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Magnetic properties of a Tb_5Sn_3 single crystal

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Abstract

Single crystals of Tb_5Sn_3 have been successfully grown by the Czochralski method. Paramagnetic susceptibility of Tb_5Sn_3 obeys Curie–Wiss law and effective paramagnetic moment obtained from reciprocal susceptibility is $10.3\mu_B$ along the *c*-axis and $10.0\mu_B$ in the *c*-plane; paramagnetic Curie temperatures along the *c*-axis and in the *c*-plane are −0.78 and 22.2 K, respectively. Tb_5Sn_3 takes antiferromagnetic phase below $T_N = 62$ K as reported by Semitelou and Yakinthos; a metamagnetic transition or a spin flop was observed at 4.2 K under up to 50 kOe external magnetic field. Another magnetic phase can exist below $T_t = 3.8$ K. Electrical resistivity along the *c*-axis shows a pronounced hump just below T_N due to a superzone gap formation along the new Brillouin zone boundaries.

Keywords: Tb_5Sn_3; Magnetization; Magnetic susceptibility; Electrical resistivity; Antiferromagnetic

1. Introduction

The intermetallic compounds R_5Sn_3 crystallize in the Mn_5Si_3 type hexagonal structure with the space group $P6_3/mcm$ [1,2]. Since many rare earth elements form isostructural compounds with Sn in R_5Sn_3, systematic studies can be made for the magnetic and transport properties in the isostructural compounds having different rare earth ions. In the previous works, magnetic properties were studied by polycrystalline samples for Ce_5Sn_3, Sm_5Sn_3 and Tb_5Sn_3 [3–5]. However for the other R_5Sn_3 compounds, no investigation on magnetic or electrical properties has been reported as far as we know. Furthermore, single crystals are required for precise studies in these compounds. Thus we have started to study the magnetic and electrical properties of R_5Sn_3 system using single crystals. Among the R_5Sn_3 compounds, Tb_5Sn_3 is the only compound for which magnetic structure was examined by neutron diffraction studies [5]; an incommensurate and commensurate transverse amplitude modulated structures were proposed at just below the Neel temperature of $T_N = 62$ K and at 1.8 K, respectively. In this report, we will present the results of magnetic and electrical measurements for Tb_5Sn_3 single crystals.

2. Experimental

The polycrystalline ingots were prepared by arc-melting the constituent 99.9% pure Tb and 99.99% pure Sn elements under high purity argon atmosphere. Before this treatment, Tb ingots were pre-reacted with liquid phase Sn in an evacuated quarts tube at 1050 °C for 72 h to prevent the loss of Sn atoms in the arc-melting process. The obtained Tb_5Sn_3 polycrystalline compounds were found to be in a single phase by powder X-ray diffraction measurements. Single crystals of Tb_5Sn_3 were grown by the Czochralski method from single-phase polycrystalline samples using a tri-arc furnace. The obtained single crystal ingots were formed into sphere and rectangular shape and annealed at 300 °C for 24 h in an evacuated quarts tube. The crystal orientation was determined by the back reflection Laue method. The magnetization and magnetic susceptibility were measured by a vibrating-sample magnetometer at 4.2 K up to 50 kOe and from 4.2 to 300 K at 10 kOe external magnetic field. The ac magnetic susceptibility was measured using a standard Hartshorn bridge circuit in

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the temperature range from 1.5 to 100 K. Electrical resistivity measurements were made by a dc four-terminal method.

3. Results and discussion

Fig. 1 shows the magnetization curves for Tb_5Sn_3 at 4.2 K in the three directions. Here, the *b*-axis refers to the [1 2 0] direction in the hexagonal cell. Along the *c*-axis, *M* increases linearly with increasing magnetic field *H* in low magnetic fields. Thus Tb_5Sn_3 is considered to be in an antiferromagnetic state at 4.2 K. Magnetization curve deviates from the linear increase at about $H_C = 30$ kOe along the *b*-axis and at 35 kOe along the *a*-axis. Therefore, a metamagnetic transition or a spin flop takes place in the *c*-plane; magnetization value at 50 kOe is about 1/5 of the *gJ* value of Tb^{3+}, $9.0\mu_B$. Easy magnetization axis is in the *c*-plane. Higher magnetic fields are required to clarify the further magnetization process.

Magnetic susceptibility χ and reciprocal susceptibility χ^{-1} are shown in Fig. 2 as a function of temperature for three axes. High temperature magnetic susceptibility obeys Curie–Weiss law and the χ shows a cusp at 62 K for all axes; no magnetic anisotropy is observed in the *c*-plane. The value of χ along the *c*-axis is smaller than that along the other two axes. These results suggest that the magnetic moment of Tb lies in the *c*-plane. From the results of powder neutron diffraction studies for Tb_5Sn_3 [5], Semitelou and Yakinthos suggested the amplitude modulated magnetic structure where magnetic moments are oriented along the *a*-axis; there is no contradiction between our results and the suggested magnetic structure. Though the reciprocal susceptibility χ^{-1} shows a liner relation in the *c*-plane above 100 K, the χ^{-1} along the *c*-axis deviates from the linear variation at around 200 K when temperature decreases. Deviation of the reciprocal susceptibility from linear relation is considered to be originated by the crystalline electric field (CEF) effect. There are two different crystallographic sites of Tb ion in Tb_5Sn_3; the length of the *c*-axis is shorter than that of the *a*-axis. Thus magnetic interactions or the CEF effect along the *c*-axis may be more considerable than that in the *c*-plane. Obtained effective magnetic moment μ_{eff} is $10.0\mu_B$/Tb in the *c*-plane by using the data from 100 to 300 K. For the *c*-axis, μ_{eff} was determined to be $10.3\mu_B$/Tb by using the data from 200 to 300 K. However this value can change according to the fitting temperature range; this value includes the error of $\pm 1.0\mu_B$/Tb. The μ_{eff} values are considered to be in reasonable agreement with the theoretical value of $9.72\mu_B$ for the Tb^{3+} free ion, but higher temperature data is required to obtain the precise μ_{eff} value for the *c*-axis. Estimated asymptotic Curie temperatures θ_p are 22.2 K for the *c*-plane and −0.78 K for the *c*-axis.

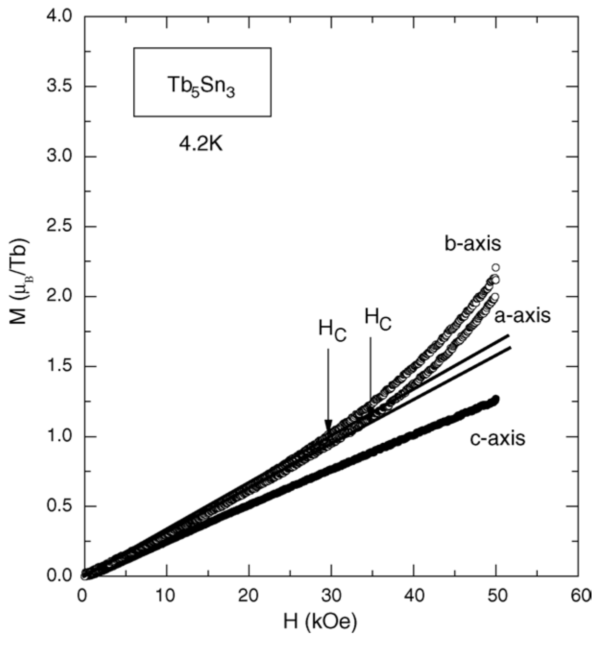

Fig. 1. Magnetization curves for Tb_5Sn_3 at 4.2 K. The H_C indicates the critical magnetic field.

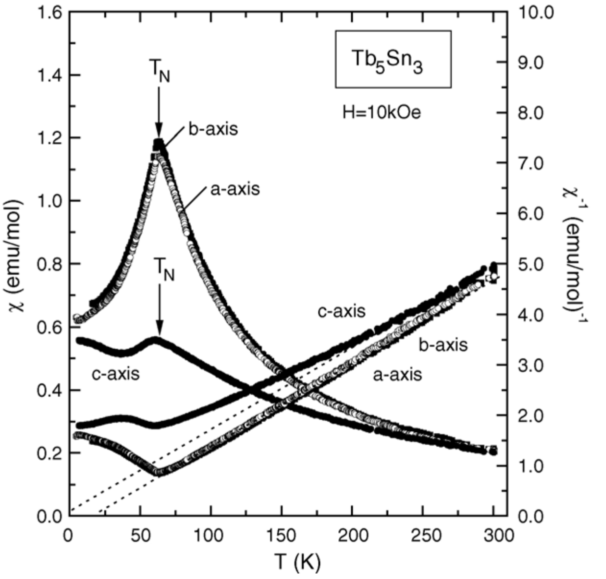

Fig. 2. Magnetic susceptibility χ and reciprocal susceptibility χ^{-1} for Tb_5Sn_3 as a function of temperature.

Fig. 3 shows the temperature variation of ac magnetic susceptibility χ_{ac} along the *c*-axis and the *a*-axis at low temperatures. The χ_{ac} has a cusp at 3.8 K in both axes. This anomaly indicates the existence of another magnetic phase below $T_t = 3.8$ K though the possibility of the effect of the trace of Sn impurity might be undeniable. Thus more detailed studies are required for Tb_5Sn_3 using single crystals.

Electrical resistivity ρ of Tb_5Sn_3 is shown in Fig. 4 for the *a*- and *c*-axes as a function of temperature. Initially, electrical resistivity decreases with decreasing temperature; ρ has a minimum at around 100 K and increases. Then ρ has a hump at just below T_N along the *c*-axis, and a break along the *a*-axis. It is considered that the hump is attributed to an energy gap formation along the new Brillouin zone boundaries due

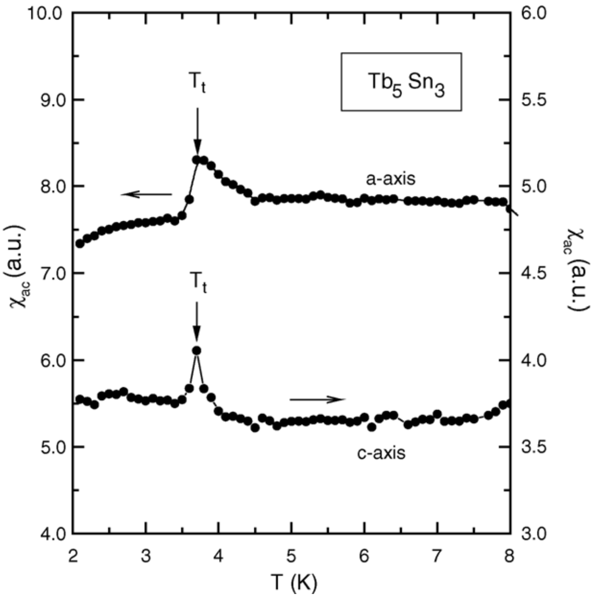

Fig. 3. The ac magnetic susceptibility χ_{ac} for Tb_5Sn_3 as a function of temperature.

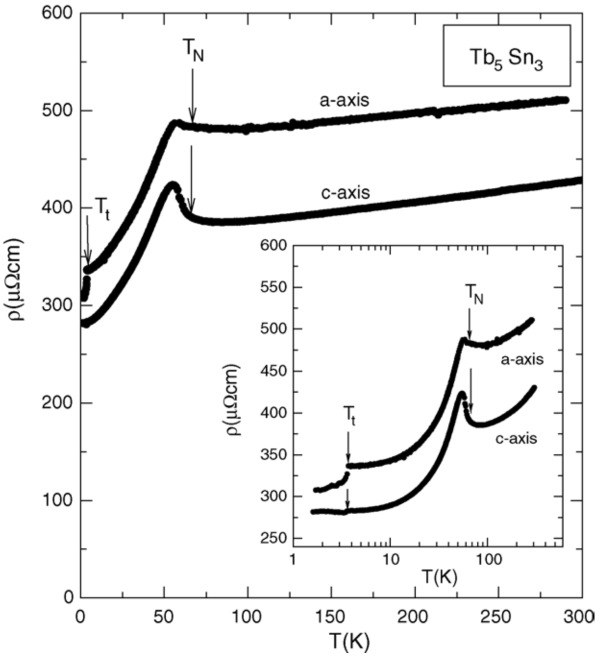

Fig. 4. Electrical resistivity ρ for Tb_5Sn_3 as a function of temperature. The inset shows the variation of ρ with log T.

to antiferromagnetic order. From the powder neutron diffraction experiments, propagation vector of magnetic moment is $Q_z = (0\ 0\ 1/2)$ at 1.8 K. This indicates that a new boundary is formed along the *c*-axis. Therefore, pronounced increase of ρ along the *c*-axis is in a good agreement with this magnetic structure. Though the origin of the above mentioned increase of ρ above T_N is not clear, the structure of state density spectrum at the Fermi level may be connected to this property [6]. Such kind of temperature variation of electrical resistivity in paramagnetic state can be seen in R_5Si_3 and R_5Ge_3 systems (R = Gd and Tb) [7,8]. Further, electrical resistivity shows a kink at 3.8 K in both axes as shown in the inset of Fig. 4. This anomaly corresponds to another magnetic transition denoted by T_t. Since the anomaly of ρ is larger along the *a*-axis than that along the *c*-axis, it is considered that the some magnetic structure change in the *c*-plane exists at T_t.

4. Conclusion

Magnetic and electrical properties of Tb_5Sn_3 have been studied using single crystals. Tb_5Sn_3 possesses an antiferromagentic phase below $T_N = 62$ K; another magnetic phase can exist below $T_t = 3.8$ K. A metamagnetic transition or a spin flop occurs at 4.2 K below 50 kOe external magnetic field. Electrical resistivity along the *c*-axis shows an anomaly just below T_N indicating a superzone gap formation along the new brillouin zone boundaries. For further investigations, magnetization measurement in the higher magnetic fields, neutron diffraction studies for single crystals and specific heat measurements are now in progress.

References

[1] A. Iandelli, A. Palenzona, in: K.A. Gscheneidner Jr., L. Eyting (Eds.), Handbook on the Physics and Chemistry of Rare Earths, North-Holland, 1979, p. 16.
[2] W. Jeitschko, E. Parthe, Acta Crystallogr. 22 (1967) 551.
[3] A. Palenzona, F. Merlo, Atti Acad. Naz. Lincei, Rend. Sci. Fis. Mater. Nat. XL (1966) 617.
[4] A. Parcheron, P. Lethuillier, R. Acad. Sci., Paris 271 (1972) 1129.
[5] J.P. Semitelou, J.K. Yakinthos, J. Magn. Magn. Mater. 186 (1998) 107.
[6] P.B. Allen, Phys. Rev. B 17 (1978) 3275.
[7] F. Canepa, S. Cirafici, F. Merlo, A. Palenzona, J. Magn. Magn. Mater. 118 (1993) 182.
[8] T. Tsutaoka, Y. Nishiume, T. Tokunaga, J. Magn. Magn. Mater. 272–276 (2004) e421.