Heat capacity of the twisted spin tube $[(CuCl_2tachH)_3Cl]Cl_2$*

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Introduction

Spin tubes constitute a special class of quasi-one dimensional spin ladder systems characterized by periodic boundary conditions in the rung direction. The magnetic compound $[(CuCl_2tachH)_3Cl]Cl_2$ [1] is a geometrically frustrated triangular spin tube, the frustration being related both to the triangular arrangement of its rungs and to the twisted geometry of the legs [see Fig. 1(a)].

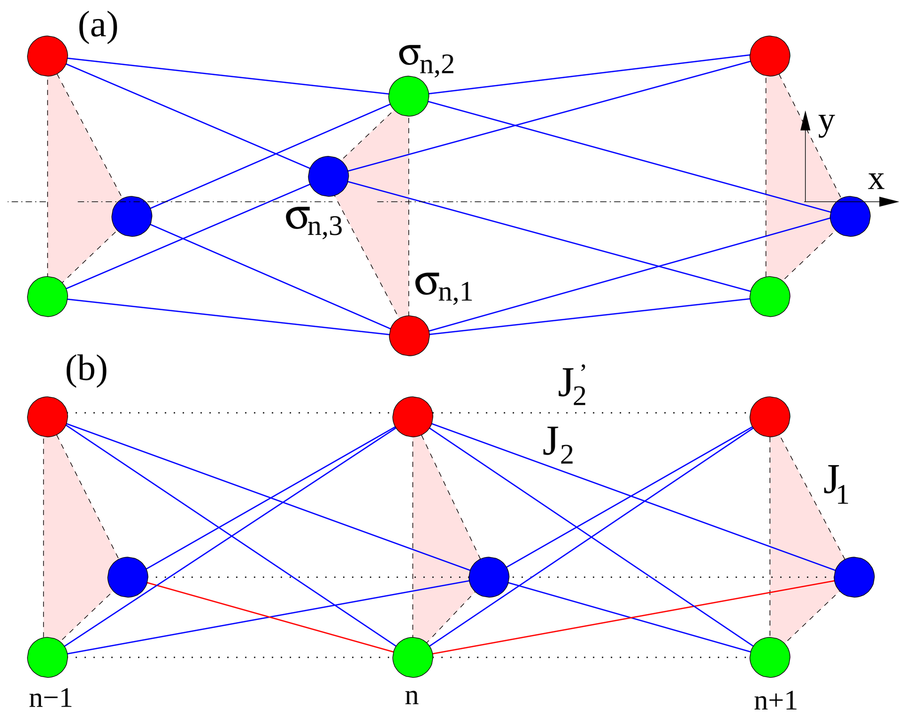

Figure 1: Sketch of (a) the twisted spin-tube system and (b) the equivalent spin model. J_2' is added for clarity.

The Hamiltonian describing the magnetic properties of $[(CuCl_2tachH)_3Cl]Cl_2$ reads

$$\mathcal{H}=\sum_{\alpha,n=1}^{L}\left[J_1\,\boldsymbol{\sigma}_{n,\alpha}\cdot\boldsymbol{\sigma}_{n,\alpha+1}+J_2\,\boldsymbol{\sigma}_{n,\alpha}\cdot(\boldsymbol{\sigma}_{n+1,\alpha+1}+\boldsymbol{\sigma}_{n+1,\alpha-1})\right].$$

Here $\boldsymbol{\sigma}_{n,\alpha}$ ($\alpha=1,2,3$) are spin-1/2 operators. n is the rung index ($n=1,\ldots,L\equiv N/3$). The twisted spin tube may also be thought of as a three-leg ladder with periodic boundary conditions in the rung direction. $[(CuCl_2tachH)_3Cl]Cl_2$ is characterized by the parameters $J_1/k_B = 1.8$ K and $J_2/k_B = 3.9$ K [2].

In the case $J_1/J_2 \gg 1$, $\mathcal{H}$ maps onto an effective spin-chirality model with additional chirality degrees of freedom, whereas for $J_1/J_2 \ll 1$ it maps onto an effective spin-3/2 antiferromagnetic (AF) Heisenberg chain containing additional biquadratic exchange couplings [3]. Below we demonstrate that in the spin tube material $[(CuCl_2tachH)_3Cl]Cl_2$, characterized by $J_1/J_2 = 0.46$, the second scenario with the effective exchange coupling $J_{\rm eff} = 2J_2/3$ is realized.

At higher temperatures, the measured specific heat exhibits a big Schottky-type peak located around $T \approx 2$ K. A detailed analysis implies that the main contribution to the specific-heat peak comes from the lowest-lying gapped magnon-type excitations resulting from the internal degrees of freedom of the composite rung spins.

Low-lying spin excitations

The contributions to the low-temperature specific heat of the spin tube material $[(CuCl_2tachH)_3Cl]Cl_2$ are dominated by three types of low-lying spin excitations. Apart from the gapless excitations, characteristic of any half-integer AF Heisenberg chain, important contributions come from two additional gapped branches of low-lying magnon-like modes related to the chirality degrees of freedom of the local triangles.

A qualitative picture of the low-lying spin excitations gives the semiclassical theory based on the Néel configuration $|S_t, -S_t, \ldots\rangle$, where S_t is the maximal value of the rung spin. The solid curves $E_m(k_x)$ in Fig. 2 show three branches of spin-wave excitations holding the rung wave vectors $k_y = 2\pi m/3$ ($m = 1, 2$, and 3):

$$E_0(k_x) = v_s|\sin k_x|, \quad v_s = (6S)\frac{2J_2}{3}, \tag{1}$$

$$E_{1,2}(k_x) = \sqrt{\Delta^2 + 4S^2J_2^2\sin^2\left(k_x \mp \frac{2\pi}{3}\right)}. \tag{2}$$

Here S is the on-site spin and $\alpha = J_1/J_2$. The gap Δ measures the energy of the lowest-lying excitations at $k_x = \pi/3$ and $2\pi/3$. $E_0(k_x)$ coincides with the semi-classical results describing the gapless excitations of a spin-($3S$) AF Heisenberg chain with the exchange constant $J_{\rm eff} = 2J_2/3$.

Using the approach of Ref. [4], the following estimates for v_s and Δ result from extrapolations of the exact-diagonalization (ED) data ($L = 6, 8, 10$, and 12) (see Fig. 3):

$$v_s/k_B = 10.06\,{\rm K} = 3.87\,\frac{2J_2}{3k_B}\,,\quad \Delta/k_B = 5.32\,{\rm K}\,. \tag{3}$$

$3v_s/(2J_2) = 3.87$ exactly reproduces the density-matrix renormalization group estimate [4] concerning the spin-3/2 AF Heisenberg chain with an exchange constant $2J_2/3$.

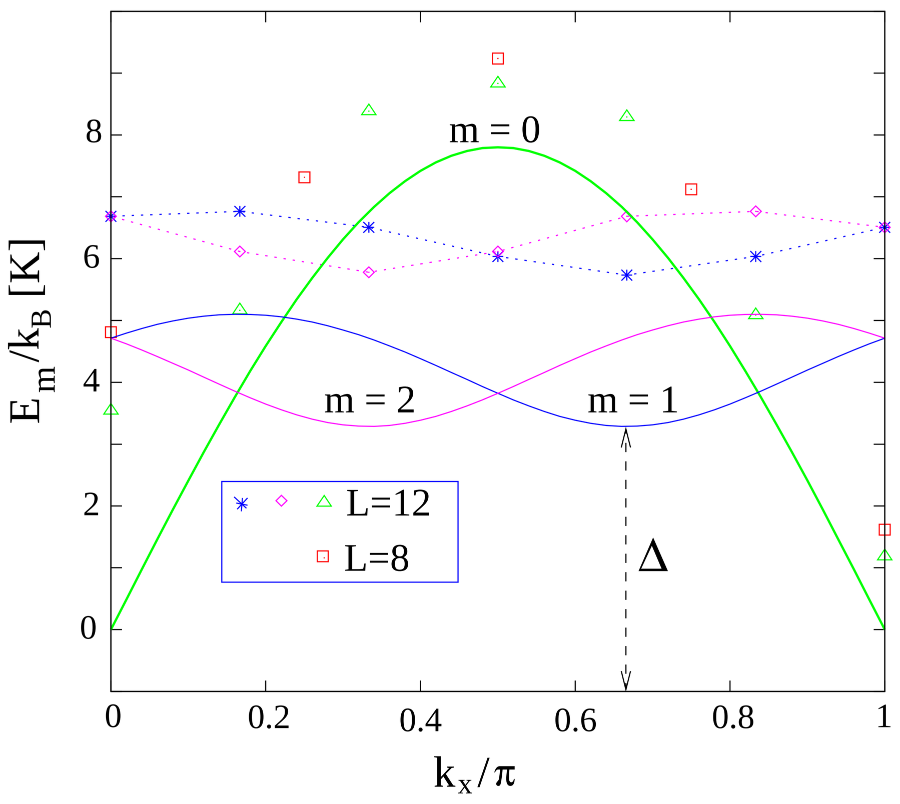

Figure 2: Spin-wave excitation modes, Eqs. (1) and (2), compared with the lowest-lying triplet excitations in two periodic clusters.

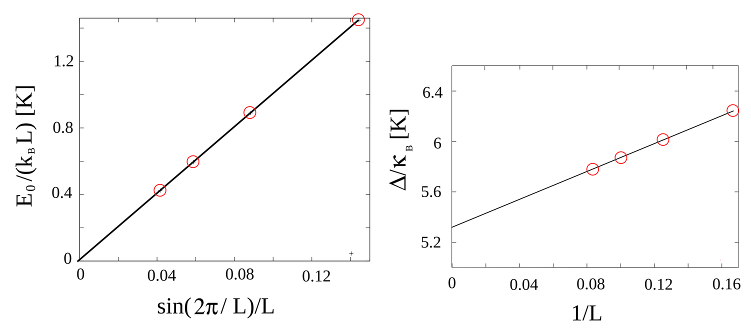

Figure 3: Extrapolation of the ED data giving the estimates for v_s (l.h.s.) and Δ (r.h.s.) in Eq. (3).

Low-temperature specific heat

The specific heat, Figs. 4 and 5, is measured at the Institute for Materials Research (IMR) of Tohoku University using powder samples of $[(CuCl_2tachH)_3Cl]Cl_2$. The solid curve in Fig. 4 depicts $C(T)$ of a spin-3/2 periodic AF Heisenberg chain ($L = 100$) with the exchange constant $J_{\rm eff} = 2/3J_2$, as obtained from a quantum Monte Carlo (QMC) method employing the ALPS code [5]. As can be seen in Fig. 4, the QMC result nicely reproduces the experimental data in the region $T \le 0.5$. In a recent report, Nuclear Magnetic Resonance measurements also indicate a gapless spin state in the same material based on estimates for the extremely low-temperature part of the magnetic susceptibility [6].

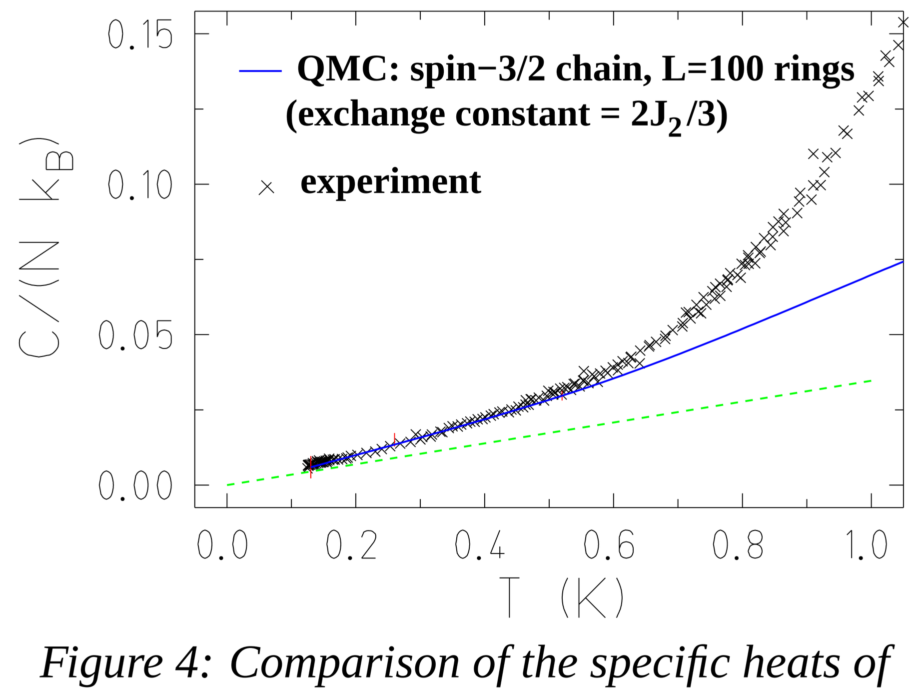

Figure 4: Comparison of the specific heats of $[(CuCl_2tachH)_3Cl]Cl_2$ and of a spin-3/2 AF chain with the exchange constant $2J_2/3 = 2.6$ K. The dashed line is the specific heat of a Tomonaga-Luttinger liquid, $C(T)/(Nk_B) = \pi T/(9v_s)$, for $v_s = 10.06$ K.

To fit the experimental data in Fig. 5, we use the two-level approximation $E_{1,2}(k_x) \approx \Delta$, which produces the Schottky-type expression

$$\frac{C}{Nk_B} = A\frac{r(\Delta/T)^2\exp(\Delta/T)}{[\exp(\Delta/T)+r]^2}\,. \tag{4}$$

Here $r = 2$ reflects the double degeneracy of the excited state (in respect to the ground state) and A is an overall parameter used to fix the height of the Schottky peak. The related curve with $\Delta/k_B = 5.32$ K is plotted in Fig. 5 by a thick line together with the experimental data.

One observes that it reproduces very well not only the position of the peak ($T_m \approx 2$ K) but also the behavior of the specific heat down to $T \approx 0.7$ K. However, turning to higher temperatures, one indicates pronounced discrepancies with the numerical results related with the increased phonon contribution to $C(T)$: since the detailed phonon spectral density is unknown, the raw experimental data for $C(T)$ was – as usually –corrected by subtracting a reasonable Debye-like specific heat contribution [7].

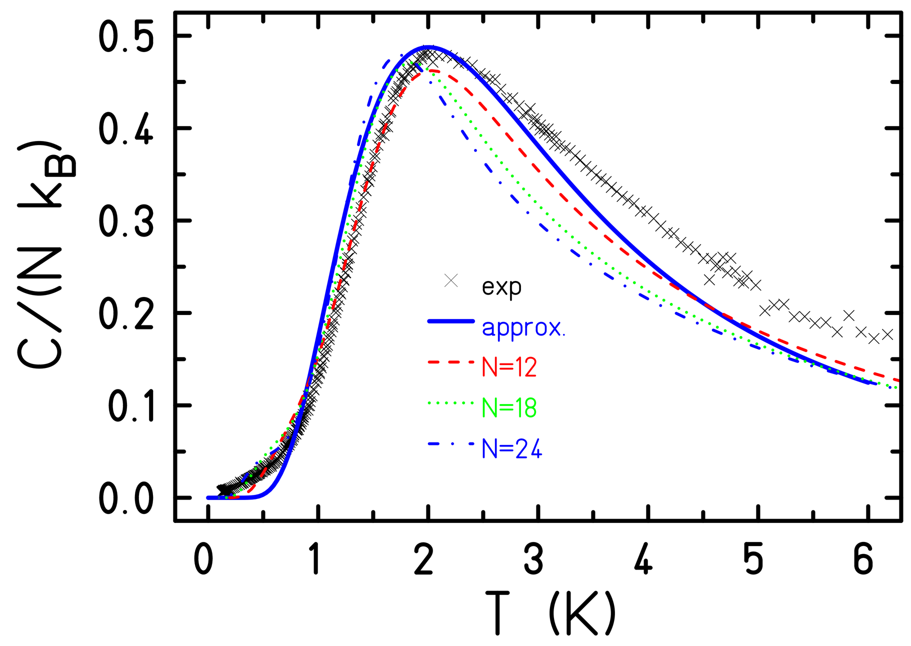

Figure 5: Specific heat of $[(CuCl_2tachH)_3Cl]Cl_2$. The symbols denote the experimental values, whereas the solid curve depicts the two-level approximation. The broken curves denote the specific heat for three complete diagonalizations for finite sizes.

Summary of the results

We demonstrated that at low enough temperatures the specific-heat behavior of $[(CuCl_2tachH)_3Cl]Cl_2$ points towards a Tomonaga-Luttinger liquid ground state in this spin-tube material, which corresponds to an effective spin-3/2 AF Heisenberg chain with the exchange constant $J_{\rm eff} = 2J_2/3$. On the other hand, a detailed analysis of the specific-heat data–combining the semiclassical approach with a number of numerical techniques– implies that the main contribution to the specific-heat peak located around $T \approx 2$ K comes from the low-lying gapped magnon excitations resulting from the internal degrees of freedom of the composite rung spins.

Acknowledgments

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