Smeared Ferromagnetic Quantum Phase Transition in $CePd_{1-x}Rh_x$

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The phenomena occurring at the disappearance of a magnetically ordered state upon tuning a non-thermal parameter like pressure, composition, or magnetic field are presently one of the central topics in the field of strongly correlated electron systems. Of strongest interest are compounds in which the transition temperature decreases continuously down to $T = 0$ K, leading to a quantum critical point (QCP). Among Ce-based compounds, there is a huge number of systems in which the disappearance of an antiferromagnetic state can be investigated, while systems appropriate for the study of the disappearance of a ferromagnetic state are extremely rare: At the beginning of our project, there was no good example for the continuous disappearance of a ferromagnetic state in a Ce-based compound [1]. From theoretical considerations, one expects strong differences between a ferromagnetic and an antiferromagnetic critical point. Thus, finding an example for a ferromagnetic critical point in a Ce-based system is a very challenging task. Upon screening appropriate compounds, we realized that the alloy $CePd_{1-x}Rh_x$ is a good candidate. While CePd is one of the few ferromagnetic Ce-based compounds with an ordering temperature $T_c = 6.6$ K, CeRh is a non-magnetic intermediate valence system, with a characteristic $4f$ valence fluctuation energy of the order of 300 K [2]. An earlier investigation of the $CePd_{1-x}Rh_x$ alloy showed a continuous decrease of T_c with increasing x, down to $T_c = 2.7$ K at $x = 0.6$, and no magnetic order above 0.5 K for $x \geq 0.8$ [2]. However, the region where T_c disappears was not thoroughly investigated. Alloys are usually considered to be less attractive for the investigation of QCP's, because the inherent disorder might mask some of the interesting properties or even inhibit interesting phenomena like, e.g., the onset of unconventional superconductivity. However, in the case of a ferromagnetic system, some disorder might be a prerequisite for the observation of a quantum critical point [3]. Because in a pure system without disorder the ferromagnetic state is suspected to vanish with a first order transition line ending in a classical critical point at finite temperature [4].

We therefore decided to look in more detail at $CePd_{1-x}Rh_x$ in the context of a DAAD-Antorchas collaboration project with the Centro Atomico Bariloche (CAB) in Argentina. A large number of polycrystalline samples with different Rh-contents were prepared and structurally characterized at the MPI-CPfS. The specific heat $C(T)$ above 0.4 K, the dc-susceptibility $\chi_{dc}(T)$ above 2 K and the ac-susceptibility $\chi_{ac}(T)$ above 0.4 K were measured at the CAB. Additional measurements of the thermal expansion and of the ac-susceptibility down to very low temperatures, 20 mK, were performed at the MPI-CPfS on samples close to the critical regime.

The results meet our expectations [5]. In particular, the ferromagnetic state does not switch to an antiferromagnetic one before the disappearance of magnetic order, which was the major problem encountered in other ferromagnetic Ce-based systems tuned to the critical point. In $CePd_{1-x}Rh_x$, the magnetic order keeps its ferromagnetic signature until it disappears. The evidence stems from the ac-susceptibility measurements (Fig. 1). Here, one expects a large and sharp maximum at a ferromag-

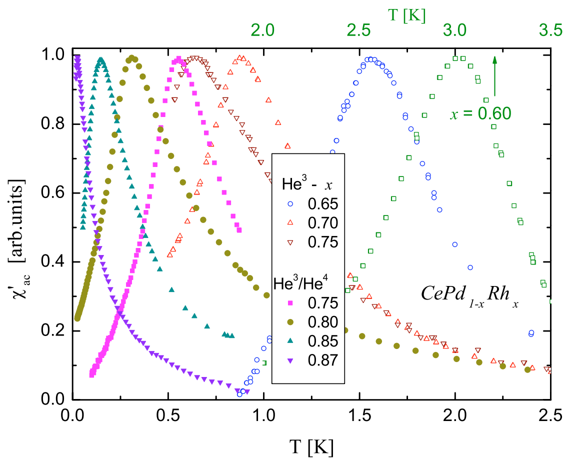

Fig. 1: Temperature dependence of the ac-susceptibility of $CePd_{1-x}Rh_x$ for samples in the region $0.6 < x \leq 0.87$, showing the peak in χ_{ac} shifting to lower temperature with increasing Rh content. All the curves were scaled with respect to the maximum value of χ_{ac}. Note the shifted upper temperature scale for $x = 0.6$. Samples with $x \leq 0.75$ were measured in a He^3 cryostat, while samples with $x \geq 0.75$ were measured in a He^3/He^4 dilution refrigerator.

netic transition, while antiferromagnetic ordering would lead to an extremely weak signature only. A large maximum was observed in all investigated samples with $x < 0.9$. The temperature of the maximum, which can be taken as the ordering temperature, shifts continuously to lower temperatures with increasing Rh content, down to $T_c = 24$ mK at $x = 0.87$. No maximum is visible above 18 mK in samples with $x \geq 0.9$ for which other types of magnetic order can be excluded from the overall behavior.

Our main results are collected in the phase diagram presented in Fig. 2. It shows the transition temperatures T_c deduced from different kind of measurements as a function of composition. The inset shows an enlarged view of the critical region where $T_c \to 0$ K. From $x = 0$ up to $x = 0.6$, T_c decreases rather smoothly, with a negative curvature as usually expected. However, for $x > 0.6$ the curvature becomes positive leading to a long tail towards higher Rh contents. At the end of this tail, T_c seems to decrease very abruptly. It is presently not clear whether this abrupt decrease corresponds to a first order transition (as generally expected for a ferromagnet in the absence of disorder), to the break up of percolation of magnetically ordered domains related to a distribution of local Kondo temperatures (see below), or to experimental limitations due to slight variations of the real Rh-content in the measured samples. This decrease of T_c over more than two decades in temperature, from 6.6 K at $x = 0$ to 24 mK at $x = 0.87$, is presently by far the best example for the continuous disappearance of ferromagnetic order in a Ce-based system.

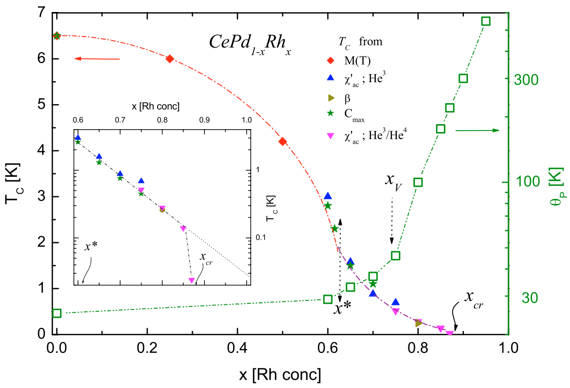

Fig. 2: Magnetic phase diagram of $CePd_{1-x}Rh_x$: Composition dependence of the ordering temperature T_c deduced from different kinds of measurements (left scale), and of the Weiss temperature θ_p (right scale). The inset shows an enlarged view of $T_c(x)$ in the critical region.

At $x \cong 0.62$, where $T_c(x)$ changes its curvature, the Kondo temperature T_K, which reflects the hybridization of the f electrons with conduction electrons, starts to increase dramatically. This is evidenced in Fig. 2 by the composition dependence of the Weiss temperature θ_p obtained from fits of $1/\chi_{dc}(T)$ versus T at high temperatures. In a first approximation θ_p is proportional to T_K. θ_p increases by only 15 % between $x = 0$ and $x = 0.6$, while between $x = 0.6$ and 0.75, the increase amounts to 60%, and to one order of magnitude between $x = 0.75$ and 0.95. The same result can be inferred from the evolution of the entropy and of the lattice parameters. The most relevant parameter governing T_K is the hybridization of the 4f electron of a Ce-atom with the valence electrons of the surrounding ligands. In Ce-based compounds, Rh ligands are known to lead to much larger T_K than Pd ligands. Thus, it seems that in $CePd_{1-x}Rh_x$ — above a critical number of Rh neighbors — the effect of the Rh-ligands strongly overcomes the effect of the Pd-ligands.

This could explain a further result of our χ_{ac} measurements. For $x \geq 0.7$, we observe a change in the behavior of χ_{ac}: The position of the maximum becomes frequency dependent: it shifts to higher temperature with increasing frequency of the excitation field, which is an evidence for spin-glass-like behavior. This indicates that in this critical region the order is not anymore of true long-range kind, but has a finite correlation length. We suspect that in this region the very rapid increase of the Kondo scale with Rh content leads to a local variation of T_K, each Ce atom having a different T_K related to the number of its Pd and Rh first neighbors. Then the strength of the Kondo screening is different on each Ce atom, which should lead to some regions where the moments are still rather stable and thus keep a strong magnetic character, while in other regions the moments are already fully screened and thus have lost their magnetic character. This is supported by the presence of a Curie-Weiss tail in $\chi_{dc}(T)$ at low T, whose effective moment is strongly decreasing with increasing x for $x > 0.6$, and by the temperature dependence of the specific heat and the entropy, which indicated a very broad distribution of energy scales for $x > 0.7$. Such a distribution of local Kondo temperatures with magnetic and non- magnetic regions should lead to some kind of cluster glass (Kondo-cluster glass) which can easily explain the frequency dependence observed in χ_{ac}.

This Kondo-cluster glass may explain why the phase-transition anomaly in the specific heat $C(T)$ gets strongly broadened for $x > 0.6$. For $x > 0.75$ the maximum in the specific heat even disappears completely, despite the presence of a sharp maximum in χ_{ac}, and is replaced by a logarithmic divergence of C/T towards low temperatures as observed in many other systems close to a quantum critical point. Preliminary measurements at very low temperatures showed some unusual behavior in the thermodynamic properties [6]. Thus, for $x \geq 0.87$, once magnetic order is completely suppressed, the logarithmic divergence in C/T is replaced by a power law divergence $C/T \propto T^n$ with n close to -0.5. The same power law is also observed in the thermal expansion $\beta(T)/T$. As a result the Grüneisen parameter Γ, i.e. the ratio between the thermal expansion and the specific heat $\Gamma(T) \propto \beta(T)/C(T)$, becomes temperature independent, in contrast to the divergence observed in other quantum critical systems. Further on, the concentration dependence of Γ is also very unusual. In all Ce-based compounds investigated up to now, Γ changes its sign at the antiferromagnetic quantum critical point and becomes positive on the non-magnetic side, with a magnitude decreasing with increasing distance from the quantum critical point. In contrast, in $CePd_{1-x}Rh_x$, Γ becomes negative at the disappearance of magnetic order at $x = 0.87$ and its absolute value still increases from $x = 0.87$ to $x = 0.9$. Presently we are investigating and analyzing these unusual low temperature properties in more detail. One of the questions is, e.g., whether these observations can be accounted for by the Griffiths phase scenario, one of the theoretical scenarios for a disordered quantum phase transition [7]. It predicts e.g. a power law in the specific heat, as we observed in $CePd_{1-x}Rh_x$. In addition, we have started the growth of single crystals in order to investigate and take into account the expected strong anisotropy of this orthorhombic compound [8].

In summary, we have found a Ce-based system in which a ferromagnetic transition can be continuously tuned to $T_c \rightarrow 0$ K. In the ac-susceptibility of $CePd_{1-x}Rh_x$, T_c can be traced over more than two decades in temperatures, from $T_c = 6.6$ K at $x = 0$ to $T_c = 24$ mK at $x = 0.87$. Thus, this compound presents a unique opportunity to study the continuous suppression of a ferromagnetic state. For $x > 0.6$ the maximum in the ac-susceptibility starts to become frequency dependent, which suggests the formation of a Kondo-cluster glass. This might be due to a distribution of local Kondo temperatures related to a huge increase of the hybridization between 4f and valence electrons with increasing Rh content for $x > 0.6$. At and beyond the disappearance of magnetic order, we found a power law divergence of the specific heat and of the thermal expansion, $C/T \propto \beta/T \propto T^n$, with an exponent $n \cong -0.5$. The temperature and concentration dependence of the Grüneisen parameter $\Gamma(T) = \beta(T)/C(T)$ is in contradiction with the results observed at quantum critical points in other Ce-based systems. We are presently investigating the unusual low temperature properties of this system in more details in order to develop a phenomenological understanding.

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