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Cluster and Percolation Inequalities for Lattice Systems with Interactions

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For a lattice gas with attractive potentials of finite range we use the inequalities of Fortuin, Kasteleyn, and Ginibre (FKG) to obtain fairly accurate upper and lower bounds on the equilibrium probability $p(K)$ of finding the set of sites K occupied and the adjacent sites unoccupied, i.e., on the probabilities of finding specified clusters. The probability that a given site, say the origin, is empty or belongs to a cluster of at most l particles is shown to be a nonincreasing function of the fugacity z and the reciprocal temperature $\beta = (kT)^{-1}$; hence the percolation probability is a nondecreasing function of z and β. If the forces are not entirely attractive, or if the ensemble is restricted by forbidding clusters larger than a certain size, the FKG inequalities no longer apply, but useful upper and lower bounds on $p(K)$ can still be obtained if the density of the system and the size of the cluster K are not too large. They are obtained from a generalization of the Kirkwood–Salsburg equation, derived by regarding the system as a mixture of different types of cluster, whose only interaction is that they cannot overlap or touch.

KEY WORDS: Percolation; finite-temperature lattice systems; inequalities; clusters.

1. INTRODUCTION

If particles are placed on some of the sites of a lattice, such as the plane square or simple cubic lattice, the resulting configuration can often usefully be described in terms of clusters, which are sets of occupied sites completely surrounded by vacant sites. An assignment of probabilities to different configurations, such as is given by Gibbs ensembles, then also induces probabilities for the occurrence of different clusters. These cluster probabilities

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are of interest in the percolation problem[1–4] and in the theory of metastable states and nucleation processes in a lattice gas or Ising spin system.[5,6]

In this paper we shall obtain some inequalities for the probabilities of finding particular types of cluster in a lattice gas, and also for the percolation probability, which can be loosely described as the probability that some specified site contains a particle belonging to an infinite cluster. These probabilities will be calculated in a grand canonical ensemble, but the formalism is sufficiently general to apply to a restricted grand canonical ensemble of the type used by Capocaccia *et al.*[5] for the static description of metastable states. In these ensembles the configurations are restricted to those that contain only clusters of specified types (for example, we might require all the clusters to contain less than a specified number of particles). The grand canonical ensemble includes as a special case $\beta = 0$, the ensemble for which the percolation problem has usually been studied,[1] in which the occupation numbers of the different sites are independent and identically distributed (taking the values 0, 1 only); but it also includes the case studied more recently[3,4] in which the occupation numbers are correlated as in a finite-temperature ensemble.

We use two methods for studying the distribution of clusters and the percolation probability. One of them, based on the inequalities of Fortuin, Kasteleyn, and Ginibre (FKG),[7] applies to systems with general one-body and attractive two-body and many-body interactions; it gives good upper and lower bounds on the cluster probabilities for a low-density equilibrium system (a comparison with Monte Carlo estimates of these probabilities is given in the following) and also yields information about percolation probabilities. The other method is more general, applying in particular to the type of restricted ensemble mentioned earlier, to which the FKG inequalities do not apply; it is based on the idea of Minlos and Sinai[8] that a lattice system can be regarded as a "gas" of clusters whose only "interaction" is a repulsion arising from the fact that they cannot overlap or touch. This makes it possible to apply the method of obtaining inequalities for the pressure and correlation functions of systems with repulsive interactions due to Lieb[9] and developed by Penrose[10] and by Lebowitz and Percus.[11] This method is also related to some of the inequalities used by Mürmann[2] in his analysis of clusters in continuous systems.

For the percolation problem, there already exists a large amount of information, based on a variety of analytical and numerical methods, about the "infinite-temperature" case in which the sites are independent. For the Bethe lattice, even the finite-temperature problem can be solved exactly[3]; but for the more interesting two-dimensional square lattice, all that has been proven so far is that for nearest-neighbor attractive interactions and densities $\leqslant \frac{1}{2}$ the percolation probability is zero for $T \geqslant T_c$, the critical temperature, but nonzero for temperatures below T_c if the density is that of the high-density phase.[4] (This last result was also shown to hold for higher dimensionalities.) In this paper our main result concerning percolation is that, when the FKG inequality applies, the probability that a given site is unoccupied or belongs to a cluster containing at most l particles is a nonincreasing function of the reciprocal temperature β and the fugacity z. It follows from this that the percolation probability is a nondecreasing function of β and z. [These results, Eqs. (32) and (33), are independent of the bounds in Section 3 and the interested reader may go directly from Eq. (14) to Section 4.]

2. DEFINITIONS

We consider a lattice gas on a finite set Λ of sites. Each configuration of the lattice gas can be specified by specifying which set of sites is occupied; thus each configuration C corresponds to a subset of Λ (and we simply denote by C the set of occupied sites). The energy of the lattice gas when its configuration is C will be denoted by $E(C)$, and the number of occupied sites by $N(C)$. In a grand canonical ensemble at temperature T and fugacity z the probability of the configuration C is

$$\mu(C) = \frac{z^{N(C)}\{\exp[-\beta E(C)]\}}{\Xi(z, \beta; \Lambda)} \tag{1}$$

where $\Xi(z, \beta; \Lambda)$ is the grand partition function, $\Xi = \sum_C z^N \exp(-\beta E)$, and $\beta = 1/kT$.

For the time being we shall assume that the energy function E is a sum of one-body and two-body terms only (this condition will be relaxed in Section 5). Then $E(C)$ has the form

$$E(C) = \sum_{i\in\Lambda} U_i n_i(C) + \sum_{i<j} V_{ij} n_i(C) n_j(C) \tag{2}$$

where n_i is the occupation number of the ith site, which, for a given configuration C, takes on the value 1 if site i is occupied and the value 0 if site i is unoccupied, i.e.,

$$n_i(C) = \begin{Bmatrix} 1 & \text{if} \quad i \in C \\ 0 & \text{if not} \end{Bmatrix} \tag{3}$$

The sum $\sum_{i<j}$ in Eq. (2) goes over all pairs (i, j) of different sites in Λ, each pair being counted once only.

In order to define clusters, it is necessary to specify which pairs of sites in Λ are to be regarded as *adjacent*. This is done by defining a set B of *bonds* associated with Λ; the bonds are unordered pairs of different sites in Λ and we define two sites to be adjacent if and only if they belong to the same bond. We require that B must include every pair (i, j) for which $V_{ij} \neq 0$; other pairs may be included if desired. For this definition to be useful we want most of the V_{ij} to be zero; this condition is satisfied if Λ is a subset of some regular infinite lattice and there is a finite "range" R such that $V_{ij} = 0$ whenever the Euclidean distance between i and j exceeds R. In particular, if $R = 1$, i.e., nearest neighbor interactions, then bonds can be simply defined as all nearest neighbor connections. Most analyses of clusters are based on such a model.

Given any configuration C, we can now partition it into subsets called *clusters*, in such a way that no member of any cluster is adjacent to a member of another cluster, but if a cluster is divided into two nonempty sets of sites, then at least one site in one of the sets is adjacent to a site in the other, i.e., clusters are sets of occupied sites connected directly or indirectly by bonds. Since the clusters are disjoint, we have

$$N(C) = N(C_1) + N(C_2) + \cdots + N(C_r) \tag{4}$$

where $N(C)$ is the number of sites in C, and $C_1, \ldots, C_r$ are the clusters constituting C. Also, since no sites in different clusters are adjacent we have, from (2),

$$E(C) = E(C_1) + E(C_2) + \cdots + E(C_r) \tag{5}$$

Equations (4) and (5) show that, once we have taken into account the fact that clusters cannot overlap or be adjacent, we can regard them as independent systems. This observation is the basis of most of this paper.

To make use of this cluster description of lattice gas configurations we consider the statistical properties of the clusters. Let K be any subset of Λ. Define $p(K)$ as the probability that all the sites in K are occupied and all the sites not in K that are adjacent to sites in K are unoccupied. If K is connected, then $p(K)$ is just the probability of finding the cluster K. If K is not connected, we can decompose it into connected clusters $K_1, K_2, \ldots$, and $p(K)$ is then the probability of finding all the clusters $K_1, K_2, \ldots$.

Writing $B(K)$ to denote the set of sites not in K that are neighbors of sites in K, i.e., the border or perimeter sites of K, we have, by the rule for conditional probabilities,

$$\begin{aligned} p(K) &= \text{prob}\{\text{all sites of } B(K) \text{ are vacant}\} \\ &\quad \times \text{prob}\{\text{all sites of } K \text{ are occupied} \mid \text{all sites of } B(K) \text{ are vacant}\} \end{aligned} \tag{6}$$

where prob $\{E \mid F\}$ denotes the conditional probability of event E given event F. In the subensemble where all sites of $B(K)$ are vacant, Eqs. (4) and (5) ensure that the sites constituting K are independent of those outside; therefore the conditional probability in (6) is the same as if the sites outside K were removed altogether, and is equal to $\theta(K)/\Xi(K)$, where

$$\theta(K) = z^{N(K)} \exp[-\beta E(K)] \tag{7}$$

and $\Xi(K)$ is an abbreviation for $\Xi(z, \beta; K)$, the grand partition function for a lattice gas confined to the set of sites K. The other factor in (6) can be written in terms of the characteristic function of the event that all the sites of $B(K)$ are vacant, which is $\prod_{i \in B(K)} [1 - n_i(C)]$, and so we obtain from (6)

$$p(K) = \left\langle \prod_{i \in B(K)} (1 - n_i) \right\rangle \theta(K)/\Xi(K) \tag{8}$$

The quantity $\Xi^{-1}(K)$ is the conditional probability that all sites in K are vacant given that all sites in B are vacant. Using this fact in (8), we obtain an alternative expression for $p(K)$,

$$p(K) = \left\langle \prod_{i \in \bar{K}} (1 - n_i) \right\rangle \theta(K) \tag{9}$$

where $\bar{K} = K \cup B(K)$ is the set consisting of all sites in K and in $B(K)$. Equation (9) tells us that $p(K)/\theta(K)$ is equal to the probability, in the grand canonical ensemble on Λ, that all sites in K and $B(K)$ are vacant. This probability is also equal to the ratio of the grand partition functions on $\Lambda - \bar{K}$ and on Λ, so that we obtain the further expression

$$p(K)/\theta(K) = \Xi(\Lambda - \bar{K})/\Xi(\Lambda) \tag{10}$$

All three of these expressions will be used in the next sections to derive upper and lower bounds on $p(K)$. Finally we note that from the definition of clusters we have

$$p(K) = \left\langle \prod_{i \in K} n_i \prod_{j \in B} (1 - n_j) \right\rangle \tag{11}$$

[For the independent-site case, (11) gives $p(K) = \prod_{i \in K} \langle n_i \rangle \prod_{j \in B} (1 - \langle n_j \rangle) = \rho^{N(K)}(1 - \rho)^{N(B)}$, the second equality holding when $\langle n_i \rangle = \rho$ for all i.]

3. ATTRACTIVE FORCES

In this section we consider the case where the potential is attractive:

$$V_{ij} \leqslant 0 \qquad \text{for all} \quad i, j \in \Lambda \tag{12}$$

We obtain simple upper and lower bounds on $p(K)/\theta(K)$ that are correct to first order in the density ρ or fugacity z, and somewhat more complicated bounds that are correct to second order in ρ or z.

Most of these bounds depend on the FKG inequality,[7] which applies, when (12) holds, to any pair f, g of functions of the configuration that are nondecreasing in the sense that

$$A \supset B \quad \text{implies} \quad f(A) \geqslant f(B) \quad \text{and} \quad g(A) \geqslant g(B) \tag{13}$$

An example of such a function is $n_i(C)$, the occupation number at the site i, which clearly can only remain the same or increase (from 0 to 1) when the set of occupied sites is increased.

The FKG inequality is

$$\langle fg \rangle \geqslant \langle f \rangle \langle g \rangle \tag{14}$$

where the averages are taken in the grand canonical ensemble with probability distribution given by (1): e.g., $\langle n_i n_j \rangle \geqslant \langle n_i \rangle \langle n_j \rangle$, which is equivalent to saying that, for attractive interactions, the presence of a particle at site i increases the probability (for a given β and z) that there will be a particle at the site j. Moreover, if $f_1, f_2, \ldots, f_r$ are nondecreasing and nonnegative functions of the configuration (with $r > 2$), then it follows by induction that

$$\langle f_1 f_2 \cdots f_r \rangle \geqslant \langle f_1 \rangle \langle f_2 \rangle \cdots \langle f_r \rangle \tag{15}$$

The inequalities (14) and (15) also hold if f, g, and f_i are nonincreasing functions; to prove this for (14), replace f and g by $-f$ and $-g$, and for (15) use induction as before.

To obtain a simple lower bound on $p(K)$, we take $f_i = 1 - n_i$, where $i \in \bar{K}$; then (9) and (15) give

$$p(K) \geqslant \theta(K) \prod_{i \in \bar{K}} (1 - \rho_i) \tag{16}$$

where $\rho_i = \langle n_i \rangle$ is the probability that site i is occupied. This bound is correct (as an approximate equality) to first order in the density. For a more accurate bound, we can apply (15) to (8) instead of to (9), obtaining

$$p(K) \geqslant [\theta(K)/\Xi(K)] \prod_{i \in B(K)} (1 - \rho_i) \tag{17}$$

This bound is stronger than (16), because in deriving it we apply (15) to fewer factors $1 - n_i$.

The bound (17) is, in fact, correct to second order in the density if $V_{ij} = 0$ for all sites i, j in $B(K)$; on a plane square lattice or simple cubic lattice with nearest neighbor interactions this is true for monomers (i.e., when K is a single site) but not for larger clusters. The reason for this is that the difference between the right-hand sides of (8) and (17) is

$$[\theta(K)/\Xi(K)] \sum_{i < j \in B(K)} [\langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle] + O(\rho^3)$$

and if there is no direct interaction between sites i and j, then $[\langle n_i n_j \rangle - \langle n_i \rangle \langle n_j \rangle]$ is also $O(\rho^3)$.

Upper bounds on $p(K)$ can be obtained in a similar manner. We note that, by FKG, $\langle \prod (1 - n_i) \rangle$ is increased if we weaken the pair interactions. Hence we obtain $\langle \prod (1 - n_i) \rangle \leqslant \prod (1 + ze^{-\beta U_i})^{-1}$, where $(1 + ze^{-\beta U_i})^{-1}$ is the value of $\langle 1 - n_i \rangle$ when there are no pair interactions at all, so that all sites are independent. Hence, using (8) and (9), we obtain

$$p(K) \leqslant [\theta(K)/\Xi(K)] \prod_{i \in B(K)} (1 + ze^{-\beta U_i})^{-1} \leqslant \theta(K) \prod_{i \in \bar{K}} (1 + ze^{-\beta U_i})^{-1} \quad (18)$$

Considered as approximate equalities, both these bounds on $p(K)$ are accurate to first order in z.

To obtain stronger upper bounds on $p(K)$, we define the interaction between the sites in $\bar{K}$ and those outside to be

$$W = \sum_{i \in \bar{K}} \sum_{j \in \Lambda - \bar{K}} V_{ij} n_i n_j \quad (19)$$

Then the definition of Ξ gives

$$\Xi(\Lambda) = \Xi(\Lambda - \bar{K}) \Xi(\bar{K}) \langle e^{-\beta W} \rangle_0 \quad (20)$$

where $\langle \ \rangle_0$ denotes an expectation with respect to a grand canonical ensemble with no interaction between the sites in $\bar{K}$ and those outside. Using (20) in (10), we obtain

$$p(K) = \theta(K)/[\Xi(\bar{K}) \langle e^{-\beta W} \rangle_0] \quad (21)$$

Considered as an equality for $p(K)/\theta(K)$, this is accurate to first order in z.

We now need bounds on $\langle e^{-\beta W} \rangle_0$. Since $n_i n_j$ takes the values 0 and 1 only, and $e^{-\beta V_{ij}} - 1 \geqslant 0$, we have, setting $U_i = 0$ for simplicity,

$$\begin{aligned} \langle e^{-\beta W} \rangle_0 &= \left\langle \prod_{i \in \bar{K}} \prod_{j \in \Lambda - \bar{K}} [1 + n_i n_j (e^{-\beta V_{ij}} - 1)] \right\rangle_0 \\ &\geqslant 1 + \sum_{i \in \bar{K}} \sum_{j \in \Lambda - \bar{K}} \langle n_i n_j \rangle_0 (e^{-\beta V_{ij}} - 1) \\ &\geqslant 1 + \sum_{i \in \bar{K}} \sum_{j \in \Lambda - \bar{K}} [z^2/(1 + z)^2](e^{-\beta V_{ij}} - 1) \end{aligned} \quad (22)$$

The last line is a consequence of the FKG inequality, which implies that $\langle n_i n_j \rangle_0$ is decreased if we weaken any of the (attractive) interactions and is therefore greater than the value it would take if the interactions were removed altogether, which is $[z/(1 + z)]^2$. By substituting (22) into (21), we obtain an upper bound on $p(K)$ that is accurate to second order in z.

A different lower bound on $\langle e^{-\beta W}\rangle_0$ can be obtained by using the convexity of the exponential function and then the FKG inequality as in (22). This bound is, from (19)

$$\langle e^{-\beta W}\rangle_0 \geqslant \exp(-\beta\langle W\rangle_0) \geqslant \exp\left\{-\beta \sum_{i\in\bar{K}} \sum_{j\in\Lambda-\bar{K}} [z^2/(1+z)^2] V_{ij}\right\} \tag{23}$$

but it is weaker than (22) in most practical cases, even though both (22) and (23) are accurate to second order in z.

The methods we have used for upper bounds on $p(K)$ can be adapted to give further lower bounds, which are accurate to second order in the density, but which require a knowledge of the pair distribution. To derive them, we start from the following identity, a companion to (20):

$$\Xi(\Lambda - \bar{K})\Xi(\bar{K}) = \Xi(\Lambda)\langle e^{\beta W}\rangle \tag{24}$$

When combined with (10) this gives

$$p(K) = [\theta(K)/\Xi(\bar{K})]\langle e^{\beta W}\rangle \tag{25}$$

A lower bound on $\langle e^{\beta W}\rangle$, analogous to the bound on $\langle e^{-\beta W}\rangle_0$ given in (22) is

$$\begin{aligned}\langle e^{\beta W}\rangle &= \left\langle \prod_{i\in\bar{K}} \prod_{j\in\Lambda-\bar{K}} [1 - n_i n_j(1 - e^{\beta V_{ij}})] \right\rangle \\ &\geqslant \prod_{i\in\bar{K}} \prod_{j\in\Lambda-\bar{K}} [1 - \langle n_i n_j\rangle(1 - e^{\beta V_{ij}})] \end{aligned} \tag{26}$$

where the inequality follows from (15), since $0 \leqslant 1 - e^{\beta V_{ij}} \leqslant 1$. An alternative, and somewhat weaker, lower bound, obtained from the convexity of the exponential function, is

$$\langle e^{\beta W}\rangle \geqslant \exp\left\{\beta \sum_{i\in\bar{K}} \sum_{j\in\Lambda-\bar{K}} \langle n_i n_j\rangle V_{ij}\right\} \tag{27}$$

If Λ is a periodic box and there are interactions between nearest neighbors only, then we can evaluate the lower bounds (26) or (27) if we know the mean energy of the system, since this is expressible in terms of $\langle n_i n_j\rangle$ with (i, j) any pair of nearest neighbors. We can then use the resulting lower bound on $\langle e^{\beta W}\rangle$ in (25) to obtain a lower bound on $p(K)$ that is accurate to second order in the density.

Table I gives a comparison of the main upper and lower bounds derived in this section with the results of a computer simulation[14] of a lattice gas on a finite, simple cubic lattice with nearest neighbor interactions, periodic boundary conditions, and the one-body potential U_i set equal to zero at every lattice site i. The computer counted the number m_l of clusters of each size l; the bounds on the expectation of this number are calculated by summing the

Table I. Comparison of Calculated Bounds on $\langle m_l \rangle$, the Expectation Number of l-Particle Clusters, with Values Estimated from Computer Simulation, for a 50 × 50 × 50 Simple Cubic Lattice with Periodic Boundary Conditions and Attractive Nearest Neighbor Interactions[a]

Cluster size l	Simple lower bound from (16)	More accurate lower bound from (17)	Estimated value from simulation with standard error	Accurate upper bound from (21) and (22)	Simple upper bound from (18)
1	1252.8	1257.4	1266 ± 4	1266.3	1285.2
2	173.8	175.0	173 ± 2	177.6	181.7
3	40.2	—	41 ± 1.8	—	43.3
4	12.0	—	12.7 ± 0.6	—	13.4
5	4.0	—	4.4 ± 0.3	—	4.7
6	1.5	—	1.6 ± 0.2	—	1.7

[a] The interaction energy V_{nn} and the reciprocal temperature β are related by $\beta|V_{nn}| = 1.5$ (i.e., $T \simeq 0.59T_c$) and the density, 0.0146 particle per site, is the density of the low-density phase at coexistence, estimated by means of a Padé approximant.[14] The fugacity is $e^{-4.5} = 0.0111089$.

bounds given in this section over all l-particle clusters that are possible on the chosen lattice. For example, there are N possible one-particle clusters, where N is the number of lattice sites, and so $\langle m_1 \rangle = Np(\mathbf{1})$, where $\mathbf{1}$ denotes some one-particle cluster. Similarly, we find $\langle m_2 \rangle = 3Np(\mathbf{2})$, where $\mathbf{2}$ is some two-particle cluster, etc. Hence the bounds (16), (17), (21) with (22), and (18) give [noting that on the simple cubic lattice $N(B(\mathbf{1})) = 6$, $N(\bar{\mathbf{1}}) = 7$]

$$Nz(1-\rho)^7 \leqslant Nz(1+z)^{-1}(1-\rho)^6 \leqslant \langle m_1 \rangle \tag{28a}$$

and

$$\begin{aligned} \langle m_1 \rangle &\leqslant Nz[1 + 7z + (6y + 15)z^2]^{-1}[1 + 30(y-1)z^2/(1+z)^2]^{-1} \\ \langle m_1 \rangle &\leqslant Nz(1+z)^{-7} \end{aligned} \tag{28b}$$

where $y = \exp(\beta|V_{nn}|)$, V_{nn} is the value of V_{ij} when (i, j) are nearest neighbors, and we have slightly weakened the better upper bound [line one of Eq. (28b)] by dropping the terms in z^3 and above from $\Xi(\bar{K})$.

The corresponding bounds on $\langle m_2 \rangle$, the expected number of two-particle clusters, are

$$\begin{aligned} 3Nz^2y(1-\rho)^{12} &\leqslant \langle m_2 \rangle \\ 3Nyz^2(1 + 2z + yz^2)^{-1}(1-\rho)^{10} &\leqslant \langle m_2 \rangle \end{aligned} \tag{29a}$$

and

$$\begin{aligned} \langle m_2 \rangle &\leqslant 3Nyz^2[1 + 12z + (15y + 51)z^2]^{-1}[1 + 42(y-1)z^2/(1+z)^2]^{-1} \\ \langle m_2 \rangle &\leqslant 3Nyz^2(1+z)^{-12} \end{aligned} \tag{29b}$$

For $l \geqslant 3$ we have used only the first-order formulas (16) and (18), and weakened the resulting bounds a little further to obtain

$$NQ_l z^l (1 - \rho)^{2+5l} \leqslant \langle m_l \rangle \leqslant NQ_l z^l (1 + z)^{-b(l)-l} \tag{30}$$

where Q_l is the "cluster partition function" defined by

$$Q_l z^l = \sum_{K:N(K)=l} \theta(K) \tag{31}$$

where the sum goes over all *translationally nonequivalent* l-particle clusters that are possible on the chosen lattice. The number $2 + 5l$ is the largest value of $N(B(K))$ compatible with $N(K) = l$, and $b(l)$ is defined as the smallest value of $N(B(K))$ compatible with $N(K) = l$. Numerical data about Q_l and $b(l)$ are given in Table II.

4. PERCOLATION AND RELATED PROBLEMS

If O is the origin of some infinite lattice $\mathscr{L}$, the probability (with respect to some specified measure on configurations in $\mathscr{L}$) that O belongs to an infinite cluster in $\mathscr{L}$ is called the percolation probability. We can use the FKG inequalities to show that for a system in equilibrium at fugacity z and temperature β^{-1} with interactions $V_{ij} \leqslant 0$, the percolation probability is a nondecreasing function of β and z. To do this we first consider the grand canonical ensemble in Λ, where Λ is any finite subset of $\mathscr{L}$ that contains the origin, and define $F_l(C)$ (where $C \subset \Lambda$) as the characteristic function of the event that O is either empty or part of a cluster comprising at most l sites. F_l is a nonincreasing function of C, since adding sites to C cannot reduce the

Table II. Values of $Q_l(\beta)$, for $\beta = 1.5/|V_{nn}|$, and of $b(l)$

l	Q_l	$b(l)$
1	1	6
2	13.445	10
3	301.3	13
4	8,682	15
5	282,852	17
6	10,037,271	18
7	3.7790×10^{8}	—
8	1.48907×10^{10}	—
9	6.07556×10^{11}	—
10	2.55024×10^{13}	—

size of the cluster containing O. It follows, then, from the FKG inequalities that

$$\frac{\partial}{\partial V_{ij}}\langle F_l\rangle_\Lambda = -\beta[\langle F_l n_i n_j\rangle - \langle F_l\rangle\langle n_i n_j\rangle] \geqslant 0, \qquad \frac{\partial\langle F_l\rangle_\Lambda}{\partial U_i} \geqslant 0 \tag{32}$$

and hence that

$$\frac{\partial\langle F_l\rangle_\Lambda}{\partial\beta} \leqslant 0, \qquad \frac{\partial\langle F_l\rangle_\Lambda}{\partial z} \leqslant 0 \tag{33}$$

where we have indicated the region Λ explicitly. Since (32) and (33) hold for every Λ, they also hold for $\langle F_l\rangle$, the expectation value of F_l in the infinite-volume equilibrium state. These states, whether defined as limits of finite-volume states or by the Dobrushin–Lanford–Ruelle equations,[12] need not of course be unique for every β and z. For the case in which we are interested here, however, with $V_{ij} = V(i-j) \leqslant 0$, it is known that the state is unique except possibly when $z = \exp[-\frac{1}{2}\beta\sum_j V(i-j)]$ (corresponding to zero magnetic field in spin language) and β is larger than some minimum value, so that $T \leqslant T_c$.[13]

Since $\langle F_l\rangle$ is monotone nondecreasing in l and $0 \leqslant \langle F_l\rangle \leqslant 1$, its limit as $l \to \infty$, which we shall call $\langle F_\infty\rangle$, exists. We define the percolation probability as[1]

$$P_c(\beta, z) = 1 - \langle F_\infty\rangle \tag{34}$$

which by our inequalities will therefore be a nondecreasing function of β and z.

We can also use the FKG inequality to study the effect of boundary conditions on the percolation probability. The inequality (32) implies that if the one-particle potential is made more attractive at some site i, then $\langle F_l\rangle_\Lambda$ will decrease. This would occur, for example, if the "boundary conditions" outside $\Lambda \subset \mathscr{L}$ were changed from "empty" to "full," decreasing U_i from zero to $U_i = \sum_{j\notin\Lambda} V_{ij} \leqslant 0$. In spin language this corresponds to changing from "minus" to "plus" boundary conditions. This implies in particular that $\langle F_l\rangle_\Lambda$ is monotone nonincreasing (nondecreasing) as Λ grows for empty (full) boundary conditions. It also implies that $P_c(\beta, z)$ for a state obtained as a limit $\Lambda \to \infty$ with empty boundary conditions is not greater than that for a state with full boundary conditions, a result already derived by Coniglio *et al.*[4]

The inequalities we have used for $\langle F_l\rangle$ also apply to the probability that the origin is part of a cluster that is contained in some specified set A (where $O \in A$). Hence the probability that the origin is part of a cluster *not* contained within A increases with β and z.

All our results also apply in an ensemble where we specify that there is a particle at the origin, or, for that matter, particles on all the sites in a specified set B. This means in particular that, for a given Λ, the expected fraction of particles contained in clusters of sizes $\leqslant l$ is a decreasing function of β and z for all l.

We believe that this expected fraction of particles will also decrease when β is increased, while the number of particles in Λ is kept fixed (canonical ensemble), if we have, for example, nearest neighbor attractive interactions with adjacent sites defined as nearest neighbor sites. This is true for the Bethe lattice[3] and is borne out by some computer simulations on the simple cubic lattice.[14] However, we have been unable to prove it in general.

We note here that the event "the origin belongs to an infinite cluster" does not belong to the algebra of local, or quasilocal, observables[13]; its occurrence cannot be determined by looking only at finite regions of an infinite system, nor does it belong to the tail field. The probability of percolation can therefore be, and generally is, a nonanalytic function of the thermodynamic parameters, such as the fugacity z, even when all the correlation functions are analytic in z, e.g., when there are no interactions between the particles.

5. GENERAL INTERACTIONS

If the potential energy function $E(C)$ is not attractive, we can no longer use the FKG inequalities, but we can still use the additivity conditions (4) and (5) to obtain some potentially useful upper and lower bounds. Our reason for looking at these more general energy functions is that we are interested in describing metastable states by means of a restricted ensemble,[5,15] which is a suitably chosen subensemble of the grand canonical ensemble. One way of doing this is to use the subensemble consisting of the configurations whose clusters all have sizes less than or equal to some specified size l^*. In the cluster formalism we have described in Section 2, such an ensemble can be described using the usual grand canonical distribution formula (1) if we redefine the energy function to be equal to $+\infty$ if any cluster containing more than l^* particles is present. This energy function satisfies our basic additivity condition (5), but violates the condition of applicability of the FKG inequalities.

A fairly crude upper bound on $p(K)$ can be obtained from Eq. (8), by using the fact that $0 \leqslant 1 - n_i \leqslant 1$; this bound is

$$p(K) \leqslant \theta(K)/\Xi(K) \tag{35}$$

This may be simplified, without appreciable weakening, by using the inequality $\Xi(K) \geqslant 1 + z \sum_{i \in K} e^{-\beta U_i}$ (obtained by dropping terms in which there is more than one particle present in K), to obtain

$$p(K) \leqslant \theta(K)\Big/\Big(1 + z \sum_{i \in K} e^{-\beta U_i}\Big) \tag{36}$$

These bounds on $p(K)$ are correct only to lowest order in z. More accurate bounds will be derived in Section 6.

For a lower bound on $p(K)$ we may turn again to Eq. (8), and this time use the fact that $\prod_i (1 - n_i) \geqslant 1 - \sum_i n_i$. This gives

$$p(K) \geqslant [\theta(K)/\Xi(K)]\left[1 - \sum_{i \in B(K)} \rho_i\right] \tag{37}$$

where $\rho_i = \langle n_i \rangle$. Considered as an equality, (37) is correct to first order in the density. A simpler but somewhat less accurate lower bound, also correct to first order, can be obtained by applying the same method to (9) instead of (8); this bound is

$$p(K) \geqslant \theta(K)\left(1 - \sum_{i \in \bar{K}} \rho_i\right) \tag{38}$$

In Table III the bounds (36)–(38), together with a more accurate upper bound derived in the next section, are compared with the computer simulation results already quoted in Table I.

6. ALTERNATING BOUNDS

Another way of obtaining both upper and lower bounds on $p(K)$ is to use the idea of Minlos and Sinai[8] that the lattice gas can be regarded as a mixture of clusters, which interact in the same way as the particles in a hard-sphere system—through the fact that they cannot overlap. This makes it possible to apply the type of inequality discovered by Lieb.[9] Our work is also related to that of Mürmann.[2]

Table III. Comparison of Bounds on $\langle m_l \rangle$, Calculated Without Using the Attractive Character of the Interactions, with the Values Estimated from the Computer Simulation Used in Table 1

Cluster size l	Lower bounds from (38)	Lower bounds from (37)	Values from simulation	Upper bound from (49)–(51)	Crude upper bound from (36)
1	1246.7	1253.0	1266 ± 4	1263.8	1373.3
2	171.0	173.2	173 ± 2	179.0	202.8
3	38.8	39.7	41 ± 1.8	—	49.9
4	11.2	—	12.7 ± 0.6	—	15.9
5	3.6	—	4.4 ± 0.3	—	5.7
6	1.2	—	1.6 ± 0.2	—	2.2

For any pair K, K' of subsets of Λ, we define the functions $e(K, K')$ and $f(K, K')$ by

$$e(K, K') = 1 + f(K, K') = \begin{cases} 1 & \text{if } K \text{ and } K' \text{ do not overlap or touch} \\ 0 & \text{if they do overlap or touch, or are identical} \end{cases} \tag{39}$$

Then, in view of the additivity conditions (4) and (5), the definition (6) of $p(K)$ can be written

$$p(K) = \theta(K) \sum_{K'} \theta(K')e(K, K')/\Xi(\Lambda) \tag{40}$$

where the sum goes over all possible configurations K' on Λ. Let the decompositions of K and K' into clusters be

$$K = K_1 \cup K_2 \cup \cdots \cup K_r, \qquad K' = K_1' \cup \cdots \cup K_s' \tag{41}$$

Using additivity again, we have

$$\theta(K) = \theta(K_1)\theta(K_2) \cdots \theta(K_r) \tag{42}$$

and

$$e(K, K') = \prod_{j=1}^{s} [1 + f(K_1, K_j')]e(K_2 \cup K_3 \cup \cdots \cup K_r, K') \tag{43}$$

Since $-1 \leqslant f(K, K') \leqslant 0$, we have[16] [writing f_j for $f(K_1, K_j')$]

$$\begin{aligned} &\prod_j (1 + f_j) \leqslant 1 \\ &\prod_j (1 + f_j) \leqslant 1 + \sum_j f_j \\ &\prod_j (1 + f_j) \leqslant 1 + \sum_j f_j + \sum_{i<j} f_i f_j, \quad \text{etc.} \end{aligned} \tag{44}$$

Substituting the first of these inequalities into (43) and then using the result, with (42), in (40), we find

$$\begin{aligned} p(K) &\leqslant \theta(K_1) \cdots \theta(K_r) \sum_{K'} e(K_2 \cup \cdots \cup K_r, K')\theta(K')/\Xi(\Lambda) \\ &= \theta(K_1)p(K_2 \cup \cdots \cup K_r) \end{aligned} \tag{45}$$

If instead we substitute the second line of (44) into (43) and proceed as before, we find after some manipulations

$$\begin{aligned} p(K) \geqslant{} & \text{right-hand side of (45)} \\ & + \theta(K_1) \sum_{K_1'} f(K_1, K_1')p(K_2 \cup \cdots \cup K_r, K_1') \end{aligned} \tag{46}$$

where we have defined, for any two subsets A, B of Λ,

$$p(A, B) = e(A, B)p(A \cup B) \tag{47}$$

so that $p(A, B) = p(A \cup B)$ if A, B are compatible, so that $e(A, B) = 1$, and $p(A, B) = 0$ if not.

The general form for inequalities such as (45) and (46) is

$$p(K) \lesseqgtr \theta(K_1) \sum_{q=0}^{t} (1/q!) \sum_{K_1'} \cdots \sum_{K_q'} \times \prod_{j=1}^{q} f(K_1, K_j')p(K_2 \cup \cdots \cup K_r, K_1', \ldots, K_q') \tag{48}$$

where the symbol $\lesseqgtr$ means $\leqslant$ if t is even and $\geqslant$ if t is odd.

As an example of the use of these inequalities, we calculate an upper bound on $p(\mathbf{1})$, the probability of a one-particle cluster, for an infinite cubic lattice. Using (48) with $t = 2$, we obtain the formula

$$p(\mathbf{1}) = z\left\{1 - \sum{}' p(K_1') + \tfrac{1}{2} \sum{}'' p(K_1', K_2')\right\} \tag{49}$$

where the first sum goes over clusters K_1' that intersect $\bar{\mathbf{1}}$ and the second goes over ordered pairs K_1', K_2' of compatible clusters that intersect $\bar{\mathbf{1}}$. The sum $\sum'$ can be bounded below using the lower bound (38) on $p(K_1')$; the result is

$$\sum{}' p(K_1') \geqslant \sum_{l} [l + b(l)]Q_l z^l[1 - (2 + 5l)\rho] \tag{50}$$

where $b(l)$ and Q_l are defined just after Eq. (30), and we use as many terms of the sum as we like, provided they are all nonnegative.

To bound the sum $\sum''$, the method we use depends on whether or not the ensemble is restricted to a finite set of clusters. In the restricted ensemble, we can use the upper bound (45), which gives

$$\sum{}'' p(K_1', K_2') \leqslant \sum{}'' \theta(K_1')p(K_2') \leqslant 30\bar{\rho}\rho \tag{51}$$

where the factor 30 is the number of ordered pairs of nonadjacent sites in $\bar{K}$, ρ is the density $[= \sum_{K_2'} N(K_2')p(K_2')]$, and $\bar{\rho}$ is defined by

$$\bar{\rho} = \sum_{K_1'} N(K_1')\theta(K_1') = \sum_{l} lQ_l z^l \tag{52}$$

This series terminates, because we are using a restricted ensemble.

As an example of the use of (49) with (50) and (51), we have compared the observed number m_1 of one-particle clusters observed in the simulation mentioned earlier with the upper bound on $\langle m_1 \rangle$ given by (49) for an ensemble restricted to clusters of ten particles or less, using the same fugacity as before. The upper bound on $\langle m_1 \rangle$ given by this method, and also the analogous upper bound on $\langle m_2 \rangle$, are shown in the fifth column of Table III. To be strictly accurate, these bounds should be compared with the averages of m_1 and m_2 taken only over the sample configurations in which no clusters larger than ten particles were present. These averages are very slightly higher than the ones recorded in column four of Table III, but the difference is small because only two clusters larger than ten were observed in the 60 or so configurations sampled.

When the full grand canonical ensemble is used, the upper bound (51) on $\sum'' p(K_1', K_2')$ does not work since we have no general upper bound on $\bar{\rho}$ (in fact the series defining $\bar{\rho}$ need not now converge.) This difficulty can be overcome by considering separately the parts of the sum $\sum'' p(K_1', K_2')$ in which $N(K_1') \leqslant 2$ and in which $N(K_1') > 2$. We omit the derivation of the resulting upper bound, which is

$$\sum{}'' p(K_1', K_2'') \leqslant 30(Q_1 z + 2Q_2 z^2)\rho + 30[\rho - z(1 - 7\rho) - 6z^2 y(1 - 12\rho)]$$

giving $\langle m_1 \rangle \leqslant 1302$ for the case considered in Tables I and III.

The inequalities (48) are the analogs of the ones forming the "truncated Kirkwood–Salsburg equation"[10] for a gas of particles with repulsive forces, and can be solved iteratively, as in Ref. 10, to give a succession of alternating upper and lower bounds on the $p(K)$'s. We shall not discuss this question further here, however, because the formulas so obtained do not appear to be as useful for practical calculations as the ones we have described.

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