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Invited paper

Analysis of the very low temperature phase diagrams of two Ce compounds

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A rich variety of behaviours have been found studying the physical properties of Ce-lattice systems tuned around their critical regions, particularly in the few phase boundaries traced along more than a decade of temperature. The phase diagram of two Ce-systems: the antiferromagnetic $CeIn_{3-x}Sn_x$ and ferromagnetic $CePd_{1-x}Rh_x$ are analyzed covering the $10K > T > 0.02K$ range of temperature. Their respective magnetic phase diagrams are described as an interplay between different components determined after analyzing their thermodynamic properties. In the former compound, signs for the formation of a new phase are observed at very low temperature, whereas in the latter a field induced contribution is detected behind the critical point.

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1. Magnetic phase diagrams of Ce compounds

Since a Quantum Critical Point occurs by driving a second order transition to zero by a non thermal parameter [1], such a point is behind the sky line of the experimental possibilities. Nevertheless, the halo of its presence can be detected by its associated effects at finite temperature. In systems governed by the competition between Kondo screening and RKKY interaction [2], to tune the ordering temperature ($T_{N,C}$) down to very low temperature has shown to be unachievable in many cases. In Ce compounds under pressure, for example, $T_N(p)$ seems to vanish at finite temperature (at $T > 2$K) [3]. When they are driven by pressure (see Fig. 1a), that critical pressure coincides with the formation of a superconductive phase at lower temperature [4]. Similar limit of the phase boundary is also observed in most of Ce-systems driven by chemical potential variation, realized by doping (x) Ce-ligand elements [5], also included in Fig. 1a. This observation suggests that such a temperature ($3 > T > 2$K) may be related to an energy threshold which is present in all phase diagrams.

Coincidentally, the few phase boundaries traced below that temperature by doping, show a change of curvature in $T_{N,C}(x)$ (see Fig.1b), indicating a crossover between different magnetic ground state behaviours. Although such a crossover occurring at $x = x^*$ only signs the limit of the long range magnetic order, many authors had confused it with the critical one after arbitrary extrapolations of the phase boundary from $T > 2$K to $T = 0$ by following the negative $T_{N,C}(x)$ curvature. The few investigations performed down to at least one decade below that temperature have shown clear changes in the ground state behaviour before the actual critical concentration (x_{cr}) is reached. An unquestionable feature for the modification of the ground state at $x = x^*$ is evidenced by the presence of a first order transition around that concentration as shown by $CePd_2(Ge_{1-x}Si_x)_2$ [6] and $CeIn_{3-x}Sn_x$ [7]. This phenomenology allows to distinguish the $x^* > x > x_{cr}$ range as a *pre-critical* region, well differentiated from the magnetically ordered phase. Besides these properties, there are further characteristics systematically observed in the *pre-critical* region, like the nearly constant value of the maximum of the specific heat C_P/T [5] coincident with a broadening of the magnetic transition.

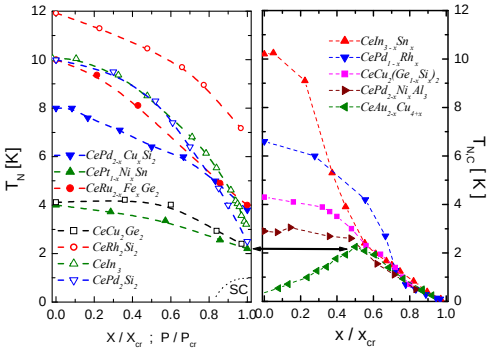

Fig. 1. Comparison of two types of phase diagrams including: a) compounds whose ordering temperature (T_N) vanish above 2K under pressure (P) or alloying (x), normalised to their respective critical values. b) systems whose $T_{N,C}(x)$ were traced by more than one decade showing a change of curvature, also around 2K.

Such a broadening in alloyed systems is frequently attributed to local disorder produced on the Ce-lattice by the atomic size differences of the Ce-ligands. Atomic disorder certainly plays an important role in systems containing ligands with large differences in atomic size (e.g. $CePt_{1-x}Ni_x$ [8] or $CeSi_{2-x}Ge_x$ [9]) because in all of them the transition smears out above the mentioned threshold value. On the contrary, for ligands with irrelevant atomic size difference (e.g. neighbours in a row of the periodic table like in $CeIn_{3-x}Sn_x$ [7] or $CePd_{1-x}Rh_x$ [10]) the transition is observed for more than two decades of temperature. Although from Nordhiem criterion the maximum disorder is expected at 50% of mixture, the respective *pre-critical* regions of the mentioned systems correspond to quite opposite ranges of composition: *6>In/Sn>3* for the former and *2/3>Pd/Rh>1/10* for the latter. Furthermore, the sharp $C_P(T)$ transition observed in the $CePd_{0.5}Rh_{0.5}$ alloy [10] proves that disorder effects are irrelevant at that concentration.

Besides the peculiarities shown by these two Ce-systems in the upper decade of their ordering temperatures (i.e. from $T_{N,C}$ ($x=0$) to $0.1T_{N,C}$ ($x=0$)), their respective phase diagrams show novel behaviors in the lowest investigated decade (i.e. $0.1\ T_{N,C}(x=0) \rightarrow\ 0.01T_{N,C}(x=0)$). In the next section we discuss the thermal properties of the cubic $CeIn_{3-x}Sn_x$, following with those of the orthorhombic $CePd_{1-x}Rh_x$, in their respective $T < 2$K regions.

2. Antiferromagnetic $CeIn_{3-x}Sn_x$

Stoichiometric $CeIn_3$ orders at T_N ($x=0$) $=10.2$K. By doping with *Sn*, T_N (x) decreases following Doniach´s description down to T_N (x^*) $\approx$ *2.2*K. Once in the pre-critical region $T_N(x)$ decreases linearly, extrapolating the critical concentration at $x_{cr} \approx 0.65$ [7,11]. In addition to the characteristic properties of the pre-critical region, thermodynamic properties (c.f. specific heat [11] and thermal expansion [12]) measured in $CeIn_{3-x}Sn_x$ down to the mili-Kelvin range show the onset of a new feature between $x = 0.55$ and *0.60*. As shown in Fig.2a for $x = 0.55$, the extrapolation of the C_P/T ($T > T_N$) tail (hereafter C_{NFL} because of its non-Fermi liquid behavior) crosses C_P/T below T_N. This indicates that in the ordered phase the entropy difference (ΔS) between measured (C_P/T) and extrapolated (C_{NFL}/T) function is nearly compensated, i.e. $\Delta S = S - S_{NFL} \rightarrow 0$ for $T > T_N$. Nevertheless, such is not the case for $0.60 \leq x \leq 0.80$ because C_{NFL}/T clearly extrapolates below C_P/T for $T < T_N$. The temperature dependence is obtained by fitting C_{NFL}/T ($T > T_N$) using the $C_{NFL}/T = D / (T^{\mu} + T_E)^q$ function described in ref. [3]. The dispersion of such a fit is smaller than 1%. This analysis indicates that a surplus of entropy arises between $0.60 \leq x < 0.80$, suggesting the presence of a new contribution: $\Delta C/T = C_P/T - C_{NFL}/T$ at very low temperature when approaching the critical concentration.

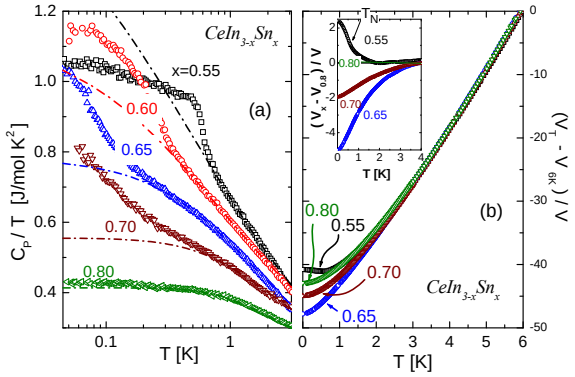

Fig 2. a) Very low temperature specific heat of $CeIn_{3-x}Sn_x$ (after Ref. [11]) and fits of C_P/T for $T > T_N$ (C_{NFL}/T, dot-dashed curves), showing an extra contribution ($\Delta C/T = C_P/T - C_{NFL}/T$) at $T < T_N$ for the $0.60 \leq x < 0.80$ samples (see the text). b) Temperature dependence of the volume: V_T (obtained form thermal expansion after Ref. [13]) for different concentrations, referred to the respective values at T = 6K: $(V_T - V_{6K})/V$. Inset: relative expansion (or shrinking) respect to $x = 0.80$ $(V_x - V_{0.80})/V$.

The field dependence of specific heat supports this interpretation because $\Delta C/T$ vanishes at a maximum critical field of $B_{cr}=6T$ [12], whereas C_{NFL}/T (B) keeps decreasing monotonously up to the highest measured field (B =10T) as expected for a NFL system. These different field dependences evidence an independent origin of those contributions. The T_E term is related to the crossover between the power law dependence at high temperature and the flattening at $T \rightarrow 0$, where $C_{NFL}/T \rightarrow D/T_E^{\ q}$. The $T_E(x)$ dependence is included in the phase diagram of Fig. 3. From the fitting procedure one obtains that (for each concentration) T_E increases with field as $T_E \sim B^2$ whilst coefficient D and exponent q show negligible variations with field [3].

In order to check whether this excess of entropy is related to the evolution of the phase boundary at very low temperature or it arises from another contribution, we have analysed the T dependence of the cell volume, extracted from thermal expansion (β) measurements [13] as: $V_T = \int \beta\, dT$. From thermodynamic laws one knows that $\beta \rightarrow 0$ as $T \rightarrow 0$ with the actual value of V_T at $T \rightarrow 0$ (V_0) remaining unknown. After the observed anomaly in the specific heat results, a non monotonous variation of $V_0(x)$ can be expect between $0.60 \leq x < 0.80$. Since the key parameter is $V_0(x)$ and not its absolute value, one may superpose the $V_T(x)$ data at T=6K (hereafter V_{6K}), well above the investigated anomaly as depicted in Fig. 2b. A slight relative deviation is observed around 2K, which becomes significant below 1K. Such a deviation can be better observed taking as a reference sample $x = 0.80$ where the investigated effect is already negligible. In the inset of Fig.2b one can appreciate the relative *expansion* of sample $x = 0.55$ upon cooling related to the transition at $T_N = 0.7$K, whereas sample $x = 0.65$ located at the quantum critical point [13], shows the largest relative *shrinking*.

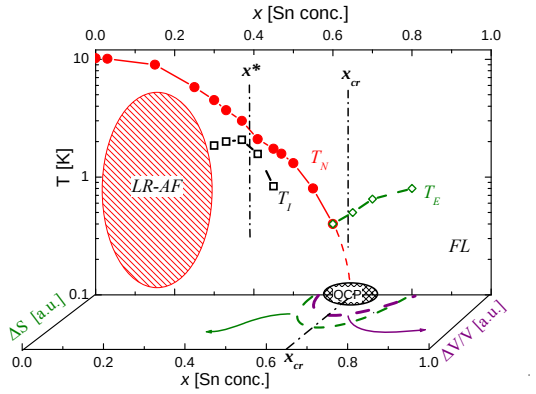

Fig. 3. Two decades phase diagram of $CeIn_{3-x}Sn_x$ in a `log T' scale, showing the phase boundary and related characteristic concentrations. T_I indicates a first order transition around x, and T_E are the values obtained from the fits (see text). The low temperature extra entropy ΔS and volume contributions $\Delta V/V$ are represented as two domes in a perpendicular plane around QCP.*

These thermal properties observed at very low temperature are collected in a phase diagram presented in Fig.3. Since the studied temperature range is now extended by another decade to lower temperature, the figure is displayed in a logarithmic T scale. Around the critical point, $\Delta S(x)$ and $\Delta V/V(x) = (V_T - V_{6K})/V$ variations are projected as a dome in a perpendicular plane at the lower part of the figure. These anomalous specific heat and volume variation at $T \rightarrow 0$ points to the presence of a new phase around the critical concentration, which involves a small amount of entropy: $\Delta S(0.65) \approx 0.7\%\ RLn2$ (in compliance with similar anomalies observed in other Ce base compounds [3]), and a $\Delta V/V \approx 7\ 10^{-6}$ between $x=0.55$ and 0.65.

3. Ferromagnetic $CePd_{1-x}Rh_x$

The Curie temperature of *CePd* occurs at $T_C\ (x=0) =$ 6.6K and, under doping with *Rh,* it also decreases according to Doniach´s prediction down to $T_C \approx$ 2.5K at $x^* \approx 0.6$. Behind that concentration it deviates from the canonical behaviour with a change of curvature in the phase boundary from negative to positive [10], indicating the crossover to the pre-critical region. The other limit of this region is determined by ac-susceptibility (χ_{ac}) measurements [14] around $x = 0.87$, where $T_C\ (x)$ turns down to zero with a linear concentration dependence. However, this magnetic property does not fully coincide with the specific heat [15] and thermal expansion [16] results at that range of concentration.

The discrepancy between those parameters is more evident in the $x = 0.85$ sample. As it can be seen in Fig. 4a, $T_C\ (x)$ determined by the cusp of $\chi_{ac}(T)$ nearly coincides with that of C_P/T up to $x=0.80$. However, an apparent logarithmic T dependence is observed in C_P/T (T) at $x = 0.85$, instead of an anomaly, which transforms into a power law at $x=0.87$, as depicted in Fig. 4b in a double logarithmic representation. Thermal expansion introduces a further discrepancy because at $x=0.85$ it shows an anomaly at around 0.35K in one of the measured directions (α_1) [16], in contrast to the cusp of $\chi_{ac}(T)$ at 0.14K and the lack of anomaly in C_P/T. Notice that in this anisotropic system, perpendicular thermal expansion coefficients: α_2(T) and α_3(T) are negative at this and following concentrations up to x=0.95.

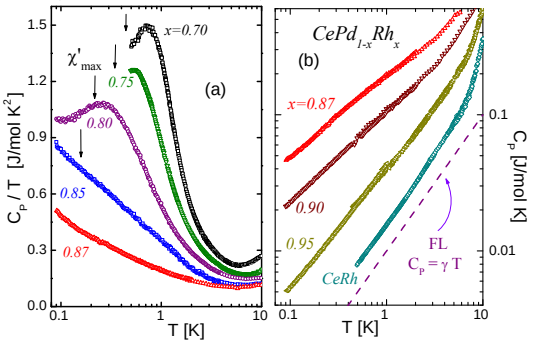

Fig. 4. a) Specific heat of samples ranging between $x=0.70$ and 0.87 in a C_P/T vs log T representation; arrows indicate the cusp of $\chi_{ac}(T)$, after Ref. [14]. b) Samples between $x = 0.87$ and 1 (CeRh) in a log C_P vs log T representation; the FL dotted-line indicates the temperature dependence for a Fermi Liquid behaviour. Data after Refs.[10, 15].

In order to clarify the apparent contradiction between these experimental results, we have analysed the field dependence of C_P/T in sample $x = 0.85$ [15]. In Fig.5a, a clear anomaly in $C_P/T(T,B)$ can be seen between 0.5 and 3Tesla, with a maximum at $T=T_{max}$ which increases linearly with field. Because $T_{max}(B)$ extrapolates around 0.3K for $B \rightarrow 0$, it arises the question whether the $C_P/T \propto -log(T)$ dependence is an artefact hiding two contributions which compensate each other in an apparent logarithmic dependence. The possibility of two components scenario is supported by the fact that $\chi_{ac}(T)$ and $\alpha_1(T)$ have their respective maxima at clearly different temperatures.

That possible of scenario is discussed by Vojta et al. [17], in a model for smeared phase transitions. According to that description: "At a smeared transition, the system divides itself up into spatial regions which independently undergo the transition at different values of the control parameter. Once static order has developed on some of the Rare Regions (RR), their order parameter can be aligned by infinitesimally small interaction of external field". Furthermore, "Such a RR (or *Griffiths islands*) can develop local order while the bulk system is disordered…". This pattern may apply to the system at hand if the *Griffiths islands* have a different dynamic and size from the bulk system because what χ_{ac} detects is not detected either by C_P/T or α, and vice-versa. In fact, magnetic field allows to distinguish between two different

behaviours, one associated to the $\chi_{ac}(T)$ anomaly and the other to $C_P/T(T)$. The intensity of the former anomaly is significantly reduced by field (about 50% with B =15mT [14]), whereas its position in temperature is practically not affected. On the contrary, $C_P/T(T)$ anomaly increases with field in both intensity and temperature, as shown in Fig. 5.

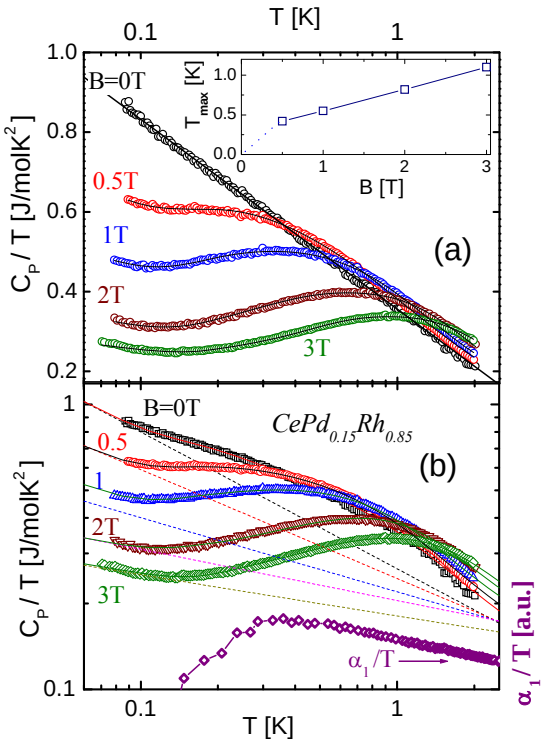

Fig. 5. Comparison of specific heat results for $CePd_{0.15}Rh_{0.85}$ in different magnetic fields, analyzed using logarithmic (a) and power law (b) dependencies (continuous curves), see the text. Inset in (a): show the discontinuity of T_{max} between B=0 and 0.5T. The α_1/T anomaly is included in (b) for comparison.

Since the following concentration (x=0.87) shows a clear $C_P/T(T) \propto 1/T^q$ dependence, we have analyzed the $C_P/T(T,B)$ curves of x = 0.85 according to a two components scenario: one accounting for a non-Fermi liquid (NFL) contribution (C_{NFL}) and the other for the magnetic *islands* (C_{mag}). In Fig.5b we show the fits performed on x=0.85 for B=0, 0.5, 1, 2 and 3T, applying the $C_P/T = C_{NFL}/T + C_{mag}/T$ criterion, with $C_{NFL}/T(T) \propto 1/T^q$ and $C_{mag}/T \propto [(\Delta/T)/\cosh(\Delta/T)]^2/T$ (a Schottky type anomaly). According with this description, T_{max} = 0.35K coincides with that of $\alpha_1(T)$ at zero field. Comparing the reliability of the $C_{NFL}/T(T) \propto 1/T^q$ dependence respect to the eventual $C_{NFL}/T(T) \propto -log(T)$, one sees that the power law has lower dispersion (WSSR) than the logarithmic one by a factor two. Therefore, the observed experimental behaviour of sample x=0.85 can be described as: i) due to the presence of Rare Regions imbed in a weakly interacting NFL medium, with ii) a power law T dependence .

A further test for this description is performed extending this analysis to C_P/T (T) results of all studied samples (x = 0.75, 0.80, 0.85, 0.87 and 0.90), including the five mentioned fields. For the fit of this set of 25 curves we have used 2 free parameters for the NFL component: $C_{NFL}/T = D/T^q$, and 4 for the Schottky-like anomaly: $C_{mag} = \sum A_i [\Delta_i/T \cosh(\Delta_i/T)]^2$, with i=1,2,3. Here, three free parameters correspond to A_i and one to Δ_i since they are fixed by a $2\Delta_1=\Delta_2=1/2\Delta_2$ relationship. Such a distribution of Δ_i is required to simulate the observed broadening of the anomaly, which is probably due to the remaining effect of the Kondo screening on the ground state [10].

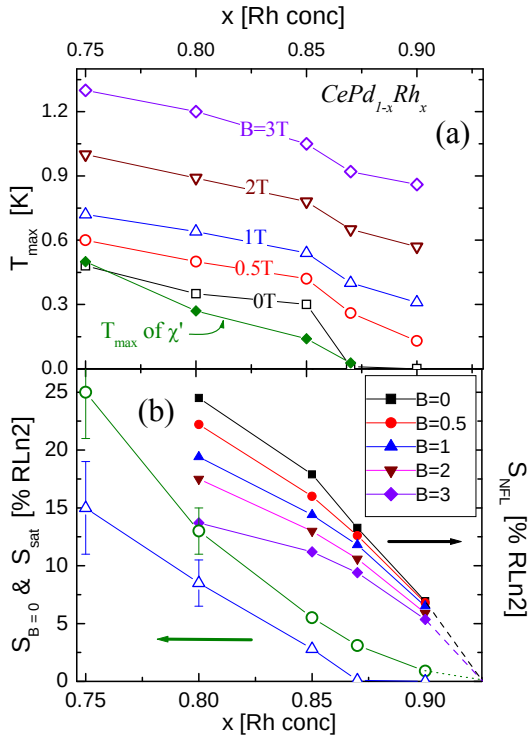

Fig. 6: a) Concentration dependence of T_{max} at different applied fields, compared with the maximum of χ'_{ac} (at B=0). b) Concentration dependence of the magnetic entropy at zero $S_{B=0}$ and saturation S_{Sat} (left axis), and that of S_{NFL} (right axis) at different fields.

Not a minor detail is the Schottky-like description required for the fit of the C_{mag}/T component, because it is based on Fermi Dirac statistic. On the contrary, models used to describe *static clusters* in disordered alloys (see e.g. Matthews et al. [18]) apply Einstein functions: $E(\theta/T)$-$E(\eta\theta/T)$, obeying Bose Einstein statistic. We have also tested this possible behavior using the same criteria, obtaining a better fit of the tail at high temperature. This means that, despite the randomness of those Rare Regions formation, their access to certain energy level depends on the number of similar entities having the same energy,

suggesting a correlated dynamic behaviour. These two scenarios are accounted in Vojta's model, where a distinction is done between smeared phase transitions and Griffiths phenomena: "…while in the former case true static order develops, in the latter the order parameter of RR fluctuates slowly" [17]. In any case a conclusive distinction between those possible scenarios for this compound requires microscopic time sensitive measurements.

At $B=0$, C_{mag}/T vanishes at $x = 0.87$ with a discontinuity in the $T_{max}(x)$ between $x=0.85$ and 0.87, as it is depicted in Fig. 6a. At that concentration none of the measured properties: χ_{ac}, C_P nor β, show any anomaly at least down to 50mK. Nevertheless, the C_{mag}/T anomaly persists under applied field ($0.5 \leq B \leq 3T$) like a field induced effect, whereas a small jump in $T_{max}(x,B\neq0)$ between 0.85 and 0.87 reflects the presence of a critical point between those concentrations. Similar field induced anomaly in specific heat was observed in the nearly intermediate valence $CeSn_3$ [19].

Such a filed induced contribution indicates that the system does not enter into a truly non-magnetic (or Fermi liquid) ground state at that concentration. In order to determine at which Rh concentration the crossover to the intermediate valence (IV) ground state actually occurs, we have evaluated the variation of the degrees of freedom of each component as a function of x. This information is achieved by computing the respective contributions to the entropy as: $S_{NFL} = \int C_{NFL}/T\, dT$ and $S_{mag} = \int C_{mag}/T\, dT$ respectively. S_{NFL} can be analytically computed avoiding the $T \rightarrow 0$ divergence with a cut-off at low enough temperature, with an upper limit taken at 4K, where $S_{mag}(T)$ tends to saturate above the Schottky-like anomaly. These arbitrary limits of integrations introduce a minor error in the evaluation of the entropy, nevertheless our main task is to analyze its B and x dependences and not the actual value. Comparing S_{NFL} and S_{mag} evolutions as a function of field, it seems that some degrees of freedom are "pumped" from the S_{NFL} component into the S_{mag} one as B increases. This effect becomes more evident in the case of $x > x_{cr}$ samples (i.e. 0.87 and 0.90) since $S_{mag}(B)$ arises continuously from zero at $B=0$, showing a tendency to saturation (S_{Sat}) at high field ($B=3T$). Fig. 6b shows how S_{NFL} (B, x) and $S_{Sat}(x)$ extrapolate to zero at the same concentration. This indicates that NFL and magnetic components vanish simultaneously at $x \approx 0.93$, where the system finally enters its intermediate valence regime, characterised by a Fermi liquid behaviour.

The existence of magnetic response behind the critical point simply indicates that the weakened Ce moments do not vanish completely at x_{cr} though the spontaneous magnetic interaction does. Also Ferromagnetic MnSi shows NFL behaviour above its critical pressure [20], despite its magnetic transition was suppressed. Under magnetic field, magnetic interactions may reappear once the remaining weakened moments are re-aligned by the external field. In any case, spectroscopic investigations are required to clarify this microscopic behaviour.

This analysis provides some valuable information: i) it confirms that at $B = 0$ the degrees of freedom related to the S_{mag} contribution are exhausted at $x = 0.87$; ii) the Rare Regions increase their degrees of freedom under magnetic field up to a saturation value, which then decrease with concentration, iii) this field induced enhancement tends to zero together with the NFL component at x ≈ 0.93, i.e. clearly behind the "zero field" critical point at x_{cr} and iv) the nature of the NFL component seems not to be affected undergoing the critical concentration because the q exponent (≈ 0.35) is dependent of concentration. On the contrary, the D coefficient decreases proportionally to the involved degrees of freedom (i.e. S_{NFL}) from $D \approx 0.38$ (at $x=0.80$) to 0.10 (at $x=0.90$) at zero field, and from 0.20 to 0.08 under $B=3T$ at those respective concentrations.

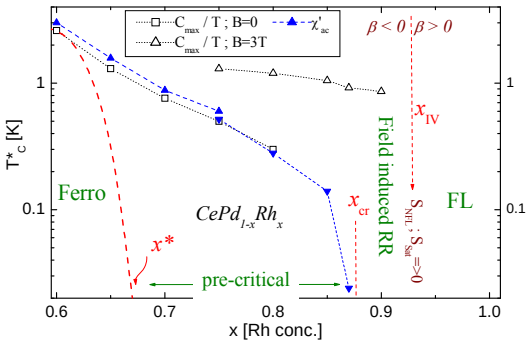

Fig 7. Low temperature phase diagram of $CePd_{1-x}Rh_x$ in a `log T' scale, showing three characteristic concentrations: x^* defines the crossover from ferro-magnetic to pre-critical region, which ends at the critical point x_{cr} . The border between the field induced Rare Regions and the truly non-magnetic (FL) phase: x_{IV} , occurs where the entropy of NFL (S_{NFL}) and magnetic (S_{Sat}) components extrapolate to zero whereas β becomes positive.

A detailed phase diagram for the very low temperature range is presented in Fig. 7. There, different characteristic concentrations are identified, the first at $x=x^*$ where the crossover from LR ferromagnetic phase occurs. The second at $x=x_{cr}$, where the maximum of the χ_{ac} signal extrapolates to zero, and the third one where also the field induced magnetic component vanishes at $x_{IV} \approx 0.93$. The relevance of this concentration is confirmed by thermal expansion measurements [15] because the negative sign of β between $0.85 \leq x \leq 0.90$ turns to $\beta > 0$ at $x = 0.95$. Positive thermal expansion sign is expected in Ce-IV systems (e.g. CeN [21], which is the IV prototype), because applied pressure reduces Ce-ion volume by delocalizing its *4f* electron.

4. Conclusions

We have seen that the phase boundaries of *Ce* compounds either vanish or show a change of curvature at a sort of threshold temperature around 2K. Although that

temperature seems to be quite high for eventual quantum effects related to a $T = 0$ critical point, neutron scattering Bragg reflections on $CeCu_{6-x}Au_x$ at the quantum critical concentration [22] show that "Bragg peaks are replaced by magnetic critical scattering which are greatly (but not completely) suppress at 1.5K".

In the two doped systems traced below that temperature, a pre-critical region was identified between $x^* < x < x_{cr}$, with clearly different properties than those of the long range ordered phase (i.e. between $0 < x < x^*$). In the case of $CeIn_{3-x}Sn_x$, the anomalous concentration dependence of C_P /T and V_T at $T \rightarrow 0$ suggest the possibility of the existence of a new phase at very low temperature. In the other case, the behaviour of $CePd_{1-x}Rh_x$ approaching its critical point was described using a two component pattern in agreement with Vojta´s model for smeared transitions. According to these results Rare Regions may form imbed into a magnetically disordered environment. Since these regions increase their degrees of freedom with magnetic field and fit into a Schottky-like description, their behaviour is not suitable for a static-cluster scenario. There is a relatively short range behind the critical point where the Rare Regions are induced by field, limited by the onset of the intermediate valence (i.e. FL) ground state. This occurs in coincidence with the change of thermal expansion sign (from negative to positive) and the T^2 dependence of the electrical resistivity.

Acknowledgments

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