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# AN INTERPOLATION APPROACH TO THE GREEN FUNCTION THEORY OF FERROMAGNETISM

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## 1. INTRODUCTION

The problem of determining the thermodynamics of a Heisenberg ferromagnet, as a function of the temperature and the magnetic field, has been rigorously studied in the low temperature region[1-5] where a series expansion in powers of $\frac{T}{Tc}$ (where Tc is the Curie Temperature) is valid. Similarly, at high temperatures the thermodynamic perturbation theory[6-9] has been used to evaluate the free energy as a power expansion in $^{Tc}/_{T}$. These methods, however, are applicable only far from the transition region and therefore approximate theories, such as the Weiss molecular field[10] theory and the various cluster theories,[11-16] have been proposed which give qualitatively good results near and beyond the transition temperature. The Curie temperatures can also be estimated from these methods. The best estimates, however, are obtained by extrapolating the high temperature results to the transition region.[6-9]

All these methods suffer from the drawback that their applicability is limited to a particular range of temperatures.

Recently the technique of double time-temperature dependent Green functions[17-22] has been successfully applied to the ferromagnetic problem. The virtue of this method is that it provides the temperature and the magnetic field dependence of the magnetization over the entire temperature range with reasonable accuracy. Furthermore, the Curie temperatures provided by this theory seem to be in close agreement with the best estimates to date (compare references 8 and 21). However these approximate Green function theories suffer from the weakness that they lack a detailed agreement with the exact low temperature and the high temperature expansions. For example, the low temperature result for the magnetization differs from the spin wave theory result in the order $(T/Tc)^3$. Similarly, at high temperatures, the susceptibility disagrees with the exact result in the order $(Tc/T)^3$.

The problem of improving on the Bogolyubov and Tyablikov[17-18], and Tahir-Kheli and ter Haar,[21] approximation has recently been studied by Callen[23]. In his method the higher order Green functions, occurring in the equations of motion of the Green functions of the spin operators, are decoupled by a plausible physical criterion. Callen's ingenious approximation consists essentially in taking into account the fluctuations of the operator $S^z_g$ i.e. the z-component of the spin operator referred to lattice site g, around its statistical average $\langle S^z \rangle$. These fluctuations were neglected in the earlier work. Callen's theory successfully predicts the correct spin wave energies at low temperatures and also leads to an accurate estimate of the Curie temperatures in the limit of large spin values. For low spins, however, the results are less accurate and for the particular case of $S = 1/2$ the expression for the low temperature magnetization is still found to contain the anomalous $(T/Tc)^3$ term.

At high temperatures, Callen's theory behaves rather similarly to the random phase theory*, and the susceptibility agrees with the

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*Here and henceforth we shall refer to the Tyablikov[18], and Tahir-Kheli and ter Haar,[21] approximation as the 'random phase' approximation (R.P.A.) because of its equivalence to Engelert's[24] approximation.

exact results up to order $(Tc/_T)^2$.

From a formal point of view both the random phase approximation[21] and the Callen approximation[23] assume that the spin Green function obeys the Dyson equation, the higher order Green function being written as a product of a mass operator and a lower order Green function. These theories differ in the form that they postulate for the mass operator.

In a recent study of the problem, Wortis[5] has shown that the Green functions of a Heisenberg ferromagnet, in common with those pertaining to condensed Bose systems[25], do not obey a Dyson equation with a simple mass operator; alternatively, if an "effective mass operator" is defined by a Dyson equation, it is found to possess an anomalous structure. It is our purpose here to investigate the form of the effective mass operator. We find that a particularly convenient representation of the "effective mass operator" is such that the higher order Green function is the sum of an anomalous additive term plus the product of a simple mass operator and a lower order Green function. Knowing the detailed form of the spin wave dispersion law,[4,26,27,] and keeping in mind the results of the R.P.A.[21] and the Callen theory[23], we infer the form of the anomalous term and of the remaining mass operator. Similarly, at high temperatures, we invoke the series expansions for the susceptibility and the magnetic energy, derived by Rushbrooke and Wood[8] and Domb and Sykes[9].

We find that the mass operator is almost exactly that proposed by Callen[23]. At low temperatures, the anomalous term is quite important for spin $\frac{1}{2}$ , but it becomes insignificant for higher spins. Again, at high temperatures, we find that the anomalous term contributes only weakly, i.e. in the order $(Tc/_T)^4$, to the susceptibility and is less important the larger the spin S. Near the Curie point, however, the anomalous term makes a significant contribution even for moderately large S, i.e. $S \approx 10$. For $S >> 1$, the anomalous term is again small and our results are identical with those of Callen[23].

The Green function theory here obtained provides an interpolation scheme between high and low temperatures. The results for the Curie temperatures and the critical magnetic energy agree with the estimates of Domb and Sykes and Rushbrooke and Wood to about 1% for all spins. The critical behavior of the susceptibility, as T approaches Tc from above, and of the magnetization, as T approaches Tc from below, is investigated. It is found that within the callen[23] and the random phase approximations[21], the susceptibility just above the Curie temperature has a form $\chi = \text{const.}(1 - Tc/_T)^{-2}$ whereas the magnetization just below the Curie temperature approaches zero as $(1 - T/_{Tc})^{1/2}$. The present theory, however, can be set to achieve agreement of $\chi$ with the high temperature series result of Domb and Sykes, i.e $\chi = \text{const.} \cdot (1-Tc/_T)^{-4/3}$. If this is done we find that just below the Curie temperature the magnetization, M(T), obeys a relation of the form $M(T) = \text{const.}\ (1-T/_{Tc})^{1/3}$.

## 2. THE GREEN FUNCTION

The mathematics of the retarded and advanced-double time-temperature dependent Green functions has been given by Zubarev[19], and by Bonch-Bruevich and Tyablikov[20] to whom we refer for details. We outline here those features of this technique which are relevant to the present work.

The Green function of operators A and B, <<A(t); B(t'>>, is defined as follows:

$$\langle\langle A(t) ; B(t') \rangle\rangle^{\text{ret.}} \equiv -\frac{i}{\hbar}\, \Theta(t-t') \left\langle \left[ A(t) , B(t') \right]_- \right\rangle$$

$$\langle\langle A(t) ; B(t') \rangle\rangle^{\text{adv.}} \equiv +\frac{i}{\hbar}\, \Theta(t'-t) \left\langle \left[ A(t) , B(t') \right]_- \right\rangle \tag{2.1}$$

where A(t) is the Heisenberg operator referred to time t, i.e.

$$A(t) = e^{i\left(\frac{H}{\hbar}\right)t}\; A \; e^{-i\left(\frac{H}{\hbar}\right)t} \tag{2.2}$$

and where H is the system Hamiltonian, $2\pi\hbar$ the Planck constant, A the Schroedinger (time independent) operator, square brackets denote a commutator, and single pointed brackets denote an ensemble average

$$\langle \cdots \rangle = \frac{\mathrm{Tr}\left(e^{-\beta H} \cdots\right)}{\mathrm{Tr}\left(e^{-\beta H}\right)} \tag{2.3}$$

$\Theta(x)$ is the step function

$$\Theta(x) = \begin{cases} 1 , & x > 0 \\ 0 , & x < 0 \end{cases} \tag{2.4}$$

and $\beta = \frac{1}{k_B T}$ ($k_B$: Boltzmann's constant; T: absolute temperature).

Differentiating the Green functions given in (2.1) with respect to t we get

$$i\hbar\frac{d}{dt}\langle\langle A(t);B(t')\rangle\rangle = \delta(t-t')\langle[A(t),B(t)]_{-}\rangle + \langle\langle[A(t),H]_{-};B(t')\rangle\rangle \tag{2.5}$$

where δ(···) denotes the Dirac delta function and the superscripts ret. and adv. have been dropped because (2.5) is the same for either of the two cases. The Green function <<[A(t),H]$_{-}$: B(t')>> in general involves Green functions of higher order than the original <<A(t); B(t')>>, except, of course, for the trivial cases of non interacting systems where exact solutions can be obtained. One has therefore to linearize the equation (2.5) by a suitable decoupling approximation. Once (2.5) has been solved for <<A(t); B(t')>>, the spectral theorems[19-20] may be invoked to get the time correlation functions,

$$\begin{aligned}&\langle B(t')\,A(t)\rangle\\ &= \lim_{\epsilon\to+0}\ \frac{i}{\hbar}\int_{-\infty}^{+\infty}\frac{[\langle\langle A;B\rangle\rangle_{E+i\epsilon} - \langle\langle A;B\rangle\rangle_{E-i\epsilon}]}{e^{\beta E}-1}\ e^{-i\left(\frac{E}{\hbar}\right)(t-t')}\,.\,dE\\ &= \langle A(t)\,B(t'+i\hbar\beta)\rangle\end{aligned} \tag{2.6}$$

where <<A;B>>$_{(E)}$ denotes the energy Fourier transform of <<A(t); B(t')>>. Equations (2.5) and (2.6) are the only two equations required for our calculations here.

## 3. CORRELATION FUNCTION OF THE TRANSVERSE COMPONENTS OF SPINS

We shall assume the ferromagnetic spin system to be described by a Heisenberg type interaction with isotropic exchange and in the presence of a spatially homogeneous time independent magnetic field, B, directed along the positive z-axis. The Hamiltonian, H, therefore is

$$H = -\frac{E_0}{\hbar}\sum_f \overset{z}{S}_f - \sum_{f,m} I(f-m)\,\vec{S}_f\cdot\vec{S}_m \tag{3.1}$$

where $E_0 = +\frac{\mu\cdot B}{S}$, $\mu$ is the magnetic moment per ion, $S_f^{x,y,z}$ the cartesian components of the spin operator for the site f and I(f-m) the exchange integral between ions at sites f and m. As usual, we assume that the self-exchange terms vanish i.e. I(f-f) = 0. We shall consider here the following Green functions:

$$G^{(n)}_{g;l}(t-t') \equiv \langle\langle \overset{+}{S}_g(t)\,;\,C^{(n)}_l(t')\rangle\rangle \tag{3.2}$$

where

$$\overset{\pm}{S}_g = \overset{x}{S}_g \pm i\,\overset{y}{S}_g \tag{3.3}$$

and

$$C^{(n)}_l(t') = \left[\overset{z}{S}_l(t')\right]^n \overset{-}{S}_l(t') \tag{3.4}$$

where n is a positive integer or zero.

From equations (2.5) and 3.1) we find the equation of motion of $G^{(n)}_{g;l}(t-t')$,

$$\left[i\hbar\frac{d}{dt} - E_0\right] G^{(n)}_{g;l}(t-t') = \delta(t-t')\,Q(n)\,\delta_{g,l} + 2\hbar\sum_f I(g-f)\,\langle\langle \overset{z}{S}_f(t)\overset{+}{S}_g(t) - \overset{z}{S}_g(t)\overset{+}{S}_f(t)\,;\,C^{(n)}_l(t')\rangle\rangle \tag{3.5}$$

where

$$Q(n) = \left\langle\left[\overset{+}{S}_g\,,\,\left[\overset{z}{S}_g\right]^n \overset{-}{S}_g\right]\right\rangle \tag{3.6}$$

$$= \left\langle\left[\left(\overset{z}{S}_g-\hbar\right)^n - \left(\overset{z}{S}_g\right)^n\right]\left[S(S+1)\hbar^2 + \hbar\overset{z}{S}_g - \left(\overset{z}{S}_g\right)^2\right] + 2\left(\overset{z}{S}_g\right)^{n+1}\hbar\right\rangle$$

In order to solve Eq. (3.5) for the Green function $G^{(n)}$, it must first be linearized. The simplest approximation for this purpose is the R.P.A.[18,21,24] which neglects the dynamical correlations between $S_g^z(t)$ (or $S_f^z(t)$ ) and the remaining operators and replaces it by the statistical average $\langle S^z \rangle$. In order to take into account the fluctuations of $S_g^z$ around its average $\langle S^z \rangle$, Callen[23] has recently suggested a decoupling scheme of the following type:

$$\hbar \langle\langle S_g^z(t)\, S_f^+(t)\, ;\, C_\ell^{(n)}(t') \rangle\rangle \xrightarrow[\text{CALLEN}]{} \hbar \langle S^z \rangle\, G_{f;\ell}^{(n)}(t-t') - \alpha \cdot \langle S_g^- S_f^+ \rangle\, G_{g;\ell}^{(n)}(t-t') \qquad (3.7)$$

Callen chooses $\alpha$ on the grounds that since the random phase approximation, represented here by the choice $\alpha = 0$, affords a reasonable first approximation, the additional part $S_g^z - \langle S^z \rangle$ introduced here must be self consistently small at all temperatures. This requirement can be satisfied if $\alpha = \frac{1}{2S}$ at low temperatures and if $\alpha$ decreases at least as fast as $\langle S^z \rangle$ at high temperatures. Callen's choice of $\alpha = \langle S^z \rangle / 2S^2$ incorporates both these features.

In order to look for an improvement over these approximations, we proceed as follows. Rather than decoupling the Green functions $\langle\langle S_g^z(t)\, S_f^+(t);\, C_\ell^{(n)}(t') \rangle\rangle$ and $\langle\langle S_f^z(t)\, S_g^+(t)\, ;\, C_\ell^{(n)}(t) \rangle\rangle$ separately, we notice that in equation (3.5) the relevant expression, $F^{(n)}$, to be decoupled is a function of the difference of these Green functions, i.e.

$$F^{(n)} \equiv 2\hbar \sum_f I(g-f) \langle\langle S_f^z(t)\, S_g^+(t) - S_g^z(t)\, S_f^+(t)\, ;\, C_\ell^{(n)}(t') \rangle\rangle \qquad (3.8)_a$$

Because of the translational and the time invariance of the Hamiltonian and the translational invariance of the lattice, $F^{(n)}$ must be a function of the vector spatial separation $(g-\ell)$ and the time difference $(t-t')$, i.e.

$$F^{(n)} \equiv F^{(n)}(g-\ell\,;\,t-t') \tag{3.8$_b$}$$

In the R.P.A. the function $F^{(n)}$ takes the following simple form[21].

$$F^{(n)}(g-\ell\,;\,t-t') \xrightarrow[\text{R.P.A.}]{} 2\hbar\,\langle S^z\rangle \sum_f I(g-f)\left[G^{(n)}_{g;\ell}(t-t') - G^{(n)}_{f;\ell}(t-t')\right] \tag{3.9}$$

In view of the fact that the R.P.A. constitutes a reasonable first approximation, we propose to investigate a decoupling scheme of the form:

$$F^{(n)}(g-\ell\,;\,t-t')$$
$$=2\hbar\langle S^z\rangle\sum_f I(g-f)\left[G^{(n)}_{g;\ell}(t-t') - G^{(n)}_{f;\ell}(t-t')\right] + \Delta^{(n)}(g-\ell\,;\,t-t') \tag{3.10}$$

where $\Delta^{(n)}$ represents the corrections to the R.P.A.

Introducing (3.10) into (3.5) we get the equation of motion in the following form: $\left[i\hbar\frac{d}{dt} - E_0\right] G^{(n)}_{g;\ell}(t-t') - \Delta^{(n)}(g-\ell\,;\,t-t')$

$$= \delta(t-t')\,\delta_{g,\ell}\,Q(n) + 2\hbar\langle S^z\rangle\sum_f I(g-f)\left[G^{(n)}_{g;\ell}(t-t') - G^{(n)}_{f;\ell}(t-t')\right] \tag{3.11}$$

The translational invariance dictates Fourier transformation with respect to the inverse lattice, i.e.

$$G^{(n)}_{g;\ell}(t-t') = \frac{1}{N}\sum_K G^{(n)}_K(t-t')\,e^{i\vec{K}\cdot(g-\ell)} \tag{3.12}$$

$$\Delta^{(n)}(g-\ell\,;\,t-t') = \frac{1}{N}\sum_K \Delta^{(n)}(K\,;\,t-t')\,e^{i\vec{K}\cdot(g-\ell)} \tag{3.13}$$

$$\delta_{g,\ell} = \frac{1}{N}\sum_K e^{i\vec{K}\cdot(g-\ell)} \tag{3.14}$$

where N is the total number of sites in the lattice and the inverse lattice sums are restricted to the first Brillouin zone. Fourier transformation with respect to the energy E is also possible, i.e.

$$G_K^{(n)}(t-t') = \frac{1}{\hbar}\int_{-\infty}^{+\infty} G_K^{(n)}(E)\, e^{-i\left(\frac{E}{\hbar}\right)(t-t')} . dE \qquad (3.15)$$

$$\Delta^{(n)}(K;t-t') = \frac{1}{\hbar}\int_{-\infty}^{+\infty} \Delta^{(n)}(K;E)\, e^{-i\left(\frac{E}{\hbar}\right)(t-t')} . dE \qquad (3.16)$$

$$2\pi . \delta(t-t') = \frac{1}{\hbar}\int_{-\infty}^{+\infty} e^{-i\left(\frac{E}{\hbar}\right)(t-t')} . dE \qquad (3.17)$$

Using (3.12) - (3.17), equation (3.11) takes the form

$$[E - E_K]\, G_K^{(n)}(E) = (1/2\pi)\, Q(n) + \Delta^{(n)}(K;E) \qquad (3.18)$$

where $E_K$ is the elementary excitation energy obtained in the R.P.A. (compare equation (3.11) of reference 21.), i.e.

$$E_K = E_0 + 2\hbar \langle \overset{z}{S} \rangle\, J(0,K) \qquad (3.19)$$

Here we have used the notation

$$\sum_f I(f)\, e^{i\vec{K}.f} = J(K) \qquad (3.20)$$

$$J(K) - J(K') = J(K,K') \qquad (3.21)$$

It is clear that in a simple mass operator type approximation, the correction $\Delta^{(n)}$ will be of the form:

$$\Delta^{(n)}(K;E) \longrightarrow \Sigma_K^{(n)}(E) . G_K^{(n)}(E) \qquad (3.22)$$

where $\Sigma_k^{(n)}(E)$, in general, would be complex and would not involve terms proportional to the inverse of $G_k^{(n)}(E)$. Recently, Wortis[5] has carried out a careful diagrammatic study of the structure of the Green function approximations in the context of thermodynamic perturbation theory relevant to the low temperatures. He observes that the Green function pertaining to the spin problems does not, in general, have the structure common to Green functions for normal particle systems and consequently, the mass operator - the generalized analogue of $\Sigma_k^{(n)}(E)$ defined in (3.22) - has an anomalous structure unique to the spin systems. In fact, Wortis notes that whereas in the case of the normal particle systems the mass operator is a functional of the Green function $G^{(n)}$, for the spin systems an effective mass operator must be considered to be a functional also of the inverse of the Green function. (Compare argument leading to equation (5.5) of reference 5). For the present purposes, it is convenient to extract the part proportional to the inverse of $G_k^{(n)}(E)$ from the generalized, effective mass operator, i.e.

$$\Delta^{(n)}(k;E) = M_k(\text{[illegible]}E)\, G_k^{(n)}(E) + D^{(n)} \tag{3.23}$$

Here $D^{(n)}$ is a function of the system temperature and of the variables n, k and E, and M(k;E) is the analogue of the usual mass operator. In order to gain some insight into the form of $D^{(n)}$, we Fourier transform (3.23) as follows:

$$\Delta^{(n)}(g-l;t-t') \tag{3.24}$$

$$= \frac{1}{N\hbar}\int_{-\infty}^{+\infty}\sum_k M_k(\text{[illegible]}E)\cdot G_k^{(n)}(E)\, e^{i\vec{k}\cdot(g-l)-i\left(\frac{E}{\hbar}\right)(t-t')}\cdot dE + D^{(n)}(g-l;t-t')$$

A convenient starting approximation for $D^{(n)}$ can now be arrived at if we refer to equations (3.10) and (3.11). It is reasonable to assume that because of the discontinuous nature of the Green function in the time variable (t-t') - refer equations (2.1) - (2.4) - $\Delta^{(n)}$ will also be a discontinuous function of (t-t'). The first term on the r.h.s. of equation (3.24) already incorporates this discontinuity because of the presence of the Green function $G^{(n)}$. A delta function dependence would therefore seem to suggest itself for the remaining term $D^{(n)}$, i.e.

$$D^{(n)}(g-\ell\,;t-t') \longrightarrow \delta(t-t')\cdot R(g-\ell)\cdot Q(n)\cdot Y(T) \tag{3.25}$$

where $Y(T)$ is an arbitrary function of the temperature T, $R(g-\ell)$ is a suitable function of the vector $g-\ell$ and the temperature. Here Q(n) has been introduced for convenience in later calculations and is the same as in (3.6). Equations (3.25), (3.10) and $(3.8)_{a,b}$ contain the essentials of the form of the basic approximation of our theory. Introducing these into the equation of motion (3.5) and carrying out the Fourier transformations described in (3.12) - (3.17) we finally get:

$$\left[E - E_k - M_k(E)\right] G_k^{(n)}(E) = (1/2\pi)\, Q(n)\left[1 + Y(T)\, R(k)\right] \tag{3.26}$$

(compare, equation (3.18).

In view of the fact that the Fourier transform of the Green function, $G_k^{(n)}(E)$, has poles at $E = E_k + M_k(E)$, we may recognize it to be the analogue of the elementary excitations of the system (see ref. 20 for a discussion of this point). In generak $M_k(E)$, which represents the shift of the true energy spectrum from that obtained in the R.P.A., will be complex. For the present purposes, however, the imaginary part of $M_k(E)$ will be ignored. It should be emphasized that strictly speaking the damping is a very important parameter of the system excitation. It determines, in fact, the limits of the applicability of the concept of quasi-stationarity with regard to the elementary excitation. One cannot, therefore, claim with any certainty that the excitation $\widetilde{E}_k$ thus chosen represents the true elementary excitation of the system until it can be ascertained that the associated damping can be neglected. At low temperatures, the applicability of this concept is not in doubt because the associated damping[4] is much smaller than $\widetilde{E}_k$. Bearing in mind this conditional interpretation of $\widetilde{E}_k$, we may proceed as follows:

From equations (3.26) and (2.6) and the identity that for real E and $\widetilde{E}_k$

$$\lim_{\epsilon \to +0} \left[ \frac{1}{E - \widetilde{E}_k + i\epsilon} - \frac{1}{E - \widetilde{E}_k - i\epsilon} \right] = -2\pi i \, \delta(E - \widetilde{E}_k) \tag{3.27}$$

we get the following expression for the static correlation function of $C_{\ell}^{(n)}$ and $S_g^{+}$ :

$$L^{(n)}(\ell - g) = \frac{Q(n)}{N} \sum_k \Phi(k) \left[ 1 + R(k) \cdot \Upsilon(T) \right] e^{i \vec{k} \cdot (g - \ell)} \tag{3.28}$$

where

$$L^{(n)}(\ell - g) = \left\langle \left( S_\ell^{z} \right)^{n} S_\ell^{-} S_g^{+} \right\rangle \tag{3.29}$$

$$\Phi(k) = \frac{1}{\left(e^{\beta \tilde{E}_k} - 1\right)} \qquad (3.30)$$

and

$$\tilde{E}_k = E_k + M_k(\tilde{E}_k) \qquad (3.31)$$

Considering the fact that

$$\overset{-}{S}_\ell \overset{+}{S}_\ell = S(S+1)\hbar^2 - \hbar \overset{z}{S}_\ell - \left(\overset{z}{S}_\ell\right)^2 \qquad (3.32)$$

when $\ell = g$, equation (3.31) contains both on the left and right-hand sides a sum of averages of powers of $S^z$. We can therefore write down 2S independent, simultaneous linear equations in $\langle S^z \rangle$, $\langle (S^z)^2 \rangle, \ldots\ldots, \langle (S^z)^{2S} \rangle$ by putting n in equation (3.28) equal to 1, 2, 3, ..., 2S consecutively. The equations with $n > 2S$ are not independent of the earlier ones because of the following operator relation satisfied by the spin operators:

$$\prod_{r=-S}^{r=+S} (\overset{z}{S} - r\hbar) = 0 \qquad (3.33)$$

where r takes on integral or half-odd-integral values according as S is integral or half-odd-integral.

The results for the average $\langle S^z \rangle$ can be written as follows:*

$$\langle \overset{z}{S} \rangle = \hbar\left[(S - \tilde{\Phi})(1+\tilde{\Phi})^{2S+1} + (S+1+\tilde{\Phi})(\tilde{\Phi})^{2S+1}\right] \Big/ \left[(1+\tilde{\Phi})^{2S+1} - \tilde{\Phi}^{2S+1}\right] \qquad (3.34)$$

where

$$\tilde{\Phi} = \Phi + \gamma(T) = \Phi\left[1 + \gamma'(T)\right]; \quad \Phi = \frac{1}{N}\sum_k \Phi(k) \qquad (3.35)$$

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*Similar expressions were empirically arrived at[28] after solving explicitly the 2S simultaneous equations for several different values of S. Callen[23] has since derived these results by a much more elegant method.

The expression for the average $<(S^z)^2>$ can also now be easily obtained from the relation (3.32).

On proceeding from equations (3.28) and (3.29) to equations (3.34), (3.35) we made the following choice for the function R(K):

$$R(K) = 1/\Phi(K) \tag{3.36}$$

The reasons for this choice become clear if we study the form of the transverse correlation function of the spins, $L^{(0)}(\ell-g)$. Putting n=0 in (3.28) we get:

$$\begin{aligned} L^{(0)}(\ell-g) &= \langle \overset{-}{S}_\ell \overset{+}{S}_g \rangle \\ &= 2\frac{\langle \overset{=}{S} \rangle}{N}\cdot\sum_K \Phi(K)\, e^{i\vec{K}\cdot(g-\ell)} + 2\cdot\frac{\langle \overset{=}{S} \rangle}{N}\cdot\Upsilon(T)\cdot\sum_K \Phi(K) R(K)\, e^{i\vec{K}\cdot(g-\ell)} \end{aligned} \tag{3.37}$$

At low temperatures, where it has already been argued that $\tilde{E}_k$ of equation (3.31) will represent the true spin wave dispersion law, the form of the correlation function $L^{(0)}(\ell-g)$ is known from the spin wave theory. It is known[29] that for $\ell \neq g$, this correlation function is, to a good approximation, given by the first term on the right-hand side of (3.37). Therefore, a convenient choice for R(k) is given in (3.36) because then the remaining term in (3.37) contributes only when $\ell = g$, i.e. $L^{(0)}(\ell-g)$

$$= 2\cdot\frac{\langle \overset{=}{S} \rangle}{N}\cdot\sum_K \Phi(K)\, e^{i\vec{K}\cdot(g-\ell)} + 2\langle \overset{=}{S} \rangle\cdot\Upsilon(T)\cdot\delta_{g,\ell} \tag{3.38}$$

Having thus specified, up to an arbitrary temperature dependent parameter $\gamma(T)$, the form of the function $D^{(n)}$ of equation (3.24) we investigate the function $M_k(\tilde{E}_k)$. Once again we invoke a result known from the rigorous spin wave theory that at low temperatures

$$\tilde{E}_K - E_K = M_K(\tilde{E}_K) \underset{T \ll T_c}{=} \frac{2\hbar^2}{N}\cdot J(0,K)\cdot\sum_{K'} \Phi(K')\, J(K')/J(0) \tag{3.39}$$

(compare equation (3.7) of reference 4). This suggests the following form for $M_k(E_k)$:

$$M_k(\tilde{E}_k) = \sigma(T) \cdot \frac{2\hbar^2}{N} J(0,k) \sum_{k'} \Phi(k') J(k')/J(0) \qquad (3.40)$$

where $\sigma(T)$ is to be a temperature dependent parameter such that in the limit of low temperatures $\sigma(T)$ approaches unity. Equations (3.28) -(3.31), (3.34)- (3.35) and (3.40) constitute a set of coupled equations which must be solved self consistently to determine the magnetization and the transverse correlation function.

The formalism developed so far is valid irrespective of the lattice structure and the spatial dependence of the exchange integral I(g-f). In order to facilitate the comparison with the results of other theories we shall, in what follows, restrict consideration to lattices of cubic symmetry with nearest neighbor exchange interaction.

$$I(g-f) = I, \quad \text{if f and g are nearest neighbors,}$$
$$= 0, \qquad (3.41)$$

With these simplifying assumptions, the elementary excitation energies $\tilde{E}_k$, of equations (3.31) and (3.40), take the following simple form:

$$\tilde{E}_k = E_0 + 2\hbar J(0,k)\left[\langle \bar{S} \rangle + \frac{\sigma(T)\cdot\hbar}{N} \sum_{k'} \Phi(k') J(k')/J(0)\right] \qquad (3.42)$$

## 4. THE LOW TEMPERATURE REGION

In this section we analyze the results appropriate to temperatures low compared to the Curie temperature.

At these temperatures the magnetization is close to the saturation value and the average $\langle \overset{z}{S} \rangle$ is little different from S. The function $\tilde{\Phi}$ therefore is small compared with unity and equation (3.34) can be expanded in powers of $\tilde{\Phi}$ to give

$$\langle \overset{z}{S} \rangle = \hbar \left[ S - \tilde{\Phi} + (2S+1)\tilde{\Phi}^{2S+1} - (2S+1)^2 \tilde{\Phi}^{2S+2} + o(\tilde{\Phi})^{2S+3} \right] \qquad (4.1)$$

$$\langle (\overset{z}{S})^2 \rangle = \hbar^2 \left[ S^2 - (2S-1)\tilde{\Phi} + 2\tilde{\Phi}^2 - (2S+1)\tilde{\Phi}^{2S+1} + (4S^2-1)\tilde{\Phi}^{2S+2} + o(\tilde{\Phi})^{2S+3} \right] \qquad (4.2)$$

So far the arbitrary function γ'(T) remains unspecified. It is clear from the definition of $\Phi$ (see eqns. (3.30, (3.42) with $\sigma(T) = 1$) and eqn. (3.35) that at low temperatures it is the strict analogue of the thermodynamic average of the number of spin waves excited per lattice site. This requires

$$\langle \overset{z}{S} \rangle \underset{T \ll T_c}{=} \hbar \left[ S - \Phi \right] \qquad (4.3)$$

and therefore γ'(T) is completely specified:

$$\gamma'(T) = \gamma(T)/\Phi \underset{T \ll T_c}{=} \left( \frac{(2S+1)\,\Phi^{2S} + \cdots}{1 + (2S+1)\Phi - (2S+1)^2 \Phi^{2S} + \cdots} \right] \qquad (4.4)$$

As expected, γ'(T) is small and decreases rapidly with the increase in S. In fact, γ'(T) is significant only for S∼1. Thus the simple mass operator approximation, which ignores γ', is sufficient for large S.

The calculation of $\tilde{E}_k$ involves an integration (or rather a summation) over the inverse lattice vector k'. The integrand, $\Phi(k')\,J(k')$, on the other hand, depends on the form of $\tilde{E}_k$. Therefore an iteration process has to be used. We first calculate $\langle \overset{z}{S} \rangle$, and the integral over k' in equation (3.42), in the R.P.A. The second iteration is obtained by introducing these results into the expression for $\tilde{E}_k$ and then recalculating $\langle \overset{z}{S} \rangle$ and the integral $\frac{1}{N}\sum_{k'}\Phi(k')J(k')$ At low temperatures, this iteration process converges very fast and it turns out that no further iteration is necessary beyond the first iteration cycle because the terms not included in the first cycle contribute in a higher order in the ratio (T/Tc) than the ones retained.

The results for the spin wave energies, $\tilde{E}_k$, and the magnetization M(T) are found to be as follows:

$$E_K \underset{T \ll T_c}{=} E_0 + 2S\hbar^2 J(0,K)\left[1 - (\pi\cdot\nu/S)\, Z_{5/2}\, \Theta^{5/2} - o(\Theta^3)\right] \quad (4.5)$$

$$\begin{aligned} M(T) &= M(0)\,\langle \overset{z}{S} \rangle / S\hbar \\ &\underset{T \ll T_c}{=} (M(0)/S)\left[S - \alpha_0\Theta^{3/2} - \alpha_1\Theta^{5/2} - \alpha_2\Theta^{7/2} - \alpha_3\Theta^4 - o(\Theta^{9/2})\right] \end{aligned} \quad (4.6)$$

where

$$\Theta = \left[3K_B T / J(0)\hbar^2 4\pi\nu S\right] \quad (4.7)$$

$$\begin{aligned} \nu &= 1\text{, for simple cubic lattice} \\ &= (3/4)(2)^{2/3}\text{, body-centered cubic} \\ &= (2)^{1/3}\text{, face-centered cubic} \end{aligned} \quad (4.8)$$

$$Z_n = \sum_{r=1}^{\infty}\left[e^{-rE_0\beta} / r^n\right] \quad (4.9)$$

$$\begin{aligned}
\alpha_0 &= Z_{3/2} \\
\alpha_1 &= (3/4)\pi\nu Z_{5/2} \\
\alpha_2 &= \pi^2 \omega \nu^2 Z_{7/2} \\
\alpha_3 &= (3/2)\pi\nu Z_{3/2} \cdot Z_{5/2} \cdot (1/S)
\end{aligned} \tag{4.10}$$

and $\omega = \frac{33}{32}$, simple cubic

$= \frac{281}{288}$, body-centered cubic

$= 15/16$, face-centered cubic (4.11)

These results are the same as obtained by Dyson[1] (neglecting the small corrections arising from the 2nd, and the higher, Born approximation spin wave scattering). The results for the average $\langle (S^z)^2 \rangle$ are obtained in a similar fashion and we get:

$$\begin{aligned}
\langle (S^z)^2 \rangle = \hbar^2 \Big[ S^2 &- (2S-1)\{\alpha_0 \theta^{3/2} + \alpha_1 \theta^{5/2} + \alpha_2 \theta^{7/2} + \alpha_3 \theta^4\} \\
&+ 2 \{\alpha_0^2 \theta^3 + 2\alpha_0 \alpha_1 \theta^4\} \\
&- 2S(2S+1)\{\alpha_0^{2S+1} \theta^{3S+3/2} + (2S+1)\alpha_0^{2S} \alpha_1 \theta^{3S+5/2}\} \\
&+ 0(\theta^{9/2}) \Big]
\end{aligned} \tag{4.12}$$

For the case S = 1/2, the r.h.s. of equation (4.12) is exactly equal to $\hbar^2/4$.

Combining equations (3.38), (4.4)-(4.11), we can find the correlation function of the transverse components of spins appropriate to the low temperatures,

$$\begin{aligned}
\langle S_g^x S_l^x \rangle &= \langle S_g^y S_l^y \rangle \\
&\underset{T \ll T_c}{=} \frac{\hbar \langle S^z \rangle}{N} \sum_k \Phi(k) e^{i\vec{k}\cdot(g-l)} + \hbar \langle S^z \rangle \cdot Y(T)\, \delta_{g,l}
\end{aligned} \tag{4.13}$$

It will be noticed that (4.13) is actually an improvement on the spin wave result in that for g $= \ell$, it also includes the term (last one on the r.h.s. of 4.13) which helps fulfill the stringent requirements of the spin kinematics. For example, for the simplest case S = 1/2, we have the exact equality

$$\langle S_g^- S_g^+ \rangle \underset{(S=1/2)}{=} \hbar^2/2 - \hbar \langle S^z \rangle \tag{4.14}$$

This equality is satisfied by (4.13) whereas the usual spin wave result, comprising only the first term on the r.h.s. of (4.13), does not satisfy it.

## 5. THE HIGH TEMPERATURE REGION

In this section we consider the high temperature expansions for the susceptibility and the magnetic specific heat.

In the presence of a small magnetic field B, the magnetization, M(T), is small and is proportional to B. In the limit $B = 0$, the energy $\tilde{E}_k$ (see equation 3.42) is therefore proportional to $\sigma(T)/B$ as long as $\sigma(T)$ does not go to zero faster than $B^2$. It is clear from the results of references 18, 21, 23 that the correct high temperature behavior of the zero field susceptibility, $\chi$, requires the proportionality of $\tilde{E}_k$ to $\langle S^z \rangle$. Therefore, it is convenient to put

$$\sigma(T) = 2\, A(T) \cdot \langle S^z \rangle^2 / \hbar^2 \tag{5.1}$$

where A(T) is a function of the temperature and is to be non-infinite, i.e. small compared with (1/B). The energies $\tilde{E}_k$ thus are (see 3.42) of the following form:

$$\tilde{E}_k = E_0 + 2\hbar \langle S^z \rangle J(0,k) \left[ 1 + X(T) \right] \tag{5.2}$$

where

$$X(T) = \left( A(T)/\hbar^2 N \right) \sum_{k'} L^{(0)}(k')\, J(k')/J(0) \tag{5.3}$$

and $L^{(0)}(k')$ is the Fourier transform of the transverse correlation function $L^{(0)}(\ell - g)$ given in (3.38). As the self exchange integral I(f-f) has been taken to zero, the sum $\sum_{k'} J(k')$ vanishes and therefore (5.3) is independent of any explicit dependence on $\gamma(T)$.

In this temperature range the quantity $\tilde{\Phi}$ is large compared with unity. Therefore equation (3.34) can be expanded in inverse powers of $\tilde{\Phi}$ to give

$$\langle S^z \rangle = \hbar \left[ S(S+1)/3\tilde{\Phi} \right] \left[ 1 - (2\tilde{\Phi})^{-1} - (2S^2+2S-9)(30\tilde{\Phi}^2)^{-1} - \dots \right] \quad (5.4)$$

$$\langle (S^z)^2 \rangle = \hbar^2 \frac{S(S+1)}{3} \left[ 1 + (4S^2+4S-3)/(30\tilde{\Phi}^2) + o(\tilde{\Phi})^{-3} \right] \quad (5.5)$$

The zero field susceptibility, $\chi$, is therefore given by the following

$$\chi = \operatorname*{Lt}_{B=0} \left[ \mu \cdot \langle S^z \rangle / S\hbar . B \right] = \left[ \mu \cdot (S+1) / 3B\tilde{\Phi} \right] \quad (5.6)$$

where we have assumed that a reasonable choice of $\gamma'$ will be such that

$$\gamma'(T) \ll B^{-1} \quad (5.7)$$

The quantity $(\tilde{\Phi} . B^{-1})$ can be expanded in inverse powers of $\gamma$ where

$$\gamma^{-1} = \beta \, J(0) \hbar^2 \quad (5.8)$$

Introducing the quantities $t_0$ and $t_1$,

$$t_0 = \tanh(\beta E_0/2) \quad (5.9)$$

$$t_1 = \tanh\left[ \langle S^z \rangle J(0,k)(1+X(T)) / \hbar \gamma J(0) \right] \quad (5.10)$$

and using equation (5.2) we can write equation (3.30) as follows:

$$\Phi(k) = -(1/2) + (1/2)\left[ 1 + t_0 t_1 / t_0 + t_1 \right] \quad (5.11)$$

$$= -(1/2) + (1/2t_0)\left[ 1 + (1 - t_0^2) \sum_{r=1}^{\infty} (-1)^r (t_1/t_0)^r \right]$$

Expanding the hyperbolic tangent $t_1$ and ignoring terms proportional to B in comparison with unity we get:

$$\begin{aligned}\Phi &= \frac{1}{N}\sum_k \Phi(k) \\ &= \left(\frac{1}{2t_0}\right)\sum_{r=0}^{\infty}(-1)^r F(r)\left[(1+X(\tau))\langle S^z\rangle/\hbar\gamma t_0\right]^r\end{aligned} \tag{5.12}$$

where

$$F(r) = \frac{1}{N}\sum_k \left[J(0,k)/J(0)\right]^r \tag{5.13}$$

Before discussing further the evaluation of $\Phi$, we recall that the R.P.A. (represented here by putting X = 0 and γ = 0) yielded the correct high temperature expansion for the susceptibility, up to, and including, terms proportional to ( $1/\tau^2$). Therefore γ must involve terms which decrease with temperature at least as fast as $1/\tau^3$.

In order to determine the correct expressions for the two parameters A(T) and γ'(T), we need also to consider the correlation function $L^{(0)}(\ell-g)$. A general expression for $L^{(0)}(\ell-g)$ is not available but the system energy and the magnetic specific heat, for which exact expansions are known,[8,9] depend sensitively on $L^{(0)}(\ell-g)$. The computation of the magnetic energy requires knowledge of the correlation functions of the transverse as well as the longitudinal components of spins. In the paramagnetic region, spatial isotropy requires that in the limit B = 0 the longitudinal correlation be equal to the transverse one, i.e.

$$\langle S_\ell^z S_g^z\rangle \underset{(T>T_c\,;\,B=0)}{=\!=} \langle S_\ell^x S_g^x\rangle = \langle S_\ell^y S_g^y\rangle \tag{5.14}$$

Thus the magnetic energy, <H>, takes the following simple form

$$\langle H \rangle \underset{(T \geq T_c\,;\,B=0)}{=} (-3/2) \sum_{f,m} I(f-m) \langle S_f^{+} S_m^{-} \rangle = N \cdot \mathcal{E} \qquad (5.15)a$$

Combining eqns. (3.38) and (5.2) we get the average magnetic energy per ion $\mathcal{E}$

$$\mathcal{E} \underset{(T \gg T_c\,;\,B=0)}{=} \left[ -3 \langle S^{z} \rangle \hbar / N \right] \cdot \sum_{K} \Phi(K) J(K) \qquad (5.15)b$$

Thus the magnetic energy is explicitly independent of the parameter γ'(T) and depends only the parameter A(T) (or X(T) ).

The procedure for the determination of A(T) and γ'(T) is now clear. We choose A(T) first to achieve agreement of eqn. $(5.15)_b$ with the result of the known high temperature expansion for the magnetic energy. γ'(T) is to be chosen next by fitting the susceptibility expansion of eqns. (5.6), (5.12) with the exact one.

Using eqn. (5.11), eqn. (5.15)b can be expanded in inverse powers of $\gamma$ to give

$$\mathcal{E} = (3/2) \bar{S} J(0) \hbar \sum_{r=1}^{\infty} (-1)^{r} \left[ \bar{S} (1 + X) / \hbar \gamma \right] \left[ F(r+1) - F(r) \right] \qquad (5.16)$$

where

$$\bar{S} = \underset{B=0}{\mathrm{Lt}} \left[ \langle S^{z} \rangle / t_0 \right] \qquad (5.17)$$

and F(r) is as in (5.13).

The sums F(r) depend only on the crystal structure and can be calculated easily for r>0. (see Appendix A.) The quantity $\bar{S}$ is proportional to the zero field susceptibility for which the exact expansion is known.

After a little algebra one can now determine the high temperature expansion for X(T) such that agreement between eqn. (5.15)b and the exact expansion for the magnetic energy is obtained. Below we give the results for the two leading terms in the expansions for X(T) and the related quantity A(T). (cf. eqns. 5.1 and 5.3)

$$X(\tau) = X_1/\tau + X_2/\tau^2 + \cdots \qquad (5.18)$$

where

$$X_1 = (-1/2z) \qquad (5.19)a$$

$$X_2 = (8/45z^2)\,s^2(s+1)^2 - \xi \cdot s(s+1) + (4/15z^2) \qquad (5.19)_b$$

$(10^3)\cdot\xi = 29.630$ f.c.c.; 52.778 b.c.c; 69.136 simple cubic

(5.19)$_c$

and where z denotes the number of nearest neighbors. Similarly, we have

$$A(\tau) = A_0 + A_1/\tau + \cdots \qquad (5.20)$$

$$A_0 = -9/8\,s^2(s+1)^2 \qquad (5.21)a$$

$$A_1 = (2/5z) - (3/s(s+1)) + \left[3/80z\,s^2(s+1)^2\right] \qquad (5.21)_b$$

$$\xi = 2.05,\ \text{f.c.c.};\ 2.45,\ \text{b.c.c.};\ 2.43,\ \text{s.c.} \qquad (5.21)_c$$

These expansions are sufficient to achieve agreement for the magnetic specific heat calculated from (5.15) to the order $(1/\gamma)^4$.

The susceptibility expansion (see eqns. 5.6 and 5.12) can now be similarly derived by using the above results for X(T). An interesting fact observed is that agreement of the leading three terms in the $1/\gamma$ expansion for $\chi$ is obtained without any assistance from the parameter $\gamma'(T)$. The fourth term is in agreement with the exact results only for large values of S.

This, once again, demonstrates that the simple mass operator type approximation, $\gamma' = 0$, is sufficient to describe the behavior of spin Green functions in the limit of large spins. For general S the $\gamma'(T)$ necessary to achieve agreement for the susceptibility up to and including the term proportional to $(1/\gamma)^4$, are as follows:

$$-10^3 \cdot \left[\gamma^3/S^2(S+1)^2\right] \cdot \gamma'(\gamma) = 14.4, \text{ f.c.c.}; \; 32.4, \text{ b.c.c.}; \; 57.6, \text{ s.c.} \qquad (5.23)$$

As long as the exact results for the susceptibility and the magnetic specific heat are available for comparison, the fore-going procedure can be used to determine X(T) and $\gamma'(T)$, and thus the dynamical spin correlation functions, to any order in $1/\gamma$.

## 6. Temperatures Close to Tc

Unlike the case of the very low or the very high temperatures, no rigorous expansions for thermodynamic functions exist for the range of intermediate temperatures in the vicinity of the Curie point. It is therefore difficult to construct a formalism which relies on the knowledge of these expansions for the determination of the arbitrary constants $\sigma(T)$ and $\gamma(T)$.

A number of results are, however, known for the transition temperature itself. For instance, the high temperature series extrapolation techniques[8,9] provide fairly accurate estimates for the Curie temperature, Tc, as a function of the exchange integral, I. Similarly, the magnetic energy at the critical point, $\langle H\rangle_c$, can also be estimated by these methods. It turns out that these estimates can be used to get information about the functions $\sigma(T)$ and $\gamma(T)$ in this temperature range. This investigation forms the contents of the present section.

In the absence of applied magnetic field, (e.i.) B = 0, the magnetization M(T), and therefore $\langle S^z\rangle$, is small and $\tilde{\Phi}$ (being, under these conditions, proportional to the inverse of $\langle S^z\rangle$) is large. Eqn. (3.34) can therefore again be expanded in inverse powers of $\tilde{\Phi}$. The most convenient expansion, it turns out, is the following (compare eqns. 5.4 and 5.5):

$$\langle S^z\rangle = \hbar\,\frac{2S(S+1)}{3\left[2\tilde{\Phi}+1+(C_1/\tilde{\Phi})-(C_1/2\tilde{\Phi}^2)+(C_2/\tilde{\Phi}^3)+\cdots\right]} \qquad (6.1)$$

where

$$C_1 = \left[2S(S+1)-3/2\right]/15 \qquad (6.2)$$

$$C_2 = \left[\frac{-12S^4-24S^3+126S^2+138S-96.75}{3150}\right] \qquad (6.3)$$

The energies $\tilde{E}_k$ can once again be written in the form (5.2). However, the parameters A(T) and X(T), defined in equations (5.1) and (5.3), will in general have different temperature dependence from that determined in the previous section. The appropriate expansion for equation (3.30) is now the following (compare (5.11) ):

$$\tilde{\Phi} = \Upsilon(T) - \tfrac{1}{2} + \tfrac{1}{2}\left[\frac{F(-1)}{\gamma} + \frac{\gamma}{3} - \gamma^3 \frac{F(3)}{45} + o(\gamma^5)\right] \qquad (6.4)$$

where

$$\gamma = \langle S^z \rangle \left[1 + X(T)\right] / kT \qquad (6.5)$$

Combining equations (6.1)-(6.5) we get:

$$\left[2\frac{S(S+1)}{3}k - \frac{F(-1)kT}{1+X(T)} - 2\Gamma(T)\right]$$
$$= \langle S^z \rangle^2 \left[\frac{1+X(T)}{3kT} + 2C_1 \frac{1+X(T)}{F(-1)kT}\right] + 0.\langle S^z \rangle^4 \qquad (6.6)$$

where we have used the notation:

$$\Gamma(T) = \langle S^z \rangle \cdot \Upsilon(T) \qquad (6.7)$$

The fummation F(-1) (see equation 5.13) is well known[30] and has the following values:

F(-1) = 1.34466, f.c.c.; 1.39320, b.c.c.; 1. 51638, s.c. (6.8)

The Curie temperature, $T_c$, is obtained by requiring that (since B = 0) $\langle S^z \rangle \rightarrow 0$ as T approached $T_c$ from below. Thus (6.6) gives:

$$T_c = \frac{\left[1 + X(T)\right] 2S(S+1)}{3F(-1)}\left[1 - 3\frac{\Gamma(T_c)}{kS(S+1)}\right] \qquad (6.9)$$

We find, empirically, that an extremely good fit of the results following from (6.9) with those known from references 8 and 9 can be obtained (refer Tables I, II, III) if we have:

$$\left[1+X(T_c)\right]\left\{1-\frac{3\Gamma(T_c)}{S(S+1)k}\right]=1+\left[\frac{F(-1)-1}{F(-1)}\right]\left(\frac{S-1}{3S}\right) \qquad (6.10)$$

Eqn. (6.10) contains two constants, X(Tc) and $\Gamma$(Tc). In order to specify these completely, we also need the expression for the critical value of the magnetic energy,$_z$ $\langle H\rangle_{T=Tc}$. Using eqns. $(5.15)_{a,b}$, expanding $\Phi_z(k)$ in powers of $\langle S\rangle$ (cf., eqn. 5.16), and proceeding to the limit $\langle S\rangle = o$ we get:

$$\frac{\mathcal{E}_c}{k_B T_c} \equiv \frac{\langle H\rangle_{T=T_c}}{N(k_B T_c)} = -\left(3/2\right)\left[\frac{F(-1)-1}{1+X(T_c)}\right] \qquad (6.11)$$

If the results for $\left(\mathcal{E}c/K_B Tc\right)$ were available for all values of S and z, equations (6.10) and (6.11) would determind $X(T_c)$ and $\Gamma(T_c)$. This, however, is not the case.

It is convenient, at this point, to establish a correspondence between the present work and that of ref. 23. Callen achieves a mass operator type solution which, in our notation, is equivalent to putting (at all temperatures):

$$A(T)\xrightarrow[\text{Callen}]{}\left(1/2\overset{2}{S}\right)\ ;\quad \gamma(T)\xrightarrow[\text{Callen}]{}0 \qquad (6.12)$$

At low temperatures, our results were obtained by choosing $A(T) = \left(1/2\overset{2}{S}\right)$(cf. eqns. 3.40 and 5.1) and $\gamma(T) \sim \left(T/_{Tc}\right)^{3S+3/2}$. Encouraged by this correspondence we postulate

$$A(T) \xrightarrow[T\to T_c]{} \left[1/2\overset{2}{S}\right] \qquad (6.13)$$

However, unlike Callen, we retain the anomalous contribution of the mass operator, $\gamma(T)$. Inserting eqn. (5.1) into (5.3) and putting T = Tc we now get

$$X(T_c)\left[1 + X(T_c)\right]/\gamma_c = \left(1/2\,\overset{z}{S}\right)\left[F(-1) - 1\right] \qquad (6.14)$$

The quantity $\left(\mathcal{E}/k_B T_c\right)$ can easily be calculated from eqns. (6.11) and (6.14) and the results are listed in Tables IV, V, and VI. We notice that these results are in good agreement with those available from reference 9 and are a considerable improvement on those following from the R.P.A.

The parameter $\Gamma$(Tc) is now completely determined and may easily be obtained with the use of equation (6.10) and X(Tc) given in Tables IV-VI.

We shall consider next the behavior of the parallel susceptibility at temperatures just beyond the Curie point. It is convenient here to recast equation (6.1) into the following form:

$$\tilde{\Phi} = \frac{\hbar S(S+1)}{3\langle \overset{z}{S} \rangle} - \frac{1}{2} - \frac{\langle \overset{z}{S} \rangle}{5\hbar S(S+1)}\left[S(S+1) - \frac{3}{4}\right] + 0\cdot\langle \overset{z}{S} \rangle^2 \qquad (6.15)$$

Let us put

$$\langle \overset{z}{S} \rangle = \chi' \cdot E_0 \qquad , \qquad (6.16)$$

where $\chi'$ is proportional to the zero field susceptibility $\chi$:

$$\chi' = \left(S^2 \hbar/\mu^2\right)\cdot\chi \qquad (6.17)$$

Introducing (6.17) into (6.15), expanding $\tilde{\Phi}$, and proceeding to the limit $E_0 = 0$ we get:

$$\frac{2}{k_B T}\left[\hbar\frac{S(S+1)}{3} - \Gamma(T)\right] = \frac{1}{N}\sum_k \frac{1}{\frac{1}{2\chi'} + \hbar J(0,k)\left[1 + X(T)\right]} \qquad (6.18)$$

Eqns. (6.9) and (6.10), which define the Curie temperature, Tc, can be rewritten as follows:

$$\frac{2}{k_B T_c}\left[\hbar\,\frac{S(S+1)}{3} - \Gamma(T_c)\right] = \frac{1}{\hbar[1+\chi(T_c)]}\,\frac{1}{N}\sum_{k}\frac{1}{J(0,k)} \qquad (6.19)$$

Subtracting (6.18) from (6.19) and carrying out the summations over k (see Appendix B) we find that just beyond Tc the susceptibility $\chi$ has the following form:

$$\chi = \Lambda \cdot \left\{(\beta_c - \beta) + \frac{3S(S+1)}{\hbar}\left[\beta\Gamma(T) - \beta_c\Gamma(T_c)\right]\right\}^{-2} \qquad (6.20)$$

where

$$\Lambda = \left(\frac{v}{a^3}\right)^2 \frac{9\mu^2}{\hbar^3 I^3 [1+\chi(T_c)]^3 \cdot 128\pi^2 \hbar^4 S^4 (S+1)^2} \qquad (6.21)$$

If we follow the R.P.A. and the Callen assumption and put $\Gamma(T) = 0$, or alternatively, if we make the approximation that in the vininity of Tc, $\Gamma(T)$ is equal to $\Gamma(T_c)$, we find that (6.20) leads to the following

$$\chi \underset{(R.P.A.;\,Callen)}{=\!=} \Lambda \cdot (\beta_c - \beta)^{-2} \qquad (6.22)$$

This result is similar to that of the spherical model[31] and was earlier obtained for the spin 1/2 case by Englert[24] and by Kawasaki and Mori[32]. Within the above approximation we can also find the magnetization just below the Curie temperature from eqn. (6.6).

$$\langle S\rangle^2 \underset{(R.P.A.;\,Callen)}{=\!=} P\cdot(1 - T/T_c) + O\cdot(1 - T/T_c)^2 \qquad (6.23)$$

where

$$P = \left[\frac{k T_c\ F(-1)}{1+X(T_c)}\right]^2 \left[\frac{3}{F(-1)+6C_1}\right] \qquad (6.24)$$

(compare ref. 21).

The above result is similar in form to that of the molecular field approximation (M.F.A.)

$$\langle \overset{z}{S} \rangle^2 \underset{\text{M.F.A.}}{=} (5/3)\frac{S^2(S+1)^2}{(S^2+S+1/2)}\left(1-T/\Theta_c\right)+O\left(1-T/\Theta_c\right)^2 \qquad (6.25)$$

where $\Theta_c$ is the Curie temperature in the M.F.A.

Recently Domb and Sykes$^9$ have reported, after a painstaking examination of the systematics of the extrapolation of high temperature series, that just beyond Tc the zero field susceptibility, $\chi$, obeys a relation of the form

$$\chi \underset{(\text{Domb \& Sykes})}{=} A\cdot(\beta_c-\beta)^{-4/3} \qquad (6.26)$$

where A is a constant independent of the temperature. It is therefore clear that the approximation $\Gamma(T) = \Gamma(Tc)$, which led to the result (6.22), is unsatisfactory. In order to secure agreement of our result of eqn. (6.20) and the above result, (6.26), the function $\Gamma(T)$, in the immediate vicinity of Tc, has to satisfy the following relation

$$\left\{(\beta_c-\beta)+\frac{3S(S+1)}{k}\left[\beta\Gamma(T)-\beta_c\Gamma(T_c)\right]\right\}^2 \qquad (6.27)_a$$

$$= C^2\cdot(\beta_c-\beta)^{4/3} + O(\beta_c-\beta)^n$$

which is equivalent to putting:

$$\Gamma(T) = \frac{\pm Ck}{3S(S+1)\beta}(\beta_c-\beta)^{2/3}+\left(\beta_c/\beta\right)\Gamma(T_c)+o(\beta_c-\beta)^{n/2} \qquad (6.27)_b$$

where C is a constant independent of the temperature and n > 4/3.

Having thus inferred the form of Γ(T) close to Tc, we utilize it in the determination of the magnetization just below the Curie temperature. Inserting eqn. $(6.27)_b$ into (6.6) we get:

$$\langle S^z\rangle^2 = \mp\frac{2C\,F(-1)\,(\beta-\beta_c)^{2/3}+o(\beta-\beta_c)^{n/2}}{S(S+1)\,\beta\,J(0)\,[1+X(T)]\,[F(-1)+6C_1]} \qquad (6.28)$$

Using eqns. (3.35), (3.38), (5.3), (6.4) and (6.13), X(T) can easily be expanded in powers of $\langle S^z\rangle$ and we get:

$$X(T)[1+X(T)] = \left(\Upsilon/2S^2\right)\left[\{F(-1)-1\} - \frac{\langle S^z\rangle^2\{1+X(T)\}^2}{k^2\,\Upsilon^2\,\gamma}\right] \qquad (6.29)$$

(compare eqn. 6.14).

and therefore

$$X(T) = X(T_c) - \frac{\langle S^z\rangle^2[1+X(T)]^2}{4k^2\Upsilon_c^2\,\gamma\sqrt{1+2(\Upsilon_c/S^2)[F(-1)-1]}} + o(1-T/T_c) \qquad (6.30)$$

Thus to the leading power in the difference $(1-T/_{Tc})$ we may replace X(T) by X(Tc) in (6.28). Eqn. (6.28) now easily leads to the result

$$\langle S^z\rangle = K\cdot(\beta-\beta_c)^{1/3} + o(\beta-\beta_c)^{n/4} \qquad (6.31)$$

where K is to be a real quantity and is related to the constant C of eqn. $(6.27)_b$ by the relation:

$$K = \left[\frac{2C\,F(-1)}{S(S+1)\,\beta_c\,J(0)\,[1+X(T_c)]\,[F(-1)+6C_1]}\right]^{1/2} \qquad (6.32)$$

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# APPENDIX A

The inverse lattice sums in equation (5.13) have to be restricted to the first Brillouin zone. It is convenient therefore to transform the function J(o,k)/J(o) to a system of coordinates such that the integration limits are simplified (the boundary of the Brillouin zone in the cases of b.c.c. and f.c.c. lattices is somewhat complicated being respecively similar in structure to f.c.c. and b.c.c. lattices). This is done by choosing the coordinate axes in the direction of the basis-vectors of the reciprocal lattice space, (for the basis vectors see reference 33). It turns out that

$$\begin{aligned} J(o,k)/J(o) &\underset{\text{s.c.}}{\equiv} \tfrac{1}{3}\left[3 - \cos k_1 - \cos k_2 - \cos k_3\right] \\ &\underset{\text{b.c.c.}}{\equiv} (1/4)\left[4 - \cos k_1 - \cos k_2 - \cos k_3 - \cos(k_1+k_2+k_3)\right] \\ &\underset{\text{f.c.c.}}{\equiv} (1/6)\left[6 - \cos k_1 - \cos k_2 - \cos k_3 - \cos(k_1-k_2) - \cos(k_2-k_3) - \cos(k_3-k_4)\right] \end{aligned} \qquad (A.1)$$

where, since the Jacobian is unity for the above transformation, we use the following prescription for changing sums into integrals:

$$\frac{1}{N}\sum_{k}(\cdots) \longrightarrow (\text{[illegible]}) \iiint_{-\pi}^{+\pi} dk_1\cdot dk_2\cdot dk_3\cdot(\cdots) \qquad (A.2)$$

The sums F(r) of equation (5.13) are now elementary. The results for the first several cases are tabulated below:

| F(r) | f.c.c. | b.c.c. | s.c. |
|---|---|---|---|
| r = 1 | 1.000 000 | 1.000 000 | 1.000 000 |
| r = 2 | 1.083 333 | 1.125 000 | 1.166 667 |
| r = 3 | 1.222 222 | 1.375 000 | 1.500 000 |
| r = 4 | 1.414 931 | 1.802 734 | 2.069 444 |
| r = 5 | 1.668 403 | 2.513 672 | 3.013 889 |
| r = 6 | 1.995 049 | 3.696 533 | 4.581 533 |
| r = 7 | 2.412 423 | 5.684 326 | 7.209 619 |
| r = 8 | 2.943 934 | 9.066 343 | 11.670 664 |
| r = 9 | 3.620 184 | 14.892 007 | 19.338 445 |

## APPENDIX B

In the vicinity of Tc, the difference between X(T) and X(Tc) is of the order $\langle \overset{z}{S} \rangle^2$ (see Eqn. (6.3[illegible]). Therefore, subtracting eqn. (6.20) from eqn. (6.21) we get for temperatures just above the Curie temperature

$$[2\hbar S(S+1)/3]\left[(\beta_c-\beta)+\frac{3S(S+1)}{\hbar k_B}\left\{\left(\Gamma(T)/T\right)-\left(\Gamma(T_c)/T_c\right)\right\}\right]$$

$$\cong \frac{1}{2\chi'\hbar^2[1+X(T_c)]^2}\cdot\frac{1}{N}\sum_k\frac{1}{[V+J(0,k)]J(0,k)} \qquad \text{(B-1)}$$

where

$$V = \frac{1}{2\chi'[1+X(T)]\hbar} \qquad \text{(B-2)}$$

The inverse lattice summation in (B-1) can be approximately performed as follows. As the susceptibility $\chi$ is large, $V\hbar^2$ is small compared with unity. The dominant contribution to the summation therefore comes from small values of k. As a rough approximation we may therefore extend the integration limits to the whole of the k-space and also use the long wave length approximation for J(o,k), i.e.

$$J(0,k) = I(k^2a^2) + O(ka)^4 \qquad \text{(B-3)}$$

where the nearest neighbor distance has been taken to be $[6(a^2/z)]^{1/2}$ (z is the coordination number). In this manner we have:

$$\frac{1}{N}\sum_k\frac{1}{[V+J(0,k)]J(0,k)} \cong \left(\frac{v}{2\pi^2 I a^2}\right)\int_0^\infty \frac{dk}{V+Ik^2} \qquad \text{(B-4)}$$

where v is the volume per ion ($v = a^3$, s.c.; $a^3/2$, b.c.c.; $a^3/4$, f.c.c.).

Therefore, eqns. (B—1) and (6.20) give

$$\chi = \left[\frac{9a^2(v/a^3)^2}{[\hbar I(1+X(T_c))]^3\cdot 128\,\pi^2\hbar^4 S^4(S+1)^4}\right]\left[(\beta_c-\beta)+\frac{3S(S+1)}{\hbar}\{\beta\Gamma(T)-\beta_c\Gamma(T_c)\}\right]^{-2} \qquad \text{(B-5)}$$

## TABLE I THE FACE-CENTERED CUBIC LATTICE

| S | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Rushbrooke and Wood | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Ours | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ R.P.A. | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Callen | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Domb and Sykes |
|---|---|---|---|---|---|
| 1/2 | 4.15 | 4.08 | 4.46 | 5.6 | 4.07 |
| 1 | 12.0 | 11.90 | 11.90 | 13.9 | 12.0 |
| 3/2 | 23.0 | 22.95 | 22.31 | 25.5 | - |
| 2 | 37.25 | 37.23 | 35.70 | 40.3 | - |
| 5/2 | 54.8 | 54.73 | 52.06 | 58.3 | - |
| 3 | 75.4 | 75.46 | 71.39 | 81.3 | - |

$$\lim_{S\to\infty}\left(\frac{3K_BT_c}{2J(0)S(S+1)}\right)$$

$= 0.788$, Rushbrooke and Wood

$= 0.807$, (ours and Callen's)

$\doteq 0.798$, Domb and Sykes

$= 0.744$, Random phase Approx.

## TABLE II THE BODY-CENTERED CUBIC LATTICE

| S | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Rushbrooke and Wood | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Ours | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ R.P.A. | $\frac{K_BT_c}{I\hbar^2}$ Callen |
|---|---|---|---|---|
| 1/2 | 2.60 | 2.60 | 2.87 | 3.7 |
| 1 | 7.55 | 7.66 | 7.66 | 9.1 |
| 3/2 | 14.7 | 14.81 | 14.36 | 16.6 |
| 2 | 23.9 | 24.05 | 22.97 | 26.2 |
| 5/2 | 35.2 | 35.39 | 33.50 | 37.9 |
| 3 | 48.5 | 48.82 | 45.94 | 51.7 |

$$\lim_{S\to\infty}\left(\frac{3K_BT_c}{2J(0)S(S+1)}\right)$$

= 0.752, Rushbrooke and Wood

= 0.785, (Ours and Callen's)

= 0.718, Random phase Approx.

## TABLE III THE SIMPLE CUBIC LATTICE

| S | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Rushbrooke and Wood | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Ours | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ R.P.A. | $\left(\frac{K_BT_c}{I\hbar^2}\right)$ Callen |
|---|---|---|---|---|
| 1/2 | 1.7 | 1.75 | 1.98 | 2.7 |
| 1 | 5.25 | 5.28 | 5.28 | 6.5 |
| 3/2 | 10.2 | 10.27 | 9.89 | 11.8 |
| 2 | 16.65 | 16.73 | 15.83 | 18.5 |
| 5/2 | 24.75 | 24.65 | 23.08 | 26.8 |
| 3 | 33.9 | 34.04 | 31.65 | 36.4 |

$$\lim_{S\to\infty}\frac{3K_BT_c}{2J(0)S(S+1)}$$

= 0.716, Rushbrooke and Wood

= 0.734, (Ours and Callen's)

= 0.659, Random phase approx.

## TABLE IV THE FACE-CENTERED LATTICE

$$\left(\frac{-E_c}{K_B T_c}\right)_{\text{R.P.A.}} = 0.517, \text{ for all } S$$

| S | $\left(\frac{-E_c}{K_B T_c}\right)$ ours | $\left(\frac{-E_c}{K_B T_c}\right)$ Domb and Sykes | X(Tc) |
|---|---|---|---|
| 1/2 | 0.433 | 0.439 | 0.195 |
| 1 | 0.450 | 0.449 | 0.150 |
| 3/2 | 0.458 | - | 0.130 |
| 2 | 0.463 | - | 0.116 |
| 5/2 | 0.465 | - | 0.113 |
| 3 | 0.467 | - | 0.108 |
| $\infty$ | 0.476 | 0.474 | 0.085 |

## TABLE V THE BODY-CENTERED CUBIC LATTICE

$$\left(\frac{-E_c}{K_B T_c}\right)_{\text{R.P.A.}} = 0.590, \text{ for all } S.$$

| S | $\left(-E_c/K_B T_c\right)$ | X(Tc) |
|---|---|---|
| 1/2 | 0.487 | 0.211 |
| 1 | 0.508 | 0.160 |
| 3/2 | 0.517 | 0.141 |
| 2 | 0.522 | 0.130 |
| 5/2 | 0.525 | 0.123 |
| 3 | 0.528 | 0.118 |
| $\infty$ | 0.539 | 0.094 |

## TABLE VI THE SIMPLE CUBIC LATTICE

$$\left(-E_c/K_BT_c\right)_{R.P.A.} = 0.775$$

| S | $\left(-E_c/K_BT_c\right)$ | X(Tc) |
|---|---|---|
| 1/2 | 0.626 | 0.237 |
| 1 | 0.651 | 0.190 |
| 3/2 | 0.664 | 0.167 |
| 2 | 0.671 | 0.155 |
| 5/2 | 0.674 | 0.149 |
| 3 | 0.678 | 0.142 |
| $\infty$ | 0.695 | 0.114 |

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