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THERMODYNAMICS OF A HEISENBERG FERROMAGNET IN THE RANDOM PHASE APPROXIMATION

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ABSTRACT

The development of a formalism for the magnetic and thermal properties of the Heisenberg ferromagnet in an applied magnetic field is presented. By the use of Green's function theory the magnetization and susceptibility are determined in the random phase approximation. It is shown that the thermodynamic quantities can be expressed in analytical form for all temperatures, both above and below the Curie point.

The object of this work is to evaluate the use of ferromagnetism in a refrigeration system. From the analytical results on magnetization and susceptibility, equations describing adiabatic and isothermal processes are presented. Magnetization and entropy are presented as functions of temperature and field.

Keywords: Green's function, Thermodynamics, Entropy, Refrigeration, Critical point, Magnetocaloric effect

INTRODUCTION

According to the Weiss theory a ferromagnetic body possess 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. The process by which this change from ferromagnetism to paramagnetism occurs is referred to as the ferromagnetic phase transition. Investigations have shown that electrical, mechanical, and many thermodynamic properties of a material are altered when the material undergoes a phase transition.

The effect of an external magnetic field is twofold: (1) the magnetization is increased somewhat above its zero field value and (2) the critical point ceases to exist. 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 more gradual transition.

This report is concerned with the investigation of the thermodynamic properties of the Heisenberg ferromagnet in an external magnetic field. The quantities studied are the magnetization, susceptibility and the derivative of magnetization with respect to temperature. The last of these determines thermodynamic quantities, such as entropy and adiabatic demagnetization (magneto - caloric effect), which are useful for investigating various recently proposed refrigeration systems.[1-2]

MODEL

The Heisenberg model is based on a solid where the magnetic electrons are in states localized about the lattice sites with exchange inter-

actions between electrons taking place between nearest neighbor pairs. The model does not take into account an itinerant electron picture. The model is thought to be very good for insulating ferromagnets such as EuO 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 Arglye, Charap and Pugh.[3] It should be noted however, that the Heisenberg model with its surprising success 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[4] using the techniques of double-time, temperature dependent Green's functions. A convenient review of Green's functions and Tyablikov's application of them to ferromagnetism is given by D. N. Zubarev[5] and references contained therein.

The Hamiltonian for the Heisenberg model is

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

where

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

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

CALCULATION OF THERMODYNAMIC QUANTITIES

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

Starting with the Green's function $\langle\langle S_g^+; S_m^- \rangle\rangle$ one can derive the correlation function in the random phase approximation. The results are

$$\langle S_m^- S_g^+ \rangle = \frac{1}{N} \sum_K 2 \langle S_g^z \rangle e^{ik \cdot (g-m)} \left(e^{\beta E_k} - 1 \right)^{-1} \tag{3}$$

where

$$E_k = 2 \langle S^z \rangle J(0) \left[1 - \frac{J(k)}{J(0)} \right] + \mu_B g H_o \tag{4a}$$

and

$$J(k) = \sum_m J_{gm} e^{ik \cdot (g-m)} \tag{4b}$$

The sum in equation (3) goes over all N lattice vectors in the first Brillouin zone.

Application of a magnetic field is assumed sufficient to orient the net magnetization 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 obtain the magnetization one calculates $\langle S^z \rangle$ by using the relation

$$\langle S^z \rangle = \frac{1}{2[1 + 2\varphi]} \tag{5}$$

where

$$\varphi = \left(\frac{1}{N}\right)\sum_k \left(e^{\beta E_k} - 1\right)^{-1} = \left(\frac{1}{2N}\right)\sum_k \left[\coth(\beta E_k) - 1\right] \tag{6}$$

The sum in equation (6) 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. Such numerical solutions are, however, somewhat difficult. One of the purposes of this paper is to show that analytical solutions of equation (6) are possible.

Consider only crystals with cubic symmetry such as bcc. One can replace the sum which appears in equation (6) by an integral. Using the same techniques developed by Flax and Raich[6-8] one can obtain the magnetization:

$$\langle S^z \rangle = \frac{1}{2\Delta} \tag{7}$$

where

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

$$P = \frac{\alpha}{2} + Q \tag{9}$$

$$\alpha = \frac{\mu_B g H_o}{[J(0)\tau]} = \frac{H'}{\tau} \tag{10}$$

$$\tau = \frac{k_B T}{J(0)} \tag{11}$$

$$Q = \frac{\langle S^z \rangle}{\tau} \tag{12}$$

$K(k)$ = complete elliptic integral of first kind (13)

and

$$k^2 = \left(\frac{1}{2}\right)\left[1 - \sqrt{1 - \left(\frac{Q}{P}\right)^2}\right] \tag{14}$$

The solution of (8) give as a function of temperature, magnetic field and magnetization. Hence by substituting equation (8) into equation (7) the magnetization is obtained.

For an adiabatic process, that is, when there is no change in entropy,

$$d\tau = -\frac{\tau}{C_{H'}}\left(\frac{\partial\langle S^z\rangle}{\partial\tau}\right)_{H'} dH' \tag{15}$$

where C_H is the magnetic specific heat. Since for a ferromagnet $\left(\partial\langle S^z\rangle/\partial\tau\right)_{H'}$ is negative, an increase in field produces an increase in temperature; moreover, the increase will be expected to be largest near the critical point. This is known as the magnetocaloric effect.

The entropy, which is a measure of the order of the system, can be calculated from

$$S = \int_0^{\tau} \frac{C_{H'}}{\tau}\, d\tau + \int_0^{H'} \left(\frac{\partial\langle S^z\rangle}{\partial T}\right)_{H'} dH'' \tag{16}$$

The change of magnetization with respect to the reduced temperature can be derived from equation (5) as follows:

$$\frac{d\langle S^z\rangle}{d\tau} = -\left(\frac{1}{\Delta}\right)^2\left[\left(\frac{\partial\Delta}{\partial\langle S^z\rangle}\right)_{\tau}\frac{d\langle S^z\rangle}{d\tau} + \left(\frac{\partial\Delta}{\partial\tau}\right)_{\langle S^z\rangle}\right] \tag{17}$$

$$\frac{d\langle S^z\rangle}{d\tau}_{H'} = \left(\frac{\partial\langle S^z\rangle}{d\tau}\right)_{H'} = -\frac{\left(\frac{\partial\Delta}{\partial\tau}\right)\langle S^z\rangle}{\left[2\Delta^2 + \left(\frac{\partial\Delta}{\partial\langle S^z\rangle}\right)_\tau\right]} \tag{18}$$

RESULTS AND DISCUSSION

In the absence of an applied magnetic field the random phase approximation predicts that the magnetization shows a second order transition and has a critical temperature which can be written as

$$0.3589 = \frac{k_B T_c}{J(0)} \qquad (H_o = 0) \tag{19}$$

where T_c is the Curie temperature. It is then feasible to relate H_o and H' as follows:

$$H' = \frac{\mu_B g H_o}{j(0)} = \frac{\mu_B g H}{(k_B T_c)} \times 0.3589 \tag{20}$$

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 a certain temperature T_c which is called the Curie temperature. This is the point where the long-range order would disappear and above this point there is 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 forming a "Curie region" in which the transition is smeared out. This is marked by the appearance of a "tail" on the magnetization curve. The cause of the tail is that the long-range order persists because the spins feel the effect of an applied magnetic field. The higher the field the greater the broadening of the transition.

Figure 2 shows the values of the temperature change in the magnetocaloric effect as a function of temperature. These results were derived from equations (15) and (18). An expression for C_H, which is required in equation (15) was derived from the expression $(\partial E/\partial \tau)_{H'}$ where E is the enthalpy. The enthalpy is given by $E = U - H\langle S^z \rangle$. Where U is the internal energy of the system, and can be found from a knowledge of the Hamiltonian.[9] As indicated in figure 2, the magnetocaloric effect is largest in the neighborhood of the Curie temperature. The magnitude of the magnetocaloric effect is dependent on the internal magnetic field as well as the temperature. For very large fields the magnetocaloric effect rises indefinitely as for an ideal paramagnet. (Note that the lattice specific heat is not taken into account in this calculation).

Figure 3 shows a plot of S as a function of temperature for several values of H'. The curves show a characteristic behavior for ferromagnetic to paramagnetic transition. It is included here to illustrate that the entropy can be calculated for a ferromagnet as a function of temperature, magnetization and applied field. Such curves are useful for the design of refrigeration cycles.

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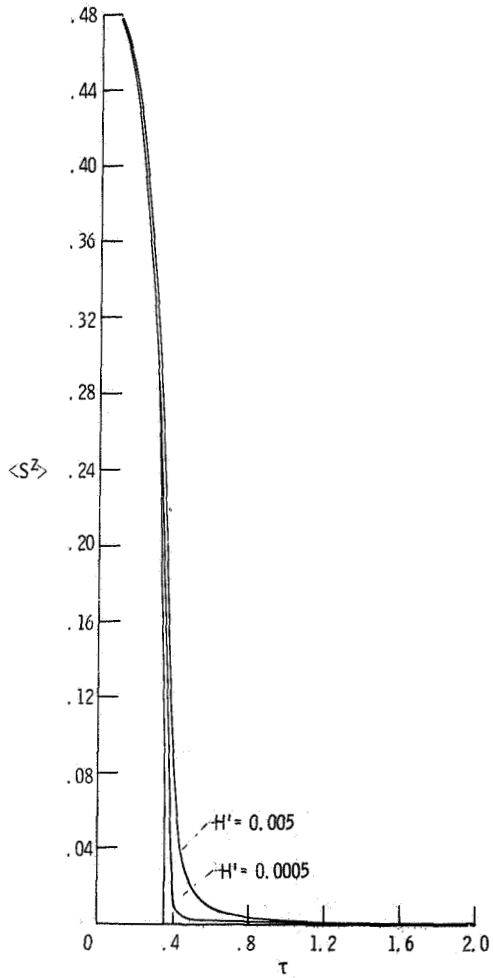

Figure 1. - Magnetization as a function of temperature.

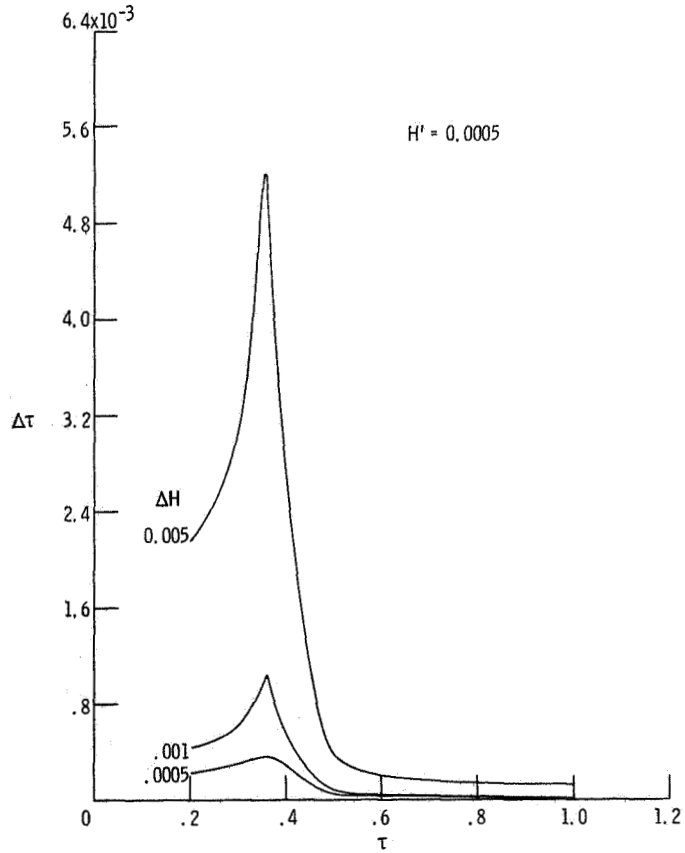

Figure 2. - Temperature change as a function of temperature.

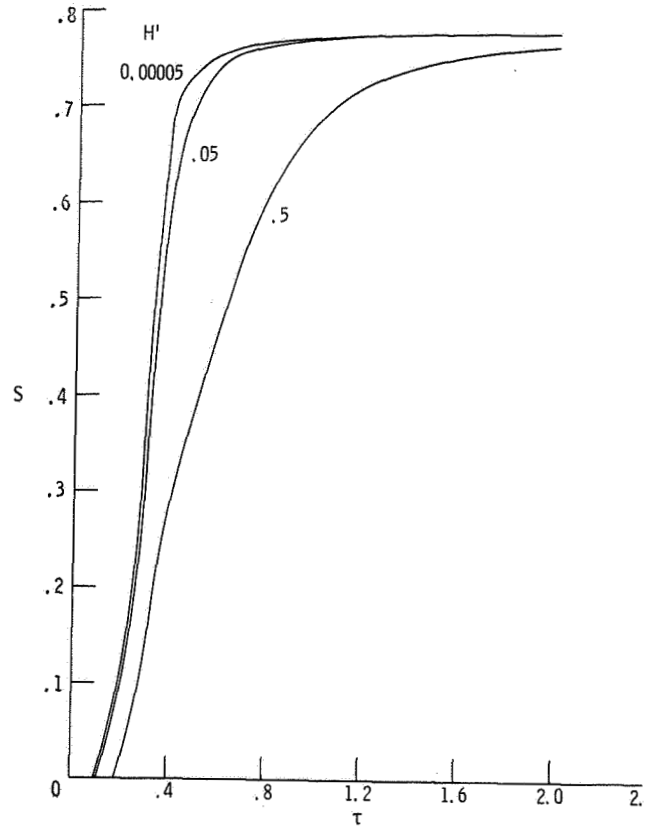

Figure 3. - Entropy as a function of temperature.