Irreversible field induced magnetostriction at temperatures above and below the order-disorder transition in single crystal $Tb_5Si_{2.2}Ge_{1.8}$

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This paper reports on the behavior of single crystal $Tb_5Si_{2.2}Ge_{1.8}$ in the vicinity of its order-disorder and order-order phase transition from a higher temperature paramagnetic/monoclinic state to a lower temperature ferromagnetic/orthorhombic state. Measurements have been made of thermal and field induced changes in strain along the crystallographic *a* axis. The material exhibits large strains of up to 1500 ppm when a magnetic field is applied to it in its paramagnetic state but much smaller strains when a field is applied to it in its ferromagnetic state. These field induced strains are different from conventional magnetostriction because they result mostly from the change in crystal structure. As a result of this the field induced strain changes that accompany the phase transitions of this material are not fully reversible. The shape and slope of the strain versus magnetic field curves were distinctly different depending on whether the material started from above the Curie temperature (where the application of a magnetic field of sufficient strength induced a structural phase transformation) or started from below the Curie temperature (where the application of a field merely stabilized the existing magnetic order). © *2007 American Institute of Physics*.
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I. INTRODUCTION

The present work has investigated the transformation of single crystal $Tb_5Si_{2.2}Ge_{1.8}$ from a high temperature monoclinic state to a low temperature orthorhombic state. This transformation is associated with a large discontinuous change in the *a* axis strain of the sample with temperature. The temperature and field induced strains have been measured over the range of 20–150 K and 0–20 kOe. From the results, which show dramatically different behavior of strain as a function of field above and below the transition temperature, details of the magnetic and structural phase transitions have been inferred.

The $Tb_5Si_xGe_{4-x}$ alloy system is similar to the better known $Gd_5Si_xGe_{4-x}$, except that it has a more complex magnetic and structural phase diagram. The $Gd_5Si_xGe_{4-x}$ system 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. $Tb_5Si_xGe_{4-x}$ also exhibits many of these features. Large changes in strain also accompany the phase transition of single crystal $Tb_5Si_xGe_{4-x}$ from a high temperature paramagnetic/monoclinic state to a low temperature ferromagnetic/orthorhombic state.

There has been some uncertainty over whether the structural transition temperature T^* and the magnetic transition temperature T_C coincide in the Tb alloys.[1–3] The phase diagram published by Ritter *et al.*[3] was based on polycrystalline data and the expected transition temperatures for the alloy composition under investigation here can be determined by interpolation of the data of Ritter *et al.* to be $T_C=115$ K and $T^*=70$ K. Morellon *et al.*[4] also using the data from polycrystalline materials have suggested that ferromagnetism arises in the monoclinic phase at the Curie temperature with a second order phase transformation, and that the first order structural transition from monoclinic to orthorhombic occurs at a lower temperature. Morellon *et al.*[5] suggested on the basis of linear thermal expansion, magnetization, and neutron diffraction studies that under zero stress the Curie temperature $T_C=110$ K and the structural transition temperature $T^*=92$ K for $Tb_5Si_2Ge_2$, but as hydrostatic pressure increases the two transition temperatures gradually converge until they coincide at 115 K under a pressure of 8.6 kbars. In a more recent paper from the same group the structural transition temperature was reported to be $T^*=100$ K.[6]

The present results on single crystal samples of $Tb_5Si_{2.2}Ge_{1.8}$ include both thermal expansion measurements at constant magnetic field and magnetostriction measurements at constant temperature. In both cases the strains were measured along the *a* axis, which is the crystallographic axis that exhibits the largest strain. Thermal expansion measurements, particularly at zero field, give a direct indication of the structural changes occurring in the material. Magnetostriction reflects aspects of both structural and magnetic changes. We rely also on previous data on magnetic susceptibility to confirm the Curie temperature. Present results show that large irreversible changes in strain accompany the structural transition and that these occur at temperatures in the vicinity of 106 K. Although these results do not prove

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definitively that T_C and T^* coincide, they do show that the two temperatures are much closer than was previously anticipated.

II. EXPERIMENTAL DETAILS

Single crystal $Tb_5Si_{2.2}Ge_{1.8}$ was produced via the Bridgman method.[7] A clean, planar surface containing the crystallographic *a* and *b* axes was produced via electron discharge cutting. This surface was prepared with ethanol and a strain gauge was bonded to the surface to measure strain along the *a* axis. The sample was mounted on a copper block using a thermally conducting silver epoxy. A cernox temperature sensor was mechanically attached to this block with a screw together with thermally conducting paste. The copper block was mounted using the same thermally conducting paste to the cold finger of a closed cycle helium refrigeration system. The sample was oriented so that the applied field was along the *a* axis, parallel to the direction of strain measurement.

The magnetic field was applied using an electromagnet with a computer programmable power supply. A gaussmeter was used to measure the magnetic field. 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. The cryogenic system with the sample mounted inside was pumped down to 10^{-7} torr and the system was cooled to 14 K. Measurements were made of λ vs *H* for samples that had been (i) cooled from a temperature above T_C or (ii) heated from a temperature below T_C after thermal demagnetization. A comparison was made of the behavior of the material in these two regimes.

III. RESULTS AND DISCUSSION

Magnetization data, particularly susceptibility, are suitable for distinguishing between magnetic structures such as paramagnetic and ferromagnetic orders and the Curie temperature. Our earlier results[7] on magnetic susceptibility led us to the conclusion that T_C=110 K, which is consistent with the present results.

A. Thermal expansion: Effect of applied field on the phase transition

Thermal expansion measurements at different fixed field strengths are shown in Fig. 1. The large temperature induced strains should occur when the material is transformed from a monoclinic to an orthorhombic structure. If we therefore assume that the high slope $(d\lambda/dT)_{\max}$ corresponds to the structural transition, the present results suggest that at zero applied field this occurs at a temperature T^* of between 106 and 110 K, depending on whether the temperature is increasing or decreasing. With the application of a magnetic field along the *a* axis the transition temperature T^*, as determined by the maximum rate of change of strain as a function of temperature $(d\lambda/dT)_{\max}$, increased with the strength of the magnetic field *H*. The change in transition temperature with applied field appeared to be nonlinear, as shown in Fig. 2. For example, a change in transition temperature of 1.5° from 108.6 to 110.1 K occurred when the applied field was changed from 0 to 10 kOe, whereas an increase of 5.5° from 110.1 to 115.6 K was caused by an equal increase in field from 10 to 20 kOe. The transition temperature varies with field according to the relation $T^*=a+bB+cB^2$, where $a=108.5$ K, $b=0$, and $c=1.75$ K^{-2}.

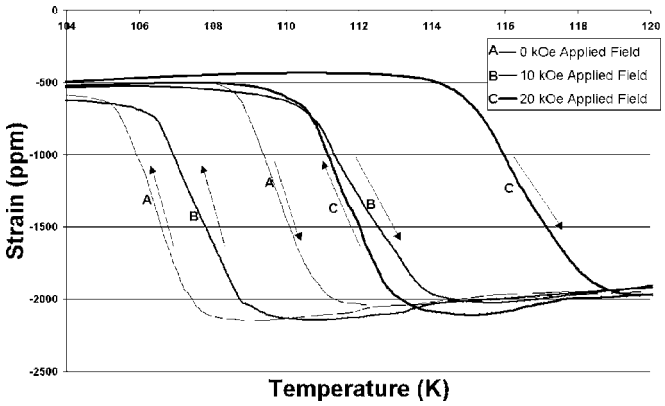

FIG. 1. Temperature dependence of strain measured along the *a* axis for single crystal $Tb_5Si_{2.2}Ge_{1.8}$ at 0, 10, and 20 kOe applied field.

B. Magnetostriction: Effect of temperature on the phase transition

Magnetostriction measurements were made at fixed temperatures of 114.5, 112.5, 110.6, 108.6, 105.8, 103.0, and 97.1 K in order to examine the behavior of the properties above and below T_C. Measurements of strain versus applied field were made at temperatures above T_C after the material was cooled from well above T_C to near T_C. The results are shown in Fig. 3. At 114.5 K, the material was too far above the transition temperature so that no field induced transition took place even for fields of 20 kOe. At 112.5 and 110.6 K, the magnetostriction curves show that the transition from monoclinic to orthorhombic can be caused by the application of a magnetic field, but the material was still close enough to T^* that it returned to a monoclinic crystal structure on the removal of the applied field. More field was required to cause the transition at 112.5 K than at 110.6 K. At 108.6 K, there was insufficient thermal energy to cause a reversion from orthorhombic to monoclinic on the removal of the field.

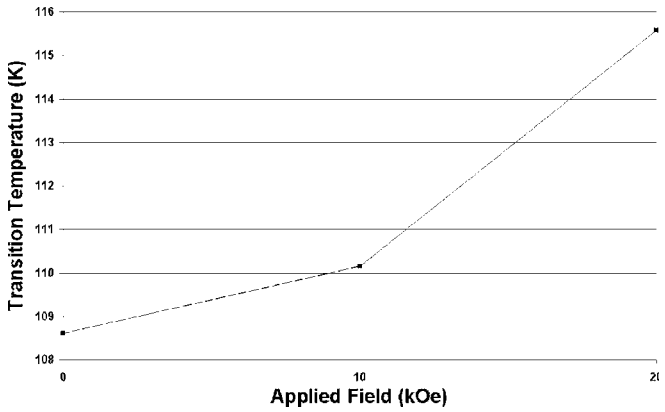

FIG. 2. Variation of the transition temperature T^* for single crystal $Tb_5Si_{2.2}Ge_{1.8}$ as determined from the maximum slope $(d\lambda/dT)_{\max}$.

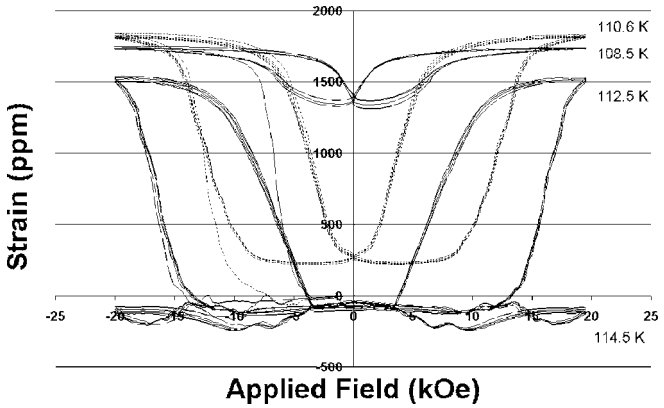

FIG. 3. Magnetostrictive strain vs applied field, both measured along the *a* axis for single crystal $Tb_5Si_{2.2}Ge_{1.8}$ at various temperatures above the transition temperature T_C.

Magnetostriction measurements were also made at temperatures from well below T_C to near T_C. Commonly occurring "butterfly-shaped" magnetostriction curves were observed, as shown in Fig. 4, with strain amplitudes of 200 ppm or less. The amplitude decreased with decreasing temperature. The results of the tests shown in Fig. 4 have similarities to those obtained in previous studies.[2]

IV. CONCLUSIONS

The large thermal expansion and magnetostrictive strains arose from changes in either the crystal structure or the magnetic state, respectively. The steepness of the rate of change of strain versus temperature, its hysteresis, and the temperature dependence of strain versus field are indicative of a first order phase transition, which can be activated either by an applied magnetic field or a change in temperature. The application of a magnetic field increases the transition temperature, so that when the sample was already below the zero field transition temperature the application of a magnetic field only had the effect of stabilizing the existing magnetic structure.

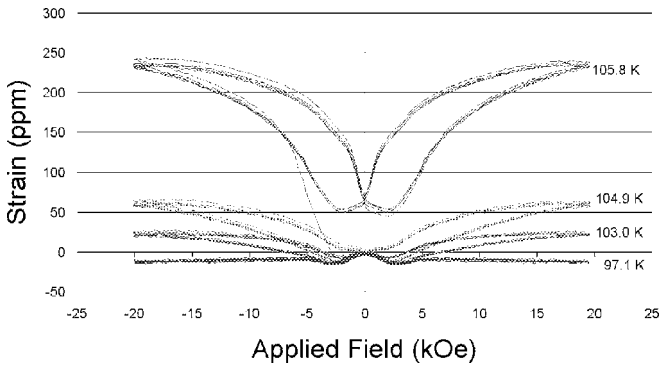

FIG. 4. Magnetostrictive strain vs applied field, both measured along the *a* axis for single crystal $Tb_5Si_{2.2}Ge_{1.8}$ at various temperatures below the transition temperature T_C.

At some temperatures, above T^* but sufficiently close to T^*, the application of a magnetic field causes a structural change from monoclinic to orthorhombic, together with the associated magnetic change from paramagnetic to ferromagnetic. The subsequent removal of the magnetic field may result in a reversion to the paramagnetic state, if the temperature is sufficiently high, or the material may not revert if the temperature is sufficiently close to the zero field Curie temperature.

In most magnetic materials the Curie point transition is independent of field strength. $Tb_5Si_xGe_{4-x}$ is unusual in that the order-disorder transition is of the first order and therefore over at least part of the composition range these alloys exhibit hysteresis and field dependence of the structural and magnetic transitions that is rarely seen. An applied magnetic field will induce the first order structural transition, which is detected by the strain that accompanies crystallographic change, to occur at a higher temperature. This effect occurs both while sweeping temperature at a fixed applied field strength (thermal expansion) and while holding temperature steady and sweeping the applied magnetic field (magnetostriction).

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