

Ensemble Theory for Stealthy Hyperuniform Disordered Ground States

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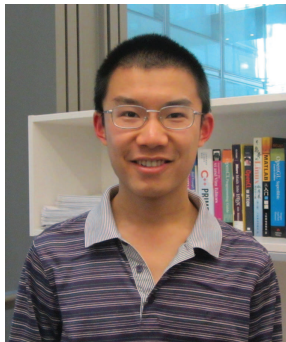
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Princeton University

<http://cherrypit.princeton.edu>

Joint work with Ge Zhang and Frank Stillinger, Princeton University

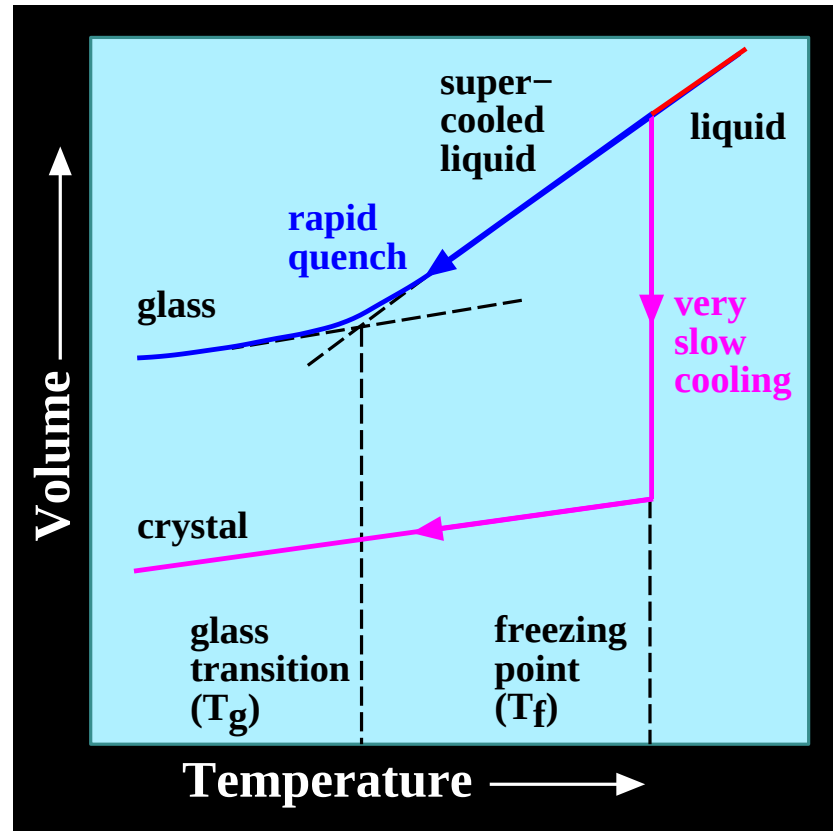


Slow and Rapid Cooling of a Liquid

- Classical **ground states** are those classical particle configurations with **minimal** potential energy per particle.

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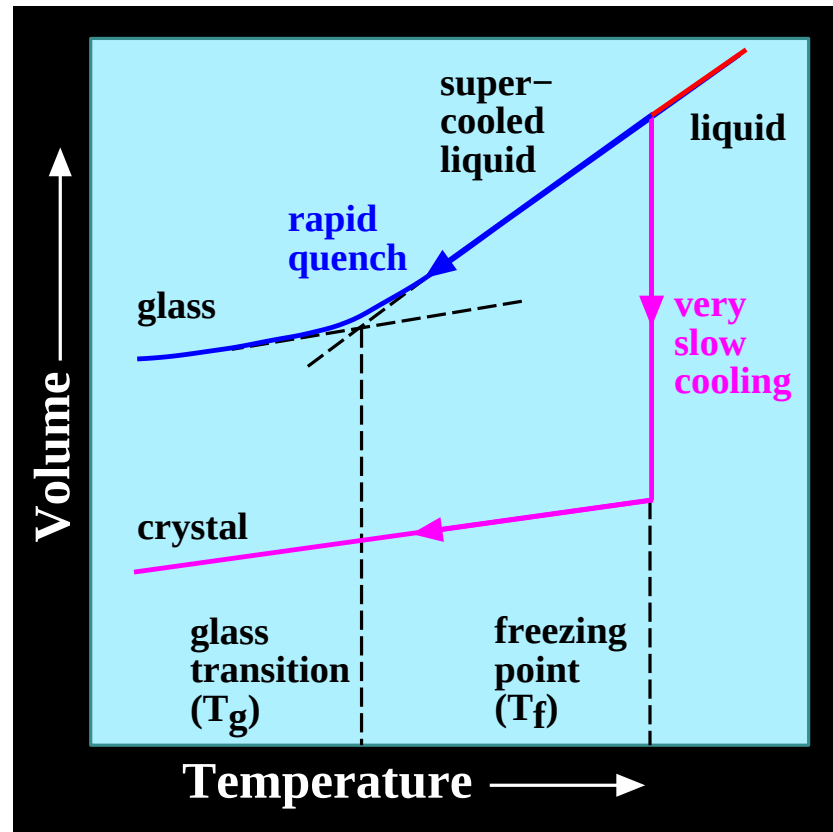
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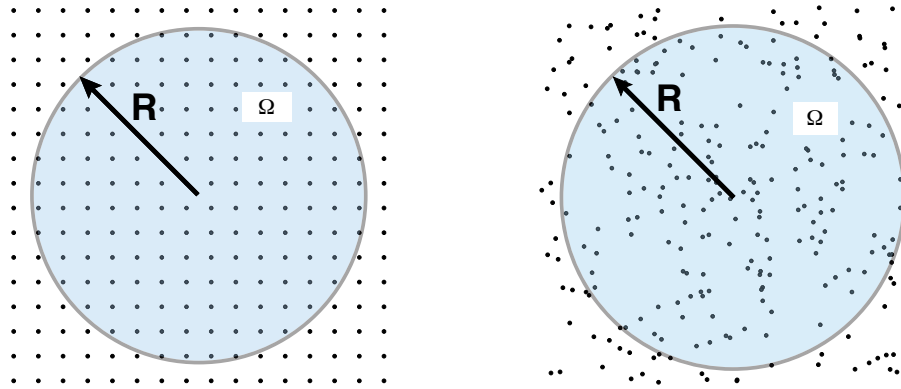
- Typically, ground states are **periodic with high crystallographic symmetries**.
- Can classical ground states ever be **disordered**?

Hyperuniform Point Processes

Torquato & Stillinger, PRE (2003)

Zachary & Torquato, J. Stat. Mech.: Th. & Exp. (2009)

- Points can represent molecules of a material, stars in a galaxy, or trees in a forest. Let Ω represent a spherical window of radius R in d -dimensional Euclidean space $\mathbb{R}^d$.



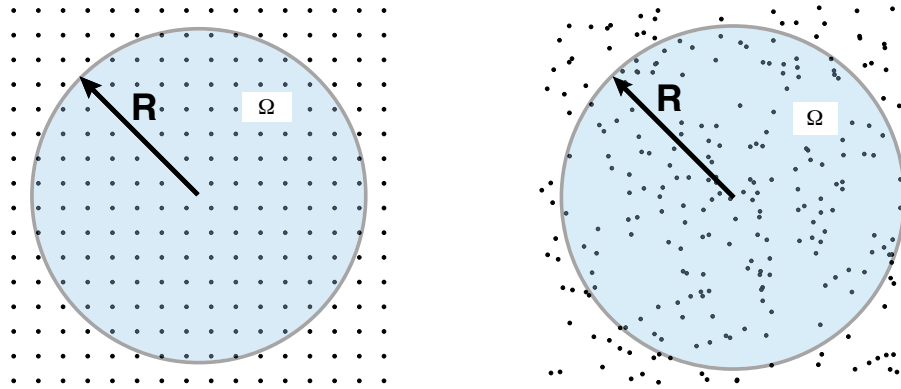
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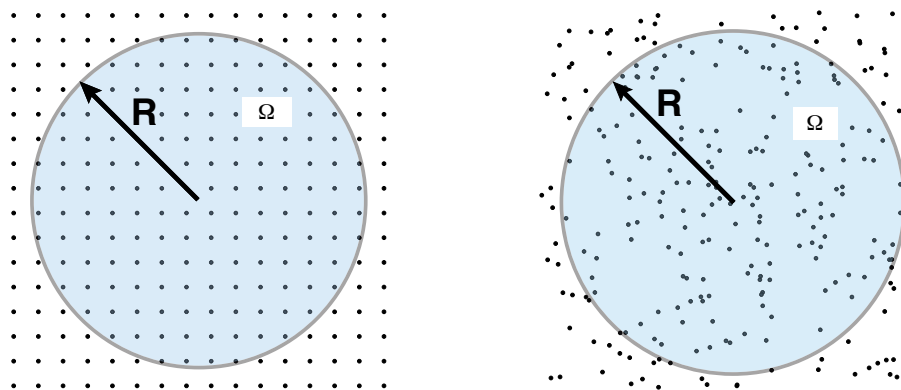
- For a **Poisson** point pattern and many **disordered** point patterns, $\sigma^2(R) \sim R^d$.

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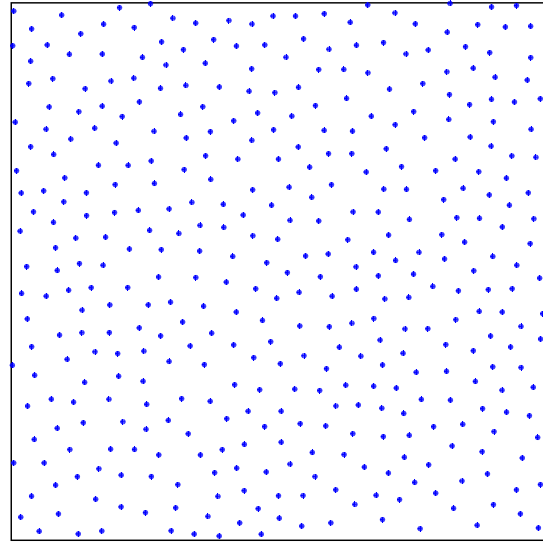
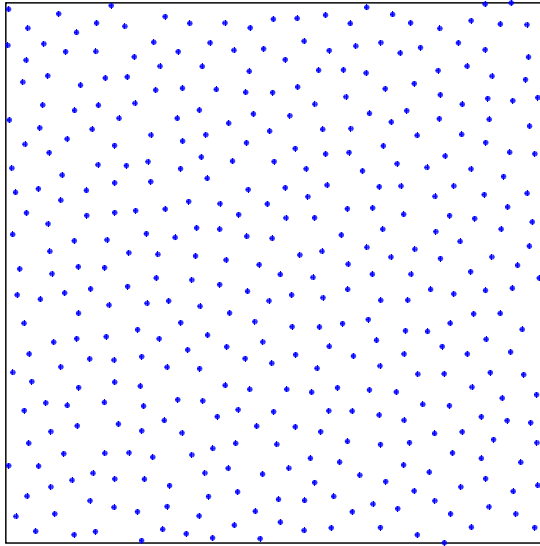
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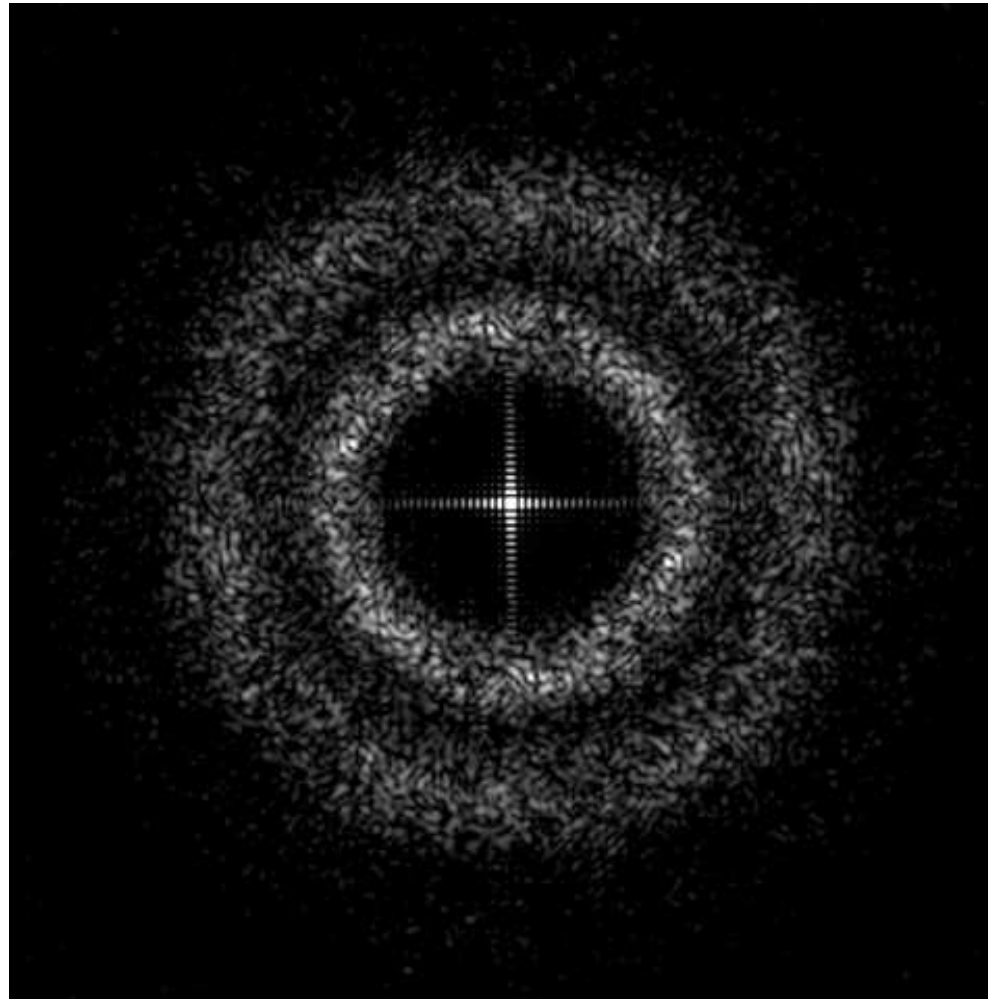
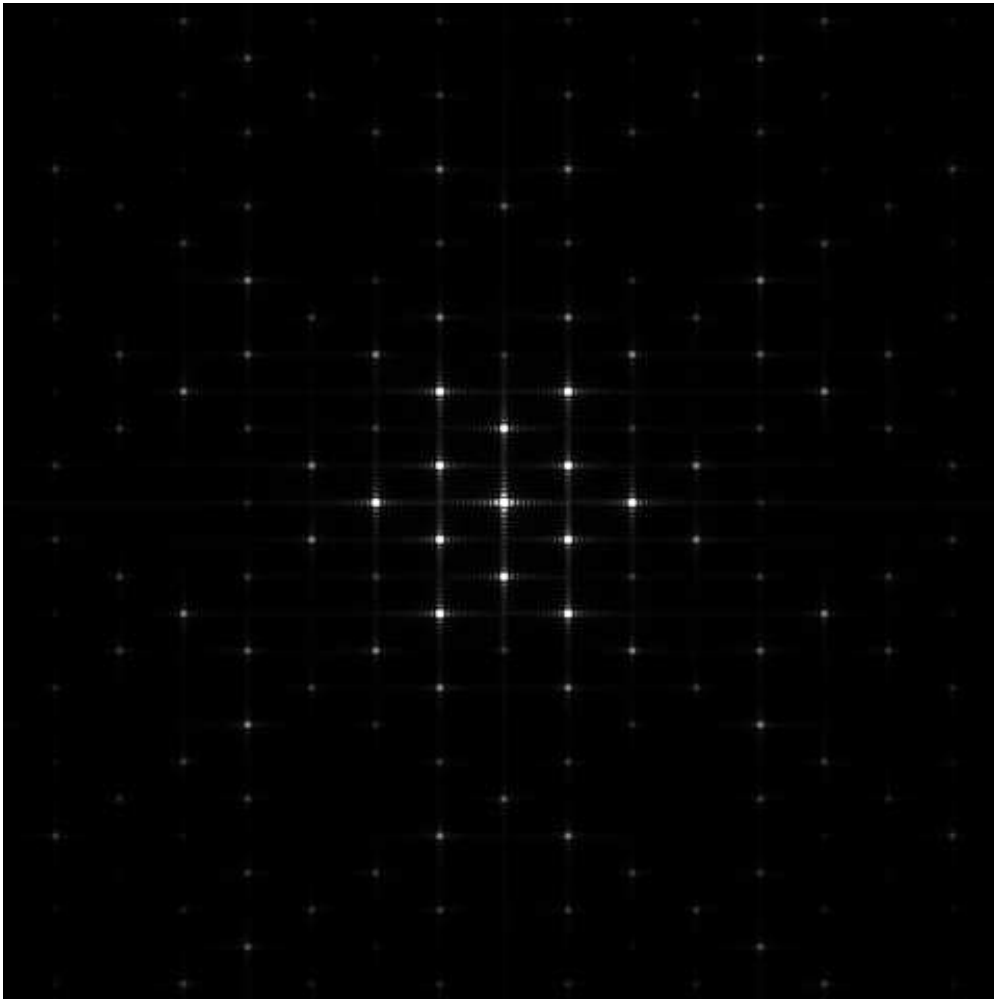
- For a **Poisson** point pattern and many **disordered** point patterns, $\sigma^2(R) \sim R^d$.
- We call point patterns whose variance grows more slowly than R^d (window volume) **hyperuniform**, implying that **structure factor** $S(k) \rightarrow 0$ for $k \rightarrow 0$.
- All **perfect crystals and quasicrystals** are hyperuniform such that $\sigma^2(R) \sim R^{d-1}$: number variance grows like **window surface area**, but the degree to which they suppress large-scale density fluctuations varies and is quantifiable.

Hidden Order on Large Length Scales



Which is the hyperuniform pattern?

Hyperuniform Diffraction/Scattering Patterns



DISORDERED HYPERUNIFORM STATES OF MATTER

● **Disordered hyperuniform** many-particle systems can be regarded to be **new distinguishable states of disordered matter** in that they

(i) *behave more like perfect **crystals or quasicrystals** in the manner in which they suppress **large-scale density fluctuations**, and yet are also like **liquids and glasses** because they are statistically isotropic structures with no Bragg peaks;*

(ii) *can exist as both as **equilibrium** and **quenched nonequilibrium** phases;*

(iii) *and, appear to be endowed with **unique bulk physical properties**.*

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- The **hyperuniformity** concept provides a **unified means of categorizing and characterizing** crystals, quasicrystals and such **special disordered** systems according to the degree to which they **suppress large-scale density fluctuations**.

Creation of Disordered Hyperuniform Ground States

Fan, Percus, Stillinger & Stillinger, Phys. Rev. A, 1991

Uche, Stillinger & Torquato, Phys. Rev. E, 2004

Batten, Stillinger & Torquato, Phys. Rev. E, 2008

Collective-Coordinate Simulations

- Consider a system of N particles with configuration $\mathbf{r}^N \equiv \mathbf{r}_1, \mathbf{r}_2, \dots$ in a **fundamental region** Ω (under periodic boundary conditions). The **complex collective density variable** is defined by

$$\rho(\mathbf{k}) = \sum_{j=1}^N \exp(i\mathbf{k} \cdot \mathbf{r}_j)$$

- The **real nonnegative structure factor** is

$$S(\mathbf{k}) = \frac{|\rho(\mathbf{k})|^2}{N}$$

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- Consider stable **bounded** pair potentials $v(\mathbf{r})$ in $\mathbb{R}^d$ with Fourier transform $\tilde{v}(\mathbf{k})$. The **total energy** is

$$\begin{aligned} \Phi_N(\mathbf{r}^N) &= \sum_{i < j} v(\mathbf{r}_{ij}) \\ &= \frac{N}{2|\Omega|} \sum_{\mathbf{k}} \tilde{v}(\mathbf{k}) S(\mathbf{k}) + \text{constant} \end{aligned}$$

- When $\tilde{v}(\mathbf{k})$ **positive** $\forall 0 \leq |\mathbf{k}| \leq K$ and zero otherwise, finding configurations in which $S(\mathbf{k})$ is **constrained** to be zero where $\tilde{v}(\mathbf{k})$ has support is equivalent to globally **minimizing** $\Phi(\mathbf{r}^N)$. These **hyperuniform** ground states are called “**stealthy**” and generally **highly degenerate**.

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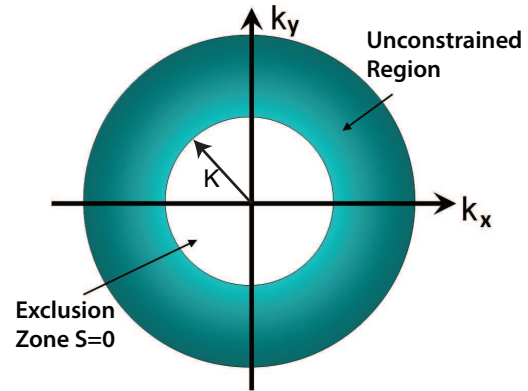
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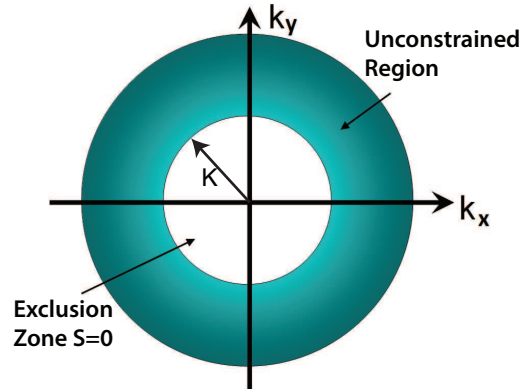
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- Stealthy patterns can be **tuned** by varying the parameter χ : **number of independently constrained degrees of freedom to the total number of degrees of freedom** $d(N - 1)$.

Creation of Disordered Stealthy Ground States



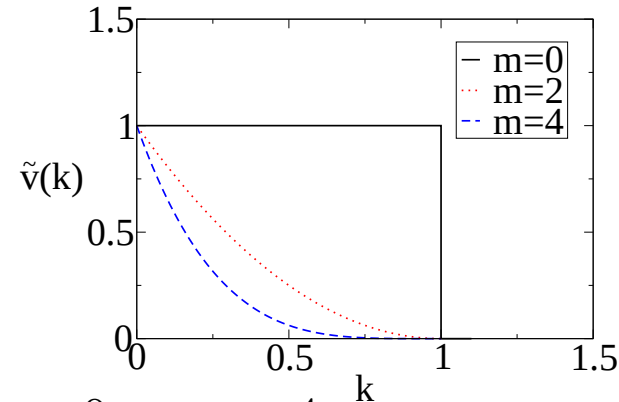
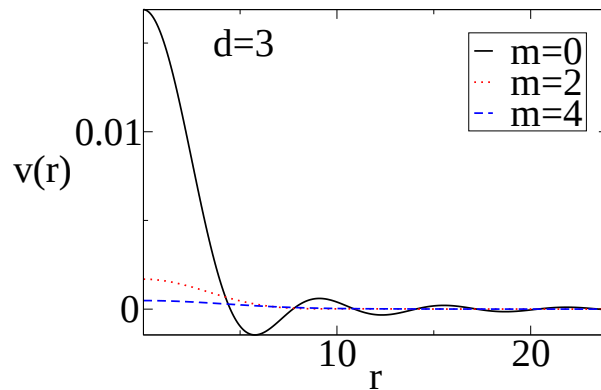
Creation of Disordered Stealthy Ground States



One class of stealthy potentials involves the following **power-law** form:

$$\tilde{v}(k) = v_0(1 - k/K)^m \Theta(K - k),$$

where n is any whole number. The special case $n = 0$ is just the simple step function.

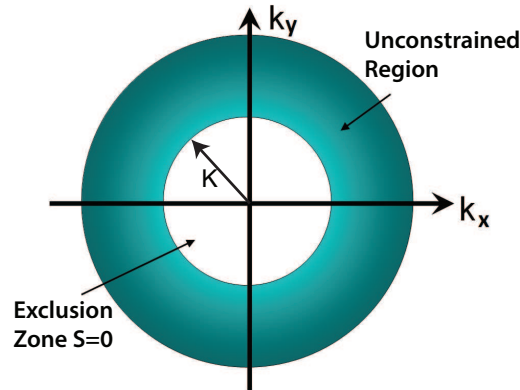


In the large-system (**thermodynamic**) limit with $m = 0$ and $m = 4$, we have the following **large- r asymptotic behavior**, respectively:

$$v(r) \sim \frac{\cos(r)}{r^2} \quad (m = 0)$$

$$v(r) \sim \frac{1}{r^4} \quad (m = 4)$$

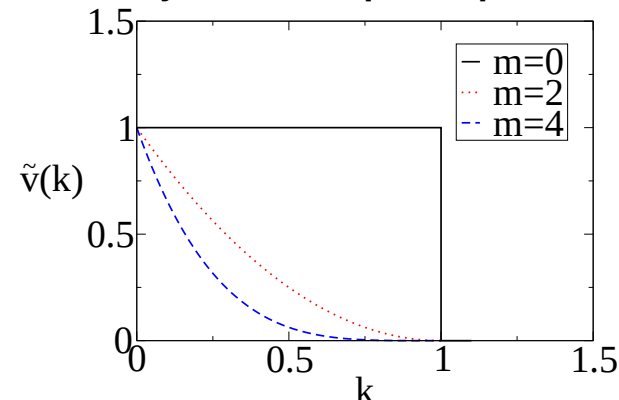
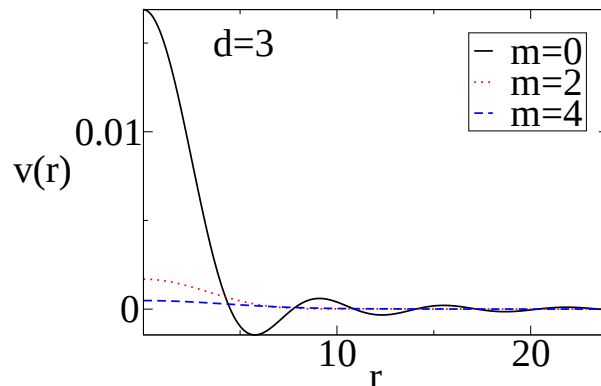
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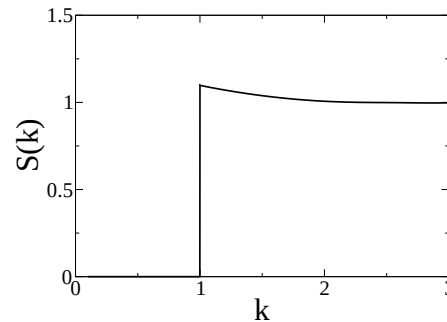
While the specific forms of these stealthy potentials lead to the same convergent ground-state energies, this may not be the case for the **pressure** and other thermodynamic quantities.

Creation of Disordered Stealthy Ground States

- In our **previous simulations**, we began with an initial **random distribution** of N points and then found the **energy minimizing configurations** (with extremely high precision) using optimization techniques.

Creation of Disordered Stealthy Ground States

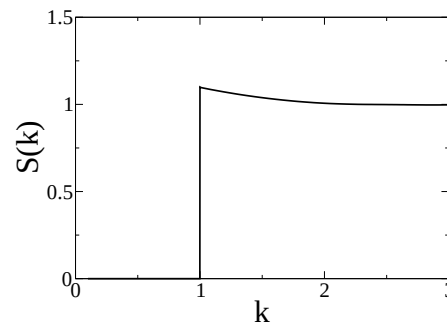
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- The **success rate** to achieve any disordered ground state is 100%, independent of the dimension. It turns out that $\chi = 1/2$ is the limit at which there are no longer degrees of freedom that can be constrained independently.

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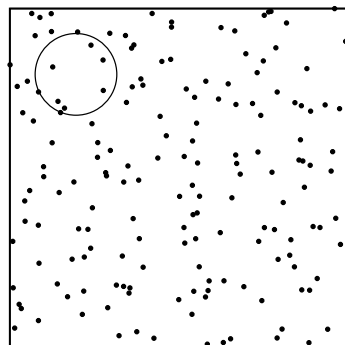


- The **success rate** to achieve any disordered ground state is 100%, independent of the dimension. It turns out that $\chi = 1/2$ is the limit at which there are no longer degrees of freedom that can be constrained independently.
- These **exotic** disordered ground states possess a **hidden order** on large length scales.

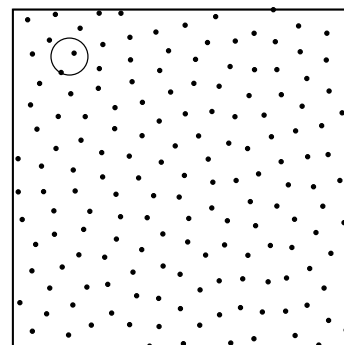
Creation of Disordered Stealthy Ground States



Two Dimensions



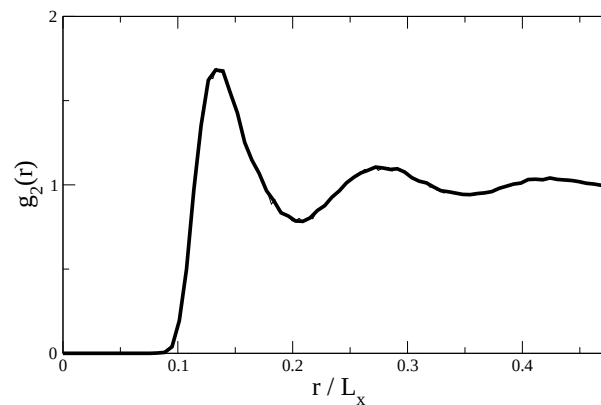
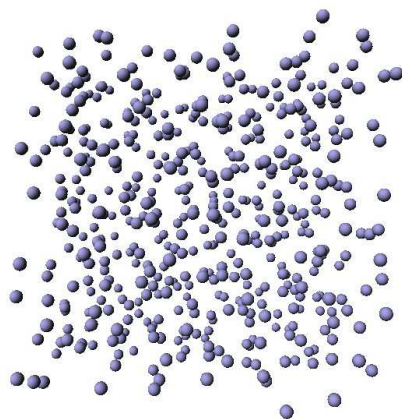
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(b) $\chi = 0.41071$

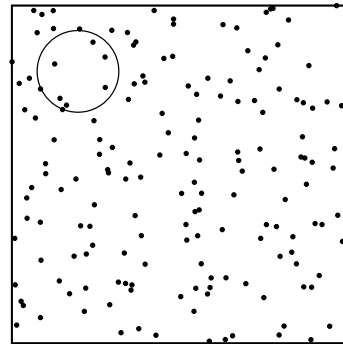


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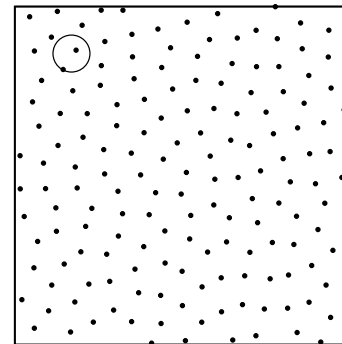


Creation of Disordered Stealthy Ground States

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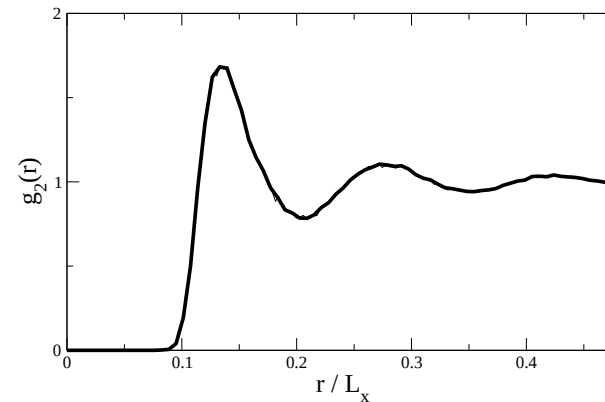
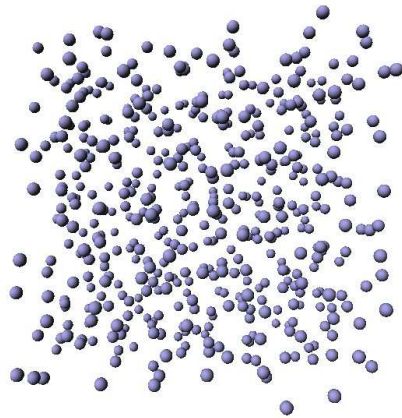


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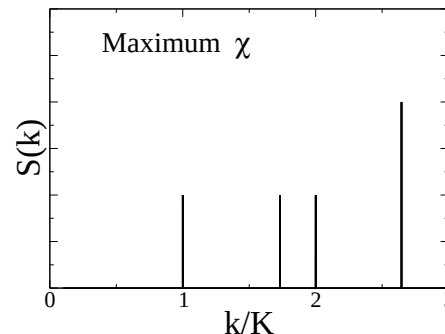


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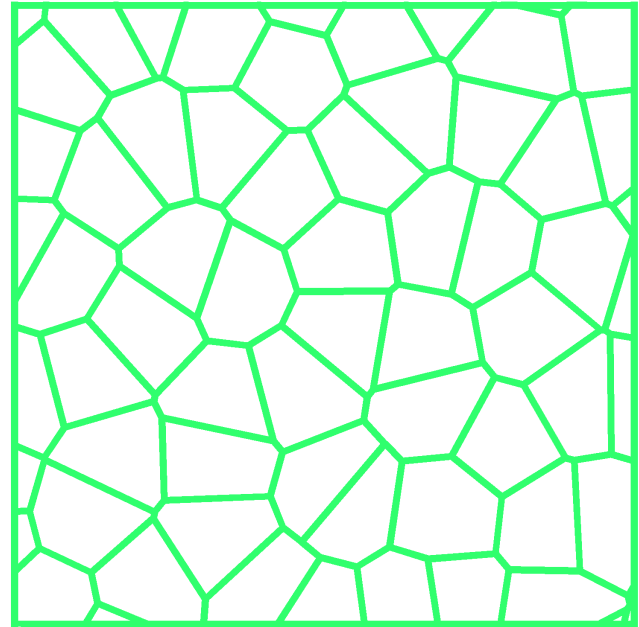
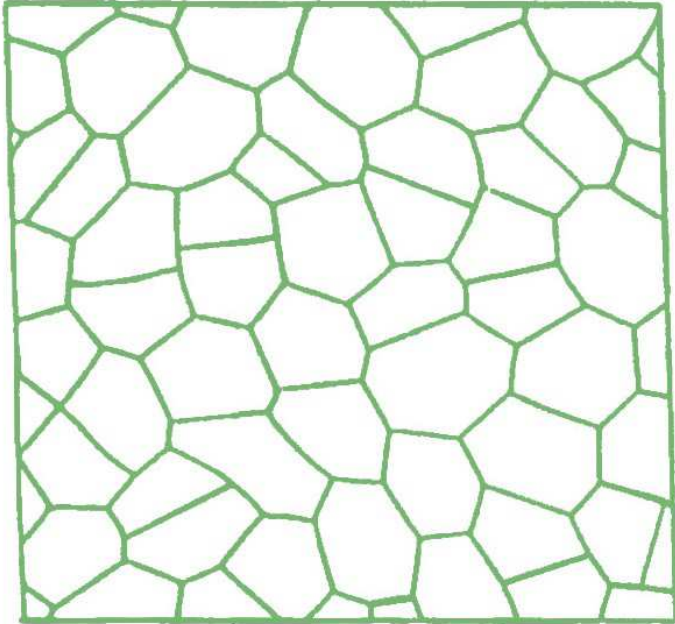
Three Dimensions



Increasing χ induces **strong short-range ordering**, and eventually the system **runs out of degrees of freedom** that can be constrained and **crystallizes**.



Which is the Cucumber Image?



Ensemble Theory of Disordered Ground States

Torquato, Zhang & Stillinger, Phys. Rev. X, 2015

● **Nontrivial:** Dimensionality of the configuration space depends on the number density ρ (or χ) and there is a multitude of ways of sampling the ground-state manifold, each with its own probability measure **Which ensemble?** How are **entropically favored** states determined?

● For **some ensemble** at fixed density ρ , the average energy per particle u for radial potentials in the thermodynamic limit is given by

$$\begin{aligned} u \equiv \left\langle \frac{\Phi(\mathbf{r}^N)}{N} \right\rangle &= \frac{\rho}{2} \int_{\mathbb{R}^d} v(r) g_2(r) d\mathbf{r} \\ &= \frac{\rho}{2} \tilde{v}(k=0) - \frac{1}{2} v(r=0) + \frac{1}{2(2\pi)^d} \int_{\mathbb{R}^d} \tilde{v}(k) S(k) d\mathbf{k}. \end{aligned}$$

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● Consider the same class of **“stealthy”** radial potential functions $\tilde{v}(k)$ in $\mathbb{R}^d$. Whenever particle configurations in $\mathbb{R}^d$ exist such that $S(k)$ is constrained to be its **minimum value of zero** where $\tilde{v}(k)$ has support, the system must be at its **ground state** or **global energy minimum**:

$$u = \frac{\rho}{2} v_0 - \frac{1}{2} v(r=0)$$

Remark: Ground-state manifold is generally **highly degenerate**.

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- In the thermodynamic limit, parameter χ is related to the number density ρ in any dimension d via

$$\rho \chi = \frac{V_1(K)}{2d(2\pi)^d},$$

where $V_1(K)$ is the volume of a d -dimensional sphere of radius K .

Remarks: We see that χ and ρ are **inversely proportional to one another**. Thus, for fixed K and d , as χ tends to zero, ρ tends to infinity, which corresponds counterintuitively to the **uncorrelated ideal-gas limit** (Poisson distribution). As χ increases from zero, the density ρ decreases.

Ensemble Theory of Disordered Ground States

- **Any periodic crystal** with a finite basis is a stealthy ground state for all positive χ up to a **maximum** $\chi_{max} (\rho_{min})$ determined by its **first positive Bragg peak** (minimal vector in Fourier space).

 **Lemma:** At fixed K , a configuration comprised of the union of m different stealthy ground-state configurations in $\mathbb{R}^d$ with $\chi_1, \chi_2, \dots, \chi_m$, respectively, is itself stealthy with a value χ value given by

$$\chi = \left[\sum_{i=1}^m \chi_i^{-1} \right]^{-1},$$

which is the harmonic mean of the χ_i divided by m .

- These last two facts can be used to show how **disordered patterns** are possible ground states.

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Table 1: Periodic stealthy ground states in $\mathbb{R}^2$ with $K = 1$.

Structure	χ_{max}	ρ_{min}
Kagomé crystal	$\frac{\pi}{3\sqrt{12}} = 0.3022 \dots$	$\frac{3\sqrt{3}}{8\pi^2} = 0.06581 \dots$
Honeycomb crystal	$\frac{\pi}{2\sqrt{12}} = 0.4534 \dots$	$\frac{\sqrt{3}}{4\pi^2} = 0.04387 \dots$
Square lattice	$\frac{\pi}{4} = 0.7853 \dots$	$\frac{1}{4\pi^2} = 0.02533 \dots$
Triangular lattice	$\frac{\pi}{\sqrt{12}} = 0.9068 \dots$	$\frac{\sqrt{3}}{8\pi^2} = 0.02193 \dots$

Canonical Ensemble Theory of Disordered Ground States



We consider the Gibbs **canonical ensemble** in which the partition function Z is a function of ρ and absolute temperature T . Our main interest is in a theory in the limit $T \rightarrow 0$, i.e., the **entropically favored ground states in the canonical ensemble**.

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Ground-State Pressure and Isothermal Compressibility

- **Energy Route:** The pressure in the thermodynamic limit at $T = 0$ can be obtained from the energy per particle (taking $v_0 = 1$) via the relation

$$p = \rho^2 \left(\frac{\partial u}{\partial \rho} \right)_T .$$

Therefore, for stealthy potentials,

$$p = \frac{\rho^2}{2} .$$

- The isothermal compressibility $\kappa_T \equiv \rho^{-1} \left(\frac{\partial \rho}{\partial p} \right)_T$ of such a system is

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$$\kappa_T = \frac{1}{\rho^2}.$$

- **Virial Route:** An alternative route to the pressure is through the “virial” equation, which at $T = 0$ is given by

$$\begin{aligned} p &= -\frac{\rho^2}{2d} s_1(1) \int_0^\infty r^d \frac{dv}{dr} g_2(r) dr \\ &= -\frac{\rho^2}{2d} \left[\tilde{f}(k=0) + \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{f}(k) \tilde{h}(k) d\mathbf{k} \right] \end{aligned}$$

where $\tilde{f}(k)$ is the Fourier transform of $f(r) \equiv r dv/dr$, **when it exists**.

Pair Statistics of Disordered Ground States

- In the ground state of a stealthy potential, the structure factor attains its minimum value $S(k) = 0$ for $0 < k \leq K$, and hence has the general form

$$S(k) = \Theta(k - K)[1 + \tilde{Q}(k)],$$

where

$$\Theta(x) = \begin{cases} 0, & x < 0, \\ 1, & x \geq 0 \end{cases}.$$

- Therefore,

$$\rho \tilde{h}(k) = \tilde{f}(k) + \tilde{P}(k),$$

where

$$\tilde{f}(k) = -\Theta(K - k)$$

and

$$\tilde{P}(k) = \Theta(k - K)\tilde{Q}(k).$$

Remark: The function $\tilde{f}(k)$ is identical to the “Mayer- f ” function for a Gibbs-ensemble hard-sphere system in direct space.

- It easily shown that the functions $v(r)$ and $P(r)$ are orthogonal to one another:

$$\int_{\mathbb{R}^d} v(r)P(r)d\mathbf{r} = 0.$$

Pair Statistics of Disordered Ground States

Behavior of the Pair Correlation Function Near the Origin

- Using the general form for $S(k)$ for stealthy ground states and elementary properties of Fourier transforms, it is easy to show pair correlation function at the **origin** is given by

$$g_2(r=0) = 1 - 2d\chi + 2d^2\chi \int_K^\infty k^{d-1} \tilde{Q}(k) dk$$

- Since $g_2(r)$ must be nonnegative for all r , we have the following integral condition on $\tilde{Q}(k)$:

$$2d^2\chi \int_K^\infty k^{d-1} \tilde{Q}(k) dk \geq 2d\chi - 1$$

Hence, this integral must be positive for $\chi \geq \frac{1}{2d}$.

- Moreover, if $g_2(r)$ has **positive curvature** at the origin, $\tilde{Q}(k)$ must obey the additional integral condition:

$$(d+2) \int_K^\infty k^{d+1} \tilde{Q}(k) dk \leq 1.$$

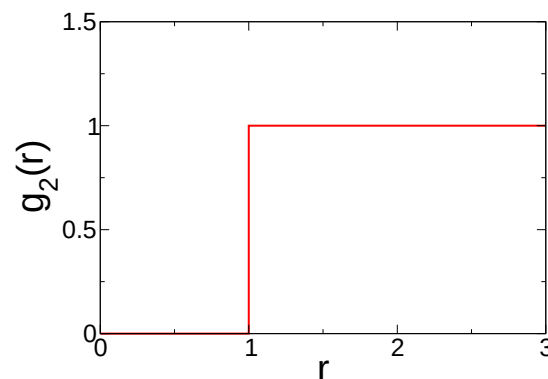
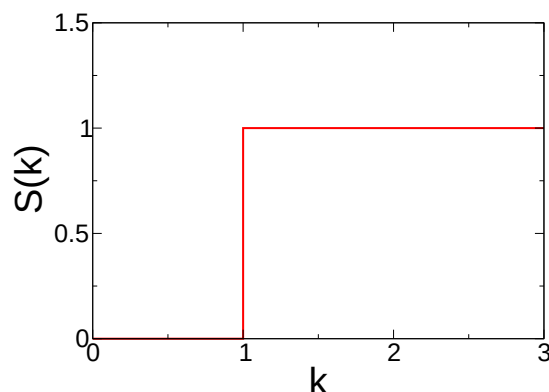
- Finally, when $g_2(r=0) = 0$, the results above yield the equality

$$\int_K^\infty k^{d-1} \tilde{Q}(k) dk = \frac{2d\chi - 1}{2d^2\chi}$$

and, because the curvature must be positive in this instance, the previous inequality must generally be obeyed.

Pseudo-Hard Spheres in Fourier Space

- From previous considerations, we see that that an important contribution to $S(k)$ is a simple hard-core step function $\Theta(k - K)$, which can be viewed as a **disordered hard-sphere system in Fourier space** in the limit that χ tends to zero, i.e., as the number density ρ tends to infinity.



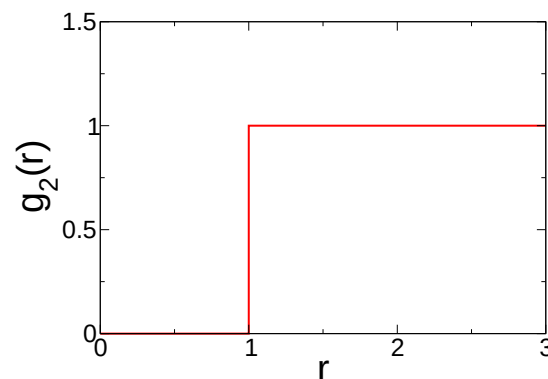
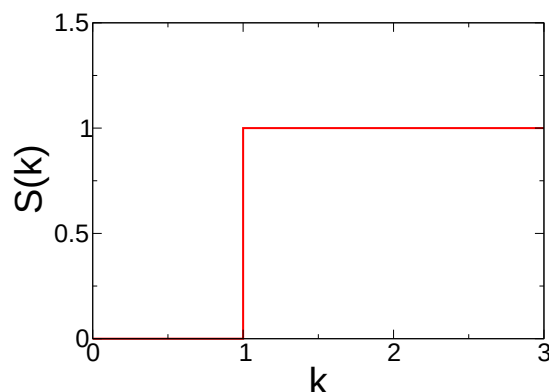
- That the structure factor must have the behavior

$$S(k) \rightarrow \Theta(k - K), \quad \chi \rightarrow 0$$

is perfectly reasonable; it is a **perturbation about the ideal-gas limit in which $S(k) = 1$ for all k .**

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- Imagine carrying out a series expansion of $S(k)$ about $\chi = 0$. We make the **ansatz** that for sufficiently small χ , $S(k)$ in the canonical ensemble for a stealthy potential can be mapped to $g_2(r)$ for an **effective disordered hard-sphere system for sufficiently small density.**

Pseudo-Hard Spheres in Fourier Space

- Let us define

$$\tilde{H}(k) \equiv \rho \tilde{h}(k) = h_{HS}(r = k)$$

There is an **Ornstein-Zernike integral** equation that defines the appropriate direct correlation function $\tilde{C}(k)$:

$$\tilde{H}(k) = \tilde{C}(k) + \eta \tilde{H}(k) \otimes \tilde{C}(k),$$

where $\eta = b\chi$ is an **effective packing fraction**, where b is a constant to be determined. Therefore,

$$H(r) = \frac{C(r)}{1 - (2\pi)^d \eta C(r)}.$$

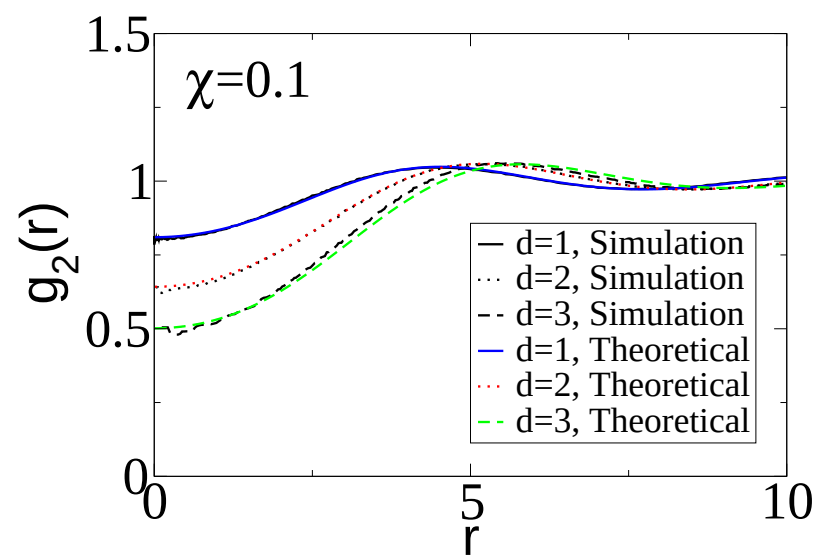
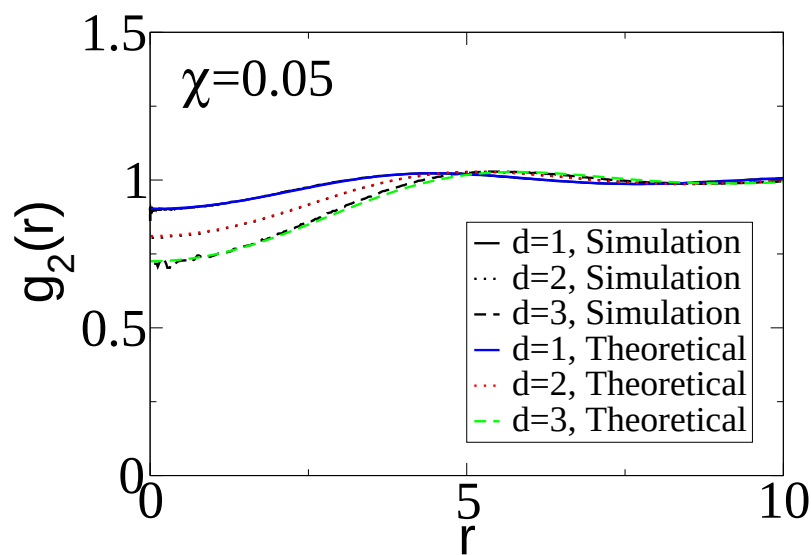
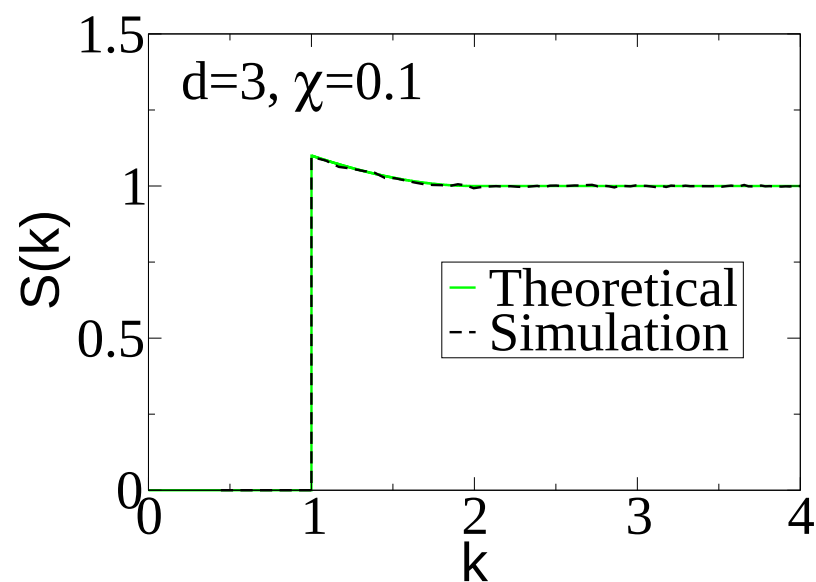
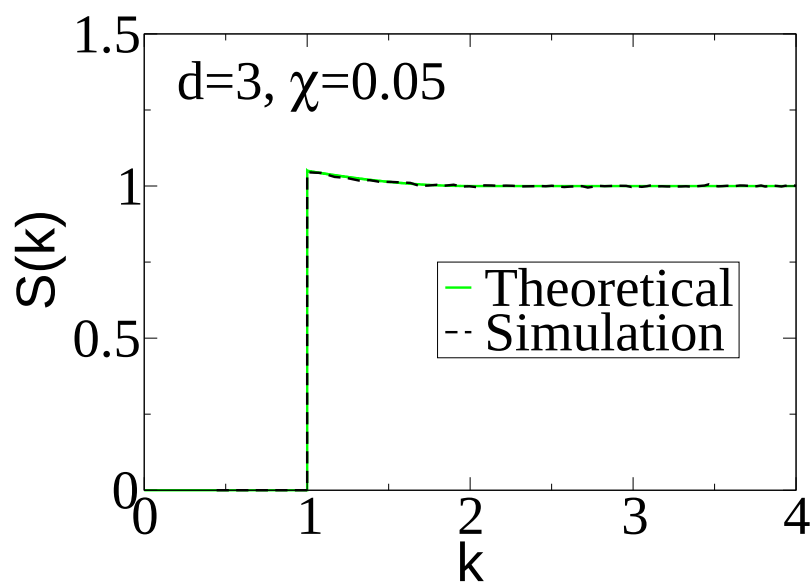
This mapping enables us to exploit the well-developed accurate theories of **standard Gibbsian disordered hard spheres in direct space**.

- To get an idea of the **large- r asymptotics**, consider case $\chi \rightarrow 0$ for any d :

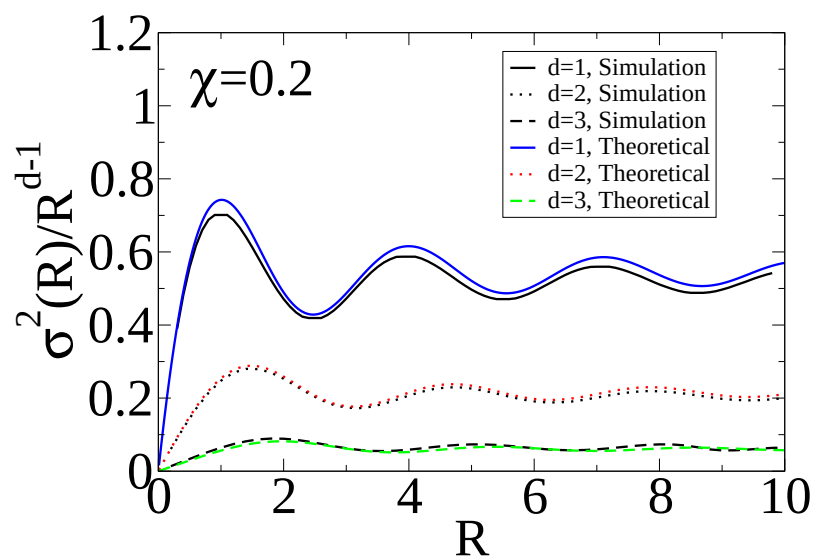
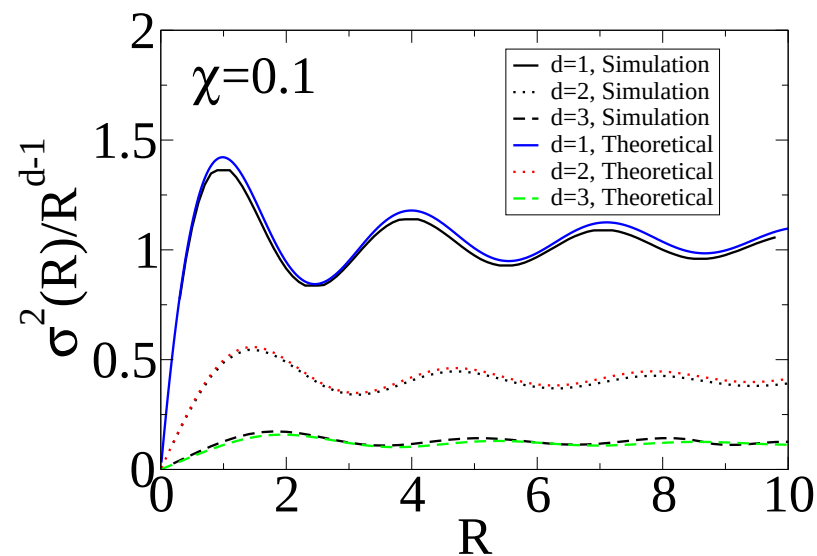
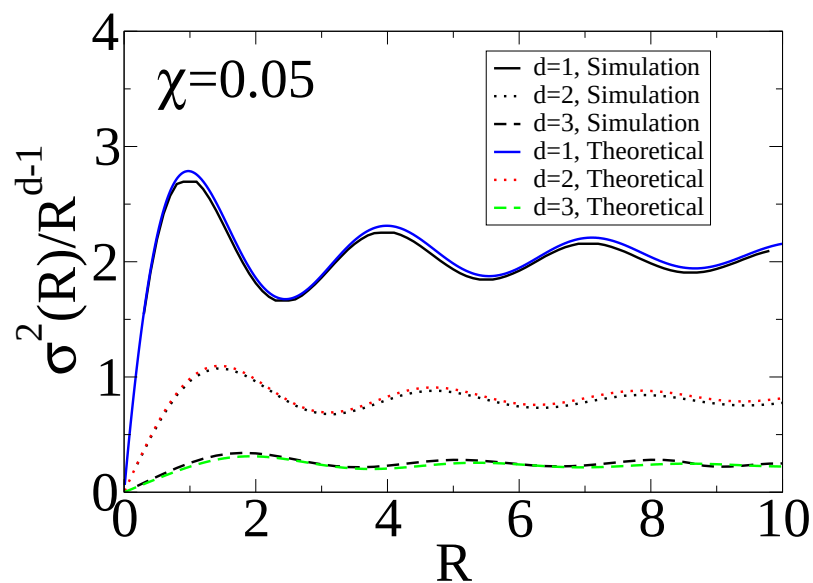
$$\rho h(r) \sim -\frac{1}{r^{d/2}} J_{d/2}(r) \quad (\chi \rightarrow 0)$$

$$\rho h(r) \sim -\frac{1}{r^{(d+1)/2}} \cos(r - (d+1)\pi/4) \quad (r \rightarrow +\infty)$$

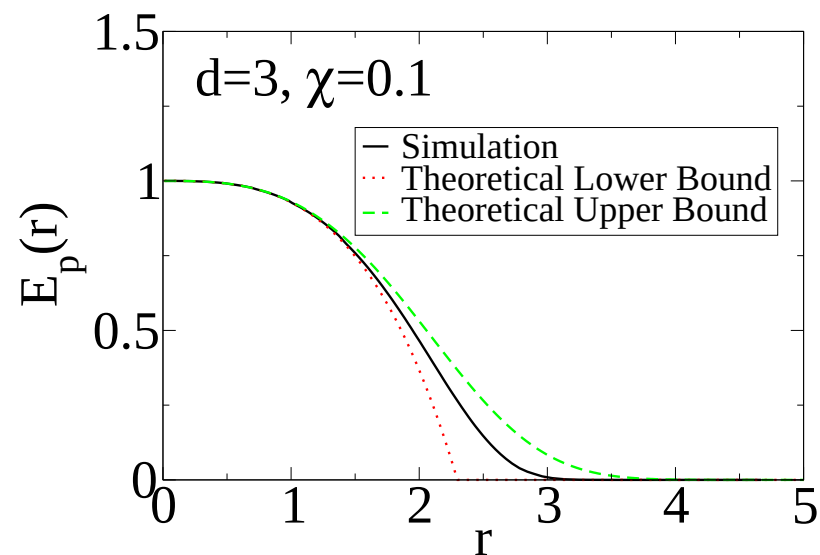
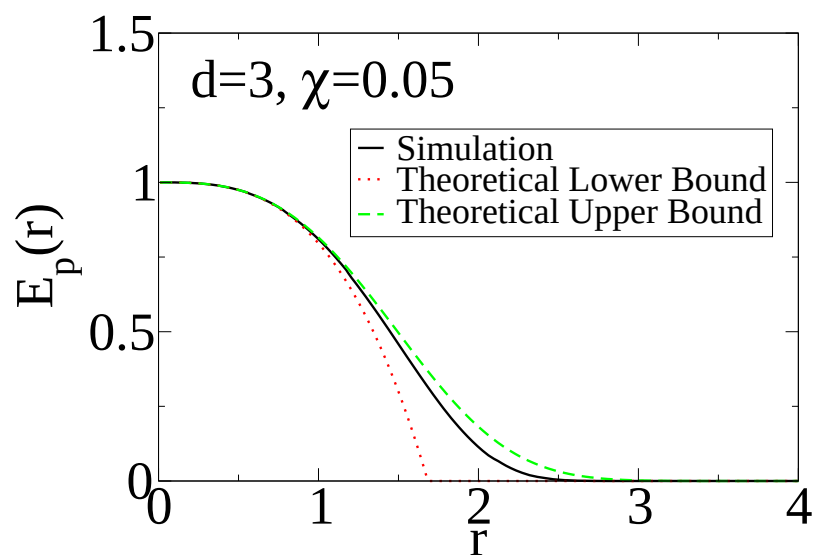
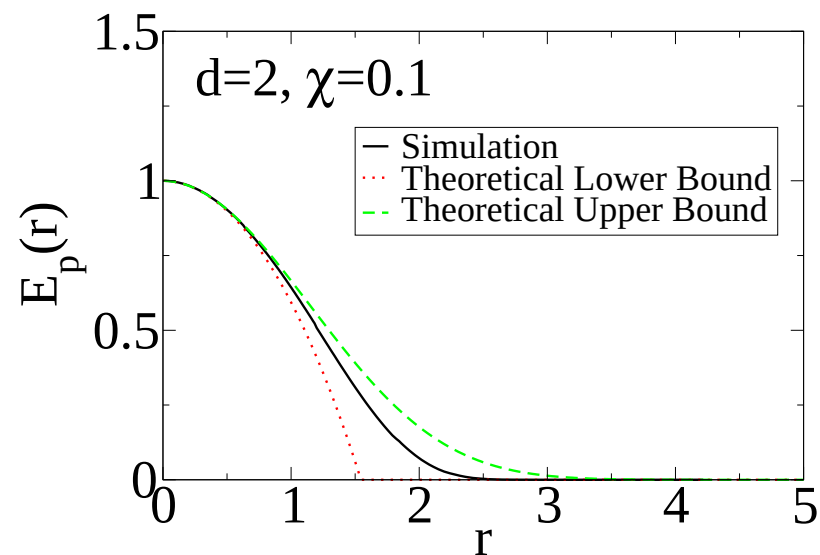
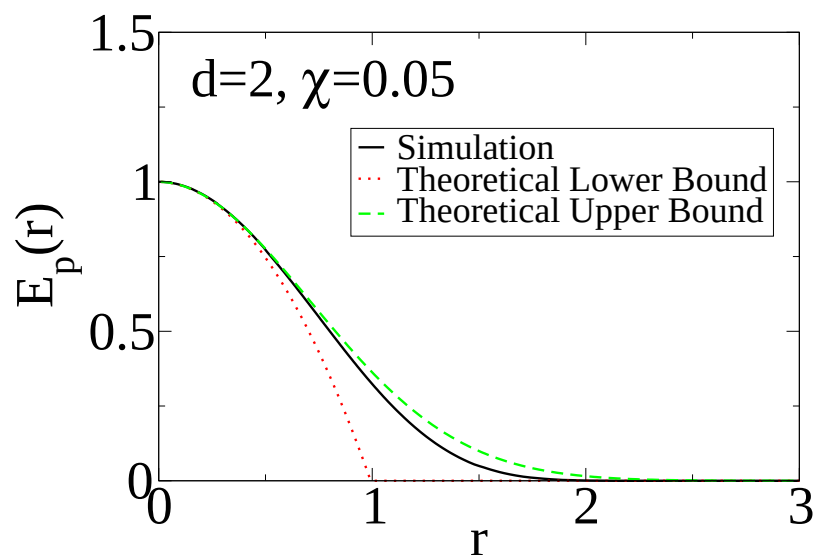
Pair Statistics: Theory vs. Simulations



Number Variance $\sigma^2(R)$: Theory vs. Simulations



Nearest-Neighbor Function: Theory vs. Simulations



Remark About Excited States

- For a system in **equilibrium**, the **compressibility relation** relates the isothermal compressibility κ_T to the structure factor at $k = 0$:

$$S(0) = \rho k_B T \kappa_T,$$

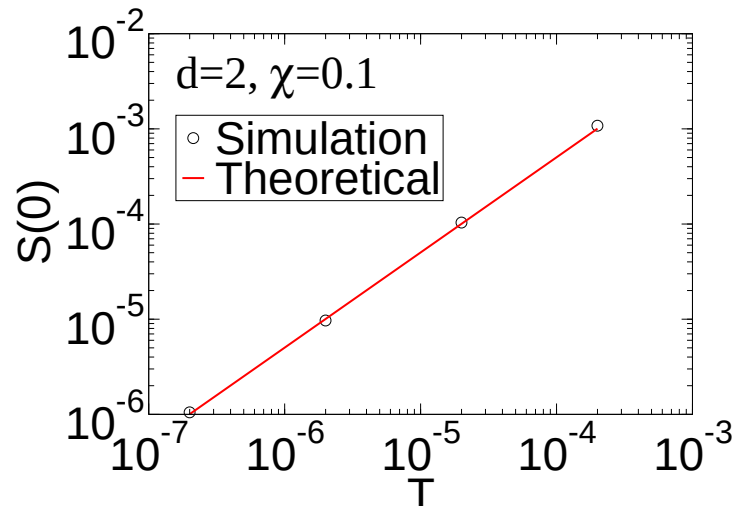
where k_B is Boltzmann's constant. Because κ_T is bounded according to $\kappa_T = \rho^{-2}$ for finite ρ , $S(0) = 0$ because $T = 0$, which clearly must be the case for stealthy ground states.

- Now consider excited states infinitesimally close to the stealthy ground states, i.e., when temperature T is positive and infinitesimally small.
- Under the assumption that the structure of such excited states will be **infinitesimally near the ground-state configurations**, we can estimate to an excellent approximation how $S(0)$ varies with T for such excited states:

$$S(0) \sim c(d) \chi T$$

in units $k_B = v_0 = 1$. Moreover, it is expected that this positive value of $S(0)$ will be the **uniform value of $S(k)$ for $0 \leq k \leq K$** .

- This behavior of $S(k)$ has indeed been verified by **molecular dynamics simulations in the canonical ensemble**:



CONCLUSIONS

- **Hyperuniformity** has connections to **physics** (e.g., ground states, quantum systems, random matrices, novel materials, etc.), **mathematics** (e.g., geometry and number theory), and **biology**.
- Disordered hyperuniform systems are **new distinguishable states of matter** somewhere between a liquid and a perfect crystal, and appear to be endowed with **novel physical properties**
- **Disordered ground states** are necessarily **highly degenerate** and their existence for **radial pair potentials** in low d is surprising.
- Devising a corresponding **ensemble theory** is highly nontrivial (many ensembles from which to choose) and until recently had been lacking.
- We have derived **exact general** results that apply to **any** ensemble of **stealthy ground states**, disordered or not.
- For the specific case of the **canonical ensemble**, our theory enables us to obtain predictive analytical results for the **structure and thermodynamics** of the **entropically** favored disordered ground states.
- Extensions of these results to **compact spaces** would be very interesting.