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Thermal expansion and Grüneisen ratio near quantum critical points

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Abstract

Recently it has been shown that at any quantum critical point (QCP) thermal expansion is more singular than specific heat and that the resulting Grüneisen ratio divergence characterizes the nature of the critical fluctuations. Here, we report a comparative study on heavy fermion and d-metal systems. In $Ce_{0.98}Ni_{2+x}Ge_{2-x}$ polycrystals ($0 \leqslant x \leqslant 0.015$) an extreme sensitivity of the ground-state behavior on small changes of composition is observed. For $YbRh_2(Si_{1-x}Ge_x)_2$ the Grüneisen ratio changes sign between the antiferromagnetic and paramagnetic state and diverges with a fractional exponent in the quantum critical regime. In the alloy Ni_xPd_{1-x} the absence of a Grüneisen ratio divergence is incompatible with a ferromagnetic QCP.

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1. Introduction

The strong fluctuations between different ground states close to magnetic quantum critical points (QCPs) in f- and d-electron metals lead to a breakdown of Landau's Fermi liquid (LFL) theory. Recently, it has been shown that dilatometric studies are of particular interest, because thermal expansion, β, probing the pressure dependence of the entropy being accumulated close to the instability, is more singular than specific heat, C [1]. Consequently, the critical Grüneisen ratio $\Gamma^{cr} \sim \beta^{cr}/C^{cr}$, where β^{cr} and C^{cr} denote the critical contributions, must diverge at any QCP being sensitive to pressure.

We have recently observed divergences $\Gamma^{cr} \propto 1/T^x$ in the two quantum critical heavy fermion systems $CeNi_2Ge_2$ and $YbRh_2(Si_{0.95}Ge_{0.05})_2$ [2]. For the Ce-system, $x = 1$ in accordance with the expectation from scaling analysis for a spin-density wave (SDW) QCP [1] has been found. By contrast, the fractional exponent $x = 0.7$ observed in the Yb-system is incompatible with this scenario [2]. Furthermore, a weak logarithmic divergence of $\Gamma(T)$ has been found at the QCP in $CeCu_{6-x}Ag_x$ which excludes an itinerant description for quantum criticality in this latter system as well [3].

In this paper, we report a systematic investigation of different heavy fermion and d-metal systems close to quantum phase transitions by means of thermal expansion and Grüneisen ratio using the technique described in Ref. [2]. In Section 2, new results on slightly off-stoichiometric $Ce_{0.98}Ni_{2+x}Ge_{2-x}$ polycrystals are discussed. As shown in Section 3 for undoped ($x = 0$) and Ge-doped ($x = 0.05$) $YbRh_2(Si_{1-x}Ge_x)_2$, the Grüneisen ratio changes sign between the ordered and quantum critical regime. For the solid solution Ni_xPd_{1-x} a ferromagnetic QCP at $x_c = 0.025$ has been proposed previously [4,5]. In Section 4, we report thermal expansion and Grüneisen ratio measurements on this system.

2. $CeNi_2Ge_2$

$CeNi_2Ge_2$ is a paramagnetic heavy fermion system with a single-ion Kondo scale of $T_K = 30\,K$ [6]. At zero magnetic field, pronounced non-Fermi liquid (NFL) effects have been observed in thermodynamic and electrical

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transport experiments [7] related to a nearby magnetic instability. Indeed, the substitution of Ni with the larger Pd in $Ce(Ni_{1-x}Pd_x)_2Ge_2$ induces long-range antiferromagnetic (AF) ordering below $T_N = 2\,K$ for $x = 0.2$ (Ref. [8]). Furthermore, by applying magnetic fields, a LFL state is induced with a coefficient $A(B)$ derived from the electrical resistivity $\Delta\rho = AT^2$ that diverges for $B \to 0$ [7]. Nevertheless, there are conflicting specific heat results about whether NFL behavior is held 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 [6] and more recent work revealed a cross-over from a logarithmic increase above 0.3 K to a saturation at lower temperatures [9]. 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 [10]. Finally, for the single crystal investigated in Ref. [2], the observed T dependencies of both the thermal expansion and the critical Grüneisen are, down to the lowest measured temperature of 50 mK, in full agreement with the 3D SDW prediction for a system at an AF QCP.

These varying results point to a strong dependence of the physical properties on composition. In order to understand the unclarified nature of the groundstate of $CeNi_2Ge_2$, we performed a systematic study of the thermal expansion $\beta(T)$ on slightly off-stoichiometric $CeNi_2Ge_2$ polycrystalline samples. The chemical composition is described by the formula $Ce_{0.98}Ni_{2+x}Ge_{2-x}$ with $0 \leqslant x \leqslant 0.015$. Because of the particularities of the Ce–Ni–Ge ternary chemical phase diagram [11], the $x = 0$ sample has a Ge-rich 122-phase, whereas in the $x = 0.015$ sample the stoichiometric 122 phase is expected. This is in accordance with the lowest residual resistivity ($\rho_0 \approx 0.16\,\mu\Omega\,cm$) for $x = 0.015$ and enhanced values ($\rho_0 \approx 2$–$3\,\mu\Omega\,cm$) for $x = 0$ and 0.005 samples.

Fig. 1 compares the T dependence of the volume expansion for the three different polycrystals with that of the single crystal studied in Ref. [2]. Above 0.3 K, all samples show a $\beta(T) = a\sqrt{T} + bT$ dependence, expected at a 3D AF QCP [1]. Below 0.3 K, similar as already observed in specific heat measurements on the same polycrystals [12], an extreme sensitivity against small variations of the Ni–Ge composition is observed. In particular, β/T for the $x = 0.015$ crystal passes a sharp cusp around 0.2 K and saturates at lowest temperatures.

To get further information, measurements of the linear thermal expansion have been performed at various magnetic fields (see Fig. 2). For the $x = 0$ crystal, a field-induced LFL behavior $\alpha/T = a_0(B)$ with diverging coefficient $a_0 \sim B^{-0.5}$ is observed. By contrast, more complicated behavior is found for $x = 0.015$. Contrary to the zero-field saturation of α/T, a divergence is observed at 0.5 and 0.6 T. LFL behavior is only observed for $B \geqslant 1\,T$ in agreement with corresponding specific heat measurements [13].

Remarkably, this complicated behavior is found only for the polycrystal with the lowest residual resistivity that

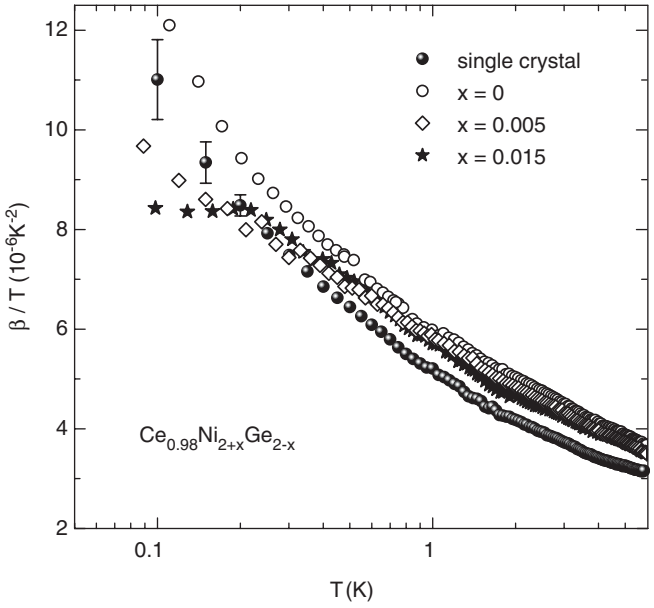

Fig. 1. Volume thermal expansion coefficient for the $CeNi_2Ge_2$ single crystal studied in Ref. [2] as well as various polycrystals with nominal composition $Ce_{0.98}Ni_{2+x}Ge_{2-x}$ as β/T vs. T.

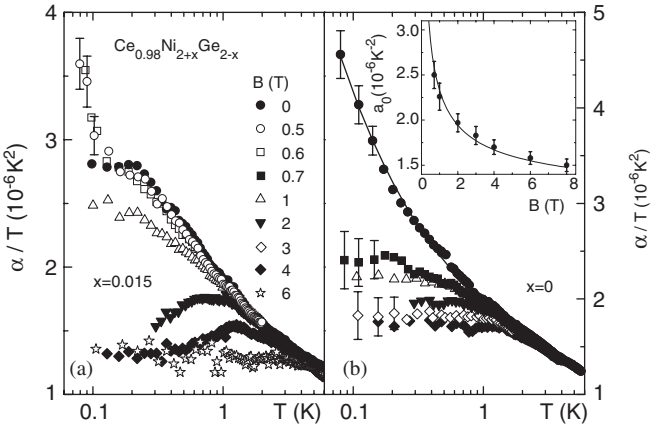

Fig. 2. Linear thermal expansion coefficient for $Ce_{0.98}Ni_{2+x}Ge_{2-x}$ polycrystals (a: $x = 0.15$, b: $x = 0$) at different magnetic fields as α/T vs. T. Line represents $T^{-0.5}$ dependence. Inset in (b) displays field dependence of Fermi liquid coefficient, see text.

should be located closest to the stoichiometric 1:2:2 composition. This raises the question whether the observed behavior might indicate an AF ground state below $T_N \approx 0.2\,K$ in that sample, suppressed by a critical magnetic field of about 0.7 T. Further measurements, in particular on single-crystalline samples are needed for clarification.

3. $YbRh_2(Si_{1-x}Ge_x)_2$

In $YbRh_2Si_2$ pronounced NFL behavior is observed above a weak AF ordering at $T_N = 70\,mK$ [14]. The ordering is further weakened by a tiny volume expansion induced by the substitution of nominally 5 at% of Si with the larger but isoelectronic Ge in $YbRh_2(Si_{0.95}Ge_{0.05})_2$ [15].