S(k) for Haldane Gap Antiferromagnets: Large-scale Numerical Results vs. Field Theory and Experiment

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Abstract

The structure function, $S(k)$, for the $s = 1$, Haldane gap antiferromagnetic chain, is measured accurately using the recent density matrix renormalization group method, with chain-length 100. Excellent agreement with the nonlinear $\sigma$ model prediction is obtained, both at $k \approx \pi$ where a single magnon process dominates and at $k \approx 0$ where a two magnon process dominates. We repeat our calculation with crystal field anisotropy chosen to model NENP, obtaining good agreement with both field theory predictions and recent experiments. Correlation lengths, gaps and velocities are determined for both polarizations.

75.10.-b, 75.10.Jm, 75.40.Mg
It was first argued by Haldane [1] that integer-spin \( s \), antiferromagnetic chains exhibit a gap to a triplet magnon above a singlet ground state. This is by now well confirmed both experimentally [2,3] and numerically [4–8]. Much insight into these systems can be obtained from the nonlinear \( \sigma \) (NL\( \sigma \)) model, the long-distance field theory limit. Since this model only becomes an exact representation for large \( s \), it is a priori unclear how accurate it is for \( s = 1 \), the case of greatest experimental interest. Furthermore, the NL\( \sigma \) model itself is not very tractable and is therefore often approximated by a weakly interacting or free boson (Landau-Ginsburg) model [9]. The NL\( \sigma \) model gives information about two limiting wave-vectors: \( k \rightarrow \pi \) and \( k \rightarrow 0 \). The excitations near \( \pi \) are predicted to be a completely stable single-magnon, plus a three (and higher) magnon continuum. Near 0 the excitations are a two (and higher) magnon continuum. The single-magnon contribution to the structure function has a simple square root Lorentzian (SRL) form near \( k \approx \pi \) which only depends on the magnon interactions via a renormalization of the overall scale. On the other hand, near \( k \approx 0 \) the form of \( S(k, \omega) \) depends quite strongly on the interactions, but the overall scale is fixed by a sum rule since the total spin operator must obey the spin commutation relations. One non-trivial exact result is known about the NL\( \sigma \) model which is of relevance to possible experiments on spin chains: the explicit form of the two-magnon contribution to \( S(k, \omega) \) at small \( k \) [10]. The NL\( \sigma \) model does not predict how \( S(k, \omega) \) crosses over from one magnon to two magnon behavior as \( k \) is swept across the Brillouin zone. Anisotropy may be included in the NL\( \sigma \) model, but this destroys the integrability and we must then use the free boson approximation to the two-magnon contribution.

Recently, detailed inelastic neutron scattering experiments [11] were carried out on Ni(C\(_2\)H\(_8\)N\(_2\))\(_2\)NO\(_2\)(ClO\(_4\)) (NENP), measuring \( S(k, \omega) \) for \( k \geq 3\pi \). At all \( k \), the peak width appeared to be resolution limited. With a ratio of inter- to intra chain coupling estimated [12] to be \( 4 \times 10^{-4} \), NENP is a highly one dimensional compound. It is, however, not isotropic. The largest of the anisotropies is the single ion anisotropy, \( D \). Neglecting other smaller anisotropies NENP is well described by the Hamiltonian
\[ H = J \sum_i \{ \mathbf{S}_i \cdot \mathbf{S}_{i+1} + D(S^z_i)^2 \}, \]

with \( D/J = 0.18 \) and \( J = 3.75 \text{meV} \). Calculations of \( S(k, \omega) \) using exact diagonalization for \( L \leq 18 \) \([8,14]\) (\( L \leq 16 \) \([15]\)) or Monte Carlo for \( L \leq 32 \), \([8]\) have found some evidence for the two-particle nature of the small \( k \) excitations from the presence of spectral weight above the lowest-lying state.

Here, we present accurate results on the equal time structure function \( S(k) \), using the density matrix renormalization group (DMRG) method recently proposed by White \([6]\), with chain-length, \( L = 100 \). Details of our calculation will be given elsewhere \([16]\). Our results for \( S(k) \) are similar to those published previously for \( D/J = 0.0 \) (\( L = 18 \) \([8]\), \( D/J = 0.18 \) (\( L = 16 \) \([15]\)) except for a significant reduction of finite-size effects near \( k = \pi \) and the presence of data at smaller \( k \). This improved precision is crucial for much of our analysis. We compare our results on the pure Heisenberg model to the NL\( \sigma \) model predictions. The SRL form is obeyed accurately for \( k \geq 0.8\pi \), with a large renormalization of the amplitude. The two-magnon NL\( \sigma \) model result, including the overall amplitude, is remarkably accurate for \( k \leq 0.4\pi \). Using previous Monte Carlo results \([3]\) for \( \omega_e(k) \), the minimum energy excitation at wave-vector \( k \), we show that the single mode approximation (SMA) to \( S(k) \) is very good for \( k \approx \pi \), indicating that the three (and higher) magnon contribution is small. However, the SMA fails at small \( k \) suggesting the two-particle nature of the excitations there. We also obtain results for a single-ion anisotropy term with \( D/J = 0.18 \) to model NENP. We determined precise values for the singlet and doublet gaps, \( \Delta_\parallel/J = 0.6565(5) \), \( \Delta_\perp/J = 0.2998(1) \) (in good agreement with \([13]\)), correlation lengths, \( \xi_\parallel = 3.69(5) \), \( \xi_\perp = 8.35(7) \) and velocities, \( v_\parallel/J = 2.38(1) \), \( v_\perp/J = 2.53(1) \), and the ground state energy per spin \( e_0/J = -1.2856861 \). These obey the relativistic relationship, \( \xi_a = \hbar v_a/\Delta_a \) to within 2% for both polarizations. Again the SRL form is very accurate near \( k \approx \pi \) and the free boson model is fairly accurate at small \( k \). The SMA is very accurate at large enough \( k \) but fails badly at small \( k \). Good agreement is obtained with the experiment apart from an overall scale factor of about 1.25 which was to be expected since the experimental total intensity
exceeded the exact sum rule: 
\[ \frac{1}{L} \sum_{\alpha,k} S^{\alpha\alpha}(\vec{k}) = s(s+1) \] by 30\% (± 30\%). The behavior of \( S(k) \) suggests that slightly higher resolution and lower \( k \) neutron scattering experiments may reveal the two-particle nature of the excitations at small \( k \).

We implement the DMRG using density matrices of size \( 243 \times 243 \) keeping 81 eigenvectors at each iteration. Our results are obtained using a finite lattice method on a chain of length 100. For a discussion of the numerical procedure we refer the reader to the discussion by White [6]. The DMRG yields a higher accuracy if open boundary conditions are used, and all of our results are therefore obtained for open ended chains. If subject to open boundary conditions the \( s = 1 \) Heisenberg antiferromagnetic chain has a singlet ground state with positive parity, \( 0^+ \), and an exponentially low lying triplet, \( 1^- \) [17,18]. In the thermodynamic limit the ground state is thus four fold degenerate. We have performed our calculations in the \( 1^- \) state since this is computationally more convenient. We have repeated some of the calculations for the \( 0^+ \) state and for a smaller system of 60 site. In none of these cases were the results seen to differ aside from boundary effects which were seen to diminish with increasing chain length.

Let us first discuss our results for the isotropic chain where \( D = 0.0 \) in Eq. (1). The gap \( \Delta/J = 0.4107 \) [6,7,18] and the velocity \( v/J = 2.49(1) \) [18] is known. Using the DMRG method we calculated the bulk correlation function \( < S^z_{70} S^z_i > \), from which the correlation length \( \xi = 6.03(1) \) can be determined [16] in agreement with previous results [7]. Since our calculations are performed in the \( 1^- \) state \( < S^z_i > \) has a non zero expectation value giving rise to a disconnected part in the correlation function. The bulk correlation function we consider is therefore \( < S^z_{70} S^z_i > = < S^z_{70} > < S^z_i > \). We find that end effects are so small that we may assume this correlation function to be equal to the correlation function for a periodic chain. The static structure factor, \( S(k) \), can now be calculated by a simple Fourier transform \( S^{aa}(k) = \sum_r \exp(ikr)S^{aa}(r) \), where \( S^{aa}(r) \) is the above equal time correlation function in real space. Our results for \( S = S^{xx} = S^{yy} = S^{zz} \) are shown in Fig. 1 as the open squares. The results obey the sum rule \((1/L) \sum_{a,k} S^{aa}(k) = s(s+1)\) to within numerical
precision up to small boundary effects.

As already mentioned the isotropic \( s = 1 \) Heisenberg antiferromagnetic chain can be described by the NL\( \sigma \) model \([1]\), with Lagrangian density

\[
L_{nl} = \frac{1}{2g} \left[ \frac{1}{v} \left( \frac{\partial \phi}{\partial t} \right)^2 - v \left( \frac{\partial \phi}{\partial x} \right)^2 \right], \quad g = \frac{2}{s}, \quad v = 2Jas,
\]

where \( a \) is the lattice spacing and \( \phi \) describes the sublattice magnetization. We have approximately \( S_i = (-1)^i s \phi + a I \), with \( I = 1/(v g) \phi \times (\partial \phi/\partial t) \). Since we have set \( \hbar = 1 \), the velocity, \( v \), has dimension energy times length. The massive triplet of fields \( \phi \) is restricted to have unit magnitude, \( \phi^2 = 1 \). The lowest energy excitation corresponds to a single magnon with \( k = \pi \) and a relativistic dispersion \( E(k) = \sqrt{(\Delta)^2 + (v)^2(k - \pi)^2} \). Near \( k = \pi \) our numerical results for \( S(k) \) have a maximum corresponding to this stable single magnon, exhibiting the relativistic SRL form:

\[
gv \frac{1}{2 \sqrt{\Delta^2 + v^2(k - \pi)^2}}.
\]

Excellent agreement between this form and the numerical results is seen for \( k/\pi \geq 0.8 \) (see Fig. 1). Since \( v \) and \( \Delta \) are known from independent calculations this allows us to determine the coupling constant, \( g \approx 1.28 \). This result can be compared to the usual large \( s \) result of \( g = 2/s = 2 \) and tentatively to the \( 1/s \) expansion result \([19]\) \( g \approx 1.44 \) for the bare coupling.

Near \( k = 0 \) \( S(k) \) approaches zero as expected since the ground state is a singlet. In this region \( S(k) \) is expected to be dominated by a two (and higher) magnon continuum \([4]\) corresponding to the excitation of two magnons with momenta \( \pm \pi \). Exact results for the two magnon contribution to \( S(k, \omega) \) within the NL\( \sigma \) model framework have been obtained in Ref. \([10]\):

\[
a^2 |G(\theta)|^2 \frac{v k^2}{2 \pi} \frac{\sqrt{\omega^2 - (vk)^2 - 4\Delta^2}}{(\omega^2 - (vk)^2)^{3/2}}, \quad \omega^2 - (vk)^2 > 4\Delta^2.
\]

Here \( \omega^2 - (vk)^2 = 4\Delta^2 \cosh^2(\theta/2) \) and \( |G(\theta)|^2 \) is given by the expression

\[
|G(\theta)|^2 = \pi^4 \frac{1 + (\theta/\pi)^2}{64 \left(1 + (\theta/2\pi)^2 \right) } \left( \tanh(\theta/2) \right)^2.
\]
This result can be numerically integrated over $\omega$ to yield the two magnon contribution to the static structure factor. The result is shown as the solid line in Fig. [1] and, as can be seen, it is remarkably accurate out to $k/\pi \leq 0.4$.

If the nonlinear constraint, $\phi^2 = 1$, on the field $\phi$ in the NL$\sigma$ model is lifted, a simpler model of almost free bosons can be arrived at [9] with Lagrangian density

$$
\mathcal{L} = \sum_{i=1}^{3} \left[ \frac{1}{2v_i} \left( \frac{\partial\phi_i}{\partial t} \right)^2 - \frac{v_i}{2} \left( \frac{\partial\phi_i}{\partial x} \right)^2 - \frac{\Delta^2}{2v_i} (\phi_i)^2 \right] - \lambda \phi^4.
$$

(6)

By allowing the 3 velocities and gaps to be phenomenologically adjusted, anisotropy can be included. If the $\lambda \phi^4$ term is neglected a mean field theory of free bosons is obtained. In the isotropic case, within this free boson model, the two magnon contribution to $S(k)$ can be shown [10] to be equal to Eq. (4) with $|G(\theta)|^2 \equiv 1$. The free boson estimate is shown as the long dashed line in Fig. 1. While in good qualitative agreement with the numerical results near $k = 0$ the discrepancy at larger $k$, between this result and the exact NL$\sigma$ prediction, demonstrate the importance of interaction effects that are neglected in the free boson theory.

Within the SMA the dynamical structure factor $S(k, \omega)$ is approximated by a $\delta$-function at the single mode frequency $\omega_{\text{SMA}}$, $S(k, \omega) = S_0(k) \delta[\omega - \omega_{\text{SMA}}(k)]$. Since the first moment of the dynamical structure factor obeys a sum rule [20] this implies [16] that

$$
\omega_{\text{SMA}}^{zz}(k) = -J(F_x + F_y)(1 - \cos(k))/S^{zz}(k),
$$

(7)

with $F_a = \langle S^a_i S^a_{i+1} \rangle$. Denoting the lower edge of the spectrum by $\omega_e$, one can show [21,16] that the inequality $\omega_{\text{SMA}}(k) \geq \omega_e(k)$ is obeyed. Furthermore it can be shown [13] that if $S(k, \omega)$ has a $\delta$-function peak at the frequency $\omega_\delta(k)$, which is below the lower edge of the continuous part of the spectrum, $\omega_e(k)$, then $\omega_{\text{SMA}}(k) \geq \omega_\delta(k)$. This inequality breaks down if $\omega_\delta(k) > \omega_e(k)$. Using the numerical results $F_a = e_0/3 = -0.4671613$ for the isotropic chain we can now calculate $\omega_{\text{SMA}}(k)$ from $S(k)$. Our results are shown in Fig. 2 along with the quantum Monte Carlo (QMC) results of Takahashi [3] for the lower edge, $\omega_e$, of the spectrum. Also shown is the relativistic dispersion relation (solid line), which should be very accurate near $k = \pi$, and the bottom of the two-magnon continuum (long dashed line),
\[ 2\sqrt{\Delta^2 + v^2(k/2)^2}, \text{ valid near } k = 0. \] As can be seen in Fig. 2 both predictions coincides with the QMC results. While highly accurate near \( k = \pi \) the SMA is clearly seen to break down near \( k = 0 \). From our results for the structure factor we expect the two magnon scattering to become important around \( k \sim 0.4\pi \) which seems to be consistent with the divergence of \( \omega_{\text{SMA}} \) from the QMC results. If compared to the relativistic dispersion, which should represent the single magnon dispersion relation \( \omega_k \) near \( k = \pi \) very accurately, it is seen that also here there must be a small multi-magnon contribution since \( \omega_{\text{SMA}} \) is above \( \omega_k \).

We now turn to a discussion of our results for the anisotropic chain with \( D/J = 0.18 \). Using the DMRG we have calculated \( \langle S_{50}^z S_i^z \rangle \) and \( \langle S_{50}^x S_i^x \rangle \) in the \( 1^- \) state. In this case the disconnected part, \( \langle S_{50}^a S_i^a \rangle \), is so small that we can neglect it. Fourier transforming we obtain \( S^\parallel = S^{zz} \) and \( S^\perp = S^{xx} = S^{yy} \), respectively. Our results are shown in Fig. 3. We estimate end effects to give an error of 0.05\% in \( S^\perp(k = \pi) \), somewhat larger at \( k = 0 \). End effects are judged to be negligible for \( S^\parallel \). Also shown in Fig. 3 are the INS results of Ma et al. [11] which are directly comparable to our results. The open triangles are points where to within experimental accuracy \( S^\parallel \) and \( S^\perp \) were identical. The full squares and circles are data points where \( S^\parallel \) and \( S^\perp \), respectively, could be resolved experimentally. At \( k = \pi \) the experimental results are about 20-30\% higher than our numerical results. Taking into account a 25\% [22] uncertainty in the overall (multiplicative) normalization of the experimental results we find a good agreement between the numerical and experimental results.

Allowing for different velocities, gaps and coupling constants for the two modes we expect \( S^{aa}(k) \) to be well described by the SRL form Eq. (3) near \( k = \pi \). With the gaps and velocities mentioned above and with \( g_\parallel = 1.44, g_\perp = 1.17 \) this SRL form is shown as the dashed lines in Fig. 3. Very good agreement when \( k \geq 0.8\pi \) between the SRL form and the numerical results is evident for both \( S^\parallel \) and \( S^\perp \).

In the region near \( k = 0 \) we see that since rotational symmetry around the x and y axis is broken, \( S^\perp \) can take on a non zero value at \( k = 0 \). This is clearly seen in the numerical
results for $S^\perp$. We can extend the results of Ref. [10] for the free boson prediction to allow for different velocities. For the two magnon contribution to $S^{aa}(k)$ near $k = 0$ we obtain [16]

$$a^2 \int \frac{dk'dk''}{8\pi} \left( \frac{\omega_k^b v_c}{\omega_k^c v_b} + \frac{\omega_k'^b v_b}{\omega_k'^c v_c} - 2 \right) \delta(k - k' - k''),$$

with cyclic permutation of the indices $a, b, c$. Here $\omega_k^a = \sqrt{\Delta_a^2 + (v_a k)^2}$. These results are shown as the solid lines in Fig. 3. As was the case for the isotropic chain the free boson prediction seems to be qualitatively correct near $k = 0$. The crossings of $S^{||}(k)$ and $S^\perp(k)$ around $k = 0.85\pi$ and $0.1\pi$ (see Fig. 3) are reproduced correctly by the free boson model. In the range $0.1\pi < k < 0.85\pi$ $S^{||}$ is the larger of the two structure factors.

We have repeated the SMA calculation for the anisotropic case finding qualitatively similar behavior as for the isotropic chain. Again the SMA fails for small $k$ while giving relatively precise results near $k = \pi$, indicating a small but non zero contribution from the multi magnon continuum at $k = \pi$.

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FIGURES

FIG. 1. The structure factor, $S$, vs. $k/\pi$ for a 100 site isotropic chain. The numerical results are shown as open squares.

FIG. 2. Dispersion of excitations vs. $k/\pi$ for a 100 site isotropic chain. The points are QMC data from Ref. [5] for $L = 32$. The short dashed line is $\omega_{\text{SMA}}(k)$ obtained from $S(k)$ for $L = 100$.

FIG. 3. $S_{\parallel}$ and $S_{\perp}$ vs. $k/\pi$ for a 100 site anisotropic chain. The numerical results are shown as open squares and circles, respectively. The data points are INS data from Ref. [11].