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Magnetic anisotropy and phase transitions in single-crystal $Tb_5(Si_{2.2}Ge_{1.8})$

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The $Tb_5(Si_xGe_{4-x})$ alloy system has many features in common with the $Gd_5(Si_xGe_{4-x})$ system although it has a more complex magnetic and structural phase diagram. This paper reports on the magnetic anisotropy and magnetic phase transition of single-crystal $Tb_5(Si_{2.2}Ge_{1.8})$ which has been investigated by the measurements of *M-H* and *M-T* along the *a*, *b*, and *c* axes. The variation of $1/\chi$ vs *T* indicates that there is a transition from paramagnetic to ferromagnetic at $T_c=110$ K. Below this transition temperature *M-H* curves show very strong anisotropy, and it is believed that this is due to the complex spin configuration. *M-H* measurements at $T=110$ K show that the *a* axis is the easy axis, and that the saturation magnetization is 200 emu/g. The *b* axis is the hard axis, which needs an external magnetic field much higher than 2 T to saturate the magnetization in that direction, indicating a high magnetocrystalline anisotropy. The *c* axis is of intermediate hardness. The magnetic properties of this material are therefore very different from those of the related $Gd_5Si_2Ge_2$ system, in which the *b* axis was found to be the easy axis and the magnitude of the anisotropy was smaller. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1855196]

INTRODUCTION

A number of pseudobinary compounds $R_5(Si_xGe_{4-x})$, where *R* is La, Lu, Gd, Nd, or Dy, have been investigated by Gschneidner *et al.*[1] The alloy $Gd_5(Si_xGe_{1-x})_4$ which has received much attention recently has several unique properties including a giant magnetocaloric effect,[2] giant magnetoresistance,[3] and giant magnetostriction.[4] Morellon *et al.*[5,6] have investigated phase transitions and the magnetocaloric effect in the alloys with $R=$Tb and Thuy *et al.*[7] have studied both magnetic properties and magnetocaloric effect in polycrystalline samples with specific compositions $Tb_5(Si_2Ge_2)$ and $Tb_5(Si_3Ge_1)$. Although $Tb_5(Si_xGe_{1-x})_4$ has many similarities with $Gd_5(Si_xGe_{1-x})_4$, it shows a more complicated magnetocrystallographic transformation according to Ritter *et al.*[8] Spichkin *et al.*[9] have found that the magnetic ordering temperatures of these alloys range from a Curie temperature of $T_c=225$ K in Tb_5Si_4, to a Néel temperature T_N of 91 K in Tb_5Ge_4. This paper reports on the magnetic anisotropy and magnetic phase transition of single crystal $Tb_5(Si_xGe_{1-x})_4$, where $x=0.55$, which has been investigated by superconducting quantum interference device (SQUID) measurements of *M-H* and *M-T* characteristics along the *a*, *b*, and *c* axes.

EXPERIMENTAL DETAILS

Appropriate quantities of terbium 99.98% (metals basis, wt. %), silicon (99.9999%), and germanium (99.999%) were cleaned and arc-melted several times under an argon atmosphere. The buttons were then remelted to ensure compositional homogeneity throughout the ingot and the alloy was drop cast into a copper chill cast mold. The as-cast ingot was electron-beam welded in a tungsten Bridgman style crucible for crystal growth. The ingot was heated in a tungsten mesh resistance furnace under a pressure of 8.8×10^{-5} Pa up to 1700 °C to degas the crucible and charge. The chamber was then backfilled to a pressure of 3.4×10^{4} Pa with high-purity argon. This overpressurization was used in order to equalize the pressure inside and outside of the crucible at the final temperature. The ingot was then heated to 2050 °C after which it was withdrawn from the heat zone at a rate of

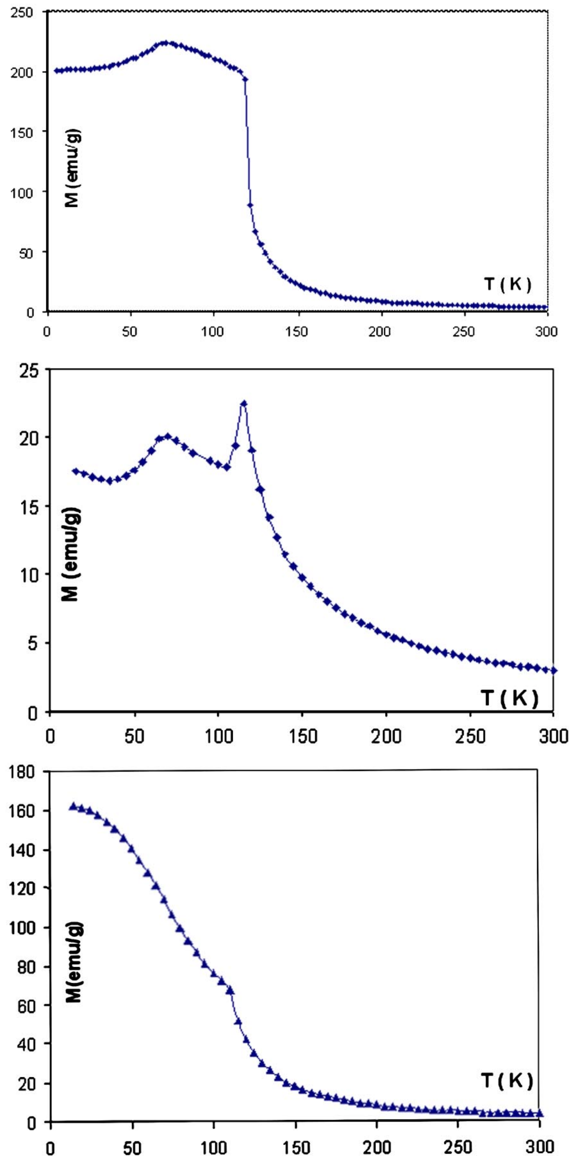

FIG. 1. Magnetization as a function of temperature measured along the *a* axis under an applied field of $H=10$ kOe. (b) Magnetization as a function of temperature measured along the *b* axis under an applied field of $H=10$ kOe. (c) Magnetization as a function of temperature measured along the *c* axis under an applied field of $H=10$ kOe.

8 mm/h. The as-grown crystal was oriented by back-reflection Laue and the crystallographic directions assigned using x-ray diffraction two-theta scans of the single crystal. The sample was cut by electrical discharge machining (EDM) and the oriented faces were prepared using standard metallographic techniques to yield flat, parallel faces.

Magnetization versus temperature measurements were conducted with magnetic field applied along the three principal crystal axes in an MPMS-5S SQUID magnetometer. Measurements were made over the range from room temperature to $T=15$ K. In order to investigate the effect of magnetic field on the transition temperature, two different magnitudes of the magnetic fields were applied: 10 kOe was applied along the *a*, *b*, and *c* axes, and 20 kOe was applied also along the *a* axis. Magnetization versus field measurements were then made at a fixed temperature of 110 K.

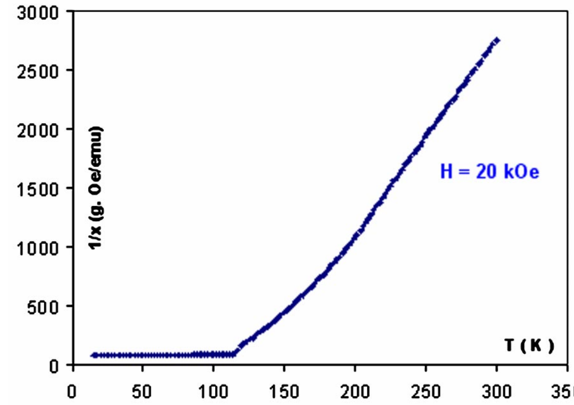

FIG. 2. Variation of $1/\chi$ at a field 20 kOe along the *a* axis with temperature *T* over the range 10–300 K showing Curie–Weiss behavior at higher temperatures and a transition to an ordered magnetic state below 110 K.

RESULTS AND DISCUSSION

The variation of magnetization with temperature under a field of 10 kOe along the *a* axis is shown in Fig. 1(a), and similarly along the *b* and *c* axes in Figs. 1(b) and 1(c), respectively. From these results, particularly the rapid change in magnetization along the *a* axis with temperature, the lambda anomaly along the *b* axis, and the discontinuity in slope along the *c* axis, it is clear that a phase transition occurs at a temperature of 110 K. The magnetization is strongly dependent on the crystallographic direction along which the field is applied, which indicates a strong magnetocrystalline anisotropy. In addition, it is interesting to find that, unlike the $Gd_5(Si_{1.95}Ge_{2.05})$ in which the transition temperature changes by 5 K/T,[10] the transition temperature in $Tb_5(Si_{2.2}Ge_{1.8})$ does not show a dependence on the magnetic field. This finding is consistent with the results reported for polycrystalline $Tb_5(Si_2Ge_2)$.[6]

The variation of reciprocal susceptibility $1/\chi$ with temperature along the *a* axis under a field of 20 kOe (2 T) is shown in Fig. 2. For temperatures above 110 K the behavior is Curie–Weiss like which is indicative of a paramagnetic state with weak interactions between localized magnetic moments. This can be compared with the magnetic order/disorder transition temperature of 268 K in $Gd_5Si_2Ge_2$.[10] According to Ritter *et al.*,[8] the transition observed here is a first-order transition from a higher-temperature phase, which is paramagnetic and monoclinic (space group $P112_1/a$), into a lower-temperature phase, which is ferromagnetic and orthorhombic (space group *Pnma*). However, according to Morellon *et al.*,[6] unlike $Gd_5(Si_2Ge_2)$, the magnetic and structural transitions observed in $Tb_5Si_2Ge_2$ do not occur together, but rather appear to be separated by a temperature difference of about 8 K. The magnetic behavior that we observe below the transition temperature in Fig. 1 is believed to be due to the separation of the magnetic and structural phase transitions, and the more complex magnetic structure of $Tb_5(Si_2Ge_2)$ below the Curie temperature, which has been discussed in detail by Morellon *et al.*[6] and Ritter *et al.*[8]

M-H measurements have been conducted along the *a*, *b*, and *c* axes at 110 K, as shown in Fig. 3. These results show that the *a* axis is the easy axis, the *c* axis is of intermediate

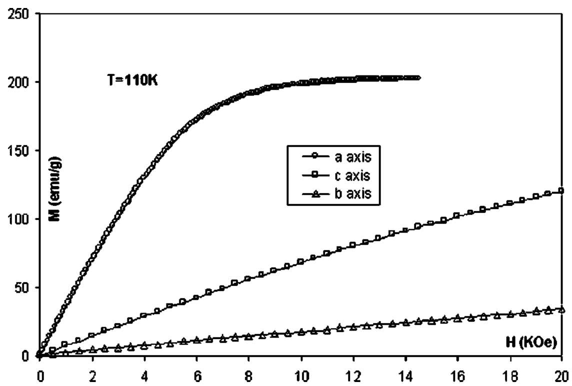

FIG. 3. Magnetization curves along the crystallographic *a*, *b*, and *c* axes at $T=110$ K.

hardness, and the *b* axis is the hard axis. The saturation magnetization is 200 emu/g. The calculation of the magnetic anisotropy from these magnetization curves gave a value of approximately 8.8×10^7 J/m^3 (8.8×10^8 emu/cm^3), which is the same order of magnitude as single-crystal pure Tb metal, which is about 6×10^8 emu/cm^3,[11] although the anisotropy in pure Tb metal is planar rather than axial. For comparison, measurements on $Gd_5Si_2Ge_2$ at 260 K, which is 9 K below its magnetic structural transition temperature, showed that the *b* axis is the easy axis, with a saturation magnetization of $M_s=0.6\times10^6$ A/m (600 emu/cm^3) and a magnetic anisotropy of $K=4.1\times10^4$ J/m^3.[12] Thus $Tb_5Si_{2.2}Ge_{1.8}$ has a much larger magnetic anisotropy than $Gd_5Si_2Ge_2$, by more than three orders of magnitude. This results from the nonzero orbital angular momentum L for the Tb atom ($L=3$ for Tb, $L=0$ for Gd).

CONCLUSIONS

Magnetic property measurements have been made on single-crystal $Tb_5Si_{2.2}Ge_{1.8}$ and have been found to be significantly different from the magnetic properties of the related $Gd_5Si_2Ge_2$ system which was studied previously. The variation of magnetization with temperature has shown that in this alloy the Curie point occurs at 110 K compared with 268 K in $Gd_5Si_2Ge_2$.[10] The variation of magnetization as a function of magnetic field shows that the a axis is the magnetic easy axis, with a magnetocrystalline anisotropy of approximately 8.8×10^7 J/m^3 and a saturation magnetization of $M_s=200$ emu/g (which with a density of 7.6 gm/cm^3 gives $M_s=1520$ emu/cm^3, or 1.52×10^6 A/m). This compares with $Gd_5Si_2Ge_2$ which has the *b* axis as the easy axis with a lower anisotropy of 4.1×10^4 J/m^3 and a saturation magnetization of $M_s=0.6\times10^6$ A/m (600 emu/cm^3).[12]

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