Magnetization of Highly Frustrated Zn-Chromium Spinel Oxides up to a Megagauss Magnetic Field

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Abstract. Precise magnetization processes of ZnCr$_2$O$_4$ were obtained by means of the Faraday rotation method subjected to ultra high magnetic fields up to 190 T generated by a single turn coil technique. In magnetization processes of ZnCr$_2$O$_4$, there observed a coplanar 2:1:1 canted state (the cant 2:1:1 phase) which is predicted by the theoretical model including a spin-lattice coupling. A magnetic-field width of the 1/2 plateau widens as the temperature increases.

1. Introduction

Geometrical frustration has been an intriguing issue in condensed matter physics for a few decades. Geometrically frustrated magnet has a macroscopically degenerated ground state such as a spin liquid state or a spin ice state in contrast to the case of a long-range Néel order. However, the macroscopic degeneracy is easily lifted by small perturbations induced by a spin-lattice coupling, far-neighbor interactions or quantum or thermal fluctuations. Therefore, they cause peculiar magnetic phases such as the plateau phase which is frequently observed in a frustrated magnet. Moreover, what is peculiar is that the quantum or thermal fluctuations select the characteristic spin configuration (the order phase) among the macroscopic spin configurations, which is the so-called “order from disorder” phenomenon [1,2,3].

ZnCr$_2$O$_4$ belongs to chromium spinel oxides ACr$_2$O$_4$ in which Cr$^{3+}$ ions (S=3/2) form a pyrochlore lattice. Therefore, ACr$_2$O$_4$ is a good candidate as the geometrically frustrated magnet in the three dimensional system. In this system, Néel temperature (-32 K in HgCr$_2$O$_4$, -70 K in CdCr$_2$O$_4$ and -390 K in ZnCr$_2$O$_4$) is far different from Curie-Weiss temperature (5.8 K in HgCr$_2$O$_4$, 7.8 K in CdCr$_2$O$_4$ and 12.5 K in ZnCr$_2$O$_4$) determined by susceptibility measurements [4,5]. This indicates that ZnCr$_2$O$_4$ bears the strongest spin frustration among the chromium spinel oxides. Therefore, magnetic properties of ZnCr$_2$O$_4$ in zero magnetic field were intensively studied as a prototype of a three dimensional frustrated magnet [6]. However, the magnetic properties of ZnCr$_2$O$_4$ have been less investigated under a magnetic field since the large antiferromagnetic exchange energy requires an extremely high magnetic field over 100 T. On the other hand, those of HgCr$_2$O$_4$ and CdCr$_2$O$_4$ in magnetic fields were well investigated and the field-induced phase transitions and the 1/2 magnetization plateau were
observed [4,5]. Penc et al. presented a theory of the magnetization process of a pyrochlore spin system taking account of a spin-lattice coupling [7]. In this model, an effective Hamiltonian is described by the bilinear and biquadratic terms in which the spin-lattice interaction term is incorporated. The latter term stabilizes the 3-up 1-down collinear spin configuration (the 1/2 plateau phase). Moreover, it is suggested in this model that there appears the canted 2:1:1 coplanar spin configuration (the Cant 2:1:1 phase) in a limit of the weak spin-lattice interaction. The strength of a coefficient “b” of a biquadratic term plays a key role on determination of the magnetic phases. The magnetization at finite temperatures was studied by Monte-Carlo calculations by Motome et al. and showed a good agreement with those of experiments for HgCr$_2$O$_4$ and CdCr$_2$O$_4$[8].

In this paper, we report the results of the magnetization of ZnCr$_2$O$_4$ measured at various temperatures by means of the Faraday rotation method up to 190 T generated by a single-turn coil technique and compared them with those of the theoretical simulation.

2. Experimental methods

The single-turn coil method was employed to generate a mega-gauss magnetic field up to 190 T. The magnetization measurements by using an induction method under a mega-gauss field are quite difficult because of a huge background induction voltage. Therefore, we applied the Faraday rotation method to the magnetization measurements under a mega-gauss field. The magnetization deduced from the Faraday rotation angle measured up to 140 T in CdCr$_2$O$_4$ was reported in our previous report [9]. The experimental set-up was similar to that in Ref. [9].

The magnetic field was measured by means of a calibrated pick-up coil wound near the sample. The estimated error of the magnetic field was about ±3%. A semiconductor laser (a Coherent “Cube”) having a central wavelength of 635 nm was used as a light source. The single crystal of ZnCr$_2$O$_4$ was grown by the flux method. The sample was cut parallel to the (111) crystal surface, attached on a quartz substrate and polished into about 100 µm thickness. The samples were cooled using a handmade liquid-He flow type cryostat made totally of a “stycast” resin.

3. Results and discussion

The red line in Fig. 1 shows the Faraday rotation angle of ZnCr$_2$O$_4$ at 9 K up to 190 T. A negative linear component arising from diamagnetic contribution of both the sample and the quartz substrate were subtracted from the original signal. The black dotted line in Fig. 1 shows the magnetization of ZnCr$_2$O$_4$ measured at 4.2 K by an induction method using a long pulse magnet [5]. It is evident from this plot that the Faraday rotation angle is proportional to the magnetization up to 50 T.

Figure 1. Red line: The magnetization process of ZnCr$_2$O$_4$ at 9 K up to 190 T by Faraday rotation method. Dotted line: The magnetization process of ZnCr$_2$O$_4$ at 4.2 K up to 50 T measured by a long pulse magnet. Symbols (circle, triangle, and square) show points of the magnetic phase transition.
From Fig. 1, we found that there observed clearly a first-order phase transition (a circular symbol at 120 T in Fig. 1) and two second order phase transition (a triangular symbol at 135 T and a square symbol at 158 T in Fig. 1), and that a magnetization was constant with a half of full magnetization moment (the 1/2 plateau phase) between the triangular and the square symbols in Fig. 1. The phase between the circular and the triangular symbols (phase I) was observed only in ZnCr$_2$O$_4$ but not in HgCr$_2$O$_4$ or CdCr$_2$O$_4$ [5,9]. We compared this magnetization process with the model calculation by Penc et al. and obtained a fairly good agreement with theory when a coefficient “b” of the biquadratic term defined by Penc et al. is very small [7]. From this comparison, the phase I observed in ZnCr$_2$O$_4$ is regarded as the Cant 2:1:1 phase (the canted 2:1:1 coplanar spin configuration). In Ref. [10], the coefficient “b” of the biquadratic term was studied by examining the magnetic properties under pressure and it was concluded that the “b” of ZnCr$_2$O$_4$ was much smaller than those of HgCr$_2$O$_4$ or CdCr$_2$O$_4$. Their conclusion is consistent with ours, since the Cant 2:1:1 phase could only be observable when b is small according to the calculation by Penc et al.

We have measured the magnetization processes of ZnCr$_2$O$_4$ at various temperatures, and found a strong temperature dependence as shown in Fig. 2 (a). We conducted a finite temperature Monte-Carlo simulation, and calculated results are presented in Fig. 2 (b) for comparison. The calculation was carried out similarly to the model in Ref. [8], and the coefficient “b” of the biquadratic term was chosen as 0.02. Symbols show the positions of magnetic phase transitions as in Fig. 1. Circular and triangular symbols shift toward a low magnetic field side and the square symbol hardly shifts as the temperature increases until the Cant 2:1:1 phase disappears.

Fig. 3 shows a magnetic phase diagram determined from the magnetization processes in Fig. 1 and 2 (a). The width of the 1/2 plateau widens and the Cant 2:1:1 phase narrows as temperature increases. This indicates that thermal fluctuations stabilize the collinear spin configuration (the 1/2 plateau phase) rather than non-collinear spin configuration (the Cant 2:1:1 phase) [11]. In Ref. [5,9], magnetic phase diagrams of HgCr$_2$O$_4$ and CdCr$_2$O$_4$ were determined, where the width of the 1/2 plateau phase exhibited less temperature dependence. ZnCr$_2$O$_4$ showed more prominent stabilization of the collinear spin configuration by thermal fluctuation than HgCr$_2$O$_4$ or CdCr$_2$O$_4$. It is expected that this effect becomes more evident as a coefficient “b” of a biquadratic term goes smaller. Our results suggest that the thermal fluctuation favors the collinear spin configuration among the macroscopically degenerated ground states at b ≈ 0. This is so called the “order from disorder” phenomenon [11,12]. Monte-Carlo calculations suggest the similar scenario, and the issue about the “order from disorder” phenomenon at b = 0 will be discussed elsewhere [13].
4. Summary
We investigated the magnetization processes of ZnCr$_2$O$_4$ at various temperatures up to 190 T by the Faraday rotation method. We observed the Cant 2:1:1 phase which was predicted by the theoretical model including the spin-lattice coupling and this phase has not been observed in HgCr$_2$O$_4$ or CdCr$_2$O$_4$. This arises from the fact that the effective spin-lattice interaction in ZnCr$_2$O$_4$ is much smaller than that in the others. The magnetic phase diagram is determined for ZnCr$_2$O$_4$. We found that the 1/2 plateau phase of ZnCr$_2$O$_4$ was stabilized by thermal fluctuations, which is more evident than that of HgCr$_2$O$_4$ or CdCr$_2$O$_4$.

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Figure 3. A magnetic phase diagram of ZnCr$_2$O$_4$ determined from Fig. 2 (a).