## Non-Fermi liquid behavior in Ce $(Cu_{1-x}Ni_x)_2Ge_2$ and Ce $Cu_2(Si_{1-y}Ge_y)_2$

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## Abstract

The heavy-fermion compounds  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  and  $CeCu_2(Si_{1-y}Ge_y)_2$  were explored using electrical transport, magnetic susceptibility, and neutron scattering measurements. The hybridization of  $CeCu_2Ge_2$  can be changed by substituting either Cu by Ni or Ge by Si. From the local type of antiferromagnetism in  $CeCu_2Ge_2$  to the heavy-fermion superconductor  $CeCu_2Si_2$ , the systems reveal a rich magnetic phase diagram with up to three distinct phase transitions below 5 K. In the case of substituting Cu by Ni, the system shows a transition from a local-type of ordering to a band-like magnetism of heavy-fermions. For the concentration  $x \approx 0.8$  the magnetic order is supressed and systematic deviations from the Fermi-liquid behavior are observed in the temperature dependence of the resistivity.

In CeCu<sub>2</sub>Ge<sub>2</sub> the energy scales of the intersite RKKY and the onsite Kondo interactions are of the same magnitude. The antiferromagnetic (AFM) order below  $T_{\rm N} = 4.15$  K is an incommesurate sinusoidal amplitude-modulated structure of localized, Kondo-compensated moments of  $1.05 \pm 0.1 \mu_{\rm B}$  [1,2]. From single crystal neutron scattering it follows that below approximately 1 K the propagation vector does not change and there are indications of a lock-in phase transition [2] to a commensurable structure. This is confirmed by measurements of the specific heat, which show several anomalies in the temperature range between 1 and 2 K [3].

To study the competition between the RKKY and the Kondo interactions, we have investigated the systems  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  and  $CeCu_2(Si_{1-y}Ge_y)_2$  in great detail. The effect of alloying is twofold, first the volume of the unit-cell is reduced and second the electronic density of states at the Fermi surface is changed. As seen in Fig. 1(a), in the case of substituting Ge by Si the AFM order temperature  $T_N$  decreases with increasing Ge concentration. For y = 0.05 no magnetic order could be detected but a broad transition to a superconducting ground state appears at  $T \approx 0.35$  K. CeCu<sub>2</sub>Si<sub>2</sub> is a superconductor with  $T_c \approx 0.67$  K [4]. The supression of the magnetic order corresponds to the increasing hybridization strength which is measured by the Kondo-lattice temperature  $T^*$  and is mainly due to the reduction of the volume [5].

For Ge concentrations y > 0.3, a second transition temperature  $T_2$  is seen in resistivity and in AC susceptibility measurements [6]. The magnetic structure below  $T_N$  is almost the same as in pure CeCu<sub>2</sub>Ge<sub>2</sub>. The transition at  $T_2$  is due to a change of the spin direction, for  $T > T_2$  the spins are confined to the [110] planes, below  $T_2$  they point perpendicular

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Fig. 1. Magnetic phase diagrams of  $CeCu_2(Si_{1-y}Ge_y)_2$  and  $Ce(Cu_{1-x}Ni_x)_2Ge_2$ . The solid line indicates the Néel temperature  $T_N$ . In  $CeCu_2(Si_{1-y}Ge_y)_2$  two additional transitions at  $T_2$  and  $T_3$  can be seen.  $T_c$  gives the border to the superconducting phase. In  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  two different antiferromagnetic phases are displayed (see Ref. [1]). The dash-dotted line represents the Kondo-lattice temperature  $T^*$  (determined by  $T(\rho_{max})$ ) and refers to the right axis. The top axis indicates the unit cell volume.

to these planes. The propagation vector does notchange at  $T_2$  and is very similar for the measured concentrations y = 0.4, 0.6 and 0.8. The origin of the third transition at  $T_3$  is not completely resolved but may be due to a lock-in phase transition as discussed in Ref. [2] for CeCu<sub>2</sub>Ge<sub>2</sub>.

The hybridization of the system can also be changed by substituting Cu by Ni.  $T^*$  increases in this system from  $T^* \approx 7$  K for x = 0 to  $T^* \approx 30$  K in CeNi<sub>2</sub>Ge<sub>2</sub>. In contrast to the Si–Ge alloy, the ordering of the local magnetic moments is suppressed for small Ni concentrations. For x < 0.3 two magnetic phase transitions occur at  $T_{\text{N1H}}$  and  $T_{\text{N1L}}$ , the structure of these phases are similar to each other. Up to now this regime of the phase diagram is not clarified. The system shows a transition from a local type of AFM ordering to a heavy-fermion band magnetism for concentrations 0.5 < x < 0.75 [7]. For  $x \ge 0.8$  no magnetic order can be detected.

The main aim of the present investigation is the search for non-Fermi liquid behavior close to the critical concentrations were the magnetic order is suppressed. This regime may well be described using the concept of a T = 0 K quantum phase transition in an itinerant fermion system [8, 9]. In addition to magnetic susceptibility and neutron scattering measurements [2, 6], detailed investigations of the electrical resistivity were performed for  $0.1 \le T \le 300$  K.



Fig. 2. Temperature dependence of the normalized resistivity  $\rho(T)/\rho(300 \text{ K})$  of  $\text{CeCu}_2(\text{Si}_{1-y}\text{Ge}_y)_2$  and  $\text{Ce}(\text{Cu}_{1-x}\text{Ni}_x)_2\text{Ge}_2$ . The curves for different y and x are shifted for clarity. The arrows indicate two magnetic transitions at  $T_{\text{NIL}}$  and  $T_{\text{NIH}}$  for  $\text{Ce}(\text{Cu}_{0.95}\text{Ni}_{0.05})_2\text{Ge}_2$ .

As displayed in Fig. 2, the temperature dependence of the resistivity exhibits two distinct maxima for all concentrations of  $CeCu_2(Si_{1-y}Ge_y)_2$ . The maxima at  $T_{CF} \approx 100$  K is due to crystal field excitations and does not significantly change for different concentrations. The low temperature maximum corresponds to the Kondo-lattice temperature  $T^*$  [1, 10]. In  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  the maxima at  $T_{CF}$  and  $T^*$  can barely be seen for concentrations higher than x = 0.75. For  $CeNi_2Ge_2$  the resistivity decreases monotonically to lower temperatures and the Kondo and crystal field peaks cannot be seperated any more.

In Fig. 3 the low temperature resistivity of  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  is plotted for the concentrations x = 0.65, 0.8 and 1.0. The residual resistivity  $\rho_0$  for  $CeNi_2Ge_2$  is  $\rho_0 = 7 \mu\Omega$  cm and increase to  $\rho_0 = 600 \mu\Omega$  cm for x = 0.65 due to the increasing disorder and can be described by the Nordheim factor x(1-x). We find  $(\rho - \rho_0) = AT^n$  up to nearly 2 K where the exponent *n* strongly depends on the concentration *x*. In the AFM regime (x < 0.8) the resistivity is proportional to  $T^2$  as predicted by the spin-fluctuation theory for a weak itinerant AFM in the ordered phase [11]. The same parabolic temperature dependence, however, is also characteristic for a coherent heavy Fermi-liquid. The prefactor *A* is correlated to the Kondo-lattice temperature via



Fig. 3. Low temperature resistivity  $(\rho - \rho_0)$  versus T of  $Ce(Cu_{1-x}Ni_x)_2Ge_2$ . The lines represent fits with a power law  $T^n$  up to 1.3 K. The arrow indicates the magnetic order for x = 0.65 at 3.2 K. For  $x \ge 0.8$  no magnetic order is detected. The curves for different x are displaced for clarification. The inset shows the exponent n as a function of Ni concentration x.

 $\sqrt{A} \propto 1/T^* \propto \gamma \propto m^*$  which agrees with the values obtained by the present experiments. For  $0.8 \le x \le 1$ no magnetic phase transition can be seen down to 0.1 K. Here we found significant deviations from the  $T^2$  law and the exponent yields values  $1.5 \le n \le 1.75$ (see inset of Fig. 3). The exponent  $n = \frac{3}{2}$  observed in CeNi<sub>2</sub>Ge<sub>2</sub> points to a non-Fermi liquid behavior in this compound. This value is expected for a heavy fermion system around an AFM instability [8, 9]. In the compound CeCu<sub>2</sub>(Si<sub>1-y</sub>Ge<sub>y</sub>)<sub>2</sub> the critical concentration (where  $T_N \rightarrow 0$ ) shifts towards y = 0. Depending on the sample preparation, CeCu<sub>2</sub>Si<sub>2</sub> reveals two distinct modifications. The magnetic 'A phase' develops out of a Fermi-liquid regime [12]. In difference the superperconducting 'S phase' possibly evolves from a non-Fermi liquid regime.

In conclusion, we have studied the magnetic state and the hybridization in  $Ce(Cu_{1-x}Ni_x)_2Ge_2$  and  $CeCu_2(Si_{1-y}Ge_y)_2$ . With decreasing Ge concentration  $T_N$  decreases due to the volume reduction, however, the magnetic structure does not change significantly. The substitution of Cu by Ni leads to a more drastical change of the magnetic state. The weak itinerant AFM order is supressed for x > 0.8and a crossover to a non-Fermi liquid behavior in  $CeNi_2Ge_2$  can be obtained.

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