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High temperature free energy of the Schrödinger model of ferromagnetism for arbitrary single-ion contributions

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Abstract. We consider a system described by the hamiltonian

$$\mathscr{H} = -J \sum_{\langle i,j \rangle} P_{ij} - \sum_{i=1}^{N} Q_i(\xi)$$

where the first term corresponds to the Schrödinger spin S exchange model and the second term represents an arbitrary single-ion contribution (characterized by the set of parameters ξ). Terms in the free energy through $(J/k_B T)^7$ have been obtained for the face-centred cubic, body-centred cubic, simple cubic, plane triangular and plane square lattices; explicit results for the face-centred cubic lattice are given in an appendix.

1. Introduction

Consider a system of N spin S particles on a lattice interacting via the hamiltonian

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1 \tag{1.1}$$

where

$$\mathscr{H}_0 = -\sum_{i=1}^{N} Q_i(\xi) \tag{1.2}$$

and

$$\mathscr{H}_1 = -J \sum_{\langle i,j \rangle} P_{ij} \equiv -J\mathscr{P}. \tag{1.3}$$

Here $Q_i(\xi)$ denotes any single-particle operator acting on spin i; ξ is a set of characteristic parameters for Q_i. J is the magnitude of an exchange interaction between the pair of nearest neighbours $\langle i,j \rangle$. P_{ij} is the Schrödinger exchange operator (Schrödinger 1941) which has the property that it permutes the spin states of the two spins i and j. In general, P_{ij} is a polynomial of degree $2S$ in $(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)$. When $S = \frac{1}{2}$, P_{ij} becomes identical with the usual Heisenberg operator.

For the case $Q_i(H) = mHS_{iz}$ and $S = \frac{1}{2}$, H being a z-directed magnetic field, Baker *et al* (1967a, 1970) have computed the first eight terms in the high temperature expansion of the free energy for arbitrary H. On the other hand, Chen and Joseph (1972) have calculated the first seven terms in the high temperature expansion of the zero field free

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energy and susceptibility for arbitrary S. The purpose of the present paper is to develop the high temperature theory for the case of general S and for arbitrary functions $Q_i(\xi)$. The first seven terms of the free energy have been obtained for the face-centred cubic, body-centred cubic, simple cubic, plane triangular and plane square lattices; explicit results for the face-centred cubic lattice are given in an appendix.

2. Theory

The free energy F of the system is given by

$$F(\xi, T) = -k_B T \ln \operatorname{Tr} \exp[-\beta(\mathscr{H}_0 + \mathscr{H}_1)] \tag{2.1}$$

where k_B is the Boltzmann constant, T the thermodynamic temperature and $\beta = 1/k_B T$. Since $\mathscr{H}_0$ and $\mathscr{H}_1$ commute, we may expand F in the form

$$-\beta F = \ln \operatorname{Tr}\left(\sum_{n=0}^{\infty} \frac{K^n}{n!} \mathscr{P}^n \mathrm{e}^{-\beta \mathscr{H}_0} \right) = \ln \operatorname{Tr} \mathrm{e}^{-\beta \mathscr{H}_0} + \ln\left(1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} \langle \mathscr{P}^n \rangle \right) \tag{2.2}$$

where $K = J/k_B T$ and

$$\langle \mathscr{O} \rangle \equiv \frac{\operatorname{Tr} \mathscr{O} \mathrm{e}^{-\beta \mathscr{H}_0}}{\operatorname{Tr} \mathrm{e}^{-\beta \mathscr{H}_0}}. \tag{2.3}$$

We choose to evaluate these traces in the basis of eigenstates of Q_i, that is, in the basis $|M_1 M_2 \ldots M_N\rangle$ defined by

$$Q_i(\xi)|M_1 M_2 \ldots M_N\rangle = q(M_i; \xi)|M_1 M_2 \ldots M_N\rangle, \qquad i = 1, 2, \ldots, N. \tag{2.4}$$

Here $q(M_i; \xi)$ denotes an eigenvalue of $Q_i(\xi)$ and the M_i each have $2S+1$ possible values.

At this point it is convenient to define the set of functions G_l by

$$g_l \equiv \sum_{M} \mathrm{e}^{\beta l q(M;\xi)}, \tag{2.5}$$

$$G_l \equiv g_l/(g_1)^l \tag{2.6}$$

where the summation is over the $2S+1$ possible values of M, and l is a positive integer.

The permutation operators P_{ij} and their products form a symmetric group of order N, S_N. If the permutations of N integers are represented by the cyclic notation (Littlewood 1940), those elements of S_N with the same cycle structure form a class so that each partition of N defines a class of S_N. This class is denoted by the symbol $[1^{\alpha_1} 2^{\alpha_2} \ldots N^{\alpha_N}]$, $\sum_{l=1}^{N} l\alpha_l = N$, or more concisely by k.

Now, we may write the quantity $\mathscr{P}$ in the form

$$\mathscr{P} = \sum_{\langle i,j \rangle} P_{ij} = \sum_{t=1}^{x} p_t \tag{2.7}$$

where x is the number of nearest-neighbour pairs and p_t denotes an element in the class $[1^{N-2}2]$. We then have that

$$\mathscr{P}^n = \sum_{t_1=1}^{x} \cdots \sum_{t_n=1}^{x} p_{t_1} \cdots p_{t_n} = \sum_{r} p_r \tag{2.8}$$

where p_r is some element of S_N and the r summation runs over x^n terms.

Consider now a cycle of length l, denoted $(i_1 i_2 \dots i_l)$. When we operate with this on the substate $|M_{i_1} M_{i_2} \dots M_{i_l}\rangle$ we find that

$$(i_1 i_2 \dots i_l)|M_{i_1} M_{i_2} \dots M_{i_l}\rangle = |M_{i_2} \dots M_{i_l} M_{i_1}\rangle \tag{2.9}$$

by the definition of this operator. Hence we have that

$$\begin{aligned}\sum_{M_{i_1}} \dots \sum_{M_{i_l}} \langle M_{i_1} \dots M_{i_l}|(i_1 \dots i_l) \exp\left(\beta \sum_{r=1}^{l} Q_{i_r}(\xi)\right)|M_{i_1} \dots M_{i_l}\rangle \\ = \sum_{M_{i_1}} \dots \sum_{M_{i_l}} \langle M_{i_1} \dots M_{i_l}|M_{i_2} \dots M_{i_l} M_{i_1}\rangle \exp\left(\beta \sum_{r=1}^{l} q(M_{i_r}; \xi)\right) \\ = \sum_{M} e^{\beta l q(M;\xi)} = g_l. \end{aligned} \tag{2.10}$$

Consequently using the fact that $\operatorname{Tr} e^{-\beta \mathscr{H}_0} = (g_1)^N$, together with equation (2.10), and noting that p_r has the cycle structure $[1^{\alpha_1} 2^{\alpha_2} \dots N^{\alpha_N}]$ we have that

$$\langle p_r \rangle = \frac{(g_1)^{\alpha_1}(g_2)^{\alpha_2} \dots (g_N)^{\alpha_N}}{(g_1)^N} = (G_2)^{\alpha_2} \dots (G_N)^{\alpha_N} \equiv G^{(k)} \tag{2.11}$$

since $\sum_{l=1}^{N} l\alpha_l = N$ and $G_1 = 1$ by definition. Here $G^{(k)}$ denotes a function characteristic of the class k which is characterized by the partition $[1^{\alpha_1} 2^{\alpha_2} \dots N^{\alpha_N}]$. It then follows from equation (2.11) that $\langle p_r \rangle$ is determined solely by its cycle structure so that $\langle p_r \rangle$ is the same for all p_r in the same class.

Upon combining equations (2.8) and (2.11) we have that

$$\langle \mathscr{P}^n \rangle = \sum_r \langle p_r \rangle = \sum_k a_{n,k} G^{(k)} \tag{2.12}$$

where the k summation runs over all the classes of S_N and $a_{n,k}$ is just the number of elements which belong to the class k among the x^n elements of p_r.

The quantities $a_{n,k}$ in equation (2.12) can be expressed in terms of the irreducible representations of $\mathscr{P}$ in the following way. Let $\mathscr{P}^{(\nu)}$ denote the νth irreducible representation of $\mathscr{P}$. It then follows from equation (2.8) that

$$\operatorname{Tr}(\mathscr{P}^{(\nu)})^n = \operatorname{Tr} \sum_r p_r^{(\nu)} = \sum_k a_{n,k} \chi_k^{(\nu)} \tag{2.13}$$

where $\chi_k^{(\nu)}$ is the character of the kth class in the νth irreducible representation (Littlewood 1940). Using the orthogonality of characters, ie,

$$\sum_{\nu} \frac{h_k}{h} \chi_k^{(\nu)*} \chi_{k'}^{(\nu)} = \delta_{kk'}$$

we get from equation (2.13) the result

$$a_{n,k} = \frac{h_k}{h} \sum_{\nu} \chi_k^{(\nu)*} \operatorname{Tr}(\mathscr{P}^{(\nu)})^n. \tag{2.14}$$

Here h_k is the number of elements of the class k, $h = N!$ is the total number of elements of S_N and the ν summation is over all irreducible representations of S_N. Equation (2.14) is the basic result which we shall use in our calculations.

Combining equations (2.2) and (2.12) we have that

$$-\beta F = N \ln g_1 + \ln\left(1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} \sum_k a_{n,k} G^{(k)}\right). \tag{2.15}$$

In the thermodynamic limit we have

$$a_{n,k} = N b_{n,k} + O(N^2). \tag{2.16}$$

On making the moment to cumulant transformation, terms of $O(N^2)$ cancel exactly so that we may write the free energy in the final form

$$\frac{-\beta F}{N} = \ln g_1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} \sum_k b_{n,k} G^{(k)} = \ln g_1 + \sum_{n=1}^{\infty} \frac{K^n}{n!} F_n(\xi). \tag{2.17}$$

Since all of the p_t in equation (2.7) are in the class $[1^{N-2}2]$ we then see that the general structure of the term proportional to K^n in equation (2.17) is such that for $n = 1$ we have only a term G_2, while for $n = 2$ we have terms proportional to unity, $(G_2)^2$, and G_3 and for $n = 3$ terms in G_2, G_4, G_2G_3 and $(G_2)^3$, and so on.

3. The calculation

For an explicit evaluation of the quantities $b_{n,k}$ which enter into equation (2.17) we have made use of the finite cluster method of Domb (1960) and Rushbrooke (1964) as discussed in detail by Baker *et al* (1967a) and Chen and Joseph (1972). For a cluster of l lines and m vertices the quantity $a_{n,k}$ of equation (2.15) was evaluated by means of equation (2.14). In this case $\mathscr{P}$ will be the sum of p_t over the l bonds of the cluster while the order of the symmetric group is m. The irreducible representations of $P_{12}, P_{23}, \ldots, P_{m-1m}$ for S_m can be constructed from those of S_{m-1}, S_{m-2} by the method of Yamanouchi (1937). By multiplying these matrices we can get the irreducible representations of any P_{ij} appearing in the cluster.

The explicit calculations were performed on a high speed computer (CDC 6600). For $m = 2, 3, 4, 5, 6, 7, 8, 9, 10$ the largest sizes of the irreducible representation matrices of S_m are 1, 2, 3, 6, 16, 35, 90, 216, 768, respectively (Littlewood 1940). We were forced to stop our calculations at 7 line clusters, that is, we could only compute terms through K^7 in equation (2.17) because the matrices for 9 vertex clusters were too large for the computer to handle. A general listing of all necessary graphs, lattice constants, and the so called T matrix necessary for the computation can be found in the monumental work of Baker *et al* (1967b). The results of our calculation for the quantities $F_n = \Sigma_k b_{n,k} G^{(k)}$, $n = 1-7$, for the face-centred cubic lattice are given in the appendix. These quantities have also been computed for the body-centred cubic, simple cubic, plane triangular and plane square lattices and can be obtained from the authors on request. The functions G_l are given by equations (2.5) and (2.6). It is only through these terms that the function Q_i enters.

Since in a calculation of this kind numerical errors can enter at any stage, it is essential to have a careful checking procedure. The following four checks were made. First, it follows directly from equation (2.12) that for a l line cluster

$$\sum_k a_{n,k} = l^n. \tag{3.1}$$

This provides a simple and direct check. A second direct check was to confirm that a

cluster of l lines did not contribute to the term K^n when $n < l$. Our final two checks were to reduce our results to the independent results of Chen and Joseph (1972) and Baker *et al* (1970). In the former case we set $Q_i = mHS_{iz}$ and computed the zero field free energy and susceptibility by simply letting

$$G^{(k)} = Y^{-\Sigma_{l\geqslant 2}(l-1)\alpha_l}, \qquad \text{for the free energy;} \tag{3.2}$$

and

$$G^{(k)} = \tfrac{1}{3}S(S+1)Y^{-\Sigma_{l\geqslant 2}(l-1)\alpha_l}\left(\sum_{l\geqslant 2} l(l-1)\right), \qquad \text{for the zero field susceptibility;} \tag{3.3}$$

and collecting like powers of $Y(=2S+1)$ where $k = [1^{\alpha_1}2^{\alpha_2}\ldots l^{\alpha_l}\ldots]$. For the latter comparison we set $S = \frac{1}{2}$ and $Q_i = 2mHS_{iz}$. It then follows from equations (2.5) and (2.6) that

$$\begin{aligned} g_l &= 2\cosh ly \\ G_l &= \frac{1}{2^{l-1}}\sum_{j=0}^{E[\frac{1}{2}l]}\binom{l}{2j}(\tanh y)^{2j}, \end{aligned} \tag{3.4}$$

where $y = mH/k_BT$ and $E[x]$ is the largest integer not exceeding x. In this case $G^{(k)}$ is a finite polynomial in $(\tanh y)^2$. By grouping like powers of $(\tanh y)^2$ the free energy can be written in the form

$$\frac{-F(H,T)}{Nk_BT} = \ln(2\cosh y) + \sum_{n=1}^{\infty}\frac{K^n}{n!}\sum_{j=0}^{n} C_{n,j}(\tanh y)^{2j}. \tag{3.5}$$

The magnetization M was obtained by taking a derivative with respect to y and the resultant series was then inverted to the form

$$\tanh y = MC_0(K) + M^3C_1(K) + \ldots \tag{3.6}$$

so as to be able to compare our results directly with those presented by Baker *et al* (1970). Needless to say our results are consistent with all four checking procedures.

4. Discussion

Since our final results do not specify an explicit form for the single-ion function $Q_i(\xi)$, various kinds of high temperature series can be derived from the free energy. In view of the recent interest in the critical point behaviour of multicomponent systems (Griffiths and Wheeler 1970) and the universality hypothesis (Griffiths 1970) it seems desirable to analyse our series for a 'multi-field' system to see what effect variation of certain parameters might have on the properties of the system. A simple example is a system with $Q_i(H,D) = mHS_{iz} - DS_{iz}^2$ where D is the magnitude of a single-ion anisotropy. The properties of this system in the limits $D/k_BT \to \pm\infty$ have been studied in some detail by Kim and Joseph (1973a, b). When $S = 1$ a simple mean field treatment of this case shows that the system possesses a tricritical point in qualitative agreement with the spin one Ising interaction with quadratic terms considered by Blume *et al* (1971). Numerical analysis of our series is now underway to study this situation in more detail.

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Appendix. Face-centred cubic lattice

$$F_1 = 6G_2$$

$$F_2 = 6 + 132G_3 - 138G_2^2$$

$$F_3 = 36G_2 + 5\,532G_4 - 13\,320G_2G_3 + 7\,752G_2^3$$

$$\begin{aligned} F_4 = {} & -48 + 3\,252G_3 - 2\,952G_2^2 + 351\,000G_5 - 975\,312G_2G_4 - 421\,920G_3^2 \\ & + 1\,766\,736G_2^2G_3 - 720\,756G_2^4 \end{aligned}$$

$$\begin{aligned} F_5 = {} & -744G_2 + 351\,120G_4 - 740\,880G_2G_3 + 30\,107\,520G_6 + 391\,800G_2^3 \\ & - 95\,454\,720G_2G_5 - 76\,387\,680G_3G_4 + 186\,333\,840G_2^2G_4 \\ & + 161\,395\,200G_2G_3^2 - 300\,739\,680G_2^3G_3 + 94\,744\,224G_2^5 \end{aligned}$$

$$\begin{aligned} F_6 = {} & 2\,700 - 48\,672G_3 + 57\,132G_2^2 + 45\,874\,776G_5 - 113\,823\,984G_2G_4 \\ & - 47\,774\,232G_3^2 + 186\,401\,880G_2^2G_3 + 3\,267\,603\,360G_7 - 4\,116\,168\,720G_4^2 \\ & - 11\,672\,959\,680G_2G_6 - 8\,896\,664\,160G_3G_5 - 70\,723\,440G_2^4 \\ & + 5\,732\,579\,520G_3^3 + 39\,705\,197\,760G_2G_3G_4 + 24\,765\,665\,760G_2^2G_5 \\ & - 41\,557\,487\,040G_2^3G_4 - 54\,041\,562\,720G_2^2G_3^2 + 62\,998\,179\,840G_2^4G_3 \\ & - 16\,184\,350\,080G_2^6 \end{aligned}$$

$$\begin{aligned} F_7 = {} & 123\,360G_2 + 352\,800G_4 + 5\,972\,400G_2G_3 + 7\,072\,108\,680G_6 - 5\,349\,120G_2^3 \\ & - 20\,348\,137\,152G_2G_5 - 15\,692\,814\,480G_3G_4 + 429\,825\,281\,760G_8 \\ & + 36\,218\,988\,288G_2^2G_4 + 30\,607\,129\,224G_2G_3^2 - 1\,111\,793\,558\,400G_4G_5 \\ & - 1\,260\,893\,632\,320G_3G_6 - 1\,710\,559\,408\,320G_2G_7 - 53\,867\,000\,880G_2^3G_3 \\ & + 3\,943\,181\,723\,040G_2^2G_6 + 16\,007\,110\,560G_2^5 + 6\,022\,790\,605\,440\,G_2G_3G_5 \\ & + 2\,788\,308\,411\,840G_2G_4^2 + 2\,414\,839\,452\,480G_3^2G_4 \\ & - 6\,993\,526\,901\,760G_2^3G_5 - 16\,842\,341\,154\,240G_2^2G_3G_4 \\ & - 4\,867\,172\,755\,200G_2G_3^3 + 18\,710\,029\,991\,520G_2^3G_3^2 \\ & + 10\,782\,927\,912\,960G_2^4G_4 - 15\,716\,035\,601\,280G_2^5G_3 \\ & + 3\,410\,421\,148\,800G_2^7. \end{aligned}$$

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