Magnetic Properties of the Quantum Critical Point in $YbRh_2Si_2$

Philipp Gegenwart, Jeroen Custers, Yoshifumi Tokiwa, Christoph Geibel, Frank Steglich, and Qimiao Si [1]

The ground-state behavior of 4*f*-based heavy fermion (HF) metals is determined by the interplay of two competing interactions which both depend on the strength of the 4*f*-conduction electron hybridization J [1]. Whereas the Kondo interaction leads to a screening of the local moments resulting in a paramagnetic ground state with itinerant 4*f*-electrons, the indirect exchange coupling (RKKY-interaction) can mediate long-range magnetic ordering. By variation of J, these systems can be tuned continuously from the non-magnetic HF state through a magnetic quantum critical point (QCP) into a long-range magnetically ordered state. The important question arises whether the heavy quasiparticles retain their itinerant character and form a spin-density wave (SDW) at the QCP [2,3] or, alternatively, decompose due to the destruction of the Kondo screening [4,5]. In the latter case, the magnetic order should be caused by localized 4*f*-electrons that do not contribute to the Fermi surface.

In recent years, the tetragonal $YbRh_2Si_2$ [6] has become a prototypical system for the study of quantum critical phenomena. It is a HF metal with a characteristic Kondo temperature of T_K = 25 K which shows very weak antiferromagnetic (AF) ordering below T_N = 70 mK. The magnetic order can continuously be suppressed by the application of small magnetic fields, and a field-induced QCP is observed at B_c = 0.06 T and 0.7 T for fields applied perpendicular and parallel to the *c*-axis, respectively [7]. As pressure increases T_N in Yb-based HF systems, a volume expansion weakens the AF ordering. Indeed, the partial substitution of Si- with the isoelectronic but larger Ge-atoms in $YbRh_2(Si_{1-x}Ge_x)_2$ results, for a single crystal with nominal Ge concentration x = 0.05, in T_N = 20 mK and a reduced critical field of 0.027 T (for fields in the tetragonal plane) [8]. It has been shown that magnetic fields are ideally suited for tuning this material as close as possible to the QCP, where the most intense effects of quantum criticality can be studied [7,8]. When approaching the QCP at the critical field by cooling, pronounced non-Fermi liquid behavior is observed: The temperature-dependent part of the electrical resistivity follows a linear dependence, $\Delta\rho \sim T$ [6,7,8] from 10 mK up to 10 K, whereas the Sommerfeld coefficient of the electronic specific heat diverges stronger than logarithmic, i.e., $C_{el}/T \sim T^{\alpha}$ with $\alpha \approx 1/3$ [8] below 0.4 K. This latter behavior is incompatible with the predictions of the itinerant SDW scenario for an AF QCP, regardless of the dimensionality of the fluctuation spectrum. Temperature over magnetic field scaling observed in both properties indicates that the characteristic energy of the heavy quasiparticles vanishes at the QCP [8]. This suggests a destruction of the Kondo resonance in accordance with the locally-critical scenario for the QCP [4,5]. The fractional exponent $\varepsilon \approx 0.7$ of the Grüneisen-ratio divergence, $\Gamma \sim T^{\varepsilon}$, also demonstrates the failure of the itinerant scenario (cf. ref. [9]). Recently, the Hall-effect evolution across the QCP has been studied in great detail at low temperatures [10] (cf. *"Hall-Effect Evolution across a Heavy-Fermion Quantum Critical Point"*). A new line in the temperature-field phase diagram has been discovered across which the isothermal Hall-resistivity as a function of the applied magnetic field changes. Upon decreasing the temperature this feature sharpens, suggesting a sudden change of the Fermi surface at the QCP as $T \rightarrow 0$ [10].

Here, we focus on the magnetic properties close to the QCP in $YbRh_2(Si_{0.95}Ge_{0.05})_2$. Figure 1 displays the temperature dependence of the low-frequency ac susceptibility of the single crystal studied previously by specific heat [8] and thermal-expansion measurements [9]. Its residual resistivity amounts to 5 μΩcm. We first concentrate on the data without superposed constant field B. Upon cooling to below 10 K, a strong increase is observed that, above 0.3 K, can be approximated by a power-law divergence. At lower temperatures, $\chi(T)$ tends to saturate and is well described by a Curie-Weiss (CW) law with a negative Weiss temperature of $\Theta = -0.32$ K. The value of the slope in $\chi^{-1}(T)$ indicates a large effective moment, $\mu_{eff} = 1.4\mu_B$ per Yb^{3+}, and the negative sign of the Weiss temperature suggests some AF correlations. No signature of magnetic ordering is observed because the experiments were performed above 20 mK. Upon superposing constant fields B to the field modula-

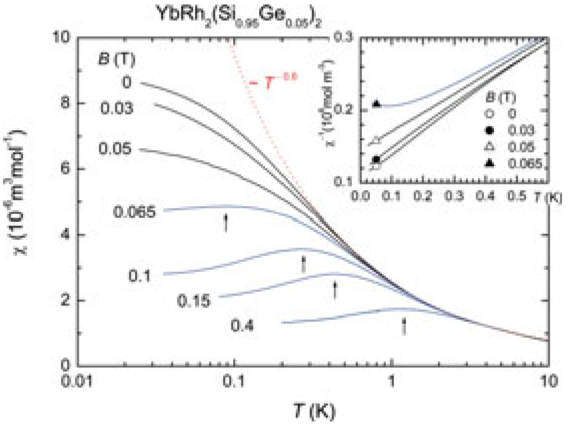

Fig. 1: Low-frequency ac susceptibility χ vs T (on a logarithmic scale) and χ^{-1} vs T (inset) of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at varying superposed static magnetic fields applied perpendicular to the c-axis [13]. Dotted line indicates $(\chi(T)–c) \sim T^{-0.6}$ with $c = 0.215\cdot10^{-6}$ m^3/mol. Arrows indicate susceptibility maxima.

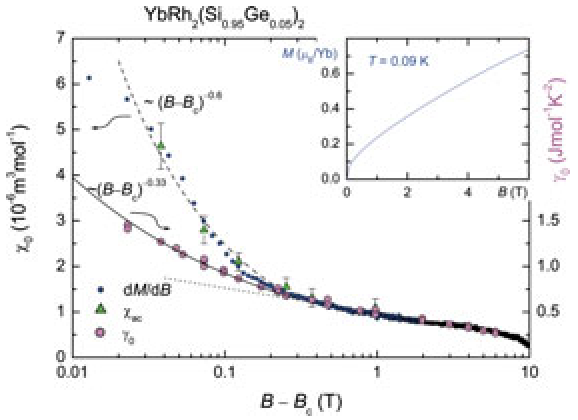

Fig. 2: Field dependence of the Pauli magnetic susceptibility χ_0 determined from the differential susceptibility dM/dB at 0.09 K (solid circles, left axis) and the $T \rightarrow 0$ extrapolation of the ac susceptibility $\chi(T)$ (open triangles, left axis) as well as Sommerfeldt coefficient γ_0 ([8], open circles, right axis). Solid, dashed and dotted lines indicate $\gamma_0 \sim (B–B_c)^{-0.33}$ (B_c =0.027 T), $\chi_0 \sim (B–B_c)^{-0.6}$ (B_c = 0.027 T) and logarithmic behavior, respectively. Inset shows magnetization $M(B)$ at T = 0.09 K.

tion, the low-temperature susceptibility decreases. For small fields the temperature dependence does not change significantly, and the CW law is observed for $B \leq 0.05$ T (see inset). At fields larger than 0.05 T, the behavior changes drastically: Upon cooling, χ(T) passes through a maximum followed by a T^2 dependence at low temperatures, indicating the formation of a field-induced LFL state also observed in specific heat and electrical resistivity measurements [8]. The extrapolated saturation values $\chi_0(B)$, therefore, are interpreted as being an enhanced Pauli susceptibility.

In Figure 2, we show that these values (open triangles) agree well with the slope d$M(B)$/dB (solid circles) of the low-temperature dc magnetization (displayed in the inset). The Sommerfeld coefficient $\gamma_0(B – B_c)$ in the field-induced LFL state at $B > B_c$ has been found to diverge in the approach of the critical field [8], and we now compare its field dependence with that of the Pauli susceptibility. For fields larger than about 0.3 T, both properties show similar field dependences (cf. Figure 2). Below 0.3 T, however, they behave differently, both showing a stronger than logarithmic increase. Whereas $\gamma_0(b) \sim b^{1/3}$ with b the difference between the applied and the critical field, $b = B – B_c$ [8], the Pauli susceptibility can be described by $\chi_0(b) \sim b^{-0.6\pm0.1}$. Note, however, that this power-law divergence, in contrast to that observed for the Sommerfeld coefficient, does not continue towards $b \rightarrow 0$: The CW law observed for fields below 0.05 T with a negative Weiss temperature that does not vanish at the critical field indicates that χ_0 remains finite at the QCP.

Next we compare the evolution of the three characteristic parameters χ_0, γ_0 and A (the coefficient of the T^2 term in the electrical resistivity) of the LFL, induced for $b > 0$ upon tuning the system into the QCP. This provides information on how the heavy quasiparticles decay into the quantum critical state. Figure 3a shows the field dependence of the so-called Kadowaki-Woods ratio, A/γ_0^2 [8]. At larger distances from the QCP, $A/\gamma_0^2 = const$ is observed. The weak divergence for $b \rightarrow 0$ indicates that the characteristic length scale for singular scattering grows much slower than expected by the itinerant spin fluctuation theory [8]. Next we focus on the (dimensionless) Sommerfeld-Wilson ratio $R_W = K\,\chi_0 / \gamma_0$, where $K = \pi^2 k_B^2/(\mu_0\mu_{eff}^2)$ with $\mu_{eff} = 1.4\ \mu_B$ (see above). As shown in Figure 3b, it is b independent in the same field range for which a constant Kadowaki-Woods ratio has been found, and the value of 17.5 $\pm$ 2 is highly enhanced compared to simple metals as well as typical HF metals. This suggests strong ferromagnetic (FM) fluctuations in the approach of the QCP in $YbRh_2Si_2$. We also analyze the field dependence of the ratio A/χ_0^2, which compares the effective quasiparticle-quasiparticle scattering cross section with the Pauli susceptibility. In contrast to both the Kadowaki-Woods and Sommerfeld-Wilson ratio, A/χ_0^2 is approximately constant in the entire field interval above

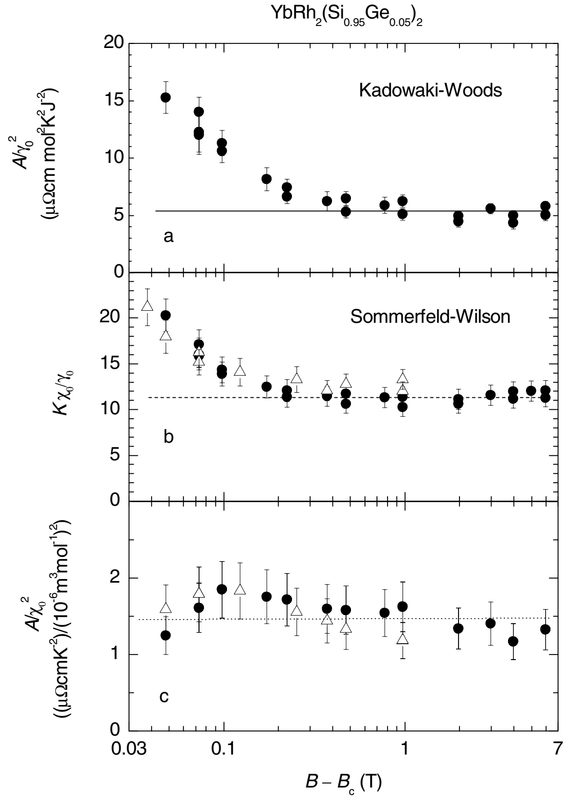

Fig. 3: Field dependence of the Kadowaki-Woods ratio A/γ_0^2 (a), the Sommerfeld-Wilson ratio $R_W = K\ \chi_0 / \gamma_0$ (b) and the ratio A/χ_0^2 (c) of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ [13].

0.065 T. Since the electrical resistivity is influenced most strongly by large-**q** scattering, one would not expect the *A*-coefficient to scale with the **q** = 0 susceptibility in the approach of an *antiferromagnetic* QCP. Furthermore, the ^{29}Si NMR-derived Korringa ratio, $S = 1/T_1K^2$ (with the 4*f*-derived nuclear spin-lattice relaxation rate T_1 and isotropic part of the Knight shift *K*), is very small and amounts to only 10 % of the value expected for a system of non-interacting electrons [12].

These observations raise the question of the role of FM fluctuations in the quantum criticality in $YbRh_2Si_2$ [13]. It may be tempting to consider the **q** = 0 magnetic fluctuations as dominant, especially since an itinerant ferromagnetic QCP would yield a Grüneisen exponent of $1/z\nu = 2/3$, close to what is observed in $YbRh_2Si_2$ [9]. Below we argue that, by contrast, it is more natural to associate the FM fluctuations as one soft part of the overall local quantum criticality.

In $CeCu_{5.9}Au_{0.1}$, the dynamical spin susceptibility at different wave vectors obeys the scaling form $\chi^{-1}(\mathbf{q},T) = \{T^\alpha + [-\Theta(\mathbf{q})]^\alpha\}/c$ with a fractional exponent $\alpha \approx 0.75$ [14]. Figure 4a displays

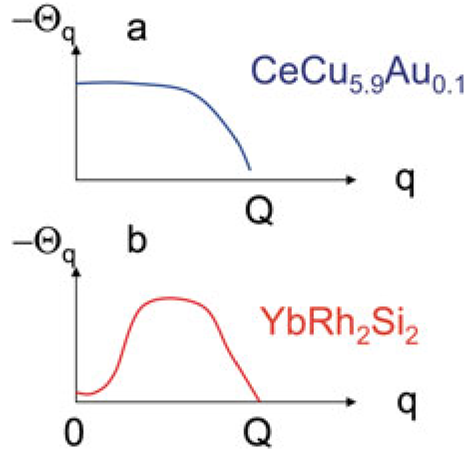

*Fig. 4a: Sketch of the **q** dependence of the Weiss temperature **Θ** as inferred from neutron scattering experiments on $CeCu_{5.9}Au_{0.1}$ [14]. b: the same quantity suggested for $YbRh_2Si_2$.*

schematically the wave vector dependence of the Weiss temperature which vanishes at the critical wave vector **q** = **Q** for this compound [14]. In the following we assume a similar form of the dynamical susceptibility for $YbRh_2Si_2$. Unlike for $CeCu_{5.9}Au_{0.1}$, the **q** = 0 value of the Weiss temperature (≈ – 0.3 K) appears to be very small (cf. Figure 4b). When **q** moves away from either **0** or **Q**, |Θ(**q**)| increases to an order of the RKKY interaction or the bare Kondo scale (~ 25 K [6]). This picture could explain a number of experimental observations. First, for a wide field range larger but not too close to B_c, $\chi(\mathbf{q}{=}0)$ would be roughly proportional to $\chi(\mathbf{Q})$. As finite wave vectors are efficient in affecting the charge transport, a constant $A/\chi(\mathbf{Q})^2$ is plausible. Second, it directly follows that the static susceptibility obeys $\chi^{-1} \sim T^\alpha$ consistent with the experimental observation of $\alpha \approx 0.6$ for $0.3\ K \leq T < 10\ K$ [13], cf. Figure 1. At lower temperatures the data instead follow a CW law, but the smallness of the entropy (≈ 0.03 *R* ln 2) associated with the latter suggests that it is not arising from simple isolated moments but rather is a collective effect, perhaps being associated with the simultaneously small $|\Theta_{\mathbf{q}=0}|$ and $|\Theta_{\mathbf{Q}}|$. Third, since **q** = 0 fluctuations are soft, the enhancement of the Knight shift *K* is considerably stronger than that of $1/T_1$ and the small Korringa ratio $S = 1/T_1K^2$ follows quite naturally. Finally, the observed relation $1/T_1 \approx$ *const* for $YbRh_2Si_2$ [11] would be in disagreement with the expectation for a FM quantum critical point of itinerant type but compatible with the locally critical scenario [5].

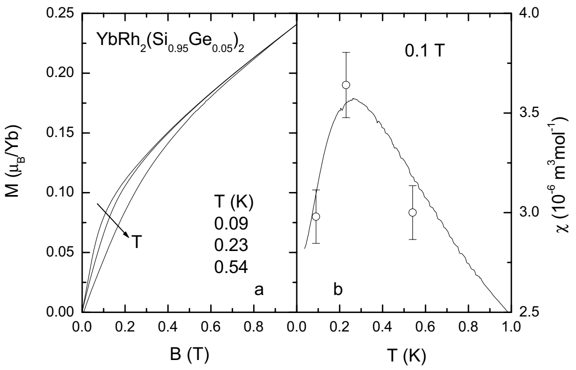

Fig. 5: a: Isothermal dc-magnetization for $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at different temperatures. b: Comparison of the ac-susceptibility $\chi(T)$ at 0.1 T (line) with the differential magnetization dM/dB (open circles), obtained at 0.1 T from the data shown in (a).

At last, we discuss the origin of the characteristic maximum in $\chi(T)$ (cf. Figure 1). As shown in Fig. 5a, the isothermal magnetization $M(B)$ is strongly nonlinear at small fields. The differential susceptibility dM/dB at constant temperature agrees well with the real part of the ac-susceptibility registered continuously at this field (see Fig. 5b). This proves that the susceptibility maximum corresponds to the decrease in slope of the magnetization which indicates a partial (about 0.1 μ_B/Yb) FM polarization of fluctuating moments. These anomalies define a line $T_\chi(B)$ which increases in field with increasing temperature. Most interestingly, $T_\chi(B)$ matches $T_{Hall}(B)$, i.e. the line at which the drastic change of the Hall coefficient has been observed [10] (for comparison with the Hall data taken along the *c*-axis, the field values need to be divided by the anisotropy factor 11). This coincidence raises the important question about the relationship between the FM fluctuations and the Hall-effect signature. Within the above picture of a coexistence of $\mathbf{q} = 0$ and $\mathbf{q} = \mathbf{Q}$ fluctuations (cf. Figure 4), the simultaneous signatures in the Hall coefficient and magnetic susceptibility would be compatible with a locally critical QCP arising from the destruction of the Kondo screening [4,5]. The dramatic change of the Fermi-surface volume naturally results in smeared thermodynamic signatures at finite temperature. The change of the magnetization slope could be one such signature, the very recently observed change in slope of the magnetostriction coefficient [15] at the $T_\chi(B)$ line another one.

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1 Rice University, Houston, USA