Heat capacity of $Cu_2(C_5H_{12}N_2)_2Cl_4$ in a magnetic field

Masayuki Hagiwara [a,1], Hiroko Aruga Katori [a], Uli Schollwöck [b], Hans-Jürgen Mikeska [c]

[a] *The Physical and Chemical Research (RIKEN), Wako, Saitama 351-0198, Japan*

[b] *Sektion Physik, Ludwig-Maximilians-Universität München, Theresienstr. 37, 80333 München, Germany*

[c] *Institut für Theoretische Physik, Universität Hannover, Appelstr. 2, D- 30167 Hannover, Germany*

Abstract

Heat capacity measurements down to 0.5 K in a magnetic field up to 12 T have been performed on a single crystal sample of a spin-ladder like compound $Cu_2(C_5H_{12}N_2)_2Cl_4$. The singlet ground state with an energy gap becomes magnetic and gapless between 7 T (H_{c1}) and 13 T (H_{c2}). In the temperature dependence of the heat capacity of this compound, we have observed a sharp and a round peak in the gapless region between H_{c1} and H_{c2}. The former peak most likely arises from the long range order and the latter one is probably attributable to the low energy excitation structure in a gapless spin liquid system.

Keywords: heat capacity; spin-ladder; $Cu_2(C_5H_{12}N_2)_2Cl_4$; spin liquid

The singlet ground state with an energy gap in a one-dimensional (1D) antiferromagnet becomes magnetic and gapless under a certain applied magnetic field. At this critical field (H_{c1}), the features of the system change dramatically. There are several gapped examples to study the low temperature properties at this critical point and in the gapless region. One of the adequate examples is $Cu_2(C_5H_{12}N_2)_2Cl_4$ (abbreviated as CHpC), because the critical field ($H_{c1} \sim 7$ T) and the saturation field ($H_{c2} \sim 13$ T) are easily accessible by a conventional magnet.

In this paper, we report the results of heat capacity measurements on a single crystal sample of CHpC and the comparison with the calculated heat capacity.

Single crystals of CHpC were obtained by the slow evaporation method from a methanol solution of powder samples of CHpC which were synthesized following the method reported in ref. [1]. This compound crystallizes in the monoclinic system (space group $P2_1/C$).[1] The Cu dimer formula units in CHpC stack to form double chains along the [101] direction. Correspondingly, the magnetic interactions between Cu spins are thought to yield a spin-ladder with a spin Hamiltonian written as

$$H = J_\perp \sum_{j=1}^{N/2} S_{1,j} \cdot S_{2,j} + J_\parallel \sum_{i=1}^{2} \sum_{j=1}^{N/2} S_{i,j} \cdot S_{i,j+1}, \quad (1)$$

where $J_\perp$ and $J_\parallel$ are the exchange constants along the rung and the leg, respectively and $S_{i,j}$ an$S$=1/2 spin on the i-th leg and the j-th rung. The ratio of $J_\perp$ to $J_\parallel$ was evaluated to be about five by the fitting of the susceptibility and mag-

[1] Corresponding author. Fax:+81-48-462-4649; E-mail: hagiwara@postman.riken.go.jp

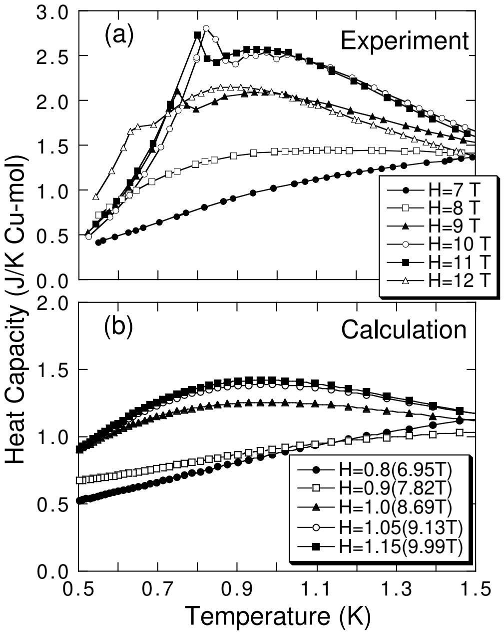

Fig. 1. (a). Temperature dependence of heat capacity of CHpC for designated magnetic fields. (b). Temperature dependence of heat capacity calculated with eq. (1) for $J_\perp/J_\parallel$=5.

netization data to the calculated ones.[2] Heat capacity measurements down to 0.5 K in a magnetic field (H) up to 12 T were performed using Mag Lab^{HC} (Oxford Instruments) installed in RIKEN.

Figure 1(a) shows the temperature (T) dependence of the heat capacity of a single crystal of CHpC under the designated magnetic fields above H_{c1} applied along the chain. The lattice contribution of the heat capacity is estimated from the heat capacity at H=0 T and is less than 1% of the total amount of the heat capacity below 1.5 K, so that the heat capacity shown in this figure mostly comes from the magnetic contribution. A sharp and a round peak have been observed. The former peak most likely arises from the long range order. The peak position shifts to high T with increasing the magnetic field until about 10 T, and then shifts to lower T with further increase in magnetic fields. The latter round peak is probably attributable to the low energy excitation structure in a gapless spin liquid system and the tendency of this peak to shift with change of fields is similar to that of the former peak.

In the following, we concentrate on the latter peak and compare the observed heat capacity with the calculated one (temperature dependent density-matrix-renormalization-group (DMRG) method). We show in Fig.1(b) the temperature dependence of the heat capacity calculated with eq.(1) for $J_\perp/J_\parallel$=5. Here, we use the exchange constant $J_\perp/\mathrm{k_B}$=12.3 K and the g-value g=2.1. These values are almost the same as those obtained in other experiments.[2] We clearly see that the calculated heat capacity has a round peak at nearly the same temperature as the observed one. The shift of the calculated peak with change of fields, although it is very weak, is similar to that of the observed peak. But the magnitude of the calculated peak is smaller than that of the observed one except at 7 T. No more numerical errors are expected in the theoretical curves for the 1D model as given in eq.(1). The difference may come from 3D interactions. A lattice instability associated with the critical field transition is suggested by Nagaosa and Murakami[3] and may be one of the candidates to explain this discrepancy. The round peak is attributable to a spinon band in the spin-liquid system discussed in ref. [4]. According to this theory[4], the splitting of the peak due to two different spinon bands is expected for magnetic fields off the symmetry field $\frac{1}{2}(H_{c1}+H_{c2})$. We, however, observed only one peak. On the contrary, the second higher temperature peak (not shown) is quantitatively quite well reproduced. A more elaborate theory is needed to clarify the quantitative discrepancy for the low-T peak.

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