Full-magnetization of geometrically frustrated $CdCr_2O_4$ determined by Faraday rotation measurements at magnetic fields up to 140 T

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(Received 10 April 2008; revised manuscript received 22 May 2008; published 19 June 2008)

The total magnetization process of $CdCr_2O_4$, a prototype of the geometrically frustrated Heisenberg spin system, was revealed by Faraday rotation measurements conducted up to 140 T at a wide range of temperatures. We have shown that the Faraday rotation angle is proportional to the magnetization of the system up to the highest magnetic fields. The magnetization processes were well described by a four sublattice Heisenberg spin model [Penc *et al.*, Phys. Rev. Lett. **93**, 197203 (2004); Motome *et al.*, J. Magn. Magn. Mater. **300**, 57 (2006)] including spin-lattice interactions.

DOI: 10.1103/PhysRevB.77.212408 PACS number(s): 75.30.Kz, 75.50.Ee, 78.20.Ls

Geometrical frustration, the competition of interactions due to a geometrical arrangement, is one of the general phenomena that appears in a wide range of solid-state physics. Geometrical frustration appears in various degrees of freedom of electrons (orbital, charge, and spin) and gives rise to unusual macroscopic states of matter.[1–3]

In a frustrated antiferromagnetic Heisenberg spin system, the existence of a macroscopically degenerated ground state (a spin liquid) is theoretically predicted and a real material possessing a spin liquid ground state has been explored over the last several decades. Although many frustrated spin systems have been found, most of them at a low temperature do not show a spin liquid state but an ordered state. This is because the macroscopically degenerated state is metastable and the perturbations (lattice distortions, quantum fluctuations, etc.) lift the degeneracy and reduce the frustration. A frustrated state is very sensitive to any type of perturbations. It is expected that such a frustrated system will show a variety of phases depending on the temperature and the magnetic field.[4]

To understand the essence of the physics of geometrically frustrated spin systems, it is very important to investigate the magnetic phases in magnetic fields up to the full saturation of magnetic moments in a simple real system and compare the observed magnetic properties with those of theoretical models. Chromium spinel oxide $CdCr_2O_4$, in which Cr^{3+} ions form a pyrochlore lattice, is a prototype of geometrically frustrated antiferromagnets. This system is ideal for studying geometrical frustration in that it has the following characteristics: (1) A strong geometrical frustration. Its Curie-Weiss temperature Θ_{CW} is about -70 K. However, antiferromagnetic ordering occurs at 7.8 K, which is well below $|\Theta_{CW}|$. (2) A lack of an orbital degree of freedom (a feature for the Cr^{3+} ion in cubic symmetry). Therefore, the system behaves like a Heisenberg spin system and the Jahn-Teller effect is elucidated. (3) Its exchange energy is relatively large $J(\sim 0.6$ meV). Due to (2) and (3), the system can be described by a simplified model Hamiltonian with a small number of terms. $HgCr_2O_4$ is another candidate for this purpose. Total magnetization process of $HgCr_2O_4$ up to 50 T has already been observed by an induction method.[5] However, the observed field-induced phase transition in $HgCr_2O_4$ is somewhat smeared out probably because of low quality of the sample, such as vacancy of Hg atoms, polycrystalline feature, etc. Therefore, $CdCr_2O_4$ is more suitable for our purpose if a high magnetic field to achieve full saturation of the total spin is available.

It is known that lattice distortions occur in a synchronized manner with antiferromagnetic ordering.[6] It is thought that spin-lattice interactions reduce strong geometrical frustration and lift the macroscopic degeneracy in this system.

Ueda *et al.*[7] reported magnetization measurements of $CdCr_2O_4$ in magnetic fields up to 50 T. They found a first-order phase transition at 28 T followed by a wide magnetization plateau with a half-of-full moment (1/2 plateau). They considered that spins form a 3-up 1-down collinear configuration (3:1) in the 1/2 plateau phase. Their magnetostriction data showed that lattice distortion takes place in association with the first-order phase transition. Recently, Mitamura *et al.*[8] performed the magnetization measurement in magnetic fields up to 80 T at 6 K by the induction method. They have reported a second-order phase transition at 61 T and a first-order phase transition at 77 T.

Penc *et al.*[9] calculated the magnetization process of a pyrochlore spin system under a high magnetic field based on a simple theoretical model that included spin-lattice interaction. They considered that lattice distortion stabilizes the 3-up 1-down collinear spin configuration and causes the 1/2 magnetization plateau. A classical Monte Carlo simulation was extended to a finite temperature by Motome *et al.*,[10] and a detailed phase diagram and high-field magnetization process were predicted for the same system.

On the other hand, recent theoretical reports (Bergman *et al.*[11] or Schulenburg *et al.*[12]) suggested that new phenomena related to Bose-Einstein condensations of magnons (a quantum jump) could occur in geometrically frustrated antiferromagnets in the vicinity of the full moment saturation. In order to justify these theories, an experimental study of magnetization process up to a full moment saturation in $CdCr_2O_4$ has been strongly desired.

A fairly large antiferromagnetically coupled exchange J in this system requires an ultrahigh magnetic field over 100 T for achieving a full moment saturation in the total magnetization process. However, direct magnetization measurements using the induction method are extremely difficult in such extreme conditions like a 100 T region accompanied with

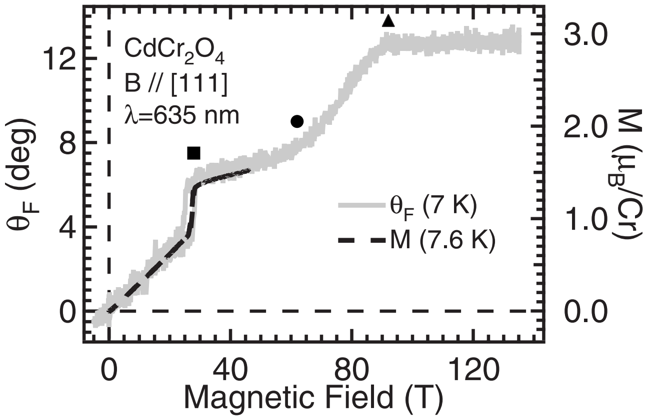

FIG. 1. Solid line: the Faraday rotation angle θ_F at 7 K. A diamagnetic component is subtracted from the raw data. Dotted line: the magnetization M at 7 K up to 50 T at 7.6 K taken from Ref. 7. Symbols (square, circle, and triangle) show critical fields.

noisy environments. The method has been successful only to the materials with sufficiently large absolute value of magnetization.[13,14] Therefore, we adopted a magneto-optical Faraday rotation method as a magnetization probe. This method has been employed for studies of the magnetic properties of materials under ultrahigh magnetic fields.[15,16]

We adopted a single-turn coil method to generate ultrahigh magnetic fields of up to 140 T. The magnetic field was measured by means of a calibrated pick-up coil wound near the sample. The field measurement error was $\pm 3\%$. A semiconductor laser (a coherent "cube") having a central wavelength of 635 nm was used as a light source. A Faraday rotation measurement was performed using a similar method shown in Ref. 15. The detection limit of $d\theta_F/dB$ (θ_F shows a Faraday rotation angle) was 0.1 deg/T. The sample was a single crystal of $CdCr_2O_4$ grown using the flux method. The sample was cut parallel to the [111] crystal surface. In order to ensure an adequate light intensity, the sample thickness was adjusted to an optical density of about one because of the field-duration-limited exposure time ($\sim$ μs) for the optical detection. The adjusted sample thickness was about 40 μm. The magnetic field was applied in the [111] direction. The sample was cooled in a handmade liquid-He flow cryostat made up of phenol resin. The sample temperature was monitored using an Au-Fe/Chromel thermocouple with an accuracy of +1 and −2 K.

θ_F at 7 K is plotted by the gray line in Fig. 1. A diamagnetic component, which is often observed in the high-field region,[17] is subtracted from the raw data. The θ_F exactly coincides with M up to 50 T. Such a rigorous coincidence of the two data up to the high field is nontrivial, since, due to the contributions of higher-order terms, θ_F in general deviate from M as a field increases.[15,16] This close correlation has been obtained by a proper choice of photon energy as the incident laser light in our experiment. This is the first data that allows us to discuss the absolute values of the ultrahigh magnetic-field magnetization processes of solid-state materials.

Several phase transitions (indicated by symbols) can be noticed in θ_F shown in Fig. 1 at 28 T (indicated by a square), 62 T (indicated by a circle), and 90 T (indicated by a triangle). The 1/2 plateau phase (28 T$\sim$62 T) can be clearly observed, and additionally, two phases are observed in the high-field region, one between 62 and 90 T and the other above 90 T.

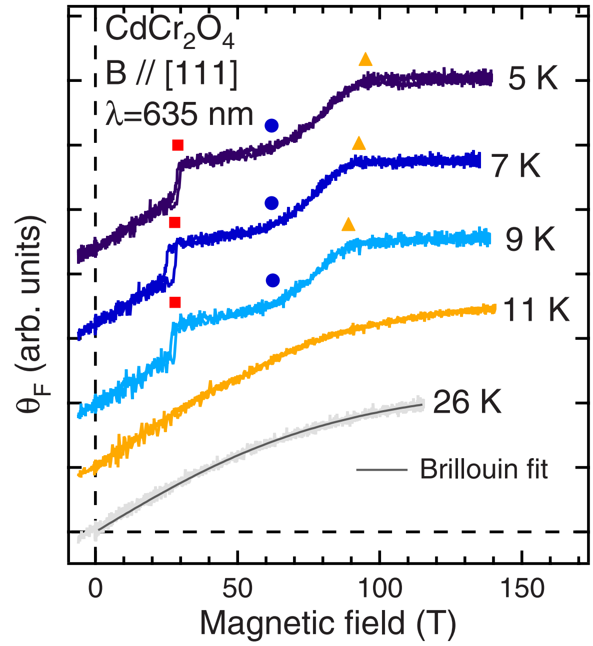

FIG. 2. (Color online) Temperature dependence of θ_F. Solid line (Brillouin fit) shows a fitting curve using a Brillouin function. Symbols (squares, circles, and triangles) show critical fields.

The general features of θ_F in Fig. 1, which can be regarded as proportional to M, and the critical fields (a square, a circle, and a triangle) are in close agreement with those calculated by Penc *et al.*[9] and also by Motome *et al.*[10] Their calculation predicts that a canted 3-up 1-down (Cant 3:1) phase and a moment-saturated (Ferro) phase should appear in higher fields than the 1/2 plateau phase. The θ_F between 62 and 90 T behaves similarly to the Cant 3:1 phase. The region above 90 T should be the Ferro phase since the moment is fully saturated.

A dM/dB measured up to 80 T recently conducted by the induction method[8] showed a very similar behavior to ours except the region of the highest magnetic field where the induction method loses its sensitivity.

The temperature dependence of θ_F is shown in Fig. 2. The θ_Fs below 9 K show almost the same behaviors. However, the position of the triangles shifts to a lower magnetic field by about 10 T on increasing temperature while that of the circles remains almost same. However, that of the squares shows a very weak low-field shift. There a drastic change in θ_F between 9 and 11 K was observed. At 11 K, θ_F at critical fields is smeared out due to thermal effects. These behaviors of the magnetization process shown in Fig. 2 are similar to those of M calculated by Motome *et al.*[10] However, there are two differences. One is that our observed magnetization processes show weaker temperature dependences below 9 K than those calculated. The other is that the θ_Fs below 9 K show full moment plateaus just after the final phase transitions (triangles), whereas the calculated magnetizations show

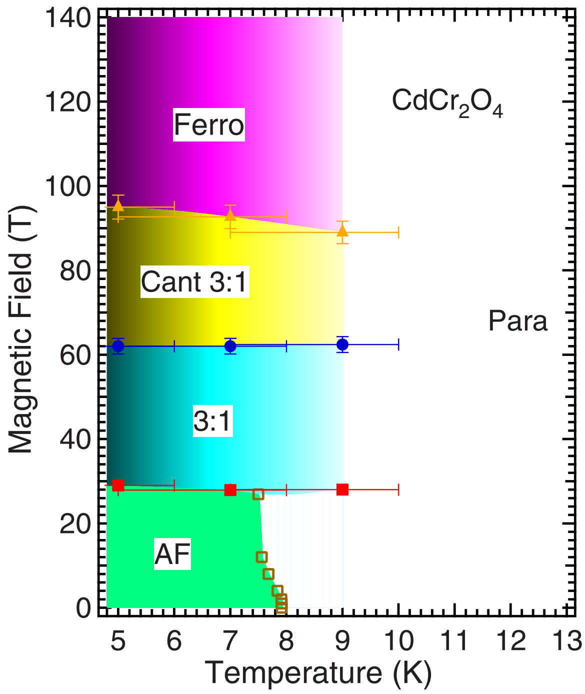

FIG. 3. (Color online) A magnetic phase diagram for $CdCr_2O_4$ determined from the temperature dependence of θ_F shown in Fig. 2. Open squares are taken from Ref. 7.

gradual increase after the final phase transitions. The θ_F at 26 K, which is well above the Neél temperature (=7.8 K), is well fitted by a Brillouin function (the lowest curve in Fig. 2). The effective temperature T_{eff} obtained by the curve fitting is about 106 K. Thus, $\Theta_{\mathrm{CW}}=T-T_{\mathrm{eff}}=-80$ K, which is close to that obtained from the magnetic-susceptibility measurement, −70 K (Ref. 7).

Figure 3 shows a phase diagram made from the critical fields in Fig. 2. The AF-3:1 phase boundary shows a very weak but finite low-field shift on increasing temperature, which corresponds to the previous magnetization measurements up to 50 T by Ueda *et al.* The temperature dependence of the 3:1-Cant 3:1 phase boundary is also weak and the 3:1 phase stably exists between 28 and 62 T below 9 K. On the other hand, the Cant 3:1-Ferro phase boundary shows a clear low-field shift as temperature increases. These show collinear spin configurations such as the 3:1 phase is more robust against thermal fluctuations than noncollinear spin configurations like Cant 3:1 because the biquadratic terms stabilize collinear spin configurations as predicted by Motome *et al.*[10] The boundaries of the Para-3:1 phase as well as the Para-Cant 3:1 phase are hardly identified according to the limit of our measurement.

Here, the values of the critical field of the first-order phase transition and the field width of the plateau at a given temperature observed in $CdCr_2O_4$ are compared with those in $HgCr_2O_4$. We obtained $\beta_{CdCr_2O_4}\sim 0.31(B_c\sim 28\text{ T},B_{\mathrm{sat}}\sim 90\text{ T})$ and $\omega_{CdCr_2O_4}\sim 0.38(W_{\mathrm{plateau}}\sim 34\text{ T})$ for $CdCr_2O_4$, and $\beta_{HgCr_2O_4}\sim 0.24(B_c\sim 10\text{ T},B_{\mathrm{sat}}\sim 42\text{ T})$ and $\omega_{HgCr_2O_4}\sim 0.40(W_{\mathrm{plateau}}\sim 17\text{ T})$ for $HgCr_2O_4$ (Ref. 5). These show $\omega_{CdCr_2O_4}<\omega_{HgCr_2O_4}$ and $\beta_{CdCr_2O_4}>\beta_{HgCr_2O_4}$. Here, the dimensionless parameters, the critical field $\beta\equiv B_c/B_{\mathrm{sat}}$ (B_c is the critical field of the first-order phase transition and B_{sat} is the field at which the moment reaches its full saturation) and $\omega\equiv W_{\mathrm{plateau}}/B_{\mathrm{sat}}$ (W_{plateau} is a width of the plateau) are used. These relations were well explained by the theoretical model of Penc *et al.*[9] when we assume that the coefficient "b" of the biquadratic term "$b(\mathbf{S}_i\cdot\mathbf{S}_j)^2$" in each material holds the relation $b_{CdCr_2O_4}<b_{HgCr_2O_4}$. The spin-lattice interaction in $CdCr_2O_4$ is less strong than that in $HgCr_2O_4$.

Most of the magnetization process in $HgCr_2O_4$ is similar to that in $CdCr_2O_4$, except near the full saturation.[5] At low temperature, θ_Fs in $CdCr_2O_4$ show full moment plateau suddenly after the final phase transition (shown by triangles in Fig. 2), while those in $HgCr_2O_4$ show gradual increase until the full moment saturation. One of the possible reasons is a difference of spin-lattice interaction between the two materials.

In summary, we investigated the magnetic properties of geometrically frustrated $CdCr_2O_4$ up to the full moment under an ultrahigh magnetic field through the use of a high-resolution Faraday rotation measurement. We found surprising close correlation between θ_F and M, which enabled us to discuss the behavior of θ_F in detail and compare it with the existing theoretical models. We observed two phases, Cant 3:1 phase and Ferro phase, in the high-field region above 60 T. The global features of θ_F and the phase diagram coincide closely with the results of model calculations incorporating a biquadratic term. From the present high magnetic-field measurement, the theories proposed by Penc *et al.* and by Motome *et al.*, regardless of their simplicity, describe the essence of the underlying physics of the present magnetic system, which possesses diverse phases arising from the frustrated spins interacting with phonons via the strong spin-lattice coupling. It was found in the present work that the relation $b_{CdCr_2O_4}<b_{HgCr_2O_4}$ holds.

We thank H. A. Katori, H. Mitamura, and Y. Motome for useful and valuable discussions. This work was supported by a Grant-in-Aid for Scientific Research on Priority Areas "High Field Spin Science in 100 T" (Grant No. 451) and a Grant-in-Aid for Scientific Research (Grant No. 20740190) from the Ministry of Education, Culture, Sports, Science, and Technology of Japan (MEXT) and by a Grant-in-Aid for Scientific Research (Grant No. 18740194) from the Japan Society for the Promotion of Science.

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