Field-induced suppression of the heavy-fermion state in YbRh$_2$Si$_2$

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We report DC magnetization measurements on YbRh$_2$Si$_2$ 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 $B^*=9.9$ 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 $B^*$. Hydrostatic pressure experiments reveal a clear correspondence of $B^*$ 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 metamagnetic-like transition from the itinerant to the localized $f$-electron state which is in contrast to the smooth cross-over in temperature variation at zero field. Such metamagnetic-like behavior has been observed in Ce- and U-based HF compounds such as CeRu$_2$Si$_2$, CeCu$_6$ and UPt$_3$. The metamagnetic transition in CeRu$_2$Si$_2$ 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. Reflecting the localization of $4f$-electrons, the Sommerfeld coefficient $\gamma$ in CeRu$_2$Si$_2$ is strongly suppressed above the critical field $B_M$. It is also noteworthy that the transition is accompanied with a sharp step in magnetostriction $\Delta L(B)/L$. The step produces a sudden change in hybridization between $4f$- and conduction electrons. In the case of Yb-based compounds YbCu$_{3-x}$Ag$_x$ shows a metamagnetic-like smooth cross-over from valence-fluctuating state to a stable Yb$^{3+}$ state with localized magnetic moments.

In this Letter we report a new type of field-induced suppression of the HF state in the Yb-based compound YbRh$_2$Si$_2$. 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 $T_K = 70$ mK. 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. 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. 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 $^{28}$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. These FM fluctuations lead to a strongly enhanced magnetic susceptibility as evidenced by a Sommerfeld-Wilson ratio of $R \approx 14$.

The Gruneisen 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. Assuming a correlation between the characteristic field $B^*$, necessary to suppress the Kondo state, and $T_K$, one would expect a strong volume dependence of $B^*$ 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 $B^*$ with pressure is expected. In order to search for an itinerant-localized $4f$-electron transition in YbRh$_2$Si$_2$ under magnetic field, we have performed demagnetization 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 ($\rho_0 = 1 \mu\Omega\cdot cm$) were grown from In-flux as described earlier. The DC magnetization was measured utilizing a high-resolution capacitive Faraday magnetometer. 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$_2$Si$_2$ 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$_2$Si$_2$ 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 quasi-adiabatic 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 $B^\ast = 9.9$ T, we ascribe, as discussed, to the itinerant-localized transition of the 4$f$-electrons. For magnetic fields $B > B^\ast$, 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 $B^\ast = 6.2$ 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 [13]. 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 $\sim 1$ GPa. As shown in the inset of Fig.1, the critical field $B_L$ 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 antiferromagnetically ordered state.

As shown in Figure 2, both the differential susceptibility $\chi(B) = dM/dB$ at 0, 0.64 and 1.28 GPa (left axis) as well as the Sommerfeld coefficient $\gamma$ at ambient pressure (right axis). Arrows indicate critical field $B^\ast$ above which the 4$f$-states are localized.
low $B^*$. The step-like decrease at $B^*$, 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 [10]. 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 [10]. Further on, at $P=0$ and above $B^*$, $\gamma(B)$ slightly deviates from the relation $\gamma \propto \chi$. This implies that $R_W$ decreases significantly at $B^*$. Note, that $\gamma$ above $B^*$ has a residual value $\sim 100\text{mJ/mol-K}^2$ which is still large for a local moment system. This might indicate residual Kondo-type interactions persisting even above $B^*$. In CeRu$_2$Si$_2$, too, relatively large $\gamma$ values beyond the metamagnetic transition have been found at magnetic fields far above $B_M(\sim 80\text{mJ/mol-K}^2$ 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$_2$Si$_2$ and those observed at metamagnetic transitions in, e.g., CeRu$_2$Si$_2$ 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$_2$Si$_2$ 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 $B^*$ 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 [6], 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 [13]. As shown in Fig. 3, a correlation between $T_0$ and $B^*$ is very probable. The exponential decrease with increasing pressure is compatible with the Kondo temperature $T_K \propto \exp(-1/J_{sf}D_T(\varepsilon_F))$ being determined by the product of the $4f$-conduction electron exchange integral, $J_{sf}$, and conduction electron density of states at the Fermi energy, $D_T(\varepsilon_F)$.

Using the isothermal compressibility $\kappa_T = 5.3 \times 10^{-12} \text{ Pa}^{-1}$ [21] we obtain the “thermal” Gr"uneisen parameter $\Gamma_T = 1/\kappa_T \times \partial \ln T_K/\partial P = -132 \pm 6$. The “magnetic” Gr"uneisen parameter, derived from the pressure dependence of the characteristic field $B^*$, $\Gamma_B = 1/\kappa_T \times \partial \ln B^*/\partial P$, equals $\Gamma_T$ because of the same slope for $T_0$ and $B^*$ in their pressure dependences. This resembles the case of CeRu$_2$Si$_2$ for which $\Gamma_B$, determined from the pressure dependence of the metamagnetic transition equals $\Gamma_T$ as well. One important difference of YbRh$_2$Si$_2$ compared to CeRu$_2$Si$_2$ is, however, the strongly enhanced Sommerfeld-Wilson ratio in the former system. A systematic comparison of thermal and magnetic Gr"uneisen parameters for various systems has been made by A. B. Kaiser and P. Fulde [19]. 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 Gr"uneisen parameter is much larger than the thermal one. The enhancement of the magnetic compared to the thermal Gr"uneisen parameters in these systems results from the strong volume dependence of $R_W$ [19]. In this respect YbRh$_2$Si$_2$ is different to nearly ferromagnetic metals, as the observed scaling behavior between $T_K$ and $B^*$ 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 [20].

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 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 [13]. In all $M(B)$ measurements under pressure, the field $B^*$ 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 $B^*$ by comparing it with the metamagnetic-like transition in HF compounds such as CeRu$_2$Si$_2$ and CeCu$_6$. Both these compounds as well as UPt$_3$ have been reported to exhibit AF short-range correlations at low temperatures [23, 27]. In
CeRu$_2$Si$_2$ the AF correlations show a step-like 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 4$f$-state without AF correlations. It seems reasonable to suppose that the intensity of AF correlations is related to the strength of the metamagnetic-like 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$_2$Si$_2$. In YbRh$_2$Si$_2$, 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$_2$Si$_2$, 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 4$f$-electrons happen at the same field. Because of the very weak RKKY intersite interaction in YbRh$_2$Si$_2$, 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 metamagnetic like behavior in other HF systems, resulting from the cross-over from an AF correlated itinerant to a ferromagnetically polarized localized 4$f$-moment state, the $B^\star$ anomaly in YbRh$_2$Si$_2$ 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 4$f$-electrons.

To conclude, a new type of field-induced suppression of the HF state has been discovered by low-temperature magnetization measurements on YbRh$_2$Si$_2$. 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$_2$Si$_2$ compared to all other HF systems.

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