On the dynamics of coupled $S = 1/2$ antiferromagnetic zigzag chains

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Abstract. We investigate the elementary excitations of quasi-one-dimensional $S = \frac{1}{2}$ systems built up from zigzag chains with general isotropic exchange constants, using exact (Lanczos) diagonalization for 24 spins and series expansions starting from the decoupled dimer limit. For the ideal one-dimensional zigzag chain we discuss the systematic variation of the basic (magnon) triplet excitation with general exchange parameters and in particular the presence of practically flat dispersions in certain regions of phase space. We extend the dimer expansion in order to include the effects of three-dimensional interactions on the spectra of weakly interacting zigzag chains. In an application to KCuCl$_3$ we show that this approach allows us to determine the exchange interactions between individual pairs of spins from the spectra as determined in recent neutron scattering experiments.

1. Introduction

Spin systems consisting of chainlike or ladderlike structures as basic building blocks have recently attracted much attention. These systems are of interest on the one hand as one-dimensional (1D) model systems allowing one to study quantum phase transitions related to the existence of a spin gap and their dependence on the exchange parameters [1, 2]; on the other hand they describe an increasing number of real materials when an additional (small) exchange coupling in the remaining two dimensions is introduced [3]. A material of particular recent experimental interest is KCuCl$_3$ [4, 5].

We have performed an investigation of the dynamics of such systems with a twofold aim:

(i) We discuss the spectrum $\omega(q_x)$ of the low-lying triplet excitations in the ideal 1D system over a wide range of exchange parameters using both series expansions and exact diagonalization, in order to determine the range of applicability of the series expansion approach and to study the validity of using an effective interaction between dimers. We find and discuss in particular a regime in phase space with extremely small dispersion and a minimum of $\omega(q_x)$ at finite wavevector $0 < q_x < \pi$.

(ii) We extend the dimer expansion to include 3D couplings and apply this method in particular to a discussion of the dynamics of the quasi-zigzag-ladder material KCuCl$_3$ in terms of microscopic exchange parameters.

Of particular interest as a 1D building block for this type of material is the $S = \frac{1}{2}$ zigzag chain, as shown in figure 1 and defined by the following Hamiltonian:

$$H = \sum_{i=1}^{L} J \vec{S}_{1,i} \cdot \vec{S}_{2,i} + J_1 \vec{S}_{1,i} \cdot \vec{S}_{2,i+1} + J_2 (\vec{S}_{1,i} \cdot \vec{S}_{1,i+1} + \vec{S}_{2,i} \cdot \vec{S}_{2,i+1}). \tag{1}$$
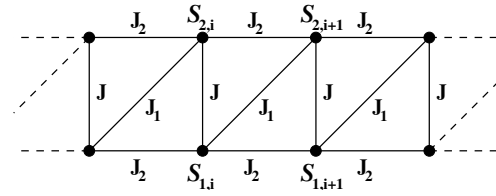
Figure 1. The zigzag chain as defined by (1).

On the theoretical side, this generic 1D model interpolates between a number of seemingly different limiting cases: it is an alternative way to formulate the Hamiltonian for the generalized $S = \frac{1}{2}$ spin ladder, generalized to include one diagonal interaction, or equivalently the $S = \frac{1}{2}$ chain with nearest-neighbour (NN) alternating exchange and next-nearest-neighbour exchange (we use the shorthand 'NNNA chain' in the following). It thus covers the well-known limiting models of the isotropic $S = \frac{1}{2}$ Heisenberg chain (HAF, $J = J_1, J_2 = 0$), the standard antiferromagnetic $S = \frac{1}{2}$ ladder ($J_1 = 0, J = J_2 > 0$), the weakly interacting dimer chain ($J_1, J_2 \ll J$) and the $S = 1$ antiferromagnetic (Haldane) chain ($J_1 \rightarrow -\infty, J + 2J_2 > 0$). It can alternatively be considered as a two-legged spin ladder with rung coupling J, leg coupling J_2 and additional diagonal coupling J_1.

The theoretical interest in the dynamics of the NNNA chain goes back to the work of Shastry and Sutherland [6], who identified the elementary excitations without alternation as free particles (spinons), which may become bound. A variational approach to the excitations of the NNNA chain based on this concept [7] has recently been shown to cover qualitatively the transition from free spinons to the Haldane triplet.

In a first approximation real materials are often considered as examples of 1D chains with a Hamiltonian as given in equation (1); then they realize different points in the phase diagram spanned by the interaction constants J_1, J_2 and show why it is of interest to describe systematically the variation of static and dynamic properties with the parameters J_1, J_2. Static properties, such as susceptibility and specific heat, however, have turned out to be rather insensitive to the details of the microscopic Hamiltonian [8] and, in the case of $(VO)_2P_2O_7$ [9–11], have even not been able to reveal the basic interactions as two instead of one dimensional.

Thus for a description of real materials a systematic microscopic treatment of the dynamics is of particular importance. In section 2 we present a systematic overview of the dynamics of the 1D system, discussing both general properties as well as comparing results from exact diagonalization to results from series expansions. In section 3 we use the series expansion approach to calculate the low-lying excitations in a 3D material with the structure of $KCuCl_3$ in terms of the microscopic Hamiltonian. This will allow us to go beyond the determination of effective dimer exchange parameters in recent work [4, 5] and to determine the microscopic exchange parameters. A summary will be given in the concluding section 4.

2. Elementary excitations of the 1D zigzag chain

We start with a short summary of the symmetries of the 1D system (the chain direction is denoted as the x-axis): translational symmetry is described by the wavevector q_x defined in a Brillouin zone $-\pi/a < q_x < +\pi/a$; the unit cell of length a contains two spins, or equivalently one dimer (one singlet in the limit $J_1 = J_2 = 0$). We thus use the conventional notation for ladders: a is the distance between rungs, whereas the distance between spins in

the NNNA-chain picture is $a/2$. We thus expect two basic excitations per unit cell. We will use units with $a = 1$ in the following. The excitation frequencies at wavevectors q_x and $-q_x$ are equal owing to reflection symmetry along the chain.

For special points in the phase diagram, additional symmetries exist:

- Without alternation ($J_1 = J$) it is natural to use a unit cell of length $\tilde{a} = \frac{1}{2}a$ containing only one spin. Our Brillouin zone is half of this Brillouin zone of the uniform chain and the excitations of the conventional spin chain will appear folded back to our smaller Brillouin zone.

- For the ladder symmetry ($J_1 = 0$) there exists a quantum number parity, P, resulting from the interchange of the two legs and we can classify states as positive or negative under this reflection. Each dimer in the singlet (triplet) state contributes a factor of -1 ($+1$) to this parity. An alternative notation introduces the component $q_\perp$ with values 0 (corresponding to $P = +1$) and π (corresponding to $P = -1$).

The ground state is a singlet in the whole phase plane and the lowest excited state is generally a triplet. The ground states for the ladder symmetry ($J_1 = 0$) have parity $P = +1$ for L even.

We have studied the dispersion $\omega(q_x)$ of the basic triplet excitation for a typical variety of paths in the J_1–J_2 parameter space by two methods:

(i) By exact numerical diagonalization, using the Lanczos algorithm, we have calculated $\omega(q_x)$ for the lowest excited states (between two and four states) for 24 spins, i.e. for seven different values of wavevector q_x.

(ii) We have performed series expansions around the dimer point, $J_1 = J_2 = 0$, up to third order analytically and up to tenth order after implementation of the cluster algorithm [12, 13] on an Alpha workstation. Thus we have obtained the ground-state energy E_0 and an effective Hamiltonian which can be diagonalized by a Fourier transformation. Finally we get the dispersion relation for the lowest excited state expressed as the series $\sum_n a_n \cos(nq_x)$.

In the following we present a number of results for the 1D zigzag chain which prepare the stage for the first application of the method to a non-trivial 3D system in section 3 and also add some new aspects to the large number of previous studies on the 1D system defined by equation (1) in recent years. To give a short review of existing work we mention first that the dimer series expansion approach started when the work of Brooks-Harris [14] was revived by Uhrig [15] in the context of $CuGeO_3$. The expansion for the triplet dispersion was extended to high orders recently by Oitmaa *et al* [16] for ladders to eighth order, by Barnes *et al* [17] for the Heisenberg alternating chain to ninth order and by Singh and Zheng [18] for the disorder line to the twenty-third order (using a special symmetry on this line). The model of equation (1) was also treated by alternative methods involving random-phase approximation [19], Brückner theory for the equivalent dilute Bose gas [20, 21], exact diagonalization [22] and continued-fraction expansion based on ED results [23] and the DMRG [24]. From these studies a rather complete picture of the low-energy dynamics of the 1D zigzag chain has emerged. In this section we supplement this picture with the following two remarks.

We start from the neighbourhood of the dimer point where an expansion in J_1, J_2 to low orders is sufficient. Up to third order the following result for the dispersion is obtained (as given in reference [15]; frequency and exchange constants are measured in units of the intradimer

exchange J from now on):

$$\omega(q_x) = 1 - \frac{J_1^2}{4}(1 + J_2) + \frac{3}{8}\left(J_2 - \frac{J_1}{2}\right)^2\left(2 + J_2 - \frac{J_1}{2}\right)$$
$$+ \left[J_2 - \frac{J_1}{2} - \frac{J_1^2}{4}(1 + J_2) - \frac{1}{4}\left(J_2 - \frac{J_1}{2}\right)^3\right]\cos q_x$$
$$- \frac{1}{4}\left(J_2 - \frac{J_1}{2}\right)^2\left(1 + J_2 + \frac{J_1}{2}\right)\cos 2q_x + \frac{1}{8}\left(J_2 - \frac{J_1}{2}\right)^3\cos 3q_x. \tag{2}$$

In first-order perturbation theory in J_1 and J_2, the spectrum is dispersionless on the Shastry–Sutherland line $J_1 = 2J_2$ (also known as the 'disorder line'). Equation (2) also shows the general feature that the location of the minimum of the dispersion curve shifts from $q_x = 0$ to $q_x = \pi$ somewhere close to crossing this line.

In recent approximate theoretical treatments of interacting dimer systems [5, 25] the exchange interactions between individual dimers were reduced to an effective interaction between dimers. For the present 1D zigzag chain the result of this approximation is that only the combination $J_2 - \frac{1}{2}J_1$ enters into the dispersion. In more detail, the result is

$$\omega(q_x) = \sqrt{1 + 2\,\delta\omega^{(1)}(q_x)} \tag{3}$$

where $1 + \delta\omega^{(1)}(q_x)$ is the dispersion in lowest order, i.e. the result of simple propagation of an excited dimer triplet without considering its coupling to higher-energy modes. Evidently this is true in lowest order of the expansion; it is seen, however, already from equation (2) that additional terms which depend on the individual exchange interactions enter in higher order. Comparing the higher-order coefficients in the series expansion we find that the effective dimer approximation of equation (3) amounts to keeping only the leading (i.e. lowest) powers in J_1, J_2 for each coefficient a_n of $\cos(nq_x)$.

For a quantitative check of the effective dimer approximation we refer first to reference [18] where the dispersion on the disorder line, i.e. for fixed $J_2 - \frac{1}{2}J_1 = 0$, is shown. The effective dimer approximation is reasonable for a large part of the line but deteriorates rapidly when the non-alternating limit $J_1 = 1$, i.e. the Majumdar–Ghosh point, is approached. For the more generic value $J_2 - \frac{1}{2}J_1 = \frac{3}{8}$ we show the dispersion in figure 2; it is seen that the effective dimer approximation is of very limited value in this case. In addition to these results, by comparing the series expansion results to spectra from exact diagonalization, we find the following limits of validity for the dimer series expansion method: whereas the tenth-order spectra coincide with exact-diagonalization results within a few per cent for $|J_1| \leqslant 1$ and $J_2 < 0.5$, the accuracy deteriorates rapidly when the symmetric ladder ladder configuration ($J_1 = 0$, $J_2 = 1$) is approached.

Whereas the dispersion is flat on the disorder line to first order in J_1, J_2, but develops maxima and minima in higher orders with an apparent jump of the minimum energy from $q_x = 0$ to $q_x = \pi$ close to this line, it has been noted already [2,7] from exact-diagonalization results that a regime with extremely flat dispersion and a minimum at finite wavevector $0 < q_x < \pi$ might exist in a narrow regime in J_1–J_2 parameter space. We have investigated this point again using the series expansions to tenth order, which provide us with a continuous wavevector dependence, and have confirmed the earlier speculation: in figures 3, 4 we show two examples for spectra on the lines $J_1 = 1 - 2J_2$ and $J_1 = \frac{1}{2}(1 - J_2)$ respectively; the points in figure 3 were discussed before in reference [2]. Figure 3 demonstrates that the shallow minimum of the dispersion curve at a wavevector in the middle of the Brillouin zone is reproduced in the series expansions. As a further example we show in figure 4 the flat dispersion for somewhat smaller values of J_1.
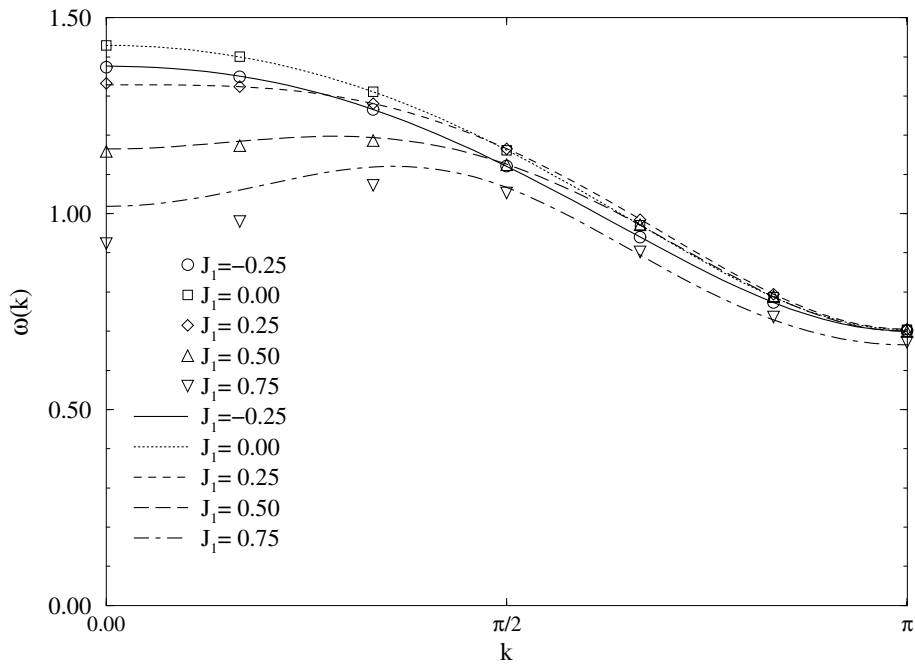
Figure 2. Comparison of excitation spectra for fixed effective dimer interaction $J_2 - \frac{1}{2}J_1 = \frac{3}{8}$.
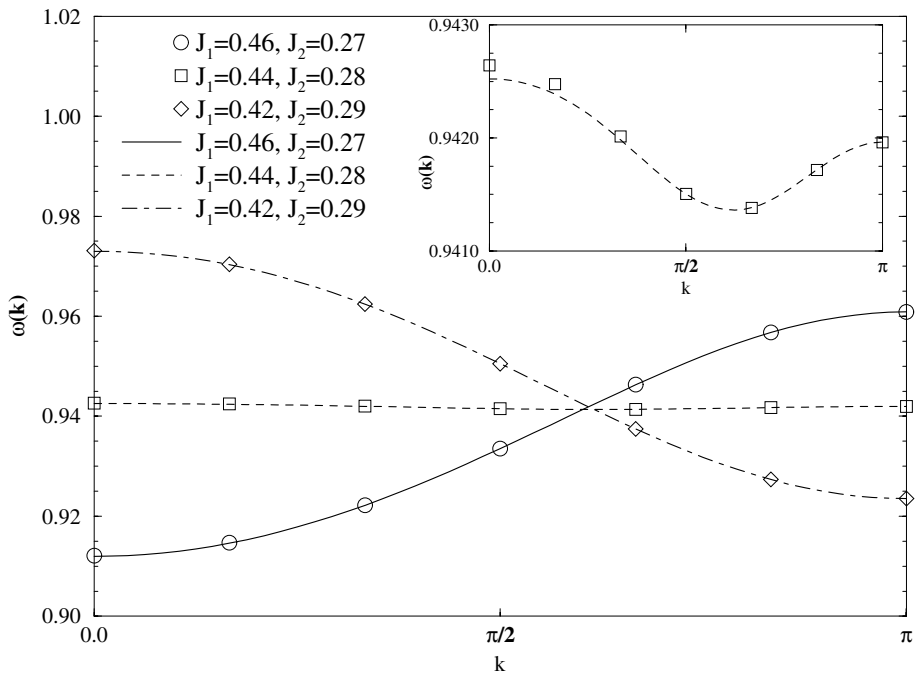
Figure 3. Variation of the dispersion curves when the incommensurate region is crossed. The inset corresponds to the 'flat' dashed curve: it shows with high resolution an example of a dispersion curve with a minimum at a finite wavevector as obtained from exact (Lanczos) diagonalization as well as from the dimer expansion method.
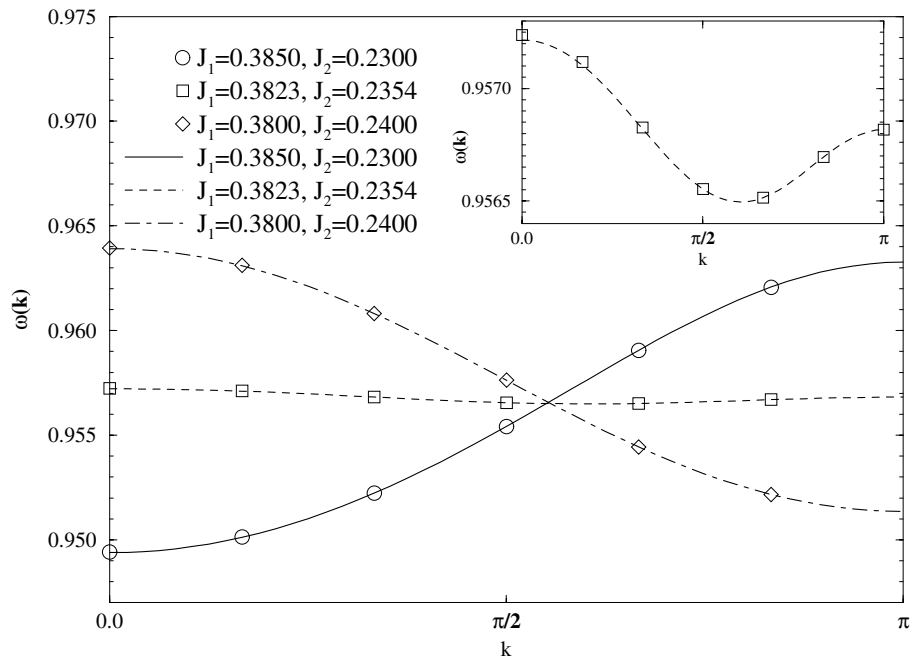
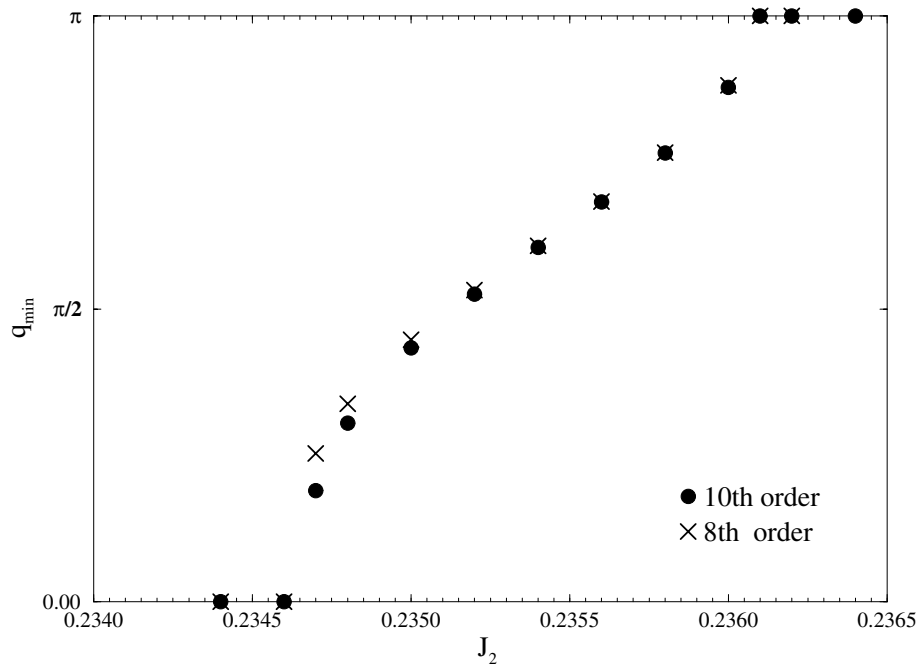**Figure 4.** As figure 3, but for different couplings.

Figure 5. The wavevector $q_{\min}$ for the minimum value of $\omega(q)$ on the line $J_1 = \frac{1}{2}(1 - J_2)$.

In figure 5 we show the development of the wavevector q_{min} for the minimum value of $\omega(q)$ on the line $J_1 = \frac{1}{2}(1 - J_2)$ in the J_1–J_2 parameter space (corresponding to the case of figure 4). The gradual transition of q_{min} between $q_{min} = 0$ and $q_{min} = \pi$, possibly with infinite slope at the end points, is clearly seen. The comparison between results in eighth and tenth orders illustrates the convergence of the expansion. At present, however, the series expansions do not give a hint as regards a possible fundamental reason for the nearly flat dispersion: we have examined the expansion coefficients a_n of $\cos(nq_x)$ up to tenth order, but we do not find any indication of a convergence to zero for $n \neq 0$.

3. Interacting zigzag chains

In real materials consisting of weakly interacting chains such as $KCuCl_3$ [4,5] and $CuGeO_3$ [26] it is clear from inelastic neutron scattering experiments that there is considerable dispersion for wavevectors perpendicular to the double-chain direction. For weakly interacting chains, the series expansion approach is to be considered as the only reliable systematic approach which (by comparing results in subsequent orders) allows a consistency check. Whereas the series expansions have been extended to cover systems coupled in 2D and interesting results have been obtained for the spin–Peierls material $CuGeO_3$ [15], for the 1/5 depleted square material CaV_4O_9 [27] as well as for general parameters [28], we present in the following the first results using the expansion for weakly interacting dimers for a 3D coupled system with particular application to investigating the magnon dispersion in the material $KCuCl_3$. The zigzag chain system $KCuCl_3$ actually appears to be closer to the dimer point than the systems treated so far and therefore is supposed to be a better candidate for this expansion.

The structure of $KCuCl_3$ is shown schematically in figure 6 in a projection to what is conventionally called the xz-plane: the fundamental dimers which are shown as solid lines form zigzag chains in the x-direction and neighbouring zigzag chains are shifted with respect to each other in the y-direction by half a lattice constant, $b/2$; this shift as well as the tilting of the internal dimer direction are indicated in figure 6 by giving the y-coordinate for each line of spins in the x-direction (n is an integer which numbers the different planes). The elementary cell consists of two dimers, dimer D_1 at the origin $\vec{R}_1 = 0$ and dimer D_2 at position $\vec{R}_2 = (0, \frac{1}{2}, \frac{1}{2})$, and the two spins forming each dimer, 1 and 2, are at positions $\vec{R}_i + \vec{d}_i$ for spin 1 and $\vec{R}_i - \vec{d}_i$ for spin 2; for definiteness we take $d_{i,z} > 0$.

For the exchange interactions between spins we will use the following notation: the main intradimer exchange is denoted as J. The exchange interaction per bond between spins in dimers separated by a lattice vector $\vec{R} = la\vec{e}_x + mb\vec{e}_y + nc\vec{e}_z$ is denoted as $J_{(lmn)}$ for the exchange between equivalent spins (pairs (11) or (22)) of the corresponding dimers and as $J'_{(lmn)}$ for the exchange between non-equivalent spins (pairs (12) or (21)). The following exchange interactions will be considered: $J_{(100)}$ for pairs (11) and (22), $J'_{(100)}$ for the pair (12) and $J'_{(201)}$ for the pair (12), and for the two cases $p = 0$ and $p = 1$: $J_{(p\frac{1}{2}\frac{1}{2})}$ for the pair (11) starting from dimer D_1 and for the pair (22) starting form dimer D_2, $J_{(p-\frac{1}{2}\frac{1}{2})} = J_{(p\frac{1}{2}\frac{1}{2})}$ for the pair (22) starting from dimer D_1 and for the pair (11) starting from dimer D_2, $J'_{(p-\frac{1}{2}\frac{1}{2})} = J'_{(p\frac{1}{2}\frac{1}{2})}$ for pairs (12) starting from either dimer D_1 or dimer D_2.

Alternatively we can look at the structure projecting on the plane spanned by the directions $\vec{e}_y$ and $\vec{e}_x + \frac{1}{2}\vec{e}_z$. In a schematic picture which shows the topology of the exchange interactions only, we obtain figure 7; the 3D structure of $KCuCl_3$ results when identical planes are stacked and the fundamental dimers are connected by zigzag interactions. Evidently the planar structure of figure 7 can be reduced to a number of limiting cases including coupled alternating chains ($J_{(1\pm\frac{1}{2}\frac{1}{2})} = 0$) or coupled ladders ($J_{(201)} = J'_{(1\frac{1}{2}\frac{1}{2})} = 0$).
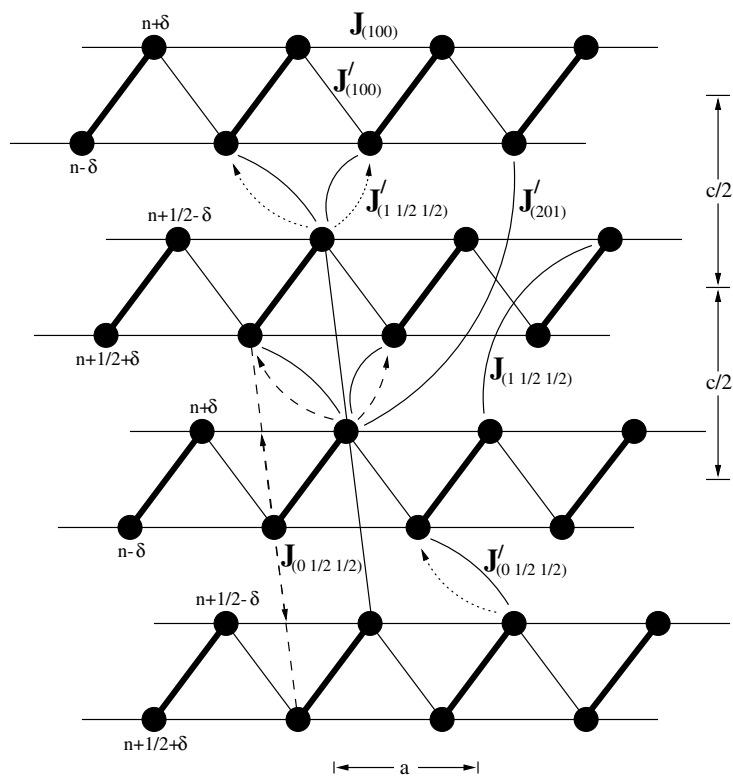
Figure 6. KCuCl$_3$ structure projected on the xz-plane. Thick full lines denote the basic dimers; the height above the reference plane $y = nb$ is given for the two spins of one dimer in each zigzag chain. Thin lines show interdimer interactions: full thin lines denote interactions of spins in dimers with identical values of n; dashed and dotted thin lines denote interactions of spins in dimers with different values of n. As indicated by the arrows, the dashed (dotted) lines start at n and end at $n - 1(n + 1)$. Only one interaction of each type is shown.
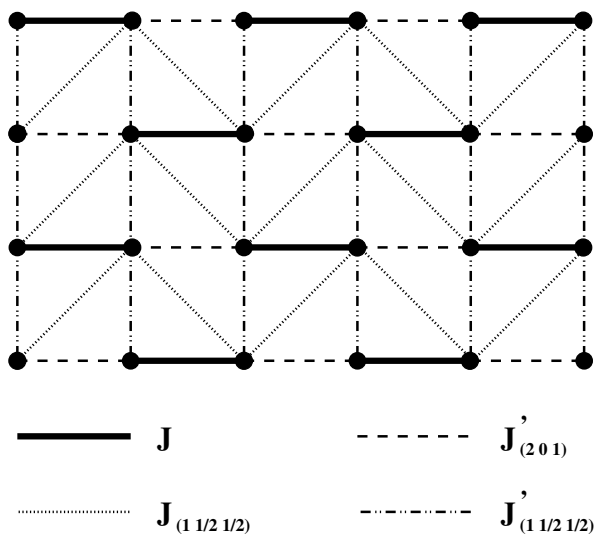

Figure 7. An alternative schematic view of the structure of KCuCl$_3$.

If the dispersion is considered only to first order [4] or if higher orders are included in an RPA-like approximation [5], J and J' for a given (lmn) enter only in the combination $J^{\text{eff}} = \frac{1}{2}(pJ - p'J')$, where $p, p' = 1, 2$ is the number of exchange paths between the interacting dimers; J^{eff} is called the effective dimer interaction. As already seen in the 1D case of section 2, a correct treatment beyond first order involves J and J' independently. According to previous work [4, 5] the main exchange interactions in addition to the basic intradimer exchange are between dimers separated by $(lmn) = (100), (201), (1 \pm \frac{1}{2}\frac{1}{2})$. Except for (201) these dimer–dimer interactions involve both J and J' and it is our aim in the following to discuss the validity of the effective dimer approximation for $KCuCl_3$ and to investigate to what extent the exchange interactions between individual spins can be determined from the present status of experimental results.

A calculation up to second order in the ratios $J_{(lmn)}/J$ leads to the following expression for the frequency of the basic triplet (frequency and coupling constants are measured in units of the basic dimer exchange constant J and wavevectors q_i are given in units with the crystallographic lattice constants a, b, c set equal to unity):

$$\omega(\vec{q}) = 1 + \delta\omega^{(1)}(\vec{q}) - \frac{1}{2}\delta\omega^{(1)2}(\vec{q}) + J_{(100)}(J_{(100)} - J'_{(100)}) - \frac{1}{4}J'^2_{(100)}\cos q_x$$

$$+ J_{(0\frac{1}{2}\frac{1}{2})}J'_{(0\frac{1}{2}\frac{1}{2})} + J_{(1\frac{1}{2}\frac{1}{2})}J'_{(1\frac{1}{2}\frac{1}{2})} + \frac{1}{2}(J^2_{(0\frac{1}{2}\frac{1}{2})} - J'^2_{(0\frac{1}{2}\frac{1}{2})})\cos\frac{q_y}{2}\cos\frac{q_z}{2}$$

$$+ \frac{1}{2}(J^2_{(1\frac{1}{2}\frac{1}{2})} - J'^2_{(1\frac{1}{2}\frac{1}{2})})\cos\frac{q_y}{2}\cos\frac{2q_x + q_z}{2} - \frac{1}{4}J'^2_{(201)}\cos(2q_x + q_z). \tag{4}$$

Here

$$\delta\omega^{(1)}(\vec{q}) = \frac{1}{2}(2J_{(100)} - J'_{(100)})\cos q_x + (J_{(0\frac{1}{2}\frac{1}{2})} - J'_{(0\frac{1}{2}\frac{1}{2})})\cos\frac{q_y}{2}\cos\frac{q_z}{2}$$

$$+ (J_{(1\frac{1}{2}\frac{1}{2})} - J'_{(1\frac{1}{2}\frac{1}{2})})\cos\frac{q_y}{2}\cos\frac{2q_x + q_z}{2} - \frac{1}{2}J'_{(201)}\cos(2q_x + q_z) \tag{5}$$

is the dispersion to first order in the exchange constants, i.e. the effect of simple propagation of an excited dimer triplet.

In equation (4) we have used for simplicity an extended-zone scheme: since there are two dimers in the proper crystallographic unit cell, there are two branches of triplet excitations for each wavevector in the crystallographic Brillouin zone ($-\pi < q_y b, q_z c \leqslant \pi$). Owing to the symmetry of the exchange interactions in the Hamiltonian, these two branches join to the unique smooth expression given in equation (4), when the doubled zone

$$-2\pi < q_z c + q_y b, q_z c - q_y b \leqslant 2\pi$$

is used. Each triplet excitation, however, is present at all equivalent wavevectors of the crystallographic reciprocal lattice. For a discussion of dipole transition amplitudes the non-interacting triplet approximation can be used as sufficient guide and gives the following results [5]: for momentum transfer perpendicular (parallel) to $\vec{e}_y$ the branch of equation (4) in the first (second) crystallographic Brillouin zone gives the only non-vanishing contribution. (A change form the first to the second crystallographic Brillouin zone then corresponds to a change in either q_y or q_z by 2π, i.e. to a change of sign of the interactions in the $(0 \pm \frac{1}{2}\frac{1}{2})$ and $(1 \pm \frac{1}{2}\frac{1}{2})$ directions.)

We note that the wavevector q_z should be distinguished from the quantity $q_\perp$ which is often used to denote the two values of the quantum number parity discussed in section 2 as $q_\perp = 0, \pi$. This wavevector is measured in units of $\tilde{c}^{-1}$, where $\tilde{c}$ is the rung length, which differs from the reduced lattice constant $c/2$. In experiments, so far only the basic triplet with $q_\perp = \pi$ has been observed and the interchange of minima with variation of q_z is an effect

of the crystallographic lattice geometry and not of the 1D ladder geometry. Excitations with $q_\perp = 0$ are excitations with two dimer quanta and have a minimum energy of twice the gap energy Δ.

In order to obtain results which are quantitatively reliable we have performed series expansions for the 3D coupled system to fourth order following the lines described in section 2. Because of the complex lattice we did not characterize the clusters which result in a large number of clusters: 5532 clusters in fourth order. The convergence of these series expansion results is excellent for the small values of the expansion parameters $J_{(lmn)}/J \leqslant 0.4$ in the application to $KCuCl_3$.

In figures 8 and 9 we show results for dispersions along typical lines in $\vec{q}$-space together with the data points from reference [5]. Dispersions are plotted only for the branch which has non-vanishing dipole transition amplitude in lowest order leading to $+$ or $-$ signs in
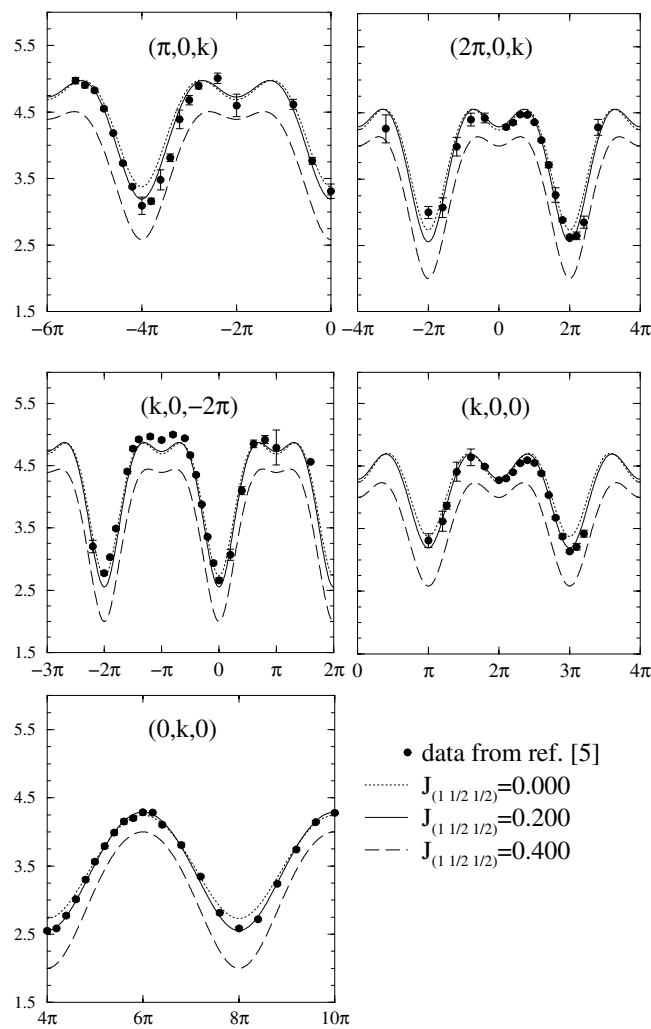

Figure 8. Dispersion curves for $KCuCl_3$ in various directions in $\vec{q}$-space showing the variation with $J'_{(1\frac{1}{2}\frac{1}{2})}$ at fixed effective dimer interaction $J_{(1\frac{1}{2}\frac{1}{2})} - J'_{(1\frac{1}{2}\frac{1}{2})} = 0.160$.
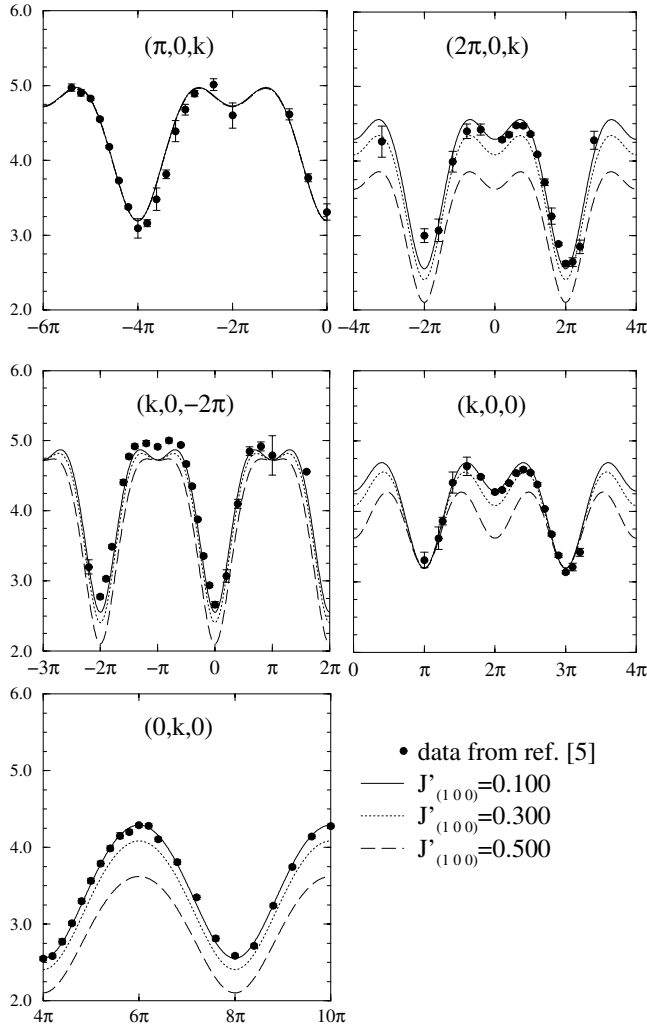
Figure 9. Dispersion curves for KCuCl$_3$ in various directions in $\vec{q}$-space showing the variation with $J'_{(100)}$ at fixed effective dimer interaction $J_{100} - \frac{1}{2}J'_{(100)} = -0.055$.

equation (4) as discussed above. We assume that the effective dimer exchange takes the values determined in previous work [4, 5] and have therefore fixed the following combinations of exchange parameters:

$$
\begin{aligned}
J'_{(201)} &= -2J^{\text{eff}}_{(201)} = 0.188 \\
2J_{100} - J'_{(100)} &= 2J^{\text{eff}}_{(100)} = -0.110 \\
J_{(1\frac{1}{2}\frac{1}{2})} - J'_{(1\frac{1}{2}\frac{1}{2})} &= 2J^{\text{eff}}_{(1\frac{1}{2}\frac{1}{2})} = 0.160.
\end{aligned}
\tag{6}
$$

The interactions $J_{(0,\frac{1}{2},\frac{1}{2})}$, $J'_{(0,\frac{1}{2},\frac{1}{2})}$ are found to be negligibly small. In order to demonstrate the relevance of the individual spin exchange parameters (as opposed to the effective description) we discuss the separations (100) and $(1 \pm \frac{1}{2}\frac{1}{2})$ independently: in figure 8 we present the variation of the dispersions $\omega(\vec{q})$ in selected directions in $\vec{q}$-space for different values of the coupling $J_{(1\frac{1}{2}\frac{1}{2})} = 0, 0.2, 0.4$ with the remaining parameters fixed as given

above. It is seen that the distribution of the effective dimer interaction between parallel and diagonal terms essentially shifts the dispersion curve by constant amounts. A comparison to the corresponding neutron scattering results leads to the conclusion that $J_{(1\frac{1}{2}\frac{1}{2})} = 0.200$ is the most likely value. The analogous results for different values of the diagonal (zigzag) coupling $J'_{(100)} = 0.1, 0.3, 0.5$ are shown in figure 9. Here the frequency $\omega(q_x = 0)$ (which is the energy gap for $q_z = \pi$ and the dip energy for $q_z = 0$) depends only on the effective interaction, whereas the frequency $\omega(q_x = \pi)$ (which is minimum for $q_z = 0$ and a dip energy for $q_z = 2\pi$) allows us to determine the exchange between individual spins.

Comparison of our series expansion results to the neutron scattering data then leads to the following values for the exchange constants for individual spins, not yet determined by the values for the effective dimer interactions published so far:

$$J_{(100)} \approx -0.005$$
$$J'_{(100)} \approx 0.100.$$

These values imply that the ladder system in $KCuCl_3$ is much closer to an alternating spin chain than was believed so far and the leg interaction tends to be ferromagnetic if it is non-zero at all. The results for the interchain interactions in the $(1\frac{1}{2}\frac{1}{2})$ direction are much less conclusive. The most likely values are

$$J_{(1\frac{1}{2}\frac{1}{2})} \approx 0.200$$
$$J'_{(1\frac{1}{2}\frac{1}{2})} \approx 0.040.$$

However, the error is large and the data may actually be compatible also with smaller values for $J_{(1\frac{1}{2}\frac{1}{2})}$.

4. Conclusions

We have investigated the dispersion curve for the low-energy triplet excitations of one-dimensional and of weakly coupled zigzag chains starting from the limit of non-interacting dimers and performing an expansion in the interdimer interactions. The series up to tenth order in the 1D case and up to fourth order in the 3D case were evaluated explicitly after implementation on a workstation. In the 1D case the dispersion curves agree with those obtained from exact diagonalization using the Lanczos algorithm for a large regime around the dimer point; in this regime the method provides a reliable approach for calculating the dispersion in its continuous dependence on the wavevector. In a narrow regime close to the Shastry–Sutherland line we find the minimum of the dispersion curve at an intermediate wavevector $k_{min}, 0 < k_{min} < \pi$. As an application of the 3D case we present the application of the method to the $KCuCl_3$ structure. Fitting to the dispersion as measured in inelastic neutron scattering experiments [5], we determine the exchange interchange interactions between individual spins in addition to the effective interaction between dimers determined before. It is shown that the effective dimer approximation, when treated in the random-phase approximation, sums up the leading powers of the dimer expansion.

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