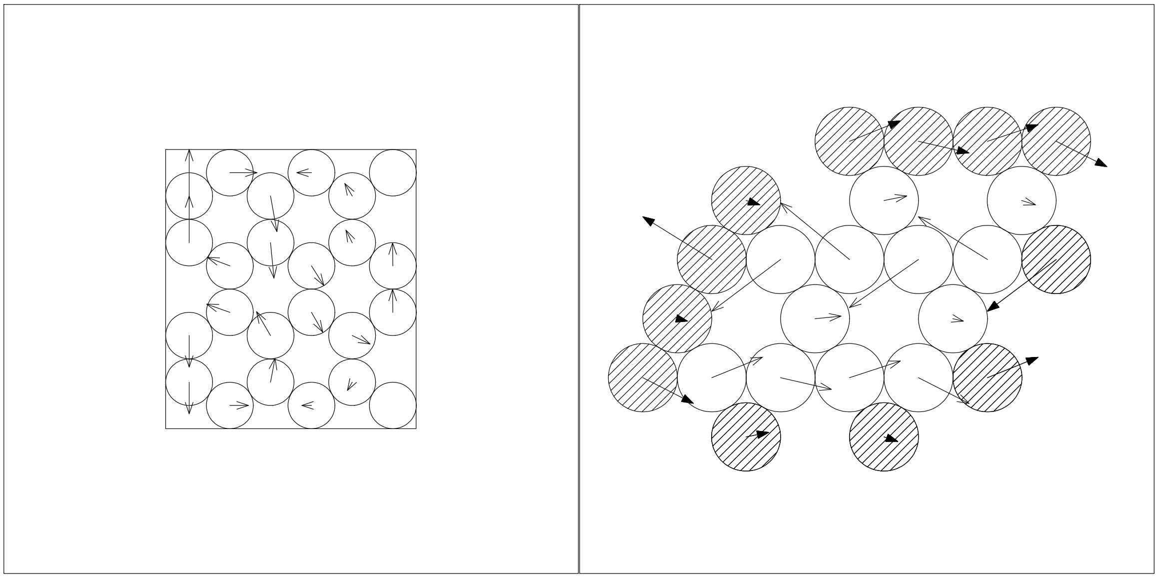

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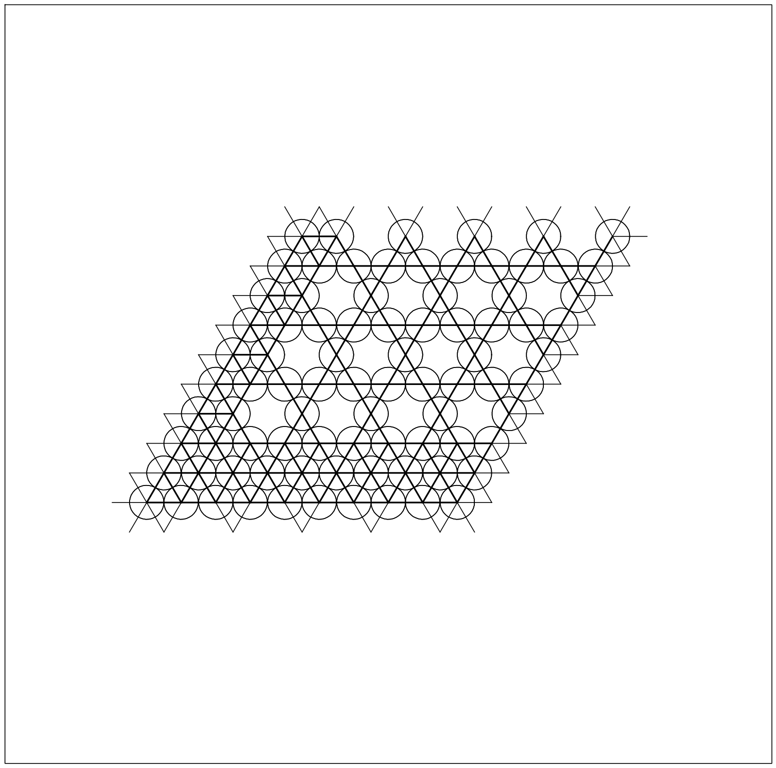

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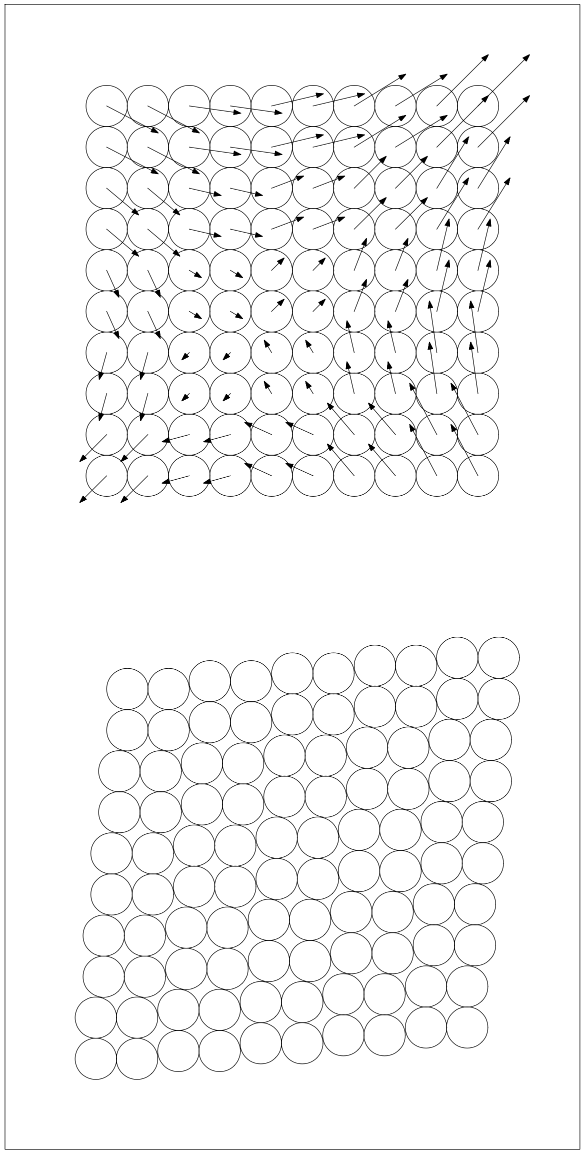

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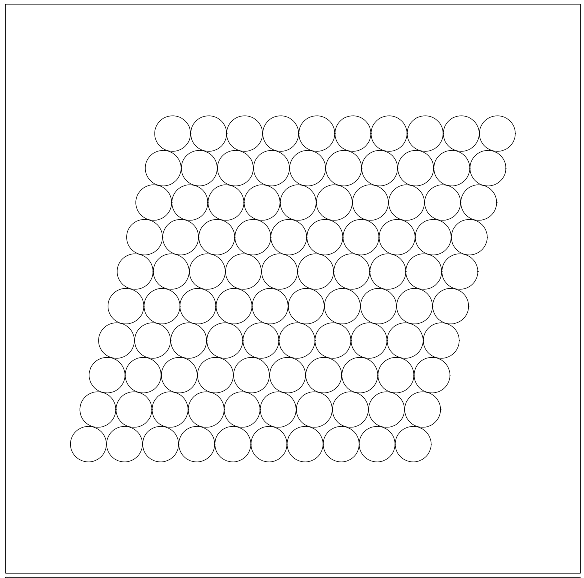

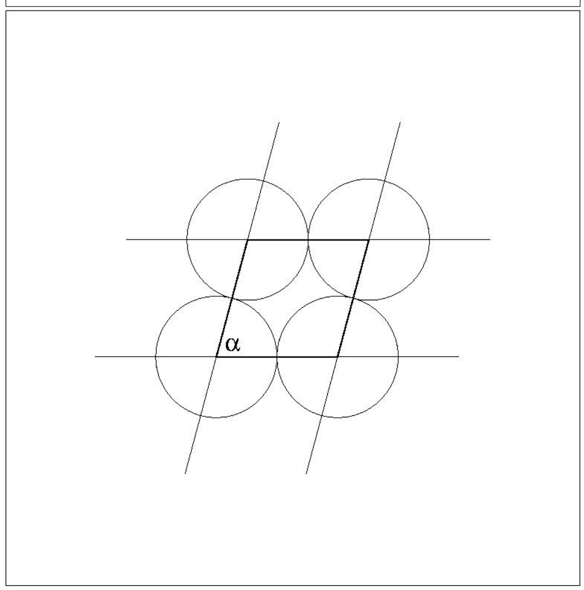

Figure 6: Torquato et al.

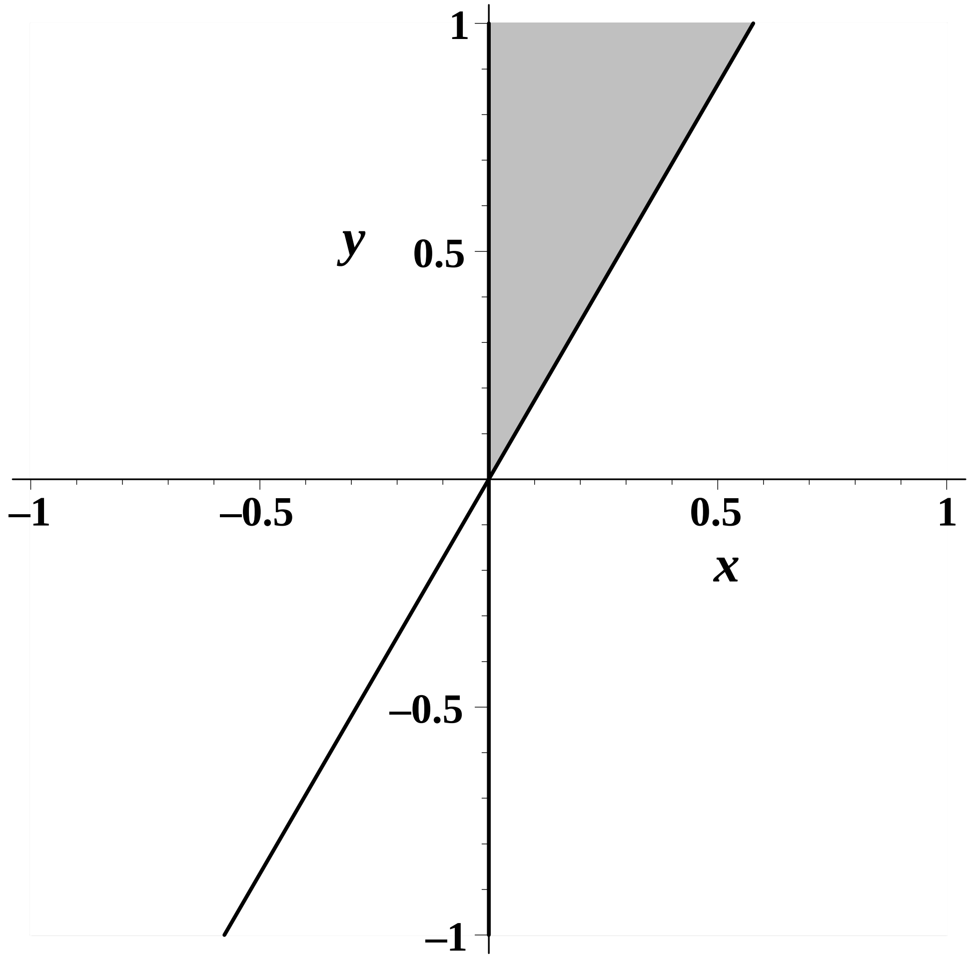

Figure 7: Torquato et al.

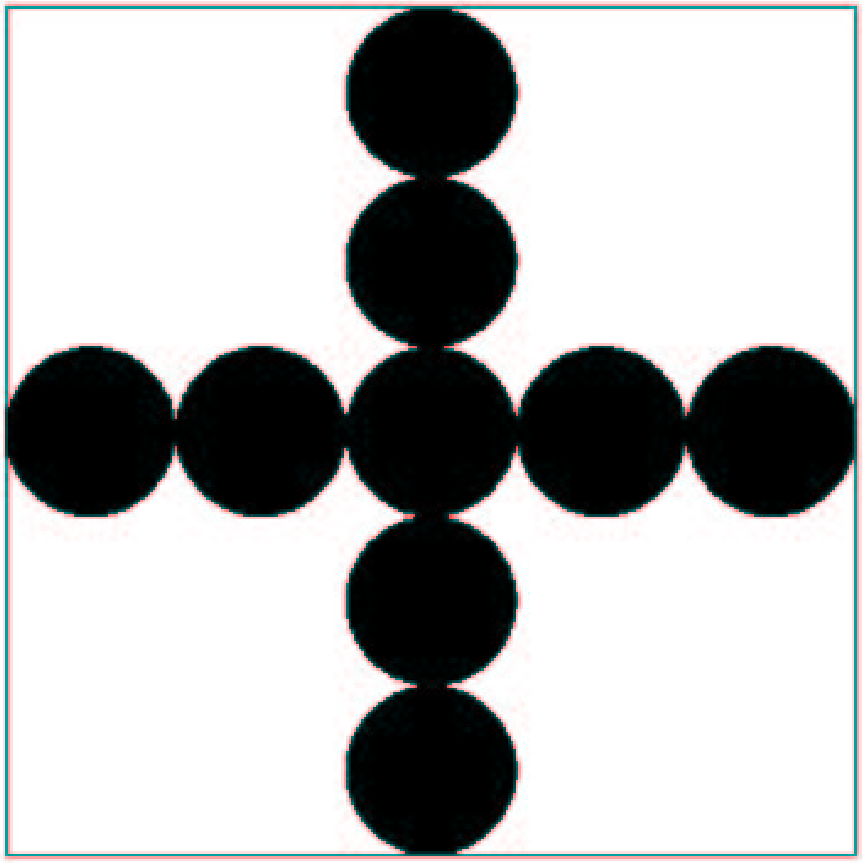

Figure 8: Torquato et al.

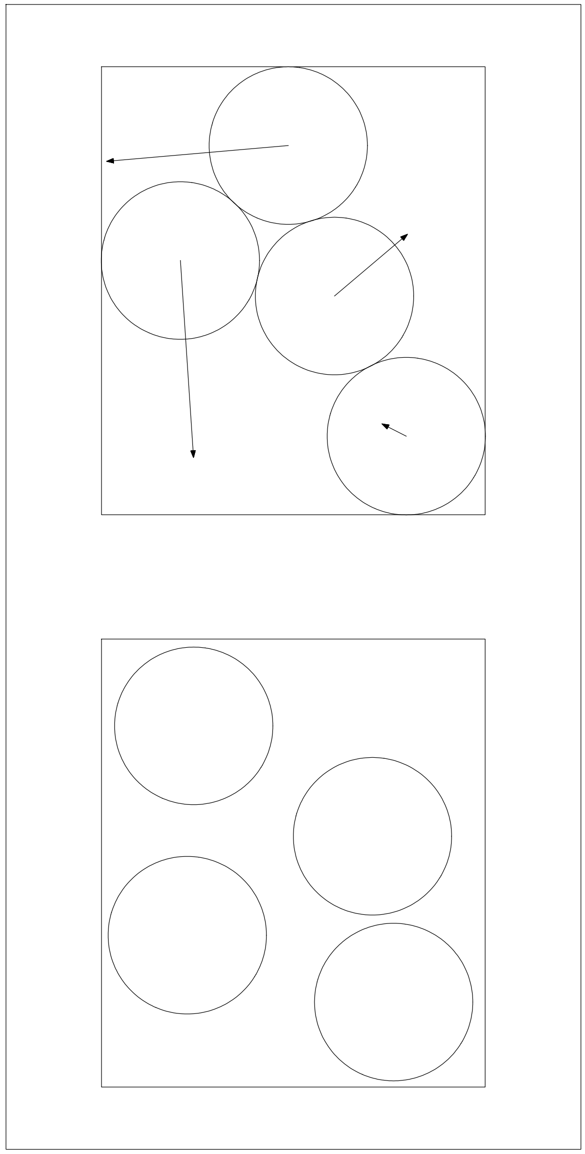

Figure 9: Torquato et al.