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Specific heat and high-field magnetization of a TbPdSn single crystal

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Abstract

The thermal variation of the specific heat and magnetization curves has been measured in the orthorhombic TbPdSn single crystal. The successive magnetic transitions have been identified at $T_1 = 2.5\,\mathrm{K}$, $T_2 = 12.2\,\mathrm{K}$ and $T_\mathrm{N} = 23.8\,\mathrm{K}$. The magnetic entropy has been evaluated and discussed in the framework of crystal electric field (CEF) analysis performed in the previous work. The CEF energy scheme is presented. The high-field magnetization measurement up to 14 T along the *b*-axis exhibits a multistep metamagnetic transition. The metamagnetic step is described by $n/20 \times 7.7\mu_\mathrm{B}/\mathrm{f.u.}$ (*n*: integer).
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1. Introduction

The magnetic properties of the equiatomic ternary compounds of RMX (R = rare earth, M = transition metal and X = semimetal) with the orthorhombic ε-TiNiSi-type crystal structure have been extensively studied [1]. Among the RMX compounds, Tb compounds of TbNiSn and TbRhGe exhibit strongly anisotropic physical properties reflecting their crystal symmetry. The existence of successive magnetic transitions with temperature as well as the multistep metamagnetism under high field has been clarified for TbNiSn and TbRhGe single crystals [2–7]. For another isostructural compound TbPdSn, the results of bulk magnetic measurements on polycrystalline samples have been reported so far [8–10]; the existence of successive magnetic phase transitions and multistep metamagnetism at low temperatures was suggested. Our previous magnetic measurements on a single crystalline TbPdSn have indicated three magnetic anomalies at low temperature and multistep metamagnetic transition at 1.8 K [11]. The magnetic structure of TbPdSn was reported to be of a sine modulated type with the propagation vector $\boldsymbol{k} = (0.0, 0.25, 0.0)$ at 10 K and of a cone spiral type with the propagation vector $\boldsymbol{k} = (0.0, 0.25, 0.075)$ at 1.7 K by the powder neutron diffraction measurements [12].

In the present study, we have carried out the specific heat and high-field magnetization measurements on a TbPdSn single crystal up to 14 T to examine the successive magnetic phase transitions and to compare metamagnetism among the isostructural Tb equiatomic ternary compounds.

2. Experimental

The polycrystalline ingots were prepared by arc-melting the stoichiometric amounts of the constituent elements of Tb (99.9% purity), Pd (99.99% purity) and Sn (99.999% purity). A single crystal was grown by a Czochralski pulling method using a tri-arc furnace.

The low-temperature specific heat was measured by means of the relaxation method on a plate of the single crystal sample

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with 36.35 mg weight using a PPMS apparatus (Quantum Design) in the temperature range from 1.8 to 50 K. The magnetization measurements were carried out using a SQUID magnetometer (Quantum Design) at the magnetic fields up to 5.5 T. An extraction magnetometer was employed for the high-field magnetization measurements up to 14 T at 1.8 K.

3. Results and discussion

3.1. Successive phase transitions

Fig. 1 shows the thermal variation of the specific heat C_p of a TbPdSn single crystal. Two anomalies are indicated in the $C_p(T)$ at $T_2 = 11.5$ K and $T_N = 23.6$ K, respectively. The anomaly associated with the transition that the magnetic susceptibility has indicated at $T_1 = 2.5$ K is not very distinct in the $C_p(T)$ shown in Fig. 1. It can be clearly visible at 2.5 K in the magnetic specific heat divided by the temperature C_{mag}/T plotted in Fig. 2.

The successive phase transitions should be a common feature for the orthorhombic Tb ternary compounds of TbNiSn, TbRhGe and TbPdSn. The minor difference in the magnetic properties among these three Tb compounds is found in that TbPdSn has three magnetic phases below the T_N, whereas in TbNiSn and TbRhGe four magnetic phases exist below T_N. There is a transition at higher temperature T_3 by a few Kelvin than T_2 in TbNiSn and TbRhGe. It should be noted that the transition at T_3 is not intrinsic; the T_3 is merged into T_2 by applying moderate pressure in TbNiSn [5].

3.2. Magnetic entropy and crystal electric field (CEF) effect

The magnetic contribution to the specific heat of TbPdSn, C_{mag}, has been evaluated as follows. The total specific heat is assumed to be composed of the three parts: electron, lattice and magnetic contributions. Here, the electronic part is neglected. The main issue is to evaluate the lattice one in the magnetic compound TbPdSn. We have used the conventional Debye model in which the effective Debye temperature θ_D of magnetic TbPdSn is renormalized by multiplying it by a scaling factor [14] in the specific heat of isomorphous non-magnetic compound LaNiSn [13].

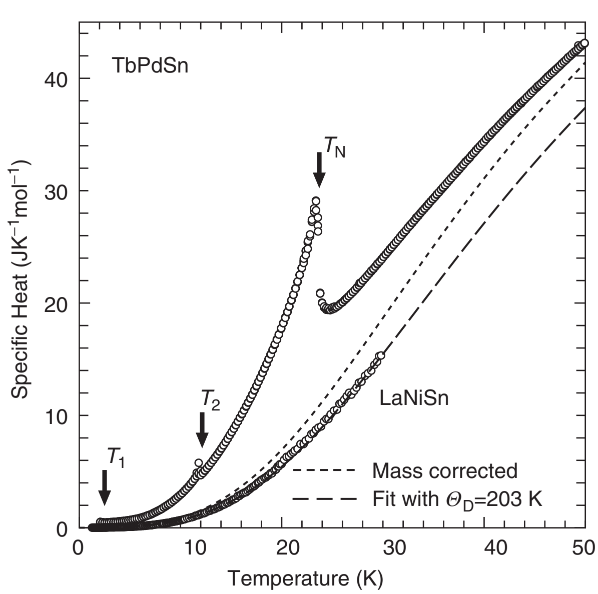

Fig. 1. The specific heat of a TbPdSn single crystal and a LaNiSn polycrystal.

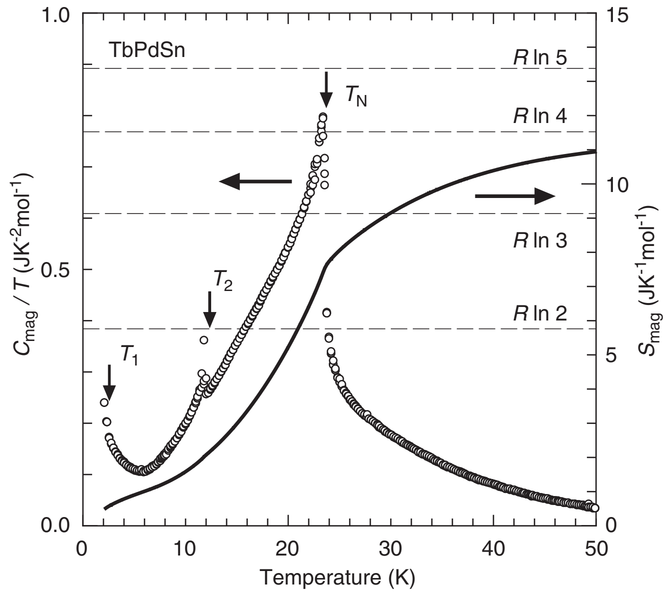

Fig. 2. The temperature dependence of magnetic specific heat divided by temperature and magnetic entropy of a TbPdSn single crystal.

First, the θ_D of LaNiSn was determined so as to fit the Debye function to the measured C_p of the compound. The dashed line in Fig. 1 gives the best fit to the C_p of LaNiSn. The 203 ± 5 K was deduced for the θ_D of LaNiSn. Then the θ_D of TbPdSn was calculated: $0.916 \times 203\,\mathrm{K} = 186$ K. The dotted line in Fig. 1 represents the calculated lattice contribution of TbPdSn. The C_{mag} was obtained by subtracting thus obtained lattice contribution from the C_p.

The C_p/T and magnetic entropy S_{mag} of TbPdSn are plotted in Fig. 2. Three transitions are indicated more distinctly in the plot than the C_p vs. T in Fig. 1. The S_{mag} reaches only 11.0 J K^{-1} mol^{-1} at 50 K which is smaller than R ln 4. However, it is very interesting to note that the calculated S_{mag} gives a comparable magnitude of 11.4 J K^{-1} mol^{-1} at 50 K for the CEF energy values determined by the analysis of the anisotropic magnetic susceptibility of the compound [11]. The set of CEF energy values are listed in Table 1. Here, we conclude only that the CEF scheme determined by the previous magnetic measurements is good enough to account for the C_{mag} of the compound.

3.3. Magnetization curves

3.3.1. High-field magnetization

Fig. 3 shows the high-field magnetization curve for the *b*-axis at 1.8 K. The magnetization along the *b*-axis exhibits

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