[27]The fact that we might not be in the asymptotic regime would also manifest itself in higher-order terms in the expression for κ, that is, we could have

$$\kappa = \kappa_0 \tau^{\nu}(1 + a\tau + b\tau^2 \cdots) \ .$$

If a and b were much greater than unity, then our values for ν obtained from a simple power law ($a, b = 0$) could be grossly in error. With our present data we certainly cannot distinguish between various analytic forms for κ; hence we must simply assume a simple power law for the reduced temperature dependence until a detailed theory dictates otherwise. It is perhaps worth commenting, however, that the pertinent parameter may well be κ itself rather than τ and in that case, it is more likely that we are in the asymptotic regime.

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New Approach to Green's-Function Decoupling in Magnetism with Specific Application to Two-Dimensional Systems

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Existing first-order Green's-function theories of the Heisenberg ferromagnet all lead to magnon energies for which the temperature renormalization is wave vector independent. Such theories can describe phase transitions only at a temperature T_C for which *all* spin-wave excitations have vanishingly small energy. It has become increasingly evident, particularly for systems of low dimensionality, that such an approximation is quite unphysical, paramagnetic magnons often being physically well defined over much of the Brillouin zone to quite elevated temperatures. This paper describes a rather general method for introducing wave-vector-dependent magnon renormalization into the Green's-function formalism, enabling approximations of obvious physical significance to be made directly in terms of the magnon dispersion relation. The theory is developed in detail for the simplest nontrivial approximation and applied to the problem of the two-dimensional Heisenberg ferromagnet. A phase transition is found to a state of zero magnetization and infinite susceptibility. We also discuss the weakly anisotropic two-dimensional ferromagnet, which supports long-range order at low temperatures, and study the approach to the isotropic limit.

I. INTRODUCTION

Over the past several years, the technique of double-time temperature-dependent Green's functions has been applied with some success by many authors to a varied selection of statistical problems in ferro-, ferri-, and antiferromagnetism. Formally, the method produces solutions for the desired expectation values as functions of temperature, but these solutions are in the form of an infinite set of coupled first-order differential equations, and the development of a tractable formalism makes necessary a decoupling approximation.

The majority of decoupling procedures devised for Heisenberg systems, particularly in approximations applicable for general spin quantum number and aiming for validity over the entire temperature range, are made at the earliest possible stage of the calculation (i.e., in the differential equation involving the lowest-order Green's functions). This is done almost from necessity rather than choice, because of the enormous increase in mathematical complexity which results from delaying the decoupling to a later stage, [1] unless further approximations restricting the temperature range of validity are also introduced. A short list of publications, sufficient to give an indication of the developing sophistication in first-order Green's-function decoupling schemes, is given in Refs. 2–10.

Regardless of their degree of sophistication, these approximations all produce excitation energies for which the scaling with temperature is *wave vector independent*. That is, they all predict that short-wavelength and long-wavelength spin waves renormalize in the same way. A corollary is that they can describe well-behaved phase transitions only at the temperature for which *all* spin-wave-like excitations have vanishingly small energy. However, it is now well established experimentally that real magnetic second-order phase transitions are not at all like this but that the phase transition affects significantly only long-wavelength excitations. Both neutron diffraction[11,12] and Raman[13,14] experiments on magnetic systems reveal that short-wavelength excitations undergo no obvious anomaly at a phase transition temperature T_c but continue to exist well into the paramagnetic phase as propagating excitations. The phenomenon is most marked in systems of low dimensionality,[15,16] where all but the very long-wavelength spin waves are essentially temperature independent up to tem-

peratures in excess of T_c, although the zone-center (zero-wave-vector) magnons scale with temperature closely like the magnetization (or sublattice magnetization for antiferromagnets) and have energies going to zero at T_c.[17]

It seems clear that any approximation hoping to describe this situation realistically, particularly for systems of low dimensionality, must at the very least introduce some wave-vector dependence into the temperature variation of the associated dispersion relationship. It is the purpose of the present paper to demonstrate, within the Green's-function framework, a rather general method for doing this; a method which effectively bypasses the necessity for devising new and even more mystical recipes for decoupling three-spin from two-spin Green's functions and enables approximations of obvious physical significance to be made directly in terms of the excitation spectrum for magnons.

The theory is developed in detail for the simplest nontrivial approximation and applied to the topical problem concerning the possible existence of a phase transition for two-dimensional Heisenberg systems.[10,18–22] We find that a phase with zero magnetization and infinite susceptibility does exist for the isotropic Heisenberg ferromagnet. We are also able to investigate the reaction of this new phase to the introduction of anisotropy and to answer, within the present approximation, the question of whether two phase transitions (one to a zero-magnetization infinite-susceptibility phase, and a second to a spontaneously magnetized phase) can exist in systems with finite but arbitrarily small anisotropy. The question arises because the ground state of the isotropic two-dimensional Heisenberg system has a long-range order which is destroyed by thermal energy at any nonzero temperature, formally at least pointing to $T = 0\,^{\circ}\mathrm{K}$ as a second phase transition temperature for the isotropic limit.

II. GREEN'S-FUNCTION APPROXIMATION

The application of double-time temperature-dependent Green's functions to the statistical problems in magnetism has been discussed in great detail many times before, and we may refer the reader to the literature for background.[1–10,23,24]

The retarded Green's function for Heisenberg operators A and B is defined as

$$\langle\langle A(t); B(t')\rangle\rangle \equiv -i\theta(t-t')\langle [A(t), B(t')]\rangle , \quad (2.1)$$

in which square brackets denote a commutator, angular brackets denote ensemble averages, and $\theta(x)$ is the step function (equal to 1 for $x > 0$, 0 for $x < 0$). We have adopted units for which Planck constant $h = 2\pi$. The equation of motion of the Green's function so defined is

$$i\frac{\partial}{\partial t}\langle\langle A(t); B(t')\rangle\rangle = \delta(t-t')\langle [A(t), B(t')]\rangle + \langle\langle [A(t), \mathcal{H}]; B(t')\rangle\rangle , \quad (2.2)$$

where $\mathcal{H}$ is the system Hamiltonian – the final term being, in general, a Green's function of higher order. If (2.2) can be linearized by a suitable decoupling approximation, the original Green's function can be evaluated and correlation functions $\langle B(t')A(t)\rangle$ follow from the formula

$$\langle B(t')A(t)\rangle = \lim_{\epsilon\to+0} i\int_{-\infty}^{\infty} \frac{[\langle\langle A; B\rangle\rangle_{E+i\epsilon} - \langle\langle A; B\rangle\rangle_{E-i\epsilon}]}{e^{E/kT}-1} \times e^{-iE(t-t')}\,dE , \quad (2.3)$$

where $\langle\langle A; B\rangle\rangle_E$ is the Fourier transform of $\langle\langle A(t); B(t')\rangle\rangle$ with respect to $t - t'$.

To demonstrate the principle behind the present method, let us consider initially a simple Heisenberg Hamiltonian for an isotropic ferromagnetic system, of the form

$$\mathcal{H} = \sum_{\langle i,j\rangle} -2J_{ij}\vec{S}_i\cdot\vec{S}_j , \quad (2.4)$$

where the summation runs over all pairs of neighbors i and j interacting via an exchange J_{ij}. The equation of motion for a two-time Green's function $\langle\langle S_g^+; (S_h^z)^n S_h^-\rangle\rangle_E$ then follows quite formally from (2.2) and is

$$E\langle\langle S_g^+; (S_h^z)^n S_h^-\rangle\rangle_E = \frac{F}{2\pi}\delta_{gh} - \sum_{j-g} 2J_{jg}\langle\langle (S_g^z S_j^+ - S_g^+ S_j^z); (S_h^z)^n S_h^-\rangle\rangle_E , \quad (2.5)$$

where n is a positive integer, $S^{\pm} = S^x \pm iS^y$, and

$$F = \langle [S_h^+, (S_h^z)^n S_h^-]\rangle . \quad (2.6)$$

Making use of the translational invariance of the system with respect to $g - h$, we may define Fourier transforms

$$G_{\vec{K}}(E) = \sum_{g-h}\langle\langle S_g^+; (S_h^z)^n S_h^-\rangle\rangle_E e^{-i\vec{K}\cdot(\vec{g}-\vec{h})} , \quad (2.7)$$

$$\Delta_{\vec{K}}(E) = -\sum_{g-h}\Big\{\sum_{j-g} 2J_{jg}\langle\langle (S_g^z S_j^+ - S_g^+ S_j^z); (S_h^z)^n S_h^-\rangle\rangle_E\Big\} \times e^{-i\vec{K}\cdot(\vec{g}-\vec{h})} , \quad (2.8)$$

in terms of which Eq. (2.5) becomes

$$EG_{\vec{K}}(E) = F/2\pi + \Delta_{\vec{K}}(E) . \quad (2.9)$$

The problem of linearization and the development of a first-order Green's-function approximation therefore consists of finding a relationship

$$\Delta_{\vec{K}}(E) = M_{\vec{K}}(E)G_{\vec{K}}(E) + D_{\vec{K}}(E) , \quad (2.10)$$

in terms of which Eq. (2.5) is formally solvable for $G_{\vec{K}}(E)$ as follows:

$$G_{\vec{K}}(E) = \frac{F/2\pi + D_{\vec{K}}(E)}{E - M_{\vec{K}}(E)} . \quad (2.11)$$

In all the previously cited decoupling approximations decoupling is performed in real space by relating $\langle\langle S_g^z S_j^+; (S_h^z)^n S_h^-\rangle\rangle_E$ and $\langle\langle S_g^+ S_j^z; (S_h^z)^n S_h^-\rangle\rangle_E$ directly to $\langle\langle S_g^+; (S_h^z)^n S_h^-\rangle\rangle_E$, although often in quite an involved way which may not always be physically transparent. The result, in general (the exception being Tahir-Kheli's work[6]), is a more modest decoupling of form

$$\Delta_{\vec{K}}(E) = M_{\vec{K}} G_{\vec{K}}(E) , \tag{2.12}$$

where $M_{\vec{K}}$ is real, independent of E, and related to simple (unrenormalized) spin-wave energy $E_0'(\vec{K})$ by

$$M_{\vec{K}} = \xi(T) E_0'(\vec{K}) , \tag{2.13}$$

in which $\xi(T)$ is a dimensionless temperature-dependent *but wave-vector-independent* function which differs from one decoupling scheme to another. Thus, for example, with Hamiltonian (2.4) and the simplest random-phase decoupling approximation,[2,3] we have

$$E_0'(\vec{K}) \equiv S E_0(\vec{K}) = S \sum_{j-g} 2J_{jg}[1 - e^{i\vec{K}\cdot(\vec{j}-\vec{g})}], \tag{2.14}$$

where S is the spin quantum number and $\xi(T) = \langle S^z\rangle/S$. The more sophisticated decoupling schemes include spin-pair correlations in $\xi(T)$, but are often tractable only for the nearest-neighbor (nn) exchange restriction.

In view of the fact that $G_{\vec{K}}(E)$ from Eq. (2.11) has poles at $E = M_{\vec{K}}(E)$, we recognize these poles to be the elementary excitations (i.e., magnons) of the system. The approximations (2.12) and (2.13) can now be seen to involve three basic assumptions as follows:

(i) $\Delta_{\vec{K}}(E)$ and $G_{\vec{K}}(E)$ can be related through a "mass operator" equation $\Delta_{\vec{K}}(E) = M_{\vec{K}}(E) G_{\vec{K}}(E)$, where $M_{\vec{K}}(E)$ does not involve terms proportional to the inverse of $G_{\vec{K}}(E)$. This assumption, which amounts to putting $D_{\vec{K}}(E)$ of Eq. (2.10) equal to zero, is valid for normal boson or fermion interacting particle systems but not, in general, for spin systems, where the more complex spin kinematics give rise to a mass operator of anomalous form [with $D_{\vec{K}}(E) \neq 0$]. The associated restriction is essentially to high spin quantum number; a more detailed analysis of the approximation involved is available in the literature.[6]

(ii) $[E - M_{\vec{K}}(E)]^{-1}$ has a simple pole on the real axis, say, at $E = M_{\vec{K}}$. This amounts to a neglect of magnon lifetime effects; an approximation which profoundly affects the spin dynamics but which is nearly always assumed for calculation of equilibrium properties.

(iii) $M_{\vec{K}}$ can be related to low-temperature magnon dispersion by a simple proportionality, i.e., Eq. (2.13).

It is only restriction (iii), implying that all magnons renormalize in the same way with temperature, which will be removed in this paper. Thus, we shall define a wave-vector-dependent renormalization $\xi_{\vec{K}}(T)$, and, whatever criterion is used to ascertain a physically reasonable form for it, it may be incorporated directly into (2.11) to give

$$G_{\vec{K}}(E) = \frac{F/2\pi}{E - \xi_{\vec{K}}(T) E_0'(\vec{K})} , \tag{2.15}$$

where we have put $D_{\vec{K}}(E) = 0$. Use of (2.3) and (2.7) now lead to findings for spin correlations as follows:

$$\langle (S_h^z)^n S_h^- S_g^+\rangle = \frac{F}{N} \sum_{\vec{K}} \frac{e^{i\vec{K}\cdot(\vec{g}-\vec{h})}}{e^{\xi_{\vec{K}}(T) E_0'(\vec{K})/kT} - 1} , \tag{2.16}$$

where k is Boltzmann's constant, N is the number of spins in the lattice, and the summation runs over the N allowed values in the first Brillouin zone of the reciprocal lattice.

By the now quite standard procedures of first-order Green's-function theory,[5,25] equations for magnetization $\bar{S} \equiv \langle S^z\rangle$ and transverse two-spin correlations follow directly from (2.16) in the form

$$\langle S_h^- S_g^+\rangle = (\bar{S}/N) \textstyle\sum_{\vec{K}} \cos[\vec{K}\cdot(\vec{g}-\vec{h})] \times \{\coth[\xi_{\vec{K}}(T) E_0'(\vec{K})/2kT] - 1\}, \tag{2.17}$$

$$\frac{2\bar{S}+x}{2S+1} = \frac{(x+1)^{2S+1} + (x-1)^{2S+1}}{(x+1)^{2S+1} - (x-1)^{2S+1}} , \tag{2.18}$$

where

$$x = (1/N) \textstyle\sum_{\vec{K}} \coth[\xi_{\vec{K}}(T) E_0'(\vec{K})/2kT] . \tag{2.19}$$

Equation (2.18) can also be written more succinctly as

$$\bar{S}/S = B_S \times 2S \coth^{-1} x , \tag{2.20}$$

where B_S is the Brillouin function for spin S.

To determine the problem it now merely remains to choose $\xi_K(T)$. At low temperatures, the evidence from spin-wave theories[26] points to a wave-vector-independent renormalization proportional to $\langle \vec{S}_0 \cdot \vec{S}_a\rangle^{1/2}$, where a is a nn distance. In spite of this, as remarked in the Introduction, it is now experimentally well established that outside the low-temperature domain the dependence of spin-wave energies on temperature can be very sensitive, indeed, to wavelength.[11–17]

It is helpful to appeal to a physical picture. Consider a system with well-developed short-range order. Even above the Curie temperature there is, in general, a finite-temperature-dependent volume V and associated length $L \sim V^{1/d}$ (where d is dimensionality) over which spins are closely correlated in some sense. Let us refer to L as a coherence length. Magnons with wavelength $\lambda \gg L$ have a frequency small compared to the reciprocal relaxation time of the background disorder and can therefore be described to a first approximation as displace-

ments from the time-averaged background. This leads to renormalization of magnon energy according to magnetization (i.e., as $\bar{S}$). Clearly, however, such an approximation is quite inappropriate for short-wavelength spin waves with $\lambda \ll L$. The latter have a frequency large compared to the background reciprocal relaxation time and can therefore, to a first approximation, be described as displacements from the instantaneous background. To use an analogy first introduced by Keffer and Loudon,[27] the additional magnon excitations are like ripples superimposed on the instantaneous nonequilibrium position of the existing waves. For nn exchange effects, this concept leads[26,27] to a magnon energy renormalization as $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$, where S_0 and S_a are nn, and is equivalent to describing spin-wave motion as small deviations from the ordered spin array in which each spin S_i is given a magnitude $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$.

In a less phenomenological vein it is useful to relate the above to the more formal Green's-function decoupling language of Callen.[5] If the Green's function $\langle\langle S_g^- S_g^+ S_f^+; B \rangle\rangle$ is decoupled in the symmetric form

$$\langle\langle S_g^- S_g^+ S_f^+; B \rangle\rangle \underset{g \neq f}{\rightarrow} \langle S_g^- S_g^+ \rangle \langle\langle S_f^+; B \rangle\rangle + \langle S_g^- S_f^+ \rangle \langle\langle S_g^+; B \rangle\rangle , \quad (2.21a)$$

and similarly for $\langle\langle S_g^+ S_g^- S_f^+; B \rangle\rangle$, then the two renormalization schemes $\xi_{\vec{K}}(T) = \bar{S}/S$ and $\xi_{\vec{K}}(T) = \langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}/S$ of the previous paragraph follow, respectively, by relating transverse spin operators S_g^+ and S_g^- to S_g^z by use of the identities

$$S_g^z = \tfrac{1}{2}(S_g^+ S_g^- - S_g^- S_g^+) , \quad (2.21b)$$

$$S_g^z = S(S+1) - (S_g^z)^2 - S_g^- S_g^+ , \quad (2.21c)$$

but (for $S > \frac{1}{2}$) neglecting the fluctuations in $(S_g^z)^2$ in (2.21c).

Decoupling on the basis of identity (2.21b) treats approximately the deviations of S_g^z from zero, whereas decoupling on the basis of identity (2.21c) treats approximately quantities which are small when $S_g^z \rightarrow +S$. Callen uses the former when $\langle S^z \rangle$ is small, and the latter when $S - \langle S^z \rangle$ is small, to give, in each case, a wave-vector-independent renormalization. The earlier physical arguments suggest that a more appropriate criterion should relate to the respective magnitudes of magnon frequency and reciprocal relaxation time of the nonequilibrium instantaneous background. For the Heisenberg ferromagnet, in view of the simple character of the magnon dispersion (with excitation energy increasing monotonically with wave-vector magnitude), this relaxation time is directly correlated to the short-range order allowing us to introduce the concept of coherence length L.

Thus, we find the renormalization $\xi_{\vec{K}}(T) = \bar{S}/S$ when wavelength $\lambda \gg L$ and $\xi_{\vec{K}}(T) = \langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}/S$ when $\lambda \ll L$. Clearly, for intermediate wavelengths the renormalization function should vary smoothly between these extremes. Kuramoto[28] has argued in favor of a particular form which (he claims) shows a general accord with the existing concepts of dynamic scaling.[29] Our immediate concern, however, is tractability and, for computational ease, we have decided initially to adopt the simplest conceivable approximation in accord with the limits above. We shall renormalize magnons in one of only two ways; either as $\bar{S}/S$ (for long-wavelength magnons with $K < \pi/L = K_C$) or as $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}/S$ (for short wavelength magnons with $K > \pi/L = K_C$). Thus, we expect L to define some sort of coherence length[30] for which magnons with $K > \pi/L$ experience much less damping and renormalization than do magnons with $K < \pi/L$.

We take care to distinguish L from the correlation length L_{corr} since we shall later show them to be quite distinct. In three dimensions L remains microscopic in the critical region, while in two dimensions (for the isotropic system) it becomes macroscopic, but proportional to L_{corr}, thereby not violating the main principle of static scaling theory.[31]

Qualitatively we shall associate the coherence length L with the spin separation for which the correlation $\langle \vec{S}_0 \cdot \vec{S}_L \rangle$ falls to some fixed fraction of its near neighbor value. However, we require the cutoff wave vector $K_C = \pi/L$ to be a continuous variable (approaching the farthest reaches of the first Brillouin zone at extremes of high temperature). Correlation $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$, on the other hand, as evaluated in a self-consistent fashion from the theory, only has physical significance at the actual spin sites. We wish to extend the concept of spin correlation to interpolate smoothly between the physically significant R values.

A function which is fairly adequate in this sense is the self-consistent Green's-function result (2.17) itself, evaluated in the first Brillouin zone as a continuous function of $R = |g - h|$. It is not ideal[31a] because of a developing oscillatory behavior at high temperatures. We note from (2.14) and (2.17) that these correlations are spherically symmetric (in a cubic lattice) or circularly symmetric (square lattice) for large R. For arbitrary R the deviations from spherical or circular symmetry are small and will, for mathematical simplicity, be neglected. We assume, therefore, a spherical cutoff surface in three dimensions with $R = L$, leading to a spherical cutoff surface $K = K_C = \pi/L$ in reciprocal space. The analogous two-dimensional situation is obvious.

Quantitatively we define L via the equation

$$\langle \vec{S}_0 \cdot \vec{S}_L \rangle = \Phi \langle \vec{S}_0 \cdot \vec{S}_{\eta a} \rangle , \quad (2.22)$$

where Φ is a numerical constant to be determined,

and where the right-hand side contains a lattice-dependent parameter η which can be determined from the condition that the theory must go over smoothly to the random-phase (RPA) Green's-function approximation in the high-temperature limit – i.e., K_C goes to a first-zone corner as $T \to \infty$. For a smooth monotonic generalized spin correlation, $\langle \vec{S}_0 \cdot \vec{S}_R \rangle \to 0$ for arbitrary nonzero R as $T \to \infty$. By inspection of (2.22) it then follows that $\eta = 1/d^{1/2}$ (where d is dimensionality) for the linear chain, quadratic-layer, and simple-cubic lattices. For correlations (2.17), this simple form is not exact and η becomes Φ dependent. However, this Φ dependence is small for the lattice of direct concern below (quadratic layer) and $\eta = 1/d^{1/2}$ is quite adequate in the present context. At low temperatures we shall find that $L \to \infty$, for which case all magnons renormalized as $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$ and we recover the essence of interacting spin-wave theory.

The theory is now determined except for the single adjustable parameter Φ which will be chosen to give the best agreement between the results of the theory and those of high-temperature series expansions. For the single lattice which we have computed in detail (the quadratic-layer or square lattice) we find $\Phi = 0.74$ for the ferromagnetic Heisenberg system. For this two-dimensional case we find a phase transition for the isotropic system at a finite temperature $T_C^{(2)}$ supporting the series expansion findings of Stanley and Kaplan.[19] We note in passing that the present method is not open to the same criticism[32] leveled against the other closed-form demonstrations of this effect.[10,33]

Our final operating equations may now be summarized as

$$\langle S_0^- S_R^+ \rangle = \frac{\bar{S}}{N} \Bigg\{ \sum_{K<K_C} \cos(\vec{K}\cdot\vec{R}) \left[\coth\left(\frac{\bar{S} E_0(\vec{K})}{2kT} \right) - 1 \right] + \sum_{K>K_C} \cos(\vec{K}\cdot\vec{R}) \left[\coth\left(\frac{\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2} E_0(\vec{K})}{2kT} \right) - 1 \right] \Bigg\} , \tag{2.23}$$

where $K_C = \pi/L$, $K \equiv |\vec{K}|$,

$$\langle \vec{S}_0 \cdot \vec{S}_L \rangle / \langle \vec{S}_0 \cdot \vec{S}_{a/\sqrt{d}} \rangle = \Phi , \tag{2.24}$$

and

$$\bar{S}/S = B_S [2S \coth^{-1} x] , \tag{2.25}$$

where

$$x = \frac{1}{N} \left[\sum_{K<K_C} \coth\left(\frac{\bar{S} E_0(\vec{K})}{2kT} \right) + \sum_{K>K_C} \coth\left(\frac{\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2} E_0(\vec{K})}{2kT} \right) \right] . \tag{2.26}$$

The equations simplify very considerably for a paramagnetic phase and, as we shall see, the detailed form of the short-wavelength renormalization is not important for static paramagnetic calculations. Consider, for example, the situation $\bar{S} \to 0$ in the absence of an applied field, i.e., $T \to T_C$. In such a limit, long-wavelength magnons ($K < K_c$) have energies which go to zero while short-wavelength magnons have energies which (regardless of detailed form) remain finite. It follows that, in this limit, (2.23) simplifies to

$$\langle S_0^- S_R^+ \rangle = \frac{1}{N} \sum_{K<K_C} 2kT_C \frac{\cos(\vec{K}\cdot\vec{R})}{E_0(\vec{K})} , \tag{2.27}$$

which for $R \to 0$ becomes

$$\frac{S(S+1)}{3kT_C} = \frac{1}{N} \sum_{K<K_C} \left(\frac{1}{E_0(\vec{K})} \right) , \tag{2.28}$$

where we have made the isotropy assumption $\langle (S^z)^2 \rangle = \langle (S^x)^2 \rangle = \langle (S^y)^2 \rangle = \frac{1}{3} S(S+1)$. This equation for Curie temperature, valid for any short-wavelength magnon renormalization which allows for $K > K_C$ paramagnetic propagating magnons, immediately reveals to us the qualitative nature of the coherence length near the Curie temperature. Thus, for three-dimensional systems, Eq. (2.28) predicts a noninfinite Curie temperature only if the coherence length at T_C is finite. For two-dimensional isotropic Heisenberg systems, however, it predicts $T_C = 0$ (the RPA result[20]) if coherence length $L (= \pi/K_C)$ remains finite at T_C but, possibly, a finite T_C if L diverges in this limit. The actual situation is determined unequivocally, of course, by the simultaneous solution of (2.23) and (2.24). This solution, which is obtained numerically below for the two-dimensional case, reveals that L *does* diverge (and that T_C *is* therefore finite) for all values of parameter $\Phi < 1$, $\Phi = 1$ being the RPA limit.

It seems evident that the greatest improvement of the present theory over the existing first-order Green's function and renormalized spin-wave approximations will result for systems in which paramagnetic short-range order effects are most pronounced, namely, spin lattices of low dimensionality. Such systems will support paramagnetic magnons with long lifetimes over most of the first Brillouin zone up to comparatively elevated temperatures,[15] and these effects are neglected entirely in all the earlier Green's-function approximations.

For this reason we have decided to compute the theory in detail for the quadratic-layer (square) lattice. Discussions concerning two-dimensional magnetic systems have flourished in the literature of late.[10,15–22] Most experimental information concerns the quadratic-layer lattice. The question of a possible phase transition for the isotropic Heisenberg ferromagnet and antiferromagnet is of prime academic interest. On the other hand, the reaction

of such a system to the introduction of small anisotropy and the stabilization of a truly ordered phase for this case is of equal importance to experimental understanding. Both questions will be discussed at length.

III. TWO-DIMENSIONAL QUASI-HEISENBERG FERROMAGNET

We now consider a Hamiltonian

$$\mathcal{H} = \sum_{nn} -2J[S_i^x S_j^x + S_i^y S_j^y + (1+\Delta) S_i^z S_j^z] - h\sum_i S_i^z , \tag{3.1}$$

where (in an obvious notation) we restrict exchange interactions to nearest neighbors for computational convenience, but where we allow for the presence of small axial anisotropy Δ and also introduce an applied-magnetic-field energy $h = g\mu_B H$, where H is an external field applied in direction z.

With this Hamiltonian the theoretical development of Sec. II remains valid in its entirety but with spin-wave energy $M_{\vec{K}}$ of (2.13) replaced by $h + M_{\vec{K}}$ and where $M_{\vec{K}} = \xi_{\vec{K}}(T) E_0'(\vec{K})$, for the specific case of a quadratic-layer lattice with Hamiltonian (3.1), is

$$M_{\vec{K}} = \xi_{\vec{K}}(T) 4JS[2(1+\Delta) - \cos K_x a - \cos K_y a] , \tag{3.2}$$

where K_x and K_y are components of the wave vector $\vec{K}$ along the square-lattice axes, i.e., z is normal to the plane. In particular, Eqs. (2.23)–(2.26) remain valid if we put

$$E_0(\vec{K}) = 4J[2(1+\Delta) - \cos(K_x a) - \cos(K_y a)] + (h/\bar{S}) , \qquad K < K_C \tag{3.3}$$

and

$$E_0(\vec{K}) = 4J[2(1+\Delta) - \cos(K_x a) - \cos(K_y a)] + h/\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}, \quad K > K_C . \tag{3.4}$$

Let us first look at the paramagnetic phase in the limit $h \to 0$. For this case we know physically that $\bar{S} \to 0$ but that nn correlation $\langle \vec{S}_0 \cdot \vec{S}_a \rangle$ goes to zero only in the limit of infinite temperature. It follows that (2.23) simplifies to

$$\langle S_0^- S_R^+ \rangle = 2kT \frac{1}{N} \sum_{K<K_C} \frac{\cos\vec{K}\cdot\vec{R}}{E_0(\vec{K})}, \tag{3.5}$$

with $E_0(\vec{K})$ given by (3.3). For the case $R = 0$, Eq. (3.5) becomes

$$S(S+1) - \langle (S^z)^2 \rangle = 2kT \frac{1}{N} \sum_{K<K_C} \frac{1}{E_0(\vec{K})} . \tag{3.6}$$

For the limit $\bar{S} \to 0$, $h \to 0$, we find $\langle (S^z)^2 \rangle = \frac{1}{3} S(S+1)$ directly from the $n = 1$ form of Eq. (2.16). This result is rather dependent upon the form in which anisotropy is included; it is not valid, in the Green's-function approximation, for single-spin crystal-field anisotropy, for example.[34] Noting that $\bar{S}/h = \chi/Ng^2\mu_B^2$, where χ is magnetic susceptibility, we define a reduced susceptibility $\chi_0 = \bar{S}/h$ in terms of which (3.6) becomes

$$\frac{4JS(S+1)}{3kT} = \frac{1}{N} \sum_{K<K_C} [2(1+\Delta) - \cos(K_x a) - \cos(K_y a) + (4J\chi_0)^{-1}]^{-1} . \tag{3.7}$$

Restricting our investigations to quasi-Heisenberg systems ($\Delta \ll 1$) and, in particular, to the isotropic limit itself, we neglect any small anisotropy of $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ in the paramagnetic phase to write (3.5) as

$$\frac{4J\langle \vec{S}_0 \cdot \vec{S}_R \rangle}{3kT} = \frac{1}{N} \sum_{K<K_C} \frac{\cos(\vec{K}\cdot\vec{R})}{2(1+\Delta) - \cos(K_x a) - \cos(K_y a) + (4J\chi_0)^{-1}} . \tag{3.8}$$

Defining the coherence length $L = \pi/K_C$, we can now express Eq. (2.24) as

$$\sum_{K<\pi/L} \frac{\cos(K_x L)}{2(1+\Delta) - \cos(K_x a) - \cos(K_y a) + (4J\chi_0)^{-1}} = \Phi \times \sum_{K<\pi/L} \frac{\cos(K_x a/\sqrt{2})}{2(1+\Delta) - \cos(K_x a) - \cos(K_y a) + (4J\chi_0)^{-1}} , \tag{3.9}$$

where, we recall, $K = \pi/L$ defines a spherical cutoff condition within the first Brillouin zone. Together, Eqs. (3.7) and (3.9) relate magnetic susceptibility χ_0 and coherence length L to temperature T, for arbitrary values of the adjustable parameter Φ. We note, in particular, that the susceptibility appears only in conjunction with anisotropy parameter Δ in the form

$$D = 2\Delta + (4J\chi_0)^{-1} . \tag{3.10}$$

Before solving these equations numerically by computer, there are one or two useful qualitative observations which can be made. First, for $L = a/\sqrt{2}$, Eq. (3.9) has the general solution $\Phi = 1$. For this case, all spin waves in the first Brillouin zone have $K < K_C$ and therefore renormalize like the magnetization. In other words, $\Phi = 1$ is just the RPA Green's-function approximation.

Also, consider the limit of infinite susceptibility for the isotropic case, viz., $D \to 0$. We shall find below that the ratio L/a ($= Q$, say) becomes large for small D in two-dimensional systems. Defining $\theta_\alpha \equiv K_\alpha L$ ($\alpha = x, y$), Eq. (3.9) then becomes, for an infinite system

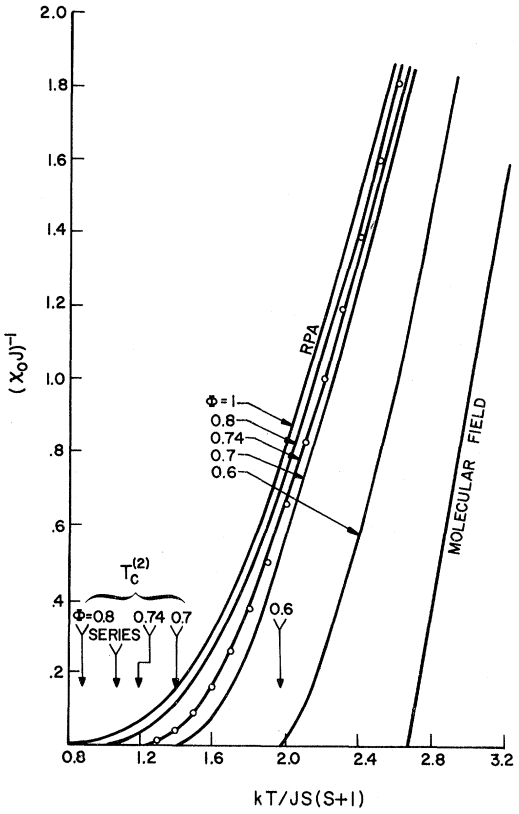

FIG. 1. Temperature dependence of magnetic susceptibility for the isotropic quadratic-layer lattice as calculated from the theory of the present paper for different values of parameter Φ [Eq. (2.24)]. The open circles are the high-temperature series result for classical spins. Also shown is the molecular-field approximation and (by arrows) the limiting values of temperature $T_C^{(2)}$ for which the different curves indicate a divergent susceptibility. Note that the $\Phi=1$ curve is the RPA Green's function with $T_C^{(2)}=0$.

$$\iint_{\substack{\text{circle}\\ \text{radius }\theta=\pi}} \frac{\cos\theta_x}{DQ^2+\frac{1}{2}\theta^2} = \Phi \times \iint_{\substack{\text{circle}\\ \text{radius }\theta=\pi}} \frac{1-(\theta_x^2/2Q^2)}{DQ^2+\frac{1}{2}\theta^2}\,, \tag{3.11}$$

where $\theta^2=\theta_x^2+\theta_y^2$. Using (3.7) in this same limit, i.e.,

$$\frac{4JS(S+1)}{3kT_C^{(2)}} = \frac{1}{4\pi^2} \times \iint_{\substack{\text{circle}\\ \text{radius }\theta=\pi}} \frac{1}{DQ^2+\frac{1}{2}\theta^2}\,, \tag{3.12}$$

we find (to lowest order in $1/Q$) on substituting into (3.11)

$$\frac{4JS(S+1)\Phi}{3kT_C^{(2)}} = \frac{1}{4\pi^2} \times \iint_{\substack{\text{circle}\\ \text{radius }\theta=\pi}} \frac{\cos\theta_x}{DQ^2+\frac{1}{2}\theta^2}\,, \tag{3.13}$$

where we have written $T_C^{(2)}$ for the temperature at which the susceptibility diverges for the isotropic quadratic-layer system. We see that the role played by DQ^2 is all important. If $DQ^2\to 0$ as $D\to 0$ it follows from (3.11) that $\Phi=1$ and from (3.13) that $T_C^{(2)}=0$. This is just RPA Green's-function theory with its, by now, familiar prediction of no susceptibility divergence at finite temperatures in two dimensions. However, for all nonzero limits of DQ^2 we find $\Phi<1$ and $T_C^{(2)}>0$. In other words, *any* value of Φ less than unity precipitates a nonzero "Stanley-Kaplan" temperature $T_C^{(2)}$.[19] Also, since DQ^2 remains finite for any $\Phi<1$ as $T_C^{(2)}$ is approached from above ($D\to 0$), the coherence length $L=Qa$ must diverge as the square root of the susceptibility. From (3.10), we see that this is true only for the isotropic system. When small anisotropy Δ is present, D remains finite as the susceptibility diverges and the coherence length does not diverge at the phase transition temperature. The actual limiting value of DQ^2 will, of course, depend on Φ and it is possible to consider the situation for $DQ^2\to\infty$, which would allow a diverging coherence length for the anisotropic case. We find that this situation requires $\Phi=0.18$ [from (3.11)] and $T_C^{(2)}\to\infty$ [from (3.13)]; a rather unphysical situation (no paramagnetic phase).

Of all the possible values of our single adjustable parameter $\Phi(0.18<\Phi<1)$ we shall now choose the one most closely approximating physical reality by numerically solving (3.7) and (3.9) simultaneously, plotting a family of susceptibility-vs-temperature curves with Φ as parameter, and then fitting to the best high-temperature series-expansion results available. The computer solutions are plotted in Figs. 1 and 2. In Fig. 3 we show $T_C^{(2)}$ as a function of Φ. The series-expansion results are calculated as described in the Appendix and Fig. 4. They are also displayed in Figs. 1 and 2.

From Fig. 1 we observe that there is quite close agreement between the present theory and the high-temperature series results for values of $\Phi\approx 0.74$, although the sensitivity of the Green's-function curves to the parameter Φ becomes quite acute as Φ decreases. For $\Phi=0.74$ the theories differ seriously only for values of the temperature very close to $T_C^{(2)}$, e.g., for values of $\chi_0 J>\sim 100$ (Fig. 2). There is agreement to within a few percent for temperatures down to $\tau\approx 1.27$ $[\tau=kT/JS(S+1)]$ but for lower temperatures the Green's-function susceptibility diverges more rapidly to give $kT_C^{(2)}\approx 1.19JS(S+1)$ compared to the series result $kT_C^{(2)}\approx 1.07JS(S+1)$. To put other theoretical approaches into perspective, molecular-field theory predicts a divergent susceptibility for $kT=(8/3)JS(S+1)$ while the RPA Green's-function theory, at the temperature $\tau=1.27$ for which the $\Phi=0.74$ and series-expansion findings are still closely in agreement, is already in error by *more than a factor of* 10. Also, at $\tau=1.27$, more than 50 terms in the series expansion are required

for quantitative estimate of susceptibility. We must therefore emphasize that the "exact series" results at and below this temperature range are far from exact, depending on the extrapolation procedure[35] quite sensitively. They do, however, represent the best approximation available and the discrepancy between $\Phi = 0.74$ and the series results below $\tau \approx 1.27$ is probably real since we have taken, if anything, a high limit for $T_C^{(2)}/T_C^{mf}$ in the series.[35]

The computed results of theory in Figs. 1–3 have so far been discussed only with reference to the isotropic-layer system ($\Delta = 0$), primarily because the high-temperature series coefficients are known only for that case. However, from (3.10), we note that the $\Phi = 0.74$ results in Figs. 1 and 2 can be interpreted more widely. They are also valid for weakly anisotropic systems if $(\chi_0 J)^{-1}$ in the ordinates is replaced by $(\chi_0 J)^{-1} + 8\Delta$. This allows us immediately to plot the susceptibility-vs-temperature (paramagnetic) curves for the anisotropic cases, noting that

$$(\chi_0 J)^{-1}_{\text{anis}} = (\chi_0 J)^{-1}_{\text{iso}} - 8\Delta \,. \tag{3.14}$$

In particular we can calculate $T_C^{(2)}$ as a function of anisotropy. The result is shown in Fig. 5. Taking

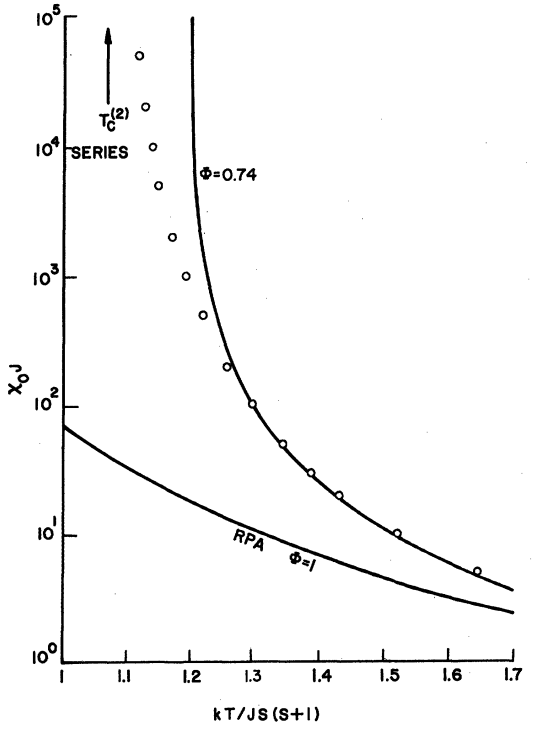

FIG. 2. Expanded scale plot of Fig. 1 for temperatures close to the high-temperature series $T_C^{(2)}$ comparing the "series" susceptibility with that of the "best-fit" Green's-function curve $\Phi = 0.74$. Also shown for comparison is the RPA Green's-function susceptibility in this same restricted temperature range.

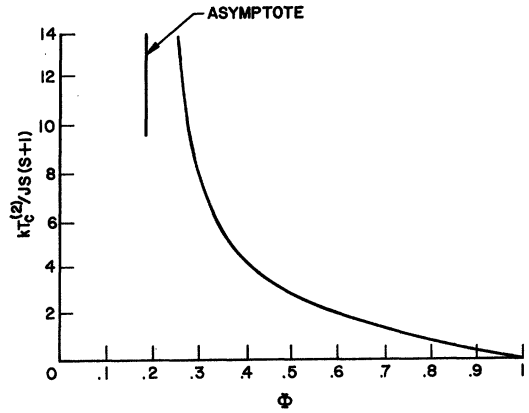

FIG. 3. Variation of $T_C^{(2)}$ with Green's-function parameter Φ (see text) calculated for the isotropic Heisenberg quadratic-layer lattice using the theory of the present paper. Note that $T_C^{(2)}$ goes to zero in the RPA ($\Phi = 1$) Green's-function limit and that $T_C^{(2)}$ is infinite for $\Phi < 0.18$ (no paramagnetic phase).

the Green's-function finding (3.14) and assuming it valid for the series-expansion susceptibility, we may also calculate $T_C^{(2)}$ as a function of Δ for the series approximation. This together with RPA Green's-function and molecular-field estimates are all shown in Fig. 5. The $\Phi = 0.74$ and series-expansion results differ only for systems with $\Delta \lesssim 5 \times 10^{-4}$, and even then by only a few percent.

A word about symbolism is necessary at this juncture. We shall refer to the temperature for which paramagnetic susceptibility diverges as $T_C^{(2)}$, and the temperature for which spontaneous mag-

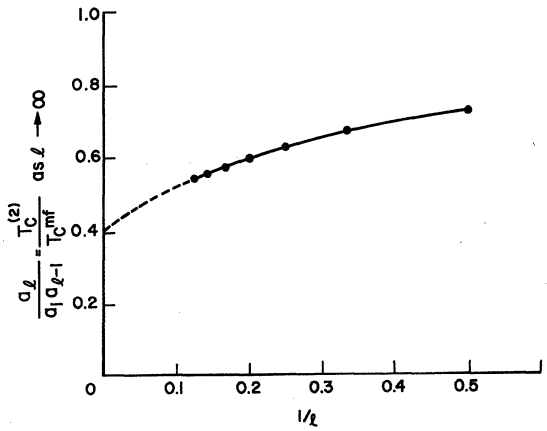

FIG. 4. Ratio of coefficients $a_l/a_1 a_{l-1}$ from high-temperature susceptibility series (A1) are plotted as a function of $1/l$ and extrapolated graphically (see Appendix) to $1/l = 0$, for which limit the ordinate equals the ratio of $T_C^{(2)}$ to the molecular-field Curie temperature T_C^{mf}. The filled circles are the exact-series coefficients for the classical spin isotropic quadratic-layer lattice, taken from Ref. 35.

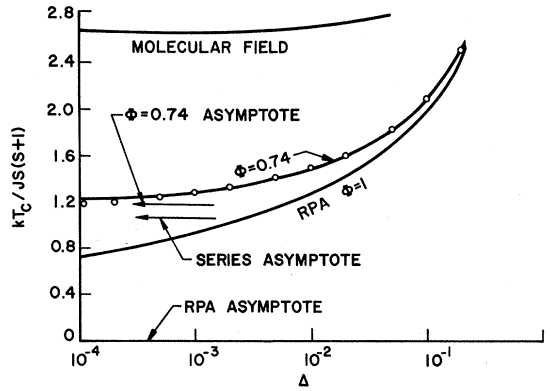

FIG. 5. Curie temperature T_C as a function of anisotropy Δ (the ratio of anisotropy field to exchange field) calculated for the weakly anisotropic quadratic-layer lattice by Green's-function theory (RPA and $\Phi = 0.74$). Also shown is the molecular-field curve and (open circles) the result of a calculation utilizing the high-temperature series susceptibility (see text). The isotropic limits $T_C \to T_C^{(2)}$ are indicated by arrows.

netization goes to zero as $T_C^{(1)}$. For three-dimensional systems, all indications are that $T_C^{(2)} = T_C^{(1)}$, so that we may always refer to a single phase transition temperature and call it the Curie temperature T_C. For isotropic Heisenberg systems in two dimensions, $T_C^{(1)} = 0$, a rigorous result,[18] while indications are that $T_C^{(2)}$ may well be nonzero. For real systems, anisotropy Δ is necessarily present and we shall be concerned with the question "Are there two phase transition temperatures $T_C^{(1)} \neq T_C^{(2)}$ for weakly anisotropic systems?" That is, for real systems, is it conceivable to have three separate phases: a ferromagnetic one, a paramagnetic one, and a zero-magnetization infinite-susceptibility intermediate one? A similar question can be asked for the antiferromagnetic analog.

For the ferromagnetic case already discussed, the answer within the present Green's-function approximation is readily deduced from a comparison of (3.7) and (3.9) in the limit $\chi_0^{-1} \to 0$ on the one hand (which measures $T_C^{(2)}$) and (2.24), (2.27), and (2.28) for the case $h = 0$ (which measures $T_C^{(1)}$) on the other. It is clear that for any nonzero value of Δ, the sets of equations are identical, i.e., $T_C^{(1)} = T_C^{(2)}$. For the isotropic case the equation for $T_C^{(1)}$ is indeterminate. Thus, in this approximation at least, the isotropic case alone requires us to differentiate between $T_C^{(1)}$ and $T_C^{(2)}$ and the zero-magnetization infinite-susceptibility phase can exist *only* for this limit. Thus, $T_C^{(1)}$ moves discontinuously from $T_C^{(2)}$ to zero at the isotropic limit. The manner in which the magnetization curves approach the isotropic limit will be discussed below [see, for example, Fig. 12]. We can therefore refer unambiguously to a Curie temperature T_C for the two-dimensional system in the presence of any nonzero anisotropy.

The Stanley-Kaplan formula (A2) indicates that the proper dependence of $T_C^{(2)}$ on spin quantum number is more complicated than the simple $S(S+1)$ proportionality dictated by conventional effective-mass theories and exemplified by the results plotted in Fig. 5. For application to real (i.e., anisotropic) systems, and in order to produce the best available theoretical results for two-dimensional Curie temperature $T_C = T_C^{(1)} = T_C^{(2)}$, it seems logical to combine the spin dependence of (A2) with the anisotropy dependence of Fig. 5. This is accomplished if the ordinate of Fig. 5 is replaced for $S > \frac{1}{2}$ by $kT_C/J[S(S+1) - \frac{1}{2}]$. A similar form may possibly result if the present Green's-function theory has Φ chosen separately for each S by fitting paramagnetic susceptibility findings to nonclassical series results.

IV. ISOTROPIC SYSTEM

For the isotropic (Heisenberg) quadratic-layer ferromagnet we noted in Sec. III that coherence length $L = Qa$ diverges as the susceptibility diverges $[T \to T_C^{(2)}]$ in such a way that DQ^2 remains finite [where D, defined in (3.10), is $(4J\chi_0)^{-1}$ for the isotropic system]. The limiting value of DQ^2 depends on the parameter Φ, and for the optimum value $\Phi = 0.74$ we find

$$\lim_{Q\to\infty;\, D\to 0} DQ^2 \approx 0.0045 \,. \tag{4.1}$$

It is of considerable interest to ascertain the temperature dependence of susceptibility, coherence length, and also correlation length L_{corr} as functions of $t \equiv T - T_C^{(2)}$ in the vicinity of the singularity. In Fig. 6 we plot reciprocal susceptibility against t^2 for $\Phi = 0.74$ to reveal that $\chi_0 \propto t^{-2}$ over quite an extensive temperature range above $T_C^{(2)}$. In fact,

$$(\chi_0 J)^{-1} = 1.45\,(t/T_C^{(2)})^2 \tag{4.2}$$

is accurate (within the theory) to $\pm 3\%$ up to $T \sim 2T_C^{(2)}$. For $t \lesssim 0.4\,T_C^{(2)}$ the accurate coefficient in (4.2) is close to 1.43, which gives the remarkably simple formula

$$(\chi_0 J)^{-1} = [kt/JS(S+1)]^2 \,, \tag{4.3}$$

accurate to within $\pm 1\%$. It follows from (4.1) that

$$Q \sim 0.134 JS(S+1)/kt = 0.112 T_C^{(2)}/t \tag{4.4}$$

as $t \to 0$. However, the temperature range over which the coherence length varies linearly with $1/t$ is quite small. The computed variation of $a/L = Q^{-1}$ against temperature over a wider paramagnetic range is shown in Fig. 7.

To compute the correlation length L_{corr}, we note that the Fourier transform $\langle \vec{S}_{\vec{K}} \cdot \vec{S}_{-\vec{K}} \rangle$ of the spin correlation $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ can be written for wave-vector magnitude $K < \pi/L$ as

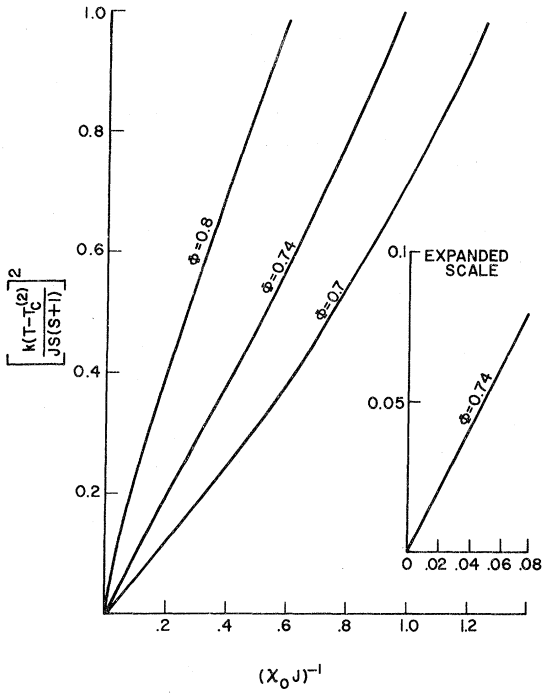

FIG. 6. Temperature dependence of paramagnetic susceptibility in the vicinity of the divergence at $T_C^{(2)}$ plotted for a series of Green's-function decouplings (for the isotropic quadratic-layer system) and exhibiting the close proportionality of susceptibility to $[T-T_C^{(2)}]^{-2}$ for the "best-fit" $\Phi=0.74$ case.

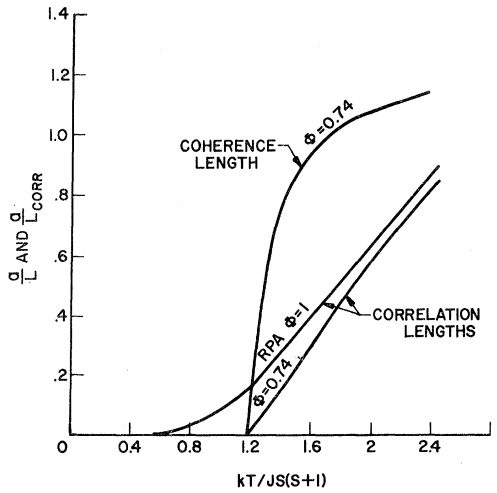

FIG. 7. Temperature dependence of coherence length L and correlation length L_{corr} as calculated by the Green's function method of the present paper for the isotropic Heisenberg quadratic-layer lattice. Also plotted for comparison is the correlation length calculated from RPA Green's-function theory.

$$\langle \vec{S}_{\vec{K}} \cdot \vec{S}_{-\vec{K}} \rangle = 3kT/\{\chi_0^{-1} + 4J[2-\cos(K_x a)-\cos(K_y a)]\} . \tag{4.5}$$

In the limit $K \to 0$, this reduces to

$$\langle \vec{S}_{\vec{K}} \cdot \vec{S}_{-\vec{K}} \rangle = (3kT/2J)/[(K_{corr}^2 + K^2)a^2] , \tag{4.6}$$

where

$$a^2 K_{corr}^2 = a^2/L_{corr}^2 = \tfrac{1}{2}(\chi_0 J)^{-1} . \tag{4.7}$$

We recognize the Ornstein-Zernike form of the relationship. From (4.7), knowing the temperature dependence of susceptibility from our computations above, we can now relate correlation length directly to temperature. The result is shown in Fig. 7 where it is contrasted with the equivalent finding for the RPA Green's function[36] and also with the temperature variation of coherence length reported above. From (4.7) [using (4.2)–(4.4)] we can obtain the behavior of L_{corr} as $t \to 0$. We find

$$L_{corr}/a \sim 1.18(T_C^{(2)}/t), \quad t \to 0 \tag{4.8}$$

and

$$L_{corr} \sim 10.5\, L , \quad t \to 0 . \tag{4.9}$$

Also of interest, and readily computed from (3.8) now that we know the temperature variation χ_0, are the near-neighbor correlations $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ for the isotropic system. We have chosen nn and next nn along an axis (x or y) and the computed results are given in Fig. 8 where we also show the RPA Green's-function result ($\Phi = 1$) for nn.

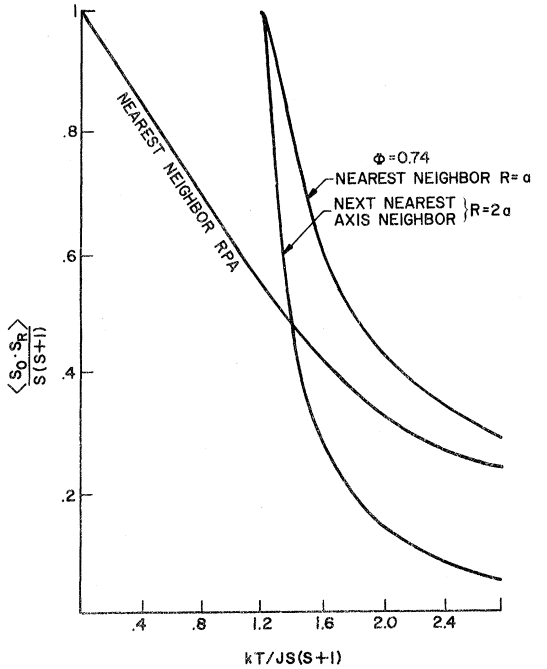

FIG. 8. Temperature dependence of nn ($R=a$) and next-nearest axis neighbor ($R=2a$) correlations $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ as calculated by the theory of the present paper for the isotropic quadratic-layer ferromagnet. Also shown for comparison is the RPA Green's-function result for nn.

Finally, for this isotropic system in the limit of zero applied field, let us consider what happens to correlations at arbitrary distance R as $T_C^{(2)}$ is approached from above. For any large but noninfinite R, as $T \to T_C^{(2)}$, we shall pass through successive situations $L \ll R$, $L \sim R$, and $L \gg R$ which, from our defining equations (3.7)–(3.9) correspond, respectively, to $\langle \vec{S}_0 \cdot \vec{S}_R \rangle \ll 0.74\, S(S+1)$, $\langle \vec{S}_0 \cdot \vec{S}_R \rangle \sim 0.74\, S(S+1)$, and $\langle \vec{S}_0 \cdot \vec{S}_R \rangle \to S(S+1)$. Thus, in the limit $T \to T_C^{(2)}$, the correlation $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ has its maximum value $S(S+1)$ for any finite R. On the other hand, if we take the limit $R \to \infty$ before the limit $T \to T_C^{(2)}$, then $\langle \vec{S}_0 \cdot \vec{S}_R \rangle = 0$. In particular $\langle \vec{S}_0 \cdot \vec{S}_L \rangle = 0.74 S(S+1)$ is always less than its maximum value, and for $R = L_{\text{corr}}$ the correlation $\langle \vec{S}_0 \cdot \vec{S}_R \rangle$ is always small compared to $0.74 S(S+1)$. In other words, the limits $R \to \infty$ and $t \to 0\,[T \to T_C^{(2)}]$ are not commutative. The low-temperature phase must be qualitatively very similar to that found by Mubayi and Lange[10] for their spin-$\frac{1}{2}$ wave-vector-independent decoupling scheme, although the details are rather different, e.g., in Ref. 10 one finds $T_C^{(2)}$ equal to the molecular-field Curie temperature, while the theory of the present paper with $\Phi = 0.74$ gives $T_C^{(2)} = 0.45\, T_C^{\text{mf}}$. Experimentally Miedema[37] finds $T_C/T_C^{\text{mf}} \sim 0.55$ for experimental $S = \frac{1}{2}$ ferromagnets which would make the results of Fig. 5 for the weakly anisotropic ferromagnet fortuitously good for $S = \frac{1}{2}$. Stanley and Kaplan[19] exclude spin $\frac{1}{2}$ from result (A2) because of lack of "smoothness" of the known series coefficients. From Ref. 37, (A2) would seem to be quite inappropriate for $S = \frac{1}{2}$.

V. CONDENSED PHASE

In this section we shall consider the ordered phase for the anisotropic system. We shall, in particular, perform a quantitative calculation of magnetization as a function of temperature for the case $\Delta = 0.01$ (a magnitude of anisotropy which is fairly representative for quadratic-layer systems containing magnetic ions with orbitally nondegenerate ground states) and we shall also examine the behavior of magnetization in the limit $\Delta \to 0$, $h \to 0$.

The first problem which manifests itself for the ordered phase is that the Eqs. (2.23)–(2.26) are not complete when the isotropy restriction $\langle S_0^x S_R^x \rangle = \langle S_0^z S_R^z \rangle$ (used in the paramagnetic calculations) is relaxed. We are confronted with the well-known difficulty of evaluating longitudinal correlations in the Green's-function context.[1,38–41] The exact relation between transverse and longitudinal correlations expressed through the medium of Green's-function theory[40] is only immediately useful when the exact Green's functions are known. For RPA Green's functions in a ferromagnet it leads to a violation of symmetry for the paramagnetic phase, indicating quite unphysical antiferromagnetically correlated $\langle S_0^z S_R^z \rangle$ for nn of equal magnitude but opposite sign to the transverse correlations. A simple physical criterion, which has some justification within first-order effective-mass decoupling[39] and is quite accurate in the low-temperature spin-wave region (although it also violates symmetry conditions at the Curie temperature) is

$$\langle S_0^z S_R^z \rangle = \langle S^z \rangle^2 \equiv (\overline{S})^2 \,. \tag{5.1}$$

For reasons of simplicity, this is the approximation which we shall now use in (2.24), even though we recognize that, at the expense of a considerable increase in labor, a method could be developed[38,41] which would enable us to avoid symmetry violation within the simple decoupling scheme of the present paper. Thus, using (5.1), Eq. (2.24) becomes for a two-dimensional lattice ($d = 2$)

$$\overline{S}^2 + 2\langle S_0^x S_L^x \rangle = \Phi(\overline{S}^2 + 2\langle S_0^x S_{a/\sqrt{2}}^x \rangle) \,, \tag{5.2}$$

where Φ, for the quadratic-layer lattice at least, is 0.74. The approximation (5.1), as incorporated into (5.2), retains the necessary feature that as $\overline{S} \to 0$ (i.e., $T \to T_C$ for any finite anisotropy case) it shows $T_C^{(1)} = T_C^{(2)}$.

An additional simplifying feature which we have adopted for the weakly anisotropic quadratic-layer lattice is to make use of the fact that nn correlations are so nearly complete at T_C that we can without incurring great error assume that short-wavelength magnons ($K > \pi/L$) are completely unrenormalized throughout the ordered phase. This feature of two-dimensional quasi-Heisenberg behavior has already been observed experimentally[15] for the antiferromagnet K_2NiF_4. This enables us to replace short-wavelength magnon energies by simple spin-wave energies in the ordered phase (an approximation which would be far less appropriate for three-dimensional systems).

Thus, our operating equations will be (2.23), (2.25), (2.26), and (5.2) but where $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$ is replaced by spin quantum number S, and where $E_0(K)$ is given by (3.3) and (3.4), the latter also containing the $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2} \to S$ substitution. In short, spin waves with $K < \pi/L$ are treated in RPA and those with $K > \pi/L$ in simple spin-wave approximation with coherence length L determined self-consistently from (5.2). In general, L increases with decreasing temperature and goes to infinity for some temperature $T < T_C$, below which Eq. (5.2) has no solution. This has an obvious physical interpretation, the spin correlations at infinity being then greater than that used to define coherence length, i.e., all spin waves below this temperature are short-wavelength spin waves and renormalize as $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$ in general (the usual interacting spin-wave approximation[26]) but are temperature independent in the approximation being used in the present section for two-dimensional systems.

Solving our equations numerically for the case

$h=0$, $\Delta=0.01$ we obtain the magnetization curves shown in Figs. 9 and 10 (for spin $\frac{1}{2}$ and 1, respectively). Also shown in these figures are the results for three other very interesting approximations; one is simple spin-wave theory, the others are the two Green's-function extremes $\Phi=1$ (which is RPA theory with all magnons renormalizing in long-wavelength fashion according to $\bar{S}$) and the unrenormalized Green's-function approximation (in which magnon energies are completely unrenormalized at all temperatures) which, for the present two-dimensional approximation for short-wavelength magnons, corresponds to the limit $L=\infty$. The latter two theories are very useful, since they always bound the $\Phi=0.74$ theory and, being very much simpler to analyze in the limit of $h\to 0$ and/or $\Delta\to 0$, give us valuable information concerning the qualitative behavior of the constant-Φ theories as the Heisenberg limit is approached.

For spin $\frac{1}{2}$ the $\Phi=0.74$ magnetization curve coincides exactly with the unrenormalized Green's-function curve (and almost exactly with the simple spin-wave curve) up to a temperature $T/T_C=0.52$. The equivalent result for spin 1 is $T/T_C=0.44$. This is in great contrast with RPA Green's-function magnetization which deviates from the spin-wave result significantly at much lower temperatures. A whole family of Φ curves can, of course, be readily computed to lie between the RPA and unrenormalized Green's-function curves. We note that the Curie temperature in the unrenormalized approximation is infinite and that from Fig. 3 this is also true for all theories with $\Phi<0.18$.

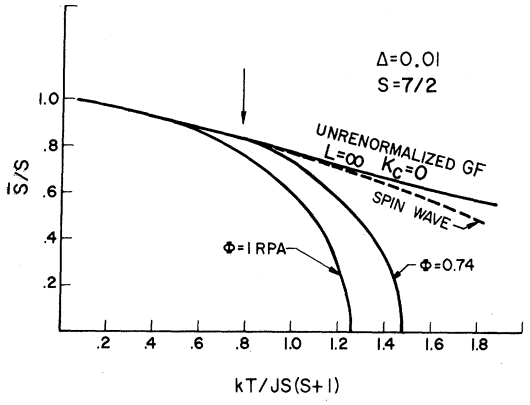

FIG. 9. Magnetization $\bar{S}$ as a function of temperature for the anisotropic quadratic-layer ferromagnet, calculated for anisotropy parameter $\Delta=0.01$ and spin $=\frac{1}{2}$. The theory of the present paper ($\Phi=0.74$) is compared with other approximations, viz., RPA Green's-function theory, simple spin-wave theory, and unrenormalized Green's-function theory (see text). The arrow signifies the temperature for which $\Phi=0.74$ theory begins to differ from the unrenormalized Green's-function and simple spin-wave approximations.

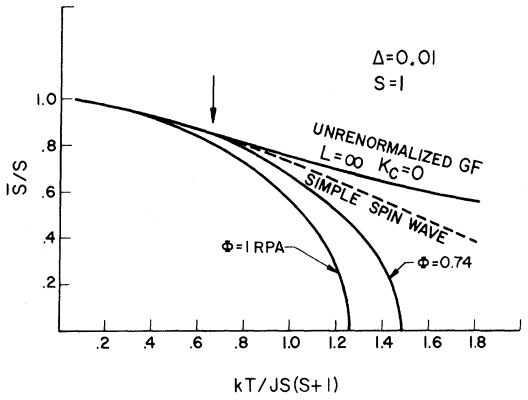

FIG. 10. As Fig. 9 but for spin quantum number $S=1$.

As anisotropy Δ is decreased, we know that T_C (RPA) tends towards zero (Fig. 5) while $T_C(\Phi=0.74)$ tends toward $T_C^{(2)}=1.19JS(S+1)/k$. The former trend is a very slow (logarithmic) one and in Fig. 11 we show the computed results for the relevant theories in the case $S=\frac{1}{2}$, $h=0$, and $\Delta=0.0001$, an anisotropy value already more than an order of magnitude smaller than that which appears to occur naturally in the least anisotropic experimental examples available to date. The qualitative behavior of magnetization curves as we approach the isotropic limit now becomes apparent and is sketched in Fig. 12.

From an academic standpoint, however, the isotropic limit and the approach to it are of considerable importance. Algebraically the easiest approach is to estimate an upper bound on magnetization as given by unrenormalized Green's-function theory. In this approximation x of Eq. (2.26) is given by

$$Nx=\sum_{\vec{K}}\coth[SE_0(\vec{K})/2kT], \tag{5.3}$$

where the summation is over the entire first Brillouin zone of the reciprocal lattice, and where

$$E_0(K)=4J[2(1+\Delta)-\cos(K_xa)-\cos(K_ya)]+(h/S). \tag{5.4}$$

As Δ and h tend towards zero, the summation in (5.3) is dominated by long-wavelength contributions and, replacing the sum by equivalent integral, we find to lowest order

$$x=-(kT/4\pi JS)\ln[2\Delta+(h/4JS)]. \tag{5.5}$$

Since this value of x tends towards infinity as Δ and h tend towards zero, the equation for $\bar{S}$ as a function of x [viz., (2.18)] may be replaced by its form in this limit which is

$$\bar{S}/S=2(S+1)/3x, \quad x\to\infty \tag{5.6}$$

which, on substitution from (5.5) gives

$$\frac{\bar{S}}{S}=-\frac{8\pi JS(S+1)}{3kT\ln[2\Delta+(h/4JS)]}. \tag{5.7}$$

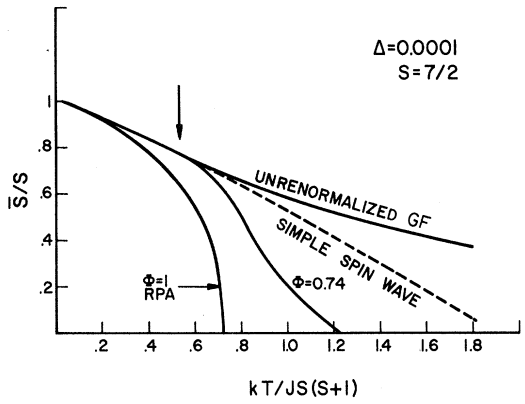

FIG. 11. As Fig. 9 but for $\Delta = 0.0001$.

Here we have two results. In the absence of applied field, magnetization goes to zero in the isotropic limit as $[T\ln(1/\Delta)]^{-1}$ for any nonzero temperature and also, for an isotropic system, magnetization goes to zero for vanishing applied field h as $[T\ln(J/h)]^{-1}$ for any nonzero temperature. These we recall are upper bounds (the latter, in particular, coincides with the finding of Mubayi and Lange[10]) and therefore demonstrate the absence of long-range order for the isotropic system in the absence of applied field and for any nonzero temperature.

Also readily calculated explicitly within the unrenormalized Green's-function theory for the $\Delta \to 0$, $h \to 0$ limit are the transverse components of two spin correlations. For nn we find

$$\langle S_0^x S_a^x \rangle = (kT\bar{S}/8\pi JS)\ln[2\Delta + (h/4JS)]^{-1} , \quad (5.8)$$

which, using (5.7) for $\bar{S}/S$, reduces to

$$\langle S_0^x S_a^x \rangle = \tfrac{1}{3}S(S+1) . \quad (5.9)$$

For arbitrary remote neighbors S_0 and S_R we find

$$\langle S_0^x S_R^x \rangle = \frac{kT\bar{S}}{16\pi^2 JS}\iint_{BZ} \frac{e^{i\vec{K}\cdot\vec{R}}}{\frac{1}{2}K^2a^2 + 2\Delta + (h/4JS)} , \quad (5.10)$$

where the integral $\int\int_{BZ}$ is over the first Brillouin zone for the quadratic-layer lattice. We see that for any finite value of R

$$\lim_{\Delta\to 0;\, h\to 0} \langle S_0^x S_R^x \rangle = \tfrac{1}{3}S(S+1) , \quad (5.11)$$

but that for any nonzero value of Δ or h

$$\lim_{R\to\infty} \langle S_0^x S_R^x \rangle = 0 . \quad (5.12)$$

Once again we find uncommutative limits; this time for $R \to \infty$ and the approach $\Delta \to 0$ and $h \to 0$ to the isolated isotropic Heisenberg system.

Once we have established that $\bar{S}$ goes to zero for the isotropic limit, we can obtain some algebraically simple results for the constant-Φ approximation itself. One readily establishes that

$$\frac{3\langle S_0^x S_a^x \rangle}{S(S+1)} \to \sum_{K<K_C} \frac{\cos(\vec{K}\cdot\vec{a})}{E_0(\vec{K})} \Big/ \sum_{K<K_C} [E_0(\vec{K})]^{-1} \to 1 \quad (5.13)$$

as $\Delta \to 0$ and $h \to 0$, i.e., nn transverse correlations $\langle S_0^x S_a^x \rangle \to \frac{1}{3}S(S+1)$. Indeed, the spins will have this maximum correlation for any spin separation $R \ll L$ (the coherence length). By definition, $\langle S_0^x S_L^x \rangle \to \frac{1}{3}\Phi S(S+1)$. As we approach the isotropic limit, the coherence length diverges and, for the particular case $\Phi = 0.74$ of the present paper $DQ^2 \to 0.0045$ [Eq. (4.1)], where $Q = L/a$ and $D = 2\Delta + (4J\chi_0)^{-1}$. Thus, for any finite spin separation R, the spins will become fully correlated $\langle S_0^x S_R^x \rangle \to \frac{1}{3}S(S+1)$ in the isotropic limit, but for a system with infinitesimal anisotropy or applied field the limiting value of transverse correlations as $R \to \infty$ is zero. When R is the coherence length the correlations are $\frac{1}{3}\Phi S(S+1)$ and when R is the correlation length L_{corr}, the transverse correlation will be small compared to $\frac{1}{3}S(S+1)$.

We note from (4.1) that when $h = 0$ the coherence length diverges in the isotropic limit closely as

$$L \to a/(21\Delta^{1/2}), \quad \Delta \to 0, \quad h = 0 . \quad (5.14)$$

The calculation of magnetization in the constant-Φ approximation is not readily expressed in simple algebraic form because of the temperature dependence of coherence length as dictated by (5.2). Nevertheless, another simple approximation can be used to give a lower bound to $\bar{S}$ as we approach the isotropic limit. This can then be used in conjunction with the upper bound (5.7) from unrenormalized Green's-function theory. The lower-bound approximation replaces the coherence length at $T < T_C$ by its value at the Curie temperature. Using (2.25), (2.26), (3.3), and (3.4) (but with $\langle \vec{S}_0 \cdot \vec{S}_a \rangle^{1/2}$ replaced

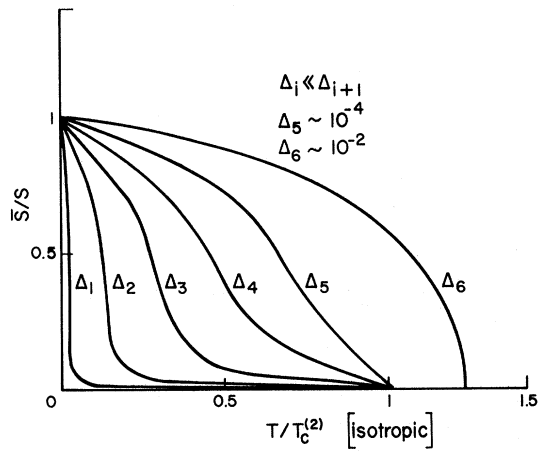

FIG. 12. Qualitative sketch of the temperature dependence of the magnetization as we approach the Heisenberg limit $\Delta = 0$ for a two-dimensional spin system.

by S, as explained above, for two-dimensional systems) we find, in the limit of small Δ and h

$$\langle 1/E_0(\vec{K})\rangle_<^T - \langle 1/E_0(\vec{K})\rangle_<^{T_C} + (\bar{S}/S)\langle 1/E_0(\vec{K})\rangle_>^{T_C} = S(S+1)t/3kT_C^2 , \quad (5.15)$$

where

$$\langle\cdots\rangle_<^T \equiv \frac{1}{N}\sum_{K<K_C(T)}\cdots, \langle\cdots\rangle_>^T \equiv \frac{1}{N}\sum_{K>K_C(T)}\cdots , \quad (5.16)$$

and where $t \equiv T_C - T \ll T_C$. Noting that the first two terms in (5.15) make up a negative quantity for $T < T_C$ [$K_C(T)$ decreases with decreasing temperature], we can write the inequality

$$(\bar{S}/S)\langle 1/E_0(\vec{K})\rangle_>^{T_C} > S(S+1)t/3kT_C^2 . \quad (5.17)$$

Consider first the case $h = 0$ and $\Delta \to 0$. Writing

$$\langle 1/E_0(\vec{K})\rangle_>^{T_C} = \langle 1/E_0(\vec{K})\rangle - \langle 1/E_0(\vec{K})\rangle_<^{T_C}, \quad (5.18)$$

where the first term on the right-hand side involves an average over the entire first Brillouin zone, and noting that

$$\langle 1/E_0(\vec{K})\rangle_<^{T_C} = S(S+1)/3kT_C , \quad (5.19)$$

Eq. (5.17) becomes

$$(\bar{S}/S) > \frac{\alpha(t/T_C)}{\ln(1/2\Delta) - \alpha}, \quad h = 0, \quad \Delta \to 0 \quad (5.20)$$

where $\alpha = 8\pi JS(S+1)/3kT_C$, which for the quadratic-layer ferromagnet ($\Phi = 0.74$) is almost exactly equal to 7.0.

A similar calculation for the limit $\Delta = 0$, $h \to 0$ gives

$$\frac{\bar{S}}{S} > \frac{\alpha(t/T_C)}{\ln Q^2}, \quad \Delta = 0, \quad h \to 0 \quad (5.21)$$

where $Q = L(T_C)/a$. But, in this same limit for the quadratic-layer lattice

$$hQ^2/(\bar{S}) = 0.018J, \quad (5.22)$$

from (3.10) and (4.1). Eliminating Q between (5.21) and (5.22) gives

$$\left(\frac{\bar{S}}{S}\right) > \frac{\alpha(t/T_c)}{\ln[(t/T_c)(0.018JS/h)\alpha]} , \quad \Delta = 0, \quad h \to 0 . \quad (5.23)$$

Comparing (5.20) and (5.23) with upper-bound result (5.7) allows us to conclude that in the constant-Φ approximation, for $T < T_c$,

$$\bar{S} \sim [\ln(1/\Delta)]^{-1}, \quad h = 0, \quad \Delta \to 0 \quad (5.24)$$

$$\bar{S} \sim [\ln(J/h)]^{-1}, \quad \Delta = 0, \quad h \to 0 \quad (5.25)$$

and

$$\chi_0 \sim L^2 \sim [h\ln(J/h)]^{-1}, \quad \Delta = 0, \quad h \to 0. \quad (5.26)$$

APPENDIX

Since the present theory is most valid for high spin quantum numbers, we use the classical series results for determining Φ. For the classical-spin quadratic-layer isotropic Heisenberg lattice the high-temperature susceptibility series is

$$\chi/\chi_{\text{Curie}} = 1 + \sum_{l=1}^{\infty} a_l/\tau^l , \quad (A1)$$

where $\tau = kT/JS(S+1)$ and $\chi_{\text{Curie}} = Ng^2\mu_B^2 S(S+1)/3kT$ and has coefficients a_l which are known exactly[35] out to $l = 9$.

In Fig. 4 we show a plot of $a_l/a_1 a_{l-1}$ vs $1/l$ which should extrapolate to $T_c^{(2)}/T_c^{\text{mf}}$ as the abscissa goes to zero [where $T_C^{\text{mf}} = 8JS(S+1)/3k$ is the molecular-field Curie temperature]. In an earlier assessment of the shorter nonclassical series[19] a formula

$$kT_C^{(2)} \approx \tfrac{1}{5}J(z-1)[2S(S+1)-1] , \quad S > \tfrac{1}{2} \quad (A2)$$

was proposed (where $z = 4$ for our case giving $T_C^{(2)}/T_C^{\text{mf}} \approx 0.45$ as $S \to \infty$). Later analysis of the longer classical series[35] points to a lower $T_C^{(2)}/T_C^{\text{mf}}$ value of perhaps between 0.3 and 0.4.

In Fig. 4 we extrapolate smoothly to an ordinate 0.40. Quantitatively the extrapolation is very closely

$$a_l/a_1 a_{l-1} = 0.40 + 1.4(1/l) - 2.3(1/l)^2, \quad l > 9. \quad (A3)$$

Using (A3) together with the exact terms $l \leq 9$ we have computed the "exact series" curves of Figs. 1 and 2.

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High-Temperature Magnetic Susceptibility of Interacting Electrons in a Solid. II*

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A new alternative expression for the third virial coefficient for fermions is derived. It is used to deduce an expression for the high-temperature magnetic susceptibility of electrons which interact according to the Hubbard Hamiltonian. The work extends a previous calculation which considered only the second virial coefficient. The results are compared with the Stoner theory of the ferromagnetism of itinerant electrons. The inclusion of the third virial coefficient modifies the usual expression for the susceptibility and indicates an increased tendency toward ferromagnetism.

I. INTRODUCTION

In a previous calculation,[1] the virial-expansion technique was used to examine the magnetic susceptibility of a system of interacting electrons described by the single-band Hubbard Hamiltonian.[2] The virial-expansion technique is applicable to low-density systems and gives an approach to the study of such systems at temperatures higher than the Curie temperature of a possible ferromagnetic transition. The work mentioned above contained a rigorous determination of the second virial coefficient using the methods of solid-state scattering theory.[3] The spin susceptibility which was obtained agreed with the high-temperature susceptibility in the Stoner theory of the ferromagnetism of itinerant electrons.[4] This enabled the establishment of a relation between the paramagnetic Curie temperature of that theory and an element of the t matrix for particle-particle scattering.

The object of this paper is to consider the third virial coefficient for the same system. We are as yet unable to calculate this exactly as was done for the second virial coefficient, and our final result still involves one significant approximation, the nature of which will be specified subsequently.

Several authors have discussed the quantum theory of the third virial coefficient.[5–8] The derivation of this quantity is considered in Sec. II of this paper. The determination of the third virial coefficient b_3 requires information concerning three-particle scattering processes. We employ the Faddeev equations[9] to obtain a concise expression for b_3 [Eq. (2.25)] which contains a free-particle part and a term involving only connected diagrams for three particles. This expression is rigorous, general,