Angular-field magnetic phase diagram of b-plane at 4 K of YAlGe-type TbAlGe with zigzag-chain

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Abstract. Orthorhombic YAlGe-type TbAlGe is expected to have an interesting magnetic anisotropy due to zigzag chains of the Tb ions. We have grown the single crystal for the first time and measured the AC magnetic susceptibility and specific heat from 1.3 K to 60 K, and the vector magnetization for the b-plane up to 7 T at 4 K. The specific heat and AC magnetic susceptibility indicate that there are two antiferromagnetic transitions at $T_{N1} = 38$ K and $T_{N2} = 7.6$ K, where the transition at $T_{N2}$ is first-order like. The magnetization curve at 4 K for the a-axis shows a large hysteresis, and metamagnetic transition appears at $H_1 = 1.6$ T in the field increasing process, and another metamagnetic transition at $H_2 = 3.5$ T in addition to $H_1$ in the decreasing field process. The magnetization curves of the b- and c-axis are linear up to 7 T. The measurement of vector magnetization at 4 K reflects the hysteresis of the magnetization curve, and there is a large hysteresis. From this vector magnetization measurement, we have made the angular magnetic field phase diagram at 4 K for the b-plane. In this phase diagram, there are phase lines that cannot be obtained by ordinary magnetization measurement.

1. Introduction
RAIGe ($R$ = rare earth) crystallizes in the tetragonal α-ThSi$_2$ type in light rare earths and the orthorhombic YAlGe type in heavy rare earths. The boundary is $R = $ Gd, where the high temperature phase is α-ThSi$_2$type, and the low temperature phase is YAlGe type [?]. These two structures are related each other that they have a triangular prism with Ge in the body. The α-ThSi$_2$ type is a triangular prism with [R6] as the vertices, and the YAlGe type is a triangular prism with [R2Al4] as the vertices. One feature of the YAlGe type structure is that the $R$ ions have a zigzag-chain in the a-plane and extend in the c-axis. Zigzag-chain of $R$ ions is also found in YFb$_2$Al$_{10}$ type structures and is studied by Ohno et al. [?]. They found that TbRu$_2$Al$_{10}$ has a very peculiar magnetic phase diagram that depends on the angle, and therefore TbAlGe is also expected to be interesting. However, the existence of YAlGe-type RAIGe has only been clarified only as a polycrystal, and its physical properties have not been measured. The purpose of this study is to grow a single crystal of TbAlGe, and measure the vector magnetization.

2. Experiment procedure
Single crystal growth of TbAlGe was carried out by the Al self-flux method. The growth was carried out in a Tanman crucible enclosed in a quartz tube with an argon atmosphere. The starting composition ratio was Tb:Al:Ge = 1:1:10. The quartz tube was heated to 1050°C, maintained for 8 hours, and then cooled to 700 °C at rate of 3.5 °C/h. The Al fulx was removed
by centrifugation at the temperature, the remaining flux was removed by etching with 3M NaOH aqueous solution. The resulting single crystal axes were determined by an XRD.

The AC magnetic susceptibility was measured by the usual Hartshorn bridge method, and the specific heat was measured by the adiabatic method using a GM refrigerator [?], both at 1.4K to 60K. The vector magnetization was measured at 4K for the b-plane using our hand-made syste [?].

3. Results and Discussion

Figure 1 shows the temperature dependence of the AC magnetic susceptibility $\chi$. Two cusps corresponding to the antiferromagnetic transition are found at $T_{N1}(\chi) = 41.5$ K and $T_{N2}(\chi) = 8.2$ K. Furthermore, $\chi$ increases below 4 K, suggesting that there is a further phase transition at low temperatures. Figure 2 shows the temperature dependence of the specific heat $C$ divided by temperature $T$. Anomalies are seen corresponding to the two anomalies in $\chi$, but the temperatures are slightly different, $T_{N1}(C) = 39$ K and $T_{N2}(C) = 7.6$ K. The phase transition at $T_{N2}(C)$ seems to be divergent, suggesting a first-order transition. In addition, although there is a slight upturn below 1.4 K that seems to correspond to the increase in $\chi$. Lower temperature measurements are needed.

Figure 3 shows the magnetic field ($H$)-dependence of the magnetization parallel to H direction ($M_x$) at 4 K along the $a$- and $c$-axis. On the $a$-axis, there are metamagnetic transitions (MT) with large hysteresis. The MT is observed at $H_1 = 1.7$ T in the $H$-up process, and at $H_2 = 3.5$ T in addition to $H_1$ in the $H$ down process. At $H > 6$ T, the magnetization appears to be saturated, but the magnetic moment is smaller than the theoretical value of Tb (9.0 $\mu_B$). Therefore, there may be another MT at higher fields. The $c$-axis magnetization curve is linear without hysteresis.

Figures 3 (b) and 3 (c) show the magnetization curves when $H$ is applied at an angle formed by the H-direction with the $a$-axis, divided into $H$-up process and $H$-down process, respectively. In Fig. 3 (c), there is a bend near 4 T at 50$^\circ$, while it disappears at 60$^\circ$. Comparing the $H$-up process and the $H$-down process, the hysteresis disappears from 60$^\circ$. The MT at $H = 1.7$ T becomes broad in the $H$-down process.

We have measured the angle dependence of $M_x$ and $M_y$ (the perpendicular magnetization to $H$-direction) of clockwise (cw) and counterclockwise (ccw) rotation at intervals of 0.5 T up to 6.5 T at 4 K. Figure 4 (a) shows the results at 6.5 T, where black and red points are cw and ccw, respectively. $\theta = 0^\circ$ and 90$^\circ$ correspond to $a$- and $c$-axis, respectively. Since the peculiar behavior of cw and ccw is not clear in $M_x$, we pay attention to $M_y$. We can see that there is a large hysteresis of angular dependence. It can also be seen that the period of magnetization is 180$^\circ$. There is also an interesting magnetization behavior at 0$^\circ$ and 360$^\circ$. These angles are on the same $a$-axis, but the magnetization is not the same. This is due to the rotation hysteresis, and depends on the start angle. In Fig. 4 (a), there is no peak at 10$^\circ$ that appears at 190$^\circ$, because the measurement started at 0$^\circ$. However, when we start the measurement from −20$^\circ$, we can see this peak at 10$^\circ$. Figures 4 (b) and (c) show $M_x$ and $M_y$ in cw each 0.5 T from 0$^\circ$ to

![Figure 1. The AC magnetic susceptibility of TbAlGe along the a-axis.](image1)

![Figure 2. The C/T vs T plot of TbAlGe.](image2)
Figure 3. Magnetic field \( (H) \) dependence of \( M_x \) of TbAlGe at \( T = 4 \) K (a) \( H_{\text{up}} \) and \( H_{\text{down}} \) process of \( a \)-axis and \( c \)-axis (b) \( H_{\text{up}} \) process every 10° (c) \( H_{\text{down}} \) process each 10°.

Figure 4. Angular dependence of magnetization for TbAlGe at \( T = 4 \) K. (a) \( b \)-plane, \( H = 6.5 \) T. The abbreviation of cw and ccw indicate clockwise and counterclockwise rotation, respectively. Angular dependence of (b) \( M_x \) and (c) \( M_y \) for several fields.

At high fields, it is difficult to see the change in \( M_x \), but if we focus on \( M_y \), we can see the angle at which the \( M_y \) changes abruptly (10°, 70°, 100°, 190°, 250°, 280° for cw).

Figure 5 shows a magnetic phase diagram obtained in this study for the angle and field at 4 K for \( b \)-plane in polar coordinate, where the angular rotation is the cw rotation. The blue and red circles are the anomalies that represent the obtained from the magnetization curve (Fig. 3) and the angular rotation (Fig. 4), respectively. For convenience of explanation, the phase lines are shown in the alphabet in the figure.

First, we consider the phase diagram from 0° to 90°. Since this corresponds to the \( H_{\text{down}} \) process of the magnetization curve, two MT appear on the \( a \)-axis (0°). The phase line (A) from \( H_1 = 2.0 \) T and the phase line (B) from \( H_2 = 3.5 \) T extend along the \( c \)-axis. The phase line from A extends in the circumferential direction and disappears at 60°. The anomaly is also seen...
in angular rotation, where the phase line of the magnetization curve extends further along the c-axis from where it disappears (D), but disappears at 4.0 T. Although the phase line B extends along the circumference and disappears at 40°, the anomaly of angular rotation connects from the phase line of B and extends along the c-axis (E). The phase line of F is found only in angular rotation and appears in a higher magnetic field than the phase line of A.

Next, we consider the phase diagram from 90° to 180°. This rotation corresponds to the $H$-up process and there is only one MT. The phase line (C) from the MT at $H_1 = 1.7$ T on the a-axis (180°) disappears at 50°, but continues to the anomalies in the angular rotation, and the phase line is along the c-axis (G).

Finally, let us consider the relationship between the magnetic moment $\mu$ as vector and the phases. Since we measured both $M_x$ and $M_y$, the direction and magnitude of $\mu$ at each point can be evaluated. The $\mu(H, \theta)$ is represented by an arrow, and the starting point is the coordinates $(H, \theta)$. The arrow length is drawn exactly in proportion to the magnitude of $\mu$. The $\mu$ increases strongly across the phase line A obtained by the magnetization curve. This reflects the MT. When the phase line of angular rotation is straddled in the angular direction, the $\mu$ direction changes. The phase line of E appears to continue from the position where the phase line disappeared on the magnetization curve. In the result of the magnetization curve, the hysteresis disappears at 60°, but with angular rotation, there is a hysteresis up to 70°. This result is a hysteresis that appears depending on the angle of rotation and is not expected from the hysteresis seen in the magnetization curve. Similarly, Similarly, it is difficult to infer the phase line F independent on fields from the magnetization curve.

4. Summary
We have measured the field and angular dependence of parallel and perpendicular magnetization for b-plane of TbAlGe at 4 K under fields up to 7 T, and obtained a field and angular magnetic phase diagram. As a result, the magnetic anomaly such as MT and hysteresis was observed in the a-axis direction, and the behavior of magnetization changed depending on the direction of rotation, and a hysteresis loop with a period of 180° was observed in $M_y$. The a-axis, in which MTs are clearly seen, is the direction perpendicular to the zigzag chain, which is very similar to TbFe$_2$Al$_{10}$, though there is no hysteresis in TbFe$_2$Al$_{10}$ [?]. Therefore, from an experimental point of view, this strange magnetic phase diagram and the zigzag structure are considered to be related.

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