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Quantum criticality in layered $\text{CeRhIn}_{5-x}\text{Sn}_x$ compared with cubic $\text{CeIn}_{3-x}\text{Sn}_x$

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Abstract – We report low-temperature thermal-expansion measurements on single crystals of the *layered* heavy fermion system $\text{CeRhIn}_{5-x}\text{Sn}_x$ ($0.3 \leq x \leq 0.6$) and compare it with a previous study on the related *cubic* system $\text{CeIn}_{3-x}\text{Sn}_x$ (KÜCHLER R. *et al.*, *Phys. Rev. Lett.*, **96** (2006) 256403). Both systems display a quantum critical point as proven by a divergent Grüneisen ratio. Most remarkably, the three-dimensional itinerant model explains quantum criticality in *both* systems, suggesting that the crystalline anisotropy in $\text{CeRhIn}_{5-x}\text{Sn}_x$ is unimportant. This is ascribed to the effect of weak disorder in these doped systems.

Quantum critical points (QCPs) in intermetallic compounds are of great scientific interest, as they provide the origin of non-Fermi liquid (NFL) behavior and novel ground states like unconventional superconductivity (SC). Heavy fermion (HF) systems, *i.e.* rare-earth or actinide-based compounds with competing Kondo and exchange interactions are prototype systems for the investigation of QCPs, and different classes of QCPs have been identified [1]. In one class, the observed properties are in agreement with the predictions of the spin-density-wave (SDW) theory, which considers the *f*-electrons as itinerant in the entire regime close to the QCP. In another class of materials (most prominent examples include $\text{CeCu}_{6-x}\text{Au}_x$ [2] and YbRh_2Si_2 [3–5]) there are strong indications for a localization-transition of the *f*-electrons due to the breakdown of Kondo screening at the QCP. SC has been observed in some but not all compounds close to QCPs and may even occur near first-order quantum phase transitions (QPTs) like in CeRh_2Si_2 [6,7] under pressure, which lack any signatures of NFL behavior.

There are several indications that magnetic anisotropy may be a crucial parameter for quantum criticality: i) quasi-two-dimensional (2D) magnetic fluctuations have been observed at the QCP in orthorhombic $\text{CeCu}_{5.9}\text{Au}_{0.1}$ [8] with an anomalous energy over

temperature scaling of the dynamical susceptibility [2], which strongly violates the predictions of the itinerant SDW theory; ii) a locally critical QCP has been predicted for the case of 2D magnetic fluctuations [9]; iii) SC in layered CeTIn_5 ($\text{T} = \text{Co}, \text{Ir}, \text{Rh}$) occurs at ten times higher temperatures compared to the cubic relative CeIn_3 [10,11]; and iv) spin-liquid formation among the local moments, proposed in the presence of strong geometrical frustration (which may possibly be enhanced in 2D magnetic systems), may act as competing mechanism against the Kondo-singlet formation [12,13].

In order to systematically investigate the relevance of magnetic anisotropy on quantum criticality, a comparison of cubic CeIn_3 with layered CeTIn_5 is most promising. The cubic point symmetry of Ce atoms in the former must lead to isotropic magnetic fluctuations. By contrast, in CeTIn_5 the alternating series of CeIn_3 and TIn_2 , stacked along the *c*-axis (for the crystal structures see fig. 1), is responsible for a strongly 2D character of the Fermi surface [14] and may also lead to quasi-2D magnetic fluctuations [15], although the magnetic correlation length in CeRhIn_5 above T_N [16] as well as in superconducting CeCoIn_5 [17] shows only a moderate anisotropy.

Hydrostatic pressure experiments have been performed on cubic CeIn_3 (Néel temperature T_N at ambient pressure about 10 K) as well as layered CeRhIn_5 ($T_N = 3.8$ K). In both cases, the AF ordering vanishes discontinuously

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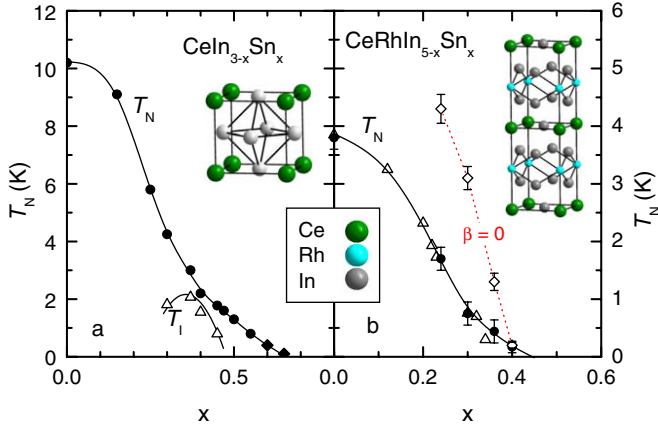


Fig. 1: (Color online) Magnetic phase diagrams for cubic $\text{CeIn}_{3-x}\text{Sn}_x$ [22] (a) and tetragonal $\text{CeRhIn}_{5-x}\text{Sn}_x$ (b). The open triangles and closed circles in (b) represent Néel temperature as determined from specific-heat [23] and thermal-expansion ([24] and this study) measurements. The red dotted line through open diamonds indicates zero crossing of the volume expansion with $\beta(T) > 0$ (< 0) on its right (left) side.

as a function of applied pressure [18–20], and the nature of the f -electrons, as determined from de Haas-van Alphen (dHvA) experiments at low temperatures and high magnetic fields, changes from localized to itinerant at the critical pressure [21]. An important difference between the two systems is that SC in CeIn_3 occurs only in a very narrow pressure regime and below 0.2 K, whereas T_c values above 2 K are observed in CeRhIn_5 between 2 and 4 GPa. Although the low- T electrical resistivity of CeIn_3 has shown an anomalous exponent of 1.6 [10], nuclear quadrupole resonance suggests a Landau Fermi liquid ground state [18], and the cyclotron mass derived from dHvA experiments is constant near the discontinuous QPT [21]. For CeRhIn_5 , the cyclotron mass $m^*(p)$ [21] and the coefficient $A(p)$ of T^2 behavior in the electrical resistivity at 15 T [20] show diverging behavior, suggesting a field-induced QCP close to $p_c \approx 2.5$ GPa. Previously it has been demonstrated, that Sn-doping in $\text{CeIn}_{3-x}\text{Sn}_x$ [22] as well as $\text{CeRhIn}_{5-x}\text{Sn}_x$ [23,24] leads to a continuous suppression of AF order without formation of SC around the QCP. Therefore, and because the *same* control parameter (Sn-doping) is used to tune the QCP¹ these two systems seem to be ideally suited to perform the desired comparative study on the effect of lattice anisotropy on quantum critical behavior. Figure 1 compares the phase diagrams of the two systems.

Since Sn has one more p -electron compared to In, the partial substitution of In- by Sn-atoms increases the conduction electron density of states and thus the Kondo

¹In a previous paper [25], the *field*-induced QCP was studied in Sn-doped CeCoIn_5 . These data can, however, not *a priori* be compared with those obtained at the *doping*-induced QCP in $\text{CeIn}_{3-x}\text{Sn}_x$ [22]. For, magnetic field-tuning has been found, *e.g.* in $\text{CeCu}_{6-x}\text{Au}_x$, to furnish quite different quantum critical behavior than tuning by chemical doping [26].

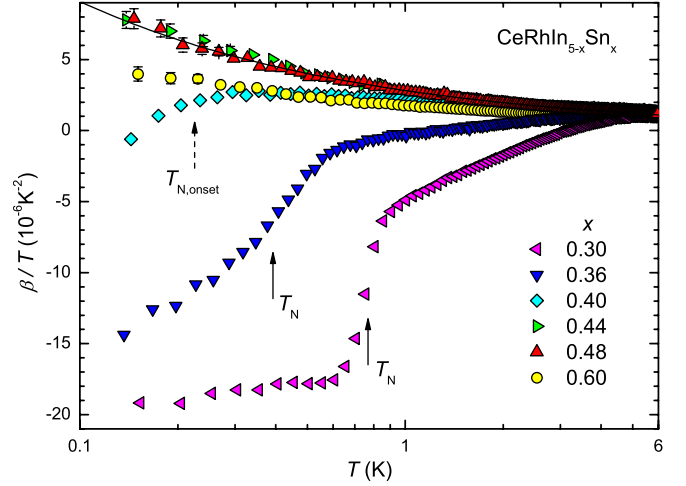


Fig. 2: (Color online) Temperature dependence of the volume thermal-expansion coefficient $\beta = \alpha_{\parallel} + 2\alpha_{\perp}$ of $\text{CeRhIn}_{5-x}\text{Sn}_x$ as β/T vs. T (on a logarithmic scale). The solid and dotted arrows indicate T_N and onset of (broadened) Néel transition, respectively. Line displays $\beta(T)/T = b_0 + b_1/\sqrt{T}$ dependence with $b_0 = -0.08 \times 10^{-6} \text{ K}^{-2}$ and $b_1 = 2.9 \times 10^{-6} \text{ K}^{-1.5}$.

temperature, leading to a suppression of the AF ordering (a small increase of lattice constants with Sn-doping is subdominant). For $\text{CeIn}_{3-x}\text{Sn}_x$ a clear change in the slope of $T_N(x)$ occurs close to a presumed tetracritical point at $x \approx 0.4$ [27] beyond which a quasi-linear suppression of the Néel temperature towards a QCP at $x_c = 0.65$ has been found [22]. The phase diagram of $\text{CeRhIn}_{5-x}\text{Sn}_x$ as displayed in fig. 1(b) is rather similar and also shows a change of the $T_N(x)$ slope before the QCP is reached. Previous low-temperature specific-heat and electrical resistivity measurements down to 0.4 K suggest a QCP near $x = 0.4$. Our thermal-expansion measurements at $T \geq 0.08$ K, discussed below, reveal $x_c \simeq 0.46$.

For this study, we have used the same single crystals studied in [23,24], as well as $x = 0.44$ and $x = 0.6$ single crystals prepared similarly. We always refer to the actual Sn concentration x determined by microprobe analysis with an uncertainty of less than 1%. The residual resistivity of the $\text{CeRhIn}_{5-x}\text{Sn}_x$ crystals increases monotonically with x and reaches $28 \mu\Omega\text{cm}$ at $x = 0.48$ [23]. Extended X-ray absorption fine-structure measurements in $\text{CeCoIn}_{5-x}\text{Sn}_x$ have revealed that the Sn atoms preferentially occupy the In-(1) position within the CeIn_3 planes of the layered system [28]. Similar behavior arises in $\text{CeRhIn}_{5-x}\text{Sn}_x$ [29]. The linear thermal expansion $\alpha(T) = d[\Delta L(T)/L]/dT$ has been determined with the aid of a high-resolution capacitive dilatometer, attached to a dilution refrigerator. The volume expansion coefficient displayed in fig. 2 has been determined by $\beta = \alpha_{\parallel} + 2\alpha_{\perp}$.

Thermal-expansion measurements on $\text{CeRhIn}_{5-x}\text{Sn}_x$ to investigate the long-range antiferromagnetism have been discussed previously [24]. For $x \leq 0.24$, a positive discontinuity $\Delta\beta > 0$ has been observed at the Néel

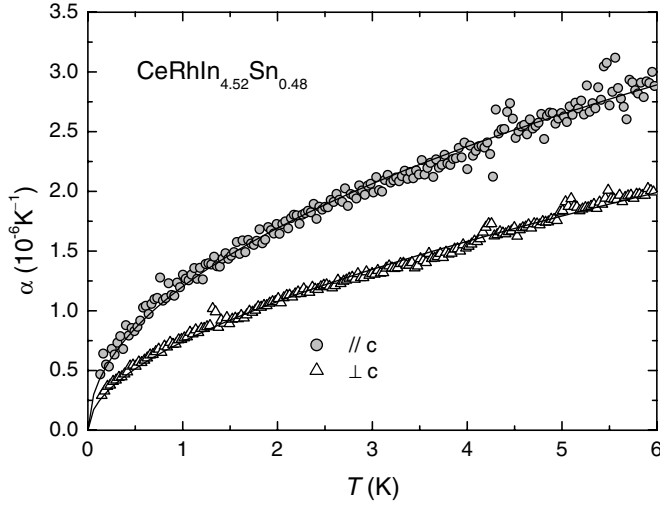


Fig. 3: CeRhIn_{4.52}Sn_{0.48} linear thermal-expansion coefficient along (circles) and perpendicular (triangles) to the c -axis. The lines indicate $\alpha(T) = a_0T + a_1\sqrt{T}$ with $a_0 = -0.01 \times 10^{-6} \text{ K}^{-2}$ ($0.05 \times 10^{-6} \text{ K}^{-2}$) and $a_1 = 1.2 \times 10^{-6} \text{ K}^{-1.5}$ ($0.7 \times 10^{-6} \text{ K}^{-1.5}$) for $\alpha_{||}$ (α_{\perp}). The noise for $\alpha_{||}$ is relatively large, because the sample length along the c -axis is rather small.

temperature, reflecting an increase of T_N with hydrostatic pressure. These samples are thus located on the left side of the maximum of $T_N(P)$ expected within the Doniach diagram. Beyond $x = 0.24$, where the change in slope in $T_N(x)$ occurs (cf. fig. 1(b)), $\Delta\beta < 0$, indicating that the system approaches the QCP. A change of sign in the volume thermal expansion $\beta(T)$ which indicates an accumulation point of entropy [30] occurs for CeIn_{3-x}Sn_x very close to $T_N(x)$ [22]. By contrast it is located much above the Néel temperature for layered CeRhIn_{5-x}Sn_x (cf. the red dotted line in fig. 1(b)). This may indicate a largely extended Ginzburg regime in which classical critical fluctuations dominate. However, specific-heat measurements have shown a Schottky-like anomaly in $C(T)/T$ very close to this line. Thus, an additional energy scale exists in this system which is likely related to short-range magnetic correlations [23]. Most interestingly, it also vanishes in the vicinity of the QCP, *i.e.* in the range $0.4 < x < 0.44$ (cf. fig. 2). The QCP is located in between the concentrations $x = 0.44$, and $x = 0.48$ for which, within experimental resolution, the same divergent behavior in $\beta(T)/T$ is found down to the lowest temperatures. At higher Sn content, $x = 0.60$, $\beta(T)/T$ tends to saturate at lowest temperatures, indicative for a crossover towards Landau Fermi liquid behavior. In the following, we will analyze quantum criticality for $x = 0.48$.

Figure 3 displays measurements of the linear thermal-expansion coefficient of CeRhIn_{4.52}Sn_{0.48} along and perpendicular to the c -axis. Despite moderate anisotropy ($\alpha_{||}/\alpha_{\perp} = 1.7$), similar behavior is found along both directions, namely a square-root behavior in $\alpha(T)$. Such a temperature dependence is expected within the itinerant SDW theory for a 3D AF QCP [31]. By contrast,

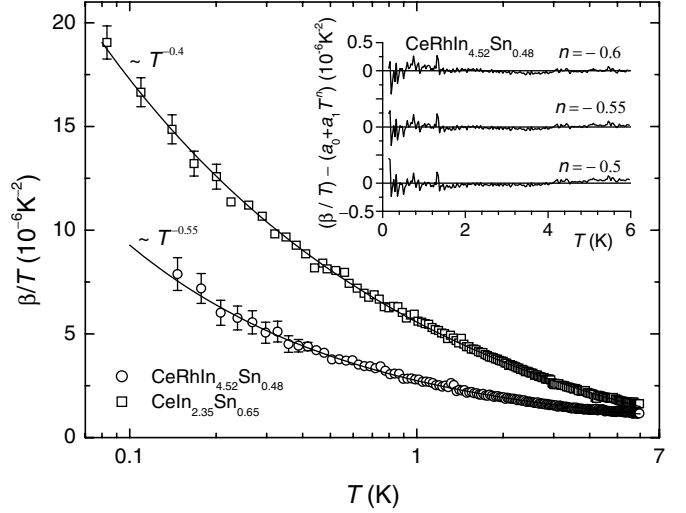


Fig. 4: Volume expansion coefficient as β/T vs. T on logarithmic scale for CeRhIn_{4.52}Sn_{0.48} (circles) and CeIn_{2.35}Sn_{0.65} (squares, [22]). The solid lines indicate power law behavior. The inset displays the deviation of $\beta(T)/T$ data for CeRhIn_{4.52}Sn_{0.48} from the best-fit description $\beta(T)/T = a_0 + a_1T^n$ using different exponents n . The constant term is negligible in all cases, $|a_0| \leq 0.2 \times 10^{-6} \text{ K}^{-2}$. For clarity, the three data sets have been shifted by different amounts vertically.

$\alpha(T) \approx \text{const}$ is expected in the 2D case. Further evidence for the 3D nature of quantum criticality in CeRhIn_{5-x}Sn_x is provided by the analysis of the volume thermal expansion and Grüneisen ratio and comparison with the case of cubic CeIn_{3-x}Sn_x. In the latter case the AF QCP is located at $x = 0.65$, and the volume thermal expansion has been described by $\beta(T)/T = a_0 + a_1T^n$, yielding $n = -0.4$ for a fit in the temperature range $0.1 \text{ K} \leq T \leq 6 \text{ K}$ and $n = -0.5$ for a fit at temperatures below 1 K [22]. A value of $n = -0.5$ agrees with the prediction of the SDW theory for a 3D AF QCP, whereas $n = -1$ is expected for the 2D case [31]. In fig. 4, we compare β/T for CeIn_{2.35}Sn_{0.65} with respective data on CeRhIn_{4.52}Sn_{0.48}. The best-fit description of the latter system reveals an exponent $n = -0.55 \pm 0.05$ (cf. the inset which displays the deviation from power law fits with variable exponent). Thus, thermal expansion does not reveal a significant difference between quantum criticality in layered CeRhIn_{5-x}Sn_x compared to cubic CeIn_{3-x}Sn_x.

The specific heat of CeRhIn_{5-x}Sn_x has been investigated down to 0.4 K [23]. As shown in the inset of fig. 5, $C(T)/T$ follows a $\log T$ -dependence between 0.4 and 20 K, similar as observed in many other NFL systems [32,33]. This temperature dependence is expected within the 3D SDW theory in an intermediate range [34], while at lowest temperatures a crossover to a square-root temperature dependence is predicted. Such an expected crossover has indeed been found in CeIn_{2.35}Sn_{0.65} around 0.4 K [35], although below 0.2 K the specific heat is dominated by the nuclear quadrupolar contribution of indium. As already discussed for the case of CeCoIn_{5-x}Sn_x [25],

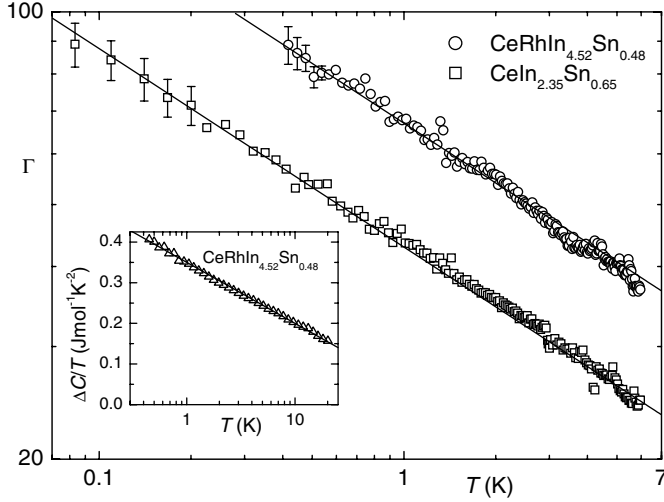


Fig. 5: Temperature dependence (on double-log scales) of the Grüneisen parameter $\Gamma = (V_{mol}\beta)/(\kappa_T\Delta C)$ of $\text{CeRhIn}_{4.52}\text{Sn}_{0.48}$ and $\text{CeIn}_{2.35}\text{Sn}_{0.65}$ [22], where V_m and κ_T denote the molar volume and the isothermal compressibility, respectively. Solid lines display $T^{-0.31}$ behavior. The inset displays the specific-heat increment $\Delta C = C - C_{\text{Phonon}}$ of $\text{CeRhIn}_{4.52}\text{Sn}_{0.48}$ as $\Delta C/T$ vs. T on a logarithmic scale. The phonon contribution has been determined from data on LaRhIn_5 [23].

thermal expansion and the Grüneisen parameter are thus ideally suited to investigate the nature of quantum criticality in these systems as they mitigate the obscuring effects of the nuclear contribution to specific heat. Theory predicts a stronger than logarithmic divergence of the Grüneisen parameter $\Gamma(T)$ for any pressure-sensitive QCP [31], otherwise, as recently found in $\text{CePd}_{1-x}\text{Rh}_x$ [36], quantum criticality as source of NFL behavior could be excluded. As shown in fig. 5, which displays $\Gamma(T)$ on double-logarithmic scales, such stronger-than $\log T$ divergence is indeed present for $\text{CeRhIn}_{4.52}\text{Sn}_{0.48}$ and $\text{CeIn}_{2.35}\text{Sn}_{0.65}$. Most interestingly, a very similar T -dependence is found for the two systems. In order to compare with the theoretical predictions, the *critical* Grüneisen ratio $\Gamma^{cr} \propto \beta^{cr}/C^{cr}$ must be analyzed, where β^{cr} and C^{cr} denote the volume thermal expansion and specific heat after subtraction of non-critical, *i.e.*, Fermi-liquid-like contributions. Within the 3D SDW model, the noncritical contribution to specific heat is given by the saturation value of $C(T)/T$ as $T \rightarrow 0$ [31]. Assuming a saturation of the specific-heat coefficient at either 0.5, 0.55 or $0.6 \text{ Jmol}^{-1}\text{K}^{-2}$, respectively, would yield values of -1.2 , -1 or -0.93 for the Grüneisen exponents within the temperature interval $0.4 \text{ K} \leq T \leq 6 \text{ K}$. For $\text{CeIn}_{2.35}\text{Sn}_{0.65}$, a value of -1.1 ± 0.1 has been found [22]. Thus, the critical Grüneisen analysis also suggests strong similarities in quantum critical behavior of the two systems.

Weak disorder may strongly influence the nature of quantum criticality and the dimensionality of the critical fluctuations, whereas strongly disordered systems like

$\text{UCu}_{5-x}\text{Pd}_x$ do not display a QCP, and NFL behavior in such systems appears to be disorder-driven [37]. Previously, we have investigated the influence of disorder on quantum criticality in $\text{CeCoIn}_{5-x}\text{Sn}_x$ [25]. With increasing x , a field-tuned QCP remains pinned to the upper critical field $H_{c2}(x)$ of heavy-fermion superconductivity, which is linearly suppressed to 0 at $x = 0.18$ [38]. While the specific heat remains virtually unchanged with x at the respective critical fields, thermal expansion delineates a crossover scale $T^*(x)$ separating 2D from 3D quantum critical behavior [25]. This crossover scale increases from 0.3 K at $x = 0$ to 1.4 K at $x = 0.18$ with increasing disorder (x), characterized by a residual resistivity $\rho_0 = 15 \mu\Omega\text{cm}$ for $x = 0.18$. In $\text{CeRhIn}_{5-x}\text{Sn}_x$, even four-times higher Sn-concentrations (resulting in $\rho_0 = 28 \mu\Omega\text{cm}$) are required to access the QCP. We conjecture that 3D behavior therefore extends up to at least 6 K. This interpretation assumes that isotropic impurity scattering due to the In-Sn site disorder is effective in smearing out the anisotropy of the quantum critical fluctuations. Nevertheless, as evidenced by the divergent Grüneisen ratio, a truly pressure-sensitive QCP emerges. This is in contrast to strongly disordered systems mentioned above which do not display a QCP. Furthermore, we note that $\rho_0 \approx 40 \mu\Omega\text{cm}$ of $\text{CeCu}_{5.9}\text{Au}_{0.1}$ [39] is even 40% larger as in $\text{CeRhIn}_{4.52}\text{Sn}_{0.48}$ but $\text{CeCu}_{5.9}\text{Au}_{0.1}$ nevertheless displays quasi-2D quantum critical fluctuations [8]. This indicates that the CeMIn_5 systems are rather sensitive to disorder within the tetragonal CeIn_3 plane and, in general, that weak disorder can drastically influence quantum criticality.

We conclude by stating that weak disorder as introduced by low Sn-doping in layered CeRhIn_5 stabilizes *three-dimensional* quantum critical behavior. Thus, no significant differences in the low- T thermal expansion and Grüneisen ratio to the corresponding quantities in cubic $\text{CeIn}_{3-x}\text{Sn}_x$ could be resolved. The distinct effect of weak disorder on the quantum criticality in the 115 systems appears to be independent of the experimental tuning parameter, as this had already been observed at the *magnetic field-induced QCP* in Sn-doped CeCoIn_5 [25].

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