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### Angaben zur Veröffentlichung / Publication details:

Trovarelli, O., C. Geibel, S. Mederle, C. Langhammer, F. M. Grosche, Philipp Gegenwart, M. Lang, G. Sparn, and F. Steglich. 2000. "YbRh<sub>2</sub>Si<sub>2</sub>: pronounced non-Fermi-liquid effects above a low-lying magnetic phase transition." *Physical Review Letters* 85 (3): 626–29.  
<https://doi.org/10.1103/physrevlett.85.626>.



## YbRh<sub>2</sub>Si<sub>2</sub>: Pronounced Non-Fermi-Liquid Effects above a Low-Lying Magnetic Phase Transition

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(Received 3 August 1999)

We report the first observation of non-Fermi-liquid (NFL) effects in a clean Yb compound at ambient pressure and zero magnetic field. The electrical resistivity and the specific-heat coefficient of high-quality single crystals of YbRh<sub>2</sub>Si<sub>2</sub> present a linear and a logarithmic temperature dependence, respectively, in more than a decade in temperature. We ascribe this NFL behavior to the presence of (presumably) quasi-2D antiferromagnetic spin fluctuations related to a very weak magnetic phase transition at  $T_N \approx 65$  mK. Application of hydrostatic pressure induces anomalies in the electrical resistivity, indicating the stabilization of magnetic order.

PACS numbers: 71.10.Hf, 71.27.+a

An increasing number of  $f$ -electron systems, mostly Ce-based heavy fermion (HF) metals, show pronounced deviations from the properties of a conventional Landau Fermi liquid (LFL) when they are tuned through an antiferromagnetic (AF) quantum critical point (QCP) by varying a control parameter such as doping, pressure, or magnetic field [1–3]. It is generally assumed that, close to the value of the control parameter at which  $T_N \rightarrow 0$ , the abundance of low-lying and extended spin fluctuations mediating the interactions between the quasiparticles gives rise to strongly temperature-dependent quasiparticle masses and scattering cross sections [4–6]. This is manifested, for instance, in the electrical resistivity  $\Delta\rho = \rho - \rho_0$  ( $\rho_0$ , residual resistivity) and in the  $4f$ -derived increment to the specific heat  $\Delta C$ , which do not behave as expected for a LFL ( $\Delta\rho \propto T^2$ ,  $\Delta C/T = \text{const}$ ), but instead follow “non-Fermi-liquid” (NFL) temperature dependencies such as power laws ( $\Delta\rho \propto T^\varepsilon$ , with  $1 \leq \varepsilon < 2$ ) or logarithmic divergencies ( $\Delta C/T \propto -\ln T$ ) over a substantial temperature range, in a few cases as large as one decade in  $T$ . The observation of quantum-critical phenomena is, in principle, expected to be present in the paramagnetic regime not only *at* but also on both sides of the QCP provided the temperature is sufficiently close to absolute zero [4]. This has been observed in alloys such as CeCu<sub>6-x</sub>R<sub>x</sub> ( $R = \text{Au, Ag}$ ) [1,3], in which the heavy LFL at  $x = 0$  is driven to a magnetically ordered state via doping, or in AF-ordered HF compounds such as CeIn<sub>3</sub> [2] or CePd<sub>2</sub>Si<sub>2</sub> [2,7], in which magnetic order is suppressed by applying hydrostatic pressure  $p$  [8]. So far, the observation of NFL effects in *undoped* compounds at *ambient* pressure is restricted to only a few prototypical HF metals, e.g., normal-state UBe<sub>13</sub> [9], CeNi<sub>2</sub>Ge<sub>2</sub> [10,11], and CeCu<sub>2</sub>Si<sub>2</sub> [12]. So far, the existence of an AF-QCP could be established only in the case of CeCu<sub>2</sub>Si<sub>2</sub>, either via choosing slight off-stoichiometry or via applying a tiny hydrostatic pressure [12].

In comparison to Ce- and U-based materials, quantum-critical phenomena in Yb compounds have been rather less investigated. The hole-electron analogy between the

$4f^{13}\text{-Yb}^{3+}$  and the  $4f^1\text{-Ce}^{3+}$  electronic configurations offers an interesting alternative way of studying the physics close to a QCP: contrary to the Ce case, in Yb-based compounds 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 follow the evolution of magnetism in the vicinity of the QCP. Pressure-induced onset of magnetism has indeed been observed in the nonmagnetic compounds Yb<sub>2</sub>Ni<sub>2</sub>Al [13], YbCuAl [14], and YbCu<sub>2</sub>Si<sub>2</sub> [15]. Since for these materials the critical pressure  $p_c$  necessary to induce magnetism is as large as or even larger than 8 GPa, the thermodynamic measurements necessarily required for a study of quantum-critical behavior [1,3,9,11,12] are inaccessible at present. Thus no conclusive evidence of NFL behavior has yet been reported in any stoichiometric Yb-based compound close to  $p_c$ . Recent doping experiments on YbCu<sub>5-x</sub>Al<sub>x</sub> were successful in “tuning” a valence change of Yb towards 3+, accompanied by the occurrence of AF order, and NFL effects were noticed close to a critical concentration  $x_c \approx 1.5$  [16]. However, disorder may strongly influence or even produce [17] the observed NFL phenomena. It is fair to state that stoichiometric Yb-based HF compounds are lacking, which could be taken as suitable “starting systems” to explore the physics near a magnetic instability. This is mainly due to difficulties in the sample preparation owing to the high vapor pressure of Yb [18].

In this Letter we report the first observation of NFL effects at ambient pressure in high-quality single crystals of an undoped Yb-based HF compound: YbRh<sub>2</sub>Si<sub>2</sub>. Below  $T < 10$  K,  $\rho(T)$  shows an almost linear temperature dependence, whereas  $\Delta C(T)/T$  increases logarithmically upon cooling over more than a decade in temperature. These NFL effects are in fact related to the proximity of a magnetic instability: evidence for AF order at  $p = 0$  is observed at the extremely low ordering temperature of  $\approx 65$  mK. Using hydrostatic pressure as a control

parameter, well-defined anomalies are observed to develop in  $\rho(T)$ , indicating that complex long-range magnetic order is stabilized upon increasing  $p$ .  $\text{YbRh}_2\text{Si}_2$  is, therefore, identified as a model substance to study the evolution of magnetism out of a QCP in an atomically well-ordered HF system, which cannot be performed with paramagnetic Ce- (e.g.,  $\text{CeCu}_6$  [1] and  $\text{CeRu}_2\text{Si}_2$  [19]) and, perhaps, U-based HF compounds without introducing some “negative” pressure via suitable alloying.

Single crystalline platelets of  $\text{YbRh}_2\text{Si}_2$  were grown from In flux, using a molten-metal-solvent technique in closed Ta crucibles, as described elsewhere [20]. X-ray powder-diffraction patterns showed single-phase samples with the proper tetragonal structure (space group  $I4/mmm$ ) and lattice parameters  $a = 4.007 \text{ \AA}$  and  $c = 9.858 \text{ \AA}$ , in agreement with Ref. [21]. All measurements at ambient pressure were made using standard techniques on single crystals with typical residual resistivity  $\rho_0$  of the order of  $2 \mu\Omega \text{ cm}$  and a ratio  $\rho_{300 \text{ K}}/\rho_0 \sim 35$ , the latter being almost a factor of 10 larger than that of the polycrystalline sample previously investigated [22]. The measurements under pressures  $p < 3 \text{ GPa}$  were performed within a conventional piston cell, filled with an equal mixture of isopropanol and  $n$ -pentane as a pressurizing medium. The pressure was determined to  $\pm 0.05 \text{ GPa}$  from the superconducting transition of a In manometer.

The high-temperature magnetic susceptibility of  $\text{YbRh}_2\text{Si}_2$  measured along both crystallographic directions ( $a$  and  $c$ ) follows a Curie-Weiss law above  $T \approx 200 \text{ K}$  with effective magnetic moments very close to the value of free  $\text{Yb}^{3+}$  ( $\mu_{\text{eff}} = 4.5 \mu_B$ ); but due to strong magneto-crystalline anisotropy there is a marked difference in the respective extrapolated values for the Weiss temperature,  $\Theta_p^a \approx -9 \text{ K}$  and  $\Theta_p^c \approx -180 \text{ K}$  [23]. The magnetic susceptibility measured along the basal plane of the tetragonal structure at  $0.1 \text{ K}$  is almost 100 times larger than that measured along the  $c$  axis, indicating that in  $\text{YbRh}_2\text{Si}_2$  the  $\text{Yb}^{3+}$  moments form an “easy-plane” square lattice with a strongly anisotropic magnetic response [23]. The data down to very low temperature indicate a weak AF-like transition  $T_N \approx 65 \text{ mK}$  (Fig. 1). This temperature is one of the lowest reported for the onset of magnetism in any strongly correlated  $f$ -electron compound. A magnetic field of only  $45 \text{ mT}$  is sufficient to suppress  $T_N$  to below our temperature limit of  $20 \text{ mK}$ , further evidencing the weakness of this ordered state [24].

The  $p = 0$  electrical resistivity of  $\text{YbRh}_2\text{Si}_2$  is almost constant above  $100 \text{ K}$ , but decreases towards lower temperatures, indicating the development of coherence effects (inset of Fig. 1). This temperature dependence is typical of HF compounds with a strong interaction between the localized  $4f$  and the conduction electrons and resembles that of, e.g., the NFL systems  $\text{CeNi}_2\text{Ge}_2$  at  $p = 0$  [10,11] and  $\text{CePd}_2\text{Si}_2$  at the critical pressure necessary to suppress magnetism [2,7,10]. Figures 2 and 3 illustrate for  $\text{YbRh}_2\text{Si}_2$  the existence of NFL effects in  $\rho(T)$  and

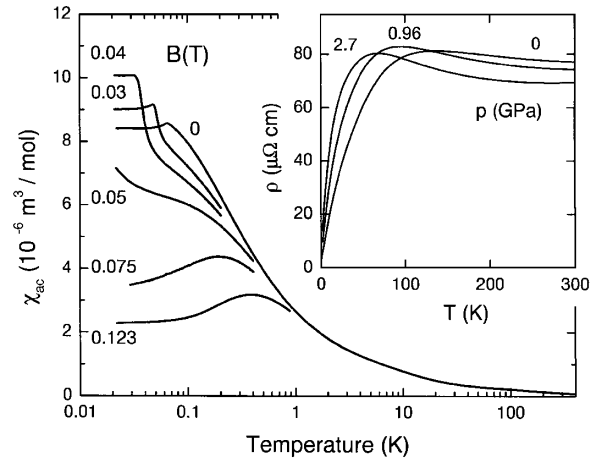


FIG. 1. ac magnetic susceptibility of  $\text{YbRh}_2\text{Si}_2$  as  $\chi_{ac}$  vs  $T$  (on a logarithmic scale), measured along the tetragonal plane at varying fields. Inset: high-temperature electrical resistivity at different pressures.

$\Delta C(T)/T$  at low temperatures. The electrical resistivity measured along the  $a$  axis follows a power-law dependence which can be expressed by  $\rho = \rho_0 + bT^\varepsilon$ , with  $b = 1.8 \mu\Omega \text{ cm/K}$  and an exponent  $\varepsilon \approx 1$ , over more than a decade in temperature (Fig. 2). Interestingly enough, *no* evidence for a phase transition is detectable within our experimental resolution down to  $20 \text{ mK}$ . This should be contrasted to “classical” HF-AF compounds, in which a  $T^2$  law is recovered below the AF-ordering temperature (e.g.,  $\text{CePd}_2\text{Si}_2$  below  $T_N \approx 8 \text{ K}$  at  $p = 2.5 \text{ GPa}$ , Ref. [7]). For a more quantitative analysis, we consider the logarithmic derivative of  $\Delta\rho(T) = \rho(T) - \rho_0$ ,  $\partial \ln \Delta\rho / \partial \ln T$ , i.e., the temperature-dependent resistivity

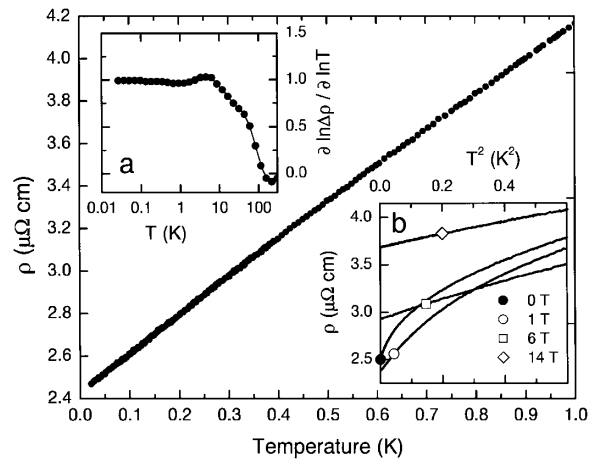


FIG. 2. Low-temperature electrical resistivity of  $\text{YbRh}_2\text{Si}_2$  at  $p = 0$  measured along the  $a$  axis as a function of temperature, obeying  $\rho(T) = \rho_0 + bT^\varepsilon$ , with  $\varepsilon \approx 1$ . (a) Temperature dependence of the effective exponent  $\varepsilon$ , defined as the logarithmic derivative of  $\Delta\rho = \rho - \rho_0$  with respect to  $T$ . (b)  $\rho(T)$ , plotted as  $\rho$  vs  $T^2$ , for  $B \leq 14 \text{ T}$  applied along the  $c$  axis. The position of the symbols indicates the crossover temperature below which a  $T^2$  law is recovered.

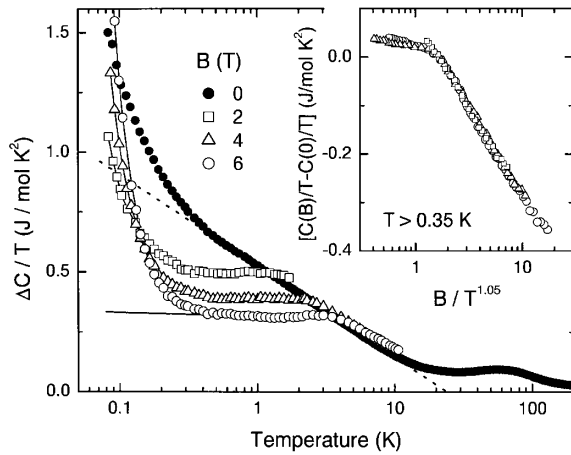


FIG. 3. Yb increment to the specific heat as  $\Delta C/T$  vs  $T$  (on a logarithmic scale) at varying fields applied along the  $a$  axis. The dotted line, representing  $\Delta C/T = \gamma'_0 \ln(T_0/T)$ , is a guide to the eye. The thick solid line represents the  $B = 6$  T data after subtracting the hyperfine contribution. Inset: scaling of the data as  $[C(B)/T - C(0)/T]$  vs  $B/T^\beta$ , with  $\beta = 1.05 \pm 0.05$ , in the temperature range  $0.35 < T < 10$  K.

exponent. As displayed in Fig. 2(a),  $\partial \ln \Delta \rho / \partial \ln T$  remains within the value  $(1 \pm 0.05)$  in a wide range of temperatures, up to almost 10 K. The application of a magnetic field leads to a gradual recovery of a  $\Delta \rho(T) = T^2$  law below a crossover temperature that increases upon increasing  $B$  [see Fig. 2(b)].

In Fig. 3 the Yb increment to the specific heat, as  $\Delta C/T$  vs  $T$ , is shown between 80 mK and 250 K. Since the entropy gain at 10 K accounts for  $0.45R \ln 2$ , while the full entropy expected for the  $J = 7/2$  multiplet of  $\text{Yb}^{3+}$  is recovered at room temperature, the broad hump at  $T \approx 60$  K indicates pronounced crystal-field splitting of the  $\text{Yb}^{3+}$  ion, implying a doublet ground state. In accordance with the linear  $T$  dependence of  $\Delta \rho(T)$ ,  $\Delta C/T$  is found to be proportional to  $-\ln T$  in a wide temperature range,  $0.3 < T < 10$  K. Here the data can be described by  $\Delta C/T = \gamma'_0 \ln(T_0/T)$ , with  $\gamma'_0 = 0.17$  J/mol K<sup>2</sup> and  $T_0 \approx 24$  K,  $T_0$  being a characteristic spin-fluctuation temperature [6]. Below 300 mK, an additional upturn appears in  $\Delta C/T$  (see Fig. 3), which might be related to the AF transition observed in the  $\chi(T)$  measurements just below the temperature limit of our calorimeter and/or to the unexplained upturns observed in the *nonmagnetic* HF compounds  $\text{UBe}_{13}$  [9], “S-type”  $\text{CeCu}_2\text{Si}_2$  [12],  $\text{CeNi}_2\text{Ge}_2$  [25], and  $\text{CeRu}_4\text{Sb}_{12}$  [26]. Application of a magnetic field parallel to the tetragonal plane leads to a gradual recovery of the properties of a LFL, in agreement with  $\Delta \rho(T)$  and similar observations made in other NFL compounds [1,3,10,11,16]. The inset of Fig. 3 shows that, in the temperature range  $0.35 < T < 10$  K, the data collapse on one universal function when plotted as  $[C(B)/T - C(0)/T]$  vs  $B/T^\beta$ , with  $\beta = 1.05 \pm 0.05$  [27]. Such scaling behavior [28] has been observed experimentally for several NFL systems in the crossover regime near a QCP [27,29]

and may be taken as further evidence of the proximity of  $\text{YbRh}_2\text{Si}_2$  to a magnetic instability.

In summary, our results at ambient pressure demonstrate that in the stoichiometric HF compound  $\text{YbRh}_2\text{Si}_2$  the low-temperature electrical resistivity and specific heat display, in an extended temperature range, pronounced deviations from the properties of a conventional LFL, namely,  $\Delta \rho \propto T$  and  $\Delta C/T \propto -\ln T$ .

Below we discuss our resistivity results obtained at finite pressures. As seen in the inset of Fig. 1, the low- $T$  drop of  $\rho(T)$  becomes steeper and shifts continuously towards lower temperatures as  $p$  increases up to  $\approx 3$  GPa, indicating a pronounced reduction of the interaction between the  $4f$  and the conduction electrons as observed for other Yb compounds [13–15,22]. With such a pressure dependence, one can expect magnetism to become stabilized at low temperature. In fact, two anomalies in  $\rho(T)$ , not visible at  $p = 0$ , develop at temperatures denoted  $T_m$  and  $T_l$ , respectively, upon increasing pressure [see Fig. 4(a)]. The phase diagram displayed in Fig. 4(b) suggests an evolution towards complex magnetic ordering for  $p > 0$ , a subject that deserves closer scrutiny in the future. An extrapolation of the data for  $p < 1.5$  GPa indicates that the ordering temperature  $T_m$  matches  $T_N$  at  $p = 0$  and vanishes at a critical pressure  $p_c$  near  $-(0.4 \pm 0.1)$  GPa. This pressure corresponds to a tiny volume change of only  $\sim 0.3\%$ , using the bulk modulus  $B_0 \sim (130 \pm 20)$  GPa of other isostructural Yb compounds [15]. We are presently trying to find out whether this phase transition in undoped  $\text{YbRh}_2\text{Si}_2$  can be suppressed just by changes in the Si-to-Rh ratio, like “phase A” disappears upon choosing a slight excess of Cu in  $\text{CeCu}_2\text{Si}_2$  [12].

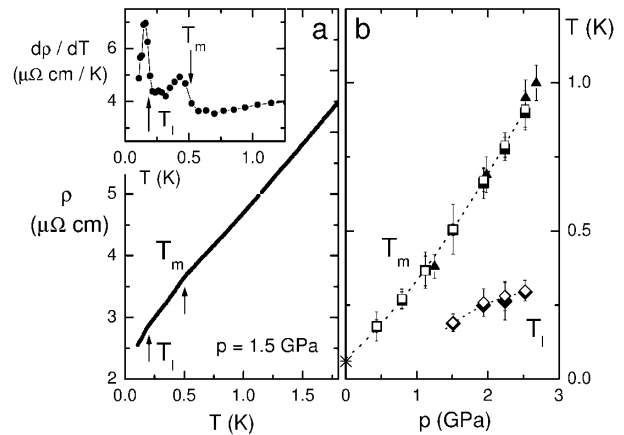


FIG. 4. (a) Low-temperature electrical resistivity of  $\text{YbRh}_2\text{Si}_2$  for  $p = 1.5$  GPa. Anomalies at  $T_m$  and at a lower temperature denoted  $T_l$  are observed to develop from a linear dependence above  $T_m$ . Inset: temperature derivative of  $\rho(T)$  showing well-defined, second-order-type transitions at  $T_m$  and  $T_l$ . (b) Pressure dependence of  $T_m$  and  $T_l$  measured in three different samples. The symbol at  $p = 0$  corresponds to the value of  $T_N$  extracted from the magnetic susceptibility. The dashed lines are guides to the eye.

Such an observation of  $\Delta\rho(T) \propto T$  and  $\Delta C(T)/T \propto -\ln T$  in a temperature range substantially larger than a decade has hitherto not been made for any clean stoichiometric HF metal at  $p = 0$  and  $B = 0$ . We note that this behavior is in apparent contradiction to the asymptotic ( $T \rightarrow 0$ ) dependencies predicted by the spin-fluctuation theory at the AF-QCP in 3D, namely,  $\Delta\rho \propto T^{3/2}$  and  $\Delta C/T = \gamma_0 - \alpha T^{1/2}$ . Here the resistivity exponent is given by  $\varepsilon = d/z$ , where  $d$  is the spatial dimension and  $z$  is the dynamical exponent of the spin fluctuations [4–6]. Although a “crossover” regime to  $\Delta\rho \propto T$  and  $\Delta C/T \propto -\ln T$  is expected by the theory in an intermediate temperature range, it should be restricted to *less* than a decade in  $T$  [6], in apparent contrast to the present case (cf. Figs. 2 and 3). A similar phenomenology has been observed in the alloy system  $\text{CeCu}_{1-x}\text{Au}_x$  close to the AF-QCP at  $x \approx 0.1$ . This was ascribed to quasi-2D spin fluctuations coupled to a 3D system of carriers [30], a behavior that is expected [4–6] if the transition at the QCP is continuous and both  $\rho_0$  and  $T$  are not *too* low [31,32]. On the other hand, in the case of 3D spin fluctuations,  $\varepsilon \rightarrow 1$  is predicted at the QCP for extremely clean samples only [32]. Since  $\rho_0$  of our  $\text{YbRh}_2\text{Si}_2$  sample is an order of magnitude larger than that of, e.g., the best  $\text{CeNi}_2\text{Ge}_2$  samples [10,11], and comparable to that of  $\text{CePd}_2\text{Si}_2$  samples close to  $p_c$  [2,7,10], for which  $\varepsilon > 1.1$ , the simplest interpretation of our  $\rho(T)$  data would indicate either the presence of spin fluctuations of AF character ( $z = 2$ ), with an effective spatial dimension close to  $d = 2$ , or an anomalous situation in which  $d \approx z > 2$ . It should be noted that the  $\text{ThCr}_2\text{Si}_2$  structure favors a geometrically frustrated spin coupling along the  $c$  direction leading to a decoupling of adjacent Yb planes and thus to a quasi-2D spin-fluctuation spectrum [2,10]. This has to be clarified by inelastic neutron-scattering experiments [33].

We conclude by stating that  $\text{YbRh}_2\text{Si}_2$  is a clean, stoichiometric Yb-based compound which, at ambient pressure, is located at the *magnetic* side very close to the QCP. The observed NFL effects in  $\text{YbRh}_2\text{Si}_2$  are ascribed to quasi-2D spin fluctuations related to a weak, low-lying AF transition. Contrary to the behavior of Ce compounds, however, application of pressure allows one to study the evolution of magnetism out of the QCP. Theoretical as well as experimental studies are needed in order to shed light on the microscopic mechanisms leading to the observed quantum-critical phenomena in  $\text{YbRh}_2\text{Si}_2$ .

We are grateful to Greg Stewart, Siegfried Horn, and Mohsen Abd-Elmeguid for valuable conversations.

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