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Linked cluster expansion for quantum spins

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Abstract. A vertex renormalised self consistent graph model for a quantum spin system is presented with the ordering transition temperature appreciably lowered from the classical value for a small spin quantum number I. The model applies e.g. to the dipole–dipole interaction in addition to the exchange interactions. The derivation is restricted to zero external field.

1. Introduction

The properties of simple models such as the Gaussian model or the spherical model have long provided guidelines to the thermodynamics of magnetic phase transitions to theoreticians and experimentalists alike. A great deal of effort has gone into the systematic first principles derivation of such model results and hopefully improved versions of the same mostly with the aid of (diagrammatic) cluster expansions (Wortis 1974).

Recently the problem of evaluating magnetic transition temperatures starting with more or less precisely known interactions has become interesting in the context of nuclear spin systems (Ehnholm *et al* 1979, Kubota *et al* 1980). The extension of high-temperature series expansions is tedious if the spin–spin interaction is not restricted to the nearest-neighbour exchange. We have therefore been led to be interested in models of the type described above for *quantum* spin systems. It is not trivial to include the quantum nature of the spins in the popular models mentioned above. In the Gaussian model, spin commutators make the renormalisation of the bonds or vertices of the relevant diagrams difficult as a way of improving on it. The spherical model is essentially a vertex renormalised version of the Gaussian model for Ising (commuting) spins. Stinchcombe *et al* (1963) have taken the diagrams of the spherical model to obtain a formal vertex renormalisation for quantum spins. Unfortunately the result reduces to the spherical model in the absence of an external magnetic field.

In this paper we derive a vertex renormalisation for a quantum spin system with nontrivial consequences for the transition temperature as a function of $I(I+1)$. Our renormalisation is equivalent to that of Horwitz and Callen (Horwitz and Callen 1961) in the classical limit. We make one further approximation in the quantum case

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concerning the permutability of spin operators in a given graph which involves only a part of the graphs and which probably does not affect our prediction of T_c. The critical properties of our model are essentially the same as those of the spherical model and independent of the spin quantum number in agreement with the expectation that the spin quantum number is an 'irrelevant' variable (Kadanoff 1976).

We find that a quantum spin should lead to a lowered transition temperature in general. Compared as it stands with the classical and spin-$\frac{1}{2}$ nearest-neighbour Heisenberg model results from high-temperature series expansions (Rushbrooke *et al* 1974), the present calculation delivers too low a T_c for both. This is not too alarming as the quantum spins certainly make impossible even an approximative bond renormalisation. The vertex renormalisation is insufficient for short-range interactions. However, in view of the simple structure of our results, one can make the statement that it should be possible to simulate the bond renormalisation with some *ad hoc* procedure to shift upward the transition temperatures and thus describe correctly the classical *and* the spin-$\frac{1}{2}$ Heisenberg models. In this sense we seem to describe the quantum effects correctly. The accuracy of our model is also improved for long-range interactions.

Our derivation is limited to zero external field. The free energy is thermodynamically consistent in the sense discussed by Englert (1963) as the fluctuation of the (staggered) magnetisation is equal to the susceptibility by construction. This is not true in general for pure vertex renormalisations of the Horwitz and Callen (1961) type. In the case of the classical Heisenberg model our result is independent of the spin dimensionality, d. We thus do not get the expected spherical model behaviour in the limit of infinite d. The discrepancy would nevertheless be small in any conceivable practical case. At the high-density limit we correctly recover the spherical model.

2. Single diagram contribution

In this section we derive the contribution of one Husimi tree diagram in the expansion of the free energy. No external field is included so the calculation applies only above the critical temperature. We obtain an approximate general expression for a quantum spin system which reduces to the classical zero-field result at the appropriate limit, $I \to \infty$.

The Hamiltonian is given by

$$H = -\tfrac{1}{2}\sum_{i,j} J_{ij}^{\alpha\beta} I_i^{\alpha} I_j^{\beta} \tag{2.1}$$

with sum implied over repeated Greek spin indices. The coupling $J_{ij}^{\alpha\beta}$ must have a particular symmetry, essentially cubic, to be specified later (equation (2.9)). The spin operators obey the usual commutation relations

$$[I_i^{\alpha}, I_j^{\beta}] = \mathrm{i}\varepsilon^{\alpha\beta\gamma} I_i^{\gamma} \delta_{ij}. \tag{2.2}$$

We calculate the free energy from the moment expansion (Rushbrooke *et al* 1974).

$$-\beta F = N \log(2I+1) + \sum_{n=1}^{\infty} \frac{1}{n!} \langle (-\beta H)^n \rangle \tag{2.3}$$

with the expectation value

$$\langle 0 \rangle = \mathrm{Tr}_N(0)/(2I+1)^N \tag{2.4}$$

where the trace is calculated over the $(2I+1)^N$-dimensional phase space without a density operator. It is customary to do the bookkeeping of the different combinations of Hamiltonians in (2.3) with diagrams in which each line (bond) corresponds to one coupling $J_{ij}^{\alpha\beta}$ and each point (vertex) to the trace of spin operators at a lattice site. In the lattice sum no two lattice indices may coincide as this would lead to a different diagram. For example, diagram (j) in figure 1 would duplicate diagrams (a) and (b). Irrespective of the indices in the lattice sums, the total effect of spin traces in a given combination of Hamiltonians can be expressed as a single factor. The components of a given disconnected diagram are not independent as a result of the restrictions on lattice sums. The diagram (j) in figure 1, say, is *not* completely cancelled in the free energy expansion (2.3). The restrictions on the lattice sums of disconnected diagrams thus lead to connected contributions. These would be taken into account in the definition of the cumulant (Rushbrooke *et al* 1974) but the moment expansion turns out to be more suitable for our specific purposes.

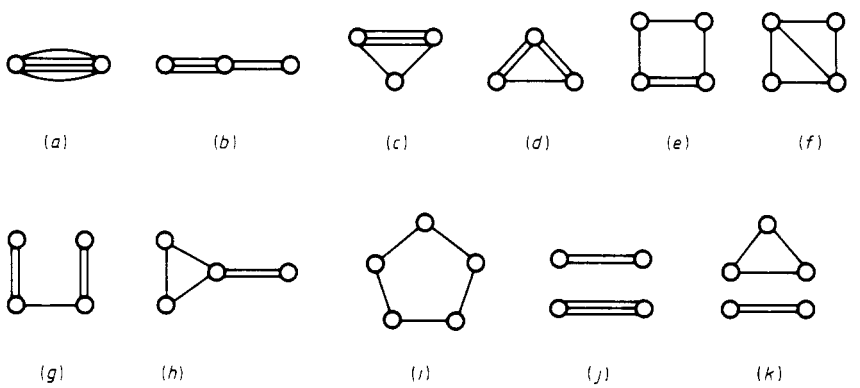


Figure 1. All fifth-order diagrams. Diagrams having vertices with only one impinging bond are not included as the trace of a single spin operator is zero. Spin traces additionally eliminate diagrams (f) and (g).

We select the 'Husimi trees' (see Domb 1974) to work with. They consist of loops of bonds in such a way that each bond belongs to only one loop. In figure 1 all fifth-order diagrams are displayed, out of which (h), (i) and (k) are included in our analysis†. Disconnected diagrams contribute only via the corrections on account of the restrictions on the lattice sums (of equation (2.15)). We will return to our particular choice of diagrams in § 3. Roughly speaking one can say that the Husimi trees dominate the free energy expansions above the critical temperature in zero magnetic field.

Let us now calculate the contribution of the diagram in figure 2. There are $n!/g$ different ways of assigning the n Hamiltonians $(-\beta H)$ to the bonds of the diagram where g is the symmetry number (Domb 1974) dividing the contribution with the number of configurations which do not change the topology of the diagram. We calculate a trace over the spin operators at each vertex and sum over the lattice sites for each permutation. These $n!/g$ terms are then added to give the total contribution of a diagram (for details, see Rushbrooke *et al* 1974). The restrictions on the lattice

† The double bond is also taken to be a loop.

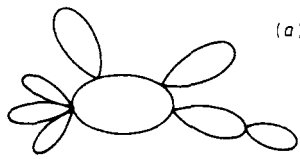


Figure 2. A general Husimi tree. Individual bonds are not shown: each loop may contain an arbitrary number of bonds.

sums are ignored for the time being. With the fourier transform

$$J^{\alpha\beta}(\boldsymbol{k}) = \frac{1}{N}\sum_{i,j} \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}) J_{ij}^{\alpha\beta} \tag{2.5}$$

and its inverse

$$J_{ij}^{\alpha\beta} = \frac{1}{V_k}\int \mathrm{d}^3k J^{\alpha\beta}(\boldsymbol{k}) \exp(-i\boldsymbol{k}\cdot\boldsymbol{r}_{ij}) \tag{2.6}$$

the loop 'a' of m vertices in the diagram of figure 2 gives the effective coupling

$$H_{\mathrm{eff},m}^{\alpha\beta} = \left(\frac{\beta}{2}\right)^m \left(\frac{\sigma_2}{3}\right)^{m-1} \frac{1}{V_k}\int \mathrm{d}^3k J^{\alpha\gamma}(\boldsymbol{k}) J^{\gamma\delta}(\boldsymbol{k}) \ldots J^{\omega\beta}(\boldsymbol{k}) \tag{2.7}$$

for each permutation of the Hamiltonians. The spin trace

$$\mathrm{Tr}(I^\alpha I^\beta) = \tfrac{1}{3}I(I+1)\delta^{\alpha\beta} = \sigma_2\delta^{\alpha\beta}/3 \tag{2.8}$$

is included in equation (2.7). In addition to the effective coupling, the loop 'a' has the spins I^α and I^β operating on the articulation vertex connecting the loop with the rest of the diagram. They are not explicitly shown in equation (2.7) as their relative position with respect to the other spins at that vertex depends on the particular permutation of the Hamiltonians.

The coupling $J_{ij}^{\alpha\beta}$ is now required to be such that $H_{\mathrm{eff},m}^{\alpha\beta}$ is independent of the spin indices α and β

$$H_{\mathrm{eff},m}^{\alpha\beta} = \delta^{\alpha\beta}(\sigma_2/3)^{m-1}\boldsymbol{J}_m. \tag{2.9}$$

This holds for isotropic exchange interactions and also, on a cubic lattice, for the dipolar interaction†. The trace of spin operators is non-zero only if the components $\{I^x, I^y, I^z\}$ occur in it either all odd or all even numbers of times (Ambler *et al* 1962). The articulation vertex of the loop 'a' then carries a delta function in spin space in addition to the trace depending on the particular permutation. We can repeat the above argument for the other loops as well to arrive at the conclusion that each assignment of the Hamiltonians to the bonds in a diagram (of k loops, $\{l_i\}_{i=1}^k$ bonds and r vertices) contributes to the free energy

$$N\frac{1}{2^n}\left(\prod_{i=1}^{k}\boldsymbol{J}_{l_i}\right)\frac{1}{3^n}\left[\prod_{i=1}^{r}(\mathrm{Tr}(I)_i/(2I+1))\right]. \tag{2.10}$$

$\mathrm{Tr}(I)_i$ represents the trace at the vertex i for the particular permutation.

† In fact one should require the macroscopic sample to be a sphere for (2.9) to hold for the dipole–dipole interaction whose $\boldsymbol{k}=0$ component depends on the shape of the sample. In the integral (2.7) this may be ignored, however (cf Griffiths 1968).

Neglecting the restrictions on the lattice sums, the total contribution of one Husimi tree is

$$\langle(-\beta H)^n\rangle = N \sum_{\{p\}} \left\{ \frac{1}{2^n} \prod_{i=1}^{k} (\boldsymbol{J}_{l_i}) \frac{1}{3^n} \prod_{i=1}^{r} [\mathrm{Tr}(I)_i/(2I+1)] \right\} \tag{2.11}$$

where the sum is over all the $n!/g$ permutations of the n Hamiltonians. These permutations only affect the spin traces. Thus the permutations can be approximately replaced by permuting just the spin operators. Permutations of Hamiltonians obviously affect the spin traces at the articulation vertices but the two spin operators from each loop carry the same spin index. Thus their relative order is immaterial. In a large portion of the Husimi trees this allows enough freedom to make the modification exact. In the rest of the diagrams, the modification is approximate and we will return to it in § 3 and in appendix 2. The approximate form of (2.11) is

$$\langle(-\beta H)^n\rangle = \frac{N}{g} \left(\prod_{i=1}^{k} \boldsymbol{J}_{l_i} \right) \frac{1}{3^n} \frac{n!}{(2n)!} \sum_{\{p'\}} \left[\prod_{i=1}^{r} \mathrm{Tr}(I)_i/(2I+1) \right] \tag{2.12}$$

where the sum is over the $(2n)!$ permutations of the individual spin operators. It follows that

$$\langle(-\beta H)^n\rangle = \frac{N}{g} \frac{n!}{3^n} \left(\prod_{i=1}^{k} \boldsymbol{J}_{l_i} \right) \left(\prod_{i=1}^{r} \sigma_{2v_i} \right) \tag{2.13}$$

where

$$\sigma_{2v_i} = \frac{1}{2I+1} \frac{1}{(2v_i)!} \sum_{\{P_{2v_i}\}} \mathrm{Tr}(I^{\alpha} I^{\alpha} \ldots I^{\omega} I^{\omega}). \tag{2.14}$$

In equation (2.13) $2v_i$ is the number of bonds impinging on the vertex i, and in equation (2.14) the sum is over the permutations of the corresponding spin operators. A recursion relation of the spin trace Σ is derived in appendix 1.

We now return the restrictions on the lattice sums. To account for them we have to subtract all the restricted sums with two, three, etc. lattice indices set equal (Percus 1971). With the restrictions indicated by a prime this reads

$$\begin{aligned} \Sigma'_{i,j,k,l,\ldots} A = {} & \Sigma_{i,j,k,l,\ldots} A - \Sigma'_{i,j,k,l,\ldots} A(\delta_{ij} + \delta_{ik} + \delta_{jk} + \ldots) \\ & - \Sigma'_{i,j,k,l,\ldots} A(\delta_{ij}\delta_{jk} + \delta_{jk}\delta_{kl} + \ldots) \\ & - \Sigma'_{i,j,k,l,\ldots} A(\delta_{ij}\delta_{jk}\delta_{kl} + \ldots) - \ldots . \end{aligned} \tag{2.15}$$

The spin traces must be calculated before this step. In order to be consistent, we keep track of those corrections only whose lattice sums can be expressed in terms of the effective coupling $\boldsymbol{J}_1$. According to an entirely topological argument of Horwitz and Callen (1961), this is equivalent to 'opening'† the vertices of a given tree in all possible ways to produce either connected or disconnected Husimi trees and subtracting these terms from the spin trace Σ. In disconnected diagrams only those terms are kept on the right-hand side of equation (2.15) which the delta functions force connected. The restricted lattice sums can thus be made unrestricted by replacing the spin traces with new effective traces $\boldsymbol{\Sigma}$.

† The meaning of 'opening' becomes obvious by considering the procedure (2.15) with the spin traces.

Consider opening the spin trace σ_{2n} into σ_{2v} and $\sigma_{2(n-v)}$. One has to choose at least two distinct indices out of the $2n$ to get a connected contribution as each loop corresponds to two identical spin indices. As

$$\begin{aligned}&\mathrm{Tr}(I^{\omega}I^{\alpha}\ldots I^{\beta}I^{\delta})\,\mathrm{Tr}(I^{\omega}I^{\gamma}\ldots I^{\phi}I^{\delta})\\&\quad=\tfrac{1}{3}\,\mathrm{Tr}(I^{\omega}I^{\alpha}\ldots I^{\beta}I^{\omega})\,\mathrm{Tr}(I^{\delta}I^{\gamma}\ldots I^{\phi}I^{\delta})\end{aligned}\tag{2.16}$$

the connected contribution is

$$-\tfrac{1}{3}[(2n/2)-n]\binom{n-2}{v-1}\boldsymbol{\Sigma}_{2v}\boldsymbol{\Sigma}_{2(n-v)}.\tag{2.17}$$

The effective spin traces arise because of the recursive nature of (2.15). The disconnected contribution is readily found to be

$$-\binom{n}{v}\boldsymbol{\Sigma}_{2v}\boldsymbol{\Sigma}_{2(n-v)}.\tag{2.18}$$

A straightforward generalisation leads to

$$\boldsymbol{\Sigma}_{2n}=\Sigma_{2n}-\sum_{p=2}^{n}\frac{n!}{p!}\sum_{\{v_i\}}\left[\!\!\left[\prod_{i=1}^{p}\left\{\frac{\sigma_{2v_i}}{v_i!}\left[1+\frac{2}{3}v_i\left(n-\sum_{l=1}^{i}v_l\right)\right]\right\}\right]\!\!\right].\tag{2.19}$$

Here we sum over all the combinations $\{v_i\}$ such that $\Sigma_{i=1}^{p}v_i=n$.

In conclusion, the approximate contribution of a single Husimi tree to the free energy expansion is

$$\langle(-\beta H)^n\rangle=\frac{N}{g}\frac{n!}{3^n}\left(\prod_{i=1}^{k}\boldsymbol{J}_{l_i}\right)\left(\prod_{i=1}^{r}\boldsymbol{\Sigma}_{2v_i}\right)\tag{2.20}$$

where

$$\boldsymbol{J}_l=\frac{\beta^l}{V_k}\int \mathrm{d}^3k J^{\alpha\beta}(\boldsymbol{k})J^{\beta\gamma}(\boldsymbol{k})\ldots J^{\omega\alpha}(\boldsymbol{k}).\tag{2.21}$$

In the diagram there are k loops of l_i bonds ($\Sigma_{i=1}^{k}l_i=n$) and r vertices with $2v_i$ impinging loops ($\Sigma_{i=1}^{r}v_i=n$). In equation (2.20) a Husimi tree refers to a diagram in fourier space, not on the real lattice. Defining the eigenvalues of the matrices $J^{\alpha\beta}(\boldsymbol{k})$ as $\lambda_\gamma(\boldsymbol{k})$ we arrive at

$$\boldsymbol{J}_l=\frac{\beta^l}{V_k}\sum_{\gamma=1}^{3}\int \mathrm{d}^3k(\lambda_\gamma(\boldsymbol{k}))^l.\tag{2.22}$$

The simplicity of the expression (2.20) with the effective spin traces $\boldsymbol{\Sigma}$ is the reason for which we chose to use the moment expansion as opposed to the cumulant expansion.

In the customary linked cluster expansion of the quantum Heisenberg model, the spin traces $\boldsymbol{\Sigma}$ are replaced with time-ordered semi-invariants. This limits practicable calculations to much more restricted sets of diagrams than ours. Typically then the quantum and classical systems differ above the transition temperature only in the case of non-zero external field. In our expansion the spin traces $\boldsymbol{\Sigma}$ bring about an essential difference between the two. The extension of the free energy expansion to high orders is straightforward.

3. The sum of the Husimi tree diagrams

An analytic expression for the free energy can be derived by extending the sum of the Husimi trees to infinite order. The procedure leads to two coupled equations via which the spin quantum number and the spectrum of energy eigenvalues determine the thermodynamics of the system. In this section we also consider the non-Husimi diagrams left out of our analysis. This section is closely parallel to the Ising model discussion of Horwitz and Callen (1961) henceforth referred to as HC. We thus concentrate on the main lines and refer to the HC paper for details.

The contribution of a single diagram, equation (2.20), is extremely useful in that one can generate all the Husimi trees by just successively adding new loops. The blind application of such a method results in overcounting of diagrams. One cannot compensate for this by means of the symmetry numbers g. The HC procedure (which is not restricted to the Ising model) generates diagrams in two separate sequences starting from two different sets of basic elements (either 'hyperbonds' or 'hypervertices'.) The difference of these two expansions gives the correct free energy. It is then assumed that one may exchange the order of the sum over the number of bonds in a given loop and the integral in equation (2.22). After a little algebra the free energy emerges as

$$-\beta F = N\log(2I+1) - \frac{N}{2}\int \frac{\mathrm{d}^3k}{V_k}\sum_{\alpha=1}^{3}\log\left(\frac{1-\beta\lambda_\alpha(\boldsymbol{k})\sigma_2\chi_0}{3}\right)$$
$$-\frac{3}{2}NR\chi_0 + N\sum_{n=0}^{\infty}\frac{1}{(n+1)!}\frac{\Sigma_{2n+2}}{\sigma_2^{n+1}}\left(\frac{3R}{2}\right)^{n+1} \tag{3.1}$$

R and χ_0 are given by the coupled equations

$$\chi_0 = \sum_{n=0}^{\infty}\frac{1}{n!}\frac{\Sigma_{2n+2}}{\Sigma_2^{n+1}}\left(\frac{3R}{2}\right)^{n} \tag{3.2}$$

$$R = \int \frac{\mathrm{d}^3k}{V_k}\frac{1}{3}\sum_{\alpha=1}^{3}\frac{\beta\lambda_\alpha(\boldsymbol{k})\sigma_2/3}{1-\beta\lambda_\alpha(\boldsymbol{k})\sigma_2\chi_0/3}. \tag{3.3}$$

The factor R describes the effect of adding one loop of all lengths to a vertex, save the change in the spin trace of that vertex. χ_0 then accounts for the adding of several loops. The functional dependence of χ_0 on R is determined by the spin quantum number. On the other hand, R as a function of $\beta\chi_0$ measures the eigenvalue spectrum. The last term in the expression (3.1) compensates for the overcounting. The HC free energy of the Ising model is analogous to (3.1).

The following expressions for the entropy and the specific heat are easily derived

$$S/Nk_B = \log(2I+1) - \frac{1}{2}\int \frac{\mathrm{d}^3k}{V_k}\sum_{\alpha=1}^{3}\log(1-\beta\lambda_\alpha(\boldsymbol{k})\sigma_2\chi_0/3)$$
$$-3R\chi_0 + \sum_{n=0}^{\infty}\frac{1}{(n+1)!}\frac{\Sigma_{2n+2}}{\sigma_2^{n+1}}\left(\frac{3R}{2}\right)^{n+1} \tag{3.4}$$

$$C/Nk_B = -\tfrac{3}{2}\beta(\mathrm{d}/\mathrm{d}\beta)(R\chi_0) + \tfrac{3}{2}R\chi_0. \tag{3.5}$$

For the staggered susceptibility $\chi(\boldsymbol{q})$ in zero field we have to include the Zeeman interaction

$$H_Z = B^\alpha\sum_i \exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r}_i)I_i^\alpha \tag{3.6}$$

in the Hamiltonian of equation (2.1). Here $\boldsymbol{B}$ is the scaled external field. The free energy expansion is then composed of diagrams with two Zeeman bonds in addition to the original spin–spin bonds. Within the Husimi tree approximation this expansion arises from the zero-field expansion when we choose in turn each vertex of the original diagrams with *two* bonds impinging on it. These two are then replaced by Zeeman bonds. There will be no overcounting as the chosen vertex is a root (see Domb 1974). The loop with the Zeeman bonds does not satisfy equation (2.9), but this is of no consequence as the other loops still do. The free energy is given by

$$-\beta F = \frac{\beta^2 B^2}{2} \frac{\chi_0 \sigma_2}{3} \sum_{n=0}^{\infty} (\beta \sigma_2 \lambda_Z(\boldsymbol{q}) \chi_0 / 3)^n \tag{3.7}$$

where we have taken a staggered external field $\boldsymbol{B} = B\hat{\boldsymbol{e}}_Z$ parallel to an eigenvector of the fourier transformed spin–spin interaction and with this vector as its wavenumber. The zero-field susceptibility readily emerges as

$$\chi^{-1}(\boldsymbol{q}) = (\beta \sigma_2 \chi_0 / 3)^{-1} - \lambda_Z(\boldsymbol{q}). \tag{3.8}$$

Choosing $\boldsymbol{q}$ as belonging to the maximum eigenvalue, $\lambda_{\max}$, the staggered susceptibility gives the critical point

$$k_B T_c = \tfrac{1}{3} \lambda_{\max} \sigma_2 \chi_0(T_c) = \chi_0(T_c) k_B T_c^{\mathrm{mf}} \tag{3.9}$$

where T_c^{mf} is the mean field estimate. This is also a singular point of the factor R (equation (3.3)). R must remain finite at the critical point to keep the free energy finite. The thermal properties of the system thus display only a weak singularity (at T_c the specific heat is $C/Nk_B = 3R\chi_0/2$).

We defer to the next section further discussion about the properties of our model and its comparison with other results. Instead we now return to the two approximations made in § 2. First consider the choice of the Husimi diagrams. There is a sum implied over the lattice for each vertex in a diagram. One thus expects diagrams with the largest number of vertices to dominate. This holds exactly for long-range interactions with high effective coordination numbers (cf Brout 1961). For a given number of bonds, the diagrams made of chains of bonds thus seem dominant. We note that a vertex with only one line impinging on it will make a diagram vanish, as the trace over a single spin operator is zero. Most vertices with an odd number of bonds also eliminate a diagram because of the antisymmetric properties of the spin traces. These are the basic facts leading to the choice of the Husimi trees.

The leading correction to the Husimi diagrams would be the inclusion of multiple bonds. Consider replacing the central bond in figure 3(a) with a doublet (figure 3(b)). The central single bond gives the Hamiltonian

$$\beta J^{\alpha\beta}(\boldsymbol{q}) I_1^{\alpha} I_2^{\beta} \tag{3.10}$$

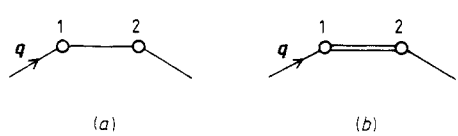
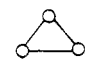
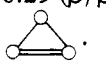


Figure 3. The indices shown refer to lattice sites (see text).

and the double bond, the effective one

$$-\frac{\beta^2}{24}\int \frac{d^3k}{V_k} J^{\alpha\beta}(\boldsymbol{k}) J^{\gamma\delta}(\boldsymbol{k}+\boldsymbol{q}) \varepsilon^{\alpha\gamma\mu} \varepsilon^{\beta\delta\kappa} I_1^{\mu} I_2^{\kappa} \tag{3.11}$$

if the adjacent bonds are single. The dominant contribution of (3.10) to the loop integral $\boldsymbol{J}_1$ (equation (2.20)) arises from the maximum eigenvalue of $J^{\alpha\beta}(\boldsymbol{q})$, $\lambda_{\max}$. Thus $\beta J^{\alpha\beta}(\boldsymbol{q})$ is the order of magnitude of unity or less in the critical region. One expects the contribution of (3.11) to be essentially smaller as the integral includes the smaller eigenvalues. On the other hand, the effect of one double bond grows larger with the number of bonds, one of which may be doubled. For the dipole–dipole interaction on the FCC lattice, we get the free energy contributions $-0.29\,(\beta/\beta_c^{mf})^3$ and $0.0014\,(\beta/\beta_c^{mf})^4$ corresponding to the diagrams  and  . The double bond becomes important in diagrams with roughly 600 bonds for $\beta \simeq \beta_c^{mf}$. One would certainly get a large correction even fairly far from the critical temperature by adding all the multiple bond (i.e. ladder) diagrams. It is clear then that the Husimi trees are severely inadequate in the critical region. Their representativity is much better at higher temperatures where only diagrams of low orders in β are important. On comparing the entropy of equation (3.4) with the exact high-temperature expansion for copper ($I = 3/2$) (Niskanen and Kurkijärvi 1981) we found good agreement down to temperatures of twice the mean-field critical point, below which the available expansion does not give the entropy accurately.

The second approximation of § 2 is the factorisation of the spin permutations. As we concentrate on the quantum effects on T_c, we do not believe this approximation to be critical. The corrections would be haphazard, positive or negative on a rather regular series of contributions which leads to equations (3.1)–(3.3). It seems highly unlikely to us that such corrections could affect the critical point of these equations or introduce new singularities. We expect in fact the spin factors of any spin trace to be exact to two orders of Σ_2 with apparently small errors in higher orders as discussed in appendix 2.

4. Results

In this section we discuss quantum spin effects on the thermodynamics of the system, particularly in the critical region. Our results are consistent with earlier calculations. The quantum system has a definitely lower transition temperature but is otherwise not different from the classical.

Consider first the expansion with only simple loops included, leading to the Gaussian approximation. The free energy is then given by

$$-\beta F = N \log(2I+1) - \frac{N}{2}\int \frac{d^3k}{V_k} \log(1-\beta\lambda_\alpha(\boldsymbol{k})\sigma_2/3) \tag{4.1}$$

and we see that the free energy of equation (3.1) reduces to the Gaussian free energy if we set

$$\chi_0 = 1 \quad \text{and} \quad R = 0. \tag{4.2}$$

At the critical point ($T = T_c^{mf}$) the Gaussian model behaves in an unphysical fashion (cf Brout 1960). In particular, the order parameter (staggered magnetisation) diverges.

Our expansion is essentially a vertex renormalisation of the Gaussian approximation like that of Horwitz and Callen (among others). One can say in general that vertex renormalised linked cluster expansions reduce the fluctuations of the order parameter and result in more reasonable thermodynamics.

In order to extract the thermodynamics from our calculation, we need the quantities χ_0 and R of equations (3.2) and (3.3) as functions of β. Equation (3.3) allows us to calculate $R\chi_0$ as a function of $\beta\chi_0$ for each interaction and lattice. The integral must be evaluated as a series expansion of $\beta\chi_0$; numerical integration with a discrete eigenvalue spectrum would lead to a divergence at the critical point because of the singularity there. The series expansion can be analysed with methods described by Joyce (1972). From the expansion of equation (3.2) we can then calculate $R\chi_0$ as a function of χ_0 alone. For $R \geqslant 1$ the expansion was analytically continued with Padé approximants (see e.g. Graves-Morris 1973). $R\chi_0$ as a function of χ_0 is plotted in figure 4 for representative values of the spin quantum number I. All in all, we know χ_0 and R as function of β, i.e. we have the full thermodynamics of the system.

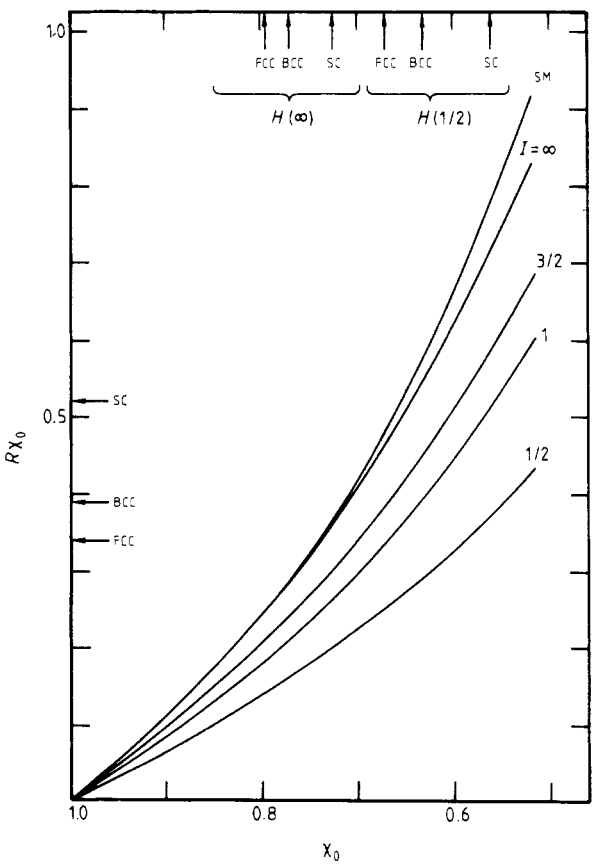


Figure 4. $R\chi_0$ as a function of χ_0 (equation (3.2)) for representative values of the spin quantum number I. SM denotes the spherical model result, equation (4.5). The arrows on the vertical axis display $(R\chi_0)_{max}$ for the cubic lattices. The arrows on the horizontal axis give the best known estimates of χ_{0C} (via equation (4.3)) for the Heisenberg models $I = 1/2(H(1/2))$ and $I = \infty(H(\infty))$ (Rushbrooke *et al* 1974).

In figure 4 we see that quantum spins affect the behaviour of the system appreciably. The maximum of $R\chi_0$ gives the critical value of χ_0, χ_{0c}, and the critical temperature as

$$T_c = \chi_{0c} T_c^{mf} \tag{4.3}$$

Watson has calculated $(R\chi_0)_{max}$ for the nearest-neighbour interaction on cubic lattices (Joyce 1972). These are indicated by arrows on the vertical axis of figure 4. The most accurate T_c's for the quantum Heisenberg nearest-neighbour interaction model have been provided by Rushbrooke *et al* (1974). These results, as interpreted via equation (4.3), are included in figure 4. Our model gives too low a T_c for all the cases included. If the maximum of $R\chi_0$ were lower, say $\simeq 0.37$ for the SC lattice, our calculation would give correct T_c's for *both* $I = \infty$ and $I = \frac{1}{2}$. We thus conclude that some simple imitation of the correct bond renormalisation should exist which would make our calculation a reliable tool for predicting T_c in the presence of quantum effects. For the Ising $I = \frac{1}{2}$ model our results agree with the zero-field limit of Horwitz and Callen.

It is interesting to take a look at the spherical model (SM) result (see Joyce 1972, for a review). The SM free energy is

$$-\beta F = N \log(2I+1) - \frac{N}{2} \int \frac{d^3k}{V_k} \sum_{\alpha=1}^{3} \log(1 - \beta\lambda_\alpha(\boldsymbol{k})\sigma_2\chi_0/3) + \tfrac{3}{2}N \log \chi_0 + \tfrac{3}{2}NR\chi_0 \tag{4.4}$$

with

$$R\chi_0 = (1/\chi_0) - 1 \tag{4.5}$$

to be compared with equations (3.1)–(3.3) of the Husimi expansion. The spherical model (SM) was initially derived for the Ising model (Berlin and Kac 1952) but there is nothing in the diagrammatic version of Brout (1961) which should make it different from the (quantum) Heisenberg model in zero external field above the transition temperature. In fact the only previous quantum vertex renormalisation is based on the same diagrams as SM (cf Stinchcombe *et al* 1963). The SM counts only a subclass of all the Husimi tree diagrams. In spite of this inferiority in terms of diagrams, the SM is essentially as good as our renormalisation for a classical spin system, as the SM result for $R\chi_0$ agrees closely with the Heisenberg model $I = \infty$. For a quantum system, the vertex renormalisation of SM is clearly inadequate. This also holds for the 'quantum SM' of Stinchcombe *et al* (1963). It should be mentioned that there is a limit at which SM is actually better than our seemingly more exact calculation. This is the classical $d = \infty$ limit† where our model cannot distinguish between $d = 3$ and $d = \infty$. Our approximation puts the $d = \infty$ limit on the line $I = \infty$ of figure 4 rather than on the SM line where it belongs according to the exact proof of Stanley (1969).

The critical indices of our model are, in close agreement with SM, entirely determined by the density of energy eigenvalues of the mean-field phases at the upper edge of the spectrum. Thus a transition is predicted to the mean-field structure. Assume the density of states near the edge $\varepsilon = 1$ given by $(\varphi < 1)$

$$g(\varepsilon) = g_0(1-\varepsilon)^\varphi + O[(1-\varepsilon)^{\varphi+1}] + \text{nonsingular terms.} \tag{4.6}$$

A little manipulation then leads to

$$\gamma = 1/\varphi \tag{4.7}$$

† Here d is the dimensionality of the spin space.

as in the spherical model (see Joyce 1972). Our expansion, however, does not predict a diverging temperature derivative of the specific heat at T_c in contrast to SM. For long-range interactions we get $\gamma = 1$ in agreement with the exact mean-field result. For nearest-neighbour interactions in cubic lattices, the SM result is $\gamma = 2$ (Joyce 1972) which is far from the best estimates $\gamma \simeq 5/4 \ldots 4/3$.

5. Conclusion

We present an approximate vertex renormalisation for a rather general quantum spin model. The results, restricted to the case of zero external field, display essential differences between the quantum and classical systems which are not present in the renormalisation of Stinchcombe *et al* (1963). The transition temperature estimates depend appreciably on the spin quantum number I.

Acknowledgments

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Appendix 1

In this appendix a recursion relation is derived for the spin trace σ defined in equation (2.14). Let us denote the spin operator I^α with the index α only. σ_{2m} is then defined as

$$\sigma_{2m} = \frac{1}{(2m)!}\frac{1}{(2I+1)}\sum_{\{P_{2m}\}} \mathrm{Tr}(\alpha\alpha\beta\beta \ldots \omega\omega) \tag{A1}$$

with m indices $\{\alpha, \beta, \ldots \omega\}$. We can calculate σ_{2m+2} by adding to σ_{2m} two 'new' spin operators Ω in all possible ways. Define an auxiliary function of the permutations $\{P'\}$ of the $2m$ 'old' spins

$$P_{2m}(j) = \sum_{\{P'\}} \mathrm{Tr}(\Omega\alpha\alpha\beta\beta \ldots \Omega \ldots \omega\omega). \tag{A2}$$

In $P_{2m}(j)$ there are j (or equally $2m-j$) old spins between the new. Because of the invariance of a trace in cyclic permutations

$$\sigma_{2m+2} = \frac{1}{(2m+2)!}\frac{2m+2}{2I+1}\sum_{j=0}^{m}(2-\delta_{j,m})P_{2m+2}(j) \tag{A3}$$

Using the commutator relations (2.2) and the definition (A2) we get

$$P_{2m+2}(j) = 2P_{2m+2}(j-1) - P_{2m+2}(j-2) - (2m)!\,(2I+1)\sigma_{2m} + 2m(2m+1)P_{2m}(j-2). \tag{A4}$$

It is easy to prove by induction that

$$P_{2m+2}(j)=\sum_{l=1}^{m}\frac{2I+1}{2(m-l)+3}(f_1(j,1)\sigma_2\sigma_{2(m+1-l)}-f_0(j,l)\Sigma_{2(m+1-l)}) \tag{A5}$$

where

$$\begin{aligned} f_1(j,l)&=\prod_{k=0}^{2l+3}(j-k)/(2l-2)! \\ f_0(j,l)&=\prod_{k=-1}^{2l+2}(j-k)/(2l)!. \end{aligned} \tag{A6}$$

Therefore the spin trace is given recursively by

$$\begin{aligned} \sigma_{2m+2}=\sum_{l=1}^{m}\frac{1}{2l+1}\Bigg[&2\binom{m}{2l-m-1}\sigma_2\sigma_{2l}-2\binom{m+1}{2l-m-2}\sigma_{2l} \\ &+\binom{m}{2l-m}\sigma_2\sigma_{2l}-\binom{m+1}{2l-m-1}\sigma_{2l}\Bigg] \end{aligned} \tag{A7}$$

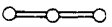
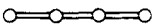
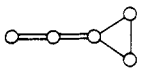
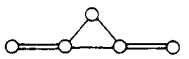
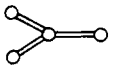
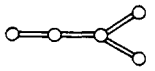
with the definition

$$\binom{p+q}{p}=0 \qquad \text{if } p<0 \text{ or } q<0. \tag{A8}$$

For example

$$\sigma_2=I(I+1) \qquad \sigma_4=\sigma_2^2-\tfrac{1}{3}\sigma_2 \qquad \sigma_6=\sigma_2^3-\sigma_2^2+\tfrac{1}{3}\sigma_2 \tag{A9}$$

Table 1. The exact mean reduced traces of Rushbrooke *et al* (1974) (they use X for σ_2) compared with our approximate results for some diagrams. We have omitted the inessential symmetry factors.

Diagram	Mean reduced trace	Our result
	$\sigma_2^4\left(1-\frac{1}{3\sigma_2}\right)$	$\sigma_4\sigma_2^4=\sigma_2^4\left(1-\frac{1}{3\sigma_2}\right)$
	$\sigma_2^6\left[\left(1-\frac{1}{3\sigma_2}\right)^2+\frac{1}{9\sigma_2^2}\right]$	$\sigma_4^2\sigma_2^2=\sigma_2^6\left(1-\frac{1}{3\sigma_2}\right)^2$
	$\sigma_2^7\left[\left(1-\frac{1}{3\sigma_2}\right)^2+\frac{1}{9\sigma_2^2}\right]$	$\sigma_4^2\sigma_2^3=\sigma_2^7\left(1-\frac{1}{3\sigma_2}\right)^2$
	$\sigma_2^7\left(1-\frac{1}{3\sigma_2}\right)^2$	$\sigma_4^2\sigma_2^3$
	$\sigma_2^8\left[\left(1-\frac{1}{3\sigma_2}\right)^3+\frac{14}{315\sigma_2^2}-\frac{16}{315\sigma_2^3}\right]$	$\sigma_4^3\sigma_2^2=\sigma_2^8\left(1-\frac{1}{3\sigma_2}\right)^3$
	$\sigma_2^6\left(1-\frac{1}{\sigma_2}+\frac{1}{3\sigma_2^2}\right)$	$\sigma_6\sigma_2^3=\sigma_2^6\left(1-\frac{1}{\sigma_2}+\frac{1}{3\sigma_2^2}\right)$
	$\sigma_2^8\left[\left(1-\frac{1}{3\sigma_2}\right)\left(1-\frac{1}{\sigma_3}+\frac{1}{3\sigma_2^2}\right)\right.$	$\sigma_6\sigma_4\sigma_2^2=$
	$\left.+\frac{14}{315\sigma_2^2}-\frac{8}{315\sigma_2^3}\right]$	$\sigma_2^8\left(1-\frac{1}{3\sigma_2}\right)\left(1-\frac{1}{\sigma_2}+\frac{1}{3\sigma_2^2}\right)$

Appendix 2.

Rushbrooke *et al* (1974) have calculated the exact contributions of the quantum spins for diagrams with eight bonds or less (with a pure exchange interaction). We list their mean reduced traces in table 1 together with our approximate spin traces for some representative diagrams. On the basis of the table we conjecture that the error in the spin traces of any Husimi tree is small, of the order of magnitude $0.1\,\sigma_2^{-2}$.

We have also studied the breakdown of our approach in several diagrams. On that basis it seems safe to state that our result is correct for all diagrams with no loops containing only articulation vertices. We have, however, not proved any general theorem about the accuracy of our spin factorisation.

References

Ambler E, Eisenstein J C and Schooley J F 1962 *J. Math. Phys.* **3** 118
Berlin T H and Kac M 1952 *Phys. Rev.* **86** 821
Brout R 1960 *Phys. Rev.* **118** 1009
—— 1961 *Phys. Rev.* **122** 469
Domb C 1974 in *Phase Transitions and Critical Phenomena* vol 3, ed C Domb and M S Green (New York: Academic)
Ehnholm G J, Ekström J P, Jacquinot J F, Loponen M T, Lounasmaa O V and Soini J K 1979 *Phys. Rev. Lett.* **42** 1702
Englert F 1963 *Phys. Rev.* **129** 567
Graves-Morris P R (ed) 1973 *Padé Approximants and their Applications* (New York: Academic)
Griffiths R B 1968 *Phys. Rev.* **176** 655
Horwitz G and Callen H B 1961 *Phys. Rev.* **124** 1757
Joyce G S 1972 in *Phase Transitions and Critical Phenomena* vol 2, ed C Domb and M S Green (New York: Academic) p 375ff
Kadanoff L P 1976 *Phase Transitions and Critical Phenomena* vol 5a, ed C Domb and M S Green (New York: Academic) p 16ff
Kubota M, Folle H R, Buchel A, Mueller R M and Pobell F 1980 *Phys. Rev. Lett.* **45** 1812
Niskanen K J and Kurkijärvi J 1981 *J. Phys. C: Solid St. Phys.* **14** 5517
Percus J K 1971 *Combinatorial Methods* (New York: Springer-Verlag) p 7
Rushbrooke G S, Baker G A and Wood P J 1974 in *Phase Transitions and Critical Phenomena* vol 3, eds C Domb and M S Green (New York: Academic) p 245ff
Stanley H E 1969 *Phys. Rev.* **179** 570
Stinchcombe R B, Horwitz G, Englet F and Brout R 1963 *Phys. Rev.* **130** 155
Wortis M 1974 *Phase Transitions and Critical Phenomena* vol 3, ed C Domb and M S Green (New York: Academic) p 114ff
Wortis M, Jasnow D and Moore M A 1969 *Phys. Rev.* **185** 805