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The Δ chain with different base–base and base–vertex interactions

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Abstract

On the basis of the recently determined crystallographic structure of the delafossite $YCuO_{2.5}$, we argue that the Cu–O network has nearly independent Δ chains, with however different interactions between the $s = 1/2$ spins, owing to the different angles, distances and coordinations of the Cu ions. Although band-structure calculations are still lacking, motivated by this observation we study here the sawtooth lattice for different ratios of the base–base and base–vertex interactions, J_{bb}/J_{bv}. By exact diagonalization and extrapolation to the infinite-size limit, we show that the elementary excitation spectrum is the same for total spins $S_{tot} = 0$ and 1, but not for $S_{tot} = 2$, and has a gap only in the interval $0.487\,42(2) \leqslant J_{bb}/J_{bv} \leqslant 1.53(1)$. The gap, dispersionless for $J_{bb} = J_{bv}$, acquires increasing k-dependence as the ratio J_{bb}/J_{bv} moves away from unity, with the minimum energy excitations for $k = 0$ ($k = \pi$) when $J_{bb}/J_{bv} < 1$ ($J_{bb}/J_{bv} > 1$). Finally, we show that the gap closure is related to the instability of the dimers in the ground state as the difference between the interactions increases.

1. Introduction

More than 20 years ago, Shastry and Sutherland (SS) [1] introduced a new class of quantum topological excitations: isolated defects separating different regions of broken translational symmetry. Since their proposal, the search for models and real systems showing this behaviour has not stopped. The typical example is the symmetric zigzag spin ladder, first addressed by Majumdar and Ghosh (MG) [2], in which nearest-neighbour (NN) triangles sharing a base site are also vertex–vertex coupled. Its lowest-energy excitations are *kinks* (K) and *antikinks* (AK), defects separating domains corresponding to one or the other of the twofold degenerate ground states, with similar characteristics and giving rise to a finite gap, $\Delta E \approx 0.234 J_1$ when the interaction J_2 between next-nearest neighbours (NNN) is half that of NN spins J_1 [3]. Then attention turned to the sawtooth or Δ chain, which consists of coupled $s = 1/2$ Heisenberg spins forming triangles aligned in a chain with a common base site, like the zigzag ladder but without the vertex–vertex coupling. Studies of this lattice [4, 5], all with bonds having

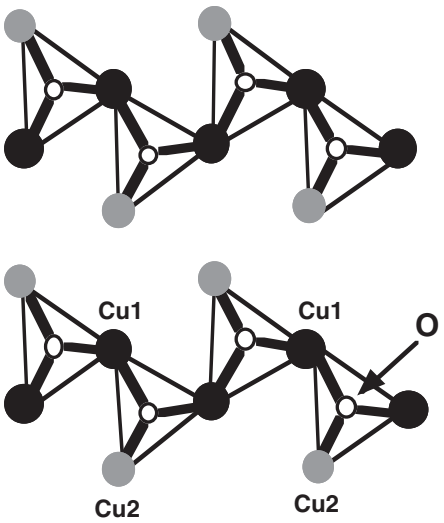

Figure 1. Sawtooth chains in the triangular Cu planes of the delafossite $YCuO_{2.5}$. The extra O ions (white circles) for $x = 0.5$ are located at the centres of particular triangles of Cu ions, creating AF superexchange only within these triangles. This gives nearly independent Δ chains, indicated by thin black lines. While Cu1 (black circles) adopts tetrahedral coordination with two O ions in this plane (and with two other O out of the plane), Cu2 (grey circles) adopts triangular coordination with just one O in this plane. The angles and distances [9] suggest a weaker J_{bb} interaction between Cu1–O–Cu1 bonds (bases of the triangles) than for the Cu1–O–Cu2 base–vertex bond J_{bv}.

the same interaction, have shown remarkable properties: the K–AK symmetry of the MG model is broken here, yielding however a similar dispersionless reduced gap for the low-lying excitation modes. Recently, the crossover from the MG model to the symmetric Δ chain has been discussed [6].

Despite all this theoretical work, there has to date been no clear physical realization of any of the various models discussed. However, experimental results that could display quantum topological excitations of the SS type have recently become available. Specifically, overdoped $RCuO_{2+x}$ (R = Y, La, etc) delafossites [7] have opened up new possibilities for studying frustrated hexagonal Cu planes with AF interactions between the Cu^{2+} ions. Depending on the O doping, different $s = 1/2$ effective lattices are obtained, although with weaker interactions than high-T_c systems, which have comparable bond lengths but 180° Cu–O–Cu angles. Studies [8] of the diluted Kagomé lattices for $x = 0.66$ predicted interesting properties. The recent synthesis of orthorhombic 2H single-phase samples of $YCuO_{2.5}$ has allowed us to elucidate its detailed structure [9], which appears as a nice realization of the sawtooth lattice (figure 1). The additional $x = 0.5$ O ions are located at the centre of alternating sets of triangles, providing superexchange paths between $s = 1/2$ spins on nearly independent Δ chains. However, the measured angles and distances indicate different interactions between the two spins on the base J_{bb} and between the base–vertex NN spins J_{bv} of the triangles. While the case $J_{bb} = J_{bv}$ has been studied theoretically [4, 5], to the best of our knowledge the case $J_{bb} \neq J_{bv}$ has not been considered before.

Therefore, we analyse here the sawtooth lattice for various ratios J_{bb}/J_{bv} of these AF couplings [10]. The Hamiltonian is given by

$$H = J_{bb} \sum_{i=1}^{N} \mathbf{s}_{2i-1} \cdot \mathbf{s}_{2i+1} + J_{bv} \sum_{i=1}^{N} (\mathbf{s}_{2i-1} \cdot \mathbf{s}_{2i} + \mathbf{s}_{2i} \cdot \mathbf{s}_{2i+1}), \tag{1}$$

Table 1. Lowest-energy excitations of the sawtooth lattice for $J_{bb} = J_{bv} = J$ with total spin S_{tot} and wavevector k, after extrapolation to $N \to \infty$. Units of J.

S_{tot}	Gap ($k = 0$)	Gap ($k = \pi/2$)	Gap ($k = \pi$)
0	0.2153(8)	0.22(1)	0.216(2)
1	0.2156(2)	0.214(10)	0.216(2)
2	0.46(1)	0.49(8)	0.46(6)

where N is the number of triangles ($2N$ spins) in the chain, and $\mathbf{s}_i$ is the spin-1/2 operator at site i. There has been no *ab initio* calculation for the J_{bb}/J_{bv} ratio in $YCuO_{2.5}$. Now, for either J_{bb} or $J_{bv} = 0$ the system is equivalent to the Heisenberg chain, while for $J_{bb} = J_{bv}$ we retrieve the symmetric Δ chain studied by Nakamura and Kubo [4] and by Sen *et al* [5]. Thus, to understand the $YCuO_{2.5}$ compound, it is important to study the entire evolution of the elementary excitations from the sawtooth lattice to the Heisenberg chain as a function of J_{bb}/J_{bv}. The transition between these two limits is not obvious: the symmetric Δ chain has a dispersionless small gap with K and AK excitations, while the isotropic Heisenberg chain has no gap and pairs of spinon excitations exhibiting a strongly dispersive spectrum.

2. Calculations and results

We are unable to solve analytically and exactly for the wavefunction and dispersion for arbitrary J_{bb}/J_{bv}, but important features of the spectrum can be obtained with high precision by exact diagonalization and extrapolation procedures. We diagonalize the spin Hamiltonian equation (1) via the Lanczos algorithm (all sizes from $N = 4$–12 triangles) using periodic boundary conditions. We find that the simplest and one of the best methods for extrapolating excitation energies to $N \to \infty$ is to take the finite-size term to be a polynomial in $1/N$, whose coefficients are determined by fitting.

To test this procedure, which includes larger clusters than before, we first briefly reconsider and extend results for the symmetric Δ chain. When $J_{bb} = J_{bv}$, equation (1) has two degenerate ground states with N dimers [11]. They may be written as states in which each spin on the base of a triangle forms a singlet either with the following vertex spin (right, R-dimer) or with the previous one (left, L-dimer state), that is,

$$|\mathrm{R}\rangle = \prod_{i=1}^{N} [2i-1, 2i], \qquad |\mathrm{L}\rangle = \prod_{i=1}^{N} [2i, 2i+1], \tag{2}$$

where $[i, j] \equiv (|\alpha_i \beta_j\rangle - |\beta_i \alpha_j\rangle)/\sqrt{2}$, with α_i (β_i) denoting states with $s_i^z = 1/2$ ($-1/2$) at site i. These two states are linearly independent and become orthogonal for $N \to \infty$. The existence of an excitation gap was also rigorously proved [11]. The elementary excitations are well-separated K–AK-type domain walls separating regions of R- and L-dimers [4, 5]. A K has a dimer in its triangle, while an AK does not. Curiously, they have very different characteristics in this system. A K has no excitation energy and is localized, but an AK propagates with kinetic energy within a region bounded by Ks. As a consequence of the former property the low-lying excitation spectrum is dispersionless, and owing to the second one, the gap is considerably reduced compared to the energy of a trivial triplet replacing a singlet dimer of the ground state.

Table 1 displays the gaps thus found for $k = 0, \pi/2$, and π for excitations with total spin $S_{tot} = 0$, 1 and 2, when $J_{bb} = J_{bv} = J$. Our results confirm the gap to be dispersionless. The gap for $k = 0$ and $S_{tot} = 1$, $\Delta E = 0.2156(2)J$, agrees with, but is more precise than, previous estimates [4, 5]. We also find that, within numerical error, the $S_{tot} = 0$ low-lying

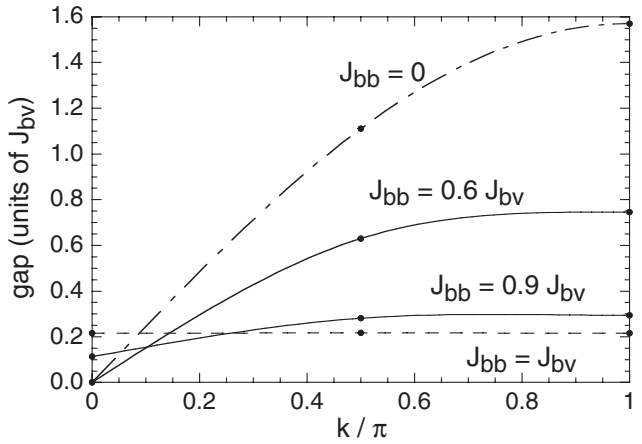

Figure 2. Low-lying excitation spectra of the sawtooth lattice for $J_{bb}/J_{bv} < 1$. The dashed line corresponds to the $J_{bb} = J_{bv}$ case, while the $J_{bb} = 0$ dotted–dashed curve follows the dispersion of the isotropic Heisenberg chain (see the text).

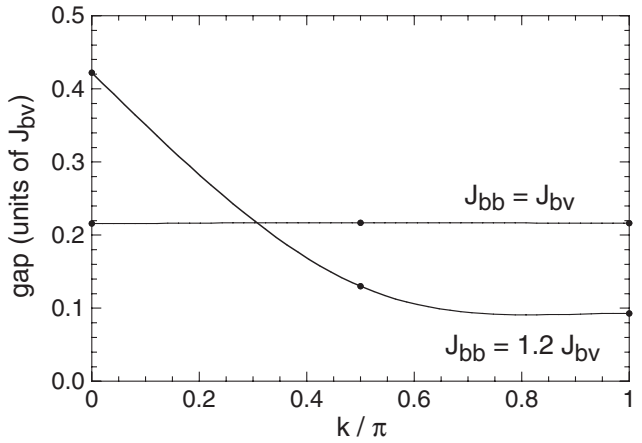

Figure 3. The dispersion curve for the gap to $S_{tot} = 1$ excited states of the sawtooth lattice with $J_{bb}/J_{bv} > 1$, compared to the $J_{bb}/J_{bv} = 1$ case.

excitations become degenerate with the $S_{tot} = 1$ gap as $N \to \infty$, as conjectured by Kubo [12]. Furthermore, the spectrum for $S_{tot} = 2$ appears also to be dispersionless, but with a gap about twice that for $S_{tot} = 1$ or 0. This new result is contrary to previous speculation [12] that the excitation energies for higher spins might converge to the same value as $N \to \infty$.

Figure 2 shows the low-lying triplet ($S_{tot} = 1$) excitation spectra for $J_{bb}/J_{bv} \leqslant 1$ (for $N \to \infty$). As J_{bb} decreases, the triplet excitation energy decreases at $k = 0$ until it vanishes near $J_{bb}/J_{bv} \approx 0.5$, while for $k = \pi$ it goes up. Progressively a stronger k-dispersion appears, yielding for $J_{bb} = 0$ the famous lower-boundary expression [13] for the continuum of excited triplet states for the isotropic $s = 1/2$ Heisenberg chain, $\Delta E_L(k) = (\pi/2)J_{bv}|\sin k/2|$ (here rewritten keeping the k definition for our Δ chain).

Figure 3 shows the evolution of the $S_{tot} = 1$ gap dispersion curves for $J_{bb}/J_{bv} > 1$. The minimum gap is now found for $k = \pi$ and decreases with increasing interaction ratio. On the other hand, for very large interaction ratios J_{bb}/J_{bv} the low-lying states become nearly

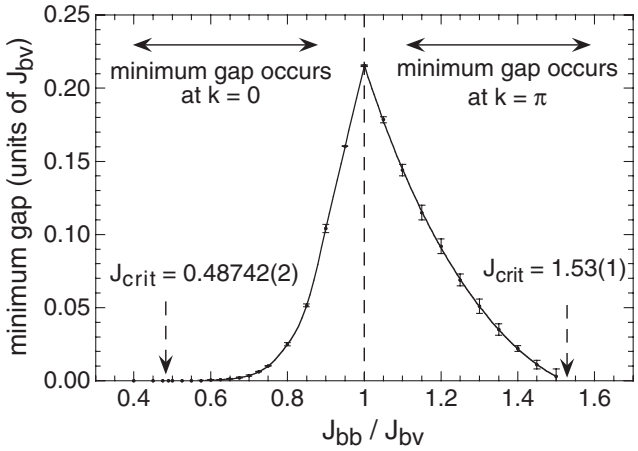

Figure 4. The gap to the lowest $S_{\rm tot} = 1$ excited states of the sawtooth lattice versus $J_{\rm bb}/J_{\rm bv}$ for $N \to \infty$. The error is greater for $J_{\rm bb}/J_{\rm bv} > 1$ because states with $k = \pi$ exist only for even N, giving fewer points in the extrapolation. See [10].

degenerate. This can be understood: we are again approaching the Heisenberg chain, though just for the N spins on the bases of the triangles, while the remaining N spins on the vertex are only loosely coupled, leading to a complex of 2^N nearly degenerate states. Correspondingly, the discussion of the critical ratio $J_{\rm bb}/J_{\rm bv} > 1$ for the closure of the gap becomes more complicated than above.

Figure 4 summarizes our main results for the low-lying excitations of the sawtooth chain. A finite gap is found only for interaction ratios within the interval $0.487\,42(2) \leqslant J_{\rm bb}/J_{\rm bv} \leqslant 1.53(1)$. Thus, the curve is not completely symmetric around $J_{\rm bb} = J_{\rm bv}$. The dispersionless gap found at this point becomes k-dependent, having its minimum value at $k = 0$ when $J_{\rm bb} < J_{\rm bv}$, and at $k = \pi$ when $J_{\rm bb} > J_{\rm bv}$.

As mentioned above, the ground state $|0\rangle$ when $J_{\rm bb} = J_{\rm bv}$ consists purely of singlets between NN spins, the R- or L-dimers; equation (2). In order to get some insight into its change of character as a function of $J_{\rm bb}/J_{\rm bv}$, we plot in figure 5 the ground state *dimerization fraction* $D_{\rm frac}$ for $J_{\rm bb}/J_{\rm bv} \leqslant 1$, defined as[3]

$$D_{\rm frac} = |\langle 0|{\rm R}\rangle|^2 + |\langle 0|{\rm L}\rangle|^2. \tag{3}$$

We calculate $D_{\rm frac}$ for up to 12 triangles; figure 5 shows the importance of the $N \to \infty$ extrapolation. We have tried several extrapolation methods and conclude that the numerical error in $D_{\rm frac}$ is of order 2% or less. In figure 5, $D_{\rm frac}$ drops from unity at $J_{\rm bb} = J_{\rm bv}$ (perfect dimerization) towards zero as $J_{\rm bb}/J_{\rm bv}$ decreases, showing that the gap reduction is related to increasing fluctuations of the dimer state. For $J_{\rm bb}/J_{\rm bv} < (J_{\rm bb}/J_{\rm bv})_{\rm crit}$, we find $D_{\rm frac} \leqslant 0.02$, which is our level of numerical error. Thus, as $J_{\rm bb}/J_{\rm bv}$ decreases, the sawtooth chain evolves from a gapped system with nonzero $D_{\rm frac}$ to a gapless system with $D_{\rm frac}$ equal to zero, within numerical error.

While our above discussion has been mainly for $S_{\rm tot} = 1$ excitations, we note that we have also been able to calculate many features of the spectra for $S_{\rm tot} = 0$ with a numerical accuracy of better than a few per cent, finding agreement with the $S_{\rm tot} = 1$ values in all cases. For example, we give in table 2 the singlet and triplet excitation energies for $k = 0$ and π for

[3] Since $|{\rm R}\rangle$ and $|{\rm L}\rangle$ are not orthogonal for N finite (but are linearly independent), for N finite we define $D_{\rm frac} = |\langle 0|{\rm R}'\rangle|^2 + |\langle 0|{\rm L}'\rangle|^2$, where $|{\rm R}'\rangle$ and $|{\rm L}'\rangle$ are two orthonormal states in the subspace spanned by $|{\rm R}\rangle$ and $|{\rm L}\rangle$.

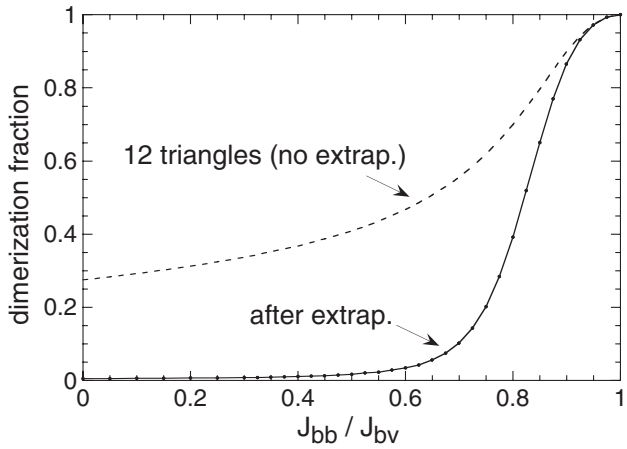

Figure 5. The ground state dimerization fraction (see the text) versus the interaction ratio.

Table 2. Excitation energies of singlet and triplet states. Units of J_{bv}.

J_{bb}/J_{bv}	$(S_{tot} = 0, k = 0)$	$(S_{tot} = 1, k = 0)$
0.3	0.000(1)	0.000(1)
0.6	0.000(1)	0.0004(1)
0.8	0.00(5)	0.025(1)
0.9	0.106(2)	0.104(3)
1.0	0.2153(8)	0.2156(2)
J_{bb}/J_{bv}	$(S_{tot} = 0, k = \pi)$	$(S_{tot} = 1, k = \pi)$
0.3	1.19(1)	1.204(4)
0.6	0.73(2)	0.743(2)
0.8	0.423(4)	0.425(2)
0.9	0.301(15)	0.304(15)
1.0	0.216(2)	0.216(2)

$J_{bb}/J_{bv} \leqslant 1$; they agree within numerical error. We also find agreement for $1 < J_{bb}/J_{bv} \leqslant 1.5$. Finally, for $N \to \infty$, the critical interaction ratios for singlet $[(J_{bb}/J_{bv})_{crit} = 1.51(3)]$ and triplet $[(J_{bb}/J_{bv})_{crit} = 1.53(1)]$ gap closure for $J_{bb}/J_{bv} \geqslant 1$ are also in agreement with each other, within numerical error. This provides strong numerical evidence that the lowest excitation spectra are in fact fourfold degenerate (for $N \to \infty$) for all $0 \leqslant J_{bb}/J_{bv} \leqslant 1.5$, thus generalizing the known results for the isotropic $s = 1/2$ Heisenberg chain [14] and the symmetric Δ chain.

3. Discussion

Now that good samples are available, measurements are in progress to distinguish the different interactions and to obtain a precise value of the gap, if one exists. It will also be worthwhile to synthesize single crystals of $YCuO_{2.5}$ to study the dispersion of the elementary excitations. Comparison with our results will then allow us to assess the applicability of the sawtooth lattice model to this system, and if appropriate to determine whether $J_{bb}/J_{bv} > 1$ or <1. We hope that this first study for $J_{bb} \neq J_{bv}$ will help with the interpretation of experiments for these interesting systems.

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