Divergence of the Magnetic Grüneisen Ratio at the Field-Induced Quantum Critical Point in \(YbRh_2Si_2\)

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The heavy-fermion metal \(YbRh_2Si_2\) is studied by low-temperature magnetization \(M(T)\) and specific heat \(C(T)\) measurements at magnetic fields close to the quantum critical point (\(H_c = 0.06 \text{T} \), \(H \perp c\)). Upon approaching the instability, \(dM/dT\) is more singular than \(C(T)\), leading to a divergence of the magnetic Grüneisen ratio \(\Gamma_{\text{mag}} = -(dM/dT)/C\). Within the Fermi liquid regime, \(\Gamma_{\text{mag}} = -G_c(H - H^{(c)})\) with \(G_c = -0.30 \pm 0.01\) and \(H_c^{(c)} = (0.065 \pm 0.005)\text{T}\) which is consistent with scaling behavior of the specific-heat coefficient in \(YbRh_2(Si_{0.98}Ge_{0.02})_2\). The field-dependence of \(dM/dT\) indicates an inflection point of the entropy as a function of magnetic field upon passing the line \(T^*(H)\) previously observed in Hall- and thermodynamic measurements.

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Quantum criticality is a topic of extensive current research interest in condensed matter physics because it results in unusual finite-temperature properties and promotes the formation of novel states like unconventional superconductivity (for recent reviews see [1, 2, 3]). A quantum phase transition occurs at \(T = 0\) upon tuning an external parameter \(r\) like pressure, doping or magnetic field to a critical value \(r_c\). For a continuous (second-order) phase transition at \(r_c\), a quantum critical point (QCP) emerges, which has wide reaching influence on the temperature dependence of thermodynamic and transport properties. In metallic systems near magnetic QCPs pronounced non-Fermi liquid (NFL) effects have been observed, which depend on the dimensionality (\(D\)), type of magnetic interaction, i.e. antiferromagnetic (AF) or ferromagnetic (FM), as well as type of quantum criticality (itinerant, locally critical, Kondo-breakdown, etc.) [4, 5, 6, 7, 8, 9]. Such NFL effects are related to an anomalous enhancement of the entropy \(S(T, r)\) near the QCP, resulting from quantum critical fluctuations. For a pressure-tuned QCP \((r \propto p - p_c, p_c:\text{ critical pressure})\) it has been pointed out, that the Grüneisen ratio \(\Gamma \propto \alpha/C\) of thermal expansion \(\alpha\) to specific heat \(C\) diverges in the approach of the QCP [10, 11], as recently found for various systems [12, 13, 14, 15]. When the control parameter to tune the system to the QCP is the magnetic field \((r = H - H_c, H_c:\text{ critical magnetic field})\), a corresponding divergence has been predicted for the magnetic Grüneisen parameter \(\Gamma_{\text{mag}} = -(dM/dT)/C\) [11]. This property equals the magnetocaloric effect, i.e. is proportional to the slope of isentropes in the temperature vs. magnetic field phase diagram [11]. Below, we demonstrate the use of magnetization for characterizing a NFL and report for the first time a divergence of \(\Gamma_{\text{mag}}\) in the approach of a (field-induced) QCP.

We focus on tetragonal \(YbRh_2Si_2\), which is a clean and stoichiometric heavy-fermion metal that displays a magnetic field-tuned QCP at \(H_c = 0.06 \text{T} (0.66 \text{T})\) for \(H \perp c\) \((H \parallel c)\) [10]. This QCP arises when very weak AF ordering at \(T_N = 70 \text{mK}\), with an ordered moment as small as \(2 \times 10^{-5}\mu_B/\text{Yb}\) [17], is continuously suppressed by magnetic field. Various thermodynamic, magnetic and transport experiments on \(YbRh_2Si_2\) as well as its slightly Ge-doped variant \(YbRh_2(Si_{1-x}Ge_x)_2\) \((T_N = 20 \text{mK}, H_c = 0.027 \text{T}, H \perp c)\) have revealed evidence for quantum criticality, that is controlled by magnetic field [16, 18]: At \(H = H_c\), \(C(T)/T\) exhibits a stronger than logarithmic divergence while \(\rho\) has \(T\)-linear temperature dependence. For \(H > H_c\), Fermi-liquid (FL) behavior is induced, as evidenced from the electrical resistivity, described by \(\rho(T) = \rho_0 + AT^2\), with the coefficient \(A(H)\) diverging towards \(H_c\) proving a field-induced QCP [16, 18]. The Sommerfeld coefficient \(\gamma(H)\) in the FL regime \((H > H_c)\) displays a \((H - H_c)^{-1/3}\) divergence, which is incompatible with the itinerant theory for a spin-density-wave QCP [18]. Bulk susceptibility [19, 20] and nuclear magnetic resonance experiments [21] indicate that in a wide regime of the \(T - H\) phase diagram the critical fluctuations have a FM character. Only for temperatures below 0.4 K and fields below 0.25 T, i.e. very close to the AF ordered phase, AF fluctuations dominate. For \(H > H_c\), a strongly enhanced Sommerfeld-Wilson ratio that exceeds a value of 30 upon approaching the critical field has been observed [19]. Note, however, that the observed temperature dependence of the bulk susceptibility and spin-lattice relaxation rate in the quantum critical regime cannot be explained within the itinerant theory for FM quantum critical fluctuations in either 2D or 3D [22].

The critical Grüneisen ratio \(\Gamma_{cr} = V_{mol}/\kappa_T \times \alpha_{cr}/C_{cr}\) (\(V_{mol}\): molar volume, \(\kappa_T\): isothermal compressibility), where \(\alpha_{cr}\) and \(C_{cr}\) denote the volume thermal expansion and specific heat after subtraction of non-critical contributions [11] has been studied for \(YbRh_2(Si_{1-x}Ge_x)_2\) at zero magnetic field and temperatures down to 80 mK [12]. Below 0.6 K, a \(\Gamma_{cr} \propto T^{-0.7}\) divergence has been obtained, which may indicate a local type of QCP [12]. Indeed, detailed studies of the evolution of the Hall coefficient upon field-tuning the system suggest a drastic change of the Fermi volume due to a localization of the \(4f\)-electrons at the QCP [23]. Furthermore, thermodynamic evidence for an additional energy scale \(T^*(H)\), possibly related to the Kondo-breakdown was obtained from mag-
netostriction and magnetization experiments \[^{[22]}\]. Below, we address the nature of quantum criticality and the evolution of the entropy across \(T^* (H)\) by means of the magnetic Grüneisen ratio, which is the appropriate thermodynamic property for a field-induced QCP.

For our measurements, we used high-quality single crystals \((\rho_0 = 1 \mu \Omega \text{cm})\) characterized before \[^{[10]}\]. The magnetization was measured utilizing a high-resolution capacitive Faraday magnetometer \[^{[24]}\]. Specific-heat measurements have been performed with the quasi-adiabatic heat-pulse technique.

Figure 1 shows the magnetization divided by field \(M/H\) as a function of temperature. For fields above 0.1 T, \(M(T)/H\) tends to saturate, while it keeps increasing down to the lowest temperature below 0.1 T. The strong increase of \(M(T)/H\) with decreasing temperature at the critical field \(H_c = 0.06\) T is reflected in the negative curvature in the inverse of \(M/H\) (see the inset). The deviation from the Curie-Weiss law above 0.3 K may be due to FM correlations in this system. Linear fitting of the data in the low-temperature region \((T < 0.3\) K) yields a Weiss temperature \(\theta = -0.46\) K and an effective magnetic moment \(\mu_{\text{eff}} = 1.6 \mu_B/\text{Yb}^{3+}\) ion, close to the reported values at zero field in a previous ac-susceptibility study \[^{[16]}\]. At \(H > H_c\), \(M(T)/H\) saturates and does not reach zero. At a FM QCP, the magnetization is expected to diverge as \(T \to 0\) with \(\theta = 0\). The finite negative \(\theta\) at \(H_c\) indicates that AF fluctuations dominate close to the QCP \[^{[21]}\].

The temperature derivative of the magnetization for various different magnetic fields at and above the critical field has been investigated between 70 mK and 3 K. In Figure 2, we compare the temperature dependence of \(-(dM/dT)/T\) displayed in part (a) with that of the respective specific heat coefficient \(C(T)/T\) in part (b) and analyze the ratio between both quantities, \(\Gamma_{\text{mag}}\) (Fig. 2c). At the critical field \(H_c = 0.06\) T, both quantities diverge upon cooling, but the divergence in \(-(dM/dT)/T\) is much stronger, resulting in a divergent magnetic Grüneisen ratio. At \(H \approx 0.1\) T, \(-(dM/dT)/T\) diverges even stronger than at 0.06 T in the investigated temperature regime. However, the analysis of the field-dependence of the

![FIG. 1](image1.png)

**FIG. 1:** Magnetization divided by field \(M/H\) of YbRh\(_2\)Si\(_2\) as a function of temperature. Inset: Inverse of \(M/H\) vs temperature. Dashed line indicates linear fit for low-temperature region \((T < 0.3\) K).

![FIG. 2](image2.png)

**FIG. 2:** (Color online) (a) Temperature derivative of the magnetization as \(-dM/dT)/T\) (left axis), (b) electronic specific heat as \(C(T)/T\) and (c) magnetic Grüneisen ratio \(\Gamma_{\text{mag}}\) vs temperature (on a logarithmic scale) for YbRh\(_2\)Si\(_2\) in magnetic fields applied perpendicular to \(c\)-axis. Volume thermal expansion of YbRh\(_2\)(Si\(_{1.05}\)Ge\(_{0.95}\))\(_2\) as \(-\beta/T\) at zero field is shown for comparison in (a) (right axis) \[^{[12]}\]. Inset in (a): Expanded plot for \(-dM/dT)/T\) vs \(T\) for \(H = 0.06\) T. The arrow indicates the position of the maximum. Inset in (b): \(\Gamma_{\text{mag}}\) vs temperature for YbRh\(_2\)Si\(_2\) on a log-log plot. The solid lines represent \(\Gamma_{\text{mag}}(T) \propto T^{-\epsilon}\) with \(\epsilon = 0.7\) and 2.0 for low- and high-temperature regions, respectively. Inset in (c): Field dependence of saturated \(\Gamma_{\text{mag}}\), as \(T \to 0\). The solid line indicates \(-G_r (H - H_{c}^{0})^{-1}\) with \(G_r = -0.3\) and \(H_{c}^{0} = 0.065\) T.
magnetic Grüneisen ratio, discussed below, is consistent with a QCP at 0.06 T and suggests a saturation of the 0.1 T data due to a crossover from NFL to FL behavior at temperatures below 70 mK. Such crossover scales are known to be different for different physical quantities [5]. Indeed, the specific heat coefficient $C(T)/T$ displays such a NFL to FL crossover for $H = 0.1$ T already around 100 mK, whereas the respective crossover in electrical resistivity at the same field is located at 80 mK [16].

In Fig. 2(a) (right axis), we also show the volume thermal expansion as $\beta/T$ for YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$) [12] for comparison. The divergence of $-\langle dM/dT \rangle/T$ is stronger at high temperatures and becomes comparable to $\beta/T$ at low temperatures. The ratio between $\beta$ and $dM/dT$ gives the pressure dependence of the critical field as expressed by $dH_c/dp = V_m \beta / (dM/dT)$, where $V_m$ is the molar volume [11]. This ratio is temperature dependent and decreases with decreasing temperature (not shown). At low temperatures it tends to saturate and the extrapolated value of $V_m \beta / (dM/dT)$ for $T \to 0$ is $(0.19 \pm 0.05) \, \text{T/GPa}$ which is consistent with previous hydrostatic pressure experiments [20].

Interestingly, $-\langle dM/dT \rangle/T$ for $H \geq 0.2$ T shows a maximum (cf. inset of Fig. 2(a) for data at 0.3 T) whose position coincides well with the temperature where the specific heat coefficient passes a maximum, $T_{\text{max}}$. Similar behavior in $C(T)/T$ has also been observed in other heavy-fermion compounds such as CeCu$_{5.9}$Au$_{0.1}$ and Ce$_{0.8}$Y$_{0.2}$Cu$_2$Si$_2$ [25, 26]. The origin of such maxima may be understood qualitatively by the Zeeman splitting of the Kondo resonance [27] which results in a more polarized state. Our results as well as those for CeCu$_{5.9}$Au$_{0.1}$ and Ce$_{0.8}$Y$_{0.2}$Cu$_2$Si$_2$ [25, 26] show that the maximum temperature in $C(T)/T$ increases linearly with increasing field. For the magnetic Grüneisen ratio $\Gamma_{\text{mag}}(T)$, the maxima in $C(T)$ and $-\langle dM/dT \rangle/T$ cancel each other, leading to a monotonous crossover from NFL to FL behavior at fields larger than 0.2 T (Fig. 2(c)).

Before discussing details of $\Gamma_{\text{mag}}(T)$ data, we first summarize the important theoretical conclusions most related to the present study with respect to the degree of assumptions.

1. $\Gamma_{\text{mag}}$ diverges in the approach of any field-induced QCP whenever the characteristic energy scale of a system is continuously suppressed to absolute zero [10].

2. Assuming scaling, the critical behavior is governed by the correlation length $\xi$ and the correlation time $\xi \tau$ such that the temperature dependence in the quantum critical regime $\Gamma_{\text{mag}}(T, H = H_c) \propto T^{-1/vz}$ ($v$ and $z$ are the correlation-length exponent and dynamical critical exponent, respectively) for the field dependence in the FL regime, scaling analysis remarkably predicts not only a universal functional dependence but even its prefactor without any adjustable parameter, i.e. $\Gamma_{\text{mag}}(T = 0, H) = -G_r (H - H_c)^{-1/2}$, with $G_r = v (d - z)$ (d: dimensionality of the critical fluctuations) [10, 11]. Furthermore, this prefactor equals the exponent in the divergence of the Sommerfeld coefficient $\gamma \propto (H - H_c)^{G_r}$ [10, 11]. Thus, the character of the QCP is completely determined by the values of $\gamma$, $z$, and $d$.

3. Within the itinerant theory, the correlation length exponent $v = 1/2$ and the dynamical critical exponent $z$ equals 2 and 3 for AF and FM case, respectively [4].

We now check the consistency of our experimental results with these theoretical predictions. Obviously $\Gamma_{\text{mag}}$ diverges as a function of temperature at $H = H_c = 0.06 \, \text{T}$ (Fig. 2(c)), diverging like $(H - H_c)^{-1}$ as expected from scaling (assumption 2). The fit reveals $-G_r (H - H_c)^{-1}$ with $G_r = (0.065 \pm 0.004) \, \text{T}$ and $G_r = -0.30 \pm 0.01$. For YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$, the field dependence of $\gamma$ within the FL regime has been studied in detail revealing $\gamma \propto (H - H_c)^{-0.33}$ [18] which has an exponent very close to our value of $G_r$. This underlines the thermodynamic consistency of the data. We note also that the obtained value of $G_r$ sets strong constraints for the scaling parameters $\nu$, $d$, and $z$. Within the itinerant theory (assumption 3), in which $\nu = 1/2$, the observed $G_r$ would require $d - z = 2/3$ which is impossible. By contrast, a critical Fermi surface model which may be relevant for a Kondo-breakdown QCP has been proposed [28], in which the electronic criticality is described by $\nu = 2/3$, $z = 3/2$, and $d = 1$, yielding $G_r = -1/3$ similar to our experiments.

Next, we focus on the temperature dependence of the magnetic Grüneisen ratio within the quantum critical regime. Interestingly, divergent behavior is found not only at $H_c$ but also at 0.1 and 0.15 T (see Fig. 2c). However, the obtained
The magnetic entropy follows the field dependence of the quantum critical regime discussed previously in the Hall coefficient [22]. A reduction of spin entropy when entering the entangled heavy-electron fluid upon increasing the magnetic field is consistent with the delocalization of f-electrons upon field tuning through the QCP.

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