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Nonlinear Spin-Wave Theory for the Anisotropic Heisenberg Antiferromagnet

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The theory of the anisotropic Heisenberg antiferromagnet is developed in the nonlinear spin-wave approximation. It is shown that the thermodynamic quantities of interest depend on two renormalization parameters. These parameters can be calculated analytically over the entire temperature range of interest. It appears that nonlinear spin-wave theory does not give a good estimate of the transition temperature as the anisotropy increases.

INTRODUCTION

One of the most important methods in studying the thermodynamic behavior of ferromagnetism or antiferromagnetism is by spin-wave theory, first initiated by Bloch.[1] The assumption is that spin-wave excitations do not interact. By the use of a Holstein-Primakoff transformation the spin-wave operators can be cast into the form of boson operators. This description of the system in terms of noninteracting bosons loses its validity as the temperature is increased. The theory suffers from the drawback that its applicability is limited to a narrow range of temperatures.

With the use of a suitable transformation, Dyson[2] transformed the Hamiltonian of spin operators into a boson form which consisted of quadratic and quartic operators. The quadratic part yielded the Bloch spin-wave theory, and the nonquadratic part leads the now famous T^4 term in the magnetization. He was therefore able to calculate the effects of spin-wave interaction for the Heisenberg ferromagnet.

Bloch[3] extended the regions of applicability of the spin-wave theory by truncating Dyson's Hamiltonian and finding solutions at higher temperatures.

Liu[4] extended the Bloch theory to a Heisenberg antiferromagnet. Using Green's-function formalism, the model was studied at high temperatures for spin-wave interactions. It was shown that the

sublattice magnetization of the system depended on a renormalization parameter $\alpha(T)$. This parameter satisfied an implicit equation which depends on temperature. The results of this investigation showed that the estimated temperature was within a few percent of the theoretical values of the Néel temperature. At low temperatures, a power-series expansion approximation agreed with Oguchi's[5] theory of spin waves.

The purpose of this paper is to investigate nonlinear spin-wave interactions for the anisotropic Heisenberg antiferromagnet for the spin-$\frac{1}{2}$ case. With the use of thermodynamic Green's function the sublattice magnetization of the system is investigated for all temperature regions of interest. The sublattice magnetization for the anisotropic model depends on two renormalization parameters $\Delta(T)$ and $\beta(T)$. These parameters satisfy equations which can be solved self-consistently to determine their dependence on temperature.

HAMILTONIAN

The nearest-neighbor-exchange interaction in the absence of an external field is given by the Hamiltonian

$$H = J\sum_e \sum_\delta \Big[S^z_{1i} S^z_{2,\,i+\delta} + \frac{\eta}{2}\,(S^+_{1i+\delta} S^-_{2,\,i+\delta} + S^-_{1i} S^+_{2,\,i+\delta})\Big] + J\sum_2 \sum_\delta \Big[S^z_{i,\,j+\delta} S^z_{2j} + \frac{\eta}{2}\,(S^z_{1,\,j+\delta} S^-_{2,\,j+\delta} + S^-_{1,\,j+\delta} S^+_{2j})\Big], \tag{1}$$

where the sums on i and j extend over all sites, S_i is the spin operator for $S=\frac{1}{2}$ for a spin at site i, and J is the exchange integral. The indices 1, 2 refer to the two sublattices, $2, i+\delta$ is a nearest neighbor of $1j$ or $1j$, and η is the anisotropy parameter ($0 \le \eta \le 1$). The Ising and isotropic Heisenberg models are obtained by setting $\eta = 0, 1$, respectively.

The notation in this paper is similar to that used by Liu[4] whose calculation corresponds to the special case $\eta = 1$.

Dalton and Wood,[6] and Flax and Raich[7] have studied the critical behavior of the anisotropic ferromagnetic model for intermediate values of η by the Green's-function method.

Using the Maleev[8] transformation the spin operators can be expressed as boson operators in the form

$$\begin{aligned} S^-_{1i} &= b_{1i} - b^\dagger_{1i}\, b_{1i} b_{1i}\,, \\ S^-_{2j} &= b^\dagger_{2j} - b^\dagger_{2j} b_{2j} b_{2j}\,, \\ S^+_{1i} &= b^\dagger_{1i},\quad S^+_{2j} = b_{2j}\,, \\ S^z_{1i} &= -\tfrac{1}{2}\,(1 - 2b^\dagger_{1i} b_{1i})\,, \\ S^z_{2j} &= \tfrac{1}{2}\,(1 - 2b^\dagger_{2j} b_{2j})\,, \end{aligned} \tag{2}$$

where the operators b_i and $b^\dagger_i$ are to be regarded as spin-wave creation and annihilation operators which satisfy the Bose commutation rules. However, as was pointed out in Ref. 4, the commutation relations between the spin operators are still valid, but S^+ and S^- are no longer Hermitian conjugates.

The Fourier transforms of $b^\dagger_i$ and b_i are introduced where

$$\begin{aligned} b_{1i} &= \left(\frac{2}{N}\right)^{1/2} \sum_k c_k\, e^{i\vec{k}\cdot\vec{R}_{1i}} \\ b^\dagger_{1i} &= \left(\frac{2}{N}\right)^{1/2} \sum_k c^\dagger_k\, e^{-i\vec{k}\cdot\vec{R}_{1i}}\,, \\ b_{2j} &= \left(\frac{2}{N}\right)^{1/2} \sum_k d_k\, e^{-i\vec{k}\cdot\vec{R}_{2j}}\,, \\ b^\dagger_{2j} &= \left(\frac{2}{N}\right)^{1/2} \sum_k d^\dagger_k\, e^{i\vec{k}\cdot\vec{R}_{2j}}\,, \end{aligned} \tag{3}$$

where N is the total number of lattice sites, $\vec{R}_{1i}$ is the position of the ith site of sublattice 1, and $\vec{R}_{2j}$ is the position of the jth site for sublattice 2. (Vector notation is suppressed in subscripts.) In terms of these operators, the transformed Hamiltonian becomes

$$\begin{aligned} H = -\frac{JNz}{4} &+ Jz\sum_k (d^\dagger_k d_k + c^\dagger_k c_k) - \frac{4Jz}{N}\sum_{k,k',q} \gamma(\vec{k}-\vec{k}')\, c^\dagger_k c_{k'} d^\dagger_{k+q} d_{k'+q} + \eta J z\Big(\sum_k \gamma(k)\,(c^\dagger_k + d^\dagger_k + c_k d_k) \\ &- \frac{1}{N}\sum_{k,k'q} \gamma(k)\, c^\dagger_k d^\dagger_{k+q} d^\dagger_{k'-q} d_{k'} - \frac{1}{N}\sum_{k,k',q} \gamma(k+q)\, c^\dagger_{k'} c_k c_{k'+q} d_{k+q} \\ &- \frac{1}{N}\sum_{k,k',q} \gamma(k)\, c_k d^\dagger_{k'} d_{k'-q} d_{k+q} \\ &- \frac{1}{N}\sum_{k,k',q} \gamma(k+q)\, c^\dagger_{k'+q} c^\dagger_k c_{k'} d^\dagger_{k+q}\Big), \end{aligned} \tag{4}$$

where z is the number of nearest neighbors,

$$\gamma(k) = (1/z) \sum_{\delta} e^{i\vec{k}\cdot\vec{\delta}} \tag{5}$$

and $\vec{\delta}$ is a vector connecting nearest neighbors. The last two terms in Eq. (4) come from the requirement that the total Hamiltonian must be Hermitian.

The operators c_k and d_k satisfy the basic commutation relations

$$(c_k c_{k'}^{\dagger}) = \delta_{kk'}, \ (d_k^{\dagger} d_{k'}) = -\delta_{kk'}, \ (c_k d_k) = (d_k^{\dagger} c_k^{\dagger}) = 0 . \tag{6}$$

EQUATION OF MOTION FOR GREEN'S FUNCTION

The Fourier transform $\langle\langle A;B \rangle\rangle$ of the time-dependent Green's function for the two operators A and B satisfies the equation of motion[9]

$$\langle\langle A; B\rangle\rangle = (2\pi)^{-1} \langle [A, B]\rangle + \langle\langle [A, H]; B\rangle\rangle, \tag{7}$$

where the single brackets $\langle\ \rangle$ indicate averages over the canonical ensemble at temperature T.

Starting with the Green's functions $\langle\langle c_k; c_k^{\dagger}\rangle\rangle$ and $\langle\langle d_k^{\dagger}; c_k^{\dagger}\rangle\rangle$ which are denoted by G_{11} and G_{21}, respectively, one obtains in the random-phase approximation

$$EG_{11} = (2\pi)^{-1} + [\alpha + Jz]\, G_{11} + [\Delta + Jz\eta]\gamma(k) G_{21} , \tag{8}$$

$$EG_{21} = -[\Delta + Jz]\gamma(k)\, G_{11} - [\alpha + Jz] G_{21} , \tag{9}$$

where

$$\alpha = -(4Jz/N) \sum_q [n_q + \eta\gamma(q)\xi_q] , \tag{10}$$

$$\Delta = -(4Jz/N) \sum_q [\gamma(q)\xi_q + \eta n_q] , \tag{11}$$

$$n_k = \langle c_k^{\dagger} c_k\rangle = \langle d_k^{\dagger} d_k\rangle , \tag{12}$$

$$\xi_k = \langle c_k d_k\rangle = \langle d_k^{\dagger} c_k^{\dagger}\rangle ,$$

and where one has made use of the nearest-neighbor approximation. One obtains for G_{11} and G_{21} therefore

$$G_{11} = (E + E_0)/2\pi(E^2 - E_0^2 + W^2) , \tag{13}$$

$$G_{21} = -W/2\pi(E^2 - E_0^2 + W^2) , \tag{14}$$

where

$$E_0 = \alpha + Jz , \tag{15}$$

and

$$W = (\Delta + Jz\eta)\gamma(k) . \tag{16}$$

The correlation function is found from the relation

$$\langle B(t')A(t)\rangle = \lim_{\epsilon \to +0} i \int_{-\infty}^{\infty} \frac{(\langle\langle A; B_{E+i\epsilon}\rangle\rangle - \langle\langle A; B_{E-i\epsilon}\rangle\rangle) e^{-iE(t-t')} dE}{e^{\beta E} - 1} , \tag{17}$$

where $\beta = 1/KT$. Using Eq. (17), an elementary integration yields

$$\langle c_k^{+} c_k\rangle = n_k = \tfrac{1}{2}[(E_0/E_1)\coth \tfrac{1}{2}\beta E_1 - 1], \tag{18}$$

$$\langle d_k^{+} c_k^{+}\rangle = \xi_k = -(W/2E_1)\coth \tfrac{1}{2}\beta E_1 , \tag{19}$$

where

$$E_1^2 = E_0^2 - W^2 . \tag{20}$$

Substituting Eqs. (18) and (19) into Eqs. (10) and (11) thus results

$$\alpha = Jz\left\{1 - \frac{2}{N}\sum_k \frac{1}{E_1}\,[E_0 - \eta\gamma(k)W]\coth \tfrac{1}{2}\beta E_1\right\} , \tag{21}$$

$$\Delta = Jz\left\{\eta - \frac{2}{N}\sum_k \frac{1}{E_1}\,[\eta E_0 - W\gamma(k)]\coth \tfrac{1}{2}\beta E_1\right\} . \tag{22}$$

To obtain the spontaneous sublattic magnetization one calculates $\langle S^z\rangle$ from Eq. (2). The result is

$$\langle S^z\rangle = \frac{1}{2}\left[1 + \frac{2}{N}\sum_k \left(1 - \frac{E_0}{E}\coth \tfrac{1}{2}\beta E_1\right)\right] . \tag{23}$$

Equations (21)–(23) must now be solved simultaneously in order to yield the magnetization.

ANALYTICAL SOLUTION FOR MAGNETIZATION

To calculate the magnetization as a function of temperature, the sums in Eqs. (21)–(23) 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, numerical solutions are difficult. One of the purposes of this paper is to show that analytical solutions of Eq. (23) are possible.

Consider only crystals with cubic symmetry such as a bcc. For this lattice one can replace the sum which appears in the equation by an integral

$$\frac{2}{N}\sum_k \frac{1}{\pi^3}\int_0^{\pi}\int_0^{\pi}\int_0^{\pi} dx\, dy\, dz , \tag{24}$$

$$x = \tfrac{1}{2}k_x a, \quad y = \tfrac{1}{2}k_y a, \quad z = \tfrac{1}{2}k_z a . \tag{25}$$

One may write Eqs. (21)–(23) in the form

$$\alpha = Jz(1 - I) , \tag{26}$$

$$\Delta = Jz(\eta + II) , \tag{27}$$

$$\langle S^z\rangle = 1 - \tfrac{1}{2} I_a , \tag{28}$$

where

$$I=\left(\frac{1}{\pi}\right)^3\int_0^\pi\int_0^\pi\int_0^\pi[E_0-\eta\gamma(k)W]\frac{1}{E_1}\coth\tfrac{1}{2}\beta E_1\,dx\,dy\,dz, \tag{29}$$

$$II=\left(\frac{1}{\pi}\right)^3\int_0^\pi\int_0^\pi\int_0^\pi[\gamma(k)W-\eta E_0]\frac{1}{E_1}\coth\tfrac{1}{2}\beta E_1\,dx\,dy\,dz, \tag{30}$$

$$I_a=\left(\frac{1}{\pi}\right)^3\int_0^\pi\int_0^\pi\int_0^\pi\frac{E_0}{E_1}\coth\tfrac{1}{2}\beta E_1\,dx\,dy\,dz. \tag{31}$$

For the case of the bcc lattice, $\gamma(k)$ is given by

$$\gamma(k)=\cos\tfrac{1}{2}k_xa\cos\tfrac{1}{2}k_ya\cos\tfrac{1}{2}k_za. \tag{32}$$

Using the identity

$$\coth\pi t=(\pi t)^{-1}+\frac{2}{\pi}\sum_{R=1}^{\infty}\frac{t}{R^2+t^2} \tag{33}$$

and the techniques developed by Flax and Raich[7] one can obtain the following:

$$I=I_a-\eta I_b, \tag{34}$$

$$II=I_b-\eta I_a, \tag{35}$$

where

$$I_a=(1/\pi p)\{[(2/\pi)K(k)]^2-1-\tfrac{1}{2}m^2\}+(2+m^2/16)\coth\pi p+(m^2/16)\pi p\,\operatorname{csch}\pi p, \tag{36}$$

$$I_b=(1/\pi p)\{(1/m)[(2/\pi)K(k)]^2-1/m-m/8-27m^2/512\}+(m/8+27m^3/1024)\coth\pi p+(27/1024)m^3(\pi p)\operatorname{csch}^2\pi p, \tag{37}$$

$$T_{\rm red}=(KT/Jz), \tag{38}$$

$$\pi p=(\alpha_1/2T_{\rm red}), \tag{39}$$

$$\alpha_1=[\alpha/(Jz)+1], \tag{40}$$

$$\Delta_k=[\Delta/(Jz)+\eta], \tag{41}$$

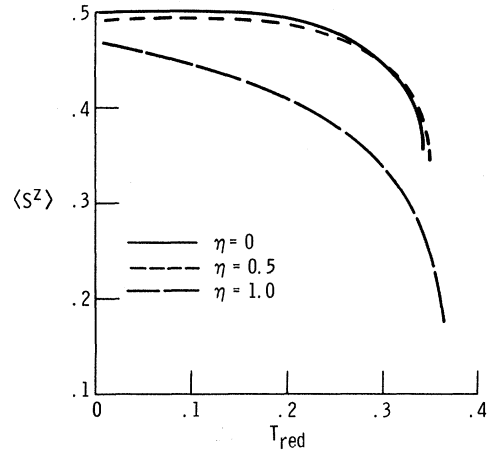

FIG. 1. Sublattice magnetization against the reduced temperature.

TABLE I. Comparison of $T_{\rm max}$ with estimations of T_N for a bcc lattice with spin $\frac{1}{2}$.

η	KT_N/J BPW Ref. 10 approx.	KT_J Tyablikov Ref. 11 approx.	$T_{\rm max}/J$ Liu Ref. 4 approx.	$KT_{\rm max}/J$ Present calculations
1	3.18	2.87	2.91	2.89
0.5				2.78
0				2.74

$K(k)$ = complete elliptic integral of first kind, (42)

$$k^2=\tfrac{1}{2}[1-(1-m^2)^{1/2}], \tag{43}$$

and

$$m=(\Delta_1/\alpha_1). \tag{44}$$

The solution of Eqs. (26) and (27) gives α and Δ as a function of temperature. Hence by substituting Eqs. (36)–(38) into Eq. (28) the sublattice magnetization is obtained.

RESULTS AND DISCUSSION

Figure 1 shows a plot of $\langle S^z\rangle$ as a function of temperature $T_{\rm red}$ for several values of η for the bcc lattice. There are no solutions for $\langle S^z\rangle$ above a certain temperature $T_{\rm max}$, which varies for different η. The slope of the magnetization approaches infinity as $T_{\rm red}$ approaches $T_{\rm max}$. As pointed out by Liu,[4] the nonlinear spin-wave theory ignores the kinematic interactions between the spin waves, so that the nature of the instability at $T_{\rm max}$ is quite different from the phase transition at the Néel temperature T_N. Nevertheless, for $\eta=1$ one obtains values of $T_{\rm max}$ which are very close to other theoretical estimates of the Néel temperature. In Table I, the values for the temperature $T_{\rm max}$ are given for values of the parameter η for the bcc lattice. Also shown are results for T_N and $T_{\rm max}$ calculated by other approximation.

The random-phase approximation predicts that the magnetization shows a second-order phase transition independent of the anisotropy. One effect of the anisotropy parameter is to shift the critical temperature. The critical temperature should decrease smoothly as η increases. Physically this is due to the energy it takes to reverse a spin. For the Ising model, since there is only one spin component present, an energy gap exists and additional energy is needed to create an excitation. In the isotropic model, the gap vanishes owing to spin waves. Figure 1 shows that these physical arguments are not supported since $T_{\rm max}$ decreases slightly as η decreases from unity.

One must therefore conclude that the nonlinear spin-wave theory for the antiferromagnet, although accurate at low temperatures, does not appear to give a good estimate of the transition temperature for $\eta \neq 1$.

SUMMARY

A single analytical expression for the magnetization was obtained, which is valid for all temperatures. The demonstrated technique of evaluating certain temperature-dependent sums and integrals was employed without necessitating the use of various series approximations or entensive numerically computed summations. Other thermodynamic quantities, such as specific heat, can be calculated in the same manner.

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Magnetic Susceptibility of Dilute Al-Mn Alloys

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The temperature dependence of the magnetic susceptibility of pure aluminum and three aluminum alloys containing 0.045, 0.21, and 0.35 at.% manganese has been measured between 2 and 300 °K. All of the susceptibility results can be fitted to an equation of the form $A + BT^2$. The experimental values of the coefficients A and B for pure aluminum are $19.2 \pm 0.1 \times 10^{-7}$ emu/cc and $3.7 \pm 0.1 \times 10^{-12}$ emu/cc/(°K^2), which are in good agreement with theoretical values derived from the known band structure of aluminum. An additional susceptibility component is observed in th Al-Mn alloys, and the experimental results can be characterized by the equation $\Delta K_{\mathrm{Mn}}(T)/C$ emu/cc at.% Mn $= 13.8 \pm 0.8 \times 10^{-7} - (14 \pm 3 \times 10^{-13})T^2$. The experimentally determined coefficients can be explained by considering an exchange enhancement of the Pauli paramagnetism within the framework of a Hartree-Fock approximation. Values of the exchange enhancement factor η, and the width of the virtual bound d states, Δ, can be uniquely determined and have the values 4.4 ± 1 and 0.33 ± 0.05 eV, respectively.

It has been known for some time that a transition-metal impurity added to a monovalent metal gives rise to a Curie-Weiss law at high temperatures, e.g., for Cu-Mn $\mu_{eff} = 4.9\ \mu_B$.[1]

In a divalent host, the higher electron density may result in greater mixing of the conduction electron and d states with a resulting decrease in the moment carried on the manganese, e.g., for Zn-Mn $\mu_{eff} = 3.9\ \mu_B$.[1] Both of the above systems exhibit a Kondo effect, i.e., a ln-T term in the low-temperature resistivity. In going to a trivalent solvent such as aluminum with a still higher electron density, the manganese ion does not obey a Curie-Weiss law and no Kondo effect is observed.

The present investigation reports on some measurements of the low-temperature static volume susceptibility, $K(T)$ of Al-Mn alloys and pure aluminum. A negative T^2 term is found at low temperatures. The magnitude of the additional observed T^2 term due to the presence of manganese in the Al-Mn alloys is consistent with the value of susceptibility calculated in the Hartree-Fock approximation of Friedel[2] and Andersen.[3,4] These theories lead to an exchange of the band paramagnetism.

The aluminum sample was made from Cominco 99.9999+% pure aluminum and the Al 0.045 at.% Mn alloy was made by the Aluminum Company of Canada.[5] The two other alloys were made from the 0.045 at.% Al-Mn alloy and 99.99% pure manganese. The alloys were melted at 850 °C and quenched in ice water from 600 °C after a 60-h homogenizing anneal. The pure aluminum specimen was given