The Direct Correlation Function of an Inhomogeneous Lattice Gas

Shinn-Tyan Wu

Battelle Memorial Institute, 505 King Ave., Columbus, Ohio and
Department of Metallurgy and Material Science, Cheng Kung University Tainan, Taiwan 700

(Received July 21, 1975)

Two terms beyond mean field approximation are obtained for the direct correlation function of a general lattice gas. Use is made of Mayer’s cluster expansion extended to inhomogeneous systems. The resulting expression is as simple and as readily applicable as the mean field approximation. It can deal with ferromagnetic or antiferromagnetic interaction, below or above transition temperature, with or without far neighbor interactions. Specializing to an Ising model its accuracy is better than that obtained by the Bethe method. A new high temperature expansion for lattice gases is discovered. In the new expansion scheme only doubly-connected graphs need to be considered. As far as the counting of graphs is concerned the present method is the simplest among all the existing series expansions for lattice gas.

§ 1. Introduction

During their study of critical opalescence Ornstein and Zernike realized that the direct correlation function is a better physical variable to work with than the pair correlation function when fluctuations become substantial. Despite its importance in describing scattering experiments a practical method of calculating the direct correlation function is yet to be devised. The integral equation approaches can, with the aid of computers, provide useful numerical information. However, the mathematical complexity and the considerable numerical work obscure much of their physical contents. Also it is difficult, if not impossible, to go to higher orders. Thus a simple, systematic method of calculating the direct correlation function for a condensed system is highly desirable. For a continuous fluid this is not feasible at present. Hence we turn to lattice gas model in the hope that some of results may be similar to those of a real fluid. Furthermore it is well known that lattice gas model can be well approximated by some real systems. Hence it seems justified as a preliminary step to study the direct correlation function of a lattice gas model.

There are numerous methods for calculating the thermodynamic variables of lattice gas model yet none of them are suitable for our purpose. Instead we found that Mayer’s cluster series for an inhomogeneous system can be used
to obtain a high temperature series expansion for the internal field (defined by Eq. (4)) of an inhomogeneous lattice gas. Differentiating the series with respect to density function\textsuperscript{131} a high temperature series for the direct correlation function results. The first four terms are worked out explicitly. The result is simple mathematically and has a wide range of applicability. It can deal with ferromagnetic or antiferromagnetic interaction, above or below transition temperature. The interaction beyond nearest neighbors causes no additional complication.

In § 2 the virial series for an imperfect, inhomogeneous gas are introduced. The basic concept of the self potential of a small cluster is explained. Then it is shown that the evaluation of self potential is equivalent to solving a set of algebraic equations. In § 3 the self potentials of a single lattice site and a pair are worked out explicitly. Then we show, with the aid of self potential, that the internal field at a lattice site can be decomposed into contributions from various clusters. In § 4 the direct correlation function is obtained. In the last section we conclude. We derive, in Appendix A, the third order term. Appendix B contains the proof of a theorem.

§ 2. The self potential of a cluster of lattice sites\textsuperscript{121}

2.1 Mayer's virial series for an inhomogeneous gas

If a system of gas is ideal but under the influence of an external potential \( V_1(\mathbf{r}) \), then the density of gas satisfies the following equation:

\[
\rho(\mathbf{r}) = \frac{1}{v_0} \exp\{-\beta [V_1(\mathbf{r}) - u]\},
\]

where

\[
v_0 = \left( \frac{\hbar^2}{2\pi mk_B T} \right)^{3/2},
\]

\( u \) is chemical potential. If a two-body potential, \( V_2(\mathbf{r}, \mathbf{r}') \), is switched on then the density function satisfies

\[
\rho(\mathbf{r}) = \frac{1}{v_0} \exp\{-\beta [V_1(\mathbf{r}) + V_{\text{int}}(\mathbf{r}) - u]\},
\]

where the internal field \( V_{\text{int}}(\mathbf{r}) \) originates from \( V_2(\mathbf{r}, \mathbf{r}') \). It has the following graphical expansion:\textsuperscript{120,19}

\[
-\beta V_{\text{int}}(\mathbf{r}) = \bigoplus \bigoplus \bigoplus \bigoplus + \cdots,
\]

where the full circle stands for \( \mathbf{r} \) at which the internal field is being evaluated. Each open circle stands for a dummy vector to be weighted by \( \rho(\mathbf{r}) \) and then integrated. The bonds stand for Mayer's \textit{f} function whose product is the integrand.
The Direct Correlation Function of an Inhomogeneous Lattice Gas

If $V_1(r)$ is switched off then $\rho(r)$ becomes a constant function and the familiar virial series is recovered. Adapting the virial series to a lattice gas we have the following:

$$\rho(i) = \exp\{-\beta[V_1(i) + V_{\text{int}}(i) - u]\}$$

(4)

and

$$-\beta V_{\text{int}}(i) = \sum_{\substack{k \in \mathcal{L} \setminus \mathcal{Q} \setminus \{i\}}} f(i, j) f(j, k) f(k, i),$$

(5)

Now $i$ stands for a lattice vector. The open circle stands for dummy variables to be weighted by $\rho(i)$ and summed over lattice sites. For example

$$f(i, j) = \exp[-\beta V_2(i, j)] - 1.$$  

(7)

For lattice gas one assumes that $V_2(i, j) = 0$ for $i = j$.

As a result one lattice site can at most accommodate one particle.

2.2 The self potential of a cluster

Let a cluster, denoted by $\mathcal{Q}$, stand for a subset of the lattice. Then the density function can be split into two parts:

$$\rho(i) = \rho^*(i) + \rho'(i),$$

(9)

where $\rho^*(i)$ is equal to $\rho(i)$ if $i$ belongs to $\mathcal{Q}$ and zero otherwise. Then $\rho'(i)$ is equal to $\rho(i)$ for $i$ outside $\mathcal{Q}$ and vanishes inside. Thus

$$\rho^*(i) = \begin{cases} \rho(i) & \text{if } i \in \mathcal{Q}, \\ 0 & \text{if } i \notin \mathcal{Q}. \end{cases}$$

(10)

In the virial series of (5) the internal field is expanded in power series of $\rho(i)$. Using (9) the series can be turned into a power series in $\rho'(i)$. The coefficients depend on $f(i, j)$ and $\rho^*(i)$. The zeroth order is independent of $\rho'(i)$ and depends only on the particle distribution inside $\mathcal{Q}$. This portion of the internal field of $\mathcal{Q}$ is defined as the self potential of $\mathcal{Q}$ and denoted by $V_s(i)$. It is independent of the particle distribution outside $\mathcal{Q}$. Thus it can be evaluated by considering

\footnote{This can be understood better by considering a Taylor series $f(x) = ax + bx^2 + cx^3 + \cdots$. If $x = y + z$ then $f(x) = A + a'z + b'z^2 + c'z^3 + \cdots$, where the zeroth order, $A$, is given by $ay + by^2 + cy^3 + \cdots$. It is independent of $z$.}
a reference system in which the particle distribution outside $\Omega$ is "switched off", i.e., a system with a particle distribution equal to $\rho^*(i)$. Then

$$V_s(i) = V_{\text{int}}^*(i) \quad \text{for} \quad i \in \Omega,$$

(11)

where the quantities with asterisk are those of the reference system. Since $\rho^*(i) = 0$ if $i$ is outside $\Omega$ its external potential, $V^*_1(i)$, should be infinite because of Eq. (4). Thus the reference system is contained in $\Omega$. If $\Omega$ has only a finite number of lattice sites the grand partition function of the reference system expanded in power series of activity, is truncated. It becomes a polynomial because each lattice site can at most accommodate only one particle. Thus

$$Z^* = \sum_{m=0}^{\infty} \frac{1}{m!} \sum_{i_1}^{\infty} \sum_{i_2}^{\infty} \cdots \sum_{i_m}^{\infty} \exp \{-\beta [U^*_m(i_1, i_2, \ldots, i_m) - mu]\},$$

(12)

where the potential energy $U^*_m$ is given by

$$U^*_m(i_1, i_2, \ldots, i_m) = \sum_{s=1}^{m} V^*_1(i_s) + \sum_{i_s < i_t} V_1(i_s, i_t).$$

(13)

The density function $\rho^*(i)$ satisfies

$$Z^* \rho^*(i) = -\frac{\partial Z^*}{\partial \beta V^*_1(i)}, \quad i = 1, 2, \ldots, n.$$  

(14)

Let an activity function be defined by

$$z^*(i) = \exp \{-\beta [V^*_1(i) - u]\}.$$  

(15)

Then we can regard (14) as an equation for solving $z^*$ in terms of $\rho^*$. There are $n$ algebraic equations for $n$ unknowns. If the solution is found then from (4) the internal field $V_{\text{int}}^*(i)$ is obtained as a function of $\rho^*$. This combined with (11) gives us $V_s(i)$ for $\Omega$. On the other hand $-\beta V_s(i)$ can formally be evaluated by summing the virial series of (5) while restricting all the open circles to lie inside $\Omega$. Thus if the dummy variables of virial series are confined within $\Omega$ the series can be summed to infinite order by solving the algebraic equation of (14). This is an important point to be emphasized. Although (12) \text{\sim} (14) was discovered long ago its significance as a method of summing subseries to infinite order for an inhomogeneous system is new.

§ 3. The new high temperature series

When $\Omega$ contains only one lattice site the self potential is easily obtained from (14) as

$$-\beta V_s(i) = \ln [1 - \rho(i)],$$

(15)

when there is a pair $(1, 2)$ in $\Omega$ then solving for $h(1, 2)$ with the aid of

$$h(1, 2) = [\rho(1) - \rho(1)\rho(2) - h(1, 2)] [\rho(2) - \rho(1)\rho(2) - h(1, 2)] \times f(1, 2).$$

(17)

Equation (17) follows from (A\cdot 8) and (A\cdot 9) if one sets $\rho(3) = h(13) = h(23) = h(123) = 0$ or equivalently $z(3) = 0$. This reduces the triplet problem of Appendix A to a pair problem.
The Direct Correlation Function of an Inhomogeneous Lattice Gas

one obtains

\[-\beta V_z(1) = \ln[1 - \rho(1)] + \ln\left\{1 + \left[\rho(2) - \frac{h(1,2)}{1 - \rho(1)}\right]f(1,2)\right\}.\]  

(18)

The left-hand side is the sum of virial series restricting all the dummy variables to \((1,2)\). The first term on the right-hand side is the sum of the series restricting all the dummy variables to the site 1. Therefore the second term is the sum of virial series restricting all the dummy variables to within \((1,2)\), in addition, at least one of the dummy variables should be at site 2. Thus it is natural to consider this term as the net contribution to the internal field at site 1 from the pair \((1,2)\). Summing all the contributions from pairs we obtain an approximation to the internal field:

\[-\beta V_{\text{int}}(1) = \ln[1 - \rho(1)] + \sum_{i \neq 1} \ln\left\{1 + \left[\rho(i) - \frac{h(1,1)}{1 - \rho(1)}\right]f(1,i)\right\}.\]  

(19)

To go to higher orders we have to evaluate, using (14), the self potentials of larger clusters. Afterwards the contributions of subclusters are substracted to obtain net contributions. In Appendix B\textsuperscript{20} we show that only doubly-connected clusters need to be considered.\textsuperscript{a)1} Thus to the next order Eq. (14) is to be solved explicitly hence we resort to high temperature series expansion. Since a triangle has three \(f\)-bonds it is third order in \(\beta V_z\). Hence the first three terms are obtained from (17) \(-\sim\) (19) as

\[-\beta V_{\text{int}}(1) = \ln[1 - \rho(1)] - \beta \sum_{i \neq 1} \rho(i) V_z(i,1)\]

\[+ \left[\frac{1}{2} - \rho(1)\right] \sum_{i \neq 1} \rho(i) [1 - \rho(i)] \beta^2 V_z^2(1,i).\]  

(20)

First order is the mean field and second order is the fluctuation of internal field. For an Ising model a result equivalent to (20) has been obtained using a very different method.\textsuperscript{14} The next order is calculated in Appendix A. Thus a new high temperature series expansion is constructed. The combinatorial problem is much simplified because only doubly-connected clusters need to be counted. In passing it is interesting to observe that the pair approximation is exact for a Bethe lattice with nearest neighbor interaction. This follows immediately from the theorem of Appendix B.

\[
\text{§ 4. The formula for direct correlation function}
\]

The direct correlation function \(C(i,j)\) is related to the internal field\textsuperscript{22} through

\[\text{\footnotesize \cite{21}}\]

\[\text{\footnotesize We give a different proof than that of Ref. 21 because:}\]

\[\text{\footnotesize (a) We deal with inhomogeneous systems while in Ref. 21) only homogeneous systems are considered.}\]

\[\text{\footnotesize (b) With very slight modification our proof is equally applicable to multi-component systems with multi-site interaction.}\]

\[\text{\footnotesize (c) Our proof is very much shorter yet clearer than the old ones.}\]
the equation
\[ C(1, 2) = \frac{\partial [-\beta V_{\text{int}}(1) \beta^2 V_t^*]}{\partial \rho(2)}. \]  

Combining (21), (20) and (A·1) we obtain
\[
C(1, 1) = \frac{-1}{1-\rho(1)} - \sum_{i \neq 1} \rho(i) [1-\rho(i)] \beta^2 V_t^*(1, i) \\
+ [1-2\rho(1)] \sum_{i \neq 1} \rho(i) [1-\rho(i)] [1-2\rho(i)] \\
\times \beta^3 V_t^*(i, 1) \\
+ 2 \sum_{i \neq j \neq k} \rho(j) [1-\rho(j)] \rho(k) [1-\rho(k)] \\
\times \beta^3 V_t(1, j) V_t(j, k) V_t(k, 1). \tag{22}
\]

If 1 and 2 denotes different lattice sites then
\[
C(1, 2) = -\beta V_t(2, 1) \\
[1-\rho(1)] [1-2\rho(2)] \beta^3 V_t^*(1, 2) \\
- [\frac{1}{6} - \rho(1) + \rho^3(1)] [1-6\rho(2) + 6\rho^2(2)] \beta^3 V_t^*(1, 2) \\
- 2[1-2\rho(1)] [1-2\rho(2)] \sum_{k \neq 1, \rho \neq 2} \rho(k) [1-\rho(k)] \\
\times \beta^3 V_t(1, 2) V_t(2, k) V_t(k, 1). \tag{23}
\]

The above two equations are the most important results of the present investigation. Notice that we have not said anything about \( V_t(i, j) \) other than (8). Hence it is equally applicable to ferromagnetic or antiferromagnetic, long-range or short-range interactions. Also nothing is mentioned about \( \rho(i) \) hence it is equally applicable to systems with or without long-range order. To author's knowledge the only approximation which is as general and as simple as the present one is the mean field approximation.

Since ours is an approximate method it is necessary to assess its accuracy. We do this by calculating the transition temperature \( T_c \) for Ising model. In this case the system is homogeneous so that \( \rho(i) \) is independent of \( i \). It is known that \( \rho \) is equal to one half at critical point because of the particle-hole symmetry of a homogeneous lattice gas. The relevant equation for \( T_c \) is\(^b\)
\[
1-\rho \sum_i C(i, 1) = 0, \tag{24}
\]
where \( C(i, 1) \) depends only on the relative position between \( i \) and 1 because the system is homogeneous. From the above three equations we obtain (setting \( \rho(i) = 1/2 \))
\[
0 = \frac{1}{q} + x + x^2 + \left( \frac{2}{3} - \frac{6}{q} \alpha \right) x^3 \tag{25}
\]
The Direct Correlation Function of an Inhomogeneous Lattice Gas

with $x$ defined by

$$x = \beta \frac{V}{4}. \quad (26)$$

Here $V$ is the interaction energy between a pair of particles sitting next to each other. $q$ is the number of nearest neighbors of the lattice. $\alpha$ is the number of triangles per lattice site. For a triangular lattice

$q = 6$ and $\alpha = 2$

(25) is solved to obtain the critical temperature

$$k_B T_c = -4 \left( \frac{V}{4} \right). \quad (27)$$

This can be compared with the critical temperature from mean field approximation

$$k_B T_m = -6 \left( \frac{V}{4} \right),$$

that from the Bethe-Peierls approximation

$$k_B T_{BP} = -4.93 \left( \frac{V}{4} \right),$$

and the exact result

$$k_B T_{ex} = -3.64 \cdots \left( \frac{V}{4} \right).$$

Thus the present result is substantially more accurate than mean field approximation and, to a less degree, than the Bethe-Peierls approximation. Because of its simplicity, generality and accuracy our result should be of considerable value for practical purposes.

§ 5. Discussion

Tarko and Fisher have performed accurate calculation of the direct correlation functions for Ising models. Admittedly for the specific models studied by them our results are less accurate than theirs. However their method lacks the simplicity and generality of ours. These we consider as more important for practical purposes, because it is very improbable that any real system can be described exactly by a specific Ising model. Therefore no matter how accurate the model calculations are they can never fit the experimental results exactly. Consequently for practical applications a method with simplicity, generality and reasonable accuracy is more useful. This seems to be what we have obtained. From (20) one sees that the present result is a beyond-mean-field approximation. Elliott and Marshall also used the Bethe-Peierls approach to evaluate the pair correlation function of Ising model. Their result is less accurate than ours, as was discussed.
below (27), even though the mathematics is much more involved. Also the
generality of mean field approximation is completely lost. Our result seems to be the
only improvement, without losing simplicity and generality, of the widely used
mean field approximation. However it should be noted that our series is too short
to estimate the values of critical indecies.
It is also interesting to notice that in our graphical expansion only the doubly-
connected graphs are needed in contrast to conventional approaches. This simplifi-
es tremendously the combinatorial problem of counting graphs. However, to de-
velop the present approach into a powerful method of calculating long series for
Ising model a more efficient way of dealing with small clusters is needed. If
this is found then a longer series than they are currently available may become
feasible.

Appendix A

The third order internal field is given by

\[- \beta V_{int}^{(3)}(1) = - \left[ \frac{1}{\beta} - \rho(1) + \rho^2(1) \right] \]

\[\times \sum_{i=1} \rho(i) \left[ 1 - \rho(i) \right] \left[ 1 - 2 \rho(i) \right] \beta V_z(i, 1) \]

\[- \left[ 1 - 2 \rho(1) \right] \sum_{1 \leq j \neq k} \rho(j) \left[ 1 - \rho(j) \right] \rho(k) \left[ 1 - \rho(k) \right] \]

\[\times \beta^2 V_z(1, j) V_z(j, k) V_z(k, 1). \quad (A·1)\]

The first term comes from pairs and the last from triplets. It can be easily shown
to reproduce the known results on Ising model (Wu 1972). The pair contribution
is found by solving (17) to second order

\[h(1, 2) = \rho(1) \left[ 1 - \rho(1) \right] \rho(2) \left[ 1 - \rho(2) \right] f(1, 2) \]

\[- \left[ \rho(1) + \rho(2) - 2 \rho(1) \rho(2) \right] f^2(1, 2) \quad (A·2)\]

and expanding (19) in powers of \( \beta V_z \). To evaluate the triplet contribution it is
necessary to consider (14) for a cluster of three lattice sites. Denoting them by
(1, 2, 3) it is easy to obtain

\[\frac{1}{Z} = 1 - \sum_{i=1} \rho(i) + \sum_{i<j} \rho(i, j) - \rho(1, 2, 3), \quad (A·3)\]

where \( \rho(i, j) \) and \( \rho(1, 2, 3) \) are defined by

\[\frac{z(1)}{Z} = \rho(1) - \rho(1, 2) - \rho(1, 3) + \rho(1, 2, 3) \quad (A·4)\]

and others

\[\frac{z(1)z(2)}{Z} \sigma(1, 2) = \rho(1, 2) - \rho(1, 2, 3) \quad (A·5)\]
The Direct Correlation Function of an Inhomogeneous Lattice Gas

and others

\[ \frac{z(1)z(2)z(3)}{Z} e(1, 2)e(1, 3)e(2, 3) = \rho(1, 2, 3) \quad (A·6) \]

“and others” means the other two equations which can be obtained from the written one by permuting \((1, 2, 3)\). Let \(h(1, 2, 3)\) and \(h(i,j)\) be defined by

\[ h(1, 2, 3) = \rho(1, 2, 3) - \rho(1)\rho(2, 3) - \rho(2)\rho(3, 1) - \rho(3)\rho(1, 2) + 2\rho(1)\rho(2)\rho(3), \quad (A·7) \]

\[ h(i,j) = \rho(i,j) - \rho(i)\rho(j). \quad (A·7)' \]

It is easy to obtain the following equations:

\[ \frac{z(1)z(2)}{Z} f(1, 2) = [1 - \rho(3)]h(1, 2) - h(1, 3)h(2, 3) - [1 - \rho(3)]h(1, 2, 3), \quad (A·8) \]

\[ \frac{z(1)z(2)}{Z} f(1, 2) = [1 - \rho(3)]f(1, 2) - h(1, 3)h(2, 3) - [1 - \rho(3)]h(1, 2, 3), \quad (A·9) \]

\[ \frac{1}{Z} = [1 - \rho(1)][1 - \rho(2)][1 - \rho(3)] + \sum' [1 - \rho(1)]h(2, 3) + h(1, 2, 3), \quad (A·10) \]

where \(\sum'\) denote summing over permutations of \((1, 2, 3)\). Using \((A·6)\) and the above four equation and after some lengthy yet straight-forward algebra we can obtain

\[ \frac{1}{Z}z(1)z(2)z(3) [f(1, 2)f(2, 3)f(3, 1) + \sum' f(1, 2)f(2, 3)] \]

\[ = [1 - \rho(1)][1 - \rho(2)][1 - \rho(3)]h(1, 2, 3) + 2\sum'[1 - \rho(1)][1 - \rho(2)]h(1, 3)h(2, 3) \]

\[ - h(1, 2, 3)\sum'[1 - \rho(1)]h(2, 3) + 2h(1, 2)h(2, 3)h(3, 1) + h(1, 2, 3)h(1, 2, 3). \quad (A·11) \]

From \((A·8)\), \((A·9)\) and \((A·11)\) a high temperature series solution is easily generated. The leading order of \(h(123)\) is

\[ h(1, 2, 3) = \rho(1)[1 - \rho(1)]\rho(2)[1 - \rho(2)]\rho(3)[1 - \rho(3)] \times \sum'[1 - \rho(1)]f(1, 2)f(1, 3), \quad (A·12) \]

also the following term should be added to the r.h.s. of \((A·2)\):

\[ \rho(1)[1 - \rho(1)]\rho(2)[1 - \rho(2)]\rho(3)[1 - \rho(3)]f(1, 2)f(1, 3). \quad (A·13) \]

The internal field is easier to find in the following way: Let us write

\[ Z = Y + z(1)Y', \quad (A·14) \]
where
\[ Y = 1 + z(2) + z(3) + z(2)z(3)e(2, 3) \]  
\[ \text{(A·15)} \]
and
\[ Y' = 1 + z(2)e(2, 1) + z(3)e(3, 1) + z(2)z(3)e(2, 3)e(2, 1)e(3, 1). \]  
\[ \text{(A·16)} \]
From (14) it follows that
\[ \rho(1) = \frac{z(1)Y'/Y}{1 + z(1)Y'/Y}. \]
Therefore we obtain
\[ \rho(1) = z(1)[1 - \rho(1)]Y'/Y. \]  
\[ \text{(A·17)} \]
Hence
\[ -\beta V_{\text{int}}(1) = \ln[1 - \rho(1)] + \ln(Y'/Y). \]  
\[ \text{(A·18)} \]
It is easy to show, using (A·8)∼(A·10), that
\[ \frac{Y'}{Y} = \frac{Y'/Z}{Y/Z} = \left\{ 1 + \left[ \rho(2) - \frac{h(1, 2)}{1 - \rho(1)} \right] f(1, 2) \right\} \left\{ 1 + \left[ \rho(3) - \frac{h(1, 3)}{1 - \rho(1)} \right] f(1, 3) \right\} \]
\[ + \left[ \frac{h(2, 3) - \frac{h(1, 2, 3)}{1 - \rho(1)} - \frac{h(1, 2)h(1, 3)}{1 - \rho(1)^2}}{1 - \rho(1)} \right] f(1, 2)f(1, 3). \]  
\[ \text{(A·19)} \]
From (A·19), (A·13) and (A·2) we obtain $-\beta V_{\text{int}}(1)$ to third order. Subtracting pair contributions we obtain the net contribution from a triplet as
\[ [1 - 2\rho(1)]\rho(2)[1 - \rho(2)]\rho(3)[1 - \rho(3)]f(1, 2)f(2, 3)f(3, 1). \]
Thus (A·1) is established.

**Appendix B**

A cluster $\mathcal{Q}$, which is not doubly-connected, contains at least a connecting site 0 the removal of which will separate $\mathcal{Q}$ into two disconnected subclusters $\mathcal{Q}_1$ and $\mathcal{Q}_2$ such that
\[ V_t(\alpha, \gamma) = 0, \]  
\[ \text{(B·1)} \]
where and hereafter $\alpha$ stands for a lattice site within $\mathcal{Q}_1$, while $\gamma$ for one within $\mathcal{Q}_2$. The grand partition function $Z$ of $\mathcal{Q}$ can be written as
\[ Z = Z_1Z_2 + z(0)G_1G_2. \]  
\[ \text{(B·2)} \]
$Z_t$ is the grand partition function of $\mathcal{Q}_t$. $G_t$ is the grand partition function of $\mathcal{Q}_t$ after the external field $V_t(j)$ is replaced by $V_t(j) + V_t(j, 0)$. $z(0)$ was defined by (15). Then from (14) we obtain
\[ \rho(0) = [1 - \rho(0)]z(0)\frac{G_1G_2}{Z_1Z_2} \]  
\[ \text{(B·3)} \]
The Direct Correlation Function of an Inhomogeneous Lattice Gas

Using (14) again we obtain

\[ \rho(\alpha) = -\frac{\partial}{\partial \beta V_i(\alpha)} \ln Z_i - \rho(0) \frac{\partial}{\partial \beta V_i(\alpha)} \ln \frac{G_i}{Z_i} \]  

(B.4)

and a corresponding formula for \( \rho(\gamma) \). Since \( Z_i \) and \( G_i \) depend only on \( z(\alpha) \) and not on \( z(\gamma) \) we can use (B.4) to solve \( z(\alpha) \) in terms of \( \rho(\alpha) \) and \( \rho(0) \) and then, as was indicated below (15), obtain \( -\beta V_i(\alpha) \) of \( \Omega \) as function of \( \rho(\alpha) \) and \( \rho(0) \). Thus it is shown that the self potential of \( \Omega \), within the subcluster \( \Omega_i \), depends only on \( \rho(\alpha) \) and \( \rho(0) \) and not on \( \rho(\gamma) \). Now, in evaluating the contribution of \( \Omega \) to the internal field at \( \alpha \), the contributions from all the subclusters of \( \Omega \) have to be subtracted from \( V_i(\alpha) \). The resulting net contribution involves density function at lattice sites of \( \Omega_2 \). This net contribution is clearly zero in the present case because \( -\beta V_i(\alpha) \) is independent of \( \rho(\gamma) \). Similar reasoning applies to \( \gamma \)-sites. For the connecting site the self potential from (B.3) is given by

\[ -\beta V_i(0) = \ln[1 - \rho(0)] + \ln \frac{G_i}{Z_i} + \ln \frac{G_2}{Z_2}. \]  

(B.5)

The first term on the right-hand side depends on \( \rho(0) \), the second on \( \rho(\alpha) \) and \( \rho(0) \), the last on \( \rho(\gamma) \) and \( \rho(0) \). Thus there is no cross term involving both \( \rho(\alpha) \) and \( \rho(\gamma) \). Hence the net contribution from \( \Omega \) to \( V_{\text{int}}(0) \) is also zero.

References

9) C. Domb, Adv. in Phys. 9 (1960), 149.
11) G. Stell, Ref. 6) pp. II-201.
13) G. Stell, Ref. 6) pp. II-203.
16) R. Brout, Phase Transition (Benjamin, New York, 1965).