Cluster expansion for dimerized spin systems

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Abstract. We have studied dimerized spin systems by realizing the cluster expansion to high order. We have extended our previous dimer expansion for onedimensional systems to cover weakly interacting chains for a quantitative description of three dimensional materials like \((\text{C}_4\text{H}_{12}\text{N}_2)\text{Cu}_2\text{Cl}_6\) (= PHCC) and KCuCl\textsubscript{3}. By comparison with recent inelastic neutron scattering data we are able to determine the exchange energies between individual spins. We have further investigated the incommensurate regime of zigzag chains with isotropic exchange coupling constants near the disorder-line where the dispersion curve exhibits a minimum at a finite wavevector. Our approach clearly shows the gradual transition between the minimum of the dispersion at wavevector 0 and wavevector \(\pi\) within this region. The extent of the incommensurate regime is given analytically in an expansion in the coupling constants.

1 Introduction

Spin systems consisting of dimers as basic building blocks describe a large number of real materials when additional exchange couplings of smaller magnitude between dimers are considered. For a quantitative treatment of the elementary excitations of such systems we have developed the dimer perturbation theory \[1\] to cover dimers interacting in two and three spatial dimensions up to high orders in the interdimer interactions. In this contribution we present the application of this method to study quantitatively the elementary excitations in PHCC and KCuCl\textsubscript{3} (sect. \textsuperscript{3}) and to determine the incommensurate regime of the one-dimensional zig-zag chain close to the dimer point (sect. \textsuperscript{3}). The results for PHCC and KCuCl\textsubscript{3} were motivated by and will be compared to spectra obtained in recent inelastic neutron scattering experiments.

For weakly interacting dimers the cluster expansion is a powerful tool to calculate power series for properties like groundstate energy \[2\] or one magnon excitations \[4\]. Due to the large number of different exchange couplings the obtained power series have one parameter only which is an overall scaling of the exchange couplings and finally set to one. Different sets of exchange parameters require new calculations whereas the dependence in \(\mathbf{q}\)-space is given completely. The power series which are correct for the infinite system are build from finite clusters. In our case going to sixth order the number of topological different colored clusters is 18084 for KCuCl\textsubscript{3} and to fourth order 405 for PHCC.

In our notation the basic intradimer interaction (which sets the energy scale) is denoted by \(J\); interdimer interactions are denoted by \(J_{(lmn)}^{(ij)}\) with \((lmn)\) giving the direction between the dimer centers (in units of the lattice constants) and \((ij)\) specifying the pair of spins which interact with exchange energy \(J_{(lmn)}^{(ij)}\). The \(z\)-component of the vector from spin \(i = 1\) to spin \(i = 2\) on a given dimer is taken to be \(>0\).

Using this method, it is possible to determine the exchange interactions between the individual spins forming the dimers whereas the standard method to analyze systems of interacting dimers is an RPA-like approach \[5\] which allows to determine only the effective dimer interactions \(J_{(lmn)}^{\text{eff}} = (J_{(lmn)}^{(11)} + J_{(lmn)}^{(22)} - J_{(lmn)}^{(12)} - J_{(lmn)}^{(21)})/2\). This effective approach reproduces the leading terms of the full series; the full series, however, has additional terms starting in second order which turn out to be important.

2 Chains interacting in two and three dimensions: PHCC and KCuCl\textsubscript{3}

In this section we present our results for the real interacting dimer compounds \((\text{C}_4\text{H}_{12}\text{N}_2)\text{Cu}_2\text{Cl}_6\) (Piperazinium Hexachlorodicuprate = PHCC) and KCuCl\textsubscript{3}. In both these materials \(\text{Cu}^{2+}\)–ions with \(S = \frac{1}{2}\) are responsible for the magnetic properties.

PHCC was investigated recently by inelastic neutron scattering \[6\] and found to be effectively two-dimensional.
(i.e. \( J_{(lmn)}^{(ij)} = 0 \) for \( m \neq 0 \)). The compound can be considered as built from interacting spin ladders. The inelastic neutron scattering spectra were analyzed using the effective dimer model. Satisfactory agreement with the data was obtained when effective interactions \( J_{(lmn)}^{(ij)} \) with \( (lmn) = (200) \) and \( (002) \) were introduced as nonzero; however, no obvious exchange paths exist for these separations. In the following we present an analysis of the spectra with interactions \( J_{(lmn)}^{(ij)} \) between neighboring dimers only. The results shown in figure 3 are obtained with the set of exchange interactions given in table 1. A detailed analysis shows that for PHCC the additional contributions to the dimer series which are not taken into account by the effective dimer approach are essential to lower the dispersion curves. On the other hand we find from a comparison of the complete and the fourth order result of the effective approach that higher orders can be safely neglected.

![Fig. 1 Dispersion of PHCC for two directions: solid lines denote the result obtained from cluster expansion, circles are data taken from [6].](image)

The material KCuCl\(_3\) is similar in structure to PHCC. It contains, however, interdimer interactions also in the remaining (third) spatial direction. Inelastic neutron scattering experiments on this material have been published by two groups [7, 8] and have been interpreted using the effective dimer model. In earlier work [9] we analyzed these data using dimer series expansions to fourth order. The incommensurate regime is shown in figure 3 in infinite wavevector approximation of eqs. (1). The behaviour of the wave vector \( q_{\text{min}} \) corresponding to the minimum excitation \( E_{\text{max}} \) is similar in structure to PHCC. The compound can be considered as built from interacting spin ladders. The inelastic neutron scattering spectra were analyzed using the effective dimer model. Satisfactory agreement with the data was obtained when effective interactions \( J_{(lmn)}^{(ij)} \) with \( (lmn) = (200) \) and \( (002) \) were introduced as nonzero; however, no obvious exchange paths exist for these separations. In the following we present an analysis of the spectra with interactions \( J_{(lmn)}^{(ij)} \) between neighboring dimers only. The results shown in figure 3 are obtained with the set of exchange interactions given in table 1. A detailed analysis shows that for PHCC the additional contributions to the dimer series which are not taken into account by the effective dimer approach are essential to lower the dispersion curves. On the other hand we find from a comparison of the complete and the fourth order result of the effective approach that higher orders can be safely neglected.

![Fig. 2 Dispersion of KCuCl\(_3\) for two directions: solid lines denote the result obtained from cluster expansion, circles are data from [8].](image)

### 3 Incommensurate regime in zig-zag chains

In this section we consider a one-dimensional array of dimers forming an interacting zig-zag chain, \( J_{(100)}^{(11)} = J_{(100)}^{(22)} = J_2 \), \( J_{(100)}^{(21)} = J_1 \). This system has the interesting feature of an incommensurate regime where the minimum of the basic magnon dispersion curve is at finite wavevector \( q = q_{\text{min}} \), \( 0 < q_{\text{min}} < \pi \). The incommensurate regime is close to the disorder line (Shastry-Sutherland line) \( J_1 = 2J_2 \), where noninteracting dimers form the exact ground state, but requires slightly larger values of \( J_2 \) for given \( J_1 \). Using the dimer expansion to sixth order we have determined the limiting lines of the incommensurate regime as

\[
J_{1d}(J_2) \leq J_1 \leq J_{1u}(J_2)
\]

\[
J_{1d}(J_2) = 2J_2 - 2J_2^2 + 2J_2^3 - \frac{5}{2}J_2^4 + \frac{21}{4}J_2^5 + O(J_2^6)
\]

\[
J_{1u}(J_2) = 2J_2 - 2J_2^2 + 2J_2^3 - \frac{1}{2}J_2^4 + \frac{3}{4}J_2^5 + O(J_2^6)
\]

Thus the deviation from the Shastry-Sutherland line is of second order in the interdimer interaction whereas the width of the incommensurate regime is of fourth order. The incommensurate regime is shown in figure 3 in a comparison of the results from high order expansions (which agree with the findings from the diagonalization of finite systems using the Lanczos algorithm) and the approximation of eqs. (1). The behaviour of the wave vector \( q_{\text{min}} \), corresponding to the minimum excitation frequency is also shown for one path through the incommensurate regime in twelfth order (thus improving on the results shown in ref. [8]).

Evidently, the incommensurate regime is tiny and therefore so far mostly of academic interest; however, if a material becomes available which allows tuning the exchange parameters by e.g. alloying or applying external
pressure, our results provide a quantitative guide where to expect this regime of considerable qualitative interest.

Fig. 3 Incommensurate regime: The solid line denotes the Shastry-Sutherland line, the other lines are eqs. \[ \text{(i)} \] up to fifth order. The inset shows the wavevector \( q_{\text{min}} \) for the minimum value of the dispersion on the line \( J_1 = \frac{1}{2}(1.2 - J_2) \). All symbols are obtained from expansions up to twelfth order.

4 Conclusions

Using the dimer expansion after computer implementation we have investigated the low energy dynamics of the real materials PHCC and KCuCl\(_3\) and of the zig-zag chain, a model of theoretical interest in its incommensurate regime. The extent of the incommensurate regime in the zig-zag chain is satisfactorily obtained in an analytical approximation close to the dimer point and this result provides simple quantitative predictions for possible candidates for this interesting phase. The magnon spectra of PHCC and KCuCl\(_3\) as measured in inelastic neutron scattering experiments can be used to determine the exchange energies between the individual spins forming the basic dimers by applying the dimer expansion to fourth, resp. sixth order. KCuCl\(_3\) turns out to be appropriately described as a system of interacting chainlike structures with alternating couplings rather than ladders. For PHCC a consistent set of exchange energies is presented for interdimer interactions between dimers which are nearest neighbours only. For the material TICuCl\(_3\), which is similar to, but stronger interacting than KCuCl\(_3\), the application of the dimer expansion approach to the magnon spectra will be presented together with new experimental results \[ \text{\cite{ref}} \].

Table 1 Considered interaction constants of KCuCl\(_3\) and PHCC. The energy scale is given by \( J_{(000)} \).

| \( (nlm) \) | \( (ij) \) | \( J_{(nlm)}^{(ij)} \) | \( (ij) \) | \( J_{(nlm)}^{(ij)} \) |
|---|---|---|---|---|
| (100) | (11), (22) | 0.000 | (11), (22) | 0.32 |
| | (12) | 0.100 | (12), (21) | 0.08 |
| (201) | (21) | 0.188 | - | - |
| \( (1 \pm \frac{1}{2}) \) | (11), (22) | 0.200 | - | - |
| | (12), (21) | 0.040 | - | - |
| (001) | - | - | (11), (22) | 0.13 |
| | - | - | (12) | 0.00 |
| (101) | - | - | (12) | 0.09 |
| (10−1) | - | - | (21) | 0.09 |
| (000) | | 4.250 meV | 2.05 meV |

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