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Systematic study of the Grüneisen ratio near quantum critical points

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Abstract

At any pressure-sensitive quantum critical point (QCP), the volume thermal expansion coefficient is more singular than the specific heat. Consequently, the resulting critical Grüneisen ratio \( G_{\text{cr}} / C_{24} \) diverges. The related critical exponent \( \varepsilon \) in \( G_{\text{cr}} / C_{24} \) can be used to characterize the nature of the underlying quantum critical fluctuations. We have performed a comparative study on various heavy fermion (HF) systems close to antiferromagnetic QCPs. In particular, we have studied (i) CeIn₃ₓSnₓ, (ii) CeNi₂Ge₂, (iii) YbRh₂(Si₀.₉₅Ge₀.₀₅)₂, as well as (iv) CeCu₅.₈Ag₀.₂, all of which show a divergent Grüneisen ratio. For the two former systems the critical exponent \( \varepsilon = 1 \) is compatible with the predictions of the well-established Hertz–Millis–Moriya theory for three-dimensional extended quantum critical fluctuations. By contrast, for the two latter systems \( \varepsilon < 1 \) is found to be incompatible with “conventional” quantum criticality. Our results thus suggest the existence of at least two different classes of QCPs in HF systems.

Keywords: Critical point; Phase; Fluctuations; Temperature; Instability; Electron; Metals

1. Introduction

Heavy fermion (HF) systems, i.e. intermetallics of certain rare earths and actinides that contain a lattice of 4f- or 5f-derived moments, have turned out to be prototypical systems for the study of non-Fermi liquid (NFL) behavior [1]. Their ground-state behavior is determined by the competition between the on-site Kondo and the inter-site exchange interaction. When both interactions are balanced, a quantum critical point (QCP), separating a paramagnetic from a magnetically ordered ground state, occurs. The nature of quantum criticality in HF systems has been the focus of interest in recent years. Different scenarios for the QCP, where long-range antiferromagnetic (AF) order emerges from the HF state, have been proposed, e.g. a spin density wave (SDW) and a localized moment scenario. In the traditional SDW-picture [2–4], the quasiparticles retain their itinerant character when these materials are tuned into the long-range ordered state and, as a consequence, form an SDW-type of AF order. Recent experiments have shown that at least in some HF systems this picture fails [5,6]. Consequently, a new type of QCP has been proposed at which the quasiparticles break up into conduction electrons and local 4f moments, which order antiferromagnetically at sufficiently low temperatures. This picture has been labelled a “locally critical” scenario and is based on the destruction of the local Kondo resonances at the QCP [7,8].

In this paper, we give an overview on the use of measurements of the thermal expansion and critical Grüneisen ratio. While the Grüneisen parameter \( G/C \), where \( C \) denotes the specific heat and \( G \) the volume thermal expansion, has long been used to describe the volume dependence of various physical processes [9], our results establish this quantity as highly suited to characterize QCPs. As a consequence of the entropy accumulation close to the zero-temperature instability, the critical Grüneisen ratio \( G_{\text{cr}}(T) \sim \beta_{\text{cr}}(T)/C_{\text{cr}}(T) \) (here \( \beta_{\text{cr}}(T) \) and \( C_{\text{cr}}(T) \) denote the quantum critical contributions to the volume thermal expansion and the electronic specific heat, respectively) diverges in the approach of any pressure-sensitive QCP [10]. Within the SDW theory, the divergence is given by
$I^\text{ex} \sim 1/T^{4/\nu}$ with the spatial correlation-length exponent $\nu$ and the dynamical exponent $z$. Measurements of the Grüneisen ratio are therefore apt to check the very existence of a QCP and to characterize its properties. In the following, we focus on CeNi$_2$Ge$_2$ [11], CeIn$_3$–Sn$_x$ [12], YbRh$_2$Si$_2$Ge$_2$ [6,13] and CeCu$_{5.8}$Ag$_{0.2}$ [14]. All four of them are well-characterized HF systems in the close vicinity of AF QCPs.

2. CeNi$_2$Ge$_2$

CeNi$_2$Ge$_2$ is a paramagnetic HF system with a single-ion Kondo scale of $T_K \approx 30$ K, crystallizing in the tetragonal ThCr$_2$Si$_2$ structure [15]. At zero magnetic field, pronounced NFL effects have been observed in thermodynamic and electrical transport experiments [11] related to a nearby magnetic instability. Indeed, substitution of Ni with the larger Pd in Ce(Ni$_{1-x}$Pd$_x$)$_2$Ge$_2$ induces long-range AF order below $T_N = 2$ K for $x = 0.2$ [16]. Furthermore, by applying magnetic fields, a Landau–Fermi liquid (LFL) state is induced with a coefficient $A(B)$, derived from the electrical resistivity $\Delta \rho = AT^2$, that diverges for $B \to 0$ [11]. Nevertheless, there are conflicting specific heat results about whether NFL behavior exists down to lowest temperatures or whether a cross-over to LFL behavior occurs. The earliest measurements show a small cusp in $C(T)/T$ at 0.3 K [15], while more recent work revealed a cross-over from a logarithmic increase above 0.3 K to a saturation at lower temperatures [17]. By contrast, $C(T)/T$ of a high-quality sample with very low residual resistivity does not saturate but shows an upturn at lowest temperatures [18]. We measured both the low-temperature volume thermal expansion, $\beta$, and specific heat, $C$, on high-quality single crystals with a residual resistivity of 5 $\mu$cm. Fig. 1b displays the volume expansion coefficient $\beta$, plotted as $\beta(T)/T$. $\beta(T)$ has been calculated from the linear thermal expansion coefficients of CeNi$_2$Ge$_2$ measured along the tetragonal $a$- and $c$-axis: $\beta = 2\beta_a + \beta_c$. $\beta(T)/T$ is not constant on cooling, as expected for an LFL, but shows a divergence over more than two decades in temperature from 6 K down to at least 50 mK. This clean observation of NFL behavior provides striking evidence that the single crystal is located very close to a QCP. As shown by the solid red line, the data can be described in the entire $T$ range investigated by the temperature dependence predicted by the three-dimensional (3D)-SDW scenario [10], i.e. the sum of a singular ($1/\sqrt{T}$) and a normal (constant) contribution. The corresponding fit function is described by $\beta(T)/T = a/\sqrt{T} + b$, with $a = 3.5 \times 10^{-8}$ K$^{-1.5}$ and $b = 1.7 \times 10^{-6}$ K$^{-2}$. We now discuss the low-temperature specific heat (Fig. 1a) measured on the same sample. Below 3 K the electronic contribution of the specific heat could be described by $C(T)/T = \gamma_0 - c\sqrt{T}$ (red line), compatible with the 3D-SDW QCP scenario. Here we have assumed that the low-temperature upturn $C/T \sim a/T^3$ (dotted line), also present in this single crystal, is described by the high-temperature tail of a (nuclear) Schottky anomaly. However, the latter is yet unknown, since internal magnetic fields of the order of 30 T acting on the nuclear $^{61}$Ni or $^{75}$Ge spin states would be necessary to produce this contribution to our $B = 0$ data [18]. Of particular importance is the very fact that this upturn cannot be part of the quantum critical behavior: It remains unchanged in a magnetic field of about 2 T (yellow curve) that tunes the system toward the LFL regime. The influence of the low-temperature or whether a cross-over to LFL behavior in this system arises, whether the mechanism of NFL behavior in this system can be described by the itinerant 3D-SDW QCP

3. CeIn$_{3-x}$Sn$_x$

Next, we focus on the HF system CeIn$_{3-x}$Sn$_x$, which crystallizes in the Cu$_3$Au structure with cubic point symmetry of the Ce-atoms (see inset of Fig. 3). It has been chosen for a detailed study, as here low-dimensional spin-fluctuations can be ruled out. Thus, the interesting question arises, whether the mechanism of NFL behavior in this system can be described by the itinerant 3D-SDW QCP.
theory. The magnetic ($x, T$) phase diagram of polycrystalline CeIn$_{3-x}$Sn$_x$ has been thoroughly studied for $0 \leq x \leq 1$ by susceptibility [12], specific heat [21] and resistivity measurements [22,23] (see the inset of Fig. 3). Whereas $T_N$ for undoped CeIn$_3$ vanishes abruptly below 3 K under hydrostatic pressure [24], it can be traced down to 0.1 K for CeIn$_{3-x}$Sn$_x$. These differences are related to the change of the electronic structure induced by Sn doping. Beyond a possible tetracritical point at $x \approx 0.4$ [21], an almost linear dependence of $T_N(x)$ is observed. Fig. 3 shows the volume thermal expansion $\beta$ of single crystals of CeIn$_{3-x}$Sn$_x$ with $x = 0.55, 0.65, 0.7$ and 0.8 plotted as $\beta(T)/T$ vs. log $T$. For $x = 0.55$ the broadened step-like decrease in $\beta(T)/T$ at $T_N \approx 0.6$ K marks the AF phase transition, in perfect agreement with specific heat measurements on the same single crystal [25]. On increasing the concentration, we find for $x = 0.65$ and 0.7 diverging behavior over nearly two decades in $T$ down to 80 mK. These data suggest that $T_N$ is suppressed at a critical concentration $x_c \approx 0.67 \pm 0.03$, also consistent with specific heat measurements performed on the same samples [25]. Finally, for $x = 0.8$ we recover LFL behavior, $\beta(T)/T \approx \text{const.}$ for $T \rightarrow 0$. According to the expression $\beta(T)/T = a + bT^0$, the analysis of the $x = 0.65$ data reveals a fit of excellent quality (cf. the red dashed line in Fig. 3) for temperatures up to 1 K. The resulting exponent $\delta$ equals $-0.5$, i.e. the value predicted by the 3D-SDW QCP scenario [10] (for detailed analysis see [26]). Fig. 4 displays

![Fig. 2. Temperature dependence of the Grüneisen ratio $\Gamma' = V_m \kappa \beta/C$ [28], where $V_m$ and $\kappa$ are the molar volume and isothermal compressibility, respectively. The black line has not been measured for CeNi$_2$Ge$_2$, we use $\kappa_T = 8.33 \times 10^{-12} \text{ Pa}^{-1}$ obtained for the isostructural and related HF compound CeCu$_2$Si$_2$ [19]. The inset shows the critical component of the Grüneisen ratio $\Gamma_{cr} = V_m \kappa_T \beta_{cr}/C$ as log $\Gamma_{cr}$ vs. $\log T$ (at $B = 0$) with $\beta_{cr} = \beta(T) - \beta$ and $C_{cr} = C(T)/(\gamma T + \delta/T)^2$. The solid red line represents $\beta_{cr} \approx 1/T^\varepsilon$ with $\varepsilon = 1$.](image)

![Fig. 3. Volume thermal expansion coefficient $\beta$ of CeIn$_{3-x}$Sn$_x$ single crystals as $\beta(T)/T$ vs. $\log T$ [26]. The red dashed line shows $T^{-0.5}$ behavior. The arrow indicates AF phase transition. The inset shows the magnetic phase diagram for cubic CeIn$_{3-x}$Sn$_x$ ($x \leq 1$). Closed circles and diamonds indicate $T_{N}$ determined from specific heat [21] and electrical resistivity [22] measurements, respectively. Open diamonds marks $T^*$, the upper limit of Landau Fermi-liquid behavior, e.g. $\Delta p(T) \sim T$ [23]. Open triangles indicate the first-order transition $T_T$ [21].](image)

![Fig. 4. Temperature dependence of the Grüneisen parameter $\Gamma' = V_m \kappa \beta/C$ for CeIn$_{3-x}$Sn$_x$, ($x = 0.65$) [26]. $V_m = 6.25 \times 10^{-5} \text{ m}^3 \text{mol}^{-1}$ and $\kappa_T = 1.49 \times 10^{11} \text{ Pa}^{-1}$ [27] are the molar volume and isothermal compressibility, respectively. The inset shows the critical component of the Grüneisen ratio $\Gamma_{cr} = V_m \kappa_T \beta_{cr}/C$ of the same single crystal as log $\Gamma_{cr}$ vs. $\log T$ with $\beta_{cr} = \beta(T) - \beta$ and $C_{cr} = C(T)/(\gamma T + \delta/T)^2$ derived after subtraction of background contributions (see text). The solid red line represents $\beta_{cr} \approx 1/T^\varepsilon$ with $\varepsilon = 1.1 \pm 0.1$.](image)
the temperature dependence of the Grüneisen ratio $\Gamma(T)$ for the $x = 0.63$ sample calculated from the specific heat and thermal expansion measured on the same single crystal. Divergent behavior is observed down to the lowest accessible temperature with very large $\Gamma$ values at 0.1 K, which are of similar size as those found for the other quantum critical HF systems discussed in this paper. The fact that the divergence of $\Gamma(T)$ in the quantum critical regime is stronger than logarithmic provides clear evidence for a pressure-sensitive QCP in the system. If the disorder present in the system would lead to a “smeared” quantum critical regime, $\Gamma(T)$ could diverge at most logarithmically [10]. In order to compare our results with the theoretical predictions for an SDW QCP [10], we analyze the critical Grüneisen ratio [26] that is defined on double logarithmic scales in the inset of Fig. 4. We find $\Gamma^c \sim T^{-\varepsilon}$ with an exponent close to 1, similar as for CeNi$_2$Ge$_2$, providing evidence for the applicability of the 3D itinerant theory in both systems.

4. YbRh$_2$Si$_2$

We now turn to YbRh$_2$Si$_2$, for which pronounced NFL behavior is observed above a weak AF ordering at $T_N = 70$ mK [13]. The ordering can be further weakened by a tiny volume expansion induced by the substitution of nominally 5 at% Ge for the smaller but isoelectronic Si in YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$ [6]. Here, $T_N = 20$ mK, and a field-induced QCP occurs at a magnetic field of 0.027 T applied within the tetragonal plane. For temperatures above 50 mK, no effect of the AF transition is detected and the zero-field data above 50 mK do probe the true quantum critical behavior [6]. In YbRh$_2$Si$_2$, the volume thermal expansion coefficient $\beta(T)$ has a negative sign reflecting the decrease of the Kondo temperature with pressure, which is opposite as in the case of Ce-based HF systems. In Fig. 5, we compare the temperature dependence of the electronic specific heat $C_{el}(T)$ with that of the volume expansion coefficient $\beta(T)$. At $T > 1$ K, $\beta(T)$ can be fit by $-T\log(T_0/T)$ with $T_0 \approx 13$ K (see the left solid black line in Fig. 5). At $T < 1$ K, the best fit is given by $a_1 + a_0 T$. Both fits are not only different from the expected 3D-SDW results discussed earlier, but also weaker than the log-log $T$ form [10] expected in a 2D-SDW picture. The maximum at 20 mK in $C_{el}(T)/T$ marks the onset of very weak AF order [6]. This is suppressed by $B_c = 0.027$ T applied in the easy plane. At $B = B_c$, a power law divergence $C_{el}(T)/T \propto T^{-1/3}$ is observed for $T < 0.3$ K, which is clearly incompatible with the 2D-SDW picture [6]. At higher temperatures, the specific heat coefficient also varies logarithmically [6]. Because of the different slope compared with that of $\beta(T)/T$, the Grüneisen ratio is strongly temperature dependent above 1 K. Most importantly, below 1 K the critical Grüneisen ratio (see the inset of Fig. 5) diverges as $T^{-2/3}$, incompatible with the prediction of the SDW theory for an AF QCP. By contrast, such a fractional exponent would be consistent with the locally quantum critical picture [7,29]. Hall effect measurements have recently revealed a dramatic change of the Hall coefficient when tuning YbRh$_2$Si$_2$ across the QCP by varying the magnetic field [30]. This change is observed when crossing a scale $T^* (B)$ that merges at $T = 0$ at the critical field of 0.05 T. Thermodynamic measurements prove that $T^*(B)$ is an intrinsic energy scale (in the equilibrium spectrum) that vanishes at the QCP [31]. These results indicate that quantum criticality in YbRh$_2$Si$_2$ cannot be described within the conventional SDW theory. Unconventional quantum criticality with multiple vanishing energy scales seems to incorporate inherently quantum degrees of freedom that are not included in conventional order-parameter fluctuation theory.

5. CeCu$_{5.8}$Ag$_{0.2}$

Finally, we concentrate on the thermal expansion and critical Grüneisen ratio analysis of orthorhombic CeCu$_{5.8}$Ag$_{0.2}$. Our experiments are motivated by INS experiments on the related system CeCu$_{5.8}$Au$_{0.2}$, which have revealed an anomalous $q$-independence in the critical response and led to the proposal of a locally critical QCP scenario [5]. Fig. 6 displays the low-temperature specific heat divided by temperature, $C(T)/T$, of various CeCu$_{5.8}$Ag$_{0.2}$ samples on a logarithmic temperature scale [32]. Long-range AF order is observed for $x \gtrsim 0.3$ and manifests itself by broadened jumps in $C(T)/T$. The inset shows $T_N(x)$ as determined by an (entropy-conserving)
equal-areas construction and the maximum of the derivative $d\rho(T)/dT$ in corresponding electrical resistivity measurements. Extrapolation of $T_N$ to zero temperature yields a critical concentration $x_c = 0.2$ (see the inset of Fig. 6). CeCu$_{5.8}$Ag$_{0.2}$ belongs to a class of CeCu$_6$-based HF systems in which the AF QCP is reached by doping the Cu-site. Common to all of these systems is a universal $C/T\propto\log(T_0/T)$ dependence ($T_0 \approx 6\text{ K}$) of the specific heat coefficient over nearly two decades in temperature down to $50\text{ mK}$ (see the green curve in Fig. 6). This behavior would be compatible with theoretical predictions of the itinerant scenario for an AF QCP in the presence of 2D critical spin-fluctuations. Indeed, INS experiments on CeCu$_{5.9}$Au$_{0.1}$ revealed rod-like structures of high intensity in $q$-space translating to quasi-2D fluctuations in real space while the critical fluctuations. For the SDW theory, both in 3D and 2D, a $1/T$ divergence of the critical Grueneisen ratio is expected. On the other hand, there are indications for a more complicated structure of the critical magnetic fluctuations in both systems. For CeCu$_{5.9}$Au$_{0.1}$, closely related to CeCu$_{5.8}$Ag$_{0.2}$, INS has revealed quasi-2D critical fluctuations, while for YbRh$_2$Si$_2$ competing AF and ferromagnetic quantum critical fluctuations have been observed. For these latter two systems, evidence for a locally critical QCP has

Fig. 6. Specific heat as $C/T$ vs. $T$ (on a logarithmic scale) for different CeCu$_{x-Ag_0}$ polycrystals. The inset shows the evolution of the antiferromagnetic phase transition temperature $T_N$ vs. $x$ as derived from specific heat (squares: this study, circles: [33]) and electrical resistivity [32] results.

Fig. 7. Electronic specific heat as $C_{el}/T$ vs. $T$ (left axis) and volume thermal expansion as $\beta/T$ (right axis) vs. $T$ (on a logarithmic scale) for CeCu$_{5.8}$Ag$_{0.2}$ at $B = 0$ [38]. Red solid lines indicate $\log T$ dependences for $C_{el}/T$ and $\beta/T$. The inset shows the temperature dependence of the Grueneisen parameter $\Gamma = \beta_{\text{el}}/\kappa T^2/\beta' C$ with molar volume $V_m = 6.37 \times 10^{-5}\text{ m}^3\text{mol}^{-1}$ and isothermal compressibility $\kappa_T = 1 \times 10^{-11}\text{ Pa}^{-1}$ [37] on a logarithmic temperature scale. The solid red line represents a $\log(T)$ dependence.

6. Summary

To summarize, thermal expansion is more singular than specific heat in the approach of any pressure-sensitive QCP. The critical Grueneisen ratio of thermal expansion to specific heat is the most suitable thermodynamic property to characterize QCPs. The critical Grueneisen ratio diverges with an exponent that is directly related to the symmetry of the critical fluctuations. For the SDW theory, both in 3D and 2D, a $1/T$ divergence of the critical Grueneisen ratio is predicted. We have used a comparative study of the low-temperature thermal expansion and critical Grueneisen ratio to QCPs in the different HF systems. For cubic CeIn$_{3-x}$Sn$_x$ [26] and tetragonal CeNi$_2$Ge$_2$ [29] for which latter system INS measurements revealed 3D low-energy magnetic fluctuations, the critical Grueneisen ratio shows a $1/T$ divergence, in perfect agreement with the SDW scenario. By contrast, for YbRh$_2$(Si$_{0.95}$Ge$_{0.05}$)$_2$ [29] and CeCu$_{5.8}$Ag$_{0.2}$ [38], a divergence with a significantly smaller exponent is observed, incompatible with SDW theory. On the other hand, there are indications for a more complicated structure of the critical magnetic fluctuations in both systems. For CeCu$_{5.9}$Au$_{0.1}$, closely related to CeCu$_{5.8}$Ag$_{0.2}$, INS has revealed quasi-2D critical fluctuations, while for YbRh$_2$Si$_2$ competing AF and ferromagnetic quantum critical fluctuations have been observed. For these latter two systems, evidence for a locally critical QCP has
been found [5,6]. Our comparative study of the Grüneisen ratio divergence may thus suggest that a conventional AF QCP scenario, implying intact Kondo singlets and "composite" HF's at sufficiently low temperatures in the whole phase diagram, requires 3D critical spin-fluctuations.

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