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LETTER TO THE EDITOR

Pressure-induced residual resistivity anomaly in $CeCu_5Au$

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Abstract

The electrical resistivity of the magnetically ordered $CeCu_5Au$ has been investigated under pressure up to 8.5 GPa. In the magnetically ordered region ($p < 3.4$ GPa) the residual resistivity ρ_0 shows a pronounced maximum as a function of pressure. Even in the non-magnetic region ρ_0 decreases monotonically by more than a factor of three. These two effects can be qualitatively explained in terms of the interplay of pressure, magnetism and disorder in a strongly correlated electron system with weak disorder.

The behaviour of the electrical resistivity ρ of metals when approaching zero temperature T has been of interest since the beginning of solid state physics. While it is clear that the residual resistivity ρ_0 of conventional metals is largely governed by lattice defects and impurities, the situation is much less clear for metals with strong electronic correlations and/or magnetic order at low T. In the independent-electron approximation, $\rho_0 = m^*/e^2 n\tau = \frac{3}{2}\pi(h/e^2)/(k_F^2 l)$, where m^* is the effective mass, τ the scattering time arising from electron scattering by defects and impurities, l the corresponding mean free path, n the electron density and k_F the Fermi wave-vector. In heavy-fermion (HF) systems, m^* is enhanced by up to a factor of 1000 due to the Kondo effect arising from the exchange coupling of conduction electrons and nearly localized f-electrons. Nevertheless, ρ_0 is rather low in stoichiometric compounds: the quasi-particles obey Bloch's theorem and scatter at $T = 0$ only from rare defects. The simple independent-electron expression above suggests that hydrostatic pressure p should affect ρ_0 only to a minor extent since both k_F and l should depend on p only through the (small) change of electron density and interatomic distances.

In this letter, we report ρ_0 measurements under quasi-hydrostatic pressure on the stoichiometric HF compound $CeCu_5Au$ where heavy quasi-particles, leading to a Sommerfeld coefficient $\gamma = 0.64$ J mol^{-1} K^{-2} for $T \to 0$, coexist with incommensurate antiferromagnetic (AF) order [1]. The magnetic ordering temperature T_N in this system can be suppressed by pressure [2]. Surprisingly, a strong dependence of ρ_0 on p is found: starting from $\rho_0 = 28$ $\mu\Omega$cm for $p = 0$, ρ_0 passes over a pronounced maximum of 58 $\mu\Omega$cm at 1.8 GPa and

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decreases to 20 $\mu\Omega$cm at ≈ 3 GPa. Even in the non-magnetic Fermi-liquid state ($p \gtrsim 3.4$ GPa), ρ_0 decreases substantially upon further pressure increase, reaching 6 $\mu\Omega$cm at 8.5 GPa.

The single crystal of $CeCu_5Au$ (space group *Pnma*) used in this study was grown with the Czochralski method from the starting constituents Ce (4N), Cu (5N), and Au (4N). A part of the crystal was used for neutron experiments which revealed a very good quality of the single-phase sample [3]. Two small rectangular pieces ($35 \times 73 \times 861$ μm^3 and $28 \times 85 \times 561$ μm^3) cut from the same crystal were used in two pressure experiments. A clamped-anvil high-pressure device [4], capable of reaching 10 GPa, was used to measure $\rho(T)$ by the four-point method for a current I along the crystallographic b direction, i.e. perpendicular to the magnetic ordering vector $\boldsymbol{Q}$ [3]. The sample and the pressure gauge (a thin Pb foil) were embedded in a soft pressure-transmitting medium (steatite) to ensure quasi-hydrostatic pressure conditions in the pressure range explored here. Both samples showed the same $\rho_0(p)$ behaviour, ruling out an error due to a possible rearrangement of the contact leads under pressure.

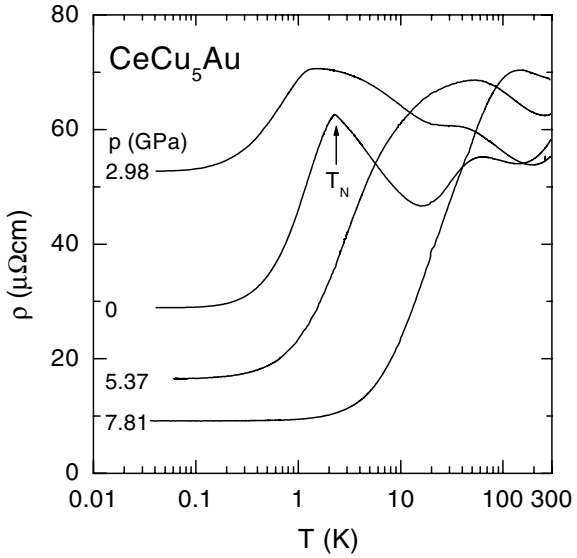

Figure 1. Electrical resistivity $\rho(T)$ of $CeCu_5Au$ at selected pressures p in a semilogarithmic temperature scale.

The $\rho(T)$ data of the larger sample are shown in figure 1. The AF phase transition at ambient pressure occurs at $T_N = 2.35$ K, in perfect agreement with specific-heat and magnetization measurements [1]. Reducing the unit-cell volume by external pressure tunes T_N to zero [2]. This behaviour is well known for a number of HF systems and is attributed to an increase of hybridization between conduction electrons and local moments, thus favouring the Kondo effect over the RKKY interaction. The detailed analysis of these data [2] reveals a critical pressure $p_c \approx 3.4$ GPa for the quantum-critical point (QCP) where $T_N \to 0$.

Figure 2 shows the pressure dependence of ρ_0, determined in the T range 30 mK $< T <$ 50 mK. The particularly striking observations are (i) the strong non-monotonic variation of ρ_0 within a few GPa where T_N is monotonically suppressed to zero, and (ii) the strong decrease of ρ_0 even in the paramagnetic state (for $p > 3.4$ GPa). The maximum in $\rho_0(p)$ is centred at $p \approx 1.8$ GPa, i.e., well inside the magnetic phase. The reversibility of this strong pressure effect was tested in one experiment. As the non-magnetic phase was reached ($p = 4.2$ GPa) the pressure was released down to 3.3 GPa and then increased

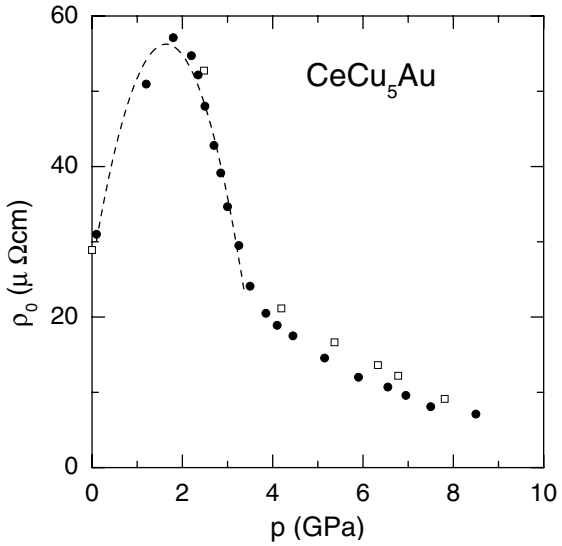

Figure 2. Residual resistivity ρ_0 of $CeCu_5Au$ against pressure p for a current I along the b axis. The two symbols denote results obtained on different samples. The line is a guide to the eye.

again. The ρ_0 values obtained in this pressure cycle (two during pressure release and two upon pressure increase) confirmed the strong $\rho_0(p)$ variation. The $\rho_0(p)$ in the magnetic region is in a superficial regard qualitatively similar to the $\rho_0(x)$ variation in $CeCu_{6-x}Au_x$ [5]. However, $\rho_0(x)$ with a maximum for $x = 0.5$ arises from x-dependent disorder and negative lattice pressure while external pressure is not expected to change the number of impurities in $CeCu_5Au$.

Pressure is not the only parameter to change ρ_0 drastically. Likewise, an external magnetic field B, applied along the (easy) crystallographic c direction, yields a reduction of ρ at low T. Figure 3 shows $\rho(T)$ curves at $p = 2.2$ and 3.5 GPa for $B = 0$ and 7.5 T. Well inside the magnetic region (for $p = 2.2$ GPa, $T_N = 1.35$ K), $B = 7.5$ T destroys the AF order and causes a decrease of ρ_0 by more than 60%. In the non-magnetic phase ($p = 3.5$ GPa), B affects $\rho(T)$ as well, but here the influence becomes weaker as $T \to 0$. Hence, the 'magnetic' contribution to ρ_0 decreases as the system is tuned away from the magnetic–non-magnetic instability. Furthermore, the variation of $\rho(p)$ in the external magnetic field supports that uniaxial stress does not play a role in our experiment.

The impressive $\rho_0(p)$ dependence of $CeCu_5Au$ should be regarded in the context of the p-induced ρ_0 changes in other HF systems. For $CeCu_2Ge_2$ a maximum is found well inside the non-magnetic phase as for $CeCu_2Si_2$ [6]. The magnitude of the ρ_0 peak in superconducting $CeCu_2Ge_2$ at a pressure close to the $T_c(p)$ maximum, can be associated with charge fluctuations [7] or even a valence transition. For $YbCu_2Si_2$ only a fraction of the Yb ions order magnetically at p_c [8] and the broad and shallow $\rho_0(p)$ maximum may correspond to full ordering at higher p. In the case of $CeRu_2Ge_2$ the $\rho_0(p)$ variation at low p might be related to a change of the magnetic ordering vector between low and intermediate p [9], but the effect is rather small.

The following discussion treats our major observations for $CeCu_5Au$, i.e., the (i) non-monotonic $\rho_0(p)$ dependence in the AF phase and (ii) the strong ρ_0 decrease for $p > p_c$, in terms of models that are generic to strongly correlated systems with some disorder.

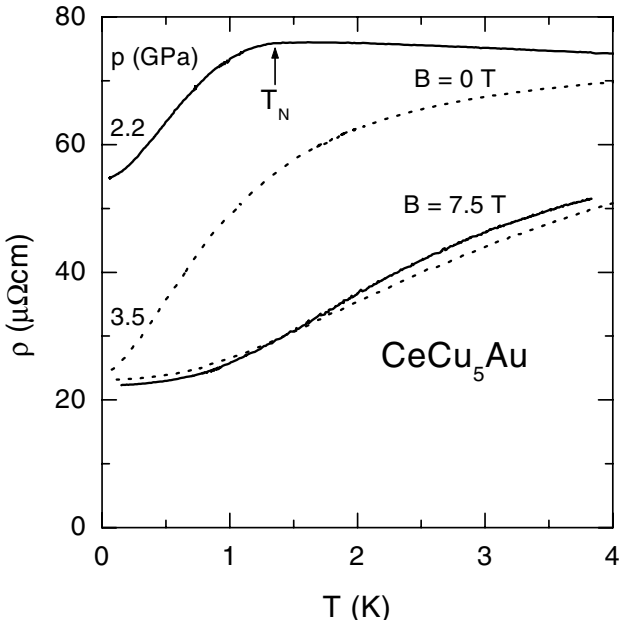

Figure 3. Electrical resistivity $\rho(T)$ of $CeCu_5Au$ for $p = 2.2$ GPa (solid lines) and $p = 3.5$ GPa (dotted lines) in zero magnetic field and $B = 7.5$ T.

We first focus on the AF phase and argue that the strong $\rho_0(p)$ dependence is a consequence of the quasi-particle scattering from local variations in the staggered magnetization. The qualitative picture is as follows: impurities change the chemical environment of the Ce atoms and lead to a large spatial variation of the local susceptibilities $\chi_L(\boldsymbol{r}) \approx 1/T_K(\boldsymbol{r})$ with a 'Kondo temperature' $T_K(\boldsymbol{r})$ which depends *exponentially* on the local environment. In the AF phase this leads to a variation of the local magnetization $\Delta M_L(\boldsymbol{r}) \propto f[H_{AF}/T_K^{imp}] - f[H_{AF}/T_K^{bulk}]$, where $H_{AF}(\boldsymbol{r})$ is a staggered (exchange) field proportional to the AF order parameter and $f[x]$ describes the crossover from weak magnetization $f[x \to 0] \propto x$ to a saturation of the magnetic moment for large H_{AF} (upper inset of figure 4). The electrons scatter off the variations of the (local) magnetization ΔM_L. Close to the QCP, $|\Delta M_L|$ increases with increasing staggered bulk magnetization M_{AF}. Deep in the AF phase, the local magnetization saturates, $H_{AF} \gtrsim \max[T_K^{imp}, T_K^{bulk}]$, and $|\Delta M_L|$ decreases in size. This explains qualitatively the pronounced maximum of $\rho_0(p)$. It also corresponds to the observation that close to the $\rho_0(p)$ maximum, T_N is of the order of the coherence temperature, measured by the maximum in $\rho(T)$ [2]. This signals a smooth crossover from a spin-density wave with small magnetic moments just below p_c to local-moment AF order with large moments as p approaches zero.

To substantiate this qualitative picture we have performed a microscopic calculation [10] for an Anderson-lattice model in the limit $U \to \infty$: $H = \sum_{ij,\sigma} t_{ij} c^\dagger_{i\sigma} c_{j\sigma} + V c^\dagger_{i\sigma} f_{i\sigma} + \epsilon^i_f f^\dagger_i f_i + U f^\dagger_{i\uparrow} f_{i\uparrow} f^\dagger_{i\downarrow} f_{i\downarrow}$. We mimic changes of p by a variation of the hybridization V. Increasing the values of V corresponds to a p increase. The local physics of this strongly correlated system was treated in the dynamical mean-field theory (MFT), the resulting self-consistent Anderson model was solved within slave-boson MFT [11]. While this approximation fails to describe the inelastic processes correctly, it is expected to give a qualitatively correct description for low T. The disorder is modeled by a small density of f-electron sites with a different local energy

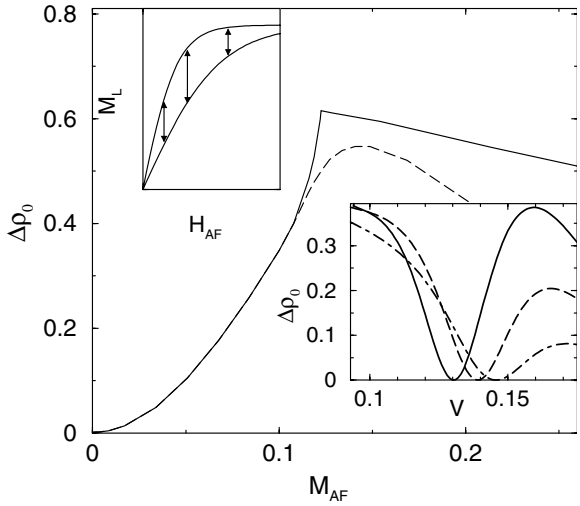

Figure 4. Upper inset: schematic plot of the local magnetization $M_{\rm L}$ as a function of a local magnetic field $H_{\rm AF}$ at a bulk site and an impurity site. In the AF phase the electrons scatter from local variations of the magnetization $\Delta M_{\rm L} = M_{\rm L}^{\rm imp} - M_{\rm L}^{\rm bulk}$ which leads to the pronounced maximum in $\Delta\rho_0$, shown in the main figure as a function of the bulk magnetization $M_{\rm AF}$ (for $\epsilon_f^{\rm imp} < \epsilon_f^{\rm bulk} < 0$). The cusp in the solid line is an artefact of the approximation used. The dashed line shows schematically the smooth maximum expected in a more realistic theory. Lower inset: $\Delta\rho_0$ in the paramagnetic phase as a function of the hybridization V (larger V corresponds to higher p) for three different values of $\epsilon_f^{\rm imp} > \epsilon_f^{\rm bulk}$.

$\epsilon_f^i = \epsilon_f^{\rm imp} \neq \epsilon_f^{\rm bulk}$. For simplicity, the long-range order is described by a staggered magnetic field with ordering wave vector $\boldsymbol{Q} = (1/2\,0\,0)$. In figure 4, $\Delta\rho_0$ in a direction perpendicular to $\boldsymbol{Q}$ (like in the experiment) as a function of $M_{\rm AF}$ shows indeed the maximum which we expected from the arguments given above. The fact that we obtain a sharp cusp instead of a smooth maximum in $\Delta\rho_0(M_{\rm AF})$ is a well known artefact of the MFT treatment of the single-impurity Anderson model where a spurious phase transition is induced when the magnetization is close to saturation.

The approximation used above neglects non-local effects which should be important in the AF phase close to the QCP, when the correlation length ξ is large. We estimate this effect in perturbation theory by considering the scattering from non-local variations of the staggered magnetization $\Delta M(\boldsymbol{r}) \approx \int \chi(\boldsymbol{r} - \boldsymbol{r}')\Delta M_{\rm L}(\boldsymbol{r}')/\chi_{\rm L}{\rm d}\boldsymbol{r}'$, where χ is the susceptibility of the AF phase, characterized by ξ. For independent impurities, small ΔM and $\Delta\rho_0 \ll \rho_0$ [12], we find an enhancement of $\Delta\rho_0$ for large ξ, $\Delta\rho_0 \propto \xi|\Delta M_{\rm L}|^2$. Unfortunately, there are no data on the p dependence of the magnetization $M_{\rm AF}$ or ξ. From MFT, we expect $\Delta M_{\rm L} \propto \sqrt{p_c - p}$ and $\xi \propto 1/\sqrt{p_c - p}$ and therefore $\Delta\rho_0 \sim \sqrt{p_c - p}$ close to p_c. This is not observed as $\rho_0(p)$ appears to rise linearly with $p_c - p$ and may be attributed to the fact that MFT strongly overestimates the increase of the size of $M_{\rm AF}$ as suggested by the x-dependence of $M_{\rm AF}$ in $CeCu_{6-x}Au_x$ [5]. Also the assumption of independent impurities breaks down close to the QCP. Furthermore, ξ may be too short in the relevant p regime (inelastic neutron scattering at the QCP of $CeCu_{6-x}Au_x$ reveals relatively broad structures [13]).

Other potentially important effects like the opening of a gap in some parts of the Fermi surface and an associated reduction of scattering rates [5] have not been included in our model calculation. We do not expect them to dominate the p dependence of ρ_0 measured

perpendicular to $\boldsymbol{Q}$. Likewise, our qualitative picture should hold also for the incommensurate ordering vector $\boldsymbol{Q}$ = (0.56 0 0) for $CeCu_5Au$ [5]. A semi-quantitative check for the local picture is a comparison with the magnetoresistivity at the QCP, where according to our local approximation a similar rise of ρ_0 is expected for not too strong fields. Indeed, in $CeCu_{5.9}Au_{0.1}$, ρ_0 (measured for I along the b direction) rises by approximately 20% for a local magnetization of 0.3 μ_B [14]. This is of the expected order of magnitude. A more precise comparison to our pressure data is not possible due to the different nature of the impurities in $CeCu_{5.9}Au_{0.1}$. Finally, we note that the strong field dependence in the range where $\rho_0(p)$ is large, is in qualitative agreement with our model.

We now turn to the pronounced $\rho_0(p)$ dependence in the paramagnetic region of the phase diagram which is indeed surprising from a theoretical point of view since in a Kondo lattice with non-magnetic impurities, ρ_0 is only weakly renormalized by interactions [15]. It is tempting to interpret the increase of ρ_0 upon approaching the QCP from the paramagnetic side as the signature of the spontaneous formation of magnetic domains of size ξ around certain disorder configurations. Such an interpretation seems not viable as the same strong variation of ρ_0 is observed at a considerable distance from the QCP, where the number of those domains should be exponentially small. We have therefore theoretically investigated the question whether a strong p dependence may arise from purely local effects.

Each impurity, i.e., a Kondo ion with a local environment differing from the bulk due to disorder, contributes according to Friedel's sum rule a factor $\Delta\rho \sim \sin^2 \pi \Delta n$ to ρ_0 if s-wave scattering dominates and the density of impurities is small. Δn is the total charge accumulated (in the s-wave channel) around the impurity. For the following argument we consider an impurity site where the f-electron is more strongly coupled to the environment than a typical bulk site ($\epsilon_f^{\text{bulk}} < \epsilon_f^{\text{imp}} < 0$). For high p or large V, the occupation of the f-orbital at the impurity site is therefore reduced and Δn is negative. For small V both the impurity site and bulk sites are deep in the Kondo regime and the number of f-electrons per site and spin is fixed at 1/2. Nevertheless, extra charge is accumulated around sites with the larger local T_K. For a *single-impurity* Kondo model this effect is of order T_K/E_F, with E_F the Fermi-energy, and therefore negligible. For a lattice Δn is of order 1 and positive for a particle-like Fermi surface. In a generic situation one therefore expects a sign change in Δn (for a particle-like Fermi surface) and correspondingly a *strong* p dependence of $\Delta\rho_0 \sim \sin^2 \pi \Delta n$ as is shown in the lower inset of figure 4. A similar argument holds in the case $\epsilon_f^{\text{bulk}} > \epsilon_f^{\text{imp}}$. Remarkably, in the limit $T_K^{\text{imp}} \ll T_K^{\text{bulk}}$ (or $T_K^{\text{imp}} \gg T_K^{\text{bulk}}$) Δn takes on a universal value which depends only on bulk properties [10].

With this mechanism, it is possible to explain the unexpected large variation of $\rho_0(p)$ in the paramagnetic phase as an intrinsic property of a weakly disordered Kondo lattice. We obtain changes of ρ_0 by a factor of ~ 3 accompanied by changes of V by a few percent and variations of T_K by a factor of 2–3, in reasonably good agreement with experiment. Our data impose a serious constraint towards theories for the non-Fermi liquid behaviour in $CeCu_5Au$. For example, we could not find any signature of a collapse of the Fermi surface due to a complete breakdown of the Kondo effect which has been suggested in [17]. Note that our analysis does not contradict the results of Kotliar and Varma [15] who find that in a Kondo lattice ρ_0 is only weakly renormalized by interactions due to non-magnetic impurities: the strong $\rho_0(p)$ variations are indeed 'weak' as they are not of order $1/T_K$. A prediction of this scenario is that ρ_0 rises again for even higher p upon entering the mixed-valence regime. From the fact that $\rho_0(p)$ in $CeCu_6$ decreases for p up to 8.5 GPa [16], we deduce that in $CeCu_5Au$ such an increase, if present, might occur at pressures well above 10 GPa.

In conclusion, we have observed a surprisingly strong variation of the residual resistivity

with pressure in both the antiferromagnetic and paramagnetic phase of $CeCu_5Au$. We argued that the pronounced $\rho_0(p)$ maximum in the AF phase can be attributed to scattering from inhomogeneities in the magnetization. The explanation for the strong decrease of ρ_0 in the paramagnetic phase by a factor of three is less obvious: a possible mechanism can nevertheless be found even in a purely local picture where the sign of the charge, which is accumulated in a s-wave channel, changes under typical conditions. More experimental and theoretical studies are needed to understand the interplay between the residual resistivity and the complex low-temperature properties of strongly correlated metals.

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