High pressure phase diagram of CeCoGe\textsubscript{2.2}Si\textsubscript{0.8}

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Abstract. We have investigated the temperature-pressure phase diagram of the heavy fermion compound CeCoGe\textsubscript{2.2}Si\textsubscript{0.8} by DC magnetic susceptibility measurements, $\chi_{DC}(T)$, under high pressure. The Néel temperature of $T_N = 4$ K in zero pressure is reduced by pressure up to 3 kbar. At higher pressures antiferromagnetic order appears to gradually transform into a spin glass like-state. Magnetic field decreases both $T_N$ and the spin glass freezing temperature $T_f$. At 3 T and 6.5 kbar a divergence of $\chi_{DC}(T)$ is observed with a power law that is consistent with a disorder-dominated quantum criticality.

1. Introduction
A challenging problem in the physics of strongly correlated electron systems is to understand the role of disorder, when matter approaches a quantum phase transition. The non-Fermi liquid (NFL) behavior and non-canonical phase diagrams around the quantum critical point (QCP) of a number of U and Ce-based heavy fermion (HF) systems [1, 2, 3] appear to be well described by the quantum Griffiths model [4, 5]. It describes the coexistence of a Fermi liquid bath, in which the Kondo interactions dominates and rare magnetically ordered regions, in which the RKKY interaction dominates. To date there are, however, few systematic investigations on the role of disorder on the quantum critical properties at pressure or magnetic field tuned QCPs.

The pseudo-ternary HF compound CeCoGe\textsubscript{3-x}Si\textsubscript{x} [6] with tetragonal BaNiSn\textsubscript{3}-type structure and antiferromagnetic (AF) order for $x = 0.75$ ($T_N = 6$ K) and 0.9 ($T_N = 3.9$ K) is one of the few examples of heavy fermion compounds that can be driven to a QCP under moderate high pressures ($< 10$ kbar) [7, 8]. Recent electrical resistivity measurements under pressure ($P$) [8] for $x = 0.9$ have suggested that the QCP ($P_C = 6.2$ kbar) might be governed by the coexistence or competition between a Griffiths phase and critical spin fluctuations. Here, we report DC magnetic susceptibility measurements under high pressures for CeCoGe\textsubscript{2.2}Si\textsubscript{0.8}, a material with a Si content similar to that recently investigated by electrical resistivity measurements under pressure [8].

2. Experimental
Polycrystalline CeCoGe\textsubscript{2.2}Si\textsubscript{0.8} samples were prepared in two steps as described in Ref.6. Precursors were first melted in an arc furnace and, in the second step, annealed at 900 °C for
two weeks in a sealed quartz tube under low pressure of argon. Powder X-ray diffraction, EDX and magnetic susceptibility measurements did not reveal any secondary phases. DC magnetic susceptibility measurements were carried out in a S700X SQUID magnetometer down to 2 K. Hydrostatic pressures up to 10 kbar were applied on a sample with dimension 5.5 mm×1.55 mm×1.35 mm using a CuBe pressure cell (easy lab M10) and Sn as in-situ manometer. The absolute value of the sample magnetic susceptibility was obtained by subtracting the pressure cell contribution at different pressures and magnetic fields from the measured data.

3. Result and discussion

Figure 1(a) shows the temperature variation of the magnetic susceptibility, $\chi_{DC}(T)$, at all investigated pressures. $\chi_{DC}(T)$ at $P = 0$ kbar shows the typical profile of a material that undergoes long range antiferromagnetic ordering at the Néel temperature $T_N$, defined as the temperature where $\chi_{DC}(T)$ has a maximum. The extracted value of $T_N = 4$ K is in good agreement with values recently derived from specific heat and electrical resistivity measurements on samples of the same batch (not shown). The residual resistivity $\rho_0 = 24 \mu\Omega$ cm is comparable to values reported by Eom et al. [6], but three times smaller than the value reported for a polycrystalline sample with $x=0.9$ [8]. This indicates that our sample is of good quality. We would also like to note that $\rho_0$ of our CeCoGe$_2$Si$_{0.8}$ samples is comparable to $\rho_0$ values of a number of other heavy fermion systems that are not considered to be disorder dominated [3, 9].

Pressure reduces the magnitude of $\chi_{DC}$ and shifts $T_N$ continuously to lower temperatures, at least up to 5 kbar (Fig. 1a). Beyond that pressure, the reduction of the magnitude of $\chi_{DC}$ is less pronounced and a different profile of $\chi_{DC}(T)$ develops, with a rounded hump centered at $T_f$. With increasing pressure $T_f$ first increases slightly up to 8 kbar and then becomes constant. This indicates a change in the type of order at a pressure of about 5 kbar. Further evidence for this conjecture comes from zero field cooling (ZFC) and field cooling (FC) $\chi_{DC}(T)$ measurements in an applied field of 0.1 T. While for pressures below 5 kbar a clear maximum is present for both the ZFC and the FC $\chi_{DC}(T)$ curves (Fig. 1 b), for pressures above 5 kbar only the ZFC curve displays a very broad maximum (Fig. 1 c, d). The hysteresis in the former case (Fig. 1 b) is characteristic of antiferromagnets with strong magnetic anisotropy and pronounced domain effects. The hysteresis in the latter case (Fig. 1 c, d) is reminiscent of a spin glass-type behavior.
(SG), probably governed by an interplay of AF and ferromagnetic (FM) interactions. This is summarized in the temperature-pressure phase diagram shown in Fig. 2.

![Figure 2. Temperature-pressure phase diagram of CeCoGe$_2$Si$_{0.8}$ obtained by magnetic susceptibility experiments (Fig. 1). Dashed colored lines separate the antiferromagnetic (AF) and the spin glass-like behavior (SG) regimes. The fully colored area under the square symbols separates the AF and SG regimes and represents an intermediate regime where the magnetic susceptibility show a non-monotonic change with pressure.](image)

A pressure-induced change of the AF ground state may also be inferred from our $\chi_{DC}(T)$ measurements in different magnetic fields (Fig. 3). At zero pressure, $\chi_{DC}(T_N)$ is continuously reduced by the magnetic field (Fig. 3 a) as is $T_N$ (inset of Fig. 3 a). For 6.5 kbar, on the other hand, $\chi_{DC}(T_f)$ at first increases with increasing field (Fig. 3 b). Unfortunately, $T_f$ quickly moves out of the investigated temperature window, calling for measurements below 2 K. At 3 T, $\chi_{DC}(T)$ increases strongly with decreasing temperature and may be approximated by $\chi_{DC} \propto T^{-0.14(2)}$. A power law divergence $\chi \propto T^{-\eta}$ with $0.1 < \eta \leq 0.3$ is expected for a disorder dominated QCP [5].

![Figure 3. Temperature dependence of the magnetic susceptibility of CeCoGe$_2$Si$_{0.8}$ at different magnetic fields for (a) 0 kbar and (b) 6.5 kbar, measured at ZFC conditions with $B = 0.1$ T. The arrow at $T_N$ marks the onset of antiferromagnetic order while the arrow at $T_f$ is the freezing temperature that marks the onset of spin glass-like behavior. The inset shows the magnetic field variation of $T_N$. The black full line on the $\mu_0H = 3$ T data represents a fit $\chi_{DC} \propto T^{-0.14(2)}$ below 9 K.](image)

We speculate that the putative SG state induced by pressured above 5 kbar consists of FM domains coupled via short range AF correlations to each other. This scenario is compatible with the formation of a Griffith phase: random substitution of Ge by Si leads to the formation of rare magnetic regions which become critical and play a relevant role when the system approaches the QCP [10].
In conclusion, we have investigated the temperature-pressure phase diagram of the heavy fermion material CeCoGe$_2$Si$_{0.8}$ using magnetic susceptibility measurements down to 2 K, at pressures up to 10 kbar. The data suggest that an antiferromagnetically ordered ground state characterized by strong magnetic anisotropy and domain effects is successfully suppressed by pressure and transformed into a state reminiscent of a spin glass. It is noteworthy that for CeCoGe$_{2.1}$Si$_{0.9}$ a faster suppression of $T_N$ with pressure was observed [8]. From that behavior we would have expected to reach a quantum critical point in CeCoGe$_{2.2}$Si$_{0.8}$ at a pressure of about 7 kbar. While we could not confirm this, we can also not exclude it since the putative spin glass phase appearing at pressures above 5 kbar might cover an underlying quantum critical point.

Application of magnetic fields suppresses both the antiferromagnetic and the putative spin glass order. Measurements below 2 K are required to further explore field-tuned quantum critical points at different pressures.

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