Crossover from Single-Ion to Coherent Non-Fermi Liquid Behavior in Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$

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Abstract

We report specific heat and magneto-resistance studies on the compound Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ for various concentrations over the entire stoichiometric range. Our data reveal single-ion scaling with Ce-concentration between $x = 0.1$ and 0.95. Furthermore, CeNi$_9$Ge$_4$ turns out to have the largest ever recorded value of the electronic specific heat $\Delta c/T \approx 5.5$ J K$^{-2}$mol$^{-1}$ at $T = 0.08$ K which was found in Cerium f-electron lattice systems. In the doped samples $\Delta c/T$ increases logarithmically in the temperature range between 3 K and 50 mK typical for non-Fermi liquid (nFl) behavior, while $\rho$ exhibits a Kondo-like minimum around 30 K, followed by a single-ion local nFl behavior. In contrast to this, CeNi$_9$Ge$_4$ flattens out in $\Delta c/T$ below 300 mK and displays a pronounced maximum in the resistivity curve at 1.5 K indicating a coherent heavy fermion groundstate. These properties render the compound Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ a unique system on the borderline between Fermi liquid and nFl physics.

Key words: non Fermi liquid, Heavy Fermion, single ion effect

Cerium-based intermetallic compounds exhibit a rich variety of ground state properties due to the competition between three different types of interactions: i) crystal-field effects; ii) Ce-Ce intersite correlations which result in long-range magnetic order and iii) on-site correlation between 4f- and conduction-electron states leading to a formation of a local Kondo singlet. In order to minimize the intersite correlation by increasing the Ce-sublattice space CeNi$_9$Si$_4$ and CeNi$_9$Ge$_4$ turn out to be good candidates. Both compounds crystallize in a tetragonal structure with space group $I4/mcm$ and a minimum Ce-Ce distance of 5.5479 Å for Si [1] and of 5.6357 Å for Ge [2]. While CeNi$_9$Si$_4$ is a typical Kondo lattice system with a moderate Sommerfeld coefficient $\gamma = 155$ mJ K$^{-2}$mol$^{-1}$ [1], CeNi$_9$Ge$_4$ exhibits non-Fermi liquid behavior with the largest ever recorded value of the electronic specific heat $\Delta c/T \approx 5.5$ J K$^{-2}$mol$^{-1}$ at $T = 0.08$ K without showing any trace of magnetic order [2]. Recently, specific heat and susceptibility studies on La-substituted samples Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ indicate that the large electronic specific heat value $\Delta c/T$ of CeNi$_9$Ge$_4$ is mainly caused by single ion effects [3]. In addition the observed nFl-behavior in $\Delta c/T$ and $\rho$ and the Kondo-like behavior in $\chi$ suggests that in the Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ system two channel Kondo physics takes place [3].

The specific heat divided by temperature of polycrystalline samples Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$(with $x$ ranging from 0 to 1) is displayed in Fig. 1. For all La substituted samples a nearly logarithmic increase of $c/T$ below 1.5 K is observed which is characteristic for nFl phys...
Fig. 1. A semi-logarithmic plot of the specific heat divided by temperature of Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ in the temperature range 0.05K $< T < 20$K. The insert shows the electronic contribution to the specific heat $\Delta c/T$ normalized per Ce-mol.

Fig. 2. The electrical resistivity $\rho(T)$ of various samples normalized at 300 K to that of LaNi$_9$Ge$_4$ which was detected with the Vander-Pauw method. Notice that the unexpected smaller residual resistivity ratio of the stochiometric single crystal sample [4] may be due to anisotropy effects and stress. The insert shows for CeNi$_9$Ge$_4$ the shift of the resistivity maximum at 1.5 K with increasing magnetic field up to higher temperatures.

Fig. 3. Magneto resistance of Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ at 2 K. Insert: percentage decrease of magneto resistance between 0 and 9T.

The concentration dependent crossover from single-ion nFl to coherent behavior in Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ is also supported by resistivity measurements (Fig. 2). With the exception of CeNi$_9$Ge$_4$ and LaNi$_9$Ge$_4$ the resistivity of all samples passes through a Kondo-like minimum around 30 K, followed by a logarithmic temperature increase and saturates below 2 K with $\rho(T) - \rho(0) \propto T^c$, $c = 0.8\pm0.2$ indicating single-ion nFl-like behavior. While LaNi$_9$Ge$_4$ shows normal metallic behavior, only CeNi$_9$Ge$_4$ has a maximum at low temperature indicating a concentration dependent crossover from a single-ion to a coherent Kondo lattice state below 1 K. Fig. 3 shows the magneto-resistance between 0 and 9 T at 2K. The percentage decrease of magneto resistance between $\rho_{\text{imp}}(0)$ and $\rho_{\text{imp}}(9T)$ increases linearly with rising Ce-concentration starting at $x = 0.9$ (insert Fig. 3). Only for the non-diluted CeNi$_9$Ge$_4$ this reduction deviates from the linearity to higher values, again indicating a concentration dependent crossover from single-ion nFl to coherent behavior. Therefore the magneto-resistance data are in good agreement with those of the specific heat data [3].

References

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