Approaching Quantum Criticality in ferromagnetic Ce$_{2}$(Pd$_{1-x}$Rh$_{x}$)$_{2}$In alloys

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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 a quantum critical point (QCP) by doping Pd with Rh. As expected, the magnetic transition observed at $T = 4.1$ K in the parent alloy decreases with increasing Rh concentration. Nevertheless it 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 QC character of the $T_N \rightarrow 0$ point arises 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 $C_m(T_N)/T$ as observed in exemplary Ce compounds for $T_N \rightarrow 0$. Beyond the critical concentration the unit cell volume deviates from the Vegard’s law in coincidence with a strong increase of the Kondo temperature.

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I. INTRODUCTION

The Ce$_{2+x}$Pd$_{2+y}$In$_{1-z}$ family of alloys shows an extended range of solid solution [1] with a peculiar magnetic behavior since the Ce-rich branch ($u > 0 > y$) behaves ferromagnetic (FM) whereas the Pd-rich ($y > 0 > u$) is antiferromagnetic (AF). Such a difference of magnetic structure under a small 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 Yb$_2$Pd$_2$In$_{1-x}$Sn$_x$ [2], which orders magnetically at intermediate $x$ values despite stoichiometric limits are not magnetic. Consequently this family of alloys are appropriated materials for testing the stability of exotic order parameters.

For this work, we have profited from the FM character of the Ce$_{2.15}$Pd$_{1.95}$In$_{0.9}$ composition to search for a quantum critical point (QCP) [3][4] by tuning the chemical potential through the doping of the Pd lattice with Rh, like in the previously studied pseudobinary compound CePd$_{1-x}$Rh$_x$ [5]. 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$_{0.9}$.

II. EXPERIMENTAL AND RESULTS

This alloyed system forms continuously all along the complete Pd/Rh concentration range with the Mo$_2$B$_2$Fe 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 in this work covers more than 50% of the total Rh by Pd substitution, i.e. $0 \leq x \leq 0.55$. From structural properties, one observes that the volume of the unit cell decreases with Rh content following a Vegard’s law up to $x = 0.3$. Beyond that concentration the volume decreases faster indicating that hybridization effect become relevant. The main change observed as a function of concentration occurs along the c-
axis.

The low temperature behavior was investigated on thermal and magnetic properties. Concerning $C_p(T)$ measurements, in Fig. 1 we present the thermal dependence of the magnetic contribution divided by temperature ($C_m/T$), after phonon subtraction for different concentrations extracted from the non-magnetic La isotypic reference compound. 

In the following we will analyze the evolution of the low temperature properties for further increase of the Rh content. The splitting between $T_N$ and $T_C$ becomes more clear in the thermal and magnetic results from the Ce$_{2.15}$(Pd$_{1-x}$Rh$_x$)$_{1.95}$In$_{0.9}$ sample at $x = 0.1$. That feature is observed as an incipient structure in the cusp of $C_m/T$, see Fig. 1 and Fig. 2. The first order character of the (lower) $T_C$ transition is evidenced by an hysteresis at the $C_m(T)$ transition which shows a shift between heating or cooling procedures as depicted in the inset of 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 $C_m(T)$ [8]. 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 ($T_N = 3.3$ K and $T_C = 2.5$ K) as observed in Fig. 1.

As it can be appreciated in Fig. 1 the nature of both transitions changes 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 1.8 K ($x = 0.4$), 0.9 K ($x = 0.5$) and 0.5 K ($x = 0.55$) respectively. Those maxima are followed at higher temperature by a large tail resembling the non-Fermi-liquid behavior. Coincidentally, the value of the $C_m/T$ maxima decrease and tends to saturate at around 1.5 J/molK$^2$ (see Fig. 1) as usually observed at the proximity of a QCP [9]. Particularly, sample $x = 0.55$ follows a power law divergence $C_m/T \propto 1/(T^{1.25} + 1)$ once the $T_S$ anomaly is subtracted.

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$_x$ [5] at a similar Rh concentration. Coincidentally, the height of that anomaly nicely fits with the excess of Ce respect to the stoichiometric value, c.f. $2 + u = 2 + 0.15$, that represents a 7.5% excess of Ce (remind that this compound contains 2 Ce atoms per formula unit). This observation is in agreement with the nearly constant entropy contribution of the anomaly. Both, $T_S(x)$ and entropy behaviors do not fit into a random concentration spurious contribution. This unexpected feature can
be explained by analyzing the distribution of Ce atoms in this complex crystalline structure. As reported in Ref. [1] the Ce-plane, at \( z = 0.5 \), contains Ce atoms placed in the crystallographic site 4\( h \) whereas Ce atoms exceeding stoichiometric concentration (i.e. \( u = 0.15 \)) replace In atoms at the 2\( a \) site at the \( z = 0.1 \) planes. A further evidence for the intrinsic nature of this anomaly is given by scaling properties since for samples with \( x \leq 0.4 \) \( C_m(x,T = T_S) \) nicely coincide if represented as a function of a normalized temperature \( t = T/T_S \). \( T_S(x) \) extrapolates to \( T = 0 \) for \( x \approx 0.75 \), beyond the studied concentration range. Preliminary magnetic measurements indicate that this anomaly is suppressed by the application of a moderate magnetic field (\( B \approx 0.1 \) T). Such a rapid suppression confirms that it is not due to a spurious CePd\(_{1-x}\)Rh\(_x\) contribution because that compound has shown to behave differently under magnetic field [10].

III. 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.

From the magnetic behavior, the intermediate phase at \( T_N \leq T \leq T_C \) seems to behave as a Shastry-Sutherland lattice [11], as it occurs in the isotypic compound Ce\(_2\)Pd\(_2\)Sn [12]. This exotic phase, which requires of Ce-dimers formation, is suppressed once those dimers do not form anymore because of the weakening of Ce magnetic moments due to the arising Kondo screening.

Fig. 4 presents the concentration dependence of the satellite \( T_S(x) \) anomaly with its extrapolation to \( x \approx 0.75 \), and the Kondo temperature \( T_K(x) \) evaluated using the Desgranges-Schotte criterion [13]. 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. 4. Further studies at higher Rh concentration are in progress to better determine \( T_S(x) \) at lower temperature and its dependence on magnetic field.

Fig. 3: Magnetic phase diagram showing \( T_N(x) \) and \( T_C(x) \) phase boundaries decrease. SSL indicates the region where a Shastry Sutherland lattice is expected to form.

Fig. 4: Rh concentration dependence of the satellite \( T_S \) and the \( T_K(x) \) increase (right axis).

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