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# Evolution of single-ion crystal field and Kondo features in $Ce_{0.5}La_{0.5}Ni_{9-x}Cu_xGe_4$

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**Abstract.** Starting with the heavy fermion compound $CeNi_9Ge_4$, 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 $CeNi_{9-x}Cu_xGe_4$ 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 $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ 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 $CeNi_{9-x}Cu_xGe_4$ 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 $CeNi_8CuGe_4$.

## 1. Introduction

One of the most outstanding Fermi-liquid systems is the heavy fermion compound $CeNi_9Ge_4$, which turns out to display the largest ever recorded value of the electronic specific heat $\Delta C/T \approx 5.5\,\mathrm{Jmol^{-1}K^{-2}}$ without showing any magnetic order [1]. The dilution of the magnetic Cerium $4f$-moments in $Ce_{1-x}La_xNi_9Ge_4$ reveals single-ion scaling with regard to the Ce-concentration [2]. Therefore, the unique behavior of $CeNi_9Ge_4$ 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 $CeNi_{9-x}Cu_xGe_4$ for $x > 0.4$ which culminates in a transition temperature of $T_N = 175\,\mathrm{mK}$ for $x = 1$ [3]. Even though the maximum of the magnetic specific heat $\Delta C(T_N)$ of $CeNi_8CuGe_4$ 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 $x \simeq 0.4$, 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 $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ and compared them to the pure $CeNi_{9-x}Cu_xGe_4$ series.

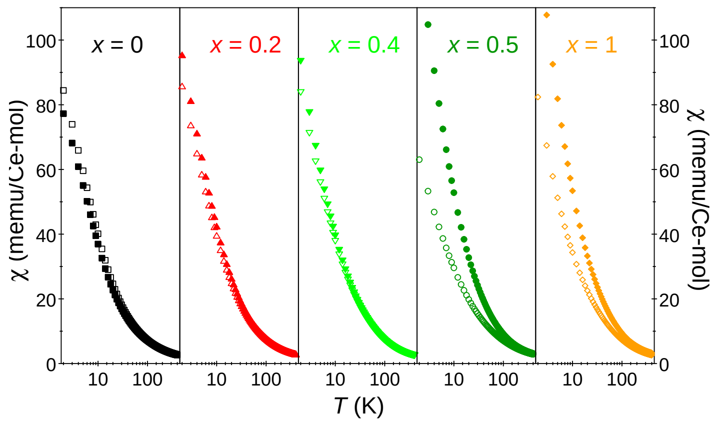


**Figure 1.** (Color online) The magnetic susceptibility $\chi$ of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ normalized per Ce-mol. The filled symbols represent the La-diluted samples, while the open symbols represent the pure Ce-compounds $CeNi_{9-x}Cu_xGe_4$ for identical Cu-contents.

## 2. Experimental Details

Polycrystalline samples of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ were prepared by arc melting the pure elements under argon atmosphere. Subsequently the samples were annealed at 950° 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 $LaFe_9Si_4$-type structure (space group $I4/mcm$). For details of the preparation and the measurements on the pure Ce-compounds $CeNi_{9-x}Cu_xGe_4$ see [3].

Figure 1 shows the magnetic susceptibility $\chi(T)$ in the temperature range $2\,K < T < 400\,K$ of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ 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 $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ with the corresponding solid solution $CeNi_{9-x}Cu_xGe_4$ 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 ($\chi_{\mathrm{Ce-mol}}$), we first subtracted the nonparamagnetic La-contribution and then scaled the data with the Ce-concentration, using the following equation:

$$\chi_{\mathrm{Ce-mol}} = 2 \cdot \Big( \chi(La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4) - 0.5 \cdot \chi(LaNi_9Ge_4) \Big) \qquad (1)$$

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 $\mu_{\mathrm{eff}} \approx 2.5\mu_B$, 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 $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ in the temperature range between 0.05 K and $T < 300\,K$. As already

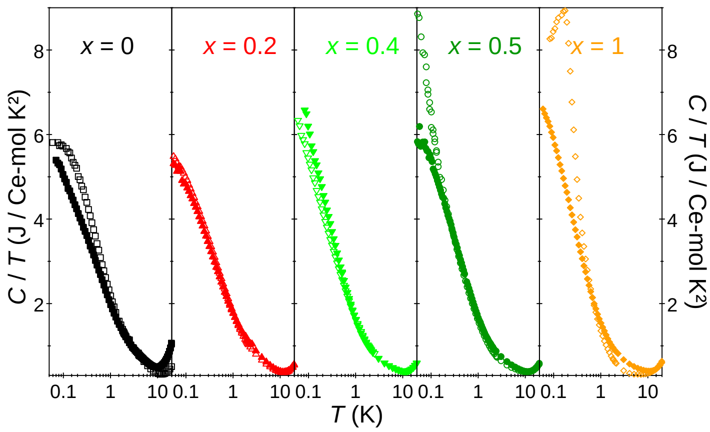


**Figure 2.** (Color online) The specific heat $C$ divided by temperature $T$ of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ normalized per Ce-mol (filled symbols) and of the undiluted Ce-compounds $CeNi_{9-x}Cu_xGe_4$ (open symbols).

known from literature [2] a normalization to $CeNi_9Ge_4$ 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 $CeNi_8CuGe_4$ 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 $CeNi_8CuGe_4$. The estimated value of the entropy $S = 1.2$ J/molK is about 20% of the theoretical expected value of $R \ln 2 \approx 5.8$ 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 $J = 2.3$ K and a Kondo temperature $T_K = 1.3$ K approximately reproduces the reduced magnitude of the AF specific heat anomaly and the enhanced electronic specific heat anomaly of $CeNi_8CuGe_4$. 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 $CeNi_8CuGe_4$ we expect some reduction of $\mu_c$ as compared to $CeNi_9Ge_4$ with $\mu_c = 2\mu_B$ [4] for the CF ground state. The Kondo-screened ordered moments of $CeNi_8CuGe_4$ 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 $H = 0.35$ J/mol an internal magnetic field of $B = 0.13$ T is determined, using

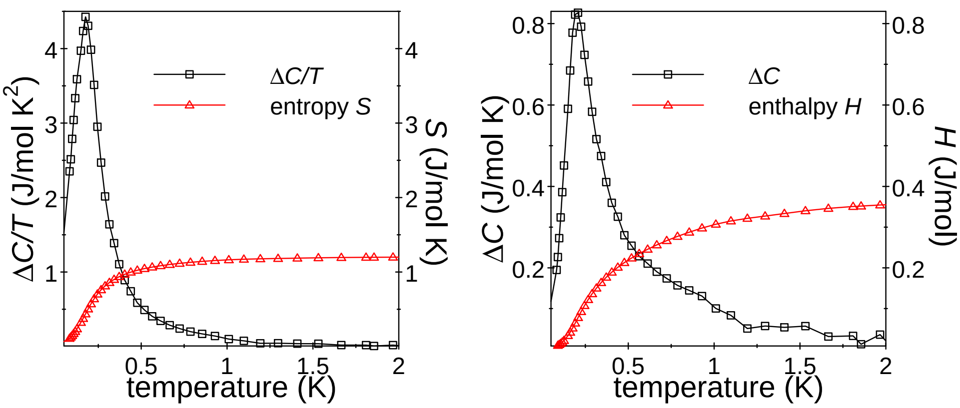


**Figure 3.** (Color online) The differences of $C/T$ (left panel) and $C$ (right panel) of $CeNi_8CuGe_4$ and $La_{0.5}Ce_{0.5}Ni_8CuGe_4$ and the resulting entropy $S$ and enthalpy $H$, respectively.

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 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 $T_N = 87$ mK can be made which is by a factor of two smaller than the observed Néel-temperature $T_N \approx 175$ mK of $CeNi_8CuGe_4$ [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 $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ verify that the behavior of the pure Ce system $CeNi_{9-x}Cu_xGe_4$ 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.

## 5. Acknowledgments

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