Anomalous two-stage spin-flop transition in $BaCu_2Si_2O_7$

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Abstract

Anomalous two-stage spin-flop transition in $BaCu_2Si_2O_7$ was investigated in detail. This compound shows antiferromagnetic long-range order below $T_N = 9.2$ K with its c axis as an spin easy axis. Magnetic field applied to the c direction induces spin-flop transitions twice; spins are first flopped roughly to the b axis and next to the a axis. This peculiar behavior is explained neither by the conventional spin-flop mechanism nor by the competition between inter-chain Heisenberg interactions and intra-chain Dzyaloshinskii-Moriya interactions as was previously proposed.

Key words: quasi one dimensional; antiferromagnet; spin flop

1. Introduction

Recent extensive studies have revealed various new phenomena in one-dimensional (1D) quantum spin systems. In a real compound, however, not only intra-chain interactions but also inter-chain interacions play a significant role on its magnetic behavior, especially at low temperatures. $BaCu_2Si_2O_7$ is a good example of such particular interest, and has been investigated in detail [1–3]. One of the remarkable findings is two-stage spin-flop transition observed in $BaCu_2Si_2O_7$ under $H \parallel c$ configuration [4]. We discuss this strange magnetic behavior based on the recently determined spin structure [5].

2. Experimental results and discussion

Single crystal samples were prepared by a conventional floating-zone method. For magnetization and heat capacity measurement, one of the crystals was cut into a rectangular shape with each face normal to one of the principal axes. For neutron diffraction measurement, one or two as-grown single crystal rods were used without cutting.

$BaCu_2Si_2O_7$ shows antiferromagnetic transition at $T_N = 9.2$ K [1]. Figure 1(a) shows field dependence of the magnetization measured along the c axis at $T = T_N$ and $T < T_N$. It is clearly seen that a spin-flop like magnetization jump occurs twice, which is absent in the b-axis magnetization data. The neutron diffraction measurement under several magnetic fields has revealed the spin structure as shown also in Fig. 1 (a) [5]. At the first transition ($H_{c1} = 2.0$ T), spins flop roughly to the b-axis direction with a finite canting angle toward the a axis. In the intermediate field range (2.0 T $< H <$ 4.9 T), spins gradually rotate toward the b-axis direction, and finally become almost parallel to the b-axis at the second transition ($H_{c2} = 4.9$ T). Above H_{c2} spins flop to the a-axis direction. This spin-flop behavior is inconsistent with the previously proposed model [4], where the competition between inter-chain Heisenberg interactions and intra-chain Dzyaloshinskii-Moriya (DM) interactions flip spins first to the a axis and next to the b axis.

To see the difference of the two spin-flop phases, we have carried out heat capacity measurement under three magnetic fields as shown in Fig. 1(b). A steep jump of the specific heat is consistent with the anti-

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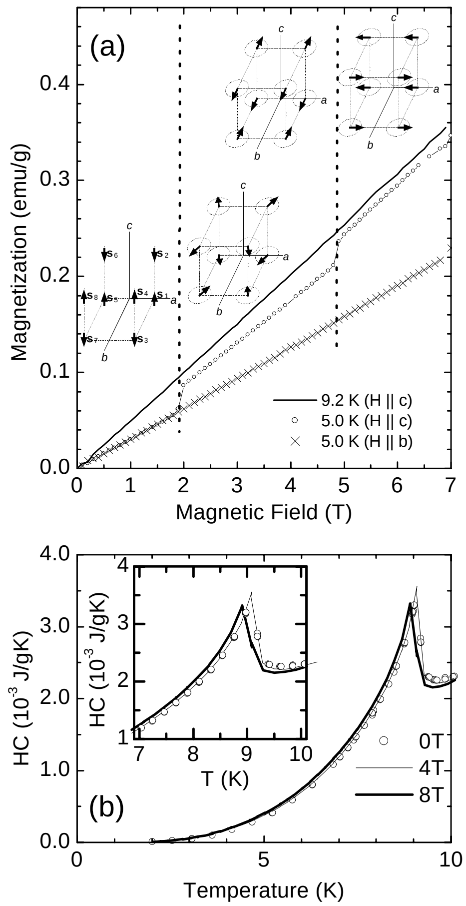

Fig. 1. (a) Field dependence of magnetization along the c and b axes. The spin structure determined by the neutron diffraction is also shown. (b) Temperature dependence of heat capacity under three different fields.

ferromagnetic transition at $T_N = 9.2$ K. Importance is that temperature dependence of the heat capacity taken at $H = 4$ T is almost the same as that at $H = 0$ T, while the data at $H = 8$ T is clearly different from the lower-field ones. This result suggests that the low-energy magnetic excitation spectrum is almost the same between the low- and the intermediate-field phases, while that of the high-field phase is qualitatively different from the lower-field phases. Such discontinuous behavior across H_{c2} is observed also in the thermal conductivity [6], which is strongly related to the low-lying magnetic excitations. Since thermal conductivity is known to show discontinuous behavior through a spin-flop transion [7], the spin-flop transition at H_{c2} is considered to be the same as the conventional one, and therefore, the transition at H_{c1} is probably new one.

Our next issue is to explain why this first transition occurs in $BaCu_2Si_2O_7$. We must first explain the small canting of spins toward the a-axis direction at the field slightly above H_{c1}, and for this purpose, all the possible inter-chain interactions should be evaluated more rigorously.

3. Summary

The exotic two-stage spin-flop transition has not yet been fully understood. However, such a quasi one-dimensional quantum spin system as $BaCu_2Si_2O_7$ is very susceptible to various external perturbation, and our finding is one of the good exhibitions to demonstrate rich physics of low-dimensional quantum magnets.

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