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Percolation in Hard-Core Lattice Gases and a Model Ferrofluid

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A lattice gas on $\mathbb{Z}^3$ consisting of hard spheres with exclusions extending through third neighbors is proved to undergo a percolation transition. If spins with ferromagnetic couplings are attached to the spheres, spontaneous magnetization is proved to occur. This may provide a model for a "ferrofluid," a system which exhibits spontaneous magnetization without crystalline order. Similar results are also obtained for an analogous model on $\mathbb{Z}^2$.

KEY WORDS: Lattice gas; percolation; ferrofluid.

1. INTRODUCTION

Order-disorder phase transitions have been proved to exist in several hard core lattice gases. A proof of the presence or absence of a phase transition in a lattice gas of hard disks on $\mathbb{Z}^2$ with first- and second-neighbor exclusions has, however, never been constructed. Configurations in this model at closest packing consist of every second row of sites being vacant, the other rows of sites containing disks. There are a large number of such configurations, for the rows of disks can "slide" with respect to one another. Consequently, this model has been difficult to treat by the Peierls argument.[1] Numerical calculations indicate the possibility of an order–disorder transition at very large activities, but the evidence is far from conclusive on this point.[2–5]

A three-dimensional analog of the above model consists of a lattice gas of hard spheres on $\mathbb{Z}^3$ with first-, second-, and third-neighbor exclusions. There are many configurations at closest packing in this model as well, for

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in addition to the sliding of alternate rows of occupied sites within a plane, there is also the possibility of sliding alternate planes of occupied spheres.

In Section 2 we study this hard sphere model on $\mathbb{Z}^3$ from the point of view of percolation.[6,7] In particular, we use reflection positivity[1] to prove that infinite connected sets of occupied unit cubes exist in the system with probability one at sufficiently large activity and with probability zero at sufficiently small activity. This proves the existence of a percolation transition in the model.

We then consider a weaker notion of connectivity.[8,9] In this regard, we define an "aggregate" to be a set D in which every proper subset $D_0 \subset D$ has the property that dist $(D_0, D \backslash D_0) \leqslant d(\text{diam } D_0)^\delta$, where d is some distance and $\delta < 2$. We will show that at sufficiently low (high) activity, the probability a given cube in the model belongs to an aggregate of occupied (vacant) cubes decreases exponentially as the size of the aggregate increases.

The percolation ideas we use for the model on $\mathbb{Z}^3$ are also applicable for the analogous model on $\mathbb{Z}^2$. We give the corresponding results for the two-dimensional model at the end of Section 2.

Infinite clusters of sites or cubes have been used in other models to prove the existence of ordered phases at sufficiently low temperatures.[10–12] This has been a particularly useful technique for cases in which the ground state configurations are highly degenerate. We hasten to point out that we prove neither the presence nor the absence of an order–disorder transition in the present model.

Since the present model may well be disordered at all activities, it provides a lattice gas model for a "ferrofluid," a system which possesses spontaneous magnetization but not crystalline order. In Section 3 we attach ferromagnetic spins to the spheres. We also introduce sphere–sphere attractions to provide for more realistic phase behavior. Using a "combination" of reflection positivity[1,13] and the standard Peierls argument,[14–16] we prove that spontaneous magnetization occurs in the model at sufficiently low temperatures. The corresponding results for the analogous two-dimensional version of the model are given at the end of Section 3.

2. PERCOLATION IN A LATTICE GAS OF HARD SPHERES

Consider a simple cubic lattice Λ with cyclic boundaries given as $\Lambda = \{(a, b, c)\colon a, b, c = 0, 1,..., 2M-1\}$. The coordinates (x, y, z) are computed modulo $2M$ onto $0 \leqslant x, y, z < 2M$.

We consider a lattice gas of hard spheres on Λ with exclusions extending through third neighbors. We let $S_i = 0$ correspond to a vacancy at site $i \in \Lambda$, and $S_i = 1$ correspond to site i being occupied by a sphere. A configuration is given as $\xi \in \{0, 1\}^{|\Lambda|}$. The Hamiltonian for a configuration ξ can be written as

$$H(\xi) = -\mu \sum_{i \in \Lambda} S_i \tag{2.1}$$

We introduce a characteristic function on allowed configurations as

$$\chi(\xi) = \prod_{(ij)} (1 - S_i S_j) \tag{2.2}$$

where the product is over all first-, second-, and third-neighbor pairs of sites. If C is the set of allowed configurations on Λ, then the grand canonical partition function for the system can be written as

$$\Xi = \sum_{\xi} \chi(\xi)\, e^{-H(\xi)/kT} = \sum_{\xi \in C} e^{-H(\xi)/kT} \tag{2.3}$$

In an allowed configuration, at most one vertex of any unit cube in Λ can be occupied by a sphere.

As is shown in Appendix A, reflection positivity[1] is satisfied for reflections through planes of lattice sites which are perpendicular to one of the coordinate axes. A pair of such planes, $P^{\pm}$, divides Λ into two equal parts, Λ^+ and Λ^-, plus $\Lambda^0 = (P^+ \cup P^-) \cap \Lambda$. If f is any function of the configurations on $\Lambda^+ \cup \Lambda^0$, and θf is the reflection of f through the planes $P^{\pm}$ (θf is thus a function of the configurations on $\Lambda^- \cup \Lambda^0$), then reflection positivity is said to be satisfied if $\langle \bar{f} \theta f \rangle \geqslant 0$. If reflection positivity is satisfied, then it follows by a standard Cauchy–Schwartz argument that $|\langle fg \rangle|^2 \leqslant \langle \bar{f} \theta f \rangle \langle \bar{g} \theta g \rangle$, where f is any function of the configurations on $\Lambda^+ \cup \Lambda^0$, and g is any function of the configurations on $\Lambda^- \cup \Lambda^0$.

This latter inequality is useful for obtaining bounds to several quantities of interest. In particular, let $P_{L,0}(P_{L,v})$ be the probability that a set L of unit cubes in Λ are all occupied by spheres (all vacant). In Appendix B, reflection positivity is used to show that

$$P_{L,0} \leqslant z^{|L|/8}, \qquad P_{L,v} \leqslant z^{-|L|/8} \tag{2.4}$$

where $z = \exp(\mu/kT)$ is the activity. We shall use these bounds to prove the existence of a percolation transition in the model.

We first give a definition of a rather weak type of connectivity.[8,9] (The notions and results between Definition 2.1 and Theorem 2.3 are more general than what will be needed, later, in this paper. However, we feel they are sufficiently interesting in this context to be briefly recalled.)

Definition 2.1. $D \subset \mathbb{Z}^\nu$ is (δ, d)-connected if $\forall D_0 \subset D$,

$$\operatorname{dist}(D_0, D \backslash D_0) \leqslant \begin{cases} d(\operatorname{diam} D_0)^\delta, & \text{if } |D_0| > 1 \\ d, & \text{if } |D_0| = 1 \end{cases}$$

We now proceed to obtain a useful bound on the number of (δ, d)-connected sets, $\delta < 2$, which contain the origin and fewer than m other sites of $\mathbb{Z}^\nu$. The following notion[9] of the volume, $V(D)$, of a set D will prove helpful for obtaining such a bound. If $\mathbb{C}_n(D)$ is a minimal family of cubes which covers D, each cube having edges of length 2^n, then $V(D) \equiv \sum_0^{n_0} |\mathbb{C}_n(D)|$, where n_0 is the smallest integer for which $|\mathbb{C}_{n_0}(D)| = 1$. We also define[9] the volume, $V'(D)$, of "isolated points" as $V' \equiv \sum_1^{n_0} |\mathbb{C}'_n|$, where $\mathbb{C}'_n(\alpha, M) = \{c: c \in \mathbb{C}_n, \text{ dist } (c, c') \geqslant M 2^{\alpha n}\ \forall c' \in \mathbb{C}_n \backslash c\}$. Here $1 \leqslant \alpha < 2$ and $M < \infty$.

Lemma 2.2. If D is (δ, d)-connected and $\delta < 2$, $\exists K(\delta, d) < \infty \ni V(D) \leqslant K(\delta, d)\, V_0(D)$.

Proof. By Theorem 4.4 of Ref. 9, $\forall M > 0, 1 < \alpha < 2$, $V(D) \leqslant K_0(\alpha, M)\, V_0(D) + K'(\alpha, M)\, V'(D)$. Pick any $M > d$ and $2 > \alpha > \max(1, \delta)$. If $V'(D) \neq 0$, then $\exists n \geqslant 0 \ni \mathbb{C}'_n(D) \neq \phi$. $\Rightarrow \exists D_0 \subset D \ni M 2^{\alpha n} \leqslant \operatorname{dist}(D_0, D \backslash D_0) \leqslant d 2^{n\delta}$, a contradiction. ∎

Theorem 2.3. If $\delta < 2$, $\exists K_\nu(\delta, d) < \infty \ni N(\delta, m) = \#\{D: D$ is (δ, d)-connected, $0 \in D, |D| \leqslant m\} \leqslant e^{K_\nu(\delta, d) m}$.

Proof. By Theorem 4.2 of Ref. 9, $\#\{D: D$ is (δ, d)-connected, $0 \in D, V(D) = V\} \leqslant e^{K_\nu V}$. $\Rightarrow \#\{D: D$ is (δ, d) connected, $0 \in D$, $V(D) \leqslant m\} \leqslant C_\nu e^{K_\nu m}$ for some $C_\nu > 0$. From Lemma 2.2, if $|D| \equiv V_0(D) \leqslant m$, then $V(D) \leqslant K(\delta, d) m$ if $\delta < 2$. $\Rightarrow N(\delta, m) \leqslant \#\{D: D$ is (δ, d)-connected, $0 \in D$, $V(D) \leqslant K(\delta, d) m\} \leqslant C_\nu e^{K_\nu K(\delta, d) m}$ if $\delta < 2$. ∎

We show in Appendix C that Theorem 2.3 is false for all $d \geqslant 1$ if $\delta \geqslant 2$. (δ, d)-connected sets, $\delta < 2$, thus provide a useful new notion of connectivity. We now give some physical meaning to such sets.

A "molecule" is a $(0, d)$-connected set of atoms, where d is the longest bond length in the molecule. For $\delta < 2$, we shall call a (δ, d)-connected set an "aggregate." From Theorem 2.3, the logarithm of the number of (δ, d)-connected aggregates, containing the origin and a number of elements not exceeding m, is proportional to m. This logarithm is related to the entropy of the aggregate.

We shall now apply Theorem 2.3 to the hard sphere lattice gas. Consider a unit cube C_r with center at r. Let $\Lambda' = \mathbb{Z}^3$ be the lattice of cube midpoints. Two cubes are said to be connected by an edge if their midpoints are separated by a distance $d \leqslant \sqrt{2}$. A set L of cubes is said to be a "connected cluster" if there exists a walk, having step length $\leqslant \sqrt{2}$, on midpoints of cubes in L which connects the midpoints of any two cubes in L. By Definition 2.1, a connected cluster of cubes is also a $(0, \sqrt{2})$-connected subset of $\mathbb{Z}^3$.

From Theorem 2.3, the number, N_L, of connected clusters, each containing $\leqslant |L|$ cubes (including the cube with center at $0 \in \Lambda'$), is bounded as $N_L \leqslant \exp(|L| \, K_3(0, \sqrt{2})) \equiv K^{|L|/8}$. Then from Eq. 2.4, the probability that the origin belongs to a connected cluster of $|L|$ vacant cubes is less than $(K/z)^{|L|/8}$.

In particular, the probability is zero that the origin belongs to an infinite connected cluster ($|L| \to \infty$) of vacant cubes if $z > K$. By translational invariance, it follows that the probability is zero that an infinite connected cluster of vacant cubes is anywhere in $\mathbb{Z}^3$ if $z > K$.

Similarly, from Theorem 2.3 and Eq. 2.4, the probability is zero that there is an infinite connected cluster of occupied cubes in the system if $z < K^{-1}$.

More precisely, our estimates (2.4) and Theorem 2.3 permit us to show that if $z > K$ the probability that there exists a connected cluster of n vacant cubes containing a given point is bounded above by $e^{-C(z)n}$, for some constant $C(z)$ (tending to $+\infty$ as $z \to \infty$), and if $z < K^{-1}$ the probability that there exists a connected cluster of n occupied cubes containing a given point is bounded above by $e^{-C(z^{-1})n}$. Thus, we see, using a standard Peierls estimate, that there exists a constant $K' \geqslant K$, such that the probability is one that the system contains an infinite cluster of occupied cubes if $z > K'$, and the probability is one that the system contains an infinite cluster of vacant cubes if $z < K'^{-1}$. Therefore, there is a percolation transition of vacant cubes at $z_{c,v} \in (K'^{-1}, K')$ and a percolation transition of occupied cubes at $z_{c,0} \in (K'^{-1}, K')$. Since every configuration is expected to contain either an infinite cluster of vacant cubes or an infinite cluster of occupied cubes, then $z_{c,0} \leqslant z_{c,v}$.

An argument, completely analogous to that given above, can be constructed to prove the existence of a percolation transition in the analogous two-dimensional model, the lattice gas of hard disks on $\mathbb{Z}^2$ with exclusions extending through 2nd neighbors. For this model, there exists a constant $K'' \geqslant \exp(4K_2(0, \sqrt{2})$, such that the probability is zero (one) that the system contains an infinite connected cluster of vacant (occupied) unit squares if $z > K''$. Likewise, the probability is zero (one) that the system contains an infinite connected cluster of occupied (vacant) unit squares if $z < K''^{-1}$.

We can also obtain bounds on the number of finite aggregates of all occupied or all vacant cubes which contain the origin (or any other specified site). From Theorem 2.3 and Eq. 2.4, the probability the origin is a member of a (δ, d)-connected aggregate of $|L|$ occupied cubes is less than $(\exp(K_3(\delta, d))\, z^{1/8})^{|L|}$, which decreases exponentially with increasing $|L|$ if $z < \exp(-8K_3(\delta, d))$. Likewise, the probability the origin is a member of a (δ, d)-connected aggregate of $|L|$ vacant cubes is less than $(\exp(K_3(\delta, d))\, z^{-1/8})^{|L|}$, which decreases exponentially with increasing $|L|$ if $z > \exp(8K_3(\delta, d))$. Analogous bounds are also easily obtained for the two-dimensional analog of this model.

Whether or not the two- or three-dimensional model has an order-disorder transition, in addition to the percolation transition, is not known. There is some numerical evidence for such a transition in two dimensions, but the results are inconclusive on this point.[2–5] In fact, the numerical evidence can be interpreted to imply that the system is disordered at all finite activities.[4,5]

The possibility that these hard-core systems may lack long range order at all finite activities makes these models attractive as a possible starting point for constructing a model ferrofluid. A ferrofluid is a system which exhibits spontaneous magnetization at low temperatures without long-range translational ordering. If ferromagnetic spins are attached to the spheres or disks of the above models, perhaps the resulting systems will exhibit ferrofluid-like behavior. We address this interesting possibility in Section 3.

3. A MODEL FERROFLUID

We now add ferromagnetic spins to the hard spheres of the lattice gas model on Λ discussed in Section 2. In addition, we allow for the possibility of van der Waals attractions between the spheres. If we let $S_i = \pm 1$ represent a sphere at site $i \in \Lambda$ with a $\pm$ spin attached, and let $S_i = 0$ represent a vacancy at site i, then the Hamiltonian for the model can be written as

$$H(\xi) = -\sum_{(ij)} J_{i,j} S_i S_j - \sum_{(ij)} \varepsilon_{i,j} S_i^2 S_j^2 - \mu \sum_{i \in \Lambda} S_i^2 \tag{3.1}$$

where $\xi \in \{-1, 0, 1\}^{|\Lambda|}$, $J_{i,j} > 0$, and $\varepsilon_{i,j} \geqslant 0$. The characteristic function, $\chi(\xi)$, on allowed configurations of hard spheres is given as

$$\chi(\xi) = \prod_{(ij)} (1 - S_i^2 S_j^2) \tag{3.2}$$

where i and j are first-, second-, or third-neighbor pairs of sites. The grand canonical partition function is of the same form as given in Eq. (2.3).

Models having the Hamiltonian of Eq. (3.1), but not the specific hard-core repulsion we consider, have been considered by other authors within the framework of the mean field approximation.[17,18]

In order to model a ferrofluid, we wish to allow configurations containing a maximum number of spheres to have a highly degenerate ground state. That is, we wish to choose $\{J_{i,j}\}$ and $\{\varepsilon_{i,j}\}$ so that the energy of a ground state configuration, containing the maximum number of spheres, is unaffected by the sliding of rows of occupied spheres. This can be accomplished by choosing the couplings as follows:

$$(J_{i,j}, \varepsilon_{i,j}) = \begin{cases} (4J, 4\varepsilon) & \text{if } i, j \text{ fourth neighbors} \\ (2J, 2\varepsilon) & \text{if } i, j \text{ fifth neighbors} \\ (J, \varepsilon) & \text{if } i, j \text{ sixth neighbors} \\ (0, 0) & \text{otherwise} \end{cases} \tag{3.3}$$

The location of fourth, fifth, and sixth neighbors is illustrated in Fig. 1.

At absolute zero, if $\mu > -12(J+\varepsilon)$, the ground states of the system contain the maximum number of spheres, all with aligned spins. There are a large number of such configurations, for the energy is invariant with respect to the sliding of occupied rows of spheres.

It is unclear whether or not the system has long-range translational order as the temperature is increased above zero. Energetically, a pair of vacancy defects will favor residing on a pair of fourth-neighbor sites. Whether or not such an energetic ordering tendency is overcome by entropy considerations is difficult to ascertain. It is quite possible, however, that the system is noncrystalline at low temperatures.

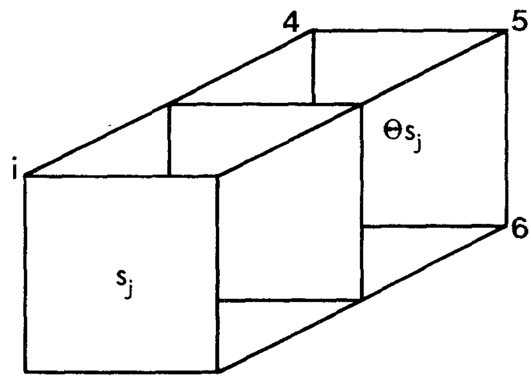

Fig. 1. Two unit cubes of Λ are illustrated. Pictured are a square, s_j, and its reflection, θs_j, through the plane containing the common face of the two unit cubes. Also pictured are examples of fourth-, fifth-, and sixth-neighbor sites to a site i.

We shall now prove that spontaneous magnetization occurs in the model at sufficiently low temperature if the chemical potential is sufficiently large. Let S_i' be the sum of the spins at the vertices of a unit cube with center at $i \in \Lambda'$. Clearly, $S_i' = 0$ if the cube is vacant, and $S_i' = \pm 1$ if the cube is occupied by a sphere with a $\pm$ spin. There is spontaneous magnetization in the model if there exists a constant $c > 0$, independent of i and j, such that $\langle S_i' S_j' \rangle \geqslant c$. Let $p_{S_i', S_j'}$ be the probability that the sum of the spins on the cube at i is S_i' and on the cube at j is S_j'. (A dot will indicate the spin is unspecified.) Since $\langle S_i' S_j' \rangle = 2(p_{+,+} - p_{+,-})$ and $2(p_{+,+} + p_{+,-} + p_{0,\cdot}) - p_{0,0} = 1$, then there is spontaneous magnetization in the model if there exists a constant $c' < 1$, independent of i and j, such that $4p_{+,-} + 2p_{0,\cdot} \leqslant c'$.

In Appendix A we prove that the model satisfies reflection positivity[1] with respect to reflection through planes of lattice sites which are perpendicular to one of the coordinate axes. Using reflection positivity, we show in Appendix B that the probability $P_{L,v}$ that a specified set, L, of cubes are all vacant is bounded as

$$P_{L,v} < \exp(-\varepsilon_0 \, |L|/kT), \qquad \text{where} \quad \varepsilon_0 = 3\varepsilon/2 + 3J/2 + \mu/8$$

As a consequence, $p_{0,\cdot} = P_{1,v} < \exp(-\varepsilon_0/kT)$.

We now use the Peierls argument[14-16] to obtain a bound on $p_{+,-}$. If $S_i' = +1$ and $S_j' = -1$, then either the cube centered at i or the cube centered at j is surrounded by an edge connected set of cubes (a contour), each cube of the contour being either vacant or occupied by a sphere with a spin opposite to the spin on cubes which are interior to the contour. Clearly, spins exterior to the contour do not interact with spins which are interior to the contour.

The probability P_γ that a contour γ occurs in a configuration is given as

$$P_\gamma = \Xi^{-1} \sum_{\xi \supset \gamma} \chi(\xi) \, e^{-H(\xi)/kT} \tag{3.4}$$

Let $C_\gamma = \{\xi : \gamma \subset \xi \in C\}$, and let $C_\gamma^* = \{\xi^*\}$, where ξ^* is the configuration obtained from $\xi \in C$ by flipping all spins in ξ which are interior to γ. If N_0 is the number of occupied cubes in the contour γ, then

$$P_\gamma < \frac{\sum_{\xi \in C_\gamma} e^{-H(\xi)/kT}}{\sum_{\xi^* \in C_\gamma^*} e^{-H(\xi^*)/kT}} \leqslant e^{-2JN_0/kT} \tag{3.5}$$

If N_v is the number of vacant cubes in γ, then the reflection positivity estimate gives $P_\gamma < \exp(-\varepsilon_0 N_v/kT)$. Since $N_0 + N_v = |\gamma|$, then either N_0 or

N_v must be as large as $|\gamma|/2$. Hence $P_\gamma < \exp(-\alpha\,|\gamma|/kT)$, where $\alpha = \min\{J, \varepsilon_0/2\}$.

The standard Peierls argument then gives

$$p_{+,-} < a \sum_{|\gamma|} |\gamma|^{3/2} (be^{-\alpha/kT})^{|\gamma|} \tag{3.6}$$

where a and b are positive constants. If $\mu > -12(J+\varepsilon)$, then $\alpha > 0$. Therefore, there is spontaneous magnetization in the model at sufficiently low temperature if $\mu > -12(J+\varepsilon)$.

In the corresponding model on $\mathbb{Z}^2$, the hard core exclusions extend only through first and second neighbors, and the other interactions are given as

$$(J_{i,j}, \varepsilon_{i,j}) = \begin{cases} (2J, 2\varepsilon) & \text{if} \quad i, j \text{ third neighbors} \\ (J, \varepsilon) & \text{if} \quad i, j \text{ fourth neighbors} \\ (0, 0) & \text{otherwise} \end{cases} \tag{3.7}$$

where $J > 0$ and $\varepsilon \geqslant 0$. A Peierls argument, completely analogous to the one given above, proves the existence of spontaneous magnetization in the model at sufficiently low temperatures provided $\mu > -4(J+\varepsilon)$.

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APPENDIX A: REFLECTION POSITIVITY

Consider a simple cubic lattice Λ with cyclic boundaries given as $\Lambda = \{(a, b, c): a, b, c = 0, 1,..., 2M-1\}$. The coordinates (x, y, z) are computed modulo $2M$ onto $0 \leqslant x, y, z < 2M$.

We consider a lattice gas on Λ with the Hamiltonian defined by Eq. (3.1), together with the characteristic function given by Eq. (3.2).

We define reflection planes $P_a^\pm$ for $0 \leqslant a \leqslant M-1$ as $P_a^- = \{(a, y, z): y, z \in \mathbb{R}\}$ and $P_a^+ = P_{a+M}^-$. Similarly, we define reflection planes $P_b^\pm$ and $P_c^\pm$. The planes $P_a^\pm$ divide Λ into three disjoint regions:

$$\Lambda_a^+ = \Lambda \cap \{(x, y, z): M+a < x < 2M+a,\ \ y, z \in \mathbb{R}\}$$

$$\Lambda_a^- = \Lambda \cap \{(x, y, z): a < x < M+a,\ \ y, z \in \mathbb{R}\}$$

$$\Lambda_a^0 = \Lambda \cap (P_a^- \cup P_a^+)$$

There is a natural involution θ_a: $(x, y, z) \to (2a-x, y, z)$ which reflects the coordinates through the reflection planes $P_a^{\pm}$. If C_a^+ is the set of configurations on Λ_a^+ (and similarly for C_a^- and C_a^0), then $\theta_a C_a^{\mp} = C_a^{\pm}$ and $\theta_a C_a^0 = C_a^0$, the latter being invariant.

For any function f: $C \to \mathbb{C}$, we define $\theta_a f$ as $(\theta_a f)(\xi) = f(\theta_a(\xi))$. We denote ξ as a triple $\xi = (\xi_a^-, \xi_a^0, \xi_a^+)$, where $\xi_a^{\pm} \in C_a^{\pm}$ and $\xi_a^0 \in C_a^0$. Let $F_a^+ = \{f: f(\xi) = f(\xi_a^0, \xi_a^+)\ \forall \xi = \{S_i\}\}$. Then $\theta_a f(\xi) = f(\xi_a^0, \xi_a^-)$ if $f \in F_a^+$. We also define a set of functions F_a^- in an analogous fashion.

Let $s_{j,a}$ denote a unit square in Λ_a^+ which is parallel to and neighboring P_a^+ or P_a^-. One such square is illustrated in Fig. 1. The Hamiltonian can then be written as

$$H(\xi) = H_a^+(\xi) + \theta_a H_a^+(\xi) - \sum_{s_{j,a}} (g_{j,a}^+(\xi)\, \theta_a g_{j,a}^+(\xi) + h_{j,a}^+(\xi) \theta_a h_{j,a}^+(\xi)) \tag{A1}$$

where $g_{j,a}^+ = \sqrt{J} \sum_{i \in s_{j,a}} S_i$ and $h_{j,a}^+ = \sqrt{\varepsilon} \sum_{i \in s_{j,a}} S_i^2$. As such, H_a^+, $g_{j,a}^+$, and $h_{j,a}^+$ are all elements of F_a^+. Moreover, $\chi_a(\xi) = \chi_a^+(\xi)\, \theta_a \chi_a^+(\xi)$, where $\chi_a^+ \in F_a^+$. (Note that each fourth-neighbor interaction occurs four times in the sum, each fifth-neighbor interaction occurs twice, and each sixth-neighbor interaction occurs once.)

If $f \in F_a^+$, then

$$\sum_{\xi} \bar{f}(\xi)\, \theta_a f(\xi) = \sum_{\xi_a^0} \overline{\sum_{\xi_a^+} f(\xi)} \sum_{\xi_a^-} \theta_a f(\xi) \geqslant 0$$

since it is a sum of squares. We can expand $\exp[-H(\xi)/kT]$ in a convergent power series with terms of the form $\alpha \bar{f} \theta_a f$, where $f \in F_a^+$ and $\alpha > 0$. [The minus sign in $H(\xi)$ before the sum over $s_{j,a}$ is crucial to insure $\alpha > 0$.[13]] Hence, for any $f \in F_a^+$, $\langle \bar{f} \theta_a f \rangle \geqslant 0$, which is the condition known as reflection positivity.

A similar argument can be used to show that the two-dimensional analog of this model is reflection positive about lines of lattice sites which are perpendicular to a coordinate axis. The model on $\mathbb{Z}^2$ has a Hamiltonian defined by Eqs. (3.1) and (3.7), together with a characteristic function to account for the hard core exclusion of first and second neighbors. For this model, s_j in Eq. (A.1) will represent an edge of a unit square which borders the reflection line.

The case in which the only interaction is the hard core repulsion is more easily treated. In this case, $H(\xi) = -\mu \sum_i S_i$ where $S_i \in \{0, 1\}$ and $\xi \in \{0, 1\}^{|\Lambda|}$. Then $H(\xi) = H_a^+(\xi) + \theta_a H_a^+(\xi)$ and $\chi(\xi) = \chi_a^+(\xi)\, \theta_a \chi_a^+(\xi)$. The argument given above then yields $\langle \bar{f} \theta_a f \rangle \geqslant 0$ if $f \in F_a^+$.

For all the models, it follows by a standard Cauchy–Schwartz proof that $|\langle fg \rangle|^2 \leqslant \langle \bar{f} \theta_a f \rangle \langle \bar{g} \theta_a g \rangle\ \forall f \in F_a^+, g \in F_a^-$.

APPENDIX B

Here we shall use reflection positivity to obtain upper bounds to the probability a set of unit cubes are all vacant or all occupied. Let C_r be the unit cube with center at r. Let Q_r be the projection onto configurations in which C_r is vacant; that is,

$$Q_r(\xi) = \begin{cases} 1 & \text{if } C_r(\xi) \text{ is vacant} \\ 0 & \text{otherwise} \end{cases}$$

Let L be a (nonempty) set of cubes. Define

$$Q(L) = \prod_{r \in L} Q_r(\xi)$$

The probability $P_{L,v}$ that L is a set of vacant cubes is then bounded as $P_{L,v} = \langle Q(L) \rangle \leqslant g^{|L|}$, where

$$g = \max_L \langle Q(L) \rangle^{1/|L|}$$

Since $Q(L) = Q^+(L)\, Q^-(L)$, where $Q^+ \in F_a^+$ and $Q^- \in F_a^-$, then (see Appendix A), $\langle Q \rangle^2 \leqslant \langle Q^+ \theta_a Q^+ \rangle \langle Q^- \theta_a Q^- \rangle$. If L_m maximizes $\langle Q \rangle^{1/|L|}$, then it also maximizes $\langle Q^+ \theta_a Q^+ \rangle^{1/|L|}$. Hence if $r \in L_m$, then $\theta_a r \in L_m$ as well. But since this is true for θ defined as a reflection through any pair of planes $P_a^\pm$, $P_b^\pm$, $P_c^\pm$, then L_m contains all the $|\Lambda| = 8M^3$ cubes in Λ and corresponds to the completely vacant configuration. Hence

$$g \leqslant \left(\frac{1}{\Xi} \right)^{1/|\Lambda|}$$

Therefore, $g \leqslant \exp(-P_\Lambda/kT)$, where $P_\Lambda = |\Lambda|^{-1}\, kT \ln \Xi$.

A weaker but somewhat more useful bound on g is obtained by noting that $\Xi > \exp(\varepsilon_0 |\Lambda|/kT)$, where $-\varepsilon_0 |\Lambda| = -(3\varepsilon/2 + 3J/2 + \mu/8)|\Lambda|$ is the value of the Hamiltonian corresponding to an allowed configuration in which every cube is occupied and all the spins are aligned. Therefore, $P_{L,v} < \exp(-\varepsilon_0 |L|/kT)$.

A similar argument can be used to bound the probability $P_{L,0}$ that a set L of cubes are all occupied by spheres. In this case, where $|\Lambda| = 8M^3$,

$$g < \left(\frac{6(2^{M+1})^M\, e^{\varepsilon_0 8M^3/kT}}{\Xi} \right)^{1/8M^3}$$

Here $-\varepsilon_0 |\Lambda|$ is the minimum value of the Hamiltonian for configurations in which every cube is occupied. Simple packing considerations indicate that $6(2^{M+1})^M$ is the maximum number of configurations in which every cube is occupied. In the thermodynamic limit, $|\Lambda| \to \infty$, $P_{L,0} < \exp(\varepsilon_0 |L|/kT)$.

For the two-dimensional model, an analogous argument can be used to obtain an upper bound to the probability that a set of $|L|$ unit squares are all occupied by disks or that they are all vacant. The bounds are of the same form as those given above, the only difference being that $\varepsilon_0 = J + \varepsilon + \mu/4$ for the two-dimensional model.

If the only interaction in the model is hard core repulsion, bounds for $P_{L,v}$ and $P_{L,0}$ can be obtained from the bounds given above by setting $J = \varepsilon = 0$. In three dimensions, $P_{L,v} < z^{-|L|/8}$ and $P_{L,0} < z^{|L|/8}$, where $z = \exp(\mu/kT)$. In two dimensions, $P_{L,v} < z^{-|L|/4}$ and $P_{L,0} < z^{|L|/4}$.

APPENDIX C

By counterexample we now show that Theorem 2.3 is false for all $d \geq 1$, if $\delta \geq 2$. Note that sets which are (δ, d)-connected are also (δ', d')-connected for all $d' \geq d$, $\delta' \geq \delta$.

Consider the set $S_0 = \{0, 1, 2, \ldots, p\}$, where $p \geq 2$. Define recursively $S_i = \{S_{i-1}, T^{p^{\delta^i}} S_{i-1}\}$, $1 \leq i \leq j$, where $T^a x \equiv x + a$. Sets S_0, S_1, and S_2 are illustrated in Fig. 2 for the case $p = 2$ and $\delta = 2$. Form new sets S'_j from S_j by translating the outer set (one with larger coordinates) of each neighboring pair of sets S_i, $0 \leq i < j$, from 1 to p^{δ^i} sites away from the neighboring S_i set. The sets S'_j so formed each contain $(p+1)\, 2^j$ elements, including the origin. Moreover, since $\delta \geq 2$ and $p \geq 2$, each set S'_i finally formed from a set S_i has $\operatorname{diam}(S'_i) \geq p^{\delta^i}$ and $1 \leq \operatorname{dist}(S'_i, S'_j \backslash S'_i) \leq p^{\delta^{i+1}}$. Hence the sets S'_j are (δ, d)-connected $\forall d \geq 1$.

Since there are 2^{j-i-1} pairs of S'_i sets, then the number of sets S'_j is

$$\prod_{i=0}^{j-1} (p^{\delta^i})^{2^{j-i-1}} \geq p^{2^{j-1} j}$$

If $p^{2^{j-1} j} \leq e^{K_v(\delta, d)(p+1) 2^j}$, then $K_v(\delta, d) \geq j(\ln p)/(2p+2)$, which increases without bound as j increases.

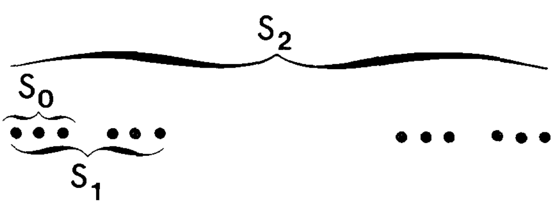

Fig. 2. Sets S_0, S_1, and S_2 are illustrated for the case $p = 2$ and $\delta = 2$.

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