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Peculiar quantum criticality in ferromagnetic $CePd_{1-x}Rh_x$

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Abstract

The alloy $CePd_{1-x}Rh_x$ is a unique example undergoing quantum critical behavior between ferromagnetic CePd ($T_C = 6.6\,K$) and mixed valence CeRh. The negative curvature of $T_C(x)$ holds up to $x = 0.6$, followed by a positive *tail* with the lowest measured $T_C = 0.25\,K$ at $x = 0.80$. We report on a detailed investigation within the $0.80 \leqslant x \leqslant 1$ range, using specific heat (C_m) and thermal expansion (β) techniques. A change of regime is observed in the T dependence of both parameters at $x_{cr} = 0.85$, from $\ln(T_0/T)$ to $A(x)T^{-q}$, with $q = 0.54$ at $x = 0.87$ and the coefficient $A(x)$ vanishing as $x \to 1$. Simultaneously, a change of sign in $\beta(T)$ is observed at x_{cr}. The vanishing $C_m/T = A(x)T^{-q}$ contribution coexists beyond x_{cr} with a Fermi Liquid component.

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Ferromagnetic (F) quantum phase transitions are being investigated in a number of itinerant stoichiometric compounds, where the critical conditions are tuned by applying pressure. In these compounds, namely MnSi, UGe_2 and $ZrZn_2$ [1], the second-order F-transition reveals notable differences from the classical behavior. Notably, the phase boundary disappears at finite temperature in a first-order transition. Although present theories predict a first-order quantum critical (QC) point in pure itinerant systems [2], disorder is expected to induce smeared QC effects, extending the second-order phase boundary to lower temperatures. This allows to trace the F-transition closer to the critical point, where the competition between thermal and non-thermal fluctuations arise novel behaviors.

F-$CePd_{1-x}Rh_x$ gives the opportunity to investigate the QC region by tuning the concentration of alloyed Ce-ligands. In this case, the driving force is the *chemical potential* rather than *chemical pressure*, since the main difference between Pd and Rh is one 4d-band electron rather than the atomic size (≈ 2%).

This system can be continuously driven from a F-ground state in CePd (with $T_C = 6.6\,K$), to a mixed valence state in CeRh. Its Curie temperature, $T_C(x)$, was traced in more than one decade down to $T_C = 0.25\,K$ at $x = 0.80$ [3] showing that the classical negative curvature of $T_C(x)$ holds up to $x = 0.60$. Beyond that concentration a positive curved *tail* sets on, with the critical concentration (where $T_C(x) \to 0$) extrapolated to $x_{cr} = 0.85$.

Previous analysis on the specific heat T dependence (C_m/T) showed that a $-\ln T$ dependence develops as $x \to x_{cr}$, in accordance with theoretical predictions for a 3D F-spin-density wave scenario [4]. Above 10 K this $-\ln T$ dependence merges into the electronic (γ_0) contribution, enhanced by the strongly hybridized excited crystal field levels, in coincidence with the rapid increase of the Curie–Weiss temperature for $x > 0.70$.

Owing to the positive curvature of T_C ($x > 0.6$) *tail*, its extrapolation to $T_C = 0$ tends to be asymptotic. Hence, a detailed investigation at, and beyond x_{cr}, is required in order to determine how the expected Fermi liquid (FL) phase arises in a vanishing magnetic medium. In this paper, we report the results of such a study performed within the $0.80 \leqslant x \leqslant 1$ range, using C_m and thermal expansion (β) techniques.

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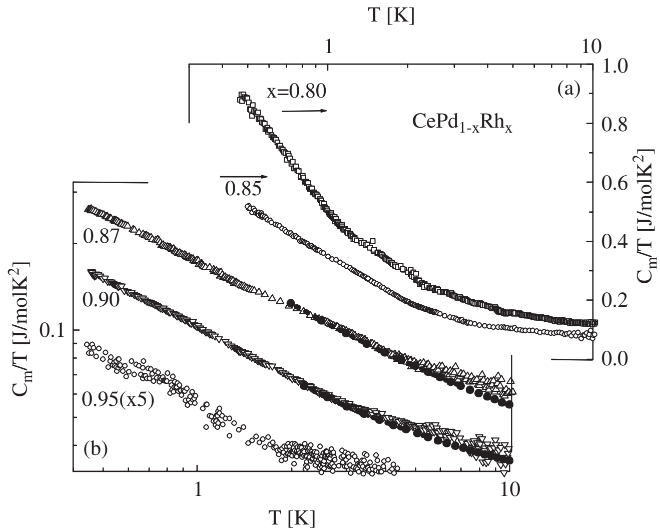

Fig. 1. (a) Logarithmic T dependence of specific heat and (b) in a double logarithmic scale. For $x = 0.90$, $\gamma_0 = 0.028\,J/mol\,K^2$ is subtracted, see the text. Results of χ ($T > 2\,K$) from $x = 0.87$ and 0.90 (•) are included for comparison {in arbitrary units}.

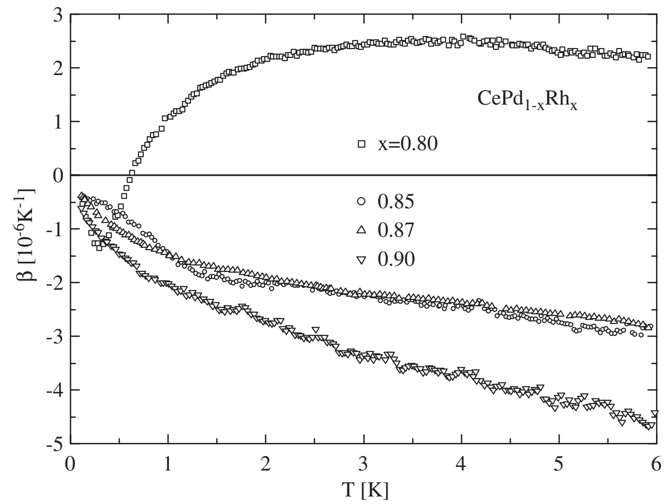

Fig. 2. Temperature dependence of volume thermal expansion.

The most significant feature at $x = x_{cr}$ is the C_m/T vs. T dependence change from a $\propto -\ln T$ to a power law: $A(x)T^{-q}$, for $x \geqslant 0.87$. In Fig. 1 we compare those behaviors in a simple logarithmic representation for $x \leqslant 0.85$ (Fig. 1a) and a double logarithmic one for $x \geqslant 0.87$ (Fig. 1b). From the latter we extract an exponent $q = 0.54 \pm 0.01$. Since $A(x)$ decreases asymptotically as $\propto -\log(b \times x)$, a remnant contribution is still observed in CeRh. Such a vanishing contribution coexists with a FL one, recognized from the $\gamma_0 = 0.028$ and $0.014\,J/mol\,K^2$ values for $x = 0.95$ and 1, respectively. The latter value is in perfect agreement with the Wilson ratio (χ_0/γ_0) for a six-fold mixed valence ground state (with $J = 5/2$) since for CeRh $\chi_0 = 4.6 \times 10^{-4}$ emu/mol. The $\chi(T, x)$ results also exhibit anomalous departures from CW-law at high T with fractional exponents (≈ 0.8) around x_{cr}. At low T, a weak, but power law, contribution is observed with similar exponent to that of C_m/T (see bullets in Fig. 1b). Though the lack of saturation at high fields excludes an impurity origin, measurements below 2 K are required to confirm this behavior. Electrical resistivity (ρ) confirms the presence of a remnant magnetic contribution at low temperature since $\rho(T)$ deviates from the FL $-\rho \propto T^2$ dependence below 20 K.

The change from a $C_m/T \propto -\ln T$ dependence to a T^{-q} one at $x = 0.87$ cannot be attributed to a sudden increase of atomic disorder. Its limited influence is evidenced by the sharp $\chi'_{ac}(T, x)$ maxima and C_m/T_{max} widths [3]. Further indications in that sense is given by $\rho(T)$. This system shows the maximum of its residual resistivity $\rho(T \to 0)$ at $x \approx 0.65$, where the valence instability sets on [5] instead of at $x = 0.5$ as expected from Nurdheim's rule. In fact, the RRR factor ($\rho_{300\,K}/\rho_0$) grows more than one decade between $x = 0.80$ and CeRh.

Similar evolution is shown by the thermal expansion, β, see Fig. 2. The sharp minimum of $\beta(T)$ at 0.25 K in the $x = 0.80$ sample marks the lowest, but still well defined, T_C value. Together with a change of sign, a non-monotonous T dependence is observed in sample $x = 0.85$. This reveals a competition between different effects, which may occur in different directions of this strongly anisotropic structure. Even these samples being polycrystalline, perpendicular linear thermal expansion components show different T dependencies [6]. At $x = 0.87$, a $\beta \propto -T \ln T$ dependence is observed up to about 2 K, whereas at $x = 0.90$ it coincides with the $C_m(T)$ power-law dependence. This makes the β/C_m ratio weakly temperature dependent but growing between $x = 0.80$ and 0.90.

These thermodynamical properties of $CePd_{1-x}Rh_x$ around x_{cr} show that, despite the changes in the T dependence of C_m and β, the QC region extends beyond x_{cr} and coexist with a FL contribution from the mixed valence component.

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