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Effect of Crystal-Field Anisotropy on Magnetically Ordered Systems*†

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The statistical mechanics of a general spin-S magnetic system, which is described by a Heisenberg-Dirac isotropic exchange Hamiltonian with single-ion anisotropy included, has been studied with the aid of the Green-function technique. This problem has been set up in a new formalism in which it is *not* necessary to decouple the anisotropy Green functions. A scheme has been found for decoupling each of the exchange Green functions. For zero anisotropy, our results reduce to the usual random-phase approximation. For finite values of the anisotropy parameter D, the ensemble averages $\langle (S_g^z)^n \rangle$, for n integer, show a greater dependence on D than they do in the molecular-field-theory (MFT) calculation. Unlike the results of some of the previous decoupling schemes used on this problem, our prediction for the transition temperature $T_c(D)$, as a function of D, remains finite as the anisotropy becomes infinite. The asymptotic value of $T_c(D)$ as $D \to \infty$ for our Green-function calculation is the same as the asymptotic value of the MFT prediction, $T_c^{\mathrm{MFT}}(D \to \infty)$. We present here the appropriate formalism for antiferromagnetic as well as for ferromagnetic systems.

I. INTRODUCTION

Starting from the Hamiltonian

$$\mathcal{H} = -\sum_i \sum_j J_{ij} \vec{S}_i \cdot \vec{S}_j - \sum_i D (S_i^z)^2 , \qquad (1.1)$$

this paper will discuss in a quantitative way the effects of the crystal-field anisotropy D on the statistical mechanical quantities associated with a general-spin-S magnetic system. The first term in the Hamiltonian is the usual Heisenberg-Dirac exchange Hamiltonian which accounts for the isotropic coupling between the highly localized electrons in an insulating magnetic salt. The sums over i and j represent sums over all the lattice points which have a spin operator $\vec{S}$ associated with them. The second term represents the splitting of the spin states owing to the presence of a crystalline electric field.

The particular statistical mechanical quantities we shall be interested in here are the $2S$ moments of the operator S_i^z, $\langle (S_i^z)^n \rangle$, where $n = 1, 2, \ldots, 2S$, and also the transition temperature T_c. The angular brackets denote the canonical ensemble average: $\langle (S_i^z)^n \rangle = \mathrm{Tr}[(S_i^z)^n e^{-\mathcal{H}/\kappa T}] / \mathrm{Tr}(e^{-\mathcal{H}/\kappa T})$, where κ is the Boltzmann constant. The temperature range to be considered is $0 \leq T \leq T_c$.

The molecular-field-theory (MFT) approximation is presented in Sec. II. The moments are calculated for this theory and it is pointed out that these quantities change very little even with large changes in D. This indicates that the MFT underestimates the effect of the crystal-field anisotropy since we know that these moments should change considerably over the range $0 \leq D \leq \infty$.

Section III presents the rudiments of the Green-function formalism and also the earlier decoupling schemes of Narath,[1] Anderson and Callen,[2] and Lines.[3] Defects in each of these schemes are pointed out, the most serious being the prediction of $T_c \to \infty$ as $D \to \infty$. These defects each arise through attempts to perform a random-phase approximation (RPA) on the anisotropy Green function itself.

The particular formalism of this paper is introduced in Sec. IV. We first point out that the problem can easily be set up so that *none* of the anisotropy Green functions need to be decoupled. Second, we then decouple the exchange-term Green functions in a manner which is consistent with the usual RPA. We show that it is possible to obtain $2S$ independent equations in the $2S$ unknown moments, thereby determining all the necessary statistical information. The solution of the problem for an antiferromagnetic lattice is then covered in Sec. V. The method for obtaining expressions for T_c is presented in Sec. VI.

Numerical solutions of the equations have been obtained for the spin $S = 1$ ferromagnet, the spin $S = 1$ antiferromagnet, and the spin $S = \frac{3}{2}$ ferromagnet, and these are presented in Sec. VII. Graphs have been plotted for the more important results. All of the results show a stronger dependence on D than the equivalent results obtained from the MFT.

Murao and Matsubara[4] have attempted a decoupling of the exchange terms such as ours for the spin $S = 1$ ferromagnet. Their final results, though, differ considerably from ours because of their particular choice for the spin operators inside the Green function. A discussion of their errors and also a comment on Green-function decoupling procedures in general are included in the Appendix.

II. MFT

The MFT is, by far, the simplest of all the approximations to apply. Consider the Hamiltonian for the ith spin:

$$\mathcal{H}_i = -2\textstyle\sum_j J_{ij}\vec{S}_i\cdot\vec{S}_j - D(S_i^z)^2 . \qquad (2.1)$$

The molecular-field approximation assumes the form of replacing $\vec{S}_j$ by its ensemble average $\langle S_j^z\rangle$. Then the molecular-field Hamiltonian for a single spin becomes

$$\mathcal{H}_i^{\mathrm{MFT}} = -2\textstyle\sum_j J_{ij}\langle S_j^z\rangle S_i^z - D(S_i^z)^2 , \qquad (2.2)$$

and for the total Hamiltonian we then have

$$\mathcal{H}^{\mathrm{MFT}} = -2\textstyle\sum_i\sum_j J_{ij}\langle S_j^z\rangle S_i^z - D\sum_i(S_i^z)^2 . \qquad (2.3)$$

Since $\langle S_i^z\rangle$ is independent of the lattice-site index i, we may define $\bar{S} = \langle S_i^z\rangle$. We shall also define $J(0) = \sum_j J_{ij}$. Then the eigenvalues of $\mathcal{H}_i^{\mathrm{MFT}}$ are

$$E_i(m) = -2\bar{S}J(0)m - Dm^2 , \qquad (2.4)$$

where m is the azimuthal quantum number which ranges in integral steps from $-S$ to $+S$. By the definition of the ensemble average we have for the moments

$$\langle (S_i^z)^n\rangle = \sum_{m=-S}^{m=S} m^n e^{[2\bar{S}J(0)m + Dm^2]/\kappa T} \Big/ \sum_{m=-S}^{m=S} e^{[2\bar{S}J(0) + Dm^2]/\kappa T} \qquad (2.5)$$

In order to obtain expressions for T_c we let $\bar{S}\to 0$. Using $n = 1$ in (2.5) one obtains in this limit

$$\kappa T_c = 2J(0)\sum_{m=-S}^{m=S} m^2 e^{Dm^2/\kappa T_c} \Big/ \sum_{m=-S}^{m=S} e^{Dm^2/\kappa T_c} \qquad (2.6)$$

or, equivalently,

$$\kappa T_c = 2J(0)\langle (S_i^z)^2\rangle\big|_{T=T_c} . \qquad (2.7)$$

Two special cases of this are as follows:

$$\kappa T_c = \tfrac{2}{3}S(S+1)J(0) \quad \text{for } D = 0 , \qquad (2.8\text{a})$$

$$\kappa T_c = 2S^2 J(0) \quad \text{for } D = \infty . \qquad (2.8\text{b})$$

Figures 3 and 4 of Sec. VII contain, respectively, plots of $\bar{S}$ and $\langle (S^z)^2\rangle$ as functions of temperature for a spin $S = 1$ simple cubic ferromagnet. These MFT curves do not change shape noticeably over the whole range of possible anisotropies. Clearly, then, the MFT grossly underestimates the effect of anisotropy on magnetic systems.

III. GREEN-FUNCTION TECHNIQUE AND PREVIOUS DECOUPLING SCHEMES

We shall use Zubarev's[5] definition of the double-time temperature-dependent Green function:

$$\langle\langle A(t); B(t')\rangle\rangle_E = -i\,\theta(t-t')\langle [A(t), B(t')]\rangle , \qquad (3.1)$$

where the double angular brackets denote the Green function, and the square brackets denote the commutator. By Fourier transforming with respect to the variable $t - t'$ one may obtain a Green function $\langle\langle A; B\rangle\rangle_E$ which satisfies the equation of motion

$$E\langle\langle A; B\rangle\rangle_E = (1/2\pi)\langle [A, B]\rangle + \langle\langle [A, \mathcal{H}]; B\rangle\rangle_E , \qquad (3.2)$$

where E is now the dependent variable. The Green function $\langle\langle A; B\rangle\rangle_E$ can be related back to the time-dependent correlation function $\langle B(t')A(t)\rangle$ via the spectral theorem

$$\langle B(t')A(t)\rangle = i\lim_{\epsilon\to 0}\int_{-\infty}^{+\infty} \frac{\langle\langle A; B\rangle\rangle_{E=\omega+i\epsilon} - \langle\langle A; B\rangle\rangle_{E=\omega-i\epsilon}}{e^{\omega/\kappa T} - 1} \times e^{-i\omega(t-t')}d\omega . \qquad (3.3)$$

If $\langle\langle A; B\rangle\rangle_E$ has only simple poles, (3.3) can be simplified greatly in the following way: Suppose $\langle\langle A; B\rangle\rangle_E$ has the form

$$\langle\langle A; B\rangle\rangle_E = \frac{1}{2\pi}\sum_{i=1}^{n}\frac{A_i}{E - E_i} , \qquad (3.4)$$

where the E_i are the location of the n simple poles, and the A_i are their residues. Then using (3.3) yields

$$\langle B(t')A(t)\rangle = \sum_{i=1}^{n}\frac{A_i}{e^{E_i/\kappa T} - 1} e^{-iE_i(t-t')} . \qquad (3.5)$$

This relationship will be useful later in this paper.

The usual procedure for magnetic problems is to choose the operator A to be S_g^+ and the operator B to be some function of the spin operators associated with lattice site h. We denote this second operator as B_h for the time being. By considering the equation of motion of the Green function $\langle\langle S_g^+; B_h\rangle\rangle$ as generated by the Hamiltonian in (1.1) one has

$$\begin{aligned} E\langle\langle S_g^+; B_h\rangle\rangle = {} & (1/2\pi)\,\delta_{g,h}\langle [S_g^+, B_g]\rangle \\ & + 2\textstyle\sum_f J_{gf}\langle\langle (S_f^z S_g^+ - S_g^z S_f^+); B_h\rangle\rangle \\ & + D\langle\langle (S_g^+ S_g^z + S_g^z S_g^+); B_h\rangle\rangle , \end{aligned} \qquad (3.6)$$

where $\delta_{g,h}$ is the Kronecker δ function. (Note that the subscript E has now been dropped from each of the Green functions, since all further Green functions that we will be using are of this transformed variety.) Clearly, one cannot solve this equation for the Green function $\langle\langle S_g^+; B_h\rangle\rangle$ without first determining the exchange function $\langle\langle (S_g^+ S_f^z - S_g^z S_f^+); B_h\rangle\rangle$ and the anisotropy function $\langle\langle (S_g^+ S_g^z + S_g^z S_g^+); B_h\rangle\rangle$. Decoupling procedures are generally employed at this point to reduce these higher-order functions to the original Green function. The exchange function can be decoupled by the RPA[6]:

$$\langle\langle S_f^z S_g^+ ; B_h\rangle\rangle \cong \langle S_f^z\rangle \langle\langle S_g^+ ; B_h\rangle\rangle \quad \text{for } f \neq g . \qquad (3.7)$$

This approximation, or factorization, says, in effect, that the transverse motion of the spin at site g is completely uncorrelated with the longitudinal motion of the spin at site f. This is certainly a reasonable approximation as long as $f \neq g$. For the special case $f = g$, such an approximation is less valid since we can no longer separate the longitudinal and transverse motions from each other. In fact such motions must be intimately correlated when they refer to the same lattice site. Nevertheless there have been three distinct attempts to decouple the anisotropy function. They are each of the form

$$\langle\langle (S_g^z S_g^+ + S_g^+ S_g^z); B_h\rangle\rangle \cong \Gamma_g \langle\langle S_g^+ ; B_h\rangle\rangle . \qquad (3.8)$$

The three choices made for Γ_g have been

Narath:

$$\Gamma_g = 2\langle S_g^z\rangle , \qquad (3.9)$$

Anderson and Callen:

$$\Gamma_g = [2 - (1/2S^2)\langle S_g^z S_g^+ + S_g^+ S_g^z\rangle]\langle S_g^z\rangle , \qquad (3.10)$$

Lines:

$$\Gamma_g = \langle 3(S_g^z)^2 - S(S+1)\rangle / \langle S_g^z\rangle \quad \text{for } B_h = S_h^- , \qquad (3.11\text{a})$$

$$\Gamma_g = \frac{\langle 2(S_g^z)^4 + 3(S_g^z)^3 - (2S^2 + 2S - 1)(S_g^z)^2 - S(S+1)S_g^z\rangle}{\langle (S_g^z)^3 + (S_g^z)^2 - S(S+1)S_g^z\rangle} \quad \text{for } B_h = S_h^z S_h^- . \qquad (3.11\text{b})$$

Each of these approximations produces a number of invalid results. The first two approximations predict that at $T = T_c$, $\langle (S_g^z)^2\rangle = \frac{1}{3}S(S+1)$ for all values of the parameter D. But only for an isotropic system can $\langle (S_g^z)^2\rangle = \frac{1}{3}S(S+1)$. For all finite values of D one must have $\langle (S_g^z)^2\rangle$ larger than $\frac{1}{3}S(S+1)$. All three approximations predict $T_c(D) \to \infty$ as $D \to \infty$. This is an unphysical result for the following reason: As $D \to \infty$ the effective spin state at each site becomes just $|S\rangle$ and $|-S\rangle$. Even in this case the spin system must still possess a finite transition temperature T_c. This particular T_c is, of course, somewhat larger than if $D = 0$, but it is still finite.

These three decoupling approximations are at best valid only for D very small.

IV. GENERAL SPIN-S SOLUTION

The problem of decoupling the anisotropy function can be avoided by writing the equation of motion of the anisotropy function. This procedure will generate a new anisotropy function. By continuing this process of writing the equation of motion of each anisotropy function generated, one will see that, after $2S$ such equations, no new linearly independent anisotropy functions will be generated. Hence this set of $2S$ equations will be closed in the anisotropy functions. Therefore we will not need to decouple any of them. The problem then reduces to finding a decoupling appropriate for the exchange functions generated.

To keep track of all the spin operators necessary for this problem we make the following definitions for the operators used inside of the anisotropy functions:

$$A_g^1 \equiv S_g^+ , \qquad (4.1\text{a})$$

$$A_g^i \equiv -[A_g^{i-1}, (S_g^z)^2] \quad \text{for } 2S \geq i \geq 2 . \qquad (4.1\text{b})$$

Second, we shall define additional sets of operators $\{B_g^i\}$ and $\{C_g^i\}$ as follows:

$$B_g^i \equiv [A_g^i, S_g^+] \quad \text{for each } i , \qquad (4.2\text{a})$$

$$C_g^i \equiv [A_g^i, S_g^-] \quad \text{for each } i . \qquad (4.2\text{b})$$

Note that the A_g^i each satisfy

$$[A_g^i, S_g^z] = -A_g^i \quad \text{for each } i . \qquad (4.3)$$

The particular commutation relationship that closes the set of anisotropy operators is

$$[A_g^{2S}, (S_g^z)^2] = \sum_{j=1}^{2S-1} a_j A_g^j , \qquad (4.4)$$

where the particular values for the c numbers a_j depend on the size of the spin of the system.

The equations of motion for these anisotropy functions then assume the form

$$E\langle\langle A_g^i ; S_h^-\rangle\rangle = (1/2\pi)\delta_{g,h}\langle C_g^i\rangle + D\langle\langle A_g^{i+1} ; S_h^-\rangle\rangle + \textstyle\sum_f J_{fg}\langle\langle (2A_g^i S_f^z - B_g^i S_f^- - C_g^i S_f^+); S_h^-\rangle\rangle \quad \text{for } i \leq 2S - 1 , \qquad (4.5\text{a})$$

$$E\langle\langle A_g^{2S} ; S_h^-\rangle\rangle = \frac{1}{2\pi}\delta_{g,h}\langle C_g^{2S}\rangle - D\sum_{j=1}^{2S-1} a_j \langle\langle A_g^j ; S_h^-\rangle\rangle + \sum_f J_{fg}\langle\langle (2A_g^{2S} S_f^z - B_g^{2S} S_f^- - C_g^{2S} S_f^+); S_h^-\rangle\rangle . \qquad (4.5\text{b})$$

The decoupling of the exchange terms that we have chosen is as follows:

$$\langle\langle A_g^i S_f^z ; S_h^-\rangle\rangle \cong \langle S_f^z\rangle \langle\langle A_g^i ; S_h^-\rangle\rangle , \qquad (4.6\text{a})$$

$$\langle\langle B_g^i S_f^- ; S_h^-\rangle\rangle \cong 0 , \qquad (4.6\text{b})$$

$$\langle\langle C_g^i S_f^+ ; S_h^-\rangle\rangle \cong \langle C_g^i\rangle \langle\langle S_f^+ ; S_h^-\rangle\rangle . \qquad (4.6\text{c})$$

The operators C_g^i are linear combinations of the powers of the operator S_g^z and hence correspond to longitudinal motions of the spins. The operators A_g^i and B_g^i are of the form of raising and lowering operators and therefore represent the transverse motions of the spins. Since it is only the operators C_g^i which have finite ensemble averages, the decouplings in (4.6) represent the same type of factorizations as in (3.7).

Assuming translational invariance, we may define

$$G^i(\vec{k},E)=\sum_{\vec{R}_g-\vec{R}_h}\langle\langle A_g^i;S_h^-\rangle\rangle e^{i\vec{k}\cdot(\vec{R}_g-\vec{R}_h)}, \tag{4.7a}$$

$$J(\vec{k})=\sum_{\vec{R}_g-\vec{R}_h}J_{gh}e^{-i\vec{k}\cdot(\vec{R}_g-\vec{R}_h)}, \tag{4.7b}$$

$$Z_i=\langle C_g^i\rangle . \tag{4.7c}$$

Then (4.5) can be written more concisely as

$$[E-2\bar{S}J(0)]\,G^i(\vec{k},E)+Z_iJ(\vec{k})G^1(\vec{k},E)$$
$$-DG^{i+1}(\vec{k},E)=Z_i/2\pi \quad \text{for } i\leqslant 2S-1\ , \tag{4.8}$$

$$[E-2\bar{S}J(0)]\,G^{2S}(\vec{k},E)+Z_{2S}J(\vec{k})G^1(\vec{k},E)$$
$$+\sum_{j=1}^{2S-1}Da_jG^j(\vec{k},E)=Z_{2S}/2\pi . \tag{4.9}$$

This set of 2S linear equations can easily be solved. The solution for each Green function can be written in the form

$$G^i(\vec{k},E)=\frac{1}{2\pi}\sum_{j=1}^{2S}\frac{R_j^i(\vec{k})}{E-E_j(\vec{k})}, \tag{4.10}$$

where the $E_j(\vec{k})$ are the 2S possible poles of the Green functions, and $R_j^i(\vec{k})$ is the residue of the *j*th pole for the *i*th Green function. By Fourier-transforming back into the spatial variable and using the relationship in (3.5), the equal-time correlation function can be written as

$$\langle S_h^-A_g^i\rangle=\frac{1}{N}\sum_{\vec{k}}\sum_{j=1}^{2S}\frac{R_j^i(\vec{k})e^{-i\vec{k}\cdot(\vec{R}_g-\vec{R}_h)}}{e^{E_j(\vec{k})/\kappa T}-1}, \tag{4.11}$$

where we have assumed that there are N points in the lattice and where the sum over $\vec{k}$ is performed over all points in the first Brillouin zone of the lattice. For $g=h$ this becomes

$$\langle S_g^-A_g^i\rangle=\frac{1}{N}\sum_{\vec{k}}\sum_{j=1}^{2S}R_j^i(\vec{k})/(e^{E_j(\vec{k})/\kappa T}-1)$$
$$\text{for } i=1,\ 2,\ \ldots,\ 2S . \tag{4.12}$$

The operators $S_g^-A_g^i$ can be expressed directly as linear combinations of the powers of S_g^z, and therefore the left-hand side of (4.12) can be written as a linear combination of the moments. Hence the 2S equations of (4.12) completely determine the 2S moments for all temperatures. Numerical methods must be used in obtaining solutions of the equations in (4.12) since they cannot be solved analytically.

For the special case $D=0$ the poles and their residues simplify as

$$E_i(\vec{k})=2\bar{S}[J(0)-J(\vec{k})] \quad \text{for each } i\ , \tag{4.13a}$$

$$R_j^i(\vec{k})=Z_i\delta_{i,j} . \tag{4.13b}$$

Therefore the equations in (4.12) reduce to

$$\langle S_g^-A_g^i\rangle=Z_i\Phi=\langle[A_g^i,S_g^-]\rangle\,\Phi\ , \tag{4.14}$$

where

$$\Phi=(1/N)\textstyle\sum_k(e^{E(\vec{k})/\kappa T}-1)^{-1}, \tag{4.15a}$$

$$E(\vec{k})=2\bar{S}[J(0)-J(\vec{k})] . \tag{4.15b}$$

Equations (4.14) and (4.15) are the same relations Tahir-Kheli and ter Haar[6] obtained for their solution of the general spin S isotropic ferromagnet. Hence our decoupling procedure (4.6) is completely equivalent to the usual RPA when $D=0$.

Finally, we note that setting $J(\vec{k})$ equal to zero in (4.12) will reproduce the results of the MFT. This can easily be seen by noting that we would have gotten Eqs. (4.12) with the $J(\vec{k})$ terms set equal to zero if we had started out by using the MFT Hamiltonian of (2.3), rather than the Hamiltonian of (1.1).

V. ANTIFERROMAGNETIC SOLUTION

The solution for an antiferromagnetic lattice can be carried out along the lines set out by Anderson and Callen[2] and Lines.[7] We shall assume that the lattice can be divided up into two sublattices, the first of which contains the "up" spins and the second of which contains the "down" spins. The extension to cases where there are more than two sublattices can be deduced from this case rather easily.

The points on the first sublattice will be denoted by indices of the form g_1, and those on the second sublattice by g_2 indices. The equations in (4.5) then become

$$E\langle\langle A_{g_\mu}^i;S_{h_1}^-\rangle\rangle=\frac{1}{2\pi}\delta_{g_\mu,h_1}\langle C_g^i\rangle+\sum_{\nu=1}^{2}\sum_{f_\nu}J_{f_\nu g_\mu}$$
$$\times\langle\langle(2A_{g_\mu}^iS_{f_\nu}^z-B_{g_\mu}^iS_{f_\nu}^--C_{g_\mu}^iS_{f_\nu}^+);S_{h_1}^-\rangle\rangle$$
$$+D\langle\langle A_{g_\mu}^{i+1};S_{h_1}^-\rangle\rangle \quad \text{for } \mu=1,2\ , \tag{5.1a}$$

$$E\langle\langle A_{g_\mu}^{2S};S_{h_1}^-\rangle\rangle=\frac{1}{2\pi}\delta_{g_\mu,h_1}\langle C_g^{2S}\rangle+\sum_{\nu=1}^{2}\sum_{f_\nu}J_{f_\nu g_\mu}$$
$$\times\langle\langle(2A_{g_\mu}^{2S}S_{f_\nu}^+-B_{g_\mu}^{2S}S_{f_\nu}^--C_{g_\mu}^{2S}S_{f_\nu}^z);S_{h_1}^-\rangle\rangle$$
$$-D\sum_{j=1}^{2S-1}a_j\langle\langle A_{g_\mu}^j;S_{h_1}^-\rangle\rangle \quad \text{for } \mu=1,2 . \tag{5.1b}$$

We shall decouple the Green functions in (5.1) using the same scheme as in (4.6). Using the translational invariance of each sublattice we may define

$$G_\mu^i(\vec{k},E)=\sum_{\vec{R}_{g_\mu}-\vec{R}_{h_1}}\langle\langle A^i_{g_\mu};S^-_{h_1}\rangle\rangle\, e^{i\vec{k}\cdot(\vec{R}_{g_\mu}-\vec{R}_{h_1})}\,, \tag{5.2a}$$

$$J_{\mu\nu}(\vec{k})=\sum_{\vec{R}_{g_\mu}-\vec{R}_{f_\nu}} J_{g_\mu f_\nu}\, e^{-i\vec{k}\cdot(\vec{R}_{g_\mu}-\vec{R}_{f_\nu})}\,, \tag{5.2b}$$

where μ and ν assume the values 1 and 2. For an antiferromagnet the quantities $\langle C_g^i\rangle$ are no longer independent of the lattice site index. The ensemble averages of the odd powers of S_g^z now have different signs on each of the sublattices. It can be shown that C_g^i is a linear combination of the odd powers of S_g^z for i odd, and a linear combination of the even powers of S_g^z for i even. Therefore $\langle C_g^i\rangle$ changes sign from one sublattice to the next only when i is odd. Hence we may define for each i

$$\langle C^i_{g_\mu}\rangle = Z_i \qquad \text{for } \mu=1\,, \tag{5.3a}$$

$$\langle C^i_{g_\mu}\rangle = (-1)^i Z_i \quad \text{for } \mu=2\,. \tag{5.3b}$$

This can be written more succinctly as

$$\langle C^i_{g_\mu}\rangle=[\delta_{\mu,1}+\delta_{\mu,2}(-1)^i]Z_i \quad \text{for each } i\,. \tag{5.4}$$

After decoupling and Fourier transforming, (5.1) becomes

$$\begin{aligned}EG^i_\mu(\vec{k},E)&=Z_i\,\delta_{\mu,1}/2\pi+2\bar{S}\textstyle\sum_\nu J_{\nu\mu}(0)\,(-1)^{\nu+1}G^i_\mu(\vec{k},E)\\&\quad-Z_i\textstyle\sum_\nu J_{\nu\mu}(\vec{k})\,G^1_\nu(\vec{k},E)\,[\delta_{\mu,1}+\delta_{\mu,2}(-1)^i]\\&\quad-D\,G^{i+1}_\mu(\vec{k},E)\\&\qquad\qquad\text{for } \mu,\nu=1,\ 2 \text{ and } i\leqslant 2S-1\,,\end{aligned} \tag{5.5a}$$

$$\begin{aligned}EG^{2S}_\mu(\vec{k},E)&=Z_{2S}\,\delta_{\mu,1}/2\pi+2\bar{S}\textstyle\sum_\nu J_{\nu\mu}(0)\\&\quad\times(-1)^{\nu+1}G^{2S}_\mu(\vec{k},E)\\&\quad-Z_{2S}\textstyle\sum_\nu J_{\nu\mu}(\vec{k})\,G^1_\nu(\vec{k},E)\,[\delta_{\mu,1}+\delta_{\mu,2}(-1)^{2S}]\\&\quad-D\textstyle\sum_j a_j G^j_\mu(\vec{k},E)\quad\text{for } \mu,\nu=1,\ 2\,.\end{aligned} \tag{5.5b}$$

This set of $4S$ linear equations can easily be solved for each of the Green functions. The functions of the form $G^i_1(\vec{k},E)$ can be related back to the correlation function $\langle S^-_{g_1}A^i_{g_1}\rangle$ with the aid of the spectral theorem. Since the poles of the Green function come in positive and negative pairs for an antiferromagnet, $G^i_1(\vec{k},E)$ has the form

$$G^i_1(\vec{k},E)=\frac{1}{2\pi}\sum_{m=1}^{2S}\left(\frac{R^i_m(\vec{k})}{E-E_m(\vec{k})}+\frac{T^i_m(\vec{k})}{E+E_m(\vec{k})}\right)\,, \tag{5.6}$$

where the $E_m(\vec{k})$ are the positive poles, the $R^i_m(\vec{k})$ are the residues of the positive poles, and the $T^i_m(\vec{k})$ are the residues of the negative poles. Relating this Green function back to the correlation function yields

$$\begin{aligned}\langle S^-_{g_1}A^i_{g_1}\rangle=\frac{2}{N}\sum_{\vec{k}}\sum_{m=1}^{2S}\,&[R^i_m(\vec{k})\,(e^{E_m(\vec{k})/\kappa T}-1)^{-1}\\&+T^i_m(\vec{k})(e^{-E_m(\vec{k})/\kappa T}-1)^{-1}]\,,\end{aligned} \tag{5.7}$$

where we have assumed that there are N points in the full lattice. The sum over $\vec{k}$ is over all points in the first Brillouin zone of the sublattice. This set of $2S$ equations completely determines the $2S$ moments for all temperatures.

VI. TRANSITION TEMPERATURE

The transition temperature T_c is defined as the temperature at which $\bar{S}$ first vanishes as we increase T from absolute zero. We note here also that when $\bar{S}$ vanishes, *all* of the odd moments of S_g^z must vanish too. Since the Z_i, for i odd, depend only on the odd moments of S_g^z, they vanish likewise. Some care must be taken in trying to obtain a set of equations which can be solved for T_c. By taking the limit $\bar{S}\to 0$ in either of the $2S$ ferromagnetic equations of (4.12) or the $2S$ antiferromagnetic equations of (5.7), we will obtain S equations ($S-\frac{1}{2}$ equations) in the S unknown ($S-\frac{1}{2}$ unknown) Z_i for i even when S is integer (S is half-integer). These equations are valid for all $T\geqslant T_c$. That is, the limit-taking process yields a set of equations which are valid over the whole paramagnetic region, and they in no way determine the value of T_c.

The correct procedure for obtaining the value of T_c is as follows: Define

$$\beta_i=Z_i/\bar{S} \quad \text{for } i \text{ odd.} \tag{6.1}$$

($\beta_1\equiv 2$ for all spin systems since $Z_1=2\bar{S}$.) Then replace each Z_i for i odd with the quantity $\beta_i\bar{S}$ in the $2S$ original equations, i.e., (4.12) or (5.7). Now expand these equations in a power series in $\bar{S}$ about the point $\bar{S}=0$. The coefficients of each power of $\bar{S}$ must be the same on each side of each equation. A comparison of the zeroth power of $\bar{S}$ will yield the paramagnetic equations referred to in the paragraph above. The coefficients of the first powers of $\bar{S}$ in each equation will yield S additional ($S+\frac{1}{2}$ additional) equations when S is an integer (S is a half-integer). Hence we will then have a total of $2S$ equations. The $2S$ unknowns are then the Z_i for i even, the β_i for i odd ($i\neq 1$), and T_c. The reason T_c now appears as an unknown is that the comparison of the coefficients for the first power of $\bar{S}$ gives equations which are *not* valid for $T>T_c$, and hence they are valid only at the single temperature T_c, which is, as yet, undetermined. Numerical solution of the equations then yields T_c and the quantities Z_i for i even and the β_i for i odd which are all evaluated at $T = T_c$.

VII. RESULTS FOR SOME SPECIAL CASES

A. Spin $S = 1$ Ferromagnet

For $S = 1$ we need only the operators A_g^1 and A_g^2, which are S_g^+ and $S_g^z S_g^+ + S_g^+ S_g^z$, respectively. Then the equations of motion of (4.8) become

$$\begin{bmatrix} E - 2\bar{S}[J(0) - J(\vec{k})] & -D \\ Z_2 J(\vec{k}) - D & E - 2\bar{S}J(0) \end{bmatrix} \begin{bmatrix} G^1(\vec{k}, E) \\ G^2(\vec{k}, E) \end{bmatrix} = \frac{1}{2\pi} \begin{bmatrix} 2\bar{S} \\ Z_2 \end{bmatrix}. \tag{7.1}$$

The locations of the poles of the Green functions are gotten by equating the determinant of the coefficient matrix in (7.1) to zero and solving for the roots of this equation. The results are

$$E_1(k) = \bar{S}[2J(0) - J(\vec{k})] + \{D^2 - Z_2 D J(\vec{k}) + [\bar{S}J(\vec{k})]^2\}^{1/2}, \tag{7.2a}$$

$$E_2(k) = \bar{S}[2J(0) - J(\vec{k})] - \{D^2 - Z_2 D J(\vec{k}) + [\bar{S}J(\vec{k})]^2\}^{1/2}. \tag{7.2b}$$

Equations (4.11) reduce to the following for this case:

$$\langle S_g^- S_g^+ \rangle = \frac{1}{N} \sum_{\vec{k}} \left(\frac{R_1^1(\vec{k})}{e^{E_1(\vec{k})/\kappa T} - 1} + \frac{R_2^1(\vec{k})}{e^{E_2(\vec{k})/\kappa T} - 1} \right), \tag{7.3a}$$

$$\langle S_g^- (S_g^z S_g^+ + S_g^+ S_g^z) \rangle = \frac{1}{N} \sum_{\vec{k}} \left(\frac{R_1^2(\vec{k})}{e^{E_1(\vec{k})/\kappa T} - 1} + \frac{R_2^2(\vec{k})}{e^{E_2(\vec{k})/\kappa T} - 1} \right), \tag{7.3b}$$

where the residues are

$$R_1^1(\vec{k}) = [2\bar{S}E_1(\vec{k}) - 4\bar{S}^2 J(0) + Z_2 D] / [E_1(\vec{k}) - E_2(\vec{k})], \tag{7.4a}$$

$$R_2^1(\vec{k}) = [2\bar{S}E_2(\vec{k}) - 4\bar{S}^2 J(0) + Z_2 D] / [E_2(\vec{k}) - E_1(\vec{k})], \tag{7.4b}$$

$$R_1^2(\vec{k}) = [Z_2 E_1(\vec{k}) - 2\bar{S} Z_2 J(0) + 2\bar{S}D] / [E_1(\vec{k}) - E_2(\vec{k})], \tag{7.4c}$$

$$R_2^2(\vec{k}) = [Z_2 E_2(\vec{k}) - 2\bar{S} Z_2 J(0) + 2\bar{S}D] / [E_2(\vec{k}) - E_1(\vec{k})]. \tag{7.4d}$$

By writing out the angular momentum matrices explicitly one finds that

$$Z_2 = 6\langle (S^z)^2 \rangle - 4, \tag{7.5a}$$

$$\langle S^- S^+ \rangle = \tfrac{1}{6}(8 - 6\bar{S} - Z_2), \tag{7.5b}$$

$$\langle S^- (S^z S^+ + S^+ S^z) \rangle = \bar{S} - \tfrac{1}{2} Z_2. \tag{7.5c}$$

For any given temperature (7.3) together with (7.4) and (7.5) determine $\bar{S}$ and Z_2 completely in terms of the exchange integrals J_{ij} and the anisotropy D. Using the technique outlined in Sec. VI one obtains the following equations which determine T_c and Z_2 evaluated at T_c:

$$\frac{8 - Z_2}{Z_2} = \frac{1}{N} \sum_{\vec{k}} \frac{D}{E_0(\vec{k})} \coth \frac{E_0(\vec{k})}{2\kappa T_c}, \tag{7.6a}$$

$$\frac{4(Z_2 - 2)}{3Z_2^2} = \frac{1}{N} \sum_{\vec{k}} \left(\frac{[J(\vec{k}) - 2J(0)]\, e^{E_0(\vec{k})/\kappa T_c}}{\kappa T_c (e^{E_0(\vec{k})/\kappa T_c} - 1)^2} - \frac{1}{2} \frac{J(\vec{k})}{E_0(\vec{k})} \coth \frac{E_0(\vec{k})}{2\kappa T_c} \right), \tag{7.6b}$$

where

$$E_0(\vec{k}) = [D^2 - Z_2 D J(\vec{k})]^{1/2}.$$

Figures 1 and 2 contain plots of $T_c(D)/T_c(0)$ as a function of the ratio D/J for a simple cubic (sc) and body centered cubic (bcc) lattice, respectively. We have assumed here that the exchange J is between nearest neighbors only, hence the variable $J(0) = zJ$, where z is the number of nearest neighbors. At very low anisotropy our results agree very closely with those of Lines. This is not too surprising since his decoupling technique is certainly the most sophisticated of all the decouplings applied to the anisotropy Green function, and therefore should be reasonably accurate for small values of D/J. For extremely large anisotropy, one may expand the functions in (7.6) to obtain $\kappa T_c = 2J(0)$ at $D/J = \infty$. Hence for large values of the anisotropy our RPA results approach the results of the MFT. This is in sharp contrast with the earlier decoupling schemes of (3.9), (3.10), and (3.11) which all predict $T_c \to \infty$ as $D/J \to \infty$.

Figure 3 contains plots of the sublattice magnetization $\bar{S}$ for a sc lattice as a function of T/T_c for three different values of D/J. Note that the larger D/J becomes the "squarer" the curve becomes. This is to be expected, since the larger D/J becomes the higher one must go in temperature, i.e., closer to T_c, before the intermediate spin states $|S-1\rangle$, $|S-2\rangle$, ..., $|-S+1\rangle$ become populated enough to reduce the ensemble average significantly. Also plotted in Fig. 3 is the MFT prediction of $\bar{S}$ vs T/T_c. This particular MFT

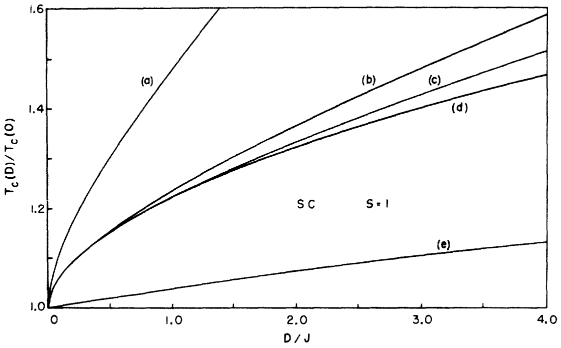

FIG. 1. The ratio $T_c(D)/T_c(0)$ vs D/J for a sc spin $S=1$ ferromagnet. (a) Narath's results; (b) Anderson and Callen's results; (c) Lines's results; (d) the results of this paper; (e) the MFT results.

curve could just as well represent the values $D/J=1.0$, 0.1, or 0.01, since the MFT does not distinguish between these values within the linewidth that we have chosen for our graph. Hence the Green-function calculation predicts greater changes in $\bar{S}$ vs T/T_c for different ratios of D/J than does the MFT. This is consistent with the fact that the MFT generally underestimates the effect of the crystal-field anisotropy on all of the statistical mechanical quantities.

In Fig. 4 we have plotted the second moment of S^z, $\langle (S_g^z)^2\rangle$ vs T/T_c for a sc lattice with $D/J=1.0$. For $T \leqslant T_c$ the Green-function curve lies beneath the MFT curve, but for $T \geqslant T_c$ the reverse is true. This particular behavior is typical for all values of D/J. The ensemble average $\langle (S_g^z)^2\rangle$ is greater than the isotropic value $\frac{1}{3}S(S+1)$ for all finite values of D/J. Only for $D/J=0$ will $\langle (S_g^z)^2\rangle = \frac{1}{3}S(S+1)$.

B. Spin $S = 1$ Antiferromagnet

For a spin $S=1$ antiferromagnet the equations of (5.5) can be written as

$$\begin{bmatrix} E-2\bar{S}[J(0)-J_{11}(\vec{k})] & -D & 2\bar{S}J_{21}(\vec{k}) & 0 \\ Z_2J_{11}(\vec{k})-D & E-2\bar{S}J(0) & Z_2J_{21}(\vec{k}) & 0 \\ -2\bar{S}J_{12}(\vec{k}) & 0 & E+2\bar{S}[J(0)-J_{11}(\vec{k})] & -D \\ Z_2J_{12}(\vec{k}) & 0 & Z_2J_{11}(\vec{k})-D & E+2\bar{S}J(0) \end{bmatrix} \begin{bmatrix} G_1^1(\vec{k},E) \\ G_1^2(\vec{k},E) \\ G_2^1(\vec{k},E) \\ G_2^2(\vec{k},E) \end{bmatrix} = \frac{1}{2\pi}\begin{bmatrix} 2\bar{S} \\ Z_2 \\ 0 \\ 0 \end{bmatrix} \qquad (7.7)$$

where we have used the notation $J(0)=J_{11}(0)-J_{21}(0) = J_{22}(0)-J_{12}(0)$. Clearly, there are four simple poles to be associated with each of the Green functions, but as we noted in Sec. V, they come in positive and negative pairs. Assuming $J_{12}(\vec{k})=J_{21}(\vec{k})$, the two positive poles are located at

$$E_1(\vec{k})=\{\alpha_1+\alpha_2+[4\alpha_1\alpha_2+(\alpha_3)^2]^{1/2}\}^{1/2}, \qquad (7.8a)$$

$$E_2(\vec{k})=\{\alpha_1+\alpha_2-[4\alpha_1\alpha_2+(\alpha_3)^2]^{1/2}\}^{1/2}, \qquad (7.8b)$$

where

$$\alpha_1=D^2-Z_2DJ_{11}(\vec{k}) + \bar{S}^2[J_{11}(\vec{k})-J_{12}(\vec{k})]\,[J_{11}(\vec{k})+J_{12}(\vec{k})], \qquad (7.9a)$$

$$\alpha_2=\bar{S}^2[J_{11}(\vec{k})-J_{12}(\vec{k})-2J(0)] \times [J_{11}(\vec{k})+J_{12}(\vec{k})-2J(0)], \qquad (7.9b)$$

$$\alpha_3=J_{12}(\vec{k})\,[Z_2D-4\bar{S}^2J(0)]. \qquad (7.9c)$$

Using the information contained in (7.5), the equations of (5.7) become

$$\frac{8-6\bar{S}-Z_2}{6}=\frac{2}{N}\sum_{\vec{k}}\left(\frac{R_1^1(\vec{k})}{e^{E_1(\vec{k})/\kappa T}-1}+\frac{T_1^1(\vec{k})}{e^{-E_1(\vec{k})/\kappa T}-1} + \frac{R_2^1(\vec{k})}{e^{E_2(\vec{k})/\kappa T}-1}+\frac{T_2^1(\vec{k})}{e^{-E_2(\vec{k})/\kappa T}-1}\right), \qquad (7.10a)$$

$$\frac{\bar{S}-Z_2}{2}=\frac{2}{N}\sum_{\vec{k}}\left(\frac{R_1^2(\vec{k})}{e^{E_1(\vec{k})/\kappa T}-1}+\frac{T_2^2(\vec{k})}{e^{-E_1(\vec{k})/\kappa T}-1} + \frac{R_2^2(\vec{k})}{e^{E_2(\vec{k})/\kappa T}-1}+\frac{T_2^2(\vec{k})}{e^{-E_2(\vec{k})/\kappa T}-1}\right), \qquad (7.10b)$$

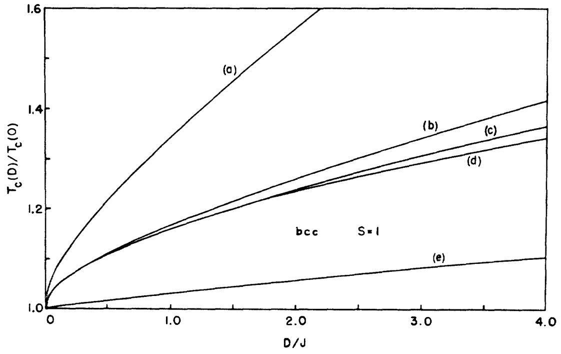

FIG. 2. The ratio $T_c(D)/T_c(0)$ vs D/J for a bcc spin $S=1$ ferromagnet. (a) Narath's results; (b) Anderson and Callen's results; (c) Lines's results; (d) the results of this paper; (e) the MFT results.

where the residues have the forms

$$R_1^1(\vec{k}) = [E_1^3(\vec{k})B_1 + E_1^2(\vec{k})B_2(\vec{k}) + E_1(\vec{k})B_3(\vec{k}) + B_4(\vec{k})] / \{2E_1(\vec{k})[E_1^2(\vec{k}) - E_2^2(\vec{k})]\}., \quad (7.11a)$$

$$R_1^2(\vec{k}) = [E_1^3(\vec{k})C_1 + E_1^2(\vec{k})C_2(\vec{k}) + E_1(\vec{k})C_3(\vec{k}) + C_4(\vec{k})] / \{2E_1(\vec{k})[E_1^2(\vec{k}) - E_2^2(\vec{k})]\} . \quad (7.11b)$$

The remaining residues have very similar forms: $R_2^1(\vec{k})$ is like $R_1^1(\vec{k})$ with $E_1(\vec{k})$ interchanged with $E_2(\vec{k})$, $T_1^1(\vec{k})$ is like $R_1^1(\vec{k})$ with $E_1(\vec{k})$ replaced by $-E_1(\vec{k})$, and $T_2^1(\vec{k})$ is like $T_1^1(\vec{k})$ with $E_1(\vec{k})$ interchanged with $E_2(\vec{k})$. These statements also hold true when the superscript is 2 rather than 1. The coefficients B_i and C_i have the forms

$$B_1 = 2\bar{S}, \quad B_2(\vec{k}) = Z_2D + 4\bar{S}^2[J(0) - J_{11}(\vec{k})], \quad B_3(\vec{k}) = 4\bar{S}Z_2DJ(0) - 16\bar{S}^3J(0)^2 - 2\bar{S}D^2, \quad (7.12a)$$

$$B_4(\vec{k}) = Z_2^2D^2J_{11}(\vec{k}) + 4\bar{S}Z_2DJ(0)[J(0) - 2J_{11}(\vec{k})] - 4\bar{S}^2J(0)D^2 - Z_2D^3 - 16\bar{S}^4J(0)^2[J(0) - J_{11}(\vec{k})],$$

$$C_1 = Z_2, \quad C_2(\vec{k}) = 2\bar{S}D - 2\bar{S}[J(0) - J_{11}(\vec{k})],$$

$$C_3(\vec{k}) = -Z_2D^2 + Z_2^2DJ_{11}(\vec{k}) - 4\bar{S}^2\{J(0)[J(0) - J_{11}(\vec{k})] + [2J(0) - J_{11}(\vec{k})][Z_2J(0) - D]\}, \quad (7.12b)$$

$$C_4(\vec{k}) = -2\bar{S}Z_2DJ_{11}(\vec{k}) - 2\bar{S}D^3 + 4\bar{S}Z_2D^2J_{11}(\vec{k}) - 2\bar{S}D[J(0) - J_{11}(\vec{k})][Z_2J_{11}(\vec{k})] + 8\bar{S}^3J(0)[J(0) - J_{11}(\vec{k})][Z_2J(0) - D] .$$

Equations (7.10) are the equations that determine $\bar{S}$ and Z_2 for all temperatures as a function of D and the J_{ij}.

Using the technique outlined in Sec. VI, we can obtain the following two equations which determine the Néel temperature T_c, and Z_2 evaluated at $T = T_c$:

$$\frac{8 - Z_2}{6} = \frac{2}{N}\sum_{\vec{k}}\left(\frac{Z_2D}{2E_0(\vec{k})}\coth\frac{E_0(\vec{k})}{2\kappa T_c} + \frac{Z_2D}{2E_{00}(\vec{k})}\coth\frac{E_{00}(\vec{k})}{2\kappa T_c}\right), \quad (7.13a)$$

$$\frac{8(Z_2 - 2)}{3Z_2^2} = \frac{2}{N}\sum_{\vec{k}}\left(F_1(\vec{k})\coth\frac{E_0(\vec{k})}{2\kappa T_c} + F_2(\vec{k})\coth\frac{E_{00}(\vec{k})}{2\kappa T_c}\right), \quad (7.13b)$$

where

$$F_1(\vec{k}) = \frac{[2J(0) - J_{11}(\vec{k})][D^2 - Z_2DJ_{11}(\vec{k})] + Z_2DJ_{12}(\vec{k})[J(0) - J_{11}(\vec{k})]}{Z_2DJ_{12}(\vec{k})E_0(\vec{k})}, \quad (7.14a)$$

$$F_2(\vec{k}) = -\frac{[2J(0) - J_{11}(\vec{k})][D^2 - Z_2DJ_{11}(\vec{k})] + Z_2DJ_{12}(\vec{k})[J(0) - J_{11}(\vec{k})]}{Z_2DJ_{12}(\vec{k})E_{00}(\vec{k})}, \quad (7.14b)$$

$$E_0(\vec{k}) = [D^2 - Z_2D(J_{11}(\vec{k}) - J_{12}(\vec{k}))]^{1/2}, \quad (7.14c)$$

$$E_{00}(\vec{k}) = [D^2 - Z_2D(J_{11}(\vec{k}) + J_{12}(\vec{k}))]^{1/2} . \quad (7.14d)$$

Numerical solutions have been obtained for (7.13) for a sc and bcc lattices, assuming only nearest-neighbor exchange. The numerical results obtained for the critical temperature of an antiferromagnet are equal to the results obtained for the critical temperature of a ferromagnet under the same conditions. The value of Z_2 at T_c is also the same in both cases under the same conditions. Figures 1 and 2 are therefore also appropriate for a spin $S = 1$ antiferromagnet.

Figure 5 shows $\bar{S}$ vs T/T_c for an antiferromagnet for the same three values of D/J as we used in Fig. 3. These curves are quite different from the ones for the ferromagnet, especially at low temperatures. This is because of the well-known zero-point fluctuations of an antiferromagnet. In an antiferromagnet the sublattice magnetization $\bar{S}$ does not reach its saturation value of unity for any finite value of D/J, whereas in a ferromagnet $\bar{S} = 1$ at $T = 0$ for all values of D/J. As the ratio of D/J is increased, the value of $\bar{S}$ at $T = 0$ will also increase and in the limit $D/J = \infty$, $\bar{S}$ will finally be completely saturated.

The average $\langle (S^z)^2 \rangle$ is plotted in Fig. 6 as a function of T/T_c and it possesses similar characteristics to the sublattice magnetization curve. For $T=0$ the average $\langle (S^z)^2\rangle$ does not approach its ferromagnetic value of unity, but some other value which is slightly less than unity. The larger D/J is, the larger the $T=0$ value of $\langle (S^z)^2\rangle$. The results for $T>T_c$ are the same for the average $\langle (S^z)^2\rangle$ in the antiferromagnetic case as for the ferromagnetic case which we have plotted in Fig. 4.

C. Spin $S=\frac{3}{2}$ Ferromagnet

For a spin $S=\frac{3}{2}$ ferromagnet, Eqs. (4.8) become

$$\begin{bmatrix} E-2\bar{S}[J(0)-J(\vec{k})] & -D & 0 \\ Z_2 J(\vec{k}) & E-2\bar{S}J(0) & -D \\ Z_3 J(\vec{k}) & -4D & E-2\bar{S}J(0) \end{bmatrix} \begin{bmatrix} G_1(\vec{k},E) \\ G_2(\vec{k},E) \\ G^3(\vec{k},E) \end{bmatrix} = \frac{1}{2\pi} \begin{bmatrix} 2\bar{S} \\ Z_2 \\ Z_3 \end{bmatrix} . \tag{7.15}$$

As before one can solve the secular equation of the above equation and obtain the three distinct poles of the Green functions. Then one can proceed to find the residues of these poles for each of the Green functions, and thereby obtain the specific form of Eqs. (4.11) appropriate for a spin $S=\frac{3}{2}$ ferromagnet. Then considering the limit of $T\to T_c$, by the technique described in Sec. VI, one can obtain the following three equations:

$$\kappa T_c = \left(4-\frac{2Z_2}{3}\right)\bigg/\left[\frac{1}{N}\sum_{\vec{k}} F(\vec{k})\right] , \tag{7.16a}$$

$$6=\frac{1}{N}\sum_{\vec{k}}\left(\frac{2Z_2D}{E_0(\vec{k})}\coth\frac{E_0(\vec{k})}{2\kappa T_c}+\kappa T_c H(\vec{k})\right), \tag{7.16b}$$

$$\begin{aligned}\frac{3-\beta_3}{8}=\frac{1}{N}\sum_{\vec{k}}\Bigg(&M(\vec{k})\coth\frac{E_0(\vec{k})}{\kappa T_c} \\ &-\frac{Z_2N(\vec{k})e^{E_0(\vec{k})/\kappa T_c}}{\kappa T_c E_0^2(\vec{k})\,(e^{E_0(\vec{k})/\kappa T_c}-1)^2} \\ &-[\kappa T_c/E_0^2(\vec{k})]\,[Z_2L(\vec{k})-4Z_2J(0)+\beta_3 D] \\ &+[4Z_2J(0)^2-2\beta_3DJ(0)]/L(\vec{k})\Bigg),\end{aligned} \tag{7.16c}$$

where

$$\beta_3 = Z_3/\bar{S} , \tag{7.17a}$$

$$E_0(\vec{k})=[4D^2-Z_2DJ(\vec{k})]^{1/2} , \tag{7.17b}$$

$$\begin{aligned}F(\vec{k})=4(8-\beta_3)D^2/\{&2[J(0)-J(\vec{k})]E_0^2(\vec{k}) \\ &-2Z_2DJ(\vec{k})^2+\beta_3D^2J(\vec{k})\} ,\end{aligned} \tag{7.17c}$$

$$H(\vec{k})=Z_2DJ(\vec{k})F(\vec{k})/E_0^2(\vec{k}) , \tag{7.17d}$$

$$M(\vec{k})=D[Z_2J(\vec{k})-8D]\,[Z_2J(\vec{k})-\tfrac{1}{2}\beta_3D]/[2E_0^3(\vec{k})] , \tag{7.17e}$$

$$\begin{aligned}N(\vec{k})=16D^2J(0)-&4Z_2DJ(0)J(\vec{k}) \\ &+2Z_2DJ(\vec{k})^2-\beta_3D^2J(\vec{k}) ,\end{aligned} \tag{7.17f}$$

$$L(\vec{k})=4(8-\beta_3)D^2/[F(\vec{k})E_0^2(\vec{k})] . \tag{7.17g}$$

We have solved the above equations numerically for a sc lattice. The results are plotted in Fig. 7. As with the spin $S=1$ case, our results closely approxi-

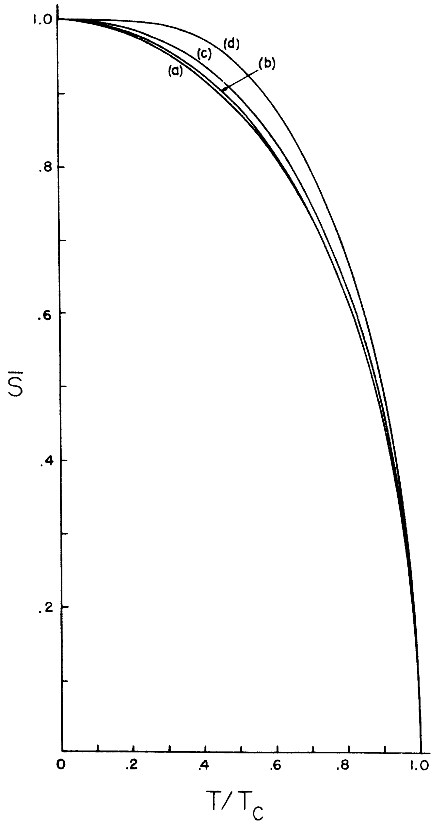

FIG. 3. The sublattice magnetization vs T/T_c for a sc spin $S=1$ ferromagnet. (a) $D/J=0.01$; (b) $D/J=0.1$; (c) $D/J=1.0$. Curve (d) is the MFT for the same three values of D/J, but in this case the three lines are indistinguishable.

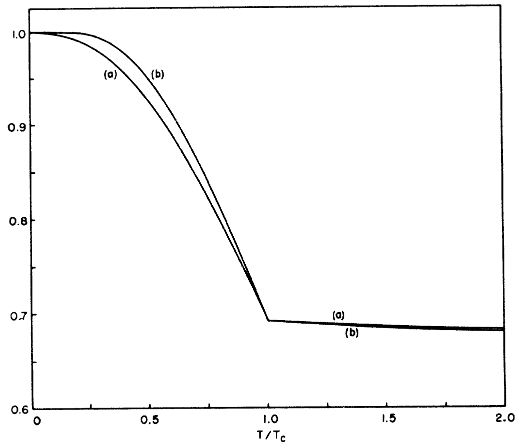

FIG. 4. The ensemble average $\langle (S^z)^2\rangle$ vs T/T_c for a sc spin $S=1$ ferromagnet. (a) Green-function calculation of this paper for $D/J=1.0$; (b) MFT prediction for $D/J=1.0$. Note that at $T=T_c$ the curves cross each other.

mate Lines's results for small anisotropy. For very large anisotropy the value of κT_c approaches the value predicted by the MFT which is $\frac{9}{2}J(0)$. The values of Z_2 and β_3 at T_c agree with the MFT values for each value of the ratio D/J, just as they did for the $S=1$ case.

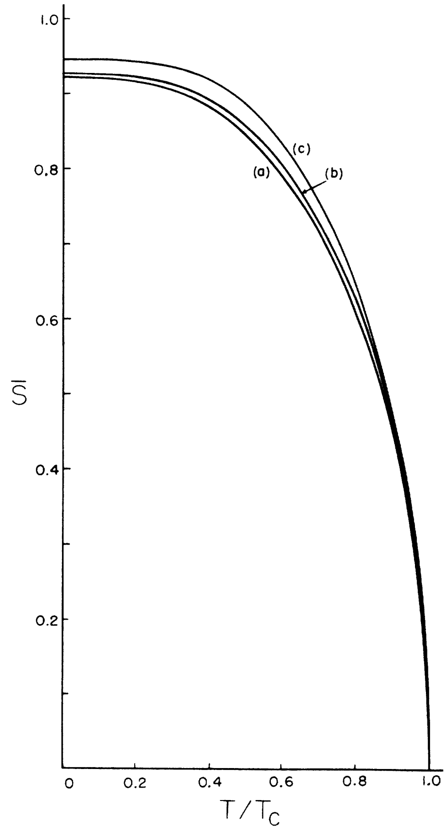

FIG. 5. The sublattice magnetization T/T_c for a sc spin $S=1$ antiferromagnet. (a) $D/J=0.01$; (b) $D/J=0.1$; (c) $D/J=1.0$.

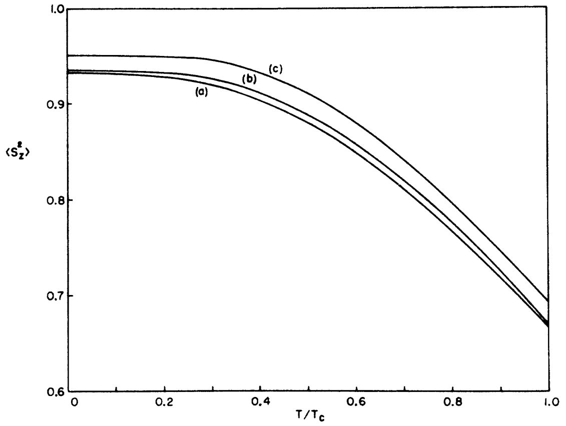

FIG. 6. The ensemble average $\langle (S^z)^2\rangle$ vs T/T_c for a sc spin $S=1$ antiferromagnet. (a) $D/J=0.01$; $D/J=0.1$; (c) $D/J=1.0$.

VIII. SUMMARY AND CONCLUSIONS

We have presented two significant ideas concerning the RPA decoupling of a magnetically ordered system which possesses crystal-field anisotropy.

First, we presented a convenient set of spin operators in (4.1) which form a closed set under commutation with the anisotropy term in the Hamiltonian. Therefore the anisotropy Green functions which are formed from these operators were also closed within each other by the anisotropy term. This formalism eliminated completely the need to decouple the anisotropy functions.

Second, we have found a decoupling scheme for the exchange Green functions which reproduces the usual RPA results when $D=0$.

Our technique is valid for all sizes of the spin of

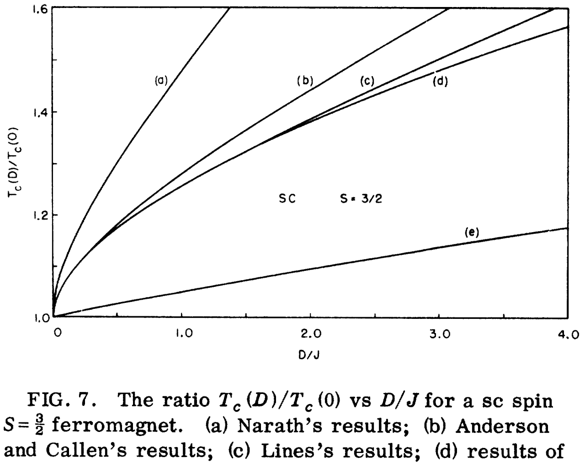

FIG. 7. The ratio $T_c(D)/T_c(0)$ vs D/J for a sc spin $S=\frac{3}{2}$ ferromagnet. (a) Narath's results; (b) Anderson and Callen's results; (c) Lines's results; (d) results of this paper; (e) MFT results.

the system. In addition, we have shown how the formalism is appropriate for antiferromagnetic as well as ferromagnetic systems.

The decoupling scheme we have presented here does remarkably well in comparison with the earlier theories on this subject. The transition temperature $T_c(D)$, as a function of the anisotropy parameter, is finite for all values of D. Our particular scheme shows that $T_c(D)$ depends more strongly on D for low anisotropy than it does in the MFT. For anisotropies of the order of the exchange J or smaller our results are equivalent to those of Lines. At the transition temperature the moments of $\langle S^z \rangle$ as predicted by our RPA calculation and as predicted by the MFT are the same, though the respective transition temperatures are quite different. The temperature dependence of the moments below T_c show a greater variation with changes in D/J than they do in the MFT. For very large anisotropies our results approach those of the MFT, and at $D/J=\infty$ both theories agree exactly.

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APPENDIX: COMMENTS ON MURAO AND MATSUBARA'S DECOUPLING APPROXIMATION

Murao and Matsubara attempted to solve the spin $S=1$ ferromagnet by a technique similar to ours. Their results, however, are unsatisfactory in a number of important respects. First, they predict a ratio of $T_c(D)/T_c(0)$ which is smaller than the MFT prediction for each corresponding value of D. But, as we have pointed out in an earlier part of this paper, the MFT underestimates the effect of the anisotropy, especially when D is small. Therefore Murao and Matsubara's results are an even further underestimation. Second, when D is set equal to zero in their calculation the result they obtain is not the usual RPA but the MFT. Both of these defects in their theory resulted from the particular Green functions they chose to decouple.

For the operators that appear on the left-hand side of the Green functions, they chose $A_g^1=S_g^+$ and $A_g^2=S_g^zS_g^+ + S_g^+S_g^z$, just as we had. In our problem we fixed the operator in on the right-hand side of the Green functions to be S_h^-. Murao and Matsubara chose to use the adjoints of both the operators A_h^1 and A_h^2, i.e., S_h^- and $S_h^-S_h^z+S_h^zS_h^-$, to be inserted in on the right-hand side of the Green functions. Hence, they obtained twice as many equations of motions as we did because they had not only our set of equations with S_h^- replaced by $S_h^zS_h^- + S_h^-S_h^z$, but also two of their four equations turned out to be equivalent, leaving three independent equations to be decoupled. They therefore overdetermined the problem, since only two moments were unknown. Their resolution of the overdeterminancy was to drop the one equation which they felt, for heuristic reasons, neglected the more important correlations. The remaining two equations produced the inadequacies mentioned above.

This error of Murao and Matsubara points out an important point about decoupling schemes that is worth emphasizing. Suppose operators A and B are relatively uncorrelated, and that only A has a finite ensemble average. We might try to decouple a Green function which contains A and B such that

$$\langle\langle A\,B;C\rangle\rangle=\langle A\rangle\ \langle\langle B;C\rangle\rangle\ , \tag{A1}$$

where C is some other operator. The question now is whether this is automatically a valid thing to do for all possible choices for the operator C. The point we wish to make here is that (A1) may be valid for some choices for C, and quite bad for some other choices. Since Murao and Matsubara performed approximations of the form of (A1) for two different choices for C, i.e., S_h^- and $S_h^zS_h^- + S_h^-S_h^z$, they had no guarantee that both would be equally valid approximations. Evidently, their choices were mutually conflicting since they did obtain thermodynamic inconsistencies.

The particular decoupling made by Lines, which is requoted in (3.11), emphasizes the point we are making here: The decoupling scheme may depend strongly on the operator in the right-hand side of the Green function.

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