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The Effect of Ce Dilution on the Ferromagnetic Ordering in CeAuGe

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Transport and thermodynamic properties of the well-ordered hexagonal CeAuGe compound have been studied. This compound is known to order ferromagnetically at $T_C = 10$ K with well-defined anomalies in magnetic susceptibility $\chi(T)$, electrical resistivity $\rho(T)$ and specific heat $C_p(T)$ characterising the phase transition. The location of T_C has been observed to be unstable and enhanced even in moderate applied magnetic fields. However, the dilution of magnetic species, Ce, with the non-f electron element, La, is shown in this work to achieve a continuous suppression of T_C to 0 K. The integrity of the space group and the details of the unit cell occupation are retained throughout the substitution series, as is the high-temperature localized Ce-effective magnetic moment $\mu_{\text{eff}} = 2.54\ \mu_B$/(mol Ce). Our studies of physical properties down to 0.05 K show a quantum critical form of non-Fermi liquid behaviour, characterised by a logarithmic divergence in $C_p(T)/T$ data in the very dilute Ce content.

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1. Introduction

An ongoing interest into the ground state of cerium-based intermetallic compounds both experimentally and theoretically is due to their interesting physical properties such as the Kondo effect, heavy fermion behavior, valence fluctuation, superconductivity, and magnetic properties [1, 2]. Some of these materials have been found to have unusual low temperature properties that appear to violate the Landau–Fermi-liquid (LFL) paradigm [3, 4]. These non-Fermi-liquid (NFL) materials exhibit weak power-law or logarithmic T dependences in various physical properties such as in $\chi(T)$, $\rho(T)$ and $C_p(T)$, which are contrary to those of LFL where $C_p(T)/T$ and $\chi(T)$ are constant and $\rho(T) \propto T^2$. These kinds of unusual low T behaviour are most often observed at a quantum phase transition (QPT), in close proximity to where a magnetic phase transition is suppressed to zero temperature [5]. Most research on NFL physics has focused more on antiferromagnetic (AFM) QPTs; comparatively little is known about ferromagnetic (FM) QPTs.

In this paper we report on the initial studies of the investigation of the possibility of NFL behaviour in ferromagnetic CeAuGe compound. It has been established from the previous work that in CeAuGe, Ce carries a local moment with compound ordering ferromagnetically at 10 K [6, 7]. The magnetic order manifests anomalies in $\chi(T)$, $\rho(T)$ and $C_p(T)$ at T_C [7, 8]. The anomaly has been evidenced to shift upwards in T with the application of magnetic field. In this work, the first results of the dilution of the Ce-site with non-magnetic element (La) will be reported.

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2. Experimental procedure

Polycrystalline samples of $Ce_{1-x}La_xAuGe$ ($x = 0$–1) compounds were prepared by arc-melting stoichiometric quantities of high-purity elements under pressure in an ultra-high purity argon gas. The as-cast samples were characterized through powder X-ray diffraction (XRD) and the Rietveld refinement profile using general structure analysis software (GSAS). The $C_p(T)$ measurements were performed using Physical Properties Measurement System (PPMS) from Quantum Design (San Diego), between 0.05 K $\leq T \leq$ 300 K. The $\chi(T)$ measurements were performed using Magnetic Properties Measurement System (MPMS) from the same suppliers, in small applied magnetic field of 0.005 T between 1.9 K and 350 K.

3. Results and discussion

3.1. Powder X-ray diffraction and sample characterisation

CeAuGe structure was confirmed to crystallize in hexagonal NdPtSb-type structure with space group $P6_3mc$ (186) [6] which remained unchanged in dilution compounds, aside from expected chemical compression effect, throughout the series. The point group of 3 or $3m$ is exhibited by Ce atom in this hexagonal point symmetry. The Rietveld refinement of XRD data was performed. The lattice parameters, a, c and the volume V of the unit cell of dilution compounds increase as the La content in the compound increases (see Fig. 1a–c). The figures show a positive linear correlation between the lattice parameters and La content as expected from Vegard's law.

3.2. Magnetic properties

Magnetic studies of CeAuGe have been discussed in prior work [7]. Higher temperature $\chi^{-1}(T)$, ($T > 150$ K)

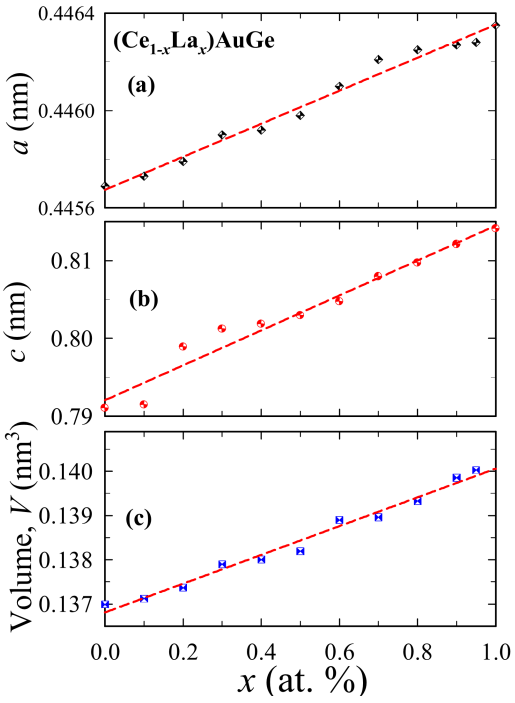

Fig. 1. (a) lattice parameters a, (b) c and (c) unit cell volume V obtained from the XRD spectra refinements of the series of dilution compounds.

data of dilution compounds studied in this paper were fitted using the Curie–Weiss law. The calculation of the effective magnetic moment, $\mu_{\rm eff}$, for a certain range ($0 \leq x \leq 0.7$) of La content were observed to be close to the theoretical full magnetic moment for Ce^{3+} ion. These varied between 2.52 and 2.54 $\mu_{\rm B}$/(mol Ce). It has also been observed that for Ce-rich compositions ($0 \leq x \leq 0.5$), the fitted θ_p yielded negative values indicating predominant AFM exchange interactions within the paramagnetic region, despite the known FM ground state in this system. Positive θ_p-values were obtained for the La-rich compounds ($0.6 \leq x \leq 0.9$), which is an indication of the emerging dominance of net FM exchange. Thus, separation of Ce ions, typical of stabilising the f–c (f- and conduction electrons, respectively) hybridization, seem to favour FM exchange.

Figure 2 shows the inverse magnetic susceptibility data, $\chi^{-1}T$) (see main parts of Fig. 2a and b), with the inset showing the low-T susceptibility. The anomaly associated with $T_{\rm C}$ (when considering the inflection point as the FM ordering temperature, as indicated by arrow in Fig. 2a) is suppressed and shifts down in temperature with increasing La content. This downward shift in $T_{\rm C}$ persists, and for $x \leq 0.7$ there was no observable anomaly associated with $T_{\rm C}$ as also revealed in Fig. 2b (a representative data with no detectable $T_{\rm C}$ anomaly). For the lowest Ce content ($Ce_{0.1}La_{0.9}AuGe$ and $Ce_{0.2}La_{0.8}AuGe$), a follow up on the behaviour of the system was done using a specific heat study.

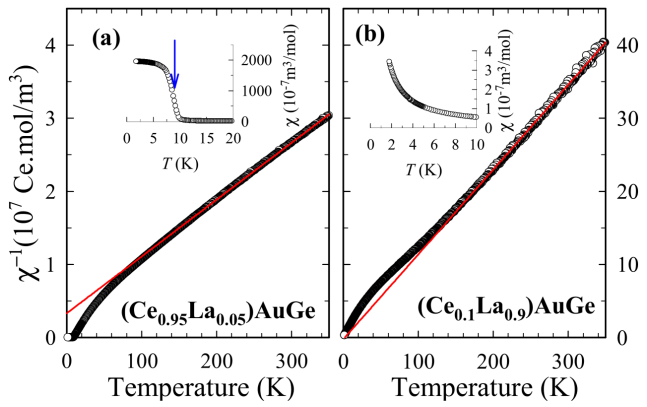

Fig. 2. Inverse susceptibility $\chi^{-1}(T)$ (main panels) and low-T susceptibility for the two representative members in the dilution series.

3.3. Specific heat

Figure 3a shows the specific heat data in the form $C_p(T)/T$ vs. T of selected high Ce content compounds ($Ce_{0.95}La_{0.05}AuGe$, $Ce_{0.9}La_{0.1}AuGe$ and $Ce_{0.8}La_{0.2}AuGe$) where the arrow indicates $T_{\rm C}$, whilst Fig. 3b shows the corresponding $C_p(T)/T$ vs. T^2 data.

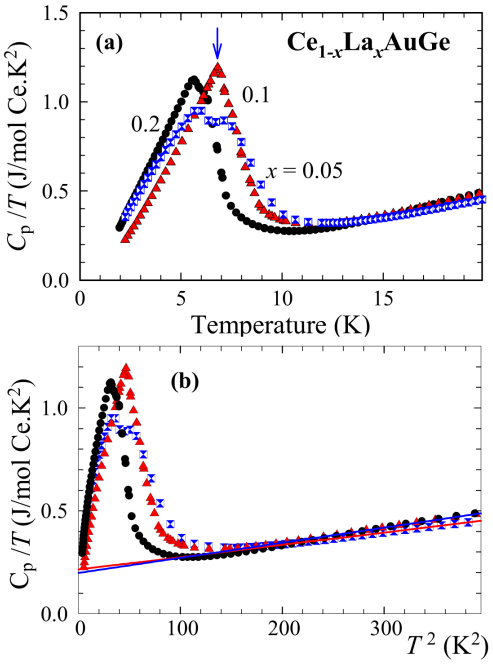

Fig. 3. (a) $C_p(T)/T$ vs T and (b) $C_p(T)/T$ vs T^2 of $Ce_{1-x}La_xAuGe$ for selected compositions.

The anomalies observed around the ordering temperature in compounds with $x = 0.05$ and 0.2 are ascribed to instrumental error, since no evidence of extra phases were detected in these compounds. The region just above the ordering temperature of these data

was fitted for the Sommerfeld coefficient according to the expression $C_p(T)/T = \gamma + \beta T^2$, and fits are shown by the solid lines in Fig. 3b. It was observed that the Sommerfeld coefficient γ increased with the increase in La content, as it was fitted to be 175(3); 198(3) and 215(3) mJ/(mol Ce K^2) for $Ce_{0.95}La_{0.05}AuGe$, $Ce_{0.9}La_{0.1}AuGe$ and $Ce_{0.8}La_{0.2}AuGe$, respectively.

This enhancement of γ in the Ce-rich region is an indication of the appreciable hybridization of the localized Ce $4f$ states with the conduction electron states [9] associated with an increase in La atomic percent. It was observed that for $x \geq 0.5$, γ decreases as La content increased.

Figure 4 depicts $C_p(T)/T$ data of $Ce_{1-x}La_xAuGe$ ($x = 0.50-0.9$) measured from 6–0.36 K. It is observed that there is a persistent downward shift in T_C as a function of La content, until T_C is not detectable in compounds $Ce_{0.2}La_{0.8}AuGe$ and $Ce_{0.1}La_{0.9}AuGe$ at the measured temperature ranges. These two compounds were then measured further at very low temperatures, down to 0.05 K As the anomaly associated with T_C disappears out of measurable range as $x \to 1$, a sharp increase in $C_p(T)/T$ is observed as $T \to 0$.

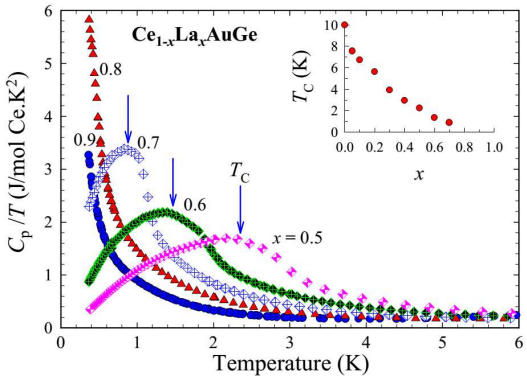

Fig. 4. $C_p(T)/T$ vs T of $Ce_{1-x}La_xAuGe$ ($x = 0.5$, 0.6, 0.7, 0.8 and 0.9) with arrow indicating T_C.

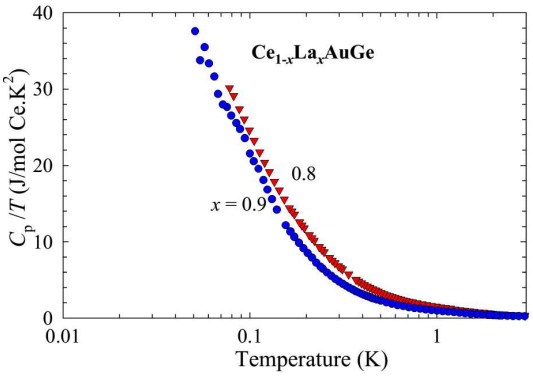

Fig. 5. $C_p(T)/T$ vs. $\log T$ of $Ce_{1-x}La_xAuGe$ ($x = 0.9$ and 0.8).

Figure 5 shows the $C_p(T)/T$ vs. T data (on a semi-log plot) of $Ce_{0.1}La_{0.9}AuGe$ and $Ce_{0.2}La_{0.8}AuGe$ measured at very low temperature ranges, to find the traces of FM order very close to 0 K. It is revealed that there exists a $C_p(T)/T \sim -\log T$ behaviour as $T \to 0$, this observation indicates a divergence from LFL ($C_p(T)/T \sim$ constant) behaviour, in both dilution compounds.

4. Conclusions

The dilution of Ce magnetic species with La resulted in the suppression of $T_C = 10$ K in CeAuGe down to about 0.87 K obtained in $Ce_{0.3}La_{0.7}AuGe$ compound. The extreme two Ce-dilute compounds, $Ce_{0.1}La_{0.9}AuGe$ and $Ce_{0.2}La_{0.8}AuGe$ appear to remain paramagnetic down to $T \to 0$. Analysis of these two dilution compounds as shown by the results of $C_p(T)/T$ showed a characteristic form of NFL behaviour where $C_p(T)/T$ followed a $-\log T$ dependence in the temperature range 0.05 K $\leq T \leq$ 0.2 K. Magnetic susceptibility, $\chi(T)$ and $\rho(T)$ measurements at very low temperatures are in progress, to support the $C_p(T)$ data.

Acknowledgments

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