

The evanescence of ferromagnetic order in the $\text{Ce}_{1-x}\text{Y}_x\text{Ni}_{0.8}\text{Pt}_{0.2}$ dense Kondo system

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Abstract. The experimental study of the low temperature (down to 0.4 K) specific heat of the Kondo ferromagnet system $\text{Ce}_{1-x}\text{Y}_x\text{Ni}_{0.8}\text{Pt}_{0.2}$ is presented. Special attention has been paid to the $\text{Ce}_{0.8}\text{Y}_{0.2}\text{Ni}_{0.8}\text{Pt}_{0.2}$ composition, which is situated near the Zero Temperature Phase Transition. This compound presents a marked divergence of the specific heat at low temperatures, signature of a Non Fermi Liquid behaviour. Different frameworks which could account this behaviour are analysed, being the most plausible a “strong coupling” situation with disorder effects. The importance of these effects is discussed in the general scenario of the Non Fermi Liquid compounds considering the differences between “ligand” and “Kondo hole” systems.

PACS. 75.30.Mb Valence fluctuation, Kondo lattice and heavy-fermion phenomena – 71.10.Hf Non-Fermi-Liquid ground states, electron phase diagrams and phase transitions in model systems

1 Introduction

It is generally accepted that the competition between intersite Ruderman-Kittel-Kasuya-Yosida (RKKY) and intrasite Kondo interactions governs the magnetic behaviour of the Kondo lattice compounds. The Doniach model provides a useful description considering that both the RKKY and Kondo interactions have different dependencies on the exchange parameter J [1,2] and they can be modified by applied or chemical pressure.

The actual interest is focussed on the region of the phase diagram where the magnetic order is suppressed, occurring at a given critical pressure P_C or, in the case of induced chemical pressure, at a critical concentration x_C . In this region, corresponding to a Zero Temperature Phase Transition (ZTPT), anomalous behaviours of the specific heat, resistivity or susceptibility, which are not expected for Fermi Liquids have been found. These “Non Fermi Liquid” (NFL) [3] compounds appear as a suggestive area of scientific activity in the field of the strongly correlated electron systems [4,5]. Several theoretical approaches try to explain the physics involved in these processes. Among them, we can mention those considering spin fluctuations that exist close to a Quantum Critical Point [6–9], originally studied in itinerant electron systems. This point of view corresponds to a “weak coupling” approach, fol-

lowing the Coleman description [10]. In the “strong coupling” approach, the Kondo effect breaks directly down at the transition and local fluctuations play the dominant role [10,11]. The effects of disorder have also been considered introducing a distribution of Kondo temperatures (T_K) [12] or small magnetically ordered regions embedded in a paramagnetic matrix, corresponding to a Griffiths phase situation [13]. The reader is referred to a recent review [14] for an actualized and comprehensive analysis of the different approaches and the state of the art of the most significant experimental examples of Non Fermi Liquids.

A large number of experimental examples have been reported during the last years. However, the observed behaviours were rather diverse and the analysis were, unfortunately, not always conclusive about the existence and the origin of the NFL behaviour. To illustrate this point, it is convenient to mention several examples. In CeRh_2Si_2 , the NFL behaviour is not observed when the ZTPT is reached by applying pressure while it is well detected in the $\text{CeRh}_{2-x}\text{Ru}_x\text{Si}_2$ series close to the critical composition $x_C = 0.95$ [15,16]. These authors claim for the importance of disorder in producing NFL. In contrast, another report shows that no difference was found between applied pressure in CeRu_2Ge_2 and chemical pressure in $\text{CeRu}_2(\text{Ge}_{1-x}\text{Si}_x)_2$, concluding that disorder is not a dominant factor in the physics of such system [17].

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The importance of disorder has also been studied in $\text{UCu}_{5-x}\text{Pd}_x$ [18–20], first interpreted as an example of NFL consistent with the Griffiths phase scenario, although more recently it has been proposed the annealing as a control parameter to tune the NFL behaviour, questioning the pure disorder effects [21]. Also controversial is the case of the $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)\text{Ge}_2$ and $\text{CeNi}_2(\text{Ge}_{1-y}\text{Si}_y)_2$ compounds, where the NFL behaviour can not uniquely be explained with the Griffiths phase or the T_K distribution models [22].

It is clear that new examples are needed to obtain further experimental data and to provide insight for the current theories. In this way, there exist two very close systems, considered as the earliest examples of the usefulness of the Doniach diagram: $\text{CePt}_{1-x}\text{Ni}_x$ [23] and $\text{CePd}_{1-x}\text{Ni}_x$ [24]. In the Pt/Pd rich side, both series are some of the few examples of Kondo ferromagnets. The $4f$ -conduction band hybridization increases when the cell volume decreases (approaching CeNi), and the suppression of the long-range magnetic order occurs for both systems around $x = 0.95$. The variation of the Curie temperature (T_C) and T_K with composition agrees with the Doniach diagram and is pretty similar in both series. Recently, it has been demonstrated by X-ray absorption spectroscopy that in $\text{CePt}_{1-x}\text{Ni}_x$ a progressive Kondo screening develops with the Ni introduction, but keeping the integral $3+$ valence for the magnetically ordered compounds ($x \leq 0.9$) [25].

The existence of NFL behaviour close to the ferromagnetic instability has been investigated in $\text{CePd}_{0.05}\text{Ni}_{0.95}$ by Kappler *et al.* [26]. The experimental data of specific heat, resistivity and saturation magnetization suggest the existence of a NFL in which the disorder seems not to play a significant role.

Another possible way to reach the evanescence of the long-range magnetic order in these systems is by substituting Ce atoms by smaller Y ones [27], introducing a supplementary disorder reminiscent of the well-known $\text{U}_{1-x}\text{Y}_x\text{Pd}_3$ system [28]. The $\text{CeNi}_{0.8}\text{Pt}_{0.2}$ compound orders at $T_C = 8.8$ K, whilst for $\text{Ce}_{0.9}\text{Y}_{0.1}\text{Ni}_{0.8}\text{Pt}_{0.2}$ the Curie temperature is 4.5 K [29]. A linear extrapolation indicates that the ZPTT takes place in the proximity of $\text{Ce}_{0.8}\text{Y}_{0.2}\text{Ni}_{0.8}\text{Pt}_{0.2}$. These different ways to approach the ZPTT have been identified by some authors [30] as “ligand” and “Kondo hole” systems, respectively, and our compounds provide an interesting possibility to discuss together both approaches.

In the actual paper we present the low temperature specific heat measurements in $\text{Ce}_{1-x}\text{Y}_x\text{Ni}_{0.8}\text{Pt}_{0.2}$ down to 0.4 K. Previous data of resistivity and magnetization obtained for $T \geq 2$ K [27, 31] have been considered for the final discussion of our results.

2 Experimental and crystallographic analysis

Polycrystalline samples with nominal compositions $x = 0, 0.1, 0.2, 0.3$ and 0.5 were prepared in a cold-crucible induction furnace under argon atmosphere, as reported in previous works [27].

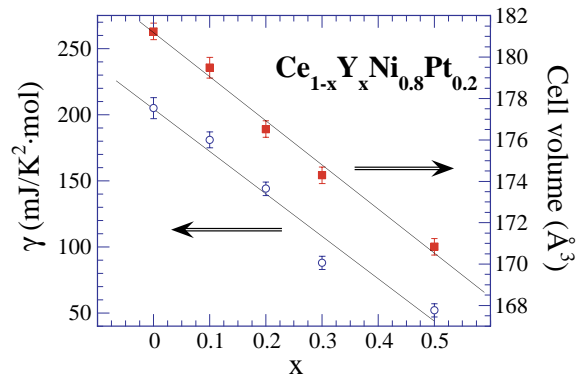


Fig. 1. Cell volume and specific heat electronic coefficient (γ) obtained as explained in the text.

The X-ray diffraction analysis of the samples confirmed that they crystallize in the CrB-type crystallographic structure (Cmcm space group) and no indications of spurious phases were appreciated. The Rietveld analysis was performed considering Ce and Y atoms randomly distributed on the same 4c) site while the Pt and Ni lie on the other 4c) site. The cell volume variation at room temperature is presented in Figure 1. The proportion of the constituents (real stoichiometry) and the homogeneity of the samples were checked by total X-ray reflection fluorescence technique TXRF [32] confirming the analysis of the X-ray diffraction data.

The specific heat measurements were carried out down to 0.4 K in an Oxford’s ^3He cryostat equipped with a mechanical switch used as a thermal switch in order to measure by means of the quasi-adiabatic method (for a recent review, see for example Ref. [33]) in a home-built low-addenda calorimeter [34] with the appropriate conditions to perform such measurements.

3 Specific heat results

Figure 2 displays the specific heat (C_p) versus temperature up to 10 K for all the studied compounds. According to previous results of magnetization and resistivity [27], the anomaly corresponding to the magnetic ordering temperature is only observed for the $x = 0$ and $x = 0.1$ compounds. For the $x = 0.1$ one, the anomaly is broadened and its maximum value strongly decreases. The Curie temperatures, defined as the inflexion point above the maximum $C_p(T)$ value, are 8.8 and 4.5 K respectively. These values are presented in the inset of Figure 2, together with the corresponding linear extrapolation showing the composition for which the long-range magnetic order is expected to disappear. For $x = 0.2, 0.3$ and 0.5 , no anomalies related to a Curie temperature were found down to 0.4 K. For the $x = 0.3$ and 0.5 samples, the specific heat thermal dependence is consistent with a metallic non-magnetic behaviour, as expected for an enhanced Pauli paramagnet. This description points out that the disappearance of the magnetic order takes place around $x = 0.2$.

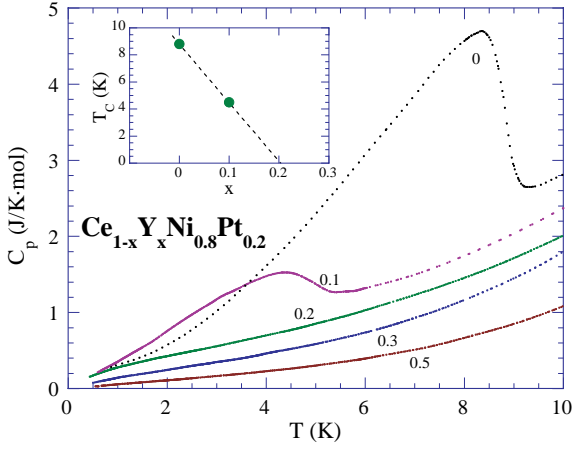


Fig. 2. Total specific heat (C_p) variation with temperature for the studied compounds. The inset shows the linear extrapolation of T_C (obtained from these measurements) with composition.

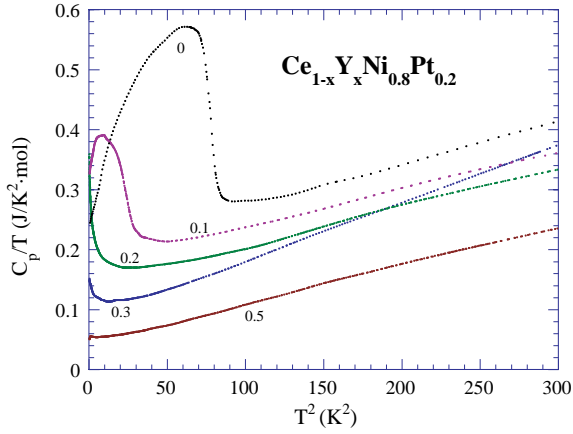


Fig. 3. Specific heat plotted as C_p/T vs. T^2 showing the temperature range with a linear behaviour whose extrapolation to $T = 0$ K allows to estimate the temperature independent γ coefficient (see text).

The C_p/T vs. T^2 plot of the specific heat is presented in Figure 3 and it reveals a large linear region for all the compositions. The extrapolation to $T = 0$ K of this linear behaviour yield the “temperature independent electronic coefficient” (γ) values that are plotted in Figure 1 together with the cell volume variation. A direct relationship is evidenced between both quantities, as already was pointed out in a preliminary work [31]. The γ values obtained in this way are only indicative of the electronic correlation enhancement, *i.e.*, how far is the corresponding compound from the free electron picture.

The used procedure does not take into account the contribution of the crystalline electric field (CEF). However, as discussed in reference [29], the CEF contribution for these compounds is very small in the limited temperature range used in the present study.

The phonon contribution to the specific heat can be estimated from the slope in the linear range of Figure 3.

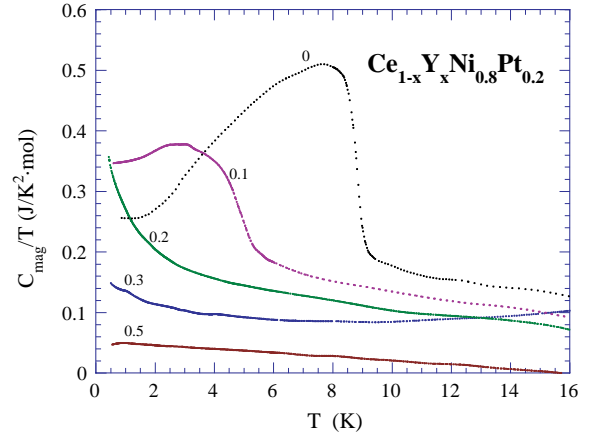


Fig. 4. C_{mag}/T vs. T representation of the specific heat. Note the low temperature divergence for the $x = 0.2$ sample.

The Debye temperatures, extracted from these slopes, are rather similar for all the compounds and they are in good agreement with those reported for the non-magnetic compounds [35]. The larger slope observed for the $x = 0.3$ compound is certainly due to lattice effects, and it yields a Debye temperature value 10% smaller. However, when an isomorphous non-magnetic compound exists, its specific heat is usually taken as the “non-magnetic” contribution. In our case, we can consider $\text{LaNi}_{0.8}\text{Pt}_{0.2}$ [29] as the non-magnetic corresponding compound. The “magnetic” contribution to the specific heat (C_{mag}) is then obtained by subtraction of the non-magnetic one, after applying the corresponding mass correction [36] for each compound. In this case, C_{mag} reflects any effect related to the magnetic order transition and/or the temperature dependence of the electronic term. We have presented this information in Figure 4 as the thermal dependence of C_{mag}/T . Taking a look to the low temperature side of this figure, we can observe an almost constant behaviour for $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Ni}_{0.8}\text{Pt}_{0.2}$, with a value of 52 mJ/K²mol, then confirming the “Fermi Liquid” character of this compound, with a moderate γ value. The negligible C_{mag}/T value for this composition around 15 K evinces the intrinsic difficulty of an accurate estimation of the phonon contribution, being the specific heat of the isomorphous La compound just a reasonable approach. Coming back to the low temperature side of these data, a small increase of the C_{mag}/T value is observed for $x = 0.3$, while $x = 0.2$ exhibits a clear divergence when approaching $T = 0$ K.

For the ordered compounds, the contribution due to the magnetic transition makes difficult to analyse their low temperature limit but, on the other hand, we can obtain the magnetic entropy for these compounds as $S_{mag} = \int (C_{mag}/T) dT$. After normalizing these entropies per Ce mol, we have displayed them in Figure 5. When Ce^{3+} ions lie in a low symmetry site (the $J = 5/2$ ground state splits into three doublets) and long-range magnetic order exists, the $R \ln 2$ value of the magnetic entropy should be reached at T_C . A reduction from this value is interpreted as due to the Kondo interactions and then,

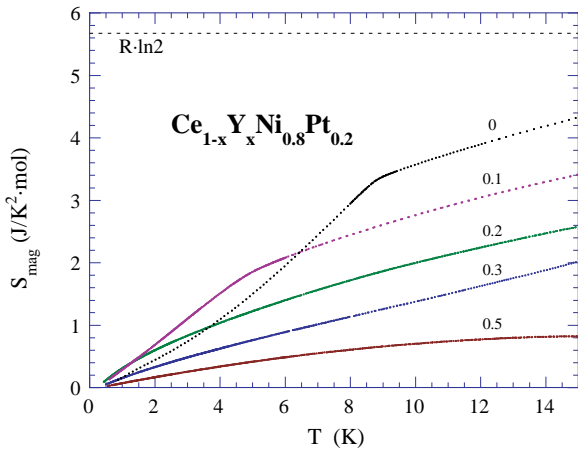


Fig. 5. Magnetic entropy, estimated as $S_{mag} = \int (C_{mag}/T)dT$. The changes of slope for the samples with $x = 0$ and 0.1 correspond to the Curie temperatures.

a T_K value can be estimated considering the $S_{mag}(T_C)$ and T_C ones [37]. From the present data, the obtained T_K values are 15.9 K for $CeNi_{0.8}Pt_{0.2}$ and 12.7 K for $Ce_{0.9}Y_{0.1}Ni_{0.8}Pt_{0.2}$, in very good agreement with those obtained for different samples of the same nominal compositions [29].

For the non-ordered compounds ($x \leq 0.2$), the estimation of T_K needs another kind of analysis. Taking into account the Sacramento and Schlottmann calculations for a $J = 1/2$ impurity [38], one could estimate T_K as the value at which 45% of the total $R \ln 2$ entropy is recovered. But it can also be estimated from the relationship between T_K and γ [39]. The absolute values obtained from these procedures are different, but the common fact for our samples is that T_K increases from $x = 0.2$ to $x = 0.3$. For the non-ordered $Ce_{0.5}Y_{0.5}Ni_{0.8}Pt_{0.2}$ compound, the estimate of T_K from entropy considerations lacks of physical meaning due to the uncertainty inferred by the lattice contribution subtraction, that is significant in relative value for this composition, as explained above.

It is remarkable that for $x = 0.2$, which corresponds to the evanescence of the magnetic order, a divergence is found in C_{mag}/T at low temperatures. In absence of measurements below 0.4 K, the existence of a transition at lower temperatures can not fully be ruled out. However, it is reasonable to think that, in that case, the magnetic contribution should be much smaller, as will be commented in the discussion. Therefore, the low temperature dependence of C_{mag}/T must be carefully analysed in this compound.

4 Discussion

The substitution of Ce by Y in $CeNi_{0.8}Pt_{0.2}$ provokes two main effects:

- A dilution of the Ce atoms which induces a decrease of the average magnetic molecular field.

- A decrease in the cell volume leading to an effective increase of the $4f$ -conduction band hybridization, with a progressive screening of the Ce magnetic moment.

The rapid decrease of T_C with composition and the strong reduction of the entropy associated to the magnetic transition, which are both not proportional to the Y content, indicate that the second effect is quite relevant in this series. Furthermore, the dilution causes a rapid decrease of the coherence in the Kondo lattice and the long-range magnetic order disappears at least down to 0.4 K for the $Ce_{0.8}Y_{0.2}Ni_{0.8}Pt_{0.2}$ compound.

The samples with $x = 0$ and 0.1 are well characterized as Kondo ferromagnets in which the introduction of Y induces a modification in the balance between the Kondo and RKKY interactions, leading to an increase of the T_K/T_C ratio with the increasing Y content, in agreement with the evolution of the paramagnetic Curie temperatures (-49 , -81 and -89 K for $x = 0$, $x = 0.1$ and $x = 0.2$ respectively [27]).

On the other hand, from the low temperature specific heat, $Ce_{0.5}Y_{0.5}Ni_{0.8}Pt_{0.2}$ could be classified as a Fermi Liquid with a moderate γ value. In principle, the Fermi Liquid theory should only be applied at very low temperatures, where it describes the low lying excitations of the quasiparticles (interacting fermions). However, it is usually assumed to extend this label (or the equivalent “enhanced Pauli paramagnets” [40]) to compounds presenting a constant electronic specific heat and magnetic susceptibility coefficient and a T^2 dependence of the electrical resistivity. This is just the case of the C_{mag}/T (present work) and the magnetic susceptibility [27] corresponding to the $x = 0.5$ compound. The minimum found in the electrical resistivity of this compound around 16 K [27] is a signature of a Kondo impurity behaviour. This means that the coherence of the Kondo lattice has been broken by the dilution of the Ce ions which individually present an almost full Kondo screening, then behaving as Kondo impurities.

The crossover between ordered and non-ordered compounds is found at the $x = 0.2$ composition. The C_{mag}/T curve corresponding to $Ce_{0.8}Y_{0.2}Ni_{0.8}Pt_{0.2}$ exhibits a clear divergence at low temperatures, whose extrapolation to $T = 0$ K is 450 mJ/K²mol, a rather enhanced value respect to that expected for a normal metal (pure free electron picture). The origin of this divergence could be linked to intrinsic electronic effects or to a magnetic transition at lower temperatures. This last possibility is not the most likely for $Ce_{0.8}Y_{0.2}Ni_{0.8}Pt_{0.2}$. In fact, if there were a magnetic transition, its contribution to the specific heat would be much smaller than those found for the $x = 0$ and 0.1 compounds because the hybridization enhancement reflected by the T_K/T_C evolution involves a decrease of the Ce magnetic moment (see Ref. [29]). Then, the specific heat jump at T_C should be much smaller than that of the $x = 0.1$ compound, and this is not quite in agreement with the experimental values (see Fig. 4). Another reasoning can be argued considering the mean-field calculations that account the short-range correlations preceding long-range magnetic ordering [36,41]. They should

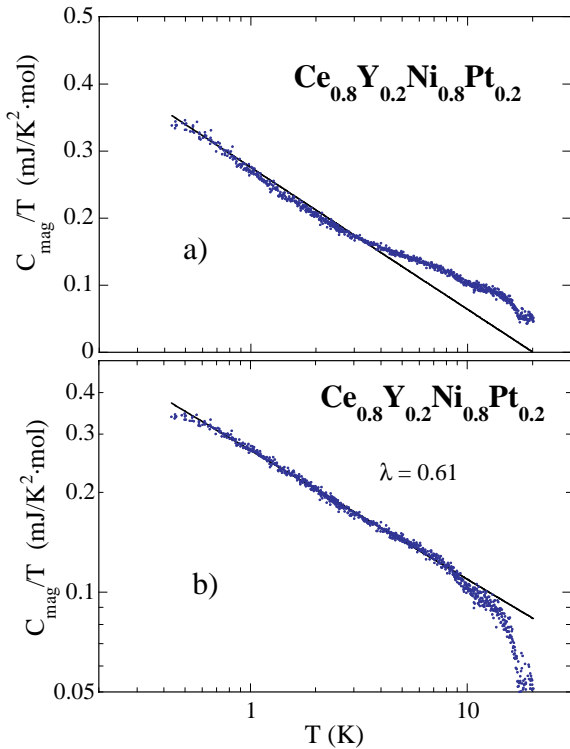


Fig. 6. a) C_{mag}/T vs. $\log T$ and b) $\log(C_{\text{mag}}/T)$ vs. $\log T$ plots for the $x = 0.2$ compound. The reported λ value is the obtained exponent from the $C_{\text{mag}}/T \propto T^{-1+\lambda}$ law (see text).

follow a T^{-3} law in a C_{mag}/T representation, which is not the case for this low temperature divergence. Then, a NFL behaviour in the ZTPT is revealed from our specific heat measurements.

In order to check the origin of the Non Fermi Liquid behaviour we have plotted C_{mag}/T vs. T in logarithmic and semilogarithmic scales. In Figure 6 we show these plots for $x = 0.2$ with fits corresponding to $C_{\text{mag}}/T \propto -\log T$ (Fig. 6a), as observed in the case of magnetic fluctuations [7–9], and $C_{\text{mag}}/T \propto T^{-1+\lambda}$ (Fig. 6b) as proposed by Castro Neto [13], who considered the existence of Griffiths phases consisting in magnetic clusters embedded in a non-magnetic matrix. Between these two laws, the $T^{-1+\lambda}$ one displays a better agreement with the experimental results, matching them for more than one temperature decade (from 0.5 to 10 K), while the $-\log T$ law only fits a more reduced temperature range (0.4–3 K). The temperature range of the fit and the value obtained for the λ coefficient (0.61) are similar to those found in other NFL with disorder effects such as $\text{Th}_{1-x}\text{U}_x\text{Pd}_2\text{Al}_3$ ($\lambda = 0.6$), $\text{UCu}_{5-x}\text{Pd}_x$ ($\lambda = 0.8$), UCu_4Pt ($\lambda = 0.83$) or $\text{Y}_{0.8}\text{U}_{0.2}\text{Pd}_3$ ($\lambda = 0.76$) [42].

We are aware that these fits represent a mere parametrisation and before proposing a final statement, it would be useful to analyse the different models from a wider perspective. The spin fluctuation theory of the Quantum Critical Point was originally developed for itinerant d-electron systems, but several extensions have tried

to apply it to systems with local magnetic moments close to a ferro or antiferromagnetic instability [6–9, 43]. In most cases, the asymptotic behaviour at low temperatures is used to check the validity of the models (see Ref. [14] for a comprehensive review of the different approaches). In particular, the temperature law arising from the different spin fluctuation calculations [14] is $C_{\text{mag}}/T \propto \log T$ for three-dimensional ferromagnets. This law, as seen in Figure 6, accounts a bit better our lowest temperature data.

However, the application of the spin fluctuation model to nearly localized f-electron systems is a matter of controversy [44], and considering the general behaviour of our series, we are more inclined to think that our system is better understood as a “strong coupling” one [3] where the instability is approached from the magnetic side where local magnetic moments exist. Supporting this assumption, we have to remind the reader that indications of Kondo diluted impurities were found for the $x = 0.5$ compound [27]. Then, localized magnetic moments (in a lattice or as impurities) seem to be present all along the series. Keeping in mind this argumentation, the lower temperature deviation of C_{mag}/T from the power law can also be accounted by a Kondo temperature distribution model [15]. Other authors [16] also mention the possibility of recovering the Fermi Liquid behaviour close to zero temperature or the possibility of a superparamagnetic freezing associated to the existence of spin-clusters [18, 19], compatible with the Griffiths phase scenario.

Unfortunately, neither resistivity nor susceptibility measurements can give us supplementary hints to support one or another option. Resistivity measurements exist only down to 2 K and their fits are of very limited intrinsic value due to the brittleness of the samples and their problems of crack propagation [27].

The magnetic susceptibility measurements [31] show a strong dependence on the applied magnetic field. The power law is only found in a very limited temperature range, with exponent values always greater than -1 , in clear disagreement with the $T^{-4/3}$ law predicted for three-dimensional ferromagnets by the spin fluctuation models [14]. On the other hand, if we look for λ values similar to that found from the specific heat, they are only found for high applied magnetic fields (>2 T). These magnetic susceptibility measurements confirm neither a spin fluctuation origin nor the Griffiths phase scenario. However, the strong magnetic field dependence of the magnetic susceptibility supports the existence of magnetic impurities (Ce ions with incomplete Kondo screening) or clusters (isolated aggregates of magnetic ions). The existence of clusters or aggregates has been proposed in a parallel series ($\text{CeCu}_{1-x}\text{Ni}_x$) that exhibits a spin-glass-like phase with competing positive and negative interactions [45]. In our case, the ferromagnetism persists along the series until the evanescence of the long-range magnetic order and the conditions to obtain a spin-glass are not fulfilled.

A last comment appears when the existing literature is analysed. Although most of the examples are subject of controversy, it seems that disorder driven theories provide at least a tentative description of the compounds

reaching the ZTPT by dilution of the magnetic ions, as occurs in $U_{1-x}Y_xPd_3$ [28] and $Th_{1-x}U_xPd_2Al_3$ [42], and also for compounds with non-isoelectronic substitutions on the non-magnetic ions. Examples of such analysis can be found in $UCu_{5-x}Pd_x$ [19,42], UCu_4Pt [42], $CeRh_{2-x}Ru_xSi_2$ [17,18]. In some of these cases it is not surprising to find spin-glass behaviours.

On the contrary, when the substitutions are done with ions of the same column of the periodic table, then providing the same number of electrons to the conduction band, the appearance of NFL behaviour does not need disorder to be explained. This occurs in $CeRh_2Si_2$ under pressure, $CeRu_2(Ge_{1-x}Si_x)_2$ [15], $CeCu_{6-x}Au_x$ [46] or U_2Co_2Sn [47], and also in $CePd_{0.05}Ni_{0.95}$ [26], that was already cited in the introduction. In this last compound, the Kondo effect breaks down just at the composition at which the long-range order disappears, without any implication of disorder effects. This is supported by the X-ray absorption spectroscopy measurements performed in the parallel “strong coupling” $CePt_{1-x}Ni_x$ system [25].

The experimental results presented here, together with the arguments developed in this discussion, do not allow to extract a definitive conclusion about the origin of the NFL behaviour. However, we are more inclined to consider an inhomogeneous magnetic picture as the most adequate microscopic description of these strong coupling “Kondo hole” compounds, and then, the disorder driven theories would be at the basis of their magnetic behaviour. Certainly, this is not the thorough and definitive description of this system. On the one hand, extended theories are needed in two different directions: considering the interplay between spin fluctuations and disorder, as already started by some authors [48], and considering inhomogeneous distributions of interacting clusters, as claimed by other ones [20]. On the other hand, more experiments involving lower temperatures and thermal treatments (as done for UCu_4Pd [20,21]) could offer us new clues to understand this complex system.

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