Magnetic property of $Ba_2CoGe_2O_7$

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Abstract

A two-dimensional antiferromagnet ($S = 3/2$), $Ba_2CoGe_2O_7$, was studied, which is isostructural to $S = 1/2$ spiral antiferromagnet, $Ba_2CuGe_2O_7$. A single crystal of $Ba_2CoGe_2O_7$ was grown and its magnetization was measured. A weak ferromagnetism was observed at 6.7 K, while the spiral antiferromagnetism was reported in $Ba_2CuGe_2O_7$. The origin of the ordered states in these isostructural materials are Dzyaloshinskii-Moriya interaction and the change of the direction of $\boldsymbol{D}$ vector results in the different spin configurations. We have also synthesized polycrystalline samples of $Ba_2Cu_{1-x}Co_xGe_2O_7$, and observed the change of the ordered state with x.

Key words: 2D spiral antiferromagnetism; magnetization measurement; weak ferromagnetism

1. Introduction

$Ba_2CoGe_2O_7$ is a two-dimensional spin system and it is isostructural to $Ba_2CuGe_2O_7$ which is a spiral antiferromagnet due to Dzyaloshinskii-Moriya (DM) interaction [1]. The in-plane structure and the components of the Dzyaloshinskii vector $\boldsymbol{D}$ is shown in Fig. 1 of ref. 3. The allowed components of $\boldsymbol{D}$ are (1, -1, 0) and (0, 0, 1). The sign of (1, -1, 0) component is the same and that of (0, 0, 1) alternates [2]. The spiral antiferromagnets, therefore, is caused by the (1, -1, 0) component [3].

The ground state of $Ba_2CuGe_2O_7$ is not a simple spiral state but a solitary state even when $\boldsymbol{H}$=0. This spiral-soliton state is realized by considering the Kaplan−Shekhtman−Entin-Wohlman−Aharony (KSEA) interaction [4–6]. $Ba_2CuGe_2O_7$ is the first experimental evidence for the KSEA interaction. In the substituted system, $Ba_2CoGe_2O_7$ ($S = 3/2$ spins on Co^{2+} ion), a single-ion anisotropy term $DS_z{}^2$ and the change of the direction of Dzyaloshinskii vector $\boldsymbol{D}$ will be expected. Hence, we study the magnetism of $Ba_2CoGe_2O_7$ by means of the magnetization measurement and weak ferromagnetism was observed. We also measured the magnetization of $Ba_2Cu_{1-x}Co_xGe_2O_7$ and observed the change of the ordered state with x.

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2. Experiments

We obtained single crystals of $Ba_2CuGe_2O_7$ and $Ba_2CoGe_2O_7$ by the floating zone method. Mixed polycrystalline samples of $Ba_2Cu_{1-x}Co_xGe_2O_7$ (x=1.00, 0.90, 0.70, 0.50, 0.30, 0.10, 0.05, 0.03, and 0.00) were synthesized by the solid reaction method. The magnetization was measured by a commercial dc-SQUID magnetometer (Quantum Design MPMS-XL7).

3. Results and Discussion

Figure 1 shows the magnetization of $Ba_2CoGe_2O_7$ single crystal. The sharp increase of magnetization was observed at 6.7 K. Below T_c, magnetization remained constant in a field cooling (FC) process. We also measured the magnetization in a zero field cooling (ZFC) process and the hysteresis was observed between the FC and the ZFC processes. The results suggest that a weak ferromagnetism which is caused by Dzaloshinskii-Moriya interaction exists in this material. The anisotropy of magnetization was also observed between magnetic fields parallel to 2D plane ($\boldsymbol{H} \perp c$) and perpendicular to the plane ($\boldsymbol{H} \parallel c$). The magnetization of $\boldsymbol{H} \perp c$ is twice as much as that of $\boldsymbol{H} \parallel c$ at low temperatures, while the anisotropy is not so large at high temperatures. So, at low temperatures, the in-plane component of the spins becomes larger. Therefore it is concluded that $Ba_2CoGe_2O_7$ has an easy-plane type anisotropy. The origin of this anisotropy is the single-ion anisotropy $DS_z{}^2$ or the anisotropy in exchange energy J.

Figure 2 shows the temperature dependence of magnetization of $Ba_2Cu_{1-x}Co_xGe_2O_7$ in the FC process. The weak ferromagnetism was observed at $x \geq 0.1$. A small anomaly is observed near 3 K in the samples of $Ba_2Cu_{1-x}Co_xGe_2O_7$ ($x \leq 0.05$) which is assigned to the spiral state. Therefore at lower Co concentration, the ground state is the spiral antiferromagnetic state. The critical concentration for the disappearance of the spiral phase is $x \sim 0.05$.

What is the origin of the difference in the spin configurations between Co-rich and Cu-rich materials? The ordered state is expected to be a spiral antiferromagnetic state when $\boldsymbol{D}$ is parallel with each other, and the one is a weak ferromagnetic state when $\boldsymbol{D}$ is antiparallel. In $Ba_2Cu_{1-x}Co_xGe_2O_7$ system the allowed components of $\boldsymbol{D}$ are (1, -1, 0) and (0, 0, 1), which are the same as those in $Ba_2CuGe_2O_7$. The sign of (1, -1, 0) component is the same and that of (0, 0, 1) alternates according to the crystal symmetry. In Cu-rich materials (1, -1, 0) component is dominant and the spiral phase appears. On the other hand, in Co-rich materials (0, 0, 1) component would be dominant and weak ferromagnetism appears.

In summary we measured the magnetization of $Ba_2Cu_{1-x}Co_xGe_2O_7$. We found the weak ferromagnetism at $x \geq 0.1$. This is because the direction of Dzyaloshinskii vector $\boldsymbol{D}$ is changed by the cobalt substitution. Especially, $Ba_2CoGe_2O_7$ has a large in-plane type anisotropy and exhibits weak ferromagnetism at 6.7 K.

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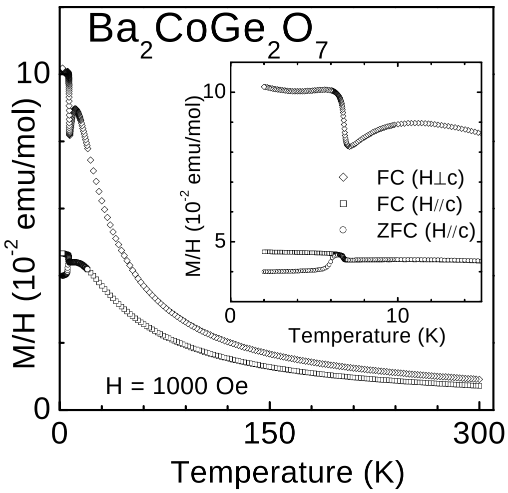

Fig. 1. Temperature dependence of the magnetization in the single crystal $Ba_2CoGe_2O_7$. A large anisotropy is observed.

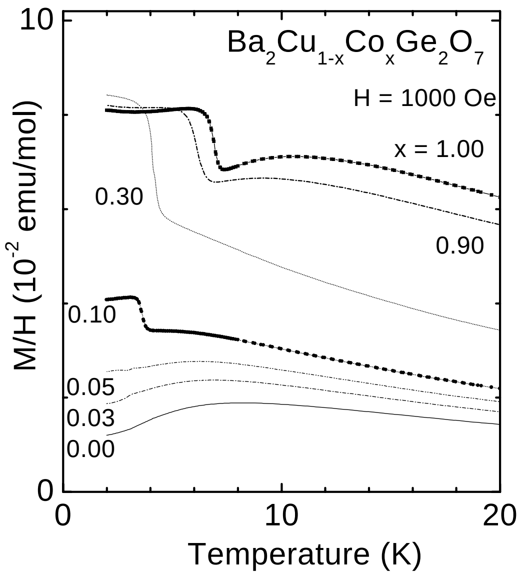

Fig. 2. Temperature dependence of the magnetization in the polycrystalline samples of $Ba_2Cu_{1-x}Co_xGe_2O_7$. These data were obtained in the field cooling process.

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