LOW-TEMPERATURE MAGNETIC AND TRANSPORT PROPERTIES OF THE CLEAN NFL SYSTEM $YbRh_2(Si_{1-x}Ge_x)_2$*

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High-quality single crystals of $YbRh_2(Si_{1-x}Ge_x)_2$ ($x = 0, 0.05$) have been studied by low-temperature (T) and high-magnetic-field (B) measurements of the electrical resistivity $\rho(T, B)$, dc-magnetization $M(T, B)$ and magnetic ac-susceptibility $\chi_{ac}(T, B)$. For the undoped compound ($x = 0$) a weak AntiFerromagnetic (AF) transition at $T_N \simeq 70\,\mathrm{mK}$ is observed which can be suppressed for small fields $B_c \simeq 0.06\,\mathrm{T}$ and $0.7\,\mathrm{T}$ applied along the crystallographic a- and c-direction, respectively. Below T_N, a $\rho(T) = \rho_0 + aT^2$ behavior is found with very low residual resistivity $\rho_0 \simeq 1\,\mu\Omega\,\mathrm{cm}$. Above T_N pronounced Non-Fermi-Liquid (NFL) effects are observed with a linear T-dependence of the electrical resistivity. A small volume expansion of $\Delta V \approx +0.3\,\%$ is sufficient to tune the system $YbRh_2(Si_{1-x}Ge_x)_2$ to its Quantum Critical Point (QCP) at $x_c = (0.06 \pm 0.01)$. The $x = 0.05$ compound shows a $\rho(T) = \rho_0 + bT$ dependence from below 10 mK up to above 10 K. The low $\rho_0 \approx 5\,\mu\Omega$cm proves that this NFL behavior is intrinsic due to the proximity to the QCP and not related to the disorder induced by Ge-alloying. No evidence for a metamagnetic phase transition in fields up to 56 T applied along the magnetic hard direction ($\boldsymbol{B} \parallel c$) has been observed.

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1. Introduction

In the last decade a lot of work has been done on Heavy Fermion (HF) f-electron systems showing pronounced deviations from conventional Landau Fermi Liquid (LFL) behavior [1]. By chemical doping as well as the application of pressure or magnetic field these HF systems can be tuned through their AntiFerromagnetic (AF) instability. Close to this AF Quantum Critical Point (QCP) at $T_{\mathrm{N}} \to 0$, low-lying and extended spin fluctuations mediate the interaction between the quasiparticles. Instead of being constant as in the case of a LFL, the effective quasiparticle mass and the effective quasiparticle–quasiparticle scattering cross section hence become strongly energy dependent [2–5]. This is manifested in the $4f$-increment to the Sommerfeld coefficient in the electronic specific heat, $\Delta C/T \sim -\ln T$ and the electrical resistivity $\Delta\rho = \rho(T) - \rho_0$ (ρ_0: residual resistivity), for which an unusual T-dependence $\Delta\rho \sim T^{\varepsilon}$, with $1 \leq \varepsilon < 2$ is observed [2]. Because of the electron–hole analogy between $4f^1$-Ce^{3+} and the $4f^{13}$-Yb^{3+} electronic configurations, Yb-based heavy fermion compounds have attracted some attention, since they open an alternative way of studying the physics close to an AF–QCP. In contrast to the Ce case, the exchange interaction between the local $4f$ moments and the conduction electrons decreases upon increasing pressure. It is therefore possible to drive a nonmagnetic Yb system into a magnetically ordered state under pressure and to investigate the upcoming magnetism in the vicinity of the QCP. Owing to the high Yb vapor pressure up to now only a few nonmagnetic Yb-based systems were driven into the magnetically ordered state [6–8]. Since for these materials the critical pressure p_{c} necessary to induce magnetism is equal or even larger than 8 GPa, thermodynamic measurements required for a study of quantum critical behavior are inaccessible at present.

2. Experimental details

Single crystalline platelets of $YbRh_2(Si_{1-x}Ge_x)_2$ ($x = 0, 0.05$) were grown from In flux, using a molten-metal-solvent technique in closed Ta crucibles, as described in Ref. [9]. X-ray powder-diffraction patterns showed single-phase samples with the proper tetragonal structure (space group I4/mmm) and lattice parameters $a = 4.007\,\text{Å}$ and $c = 9.858\,\text{Å}$ for $x = 0$.

The low-T ac-susceptibility and -resistivity was measured by utilizing a low-frequency (17 Hz) lock-in technique adapted to ^{3}He/^{4}He dilution refrigerator. The absolute values of χ_{ac} have been determined from a comparison in the temperature range $2\,\mathrm{K} \leq T \leq 6\,\mathrm{K}$ with the results of the dc-susceptibility measured in 50 mT using a Quantum Design SQUID mag-

netometer. To perform dc-magnetization measurements down to 50 mK we used a high-resolution capacitive Faraday magnetometer developed in our institute [10].

High-field magnetization experiments were performed in the Los Alamos High Magnetic Field Laboratory using a short pulse (25 msec) 60 T magnet. Absolute values have been obtained from a comparison with the results of an Oxford Instruments magnetometer in the field range $0\,\mathrm{T} < B \leq 18\,\mathrm{T}$.

3. Magnetic properties

At high temperatures ($T \geq 200\,\mathrm{K}$) the magnetic susceptibility of $YbRh_2Si_2$ measured along both major crystallographic directions (a and c) follows a Curie–Weiss law with effective magnetic moments very close to the value of free Yb^{3+} ($\mu_{\mathrm{eff}} = 4.5\mu_{\mathrm{B}}$); but due to the strong magnetocrystalline anisotropy there is a marked difference in the respective extrapolated values for the Weiss temperatures $\Theta_P^a \approx -9\,\mathrm{K}$ and $\Theta_P^c \approx -180\,\mathrm{K}$ [11]. At $T = 2\,\mathrm{K}$ the magnetic susceptibility measured along the basal plane is about 20 times larger compared to the value measured with applied field parallel to the c-axis. Figure 1(a) is indicating that in $YbRh_2Si_2$ the Yb^{3+} moments form an "easy-plane" square lattice with a strongly anisotropic response. This anisotropy is also reflected in the isothermal magnetization. For fields applied along the magnetic hard direction ($\boldsymbol{B} \parallel c$) an almost linear magnetization curve up to 56 T is observed without any indication of a metamagnetic transition, figure 1(b).

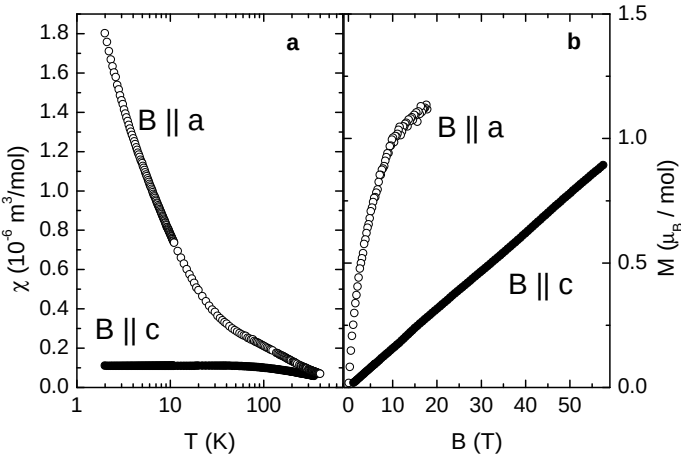

Fig. 1. Magnetic susceptibility $\chi(T) = M/B$ *vs* T in $B = 1\,\mathrm{T}$ (a) and isothermal magnetization M *vs* B at 2 K (b) for $YbRh_2Si_2$ in magnetic fields applied along the a- (∘) and c-axis (•). Pulsed-field data in (b) for $\boldsymbol{B} \parallel c$ were taken on $YbRh_2(Si_{0.95}Ge_{0.05})_2$ and agree at least for $B \leq 14\,\mathrm{T}$ with those obtained from pure $YbRh_2Si_2$.

4. Weak antiferromagnetic order

Previous low-T ac-susceptibility measurements on $YbRh_2Si_2$ have detected an AF transition at $T \approx 65\,\mathrm{mK}$ in zero field [11]. A magnetic field of only about 0.05 T applied along the easy direction ($\boldsymbol{B} \parallel a$) was found to be sufficient to suppress the ordering. This indicates the weakness of the AF-state. dc-magnetization measurements for $\boldsymbol{B} \parallel a$ have confirmed these observations. As presented in figure 2 for $T \leq T_\mathrm{N}$ a sharp decrease of the magnetization curve sets in for $B \leq B_\mathrm{c}$, with $B_\mathrm{c} \approx 50\,\mathrm{mT}$ and $\boldsymbol{B} \parallel a$.

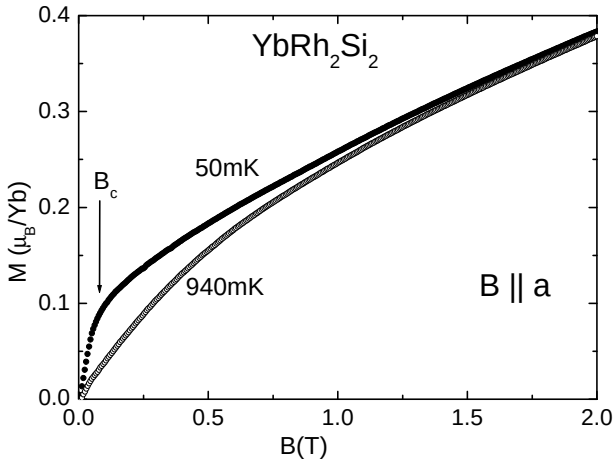

Fig. 2. DC magnetization M *vs* B for $\boldsymbol{B} \parallel a$ at two different temperatures $T < T_\mathrm{N}$ and $T > T_\mathrm{N}$. The arrow marks the critical field $B_\mathrm{c} = 0.05\,\mathrm{T}$.

The staggered magnetization per Yb-atom, as extrapolated from the data in figure 2 for $B \to 0$, is low, only $\mu_\mathrm{Yb} \approx 0.1\mu_\mathrm{B}$, indicating that the saturation moments involved in the AF ordered state are very small. The evolution of the weak AF transition has been followed in resistivity measurements too [12]. For this purpose, high-quality single crystals with residual resistivities as low as $\rho_0 \approx 1\mu\Omega\mathrm{cm}$ corresponding to a Residual Resistivity Ratio (RRR) $= \rho(300\mathrm{K})/\rho_0 \sim 68$ were studied in magnetic fields along both directions $\boldsymbol{B} \parallel a$ and $\boldsymbol{B} \parallel c$. The electrical resistivity in zero field shows a sharp kink at $T \approx 70\,\mathrm{mK}$ independent on the current direction $\boldsymbol{j} \parallel a$ and $\boldsymbol{j} \parallel c$ (figure 3(a), inset figure 4(b)). Above this characteristic temperature the resistivity follows a power-law dependence $\rho \sim T^\varepsilon$ with $\varepsilon = 1$ over more than one decade in T, whereupon the resistivity data below the transition can be best described by a $\Delta\rho \sim T^2$ behavior, in the temperature range $20\,\mathrm{mK} < T < 60\,\mathrm{mK}$. With increasing magnetic field B the resistivity anomaly shifts to lower temperatures. Figure 3(b) shows the field dependence of the Néel temperature as determined from the maximum value of

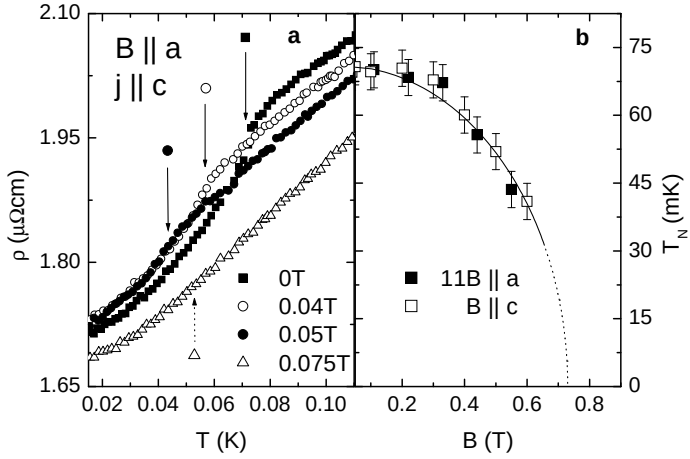

Fig. 3. (a) — Low-temperature resistivity $\rho(T)$ of $YbRh_2Si_2$ in various applied magnetic fields. The arrows mark the Néel temperature T_N deduced from $\partial\rho/\partial T$. The dashed arrow indicates the cross-over temperature $T^*(B)$, see text. (b) — Phase diagram derived from resistivity measurements for $\boldsymbol{B} \parallel c$ and $\boldsymbol{B} \parallel a$. Note, for the latter one the critical-field values have been multiplied by a factor 11.

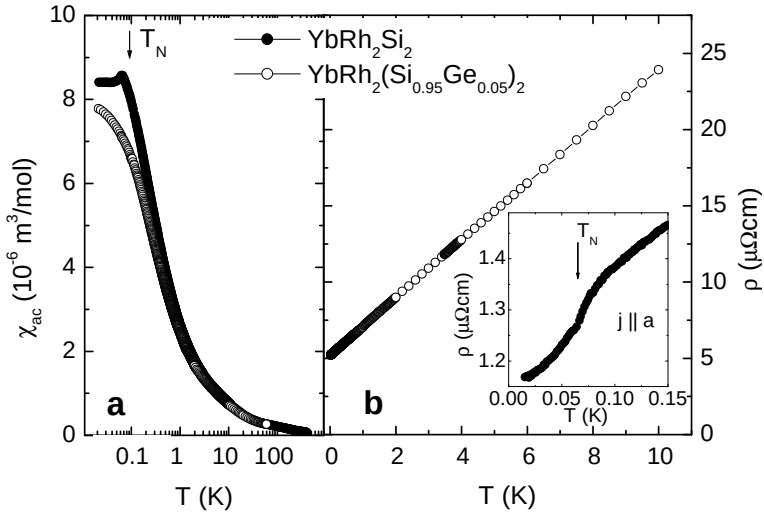

Fig. 4. (a) — ac magnetic susceptibility of $YbRh_2(Si_{1-x}Ge_x)_2$ ($x = 0$ (•), $x = 0.05$ (○)) as χ_{ac} *vs* T (on a logarithmic scale), measured along the tetragonal plane in zero dc-field. (b) — Electrical resistivity ρ of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ showing a linear behavior $\rho \sim T$ over a large temperature range $10\,\mathrm{mK} \leq T < 10\,\mathrm{K}$. Inset: low-temperature electrical resistivity of pure $YbRh_2Si_2$ in zero field, arrow marks decrease at the Néel temperature.

the derivative $\partial\rho(T)/\partial T$. For the field orientation $\boldsymbol{B} \parallel a$ the determined B–T phase diagram (figure 3(b)) agrees perfectly with earlier results obtained from ac-susceptibility and dc-magnetization studies [11, 13, 14]. The critical fields for $\boldsymbol{B} \parallel c$ are about 11 times larger than those obtained for $\boldsymbol{B} \parallel a$. This reflects the strong magnetic anisotropy of $YbRh_2Si_2$ as already discussed before. The application of magnetic fields larger than B_c, with $B_c^a \approx 0.06\,\mathrm{T}$ ($\boldsymbol{B} \parallel a$), see figure 3(a) ($\triangle$) data, and $B_c^c \approx 0.7\,\mathrm{T}$ ($\boldsymbol{B} \parallel c$) leads to a field-induced LFL state [5] below some cross-over temperature $T^*(B)$ which increases upon increasing field as evidenced by the change from $\Delta\rho \sim T$ ($T > T^*(B)$) to $\Delta\rho \sim T^2$ ($T < T^*(B)$).

5. Tuning $YbRh_2(Si_{1-x}Ge_x)_2$ through the QCP

Pronounced NFL effects in $YbRh_2Si_2$ have not only been observed in electrical-transport properties, but also in specific-heat measurements. Here a logarithmic temperature dependence of $C/T \sim \ln T$ is found over a wide temperature range [11]. Both resistivity and specific-heat measurements under hydrostatic pressure p show an increase of T_N with increasing p [16] — as expected for Yb-based alloys. This allows one to define a negative critical pressure $p_c = -(0.3 \pm 0.1)\,\mathrm{GPa}$, which corresponds to a small volume expansion ΔV of about $+0.3\%$. Such a small expansion of the unit-cell volume can be achieved by *e.g.* alloying the Si-sites of the $ThCr_2Si_2$-structure with isoelectronic Ge. The evolution of the unit-cell volume in the series $YbRh_2(Si_{1-x}Ge_x)_2$ for $x > 0$ indicates that ΔV corresponds to a critical Ge-concentration of only $x_c = (0.06 \pm 0.01)$. For such low x value, the electronic properties will be almost unaffected and nearly no significant disorder is introduced to the lattice. In a first attempt single crystals with $x = 0.05$ were grown from In flux, as reported elsewhere [9, 17]. It could be shown that the effect of Ge-alloying is opposite to the effect due to the application of hydrostatic pressure [17]. Therefore magnetic order should be suppressed completely by choosing a Ge-content of x_c. Indeed, as is shown in figure (4a), in ac-susceptibility measurements down to 10 mK a peak signaling the onset of AF order as has been observed in pure $YbRh_2Si_2$ could not be resolved. Additionally, in the zero-field electrical resistivity, performed in zero-field ($\boldsymbol{j} \parallel a$-axis) no low-temperature anomaly is detected, see figure 4(b). Instead, the linear temperature dependence $\Delta\rho(T) \sim T$, observed for pure $YbRh_2Si_2$ from above 10 K down to T_N, extends for $YbRh_2(Si_{0.95}Ge_{0.05})_2$ down to below 10 mK. A linear T-dependence of the resistivity over three decades in temperature has so far not been observed for any heavy-fermion compound. The residual resistivity is larger than in the undoped compound by only a factor of five ($\rho_0 \cong 5\mu\Omega\mathrm{cm}$), strongly suggesting that this behavior is, indeed, due to the proximity of the AF-QCP in $YbRh_2(Si_{1-x}Ge_x)_2$ at $x \cong x_c$ rather than being an effect of disorder introduced by doping.

6. Conclusion

$YbRh_2Si_2$ is a clean stoichiometric Yb-based compound which, at ambient pressure, is located at the magnetic side close to the AF-QCP. Below 10 K down to T_N, the electrical resistivity shows a linear T dependence. The phase transition is reflected by a significant increase in the slope of $\rho(T)$, while the resistivity is dominated by a T^2 term for $T < T_N$. Application of small magnetic fields suppresses the magnetic order and leads to a recovery of LFL state. From dc-magnetization measurements it is inferred that only small moments are involved in the AF order. Magnetization data up to fields of 56 T reveal no evidence for another magnetically ordered state. Applying negative pressure by slightly doping with Ge, one can tune the compound through the QCP. $YbRh_2(Si_{0.95}Ge_{0.05})_2$ is located extremely close to the QCP: a linear temperature dependence in the electrical resistivity over three decades of temperature was observed.

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