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 ($H_c = 0.06$ 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 $\Gamma_{mag} = -(dM/dT)/C$. Within the Fermi-liquid regime, $\Gamma_{mag} = -G_r(H - H_c^{fit})$ with $G_r = -0.30 \pm 0.01$ and $H_c^{fit} = (0.065 \pm 0.005)$ T which is consistent with scaling behavior of the specific-heat coefficient in $YbRh_2(Si_{0.95}Ge_{0.05})_2$. The field dependence of dM/dT indicates an inflection point of the entropy as a function of magnetic field upon passing the line $T^\star(H)$ 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 [1–3]). 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. 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.) [4–9]. 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: critical pressure) it has been pointed out, that the Grüneisen ratio $\Gamma \propto \alpha/C$ of thermal expansion α to specific heat C diverges [10,11], as recently found for various systems [12–15]. When the control parameter to tune the system to the QCP is the magnetic field ($r = H - H_c$, H_c: critical magnetic field), a corresponding divergence has been predicted for the *magnetic* Grüneisen parameter $\Gamma_{mag} = -(dM/dT)/C$ [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].

We focus on tetragonal $YbRh_2Si_2$, which is a clean and stoichiometric heavy-fermion metal that displays a magnetic field-tuned QCP at $H_c = 0.06$ T (0.66 T) for $H \perp c$ ($H \parallel c$) [16]. This QCP arises when very weak AF ordering at $T_N = 70$ mK, with an ordered moment as small as $2 \times 10^{-3} \mu_B$/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 $YbRh_2(Si_{1-x}Ge_x)_2$ ($T_N = 20$ mK, $H_c = 0.027$ 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 ρ 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 $\Gamma_{cr} = (V_{mol}/\kappa_T)(\alpha_{cr}/C_{cr})$ (V_{mol}: molar volume, κ_T: isothermal compressibility), where α_{cr} and C_{cr} denote the volume thermal expansion and specific heat after subtraction of noncritical contributions [10] 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 $T^\star(H)$, possibly related to the Kondo-breakdown was obtained from magnetostriction and magnetization experiments [22]. Below, we address the nature of quantum criticality and the evolution of the entropy across $T^{\star}(H)$ 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 ($\rho_0 = 1\ \mu\Omega$ cm) characterized before [16]. The magnetization was measured utilizing a high-resolution capacitive Faraday magnetometer [24]. Specific-heat measurements have been performed with the quasiadiabatic 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 $\mu_{\rm eff} = 1.6\mu_B/{\rm Yb}^{3+}$ ion, close to the reported values at zero field in a previous ac-susceptibility study [16]. At a FM QCP, the magnetization is expected to diverge as $T \rightarrow 0$ with $\theta = 0$. The finite negative θ 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 0.07 and 3 K. In Fig. 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, $\Gamma_{\rm mag}$ [Fig. 2(c)]. 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 $H = 0.1$ 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 magnetic Grüneisen ratio, discussed below, is consistent with a QCP at 0.06 T. We therefore expect 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].

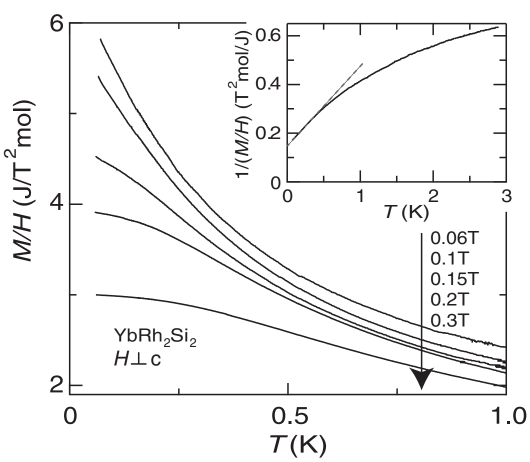

FIG. 1. Magnetization divided by field M/H of $YbRh_2Si_2$ as a function of temperature. Inset: Inverse of M/H vs temperature. The dashed line indicates linear fit for low-temperature region.

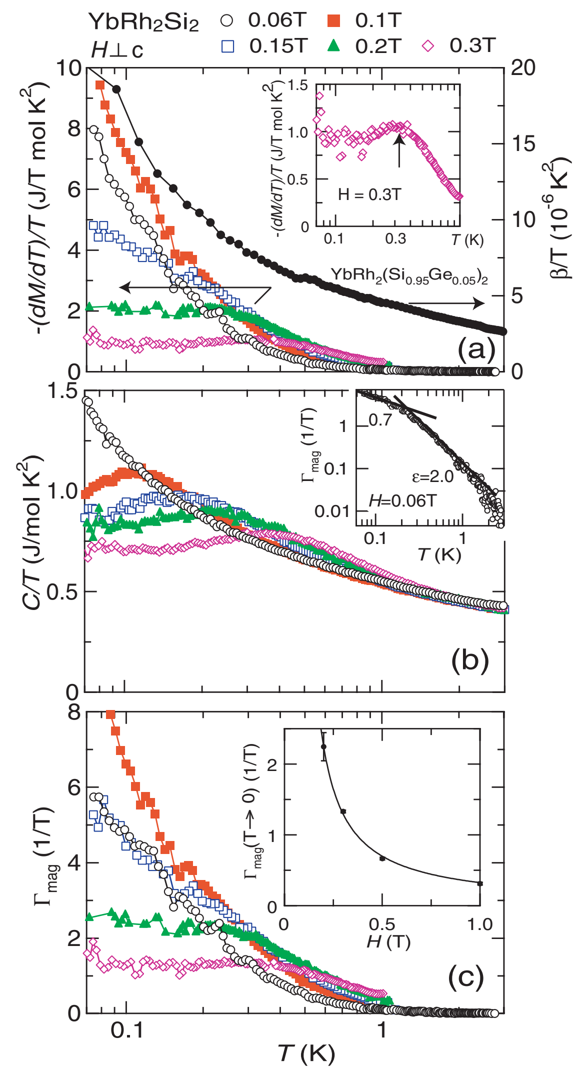

FIG. 2 (color online). (a) Temperature derivative of the magnetization as $-(dM/dT)/T$ (left axis), (b) electronic specific heat as $C(T)/T$, and (c) magnetic Grüneisen ratio $\Gamma_{\rm mag}$ vs temperature (on a logarithmic scale) for $YbRh_2Si_2$ in magnetic fields applied perpendicular to c axis. Volume thermal expansion of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ 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 $H = 0.3$ T. The arrow indicates the position of the maximum. Inset in (b): $\Gamma_{\rm mag}$ vs temperature for $YbRh_2Si_2$ on a log-log plot. The solid lines represent $\Gamma_{\rm mag}(T) \propto T^{-\epsilon}$ with $\epsilon = 0.7$ and 2.0 for low- and high-temperature regions, respectively. Inset in (c): Field dependence of saturated $\Gamma_{\rm mag}$, as $T \rightarrow 0$. The solid line indicates $-G_r(H - H_c^{\rm fit})^{-1}$ with $G_r = -0.3$ and $H_c^{\rm fit} = 0.065$ T.

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 β/T for $YbRh_2(Si_{0.95}Ge_{0.05})_2$ [12] for comparison. The divergence of $-(dM/dT)/T$ is stronger at high temperatures and becomes comparable to β/T at low temperatures. The ratio between β and dM/dT gives the pressure dependence of the critical field which is temperature dependent and decreases with decreasing temperature (not shown). At low temperatures it tends to saturate and the extrapolated value is (0.19 ± 0.05) T/GPa which is consistent with previous hydrostatic pressure experiments [20].

Interestingly, $-(dM/dT)/T$ 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_{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 $Ce_{0.8}Y_{0.2}Cu_2Si_2$ [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 $Ce_{0.8}Y_{0.2}Cu_2Si_2$ [25,26] show that the maximum temperature in $C(T)/T$ increases linearly with increasing field. For the magnetic Grüneisen ratio $\Gamma_{mag}(T)$, the maxima in C/T and $-(dM/dT)/T$ cancel each other, leading to a monotonic crossover from NFL to FL behavior at fields larger than 0.2 T [Fig. 2(c)].

Before discussing details of $\Gamma_{mag}(T)$ data, we first summarize the important theoretical conclusions most related to the present study with respect to the degree of assumptions. (1) Γ_{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 ξ and the correlation time ξ_τ such that the temperature dependence in the quantum critical regime $\Gamma_{mag}(T, H = H_c) \propto T^{-1/\nu z}$ (ν 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., $\Gamma_{mag}(T = 0, H) = -G_r(H - H_c)^{-1}$, with $G_r = \nu(d - z)$ (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 ν, z and d. (3) Within the itinerant theory, the correlation-length exponent $\nu = 1/2$ 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 Γ_{mag} diverges as a function of temperature at $H = H_c = 0.06$ T [Fig. 2(c)] proving the existence of a field-induced QCP (assumption 1). The saturation values of Γ_{mag} for $T \rightarrow 0$ as a function of field, plotted in the inset of Fig. 2(c), diverge like $(H - H_c)^{-1}$ as expected from scaling (assumption 2). The fit reveals $-G_r(H - H_c^{fit})^{-1}$ with $H_c^{fit} = (0.065 \pm 0.004)$ T and $G_r = -0.30 \pm 0.01$. For $YbRh_2(Si_{0.95}Ge_{0.05})_2$, the field dependence of γ 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 ν, 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 as in 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 $H_c^{fit} = 0.065$ T is very close to 0.06 T, supporting the QCP at $H_c = 0.06$ T. Furthermore, the extrapolated values of the saturated Γ_{mag} at $H = 0.1$ T and 0.15 T from $-G_r(H - H_c^{fit})^{-1}$ are 11.8 and 26.6 T^{-1}, respectively, strongly exceeding the largest *measured* values shown in Fig. 2(c). Thus, the lowest temperature in our study (~70 mK) is not low enough to observe saturation of Γ_{mag} at fields very close to the QCP and thereby explains the absence of saturation in $-(dM/dT)/T$ at 0.1 and 0.15 T. As shown in the double-log plot in the inset of Fig. 2(b), $\Gamma_{mag} \propto T^{-\epsilon}$ with $\epsilon \approx 2.0$ from 3 down to 0.3 K. This temperature dependence is much stronger than any expectation within the itinerant theory. At lower temperatures, a crossover to a weaker divergence is found with $\epsilon \approx 0.7$ below 0.25 K, which interestingly is similar to the exponent obtained by the study of the thermal Grüneisen ratio for $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at zero field [12]. The crossover around 0.3 K may be related to the interplay of AF and FM fluctuations, as it is of the same order of magnitude as the absolute value of the Weiss temperature [19]. The fact that no single power-law behavior is found over a larger temperature regime makes the interpretation of the Grüneisen ratio within the quantum critical regime difficult.

At last, we study the evolution of the magnetic entropy upon crossing $T^\star(H)$. For this purpose, it is important to follow the field dependence of dM/dT, which according to the Maxwell relation equals the field dependence of dS/dH. Using isothermal magnetization scans at different temperatures, we calculate the difference in magnetization divided by the temperature increment, $-\Delta M/\Delta T = -\{M(T + \Delta T, H) - M(T - \Delta T, H)\}/2\Delta T$ vs magnetic field, plotted in Fig. 3. For each curve, we have used two

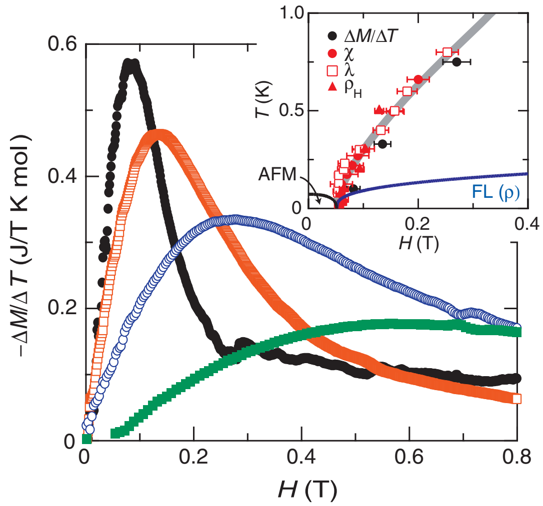

FIG. 3 (color online). Magnetization difference divided by temperature increment $-\Delta M/\Delta T$ (see text) vs magnetic field for $YbRh_2Si_2$ at $T = 0.08$ K (black circles), 0.33 K (red open squares), 0.75 K (blue open circles) and 1.5 K (green squares). Inset: H-T phase diagram of $YbRh_2Si_2$ with the peak positions in $-\Delta M/\Delta T$ (black solid circles) and $T^\star(H)$ (gray solid line) as determined from transport and thermodynamic properties (red or gray symbols) [22,23]. The black and blue or dark gray lines represent the phase boundary of the antiferromagnetic (AFM) ground state and crossover to the Fermi-liquid (FL) regime, respectively [22].

sets of isothermal magnetization $M(H)$ data at temperatures $T - \Delta T$ and $T + \Delta T$. The $-\Delta M/\Delta T(H)$ traces show a peak whose position shifts to larger field with increasing temperature as displayed in the temperature versus field phase diagram in the inset of Fig. 3. The peak positions agree satisfactorily with the energy scale $T^\star(H)$ found previously in the Hall coefficient [23], magnetostriction and susceptibility [22] [cf. red (or gray) symbols in the inset of Fig. 3]. Through $\Delta M/\Delta T \approx dM/dT = dS/dH$, the field dependence of the entropy $S(H)$ could be obtained by integrating $\Delta M/\Delta T$ over the field. Since $dM/dT < 0$ in the entire field range (at all accessible temperatures), the magnetic entropy decreases with increasing field. The positions of the maxima in $-\Delta M/\Delta T$ correspond to inflection points in $S(H)$; i.e., they mark characteristic magnetic fields, at which entropy $S(H)$ at constant temperature is reduced most strongly with increasing field. Remarkably, these inflection points of the entropy agree very well with $T^\star(H)$ determined from Hall effect [23], magnetostriction, and magnetization [22] and thus provide further thermodynamic confirmation of this additional energy scale in $YbRh_2Si_2$.

In conclusion, our study of the low-temperature magnetization and specific heat of $YbRh_2Si_2$ reveals for the first time in any system a divergence of the magnetic Grüneisen ratio $\Gamma_{\mathrm{mag}} = -(dM/dT)/C$ at a magnetic field-tuned QCP. This property provides information on the scaling of the magnetic entropy due to quantum critical fluctuations. The field dependence of the magnetic Grüneisen ratio within the FL regime follows $\Gamma_{\mathrm{mag}}(T \to 0) = -G_r(H - H_c)^{-1}$ [10] with a prefactor $G_r = -0.3 \pm 0.01$ that is consistent with a recent critical Fermi surface model [28]. Within the quantum critical regime, complicated behavior with a crossover scale around 0.3 K is found which may be related to the interplay of AF and FM fluctuations in this system. We also observe an inflection point in the entropy at the scale $T^\star(H)$ previously observed in Hall- and thermodynamic measurements [22,23]. 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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