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# Extremely broad hysteresis in the magnetization process of $\alpha$-$Dy_2S_3$ single crystal induced by high field cooling

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**Abstract**. $\alpha$-$Dy_2S_3$ possesses orthorhombic crystal structure having two crystallograpically inequivalent Dy sites. Magnetization process of $\alpha$-$Dy_2S_3$ single crystal after cooling in the high magnetic field of 18 T has been investigated. The magnetization under the field of 18 T along the *a*-axis on the cooling process from 150 K shows step-like rises at 70 and 40 K and reaches about 9 $\mu_B$ per one $Dy^{3+}$ at 1.5 K. This value, which corresponds to 90 % of full saturation moment of $Dy^{3+}$, is much larger than 6 $\mu_B$ obtained at the same conditions after cooling in no magnetic field (zero-field cooling; ZFC). After cooling to 1.5 K, the magnetization while decreasing field shows abrupt drops at 3.0 and 1.7 T, and then comes to 0 $\mu_B$ at 0 T. Subsequently, while increasing field, the magnetization demonstrates a similar curve to that obtained after ZFC without step-like rise below 13.1 T. At $\mu_0 H$ = 13.1 T, the magnetization rises suddenly and agrees with the curve for the decreasing process. This irreversible magnetization process yields extremely broad hysteresis having width of $\mu_0 \Delta H$ = 11.4 T. Broader hysteresis and narrower one are also observed at 4.2 and 10 K, respectively.

## 1. Introduction

The compound $\alpha$-$Dy_2S_3$ has the orthorhombic crystal structure shown in figure 1, which is one type of four structural modifications for dysprosium sesquisulfide [1-3]. There are two crystallographically inequivalent Dy sites labeled Dy1 and Dy2 in this structure. The Dy1 atoms form planes with buckling in the *ab*-plane. The Dy2 atoms are connected to these planes. The decorated planes are stacked along the *c*-axis. Novel physical properties in the isostructural compounds $\alpha$-$R_2S_3$ ($R$ = Sm, Gd, Tb and Dy) have been investigated within recent years [4-13]. In particular, magnetic and electrical properties in $\alpha$-$Dy_2S_3$ are fascinating [10-12]. Successive magnetic transitions occurs at $T_{N_1}$ = 11.5 K and $T_{N_2}$ = 6.4 K. The behavior of the magnetization in the vicinities of $T_{N_1}$ and $T_{N_2}$ is extremely anisotropic [10] and the electrical resistivity shows anomalous enlargement between $T_{N_1}$ and $T_{N_2}$ [11]. Recently, clear peaks in the temperature dependence of the specific heat at both transition temperatures have also been observed, which will be reported elsewhere. The magnetization process under the magnetic field up to 18 T after cooling down to 1.5 K in no magnetic field (zero-field cooling; ZFC) is also anisotropic, and the magnetization along the *a*-axis is smallest among those along three crystal axes at 18 T [12]. The value per one Dy atom is small as 6 $\mu_B$, which corresponds to 60 % of full saturation moment of

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$Dy^{3+}$. In this paper, we report extremely broad hysteresis in the magnetization process of $\alpha$-$Dy_2S_3$ single crystal induced by cooling in the high magnetic field of 18 T.

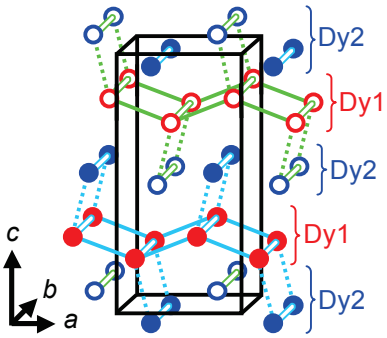


**Figure 1.** Crystal structure of $\alpha$-$Dy_2S_3$, in which S atoms are omitted for clarity. Two crystallographically inequivalent Dy sites are shown as Dy1 and Dy2. Each site atoms are stacked alternately along the *c*-axis. The open circles represent Dy atoms on (0, 1/4, 0) plane and the filled circles are Dy atoms on (0, 3/4, 0) plane.

## 2. Experimental

The single crystals of $\alpha$-$Dy_2S_3$ were grown by a chemical transport reaction method from the powder sample prepared by sulfurizing method with iodine as a carrier [4]. Identification of the powder sample and determination of the crystal orientation were made by X-ray (Cu $K_\alpha$-radiation) diffraction measurements. Magnetization was measured in the magnetic-field range of 0-18 T by using extraction-type magnetometer [14] and a superconducting magnet. The needle-shaped single crystal having a 3.5-mm length along the *b*-axis, an around 0.3-mm length of the hexagonal cross section in the *ac*-plane and a mass of 3.4 mg was mounted with epoxy resin for the measurement.

## 3. Results and discussion

The temperature dependence of the magnetization while cooling in the magnetic field of 18 T along the *a*-axis (experiment 1; abbreviated as exp. 1 here) is shown in figure 2. The magnetic field was applied when the temperature of the sample was 150 K. The solid curve shows the magnetization of a paramagnetic $Dy^{3+}$ ion in the magnetic field of 18 T calculated as a function of temperature by

$$M_{\mathrm{cal}} = g_J \mu_{\mathrm{B}} J B_J\left(\frac{g_J \mu_{\mathrm{B}} JH}{k_{\mathrm{B}} T}\right), \tag{1}$$

where $\mu_{\mathrm{B}}$ is the Bohr magneton, $k_{\mathrm{B}}$ is the Boltzmann constant and $B_J$ is the Brillouin function. The Landé *g*-factor; $g_J = 4/3$ and the total angular momentum $J = 15/2$ for $Dy^{3+}$ ion are used. The cernox™ thermometer used has not been calibrated in magnetic field, thus the temperature below 10 K has errors within 1 %. However, the experimental values of the magnetization at these temperatures are almost constant, therefore it does not affect qualitative consideration. The magnetization rises

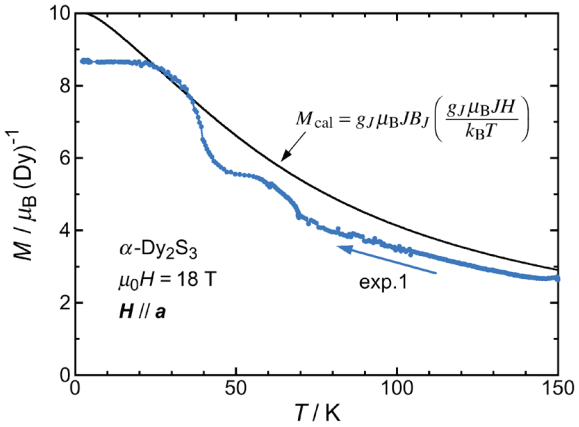


**Figure 2.** Temperature dependence of the magnetization per one Dy atom on the cooling process in the field of 18 T along the *a*-axis of $\alpha$-$Dy_2S_3$ single crystal. The calculated $M(T)$ curve for free $Dy^{3+}$ ion is also shown.

gradually with decreasing temperature from 150 K, and the curve is lower than the calculated one. The magnetization shows relatively steep rises at around 70 and 40 K, and subsequently increases up to almost the same value to the calculated one below 40 K. Then, below 20 K, it shows almost constant value of about 8.7 $\mu_B$. This value is fairly larger than the value of 6 $\mu_B$ obtained at 1.5 K after ZFC. Moreover, it is larger than the *b*-axis magnetization of 7.5 $\mu_B$ at 1.5 K, which is highest value among those for three crystal axes [12].

After exp. 1 the magnetization was measured while decreasing magnetic field (exp. 2) and then while increasing field (exp. 3) at the same temperature of 1.5 K. Then the sample was suffered thermal cycle of 1.5-200 K and field cycle of 0-18 T randomly. Subsequently, the decreasing process from 18 T (exp. 4) and increasing process from 0 T (exp. 5) at 1.5 K were repeated. These isothermal magnetization curves are shown in figure 3(a). Although some discrepancies in magnitude between the curves of the same-type process and scattering of the data are seen, reproducible features are also observed clearly. The decreasing processes (exp. 2, 4) show two-step abrupt drops at around 3.1 and 1.7 T. Then the *M* decreases down to 0 $\mu_B$ at 0 T without remanent magnetization. On the other hand, the increasing processes (exp. 3, 5) show no anomalies at 1.7 and 3.1 T. The curves rather resemble that of ZFC curve reported previously [12] until 13.1 T; however, they show sudden steep rises at 13.1 T and overlaps with the curves of the decreasing process. This irreversible magnetization process yields extremely broad hysteresis having width of $\mu_0\Delta H$ = 11.4 T.

The isothermal magnetization at 4.2 K measured after exp. 5 is shown in figure 3(b). The increasing process (exp. 6) shows step-like rise clearly at 13.4 T and the following decreasing process (exp. 7) demonstrates two-step drops at 3.4 and 1.6 T. Broader hysteresis having width of $\mu_0\Delta H$ = 11.8 T than that for 1.5 K is confirmed. This strange broadening of the hysteresis with increasing temperature might be brought about by complicated competition of magnetic interactions between both Dy sites. The isotherms of exp. 2-7 were measured at the temperatures below $T_{N2}$ = 6.4 K determined from the $M(T)$ in lower magnetic field [10]. The isotherms at 10 K between $T_{N1}$ and $T_{N2}$ measured after exp. 7 are shown in figure 3(c). A clear step-like rise in the increasing process (exp. 8)

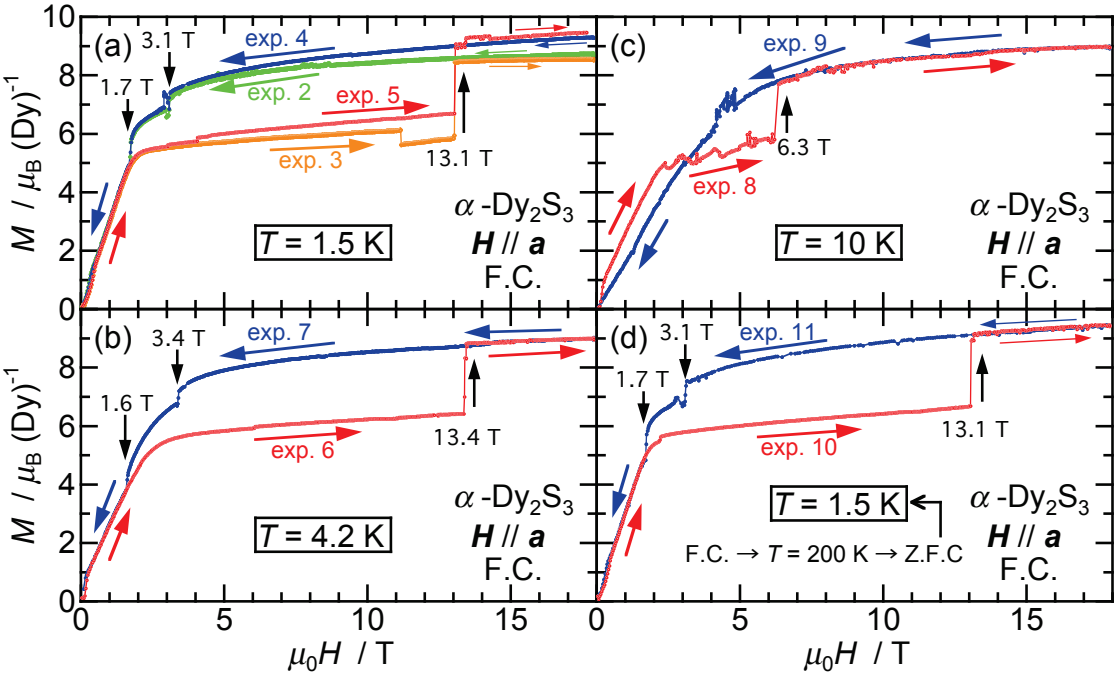


**Figure 3.** Magnetization curves along the *a*-axis of $\alpha$-$Dy_2S_3$ single crystal at some temperatures. The curves in the figures of (a)-(c) are obtained after field cooling shown in Fig. 2. Before the exp. 10 shown in the figure (d) was performed, the sample had been warmed up to 200 K and then cooled down to 1.5 K in no magnetic field.

at 6.3 T is observed; however, no clear drops are observed in the decreasing process (exp. 9). Although suspicion remains because of scattering data, the curve of exp. 8 below 3.2 T exists over the curve of exp. 9. A hysteresis with width of about 3 T is observed also in this case. After the process of exp. 9, the sample suffered heating up to 200 K followed by cooling down to 1.5 K without applying magnetic field. The magnetization process of increasing (exp. 10) and decreasing (exp. 11) at 1.5 K reproduces broad hysteresis as shown in figure 3(d).

The sample was warmed up to room temperature in no magnetic field, and then the anisotropy of the high-field cooling effect from 150 K was investigated. The *c*-axis magnetization measured after the experiment for the *a*-axis direction did not show such effect mentioned above. As for the magnetization process of *b*-axis direction, the decreasing curve from 18 T just after cooling to 1.5 K showed a small drop at 4.4 T and the increasing process showed a slight lower curve at the magnetic field range higher than 4.4 T; however, the curve indicated no step-like rise until 18 T.

Cooling in the magnetic field of 18 T along the *a*-axis of $\alpha$-$Dy_2S_3$ single crystal induced larger magnetization than that of ZFC. The magnetization around 9 $\mu_B$ indicates that 90 % component of $Dy^{3+}$ moments was aligned along the *a*-axis forcibly by the magnetic field. Magnetic structure stabilized after decreasing field must be different from that in the case of ZFC. The hysteresis in the $M(H)$ induced by high field cooling suggests the existence of multiple metastable magnetic structures and complicated magnetic interactions between two crystallographic Dy sites. It is considered that the drastic changes of magnetization [10] originate from change of magnetic structure.

## 4. Summary

We have found out extremely broad hysteresis in the $M(H)$ of $\alpha$-$Dy_2S_3$ single crystal induced by cooling in high magnetic field. It suggests multiple metastable magnetic structures exist in $\alpha$-$Dy_2S_3$. It is considered that novel physical properties in $\alpha$-$Dy_2S_3$ are brought about by transitions among metastable magnetic phases.

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