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Unusual non-fermi liquid behavior of $Ce_{1-x}La_xNi_9Ge_4$ analyzed in a single impurity Anderson model with crystal field effects

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Abstract

$CeNi_9Ge_4$ exhibits unusual non-Fermi liquid behavior with the largest ever recorded value of the electronic specific heat $\Delta C/T \cong 5.5\,\mathrm{J\,K^{-2}\,mol^{-1}}$ without showing any evidence of magnetic order. Specific heat measurements show that the logarithmic increase of the Sommerfeld coefficient flattens off below 200 mK. In marked contrast, the local susceptibility $\Delta\chi$ levels off well above 200 mK and already becomes constant below 1 K. Furthermore, the entropy reaches $2R\ln 2$ below 20 K corresponding to a four-level system. An analysis of C and χ was performed in terms of an $SU(N=4)$ single impurity Anderson model with additional crystal electric field (CEF) splitting. Numerical renormalization group calculations point to a possible consistent description of the different low-temperature scales in Δc and $\Delta\chi$ stemming from the interplay of Kondo effect and crystal field splitting.

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Strongly correlated electron materials have been of interest for many years since they allow to test the limits of Landau's Fermi liquid (FL) theory. Specific heat and susceptibility measurements are good tools to distinguish whether electronic correlations renormalize the Fermi liquid parameters or lead to a new non-Fermi liquid (nFL) state. The breakdown of FL theory and the borderline between these two regimes continues to attract much interest [1]. Recently, we have shown that $Ce_{1-x}La_xNi_9Ge_4$ is a very interesting system for studying this borderline [2,3].

The stoichiometric system ($x=0$) shows the largest ever recorded value of the electronic specific heat $\Delta C/T \cong 5.5\,\mathrm{J\,K^{-2}\,mol^{-1}}$ without any magnetic order [4]. This behavior is mainly driven by single ion (Ce) effects [2]: the low-temperature behavior of the Sommerfeld coefficient ($\gamma \sim \Delta c(T)/T$) and the local susceptibility ($\chi(T)$) are proportional to $1-x$ (except for $x=0$ which shows additional collective effects). As representative of all dilute samples, we discuss $Ce_{0.5}La_{0.5}Ni_9Ge_4$ in this paper. In order to extract the electronic contribution to its specific heat, we calculated the phonon contribution of the non-f-electron system $LaNi_9Ge_4$. The solid line in Fig. 1a is a phonon-fit which is parametrized using a Debye term ($\Theta_D = 135\,\mathrm{K}$, three degrees of freedom (dof)) and two Einstein modes ($\Theta_E = 154\,\mathrm{K}$, 12 dof; $\Theta_E = 301\,\mathrm{K}$, 27 dof). This result is in good agreement with recent neutron scattering measurements, where the inelastic response in both $CeNi_9Ge_4$ and $LaNi_9Ge_4$ was dominated by strong phonon peaks centered at 15 and 35 meV [5].

Notice that the Sommerfeld coefficient $\Delta c/T$ of $Ce_{0.5}La_{0.5}Ni_9Ge_4$ shows nFL-behavior (logarithmic increase) down to 60 mK, while $\chi(T)$ becomes constant

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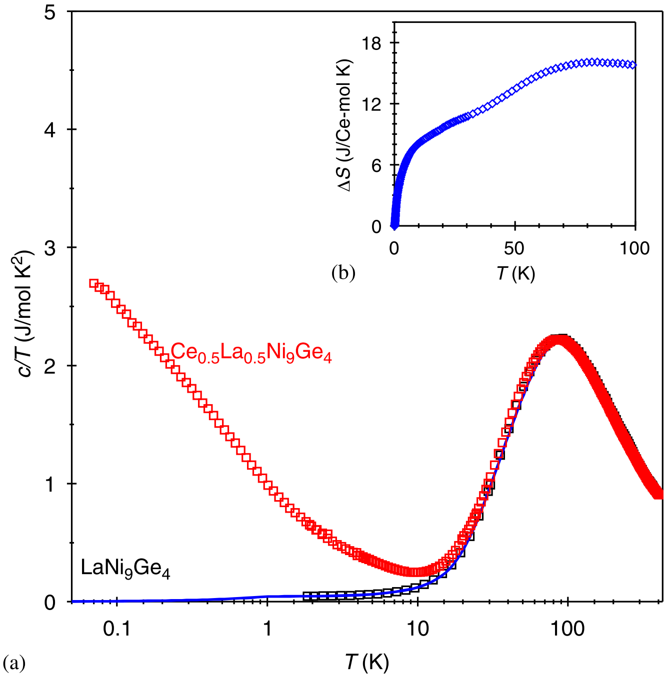

Fig. 1. (a) A semi-logarithmic plot of c/T vs. T of $Ce_{0.5}La_{0.5}Ni_9Ge_4$ and $LaNi_9Ge_4$. The solid line mirrors the phonon contribution of $LaNi_9Ge_4$ (see text). (b) The electronic part of the entropy of $Ce_{0.5}La_{0.5}Ni_9Ge_4$ as calculated from the data of Fig. 2a.

(Fermi liquid like) below 1 K (Fig. 2). From an entropy calculation ($S = 2R\ln 2$ for $T < 20$ K) one could suggest that the crystal electrical field (CEF) ground state quartet of Ce^{3+} splits into two doublets leading to an interplay between Kondo effect and CEF-splitting on the same energy scale [2]. In this work, we use a numerical renormalization group calculation (NRG) to establish a consistent description of the different low-temperature scales in Δc and $\Delta\chi$. We use the SU(4)-Anderson impurity model with additional crystal field splitting as a possible theoretical minimal model:

$$H = H_{\text{cond}} + H_{\text{imp}} + H_{\text{hyb}}, \tag{1}$$

$$H_{\text{cond}} = \sum_{\sigma}\sum_{\alpha=1,2} \varepsilon_{k\alpha\sigma} c^{\dagger}_{k\alpha\sigma} c_{k\alpha\sigma}, \tag{2}$$

$$H_{\text{imp}} = \sum_{\alpha\sigma} E_{\alpha} |\alpha\sigma\rangle\langle\alpha\sigma|, \tag{3}$$

$$H_{\text{hyp}} = \sum_{k\alpha\sigma} V_{\alpha\sigma}(c^{\dagger}_{k\alpha\sigma}|0\rangle\langle\alpha\sigma| + |\alpha\sigma\rangle\langle 0|c_{k\alpha\sigma}). \tag{4}$$

H_{imp} describes the dynamics of the Ce 4f-states under the assumption that only the unoccupied state $|0\rangle$ and singly occupied states $|\alpha\sigma\rangle$ play a role for the low-temperature physics. Therefore, the Schottky peak at about 60 K due to the $J = \frac{5}{2}$ states is necessarily absent in the NRG fits (see Fig. 2a). We found the best agreement with the

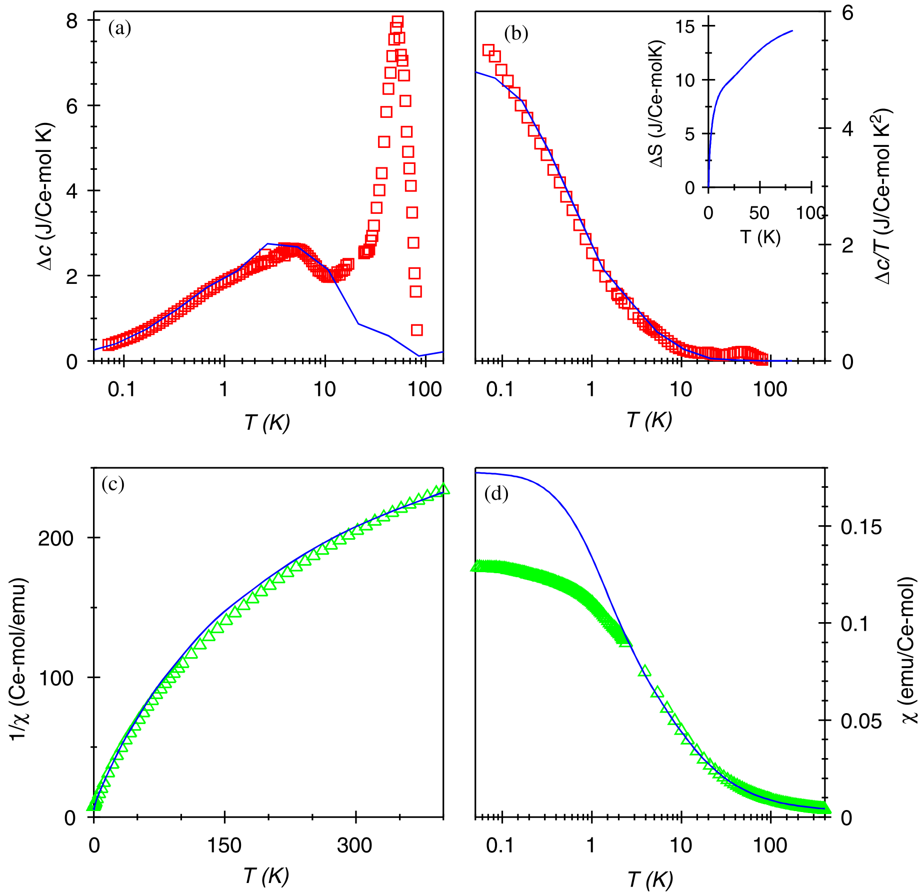

Fig. 2. $Ce_{0.5}La_{0.5}Ni_9Ge_4$: (a) a semi-logarithmic plot of the electronic contribution of the specific heat Δc and (b) $\Delta c/T$. (c) The inverse susceptibility vs. T and (d) the local susceptibility down to 30 mK. The lines always show the fit results from the NRG calculation (see text).

low-temperature data for a crystal field splitting $\Delta = E_2 - E_1 \approx 10\,\mathrm{K}$ for the ground state doublet $|\alpha\sigma\rangle$ and approximately the same Kondo temperature T_K due to the hybridization matrix elements $V_{\alpha\sigma}$ [6]. From Fig. 2c and d one concludes that while the quantitative agreement at very low temperatures is not yet satisfactory, the interplay of crystal field effects and Kondo physics can lead to temperature scales for the onset of Fermi liquid-like behavior in the specific heat and magnetic susceptibility that differ by a factor of about 10. This would imply that such an interplay can result in an extended NFL-like logarithmic behavior of $\Delta c/T$, while eventually the system does become a Fermi liquid at even lower temperatures.

This observation based on our NRG calculations is of general importance for analyzing NFL-like behavior in heavy fermion systems. Whether our theoretical model can quantitatively explain the experimental data for $Ce_{1-x}La_xNi_9Ge_4$, however, still needs to be explored further. Work along these lines is in progress.

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