Spin-Wave Description of Haldane-gap antiferromagnets

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(Received 13 December 2002)

PACS numbers: 75.10.Jm, 05.30.Jp, 75.40.Mg

Haldane’s conjecture [1,2] that one-dimensional Heisenberg antiferromagnets should exhibit qualitatively different low-energy structures based on whether the constituent spins are integral or fractional sparked renewed interest in the field of quantum magnetism. An energy gap immediately above the ground state was indeed observed in a quasi-one-dimensional Heisenberg antiferromagnet Ni(C$_2$H$_6$N$_2$)$_2$NO$_2$(ClO$_4$) [3] and a rigorous example of such a massive phase was also found out [4,5]. The energy gaps in magnetic excitation spectra, that is, spin gaps, are now one of the most attractive and important topics. In the context of theoretical progress, we may be reminded of quantized plateaux in the ground-state magnetization curves [6], a dramatic crossover from one- to two-dimensional quantum antiferromagnets [7], and an antiferromagnetic excitation gap accompanied by ferromagnetic background [8–11]. From the experimental point of view, metal oxides such as spin-Peierls compounds Cu$_{1-x}$M$_x$GeO$_3$ (M = Zn, Mg) [12,13], Haldane-gap antiferromagnets R$_2$BaNiO$_3$ (R = rare earth) [14] and ladder materials Sr$_{n-1}$Cu$_n$O$_{2n}$ (n = 3, 5, 7, ···) [15] have significantly contributed to systematic investigations of the mechanism of gap formation.

In this article, we aim to do away with our vague but persistent impression that the spin-wave scheme hardly works for one-dimensional quantum antiferromagnets. Spin-2 Haldane-gap antiferromagnets [36,37], as well as those of spin 1, have recently been synthesized and more explorations into novel quantum phenomena in one dimension are expected in the future. In such circumstances, we make our first attempt to construct a modified spin-wave theory for Haldane-gap antiferromagnets.

We consider integer-spin antiferromagnetic Heisenberg chains

\[ \mathcal{H} = J \sum_{j=1}^{L} \mathbf{S}_j \cdot \mathbf{S}_{j+1}; \quad S^z_j = S(S+1). \] (1)

We define bosonic operators for the spin deviation in each sublattice via

\[ S^+_{2n-1} = \sqrt{2S - a^+_n a_n} \quad a_n, \quad S^-_{2n-1} = S - a^+_n a_n, \] \[ S^+_{2n} = b^+_n \sqrt{2S - b^+_n b_n}, \quad S^-_{2n} = -S + b^+_n b_n. \] (2)

The Fourier-transformed operators are introduced as

\[ a_k = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{i k (n-1/4)} a_n, \] \[ b_k = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{-i k (n+1/4)} b_n, \] (3)

where twice the lattice constant 2a is set equal to unity and therefore \( k = 2 \pi n / N \) (\( n = 0, 1, \ldots, N-1; \ N = L/2 \)). Through the Bogoliubov transformation

\[ a^+_k = \alpha^+_k \cosh \theta_k - \beta_k \sinh \theta_k, \] \[ b^+_k = \beta^+_k \cosh \theta_k - \alpha_k \sinh \theta_k, \] (4)
we obtain
\[ \mathcal{H} = E_N + E_1 + E_0 + \mathcal{H}_1 + \mathcal{H}_0 + O(S^{-1}), \]
where
\[ E_N = -2NJ S^2, \quad (6a) \]
\[ E_1 = -4NJS (\Gamma - \Lambda), \quad (6b) \]
\[ E_0 = -2NJ (\Gamma - \Lambda)^2, \quad (6c) \]
\[ \mathcal{H}_1 = J \sum_k [\omega_1(k) (a_k^\dagger a_k + b_k^\dagger b_k)] \]
\[ + \gamma_1(k) (a_k^\dagger b_k + a_k^\dagger b_k^\dagger)] \]
\[ + \gamma_0(k) (\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k)], \quad (7a) \]
\[ \mathcal{H}_0 = J \sum_k [\omega_0(k) (a_k^\dagger a_k + b_k^\dagger b_k)] \]
\[ + \gamma_0(k) (\alpha_k^\dagger \beta_k + \beta_k^\dagger \alpha_k)], \quad (7b) \]
with
\[ \Gamma = \frac{1}{2N} \sum_k \cos \frac{k}{2} \sinh 2 \theta_k, \quad (8a) \]
\[ \Lambda = \frac{1}{2N} \sum_k (\cosh 2 \theta_k - 1), \quad (8b) \]
\[ \omega_1(k) = 2S \left( \cosh 2 \theta_k - \cos \frac{k}{2} \sinh 2 \theta_k \right), \quad (9a) \]
\[ \omega_0(k) = 2(\Gamma - \Lambda) \left( \cosh 2 \theta_k - \cos \frac{k}{2} \sinh 2 \theta_k \right), \quad (9b) \]
\[ \gamma_1(k) = 2S \left( \cos \frac{k}{2} \cosh 2 \theta_k - \sinh 2 \theta_k \right), \quad (10a) \]
\[ \gamma_0(k) = 2(\Gamma - \Lambda) \left( \cos \frac{k}{2} \cosh 2 \theta_k - \sinh 2 \theta_k \right). \quad (10b) \]
The naivest diagonalization of the Hamiltonian (5), whether up to $O(S^1)$ or up to $O(S^0)$, results in diverging sublattice magnetizations even at zero temperature. Although eq. (2) assumes that $a_n^\dagger a_n \leq 2S$ and $b_n^\dagger b_n \leq 2S$, the conventional spin-wave theory cannot reasonably control the boson numbers. Then we consider introducing a grand canonical constraint to the noncompact Hamiltonian (5).

Isotropic magnets should lie in the state of zero magnetization $\sum_j S_j^z = 0$ and the minimization of the free energy under such a condition indeed yields an excellent description of the low-temperature thermodynamics of ferromagnets [23]. However, in the cases with antiferromagnetic exchange interactions, the zero-magnetization constraint, claiming that $\sum_n (a_n^\dagger a_n - b_n^\dagger b_n) = 0$, still fails to overcome the divergence of the numbers of the sublattice bosons. Hence we minimize the free energy constraining the sublattice magnetizations to be zero:
\[ \sum_n a_n^\dagger a_n = \sum_n b_n^\dagger b_n = SN. \quad (11) \]

Within the conventional spin-wave theory, spins on one sublattice point predominantly up, while those on the other predominantly down. This condition (11) restores the sublattice symmetry. In order to enforce the constraint (11), we first introduce a Lagrange multiplier and diagonalize
\[ \tilde{\mathcal{H}} = \mathcal{H} + 2J \lambda \sum_k (a_k^\dagger a_k + b_k^\dagger b_k). \quad (12) \]

Then the ground-state energy and the dispersion relation are obtained as
\[ E_g = E_N + \tilde{E}_1; \quad \tilde{E}_1 = E_1 + 4N J \lambda \Lambda, \quad (13) \]
\[ \omega(k) = \tilde{\omega}_1(k); \quad \tilde{\omega}_1(k) = \omega_1(k) + 2\lambda \cosh 2 \theta_k, \quad (14) \]
within the linear modified spin-wave scheme and as
\[ E_g = E_N + \tilde{E}_1 + E_0, \quad (15) \]
\[ \omega(k) = \tilde{\omega}_1(k) + \omega_0(k), \quad (16) \]
within the up-to-$O(S^0)$ interacting modified spin-wave scheme. For eqs. (13) and (14), $\theta_k$ is given by $\gamma_1(k) - 2\lambda \sinh 2 \theta_k \equiv \tilde{\gamma}_1(k) = 0$, whereas for eqs. (15) and (16), $\theta_k$ may be determined in two ways. One idea is the perturbational treatment of $\mathcal{H}_0$, which is referred to as the perturbational interacting modified spin-wave scheme, where $\theta_k$ is still given by $\tilde{\gamma}_1(k) = 0$ and the $O(S^0)$ quantum correction is the $O(S^1)$-eigenstate average of $\mathcal{H}_0$. The other is the full diagonalization of $\mathcal{H}_1 + \mathcal{H}_0$, which is referred to as the full-diagonalization interacting modified spin-wave scheme, where $\theta_k$ is given by $\tilde{\gamma}_1(k) + \gamma_0(k) = 0$. Once $\theta_k$ is given, we calculate the free energy and obtain the optimum thermal distribution functions as
\[ \langle a_k^\dagger a_k \rangle = \langle b_k^\dagger b_k \rangle \equiv \tilde{n}_k = \frac{1}{e^{\gamma_0/k_B T - 1}}, \quad (17) \]
where $\lambda$ is self-consistently determined by the condition
\[ \sum_k (2\tilde{n}_k + 1) \cosh 2 \theta_k = (2S + 1)N. \quad (18) \]

First, let us evaluate the ground-state energy. We compare the modified spin-wave calculations with the highly accurate quantum Monte Carlo estimates [38] in Table I. The modified spin-wave findings are generally in good agreement with the quantum Monte Carlo results. The interacting modified spin waves describe the ground-state correlations much better than the linear ones and their description becomes increasingly refined with increasing $S$. The interacting modified spin-wave findings miss the correct value by 0.5 percent for $S = 1$ and by only 0.008 percent for $S = 3$.

Secondly, we consider the Haldane gap $\omega(ak = \pi) \equiv \Delta(T)$. Table II shows that the modified spin-wave scheme, in contrast with the conventional spin-wave theory, succeeds in generating the gap but considerably underestimates it. Unavailability of the absolute energy
The effective Hamiltonian \((12)\). Then we examine the present scheme by scaling \(\Delta(T)\) to its zero-temperature value \(\Delta(T=0) = \Delta_0\). Such an argument is quite usual with field-theoretical investigations, which do not lead to an estimate of the normalization factor but derive finite-temperature expressions involving only ratios such as \(k_B T / \Delta_0\). In Fig. 1, we compare the modified spin-wave calculations with numerical \([39]\), field-theoretical \([40]\) and experimental \([41]\) findings. Now the superiority of the modified spin-wave scheme is clear at a glance. Of all the theoretical tools, the full-diagonalization interacting modified spin-wave approach is the most successful to reproduce the observed upward behavior of \(\Delta(T)\) with increasing temperature \([41–43]\). The nonlinear \(\sigma\)-model treatment \([40,44]\) is justified well in the low-temperature region \(k_B T \ll \Delta_0\), while the maximum-entropy technique \([39]\) works less with increasing temperature. Nickel compounds such as \(Y_2BaNiO_5\) \([41]\) and \(Ni(C_2H_5N_2)_2NO_2(ClO_4)\) \([42]\) are good candidates for spin-1 Haldane-gap antiferromagnets, but magnetic anisotropy and interchain interaction are not negligible there. They split the lowest excitation gap of the isotropic chain \((1)\) into several levels and smear the intrinsic behavior of the ideal integer-spin chains. Considering such practical factors, the present theory satisfactorily interprets the observations.

Lastly, we show the modified spin-wave calculations of the magnetic susceptibility in Fig. 2. Considering the total breakdown of the conventional spin-wave theory in one-dimensional thermodynamic calculations, the modified spin-wave achievement is highly successful. All the calculations are guaranteed to reproduce the paramagnetic susceptibility \(\chi/Lg^2 \mu_B^2 = S(S+1)/3k_B T\) at high temperatures. Since the interacting modified spin-wave scheme gives better estimates of the gap than the linear modified spin-wave one (Table I), it is somewhat better at describing the low-temperature behavior. With increasing \(S\), the activation-type initial behavior is suppressed and the antiferromagnetic peak is broadened. The \(S = 1\) modified spin-wave calculations are in fine agreement with the quantum Monte Carlo findings over a wide temperature range, while those for \(S = 2\) look somewhat poorer at intermediate temperatures. It may be closely related to the fact that the modified spin-wave estimates of the \(S = 2\) gap are worse than those of the \(S = 1\) gap (Table II). However, Table II suggests that the validity of the modified spin-wave scheme for excitations significantly improves with increasing \(S\), possibly in a staggered way. Thermodynamic calculations, whether by quantum Monte Carlo or density-matrix renormalization group, for systems with large degrees of freedom are less feasible numerically, in particular, at low temperatures. The present scheme has the advantage of saving time and computational resources.

We have demonstrated the applicability of the new spin-wave scheme to Haldane-gap antiferromagnets. This is the first comprehensive attempt to describe one-dimensional spin-gapped antiferromagnets in terms of spin waves. The modified spin waves are free from their thermal as well as quantum divergence and can therefore microscopically interpret various magnetic properties. Besides the magnetic susceptibility, the spin correlation function and the nuclear spin-lattice relaxation time can be revealed. Unfortunately, we have less information on the bare energy spectrum, because we get rid of the quantum divergence at the cost of the original Hamiltonian. However, with the zero-temperature spin gap, which is readily and precisely available through numerical calculations within a canonical ensemble \([38]\), we can still argue the energy structure quantitatively.
Haldane-gap antiferromagnets in a magnetic field provide further interesting issues. With the increase of an applied field, the gap is reduced and the ground state is mixed increasingly with the first excited state. Indeed the nuclear spin-lattice relaxation is accelerated in the vicinity of the critical field, but its overall behavior as a function of an applied field and temperature [45] is far from understandable at a glance. We may expect the present new scheme to open the way for the total understanding of low-dimensional spin-gapped antiferromagnets.

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TABLE I. The linear (LMSW), perturbational interacting (PIMSW) and full-diagonalization interacting (FDIMSW) modified spin-wave calculations of the ground-state energy per site compared with quantum Monte Carlo (QMC) estimates [38].

| S  | LMSW        | PIMSW       | FDIMSW     |
|----|-------------|-------------|------------|
| 1  | −1.361879   | −4.726749   | −10.0901   |
| 2  | −1.394853   | −4.759760   | −10.1231   |
| 3  | −1.394617   | −4.759759   | −10.1231   |
|    | −1.401481(4)| −4.761249(6)| −10.1239(1) |

TABLE II. The linear (LMSW), perturbational interacting (PIMSW) and full-diagonalization interacting (FDIMSW) modified spin-wave calculations of the lowest excitation gap \(\Delta(T=0) \equiv \Delta_0\) compared with quantum Monte Carlo (QMC) estimates [38].

| S  | LMSW       | PIMSW       | FDIMSW     |
|----|------------|-------------|------------|
| 1  | 0.07200    | 0.00626     | 0.00279    |
| 2  | 0.07853    | 0.00655     | 0.00287    |
| 3  | 0.08507    | 0.00683     | 0.00295    |
|    | 0.41048(6) | 0.08917(4)  | 0.01002(3) |