Eur. Phys. J. B **31**, 453–456 (2003)
DOI: 10.1140/epjb/e2003-00054-2

THE EUROPEAN PHYSICAL JOURNAL B

Quantum topological excitations: from the sawtooth lattice to the Heisenberg chain

S.A. Blundell[1,a] and M.D. Núñez-Regueiro[2,b]

[1] Département de Recherche Fondamentale sur la Matière Condensée, CEA-Grenoble/DSM
17 rue des Martyrs, 38054 Grenoble Cedex 9, France
[2] Laboratoire de Physique des Solides, bâtiment 510, Université Paris-Sud, 91405 Orsay Cedex, France

Received 29 October 2002 / Received in final form 14 January 2003
Published online 6 March 2003 –

Abstract. From the recently determined structure of the delafossite $YCuO_{2.5}$, we argue that the Cu-O network has nearly independent Δ chains but with different interactions between the $s = 1/2$ spins. Motivated by this observation, we study the Δ chain for different ratios of the base-base and base-vertex interactions, $J_{\rm bb}/J_{\rm bv}$. By exact diagonalization and extrapolation, we show that the elementary excitation spectrum is the same for total spins $S_{\rm tot} = 0$ and 1, but not for $S_{\rm tot} = 2$, and has a gap only in the interval $0.4874(1) \leq J_{\rm bb}/J_{\rm bv} \leq 1.53(1)$. The gap, known to be dispersionless for $J_{\rm bb} = J_{\rm bv}$, is found to acquire increasing k-dependence as $J_{\rm bb}/J_{\rm bv}$ moves away from unity.

PACS. 75.50.Ee Antiferromagnetics – 75.10.Jm Quantized spin models – 75.10.-b General theory and models of magnetic ordering – 75.40.Mg Numerical simulation studies

Rapid Note

There is great interest in $s = 1/2$ antiferromagnetic (AF) lattices including triangles, where the interplay between frustration and quantum effects yields interesting physics. Twenty years ago, Shastry and Sutherland [1] (SS) 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 behavior has not stopped. The typical example is the symmetric zigzag spin ladder, first addressed by Majumdar and Ghosh [2] (MG), in which nearest-neighbor (NN) triangles sharing a base site are also vertex-vertex coupled. Its lowest-energy excitations are so-called *kinks* (K) and *antikinks* (AK), defects separating domains corresponding to one or the other of the twofold degenerate ground state, both 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-neighbors (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, but without the vertex-vertex coupling. The Δ chain has unique properties. In fact, studies of this lattice [4,5], all with bonds having the same interaction, have shown a remarkable feature: 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 is a lack of clear physical realizations of any of the various models discussed. However, experimental results that hold the promise of displaying 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 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 much 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 of the $x = 0.66$ case predicted interesting properties. The recent synthesis [9] of orthorhombic 2H single-phase samples of $YCuO_{2.5}$ has allowed one to elucidate its detailed structure [10], which appears as a realization of the sawtooth lattice (Fig. 1). The additional $x = 0.5$ oxygen ions are located at the center of alternating sets of triangles, providing super-exchange paths between $s = 1/2$ spins on nearly independent Δ chains, as previously suggested [4,5]. However, by carefully considering the measured angles and distances, we conclude that the interactions between the two spins on the base $J_{\rm bb}$ and between the base-vertex NN spins $J_{\rm bv}$ of the triangles are different, most probably with $J_{\rm bb} < J_{\rm bv}$ owing to the smaller Cu-O-Cu angle. While the case $J_{\rm bb} = J_{\rm bv}$ has been quite extensively studied theoretically [4,5], to

[a] e-mail: sblundell@cea.fr
[b] e-mail: nunezreg@lps.u-psud.fr

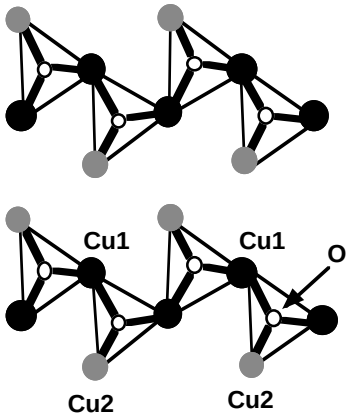

Fig. 1. Sawtooth chains in the triangular Cu planes of the delafossite $YCuO_{2.5}$. The extra O ions (white circles) for $x = 0.5$ are located at the center of particular triangles of Cu ions, creating AF super-exchange only within these triangles. This gives nearly independent Δ chains, shown by thin 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 (gray circles) adopts triangular coordination with just one O in this plane. The angles and distances [10] suggest a weaker $J_{\rm bb}$ interaction between Cu1-O-Cu1 bonds (bases of the triangles) than for the Cu1-O-Cu2 base-vertex bond $J_{\rm bv}$.

the best of our knowledge the case $J_{\rm bb} \neq J_{\rm bv}$ has never been considered before.

Therefore, we here analyze the sawtooth lattice for arbitrary ratios $J_{\rm bb}/J_{\rm bv}$ of these AF couplings. The Hamiltonian is given by

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

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 $J_{\rm bb}/J_{\rm bv}$ ratio in $YCuO_{2.5}$. Now, for either $J_{\rm bb} = 0$ or $J_{\rm bv} = 0$ the system is equivalent to the Heisenberg chain, while for $J_{\rm bb} = J_{\rm bv}$ 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 $J_{\rm bb}/J_{\rm bv}$. The transition between these two limits is not immediately 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.

We are unable to solve analytically and exactly for the wave function and dispersion for arbitrary $J_{\rm bb}/J_{\rm bv}$, but many of the important features of the spectrum can be obtained with high numerical precision by a careful exact diagonalization and extrapolation procedure. To test this method, which includes larger clusters than before, and to make contact with earlier work, we first briefly reconsider and extend results for the symmetric Δ chain ($J_{\rm bb} = J_{\rm bv}$). In this case, equation (1) has two degenerate ground states with N dimers [11]. These ground states 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 state) or with the previous one (left, L-dimer state), that is,

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

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

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

$S_{\rm tot}$	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)

We diagonalize the spin Hamiltonian in equation (1) by the Lanczos algorithm, using periodic boundary conditions with $\mathbf{s}_{2N+1}$ identified with $\mathbf{s}_1$. All sizes from $N = 4$–12 triangles are calculated. After some experimentation, including also forms containing exponentials, we found that the simplest and one of the best methods to extrapolate 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. Details will be given elsewhere. Table 1 displays the gaps found for $k = 0$, $\pi/2$, and π for excitations with total spin $S_{\rm tot} = 0$, 1, and 2, when $J_{\rm bb} = J_{\rm bv} = J$. (The wavevector k of an eigenstate $|\psi\rangle$ is here defined such that $T_n|\psi\rangle = \mathrm{e}^{\mathrm{i}kn}|\psi\rangle$, where T_n is the translation operator by n triangles or $2n$ spins.) The quoted numerical error, which arises entirely from the extrapolation, is defined to be twice the change in the result upon discarding the data of the largest system calculated (12 triangles) and repeating the extrapolation. Table 1 confirms the gap to be dispersionless, within numerical error [4,5]. The gap for $k = 0$ and $S_{\rm tot} = 1$, $\Delta E = 0.2156(2)J$, agrees with, but is more precise than,

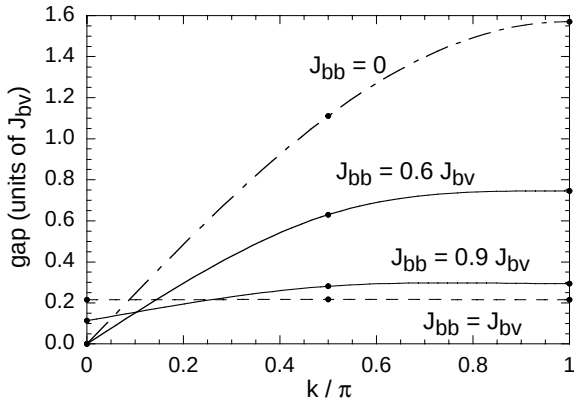

Fig. 2. Low-lying excitation spectra of the sawtooth lattice for $J_{\rm bb}/J_{\rm bv} < 1$. The dashed line corresponds to $J_{\rm bb} = J_{\rm bv}$, while the $J_{\rm bb} = 0$ dotted-dashed curve to the Heisenberg chain (using the definition of k for the Δ chain). The gap was calculated for the points shown by the dots, which were interpolated by a polynomial with the constraint of zero gradient at $k = \pi$.

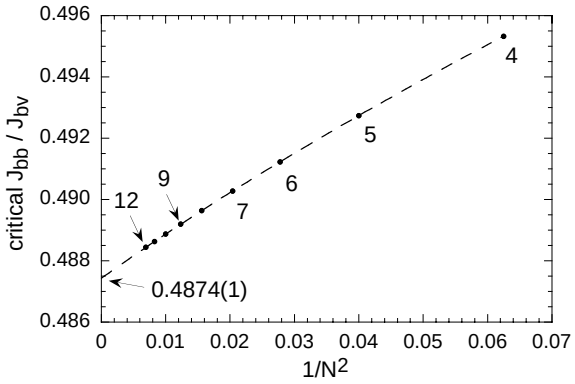

Fig. 3. Variation of the critical interaction ratio for gap closure when $J_{\rm bb}/J_{\rm bv} < 1$, as a function of the number of triangles N. The polynomial extrapolation in $1/N^2$ yields the best estimate of this critical value.

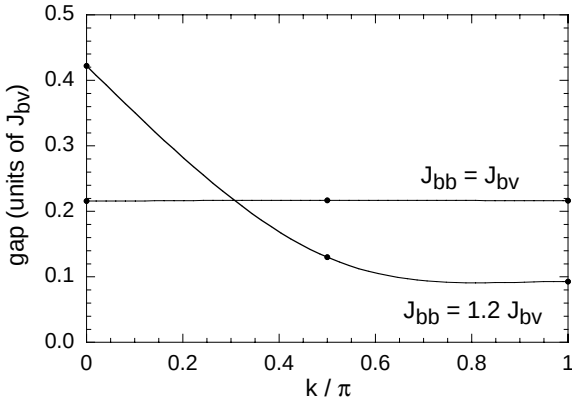

Fig. 4. Dispersion curve for the gap to $S_{\rm tot} = 1$ excited states of the sawtooth lattice with $J_{\rm bb}/J_{\rm bv} > 1$, compared to the $J_{\rm bb}/J_{\rm bv} = 1$ case.

previous estimates [4,5]. We also find that, within numerical error, the $S_{\rm tot} = 0$ low-lying excitations become degenerate with the $S_{\rm tot} = 1$ gap as $N \to \infty$, as conjectured by [12]. However, the spectrum for $S_{\rm tot} = 2$ appears also to be dispersionless with a gap about twice that for $S_{\rm tot} = 1$ or 0. This new result is contrary to a speculation [12] that the excitation energies for higher spins might converge to the same value as $N \to \infty$.

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

An accurate determination of the critical ratio $J_{\rm bb}/J_{\rm bv} < 1$ required to produce a triplet gap may be made by the method proposed in reference [14]. The idea is to map this problem to a continuum field theory and take into account the fact that in a fermion system with a fixed number of particles umklapp scattering is the only interaction that splits the degeneracy of the two lowest excited states. Therefore, the difference of their energies provides a precise measure of the umklapp processes, which vanish at the critical interaction ratio. For the Δ chain, with N finite and $J_{\rm bb} < J_{\rm bv}$, we find the first two excited states to have spin-parity 0^- and 1^+ and to undergo a level-crossing near $J_{\rm bb}/J_{\rm bv} \approx 0.5$. As shown in Figure 3, the value of $J_{\rm bb}/J_{\rm bv}$ at the level crossing follows a polynomial in $1/N^2$, and its extrapolation to $N \to \infty$ yields $(J_{\rm bb}/J_{\rm bv})_{\rm crit} = 0.4874(1)$.

Turning to the case $J_{\rm bb}/J_{\rm bv} > 1$, we show in Figure 4 how the $S_{\rm tot} = 1$ gap dispersion curves are modified. The minimum gap is now found for $k = \pi$ and decreases with increasing interaction ratio. On the other hand, for large interaction ratios the low-lying states become nearly 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. The evaluation of the critical ratio for the closure of the gap thus becomes more complicated (details will be given elsewhere), but an approach related to that used above gives $(J_{\rm bb}/J_{\rm bv})_{\rm crit} = 1.53(1)$.

Figure 5 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 $0.4874(1) \leq J_{\rm bb}/J_{\rm bv} \leq 1.53(1)$. The curve is asymmetric about $J_{\rm bb} = J_{\rm bv}$.

While our above discussion of the excitation gap has been explicitly just 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 percent, finding agreement with the $S_{\rm tot} = 1$ values in all cases. Examples where accurate calculations are possible: the excitation energy at $k = \pi$ for $0 \leq J_{\rm bb}/J_{\rm bv} \leq 1.5$ and at $k = 0$ for $0.9 \leq J_{\rm bb}/J_{\rm bv} \leq 1.0$. Also, for $S_{\rm tot} = 0$ gap closure we find $(J_{\rm bb}/J_{\rm bv})_{\rm crit} = 1.51(3)$

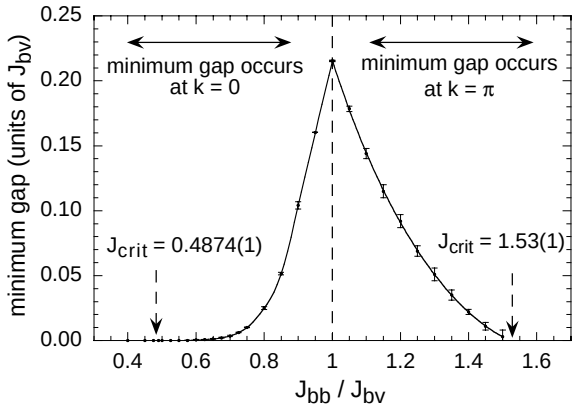

Fig. 5. Gap to the lowest $S_{\rm tot} = 1$ excited states of the sawtooth lattice *vs.* $J_{\rm bb}/J_{\rm bv}$, after extrapolation to $N \to \infty$. The error is greater for $J_{\rm bb}/J_{\rm bv} > 1$ because states with $k = \pi$ exist only for even N, giving less points in the extrapolation.

compared to $(J_{\rm bb}/J_{\rm bv})_{\rm crit} = 1.53(1)$ for $S_{\rm tot} = 1$. This provides strong numerical evidence that the lowest excitation spectra are in fact four-fold degenerate (in the limit $N \to \infty$) for all $0 \leq J_{\rm bb}/J_{\rm bv} \leq 1.5$, like the isotropic $s = 1/2$ Heisenberg chain [15] and the symmetric Δ chain limits. Further details will be published elsewhere.

The recent crystallographic study of $YCuO_{2.5}$ gives values for the three sides of the triangles and for the Cu-O-Cu angles. The consideration of these parameters and of the different coordination of the Cu ions (see Fig. 1 and Ref. [10]) justifies taking the same base-vertex interaction $J_{\rm bv}$ for both sides of the triangle, but a different $J_{\rm bb}$ along the base, as done here. The effect of a stronger AF interaction in just one of the base-vertex bonds will be to stabilize the dimer singlet ordering corresponding to that direction, lifting the degeneracy of the gapped ground state [16].

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 $YCuO_{2.5}$ in order to study the dispersion of the elementary excitations. Comparison with Figures 2 and 4 will then allow one to assess the applicability of the sawtooth lattice model, equation (1), to this system, and if appropriate to determine directly whether $J_{\rm bb}/J_{\rm bv} > 1$ or $J_{\rm bb}/J_{\rm bv} < 1$. We hope that this first study for $J_{\rm bb}/J_{\rm bv} \neq 1$ will help with the interpretation of the experimental results for these interesting systems.

We acknowledge D. Sen for useful correspondence. M.D. N.-R. is indebted to the Grenoble High Magnetic Field Laboratory for hospitality at the beginning of this work.

References

1. B.S. Shastry, B. Sutherland, Phys. Rev. Lett. **47**, 964 (1981)
2. C.K. Majumdar, D.K. Ghosh, J. Math. Phys. **10**, 1388 and 1399 (1969); C.K. Majumdar, J. Phys. C **3**, 911 (1970)
3. W.J. Caspers, K.M. Emmett, W. Magnus, J. Phys. A **17**, 2687 (1984)
4. T. Nakamura, K. Kubo, Phys. Rev. B **53**, 6393 (1996), and references therein
5. D. Sen, B.S. Shastry, R.E. Walsted, R.J. Cava, Phys. Rev. B **53**, 6401 (1996)
6. S. Chen, H. Buttner, J. Voit, Phys. Rev. Lett **87**, 087205 (2001); `cond-mat/0201004`; S. Sarkar, D. Sen, Phys. Rev. B **65**, 172408 (2002)
7. R.J. Cava *et al.*, J. Solid State Chem. **104**, 437 (1993)
8. M.E. Simón, A.A. Aligia, M.D. Núñez-Regueiro, Phys. Rev. B **51**, R15642 (1995); M.D. Núñez-Regueiro, C. Lacroix, B. Canals, Phys. Rev. B **54**, R736 (1996)
9. O. Garlea, Thesis Grenoble University (2001)
10. G. Van Tendeloo *et al.*, J. Solid State Chem. **156**, 428 (2001)
11. F. Monti, A. Süto, Phys. Lett. A **156**, 197 (1991); Helv. Phys. Acta **65**, 560 (1992)
12. K. Kubo, Phys. Rev. B **48**, 10552 (1993)
13. J. des Cloizeaux, J.J. Pearson, Phys. Rev. **128**, 2131 (1962)
14. J.L. Black, V.J. Emery, Phys. Rev. B **23**, 429 (1981); V.J. Emery, C. Noguera, Phys. Rev. Lett **60**, 631 (1988)
15. P. Fazekas, P. Süto, Solid State Commun. **19**, 1045 (1976)
16. A similar problem occurs in $CuGeO_3$, where the ground state degeneracy is lifted by a small structural distortion; see D. Augier, D. Poilblanc, E. Sorensen, I. Affleck, Phys. Rev. B **58**, 9110 (1998); G. Castilla, S. Chackravarty, V. J. Emery, Phys. Rev. Lett. **75**, 1823 (1995)

Rapid Note