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Phase Transition in the Two-Dimensional Heisenberg Ferromagnet

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We develop a Green-function theory to describe the thermodynamic behavior of a plane square lattice with spins of magnitude one-half located at the lattice sites interacting via a nearest-neighbor Heisenberg ferromagnetic coupling. Our approximation technique involves a decoupling of the hierarchy of Green-function equations similar in some respects to that found in the random-phase approximation (RPA) but improved to include spin correlations neglected in the RPA. Such an improvement is essential for the two-dimensional problem. Our theory predicts a phase transition at the temperature given by $kT_c=2J$, where J is the exchange parameter. As T approaches T_c from above, the static susceptibility diverges as $1/(T-T_c)$. The spontaneous magnetization is zero at all nonzero temperatures, both above and below the critical point. Therefore, our theory is consistent with the existing rigorous proof of Mermin and Wagner that the spontaneous magnetization must be zero for $T\neq0$, and displays the divergent susceptibility predicted by Stanley and Kaplan from an analysis of high-temperature expansions for related two-dimensional spin systems.

1. INTRODUCTION

RECENTLY, the two-dimensional lattice with Heisenberg ferromagnetic exchange coupling between the lattice spins has been the object of two stimulating theoretical observations. Stanley and Kaplan[1] searched for a phase transition in such a system by studying the high-temperature series expansions for the zero-field, static magnetic susceptibility. They find that the susceptibility diverges at a finite temperature as had been found previously with similar techniques for the three-dimensional lattice.[2] Within the context of their approach they predict a phase transition, though, as they suggest, not necessarily to a state with spontaneous magnetization, at some finite temperature. On the other hand, Mermin and Wagner,[3] adapting a technique of Hohenberg[4] which utilizes the Bogoliubov inequality,[5] have rigorously shown that there can be no spontaneous magnetization at any finite temperature in the two-dimensional lattice with Heisenberg exchange interactions between the spins. The reason is related to the fact that if there is magnetization, there will be modes, as predicted by the Goldstone theorem[6] which will have sufficient weight at long wavelengths to cause severe fluctuations at any finite temperature. The proof consists of showing that, in fact, the fluctuations would be so large that inconsistencies arise which can only be removed by requiring that there be no spontaneous magnetization in the first place. So, if there does exist a phase transition, ferromagnetic in some sense, it will not involve the onset of spontaneous magnetization of the usual sort. The problem of building a theory for this model becomes of interest, since it promises to increase our understanding of phase transitions and the types of order which are possible at low temperatures.

The RPA[7] predicts no critical temperature of any sort for the two-dimensional lattice. Since this approximation really involves only a single parameter, the spontaneous magnetization, anything interesting in the way of temperature dependence of the susceptibility would have to involve a nonzero temperature-dependent spontaneous magnetization. Rather than violate the rigorous results of Ref. 3, the RPA predicts nothing interesting at any temperature.

What we succeed in doing in this paper is constructing a theory which in spirit and technique is similar to the RPA. However, the effective field parameter in our theory is more complicated than the magnetization, so that this magnetization can be zero at finite temperature without washing out all other possible predictions. In fact, we do find that for the case of spins of magnitude one-half situated on a plane square lattice, and interacting via nearest-neighbor isotropic exchange forces,

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our theory is consistent with the results of Stanley and Kaplan as well as those of Mermin and Wagner. We find that the spontaneous magnetization, defined in the usual way,[8] is zero at all nonzero temperatures, while the static zero-field susceptibility diverges at a finite temperature. At very low temperatures, our calculations predict that the specific heat varies linearly with temperature. This is in agreement with recent experiments of Miedema *et al.*[9] on the heat capacity of compounds which, from a magnetic point of view, have two-dimensional crystal structures.

2. GREEN-FUNCTION APPROXIMATION

The application of double-time temperature-dependent Green functions to the theory of the Heisenberg ferromagnet has been discussed many times before, and we refer the reader to the extensive literature on the subject.[10] For our purposes the following well-known results will be sufficient.

The retarded Green function of operators $A(t)$ and $B(t')$ (which are in the Heisenberg representation) is defined as

$$\langle\langle A(t);B(t')\rangle\rangle = -i\theta(t-t')\langle[A(t);B(t')]\rangle. \quad (1)$$

Here $\theta(t)$ is the step function, square brackets denote a commutator, and the angular brackets denote an average over the canonical ensemble,

$$\langle X\rangle = \mathrm{Tr}\exp(-\beta\mathcal{H})X/\mathrm{Tr}\exp(-\beta\mathcal{H}),$$

where $\beta=1/k_BT$, k_B being the Boltzmann constant, T the absolute temperature, and $\mathcal{H}$ the Hamiltonian of the system.

The equation of motion of the Green function so defined is

$$i(\partial/\partial t)\langle\langle A(t);B(t')\rangle\rangle = \langle[A(t);B(t')]\rangle\delta(t-t') + \langle\langle[A(t),\mathcal{H}];B(t')\rangle\rangle. \quad (2)$$

The second term on the right-hand side is, in general, a Green function of higher order and so an approximation or "decoupling" can be used to solve the equation of motion for $\langle\langle A(t);B(t')\rangle\rangle$. Once this decoupling has been done and $\langle\langle A(t);B(t')\rangle\rangle$ has been evaluated, the correlation function $\langle B(t')A(t)\rangle$ can be obtained from the formula

$$\langle B(t')A(t)\rangle = \lim_{\epsilon\to 0^+} i\int \frac{\langle\langle A;B\rangle\rangle_{E+i\epsilon}-\langle\langle A;B\rangle\rangle_{E-i\epsilon}}{e^{\beta E}-1} e^{-iE(t-t')}dE, \quad (3)$$

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

The Heisenberg Hamiltonian for a system of N spins $\mathbf{S}_i$ localized at each lattice site i, interacting through an isotropic, ferromagnetic exchange coupling J, in the presence of a uniform time-independent magnetic field H directed along the z-axis is of the form

$$\mathcal{H} = -gH\sum_{i=1}^{N} S_i^z - \sum_i\sum_j J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j, \quad (4)$$

$J_{ij}=J_{ji}$, $J_{ii}=0$, $J_{ij}>0$. gS is the magnetic moment per ion at each lattice site of spin S. The spin operators obey the usual commutation rules:

$$[S_f^x,S_g^y] = iS_f^z\delta_{fg},$$
$$[S_f^z,S_g^\pm] = \pm S_f^\pm\delta_{fg},$$
$$[S_f^+,S_g^-] = 2S_f^z\delta_{fg},$$

where

$$S_f^\pm = S_f^x \pm iS_f^y.$$

For spin one-half, we have the additional relations

$$S_f^z = \tfrac{1}{2} - S_f^-S_f^+, \quad (5)$$

$$(S_f^+)^2 = (S_f^-)^2 = 0. \quad (6)$$

In what follows, we shall restrict our analysis to spin one-half.

We consider the Green function as defined in Eq. (1) with $A(t)=S_i^+(t)$ and $B(t')=S_j^-(t')$:

$$G_{ij}(t-t') = \langle\langle S_i^+(t);S_j^-(t')\rangle\rangle = -i\theta(t-t')\langle[S_i^+(t),S_j^-(t')]\rangle.$$

Its equation of motion is

$$i(\partial/\partial t)G_{ij}(t-t') = 2\langle S^z\rangle\delta_{ij}\delta(t-t') + g\mathrm{H}G_{ij}(t-t') - 2\sum_l J_{il}\{\langle\langle S_l^z(t)S_i^+(t);S_j^-(t')\rangle\rangle - \langle\langle S_i^z(t)S_l^+(t);S_j^-(t')\rangle\rangle\}. \quad (7)$$

So the higher-order Green function to be decoupled is of the form

$$\langle\langle S_l^z(t)S_i^+(t);S_j^-(t')\rangle\rangle. \quad (8)$$

The RPA decoupling,[7]

$$\langle\langle S_l^z(t)S_i^+(t);S_j^-(t')\rangle\rangle \xrightarrow[\mathrm{RPA}]{} \langle S^z\rangle\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle, \quad (9)$$

neglects the correlations between the z component of the spin at the site l and the transverse components at the other sites i and j and replaces $S_l^z(t)$ by its average value $\langle S^z\rangle$.

To gain some insight into the effect of these correlations, we use Eq. (5), valid for spin one-half, to rewrite expression (8) as

$$\tfrac{1}{2}\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle - \langle\langle S_l^-(t)S_l^+(t)S_i^+(t);S_j^-(t')\rangle\rangle. \quad (10)$$

[8] This is the definition used, for example, by Mermin and Wagner, Ref. 3. See also, Sec. II A of R. B. Griffiths, Phys. Rev. **152**, 240 (1966).

[9] J. Koppen, R. Hamersma, J. V. Lebesque, and A. R. Miedema, Phys. Letters **25A**, 376 (1967).

[10] See, for example, D. N. Zubarev, Usp. Fiz. Nauk **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)]; also, V. L. Bonch-Bruevich and S. V. Tyablikov, in *The Green Function Method in Statistical Mechanics*, edited by D. ter Haar (North-Holland Publishing Company, Amsterdam, 1962).

We see that the second term in expression (10) has a strong dependence on the indices i and l since this term vanishes identically when $i=l$ as a consequence of Eq. (6). Similarly, at equal time, $t=t'$, there is a strong dependence on the sites l and j. However, this spatial dependence which leads to correlations between the spin operators at the respective sites, i, l, and j is lost completely as soon as a decoupling of the form given by (9) is introduced. To look for an improvement over the RPA, we must somehow take into account the correlations neglected by the RPA.

The decoupling approximation we propose (for the $S=\frac{1}{2}$ case) is

$$\begin{aligned}\langle\langle S_l^z(t)S_i^+(t);S_j^-(t')\rangle\rangle=&\tfrac{1}{2}\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle\\&+\alpha_{il}\langle S_l^-S_l^+\rangle\langle\langle S_i^+(t);S_j^-(t')\rangle\rangle\\&+\gamma_{il}\langle S_l^-S_i^+\rangle\langle\langle S_l^+(t);S_j^-(t')\rangle\rangle,\end{aligned}\qquad(11)$$

where α_{il} and γ_{il} are parameters which are determined by two conditions described below. This decoupling reduces to the RPA for the following particular values:

$$\alpha_{il}=-1,\quad\langle S_l^-S_i^+\rangle=0,\quad i\neq l.$$

The idea of decoupling the higher-order Green function with a parameter was first introduced by Gallen,[11] who determined the parameter, using a plausible physical argument. In our case, the first condition we use is essentially one of self-consistency, while the second condition incorporates the stringent requirements of spin kinematics.

The first condition uses the fact that the difference of the higher-order Green functions

$$\langle\langle S_l^z(t)S_i^+(t);S_j^-(t')\rangle\rangle-\langle\langle S_i^z(t)S_l^+(t);S_j^-(t')\rangle\rangle$$

occurring on the right-hand side of (7) can be evaluated exactly in the equal time $t=t'$ limit. We can also evaluate it, in the same limit, after introducing the decoupling. If we require, as a consistency condition on our parameters, that these two evaluations be equal,[12] we obtain the relation

$$\begin{aligned}\langle 2S_l^zS_i^z+S_i^-S_l^+\rangle=&\langle S^z\rangle+2\alpha_{il}\langle S^-S^+\rangle\langle S^z\rangle\\&-2\gamma_{il}\langle S_i^-S_l^+\rangle\langle S^z\rangle.\end{aligned}\qquad(12)$$

(We have made use of the fact that $\langle S_i^z\rangle$ and $\langle S_i^-S_i^+\rangle$ are independent of the lattice index i on account of translational invariance of the lattice.) This provides one equation connecting the parameters.

The second condition makes use of what has come to be known in the literature as "Dyson's kinematical interaction." This is the fact that reversing a spin more than $2S$ times at any one lattice site gives zero, and it is embodied in Eq. (6). From Eq. (3) we note that the correlation function $\langle S_i^-(t')S_i^-(t)S_i^+(t)S_l^+(t)\rangle$, which is identically zero for $t=t'$, can be written as an integral over the discontinuity of the corresponding Green function:

$$\begin{aligned}0=&\lim_{t\to t'}\langle S_i^-(t')S_i^-(t)S_i^+(t)S_l^+(t)\rangle\\=&\lim_{\epsilon\to0^+,t\to t'} i\int\frac{\langle\langle S_i^-S_i^+S_l^+;S_i^-\rangle\rangle_{E+i\epsilon}-\langle\langle S_i^-S_i^+S_l^+;S_i^-\rangle\rangle_{E-i\epsilon}}{e^{\beta E}-1}e^{-iE(t-t')}dE.\end{aligned}$$

On introducing our decoupling into the Green functions on the right-hand side we obtain for $i\neq l$

$$\begin{aligned}0=&\alpha_{il}\langle S^-S^+\rangle\lim_{\epsilon\to0^+} i\int\frac{\langle\langle S_l^+;S_i^-\rangle\rangle_{E+i\epsilon}-\langle\langle S_l^+;S_i^-\rangle\rangle_{E-i\epsilon}}{e^{\beta E}-1}dE+\gamma_{il}\langle S_i^-S_l^+\rangle\lim_{\epsilon\to0^+} i\int\frac{\langle\langle S_i^+;S_i^-\rangle\rangle_{E+i\epsilon}-\langle\langle S_i^+;S_i^-\rangle\rangle_{E-i\epsilon}}{e^{\beta E}-1}dE\\=&\alpha_{il}\langle S^-S^+\rangle\langle S_i^-S_l^+\rangle+\gamma_{il}\langle S_i^-S_l^+\rangle\langle S^-S^+\rangle.\end{aligned}$$

For $\langle S^-S^+\rangle$ and $\langle S_i^-S_l^+\rangle$ not equal to zero, this gives

$$\alpha_{il}=-\gamma_{il}.\qquad(13)$$

Making use of Eq. (5), we can write, for $S=\frac{1}{2}$

$$\langle S_l^zS_i^z\rangle=\tfrac{1}{4}-\langle S^-S^+\rangle+\langle S_l^-S_l^+S_i^-S_i^+\rangle,\quad i\neq l.$$

The correlation function $\langle S_l^-S_l^+S_i^-S_i^+\rangle$ can be expressed in terms of $\langle S^-S^+\rangle$ and $\langle S_i^-S_l^+\rangle$, and the parameters, if, as before, we use Eq. (3) with the decoupling introduced into the Green functions. We then arrive at the following relation:

$$\langle S_l^zS_i^z\rangle=\tfrac{1}{4}-\langle S^-S^+\rangle-\alpha_{il}\langle S^-S^+\rangle^2-\gamma_{il}\langle S_i^-S_l^+\rangle^2.\qquad(14)$$

Equations (12), (13), and (14) can be solved along with Eq. (5) to obtain

$$\alpha_{il}=\frac{-\frac{1}{2}+\langle S^z\rangle+\langle S_i^-S_l^+\rangle}{\frac{1}{2}-\langle S^z\rangle+2\langle S^z\rangle\langle S_i^-S_l^+\rangle-2\langle S_i^-S_l^+\rangle^2}.\qquad(15)$$

Along with Eq. (13), this determines both parameters in terms of $\langle S^z\rangle$ and the transverse correlation function $\langle S_i^-S_l^+\rangle$.

[11] H. B. Callen, Phys. Rev. **230**, 590 (1963).

[12] The reason for doing this is that within the Green-function decoupling technique, the energy of the elementary excitations (i.e., the location of the pole of the spectral Green function) can be obtained either by introducing the decoupling into the equal-time correlation function or directly into the Green function. In the RPA, for example, these two ways of calculating the energy lead to different results. So our consistency condition that the two evaluations be equal amounts to a consistency requirement on the energy.

We note that our decoupling technique and method of obtaining the parameters has taken into account the correlations that we earlier mentioned were neglected by the RPA. Indeed, for $i=l$, the second term of (10) does identically vanish as required by spin kinematics [this can be seen immediately from Eqs. (11) and (13)], while the correlation between the sites j and l at equal time has been explicitly taken into account in the second condition used to determine the parameters.

The equation of motion (7) can be Fourier-transformed with respect to the time difference $t-t'$, giving the Green-function dependence on the energy variable E. Since the parameters α_{il} and γ_{il} are not functions of time, they do not complicate this operation and we get simply

$$EG_{ij}(E)=(2\langle S^z\rangle/2\pi)\delta_{ij}+gHG_{ij}(E)-2\sum_l J_{il}(G_{ij}(E)-G_{lj}(E))(\tfrac{1}{2}+\alpha_{il}\rho-\gamma_{il}\mu_{il}), \quad (16)$$

where $\rho=\langle S^-S^+\rangle$ and $\mu_{il}=\langle S_i^-S_l^+\rangle$. Translational invariance allows spatial Fourier transformation,

$$G_{ij}=\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(r_i-r_j)}G(\mathbf{k}),$$

$$J_{ij}=\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(r_i-r_j)}J(\mathbf{k}),$$

$$\delta_{ij}=\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(r_i-r_j)}, \quad \text{etc.,}$$

and the inverse transformations

$$G(\mathbf{k})=\sum_{|i-j|} e^{-i\mathbf{k}\cdot(r_i-r_j)}G_{ij},$$

$$J(\mathbf{k})=\sum_{|i-j|} e^{-i\mathbf{k}\cdot(r_i-r_j)}J_{ij}, \quad \text{etc.}$$

We assume periodic boundary conditions, so the reciprocal lattice vectors $\mathbf{k}$ are summed over the first Brillouin zone.

We shall restrict J to nearest-neighbor interactions. That is,

$$J_{ij}=J \quad \text{if} \quad r_i-r_j=\boldsymbol{\delta},$$

where $\boldsymbol{\delta}$ is a lattice vector connecting nearest neighbors, and

$$J_{ij}=0 \quad \text{otherwise.}$$

On account of this restriction, a very useful property of certain spatial Fourier transforms becomes valid. If A_{ij} is any function of the lattice separation r_i-r_j and A_δ, the nearest-neighbor part of A_{ij}, is independent of the direction of $\boldsymbol{\delta}$ and equals A_δ (this is the assumption of equivalent nearest neighbors), then

$$\sum_j J_{ij}A_{ij}e^{-i\mathbf{k}\cdot(r_i-r_j)}=A_\delta\sum_j J_{ij}e^{-i\mathbf{k}\cdot(r_i-r_j)}=A_\delta J(\mathbf{k}).$$

We exploit this simplifying property, and introducing the notation, with the assumption that all nearest neighbors are equivalent,

$$\mu_\delta\equiv\mu,$$
$$\alpha_\delta\equiv\alpha,$$
$$\gamma_\delta\equiv\gamma,$$

we get the following form for Eq. (16) after spatial Fourier transformation:

$$[E-gH-E(\mathbf{k})]G(\mathbf{k},E)=2\langle S^z\rangle/2\pi, \quad (17)$$

where

$$E(\mathbf{k})=(1+2\alpha\rho-2\gamma\mu)[J(0)-J(\mathbf{k})].$$

The previous calculations relating α and γ to μ and $\langle S^z\rangle$ can be used to rewrite this as

$$E(\mathbf{k})=(\langle S^z\rangle/\tfrac{1}{2}-\mu)[J(0)-J(\mathbf{k})]. \quad (18)$$

In the above equation, μ is the nearest-neighbor part of the transverse correlation function

$$\mu=\langle S_i^-S_{i+\delta}^+\rangle=\langle S_i^xS_{i+\delta}^x\rangle+\langle S_i^yS_{i+\delta}^y\rangle.$$

Equation (18) becomes identical with the RPA result if we set $\mu=0$. We thus see that, in our decoupling approximation, the effective field parameter renormalizing the free spin wave energies is not simply the magnetization, as it is in the RPA, but is a function both of the magnetization and a quantity related to the short-range order in the system.

3. BASIC EQUATIONS

From Eq. (3) and the fact that for real ω and E

$$\lim_{\epsilon\to 0^+}\left[\frac{1}{\omega-E+i\epsilon}-\frac{1}{\omega-E-i\epsilon}\right]=-2\pi i\delta(\omega-E),$$

we obtain

$$\langle S_i^-S_j^+\rangle=2\langle S^z\rangle\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(r_i-r_j)}/e^{\beta[gH+E(\mathbf{k})]}-1.$$

Using the identity

$$\frac{1}{e^{\beta\omega}-1}=\tfrac{1}{2}\coth\tfrac{1}{2}\beta\omega-\tfrac{1}{2},$$

we get

$$\langle S_i^-S_j^+\rangle=\langle S^z\rangle\frac{1}{N}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(r_i-r_j)}\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})]-\langle S^z\rangle\delta_{ij}. \quad (19)$$

The relative magnetization is given by the expression

$$\sigma=\langle S^z\rangle/S=2\langle S^z\rangle \quad \text{for spin one-half.}$$

Putting $i=j$ in Eq. (19), we find the relative magnetization is given by

$$\frac{1}{\sigma}=\frac{1}{N}\sum_{\mathbf{k}}\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})],\qquad(20)$$

while the equation for the transverse correlation function for $i\neq j$ is

$$\frac{\langle S_i^-S_j^+\rangle}{\sigma}=\frac{1}{2N}\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot(\mathbf{r}_i-\mathbf{r}_j)}\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})].\qquad(21)$$

An important quantity in our model is the nearest-neighbor part of the transverse correlation function, μ, given by

$$\frac{\mu}{\sigma}=\frac{1}{2N}\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\delta}\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})].\qquad(22)$$

We consider the case of a plane square lattice with a lattice constant$=a$ and the coordination number $z=4$. For this case,

$$J(\mathbf{k})=J\sum_1^z e^{-i\mathbf{k}\cdot\delta}=\tfrac{1}{2}J(0)[\cos k_1a+\cos k_2a],$$

where $J(0)=zJ$. The sums over $\mathbf{k}$ can be replaced by integrals, using the familiar prescription

$$\frac{1}{N}\sum_{\mathbf{k}}=\frac{\Omega}{(2\pi)^n}\int d^nk,$$

where n is the number of dimensions and Ω is the volume in n dimensions of a unit cell. In our case, this becomes

$$\frac{1}{N}\sum_{\mathbf{k}}=\frac{\Omega}{(2\pi)^2}\int d^2k,$$

where $\Omega=a^2$ and, since $\mathbf{k}$ is restricted to the first Brillouin zone, the integration limits are $-\pi/a$ to $+\pi/a$. With this prescription, and the fact that $E_{\mathbf{k}}=E_{-\mathbf{k}}$, Eqs. (20), (21), and (22) become

$$\frac{1}{\sigma}=\frac{\Omega}{(2\pi)^2}\int\int_{-\pi/a}^{\pi/a}d^2k\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})],\qquad(23)$$

$$\frac{\langle S_i^-S_j^+\rangle}{\sigma}=\frac{\Omega}{2(2\pi)^2}\int\int_{-\pi/a}^{\pi/a}\cos\mathbf{k}\cdot(r_i-r_j)\times\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})]d^2k,\qquad(24)$$

and

$$\frac{\mu}{\sigma}=\frac{\Omega}{2(2\pi)^2}\int\int_{-\pi/a}^{\pi/a}d^2k\cos k_1a\times\coth\tfrac{1}{2}\beta[gH+E(\mathbf{k})].\qquad(25)$$

The last equation incorporates the fact that for any pair of nearest neighbors,

$$\cos\mathbf{k}\cdot\mathbf{\delta}=\cos k_1a\quad\text{or}\quad\cos k_2a.$$

In these expressions,

$$E(\mathbf{k})=\frac{\langle S^z\rangle}{\frac{1}{2}-\mu}[J(0)-J(\mathbf{k})]=\frac{\sigma}{\frac{1}{2}-\mu}2J(1-\tfrac{1}{2}\cos k_1a-\tfrac{1}{2}\cos k_2a).$$

If we set $2J=g=1$, β and H become dimensionless quantities. β now measures inverse temperature in units of $(k_B/2J)$ and H is the external field (in the z direction) in units of $2J/g$.

At this point, we also change the integration variables to eliminate the lattice spacing, a, and Eqs. (23) and (25) become

$$\frac{1}{\sigma}=\frac{1}{(2\pi)^2}\int\int_{-\pi}^{\pi}d^2k\times\coth\tfrac{1}{2}\beta\left(H+\frac{\sigma}{\frac{1}{2}-\mu}(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)\right),\qquad(26)$$

$$\frac{\mu}{\sigma}=\frac{1}{2(2\pi)^2}\int\int_{-\pi}^{\pi}d^2k\cos k_1\times\coth\tfrac{1}{2}\beta\left(H+\frac{\sigma}{\frac{1}{2}-\mu}(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)\right).\qquad(27)$$

4. DETERMINATION OF C

If we define a quantity C by

$$C=\sigma/(\tfrac{1}{2}-\mu),$$

then Eqs. (26) and (27) can be combined to give

$$\frac{1}{C}=\frac{1}{2(2\pi)^2}\int\int d^2k(1-\cos k_1)\times\coth\tfrac{1}{2}\beta(H+C[1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2]).\qquad(28)$$

For a spin one-half system, $0\leq\sigma\leq1$ and $0\leq|\mu|\leq\frac{1}{2}$ so the quantity C is non-negative. The hyperbolic cotangent has the property

$$\coth x\geq|1/x|.\qquad(29)$$

Introducing this into Eq. (28) we obtain

$$\frac{1}{C}\geq\frac{1}{\beta}\frac{1}{(2\pi)^2}\int\int\frac{d^2k(1-\cos k_1)}{H+C[1-\frac{1}{2}\cos k_1-\frac{1}{2}\cos k_2]}.\qquad(30)$$

We make use of the fact that

$$\frac{1}{(2\pi)^2}\int\int_{-\pi}^{\pi}\frac{\cos k_1 d^2k}{1+H/C-\frac{1}{2}\cos k_1-\frac{1}{2}\cos k_2}$$

$$=\frac{1+H/C}{(2\pi)^2}\int\int_{-\pi}^{\pi}\frac{d^2k}{1+H/C-\frac{1}{2}\cos k_1-\frac{1}{2}\cos k_2}-1$$

and

$$\frac{1}{(2\pi)^2}\int\int_{-\pi}^{\pi}\frac{d^2k}{1+H/C-\frac{1}{2}\cos k_1-\frac{1}{2}\cos k_2}$$

$$=\frac{2}{\pi}\frac{1}{1+H/C}K\left(\frac{1}{1+H/C}\right), \quad (31)$$

where $K(k)$ is a complete elliptic integral of the first kind defined by[13]

$$K(k)=\int_0^{\pi/2}\frac{d\phi}{1-k^2\sin^2\varphi}$$

to rewrite (30) as

$$(1-\xi)K(\xi)\geq\tfrac{1}{2}\pi(1-\beta), \quad (32)$$

where $\xi=(1+H/C)^{-1}$.

We wish to evaluate C as a function of the external field H, in the limit as H becomes small, in various temperature ranges. In this connection, the following property of $K(\xi)$[14] is useful: For $\xi\lesssim 1$,

$$K(\xi)\simeq\ln\frac{4}{\xi'}+\frac{1}{2}\left(\ln\frac{4}{\xi'}-1\right)\xi'^2+\cdots,$$

when $\xi'^2=1-\xi^2$. With the definition of ξ given above, we have that for $\xi\lesssim 1$,

$$K(\xi)\simeq\tfrac{1}{2}|\ln(H/C)|+\cdots$$

and

$$(1-\xi)K(\xi)\simeq\tfrac{1}{2}(H/C)|\ln(H/C)|+\cdots. \quad (33)$$

a. $\beta<1$

For $\beta<1$, relation (32) shows that $\lim_{H\to 0}H/C$ must tend to a definite nonzero limit. If it approached zero, then the fact that $\lim_{x\to 0}x\ln x=0$ would make the left-hand side of relation (32) approach zero, which would contradict $\beta<1$. In this temperature region, then,

$$\lim_{H\to 0}C=b(\beta)H, \quad (34)$$

where b is independent of H but depends on the temperature. This is essentially the region of paramagnetic response. Further, with such a behavior of C it is clear that, in this temperature region, the inequality (32) reduces to an equality in the limit $H=0$. This can be seen directly by substituting the series expansion[15]

$$\coth x=\frac{1}{x}+\sum_{n=1}^{\infty}\frac{(-1)^{n-1}2^{2n}}{(2n)!}B_n x^{2n-1}$$

for the hyperbolic cotangent into Eq. (28) and noting that, apart from the first term [which leads to (32)], the rest of the expansion is a power series in H and C which tends to zero as H tends to zero.

A numerical solution for $\lim_{H\to 0}C/H$ as a function of β treating relation (32) as an equality leads to the result that $\lim_{H\to 0}C/H$ gets larger as β gets closer to unity. This suggests intuitively that $\lim_{H\to 0}C$ is going to zero more and more slowly compared to H as β gets closer to the value 1.

b. $\beta=1$

We have seen that for $\beta<1$, the replacement of the hyperbolic cotangent in Eq. (28) by its first term in a series expansion is justified in the $H\to 0$ limit since

$$\tfrac{1}{2}\beta[H+C(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)]\ll 1 \quad \text{for all } k\subset\{-\pi,\pi\}$$

and the higher-order terms in the expansion will make small contributions compared to $1-\beta$ on the right-hand side of the inequality (32).

At $\beta=1$, however, $1-\beta$ vanishes and we need at least the next term in the coth expansion. We now use the inequality

$$\coth x\leq 1/x+\tfrac{1}{3}x.$$

Substitution in Eq. (28) yields

$$(2/\pi)(1-\xi)K(\xi)\leqq(1-\beta)+(\beta^2/12)HC+(5/48)\beta^2C^2. \quad (35)$$

In the limit $H\to 0$, this becomes an equality for reasons similar to those given previously. We have also established that for $\beta\lesssim 1$, $K(\xi)$ can be replaced to a very good approximation by $\frac{1}{2}|\ln(H/C)|$. At $\beta=1$ and H approaching zero, relation (35) then becomes

$$\frac{H}{C}\left|\ln\frac{H}{C}\right|=\frac{5}{48}\pi C^2. \quad (36)$$

Solving Eq. (36), we obtain

$$\lim_{H\to 0}C=H^{\frac{1}{3}}|\ln H|^{\frac{1}{3}}\left(\frac{32}{15\pi}\right)^{\frac{1}{3}}. \quad (37)$$

Since $|\ln H|$ varies much more slowly than H in the region $H\to 0$, we may regard Eq. (37) as the power law relation for $\beta=1$:

$$\lim_{H\to 0}C\propto H^{\frac{1}{3}}. \quad (38)$$

[13] E. Jahnke and F. Emde, *Tables of Functions* (Dover Publications, Inc., New York, 1943).
[14] E. Jahnke and F. Emde, Ref. 13.
[15] H. B. Dwight, *Tables of Integrals and Other Mathematical Data* (The Macmillan Company, New York, 1963).

c. $\beta > 1$

For $\beta>1$, the relation (35) already shows that $\lim_{H\to 0} C$ becomes independent of H. In fact, for small but positive $\beta-1$ we have

$$C \propto (\beta-1)^{\frac{1}{2}}+\cdots. \tag{39}$$

In summary, the behavior of $\lim_{H\to 0} C$ as a function of H in various temperature ranges is the following:

$$\begin{aligned}\lim_{H\to 0} C &= \text{const}\times H, & \beta<1\\ &= (\text{const}\times |\ln H|^{\frac{1}{3}})\times H^{\frac{1}{3}}, & \beta=1\\ &= \text{independent of } H, & \beta>1.\end{aligned}$$

Since C is the effective field parameter in our theory, a shift from a linear to a power-law behavior signifies a change at $\beta=1$ in the response of the spin system to an infinitesimal external field. In the next section, we show that the static susceptibility diverges at $\beta=1$.

5. DIVERGENCE OF STATIC SUSCEPTIBILITY

We proceed to analyze the static zero-field susceptibility χ given by

$$\lim_{H\to 0} \chi = \lim_{H\to 0} \frac{\sigma}{H},$$

as we approach $\beta=1$ from the high-temperature side. Equation (26) for the magnetization may now be utilized to obtain

$$\frac{1}{\chi H} = \frac{1}{(2\pi)^2} \iint_{-\pi}^{\pi} d^2k \times \coth\tfrac{1}{2}\beta[H+C(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)]. \tag{40}$$

Substitution of Eqs. (29) and (31) into Eq. (40) yields

$$\frac{1}{\chi} \geq \frac{4}{\pi\beta}(1-\xi)K(\xi). \tag{41}$$

For $\beta<1$, (41) becomes an equality in the limit $H\to 0$ (for reasons pointed out earlier) and comparing with relation (32) we get

$$\lim_{H\to 0} \chi = \frac{\beta}{2(1-\beta)}. \tag{42}$$

This shows that the zero-field static susceptibility diverges at $\beta_c=1$, with a power-law behavior

$$\chi \sim (\beta_c-\beta)^{-\gamma},$$

where the exponent $\gamma=1$. Since we are measuring temperatures in units of $2J$, the critical temperature turns out to be $T_c=2J/k_B$. This value is exactly that obtained in molecular field theory for the particular lattice we are considering. While we do not insist that these values of γ and T_c are necessarily those which a more correct theory would give, our theory does predict critical behavior similar to that found by Stanley and Kaplan for $S>\frac{1}{2}$ from their analysis of the exact high-temperature series expansions for the susceptibility. At $\beta=\beta_c$, we can evaluate how χ diverges as a function of H. Equation (35) combined with Eq. (41) gives

$$\chi = (24/5)H^{-1/6},$$

so that $\lim_{H\to 0}\chi$ goes to infinity as the $\frac{1}{6}$ inverse power of H at the critical temperature β_c.

6. ABSENCE OF SPONTANEOUS MAGNETIZATION

We now demonstrate the absence of spontaneous magnetization in our model by showing that $\lim_{H\to 0}\sigma=0$ for all finite temperatures.

For $\beta\leq 1$, we have shown that

$$\lim_{H\to 0} C = \lim_{H\to 0} \frac{\sigma}{\frac{1}{2}-\mu} = 0.$$

Since μ is bounded, this can happen only if $\lim_{H\to 0}\sigma=0$. For $\beta>1$, we found $\lim_{H\to 0} C\neq 0$. From Eq. (26) and the property of the hyperbolic cotangent that we have used earlier, $\coth x \geq |1/x|$, we obtain

$$\frac{1}{\sigma} \geq \frac{2}{\beta C(2\pi)^2} \iint_{-\pi}^{\pi} \frac{d^2k}{1+H/C-\frac{1}{2}\cos k_1-\frac{1}{2}\cos k_2}.$$

Using Eq. (31), this becomes

$$\frac{1}{\sigma} \geq \frac{2}{\pi\beta C} K\left(\frac{1}{1+H/C}\right)\frac{1}{1+H/C}.$$

In the limit of small H, this reduces to

$$\sigma \leq \frac{C}{T} \times \frac{\text{const}}{|\ln H|}, \tag{43}$$

since $\lim_{H\to 0} C\neq 0$. This expression can be compared with the corresponding result of Wagner and Mermin[3] who found, rigorously, that for a two-dimensional lattice

$$\sigma < \frac{\text{const}}{T^{\frac{1}{2}}} \times \frac{1}{|\ln H|^{\frac{1}{2}}}.$$

Our result is quite consistent with that of Mermin and Wagner and is, in fact, somewhat stronger as far as the upper bound on the spontaneous magnetization is concerned.

7. ANALYSIS OF LOW-TEMPERATURE PHASE

In the low-temperature, $T<T_c$, phase, $\lim_{H\to 0}\mu$ has the value

$$\lim_{H\to 0}\mu=\tfrac{1}{2}.$$

This can be obtained either by explicit calculation from Eq. (27) or from the fact that to satisfy both

$$\lim_{H\to 0}C\neq 0$$

and

$$\lim_{H\to 0}\sigma=0,$$

we must have

$$\lim_{H\to 0}\mu=\tfrac{1}{2}.$$

Since this is the maximum value that μ can have, the transverse (x and y) components of the nearest-neighbor spins appear to become perfectly correlated at low temperatures as the external field (in the z direction) is turned off. To investigate further, we look at the ratio of μ to the correlation function of arbitrarily separated sites, $\langle S_i^- S_j^+\rangle$. Using Eqs. (24) and (25), we find

$$\frac{\langle S_i^- S_j^+\rangle}{\mu}=\int\int_{-\pi}^{\pi} d^2k\cos(k_1N_1+k_2N_2)\coth\tfrac{1}{2}\beta[H+E(\mathbf{k})]\Big/\int\int_{-\pi}^{\pi} d^2k\cos k_1\coth\tfrac{1}{2}\beta[H+E(\mathbf{k})]. \tag{44}$$

We have eliminated the lattice spacing by a transformation which replaces k by ka. We have also used the fact that the spatial separation r_i-r_j can be written as $N_1a\hat{e}_x+N_2a\,\hat{e}_y$, where $\hat{e}_x$ and $\hat{e}_y$ are unit vectors in the x and y directions. N_1 and N_2 are integers so that $\mathbf{k}\cdot(r_i-r_j)=k_1N_1a+k_2N_2a$. We calculate the right-hand side of Eq. (44) in two limits.

First, if H is finite and N_1 and/or N_2 tends to infinity, the right-hand side is zero. This can be made mathematically precise but intuitively follows from the observation that as long as H is finite, the denominator integral is finite, and the oscillation of the $\cos(k_1N_1+k_2N_2)$ factor in the numerator integral, for N_1 and/or N_2 large, makes this integral zero.

The other limit is that in which N_1 and N_2 are finite and H tends to zero. To analyze this case, we first replace $\cos(k_1N_1+k_2N_2)$ in the numerator integral by $\cos k_1N_1\cos k_2N_2$. We may do this since

$$\int\int_{-\pi}^{\pi} dk_1dk_2\sin k_1N_1\sin k_2N_2\, f(k_1,k_2)=0,$$

where $f(k_1,k_2)$ is any even function of k_1 and k_2. A further manipulation gives

$$\frac{\langle S_i^- S_j^+\rangle}{\mu}=1+\frac{\mathfrak{N}(H)}{\mathfrak{D}(H)},$$

where

$$\mathfrak{N}(H)=\int\int_{-\pi}^{\pi} dk_1dk_2[\cos k_1N_1\cos k_2N_2-\cos k_1]\times\coth\tfrac{1}{2}\beta[H+E(\mathbf{k})]$$

and $\mathfrak{D}(H)$ is the denominator integral in Eq. (44). It is shown in Appendix B that

$$\lim_{H\to 0}\mathfrak{N}(H)<\infty,$$

and since

$$\lim_{H\to 0}\mathfrak{D}(H)=\infty$$

we get, for finite N_1 and N_2,

$$\lim_{H\to 0}\frac{\langle S_i^- S_j^+\rangle}{\mu}=1.$$

These results show that the transverse correlation function $\langle S_i^- S_j^+\rangle$ goes to zero as the separation $|\mathbf{i}-\mathbf{j}|$ becomes infinite, as long as the external field is finite, but, if the external field is turned off, $\langle S_i^- S_j^+\rangle$ has its maximum value for all finite separations.

The fact that the limits $|\mathbf{i}-\mathbf{j}|\to\infty$ and $H\to 0$ are not commutative implies an important difference between the low-temperature phase in the present system, as compared to the condensed phase of the usual three-dimensional ferromagnet. In the latter case, the long-range correlation existing between spins at widely (even infinitely) separated lattice sites is unaffected by the order in which the above limits are taken. To get a more detailed idea of the spatial dependence underlying the limiting results

$$\text{finite } H,\qquad \lim_{|r_i-r_j|\to\infty}\langle S_i^- S_j^+\rangle=0, \tag{45}$$

$$\text{finite } |r_i-r_j|,\qquad \lim_{H\to 0}\langle S_i^- S_j^+\rangle=\tfrac{1}{2}, \tag{46}$$

we approximately analyze the behavior of $\langle S_i^- S_j^+\rangle$ for large separation. The expression we must evaluate is again

$$\langle S_i^- S_j^+ \rangle = \frac{1}{2}\int\!\!\int_{-\pi}^{\pi} dk_1 dk_2 e^{ik\cdot[(r_i-r_j)/a]} \coth\tfrac{1}{2}\beta[H+C(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)] \Big/ \int\!\!\int_{-\pi}^{\pi} dk_1 dk_2 \coth\tfrac{1}{2}\beta[H+C(1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2)].$$

We will assume that for very large separation the integrals will be dominated by the small momenta behavior of the integrands so that we may approximate the first Brillouin zone by a circle and expand the integrands to get approximately

$$\langle S_i^- S_j^+ \rangle \simeq \left[\frac{4\pi}{1+H/C} K\left(\frac{1}{1+H/C}\right)\right]^{-1} \times \int_0^{2\pi}\int_0^{\pi A} \frac{e^{ikr\cos\theta} k dk d\theta}{H/C+k^2},$$

where $r=(1/a)|r_i-r_j|$ and A is a number on the order of unity whose precise value is unimportant. The integral representation of the Bessel function,[16]

$$J_0(x)=\frac{1}{2\pi}\int e^{ix\cos y}dy,$$

allows us to perform the θ integration and we get, after a change of integration variable,

$$\langle S_i^- S_j^+ \rangle \simeq \left[\frac{1}{1+H/C} K\left(\frac{1}{1+H/C}\right)\right]^{-1} \int_0^{\pi A r} \frac{xJ_0(x)dx}{r^2H/C+x^2},$$

which may be rewritten as

$$\langle S_i^- S_j^+ \rangle \simeq \left[\frac{1}{1+H/C} K\left(\frac{1}{1+H/C}\right)\right]^{-1} \times \{K_0(r\surd(H/C)) - D(r, r\surd(H/C))\}, \quad (47)$$

where $K_0(z)$ is the modified Bessel function defined by[17]

$$K_0(z)=\int_0^\infty \frac{xJ_0(x)dx}{z^2+x^2}$$

and

$$D(r, r\surd(H/C)) = \int_{\pi A r}^\infty \frac{xJ_0(x)dx}{r^2H/C+x^2}.$$

We showed previously that for some fixed, small field H the transverse correlation is zero for infinite separation. As the separation decreases, the correlation increases, becoming equal to $\frac{1}{2}$ for nearest-neighbor distances, in our approximation, and very close to that value over some finite range. The somewhat complicated expression in (47) shows how this correlation begins to grow as we decrease the separation distance from infinity. There is the coefficient

$$[(1+H/C)^{-1}K(1+H/C)^{-1}]^{-1},$$

which plays no role in the spatial dependence. The r dependence lies in the two terms within the bracket, $K_0(r\surd(H/C))$ and $D(r, r\surd(H/C))$. So long as

$$H/C \ll \pi^2 A^2,$$

a condition easily met by the small field we are considering (and may be taken as what we mean by small field in the present context), $D(r, r\surd(H/C))$ is an oscillating term tending to zero as $r\to\infty$ and essentially independent of H. More important is the term $K_0(r\surd(H/C))$ which, as we decrease r from infinity will begin to increase like

$$e^{-r\surd(H/C)}/r\surd(H/C).$$

Therefore, except for the field-independent oscillating term, the growth of the correlation as r decreases from infinity, for fixed field, is of exponential form with the characteristic parameter being

$$(H/C)^{1/2}|r_i-r_j|.$$

This approximate derivation provides some insight into the difference between the condensed phase of three-dimensional ferromagnets which possess long-range order, in the sense that correlations between spins are of truly infinite range, and our two-dimensional system which possesses what may be called quasi-off-diagonal long-range order (QODLRO). QODLRO may occur in those systems[18] where the correlation range (on which the notion of ordering is based) is related to the external field H in such a way that one obtains different results [as exemplified by Eqs. (45) and (46)] depending on whether one takes the separation between the spins to tend to infinity or the external field to tend to zero, first. The three-dimensional Heisenberg ferromagnet, for example, which one as-

[16] *Handbook of Mathematical Functions*, edited by Milton Abramovitz and I. A. Stegun (Dover Publications, Inc., New York, 1965).

[17] G. N. Watson, *Theory of Bessel Functions* (Cambridge University Press, New York, 1958).

[18] A generalized model of a Bose system with similar properties is discussed by M. D. Girardeau, J. Math. Phys. 6, 1083 (1965).

sumes possesses conventional long-range order at sufficiently low temperatures is characterized by the following properties[19]:

Let N be the total number of spins and H the external field. Then, for $T<T_c$, $N\to\infty$, $H\to 0$:

(1) If $NH\to$ constant, then

$$\lim_{|r_i-r_j|\to\infty}\langle S_i^-S_j^+\rangle\neq 0.$$

(2) If $NH\to\infty$, then

$$\lim_{|r_i-r_j|\to\infty}\langle S_i^-S_j^+\rangle=0.$$

In both cases,

$$\lim_{|r_i-r_j|\to\infty}\langle S_i^zS_j^z\rangle\neq 0.$$

These facts show that, in three-dimensions, if the external field H is of $O(1/N)$ [or of $O(r^{-d})$, r being a length and d the dimensionality of the system], the correlation functions, both longitudinal and transverse, become essentially independent of H and also of N at sufficiently low temperatures. In an analogous fashion, if our two-dimensional system was conventionally ordered, a field H of $O(1/r^2)$ would display the independence of the spin correlation range (in our particular model, the range of the transverse correlation function $\langle S_i^-S_j^+\rangle$) on r in the limit of large r. To check this we substitute $H\sim 1/r^2$ in expression (47) for the transverse correlation function to obtain

$$\langle S_i^-S_j^+\rangle\sim\frac{1}{|\ln r|},\quad r=|r_i-r_j|$$

for large r. This shows explicitly that long-range order of the conventional kind is not present in our theory. A field of strength $1/N$ is not sufficient to produce order over the entire sample. However, the correlation function drops off very slowly with distance when this field is present, and it is not surprising that we find essentially perfect correlation at all finite distances when we take H to be strictly zero.

We cannot make a similar statement about the range of the longitudinal correlation function $\langle S_i^zS_j^z\rangle$ since we are not able to calculate it unambiguously in our simple decoupling approximation scheme. Our approximation involves decoupling the first higher-order Green function. However, $\langle S_i^zS_j^z\rangle$ contains $\langle S_i^-S_i^+S_j^-S_j^+\rangle$ and to determine this correlation function unambiguously we would have to go to the next higher-order Green function. We expect, nevertheless, on physical grounds that for an external field H of $O(1/N)$ it would display the same behavior as the transverse correlation function.

Though the low-temperature phase in our model has the appealing features of possessing neither spontaneous magnetization (which would contradict the rigorous results of Mermin and Wagner) nor long-range order of the conventional type, it nevertheless suffers from one shortcoming. The magnitude of the QODLRO as given by Eq. (46), viz.,

$$|r_i-r_j|\ \text{finite},\quad \lim_{H\to 0}\langle S_i^-S_j^+\rangle=\tfrac{1}{2}$$

is larger than an upper limit set by a rigorous inequality[20] derived from spin kinematics. According to this inequality (derived in Appendix C)

$$(\langle S_i^-S_j^+\rangle)_{\text{av}}\leq\frac{N}{4(N-1)},\quad i\neq j \tag{48}$$

for a system of N spins of magnitude $S=\frac{1}{2}$, where the notation $(\ )_{\text{av}}$ denotes the average value of the correlation function $\langle S_i^-S_j^+\rangle$ for $i\neq j$ over the N-spin system. For $N>2$, the magnitude of $\lim_{H\to 0}\langle S_i^-S_j^+\rangle$ clearly violates (48).

In view of the fact that every Green-function approximation scheme undoubtedly violates some exact constraints, it is not surprising that some such violation occurs in our approximation. However, as we presently show, neither the divergence of the static susceptibility nor the absence of spontaneous magnetization in our theory are crucially dependent on the magnitude of $\langle S_i^-S_j^+\rangle$ at finite separation in strictly zero field in the low-temperature region. We can then regard the violation as pointing more to the approximate character of the theory, rather than invalidating the main results derived from it. We emphasize that the nature of the low-temperature order as embodied in Eq. (47) is not inconsistent with inequality (53). The same sort of QODLRO with a smaller magnitude for the correlation would be completely satisfactory with respect to any rigorous test we can devise.

The absence of spontaneous magnetization is basically related to the large fluctuations associated with the long-wavelength modes, and is quite independent of the precise magnitude of the spin correlations at finite distances. An understanding of the divergence of χ is obtained by analyzing the behavior of the effective field parameter C in our theory as we approach the critical point from above. Referring to the section on the determination of C we have the following results:

$$T>T_c:\quad \lim_{H\to 0}\frac{H}{C}\neq 0,$$

$$\lim_{H\to 0}C=0;$$

$$T=T_c:\quad \lim_{H\to 0}C\sim H^{\frac{1}{3}}.$$

The change in the behavior of C from a linear to an approximate power-law behavior with an infinite slope

[19] S. P. Heims, Phys. Rev. Letters 14, 850 (1965).

[20] Dr. V. Korenman (private communication).

at $H=0$ as we approach $T=T_c$ from the high-temperature side is responsible for the divergence of χ. That $\mu=\frac{1}{2}$ for $T<T_c$ at $H=0$ is not crucial here. It is only for $T<T_c$ when $\lim_{H\to0} C\neq 0$ that $\lim_{H\to0}\mu$ becomes equal to $\frac{1}{2}$ in our approximate theory. The χ divergence does not depend on the value that $\lim_{H\to0}\mu$ and $\lim_{H\to0}\langle S_i^- S_j^+\rangle$ acquire at lower temperatures.

The behavior of the effective field parameter and the role it plays in the divergence of the susceptibility also clarifies the difference between our decoupling and the RPA. In the RPA, the effective field parameter is the magnetization σ and if we carry out an analysis similar to the one used for determining C, we arrive at the following result: For all $T>0$,

$$\lim_{H\to 0}\frac{H}{\sigma}\neq 0.$$

This implies that for all nonzero temperatures the response of the RPA effective field parameter to the external field is linear. In other words, the two-dimensional lattice remains paramagnetic in the RPA, so the susceptibility cannot diverge at any finite temperature.

8. SPECIFIC HEAT AT LOW TEMPERATURES

Recently, Miedema *et al.* have reported some measurements[9] on the heat capacity at low temperatures of (spin one-half) ferromagnetic $Cu(C_2H_5NH_3)_2Cl_4$ and $Cu(CH_3NH_3)_2Cl_4$. These compounds have crystal structures which can perhaps be regarded, from a magnetic point of view, as being two dimensional since the interactions between the magnetic copper atoms within a layer are much stronger than the interactions between copper atoms in different layers. For both compounds, a plot of the specific heat versus temperature reveals two transition temperatures. At temperatures below 4°K, the results are described by a dominant linear dependence of the specific heat on temperature. Miedema *et al.* identify the higher of the transition temperatures with the predictions of Stanley and Kaplan, while they attribute the lower one to possible anisotropy or departure from two dimensionality, either of which could conceivably lead to ferromagnetic ordering of the usual kind. As pointed out in Ref. 9, a dominant linear dependence on temperature of the specific heat strongly supports the idea of a two-dimensional ferromagnet. However, in an elementary spin-wave treatment of the problem, one needs an anisotropic interaction to achieve a linear dependence of the specific heat on temperature and anisotropy leads, of course, to ferromagnetic ordering. In our treatment, as we shall presently show, we obtain, at sufficiently low temperatures, a linear dependence of the specific heat on temperature, even for the completely isotropic Heisenberg model without, of course, ferromagnetic ordering of the conventional kind. Thus, there is no need to assume anisotropy to explain the low-temperature portion of the heat-capacity curve, since we can explain this result even for the completely isotropic Heisenberg interaction. Of course, the presence of two transition temperatures in the experiments of Ref. 9 may, in fact, indicate the presence of anisotropy which leads to ferromagnetic ordering at a lower temperature. What we point out, however, is that the linear behavior of the specific heat is not a sufficient condition that ferromagnetic ordering, caused by anisotropy, or departure from two-dimensionality exist, for even the completely isotropic interaction gives the same result at sufficiently low temperatures.

To calculate the specific heat, we need the internal energy per spin, which is given by

$$U=\frac{\langle\mathcal{H}\rangle}{N}=-\frac{gH}{N}\langle\sum_i S_i^z\rangle-\frac{1}{N}\sum_f\sum_g J(\mathbf{f}-\mathbf{g})\langle\mathbf{S}_f\cdot\mathbf{S}_g\rangle.$$

By standard techniques of Green-function analysis,[21] this can be written in the form

$$\begin{aligned}U=&-\tfrac{1}{2}gH-\tfrac{1}{4}J(0)+(gH+\tfrac{1}{2}J(0))\frac{1}{N}\sum_f\int\tau_{ff}(\omega)d\omega\\&+\frac{1}{2N}\sum_f\int\omega\tau_{ff}(\omega)d\omega\\&-\tfrac{1}{2}\sum_f\sum_g J(f-g)\int\tau_{fg}(\omega)d\omega,\quad(49)\end{aligned}$$

where

$$\tau_{fg}(\omega)=\frac{1}{N}\sum_k e^{i\mathbf{k}\cdot(\mathbf{f}-\mathbf{g})}\tau(\mathbf{k},\omega)$$

and

$$\tau(\mathbf{k},\omega)=\lim_{\epsilon\to0} i\frac{G(\mathbf{k},\omega+i\epsilon)-G(\mathbf{k},\omega-i\epsilon)}{e^{\beta\omega}-1}.$$

Combining Eq. (49) with Eqs. (17), (18), (20), and (28) we obtain

$$\begin{aligned}U=&\tfrac{1}{4}gH-\langle S^z\rangle[\tfrac{3}{2}gH+\tfrac{1}{2}J(0)]\\&+C[\tfrac{1}{8}J(0)-\tfrac{1}{4}J(0)\langle S^z\rangle-\tfrac{1}{2}J(0)\mu]-\mu J(0).\quad(50)\end{aligned}$$

The specific heat at constant field, C_H, is given by $C_H=(\partial U/\partial T)_H$. In particular, for $H=0$, we have from Eq. (50), on remembering that

$$\lim_{H\to0}\langle S^z\rangle=0\quad\text{and}\quad\lim_{H\to0}\mu(T<T_c)=\tfrac{1}{2},$$

$$C_{H=0}(T)=-\tfrac{1}{8}J(0)\frac{\partial}{\partial T}\lim_{H\to0}C.\quad(51)$$

$C(T)$ at low temperatures, $T\ll T_c$, is calculated in Appendix B using techniques appropriate to a low-temperature expansion in powers of (T/T_c). Referring to Eq. (B6), we then have for the specific heat

$$C_H=aT+bT^2+\cdots,$$

[21] See, e.g., S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Plenum Press, Inc., New York, 1967).

a dominant linear dependence on temperature with

$$a=\frac{\zeta(2)}{4\pi}\frac{k_B^2}{J}\ \mathrm{erg}/(°\mathrm{K})^2=0.13\frac{k_B^2}{J}\ \mathrm{erg}/(°\mathrm{K})^2$$

and

$$b=\frac{3\zeta(3)}{32\pi}\frac{k_B^3}{J^2}\ \mathrm{erg}/(°\mathrm{K})^3=0.04\frac{k_B^3}{J^2}\ \mathrm{erg}/(°\mathrm{K})^3.$$

9. CONCLUSION

Our theory is not exact, and if a phase transition does exist in the two-dimensional Heisenberg ferromagnet a detailed and correct description of the low-temperature phase will have to await more elaborate approximations. However, simple as it is, our model does represent a first step toward providing some insight into phase transitions which, unlike those in three-dimensional systems, do not arise from a broken symmetry.

An alternative description[1,22] of the two-dimensional spin system, which would fulfill the requirements of a divergent susceptibility and no spontaneous magnetization, has been proposed along the following lines: As long as $\sum_r \langle \mathbf{S}_0\cdot\mathbf{S}_r\rangle$ diverges, the susceptibility will also diverge and, therefore, a $\langle \mathbf{S}_0\cdot\mathbf{S}_r\rangle$ function which behaves like $r^{-\lambda}$ with $\lambda\leq 2$ at large distances will be sufficient to guarantee a divergent susceptibility. Dyson[22] went on to actually make an heuristic derivation of this asymptotic behavior, thereby obtaining an explicit expression for λ. While it is attractive to speculate that this kind of power-law dependence for the correlation function sets in at low temperatures, our theory suggests a different form for the correlation in the system which is consistent with a divergent susceptibility and no spontaneous magnetization. This is the fact that the two-dimensional lattice may possess quasi-off-diagonal long-range order of the kind we have analyzed earlier, and which is suggested by our theory. QODLRO would mean that there is much larger correlation in the spin system than is allowed for in the suggestion of Ref. 22, where the correlations would die down rather rapidly as the spin sites are separated. In view of the fact that the two-dimensional Heisenberg spin system is on the edge of being spontaneously magnetized (the upper limit for $\langle S^z\rangle$, $1/(\ln H)^{\frac{1}{2}}$, goes very slowly to zero, and for $H\sim 1/N$, the upper limit is only about $\frac{1}{10}$ for a finite system), it is not unreasonable to suppose that the correlations between the spins are rather large at low temperatures. QODLRO may then reflect the fact that the low-temperature phase possesses a large total spin, which serves to minimize the exchange energy, and zero $\langle S^z\rangle$ rather than a large total spin and finite or maximum $\langle S^z\rangle$ which is forbidden by the large fluctuations associated with a low dimensionality.

The shortcoming of our calculation lies in the fact that the transverse correlation $\langle S_i^xS_j^x\rangle+\langle S_i^yS_j^y\rangle$ is larger than an upper limit set by spin kinematics. However, as we have pointed out previously, every calculation made on the basis of a Green-function approximation scheme will undoubtedly violate some exact constraints. What has to be considered is whether the violations invalidate the principal results derived in the particular approximation scheme or whether they point only to the approximate character of the theory while leaving its main conclusions substantially unaltered. As we have analyzed earlier, neither of the two principal results of our theory, the divergence of the static susceptibility or the absence of spontaneous magnetization, are crucially linked to the precise value that the transverse correlation function attains in the low-temperature phase as the external magnetic field is completely switched off. We can thus justifiably regard the violation of the inequality as being a shortcoming, which a better approximation should eliminate, but not a drastic enough one to invalidate our results concerning the existence of the transition and the qualitative nature of the low-temperature phase.

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APPENDIX A

We wish to show that $\mathfrak{N}(H)$, given by

$$\mathfrak{N}(H)=\int\!\!\int_{-\pi}^{\pi} dk_1dk_2 \times\{\cos k_1N_1\cos k_2N_2-\cos k_1]\coth\tfrac{1}{2}\beta[H+E(\mathbf{k})]\}$$

is bounded for N_1 and/or N_2 finite as H tends to zero. The only possible reason for a divergence of $\lim_{H\to 0}\mathfrak{N}(H)$ would be a divergence of this integral due to the behavior of the integrand for k_1 and k_2 near zero. The analysis is simplified if we first utilize obvious symmetries to write

$$\mathfrak{N}(H)=2\int_0^\pi\int_0^\pi dk_1dk_2 \times[\cos k_1N_1\cos k_2N_2-\cos k_1N_2\cos k_2N_1-\cos k_1-\cos k_2] \times\coth\tfrac{1}{2}\beta[H+E(\mathbf{k})].$$

For fixed N_1 and N_2 the integrand behaves ultimately according to the obvious small k_1, k_2 expansions so that

[22] F. J. Dyson, lecture given at the Brandeis Summer Institute for Theoretical Physics, 1966 (unpublished).

we get

$$\mathfrak{N}(H)=\int_0^\pi\int_0^\pi dk_1dk_2I(k_1,k_2,H),$$

where for k_1, $k_2\to 0$, and $H\to 0$

$$I(k_1,k_2,H)\sim\frac{2}{\beta}\frac{(\frac{1}{2}-\frac{1}{2}N_1^2-\frac{1}{2}N_2^2)(k_1^2+k_2^2)}{[H+\frac{1}{4}C(k_1^2+k_2^2)]}.$$

Clearly for H going to zero and C finite (we are in the low-temperature phase), there is no divergence problem of any sort at the zero limits so that

$$\lim_{H\to 0}\mathfrak{N}(H)<\infty.$$

APPENDIX B

To evaluate C as a function of temperature for $T\ll T_c$ we use techniques appropriate to an expansion in powers of T/T_c. Rewriting Eq. (28) as

$$\frac{1}{C}=\frac{1}{2}+\frac{1}{(2\pi)^2}\int_{-\pi}^{\pi}\int\frac{d^2k(1-\cos k_1)}{e^{\beta[H+C\epsilon(k)]}-1}, \tag{B1}$$

making use of the identity

$$\frac{1}{e^x-1}=\sum_{n=1}^{\infty}e^{-nx}$$

and the following integral representation[17] of the Bessel function of pure imaginary argument

$$I_n(x)=\frac{(i)^n}{(2\pi)^n}\int_{-\pi}^{\pi}e^{-x\cos y}\cos ny\,dy,$$

we obtain

$$1/C=\tfrac{1}{2}+\phi, \tag{B2}$$

with

$$\phi=\sum_{n=1}^{\infty}e^{-\beta Hn}[e^{-nx}I_0(nx)]^2-\sum_{n=1}^{\infty}e^{-\beta Hn}[e^{-nx}I_1(nx)][e^{-nx}I_0(nx)] \tag{B3}$$

and $x=\beta C/2$. In the above we have made use of the fact (see Eq. 28) that

$$\epsilon(k)=1-\tfrac{1}{2}\cos k_1-\tfrac{1}{2}\cos k_2.$$

β is in the dimensionless units defined earlier. We put $\tau=1/\beta$ (τ is now the temperature in our units) so that low temperatures means $\tau\to 0$ and C/τ is a large quantity (we remember that $\lim_{H\to 0}C\neq 0$ at low temperatures). The following asymptotic expansions[15] are then convenient:

$$\begin{aligned}e^{-x}I_0(x)&=\frac{1}{(2\pi x)^{\frac{1}{2}}}\left(1+\frac{1}{8x}+\frac{9}{128x^2}+\cdots\right),\\ e^{-x}I_1(x)&=\frac{1}{(2\pi x)^{\frac{1}{2}}}\left(1-\frac{3}{8x}-\frac{15}{128x^2}+\cdots\right).\end{aligned} \tag{B4}$$

Equation (B2) is a coupled equation for C and the most convenient solution is obtained by substituting (B4) in (B3) and then solving iteratively for C to any desired power in τ. In the limit $H\to 0$, the solution correct through $O(\tau^3)$ is

$$\lim_{H\to 0}C=2-\frac{\zeta(2)}{\pi}\tau^2-\frac{\zeta(3)}{2\pi}\tau^3\cdots, \tag{B5}$$

where $\zeta(p)$ is the Riemann zeta function defined by

$$\zeta(p)=\sum_{n=1}^{\infty}\frac{1}{n^p}.$$

Using the fact that the critical temperature $T_c=2J/k_B$ and that β (and τ) is being measured in units of $k_B/2J$, we can rewrite (B5) as

$$\lim_{H\to 0}C=2-\frac{\zeta(2)}{\pi}\left(\frac{T}{T_c}\right)^2-\frac{\zeta(3)}{2\pi}\left(\frac{T}{T_c}\right)^3\cdots\quad\text{for } T\ll T_c. \tag{B6}$$

APPENDIX C

To derive the inequality mentioned in (53), consider a (sub) system of N spins of magnitude $S=\frac{1}{2}$ and compute, in any state, the quantity

$$\begin{aligned}\langle(\sum_{i=1}^{N}S_i)^2\rangle&=\langle S_{\text{tot}}^2\rangle\\ &=\sum_i\langle S_i^2\rangle+\sum_{i\neq j}\sum\langle S_i^zS_j^z\rangle\\ &\quad+\sum_{i\neq j}\sum\langle S_i^-S_j^+\rangle.\end{aligned} \tag{C1}$$

Since $S_{\text{tot}}^2\le\frac{1}{2}N(\frac{1}{2}N+1)$, $S_i^2=\frac{3}{4}$, $(S_i^z)^2=\frac{1}{4}$ for $S=\frac{1}{2}$, and

$$\sum_{i\neq j}\sum\langle S_i^zS_j^z\rangle=\sum_i\langle S_i^z(S_{\text{tot}}^z-S_i^z)\rangle,$$

Eq. (C1) can be reexpressed as

$$\tfrac{1}{2}N(\tfrac{1}{2}N+1)\ge\tfrac{3}{4}N-\tfrac{1}{4}N+\langle(S_{\text{tot}}^z)^2\rangle+N(N-1)(\langle S_i^-S_j^+\rangle)_{\text{av}}, \tag{C2}$$

where the notation $(\ )_{\text{av}}$ denotes the average value of $\langle S_i^-S_j^+\rangle$ over the $N(N-1)$ pair of spins in the system. Since $\langle(S_{\text{tot}}^z)^2\rangle\ge 0$, we then have the inequality

$$(\langle S_i^-S_j^+\rangle)_{\text{av}}\le\frac{N}{4(N-1)}. \tag{C3}$$

It is obvious that a similar inequality holds for $(\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle)_{\text{av}}$, viz.,

$$(\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle)_{\text{av}}\le\frac{N}{4(N-1)}.$$