Green Function Theory of Ferromagnetism

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ABSTRACT

A theory of ferromagnetism for general spin, approximately valid through the entire temperature range, is given. At low temperatures the magnetization agrees with the Dyson results, having no term in T^3 and having a term in T^4 equal to that found by Dyson in first Born approximation; terms arising from the approximations of the theory first appear in order $T^{3(2S + 1)/2}$, so that a spurious T^3 term does appear for $S = 1/2$, but for no other spin. Curie temperatures are within a few percent of the Brown and Luttinger estimates for spins greater than unity, and agree within one percent of the Domb and Sykes estimate of the rge spin limit. The susceptibility at high temperatures agrees with the Opechowski expansion to terms in $1/T^2$. The quasi-particle energies are renormalized by the energy at low temperature and by the magnetization at higher temperature. The Green function is decoupled by a physical criterion involving self-consistency of the decoupling at all temperatures. The Green function method is extended to higher spin by a technique of parametrizing the Green function and explicitely finding the functional dependence on this parameter by solution of an auxiliary differential equation.

Introduction

The Heisenberg model of a ferromagnet has been theoretically analyzed by Dyson[1] by series expansion in powers of T, valid at low temperatures, and by Opechowski[2] by series expansion in $\frac{1}{T}$, valid at very high temperatures. We here develop an approximate theory which covers the entire temperature range, including the particularly interesting intermediate region in the neighborhood of the Curie temperature, and which agrees satisfactorily with the rigorous results at both very low and very high temperatures.

At low temperatures the magnetization has terms of order $T^{3/2}$, $T^{5/2}$, $T^{7/2}$ which agree with the Dyson results, it properly has no term in T^3, and the term in T^4 is equal to that found by Dyson in first Born approximation. Terms arising from the approximation in the theory first arise in order $T^{3(2S + 1)/2}$, so that the case of $S = 1/2$ is an exception to the above statements, having a spurious T^3 term. Curie temperatures are quite close ($\simeq 3\%$ for spin 2) to the values estimated by Brown and Luttinger[3] by extrapolation of the high temperatures series expansion, except for very small values of spin (again the case of spin 1/2 is particularly unsatisfactory). The Curie temperatures agree within 1% with the estimate of Domb and Sykes[4] for the high spin limit. The susceptibility at high temperatures for all spin values agrees with the Opechowski[2]

expansion to terms in $1/T^2$.

The quasi-particle energies are equivalent to simple spin wave energies "renormalized" by a factor which is proportional to the thermodynamic energy at low temperatures, but which becomes proportional to the magnetization at higher temperatures.

An heuristic interpretation of the renormalization of quasi-particle energies has been given by Keffer and Loudon[5]. They point out that, at low temperatures where only long wave length spin waves are excited, the local magnetization direction varies slowly through the crystal. Excitation of an additional spin wave is analogous to excitation of a ripple relative to this slowly varying local magnetization. The effective exchange integral determining the energy of this ripple is influenced by the angle between neighboring spins in the slowly varying background medium. This angle also determines the thermodynamic energy; hence the renormalization of the spin wave energy by the thermodynamic energy. However at higher temperatures the thermally excited excitations have wave lengths comparable to the inter-spin distance, and the correlation distance in the background medium is as short, or shorter, than the wave length of the particular excitation being considered. In this region our results indicate that the effective exchange integral is influenced by the angle

relative to the average magnetization; that is, the spin wave

energies are renormalized by the average magnetization.

The Heisenberg ferromagnet with spin 1/2 was analyzed by

Tyablikov[6] using the technique of double-time temperature-

dependent Green functions[7,8]. Extension of the theory to

higher spin has been achieved recently by Tahir-Kheli and

ter Haar[9]. The present theory differs from those applica-

tions of the Green function method in two respects. First,

the decoupling of the higher order Green functions is guided

by a plausible physical criterion, Second, the method of employing

Green functions for general spin is simplified; the Green

function is parametrized and the functional dependence on

this parameter is found explicitely by solution of an

auxiliary differential equation. This single differential

equation replaces the 2S simultaneous Green function

equations in the method of Tahir-Kheli and ter Haar[9].

2. The Green Function

The problem to be considered is the statistical
mechanics of the system described by the Hamiltonian

$$\mathcal{H} = -\mu H \sum_{g} S_{g}^{z} - \sum_{g,f} J(g-f)\, \vec{S}_{g} \cdot \vec{S}_{f} \qquad (1)$$

where μS is the magnetic moment per ion; H is the applied
magnetic field (in the positive z-direction); S_{g} is the
spin operator (in units of $\hbar$) for the ion at site g, and
$J(g-f)$ is the exchange integral between ions at sites g and f.
The exchange integral is assumed to be a function only of the
distance between ions; it is not restricted to nearest
neighbors or to only positive values, but it is assumed that
the ground state of the system is a ferromagnetically aligned
state.

The temperature-dependent retarded Green function
involving the two operators A and B, $<\!<\!A(t); B\!>\!>$, is defined
by [8]

$$\langle\!\langle A(t); B \rangle\!\rangle \equiv -i\, \theta(t) \langle\, [A(t), B]\, \rangle \qquad (2)$$

where A(t) is the Heisenberg operator at time t; θ(t) is

unity for positive t and zero for negative t; square
brackets denote a commutator; and single angular brackets
denote an average with respect to the canonical density
matrix of the system at temperature T. The Fourier transform
of the Green function is a function of ω (or of $E = \hbar\omega$),
and is denoted by $<<A;B>>_E$. It satisfies the equation of motion[8]

$$E \langle\!\langle A;B \rangle\!\rangle_E = \frac{1}{2\pi} \langle [A, B] \rangle + \langle\!\langle [A(t), H] ; B \rangle\!\rangle_E$$

(3)

If this equation can be solved for $<<A;B>>_E$ one then
extracts knowledge of the correlation function $<BA(t)>$ from
the relation[8]

$$\langle BA(t) \rangle = \lim_{\epsilon \to 0} i \int_{-\infty}^{\infty} \frac{\langle\!\langle A;B \rangle\!\rangle_{\hbar\omega+i\epsilon} - \langle\!\langle A;B \rangle\!\rangle_{\hbar\omega-i\epsilon}}{\exp(\hbar\omega/k_T) - 1} e^{-i\omega t} d\omega$$

(4)

Equations (3) and (4) are the only equations required for
the application of the Green function method.

For reasons which will become evident subsequently we consider the Green function

$$G_t^a (g,\ell) \equiv \langle\langle S_g^+ (t) ; e^{a S_\ell^z} S_\ell^- \rangle\rangle \tag{5}$$

where a is a parameter. The Fourier transform of this Green function, G_E^a (g,1), satisfies the equation of motion (eqn. 3)

$$E G_E^a (g,\ell) = \frac{1}{2\pi} \Theta (a) \delta_{g,\ell} + \langle\langle [S_g^+(t), H]; e^{a S_\ell^z} S_\ell^- \rangle\rangle_E \tag{6}$$

where

$$\Theta (a) \equiv \langle [S^+, e^{a S^z} S^-] \rangle \tag{7}$$

The commutator of S_g^+ with the Hamiltonian, required in the last term of equation (6), is easily computed, giving

$$E G_E^a (g,\ell) = \frac{\Theta(a)}{2\pi} \delta_{g,\ell} + \mu H G_E^a (g,\ell) - \sum_f J(g-f) \langle\langle S_g^z S_f^+ - S_f^z S_g^+ ; e^{a S_\ell^z} S_\ell^- \rangle\rangle_E \tag{8}$$

The remaining problem is to express the higher-order Green function on the right in terms of two-particle Green functions, so that equation (8) can be explicitly solved for G_E^a (g,1).

3. A Decoupling Approximation

The essential approximation in the methods of Tyablikov and of Tahir-Kheli and ter Haar consists of ignoring the fluctuations of S_g^z, replacing this operator by its average value:

$$\langle\langle S_g^z S_f^+ ; B \rangle\rangle \xrightarrow[f \neq g]{} \langle S^z \rangle \langle\langle S_f^+ ; B \rangle\rangle \qquad (Tyablikov) \qquad (9)$$

This approximation results in the magnetization renormalization of quasi-particle energies, in disagreement with the low-temperature theory.

The decoupling approximation to be used here is most clearly described for the special case of spin $\frac{1}{2}$. In that case we can write S_g^z in either of the following forms

$$S_g^z = S - S_g^- S_g^+ \qquad (s = \tfrac{1}{2}) \qquad (10)$$

$$S_g^z = \tfrac{1}{2}(S_g^+ S_g^- - S_g^- S_g^+) \qquad (11)$$

or, multiplying the first of these equations by an arbitrary parameter α and the second by $(1 - \alpha)$ and adding,

$$S_g^z = \alpha S + \frac{1-\alpha}{2} S_g^+ S_g^- - \frac{1+\alpha}{2} S_g^- S_g^+ \qquad (s = \tfrac{1}{2}) \qquad (12)$$

The Green function $\langle\langle S_g^- S_g^+ S_f^+ ; B \rangle\rangle$ is reasonably decoupled in the symmetric form*

$$\langle\langle S_g^- S_g^+ S_f^+ ; B \rangle\rangle \xrightarrow[g \neq f]{} \langle S_g^- S_g^+ \rangle \langle\langle S_f^+ ; B \rangle\rangle + \langle S_g^- S_f^+ \rangle \langle\langle S_g^+ ; B \rangle\rangle \qquad (13)$$

and similarly for the Green function $\langle\langle S_g^+ S_g^- S_f^+ ; B \rangle\rangle$.
Thus the identity (12) leads to

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*The remaining "contraction", $\langle S_g^+ S_f^+ \rangle \langle\langle S_g^- ; B \rangle\rangle$ vanishes because the operator $S_g^+ S_f^+$ is not diagonal in the total z-component of spin.

$$\langle\langle S_g^z S_f^+ ; B \rangle\rangle \xrightarrow[g \neq f]{} \langle S^z \rangle \langle\langle S_f^+ ; B \rangle\rangle - \alpha \langle S_g^- S_f^+ \rangle \langle\langle S_g^+ ; B \rangle\rangle \qquad (14)$$

If α is chosen as unity the result corresponds to decoupling on the basis of identity (10); $\alpha = 0$ corresponds to decoupling on the basis of identity (11); $\alpha = -1$ corresponds to decoupling on the basis of the identity $S_g^z = -S + S_g^+ S_g^-$. Thus we are faced with the possibility of obtaining a correction to the Tyablikov decoupling with either a positive or negative sign, or no correction at all, or any intermediate value, depending on the choice of α. Clearly a physical criterion is required at this point.

The operator $S^- S^+$ in equation (10) represents the deviation of S^z from $+S$. It is this operator $S^- S^+$ which is treated approximately when decoupling on the basis of equation (10). It therefore seems reasonable to use equation (10) when the deviation from $S^z = S$ is small; that is, when $\langle S^z \rangle \simeq S$.

Similarly, the operator $\frac{1}{2}(S^+ S^- - S^- S^+)$ in equation (11) represents the deviation of S^z from zero, and it therefore is reasonable to use equation (11) as the basis of decoupling when $\langle S^z \rangle \simeq 0$.

Both of the above observations are contained in the choice

$$\alpha = \frac{\langle S^z \rangle}{S} \qquad (S = 1/2) \qquad (15)$$

for then equation (12) becomes

$$S_g^z = \langle S^z \rangle + \left[\frac{S - \langle S^z \rangle}{2S} S_g^+ S_g^- - \frac{S + \langle S^z \rangle}{2S} S_g^- S_g^+ \right] \qquad (16)$$

The operator in brackets, which is to be decoupled, represents the

deviation of S^z from $\langle S^z \rangle$ and should be self-consistently small in all temperature regions.

Inserting the above value of α into the decoupling equation (19) gives

$$\langle\langle S_g^z S_f^+ ; B \rangle\rangle \xrightarrow[g \neq f]{} \langle S^z \rangle \langle\langle S_f^+ ; B \rangle\rangle - \frac{\langle S^z \rangle}{S} \langle S_g^- S_f^+ \rangle \langle\langle S_g^+ ; B \rangle\rangle \quad , \quad (s = \frac{1}{2}) \quad (17)$$

This is the basic decoupling approximation for spin $\frac{1}{2}$; we now generalize it for higher spin.

The analogue of equation (10), for general spin, is

$$S_g^z = S(S+1) - (S_g^z)^2 - S_g^- S_g^+ \quad (18)$$

whereas equation (11) remains true. Decoupling as before, and neglecting* the fluctuations of $(S^z)^2$, we find in this case as well

$$\langle\langle S_g^z S_f^+ ; B \rangle\rangle \xrightarrow[g \neq f]{} \langle S^z \rangle \langle\langle S_f^+ ; B \rangle\rangle - \alpha \langle S_g^- S_f^+ \rangle \langle\langle S_g^+ ; B \rangle\rangle \quad (19)$$

where α is the fractional contribution of the identity (18), and $(1-\alpha)$ is the contribution of the identity (11), to this result. Unfortunately $S_g^- S_g^+$ is no longer the only operator treated approximately in decoupling equation (18), and the interpretation of the decoupled operator as being the deviation from $S^z = + S$ is no longer true. Hence the choice of α is no longer quite so evident. However,

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*The Green function $\langle\langle (S_g^z)^2 S_f^+ ; B \rangle\rangle$ also can be symmetrically decoupled, but the results are of the same form as those obtained by the simpler procedure above.

we determine it by the following requirements:

a) For $S = \frac{1}{2}$, α should reduce to our previous result, or to $\alpha = \langle S^z \rangle / S$.

b) For $\langle S^z \rangle = 0$, α should vanish. This requirements follows from the fact that identity (11) retains its interpretation for arbitrary S.

c) For $\langle S^z \rangle \simeq S$ we expect that S^z should have the form $S^z \simeq S - n$, where n is a deviation which is of order unity rather than of order S.

Requirement c) implies that $\alpha \langle S_g^- S_f^+ \rangle$ should be of order unity, rather than of order S, at low temperatures. Now $\frac{1}{2S} \langle S_g^- S_g^+ \rangle$ is the spin deviation in lowest order *; similarly $\langle S_g^- S_f^+ \rangle$ will be of order 2S if f and g are closely coupled. Hence we take

$$\alpha = \frac{1}{2S} \frac{\langle S^z \rangle}{S} \qquad (20)$$

and it is clear that this satisfies all of the physical requirements above. Equations (19) and (20) characterize the basic decoupling approximation of the theory.

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* The appearance of 2S here corresponds to the identification of S^+ and S^- respectively as $\sqrt{2S}\, a^+$ and $\sqrt{2S}\, a^-$ in the leading terms both of the Dyson and the Holstein-Primakoff transformations. Here a^+ and a^- are destruction and creation operators of elementary boson-type excitations,

4. Solution of the Green Function Equation

Inserting the decoupling approximation into the equation of motion (8) gives

$$E G_E^a(g,\ell) = \frac{\Theta(a)}{2\pi} + \mu H G_E^a(g,\ell) - 2\langle s^z \rangle \sum_f J(g-f)[G_E^a(f,\ell) - G_E^a(g,\ell)] +$$

$$+ \frac{\langle s^z \rangle}{S} \sum_f J(g-f)[\langle S_g^- S_f^+ \rangle G_E^a(g,\ell) - \langle S_f^- S_g^+ \rangle G_E^a(f,\ell)] \qquad (21)$$

These equations are a set of coupled equations for various pairs of sites (g, l), (f, l). Translational invariance dictates consideration of the spatial Fourier transforms

$$G_E^a(\vec{k}) \equiv \sum_{g-\ell} e^{-i(\vec{g}-\vec{\ell})\cdot\vec{k}} G_E^a(g,\ell) \qquad (22)$$

$$J(\vec{k}) \equiv \sum_{g-\ell} e^{-i(\vec{g}-\vec{\ell})\cdot\vec{k}} J(g-\ell) \qquad (23)$$

$$\Psi(\vec{k},a) \equiv \sum_{g-\ell} e^{-i(\vec{g}-\vec{\ell})\cdot\vec{k}} \langle e^{a S_f^z} S_f^- S_g^+ \rangle \qquad (24)$$

Here $\vec{g}\cdot\vec{k}$ denotes the vector product $\vec{R}_g\cdot\vec{k}$, where $\vec{R}_g$ is the position vector of the g-site. Equation (21) then implies

$$E G_E^a(\vec{k}) = \frac{\Theta(a)}{2\pi} + \mu H G_E^a(\vec{k}) - 2\langle s^z \rangle [J(\vec{k}) - J(0)] G_E^a(\vec{k}) +$$

$$+ \frac{\langle s^z \rangle}{S} \sum_{k'} [J(\vec{k}') - J(\vec{k}'-k)] \Psi(\vec{k}',0) G_E^a(\vec{k}) \qquad (25)$$

or

$$G_E^a(\vec{k}) = \frac{\Theta(a)}{2\pi(E - E(\vec{k}))} \qquad (26)$$

where

$$E(\vec{k}) = \mu H + 2\langle S^z \rangle [J(o) - J(\vec{k})] + \frac{\langle S^z \rangle}{NS^2} \sum_{\vec{k'}} [J(\vec{k'}) - J(\vec{k'} - \vec{k})] \Psi(\vec{k'}, o) \quad (27)$$

The correlation function which can be obtained from $G_E^a(\vec{k})$ by equation (4) is the quantity $\Psi(\vec{k}, a)$, defined in equation (24). Inserting equation (26) in (4), and taking $t = o$, we thereby obtain

$$\Psi(\vec{k}, a) = \frac{\Theta(a)}{e^{E(\vec{k})/kT} - 1} \quad (28)$$

Equation (28), together with eq. (27) defining $E(\vec{k})$, equation (24) defining $\Psi(\vec{k}, a)$, and equation (7) defining $\Theta(a)$, is the basic equation of the theory. It must, however, be augmented by an explicit relationship between Ψ and Θ, and it is this step which complicates the problem for spin $> \frac{1}{2}$.

The problem which distinguishes the simple case of spin $\frac{1}{2}$ from the more complicated case of higher spin becomes evident if we particularize our solution temporarily to $S = \frac{1}{2}$. We also take $a = o$, and we note from eqs.(24) and (10) that

$$\overline{\Psi}(o) \equiv \frac{1}{N} \sum_k \Psi(\vec{k}, o) = \langle S_g^- S_g^+ \rangle = \frac{1}{2} - \langle S^z \rangle \quad , \quad (S = \tfrac{1}{2}) \quad (29)$$

Thus $\overline{\Psi}(o)$ is simply related to the magnetization. $N\overline{\Psi}(o)$ is the total number of spin reversals in the crystal, and $\Psi(\vec{k}, o)$ is the occupation number of the basic excitation of wave vector $\vec{k}$ Similarly, from equation (7)

$$\Theta(o) = 2\langle S^z \rangle \quad (30)$$

← Hence equations (28) and (30) determine $\bar{\psi}(\vec{k},o)$ as a function of $\langle S^z \rangle$, and equation (29) provides a requirement of self-consistency which determines $\langle S^z \rangle$.

For higher spin values equation (29) is replaced by

$$\bar{\psi}(o) \equiv \frac{1}{N}\sum_{k}\psi(\vec{k},o) = \langle S_j^- S_j^+ \rangle = S(S+1) - \langle (S^z)^2 \rangle - \langle S^z \rangle$$

(31)

This introduces the new quantity $\langle (S^z)^2 \rangle$, which is not determined by equation (29). Tahir-Kheli and ter Haar[9] therefore introduce the Green function $\langle\langle S_j^+ ; (S_\ell^-) S_\ell^+ \rangle\rangle$ to evaluate this quantity, but its solution introduces $\langle (S^z)^3 \rangle$. By introducing 2S such Green functions, and by recalling that $(S^z)^{2S+1}$ is related by an identity to lower powers of S^z, they are thereby able to obtain a solution. That method gets laborious very quickly, but they have given explicit solutions to $S = 3$.

By exploiting the functional dependence of $\bar{\theta}(a)$ and $\bar{\psi}$ on the parameter a, which was inserted in equation (5) for just this purpose, both $\bar{\theta}$ and $\bar{\psi}$ can be explicitly related to $\langle S^z \rangle$ for arbitrary spin, in close analogy with the case of spin $\frac{1}{2}$.

5. <u>Relationship of θ to $\langle S^Z \rangle$</u>

Calculation of the commutator defining $\theta(a)$ (eq. (7)) is
facilitated by the identity

$$\left[S^+, (S^z)^n \right] = \left\{ (S^z-1)^n - (S^z)^n \right\} S^+ \tag{32}$$

which is easily corroborated for n = 1, 2 and extended to higher n
by mathematical induction. It follows that

$$\left[S^+, e^{aS^z} \right] = (e^{-a}-1) e^{aS^z} S^+ \tag{33}$$

and thence

$$\theta(a) = 2\langle e^{aS^z} S^z \rangle + (e^{-a}-1)\langle e^{aS^z} S^+ S^- \rangle \tag{34}$$

Expressing $S^+ S^-$ in terms of S^z by the identity (18),

$$\theta(a) = S(S+1)(e^{-a}-1)\langle e^{aS^z} \rangle + (e^{-a}+1)\langle e^{aS^z} S^z \rangle - (e^{-a}-1)\langle e^{aS^z} (S^z)^2 \rangle \tag{36}$$

Finally, it is convenient to introduce the quantity

$$\Omega(a) \equiv \langle e^{aS^z} \rangle \tag{36}$$

and the notation

$$D \equiv \frac{d}{da} \tag{37}$$

Then the two quantities of interest can be written

$$\theta(a) = S(S+1)(e^{-a}-1)\Omega + (e^{-a}+1)D\Omega - (e^{-a}-1)D^2\Omega \tag{38}$$

and

$$\Psi(a) = \langle e^{aS^z} S^- S^+ \rangle = S(S+1)\Omega - D\Omega - D^2\Omega \tag{39}$$

The relationship (28) between $\Psi(\vec{k},a)$ and $\theta(a)$ can be recast
in a more convenient form as well, by defining

$$\phi(\vec{k}) \equiv \frac{1}{e^{E(\vec{k})/kT}-1} \tag{40}$$

whence

$$2\psi(\vec{k}, a) = \phi(\vec{k}) \, \theta(a)$$

(41)

The self-consistency requirement on the Green function is now contained in the condition

$$\overline{2\psi}(a) = \Phi \, \theta(a)$$

(42)

where

$$\Phi \equiv \frac{1}{N} \sum_k \phi(\vec{k})$$

(43)

This condition should determine $\theta(a)$, and thence $\langle S^z \rangle$, which is just $\frac{1}{2}\theta(o)$, as we see from equation (25). It is more convenient to determine $\Omega(a)$, and thence to find $\theta(a)$ by equation (38). In fact inserting equations (38) and (39) into (42) gives a differential equation for $\Omega(a)$.

$$D^2\Omega + \frac{(1+\Phi)e^a + \Phi}{(1+\Phi)e^a - \Phi} D\Omega - S(S+1)\Omega = 0$$

(44)

This differential equation is the analogue of the set of 2S coupled equations of Tahir-Kheli and ter Haar.

To completely determine the solution we require two boundary conditions. From the definition (36) we observe

$$\Omega(o) = 1$$

(45)

The second condition is provided by the operator identity

$$\prod_{p=-S}^{S} (S^z - p) = 0$$

(46)

Taking the average of this equation it can be written in the form

$$\prod_{p=-S}^{S} (D - p) \, \Omega(a=o) \equiv \alpha_S \, \Omega(o) = 0$$

(47)

In the Appendix it is shown that the solution of the differential equation (44) satisfying the boundary conditions (45) and (47) is

$$\Omega(a) = \frac{\Phi^{2S+1} e^{-Sa} - (1+\Phi)^{2S+1} e^{(S+1)a}}{[\Phi^{2S+1} - (1+\Phi)^{2S+1}][(1+\Phi)e^a - \Phi]} \tag{48}$$

from which S^z or $\Omega(a)$ can be found by differentiation.

6. The Formal Solution

For convenience and clarity we recapitulate the final form of the equations, preparatory to analyzing their low-temperature and high temperature behavior, Curie temperature, etc.

Given a spin magnitude S, a temperature T, a magnetic field H, and an exchange interaction with Fourier components $J(\vec{K})$, the quasi-boson occupation number $\phi(\vec{K})$ is

$$\phi(\vec{K}) = \frac{1}{e^{E(\vec{K})/kT} - 1} \tag{49}$$

where

$$E(\vec{K}) = \mu H + 2\langle S^z \rangle \left\{ [J(0) - J(\vec{K})] + \frac{\langle S^z \rangle}{NS^2} \sum_{K'} [J(\vec{K}') - J(\vec{K}'-\vec{K})]\, \phi(\vec{K}') \right\} \tag{50}$$

This is an implicit equation for $\phi(\vec{K})$, involving the unknown quantity $\langle S^z \rangle$. However $\langle S^z \rangle$ is given in terms of

$$\Phi = \frac{1}{N} \sum_K \phi(\vec{K}) \tag{51}$$

by

$$\langle S^z \rangle = D\Omega(0) = \frac{(S-\Phi)(1+\Phi)^{2S+1} + (S+1+\Phi)\,\Phi^{2S+1}}{(1+\Phi)^{2S+1} - \Phi^{2S+1}} \tag{52}$$

Thus equations (49) - (50) constitute a set of coupled equations which must be solved self-consistently for $\langle S^z \rangle$.

7. Nearest Neighbor Interaction

For simple lattices with nearest neighbor interaction the formalism simplifies markedly. In this case the exchange $J(\vec{k})$ is

$$J(\vec{k}) = J \sum_{\delta} e^{i\vec{k}\cdot\vec{\delta}} \tag{53}$$

where $\vec{\delta}$ goes over the nearest neighbors of a representative ion; we assume all magnetic ions are crystalographically equivalent. Consider the sum

$$\mathcal{H} \equiv \sum_{k'} [J(\vec{k'}) - J(\vec{k'}-\vec{k})] \phi(\vec{k'}) \tag{54}$$

which appears in equation (50) for $E(\vec{k})$. Then

$$\mathcal{H} = J \sum_{\delta} (1 - e^{-i\vec{k}\cdot\vec{\delta}}) \sum_{k'} e^{ik'\cdot\vec{\delta}} \phi(\vec{k'}) \tag{55}$$

The sum over $\vec{k'}$ is clearly independent of $\vec{\delta}$, by symmetry, so that it can be replaced by $\mathfrak{z}^{-1} \sum_{\delta'} \sum_{k'} \exp(i\vec{k'}\cdot\vec{\delta'}) \phi(\vec{k'})$ where $\mathfrak{z}$ is the number of nearest neighbors. We thus obtain

$$\mathcal{H} = J^{-1}(o) [J(o) - J(\vec{k})] \sum_{k'} J(\vec{k'}) \phi(\vec{k'}) \tag{56}$$

where $J(o) = \mathfrak{z} J$ is the k = o Fourier component of the exchange interaction. This equation was first pointed out by Michelene Bloch.[10] The sum $\sum J(\vec{k'}) \phi(\vec{k'})$ is a function only of the temperature, $\langle S^z \rangle$, and H (and, of course, of J and the lattice structure).

$$\frac{1}{N J(o)} \sum_{k} J(\vec{k}) \phi(\vec{k}) = f(T, \langle S^z \rangle, H) \tag{57}$$

and

$$E(\vec{k}) = \mu H + 2\langle S^z \rangle [J(o) - J(\vec{k})] \left[1 + \frac{\langle S^z \rangle}{S^2} f\right] \tag{58}$$

Thus, for simple lattices (including simple cubic, body-centered

cubic, and face-centered cubic lattices in particular) and for

nearest neighbor interaction the simple spin wave energies are re-

normalized by a factor depending only on the temperature (for zero

field), independent of the wave vector $\vec{k}$. Equations (57) and (58),

together with the definition of $\phi(\vec{k})$ in terms of $E(\vec{k})$, constitute

a pair of coupled equations for the renormalization function f. For

these lattices and nearest neighbor interaction the complete solution

would be obtained explicitly by solving eqns. (57) and (58) for f,

thereby obtaining $\phi(\vec{k})$ or Φ in terms of T, $\langle S^z \rangle$ and H. Then eliminating

Φ between this equation and equation (52) would give $\langle S^z \rangle$ as a function

of T and H.

8. The Low Temperature Region

The summation (or integral) over $\vec{\kappa}'$ involved in the calculation of Φ for H = o occurs in simple spin wave theory and has been carried out by Dyson[1] and others by standard series expansion procedures appropriate to low temperatures. Thus, if

$$E(\vec{\kappa}) = 2SR\left[J(0) - J(\vec{\kappa})\right] \tag{59}$$

and if a reduced temperature is defined by

$$\tau \equiv \frac{3 k T}{4\pi_3 J S \nu} \tag{60}$$

then[1]

$$\Phi = \zeta(\tfrac{3}{2})\left(\tfrac{\tau}{R}\right)^{3/2} + \tfrac{3}{4}\pi\nu\,\zeta(\tfrac{5}{2})\left(\tfrac{\tau}{R}\right)^{5/2} + \pi^2\omega\,\nu^2\,\zeta(\tfrac{7}{2})\left(\tfrac{\tau}{R}\right)^{7/2} + \cdots \tag{61}$$

Similarly

$$f = \frac{1}{N_3 J}\sum_{\kappa} J(\vec{\kappa})\,\phi(\vec{\kappa}) = \zeta(\tfrac{3}{2})\left(\tfrac{\tau}{R}\right)^{3/2} - \tfrac{1}{4}\pi\nu\,\zeta(\tfrac{5}{2})\left(\tfrac{\tau}{R}\right)^{5/2} + (\omega-\tfrac{5}{4})\pi^2\nu^2\,\zeta(\tfrac{7}{2})\left(\tfrac{\tau}{R}\right)^{7/2} + \cdots \tag{62}$$

The constants ν and ω depend upon the lattice structure and are defined by

$$\nu = 1 \;,\quad \omega = 33/32 \qquad \text{for simple cubic} \tag{63}$$

$$\nu = \tfrac{3}{4}\cdot 2^{2/3} \;,\quad \omega = 281/288 \quad \text{for body-centered cubic} \tag{64}$$

$$\nu = 2^{1/3} \;,\quad \omega = 15/16 \quad \text{for face-centered cubic} \tag{65}$$

To apply these results to our case we take (compare eqns. (59) and (58))

$$R = \frac{\langle S^2 \rangle}{S}\left[1 + \frac{\langle S^2 \rangle}{S^2} f \right] \tag{66}$$

The remaining equation is eq. (52) for $\langle S^z \rangle$ in terms of Φ; expanding it in powers of Φ (which is small at low temperature), we find

$$\langle S^z \rangle = S - \Phi + (2S+1)\Phi^{2S+1} - (2S+1)^2 \Phi^{2S+2} + O(\Phi^{2S+3}) \tag{67}$$

To disentangle the coupled equations we first substitute equation (66) for R into equation (62), solving for f as a series in τ.

$$f = \zeta(\tfrac{3}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{3/2} - \tfrac{1}{4}\pi\nu\,\zeta(\tfrac{5}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{5/2} + (\omega - \tfrac{5}{4})\pi^2\nu^2\,\zeta(\tfrac{7}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{7/2} + \cdots$$
$$- \tfrac{3}{2}\zeta^2(\tfrac{3}{2})\frac{S}{\langle S^2 \rangle^2}\tau^3 - \tfrac{5}{8}\pi\nu\,\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\frac{S^2}{\langle S^2 \rangle^3}\tau^4 + \cdots \tag{68}$$

Inserting this series into equation (61) for Φ we find

$$\Phi = \zeta(\tfrac{3}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{3/2} + \tfrac{3}{4}\pi\nu\,\zeta(\tfrac{5}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{5/2} + \pi^2\omega\nu^2\,\zeta(\tfrac{7}{2})\left(\frac{S\tau}{\langle S^2 \rangle}\right)^{7/2} + \cdots$$
$$- \tfrac{3}{2}\zeta^2(\tfrac{3}{2})\frac{S}{\langle S^2 \rangle^2}\tau^3 - \tfrac{3}{2}\pi\nu\,\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\frac{S^2}{\langle S^2 \rangle^3}\tau^4 + \cdots \tag{69}$$

Finally, inserting this equation into equation (67) we find

$$\langle S^z \rangle = S - \zeta(\tfrac{3}{2})\tau^{3/2} - \tfrac{3}{4}\pi\nu\,\zeta(\tfrac{5}{2})\tau^{5/2} - \pi^2\omega\nu^2\,\zeta(\tfrac{7}{2})\tau^{7/2} + \cdots$$
$$- \tfrac{3}{25}\pi\nu\,\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^4 \ldots + (2S+1)\,\zeta^{2S+1}(\tfrac{3}{2})\,\tau^{3S+3/2} +$$
$$+ (2S+1)^2\,\tfrac{3\pi\nu}{4}\,\zeta^{2S}(\tfrac{3}{2})\,\zeta(\tfrac{5}{2})\,\tau^{3S+5/2} + \cdots \tag{70}$$

Neglecting, for the moment, the terms in $\tau^{3S + 3/2}$, $\tau^{3S + 5/2}$ we see that the coefficients of $\tau^{3/2}$, $\tau^{5/2}$, $\tau^{7/2}$ are in exact agreement with the Dyson result. The term in τ^3 is absent, as required by Dyson. The term involving τ^4 is given by Dyson[1] as

$$- \frac{3}{2S} \pi \nu \, \zeta(\tfrac{3}{2}) \, \zeta(\tfrac{5}{2}) \, Q \tau^4 \tag{71}$$

where Q, the ratio the ratio of this term to our t^4 term, is

$$Q = 1 + \frac{4}{3} [GS - 1]^{-1} + \frac{\alpha}{3S} \tag{72}$$

with

$$G = 10, 16, 24 \qquad \text{for S.C., B.C.C., F.C.C.} \tag{73}$$

$$\alpha = .52, .39, .34 \quad \text{for S.C., B.C.C., F.C.C.} \tag{74}$$

Thus our τ^4 term corresponds to the leading term (Q = 1) of the Dyson result; that is, to the result which Dyson finds in the first Born approximation.

The terms $\tau^{3S + 3/2}$, $\tau^{3S + 5/2}$,...in equation (70) seem to be spurious results of the Green function approximation. For S = 1/2 they gave incorrect contributions to the τ^3 and τ^4 terms. For spin unity the first spurious contribution appears in the $\tau^{9/2}$ term, and for spin 3/2 it appears first in the τ^6 term; thereafter it moves rapidly to higher order in τ. Consequently the spurious terms are of consequence in the low temperature region only for spin 1/2.

Finally, it is of interest to substitute equation (70) into equations (69), (68), and (66), to obtain Φ, f and the renormalization factor R explicitly as series in τ. We thus find

$$\Phi = S - \zeta(\tfrac{3}{2})\tau^{3/2} - \tfrac{3}{4}\pi\nu\zeta(\tfrac{5}{2})\tau^{5/2} - \pi^2\omega\nu^2\zeta(\tfrac{7}{2})\tau^{7/2} + \cdots + \tag{75}$$
$$- \tfrac{3}{2S}\pi\nu\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^4 + \cdots$$

$$f = \zeta(\tfrac{3}{2})\tau^{3/2} - \tfrac{1}{4}\pi\nu\zeta(\tfrac{5}{2})\tau^{5/2} + (\omega - \tfrac{5}{4})\pi^2\nu^2\zeta(\tfrac{7}{2})\tau^{7/2} + \cdots + \tag{76}$$
$$- \tfrac{1}{8S}\pi\nu\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^4 + \cdots$$

and

$$R = 1 - \tfrac{\pi\nu}{S}\zeta(\tfrac{5}{2})\tau^{5/2} - \tfrac{5}{4S}\pi^2\nu^2\zeta(\tfrac{7}{2})\tau^{7/2} + \cdots + \tag{77}$$
$$- \tfrac{2}{S^2}\zeta(\tfrac{3}{2})\tau^3 - \tfrac{21}{8S^2}\pi\nu\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^4 + \cdots +$$
$$+ \tfrac{2S+1}{S}\zeta^{2S+1}(\tfrac{3}{2})\tau^{3(2S+1)/2} + \tfrac{3(2S+1)^2}{4S}\pi\nu\zeta^{2S}(\tfrac{3}{2})\zeta(\tfrac{5}{2})\tau^{3S+5/2}$$

It will be recalled that R is the "renormalization factor"; the ratio of the actual quasi-particle energies to the simple spin wave energies. At low temperatures the leading temperature dependence of R arises in the $\tau^{5/2}$ term. Hence the spin wave energies are renormalized by a factor proportional to the thermodynamic energy rather than the magnetization in this temperature region.

9. The Curie Temperatures, and the High Temperature Between

Just below the Curie temperature (assuming H = o) the average magnetization $\langle S^z \rangle$ is small, and the mean number of excited quasi-particles is large. In fact, expanding equation (52) in powers of Φ^{-1} we find

$$\langle S^z \rangle = \frac{S(S+1)}{3} \Phi^{-1} + O(\Phi^{-2})$$

(78)

Furthermore, since $E(\vec{k})$ is proportional to $\langle S^z \rangle$ the exponential in the Bose distribution can be expanded, giving

$$\Phi = \frac{1}{N} \sum_k \left\{ \frac{2 \langle S^z \rangle}{kT} [J(0) - J(\vec{k})] [1 + \frac{\langle S^z \rangle}{S^2} f] \right\}^{-1}$$

(79)

Multiplying equation (79) by $\langle S^z \rangle$, and replacing $\langle S^z \rangle \Phi$ by $S(S+1)/3$ (from equation 78) we then have, in the limit $\langle S^z \rangle \to 0$,

$$\frac{S(S+1)}{3} = \frac{kT}{2 J(0) [1 + \frac{\langle S^z \rangle}{S^2} f]} F(-1)$$

(80)

where $F(-1)$ denotes the summation

$$F(-1) \equiv \frac{1}{N} \sum_k \frac{J(0)}{J(0) - J(\vec{k})}$$

(81)

This summation has been evaluated by Watson; it has the values

$$F(-1) = 1.51638 (S.C.); 1.39320 (B.C.C.); 1.34466 (F.C.C.)$$

(82)

Equation (80) determines the Curie temperature. However we must evaluate the limiting value of the quantity $\frac{\langle S^z \rangle}{S^2} f$ which appears in that equation. From equation (57) defining f, again expanding the Bose factor,

$$\frac{\langle S^z \rangle}{S^2} f = \frac{\langle S^z \rangle}{NS^2 J(0)} \sum_k J(\vec{k}) \left\{ \frac{2 \langle S^z \rangle}{kT} [J(0) - J(\vec{k})] [1 + \frac{\langle S^z \rangle}{S^2} f] \right\}^{-1}$$

(83)

or

$$\frac{\langle S^z \rangle}{S^2} f [1 + \frac{\langle S^z \rangle}{S^2} f] = \frac{F(-1) - 1}{2 S^2 J(0)} kT$$

(84)

Eliminating $\frac{\langle s^z \rangle}{s^z}$ between this equation and equation (80) we finally find the equation determining the Curie temperature.

$$\frac{kT_c}{J} = \frac{2\beta}{9} \frac{(S+1)}{F^2(-1)} \left[(4S+1) F(-1) - (S+1) \right] \tag{85}$$

In Table I we give the values of kT_c/J as estimated by Brown and Luttinger[3] by extrapolation of the series expansion for the susceptibility in powers of $1/T$. We also list the values obtained by Tahir-Kheli and ter Haar[9], and the values calculated from equation (85). It will be noted that our values are higher than those of Brown and Luttinger, whereas those of Tahir-Kheli and ter Haar are lower. For spin 2 the deviation of our results from the Brown and Luttinger values is of the order of 3%, and the agreement improves with increasing spin. In fact Domb and Sykes have recently published[4] an estimate of the limiting value $\frac{kT_c}{JS(S+1)}$ for large S, for the face centered cubic lattice. This estimate was obtained by a painstaking examination of the systematics of the extrapolation of high temperature series. They obtain the value of 6.384, with which our value of 6.45 agrees within 1%.

Domb and Sykes[4] also give estimates of kT_c/J for $S = \frac{1}{2}$ and $S = 1$ only, for the face-centered cubic lattice; their values are 4.07 and 11.95 respectively. These are lower than the Brown and Luttinger results, and further aggravate the disparity between our values and the estimated values for these small spins. It is apparent, both from the Curie temperatuees and from the low temperature results, that our approximations are more reliable for large spin, being particularly bad for spin $\frac{1}{2}$. Fortunately most cases of practical interest are associated with spins greater than $\frac{1}{2}$, for which the results appear to be quite reliable.

TABLE I

Curie Temperatures for Cubic Lattices, Nearest Neighbor Interaction

S	Simple Cubic			Body Centered Cubic			Face Centered Cubic		
	Brown Lutt.	T-Kheli t. Haar	Callen	Brown Lutt.	T-Kheli t Haar	Callen	Brown Lutt.	T-Kheli t Haar	Callen
$\frac{1}{2}$	1.9	2.0	2.7	2.39	2.9	3.7	4.2	4.5	5.6
1	5.4	5.3	6.5	7.82	7.7	9.1	12.7	11.9	13.9
$\frac{3}{2}$	10.6	9.4	11.2	15.42	14.4	16.6	24.7	22.3	25.5
2	17.5	15.8	17.9	25.17	23.0	26.2	40.0	35.7	40.1
$\frac{5}{2}$	25.8	23.1	26.8	37.10	33.5	37.9	58.7	52.1	58.3
3	35.7	31.6	36.4	51.19	45.9	51.6	80.9	71.4	79.5

For the face-centered cubic Domb and Sykes[4] give

$$\lim_{S\to\infty}\left(\frac{kT_c}{JS(S+1)}\right) = 6.38 \text{ (Domb and Sykes)}$$

$$= 5.95 \text{ (Tahir-Kheli and ter Haar)}$$

$$= 6.45 \text{ (Callen)}$$

It should, perhaps, be noted that the Curie temperatures are quite sensitive to the decoupling parameter α, of equation (20). If the chosen value of α were to be multiplied by $\left(\langle S^z \rangle / S\right)^\epsilon$, where ϵ is any positive constant no matter how small, the Curie temperatures would become identical to those obtained by Tahir-Kheli and ter Haar, whereas the low temperatures and the high temperature behavior of the theory would remain unaltered.

Finally, the high temperature expansion of the susceptibility χ is of interest. We assume $\langle S^z \rangle$ small, and maintain only terms proportional to the applied magnetic field. Equation (78) remains valid, equation (79) contains the additional Zeeman term, and the analogue of equation (80) becomes

$$\frac{S(S+1)}{3} = \frac{\chi kT}{\mu^2} \frac{1}{N} \sum_K \left\{ 1 + \frac{2\chi}{\mu^2} \left[J(0) - J(\vec{K}) \right] \left[1 + \frac{\langle S^z \rangle}{S^2} f \right] \right\}^{-1} \tag{86}$$

Similarly the analogue of equation (83) is

$$\frac{\langle S^z \rangle}{S^2} f = \frac{\chi kT}{\mu^2} \frac{1}{S^2 J(0)} \frac{1}{N} \sum_K J(\vec{K}) \left\{ 1 + \frac{2\chi}{\mu^2} \left[J(0) - J(\vec{K}) \right] \left[1 + \frac{\langle S^z \rangle}{S^2} f \right] \right\}^{-1} \tag{87}$$

It is convenient to denote

$$\left[1 + \frac{\langle S^z \rangle}{S^2} f \right] \equiv X \tag{88}$$

and to define a quantity χ_1, by

$$\chi \equiv \lim_{H \to 0} \frac{\mu \langle S^z \rangle}{H} \equiv \frac{\mu^2 S(S+1)}{3kT} \left(1 + \chi_1 \right) \tag{89}$$

Then multiplying equation (86) by $[1 + \frac{2\chi}{\mu^2} J(o) X]$

and equation (87) by $- \frac{2\chi}{\mu^2} S^2 J(o) X$

and adding, we find

$$X^2 - \frac{4S+1}{3S} X + \frac{kT}{2S^2 J(o)} \frac{\chi_1}{1+\chi_1} = 0 \tag{90}$$

Furthermore, expanding the summand in equation (86) we find

$$1 = (1+\chi_1)\left[1 - \frac{2J(o)}{\mu^2} X \chi + \frac{4J^2(o)}{\mu^4} X^2 \chi^2 \frac{z+1}{3} + \cdots \right] \tag{91}$$

In this summation we have employed the identities[9]

$$F(1) = 1 \quad , \quad F(2) = \frac{z+1}{3} \tag{92}$$

where

$$F(n) \equiv \frac{1}{N} \sum_{\kappa} \left(\frac{J(o) - J(\vec{\kappa})}{J(o)} \right)^n \tag{93}$$

The quantity X can now be eliminated between equations (90) and (91), enabling χ_1 to be evaluated in a series in $1/T$. In this way we find

$$\chi = \frac{\mu^2 S(S+1)}{3kT} \left[1 + \frac{T_M}{T} + \left(1 - \frac{2S-1}{3zS} \right)\left(\frac{T_M}{T} \right)^2 + O\left(\frac{1}{T^3} \right) \right] \tag{94}$$

where T_M is the Curie temperature of the molecular field theory:

$$kT_M \equiv \frac{2}{3} z J S(S+1) \tag{95}$$

The two leading terms in this expansion are in agreement with the values found[3] by a rigorous expansion of the susceptibility in powers of $1/T$.

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Appendix

It is easily corroborated that the solution of the differential equation (44), satisfying the boundary conditions (46) and (47) is

$$\Omega(a) = \frac{1}{1+\Phi} \frac{\omega(-s,a)\, \mathcal{D}_s\, \omega(s+1,0) - \omega(s+1,a)\, \mathcal{D}_s\, \omega(-s,0)}{\mathcal{D}_s\, \omega(s+1,0) - \mathcal{D}_s\, \omega(-s,0)} \tag{A.1}$$

where

$$\omega(x,a) \equiv \frac{e^{ax}}{(1+\Phi)e^{a} - \Phi} \tag{A.2}$$

The evaluation of the derivatives $\mathcal{D}_s\, \omega(x,0)$ is then required to reduce this result to equation (48). Consider

$$\mathcal{D}_s\, \omega(x,a) = \prod_{-s}^{s} (D-p) \frac{e^{ax}}{(1+\Phi)e^{a} - \Phi} \tag{A.3}$$

$$= \prod_{-s}^{s} \left(y \frac{d}{dy} - p\right) \frac{y^{x}}{(1+\Phi)y - \Phi} \tag{A.4}$$

where

$$y = e^{a} \tag{A.5}$$

Expanding in powers of y, and noting that $y \frac{d}{dy} y^{n} = n y^{n}$ we find

$$\mathcal{D}_s\, \omega(x,a) = -\frac{1}{\Phi} \sum_{n=x}^{\infty} \left(\frac{1+\Phi}{\Phi}\right)^{n-x} y^{n} \prod_{p=-s}^{s} (n-p) \tag{A.6}$$

We now take a = o (or y = 1), let x = S + 1, and change variables from n to m = n - S and from p to r = S - p. Then

$$\mathcal{D}_s\, \omega(s+1,0) = \frac{-1}{\Phi} \sum_{m=0}^{\infty} \frac{(m+2s+1)!}{m!} \left(\frac{1+\Phi}{\Phi}\right)^{m} \tag{A.7}$$

Similarly, taking a = o, letting x = -S and changing variables from n to m = n - S - 1, and from p to r = S + 1 - p we find

$$\alpha_s \, \omega(-S, 0) = - \frac{(1+\Phi)^{2S+1}}{\Phi^{2S+2}} \sum_{m=0}^{\infty} \frac{(m+2S+1)!}{m!} \left(\frac{1+\Phi}{\Phi} \right)^m \qquad (A.8)$$

From equations (A.7) and (A.8) we note that the ratio of the relevant quantities is

$$\frac{\alpha_s \, \omega(-S, 0)}{\alpha_s \, \omega(S+1, 0)} = \frac{(1+\Phi)^{2S+1}}{\Phi^{2S+1}} \qquad (A.9)$$

Finally inserting this ratio into equation (A.1) gives equation (48).

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