Yb-based heavy-fermion metal situated close to a quantum critical point

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We report low-temperature thermodynamic, magnetic, and transport measurements on high-quality single crystals of the tetragonal heavy-fermion system $YbIr_2Si_2$. Depending on synthesis conditions it crystallizes either in the *I*-type body-centered $ThCr_2Si_2$ or *P*-type $CaBe_2Ge_2$ structure. The *P*-type system has a low Kondo scale $T_K=2$ K and orders magnetically below 0.7 K whereas the *I*-type system remains paramagnetic and shows, above 0.5 K, a temperature dependence of the resistivity and the specific heat similar to that of $YbRh_2Si_2$. In contrast to $YbRh_2Si_2$ which exhibits non-Fermi-liquid behavior, $YbIr_2Si_2$ reaches a Landau Fermi-liquid state below 0.2 K and is thus ideally suited for pressure studies across the quantum critical point.

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The zero-temperature quantum phase transition (QPT) from a magnetically ordered to a nonmagnetic ground state upon varying a control parameter other than temperature is presently one of the hot topics within condensed matter physics. In several cases, the disappearance of the magnetic order is connected with very interesting phenomena, such as, e.g., formation of heavy fermions (HFs), onset of unconventional superconductivity, and appearance of non-Fermi-liquid (NFL) behavior.[1–6] HF systems, intermetallic compounds mostly based on Ce, Yb, or U, are ideal materials for the study of these phenomena, because these elements present an unstable *f* shell which can be easily tuned from a magnetic to a nonmagnetic configuration by alloying or applying pressure. Increasing the hybridization between the localized *f* and the conduction electrons leads to a weakening of the magnetic order, which eventually disappears. In several systems, it is suggested that the magnetic ordering temperature decreases continuously down to $T=0$, leading to a quantum critical point (QCP). These QPTs attract presently wide interest because of the observation of unusual transport and thermodynamic properties and because, in some cases, one observes the onset of unconventional superconductivity. A typical example is $CePd_2Si_2$, an antiferromagnetically ordered HF system in which the Néel temperature is continuously suppressed from 10 K at ambient pressure to $T=0$ at a critical pressure of 2.8 GPa. Close to this QCP the system shows HF superconductivity in very clean samples only.[3] Neither the nature of the superconducting state, nor the nature of the fluctuations responsible for the unusual properties at the QCP are presently settled.

While a huge number of Ce- and U-based HF systems have been discovered and thoroughly investigated up to now, much less Yb-based HF systems are presently known. Only a very few of them are located close to the magnetic QPT on the magnetically ordered side, and there seems to be no example for an Yb system close to the QPT located on the nonordered side. Furthermore, we do not yet know of any Yb-based HF superconductor. These facts motivated us to look for Yb-based systems situated close to the borderline of long-range magnetic order. In recent years $YbRh_2Si_2$ has attracted considerable attention due to its proximity to a QCP.[6–11] This is evidenced by the onset of very weak magnetic order below $T_N=70$ mK, which is expected to disappear under a negative pressure of only 0.3 GPa (since the magnetic trivalent Yb state is smaller than the nonmagnetic divalent one, pressure increases the magnetic character of Yb compounds). A small magnetic field $B_c=0.06$ T applied in the easy magnetic plane, or slightly enlarging the unit cell using Ge- or La doping eventually suppresses the magnetic order leading to a QCP where the characteristic energy of the heavy quasiparticles seems to vanish. While at $B=B_c$ pronounced NFL behavior is observed in the resistivity and in the specific heat, for $B>B_c$ a field induced Landau Fermi liquid is formed below a cross over temperature which increases with increasing field.[6–8] The QCP in $YbRh_2Si_2$ is believed to be of unconventional nature[12,13] as in the case of $CeCu_{6-x}Au_x$.[14] $YbRh_2Si_2$ is the first dense Kondo system where one could surprisingly observe a sharp ESR line of the Kondo ion well below the Kondo temperature ($T_K=25$ K).[10] Most likely, these unique properties of $YbRh_2Si_2$ are connected with very specific interactions in this compound, such as, e.g., pronounced ferromagnetic fluctuations competing with antiferromagnetic ones, but they might be also a consequence of a more general difference between Yb- and Ce-based compounds.

A drawback of $YbRh_2Si_2$ is that the QCP can only be reached by applying an external magnetic field or some chemical substitution, which imply destroying the time reversal symmetry or introducing disorder. Both are expected to affect the behavior in the quantum critical regime, in particular, the appearance of possible unconventional superconductivity. The conceptually better way to cross the QCP with pressure requires, in the case of Yb compounds, the system to be on the nonmagnetic side of the QCP at ambient pressure. Ir is located just below Rh in the periodic table, has thus the same chemical valence but is slightly larger. To the best of our knowledge, no investigation of $YbIr_2Si_2$ has yet been published. We have now been able to synthesize this compound and investigate its low-*T* properties. Our results evidence a paramagnetic Fermi-liquid ground state and suggest this compound to be located close to the QCP.

Single crystals were grown from In flux in a closed Ta crucible. We found that, depending on preparation condi-

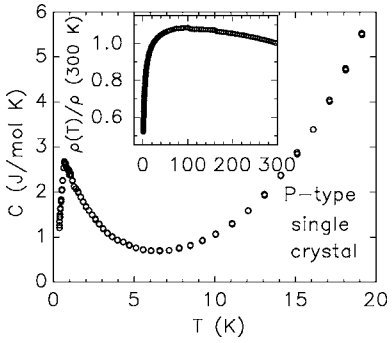

FIG. 1. Specific heat and (inset) resistivity of *P*-type $YbIr_2Si_2$. The specific heat shows a clear anomaly at 0.7 K which we attribute to a phase transition, presumably of antiferromagnetic type. The temperature dependence of the resistivity evidence a pronounced Kondo lattice behavior.

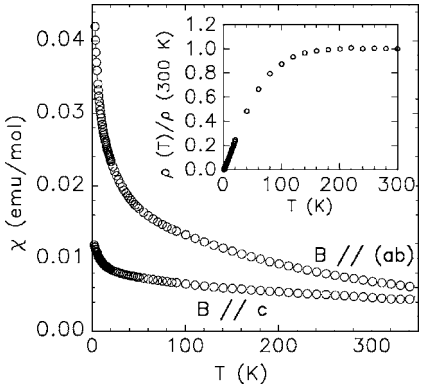

FIG. 2. Magnetic susceptibility of *I*-type $YbIr_2Si_2$ with $B//a$ and $B//c$, showing the pronounced easy plane anisotropy. Inset: temperature dependence of the resistivity with $I//ab$ up to 300 K. The resistivity behavior is typical of heavy-fermion compounds.

tions, $YbIr_2Si_2$ crystallizes in two different structures, either in the primitive tetragonal $CaBe_2Ge_2(P)$ type or in the body-centered tetragonal $ThCr_2Si_2$-(I) type, both being ordered variants of the $BaAl_4$ type and differing by the interchange of one Ir and one Si layer. This interchange leads to the loss of a mirror plane at the Yb site in the *P* type, resulting in a profound change of the local symmetry at the Yb site, although the lattice parameters are almost identical with $a=4.032$ Å, $c=9.826$ Å for the *I* type and $a=4.036$ Å, $c=9.900$ Å for the *P* type. A detailed structure determination with single crystal XR diffraction is underway.[16] The occurrence of these two different structures in RIr_2Si_2 is well documented[15] for other rare-earth elements *R*, but there was no report on $YbIr_2Si_2$. We are able to grow both types of crystals by slightly changing the Yb content in the starting composition. A slight excess (10%) of Yb favors the primitive structure, while stoichiometric starting composition produces the body-centered structure. While the *I*-type single crystals seem to have a rather perfect crystallinity as deduced from the very high residual resistivity ratio RRR >200, the much lower RRR ≈ 2 of the *P* single crystal suggests some disorder there, likely some Ir-Si interchange.

First we briefly discuss the properties of a *P*-type sample. The magnetic susceptibility (not shown) shows the behavior typical for a system with well localized *f* moments, following a Curie Weiss law above 30 K with an effective moment $\mu_{\text{eff}}=4.4\mu_B$, close to that expected for free Yb^{3+} ions $(4.54\mu_B)$. The temperature dependence of the resistivity (inset of Fig. 1) is typical of Kondo lattice system: at high temperature $\rho(T)$ increases with decreasing temperature, passes through a broad maximum at around 70 K followed by a rapid decrease below 50 K. The specific heat (main part of Fig. 1) shows a pronounced anomaly at 0.7 K, which we attribute to magnetic ordering. The magnetic entropy reaches $0.5R\ln 2$ at 1 K and is close to $R\ln 2$ at 5 K, indicating a rather small Kondo temperature of a low-lying crystal-field derived Kramers doublet ($S_{\text{eff}}=1/2$), with T_K being of the order of 2 K. This compound is, therefore, assumed to have a more stable magnetically ordered state than $YbRh_2Si_2$ and should be located further away from the QCP on the magnetic side.

In the following we focus on *I*-type $YbIr_2Si_2$ that is located on the nonmagnetic side of the QCP and thus represents an ideal complement of $YbRh_2Si_2$. A substantially larger Kondo temperature than for the *P*-type sample, is evidenced, e.g., by the behavior of both the susceptibility and of the resistivity at elevated temperatures (Fig. 2). The magnetic susceptibility is found to be highly anisotropic, $\chi(T)$ along the *a* axis being much higher than $\chi(T)$ along the *c* axis. A similar, but even more pronounced easy plane anisotropy has been reported for $YbRh_2Si_2$. A Curie-Weiss law can be fitted to the $\chi(T)$ curves only at high temperatures, above 100 K [for $B//(ab)$]. The effective moment obtained from the fits $(4.5\mu_B)$ points to an Yb valence close to 3+. The Weiss temperatures $\Theta_P{}^a=-82$ K and $\Theta_P{}^c>-400$ K are very different, reflecting the large magnetocrystalline anisotropy. The large Θ_P values derived from the high temperature susceptibility provide a first indication for a large Kondo energy scale.

Further support for this stems from the temperature dependence of the resistivity, which shows the typical behavior for a Kondo-lattice system. Below 300 K, $\rho(T)$ increases slightly with decreasing temperature, then goes through a broad maximum at around 150 K, before decreasing strongly below 100 K (inset of Fig. 2). The residual resistivity is as low as 0.3 $\mu\Omega$ cm and corresponds to RRR=225. The overall resistivity behavior is very similar to that of the $YbRh_2Si_2$. However, while the Rh-based compound exhibits a linear temperature dependence down to $T_N=70$ mK, *I*-type $YbIr_2Si_2$ shows a clear positive curvature in $\rho(T)$ below 10 K. A fit with a power law $\rho(T)=\rho_0+AT^n$ reveals an increase of *n* when the fitting range is shifted to lower temperatures (upper inset of Fig. 3). Below 200 mK, $\rho(T)$ perfectly follows $\rho(T)=\rho_0+AT^2$ with $\rho_0=0.3$ $\mu\Omega$ cm and $A=0.7$ $\mu\Omega$ cm/K^2 (lower inset of Fig. 3). Despite the very low residual resistivity, which confirms the high quality of the single crystals, we could not resolve any signature of magnetic order or superconductivity down to 20 mK.

A magnetically nonordered Fermi-liquid (FL) ground state is also supported by the specific-heat and susceptibility results at low temperatures. In Fig. 4 the 4*f* part of the specific heat $\Delta C(T)$ obtained after subtracting $C(T)$ of $LuIr_2Si_2$ is plotted as $\Delta C/T$ versus ln T. A nuclear contribution $C_n=DT^{-2}$ with $D=80\times 10^{-6}$J K mol^{-1} was subtracted below 200 mK. Below 10 K, $\Delta C/T$ increases with decreasing tem-

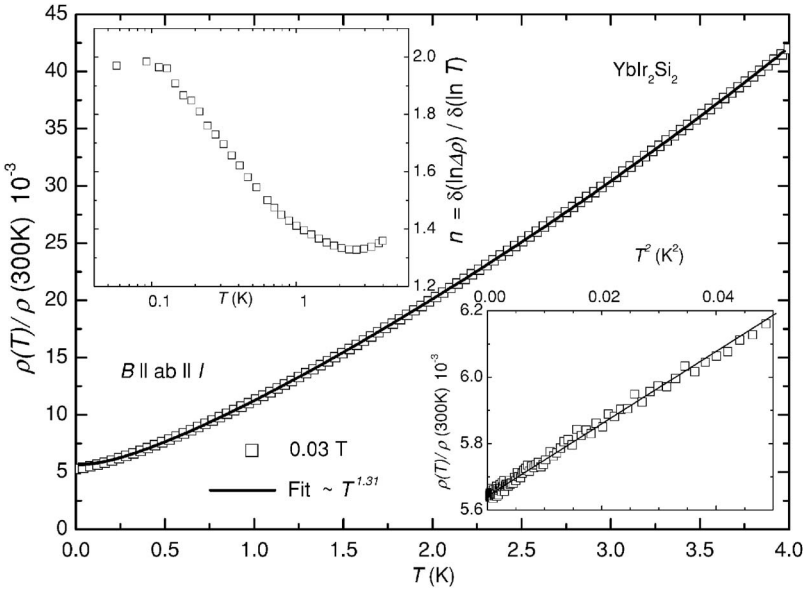

FIG. 3. Electrical resistivity of I-$YbIr_2Si_2$ with $I//ab$ at low temperatures in a field of 0.03 T in order to suppress the effect of superconductivity in In inclusions. The inset shows the temperature dependence of the exponent n of a fit $\rho(T)=\rho_0+AT^n$. n increases towards low temperatures and levels out at the value $n=2$, indicating the formation of a Fermi-liquid ground state. Lower inset: Plot of $\rho(T)$ versus T^2 showing the Fermi-liquid behavior at low temperature

perature and in the temperature range 0.5 to 2 K it follows a relation $\Delta C/T=\gamma \ln(T_0/T)$ with $\gamma=0.082$ J/mol K^2 and the characteristic temperature $T_0=44$ K. This temperature dependence is similar to that observed in $YbRh_2Si_2$($T_0=24$ K). However, in contrast to $YbRh_2Si_2$ where the increase in $\Delta C/T$ gets more pronounced below 300 mK, in $YbIr_2Si_2$ we observe below 400 mK a saturation of $\Delta C/T$ at a Sommerfeld coefficient $\gamma_0=0.37$ J/mol K^2. The crossover from the logarithmic increase to the FL behavior around 400 mK is quite abrupt. Similar behavior has been observed in other systems close to a QCP, e.g., in $YbRh_2Si_2$ in a magnetic field large enough to suppress the AF state. The magnetic entropy reaches $0.5R$ ln 2 at 20 K, giving a first estimation for the Kondo temperature $T_K \simeq 40$ K. This is almost twice the value deduced for $YbRh_2Si_2$. The application of magnetic fields extends the FL regime to higher temperatures.

The inset of Fig. 4 shows the temperature dependence of the magnetization (M) taken at $B=1$ T and at low temperatures, down to 50 mK. $\chi(T)=M/B$ continues to increase down to 400 mK, but the increase amounts only to 15%, i.e., much less than in $YbRh_2Si_2$. Below 400 mK, the susceptibility shows a tendency to saturation, a further signature of Landau Fermi-liquid behavior. As in the specific heat, the crossover from an increasing $\chi(T)$ to the almost constant value $\chi_0=5.25\times10^{-2}$ emu/mol is well defined. This crossover, which is more sudden than in typical Ce-based HF system such as $CeRu_2Si_2$, might be an intrinsic property of Yb systems connected with the smaller extend of the f-wave function in Yb and thus a more localized character. However, we cannot exclude the broad kink in $\chi(T)$ and $C(T)$ to be due to a very small amount of P-type phase with a broadened magnetic transition around 400 mK. We can exclude these broad kinks to originate from an intrinsic magnetic transition in I-type $YbIr_2Si_2$, because with such a high RRR one expects to observe a sharp transition, sharper than that observed in $YbRh_2Si_2$ with an RRR of 30–60. Further on, one would then also expect to see an anomaly in $\rho(T)$. The magnetization as a function of field (not shown) shows a slight negative curvature similar to that in many nonmagnetic HF compounds. Differences in the magnetization between 50 mK and 2 K are rather small and can only be resolved at low fields, confirming an intrinsic T-independent susceptibility expected for a Landau Fermi-liquid ground state.

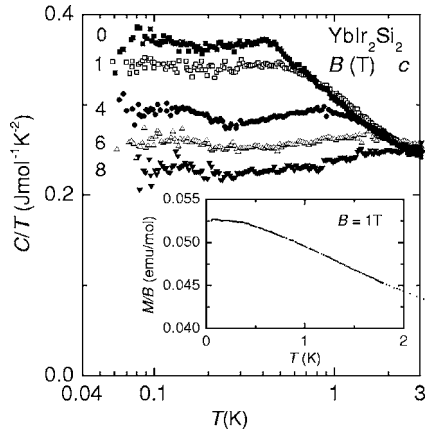

FIG. 4. Specific heat of $YbIr_2Si_2$ plotted as delta C/T vs ln$T$. C/T increases logarithmically with decreasing temperature but levels out below 400 mK as expected for a Fermi liquid. Inset: Magnetic susceptibility at low temperatures. Note the tendency to saturation below 400 mK.

For a Landau Fermi liquid, one expects the Sommerfeld coefficient to be related to the coefficient A of the T^2 term in the resistivity and with the value of the constant (Pauli-) susceptibility at low temperatures. For I-type $YbIr_2Si_2$, we get a Kadowaki-Woods ratio $A/\gamma^2=5.1$ $\mu\Omega$ cm K^2 mol^2 J^{-2} a factor of 2 smaller than that found in many Ce-based HF systems. In many of the Yb-based intermediate valent systems such as $YbAl_3$, the Kadowaki-Woods ratio is even smaller by an order of magnitude, which has been explained to be due to the higher degeneracy of the f state, since in an intermediate valent system the full Hund's rule multiplet is involved instead of only the crystal-field ground state doublet.[17] Then, the observed value of A/γ^2 in $YbIr_2Si_2$ is in full accordance with its Kondo energy scale $T_K \simeq 40$ K being

higher than that of typical Ce-based heavy-fermion systems, but lower than that of intermediate valent Yb compounds. Interestingly, the Sommerfeld-Wilson ratio is highly enhanced similar to $YbRh_2Si_2$.[18]

To summarize, we present in this paper an investigation of $YbIr_2Si_2$. We found that this compound can crystallize in two different, but closely related tetragonal structures, the primitive $(P)CaBe_2Ge_2$ type or the body centered $(I)ThCr_2Si_2$ type. Both compounds exhibit Kondo-lattice behavior. In P-$YbIr_2Si_2$, the Kondo temperature is rather low, $T_K \simeq 2$ K, resulting in a magnetically ordered state below 0.7 K. By contrast, the Kondo temperature in I-$YbIr_2Si_2$ is much larger, $T_K \simeq 40$ K, a factor of 2 larger than in the structural homolog $YbRh_2Si_2$ and no phase transition is observed above 20 mK in this variant. Instead, one finds Landau Fermi-liquid behavior below 0.2 K. Thus, I-$YbRh_2Si_2$ is found to be a new Yb-based HF compound with a magnetically nonordered Landau Fermi-liquid ground state. Its Sommerfeld coefficient $\gamma_0 = 370$ mJ/K^2 mol is one of the highest value among magnetically nonordered Yb systems. Our results indicate I-$YbIr_2Si_2$ to be placed on the nonmagnetic side of the QCP, in contrast to its Rh homologue which is just on the magnetic side. Although over a much narrower temperature range, the logarithmic increase of C/T with decreasing temperature and non quadratic temperature dependence of resistivity are similar to those observed in $YbRh_2Si_2$ which suggests the Ir compound to be rather close to the QCP. Since the volume of $YbIr_2Si_2$ is only 1.9% larger than that of $YbRh_2Si_2$, a moderate pressure of 2 to 3 GPa is expected to be sufficient to cross the QCP from the nonmagnetic side, providing a unique opportunity to investigate the properties at and around the QCP in a very clean stoichiometric Yb system. There is presently a lot of speculation on whether and how the unconventional superconducting state observed in HF systems and high-temperature superconductors might be caused by the disappearance of magnetic order due to the delocalization of f or d electrons. In HF systems, the onset of unconventional superconductivity is suspected to occur only if the samples are very clean. Being one of the cleanest Yb-based HF systems and being located on the nonmagnetic side of the QCP, $YbIr_2Si_2$ appears highly suited to searching for pressure-induced superconductivity.

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