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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 χ 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 $T_\mathrm{N} = 175\,\mathrm{mK}$ for $x = 1$ [3]. Even though the maximum of the magnetic specific heat $\Delta C(T_\mathrm{N})$ 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 χ 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.

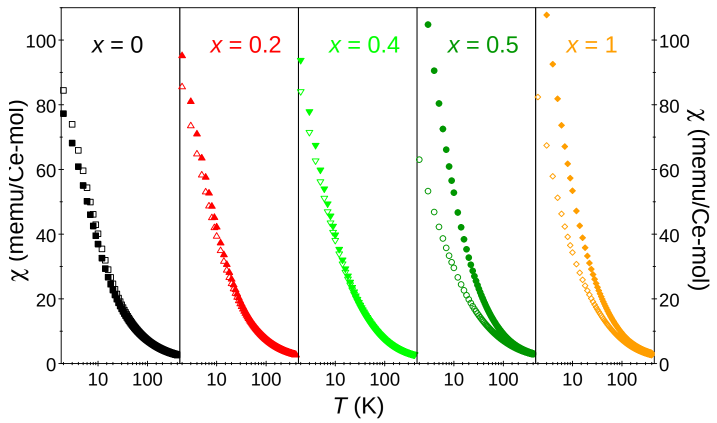

Figure 1. (Color online) The magnetic susceptibility χ of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ normalized per Ce-mol. The filled symbols represent the La-diluted samples, while the open symbols represent the pure Ce-compounds $CeNi_{9-x}Cu_xGe_4$ for identical Cu-contents.

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 (χ_{Ce-mol}), we first subtracted the nonparamagnetic La-contribution and then scaled the data with the Ce-concentration, using the following equation:

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

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 $\mu_{eff} \approx 2.5\mu_B$, 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

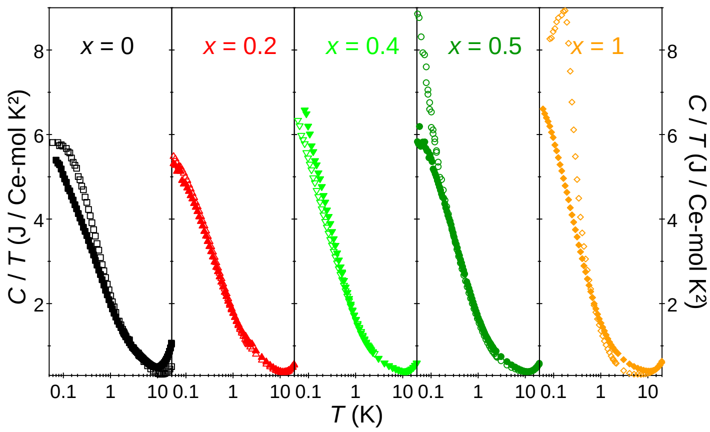

Figure 2. (Color online) The specific heat C divided by temperature T of $La_{0.5}Ce_{0.5}Ni_{9-x}Cu_xGe_4$ normalized per Ce-mol (filled symbols) and of the undiluted Ce-compounds $CeNi_{9-x}Cu_xGe_4$ (open symbols).

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 μ_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 μ_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 ΔC 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

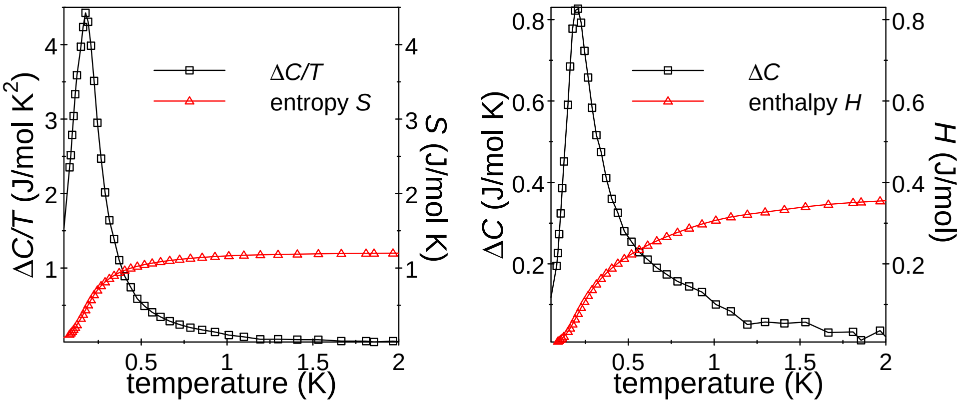

Figure 3. (Color online) The differences of C/T (left panel) and C (right panel) of $CeNi_8CuGe_4$ and $La_{0.5}Ce_{0.5}Ni_8CuGe_4$ and the resulting entropy S and enthalpy H, respectively.

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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