Journal of Statistical Physics, Vol. 9, No. 1, 1973

Phase Transition of Hard Hexagons on a Triangular Lattice

Ole J. Heilmann[1] and Eigil Praestgaard[1]

Received April 2, 1973

Systems of hard hexagons on a triangular lattice are investigated. The orientation of the hexagons is kept fixed, while the size of the hexagons is varied. The existence of a phase transition is proved for all sizes by means of the Peierls'argument. The proof does not imply a phase transition in the continuous limit.

KEY WORDS: Lattice gas; phase transition; Peierls' argument.

1. INTRODUCTION

The Peierls'argument is one of the few methods for proving the existence of a phase transition without explicit calculation. The idea goes back to Peierls[1] and the method was given a rigorous formulation for the ordinary, ferromagnetic Ising model by Griffiths[2] and Dobrushin.[3] The argument was later extended to other lattice models with similar properties[4] and most recently Ruelle[5] has succeeded in applying the method in the first rigorous proof of the existence of a phase transition in a continuous model.

The first application of Peierls'argument to lattice gases with repulsive potential is due to Dobrushin,[6] who modified the argument so that it could be applied to the case of a simple cubic lattice in ν dimensions ($\nu \geqslant 2$) with either nearest-neighbor repulsion or nearest-neighbor exclusion. Later

Work supported by the U. S. Air Force under Contract No. F 44620-71-C-0013.

[1] Department of Chemistry, H. C. Ørsted Institute, University of Copenhagen, DK-2100 Copenhagen, Denmark.

© 1973 Plenum Publishing Corporation, 227 West 17th Street, New York, N. Y.10011.

Heilmann[7] extended this method to the case of a triangular lattice and a hexagonal lattice.

The problem of nearest-neighbor exclusion on the triangular lattice is identical to the problem of placing hard hexagons of a certain size and with a fixed orientation on a triangular lattice. In this paper we let the size of the hexagons increase and show that the phase transition persists as the size goes to infinity. However, the lower bound on the fugacity above which the existence of a solid can be proved increases as N^N, where N is a measure of the linear size of the hexagons, and this is too fast to prove the existence of a solid phase for a continuous system of hard, oriented hexagons.

The problem of a phase transition on a triangular lattice has been investigated for some of the smallest (nearest-, next-nearest-, and third-nearest neighbor exclusion) hexagons by numerical methods.[8] There is general agreement that these systems do have a phase transition from a gaseous to a solid state.

2. PEIERLS' ARGUMENT

In this section we shall give a general description of the traditional version of Peierls'argument, primarily to serve as a reference for the following sections. The aim of the argument is to locate values of the thermodynamic variables where two or more different phases (or states) can exist simultaneously. In order to avoid unnecessary complication, we describe the case of a two-dimensional system which can exist in only two states.

Call the two states A and B. The first step is to define the two states; more precisely, one has to associate with each state a definite local structure such that for a given configuration of the system one can say for each vertex whether it is in state A or state B. A given configuration is then associated with a system of contours separating regions with different local structure, and one could alternatively consider a system of contours as defining the configuration.

If the whole boundary of the system is fixed to have the A structure, then the system consists of closed contours; a vertex with the B structure is then surely inside at least one contour. An upper bound on the probability of finding the B structure at a vertex, given that the boundary of the system is A, is then furnished by the probability of having an outer contour (i.e., a contour that is not inside another contour) around the vertex.

We introduce Z for the partition function

$$Z = \sum_{\text{conf}} w(\text{conf}) \tag{1}$$

where the sum runs over all configurations and $w(\text{conf})$ is the appropriate canonical weight of the configuration in question. The probability of a given (closed) contour C_0 is then given by

$$p(C_0) = \sum_{\mathrm{conf} \ni C_0} w(\mathrm{conf})/Z \tag{2}$$

where the sum is the same as in Eq. (1), but is now restricted to configurations that contain the contour C_0 as an outer contour. An upper bound on $p(C_0)$ can be obtained by making the denominator smaller. The way to do that is to delete terms in Z such that one gets a one-to-one correspondence between terms in numerator and denominator. A term in the denominator is obtained from the corresponding term in the numerator by deleting the contour C_0 but keeping the rest of the system of contours, possibly with a slight adjustment of the contours that were inside C_0.

If the thermodynamic variables are chosen such that the "free energy" of a region where the configuration is all A is the same as the "free energy" for a region of the same shape with the configuration B throughout, then the canonical weight of a configuration can be considered as a product of contributions from the contours. If, furthermore, the contribution from the contours are independent of which state is on which side, then the above estimate of $p(C_0)$ yields

$$p(C_0) \leqslant w(C_0) \tag{3}$$

where $w(C_0)$ is the contribution to the canonical weight from the contour C_0. In general one should be able to make the estimate

$$w(C_0) \leqslant \lambda^{|C_0|} \tag{4}$$

where $| C_0 |$ is the length of the contour C_0 and λ is the canonical weight per unit contour. It is essential for the argument that the two states have been chosen such that $\lambda < 1$ and it should even be possible to make λ much smaller than one by a suitable choice of the thermodynamic variables. (The condition $\lambda < 1$ is obviously necessary from a physical viewpoint since it implies that the contours should be energetically unfavorable.)

To finish the estimate of the probability of finding a vertex with configuration B, one has to sum $p(C_0)$ over all possible contours C_0 that enclose the vertex. The number of different contours of length l surrounding a fixed point can in general be bounded by γq^l, where γ and q are some constants.

With σ for the smallest possible length of a contour, one finds the following estimate for the probability of finding B if the boundary is A:

$$p_B \leqslant \gamma' \sum_{l=\sigma}^{\infty} (\lambda q)^l \tag{5}$$

It is easily seen that with sufficiently small λ one can get the bound on p_B to be less than $\frac{1}{2}$. By interchanging the roles of A and B, one then finishes the argument by observing that the state of the system consequently depends on the boundary condition.

3. THE MODEL

The model is the lattice gas model, the lattice is the triangular lattice, and the particles are hard hexagons. The contours of the hexagons are confined to the lattice points (the usual lattice gas restriction). The orientation of the hexagons is fixed by the requirement that the edges of the hexagons should be parallel to the edges of the lattice. For convenience we keep the spacing of the lattice fixed and let the size of the hexagons increase. Since we want to be able to cover the lattice with nonoverlapping hexagons, the length of an edge of a hexagon has to be an integer multiple of the lattice constant (the distance between neighboring points in a lattice); we will denote this integer by N. The case $N = 1$ corresponds to the problem considered earlier of nearest-neighbor exclusion. In the following we consider for convenience $N > 1$.

The number of lattice points inside a hexagon will be $M = 3N^2$ if the points on the edges of the hexagons are counted properly. Having defined the model, we proceed to define the concepts necessary for the Peierls argument. A region with one state is a region with close-packed hexagons. The structures of different states are generated by translating one such ordered state on the triangular lattice by an amount less than the diameter of the hexagons. The number of different states equals M.

In the earlier applications of Peierls'argument one had essentially only one type of contour; in this case, however, the large number of different states leads to many different types of contour.

If a configuration contains a void large enough to accommodate additional hexagons, we imagine it to be filled up with virtual hexagons. The problem of distinguishing between real and virtual hexagons will be considered later; for the moment we shall treat them alike and consequently we can assume that no configuration contains a void large enough to accommodate a hexagon.

Definition 1. An element of contour is either an area not covered by hexagons or a common part of the edges of two hexagons belonging to two different states.

Definition 2. One contour is a set of connected elements of contour not connected to any other elements of contour (see Fig. 1). This definition clearly allows contours within contours.

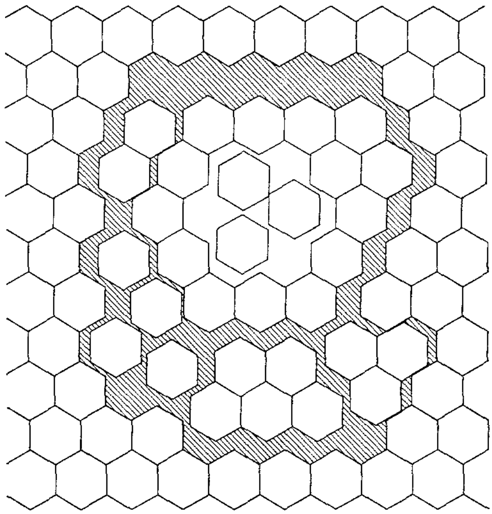

Fig. 1. An outer contour (shaded area and heavy lines) with another contour inside it.

Definition 3. The area $A(C)$ of a contour C equals one-half the number of elementary triangles constituting the empty area of the contour.

As usual we consider the boundary of the system to have one fixed structure which we will call the A structure. A point inside the boundary not covered by a hexagon belonging to the A structure must necessarily be surrounded by or part of a contour. We only need to consider contours not surrounded by other contours; such contours necessarily are surrounded by the A structure.

We shall now consider how one such contour is specified. Imagine the whole space covered with hexagons of the A structure. The edges of these hexagons make up a hexagonal lattice. In order to form a contour, we start by making a hole in the A structure. This hole is specified by a self-avoiding polygon on the hexagonal lattice mentioned above. This polygon, which constitutes the boundary between the outer A structure and the contour, is denoted by B. Besides specifying this outer boundary, we also have to specify the inner boundaries of the contour in order to get a complete specification of the contour. This is done by giving the positions of all the hexagons next to the boundaries.

A contour C divides the interior hexagons into regions which are separated from each other and from the outer A structure by C; we term these regions *C-interior regions*. The hexagons along the boundary of a region (i.e., the hexagons adjacent to C) will necessarily all have the same structure;

we term this structure *the boundary structure of the region.* (Inside a region there might very well be additional contours.)

According to the method considered in Section 2, we now have to describe how to delete terms in the denominator on the right-hand side of Eq. (2) in order to secure a one-to-one correspondance between the configurations included in the summation in the denominator and the configurations making up the numerator. The hexagons of a C-interior region are translated by an amount which transforms the boundary structure of the region into the A structure. If all the C-interior regions are translated in essentially the same direction (it is always possible to chose a direction within a fixed angle of 60°) and if no region is translated longer than necessary, then this transformation will not cause any hexagons to overlap. By a trivial conservation law one has the same total area [namely $A(C)$] left uncovered before and after the transformation; but after the transformation all the hexagons around this uncovered area have the A structure and the void can consequently be completely covered by hexagons with the A structure. If this is done, one gains $A(C)/M$ hexagons.

With

$$z = e^{\beta\mu} \tag{6}$$

for the fugacity of a hexagon, one then obtains

$$p(C) \leqslant e^{-A(C)\beta\mu/M} \tag{7}$$

The probability of a given point x being surrounded by or part of a contour is given by

$$\tilde{p}(x) = \sum_{C \ni x} p(C) \tag{8}$$

where the sum runs over all outer contours C with the property that the point x is inside the outer boundary B of C. Introducing (7) into (8), one obtains

$$\tilde{p}(x) \leqslant \sum_{C \ni x} e^{-A(C)\beta\mu/M} \tag{9}$$

or, introducing the destinction between the outer boundaries and the inner hexagons next to C,

$$\tilde{p}(x) \leqslant \sum_{B \ni x} \sum_{\substack{\text{inner} \\ \text{hex}}} e^{-A(C)\beta\mu/M}$$

or,

$$\tilde{p}(x) \leqslant \sum_{B \ni x} S(B) \tag{10}$$

$$S(B) = \sum_{\substack{\text{inner} \\ \text{hex}}} e^{-A(C)\beta\mu/M} \tag{11}$$

The summation over the inner hexagons can be split up into a summation over the outer inner hexagons, i.e., the hexagons nearest to B and a summation over the rest:

$$S(B) = \sum_{\substack{\text{outer}\\ \text{inner hex}}} \sum_{\substack{\text{inner}\\ \text{inner hex}}} e^{-A(C)\beta\mu/M} \tag{12}$$

The maximum number of hexagons nearest to B will be given by the shape of B; let us call this number n. The first summation will then be over the possible values of their position coordinates, $j(1), j(2),..., j(n)$. The problems arising in connection with hexagons which are not placed will be dealt with in the appendix. Similarly, the maximum number of inner inner hexagons will be given by the shape of B; let us denote this number by m. The second summation will then be a summation over the position coordinates of these hexagons, $j(n + 1),..., j(n + m)$; we again defer the proboem of unplaced hexagons to the appendix.

Then $S(B)$ can be written as

$$S(B) = \sum_{j(1)} \sum_{j(2)} \cdots \sum_{j(n)} \sum_{j(n+1)} \cdots \sum_{j(n+m)} e^{-A(C)\beta\mu/M} \tag{13}$$

The value of $A(C)$ depends on B and all the position coordinates $j(1),..., j(n + m)$, and we introduce a set of rules on how to divide $A(C)$ into contributions from each of the $n + m$ hexagons. The details of these rules are described in the appendix; for the present it suffices that we can write

$$A(C) = \sum_{i=1}^{n+m} a_i \tag{14}$$

where a_i is the part of $A(C)$ attributed to the ith hexagon. Making the rules for the division of $A(C)$ such that a_k is independent of $j(l)$, $l > k$, and introducing the abreviation

$$b_i = e^{-a_i\beta\mu/M} \tag{15}$$

we can now write Eq. (13) in the form

$$S(B) = \sum_{j(1)} b_1 \cdots \sum_{j(n)} b_n \sum_{j(n+1)} b_{n+1} \cdots \sum_{j(n+m)} b_{n+m} \tag{16}$$

In the appendix we show the following inequality:

$$\sum_{j(i)} b_i \leqslant \max\{\delta(\beta\mu/M, N), 1\}, \qquad i > n \tag{17}$$

for fixed values of the positions of the preceding hexagons $j(1),..., j(i - 1)$; it

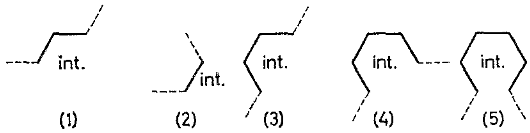

Fig. 2. The five types of segments into which the outer boundary B of a contour can be broken. The segments are shown by heavy lines and the continuation of B is shown dashed; the interior side is marked int. Note that the segments are terminated by corners where the interior angle is 240°, while segments continue over corners where the interior angle is 120°.

is important that the bound δ is independent of these positions. (Note that $i > n$ implies that the ith hexagon is an inner inner hexagon.) If Eqs. (16) and (17), are combined we obtain the inequality

$$S(B) \leqslant 1 \sum_{j(1)} b_1 \cdots \sum_{j(n)} b_n \tag{18}$$

provided $\delta \leqslant 1$.

Since the outer boundary B is a self-avoiding polygon on a hexagonal lattice, it can be considered to be made up of the five types of segments shown in Fig. 2. The segment type 5 cannot, in fact, occur since it would have been filled with a virtual hexagon and the boundary would then look different.

Calling the length of the boundary l (in units of the edges of the hexagons), the number of B's with a given length l surrounding a fixed hexagon ν is less than $\nu(l)$:

$$\nu(l) = \left(\frac{3 \cdot 2^{l-3}}{2^l}\right)\left(\frac{1}{48}\, l^2 + \frac{1}{4}\right) \tag{19}$$

The number in the first parentheses is an upper bound on the number of shapes for given l, and the number in the next parentheses is an upper bound on the number of hexagons that can be inside a polygon of length l. It is clear that l has to be even. The lowest possible value for l is 12 (see Fig. 3). If

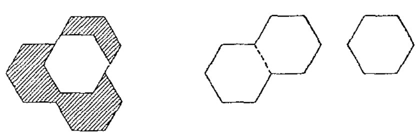

Fig. 3. Left: A contour with the smallest possible outer boundary. Right: Two possibilities which will not occur since the voids will necessarily be covered by virtual hexagons.

we introduce $\eta(i)$ for the number of segments of type i, we find the following expression for l:

$$l = 2\eta(1) + \eta(2) + 3\eta(3) + 4\eta(4) \tag{20}$$

Since B is a self-avoiding polygon, it follows that the numbers of segments of type 2, 3, and 4 are related by

$$\eta(2) + 6 = \eta(3) + 2\eta(4) \tag{21}$$

In general it will be possible to place one outer inner hexagon outside each segment. However, the hexagons placed outside segments of type 2 will actually be regarded as inner inner hexagons. For the other possibilities one obtains bounds of the same type as Eq. (17):

$$\sum_{j(i)} b_i \leqslant \gamma_k(\beta\mu/M, N), \qquad i \leqslant n \tag{22}$$

where the index k refers to the type of segment next to which the ith hexagon is located, $k = 1, 3, 4$. In the appendix we obtain the following values for γ_k :

$$\gamma_1 = \frac{e^{-\alpha N}(2 - e^{-\alpha N})}{(1 - e^{-\alpha N})^2} + 2N^2 \exp(-\alpha N^2) \tag{23}$$

$$\gamma_3 = \frac{e^{-2\alpha N}(2 - e^{-2\alpha N})}{(1 - e^{-\alpha N})^2} + 2N^2 \exp(-2\alpha N^2) + \exp(-\tfrac{3}{2}\alpha N^2) \tag{24}$$

$$\gamma_4 = \frac{e^{-2\alpha N}}{1 - e^{-2\alpha N}} \tag{25}$$

where α is defined as

$$\alpha = \tfrac{1}{3}\beta\mu/N^2 \tag{26}$$

One can find an α_0 independent of N such that for $\alpha > \alpha_0/N$ $(N > 1)$

$$\gamma_4 > {\gamma_2}^2$$

and

$$\gamma_4 > {\gamma_1}^3 \tag{27}$$

If one substitutes (27) and (20)–(22) one obtains

$$S(B) \leqslant \gamma_4^{l/4} \tag{28}$$

If one substitutes (19) and (28) into Eq. (10) one now obtains (using $i = 2l$ as summation variable)

$$\begin{aligned}\tilde{p}(x) &\leqslant \frac{1}{32} \sum_{i=6}^{\infty} \left(i + \frac{3}{i}\right)(16\gamma_4)^{i/2} \\ &\leqslant \frac{7}{32} \frac{(16\gamma_4)^3}{(1 - 4\,\gamma_4^{1/2})^2}\end{aligned} \tag{29}$$

provided $\alpha > \alpha_0/N$ and $\delta \leqslant 1$.

In the appendix the following expression for δ is proven:

$$\begin{aligned}\delta = {} & [\exp(-\tfrac{1}{2}\alpha)]\{1 - \exp[-\alpha(N-1)]\}^{-1}\{1 - \exp[-\alpha(N+1)]\}^{-1} \\ & + 5N^2 \exp(-\alpha N^2/2)\end{aligned} \tag{30}$$

For $N > 1$ one can find a positive constant ω_0 such that $\delta \leqslant 1$ provided

$$\beta\mu = 3N^2\alpha \geqslant \omega_0 N \ln N \tag{31}$$

It is easy to chose ω_0 so that Eq. (31) implies $\alpha > \alpha_0/N$. Actually one finds for N large enough that the best possible estimate for ω_0 is 3. This expression is substituted into (29):

$$\tilde{p}(x) \leqslant \omega N^{-2\omega_0}$$

where ω is a constant independent of N for a given ω_0.

There remains the problem of the virtual hexagons. If a point is covered with a virtual hexagon not belonging to the outer A structure, the case is already treated by the preceding argument since the hexagon will necessarily be inside a contour. The probability of a point being covered by a virtual hexagon belonging to the outer A structure is obviously less than $e^{-\beta\mu}$,

$$p_v(x) < e^{-\beta\mu} \leqslant e^{-\omega_0 N \ln N} \tag{33}$$

The total probability of a given point x not being covered by a hexagon belonging to the outer A structure is

$$p_{\bar{A}}(x) \leqslant \tilde{p}(x) + p_v(x) \tag{34}$$

Combining Eqs. (32)–(34), it is possible by chosing ω_0 sufficiently large to ensure that

$$p_{\bar{A}}(x) < 1 - (1/3N^2) \tag{35}$$

Since we have all together $3N^2$ different structures, the inequality (35) implies that the A structure is strictly more probable than at least one of the other structures; from this one easily finishes the argument by concluding that the structure depends on the boundary; consequently the system is not in the fluid state.

4. CONCLUSION

Using the Peierls argument, we have proved the existence of phase transitions in a system of hard hexagons on a triangular lattice for values of the fugacity

$$z > N^{\omega N}$$

We observe, however, that this bound on the fugacity gives no proof of a phase transition in a continuous system of hard hexagons, and one should not expect a possible phase transition to be provable by this method since the ordered structure obtained in the continuous limit clearly has no phase volume. If the ordered state exists of a given fugacity, and if it consists of large areas covered with close-packed hexagons, then this phase has little to do with the solid phase conjectured for hard disks in the continuum case.

The above proof for the coexistence of solid states is based on the improbability of a liquid phase (it is $\delta \leqslant 1$ which determines the bound on $\beta\mu$), and once this is fulfilled the surface of the liquid drop is automatically so unfavorable that the liquid drop becomes very unlikely

The final estimate

$$\beta\mu \geqslant 3N \ln N, \qquad N > N_0$$

seems remarkably independent of the details of the proof. The above proof easily extends to other orientations of the hexagons relative to the triangular lattice, among which cases are the case of second-nearest-neighbor exclusion on the triangular lattice.

APPENDIX

In this appendix we prove the upper bounds to the contributions from single hexagons given in Eqs. (23)–(25) and (30). The starting point is Eqs. (14)–(16).

The first problem is to define a division of the free area $A(C)$ into contributions from the individual hexagons as indicated in Eq. (14). Since we want an upper bound, it is clear that we are allowed to neglect part of the area.

The general principle for attributing empty area to a hexagon will be to consider the empty area that a given hexagon prohibits any other hexagon from covering as belonging to that hexagon. The problem is then to avoid counting the same area twice. Let us first consider the outer inner hexagons.

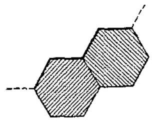

Fig. 4. Two adjacent segments of the outer boundary of a contour. The segments are both of type 1; they are shown by heavy lines. The shaded hexagons next to the segments are on the interior side.

Figure 4 shows two pieces of contour of type 1. For the orientation shown in the figure the hexagon considered to be the adjacent hexagon to a given piece of contour will be the hexagon with its upper left corner inside the shaded area of hexagonal shape shown next to the piece of contour. With this definition there can be no or one hexagon adjacent to a piece of contour, but never more than one. For a given position of the adjacent hexagon the area not coverable is calculated as if no other hexagon had yet been placed. In order to avoid double counting of uncoverable area, we make the main convention of only counting such an area if it belongs to the aforementioned shaded area. However, we also include an uncoverable area outside the shaded area if it is certain that it cannot be made uncoverable by another hexagon; see Fig. 5 for an example.

In placing the adjacent hexagon, six principally different positions will occur corresponding to six different shapes of the uncoverable area (see Fig. 6). The two cases denoted A and B on the figure will give identical contributions because of symmetry.

Figure 7 shows an example of case A. Introducing x and y coordinates

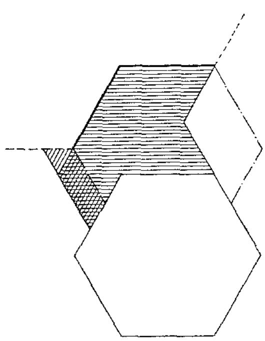

Fig. 5. A boundary segment of type 1 (heavy line) and the adjacent hexagon. The boundary of the region of hexagonal shape inside which uncoverable area always is counted is indicated with dot-dashed lines. The actual uncoverable area inside this region is shaded horizontally. Further uncoverable area is shown obliquely shaded (not counted) and cross-hatched (counted, since it can not be made uncoverable by another hexagon).

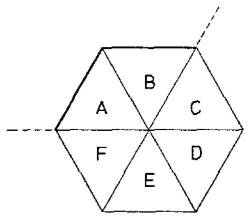

Fig. 6. If the position of the hexagon is given by the position of the upper left corner, each of the six different shapes of the uncoverable area will correspond to the corner in one of the six triangles shown on the figure.

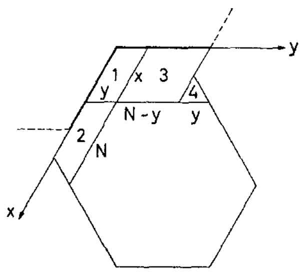

Fig. 7. A segment of type 1 and the adjacent hexagon in a position corresponding to case A. The x and y coordinates which give the positions of the hexagons are shown. The part of the uncoverable area that is counted is divided into pieces numbered from 1 to 4 in the order which corresponds to the order of the terms in Eq. (A.1).

as shown in the figure and taking the lattice constant of the triangular lattice as unit length, one finds the uncoverable area

$$\begin{aligned} a' &= xy + \tfrac{1}{2}y(N + N - y) + x(N - y) + \tfrac{1}{2}y^2 \\ &= N(x + y) \end{aligned} \tag{A.1}$$

(this corresponds to the unit of area being two elementary triangles as introduced in Definition 3).

For the cases C–F one finds that the area is certainly larger than N^2. Also, if no hexagon is placed (for example, see Fig. 8), there will be an uncoverable area of at least N^2 which is not counted in connections with other hexagons (such an area is shown shaded in the figure).

Using the definition of b, Eq. (15), one finds on summing over all possible positions of the hexagon including the possibility of not placing a hexagon

$$\sum b < \sum_{x=0}^{N} \sum_{y=0}^{N} \exp[-\alpha N(x + y)] - 1 + 2N^2 \exp(-\alpha N^2) \tag{A.2}$$

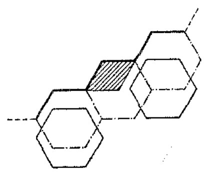

Fig. 8. Three consecutive segments of type 1. The two adjacent hexagons figure belong to the first and third segments. No hexagon can be placed next to the middle segment. The uncoverable area that is counted for the middle segment is shaded.

The minus one comes from the fact that the position $x = y = 0$ is not allowed since that would mean that the hexagon would belong to the outer A structure. Extending the summation to infinity, one easily confirms Eq. (23).

Next we consider a piece of contour of type 3. For the orientation shown in Fig. 9 the hexagon adjacent to the given piece of contour has its upper left corner inside the shaded area.

We do not get more possibilities for the position of the upper left corner than the ones inside the shaded area because we also want the upper right corner to be inside the hexagon on the figure in order to ensure that neighboring pieces of contour are treated consistently.

For a given position of the adjacent hexagon the uncoverable area must belong to the aforementioned hexagon in order to avoid double counting. However, we also include here the uncoverable area outside the hexagon if it cannot be made uncoverable by another hexagon.

Two principally different positions of the adjacent hexagon will occur corresponding to two different shapes of the uncoverable area (see Fig. 10). In Fig. 11 an example of case A is shown. Introducing x and y coordinates as shown in the figure one finds the uncoverable area

$$\begin{aligned} a' &= \tfrac{1}{2}x(x + y + N + N + y) + \tfrac{1}{2}(x + y)(N - x + N + y) \\ &\quad + \tfrac{1}{2}y(N + N - y) \qquad\qquad (\text{A}.3) \\ &= 2N(x + y) + xy \geqslant 2N(x + y) \end{aligned}$$

For case B one finds that the area is certainly larger than $2N^2$. When no

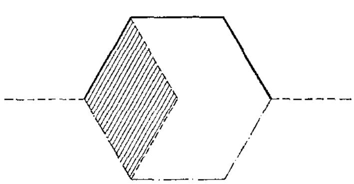

Fig. 9. A boundary segment of type 3 (heavy lines). The boundary of the hexagonal region inside which uncoverable area always is counted is indicated with dot-dashed lines. The upper left corner of the adjacent hexagon must be inside the shaded region.

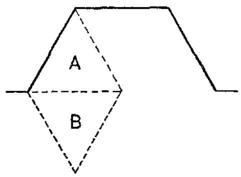

Fig. 10. The two regions A and B that give rise to two different shapes of the uncoverable area when the upper left corner of the adjacent hexagon is placed inside them.

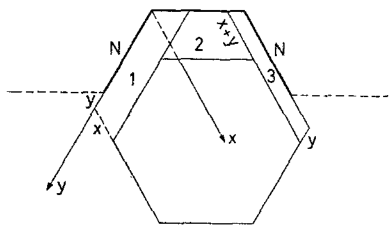

Fig. 11. A segment of type 3 and the adjacent hexagon in a position corresponding to case A. The x and y coordinates which give the positions of the hexagons are shown. The part of the uncoverable area that is counted is divided into three pieces numbered from 1 to 4 in the order which corresponds to the order in Eq. (A.3).

hexagon is placed one obtains an area larger than $\frac{3}{2}N^2$. Using the definition of b [Eq. (15)], one finds on summing over all possible positions of the hexagon (including no hexagon)

$$\sum b \leqslant \sum_{x=0}^{x+y+N} \sum_{y=0} \exp[-\alpha 2N(x+y)] - 1 + \tfrac{1}{2}N^2 \exp[-2\alpha N^2] + \exp(-\tfrac{3}{2}\alpha N^2) \tag{A.4}$$

Extending the upper limit of summation to infinity, one easily confirms Eq. (24).

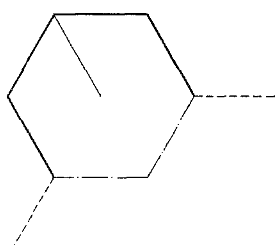

Fig. 12. A boundary segment of type 4 (heavy lines) and the hexagonal region inside which uncoverable area is counted (dot-dashed lines). The adjacent hexagon will have the upper left corner on the thin line.

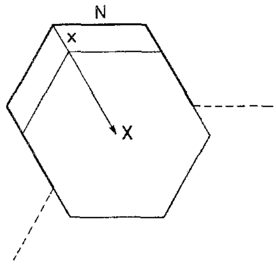

Fig. 13. An adjacent hexagon placed next to a boundary segment of type 4 (heavy lines).

The last piece of boundary to consider is type 4. For the orientation shown on Fig. 12 the adjacent hexagon has its upper left corner on the thin line. This time we only include uncoverable area inside the hexagon indicated on the figure. Figure 13 shows an example. Introducing the x coordinate as shown on the figure, one finds the uncoverable area

$$a = 2Nx \tag{A.5}$$

Again using the definition of b one finds on summing over all possible values of x

$$\sum b = \sum_{x=1}^{N} e^{-\alpha 2Nx} \tag{A.6}$$

Extending the summation to infinity, one confirms Eq. (25).

We have thus dealt with the outer inner hexagons in full agreement with the convention introduced for splitting up the area of the contour into contributions from the individual inner hexagons.

While the outer inner hexagons were placed independently of each other with no preferred order, the inner inner hexagons will be placed in fixed order such that an inner hexagon is placed close to already fixed hexagons, thereby enlarging the uncoverable area, and thus stepwise continuing the

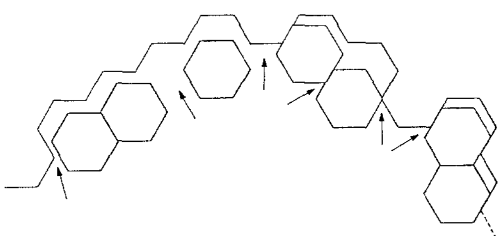

Fig. 14. Part of the outer boundary of a contour with the adjacent hexagons placed. The arrows indicate where to start placing the inner hexagons.

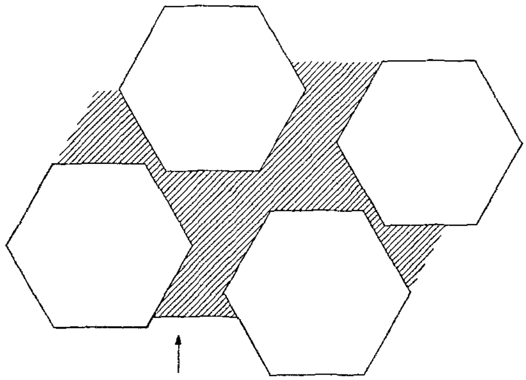

Fig. 15. The general situation that arises when an inner inner hexagon has to be placed. Some hexagons have already been placed and some area secured as uncoverable (cross-hatched on the figure). The next hexagon is to be placed as indicated by the arrow.

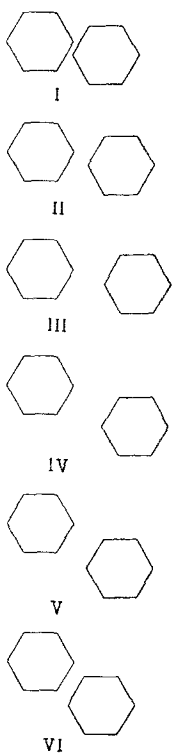

Fig. 16. The six principal positions of the two preceding hexagons.

contour. The starting point will be that we have placed all the outer inner hexagons. In Fig. 14 part of the boundary is shown with the adjacent hexagons in position. The arrows then indicate where the first inner inner hexagons are to be placed. The general situation is the one shown in Fig. 15. The next hexagon is to be placed as indicated by the arrow and the area that has already been secured as uncoverable is cross-hatched; for our purpose the position of the last two hexagons will give sufficient information about the preceding part of the contour.

Taking full account of the symmetry of the underlying triangular

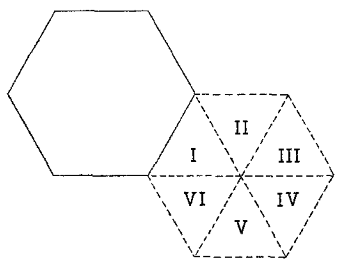

Fig. 17. When the leftmost corner of a hexagon is placed consecutely in each of the six triangles the six main positions shown in Fig. 16 arise.

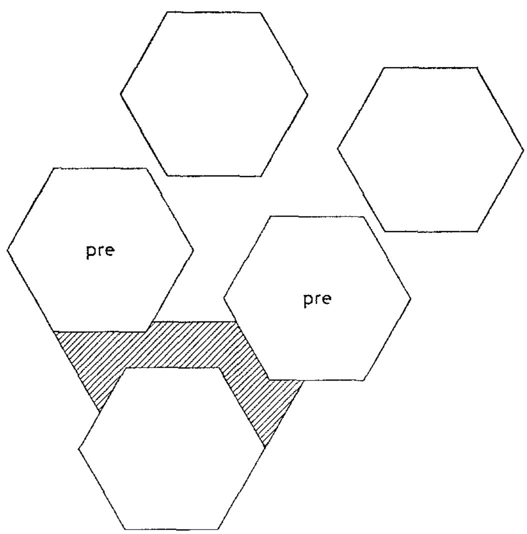

Fig. 18. The enlargement of the uncoverable area (shaded) that is obtained by placing a hexagon next to two preceding hexagons (marked pre).

lattice, one finds six principal positions of the two preceding hexagons (see Fig. 16). These six main positions arise when the leftmost corner of the rightmost hexagon is restricted to each of the six triangles shown in Fig. 17. The uncoverable area a attributed to an inner inner hexagon is the enlargement of the uncoverable area obtained by placing this hexagon relative to the two preceding ones as shown in Fig. 18. It is clear that this convention ensures that we do not include uncoverable area twice.

The possible ways of placing this hexagon are limited by the requirement that it should be the next inner inner hexagon and the fact that large enough voids are filled with virtual hexagons. In a few special cases this will force us to consider the placement of two hexagons simultaneously or to use the positions of the three preceding hexagons to gain sufficient information about the preceding parts of the contour.

Each of the six main cases will give rise to a large number of principally different positions of the next hexagon; we shall not amuse the reader by working out all the cases but confine ourselves to case VI, which turns out to give the largest value of $\sum_j b(j)$.

A number of principally different positions for the next hexagon arise each corresponding to a different shape of the uncoverable area. Figure 19 shows an example, with the two preceding hexagons placed in accordance with case VI, the relative positions of the two hexagons being determined by introducing coordinates u and v as shown in the figure. Defining the position of the next hexagon by the position of the upper right corner, each of the numbered areas will correspond to principally different shapes of the uncoverable area. Additional principally different positions exist outside the numbered areas.

In Fig. 20 is shown an example of the next hexagon being in position 1.

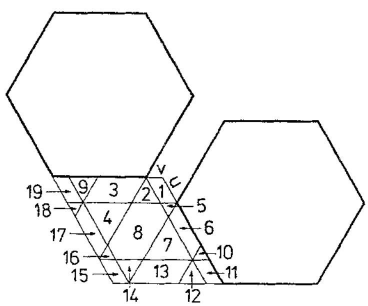

Fig. 19. The two preceding hexagons placed in accordance with case VI; the relative position is defined by u and v as shown. If the position of the next hexagon is defined by the position of the upper right corner, each of the numbered regions corresponds to principally different positions of this hexagon.

Introducing x and y coordinates as shown in the figure, one finds the uncoverable area

$$\begin{aligned} a' &= \tfrac{1}{2}(v-x)^2 + y(v+N) + x(N+u-y) + \tfrac{1}{2}(u-y)^2 \\ &= \tfrac{1}{2}u^2 + \tfrac{1}{2}v^2 + N(x+y) + (x-y)(u-v) + \tfrac{1}{2}(x-y)^2 \end{aligned} \tag{A.7}$$

Figure 21 shows an example of the next hexagon being in position 4. Just as for the hexagon in position 1, one finds the uncoverable area

$$a' = \tfrac{1}{2}u^2 + \tfrac{1}{2}v^2 + N(x+y) + (x-y)(u-v) + \tfrac{1}{2}(x-y)^2$$

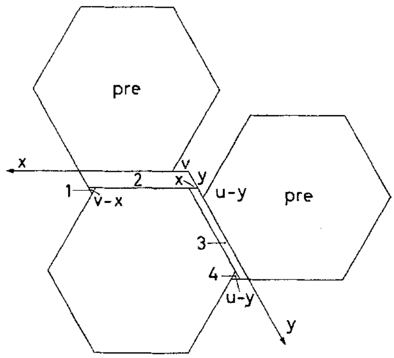

Fig. 20. The two preceding hexagons (marked pre) placed as case VI and the next hexagon placed in position 1 (see Fig. 19). The position of the next hexagon is given by x and y coordinates as shown. The new uncoverable area is divided into parts numbered in correspondance with the order of the terms appearing in Eq. (A.7).

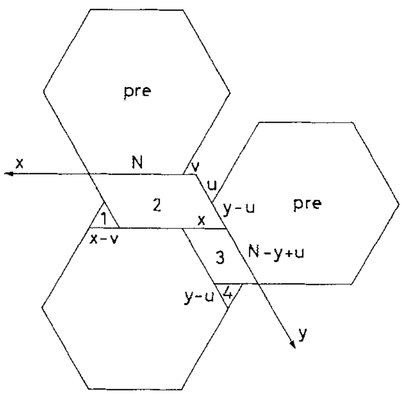

Fig. 21. The two preceding hexagons (marked pre) placed as case VI and the next hexagon placed in position 4 (see Fig. 19). The position of the next hexagon is given by x and y coordinates as shown. The new uncoverable area is divided into parts numbered in accordance with the numbering on Fig. 20.

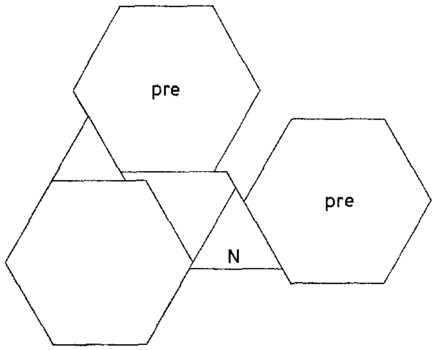

Fig. 22. The two preceding hexagons (marked pre) placed as case VI and the next hexagon placed in position 9 (see Fig. 19). It is easily seen that the uncoverable area exceeds $\frac{1}{2}N^2$.

which is the same result as for position 1; in fact one obtains the same result for position 1–8.

In Fig. 22 is shown an example of position 9. One obtains the result

$$a' \geqslant \tfrac{1}{2}N^2 \tag{A.8}$$

a result which actually holds for all the remaining cases. If there is no hexagon with its upper right corner inside the heavy line shown in Fig. 23, one can surely place a hexagon as indicated. The total number of positions inside this area is less than $5N^2$.

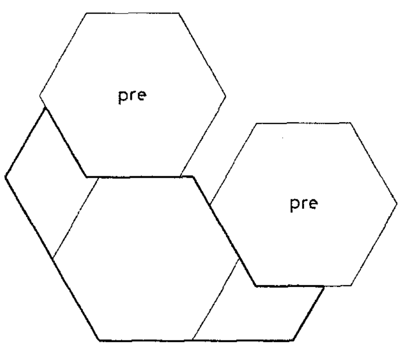

Fig. 23. If no hexagon has been placed with its upper right corner inside the heavily bordered region, a hexagon can be placed as indicated.

Summing over all positions of the hexagon, one then obtains

$$\begin{aligned}
&\{\exp[-\tfrac{1}{2}\alpha(u^2 + v^2)]\}\{1 - \exp[-\alpha(N + u - v)]\}^{-1} \\
&\qquad \times \{1 - \exp[-\alpha(N + v - u)]\} + 5N^2 \exp(-\alpha N^2/2) \\
&\leqslant [\exp(-\tfrac{1}{2}\alpha)]\{1 - \exp[-\alpha(N - 1)]\}^{-1}\{1 - \exp[-\alpha(N + 1)]\}^{-1} \\
&\qquad + 5N^2 \exp(-\alpha N^2/2) \\
&\quad = \delta
\end{aligned}$$

As one continues to place the inner hexagons one will eventually reach a situation where it is clear that the contour is finished. If there remain hexagons, i.e., if we have placed a number less than m, the remaining hexagons cannot be placed; this corresponds to $a_k = 0$ and $\sum b_k = 1$.

ACKNOWLEDGMENT

The authors want to thank Professor E.H. Lieb for valuable discussions.

REFERENCES

1. R. E. Peierls, *Proc. Camb. Phil. Soc.* **32**:477 (1936).
2. R. Griffiths, *Phys. Rev.* **136A**:437 (1964).
3. R. L. Dobrushin, *Theor. Prob. and Appl.* **10**(2):209 (1965); in *Vth Symp. Mathematical Stat. and Prob.*, Vol. 3 (1967), p. 73.
4. F. A. Berezin and J. G. Sinai, *Trans. Moscow Math. Soc.* **17**:219 (1967); J. Ginibre, A. Grossmann, and D. Ruelle, *Commun. Math. Phys.* **3**:187 (1966); J. L. Lebowitz and G. Gallavotti, *J. Math. Phys.* **12**:7 (1971).
5. D. Ruelle, *Phys. Rev. Lett.* **27** (1971), 1040.
6. R. L. Dobrushin, *Funct. Anal. Appl.* **2**(4):44 (1968) [English transl. **2**:302 (1968)].
7. O. J. Heilmann, *Lett. Nuovo Cimento* **3**:95 (1972).
8. L. K. Runnels and L. L. Combs, *J. Chem. Phys.* **45**:2482 (1966).
9. J. Orban and A. Bellemans, *J. Chem. Phys.* **49**:363 (1968).
10. L. K. Runnels, J. R. Craig, and H. R. Streiffer, *J. Chem. Phys.* **54**:2004 (1971).