Second order phase transition temperature of single crystals of Gd₅Si₁.₃Ge₂.₇ and Gd₅Si₁.₄Ge₂.₆

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Second order phase transition temperature of single crystals of Gd$_5$Si$_{1.3}$Ge$_{2.7}$ and Gd$_5$Si$_{1.4}$Ge$_{2.6}$

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Gd$_5$(Si$_{x}$Ge$_{1-x}$)$_4$ has mixed phases in the composition range $0.32 < x < 0.41$, which have not been widely studied. In this paper, we have synthesized and indexed single crystal samples of Gd$_5$Si$_{1.3}$Ge$_{2.7}$ and Gd$_5$Si$_{1.4}$Ge$_{2.6}$. We have investigated the first order and second order phase transition temperatures of these samples using magnetic moment vs. temperature and magnetic moment vs. magnetic field at different temperatures. We have used a modified Arrott plot technique that was developed and reported by us previously to determine the “hidden” second order phase transition temperature of the orthorhombic II phase.

INTRODUCTION

Gd$_5$(Si$_{x}$Ge$_{1-x}$)$_4$ still remains a magnetocaloric material of great interest due to its giant magnetocaloric effect near room temperature. The material has been widely studied over the composition range $0.41 < x < 0.51$, where the coupled magnetic and structural first order phase transitions occur close to room temperature. Gd$_5$(Si$_{x}$Ge$_{1-x}$)$_4$ has a mixed phase region in the phase diagram with both Sm$_2$Ge$_3$ type (orthorhombic I) and Gd$_5$Si$_4$ type (orthorhombic II) phases for compositions $0.32 < x < 0.41$. There has been little research on specific composition in this region. Arrott plots have been widely used to determine the nature of phase transitions in many magnetic materials. Previously, we have used modified Arrott plots to determine the second order phase transition temperature when it is suppressed by the first order phase transition in samples with compositions $x < 0.51$. In our previous work, we also used these modified Arrott plots on the mixed phase composition of Gd$_5$Si$_{1.2}$Ge$_{2.5}$ $(x = 0.375)$ to determine the second order phase transition temperatures of both the monoclinic and the orthorhombic II phases. In the present work, we have investigated two more single crystal samples of Gd$_5$Si$_{1.3}$Ge$_{2.7}$ and Gd$_5$Si$_{1.4}$Ge$_{2.6}$ whose compositions fall in the mixed phase regions of orthorhombic I and orthorhombic II in the phase diagram. The second order phase transition temperatures of the samples were estimated to be 383 K for Gd$_5$Si$_{1.3}$Ge$_{2.7}$ and 365 K for Gd$_5$Si$_{1.4}$Ge$_{2.6}$. These temperatures are much higher than the expected second order phase transition temperature of orthorhombic II phase (280 K). This may be due to the presence of the orthorhombic I phase in larger volume fraction.

EXPERIMENTAL DETAILS

Single crystal samples of Gd$_5$Si$_{1.3}$Ge$_{2.7}$ $(x = 0.325)$ and Gd$_5$Si$_{1.4}$Ge$_{2.6}$ $(x = 0.35)$ were prepared by the tri-arc crystal pulling method at the Materials Preparation Center of Ames Laboratory. The samples were heat treated at 1273 K for 24 h to reduce or eliminate the amount of orthorhombic I phase present in the sample. They were then cooled at a rate of 10$^\circ$/min. It is our experience that slow cooling produces a more phase pure sample than quenching. The ingots were cut using an electrical discharge machine (EDM) to limit any stress on the final cut sample. The samples were indexed using Laue X-ray diffraction technique. The Laue patterns of “a” and “c” axes were not distinguishable by this method due to their similar lattice parameters, so, they were distinguished by additional XRD measurements of the faces of the bulk samples. Magnetization measurements were carried out on a SQUID magnetometer. For the M vs. T measurements, the rate of heating/cooling of the samples was 10$^\circ$/min and the applied field was 100 Oe (8 kA/m).

RESULTS AND DISCUSSION

Figs. 1(a) and 1(b) show magnetization vs. temperature at an applied magnetic field of 100 Oe (8 kA/m) along the “a” axis of single crystal samples of Gd$_5$Si$_{1.3}$Ge$_{2.7}$ and Gd$_5$Si$_{1.4}$Ge$_{2.6}$. In both samples, the transition has a small thermal hysteresis and is abrupt, indicating that it is a first order phase transition. Gd$_5$Si$_{1.4}$Ge$_{2.6}$ shows a change in magnetic moment below the first order phase transition temperature, between 50 and 150 K, which may be due to the presence of a secondary phase of orthorhombic I phase.

Magnetic moment as a function of magnetic field for several temperatures near the first order phase transition temperature was also measured. Fig. 2 shows these...
measurements for (a) Gd$_{5}$Si$_{1.3}$Ge$_{2.7}$ (x = 0.325) and (b) Gd$_{5}$Si$_{1.4}$Ge$_{2.6}$ (x = 0.35) single crystal samples. A prominent feature of the MH isotherms is the large hysteresis indicating that these samples undergo a magnetic field induced first order phase transition. The hysteresis area within the forward sweep and reverse sweep of the magnetic field reduces with increase in the temperature similar to other samples that undergo first order phase transition temperature. The rate of change of transition temperature with respect to applied field is reported to be 5°–7°/T. Low magnetic field regions have a different slope from high magnetic field regions during the transition indicating that there is the presence of a residual phase in the samples.

M vs. H measurements at various temperatures close to the first order phase transition temperature were used in the Arrott plots, which show a set of parallel lines as shown in Fig. 3. Some of the isotherms have negative slope at lower magnetic fields indicating the presence of more than one phase transition and hence were not included here. The critical exponents β and γ used in these plots were 0.5 and 1 giving straight lines in the high field region indicating that the interaction between the spins follows mean-field behavior. These modified Arrott plots were used to estimate the second order phase transition temperatures of the samples similar to Hadimani et al. Figs. 3(a) and 3(b) show the Arrott plots for Gd$_{5}$Si$_{1.3}$Ge$_{2.7}$ and Gd$_{5}$Si$_{1.4}$Ge$_{2.6}$.
respectively. The projected second order phase transition temperatures are estimated to be 383 K for Gd₅Si₁.₃Ge₂.₇ and 365 K for Gd₅Si₁.₄Ge₂.₆. These estimated second order phase transition temperatures are significantly higher than the transition temperatures of sample with compositions 0.375 < x < 0.51, which may be due to presence of the orthorhombic I phase in the samples. In addition, one can also see that the transition temperature increases with decreasing amount of Si for this phase as opposed to the Si-rich region of Gd₅(Si₁-xGe₁-x)₄. In the mixed phase region of the phase diagram of Gd₅(Si₄Ge₁-x)₄ single crystal sample of Gd₅Si₁.₅Ge₂.₅ obeys the modified Arrott plot technique to determine the “hidden” second order phase transition temperature of the orthorhombic II and monoclinic phases but not the compositions of Gd₅Si₁.₃Ge₂.₇ and Gd₅Si₁.₄Ge₂.₆ as shown in Fig. 4.

CONCLUSION

We have investigated the first and second order phase transition temperatures of single crystal samples of Gd₅Si₁.₃Ge₂.₇ and Gd₅Si₁.₄Ge₂.₆ whose compositions fall in a mixed phase region of the phase diagram. The second order phase transition temperatures of the orthorhombic II phase of the samples were determined to be 365 K for Gd₅Si₁.₄Ge₂.₆ and 383 K for Gd₅Si₁.₃Ge₂.₇. These temperatures are much higher than the expected second order phase transition temperature of the orthorhombic II phase due to the presence of orthorhombic I phase as a residual phase in the samples. The estimated second order phase transition temperatures did not follow the trend in the phase diagram similar to the single crystal sample of Gd₅Si₁.₅Ge₂.₅, which also falls in the mixed phase region.

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