Successive field-induced transitions in a frustrated antiferromagnet $HgCr_2O_4$

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A new geometrically frustrated chromium spinel $HgCr_2O_4$ was synthesized and characterized. $HgCr_2O_4$ is an antiferromagnet whose exchange constant J is significantly smaller than that of other chromium spinels MCr_2O_4 (M=Mg, Zn, and Cd). We found evidence for antiferromagnetic ordering accompanied by orthorhombic lattice distortions at 5.8 K. By applying a magnetic field, we observed successive transitions from an antiferromagnetic to an intermediate phase and, finally, to a ferromagnetic state. The intermediate state appears as a very wide magnetization plateau from 10 T to 27 T at one-half the full moment of Cr^{3+}, which is very similar to the behavior of $CdCr_2O_4$ above 28 T. The magnetic field required to realize the magnetization plateau phase is considerably small, reflecting the small value of J, which makes it possible to make a more complete study of the physical properties of the system. The facts that the nearest-neighbor interaction is antiferromagnetic and that thermal fluctuation stabilizes the magnetization plateau phase indicate that a collinear three-up and one-down state is realized in this phase. From the obtained H-T phase diagram, this phase is stable even at T=0. We conclude that lattice distortions, in addition to thermal fluctuations, are the most probable cause of the plateau phase.

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I. INTRODUCTION

In recent years, cubic spinels AB_2X_4, which have nonmagnetic A-site cations and magnetic B-site cations, have attracted considerable attention. Antiferromagnetic interactions on a magnetic B-sublattice, which consists of corner-sharing tetrahedra called a pyrochlore lattice, cause highly degenerate ground states. Due to this spin frustration, the system is prevented from ordering and remains in the paramagnetic phase down to very low temperatures. The system is then likely to enter an unconventional ground state displaying unique properties.[1]

However, to achieve an ordered phase, spins tend to couple with some degree of freedom, such as an orbital degree of freedom. If octahedrally coordinated B-site ions have orbital degeneracy, the Jahn-Teller effect dominates the system, and it is difficult to observe the effect of spin frustration. For this reason, we have chosen to study chromium spinels, since Cr^{3+} has three d electrons in triply degenerated t_{2g} orbitals. Without an orbital degree of freedom, chromium spinels with nonmagnetic A-site cations marginally achieve magnetic ordering with the help of lattices. All well-known chromium spinel oxides, $MgCr_2O_4$,[2] $ZnCr_2O_4$,[3] and $CdCr_2O_4$,[2] undergo a transition from a paramagnetic cubic phase to a Néel-ordered tetragonal one with lattice distortions, which can be interpreted as analogues of the spin-Peierls transition to relieve spin frustration.[3] In the presence of spin-lattice couplings, lattice distortions relieve spin frustration, which has been studied theoretically[4–7] and tested experimentally in other systems with a pyrochlore lattice.[8,9]

Recently, we found a metamagnetic transition at 28 T and a subsequent very wide magnetization plateau with one-half of the full moment of S=3/2 Cr^{3+} in $CdCr_2O_4$.[10] This is the first example of a magnetization plateau found in a pyrochlore Heisenberg antiferromagnet. The origin of this phenomenon is very likely to be different from that of the plateau found in Ising pyrochlore magnets.[11–13] We have proposed a collinear three-up and one-down spin configuration out of four spins on a Cr tetrahedron. In this spin configuration, three antiferromagnetic bonds, which are favored by predominant antiferromagnetic interactions, are realized out of six bonds in one tetrahedron to reduce the magnetic energy. However, experimental results offering further insight into this phenomenon are still lacking, as the critical field of 28 T is too high to be studied using a conventional superconducting magnet.

In this paper, we focus on another member of chromium spinels with Hg^{2+} as the A-site cation, of which only the synthesis has been previously reported.[14,15] By comparison with other chromium spinels, we expect that $HgCr_2O_4$ is an antiferromagnet with small exchange energy J that shows a half-magnetization plateau at a smaller magnetic field. In this paper, we report the basic physical properties of $HgCr_2O_4$ and the behaviors of its magnetization process under high magnetic fields. At low temperatures, $HgCr_2O_4$ shows a transition to a magnetically ordered state with orthorhombic structural distortions. As is expected, we found a magnetization jump followed by a plateau with a half moment under a relatively small magnetic field due to the small J. Furthermore, we clearly observed the magnetization process up to the full moment and then found another transition to a ferromagnetic state. As a result, we obtained a complete H-T phase diagram of this material. From the effect of thermal fluctuation, it is strongly indicated that the magnetization plateau phase has a collinear spin structure. From the stability at T=0, we conclude that spin-lattice couplings play a crucial role to stabilize the plateau phase.

II. EXPERIMENTAL DETAILS

Powder samples of $HgCr_2O_4$ were prepared by a thermal decomposition of Hg_2CrO_4 (Refs. 14 and 15) in an evacuated silica tube. As references, other chromium spinels were pre-

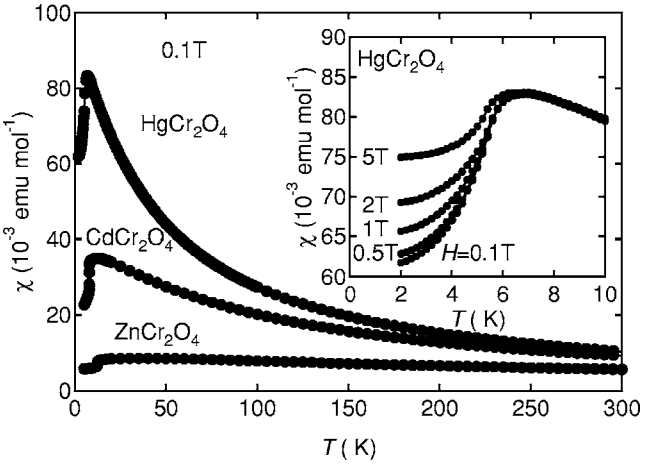

FIG. 1. Temperature dependence of the magnetic susceptibility of $HgCr_2O_4$ with those of $ZnCr_2O_4$ and $CdCr_2O_4$. The inset exhibits the field dependence at low temperatures.

pared as follows. All polycrystalline samples were synthesized by conventional solid-state reactions. Single crystals of $CdCr_2O_4$ were grown by the flux method. Single crystals of $ZnCr_2S_4$ and $ZnCr_2Se_4$ were obtained by the vapor transport methods. The dc magnetic susceptibilities up to 5 T were measured using a commercial superconducting quantum interference device magnetometer. Magnetization measurements up to 47 T were performed using an induction method with a multilayer pulse magnet at the ultrahigh magnetic field laboratory of the Institute for Solid State Physics at the University of Tokyo. X-ray diffraction measurements at low temperatures were conducted in a He gas flow cryostat using a diffractometer with a Cu $K\alpha$ source. The signals from Cu $K\alpha_2$ were numerically subtracted from raw data.

III. RESULTS AND DISCUSSIONS

A. Magnetic properties

Among various chromium spinels with nonmagnetic A-site cations, all oxide spinels are antiferromagnets, whereas sulfide and selenide spinels have predominantly ferromagnetic interactions.[16] $HgCr_2O_4$ is expected to have antiferromagnetic interactions, which cause spin frustration on a pyrochlore lattice. The temperature dependence of magnetic susceptibility χ of $HgCr_2O_4$ is shown in Fig. 1 together with those of $ZnCr_2O_4$ and $CdCr_2O_4$. Susceptibility data at high temperatures are well fitted by the Curie-Weiss law, which gives an effective moment p_{eff} of $3.72\mu_B$, and the Curie-Weiss temperature Θ_{CW} of -32 K. The value of p_{eff} is reasonable compared with $3.87\mu_B$, which is the value expected for $S=3/2$ and $g=2$. The magnitude of $|\Theta_{\mathrm{CW}}|$ for $HgCr_2O_4$ is substantially small compared with those for $MgCr_2O_4$, $ZnCr_2O_4$, and $CdCr_2O_4$, whose Θ_{CW} are -370 K, -390 K,[17] and -70 K,[10] respectively. Based on molecular field approximation, assuming that only nearest-neighbor interactions are considered, the exchange constant $J=-3k_B\Theta_{\mathrm{CW}}/zS(S+1)$ is proportional to $|\Theta_{\mathrm{CW}}|$, where $z=6$ is the number of nearest-neighbor interactions. Going from $MgCr_2O_4$ to $HgCr_2O_4$, J is reduced by the increase of the ionic radius of the A-site cation. In addition to the variation of the Cr-Cr distances, the Cr-O-Cr angles are also affected. Even though the oxygen parameters u and the Cr-O-Cr angles are almost the same for $MgCr_2O_4$, $ZnCr_2O_4$, and $CdCr_2O_4$, the J of $CdCr_2O_4$ is much smaller than those of $MgCr_2O_4$ and $ZnCr_2O_4$. Hence, we are convinced that the reduction of J is attributed mainly to the variation of the Cr-Cr distances and, therfore, the decrease of orbital overlapping. This is reasonably explained by considering that chromium ion is a t_{2g} system. The t_{2g} orbital of a chromium atom points toward the next chromium atom. Therefore, direct exchange interaction is more important than the superexchange through an oxygen atom.

Below 5.8 K, the susceptibility drops steeply, indicating an antiferromagnetic ordering. There is no difference between field-cooled and zero-field-cooled measurements at $H=0.1$ T. In the low field, the value of χ extrapolated to 0 K is about two thirds of χ just above the transition, which is typical for three-dimensional antiferromagnets. In the inset of Fig. 1, susceptibilities within a low-temperature region are plotted under various magnetic fields. Below the transition temperature, χ is field-dependent, as in $ZnCr_2O_4$,[18] which shows antiferromagnetic orderings. This field dependence of χ is well explained to be due to the movement of antiferromagnetic domains, which suggests that almost the same situation takes place in $HgCr_2O_4$. Indeed, below this transition temperature, an antiferromagnetic ordering is confirmed by neutron-scattering measurements.[19]

The Néel temperature T_N of $HgCr_2O_4$ is smaller than those of $MgCr_2O_4$, $ZnCr_2O_4$, and $CdCr_2O_4$, which are 12.5 K, 12 K, and 7.8 K, respectively.[17] Considering the values of Θ_{CW}, the index of frustration $T_N/|\Theta_{\mathrm{CW}}|$ for $HgCr_2O_4$ is notably larger than those of other chromium spinels, suggesting that the frustration is less strong in $HgCr_2O_4$. The increase of $T_N/|\Theta_{\mathrm{CW}}|$ is due to the enhancement of T_N compared with J. There should be a certain interaction to stabilize an ordered state. One likely origin is further-neighbor interactions. Further-neighbor interactions play an important role in stabilizing magnetic ordering in an antiferromagnetic pyrochlore lattice. Although we cannot estimate the sign or the value of further-neighbor coupling constants at present, order-favoring interactions seem to work effectively in $HgCr_2O_4$. Another interpretation is the effect of strong spin-lattice couplings. Antiferromagnetic orderings in chromium spinels are always observed with lattice distortions. If spin-lattice couplings are strong enough, Néel ordering with a lattice distortion is stabilized even at relatively higher temperatures. Assuming that $HgCr_2O_4$ has strong spin-lattice couplings, a structural transition should be expected.

B. Structural distortions

For other chromium spinels with non-magnetic A-site cations, the transition to the Néel-ordered state is of the first order, and a structural transition takes place simultaneously at T_N due to spin-lattice couplings. Similarly, we expected a structural transition for $HgCr_2O_4$, although the cooling and heating measurements of χ gave no hysteresis as a function of the temperature, thus giving no indication of a first-order transition. We measured the powder x-ray diffraction at low

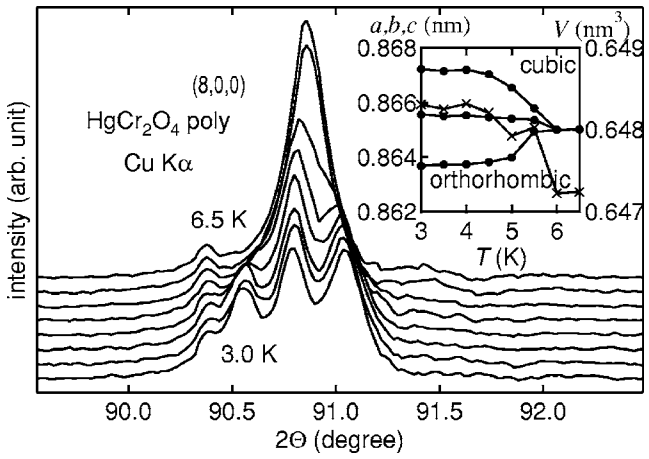

FIG. 2. X-ray diffraction spectrum of $HgCr_2O_4$ obtained at low temperatures. The lines on this graph are drawn every 0.5 K from 3.0 K to 6.5 K and are shifted to avoid overlapping. The inset represents the temperature dependence of the lattice parameters (filled circles) and cell volume (crosses) calculated from the (8, 0, 0) diffraction.

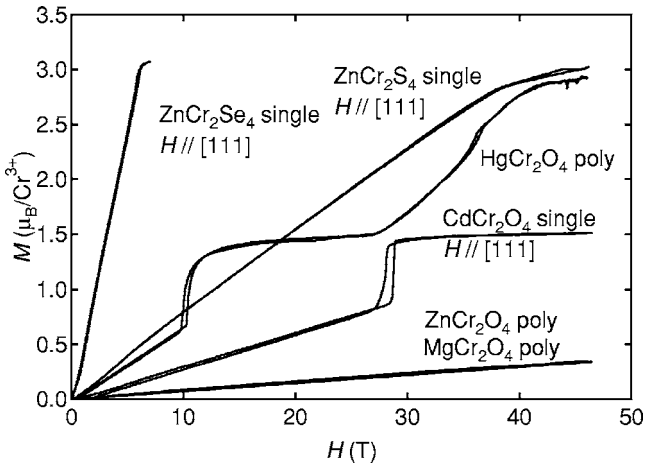

FIG. 3. Magnetization curves of various chromium spinels that exhibit Néel ordering at low temperatures. $HgCr_2O_4$ and $CdCr_2O_4$ are measured at 1.8 K, and others, at 4.2 K.

temperatures. Figure 2 represents the temperature dependence of the diffraction pattern of the (8, 0, 0) signal. Above 6 K, only one peak is observed, reflecting the cubic symmetry of the system. The signal around 90.4° is due to an impurity. Below 6 K, the signal becomes broad and finally splits into three peaks, suggesting a structural transition. This transition temperature is the same as the magnetic-ordering temperature within the experimental error. We conclude that this structural change is accompanied by magnetic ordering. The splitting of the (8, 0, 0) diffraction implies that the length of the lattice parameters $a=b=c$ achieves three different values, namely, the crystal structure varies from a cubic to at least an orthorhombic symmetry. No extra diffraction that violates the extinction rule is observed. Only splitting and broadening of each diffraction are observed due to the change of a,b,c. Hence, the space group is supposed to be an orthorhombic *Fddd*. This is in clear contrast with other chromates with a tetragonal $I4_1/amd$ symmetry at low temperatures. In chromium spinels, lattice distortions in the low-temperature phase are closely related to the magnetic structure, since there is no Jahn-Teller effect. For $HgCr_2O_4$, the different crystal structure suggests that a different magnetic structure is realized at low temperature. It is likely that the subtle interplay of the spin and lattice causes different magnetic structures and different structural distortions in chromium spinels.

The temperature variation of the lattice parameters and the volume of unit cell V are plotted as a function of the temperature in the inset of Fig. 2. The volume change at low temperature is $\Delta V/V=1.6\times10^{-3}$. For other spinels, $ZnCr_2O_4$ and $CdCr_2O_4$,[20] $|\Delta V/V|<0.5\times10^{-3}$. The large volume change in $HgCr_2O_4$ is a clue to the softness of the lattice, which is likely due to the large A-site cation. Since different magnetic ordering patterns are realized among $ZnCr_2O_4$, $CdCr_2O_4$, and $HgCr_2O_4$ at the ground state,[19] it is difficult to compare the strength of the spin-lattice couplings of these three systems. However, from the softening of the lattice, spin-lattice couplings appear to be significantly enhanced in $HgCr_2O_4$. This is the most likely origin of the small frustration index, as previously discussed.

C. Magnetization process

The predominant antiferromagnetic interactions in $HgCr_2O_4$ led us to think about the half-magnetization plateau observed in $CdCr_2O_4$.[10] In the magnetization process of $CdCr_2O_4$, we found a magnetization jump at 28 T followed by a magnetization plateau at the value of one half-moment of Cr^{3+}, as shown in Fig. 3. To understand the behavior of the magnetization plateau with one-half moment, we proposed a collinear spin arrangement in which three spins are up and one spin is down in one chromium tetrahedron. Under a very high magnetic field, up spins are favored. Hence, ferromagnetic bonds tend to increase in general. If antiferromagnetic interactions are dominant, the system will try to maintain the number of antiferromagnetic bonds. In the three-up and one-down collinear spin structure, there are three antiferromagnetic bonds out of six bonds in one tetrahedron. This spin arrangement satisfies both conditions, i.e., that magnetic moment is one-half of the full moment and that there remains a relatively large number of antiferromagnetic bonds in spite of the many up spins. A very wide magnetization plateau quite likely indicates the appearance of this collinear spin structure.

This magnetization plateau is achieved by the balance of the magnetic interaction J and the applied field H. Given the small $|\Theta_{CW}|$ and J values of $HgCr_2O_4$, a plateau behavior can be expected for relatively small magnetic fields. A magnetization curve up to 47 T, measured at 1.8 K, is plotted in Fig. 3, together with those of other chromium spinels that show Néel ordering for comparison. As expected, the magnetization of $HgCr_2O_4$ jumps at 10 T, and the following magnetization plateau up to 27 T is observed. The magnetization jump has a pronounced hysteresis, as is observed in

$CdCr_2O_4$, indicating a first-order transition. A structural change is also expected to take place at this transition, as in $CdCr_2O_4$.[10] This step feature of $HgCr_2O_4$ is somewhat round compared with that of $CdCr_2O_4$. We suppose that this is due to the effect of disorder in the *A*-site cation, since a similar rounded shape is observed in roughly synthesized $CdCr_2O_4$ polycrystalline samples, which likely contain a significant amount of disorder. The behaviors of $HgCr_2O_4$ and $CdCr_2O_4$ are in clear contrast to the magnetization curves of $ZnCr_2S_4$ and $ZnCr_2Se_4$, which are almost linear up to the full moment of Cr^{3+}. Considering that these chalcogenide spinels have predominant ferromagnetic interactions, antiferromagnetic interactions in oxide spinels are essential for this half-magnetization plateau. As mentioned before, we have proposed a collinear three-up and one-down spin configuration to explain the half-magnetization plateau of $CdCr_2O_4$.[10] It is very likely that the same situation takes place in $HgCr_2O_4$ as well. These ideas are supported by a recent theory.[21] For $MgCr_2O_4$ or $ZnCr_2O_4$, no indication of a magnetization plateau is observed in these experiments. Even if they could demonstrate such behavior, the magnetic fields in our measurements are too small to stabilize a half-magnetization plateau phase due to their large *J* values.

The critical field $H_c = 10$ T of the plateau phase is much smaller than that of $CdCr_2O_4$ of 28 T, reflecting a difference in *J*. To compare these two materials, we must normalize the magnetic field *H* by *J*. In doing so, the initial magnetization curves before the plateau phase almost overlap. However, the normalized critical fields are different for these two compounds, which appear as different values of magnetization just before the plateau phase in the figure. Assuming $g=2$, we obtain normalized critical fields $H_c g\mu_B/J$ of 2.25 and 1.735 for $CdCr_2O_4$ and $HgCr_2O_4$, respectively, using *J* values obtained from a molecular field approximation. The normalized field $Hg\mu_B/J$ corresponds to h/J, which is used in the letter regarding theory.[21] The theory suggests that lattice distortions play a crucial role in the stabilization of the collinear spin configuration and the half-magnetization plateau state and that larger spin-lattice couplings lead to a smaller critical field. The reduction of $H_c g\mu_B/J$ in $HgCr_2O_4$ implies rather strong spin-lattice couplings in this system. The theory predicts that the transition to the plateau phase occurs where h/J is within the range of 2.4 to 3.2. The value for $CdCr_2O_4$ is in good accordance with this theory; however, it is remarkably small for $HgCr_2O_4$. One possible reason for this discrepancy is the overestimation of *J* by a mean field approximation. Another likely origin is due to further-neighbor interactions and the quantum effect, which are not considered in the theory. In particular, further-neighbor interactions should have extremely large effects on the selection of one certain spin configuration from all possible configurations that satisfy the condition that each chromium tetrahedron contain three-up and one-down spins.

In addition to the observation of the magnetization plateau, we obtained the magnetization curve up to the full moment at the lowest temperature, 1.8 K. Above 27 T, the magnetization again starts to increase. This magnetization increase after the plateau phase apparently corresponds to a canted three-up one-down state. Together with this, around 37 T, we observe a sudden change of the magnetization slope, which seemingly has a hysteresis. This very likely implies the appearance of another phase, which, we suppose, is an unsaturated ferromagnetic state. Then, the magnetization increases up to the full moment of Cr^{3+} at about 42 T, for which we have a field-stabilized ferromagnetic state.

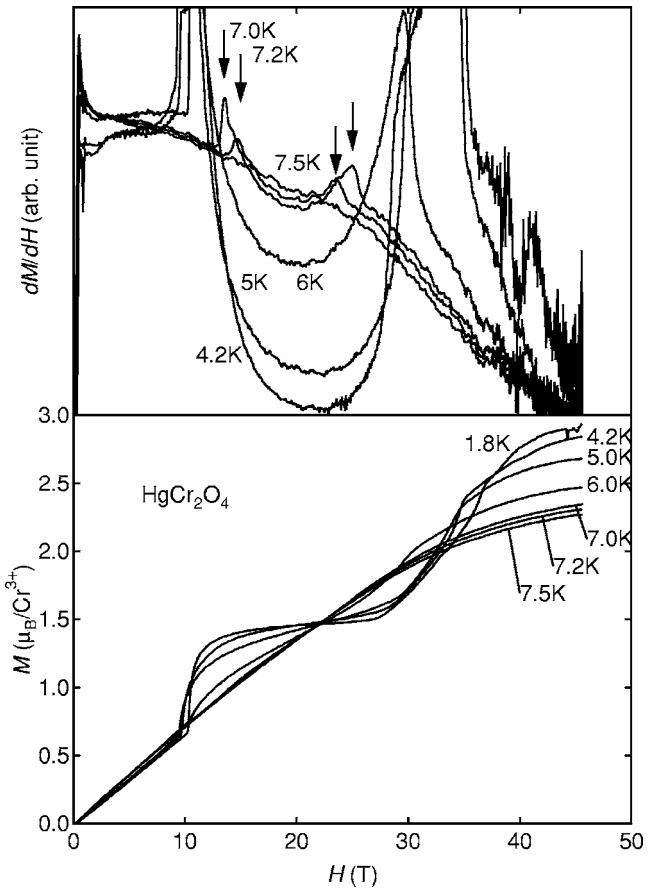

FIG. 4. Temperature dependence of the magnetization curves of $HgCr_2O_4$ measured in the process of field increase. The upper panel describes the derivation of magnetization dM/dH above 5 K, and the arrows indicate the signals that correspond to the beginning and ending of the plateau phase.

To clarify the stability of the magnetization plateau phase against the temperature, we measured the temperature dependence of the magnetization curves, as shown in Fig. 4. Please note that the slope dM/dH in the low-field region is different below and above T_N, which is consistent with the temperature dependence of magnetic susceptibility. Although the plateau behaviors can be clearly distinguished up to 6 K, it is hard to find them above 6 K. However, from the curves of the derivatives given in the upper panel of Fig. 4, we can easily detect the signals that correspond to the boundaries of the plateau phase, which are indicated as arrows. Anomalies that probably indicate the phase boundary between the canted three-up one-down phase and the unsaturated ferromagnetic phase also are observed. Finally, at 7.5 K, no signals that suggest the existence of a phase transition are observed.

These phase boundaries are plotted in Fig. 5 to draw a *H-T* phase diagram. This phase diagram within a low-field region is very similar to that of $CdCr_2O_4$.[10] Below T_N, the

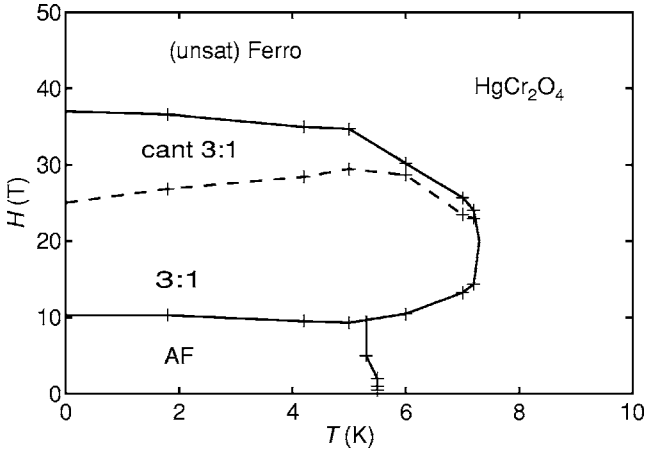

FIG. 5. *H-T* phase diagram of $HgCr_2O_4$. The solid lines represent a first-order transition with a structural change, and the broken line, a second-order transition. The abbreviations used are as follows: Para, paramagnetic; AF, antiferromagnetic; 3:1, three-up one-down; cant 3:1, canted three-up one-down; (unsat) Ferro, ferromagnetic or unsaturated ferromagnetic.

critical field to the plateau phase slightly decreases with raising temperature and shows an upturn above T_N. Conversely, the upper field limit of the plateau phase increases with temperatures below T_N and then decreases above T_N. In addition, the phase boundary that we assign between the canted three-up one-down phase and the unsaturated ferromagnetic phase monotonically decreases with temperature. At around 7.5 K, these phase boundaries are probably connected. These features are similar to recent Monte Carlo simulations,[22] except for the behavior of the upper limit of the plateau phase.

Due to strong spin-lattice couplings in this compound, the lattice structures are supposed to be different among different magnetic phases in this phase diagram. Indeed, a structural change is observed in the half-magnetization plateau region for $CdCr_2O_4$. Considering the symmetry of the magnetic orderings in this *H-T* phase diagram, we predict that three kinds of lattice structures are realized, namely, a cubic (unsaturated) ferromagnetic phase that connects to the paramagnetic phase, an orthorhombic antiferromagnetic phase, and a (canted) three-up one-down phase with an unknown lattice structure, which are separated by solid lines in the phase diagram. We suggest that two successive field-induced structural transitions take place below T_N, between the antiferromagnetic phase and the three-up one-down phase, as is observed for $CdCr_2O_4$, and between the canted three-up one-down phase and the ferromagnetic phase. At these phase boundaries around 10 T and 37 T, first-transition natures are observed as a hysteresis of the magnetization curve measured at 1.8 K in Fig. 3. For $T_N < T < 7.5$ K, reentrant structural changes are expected to be observed.

This *H-T* phase diagram provides important information about the origin of the half-magnetization plateau phase. Now, we are able to compare the phase diagrams of the magnetization plateau in other Heisenberg antiferromagnets on a frustrated lattice, such as triangular[24] and kagomé lattices.[25] By applying a magnetic field, a collinear two-up and one-down spin configuration, which is observed as a one-third-magnetization plateau, is realized for them. The origin of the magnetization plateau in these systems is thermal and quantum fluctuations, which favor a collinear spin configuration.[23] At $T=0$, a collinear structure appears due to quantum fluctuations, normally in a very narrow region. By raising the temperature, thermal fluctuations stabilize collinear spin configurations, and the magnetization plateau phase expands on the *H-T* phase. Our observation of the magnetization plateau in chromium spinels corresponds to such a phenomenon on a pyrochlore lattice. Indeed, by raising the temperature, the plateau phase becomes more stable below T_N, suggesting that the magnetization plateau phase takes a collinear spin configuration. In this respect, the *H-T* phase diagram of $HgCr_2O_4$ is qualitatively similar to those for triangular and kagomé lattices. However, the feature that distinguishes chromium spinels from these is their quite wide magnetization plateau phase even at the $T=0$ limit. The half-magnetization plateau phase extends over more than 10 tesla for both $CdCr_2O_4$ and $HgCr_2O_4$. This robustness against the magnetic field strongly indicates the existence of additional effects to stabilize the plateau phase. Considering that the crystal structure is quite likely deformed in the magnetization plateau phase, lattice distortions are the most likely candidate, as is proposed by our previous paper[10] and the paper regarding theory.[21]

Likewise, $MgCr_2O_4$ and $ZnCr_2O_4$ are known as systems with large spin-lattice couplings. Hence, we can expect that spin-lattice couplings in these materials stabilize the half-magnetization plateau under a high magnetic field. If we assume that the normalized critical field has almost the same value as $HgCr_2O_4$ or $CdCr_2O_4$, the critical field will be 10 times larger, namely, over 100 T due to the large *J* values. It is very difficult to measure magnetization over 100 T, but it is not impossible. Exploration into the magnetization plateau phase in these systems is under planning.

IV. CONCLUSIONS

We synthesized a new geometrically frustrated chromium spinel, $HgCr_2O_4$, and evaluated the physical properties of this material. $HgCr_2O_4$ can be regarded as an antiferromagnet with a small exchange constant. Like other chromium spinels, it shows a magnetic ordering with structural distortions at low temperatures; however, the symmetry of the lattice is orthorhombic as opposed to tetragonal, and the volume change at the transition is substantially larger. It is very likely that $HgCr_2O_4$ has rather strong spin-lattice couplings. By applying a magnetic field, a very wide magnetization plateau phase was realized, as is observed for $CdCr_2O_4$, and another transition to the ferromagnetic state was found. We successfully obtained a complete *H-T* phase diagram, which suggests that lattice distortions play a key role augmented by the effect of thermal fluctuations. Similar behaviors are expected for $MgCr_2O_4$ and $ZnCr_2O_4$ with strong spin-lattice couplings. The critical field for $HgCr_2O_4$ is relatively small and can be achieved by conventional superconducting magnets. Further experiments to clarify the physics behind the magnetization plateau behavior observed in the Heisenberg antiferromagnet on a pyrochlore lattice are in progress.

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