Specific heat of an S=1 quasi-1D bond alternating antiferromagnet in a magnetic field

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Abstract

We report the results of specific heat measurements in a magnetic field on single crystal samples of the S=1 quasi-1D bond alternating antiferromagnet $Ni(C_9H_{24}N_4)NO_2(ClO_4)$. The compound has the singlet ground state with an excitation energy gap and the gap closes by application of magnetic field (Hc). We observed a sharp peak corresponding to the long-range ordering above Hc both parallel and perpendicular to the chain at low temperatures. The temperature-magnetic field phase diagram is discussed in comparison with those of the Haldane system $Ni(C_5H_{14}N_2)_2N_3(PF_6)$.

Key words: bond alternating antiferromagnet; long-range-ordering; temperature-magnetic field phase diagram; specific heat

1. Introduction

Recently, field-induced phenomena have attracted considerable interest. On the one hand, magnetization plateaux attributed to many body quantum effect have been studied extensively and, on the other hand, much attention has been paid to the field induced long-range-ordering in the gapped low-dimensional magnets[1,2].

In the present study, we report the results of specific heat measurements in a magnetic field on single crystal samples of $Ni(C_9H_{24}N_4)NO_3(ClO_4)$, abbreviated as NTENP, which is regarded as the S=1 quasi-1D bond alternating antiferromagnet. This compound has an excitation energy gap between the singlet ground state and the excited triplet one at zero field. When the magnetic field is applied, one of the triplet branches goes down and intersects the singlet state, and magnetism recovers at this critical field (Hc). Then, we expect the long-range-ordering to occur above Hc because the interchain interaction exists in a real material.

NTENP crystallizes in the triclinic system, space group $P\bar{1}$. The chain consists of Ni^{2+} ions bridging through nitrito groups and runs along the a-axis. The most important feature is that the inversion centers are situated not on the Ni^{2+} ions but on the nitrito groups and two different bond distances exist between Ni^{2+} ions [3]. As a result, this compound is expected to have bond alternation. Magnetic properties of this compound were investigated by magnetic susceptibility and magnetization measurements. By comparing the experimental susceptibility data with the calculated ones, the bond alternating ratio between the neighboring exchange constants is estimated to be 0.45, which means that the ground state of this compound is predicted to be in the singlet-dimer phase [4]. This prediction was confirmed by experiment on single crystal samples of NTENP doped with nonmagnetic impurities [5].

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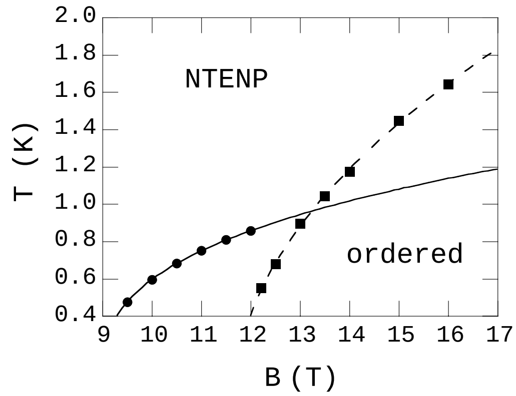

Fig. 1. Temperature versus magnetic field phase diagram of NTENP obtained from the specific heat measurements. Filled circles and squares are peak positions of the specific heat for H//chain and H⊥chain, respectively. Solid and broken lines are the results of the fittings.

2. Experimental

Single crystals of NTENP were grown by the slow evaporation method reported in Ref. 3. The characterization of the present sample is described in Ref. 5. The specific heat measurements under magnetic fields were carried out with a Mag Lab^{HC} microcalorimeter (Oxford Instruments, UK) installed at RIKEN for H//chain up to 12 T and a home made specific heat measuring system installed at KYOKUGEN, Osaka University for $H\perp$chain up to 16 T.

3. Results and discussion

We observed a sharp peak in the temperature dependence of the specific heat in the external field above 9.5 T for H//chain and above 12.2 T for $H\perp$chain. The position of the peak shifts to higher temperature with increasing magnetic field. Since the high-field magnetization process indicated that this region is in the gapless state, the peak of the specific heat corresponds obviously to the phase transition to the long-range-ordered state. The field dependence of the peak is plotted in Fig. 1. The temperature vs. magnetic field phase diagram is similar to those of the S=1 quasi-1D Heisenberg antiferromagnets $Ni(C_5H_{14}N_2)_2N_3(PF_6)$, (NDMAP) [1] and $Ni(C_5H_{14}N_2)_2N_3(ClO_4)$, (NDMAZ) [2] where the anisotropic behavior for the field direction is explained by the existence of the single ion anisotropy D. [1] NTENP has the single ion anisotropy constant D/k$_B$ = 13.6 K of easy plane type and the principal axis of single ion anisotropy is close to the chain axis. These properties are very similar to those of NDMAP [1] and NDMAZ [2].

The peak position for $H\perp$chain shifts largely with increasing magnetic field, while that for H//chain does not move so much especially at high fields. Solid and broken lines in the figure show the results of the fitting of the peak to the expression of T(K)=a| H(T)-Hc|$^\alpha$ for H//chain and $H\perp$chain, respectively. The parameters obtained from the fitting are a=0.78±0.05, Hc=11.7±0.1 and α=0.52±0.04 for the former, and a=0.59±0.01, Hc=8.98±0.02 and α=0.334±0.004 for the latter. Similar results were obtained in NDMAP [1]. The reported exponents (α=0.326 for H//chain and α=0.513 for $H\perp$chain) are very close to those of NTENP. As mentioned above, the ground state of NTENP is in the singlet-dimer phase, but that of NDMAP is in the Haldane phase. On applying magnetic field and quenching the energy gap, these two systems enter the same gapless phase as shown theoretically in the magnetic field vs. bond alternating ratio phase diagram[6]. Then, the long-range-ordering takes place in the same gapless phase with the help of interchain coupling. Therefore, it might be concluded that the exponents are very close to each other in NTENP and NDMAP.

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