PHYSICAL REVIEW B **67**, 180403(R) (2003)

Unusual behavior of the low-moment magnetic ground state of $YbRh_2Si_2$ under high pressure

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(Received 21 March 2003; published 19 May 2003)

High-pressure investigations of the ground state properties of the non-Fermi-liquid system $YbRh_2Si_2$ (^{170}Yb-Mössbauer effect, electrical resistance and x-ray diffraction) reveal unusual features that are different from those found in Ce-related systems. We show that the ordered state with a low magnetic moment persists in a very large pressure range up to ≈ 10 GPa and is associated with spin fluctuations. At 10 GPa the system undergoes a first-order magnetic phase transition to a high-moment state with $\mu_{Yb}=1.9(1)\mu_B$ and magnetic ordering temperature $T_m=7.5(4)$ K at 16.5 GPa. A magnetic phase diagram for $YbRh_2Si_2$ in the (p,T) space is suggested.

DOI: 10.1103/PhysRevB.67.180403 PACS number(s): 75.20.Hr, 75.30.Mb, 76.80.+y, 71.27.+a

In recent years a growing interest has been devoted to investigate strongly correlated electron systems, in particularly heavy fermion (HF) metals which are located at or close to a magnetic quantum critical point (QCP).[1] This is due to the fact that such systems (Ce-, U- or Yb-based HF compounds and alloys) develop low temperature thermodynamic, transport, and magnetic properties [so called non-Fermi-liquid (NFL) behavior] that essentially deviate from those predicted by Landau Fermi-liquid theory.[2] Most interesting is that the ground state properties of this class of materials can be *tuned* around a magnetic QCP ($T_m \rightarrow 0$ K) by a control parameter such as doping, pressure, or magnetic field, and thereby allows one to gain new information about this unconventional metallic state.

In fact, there are increasing number of examples of Ce- and U-based systems which have been found to exhibit magnetic quantum critical points either by doping[3–6] or by applying external pressure.[7–9] Well known examples for a pressure tuning of a QCP are $CeCu_{6-x}Au_x$,[7] $CePd_2Si_2$,[8] and $CeIn_3$,[9] which undergo a *second-order* magnetic-to-nonmagnetic transition through a QCP. In the case of $CePd_2Si_2$ and $CeIn_3$ even superconductivity appears near the antiferromagnetic QCP. Among this class of systems, $YbRh_2Si_2$ has attracted considerable excitement as being the *first* ordered Yb-based HF compound which is located in the vicinity of a QCP and exhibits a pronounced NFL behavior at ambient pressure over a temperature range of more than one decade as obtained from low temperature resistivity ($\Delta\rho \propto T$) and specific heat ($\triangle C/T \propto \ln T$) measurements.[10] The proximity of $YbRh_2Si_2$ to a magnetic QCP is manifested by the observation of an antiferromagnetically (AF) ordered state with extremely low value of $T_N=70$ mK, which can either be suppressed to $T_N \rightarrow 0$ by lattice expansion via Ge substitution (x) of $\sim 5\%$, $YbRh_2(Si_{1-x}Ge_x)_2$,[11] or by applying a very small external magnetic field.[12] On the other hand, T_N increases with pressure [$p \leqslant 2.7$ GPa; $T_N \approx 1$ K at 2.7 GPa Refs. (10 and 11)]. A further interesting feature of the AF ground state is its very tiny value of the ordered (static) magnetic moment [$\mu_{Yb} \approx (10^{-2}$–$10^{-3})\mu_B$] as reported from very recent muon spin relaxation (μSR) measurements.[13] In addition, the existence of spin fluctuations in this low-moment (LM) state has been reported from low temperature ^{29}Si NMR (Ref. 14) and μSR (Ref. 13) experiments on $YbRh_2Si_2$.

Regarding the pressure effect on the ground state properties of Yb HF systems, unlike in Ce-based compounds ($4f^1$,Ce^{3+}) where pressure suppresses magnetism, pressure induces (enhances) magnetism in Yb-based ($4f^{13}$,Yb^{3+}) compounds. This is due to the fact that the hybridization strength parameter J between f and conduction electrons which determines the ratio between the characteristic energies for the Kondo effect (T_K) and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction (T_{RKKY})is decreased by applying pressure for Yb systems.[15] Since $YbRh_2Si_2$ is located in the vicinity of a QCP and displays weak magnetic order with a low magnetic moment (i.e., the ratio T_K/T_{RKKY} is just below the critical value for disappearance of magnetism), application of pressure offers a unique possibility to investigate the nature of the LM state as well as its expected evolution towards a high-moment (HM) magnetic state at sufficiently high pressures.[16] Such information is of fundamental interest for a better understanding of quantum critical phenomena in this class of systems.

In this Communication, we show that the LM state of $YbRh_2Si_2$ persists up to a very high pressure of about 10 GPa, and exhibits spin fluctuations in this pressure range. At 10 GPa we find a first-order magnetic phase transition from the LM state to a magnetically ordered state with a HM [$\mu_{Yb}=1.9(1)\mu_B$ and $T_m=7.5(4)$ K at 16.5 GPa]. Finally, we suggest a temperature-pressure phase diagram for $YbRh_2Si_2$.

High quality $YbRh_2Si_2$ single crystals (residual resistivity ~ 1 $\mu\Omega$ cm) were prepared as described in Ref. 10. The pressure dependence of the lattice constants at 300 K up to 21 GPa was measured on powdered samples by energy dispersive x-ray diffraction at HASYLAB using the diamond anvil cell (DAC) technique. The same type of DAC has been used for electrical resistance measurements up to 21 GPa between 1.7 and 295 K and 70 mK $\leqslant T \leqslant$ 25 K in an external magnetic field (B_{ex}) up to 6 T. ^{170}Yb high pressure Mössbauer effect (ME) measurements up to 17.5 GPa at temperatures 1.3 K $\leqslant T \leqslant$ 8 K have been performed with a $^{170}TmB_{12}$ source (35 mCi) using a specially designed large volume pressure setup using industrial diamond anvils.[19]

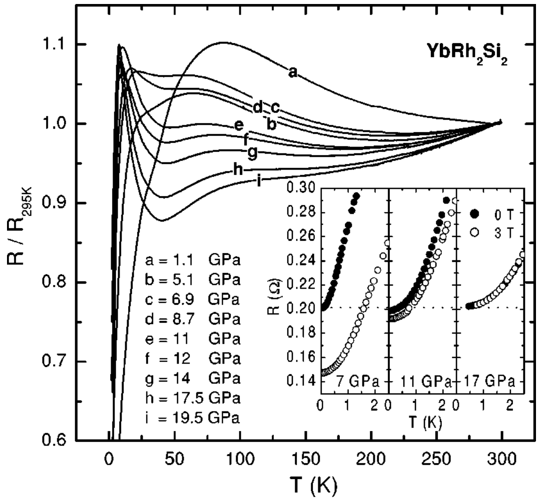

FIG. 1. Temperature dependence ($1.7\ \mathrm{K} \leqslant T \leqslant 295\ \mathrm{K}$) of the electrical resistance normalized to its room temperature value for YbRh$_2$Si$_2$ at different pressures. The inset shows selected $R(T,p)$ curves in an external magnetic field B_{ex} for $70\ \mathrm{mK} \leqslant T \leqslant 2.5\ \mathrm{K}$.

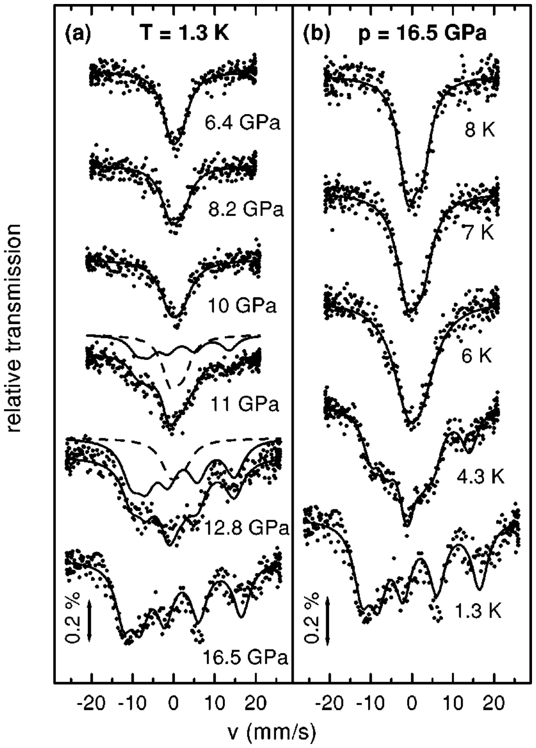

FIG. 2. ^{170}Yb Mössbauer spectra of YbRh$_2$Si$_2$: (a) at $T = 1.3$ K for some selected pressures and (b) at $p = 16.5$ GPa and different temperatures. The dashed and solid lines in the 11- and 12.8-GPa spectra represent the contibution of the paramagnetic and magnetic components, respectively.

Figure 1 displays the temperature dependence of the electrical resistance $R(T,p)$ normalized to its value at room temperature $R(295\ \mathrm{K},p)$ for some selected pressures. For $p = 1.1$ GPa the resistance shows a broad maximum around 80 K and a weak shoulder near 25 K. Upon increasing pressure a maximum develops out of the shoulder (T_{max}) at low temperature which is shifted to lower temperatures and becomes sharp at higher pressures. The low temperature maximum T_{max} which is observed in many HF compounds[15] scales with the Kondo temperature (T_K), whereas the broad maximum (T_{CEF}) at higher temperature is due to a Kondo scattering of the conduction electrons at the first excited crystal field state.

Due to the fact that up to $p = 8.7$ GPa the two maxima in the resistance at T_{max} and T_{CEF} are not well defined it is very difficult to determine the value of T_K and its pressure dependence. Nevertheless we find that T_{max} first decreases with increasing p (from 25 to 10 K) and then nearly saturates around 8 K at pressures higher than 11 GPa. This decrease of T_{max} (or T_K) with increasing p indicates a corresponding decrease of J (s. above), whereas the saturation of T_{max} at higher pressures reflects a growing of the magnetic RKKY interaction. Such a type of behavior has already been observed in nonmagnetic HF compounds, e.g., Yb$_2$Ni$_2$Al (Ref. 17) and intermediate valent compounds, e.g., YbCu$_2$Si$_2$,[18] in which a pressure-induced magnetic phase transition occurs above pressure values where T_{max} nearly saturates. Accordingly, one would expect a change of the magnetic ground state of YbRh$_2$Si$_2$ above $p = 11$ GPa. It should be mentioned that no clear anomaly indicating magnetic ordering was found in the resistance curves at least down to 1.7 K up to the highest pressure ($p = 21$ GPa). Further low temperature electrical resistance data down to 70 mK at high pressure and in external magnetic field (shown in the inset of Fig. 1) will be discussed later.

To obtain microscopic information about the effect of pressure on the low moment state of YbRh$_2$Si$_2$, we now consider the ^{170}Yb high pressure ME measurements. This experimental technique allows one to determine the pressure dependence of the Yb magnetic moment $\mu_{Yb}(p)$, the ordering temperature $T_m(p)$, and the electric field gradient (EFG) at the Yb site which reflects the Yb local site symmetry. $\mu_{Yb}(p)$ is obtained from the effective magnetic hyperfine field (hf) B_{eff} at the ^{170}Yb nucleus which is directly proportional to the value of the Yb ordered magnetic moment [$B_{eff} = C \times \mu_{Yb}$, where $C = 102\ \mathrm{T}/\mu_B$ (Ref. 20)]. The change of EFG with p is obtained from the pressure dependence of the electric quadrupole splitting $E_Q = eQV_{zz}$ where V_{zz} is the EFG at the Yb nucleus and Q is the nuclear quadrupole moment of the $I = 2$ excited state.

Figure 2(a) shows some selected ME spectra collected for YbRh$_2$Si$_2$ up to 16.5 GPa and at 1.3 K. As shown in the figure, below 10 GPa one observes paramagnetic spectra with a small value of E_Q which slightly increases with increasing pressure. At 10 GPa (1.3 K) one observes a *pressure-induced* spectral line broadening (about 20%). At $p = 11$ GPa the spectral shape changes significantly and best fit to the data is obtained by assuming a superposition of a well-defined paramagnetic and magnetic components of about 51% and 49%, respectively. A value of B_{eff} of 150 T which corresponds to a value of $\mu_{Yb} = 1.5(1)\mu_B$ at 1.3 K was deduced from the analysis of the magnetic component. By further increasing the pressure above 11 GPa the magnetic component in the ME spectra grows at the expense of the paramagnetic one, and at $p = 16.5$ GPa and $T = 1.3$ K the whole spectrum shows an asymmetrical hf splitting due to combined magnetic and quadrupole interactions with $B_{eff} = 191$ T $[\mu_{Yb} = 1.9(1)\mu_B]$ and $E_Q = 8.8(4)$ mm/s, assuming B_{eff} and V_{zz} to be parallel. This indicates a pressure-induced first order magnetic phase transition at $p_c \approx 10$ GPa from the LM state to a magnetically ordered state with a HM. Note that the HM value is significantly reduced compared to the free ion Yb^{3+} value of $4\mu_B$ due to the crystal electric field effect. The analysis of the ME spectra in the pressure range $(10\ \mathrm{GPa} < p \leqslant 16.5\ \mathrm{GPa})$ shows that the magnitude of E_Q is temperature independent between 8 and 1.3 K. This clearly shows that the onset of magnetic ordering below 7.5 K is not connected with a lattice distortion or a structural phase transition,[21] and suggests that the Yb moments in the HM phase are parallel to V_{zz}, i.e., aligned along the c axis.

To construct a magnetic phase diagram for $\mathrm{YbRh_2Si_2}$ in the (p,T) space we have determined the pressure dependence of T_m by measuring the temperature dependence of B_{eff} at different pressure points. We first consider the HM state above 10 GPa. Figure 2(b) displays the temperature dependence of the ME spectra at $p = 16.5$ GPa between 8 and 1.3 K. As is evident from the figure the total width of the paramagnetic resonance line increases rapidly by lowering the temperature below 8 K, resulting in a well-resolved magnetic hf splitting at 4.3 K which almost reaches its saturation value at 1.3 K. The analysis of the change of the linewidth with T allows us to obtain a value of $T_m = 7.5(4)$ K at 16.5 GPa. With a knowledge of $T_m(p)$ and the magnetic moment at finite temperature, the saturation value of the Yb magnetic moment μ_{Yb} can be calculated assuming a $J = 1/2$ Brillouin function appropriate for a well isolated Kramer's doublet. Performing these calculations we obtain nearly the *same* value of ordered magnetic moment $\mu_{Yb} = 1.9(1)\mu_B$ for pressure values above $p_c = 10$ GPa. This clearly confirms that the LM→HM phase transition at p_c is of first order.

Now we discuss the pressure dependence of T_m related to the LM state. In contrast to the HM state, in the LM state we find no resolved magnetically hf split ME spectra down to 1.3 K $(3.5\ \mathrm{GPa} < p < 10\ \mathrm{GPa})$. However, a *temperature-induced* broadening of the spectral lines is observed around 1.3 K, indicating an onset of magnetic order at this temperature. Since the lowest accessible temperature is 1.3 K, it is not possible to estimate the values of the Yb magnetic moments in the LM phase with increasing pressure from such line broadening. We find that T_m is nearly pressure independent in the pressure range $(3.5\ \mathrm{GPa} < p < 10\ \mathrm{GPa})$. Such a pressure independent behavior of T_m is not likely expected because of (i) the large initial increase of T_m with pressure up to 2.7 GPa $[dT_m/dp \approx 0.3$ K/GPa (Ref. 10)], and (ii) the continuous decrease of T_K with pressure $(0 < p < 10\ \mathrm{GPa})$ which reflects a corresponding decrease in the hybridization strength J.

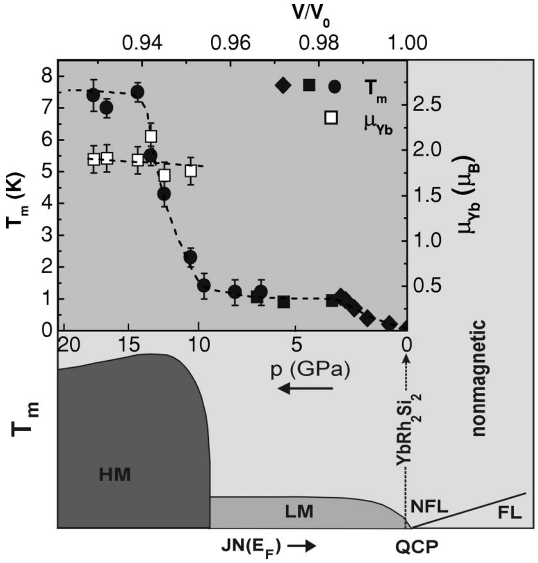

FIG. 3. The upper part shows the pressure-temperature magnetic phase diagram for $\mathrm{YbRh_2Si_2}$ based on the experimental data for the ordering temperature (T_m) determined by ME (●) and electrical resistance [(■) (Ref. 22) and (◆) (Ref. 10)]. In addition the pressure dependence of the magnetic moment μ_{Yb} (□) is shown. The lower part illustrates the suggested different phases in the $[JN(E_F),T]$-phase diagram. $N(E_F)$ is the electronic density of states at the Fermi energy.

Before discussing the pressure dependence of T_m we first consider our result of the effect of external magnetic field (B_{ex}) on $R(T,p)$ down to 70 mK. As shown in the inset of Fig. 1, application of $B_{ex} = 3$ T in the LM state (7 GPa) causes a strong reduction of the low temperature resistance $(70\ \mathrm{mK} \leqslant T \leqslant 2.5\ \mathrm{K})$ of about 28% (negative magnetoresistance) whereas above the LM→HM transition (11 GPa) and in the HM state (17 GPa) almost no change is observed. This indicates that the external magnetic field suppresses spin fluctuations in the LM state $(3\ \mathrm{GPa} < p < 10\ \mathrm{GPa})$, and thereby causes a strong reduction of the low temperature resistance. These findings clearly show that spin fluctuations survive up to $p < 10$ GPa and that their existence has to be considered to understand the pressure dependence of T_m. Despite the lack of any information about the nature of spin fluctuations in the LM state under high pressure, we would like to suggest a qualitative explanation of the unusual behavior of $T_m(p)$. The starting point is to consider the complex nature of spin fluctuations in $\mathrm{YbRh_2Si_2}$ at ambient pressure. According to very recent NMR measurements,[14] it is suggested that the spin fluctuations with finite q vectors which are associated with the formation of the low moment antiferromagnetic (LMAF) state coexists with large spin fluctuations with $q = 0$. Starting near the QCP, the large increase of T_m with p in the low pressure range up to $p = 2.7$ GPa could be explained if one assumes a corresponding increase of the static part of the LMAF state which would lead to a decrease of its associated spin fluctuations. For $3.5\ \mathrm{GPa} < p < 10\ \mathrm{GPa}$, far away from the QCP, the LMAF state becomes stable showing a pressure independent behavior of T_m. While these spin fluctuations freeze at temperatures below T_m, other spin fluctuations (not associated with LMAF order) with certain q vectors survive pressures up to 10 GPa and might lead to the formation of the HM state at higher pressures.

Collecting the results of different measurements, the pressure-temperature magnetic phase diagram and the suggested different phases of $YbRh_2Si_2$ are shown in the upper and lower parts of Fig. 3, respectively. As evident from the figure, the LM state survives in a very large pressure range up to about 10 GPa (or $\Delta V/V \approx 5\%$), and then undergoes a first-order LM→HM phase transition with $\mu_{Yb} \sim 1.9 \mu_B$ for $p \geqslant 10$ GPa. The existence of such an unusual behavior of the LM state has not been found so far in any Ce-HF system,[7–9] and is observed in $YbRh_2Si_2$ for the first time, to our knowledge.

In summary, we have used the ^{170}Yb Mössbauer effect, electrical resistance, and x-ray diffraction techniques to investigate the effect of pressure on the ground state properties of the NFL system $YbRh_2Si_2$ which is located in the vicinity of a QCP. We find that a weak AF ordered state with a low moment exists up to very high $p < 10$ GPa and is associated with spin fluctuations in this pressure range. At 10 GPa, the system undergoes a first-order magnetic phase transition from the LM state to a HM state with $\mu_{Yb} = 1.9(1) \mu_B$ and $T_m = 7.5(4)$ K at 16.5 GPa. We think that our results would stimulate further experimental and theoretical efforts for a better understanding of quantum critical phenomena and their relation to magnetic ordering in HF systems.

M.M.A. and J.P.S. are grateful for the financial support by the German/French PROCOPE program. The authors thank the European Science Foundation Program FERLIN for supporting scientific exchange.

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[21] From our x-ray diffraction measurements on $YbRh_2Si_2$ up to 21 GPa at 300 K, we find a smooth pressure-induced decrease of the volume of the tetragonal ($ThCr_2Si_2 type$) unit cell. Using the Murnaghan equation we obtain a bulk modulus $B_0 = 189(15)$ GPa and $B_0' = 5.8(1.7)$. The ratio of the lattice parameters c/a is found to be fairly constant with increasing pressure up to 21 GPa.

[22] T_m was estimated from the temperature where d^2R/dT^2 shows a minimum in the temperature range 70 mK$< T <$2.5 K.