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Green's-Function Theory of an Antiferromagnet

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A Green's-function theory is developed for a Heisenberg antiferromagnet using a decoupling scheme which conserves the frequency moments of the spectral function. The method closely follows Tahir-Kheli's decoupling scheme for a ferromagnetic system, and various correlation functions, sublattice magnetization, and susceptibilities are obtained. The theory is compared with the low- and high-temperature series expansions. The Néel temperature obtained using the theory is used to estimate $|J/k_B|$ values of some transition-metal compounds. The theory is then applied to one and two dimensions, and it is found to lend support to Stanley and Kaplan's suggestion of a second-order phase transition, while not predicting spontaneous magnetic ordering.

I. INTRODUCTION

The Heisenberg model of the spin-spin interaction to describe various magnetic properties of materials continues to be of interest even today. The model consists of a finite array (a linear chain in one dimension, a square in two dimensions, and a cubic structure in three dimensions) of points which may be thought of as vertices of unit spacing and side L . Periodic boundary conditions are assumed. The system of spins interact through an isotropic exchange between the nearest-neighbor sites. The Hamiltonian for such a system is given by

$$\mathcal{H} = J \sum'_{i,j} \vec{S}_i \cdot \vec{S}_j - \mu_B H \sum_i S_i^z, \quad (1.1)$$

where the prime on the summation indicates that the sum is only restricted to nearest-neighbor pairs. In spite of the inherent simplicity of the model, it remains essentially a many-body problem and an exact solution for quantities such as the partition function have not been found in any dimension. The ferromagnetic ground state and the low-lying excited states of the system were found by Bloch,¹ who also showed that the deviation from

the ground-state magnetization was proportional to $T^{3/2}$. Dyson² in 1956 presented a theory of spin-wave interactions, which is useful in discussing the low-temperature thermodynamics of the ferromagnetic system ($J < 0$).

The development on the antiferromagnetic Heisenberg model, however, has not been so encouraging. The exact antiferromagnetic ground state of the Hamiltonian (1.1) (with $J > 0$) is not known in any dimension even though Bethe³ and Hulthén⁴ have computed the ground-state energy of the antiferromagnetic linear chain. des Cloizeaux and Pearson,⁵ discussed the low-lying excitation spectrum of the antiferromagnetic chain and found that the long-wavelength magnons obey a linear dispersion law. The statistical theories of the model historically date back to the molecular-field theory of Néel,⁶ which successfully explained the existence of a transition temperature, a specific-heat anomaly, etc., but failed to explain the lack of short-range order above the transition temperature and many other low-temperature properties. Series expansions by Opechowski⁷ at high temperatures and spin-wave theories at low temperatures have their validities in restricted range of temperatures but are unreliable near the transition

point.

The most satisfactory approach to describe the statistical mechanics of the Heisenberg antiferromagnet has been the application of the method of double-time Green's functions.⁸ The most satisfying feature of the Green's-function formalism is its validity in both high- and low-temperature ranges, as well as near the critical temperature. The various works using Green's-function theories differ from one another in one important aspect. The exact equation of motion for Green's functions of any order involves a still higher-order Green's function and this new Green's function has to be approximated in some manner. The random-phase approximation (RPA) is unsatisfactory, and does not agree with the spin-wave theories at low temperature. Anderson and Callen⁹ proposed a decoupling scheme for an antiferromagnetic system, extending Callen's¹⁰ earlier suggestion for a ferromagnet. Lee and Liu¹¹ studied the statistical mechanics of a simple antiferromagnet over the entire range of temperature and obtained sublattice magnetization, Néel temperature, and the various correlation functions.

While the work of Lee and Liu was in excellent agreement with various other theoretical estimates, the scheme of decoupling was made from heuristic arguments. An attempt to fix the parameters of the theory from physical considerations was made by Mubayi and Lange¹² for the ferromagnetic case. Tahir-Kheli¹³ used a self-consistent version of an algorithm due to Roth¹⁴ for the solution of the Hubbard model, and found that, in the case of $S = \frac{1}{2}$, his results for the ferromagnetic Heisenberg model basically reproduced Mubayi and Lange's solution. The decoupling scheme in this new approach is fixed by the condition for conservation of various frequency moments of the spectral function.¹⁵ The scheme is made self-consistent by the additional condition that the scheme also fixes the corresponding decoupling of the correlation functions. Tahir-Kheli found that this latter condition cannot be satisfied for an anisotropic Heisenberg model.

The present work treats the statistical mechanics of an antiferromagnet with isotropic nearest-neighbor interactions by the use of Green's-function theory, using the self-consistent moment-conserving decoupling scheme (SCMD) of Tahir-Kheli. In Sec. II, we review the definitions and properties of spin Green's functions and the properties of various frequency moments of the spectral functions. We also derive the equation of motion for the Green's function, and briefly outline the various decoupling schemes proposed earlier. In Sec. III, we describe the SCMD in detail, and obtain a solution for the Green's-function equation of motion and consequently obtain the

energy spectrum. In Sec. IV, we obtain the low-temperature expansion for the magnetization and compare it with the spin-wave theory and the RPA. We also calculate the susceptibilities in the antiferromagnetic phase. In Sec. V, we determine the Néel temperature of the solid, and compare the $|J|/k_B$ value predicted by the present approach with other high-temperature estimates on various substances. We calculate the molar susceptibility at the Néel temperature and explain experimental data on several compounds. In the last sections we specialize our results in one and two dimensions and discuss these in the light of an exact theorem due to Mermin and Wagner.¹⁶

II. GREEN'S-FUNCTION EQUATIONS

For convenient reference, we give here various definitions and formulas, which we shall be using later in this paper.⁸ The double-time retarded r and advanced a Green's functions are defined by

$$\langle\langle A(t); B(t') \rangle\rangle^r \equiv G_{AB}^r(t-t') = -i\theta(t-t')\langle[A(t), B(t')]\rangle, \quad (2.1)$$

$$\langle\langle A(t); B(t') \rangle\rangle^a \equiv G_{AB}^a(t-t') = i\theta(t-t')\langle[A(t), B(t')]\rangle;$$

where the angular brackets $\langle \rangle$ denote the usual average over a grand canonical ensemble. $\theta(t)$ is the unit step function, being unity only when t is positive and is zero otherwise. The Green's functions are not defined for $t=t'$. The Fourier transform of the Green's function defined by

$$\langle\langle A; B \rangle\rangle_E^{r,a} \equiv G_{AB}^{r,a}(E) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} G_{AB}^{r,a}(t) e^{iEt} dt, \quad (2.2)$$

satisfies the following equation of motion:

$$E G_{AB}^{r,a}(E) = \frac{1}{2\pi} \langle[A, B]\rangle + \langle\langle[A, \mathcal{H}], B \rangle\rangle_E^{r,a}. \quad (2.3)$$

It is well known that these Fourier transforms of the retarded (advanced) Green's functions can be analytically continued to the upper- (lower-) half of the complex energy plane if E is to be considered complex. If a cut is made along the real axis, the function

$$G(E) = G^r(E) \quad \text{Im}E > 0 \\ = G^a(E), \quad \text{Im}E < 0$$

can be considered to be one analytic function consisting of two branches, one defined in the upper and the other in the lower half-plane.

Regarding Z as the complex energy variable we have

$$\langle\langle A; B \rangle\rangle_Z \equiv G_{AB}(Z) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \frac{F^{AB}(\omega)}{Z-\omega} d\omega, \quad \text{Im}Z \neq 0 \quad (2.4)$$

where $F^{AB}(\omega)$ is the Fourier transform of the time

$$\langle [A(t), B(t')] \rangle = \int_{-\infty}^{+\infty} d\omega e^{-i\omega(t-t')} F^{AB}(\omega) . \quad (2.5)$$

From Eq. (2.4) it follows, using the properties of δ functions, that

$$F^{AB}(\omega) = i \lim_{\epsilon \rightarrow 0^+} [G_{AB}(\omega + i\epsilon) - G_{AB}(\omega - i\epsilon)] . \quad (2.6)$$

We define the n th moment of the spectral function F^{AB} by the following relation:

$$\omega_{AB}^n = \int_{-\infty}^{+\infty} \omega^{n-1} F(\omega) d\omega . \quad (2.7)$$

[The functions $F^{AB}(\omega)$ differ from the conventional spectral functions $J_{AB}(\omega)$ of Zubarev by a factor $(e^{\beta\omega} + 1)$.] These moments are related to the equal-time thermodynamic average

$$\omega_{AB}^n = \lim_{t \rightarrow t'} \left\langle \left[\left(i \frac{\partial}{\partial t} \right)^{n-1} A(t), \left(-i \frac{\partial}{\partial t'} \right)^n B(t') \right] \right\rangle . \quad (2.8)$$

Thus, a knowledge of these moments can be expected to provide us with a knowledge of spectral functions and hence of the various properties of interest. The equation of motion satisfied by this generalized Green's function $\langle\langle A, B \rangle\rangle_Z$ is seen to be

$$Z \langle\langle A; B \rangle\rangle_Z = \frac{1}{2\pi} \omega_{AB}^1 + \langle\langle [A, \mathcal{H}]; B \rangle\rangle_Z . \quad (2.9)$$

In general, when one is dealing with a many-body system of interacting particles, Eq. (2.9) is an infinite hierarchy of equations and hence no exact solution can be found for $\langle\langle A; B \rangle\rangle_Z$. It is customary to truncate the hierarchy at some suitable stage, and one of the central problems in using a Green's-function procedure is to find a reasonable truncation, in conformity with the physical constraints.

The Hamiltonian of the system we consider is

$$\mathcal{H} = J \sum_{i \in A} \sum_{j \in B} \vec{S}_i \cdot \vec{S}_j - \mu_B H \sum_i S_i^z . \quad (2.10)$$

The sum over j in the first sum is restricted to the nearest neighbors and the second term represents the extra energy in a magnetic field directed along the negative z axis. The equation of motion for the Green's function $\langle\langle S_{\lambda i}^+; S_{\mu j}^- \rangle\rangle_Z = G_{ij}^{\lambda\mu}(Z)$ is given by

$$\begin{aligned} (Z - \mu_B H) \langle\langle S_{\lambda i}^+; S_{\mu j}^- \rangle\rangle &= \frac{\langle S_{\lambda i}^z \rangle}{\pi} \delta_{ij} \delta_{\lambda\mu} \\ &- J \sum_{\delta} \langle\langle (S_{\lambda i}^+ S_{\nu i+\delta}^z - S_{\nu i+\delta}^+ S_{\lambda i}^z); S_{\mu j}^- \rangle\rangle \\ &\quad (\lambda \neq \nu) . \end{aligned} \quad (2.11)$$

Here δ 's are the nearest-neighbor vectors, the Greek indices λ, μ, ν refer to either of the A or B sublattices. The second term on the right-hand side contains the higher-order Green's functions. The frequently used RPA decoupling scheme consists of neglecting fluctuations in S_{ν}^z [$g \equiv (\lambda, i)$], replacing the operator by its average value $\langle S_{\nu}^z \rangle$,

i. e.,

$$\langle\langle S_{\lambda i}^+ S_{\nu p}^z; S_{\mu j}^- \rangle\rangle_Z \approx \langle S_{\nu}^z \rangle \langle\langle S_{\lambda i}^+; S_{\mu j}^- \rangle\rangle . \quad (2.12)$$

This procedure neglects short-range correlations in the system and leads to a magnetization renormalization of the quasiparticle energy in disagreement with the low-temperature theory. Callen¹⁰ suggested a decoupling procedure using a parameter which was later used by Anderson and Callen⁹ and by Lee and Liu¹¹ for the antiferromagnetic problem. The parametrization is suggested by the fact that for spin one-half S_{ν}^z can be written $S - S_{\nu}^+ S_{\nu}^-$, and Callen proposed

$$\langle\langle S_{\nu}^z S_{\nu}^+; B \rangle\rangle \approx \langle S_{\nu}^z \rangle \langle\langle S_{\nu}^+; B \rangle\rangle - \alpha \langle S_{\nu}^+ S_{\nu}^- \rangle \langle\langle S_{\nu}^+; B \rangle\rangle . \quad (2.13)$$

When applied to the antiferromagnetic problem, Anderson and Callen⁹ found that one of the two equally acceptable values for this parameter led to internal inconsistency, and in that sense the procedure remained largely *ad hoc*. Liu and Lee¹¹ observed that the inconsistency was absent in the case of vanishing magnetic field. Mubayi and Lange,¹² in their study of two-dimensional ferromagnet introduced an over-all parameter on the right-hand side of Eq. (2.13) in addition to α and they determined these two parameters by two constraints: The first one, often referred to as Dyson's kinematical condition, requires $\langle S_i^z \rangle^{2s+1} = 0$, and the second condition was the assumed validity of the decoupling procedure in a certain limiting case. By doing so they obtained a phase transition in two dimensions, in accordance with the theory of Stanley and Kaplan,¹⁷ while still not obtaining a spontaneous magnetization, the non-existence of which has been rigorously established by Mermin and Wagner¹⁶ in both one and two dimensions. However, Kenan¹⁸ reported that this new scheme leads to the onset of antiferromagnetic short-range order in a ferromagnetic system.

In the self-consistent moment-conserving decoupling scheme (SCMD) of Tahir-Kheli,¹³ which we use here with the necessary modification for an antiferromagnetic system, the parameters of the decoupling scheme are chosen so as to conserve the maximum number of frequency moments $\omega_{AB}^{(n)}$. Since the parameters are assumed to be independent of Z , these moments can be computed and compared for large Z . The procedure becomes clearer in Sec. III, when we solve Eq. (2.11) by explicitly decoupling the higher-order Green's functions.

III. DECOUPLING PROCEDURE AND SPIN-WAVE SPECTRUM

A. Decoupling Procedure

In this section we shall seek an approximate solution to the Eq. (2.11) by decoupling the last term on its right-hand side. Let us propose a de-

coupling scheme following Tahir-Kheli.¹³ For $\lambda \neq \nu$,

$$\langle\langle (S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} - S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma}) ; S_{\mu j}^{\sigma} \rangle\rangle_Z = A_1^{\lambda\nu} \langle\langle S_{\lambda i}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle + A_2^{\lambda\nu} \langle\langle S_{\nu i+\delta}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle. \quad (3.1)$$

The parameters $A_1^{\lambda\nu}$ and $A_2^{\lambda\nu}$ will be fixed by the condition of conservation of the first frequency moment, and the necessity of the procedure being self-consistent. If $F(\omega)$ denotes the first frequency moment of the Green's function on the left, and $f_1(\omega)$ and $f_2(\omega)$ denote the corresponding moments for the two Green's functions $\langle\langle S_{\lambda i}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle_Z$ and $\langle\langle S_{\nu i+\delta}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle_Z$, then we have, for large Z

$$\begin{aligned} \langle\langle (S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} - S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma}) ; S_{\mu j}^{\sigma} \rangle\rangle_Z &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} d\omega \frac{F(\omega)}{Z - \omega} \\ &= \frac{1}{2\pi} \int_{-\infty}^{+\infty} F(\omega) \left[1 + \left(\frac{\omega}{Z}\right) + \left(\frac{\omega}{Z}\right)^2 + \dots \right] d\omega. \end{aligned} \quad (3.2)$$

The successive terms can be recognized as the first, second, ..., etc., moments of the spectral function $F(\omega)$. On the right-hand side of (3.1) we have

$$\begin{aligned} A_1^{\lambda\nu} \langle\langle S_{\lambda i}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle_Z + A_2^{\lambda\nu} \langle\langle S_{\nu i+\delta}^{\sigma} ; S_{\mu j}^{\sigma} \rangle\rangle_Z \\ = \frac{1}{2\pi Z} \int_{-\infty}^{+\infty} [A_1^{\lambda\nu} f_1(\omega) + A_2^{\lambda\nu} f_2(\omega)] \\ \times \left[1 + \left(\frac{\omega}{Z}\right) + \left(\frac{\omega}{Z}\right)^2 + \dots \right] d\omega. \end{aligned} \quad (3.3)$$

If the decoupling scheme is to preserve the n th moment the coefficients of Z^{-n} on the right-hand side of (3.2) and (3.3) must match. Since we are decoupling the equations for (2.11) at the very first stage, we can only preserve the first moment. More moments could be conserved by writing down the equation of motion for the Green's functions on the right-hand side of Eq. (2.11) and decoupling at a later stage. We thus require

$$\int_{-\infty}^{+\infty} F(\omega) d\omega = \int_{-\infty}^{+\infty} [A_1^{\lambda\nu} f_1(\omega) + A_2^{\lambda\nu} f_2(\omega)] d\omega. \quad (3.4)$$

Using Eq. (2.5) in the limit $t \rightarrow t'$ this leads to

$$\begin{aligned} \langle\langle (S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma}) + (S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma}) \rangle\rangle (\delta_{\lambda\mu} \delta_{i+\delta, j} - \delta_{\lambda\mu} \delta_{ij}) \\ = 2A_1^{\lambda\nu} \delta_{\lambda\mu} \delta_{ij} \langle S_{\lambda i}^{\sigma} \rangle + 2A_2^{\lambda\nu} \delta_{\nu\mu} \delta_{i+\delta, j} \langle S_{\nu i+\delta}^{\sigma} \rangle. \end{aligned} \quad (3.5)$$

Since $\lambda \neq \nu$, Eq. (3.5) determines both $A_1^{\lambda\nu}$ and $A_2^{\lambda\nu}$. Denoting

$$\varphi_{\lambda\nu}^{\delta} = \langle S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} \rangle, \quad (3.6)$$

$$\psi_{\lambda\nu}^{\delta} = \langle S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} \rangle,$$

we have, for $A_1^{\lambda\nu}$ and $A_2^{\lambda\nu}$,

$$A_1^{\lambda\nu} = -\frac{1}{2\sigma_{\lambda}} (2\varphi_{\lambda\nu}^{\delta} + \psi_{\lambda\nu}^{\delta}),$$

$$A_2^{\lambda\nu} = \frac{1}{2\sigma_{\nu}} (2\varphi_{\lambda\nu}^{\delta} + \psi_{\lambda\nu}^{\delta}), \quad (3.7)$$

where $\sigma_{\lambda} \equiv \langle S_{\lambda i}^{\sigma} \rangle$ has been assumed independent of the lattice site because of translational invariance within each sublattice.

The first-order moment-conserving decoupling scheme (3.1) will be self-consistent as well, if the correlation functions corresponding to the Green's functions are self-consistent too. This means that for $A_1^{\lambda\nu}$ and $A_2^{\lambda\nu}$ given by Eq. (3.7), we must have

$$\begin{aligned} \langle S_{\mu j}^{\sigma} (S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} - S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma}) \rangle &= A_1^{\lambda\nu} \langle S_{\mu j}^{\sigma} S_{\lambda i}^{\sigma} \rangle \\ &+ A_2^{\lambda\nu} \langle S_{\mu j}^{\sigma} S_{\nu i+\delta}^{\sigma} \rangle. \end{aligned} \quad (3.8)$$

This result is an immediate consequence of the expression for the correlation functions in terms of the Green's functions

$$\langle BA \rangle = i \lim_{\epsilon \rightarrow 0^+} \int_{-\infty}^{+\infty} \frac{\langle\langle A ; B \rangle\rangle_{E+i\epsilon} - \langle\langle A ; B \rangle\rangle_{E-i\epsilon}}{e^{\beta E} - 1} dE. \quad (3.8a)$$

Since Eq. (3.8) is to hold for all (μ, j) it must, in particular be true when $(i=j, \mu=\lambda)$, and also in the case when $(j=i+\delta, \lambda=\mu)$. Consider $S = \frac{1}{2}$ to begin with. In this case for the first choice of (μ, j) one obtains

$$\varphi_{\lambda\nu}^{\delta} = \frac{2\sigma_{\lambda}(\psi_{\lambda\nu}^{\delta} + \sigma_{\nu}^2) - \sigma_{\nu}\psi_{\lambda\nu}}{2(\sigma_{\nu} - 2\sigma_{\lambda}\psi_{\lambda\nu})}, \quad (3.9a)$$

while, if one makes the second choice for (μ, j) one gets

$$\varphi_{\lambda\nu}^{\delta} = \frac{2\sigma_{\nu}(\psi_{\lambda\nu}^{\delta} + \sigma_{\lambda}^2) - \sigma_{\lambda}\psi_{\lambda\nu}}{2(\sigma_{\lambda} - 2\sigma_{\nu}\psi_{\lambda\nu})}. \quad (3.9b)$$

The expressions (3.9a) and (3.9b) are indeed self-consistent because of obvious symmetry $\sigma_{\lambda} = -\sigma_{\nu}$, for an antiferromagnet. Thus, Eq. (3.1) together with Eq. (3.7) fixes the decoupling scheme in this case. From Eq. (3.7), and the symmetry requirement $\sigma_A = -\sigma_B$, we have

$$A_1^{\lambda\nu} = \sigma_{\lambda} (1 + 2\psi_{\lambda\nu}^{\delta}) = A_2^{\lambda\nu} = -A_1^{\nu\lambda} = -A_2^{\nu\lambda}. \quad (3.10)$$

Coming back to the case of general spin S , it is seen that $\varphi_{\lambda\nu}^{\delta}$ in this case is

$$\begin{aligned} \varphi_{\lambda\nu}^{\delta} &= \{ \sigma_{\nu}\sigma_{\lambda} [\psi'_{\lambda\nu} + \psi_{\lambda\nu}^{\delta} + L'_{\lambda\nu} - S(S+1)\sigma_{\nu}] \\ &+ \frac{1}{2}\sigma_{\nu}\psi_{\lambda\nu}^{\delta} [S(S+1) - M_{\lambda} - \sigma_{\lambda}] - \psi_{\lambda\nu}^{\delta}\sigma_{\lambda} \} \\ &\times [\psi_{\lambda\nu}\sigma_{\lambda} + M_{\lambda}\sigma_{\nu} - S(S+1)\sigma_{\nu}]^{-1}, \end{aligned} \quad (3.11)$$

where

$$\begin{aligned} \psi'_{\lambda\nu} &= \langle S_{\lambda i}^{\sigma} S_{\lambda i}^{\sigma} S_{\nu i+\delta}^{\sigma} \rangle, \quad L'_{\lambda\nu} = \langle (S_{\lambda i}^{\sigma})^2 S_{\nu i+\delta}^{\sigma} \rangle, \\ M_{\lambda} &= \langle (S_{\lambda i}^{\sigma})^2 \rangle. \end{aligned} \quad (3.12)$$

The above result is trivially derived from Eq. (3.8) by choosing $(\mu, j) = (\lambda, i)$, and using the fol-

lowing relation for the general spin:

$$S_i^+ S_i^- = S(S+1) - (S_i^z)^2 - S_i^z. \quad (3.13)$$

Thus the additional complexity of the general spin case is due to the presence of new quantities like ψ'_λ , L'_λ , and M'_λ . Before we deal with this complexity, we will find it convenient to write down the formal solution for the Green's functions, as a result of our decoupling scheme.

B. Formal Solution of Eq. [2.11] and Spin-Wave Spectrum

Let us fix μ to be the A sublattice throughout the calculation. Then on introducing the various decoupling schemes, discussed in the foregoing subsection, into Eq. (2.11) we get a set of two simultaneous equations:

$$\begin{aligned} (Z - \mu_B H - J \partial A_1^{AB}) \langle \langle S_{A_i}^+; S_{A_j}^- \rangle \rangle_Z \\ = \frac{\sigma_A}{\pi} \delta_{ij} + J A_1^{AB} \sum_0 \langle \langle S_{B_{i+\delta}}^+; S_{A_j}^- \rangle \rangle_Z \end{aligned}$$

and

$$\begin{aligned} (Z - \mu_B H + J \partial A_1^{AB}) \langle \langle S_{B_i}^+; S_{A_j}^- \rangle \rangle_Z \\ = -J A_1^{AB} \sum_0 \langle \langle S_{A_{i+\delta}}^+; S_{A_j}^- \rangle \rangle_Z, \end{aligned}$$

where ∂ is the nearest-neighbor coordination number of the lattice under consideration. In deriving the last equation, we have used the fact that $A_1^{BA} = -A_1^{AB}$. Henceforth we shall drop the superscripts. To diagonalize Eqs. (3.11) and (3.12) we introduce the Fourier transforms

$$\langle \langle S_{\lambda i}^+; S_{\lambda j}^- \rangle \rangle_Z = \left(\frac{2}{N} \right)^2 \sum_{k_1 k_2} G_Z^{\lambda A}(\vec{k}_1, \vec{k}_2) e^{i\vec{k}_1 \cdot \vec{R}_i - i\vec{k}_2 \cdot \vec{R}_j}.$$

This gives

$$\begin{aligned} G_Z^{AA}(\vec{k}_1, \vec{k}_2) = \frac{\sigma_A}{\pi} \delta_{\vec{k}_1 \vec{k}_2} \delta_{\vec{k}_1 \vec{k}_2} \\ \times \left(\frac{Z - \alpha_2}{(Z - \alpha_1)(Z - \alpha_2) + (J A_1 \partial \gamma_k)^2} \right), \quad (3.14a) \end{aligned}$$

$$G_Z^{BA}(\vec{k}) = -\frac{J A_1 \partial \gamma_k}{Z - \alpha_2} G_Z^{AA}(\vec{k}); \quad (3.14b)$$

where

$$\alpha_{1,2} = \mu_B H \pm J \partial A_1 \quad (3.15a)$$

and

$$\gamma_k = \frac{1}{\partial} \sum_0 e^{i\vec{k} \cdot \delta}. \quad (3.15b)$$

The nature of the spin-wave spectrum becomes apparent on rewriting Eq. (3.14a) in a slightly different form,

$$\begin{aligned} G_Z^{AA}(\vec{k}) = \frac{\sigma_A}{\pi} \frac{Z - \alpha_2}{2E_{\vec{k}}} \\ \times \left(\frac{1}{Z - E_{\vec{k}} - \mu_B H} - \frac{1}{Z + E_{\vec{k}} - \mu_B H} \right), \quad (3.16) \end{aligned}$$

and, using Eq. (3.14b),

$$\begin{aligned} G_Z^{BA}(\vec{k}) = -\frac{\sigma_A}{\pi} \frac{J \partial A_1 \gamma_k}{2E_{\vec{k}}} \\ \times \left(\frac{1}{Z - E_{\vec{k}} - \mu_B H} - \frac{1}{Z + E_{\vec{k}} - \mu_B H} \right), \quad (3.17) \end{aligned}$$

where

$$E_{\vec{k}} = J \partial A_1 (1 - \gamma_k^2)^{1/2}. \quad (3.18)$$

By obvious symmetry, the Green's functions G^{BB} and G^{AB} are obtained, respectively, from G^{AA} and G^{BA} by $\alpha_1 \leftrightarrow \alpha_2$, $\gamma_{\vec{k}} \leftrightarrow -\gamma_{\vec{k}}$, and $\sigma_A \leftrightarrow \sigma_B$. The energies of the spin wave in the upper and lower branches are, as can be corroborated by comparison with the conventional spin-wave theory

$$E_{u,i} = E_k \pm \mu_B H. \quad (3.19)$$

The above results now immediately lead to the following:

$$\langle S_{A_i}^- S_{A_i}^+ \rangle = \sigma_A [2\Omega(0) + \varphi_u(0) - \varphi_i(0) - 1], \quad (3.20)$$

$$\langle S_{B_i}^- S_{B_i}^+ \rangle = \sigma_B [\varphi_u(0) - \varphi_i(0) - 2\Omega(0) - 1], \quad (3.21)$$

where

$$\varphi_{u,i}(\vec{R}) = \frac{2}{N} \sum_k e^{i\vec{k} \cdot \vec{R}} f_{u,i}(\vec{k}), \quad (3.22)$$

$$\Omega(\vec{R}) = \frac{2}{N} \sum_k \frac{f_u(\vec{k}) + f_i(\vec{k}) + 1}{2(1 - \gamma_k^2)^{1/2}} e^{i\vec{k} \cdot \vec{R}}. \quad (3.23)$$

The general structure of the Green's functions and the correlation functions are thus seen to be the same as those obtained by Anderson-Callen or RPA decoupling. It was shown by Callen and Shtrikman,¹⁹ and later generalized for the case of multiple sublattices by Mills *et al.*²⁰ that an implication of such a decoupling scheme, or alternatively of Eqs. (3.20)–(3.23) is that the generating function for $\langle \langle S_\lambda^{\pm n} \rangle \rangle$ is given by

$$\begin{aligned} Q_\lambda(a) = \langle e^{aS_\lambda^+} \rangle \\ = [\varphi_\lambda^{2S+1} e^{-aS} - (1 + \varphi_\lambda)^{2S+1} e^{(S+1)a}] \\ \times [\varphi_\lambda^{2S+1} - (1 + \varphi_\lambda)^{2S+1}]^{-1} [(1 + \varphi_\lambda) e^a - \varphi_\lambda]^{-1}, \quad (3.24) \end{aligned}$$

where $\varphi_\lambda = \langle S_\lambda^- S_\lambda^+ \rangle / 2\sigma_\lambda$. The averages $\langle \langle S_\lambda^{\pm n} \rangle \rangle$ are obtained from (3.24) by

$$\langle \langle S_\lambda^{\pm n} \rangle \rangle = \left(\frac{d^n}{da^n} \right) Q_\lambda(a) \Big|_{a=0}. \quad (3.25)$$

In particular, the sublattice magnetization σ_A is given by

$$\begin{aligned} \sigma_A = S - \frac{1}{2} [2\Omega(0) + \varphi_u(0) - \varphi_i(0) - 1] \\ + (2S+1) \left[\frac{1}{2} (2\Omega(0) + \varphi_u(0) - \varphi_i(0) - 1) \right]^{2S+1} \\ \times \left\{ \frac{1}{2} [2\Omega(0) + \varphi_u(0) - \varphi_i(0) + 1] \right\}^{2S+1} \\ - \left[\frac{1}{2} [2\Omega(0) + \varphi_u(0) - \varphi_i(0) - 1] \right]^{2S+1} \Big|^{-1}. \quad (3.26) \end{aligned}$$

σ_B is trivially obtained from σ_A with the substitution $-\Omega(0)$ for $\Omega(0)$. Similarly

$$M_\lambda = \langle (S_\lambda^z)^2 \rangle = S^2 - (1 - 2S)\varphi_\lambda + 2\varphi_\lambda^2 - (1 + 2S)(1 + 2\varphi_\lambda) \\ \times \varphi_\lambda^{2S+1} / [(1 + \varphi_\lambda)^{2S+1} - \varphi_\lambda^{2S+1}] , \quad (3.27)$$

and in a manner analogous to the Tahir-Kheli method we can get

$$\psi'_\lambda = \langle S_{\lambda i}^z S_{\lambda i}^- S_{\nu i+s}^+ \rangle = (1/2\sigma_\nu) [3M_\nu - S(S+1) - \sigma_\nu] \psi_{\lambda\nu}^0 , \quad (3.28)$$

Following the method outlined by Tahir-Kheli and Callen²¹ we can obtain the function L'_λ as well, through the generating function

$$\langle S_{\lambda\nu}^z e^{aS_{\lambda\nu}^z} S_{\nu\mu}^z \rangle = \sigma_\nu \frac{dQ_\lambda(a)}{da} + \left(\frac{d^2 Q_\lambda(a)}{da^2} - \sigma_\lambda \frac{dQ_\lambda(a)}{da} \right) \\ \times \left. \frac{\langle S_{\lambda\nu}^z S_{\nu\mu}^z \rangle - \sigma_\lambda \sigma_\nu}{M_\lambda - \sigma_\lambda^2} \right|_{a=0} . \quad (3.29)$$

This gives

$$L'_\lambda = \sigma_\nu M_\lambda + (\varphi_{\lambda\nu}^0 - \sigma_\nu \sigma_\lambda) \frac{\rho_\lambda - \sigma_\lambda M_\lambda}{M_\lambda - \sigma_\lambda^2} , \quad (3.30)$$

where $\rho_\lambda = (S_\lambda^z)^2$, which is obtained from (3.25). Substituting these results in Eq. (3.11) one can now write down $\phi_{\lambda\nu}^0$ in terms of known functions $\psi_{\lambda\nu}^0$, σ_λ , and M_λ only. On inserting these in Eq. (3.7) one gets

$$A_1^{\lambda\nu} = A_2^{\lambda\nu} = \frac{\frac{1}{2} \psi_{\lambda\nu}^0 [3M_\nu + \gamma_\lambda - S(S+1)] - \sigma_\nu [S(S+1)\sigma_\nu + \gamma_\lambda \sigma_\lambda - M_\lambda \sigma_\nu]}{S(S+1)\sigma_\nu - M_\lambda \sigma_\nu + \sigma_\lambda \gamma_\lambda - \psi_{\lambda\nu}^0 \sigma_\lambda} , \quad (3.31)$$

where $\gamma_\lambda = \sigma_\nu (\rho_\lambda - \sigma_\lambda M_\lambda) / (M_\lambda - \sigma_\lambda^2)$. For the simpler case of $S = \frac{1}{2}$, $M_\lambda = M_\nu = \frac{1}{4}$, $\rho_\lambda = \frac{1}{4} \sigma_\lambda$, and $\gamma_\lambda = 0$, one easily gets back the earlier relations (3.10).

IV. ENERGY SPECTRUM, TRANSVERSE CORRELATION FUNCTIONS, AND LOW-TEMPERATURE THERMODYNAMICS

Let us introduce the Fourier transforms of the spin operators

$$\vec{S}_\lambda(\vec{k}) = \sum_i \vec{S}_{\lambda i} e^{-i\vec{k} \cdot \vec{R}_i} , \quad (4.1)$$

$$\vec{S}_{\lambda i} = \frac{2}{N} \sum_{\vec{k}} \vec{S}_\lambda(\vec{k}) e^{i\vec{k} \cdot \vec{R}_i} . \quad (4.2)$$

These obey the following commutation rules

$$[S_\lambda^+(\vec{k}_1), S_\mu^-(\vec{k}_2)] = 2S_\lambda^z(\vec{k}_1 + \vec{k}_2) \delta_{\lambda\mu} , \\ [S_\lambda^+(\vec{k}_1), S_\mu^+(\vec{k}_2)] = \pm S_\lambda^z(\vec{k}_1 + \vec{k}_2) \delta_{\lambda\mu} . \quad (4.3)$$

Using Eqs. (3.8a) and (3.21),

$$\langle S_\mu^-(\vec{k}_1) S_\lambda^+(\vec{k}_2) \rangle = (N/2E_{\vec{k}}) \delta_{\vec{k}_1, -\vec{k}_2} \delta_{\mu, \lambda} [A^{\lambda\mu} f(E_{\vec{k}} + \mu_B H) \\ + B^{\lambda\mu} f(E_{\vec{k}} - \mu_B H)] , \quad (4.4)$$

where $f(x)$ is the Bose function $(e^{2x} - 1)^{-1}$. In the absence of any external field, using Eqs. (3.20), (4.3), and the following relation for the Bose functions:

$$(e^{2x} - 1)^{-1} = \coth(\frac{1}{2} \beta x) - \frac{1}{2} , \quad (4.5)$$

the various correlation functions can be written as follows:

$$\langle S_A^-(-\vec{k}) S_A^+(\vec{k}) \rangle = \langle S_B^-(-\vec{k}) S_B^+(\vec{k}) \rangle \\ = (\frac{1}{2} N \sigma_A) [J \partial A_1 E_{\vec{k}}^{-1} \coth(\frac{1}{2} \beta E_{\vec{k}}) - 1] , \\ \langle S_A^-(-\vec{k}) S_A^-(\vec{k}) \rangle = \langle S_B^-(-\vec{k}) S_B^-(\vec{k}) \rangle \\ = (\frac{1}{2} N \sigma_A) [J \partial A_1 E_{\vec{k}}^{-1} \coth(\frac{1}{2} \beta E_{\vec{k}}) + 1] , \\ \langle S_A^+(\vec{k}) S_B^-(\vec{k}) \rangle = \langle S_B^-(-\vec{k}) S_A^+(\vec{k}) \rangle \quad (4.6)$$

$$= \langle S_A^+(\vec{k}) S_B^-(-\vec{k}) \rangle = \langle S_B^+(\vec{k}) S_A^-(-\vec{k}) \rangle \\ = -\frac{1}{2} N \sigma_A J \partial A_1 \gamma_{\vec{k}} E_{\vec{k}}^{-1} \coth(\frac{1}{2} \beta E_{\vec{k}}) .$$

The transverse correlation functions of two spins, i. e., $\langle S_{\lambda i}^z S_{\mu j}^z \rangle$ are given by the inverse Fourier transforms of Eqs. (4.6). The energy spectrum (3.18) can be written

$$E_{\vec{k}} = J \partial A_1 (1 - \gamma_{\vec{k}}^2)^{1/2} , \quad (4.7)$$

where A_1 is given by Eq. (3.31). $A_1 = \sigma_A$ is the result of decoupling by RPA, while $R = 1$ corresponds to noninteracting spin-wave theory.

We will now investigate the consequence of the foregoing results on the low-temperature thermodynamics of the system. In this range of temperature, the function $[\Omega(0) - \frac{1}{2}]$ can be expanded in powers of the reduced temperature $\tau (\equiv k_B T / J \partial)$, following standard techniques,²² which we will not repeat. For $H = 0$ it gives

$$\Omega(0) - \frac{1}{2} = \frac{1}{2} C' + a_0 (\tau / A_1)^2 + a_1 (\tau / A_1)^4 + O(\tau^6) , \quad (4.8)$$

where

$$C' = \frac{2}{N} \sum_{\vec{k}} (1 - \gamma_{\vec{k}}^2)^{-1} - 1 \\ = \begin{cases} 0.156 & \text{for sc,} \\ 0.150 & \text{for bcc,} \end{cases}$$

and the constants a_0 and a_1 are given by

$$a_0 = \frac{\zeta(2)}{\pi^2} q , \quad a_1 = \frac{\zeta(4)}{\pi^2} 6q ,$$

where $\zeta(n)$ are the Riemann ζ -function $\zeta(n) = \sum_{p=1}^{\infty} p^{-n}$, the factor q being $3^{3/2}$ for an sc and 4 for a bcc lattices. A similar expansion can be made in the Eq. (4.6) giving

$$\psi_{\lambda\nu}^0 = -2\sigma_A [\frac{1}{2} C_0 + a_0 (\tau / A_1)^2 + O(\tau^6)] , \quad (4.9)$$

where

$$C_0 = \frac{2}{N} \sum_k \gamma_k^2 (1 - \gamma_k^2)^{1/2} \\ = \begin{cases} 0.253 & \text{for sc,} \\ 0.223 & \text{for bcc.} \end{cases}$$

The series expansions of σ_A , $M_A = M_B$, and $\gamma_A = \gamma_B$ are as follows:

$$\sigma_A = S - [\Omega(0) - \frac{1}{2}] + (2S+1)[\Omega(0) - \frac{1}{2}]^2 S^{+1} + \dots \\ = -\sigma_B, \quad (4.10)$$

$$M = S^2 + (1-2S)[\Omega(0) - \frac{1}{2}] \\ + 2[\Omega(0) - \frac{1}{2}]^2 - (2S+1)[\Omega(0) - \frac{1}{2}]^2 S^{+1}, \quad (4.11)$$

$$\gamma = -S(2S-1) - (1-6S)[\Omega(0) - \frac{1}{2}] \\ - 2S^2(2S+1)^2[\Omega(0) - \frac{1}{2}]^2 S^{+1} + \dots \quad (4.12)$$

Substituting these in Eq. (3.31) we get

$$A_1 = S - [\Omega(0) - \frac{1}{2}] - \psi_{\lambda\nu}/2S + \dots \quad (4.13)$$

An iterative procedure can be now set up as follows: Starting with $A_1 = S$ and $\sigma_A = S$ in the zeroth order, $[\Omega(0) - \frac{1}{2}]$ and $\psi_{\lambda\nu}^0$ are calculated from Eqs. (4.8) and (4.9). These provide the first-order values for A_1 and σ_A . It is necessary to calculate $\psi_{\lambda\nu}^0$ and A_1 correctly to the order τ^2 , in order to calculate σ_A correctly to the order τ^4 . Neglecting all products of the small quantities C_0 and C' , we get ($C \equiv C_0 - C'$)

$$S = \frac{1}{2}: \\ \sigma_A = \frac{1}{2} - \frac{1}{2}C' - 4a_0\tau^2[1 - 2(C+C')] - 16[a_1(1 - 2C' - 4C) \\ - 2a_0^2(1 - 5C - 2C')], \quad (4.14)$$

$$S > \frac{1}{2}: \\ \sigma_A = S - \frac{1}{2}C' - a_0\tau^2[1 - (C/S)]/S^2 - \tau^4\{[1 - (2C/S)]a_1 \\ + a_0^2(C + 2C')/2S^2 - (2S+1)^2 a_0^2 S(C'/2)(\tau/S)^{4S-4}\}/S^4, \quad (4.15)$$

$$S \geq \frac{1}{2}: \\ A = S + \frac{1}{2}C - a_0\tau^2(C + 2C')/2S^3. \quad (4.16)$$

These results are to be compared with the spin-wave results of Oguchi²³

$$S \geq \frac{1}{2}: \\ \sigma_A = S - \frac{1}{2}C' - a_0[1 - (c/S)]\tau^2/S^2 \\ - a_1[1 - (2C/S)]\tau^4/S^4 + \dots \quad (4.17)$$

It is noted that the temperature-independent terms, as well as the τ^2 term, agree with the spin-wave theory for $S > \frac{1}{2}$. The corresponding RPA results gives the small correction C/S to the τ^2 term wrong. For the τ^4 term, the extra terms proportional to a_0^2 appear in our result. The deviation is somewhat large for $S=1$, but decreases sharply

for higher values of S . Anderson and Callen's⁹ coefficient of τ^4 gives better agreement with the spin-wave result for $S=1$, but for higher S values our agreement is better. For $S = \frac{1}{2}$, the temperature-independent terms agree with (4.17) for the RPA, the Anderson-Callen, and SCMD approaches. However, the correction of order C in the τ^2 term disagrees in all the three cases.

The perpendicular susceptibility is determined by the correlation functions, through the well-known relation²⁴

$$\chi^\perp = \beta\mu_B^2 \sum_{i,j} \langle S_i^x S_j^x \rangle. \quad (4.18)$$

Expressing the operators S_i^x in terms of S_i^z , one finds, using (4.6) that

$$\chi^\perp = \frac{1}{2} \mu^2 N \sigma_A / J \partial A_1. \quad (4.19)$$

Using the expansions (4.14)–(4.16), we get the following expansions for χ^\perp :

$S > \frac{1}{2}$:

$$\chi^\perp = \frac{\mu_B^2 N}{2J\partial} \left[1 + \frac{C+C'}{2S} - a_0 \left(1 - \frac{5C+4C'}{2S} \right) \frac{\tau^2}{S^3} + \dots \right], \quad (4.20)$$

$S = \frac{1}{2}$:

$$\chi^\perp = \frac{\mu_B^2 N}{2J\partial} [1 - (C+C') - 8a_0\tau^2(1 - 4C' + 3C)]. \quad (4.21)$$

The spin-wave result of Oguchi is

$$\chi^\perp = \frac{\mu_B^2 N}{2J\partial} \left[1 - \frac{C+C'}{2S} - a_0 \left(1 - \frac{C}{S} \right) \frac{\tau^2}{S^3} \right], \quad (4.22)$$

while the RPA gives the temperature-independent result

$$\chi^\perp = \mu_B^2 N / 2J\partial. \quad (4.23)$$

Thus, the temperature-independent term of SCMD agrees with the spin-wave theory, whereas the RPA result does not give the zeroth-order quantum correction. The τ^2 term again deviates from its form in (4.22) by a few percent.

V. HIGH-TEMPERATURE THERMODYNAMICS

A. Néel-Temperature Thermodynamics

In this section we will obtain expressions for some thermodynamic quantities at high temperatures. However, first we determine the Néel temperature T_N , at which the sublattice magnetization vanishes in the absence of any external field.

Putting $H=0$, the sublattice magnetization is seen from Eq. (3.26) to be given by

$$\sigma_A = -\sigma_B = S + \frac{1}{2} - \Omega(0) + \frac{(2S+1)[\Omega(0) - \frac{1}{2}]^2 S^{+1}}{[\Omega(0) + \frac{1}{2}]^2 S^{+1} - [\Omega(0) - \frac{1}{2}]^2 S^{+1}}. \quad (5.1)$$

At very high temperature, $\varphi_u = \varphi_l \equiv \varphi$ is given by

$$\varphi = -\frac{1}{2} + (\beta E_k)^{-1} + \frac{1}{12} \beta E_k + O(\beta^3)$$

and

$$\Omega(0) = I_0 / \beta J \partial A_1 + \frac{1}{12} \beta J \partial A_1 + \dots, \quad (5.2)$$

where $I_0 = (2/N) \sum_k (1 - \gamma_k^2)^{-1}$ has been evaluated by Watson.²⁵ For sc and bcc lattices I_0 has been value 1.51638 and 1.3932, respectively. Thus $[\Omega(0)]^{-1}$ can be regarded as a small parameter in which one can expand various quantities. At such temperatures σ_A , M_A , and γ_A are given by

$$\begin{aligned} \sigma_A &= x[\Omega^{-1}(0) - \Omega^{-3}(0)(\frac{1}{5}x - \frac{1}{20})] = -\sigma_B, \\ M_A &= x + 2x(\frac{1}{5}x - \frac{1}{20})\Omega^{-2}(0) = M_B, \\ \gamma_A &= x(1 - 4x)/5\Omega^2(0), \end{aligned} \quad (5.3)$$

where $x = \frac{1}{3}S(S+1)$, and the neglected terms are of the order $\Omega^{-4}(0)$ or less.

There nearest-neighbor correlation functions $\psi_{\lambda\nu}^0$ can be determined from the Eq. (3.21) to be (for $H=0$)

$$\psi_{\lambda\nu}^0 = -2\sigma_A \frac{2}{N} \sum_k (2\varphi + 1) \frac{\gamma_k^2}{2(1 - \gamma_k^2)^{1/2}} \quad (5.4)$$

For high temperatures $\psi_{\lambda\nu}^0$ can be expressed as follows:

$$\psi_{\lambda\nu}^0 = 2\sigma_A \left((1 - I_0)(J \partial A_1 \beta)^{-1} - \frac{1}{8} J \partial A_1 \beta \frac{2}{N} \sum_k \gamma_k^2 \right).$$

Using the high-temperature expansion for σ_A ,

$$\psi_{\lambda\nu}^0 \approx 2x[(1 - I_0)/I_0] + O(\beta^2). \quad (5.5)$$

If we substitute the foregoing results in the expression for A_1 , we obtain

$$A_1 = \alpha \Omega^{-1}(0) + O(\Omega^{-3}(0)), \quad (5.6)$$

where

$$\alpha = \frac{I_0}{2} \left(\frac{1 - I_0}{10I_0} (1 - 4x) + 2x \right). \quad (5.7)$$

The Néel temperature is determined by $\lim_{T \rightarrow T_N} \sigma_A = 0$. At such high temperatures σ_A is given essentially by the first term of its expansion in (5.3),

$$\lim_{T \rightarrow T_N} \sigma_A = x\Omega^{-1}(0) = x\beta_N J \partial A_1(\tau_N)/I_0, \quad (5.8)$$

where $\beta_N = 1/k_B T_N$. Thus at $T = T_N$, A_1 must vanish identically. From Eqs. (5.3) and (5.6) it is seen that to order β ,

$$\left(\frac{1}{\beta J \partial A_1} I_0 + \frac{1}{12} \beta J \partial A_1 \right) A_1 = \alpha,$$

giving $\beta J \partial A_1 = [12(\beta J \partial \alpha - I_0)]^{1/2}$. Thus, at $T = T_N$, we get

$$J \partial / k_B T_N = I_0 / \alpha. \quad (5.9)$$

In Table I we compare this with the RPA and the Anderson-Callen results. It is found that the T_N predicted here is too high compared to any of

TABLE I. Reduced Néel temperature ($k_B T_N / J \partial$) for simple cubic and body-centered-cubic lattices.

S	RPA	Anderson- Callen (Ref. 9)	Present
Simple cubic			
$\frac{1}{2}$	0.17	0.22	0.25
1	0.44	0.54	0.69
$\frac{3}{2}$	0.82	0.98	1.31
2	1.32	1.54	2.11
Body-centered cubic			
$\frac{1}{2}$	0.18	0.23	0.25
1	0.48	0.57	0.68
$\frac{3}{2}$	0.90	1.04	1.30
2	1.44	1.64	2.09

these, and is almost equal to the molecular-field value. It is found that the T_N predicted by the theory is somewhat greater than T_c value calculated by Tahir-Kheli in the same scheme. This is in conformity with Rushbrooke and Wood's estimate

$$T_N = T_c [1 + 0.63/\partial S(S+1)].$$

The high value of T_N itself is very disturbing, and the answer must lie in the very nature of the approximation. However we have no satisfactory theoretical explanation to this at present.

The perpendicular susceptibility is determined by the relation

$$\chi^{\perp} = \frac{1}{2} \mu^2 N \sigma_A / J \partial A_1. \quad (5.10)$$

From Eq. (5.8) the ratio $(\sigma_A/A_1) = \chi \beta_N J \partial / I_0$ at the Néel temperature. Thus the susceptibility at the Néel temperature is given by

$$\chi(\tau_N) = \frac{\mu^2 N}{2J \partial} \frac{S(S+1)}{3I_0} \left(\frac{J \partial}{k_B T_N} \right). \quad (5.11)$$

The molar susceptibility at the Néel temperature $\chi_M(T_N)$ can be written

$$\chi_M(T_N) = C_M / 2I_0 T_N, \quad (5.12)$$

where C_M the molar Curie constant. In Table II, we have listed the molar susceptibility of six compounds MnF_2 , FeF_2 , $LaFeF_2$, $LaCrO_2$, and $KCoF_3$. The first two of these have rutile structure in which the magnetic lattice is body centered tetragonal. However, it seems likely that the important interactions are super exchange interactions between a "central" atom, and its eight corner neighbors.²⁶ The other four have all pseudo-perovskite structures, in which the magnetic lattice is approximately sc. All these compounds conform to the nearest-neighbor model discussed here.²⁷ In calculating the theoretical numbers, we have used the experimental C_M values in some

TABLE II. Molar susceptibility at the Néel temperature. The experimental values of C_M marked with an asterisk are taken from Ref. 27.

Compounds	Z	S	T_N (°K)	C_M	$\chi_M(T_N)$	$\chi_M(T_N)$
					(cgs/mole)	Calc.
MnF ₂	8	$\frac{5}{2}$	68	4.38	0.025 1	0.023
FeF ₂	8	2	79	3.88*	0.019 6	0.018
LaFeF ₃	6	$\frac{5}{2}$	740	4.38	0.002 11	0.001 97
LaCrO ₃	6	$\frac{3}{2}$	320	1.88	0.001 96	0.001 96
KFeF ₃	6	2	115	3.38*	0.011 3	0.009 7
KCoF ₃	6	$\frac{3}{2}$	135	3.54	0.008 35	0.008 8

cases, as this enables us to take into account certain effects, such as the orbital contribution to the susceptibility, which cannot be estimated accurately by theoretical methods. Theoretical values have been taken in the other cases. The agreement is within a few percent of the experimental numbers. However it must be pointed out that $\chi_M(T_N)$ is notoriously insensitive to the theory and is correctly given by most of the decoupling schemes.

We will presently calculate the internal energy of the Heisenberg model at Néel temperature. This is given by

$$E = \langle \mathcal{H} \rangle = 2N^{-1}J \sum_{\mathbf{q}} \gamma_{\mathbf{q}} \langle \vec{S}_A(-\mathbf{q}) \cdot \vec{S}_B(\mathbf{q}) \rangle . \quad (5.13)$$

It is possible to calculate the internal energy for a system of particles with biquadratic interactions (the Ising model part of the Hamiltonian, in this case), from a knowledge of the single-particle Green's function. In this particular case of isotropic interactions, we can rewrite Eq. (5.13) as

$$E = 2N^{-1}J \sum_{\mathbf{q}} \frac{3}{2} \gamma_{\mathbf{q}} \langle S_A^-(\mathbf{q}) S_B^+(\mathbf{q}) \rangle , \quad (5.14)$$

which gives, using the expressions (4.6) for the correlation functions on the right,

$$E_N = -\frac{3}{2} \sigma_A J \sum_{\mathbf{q}} \frac{J \partial A_1 \gamma_{\mathbf{q}}^2}{E_{\mathbf{q}}} \coth \frac{\beta E_{\mathbf{q}}}{2} . \quad (5.15)$$

At an arbitrary temperature, a study of the internal energy or the resulting specific heat, is complicated, and is to be carried in a computer. At the Néel temperature T_N , the expression simplifies, and we can write

$$E_N = -\frac{3}{2} x \beta_N (I_0 - 1) / I_0 . \quad (5.16)$$

For $S = \frac{1}{2}$ it is seen that this is the same as estimated by Liu and Lee.¹¹

B. Analysis above Néel Temperature

The region above the Néel temperature ($T > T_N$) is paramagnetic, and the analysis is formally

identical to that carried out by Tahir-Kheli¹³ for the ferromagnet, except that the sign of the exchange integral is changed. We will merely quote the obvious analog of the high-temperature ($T \gg T_c$) susceptibility in our case, in order to avoid duplication of details. It is found that for $T \gg T_c$

$$\chi = \frac{x}{3k_B T} \left[1 - \left(\frac{J \partial}{k_B T} \right) x + \left(\frac{J \partial}{k_B T} \right)^2 x^2 [1 - (4x - 1) \times (20 \partial x)^{-1}] \right] + O(\tau^{-4}) . \quad (5.17)$$

It is seen that the above result agrees with the exact-series expansion only to the first two powers of $1/\tau$, as much as the other decoupling theories (cf. Refs. 10 and 22 for negative J).

The series (5.17) is in itself not very interesting. Whereas the coefficients of $1/\tau$ in the ferromagnetic phase are all positive, so that the ferromagnetic analog of (5.17) diverges at some critical point, viz., the Curie point, this is not true for the antiferromagnetic series, since the coefficients now alternate in sign. T_c still determines the radius of convergence of the high-temperature expansion, but does not correspond to a physical singularity in the susceptibility. The ordering temperature is determined by discussing the antiferromagnetic "staggered susceptibility" χ_S (the order parameter) reflecting the effect of a vanishingly small magnetic field, which changes sign at alternate sites. The Hamiltonian for discussing the staggered susceptibility is taken to be

$$\mathcal{H} = J \sum_{i \in A} \sum_{j \in B} \vec{S}_i \cdot \vec{S}_j - \mu_B H \left(\sum_{i \in A} S_i^z - \sum_{i \in B} S_i^z \right) . \quad (5.18)$$

We will take the vanishing-field limit at the end of the calculation. For (5.18), the Green's-function equations are the same as (3.16) and (3.17) except for the following replacements:

$$\begin{aligned} \alpha_1 &= -\alpha_2 = \mu_B H + J \partial A_1 , \\ E_k - \omega_k &\equiv [(J \partial A_1 + \mu_B H)^2 - (J \partial A_1 \gamma_k)^2]^{1/2} , \\ E_u &= -E_l = \omega_k . \end{aligned} \quad (5.19)$$

Equations (3.20) and (3.12) for the correlation-functions now take the following form:

$$\langle S_{A_i}^- S_{A_i}^+ \rangle = \sigma_A [2\Omega'(0) - 1] , \quad (5.20a)$$

$$\langle S_{B_i}^- S_{B_i}^+ \rangle = -\sigma_B [2\Omega'(0) + 1] , \quad (5.20b)$$

where

$$\Omega'(0) = \frac{2}{N} \sum_k \frac{1 + 2f(\omega_k)}{2\omega_k} (J \partial A_1 + \mu_B H) . \quad (5.21)$$

For temperatures above T_N , $f(\omega_k)$ can be expanded in the inverse powers of τ , and one gets, retaining only the leading term

$$\Omega'(0) = \frac{2}{N\beta} \sum_k \frac{\mu_B H + J \partial A_1}{(\mu_B H + J \partial A_1)^2 - (J \partial A_1 \gamma_k)^2} \quad (5.22)$$

Let us introduce the order parameter χ_S (the staggered susceptibility χ^A), and a quantity A , through the following relations:

$$\chi_S = \lim_{H \rightarrow 0} \sigma_A / \mu_B H \quad , \quad (5.23)$$

$$A = \lim_{H \rightarrow 0} A_1 / \sigma_A \quad . \quad (5.24)$$

Above the Néel temperature, $\sigma_A \rightarrow 0$ as $H \rightarrow 0$. As the temperature is lowered to the Néel point T_N , χ diverges in the limit of a vanishing magnetic field. Eq. (5.22) can now be written

$$\Omega'(0) = \frac{2}{N\beta\mu_B H} \sum_k \frac{1 + A\chi_S}{(1 + A\chi_S)^2 - (A\chi_S\gamma_k)^2} \quad (5.25)$$

It is seen that $\Omega'(0)$ diverges as $H \rightarrow 0$ ($T > T_N$), and thus in this limit, one can expand various quantities like σ_A , M_A , etc., in the inverse powers of $\Omega'(0)$. The expansions (5.3) for σ_A , M_A , etc., are still valid with the replacement $\Omega'(0)$ for $\Omega(0)$. Before proceeding further, we will cast (5.25) into a more convenient form. Using symmetry in the k space for cubic lattices, we can write

$$\Omega'(0) = (1/\beta\mu_B H) Q F(Q) \quad , \quad (5.26)$$

where

$$Q = (A\chi_S)^{-1} \quad (5.27)$$

and

$$F(Q) = (2/N) \sum_k (Q + 1 - \gamma_k)^{-1} \quad . \quad (5.28)$$

Substituting the expansions for σ_A , etc., in the expression (3.31) for A_1 , and using (5.27)–(5.28), we get

$$\frac{J \partial Q \chi_S}{x} = \frac{2x + \psi_0}{2x^2 - \frac{1}{20} \psi_0 (4x - 1)} \quad , \quad (5.29)$$

where $\psi_0 = \lim_{H \rightarrow 0} \psi^0$ is found to be given by

$$\psi_0 = [2x/F(Q)][1 - (1+Q)F(Q)] \quad . \quad (5.30)$$

In the same limit χ_S^{-1} is given by

$$\lim_{H \rightarrow 0} [\Omega(0)\mu_B H/x] = \chi_S^{-1} = QF(Q)/\beta x \quad . \quad (5.31)$$

The set of equations (5.29)–(5.31) can be solved simultaneously, and using Eqs. (5.9) for the Néel temperature T_N , we get the following convenient expression:

$$\begin{aligned} \frac{20k_B(T - T_N)}{J \partial} &= (24x - 1)[(1 - \beta\chi_S^{-1}x)^{-1} - 1] \\ &+ (4x - 1)Q(1 - \beta\chi_S^{-1}x)^{-1} - (4x - 1) \\ &\times [F^{-1}(Q)(1 - \beta\chi_S^{-1}x)^{-1} - F(0)^{-1}] \quad , \end{aligned} \quad (5.32)$$

where $F(0) = I_0$.

The solution of (5.23) for $S = \frac{1}{2}$ is straightforward, for in this case $4x - 1$ vanishes identically and one gets $\chi_S \sim (T - T_N)^{-1}$. For the case of $S > \frac{1}{2}$ we have to divide the range of temperature into one close to T_N and the other much higher.

(a) $(T - T_N) \ll T_N$: In this region, the susceptibility χ is expected to be small, and as such Q is expected to be small. In this case we use the expansion²⁸

$$F(Q) = F(0) - bQ^{1/2} \quad ,$$

where

$$\begin{aligned} b &= 3\sqrt{3}/\pi\sqrt{2} \quad \text{for sc} \\ &= 2\sqrt{2}/\pi \quad \text{for bcc} \\ &= 3\sqrt{3}/\pi \quad \text{for fcc.} \end{aligned}$$

Substituting this into (5.31)–(5.32), we get, for $S > \frac{1}{2}$,

$$[\chi_S]_{T - T_N \ll T_N} = \frac{(4x - 1)^2 b^2 (20x)(T/T_N - 1)^{-2}}{J \partial [(24x - 1)F^2(0) - (4x - 1)^3]} \quad . \quad (5.32)$$

Thus the exponent of the staggered susceptibility is given by

$$\begin{aligned} \gamma &= 1 \quad \text{for } S = \frac{1}{2} \\ &= 2 \quad \text{for } S > \frac{1}{2} \quad . \end{aligned} \quad (5.33)$$

The value of γ does not seem to have been estimated with much reliability, although for $S = \infty$ we must have $\gamma = 1.33$, for symmetry with the ferromagnetic case.²⁹ The RPA value for the critical exponent is 2 while the Landau theory gives $\gamma = 1$ (see p. 660 of Ref. 29).

(b) $T \gg T_N$: In this case $Q \gg 1$, and we can use

$$F(Q) = Q^{-1}[1 - Q^{-1} + (1 + 1/\partial)Q^{-2}] + O(Q^{-4}) \quad . \quad (5.34)$$

Inserting (5.34) into (5.31), we get for $T \gg T_N$

$$\chi_S^{-1} = (k_B T/x)[1 - Q^{-1} + (1 + 1/\partial)Q^{-2}] \quad (5.35)$$

and (5.23) gives

$$\frac{k_B T}{J \partial x} = 2Q + \frac{(24x - 1)(1 + 1/\partial) - 4x - 1}{20x} + O(Q^{-1}) \quad . \quad (5.36)$$

Solving (5.35)–(5.36) gives

$$[\chi_S]_{T \gg T_N} = \frac{x}{k_B T} \left[1 + \frac{2J \partial x}{k_B T} + (2 + 2\alpha) \left(\frac{J \partial x}{k_B T} \right)^2 + \dots \right] \quad . \quad (5.37)$$

Comparing (5.37) with the series expansion of Rushbrook and Wood³⁰ we find that the staggered susceptibility agrees only to first two terms.

VI. RESULTS IN RESTRICTED DIMENSIONALITIES

In this section, and in Sec. VII, we restrict ourselves to one and two dimensions. This is important because of an exact theorem by Mermin and

Wagner,¹⁶ which rules out the occurrence of the antiferromagnetic order in one and two dimensions. However, their proof does not preclude the existence of a second- or higher-order continuous phase transitions, where $(\partial^n/\partial H^n)S^e \rightarrow \infty$ while $S^e \rightarrow 0$ for $H \rightarrow 0$. It is generally believed, however, on the basis of a theorem due to VanHove,³¹ that phase transitions of any kind will not occur in an one-dimensional system, whereas in the two dimensions, Stanley and Kaplan¹⁷ have predicted a second-order phase transition. However, the usual decoupling schemes⁹⁻¹¹ predict no phase transitions in these dimensionalities, whereas the molecular-field theory predicts a phase transition in all the dimensions. In the following, we shall examine these questions within the framework of SCMD scheme. We consider the case of $S = \frac{1}{2}$ for simplicity, but the general spin case follows in an analogous fashion.

In the presence of a magnetic field, which alternate in sign from site to site, the Green's-function equations (3.14a) and (3.14b) are still valid, except for the identification

$$\alpha_1 = -\alpha_2 = \mu_B H + J \delta A_1 ,$$

instead of (3.15a). The nearest-neighbor transverse correlation function ψ_{AB}^0 , and the magnetization σ_A , are given by

$$\sigma_A^{-1} = \frac{4}{N} \sum_k \frac{J \delta A_1 + \mu_B H}{\omega_k} \coth(\frac{1}{2} \beta \omega_k) \quad (6.1)$$

and

$$\psi^0 = -\frac{2}{N} \sigma_A \sum_k \frac{(J \delta A_1 + \mu_B H)}{\omega_k} \gamma_k^2 \coth(\frac{1}{2} \beta \omega_k) , \quad (6.2)$$

where $\omega_k^2 = (\mu_B H + J \delta A_1)^2 - (J z A_1 \gamma_k)^2$. We will now construct the quantity A_1^{-1} using Eqs. (6.1) and (6.2). By definition, A_1^{-1} is positive, and is given by

$$\begin{aligned} A_{11}^{-1} &= \sigma_A^{-1} (1 + 2\psi^0) \\ &= \frac{4}{N} \sum_k \frac{(J \delta A_1 + \mu_B H)(1 - \gamma_k^2)}{\omega_k} \coth(\frac{1}{2} \beta \omega_k) . \end{aligned} \quad (6.3)$$

Now, the hyperbolic cotangent has the property

$$\coth x \geq 1/|x| , \quad (6.4)$$

and thus we can set up an inequality for the positive quantity A_1^{-1} ,

$$A_1^{-1} \geq \frac{8}{N\beta} \sum_k \frac{(J \delta A_1 + \mu_B H)(1 - \gamma_k^2)}{(J \delta A_1 + \mu_B H)^2 - (J \delta A_1 \gamma_k)^2} . \quad (6.5)$$

We will now specialize above results to one and two dimensions.

A. One Dimension

In the case of a linear chain with nearest-neighbor interactions, we have $\delta = 2$ and $\gamma_k = \cos k$.

Replacing above sum by an integration through the prescription

$$\frac{2}{N} \sum_k \rightarrow \frac{\Omega}{(2\pi)^n} \int_{-\pi}^{+\pi} d^n k , \quad (6.6)$$

where Ω is the unit cell volume, to be normalized to unity, and n the number of dimensions.

Thus in one dimension, we have

$$A_1^{-1} \geq \frac{2}{\pi\beta} \int_{-\pi}^{+\pi} \frac{(J z A_1 + \mu_B H) \sin^2 k}{(J z A_1 + \mu_B H)^2 - (J z A_1)^2 \cos^2 k} . \quad (6.7)$$

The integral on the right-hand side of Eq. (6.7) can be simplified using the various properties of the integrand, and one can obtain

$$1 \geq \frac{1}{\pi\beta J} \int_{-\pi}^{+\pi} \frac{\sin^2 k}{(1 + \mu_B H/J z A_1) - \cos^2 k} . \quad (6.8)$$

Substituting the value of the integral on the right, we get

$$\frac{\beta J}{2} - 1 \geq \frac{\mu_B H}{2 J A_1} \left[1 - \left(1 + \frac{4 J A_1}{\mu_B H} \right)^{1/2} \right] . \quad (6.9)$$

The inequality (6.9) presents the following cases of interest.

(a) $\frac{1}{2} \beta J < 1$: The left-hand side of (6.9) becomes negative, and hence the right hand side must also be so. After a little algebraic manipulation, one gets from (6.9)

$$A_1 \leq \frac{\beta}{4(1 - \frac{1}{2} \beta J)} \mu_B H , \quad (6.10a)$$

showing that in the limit of vanishing field, A_1 can be written

$$\lim_{H \rightarrow 0} A_1 = f(\beta, J) H , \quad (6.10a)$$

where $f(\beta, J)$ is a positive definite function of the temperature, and the exchange constant J .

(b) $\frac{1}{2} \beta J = 1$: In this case nothing definite can be said about the behavior of A_1 in the limit of small field by using (6.9). Hence we return to Eq.

(6.3), and use the inequality

$$\coth x \leq 1/|x| + \frac{1}{3} x . \quad (6.11)$$

This gives

$$\begin{aligned} \frac{\beta J}{2} - 1 \leq \frac{\mu_B H}{2 J A_1} \left[1 - \left(1 + \frac{4 J A_1}{\mu_B H} \right)^{1/2} \right] + \frac{1}{6} (\beta J A_1)^2 \\ + \frac{1}{12} J A_1^2 \mu_B H , \end{aligned} \quad (6.12)$$

In order that both the inequalities (6.9) and (6.12) be satisfied simultaneously at $\frac{1}{2} \beta J = 1$, it is necessary that A_1 must go to zero slower than H itself, for then it is possible to satisfy the equalities in both the cases. It can be easily checked that A_1 must go to zero no faster than $J^{-1/5} H^{1/5}$ as $H \rightarrow 0$.

(c) $\frac{1}{2} \beta J > 1$: In this case it can be easily shown that A_1 has a finite value even in the absence of

a field. Putting $H=0$, the inequalities corresponding to (6.8) and (6.12) can be worked out starting with the Eq. (6.3). They are

$$1 \leq 2/\beta J + \frac{1}{3} \beta J A_1^2 \quad (6.13a)$$

and

$$1 \geq 2/\beta J. \quad (6.13b)$$

While (6.13b) is merely a statement of the condition imposed, (6.13a) states that

$$\lim_{H \rightarrow 0} A_1 \geq (3/\beta J)[1 - 2/\beta J], \quad (6.14)$$

the right-hand side of which is positive definite. In summary, the behavior of $\lim_{H \rightarrow 0} A_1$ as a function of H in various ranges of temperature ranges is the following:

$$\begin{aligned} \lim_{H \rightarrow 0} A_1 &= \text{const} \times H & \frac{1}{2} \beta J < 1 \\ &= \text{const} \times H^{1/5} & \frac{1}{2} \beta J = 1 \\ &= \text{const}, & \frac{1}{2} \beta J > 1. \end{aligned}$$

Since A is the effective-field parameter of our theory, a shift from a linear to a power law signifies a change at $\frac{1}{2} \beta J = 1$, in the response of the spin system to an external field. We will presently examine the susceptibility and the spontaneous magnetization.

The static magnetic susceptibility of the sublattice A is defined in terms of the sublattice magnetization σ_A by the relation

$$\chi_A = \lim_{H \rightarrow 0} \sigma_A / \mu_B H. \quad (6.15)$$

For high temperatures we obtain

$$(\chi_A \mu_B H)^{-1} \geq 2(\pi\beta)^{-1} \int_{-\pi}^{+\pi} dk [(J \partial A_1 + \mu_B H) - J \partial A_1 \cos k].$$

Substituting the value of the integral on the right we get

$$\chi_A^{-1} \geq (2\mu_B H / \beta) [(J \partial A_1 + \mu_B H)^2 - (J \partial A_1)^2]^{-1/2}. \quad (6.16)$$

Further, using the inequality (6.11) for the hyperbolic cotangent

$$\begin{aligned} \chi_A^{-1} &\leq (2\mu_B H / \beta) [(J \partial A_1 + \mu_B H)^2 - (J \partial A_1)^2]^{-1/2} \\ &\quad + \frac{1}{3} \beta \mu_B H (J \partial A_1 + \mu_B H). \end{aligned} \quad (6.17)$$

As long as $\frac{1}{2} \beta J < 1$, the effective-field parameter A_1 goes to zero with H , according to (6.10a) and (6.10b). Thus only the equality signs in (6.16) and (6.17) can simultaneously be satisfied. Recognizing the role of $\frac{1}{2} \beta J$ as a turning point, let us define a critical temperature T_c by the relation

$$\frac{1}{2} \beta J = T_c / T; \quad (6.18)$$

it is immediately observed that above T_c the zero-field susceptibility goes like $(T - T_c)^{-1}$. At

the critical point itself, a similar analysis using the case (b) above shows that

$$\lim_{T \rightarrow T_c} \chi = \text{const} \times H^{-2/5}. \quad (6.19)$$

Let us now examine what happens to the sublattice magnetization itself. For $\frac{1}{2} \beta J \leq 1$, we have shown that

$$\lim_{H \rightarrow 0} A_1 = \lim_{H \rightarrow 0} \sigma_A (1 + 2\psi)^{-1} = 0.$$

Since ψ^0 is bounded, this suggests that $\lim_{H \rightarrow 0} \sigma_A = 0$. For $\frac{1}{2} \beta J > 1$, A_1 is seen to be independent of H . As before, we set up the inequality

$$\sigma_A^{-1} \geq \frac{2}{\pi\beta} \int_{-\pi}^{+\pi} \frac{dk}{(2JA_1 + \mu_B H) - 2JA_1 \cos k}. \quad (6.20)$$

The integral on the right is easily performed, and in the limit of the small H one obtains

$$\sigma_A \leq (\text{const}/T) |H|^{1/2}. \quad (6.21)$$

It is observed that there is no spontaneous magnetic ordering in a one-dimensional chain, consistent with the exact theorem of Mermin and Wagner.¹⁶ The existence of a nonzero transition temperature, for a second-order phase transition in one dimension, is somewhat disturbing, and is probably erroneous.³²

VII. RESULTS IN TWO DIMENSIONS

The analysis of the two-dimensional case is somewhat more complicated because of the fact that the integrals involved here are not expressible, in terms of simple algebraic or trigonometric functions, as before. Further, we have to restrict ourselves to a particular lattice, which we take to be quadratic structure, for convenience. In this case $\gamma = \frac{1}{2} (\cos k_x + \cos k_y)$, and the inequality for A_1^{-1} corresponding to (6.7) can be written

$$\begin{aligned} A_1^{-1} &\geq \frac{1}{\pi^2 \beta} \int_{-\pi}^{+\pi} dk_x \int_{-\pi}^{+\pi} dk_y \\ &\quad \times \frac{(J \partial A_1 + \mu_B H) [4 - (\cos k_x + \cos k_y)^2]}{4(J \partial A_1 + \mu_B H)^2 - (J \partial A_1)^2}, \end{aligned} \quad (7.1)$$

which can be simplified, first by expressing the integrand as partial fractions, and then using its various symmetry properties, to give

$$\begin{aligned} A_1^{-1} &\geq \frac{1}{2\pi^2 \beta} \int_{-\pi}^{+\pi} dk_x \int_{-\pi}^{+\pi} dk_y \\ &\quad \times \frac{4 - (\cos k_x + \cos k_y)^2}{2(J \partial A_1 + \mu_B H) - J \partial A_1 (\cos k_x + \cos k_y)}. \end{aligned} \quad (7.2)$$

The integral on the right can be performed following the method indicated in the Appendix. One obtains

$$A_1^{-1} \geq \frac{2}{\beta} \left\{ \frac{2(J\partial A_1 + \mu_B H)}{(J\partial A_1)^2} + 4I_0 \left[1 - \left(1 + \frac{\mu_B H}{J\partial A_1} \right)^2 \right] \right\}, \quad (7.3)$$

where

$$I_0 = \frac{1}{\pi J \partial A_1} \left(1 + \frac{\mu_B H}{J \partial A_1} \right)^{-1} K \left(\left(1 + \frac{\mu_B H}{J \partial A_1} \right)^{-1} \right), \quad (7.4)$$

$K(\xi)$ being the complete elliptic integral of the first kind defined through

$$K(\xi) = \int_0^{\pi/2} (1 - \xi^2 \sin^2 \varphi)^{-1/2} d\varphi. \quad (7.5)$$

The inequality (7.3) can be written

$$\beta J \geq \xi^{-1} - (2/\pi)\xi^{-1}(1 - \xi^2)K(\xi), \quad (7.6)$$

where $\xi = (1 + \mu_B H/4JA_1)^{-1}$, and we have put the coordination number of the square lattice, $\partial = 4$. With ξ so defined, it can be shown that,²³ for ξ approaching the value of unity from below ($\xi = 1 - 0^+$),

$$(1 - \xi)K(\xi) \approx \frac{1}{2} \left(\frac{\mu_B H}{4JA_1} \right) \left| \ln \frac{\mu_B H}{4JA_1} \right|. \quad (7.7)$$

As in the case of one dimension, we consider three cases of interest.

(a) $\beta J < 1$: In this case, if it is assumed that $\mu_B H/J\partial A_1 \rightarrow 0$ as $H \rightarrow 0$, then $\xi \lesssim 1$, making (7.7) valid. However, since $\lim_{x \rightarrow 0} x \ln|x| = 0$, the inequality (7.6) will imply $\beta J > \xi^{-1}$ contradicting the condition $\beta J < 1$, since ξ remains less than unity. Thus in this case, the ratio $\mu_B H/J\partial A_1$ must approach a finite nonzero value,

$$\lim_{H \rightarrow 0} A_1 = f(\beta, J)H,$$

where $f(\beta, J)$ is a function of J and β .

(b) $\beta J = 1$: In this case nothing definite can be said about the behavior of A_1 with H . Thus, as in the one-dimensional case, we consider the next term in the expansion of $\coth \frac{1}{2} \beta \omega_k$ to set up the following inequality:

$$\beta J \leq \xi^{-1} - (2/\pi)\xi^{-1}(1 - \xi^2)K(\xi) + \beta^2 J^2 A_1^2 + \frac{1}{4} J A_1 \mu_B H. \quad (7.8)$$

As $H \rightarrow 0$, conditions (7.7) and (7.8) can simultaneously be satisfied if A_1 goes to zero with H , for then both (7.7) and (7.8) become equalities. For $\xi \approx 1$, we can write as $H \rightarrow 0$;

$$(4/\pi)(1 - \xi)K(\xi) = \beta^2 J^2 A_1^2. \quad (7.8a)$$

Replacing, $K(\xi) \approx \frac{1}{2} |\ln(\mu_B H/J\partial A_1)|$, in the limit of $H \rightarrow 0$ we get

$$\frac{\mu_B H}{4JA_1} \left| \ln \frac{\mu_B H}{4JA_1} \right| = A_1^2. \quad (7.9)$$

Thus we obtain

$$\lim_{H \rightarrow 0} A_1 = \text{const} \times H^{1/3} |\ln H|^{1/3}. \quad (7.10)$$

(c) $\beta J > 1$: This case can obviously be consistent with (7.7) and (7.8) if A_1 is independent of H . Summarizing, we have shown that

$$\begin{aligned} \lim_{H \rightarrow 0} A_1 &= \text{const} \times H & \beta J < 1 \\ &= \text{const} \times (H \ln |H|)^{1/3} & \beta J = 1 \\ &= \text{const}, & \beta J > 1. \end{aligned} \quad (7.11)$$

Thus again we find that $\beta J = 1$ behaves as a singular point, at which the effective-field parameter A_1 changes its power law. As before for $\beta J \leq 1$ the vanishing of $A_1 = \sigma_A (1 + 2\psi^0)^{-1}$ as $H \rightarrow 0$ implies $\sigma_A \rightarrow 0$. For $\beta J < 1$, we have to examine the expression for the magnetization from (6.1). Thus for low temperatures

$$\begin{aligned} \sigma_A^{-1} &\geq \frac{2}{\pi^2 \beta} \int_{-\pi}^{+\pi} dk_x \int_{-\pi}^{+\pi} dk_y \\ &\times \frac{J \partial A_1 + \mu_B H}{2(J \partial A_1 + \mu_B H) - J \partial A_1 (\cos k_x + \cos k_y)}. \end{aligned} \quad (7.12)$$

Using the value of the integral on the right,

$$\sigma_A^{-1} \geq (2/\pi\beta)K(\xi). \quad (7.13)$$

In the limit of the small H , $K(\xi) \approx \frac{1}{2} |\ln |\mu_B/4JA_1||$, and thus

$$\sigma_A \leq \frac{\text{const}}{T} \frac{1}{|\ln |H||}. \quad (7.14)$$

Thus it is seen that a two-dimensional Heisenberg model cannot exhibit spontaneous antiferromagnetic order, a result which is in agreement with the exact theorem of Mermin and Wagner. It may be remarked that the exact way in which σ_A goes to zero is of no particular significance, since Mermin and Wagner's theorem is in the nature of an inequality involving the temperature T and the magnetic field H . By using a slightly different form of the inequality for the functions, it is possible to get, for instance, a stronger condition on H , while obtaining a weaker one on T , or vice versa.

The deviation from the linear behavior to a logarithmic law in the field dependence of A_1 , however, suggests existence of a continuous phase transition. The static magnetic susceptibility χ_A can be seen to obey the following inequalities:

$$\chi_A^{-1} \geq (8\mu_B H/\pi\beta)K(\xi), \quad (7.15)$$

$$\chi_A^{-1} \leq (8\mu_B H/\pi\beta)K(\xi) + \frac{1}{3} \beta \mu_B H (J \partial A_1 + \mu_B H). \quad (7.16)$$

As long as $\beta J \leq 1$, $A_1 \rightarrow 0$ as $H \rightarrow 0$, thus only the equality signs in (7.15) and (7.16) can simultaneously be satisfied:

$$\chi_A = \frac{1}{8} \beta \pi [\mu_A H K(\xi)]^{-1}. \quad (7.17)$$

The inequality (7.6) can be written in the following form, since as the temperature approaches T_c , A_1 goes to zero slower than $\mu_A H$, and very near T_c it can be assumed that $\xi \lesssim 1$. Thus one gets from (7.6), for $\beta J \lesssim 1$,

$$[\mu_B H K(\xi)]^{-1} \simeq 4JA_1/\pi(1 - \beta J)$$

giving

$$\chi_A = \beta/2(1 - \beta J) \equiv T_c/2(T_c - T) \quad (7.18)$$

Thus at a critical temperature T_c , defined by $T_c = J/k_B$, the zero-field susceptibility diverges as $(T_c - T)^{-1}$, for $T = T_c - 0^+$. At $T = T_c$, the inequality (7.8) is written as an equality for reasons mentioned earlier, and one can write

$$[\mu_B H K(\xi)]^{-1} = \text{const} \times H^{-1} \frac{1}{|\ln|H||} \quad (7.19)$$

Thus at $T = T_c$, the susceptibility χ_A diverges, according to the law $(H|\ln|H|)^{-1}$. The critical temperature T_c can be compactly written as $T_c = Jd/2k_B$, where $d = 1, 2$ denotes the dimensionality. It is noted however, that the T_c in this case, is exactly the molecular-field value, and is thus more than twice than that implied by Stanley and Kaplan series extrapolation. Our calculation seems to support Stanley and Kaplan's suggestion regarding a phase transition in two dimensions, whereas the usual decoupling schemes do not support this. All these schemes predict absence of long-range order in these dimensionalities. The validity of the existence of second-order phase transition is an open question and cannot be answered now.

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APPENDIX

The integral in (7.2) can be expressed as a linear combination of the following basic integral and other elementary ones, after suitable algebraic manipulations:

$$\mathcal{I} = \frac{1}{4\pi^2} \int_{-\pi}^{+\pi} dk_x \int_{-\pi}^{+\pi} dk_y \frac{1}{a - b(\cos k_x + \cos k_y)} \quad (A1)$$

where $a = 2(JzA_1 + \mu_B H)$ and $b = JzA_1$. It is observed that integrand does not have any pole in the x - y plane for $\text{Max}[b(\cos x + \cos y)] < a$. Thus it is possible to transform \mathcal{I} into the following form by introducing an additional integral.

$$\mathcal{I} = \frac{1}{4\pi^2} \int_0^\infty d\lambda \int_{-\pi}^{+\pi} dk_x \int_{-\pi}^{+\pi} dk_y \times \exp\{-\lambda[a - b(\cos k_x + \cos k_y)]\} \quad (A2)$$

The integrals over k_x and k_y can be performed by using the integral representation of the Bessel functions³³ of imaginary arguments:

$$J_n(iz) = \frac{(i)^n}{2\pi} \int_{-\pi}^{+\pi} e^{-iz \cos \theta} \cos n\theta d\theta \quad (n = 0, 1, 2, \dots) \quad (A3)$$

Thus, the integral \mathcal{I} can be expressed as a Laplace transform of a product of two such functions:

$$\mathcal{I} = \int_0^\infty d\lambda [J_0(i\lambda b)]^2 e^{-\lambda a} \quad (A4)$$

which can be obtained from standard tables,³⁴ to be

$$\mathcal{I} = \frac{2}{\pi a} K\left(\frac{2b}{a}\right) \quad (A5)$$

where K is the complete elliptic integral of the first kind.

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