Hyperuniform Long-Range Correlations are a Signature of Disordered Jammed Hard-Particle Packings

Chase E. Zachary, Yang Jiao, and Salvatore Torquato

Phys. Rev. Lett. **106**, 178001 — Published 29 April 2011

DOI: 10.1103/PhysRevLett.106.178001

Hyperuniform long-range correlations are a signature of disordered jammed hard-particle packings

Chase E. Zachary,[1] Yang Jiao,[2] and Salvatore Torquato[1,2,3,4*]
[1]*Department of Chemistry,* [2]*Department of Mechanical and Aerospace Engineering,*
[3]*Department of Physics,* [4]*Princeton Center for Theoretical Science,*
Princeton University, Princeton, New Jersey 08544, USA

We show that quasi-long-range (QLR) pair correlations that decay asymptotically with scaling $r^{-(d+1)}$ in d-dimensional Euclidean space $\mathbb{R}^d$, trademarks of certain quantum systems and cosmological structures, are a universal signature of maximally random jammed (MRJ) hard-particle packings. We introduce a novel hyperuniformity descriptor in MRJ packings by studying local-volume-fraction fluctuations and show that infinite-wavelength fluctuations vanish even for packings with size- and shape-distributions. Special void statistics induce hyperuniformity and QLR pair correlations.

PACS numbers:

Quasi-long-range (QLR) pair correlations with asymptotic scaling $r^{-(d+1)}$ are unique trademarks of noninteracting spin-polarized fermionic ground states [1, 2], ground-state liquid helium [3], and the Harrison-Zeldovich spectrum of the early Universe [4]. Such correlations also arise in maximally random jammed (MRJ) packings of monodisperse spheres [5], which are prototypical glasses [6] possessing maximal disorder among all jammed packings with diverging elastic moduli [7–9]. These packings were once thought to describe the structure of liquids [10], which typically possess pair correlations decaying exponentially fast [7, 11–13]. It is thus unclear why special QLR correlations occur in MRJ packings and whether this behavior extends to packings of particles with size- and shape-distributions, which possess nontrivial material properties [14, 15].

Noting that MRJ packings are structurally rigid with a well-defined contact network, Torquato and Stillinger conjectured [7] that all strictly jammed, i.e., globally incompressible and nonshearable, saturated packings of monodisperse spheres in d-dimensional real space $\mathbb{R}^d$ are hyperuniform, i.e., infinite-wavelength local-number-density fluctuations vanish [7], a proposition for which no counterexample has been found to date. Although this conjecture made no mention of QLR pair correlations, further work on 3D MRJ monodisperse sphere packings [5] established that the structure factor $S(k)$ approaches zero linearly as the wavenumber $k \to 0$, inducing a QLR power-law tail r^{-4} in the pair correlation function $g_2(r)$, which is proportional to the probability density of finding a particle within a radial distance r from a reference particle. However, neither study provided a quantitative explanation for the presence of QLR correlations in these systems, and the analysis does not blindly extend to MRJ packings of nonspherical and/or polydisperse particles in which the shape- and size-distributions of the particles are crucial. Also, it is not obvious why MRJ packings are hyperuniform since they lack the crystalline long-range order and Bragg peaks of periodic structures [16] and yet exhibit QLR pair correlations. These unusual characteristics have important implications for understanding the structural properties of MRJ and glassy material systems [17, 18]. We note that hyperuniform disordered point patterns have been used to create new materials with unusual optical properties [19] and large, complete photonic band gaps [20].

In this Letter, we characterize universal properties of polydisperse and/or nonspherical MRJ hard-particle packings. Specifically, we provide strong support for the claim that all saturated, strictly jammed particle packings, which may be polydisperse and/or nonspherical, possess vanishing infinite-wavelength local-volume-fraction fluctuations, a recently-introduced extended notion of hyperuniformity [11]. We thus provide the appropriate novel framework and descriptor to study universal QLR pair correlations in general MRJ packings. By examining binary circular disk, ellipse, and superdisk packings in $\mathbb{R}^2$, we show that infinite-wavelength density fluctuations associated with the particle centers do not vanish due to local microstructural inhomogeneities. However, infinite-wavelength local-volume-fraction fluctuations do vanish, so that the appropriate structural descriptor of MRJ packings is the two-point probability function $S_2(r)$, defined as the probability of finding both of two arbitrary points either within particles or in the exterior void space. Importantly, our methodology contains all previously-reported results for monodisperse MRJ sphere packings [5, 7] as a special case, meaning that we provide a completely general means of understanding QLR correlations in jammed hard-particle packings. We show that the rigidity of the contact network places strong constraints on the size- and spatial-distributions of the local voids, leading to hyperuniform QLR pair correlations that compete with the maximal randomness of the packings. This observation supports the important notion that the topology and geometry of the void space [21] are more basic than the particle space [14].

It is sufficient for our purposes to consider 2D packings of binary circular disks, ellipses, and superdisks, but our arguments concerning the void space are general enough to incorporate higher-dimensional packings. We use the Donev-Torquato-Stillinger algorithm [22] to generate jammed packings of hard particles in $\mathbb{R}^2$. Particles of two different sizes undergo event-driven molecular dynamics with periodic boundary conditions while simultaneously growing at a specified rate and fixed size ratio, resulting in a final configuration that is essentially strictly jammed with a packing fraction $\phi \approx 0.8475$. The configurations that we study contain a small concentration ($\sim 2.5\%$) of "rattler" particles, which are free to move within some small cage. Saturation of the packing places an upper bound on the cage size, and the rattler locations are roughly uniformly distributed in the simulation box. Removing these rattlers breaks hyperuniformity by lifting the saturation constraint [5]; these particles are thus kept in our configurations, which, by numerical protocol design, are close approximants to the MRJ state [5]. We have chosen particle concentrations $\gamma_{\text{small}} = 0.75$ and $\gamma_{\text{large}} = 0.25$ with size ratio $\beta = 1.4$ and have found that varying these parameters does not affect hyperuniformity.

Hyperuniformity is determined by pair separation distances within either a point pattern or a heterogeneous medium. Analysis of point patterns thus requires the pair correlation function g_2 or the associated structure factor $S(k) = 1 + \rho\mathfrak{F}\{g_2(r) - 1\}(k)$ with $\mathfrak{F}$ the Fourier transform. Using these functions, one can write the asymptotic behavior of the number variance $\sigma_N^2(R)$ in $\mathbb{R}^2$ as [7]:

$$\sigma_N^2(R) = 4\phi S(0)(R/D)^2 + \text{lower order terms}, \tag{1}$$

where D is an effective length scale, taken here to be the average diameter ($D = \gamma_{\text{small}}D_{\text{small}} + \gamma_{\text{large}}D_{\text{large}}$) of the particles, and R is the radius of a circular observation window. It follows from (1) that any point pattern with $S(0) = 0$ does not possess infinite-wavelength local number density fluctuations and thus is hyperuniform. For heterogeneous media consisting of finite-volume inclusions, such as the binary MRJ packings we study here, one should consider the *two-point probability function* $S_2(r)$, defined previously, and the associated *spectral density* $\hat{\chi}(k) = \mathfrak{F}\{S_2(r) - \phi^2\}(k)$. Using these functions, the variance $\sigma_\tau^2(R)$ in the *local volume fraction* τ, the volume fraction of a reference phase in an observation region, can be asymptotically written in $\mathbb{R}^2$ as [11]:

$$\sigma_\tau^2(R) = [\rho/(4\phi)]\hat{\chi}(0)(D/R)^2 + \text{lower order terms}. \tag{2}$$

As for point patterns, any heterogeneous medium with $\hat{\chi}(0) = 0$ is hyperuniform and lacks infinite-wavelength local-volume-fraction fluctuations. Differentiating between the number variance and local-volume-fraction fluctuations is essential to characterize MRJ packings with a size distribution, and such a distinction has not previously been made in the literature.

Both the structure factor and spectral density can be numerically obtained by direct Fourier transform [19] according to $S(\mathbf{k}) = N^{-1}|\sum_{j=1}^{N} \exp(i\mathbf{k}\cdot\mathbf{r}_j)|^2$ and $\hat{\chi}(\mathbf{k}) = V^{-1}|\sum_{j=1}^{N} \exp(i\mathbf{k}\cdot\mathbf{r}_j)\hat{m}(k;R_j)|^2$, where $\hat{m}(k;R) = \mathfrak{F}\{\Theta(R - \|\mathbf{r}\|)\}(k)$ is the Fourier transform of the indicator function for a disk of radius R, $\Theta(x)$ is the Heaviside step function, and $\{\mathbf{r}_j\}$ denotes the particle locations. This result can be generalized to include particles of any geometry by changing the form of $\hat{m}$; the important restriction is that it will only apply for media consisting of impenetrable inclusions. The reciprocal vectors $\mathbf{k}$ are discretized according to the shape of the simulation box. For a square simulation box of side length L, the wave vectors are given by $\mathbf{k} = (2\pi/L)\mathbf{n}$ for $\mathbf{n} \in \mathbb{Z}^2$. To obtain spherically-symmetric forms of the structure factor and spectral density, we radially average over all wavevectors within some small spherical shell in reciprocal space. We have considered system sizes up to $N = 1000000$ particles to probe effectively the small-wavenumber region with even higher resolution, due to dimensional scaling, than previous studies [5].

Figure 1 depicts both the structure factor and spectral density of the binary MRJ disk packing. The small-k behavior of $S(k)$ is nonvanishing at the origin, showing that the point pattern generated by the disk centers is not hyperuniform, a result of the binary nature of the packing. Figure 1 also shows that close-packed clusters of small particles permeate the system, but these local clusters are separated from each other by effective "grain boundaries" arising from the inclusion of large particles. These grain boundaries preclude the suppression of infinite-wavelength local density fluctuations. This result should be contrasted with previous calculations for MRJ packings of *monodisperse* spheres in 3D [5], where $S(k) \sim k$ as $k \to 0$. It follows that the Torquato-Stillinger conjecture as originally stated for the number variance cannot hold for strictly jammed, saturated sphere packings with a size distribution. Nevertheless, the physical motivation behind this conjecture is intuitively appealing, and our results for the spectral density in Fig. 1 show that it continues to hold in the aforementioned generalized sense upon analyzing local-volume-fraction fluctuations.

We have fit the small-k region of the spectral density with a minimal third-order polynomial of the form $\hat{\chi}(k) \sim \sum_{i=0}^{3} a_i k^i$ and have found $a_0 = (1.0 \pm 0.2) \times 10^{-5}$, strongly indicating that this packing is hyperuniform. Thus, like 3D MRJ monodisperse sphere packings [5], the spectral density is linear at the origin, implying that asymptotic local-volume-fraction fluctuations decay logarithmically faster than the observation window volume, i.e., $\sigma_\tau^2(R) \sim$

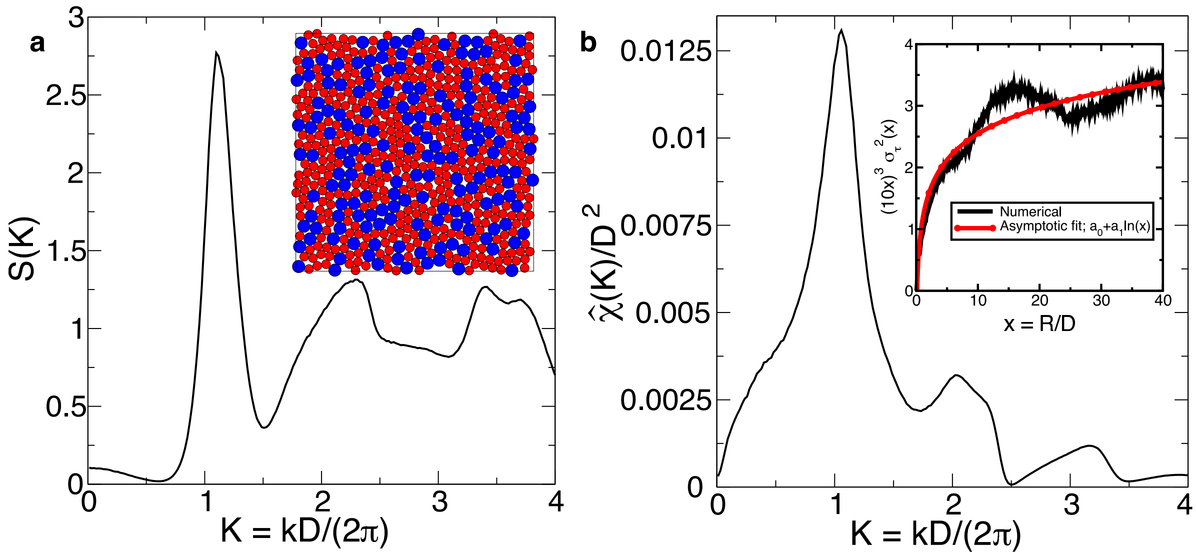

FIG. 1: (Color online) (a) Structure factor and local configuration (inset) for the binary MRJ packing. Large and small particles are shown in blue and red, respectively. (b) Corresponding spectral density with induced local-volume-fraction fluctuations (inset). The local-volume-fraction fluctuations decay logarithmically faster than the volume of an observation window, implying both hyperuniformity and the presence of quasi-long-range correlations.

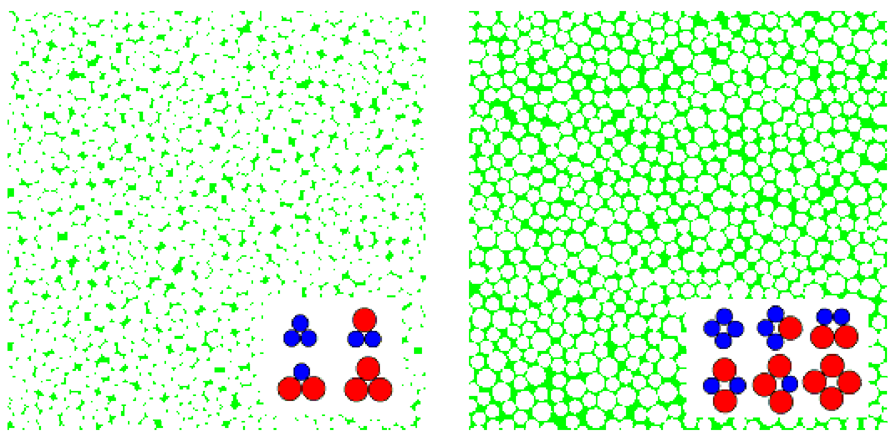

FIG. 2: (Color online) Jamming constraints on the void space. The left panel shows the MRJ state, which is dominated by three-particle loops of the types shown. The right panel is a nearly-jammed configuration with volume fraction $\phi = 0.75$, which exhibits an increasing fraction of skewed higher-order loops (also shown) as the void space becomes more connected.

$(b_0 \ln(R) + b_1)/R^3$ as $R \to +\infty$. This behavior implies that $S_2(r)$ contains a QLR power-law tail r^{-3}, extending to $r^{-(d+1)}$ in $\mathbb{R}^d$ [11].

To clarify the disparity between the number variance and local-volume-fraction fluctuations, we have examined the distribution of the void space. Since the spectral density measures both the inclusion phase and the surrounding matrix [14], hyperuniformity is invariant to the choice of the reference phase, and homogeneity in the void phase is sufficient to induce hyperuniformity. Figure 2 highlights the void phase for the binary MRJ packing in addition to a nonhyperuniform "nearly jammed" disk packing. The MRJ contact network is dominated by local three-particle contacts (or *loops*), but by moving away from the jammed state, higher-order loops become increasingly common. This observation implies that the void-space distribution is inherently constrained by strict jamming. It is true that for any strictly jammed, saturated packing of hard disks (2D) the matrix phase must be disconnected (see Fig. 2), which, although not essential for hyperuniformity, implies that the void distribution is determined completely by the contacts among the particles [24]. In a companion paper [25], we quantify the relationship between jamming and the constrained void space by measuring the distribution of pore sizes and by utilizing rigorous bounds that account for jamming and hyperuniformity.

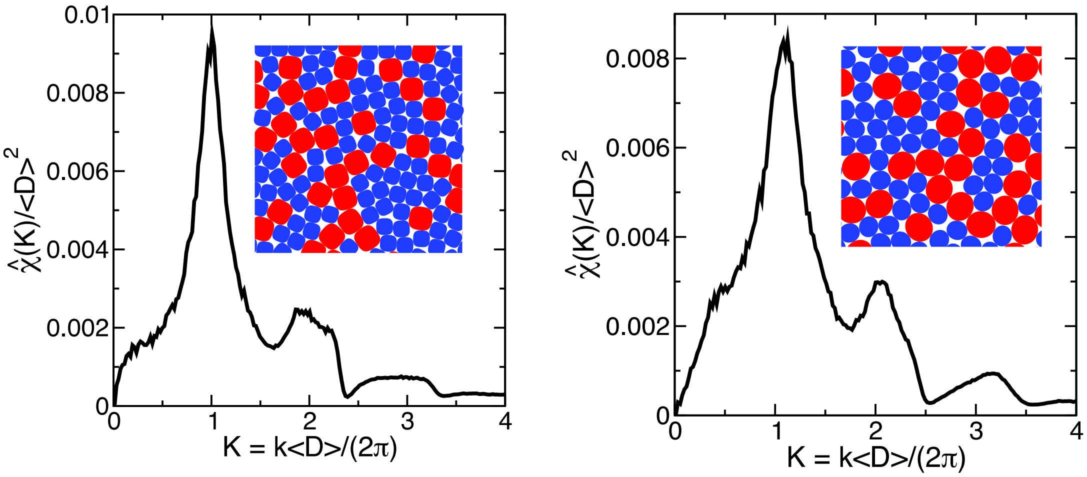

FIG. 3: (Color online) Spectral densities for binary MRJ packings of (left) superdisks ($p = 1.5$) and (right) ellipses ($\alpha = b/a = 1.1$). Also included are portions of the jammed configurations (insets). Large and small particles are in red and blue, respectively.

Jamming is a critical factor for hyperuniformity to hold; even upon moving infinitesimally away from jamming, hyperuniformity is broken. This behavior implies that knowledge of local n-particle contacts is equivalent to specifying the distribution of void sizes. Relaxing the jamming constraint generates nonuniform, connected void-space regions that eventually percolate, skewing the distribution of pore sizes. Thus, the local void sizes in the MRJ packing are inherently correlated with each other via the contact network. Specifically, the presence of a large void limits the sizes of neighboring voids since the particles are strictly jammed. The fact that hyperuniformity is lost upon breaking the contact network implies that these correlations are inherently quasi-long-ranged, thereby suggesting a fundamental relationship between the distribution of the void space and spatially-extended correlations.

We have extended this analysis to MRJ packings of noncircular and nonspherical particles. Figure 3 shows the spectral densities associated with binary MRJ packings of hard superdisks and ellipses [26]. In both cases the packings are hyperuniform even though the inclusion shapes are no longer isotropic. Although the local contacts for these packings are less uniform than for hard disks, the constraints of saturation and strict jamming again limit the distribution of pore sizes and enforce hyperuniformity. Also, the small-k behaviors of the spectral densities for both the superdisk and ellipse packings are linear.

It has been previously observed that this small-wavenumber scaling is directly related to the "degree of order" of the packing [5, 27]. Linear small-k behavior in the spectral density therefore reflects a reasonable amount of variability in the shapes and sizes of the local voids. Lower-order small-wavenumber terms in the spectral density (e.g., $\hat{\chi}(k) \sim k^{1/2}$) are associated with even greater variability in the distribution of the void sizes and therefore reflect increasing probabilities of observing large voids. These types of local-volume-fraction fluctuations are thus incompatible with strict jamming, and the small-wavenumber scaling of the spectral density is minimized to the smallest positive integer value [$\hat{\chi}(k) \sim k$]. Our results thereby suggest that QLR correlations in $S_2(r)$ as induced by a linear small-wavenumber region of the spectral density are a likely sufficient indicator of MRJ particle packings.

We have established universal features of MRJ particle packings using local-volume-fraction fluctuations. Our results generalize the QLR pair correlations of 3D MRJ sphere packings to saturated, strictly jammed hard-particle packings and provide insight into the origin of these correlations. These packings are characterized by QLR pair correlations ($r^{-(d+1)}$ in $\mathbb{R}^d$) with a linear small-wavenumber region in the spectral density, and moving even slightly off the jamming point destroys hyperuniformity. We have justified this property using the variability of the void space, which is both constrained by the local contact network and maximally disordered among all related saturated, strictly jammed packings. Our arguments incorporate higher-dimensional packings, and future work will examine MRJ packings of ellipsoids [28] and superballs [29]. We have also shown that local-number-density fluctuations of the particle centers are in general not sufficient to characterize MRJ packings because the inclusion shapes and sizes are rigorously linked to the void space, meaning that the appropriate quantities to examine are the two-point probability function $S_2(r)$ and the associated spectral density $\hat{\chi}(k)$. We mention that recent, independent work [30] has examined fluctuation-response relations in MRJ packings of polydisperse spheres with similar conclusions to our own. However, no mention is made of quasi-long-range correlations, and MRJ packings of nonspherical particles are not considered. Our arguments provide insight into the structural properties of jammed particle packings and the nature of the jamming transition and suggest certain quantum many-body systems and cosmological structures with special QLR correlations [1–3] are statistically "static" with vanishing infinite-wavelength number density fluctuations vanish and nonanalyticity in the structure factor at small wavenumbers.

This work was supported by the National Science Foundation under Grants DMS-0804431 and DMR-0820341.

* Electronic address: `torquato@princeton.edu`

[1] S. Torquato, A. Scardicchio, and C. E. Zachary, J. Stat. Mech.: Theory and Expt., P11019 (2008).
[2] A. Scardicchio, C. E. Zachary, and S. Torquato, Phys. Rev. E **79,** 041108 (2009).
[3] L. Reatto and G. V. Chester, Phys. Rev. **155,** 88 (1967).
[4] P. J. E. Peebles, 1993, *Principles of Physical Cosmology* (Princeton University Press, Princeton, NJ, 1993).
[5] A. Donev, F. H. Stillinger, and S. Torquato, Phys. Rev. Lett. **95**, 090604 (2005).
[6] We define a glass as a material with a nonzero shear rigidity and without long-range order. This notion is consistent with the description of glassy states in: P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge UP, Cambridge, UK, 1995). The MRJ state is therefore a prototypical glass in that it corresponds to the particular case of diverging elastic moduli and maximal disorder.
[7] S. Torquato and F. H. Stillinger, Phys. Rev. E **68**, 041113 (2003).
[8] S. Torquato, T. M. Truskett, and P. G. Debenedetti, Phys. Rev. Lett. **84**, 2064 (2000).
[9] S. Torquato and F. H. Stillinger, J. Appl. Phys. **102**, 093511 (2007); S. Torquato and F. H. Stillinger, Rev. Mod. Phys. **82**, 2633 (2010).
[10] J. D. Bernal, In *Liquids: Structure, Properties, Solid Interactions*. Hughel, T. J., Ed. (Elsevier, New York, 1965) pp. 25-50.
[11] C. E. Zachary and S. Torquato, J. Stat. Mech.: Theory and Expt., P12015 (2009).
[12] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge UP, New York, 2000).
[13] Exponentially fast decay of pair correlations implies that the structure factor $S(k)$ is analytic at the origin.
[14] S. Torquato, *Random Heterogeneous Materials: Microstructure and Macroscopic Properties* (Springer, New York, 2002).
[15] S. Torquato, J. Stat. Phys. **45**, 843 (1986).
[16] H. Cohn and A. Kumar, J. Am. Math. Soc. **20**, 99 (2007).
[17] G. Yatsenko and K. S. Schweizer, Langmuir **24**, 7474 (2008).
[18] G. Parisi and F. Zamponi, Rev. Mod. Phys. **82**, 789 (2010).
[19] R. D. Batten, F. H. Stillinger, and S. Torquato, J. Appl. Phys. **104**, 033504 (2008).
[20] M. Florescu, S. Torquato, and P. J. Steinhardt, Proc. Nat. Acad. Sc. **106**, 20658 (2009).
[21] The void space of a packing is exterior to the inclusions.
[22] A. Donev, S. Torquato, and F. H. Stillinger, J. Comput. Phys. **202**, 737 (2005).
[23] S. Torquato, Phys. Rev. E **82**, 056109 (2010).
[24] For higher d, the particle and void space can both be connected, but our arguments should hold since the contact network still constrains the local void geometries, inducing correlations between void sizes. Void-space percolation is thus not sufficient to break hyperuniformity.
[25] C. E. Zachary, Y. Jiao, and S. Torquato, in preparation (2010).
[26] A superdisk is defined by the region $|x_1|^{2p} + |x_2|^{2p} \leq 1$ for $p \geq 0$; similarly, an ellipse is given by $(|x_1|/a)^2 + (|x_2|/b)^2 \leq 1$ for $a, b > 0$.
[27] A. Gabrielli, M. Joyce, and S. Torquato, Phys. Rev. E **77**, 031125 (2008).
[28] A. Donev, *et. al.* Science **303**, 990 (2004).
[29] Y. Jiao, F. H. Stillinger, and S. Torquato, Phys. Rev. E **81**, 041304 (2010).
[30] L. Berthier, P. Chaudhuri, C. Coulais, O. Dauchot, and P. Sollich, Phys. Rev. Lett. **106**, 120601 (2011).