Evolution of single-ion crystal field and Kondo features in Ce$_{0.5}$La$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$

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Evolution of single-ion crystal field and Kondo features in Ce$_{0.5}$La$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$

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Abstract. Starting with the heavy fermion compound CeNi$_9$Ge$_4$, the substitution of nickel by copper leads to a dominance of the RKKY interaction in competition with the Kondo and crystal field interaction. Consequently, this results in an antiferromagnetic phase transition in CeNi$_{9-x}$Cu$_x$Ge$_4$ for $x > 0.4$, which is, however, not fully completed up to a Cu-concentration of $x = 1$. To study the influence of single-ion effects on the AFM ordering by shielding the 4f-moments, we analyzed the spin diluted substitution series La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ by magnetic susceptibility $\chi$ and specific heat $C$ measurements. For small Cu-amounts $x \leq 0.4$ the data reveal single-ion scaling with regard to the Ce-concentration, while for larger Cu-concentrations the AFM transition (encountered in the CeNi$_{9-x}$Cu$_x$Ge$_4$ series) is found to be completely depressed. Calculation of the entropy reveal that the Kondo-effect still shields the 4f-moments of the Ce$^{3+}$-ions in CeNi$_8$CuGe$_4$.

1. Introduction

One of the most outstanding Fermi-liquid systems is the heavy fermion compound CeNi$_9$Ge$_4$, which turns out to display the largest ever recorded value of the electronic specific heat $\Delta C/T \approx 5.5$ Jmol$^{-1}$K$^{-2}$ without showing any magnetic order [1]. The dilution of the magnetic Cerium 4f-moments in Ce$_{1-x}$La$_x$Ni$_9$Ge$_4$ reveals single-ion scaling with regard to the Ce-concentration [2]. Therefore, the unique behavior of CeNi$_9$Ge$_4$ could be mainly attributed to a single-ion effect. Gradually replacing Ni by Cu changes both, the 3d electron number and the lattice parameters. This substitution influences the crystal field and leads to a formation of long range antiferromagnetic order in CeNi$_{9-x}$Cu$_x$Ge$_4$ for $x > 0.4$ which culminates in a transition temperature of $T_N = 175$ mK for $x = 1$ [3]. Even though the maximum of the magnetic specific heat $\Delta C(T_N)$ of CeNi$_9$CuGe$_4$ reaches less than 15% of the theoretical expected value, the transition was discussed in terms of a reduced long range antiferromagnetic order due to the presence of the Kondo-effect [3]. At a suitable concentration of $x \approx 0.4$, where a crossover between single-ion and magnetic ordered behavior occurs, the system exhibits a quantum critical phase transition (QCP)[3].

In the present work we studied the influence of Kondo-shielding on the antiferromagnetic ordering and how far single-ion effects are still present when crossing the phase transition from a Kondo-state ($x \leq 0.4$) to an antiferromagnetic coherent state ($x \geq 0.4$). Therefore we performed magnetic susceptibility $\chi$ and specific heat $C$ measurements of the spin diluted substitution series La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ and compared them to the pure CeNi$_{9-x}$Cu$_x$Ge$_4$ series.
2. Experimental Details
Polycrystalline samples of La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ were prepared by arc melting the pure elements under argon atmosphere. Subsequently the samples were annealed at 950$^\circ$C for two weeks in evacuated quartz tubes. Less than 0.8% weight loss occurred during the melting process. X-ray powder diffraction, optical emission spectroscopy in an indicatively coupled plasma (ICP-OES) and energy dispersive X-ray spectroscopy (EDX) indicated that the samples display a single phase character. The system crystalizes in the tetragonal LaFe$_9$Si$_4$-type structure (space group $I4/mcm$). For details of the preparation and the measurements on the pure Ce-compounds CeNi$_{9-x}$Cu$_x$Ge$_4$ see [3].

Figure 1 shows the magnetic susceptibility $\chi(T)$ in the temperature range 2 K $< T < 400$ K of La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ normalized per Ce-mol (filled symbols) in comparison with the pure Ce-alloys (open symbols) taken from [3]. For a direct comparison of the magnetically dilute solid solution La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ with the corresponding solid solution CeNi$_{9-x}$Cu$_x$Ge$_4$ with undiluted Ce-sublattice, the specific heat and magnetic susceptibility data are normalized to the Ce-content. In case of the Ce-normalized magnetic susceptibility ($\chi_{\text{Ce-mol}}$), we first subtracted the nonparamagnetic La-contribution and then scaled the data with the Ce-concentration, using the following equation:

$$\chi_{\text{Ce-mol}} = 2 \cdot \left( \chi(\text{La}_{0.5}\text{Ce}_{0.5}\text{Ni}_{9-x}\text{Cu}_x\text{Ge}_4) - 0.5 \cdot \chi(\text{LaNi}_9\text{Ge}_4) \right)$$  \hspace{1cm} (1)

For $T > 80$ K, $\chi(T)$ follows a modified Curie-Weiss law, $\chi(T) = C/(T - \Theta) + \chi_0$, yielding an effective magnetic moment of $\mu_{\text{eff}} \approx 2.5\mu_B$, as theoretically expected for a Ce$^{3+}$-lattice. In the low temperature range ($T < 80$ K) the data scales for $x \leq 0.4$ with the Ce-concentration indicating single-ion behavior. For $x > 0.4$ the single-ion character vanishes and the temperature dependence of the La-substituted samples deviate from the behavior of the pure Ce-compounds, which follow a Curie-Weiss-law only down to 100 K, due to the formation of an antiferromagnetic transition at lower temperatures [3]. The different temperature dependence of $\chi(T)$ is due to the absence of antiferromagnetic correlations in the La-substituted system and results from the dilution of the magnetic moments.

The specific heat $C$ normalized per Ce-mol and divided by temperature $T$ is displayed in Fig. 2 for La$_{0.5}$Ce$_{0.5}$Ni$_{9-x}$Cu$_x$Ge$_4$ in the temperature range between 0.05 K and $T < 300$ K. As already
known from literature [2] a normalization to CeNi₉Ge₄ for \( x = 0 \) is not possible due to the coherence of the Kondo-lattice. This is, however, not true for the diluted sample, where a logarithmic increase of \( C/T \) below 1.5 K is observed, indicating non-Fermi-liquid behavior. For \( 0 < x \leq 0.4 \) the data scale with the undiluted compounds which is in agreement with single-ion effects as already observed in the magnetic susceptibility. The absence of the antiferromagnetic transition for \( x > 0.4 \) in the Ce-diluted compounds is also in line with their susceptibility behavior. The stronger increase of \( C/T \) of the pure Ce-alloys compared to the diluted compounds is due to the additional entropy required for the antiferromagnetic ordering.

3. Discussion and conclusions

In order to study the antiferromagnetic transition of CeNi₈CuGe₄ in a more quantitative manner, we take a closer look at the \( C/T \)-difference of the diluted and undiluted systems. Due to the fact, that only the pure Ce-compounds order antiferromagnetically, the difference in the specific heat \( \Delta C/T \) provides an entropy, which only belongs to the formation of long range magnetic order. The left picture in Fig. 3 displays \( \Delta C/T \), showing the contribution of the antiferromagnetic ordering, and the associated entropy \( S \) in CeNi₈CuGe₄. The estimated value of the entropy \( S = 1.2 \text{ J/molK} \) is about 20\% of the theoretical expected value of \( R \ln 2 \approx 5.8 \text{ J/molK} \). This is in line with the presence of a partially Kondo-screened long range antiferromagnetic order which has been analyzed in terms of the resonant-level model model by Schotte and Schotte in Ref. [3]. A model calculation with a RKKY coupling parameter \( J = 2.3 \text{ K} \) and a Kondo temperature \( T_K = 1.3 \text{ K} \) approximately reproduces the reduced magnitude of the AF specific heat anomaly and the enhanced electronic specific heat anomaly of CeNi₈CuGe₄. This calculation implies Kondo-screening of an ordered Ce-moment along the \( c \)-axis which reduces the Ce moment to 36\% of its CF ground state value of \( \mu_c \). Considering a reduction of the local symmetry at the Ce-sites due to substitutional disorder present in CeNi₈CuGe₄ we expect some reduction of \( \mu_c \) as compared to CeNi₉Ge₄ with \( \mu_c = 2\mu_B \) [4] for the CF ground state. The Kondo-screened ordered moments of CeNi₈CuGe₄ are thus expected to range between 0.5 – 0.7 \( \mu_B \).

Further details can be drawn from the calculation of the enthalpy \( H \). Therefore the difference of the specific heat \( \Delta C \) was integrated as displayed in the right panel in Fig. 3. From the estimated value of \( H = 0.35 \text{ J/mol} \) an internal magnetic field of \( B = 0.13 \text{ T} \) is determined, using

![Figure 2.](image-url)
Figure 3. (Color online) The differences of $C/T$ (left panel) and $C$ (right panel) of CeNi$_8$CuGe$_4$ and La$_{0.5}$Ce$_{0.5}$Ni$_8$CuGe$_4$ and the resulting entropy $S$ and enthalpy $H$, respectively.

the relation $H = N_A 0.5 \mu_B B$ with the reduced magnetic moment of $0.5 \mu_B$ discussed above. This means that an external magnetic field of about $0.13 \text{T}$ should lead to a suppression of the longrange antiferromagnetic order. With the knowledge of this critical magnetic field an estimation of the Néel-temperature $T_N = 87 \text{ mK}$ can be made which is by a factor of two smaller than the observed Néel-temperature $T_N \approx 175 \text{ mK}$ of CeNi$_8$CuGe$_4$ [3]. From our thermodynamic considerations, taking into account the experimental Néel-temperature, a reduced Ce magnetic moment of $0.24 \mu_B$ would be expected.

4. Summary
Comparative studies of the specific heat and the magnetic susceptibility on the diluted system La$_{0.5}$Ce$_{0.5}$Ni$_9-x$Cu$_x$Ge$_4$ verify that the behavior of the pure Ce system CeNi$_9-x$Cu$_x$Ge$_4$ in the none ordered magnetic region ($x \leq 0.4$) is driven by a single-ion Kondo-effect. In the magnetic ordered phase ($x > 0.4$) the Kondo-effect still influences the magnetic ordering, leading to a reduction of the magnetic moments and therefore to a reduced antiferromagnetic contribution in the specific heat at $T_N$, as it is also predicted in [3], where a resonant-level model in combination with a molecular field approach is used. Thermodynamic calculations support these results.

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