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Variational Treatment of the Heisenberg Antiferromagnet*

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A form of the Peierls free-energy variational theorem is applied to the Heisenberg Hamiltonian for a three-dimensional system with nearest-neighbor antiferromagnetic interaction. For a large magnetic field ($h\equiv g\mu H/4SJz\approx 1$) we find a phase boundary separating a region of antiferromagnetic order from one of ferromagnetic order. At low temperatures ($\theta\equiv kT/2SJz\ll 1$) the phase boundary has the leading behavior: $h=1-a\theta^{3/2}$ with $a=2\zeta(3/2)(3/2\pi)^{3/2}/S$ for a simple cubic antiferromagnetic lattice (e.g., $RbMnF_3$). At the phase boundary the magnetization is continuous; whereas a discontinuity in the susceptibility is suggested but not firmly established by this treatment. Low-temperature expressions are given for the magnetization, susceptibility, and specific heat above the boundary. Numerical calculations show that, for the approximation used, the phase boundary extends to a maximum θ at which the magnetization is nonzero. For the limiting case of $h=0$ we obtain Keffer and Loudon's renormalized spectrum and magnetization for a ferromagnet and for an antiferromagnet from a single variational calculation. Attention is also given to a reduced Hamiltonian which, when treated by the variational method, exhibits the properties of an antiferromagnetic molecular field model previously proposed by Garrett for $S=\frac{1}{2}$.

1. INTRODUCTION

WE are considering the spin-system Hamiltonian

$$\mathcal{H}=J\sum_{\mathbf{f}}\sum_{\delta}\mathbf{S}_{\mathbf{f}}\cdot\mathbf{S}_{\mathbf{f}+\delta}+g\mu H\sum_{\mathbf{f}}S_{\mathbf{f}}^{z} \qquad (1)$$

in which the double summation (over lattice sites f and nearest neighbors δ) represents the antiferromagnetic Heisenberg exchange interaction, and the single summation accounts for the Zeeman energy of the spin system in an applied magnetic field H. The symbols J, g, and μ denote the exchange energy, Lande' factor, and Bohr magneton, respectively.

Although the ground state of $\mathcal{H}$ (for $H=0$) is known[1] to be a nondegenerate singlet, neither the eigenfunction nor the energy has been given for two- and three-dimensional lattices.

For a one-dimensional infinite chain the ground-state energy[2] and the associated short-range correlation[3] $\langle\sum_{\mathbf{f}}S_{\mathbf{f}}^{z}S_{\mathbf{f}+\delta}^{z}\rangle$ are known exactly. Also to be found[4,5] in the literature are the eigenvalues and eigenfunctions of some finite chains.

If one neglects the $S_{\mathbf{f}}^{x}S_{\mathbf{f}+\delta}^{x}$ and $S_{\mathbf{f}}^{y}S_{\mathbf{f}+\delta}^{y}$ terms in (1), then what remains is the Ising model[6] which has been solved exactly at finite temperatures for a one-dimensional chain and for the two-dimensional nets, the latter for $H=0$.

Now it has been shown[7] that for H greater than a critical field $H_c=4SJz/g\mu$, the antiferromagnet Hamiltonian (1) has the same ground state as the *ferro*magnet Hamiltonian; that is, all spins are parallel to the external field.

Motivated by this latter result, which suggests using some of the relatively well-established theory of the ferromagnet, we study (1) primarily with attention given to the simple cubic lattice in a large magnetic field ($h\equiv H/H_c=g\mu H/4SJz\approx 1$), and at low temperatures ($\theta\equiv kT/2SJz\ll 1$). The symbols k, T and z denote the Boltzmann constant, the absolute temperature, and number of nearest neighbors, respectively.

We find a phase boundary which has the low-temperature form

$$h=1-a\theta^{3/2}+\cdots,$$

with

$$a=\frac{(2)\zeta(3/2)}{S}\left(\frac{3}{2\pi}\right)^{3/2}, \quad \text{(simple cubic)}.$$

The boundary separates a region of antiferromagnetic sublattice canting from a region of ferromagnetic order. Across the phase boundary the magnetization is continuous; whereas a discontinuity in the susceptibility is suggested but not firmly established by this treatment.

Our calculation is based on a modified[8,9] (weak) form of the Peierls variational theorem for the free energy. The weak form of the theorem has been applied in the study of superconductivity,[10] ferromagnetism,[11] antiferromagnetism,[12] and general many-body systems.[13–15] Although it is well known to some, we mention that this method is essentially equivalent both to first order,

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† Part of this work is based on the author's Ph.D. thesis submitted to the University of Washington, Seattle, Washington.
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[15] V. Tolmachev, Dokl. Akad. Nauk SSSR **134**, 1324 (1960) [English transl.: Soviet Phys.—Doklady **5**, 984 (1961)].

finite-temperature perturbation theory[16] and to a method of linearizing the equations of motion[13,17] (random-phase approximation).

The method of calculation was viewed with some confidence after it yielded the following results:

(a) For $h=0$ the temperature dependence of the renormalized spectrum and the sublattice magnetization are in agreement (Appendix A) with well-known results,[18,19] and the average ground-state magnetization is found to vanish.[1]

(b) For the linear chain we found[20] no phase boundary. The variationally obtained Fermion excitation spectrum for $h=0$ is linear in k in the long-wavelength limit, and the calculated ground-state energy is close to the exactly known value.

(c) For the *ferromagnet* (Appendix B) we easily obtain the renormalized spectrum and magnetization presented by Keffer and Loudon.[19]

2. METHOD OF CALCULATION

The variational theorem states that for a system described by a Hamiltonian $\mathcal{H}=\mathcal{H}_0+(\mathcal{H}-\mathcal{H}_0)$, an upper bound to the exact free energy is

$$F=F_0+\langle\mathcal{H}-\mathcal{H}_0\rangle_0, \tag{2}$$

where F_0 is the free energy associated with $\mathcal{H}_0$, and

$$\langle Q\rangle_0=\frac{\mathrm{Tr}(e^{-\beta\mathcal{H}_0}Q)}{\mathrm{Tr}e^{-\beta\mathcal{H}_0}}, \quad (\beta=1/kT). \tag{3}$$

Frequently F is written in terms of the entropy S_0:

$$F=\langle\mathcal{H}\rangle_0-TS_0,$$

where $\mathcal{H}_0$ is taken to be the free particles' Hamiltonian and S_0 is the associated entropy.

Our procedure is first to express $\mathcal{H}$ in terms of boson (or Fermion) creation and absorbtion operators $c_{\mathbf{k}}^\dagger$ and $c_{\mathbf{k}}$. Then $\mathcal{H}_0$ is selected to be of the form

$$\mathcal{H}_0=\sum_{\mathbf{k}}\epsilon_{\mathbf{k}}a_{\mathbf{k}}^\dagger a_{\mathbf{k}}, \tag{4}$$

where $\epsilon_{\mathbf{k}}$ is the single-particle spectrum to be determined. The operators $c_{\mathbf{k}}$ and $a_{\mathbf{k}}$ are related by the transformation

$$c_{\mathbf{k}}=u_{\mathbf{k}}a_{\mathbf{k}}+v_{\mathbf{k}}a_{-\mathbf{k}}^\dagger \tag{5}$$

in which $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$, both to be determined, satisfy a relationship which makes (5) a canonical transformation. When $c_{\mathbf{k}}$ are boson operators, $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ will be real even functions of $\mathbf{k}$ and satisfy

$$u_{\mathbf{k}}^2-v_{\mathbf{k}}^2=1, \quad \text{(boson case)}. \tag{6}$$

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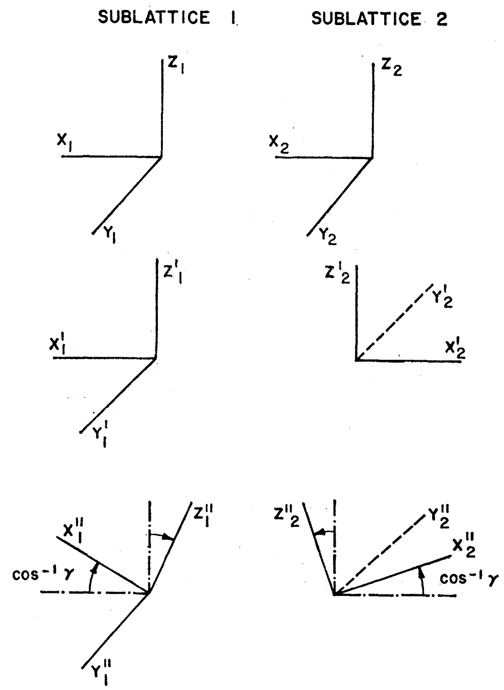

FIG. 1. Sublattice transformations.

For Fermion operators $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ will be complex functions and satisfy

$$\left.\begin{aligned} u_{\mathbf{k}}&=u_{-\mathbf{k}};\\ v_{\mathbf{k}}&=-v_{-\mathbf{k}},\\ \text{and}\quad |u_{\mathbf{k}}|^2&+|v_{\mathbf{k}}|^2=1.\end{aligned}\right\} \quad \text{(Fermion case)}. \tag{6'}$$

The trial free energy F is extremized with respect to functional variations of the transformation $v_{\mathbf{k}}$, the spectrum $\epsilon_{\mathbf{k}}$, the average occupation number $n_{\mathbf{k}}$, and variations of any free parameters. The resulting set of coupled nonlinear integral equations is solved to determine the optimum $n_{\mathbf{k}}$, $v_{\mathbf{k}}$, $\epsilon_{\mathbf{k}}$, and varied parameters. The physical averages of interest may then be calculated according to (3).

Equation (2) is seen to be equivalent to first-order perturbation theory at finite temperatures.[16] In our method we merely try to optimize our choice of unperturbed Hamiltonian $\mathcal{H}_0$. That the method described is essentially equivalent to self-consistently linearizing the equations of motion for $c_{\mathbf{k}}$ and $c_{\mathbf{k}}^\dagger$, has been discussed by Valatin and Butler.[13,17]

3. ROTATION

The Hamiltonian (1) is transformed according to the sequence of rotations shown in Fig. 1. In terms of the transformed coordinates the spin operators become

$$\begin{aligned} S_1^x&=\gamma S_1^{x''}-(1-\gamma^2)^{1/2}S_1^{z''},\\ S_1^y&=S_1^{y''},\\ S_1^z&=(1-\gamma^2)^{1/2}S_1^{x''}+\gamma S_1^{z''},\end{aligned} \tag{7}$$

and

$$S_2^x = -\gamma S_2^{x''} + (1-\gamma^2)^{1/2} S_2^{z''},$$
$$S_2^y = -S_2^{y''}, \tag{8}$$
$$S_2^z = (1-\gamma^2)^{1/2} S_2^{x''} + \gamma S_2^{z''}.$$

The transformed Hamiltonian is

$$\begin{aligned}\mathcal{H} = J \textstyle\sum_\delta \sum_f \big[\tfrac{1}{2}(1-\gamma^2)(S_f^+ S_{f+\delta}^+ + S_f^- S_{f+\delta}^-) \\ -\tfrac{1}{2}\gamma^2 (S_f^+ S_{f+\delta}^- + S_f^- S_{f+\delta}^+) - (1-2\gamma^2) S_f^z S_{f+\delta}^z\big] \\ + g\mu H \gamma \textstyle\sum_f S_f^z, \quad (9)\end{aligned}$$

where double primes are suppressed, and terms like $\gamma(1-\gamma^2)^{1/2} S_f^x S_{f+\delta}^z$ and $H(1-\gamma^2)^{1/2} S_f^x$ are dropped, because their expectation vanishes for all ensembles which we consider. In writing (9) we have employed the usual definition $S^{\pm} = S^x \pm iS^y$.

4. SIMPLE MODEL

As a simple illustration of the method, we consider only the z components of the spin operators in the rotated model (9). We take $S = \frac{1}{2}$ and write S_g^z in terms of fermion operators c_g:

$$S_g^z = c_g^\dagger c_g - S. \tag{10}$$

The problem is now to treat the Hamiltonian

$$\begin{aligned}\mathcal{H} = J \textstyle\sum_\delta \sum_f \big[(2\gamma^2-1)(c_f^\dagger c_f - S)(c_{f+\delta}^\dagger c_{f+\delta} - S) \\ + 2h\gamma(c_f^\dagger c_f - S)\big], \quad (11)\end{aligned}$$

where

$$h = g\mu H / 4SJz$$

and

$$S = \tfrac{1}{2}.$$

The Fourier Fermion amplitudes a_k are defined by

$$c_f^\dagger = N^{-1/2} \textstyle\sum_k e^{i\mathbf{k}\cdot\mathbf{f}} a_k^\dagger,$$
$$c_f = N^{-1/2} \textstyle\sum_k e^{-i\mathbf{k}\cdot\mathbf{f}} a_k. \tag{12}$$

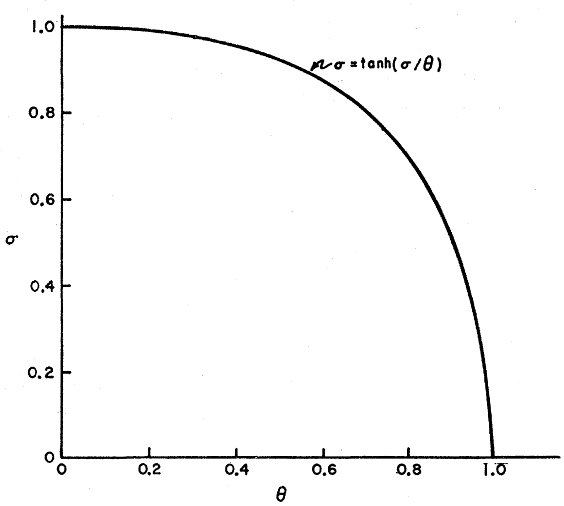

FIG. 2. Reduced sublattice magnetization (simple model).

We apply the identity transformation, i.e., (5) with $u_k = 1$ and $v_k = 0$, and calculate the thermal average of $\mathcal{H}$ according to (3) and (4). By using the finite temperature form[16] of Wick's theorem, one easily verifies the result:

$$\begin{aligned}\langle\mathcal{H}\rangle_0 / NJz = (2\gamma^2-1)[(S-A)^2 - (A-B)^2] \\ -2h\gamma(S-A), \quad (13)\end{aligned}$$

with

$$A = N^{-1} \textstyle\sum_k n_k, \tag{14}$$
$$B = N^{-1} \textstyle\sum_k (1-\gamma_k) n_k, \tag{15}$$
$$n_k = \langle a_k^\dagger a_k \rangle_0,$$

and

$$\gamma_k = z^{-1} \textstyle\sum_\delta \cos(\mathbf{k}\cdot\boldsymbol{\delta}). \tag{16}$$

By requiring that (2) be stationary with respect to

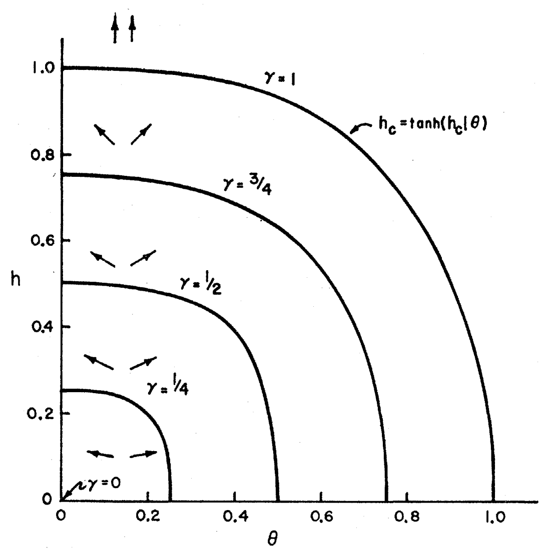

FIG. 3. Curves of constant canting of the sublattices, where the phase boundary locus is $\gamma = 1$ (simple model).

variation of ϵ_k, n_k, and γ, we find the coupled equations

$$\sigma = N^{-1} \textstyle\sum_k \tanh(\omega_k/\theta), \quad (\theta = 2kT/2SJz), \tag{17}$$
$$2(A-B) = N^{-1} \textstyle\sum_k \gamma_k \tanh(\omega_k/\theta), \tag{18}$$
$$h\sigma = \gamma[\sigma^2 - 4(A-B)^2], \quad (\gamma^2 < 1), \tag{19}$$

where the spectrum is

$$\omega_k = 2h\gamma - (2\gamma^2-1)\sigma - 2(2\gamma^2-1)(A-B)\gamma_k, \tag{20}$$

and the reduced magnetization is

$$\begin{aligned}\sigma &= |\langle S^z\rangle_0| / S \\ &= 1 - 2A.\end{aligned} \tag{21}$$

By observing that $\sum_k \gamma_k = 0$ for cubic lattices, we find the particular solution

$$B=A\,,$$
$$\sigma=\tanh(\sigma/\theta) \quad \text{for} \quad \gamma^2<1 \tag{22}$$
$$=\tanh[(2h-\sigma)/\theta] \quad \text{for} \quad \gamma^2=1\,,$$
$$h=\gamma\sigma \quad \text{for} \quad \gamma^2<1\,. \tag{23}$$

This solution is equivalent to a heuristic molecular field result previously obtained by Garrett.[21] Figure 2 shows σ, the reduced sublattice magnetization, and Fig. 3 shows a family of curves each for a particular γ (the cosine of half the angle of relative canting of the sublattices). The curve for $\gamma=1$ is the phase boundary $[h_c=\tanh(h_c/\theta_c)]$ across which the magnetization, $\gamma\sigma$, is continuous; whereas the susceptibility, $\chi=\partial(\gamma\sigma)/\partial h$, has a discontinuity $\Delta\chi=[h_c^2-(1-\theta_c)]/[\theta_c+(1-h_c^2)]$ shown in Fig. 4. Typical magnetization curves are plotted in Fig. 5, and it is clear that for this model the magnetization is temperature-independent within the region enclosed by the phase boundary.

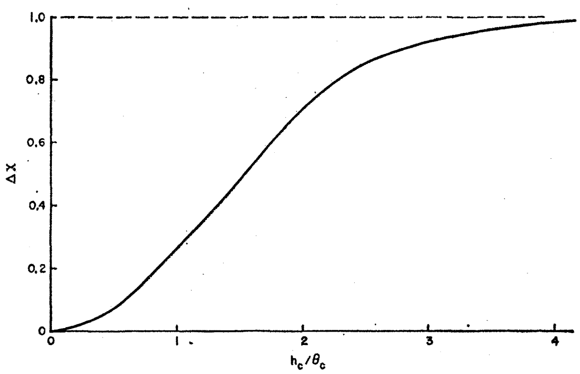

FIG. 4. Discontinuity in the magnetic susceptibility across the phase boundary (simple model).

We presented the above simple model to illustrate our method and to obtain a qualitative basis for discussing the behavior of the simple antiferromagnet in a magnetic field. One should note that at this time no proof is known of the existence of a phase transition for a system described by (1), and commonly held ideas about finite temperature magnetic phase transitions are based essentially on models resulting from modified forms of (1); e.g., molecular field and Ising models.

5. CASE FOR $(h-1)/\theta\gg 1$

We turn our attention back to the Heisenberg Hamiltonian (9) in rotated coordinates, and attempt to more carefully treat a simple cubic lattice of spin S per site. We focus on the particular region $h>1$, $\theta\ll 1$, and use the Holstein and Primakoff[22] expressions for the spin operators in terms of Boson operators:

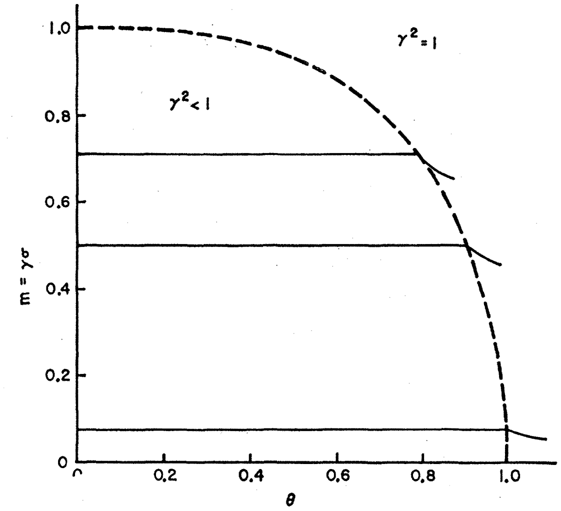

FIG. 5. Reduced magnetization versus temperature (simple model).

$$S_g^+=(2S)^{1/2}c_g^\dagger\left(1-\frac{c_g^\dagger c_g}{2S}\right)^{1/2},$$
$$S_g^-=(2S)^{1/2}\left(1-\frac{c_g^\dagger c_g}{2S}\right)^{1/2}c_g\,, \tag{24}$$
$$S_g^z=c_g^\dagger c_g-S\,,$$

with

$$c_g^\dagger c_g\leq 2S\,. \tag{25}$$

In all our calculations we replace (25) by the usual self-consistent approximation

$$\langle c_g^\dagger c_g\rangle_0\ll 2S\,. \tag{26}$$

We expect that for external fields which are sufficiently large compared to the critical field, the system will be accurately described at low temperatures by considering states of only a few spin excitations.

Now if

$$(h-1)/\theta\gg 1\,, \tag{27}$$

where

$$h=H/H_c$$
$$=g\mu H/4SJz\,; \tag{28}$$
$$\theta=kT/2SJz\,, \tag{29}$$

then we should be able to neglect terms like $c^{\dagger p}c^p$ for $p\geq 2$. On the physical consideration that at large fields the sublattice moments will be "dragged into parallelism," we take $\gamma=1$ so that the problem becomes *analogous* to the treatment of a low-temperature *ferromagnet* in a *small* magnetic field. The Hamiltonian with $\gamma=1$ is

$$\mathcal{H}=J\sum_f\sum_\delta(S_f^zS_{f+\delta}^z-S_f^+S_{f+\delta}^-)+g\mu H\sum_f S_f^z\,. \tag{30}$$

By using the leading terms in the expansion of (24), i.e., replacing $(1-(c^\dagger c/2S))^{1/2}$ by 1, the Fourier trans-

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[22] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

forms (12) (here interpreted for boson amplitudes) diagonalize the quadratic part of (30) which becomes

$$\mathcal{H}_0/2SNJz=N^{-1}\sum_{\mathbf{k}}\omega_{\mathbf{k}}c_{\mathbf{k}}^{\dagger}c_{\mathbf{k}}+(1-2h)S\,, \quad (31)$$

where the spectrum is

$$\omega_{\mathbf{k}}=2(h-1)+(1-\gamma_{\mathbf{k}})\,. \quad (32)$$

Since we are dealing with a noninteracting boson gas, we readily compute thermodynamic averages according to (3) and find the average energy

$$\langle\mathcal{H}\rangle_0/2SNJz=2(h-1)A+B+(1-2h)S\,, \quad (33)$$

and reduced magnetization

$$\begin{aligned}\sigma&=|\langle S_f^z\rangle_0|/S\\&=1-AS^{-1},\end{aligned} \quad (34)$$

where A and B are defined as in (14) and (15), except that for this (boson) system

$$n_{\mathbf{k}}=[\exp(\omega_k/\theta)-1]^{-1}\,. \quad (35)$$

Now for a simple cubic lattice of unit lattice constant, (16) is simply

$$\gamma_{\mathbf{k}}=\tfrac{1}{3}(\cos k_x+\cos k_y+\cos k_z)\,. \quad (36)$$

In the constant density limit of $N\rightarrow\infty$

$$\begin{aligned}A&=\frac{1}{(2\pi)^3}\int d^3k\; n_{\mathbf{k}}\\&\approx e^{-2(h-1)/\theta}\left[\theta'^{3/2}+\frac{3}{4}\pi\theta'^{5/2}+\frac{33}{32}\pi^2\theta'^{7/2}+0(\theta'^{9/2})\right]\\&\qquad+0(e^{-4(h-1)/\theta}\theta'^{3/2})\,,\end{aligned} \quad (37)$$

$$\begin{aligned}B&\;\frac{1}{(2\pi)^3}\int d^3k(1-\gamma_{\mathbf{k}})n_{\mathbf{k}}\\&\approx e^{-2(h-1)/\theta}\left[\pi\theta'^{5/2}+\frac{5}{4}\pi^2\theta'^{7/2}+\cdots\right]\\&\qquad+0(e^{-4(h-1)/\theta}\theta'^{5/2})\,,\end{aligned} \quad (38)$$

where

$$\theta'\equiv3\theta/2\pi\,, \quad (39)$$

and (27) obtains.

To lowest order we have the following results for the reduced magnetization, susceptibility, and specific heat, respectively:

$$\sigma=1-\left(\frac{3}{2\pi}\right)^{3/2}\frac{e^{-2(h-1)/\theta}}{S}\theta^{3/2}\,, \quad (40)$$

$$\chi=2\left(\frac{3}{2\pi}\right)^{3/2}\frac{e^{-2(h-1)/\theta}}{S}\theta^{+1/2}\,, \quad (41)$$

and

$$\frac{\partial\langle\mathcal{H}\rangle_0/N}{\partial T}=4k(h-1)^2e^{-2(h-1)/\theta}\theta^{-1/2}\,. \quad (42)$$

Observe that, as initially assumed, the deviation of σ from unity is exceedingly small for $(h-1)/\theta\gg1$. As $\theta\rightarrow0$ for $h>1$, the magnetization becomes saturated; whereas the susceptibility and specific heat both tend to zero.

6. PHASE BOUNDARY

To more accurately treat the region near the phase boundary, we introduce into (24) the truncated expansion

$$\left(1-\frac{c_{\mathbf{g}}^{\dagger}c_{\mathbf{g}}}{2S}\right)^{1/2}=1-\tfrac{1}{2}[1+(8S)^{-1}+0(S^{-2})]\frac{c_{\mathbf{g}}^{\dagger}c_{\mathbf{g}}}{2S}+0\left(\frac{c_{\mathbf{g}}^{\dagger p}c_{\mathbf{g}}^{p}}{S^2}\right)+0(S^{-3})\,,\quad(p\geq2)\,.$$

The resulting Hamiltonian contains quartic as well as quadratic terms in the c operators, and the free parameter γ will be selected variationally.

To the Fourier amplitudes $c_{\mathbf{k}}$, we apply the canonical transformation (5) in which $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ satisfy (6). Thermal averages are computed according to (3) and we find

$$\begin{aligned}&\langle\mathcal{H}\rangle_0/2SNJz\\&=(1-\gamma^2)\left[C-D-\frac{2A(C-D)+C(A-B)}{2S}\right]\\&\quad-\gamma^2\left[A-B-\frac{2A(A-B)+C(C-D)}{2S}\right]\\&\quad+(2\gamma^2-1)\left[\tfrac{1}{2}S-A+\frac{A^2+(A-B)^2+(C-D)^2}{2S}\right]\\&\quad+2h\gamma(A-S)\,,\end{aligned} \quad (43)$$

with

$$\begin{aligned}A&\equiv z^{-1}\sum_\delta\langle c_f^{\dagger}c_f\rangle_0=N^{-1}\sum_{\mathbf{k}}h_{\mathbf{k}}\,,\\B&\equiv z^{-1}\sum_\delta\langle c_f^{\dagger}c_f-c_f^{\dagger}c_{f+\delta}\rangle_0=N^{-1}\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}})h_{\mathbf{k}}\,,\\C&\equiv z^{-1}\sum_\delta\langle c_f^{\dagger}c_f^{\dagger}\rangle_0=N^{-1}\sum_{\mathbf{k}}\chi_{\mathbf{k}}\,,\\D&\equiv z^{-1}\sum_\delta\langle c_f^{\dagger}c_f^{\dagger}-c_f^{\dagger}c_{f+\delta}^{\dagger}\rangle_0=N^{-1}\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}})\chi_{\mathbf{k}}\,,\end{aligned} \quad (44)$$

and

$$\begin{aligned}h_{\mathbf{k}}&=v_{\mathbf{k}}^2(1+n_{\mathbf{k}})+n_{\mathbf{k}}(1+v_{\mathbf{k}}^2)\,,\\\chi_{\mathbf{k}}&=(1+2n_{\mathbf{k}})u_{\mathbf{k}}v_{\mathbf{k}}\,.\end{aligned} \quad (45)$$

The mean occupation number is $n_{\mathbf{k}}=\langle a_{\mathbf{k}}^{\dagger}a_{\mathbf{k}}\rangle_0$.

The requirement that F be stationary with respect to functional variations of $v_{\mathbf{k}}$, $n_{\mathbf{k}}$, $\epsilon_{\mathbf{k}}$, and variations of the parameter γ, leads to the extremum conditions:

$$\tfrac{1}{2}[(\xi_{\mathbf{k}}/\omega_{\mathbf{k}})-1], \tag{46}$$

$$u_{\mathbf{k}}v_{\mathbf{k}}=-\tfrac{1}{2}(\Delta_{\mathbf{k}}/\omega_{\mathbf{k}}), \tag{47}$$

$$\omega_{\mathbf{k}}=(\xi_{\mathbf{k}}{}^2-\Delta_{\mathbf{k}}{}^2)^{1/2}, \tag{48}$$

$$n_{\mathbf{k}}=[\exp(\omega_{\mathbf{k}}/\theta)-1]^{-1}, \tag{49}$$

$$\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial\gamma}=0, \quad (\gamma^2\lesssim 1), \tag{50}$$

$$\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial\gamma}<0, \quad (\text{for a local minimum at } \gamma=1). \tag{51}$$

The spectrum (48) is readily determined from

$$\xi_{\mathbf{k}}=\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial A}+(1-\gamma_{\mathbf{k}})\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial B}, \tag{52}$$

and

$$\Delta_{\mathbf{k}}=\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial C}+(1-\gamma_{\mathbf{k}})\frac{\partial\langle\mathcal{H}\rangle_0/2SNJz}{\partial D}. \tag{53}$$

Conditions (46) through (53) are to be taken in conjunction with (44) and (45).

At this point we refer to Appendixes A and B where we connect this approach to the low-temperature calculations previously referenced.[11,18,19] One will observe how our method simply reproduces the relatively well-established low-temperature ferromagnetic and antiferromagnetic results as special cases of a single variational calculation. With this vote of confidence, the phase boundary is approached.

We first differentiate (43) with respect to C and D and observe that all resulting terms contain either C, D, or $(1-\gamma^2)$. Thus as $\gamma^2\rightarrow 1$ we find the consistent solution $\Delta_{\mathbf{k}}=v_{\mathbf{k}}=C=D=0$; $\omega_{\mathbf{k}}=\xi_{\mathbf{k}}$. Consequently as we approach the phase boundary (defined by $\gamma=1$) from below, we pass from the nontrivial solution $v_{\mathbf{k}}$ and $\Delta_{\mathbf{k}}\neq 0$; $\omega_{\mathbf{k}}=(\xi_{\mathbf{k}}{}^2-\Delta_{\mathbf{k}}{}^2)^{1/2}$ to the trivial or identity solution $v_{\mathbf{k}}=\Delta_{\mathbf{k}}=0$, $\omega_{\mathbf{k}}=\xi_{\mathbf{k}}$. Ideally we would like to solve the coupled integral equations (44) with $v_{\mathbf{k}}\neq 0$ below the boundary; however, with $v_{\mathbf{k}}\neq 0$ the form of the spectrum leads to three-dimensional integrals which are formidable for all but very special cases. To treat the region below the boundary we select the trial function $v_{\mathbf{k}}=0$, and thus do not treat the *transformation* variationally. This selection does not alter the equations for the phase boundary which is approached from above where $v_{\mathbf{k}}$ already has vanished.

With the convenient notation

$$\sigma=1-(A/S), \tag{54}$$

$$\rho=(A-B)/S, \tag{55}$$

and $v_{\mathbf{k}}=0$, Eqs. (44)–(53) lead to

$$\begin{aligned} x&=[2h\gamma+(1-3\gamma^2)(\sigma-\rho)]/\theta,\\ 3y&=[(1-\gamma^2)\rho+\gamma^2(\sigma-\rho)]/\theta, \end{aligned} \tag{56}$$

$$\begin{aligned} h&=\gamma\left(\sigma-\rho+\frac{\rho^2}{\sigma}\right); \quad (\gamma^2<1),\\ h&\gtrsim\left(\sigma-\rho+\frac{\rho^2}{\sigma}\right); \quad (\gamma^2=1). \end{aligned} \tag{57}$$

$$\begin{aligned} \sigma&=1-S^{-1}\sum_{r=1}^{\infty}e^{-rx}[e^{-ry}I_0(ry)]^3,\\ \rho&=S^{-1}\sum_{r=1}^{\infty}e^{-rx}[e^{-ry}I_0(ry)]^2[e^{-ry}I_1(ry)], \end{aligned} \tag{58}$$

where

$$n_{\mathbf{k}}=\{\exp[x+3y(1-\gamma_{\mathbf{k}})]-1\}^{-1}. \tag{59}$$

We have used the standard notation and integral representation for the Bessel functions of imaginary argument

$$I_p(W)\equiv i^{-p}J_p(iW).$$

We first examine these equations for the leading low-temperature ($\theta\equiv kT/2SJz\ll 1$) behavior. With the asymptotic forms of Bessel functions of large argument we find for $\gamma=1$:

$$h=1-(2A/S)+0(B), \tag{60}$$

$$\omega_{\mathbf{k}}=(1-\gamma_{\mathbf{k}})+0(A), \tag{61}$$

$$A/S\approx a\theta^{3/2}/S, \quad (a=2S^{-1}\zeta(3/2)(3/2\pi)^{3/2}), \tag{62}$$

and

$$B/S=0(\theta^{5/2}).$$

Equation (60) gives the leading expression for the phase boundary

$$h\approx 1-a\theta^{3/2}, \quad (\theta\ll 1). \tag{63}$$

From (54) the reduced magnetization at the phase boundary is

$$\sigma\approx 1-\tfrac{1}{2}a\theta^{3/2}, \quad (\theta\ll 1). \tag{64}$$

Above the boundary we have

$$\omega_{\mathbf{k}}\approx 2(h-1)+4AS^{-1}+(1-\gamma_{\mathbf{k}}), \tag{65}$$

and the reduced magnetization is

$$\sigma\approx 1-\frac{a\theta^{3/2}}{2\zeta(3/2)}\sum_{m=1}^{\infty}e^{-[2(h-1)+4AS^{-1}]m/\theta}m^{-3/2}; \tag{66}$$

whereas the susceptibility is

$$\left(\frac{\partial\sigma}{\partial h}\right)_{\text{above}}\approx\frac{\pi^{1/2}a}{\zeta(3/2)}\frac{\theta}{(2(h-1)+2a\theta^{3/2}S^{-1})^{1/2}}. \tag{67}$$

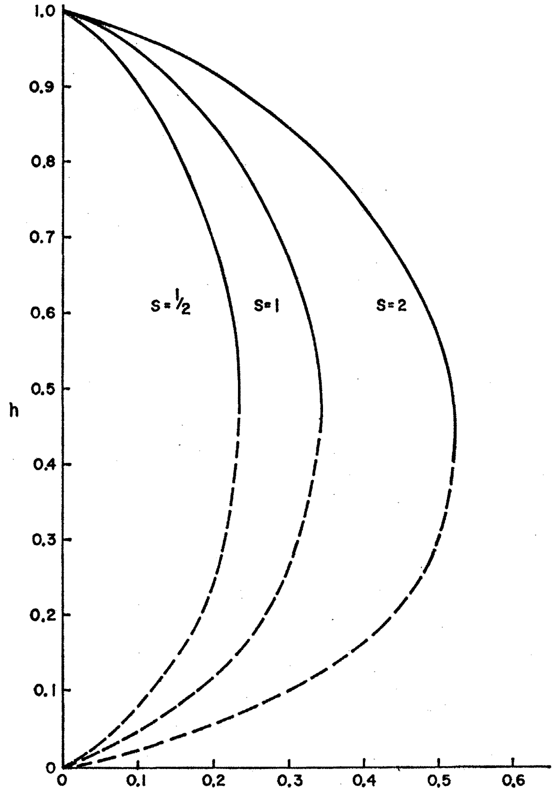

FIG. 6. Phase boundaries for $S=\frac{1}{2}$, 1, and 2 (IBM 709).

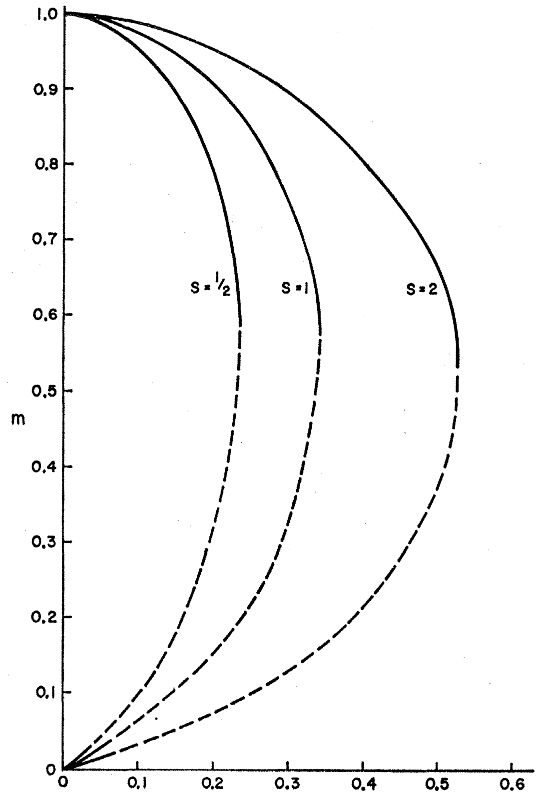

FIG. 7. Reduced magnetization along the phase boundaries (IBM 709).

Notice that (67) is valid far enough above the phase boundary so that $\partial\sigma/\partial h \ll 1$. If we try to approach the boundary from above for $\theta > 0$, the susceptibility suffers the same fluctuation[23] divergence as found for the susceptibility of the isotopic *ferro*magnetic when $h \to 0$; $\theta > 0$.

On the other hand for $\gamma^2 \lesssim 1$, Eq. (57) gives the lowest order result

$$\gamma \approx h(1+2AS^{-1}). \tag{68}$$

Since the net magnetization m is the projection of the sublattice magnetization σ, we have

$$m = \gamma(1-AS^{-1}). \tag{69}$$

Application of (60) and (68) demonstrates the continuity of the magnetization across the boundary. The susceptibility below the boundary is

$$\partial m/\partial h)_{\text{below}} \approx 1; \tag{70}$$

consequently, (67) and (70) suggest a discontinuity in the susceptibility across the boundary.

Even though the above theory is presumably most justified for $\theta \ll 1$, we were able to obtain the higher temperature behavior of the phase boundary curve. To accomplish this the coupled equations were kindly programed for the IBM-709 computer by John Wills. The phase boundaries for $S=\frac{1}{2}$, 1, and 2 are shown in Fig. 6, and the magnetization along the boundaries is given in Fig. 7. Notice that for higher spin values the magnetization at the maximum θ is decreasing. This result, as well as the degeneracy of the solution, are in resemblance to Bloch's calculation for the *ferro*magnet where $\theta_{\max}$ was suggestive of the Curie temperature. Since neither calculation has strong *a priori* justification for $\theta \approx 1$, the nonvanishing of the magnetization at $\theta_{\max}$ may manifest the inadequacy of the approximation used rather than the physical behavior of the system near the critical temperature.

[23] R. Kubo, Phys. Rev. 87, 568 (1952).

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APPENDIX A

It is easily seen that (50) is satisfied for $h=\gamma=0$. For this case we have

$$A=B,$$
$$C=0,$$
$$\xi_{\mathbf{k}}=p=1-(A-D)S^{-1},$$
$$\Delta_{\mathbf{k}}=p\gamma_{\mathbf{k}},$$

and the spectrum

$$\omega_{\mathbf{k}}=p(1-\gamma_{\mathbf{k}}^2)^{1/2}, \tag{A1}$$

which is the form exhibited by Keffer and Loudon.[19] By introducing the well-known quantities

$$K=\frac{1}{N}\sum_{\mathbf{k}}\frac{1-\gamma_{\mathbf{k}}}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}=\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}, \tag{A2}$$

$$K'=N^{-1}\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}}^2)^{1/2}, \tag{A3}$$

which have been evaluated[24] in the constant density limit $N\to\infty$, we obtain

$$A=B=\tfrac{1}{2}(K-1)+P(y), \tag{A4}$$

$$C=0, \tag{A5}$$

$$D=\tfrac{1}{2}(K-K')+P(y)+Q(y), \tag{A6}$$

where

$$P(y)=\frac{1}{(2\pi)^3}\int d^3k\frac{1}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}\cdot n_{\mathbf{k}}, \tag{A7}$$

$$Q(y)=\frac{1}{(2\pi)^3}\int d^3k(1-\gamma_{\mathbf{k}}^2)^{1/2}\cdot n_{\mathbf{k}}, \tag{A8}$$

$$y=\left[1+\frac{(1-K')-2Q(y)}{2S}\right]\Big/\theta;\ (\theta=kT/2SJz), \tag{A9}$$

$$n_{\mathbf{k}}=[\exp y(1-\gamma_{\mathbf{k}}^2)^{1/2}-1]^{-1}, \tag{A10}$$

and

$$K=1.156;\quad 1-K'=0.097.$$

For low temperatures it is easily verified that the reduced sublattice magnetization is

$$\begin{aligned}\sigma\equiv 1-AS^{-1}=1-\Bigg\{&\frac{0.156}{2}+\frac{3^{3/2}}{2\pi^2}\Bigg[\zeta(2)\left(1-\frac{(1-K')}{S}\right)\theta^2\\&+6\zeta(4)\left(1-\frac{2(1-K')}{S}\right)\theta^4\\&+234\zeta(6)\left(1-\frac{3(1-K')}{S}\right)\theta^6\Bigg]\\&+\left(\frac{3}{\pi}\right)^4\frac{\zeta(2)\zeta(4)}{S}\theta^6+\cdots\Bigg\}S^{-1},\end{aligned} \tag{A11}$$

in agreement with Oguchi's magnetization expression even though our renormalized spectrum

$$\omega_{\mathbf{k}}=\left[1+\frac{0.097}{2S}-\frac{3^{5/2}\zeta(4)}{2S\pi^2}\theta^4+0(\theta^6)\right](1-\gamma_{\mathbf{k}}^2)^{1/2}, \tag{A12}$$

agrees with Oguchi's spectrum only for $\theta=0$. We see that Keffer and Loudon's[19] results are thus obtainable variationally.

APPENDIX B

For the *ferro*magnetic we take $\gamma=1$ and J of opposite sign to find

$$\begin{aligned}A&=N^{-1}\textstyle\sum_{\mathbf{k}}n_{\mathbf{k}},\\B&=N^{-1}\textstyle\sum_{\mathbf{k}}(1-\gamma_{\mathbf{k}})n_{\mathbf{k}},\\C&=D=0,\end{aligned} \tag{B1}$$

with

$$n_{\mathbf{k}}=(e^{\omega_{\mathbf{k}}/\theta}-1)^{-1},\quad(\theta=kT/2SJz), \tag{B2}$$

and

$$\omega_{\mathbf{k}}=2h+(1-BS^{-1})(1-\gamma_{\mathbf{k}}). \tag{B3}$$

When $h=0$, the implicit spectrum is identical with Bloch's[11] result for the ferromagnet in zero external field. We easily obtain the low-temperature expressions for the spectrum

$$\begin{aligned}\omega_{\mathbf{k}}=[1-\pi S^{-1}\zeta(5/2)\theta'^{5/2}-(5/4)\pi^2S^{-1}\zeta(7/2)\theta'^{7/2}&\\+0(\theta'^{9/2})](1-\gamma_{\mathbf{k}}),&\end{aligned} \tag{B4}$$

and the reduced magnetization

$$\begin{aligned}\sigma=1-S^{-1}\zeta(3/2)[\theta'^{3/2}+(3/2)\pi S^{-1}\zeta(5/2)\theta'^4]&\\-(3/4)\pi S^{-1}\zeta(5/2)\theta'^{5/2}-(33/32)\pi^2S^{-1}\zeta(7/2)\theta'^{7/2}&\\+0(\theta'^{9/2}),\quad(\theta'=3\theta/2\pi),&\end{aligned} \tag{B5}$$

which coincide with the results of Keffer and Loudon.[19]

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[24] P. Anderson, Phys. Rev. **86**, 1 (1952).