PHYSICAL REVIEW B VOLUME 8, NUMBER 5 1 SEPTEMBER 1973

# Second-Order Green's-Function Theory of the Infinite-Chain Heisenberg Ferromagnet*

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(Received 21 March 1973)

A second-order self-consistent random-phase-approximation decoupling similar to that used by Kondo and Yamaji for spin-1/2 and by Scales and Gersch for the antiferromagnet is applied to the isotropic infinite-chain Heisenberg ferromagnet for arbitrary spin in zero applied field. At finite temperatures, well-defined spin waves and a finite correlation length are found. In the zero-temperature limit, the correlation length becomes infinite and the exact ground state and spin-wave spectrum are obtained. The correlation functions are isotropic, as required in the absence of magnetization, and are asymptotically exact in the high-temperature limit. Results are compared with extrapolations from finite chains by Bonner and Fisher for spin-1/2 and by Weng for spin 1, and with the rigorous solution by Fisher for the classical infinite-spin limit.

## I. INTRODUCTION

The infinite-chain Heisenberg magnet has been of interest to theorists[1] for some time. In the absence of an applied magnetic field, the Heisenberg Hamiltonian for a chain of $N$ atoms is given by

$$H = -(\tfrac{1}{2}J)\sum_{j=0}^{N-1}\sum_{d=\pm 1}\vec{S}_j\cdot\vec{S}_{j+d}, \qquad (1.1)$$

where $J$ is the exchange constant, $\vec{S}_j$ is the spin operator at site $j$, $d$ is the nearest-neighbor index, and where periodic boundary conditions may be applied to require $S_N = S_0$, since the limit $N\to\infty$ is to be taken. The case $J>0$ is the ferromagnet and the case $J<0$ is the antiferromagnet. Theoretical interest in linear chains has been heightened by recent experiments which show that a number of materials behave as linear-chain ferromagnets[2-4] and antiferromagnets[5] to a first approximation.

The only exact solution for the specific heat and other thermostatic quantities of the Hamiltonian given by Eq. (1.1) for arbitrary temperature is that obtained by Fisher[6] in the classical infinite-spin limit. Some of the features of his solution are that magnetization[7] can exist only at zero temperature, that the correlation functions are spatially isotropic, as required in the absence of magnetization, and that the correlation length for both the ferromagnet and antiferromagnet at low temperature varies inversely with temperature. Mermin and Wagner[8] have proven that the infinite-chain Heisenberg model cannot support magnetization[7] at nonzero temperature in the absence of an applied magnetic field. Hutchings *et al.*[5] have found that the substance $(CH_3)_4NMnCl_3$ (TMMC), a linear-chain Heisenberg antiferromagnet of spin-$\frac{5}{2}$, has a correlation length which varies inversely with temperature over a considerable temperature range and has a spin-wave spectrum which is well defined at low temperature. Steiner and Dachs[3] have found that $CsNiF_3$, a linear-chain ferromagnet, has a rather large correlation length at low temperatures that decreases with an increase in temperature. These results lead one to expect that, even though the one-dimensional ferromagnet cannot support magnetization at nonzero temperature, the spins remain highly correlated over a considerable distance at low temperature, so that spin waves whose wavelengths are short compared to the correlation length exist, at least locally. On the other hand, spin waves whose wavelengths are long compared to the correlation length may possibly not exist, and if they do exist, may have greatly renormalized excitation energies in comparison with short-wavelength spin waves.

Extrapolations from finite-chain Heisenberg ferromagnets by Bonner and Fisher[9] for spin-$\frac{1}{2}$ and by Weng[10] for spin 1 yield estimates for the specific heat and other thermostatic quantities which become rather uncertain at low temperature and shed no light on the behavior of the infinite-chain correlation length in this temperature region. Neither ordinary renormalized-spin-wave theory[1] nor the first-order self-consistent random-phase-approximation (RPA) Green's-function theory of Tyablikov[11] can be applied, because the temperature renormalization of the spin-wave excitation energy in these theories is independent of wavelength and proportional to the magnetization, which vanishes in absence of an applied field.

Self-consistent RPA decouplings of the Green's-function equations of motion at second order, rather than first, have been used by Kondo and Yamaji[12] for the spin-$\frac{1}{2}$ infinite chain and by Scales and Gersch[13,14] for arbitrary spin for one-, two-, and three-dimensional lattices. These decouplings lead to spin waves with excitation energies whose temperature renormalization is greater for longer wavelengths and proportional to the correlation functions, so that the existence of spin waves does not depend on having magnetization. Scales and Gersch[13] have also shown that self-consistent second-order RPA Green's-function theories obey the principle of detailed balance and satisfy zeroth-, first-, and second-moment relaxation-function sum rules for the spin-wave excitation energy. The ferromagnetic chain cannot be treated by the decoupling method of Ref. 13 without modification, since the proper ground state would not be found in the zero-temperature limit. In the present paper we construct a self-consistent second-order RPA Green's-function theory for the infinite-chain Heisenberg ferromagnet in zero applied field for arbitrary temperature and arbitrary spin, which yields the proper ground state in the zero-temperature limit, gives a correlation length which varies inversely with temperature at low temperature, and has the required spatial isotropy in the correlation functions in the absence of magnetization.

## II. GREEN'S FUNCTION FORMALISM

The retarded double-time temperature-dependent Green's function for operators $A$ and $B$, $\langle\langle A(t), B(0)\rangle\rangle$, is defined by[15]

$$\langle\langle A(t),\ B(0)\rangle\rangle = -i\Theta(t)\langle[A(t),\ B(0)]\rangle, \qquad (2.1)$$

where $t$ is the time, $\Theta$ is the unit step function, $[A, B] = AB - BA$, $A(t) = e^{iHt}A(0)e^{-iHt}$ (where $H$ is the Hamiltonian operator), and the average of an operator over a canonical ensemble at temperature $T$ is indicated by enclosing that operator within angular brackets.

If we denote the time Fourier transform of $\langle\langle A(t), B(0)\rangle\rangle$ as

$$\langle\langle A,\ B\rangle\rangle_\omega = (2\pi)^{-1}\int_{-\infty}^{\infty}\langle\langle A(t),\ B(0)\rangle\rangle e^{i\omega t}\,dt, \qquad (2.2)$$

the first-order equation of motion for $\langle\langle A(t), B(0)\rangle\rangle$ may be written[15]

$$\omega\langle\langle A,\ B\rangle\rangle_\omega = (2\pi)^{-1}\langle[A,\ B]\rangle + \langle\langle[A,\ H],\ B\rangle\rangle_\omega . \qquad (2.3)$$

If the operator $A$ appearing in Eq. (2.3) is replaced by the operator $[A, H]$, the equation of motion for the Green's function $\langle\langle[A, H], B\rangle\rangle_\omega$ appearing on the right-hand side of Eq. (2.3) is obtained, as

$$\omega\langle\langle[A,\ H],B\rangle\rangle_\omega = (2\pi)^{-1}\langle[[A,\ H],B]\rangle + \langle\langle[[A,\ H],\ B]\rangle\rangle_\omega . \qquad (2.4)$$

From Eqs. (2.3) and (2.4) it is seen that an infinite heirarchy of Green's-function equations of motion is obtained unless the Green's function on the right-hand side of a higher-order equation turns out to be identical to a Green's function in a lower-order equation, and this does not happen for the Hamiltonian given by Eq. (1.1). It is convenient to let $A = S_j^z$ and $B = S_0^z$ in Eqs. (2.3) and (2.4) because the equations of motion are simpler than for the frequently used choice $S_j^+$ and $S_0^-$. $\langle\langle[S_j^z, H], S_0^z\rangle\rangle_\omega$ may be eliminated in Eq. (2.3) by using Eq. (2.4), to obtain a second-order equation of motion for $\langle\langle S_j^z, S_0^z\rangle\rangle_\omega$ given by

$$\begin{aligned}\omega^2\langle\langle S_j^z,\ S_0^z\rangle\rangle_\omega = (2\pi)^{-1}J\sum_d\langle S_0^xS_d^x + S_0^yS_d^y\rangle(\delta_{j,0} - \delta_{j+d,0}) &+ J^2\sum_{dd'}\langle\langle(\vec{S}_j\cdot\vec{S}_{j+d}S_{j+d+d'}^z - \vec{S}_j\cdot\vec{S}_{j+d+d'}S_{j+d}^z),\ S_0^z\rangle\rangle_\omega \\ &+ J^2\sum_{dd'}\langle\langle(\vec{S}_{j+d}\cdot\vec{S}_{j+d'}S_j^z - S_{j+d}^z\vec{S}_j\cdot\vec{S}_{j+d'}),\ S_0\rangle\rangle_\omega , \end{aligned} \qquad (2.5)$$

where $\delta_{j,l}$ is the Kronecker $\delta$, after using the angular-momentum commutation relations together with Eq. (1.1).[16]

## III. DECOUPLING THE EQUATIONS OF MOTION

The higher-order Green's functions appearing on the right-hand side of Eq. (2.5) may be approximated in terms of a first-order Green's function of the kind found on the left-hand side, in order to close the equations of motion at second order. The decoupling approximation is rather arbitrary in comparison with that at first order, due to the combinations of more complicated operators appearing in the higher-order Green's functions.

Perhaps the most "natural" generalization of the first-order Tyablikov random-phase-decoupling approximation[11] is found by expressing the commutators of three spin operators with one spin operator in the higher-order Green's functions as sums of terms having commutators of only one spin operator with another, in order to satisfy the definition of the first-order Green's functions as given by Eq. (2.1), obtaining the approximation

$$\begin{aligned}\langle\langle S_j^\alpha S_l^\alpha S_m^z,\ S_0^z\rangle\rangle_\omega \approx{}& \langle S_j^\alpha S_l^\alpha\rangle\langle\langle S_m^z,\ S_0^z\rangle\rangle_\omega \\ &+ \langle S_j^\alpha S_m^z\rangle\langle\langle S_l^\alpha,\ S_0^z\rangle\rangle_\omega + \langle S_l^\alpha S_m^z\rangle\langle\langle S_j^\alpha,\ S_0^z\rangle\rangle_\omega ,\end{aligned} \qquad (3.1)$$

where $\alpha$ may be $x$, $y$, or $z$ and $\langle\langle S_m^z S_j^\alpha S_l^\alpha,\ S_0^z\rangle\rangle_\omega$ is approximated in the same manner. If $\alpha$ is $x$ or $y$, the correlation functions $\langle S_j^\alpha S_m^z\rangle$ and $\langle S_l^\alpha S_m^z\rangle$ vanish,[17] and the only Green's function which appears is $\langle\langle S_m^z,\ S_0^z\rangle\rangle_\omega$. When Eq. (3.1) is substituted into Eq. (2.5), the terms in this approximation for $\alpha = z$ vanish when summed over, as they must, since inspection of Eq. (2.5) reveals that the operators in the Green's-function arguments for $\alpha = z$ vanish rigorously when summed over.

The spatial isotropy and translational invariance of the Hamiltonian give the relations

$$\langle S_j^x S_l^x\rangle = \langle S_j^y S_l^y\rangle = \langle S_0^z S_{|j-l|}^z\rangle \qquad (3.2)$$

for any $j$ and $l$. The equation obtained by substituting Eqs. (3.1) and (3.2) into Eq. (2.5) can easily be solved in terms of the lattice Fourier transform of $\langle\langle S^z_m, S^z_0\rangle\rangle_\omega$,

$$G_k(\omega)=\sum_{m=0}^{N-1} e^{ikm}\langle\langle S^z_m, S^z_0\rangle\rangle_\omega\,. \tag{3.3}$$

$G_k(\omega)$ will involve the unknown correlation functions $\langle S^z_0 S^z_1\rangle$ and $\langle S^z_0 S^z_2\rangle$, which must be determined self-consistently. $\langle S^z_k S^z_{-k}\rangle$ may be found in terms of $\langle S^z_0 S^z_1\rangle$ and $\langle S^z_0 S^z_2\rangle$ from the spectral theorem[15]

$$\langle S^z_k S^z_{-k}\rangle = i\lim_{\gamma\to 0}\int_{-\infty}^{\infty}[G_k(\omega+i\gamma)-G_k(\omega-i\gamma)]\times n(\omega/k_BT)\,d\omega\,, \tag{3.4}$$

where $n(x)=(e^x-1)^{-1}$. The self-consistency equations are then given by the inverse-lattice Fourier transforms

$$\langle S^z_0 S^z_m\rangle = N^{-1}\sum_{k=-\pi}^{\pi}\langle S^z_k S^z_{-k}\rangle\cos mk\,. \tag{3.5}$$

Since

$$\langle S^z_0 S^z_0\rangle = \tfrac{1}{3}\langle \vec{S}_0\cdot\vec{S}_0\rangle = \tfrac{1}{3}S(S+1)\,, \tag{3.6}$$

Eq. (3.5) for $m=0$ yields an additional equation which $\langle S^z_0 S^z_1\rangle$ and $\langle S^z_0 S^z_2\rangle$ must satisfy, and we have three equations in two unknowns. An additional temperature-dependent unknown is required.

The arbitrariness in the decoupling approximation may be used to introduce the required unknown. It is not reasonable to use Eq. (3.1) for all values of $j$, $l$, and $m$. Different temperature dependencies of the decoupling might be expected for the cases $|j-l|=0$, 1, and 2. In addition, the decoupling may be modified when $m=j$ or $l$, which occurs in Eq. (2.5) only for $|j-l|=1$. In fact, for the special case of spin-$\frac{1}{2}$, the operators for $m=j$ or $l$ in the Green's-function arguments of Eq. (2.5) vanish rigorously when summed over.[18] Since only one additional temperature-dependent unknown may be introduced if the temperature dependencies are to be self-contained in Eq. (3.5), we choose to modify our decoupling procedure as follows, calling the new unknown $\hat{\tau}_0$. For $\alpha=x$ or $y$, $\langle\langle S^\alpha_j S^\alpha_l S^z_m, S^z_0\rangle\rangle_\omega$ is approximated as $\langle S^\alpha_j S^\alpha_l\rangle\langle\langle S^z_m, S^z_0\rangle\rangle_\omega$ if $j=l$; $\hat{\tau}_0\langle S^\alpha_j S^\alpha_l\rangle\langle\langle S^z_m, S^z_0\rangle\rangle_\omega$ if $|j-l|=1$ and $m\neq j$ or $l$; $e_1\hat{\tau}_0\langle S^\alpha_j S^\alpha_l\rangle\langle\langle S^z_m, S^z_0\rangle\rangle_\omega$ if $|j-l|=1$ and $m=j$ or $l$; and $e_2\hat{\tau}_0\langle S^\alpha_j S^\alpha_l\rangle\langle\langle S^z_m, S^z_0\rangle\rangle_\omega$ if $|j-l|=2$. $\langle\langle S^z_m S^\alpha_j S^\alpha_l, S^z_0\rangle\rangle_\omega$ is approximated in the same manner. It is not necessary to be concerned with terms for $\alpha=z$, since their net contribution to Eq. (2.5) is zero. The constants $e_1$ and $e_2$ are to be determined by physical considerations in the zero- and high-temperature limits and are given no physical significance in our theory, other than as fitting parameters that allow the decoupling to yield correct results in these temperature limits, so that the theory can give a good approximation at arbitrary temperature.

It is convenient to introduce the normalized correlation functions

$$\tau_m = \langle\vec{S}_0\cdot\vec{S}_m\rangle/Y = 3\langle S^z_0 S^z_m\rangle/Y\,, \tag{3.7}$$

where $Y=S(S+1)$, and to make the change of variables

$$\hat{\tau}_m=\hat{\tau}_0\tau_m\,, \tag{3.8}$$

where $\tau_0=1$, from Eq. (3.6). Then with our modified decoupling approximation, Eq. (2.5) becomes

$$\frac{3\omega^2\langle\langle S^z_m, S^z_0\rangle\rangle_\omega}{2YJ^2}=\frac{\hat{\tau}_1}{2\pi\hat{\tau}_0}\sum_d(\delta_{m,0}-\delta_{m+d,0})+\hat{\tau}_1\sum_{dd'}\langle\langle S^z_{m+d+d'}, S^z_0\rangle\rangle_\omega - [1+(e_1+1)\hat{\tau}_1+e_2\hat{\tau}_2]\sum_d\langle\langle S^z_{m+d}, S^z_0\rangle\rangle_\omega+2[1+(e_1-1)\hat{\tau}_1+e_2\hat{\tau}_2]\langle\langle S^z_m, S^z_0\rangle\rangle_\omega\,. \tag{3.9}$$

Taking the lattice Fourier transform of this equation, we find

$$G_k(\omega)=2YJ\hat{\tau}_1(1-\cos k)/3\pi\hat{\tau}_0(\omega^2-\epsilon_k^2)\,, \tag{3.10}$$

where $G_k(\omega)$ is defined by Eq. (3.3) and

$$\epsilon_k^2=8YJ^2\hat{\tau}_1(1-\cos k)(1-h\cos k)/3h\,, \tag{3.11}$$

where

$$h=2\hat{\tau}_1[1+(e_1-1)\hat{\tau}_1+e_2\hat{\tau}_2]^{-1}\,. \tag{3.12}$$

$G_k(\omega)$ may be written

$$G_k(\omega)=[YJ\hat{\tau}_1(1-\cos k)/3\pi\hat{\tau}_0\epsilon_k]\times[(\omega-\epsilon_k)^{-1}-(\omega+\epsilon_k)^{-1}]\,,\quad \epsilon_k>0 \tag{3.13}$$

expressing the equal *a priori* probability for the creation of a magnon of energy $\epsilon_k$ and a magnon "hole" of energy $-\epsilon_k$, as required by the spatial isotropy of the Hamiltonian.[19]

From Eqs. (3.4) and (3.13) and the identity[20]

$$\lim_{\gamma\to 0}(x\pm i\gamma)^{-1}=\mathcal{P}(x^{-1})\mp i\pi\delta(x)\,, \tag{3.14}$$

where $\mathcal{P}$ denotes "principal values of the integral," $\delta(x)$ is the Dirac $\delta$ function, and the limit $\gamma\to 0$ is to be taken after integration, we have

$$\langle S^z_k S^z_{-k}\rangle=\frac{2YJ\hat{\tau}_1(1-\cos k)[2n(\epsilon_k/k_BT)+1]}{3\hat{\tau}_0\epsilon_k}\,. \tag{3.15}$$

Then from Eqs. (3.5), (3.7), (3.8), and (3.15) we obtain

$$\hat{\tau}_m = \pi^{-1}\left(\frac{3\hat{\tau}_1 h}{2Y}\right)^{1/2} \int_0^{\pi} \left(\frac{1-\cos k}{1-h\cos k}\right)^{1/2} \times \left[2n\left(\frac{\epsilon_k}{k_B T}\right)+1\right] \cos mk\, dk \qquad (3.16)$$

in the thermodynamic limit $N \to \infty$. Equation (3.16) provides the self-consistency conditions for $m=0$, 1, and 2 and for $m>2$ expresses the remaining correlation functions in terms of $\hat{\tau}_0$, $\hat{\tau}_1$, and $\hat{\tau}_2$.

Once the values of $\hat{\tau}_m$ have been found which satisfy Eq. (3.16), the other thermostatic quantities may be found from the correlation functions. From the translational invariance of the Hamiltonian [Eq. (1.1)], the internal energy is given by

$$\langle H \rangle = -NYJ\tau_1 , \qquad (3.17)$$

so that the specific heat per ion is

$$C = -YJ\frac{\partial \tau_1}{\partial T} \qquad (3.18)$$

Assuming the dominance of excitations of wavelength long compared to the lattice spacing $a$, so that the correlation length has meaning, we have from Eq. (3.15) the Lorentzian form

$$\langle S^z_k S^z_{-k} \rangle \approx \frac{k_B T}{J\hat{\tau}_0[k^2+2(1-h)/h]}, \quad k \approx 0, \quad T>0. \qquad (3.19)$$

Then from Eq. (3.5), we see that $\langle S^z_0 S^z_m \rangle$ takes the form $e^{-|m|a/l}$, where $l$ is the correlation length, given by

$$l = a[h/2(1-h)]^{1/2}. \qquad (3.20)$$

We shall retain this definition of the correlation length in terms of $\langle S^z_k S^z_{-k} \rangle$ for small $k$ even for high temperatures at which many short-wavelength excitations are present, because this definition is appropriate for comparison with neutron quasielastic scattering measurements. The susceptibility per ion $\chi$ is found from the fluctuation relation,[21] to be

$$\chi = \mu_B^2 g^2 \sum_{m=0}^{N-1} \langle S^z_0 S^z_m \rangle / k_B T, \qquad (3.21)$$

since $\langle S^z_m \rangle = 0$ for all $m$ and the Hamiltonian is translationally invariant. In Eq. (3.21), $\mu_B$ is the Bohr magneton and $g$ is the Landé factor. From Eqs. (3.5) and (3.19), we then have

$$\chi = \frac{\mu_B^2 g^2}{k_B T} \lim_{k \to 0} \langle S^z_k S^z_{-k} \rangle = \frac{\mu_B^2 g^2 h}{2J\hat{\tau}_0(1-h)}. \qquad (3.22)$$

No further justification will be given here for our decoupling approximation, since ultimately its validity rests on its ability to produce physically meaningful results. The zero-temperature, high-temperature, and classical limits, for which the exact thermal averages are known, are investigated in the sections to follow.

## IV. HIGH-TEMPERATURE LIMIT

The first few terms of the exact high-temperature expansions are easily found using the properties of the traces of the spin operators.[22] To second order, the exact thermal averages are

$$\begin{aligned} \tau_{1\,\mathrm{ex}} &\approx (Y/3\theta)(1-\tfrac{1}{4}\theta), \\ C_{\mathrm{ex}} &\approx k_B(Y^2/3\theta^2)(1-\tfrac{1}{2}\theta), \\ \tau_{2\,\mathrm{ex}} &\approx (Y/3\theta)^2(1-\tfrac{1}{2}\theta), \\ \chi_{\mathrm{ex}} &\approx \mu_B^2 g^2 (Y/3J\theta)(1+2Y/3\theta), \end{aligned} \qquad (4.1)$$

where $\theta = k_B T/J$ and $\theta \gg 1$. In addition, to first order, one has for any $m$,

$$\tau_{m\,\mathrm{ex}} \approx (Y/3\theta)^m. \qquad (4.2)$$

To obtain results to second order in the high-temperature limit from Eq. (3.16), it is sufficient to keep only the terms $2k_BT/\epsilon_k + \epsilon_k/6k_BT$ from the expansion of $2n(\epsilon_k/k_BT)+1 = \coth(\epsilon_k/2k_BT)$. The integrals are easily evaluated[23] to obtain

$$\hat{\tau}_m \approx \frac{3\theta[1-(1-h^2)^{1/2}]^m}{2Y(1-h^2)^{1/2}h^{m-1}} + \frac{y_m \hat{\tau}_1}{3\theta}, \qquad (4.3)$$

where $y_0 = 1$, $y_1 = -\frac{1}{2}$, and $y_2 = 0$. We may substitute $\hat{\tau}_1$ from Eq. (4.3) into Eq. (3.12) to obtain an equation for $h$ as a function of $\theta$,

$$2Y\theta(1+\tfrac{1}{6}\theta)h(1-h^2)^{1/2} \approx 3[2+(1-e_1)h][1-(1-h^2)^{1/2}], \qquad (4.4)$$

where the $\hat{\tau}_2$ term is omitted because it gives only a third-order contribution to $h$. Assuming $\lim_{\theta \to \infty} h = 0$,[24] Eq. (4.4) may be solved for $h$ to the first two orders, and the thermal averages may be found up to second order from Eqs. (3.8), (3.12), and (4.3) for comparison with the exact series results. In first order, $h \approx 2Y/3\theta$, $\hat{\tau}_0 \approx 1$, and we find that all relations in Eqs. (4.1) and (4.2) are satisfied in first order by our theory. Through second order,

$$\begin{aligned} h &\approx (2Y/3\theta)[1+\tfrac{1}{6}\theta+(e_1-1)Y/3\theta], \\ \hat{\tau}_0 &\approx 1+\tfrac{1}{6}\theta+(e_1-1)Y/3\theta, \end{aligned}$$

and we find

$$\begin{aligned} \tau_1 &\approx (Y/3\theta)[1+(e_1-1)Y/3\theta], \\ \tau_2 &\approx (Y/3\theta)^2[1+\tfrac{1}{3}\theta+2(e_1-1)Y/3\theta], \\ \chi &\approx \mu_B^2 g^2(Y/3J\theta)(1+2Y/3\theta). \end{aligned} \qquad (4.5)$$

We now choose constant $e_1$ so as to make $\tau_1 = \tau_{1\,\mathrm{ex}}$ to second order, so that

$$e_1 = 1 - \tfrac{3}{4}Y = (2S-1)(2S+3)/4Y, \qquad (4.6)$$

and we see that $e_1$ vanishes for spin-$\frac{1}{2}$, as required in Sec. III. Then, through second order, $h \approx (2Y/3\theta)(1-\frac{1}{12}\theta)$, $\hat{\tau}_0 \approx 1-\frac{1}{12}\theta$, and all relations in Eq. (4.1) are satisfied by our theory except for $\tau_2$, which becomes

$$\tau_2 \approx (Y/3\theta)^2(1-\tfrac{1}{6}\theta)\,. \tag{4.7}$$

If one chooses $e_1$ such that $\tau_2$ rather than $\tau_1$ is correct to second order, then $\tau_1$, and of course the internal energy and specific heat, become incorrect in second order and the requirement that $e_1$ vanish for spin-$\frac{1}{2}$ is not satisfied. The choice for $e_1$ made in Eq. (4.6) is obviously better. Since $e_2$ does not appear in the first two orders of the high-temperature expansion, our decoupling approximation cannot give both $\tau_1$ and $\tau_2$ correct to second order. A different decoupling or additional fitting parameter is required to do this.

From Eq. (3.20), the correlation length becomes

$$l \approx (Y/3\theta)^{1/2}(1+Y/3\theta-\tfrac{1}{24}\theta)\,, \tag{4.8}$$

and we find from Eq. (3.11) that

$$\lim_{\theta\to\infty}\epsilon_k = 2J[\tfrac{1}{3}Y(1-\cos k)]^{1/2}\,, \tag{4.9}$$

so that $\epsilon_k$ is proportional to $k$ for small $k$.

## V. LOW-TEMPERATURE LIMIT

In the limit $T\to 0$ in Eq. (3.16), $h\to 1$[24] and it is more convenient to use the variable

$$q = (1-h)/2h = [1+(e_1-3)\hat{\tau}_1 + e_2\hat{\tau}_2]/4\hat{\tau}_1\,, \tag{5.1}$$

since $q\to 0$ as $T\to 0$. If we define $x=\frac{1}{2}k$, from Eq. (3.11) we see that $\epsilon_k$ may be written

$$\epsilon_k = \epsilon_x = 4J(\tfrac{2}{3}Y\hat{\tau}_1)^{1/2}\,|\sin x|\,(\sin^2 x+q)^{1/2}\,. \tag{5.2}$$

Then Eq. (3.16) becomes

$$\hat{\tau}_m = \pi^{-1}\left(\frac{6\hat{\tau}_1}{Y}\right)^{1/2}\left[2\int_0^{\pi/2}\frac{\sin x\cos 2mx}{(\sin^2 x+q)^{1/2}}\,n\!\left(\frac{\epsilon_x}{k_BT}\right)dx + p_m\right], \tag{5.3}$$

where $p_m = \int_0^{\pi/2}\sin x\cos 2mx(\sin^2 x+q)^{-1/2}\,dx$. The integrals $p_m$ are easily evaluated[23] for $m\leqslant 2$ to give

$$\begin{aligned} p_0 &= \cot^{-1}q^{1/2}\,,\\ p_1 &= qp_0 - q^{1/2}\,,\\ p_2 &= (2+3q)p_1 + q^{1/2}\,. \end{aligned} \tag{5.4}$$

As $q\to 0$, we see that the major contribution to the integrand in Eq. (5.3) comes from small $x$ values, so that the leading term of $\hat{\tau}_m$, $m\geqslant 1$, is given by setting $n(\epsilon_x/k_BT) = k_BT/\epsilon_x$. The integrals are readily evaluated[23] to obtain

$$\hat{\tau}_m \approx 3\theta[(1+q)^{1/2}-q^{1/2}]^{2m}/4Yq^{1/2}(1+q)^{1/2}\,,\quad m\geqslant 1 \tag{5.5}$$

since $p_m$ does not contribute to the leading order. From Eq. (5.5), we see that $\hat{\tau}_m = \hat{\tau}_1$ and consequently $\tau_m = \tau_1$, $m\geqslant 1$, to first order, as expected for the ground state. Equation (5.5) is not valid for $m=0$ because $p_0$ does contribute to $\hat{\tau}_0$ in first order. We may find $\hat{\tau}_0$ to first order from the difference $\hat{\tau}_0-\hat{\tau}_1$, since the integrals in Eq. (5.3) will then cancel out in first order, to yield

$$\hat{\tau}_0 - \hat{\tau}_1 \approx \pi^{-1}(6\hat{\tau}_1/Y)^{1/2}(p_0-p_1) \approx (3\hat{\tau}_1/2Y)^{1/2}\,. \tag{5.6}$$

At zero temperature, the system must be in its ground state, which in the absence of magnetization consists of an isotropic linear combination of fully aligned spins. Then at $T=0$ we must have $\langle S_0^z S_m^z\rangle = S^2$, which we can satisfy by requiring $\tau_1 = S^2/Y$ at $T=0$, since we have one remaining undetermined constant $e_2$. Then $\hat{\tau}_1 = S^2\hat{\tau}_0/Y$ and Eq. (5.6) yields $\hat{\tau}_0 = \frac{3}{2}$ to first order. From Eq. (5.2), the excitation energy becomes

$$\lim_{\theta\to 0}\epsilon_k = 2JS(1-\cos k)\,, \tag{5.7}$$

which is the expected dispersion relation for the excitation of a magnon out of the ground state at zero temperature. However, we see from Eq. (5.2) that for small $k$, $\epsilon_k$ is proportional to $k$, rather than $k^2$, when $T>0$, indicating a greater degree of temperature renormalization of the excitation energy for long-wavelength spin waves than for those of short wavelength. The requirement $\tau_1 = S^2/Y$ when $T=0$ determines $e_2$ by Eqs. (4.6) and (5.1) to be

$$e_2 = [(3-e_1)\hat{\tau}_1 - 1]\,\hat{\tau}_1^{-1}\big|_{T=0} = (4S+1)^2/12Y\,, \tag{5.8}$$

since $q=0$ when $T=0$. According to Eq. (5.5) the leading term in $q$ is

$$q \approx \theta^2/4S^4\,. \tag{5.9}$$

Then from Eqs. (3.20), (3.22), (5.1), and (5.9), we have the leading term in the correlation length,

$$l = a/2q^{1/2} \approx aS^2\theta^{-1} \tag{5.10}$$

and in the susceptibility,

$$\chi = \mu_B^2 g^2/4J\hat{\tau}_0 q \approx 2\mu_B^2 g^2 S^4/3J\theta^2\,, \tag{5.11}$$

both of which go to infinity as $T\to 0$, as expected.

As done by Kondo and Yamaji[12] for spin-$\frac{1}{2}$, the next order of terms in $\tau_m$ may be found from the differences $\hat{\tau}_1-\hat{\tau}_2$ and $\hat{\tau}_0-\hat{\tau}_1$ by the replacements $\sin x\to x$, $\cos 2mx\to 1-2m^2x^2$, and $q\to 0$ in the integrands of the differences in Eq. (5.3) to obtain

$$\hat{\tau}_m - \hat{\tau}_{m+1} = \pi^{-1}\left(\frac{6\hat{\tau}_1}{Y}\right)^{1/2}\left(4(2m+1)\times\int_0^{\pi/2}\frac{x^2\,dx}{\exp[4(\frac{2}{3}Y\hat{\tau}_1)^{1/2}x^2\theta^{-1}]-1} + p_m - p_{m+1}\right) + O(\theta^2)\,. \tag{5.12}$$

The integral may be extended to infinity, with errors at most of order $\theta^2$, to get[23]

$$\hat{\tau}_m - \hat{\tau}_{m+1} = \frac{3(2m+1)v(\theta/2S)^{3/2}}{2(S+1)}$$

$$+\pi^{-1}\left(\frac{6\hat{\tau}_1}{Y}\right)^{1/2}(p_m-p_{m+1})+O(\theta^2), \qquad (5.13)$$

where $v=(2\pi)^{-1/2}\zeta(\frac{3}{2})\approx 1.042$, $\zeta$ is Riemann's zeta function, and $\hat{\tau}_1$ has been replaced by $3S^2/2Y$ in the first term. From Eq. (5.4) we can see that $p_1-p_2$ is of order $\theta^2$, and Eqs. (5.9) and (5.13) may be solved for $\hat{\tau}_1$ and $\hat{\tau}_2$ to order $\theta^{3/2}$ to obtain

$$\hat{\tau}_1=\frac{3S^2}{2Y}\left[1-\left(\frac{3vS(4S+1)^2}{8Y^2}\right)\left(\frac{\theta}{2S}\right)^{3/2}\right]+O(\theta^2),$$
$$\hat{\tau}_2=\frac{3S^2}{2Y}\left[1-\left(\frac{9vS(8S^2+8S+3)}{8Y^2}\right)\left(\frac{\theta}{2S}\right)^{3/2}\right]+O(\theta^2). \qquad (5.14)$$

From Eq. (5.4) we see that $p_0-p_1=\frac{1}{2}\pi+O(\theta^2)$, and from Eqs. (5.13) and (5.14), we have

$$\hat{\tau}_0=\frac{3}{2}\left[1-\left(\frac{vS^2(80S^3+64S^2+14S+3)}{16Y^2}\right)\left(\frac{\theta}{2S}\right)^{3/2}\right]+O(\theta^2). \qquad (5.15)$$

Then from Eqs. (3.8), (5.14), and (5.15) we find

$$\tau_m=(S^2/Y)[1-vS^{-1}f_m(S)(\theta/2S)^{3/2}], \quad m=1 \text{ or } 2 \qquad (5.16)$$

where

$$f_1(S)=(16S^3+80S^2+40S+3)/16(S+1)^3,$$
$$f_2(S)=(64S^3+224S^2+184S+51)/16(S+1)^3. \qquad (5.17)$$

From Eqs. (3.18) and (5.16), we have the specific heat to first order,

$$C=[\tfrac{3}{4}k_B v f_1(S)](\theta/2S)^{1/2}+O(\theta). \qquad (5.18)$$

Although the simple spin-wave picture is invalid here, since $\langle S_m^z\rangle=0$, it is interesting to compare Eq. (5.18) with the spin-wave-model specific heat. The ratio of our $T^{1/2}$ term to that of the spin-wave model is just $f_1(S)$, which is $\frac{5}{6}$ for $S=\frac{1}{2}$, slightly greater than unity for $S\geq 1$, and approaches unity as $S\to\infty$. The exact solution for the specific heat, if it could be found, would probably exhibit the same power-law dependence on the temperature as our solution. However the exact solution probably would not have the same functional dependence on $S$ at low temperature, but $f_m(S)$ may be viewed as a rational function approximation to the correct result. Our low-temperature results are identical to those of Kondo and Yamaji[12] for $S=\frac{1}{2}$. In fact, for spin-$\frac{1}{2}$, $e_1=0$ and $e_2=1$, so that our self-consistency equations become identical to those of Kondo and Yamaji and the solutions are identical throughout the entire temperature domain. The specific heat $T^{1/2}$ coefficient from Eq. (5.18) for $S=\frac{1}{2}$ is about 8% larger than estimated by Bonner and Fisher[9] in their extrapolation from finite chains, and is probably within their extrapolation error.

## VI. NUMERICAL SOLUTION AT ARBITRARY TEMPERATURE

If the parameter $\lambda=2(2Y\hat{\tau}_1/3)^{1/2}\theta^{-1}$ is introduced, the solution of the self-consistency equation can be reduced to the problem of finding the value of $h$ for a given value $\lambda$ that satisfies Eq. (3.12), where $\hat{\tau}_1$ and $\hat{\tau}_2$ are given by Eq. (3.16) in terms of $h$ and $\lambda$. Then $\hat{\tau}_0$ can be found from Eq. (3.16) and the remaining quantities of interest can be easily calculated. An estimate $h'$ is picked, $\hat{\tau}_1$ and $\hat{\tau}_2$ are calculated, and $h''$ is found from Eq. (3.12). Then $h'$ and $h''$ can be used to obtain a better estimate of $h$ for a new iteration. If $h''=h'$, $h'$ is the correct solution for $h$. By noting from Eqs. (3.12) and (3.16) that $h'\lessgtr h''$ implies that the correct $h\gtrless h'$, a convergent iteration scheme may be constructed.

The stability of the iteration scheme with respect to the initial estimate of $h$ was found to be greater for smaller $\lambda$. Therefore, the solution was found for smaller $\lambda$ values first, and the initial estimates of $h$ were improved for successively larger $\lambda$ values by use of an accumulative Lagrange extrapolation.[25] A convergence criterion of $|(h'-h'')/h'|<10^{-8}$ was used in the iteration routine.

Numerical integrations were done by the Romberg algorithm[26] with an absolute convergence criterion of $10^{-9}$. For $\lambda\geq 10$, the forms for $\hat{\tau}_m$ given by Eq. (5.3) were used and the integrals were split into two parts about $x=\lambda^{-1/2}$, since the integrand peak occurs for $x\lesssim\lambda^{-1/2}$. Once $\tau_1$ had been found as a function of $\theta$, the specific heat was calculated from Eq. (3.18) using the Newton forward-and-backward divided-difference formulas of fourth order for unequal intervals.[25]

The numerical computations were performed on the Georgia Institute of Technology Univac 1108 computer for spins-$\frac{1}{2}$, 1, and -$\frac{5}{2}$, in the temperature range $10^{-3}\lesssim\theta\lesssim 10^2$. The correlation functions, specific heat, inverse correlation length, and inverse susceptibility are shown as functions of temperature in Figs. 1–4, respectively. Accuracy of the computation of all quantities shown is estimated to be better than 0.1% for $\theta>0.025$ and better than 1.0% for $\theta<0.025$.

One feature of the numerical solution which cannot be seen from the displayed figures is the approach to the low-temperature power-law behavior derived in Sec. V. Full logarithmic graphs of the specific heat, correlation length, and susceptibility, as functions of temperature (not shown here), reveal that according to our theory the low-temperature power-law behavior does not set in until $\theta$ becomes very small, $\sim 10^{-3}$ for spin-$\frac{1}{2}$, $\sim 5\times 10^{-3}$ for spin 1, and $\sim 10^{-2}$ for spin-$\frac{5}{2}$. It should be mentioned that our theory may have no meaning for real physical systems at such low temperatures,

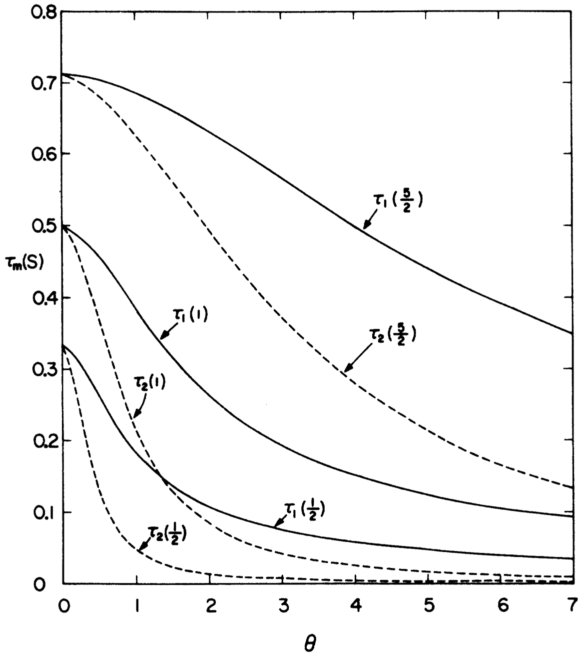

FIG. 1. Normalized correlation functions $\tau_m(S)$ for $m$th neighbor and spin $S$ vs $\theta = k_BT/J$.

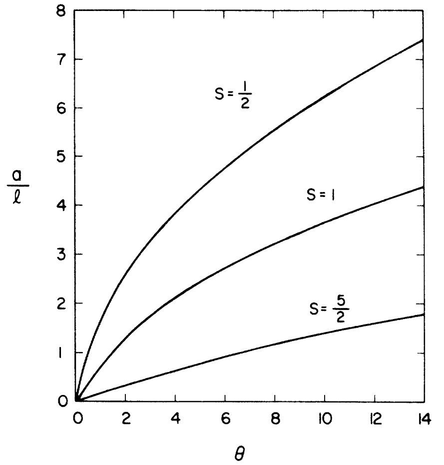

FIG. 3. Inverse correlation length $l^{-1}$ vs $\theta = k_BT/J$.

since three-dimensional effects, such as dipole-dipole interactions and interchain exchange, are likely to become important.

The specific heat is usually a rather sensitive measure for comparison, and in Fig. 1 we see that our specific heats roughly agree with extrapolation estimates from finite chains for spin-$\frac{1}{2}$[9] and spin 1.[10] The uncertainty in the extrapolation estimates from finite chains is greatest in the temperature region below and in the vicinity of the specific-heat peaks, because the peaks for finite chains have different locations and shapes than those for infinite chains and because the specific heats for finite chains approach zero exponentially as the temperature approaches zero. As pointed out in Sec. V, our low-temperature-specific-heat $T^{1/2}$ term agrees well with that estimated by Bonner and Fisher.[9] The discrepancy between the two results in the region $10^{-3} \lesssim \theta \lesssim 0.5$ is largely due to the fact that Bonner and Fisher's specific heat is close to the low-temperature power law up to $\theta \sim 0.025$. If our prediction is correct that the low-temperature power law does not set in until $\theta \lesssim 10^{-3}$ for spin-$\frac{1}{2}$, then our results should be viewed as more descriptive than theirs. The agreement be-

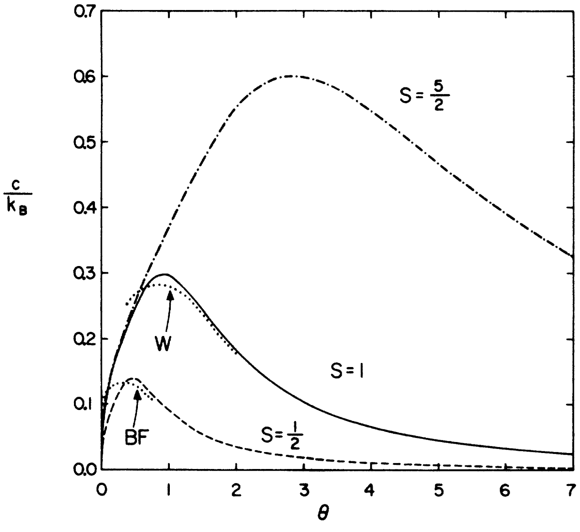

FIG. 2. Specific heat $C$ vs $\theta = k_BT/J$. Dotted lines are Bonner and Fisher's estimate BF for spin-$\frac{1}{2}$ and Weng's estimate W for spin 1, based on extrapolations from finite chains.

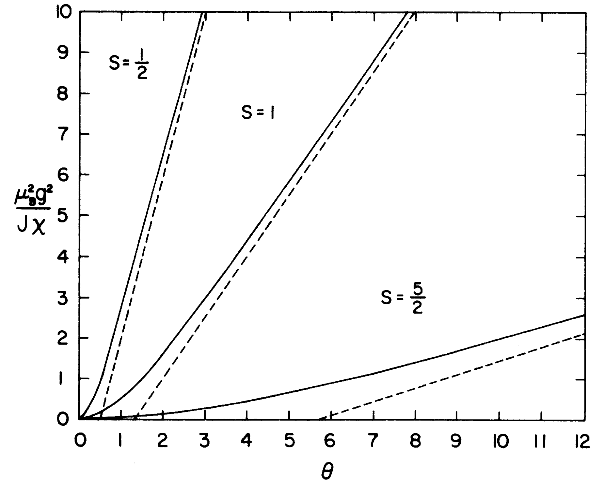

FIG. 4. Inverse susceptibility $\chi^{-1}$ vs $\theta = k_BT/J$. Dashed lines are high-temperature asymptotes $3\theta/S(S+1) - 2$.

tween our specific heats and those of Bonner and Fisher[9] and Weng[10] is fairly good for temperatures above the maxima, where there is the least uncertainty in their extrapolation estimates.

## VII. CLASSICAL LIMIT

Fisher's classical model[6] is obtained by taking the limit $S \to \infty$, $J \to 0$, $g \to 0$, such that $J_c = 2YJ$ and $g_c = 2Y^{1/2}g$ are constant. In this limit, the temperature region in which the low-temperature power-law behavior is valid shrinks to zero and a finite specific heat is obtained at zero temperature, in violation of the Nernst postulate of thermodynamics. Nevertheless, an examination of our theory in this limit is in order because the exact solution[6] of this model is known throughout the entire temperature domain and because the model may be a good approximation to high-spin systems for temperatures not too close to zero.

The classical limit of our theory is very much like the high-temperature limit. From Eq. (3.11), we see that $\epsilon_k \to 0$, so that $2n(\epsilon_k/k_BT)+1 \to 2k_BT/\epsilon_k$ in the classical limit, and we need merely to transcribe the first term of Eq. (4.3) to obtain

$$\hat{\tau}_{mc} = 3\theta_c[1-(1-h_c^2)^{1/2}]^m/2(1-h_c^2)^{1/2}h_c^{m-1}, \quad (7.1)$$

where $\theta_c = k_BT/J_c$ and the subscript $c$ indicates the classical limit of our theory. From Eqs. (3.8) and (7.1) we find

$$\tau_{1c} = [1-(1-h_c^2)^{1/2}]\,h_c^{-1} \quad (7.2)$$

and

$$\tau_{mc} = \tau_{1c}^m, \quad m \geq 1\,. \quad (7.3)$$

Equation (7.2) may be inverted to obtain

$$h_c = 2\tau_{1c}(1+\tau_{1c}^2)^{-1}\,. \quad (7.4)$$

Then, from Eqs. (3.20), (3.22), (7.1), and (7.4), we have the correlation length

$$l_c = a\tau_{1c}^{1/2}(1-\tau_{1c})^{-1} \quad (7.5)$$

and the susceptibility

$$\chi_c = \mu_B^2 g_c^2 \theta_c(1+\tau_{1c})/12J_c(1-\tau_{1c})\,. \quad (7.6)$$

According to Eq. (3.18), the specific heat is given by

$$C_c = -\left(\frac{k_B}{2}\right)\frac{\partial\tau_{1c}}{\partial\theta}\quad. \quad (7.7)$$

Inspection of Fisher's calculation[6] reveals that the exact solutions for $\tau_{mc}$, $l_c$, $\chi_c$, and $C_c$ obey Eqs. (7.3) and (7.5)–(7.7) in terms of the exact solution for $\tau_{1c}$,[27] so that our results for these quantities will approximate the corresponding exact solutions to the extent that our $\tau_{1c}$ approximates the exact solution for $\tau_{1c}$.

From Eqs. (4.6) and (5.8), we see that $e_1 \to 1$ and $e_2 \to \frac{4}{3}$ in the classical limit. Then by use of Eqs. (7.1)–(7.4) in Eq. (3.12), the relation between $\tau_{1c}$ and $\theta_c$ can be found,

$$\theta_c = (1-\tau_{1c}^2)/2\tau_{1c}(3-\tau_{1c}^2)\,. \quad (7.8)$$

In the high-temperature limit, Eq. (7.8) yields

$$\tau_{1c} \approx (\tfrac{1}{6}\theta_c)(1-\tfrac{1}{54}\theta_c^2), \quad \theta_c \gg 1 \quad (7.9)$$

to the first two nonvanishing orders in $\theta_c^{-1}$. The first term of Eq. (7.9) agrees with that for the exact solution and the coefficient of the second term is only 10% larger in magnitude than that for the exact solution. In the low-temperature limit, Eq. (7.8) yields

$$\tau_{1c} \approx 1-2\theta_c-2\theta_c^2, \quad \theta_c \ll 1 \quad (7.10)$$

where the first two terms agree with the exact solution, but the $\theta_c^2$ term is spurious. In fact, a Green's-function theory with the simple type of decoupling approximation we have used is incapable of describing $\tau_{1c}$ to higher order than $\theta_c$ in the low-temperature limit, because the remaining terms in the expansion of the exact solution are of exponential order. From Eqs. (7.7) and (7.10), we find the specific heat to be

$$C_c \approx k_B(1+2\theta_c), \quad \theta_c \ll 1 \quad (7.11)$$

where the first term agrees with the exact solution, but the $\theta_c$ term is spurious.

By finding $\theta_c$ for a wide range of $\tau_{1c}$ values from Eq. (7.8), $\tau_{1c}$ was compared to the exact solution $\coth(1/2\theta_c) - 2\theta_c$ at arbitrary temperature. Very good agreement was found throughout the full temperature range, with $\tau_{1c}$ always slightly greater than the exact solution. The maximum difference is ~1.7%, at $\theta_c \sim 0.17$, the difference being considerably less than this at most temperatures. The specific heat $C_c$ was then found at arbitrary temperature in the same manner as described in Sec. VI, and is compared to the exact solution in Fig. 5. Agreement for $C_c$ is not quite as good at low temperature as for $\tau_{1c}$, as expected, but the small peak is only ~6.5% above the exact solution and agreement is very good for $\theta_c > 0.15$.

In addition to the decoupling scheme used throughout this paper, with $e_1$ determined by Eq. (4.6) and $e_2$ by Eq. (5.8), a slightly simpler scheme was investigated in which the fitting parameter $e_2$ was eliminated by requiring that $e_2 = 1$ and $e_1$ was determined by requiring $\tau_1 = S^2/Y$ at zero temperature. It was found that $e_1 = 2(2S-1)/3S$, so that Kondo and Yamaji's results[12] for spin-$\frac{1}{2}$ are reproduced by this scheme as well. However, a somewhat different temperature dependence was found for higher spin. In particular, $\tau_1$ agreed with exact high-temperature series only to first order (except for spin-$\frac{1}{2}$), and in the low-temperature limit, $f_1(S)$ is replaced by $S(4S+13)/4(S+1)^2$ and $f_2(S)$ is replaced by $(16S^2+37S+12)/4(S+1)^2$ in Eq.

(5.16)–(5.18). In the classical limit, Eq. (7.1)–(7.7) remain valid, but $e_1 \rightarrow \frac{4}{3}$ and Eq. (7.8) is replaced by

$$\tau_{1c} = [(1 - 2\theta_c + 9\theta_c^2)^{1/2} + 3\theta_c]^{-1}, \tag{7.12}$$

leading to the high-temperature expansion

$$\tau_{1c} \approx (\tfrac{1}{6}\theta_c)(1 + \tfrac{1}{18}\theta_c), \quad \theta_c \gg 1 \tag{7.13}$$

where the second term is spurious, and the low-temperature expansion

$$\tau_{1c} \approx 1 - 2\theta_c + 4\theta_c^3, \quad \theta_c \ll 1 \tag{7.14}$$

where the $\theta_c^3$ term is spurious. From Eqs. (7.7) and (7.14), we have

$$C_c \approx k_B(1 - 6\theta_c^2), \quad \theta_c \ll 1 \tag{7.15}$$

where the $\theta_c^2$ term is spurious. As seen in Fig. 5, the simpler decoupling scheme yields better agreement with the exact classical specific heat for $\theta_c < 0.1$ and worse agreement for $\theta_c > 0.1$. The simpler scheme gives generally worse agreement with the exact classical correlation functions, the maximum difference for $\tau_{1c}$ being ~6%, at $\theta_c \sim 0.4$, with $\tau_{1c}$ always smaller than the exact solution. The simpler scheme does however yield better agreement with the exact correlation functions for $\theta_c < 0.1$. The better agreement of the simpler scheme with the classical model for $\theta_c < 0.1$ is less significant than its worse agreement for $\theta_c > 0.1$, since the classical model violates the Nernst postulate, so that the behavior of high-spin systems will deviate from the classical model at low temperatures. This scheme was rejected because it is felt that the other decoupling scheme, with $e_1$ and $e_2$ determined by Eqs. (4.6) and (5.8), yields generally superior results throughout the full temperature range.

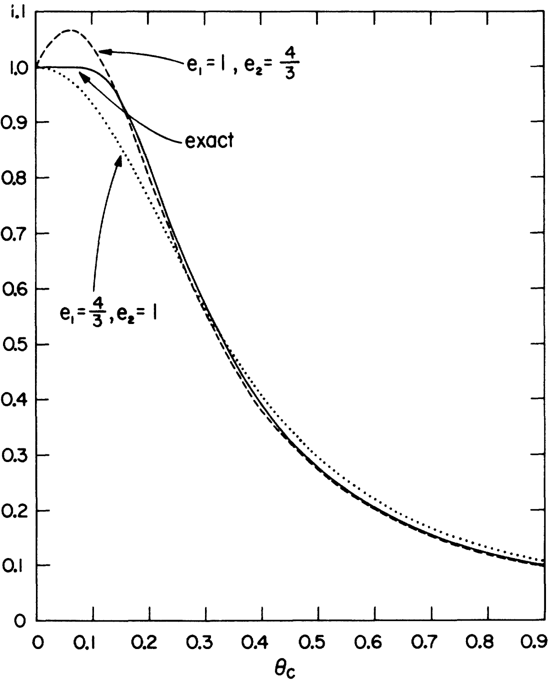

FIG. 5. Specific heat $C_c$ in the classical limit vs $\theta_c = k_B T/J_c$. Solid line is the exact solution, dashed line is our theory with accepted decoupling scheme, and dotted line is our theory with simpler decoupling scheme.

In comparing Figs. 2 and 5, one sees that in the vicinity of the specific-heat maxima our (accepted) solution bears approximately the same relationship, for spin-$\frac{1}{2}$, to Bonner and Fisher's result as it does to the exact solution in the classical limit, in that our solution gives a somewhat higher and narrower peak in the specific heat. One might be tempted to think this indicates the Bonner and Fisher result is closer to the truth than ours for spin-$\frac{1}{2}$. This is faulty logic, because the classical infinite-spin limit is a very different case from spin-$\frac{1}{2}$. For example, the simpler decoupling scheme discussed in the preceding paragraph, which gives the same result as our accepted scheme for spin-$\frac{1}{2}$, is seen in Fig. 5 to yield a specific heat in the classical limit which has a generally lower and broader peak than the exact solution. All that can be stated is that the specific heat derived from our accepted decoupling approximation may tend to have a peak which in the case of high spin is too high and too narrow to some extent.

## VIII. CONCLUSION

In the preceding sections, we have developed a Green's-function theory for the infinite-chain Heisenberg ferromagnet of arbitrary spin in zero magnetic field that yields reasonable results throughout the whole temperature domain for the equal-time spin-correlation functions, the correlation length, and related thermostatic averages. The required isotropy in the absence of magnetization is conserved by a decoupling of the equations of motion at second order with a random-phase approximation, yielding a set of three self-consistency equations for the first- and second-neighbor correlation functions and a decoupling parameter which reduce to the equations of Kondo and Yamaji[12] for spin-$\frac{1}{2}$. The solution of these equations yields well-defined spin waves with a finite correlation length at finite temperature. In the zero-temperature limit, the correlation length becomes infinite and the proper ground state and spin-wave spectrum are obtained. The correlation functions are all asymptotically exact in the high-temperature limit as well. At arbitrary temperature, fair agreement is found with extrapolations from finite chains for spin-$\frac{1}{2}$[9] and 1[10] and good agreement is obtained with the exact solution in the infinite-spin classical limit.[6]

The thermal averages obtained by extrapolation from finite chains have been found only for spins-$\frac{1}{2}$

and 1, because each spin case must be considered separately and larger matrices must be solved to obtain the larger number of eigenstates involved in this approach for higher spins. With minimal numerical computation, our theory yields results for arbitrary spin which are not of course subject to the uncertainty of such extrapolations. On the other hand, the accuracy of our decoupling approximation at arbitrary temperature is difficult to ascertain. As with any random-phase approximation, it leads to a sharp magnon excitation energy. Magnon-magnon interactions, which add a linewidth and an energy shift to the excitation energy and also result in bound magnon states of lower energy,[1] are accounted for in our theory only on an approximate average basis by the temperature-dependent renormalization of the magnon excitation energy, which in our case is wavelength dependent. We can only state that we are interested solely in thermostatic, rather than thermodynamic, averages and that the leading low-temperature term in our specific heat for spin-$\frac{1}{2}$ is very close to that found by Bonner and Fisher[9] from extrapolation of finite-chain calculations which account for all eigenstates of the Hamiltonian.

A comparison of our theory with the spin 1 linear-chain ferromagnet $CsNiF_3$[2-4] would be interesting. However, the spin of the $Ni^{++}$ ion in this crystal appears to experience a rather large uniaxial anisotropy of the easy-plane type, requiring a modification of our isotropic Hamiltonian. The proper decoupling of the Green's-function equations of motion in the presence of anisotropy and the comparison with experimental data on $CsNiF_3$ is a problem for future investigation.

## ACKNOWLEDGMENT

The authors express their appreciation to Professor H. A. Gersch for helpful comments on points which arose in this investigation.

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[16] In Eq. (2.5), $d$ and $d'$ are $\pm 1$ and $j$ is allowed to take on negative values. Periodic boundary conditions require negative $j$ values to be redefined as $N+j$.

[17] This can be seen by calculating the thermal trace using eigenstates of the total-spin $z$ component as basis, noting that these states are also eigenstates of the Hamiltonian.

[18] $S_j^\alpha S_{j+d}^\alpha S_{j+d+d'}^z$ with $d'=-d$ equals $S_{j+d}^z S_j^\alpha S_{j+d'}^\alpha$ with $d'=d$, for $\alpha = x$ or $y$ and spin-$\frac{1}{2}$. This can be shown using the commutator relation $[S_j^z, S_j^\pm] = \pm S_j^\pm$ and the special spin-$\frac{1}{2}$ identity $S_j^z S_j^\pm = \pm S_j^\pm / 2$.

[19] The Green's function $\Sigma_m e^{ikm} \langle\langle S_m^+, S_0^- \rangle\rangle_\omega$, which describes magnons and magnon "holes", can be shown to have the same form as Eq. (3.13) in our decoupling approximation. For example, see Ref. 12.

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