Observation of a novel phase transition induced by a magnetic field in the pyrochlore spin ice compound

2007 J. Phys.: Condens. Matter 19 145269

(http://iopscience.iop.org/0953-8984/19/14/145269)

IOP PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **19** (2007) 145269 (9pp) doi:10.1088/0953-8984/19/14/145269

Observation of a novel phase transition induced by a magnetic field in the pyrochlore spin ice compound

K Matsuhira[1], H Sato[2], T Tayama[2], Z Hiroi[2], S Takagi[1] and T Sakakibara[2]

[1] Department of Electronics, Faculty of Engineering, Kyushu Institute of Technology, Kitakyushu 804-8550, Japan
[2] Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581, Japan

E-mail: matuhira@elcs.kyutech.ac.jp

Received 31 August 2006
Published 23 March 2007
Online at stacks.iop.org/JPhysCM/19/145269

Abstract

We present a novel field-induced phase transition in the spin ice compound $Dy_2Ti_2O_7$ that is investigated by means of angle-resolved DC magnetization measurements. In the near [112] field, the magnetization component perpendicular to the Kagomé plane shows a first-order phase transition below 0.27 K. This transition is driven by a ferromagnetic ordering in the field-decoupled spins on the triangular lattice. Furthermore, we revealed another first-order phase transition below 0.52 K in a magnetic field between the [110] and [111] directions. This phase transition is related to a breakdown of ferromagnetic Ising spin chains along $[1\bar{1}0]$ or $[\bar{1}10]$.

1. Introduction

Recently, pyrochlore compounds have been attracting a great deal of interest because spins reside on the vertices of corner-shared tetrahedra [1–6]. In the pyrochlore lattice, a ferromagnetic coupling between the spins leads to full frustration in the case of a strong single-ion Ising anisotropy along the ⟨111⟩ axes [4, 7]. Consequently, stable spin configurations for each tetrahedron obey the ice rule, i.e., two spins pointing outward and two spins pointing inward (the 2-in 2-out state) in a basic tetrahedron are stabilized [4]. As a result, the ground state of the entire spin system is highly degenerate and a static disordered (spin ice) state is formed at low temperature, in spite of the structural order of the lattice. In the typical spin ice compound $Dy_2Ti_2O_7$ (the effective ferromagnetic coupling $J_{\rm eff} = 1.1$ K), a macroscopically degenerate ground state analogous to water ice is realized in zero magnetic field below ∼0.5 K [8]. An interesting aspect of the spin ice system is that the ground state degeneracy can be removed by applying a magnetic field [4]. Ramirez *et al* studied the effect of a magnetic field on the powdered specimen of $Dy_2Ti_2O_7$ by calorimetric measurements [8]. Interestingly, for fields higher than 1 T, the specific heat data revealed three sharp peaks at 0.34, 0.47 and 1.12 K,

suggesting new phase transitions in magnetic fields. Furthermore, their critical temperatures were insensitive to the field strength.

Since then, the field-induced phase transition in the pyrochlore spin ice has been progressively investigated in several studies. The anomaly at 1.12 K is ascribed to an ordering in a field applied along the [110] direction of the cubic unit cell [9–14]. Under the [110] field, half of the spins have their Ising axes oriented perpendicular to the field and are decoupled from the field. These spins behave as unique ferromagnetic Ising spin chains, weakly interacting with each other through a long-range dipolar interaction. An intra-chain spin freezing is expected with a rather sharp feature in the specific heat at ∼1.1 K [11]. Another sharp feature at 0.47 K is ascribed to a novel first-order phase transition associated with the collapse of a Kagomé ice state in a [111] field [15–18]. The pyrochlore lattice consists of Kagomé and triangular planes stacked alternately along the [111] direction. When a magnetic field is applied along the [111] direction, spins on the triangular planes first align along the field direction because they have their Ising axes parallel to [111]. However, spin configurations on the Kagomé planes still have a macroscopic degeneracy under the 2-in 2-out ice rule. This intermediate state with a finite residual entropy is called Kagomé ice [15]. When the applied field increases further, the spin configuration of 2-in 2-out turns into a fully polarized state (1-in 3-out or 3-in 1-out) through a first-order transition of liquid–gas-type at ∼0.9 T with the critical point $T_{cr} \sim 0.4$ K [17].

Furthermore, recently, the origin of the 0.34 K anomaly has been theoretically proposed by Ruff *et al* [14]. The new phase transition is ascribed to the ordering in the triangular lattice near the [112] field, which is slightly canted toward [111] to cancel internal nearest-neighbour exchange plus the dipolar local field. Considering that a strong field is applied along the [112] direction parallel to the $(\bar{1}\bar{1}1)$ Kagomé plane, the spins on Kagomé planes are aligned toward the field direction. On the other hand, the spins on triangular planes do not couple to the field and are free. Interestingly, Ruff *et al* pointed out that a ferromagnetic coupling arising from a long-range dipolar interaction as well as a weak antiferromagnetic third-nearest-neighbour interaction leads to a ferromagnetic ordering of the field-decoupled spins at $T_c \sim 0.35$ K [14]. The predicted phase transition near the [112] field has been studied by Higashinaka *et al* by AC magnetic susceptibility measurements on a single crystal of $Dy_2Ti_2O_7$ [19]. They obtained a signature of the predicted phase transition. However, the observed $[11\bar{1}]$ susceptibility did not show a clear divergence at the critical temperature. Based on their experiment, we cannot conclude that the anomaly is a ferromagnetic ordering, as theoretically predicted.

As mentioned above, some unsolved problems on the field-induced phase transition remain. We have researched the field-induced phase transition by means of field-angle dependent DC magnetization measurement. The field-angle dependent DC magnetization measurement is very useful in revealing a new field-induced phase transition. In the present paper, we report the results obtained for two field-induced phase transitions of $Dy_2Ti_2O_7$. One transition is a ferromagnetic ordering in the triangular lattice, as theoretically predicted. Another is a first-order phase transition below 0.5 K in a magnetic field between the [110] and [111] directions. This phase transition is related to a breakdown of ferromagnetic Ising spin chains along $[1\bar{1}0]$ or $[\bar{1}10]$. To our knowledge, this is the first investigation of this transition.

2. Experimental details

A single crystal of $Dy_2Ti_2O_7$ was grown by the floating-zone method as described previously [15]. A thin plate-like crystal of size $0.25 \times 2 \times 2$ mm^3 and having a weight of 6.99 mg was prepared for magnetization measurements. The largest face was the $(1\bar{1}0)$ plane. To minimize the demagnetization field effect, the field direction was applied along the $(1\bar{1}0)$ plane. Angle-resolved magnetization measurements were performed by the Faraday method

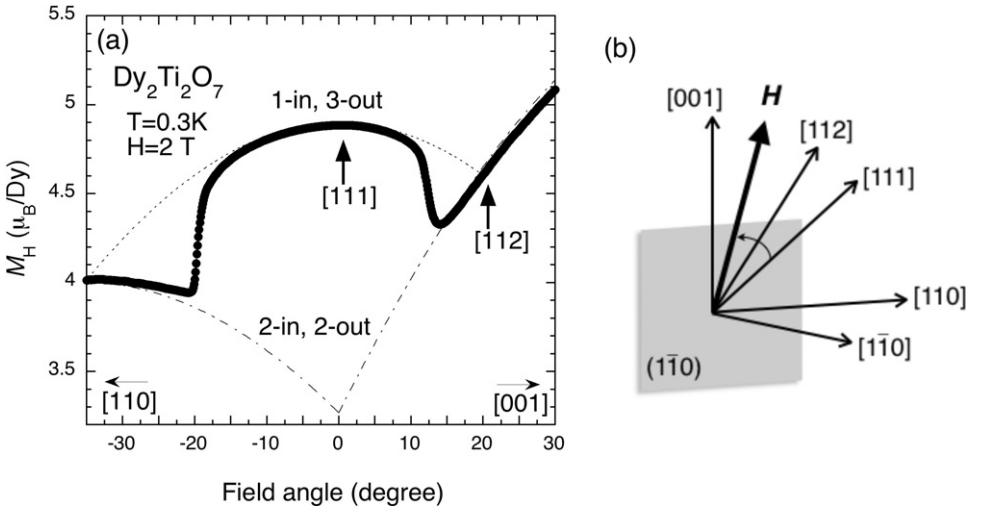

Figure 1. (a) Field-angle variation of the magnetization for $Dy_2Ti_2O_7$ obtained at 0.3 K in a field of 2 T rotated in the $(1\bar{1}0)$ plane. The broken and dash–dotted lines show the calculated angular dependence of the saturation magnetization for 1-in 3-out and 2-in 2-out states, respectively. (b) Field applied in the $(1\bar{1}0)$ plane.

using a ^{3}He cryostat, as described previously [20]. Rotating the cryostat using a stepper motor controlled by a computer, the magnetization was measured as a function of the field angle with a resolution of 0.01°. The accuracy of the crystal orientation was within 0.5°. Demagnetization field correction was performed using a demagnetization factor of $N \sim 0.08$ [20]. Here, H^* denotes the field after the demagnetization field corrections.

3. Results and discussion

Figure 1(a) shows the field-angle variation of the magnetization M_H ($\boldsymbol{M}\cdot(\boldsymbol{H}/H)$) for $Dy_2Ti_2O_7$ obtained at 0.3 K in a field of 2 T rotated in the $(1\bar{1}0)$ plane, where $\boldsymbol{M}$ is the magnetization and $\boldsymbol{H}$ is the applied magnetic field. The broken and dash–dotted lines show the calculated saturation magnetization for 1-in 3-out and 2-in 2-out states, respectively. As shown in figure 1(b), the main directions, such as [110], [111], [112] and [001], are included in the $(1\bar{1}0)$ plane. At 0.3 K in a field of 2 T, the magnetizations along the [110], [112] and [001] directions saturate to the calculated saturation magnetization for the 2-in 2-out state. On the other hand, the magnetization along the [111] direction saturates to the calculated saturation magnetization for the 1-in 3-out state. Interestingly, we observed a discontinuous change from the 2-in 2-out state to the 1-in 3-out state around field angles of $-19.6°$ and 12.4°. The discontinuous change of spin configuration suggests a phase transition.

First, we focus on the discontinuous change of M_H between the [111] and [112] directions. Recently, we have obtained the first thermodynamic evidence for a ferromagnetic transition of the field-decoupled spins on the triangular lattice in a field near the [112] direction [14, 20]. In figure 2(a), we show the field-angle variation of the magnetization M_H obtained at temperatures above 0.29 K in a field of 2 T rotated in the $(1\bar{1}0)$ plane, where θ is the angle between $\boldsymbol{H}$ and the [112] direction (figure 2(b)). The data in figure 2 were obtained by rotating the field from $\theta = 5°$ to $-25°$, passing through the [111] direction ($\theta = -19.4°$). In the field reverse rotation from $\theta = -25°$ to 5°, we detected no difference in the magnetization behaviour. The broken

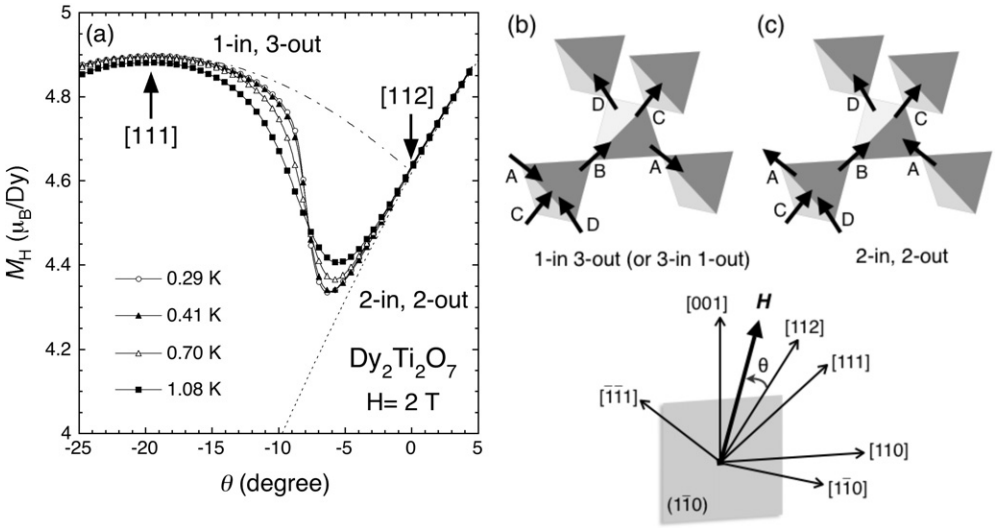

Figure 2. (a) Field-angle variation of the magnetization for $Dy_2Ti_2O_7$ obtained at various temperatures in a field of 2 T rotated in the (1$\bar{1}$0) plane. The broken and dash–dotted lines represent the field-angle variation of the calculated saturation magnetizations for the 2-in 2-out and 1-in 3-out states, respectively. The field-angle θ is measured with respect to the [112] direction (reproduced from [20]). (b) 1-in 3-out (or 3-in 1-out) state in the [111] field. The spins denoted by B, C and D in the Kagomé plane then align along the [111], [1$\bar{1}$1] and [$\bar{1}$11] directions, respectively, so as to maximize the Zeeman energy gain. (c) 2-in 2-out state in the [112] field.

and dash–dotted lines show the calculated angular dependence of the saturation magnetization for 1-in 3-out (or 3-in 1-out) and 2-in 2-out states, respectively. Near the [112] direction, the measured magnetization $M_{\rm H}(\theta)$ agrees closely with the angular variation expected for the 2-in 2-out state. Considering that a strong field is applied along the [112] direction parallel to the ($\bar{1}\bar{1}$1) Kagomé plane, as shown in figures 2(b) and (c), the Kagomé and triangular planes are stacked alternately along the [$\bar{1}\bar{1}$1] direction. For the [112] direction, the spins denoted B, C and D in the Kagomé plane align along the [111], [1$\bar{1}$1] and [$\bar{1}$11] directions in the field of 2 T, respectively. On the other hand, for the [112] direction, the spins denoted by A in the triangular plane are decoupled from the field because its Ising axis is normal to the field. In this situation, an internal field $H_{\rm i}$ that occurs as a result of the nearest neighbour interaction with spins B $\sim$ D forces all of the spins on the A sublattice to point along [$\bar{1}\bar{1}$1] in order to maintain the 2-in 2-out state on each tetrahedron, as shown in figure 2(c).

When the magnetic field is slightly tilted from the [112] direction toward the [111] direction, a field component antiparallel to [$\bar{1}\bar{1}$1] appears and couples to spin A. At critical angle $\theta_{\rm c}$, this field component compensates the internal field $H_{\rm i}$ acting on spin A. A flipping of the spins on the A sublattice toward the [11$\bar{1}$] direction occurs as the field is further tilted across $\theta_{\rm c}$. In figure 2(a), we have observed that $M_H(\theta)$ exhibits an abrupt jump to the value expected for the 1-in 3-out state at $\theta = \theta_{\rm c} \sim -8^\circ$. This discontinuous change indicates that spin flip occurs in the triangular lattices. Note that this magnetization jump at $\theta_{\rm c}$ becomes shaper with decreasing temperature.

In figure 3(a), we show $M_{\rm t}$ as a function of $H_{\rm t}^*$ at various temperatures above 0.29 K, where $M_{\rm t}$ is the magnetization component along the [$\bar{1}\bar{1}$1] direction and $H_{\rm t}$ is the magnetic field component along the [$\bar{1}\bar{1}$1] direction, i.e., $H_{\rm t} = H \sin\theta$. Figure 3(b) shows the angular relation of the magnetization of the system. Near the [112] direction, $\boldsymbol{M}$ for the 2-in 2-out

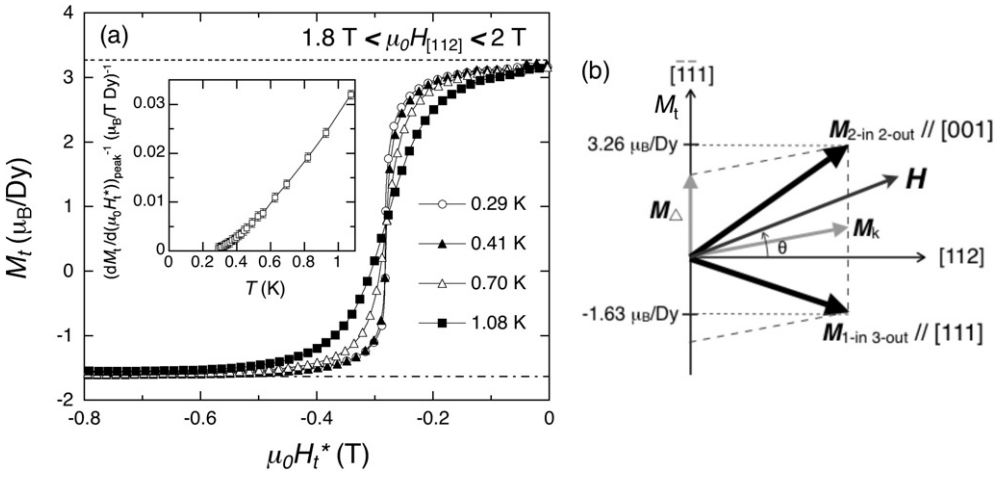

Figure 3. (a) Transverse magnetization M_t versus H_t^* for $Dy_2Ti_2O_7$ with a strong field ($\mu_0 H_{[112]} \sim$ 2 T) applied along the [112] direction. Demagnetization field correction has been performed for the transverse field H_t, as described in the text. The in-plane field component $H_{[112]}$ ($=H\cos\theta$) is in the range 1.8–2.0 T in this plot. The broken and dash–dotted lines represent the calculated saturation magnetizations for the 2-in 2-out and 1-in 3-out spin states, respectively. The inset shows the reciprocal of the peak value $(dM_t/dH_t^*)_{peak}$ as a function of temperature, where $(dM_t/dH_t^*)_{peak}$ tends to diverge just below 0.29 K, indicating the existence of a phase transition (reproduced from [20]). (b) Definition of the magnetization and the magnetic field along the [$\bar{1}\bar{1}$1] direction, where $M_\triangle$ and M_k denote the magnetization of the triangular and Kagomé lattices, respectively.

state has a component along the [001] direction. Near the [111] direction, $\boldsymbol{M}$ for the 1-in 3-out state has a component along the [111] direction. Next, $\boldsymbol{M}$ can be expressed as the sum of the Kagomé lattice ($\boldsymbol{M}_k$) and the triangular lattice ($\boldsymbol{M}_\triangle$) magnetizations, as $\boldsymbol{M} = \boldsymbol{M}_k + \boldsymbol{M}_\triangle$. Remember that the measured magnetization $M_H(\theta)$ is a component of $\boldsymbol{M}$ parallel to $\boldsymbol{H}$. Thus, $M_H = \boldsymbol{M}\cdot(\boldsymbol{H}/H) = M_t \sin\theta + M_{[112]}\cos\theta$, where $M_{[112]}$ is the component of $\boldsymbol{M}$ along [112]. At low T in a sufficiently high field, $\boldsymbol{M}_k$ is almost saturated and fixed ($\|$[113]). Assuming that $M_{[112]}$ is independent of θ and $M_{[112]} = M_H(0)$, we obtain $M_t = (M_H(\theta) - M_H(0)\cos\theta)/\sin\theta$, where M_t can be expressed as a function of H_t. The broken and dash–dotted lines indicate the expected M_t for the 2-in 2-out (3.26 μ_B/Dy) and 1-in 3-out (-1.63 μ_B/Dy) states, respectively. This result clearly indicates a spin-flip from the 2-in 2-out state to the 1-in 3-out state at the critical field of -0.28 T. We obtained the internal field $\mu_0 H_i$ of 0.28 T from the Kagomé lattice. This value is in relatively good agreement with the estimate of 0.25 T by Higashinaka *et al* [19]. Next, M_t has a constant offset of 0.82 μ_B/Dy, which comes from the component of $\boldsymbol{M}_k$ along the [001] direction. Apart from the offsets in M_t and H_t^*, it should be noted that the behaviour of M_t closely resembles that of ferromagnetic systems.

The spin-flip becomes sharp on cooling. The differential susceptibility dM_t/dH_t^* has a sharp peak at the spin-flip field. The reciprocal of the peak value $(dM_t/dH_t^*)_{peak}$ as a function of T is shown in the inset of figure 3(a). Here, $(dM_t/dH_t^*)_{peak}$ tends to diverge at $T = 0.27 \pm 0.02$ K. Our estimation of the critical point T_c is in good agreement with that of Higashinaka *et al* [19]. Our results reveal that this phase transition is driven by the ferromagnetic ordering in the spins on the triangular lattice. This spin-flip becomes a first-order transition below T_c. It is important to compare the first-order transition of liquid–gas type for the [111] direction, which is driven by a spin-flip on the Kagomé lattice. The ice-rule breaking first-order transition in the [111] field lifts the macroscopic degeneracy on the Kagomé plane.

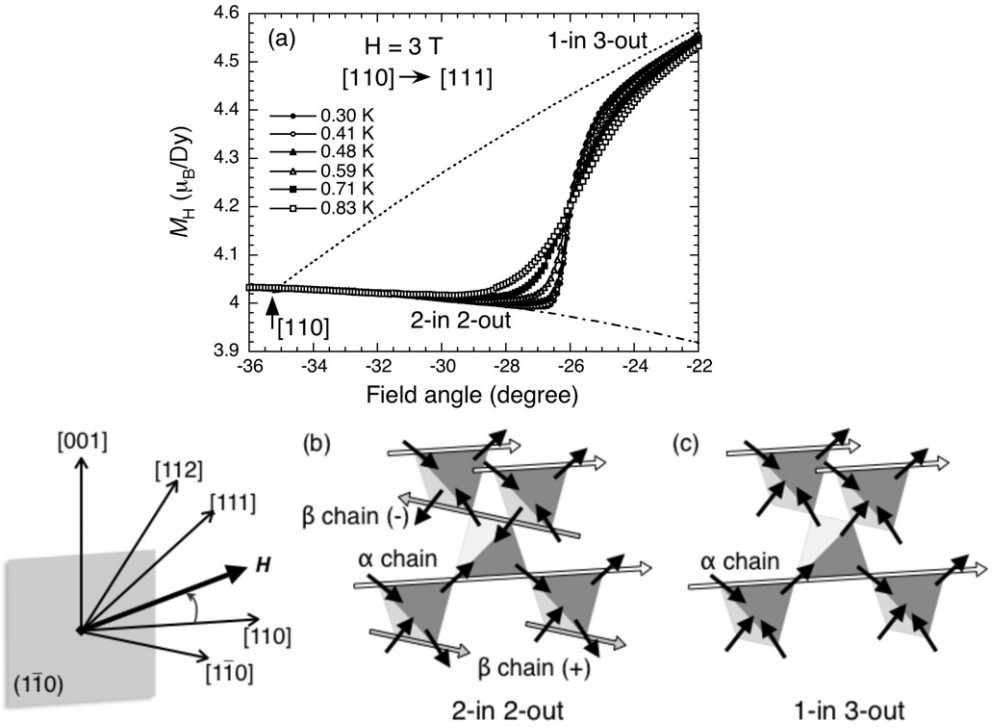

Figure 4. (a) Field-angle variation of the magnetization (M_H) for $Dy_2Ti_2O_7$ obtained at various temperatures in a field of 3 T rotated in the (1$\bar{1}$0) plane. The field direction is rotated from [110] to [111]. The broken and dashed–dotted lines represent the field-angle variation of the calculated saturation magnetizations for the 2-in 2-out and 1-in 3-out states, respectively. The field angle is measured with respect to the [111] direction. (b) 2-in 2-out state in the [110] direction. The spins denoted by the α chain have a component along [110]. The β chains behave as unique ferromagnetic Ising spin chains. The β chain (+) and β chain(−) have components along the [1$\bar{1}$0] and [$\bar{1}$10] directions, respectively. The spins on the β chain are decoupled from the [110] field. (c) 1-in 3-out state.

In contrast, this first-order spin-flip transition near the [112] direction is merely an inversion of the spins on the triangular lattice, and there is no macroscopic degeneracy in the 2-in 2-out state. Note that no entropy change is involved in the spin-flip transition.

Next, we focus on the discontinuous change of M_H between the [110] and [111] directions (see figure 1(a)). In figure 4(a), we show the field-angle variation of M_H obtained in a field of 3 T rotated from [110] to [111] in the (1$\bar{1}$0) plane. The broken and dash–dotted lines represent the field-angle variation of the calculated saturation magnetizations for 2-in 2-out and 1-in 3-out states, respectively. The field angle is measured with respect to the [111] direction. With the field applied along the [110] direction, the spin system is separated into parallel (α) and perpendicular (β) chains (figure 4(b)). The magnetic field along [110] easily aligns the spins on the α chain owing to the large Zeeman energy, while the spins on the β chain are decoupled from the field. However, according to the ice rule, the spins on the β chain are ferromagnetically aligned below 1.1 K $\sim J_{\rm eff}$. Note that there are still two possibilities for each β chain to align along the [1$\bar{1}$0] or [$\bar{1}$10] directions. These β chains weakly interact with each other by a long-range dipolar interaction. In the [110] field, since the magnetic moment from β chains is cancelled out, the magnetic moment from α chains has a component along [110]. Rotating the

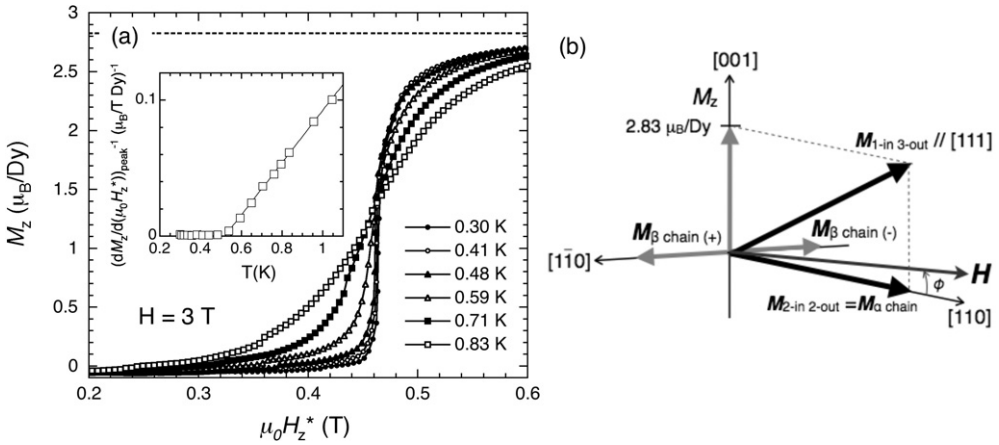

Figure 5. (a) M_z versus H_z^* for $Dy_2Ti_2O_7$ under a magnetic field of 3 T rotated from [110] to [111]. Here, M_z is the magnetization component along the [001] direction, and H_z^* denotes the magnetic field component along the [001] direction for which a demagnetization field correction is performed. The dotted lines represent the calculated M_z for the 1-in 3-out state, and M_z for the 2-in 2-out state is zero. The inset shows the reciprocal of the peak value $(dM_z/dH_z^*)_{peak}$ as a function of temperature, and $(dM_z/dH_z^*)_{peak}$ tends to diverge at $T_c = 0.52$ K. (b) For the [110] field direction, the magnetization for the 2-in 2-out state is equal to the magnetization for the α chain. The magnetizations for the β chains (+) and the β chains (−) cancel each other out.

field from the [110] direction toward the [111] direction, the magnetic field component along the [001] direction H_z increases. When the gain of the Zeeman energy due to H_z becomes larger than $\sim J_{\text{eff}}$, the ferromagnetic ordering in β chains finally collapses (figure 4(c)). The half-spins on the β chain (one spin in the tetrahedron) with an antiparallel component to the H_Z show a spin-flip. The magnetization component along the [001] direction M_z is induced by the breakdown of ferromagnetic β chains. Since a magnetic field of 3 T is rotated from [110] toward [111], the breakdown of ferromagnetic spin chains is observed at $-26°$, and in a field of 2 T, it is observed at $-19.6°$ (see figure 1(a)). It should be noticed that the magnetization jump at $-26°$ becomes sharper with decreasing temperature. A new phase transition at finite temperature is suggested.

In order to clearly observe the spin-flip behaviour, M_z versus H_z^* is plotted in figure 5(a) for various temperatures above 0.30 K. Figure 5(b) shows the angular relation of the magnetization of the system. In a similar manner to that described for the case of the near [112] direction, we obtain $M_H = \boldsymbol{M} \cdot (\boldsymbol{H}/H) = M_z \sin\phi + M_{[110]} \cos\phi$, where $M_{[110]}$ is the component of $\boldsymbol{M}$ along the [110] direction and ϕ is the angle between $\boldsymbol{H}$ and the [110] direction. Assuming that the magnetic moment of the α chains is independent of ϕ and $M_{[110]} = M_H(\phi = 0)$, we obtain $M_z = (M_H(\phi) - M_H(\phi = 0)\cos\phi)/\sin\phi$. Here, M_z can be expressed as a function of H_z. Near the [110] direction, M_z for the 2-in 2-out state is zero, and M_z for the 1-in 3-out state is expected to be 2.83 μ_B/Dy, as shown by the dotted line. The behaviour of M_z clearly shows the spin-flip from the 2-in 2-out state for $\mu_0 H_z^* < 0.4$ T to the 1-in 3-out state for $\mu_0 H_z^* > 0.55$ T. The critical field of the spin-flip H_c is obtained as 0.46 T. Calculating H_c on the basis of the nearest-neighbour spin ice model with $J_{\text{eff}} = 1.1$ K i.e. that the spins undergoing the flip do not interact with each other, H_c is estimated to be 0.57 T. The calculated value is slightly larger, suggesting a long-range interaction. The spin-flip becomes sharper upon cooling, and the differential susceptibility dM_z/dH_z^* results in a sharp peak at H_c. The inset of figure 5(a) shows the reciprocal of the peak value $(dM_z/dH_z^*)_{peak}$ plotted as a function of T. As shown in

the inset, $(\mathrm{d}M_z/\mathrm{d}H_z^*)_{\mathrm{peak}}$ tends to clearly diverge at a finite temperature $T \sim 0.5$ K, which is the critical point T_{cr}. A simple linear extrapolation of the plot gives the onset of the transition as $T_{\mathrm{cr}} \sim 0.52$ K.

Next, we will discuss this spin-flip transition between the [110] and [111] directions. First, it is important to understand the arrangement of spins undergoing the flip. In the [110] field, the experimental results show that the ferromagnetic β chains have a long but finite intrachain correlation length and a short interchain correlation, although the β chains theoretically show antiferromagnetic ordering ($Q = X$ structure) [12, 13]. These chains are interpreted as being arranged in a triangular array, which frustrates the coupling between them. More precisely, the triangular arrangement is not perfectly equilateral [11]. When the applied field is rotated slightly from the [110] direction toward the [111] direction, a ferromagnetic α chain remains to be formed. Consequently, the in-out state on the β chain in the (111) Kagomé plane is selected in accordance with the ice rule. The spins undergoing the flip only exist on the β chains in the (111) Kagomé plane (see figure 4(b)), and the spins on the (111) triangular plane are aligned along the [111] direction and lie on the α chain. Therefore, the arrangement of spins undergoing the flip is disordered (or short-ranged ordered) and has no translational symmetry.

Note that this spin-flip transition between the [110] and [111] directions is similar to the ice rule breaking the first-order transition of a liquid–gas type for the [111] direction. In the Kagomé ice state in the [111] field, the spins undergoing the flip exist in the Kagomé planes without translational symmetry in accordance with the ice rule [17]. We speculate that the transition between the [110] and [111] directions continuously connects to the transition in the [111] field from the field-angle dependence of H_{c} [21]. However, the macroscopic degeneracy in Kagomé ice is removed when the applied field is rotated from the [111] direction toward the [110] direction. In a field near the [110] direction, it is expected that the macroscopic degeneracy direction almost disappears because the intrachain correlation length on the β chain becomes larger in the [110] field. It is speculated that a slight entropy change is involved in the spin-flip transition. Further experiments and theoretical research are needed in order to clarify this spin-flip transition between the [110] and [111] directions.

In summary, we have measured the angle variation of the magnetization in the $(1\bar{1}0)$ plane by means of angle-resolved DC magnetization measurements in order to study the field-induced phase transition in the pyrochlore spin ice compound $Dy_2Ti_2O_7$. Two phase transitions caused by ice-rule breaking spin-flip are observed in a field of 2 or 3 T. One of these is a ferromagnetic ordering in the triangular lattice near the [112] direction, $T_{\mathrm{c}} = 0.27$ K at 2 T, as predicted theoretically, and the other is a first-order phase transition related to the breakdown of ferromagnetic Ising spin chains along $[1\bar{1}0]$ or $[\bar{1}10]$ in the magnetic field direction between [110] and [111], $T_{\mathrm{cr}} = 0.52$ K at 3 T. The arrangement of spins undergoing the flip has no translational symmetry. This transition is expected to be a liquid–gas type, which is similar to the a liquid–gas-type transition in the [111] field. The present result reveals the origin of the peaks at ∼0.3 K and ∼0.5 K for fields higher than 1 T, which are observed in powder samples.

Acknowledgments

This study was supported in part by a Grant-in-Aid for Scientific Research (B) (No. 16340099) and Young Scientists (B) (No. 17740234) of MEXT in Japan. KM was supported by the Visiting Researcher's Program of the Institute for Solid State Physics, University of Tokyo, and HS was supported by MEXT of Japan through the 21st Century COE program 'Quantum Extreme Systems and Their Symmetries'.

References

[1] Subramanian M A, Aravamudan G and Subba Rao G V 1983 *Prog. Solid State Chem.* **15** 55
[2] Gaulin B D, Reimers J N, Mason T E, Greedan J E and Tun Z 1992 *Phys. Rev. Lett.* **69** 3244
[3] Gingras M J P, Stager C V, Raju N P, Gaulin B D and Greedan J E 1997 *Phys. Rev. Lett.* **78** 947
[4] Harris M J, Bramwell S T, McMorrow D F, Zeiske T and Godfrey K W 1997 *Phys. Rev. Lett.* **79** 2554
[5] Gardner J S, Dunsiger S R, Gaulin B D, Gingras M J P, Greedan J E, Kiefl R F, Lumsden M D, MacFarlane W A, Raju N P, Sonier J E, Swainson I and Tun Z 1999 *Phys. Rev. Lett.* **82** 1012
[6] Raju N P, Dion M, Gingras M J P, Mason T E and Greedan J E 1999 *Phys. Rev.* B **59** 14489
[7] Bramwell S T and Gingras M J P 2001 *Science* **294** 1495
[8] Ramirez A P, Hayashi A, Cava R J, Siddharthan R and Shastry B S 1999 *Nature* **399** 333
[9] Higashinaka R, Fukazawa H and Maeno Y 2003 *Phys. Rev.* B **68** 014415
[10] Fennell T, Petrenko O A, Balakrishnan G, Bramwell S T, Champion J D M, Fak B, Harris M J and Paul D M 2002 *Appl. Phys.* A **77** S889
[11] Hiroi Z, Matsuhira K and Ogata M 2003 *J. Phys. Soc. Japan* **72** 3045
[12] Fennell T, Petrenko O A, Fak B, Gardner J S, Bramwell S T and Ouladdiaf B 2005 *Phys. Rev.* B **72** 224411
[13] Yoshida S, Nemoto K and Wada K 2004 *J. Phys. Soc. Japan* **73** 1619
[14] Ruff J P C, Melko R G and Gingras M J P 2005 *Phys. Rev. Lett.* **95** 097202
[15] Matsuhira K, Hiroi Z, Tayama T, Takagi S and Sakakibara T 2002 *J. Phys.: Condens. Matter* **14** L559
[16] Hiroi Z, Matsuhira K, Takagi S, Tayama T and Sakakibara T 2003 *J. Phys. Soc. Japan* **72** 411
[17] Sakakibara T, Tayama T, Hiroi Z, Matsuhira K and Takagi S 2004 *Phys. Rev. Lett.* **90** 207205
[18] Aoki H, Sakakibara T, Matsuhira K and Hiroi Z 2004 *J. Phys. Soc. Japan* **73** 2851
[19] Higashinaka R and Maeno Y 2005 *Phys. Rev. Lett.* **95** 237208
[20] Sato H, Matsuhira K, Tayama T, Hiroi Z, Takagi S and Sakakibara T 2006 *J. Phys.: Condens. Matter* **18** L297
[21] Sato H, Matsuhira K, Sakakibara T, Tayama T, Hiroi Z and Takagi S 2006 *Proc. Highly Frustrated Magnetism 2006* at press