Spectral weight contributions of many-particle bound states and continuum

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Cluster expansion methods are developed for calculating the spectral weight contributions of multiparticle excitations - continuum and bound states - to high orders. A complete 11th order calculation is carried out for the alternating Heisenberg chain. For $\lambda = 0.27$, relevant to the material Cu(\text{NO}_3)_2\cdot 2.5\text{D}_2\text{O}$, we present detailed spectral weights for the two-triplet continuum and all bound states. We also examine variation of the relative weights of one and two-particle states with bond alternation from the dimerized to the uniform chain limit.

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In recent years there has been a growing interest in understanding quantitatively the single and multiparticle excitation spectra in quantum spin systems. On the one hand, this understanding is important in assessing the extent of a continuum due to conventional multiparticle excitations as compared to more exotic scenarios [1]. On the other hand, the increased frequency and wave-vector resolution of inelastic neutron scattering experiments means that such spectra are being observed in real materials and need quantitative theoretical support [2].

Controlled and systematic calculation of the spin dynamics of quantum spin models remains a challenging computational task. Despite much progress in developing computational methods the multiparticle excitations remain poorly understood.

Here, we develop a general linked-cluster formalism to calculate the single-particle and multi-particle contributions to the dynamical structure factor by means of high-order series expansions. We apply the method to the alternating Heisenberg chain (AHC) model, where expansions are made around the strong coupling limit of decoupled spin dimers.

We follow the formalism of Tennant et al. [3]. The inelastic neutron scattering cross-section [3] at temperature $T = 0$ is proportional to the neutron scattering "structure factor" $S^{\alpha \beta}(k, \omega)$

$$S^{\alpha \beta}(k, \omega) = \frac{1}{2\pi N} \sum_{i,j} \int dt \exp[i(\omega t + k \cdot (r_i - r_j))]
\langle \psi_0 | S^{\alpha}_j(t) S^{\beta}_i(0) | \psi_0 \rangle$$

(1)

where $k$ is the wavevector transfer, $\omega$ is the energy, $N$ is the number of scattering centres, $\alpha, \beta = x, y, z$ are Cartesian coordinates, $i, j$ label sites of the system, and $| \psi_0 \rangle$ is the ground state of the Hamiltonian. This is just the space and time Fourier transform of the spin-spin correlation function. For AHC, the structure factor has only one independent spin component, and henceforth we concentrate our attention on $S^{++}(k, \omega)$.

Integrating equation (1) over energy, we get the integrated structure factor, i.e. the Fourier transform of the spin-spin correlation function at $t = 0$:

$$S^{++}(k) = \sum_r C^{++}(r) \exp[i(k \cdot r)]$$

(2)

Inserting a complete set of momentum eigenstates $| \psi_n \rangle$ of $H$ between the spin operators in equation (2), and using translation invariance, we can express the spin structure factor as a sum over "exclusive" structure factors

$$S^{++}_n(k, \omega) = \sum_{n} S^{++}_n(k, \omega) ,$$

(3)

$$S^{++}_n(k, \omega) = \sum_{i} \langle \psi_n(k) | S^{+}_{i*}(0) | \psi_0 \rangle \exp[i(k \cdot r_{i*})] $$

(4)

where the sum runs over all sites $i*$ in the unit cell, and $N_c$ is the number of unit cells on the lattice. Barnes et al. define the “reduced exclusive structure factor” as

$$S^{++}_n(k) = N_c \sum_{i*} \langle \psi_n(k) | S^{+}_{i*}(0) | \psi_0 \rangle \exp[i(k \cdot r_{i*})]$$

(5)

Each exclusive structure factor $S^{++}_n(q, \omega)$ gives the intensity of scattering from $| \psi_0 \rangle$ to a specific triplet excited state $| \psi_n(k) \rangle$.

We turn now to a discussion of algorithms for the calculation of exclusive structure factors within perturbation theory. Efficient linked cluster expansion methods have long been known [4] for calculating bulk properties of a quantum lattice system. Similar methods for the calculation of 1-particle spectra were developed by Gelfand [5], and were recently extended to 2-particle spectra by Trebst et al [6]. Series calculations of exclusive 1-particle structure factors or spectral weights appear in several places (e.g. [7]); and some low-order calculations for 2-particle states have recently been made [8]. Knetter et al [9] have used an alternative approach based on 'continuous unitary transformations' which is also capable of giving bound state energy spectra and structure factors to high order and in great detail.

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Let us suppose that the Hamiltonian can be decomposed
\[ H = H_0 + \lambda V \]
where \( H_0 \) is the unperturbed Hamiltonian and \( V \) is to be treated as a perturbation. We aim to expand the multiparticle dispersion relations and structure factors in powers of the parameter \( \lambda \). For illustrative purposes, we shall use the language of the AHC, but the formalism can be applied more generally.

At zeroth order, the ‘single-particle’ excitations consist of triplets on a single dimer which can be labelled \(|m, \alpha\rangle\), where \( m \) labels the position of the excited dimer, and \( \alpha \) labels the angular momentum eigenstate (i.e. \( S_z^2, S_z \)). The reduced exclusive structure factor is
\[ S^{\pm}(k) = \left| \sum_{m,i,\alpha} \langle m, \alpha | S^{\pm}_i | 0 \rangle \exp[i(k_i - k_m)] \right|^2 \]

It is easy to see that the matrix element \( \langle m, \alpha | S^{\pm}_i | 0 \rangle \equiv S_{1\alpha}^{(m,i)} \) must obey a simple ‘cluster addition’ property, such that the elements \( S_{1\alpha}^{(m,i)} \) admit a linked cluster expansion
\[ S_{1\alpha}^{(m,i)} = \sum_{\gamma} s_{\gamma,\alpha}^{(m,i)} \]
where the sum over \( \gamma \) denotes a sum over all connected clusters which are “rooted to” \( \gamma \), or contain, the positions of dimer \( m \) and site \( i \). Correspondingly, the perturbation series expansion for \( S_{1\alpha}^{(m,i)} \) could be formulated in terms of a diagrammatic expansion where only connected diagrams contribute, although we will not elaborate on this approach here.

An efficient linked cluster algorithm for calculation of the structure factors can now be formulated, following Trebst et al. [3]:

i) Generate a list of connected clusters \( \gamma \) appropriate to the problem at hand (in the present case, they will simply consist of chains of dimers of different lengths);

ii) For each cluster \( \gamma \), construct matrices for the Hamiltonian \( H \) and spin operators \( S_i^{\pm} \) in the basis of singlet and triplet dimer states corresponding to \( H_0 \);

iii) ‘Block diagonalize’ the Hamiltonian by an orthogonal transformation as outlined by Trebst et al. [3], constructed order-by-order in perturbation theory so that the 1-particle states sit in a block by themselves; simultaneously, transform the matrices for the spin operators
\[ H^{\text{eff}} = O^T H O, \quad S_i^{\text{eff}, \pm} = O^T S_i^{\pm} O \]

iv) Subtract all sub-cluster contributions;

v) Insert in equation (3), and then equation (2), to build up the exclusive structure factors order-by-order in perturbation theory.

Since two triplets can combine to give total spin 1, there will also be non-zero spin structure factors for 2-particle states. The 2-particle states can be labelled according to their unperturbed counterparts in position space \(|m,n; \alpha, \beta\rangle\) where \( m, n \) label the two dimer positions, and \( \alpha, \beta \) the corresponding angular momentum states. Then the exclusive matrix element is
\[ \langle \psi_j | S^+_i | 0 \rangle = \sum_{m,n} f_{\alpha\beta}(m,n) \langle m,n; \alpha, \beta | S^+_i | 0 \rangle \]

where the 2-particle wavefunctions \( f_{\alpha\beta}(m,n) \) can be computed by the block diagonalization procedure of Trebst et al. [3]. The 2-particle matrix elements in position space can then be found by a linked cluster algorithm essentially the same as that for the 1-particle states.

We have used this method to investigate the spectral weights for the alternating Heisenberg chain Hamiltonian, described by the following Hamiltonian
\[ H = \sum_i \left[ S_{2i} \cdot S_{2i+1} + \lambda S_{2i-1} \cdot S_{2i} \right] \]
where the \( S_i \) are spin-\( \frac{1}{2} \) operators at site \( i \), and \( \lambda \) is the alternating dimerization.

There is a considerable literature on this model, which has been reviewed recently by Barnes et al. [4]. At \( \lambda = 0 \), the system consists of a chain of decoupled dimers, and in the ground state each dimer is in a singlet state, while excited states are made up from the three triplet excited states on each dimer, with a finite energy gap. At \( \lambda = 1 \), we regain the uniform Heisenberg chain, which is gapless. Numerical studies of the model include series expansions [5,6,7], and exact diagonalizations for finite lattices [8,9,10].

The 2-magnon bound states were previously studied by Uhrig and Schulz [17] using an RPA approach. They found a singlet bound state below the 2-particle continuum for all momenta \( k \) and over the whole range of \( \lambda < 1 \). They also predicted a triplet bound state and a quintuplet antibound state near \( k = \pi/2 \) for \( \lambda \) not too large. These conclusions were supported in later studies [18,19,20]. Trebst et al. [21] found in a high-order series expansion study that in fact there are two \( S = 0 \) (\( S_1 \) and \( S_2 \)) and two \( S = 1 \) bound states (\( T_1 \) and \( T_2 \)), together with two \( S = 2 \) antibound states (\( Q_1 \) and \( Q_2 \)), if one goes sufficiently close to \( k = \pi/2 \). Hence the model displays some interesting multi-particle dynamics which can be explored both theoretically and in experiments. Neutron scattering experiments, however, will only be sensitive to the triplet bound states.

Barnes et al. [4] and Tennant et al. [2] have shown how to calculate exclusive structure factors for the 2-particle states by low-order series expansions, and have made a comparison with experimental data [22,23] for the copper nitrate material, \( Cu(NO_3)_2.2.5D_2O \).
Here we extend the series expansions to high orders. Series have been computed up to order $\lambda^{11}$ for the integrated structure factor $S(k)$, and the exclusive structure factors for the 1-particle triplet state $S_{1p}(k)$, the two 2-particle triplet bound states, $S_{T1}$ and $S_{T2}$, the 2-particle continuum $S_{2pc}$, and finally the total 2-particle structure factor (the sum rule) $S_{2p}$. The 1-particle structure factor has been computed to order $\lambda^{3}$ by Barnes et al. [4], but our series disagree with their results from second order.

The integrated structure factor for $\lambda = 0.27$, relevant to the material Cu(NO$_3$)$_2$·5D$_2$O, is shown as a function of wavevector $k$, in Fig. 1, along with some relative spectral weights. It can be seen that the structure factor peaks strongly at $kd = 2\pi$, and is dominated by the 1-particle state, which carries at least 96% of the total weight. The relative weight for more than two particles (i.e. $(S - S_{1p} - S_{2p})/S$) is less than 0.0069%.

The spectral weights for 2-particle triplet bound states $T_1$ and $T_2$ are given in Fig. 2. We know from previous studies that bound states exist only in a limited range of momenta near $kd = \pi$, and the spectral weight reflects this fact. The maximum relative spectral weight for $T_1$ is about 1.2% at $kd = \pi$, while the spectral weight for $T_2$ has two peaks, with a zero in between them, and has a very small maximum relative weight about 0.023%. The two-peaked structure can be traced back to the bound-state wave function for $\lambda \to 0$, where the two triplets are separated by an odd number of dimers [21].

Figure 3 shows the structure factor for the 2-particle continuum, also at $\lambda = 0.27$, sliced at various momentum intervals. Also shown as a solid curve is the dispersion relation for the triplet bound state $T_1$. It can be seen that where the bound state merges with the continuum, a very sharp peak develops at the lower edge of the continuum.

Figure 4 shows first, the position of the peak in the continuum structure factor relative to the lower edge, $\omega_p - \omega_L$, and secondly the peak value of the structure factor $S_{2pc}^p(\omega_p, k)$, as functions of $k$, calculated with finite lattices of 600, 1200 and 2400 sites in the Fourier transform [8]). It can be seen that at the threshold points where $T_1$ and $T_2$ merge with the continuum, $(\omega_p - \omega_L)$ goes quadratically to zero, and the peak value $S_{2pc}^p(\omega_p, k)$ shows spikes which appear to be diverging to infinity for the bulk system.

We have also studied the behaviour of the spectral weight as a function of $\lambda$. Figure 5 shows, for $kd = \pi$, the integrated structure factor $S$, the relative spectral weight of the 1-particle triplet state, the total 2-particle spectral weight, and those for the individual 2-particle
triplet bound states $T_1$ and $T_2$ and continuum. It can be seen that the relative spectral weight of the 1-particle triplet state approaches zero as $\lambda \to 1$, as expected, and the spectral weight for $T_2$ vanishes before $\lambda = 1$, while that for $T_1$ vanishes at $\lambda = 1$. The 2-particle continuum has about 54% weight at $\lambda = 1$, implying a large overlap between the two-triplet and the two-spinon descriptions in the uniform limit.

These results show that detailed and accurate results for structure factors and spectral weights can be obtained using high-order series expansions, as was also demonstrated for the spin-ladder model by Knetter et al. [12].

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