First Investigation of magnetic ground states in the rare-earth intermetallic compounds $RAl_{0.9}Si_{1.1}$ ($R = Ce, Pr, Gd$)

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Abstract

We report the magnetic properties strongly varying with the rare-earth elements in the newly found ternary compounds $RAl_{0.9}Si_{1.1}$, which crystallize in the tetragonal $\alpha$-ThSi$_2$-type structure. For $R = Ce$ the alloy has a weak ferromagnetism below 11 K and for $R = Pr$ it orders ferromagnetically at 17 K, while for $R = Gd$ it is antiferromagnetic with $T_N = 30.5$ K. In addition, we find no field effect on $T_N$ of $R = Gd$ because of the large internal mean field, but significant changes in the magnetic properties of $R = Ce$ and Pr.

Key words: rare-earth compound, local moment, magnetic properties, exchange interaction, spin-glass transition
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There have been studies of Ce-based ternary compounds in the Ce-Al-(Si,Ge) phase diagram [1]. It was found for CeAl$_x$(Si,Ge)$_{2-x}$ that the crystal structure changes as the Al/Si(Ge) ratio varies [2]. The alloys of $x = 1$ and 1.2 have the tetragonal $\alpha$-ThSi$_2$-type structure, while the alloy of $x = 1.5$ has the hexagonal AlB$_2$-type structure. The magnetic properties were reported to depend on the crystal chemistry [3]; CeAlSi is ferromagnetic with a Curie temperature of 7.1 K, whereas CeAlGe orders antiferromagnetically at 4 K. In the present work, we have extended this investigation to other ternary compounds as changing

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The specific heat $C(T)$ vs. temperature for $R = \text{Ce, Pr, and Gd}$ in $\text{RAI}_{0.9}\text{Si}_{1.1}$. The solid symbols are zero-field data and the open symbols are 9T data.

the rare-earth elements, which are $\text{RAI}_{0.9}\text{Si}_{1.1}$ ($R = \text{Ce, Pr, and Gd}$) and find that the magnetic properties are strongly varying with the rare-earth elements.

The single crystals were synthesized by high-temperature flux method. Electron-probe microanalysis showed the composition of $\text{RAI}_{0.9}\text{Si}_{1.1}$ within an error of $\pm 0.05$ for $R = \text{Gd}$ and $\pm 0.03$ for $R = \text{Ce and Pr}$ without any impurity phase. X-ray powder diffraction pattern revealed that the samples are single phased with the tetragonal $\alpha$-ThSi$_2$-type structure. The magnetic susceptibility was measured in a field of 100 G using a Quantum Design superconducting quantum interference device. The specific heat was taken with a Quantum Design physical property measurement system.

The inverse magnetic susceptibility $H/M$ vs. temperature is shown in Fig. 1. A linear behavior of the Curie-Weiss law is observed above 100 K and a deviation from the linear behavior occurs below 50 K. The effective magnetic moments are estimated to be $\mu_{\text{eff}} = 2.52(5) \mu_B$, $3.31(8) \mu_B$, and $8.10(8) \mu_B$ for $R = \text{Ce, Pr, and Gd}$, respectively. These values show that the rare-earth ions are in the normal trivalent state. The paramagnetic Curie temperatures are $\theta_P = 6.91(0)$ K, $26.37(4)$ K, and $-142.43(3)$ K for $R = \text{Ce, Pr, and Gd}$, respectively, possibly indicating the development of different types of magnetic exchange interaction. The small value of $\theta_P$ for CeAl$_{0.9}$Si$_{1.1}$ might be related with a weak ferromagnetism, while the negative value of $\theta_P$ for GdAl$_{0.9}$Si$_{1.1}$ might be responsible for an antiferromagnetic order.

These different types of magnetic exchange could be achieved at low temperatures. In the inset of Fig. 1, we observe a peak in the zero-field cooled (ZFC) curve and a cusp in the field cooled (FC) curve at $T_{wF} = 11$ K, which fur-
Fig. 2. Inverse magnetic susceptibility $H/M$ vs. temperature for $R = \text{Ce, Pr, and Gd}$ in $\text{RAI}_{0.9}\text{Si}_{1.1}$. The solid lines represent linear fits of the Curie-Weiss law. The inset shows the low-temperature susceptibility $M/H$ of $R = \text{Ce}$.

ther confirms weak ferromagnetism in $\text{CeAl}_{0.9}\text{Si}_{1.1}$. The magnetic behavior of $\text{PrAl}_{0.9}\text{Si}_{1.1}$ could be also understood in a way similar to that of $\text{CeAl}_{0.9}\text{Si}_{1.1}$, because there is a difference between ZFC and FC susceptibilities below $T_C = 17$ K. However, we observe a rapid saturation of magnetization to a value of almost full moment $\sim 3\mu_B$ at 0.4 T (not shown here), indicating a ferromagnetic ordering. It is worthwhile to mention here that this difference could be associated with a spin-glass transition. On the other hand, $\text{GdAl}_{0.9}\text{Si}_{1.1}$ has no difference between ZFC and FC susceptibilities and orders antiferromagnetically below $T_N = 30.5$ K. The magnetization increases linearly with magnetic field (not shown here).

Figure 2 represents the specific heat $C(T)$ vs. temperature. A $\lambda$-type anomaly is observed in all the compounds $\text{RAI}_{0.9}\text{Si}_{1.1}$ at the magnetic transition temperatures: $T_{wF} = 11$ K, $T_C = 17$ K, and $T_N = 30.5$ K for $R = \text{Ce, Pr, and Gd}$, respectively. As the magnetic is applied, the magnetic transition temperatures for $R = \text{Ce and Pr}$ are increased, whereas the transition temperature for $R = \text{Gd}$ is unchanged. The former is characteristic of ferromagnetic materials. In the latter case, one should note that $\text{Gd}^{3+}$ ions ($L = 0$ and $J = S$) in $\text{GdAl}_{0.9}\text{Si}_{1.1}$ have a large internal mean field and thus an effect of external field could be negligible in total magnetic exchange. We may depress $T_N$ in a very high magnetic field beyond our present measurement range.

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