From an antiferromagnet to a heavy-fermion system: CeCu$_5$Au under pressure

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The electrical resistivity $\rho(T)$ of single crystalline CeCu$_5$Au under pressure was measured in the temperature range 30 mK < $T$ < 300 K. Pressure suppresses the antiferromagnetic order ($T_N = 2.35$ K at ambient pressure) and drives the system into a non-magnetic heavy-fermion state above $P_c = 4.1(3)$ GPa. The electrical resistivity shows a deviation from a $T^2$ dependence of a Fermi–liquid in the pressure range 1.8 GPa < $P$ < 5.15 GPa. The $\rho(T)$–curves can be compared with those of CeCu$_{6-x}$Au$_x$ at different Au concentrations. Just before the long-range magnetic order vanishes, a possibly superconducting phase (at $T_c = 0.1$ K and $P = 3.84$ GPa) occurs, pointing to a coexistence of antiferromagnetic order and superconductivity. This new phase is only seen in a narrow pressure interval $\Delta P = 0.4$ GPa.

\[ \text{phase transition, high pressure, electrical resistivity, non–Fermi–liquid} \]

1. Introduction

In many heavy-fermion (HF) metals antiferromagnetic (AFM) quantum critical phenomena have been observed. Ternary Ce–based compounds like CePd$_2$Si$_2$, CeRu$_2$Ge$_2$, and CeCu$_{6-x}$Au$_x$ can be tuned to a quantum critical point (QCP) either by pressure or doping. According to the spin–fluctuation theory $^4$ $^5$ $^6$ $^7$ $^8$ $^9$, the temperature dependence of the electrical resistivity $\Delta \rho(T) \propto T^\nu$ shows a deviation from the conventional Fermi–liquid (FL) behaviour ($n = 2$) in a limited temperature range. Furthermore, the Néel temperature $T_N$ approaches zero in a characteristic way: $T_N \propto (g_c - g)^m \rightarrow 0$, where $g$ and $g_c$ are the tuning parameter (like concentration $x$ or pressure $P$) and its critical value. For AFM fluctuations the exponent is $m = 2/3$. This prediction has not been found unequivocally in experiment. Recent high pressure experiments on CeRu$_2$Ge$_2$ $^2$ gave $m = 0.70 \pm 0.08$ but reported variations on other compounds are mainly linear in $x$ or $P$ (for more details see Ref. $^2$ and references therein).

The anomalous exponents found for the temperature dependence of $\rho(T)$ of CePd$_2$Si$_2$ and CeNi$_2$Ge$_2$ ($n = 1.2$ $^1$ and 1.37 $^8$, respectively) as well as the logarithmic temperature dependence of the specific heat $C/T \propto \ln(T/\lambda)$ observed in CeCu$_{6-x}$Au$_x$ (see e. g. Ref. $^3$) point to a fundamental breakdown of FL theory $^1$. The results on high quality samples of CePd$_2$Si$_2$ $^1$ and CeNi$_2$Ge$_2$ $^3$ have shown that such a non–Fermi–liquid (NFL) behaviour occurs in a small temperature interval. Furthermore, close to the QCP superconductivity emerges in some cases. On the other hand, it is not clear if disorder will modify or even produce the various NFL properties. In a recent theoretical explanation $^1$ it was argued that the anomalies in $\rho(T)$ can be attributed to the interplay between quantum critical AFM fluctuations and impurity scattering in a conventional FL. Such a consideration, however, is not relevant for the $\Delta \rho(T) \propto T$ behaviour, observed in CeCu$_{5.9}$Au$_{0.1}$ $^3$, where the anisotropy of the spin fluctuations seems to play an important role $^2$ $^3$ $^4$ $^5$.

The stoichiometric CeCu$_5$Au compound has the highest $T_N = 2.35$ K of all CeCu$_{6-x}$Au$_x$ alloys with $0.1 \leq x \leq 1.0$ $^4$ $^7$ where $T_N$ varies linearly with $x$. At the critical concentration $x_c = 0.1$ the system loses its long–range magnetic order and the Kondo effect dominates the RRKY interaction ($T_K = 6.2$ K in CeCu$_6$ $^4$). At very low temperature ($T < 0.3$ K) a FL behaviour is found in CeCu$_6$ $^4$. The $T_N(x)$ variation seems to be related to the increase of the unit–cell volume upon doping, but changes in the band structure have to be considered if the different $T_N(x)$ and $T_N(P)$ dependence at equal volume have to be explained. Since in the alloys a certain disorder and a structural transition at low temperature (for $x < 0.15$) exist, measurements on the stoichiometric and single crystalline CeCu$_5$Au $^7$ offer a unique possibility to study the pure pressure or volume effect on the magnetic ordering temperature.

Here we present electrical resistivity measurements on single crystalline CeCu$_5$Au under high pressure ($P < 8$ GPa). The four–point resistance was measured on a sample in a clamped high pressure cell which was cooled down in a dilution refrigerator ($T > 30$ mK). Details of the high pressure set–up can be found elsewhere $^3$.

2. Results and Discussion

Representative electrical resistivity curves $\rho(T)$ of CeCu$_5$Au are shown in a semi–logarithmic plot in Fig. 1. The low temperature part of the ambient pressure curve is identical to that reported in Ref. $^3$. Below $T_N = 2.35$ K the antiferromagnetically ordered phase is entered, clearly visible by the pronounced cusp in $\rho(T)$. A negative logarithmic slope is present above $T_N$ up to 10 K, reflecting the presence of the Kondo effect ($T_K = 1.8$ K $^3$). A maximum in $\rho(T)$ develops at higher temperature ($T_{\text{max}} \approx 60$ K). It becomes less pronounced and...
shifts slightly down in temperature as pressure is applied. At a moderate pressure of \( P = 2.98 \) GPa a second maximum at \( T_{\text{max}}^{\text{low}} = 3.5 \) K appears, which is well separated from the entrance into the AFM state at \( T_N = 1.4 \) K. It might be related to an already enhanced Kondo effect and could point to the development of a coherent state. The \( \rho(T) \)-curve at \( P = 3.84 \) GPa shows a peculiar low temperature behaviour. The entrance into a magnetically ordered phase at \( T_N \approx 1 \) K is still visible but at \( T = 0.1 \) K the resistivity drops suddenly by more than 10\%, indicating the occurrence of a new phase. Its possible nature will be discussed below. At this pressure the two maxima in \( \rho(T) \) are still present whereas at higher pressure only \( T_{\text{max}}^{\text{high}} \) remains and a FL behaviour is observed at low temperature. We mention that the residual resistivity exhibits a strong pressure dependence (cf. Fig. 1) and details will be given elsewhere [19].

In Fig. 2 the pressure dependence of the characteristic temperatures in \( \rho(T) \) of CeCu\(_5\)Au in a semilogarithmic plot. The Néel temperature \( (T_N) \) scales to zero at \( P_c = 4.1(3) \) GPa. Two maxima in \( \rho(T) \) at \( T_{\text{max}}^{\text{low}} \) and \( T_{\text{max}}^{\text{high}} \), related to the Kondo effect, seem to merge above 4 GPa. In one experiment (open symbols) indications of a new phase (possibly superconducting) below \( T_c = 0.1 \) K (\( \odot \)) was found. In the inset the \( T_N(P) \) dependence is shown in a linear plot.

The Kondo temperature in CeCu\(_5\)Au at low pressure is small in comparison to the crystal field (CF) splitting \( \Delta^{\text{(1)}}_{\text{CF}} \approx 100 \) K and \( \Delta^{\text{(2)}}_{\text{CF}} \approx 160 \) K [20]. Therefore, as often observed in this situation for other compounds, the magnetic resistivity has two maxima at \( T_{\text{max}}^{\text{low}} \) and \( T_{\text{max}}^{\text{high}} \), whose high temperature sides are a sign of the Kondo scattering on the ground state and excited CF levels, respectively, and whose low temperature sides reflect the onset of a coherent heavy–fermion state and the freezing of scattering from CF levels. The pressure variation of \( T_{\text{max}} \) might point to the possibility of an enhanced screening (induced by pressure) of the magnetic moments by the conduction electrons and thus to an strengthened role of the Kondo effect. Consequently, the anomaly at \( T_{\text{max}}^{\text{low}} \) has to be related to \( T_K \). Both anomalies in \( \rho(T) \) seem to merge above 4 GPa, indicating the entrance into an intermediate valence regime where the Kondo temperature becomes of the order of the CF splitting.

In the case of two excited CF levels Hanazawa et al. [21] have introduced a second Kondo temperature at high temperature \( T_K = \sqrt[\text{3}]{T_K^{(1)}} \Delta^{(1)}_{\text{CF}} \Delta^{(2)}_{\text{CF}} \). With the assumption that \( \Delta^{(1)}_{\text{CF}} \) and \( \Delta^{(2)}_{\text{CF}} \) are hardly changed at low pressure (i. e. \( P < 4 \) GPa), the \( T_K \) values can be calculated if \( T_K \) is known. For some CeCu\(_{6-x}\)Au\(_x\) compounds \( T_K \) has been
determined [22]. To transform the $T_K(V(x))$–dependence in a $T_K(P)$–dependence the relative unit–cell volumes $V(x)/V_0$, with $V_0$ the unit–cell volume of CeCu$_5$Au at ambient pressure, have to be transformed into the corresponding pressure values. With the Murnaghan equation of state (EOS) [23] a $T_K(P)$ relation, applicable for CeCu$_5$Au, can be deduced (using $B_0 = 110$ GPa and $B'_0 = 4$). This (linear) function then yields the $T_K(P)$ dependence. The $T_K(P)$ values are practically identical to $T_{\text{high}}(P)$ in the pressure range $2 < P < 3$ GPa. At lower pressure the agreement is not so good which might be related to the presence of magnetic order.

![Graph](image)

**FIG. 3.** The exponent $n$ used in the power law of eq. (1) to describe the $\rho(T)$ data of CeCu$_5$Au below $T = 0.3$ K. Around $P = 3.5$ GPa a clear deviation from a FL ($n = 2$) behaviour is observed. The dashed line is a guide to the eye.

Applying pressure to stoichiometric and single crystalline CeCu$_5$Au offers the possibility to study the low temperature properties close to the magnetic instability and to compare them with the CeCu$_{6-x}$Au$_x$ solid solution (with $x \rightarrow x_c$). The electrical resistivity of CeCu$_5$Au below 0.3 K can be described with

$$\rho = \rho_0 + A T^n$$  \hspace{1cm} (1)

at all pressures. The exponent $n$ and the coefficient $A$ are fitting parameters. The only “constraint” to the fit was the fixation of the upper temperature limit $T_A = 0.3$ K. It is a compromise between an as narrow as possible temperature interval (30 mK < $T < 300$ mK) and the reliability of the deduced parameters, i. e. $n$ and $A$. Figure 3 illustrates how the deviation from a FL description ($n = 2$) evolves with pressure. Below 1.2 GPa $n = 2$ is consistent with residual electron–magnon scattering in a magnetic system [24]. Then, at 1.8 GPa, $n$ suddenly attains a value of 1.75 and decreases further as pressure increases. At 3.5 GPa $n = 1.51$ is reached, close to the critical value $n = 3/2$, predicted by theory [18]. Increasing pressure further, leads to a higher $n$ value which finally reaches $n = 2$, well inside the non–magnetic region ($P \geq 5.37$ GPa) and comparable to CeCu$_6$ [24]. The minimum in $n$ vs $P$ is not an artefact of the limited temperature interval in the fitting procedure as the fits for various $T_A$ values (up to 0.6 K) showed always a minimum in $n(P)$ around 3.5 GPa, where $n$ is the smaller ($n = 1.2$) the higher the $T_A$ limit (0.6 K) was chosen. In the high pressure non–magnetic region however, the FL value $n = 2$ was found in temperature intervals which became enlarged with pressure.

This behaviour immediately raises the question whether these results can be compared to the observations in CeCu$_{6-x}$Au$_x$ with different Au concentrations. It is clear that the unit–cell volume variation is a crucial parameter. Thus, a correspondence of the $x$ values in CeCu$_{6-x}$Au$_x$ to the pressure values in CeCu$_5$Au is needed. Using the relation $V(x) = 420.225 \, \text{Å}^3 + 13.988x$, deduced from x–ray data [15] and e. g. the Murnaghan EOS, a relation between $x$ and $P$ can be obtained. For CeCu$_5$Au, this results in a bulk modulus $B_0 = 110$ GPa (with $B'_0 = 4$) leading to the correspondence $x = 0.5 \Leftrightarrow P = 1.8$ GPa, $x = 0.1 \Leftrightarrow P = 3.4$ GPa, and $x = 0 \Leftrightarrow P = 3.85$ GPa which should be taken as a guide rather than as a strict prediction.

![Graph](image)

**FIG. 4.** Electrical resistivity $\rho(T)$ of CeCu$_5$Au normalized at $T = 0.1$ K at pressures close to the magnetic instability. The strong drop in $\rho(T)$ at $T = 0.1$ K and $P = 3.84$ GPa is interpreted as the entrance into a (probably) superconducting phase. Inset: The anomaly in $\rho(T)$ at $\approx 1$ K is interpreted as a sign of magnetic order.

Now the peculiar low temperature behaviour of the
$\rho(T)$ curves recorded close to the magnetic instability are of particular interest (Fig. 4). At $P = 3.84$ GPa the entrance into a magnetically ordered phase at $T_N \approx 1$ K is still visible (inset Fig. 4) but at $T = 0.1$ K, $\rho(T)$ drops suddenly more than 10%. This effect can be suppressed if a small magnetic field ($\vec{B} \parallel \vec{c}$, $B = 0.2$ T) is applied. Traces of this transition are also present at $P = 4.19$ GPa, where the resistivity starts to decrease, but not as strong as at the preceding pressure. No signs of a magnetically ordered phase were found. Hence, if magnetism and superconductivity coexist, it occurs in a very narrow pressure range. Well above this pressure, no anomalies in the low temperature part of $\rho(T)$ were found (see curve at $P = 5.37$ GPa in Fig. 4).

If the new phase should be found unequivocally to be a superconducting phase, the measurements show that low residual resistivity is not an important ingredient for superconductivity for CeCu$_3$Au. Just before $\rho(T)$ of CeCu$_3$Au starts to decrease, the resistivity is close to 40 $\mu\Omega$cm (at 0.1 K and $P = 3.84$ GPa). Furthermore, as in the other pressure-induced HF superconductors, superconductivity then would emerge in the vicinity of the magnetic instability. Therefore, it is an intriguing possibility that AFM spin fluctuations may provide the attractive interaction between quasi–particles which is required to form Cooper–pairs [1]. However, additional experiments are necessary to clarify this point.

3. Conclusion

The electrical resistivity measurements on single crystalline CeCu$_3$Au showed that the long–range magnetic order is suppressed at $P_c = 4.1(3)$ GPa. Close to the magnetic instability the electrical resistivity (below $T = 0.3$ K) deviates from a Fermi–liquid behaviour. At $P = 3.84$ GPa the pronounced drop in $\rho(T)$ at $T_N = 0.1$ K might point to the existence of a superconducting phase. At this pressure an AFM order is still present ($T_N \approx 1$ K), leading to the possibility to study the coexistence of AFM order and superconductivity as well as NFL behaviour close to a quantum critical point.

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