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Searching for a Quantum Critical Point in Rh doped ferromagnetic $Ce_{2.15}Pd_{1.95}In_{0.9}$

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Abstract.

Low temperature magnetic and thermal (C_m) properties of the ferromagnetic (FM) alloys $Ce_{2.15}(Pd_{1-x}Rh_x)_{1.95}In_{0.9}$ were investigated in order to explore the possibility for tuning towards a quantum critical point (QCP) by doping Pd with Rh. As expected, the magnetic transition decreases from $T = 4.1$ K at $x = 0$ with increasing Rh concentration. However, the phase boundary splits into two transitions, the upper being antiferromagnetic (AF) whereas the lower FM. The AF phase boundary extrapolates to $T_N = 0$ for $x_{cr} \approx 0.65$ whereas the first order FM transition vanishes at $x \approx 0.3$. The quantum critical character of the $T_N \to 0$ point is inferred from the divergent T dependence of the tail of C_m/T observed in the $x = 0.5$ and 0.55 alloys, and the tendency to saturation of the maximum of $C_m(T_N)/T$ currently observed in exemplary Ce compounds when $T_N \to 0$. Beyond the critical concentration the unit cell volume deviates from the Vegard's law in coincidence with a strong increase of the Kondo temperature.

1. Introduction

The $Ce_{2\pm t}Pd_{2\mp u}In_{1-v}$ family of alloys shows an extended range of solid solution [1] with a peculiar magnetic behavior since the Ce-rich branch ($t > 0 > u$) behaves ferromagnetic (FM) whereas the Pd-rich ($t > 0 > u$) is antiferromagnetic (AF). Such a difference of magnetic structure under a slight variation of the alloy composition indicates that the energies of both phases are very similar. Another evidence for the competition between magnetic structures in this type of compounds is given by Ce_2Pd_2Sn [2], which shows a competition between ferro- and antiferromagnetic phases with a finite temperature critical point tuned by $25\% Ni$ doping. Consequently this family of alloys are appropriate materials for testing the stability of exotic order parameters.

For this work, we have exploited the FM character of the $Ce_{2.15}Pd_{1.95}In_{0.9}$ composition to search for a quantum critical point (QCP) [3] by tuning the chemical potential through the doping of the Pd lattice with Rh, like in the previously studied pseudo-binary compound $CePd_{1-x}Rh_x$ [4]. For such a purpose, we have investigated the low temperature properties through magnetic (M) and specific heat (C_P) measurements performed on the Rh doped $Ce_{2.15}(Pd_{1-x}Rh_x)_{1.95}In_{0.9}$.

2. Experimental and results

This alloyed system forms continuously all along the complete Pd/Rh concentration range with the Mo_2B_2Fe type crystalline structure. The samples were prepared using a standard arc melting

procedure under an argon atmosphere, and remelted several times to ensure good homogeneity. The range of concentration investigated covers up to 50% of the total Rh by Pd substitution and sample characterization by EPMA and X-ray diffraction indicates that a small percentage of $Ce(Pd_{1-x}Rh_x)$ with x composition slightly higher than the bulk is present.

As reported in Ref. [1] the Ce-plane, at $z = 0.5$, contains Ce atoms placed in the crystallographic site $4h$ whereas Ce atoms exceeding stoichiometric concentration (i.e. $u = 0.15$) replace In atoms at the $2a$ site at the $z = 0$ and 1 planes. Concerning structural properties, one observes that the volume of the unit cell decreases with Rh content following Vegard's law up to $x = 0.3$. The main structural variation as a function of concentration is observed along the c-axis.

The low temperature behavior was investigated through thermal and magnetic parameters. Concerning specific heat measurements, in Fig. 1 we present the thermal dependence of the magnetic contribution divided by temperature (C_m/T), after phonon subtraction extracted from the non-magnetic La isotypic reference compound.

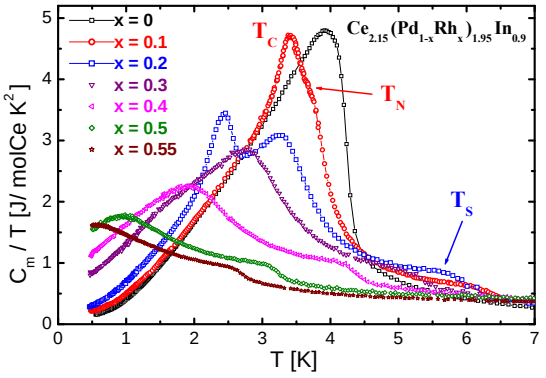

Figure 1. (Color on line) Thermal and concentration dependence of the magnetic contribution to specific heat divided temperature. T_C and T_N represent respective FM and AF transitions whereas T_S identifies a concentration dependent satellite transition discussed in the text.

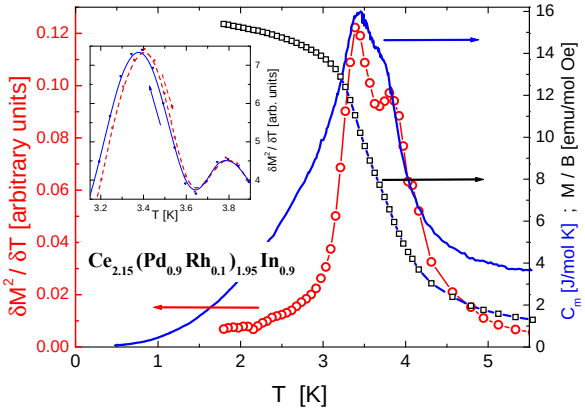

Figure 2. (Color on line) Comparison between thermal derivative of $M^2(T)$ (left axis) with $C_m(T)$ (right axis) showing the split between FM and AF transitions. The original $M(T)$ dependence is also included (right axis). Inset: hysteresis of $\partial M^2/\partial T$ between heating and cooling procedures.

As it can be seen in Fig. 1, the magnetic transition in the parent compound ($x = 0$) shows the characteristic C_m/T jump of second order character. However, a slight broadening at the maximum of C_m/T can be observed. This broadening was formerly interpreted as an intrinsic experimental effect [1]. The present results indicate that the sample at x = 0 is actually close to a bi-critical point at $T_{cr} = 4.1$ K since the phase boundary splits into two transitions by Rh doping. Similar feature was observed in the intensively studied $CeRu_2(Ge_{1-x}Si_x)_2$ system [5]. Also in Fig. 1 a Rh concentration dependent satellite anomaly ($T_S(x)$) is observed. The intrinsic character of this satellite transition is discussed below.

Magnetic $M(T)$ measurements on the $x = 0$ sample do not improve the identification of the bi-critical point because the FM signal largely exceeds the underlying AF cusp. The presence of this bi-critical point is not surprising if we consider the mentioned study of $Ce_{2\pm t}Pd_{2\mp u}In_{1-v}$ solid solutions [6]. From that study, one concludes that $T_{cr} = 4.1$ K, lies in the extrapolated values of $T_N(u)$ from the Pd-rich AF samples (i.e. $Pd \geq 2 + u$) to the Ce-rich FM-branch

(i.e. $Pd \geq 2-u$). The same coincidence is observed for the paramagnetic temperature $\theta_P(u)$ extrapolated to $Pd = 1.95$ from the $Pd \geq 2$ side.

In the following we will analyze the evolution of the low temperature properties under further increase of Rh content in $Ce_{2.15}(Pd_{1-x}Rh_x)_{1.95}In_{0.9}$. The splitting between $T_N(x)$ and $T_C(x)$ becomes more pronounced in the thermal and magnetic properties of sample $x = 0.1$. That feature is observed as an incipient structure in the cusp of C_m/T shown in Fig. 1 and Fig. 2. A further evidence for the splitting of both phase boundaries is obtained from $M(T)$ measurements. Taking into account that from thermodynamic properties the internal magnetic energy U_m of a FM phase is related to the spontaneous magnetization, i.e. $U_m \propto M^2$, its temperature derivative $\partial M^2/\partial T$ is proportional to $C_m(T)$ [7]. In Fig. 2 we compare both parameters (left axis for $\partial M^2/\partial T$ and right axis for $C_m(T)$) showing that the mentioned structure at the transition is better defined by a $\partial M^2/\partial T$ versus T dependence. The same features are observed in the $x = 0.2$ sample since the transitions increase their thermal difference ($T_N = 3.3\,\mathrm{K}$ and $T_C = 2.5\,\mathrm{K}$) as observed in Fig. 1 from C_m/T results. The first order character of the (lower) T_C transition is evidenced by an hysteresis in $\partial M^2/\partial T$ at the lower transition which shows a shift between heating or cooling procedures as depicted in the inset of Fig. 2. The first order character of the lower transition is in agreement with the fact that the AF order parameter developing from T_N suddenly changes to an FM structure at T_C.

Also from Fig. 1, a change of the nature of both transitions can be appreciated for $x \geq 0.3$. While $T_C(x)$ tends to vanish becoming a weak shoulder at $x = 0.3$, the $C_m(T_N)$ jump transforms into a cusp at that concentration and then into a broad maximum for $x \geq 0.4$ centered at $T_{max} = 1.8\,\mathrm{K}$ ($x = 0.4$), $0.9\,\mathrm{K}$ ($x = 0.5$) and $0.5\,\mathrm{K}$ ($x = 0.55$) respectively. Those maxima are followed at higher temperature by a large tail denouncing non-Fermi-liquid behavior [3]. Coincidentally, the value of the C_m/T maxima decrease, extrapolating to $\approx 1.4\,\mathrm{J/molK^2}$ for $T_{max} \to 0$ (see Fig. 1) as currently observed in the proximity of a QCP [8]. Particularly, sample $x = 0.55$ follows a modified power law divergency $C_m/T \propto 1/(T^{1.25}+1)$ [9] once the T_S anomaly contribution is subtracted.

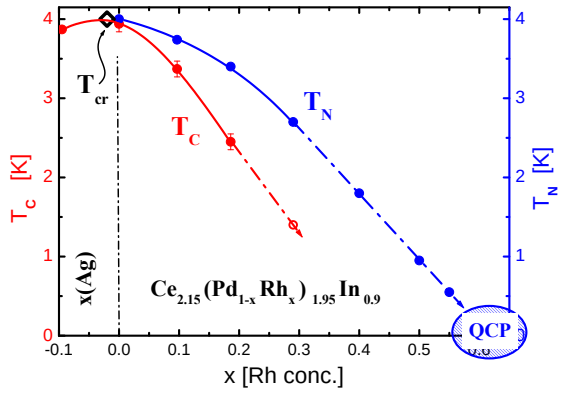

Figure 3. (Color on line) Magnetic phase diagram showing $T_N(x)$ and $T_C(x)$ phase boundaries decrease. Hollow circle in $T_C(x$=0.3) corresponds to the kink in $C_m(T)/T$ of Fig. 1. Negative x values correspond to Ag doping and hollow symbol at $Ag(x$=-0.03) to the bi-critical point.

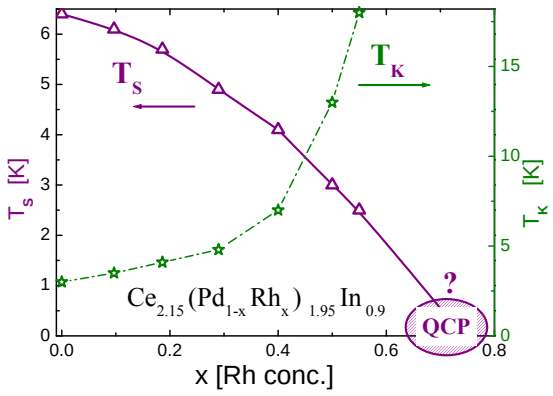

Figure 4. (Color on line) Rh concentration dependence of the satellite anomaly T_S and the $T_K(x)$ increase (right axis).

Concerning the satellite transition at $T_S(x)$, it can be mapped out to the T_C transition of

the pseudo-binary compound $CePd_{1-x}Rh_x$ [4] at slightly higher ($\approx 10\%$) *Rh* concentration. The relevant features observed in this anomaly are the following: i) the height of the $C_m(T_S)$ contribution and its associated entropy corresponds to about 7% of a $CePd_{1-x}Rh_x$ phase; ii) in samples with $x \geq 0.4$, $C_m(x, T = T_S)$ nicely coincide once represented as a function of a normalized temperature $t = T/T_S$. These characteristics do not coincide with random contributions expected from spurious phases. Preliminary magnetic measurements indicate that: iii) the T_S anomaly is suppressed by the application of a moderate magnetic field ($B \approx 0.1\,\mathrm{T}$). Such a rapid suppression is in contrast with the magnetic behavior of pure $CePd_{1-x}Rh_x$ because that compound shows a robust FM phase [4]. Coincidentally, iv) the FM signal observed around $M(T_S)$ is significantly weaker that the expected from the binary compound. All these features raise the question whether the origin of the T_S anomaly is due to a spurious $CePd_{1-x}Rh_x$ phase or to the intrinsic excess of Ce atoms placed at the '2a' crystalline sites [1], with an atomic environment mimicking that of $CePd_{1-x}Rh_x$.

3. Magnetic Phase Diagrams

In Fig. 3 we present the phase diagram showing that the upper (T_N) and the lower (T_C) transitions converge into a bi-critical point at $x \to 0$. Both boundaries are well defined for $x \leq 0.2$, but they broaden and smear respectively for $x \geq 0.3$. Since Rh doping effect is expected to introduce *holes* into the conduction band, in order to confirm the existence of that critical point we have tested the possibility to move on the other direction, i.e. introducing more *electrons* into the band. For such a purpose we investigated the Ag doped alloy $Ce_{2.15}(Pd_{0.90}Ag_{0.10})_{1.95}In_{0.9}$, whose preliminary thermal and magnetic properties indicate a clear FM behavior with the consequent disappearance on the AF phase at the bi-critical point, estimated at Ag≈ 0.03 and $T \approx 4\,\mathrm{K}$ as indicated in Fig. 3. The FM character of the $T_C(x)$ phase boundary is proved by $M(T)$ measurements on respective samples.

Fig. 4 presents the concentration dependence of the satellite $T_S(x)$ anomaly with its extrapolation to zero at $x \approx 0.75$, and the Kondo temperature $T_K(x)$ evaluated using the Desgranges-Schotte criterion [10]. According to this criterion, T_K can be computed as the temperature at which the entropy reaches the value $S(T_K) = 2/3RLn2$. It is worth noting that the rapid increase of $T_K(x)$ coincides with the extrapolation of $T_N(x)$ to the quantum critical point and the deviation of $V(x)$ from the Vegard's law. On the contrary, the degrees of freedom involved in the $T_S(x)$ anomaly seems not to be affected by Rh increase at least up to $x = 0.55$, see Fig. 1.

Further studies at higher Rh concentration are in progress to better determine $T_S(x)$ at lower temperature and its dependence on magnetic field in elucidate the origin of that anomaly. Simultaneously, other $Ce_{2.15}(Pd_{1-x}Ag_x)_{1.95}In_{0.9}$ alloys are being investigated to better tune the electronic concentration on the bi-critical point.

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