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Physica B 359–361 (2005) 41–43
PHYSICA B
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Evidence for a ferromagnetic quantum critical point in $CePd_{1-x}Rh_x$ ☆

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Abstract

In the alloy $CePd_{1-x}Rh_x$, one observes the transition from a ferromagnetic (F) state at $x=0$ to a valence fluctuating state at $x=1$. We are performing a detailed investigation of the disappearance of the F-state in this alloy using AC-susceptibility, specific heat and thermal expansion measurements and present here preliminary results. Our results indicate a continuous decrease of the ferromagnetic ordering temperature T_C with increasing x down to $T_C = 0.25$ K at $x = 0.80$. This suggests that in this alloy the ferromagnetic order disappears at a quantum critical point placed around $x_{cr} \approx 0.85$.

PACS: 71.27.+a; 75.3.Mb; 75.45.+j

Keywords: Quantum critical points; Non-Fermi liquid; Ferromagnetism; Ce compounds

The behavior of Ce-based systems at the critical point, where magnetic order is suppressed by applying pressure or alloying, is presently a subject of great interest. Almost all the current investigations are dedicated to antiferromagnetic critical points [1], while suitable candidates for the study of ferromagnetic critical points (FCP) are very scarce. Nevertheless, a study of a FCP in a Ce-based system would be of high value, because theories predict the magnetic, thermal and transport properties to differ between a FCP and an antiferromagnetic critical point [2].

The study of these critical points are preferentially performed by applying isostatic pressure on pure compounds instead of alloying, because the intrinsic disorder of an alloy induces additional effects which are not well understood. However, in pure compounds an F-state is now suspected to end with a first-order transition disappearing at a classical critical point at finite temperature. On the

☆This work was partially supported by A.v. Humboldt-Foundation (V-8100/B-ARG/1026040) and DAAD-Fundacion Antorchas program Nr.4248-7.
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doi:10.1016/j.physb.2004.12.050

contrary, disorder is suspected to lead to a quantum critical point (QCP) where $T_C(x)$ goes continuously down to $T=0$ K, making alloys more interesting for the study of FCP than pure compounds.

An attractive candidate for the study of a FCP is the alloy $CePd_{1-x}Rh_x$, which was reported to evolve from a $F-Ce^{3+}$ state at $x=0$ (with $T_C=6.6$ K [3]) to a non-magnetic-valence fluctuating one at $x=1$, with a susceptibility maximum at $T\approx 280$ K. This alloy forms a continuous solution with an orthorhombic CrB-type structure. Its cell volume decreases continuously with x showing a deviation from Vegards law at $x\approx 0.6$, while $T_C(x)$ decreases extrapolating to $T=0$ K at $x\approx 0.7$ [3]. Since the disappearance of the F-state in $CePd_{1-x}Rh_x$ has never been thoroughly investigated down to very low temperatures, we started a detailed study of this system by means of AC-susceptibility (χ_{AC}), specific heat (C_m) and thermal expansion (α) measurements and present here first results. Our χ_{AC} results show a further continuous decrease of $T_C(x)$ with increasing x below the minimum $T_C=3$ K value reported in the literature for $x=0.6$ [3]. The F-phase boundary was traced following the temperature of the maximum of the inductive signal $\chi'_{AC}(T)$ in the $x=0.60, 0.65, 0.70$ and 0.75 samples (see Fig. 1). We observe that $T_C(x)$ does not extrapolate to the previously

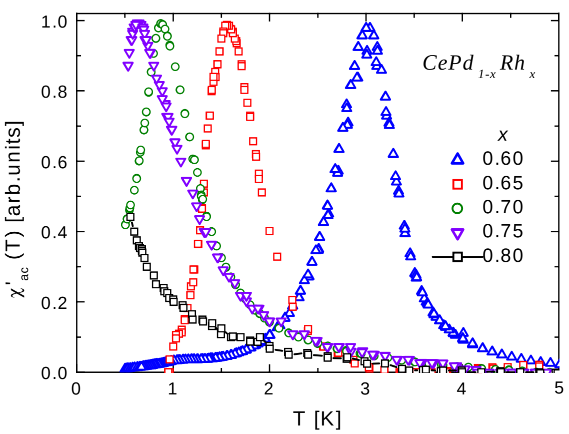

Fig. 1. Inductive signal of χ_{AC} normalized to their respective values at the maximum and at $T=5$ K. For $x=0.8$ the maximum is likely below the temperature limit of the measurement.

reported critical value ($x=0.7$) because its negative curvature changes to slightly positive between $x=0.6$ and 0.65. Our new data suggest a QCP at $x_{cr}\approx 0.85$. Approaching x_{cr}, an anomalous frequency dependence was observed in the dissipative χ''_{AC} response, which (between 1 and 22 KHz) shows a progressive shift of its maximum to higher temperatures while χ'_{AC} is not affected. The sharp maximum in χ'_{AC} show that the F-state is maintained until the critical point. The $C_m(T=T_C)$ anomaly (after phonon subtraction) broadens and decreases significantly for $x\geqslant 0.6$. However, the temperature of its maximum remains very close to that of χ'_{AC}, indicating that it is related to the ordering temperature (see Fig. 2). The maximum of $C_m(T)/T$ (not shown) occurs at lower temperature because of the broadening of the transition, but its magnitude is almost independent of x for $x\geqslant 0.6$ with a value $C_m/T_{max}\approx 1.5$ J/mol K^2 and its width ($\Delta T/T_C$, with ΔT taken at 90% of C_m/T_{max}) does not broaden further. Above T_C, C_m/T develops a $-\mathrm{Ln}\,T$ dependence, in accordance with theoretical predictions for FCP in the spin-density wave scenario [1]. This $-\mathrm{Ln}\,T$ behavior seems to merge above 10 K into a large constant electronic contribution (γ_{HT}), probably related to strong hybridization effects on the first excited crystalline field doublet. In this concentration range the recovery of the $R\,\mathrm{Ln}2$ entropy of the doublet ground state shifts to higher temperatures according to the significant increase of the hybridization strength. Accordingly, an enhanced decrease of the unit cell volume and a drastic increase of the Curie–Weiss temperature [3] form ≈ 30 K at $x=0.6$ up to $\geqslant 300$ K at $x=1$ are observed.

In order to investigate this phase diagram closer to its critical point, we started thermal expansion ($\alpha(T)$) measurements on the $x=0.80$, 0.85 and 0.87 samples. Although these measurements were performed on polycrystals, we found a strong anisotropy, indicating a pronounced texture induced by the cooling process, as well as an intrinsic magnetic anisotropy related to the orthorhombic structure. For $x=0.80$, a well-defined transition was observed at $T_C=0.25$ K in one direction, while in the others $\alpha(T)$ show only a weak temperature dependence. For $x=$

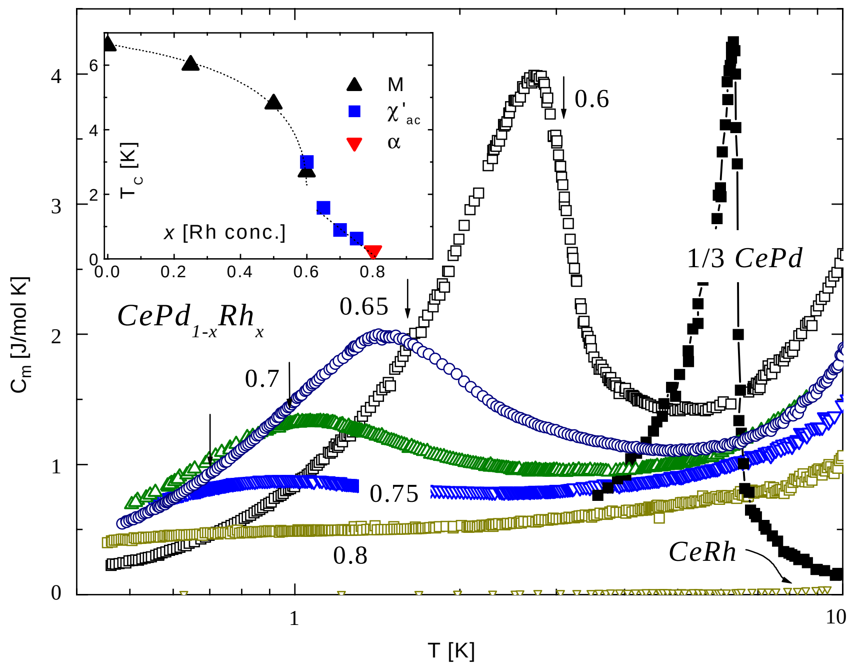

Fig. 2. Evolution of the specific heat anomaly at T_C. The arrows indicate the temperatures of the respective $\chi'(T)$ maxima. Inset: new extended magnetic phase diagram.

0.87, no transition was detected down to 90 mK, though low-energy excitations effects are still observed.

To our knowledge, $CePd_{1-x}Rh_x$ is the first Ce-based system where the decrease of $T_C(x)$ can be followed over more than one decade in T, from 6.6 to 0.25 K. This gives a strong evidence that in this alloy the F-state is disappearing at a QCP. Our results place the critical concentration where $T_C \to 0$ at $x_{cr} \approx 0.85$, significantly above (i.e. $x \approx 0.7$ suggested in previous, less detailed, investigations [3]). The difference is due to a change in the x dependence of the $T_C(x)$ phase boundary at $x \approx 0.65$, likely related to the proximity of a QCP. Such a change was also observed in $CeIn_{3-x}Sn_x$ at a similar temperature, just above an antiferromagnetic QCP [4]. The sharpness of the $\chi'_{AC}(T,x)$ maxima and the nearly constant width of C_m/T_{max} indicate that local inhomogeneities play no relevant role in this system. In summary, these results confirm $CePd_{1-x}Rh_x$ as a system proper for the experimental study of a ferromagnetic quantum critical point.

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