Field-Induced Magnetic Ordering in an Alternating Heisenberg Chain F_5PNN

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Abstract

We have found the field-induced magnetic ordering of an $S = 1/2$ alternating linear Heisenberg antiferromagnet F_5PNN by measuring magnetic field dependence of the specific heat. A cusp-like anomaly is observed below 0.2 K between 3.5 T and 6 T. From the extrapolation of H vs T phase diagram to 0 K, the lower and the upper critical fields are expected to be about 3 T and 7 T, respectively. The phase boundary is almost symmetric with respect to a horizontal line of $H = 5$ T. This behavior is consistent with the previously observed magnetization measurement in which magnetization began to increase at about 3 T and saturated at about 7 T.

Key words: alternating chain, spin-gap system, specific heat, field-induced magnetic ordering

1. Introduction

Low-dimentional quantum spin-gap systems such as Haldane systems, spin-peierls systems and two-leg ladder systems have been attracting much attention since the Haldane conjecture [1]. One of the notable quantum effects in spin-gap systems is the existence of the excitation gap Δ between the non-magnetic singlet ground state and the lowest excited state. When the magnetic field higher than the critical field $H_g = \Delta/g\mu_B$ is applied, some spin-gap systems become gapless and the three-dimensional long-range magnetic ordering (3DLRMO) occurs in these systems. Recently, it has been proposed that such transitions induced by magnetic fields can be interpreted as a Bose-Einstein condensation of magnons [2]. To clarify the nature of the field-induced magnetic ordering (FIMO), we have studied the field dependence of the specific heat of an $S = 1/2$ alternating linear Heisenberg genuine organic antiferromagnet, pentafluorophenyl nitroxide radical (F_5PNN). The alternating linear Heisenberg antiferromagnet has two antiferromagnetic interactions lying alternately in one dimension, and show the excitation gap at very low temperatures. This system is described by the following Hamiltonian:

$$H = -2\sum_{i}^{N/2}(J_1 S_{2i-1}\cdot S_{2i} + J_2 S_{2i}\cdot S_{2i+1}) \qquad (1)$$

where J_1 and J_2 are constants of inter-spin exchange interactions along the chain, S denotes the $S = 1/2$ Heisenberg type of spin operator and N is the total number of spins. Low-temperature susceptibility of F_5PNN is well reproduced by this Hamiltonian with exchange parameters, $\alpha = |J_2/J_1| \simeq 0.4$ and $2J_1/k_B = -5.6$ K. The magnetization measurement at 0.5 K indicates that the gap of F_5PNN is about 3 T [3], which is quite a small gap compared to those of other compounds studied in previous works. Moreover, magnetic

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anisotropy is extremely small because of complete quenching of angular momentum via molecular orbitals. We believe that these features of F_5PNN enable us to understand a nature of FIMO in detail.

2. Experimental

The single crystals of F_5PNN were prepared by the procedure reported in Ref.[4]. Specific heats were measured by the adiabatic heat-pulse method in several magnetic fields. Two pieces of the crystal were covered by Apiezon-J grease to ensure the thermal contact, and were attached on the Ag background cell (AgBG) in a sample holder of a ^{3}He-^{4}He dilution refrigerator. Magnetic fields were applied by a superconducting magnet. The specific heats (C) of F_5PNN was estimated by subtracting the nuclear magnetic specific heat of AgBG from the total specific heat.

3. Results and discussion

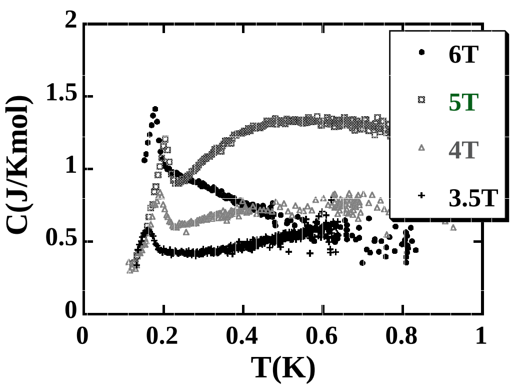

Fig. 1. Field dependence of the specific heat of F_5PNN. A cusp-like anomaly is observed for $T \leq 0.2$ K, in 3.5 T$\leq H \leq 6$ T. A broad maximum is observed above the cusp-like anomaly between 3.5 T and 5 T. But at 6 T this broad maximum disappear.

Fig.1 shows the temperature dependence of C in magnetic fields 3.5 T$\leq H \leq 6$ T. A cusp-like anomaly is observed for $T \leq 0.2$ K. On the other hand, this anomaly is not observed below 3 T. From the extrapolation of H vs T phase diagram to 0 K (Fig.2) in which the critical temperature of the anomaly is plotted, the lower and the upper critical fields are expected to be about 3 T and 7 T, respectively. The phase boundary is almost symmetric with respect to a horizontal line of $H = 5$ T. The lower and upper critical fields estimated from this phase diagram are consistent with the low-temperature magnetization measurement, in which magnetization began to increase at about 3T and saturated at about 7 T [3]. This indicates that the anomaly observed in C is caused by the occurrence of 3DLRMO triggered by small interchain interactions.

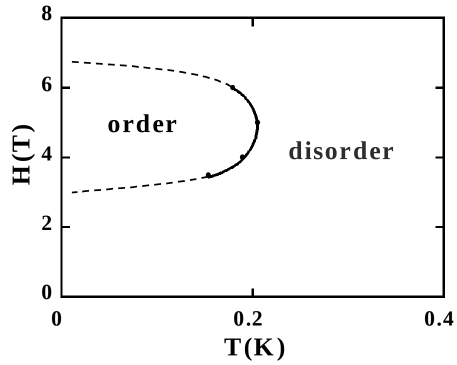

Fig. 2. H vs T diagram of the cusp-like anomaly. The lower and the upper critical fields are expected to be about 3 T and 7 T, respectively

At the temperatures above the anomaly, a broad maximum is observed below 5 T. While at 6 T, this broad maximum seems to disappear. The origin and the behaviour of the broad maximum have great interest. One possible explanation for the origin is that a broad maximum due to the short-range order in 1D system observed at 0 T is suppressed and sifted to the lower temperatures by the magnetic field. To clarify the origin and the behaviour of the broad maximum, detailed experimental and theoretical studies are needed.

4. Conclusion

We have studied the field dependence of the specific heat of the $S = 1/2$ alternating linear Heisenberg genuine organic antiferromagnet F_5PNN. A cusp-like anomaly is observed for $T \leq 0.2$ K, in 3.5 T$\leq H \leq 6$ T. From the H vs T phase diagram in which the critical temperature of the anomaly is plotted, the lower and the upper critical fields are expected to be about 3 T and 7 T, respectively. The phase boundary is almost symmetric with respect to a horizontal line of $H = 5$ T. Because the phase diagram is consistent with the magnetization measurement, we have found the origin of the anomaly is the occurrence of FIMO in F_5PNN.

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