Second-Order Green's-Function Theory of a One-Dimensional Heisenberg Antiferromagnet*

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A self-consistent second-order Green's-function theory of a one-dimensional Heisenberg antiferromagnet, exhibiting spin waves without long-range order, is shown to yield qualitatively different results than a hybrid version treated by Richards. Comparison with exact and computer solutions for energy shows that the self-consistent solution represents a substantial improvement in local spin behavior at low temperatures. Long-range spin behavior at low temperatures is not correctly predicted by the self-consistent theory.

In a recent Letter,[1] Richards showed that the results of a second-order Green's-function theory (GFT) for a one-dimensional Heisenberg antiferromagnet gave good agreement with both Fisher's classical solution[2] for the inverse correlation length and the static correlation function and with experimental neutron scattering results for these quantities in $(CD_3)_4NMnCl_3$ (TMMC) on the temperature interval $1.1 \leq T \leq 40$°K. The spin-spin correlations between nearest and next-nearest neighboring spins in Ref. 1 were taken from the classical solution, rather than obtaining them from a self-consistent Green's-function theory (SCGFT). This procedure raises the question of whether the good agreement of the GFT with the classical solution is in large measure enforced by the utilization of the classical parameters in the GFT. We carry out a SCGFT in this Letter, compare the results with the classical theory, and find that the good agreement implied in Ref. 1 largely disappears.

The second-order decoupling scheme for the Green's-function equations of motion satisfying the rotational invariance implied by the form of the Heisenberg Hamiltonian previously used to study the paramagnetic region of Heisenberg magnets[4] yields an equation for the excitation energies identical to that given by Richards[1]:

$$E_k^2 = \tfrac{16}{3} J^2 Y(1+\tau_2)(1-\cos k)(1+h\cos k), \tag{1}$$

where k is a wave vector in units of the inverse lattice spacing, $h = 2|\tau_1|/(1+\tau_2)$, $Y = S(S+1)$, and where the $\tau_i = \langle \vec{S}_0 \cdot \vec{S}_i \rangle / Y$ $(i = 1, 2)$ are normalized first- and second-nearest-neighbor spin-spin correlation functions.

The spectral theorem gives the Fourier transform of the static correlation function which is also identical with Richards's expression:

$$\langle \vec{S}_k \cdot \vec{S}_{-k} \rangle = 4J|\tau_1| Y(1-\cos k)(1+2n_k)/E_k, \tag{2}$$

where $1/n_k = \exp(\beta E_k) - 1$ and $\beta = 1/k_B T$. The Fourier lattice transform furnishes the relations $\langle \vec{S}_0 \cdot \vec{S}_n \rangle = N^{-1} \sum_k \langle \vec{S}_k \cdot \vec{S}_{-k} \rangle \cos nk$. These provide, for the values $n = 0, 1$, two linearly independent equations which uniquely determine both τ_1 and τ_2. The sums that appear in these two equations, which are explicity independent of n_k, are exactly summable in the thermodynamic limit. The self-consistency equations that follow after this summation is performed may be expressed in terms of the remaining lattice sums $L_n(k_i, k_f)$:

$$L_n(k_i, k_f) = 2N^{-1} \sum_{k_i}^{k_f} A_n(k,h) n_k, \tag{3}$$

where

$$A_n(k,h) = [\tfrac{1}{2}(1+h\cos k)]^{-1/2} \sin(\tfrac{1}{2}k) \cos nk. \tag{4}$$

The first self-consistency equation is

$$\tau_1 = -MF(\Delta), \quad M = (1+h)/h, \tag{5}$$

where

$$\tfrac{1}{2}F(\Delta) = D - (D^2-1)^{1/2}, \tag{6}$$

$$D = 1 + C(1+\Delta)^2, \tag{7}$$

$$C = 3/\pi^2 Y, \tag{8}$$

$$\Delta = \pi(h/2M)^{1/2}[\tfrac{1}{2}ML_0(0, 2\pi) + L_1(0, 2\pi)]. \tag{9}$$

The second self-consistency equation is given by

$$\ln\{[1 + (\tfrac{1}{2}M)^{1/2}](\tfrac{1}{2}M - 1)^{-1/2}\} = -\tfrac{1}{2}\pi h^{1/2} L_0(0, 2\pi) + (2C|\tau_1|)^{-1/2}. \tag{10}$$

We first give the results for the ground state for which $n_k = 0$. Setting the $L_n = 0$ in the above equations, one obtains

$$(\tfrac{1}{2}M)^{1/2} \coth^{-1}(\tfrac{1}{2}M)^{1/2} = [4F(0)C]^{-1/2}. \tag{11}$$

Equations (5) and (11) provide numerical values for the ground-state parameters given in Table I. Our values for τ_1 are considerably larger than the classical predictions, $\tau_1 = 1$, and agree quite well with the exactly known value in the case of

TABLE I. Ground-state parameters for the one-dimensional Heisenberg antiferromagnet.

Spin	h	τ_1^0 [a] (SCGFT)	τ_1^0 [a] (Exact)
1/2	0.728 80	−0.495 71	−0.591 [b]
1	0.929 35	−0.602 14	$-3/4 < \tau_1^0 < -1/2$ [c]
5/2	0.998 60	−0.769 41	$-6/7 < \tau_1^0 < -5/7$ [c]

[a] Ground-state energy per spin is $2JS(S+1)\tau_1^0$.
[b] Ref. 5.
[c] Ref. 6.

$S=\frac{1}{2}$,[5] and the rigorous bounds in the case of $S=\frac{5}{2}$.[6]

The ground-state results for $S=\frac{5}{2}$ are applicable to TMMC and provide a description of the excitation energies E_k according to Eq. (1). The experimental results of inelastic neutron magnetic scattering at 4.4°K are known to fit very closely the curve $E_k = (6.1\ \text{meV})|\sin k|$ "over the entire one-dimensional Brillouin zone."[7] Since $h=0.9986$, our E_k does not quite go to zero at $k=\pi$ [see Eq. (12) below]. However, the percentage deviation of E_k from the value

$$E_k = 4J[\tfrac{1}{3}S(S+1)(1+\tau_2)]^{1/2}\sin k,$$

is only 5% at $k=0.95\pi$ and 1.5% at $k=0.9\pi$.

Richards,[1] using classical theory values $\tau_1=\tau_2=-1$ at zero degrees, predicts

$$E_k = 4J[\tfrac{2}{3}S(S+1)]^{1/2}\sin k,$$

while Lovesey and Meserve[8] use a theory of the shape of the relaxation function to get

$$E_k = 4J[S(S+1)]^{1/2}\sin k.$$

With $J=7.7$°K, the maximum E_k at $k=\pi/2$ is 5.65 meV (present theory), 6.41 meV (Richards), and 7.85 meV (Lovesey and Meserve).

Equation (1) predicts an energy gap at $k=\pi$,

$$E_G(T) = 4J[\tfrac{2}{3}Y(1+\tau_2)(1-h)]^{1/2}, \qquad (12)$$

which at $T=0$ is 0.298 meV, and a parabolic dependence of E_k for k near π. The energy gap is, of course, a consequence of $h=2|\tau_1|/(1+\tau_2)$ being less than unity. A further implication of h being less than unity is a finite value of the staggered susceptibility at $T=0$°K since the staggered susceptibility is proportional to $h/(1-h)$. This in turn implies finite values for both κ, the inverse correlation range, and $\langle\vec{S}_\pi\cdot\vec{S}_{-\pi}\rangle^{-1}$, contrasted with the value zero predicted by the classical theory for these two quantities.

Numerical calculations[9] on finite spin systems produce E_k spectra with no evidence for such a gap, so we are led to believe its presence represents a flaw in the SCGFT. It appears that the zero-temperature SCGFT corresponds to a ground state which is deficient in the long-range spin-spin correlations and concomitant large fluctuations in sublattice magnetization about its zero average value since $h\neq 1$ implies finite staggered susceptibility and finite correlation range.

Solutions for finite temperature are now restricted to low temperatures where the principal contributions to thermal excitations have wave vectors in the neighborhood of $k=0$ and $k=\pi$. (For TMMC this corresponds to $T<25$°K.) The lattice sums in Eq. (3) involve a phononlike spectrum on the interval $0<k<\pi/2$. The sums $L_n(0, \frac{1}{2}\pi)$ become integrals in the thermodynamic limit, and we find upon using $E_k \propto k$ and extending the upper limit of integration to infinity that

$$L_n(0,\tfrac{1}{2}\pi) = \pi(1-h)/6\beta^2 E_G{}^2 + O(\beta^{-4}). \qquad (13)$$

The evaluation of $L_n(\frac{1}{2}\pi,\pi)$ is more complicated since its value is very sensitive to the height of the rotonlike minimum in the excitation spectrum given by Eq. (12). Upper and lower bounds for $L_n(\frac{1}{2}\pi,\pi)$ are obtained by the following procedure. At low temperatures, Eq. (4) indicates $L_0(\frac{1}{2}\pi,\pi) \simeq -L_1(\frac{1}{2}\pi,\pi)$ so one need only consider $L_0(\frac{1}{2}\pi,\pi)$. The excitation spectrum is approximated by $E_k \simeq E_G[(1+h\cos k)/(1-h)]^{1/2}$. Expanding n_k in a geometric series, one obtains

$$L_0(\tfrac{1}{2}\pi,\pi) = (2/\pi)(2h)^{-1/2}\sum_{r=1}^{\infty}\exp(-r\beta E_G)I_r, \qquad (14)$$

where

$$I_r = \int_0^{x_0} dx\,[x(1+\tfrac{1}{2}x)]^{-1/2}\exp(-r\beta E_G x) \qquad (15)$$

has $x_0 = -1+(1-h)^{-1/2}$ as the upper limit of integration. Substitution of the rigorous bounds, $\exp(-\frac{1}{4}x) \leq (1+\frac{1}{2}x)^{-1/2} \leq 1$ into Eq. (15) yields

$$I_r = [(r\beta E_G + \tfrac{1}{4}\eta/\pi]^{-1/2}\exp(-\beta E_G r) \times \operatorname{erf}\{[x_0(r\beta E_G + \tfrac{1}{4}\eta)]^{1/2}\}, \qquad (16)$$

for some value of η satisfying $0\leq\eta\leq 1$. For the case of $S=\frac{5}{2}$, the error function in Eq. (16) may be replaced by unity for $T<25$°K.

After inserting the lattice sums L_0 and L_1 into Eqs. (9) and (10), upper and lower bounds on τ_1 and τ_2 are determined by varying the parameter η for fixed T. The quantity $\langle\vec{S}_k\cdot\vec{S}_{-k}\rangle$ in Eq. (2) has the Lorentzian form in terms of $\tilde{k}=\pi-k$, $\langle\vec{S}_k\cdot\vec{S}_{-k}\rangle \propto (\tilde{k}^2+\kappa^2)^{-1}$, defining the inverse correlation

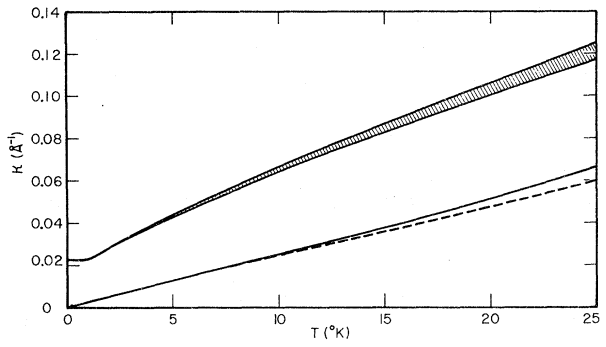

FIG. 1. Inverse correlation range κ versus temperature for TMMC ($S=\frac{5}{2}$). Solid line, Fisher's classical solution; dashed line, Richards's result; cross-sectioned curve, upper and lower bounds of present theory.

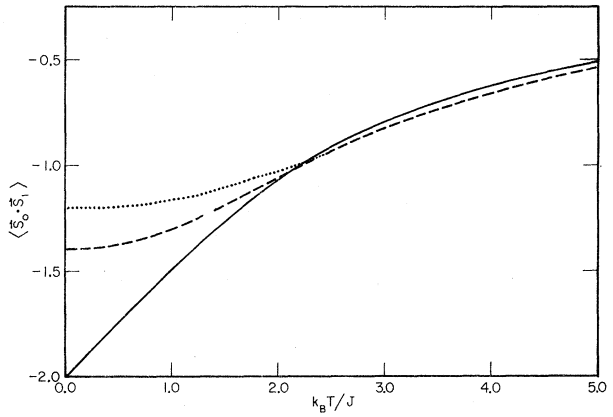

FIG. 2. Nearest-neighbor correlation function $\langle \vec{S}_0 \cdot \vec{S}_1 \rangle$ versus temperature for the linear Heisenberg antiferromagnet with $S=1$. Solid line, Fisher's classical model; dashed line, computer calculation of Weng; dotted line, present theory.

length κ as

$$(\kappa a)^2 = \frac{2(1-h)}{h}\left(1+\frac{n_\pi^2}{1+2n_\pi}\right)^{-1}. \qquad (17)$$

The inverse correlation length κ depends on the parameter η. Upper and lower bounds for κ at each temperature result by varying η between zero and unity.

Our results, Fisher's classical solution, and Richards's result for the inverse correlation length κ are shown in Fig. 1. One sees from the figure that the large discrepancy between our SCGFT and the classical solution remains for temperatures up to 25°K.[10] Inspection of Fig. 2 depicting $\langle \vec{S}_0 \cdot \vec{S}_1 \rangle$ data for $S=1$ shows the improvement of our SCGFT predictions[11] over the classical results when compared with the computer calculations of Weng.[12]

In conclusion, the SCGFT has been shown to produce an improvement over the classical-theory description of local spin behavior, as exemplified by nearest-neighbor correlations in the low-temperature region for a one-dimensional Heisenberg antiferromagnet. The long-range spin behavior is, on the other hand, much more accurately given by the classical theory. This unbalanced performance does not seem to us surprising, given that the ground state of this system is characterized by an infinite spin correlation length and concomitant macroscopic fluctuations in sublattice magnetization. These characteristics, similar to those present near an ordinary Curie or Néel point of a three-dimensional magnet, are known to receive adequate description via classical theory.

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[10]Results for $\langle \vec{S}_\pi \cdot \vec{S}_{-\pi} \rangle^{-1}$ not reported in this Letter show the same qualitative behavior as κ as a function of temperature.

[11]Both upper and lower bounds for $\langle \vec{S}_0 \cdot \vec{S}_1 \rangle$ fall within the width of the dotted curve in Fig. 2. However, our low-temperature approximations become questionable for values of $k_B T/J \gtrsim 2.0$.

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