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Giant magnetostriction behavior at the Curie temperature of single crystal Gd5(Si0.5Ge0.5)4

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Abstract

We report results of thermal expansion (TE) and magnetostriction (MS) measurements on a single crystal sample of $Gd_5(Si_{0.5}Ge_{0.5})_4$ prepared by the Bridgman method. TE and MS were measured along the *c* axis by the strain gauge method and the temperature was controlled using a closed cycle helium refrigerator. From the TE measurements the magnetic structural phase transition temperature was found to be 259.5 K on cooling and 261.5 K on heating. The abrupt change in strain and the temperature hysteresis indicate that it is a first order transition. MS measurements were conducted at 15, 258, and 265 K. At 15 K, the magnetostriction amplitude was 3–4 ppm, whereas at 258 K it was 100 ppm. At 265 K, which is just above the Curie temperature, a giant magnetostriction of 2000 ppm was found. This unusual behavior is due to the fact that the external magnetic field can increase the transition temperature above 265 K, resulting in a first order magnetic/structural phase transition. The results reveal that giant magnetostriction in$Gd_5(Si_{0.5}Ge_{0.5})_4$ only occurs as a result of the magnetic/structural transformation.

Keywords

Materials Science and Engineering, gadolinium alloys, Ge-Si alloys, crystal growth from melt, thermal expansion, magnetostriction, Curie temperature, magnetic transitions, ferromagnetic materials, paramagnetic materials, solid-state phase transformations, ferromagnetic-paramagnetic transitions

Disciplines

Condensed Matter Physics | Metallurgy

Comments

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Giant magnetostriction behavior at the Curie temperature of single crystal $Gd_5(Si_{0.5}Ge_{0.5})_4$

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We report results of thermal expansion (TE) and magnetostriction (MS) measurements on a single crystal sample of $Gd_5(Si_{0.5}Ge_{0.5})_4$ prepared by the Bridgman method. TE and MS were measured along the *c* axis by the strain gauge method and the temperature was controlled using a closed cycle helium refrigerator. From the TE measurements the magnetic structural phase transition temperature was found to be 259.5 K on cooling and 261.5 K on heating. The abrupt change in strain and the temperature hysteresis indicate that it is a first order transition. MS measurements were conducted at 15, 258, and 265 K. At 15 K, the magnetostriction amplitude was 3–4 ppm, whereas at 258 K it was 100 ppm. At 265 K, which is just above the Curie temperature, a giant magnetostriction of 2000 ppm was found. This unusual behavior is due to the fact that the external magnetic field can increase the transition temperature above 265 K, resulting in a first order magnetic/structural phase transition. The results reveal that giant magnetostriction in $Gd_5(Si_{0.5}Ge_{0.5})_4$ only occurs as a result of the magnetic/structural transformation. © *2004 American Institute of Physics.*
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I. INTRODUCTION

$Gd_5(Si_xGe_{1-x})_4$ has been found to exhibit a giant magnetocaloric effect.[1] Within the composition range $0.4 \leqslant x \leqslant 0.503$, it was found that there exists an unusual first order magnetic/structural phase transition at T_c. Above T_c the material is paramagnetic with a monoclinic crystal structure. Below T_c, it is ferromagnetic with an orthorhombic crystal structure. In addition to temperature, external magnetic field[2,3] and pressure[4,5] can also trigger this transition, by shifting T_c. For example, linear thermal expansion measurements on single crystal $Gd_5(Si_{1.95}Ge_{2.05})$ have shown T_c to increase with applied magnetic field at a rate of 4.9 K/T.[3] On the basis of thermal expansion measurements on single crystal $Gd_5(Si_{1.72}Ge_{2.28})$, the change in T_c with pressure has been predicted and in fact, predicted to be quite different for uniaxial pressure along the three different principal axes.[4] And T_c of polycrystalline $Gd_5(Si_{1.8}Ge_{2.2})$ has been observed to increase with hydrostatic pressure at the rate of 3.79 K/kbar for decreasing temperature and 3.46 K/kbar for increasing temperature.[5] In this article, we present a study of the thermal expansion and magnetostriction properties for $Gd_5(Si_{0.5}Ge_{0.5})_4$ in single crystal form. Thermal expansion results have been reported previously for other compositions.[2,3,6]

II. EXPERIMENTAL DETAILS

A single crystal of $Gd_5(Si_{0.5}Ge_{0.5})_4$ with dimensions 4 mm×4 mm×4 mm was grown by the Bridgman method. Appropriate quantities of high purity gadolinium (99.996%), silicon (99.9999%), and germanium (99.999%) were cleaned and arc melted several times under an argon atmosphere. The as-cast ingot was electron beam welded into a tungsten Bridgman style crucible for crystal growth. The ingot was then heated to 2000 °C and held at this temperature for 1 h to allow thorough mixing before withdrawing the sample from the hot zone at a rate of 4 mm/h. The as-grown crystal was oriented by back-reflection Laue x-ray diffraction and the crystallographic directions assigned using ‘‘two-theta’’ x-ray diffraction scans of the single crystal. The sample was cut by electric discharge machining and the faces were polished using standard metallographic techniques.

Thermal expansion measurements were conducted using strain-gauges in a two-stage closed cycle helium refrigera-

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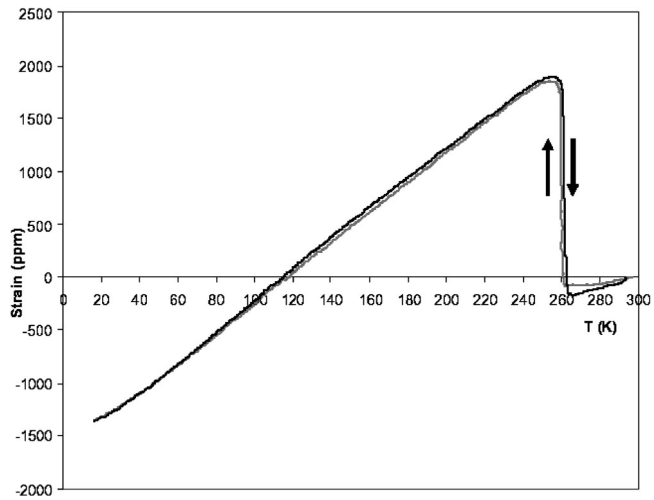

FIG. 1. Thermal expansion along the *c* axis for $Gd_5(Si_{0.5}Ge_{0.5})_4$ with $B=0$ T, on cooling $T_c=259.5$ K, and on heating $T_c=261.5$ K.

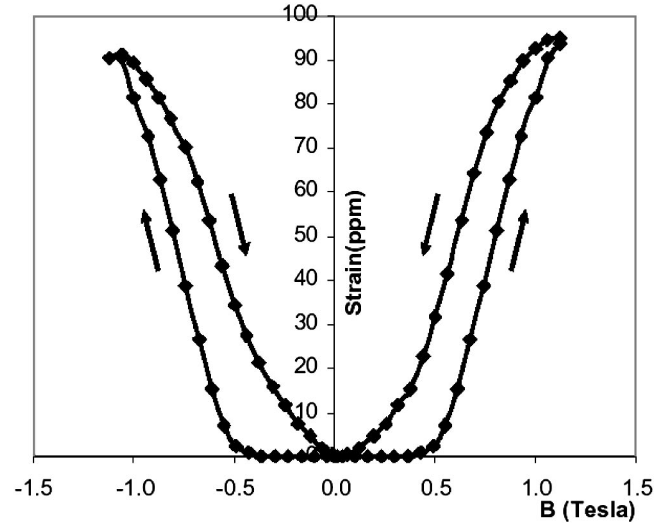

FIG. 2. Magnetostriction measurement along the *c* axis for $Gd_5(Si_{0.5}Ge_{0.5})_4$ at $T=258$ K.

tion system with temperature range 10–310 K. The strain gauge was mounted along the *c* axis using Permabond 910 adhesive which was cured at room temperature. The sample was cooled from room temperature through the phase transition down to 15 K, and then was warmed through the transition for each measuring cycle. The linear thermal expansion was measured during both cooling and heating. Measurements were made either when the strain changed by 7 ppm or when the temperature changed by 1 K. The transition temperature was determined by finding the maximum point in the derivative of strain versus temperature. For the magnetostriction measurement, the magnetic field was applied along the *c* axis, and the field was cycled from 0 to 1.2 T to -1.2 T to 0, and from 0 to 2.5 T to -2.3 T to 0 under constant temperatures $T=15$ and 258 K. The magnetostriction measurement was also repeated at 265 K with magnetic field varying over the range from -2.3 to 2.3 T.

III. RESULTS AND DISCUSSION

Thermal expansion along the *c* axis with $B=0$ T is shown in Fig. 1. An abrupt change in strain of about 2000 ppm appears at $T=259.5$ K on cooling, and at $T=261.5$ K on heating. It has been verified that this is a first order magnetic/structural transformation.[2,3] At the transition point, there are two transformations occurring simultaneously. One is the crystalline structural transition: monoclinic ↔ orthorhombic. The other is the magnetic transition: paramagnetic ↔ ferromagnetic. In most magnetic materials, it is well known that the Curie point transition is a second order transition. However, an abrupt change in strain at the transition with a temperature hysteresis about 2 K as shown in Fig. 1, indicates that this is a first order transition. The existence of a first order transition has been reported previously in the $Gd_5(Si_xGe_{1-x})_4$ system[7] when x is in the range of $0.4<x<0.503$. The change in strain at the magnetic/structural phase transition is highly anisotropic. The change in strain along the *a* axis of single crystal $Gd_5(Si_{1.95}Ge_{2.05})$ was observed to be about 8000 ppm and of opposite sign (increasing with increasing temperature).[3] Similar results (magnitudes and signs) have been reported for single crystal $Gd_5(Si_{1.72}Ge_{2.28})$ measured along its three principal crystallographic axes.[4]

Magnetostriction measurements were carried out at $T=15$ K over the ranges $B=\pm1.2$ T and $B=\pm2.4$ T. The magnetostriction was found to be only about a few parts per million (ppm). At $T=15$ K, the exchange energy between Gd atoms is much greater than the thermal energy and therefore the magnetic moments are well aligned. Therefore, the field-induced magnetostriction amplitude is very small. As reported previously, the external magnetic field increases the first order magnetic/structural transition temperature. In this case for $B=2.5$ T, $T_c=272$ K. Therefore at 15 K application of a magnetic field can not trigger the first order magnetic/structural transition.

When a magnetic field was applied in order to measure the magnetostriction, T_c increased at a rate of 4.9 K/T. Magnetostriction measurements are shown in Fig. 2 at $T=258$ K, which is just below the Curie temperature of 259.5 K in zero applied field. When the field was cycled between -1.2 and 1.2 T, the thermodynamic temperature always remained below T_c and therefore no structural transition occurred. In this case the observed magnetostriction was only due to domain reorientation. However, when a magnetic field was applied for a sample above T_c, for example at $T=265$ K, as shown in Fig. 3, the value of T_c increased from 259.5 K by an amount depending on the applied field strength. When $T_c=265$ K, a field induced first order transition occurred and there was a significant change in strain of 2000 ppm. This magnetostriction was due to the two simultaneous transitions: paramagnet–ferromagnet and monoclinic–orthorhombic. Therefore, it can be expected that a larger magnetostriction can be observed for the *a*-axis direction, for which a change in strain at the phase transition of about 8000 ppm has been observed.[3,4] Irreversible effects are noticeable as asymmetry in the strain versus field plot. These effects are probably due to the build up of residual stress in

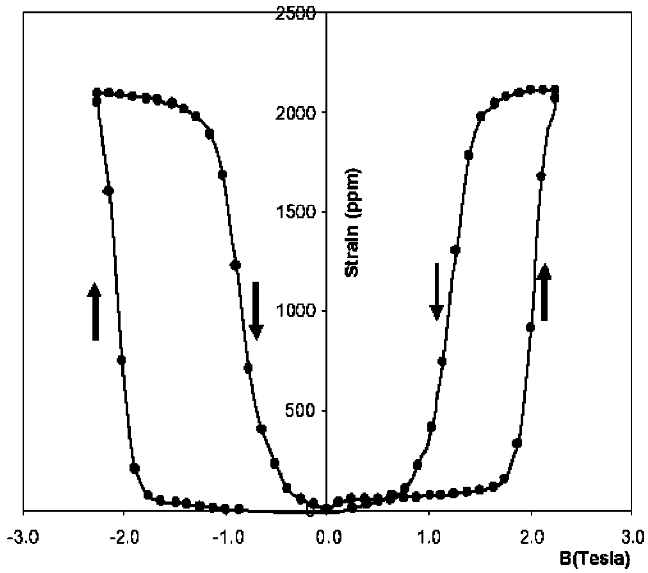

FIG. 3. Magnetostriction measurement along the c axis for $Gd_5(Si_{0.5}Ge_{0.5})_4$ at $T = 265$ K.

the sample as the field was cycled and the material underwent the structural phase transformation.

Similar results have been found for $Gd_5(Si_{0.1}Ge_{0.9})_4$ by Morellon *et al.*[6] $Gd_5(Si_{0.1}Ge_{0.9})_4$ exhibits a first order transition at $T_c = 81$ K (when $B = 0$ T), and a second order transition at $T_N = 127$ K. Below its Curie temperature, it was found there was no significant magnetostriction in applied fields of up to 12 T. At $T = 110$ K, which is above T_c within the antiferromagnetic regime, it required an 8 T applied magnetic field in order to produce giant magnetostriction.

IV. CONCLUSIONS

Thermal expansion measurements have been conducted to verify the first order transition for a single crystal sample of $Gd_5(Si_{0.5}Ge_{0.5})_4$, which was found to occur at $T_c = 259.5$ K on cooling and $T_c = 261.5$ K on heating. Magnetostriction measurements have been carried out above and below T_c. It was found that at $T = 15$ K, very little magnetostriction was observed due to the absence of a field induced structural transformation. At $T = 258$ K, a magnetostriction amplitude of about 100 ppm was observed. At $T = 265$ K giant magnetostriction was observed, with an amplitude of about 2000 ppm. This observation was due to the fact that the applied magnetic field can shift the T_c above 265 K and trigger a simultaneous magnetic/structural transformation with a high level of strain resulting from the structurally induced strain rather than from a reorientation of magnetic domains.

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