Magnetic anisotropy and phase transitions in single-crystal \( \text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8}) \)

M. Han and J. E. Snyder  
Materials and Engineering Physics Program, Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011 and Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011

W. Tang  
Materials and Engineering Physics Program, Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011

T. A. Lograsso  
Materials and Engineering Physics Program, Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011 and Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011

D. L. Schlagel  
Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011

D. C. Jiles  
Materials and Engineering Physics Program, Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, Iowa 50011 and Department of Materials Science and Engineering, Iowa State University, Ames, Iowa 50011

EXPERIMENTAL DETAILS

A number of pseudobinary compounds \( R_5(\text{Si}_{x}\text{Ge}_{4-x}) \), where \( R \) is La, Lu, Gd, Nd, or Dy, have been investigated by Gschneidner et al.\(^1\). The alloy \( \text{Gd}_5(\text{Si}_{0.5}\text{Ge}_{3.5}) \) which has received much attention recently has several unique properties including a giant magnetocaloric effect,\(^2\) giant magneto-resistance,\(^3\) and giant magnetostriction.\(^4\) Morellon et al.\(^5\) have investigated phase transitions and the magneto-caloric effect in the alloys with \( R=\text{Tb} \) and Thy et al.\(^7\) have studied both magnetic properties and magneto-caloric effect in polycrystalline samples with specific compositions \( \text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{3.5}) \) and \( \text{Tb}_5(\text{Si}_{1}\text{Ge}_{4}) \). Although \( \text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{3.5}) \) has many similarities with \( \text{Gd}_5(\text{Si}_{0.5}\text{Ge}_{3.5}) \), it shows a more complicated magnetocrystallographic transformation according to Ritter et al.\(^6\) Spichkin et al.\(^7\) have found that the magnetic ordering temperatures of these alloys range from a Curie temperature of \( T_C=225 \text{ K} \) in \( \text{Tb}_5\text{Si}_5 \) to a Néel temperature \( T_N \) of \( 91 \text{ K} \) in \( \text{Tb}_5\text{Ge}_4 \). This paper reports on the magnetic anisotropy and magnetic phase transition of single crystal \( \text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{3.5}) \), 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.
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 \( T = 15 \text{ K} \). 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.

RESULTS AND DISCUSSION

The variation of magnetization with temperature under a field of 10 kOe along the \( a \) axis is shown in Fig. 1, 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 Gd₅(Si₁.₉₅Ge₂.₀₅) in which the transition temperature changes by 5 K/T,¹⁰ the transition temperature in Tb₅(Si₂.₅Ge₁.₈) does not show a dependence on the magnetic field. This finding is consistent with the results reported for polycrystalline Tb₅(Si₂Ge₂).⁶

The variation of reciprocal susceptibility \( 1/\chi \) with temperature along the \( a \) axis under a field of 20 kOe 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 Gd₅Si₂Ge₂.¹⁰ 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 Gd₅(Si₂Ge₂), the magnetic and structural transitions observed in Tb₅Si₂Ge₂ 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 Tb₅(Si₂Ge₂) 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. 1. Magnetization as a function of temperature measured along the \( a \) axis under an applied field of \( H = 10 \text{ kOe} \). (b) Magnetization as a function of temperature measured along the \( b \) axis under an applied field of \( H = 10 \text{ kOe} \). (c) Magnetization as a function of temperature measured along the \( c \) axis under an applied field of \( H = 10 \text{ kOe} \).

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.
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 $8.8 \times 10^7$ J/m$^3$ ($8.8 \times 10^8$ emu/cm$^3$), which is the same order of magnitude as single-crystal pure Tb metal, which is about $6 \times 10^8$ emu/cm$^3$, although the anisotropy in pure Tb metal is planar rather than axial. For comparison, measurements on Gd$_5$Si$_2$Ge$_2$ 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 $M_s = 0.6 \times 10^6$ A/m ($600$ emu/cm$^3$) and a magnetic anisotropy of $K = 4.1 \times 10^4$ J/m$^3$. This compares with Gd$_5$Si$_2$Ge$_2$ which has the b axis as the easy axis with a lower anisotropy of $4.1 \times 10^4$ J/m$^3$ and a saturation magnetization of $M_s = 0.6 \times 10^6$ A/m ($600$ emu/cm$^3$).

CONCLUSIONS

Magnetic property measurements have been made on single-crystal Tb$_5$Si$_2$Ge$_1.8$ and have been found to be significantly different from the magnetic properties of the related Gd$_5$Si$_2$Ge$_2$ 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$_5$Si$_2$Ge$_2$. 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 $8.8 \times 10^7$ 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 $1.52 \times 10^6$ A/m). This compares with Gd$_5$Si$_2$Ge$_2$ which has the b axis as the easy axis with a lower anisotropy of $4.1 \times 10^4$ J/m$^3$ and a saturation magnetization of $M_s = 0.6 \times 10^6$ A/m ($600$ emu/cm$^3$).

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