Fine structure observation in magnetostriction near the transition temperature in Gd₅Si₁.₉₅Ge₂.₀₅

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Fine structure observation in magnetostriction near the transition temperature in Gd5Si1.95Ge2.05

Abstract
Gd5(Si\textsubscript{x}Ge\textsubscript{1-x})\textsubscript{4} has a complex magnetic-structural phase diagram which can be divided into three distinct regions. It exhibits an unusual first-order coupled magnetic-structural phase transition in the region \textsubscript{x} < 0.51. A series of magnetostrictive strain measurements were carried out as a function of magnetic field strength at different temperatures and as a function of temperature at near-zero magnetic field strengths. In this paper, we report for the first time the observation of fine structure in the variation of strain with magnetic field near the first-order phase transition temperature. This fine structure was observed only for the single-crystalline and polycrystalline samples of Gd5 Si 1.95 Ge2.05 but not for Gd5 Si2 Ge2 and Gd5 Si2.09 Ge1.91 samples. There was a sudden increase of about 200-300 ppm in the magnetostrictive strain just prior the field-induced first-order phase transition. In this paper, this anomaly is termed as fine structure. It was observed in measurements of both magnetostrictive strain versus magnetic field and magnetostrictive strain versus temperature. In the case of the polycrystalline Gd5Si 1.95Ge2.05 sample, this anomaly was not as sharp, and the sudden magnetostrictive strain change was about 40 ppm just before the field-induced first-order phase transition.

Keywords
first-order phase transition, Gd5 (Si\textsubscript{x}Ge\textsubscript{1-x})\textsubscript{4}, magnetocaloric effect, magnetostriction, Ames Laboratory, US Department of Energy

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Fine structure observation in magnetostriction near the transition temperature in Gd_{5}Si_{1.95}Ge_{2.05}

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Gd_{5}(Si,Ga)_{12} has a complex magnetic-structural phase diagram which can be divided into three distinct regions. It exhibits an unusual first order coupled magnetic-structural phase transition in the region 0.41 \leq x \leq 0.51. A series of magnetostrictive strain measurements were carried out as a function of magnetic field strength at different temperatures and as a function of temperature at near zero magnetic field strengths have been carried out. In this paper we report for the first time the observation of fine structure in the variation of strain with magnetic field near the first order phase transition temperature. This fine structure was observed only for the single crystalline and polycrystalline samples of Gd_{5}Si_{1.95}Ge_{2.05} but not for Gd_{5}Si_{2}Ge_{2} and Gd_{5}Si_{2.09}Ge_{1.91} samples. There was a sudden increase of about 200-300 ppm in the magnetostrictive strain just prior to the field induced first order phase transition. In this paper this anomaly is termed as fine structure. It was observed in measurements of both magnetostrictive strain versus magnetic field and magnetostrictive strain versus temperature. In the case of the polycrystalline Gd_{5}Si_{1.95}Ge_{2.05} sample this anomaly was not as sharp and the sudden magnetostrictive strain change was about 40 ppm just before the field induced first order phase transition.

Index Terms—First order phase transition, Gd_{5}(Si,Ga)_{12}, Magnetocaloric effect, Magnetostriction.

I. INTRODUCTION

Various studies have been conducted on Gd_{5}(Si,Ga)_{12} due to its giant magnetocaloric effect near room temperature. It exhibits one of the largest room temperature giant magnetocaloric effect close to its first order phase transition temperature [1, 2]. It also exhibits a colossal magnetostriction of the order of 10,000 ppm [3-5] and a giant magnetoresistance (AR/R) of the order of 25% [6, 7] close to the first order phase transition temperature for x=0.5. The room temperature giant magnetocaloric effect can be utilized for energy efficient refrigeration. The energy conversion efficiency of these refrigerators can reach as high as 60% of Carnot efficiency which is twice the efficiency of normal liquid/vapor refrigerators [8].

Gd_{5}(Si,Ga)_{12} has a complex phase diagram which can be divided into three main regions with distinct transition temperatures and two small regions that exhibit two-phase behavior, where the transition temperature is not distinct. The three main regions can be divided on the basis of their crystallographic structures at room temperature. The Sm_{5}Ge_{4} type structure occurs for the composition 0 \leq x \leq 0.31, Gd_{5}Si_{4}Ge_{2} type structure occurs for the composition 0.41 \leq x \leq 0.51 and Gd_{5}Si_{4} type structure occurs for the composition 0.575 \leq x \leq 1 [9]. Region I with the composition 0 \leq x \leq 0.31 has two kinds of transitions, first order ferromagnetic orthorhombic (Gd_{5}Si_{4}) to antiferromagnetic orthorhombic (Sm_{5}Ge_{4}) at lower temperatures and second order antiferromagnetic to paramagnetic at higher temperatures. Region II with the composition 0.41 \leq x \leq 0.51 has an unusual first order phase transition from ferromagnetic orthorhombic (Gd_{5}Si_{4}) to paramagnetic monoclinic. Region III with the composition 0.575 \leq x \leq 1 has a second order phase transition from ferromagnetic orthorhombic (Gd_{5}Si_{4}) to paramagnetic orthorhombic (Gd_{5}Si_{4}) [10]. Region II exhibits the largest magnetocaloric effect hence it is the most researched region in the phase diagram.

In this paper we report the observation of anomaly close to the first order phase transition in magnetostriction for the composition Gd_{5}Si_{1.95}Ge_{2.05} (which is in region II of the phase diagram). This anomaly near the phase transition is termed as fine structure. Magnetostrictive strain as a function of magnetic field for different temperatures and magnetostrictive strain as a function of temperature for constant magnetic field strengths were measured for single crystalline and polycrystalline samples of Gd_{5}Si_{1.95}Ge_{2.05} (x=0.487). Similar measurements were carried out on single crystalline Gd_{5}Si_{2}Ge_{2} (x=0.5) and polycrystalline Gd_{5}Si_{2.09}Ge_{1.91} (x=0.522) samples but they did not show any fine structure in the measurements.

II. SAMPLE PREPARATION

Single crystalline samples of Gd_{5}Si_{1.95}Ge_{2.05} and Gd_{5}Si_{2}Ge_{2} were prepared at Ames Laboratory, US Department of Energy [11] by the Tri-arc pulling method using 99.996 % pure gadolinium (weight basis), 99.9999 % pure silicon (weight basis) and 99.999 % germanium (weight basis). The ingot was pulled at a rate of 4 mm/hour. The crystal was subsequently heat treated at 1273 K for 24 h to reduce or eliminate the amount of orthorhombic phase present. The sample was then slow cooled at a rate of 10 degrees/minute from the annealing temperature to produce a more phase-pure sample. This ingot was cut using an electric discharge machine (EDM) to limit any stress on the final cut sample. The sample was indexed using Laue X-ray diffraction technique. The polycrystalline...
Gd$_{5}$Si$_{1.95}$Ge$_{2.05}$ and the polycrystalline Gd$_{5}$Si$_{2.09}$Ge$_{1.91}$ samples were also prepared at Ames laboratory by the arc melting. The initial materials used for the Gd$_{5}$Si$_{1.95}$Ge$_{2.05}$ sample were of the same purity mentioned above. For the Gd$_{5}$Si$_{2.09}$Ge$_{1.91}$ sample the material used was commercial grade gadolinium (99.9% pure by weight) and 99.999% pure silicon (weight basis) and 99.99% germanium (weight basis). The polycrystalline samples were heat treated similarly to the single crystalline samples and their crystal structure was confirmed with XRD technique.

### III. EXPERIMENTAL DETAILS

Magnetostrictive strain was determined by with the strain gauges mounted on the samples. Temperature and magnetic field control were achieved using a Quantum Design Physical Property Measurement System (PPMS). Vishay Micro-Measurement WK-06-031CF-350 strain gauges were used which have a resistance of 350 ohms and a gauge factor of 2.05 (± 1.0%). The strain gauges were mounted on the sample using M-Bond 610 adhesive which has an operational temperature range of 4 K to 643 K. The strain gauge-mounted samples were then heated for about 5 hours at 350 K to cure the adhesive. A quarter Wheatstone bridge was built using high tolerance 350 ohms resistors. Alignment of strain gage to the axes is not taken into consideration.

### IV. RESULTS AND DISCUSSION

The single crystalline Gd$_{5}$Si$_{1.95}$Ge$_{2.05}$ has a first order phase transition from high temperature monoclinic-paramagnetic phase to low temperature orthorhombic ferromagnetic phase at 255 K at nearly zero field. The material also exhibits field induced first order phase transition from monoclinic-paramagnetic at low field to orthorhombic ferromagnetic at high field. Magnetostrictive strain as a function of magnetic field strength was measured using the PPMS (Fig. 1) for the single crystalline Gd$_{5}$Si$_{1.95}$Ge$_{2.05}$ sample at 275 K. The magnetic field strength was varied from 0 A/m to 5.57 MA/m, 5.57 MA/m to -5.57 MA/m, -5.57 MA/m to 5.57 MA/m and 5.57 MA/m to 0 A/m. The material at 275 K is monoclinic-paramagnetic, when sufficient field is applied it transforms to orthorhombic-ferromagnetic as shown in Fig. 1. At temperature 275 K the field needed to induce the first order phase transition was about 3.18 MA/m for the forward field variation. In all these parts of measurements there was a sudden increase in the magnetostrictive strain just before the onset of the first order phase transition. Fig. 2 shows the magnetostrictive strain as a function of magnetic field strength for various temperatures for single crystalline Gd$_{5}$Si$_{1.95}$Ge$_{2.05}$ sample above its transition temperature. Note the sudden increase in the magnetostrictive strain of the order of 200-300 ppm near the field induced first order phase transition. There is a field induced first order phase transition at a critical field for all isotherms. The amount of field required to induce the transition increases with increasing temperature as reported in literature [12].
Fig. 3 shows magnetostrictive strain as a function of temperature at near zero magnetic field strength for single crystalline Gd$_5$Si$_{1.95}$Ge$_{2.05}$. It shows a sudden increase in the strain of the order of 200 ppm close to its first order phase temperature. Fig. 4 shows magnetostrictive strain as a function of magnetic field strength at various temperatures for a polycrystalline Gd$_5$Si$_{1.95}$Ge$_{2.05}$ sample. For the polycrystalline sample the sudden increase in the strain is of the order of 40 ppm which is not as high as for the single crystalline sample due to the anisotropy of strain change related to grain orientations.

Fig. 5 and Fig. 6 show magnetostrictive strain as a function of temperature at nearly zero field and as a function of magnetic field strength for different temperatures for a single crystalline Gd$_5$Si$_2$Ge$_2$ sample respectively. Fig. 7 and Fig. 8 show magnetostrictive strain as a function of temperature and as a function of magnetic field strength for different temperatures for a polycrystalline Gd$_5$Si$_{2.09}$Ge$_{1.91}$ sample.
It can be seen from Figs. 5-8 that unlike the single crystalline Gd$_5$Si$_{1.95}$Ge$_{2.05}$ (Figs. 1-3) and polycrystalline sample (Fig. 4) in these measurements there is no sudden increase in the strain close to the transition temperature. Out of the four samples measured, the anomaly in the magnetostrictive strain curves was observed only for the composition Gd$_5$Si$_{1.95}$Ge$_{2.05}$ ($x=0.487$) in both single crystalline and polycrystalline samples. The anomaly was observed both for magnetostrictive strain versus magnetic field strength and magnetostrictive strain versus temperature measurements, indicating that it is likely not the result of an experimental error in one type of measurement. However this anomaly was not observed when single crystalline Gd$_5$Si$_2$Ge$_2$ ($x=0.5$) and polycrystalline Gd$_5$Si$_{2.98}$Ge$_{1.91}$ ($x=0.522$) samples were measured for both kinds of measurements using the same set of apparatus. This indicates that there is a fine structure in the magnetostrictive curve for the composition Gd$_5$Si$_{1.95}$Ge$_{2.05}$ ($x=0.487$) near its first order phase transition temperature. An extensive search in literature show a similar fine structure in lattice parameters measurements near the first order phase transition along all three major axes, ‘a’, ‘b’ and ‘c’ in the composition Gd$_{5}$(Si$_{0.4}$Ge$_{3.6}$) [13]. Magnetostriction measurements on single crystalline Tb$_5$Si$_{2.2}$Ge$_{1.8}$ also show similar fine structures near the first order phase transition [14].

We suggest that this fine structure observation in the magnetostrictive strain curves for Gd$_5$ Si$_{1.95}$Ge$_{2.05}$ ($x=0.487$) is indicative of differences in switching field strengths for different regions of the material which could be due to the presence of different microscopic inhomogeneous phase or Griffith’s phase. The presence of secondary phase in samples lead to variation in the transition temperature or critical field in field induced phase transition depending on the amount and distribution of the secondary phases. If the secondary phase is evenly distributed in the sample in larger quantities, the global average transition temperature of the sample is different from the single phase material. The transition temperature may not vary if the amount of secondary phase is low and is not distributed evenly. When the secondary phase is large and evenly distributed in the sample and if pure phase occurs locally over a larger grain in the sample, apart from showing variation in transition temperature, the sample will also show anomalies in the measurements. This secondary phase has been referred to as Griffiths-like phase in literature [15] and has been reported in Gd$_5$(Si$_{1}$Ge$_{1}$)$_{3}$ for $x=0.1$ and Tb$_5$(Si$_{1}$Ge$_{1}$)$_{3}$ for $x=0.5$ [16] and other similar systems recently [17]. Presence of Griffiths-like phase in a material depends on material preparation method hence some samples may show Griffiths phase while others samples even with the same composition may not show if they are prepared by different methods. First principles investigations are needed to confirm the presence of this phase in Gd$_5$(Si$_{1}$Ge$_{1}$)$_{3}$.

V. CONCLUSION

Fine structure was observed in the magnetostrictive strain as a function of magnetic field strength and temperature near its first order phase transition. This fine structure in magnetostriction was of the order of 200-300 ppm for single crystalline and 40 ppm for polycrystalline Gd$_5$Si$_{1.95}$Ge$_{2.05}$ ($x=0.487$) samples. Observation of this fine structure in the composition Gd$_5$Si$_{1.95}$Ge$_{2.05}$ ($x=0.487$) is indicative of differences in switching field strengths for different regions of the material which could be due to the presence of different phases in the sample.

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REFERENCES


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