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Variation of magnetostriction with temperature in $\text{Tb}_5\text{Si}_{2.2}\text{Ge}_{1.8}$ single crystal

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The $\text{Tb}_5(\text{Si}_x\text{Ge}_{4-x})$ alloy system is similar to the better known $\text{Gd}_5(\text{Si}_x\text{Ge}_{4-x})$, except it has a more complex magnetic and structural phase diagram. $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ has received much attention recently due to its giant magnetocaloric effect, colossal magnetostriction and giant magnetoresistance in the vicinity of a first order combined magnetic-structural phase transition. The magnetostriction changes that accompany the phase transitions of single crystal $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$ have been investigated at temperatures between 20 and 150 K by measurements of magnetostriction along the a axis. Over this temperature range the shape and slope of the magnetostriction curves change, indicative of changes in the magnetic state, crystal structure, and magnetic anisotropy. The results appear to indicate a phase transition that occurs near 106 K (onset-completion range of 116–100 K). The steepness of the strain transition, its unusual hysteresis, and its temperature dependence appear to indicate a first order phase transition which is activated by applied magnetic field in addition to temperature (see Fig. 1). Magnetostriction measurements at temperature below the transition region appear to indicate a magnetostriction of small overall magnitude (about 30×10^{-6}) but high anisotropy, with anisotropy showing considerable temperature dependence. © 2006 American Institute of Physics. [DOI: 10.1063/1.2171951]

I. INTRODUCTION

The $\text{Tb}_5(\text{Si}_x\text{Ge}_{4-x})$ alloy system is similar to the better known $\text{Gd}_5(\text{Si}_x\text{Ge}_{4-x})$, except it has a more complex magnetic and structural phase diagram. $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ has received much attention recently due to its giant magnetocaloric effect, colossal magnetostriction, and giant magnetoresistance in the vicinity of a first order combined magnetic-structural phase transition. $\text{Tb}_5(\text{Si}_x\text{Ge}_{4-x})$ also demonstrates many of these features. Magnetostriction changes accompany the phase transitions of single crystal $\text{Tb}_5(\text{Si}_x\text{Ge}_{4-x})$. In this paper, linear magnetostriction along the a axis of single crystal $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$ has been investigated at different temperatures over a range of 20 to 150 K and from the results, details of the magnetic and structural phase transitions have been inferred.

II. EXPERIMENTAL DETAILS

Single crystal $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$ was produced via the Bridgman method, as described in Ref. 1. An a - b surface was produced via electron discharge cutting and cleaned with ethanol. A strain gauge was then bonded to this surface. The sample was then mounted to a copper block using a thermally conductive silver epoxy. The temperature sensor was connected to this block with a screw and thermally conductive paste. The copper block was mounted with this same thermally conductive paste to the cold finger of a closed cycle He refrigeration system. The sample was oriented so

that the applied field and strain gauge measurement directions were both along the a axis. The cryogenic system with the sample mounted inside was then pumped down to 10^{-7} torr and the system was cooled to 14 K. Before each measurement of a magnetostrictive strain versus applied field curve, the temperature was increased to the target temperature and controlled using an electric heater in the cold finger. The magnetic field was applied via a computer control system and measured by a gauss meter. The magnetostriction was measured using the strain gauge method. Both field and strain gauge readings were recorded on the computer together with the temperature at which the measurements were made.

III. RESULTS AND DISCUSSION

For the purposes of this discussion, the measurement results of the strain along the a axis as a function of magnetic field can be grouped into three different temperature regions, which follow the regions of the phase diagram determined in Ref. 2. Starting at 20 K (Fig. 1), the strain versus field (λ vs H) curve had a distinct shape. Starting at low applied field, the strain peaks at 30 ppm near 3 kOe, then decreases steeply, and becomes negative for high applied field magnitude where the trend becomes one of contraction. This trend continues until one passes from the low temperature ferromagnetic orthorhombic(I) (FM2) phase to the ferromagnetic orthorhombic(I) (FM1) phase near 70 K.² At that temperature the maximum strain at 3 kOe drops to about 10 ppm, and the slope above 3 kOe becomes zero and then for higher

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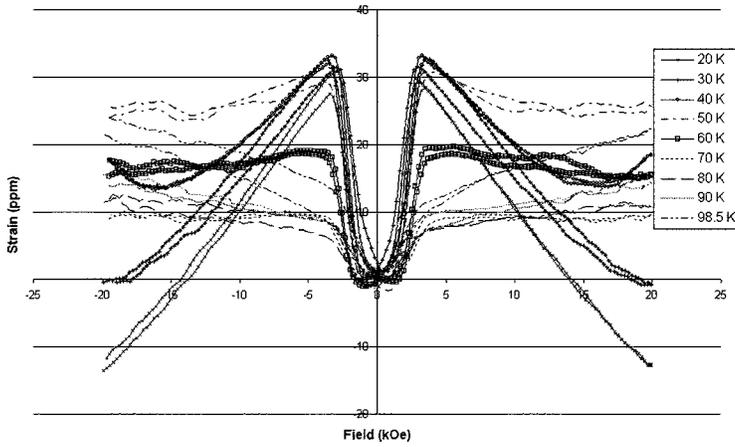


FIG. 1. Strain vs applied field for $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$, measured along the a axis (field along the a axis), for a temperature range of 20–98.5 K.

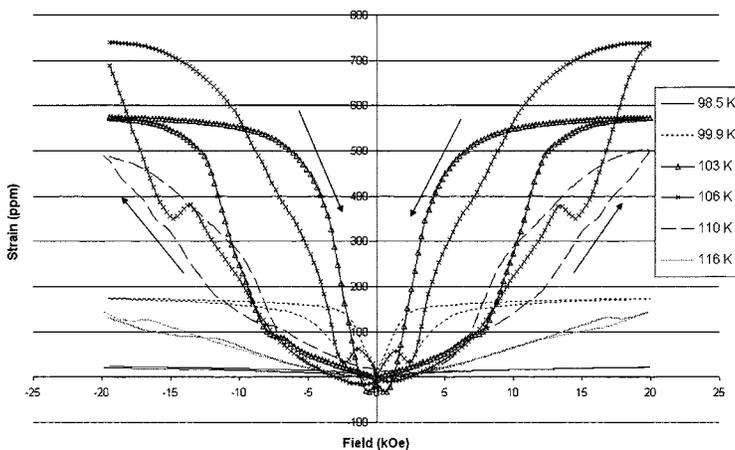


FIG. 2. Strain vs applied field for $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$, measured along the a axis (field along the a axis), for a temperature range of 98.5–116 K.

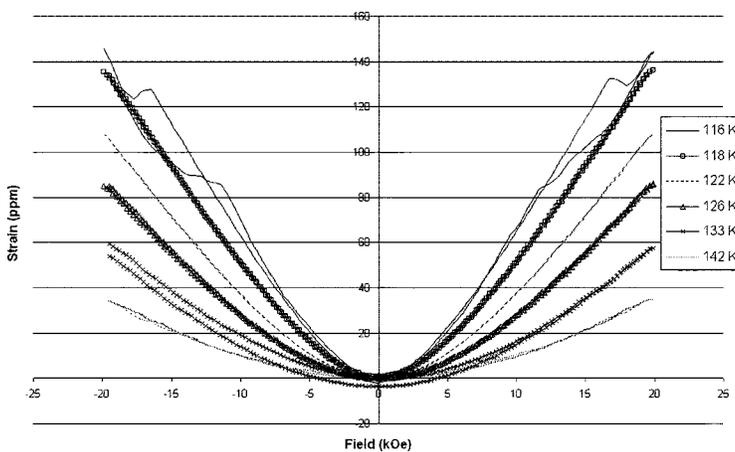


FIG. 3. Strain vs applied field for $\text{Tb}_5(\text{Si}_{2.2}\text{Ge}_{1.8})$, measured along the a axis (field along a axis), for a temperature range of 116–142 K.

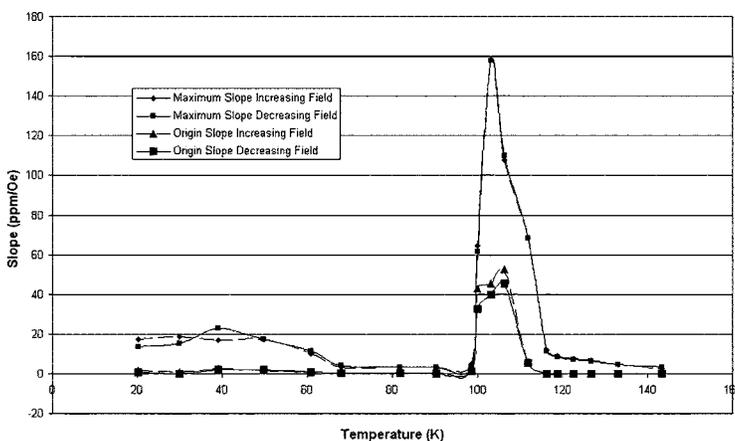


FIG. 4. Maximum slope and slope at the origin of magnetostriction along the a axis (field along the a axis), for a temperature range of 20–142 K.

temperatures becomes positive and becomes more linear. The high field slope becomes increasingly positive for increasing temperature until just below 100 K.

Between 98.5 and 116 K (Fig. 2), the structure of the material changes from a Gd_5Si_4 -type orthorhombic crystal structure to a $\text{Gd}_5\text{Si}_2\text{Ge}_2$ -type monoclinic structure.² This is the transition from a low temperature ferromagnetic to a higher temperature paramagnetic state.³ The structural change is influenced by the applied magnetic field. This field-affected structural change is demonstrated by the unusual magnetomechanical hysteresis evident in the transition area λ vs H curves. As increasing field is applied, the initial slope of strain versus applied field is low, but at higher field, there is an abrupt increase in strain, followed by a decrease of slope again. As field is decreased, strain remains large to lower applied field, then again shows an abrupt decrease before reaching zero applied field. This hysteresis region is also observed to shift toward higher applied field values as temperature increases. The maximum strain in this region of magnetically and thermally induced phase transitions is much larger than the maximum strain from the paramagnetic and ferromagnetic regions on either side. The maximum strain and the width of the hysteresis are observed to increase, peak at 106 K, then decrease again. There would appear to be a first order transition occurring in this region, triggered by an applied magnetic field (it shows an abrupt change in strain, it shows a hysteresis, and, for increased temperature, it requires more applied field to trigger the transition), however, the behavior of this material appears more complex than the first order magnetic-structural phase transformation previously observed in $\text{Gd}_5\text{Si}_2\text{Ge}_2$ (Ref. 4) and might represent that proposed by Morellon *et al.*⁵ in which in this temperature range the ferromagnetic orthorhombic phase can transform to ferromagnetic monoclinic phase before transforming to paramagnetic monoclinic phase.

Above this structural change (Fig. 3), the shape of the magnetostriction versus field (λ vs H) curve settles into a regular “U” shape. From 118 to 150 K, the material stays in the $\text{Gd}_5\text{Si}_2\text{Ge}_2$ -type monoclinic structural state.² As the material becomes paramagnetic, the magnitude drops steadily, from above 130 ppm, down to about 35 ppm. The unusual magnetomechanical hysteresis is gone from the sample in this region. It also does not show the ordinary magnetic hysteresis often observed in magnetostriction curves.

Comparing the slopes $d\lambda/dH$, both the maximum value and the value at the origin, across the range of temperatures from 20 to 150 K (Fig. 4), it is immediately evident that a large discontinuity is present between 100 and 116 K. This is attributed to the first order transition that arises because of both structural and magnetic changes occurring in the material and the fact that this transition is field dependent. This

might represent the phase transition sequence proposed by Morellon *et al.*⁵ in which in this temperature range the ferromagnetic orthorhombic phase can transform to ferromagnetic monoclinic phase before transforming to paramagnetic monoclinic phase.

IV. CONCLUSIONS

$\text{Tb}_5(\text{Si}_2\text{Ge}_2)$ has more complicated magnetostriction behavior than the related $\text{Gd}_5(\text{Si}_2\text{Ge}_2)$ compound. At low temperatures below 70 K, magnetostriction is small and positive for low fields below 3 kOe, but shows negative slope for fields above 3 kOe. Above 70 K, magnetostriction is positive throughout, but changes from higher slope below 3 kOe to lower slope above 3 kOe. In the region between 100 and 116 K, the magnetostriction shows a magnitude as high as 750 ppm and a usual high field hysteresis region, which moves up in field with increasing temperatures. Beyond 116 K, the hysteretic region can no longer be observed with 20 kOe of an applied magnetic field. In the region from 118 to 150 K, magnetostriction has a simple U shape, with magnitude decreasing with increasing temperature.

This behavior is consistent with the hypothesis that there is a phase transition from one orthorhombic ferromagnetic state to another orthorhombic ferromagnetic state at 70 K upon heating; and in the region from 100 to 116 K there is a first order phase transition, more complex than that of $\text{Gd}_5\text{Si}_2\text{Ge}_2$, which is also affected by an applied magnetic field. The latter phase transition could be that proposed by Morellon *et al.*,⁵ in which the ferromagnetic orthorhombic phase can transform to a ferromagnetic monoclinic phase before transforming to paramagnetic monoclinic phase. It appears that the giant magnetostriction of 750 ppm can be observed because the applied magnetic field is triggering a first order phase transition.

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