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Second-Order Green's Function Theory of a Heisenberg Paramagnet

Sam A. Scales[1] and H. A. Gersch[1]

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The Heisenberg paramagnet in one, two, and three dimensions is analyzed by a second-order Green's function theory similar to that used by Knapp and ter Haar. This theory, which incorporates the exact values for the zero, first, and second moments of the relaxation function as boundary conditions, yields results satisfying the rotational symmetry of the paramagnetic region as well as the principle of detailed balance. We find that our predictions for equal time properties in the classical limit are identical with the RPA Green's function theory of Liu as well as the spherical model results of Lax. The quantum limit is analyzed, and our predictions for the $1/T$ series coefficients for both internal energy and susceptibility are compared with exact results.

KEY WORDS: Heisenberg paramagnet; Green's functions; relaxation functions.

1. INTRODUCTION

The Heisenberg model has been extensively studied during the last several years using the techniques of double time temperature-dependent Green's functions. Formally, the problem of finding the solution for magnetization and time-dependent spin–spin correlation functions is reduced to the determination of the solution to an infinite set of coupled first-order differential equations. The development of a tractable formalism necessitates a decoupling approximation.

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[1] School of Physics, Georgia Institute of Technology, Atlanta, Georgia.

A majority of the decoupling procedures which have been utilized on the Heisenberg model have been made at the first stage of the calculation, since further delay introduces an enormous mathematical complexity to the problem. The random phase approximation (RPA) of Bogoliubov and Tyablikov(1-4) has been used extensively in many slightly modified forms. A short and somewhat representative sample of these theories is given by Refs. 1-15.

These first-order theories are largely designed to apply to systems in the ordered state, and therefore the failure of the excitation energies to agree with experiment and the exactly known properties of the Heisenberg model in the paramagnetic region is perhaps understandable. One finds that all these theories, with the notable exception of Lines's,(14) produce excitation energies for which the scaling with temperature is *independent of wave vector*. A further examination of these theories shows that all spin wave energies are *vanishingly small* in the paramagnetic region. Both neutron diffraction(15-19) and Raman(20,21) experiments on magnetic systems reveal that short-wavelength excitations remain finite and continue to propagate well into the paramagnetic region. This phenomenon is most apparent in one- and two-dimensional systems.(22-24) Of the literature cited,(1-14) only Lines(14) has a first-order Green's function theory for the paramagnetic region that is even qualitatively correct and his approach, which is phenomenological in nature, leaves obscured the underlying basis for the theory.

In Section 2, we define both the Green's functions and the corresponding relaxation functions which are pertinent to the Heisenberg model, as well as quote some of the spectral relations that are developed in the literature.(25,26) By using arguments based on the invariance properties of the Heisenberg paramagnet as well as exact moments of the relaxation function, we conclude that a second-order Green's function theory is the best approach because the second-order theory incorporates exactly the zero, first, and second moments of the relaxation function as boundary conditions for the Green's function equation ensuring nonvanishing excitation energies in the paramagnetic region.

In Section 3, we describe a decoupling of the Green's function equations which is a modification of the procedure used by Knapp and ter Haar(28) and is similar to theories used by Richards,(29) Lo and Halley,(30) and ourselves.(27)

In Section 4, we make a further approximation to the Green's function equations obtained from the truncation used in Section 3. We tentatively identify this approximation as the classical limit while deferring the proof to Section 6. The equal time properties predicted by our theory in the absence of a magnetic field in the classical limit are shown to be identical with the predictions of the RPA theory of Liu.(5) It is also found that our thermodynamics reduces to the spherical model results in the classical limit, as did the theory reported by Lo and Halley.(30)

In Section 5, we examine quantum effects that were neglected in the classical limit approximation of Section 4. In particular we derive corrections to the Curie and Néel temperatures predicted by the RPA and spherical models by our theory to order $1/S(S+1)$. The critical value of the nearest-neighbor spin–spin correlation function is also calculated to the same level of approximation. Comparisons of our results with $(1/T)$ series results are given in Tables I and II.

Finally, in Section 6, we examine the $1/T$ series expansion for the susceptibility and nearest-neighbor correlation function predicted by our theory, and as a by-product obtain the $1/T$ series expansions for the spherical model, the RPA theory, and the RPA theory of Liu. Our theory is found to give a much more accurate description of the high-temperature thermodynamics than either the spherical model or the RPA theory of Liu especially for small spin values, as may be seen by an examination of the $1/T$ series coefficients given in Appendices A and B.

2. GREEN'S FUNCTION THEORY

The usual retarded and advanced Green's functions reviewed by Zubarev(25) are defined by

$$G_{\substack{r\\a}}(t) = \langle\langle A(t); B(0)\rangle\rangle_{\substack{r\\a}} = \mp i\theta(\pm t)\langle[A(t), B(0)]\rangle \tag{1}$$

where

$$\theta(t) = 1, \quad t > 0; \qquad \theta(t) = 0, \quad t < 0 \tag{2}$$

$$[A, B] = AB - BA \tag{3}$$

$$A(t) = e^{iHt} A(0)\, e^{-iHt} \tag{4}$$

$$\langle\cdots\rangle = Z^{-1}\,\mathrm{Tr}(e^{-\beta H}\cdots) \tag{5}$$

$$\beta = 1/k_B T \tag{6}$$

and H is the Hamiltonian defined by

$$H = -J \sum_{j,d} \mathbf{S}_j \cdot \mathbf{S}_{j+d} \tag{7}$$

where J is the exchange constant in units of energy (we use units where Planck's constant is equal to 2π) and where the sum over the spin index j ranges over the positions of all N lattice sites, while the sum over d ranges over the set of nearest-neighbor vectors only.

Upon taking the time derivative of (1) and utilizing (3) and (4) in the resulting equation, one finds that both advanced and retarded Green's functions satisfy

$$i(\partial/\partial t)\langle\langle A(t); B(0)\rangle\rangle_{\substack{r\\a}} = \langle[A, B]\rangle\,\delta(t) + \langle\langle [A, H](t); B(0)\rangle\rangle_{\substack{r\\a}} \tag{8}$$

We have used the convention that all operators to the left of the semicolon in the Green's function appearing on the right-hand side of (8) are to be evaluated at the time t. We define the image $\tilde{A}(\omega)$ of an arbitrary function of the time $A(t)$ under the Fourier transform operator $F(\omega; t)$ via

$$\tilde{A}(\omega) = F(\omega; t)\, A(t) = (1/2\pi)\int_{-\infty}^{\infty} dt\, e^{i\omega t} A(t) \tag{9}$$

It is convenient to define a relaxation function $R(\omega)$ by

$$R(\omega) = i\omega^{-1}[F(\omega + i\epsilon; t)G_r(t) - F(\omega - i\epsilon; t)G_a(t)] \tag{10}$$

The correlation function $\langle B(0)A(t)\rangle$, which is of ultimate interest, is given by the spectral relation[25]

$$\langle B(0)A(t)\rangle = F^{-1}(t; \omega)\omega R(\omega)/(e^{\beta\omega} - 1) \tag{11}$$

where $F^{-1}(t; \omega)$ is the inverse Fourier transform operator.

The relevant Green's functions for the Heisenberg model are given by

$$G_{mn}(j, t) = \langle\langle S_j^m(t); S_0^n(0)\rangle\rangle \tag{12}$$

where m and n are labels which may take on any of the values $+$, $-$, x, y, or z. To each Green's function defined by (12) there corresponds a Green's function defined by

$$G_{mn}(k, t) = F_{k;j}G_{mn}(j, t) \tag{13}$$

where the Fourier lattice transform operator $F_{k;j}$ is defined by

$$F_{k;j}\cdots = \sum_j e^{ik\cdot j}\cdots \tag{14}$$

To each Green's function defined by (12) and (13) there corresponds a relaxation function defined by (10). The moments of the relaxation functions, which we denote by $\langle\omega_k{}^s\rangle_{mn}$, are defined by

$$\langle\omega_k{}^s\rangle_{mn} = \int_{-\infty}^{\infty} \omega^s R_{mn}(k, \omega)\, d\omega \Big/ \int_{-\infty}^{\infty} R_{mn}(k, \omega)\, d\omega \tag{15}$$

These moments are related in an important way to the boundary conditions on the Green's functions at time $t = 0$. We find

$$\langle \omega^s \rangle R(t{=}0) = (i\,\partial/\partial t)^s R(t)_{t=0} = i(i\,\partial/\partial t)^{s-1} G_r(t)_{t=0^+} \tag{16}$$

where $R(t)$ is the inverse Fourier transform of $R(\omega)$ defined by (10).

We are now in a position to determine what is wrong with the use of a first-order Green's function truncation in the paramagnetic region. We have that $G_{+-,r}(k, t = 0^-) = -2i\langle S_0^z \rangle$. Therefore it follows from (16) that

$$\langle \omega_k \rangle_{+-} = 2\langle S_0^z \rangle / R_{+-}(k, t = 0) = \langle S_0^z \rangle / \chi_k \tag{17}$$

where we have used the fact that $\frac{1}{2}R_{+-}(k, t = 0) = R_{zz}(k, t = 0) = \chi_k$ in the paramagnetic region due to the spherical symmetry.(26) The function χ_k is just the wavelength-dependent paramagnetic susceptibility. If one insists that the Green's functions have only one pole corresponding to the magnon energy, as is the case in the literature cited,(1–11) then the magnon energy must be given by

$$\tilde{E}_k = \langle \omega_k \rangle_{+-} = \langle S_0^z \rangle / \chi_k \tag{18}$$

as a result of (17). One must in fact solve the Green's function equations in the limit of vanishing magnetic field in order to get a solution since $\langle S_0^z \rangle$ is identically zero when the magnetic field is identically zero.

It is clear, however, that the Green's function for $G_{+-}(k, t)$ must have two poles in the paramagnetic region (to the extent that the concept of elementary excitation is valid) since the operators $F_{k;j} S_j^-$ and $F_{k;j} S_j^-$ in the absence of a magnetic field must by symmetry have equal probability for creating and destroying a magnon. Therefore (17) should be interpreted as a measure of the asymmetry in the location of the poles in the Green's function corresponding to the propagation of a magnon and a magnon hole.

We now consider the information contained in the boundary condition for the second moment of the relaxation function in the paramagnetic region. We obtain

$$\langle \omega_k \rangle_{zz} = 0; \qquad \langle \omega_k^2 \rangle_{zz} = 2J(\gamma_0 - \gamma_k)\langle S_0^x S_d^x + S_0^y S_d^y \rangle / \chi_k \tag{19}$$

from the relaxation function $R_{zz}(k, t)$. The lattice site d appearing in (19) refers to any one of the nearest-neighbor sites of the site 0. The function γ_k is defined by

$$\gamma_k = \sum_{d'} \exp(ik \cdot d') \tag{20}$$

where the sum over d' ranges over the set of nearest-neighbor displacement vectors. Notice that (19) is consistent with a relaxation function given by

$$R_{zz}(k, t) = \chi_k \cos(E_k t) \tag{21}$$

where

$$E_k^2 = \langle \omega_k^2 \rangle_{zz} = 2J(\gamma_0 - \gamma_k) \langle S_0^x S_d^x + S_0^y S_d^y \rangle / \chi_k \tag{22}$$

Equation (21) is obtained by assuming that for a given k, the relaxation function contains a pure negative frequency corresponding to the presence of a magnon of energy E_k and a positive frequency component corresponding to the presence of a magnon hole of energy $-E_k$. The relative amplitude and phase of the two frequency components are fixed by the principle of detailed balance. More explicitly, Marshall and Lowde(26) have shown that for crystals with inversion symmetry, $R_{zz}(k, t)$ must be an even function of t, which they show is equivalent to the statement of detailed balance. Finally, (19) determines the energy of the excitations to be given by (22).

It must be emphasized that (21) and (22) are meaningful only to the extent that it is possible to describe the behavior of the Heisenberg paramagnet in terms of elementary excitations. The experimental data(15-24) suggest this assumption is realized most strongly in systems of low dimensionality. In particular, the experimental measurement of the excitations of TMMC (a one-dimensional Heisenberg paramagnet with $S = 5/2$ and $J = -7.7$°K) have revealed the existence of elementary excitations obeying

$$E_k = (6.1 \text{ meV}) |\sin k| \tag{23}$$

over the entire Brillouin zone at 4.4°K.(21) We have elsewhere(27) shown in a preliminary account of the present Green's function theory that our Green's function theory is able to explain the excitation spectra given by (23) for TMMC at 4.4°K.

In Section 3, we shall use a truncation procedure on the Green's function equations at second order. This truncation scheme is a modification of a scheme used earlier by Knapp and ter Haar(28) and is similar to a scheme also reported by Richards(69) and Lo and Halley.(30) We choose a second-order decoupling scheme because the second moment sum rule for the relaxation function given by (19) is incorporated into the Green's function as a boundary condition. Furthermore, the symmetry of the second-order equation of motion for $G_{zz}(j, t)$ guarantees that the condition of detailed balance is satisfied, and this feature is easily retained in the truncation. In fact, our theory has a relaxation function given by (21) and excitation energies given by (22). These equations are supplemented by (42)–(46), which define a self-consistent scheme for determining the unknown susceptibility and nearest-neighbor correlation function appearing in (21) and (22).

One may justifiably ask why we should use a theory having a relaxation function given by (21) in three dimensions at high temperature when both experimental and theoretical studies indicate that the relaxation function should be Gaussian in ω for large values of k and Lorentzian in ω for small k.(26) The answer is that the dynamics predicted by (21) is certainly unrealistic; however, the static properties are determined by only low-order moments $\langle \omega_k{}^s \rangle$ of the relaxation function and these are correctly represented in our theory. For example, we find from (11) that

$$F_{k;j}\langle S_0{}^z S_j{}^z \rangle \beta = \chi_k(1 \times c_1\beta \langle \omega_k \rangle_{zz} + c_2\beta^2 \langle \omega_k{}^2 \rangle_{zz} + \cdots) \tag{24}$$

Similar remarks hold for the Heisenberg paramagnet at high temperatures in the cases of one and two dimensions.

3. DECOUPLING OF THE EQUATIONS OF MOTION

The Green's function $G_{zz}(j, t)$ is found to satisfy a second-order equation,

$$\begin{aligned}(i\,\partial/\partial t)^2\, G_{zz}(j, t) & \\ = 2J \sum_{d} \langle S_0{}^x S_d{}^x + S_0{}^y S_d{}^y \rangle(\delta_{j,0} - \delta_{j+d,0})\,\delta(t) & \\ + 4J^2 \sum_{d,d'} \langle\langle (\mathbf{S}_j * \mathbf{S}_{j+d} S^z_{j+d+d'} - \mathbf{S}_j * \mathbf{S}_{j+d+d'} S^z_{j+d})(t); S_0{}^z(0) \rangle\rangle & \\ + 4J^2 \sum_{d,d'} \langle\langle (\mathbf{S}_{j+d} * \mathbf{S}_{j+d'} S_j{}^z - S^z_{j+d} \mathbf{S}_j * \mathbf{S}_{j+d'})(t); S_0{}^z(0) \rangle\rangle &\end{aligned} \tag{25}$$

after using (8) two times and simplifying the resulting equation with the aid of the spin angular momentum commutation relations. The operation $\mathbf{S}_{j_1} * \mathbf{S}_{j_2}$ appearing in (25) is presently to be interpreted as the usual dot product of two vectors. Knapp and ter Haar(28) proceeded to approximate the Green's functions on the right-hand side of (25) by

$$\langle\langle \mathbf{S}_{j_1} * \mathbf{S}_{j_2} S^z_{j_3}(t); S_0{}^z(0) \rangle\rangle \simeq \langle \mathbf{S}_{j_1} * \mathbf{S}_{j_2} \rangle\, G_{zz}(j_3, t) \tag{26}$$

$$\langle\langle S^z_{j_3} \mathbf{S}_{j_1} * \mathbf{S}_{j_2}(t); S_0{}^z(0) \rangle\rangle \simeq \langle \mathbf{S}_{j_1} * \mathbf{S}_{j_2} \rangle\, G_{zz}(j_3, t) \tag{27}$$

Knapp and ter Haar described this truncation as a natural generalization of the first-order RPA decoupling scheme. We refer the reader to their paper for a discussion of the ambiguities that arise in their truncation procedure.

Our truncation differs from that of Knapp and ter Haar in that we notice that the Green's functions appearing on the right-hand side of (25) which involve only z-component spin operators when summed yield identically zero for all values of j and t. Therefore we interpret the operator $\mathbf{S}_{j_1} * \mathbf{S}_{j_2}$ as

$$\mathbf{S}_{j_1} * \mathbf{S}_{j_2} = S^x_{j_1} S^x_{j_2} + S^y_{j_1} S^y_{j_2} \tag{28}$$

and (25) is still exact. We then truncate the Green's function equations using (26) and (27).

We now examine the reason we think that (26) and (27) are appropriate when the $*$ product is defined by (28) and are not appropriate when the $*$ product is interpreted as the ordinary dot product. For $n = x$ or y (but not z), we use

$$\begin{aligned}\langle\langle S_{j_1}^n S_{j_2}^n S_{j_3}^z(t); S_0{}^z(0)\rangle\rangle &\simeq \langle S_{j_1}^n S_{j_2}^n\rangle\, G_{zz}(j_3\,, t)\\ &\quad + \langle S_{j_1}^n S_{j_3}^z\rangle\, G_{nz}(j_2\,, t) + \langle S_{j_2}^n S_{j_3}^z\rangle\, G_{nz}(j_1\,, t)\\ &= \langle S_{j_1}^n S_{j_2}^n\rangle\, G_{zz}(j_3\,, t) \end{aligned} \tag{29}$$

The equality in (29) follows from the fact that both the correlation functions $\langle S_{j_1}^n S_{j_3}^z\rangle$ and $\langle S_{j_2}^n S_{j_3}^z\rangle$ are rigorously zero for $n = x$ or y (but not z). This may be seen most easily if the trace is taken in a basis in which the states are eigenstates of the z-component of total spin. Similar arguments apply to (27).

We will utilize the symmetries of the isotropic Heisenberg model with nearest-neighbor interactions, periodic boundary conditions, and equivalence of nearest-neighbor sites to obtain a compact form for the truncated equations of motion contained in (26) and (27). We have

$$\langle S_{j_1}^x S_{j_2}^x\rangle = \langle S_{j_1}^y S_{j_2}^y\rangle = \langle S_{j_1}^z S_{j_2}^z\rangle = \langle S_{j_1-j_2}^z S_0{}^z\rangle \tag{30}$$

and

$$\langle S_d{}^z S_0{}^z\rangle = \langle S_{d'}^z S_0{}^z\rangle, \quad d \neq d'; \qquad \sum_{d,d'} \langle S_{d+d'}^z S_0{}^z\rangle = \gamma_0 \sum_d \langle S_{d+d_1}^z S_0{}^z\rangle \tag{31}$$

where j_1 and j_2 are arbitrary lattice vectors, d, d', and d_1 are nearest-neighbor displacement vectors, and γ_0 is the number of nearest-neighbor displacement vectors.

It is important to retain the fact that (26) and (27) are only approximately true in our truncation since there will be more equations than there are unknowns, which can easily lead to a system of equations for which there exists no solution. In anticipation of the problems that will arise, we treat (26) and (27) as exact when $|j_1 - j_2| = |d|$. This leads to

$$\sum_{d,d} \langle\langle \mathbf{S}_j * \mathbf{S}_{j+d} S_{j+d+d'}^z(t); S_0{}^z(0) = 2f \sum_{d,d'} G_{zz}(j + d + d', t) \tag{32}$$

and

$$\sum_{d,d'} \langle\langle S_{j-d}^z \mathbf{S}_j * \mathbf{S}_{j+d'}(t); S_0{}^z(0)\rangle\rangle = 2f\gamma_0 \sum_d G_{zz}(j + d, t) \tag{33}$$

where

$$f = \langle S_0{}^z S^z_{d_1} \rangle \tag{34}$$

However, when $|j_1 - j_2| \neq |d|$, we do not demand a strict equality in (26) and (27) unless $j_1 = j_2$. We instead make the approximation

$$\sum_{d,d'} \langle\langle \mathbf{S}_j * \mathbf{S}_{j+d+d'} S^z_{j+d}(t); S_0{}^z(0) \rangle\rangle = 2g\gamma_0 \sum_d G_{zz}(j+d, t) \tag{35}$$

and

$$\sum_{d,d'} \langle\langle \mathbf{S}_{j+d} * \mathbf{S}_{j+d'} S_j{}^z(t); S_0{}^z(0) \rangle\rangle = 2g\gamma_0{}^2 G_{zz}(j, t) \tag{36}$$

where

$$cg = \gamma_0^{-1} \sum_d \langle S_0{}^z S^z_{d+d'} \rangle \tag{37}$$

and where c is a constant that may deviate from unity. After combining (32)–(37), we find

$$\begin{aligned} -(\partial^2/\partial t^2)\, G_{zz}(j, t) = {} & 4J\delta(t)\, f \sum_d (\delta_{j,0} - \delta_{j+d,0}) \\ & + 8J^2 \sum_{j'} G_{zz}(j+j', t) \\ & \times \sum_{d,d'} (f\delta_{j',d+d'} - f\delta_{j',d} + g\delta_{j',0} - g\delta_{j',d}) \end{aligned} \tag{38}$$

We see that (38) is a difference equation which is easily solved by use of the Fourier lattice transform defined by (14). We obtain

$$G_{zz(^r_a)}(k, t) = \mp 4\theta(\pm t)\, Jf(\gamma_0 - \gamma_k)[\sin(E_k t)]/E_k \tag{39}$$

where

$$E_k{}^2 = 8J^2(\gamma_0 - \gamma_k)(g\gamma_0 - f\gamma_k) \tag{40}$$

The relaxation function is found to be

$$R_{zz}(k, t) = f[\cos(E_k t)]/2J(g\gamma_0 - f\gamma_k) \tag{41}$$

upon substitution of (39) into (10). The wavelength-dependent susceptibility χ_k is identified from (41) and (21) as

$$\chi_k = f/2J(g\gamma_0 - f\gamma_k) \tag{42}$$

The susceptibility given by (42) is of the Ornstein–Zernike(31) form. The present theory will be complete once the temperature dependence of the parameters f and g are determined.

Upon the substitution of the Fourier transform of (41) into (11), we obtain

$$\langle S_0^z(0)\, S_j^z(t)\rangle = F_{j;k}^{-1}\{\tfrac{1}{2}\chi_k E_k[\cosh(\tfrac{1}{2}\beta E_k + iE_k t)]/\sinh(\tfrac{1}{2}\beta E_k)\} \tag{43}$$

for the time-dependent spin–spin correlation functions. The parameters f and g are determined from (43) by the self-consistency relations,

$$S(S+1)/3 = \langle S_0^z(0)\, S_0^z(0)\rangle = \frac{1}{\beta N}\sum_k \frac{fV(k,T)}{2J(g\gamma_0 - f\gamma_k)} \tag{44}$$

and

$$f = \gamma_0^{-1}\sum_d \langle S_0^z(0)\, S_d^z(0)\rangle = \frac{1}{\beta N}\sum_k \frac{f\gamma_k V(k,T)}{2J(g\gamma_0 - f\gamma_k)\,\gamma_0} \tag{45}$$

where

$$V(k,T) = \tfrac{1}{2}\beta E_k \coth(\tfrac{1}{2}\beta E_k) \tag{46}$$

There is an additional self-consistency equation that determines the constant c given in (37). It is

$$cg = \gamma_0^{-2}\sum_{d,d'} \langle S_0^z(0)\, S_{d+d'}^z(0)\rangle = \frac{1}{\beta N}\sum_k \frac{f\gamma_k^2 V(k,T)}{2J(g\gamma_0 - f\gamma_k)\,\gamma_0^2} \tag{47}$$

Our Green's function theory is now complete. We shall find out how well this theory, which is contained in (40)–(46), describes the behavior of the Heisenberg model. One can determine qualitatively the behavior of the theory by examining the classical spin limit, which we now consider.

4. CLASSICAL LIMIT

We now proceed to recover the classical limit for the equal time properties of the present theory and show that the thermal properties of our theory are the same as those predicted by the RPA theory of Liu(5) as well as the spherical approximation on the Heisenberg model by Lax.(33) The description of the equal time properties is contained in (40), (44)–(46), and the equation

$$\langle S_0^z(0)\, S_j^z(0)\rangle = \frac{1}{\beta N}\sum_k \frac{f[\exp(-ik\cdot j)]\, V(k,T)}{2J(g\gamma_0 - f\gamma_k)} \tag{48}$$

which we deduced by substituting (42) into (43) and using the inverse of the Fourier lattice transform operator defined by (14). In Section 6, we will prove that the classical limit corresponding to this system of equations is given by (44) and (48) where one uses $V(k,T) = 1$ instead of the expression given by (46).

Our theory in the classical limit has only one unknown parameter, $h = f/g$. One can also use χ_0 determined from (42) with $k = 0$ as the unknown parameter. The classical limit of (48) is then

$$\langle S_0^z(0)\, S_j^z(0)\rangle = \frac{1}{\beta N} \sum_k \frac{\exp(-ik \cdot j)}{\chi_0^{-1} + 2J(\gamma_0 - \gamma_k)} \tag{49}$$

where the susceptibility χ_0 is determined from

$$\frac{S(S+1)}{3} = \frac{1}{\beta N} \sum_k \frac{1}{\chi_0^{-1} + 2J(\gamma_0 - \gamma_k)} \tag{50}$$

which is deduced from the classical limit of (44).

In the case of $J = -|J|$, it is more reasonable to express (49) and (50) in terms of the staggered susceptibility χ_s when it can be defined. More precisely, when a lattice is decomposable into two interpenetrating sublattices with the property that nearest neighbors of a point on one sublattice all lie on the other sublattice, then one may define a staggered susceptibility. Then there exists at least one wave vector k_0 such that $\exp(ik_0 \cdot j) = \pm 1$ for all lattice sites j. The lattice points for which $\exp(ik_0 \cdot j) = 1$ are on one sublattice, while the points for which $\exp(ik_0 \cdot j) = -1$ are on the other sublattice. The staggered susceptibility is then determined from (42) by $\chi_s = \chi_{k_0}$. The important wave vectors when $J = -|J|$ are those wave vectors for which $|k - k_0|$ is small. We define a new wave vector q by

$$q = k - k_0 \tag{51}$$

so that (49) and (50) now become

$$\langle S_0^z(0)\, S_j^z(0)\rangle = \frac{1}{\beta N} \sum_q \frac{\exp[-i(k_0 + q) \cdot j]}{\chi_s^{-1} + 2|J|(\gamma_0 - \gamma_q)} \tag{52}$$

and

$$\frac{S(S+1)}{3} = \frac{1}{\beta N} \sum_q \frac{1}{\chi_s^{-1} + 2|J|(\gamma_0 - \gamma_q)} \tag{53}$$

respectively.

The equal time spin–spin correlation functions which one obtains from (49) and (50) are identical with those obtained by Liu(5) when he restored the spherical symmetry to the RPA Green's function theory for the quantum Heisenberg model in the paramagnetic region. It remains an unanswered question why the classical limit for the equal time spin–spin correlations given by (49) and (50), which were calculated by a second-order Green's function theory with finite frequencies given by (22), are identical with the equal time spin-spin correlations obtained by Liu from the first-order RPA Green's function approximation in which the excitation energies become zero for all wave vectors in the limit of zero magnetic field in accordance with (18). This identity strengthens the parallelism between the present theory and the RPA theory mentioned by Knapp and ter Haar.(28)

About ten years ago, Tahir-Kheli and ter Haar(6) pointed out that their RPA Green's function theory produced the same critical temperatures as the spherical model of Lax.(5) We now indicate that the equal time spin-spin correlations predicted by the RPA Green's function of Liu are the same as those predicted by the spherical model of Lax. This identity is most easily obtained by a treatment of the Heisenberg model in the spherical approximation paralleling the development leading to Eq. (36) in the paper by Berlin and Kac(33) devoted to the spherical approximation to the Ising model. The transition to the spherical approximation results for the Heisenberg model from the spherical model results of Berlin and Kac requires only a redefinition of the symbols since the new degrees of freedom S_j^x and S_j^y are not dynamically coupled to S_j^z in the spherical approximation. It remains an unanswered question why the equal time spin-spin correlation functions predicted by the present theory are equivalent to both the spherical model of Lax and the RPA theory of Liu. This identity, however, gives us an insight into the behavior of our model in one, two, and three dimensions.

The classical limit of our theory as defined by (49) and (50) predicts that a transition from the paramagnetic state to a state of long-range order is possible only in three dimensions,(33) in agreement with the proof of Mermin and Wagner.(34) For $J > 0$, one finds: a second-order phase transition from the paramagnetic to the ferromagnetic state; spin-spin correlation functions of the Ornstein-Zernike(31) form in the critical region(33); susceptibility obeying a $(T - T_c)^2$ law in the critical region(33) instead of a $(T - T_c)^{1/3}$ law predicted by early $1/T$ series methods;(35) and the predicted specific heat remains finite at T_c, in disagreement with the $1/T$ series result.(36) For $J < 0$, one must distinguish between two distinct cases. For those lattices for which it is possible to define a staggered susceptibility, one finds a transition from the paramagnetic state to the antiferromagnetic state only in the three-dimensional case, in agreement with the theorem of Mermin and Wagner.(34) The critical properties are analogous to those for the ferromagnetic transition, as is evidenced by the isomorphism that exists between the set of equations (49) and (50) and the set of equations (52) and (53). When $J < 0$ and χ_s is undefined, one does not generally find a transition even in three dimensions, as is evidenced by the face-centered cubic lattice with only nearest-neighbor interactions.(37) In the case of both positive and negative exchange constants J, one finds that the classical limit is asymptotically exact at high temperatures.(6)

In the development that is to follow, we will need the RPA expressions for the Curie and Néel temperatures first obtained for arbitrary spin by Tahir-Kheli and ter Haar.[6] The expression for the Curie temperature in the RPA, which we denote by $\tilde{T}_C$, is easily obtained from (50) by setting $\chi_0 = \infty$, while the expression for the Néel temperature in the RPA, which we denote by $\tilde{T}_N$, is easily obtained from (53) by setting $\chi_s = \infty$. Both results may be expressed as

$$\tilde{T}_C = \tilde{T}_N = \frac{2|J|S(S+1)}{3k_B}\left(\frac{1}{N}\sum_k \frac{1}{\gamma_0 - \gamma_k}\right)^{-1} \tag{54}$$

It should be recalled that we introduced a parameter c in (37) which may differ from unity if necessary to ensure the existence of a solution for f and g. One can show that in the classical limit, c has the unique solution of unity and hence (37) is redundant. The situation is quite different in the quantum case, which we now consider.

5. QUANTUM RESULTS

We consider the thermodynamics predicted by our Green's function theory in the quantum case, which is defined as the solution to (40), (44)–(46), and (48). The solutions to these equations have the same qualitative features as the solution in the classical limit. In particular, one finds that χ_0 and χ_s are decreasing functions of temperature with $\chi_0 = \chi_s = 0$ at infinite temperature and with $\chi_0 = \infty$ at the Curie temperature [which implies that $f = g$ due to (42)] and with $\chi_s = \infty$ at the Néel temperature [which implies that $f = -g$ due to (42)]. One now finds from (44) that in the quantum limit, the Curie temperature, which we denote by T_C, and the Néel temperature, which we denote by T_N, are given by

$$T_{\substack{C\\N}} = [2|J|S(S+1)/3k_B]\left\{(1/N)\sum_k [V(k, T_{\substack{C\\N}})/(\gamma_0 \mp \gamma_k)]\right\}^{-1} \tag{55}$$

where one choses the minus sign for T_C and the plus sign for T_N. It no longer follows that the Curie and Néel temperatures are equal. The reason for this is that excitation energies appearing in the definition of $V(k, T)$ given by (46) are quite different. In fact, one has from (40) that

$$E_{k(T_C)} = 4Jf^{1/2}(\gamma_0 - \gamma_k), \qquad E_{k(T_N)} = 4|J|f^{1/2}(\gamma_0^2 - \gamma_k^2)^{1/2} \tag{56}$$

Notice that at T_C, the excitations have the same wavelength dependence as low-temperature ferromagnetic magnons, while at T_N, the excitations have the same wavelength dependence as low-temperature antiferromagnetic spin waves. The facts that the staggered susceptibility becomes infinite and the excitation energies have a higher periodicity in k-space consistent with a sublattice picture stongly suggest the appearance of antiferromagnetism at lower temperatures, although we are unable to explicitly demonstrate this since the assumption of a spherically symmetric phase is no longer valid below T_N. Likewise, we can only suggest the existence of the ferromagnetic phase below T_C.

Rushbrooke and Wood[38] have used a $1/T$ series expansion for the susceptibility and staggered susceptibility to determine estimates for the Curie and Néel temperatures. They find

$$(T_N^{RW} - T_C^{RW})/T_C^{RW} \simeq 0.63/\gamma_0 S(S+1) \tag{57}$$

where T_N^{RW} and T_C^{RW} are their estimates for the Néel and Curie temperatures, respectively. One should therefore expect a different expression for the Curie and Néel temperatures, as is evidenced by (55).

It will now be shown that our Green's function theory in the quantum limit is consistent with the theorem of Mermin and Wagner,[31] which states that a phase transition to the ferromagnetic state or antiferromagnetic state at finite temperature is impossible in either one or two dimensions. To do this, we note that $V(k, T)$ as defined in (46) satisfies the inequality $V(k, T) \geqslant 1$ for all values of k and T for both positive and negative exchange constant J. This inequality, when used in (55), leads to the results

$$T_C < \tilde{T}_C, \qquad T_N < \tilde{T}_N \tag{58}$$

which in view of the known values of $\tilde{T}_N$ and $\tilde{T}_C$, implies consistency with the theorem of Mermin and Wagner.

We now estimate the change in the thermal properties arising due to the quantum effects which were neglected in the classical limit presented in Section 4. The $V(k, T)$ appearing in (46) may be expanded about $\beta \to 0$. If we substitute this expansion into (44) and (45), we obtain

$$2S(S+1)\beta J = 3I(h) + 2\beta^2 J^2 \gamma_0 f + \cdots \tag{59}$$

and

$$2\beta J \gamma_0 f = [\gamma_0 I(h) h^{-1} - 1] - 2\beta^2 J^2 \gamma_0 f/3 + \cdots \tag{60}$$

where $I(h)$ is the lattice sum,

$$I(h) = (1/N) \sum_k [h/(\gamma_0 - h\gamma_k)] \tag{61}$$

and

$$h = f/g \tag{62}$$

A comparison of (44) and (45) with (24) shows that we have retained terms to order $\langle \omega_k{}^2 \rangle_{zz}$ in the expansion given in (59) and (60). It should be recalled that the exact zero, first, and second moments of the relaxation function $R_{zz}(k, t)$ have been used as boundary conditions on our Green's function at $t = 0$, as is reflected in (16) and (19). Higher moments have not been included in (59) and (60) since these are not correctly given by the relaxation function as given in (21), which contains only two frequencies. The contributions from these higher moments to the thermodynamics is small except possibly for $S = \frac{1}{2}$ systems with a small number of near neighbors for temperatures near the critical temperature in the three-dimensional case. We may rearrange (60) to obtain

$$2\beta J\gamma_0 f = [\gamma_0 I(h)h^{-1} - 1](1 + \tfrac{1}{3}\beta J)^{-1} \tag{63}$$

Furthermore, we obtain

$$2S(S+1)\beta J = 3I(h)/\{1 - [2S(S+1)]^{-1}[\gamma_0 I(h)h^{-1} - 1](1 + \tfrac{1}{3}\beta J)^{-1}\} \tag{64}$$

upon the substitution of (63) into (59). In order to motivate one further approximation, we write the term $(1 + \frac{1}{3}\beta J)$ which appears in both (63) and (64) as

$$(1 + \tfrac{1}{3}\beta J) = \{1 + [\tilde{T}_C I(1)/6S(S+1)T]\} \tag{65}$$

with the aid of (54) and (61). Equation (65) indicates that (59) and (60), when expressed as a series in $\tilde{T}_C/T$, will also be a series in inverse powers of $S(S+1)$. The same results apply to (63) and (64), which are derived from (59) and (60). If we now compare (63) and (64) with the aid of (65), we see that the denominator of (64) contains higher terms in inverse powers of $S(S+1)$ than does (63). We therefore make the furher approximation of replacing (64) by

$$2S(S+1)\beta J = 3I(h)/\{1 - [2S(S+1)]^{-1}[\gamma_0 I(h)h^{-1} - 1]\} \tag{66}$$

so that (66) reflects the same level of approximation as does (63) in inverse powers of $S(S+1)$.

In the remainder of this paper, (63) and (66) will be used to describe the high-temperature region of the Heisenberg paramagnet. The high-temperature region is defined as the entire paramagnetic region for three-dimensional paramagnets which undergo either a Curie or a Néel transition. For one- and two-dimensional paramagnets as well as those three-dimensional paramagnets that do not undergo a Néel transition when J is negative, we define the high-temperature region to be the temperature range above the Curie–Weiss temperature defined by

$$\Theta_{\mathrm{CW}} = 2|J|\gamma_0 S(S+1)/3k_{\mathrm{B}} \tag{67}$$

A comparison with experiment would require use of analytical expressions for the Watson sums $I(h)$,(33,34,39–44) to obtain the self-consistency parameters f and h appearing in (63) and (66). However, we are primarily interested in making a comparison with exactly known properties of the Heisenberg model to test the validity of the statistical approximations used in this paper. The bulk of the theoretical knowledge related to the Heisenberg paramagnet is contained in the $1/T$ series expansion results and results deduced from these series by Padé approximations.(35,36,38,45–50) In the next section, we shall express the solutions for the specific heat and susceptibility deduced from (63), (65), and (42) as a series expansion in powers of $1/T$ which may be compared with the exact results.

Before proceeding to the $1/T$ series, we examine the predicted Curie and Néel temperatures as well as values of the nearest-neighbor correlation functions at the critical temperature. We will show that the unphysical results obtained by Liu in the RPA for the $S = \frac{1}{2}$ simple cubic paramagnet in which the energy predicted at the critical temperature was lower than the ground-state energy disappears when the quantum effects present in (63) and (65) are taken into account.

Using the fact that

$$I(h) = -I(-h) \tag{68}$$

for lattices that are decomposable into two interpenetrating sublattices with the property that nearest-neighbor sites always lie on separate sublattices, one finds from (66) with $|h| = 1$ that the Curie and Néel temperature are given by

$$T_C = T_N = \tilde{T}_C\left(1 - \frac{\gamma_0 I(1) - 1}{2S(S+1)}\right) \tag{69}$$

so that the Curie and Néel temperatures are in agreement to order $1/S(S+1)$. Equation (69) is in disagreement with the predictions of Rushbrooke and Wood given in (57). One needs the following values of $I(1)$

$$I(1) = \begin{cases} 0.2527 & (\text{sc}) \\ 0.1742 & (\text{bcc}) \\ 0.1122 & (\text{fcc}) \end{cases} \tag{70}$$

for the simple cubic, body-centered cubic, and face-centered cubic lattices in order to compare our predictions for the Curie and Néel temperatures with the predictions of Rushbrooke and Wood,(36,38) which are given by (57) and

$$T_C^{\text{RW}} = 5J(\gamma_0 - 1)[11S(S+1) - 1]/96k_B \tag{71}$$

Table I. Predictions for the Curie and Néel Temperatures[a]

Lattice method	Curie $S = \frac{1}{2}$	Néel $S = \frac{1}{2}$	Curie $S = 1$	Néel $S = 1$	Curie $S = 5/2$	Néel $S = 5/2$
Simple-cubic						
$1/T$ Series(36,38)	0.629	0.717	0.683	0.719	0.709	0.718
RPA(6)	0.660	0.660	0.660	0.660	0.660	0.660
Present theory	0.433	0.433	0.575	0.575	0.641	0.641
Body-centered cubic						
$1/T$ Series(36,38)	0.661	0.730	0.735	0.764	0.744	0.750
RPA(6)	0.718	0.718	0.718	0.718	0.718	0.718
Present theory	0.530	0.530	0.647	0.647	0.702	0.702
Face centered cubic						
$1/T$ Series(36,38)	0.692	None	0.752	None	0.780	None
$1/T$ Series(35)	0.679	None	0.747	None	—	None
RPA(6)	0.743	None	0.743	None	0.743	None
Present theory	0.571	None	0.679	None	0.728	None

[a] Tabulated temperatures are multiples of the Curie–Weiss temperature, $\theta_{\mathrm{CW}} = 2\gamma_0 |J| S(S+1)/3k_{\mathrm{B}}$.

Table I gives a comparison of our predictions for the Curie and Néel temperature with both RPA and $1/T$ series predictions. Our predictions are uniformly lower than the $1/T$ series estimates.

We now consider predictions for the nearest-neighbor spin–spin correlation function predicted by our Green's function theory at the critical temperature. We obtain

$$\langle \mathbf{S}_0 \cdot \mathbf{S}_d \rangle_{\substack{\mathrm{C}\\ \mathrm{N}}}^{\mathrm{GF}} = \pm 3k_{\mathrm{B}}T_{\mathrm{C}}[\gamma_0 J(1) - 1)]/2|J|\gamma_0(1 \pm |J|/3k_{\mathrm{B}}T_{\mathrm{C}}) \tag{72}$$

upon the substitution of (69) into (63) with the use of (68). These values are considerably lower than the RPA values of Liu, which are given by

$$\langle \mathbf{S}_0 \cdot \mathbf{S}_d \rangle_{\mathrm{C}}^{\mathrm{RPAL}} = -\langle \mathbf{S}_0 \cdot \mathbf{S}_d \rangle_{\mathrm{N}}^{\mathrm{RPAL}} = S(S+1)\{1 - [1/\gamma_0 J(1)]\} \tag{73}$$

Our values of $\langle S_0 \cdot S_d \rangle_{\mathrm{C}}$ are much closer to the $1/T$ series values of Domb and Sykes(35) than the RPA values of Liu, as may be seen by an examination of Table II.

We find that the power laws in the critical region are the same as for the RPA theory and the spherical model although the numerical values of the coefficients change somewhat, as is evidenced by the change in the critical temperature and critical value of the nearest-neighbor correlation function calculated above. Since the critical behavior of the spherical model is known to be incorrect, we pursue the matter no further.

Table II. Critical Values of the Normalized Nearest-Neighbor Correlation Function $\langle \mathbf{S}_0 \cdot \mathbf{S}_d \rangle / S^2$

Lattice method	Curie $S = \frac{1}{2}$	Néel $S = \frac{1}{2}$	Curie $S = 1$	Néel $S = 1$	Curie $S = \infty$	Néel $S = \infty$
Simple cubic						
RPA (Liu)⁽⁵⁾	1.02	−1.02	0.68	−0.68	0.34	−0.34
Present theory	0.53	−0.90	0.55	−0.64	0.34	−0.34
Body-centered cubic						
RPA (Liu)⁽⁵⁾	0.84	−0.84	0.56	−0.56	0.28	−0.28
Present theory	0.54	−0.74	0.48	−0.54	0.28	−0.28
Face-centered cubic						
RPA (Liu)⁽⁵⁾	0.77	None	0.51	None	0.26	None
Present theory	0.54	None	0.46	None	0.26	None
$1/T$ Series⁽⁸⁵⁾	0.45	None	0.34	None	0.19	None

6. 1/*T* SERIES RESULTS

It is useful to introduce the notation

$$\tilde{J} = 2JS^2 \tag{74}$$

$$K = \tilde{J}/k_{\mathrm{B}}T \tag{75}$$

and

$$X = S(S+1) \tag{76}$$

so that the Hamiltonian given by (7) now becomes

$$H = -(\tilde{J}/2S^2) \sum_{j,d} \mathbf{S}_j \cdot \mathbf{S}_{j+d} \tag{77}$$

It is easier to assess the quantum effects as a function of S with the last form of the Hamiltonian, for which the ferromagnetic ground state is independent of S for fixed $\tilde{J}$ and for which the Curie temperature depends only weakly on S for a fixed value of $\tilde{J}$. Equations (63) and (66) become

$$f = S^2[\gamma_0 I(h)h^{-1} - 1]/K^2[1 + (\nu K/6S^2)] \tag{78}$$

and

$$K = 3S^2X^{-1}I(h)/\{1 + \tfrac{1}{2}\epsilon[\gamma_0 I(h)h^{-1} - 1)/X]\} \tag{79}$$

when use is made of (74)–(76). We have introduced the variable ϵ, which is equal to unity. The reason we introduced ϵ is that if one sets $\epsilon = 0$, one recovers the $1/T$ series solution for the classical limit discussed in Section 4.

We are therefore able to get $1/T$ series expressions for both the spherical model and the RPA model of Liu as well as the $1/T$ series expressions for the quantum limit of our model.

Notice that (79) gives h as a function of spin and temperature. So long as $|h| < 1$, we can expand (61) to obtain

$$I(h) = \gamma_0 h^{-1} \sum_{n=0}^{\infty} W_n h^n \tag{80}$$

where

$$W_n = N^{-1} \sum_{k} (\gamma_k/\gamma_0)^n \tag{81}$$

The lattice sums given in (81) may be interpreted with the aid of (20) as the probability of returning to the starting point after n random steps between nearest-neighbor spin sites. These values are easily obtained on a computer and are given in Table III for several lattices.

Combining (79)-(81) and inverting the series, we obtain

$$h = \sum_{n=1}^{\infty} A_n (K/S^2)^n \tag{82}$$

where the first four coefficients are given by

$$\begin{gathered} A_1 = X\gamma_0/3, \qquad A_2 = 0, \qquad A_3 = -X^2\gamma_0^2(2X + v)/54, \\ A_4 = X^3\gamma_0 V_3(2X + v)/162 \end{gathered} \tag{83}$$

where V_n is defined by

$$V_n = \gamma_0^n W_n \tag{84}$$

We can now obtain an explicit expression for f as a function of temperature by substituting (82) and (80) into (78). The resulting expression

Table III. Probabilities W_n of Returning to Starting Point After n Steps Between Nearest-Neighbor Lattice Sites

Lattice	W_0	W_2	W_3	W_4
Linear	1	$\frac{1}{2}$	0	3/8
Square	1	$\frac{1}{4}$	0	9/64
Simple cubic	1	1/6	0	5/72
Face-centered cubic	1	1/12	1/36	5/192
Body-centered cubic	1	1/8	0	27/512

is readily reduced to a $1/T$ series in temperature. We write our prediction for the normalized nearest-neighbor correlation function as

$$\langle \mathbf{S}_0 \cdot \mathbf{S}_d \rangle^{\mathrm{GF}}/S^2 = -\sum_{n=2}^{\infty} B_n^{\mathrm{GF}}(v, S)\, S^{-2n} K^{n-1} \tag{85}$$

where one obtains our prediction for the classical Heisenberg model when $v = 0$ as well as the series expansion for the RPA theory of Liu$^{(5)}$ and the spherical model of Lax,$^{(32)}$ and where one obtains our prediction for the quantum Heisenberg model when $v = 1$. The coefficients $B_n^{\mathrm{GF}}(v, S)$ are given in Appendix A, where they are compared with the exact results$^{(36)}$ $B_n^{E}(v, S)$.

Upon substituting (62) and (72) into (42) and setting $k = 0$, we obtain

$$\chi_0 = S^2 h / J\gamma_0(1 - h) \tag{86}$$

for the susceptibility. Finally, we obtain after substituting (82) into (86), an expression for the susceptibility as a function of inverse temperature. We denote our prediction for the series as

$$3k_{\mathrm{B}} T \chi_0^{\mathrm{GF}}/S(S + 1) = \sum_{n=0}^{\infty} D_n^{\mathrm{GF}}(v, S)\, S^{-2n} K^n \tag{87}$$

where the coefficients $D_n^{\mathrm{GF}}(v, S)$ are given in Appendix B and are compared with the exact coefficients obtained by Brown and Luttinger.$^{(51)}$ Again, one obtains predictions for the classical Heisenberg model when $v = 0$ and predictions for the quantum Heisenberg model when $v = 1$. Equation (87) also gives the exact $1/T$ series for the RPA theory when $v = 0$ as well as the exact $1/T$ series for the spherical model of Lax$^{(32)}$ due to the relations established in Section 4.

Notice in Appendices A and B that our $1/T$ series coefficients for the classical and quantum Heisenberg models are related in precisely the same way as the exact $1/T$ series for the classical and quantum Heisenberg models. The coefficients with subscript n are polynomials in the variable X of degree n in the quantum case, whereas the coefficients in the classical case contain only the term in the polynomial containing the highest power of the variable X. We use this rigorous relation to establish that by setting $V(k, T)$ in (46) equal to unity, we do indeed obtain the classical limit of our theory which was tentatively assumed in Section 4.

We have given arguments for the necessity for a second-order truncation in the paramagnetic region of a Heisenberg magnet in the absence of a magnetic field. This truncation, which incorporates the exact values for the zero, first, and second moments of the relaxation function as boundary conditions, yields results which satisfy the rotational symmetry of the paramagnetic region and the principle of detailed balance. The classical limit of our theory yields results for the equal time spin-spin correlations in the field-free paramagnetic region which are identical to the results of both the spherical model and the RPA theory of Liu. Quantum corrections to both the Curie and Néel temperatures as well as critical values of the nearest-neighbor correlation function are found. Our expressions for the susceptibility and internal energy are expanded in powers of the inverse temperature and the results are compared with the exact series.

APPENDIX A

The nearest-neighbor spin-spin correlation coefficients $B_n^{\mathrm{GF}}(v, S)$ defined by (85) and discussed in Section 6 are compared below with the exact coefficients, denoted by $B_n{}^E(v, S)$, which were deduced from the coefficients of Rushbrooke and Wood.[36]

Linear:

$$B_2{}^E = X^2/3, \qquad B_3{}^E = 0X^3 - vX^2/12,$$
$$B_4{}^E = X^2(-3X^2 - 8vX + 3v)/135$$

$$B_2^{\mathrm{GF}} = X^2/3, \qquad B_3^{\mathrm{GF}} = 0X^3 - vX^2/18,$$
$$B_4^{\mathrm{GF}} = X^2(-5X^2 - 10vX + 1.25v)/135$$

Square:

$$B_2{}^E = X^2/3, \qquad B_3{}^E = 0X^3 - vX^2/12,$$
$$B_4{}^E = X^2(7X^2 - 18vX - 3v)/135$$

$$B_2^{\mathrm{GF}} = X^2/3, \qquad B_3^{\mathrm{GF}} = 0X^3 - vX^2/18,$$
$$B_4^{\mathrm{GF}} = X^2(5X^2 - 20vX + 1.25v)/135$$

Simple cubic:

$$B_2{}^E = X^2/3, \qquad B_3{}^E = 0X^3 - vX^2/12,$$
$$B_4{}^E = X^2(17X^2 - 28vX - 3v)/135$$

$$B_2^{\mathrm{GF}} = X^2/3, \qquad B_3^{\mathrm{GF}} = 0X^3 - vX^2/18,$$
$$B_4^{\mathrm{GF}} = X^2(15X^2 - 30vX + 1.25v)/135$$

Body-centered cubic:

$$B_2{}^E = X^2/3, \qquad B_3{}^F = 0X^3 - vX^2/12,$$

$$B_4{}^E = X^2(57X^2 - 38vX - 3v)/135$$

$$B_2^{\mathrm{GF}} = X^2/3, \qquad B_3^{\mathrm{GF}} = 0X^3 - vX^2/18,$$

$$B_4^{\mathrm{GF}} = X^2(55X^2 - 40vX + 1.25v)/135$$

Face-centered cubic:

$$B_2{}^E = X^2/3, \qquad B_3{}^F = (4X^3/9) - (vX^2/12),$$

$$B_4{}^E = X^2(107X^2 - 78vX + 3v)/135$$

$$B_2^{\mathrm{GF}} = X^2/3, \qquad B_3^{\mathrm{GF}} = (4X^3/9) - (vX^2/18),$$

$$B_4^{\mathrm{GF}} = X^2(105X^2 - 70vX + 1.25v)/135$$

APPENDIX B

The susceptibility coefficients $D_n^{\mathrm{GF}}(v, S)$ defined by (87) and discussed in Section 6 are compared with the exact coefficients, denoted by $D_n{}^E(v, S)$, which we deduced from the coefficients of Brown and Luttinger.[51]

Linear:

$$D_0{}^E = 1, \qquad D_1{}^E = 2X/3, \qquad D_2{}^E = (2X^2/9) - (vX/6)$$

$$D_0^{\mathrm{GF}} = 1, \qquad D_1^{\mathrm{GF}} = 2X/3, \qquad D_2^{\mathrm{GF}} = (2X^2/9) - (vX/9)$$

Square:

$$D_0{}^E = 1, \qquad D_1{}^E = 4X/3, \qquad D_2{}^E = (4X^2/3) - (vX/3)$$

$$D_0^{\mathrm{GF}} = 1, \qquad D_1^{\mathrm{GF}} = 4X/3, \qquad D_2^{\mathrm{GF}} = (4X^2/3) - (2vX/9)$$

Simple cubic:

$$D_0{}^E = 1, \qquad D_1{}^E = 2X, \qquad D_2{}^E = (10X^2/3) - (vX/2)$$

$$D_0^{\mathrm{GF}} = 1, \qquad D_1^{\mathrm{GF}} = 2X, \qquad D_2^{\mathrm{GF}} = (10X^2/3) - (vX/3)$$

Body-centered cubic:

$$D_0{}^E = 1, \qquad D_1{}^E = 8X/3, \qquad D_2{}^E = (56X^2/9) - (2vX/3)$$

$$D_0^{\mathrm{GF}} = 1, \qquad D_1^{\mathrm{GF}} = 8X/3, \qquad D_2^{\mathrm{GF}} = (56X^2/9) - (4vX/9)$$

Face-centered cubic

$$D_0^E = 1, \qquad D_1^E = 4X, \qquad D_2^E = (44X^2/3) - rX$$

$$D_0^{\mathrm{GF}} = 1, \qquad D_1^{\mathrm{GF}} = 4X, \qquad D_2^{\mathrm{GF}} = (44X^2/3) - (2rX/3)$$

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