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# Field induced structural phase transition at temperatures above the Curie point in $Gd_5(Si_xGe_{1-x})_4$

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$Gd_5(Si_xGe_{1-x})_4$ exhibits a field induced first order phase transition from a monoclinic paramagnetic to an orthorhombic ferromagnetic at temperatures above its Curie temperature for $0.41 \leq x \leq 0.51$. The field required to induce the transition increases with temperature. This field induced first order phase transition was observed even above the projected second order phase transition temperature of the orthorhombic phase. This may be due to the fact that the applied magnetic field is so high that it causes the broadening to a wider range of higher temperatures of the second order phase transition of the orthorhombic phase, and at such high magnetic fields the magnetic moment is still quite large, therefore, causing the transition. This hypothesis seems to be confirmed by the various magnetic moment versus magnetic field, magnetic moment versus temperature, and strain versus magnetic field measurements carried out on single crystal $Gd_5Si_{1.95}Ge_{2.05}$ and $Gd_5Si_2Ge_2$ samples at magnetic fields of 0–9 T and at temperatures of 265–305 K. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3076419]

## I. INTRODUCTION

$Gd_5(Si_xGe_{1-x})_4$ has been called as an "extremum" material for its unusual properties such as giant magnetocaloric effect, colossal magnetostriction, and giant magnetoresistance near its first order (FO) phase transition (PT). An adiabatic temperature change of 17 °C and an entropy change of 20 J $kg^{-1}$ $K^{-1}$ can be achieved upon application of a magnetic field of 5 T.[1] An entropy change of 14.5 J $kg^{-1}$ $K^{-1}$ can be achieved for a lower magnetic field of 2 T.[2] The material also exhibits a magnetostriction of the order of 10 000 ppm (Ref. 3) and a magnetoresistance ($\Delta R/R$) of 25% near its FO PT temperature.[4,5]

The phase diagram of $Gd_5(Si_xGe_{1-x})_4$ can be divided into three regions depending on the value of $x$.[6] The central region with composition of $0.41 \leq x \leq 0.51$ and Si-rich region with composition of $0.57 \leq x \leq 1.0$ are focal points of our investigation. In the Si-rich region, the material exhibits a second order (SO) PT from orthorhombic ferromagnetic to orthorhombic paramagnetic at the Curie temperature $T_C$. For a sample from the central region, however, at transition temperature $T_{FO}$, the PT is a FO magnetic-structural PT: in which the low temperature orthorhombic ferromagnetic phase changes to high temperature monoclinic paramagnetic phase. It is in this region that the exceptional magnetocaloric effect, magnetostriction, and magnetoresistance occur.

In this region, if the thermodynamic temperature $T$ is higher than the transition temperature $T_{FO}$ ($H_{app}=0$), the field induced FO PT will occur under a sufficiently high magnetic field. The magnetic field needed to induce the field induced FO PT increases with thermodynamic temperature $T$. The inverse effect is also true; in plots of magnetic moment versus temperature, for different constant magnetic fields the transition temperature $T_{FO}$ increases with the applied magnetic field at a rate of 5 K/T.[7]

It is reasonable to expect that the orthorhombic phase has a Curie temperature $T_C^*$ as well. However this temperature $T_C^*$ cannot be directly measured as the FO PT occurs at lower temperatures, $T_{FO} < T_C^*$ but it still can be estimated by means of Arrott plot techniques.[8] It was hypothesized that for high enough temperatures, $T > T_C^*$, the field induced FO PT could not occur even for very high applied fields because the orthorhombic phase would already be paramagnetic at that temperature. In this paper we present a study of the field induced PT at high magnetic fields and high temperatures for the single crystals $Gd_5Si_{1.95}Ge_{2.05}$ and $Gd_5Si_2Ge_2$.

## II. SAMPLE PREPARATION AND EXPERIMENTAL DETAILS

Single crystal samples of $Gd_5Si_{1.95}Ge_{2.05}$, $Gd_5Si_2Ge_2$, and $Gd_5Si_{2.7}Ge_{1.3}$ were prepared at Ames Laboratory, U.S. Department of Energy by the Bridgman method using 99.996% pure gadolinium (weight basis), 99.9999% pure silicon (weight basis), and 99.999% germanium (weight basis). The samples were then annealed at 2000 °C for 1 h to remove the residual secondary phases in the material.

Transition temperatures $T_{FO}$ and $M$ versus $H$ curves for single crystal $Gd_5Si_{1.95}Ge_{2.05}$ and $Gd_5Si_2Ge_2$ samples were measured in a Quantum Design magnetic properties measurement system (MPMS) under an applied magnetic field of 8 kA/m. Strain versus magnetic field measurements were carried out in a Quantum Design physical properties measurement system. Additionally, the higher temperature $M$-$H$ isotherms above 296 K (i.e., above the SO PT temperature $T_C^*$ of the orthorhombic phase) were measured on a 9 T vibrating sample magnetometer (VSM) at the Sheffield University, UK.

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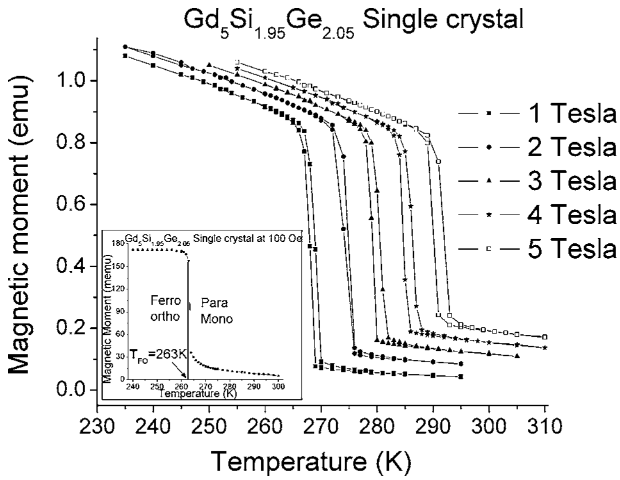


FIG. 1. *M* vs *T* at various values of applied magnetic field for single crystal $Gd_5Si_{1.95}Ge_{2.05}$ ($x$=0.4875) strengths. The inset shows the transition temperature at low magnetic field of 8 kA/m.

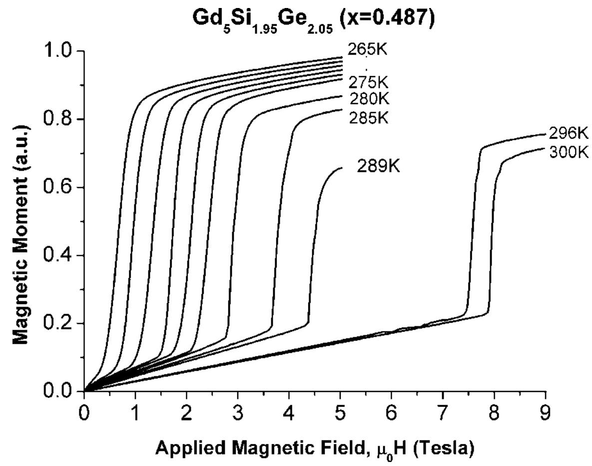


FIG. 3. *M* vs *H* for various temperatures above and below the SO PT temperature (296 K) of the orthorhombic phase for single crystal $Gd_5Si_{1.95}Ge_{2.05}$. Isotherms 265–289 K were measured in 5 T superconducting quantum interference device (SQUID) (Ref. 6) and isotherms 296 and 300 K were measured in 9 T VSM.

## III. RESULTS AND DISCUSSIONS

The transition temperatures of single crystal samples from the central region of the phase diagram, $Gd_5Si_{1.95}Ge_{2.05}$ ($x$=0.4875) and $Gd_5Si_2Ge_2$ ($x$=0.5), and Si-rich region, $Gd_5Si_{2.7}Ge_{1.3}$ ($x$=0.675), were determined using the inflection point. The FO PT temperatures $T_{FO}$ were found to be 263 K for $Gd_5Si_{1.95}Ge_{2.05}$ (see inset in Fig. 1) and 269 K for $Gd_5Si_2Ge_2$. The SO PT Curie temperature $T_C$ for single crystal $Gd_5Si_{2.7}Ge_{1.3}$ was found to be 307 K (see inset in Fig. 2).

The transition temperatures of single crystal $Gd_5Si_{1.95}Ge_{2.05}$ at different field strengths were determined for field strengths up to 5 T in a MPMS as shown in Fig. 1 which is in agreement with the strain versus magnetic field measurement of single crystal $Gd_5Si_2Ge_2$.[8] The change in the transition temperature was 4–5 K for every 1 T applied field change confirming the results reported by Han *et al.*[7] and Casanova *et al.*[9] For the measurements of magnetic moment versus temperature for single crystal $Gd_5Si_{2.7}Ge_{1.3}$ ($x$ =0.675), it was found that high applied magnetic fields broaden the transition to a wider range of higher temperatures (see Fig. 2).

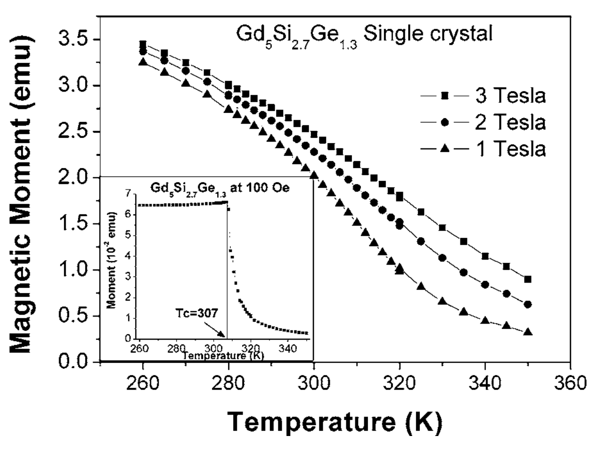


FIG. 2. *M* vs *T* at various values magnetic fields for single crystal $Gd_5Si_{2.7}Ge_{1.3}$ ($x$=0.675) at high applied magnetic fields strengths. The inset shows the identification of the transition temperature at low magnetic field of 8 kA/m.

As mentioned above, it was hypothesized that for temperatures $T$ above the SO PT temperature of the orthorhombic phase, $T > T_C^*$, that even the orthorhombic phase should be paramagnetic hence there would not be a field induced FO magnetic-structural PT for any amount of applied magnetic field. This temperature was estimated to be $T_C^*$=296 K for single crystal $Gd_5Si_{1.95}Ge_{2.05}$.[10] It is clearly seen that a field induced FO PT occurred even at temperatures higher than $T_C^*$=296 K for single crystal $Gd_5Si_{1.95}Ge_{2.05}$ (see Fig. 3). The same is observed for the single crystal $Gd_5Si_2Ge_2$ sample (see Fig. 4), which exhibits the field induced FO PT above the SO PT temperature $T_C^*$=301 K of the orthorhombic phase.[10]

Occurrence of a field induced FO PT at temperatures above the SO PT temperature of the orthorhombic phase at a high magnetic field can possibly be explained by the fact that the transition of the orthorhombic phase from ferromagnetic to paramagnetic is not distinct and it spreads over a wide range of higher temperatures. Even at temperatures higher than the Curie temperature $T_C$, at such high magnetic fields the magnetic moment is still quite large. Such a large mag-

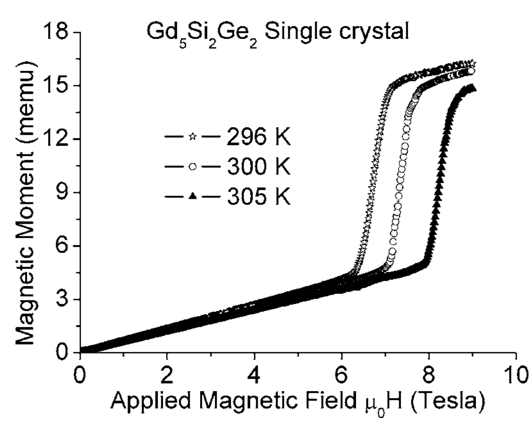


FIG. 4. *M* vs *H* for various temperatures above the SO PT temperature (301 K) of the orthorhombic phase for single crystal $Gd_5Si_2Ge_2$. Above 305 K the transition field is higher than 9 T.

netic moment of the orthorhombic phase can cause the FO PT to be favorable. The broadening of the transition to a wider range of temperatures is clearly seen in Fig. 2 which shows the measurement of magnetic moment versus temperature for single crystal $Gd_5Si_{2.7}Ge_{1.3}$ ($x=0.675$) at high applied magnetic field strengths. It can also be seen from Fig. 2 that at high applied fields there is no distinct Curie temperature $T_C$.

Observation of a field induced FO PT at high magnetic fields of up to 16 T has been reported by Casanova *et al.*,[9] but it was not compared to a projected SO PT temperature of the orthorhombic phase. It can also be seen from their paper that the error bars increased at higher magnetic fields in the transition field versus transition temperature graphs. We propose this is due to the broadening of the transition of the underlying orthorhombic phase at high magnetic fields. The magnetization versus temperature characteristics of the underlying orthorhombic phase of $Gd_5Si_{1.95}Ge_{2.05}$ and $Gd_5Si_2Ge_2$ should be quite similar to those we measured for orthorhombic $Gd_5Si_{2.7}Ge_{1.3}$ ($x=0.675$).

The rate of change in transition temperature with respect to field (5 K/T for zero applied pressure) can be lowered by applying an external compressive pressure. According to the work reported by Megan *et al.*,[11] this rate decreases with an increase in the isobaric compressive pressure on the sample. With the application of compressive pressure they obtained a field induced FO PT at temperatures above the SO PT temperature of the orthorhombic phase. Under a nonzero compressive isobaric pressure the magnetization of the ferromagnetic orthorhombic phase at any magnetic field is 25% less than the magnetization under zero compressive pressure. This is an indicative of an incomplete magnetic phase transformation of the underlying orthorhombic phase.

## IV. CONCLUSION

It was found that the field induced FO PTs of $Gd_5(Si_xGe_{1-x})_4$ occurs for the composition of $0.41 \leq x \leq 0.51$ even at temperatures above the projected SO PT temperature of the orthorhombic phase, provided sufficiently high magnetic fields are applied. This is thought to be due to the broadening of the transition to a wider range of higher temperatures. Due to this broadening of the transition, the magnetization of the material above the projected SO PT temperature of the orthorhombic phase is significant giving an impression of a magnetic PT. The rate of change in transition field with the temperature was found to be constant over all the temperature range.

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