Existence of a Phase Transition in a Continuous Classical System

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A rigorous proof is given for the existence of a phase transition in the Widom-Rowlinson model in two dimensions.

A new model exhibiting a phase transition has received much attention recently. It is a classical continuous model with two kinds of particles, A and B. The only interaction is a hard-core exclusion: An A particle and a B particle cannot be closer than a distance R; there is no restriction on the distance between two A or two B particles. When the activities of the two kinds of particles are equal to z, and z is sufficiently large, one expects that two phases coexist: The one consists predominantly of A particles; the other, of B particles. If the B particles are invisible, the resulting system of A particles yields a model for vapor-liquid phase transitions, which has been introduced and discussed by Widom and Rowlinson.[1]

Until now, a rigorous proof for the presence of a phase transition in the above models was missing although Gallavotti and Lebowitz[2] were able to handle the analogous lattice problem. In the present note a modification of the Peierls argument[3] is used to give a proof for the continuous case in two dimensions. Higher dimensions can be handled in a similar manner.

The main point in the Peierls argument is the computation of the probability of certain contours on a lattice. We use a lattice formed of $d\times d$ "little" squares with $d=R/3\sqrt{2}$ (so that R is the diagonal of a $3d\times 3d$ square). The box containing the particles is a rectangle composed of N little squares. We introduce the boundary condition that no B particle is allowed in the two rows or columns adjacent to the sides of the rectangle. If a B particle is contained in a little square, we shade the $3d\times 3d$ square centered on this little square. The boundary of the union of the shaded areas is a contour Γ, a polygon consisting of various connected components. We write Γ as a union of disjoint pieces $\gamma_1, \cdots, \gamma_n$: Each piece is a smallest set of connected components of Γ such that, if two connected components have a distance less than R, they belong to the same γ_i.

If the polygon γ_i has length ld and consists of c connected components, it can be covered by a k-step lattice walk, where $k\leqslant l+12(c-1)$. The walk starts at one point and goes back to it; it covers once the γ_i and twice the paths between the different connected components of γ_i (each path can be chosen of length $\leqslant 6d$, and, at most, $c-1$ such paths are needed). A connected component of γ_i has length of at least 12; therefore, $l\geqslant 12c$ and

$$k<l+12c\leqslant 2l.$$

An upper bound to the number $n(l)$ of polygons of length ld which may occur as pieces γ_i in the decomposition of Γ is given by

$$n(l)<N\times 3^{2l} \tag{1}$$

The factor N corresponds to the choice of an initial point for a lattice walk covering the polygon; the factor $3^{2l}>3^k$, to the number of such walks with given initial point.

We prove now that the grand canonical probability $p(\gamma)$ for a given polygon γ to occur as one of the pieces $\gamma_1, \cdots, \gamma_n$ of Γ satisfies

$$p(\gamma)\leqslant \exp(-\tfrac{1}{2}ld^2z). \tag{2}$$

We assume that γ is an outer piece of Γ, i.e., there is a path coming from infinity, reaching a point of γ without crossing Γ. We say that a point x (not on γ) is interior to γ if a path coming from infinity crosses γ an odd number of times before reaching x (see Fig. 1). Let X be any configuration of A and B particles producing a contour Γ

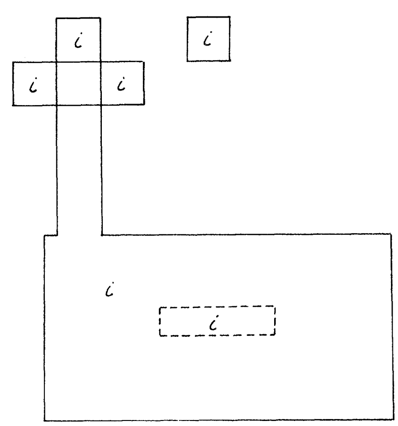

FIG. 1. γ_1 (solid line) and γ_2 (dashed line) are two pieces of Γ. The interior of γ_1 is marked i.

which contains the piece γ, and suppose that γ is an outer piece. From X we obtain a class X^* of configurations as follows: (a) All A particles interior to γ are changed to B particles, and vice versa. (b) Let G be a band around γ consisting of all little squares which have one side or corner on γ; A particles are placed in an arbitrary manner in G. Clearly, step (a) leads to permissible configurations. So does step (b) because, after (a), any B particle must be separated from G by a piece of the contour Γ different from γ, and therefore at a distance $>R$ from G. Using an idea which goes back ultimately to Peierls and Griffiths,[2,3] we write

$$p(\gamma)=Z(\gamma)/Z \leqslant Z(\gamma)/[Z(\gamma)Z_G]=1/Z_G.$$

Here Z is the grand partition function; $Z(\gamma)$ is the contribution to Z of the configurations such that Γ contains γ as an outer piece. Finally Z_G is the grand partition function for A particles in G, which is equal to the grand partition function for free particles at activity z in G. The area of G is larger than $\frac{1}{2}ld^2$. (On each side of γ one can construct a square with this side as diagonal; these squares have area $\frac{1}{2}d^2$, are contained in G, and do not overlap.) Therefore

$$Z_G \geqslant \exp(\tfrac{1}{2}ld^2z),$$

proving (2).

We can now estimate the expectation value of the number S of little squares in the shaded area surrounded by Γ: S is not greater than the sum of the numbers of little squares interior to the outer pieces which occur in the decomposition of Γ. We write $l=2m+2n$, where $2md$ and $2nd$ are the length of the horizontal and vertical part of γ, respectively. Then, using (1) and (2), we obtain

$$\langle S\rangle \leqslant \textstyle\sum_\gamma p(\gamma)\times(\text{number of little squares interior to }\gamma)$$

$$\leqslant \sum_{m=3}^{\infty}\sum_{n=3}^{\infty} mn3^{4(m+n)}\exp[-(m+n)d^2z]$$

$$=N[\sum_{m=3}^{\infty} m\exp m(4\ln 3-d^2z)]^2.$$

The probability that a little square contains at least one B particle is thus bounded by

$$N^{-1}\langle S\rangle \leqslant \varphi(d^2z-4\ln 3),$$

where $\varphi(t)$ is finite for $t>0$ and tends to 0 for $t\to\infty$.

Let T be the total number of little squares occupied by at least one particle of any kind. We shall show that for sufficiently large z

$$N^{-1}\langle T\rangle > 1/484.$$

We denote by Z_- the contribution to the grand partition function Z of the configurations such that $T<N/242$. We notice that at least $N-(11)^2T$ little squares are at a distance $>R$ from any particle. Therefore,

$$Z \geqslant Z_-[\exp(zd^2)]^{N/2} \geqslant \tfrac{1}{2}Z_-$$

for sufficiently large z. Finally,

$$\frac{1}{N}\langle T\rangle \geqslant \frac{1-Z_-/Z}{2(11)^2} \geqslant \frac{1}{4(11)^2}=\frac{1}{484}.$$

If there is only one pure thermodynamic phase, the probability that a little square contains at least one A particle is equal to the probability that this little square contains at least one B particle, and therefore

$$2N^{-1}\langle S\rangle \geqslant N^{-1}\langle T\rangle.$$

We have thus proved that there is more than one pure thermodynamic phase if

$$2\varphi(d^2z-4\ln 3)<1/484,$$

and we know that this is the case for sufficiently large z.

Remark.—The existence of a phase transition can be proven in the same manner as above if a "small" hard core is introduced for the A-A and B-B interactions. It suffices to take the corresponding distance strictly less than d (for instance $R/5$).

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