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An unconventional metallic state in $YbRh_2(Si_{1-x}Ge_x)_2$—a high pressure study

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Abstract

We present a detailed pressure study of the electrical resistivity $\rho(T)$ and the specific heat $C(T)$ of the non-Fermi-liquid (NFL) compound $YbRh_2Si_2$ and of $\rho(T)$ for a single crystal in which 5 at.% of Si is replaced by isoelectronic Ge. The magnetic phase diagram is deduced up to $p \cong 2.5$ GPa. A comparison of the effects of the volume change introduced by doping and/or by hydrostatic pressure will be given. We show that the NFL behaviour observed in $\rho(T)$ as well as the magnetic phase diagram are not influenced by the disorder introduced by alloying.

1. Introduction

The study of 3d- and 4f-electron compounds on the threshold of magnetism has brought to light a number of metals with electronic properties deviating significantly from the behaviour expected for a conventional Landau Fermi-liquid (FL) ($\Delta\rho \propto T^2$, $\Delta C/T =$ constant). On theoretical grounds, these non-Fermi-liquid (NFL) effects are expected not just at, but also in the vicinity of a quantum critical point (QCP) at which a magnetic ordering temperature vanishes continuously ($T_m \rightarrow 0$). In order to avoid the effect of disorder (e.g. produced by alloying) in the study of quantum critical phenomena it is highly desirable to investigate stoichiometric compounds. Unfortunately, only a few such heavy-fermion (HF) compounds exist which exhibit NFL behaviour at ambient pressure (e.g., UBe_{13}, $CeNi_2Ge_2$, $CeCu_2Si_2$) [1]. The number of stoichiometric candidates can be increased by utilizing hydrostatic pressure as a control parameter to drive magnetically ordering HF compounds close to a QCP (e.g., $CePd_2Si_2$, $CeIn_3$ [2]). A deeper understanding might be reached by widening the search to include Yb-based HF compounds. The electron–hole analogy between Ce- and Yb-based compounds ($4f^1$ Ce^{3+} and $4f^{13}$ Yb^{3+}) also offers an interesting alternative approach for realizing a QCP system [3] experimentally.

Recently, $YbRh_2Si_2$ was identified as a stoichiometric HF compound close to a weak antiferromagnetic phase transition at ambient pressure with a very low ordering temperature

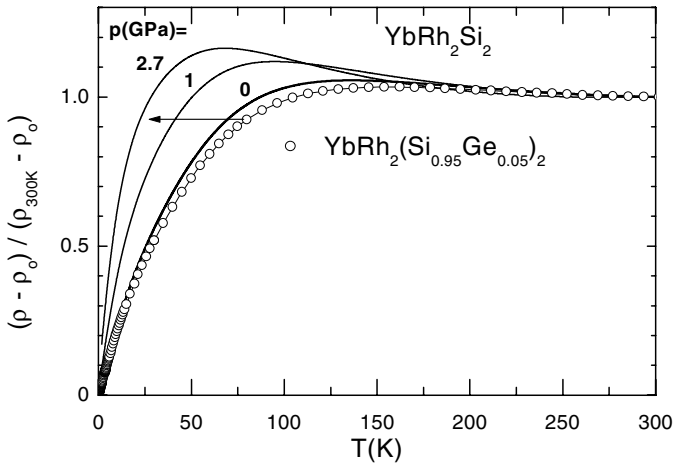

Figure 1. Electrical resistivity ρ versus T for $YbRh_2Si_2$ at ambient pressure, $p = 1$ GPa and $p = 2.7$ GPa (lines) and of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at ambient pressure (○).

T_m ($\widetilde{=}$70 mK) [4]. Therefore, $YbRh_2Si_2$ offers one of the few opportunities to determine thermodynamic quantities (like the specific heat) of a HF metal close to the QCP. This is the first observation of NFL effects in a stoichiometric Yb-based HF compound already at ambient pressure and zero magnetic field.

2. Experimental details

Single-crystalline samples of $YbRh_2Si_2$ and $YbRh_2(Si_{0.95}Ge_{0.05})_2$ (nominal Ge concentration) were grown from In flux as described elsewhere [5].

The electrical resistivity measurement was performed in a ^{3}He/^{4}He dilution refrigerator down to $T \cong 50$ mK using a standard four-point lock-in technique. Pressure was applied by utilizing a piston–cylinder cell. The pressure-transmitting medium was an equal mixture of n-pentane and 2-methylbutane to achieve almost hydrostatic conditions. Pressure was determined by monitoring resistively the pressure-induced shift of the superconducting transition temperature of Sn.

The specific heat was measured via a 'compensated heat-pulse method' [6] to establish almost adiabatic conditions for the whole assembly of the CuBe piston–cylinder pressure cell, the sample, the pressure-transmitting medium (Fluorinert) and a small piece of In to monitor the pressure. Several single crystals, with a total sample mass of about 0.5 g, were used to perform the experiment. The Yb increment to the specific heat $\Delta C(T)$ was determined by subtracting from the measured specific heat that of the Lu-based homologue $LuRh_2Si_2$.

3. Results and discussion

At ambient pressure the electrical resistivity (figure 1) of $YbRh_2Si_2$ is almost constant above $T \cong 100$ K and decreases towards lower temperatures indicating the development of coherence effects. At low temperatures, the resistivity is linear in T over a wide range in temperature [4] and shows an anomaly at $T_H \cong 70$ mK. This anomaly in the resistivity is most pronounced for the current $\boldsymbol{j}$ running perpendicular to the basal plane, but hardly resolvable for $\boldsymbol{j} \parallel a$-axis. Applying hydrostatic pressure, a maximum in the resistivity forms below $T = 100$ K, which

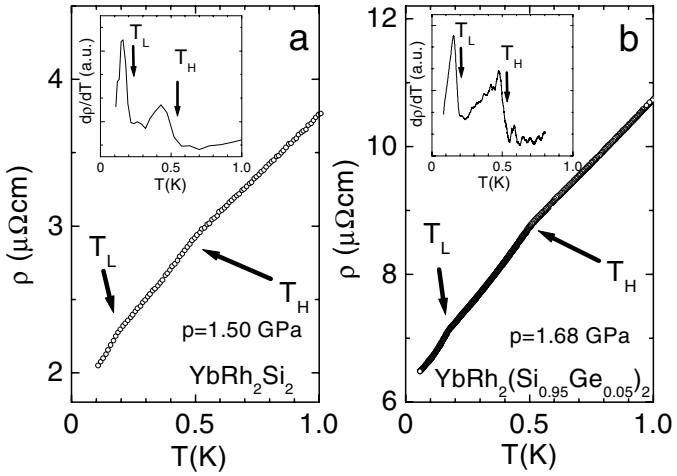

Figure 2. Resistivity ρ versus T for $YbRh_2Si_2$ at $p = 1.5$ GPa (a) and $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at $p = 1.68$ GPa (b). The derivative $d\rho/dT$ is utilized to determine T_H and T_L (insets).

shifts to lower temperatures, reflecting the decrease of the Kondo temperature T_K. The anomaly in the resistivity at T_H can be shifted to higher temperatures by applying pressure, as expected for an Yb system. At $p \geqslant 1.5$ GPa ($T_H = 0.5$ K), a second anomaly is observed at $T_L = 0.2$ K (figure 2(a)). The temperature derivative of $\rho(T)$ shows that both phase transition anomalies are of mean-field type (insets of figures 2(a) and (b)). With p increasing to above 1.5 GPa, the anomaly in $\rho(T)$ at T_H weakens, while the one at T_L becomes more pronounced (not shown).

Below $T \cong 10$ K the electronic contribution to the specific heat obeys $\Delta C(T)/T = D \log(T_0/T)$ over more than one decade in temperature [4]. On increasing the pressure, the slope of $\Delta C(T)/T$ becomes steeper, but the temperature range of $\Delta C(T)/T \propto -\log T$ is reduced to lower T, indicating the growing distance to the QCP (figure 3(a)). At $p > 1.2$ GPa, the phase transition anomaly expected from figure 2 ($T_H \cong 0.4$ K) becomes observable above the temperature limit of our ^{3}He-evaporation cryostat and shifts to $T_H \cong 0.5$ K at $p = 1.5$ GPa. The size of the jump of ΔC at T_H demonstrates that the anomalies observed in the magnetic susceptibility ($p = 0$) [4] as well as in $d\rho/dT$ are due to a bulk effect and intrinsic to this compound. The shape of $\Delta C(T)/T$ at $p = 1.5$ GPa resembles that of 'A-type' $CeCu_2Si_2$ in the vicinity of the QCP [8]. Upon increasing pressure, the jump of $\Delta C/T$ at T_H appears to be slightly reduced (figure 3(a)).

Application of a magnetic field $B = 2$ T at $p = 1.5$ GPa appears to be sufficient to suppress the magnetic phase transition rather than to shift it to $T < 0.4$ K. At $B = 8$ T and $T < 4$ K, $\Delta C(T)/T =$ constant at $p \leqslant 1.5$ GPa and decreases with increasing p. Similar sensitivity of $\Delta C(T)/T$ to B was found already at ambient pressure [4].

With the aid of these specific-heat measurements under hydrostatic pressure, the closeness of $YbRh_2Si_2$ to a magnetic instability is proven as an intrinsic property. In addition, this study shows the existence of NFL behaviour in $YbRh_2Si_2$ above T_H over a broad range in temperature and pressure.

Since T_m increases with decreasing unit-cell volume (increasing hydrostatic pressure), one expects to get even closer to the QCP when expanding the unit-cell volume. An extrapolation of the phase diagram of the pure compound to the negative pressure range gives a critical pressure of $p_c \cong -0.3$ GPa. This is the motivation for a substitution of 5 at.% of Ge for Si (which has a smaller ionic radius).

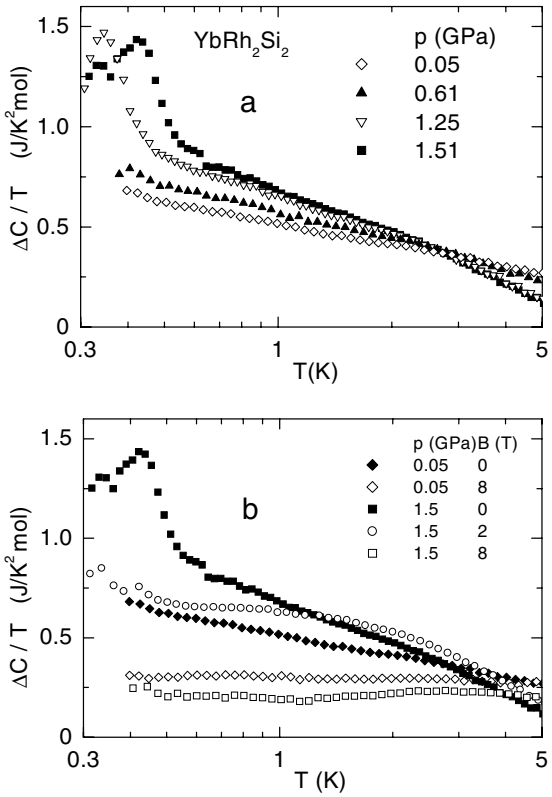

Figure 3. The Yb increment to the specific heat $\Delta C = C(YbRh_2Si_2) - C(LuRh_2Si_2)$ as $\Delta C/T$ versus $\log(T)$ for $p = 0.05$ (♦), 0.61 (▲), 1.25 (▼) and 1.51 (■) GPa (a) and for $p = 0.05$ GPa and $p = 1.51$ GPa and external magnetic field as indicated (b).

The effect of alloying on the electrical resistivity at high temperatures is compared in figure 1 to the effect of hydrostatic pressure. The steep decrease of $\rho(T)$ below $T \cong 100$ K is shifted in an opposite way by either expanding the volume or by application of pressure. This indicates an increase of the characteristic (Kondo) energy scale associated with the interaction between the 4f and the conduction electrons upon expanding the lattice. Therefore, magnetic ordering is expected to be suppressed in the doped compound. Indeed, no indication of a phase transition can be seen in measurements of the electrical resistivity and the AC magnetic susceptibility down to $T \cong 10$ mK [9]. The resistivity follows a linear temperature dependence above $T \cong 10$ mK up to $T \cong 10$ K, a remarkably large temperature range of three decades. Like in the stoichiometric samples, an anomaly can be observed in $\rho(T)$, when hydrostatic pressure is applied. For $p > 1.3$ GPa, also the second anomaly is visible in the accessible range of temperatures. The resistivity is nearly linear in T over a wide temperature range above the magnetic transition temperature within the whole pressure range explored. A comparison of the electrical resistivity for low temperatures at $p = 1.5$ GPa in the case of $YbRh_2Si_2$ and $p = 1.68$ GPa in the case of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ is shown in figure 2.

The magnetic phase diagram deduced from the measurements of the electrical resistivity is shown in figure 4 together with the data for the pure sample. For a better comparison, the data of the Ge-substituted compound are shifted by $\Delta p = -0.2$ GPa. The shifted data points and those of the stoichiometric compound fall on top of each other, demonstrating that neither

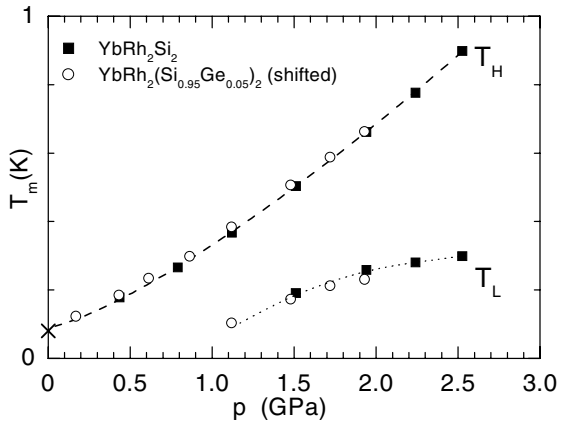

Figure 4. Pressure dependence of T_H and T_L as deduced from electrical resistivity measurements for $YbRh_2Si_2$ (■) and $YbRh_2(Si_{0.95}Ge_{0.05})_2$ (○). Data on the latter are shifted by $\Delta p = -0.2$ GPa. T_H and T_L represent the positions of the inflection points of $d\rho/dT$ versus T. The width of the transitions is $\Delta T \approx$ (20–40) mK over the pressure range explored. ×: T_H as deduced from magnetic susceptibility measurements at $p = 0$ in the pure compound. The dashed line represents $T_H \propto |\Delta p|^\alpha$ ($\alpha \cong 4/3$). The dotted line through T_L-values is a guide to the eye.

the shape of $T_H(p)$ nor that of $T_L(p)$ change upon alloying. Interestingly, the dependence of T_H on p shows positive curvature and, for the whole range of pressure investigated, can best be expressed as $T_H \propto |\Delta p|^\alpha$ with $\alpha \cong 4/3$ (figure 4).

4. Conclusions and outlook

The reported specific-heat measurements under hydrostatic pressure show that the antiferromagnetic transition in $YbRh_2Si_2$ is an intrinsic property. Slight doping with 5 at.% of Ge suppresses the magnetic anomaly, as expected from the application of 'negative' pressure in an Yb system, but does not induce changes to the NFL behaviour found in the electrical resistivity. The magnetic phase diagram of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ matches that of the stoichiometric compound by simply shifting the data uniformly by $\Delta p = -0.2$ GPa. This demonstrates that volume effects dominate the physical properties in $YbRh_2(Si_{0.95}Ge_{0.05})_2$ and that the influence of disorder caused by alloying is small. Therefore, $YbRh_2(Si_{1-x}Ge_x)_2$ offers the opportunity to tune the system exactly to the QCP by chemical substitution without being hampered by the influence of disorder. For the first time, this would allow a complete thermodynamic characterization of an Yb compound right at a QCP.

Acknowledgment

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