PHYSICAL REVIEW B VOLUME 16, NUMBER 9 1 NOVEMBER 1977

Application of the cluster variation method to the fcc Ising ferromagnet*

Shiv Kumar Aggarwal and Tomoyasu Tanaka

Department of Physics, Clippinger Research Laboratories, Ohio University, Athens, Ohio 45701

(Received 21 January 1977)

The molecular-field-type approximations when applied to the Ising ferromagnet yield inaccurate and sometimes incorrect behaviors at the transition temperature. We have advanced the mathematical approximation of the cluster variation method, which is the generalization of the molecular-field-type approximation, a few more steps so that the four- and six-body correlations are taken into account. Some remarkable features became apparent from preliminary calculations for fcc Ising ferromagnets: (i) The transition temperature is predicted within a few percent of the exact value. T_c(4-body) = 0.835 45, T_c(6-body) = 0.833 94, T_c(exact) = 0.816 27, with T_c(Weiss) = 1.000 00; (ii) When closed-form equations for the low-temperature specific heat are expanded into infinite series, all of the known exact coefficients are reproduced; (iii) The extent of mathematical analysis and the amount of numerical computation are considerably less than those involved in diagram enumeration methods.

I. INTRODUCTION

It has been widely known that the Bragg-Williams approximation of order-disorder theory of binary alloys when applied to the Ising ferromagnet yields the Weiss molecular-field approximation and describes qualitative features of the second-order phase transition. When examined more closely, however, the molecular-field approximation entails many detrimental drawbacks; an over-estimation of the transition temperature, no tail of the specific heat on the high-temperature side, and most crucially, inability to predict correct critical-point exponents. Description of the second-order transition is substantially improved when the effect of the nearest-neighbor pair correlation is included.[1] The next higher-order approximation is not necessarily unique. One of the possible choices would be the equilateral triangle approximation which is appropriate only for the fcc lattice. This approximation has been attempted but does not seem to yield any critical temperature. An algebraic equation for the transition temperature in this approximation, however, yields a complex solution which is an improved value if the small imaginary part is ignored. A remarkable advance in the theory of cooperative phenomena has been accomplished by Kikuchi.[2] The approach is called the cluster variation method and is applied, in particular, to the square lattice of the two-dimensional Ising ferromagnet. This approximation is found to be equivalent to the Kramers-Wannier[3] variational solution of the eigenvalue formulation of the two-dimensional Ising ferromagnet and the order of approximation is often referred to as Kramers-Wannier-Kikuchi approximation.

In spite of its rather remarkable success and its subsequent improvement by Kurata, Kikuchi, and Watari,[4] the cluster variation method, in its original form has not been widely used as a method of formulating the theory of second-order phase transition because the method requires a statistical ensemble consideration each time a new cluster is added. This is accomplished by means of combinatoric analysis and hence the problem of bookkeeping becomes quite tedious. A rather exhaustive review of Kikuchi's cluster variation method and its applications to other lattice systems is given by Burley.[5] In order to overcome the aforementioned formulational complexity the cluster variation method has subsequently been reformulated and generalized by Morita.[6] In this generalization it is shown that (a) the cluster variation method is based on the exact variational principle of equilibrium statistical mechanics; (b) the method is applicable to quantum-statistical systems as well as to classical systems; (c) the exact cumulant expansion of the entropy in terms of the reduced density matrices is accomplished; (d) the order of approximation is decided at the moment when the cumulant expansion of entropy is truncated, hence, no ensemble formulation is repeated; (e) the Weiss molecular-field approximation, the Bethe pair approximation, and the Kramers-Wannier-Kikuchi approximation are special cases of the generalized cluster variation method. Quite recently, Tanaka and Libelo[7] applied the cluster-variation method to an fcc Heisenberg ferromagnet in which the nearest-neighbor tetrahedral spin correlation is taken into account and found a value of the critical temperature which is within one percent of the exact value of the power-series result.[8] This encourages us to attempt establishing some conjectures concerning criteria for attaining and improving approximations in the cluster variation method when applied to the general formulation of second-order phase transition.

The spin-$\frac{1}{2}$ fcc Ising ferromagnet treated in the present investigation yielded the transition temperature to within a few percent of the exact value, and all the known coefficients, 29 in number, of the low-temperature series for the specific heat were reproduced. The cluster variation method, which is designed originally for the purpose of obtaining various correlation functions in closed form, seems to serve as a means of producing rigorous power series for the correlation functions without ever enumerating the numbers of ways to fit various diagrams into the lattice. The physical system studied in the present investigation is the spin-$\frac{1}{2}$ Ising ferromagnet for the fcc lattice. In order to clearly exhibit the effect of including higher-body correlations, the above system will be treated, successively, in the Weiss (one-body), the pair (two-body), the triangle (three-body), the tetrahedron (four-body), and the octahedron plus tetrahedron (six-body) approximations. The results of the triangle and tetrahedron approximations are previously available[4] and only the last approximation (six-body) is new.

II. WEISS APPROXIMATION

The energy per lattice site is given by

$$E/N=-\tfrac{1}{2}zJx_2\,, \qquad (2.1)$$

where $x_2=\langle\mu_1\mu_2\rangle$ is the pair-correlation function, μ is the spin variable, z is the coordination number of the lattice, and N is the total number of lattice points. In the Weiss approximation all corrections are absent, and so the pair-correlation function, x_2, is approximated as the product of the long-range order, $x_1=\langle\mu_1\rangle$. Hence,

$$x_2=x_1^2\,. \qquad (2.2)$$

The entropy per lattice point in this approximation is given by

$$-S/kN=g_1(1)\,, \qquad (2.3)$$

where $g_1(1)$ is the one-body cumulant part of the entropy. The general cumulant parts, g's are related to G's by

$$G_n(j_1,j_2,\ldots,j_n)=\sum_j g_1(j)+\sum\sum_{j<k} g_2(j,k)+\cdots+g_n(j_1,j_2,\ldots,j_n)\,, \qquad (2.4)$$

where G_n is expressed in terms of the reduced density matrix $R^{(n)}$ as

$$G_n(j_1,j_2,\ldots,j_n)=\mathrm{Tr}R^{(n)}(j_1,j_2,\ldots,j_n)\times\ln R^{(n)}(j_1,j_2,\ldots,j_n)\,, \qquad (2.5)$$

$$n=1,2,\ldots,N\,.$$

The density matrices $R^{(n)}$'s are all diagonal for the Ising model, and the diagonal elements of $R^{(1)}(1)$ are given to be

$$R_{11}=\tfrac{1}{2}(1+x_1)\ \text{ and }\ R_{12}=\tfrac{1}{2}(1-x_1)\,. \qquad (2.6)$$

The free energy F can now be written

$$F/NkT=-zJ/(2kT)x_1^2+\tfrac{1}{2}(1+x_1)\ln(1+x_1)+\tfrac{1}{2}(1-x_1)\ln(1-x_1)\,. \qquad (2.7)$$

Minimizing F with respect to x_1, one obtains

$$0=-z(J/kT)x_1+\tfrac{1}{2}\ln(1+x_1)/(1-x_1)\,. \qquad (2.8)$$

Note that $x_1=0$ is always a solution of (2.8). However, for $T<zJ/k$ there is another, nonzero, solution for x_1: this nonzero solution is called the ordered solution and corresponds to a lower free energy as can be seen by expanding (2.7). Further the ordered solution goes continuously to zero as T approaches zJ/k. x_1 may, therefore, be treated as an order parameter. In this way, the Weiss approximation predicts a second-order phase transition and the critical temperature is given by

$$T_c=zJ/k\,. \qquad (2.9)$$

For fcc ($z=12$), $T_c(\text{Weiss})=12J/k$.

III. PAIR APPROXIMATION

The entropy per lattice point in this approximation is given by

$$-S/kN=g_1(1)+\tfrac{1}{2}zg_2(1,2)\,, \qquad (3.1)$$

where the coefficient of g_2 represents the number of nearest-neighbor pairs per lattice point. Using (2.1) and (2.4) with (3.1), the following expression for the free energy is obtained:

$$\frac{F}{NkT}=-\left(\frac{zJ}{2kT}\right)x_2+\tfrac{1}{2}zG_2(1,2)-(z-1)G_1(1)\,. \qquad (3.2)$$

The diagonal elements of the one-spin density matrix $R^{(1)}$ are given by (2.6). The four diagonal elements of the two-spin density matrix $R^{(2)}(1,2)$ are calculated as

$$\begin{aligned} R_{21}&=\tfrac{1}{4}(1+2x_1+x_2)\,,\\ R_{22}&=\tfrac{1}{4}(1-2x_1+x_2)\,,\\ R_{23}&=\tfrac{1}{4}(1-x_2)\,, \end{aligned} \qquad (3.3)$$

with multiplicities 1, 1, and 2, respectively. G's are related to density matrices through (2.5). The minimization of the free energy leads to the following relations for thermal averages x_1 and x_2:

$$x_1:\ 0=\tfrac{1}{4}\ln[(1+2x_1+x_2)/(1-2x_1+x_2)] \\ -\tfrac{1}{2}(z-1)\ln[(1+x_1)/(1-x_1)]\,, \tag{3.4a}$$

$$x_2:\ \frac{zJ}{2kT}=\tfrac{1}{8}\ln\left(\frac{(1+2x_1+x_2)(1-2x_1+x_2)}{(1-x_2)^2}\right). \tag{3.4b}$$

At a temperature slightly below the critical temperature T_c, x_1 has a very small nonzero value. Therefore, Eq. (3.4a) reduces to

$$x_2=1/(z-1)\ \text{ at } T=T_c\,.$$

Consequently, Eq. (3.4b) gives

$$\frac{zJ}{2kT_c}=\tfrac{1}{4}\ln\frac{1+x_2}{1-x_2}=\tfrac{1}{4}\ln\left(\frac{z}{z-2}\right).$$

So, the critical temperature T_c in the pair approximation is given by

$$T_c=2/[z\ln(z/(z-2))]\quad(\text{in units of } zJ/k)\,.$$

For fcc, $z=12$. Hence

$$T_c(\text{pair})=0.914\,14\ T_c\ (\text{Weiss})\,. \tag{3.5}$$

IV. TRIANGLE APPROXIMATION

As is stated in Sec. I the result of this section is not new. However, the cluster-variation method as reformulated by Morita[6] and used in this paper is entirely different from the one originally introduced by Kikuchi[2] and Kurata, Kikuchi, and Watari[4]; we shall present the full account of the triangle approximation. In this formulation all terms of the cumulant entropy are immediately determined at the moment when the structure of the largest (basic) cluster is decided. In the triangle approximation these are: the equilateral triangle (g_3), nearest-neighbor pair (g_2), and the single-site cluster (g_1).

The entropy per lattice point is given by

$$-S/kN=g_1+6g_2+8g_3\,, \tag{4.1}$$

where the numerical coefficients of g_2 and g_3 represent the numbers of nearest-neighbor pairs and equilateral triangles, per lattice point, respectively. Using (2.1) and (2.4) with (4.1) the following expression for the free energy is obtained:

$$F/NkT=-(6J/kT)x_2+13G_1-18G_2+8G_3\,. \tag{4.2}$$

The elements of the one-spin density matrix $R^{(1)}$ and the two-spin density matrix $R^{(2)}$ are the same as given before by (2.6) and (3.3). The eight nonzero elements of the three-spin density matrix $R^{(3)}$ are found as

$$\begin{aligned} R_{31}&=\tfrac{1}{8}(1+3x_1+3x_2+x_3)\,,\\ R_{32}&=\tfrac{1}{8}(1-3x_1+3x_2-x_3)\,,\\ R_{33}&=\tfrac{1}{8}(1+x_1-x_2-x_3)\,,\\ R_{34}&=\tfrac{1}{8}(1-x_1-x_2+x_3)\,, \end{aligned} \tag{4.3}$$

with multiplicities 1, 1, 3, and 3, respectively. Here $x_3=\langle\mu_1\mu_2\mu_3\rangle$ is the triplet correlation. Using (2.5), the following minimization conditions are obtained:

$$\begin{aligned} x_1:&\ 0=3\ln\left(\frac{1+3x_1+3x_2+x_3}{1-3x_1+3x_2-x_3}\ \frac{1+x_1-x_2-x_3}{1-x_1-x_2+x_3}\right)\\ &\qquad-9\ln\left(\frac{1+2x_1+x_2}{1-2x_1+x_2}\right)+\tfrac{13}{2}\ln\left(\frac{1+x_1}{1-x_1}\right),\\ x_2:&\ \frac{6J}{kT}=3\ln\left(\frac{(1+3x_1+3x_2+x_3)(1-x_1-x_2+x_3)}{(1-3x_1+3x_2-x_3)(1+x_1-x_2-x_3)}\right),\\ x_3:&\ 0=\ln\left(\frac{1+3x_1+3x_2+x_3}{1-3x_1+3x_2-x_3}\right)+3\ln\left(\frac{1-x_1-x_2+x_3}{1+x_1-x_2-x_3}\right), \end{aligned} \tag{4.5}$$

From these equations one finds, by neglecting squares and higher prowers of x_1 and x_3,

$$T_c=\left[6\ln\left(\frac{(1+3x_2)^2(1-x_2)}{(1+x_2)^3}\right)\right]^{-1}, \tag{4.6}$$

where x_2 are the solutions of

$$26x_2^2-9x_2+1=0\ . \tag{4.7}$$

Since (4.7) does not have a real root, (4.6) does not yield a real critical temperature. If one goes ahead and evaluates x_2 and T_c as complex numbers, one finds

$$\begin{aligned} x_2&=(0.173\,08,\ 0.092\,23)\,,\\ T_c&=(0.841\,70,\ -0.059\,14)\,. \end{aligned} \tag{4.8}$$

A critical temperature as a complex number is unphysical. If one looks at the real part of it, however, it is a substantial improvement over its value in the pair approximation (3.5).

V. TETRAHEDRON APPROXIMATION

This approximation is also discussed by Kurata, Kikuchi, and Watari,[4] however, the full account of the new formulation is presented in this section in order to demonstrate its simplicity.

In this approximation, the four-spin mother cluster is chosen to be the smallest tetrahedron. Thus each diagram which is a part of the nearest-neighbor tetrahedron will contribute to the entropy term. Consequently, the entropy per lattice point is given by

$$-S/kN=g_1+6g_2+8g_3+2g_4\,, \tag{5.1}$$

where the coefficients of g_2, g_3, and g_4 represent the numbers of nearest-neighbor pairs, equilateral triangles, and regular tetrahedra per lattice point, respectively. One may note the fact that the entropy (5.1) has an additional term which is contributed by the tetrahedron when compared with the entropy (4.1) in the triangle approximation. In Kikuchi's formulation each time an additional cluster is included the statistical ensemble consideration must be repeated. There is no such complication in the present formulation. Using (2.1) and (2.4) together with (5.1) the following expression for the free energy is obtained:

$$F/NkT = -(6J/kT)x_2 + 2G_4 - 6G_2 + 5G_1 , \qquad (5.2)$$

G_1 and G_2 are the same as in the pair and triangle approximations. The 16 elements of the four-spin density matrix $R^{(4)}$ are calculated as

$$\begin{aligned} R_{41} &= \tfrac{1}{16}\,(1 + 4x_1 + 6x_2 + 4x_3 + x_4) ,\\ R_{42} &= \tfrac{1}{16}\,(1 - 4x_1 + 6x_2 - 4x_3 + x_4) ,\\ R_{43} &= \tfrac{1}{16}\,(1 + 2x_1 - 2x_3 - x_4) ,\\ R_{44} &= \tfrac{1}{16}\,(1 - 2x_1 + 2x_3 - x_4) ,\\ R_{45} &= \tfrac{1}{16}\,(1 - 2x_2 + x_4) , \end{aligned} \qquad (5.3)$$

with multiplicities 1, 1, 4, 4, and 6, respectively. Here

$$x_3 = \langle \mu_1\mu_2\mu_3\rangle \quad \text{and } x_4 = \langle \mu_1\mu_2\mu_3\mu_4\rangle . \qquad (5.4)$$

x_3 is the triangle correlation and x_4 is the tetrahedron correlation, respectively. Using (2.5), the following minimization conditions for F are obtained:

$$x_1 : \quad 0 = \tfrac{1}{2}\ln\left(\frac{R_{41}}{R_{42}}\right) + \ln\left(\frac{R_{43}}{R_{44}}\right) - 3\ln\left(\frac{R_{21}}{R_{22}}\right) + \tfrac{5}{2}\ln\left(\frac{R_{11}}{R_{12}}\right) , \qquad (5.5a)$$

$$x_2 : \quad \frac{6J}{kT} = \tfrac{3}{4}\ln\left(\frac{R_{41}R_{42}}{R_{45}^2}\right) - \tfrac{3}{2}\ln\left(\frac{R_{21}R_{22}}{R_{23}^2}\right) , \qquad (5.5b)$$

$$x_3 : \quad 0 = \ln(R_{41}/R_{42}) - \ln(R_{43}/R_{44}) , \qquad (5.5c)$$

$$x_4 : \quad 0 = \tfrac{1}{8}\ln[R_{41}R_{42}R_{45}/(R_{43}R_{44})^4] . \qquad (5.5d)$$

At a temperature slightly below the critical temperature T_c, x_1 and x_3 are nonzero and very small. Therefore, from Eqs. (5.5a) and (5.5c) one finds that $x_2 = \frac{1}{5}$ or $\frac{1}{3}$. It will be shown in Sec. VIII that the reduced susceptibility in the present approximation has the closed-form expression

$$\chi = (1 + 3x_2)(1 + x_2)/(1 - 3x_2)(1 - 5x_2) . \qquad (5.6)$$

At infinite temperature all correlations vanish, and hence $x_2 = 0$ and χ has its high-temperature value of unity. As the temperature decreases, the correlations increase, the value $\frac{1}{5}$ of x_2 will be attained at some finite temperature. Below this temperature (5.6) is not valid. Therefore the value $\frac{1}{3}$ for x_2 must be rejected in favor of $x_2 = \frac{1}{5}$. Substituting the value of x_2, (5.5d) reduces to the cubic equation

$$625x_4^3 - 75x_4^2 + 515x_4 - 41 = 0 ,$$

which has only one positive root between 0 and 1, namely, $x_4 = 0.079\,92$. Using the values of x_2 and x_4 in Eq. (5.5b), T_c is calculated as

$$T_c(\text{four-spin}) = 0.835\,45\,T_c\,(\text{Weiss}) . \qquad (5.7)$$

VI. OCTAHEDRON PLUS TETRAHEDRON APPROXIMATION

The next higher-order approximation would, naturally, be the octahedron approximation, however, one again finds no real critical temperature as in the triangle approximation.[4] This means that one has to go either to a still larger cluster or to a combination of two parent clusters. In the case of the triangle approximation one cannot choose the triangle and pair both as parent cluster because the pair is a part of triangle. Since the regular tetrahedron is not a part of octahedron both clusters can be chosen as the parent clusters. One should note the fact, however, that this approximation is still different from the one in which a single cluster, which is a combination of the regular tetrahedron and octahedron, is used as the parent cluster. Such an approximation would certainly be better that the one which is developed in this section. An approximation developed in this section will be called the octahedron plus tetrahedron approximation. In this approximation, each diagram which is a part of tetrahedron or octahedron will contribute to the entropy. Consequently, the entropy per lattice point is given by (see Fig. 1.)

$$\begin{aligned} -S/kN = {}& g_1(1) + 6g_2(1,2) + 3g_2(1,6) + 8g_3(1,2,3)\\ &+ 12g_3(1,2,7) + 2g_4(1,2,3,4) + 3g_4(2,3,5,7)\\ &+ 12g_4(3,5,6,7) + 6g_5(1,2,3,5,7)\\ &+ g_6(1,2,3,5,6,7) , \end{aligned} \qquad (6.1)$$

where the numerical coefficients of g_2's through g_6 represent the numbers of nearest-neighbor pairs, equilateral triangles, isosceles triangles, regular tetrahedra, squares, tetrahedra of the type 3567, pyramids, and octahedra per lattice point, respectively. One may again note the fact that six terms, which are contributions of the octahedron and its subclusters, are added to the entropy of the tetrahedron approximation, yet no change of the numerical coefficients of tetrahedron terms is required. Using (2.1) and (2.4) together with

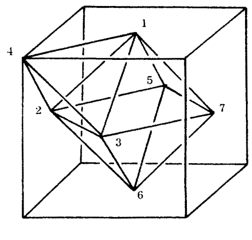

FIG. 1. Octahedron plus tetrahedron.

(6.1), the expression for the free energy is found to be

$$F/NkT = -(6J/kT)x_2 + G_6(1,2,3,5,6,7) + 2G_4(1,2,3,4) - 8G_3(1,2,3) + 6G_2(1,2) - G_1(1)\,. \tag{6.2}$$

Note that G_5 does not appear in the expression for entropy. In the general formulation of the cluster variation method, this situation is expressed in geometrical terms by saying that any diagram which is not a common part of two neighboring parent clusters does not contribute to the entropy.[6] In the present case, a pyramid is not shared by two octahedra. For similar reasons, $G_4(2,3,5,7)$, $G_4(3,5,6,7)$, $G_3(1,2,7)$,and $G_2(1,6)$ are absent from the entropy.

The elements of the one-spin density matrix $R^{(1)}(1)$, the two-spin density matrix $R^{(2)}(1,2)$, the three-spin density matrix $R^{(3)}(1,2,3)$, and the four-spin density matrix are, respectively, given by (2.6), (3.3), (4.3), and (5.3). The six-spin density matrix has sixty-four elements (only diagonal) and these are found as

$$\begin{aligned}
R_{61} &= N_0(S_{12}+L_{12})\,, \quad 1\\
R_{62} &= N_0(S_{12}-L_{12})\,, \quad 1\\
R_{63} &= N_0(S_{34}+L_{34})\,, \quad 6\\
R_{64} &= N_0(S_{34}-L_{34})\,, \quad 6\\
R_{65} &= N_0(S_{56}+L_{56})\,, \quad 12\\
R_{66} &= N_0(S_{56}-L_{56})\,, \quad 12\\
R_{67} &= N_0(S_{78}+L_{78})\,, \quad 3\\
R_{68} &= N_0(S_{78}-L_{78})\,, \quad 3\\
R_{69} &= N_0(1-3x_{10}+3x_6-x_9)\,, \quad 8\\
R_{60} &= N_0(1-4x_2+x_{10}-x_6+4x_7-x_9)\,, \quad 12
\end{aligned} \tag{6.3}$$

where N_0 is the normalization factor, $\frac{1}{64}$, multiplicities of these elements are indicated at the end of each line, and the S's (short-range part) and L's (long-range part) are given by

$$\begin{aligned}
S_{12} &= 1+12x_2+3x_{10}+3x_6+12x_7+x_9\,,\\
S_{34} &= 1+4x_2+x_{10}-x_6-4x_7-x_9\,,\\
S_{56} &= 1-x_{10}-x_6+x_9\,,\\
S_{78} &= 1-4x_2+3x_{10}+3x_6-4x_7+x_9\,,
\end{aligned} \tag{6.4}$$

$$\begin{aligned}
L_{12} &= 6x_1+8x_3+12x_5+6x_8\,,\\
L_{34} &= 4x_1-4x_8\,,\\
L_{56} &= 2x_1-4x_5+2x_8\,,\\
L_{78} &= 2x_1-8x_3+4x_5+2x_8\,.
\end{aligned} \tag{6.5}$$

Here newly introduced x's are defined by

$$\begin{aligned}
&x_5 = \langle \mu_1\mu_2\mu_7\rangle\,, \quad x_6 = \langle \mu_2\mu_3\mu_5\mu_7\rangle\,,\\
&x_7 = \langle \mu_3\mu_5\mu_6\mu_7\rangle\,, \quad x_8 = \langle \mu_2\mu_3\mu_5\mu_6\mu_7\rangle\,,\\
&x_9 = \langle \mu_1\mu_2\mu_3\mu_5\mu_6\mu_7\rangle\,, \quad x_{10} = \langle \mu_1\mu_6\rangle\,.
\end{aligned} \tag{6.6}$$

Using (2.5), the minimization of F leads to the following set of ten equilibrium conditions:

$$x_1:\ 0 = \tfrac{3}{32}\ln\left(\frac{R_{61}R_{67}}{R_{62}R_{68}}\right) + \tfrac{3}{8}\ln\left(\frac{R_{63}R_{65}}{R_{64}R_{66}}\right) + \tfrac{1}{2}\ln\left(\frac{R_{41}}{R_{42}}\right) + \ln\left(\frac{R_{43}}{R_{44}}\right) - 3\ln\left(\frac{R_{31}R_{33}}{R_{32}R_{34}}\right) + 3\ln\left(\frac{R_{21}}{R_{22}}\right) - \tfrac{1}{2}\ln\left(\frac{R_{11}}{R_{12}}\right), \tag{6.7a}$$

$$x_2:\ \frac{1}{2T} = \tfrac{3}{16}\ln\left(\frac{R_{61}R_{62}}{R_{67}R_{68}}\right) + \tfrac{3}{8}\ln\left(\frac{R_{63}R_{64}}{R_{60}^2}\right) + \tfrac{3}{4}\ln\left(\frac{R_{41}R_{42}}{R_{45}^2}\right) - 3\ln\left(\frac{R_{31}R_{32}}{R_{33}R_{34}}\right) + \tfrac{3}{2}\ln\left(\frac{R_{21}R_{22}}{R_{23}^2}\right), \tag{6.7b}$$

$$x_3:\ 0 = \tfrac{1}{8}\ln\left(\frac{R_{61}}{R_{62}}\right) - \tfrac{3}{8}\ln\left(\frac{R_{67}}{R_{68}}\right) + \tfrac{1}{2}\ln\left(\frac{R_{41}}{R_{42}}\right) - \ln\left(\frac{R_{43}}{R_{44}}\right) - \ln\left(\frac{R_{31}}{R_{32}}\right) + 3\ln\left(\frac{R_{33}}{R_{34}}\right), \tag{6.7c}$$

$$x_4:\ 0 = \tfrac{1}{8}\ln(R_{41}R_{42}) - \tfrac{1}{2}\ln(R_{43}R_{44}) + \tfrac{3}{4}\ln R_{45}\,, \tag{6.7d}$$

$$x_5:\ 0 = \tfrac{3}{16}\ln\left(\frac{R_{61}}{R_{62}}\right) - \tfrac{3}{4}\ln\left(\frac{R_{65}}{R_{66}}\right) + \tfrac{3}{16}\ln\left(\frac{R_{67}}{R_{68}}\right), \tag{6.7e}$$

$$x_6:\ 0 = \tfrac{3}{64}\ln(R_{61}R_{62}) - \tfrac{3}{32}\ln(R_{63}R_{64}) - \tfrac{3}{16}\ln(R_{65}R_{66}) + \tfrac{3}{8}\ln R_{69} + \tfrac{9}{64}\ln(R_{67}R_{68}) - \tfrac{3}{16}\ln R_{60}\,, \tag{6.7f}$$

$$x_7:\ 0 = \tfrac{3}{16}\ln\left(\frac{R_{61}R_{62}}{R_{67}R_{68}}\right) - \tfrac{3}{8}\ln\left(\frac{R_{63}R_{64}}{R_{60}^2}\right), \tag{6.7g}$$

$$x_8:\ 0 = \tfrac{1}{64}\ln\left(\frac{R_{61}}{R_{62}}\right) - \tfrac{3}{8}\ln\left(\frac{R_{63}R_{66}}{R_{64}R_{65}}\right) + \tfrac{3}{32}\ln\left(\frac{R_{67}}{R_{68}}\right), \tag{6.7h}$$

$$x_9:\ 0 = \tfrac{1}{64}\ln(R_{61}R_{62}) - \tfrac{3}{32}\ln(R_{63}R_{64}) + \tfrac{3}{16}\ln(R_{65}R_{66}) + \tfrac{3}{64}\ln(R_{67}R_{68}) - \tfrac{1}{8}\ln R_{69} - \tfrac{3}{16}\ln R_{60}\,, \tag{6.7i}$$

$$x_{10}:\ 0 = \tfrac{3}{64}\ln(R_{61}R_{62}) + \tfrac{3}{32}\ln(R_{63}R_{64}) - \tfrac{3}{16}\ln(R_{65}R_{66}) + \tfrac{9}{64}\ln(R_{67}R_{68}) - \tfrac{3}{8}\ln R_{69} + \tfrac{3}{16}\ln R_{60}\,. \tag{6.7j}$$

In the immediate neighborhood of the critical temperature, the quantities x_1, x_3, x_5, and x_8 are very small and may be set equal to zero. Then (6.7a), (6.7c), (6.7e), and (6.7h) are trivially satisfied. The solution of (6.7b), (6.7d), (6.7f), (6.7g), (6.7i), and (6.7j) can be obtained at each temperature by a computer program called MINMAX.[11]

The critical temperature is found by observing the singularity of the reduced susceptibility:

$$T_c(\text{six-spin}) = 0.833\,94\ T_c(\text{Weiss})\,. \tag{6.8}$$

VII. HIGH-TEMPERATURE SPECIFIC HEAT

The expression for the high-temperature specific heat is given by

$$(kT/J)^2 c_v(w) = \tfrac{1}{2} z (1-w^2) \frac{dx_2}{dw}\,, \tag{7.1}$$

where

$$w = \tanh(J/kT)\,. \tag{7.2}$$

Weiss approximation

At a temperature higher than the critical temperature, the only solution of Eq. (2.8) is $x_1 = 0$. So, Eq. (2.2) leads to $x_2 = 0$. Hence in the Weiss approximation (7.1) reduces to

$$(kT/J)^2 c_v(w) = 0\,. \tag{7.3}$$

Pair approximation

Since $x_1 = 0$ above the critical temperature, Eq. of (3.4b) reduces to

$$(J/kT) = \tfrac{1}{2} \ln(1+x_2)/(1-x_2) = \tanh^{-1} x_2$$

or

$$x_2 = \tanh(J/kT) = w\,. \tag{7.4}$$

Therefore, Eq. (7.1) gives

$$(kT/J)^2 c_v(w) = \tfrac{1}{2} z (1-w^2)\,.$$

For fcc $(z = 12)$,

$$(kT/J)^2 c_v = 6 - 6w^2\,. \tag{7.5}$$

Tetrahedron approximation

Eqs. of (5.5a) and (5.5c) are satisfied identically as $x_1 = 0$ and $x_3 = 0$ for $T > T_c$. Noting the fact that as T goes to infinity, x_2 and x_4 tend to zero, one writes

$$x_2 = \sum_1^\infty b_n w^n \quad \text{and} \quad x_4 = \sum_1^\infty d_n w^n\,,$$

where b's and d's are to be determined. Substituting the above expansions in Eqs. (5.5b) and (5.5d), the expressions for x_2 and x_4 are obtained as

$$\begin{aligned} x_2 &= w + 4w^2 + 20w^3 + 116w^4 + \cdots\,, \\ x_4 &= 3w^2 + 16w^3 + 87w^4 + \cdots\,. \end{aligned} \tag{7.6}$$

This leads to the following expression for specific heat:

$$(kT/J)^2 c_v = 6 + 48w + 354w^2 + \cdots\,. \tag{7.7}$$

Octahedron plus tetrahedron approximation

For $T > T_c$, Eqs. (6.7a), (6.7c), (6.7e), and (6.7h) are satisfied identically as $x_1 = x_3 = x_5 = x_8 = 0$. Equations (6.7f), (6.7g), (6.7i), and (6.7j) can be manipulated to yield

$$x_6 = x_7 \quad \text{and} \quad x_9 + x_{10} = 2x_6\,. \tag{7.8}$$

In accordance with the fact that at infinite temperature, the thermal averages $x_2, x_4, x_6, x_7, x_9, x_{10}$ approach the value zero, the following form of series expansion is assumed for the x's mentioned above:

$$x = \sum_1^\infty a_n w^n\,. \tag{7.9}$$

Substituting x's in the form (7.9), (Eqs. (6.7b), (6.7d), (6.7f), (6.7g), (6.7i), and (6.7j) yield

$$\begin{aligned} x_2 &= w + 4w^2 + 22w^3 + 136w^4 + 880w^5 + 5908w^6 + \cdots\,, \\ x_4 &= 3w^2 + 16w^3 + 108w^4 + 744w^5 + 5180w^6 + \cdots\,, \\ x_6 = x_7 &= 2w^2 + 16w^3 + 108w^4 + 736w^5 + 5144w^6 + \cdots\,, \\ x_9 &= 8w^3 + 72w^4 + 528w^5 + 3858w^6 + \cdots\,, \\ x_{10} &= 4w^2 + 24w^3 + 144w^4 + 944w^5 + 6432w^6 + \cdots\,. \end{aligned} \tag{7.10}$$

Using the series for x_2 in (7.1), the high-temperature specific heat is found to be

$$\begin{aligned} (kT/J)^2 c_v = 6 &+ 48w + 390w^2 \\ &+ 3216w^3 + 260\,04w^4 + \cdots\,. \end{aligned} \tag{7.11}$$

The exact power series result gives[9]

$$\begin{aligned} (kT/J)^2 c_v = 6 &+ 48w + 390w^2 + 3216w^3 \\ &+ 26\,844w^4 + 229\,584w^5 + 2\,006\,736w^6 \\ &+ 17\,809\,008w^7 + \cdots\,. \end{aligned} \tag{7.12}$$

Thus the six-spin approximation reproduces the first four of the eight known terms exactly.

VIII. HIGH-TEMPERATURE SUSCEPTIBILITY

In the presence of a small external magnetic field H, the equilibrium condition corresponding to x_1 is modified so that its left hand side is replaced by mH/kT. This results in nonzero values for the expectation values of some x's (which otherwise should have been zero) even at temperatures higher than the critical temperature. The reduced susceptibility per spin[10] is given by

$$\chi = (kT/m)(\delta x_1/\delta H)_{T,\,H=0}\,. \tag{8.1}$$

Weiss approximation

Equation (2.8) when modified, results in the following expression for the susceptibility:

$$\chi = 1/(1 - zJ/kT) = 1 + 12w + 144w^2 + \cdots\,. \tag{8.2}$$

Pair approximation

The susceptibility is given by

$$\chi = (1+x_2)/[1-(z-1)x_2]$$
$$= 1 + z x_2 + z(z-1)x_2^2 + z(z-1)^2 x_2^3 + \cdots .$$

Using (7.4) and $z=12$, this reduces to

$$\chi = 1 + 12w + 132w^2 + 1452w^3 + \cdots . \tag{8.3}$$

Tetrahedron approximation

Modified equations (5.5a) and (5.5c) are expanded up to first powers in x_1 and x_3. The resulting linear equations are solved to give

$$x_1 = \frac{mH}{kT}\,\frac{(1+3x_2)(1+x_2)}{(1-3x_2)(1-5x_2)} ,$$

which by using (8.1) results in (5.6). Substituting from (7.6) for x_2, one obtains

$$\chi = 1 + 12w + 132w^2 + 1404w^3 + 14\,676w^4 + \cdots . \tag{8.4}$$

Octahedron plus tetrahedron approximation

Modified equations (6.7a), (6.7c), (6.7e), and (6.7h) in the linear-response region to find an expression for x_1, in terms of H. Then using (8.1) and (7.10), the following expression for susceptibility is obtained:

$$\chi = 1 + 12w + 132w^2 + 1404w^3 + 14\,652w^4$$
$$+ 151\,116w^5 + 1\,546\,668w^6 + \cdots . \tag{8.5}$$

The exact power series result[14] gives

$$\chi = 1 + 12w + 132w^2 + 1404w^3 + 14\,652w^4$$
$$+ 151\,116w^5 + 1\,546\,332w^6 + 15\,734\,460w^7$$
$$+ 159\,425\,580w^8 + 160\,998\,770w^9 + 16\,215\,457\,188w^{10}$$
$$+ 162\,961\,837\,500w^{11} + 1\,634\,743\,178\,420w^{12}$$
$$+ 16\,373\,484\,437\,340w^{13} + \cdots . \tag{8.6}$$

Thus the six-spin approximation reproduces the first six coefficients exactly and the seventh one within 0.02%.

IX. LOW-TEMPERATURE SPECIFIC HEAT

The low-temperature specific heat is given by

$$\left(\frac{kT}{J}\right)^2 c_v = 2zu\,\frac{d(1-x_2)}{du} , \tag{9.1}$$

where

$$u = \exp(-4J/kT) . \tag{9.2}$$

At zero temperature, the thermal averages of all the correlations are unity. Consequently all the x's in the equilibrium conditions are replaced by expansions of the form

$$x = 1 - u^{\alpha} \sum_{0}^{\infty} a_n u^n , \quad a_0 \neq 0 , \tag{9.3}$$

where α's and a_n's are to be determined.

Weiss approximation

Substituting expression (9.3) for x_1 in Eq. (2.8) and comparing the coefficients of $\ln u$ on both sides, one finds

$$\alpha = \tfrac{1}{2} z . \tag{9.4}$$

A further comparison of coefficients of different powers of u in Eq. (2.8) leads to

$$x_1 = 1 - 2u^{z/2} + 2u^{z} + \cdots , \tag{9.5}$$

which on using (2.2) and (9.1) yields

$$(kT/J)^2 c_v(u) = 4z^2 u^{z/2} - 16 z^2 u^{z} + \cdots .$$

For fcc ($z=12$),

$$(kT/J)^2 c_v = 576u^6 - 2304u^{12} + \cdots . \tag{9.6}$$

Pair approximation

The following u expansion for $1-x_1$, $1-x_2$, and $1-2x_1+x_2$ are assumed in Eqs. (3.4):

$$\begin{aligned} 1-x_1 &= 2u^{\alpha_1} \sum_{0}^{\infty} a_n u^n , \quad a_0 \neq 0 , \\ 1-x_2 &= 4u^{\alpha_2} \sum_{0}^{\infty} b_n u^n , \quad b_0 \neq 0 , \\ 1-2x_1+x_2 &= 4u^{\alpha_3} \sum_{0}^{\infty} c_n u^n , \quad c_0 \neq 0 . \end{aligned} \tag{9.7}$$

A comparison of the coefficients of $\ln u$ in Eq. (3.4) leads to

$$0 = -3\alpha_3 + \tfrac{11}{2}\alpha_1 , \quad -1 = \alpha_3 - 2\alpha_2 . \tag{9.8}$$

The set (9.8) by itself is undetermined for finding a unique solution. Another physical restriction is imposed by the fact that $\frac{1}{4}(1-2x_1+x_2)$ is the probability of some configuration and the leading term in its u expansion governs its sign. Mathematically it means

$$\alpha_3 \geqslant \min(\alpha_1, \alpha_2) , \tag{9.9}$$

with equality holding only when $\alpha_1 = \alpha_2$. Equations (9.8) and (9.9) have a unique solution:

$$\alpha_1 = 6, \quad \alpha_2 = 6, \quad \alpha_3 = 11 . \tag{9.10}$$

Feeding back information (9.10) in Eq. (3.4), a further comparison of the coefficients of higher powers of u yields

$$\begin{aligned} x_1 &= 1 - 2u^6(1 + 12u^5 - 13u^6 + 198u^{10} + \cdots) , \\ x_2 &= 1 - 4u^6(1 + 11u^5 - 13u^6 + 176u^{10} + \cdots) . \end{aligned} \tag{9.11}$$

The expression for the low-temperature specific heat is given by

$$(kT/J)^2 c_v(u) = 576u^6 + 11\,616u^{11} - 14\,976u^{12} + 270\,336u^{16} + \cdots . \qquad (9.12)$$

Tetrahedron approximation

Following the same procedure as in the pair approximation, an underdetermined set of equations similar to (9.7) is obtained. This set can be uniquely solved, if the results (9.10) are assumed to be known. Feeding back this information in Eq. (5.5), the following series for the low-temperature specific heat is obtained:

$$(kT/J)^2 c_v = 576u^6 + 11\,616u^{11} - 14\,976u^{12} + 28\,800u^{15} + 172\,032u^{16} - 554\,880u^{17} + 374\,976u^{18} + 138\,624u^{19} + 768\,000u^{20} + \cdots . \qquad (9.13)$$

Octahedron plus tetrahedron approximation

In this approximation, the series for specific heat reads

$$(kT/J)^2 c_v(u) = 576u^6 + 11\,616u^{11} - 14\,976u^{12} + 28\,800u^{15} + 172\,032u^{16} - 554\,880u^{17} + 374\,976u^{18} + 138\,624u^{19} + 787\,200u^{20} + 889\,056u^{21} - 12\,568\,512u^{22} + 20\,465\,952u^{23} - 4\,564\,224u^{24} + 8\,220\,000u^{25} - 29\,235\,648u^{26} - 180\,931\,968u^{27} + 633\,948\,672u^{28} - 558\,773\,856u^{29} + 125\,758\,080u^{30} + \cdots . \qquad (9.14)$$

This series reproduces all the terms given by Baker[9] and agrees perfectly well with the latest results.[12] The details of the calculations in the last two apprximations have been omitted for their length; they are nontrivial and require some manipulational skill.

X. CONCLUSIONS

The spin-$\frac{1}{2}$ fcc Ising ferromagnet treated in the present investigation brings out the following remarkable features.

(a) The transition temperature is predicted to within a few percent of the exact value:

$T_c(\text{four-body}) = 0.83\,545$,

$T_c(\text{six-body}) = 0.83\,394$,

$T_c(\text{exact}) = 0.81\,627^{(12)}$,

where

$T_c(\text{Weiss}) = 1.000\,00$.

(b) All the known exact coefficients of the power series for the low-temperature specific heat are reproduced.

(c) The high-temperature specific heat expansion reproduces four out of the eight known exact coefficients.

(d) The high-temperature susceptibility expansion reproduces six out of the 14 known coefficients exactly and the seventh to within about 0.02%.

(e) The extent of mathematical analysis and the amount of numerical computation are much less than those involved in the diagram methods.

(f) In the process of obtaining low-temperature specific heat, series for the expectation values of correlations were also obtained. In spite of the fact that these series are not available in the current literature, except the equilateral triangle correlation,[13] they are listed in Table I. Agreement with the available result for the triplet correlation is again excellent.

(g) It is confirmed in this formulation that all the odd order parameters have the same critical-point exponent, the fact used in determining the critical temperature. This assertion seems to be supported by the diagram method of power series for the triplet order parameters.[13]

Seemingly our results are not as good for the high-temperature expansions as for the low-temperature expansions. This may be interpreted as follows: The minimum conditions for the free energy with respect to the long-range order parameters are trivially satisfied at temperatures higher than the critical temperature. This means that the number of self-consistency conditions is reduced, i.e., the number of paths through which effects of feedback are transmitted is reduced. While on the low-temperature side of the critical temperature, the long-range order parameters are determined consistently in conjunction with the short-range order parameters and hence the free energy is minimized under more-stringent conditions.

The cluster variation formulation was designed originally for the purpose of obtaining various correlation functions as solutions of coupled nonlinear equations, however, as has been demonstrated in this investigation the method may be used as a means of producing rigorous power series expansions for the correlation functions without ever enumerating the numbers of ways to fit vari-

TABLE I. Coefficients of power series for correlation functions. $\langle 123\rangle = \langle \mu_1\mu_2\mu_3\rangle$, etc. (see Fig. 1).

Correlation function	u^0	u^6	u^{11}	u^{12}	u^{15}
$\langle 1\rangle$	1	−2	−24	26	−48
$\langle 12\rangle$	1	−4	−44	52	−80
$\langle 16\rangle$	1	−4	−48	56	−96
$\langle 123\rangle$	1	−6	−60	78	−104
$\langle 124\rangle$	1	−6	−64	82	−112
$\langle 1234\rangle$	1	−8	−72	104	−128
$\langle 2357\rangle$	1	−8	−80	112	−128
$\langle 3567\rangle$	1	−8	−76	108	−128
$\langle 23567\rangle$	1	−10	−88	138	−144
$\langle 123567\rangle$	1	−12	−96	168	−160
	u^{16}	u^{17}	u^{18}	u^{19}	u^{20}
$\langle 1\rangle$	−252	720	−438	−192	−984
$\langle 12\rangle$	−448	1 360	−868	−304	−1 640
$\langle 16\rangle$	−488	1 504	−964	−368	−1 896
$\langle 123\rangle$	−588	1 920	−1 298	−384	−2 088
$\langle 124\rangle$	−636	2 064	−1 386	−416	−2 264
$\langle 1234\rangle$	−672	2 400	−1 720	−480	−2 448
$\langle 2357\rangle$	−784	2 688	−1 896	−480	−2 624
$\langle 3567\rangle$	−728	2 544	−1 816	−464	−2 544
$\langle 23567\rangle$	−828	3 088	−2 334	−512	−2 856
$\langle 123567\rangle$	−888	3 312	−2 860	−528	−3 144
	u^{21}	u^{22}	u^{23}	u^{24}	u^{25}
$\langle 1\rangle$	−1 008	12 924	−19 536	3 062	−8 280
$\langle 12\rangle$	−1 764	23 804	−37 076	7 924	−13 700
$\langle 16\rangle$	−1 664	26 312	−41 648	8 660	−15 072
$\langle 123\rangle$	−2 068	32 496	−52 932	13 818	−16 500
$\langle 124\rangle$	−2 296	35 300	−57 256	14 786	−18 112
$\langle 1234\rangle$	−1 720	39 048	−67 224	19 528	−16 632
$\langle 2357\rangle$	−2 832	45 088	−75 568	22 520	−20 240
$\langle 3567\rangle$	−2 276	42 020	−71 428	21 480	−19 108
$\langle 23567\rangle$	−2 288	49 788	−88 240	29 822	−20 472
$\langle 123567\rangle$	−1 904	55 752	−103 488	38 956	−22 224
	u^{26}	u^{27}	u^{28}	u^{29}	u^{30}
$\langle 1\rangle$	26 694	153 536	−507 948	406 056	−78 972
$\langle 12\rangle$	46 852	279 216	−943 376	802 836	−174 664
$\langle 16\rangle$	57 484	302 048	−1 057 024	908 432	−184 296
$\langle 123\rangle$	65 010	369 136	−1 308 588	1 185 348	−268 050
$\langle 124\rangle$	70 274	404 368	−1 425 780	1 286 640	−291 684
$\langle 1234\rangle$	86 856	418 528	−1 609 440	1 541 112	−318 704
$\langle 2357\rangle$	85 144	511 424	−1 847 296	1 746 736	−413 872
$\langle 3567\rangle$	86 048	464 624	−1 726 960	1 649 564	−387 488
$\langle 23567\rangle$	103 838	536 416	−2 086 524	2 090 952	−515 116
$\langle 123567\rangle$	123 876	579 232	−2 387 904	2 511 840	−669 456

ous diagrams into the lattice. For this reason, the cluster variation method may be regarded as another way of finding rigorous critical behaviors of the Ising ferromagnet when combined with the Padé approximant method.

It is generally observed that the larger the cluster, the better the approximation is. For example, for fcc, the tetrahedron approximation improves upon the results of the pair approximation, and the octahedron plus tetrahedron as mother clusters improve upon the results of the tetrahedron approximation. However, the mother cluster cannot be arbitrarily chosen. This can be illustrated by selecting a three-spin nearest-neighbor linear chain as mother cluster (not described in the text). Not only is the same value for critical temperature obtained, but the high-temperature expansions for specific heat and susceptibility are also exactly the same as those in the pair approximation. Clearly a large linear cluster does not necessarily give a better result.

It seems that the mother cluster should be spatially compact. A better critical temperature is obtained in the tetrahedron rather than in the square approximation though both of them involve four-spin mother clusters. The critical temperature seems to be inherited from some smaller clusters. For example, the octahedron approximation apparently does not yield a critical temperature, but the octahedron plus tetrahedron approximation improves upon the critical temperature of the tetrahedron approximation. A further conclusion is that the mother cluster should preferably reflect the dimensionality of the lattice. For example, the equilateral triangle approximation loses the critical temperature, which is regained in the tetrahedron approximation. In summary the present investigation indicates that the mother cluster should be (i) large, (ii) spatially compact, and (iii) reflecting the dimensionality of the lattice. These criteria agree more or less with those proposed by previous authors[4,5] except for minor details.

Finally, the critical exponents predicted by the cluster variation methods should be classical if worked out only with the closed form expressions for the correlation functions. However, when the cluster variation method is used as a means of creating power series for various correlation functions in conjunction with the Padé approximant method, the critical exponents of nonclassical nature can be predicted.

For further work on these lines, for the fcc lattice, an approximation in which the seven-spin capped octahedron (1 234 567), which contains the tetrahedron and octahedron as subclusters, is suggested.

An important and useful direction for further study will be to include the next-nearest-neighbor interaction and try to reproduce the phase diagram for magnetic systems. The lattice gas model can be studied in a better approximation. Most of the studies for these systems have so far been restricted to the Weiss approximation.

ACKNOWLEDGMENTS

One of the authors (T. T.) is grateful to Dr. Tohru Morita for his constant encouragement over the past years, which motivated the present investigation. The authors are indebted to Dr. Jeremiah Barry for his stimulating discussions and critical reading of the manuscript. Thanks are also due to Dr. R. Kikuchi for his valuable comments.

---

*Supported by NASA Contract No. NAS5-20885.

[1] H. A. Bethe, Proc. R. Soc. A 150, 150 (1935); 150, 552 (1935).

[2] R. Kikuchi, Phys. Rev. 81, 988 (1951), J. Chem. Phys. 19, 1230 (1951).

[3] H. A. Kramers and G. H. Wannier, Phys. Rev. 60, 251 (1941); 60, 263 (1941).

[4] M. Kurata, R. Kikuchi, and T. Watari, J. Chem. Phys. 21, 434 (1953).

[5] D. M. Burley, in Ref. 12, Vol. II, 1972.

[6] T. Morita, J. Phys. Soc. Jpn. 12, 753 (1957); 12, 1060 (1957); J. Math. Phys. 13, 115 (1972).

[7] T. Tanaka and L. F. Libelo, Phys. Rev. B 12, 1790 (1975).

[8] G. A. Baker, Jr., Phys. Rev. 136, A1376 (1964).

[9] G. A. Baker, Jr., Phys. Rev. 129, 99 (1963).

[10] M. F. Sykes, J. Math. Phys. 2, 52 (1961).

[11] J. Cambell, W. E. Moore, and H. Wolf, Manned Spacecraft Center, NASA, Houston, Tex., report 1964 (unpublished).

[12] *Phase Transition and Critical Phenomena*, Vol. III, edited by C. Domb and M. S. Green (Academic, New York, 1974).

[13] D. W. Wood and H. P. Griffiths, J. Phys. A 9, 407 (1976).

[14] D. C. Rapaport, J. Phys. A 7, 1918 (1974).