Ferromagnetic Quantum Critical Fluctuations in $\mathbf{YbRh_2(Si_{0.95}Ge_{0.05})_2}$

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The bulk magnetic susceptibility $\chi(T, B)$ of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ has been investigated close to the field-induced quantum critical point at $B_c = 0.027$ T. For $B \leq 0.05$ T a Curie-Weiss law with a negative Weiss temperature is observed at temperatures below 0.3 K. Outside this region, the susceptibility indicates ferromagnetic quantum critical fluctuations, $\chi(T) \propto T^{-0.6}$ above 0.3 K. At low temperatures the Pauli susceptibility follows $\chi_0 \propto (B - B_c)^{-0.6}$ and scales with the coefficient of the T^2 term in the electrical resistivity. The Sommerfeld-Wilson ratio is highly enhanced and increases up to 30 close to the critical field.

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Landau's Fermi liquid theory has been successfully used to describe the low-temperature behavior of strongly correlated electron systems. Starting from a Fermi gas, this model introduces the many-body interactions in a phenomenological way. It is based on the concept of elementary excitations, called quasiparticles, showing a one-to-one correspondence to the free electron (or hole) excitations of the Fermi gas. Furthermore, the quasiparticle motion can be described by a generalized Boltzmann equation. The quasiparticle excitations thus lead to a linear in temperature (T) specific heat, $C = \gamma_0 T$, and a constant Pauli susceptibility χ_0 at low temperatures as well as a temperature independent rate $\propto A$ of quasiparticle-quasiparticle collisions causing an electrical resistivity contribution $\Delta\rho = AT^2$. The electronic correlations result in a renormalization of the effective mass of the quasiparticles, which in the case of the heavy fermion (HF) systems can exceed the bare electron mass by a factor up to 1000. This causes huge values of γ_0, χ_0, and A that roughly scale as $\gamma_0 \propto \chi_0 \propto \sqrt{A}$. Recently, much interest has been focused on how the properties of the heavy Landau Fermi liquid (LFL) state evolve if these materials are tuned into a long-range magnetically ordered state [1]. The important question arises whether the heavy quasiparticles retain their itinerant character and form a spin-density wave (SDW) at the quantum critical point (QCP) or, alternatively, decompose due to the destruction of the Kondo screening. In the latter case, the magnetic order is caused by localized f electrons that do not contribute to the Fermi surface [2]. In order to address this question, detailed experiments on the nature of the quantum critical state in the two prototypical materials $CeCu_{5.9}Au_{0.1}$ [3] and $YbRh_2Si_2$ [4] have been performed.

In $CeCu_{5.9}Au_{0.1}$ the static susceptibility has been found to obey a modified Curie-Weiss (CW) law [5],

$$\chi^{-1}(\mathbf{q}, T) = \{T^\alpha + [-\Theta(\mathbf{q})]^\alpha\}/c, \qquad (1)$$

with a fractional exponent $\alpha \simeq 0.75$. The Weiss temperature $\Theta(\mathbf{q}) < 0$ is a function of $\mathbf{q}$ and vanishes at the critical wave vector $\mathbf{q} = \mathbf{Q}$ of the nearby antiferromagnetic (AFM) order [5]. Furthermore, the dynamical susceptibility follows energy over temperature scaling and the bulk ($\mathbf{q} = 0$) susceptibility obeys magnetic field over temperature scaling, both with the same fractional exponent α obtained from the modified CW law [5]. The momentum independence in the critical response observed in these experiments led to the proposal of a *locally critical* scenario for the HF QCP [6].

$YbRh_2Si_2$ is a clean and stoichiometric HF system located extremely close to the border of long-range magnetic order and shows pronounced non-Fermi liquid behavior in thermodynamic, electrical transport, and magnetic properties [4,7–11]. Very weak AFM ordering at $T_N = 70$ mK can be driven to zero by a small critical magnetic field B_c of 0.06 T applied in the easy magnetic plane perpendicular to the crystallographic c axis [8]. In $YbRh_2(Si_{0.95}Ge_{0.05})_2$ the partial substitution of Si atoms with the larger but isoelectronic Ge reduces T_N and B_c far closer towards zero (20 mK and 0.027 T, see Fig. 4). The observed divergences of both the quasiparticle mass and Grüneisen ratio [7,9] exclude the SDW description of the QCP in this system. Temperature over magnetic field scaling in thermodynamic and transport properties indicates that the characteristic energy of the heavy quasiparticles is governed only by the ratio of the thermal energy to the magnetic field difference $b = B - B_c$ and vanishes at $b \rightarrow 0$ [7,11]. The observed disparity in the temperature dependence of the electrical resistivity and specific heat at $b = 0$ suggests a breakup of the heavy quasiparticles in the approach of the QCP [7]. Furthermore, the observation of the Yb^{3+} electron spin resonance at temperatures at least down to 2 K, i.e., well below the single-ion Kondo scale of 25 K in that system, has been ascribed to the emergence of large unscreened local magnetic moments close to the QCP [12].

In this Letter, we use low-temperature measurements of the bulk magnetic susceptibility $\chi(T, B)$ to investigate quantum critical behavior in $YbRh_2(Si_{0.95}Ge_{0.05})_2$. Our results prove that the quantum critical fluctuations in this system have a very strong ferromagnetic (FM) component and thus are unique among all other quantum critical HF systems, including $CeCu_{5.9}Au_{0.1}$.

The measurements were performed on pieces of a high-quality single crystal of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ studied previously by specific heat and electrical resistivity [7], as well as thermal expansion measurements [9]. The residual resistivity of the crystals amounts to 5 $\mu\Omega$ cm. We obtain the magnetic susceptibility $\chi(T, B)$ from either low-temperature ac-susceptibility or dc-magnetization measurements. The ac susceptibility was determined with a low-frequency (16.67 Hz) field modulation of 0.1 mT. Constant fields B have been superposed to the modulation field using a superconducting 20 T magnet. A $B = 0$ study has already been published in [13]. The dc-magnetization measurements were performed utilizing a high-resolution Faraday magnetometer.

Figure 1 displays the temperature dependence of the magnetic ac susceptibility of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at different fields B, applied in the easy magnetic plane perpendicular to the c axis. We first concentrate on the $B = 0$ data. Upon cooling to below 10 K, a strong increase is observed that, above 0.3 K, can be approximated by a power-law divergence $\Delta\chi \propto T^{-0.6}$. Here $\Delta\chi$ is the susceptibility after subtraction of a small temperature independent contribution that amounts to 2% of the total susceptibility at 0.02 K. The previous attempt [13] to fit the data with an exponent of 0.75 is much less satisfactory. At lower temperatures, $\chi(T)$ tends to saturation and is well described by a CW law with a negative Weiss temperature of $\Theta = -0.32$ K similar to that found for pure $YbRh_2Si_2$ at $T_N < T \leq 0.3$ K [7]. The value of the slope in $\chi^{-1}(T)$ indicates a large effective moment $\mu_{\mathrm{eff}} \approx 1.4\mu_B$ per Yb^{3+}, and the sign of the Weiss temperature suggests some AFM correlations [7]. Note that the temperature dependence both above and below 0.3 K is different from that found in the bulk susceptibility of $CeCu_{5.9}Au_{0.1}$ [cf. Eq. (1)]. No signature of magnetic ordering is observed because the experiments have been performed above 20 mK. Upon superposing constant fields B to the field modulation, 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, $\chi(T)$ passes through a maximum followed by a T^2 dependence at low temperatures, indicating the formation of a field-induced LFL state [14] also observed in specific heat and electrical resistivity measurements [7]. The extrapolated saturation values $\chi_0(B)$ therefore represent the Pauli susceptibility.

Next, we focus on the field dependence of $\chi_0(B)$ in the approach of the QCP at $B_c = 0.027$ T. In Fig. 2, we show that the Pauli susceptibility, determined as discussed above from the saturation values of isofield ac-susceptibility measurements (open triangles), agrees well with the slope $dM(B)/dB|_{T=\mathrm{const}}$ (solid circles) of the low-temperature dc magnetization (see inset). The specific heat coefficient in the field-induced LFL state at $B > B_c$ has been found to diverge in the approach of the critical field [7] and we now compare its field dependence with that of the Pauli susceptibility. For fields larger than about 0.3 T, both properties show a very similar field dependence (cf. Fig. 2). Below 0.3 T, they deviate from each other, both showing a stronger than logarithmic increase. Whereas $\gamma(b) \propto b^{1/3}$, with b the difference between the applied and the critical field, $b = B - B_c$ [7], the Pauli susceptibility can be de-

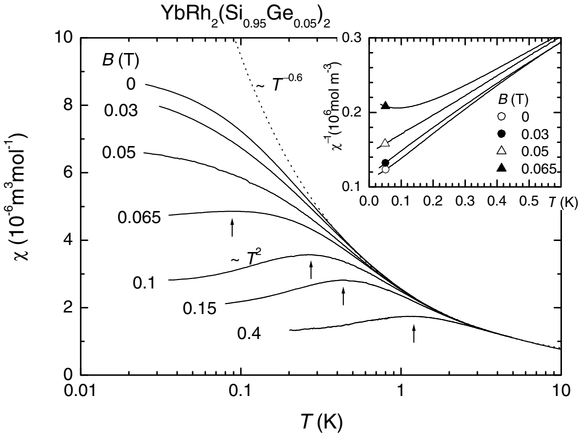

FIG. 1. Low-frequency ac susceptibility χ vs T (on a logarithmic scale) and χ^{-1} vs T (inset) of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ at varying superposed static magnetic fields applied perpendicular to the c axis. The dotted line indicates $(\chi(T) - c) \propto T^{-0.6}$, with $c = 0.215 \times 10^{-6}$ m^3 mol^{-1}. Arrows indicate susceptibility maxima.

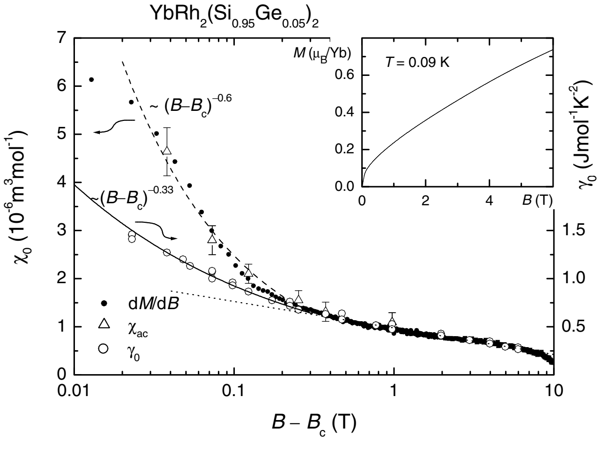

FIG. 2. Field dependence of the Pauli magnetic susceptibility χ_0 determined from the differential susceptibility dM/dB at 0.09 K (solid circles, left axis) and $T \rightarrow 0$ extrapolation of the ac susceptibility $\chi(T)$ (open triangles, left axis) as well as specific heat coefficient γ_0 ([7], open circles, right axis). Solid, dashed, and dotted lines indicate $\gamma_0 \propto (B - B_c)^{-0.33}$ ($B_c = 0.027$ T), $\chi_0 \propto (B - B_c)^{-0.6}$, and logarithmic behavior, respectively. Inset shows magnetization $M(B)$ at $T = 0.09$ K.

scribed by $\chi_0(b) \propto b^{-0.6\pm0.1}$. Note, however, that this power-law divergence, in contrast to that observed for the specific heat 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 $\chi(T \rightarrow 0)$ remains finite at the QCP.

Having determined the field dependence of the Pauli susceptibility, we may now 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 3(a) shows the field dependence of the Kadowaki-Woods ratio [7]. At larger distances from the QCP, $A/\gamma_0^2 = \text{const}$ 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 [7].

Next we focus on the Sommerfeld-Wilson ratio $R_W = K\chi_0/\gamma_0$, where $K = \pi^2 k_B^2/(\mu_0 \mu_{\text{eff}}^2)$ is a scaling factor which gives a dimensionless value of $R_W = 1$ for the free electron gas. Whereas electron-phonon interactions enhance γ_0 but not χ_0, leading to a reduction of R_W, an enhancement of R_W could be caused by electronic spin-spin interactions. For Kondo systems, a Sommerfeld-Wilson ratio of 2 is expected [15] as observed in many HF systems [16]. Nearly FM metals, due to Stoner enhancement, show very large values, e.g., $R_W = 6$–8 (Pd), 12 (TiBe_2), 40 (Ni_3Ga) [17], and 10 ($\text{Sr}_3\text{Ru}_3\text{O}_7$ [18]). For $\text{YbRh}_2(\text{Si}_{0.95}\text{Ge}_{0.05})_2$, as shown in Fig. 3(b), the Sommerfeld-Wilson ratio is b independent in the same field range for which a constant Kadowaki-Woods ratio has been found. The value of $R_W = 17.5 \pm 2$ is highly enhanced compared to all other HF systems. Upon lowering the magnetic field deviation from the QCP, R_W even increases, reaching a value larger than 30 at 0.065 T, which is the lowest field at which χ_0 could be determined (see above). This dramatic increase of R_W highlights the importance of FM fluctuations in the approach of the QCP.

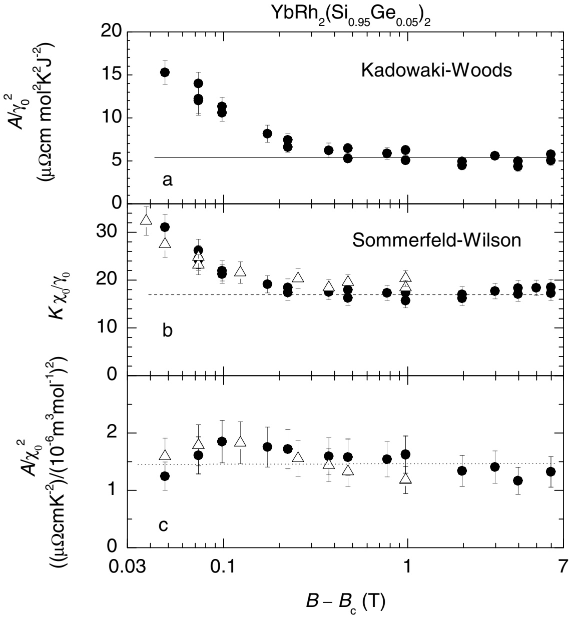

FIG. 3. Field dependence of the Kadowaki-Woods ratio A/γ_0^2 (a), the Sommerfeld-Wilson ratio $R_W = K\chi_0/\gamma_0$ (b), and the ratio A/χ_0^2 (c) of $\text{YbRh}_2(\text{Si}_{0.95}\text{Ge}_{0.05})_2$, using $\gamma_0(B)$ and $A(B)$ from Ref. [7]. Solid (open) symbols in (b) and (c) indicate data calculated by using the Pauli susceptibility χ_0 obtained from the differential susceptibility dM/dB at 90 mK and the $T \rightarrow 0$ extrapolation of the ac susceptibility (cf. Fig. 2), respectively. Solid, dashed, and dotted lines indicate $A/\gamma_0^2 = 5.3\ \mu\Omega\,\text{cm}\,\text{mol}^2\,\text{K}^2\,\text{J}^{-2}$, $R_W = 17.5$, and $A/\chi_0^2 = 1.45 \times 10^{12}\ \mu\Omega\,\text{cm}\,\text{K}^{-2}/(\text{m}^3/\text{mol})^2$, respectively.

In Fig. 3(c), the field dependence of the ratio A/χ_0^2, which compares the quasiparticle-quasiparticle scattering cross section with the Pauli susceptibility, is shown. In contrast to both the Kadowaki-Woods and Sommerfeld-Wilson ratio, A/χ_0^2 is approximately constant in the entire field interval above 0.065 T. Since the electrical resistivity is most strongly influenced by large-**q** scattering [19], one would not expect the A coefficient to scale with the $\mathbf{q} = 0$ susceptibility in the approach of an *antiferromagnetic* QCP. The fact that $A/\chi_0^2 \approx \text{const}$ over more than two decades in the field deviation from the QCP thus provides evidence for FM ($\mathbf{q} = 0$) quantum critical fluctuations in $\text{YbRh}_2(\text{Si}_{0.95}\text{Ge}_{0.05})_2$.

Figure 4 shows the temperature-field diagram for $\text{YbRh}_2(\text{Si}_{0.95}\text{Ge}_{0.05})_2$ including regimes of different magnetic response. The AFM state close to the origin is surrounded by a regime below 0.3 K that extends to fields up to 0.05 T (shaded area), in which the susceptibility follows a CW law with a negative Weiss temperature, indicating predominant AFM correlations. Outside this region, the quantum critical behavior is dominated by FM fluctuations: (i) $\chi_0(b)$ follows a $b^{-0.6}$ dependence and (ii) the temperature dependent part, $\Delta\chi(T)$, diverges as $T^{-0.6}$ for $T > 0.3$ K, suggesting a divergent $\mathbf{q} = 0$ susceptibility (see also inset b of Fig. 4). A similar temperature dependence has been observed in the ^{29}Si NMR-derived Knight shift $K_s(T, B)$ of YbRh_2Si_2 [10]. In these experiments, outside a narrow region close to the critical field, the Korringa ratio $(1/T_1T)/K_s^2$ with K_s and $1/(T_1T)$, being proportional to the bulk susceptibility and the **q**-averaged dynamical spin susceptibility, respectively, is constant, with a value similar as found for nearly ferromagnetic metals [10]. This suggests that the inverse of the zero-field susceptibility, plotted versus $T^{0.6}$ in inset b of Fig. 4, is effectively **q** independent above 0.3 K. Such behavior is even "more local" than that described in the *locally critical* scenario [6] and very different from the case of $\text{CeCu}_{5.9}\text{Au}_{0.1}$ [cf. Eq. (1)] for which in the latter system the Weiss temperature is strongly **q** dependent and van-

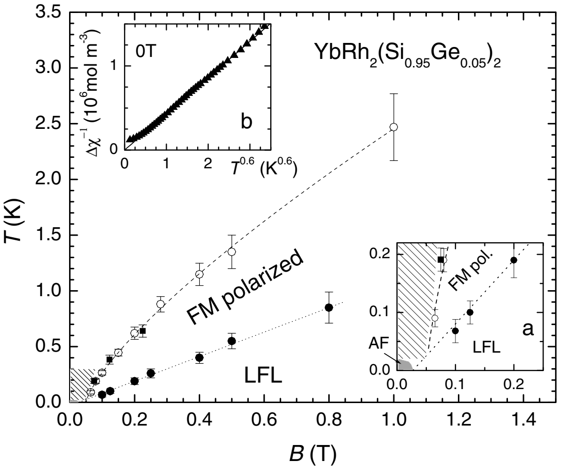

FIG. 4. T-B phase diagram for $\mathrm{YbRh_2(Si_{0.95}Ge_{0.05})_2}$, $B \perp c$. Open and closed circles indicate temperatures of maxima in $\chi(T)$ (cf. arrows in Fig. 1) and $C(T)/T$ [7] at various different fields, respectively. Positions of $\chi(T)$ maxima for $\mathrm{YbRh_2Si_2}$ [4] are indicated by small solid squares. Dashed and dotted lines represent $2.55\ \mathrm{K\,T^{0.75}} \times (B - 0.05\ \mathrm{T})^{0.75}$ and $1.1\ \mathrm{K\,T^{-1}} \times (B - 0.027\ \mathrm{T})$, respectively. The AFM state is marked by gray solid region very close to the origin. Slanted lines indicate the regime where $\chi(T)$ follows the Curie-Weiss law. For labels, see text. Inset (a) enlarges the region close to the origin. Inset (b) displays susceptibility increment as $\Delta\chi^{-1}$ vs $T^{0.6}$ with $\Delta\chi = \chi(T) - 0.215 \times 10^{-6}\ \mathrm{m^3mol^{-1}}$. Solid line indicates $\Delta\chi^{-1} \propto T^{0.6}$.

ishes only for $\mathbf{q} = \mathbf{Q}$, i.e., at the critical antiferromagnetic wave vector [5].

Finally, we discuss the characteristic maximum in $\chi(T)$ whose position shifts to lower temperatures with decreasing magnetic field extrapolating towards $B^\star \approx 0.05$ T (see dashed line in Fig. 4). Very similar behavior is observed for pure $\mathrm{YbRh_2Si_2}$ as well [4], the positions of the susceptibility maxima not being affected by the Ge substitution (cf. open circles and solid squares in Fig. 4). These positions of susceptibility maxima define a line in the temperature-field plane along which the magnetization slope dM/dB is most sensitive to a change of the applied magnetic field. This characteristic field increases with temperature as expected for a FM polarization of fluctuating magnetic moments. Most interestingly, the *ferromagnetic* fluctuations are unaffected by the Ge substitution in $\mathrm{YbRh_2(Si_{0.95}Ge_{0.05})_2}$ that has strong influence on the *antiferromagnetic* order, leading to a roughly threefold reduction of the ordering temperature, ordered moment [11], and critical magnetic field compared to the parent compound $\mathrm{YbRh_2Si_2}$. This suggests the FM fluctuations to be not directly correlated to the AFM-QCP at $B_c = 0.027$ T in the Ge-substituted system. Indeed, the line of susceptibility maxima is different from the crossover line determined from the maxima in the specific heat coefficient $C(T)/T$ that terminates at $B = B_c = 0.027$ T, i.e., $\approx \frac{1}{2}B^\star$ ([7], see solid circles in Fig. 4).

To summarize, the strong increase of the bulk susceptibility towards low temperature, the highly enhanced Sommerfeld-Wilson ratio, and the field independence of A/χ_0^2 indicate that $\mathrm{YbRh_2(Si_{0.95}Ge_{0.05})_2}$ is located very close to a FM instability. Recent experiments on itinerant ferromagnets have revealed a first-order instead of a continuous suppression of the ordering [20]. It has also been argued that close to a FM QCP a nonanalytic term in the free energy generates first-order behavior [21]. In $4f$-based heavy fermion systems, no evidence for a FM QCP has yet been found; instead, these systems first undergo a transition to an AFM state before getting paramagnetic [22,23]. $\mathrm{YbRh_2(Si_{0.95}Ge_{0.05})_2}$ is thus unique as the quantum critical behavior is dominated by FM fluctuations over wide ranges of the T-B plane, except for fields close to the critical field and temperatures below 0.3 K.

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[1] G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001).
[2] P. Coleman *et al.*, J. Phys. Condens. Matter **13**, R723 (2001).
[3] H. v. Löhneysen *et al.*, Phys. Rev. Lett. **72**, 3262 (1994).
[4] O. Trovarelli *et al.*, Phys. Rev. Lett. **85**, 626 (2000).
[5] A. Schröder *et al.*, Nature (London) **407**, 351 (2000).
[6] Q. Si *et al.*, Nature (London) **413**, 804 (2001).
[7] J. Custers *et al.*, Nature (London) **424**, 524 (2003).
[8] P. Gegenwart *et al.*, Phys. Rev. Lett. **89**, 056402 (2002).
[9] R. Küchler *et al.*, Phys. Rev. Lett. **91**, 066405 (2003).
[10] K. Ishida *et al.*, Phys. Rev. Lett. **89**, 107202 (2002).
[11] P. Gegenwart *et al.*, cond-mat/0406260.
[12] J. Sichelschmidt *et al.*, Phys. Rev. Lett. **91**, 156401 (2003).
[13] P. Gegenwart *et al.*, Acta Phys. Pol. B **34**, 323 (2003).
[14] As discussed in [11], the temperature dependent part of the susceptibility varies as $T^2 b^{-7/3}$ in the field-induced LFL, obeys T/b scaling ($b = B - B_c$), and is consistent with the field dependence of the magnetic entropy.
[15] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, England, 1993).
[16] Z. Fisk, H. R. Ott, and G. Aeppli, Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 1882 (1987).
[17] S. R. Julian *et al.*, Physica (Amsterdam) **259B–261B**, 928 (1999), and references therein.
[18] S.-I. Ikeda *et al.*, Phys. Rev. B **62**, R6089 (2000).
[19] See, e.g., K. Ueda, J. Phys. Soc. Jpn. **43**, 1497 (1977).
[20] M. Uhlarz, C. Pfleiderer, and S. M. Hayden, Phys. Rev. Lett. **93**, 256404 (2004), and references therein.
[21] D. Belitz and T. R. Kirkpatrick, Phys. Rev. Lett. **89**, 247202 (2002).
[22] S. Süllow *et al.*, Phys. Rev. Lett. **82**, 2963 (1999).
[23] N. Neemann *et al.*, Acta Phys. Pol. B **34**, 1085 (2003).