Exchange-interaction model on the simple-cubic lattice

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The ferromagnetic exchange-interaction model on the simple-cubic lattice is studied by considering the effective Hamiltonian of a cluster of eight spins which form a cubic unit cell. Through the use of the Bogoliubov inequality and a group-theoretical method, thermodynamic properties of the model are examined. Critical parameters are then determined for general spins.

I. INTRODUCTION

In the spin-$\frac{1}{2}$ Heisenberg model the interaction between a pair of spins has the property that it permutes the coordinates of the two spins. The generalization of the spin-permutation property to a system of higher spins leads to the exchange interaction (EI) model.[1,2] The EI model which contains nonlinear interactions $(\mathbf{S}_i\cdot\mathbf{S}_j)^n$ is of theoretical interest. Under the mean-field approximation, the spin-S EI model and the $(2S+1)$-state Potts model are shown to possess exactly the same thermodynamic properties.[3]

As the EI model is a generalization of the Heisenberg model, previous studies presumed that the EI model is characterized by second-order phase transitions with which the critical temperatures and critical exponents of the EI model are determined.[1,2,4,5] Recently, the order parameter of the ferromagnetic EI model has been studied by using the mean-field theory[6] and the constant-coupling approximation.[7] It is found that the system undergoes a first-order phase transition for $S>\frac{1}{2}$. In these methods, thermodynamic properties depend only on the coordination number z, rather than on the detailed structure of the lattice. Generally, the results obtained by the mean-field theory are correct only for $z\gg 1$, while the constant-coupling approximation gives a correct result for the linear chain, and predicts reasonably good results for the body-centered ($z=8$) and the face-centered ($z=12$) cubic lattices.[8]

The Hamiltonian of the EI model is isotropic in spin space. It is shown rigorously by Thorpe[9] that such a system exhibits no long-range order at finite temperatures for one- and two-dimensional lattices. Therefore, the plane triangular lattice, which has $z=6$, cannot have a phase transition. On the other hand, the simple cubic (sc) lattice, which has the same coordination number $z=6$, is believed to have a phase transition. Therefore, the constant-coupling approximation is not expected to give good results for the sc lattice. The purpose of this paper is to study the ferromagnetic EI model on the sc lattice. We consider a cluster of eight spins which form a cubic unit cell of the lattice. Interactions within the cluster are treated exactly, and the effects of other spins are described by an effective field which is then determined self-consistently.

II. CLUSTER HAMILTONIAN AND THERMODYNAMIC PROPERTIES

The Hamiltonian of the EI model is given by

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

where P_{ij} is the spin-exchange operator which permutes the spin coordinates of $\mathbf{S}_i$ and $\mathbf{S}_j$, J is the coupling constant, and the summation is taken over all nearest-neighbor pairs of spins. The exchange operator can be expressed in terms of Hermitian spin tensor operators (multipole moments) $Q_m^{(l)}$,

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

where $A(S,l)$ are constants.[6]

In the cluster approximation, we divide the system into equivalent clusters. The approximate Hamiltonian is

$$\mathcal{H}_A=\sum_n\mathcal{H}(c_n)\ , \tag{3}$$

where $\mathcal{H}(c_n)$ is the Hamiltonian of the nth cluster and the summation is over all clusters. The intracluster interactions are treated exactly and interactions between clusters are replaced by effective-field terms. Consider clusters of eight spins which form cubic unit cells. Each cluster contains twelve pair interactions $-JP_{ij}$ as shown by solid lines in Fig. 1. Each spin in the cluster has three field terms

$$-J\sum_{l,m}A(S,l)Q_m^{(l)}(\mathbf{S}_i)\langle Q_m^{(l)}\rangle$$

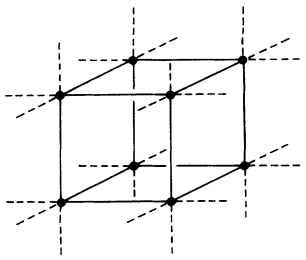

FIG. 1. A cluster of eight spins which form a cubic unit cell.

as represented by dashed lines. Here $\langle Q_m^{(l)} \rangle$ are thermal averages of the multipole moments $Q_m^{(l)}$.

The Hamiltonian of the eight-spin cluster shown in Fig. 1 is

$$\mathcal{H}(c_n) = -J \sum_{\langle ij \rangle \in c_n} P_{ij} - 3J \sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) \langle Q_m^{(l)} \rangle \sum_{i \in c_n} Q_m^{(l)}(\mathbf{S}_i) , \quad (4)$$

where $\langle ij \rangle \in c_n$ and $i \in c_n$ indicate that the summations are over bonds and sites of the cluster c_n. Since all clusters are equivalent, the subscript n may be omitted for convenience. For the ferromagnetic EI model, each spin is in the same state $|\phi\rangle$ at the zero temperature ($|\phi\rangle$ is any single-spin wave function). Since all multipole moments are equivalent for the EI model,[10] it is reasonable to assume that[6]

$$\langle Q_m^{(l)} \rangle = q(T) \langle \phi | Q_m^{(l)} | \phi \rangle \quad \text{for } l \neq 0 , \quad (5)$$

where $q(T)$ is called the polarization (or the order parameter) of the system. Equation (4) then reduces to

$$\mathcal{H}(c) = -J \sum_{\langle ij \rangle \in c} P_{ij} - 3Jq \sum_{i \in c} \rho_\phi(\mathbf{S}_i) - 24J(1-q)/(2S+1) , \quad (6)$$

where $\rho_\phi(\mathbf{S}_i)$ is the single-spin density matrix[6] for the spin $\mathbf{S}_i$ to be in the pure state $|\phi\rangle$,

$$\rho_\phi(\mathbf{S}_i) = \sum_{l=0}^{2S} \sum_{m=-l}^{l} A(S,l) \langle \phi | Q_m^{(l)} | \phi \rangle Q_m^{(l)}(\mathbf{S}_i) . \quad (7)$$

For any approximate Hamiltonian $\mathcal{H}_A$, the Bogoliubov inequality[11] is stated as

$$F_{\text{exact}} \leq F \equiv -kT \ln \operatorname{Tr} \exp(-\mathcal{H}_A/kT) + \langle \mathcal{H} - \mathcal{H}_A \rangle , \quad (8)$$

where F_{exact} is the exact free energy and $\langle \cdots \rangle$ is the thermal average taken over the Hamiltonian $\mathcal{H}_A$. For a system of N spins, there are $N/8$ clusters. It is straightforward to show that

$$\langle \mathcal{H} - \mathcal{H}_A \rangle = (N/8)(12J) \left[\sum_{l=1}^{2S} \sum_{m=-l}^{l} A(S,l) q^2 \langle \phi | Q_m^{(l)} | \phi \rangle^2 + \frac{1}{2S+1} \right] = (N/8)(12J)(2Sq^2+1)/(2S+1) . \quad (9)$$

Let $K = J/kT$. The free energy defined in Eq. (8) becomes

$$F(K,q) = \frac{N}{8} \left[-kT \ln \operatorname{Tr} \exp \left[K \sum_{\langle ij \rangle \in c} P_{ij} + 3Kq \sum_{i \in c} \rho_\phi(\mathbf{S}_i) \right] + 12J \frac{2Sq^2+2q-1}{2S+1} \right] \equiv \frac{N}{8} \left[-kT \ln Z_c(K,q) + 12J \frac{2Sq^2+2q-1}{2S+1} \right] . \quad (10)$$

As F gives an upper bound to the exact free energy, the stable solution of the order parameter $q(T)$ is the one which minimizes $F(K,q)$. Therefore, the self-consistent equation for $q(T)$ can be obtained from $\partial F/\partial q = 0$. If we define $L = Kq$, then $\partial F/\partial q = 0$ leads to

$$\frac{24(2Sq+1)}{2S+1} = \frac{\partial}{\partial L} \ln Z_c(K,L) . \quad (11)$$

For the sc lattice of N sites there are $3N$ bonds. The internal energy per spin U/N is

$$\frac{U}{N} = -3J \langle P_{ij} \rangle = -\frac{J}{4} \left\langle \sum_{\langle ij \rangle \in c} P_{ij} \right\rangle = -\frac{J}{4} \frac{\partial}{\partial K} \ln Z_c(K,L) . \quad (12)$$

The dimensionality of matrices involved is $(2S+1)^8 \times (2S+1)^8$. For example, it is $390\,625 \times 390\,625$ for $S=2$. Numerical calculations of $Z_c(K,L)$ and its derivatives are highly nontrivial, especially for large spins. We developed a group-theoretical technique which simplifies the calculations considerably.

III. APPLICATION OF GROUP THEORY

For a cluster of n spins the matrices of the operators P_{ij} and their products form a $(2S+1)^n$-dimensional representation (called the Γ representation) of the symmetric group of degree n, denoted as $\mathcal{S}_n$. The group elements can be divided into classes.[12] The number of elements in the class κ will be denoted as h_κ, and the character of this class in the irreducible representation ν will be denoted as $\chi_\kappa^{(\nu)}$. Chen and Joseph[4] have proved the following theorem which is very useful for the EI model.

Theorem I. If $\mathcal{R}$ is any matrix which commutes with all elements of $\mathcal{S}_n$ in a matrix representation Γ, then

$$\operatorname{Tr}[(\mathcal{P}^{(\Gamma)})^m \mathcal{R}] = \sum_\nu \sum_\kappa \frac{h_\kappa}{n!} \chi_\kappa^{(\nu)} \operatorname{Tr}(\mathcal{P}^{(\nu)})^m \operatorname{Tr}(P_\kappa^{(\Gamma)} \mathcal{R}) , \quad (13)$$

where m is any integer and $\mathcal{P}^{(\Gamma)}$ and $\mathcal{P}^{(\nu)}$ are sums of elements in $\mathcal{S}_n$ in the Γ- and ν-irreducible representations, respectively. P_κ is any element in the class κ and the summations are taken over all irreducible representations ν and all classes κ. This theorem can be generalized to the following.[13]

Theorem II. For any matrix $\mathcal{R}$ commuting with all elements in $\mathcal{S}_n$ in a matrix representation Γ, if $f(\mathcal{P})$ is an analytical function of $\mathcal{P}$, then

$$\operatorname{Tr}[f(\mathcal{P}^{(\Gamma)}) \mathcal{R}] = \sum_\nu \sum_\kappa \frac{h_\kappa}{n!} \chi_\kappa^{(\nu)} \operatorname{Tr}[f(\mathcal{P}^{(\nu)})] \operatorname{Tr}(P_\kappa^{(\Gamma)} \mathcal{R}) . \quad (14)$$

The proof of Theorem II is straightforward. We express $f(\mathcal{P}^{(\Gamma)})$ as a power series of $\mathcal{P}^{(\Gamma)}$, apply Theorem I, and then resume the power series of $\mathcal{P}^{(\nu)}$ to obtain $f(\mathcal{P}^{(\nu)})$.

With Theorem II we can calculate $Z_c(K,L)$ and its derivatives shown in Eqs. (10)–(12). If we define $\mathcal{P}=\sum_{\langle ij\rangle\in c}P_{ij}$,

$$f(\mathcal{P}^{(\Gamma)})=\exp(K\mathcal{P}^{(\Gamma)}) ,$$

and

$$\mathcal{R}=\exp\left[3L\sum_{i\in c}\rho_\phi(\mathbf{S}_i)\right] ,$$

then the left-hand side of Eq. (14) is equal to the partition function $Z_c(K,L)$. It is also straightforward to see that $\mathcal{P}^{(\Gamma)}$ and $\mathcal{R}$ commute. Therefore,

$$Z_c(K,L)=\sum_\nu\sum_\kappa\frac{h_\kappa}{n!}\chi_k^{(\nu)}A^{(\nu)}(K)B_k(L) , \tag{15}$$

where $A^{(\nu)}(K)$ and $B_\kappa(L)$ are defined and calculated as follows:

$$A^{(\nu)}(K)\equiv\mathrm{Tr}\left[\exp\left[K\sum_{\langle ij\rangle\in c}P_{ij}^{(\nu)}\right]\right]$$

$$=\sum_\alpha\exp(K\lambda_\alpha^{(\nu)}) , \tag{16}$$

where $\lambda_\alpha^{(\nu)}$ are eigenvalues of the operator

$$\mathcal{P}^{(\nu)}\equiv\sum_{\langle ij\rangle\in c}P_{ij}^{(\nu)} .$$

The explicit forms of $P_{ij}^{(\nu)}$ in any irreducible representation can be obtained by the technique introduced by Yamanouchi.[14] We find $P_{ij}^{(\nu)}$ and then diagonalize $\mathcal{P}^{(\nu)}$ to obtain $\lambda_\alpha^{(\nu)}$. For $\mathcal{S}_8$, there are 22 irreducible representations, and the size of the greatest matrices is of dimension 90×90 (for $\nu=[4211]$).

The matrices $P_\kappa^{(\Gamma)}$ and $\mathcal{R}$ are of dimension $(2S+1)^8\times(2S+1)^8$. But $B_\kappa(L)$ can be calculated analytically. there are 22 classes. For the class $\kappa=\{a_1a_2a_3\cdots\}$ ($a_i\geq1$, $a_i\geq a_{i+1}$, $\sum a_i=n=8$) it can be shown that

$$B_\kappa(L)\equiv\mathrm{Tr}(P_\kappa^{(\Gamma)}\mathcal{R})$$

$$=\prod_i[\exp(3a_iL)+2S] \text{ for } \kappa=\{a_1a_2a_3\cdots\} . \tag{17}$$

From Eqs. (15) to (17), we can calculate the partition function $Z_c(K,L)$ for any values of K and q ($L=Kq$). The derivatives $\partial A^{(\nu)}/\partial K$ and $\partial B_\kappa/\partial L$ can also be obtained easily from Eqs. (16) and (17). With these derivatives, $(\partial/\partial K)\ln Z_c$ and $(\partial/\partial L)\ln Z_c$ can be calculated.

We note that the method developed here for the eight-spin cluster can also be applied to the cluster of four spins which form a square, and to the cluster of two spins. For other clusters, $\mathcal{P}$ and $\mathcal{R}$ in Theorem II do not commute, and the group-theoretical method is no longer applicable. Therefore, the extension of the study to larger clusters is extremely difficult.

TABLE I. Critical parameters obtained from the eight-spin cluster.

S	kT_c/J	q_c	L/NJ
$\frac{1}{2}$	2.5363	0	0
1	1.8126	0.4917	0.2078
$\frac{3}{2}$	1.5089	0.6543	0.3924
2	1.3326	0.7351	0.5020
$\frac{5}{2}$	1.2141	0.7834	0.5663
3	1.1275	0.8155	0.6038
10	0.7231	0.9316	0.5496
20	0.5780	0.9612	0.3736
100	0.3729	0.9912	0.0646

IV. RESULTS AND DISCUSSIONS

For a given value of S the polarization $q(T)$ is obtained by solving Eq. (11) for each temperature. In general, there are three solutions: (i) the trivial solution $q=0$, (ii) a solution with $dq/dT<0$, and (iii) a solution with $dq/dT>0$. The solution with $dq/dT>0$, which is unstable, exists only in a small range of temperatures. The free energy as a function of q is a maximum at this solution. The solution with $dq/dT<0$, which describes the ordered phase, exists at low temperatures. This solution has the lowest free energy and is the stable solution below the ordering temperature T_c. It becomes metastable for temperatures above T_c. It is the trivial solution which has the lowest free energy, and is stable above T_c. In Fig. 2, we plot nontrivial solutions $q(T)$ for several spins. The stable solutions are shown by solid lines, and the metastable or unstable solutions by dashed lines. The internal energies, calculated by Eq. (12) for the stable $q(T)$, are given in Fig. 3. The phase transitions are first order for $S>\frac{1}{2}$. In Table I, we present the ordering temperatures, kT_c/J, discontinuities of q at T_c, denoted q_c, and the latent heats L/NJ (discontinuities of U/NJ at T_c) for several spins.

Consider the latent heat first. We see from Table I that L is small for small values of S and for $S\gg1$. The latent heat as a function of the spin has a maximum. This is qualitatively different from the mean-field result,

$$L/NJ=3(2S-1)^2/[2S(2S+1)] ,$$

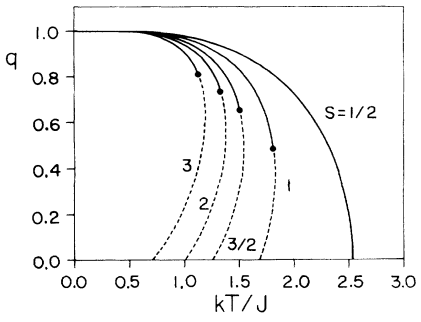

FIG. 2. Polarizations (or order parameters) $q(T)$ versus T for several spins.

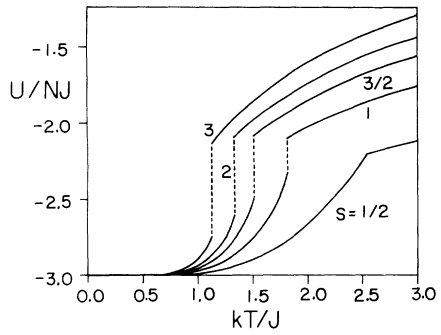

FIG. 3. Internal energies U/NJ versus T for several spins.

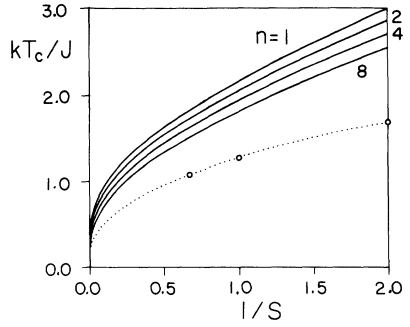

FIG. 4. Ordering temperatures kT_c/J versus $1/S$ for different cluster sizes n. The high-temperature series-expansion results are shown by open circles, and the predicted T_c for $n \rightarrow \infty$ is represented by the dotted line.

which approaches 3 for $S \gg 1$. As mentioned above, the cluster approximation described for the eight-spin ($n=8$) cluster can be applied to clusters of four ($n=4$) and two ($n=2$) spins, respectively. We have also calculated L/NJ for $n=2$ and 4. The latent heat has a peak for each $n \geq 2$. If we regard S as a continuous variable, the maxima of L/NJ occur at $1/S=0.27$, 0.23, and 0.21, for $n=2$, 4, and 8, respectively. The peak shifts to larger S when n increases. The peak might occur at $1/S=0$ when $n \rightarrow \infty$. More studies are needed to confirm this point.

In the mean-field approximation,[6] the Oguchi method and the constant-coupling approximation,[7] q_c are found to be $(2S-1)/2S$. In the present calculations for the four- and eight-spin clusters, q_c are smaller than $(2S-1)/2S$, but the differences are within a few percent.

In order to see how the ordering temperatures depend on the cluster size n, we consider S as a continuous variable and plot kT_c/J versus $1/S$ in Fig. 4 for $n=1$, 2, 4, and 8, respectively. The $n=1$ result is simply the mean-field solution[6] and the result for $n=2$ is what is obtained by the Oguchi method.[7] When $S=\frac{1}{2}$ the EI model is identical to the Heisenberg model. Based on the high-temperature susceptibility series of 10 terms,[15] kT_c/J for the sc lattice is determined to be 1.677 ± 0.003. For the EI model ($S > \frac{1}{2}$), high-temperature susceptibility series are available only for seven terms.[4] The series coefficients are irregular for large spins. For the sc lattice some series coefficients become negative for $S \geq \frac{3}{2}$. Therefore, reliable estimates of T_c are obtained only for

$$S=1 \ (kT_c/J=1.27 \pm 0.02)$$

and

$$S=\tfrac{3}{2} \ (kT_c/J=1.07 \pm 0.03) \ .^{4}$$

For comparison, kT_c/J obtained by the high-temperature series-expansion (HTSE) method are shown by open circles in Fig. 4.

Usually, HTSE results are considered to be the "best" values when exact or rigorous results are unavailable. But it is important to note that in the HTSE method, T_c is analyzed by assuming that the phase transition is second order.[16] One determines the temperature at which the susceptibility diverges. If the system undergoes a first-order phase transition, this temperature is not the ordering temperature, but is the temperature, called T_m, at which the order parameter has an infinite slope. For the EI model on the sc lattice, we see from Fig. 2 that T_m is very close to the ordering temperature, especially for small values of S. The percentaged difference between T_c and T_m for each spin is within the error bar of the estimate of T_c in the HTSE method. It is then reasonable to believe that the series-expansion results for the EI model on the sc lattice are accurate, although the phase transitions are first order.

The ordering temperatures obtained in the present study for the eight-spin cluster are higher than the HTSE results. If we assume that results of the cluster approximation will converge uniformly to the HTSE results when $n \rightarrow \infty$, then the best estimates of the ordering temperatures for general values of S may be given by the dotted line shown in Fig. 4.

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[1]R. I. Joseph, Phys. Rev. **163**, 523 (1967).
[2]G. A. T. Allen and D. D. Betts, Proc. Phys. Soc. London **91**, 341 (1967).
[3]H. H. Chen, S.-C. Gou, and Y. C. Chen, Phys. Lett. A **177**, 248 (1993).
[4]H. H. Chen and R. I. Joseph, J. Math. Phys. **13**, 725 (1972).
[5]H. H. Chen and F. Lee, Phys. Rev. B **42**, 10 540 (1990).
[6]H. H. Chen, S.-C. Gou, and Y. C. Chen, Phys. Rev. B **46**, 8323 (1992).
[7]H. H. Chen and F. Lee, Phys. Rev. B **48**, 9456 (1993).
[8]J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966).
[9]M. F. Thorpe, J. Appl. Phys. **42**, 1410 (1971).
[10]H. H. Chen and R. I. Joseph, Phys. Rev. B **2**, 2706 (1970).
[11]H. Falk, Am. J. Phys. **38**, 858 (1970).
[12]J.-Q. Chen, *Group Representation Theory for Physicists* (World Scientific, Singapore, 1989).
[13]S.-C. Gou, Ph.D. thesis, National Tsing Hua University, Taiwan, 1992.
[14]T. Yamanouchi, Proc. Phys. Math. Soc. Jpn. **19**, 436 (1937).
[15]G. A. Baker, Jr., H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Rev. **164**, 800 (1967).
[16]C. Domb and M. S. Green, eds., *Phase Transitions and Critical Phenomena* (Academic, New York, 1974), Vol. 3.