DC magnetic susceptibility of CeCoGe$_{2.36}$Si$_{0.64}$ under high pressure

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Abstract. We report an investigation of the magnetic phase diagram of the heavy fermion compound CeCoGe$_{2.36}$Si$_{0.64}$ using DC magnetic susceptibility measurements under high pressure up to 10 kbar. The antiferromagnetic order that develops at ambient pressure below about 5.5 K remains essentially unaffected by pressure in the investigated pressure range up to 10 kbar. On the other hand, moderate magnetic fields appear to induce a quantum critical point in a sample subject to a pressure of 2 kbar. We discuss the role of disorder in the series of compounds CeCoGe$_{3-x}$Si$_x$.

Introduction
CeCoGe$_{3-x}$Si$_x$ is a pseudo-ternary heavy fermion (HF) compound that crystallizes in the tetragonal BaNiSn$_3$-type structure. It orders antiferromagnetically for $0.5 < x < 1$ and shows short range magnetic order (SRMO) from $x = 1$ up to $x_C = 1.25$ [1]. The vanishing of any signature of magnetic order together with the observation of non-Fermi liquid (NFL) behavior around $x_C$ defines this critical concentration as the $x$-induced quantum critical point (QCP). It was argued that disorder, due to the possible site exchange of Co and Ge at the 2$a$ and 4$b$ sites, strongly influences the quantum criticality of this system [2]. More recently, electrical resistivity experiments under high pressure on samples with $x = 0, 0.75$ and 0.9 indicated that, for the substituted samples, unquenched magnetic moments persist even at the QCP [3, 4, 5], calling for further investigations. Here, we report DC magnetic susceptibility measurements under pressures up to 10 kbar on a sample with $x = 0.64$, which is expected to be less disordered than the above samples with higher $x$ [4, 5]. Our aim was to investigate the pressure-induced quantum criticality of CeCoGe$_{2.36}$Si$_{0.64}$ expected from the above investigations; however, up to the highest pressures reached in our experiment no QCP could be induced. Instead, a quick suppression of the Néel temperature was observed when applying a magnetic field to an $x = 0.64$ sample at a pressure of 2 kbar.

Experimental
Polycrystalline CeCoGe$_{3-x}$Si$_x$ samples with the concentrations $x = 0, 0.64$, and 0.8 were prepared by arc melting, using high-purity elements. According to EDX and x-ray diffraction all samples are single phased. Magnetic susceptibility measurements were carried out in a S700X SQUID magnetometer down to 2 K. Hydrostatic pressures up to 10 kbar were applied to samples of typical dimension $4 \text{ mm} \times 1.2 \text{ mm} \times 1.2 \text{ mm}$, using an easy lab M10 pressure cell with Sn as in-situ manometer. Absolute values of the magnetic susceptibility of the sample inside the pressure cell were obtained by subtracting the pressure cell contribution, determined at different pressures and magnetic fields.
Results and Discussion

CeCoGe$_{3-x}$Si$_x$ is one of the rare examples of HF compounds where tuning with different control parameters leads to different behavior of physical properties in the vicinity of the QCP. For instance, $x$-tuning leads to the typically observed [6] shallow bump of the residual resistivity $\rho_0$ around the critical concentration $x_C$ [1]. Pressure tuning, on the other hand, leads to a step-like decrease of $\rho_0$ near the critical pressure $P_C$ [5]. This is illustrated in the inset of Fig. 1(a) where $\rho_0$ is plotted for both experiments as function of the normalized distance to the QCP $\delta = (x-x_C)/x_C$ and $\delta = (P-P_C)/P_C$, respectively.

Fig. 1: (Color online) (a) Temperature--pressure phase diagram of CeCoGe$_{3-x}$Si$_x$ for the different Si contents $x = 0.75$ [4], $x = 0.9$[5], and $x = 0.64$ (present data). The inset shows the residual resistivity $\rho_0$ as a function of the distance $\delta$ to the quantum critical point induced by pressure for $x = 0.9$ and by Si substitution in CeCoGe$_{3-x}$Si$_x$ (see text). The dashed line separates the magnetic (M) from the non-magnetic (NM) phase. (b) Unit cell volume (left axis), and estimated ($P_C^t$, see text) and experimental determined ($P_C$) critical pressure (right axis) vs $x$. The dashed lines are guides to the eye.

Figure 1(b) shows that the unit cell volume decreases linearly with increasing Si content $x$, confirming the previously reported [1] formation of homogeneous solid solutions in CeCoGe$_{3-x}$Si$_x$.

Assuming a linear relation between volume compression and pressure, $(V-V_C)/V = \kappa \times P$, with a constant compressibility $\kappa = 1 \times 10^{-3}$ kbar$^{-1}$ as measured for CeCoGe$_3$ [1], we have estimated the critical pressure ($P_C^t$) as function of $x$. $V_C$ is the volume of CeCoGe$_{3-x}$Si$_x$ at the critical Si concentration $x_C = 1.25$. A comparison of $P_C^t$ with experimentally determined critical pressures ($P_C$) is shown in Fig.1(b). While $P_C^t$ is linear in $x$ by construction, $P_C$ decreases at first faster, and then more gradually with increasing $x$. We attribute this non-linear behavior to the increasing importance of disorder with increasing $x$. 


Fig. 2: (Color online) (a) Temperature variation of the DC magnetic susceptibility, $\chi(T)$, of CeCoGe$_{2.36}$Si$_{0.64}$ at different pressures. (b) $\chi(T)$ at 2 kbar under different magnetic fields. The arrows indicate the onset of AF order and FL behavior, respectively (see text). The inset shows the low-temperature data for 0.1 T and 5 T, measured under ZFC and FC conditions (see text). The arrow points to the temperature where a weak bifurcation in $\chi(T)$ is observed.

Figure 2(a) shows the temperature variation of the magnetic susceptibility $\chi(T)$ of the heavy fermion CeCoGe$_{2.36}$Si$_{0.64}$ at different pressures, measured after zero-field cooling (ZFC) in an applied magnetic field of $B = 0.1$ T. At ambient pressure, a pronounced and relatively narrow peak is observed that signals the onset of antiferromagnetic (AF) order below $T_N = 5.5$ K. This peak is reduced in height and broadens as $P$ increases up to 4 kbar. Beyond that pressure no further appreciable change in the profile of $\chi(T)$ is observed up to 10 kbar. $T_N$, determined as the peak position of $\chi(T)$, is plotted as function of $P$ in Fig. 1(a). $T_N$ shows only very weak pressure dependence up to the highest pressure of 10 kbar. Comparing this behavior with the $T_N(P)$ dependencies of the $x = 0.75$ and 0.9 samples one might have expected to access a QCP at pressures of the order of 10 kbar in CeCoGe$_{2.36}$Si$_{0.64}$. The apparent need for sizable higher pressures could be due to the reduced disorder in the $x = 0.64$ sample.

Figure 2(b) shows $\chi(T)$ of CeCoGe$_{2.36}$Si$_{0.64}$ at 2 kbar, under different magnetic fields measured in both ZFC and field cooling (FC) experiments. With increasing field, the AF transition is shifted to lower temperatures. Simultaneously, the peak height of $\chi(T)$ is reduced and the peak broadens. For $B > 3$ T, $\chi(T)$ does not show a maximum down to 2 K, but instead appears to saturate to a constant value below a characteristic temperature $T_{FL}$, indicating a crossover to Fermi liquid (FL) behavior. However, experiments both at lower temperatures and in smaller magnetic field steps are needed to confirm this assertion.

The inset of Fig.2(b) enlarges the low-temperature behavior at 0.1 and 5 T, for both ZFC and FC experiments. A very weak bifurcation may be discerned in the 0.1 T data below $T_N$. Recent investigations have shown a much larger difference between ZFC and FC susceptibilities for a sample with $x = 0.8$ [7]. This supports our above conclusion that disorder in CeCoGe$_{3-x}$Si$_x$ is sizable reduced with decreasing $x$. 

Fig. 2: (Color online) (a) Temperature variation of the DC magnetic susceptibility, $\chi(T)$, of CeCoGe$_{2.36}$Si$_{0.64}$ at different pressures. (b) $\chi(T)$ at 2 kbar under different magnetic fields. The arrows indicate the onset of AF order and FL behavior, respectively (see text). The inset shows the low-temperature data for 0.1 T and 5 T, measured under ZFC and FC conditions (see text). The arrow points to the temperature where a weak bifurcation in $\chi(T)$ is observed.
Conclusions

In conclusion, we have investigated the temperature--pressure phase diagram of CeCoGe$_{2.36}$Si$_{0.64}$ using DC magnetic susceptibility measurements. The Néel temperature, which is about 5.5 K at ambient pressure, is hardly affected by pressures up to 10 kbar, indicating that higher pressures are needed to reach a pressure-induced quantum critical point. However, by simultaneously applying magnetic fields, the Néel temperature is quickly suppressed. For CeCoGe$_{2.36}$Si$_{0.64}$ under a pressure of 2 kbar a quantum critical point appears to be induced by magnetic fields of the order of 5 T. The disorder in CeCoGe$_{2.36}$Si$_{0.64}$ is judged to be less pronounced than in the previous investigated CeCoGe$_{3-x}$Si$_x$ samples with $x = 0.75$ and 0.9.

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References


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