Magnetic-Field Induced Quantum Critical Point in YbRh$_2$Si$_2$

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Abstract

We report low-temperature calorimetric, magnetic and resistivity measurements on the antiferromagnetic (AF) heavy-fermion metal YbRh$_2$Si$_2$ ($T_N = 70$ mK) as a function of magnetic field $B$. While for fields exceeding the critical value $B_{c0}$ at which $T_N \to 0$ the low temperature resistivity shows an $AT^2$ dependence, a $1/(B - B_{c0})$ divergence of $A(B)$ upon reducing $B$ to $B_{c0}$ suggests singular scattering at the whole Fermi surface and a divergence of the heavy quasiparticle mass. The observations are interpreted in terms of a new type of quantum critical point separating a weakly AF ordered from a weakly polarized heavy Landau-Fermi liquid state.

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The study of quantum phase transitions has attracted the interest of many researchers in the last decades, especially since the discovery of the cuprate superconductors. Quantum phase transitions, in contrast to their classical counterparts at $T > 0$ where thermal fluctuations are important, are driven by a control parameter other than temperature, e.g., composition or pressure. A quantum critical point (QCP) commonly separates an ordered from a disordered phase at zero temperature. To study quantum critical behavior the heavy-fermion (HF) systems are very suitable since they can be tuned continuously from an antiferromagnetic (AF) to a paramagnetic (PM) metallic state by the variation of a single parameter, i.e., the strength of the $4f$-conduction electron hybridization $g$, which can be modified by the application of either external pressure or chemical substitution. According to itinerant spin fluctuation theory [1, 2, 3], close to the critical value $g_c$, at which $T_N \to 0$, the abundance of low-lying and long-range spin fluctuations, mediating the interactions between the heavy quasiparticles (QP), gives rise to pronounced deviations from Landau Fermi liquid (LFL) behavior. Instead of being constant as for a LFL, the QP mass and QP-QP scattering cross section, being proportional to the low-temperature coefficients of the electronic specific heat $\gamma(T) = C_{el}(T)/T$ and the electrical resistivity $A(T) = (\rho(T) - \rho_0)/T^2 = \Delta \rho/T^2$, respectively, show a strong increase or even divergence upon cooling to lowest temperatures. The origin of non-Fermi liquid (NFL) behavior, though observed in an increasing number of HF systems [4], is still a subject of controversy [5].

Another type of QCP arises by the suppression of AF order upon applying magnetic fields $B$. By tuning $T_N$ towards zero temperature at a critical field $B_{c0} = B_c(0)$, the AF correlations between the ordered moments are suppressed resulting in a field-aligned (FA) state for $B \geq B_{c0}$. This is very different to the destruction of the ordered moments which occurs at $B = 0$ upon ”$g$-tuning” an antiferromagnet through its QCP at $g_c$ as described above. Up to now, theoretical models for the QCP at $B_{c0}$ are lacking, and only the doped AF systems CeCu$_{6-x}$Ag$_x$ [1, 7] and YbCu$_{5-x}$Al$_x$ [8] had been tuned by magnetic field through this kind of QCP. For the latter system a substantial amount of Al-doping is necessary to induce long-range AF order leading to a broad phase-transition anomaly in zero field. It is not clear in this case, how the observed NFL behavior is influenced by disorder. For single crystalline CeCu$_{5.2}$Ag$_{0.8}$, NFL effects were observed only for fields applied along the easy magnetic direction and do not indicate a divergence of either the QP mass or the QP-QP scattering cross-section [7]. The results were described within the framework of the
self-consistent renormalization theory \[3\] originally developed for the "g-tuned" QCP.

In this letter we concentrate on the HF metal YbRh\(_2\)Si\(_2\) for which pronounced NFL phenomena, i.e., a logarithmic increase of \(C_{el}(T)/T\) and a quasi-linear \(T\)-dependence of the electrical resistivity below 10 K, have been observed above a low-lying AF phase transition \[9\]. This system is highly suited to study the properties of a \(B\)-induced QCP, because i) the AF phase transition is of second order (see below) and the ordering temperature \(T_N = 70\) mK is the lowest among all undoped HF systems at ambient pressure, ii) already very small magnetic fields are sufficient to suppress the AF state, and iii) clean single crystals can be studied, showing very sharp and well defined phase-transition anomalies which do not broaden significantly at finite fields \[11\]. The application of pressure to YbRh\(_2\)Si\(_2\) increases \(T_N\) \[9\] as expected, because the ionic volume of the magnetic 4\(f^{13}\) Yb\(^{3+}\)-configuration is smaller than that of the nonmagnetic 4\(f^{14}\) Yb\(^{2+}\) one. Expanding the crystal lattice by randomly substituting Ge for the smaller isoelectric Si atoms allows one to "g-tune" YbRh\(_2\)(Si\(_{1-x}\)Ge\(_x\))\(_2\) through the QCP at \(x_c = (0.06 \pm 0.01)\) without affecting its electronic properties and, due to the low value of \(x_c\), without introducing significant disorder to the lattice \[10\]. In YbRh\(_2\)(Si\(_{0.95}\)Ge\(_{0.05}\))\(_2\) the NFL behavior extends to the lowest accessible temperatures, in particular \(\Delta\rho(T) \sim T\) is observed from above 10 K to below 10 mK \[10\]. In the following, we report on low-temperature magnetic, thermodynamic and transport properties of undoped YbRh\(_2\)Si\(_2\) which are used to characterize its field-induced QCP.

High-quality single crystalline platelets of YbRh\(_2\)Si\(_2\) were grown from In flux as described earlier \[9\]. The new generation of crystals show a residual resistivity \(\rho_0 \simeq 1\mu\Omega\)cm, i.e., twice as low as \(\rho_0\) of the previous ones. Whereas for the latter no phase transition anomaly at \(T_N\) could be resolved in the resistivity \[9\], the new crystals show a clear kink of \(\rho(T)\) at \(T_N\), see below. For all low-temperature measurements, \(^3\)He/\(^4\)He dilution refrigerators were used. The electrical resistivity and magnetic AC-susceptibility \(\chi_{AC}\) were measured utilizing a Linear Research Co. (LR700) bridge at 16.67 Hz. Amplitudes of 0.1 mA and 1 Oe for the current and magnetic field, respectively, were chosen to determine \(\rho\) and \(\chi_{AC}\). The DC-magnetization, \(M_{DC}\), measurements were performed utilizing a high-resolution capacitive Faraday magnetometer as described in \[13\]. The specific heat was determined with the aid of a quasi-adiabatic heat pulse technique.

YbRh\(_2\)Si\(_2\) exhibits a highly anisotropic magnetic response, indicating that Yb\(^{3+}\) moments are forming an "easy-plane" square lattice perpendicular to the crystallographic \(c\)-direction.
We first discuss the magnetic properties measured with the field applied along the easy tetragonal plane, $B \perp c$. At $B \simeq 0$, $\chi_{AC}(T)$ reveals a sharp AF phase-transition at $T_N = 70$ mK (Fig. 1a). In the paramagnetic state at $T_N \leq T \leq 0.6$ K, the susceptibility follows a Curie-Weiss type behavior implying fluctuating moments of the order of $1.4 \mu_B / Yb^{3+}$-ion and a Weiss-temperature of $\Theta \simeq -0.32$ K. The isothermal magnetization (Fig. 1b) shows a strongly nonlinear response for fields $B \perp c$. For $T < T_N$ a clear reduction in slope is observed above 0.06 T which indicates the suppression of AF order and the transition into the FA state. A smooth extrapolation of $M_{DC}(B)$ for $B > 0.06$ T towards zero field reveals a value of $\mu_s < 0.1 \mu_B$ for the staggered magnetization in the AF state, indicating that the size of the ordered moments is much smaller than that of the effective moments observed in the PM state above $T_N$. Thus a large fraction of the local moments appears to remain fluctuating within the easy plane in the AF ordered state. Their continuous polarization for fields exceeding $B_{c0}$ gives rise to a strong curvature in $M(B)$ for $B \perp c$. For fields applied along the magnetic hard direction, $B \parallel c$, the magnetization shows an almost linear behavior (Fig. 1b) which was found to extend at least up to 58 T [14]. At $T < T_N$ a very tiny decrease in the $M(B)$ slope is observed at about 0.7 T which, according to the resistivity measurements discussed below, represents the critical field $B_{c0}$ for $B \parallel c$.

The low-$T$ resistivity was measured in magnetic fields applied both perpendicular and parallel to the $c$-direction, with the current perpendicular to the field direction in each case (Fig. 2). For $B = 0$ the resistivity follows a quasi-linear $T$-dependence down to about 80 mK, where a sharp decrease, independent of the current direction, is observed. We note that this behavior is not consistent with that observed for SDW systems for which an increase of $\rho(T)$ along the direction of the SDW modulation, indicating the partial gapping of the Fermi surface, should be expected. The absence of this behavior favors the interpretation of local-moment type of magnetic order in YbRh$_2$Si$_2$, compatible with the observation of large fluctuating moments in $\chi_{AC}(T)$ above $T_N$. The resistivity in the AF ordered state is best described by $\Delta \rho = AT^2$ with a huge coefficient, $A = 22 \mu \Omega cm/K^2$, for $20$ mK $\leq T \leq 60$ mK. With increasing $B$, the phase-transition anomaly in $\rho(T)$ shifts to lower temperatures and vanishes at critical fields $B_{c0}$ of about 0.06 T and 0.66 T for $B \perp c$ and $B \parallel c$, respectively. At $B = B_{c0}$, the resistivity follows a linear $T$-dependence down to the lowest accessible temperature of about 20 mK. This observation provides striking evidence for field-induced NFL behavior at magnetic fields applied along both crystallographic directions. At $B > B_{c0}$,
we find $\Delta \rho = A(B) \cdot T^2$ for $T \leq T^*(B)$, with $T^*(B)$ increasing and $A(B)$ decreasing upon raising the applied magnetic field.

Next we turn to our low-temperature specific-heat results, $C(T)$, which contain electronic and hyperfine contributions, while the phonon part can be safely ignored. We use $\Delta C = C - C_Q$, where the nuclear quadrupolar term calculated from recent Mössbauer results \cite{13} amounts to about 5\% of $C(T)$ at 40 mK. As reported in \cite{9}, the zero-field ratio $\Delta C/T$ is proportional to $-\log T$ in a wide temperature window, 0.3 K $\leq T \leq 10$ K, below which an additional (as yet unexplained) upturn occurs. The new measurements, which were performed in small magnetic fields and down to lower temperatures when compared to the previous ones, show a clear mean-field-type anomaly at $T = T_N$ (Fig. 3). Specific heat, therefore, confirms a second-order phase transition, as already concluded from our magnetization measurements. Extrapolating $\Delta C(T)/T$ as $T \to 0$ to $\gamma_0 = (1.7 \pm 0.2)$ J/K$^2$mol reveals an entropy gain at the AF phase transition of only about 0.01R $\cdot$ ln 2. This is in accordance with the small value of the staggered moments and gives further evidence for the weakness of the AF order in YbRh$_2$Si$_2$. The ratio of $A/\gamma_0^2$ taken from the $B = 0$ data in the ordered state is close to that for a LFL \cite{16}, i.e., one with very heavy quasiparticle masses. At small magnetic fields applied along the c-direction the phase-transition anomaly shifts to lower $T$ as observed in both AC-susceptibility and resistivity experiments. However, due to the strong magnetic anisotropy, the sample plate used for the specific-heat measurement could not be aligned perfectly along the hard magnetic direction. Therefore, a critical field $B_{c0}$ of only about 0.3 T was sufficient to suppress AF order completely in these experiments. As shown in Fig. 3, at $B = B_{c0}$ the specific-heat coefficient $\Delta C(T)/T$ increases down to the lowest $T$, indicative of a field-induced NFL ground state. Within 40 mK $\leq T \leq 120$ mK it follows a steep increase with a much larger slope than observed in zero-field at elevated temperatures (see dotted line in Fig. 3 of Ref. \cite{3}). While this anomalous contribution is strongly reduced upon increasing $B$, at magnetic fields $B \geq 1$ T, the nuclear contribution becomes visible at the lowest temperatures, above which a constant $\gamma_0(B)$ value is observed (Fig. 3). $\gamma_0(B)$ decreases in magnitude upon increasing the field.

The results of the low-$T$ experiments are summarized in the $T - B$ phase diagram displayed in Fig. 4. Here, the field dependence of the Néel temperature was determined from the maximum value of the corresponding $d\rho/dT$ vs $T$ curves. For fields aligned in the easy plane, the results agree perfectly well with those obtained from the AC-susceptibility
For $B > B_{c0}$, the characteristic temperature $T^*(B)$ marks the upper limit of the observed $T^2$ behavior in the resistivity.

To study the nature of the field-induced QCP in YbRh$_2$Si$_2$, we analyze the magnetic field dependence of the coefficients $A$, $\gamma_0$ and $\chi_0$ observed for $T \to 0$ in the resistivity, $\Delta \rho = A(B)T^2$, specific heat, $C/T = \gamma_0(B)$ [9], and magnetic AC-susceptibility, $\chi_{AC} = \chi_0(B)$ [9] when approaching the QCP from the field-polarized state. As shown in the inset of Fig. 5, we observe $A \sim \gamma_0^2, A \sim \chi_0^2$ and thus also $\gamma_0 \sim \chi_0$, independent of the field orientation and for all $B$ values exceeding $B_{c0}$. Thus, the FA state can be described by the LFL model, too. Like in the AF ordered state (at $B = 0$) we find that the $A/\gamma_0^2$ ratio roughly equals that observed for many HF systems [16]. This is in striking variance to the SDW scenario for a 2D spin fluid and a 3D Fermi surface [17]. Further on, a very large Sommerfeld-Wilson ratio $R = (\chi_0/\gamma_0)(\pi^2 k_B^2/\mu_0 \mu_{eff}^2)$ of about 14 ($\mu_{eff} = 1.4 \mu_B$) indicates a strongly enhanced susceptibility in the field-aligned state of YbRh$_2$Si$_2$ pointing to the importance of low-lying ferromagnetic ($q = 0$) fluctuations in YbRh$_2$Si$_2$ [19]. Since YbRh$_2$Si$_2$ behaves as a true LFL for $B > B_{c0}$ and $T < T^*(B)$, the observed temperature dependences should hold down to $T = 0$. The field dependence $A(B)$ shown in Fig. 5 measures the QP-QP scattering cross section when, by field tuning, crossing the QCP at zero temperature. Most importantly, a $1/(B - B_{c0})$ divergence is observed indicating that the whole Fermi surface undergoes singular scattering at the $B$-tuned QCP. Furthermore, the relation $A \sim \gamma_0^2$ observed at elevated fields, suggests that also the QP mass diverges, i.e., as $1/(B - B_{c0})^{1/2}$, when approaching $B_{c0}$ [20].

The extremely low value of the critical field applied along the easy plane highlights the near degeneracy of two different LFL states, one being weakly AF ordered ($B < B_{c0}$) and the other one being weakly polarized ($B > B_{c0}$). In fact, the AC-susceptibility measured for fields near $B_{c0}$ shows a sharp increase when approaching the phase transition upon cooling, see 0.04T data in Fig. 1a.

To conclude, a new type of QCP separating an antiferromagnetic from a field-aligned ground state was studied in the clean heavy-fermion metal YbRh$_2$Si$_2$. We observed pronounced NFL behavior at the critical field $B_{c0}$ necessary to suppress antiferromagnetic order. When this system is tuned, at zero temperature, from the field-aligned state towards the QCP by decreasing the applied magnetic field, both the quasiparticle scattering cross section and the quasiparticle mass appear to diverge.
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FIG. 1: Low-temperature AC-susceptibility $\chi_{AC}$ of YbRh$_2$Si$_2$ at varying fields applied perpendicular to the c-axis (a) and isothermal DC magnetization $M_{DC}$ at varying temperatures in magnetic fields applied along and perpendicular to the c-axis. Arrows indicate critical fields $B_{c0}$.

FIG. 2: Low-temperature electrical resistivity of YbRh$_2$Si$_2$ at varying magnetic fields applied along the a- (a) and c-axis (b). For clarity the different curves in $B > 0$ were shifted subsequently by 0.1 $\mu\Omega$cm. Up- and downraising arrows indicate $T_N$ - and upper limit of $T^2$ behavior, respectively. Dotted and solid lines represent $\Delta \rho \sim T^\epsilon$ with $\epsilon = 1$ and $\epsilon = 2$, respectively.
FIG. 3: Specific heat as $\Delta C/T = (C - C_Q)/T$ vs $T$ (on a logarithmic scale) for YbRh$_2$Si$_2$ at varying fields applied parallel to the $c$-axis. $C_Q \sim T^{-2}$ is the nuclear quadrupole contribution calculated from recent Moessbauer results\cite{13}. 
FIG. 4: $T - B$ phase diagram for YbRh$_2$Si$_2$ with $T_N$ as derived from $d\rho/dT$ vs $T$ and $T^*$, the upper limit of the $\Delta \rho = AT^2$ behavior, as a function of magnetic field, applied both along and perpendicular to the $c$-axis. For the latter ones the $B$-values have been multiplied by a factor 11. Lines separating the antiferromagnetic (AF), non-Fermi liquid (NFL) and Landau Fermi liquid (LFL) phase are guides to the eye. Note that the AF phase transition as a function of field is a continuous one, cf. $M_{DC}(B)$ curves in Fig. 1b.
FIG. 5: Coefficient $A = \Delta \rho / T^2$ vs field $B$. Data for $B$ perpendicular to the $c$-direction have been multiplied by 11. Dashed line marks $B_{c0}$, solid line represents $(B - B_{c0})^{-1}$. Inset shows double-log plot of $A$ vs $\gamma_0$ and $A$ vs $\chi_0$ for different magnetic fields. Solid lines represent $A/\gamma_0^2 = 5.8 \cdot 10^{-6} \mu \Omega \text{cm(Kmol/mJ)}^2$ and $A/\chi_0^2 = 1.25 \cdot 10^{12} \mu \Omega \text{cmK}^{-2}/(\text{m}^3/\text{mol})^2$. 