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Local magnetism and Kondo behaviour of Ce in U-substituted CePd₃

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Abstract

We report bulk and local magnetic studies in single-phase cubic (AuCu₃-type) Ce_{1-x}U_xPd₃ intermetallic alloys for $0 \leq x \leq 0.3$. Bulk magnetization data for all compositions exhibit paramagnetic behaviour that shows enhanced effective magnetic moment μ_{eff} and progressive reduction of θ_{p} with U content. The data—combined with the local susceptibility (χ_{loc}) of ¹⁴⁰Ce, measured by time-differential perturbed angular correlation spectroscopy—reveal a smooth cross-over from intermediate valence to nearly trivalent local moment behaviour with a sharp diminution of Kondo temperature T_{K} with x . We believe that 5f-band electrons of U strongly influence the magnetism and Kondo behaviour of Ce ions in these alloys.

1. Introduction

Depending on the degree of f conduction electron hybridization strength, Ce and U intermetallic alloys exhibit many interesting phenomena, namely the Kondo effect, valence fluctuation, heavy fermion behaviour etc. As such, electron correlation in these materials has been a major topic of experimental and theoretical study over the past several years. In particular, the compounds CePd₃ and UPd₃ have attracted considerable interest. The former, which has cubic (AuCu₃) crystal structure, has been classified as an archetypal valence fluctuating system with a high 4f spin fluctuation rate that is characterized by a large Kondo temperature $T_{\text{K}} \sim 500$ K [1–4]. On the other hand, UPd₃ forming in the Ni₃Ti-type hexagonal structure has been identified as showing classical local moment behaviour [5]. Over the past several years extensive studies have been undertaken on many alloy series of the type $M_{1-x}U_xPd_3$ with $M = \text{Sc, Y, La, Pr, Zr, and Th}$ [6]. Alloys with trivalent elements ($M = \text{Sc, Y, La, Pr}$) crystallize in cubic AuCu₃ structure. For tetravalent elements ($M = \text{Zr, Th}$) the alloys form in hexagonal UPd₃ structure. More importantly, from magnetic and transport studies it has been reported that alloys with $M = \text{La or Pr}$ exhibit Fermi liquid behaviour with very

Table 1. A summary of structural and magnetic data for $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys.

x	a (Å)	χ_0 (10^{-3} emu mol $^{-1}$)	μ_{eff} (μ_{B})	θ_{p} (K)	T_{K} (Ce)
0.0	4.125(5)	1.37(8)	2.08	−320(25)	296(20)
0.05	4.122(5)	1.66(8)	2.27	−247(15)	203(10)
0.10	4.120(5)	1.86(10)	2.42	−180(10)	135(10)
0.30	4.115(5)	2.60(10)	2.83	−120(10)	90(10)

low Kondo temperature, whereas compounds with $M = \text{Sc}$ or Y with large T_{K} values show non-Fermi liquid behaviour at low temperatures [6]. Thus it is of interest to study the structural and magnetic properties of alloys when M is an element with intermediate valency (e.g. Ce).

The 4f instability in CePd_3 has also been studied extensively in several pseudo-binary systems of the type $\text{Ce}_{1-x}\text{M}_x\text{Pd}_3$ ($M = \text{La}, \text{Y}, \text{Sc}$ etc) and $\text{Ce}(\text{Pd}_{1-x}\text{T}_x)_3$ (where T is a transition element, namely Ni, Rh, Ag, Pt etc) [7–9]. Ce site substitution by La, Y and Sc drives the system towards a non-magnetic (tetravalent) ground state; replacement of Pd by other transition metals, namely Ni, Ag, Rh etc, leads to trivalent/tetravalent behaviour, depending on the chemical pressure induced by the solvent atom. For example, Ni and Ag substitution drive Ce ions towards trivalency, whereas Rh and Pt causes a rapid loss of the Ce moment (tetravalency). These features of Ce magnetic behaviour have been explained as being due to a decrease/increase in the Kondo temperature T_{K} proportional to the Ce-4f spin fluctuation rate arising from changes in hybridization strength as a consequence of unit cell expansion or contraction. Despite the large volume of data available, it is not clear how different types of conduction electrons influence the 4f spin fluctuation rate. From a recent study of the local magnetism for extremely dilute Ce impurity in U metal, it has been proposed that—in spite of a large hybridization strength—the f–f interaction between the Ce-4f and the host U-5f band electrons suppresses spin fluctuation rates and stabilizes the magnetic moment [10, 11]. In connection to this, it is of interest to study the magnetic behaviour of Ce in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys. Such investigations are likely to provide direct information on the role of f–f interaction on the magnetism and Kondo behaviour of Ce ions. In this paper we present the magnetic behaviour of Ce ions in single-phase alloys of $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ ($0.0 \leq x \leq 0.3$) crystallized in a AuCu_3 -type cubic structure. With a progressive increase in U content, in spite of a negligible change in hybridization strength scaled by the measured lattice constant, the magnetic response of Ce obtained from measurements of bulk susceptibility and magnetic hyperfine field of ^{140}Ce 's reveal a large reduction in Kondo temperature T_{K} . We believe that the Kondo behaviour of Ce in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys is strongly influenced by the interaction between Ce-4f and U-5f band electrons.

2. Experimental details

Polycrystalline alloys of $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ were prepared by melting stoichiometric amounts of constituent elements in an argon arc furnace. The samples were characterized by room-temperature powder x-ray diffraction measurement. Alloys with $0.0 \leq x \leq 0.3$ were found to be single phase with AuCu_3 -type FCC crystal structure. For $x > 0.3$ the alloys were found to be multi-phase. The lattice constants a , determined from the (422) reflection, are listed in table 1. They show a small decrease from 4.125(5) Å for $x = 0$ to 4.115(5) Å at $x = 0.3$. It is interesting to point out that extrapolation of this data yielded $a = 4.102 \pm 0.005$ Å for hypothetical cubic UPd_3 . This shows excellent agreement with results reported by Seaman *et al* [12]. Bulk magnetization measurements were carried out in the temperature

range 4.2–300 K and in applied magnetic field up to 5.5 T using a superconducting quantum interference device (SQUID) magnetometer.

Complementary to the bulk magnetic studies, we carried out measurements of local susceptibility for Ce ions using the time-differential perturbed angular correlation (TDPAC) technique. The basic principle of the PAC technique lies in the directional correlation of γ -radiation emitted by nuclei with a preferred spin orientation and its modification by the hyperfine interaction between the nucleus and the electromagnetic fields produced by the surrounding atomic environment. Experimentally, a trace quantity of radioactive impurity is introduced into the sample under investigation. One then measures the hyperfine fields acting on the daughter nuclei as a function of physical variables such as temperature, pressure, concentration etc. In this study, we have used the probe ^{140}Ce produced via beta decay of ^{140}La . The 2084 keV, $I^\pi = 4^+$ state in ^{140}Ce with a half-life of $T_{1/2} = 3.5$ ns and a nuclear g -factor of $g_N = 1.123$ [13] serves as a sensitive probe for the detection of the magnetic hyperfine field, which in turn provides microscopic information on the static and dynamic magnetic response of Ce ions in the sample being studied. A detailed description of the PAC technique can be found in [14].

The parent ^{140}La activity was obtained by irradiating a piece of natural La metal with thermal neutrons from the CIRUS/DHRUVA reactor at BARC, Mumbai. The ^{140}La activity was introduced into the alloys through electron-beam melting under high vacuum. The concentration of La in the alloys was maintained below 0.1 at.%. The PAC spectra were recorded using a set-up that consisted of three NaI(Tl) detectors in a $\pm 135^\circ$ geometry and a standard slow-fast coincidence circuit. Measurements were carried out in an external magnetic field of 1.4 T from an electromagnet. The sample temperature was varied in the range 25–300 K using a closed-cycle He refrigerator.

In the presence of a magnetic field, the nuclear spin precesses with the Larmor frequency ω_L , resulting in a time-dependent modulation of the γ -ray intensity. From the background subtracted normalized coincidence counts $N(\theta, t)$ of each detector, spin rotation spectra $R(t)$ were generated by taking the ratio

$$R(t) = \frac{[N(135^\circ, t) - N(-135^\circ, t)]}{[N(135^\circ, t) + N(-135^\circ, t)]}.$$

These were fitted to the function

$$R(t) = \frac{3}{4} A_{22} \sin(2\omega_L t)$$

to extract the value of ω_L and the paramagnetic enhancement factor $\beta = \hbar\omega_L / (g_N \mu_N B_{\text{ext}})$. The local susceptibility χ_{loc} of Ce in the alloy can be estimated from the measured $\beta(T)$ data using the relation [15]

$$\chi_{\text{loc}} = \frac{N(\beta - 1)g_J \mu_B J}{B(0)}$$

where N is the Avogadro number and $B(0)$ is the zero-temperature hyperfine field, which is known to be 180 T for Ce^{3+} ions [16].

3. Results and discussion

Figure 1 shows the temperature dependence of the bulk magnetic susceptibility (χ) measured in an applied field of 1.0 T. Inset (a) in figure 1 displays the plot of inverse susceptibility (χ^{-1}) as a function of temperature. For the parent alloy CePd_3 , the value of χ was small and was found to be weakly temperature dependent, showing a broad maximum at around 150 K. This is consistent with the results reported in the literature [2]. From the data presented in figure 1

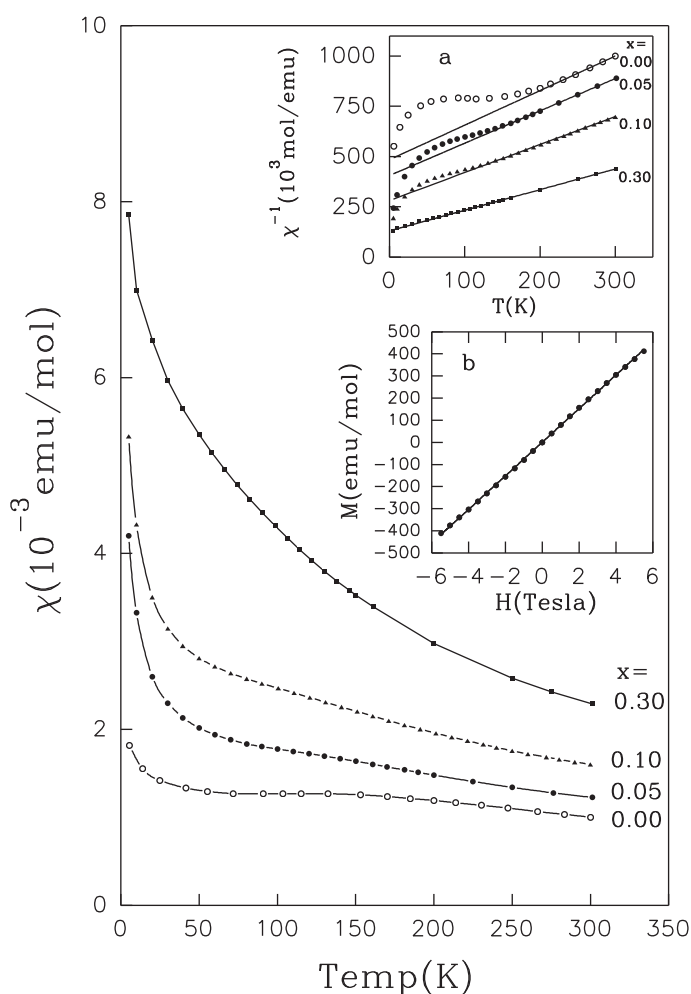


Figure 1. Magnetic susceptibility (χ) versus temperature for $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys measured in an applied field of $H = 1.0$ T. The top inset shows χ^{-1} versus T for different x values, from which μ_{eff} and θ_{p} were extracted. The bottom inset shows M versus H data for $x = 0.3$ at 4.2 K.

it can easily be seen that the behaviour of $\chi(T)$ changes remarkably with U substitution. With increasing U content, the value of χ grows and also shows much stronger temperature dependence. Below 50 K, the $\chi(T)$ data for all compositions show a sharp up-turn, which may be due to small amounts (<1%) of unidentified impurity phases present in the samples. From magnetic and transport studies in $M_{1-x}\text{U}_x\text{Pd}_3$ ($M = \text{Sc}, \text{Y}, \text{La}$) it has been reported that samples with $x \geq 0.3$ show spin-glass/antiferromagnetic ordering with irreversible magnetization below a certain temperature [6]. To examine possible spin-glass/antiferromagnetic ordering in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$, we measured M versus H at 4.2 K for the $x = 0.3$ sample cooled under the zero-field condition. The data are shown in figure 1 (see inset (b)). It is evident that magnetization in $\text{Ce}_{0.7}\text{U}_{0.3}\text{Pd}_3$ varies linearly with H and does not show any hysteresis, indicating the absence of spin-glass-like freezing in our sample.

The magnetic susceptibility measured for the paramagnetic $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys can be expressed as

$$\chi(T) = \chi_0 + C/(T + \theta_P)$$

where χ_0 is the temperature-independent susceptibility due to conduction-band electrons, and the Curie–Weiss-like contribution arises mainly from the local magnetic moments of the Ce and U ions. From the slope of the χ^{-1} versus T plots (see inset (a) in figure 1) we estimated the effective magnetic moment per unit cell ($\mu_{\text{eff}}/\text{fu}$). The results are summarized in table 1. For the parent alloy CePd₃ our χ_0 , θ_P and μ_{eff} values agree closely with the results reported earlier [2]. The derived large negative θ_P value is consistent with the high Kondo temperature of Ce ions in this alloy. From the data presented in table 1 it is evident that the values of χ_0 and μ_{eff} increase progressively with U concentration in Ce_{1-x}U_xPd₃ alloys. On the other hand, the value of θ_P decreases rapidly with x . For a simple analysis of the magnetic data, the measured effective moment per unit cell can be expressed as

$$\mu_{\text{eff}}^2 = (1 - x)\mu_{\text{eff}}^2(\text{Ce}) + x\mu_{\text{eff}}^2(\text{U}).$$

Here, $\mu_{\text{eff}}(\text{Ce})$ and $\mu_{\text{eff}}(\text{U})$ are the effective moments of Ce and U atoms, respectively. Considering that U atoms in $M_{1-x}U_x\text{Pd}_3$ ($M = \text{Sc}, \text{Y}, \text{La}$ etc) alloys carry large local magnetic moments, ranging from 3.1 to 3.4 μ_B [6], they are likely to have similar magnetic properties in Ce_{1-x}U_xPd₃ alloys as well. Assuming the moment of U to be as high as 3.58 μ_B (the free-ion value of tetravalent U ions), the magnetic moment for Ce ions in Ce_{1-x}U_xPd₃ alloys was estimated to be 2.08, 2.18, 2.25 and 2.45 μ_B for $x = 0.0, 0.05, 0.10$ and 0.30, respectively. From these values of moment, the average valency of Ce ions in Ce_{1-x}U_xPd₃ could be estimated using the relation $\mu_{\text{Ce}}^2 = (4 - v)\mu_{\text{Ce}^{3+}}^2$, yielding $v = 3.32$ and 3.07 for $x = 0$ and 0.3, respectively. The above analysis shows that Ce ions change smoothly from an intermediate-valence to a nearly trivalent state with increasing U content in CePd₃. These features, together with the observed trend in θ_P values, indicate that U substitution in CePd₃ causes a large reduction in Ce-4f spin fluctuation and hence the value of T_K .

To confirm the influence of U substitution on the magnetism and spin fluctuation behaviour of Ce in Ce_{1-x}U_xPd₃ alloys, a microscopic investigation of the local magnetic susceptibility of ¹⁴⁰Ce was carried out using the TDPAC method. Figure 2 displays typical spin rotation spectra, $R(t)$, of ¹⁴⁰Ce in some of the Ce_{1-x}U_xPd₃ alloys measured in an applied magnetic field of 1.4 T. In all cases the recorded spectra show a single frequency with high anisotropy $A_{22} \sim -0.05$. Though slightly smaller than the maximum $A_{22} = -0.096$ reported for the 329–487 keV γ – γ cascade [17], the observed anisotropy (with solid-angle correction not applied) compares well with the A_{22} values observed for ¹⁴⁰Ce in many metallic systems [10, 18]. These observations indicate that the ¹⁴⁰La probe atoms occupy a unique lattice site in the alloy, most likely substituting a Ce atom. Figure 3 shows the paramagnetic enhancement factor β of Ce in various alloys measured as a function of temperature. Here $\beta(T) = 1$ indicates non-magnetic behaviour of Ce, expected for Ce⁴⁺ ions. For trivalent Ce ions with a stable magnetic moment, due to the large positive value of the hyperfine field ($B(0) = 180$ T) the $\beta(T)$ are positive and show Curie-like temperature dependence. In intermediate-valence systems with a highly unstable 4f electron, $\beta(T)$ values are expected to lie between these two extremes. The magnetic response of Ce in the parent alloy CePd₃ shows slightly positive $\beta(T)$ values, exhibiting weak temperature dependence with a broad maximum around 150 K. This feature is similar to that observed in bulk susceptibility data (see figure 1) and is consistent with a high fluctuation rate of Ce-4f electrons. Upon U substitution, the χ_{loc} of Ce grows progressively and shows stronger temperature dependence at higher values of x . For $x = 0.3$, the measured $\beta(T)$ is close to the behaviour expected for trivalent Ce ions. It is interesting to note that, for all compositions, $\beta(T)$ shows a clear maximum that is not visible in the bulk susceptibility data. As the U content in CePd₃ increases, the peak in $\beta(T)$ becomes more pronounced and shifts to lower temperature. The data presented above clearly

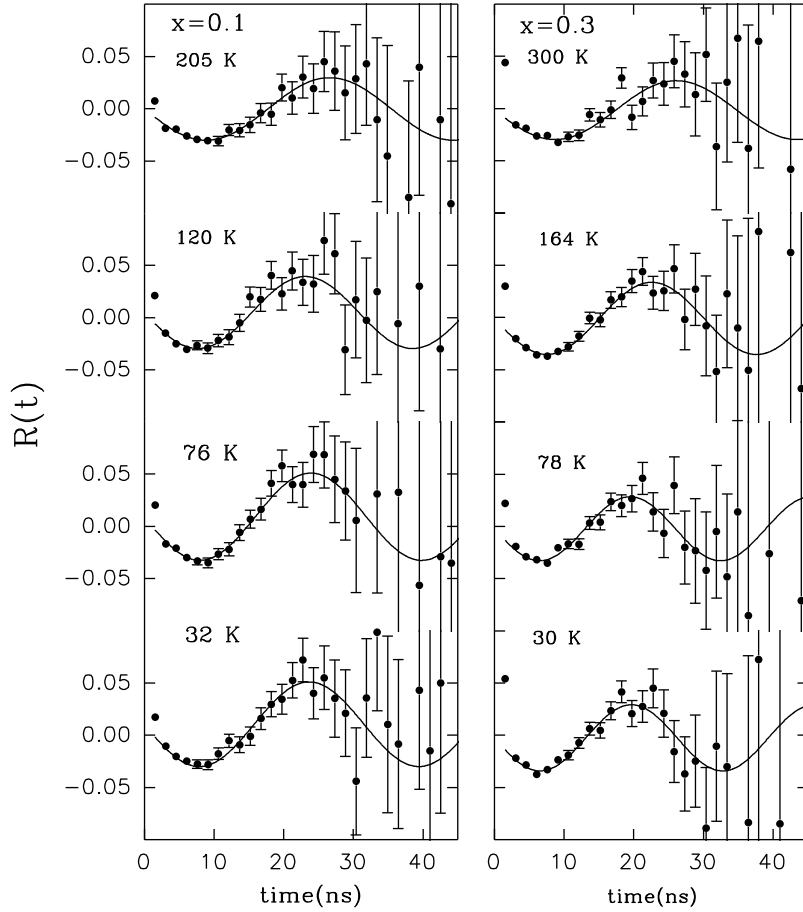


Figure 2. Typical spin rotation spectra, $R(t)$, for ^{140}Ce in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ for $x = 0.1$ and 0.3 , recorded in a magnetic field of $B_{\text{ext}} = 1.4$ T.

show a smooth transition from intermediate-valence to nearly trivalent behaviour for Ce as the U content in the alloy increases from $x = 0.0$ to 0.3 .

The local susceptibility data was analysed using the modified Coqblin–Schrieffer (CS) model, which was developed for orbitally degenerate Kondo ions [19]. In this case the dynamic susceptibility of Ce can be parameterized by a characteristic temperature, T_0 , and the zero-temperature susceptibility, χ_0 , is related by the equation

$$\chi_0 \sim \frac{N(N^2 - 1)\mu^2}{(24\pi k_B T_0)}$$

where $N = 2J + 1$ is the degeneracy factor. The Kondo temperature, T_K , can then be estimated from the relation $T_K/T_0 = 2\pi W(N)/N = 0.677$, where $W(N) = 0.6464$ is the Wilson number for $N = 6$ [20, 21]. From the numerical results given in [19], a fit of the experimentally measured $\beta(T)$ data yielded $T_K = 296 \pm 20$, 203 ± 10 , 135 ± 10 and 90 ± 5 K for $x = 0$, 0.05 , 0.10 and 0.30 , respectively. The results presented here clearly indicate that the addition of U into CePd_3 results in a considerable reduction in the Kondo temperature of Ce ions.

The Kondo temperature of a magnetic ion can be expressed as [22]

$$T_K \sim T_F \exp[-1/N(E_F)J] \sim T_F \exp[-\epsilon_f / \langle V_{kf} \rangle^2 N(E_F)]$$

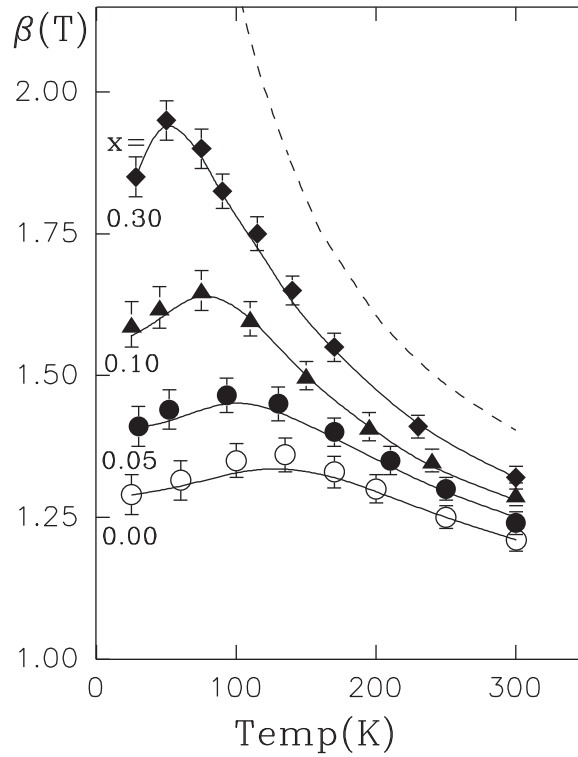


Figure 3. The temperature dependence of the paramagnetic enhancement factor, β , for ^{140}Ce in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys. The solid curves represent the fits to Rajan's model (see text). The dashed curve corresponds to the behaviour expected for a stable Ce^{3+} magnetic moment.

where T_F is the Fermi temperature, $N(E_F)$ is the density of states at E_F , $J \sim -\epsilon_f^{-1}[\langle V_{kf} \rangle]^2$ is the exchange interaction parameter (with ϵ_f being the position of the Ce-4f level with respect to the Fermi energy E_F), and $\langle V_{kf} \rangle$ is the hybridization matrix element, which in turn is proportional to the inverse of the unit cell volume. From the above relation it becomes evident that a reduction in T_K can arise due to a decrease in the value of $\langle V_{kf} \rangle$ (an increase in unit cell volume) and/or $N(E_F)$. The lattice constants measured for the alloys decrease marginally with x (see table 1), suggesting a small increase in $\langle V_{kf} \rangle$. Furthermore, upon U substitution in CePd_3 , the number of electrons per unit cell and hence $N(E_F)$ is expected to increase with x . Thus in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys, due to the larger $\langle V_{kf} \rangle$ and $N(E_F)$, Ce's T_K is expected to be higher compared to that in pure CePd_3 . In contrast, the data presented above clearly show a large reduction in T_K value. We believe that U-5f band electrons play an important role in lowering Ce's T_K in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys. From spectroscopic measurements as well as band structure calculations in Ce alloys including CePd_3 , it has been shown that the narrow 4f band is located $\sim 1-2$ eV below the Fermi energy, E_F [23]. Similarly, for cubic UPd_3 , local spin density calculations show a high density of states at E_F ($N(E_F) = 3.173$ states $\text{eV}^{-1}/\text{atom}$) that arises mainly from the 5f band electrons [24]. Thus, in the pseudo-binary system $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$, the 4f electrons of Ce are expected to overlap strongly with the 5f electrons of U, causing greater f-f interaction. Furthermore, as seen in $\text{Y}_{1-x}\text{U}_x\text{Pd}_3$ alloys [25], the position of the U-5f band shifts to lower energy with increasing x . Such a scenario in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$, if present, would lead to an even stronger overlap of the Ce-4f and U-5f energy bands. In addition, considering the high value of χ_0 in $\text{Ce}_{1-x}\text{U}_x\text{Pd}_3$ alloys (see table 1), strong f-f interaction between Ce-4f and U-5f may also

be possible indirectly via the polarization of conduction-band electrons. This f–f interaction can suppress spin fluctuations arising from f–d and f–sp interactions, leading to lower T_K values, analogous to the behaviour observed for dilute Ce impurities in U metal [10, 11].

In summary, we have performed bulk and local magnetic studies for single-phase alloys in the series $Ce_{1-x}U_xPd_3$ ($0 < x < 0.3$). With increasing U concentration, the Ce ions change smoothly from intermediate-valence to trivalent local moment behaviour with a progressive diminution in Kondo temperature, T_K . The data suggest that magnetism and Kondo behaviour of Ce in $Ce_{1-x}U_xPd_3$ alloys are strongly influenced by the f–f interaction between the Ce-4f and U-5f band electrons.

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