# Dimensional Crossover of Quantum Critical Behavior in $CeCoIn_5$

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The nature of quantum criticality in $CeCoIn_5$ is studied by low-temperature thermal expansion $\alpha(T)$. At the field-induced quantum critical point at $H = 5$ T a crossover scale $T^\star \approx 0.3$ K is observed, separating $\alpha(T)/T \propto T^{-1}$ from a weaker $T^{-1/2}$ divergence. We ascribe this change to a crossover in the dimensionality of the critical fluctuations which may be coupled to a change from unconventional to conventional quantum criticality. Disorder, whose effect on quantum criticality is studied in $CeCoIn_{5-x}Sn_x$ ($0 \le x \le 0.18$), shifts $T^\star$ towards higher temperatures.

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Quantum criticality in heavy fermion (HF) systems continues to attract interest due to the occurrence of highly anomalous metallic states with severe deviations from Landau Fermi liquid (LFL) behavior [1,2] and the emergence of unconventional superconductivity in close vicinity to antiferromagnetic (AF) quantum critical points (QCPs) [3]. Neither the nature of the non-Fermi liquid (NFL) normal state related to quantum criticality, nor the superconducting (SC) pairing mechanism has been clarified up to now. It is thus of great interest to investigate whether quantum criticality in these systems can be described by conventional theory within the framework of a spin-density-wave (SDW) instability [4,5], or whether unconventional scenarios in which the $f$ electrons localize at the magnetic QCP due to a destruction of the Kondo resonance [6–8] may be more appropriate. For the formation of the latter, magnetic frustration leading to a reduced dimensionality of the critical fluctuations may be crucial.

The $CeMIn_5$ ($M$ = Rh, Ir, Co) systems are prototypical as they display a generic phase diagram with unconventional HF superconductivity in close vicinity to an AF QCP [9]. They crystallize in a tetragonal structure which can be viewed as an alternating series of $CeIn_3$ and $MIn_2$ layers. As a result of the layered crystal structure, the Fermi surface displays a strongly two-dimensional (2D) character with cylindrical sheets along the crystallographic $c$ axis [10]. Compared with cubic $CeIn_3$, a HF superconductor with $T_c = 0.2$ K [3] in a very narrow pressure range close to the magnetic quantum phase transition at $p_c \approx 2.6$ GPa, SC transition temperatures of about 2 K are observed over wide pressure ranges for the tetragonal $CeRhIn_5$ (at $p \ge 1.6$ GPa) and $CeCoIn_5$ (at ambient pressure) [11,12]. This $T_c$ enhancement has been attributed to the layered crystal structure and, relatedly, strongly anisotropic magnetic fluctuations [12]. Indeed the nuclear magnetic relaxation rate $1/T_1$ of $CeCoIn_5$ displays a weak $T^{1/4}$ dependence in the normal state between 2 and 40 K which signals strongly anisotropic quantum critical fluctuations [13].

The aim of this Letter is a detailed investigation of the nature of quantum criticality in $CeCoIn_5$. We focus, in particular, to the region very close to the upper critical field $H_{c2}$ for superconductivity (5 T for $H \parallel c$ cf. Fig. 1) which previously has been studied by heat and charge transport [14–16] and specific heat measurements [17]. Diverging coefficients of the $T^2$ contributions to the electrical and the thermal resistivity prove the existence of a magnetic-field-induced QCP at 5 T. NFL behavior in the temperature dependence of the electronic specific heat coefficient at 5 T has been described in the frame of the SDW theory [17]. However, below 0.3 K a large nuclear contribution arising from the Zeeman splitting of In-nuclear moments needs to be subtracted. Therefore, the data do not allow to distinguish between a saturation or

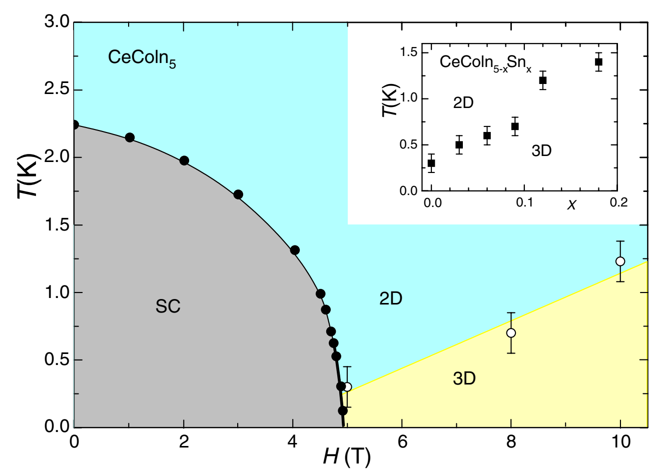


FIG. 1 (color online). Phase diagram of $CeCoIn_5$ for $H \parallel c$ as determined from thermal expansion. Superconducting phase in gray with first-order boundary below 0.7 K indicated by thick black line. Regions where thermal expansion follows 2D and 3D quantum critical behavior are marked in blue (lighter gray) and yellow (lightest gray), respectively. The inset displays the evolution of the crossover with Sn-doping in $CeCoIn_{5-x}Sn_x$ at the respective $H_{c2}(x)$.

 

logarithmic divergence at lowest temperatures and thus further *thermodynamic* measurements are needed to determine the nature of quantum criticality in the system (transport data will be discussed later).

Thermal expansion is ideally suited for this purpose. It probes the pressure dependence of the entropy which close to QCPs is accumulated at finite temperatures. Scaling arguments have revealed that thermal expansion $\alpha(T)$ is far more singular than specific heat $C(T)$ in the approach of any pressure-sensitive QCP [18]. Within the SDW theory the leading contribution to $\alpha(T)/T$ diverges like $T^{-1/2}$ and $T^{-1}$ for 3D and 2D AF QCPs, respectively [18]. Both can easily be distinguished from $\alpha(T)/T = \text{const}$ expected for a LFL. Especially important in this context, thermal expansion, in contrast to specific heat, is not affected by nuclear hyperfine contributions.

For our study, we have used high-quality single crystals of $CeCoIn_{5-x}Sn_x$ grown from In flux, whose low-temperature specific heat and electrical resistivity are discussed in [17,19,20]. For details on the sample characterization see [19–21]. Thermal expansion has been determined with the aid of high-resolution dilatometers at temperatures down to 0.04 K and in magnetic fields up to 10 T. We have measured the length change $\Delta L_c$ along the $c$ axis and determined the linear ($c$ axis) expansion coefficient $\alpha = \partial \ln L_c / \partial T$.

Figure 2 displays our thermal expansion data on undoped $CeCoIn_5$. At the upper critical field of 5 T, the thermal expansion coefficient $\alpha(T)/T$ [cf. inset (a)] grows much stronger upon cooling than the respective specific heat coefficient which diverges only logarithmically [17]. Over more than one decade in temperature, i.e., for $0.3\ \text{K} \leq T \leq 6\ \text{K}$, the data follow a $1/T$ divergence which hints at 2D AF quantum critical fluctuations [18]. The latter may result from the layered crystal structure [12,13]. At $T^\star \approx 0.3$ K, the temperature dependence changes to $\alpha \propto T^{1/2}$ [see main part and inset (b), which also displays data obtained on a second sample down to 40 mK]. Note, that $\alpha/T$ does not show a saturation excluding the formation of a LFL above the lowest measured temperature. The square-root behavior for $\alpha(T)$ is compatible with a 3D AF QCP of itinerant nature [18] as observed for $CeNi_2Ge_2$ [2] and $CeIn_{3-x}Sn_x$ [22].

As $H$ is increased above 5 T, $T^\star$ increases and $\alpha(T)$ becomes less singular; i.e., the coefficient of the square-root contribution decreases [cf. Fig. 2, inset (b)]. This suggests that the system is tuned away from the QCP, compatible with previous studies [14,17], although LFL behavior is not yet fully established in thermal expansion.

Our data on $CeCoIn_5$ are summarized in the main part of Fig. 1. We have observed a crossover scale separating 2D from 3D quantum critical behavior. To provide further evidence for this crossover and to investigate how it is influenced by weak disorder, we now focus on the series $CeCoIn_{5-x}Sn_x$ where the Sn atoms preferentially occupy the In-1 position within in the tetragonal plane [21]. Sn doping weakens superconductivity, leading to a linear suppression of $T_c$ towards zero for $x = 0.18$ [19]. The temperature-magnetic-field phase diagram of various $CeCoIn_{5-x}Sn_x$ single crystals has previously been studied by low-temperature electrical resistivity and specific heat measurements [19,20]. As $T_c$ is reduced, a corresponding reduction of $H_{c2}$ is observed (for the $x$ dependence of $T_c$ and $H_{c2}$, see Table I). For all different Sn concentrations the temperature dependence of the specific heat displays NFL behavior at the respective upper critical field and the formation of a LFL state at fields exceeding $H_{c2}(x)$ [19]. This suggests that field-induced quantum criticality is always pinned at the upper critical field $H_{c2}$ when the latter is reduced by Sn doping. Furthermore, the low-$T$ specific

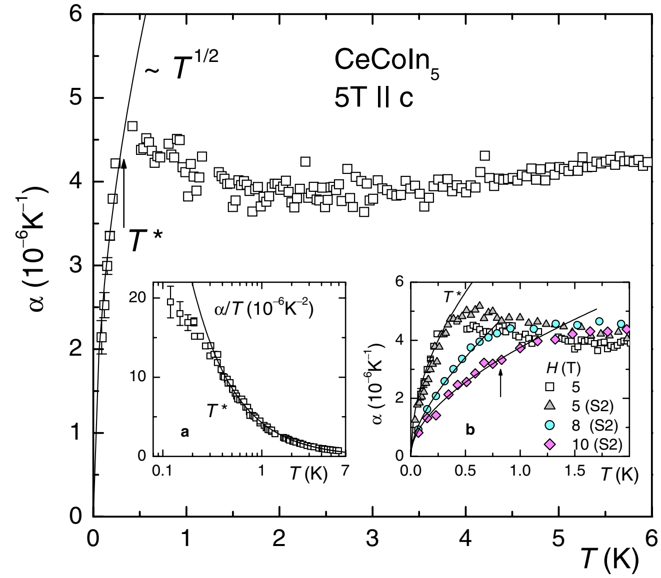


FIG. 2 (color online). Temperature dependence of the linear thermal expansion coefficient of $CeCoIn_5$ at $H = 5$ T ($\parallel c$). The line and arrow indicate $\alpha \propto \sqrt{T}$ and crossover temperature $T^\star$, defined as an upper limit for this $T$ dependence, respectively. Inset (a) displays data from the main part as $\alpha/T$ vs $T$ (on a logarithmic scale). The line indicates $T^{-1}$ dependence. Inset (b) compares data from the main part in the low-temperature regime with 5, 8, and 10 T data obtained from a second sample (S2). Lines display square-root behavior.

TABLE I. Values for the SC transition temperature $T_c$ and upper critical magnetic field $H_{c2}$ for $CeCoIn_{5-x}Sn_x$ [19].

| $x$ | $T_c$ (K) | $H_{c2}$ (T) |
|---|---|---|
| 0.00 | (2.25 ± 0.05) | (4.9 ± 0.1) |
| 0.03 | (1.80 ± 0.05) | (4.5 ± 0.1) |
| 0.06 | (1.50 ± 0.05) | (3.9 ± 0.1) |
| 0.09 | (1.15 ± 0.05) | (3.4 ± 0.1) |
| 0.12 | (0.75 ± 0.05) | (2.5 ± 0.1) |
| 0.18 | 0 | 0 |

heat coefficient at $H = H_{c2}(x)$ remains unchanged within the scatter of the data for $0 \leq x \leq 0.12$ [19]. On the other hand, the residual resistivity $\rho_0$ shows a tenfold increase for $x$ ranging from 0 to 0.18, indicating the effect of disorder scattering due to the random distribution of Sn atoms on the in-plane In site [20]. The study of $CeCoIn_{5-x}Sn_x$ thus allows us to systematically investigate the disorder dependence of NFL behavior without tuning the system away from the QCP.

Figure 3 shows $c$-axis thermal expansion data for the various studied $CeCoIn_{5-x}Sn_x$ single crystals at their respective upper critical magnetic fields (for the zero-field data see [23]). In all these samples, 2D-like quantum critical behavior $\alpha(T)/T \propto T^{-1}$ is found from 6 K down to a lower bound which increases from about 0.3 K for $x = 0$ to about 1.4 K for $x = 0.18$.

Like for $x = 0$, the low-$T$ thermal expansion of all samples studied is well described by $\alpha(T) \propto \sqrt{T}$. For $x = 0.18$, this temperature dependence holds up to $T^\star \approx 1.4$ K, i.e., over more than one decade (see Fig. 4), providing clear evidence for 3D AF quantum critical fluctuations in the latter system. Clearly, the temperature at which the dimensional crossover occurs is shifted with Sn doping in $CeCoIn_{5-x}Sn_x$ to values above 1 K; cf. the inset of Fig. 1. As stated above, the partial substitution of the In-1 site by Sn atoms enhances impurity scattering without tuning the system away from the QCP. Our observation of a shift of the crossover scale $T^\star$ with $x$ is then naturally attributed to the effect of isotropic impurity scattering, which "smears out" the magnetic anisotropy. Crossovers have also been observed in the electrical and heat transport [15,16] as well as in the Hall coefficient [24] for the current direction $j \perp c$. However, transport experiments are influenced by electronic relaxational properties, which can give rise to complicated behavior for anisotropic and multiband systems such as $CeCoIn_5$. Indeed, for $j \parallel c$ no crossover is visible in $\rho(T)$, and the Wiedemann-Franz law, which is obeyed for $j \perp c$, seems to be violated [16].

In order to clearly show that, at the QCP in $CeCoIn_{5-x}Sn_x$, a finite energy scale $k_B T^\star$ exists which marks the crossover from 2D to 3D quantum critical behavior, measurements either of the fluctuation spectrum in equilibrium, for example, by inelastic neutron scattering (INS), or of thermodynamic properties are required. **q** scans of the INS over wide regions in reciprocal space, required to decide on the dimensionality of the quantum critical fluctuations, are not possible at high fields. Therefore, our thermodynamic measurements provide the only way to investigate this question and indeed prove such a crossover.

We now address the nature of quantum criticality (SDW-type or unconventional) in the regime where 2D-like behavior is observed. Theory suggests that 2D fluctuations are necessary for the occurrence of locally critical quantum criticality [7]. The latter is well established for the magnetic-field tuned AF QCP in the heavy fermion system $YbRh_2Si_2$ and its slightly Ge-doped variant $YbRh_2(Si_{0.95}Ge_{0.05})_2$, for which the critical field is almost

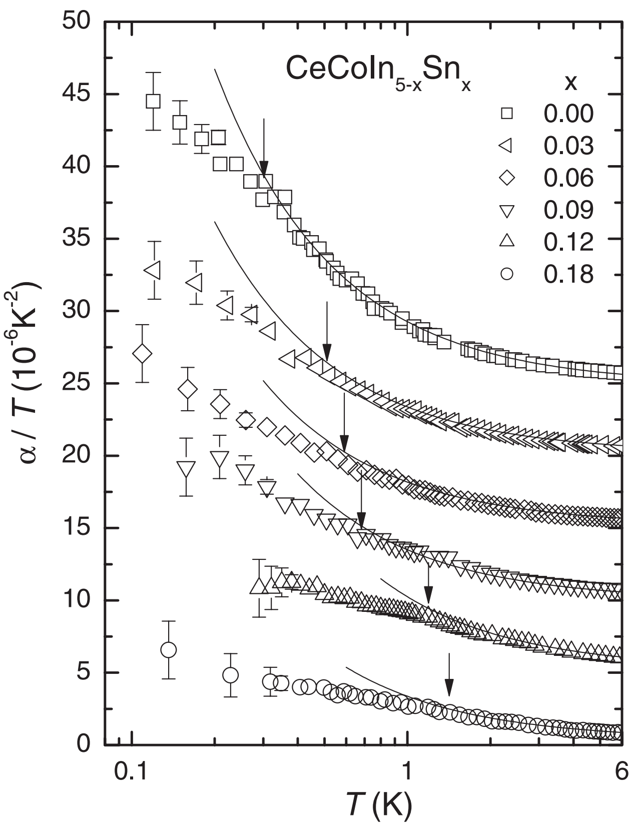


FIG. 3. Linear thermal expansion coefficient as $\alpha/T$ vs $T$ on a logarithmic scale for $CeCoIn_{5-x}Sn_x$ at $H \simeq H_{c2}(x)$ (given in Table I). Note that data sets are shifted by $5 \times 10^{-6}$ K$^{-2}$, subsequently. Arrows indicate the lower limit of $\alpha/T \propto T^{-1}$ behavior.

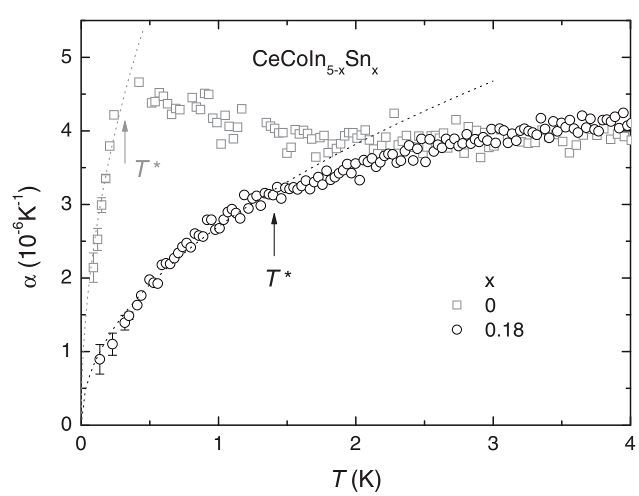


FIG. 4. Linear thermal expansion coefficient $\alpha(T)$ of $CeCoIn_5$ at $H = 5$ T ($\parallel c$, open squares), as well as $CeCoIn_{4.82}Sn_{0.18}$ at $H = 0$ (open circles). Dotted lines and arrows indicate $\alpha \propto \sqrt{T}$ and crossover temperatures $T^\star$, respectively.

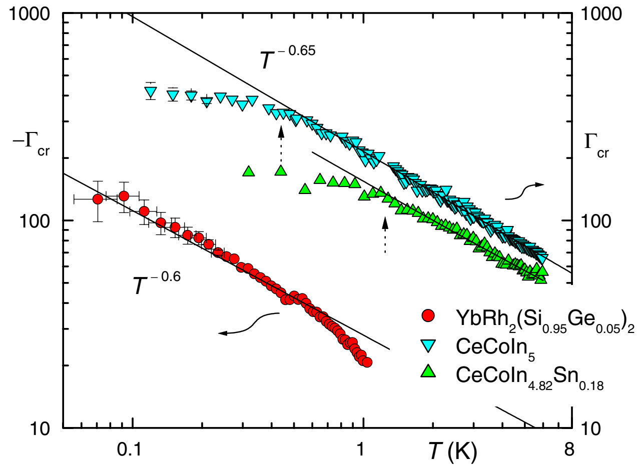


FIG. 5 (color online). Critical Grüneisen ratio $\Gamma_{cr} = (V_m/\kappa_T)(\alpha_{cr}/C_{cr})$, where $\alpha_{cr}$ and $C_{cr}$ denote thermal expansion and specific heat after subtraction of noncritical background contributions [18], of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ (left axis [2]) and $CeCoIn_{5-x}Sn_x$ ($x = 0$ at $H = 5$ T and $x = 0.18$ at 0 T, right axis). For the latter, the molar volume and isothermal compressibility equal $V_m = 9.57 \times 10^{-5}$ m$^3$/mol and $\kappa_T = (3.43 \pm 0.16) \times 10^{-3}$ GPa$^{-1}$ [25], respectively. A small background term [$0.35 \times 10^{-6}$ K$^{-2}$] has been subtracted from $\alpha/T$ for $x = 0.12$. No specific-heat background contributions have been subtracted since $C(T)/T \propto \log T$ at $T > T^\star$ ($T^\star$ indicated by dotted arrows). The so-derived critical Grüneisen ratio is invalid for $T < T^\star$, where the measured specific heat is dominated by a noncritical contribution [26]. Lines indicate power-law behavior at $T > T^\star$.

zero [2]. It is therefore very interesting to compare the low-$T$ thermodynamics of $CeCoIn_{5-x}Sn_x$ with the latter system. Of particular importance is the temperature dependence of the critical Grüneisen ratio $\Gamma_{cr}$, i.e., the ratio of the critical components of thermal expansion to specific heat. It has previously been shown, that $\Gamma_{cr}(T) \propto T^{-\epsilon}$ with $\epsilon = 1$ and 2/3 for conventional and unconventional quantum criticality, respectively [2].

Figure 5 shows striking similarities in the temperature dependence of the critical Grüneisen ratio of $YbRh_2(Si_{0.95}Ge_{0.05})_2$ and $CeCoIn_{5-x}Sn_x$ at temperatures above $T^\star(x)$, i.e., in the 2D regime: A rather similar fractional Grüneisen exponent is found which is close to the prediction for the locally critical QCP scenario in the presence of $xy$ anisotropy [7]. Theoretically, it has been shown that such behavior requires quasi-2D quantum critical fluctuations [7] supporting further the latter at $T > T^\star$ in $CeCoIn_5$. In view of the lack of superconductivity in $YbRh_2Si_2$ (at least for $T > 10$ mK), it is highly desirable to check whether or not a similar crossover towards conventional behavior at temperatures below the lower limit of previous studies (20 mK) takes place in the latter material.

To summarize, we have found thermodynamic evidence for a finite crossover scale $T^\star$ at the magnetic-field tuned QCP in $CeCoIn_5$. We associate $T^\star$ with a dimensional crossover from 2D ($T > T^\star$) to 3D ($T < T^\star$) quantum critical behavior. The introduction of disorder shifts the crossover scale towards higher temperatures.

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