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MAGNETIZATION AND SUSCEPTIBILITY OF THE HEISENBERG FERROMAGNET

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MAGNETIZATION AND SUSCEPTIBILITY OF THE HEISENBERG FERROMAGNET

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SUMMARY

The anisotropic Heisenberg ferromagnet developed by Flax and Raich is reevaluated to include an applied magnetic field. The present calculation is restricted to the case of an isotropic exchange interaction, and the random phase approximation is used. The present theory predicts a broadening of the transition region with increasing magnetic field. Thermodynamic quantities, such as magnetization and susceptibility, are studied near the Curie point.

INTRODUCTION

According to the Weiss theory a ferromagnetic body possesses an internal field which is proportional to the magnetization. As the temperature of a ferromagnet is increased, the magnetization decreases until, at a temperature known as the Curie temperature T_c, the sample becomes paramagnetic. This change from ferromagnetic to paramagnetic is referred to as the ferromagnetic phase transition. Experiments and theory have shown that electrical, mechanical, and many thermodynamic properties of a material are altered when the material undergoes a phase transition.

One way of describing the ferromagnetic phase transition is to look upon it as an order-disorder transition in the material. The property of order in a system describes the correlation between the properties of two atoms, even when the atoms are widely separated in space. The spontaneous magnetization present in a ferromagnet is due to the correlation between the spins of the atoms. Above the Curie temperature the long-range order is gone if there is no applied field and the system is paramagnetic.

At absolute zero the spontaneous magnetization $\langle S^z \rangle$ is equal to its maximum value, while at any other temperature T with no applied field the value depends on the ratio T/T_c. (Symbols are defined in the appendix.)

The effect of the external magnetic field is twofold: (1) the magnetization is increased somewhat above its zero-field value, and (2) the transition region is broadened.

Even though temperature tends to destroy this alinement, the field causes some ordering to be present. Thus, instead of an abrupt disorder at the critical temperature there is a tail due to some ordering.

This report investigates some thermodynamic properties of the Heisenberg ferromagnet is an external magnetic field. The objective is to obtain expressions for some of the thermodynamic quantities that are needed to evaluate the use of ferromagnetism for a refrigeration system (refs. 1 and 2). The thermodynamic quantities studied here are the magnetization and susceptibility. The statistical method used to determine the thermodynamics of the system is the double-time Green's function. Generalized Watson sums arise in the theory of magnetism and other disorder phenomena. These sums are evaluated analytically in order to calculate the thermodynamic quantities as a function of temperature. Previous evaluations of these sums have been based on series expansions and are valid only over limited temperature regions - usually away from the critical temperature. The methods used in references 3 to 7 and in this work enable one to obtain analytical expressions for the thermodynamic quantities without using series expansions.

HEISENBERG FERROMAGNET

The Heisenberg model assumes that the magnetic electrons are in states localized about the lattice sites with an exchange interaction taking place between the electrons. The model does not take into account an itinerant-electron picture. The model is thought to be very good for insulating ferromagnets such as europium oxide and poor for conductors such as iron and nickel. However, it appears that this model gives better results for conductors at low temperatures than those calculated from a band theory approach as shown by Argyle, Charap, and Pugh (ref. 8). The success of the Heisenberg model is somewhat surprising since it does not take into account the spreading of the electronic energy levels into bands.

The Heisenberg ferromagnet with spin 1/2 was analyzed by Bogolyubov and Tyablikov (ref. 9) using the techniques of double-time temperature-dependent Green functions. A convenient review of Green functions and Tyablikov's application of them to ferromagnetism is given by D. N. Zubarev in reference 10 and in references cited there.

The Hamiltonian for the Heisenberg model is

$$H = -g\mu_B H_0 \sum S_i^z - \sum_{i,j} J_{ij}\left[S_i^z S_j^z + \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right)\right] \tag{1}$$

where

$$S_j^{\pm} = S_j^x \pm iS_j^y \tag{2}$$

μ_B is the Bohr magneton, g is the Lande g factor, H_0 is the applied magnetic field which is assumed to be in the -z direction, S_i is the spin operator for a spin at site i, J_{ij} is the exchange interaction between spins on sites i and j, and the sum is carried over all sites in the crystal. The exchange interaction is assumed to be a function of the distance between sites only. The self-exchange terms such as J_{ii} and J_{jj} vanish.

Since the thermodynamic quantities of interest are the magnetization and susceptibility of the system, one is interested in evaluating correlation functions of the form $\langle S_m^- S_g^+ \rangle$ and hence of the Green's function $\langle\langle S_g^+; S_m^- \rangle\rangle$.

Using the Hamiltonian (1) together with the known spin commutation relations

$$\left.\begin{aligned} \left[S_i^+, S_j\right] &= 2S_j^z \delta_{ij} \\ \left[S_i^{\pm}, S_j^z\right] &= S_j^{\pm} \delta_{ij} \end{aligned}\right\} \tag{3}$$

one can write the equation of motion (see ref. 3 or 4) in the form

$$(E - \mu_B g H_0)\langle\langle S_g^+; S_m^- \rangle\rangle = \frac{1}{\pi} \langle S_g^z \rangle \delta_{gm/\pi} + 2 \sum J_{gf} \langle\langle S_g^+ S_f^z; S_m^- \rangle\rangle - \langle\langle S_f^+ S_g^z; S_m \rangle\rangle \tag{4}$$

For a system of interacting spins, the second and third terms on the right side of equation (4) contain Green's function with three operators and are called higher order Green's functions. The equation of motion for one of these higher order Green's functions involves the next higher order Green's function. In this way an infinite hierarchy of equations is generated. Thus, the exact treatment of the equations of motion involves the solution of an infinite set of coupled equations for an infinite number of Green's functions. To obtain a closed solution for a Green's function, this hierachy of equations must be truncated at some point. The procedure usually adopted is to make a so-called decoupling approximation in which the higher order Green's functions on the right side of equation (4) are expressed in terms of lower order Green's functions. By doing this, an explicit approximate solution of equation (4) for $\langle\langle S_g^+; S_m^- \rangle\rangle$ can be found.

RANDOM PHASE APPROXIMATION

The essential approximation of the random phase approximation (RPA) method of Bogolyubov and Tyablikov (ref. 9), Tahir-Kheli and ter Haar (ref. 11), Flax and Raich (refs. 3 to 7), and those cited in references 3 to 7 consists of ignoring the correlations between $\langle S_g^z \rangle$ and the other operators S_f^+ and S_m^- and replacing the operator S_g by the statistical average $\langle S^z \rangle$, that is,

$$\langle\langle S_g^z S_f^+; S_m^- \rangle\rangle \longrightarrow \langle S^z \rangle \langle\langle S_f^+; S_m^- \rangle\rangle \tag{5}$$

We may note that $\langle S_g^z \rangle = \langle S^z \rangle$ is independent of the site g on account of the translational invariance of the lattice. This method of truncating the higher order Green functions is certainly an ad hoc one since $S_g^z - \langle S^z \rangle$ is not necessarily small. At low temperatures, one may justify the use of this decoupling procedure since the spins are nearly completely alined. In the nearly alined state, the deviation of the spin (z component) at any one site from the average value would be small, and thus for a first approximation one might neglect $S_g^z - \langle S^z \rangle$. However, the low-temperature expansion of the resulting expression for the magnetization contains a T^3 term in conflict with the careful spin wave analysis of Dyson.

At high temperatures the spins are more disordered, and the RPA approximation leads to further discrepancies:

(1) The magnetization obeys a square root law in temperature in the vicinity of the Curie point as opposed to the cube root law behavior suggested by experiment.

(2) The specific heat is zero above T_c because of the abrupt disappearance of long-range order to contrast to experimental observation in which a nonzero value persists because of local or short-range order.

However, the random phase approximation shares these defects with all theories based on Landau's treatment of second-order phase transitions.

Upon applying the RPA to the equation of motion (4), one obtains

$$\left[E - \mu_B g H_0 - 2\langle S^z \rangle J(0)\right] A_{gm} = \frac{1}{\pi} \langle S_g^z \rangle \delta_{gm} - 2\langle S^z \rangle \sum_F J_{gf} A_{fm} \tag{6}$$

where for brevity

$$A_{gm} = \langle\langle S_g^+; S_m^- \rangle\rangle \tag{7}$$

Translational invariance allows the use of a Fourier transformation to the reciprocal lattice; that is,

$$A_{gm} = \frac{1}{N} \sum_{k} \exp[ik \cdot (g - m)] A_k \tag{8}$$

and

$$J(k) = \sum_{f} J_{gf} \exp[ik \cdot (f - g)] \tag{9}$$

where N is the total number of sites in the lattice and the sums are restricted to the first Brillouin zone. It was shown in references 3 to 5 and 7 that the Green's function for this model can be written in the form

$$A_k = \frac{\langle S^z \rangle}{\pi [E - E(k)]} \tag{10}$$

where the elementary excitation energy $E(k)$ is

$$E(k) = \mu_B g H_0 + 2\langle S^z \rangle J(0) \left[1 - \frac{J(k)}{J(0)}\right] \tag{11}$$

MAGNETIZATION

Application of a magnetic field is assumed sufficient to orient the net magnetization vector along the direction of the field. The field also introduces additional long-range order. The long-range order is thus due to the internal field and to the applied magnetic field. The average z component of the spin is a measure of this long-range order. The thermal average $\langle S^z \rangle$ is proportional to the magnetization.

To calculate the magnetization $\langle S^z \rangle$, one uses the commutation relation of equation (3) as well as

$$\vec{S} \cdot \vec{S} = S^- S^+ + S^z + (S^z)^2 \tag{12}$$

which fixes the magnitude of the spin. The average value of the spin correlation is

$$\langle S^- S^+ \rangle = S(S+1) - \langle S^Z \rangle - (S^Z)^2 \tag{13}$$

For spin 1/2 equation (13) reduces to

$$\langle S^- S^+ \rangle = \frac{1}{2} - \langle S^Z \rangle \tag{14}$$

In this report only spin 1/2 is considered. With the use of equation (10) one can calculate the correlation function $\langle S_m^- S_g^+ \rangle$ from the relation

$$\langle S_m^-(t') S_g^+(t) \rangle = \lim_{\xi \to 0} i \int_{-\infty}^{\infty} \frac{\left[A_{gm}(E + i\xi) - A_{gm}(E - i\xi) \right] e^{iE} \, dE^{(t-t')}}{e^{E(k)\beta} - 1} \tag{15}$$

Equations (10), (14), and (15) enable us to obtain an expression for magnetization.

The calculation of the correlation function follows directly from equation (15) in the limit $t = t'$. From the δ function representation

$$\delta(x) = \lim_{\xi \to 0} \frac{1}{2\pi i} \left(\frac{1}{x - i\xi} - \frac{1}{x + i\xi} \right) \tag{16}$$

it follows that

$$\langle S^- S^+ \rangle = 2 \langle S^Z \rangle \varphi \tag{17}$$

where

$$\varphi = \frac{1}{N} \sum_k \frac{1}{\left[e^{E(k)\beta} - 1 \right]} \tag{18}$$

in which $\beta = 1/(k_B T)$ and $E(k)$ is given by equation (11). Using equations (14) and (17), one finds that

$$\langle S^- S^+ \rangle = 2 \langle S^Z \rangle \varphi = \frac{1}{2} - \langle S^Z \rangle \tag{19}$$

The expression for $\langle S^z \rangle$ then becomes

$$\langle S^z \rangle = \frac{\frac{1}{2}}{1 + 2\varphi} \tag{20}$$

and it is easily shown that

$$\varphi = \frac{1}{2N} \sum_k \left[\frac{\coth \beta E(k)}{2} - 1 \right] \tag{21}$$

ANALYTICAL SOLUTION FOR MAGNETIZATION

To calculate the magnetization as a function of temperature, the sums in equation (21) must be evaluated over all values of k in the first Brillouin zone of the appropriate lattice. Except at the very low and high temperature limits, numerical methods are usually used. However, such numerical solutions are difficult. One of the purposes of this report is to show that analytical solutions of equation (21) are possible if the methods of references 3 to 6 are used.

Consider only crystals with cubic symmetry such as body-centered cubic (bcc). For such a lattice one can replace the sum which appears in equation (21) by an integral

$$\frac{1}{N} \sum_k \rightarrow \frac{1}{\pi^3} \int_0^\pi \int_0^\pi \int_0^\pi dx\, dy\, dz \tag{22}$$

where for a bcc lattice

$$\left.\begin{aligned} x &= \frac{k_x a}{2} \\ y &= \frac{k_y a}{2} \\ z &= \frac{k_z a}{2} \end{aligned}\right\} \tag{23}$$

where a is the length of the cell edge. Rewrite equation (20) in the form

$$\langle S^z \rangle = \frac{1}{2\Delta} \tag{24}$$

where

$$\Delta = \frac{1}{N} \sum_k \frac{\coth \beta E(k)}{2} \tag{25}$$

Using equation (22), one can write equation (25) as

$$\Delta = \frac{1}{\pi^3} \int_0^\pi \int_0^\pi \int_0^\pi \frac{\coth \beta E(k)}{2} \, dx \, dy \, dz \tag{26}$$

For the case of the bcc lattice,

$$\frac{J(k)}{J(0)} = \left(\frac{\cos k_x a}{2}\right)\left(\frac{\cos k_y a}{2}\right)\left(\frac{\cos k_z a}{2}\right) \tag{27}$$

Using the identity

$$\coth \pi t = \frac{1}{\pi t} + \frac{2}{\pi} \sum_{R=1}^{\infty} \frac{t}{R^2 + t^2} \tag{28}$$

and the techniques developed by Flax (ref. 5) and Flax and Raich (refs. 3 and 4), one can obtain

$$\Delta = \frac{1}{P}\left[\frac{2K(k)}{\pi}\right]^2 + \coth P - \frac{1}{P} + \frac{Q^2}{8}\left[\operatorname{csch}^2 P \coth P - \left(\frac{1}{P}\right)^3\right] \tag{29}$$

where

$$P = \frac{\alpha}{2} + Q = Q + \frac{\alpha}{2} \tag{30}$$

$$\alpha = \frac{\mu_B g H_0}{J(0)\tau} = \frac{H'}{\tau} \tag{31}$$

$$Q = \frac{\langle S^z \rangle}{\tau} \tag{32}$$

$$\tau = \frac{k_B T}{J(0)} \tag{33}$$

K(k) is a complete elliptic integral of the first kind, and

$$k^2 = \frac{1}{2}\left[1 - \sqrt{1 - \left(\frac{Q}{P}\right)^2}\right] \tag{34}$$

The solution of equation (29) gives Δ as a function of temperature, magnetic field, and magnetization. Hence, when equation (29) is substituted into equation (24), the magnetization is obtained.

In the absence of an applied magnetic field the random phase approximation predicts that the magnetization shows a second-order transition which has a critical temperature with the value

$$T_c = \frac{0.3589\ J(0)}{k_b} \qquad (H_0 = 0) \tag{35}$$

It is then feasible to relate H_0 and H' as follows:

$$H' = \frac{\mu_B g H_0}{J(0)} = \frac{0.3588\ \mu_B g H_0}{k_b T_c}$$

or

$$H_0 = \frac{k_b T_c H'}{0.3588\ \mu_B} \tag{36}$$

Substituting the appropriate values of the Boltzmann value, Lande g factor, and the Bohr magneton, one obtains

$$H_0 = \frac{1.143\times10^4\ T_c H'}{2} = 5.71\times10^3\ T_c H' \tag{37}$$

Although the theory is for spin 1/2, and neither iron nor nickel has this spin, as examples of the actual values of field corresponding to the nondimensional field H', consider the following. Using the values for the critical temperature of iron (1043 K) and nickel (631 K), one can write

$$\left.\begin{aligned} H_0 &= 596\times10^4\ H' \qquad \text{(iron)} \\ H_0 &= 365.6\times10^4\ H' \qquad \text{(nickel)} \end{aligned}\right\} \tag{38}$$

The values used for H' are 5×10^{-4} and 5×10^{-3}, which correspond in the case of iron, to 2480 and 24 800 gauss, respectively. In the case of nickel these values correspond to 1802 and 18 020 gauss, respectively.

Figure 1 shows a plot of $\langle S^Z \rangle$ as a function of temperature for several values of H' for a bcc lattice. When H' equals zero, there are no solutions for $\langle S^Z \rangle$ above T_c. Thus, at this point the long-range order disappears, and above this point the present theory predicts complete disorder of the spins. When H' is not equal to zero, the ferromagnetic transition occurs not at a single temperature but over a range of temperatures which forms a "Curie region," so that the transition is broadened and results in the appearance of a "tail."

The reason for the tail is that the spins feel the effect of an applied magnetic field and the long-range order persists. The higher the field, the greater the broadening of the transition.

Figure 2 shows a plot of $\langle S^Z \rangle$ as a function of H' for several values of τ for a bcc lattice. As can be seen from the figure, in the immediate neighborhood of the zero-field Curie point ($\tau_c = 0.359$) the magnetization has a nonlinear dependence on the field for all values of the field strength. When the temperature is raised, the nonlinearity remains in the weak-field region, but in the strong field there is a linear section of the magnetization curve. As one increases the temperature, the linear section of the curve extends farther toward weaker fields, and finally the magnetization curve goes entirely into a

straight line slope. The linear sections are simply Curie-law behavior. Experimental observations (ref. 12) show linear and nonlinear regions corresponding to those just described.

MAGNETIC SUSCEPTIBILITY

Let the magnetic susceptibility χ be defined as

$$\chi = \frac{\langle S^z \rangle}{H'} = \frac{Q}{\alpha} \tag{39}$$

Using equations (24) to (34) in conjunction with equation (39), one obtains

$$\chi = \frac{1}{2\alpha \Delta'_\tau} \tag{40}$$

where

$$\Delta' = \frac{1}{P}\left[\frac{2K(k_1)}{\pi}\right]^2 + \coth P - \frac{1}{P} + \frac{\alpha^2\chi^2}{8}\left[\operatorname{csch}^2 P \coth P - \left(\frac{1}{P}\right)^3\right] \tag{41}$$

$$P = \frac{\alpha}{2}(1 + 2\chi) \tag{42}$$

and

$$K_1^2 = \frac{1}{2}\left[1 - \sqrt{1 - \left(\frac{2\chi}{1 + 2\chi}\right)^2}\right] \tag{43}$$

For a given ferromagnetic material the spontaneous magnetization can occur only below a critical temperature T_c. Well above T_c such materials are found to behave paramagnetically, and to have a well-defined susceptibility which follows the Curie-Weiss law, namely,

$$\chi = \frac{C}{T - \theta} \tag{44}$$

in which C is the Curie constant and θ is called the paramagnetic Curie temperature. Experimental measurements (ref. 12) show, however, that in the immediate neighborhood of the Curie point, the dependence of $1/\chi$ on τ above T_c is not linear.

A plot of $1/\chi$ as a function of τ from equation (40) is shown in figure 3. As required the curve becomes linear at temperatures sufficiently far above the Curie point. The nonlinearity in the neighborhood of the Curie point agrees qualitatively with experiment (ref. 12). Previous theories such as molecular field theories have been unable to demonstrate this nonlinear behavior.

CONCLUSIONS

The thermodynamic properties of the Heisenberg ferromagnet in a magnetic field were investigated theoretically by using a random phase approximation. Expressions were derived from which the thermodynamic parameters can be calculated, and the following conclusions were drawn:

1. The random phase approximation predicts that in the presence of a magnetic field there is a broadening of the ferromagnetic transition which results in the appearance of a tail in the magnetization-temperature curve. A higher magnetic field increases the broadening of the transition.

2. There is a nonlinear dependence of the magnetization upon field at the Curie point. When the temperature is raised, the nonlinearity remains in the weak-field region, but in the strong-field region the magnetization becomes linear with field, in agreement with the Curie law.

3. Analytical expressions for magnetization and susceptibility are obtained which are valid for all temperatures. The analysis predicts that well above the Curie point the susceptibility follows the Curie-Weiss law. In the immediate neighborhood of the Curie point, the dependence of the reciprocal of susceptibility $1/\chi$ on temperature is nonlinear. The results are in qualitative agreement with experimental data.

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APPENDIX - SYMBOLS

C	Curie constant
E_k	excitation energy
g	Lande g factor
H	Hamiltonian
H_0	applied magnetic field
H'	$\mu_B g H_0 / J(0)$
i, j	lattice sites
$J(k)$	defined by eq. (9)
$J_{i,j}$	exchange interaction
$K(k)$	elliptic integral of first kind
k_b	Boltzmann's constant
P	$(\alpha/2) + Q$
Q	$\langle S^Z \rangle / \tau$
R	sum index
S_i	spin operator at site i
$\langle S^Z \rangle$	magnetization
T	temperature
T_c	Curie temperature
α	defined by eq. (31)
β	$1/(kT)$
τ	reduced temperature
χ	$\langle S^Z \rangle / H' = Q/\alpha$
$\langle\langle\ \rangle\rangle$	Fourier transform of Green's function
$\langle\ \rangle$	conical average of operator

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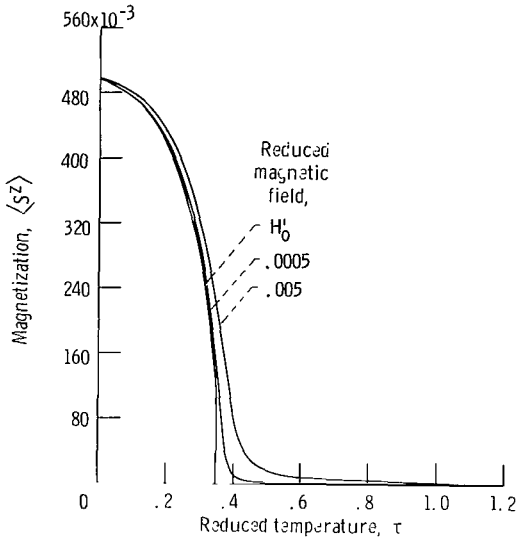

Figure 1. - Magnetization as function of reduced temperature.

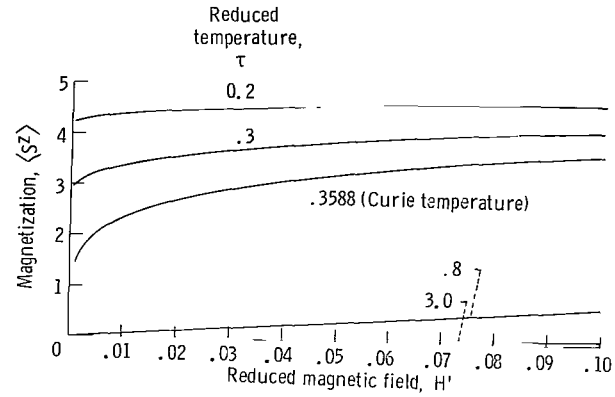

Figure 2. - Magnetization as function of reduced magnetic field.

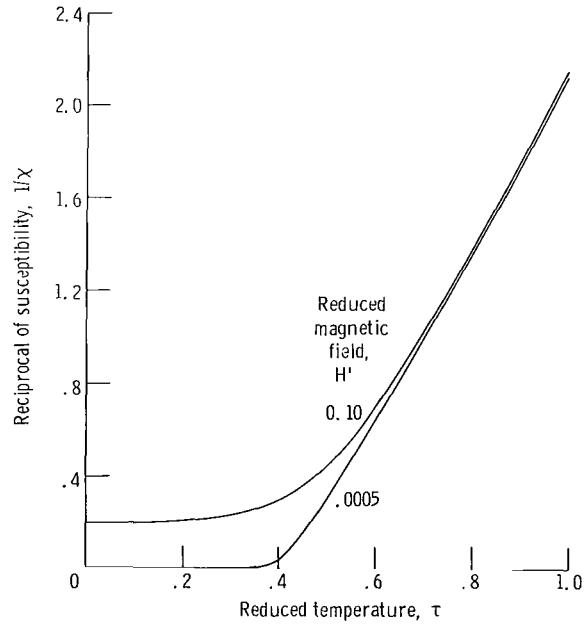

Figure 3. - Reciprocal of susceptibility as function of reduced temperature.

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