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1976 J. Phys. A: Math. Gen. 9 105

(http://iopscience.iop.org/0305-4470/9/1/016)

J. Phys. A: Math. Gen., Vol. 9, No. 1, 1976. Printed in Great Britain. © 1976

High-temperature series for $S = \frac{1}{2}$ with anisotropic exchange

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Received 9 July 1975

Abstract. For the nearest-neighbour $S = \frac{1}{2}$ Hamiltonian

$$\mathscr{H} = -2 \sum_{\langle ij \rangle} [J_\perp(S_i^x S_j^x + S_i^y S_j^y) + J_\parallel S_i^z S_j^z] + H \sum_j S_j^z$$

the expansion for the logarithm of the partition function is calculated as

$$\ln(Z) = \sum_{ij} a(J_\perp, J_\parallel)_{ij} \beta^i H^j,$$

where $\beta = 1/kT$. It is shown that from the zero-field coefficients $a(J_\perp, J_\parallel)_{i0}$, the corresponding series for the more general Hamiltonian

$$\mathscr{H}' = -2 \sum_{\langle ij \rangle} (J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_z S_i^z S_j^z)$$

can be calculated up to a certain maximum order in β. This maximum order depends on the topology of the lattice. For open cubic lattices the coefficients a_{ij} $(i \leqslant 9, j \leqslant 8)$ have been calculated, while the series for the more general Hamiltonian is determined up to and including β^9, thereby not reaching the limit imposed by these lattices.

1. Introduction

High-temperature series expansions of thermodynamical functions have been of great help in theoretical as well as in experimental magnetism. A considerable amount of labour is still devoted to the calculation of further terms in known series or the evaluation of completely new series. It seems that most attention has been paid to Hamiltonians with a model interaction (Ising, XY and Heisenberg; spin dimensionality $D = 1, 2$ and 3 respectively), mainly on lattices with nearest-neighbour interactions only. In practice however, many magnetic compounds do not behave like such a model system and experimental physicicists are confronted with a lack of data concerning the series for more general Hamiltonians. This may imply interactions of different strength or a more general form of the exchange tensor.

We wish to turn our attention to this last case and write the Hamiltonian in the axial form

$$\mathscr{H} = -2 \sum_{\langle ij \rangle} [J_\perp(S_i^x S_j^x + S_i^y S_j^y) + J_\parallel S_i^z S_j^z] + H \sum_i S_i^z \tag{1}$$

with $S = \frac{1}{2}$. The summation runs over all pairs of nearest neighbours and each pair is counted once.

In order to calculate the series expansion for the logarithm of the partition function

$$\ln(Z(\beta, H)) = \sum_{ij} \beta^i H^j \sum_{k=0}^{i} a_k^{ij} J_\perp^k J_\parallel^{i-k} \tag{2}$$

with $\beta = 1/kT$, we employed the finite cluster method (Domb 1960, Rushbrooke 1964, Rushbrooke *et al* 1974), for a set of ratios $J_\perp/J_\parallel$.

The experimental interest of our laboratory in the magnetic behaviour of certain insulators motivated us to see whether this same technique could be applied to the more general Hamiltonian

$$\mathscr{H}' = -2\sum_{\langle ij\rangle}(J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_z S_i^z S_j^z) + H\sum_i S_i^z \tag{3}$$

for which the series would read

$$\ln(Z'(\beta, H)) = \sum_{ij} \beta^i H^j \sum_{k=0}^{i} \sum_{l=0}^{i-k} \gamma_{kl}^{ij} J_x^k J_y^l J_z^{i-k-l}. \tag{4}$$

It is obvious that the extra labour involved is considerable, partly due to the higher number of γ_{kl}^{ij} but mainly due to the fact that T_z $(= \Sigma_i S_i^z)$ is no longer a good quantum number in (3). This reduces the symmetry of $\mathscr{H}'$ (as compared to (1)) and results in much larger matrices that have to be handled.

It was found however, that the coefficients in the series for the zero-field specific heat (or equivalently the γ_{kl}^{i0}) for a Hamiltonian like (3) are related to the α_k^{i0} from the axial case (1).

A unique correspondence between them holds up to a certain order in β, which depends on the type of lattice.

2. Method of calculation

The most powerful method of calculating the coefficients in a series expansion of $\ln(Z)$ for an $S = \frac{1}{2}$ Hamiltonian like (1) is probably the finite cluster method, introduced by Domb (1960). In order to find the series for an infinite lattice with this technique, the series for a number of relatively small clusters of spins are calculated and combined in a suitable way. Details of the different steps in this process are clearly described elsewhere (see, for instance, Baker *et al* 1967b, Rushbrooke *et al* 1974) and tables that describe the way in which the clusters should be combined have been published by Baker *et al* (1967a). Here we wish to emphasize only one point, concerning the arithmetic.

The coefficients in the series for any particular cluster are obtained from $\mathrm{Tr}(\mathscr{H}^i T_z^j)$ for the corresponding Hamiltonian. When dealing with an isotropic interaction tensor $(J_\perp = J_\parallel)$, this is usually done through repeated multiplication of the matrix representation of $\mathscr{H}$ on any suitable set of basis functions. For $S = \frac{1}{2}$ the basis can be chosen such that this matrix contains only integer numbers and computer calculations are then errorless. In our case of anisotropic exchange, $\mathrm{Tr}(\mathscr{H}^i T_z^j)$ should be solved for a number of ratios $J_\perp/J_\parallel$. Although it is still possible to choose $J_\perp/J_\parallel$ as an integer, thereby obtaining a matrix that contains only integers also, we found this method too cumbersome and proceeded differently.

Instead of matrix multiplications to obtain $\mathrm{Tr}(\mathscr{H}^i T_z^j)$, we calculated the eigenvalues of $\mathscr{H}$ and computed the traces from these. Since T_z is a good quantum number, the eigenvalues can be labelled according to the eigenvalues of T_z and no difficulties arise. This set up introduces some rounding errors but on the other hand the computations are less time consuming.

Solving ln (Z) for given $J_\perp$ and $J_\parallel$ results in a series

$$\ln (Z(\beta, H)) = \sum_{i \geqslant 0} \sum_{j \geqslant i} a_{ij}(J_\perp, J_\parallel)\beta^i H^j. \tag{5}$$

It should be noted that in this expression the coefficients a_{ij} are not known as *functions* of $J_\perp$ and $J_\parallel$. Their numerical values are known only for certain combinations of the two parameters. Since each coefficient $a_{ij}(J_\perp, J_\parallel)$ in (5) can be expressed in a homogeneous polynomial of degree i in $J_\perp$ and $J_\parallel$,

$$a_{ij}(J_\perp, J_\parallel) = \sum_{k=0}^{i} \alpha_k^{ij} J_\perp^k J_\parallel^{i-k} \tag{6}$$

the final coefficients α_k^{ij} can be solved by comparing the numerical values of $a_{ij}(J_\perp, J_\parallel)$ in (5) for different sets of $J_\perp$ and $J_\parallel$. As mentioned, the $a_{ij}(J_\perp, J_\parallel)$ are subject to rounding errors and an averaging is therefore desirable. We solved (5) for 14 different ratios $J_\perp/J_\parallel$ and this averaging was accomplished by requiring the squares sum

$$F_{ij} = \sum_{J_\perp, J_\parallel} \left(\sum_{k=0}^{i} a_k^{ij} J_\perp^k J_\parallel^{i-k} - a_{ij}(J_\perp, J_\parallel) \right)^2 \tag{7}$$

to reach a minimum for the α_k^{ij} to be determined. The α_k^{ij}, found in this way, are not errorless but the minimum value of F_{ij} can be used to indicate their reliability. Some of the α_k^{ij} are known exactly from the series for model Hamiltonians (Ising, XY). These may be substituted beforehand. The known series coefficients for the Heisenberg model impose an additional condition of the coefficients α, namely that their sum is correct. After solution of (7) the difference between their sum and the correct sum is distributed among the α_k^{ij} with a weight according to their respective uncertainties. In this way the series for the model Hamiltonians (Ising, XY and Heisenberg) are correctly represented by the general expression (2).

We will now turn to the possibility of calculating the series for the general Hamiltonian (3). In order to find the expressions for the coefficients $b_{ij}(J_x, J_y, J_z)$ in (4), we first define their polynomial expression in accordance with (6) as

$$b_{ij}(J_x, J_y, J_z) = \sum_{k=0}^{i} \sum_{l=0}^{i-k} \gamma_{kl}^{ij} J_x^k J_y^l J_z^{i-k-l}. \tag{8}$$

Our aim is to establish a relation between γ_{kl}^{ij} and α_m^{ij}. Since $b_{ij}(J_\perp, J_\perp, J_\parallel) = a_{ij}(J_\perp, J_\parallel)$ this is readily done by the substitution $J_x = J_y = J_\perp$, $J_z = J_\parallel$ in (8) and comparing the result with (6). This shows that for any i and j

$$\alpha_k^{ij} = \sum_{l=0}^{k} \gamma_{lk-l}^{ij} \qquad (k = 0, 1, \ldots i). \tag{9}$$

For i and j fixed, the number of unknowns on the right-hand side is $\frac{1}{2}(i+1)(i+2)$, whereas the number of coefficients on the left-hand side is just $i+1$. In general it is thus not possible to determine the values for all γ_{kl}^{ij} for given i and j from the i different α_m^{ij}. However, certain symmetries may be present and these are most helpful when $j = 0$ and $H = 0$. In that case no external preferred direction is imposed on the Hamiltonian and $\mathscr{H}'$ is invariant under any permutation of J_x, J_y and J_z. For the coefficients γ^{i0}_{kl} this implies the relations

$$\gamma^{i0}_{kl} = \gamma^{i0}_{lk} = \gamma^{i0}_{i-kl} = \gamma^{i0}_{li-k} = \gamma^{i0}_{ki-l} = \gamma^{i0}_{i-lk}. \tag{10}$$

It is obvious that this symmetry reduces the number of coefficients γ^{i0}_{kl} quite drastically. To show the effect on the calculation we will consider the coefficients in β^3 in more detail. For the general Hamiltonian (3) we find, grouping different terms in accordance with (10),

$$\begin{aligned} b_{30}(J_x, J_y, J_z) = {} & \gamma^{30}_{00}(J_x^3 + J_y^3 + J_z^3) + \gamma^{30}_{01}(J_xJ_y^2 + J_yJ_z^2 + J_zJ_x^2 + J_x^2J_y + J_y^2J_z + J_z^2J_x) \\ & + \gamma^{30}_{11}J_xJ_yJ_z \end{aligned} \tag{11}$$

and for the axial case

$$a_{30}(J_\perp, J_\parallel) = \alpha^{30}_0 J_\parallel^3 + \alpha^{30}_1 J_\parallel^2 J_\perp + \alpha^{30}_2 J_\parallel J_\perp^2 + \alpha^{30}_3 J_\perp^3. \tag{12}$$

Substitution of $J_x = J_y = J_\perp$ and $J_z = J_\parallel$ in (11) and comparison of (11) and (12) then leads to the set of equations (9), which read

$$\begin{aligned} \alpha^{30}_0 &= \gamma^{30}_{00} \\ \alpha^{30}_1 &= 2\gamma^{30}_{01} \\ \alpha^{30}_2 &= 2\gamma^{30}_{01} + \gamma^{30}_{11} \\ \alpha^{30}_3 &= 2\gamma^{30}_{00} + 2\gamma^{30}_{01}. \end{aligned} \tag{13}$$

The three different γ^{30}_{kl} are thus uniquely determined as

$$\begin{aligned} \gamma^{30}_{00} &= \alpha^{30}_0 \\ \gamma^{30}_{01} &= \tfrac{1}{2}\alpha^{30}_1 \\ \gamma^{30}_{11} &= \alpha^{30}_2 - \alpha^{30}_1 \end{aligned} \tag{14}$$

and besides, it is clear that a relation must exist between the four α^{30}_m. For higher terms the corresponding set of equations becomes still undetermined and no unique solution can be found. In fact, for $i = 4$ one would end up with five equations and six unknowns. However, a number of the coefficients γ^{i0}_{kl} can be neglected since their value must be zero. In the above example for instance, γ^{30}_{01} must vanish since in β^3 the only graphs with a non-vanshing trace are

$$\equiv \qquad \text{and} \qquad \triangle.$$

But the first gives rise to γ^{30}_{11} since any other combination would result in a vanishing trace, and the second graph contributes only to γ^{30}_{00}. (This explains the predicted extra relation between the four α^{30}_m.)

A systematic examination of all graphs yields some general rules concerning the indices i, k and l in γ^{i0}_{kl}. We may state quite generally that no graphs exist that contribute to γ^{i0}_{0l} with l odd. Other restrictions may be present that depend on the type of lattice for which the series is calculated. For all open cubic lattices (no odd-numbered rings) for instance, examination of the graphs shows that i, j and k should be all even or all odd. Conditions, imposed in this way on the coefficients, affect both the number of equations and the number of unknowns in (9). We did not study this problem in great detail for all lattices, but a first examination reveals that the set is still solvable to order β^7 for any lattice. We conclude therefore that this method is rather generally applicable.

although its use is primarily of experimental interest. For theoretical analysis of series, order β^7 is quite low. On the open lattices, where solutions can be found to order β^{11}, the restriction is less severe.

Attempts to apply the same method to the series of the susceptibility (γ_{kl}^{i2}) fail already at order β^4, due to the fact that $\mathscr{H}'$ is no longer invariant under all permutations of J_x, J_y and J_z.

3. Results

The series expansion of ln (Z) was calculated on four open lattices: the linear chain (one dimensional), square (two dimensional), simple cubic (three dimensional) and body-centred cubic (three dimensional). The polynomials representing the coefficients $b_i(J_x, J_y, J_z)$ and $a_{ij}(J_\perp, J_\parallel)(j > 0)$ are expressed as integer ratios in table 1. The table is divided in sections, corresponding to the power of H in (2). Each section contains seven columns, corresponding respectively to: the order of β, an identification of the non-zero coefficients α_m^{ij} or γ_{kl}^{i0}, a multiplication factor and the information for the four lattice types. For the terms in H^0 the identification of the γ_{kl}^{i0} is a set of three numbers listing the respective powers of J_x, J_y and J_z. It should be remembered that, as in (11), permutation of the J_x, J_y and J_z is implicit. Thus $\{1, 1, 3\}$ for example corresponds to

$$(J_xJ_yJ_z^3 + J_xJ_y^3J_z + J_x^3J_yJ_z)$$

and $\{2, 2, 2\}$ to $J_x^2J_y^2J_z^2$. The other sections of the table compile information for the axially symmetric Hamiltonian (1). In that case no permutations are allowed and (2, 4) for example should thus be read as $(J_\perp^2 J_\parallel^4)$. Each of the numbers in the last four columns must be multiplied by the appropriate constant in the third column. As was mentioned, the coefficients are not free from rounding errors. The minimum squares sum resulting from the least-squares fit (7) in the determination of the α_k^{ij}, is used to estimate the accuracy of the numbers. This is expressed by asterisks, superscripted to the numbers in the last four columns of the table. If one asterisk is attached, a deviation of ± 5 is possible. For two asterisks a maximum error of ± 50 may be present. Care was taken to ensure the correct result for Heisenberg interaction ($J_x = J_y = J_z$) (Rushbrooke *et al* 1974), for Ising exchange ($J_x = J_y = 0$) (Domb 1974) and for the XY model ($J_x = J_y, J_z = 0$) (Betts 1974) for terms in H^0; Lee (1971) for terms in H^2. Previous results on the axial Hamiltonian were used as a check also. Especially the result of Obokata *et al* (1967) for the susceptibility of (1) on the linear chain, square and simple cubic lattices, and the work of Wood and Dalton (1972). These checks were made by comparing the series for the specific heat in zero field:

$$C = \sum_{i \geqslant 2} i(i-1)a_{i0}\beta^i \tag{15}$$

and for the susceptibility in zero field,

$$\chi = \sum_{i \geqslant 2} 2a_{i2}\beta^{i-1}. \tag{16}$$

For the Heisenberg Hamiltonian, the tables of Rushbrooke *et al* (1974) offered the possibility of a direct comparison for the expansion of ln (Z).

Table 1. Coefficients in the series expansion of $\ln(Z(\beta, H))$ for four lattices. The series is written as

$$\ln Z = \sum a_{ij}\beta^i H^j$$

and the a_{ij} are tabulated. For $j = 0$, they are expressed as a function of J_x, J_y and J_z, according to the Hamiltonian (3) of the text. For $j \neq 0$, their dependence on $J_\perp$ and $J_\parallel$ is given for the axial Hamiltonian (1). The correct use of the table is explained in the text.

Terms in $\ln(Z'(\beta, 0))$

	$\{J_x^k J_y^l J_z^m\}$	Factor	Chain	Square	Cubic	BCC
β^0	{0, 0, 0}	ln(2)	1	1	1	1
β^2	{0, 0, 2}	$1/2^2 2!$	1	2	3	4
β^3	{1, 1, 1}	$6/2^3 3!$	−1	−2	−3	−4
β^4	{0, 0, 4}	$2/2^4 4!$	−1	10	33	140
	{0, 2, 2}		−4	−24	−60	−112
β^5	{1, 1, 3}	$40/2^5 5!$	3	10	21	12
β^6	{0, 0, 6}	$8/2^6 6!$	2	64	1626	11888
	{0, 2, 4}		18	−108	−810	−7560
	{2, 2, 2}		−51	846	4131	12756
β^7	{1, 1, 5}	$16/2^7 7!$	−238	224	−4662	−19600
	{1, 3, 3}		−679	−5418	−16569	13412
β^8	{0, 0, 8}	$16/2^8 8!$	−17	4250	370641	5580220
	{0, 2, 6}		−272	2306*	−313096**	−3256052**
	{0, 4, 4}		−612	−1212*	−16012**	−1021032**
	{2, 2, 4}		5424	11536	−216246	2536608
β^9	{1, 1, 7}	$64/2^9 9!$	2790	−30056	−453634*	−13922881**
	{1, 3, 5}		15480	95539*	1467052*	7873215**
	{3, 3, 3}		27840	864970*	3870492*	−13809423**

Terms in H^2 in $\ln(Z(\beta, H))$

	$(J_\perp^k J_\parallel^l)$	Factor	Chain	Square	Cubic	BCC
β^2	(0, 0)	$1/2^2 2!$	1	1	1	1
β^3	(0, 1)	$6/2^3 3!$	1	2	3	4
β^4	(0, 2)	$24/2^4 4!$	1	6	15	28
	(2, 0)		−1	−2	−3	−4
β^5	(0, 3)	$80/2^5 5!$	1	26	111	292
	(2, 1)		−3	−18	−45	−84
β^6	(0, 4)	$240/2^6 6!$	1	138	1059	3916
	(2, 2)		−2	−132	−606	−1640
	(4, 0)		6	20	42	24
β^7	(0, 5)	$672/2^7 7!$	1	902	12603	65524
	(2, 3)		5	−1110	−9345	−36100
	(4, 1)		15	350	1425	2100
β^8	(0, 6)	$896/2^8 8!$	2	13752	354450	2593136
	(2, 4)		−6	−20388	−323010	−1762294*
	(4, 2)		−225	8286	72405	197532*
	(6, 0)		−170	−916	−4806	−8792*
β^9	(0, 7)	$4608/2^9 9!$	1	60566	2904051	29907112
	(2, 5)		−63	−105630	−3139017	−24166230*
	(4, 3)		−273	54838	946617	4049008*
	(6, 1)		175	−11396	−109725	−247688*

Table 1.—continued.

	Terms in H^4 in $\ln(Z(\beta, H))$						
	$(J_\perp^k J_\parallel^l)$	Factor	Chain	Square	Cubic	BCC
β^4	(0, 0)	$2/2^4 4!$	−1	−1	−1	−1
β^5	(0, 1)	$80/2^5 5!$	−1	−2	−3	−4
β^6	(0, 2)	$120/2^6 6!$	−13	−66	−159	−292
	(2, 0)		4	8	12	16
β^7	(0, 3)	$560/2^7 7!$	−40	−608	−2424	−6208
	(2, 1)		39	198	477	876
β^8	(0, 4)	$2240/2^8 8!$	−121	−6174	−41919	−150340
	(2, 2)		212	3480	14124	36464
	(4, 0)		−51	−194	−429	−492
β^9	(0, 5)	$16128/2^9 9!$	−182	−34564	−407706	−2051288
	(2, 3)		440	28260	198420	723800
	(4, 1)		−345	−4330	−16395	−30300

	Terms in H^6 in $\ln(Z(\beta, H))$					
	$(J_\perp^k J_\parallel^l)$	Factor	Chain	Square	Cubic	BCC
β^6	(0, 0)	$16/2^6 6!$	1	1	1	1
β^7	(0, 1)	$1904/2^7 7!$	1	2	3	4
β^8	(0, 2)	$896/2^8 8!$	107	522	1245	2276
	(2, 0)		−17	−34	−51	−68
β^9	(0, 3)	$5376/2^9 9!$	602	8092	31542	80024
	(2, 1)		−321	−1566	−3735	−6828
	Terms in H^8 in $\ln(Z(\beta, H))$					
β^8	(0, 0)	$272/2^8 8!$	−1	−1	−1	−1
β^9	(0, 1)	$71424/2^9 9!$	−1	−2	−3	−4

Acknowledgments

The authors kindly acknowledge stimulating discussions with Professor P van der Leeden, Professor C Domb and Dr W J M de Jonge.

Note added in proof. Very recently van Dongen *et al* (1975) obtained results for the series expansion (4), for the linear chain. Their technique is very similar to the method described here. Actually they were able to find the γ_{kl}^{i0} from α_0^{i0} and α_i^{i0}, known from the exact solutions for the Ising and XY chain respectively. Our results are in agreement with theirs.

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