Faraday rotation of spinel oxide under ultrahigh magnetic field

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Abstract. We studied the magnetic properties of ZnCr₂O₄ under an ultra-high magnetic field by using the Faraday rotation method. We used a single turn coil system to generate a pulsed high field up to 180 T and used a He-Ne laser as a light source. We chose a 45 degree angle between the polarizer and the analyzer and measured a transient Faraday rotation signal by using a photo-diode and a transient recorder. The sample temperature was kept below Neel temperature during the experiment. We observed a discernible discontinuity in a Faraday rotation signal at 120 T. This discontinuity is considered to represent the first-order phase transition which is observed also in CdCr₂O₄ which has the same geometrical structure as ZnCr₂O₄.

1. Introduction

A geometrically frustrated antiferromagnet is an antiferromagnet which cannot satisfy the antiferromagnetic interaction for all spin pairs. In such a system, the ordering is suppressed even at low temperature and the ground state is highly degenerated. A geometrical frustration appears in some lattice systems including triangular, kagome, and pyrochlore lattices which is composed of corner sharing tetrahedra. In a frustrated antiferromagnet, the degeneracy of the ground state is sometimes lifted by quantum fluctuations, thermal fluctuations, lattice distortions, or external magnetic fields. A frustrated antiferromagnet is a very interesting system which is expected to show various phases which are dependent on magnetic fields and temperature.

In chromium spinel oxides MCr₂O₄ (M=nonmagnetic ion), Cr ions form a pyrochlore lattice. A crystal field of nearly cubic symmetry splits the 3d orbitals of Cr³⁺ into t₂g and e_g states. By Hund’s rule, the t₂g state is half-filled and the orbital degree of freedom disappears. Therefore, this material shows a very small spin anisotropy and can be well described by a Heisenberg spin system. This system shows a paramagnetism at high temperature and its magnetic susceptibility can be explained by the Curie-Weiss law. The Curie-Weiss temperatures Θ_CW are estimated to be -70 K in CdCr₂O₄ and -390 K in ZnCr₂O₄ [1]. This system shows Neel ordering at low temperatures; the Neel temperatures T_N are 8 K in CdCr₂O₄ [2] and 12 K in ZnCr₂O₄ [3]. Such a large difference between the Curie-Weiss temperature...
and the Neel temperature shows that a strong geometrical frustration prevents its ordering. In this system, lattice distortions also occur at $T_N$. Therefore, spin-lattice coupling also affects the phase transition to the ordered phase.

Ueda et al. measured the magnetization process of CdCr$_2$O$_4$ under high magnetic fields of up to 47 T by using a non-destructive pulsed magnet [1]. They found a first-order phase transition with a lattice distortion at 28 T and a wide magnetization plateau with one-half of the full moment $S=3/2$ following the phase transition. They considered that the phase transition corresponds to a transition to a 3-up 1-down collinear spin state for each Cr tetrahedron which reflects the three-dimensional geometrical frustration in this system.

Some theoretical models of the high field magnetization process in Cr spinel oxides have been suggested. Penc et al. considered that the phenomenon was classical, and that lattice distortions stabilized the collinear 3-up 1-down state through the spin-lattice coupling effect and produced a magnetization plateau [4]. On the other hand, Bergman et al. considered that a quantum fluctuation was the dominant factor of the stabilization of the plateau state [5].

We studied the magnetic properties of highly frustrated antiferromagnet ZnCr$_2$O$_4$ under a high magnetic field. To study the magnetization process in this material, a high magnetic field of over 100 T is necessary because the exchange interaction $J$ is very large. However, a magnetization measurement technique under such a high field has not yet been established. Therefore, we adopted the Faraday rotation method for the magnetization probe which is easily applicable to the measurement under ultra-high magnetic fields.

2. Experimental procedure
A destructive pulsed magnet is necessary for the generation of a magnetic field of over 100 T. We adopted a single-turn coil, which is a destructive pulsed magnet, to generate the field. We used a horizontal single-turn coil system which generates a field parallel to the ground surface. The magnetic field was measured by a pick-up coil and recorded using a transient recorder. The single crystal of ZnCr$_2$O$_4$ is grown by a flux method. The field was applied to the [111] direction of the sample. We used a He-Ne laser (632.8 nm, 1.96 eV) as a light source. We chose a 45 degree angle between the polarizer and the analyzer. The analyzer split the light transmitted through the sample into two orthogonal polarization components (a vertical and a horizontal component). Each polarization component was independently measured by a photo-detector and recorded using a transient recorder system. The Faraday rotation angle’s dependence on the field was determined by a comparison between the difference of two polarization components and the magnetic field. The sample was cooled below $T_N$ in a cryostat made of phenol resin. The capacitor bank for the horizontal single turn coil method in ISSP is a fast capacitor bank system which can store energy of up to 200 kJ (50 kV, 160 μF). The inner diameter of the single turn coil used in the experiment was 10 mm and the generated peak field was 180 T. Both surfaces of the sample were polished to mirror smoothness. The thickness of the sample was 58±4 μm. The sample was attached to a quartz substrate with a thickness of 321±10 μm.

3. Result and Discussion
Figure 1 shows the measured waveform of the magnetic field and each polarization component ($I_{sig,v}$ (vertical component), $I_{sig,h}$ (horizontal component)) of the light transmitted through the sample and the substrate. The signal ($I_{sig,v}$,$I_{sig,h}$) is converted into the angle ($\theta_{sam\text{+}sub,v}$,$\theta_{sam\text{+}sub,h}$) by using the relation $\theta_{sam\text{+}sub,v}(B(t)) = \arcsin(I_{sig,v}(B(t))/I_{sig,v}(B(0)))\times90/\pi$. The Faraday rotation angle of sample+substrate ($\theta_{sam\text{+}sub}$) was obtained by using the relation $\theta_{sam\text{+}sub} = (\theta_{sam\text{+}sub,v} - \theta_{sam\text{+}sub,h})/2$. Under the ultrahigh magnetic field, the Faraday rotation of the quartz substrate is not negligible and it should be subtracted from $\theta_{sam\text{+}sub}$. We measured the Verde constant of the quartz substrate at 1.96 eV and estimated the Faraday rotation angle of the quartz
substrate $\theta_{\text{sub}}(B(t))$. The Faraday rotation angle of the sample $\theta_{\text{sam}}$ was determined by the difference between $\theta_{\text{sam+sub}}$ and $\theta_{\text{sub}}$. 

Figure 2 shows the field dependence of the Faraday rotation angle of ZnCr$_2$O$_4$ $\theta_{\text{sam}}$. A discernible discontinuity of the Faraday rotation angle is found at 120 T. This discontinuity is considered to represent a first-order phase transition which corresponds to the one observed in CdCr$_2$O$_4$ at 28 T [1]. The critical field in this material is much higher than that in CdCr$_2$O$_4$, which reflects a very strong geometrical frustration. 

**Figure 1.** A waveform of the magnetic field and the vertical (horizontal) polarization component $I_{\text{sig,v}}(I_{\text{sig,h}})$ of light transmitted through the sample and the substrate. The photon energy of the probe light was 1.96 eV. The sample’s temperature was about 9 K which was below $T_N (=12$ K). The field was applied to the [111] direction of the sample. 

Generally, a phase transition can occur when the exchange interaction corresponds to Zeeman energy. The exchange interaction $J$ in ZnCr$_2$O$_4$ was estimated to be -4.5 meV based on the Curie-Weiss temperature [6]. The magnetic field at which the exchange interaction is comparable with Zeeman energy can be estimated to be 26 T. In the theoretical model of the high field magnetization process in this material suggested by Penc et al., a first-order phase transition is expected to occur when the Zeeman energy is about $3J$ [4]. The corresponding magnetic field is estimated to be 78 T. Both expected critical fields are different from the critical field obtained in the experiment. It is necessary that the model is refined. In the region over 120 T, we could not observe a plateau which had been observed in CdCr$_2$O$_4$. In this region, the Faraday rotation angle shows a gradual decrease with increases in the field. This phenomenon could, interestingly, correspond to the decrease of magnetization. However, there exist other possibilities regarding this phenomenon. One is that Faraday rotation in this case does not depend on magnetization only. Although magnetization plays a role in Faraday rotation, it also depends on other field dependent effects including the field dependence of both the electronic structure and optical anisotropy. Even if the magnetization shows a plateau, it is possible that the Faraday rotation does not show a plateau due to these field-dependent effects. Another possibility is the existence of a technical problem. If the removal of the quartz substrate’s Faraday rotation is incomplete, such an anomaly of Faraday rotation could result. The effects causing the decrease of Faraday rotation angle above 120 T could be investigated by changing...
the photon energy of the light source to the non-resonant region below 1.6 eV or by adopting an experimental setup without the involvement of a quartz substrate.

![Graph showing the field dependence of Faraday rotation angle in ZnCr$_2$O$_4$.]

**Figure 2.** The field dependence of Faraday rotation angle in ZnCr$_2$O$_4$.

**4. Summary**

We studied the magnetic properties of a strongly frustrated spinel oxide ZnCr$_2$O$_4$ under an ultrahigh magnetic field up to 180 T by using a Faraday rotation method. We found a finite discontinuity of Faraday rotation angle at 120 T. This is considered to correspond to a first-order phase transition which corresponds to the one observed in CdCr$_2$O$_4$ at 28 T. The critical field of the phase transition was relatively higher than the value estimated by the theoretical model suggested by Penc et al. in which lattice distortions coupled to the spin system and caused a phase transition. A refinement of the model is desired. We could not observe a plateau of Faraday rotation above 120 T, which differs from the magnetization measurement of CdCr$_2$O$_4$. Some possibilities to account for this include the field dependence of optical anisotropy and electronic structure and an incomplete removal of the Faraday rotation of the quartz substrate.

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