Field-Induced Suppression of the Heavy-Fermion State in $YbRh_2Si_2$

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We report dc-magnetization measurements on $YbRh_2Si_2$ at temperatures down to 0.04 K, magnetic fields $B \leq 11.5$ T, and under hydrostatic pressure $P \leq 1.3$ GPa. At ambient pressure a kink at $B^\star = 9.9$ T indicates a new type of field-induced transition from an itinerant to a localized $4f$ state. This transition is different from the metamagnetic transition observed in other heavy-fermion compounds, as here ferromagnetic rather than antiferromagnetic correlations dominate below $B^\star$. Hydrostatic pressure experiments reveal a clear correspondence of $B^\star$ to the characteristic spin fluctuation temperature determined from specific heat.

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The f electrons in certain lanthanide and actinide compounds can exhibit a dual character, i.e., localized as well as itinerant, and the competition between both leads to the exciting heavy fermion (HF) state. Here, the f electrons behave as local moments at temperatures above the Kondo temperature T_K, following a Curie-Weiss law in the magnetic susceptibility, while the weak hybridization between the f and conduction electrons leads at lower temperatures to the formation of heavy quasiparticles. An extremely large mass enhancement up to $\sim$1000 in certain Ce-, Yb-, and U-based compounds has been observed. The application of magnetic field destroys the HF state and can produce a sharp metamagneticlike transition from the itinerant to the localized f-electron state which is in contrast to the smooth crossover in temperature variation at zero field. Such metamagneticlike behavior has been observed in Ce- and U-based HF compounds such as $CeRu_2Si_2$, $CeCu_6$, and UPt_3 [1–3]. The metamagnetic transition in $CeRu_2Si_2$ has been studied most extensively because of the dramatic step observed in magnetization and the relatively small critical field of $B_M = 7.7$ T. Fermi surface properties studied by de Haas–van Alphen effect (dHvA) are well explained by the picture of itinerant and localized $4f$ electrons below and above B_M, respectively [4,5]. Reflecting the localization of $4f$ electrons, the Sommerfeld coefficient γ in $CeRu_2Si_2$ is strongly suppressed above the critical field B_M [6]. It is also noteworthy that the transition is accompanied with a sharp step in magnetostriction $\Delta L(B)/L$ [7]. The step produces a sudden change in hybridization between $4f$ and conduction electrons. In the case of Yb-based compounds, $YbCu_{5-x}Ag_x$ shows a metamagneticlike smooth crossover from valence-fluctuating state to a stable Yb^{3+} state with localized magnetic moments [8].

In this Letter we report a new type of field-induced suppression of the HF state in the Yb-based compound $YbRh_2Si_2$. This system with a Kondo temperature of about 25 K is located very close to a quantum critical point (QCP) related to a very weak antiferromagnetic (AF) order below $T_N = 70$ mK [9]. A tiny critical magnetic field of $B_c = 0.06$ T, applied in the easy magnetic plane perpendicular to the tetragonal c axis, is sufficient to suppress the AF order [10]. For $B > B_c$, Landau Fermi liquid (LFL) behavior is deduced from the electrical resistivity, described by $\rho(T) = \rho_0 + AT^2$, with the coefficient $A(B)$ diverging towards B_c indicating a field-induced QCP [10,11]. Correspondingly, for $B > B_c$ the low-temperature specific heat divided by temperature saturates, and the Sommerfeld coefficient $\gamma(B)$ decreases rapidly with increasing field, indicating a strongly field-dependent quasiparticle mass in the LFL state. It has been discovered by ^{29}Si-nuclear magnetic resonance experiments that the critical fluctuations in the field-induced LFL state have a strong ferromagnetic (FM) component, dominating for fields above 0.25 T [12]. These FM fluctuations lead to a strongly enhanced magnetic susceptibility as evidenced by a Sommerfeld-Wilson ratio of $R \simeq 14$ [10].

The Grüneisen parameter which describes the volume dependence of the Kondo temperature, $\Gamma = -\partial \ln T_K/\partial \ln V$, is generically large in HF systems, reaching values up to a few hundreds [13]. Assuming a correlation between the characteristic field $B^\star$, necessary to suppress the Kondo state, and T_K, one would expect a strong volume dependence of $B^\star$ as well. Contrary to the Ce case, in Yb-based systems the exchange interaction between localized $4f$ electrons and the conduction electrons decreases upon applying pressure, leading to a decrease of T_K. Correspondingly, a decrease of the field $B^\star$ with pressure is expected. In order to search for an itinerant-localized $4f$-electron transition in $YbRh_2Si_2$ under magnetic field, we have performed dc-magnetization measurements at temperatures down to 40 mK and fields up to 11.5 T both at ambient pressure, as well as—for the first time in any system at mK temperatures—under hydrostatic pressure up to 1.3 GPa.

High-quality single crystals ($\rho_0 = 1\ \mu\Omega$ cm) were grown from In flux as described earlier [9]. The dc magnetization was measured utilizing a high-resolution capacitive Faraday magnetometer [14]. In order to determine the magnetization under hydrostatic pressure, a miniaturized CuBe piston-cylinder pressure cell of 6 mm outer diameter and 3.2 g total weight has been designed. The piston is made from NiCrAl, a hard material with a relatively small magnetization. The magnetization of the pressure cell, including the 6.0 mg $YbRh_2Si_2$ single crystal mounted on the magnetometer, can be detected with a resolution as high as 10^{-5} emu. The contribution of the sample to the total magnetization of the sample and pressure cell is larger than 63% in the entire field and temperature range. The pressure is determined by the difference between the superconducting transitions of two small Sn samples: one placed inside the pressure-transmitting medium (daphne oil) together with the $YbRh_2Si_2$ sample, the other one outside the pressure cell. The T_c values are determined using a commercial SQUID magnetometer. In order to investigate the field dependence of the quasiparticle mass, specific-heat measurements have been performed with the aid of a quasiadiabatic heat-pulse technique.

Figure 1 shows the low-temperature magnetization $M(B)$ at several different pressures. At ambient pressure the magnetization curve reveals two kinks. The first one at very low fields (see inset) results from the suppression of AF order at $B_c = 0.06$ T. Note that the polarization at B_c amounts to $\sim 0.1\mu_B$ only. The remaining part of the moment is fluctuating and contributes to the strongly enhanced Pauli-paramagnetic susceptibility [10]. The second kink at $B^\star = 9.9$ T we ascribe, as discussed, to the itinerant-localized transition of the $4f$ electrons. For magnetic fields $B > B^\star$, the magnetization tends to saturate at a value of the order of $1.2\mu_B/Yb$ expected for a polarized doublet ground state. This upper kink broadens rapidly with increasing temperature and disappears above 2 K without shifting its position in field. Upon applying hydrostatic pressure, the kink shifts to $B^\star = 6.2$ and 3.7 T at 0.64 and 1.28 GPa, respectively. Thus this anomaly is very sensitive to pressure.

The pressure dependence of the AF phase transition has been studied by electrical resistivity measurements[15]. Mederle *et al.* found an increase of T_N with pressure and the indication of a second phase transition, labeled T_L inside the antiferromagnetically ordered state for pressures above ~1 GPa. As shown in the inset of Fig. 1, the critical field B_c for the AF order increases to 0.14 and 0.29 T at pressures of 0.64 and 1.28 GPa, respectively. The kink at B_c sharpens substantially under pressure. The additional anomaly observed at 0.08 T for a pressure of $p =$ 1.28 GPa indicated by the empty bracket corresponds to the suppression of the state below T_L observed by Mederle *et al.* inside the antiferromagntically ordered state.

As shown in Fig. 2, both the differential susceptibility $\chi(B) = dM(B)/dB$ and the Sommerfeld coefficient $\gamma(B)$ show a broadened step at $B^\star$ that sharpens under hydrostatic pressure. The fact that $\chi(B) \propto \gamma(B)$ for $B \leq B^\star$ proves a low-field LFL state of itinerant $4f$ electrons with strongly field-dependent quasiparticle mass. Using an effective moment of $1.4\mu_B/Yb$ [10], the resulting Sommerfeld-Wilson ratio R_W equals 18 ± 1 below $B^\star$. The steplike decrease at $B^\star$, indicates a large and sudden reduction of the quasiparticle mass. A similar feature has previously been observed for the A coefficient determined from the T^2 contribution to the electrical resistivity [16].

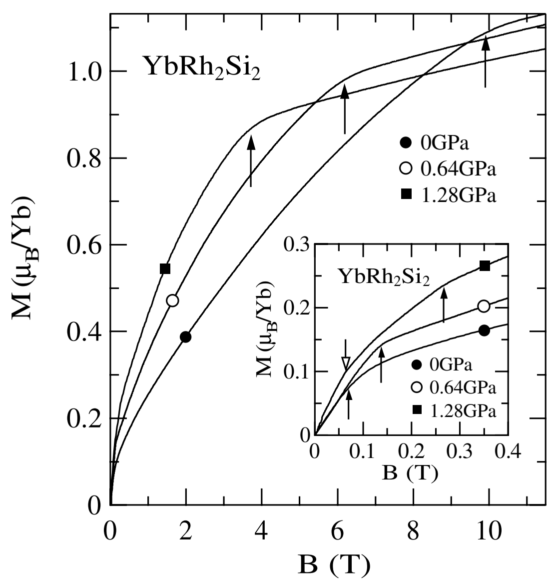

FIG. 1. Field dependence of the magnetization ($B \perp c$ and in units of effective moment per Yb) at differing pressures of 0, 0.64, and 1.28 GPa measured at 40, 40, and 60 mK, respectively. Arrows indicate respective values of critical field $B^\star$ above which the $4f$ states are localized. Inset enlarges low-field regime. Filled and open arrows indicate critical fields for the AF order and second transition induced at high pressure; see text.

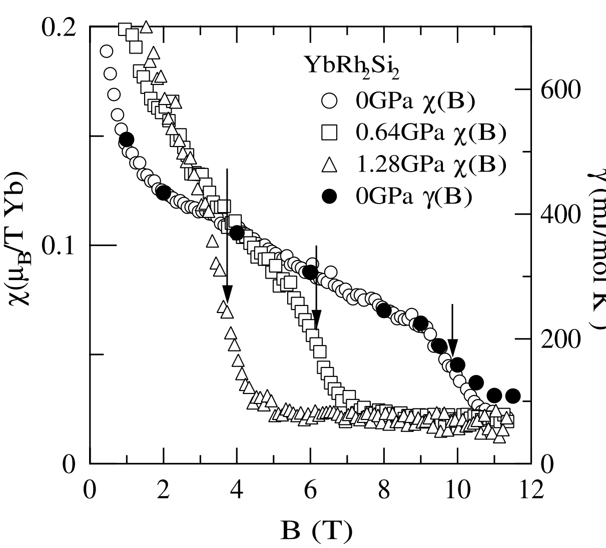

FIG. 2. Field dependence of the differential susceptibility $\chi = dM/dB$ at 0, 0.64, and 1.28 GPa (left axis) as well as Sommerfeld coefficient γ at ambient pressure (right axis). Arrows indicate critical field B^* above which the $4f$ states are localized.

Additionally, the slope of the linear magnetostriction changes at this field from negative to positive, suggesting the formation of completely localized $4f$ moments in the high-field state [16]. Further on, at $P = 0$ and above $B^\star$, $\gamma(B)$ slightly deviates from the relation $\gamma \propto \chi$. This implies that R_W decreases significantly at $B^\star$. Note that γ above $B^\star$ has a residual value $\sim$100 $\mathrm{mJ/mol \cdot K^2}$ which is still large for a local moment system. This might indicate residual Kondo-type interactions persisting even above $B^\star$. In $CeRu_2Si_2$, too, relatively large γ values beyond the metamagnetic transition have been found at magnetic fields far above B_M ($\sim$ 80 $\mathrm{mJ/mol \cdot K^2}$ at 20 T) [6], although the Fermi surface properties are in good agreement with the picture of localized $4f$ electrons. However, there is a distinct difference between our results on $YbRh_2Si_2$ and those observed at metamagnetic transitions in, e.g., $CeRu_2Si_2$ and UPt_3: for the latter systems the Sommerfeld coefficient $\gamma(B)$ shows a peak at B_M [6,17,18]. The absence of a peak in $\gamma(B)$ for $YbRh_2Si_2$ is related to the absence of a peak in the susceptibility, and the origin of this difference is discussed below.

Next we compare the pressure dependence of $B^\star$ with that of the characteristic spin fluctuation temperature T_0, estimated by fitting the zero-field low-temperature specific heat with $C(T)/T = -D\ln(T/T_0)$. Since at ambient pressure T_0 matches with the single-ion Kondo temperature T_K determined from the magnetic entropy [9], the pressure dependence of T_0 is assumed here to represent that of the Kondo temperature T_K. In order to obtain the pressure dependence of T_0, we used specific-heat data under hydrostatic pressure reported in [15]. As shown in Fig. 3, a correlation between T_0 and $B^\star$ is very probable. The exponential decrease with increasing pressure is compatible with the Kondo temperature $T_\mathrm{K} \propto \exp[-1/J_{cf}D_c(\varepsilon_F)]$ being determined by the product of the $4f$-conduction electron exchange intergral, J_{cf}, and conduction electron density of states at the Fermi energy, $D_c(\varepsilon_F)$.

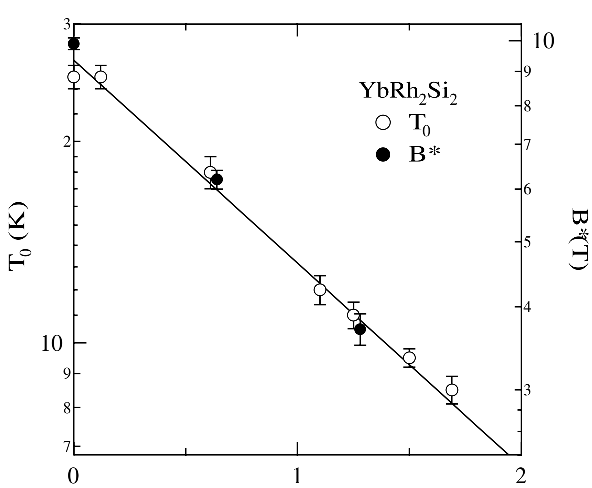

FIG. 3. Pressure dependence of the characteristic spin fluctuation temperature T_0 [15] (left axis) and the field $B^\star$ (right axis) for $YbRh_2Si_2$. Solid line represents $\exp(-0.7\ \mathrm{GPa}^{-1} \times P)$ dependence.

Using the isothermal compressibility $\kappa_T = 5.3 \times 10^{-12}\ \mathrm{Pa}^{-1}$ [19] we obtain the ''thermal'' Grüneisen parameter $\Gamma_T = 1/\kappa_T \times \partial \ln T_\mathrm{K}/\partial P = -132 \pm 6$. The ''magnetic'' Grüneisen parameter, derived from the pressure dependence of the characteristic field $B^\star$, $\Gamma_B = 1/\kappa_T \times \partial \ln B^\star/\partial P$, equals Γ_T because of the same slope for T_0 and $B^\star$ in their pressure dependences. This resembles the case of $CeRu_2Si_2$ for which Γ_B, determined from the pressure dependence of the metamagnetic transition equals Γ_T as well. One important difference of $YbRh_2Si_2$ compared to $CeRu_2Si_2$ is, however, the strongly enhanced Sommerfeld-Wilson ratio in the former system. A systematic comparison of thermal and magnetic Grüneisen parameters for various systems has been made by Kaiser and Fulde [20]. They found that in contrast to usual metals and HF systems, for strongly enhanced Pauli paramagnets with a Sommerfeld-Wilson ratio $R_W \gg 1$, the magnetic Grüneisen parameter is much larger than the thermal one. The enhancement of the magnetic compared to the thermal Grüneisen parameters in these systems results from the strong volume dependence of R_W [20]. In this respect $YbRh_2Si_2$ is different to nearly ferromagnetic metals, as the observed scaling behavior between T_K and $B^\star$ indicates $\Gamma_B \simeq \Gamma_T$. Probably, the pressure dependence of R_W is small compared to that of T_K. Indeed, a similar R_W value has been observed in $YbRh_2(Si_{0.95}Ge_{0.05})_2$ [21].

Our results are summarized in the $T - B$ phase diagram displayed in Fig. 4. Here the phase boundary of the AF state at low fields has been determined from kinks in both

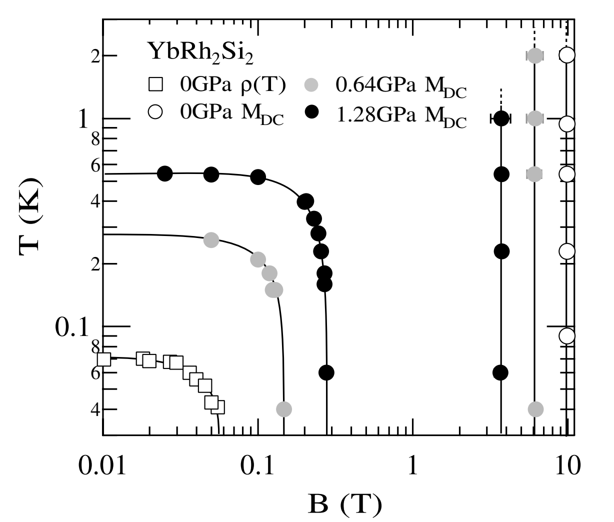

FIG. 4. Temperature-magnetic field phase diagram of $YbRh_2Si_2$ for $B \perp c$ as $\log T$ vs $\log B$. White, gray, and black symbols indicate points at 0, 0.64, and 1.28 GPa, respectively. Circles and squares are data points from magnetization and resistivity measurements, respectively. Lines are guides to the eye.

constant temperature and constant field scans, respectively. Zero-field extrapolations of the phase boundaries agree well with the $T_N(P)$ results of electrical resistivity measurements under hydrostatic pressure [15]. In all $M(B)$ measurements under pressure, the field $B^\star$ at which the kink occurs is independent of temperature. The anomaly broadens rapidly with increasing temperature, and the kink disappears around 2 K at ambient pressure and 1 K at the highest pressure of this study. Thus, the transition occurs only in the coherent regime at $T \ll T_K(P)$.

We discuss the peculiar anomaly at $B^\star$ by comparing it with the metamagneticlike transition in HF compounds such as $CeRu_2Si_2$ and $CeCu_6$. Both these compounds as well as UPt_3 have been reported to exhibit AF short-range correlations at low temperatures [22–24]. In $CeRu_2Si_2$ the AF correlations show a steplike decrease at B_M and disappear at higher fields. Therefore, metamagnetism in these HF compounds can be interpreted as a transition from an "almost" antiferromagnetically ordered HF phase below B_M to a ferromagnetically polarized localized $4f$ state without AF correlations. It seems reasonable to suppose that the intensity of AF correlations is related to the strength of the metamagneticlike behavior. The weaker peak in $\chi(B)$ at B_M for $CeCu_6$ is consistent with a smaller intensity of AF correlations compared to that in $CeRu_2Si_2$. In $YbRh_2Si_2$, on the other hand, AF correlations have been found to persist only in close vicinity to the critical field B_c, and the field-induced LFL state for $B > B_c$ is dominated by strong ferromagnetic fluctuations [12]. Their polarization with increasing B causes the large magnetization already well below the transition at $B^\star$.

In $CeRu_2Si_2$, the strength of the AF correlations and the Kondo interaction are comparable and thus the strong reduction of the former and the localization of the $4f$ electrons happen at the same field. Because of the very weak RKKY intersite interaction in $YbRh_2Si_2$, evidenced by the very low ordering temperature T_N, the AF correlations cannot persist at fields needed to destroy the Kondo interaction in this system. Thus in contrast to the metamagneticlike behavior in other HF systems resulting from the crossover from an AF correlated itinerant to a ferromagnetically polarized localized $4f$-moment state, the $B^\star$ anomaly in $YbRh_2Si_2$ may result from an itinerant to localized transition with FM polarization at both sides. It is worth noting that the relatively small critical field $B^\star = 9.9$ T is suitable for dHvA experiments to study Fermi surface properties below and above $B^\star$. These experiments will provide crucial evidence for the localization of the $4f$ electrons.

To conclude, a new type of field-induced suppression of the HF state has been discovered by low-temperature magnetization measurements on $YbRh_2Si_2$. At ambient pressure, we have observed a broadened transition at $B^\star = 9.9$ T which is accompanied by a decrease of the quasiparticle mass. The use of a miniaturized hydrostatic pressure cell for low-T magnetization experiments has revealed a clear one-to-one correlation between the transition field $B^\star$ and the Kondo temperature. Both are strongly pressure dependent with a Grüneisen parameter of about -130. Strong ferromagnetic fluctuations present in the HF state cause the unique difference of $YbRh_2Si_2$ compared to all other HF systems.

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