PHYSICAL REVIEW B VOLUME 48, NUMBER 13 1 OCTOBER 1993-I

Constant-coupling approximation of the exchange-interaction model of ferromagnetism

H. H. Chen and Felix Lee
Department of Physics, National Tsing Hua University, Hsinchu, Taiwan 30043, Republic of China
(Received 12 October 1992)

The ferromagnetic-exchange-interaction model is studied by the Oguchi method and the constant-coupling approximation (CCA). The polarization $q(T)$, which describes the ordering of the model, is determined for various spins. The phase transition is first order for all spins $S \geq 1$. The transition temperature obtained with the CCA is $kT_c/J = 2/\{\ln S(2S-1) - \ln[(2S)^{\zeta} - S(2S+1)]\}$, where ζ depends on the lattice coordination number z and is given by $\zeta = 2(z-1)/z$. Both the CCA and the Oguchi method show that the discontinuity of q at T_c is $q_c = (2S-1)/2S$, which is exactly the same as the mean-field result.

I. INTRODUCTION

The Schrödinger exchange operator P_{ij} which permutes the spin coordinates of two particles i and j has been used to form an interaction Hamiltonian for a nearest-neighbor model of magnetism[1] which contains nonlinear terms in $\mathbf{S}_i \cdot \mathbf{S}_j$. The Hamiltonian of the exchange-interaction (EI) model is given by

$$H = -J \sum_{\langle ij \rangle} P_{ij} , \tag{1}$$

where J is the coupling constant and the summation is over all nearest-neighbor pairs of sites. The EI model is of theoretical interest and has been studied by various methods.[2–7]

The mean-field approximation (MFA), which is the simplest approach for general models, is not trivial for the EI model. In a recent article,[8] the MFA of the EI model was studied. The exchange operator is expressed as inner products of Hermitian spin tensor operators $Q_m^{(l)}$,

$$P_{ij} = \sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) Q_m^{(l)}(\mathbf{S}_i) Q_m^{(l)}(\mathbf{S}_j) , \tag{2}$$

where $A(S,l)$ are coefficients.[8] This model has $4S(S+1)$ order parameters $\langle Q_m^{(l)} \rangle$, which are thermal averages of $Q_m^{(l)}$ per spin. For any single-spin wave function $|\phi\rangle$, it is clear from the permutation property of P_{ij} that the product $|\phi(\mathbf{S}_1)\rangle |\phi(\mathbf{S}_2)\rangle \cdots |\phi(\mathbf{S}_N)\rangle$ (i.e., all spins are in the same state $|\phi\rangle$) is a ground state of the ferromagnetic EI model. At $T=0$, $\langle Q_m^{(l)} \rangle = \langle \phi | Q_m^{(l)} | \phi \rangle$. By assuming that all order parameters have the same temperature dependence

$$\langle Q_m^{(l)} \rangle = q(T) \langle \phi | Q_m^{(l)} | \phi \rangle, \quad l \neq 0 , \tag{3}$$

the MFA predicts that the phase transition of the model is first order with transition temperature $kT_c/J = z(2S-1)/(4S \ln 2S)$, where z is the coordination number of the lattice and the discontinuity of q at T_c is $q_c = (2S-1)/2S$.[8] The assumption [Eq. (3)] is quite reasonable because it has been shown that thermal fluctuations of all multipole moments of the EI model have exactly the same temperature dependence for all spins and for all lattices.[9] This assumption has also been verified numerically for several spins.[8]

For general spins the EI model has been studied by high-temperature series expansions,[2] Migdal-Kadanoff renormalizations,[4] and quantum Monte Carlo simulations.[3] These studies make use of the permutation property of the exchange operators, instead of expanding the exchange operators in terms of inner products of spin tensors. Therefore order parameters of the system cannot be obtained. Critical temperatures of the EI model are determined by assuming that the phase transitions are continuous (power-law singularity). Order parameters of the EI model have been calculated for $S=1$ by the constant-coupling approximation (CCA),[6] and a continuous phase transition is found. So far, only the MFA predicts first-order transitions for the EI model for all spins.

The purpose of this article is to extend the CCA study for the spin-1 system to general spins. Meanwhile, order parameters of the EI model for general spins are also studied by the Oguchi method.[10,11] We find that phase transitions of the EI model are first order in these methods. The previous CCA study for $S=1$, which showed a continuous phase transition,[6] was qualitatively incorrect. As the CCA is superior to the Oguchi method, this article puts emphasis on the CCA; detailed results of the Oguchi method will not be presented. In Sec. II we describe the Oguchi method briefly. The constant-coupling approximation[11,12] and its results for general spins are shown in Sec. III. A summary and discussions of our results are given in Sec. IV.

II. OGUCHI METHOD

In the Oguchi method, the interaction of a pair of nearest-neighbor spins, say, $\mathbf{S}_1$ and $\mathbf{S}_2$, is treated exactly and the interactions of $\mathbf{S}_1$ and $\mathbf{S}_2$ with their neighboring spins are replaced by effective-field terms in exactly the same way as the MFA. The Oguchi Hamiltonian for a pair of spins is given by

$$H_{\mathrm{Og}}=-JP_{12}-J(z-1)\sum_{l=0}^{2S}\sum_{m=-l}^{l}A(S,l)[Q_m^{(l)}(\mathbf{S}_1)+Q_m^{(l)}(\mathbf{S}_2)-\langle Q_m^{(l)}\rangle]\langle Q_m^{(l)}\rangle \ . \tag{4}$$

By assuming that all order parameters have the same temperature dependence $q(T)$ defined by Eq. (3), the Oguchi Hamiltonian reduces to[8]

$$\begin{aligned}H_{\mathrm{Og}}&=-JP_{12}-J(z-1)q[\rho_\phi(\mathbf{S}_1)+\rho_\phi(\mathbf{S}_2)]\\&\quad+J(z-1)\varepsilon_{\mathrm{Og}}(q)\\&\equiv H_2(J,q)+J(z-1)\varepsilon_{\mathrm{Og}}(q) \ ,\end{aligned}\tag{5}$$

where $\rho_\phi(\mathbf{S}_i)$ is the single-spin density matrix for the spin $\mathbf{S}_i$ to be in the pure state $|\phi\rangle$ and

$$\varepsilon_{\mathrm{Og}}(q)=(2Sq^2+2q-1)/(2S+1) \ . \tag{6}$$

The last term in the square brackets of Eq. (4) is a constant operator. Similar to the MFA, it is necessary to include this term to obtain the correct free energy of the system.

The free energy (in units of J) per pair of spins in the Oguchi method is

$$\begin{aligned}F_{\mathrm{Og}}(K,q)/J&=-(kT/J)\ln \mathrm{Tr}\exp(-H_2/kT)\\&\quad+(z-1)\varepsilon_{\mathrm{Og}}(q)\\&\equiv -K^{-1}\ln Z(K,q)+(z-1)\varepsilon_{\mathrm{Og}}(q) \ ,\end{aligned}\tag{7}$$

where $K=J/kT$. As shown in the Appendix,

$$\begin{aligned}Z(K,q)&=\mathrm{Tr}\exp[-H_2(J,q)/kT]\\&=\{\exp[K(z-1)q]+2S\}^2\cosh K\\&\quad+\{\exp[2K(z-1)q]+2S\}\sinh K \ .\end{aligned}\tag{8}$$

The polarization $q(T)$ defined by Eq. (3) is derived in the Appendix as

$$\begin{aligned}q&=(\{\exp[2K(z-1)q]\\&\quad+(2S-1)\exp[K(z-1)q]-2S\}\cosh K\\&\quad+\{\exp[2K(z-1)q]-1\}\sinh K)/Z(K,q)\\&\equiv G(K,q) \ .\end{aligned}\tag{9}$$

The above equation can also be obtained from $\partial F_{\mathrm{Og}}/\partial q=0$.

The solution $q(T)$ can be solved numerically. It behaves qualitatively the same as the MFA.[8] When $q\ll 1$, it can be shown that $(dq/dT)>0$ for $S\geq 1$. This indicates that the phase transition is first order except for $S=\frac{1}{2}$. The phase-transition temperature kT_c/J $(=1/K_c)$ and the discontinuity of q at T_c, denoted as q_c, are determined simultaneously by Eq. (9) and

$$F_{\mathrm{Og}}(K,q)=F_{\mathrm{Og}}(K,0) \ , \tag{10}$$

where $F_{\mathrm{Og}}(K,0)$ is the free energy of the disorder phase. We find that $q_c=(2S-1)/2S$ and K_c is the solution of

TABLE I. Phase-transition temperatures kT_c/J and zero-temperature polarization $q(T\to 0)$ obtained by the constant-coupling approximation (CCA) for some spins S and coordination numbers z. T_c obtained by the mean-field approximation (MFA) and the Oguchi method are included for comparison. For the MFA and the Oguchi method, $q(0)=1$.

Lattice z	Spin S	kT_c/J MFA	Oguchi	CCA	$q(T\to 0)$ CCA
6	$\frac{1}{2}$	3.0000	2.8597	1.8205	0.95614
6	1	2.1640	2.0539	1.1467	0.92415
6	$\frac{3}{2}$	1.8205	1.7208	0.7922	0.87828
6	2	1.6230	1.5288	0.4624	0.79750
6	$\frac{5}{2}$	1.4912	1.4004	$q=0$ for all T	
6	3	1.3953	1.3069	$q=0$ for all T	
8	$\frac{1}{2}$	4.0000	3.8910	2.8854	0.99139
8	1	2.8854	2.8005	1.9768	0.98666
8	$\frac{3}{2}$	2.4273	2.3506	1.5690	0.98159
8	2	2.1640	2.0916	1.3167	0.97613
8	$\frac{5}{2}$	1.9883	1.9184	1.1356	0.97022
8	3	1.8604	1.7922	0.9931	0.96378
12	$\frac{1}{2}$	6.0000	5.9244	4.9326	0.99951
12	1	4.3281	4.2698	3.4878	0.99926
12	$\frac{3}{2}$	3.6410	3.5885	2.8692	0.99901
12	2	3.2461	3.1965	2.5038	0.99876
12	$\frac{5}{2}$	2.9824	2.9347	2.2543	0.99851
12	3	2.7906	2.7440	2.0693	0.99825

$$S(2S+1)+S(2S-1)\exp(-2K) = \exp[K(z-1)(2S-1)/2S] \; . \quad (11)$$

When $S=\frac{1}{2}$, Eq. (11) reduces to the known result for the spin-$\frac{1}{2}$ Heisenberg model: $3+2\exp(-2K)=2K(z-1)$.

Phase-transition temperatures obtained by the Oguchi method are a few percent smaller than those determined by the MFA. T_c for some values of S and z are shown in Table I.

III. CONSTANT-COUPLING APPROXIMATION

In the CCA the effective fields are not simply proportional to $\langle Q_m^{(l)} \rangle$. We introduce a dimensionless parameter $h(K,q)$, which is to be determined later, and assume that the effective fields in the Oguchi method are changed by a factor h/q. The last factor $\langle Q_m^{(l)} \rangle$ in Eq. (4), which produces the effective fields, is multiplied by h/q. $\langle Q_m^{(l)} \rangle$ in the square brackets of Eq. (4) remains unchanged as it is the thermal average of $Q_m^{(l)}$. The pair Hamiltonian in the CCA is

$$\begin{aligned} H_{\mathrm{CC}}^{(2)} &= -JP_{12} - J(z-1)h[\rho_\phi(\mathbf{S}_1)+\rho_\phi(\mathbf{S}_2)] \\ &\quad + J(z-1)\varepsilon_{\mathrm{CC}}(h,q) \\ &= H_2(J,h)+J(z-1)\varepsilon_{\mathrm{CC}}(h,q) \; , \end{aligned} \quad (12)$$

where

$$\varepsilon_{\mathrm{CC}}(h,q)=(2Shq+2h-1)/(2S+1) \; . \quad (13)$$

Apart from the constant operators, the only difference between Eqs. (12) and (5) is the field parameter in H_2. It is straightforward to see that the polarization q in the CCA is

$$q=G(K,h) \; , \quad (14)$$

where $G(x,y)$ is defined in Eq. (9).

The field parameter h is determined by requiring that both the pair Hamiltonian and the single-particle Hamiltonian predict the same polarization $q(T)$. The single-particle Hamiltonian which has the same effective fields as in Eq. (12) is

$$H_{\mathrm{CC}}^{(1)} = -Jzh\rho_\phi(\mathbf{S}) + Jz\varepsilon_{\mathrm{CC}}(h,q)/2 \; . \quad (15)$$

From the MFA,[8] the polarization for the Hamiltonian $H_{\mathrm{CC}}^{(1)}$ is

$$q=[\exp(Kzh)-1]/[\exp(Kzh)+2S] \; . \quad (16)$$

For a given temperature (or K), the polarization q and the field parameter h are determined by solving Eqs. (14) and (16) simultaneously. Figure 1 shows $q(T)$ for some values of S for the body-centered-cubic lattice ($z=8$). From $q(T)$ the field parameter h can be obtained easily since Eq. (16) can be rewritten as

$$h=(Kz)^{-1}\ln[(1+2Sq)/(1-q)] \; . \quad (17)$$

Substituting Eq. (17) into Eq. (14), we obtain

$$\exp(2K)=\frac{2Sq(2X+2S-1)-(2S-1)(X-1)}{2X^2+(2S-1)X-(2S+1)-2q[X^2+2SX+S(2S+1)]} \equiv A(q)/B(q) \; , \quad (18)$$

where

$$X(q)=[(1+2Sq)/(1-q)]^{(z-1)/z} \; . \quad (19)$$

Some results can be obtained from Eq. (18).

(a) For $q \ll 1$ we expand the right-hand side of Eq. (18) in a power series of q. The zeroth-order term gives

$$kT/J=2/\ln[(z+2S-1)/(z-2S-3)] \; . \quad (20)$$

This is the temperature, called T_0, at which $q(T)$ intersects the T axis in Fig. 1. We note that T_0 exists only for $z>2S+3$. The first-order term in the expansion gives the slope of $q(T)$ at T_0. We find that the slope is positive for $S \geq 1$. The solution $q \ll 1$ is unstable near T_0, and the system will not have a second-order phase transition at T_0 for $S \geq 1$.

(b) It is known that the effective fields in the CCA are much smaller than those in the MFA. The fields are overcorrected at low temperatures such that the polarization $q(T)$ does not reach its saturation value $q=1$ as T approaches zero. This is the main drawback of the CCA. The value $q(T \to 0)$ is a positive root of $B(q)$ in Eq. (18). For a given value of z, $B(q)$ has one positive root when $2S \leq z-3$. And there exists a spin $S_{\max}$. When $(z-3)/2 < S \leq S_{\max}$, $B(q)$ has two positive roots which are the intersections of $q(T)$ with the q axis in Fig. 1.

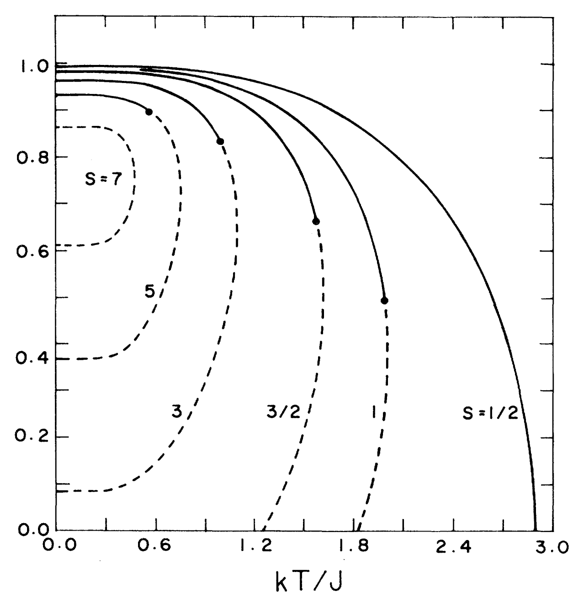

FIG. 1. Polarization $q(T)$ of the EI model obtained by the constant-coupling approximation for a body-centered-cubic lattice ($z=8$) for several spins. The dashed lines are metastable or unstable solutions.

When $S > S_{\max}$, $B(q)$ has no real roots. In this case a nontrivial solution for $q(T)$ does not exist. When $z=4$, $q(T)$ does not exist for any spin. For $z=6$, 8, and 12, we find numerically that $S_{\max}=2$, $\frac{15}{2}$, and 79, respectively. Some values of $q(T\to 0)$ are shown in Table I.

(c) As the phase transition is first order for $S \geq 1$, the phase-transition temperature should be determined by comparing the free energies of the order and disorder phases. There are two free energies involved in the CCA: the free energy of the single-particle Hamiltonian $F_{\rm CC}^{(1)}$ and that of the pair Hamiltonian $F_{\rm CC}^{(2)}$. From $F_{\rm CC}^{(i)} = -kT \ln \operatorname{Tr} \exp(-H_{\rm CC}^{(i)}/kT)$, we obtain

$$F_{\rm CC}^{(1)}(K,q,h)/J = -K^{-1}\ln[\exp(Kzh)+2S] + z\varepsilon_{\rm CC}(h,q)/2 \tag{21}$$

and

$$F_{\rm CC}^{(2)}(K,q,h)/J = -K^{-1}\ln Z(K,h) + (z-1)\varepsilon_{\rm CC}(h,q)\ , \tag{22}$$

where $Z(x,y)$ and $\varepsilon_{\rm CC}$ are given by Eqs. (8) and (13), respectively.

The phase-transition temperature T_c and the discontinuity of the polarization q_c are determined by Eq. (18), together with $F_{\rm CC}^{(1)}(K,q,h)=F_{\rm CC}^{(1)}(K,0,0)$ or with $F_{\rm CC}^{(2)}(K,q,h)=F_{\rm CC}^{(2)}(K,0,0)$. It is ambiguous whether $F_{\rm CC}^{(1)}$ or $F_{\rm CC}^{(2)}$ should be used. It turns out that the same results are obtained whether we use $F_{\rm CC}^{(1)}$ or $F_{\rm CC}^{(2)}$. We find that $q_c=(2S-1)/2S$, which is the same result as the MFA and the Oguchi method, and

$$kT_c/J = 2/\{\ln[S(2S-1)] - \ln[(2S)^{\zeta} - S(2S+1)]\}\ , \tag{23}$$

where $\zeta = 2(z-1)/z$. At T_c, the field parameter is $h_c = 2\ln(2S)/K_c z$. It can be shown that $F_{\rm CC}^{(1)}(K_c,q_c,h_c)=F_{\rm CC}^{(1)}(K_c,0,0)$ and $F_{\rm CC}^{(2)}(K_c,q_c,h_c) = F_{\rm CC}^{(2)}(K_c,0,0)$. We note that the free energy per spin in the cluster variation method[13] is $F_2=(z/2)F_{\rm CC}^{(2)} - (z-1)F_{\rm CC}^{(1)}$ for considering clusters up to two spins. Exactly the same results are obtained if we use F_2 instead of $F_{\rm CC}^{(1)}$ or $F_{\rm CC}^{(2)}$ to determine the critical properties.

For $S=\frac{1}{2}$, Eq. (23) reduces to the spin-$\frac{1}{2}$ Heisenberg result $kT_c/J=2/\ln[z/(z-4)]$. This temperature is a special case ($S=\frac{1}{2}$) of T_0 given by Eq. (20). In the CCA, T_c exists only when $z > 2\ln(2S)/\ln[4S/(2S+1)]$. Phase transitions occur for $S \leq 2$, $\frac{11}{2}$, and $\frac{57}{2}$ when $z=6$, 8, and 12, respectively. As shown in Fig. 1, the solution $q(T)$ exists for $S=7$, but T_c does not exist because this solution is either unstable or metastable at all temperatures.

IV. SUMMARY AND DISCUSSION

We have studied the ferromagnetic EI model for general spins by using the Oguchi method and the CCA. By assuming that all order parameters $\langle Q_m^{(l)}\rangle$ have the same temperature dependence $\langle Q_m^{(l)}\rangle = q(T)\langle\phi|Q_m^{(l)}|\phi\rangle$, the polarizations $q(T)$ for various lattices have been calculated. When $S=\frac{1}{2}$ the present work reproduces the results for the spin-$\frac{1}{2}$ Heisenberg model[11] $[q(T)=2\langle S_z\rangle]$. For $S \geq 1$ both the Oguchi method and the CCA predict that the system undergoes a first-order phase transition. The discontinuity of the polarization at T_c is $q_c=(2S-1)/2S$, which is exactly the same as the MFA result. It was unexpected that the MFA, Oguchi method, and CCA would predict the same q_c. Probably, this value will be correct in more exact theories.

In the MFA the EI model has a phase transition for any lattice. In the CCA a phase transition exists only when $z > 2\ln(2S)/\ln[4S/(2S+1)]$, and all transitions are first order (except for $S=\frac{1}{2}$) with the transition temperatures given by Eq. (23). There are no phase transitions for all spins when $z=4$ and for $S>2$ when $z=6$. Although the CCA is a considerable improvement over the MFA, it is the disadvantage of the CCA (and the Oguchi method) that thermodynamic properties of the system depend only on the coordination number z, but not on other details of the lattice. Both the two-dimensional (2D) square lattice and the 3D diamond lattice have $z=4$; the 2D triangular lattice and the 3D simple cubic lattice have $z=6$. As the EI model has spin-rotational symmetry, the Mermin-Wagner-Thorpe theorem[14] is valid, and there cannot be a finite T_c in two dimensions, while it is generally believed that a 3D spin model with ferromagnetic nearest-neighbor interactions has a phase transition.[15] Therefore the CCA and the Oguchi method are not expected to give good results for $z=4$ and 6. For the body-centered-cubic lattice ($z=8$) and the face-centered-cubic lattice ($z=12$), the CCA results shown in Table I should be quite reasonable. For the linear chain ($z=2$), the CCA (but not the Oguchi method) predicts the correct result that long-range order does not exist at finite temperatures. It is also expected that the CCA gives good results in high dimensions (or larger values of z).

As mentioned in Sec. I, a CCA study of the spin-1 EI model by Brown[6] reported that the phase transition of the system is second order. This result disagrees with what we have obtained. Brown assumed that the system has two order parameters $\langle S_z\rangle$ ($=\langle Q_0^{(1)}\rangle$ in our notation) and $\langle S_z^2\rangle$ [$=2(\langle Q_0^{(2)}\rangle+1)/3$ in our notation; S_z^2 is not traceless]. He obtained a second-order phase transition at the temperature $kT/J=2/\ln[(z+1)/(z-5)]$, which is the $S=1$ result of our T_0 [Eq. (20)]. But we have shown in Sec. III that T_0 is not a phase-transition temperature for $S>\frac{1}{2}$ and long-range order exists for certain temperatures higher than T_0.

Brown's study is equivalent to choosing $|\phi\rangle=|1\rangle$, $|0\rangle$, or $|-1\rangle$ in our method and allows $\langle Q_0^{(1)}\rangle$ and $\langle Q_0^{(2)}\rangle$ to have different thermal variations. For $|\phi\rangle=|\pm 1\rangle$, the nonzero moments of the spin-1 system are $\langle\phi|Q_0^{(1)}|\phi\rangle=\pm 1$ and $\langle\phi|Q_0^{(2)}|\phi\rangle=\frac{1}{2}$, and for $|\phi\rangle=|0\rangle$ the only nonzero moment is $\langle\phi|Q_0^{(2)}|\phi\rangle=-1$. We have repeated Brown's calculations. In general, we find that there are six sets of nontrivial solutions. Consider the body-centered-cubic lattice ($z=8$) for illustration. Brown did not find any solution of $kT/J > 1.82048$, but we find six solutions for each temperature $kT/J < kT_m/J = 1.99707$. For example, at $kT/J = 1.97679\ldots$, which is our T_c given by Eq. (23), the six sets of solutions in Brown's notations $(\langle S_z \rangle, \langle S_z^2 \rangle)$ are $(\frac{1}{2},\frac{5}{6})$, $(0,\frac{1}{3})$, $(-\frac{1}{2},\frac{5}{6})$, $(\frac{1}{4},\frac{3}{4})$, $(0,\frac{1}{2})$, and $(-\frac{1}{4},\frac{3}{4})$.

The six sets of solutions, when expressed in $\langle Q_0^{(l)} \rangle$, are simply $\langle Q_0^{(l)} \rangle = q(T) \langle \phi | Q_0^{(l)} | \phi \rangle$ $(l=1,2)$ for $|\phi\rangle = |1\rangle$, $|0\rangle$, and $|-1\rangle$, respectively. For each temperature $T < T_m$, $q(T)$ has two values which are the solutions of Eq. (18). When $T > T_0$ both solutions are positive, and for $T < T_0$ one solution becomes negative. The negative solution of $q(T)$ is unstable and is not shown in Fig. 1. This calculation shows that the spin-1 EI model undergoes a first-order phase transition; it also provides a numerical verification of our assumption that all order parameters have the same thermal behavior.

ACKNOWLEDGMENTS

This work was supported by the National Science Council of the Republic of China under Contract No. NSC 82-0208-M007-016.

APPENDIX: DERIVATION OF THE PARTITION FUNCTION AND THE POLARIZATION

Consider a two-particle Hamiltonian H_2 of the form

$$-H_2/kT = KP_{12} + L[\rho_\phi(\mathbf{S}_1) + \rho_\phi(\mathbf{S}_2)] , \tag{A1}$$

where $K = J/kT$ and $L = J(z-1)q/kT$ for the Oguchi method or $L = J(z-1)h/kT$ for the constant-coupling approximation. ρ_ϕ is the single-spin density matrix[8] for a pure state $|\phi\rangle$:

$$\rho_\phi(\mathbf{S}_i) = \sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) \langle \phi | Q_m^{(l)} | \phi \rangle Q_m^{(l)}(\mathbf{S}_i) . \tag{A2}$$

Since P_{12} commutes with $\rho_\phi(\mathbf{S}_1) + \rho_\phi(\mathbf{S}_2)$ and $\rho_\phi(\mathbf{S}_1)$ commutes with $\rho_\phi(\mathbf{S}_2)$, the partition function can be expressed as

$$\begin{aligned} Z &= \mathrm{Tr}\exp(-H_2/kT) \\ &= \mathrm{Tr}\exp(KP_{12})\exp[L\rho_\phi(\mathbf{S}_1)]\exp[L\rho_\phi(\mathbf{S}_2)] . \end{aligned} \tag{A3}$$

By expanding the exponential functions in a power series, applying the relations $P_{12}^{2n} = 1$, $\rho_\phi^{n+1} = \rho_\phi$, and rearranging terms, the partition function becomes

$$Z = \mathrm{Tr}(\cosh K + P_{12}\sinh K)[1 + (e^L - 1)\rho_\phi(\mathbf{S}_1)][1 + (e^L - 1)\rho_\phi(\mathbf{S}_2)] . \tag{A4}$$

Trace calculations involved in the above equation are

$$\begin{aligned} &\mathrm{Tr}1 = (2S+1)^2, \quad \mathrm{Tr}P_{12} = 2S+1, \quad \mathrm{Tr}\rho_\phi(\mathbf{S}_1) = \mathrm{Tr}\rho_\phi(\mathbf{S}_2) = 2S+1 , \\ &\mathrm{Tr}P_{12}\rho_\phi(\mathbf{S}_1) = \mathrm{Tr}P_{12}\rho_\phi(\mathbf{S}_2) = 1, \quad \mathrm{Tr}\rho_\phi(\mathbf{S}_1)\rho_\phi(\mathbf{S}_2) = \mathrm{Tr}P_{12}\rho_\phi(\mathbf{S}_1)\rho_\phi(\mathbf{S}_2) = 1 . \end{aligned} \tag{A5}$$

We obtain

$$Z = (e^L + 2S)^2 \cosh K + (e^{2L} + 2S)\sinh K . \tag{A6}$$

The order parameters are thermal averages of $Q_m^{(l)}(\mathbf{S}_1)$ [or $Q_m^{(l)}(\mathbf{S}_2)$] for $l \neq 0$. That is,

$$\begin{aligned} \langle Q_m^{(l)} \rangle &= Z^{-1}\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)\exp(-H_2/kT) \\ &= Z^{-1}\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)(\cosh K + P_{12}\sinh K)[1 + (e^L - 1)\rho_\phi(\mathbf{S}_1)][1 + (e^L - 1)\rho_\phi(\mathbf{S}_2)] . \end{aligned} \tag{A7}$$

With the aid of the relations

$$\begin{aligned} &\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1) = \mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)P_{12} = \mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)\rho_\phi(\mathbf{S}_2) = 0 , \\ &\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)\rho_\phi(\mathbf{S}_1) = (2S+1)\langle \phi | Q_m^{(l)} | \phi \rangle , \\ &\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)P_{12}\rho_\phi(\mathbf{S}_1) = \mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)P_{12}\rho_\phi(\mathbf{S}_2) = \langle \phi | Q_m^{(l)} | \phi \rangle , \\ &\mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)\rho_\phi(\mathbf{S}_1)\rho_\phi(\mathbf{S}_2) = \mathrm{Tr}Q_m^{(l)}(\mathbf{S}_1)P_{12}\rho_\phi(\mathbf{S}_1)\rho_\phi(\mathbf{S}_2) = \langle \phi | Q_m^{(l)} | \phi \rangle , \end{aligned} \tag{A8}$$

we find that

$$\begin{aligned} q &= \langle Q_m^{(l)} \rangle / \langle \phi | Q_m^{(l)} | \phi \rangle \\ &= Z^{-1}[(e^{2L} + (2S-1)e^L - 2S)\cosh K + (e^{2L} - 1)\sinh K] . \end{aligned} \tag{A9}$$

[1]G. A. T. Allan and D. D. Betts, Proc. Phys. Soc. London **91**, 341 (1967); R. I. Joseph, Phys. Rev. **163**, 523 (1967).
[2]H. H. Chen and R. I. Joseph, J. Math. Phys. **13**, 725 (1972).
[3]Y. C. Chen, H. H. Chen, and F. Lee, Phys. Lett. A **130**, 257 (1988); Y. C. Chen, Ph.D. thesis, National Tsing Hua University, Taiwan, 1988.
[4]H. H. Chen, Y. C. Chen, and F. Lee, Phys. Lett. A **125**, 235 (1987); H. H. Chen and F. Lee, Phys. Rev. B **42**, 10 540 (1990).
[5]E. B. Brown, Phys. Rev. B **40**, 775 (1989).
[6]H. A. Brown, Phys. Rev. B **31**, 3118 (1985).
[7]B. Sutherland, Phys. Rev. B **12**, 3295 (1975).
[8]H. H. Chen, S. C. Gou, and Y. C. Chen, Phys. Rev. B **46**, 8323 (1992).
[9]H. H. Chen and R. I. Joseph, Phys. Rev. B **2**, 2706 (1970).
[10]T. Oguchi, Prog. Theor. Phys. **13**, 148 (1955).
[11]J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966), Chaps. 4 and 5.
[12]P. W. Kasteleijn and J. van Kranendonk, Physica **22**, 317 (1956).
[13]T. Morita, J. Math. Phys. **13**, 115 (1972).
[14]M. F. Thorpe, J. Appl. Phys. **42**, 1410 (1971); N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).
[15]*Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1991), Vols. 1–14.