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# Zero field magnetic phase transitions and anomalous low temperature upturn in resistivity of single crystalline $\alpha$ -TmAlB<sub>4</sub>

## Abstract

In this study, pure  $\alpha$ -TmAlB<sub>4</sub> (YCrB<sub>4</sub> structure) single crystals were grown with no  $\beta$ -TmAlB<sub>4</sub> (ThMoB<sub>4</sub> structure) intergrowth, and zero magnetic field transitions were confirmed through specific heat capacity, magnetization, and electric resistivity measurements. The anomalous magnetic transition was found at approximately 6.2 K with long range antiferromagnetic transition at 5.6 K. The difference in field dependence between these two transitions indicates that they do not share a common magnetic origin. In addition, we investigated electrical resistivity down to 20 mK, and found upturn behavior at around 0.8 K. The low temperature upturn anomaly in resistivity was not found for other compounds of investigation for RAlB<sub>4</sub> (R=rare earth elements), which suggests that an  $\alpha$ -RAlB<sub>4</sub> system has a significantly different ground state, compared to a  $\beta$ -RAlB<sub>4</sub> system.

## Keywords

Physics and Astronomy, Crystal structure, Electrical resistivity, Single crystals, Antiferromagnetism, Curie point

## Disciplines

Condensed Matter Physics

## Comments

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## Zero field magnetic phase transitions and anomalous low temperature upturn in resistivity of single crystalline $\alpha$ -TmAlB<sub>4</sub>

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In this study, pure  $\alpha$ -TmAlB<sub>4</sub> (YCrB<sub>4</sub> structure) single crystals were grown with no  $\beta$ -TmAlB<sub>4</sub> (ThMoB<sub>4</sub> structure) intergrowth, and zero magnetic field transitions were confirmed through specific heat capacity, magnetization, and electric resistivity measurements. The anomalous magnetic transition was found at approximately 6.2 K with long range antiferromagnetic transition at 5.6 K. The difference in field dependence between these two transitions indicates that they do not share a common magnetic origin. In addition, we investigated electrical resistivity down to 20 mK, and found upturn behavior at around 0.8 K. The low temperature upturn anomaly in resistivity was not found for other compounds of investigation for RAlB<sub>4</sub> (R=rare earth elements), which suggests that an  $\alpha$ -RAlB<sub>4</sub> system has a significantly different ground state, compared to a  $\beta$ -RAlB<sub>4</sub> system.

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The RAlB<sub>4</sub> compounds (R=rare earth elements) have YCrB<sub>4</sub> structured type orthorhombic lattice with a space group *Pbam*. RAlB<sub>4</sub> became known after Yb<sub>2</sub>AlB<sub>6</sub> and RAlB<sub>14</sub> and was discovered as the third group of ternary rare-earth aluminum borides.<sup>1,2</sup> Recently, two phases in the structure of the LnAlB<sub>4</sub> (Ln=Yb,Lu) single crystal were reported: the YCrB<sub>4</sub> structure type in a space group *Pbam* ( $\alpha$ -LnAlB<sub>4</sub>, needlelike morphology), and the ThMoB<sub>4</sub> structure type in a space group *Cmmm* ( $\beta$ -LnAlB<sub>4</sub>, platelike morphology). Additionally, each structure phase has a distinct physical ground state.  $\beta$ -YbAlB<sub>4</sub> showed superconductivity near a quantum critical point and heavy fermionic behavior, while  $\alpha$ -YbAlB<sub>4</sub> showed no such peculiarities.<sup>3,4</sup> Furthermore, multiple antiferromagnetic transitions in  $\beta$ -ErAlB<sub>4</sub> polycrystalline have been reported as the first observation in the  $\beta$ -RAlB<sub>4</sub> compounds.<sup>5</sup> Therefore, investigation of the physical properties of LnAlB<sub>4</sub> and identifying the structure type is necessary in order to study the intrinsic properties.

Among the RAlB<sub>4</sub> compounds, TmAlB<sub>4</sub> is of interest because of its magnetic properties. TmAlB<sub>4</sub> is an antiferromagnetic ordering compound under  $T_N=5.8$  K (Ref. 6) with seven magnetic phases due to complicated multiple magnetic transitions, and has a large magnetic anisotropy with a magnetic easy axis along the *c*-axis.<sup>7</sup> In general, it is difficult to grow single crystals of one type of structure because of similar forming energies between two crystal structures. For example, magnetic anomalies of TmAlB<sub>4</sub> at low temperatures were reported in Ref. 8 with no identification of structure

type and, thus, were probably closely related to the mixture of two structure types. Moreover, the intergrowth of the  $\alpha$ -type phase was also observed in the  $\beta$ -type phase TmAlB<sub>4</sub> polycrystalline compound, while  $\beta$ -TmAlB<sub>4</sub> exhibited antiferromagnetic ordering under  $T_N=9.5$  K.<sup>9</sup>

In this study, we have grown single crystals of  $\alpha$ -TmAlB<sub>4</sub> and investigated the zero field magnetic phase transitions, specific heat capacity, and anomalous electrical property at low temperatures.

Needlelike and platelike large single crystals of TmAlB<sub>4</sub> were grown out of the aluminum flux method.<sup>7</sup> Mixture of Tm<sub>2</sub>O<sub>3</sub> powder (99.9%), crystalline boron powder (99.9%), and aluminum chips (99.999%) were prepared with a ratio of Tm<sub>2</sub>O<sub>3</sub>:B:Al=1:2:119.25 and were placed in an alumina crucible (batch A). Another assembly (batch B) with a ratio of Tm(chips, 99.95%): B(grains, 99.5%): Al(chips,

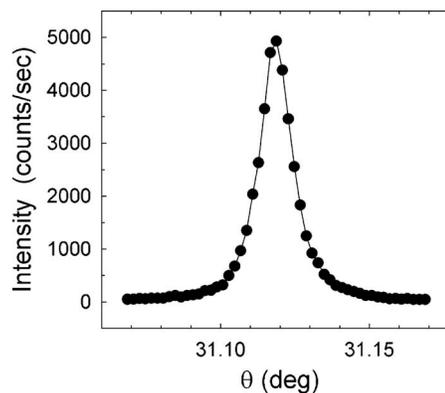


FIG. 1. Rocking scan through the (400) reflection, measured by x-ray diffraction on a needle-shaped crystal.

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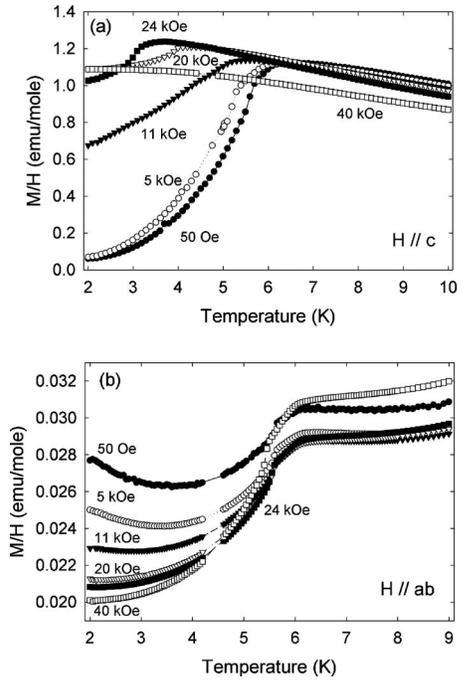


FIG. 2. Magnetization divided field ( $M/H$ ) in terms of temperature with various magnetic fields (a) parallel and (b) perpendicular to the  $c$ -axis.

99.999%)=1:4:393.2 was also prepared in an alumina crucible. The crucible for batch A was placed in a programmable tube furnace and heated under an Ar gas flow at 1450 °C for one day until a sufficient chemical reaction had been achieved, and then cooled to 900 °C at a rate of 2 °C/h. The crucible for batch B was slowly cooled from 1450 to 750 °C at a rate of 5 °C/h without dwelling. The aluminum flux was dissolved with NaOH in order to remove it from the crystals.

Two single crystals of needlelike and platelike shape from batch A were studied by x-ray diffraction to decide if the prepared material crystallizes in the alpha or the beta phase.<sup>8</sup> The Laue pattern showed two mirror planes rectangular to each other. The oriented crystals were mounted in a four circle diffractometer and studied by Cu  $K_{\alpha}$  x-ray radiation from a rotating anode source selected by a Ge (111) monochromator. The lattice parameters for the orthorhombic system were determined to  $a=5.922(1)$  Å,  $b=11.478(2)$  Å, and  $c=3.529(8)$  Å. Besides several other reflections of the (HK0) plane, the (120) reflection was observed with 1000

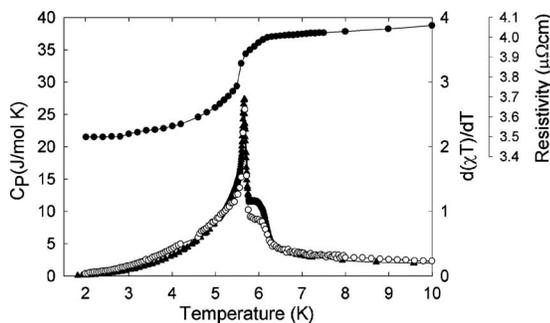


FIG. 3. Specific heat (closed triangles), temperature derivative of magnetization, divided by field, times temperature,  $\partial(\chi_c T)/\partial T$  (open circles), and resistivity (closed circles) as a function of temperature.

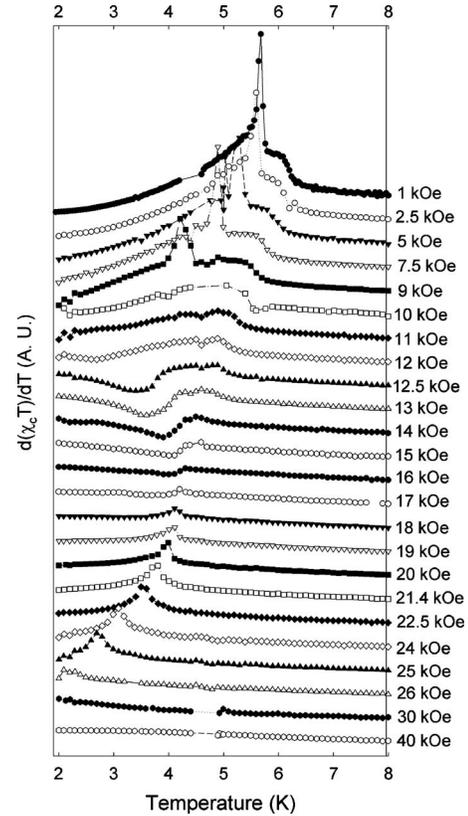


FIG. 4. Temperature derivatives of magnetization, divided by field, times temperature ( $\partial(\chi_c T)/\partial T$ ) with various magnetic field ( $H=1-40$  kOe) parallel to the  $c$ -axis.

cts/s. A reflection at this position is only consistent with the symmetry and corresponding reflection conditions for the alpha phase, it cannot appear for the beta phase. The determined lattice parameters are also consistent with the alpha phase only and, therefore, it can be concluded that both studied crystals belong to the alpha phase despite their different growth habits. The full width at half maximum of 0.013° and 0.018° in rocking scans through the (400) and (060) reflections, respectively, demonstrate the excellent mosaicity and high quality of the grown single crystals (Fig. 1).

Figure 2 shows the temperature-dependent magnetic susceptibility with various applied fields. As can be seen, the magnetic moment parallel to the  $c$ -axis is much larger than that perpendicular to the  $c$ -axis. Also, the antiferromagnetic transition is suppressed by an applied field along the  $c$ -axis, but the transition temperature remains mostly unchanged perpendicular to the  $c$ -axis in a field range of  $50 \text{ Oe} \leq H \leq 40 \text{ kOe}$ . These anisotropic behaviors are similar to previous results found in Ref. 8; however, there is no magnetic transition at approximately 3–4 K perpendicular to the  $c$ -axis, which was believed to be due to structural impurities in Ref. 8. Similar properties were also observed in the samples from batch B providing further evidence that the sample is in the alpha phase form only and has no beta-phase structure. We surmised that the difference in magnetic properties identified in this study and those in Ref. 8 originated from the crystal structures due to different growth conditions.

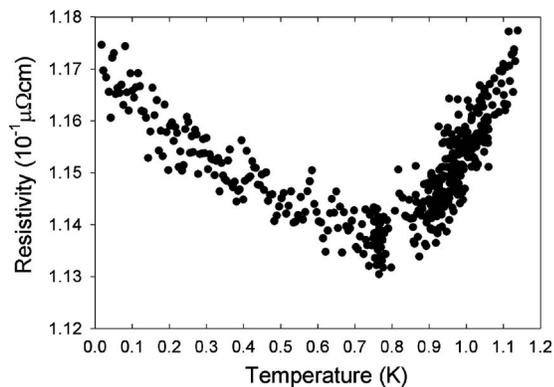


FIG. 5. Electrical resistivity as a function of temperature at low temperatures ( $20 \text{ mK} \leq T \leq 1.12 \text{ K}$ ) for an  $\alpha$ -TmAlB<sub>4</sub> single crystal.

In the electrical resistivity measurement, we observed a transition at  $T=5.6 \text{ K}$ , which corresponds to the antiferromagnetic transition temperature, and a linear slope change between  $5.6 \text{ K} \leq T \leq 6.2 \text{ K}$ , as shown in Fig. 3. Based on the heat capacity, we found a long range antiferromagnetic ordering at  $T=5.6 \text{ K}$  and an anomalous magnetic transition at  $T \approx 6.2 \text{ K}$  in the zero field. These two temperatures are nicely consistent with the temperatures of slope change in the resistivity data. The specific heat capacity of a LuAlB<sub>4</sub> single crystal was approximately  $0.03 \text{ J/mol K}$  up to  $10 \text{ K}$ . Therefore, the magnetic contribution to heat capacity is much larger than the lattice structural contribution as much as it can be ignored. The temperature dependent magnetization data, which are divided by the applied magnetic field of  $H=1 \text{ kOe}$ , was converted in  $d(\chi T)/dT$  [ $\chi=(\chi_{lc}+2\chi_{\perp c})/3$ ] to confirm the relation between the specific heat and magnetic susceptibility for an antiferromagnet<sup>10</sup> and is plotted in Fig. 3. As shown, the  $d(\chi T)/dT$  data match with the specific heat data, i.e., the sharp peaks at  $T=5.6 \text{ K}$  and the broad hump at  $T \approx 6.2 \text{ K}$ . Thus, the anomalous transition at  $T \approx 6.2 \text{ K}$ , as well as the antiferromagnetic transition, is likely due to magnetic interaction.

In order to investigate the field effects on the transition temperatures,  $d(\chi T)/dT$  along the  $c$ -axis with various fields was plotted in Fig. 4. The antiferromagnetic and the anomalous magnetic transitions shifted to lower temperatures and were suppressed by the applied magnetic field. The antiferromagnetic transition disappeared at an applied field of  $H=10 \text{ kOe}$ , while the anomalous magnetic transition was gradually suppressed by increasing the field, almost disappearing at  $H=17 \text{ kOe}$ , and appeared again with a further field increase until it disappeared again at  $H=30 \text{ kOe}$ . The origin of the anomalous magnetic transition is not obvious at present, but it is believed to be caused by a magnetic interaction different from that for long range antiferromagnetic ordering.

Temperature-dependent electrical resistivity was also measured in order to investigate the low temperature electrical property down to  $T=20 \text{ mK}$ , as shown in Fig. 5. An upturn anomaly was observed at approximately  $T=0.8 \text{ K}$  along the  $c$ -axis, as opposed to superconductivity which was observed in  $\beta$ -YbAlB<sub>4</sub>.<sup>4</sup> In general, the upturn in resistivity has several possible origins, including the Kondo effect, spin

fluctuation, localization of magnetic ions, or impurity scattering. At present, the origin in the  $\alpha$ -TmAlB<sub>4</sub> single crystal is still not fully understood; however, it can be assumed that the origin is not from the impurity scattering due to the high quality of the sample. To confirm the origin of the anomalous upturn in resistivity, further measurements are needed with an applied magnetic field. Although the origin of the anomalous upturn in resistivity is not clear, it is still an interesting behavior since this is the first observation for an  $\alpha$ -RAIB<sub>4</sub> system up to date. Therefore, the investigation of magnetic field-dependent and temperature-dependent electrical and magnetic properties of  $\alpha$ -RAIB<sub>4</sub> at low temperature is important for understanding the intrinsic ground state at low temperatures, such as in  $\beta$ -YbAlB<sub>4</sub>.

In summary, we have investigated the magnetic transitions and electrical anomalous upturn behavior at low temperatures of an  $\alpha$ -TmAlB<sub>4</sub> single crystal in a phenomenological view by measuring temperature-dependent magnetization, electrical resistivity, and specific heat capacity. The magnetic transition occurring around  $3\text{--}4 \text{ K}$  due to a structural mixing of the  $\alpha$  and  $\beta$ -phases, was not observed, which is consistent with the structural identification of the  $\alpha$ -phase. Therefore, we conclude that the zero magnetic field transitions and anomalous upturn in resistivity are intrinsic properties of an  $\alpha$ -TmAlB<sub>4</sub> single crystal. This study suggests that two types of YCrB<sub>4</sub> and ThMoB<sub>4</sub> can be separated in a TmAlB<sub>4</sub> system, similar to other cases (YbAlB<sub>4</sub>, LuAlB<sub>4</sub>, and ErAlB<sub>4</sub>),<sup>3,5</sup> although the close formation energies for the two structures were derived from calculations for the TmAlB<sub>4</sub> system.<sup>8</sup> Since we observed an anomalous behavior of electrical resistivity at low temperatures in the  $\alpha$ -TmAlB<sub>4</sub> single crystal, it would be of great interest to synthesize  $\beta$ -TmAlB<sub>4</sub> single crystals and study their low temperature properties.

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