Magnetic-Field Induced Quantum Critical Point in $YbRh_2Si_2$

P. Gegenwart,[1] J. Custers,[1] C. Geibel,[1] K. Neumaier,[2] T. Tayama,[1,3] K. Tenya,[1] O. Trovarelli,[1] and F. Steglich[1]

[1]*Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany*
[2]*Walther Meissner Institute, D-85748 Garching, Germany*
[3]*Institute of Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

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We report low-temperature calorimetric, magnetic, and resistivity measurements on the antiferromagnetic (AF) heavy-fermion metal $YbRh_2Si_2$ ($T_N = 70$ mK) as a function of magnetic field B. While for fields exceeding the critical value B_{c0} at which $T_N \rightarrow 0$ 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 [1–3], close to the critical value g_c at which $T_N \rightarrow 0$, 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 from 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 $CeCu_{6-x}Ag_x$ [6,7] 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 $YbRh_2Si_2$ for which pronounced NFL phenomena, i.e., a logarithmic increase of $C_{el}(T)/T$ and a quasilinear T dependence of the electrical resistivity below 10 K, have been observed upon cooling down to 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 [10]. The application of pressure to $YbRh_2Si_2$ increases T_N [9] as expected, because the ionic volume of the magnetic $4f^{13}$ Yb^{3+} configuration is smaller than that of the nonmagnetic $4f^{14}$ 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 without affecting its electronic properties [12]. In single crystals prepared with the nominal Ge composition $x = 0.05$ no significant disorder is introduced to the lattice and the NFL behavior

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extends to the lowest accessible temperatures: $\Delta\rho \sim T$ is observed from above 10 K to below 10 mK [12]. In the following, we report on low-temperature magnetic, thermodynamic, and transport properties of undoped $YbRh_2Si_2$ which are used to characterize its field-induced QCP.

High-quality single crystalline platelets of $YbRh_2Si_2$ were grown from In flux as described earlier [9]. The new generation of crystals shows a residual resistivity $\rho_0 \simeq 1\ \mu\Omega$ cm, i.e., twice as low as ρ_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, $^3He/^4He$ dilution refrigerators were used. The electrical resistivity and magnetic ac susceptibility χ_{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 ρ and χ_{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 quasiadiabatic heat pulse technique.

$YbRh_2Si_2$ exhibits a highly anisotropic magnetic response, indicating that Yb^{3+} moments are forming an "easy-plane" square lattice perpendicular to the crystallographic c direction [9]. 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. 1(a)]. 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. 1(b)] 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. 1(b)] 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 quasilinear 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 the local-moment type of magnetic order in $YbRh_2Si_2$, 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, $A = 22\ \mu\Omega\,\mathrm{cm/K^2}$, for $20 \leq T \leq 60$ mK (at $B = 0$). 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 and 0.66 T for $B \perp c$ and $B \parallel c$,

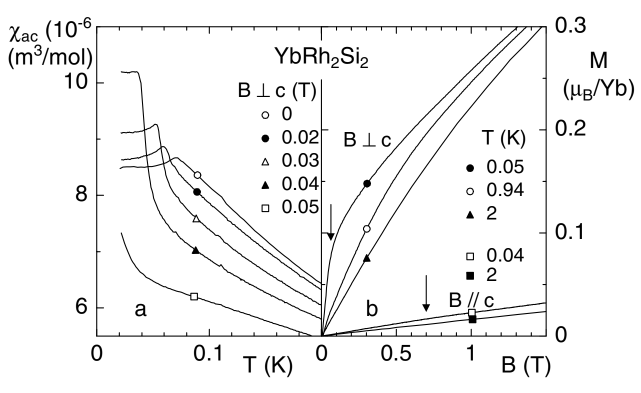

FIG. 1. Low-temperature ac susceptibility χ_{ac} of $YbRh_2Si_2$ at varying fields applied perpendicular to the c axis (a) and isothermal dc magnetization M_{dc} at varying temperatures in magnetic fields applied along and perpendicular to the c axis. Arrows indicate critical fields B_{c0}.

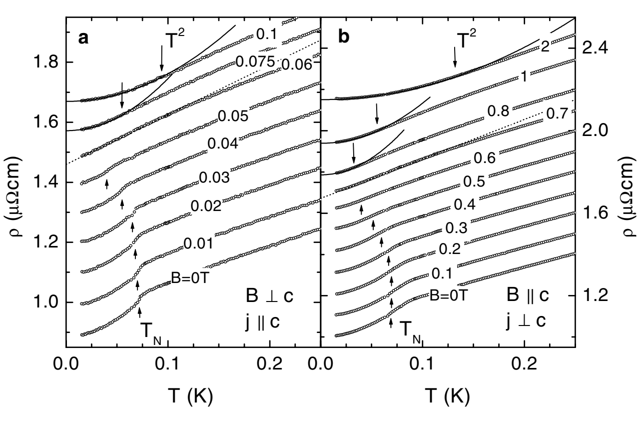

FIG. 2. Low-temperature electrical resistivity of $YbRh_2Si_2$ at varying magnetic fields applied along the a axis (a) and the c axis (b). For clarity the different curves in $B > 0$ were shifted subsequently by $0.1\ \mu\Omega$ cm. Up- and downraising arrows indicate T_N and the upper limit of T^2 behavior, respectively. Dotted and solid lines represent $\Delta\rho \sim T^\epsilon$ with $\epsilon = 1$ and 2, respectively.

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 $\Delta\rho = A(B)T^2$ 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 [15] amounts to about 5% of $C(T)$ at 40 mK. As reported in [9], the zero-field ratio $\Delta C/T$ is proportional to $-\log T$ in a wide temperature window, $0.3 \leq T \leq 10$ 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 $0.01R\ln 2$. This is in accordance with the small value of the staggered moments and gives further evidence for the weakness of the AF order in $YbRh_2Si_2$. The ratio of A/γ_0^2 taken from the $B = 0$ data in the ordered state is close to that for a LFL [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, because of 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 $40 \leq T \leq 120$ 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. [9]). 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 [Fig. 1(a)]. 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 $YbRh_2Si_2$, we analyze the magnetic field dependence of the coefficients A, γ_0, and χ_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

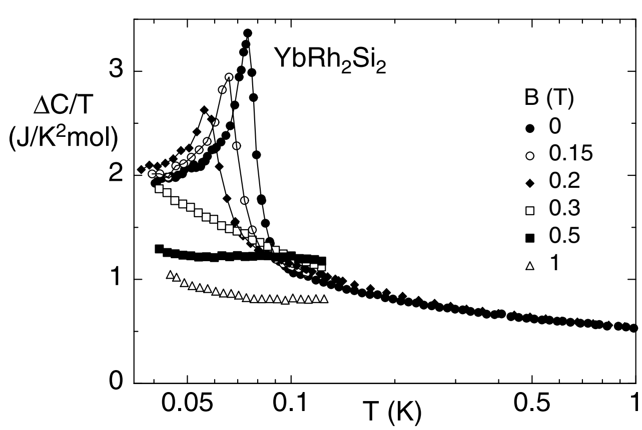

FIG. 3. Specific heat as $\Delta C/T = (C - C_Q)/T$ vs T (on a logarithmic scale) for $YbRh_2Si_2$ at varying fields applied parallel to the c axis. $C_Q \sim T^{-2}$ is the nuclear quadrupole contribution calculated from recent Mössbauer results [15].

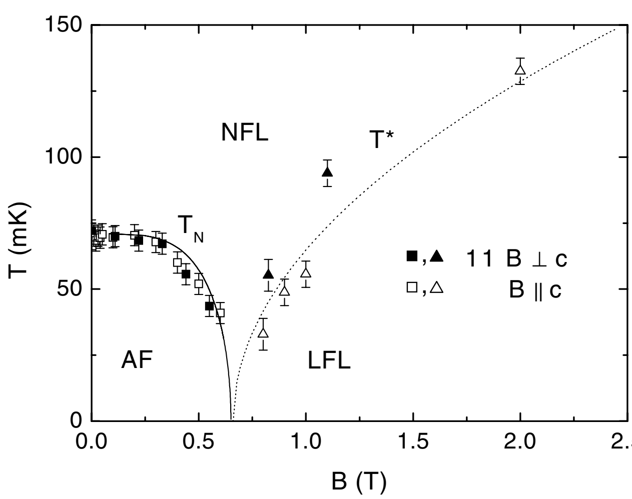

FIG. 4. T-B phase diagram for $YbRh_2Si_2$ with T_N as derived from $d\rho/dT$ vs T and T^*, the upper limit of the $\Delta\rho = AT^2$ behavior, as a function of magnetic field, applied both along and perpendicular to the c axis. For the latter ones the B values have been multiplied by a factor of 11. 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. 1(b).

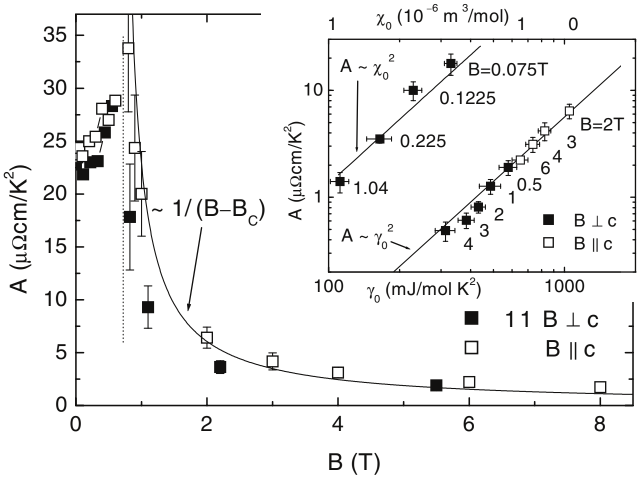

FIG. 5. Coefficient $A = \Delta\rho/T^2$ vs field B. Data for B perpendicular to the c direction have been multiplied by 11. Dashed line marks B_{c0}, and solid line represents $(B - B_{c0})^{-1}$. Inset shows double-log plot of A vs γ_0 and A vs χ_0 for different magnetic fields. Solid lines represent $A/\gamma_0^2 = 5.8 \times 10^{-6}\ \mu\Omega\,\mathrm{cm}\,(\mathrm{K\,mol/mJ})^2$ and $A/\chi_0^2 = 1.25 \times 10^{12}\ \mu\Omega\,\mathrm{cm\,K^{-2}/(m^3/mol)^2}$.

state (at $B = 0$) we find that the A/γ_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 $R = (\chi_0/\gamma_0)(\pi^2 k_B^2/\mu_0\mu_{\mathrm{eff}}^2)$ of about 14 ($\mu_{\mathrm{eff}} = 1.4\mu_B$) indicates a strongly enhanced susceptibility in the field-aligned state of $YbRh_2Si_2$ pointing to the importance of low-lying ferromagnetic ($\mathbf{q} = \mathbf{0}$) fluctuations in $YbRh_2Si_2$ [19]. Since $YbRh_2Si_2$ 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 for $B > B_{c0}$ 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 0.04 T data in Fig. 1(a).

To conclude, a new type of QCP separating an antiferromagnetic from a field-aligned ground state was studied in the clean heavy-fermion metal $YbRh_2Si_2$. 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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