

Low-temperature properties of the $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ system

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Received 9 April 2003 / Received in final form 2 September 2003

Published online 15 March 2004 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2004

Abstract. The influence of substituting Pd by Ni is described in $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ alloys with x taken up to about 0.25. Thermal and magnetization measurements point out a transition from a non-magnetic state (CePd_3) to a ferromagnetic state for $x > 0.05$, with a Curie temperature $T_c \approx 2$ K. The Ce- $L_{2,3}$ absorption edges and magnetic circular dichroism (XMCD) study reveals the coexistence of strong $4f$ hybridization and ferromagnetic order. The Ce- $L_{2,3}$ XMCD signal measured in CePd_3 demonstrates that in the Ce-based dense Kondo materials only the $4f^1$ channel gives a magnetic response.

PACS. 75.20.Hr Local moment in compounds and alloys; Kondo effect, valence fluctuations, heavy fermions – 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

Magnetism of Ce intermetallic compounds is known depending on the degree of localization of the $4f$ states to exhibit a wide variety of behaviours from Kondo systems, itinerant ferromagnetics to strongly localized magnets. For many years this subject has attracted a lot of interest of both experimentalists and theoreticians (see for example: [1–3]). A very illustrative example of the manifold behavior is magnetism of α and γ phases of Ce metal (for recent theoretical works see [4,5]). Another intriguing examples are Ce-Pd intermetallics: i) A typical Kondo system CePd_7 [6] with a Kondo temperature T_K of about 1000 K and a Ce-Ce interatomic distance of 5.67 Å. Let us just recall that T_K is the temperature below which the local moment ($4f$ in case of Ce) is compensated by the conduction electrons. ii) CePd_3 has a much lower T_K of about 240 K and a shorter Ce-Ce distance of 4.13 Å; $4f$ states in this compound are less hybridized with conduction electrons, but now a non-negligible Ce-Ce inter-site magnetic exchange coupling is present, that could lead to the magnetic ordering of Ce moments under certain conditions.

The latter compound is indeed the most extensively studied Ce-based dense Kondo material. It is generally accepted that the ground state of CePd_3 can be described as a non-magnetic (NM) Fermi-Liquid (FL). The electric resistivity ρ as a function of temperature shows a broad peak around 120 K, which is very close to the maximum in magnetic susceptibility. At lower temperatures resistivity decreases, indicating the formation of a coherent FL

state [7]. This and other measurements [9,10] suggest that the Kondo temperature of CePd_3 is about 240 K. Moreover, resonant inverse photoemission spectra at the Ce $4d$ threshold [10] show significant changes in $4f$ peak intensity in temperature range from 20 to 290 K. This observation indicates the tendency for the $4f$ electrons to become more localized at high temperatures, even though they are still itinerant around T_K .

Veenhuizen et al. [11] investigated the influence of substitutions on the low temperature properties of CePd_3 , especially in $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ alloys. On the basis of the temperature dependent susceptibility measurements the authors suggested that Ce has a tendency to a more localized $4f^1$ ground state with increase of the Ni content up to 0.2. Magnetization measurements of $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ alloys recorded at 4.2 K in field up to 35 tesla for [11] was described by the following equation:

$$M(B) = M_0 + \chi_{intr}B + bB^3, \quad (1)$$

where χ_{intr} is the intrinsic susceptibility of CePd_3 and M_0 is the magnetization of the “Ni impurity” in saturation. That means that the intrinsic susceptibility of CePd_3 is not suppressed by substitution of Pd for Ni. However, this study does not give any indication about possible transition to a ferromagnetic phase in these systems.

This kind of transitions has already been observed few years ago in $\text{CePd}_{1-x}\text{Ni}_x$ alloys, in which the Ce sublattice remains basically unchanged with the same CrB structure [12]. The highly localized $4f^1$ state of CePd leads to a ferromagnetic ordering below the Curie temperature $T_c = 6.5$ K, whereas CeNi shows the characteristic behaviour of a Kondo system. Thus, starting from CePd,

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$T_c(x)$ increases from 6.5 K, in the $0 < x < 0.5$ concentration range, to reach a plateau of about 10.5 K, in the $0.5 < x < 0.8$ range. Then, at higher Ni concentration, the T_c values decrease and further steeply drop to zero near $x = 0.95$ [13, 14]. This system has been cited in Stewart's review [14] for its non-Fermi-liquid behaviour with T_c suppressed to zero via Ni doping. Let us recall that, in general, the Fermi-liquid provides a good description of the low-temperature measurable parameters of a metal as long as the electron interactions, while T tends to zero, become temperature independent and are short ranged in both space and time. On the opposite, a non-Fermi-liquid behaviour is exhibited for a system with electron-electron interactions that are too strong to permit entry into the FL ground state at low temperatures. For the $\text{CePd}_{0.05}\text{Ni}_{0.95}$ system, an obvious explanation is probably given by the nearness to a magnetic instability in the phase diagram.

In the present paper we would like to further investigate the low temperature properties of $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ alloys, when x varies from 0 to 0.25. These systems have been characterized by magnetization measurements up to 4 T at low temperatures down to $T = 50$ mK. Temperature dependent specific heat measurements have been performed in the 0.5–20 K temperature range. Special attention is devoted to the Ce- $L_{2,3}$ absorption edges (XAS) and the magnetic circular dichroism (XMCD) spectra in order to precise the electronic and magnetic ground state of Ce.

2 Sample preparation and X-ray diffraction characterization

For this study, polycrystalline samples were prepared under a high purity argon atmosphere by triarc melting of Ce, Pd and Ni in a water-cooled copper hearth furnace, using tungsten electrodes. Appropriate amounts of pure Ce (99.99 wt.%), Pd (99.9 wt.%) and Ni (99.99 wt.%) were mixed respective binary 1:3 ratio for CePd_3 compounds. The nickel contents in the alloys varied from 0 up to 25 at %Ni, above this concentration the solid solution is not anymore well defined. In order to ensure chemical homogeneity the arc-melted buttons were turned over and re-melted ten times. Weight losses during melting were lower than 0.2 wt.%. The as-cast $\text{Ce}(\text{Pd},\text{Ni})_3$ alloys were characterized by X-ray diffraction (XRD) analysis. The XRD experiments on powders, with an average grain size $< 100 \mu\text{m}$, were performed at room temperature using monochromatic $\text{CuK}\alpha_1$ radiation. XRD patterns showed that all the compounds are crystallized in the proper Cu_3Au structure without any observable second phase. As no superlattice reflection has been detected, we suppose that the samples under study are characterized by a disordered Cu_3Au structure.

3 Experimental results and discussion

3.1 Lattice parameter

Figure 1 shows the lattice parameters i) as a function of Ni concentration x , with x taken up to 25 at% Ni, the

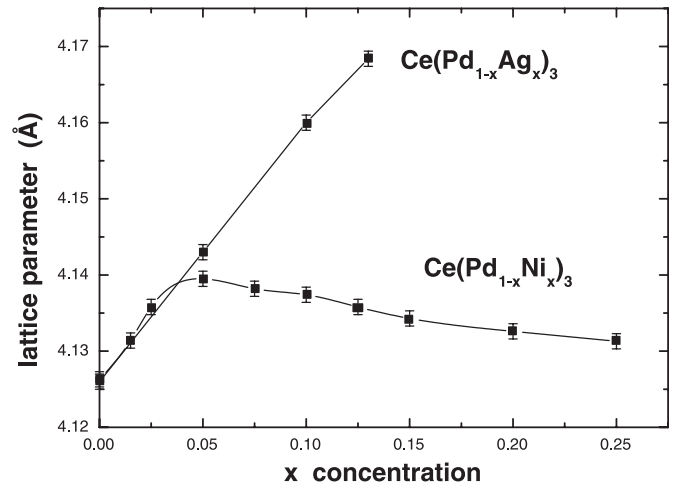


Fig. 1. Lattice parameter versus concentration x for various $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ alloys with $0 \leq x \leq 0.25$ and $\text{Ce}(\text{Pd}_{1-x}\text{Ag}_x)_3$ for $0 \leq x \leq 0.13$. The continuous lines are a guide for the eyes.

solubility limit of Ni in CePd_3 , ii) as a function of Ag concentration x , with x taken up to 13 at% Ag. At low concentration, up to $x = 0.05$, the Ni substitution drives the Ce atoms towards the $4f^1$ state limit which corresponds to a lattice parameter of about 4.16 Å (see [15] for similar situations). The lattice parameter reaches a broad maximum around $x = 0.05$ and then decreases with increasing concentration, exhibiting a complex behaviour of the Ce- $4f$ configuration not yet understood up to now. At this stage, it should be mentioned that electrical resistivity measurements $\rho(T)$ were carried out by Veenhuizen et al. [11] on similar samples. They reported that T_{max} (corresponding to ρ_{max}), which reaches a minimum value for $x = 0.05$, may be associated with the observation of the broad maximum of the lattice parameter around this x value and decrease again with increasing x . It is also interesting to point out that for a concentration equal to $x = 0.25$ the lattice parameter of the solid solution is rather close to the one for pure CePd_3 .

3.2 Magnetization

The magnetization were studied for $x < 0.20$ in the 2–20 K temperature range (not shown here) and for $x \geq 0.20$ down to $T = 50$ mK in fields up to 4 teslas. In Figure 2 we show the magnetization curves measured for $\text{Ce}(\text{Pd}_{0.75}\text{Ni}_{0.25})_3$. As the temperature is lower than 2 K the saturation of the magnetization is observed with a corresponding opening of the hysteresis loop with a coercive field about 0.1 T (inset of Fig. 2). These results evidence the existence of a ferromagnetic state below $T_c \approx 2$ K (T_c is defined at the inflexion point of the remnant magnetization variation).

The saturated magnetic moment extracted from the magnetization measurements at 50 mT is about $M_S = 0.15 \mu_B/\text{fu}$. This low M_S value can be explained by the Kondo effect acting on the Ce ground state, reducing the moment of the Γ_7 state in a cubic crystalline field,

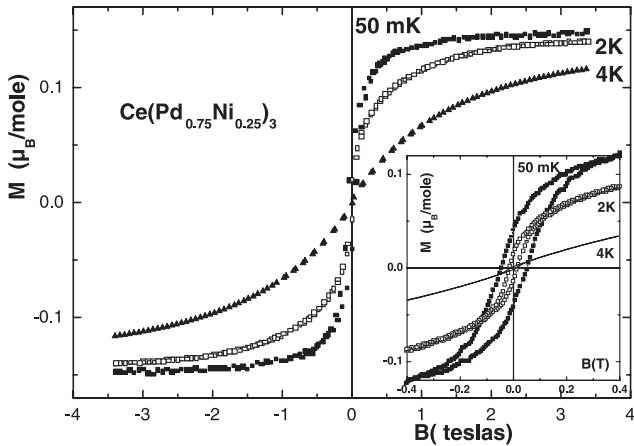


Fig. 2. Magnetization of $\text{Ce}(\text{Pd}_{0.75}\text{Ni}_{0.25})_3$ versus field for $T \leq 4$ K. Inset: Magnetization in magnetic field ≤ 0.4 T.

expected to be equal to $0.7 \mu_B$. Note that the saturation observed in the $M(B)$ is in conflict with the relation (1). A similar magnetic behaviour is also observed for the solid solution with $x = 0.20$.

3.3 Specific heat

We have studied the overall behaviour of $C_M(T)$ in the 0.5–20 K temperature range for the series of $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ samples and for comparison the $\text{Ce}(\text{Pd}_{0.87}\text{Ag}_{0.13})_3$ alloy. Results of specific heat measurements are presented in Figure 3 as a function of T . Very interesting is the almost fixed and pronounced peak which appears at $T = 1.45$ K for the various C_M curves of all samples with $x \geq 0.1$: the amplitude of this peak increases with Ni concentration but is absent in pure CePd_3 , thus it should be directly related to the Ni effect on a CePd_3 compound and more precisely to a magnetic transition, as illustrated by the magnetization measurements at low temperatures. Let us note that the specific heat peaks are as broad as the width of the T dependence of the remnant magnetization, both variations vanish around 4 K.

Compared to CePd_3 one may conclude that by increasing the Ce-Ce spacing a little bit from 4.126 Å for CePd_3 to a maximum of 4.140 Å for $\text{Ce}(\text{Pd}_{0.95}\text{Ni}_{0.05})_3$ (see Fig. 1) does not prevent the disappearance of the NM FL behaviour and the creation, instead, of a magnetically ordered state. Probably the d conduction band as well as the $4f$ - d hybridization change sufficiently from a Ce-Pd to a Ce-Ni environment to give rise to these different behaviours. In Figure 3 we also presented results of a $\text{Ce}(\text{Pd}_{0.87}\text{Ag}_{0.13})_3$ alloy: the significant increase of the electronic specific heat term, defined as the limit $(C/T)_{T \rightarrow 0}$, (about $150 \text{ mJ/K}^2\text{mol}$ for this alloy, compared to $38 \text{ mJ/K}^2\text{mol}$ for CePd_3) suggests that a heavy fermion state characterized $\text{Ce}(\text{Pd}_{0.87}\text{Ag}_{0.13})_3$ and CePd_3 , instead the specific effect of Ni impurities. The magnetic transition, with almost constant lattice parameters suggests that this transition comes more from an electronic effect than from a chemical pressure effect, as already observed in $\text{CePd}_{1-x}\text{M}_x$ system [16]. This is well illustrated by the

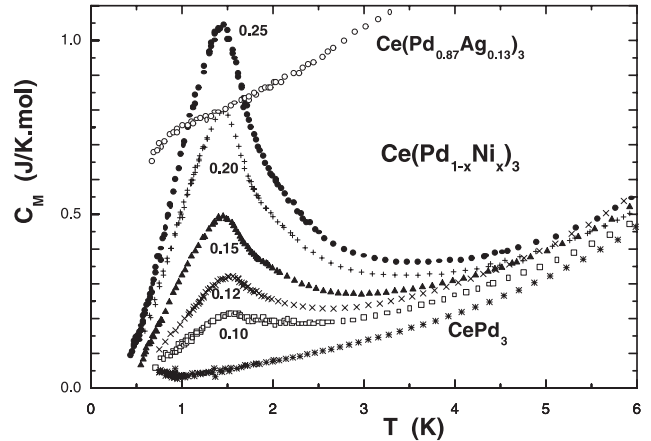


Fig. 3. Temperature dependence, for $0.5 \leq T \leq 6$ K, of the molar specific heat for various $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ intermetallic alloys, with $0 \leq x \leq 0.25$ and of the $\text{Ce}(\text{Pd}_{0.87}\text{Ag}_{0.13})_3$ alloy.

very different behaviour observed in the $\text{Ce}(\text{Pd},\text{Ag})_3$ system which does not show any magnetic anomaly in the $C(T)$ variation.

3.4 X-ray absorption and X-ray magnetic circular dichroism

The physical properties of Ce based compounds are profoundly related to the nature of the $4f$ electronic states. As already mentioned in the introduction, the reference is the isostructural α -Ce \rightarrow γ -Ce transition, interpreted as a transition from a well localized $4f$ state towards an itinerant $4f$ state. One of the key parameters which govern this behaviour is the hybridization of the $4f$ and conduction states [17]. This hybridization usually yields a NM ground state even at low temperature.

The $4f$ electronic configuration of Ce can be explored by XAS at the $L_{2,3}$ edges ($2p \rightarrow 5d$ transitions) and/or at the $M_{4,5}$ edges ($3d \rightarrow 4f$) [18]. The $2p \rightarrow 5d$ dipolar transitions might be used to probe the $5d$ band magnetism of the rare earths. In the case of highly correlated Ce systems, the $L_{2,3}$ spectra give more insight of the degree of $4f$ hybridization [20]. Moreover, the direct measure of the $4f$ moment could be done at the $M_{4,5}$ edges, unfortunately the current state of the low energy beam line possibilities in the soft X-ray range (≈ 800 eV) does not allow to measure weak signal for NM system. The XAS and XMCD experiments were performed at the Ce- $L_{2,3}$ edges in the $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ system. XMCD spectra were recorded at 4.5 K under 0–7 T magnetic fields applied perpendicularly to the surface of the polycrystalline sample (parallel or antiparallel to the photon propagation direction). The XAS and XMCD spectra are recorded on the ESRF ID12 beam line [19]. The circular polarization rate is, in the 5.5–6.2 keV energy range, in excess of 0.82.

3.4.1 XAS spectra at the Ce- $L_{2,3}$ edges

The Ce- $L_{2,3}$ X-ray absorption spectra in CePd_3 and $\text{Ce}(\text{Pd}_{1-x}\text{Ni}_x)_3$ are reproduced in Figure 4. As expected

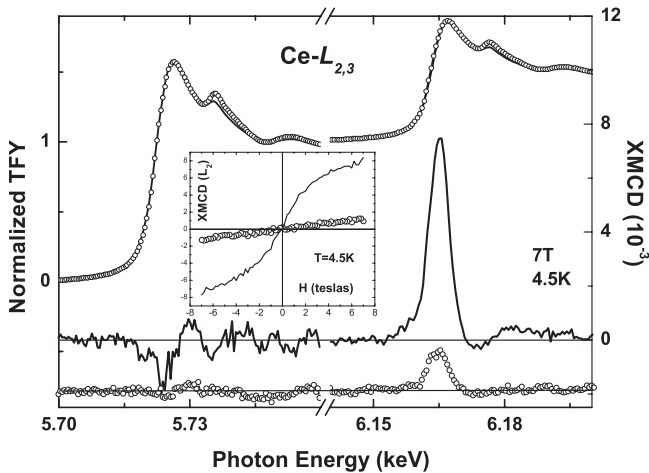


Fig. 4. XAS and XMCD at the Ce- $L_{2,3}$ edges in CePd₃ (open cycles) and Ce(Pd_{0.75}Ni_{0.25})₃ samples (line), at $T = 4.5$ K and $H = 7$ T. The CePd₃ XMCD is shifted to facilitate comparison. Inset: magnetic field dependence of the maximum XMCD signal.

they exhibit the double peak structure which is characteristic of the highly correlated Ce systems [20]: the ground state is a mixture of $|4f^1\rangle$ and $|4f^0\rangle$ states, that can be described as $|\psi_i\rangle = \alpha |4f^1\rangle + \beta |4f^0\rangle$. The double structure is, in fact, a signature of the two final states of optical transitions associated with the $4f^1$ and $4f^0$ channels. It reflects the two possible screening mechanisms of the $2p$ core hole, via $4f$ electrons or $5d$ band electrons [17]. The high-energy shoulder contribution (split from the main peak by $\Delta E(4f^1 - 4f^0) \approx 10$ eV) which was considered somewhat a fingerprint of the degree of the $4f$ hybridization in the initial state, can be taken as a measure of the fractional $4f$ occupation number n_f as well as the ground-state Kondo temperature. For the Ni doped alloy it is remarkable to note that, despite the magnetic state of this sample, the double peak structure is still observed. However, as illustrated in Figure 4, a decrease of the $4f^0$ to $4f^1$ intensity ratio leads to a n_f increasing of about 5%, when going from CePd₃ and to an alloy with $x = 0.25$.

3.4.2 XMCD at the Ce- $L_{2,3}$ edges

From XMCD studies in Ce based-systems [20] it is well known that two different cases have to be considered:

i) For well localized $4f^1$ systems, such as CePd, CeRu₂Ge₂, CeH₂ [18–20] without $3d$ partner (in other words the magnetic order is only due to the Ce atoms) the $5d$ -XMCD response is well established at once by the shape, the sign and the dichroic branching ratio. Such a reference can be given by CeRu₂Ge₂ [23]: one negative (positive) peak at the L_3 (L_2) edge, the XMCD(L_2/L_3) branching ratio being about -4 . These systems are characterized by $n_f \approx 1$ and magnetic order generally develops at low temperature ($T < 10$ K).

ii) For $4f^1$ systems with $3d$ partners, like CeFe₂, CeCo₅, Ce/Fe multi-layers [20–22], a lot of experiments have

shown that the $5d$ -XMCD signal is characterized by a double-peak structure (a replica of the edge) which indicates that the two channels are magnetic. This is due to the strong exchange $5d-3d$. Moreover, the sign (opposite to the well localized $4f^1$ systems) and the branching ratio (-1) of the dichroic signals are in agreement with the model predicted by the band theory. This is easily explained by the small contribution of the $4f$ shell, only the $5d$ spin polarization determines the dichroic signal. These systems are characterized by $n_f < 1$ and by ferromagnetic orders (mainly due to the $3d$ metal) with high Curie temperatures. The magnetic state of Ce is due to a $3d$ induced polarization effect.

Let us now consider the magnetic dichroism results in the system under study:

i) Ce(Pd_{0.75}Ni_{0.25})₃ alloy. In both $L_{2,3}$ edges the magnetic contributions consists of a main structure and possibly some tiny structures at high energy (see Fig. 4). Each main peak points 2 eV before the edge maximum, with a line width (≈ 5 eV) much narrower than the edge line (≈ 10 eV). The sign of the dichroic response is positive (negative) at the L_2 , (L_3) which corresponds to a $5d$ moment parallel to the applied magnetic field. The normalized amplitudes of the dichroic signals are about 2×10^{-3} and 8×10^{-3} at the L_3 and L_2 , respectively. The integrated XMCD signals at the $L_{2,3}$ edges give a $5d$ -XMCD branching ratio close to -4 . This reproduces the expected behavior for a well localized $4f^1$ system, as in CeRu₂Ge₂ or CeH₂/Fe multilayers [24], and consequently demonstrates that the main source of magnetism, in the Ce(Pd_{1-x}Ni_x)₃ system, is due to the Ce atoms. Preliminary XMCD measurements at the Ce- $M_{4,5}$ and Ni- $L_{2,3}$ edges show that Ce atoms bear a $4f^1$ moment and that Ni atom moment is below the actual detection limit.

ii) CePd₃ compound. The observation, at the Ce- L_2 edge (Fig. 4), of a dichroic signal in CePd₃ shows the extreme sensitivity of XMCD technique; the signal corresponds to a magnetic field induced effect only due to the $5d$ paramagnetic susceptibility, part of the total susceptibility $\chi_0(4\text{ K}) = 1.2 \times 10^{-3} \text{ cm}^3/\text{mol}$ [24]. The small amplitude (only 1.8×10^{-3}) explains the lack of signal at the L_3 edge; indeed if we suppose a branching ratio about -4 , as observed for $x = 0.25$, the amplitude of the L_3 signal should be approximately -0.5×10^{-3} , which would require by far more beam time that used for the present experiments. The magnetic field dependence of the maximum dichroic signal, presented in the inset of Figure 4, demonstrates, if needed, the validity of these experiments, as they well reproduce the magnetic character of the different samples, specially the linear variation for CePd₃.

Let us highlight that the observation of only one magnetic peak corresponding to the main $4f^1$ final state demonstrates that only this channel leads to a magnetic response, in contrast to the early studies in highly hybridized Ce systems with $3d$ partners, characterized by a double peak in the dichroic line.

Is it possible to propose a quantitative analysis of these data? It has been already discussed that the sum rules [25] do not apply in the case of $5d$ band of RE systems, due to

the $4f$ exchange. However, it is possible to make a rough comparison of the dichroic signals with those of CeRu₂Ge₂ where we know the $M_{5d}(4K)$ moment value ($\approx 0.1\mu_B$). As the ratio between the two dichroic signals is about 10, M_{5d} is approximately $0.01\mu_B$ in Ce(Pd_{0.75}Ni_{0.25})₃ and $4 \times 10^{-3}\mu_B$ in CePd₃.

4 Conclusion

This first detailed study of the thermal and magnetic properties of the series of Ce(Pd_{1-x}Ni_x)₃ solid solutions with x comprised between 0 and 0.25 shows that the considered alloys undergo a transition from a typical non-magnetic FL behaviour to a magnetically ordered system due to a few percent of Ni alloying. In other words we quit the FL regime at low concentration of Ni (when $x \geq 0.05$). We also reported on the field induced dichroism on pure CePd₃ and demonstrated that only the $4f^1$ channel contributes to the XMCD signal.

The authors express their thanks to J.G. Sereni for fruitful discussions, as well to Profs A. Kotani and S. Shin at an early stage of this work. W. Wernsdorfer and Mr E. Eyraud are gratefully acknowledged for low temperature magnetization measurements and J.-P. Lambour for specific heat measurements. We warmly thank the unknown referees for their constructive criticism.

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