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A Green Function Approach to the Paramagnetic Phase of a Heisenberg Ferromagnet

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A higher-order decoupling similar to the first-order Bogolyubov–Tyablikov decoupling is used to find various correlation functions in the paramagnetic region for a Heisenberg ferromagnet. Expressions are given for the susceptibility, the nearest-neighbor correlation, and autocorrelation at the critical point as well as at high temperatures.

KEY WORDS: Paramagnetic phase of Heisenberg ferromagnet; Green function methods.

1. INTRODUCTION

It is well known that the so-called double-time thermodynamic Green functions which were used by Bogolyubov and Tyablikov have had great success especially when applied to magnetic problems.[2–7] In the present paper we shall be concerned with the Heisenberg ferromagnet—the subject of a great many of the studies made so far. However, where most other papers have been dealing with either the low-temperature region where spontaneous ordering exists or with the case where a magnetic field was applied, so that magnetic ordering is induced, we shall be investigating the temperature region above the critical point—the paramagnetic region—in zero external magnetic field. This region has come more into the focus of attention because of recent interest in phase transitions and studies of physical behavior at

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both sides of transition temperatures. The results obtained so far for this region can be summarized as follows.[8] From series expansions in the inverse absolute temperature T^{-1} it is found that the susceptibility is positive, vanishes as T^{-1} at infinite temperature, and diverges as $(T - T_C)^{-4/3}$ at the Curie temperature T_C. Approximate methods such as the Bogolyubov–Tyablikov decoupling scheme (called in the following the RPA [random phase approximation] theory) give the correct asymptotic behavior as $T \to \infty$, but a $(T - T_C)^{-2}$ behavior for $T \to T_C$, a behavior also found in the spherical model.[9]

Brout[10] recently suggested that excitations corresponding to short-range order correlations might exist in the paramagnetic region. These excitations would be "quasi-spin-waves" with wavelengths short compared with the correlation length. A theoretical treatment by Beeby and Hubbard[11] found evidence for such quasi-excitations, and Bennett[12] has recently predicted, from considering the generalized susceptibility, propagating excitations at wave vectors k greater than a lower limit k_c, which is proportional to the square root of the inverse susceptibility. The experimental evidence for such quasi-excitations is by now considerable, as well.[13–17]

In the following we shall treat the paramagnetic region by extending the Bogolyubov–Tyablikov theory.[2,3] We introduce the usual retarded and advanced Green functions, $G_r(t, t')$ and $G_a(t, t')$ (we use units where $\hbar = 1$):

$$\underset{a}{G_r}(t, t') \equiv \langle\langle A(t); B(t')\rangle\rangle_{\underset{a}{r}} = \mp i\theta(\pm(t - t'))\langle[A(t), B(t')]\rangle \tag{1}$$

where

$$\theta(t) = 1, \quad t > 0: \qquad \theta(t) = 0, \quad t < 0 \tag{2}$$

$$[A, B] = AB - BA \tag{3}$$

$$A(t) = e^{i\mathcal{H}t}Ae^{-i\mathcal{H}t} \tag{4}$$

$$\langle\cdots\rangle = (1/Z)\,\mathrm{Tr}\{e^{-\beta\mathcal{H}}\cdots\} \tag{5}$$

$$Z = \mathrm{Tr}\{e^{-\beta\mathcal{H}}\} \tag{6}$$

$$\beta = 1/kT \tag{7}$$

(k is Boltzmann's constant) and $\mathcal{H}$ is the Hamiltonian of the system which we choose in the form

$$\mathcal{H} = -\frac{1}{2}\sum_{a,b} J_{ab}(\boldsymbol{S}_a \cdot \boldsymbol{S}_b), \qquad J_{aa} = 0 \tag{8}$$

where a and b label the sites of a lattice of N spins of magnitude S and J_{ab} $(= J_{ba})$ is an exchange energy.

For the following, we need only the following results (for detailed derivations we refer to the literature, e.g., References 4 and 5). If $\langle\langle A; B\rangle\rangle$ (without the time arguments and the subscripts) denote the Fourier transform of the $G(t, t')$,

$$\langle\langle A; B\rangle\rangle = \int_{-\infty}^{+\infty} \langle\langle A(t); B(t')\rangle\rangle\, e^{i\omega(t-t')}\, d(t - t') \tag{9}$$

one finds, first of all, that the $\langle\langle A; B\rangle\rangle$ satisfy the equations of motion

$$\omega\langle\langle A; B\rangle\rangle = \langle [A, B]\rangle + \langle\langle [A, \mathscr{H}]; B\rangle\rangle \tag{10}$$

and the value of the correlation function $\langle B(t')\, A(t)\rangle$, which we will need later, is connected with $\langle\langle A; B\rangle\rangle$ through the relation

$$\langle B(t')\, A(t)\rangle = \frac{1}{2\pi i}\int_{-\infty}^{+\infty} d\omega\, e^{-i\omega(t-t')}\, \frac{\lim_{\epsilon\to 0^+}[\langle\langle A; B\rangle\rangle_{\omega+i\epsilon} - \langle\langle A; B\rangle\rangle_{\omega-i\epsilon}]}{e^{\beta\omega} - 1} \tag{11}$$

In (10) we note the appearance of so-called higher-order Green functions. In the early RPA applications, one began with Green functions such as $\langle\langle S_a^{+}; S_b^{-}\rangle\rangle$ ($S_a^{\pm} = S_a^{x} \pm iS_a^{y}$), which are coupled by the equation of motion to Green functions such as $\langle\langle S_c^{z} S_a^{+}; S_b^{-}\rangle\rangle$. The main assumption of the RPA method was then to put

$$\langle\langle S_c^{z} S_a^{+}; S_b^{-}\rangle\rangle \cong \langle S^{z}\rangle\langle\langle S_a^{+}; S_b^{-}\rangle\rangle \tag{12}$$

The quantity $\langle S^{z}\rangle$ measures the long-range order (magnetization), and as long as it is nonvanishing it should be the main determining factor for the behavior of the system. It is determined self-consistently from the equations of motion for the Green functions (e.g., for the spin-$\frac{1}{2}$ case it is directly related to $\langle S_a^{-} S_a^{+}\rangle$ which is through (11) determined by $\langle\langle S_a^{+}; S_a^{-}\rangle\rangle$).

However, in the paramagnetic phase where there is no external field, $\langle S^{z}\rangle = 0$ and the RPA decoupling (12) ceases to be useful. We must therefore look for a different kind of decoupling, which we shall do in the next section. We shall discuss the results of solving the self-consistent equations in the later sections of the paper.

2. DECOUPLING OF THE EQUATIONS OF MOTION

We consider the following dyadics (indicated by boldface roman type):

$$\mathbf{G}_{fg}^{(1)} = \langle\langle \boldsymbol{S}_f; \boldsymbol{S}_g\rangle\rangle \tag{13}$$

We use the elementary commutation relations for spin operators

$$[S_a^{\,i}, S_b^{\,j}] = i\epsilon_{ijk} S_a^{\,k}\delta_{ab} \tag{14}$$

where ϵ_{ijk} is the antisymmetric third-rank tensor:

$$\left.\begin{aligned} \epsilon_{ijk} &= 1 \quad \text{if } ijk \text{ is an even permutation of } 123 \\ &= -1 \quad \text{if } ijk \text{ is an odd permutation of } 123 \\ &= 0 \quad \text{otherwise} \end{aligned}\right\} \tag{15}$$

and where i, j, and k are Cartesian coordinates.

From (8) and (10) we then find the equations of motion

$$\omega\langle\langle \boldsymbol{S}_f; \boldsymbol{S}_g\rangle\rangle = \langle [\boldsymbol{S}_f, \boldsymbol{S}_g]\rangle + i\sum_a J_{fa}\langle\langle [\boldsymbol{S}_f \wedge \boldsymbol{S}_a]; \boldsymbol{S}_g\rangle\rangle \tag{16}$$

By taking the (double) scalar product $(e_x + ie_y \cdot \langle\langle S_f); (S_g\rangle\rangle \cdot e_x - ie_y)$, where e_x and e_y are unit vectors along the x and y axes, we find from (16) the usual RPA equation. As we cannot use the RPA decoupling, we must go to the next order and write down the equation of motion for $\langle\langle[S_f \wedge S_h]; S_g\rangle\rangle$, which is

$$\omega\langle\langle[S_f \wedge S_h]; S_g\rangle\rangle = \langle[S_f \wedge S_h], S_g]\rangle + i\sum_a \{J_{fa}\langle\langle[[S_f \wedge S_a] \wedge S_h]; S_g\rangle\rangle + J_{ha}\langle\langle[S_f \wedge [S_h \wedge S_a]]; S_g\rangle\rangle\} \tag{17}$$

We now expand the triple vector products by using a standard identity for classical vectors, in which additional terms appear due to the noncommutativity of the spin operators. The number of new terms depends on the order of operators. For example, we have *both*

$$[[S_f \wedge S_a] \wedge S_h] = S_a(S_f \cdot S_h) - S_f(S_a \cdot S_h) + i[S_f \wedge S_h]\,\delta_{af} \tag{18}$$

and

$$[[S_f \wedge S_a] \wedge S_h] = (S_f \cdot S_h)\,S_a - (S_a \cdot S_h)\,S_f + i[S_f \wedge S_h](\delta_{af} + \delta_{hf}) - i[S_a \wedge S_h]\,\delta_{ah} \tag{19}$$

If we order the operators so as to minimize the number of commutator terms that appear, and use the fact that $J_{aa} = 0$, we have from (17)

$$\omega\langle\langle[S_f \wedge S_h]; S_g\rangle\rangle = \langle[[S_f \wedge S_h], S_g]\rangle + i\sum_a \{J_{fa}\{\langle\langle S_a(S_f \cdot S_h); S_g\rangle\rangle - \langle\langle S_f(S_a \cdot S_h); S_g\rangle\rangle\} - J_{ha}\{\langle\langle(S_f \cdot S_a)\,S_h; S_g\rangle\rangle - \langle\langle(S_f \cdot S_h)\,S_a; S_g\rangle\rangle\}\} \tag{20}$$

The commutator terms do not appear at all in this equation, and thus we may say that it expresses a sort of semiclassical approximations. If we expand the triple vector product so that the extra spin operator always stands to the right or the left of the scalar product, we find an equation of motion with an additional term, $\pm J_{fh}\langle\langle[S_f \wedge S_h]; S_g\rangle\rangle$, which obviously alters the results considerably. (The negative sign comes from using (19); the positive sign from using the corresponding equation for the other triple product.) There seems no way *a priori* of deciding which sign is correct, and in any case, application of our decoupling to (20) *with* the extra term leads to results quite unlike those known for the Heisenberg paramagnet. One way of resolving this ambiguity is to choose a symmetrized expansion of the triple product. In this, the additional term vanishes and we are left with (20) again.

We now look for a decoupling to get a closed set of equations. As we are interested in correlations in the paramagnetic region, which means that ultimately we wish to calculate (self-consistently) the correlation functions related to the $\mathbf{G}^{(1)}_{fg}$, we try the decoupling

$$\langle\langle S_a(S_f \cdot S_h); S_g\rangle\rangle \cong \langle(S_f \cdot S_h)\rangle\langle\langle S_a; S_g\rangle\rangle \tag{21}$$

and so on. We note (i) the similarity between (12) and (19), (ii) that other averages such as $\langle S_a^x S_f^x \rangle$, which we might extract from the operator $\boldsymbol{S}_a(\boldsymbol{S}_f \cdot \boldsymbol{S}_h)$, would cancel terms arising from $\boldsymbol{S}_f(\boldsymbol{S}_a \cdot \boldsymbol{S}_h)$ while averages such as $\langle S_a^x S_f^y \rangle$ vanish in a system where the total z component of spin is conserved, and (iii) that as usual we do not give any proof of the validity of the decoupling, but hope that the results will justify our procedure.

We shall use (19) and the short-hand notation (13) and

$$\langle\langle [\boldsymbol{S}_f \wedge \boldsymbol{S}_h]; \boldsymbol{S}_g \rangle\rangle = \mathbf{G}^{(2)}_{fhg} \tag{22}$$

$$\langle (\boldsymbol{S}_a \cdot \boldsymbol{S}_b) \rangle = C_{ab} \tag{23}$$

$$\langle [\boldsymbol{S}_f, \boldsymbol{S}_g] \rangle = \mathbf{I}^{(1)}_{fg} \tag{24}$$

$$\langle [[\boldsymbol{S}_f \wedge \boldsymbol{S}_h], \boldsymbol{S}_g] \rangle = \mathbf{I}^{(2)}_{fhg} \tag{25}$$

where we note that while in (13) and (22) two time arguments occur, in (23)–(25) all time arguments coincide. Equations (16) and (18) now become

$$\omega \mathbf{G}^{(1)}_{fg} = \mathbf{I}^{(1)}_{fg} + i \sum_a J_{fa} \mathbf{G}^{(2)}_{fag} \tag{26}$$

and

$$\omega \mathbf{G}^{(2)}_{fag} = \mathbf{I}^{(2)}_{fag} + i \sum_b \{J_{fb}[C_{fa}\mathbf{G}^{(1)}_{bg} - C_{ba}\mathbf{G}^{(1)}_{fg}] + J_{ab}[C_{fb}\mathbf{G}^{(1)}_{ag} - C_{fa}\mathbf{G}^{(1)}_{bg}]\} \tag{27}$$

As they stand, (26) and (27) can be solved by the usual lattice Fourier transform, based on the translational invariance of the system. Before doing this, we note that we can insert one exact relation into this approximation. In general $(\boldsymbol{S}_a \cdot \boldsymbol{S}_a) = S(S+1) \equiv \Gamma_0$. Now in (27), we always have $a \neq f$ because of (26) and the fact that $J_{aa} = 0$; however, the summed index b can equal a or f and thus $C_{aa} = C_0$ appears in front of the Green functions. We know exactly, from (20) and the above, that these are multiplied by Γ_0, and since therefore C_0 does not appear in the equations, we should replace it by Γ_0. [No additional approximation is introduced here, and in fact failure to include this exact information leads to completely unphysical results, for example, nearest-neighbor correlations proportional to the temperature. This is not surprising, for if Γ_0 is not included, as in (27), nothing in the equations specifies the magnitude of the spin S.] To preserve the symmetry of the equations, however, we find it convenient to perform the replacement by subtracting the term in C_0 and adding that in Γ_0 separately, as follows:

$$\omega \mathbf{G}^{(2)}_{fag} = \mathbf{I}^{(2)}_{fag} + i \sum_b^b \{J_{fb}[C_{fa}\mathbf{G}^{(1)}_{bg} - C_{ba}\mathbf{G}^{(1)}_{fg}] + J_{ab}[C_{fb}\mathbf{G}^{(1)}_{ag} - C_{fa}\mathbf{G}^{(1)}_{bg}]\}$$
$$+ i(C_0 - \Gamma_0) J_{fa}(\mathbf{G}^{(1)}_{fg} - \mathbf{G}^{(1)}_{ag}) \tag{28}$$

The better the decoupling approximation, the smaller the final term in (28). Why, then, is it essential? The reason will become clear when we get to the implicit equation which expresses the self-consistency condition.

We now calculate the Fourier transforms and write

$$\mathbf{G}_{\lambda}^{(1)}(\mathbf{I}_{\lambda}^{(1)}, K_{\lambda}, J_{\lambda}) = \sum_{a-b} e^{i(\lambda \cdot a-b)}\, \mathbf{G}_{ab}^{(1)}(\mathbf{I}_{ab}^{(1)}, C_{ab}, J_{ab}) \tag{29}$$

$$\mathbf{G}_{\lambda\mu}^{(2)}(\mathbf{I}_{\lambda\mu}^{(2)}) = \sum_{a-c,b-c} e^{i(\lambda \cdot a-c)+i(\mu \cdot b-c)}\, \mathbf{G}_{abc}^{(2)}(\mathbf{I}_{abc}^{(2)}) \tag{30}$$

and assume inversion symmetry of the lattice so that, e.g., $J_{\lambda} = J_{-\lambda}$.

Equations (24) and (27) now become

$$\omega \mathbf{G}_{\lambda}^{(1)} = \mathbf{I}_{\lambda}^{(1)} + \frac{i}{N} \sum_{\mu} J_{\mu} \mathbf{G}_{\lambda-\mu,\mu}^{(2)} \tag{31}$$

$$\begin{aligned} \omega \mathbf{G}_{\lambda\upsilon}^{(2)} = \mathbf{I}_{\lambda\upsilon}^{(2)} &+ i[(K_{\upsilon} - K_{\lambda}) J_{\upsilon+\lambda} + K_{\lambda} J_{\lambda} - K_{\upsilon} J_{\upsilon}]\, \mathbf{G}_{\lambda+\upsilon}^{(1)} \\ &+ i(\Gamma_0 - C_0)(J_{\lambda} - J_{\mu})\, \mathbf{G}_{\lambda+\upsilon}^{(1)} \end{aligned} \tag{32}$$

Eliminating $\mathbf{G}_{\lambda\nu}^{(2)}$ and solving for $\mathbf{G}_{\lambda}^{(1)}$, we find

$$\mathbf{G}_{\lambda}^{(1)} = \frac{\omega \mathbf{I}_{\lambda}^{(1)} + \frac{i}{N} \sum_{\mu} J_{\mu} \mathbf{I}_{\lambda-\mu,\mu}^{(2)}}{\omega^2 - \omega_{\lambda}{}^2} \tag{33}$$

where

$$\omega_{\lambda}{}^2 = \frac{1}{N} \sum_{\mu} \{K_{\mu}(J_{\mu} - J_{\lambda}) + (\Gamma_0 - C_0)\, J_{\mu}\}(J_{\mu} - J_{\lambda-\mu}) \tag{34}$$

3. EXCITATION ENERGIES

Equation (33) gives us $\mathbf{G}^{(1)}$ but in it occur the K_{μ} as parameters. We must thus express the K_{μ} by means of (11) in terms of $\mathbf{G}^{(1)}$ and then solve the resultant equation—as is done for $\langle S^z \rangle$ in the RPA theory.

We first note that the $\mathbf{G}^{(1)}$ are dyadics while the K_{μ} are scalars. We must thus use equations such as

$$\langle (\boldsymbol{S}_f \cdot \boldsymbol{S}_g) \rangle = \sum_{k} (\boldsymbol{e}_k \cdot \langle \boldsymbol{S}_f \rangle)(\boldsymbol{S}_g \rangle \cdot \boldsymbol{e}_k) \tag{35}$$

where the $\boldsymbol{e}_k$ ($k = x, y, z$) are unit vectors along the Cartesian axes and where the dots on the right-hand side of (35) indicate scalar products with the vector immediately behind or in front of the unit vector.

From the commutation relations for the spin operators one finds after straightforward calculations

$$\sum_{k} (\boldsymbol{e}_k \cdot \mathbf{I}_{\lambda\upsilon}^{(2)} \cdot \boldsymbol{e}_k) = 2i(K_{\lambda} - K_{\upsilon}) \tag{36}$$

One can now take (33), decompose the right-hand side into partial fractions, and use (11), (35), (36), and the fact that in the K_λ the time arguments coincide to find the implicit equation

$$K_\lambda = \frac{\coth \frac{1}{2}\beta\omega_\lambda}{\omega_\lambda} \frac{1}{N} \sum_\mu K_\mu (J_\mu - J_{\lambda-\mu}) \tag{37}$$

To make the discussion more transparent, we shall restrict ourselves to the case of nearest-neighbor interactions only, so that, if Δ indicates here and henceforth a nearest-neighbor vector,

$$J_{ab} = J\delta_{a,b+\Delta} \tag{38}$$

where we, moreover, have assumed J to be independent of Δ. We introduce the following notation:

$$\gamma_\lambda = \frac{1}{z} \sum_\Delta e^{i(\lambda \cdot \Delta)}, \; u_\lambda = 1 - \gamma_\lambda \tag{39}$$

where z is the number of nearest neighbors. From (39), (38), and (29), it follows that

$$J_\lambda = zJ\gamma_\lambda \tag{40}$$

We also use the helpful identity

$$\frac{1}{N} \sum_\mu F_\mu \gamma_{\lambda-\mu} = \gamma_\lambda \frac{1}{N} \sum_\mu F_\mu \gamma_\mu \tag{41}$$

where F_μ is the lattice Fourier transform of a function F_r whose value is independent of lattice direction for $r = \Delta$. The identity follows from using (39) for $\gamma_{\lambda-\mu}$, inverting the transform of F_μ, using the spherical symmetry of F_Δ, and retransforming to F_μ again.

Using (40) and (41), (37) becomes

$$K_\lambda = \frac{zJC_1}{\omega_\lambda} u_\lambda \coth \tfrac{1}{2}\beta\omega_\lambda \tag{42}$$

where C_1 is the nearest-neighbor correlation function

$$C_1 \equiv C_{a,a+\Delta} = \langle (S_a \cdot S_{a+\Delta}) \rangle \tag{43}$$

which is independent of Δ since both the Heisenberg Hamiltonian and the paramagnetic phase are spherically symmetrical.

Similarly, we can manipulate (34) and find

$$\omega_\lambda^2 = z^2 J^2 C_1 u_\lambda (\chi^{-1} + u_\lambda) \tag{44}$$

where

$$\chi^{-1} = \frac{\Gamma_0 - C_0}{zC_1} + \frac{\sum_{\Delta'} C_{a,a+\Delta+\Delta'} - zC_1}{zC_1} \tag{45}$$

Note that because of the first term, C_0 does not appear in the sum in the second term.

To see the physical meaning of χ we note that for small λ we have $u_\lambda \approx a\lambda^2$ (we use the assumed inversion symmetry), so that as $\lambda \to 0$ we have from (44) and (42)

$$K_\lambda \xrightarrow[\lambda \to 0]{} \frac{2}{\beta z J} \frac{1}{\chi^{-1} + a\lambda^2} \tag{46}$$

the Ornstein–Zernike form[18] for correlation functions near a phase transition. Noting that

$$K_\lambda = \langle | \boldsymbol{S}_\lambda |^2 \rangle \tag{47}$$

and that thermodynamics relates the zero-momentum fluctuations with the static susceptibility χ_{stat} as follows

$$\langle | \boldsymbol{S}_0 |^2 \rangle = \frac{3}{\beta \mu_B{}^2} \chi_{\text{stat}} \tag{48}$$

where μ_B is the Bohr magneton, we see that

$$\chi = \frac{3}{2} \frac{zJ}{\mu_B{}^2} \chi_{\text{stat}} \tag{49}$$

where the factor 3 comes from summing over the three spatial directions. Note that because of (44), χ^{-1} must always be positive for the Green function formalism to go through, since $\chi^{-1} < 0$ means that ω_λ is imaginary for $\boldsymbol{\lambda}$ near the origin. Thus the vanishing of χ^{-1}, which (45) indicates will occur as the temperature is lowered from infinity if the C_{ab} increase as intuitively expected, defines the breakdown of the decoupling approximation. We may interpret this as due to the onset of ferromagnetic ordering: thus the transition temperature T_c for this theory is that at which χ^{-1} vanishes.

For positive χ^{-1}, the poles of the Green function, i.e., what this theory sees as excitations, occur at the energies $\pm\omega_\lambda$ given by (45); ω_λ as a function of $\boldsymbol{\lambda}$ has quite different forms for high and low momenta, if χ^{-1} is not too large:

$$\omega_\lambda \approx zJ(C_1\chi^{-1})^{1/2} u_\lambda^{1/2}, \qquad \chi^{-1} \gg u_\lambda \tag{50}$$

$$\omega_\lambda \approx zJC_1^{1/2} u_\lambda, \qquad \chi^{-1} \ll u_\lambda \tag{51}$$

The changeover occurs at momenta of the order of λ_0, which is defined such that $u_{\lambda_0} = \chi^{-1}$. For χ^{-1} very small, since $u_\lambda \approx a\lambda^2$, we have $\lambda_0 = \sqrt{(\chi^{-1}/a)}$. Note that the high-momentum form (51) has the momentum dependence of spin-waves, which tends to confirm the idea that quasi-spin-waves should appear near the critical point for small but nonzero wave vector, and that λ_0 is proportional to $\chi^{-1/2}$, as in Bennett's theory.[12] In this order, the theory reveals nothing about the damping of such excitations, but it definitely supports the suggestions of their existence. The low-momentum form for ω_λ, (50), is linear in λ for small λ. No such excitation has been suggested so far. It would be interesting to see what the inclusion of damping does to this form.

Using the relation (in fact the definition of C_0, since it is *not* a decoupling parameter)

$$C_0 = \frac{1}{N}\sum_{\lambda} K_\lambda \tag{52}$$

we can write our self-consistent equations (42) and (44) in the form

$$C_1 = \frac{1}{N}\sum_{\lambda} K_\lambda \gamma_\lambda \tag{53}$$

$$C_1(\chi^{-1} + 1) = \frac{1}{N}\sum_{\lambda} K_\lambda \left(\gamma_\lambda{}^2 - \frac{1}{z}\right) + \frac{\Gamma_0}{z} \tag{54}$$

where K_λ satisfies (42) and ω_λ is given by (44).

4. THERMODYNAMIC RESULTS

We can find a formal solution to these equations by expanding $\coth \frac{1}{2}\beta\omega_\lambda$ in an infinite series. This allows us to evaluate the sums over momenta. The expansion is valid for $\frac{1}{2}\beta\omega_\lambda \leqslant \pi$, and since χ^{-1} must increase at most as T at high temperatures, and C_1 should decrease with temperature, the expansion should work for β sufficiently near zero. In fact, the expansion turns out to be good everywhere that the decoupling succeeds. Inserting the expansion[11] in (53), we find

$$C_1 = \frac{1}{N}\sum_{\lambda} \frac{zJC_1 u_\lambda \gamma_\lambda}{\omega_\lambda}\left[\frac{1}{\frac{1}{2}\beta\omega_\lambda} + \sum_{k=1}^{\infty} D_{2k}\left(\frac{1}{2}\beta\omega_\lambda\right)^{2k-1}\right] \tag{55}$$

where $D_{2k} = 2^{2k}B_{2k}/(2k)!$ and the B_{2k} are the Bernoulli numbers.
From (44) and a slight rearrangement, we have

$$C_1 = 2\tau\frac{1}{N}\sum_{\lambda}\frac{\gamma_\lambda}{\chi^{-1} + u_\lambda} + \frac{C_1}{2\tau}\Sigma_1 \tag{56}$$

where $\tau = 1/\beta zJ$ is the reduced temperature and

$$\Sigma_1 = \sum_{k=1}^{\infty} D_{2k}\frac{1}{N}\sum_{\lambda}(\tfrac{1}{2}\beta\omega_\lambda)^{2k-2}\, u_\lambda\gamma_\lambda \tag{57}$$

A corresponding expansion for (54) gives

$$C_1(\chi^{-1} + 1) = 2\tau\frac{1}{N}\sum_{\lambda}\frac{\gamma_\lambda{}^2 - 1/z}{\chi^{-1} + u_\lambda} + \frac{C_1}{2\tau}\Sigma_2 + \frac{\Gamma_0}{z} \tag{58}$$

where

$$\Sigma_2 = \sum_{k=1}^{\infty} D_{2k}\frac{1}{N}\sum_{\lambda}\left(\frac{1}{2}\beta\omega_\lambda\right)^{2k-2} u_\lambda\left(\gamma_\lambda{}^2 - \frac{1}{z}\right) \tag{59}$$

Of course Σ_1 and Σ_2 depend on C_1 and χ^{-1}, so (56) and (58) are still implicit equations. However a useful relation results from treating Σ_1 and Σ_2 as parameters and substituting for C_1 in (58) its value given by (56). In terms of the variable $\psi = \chi^{-1} + 1$, a lengthy rearrangement leads to the implicit equation

$$F_0(\psi) = \frac{(\psi - \Sigma_2/\Sigma_1) + (\Gamma_0/z)(1/\Sigma_1 - 1/2\tau)}{\psi(\psi - \Sigma_2/\Sigma_1) + (2\tau/z)(1/\Sigma_1 - 1/2\tau)} \tag{60}$$

Here

$$F_n(\psi) = \frac{1}{N}\sum_{\lambda} \frac{{\gamma_\lambda}^n}{\psi - {\gamma_\lambda}^n} \tag{61}$$

These functions appear frequently in the theory of ferromagnetism[20] and have been exhaustively discussed by Mannari and Kawabata.[21] In the manipulation leading to (60), we have used the identities

$$\frac{1}{N}\sum_{\lambda} \gamma_\lambda = 0 \tag{62}$$

$$F_2(\psi) = \psi F_1(\psi), \qquad F_1(\psi) = \psi F_0(\psi) - 1 \tag{63}$$

which are easy to derive from the definitions (39) and (61). Although in principle (60) can be solved, since all the functional forms are known, it is still too difficult to work with. But its important features appear if we neglect all terms of $O(1/\tau)$ and higher in the series expansions. Checks confirm that nothing significant is lost by this simplification. For cubic lattices, $F_0(\psi)$ depends on the elliptic integral $K(k^2)$[20,21]; this dependence is simplest for the body-centred cubic structure, and the following discussion will be confined to that case, where we have

$$F_0(\psi) \equiv \frac{1}{\psi} H(\psi) = \frac{1}{\psi}\frac{4}{\pi^2} K^2(k^2) \tag{64}$$

where

$$k^2 = \tfrac{1}{2} - \tfrac{1}{2}\sqrt{1 - 1/\psi^2} \tag{65}$$

Thus, since the first term of Σ_1 equals $1/3z$ and that of Σ_2 vanishes, (60) becomes

$$H(\psi) = \psi(\psi - 3\Gamma_0)/(\psi^2 - 6\tau - z^{-1}) \tag{66}$$

This equation has a single solution for all $\tau \geqslant \tau_c$, where the reduced critical temperature τ_c is

$$\tau_c = \frac{\frac{1}{2}\Gamma_0}{F_0(1)}\left[\frac{3\Gamma_0 + F_0(1)(1 - 1/z) - 1}{3\Gamma_0}\right] \tag{67}$$

As mentioned above, this critical temperature corresponds to the vanishing of χ^{-1}. Note that τ_c is not simply proportional to Γ_0, as in the RPA theory, although the factor in brackets is close to unity. The RPA value,[6] which is numerically very close

to that found in the Bethe–Peierls–Weiss (cluster) theory and by extrapolation of high-temperature series,[21] is

$$\tau_c^{(\mathrm{RPA})} = \frac{\frac{1}{3}\Gamma_0}{F_0(1)} = \frac{k}{zJ}\frac{\theta_c}{F_0(1)} \tag{68}$$

where θ_c is the transition temperature from molecular field theory. Comparison of (67) and (68) shows that in a sense the present theory describes a Heisenberg model with molecular-field transition temperature $\tilde{\theta}_c = \frac{3}{2}\theta_c$. This feature will reappear.

In an exact theory, the sum $C_0 = (1/N)\sum_\lambda K_\lambda$ equals Γ_0: the departure of $C_0 - \Gamma_0$ from zero is one measure of the accuracy of our approximation. At τ_c, we have $C_0 - \Gamma_0$ at most 10% of Γ_0 for spin $\frac{1}{2}$, and much less for higher spin. This observation brings us back to the problem that the success of the decoupling seems to be due to introducing a parameter $(C_0 - \Gamma_0)$ which goes to zero as the decoupling improves. A glance at (60) reveals the reason. The terms in $1/z$ are due to the addition and subtraction of C_0 in (28). If they are removed, the resulting equation can only be solved if one of the following two conditions is met:

(i) $H(\psi) = 1$, which can only be satisfied in the limit $\psi \to \infty$, or

(ii) $\psi - \Sigma_2/\Sigma_1 = 0$, which leads to entirely unreasonable results, as demonstrated in the appendix. But if $C_0 - \Gamma_0$ differs from zero, by however little, the additional terms appear in (60) and the character of the solution is changed entirely: it becomes reasonable, and even agrees rather well with other theories.

Near the critical point, the series expansions for χ and the specific heat C, the most relevant thermodynamic quantities, are as follows:

$$\chi = 1.10\chi_1(\tau - \tau_c)^2 - 1.37\chi_1^2\left[1.73 + \frac{4.95}{3S(S+1)-1}\right](\tau - \tau_c)^3 + O((\tau - \tau_c)^4) \tag{69}$$

$$\frac{C}{\frac{1}{2}Fk} = 0.78 + 2\tau_c\chi_1 + \left[4.92\tau_c\chi_1^2\left\{2.56 + \frac{2.48}{3S(S+1)-1}\right\} - 4\chi_1\right](\tau - \tau_c) + O((\tau - \tau_c)^2) \tag{70}$$

where $\chi_1 = 6F_0/[3S(S+1) - 1]$. The leading term in χ is the quadratic one, as in the spherical model and the RPA theory, and the specific heat C is constant as τ_c is approached from above.

At high temperatures, we expand in powers of $1/T$, and find

$$\chi_{\mathrm{stat}} = [\mu_B^2 S(S+1)/3kT][1 + (\tfrac{3}{2} + \delta)\,\theta_c/T + O(1/T^2)] \tag{71}$$

where δ is very small. The exact high-temperature series[22] starts as follows:

$$\chi_{\mathrm{stat}} = [\mu_B^2 S(S+1)/3kT][1 + \theta_c/T + O(1/T^2)] \tag{72}$$

Here again, the approximation seems to describe a model with a different θ_c, $\tilde{\theta}_c = \frac{3}{2}\theta_c$.

The nearest-neighbor correlation is proportional to T^{-1} and leads to a specific heat of the form

$$C = Nk\,(\tfrac{3}{2})^2\,\theta_c^2/T^2 + O(T^{-3}) \tag{73}$$

where the exact result is

$$C = 3Nk(\theta_c^2/T^2) + O(T^{-3}) \tag{74}$$

Finally, the sum C_0 is

$$C_0 = \Gamma_0 + \tfrac{1}{3}[S(S+1)\,zJ/4kT]^2 + O(T^{-3}) \tag{75}$$

Thus $C_0 - \Gamma_0$ is small at high temperatures as well as near the transition temperature.

5. CONCLUSIONS

We have shown that a simple Green function theory predicts the phase transition of the Heisenberg paramagnet, as well as most of the qualitative thermodynamic behavior, both at high temperatures and near the critical point. Our theory also contains several suggestions as to the microscopic nature of the paramagnet: in particular, it predicts excitations with a spectrum ω_λ given by (44), and correlation functions K_λ given by (42). These results are already supported by a certain amount of evidence, as we have indicated, but neither has been firmly established. Our theory adds weight to the evidence and suggests the way ω_λ and K_λ are related. Furthermore, (45) relates the susceptibility to the correlation functions, and it would be interesting to see whether this relation is accidental or is generally useful.

APPENDIX

Decoupling Without $(C_0 - \Gamma_0)$

Here we will discuss the solutions of the equation

$$\psi = \Sigma_2/\Sigma_1 \tag{A.1}$$

which is the appropriate self-consistent equation in our decoupling if the terms in C_0 are not eliminated from (27). As we showed in Section 4, (A.1) follows from (60) simply by removing the terms in $1/z$. (This reflects nothing profound about z; it has been retained as a handy label.) Note further that the definition of Σ_2 must also be changed, since from (59) it contains terms in $1/z$. The proper quantity for (A.1) is

$$\tilde{\Sigma}_2 = \sum_{k=1}^{\infty} D_{2k}\,\frac{1}{N}\sum_{\lambda}(\tfrac{1}{2}\beta\omega_\lambda)^{2k-2}\,u_\lambda\gamma_\lambda^2 \tag{A.2}$$

The natural way to attempt a solution to (A.1) is the series-expansion approach of Section 4. This must work at sufficiently high temperatures if the solution is to be

physically reasonable, so that if it fails, we automatically know that (A.1) is not the right equation. Quick evaluation of the leading terms in Σ_1 and $\tilde{\Sigma}_2$ gives

$$\psi = \frac{D_2/z + O(T^{-1})}{-D_2/z + O(T^{-1})} \tag{A.3}$$

where now the factor $1/z$ comes from the sums $(1/N)\sum_\lambda u_\lambda^n$, not from the term in $C_0 - \Gamma_0$. We quickly see that

$$\psi \equiv \chi^{-1} + 1 = -1 + O(T^{-1}) \tag{A.4}$$

This implies a negative susceptibility at high temperatures, and from (56) a nearest-neighbor correlation C_1 which is negative and increasing with T. Thus (A.1) leads to entirely unacceptable results, and we may therefore reject it.

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