Magnetic ordering of Mn sublattice, dense Kondo lattice behavior of Ce in (RPd₃)₈Mn (R = La, Ce)

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Abstract

We have synthesized two new interstitial compounds (RPd₃)₈Mn (R = La and Ce). The Mn ions present in ‘dilute’ concentration of just 3 molar percent form a sublattice with an unusually large Mn-Mn near neighbor distance of $\sim 8.5$ Å. While the existence of (RPd₃)₈M (where M is a p-block element) is already documented in the literature, the present work reports for the first time the formation of this phase with M being a 3d element. In (LaPd₃)₈Mn, the Mn sub-lattice orders antiferromagnetically as inferred from the peaks in low-field magnetization at 48 K and 23 K. The latter peak progressively shifts towards lower temperatures in increasing magnetic field and disappears below 1.8 K in a field of $\sim 8$ kOe. On the other hand in (CePd₃)₈Mn the Mn sublattice undergoes a ferromagnetic transition around 35 K. The Ce ions form a dense Kondo-lattice and are in a paramagnetic state at least down to 1.5 K. A strongly correlated electronic ground state arising from Kondo effect is inferred from the large extrapolated value of $C/T = 275$ mJ/Ce-mol K$^2$ at $T = 0$ K. In contrast, the interstitial alloys RPd₃Mnx ($x = 0.03$ and 0.06), also synthesized for the first time, have a spin glass ground state due to the random distribution of the Mn ions over the available 1b sites in the parent RPd₃ crystal lattice.

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I. INTRODUCTION:

The RPd$_3$ compounds have the AuCu$_3$-type crystal structure with R and Pd atoms occupying the corners (1a) and face centers (3c) of the cubic unit cell, respectively. A unique way of alloying RPd$_3$ (R = La to Lu) compounds was reported by Dhar et al., when they observed that smaller p-block atoms like boron could be incorporated interstitially in the RPd$_3$ crystal lattice. The smaller boron atoms occupy the body-center (1b) position in the RPd$_3$ unit cell forming a defect pervoskite-type structure with formula RPd$_3$B$_x$ (0 < $x$ < 1). It was later found that silicon could also be incorporated in the RPd$_3$ unit cell up to $x \sim 0.3$. Alloying with boron and silicon resulted in the expansion of the lattice and changed drastically the valence and hence the magnetic properties of CePd$_3$ and EuPd$_3$. Briefly, cerium ions changed from valence fluctuating state in CePd$_3$ to trivalent state in CePd$_3$B$_x$ for $x > 0.12$, while the lattice expansion forced the europium ions to change from the trivalent state in EuPd$_3$ to the valence fluctuating state in EuPd$_3$B$_x$ alloys, with possible charge ordering in EuPd$_3$B.

An interesting extension of the above-mentioned interstitial alloying has recently been reported by Gordon and DiSalvo. They found that alloying CePd$_3$ with an appropriate amount of Sb leads to the formation of a cubic superstructure, (CePd$_3$)$_8$Sb, closely related to the AuCu$_3$-type structure of CePd$_3$. The large sized Sb atoms deform the Pd octahedra, such that the available 1b sites in the CePd$_3$ lattice are occupied by Sb atoms only in a regular pattern forming a simple cubic sublattice with near neighbor Sb–Sb separation slightly more than twice the lattice parameter of the parent CePd$_3$ unit cell. In the superstructure the Ce atom remains coordinated by 12 Pd atoms and the Ce–Ce separation is increased by 2 to 3% over its value in CePd$_3$. A rigorous discussion of the superstructure can be found in 6,7. Later, it was found that the superstructure (CePd$_3$)$_8$M is also formed for other p-block elements M = Ga, In, Sn, Pb and Bi, and for (LaPd$_3$)$_8$In. (CePd$_3$)$_8$M compounds order magnetically below 10 K in contrast to the Pauli paramagnetic, valence fluctuating ground state in CePd$_3$, and they exhibit heavy-fermion like enhanced low temperature electronic specific heat coefficient, $\gamma$. Further investigations showed that (CePd$_3$)$_8$M (M = Al and Ge) also form, order magnetically below 10 K and exhibit a dense Kondo lattice behavior. Jones et al. also tried to synthesise (CePd$_3$)$_8$M for M.
= Zn and Te. Though their powder x-ray diffraction patterns showed the signature of superstructure formation, there were some doubts about the homogeneity and the exact stoichiometry of the samples. The case for M = Zn is interesting in the sense that Zn is not a p–block element and it motivated us to search for other atoms in the 3d series that might lead to superstructure formation.

In the present work, we report for the first time the formation of ‘dilute-Mn’ ternary compounds (RPd₃)₈Mn for R = La and Ce. The word ‘dilute’ emphasizes the fact that the Mn concentration in these compounds is low (about few molar percent) and yet the Mn ions form a periodic lattice with unusually large nearest neighbor Mn–Mn separation allowing localization of the 3d moments. Recently such ‘dilute-Mn’ compounds have attracted considerable attention. Negative colossal magnetoresistance has been reported in the single crystals of Zintl compounds Eu₁₄MnSb₁₁ and Eu₁₄MnBi₁₁, despite antiferromagnetic ordering in the later compound²³. In Yb₁₄MnSb₁₁ (where Yb ions are divalent and hence non-magnetic) the localized Mn³⁺ moments are coupled via conduction electrons and order ferromagnetically at 53 K¹⁴. Yb₁₄MnBi₁₁ exhibits two magnetic transitions: a ferromagnetic transition at 58 K followed by a second transition at 28 K¹⁴. It was pointed out in ref. 14 that similar ‘dilute’ intermetallic compounds may be a good starting point to find 3d-Kondo lattice systems, analogous to the well known and extensively studied 4f-Kondo lattice/heavy fermion systems. We find that in (RPd₃)₈Mn (R = La, Ce) the nearest neighbor Mn–Mn separation is unusually large ~ 8.5 Å. In (LaPd₃)₈Mn the Mn ions are in a localized 2+ valence state and order antiferromagnetically below ~50 K. In (CePd₃)₈Mn the Ce ions form a dense Kondo lattice and do not order magnetically at least down to 1.5 K. However, the Mn sublattice orders ferromagnetically near 35 K. In contrast the alloys RPd₃Mnx (x = 0.03 and 0.06), also synthesized for the first time, have a spin glass ground state due to the random occupation of 1b sites by the Mn ions in the RPd₃ lattice.
II. EXPERIMENT

Initially compounds of the composition \((\text{R} \text{Pd}_3)_8 \text{Mn}\) for \(\text{R} = \text{La} \) and \(\text{Ce}\) were made by the standard technique of arc melting in an inert atmosphere of argon. First an ingot of \(\text{RPd}_3\) was melted and then Mn added to it by gently focusing the arc on the pellet of \(\text{RPd}_3\) so that Mn, which is volatile, is not directly heated. The button of the ternary alloy was flipped over and re-melted several times to ensure homogenization. The initial and final weights of the pellets are practically the same and hence there is no loss of Mn. We believe that the success of arc melting technique for making these samples is partially due to the fact that the molar percentage fraction of Mn is low \((\sim 3 \%)\). Later, \(\text{LaPd}_3\text{Mn}_x\) \((x = 0.03, 0.06)\) and \(\text{CePd}_3\text{Mn}_x\) \((x = 0.02, 0.06)\) were also made by the same method. The samples were checked for their phase purity by the standard powder x-ray diffraction technique and optical micrography. The resistance between 1.5–300 K was measured by the four-probe DC method on a rectangular parallelepiped piece of uniform cross-section. Contacts with the sample were made using gold wire and silver paste. The following method was used to remove the uncertainty in the value of resistivity arising due to the error associated with the measurement of distance between the voltage probes. The two current and one voltage probes were fixed to the sample, the room temperature resistance \((R)\) of the sample was then measured as a function of distance by moving the second voltage probe in steps of \(\delta L = 0.05\)mm. The slope of the \(R\) vs \(L + \delta L\) (where \(L\) is the initial distance between the moving and fixed voltage probes; the slope is independent of the choice of \(L\)) is used to normalize the value of resistivity obtained from silver contact measurements. The heat capacity was measured by using the semi-adiabatic, heat pulse method. Magnetization as a function of temperature and field was recorded by using VSM (Oxford Instruments) and SQUID (Quantum Design) magnetometers.

III. RESULTS AND DISCUSSIONS

A. Powder X-ray diffraction patterns

A representative powder x-ray diffraction pattern of \((\text{LaPd}_3)_8\text{Mn}\) obtained using Cu-K\(\alpha\) radiations is shown in Fig. 1. The crystal structure was refined using the Rietveld
refinement program FULLPROF\textsuperscript{15} and an output of such a fitting is also presented in Fig. 1. The Bragg peaks for the powder pattern of (LaPd\textsubscript{3})\textsubscript{8}Mn, generated by the FULLPROF, are shown as two sets of vertical bars in Fig. 1. While the bigger and thicker bars correspond to the reflections one would expect from a face centered cubic LaPd\textsubscript{3} lattice, the smaller bars (labelled as satellite peaks in the figure) correspond to additional low intensity satellite peaks arising due to the periodic occupation of 1b sites by the bigger Mn atoms resulting in the formation of a cubic superstructure of size about twice the size of the LaPd\textsubscript{3} unit cell. The powder diffraction pattern of (CePd\textsubscript{3})\textsubscript{8}Mn, similarly, showed the formation of cubic superstructure. The lattice parameters \(a\) of (LaPd\textsubscript{3})\textsubscript{8}Mn and (CePd\textsubscript{3})\textsubscript{8}Mn are 8.466 and 8.415 Å respectively, which is also the nearest neighbor Mn−Mn separation in the two compounds. The powder diffraction patterns of RPd\textsubscript{3}Mn\(_x\) (\(x = 0.03\) and 0.06 for \(R = \text{La}\) and 0.02 and 0.06 for \(R = \text{Ce}\)) alloys, on the other hand, are similar to those of RPd\textsubscript{3}B\(_x\) alloys: characterized by the absence of satellite peaks and a slight lattice expansion over the parent RPd\textsubscript{3} lattice. We find that the superstructure in (RPd\textsubscript{3})\textsubscript{8}Mn (\(R = \text{La}\) and Ce) is sensitive to the heat treatment. For example, annealing the (CePd\textsubscript{3})\textsubscript{8}Mn samples at 900 °C diminishes the low intensity satellite peaks and gives rise to new peaks, indicating a degradation of the phase by the heat treatment. All the data reported in this work were taken on the as-cast samples.

B. (LaPd\textsubscript{3})\textsubscript{8}Mn

LaPd\textsubscript{3} is diamagnetic at room temperature and shows a nearly temperature independent magnetic susceptibility\textsuperscript{17}. The inverse molar susceptibility, \(\chi^{-1}\), of (LaPd\textsubscript{3})\textsubscript{8}Mn measured in an applied field of 3 kOe is shown in Fig. 2. Between 100 and 300 K the data can be fitted to a modified Curie-Weiss expression: \(\chi = \chi_0 + C/(T - \Theta_p)\) where \(C\) is the Curie constant and \(\chi_0\) is the temperature-independent contribution to the total magnetic susceptibility and \(\Theta_p\) is the paramagnetic Curie temperature. The values of the best-fit parameters are: \(\mu_{\text{eff}} = 5.9 \mu_B/\text{f.u.}\) (derived from \(C\)), \(\Theta_p = 52.1\) K and \(\chi_0 = -0.003\) emu/mol. The \(\mu_{\text{eff}}\) is comparable to the theoretical value of 5.92 \(\mu_B\) for Mn\textsuperscript{2+} (\(S = 5/2\)) ion and suggest that the Mn ions in this compound are in a 3d\textsuperscript{5} electronic configuration. The negative \(\chi_0\) (about 20% of the total susceptibility at 300 K) arises from the the diamagnetic contribution of
the LaPd$_3$ network and is comparable to the susceptibility of (LaPd$_3$)$_8$In which is reported to be $-1.06 \times 10^{-3}$ emu/mol at 293 K. The susceptibility of (LaPd$_3$)$_8$Mn measured in a field of 1 kOe is shown in the inset of Fig. 2. At low temperatures the susceptibility shows a peak at 48 K (T1) indicating an antiferromagnetic transition. In addition there is a smaller peak at 23 K (T2) indicative of a second phase transition.

Fig. 3a shows that the magnetization of (LaPd$_3$)$_8$Mn measured in zero-field-cooled (ZFC) and field-cooled (FC) mode in an applied field of 100 Oe is practically the same. Further, we do not observe any frequency dependence of the AC susceptibility measured at 1, 9 and 99 Hz (Fig. 3b). These data show conclusively that the peaks in the magnetization arise due to the long range magnetic order and not due to any spin-glass-type freezing of the Mn moments. The magnetization, $[M(T)/H]_H$, of (LaPd$_3$)$_8$Mn, measured in several applied fields below 60 K is depicted in Fig. 4. $[M(T)/H]_H$ in the ordered state exhibits very unusual behavior in relatively low applied fields. Noteworthy is the shift of T2-peak to lower temperatures: above $\sim 3$ kOe the T2-peak shifts rapidly to lower temperatures and disappears below 1.8 K in an applied field of $\sim 8.25$ kOe. The magnitude of M/H, on the other hand, increases with the increase in the magnetic field.

The isothermal magnetization $[M(H)]_T$ measured at several temperatures between 2K and 50K is shown in Fig. 5. For each T, $[M(H)]_T$ is measured by cooling the sample from above 60 K to the desired temperature in a nominal zero field. $[M(H)]_{1.8K}$ undergoes a sharp metamagnetic transition in a ‘critical field’ of $\sim 8.25$ kOe, at which the T2-peak of magnetization disappears as seen in Fig. 4. At higher temperatures the metamagnetic transition becomes less sharper and the corresponding field at which the metamagnetic transition occurs also decreases. The linearity of the magnetization at 50 K characterizes the response in the paramagnetic state. $[M(H)]_{2K}$ (shown as inset in Fig. 5 up to 120 kOe) has a tendency towards saturation attaining a value of $\mu_s = 4.6 \mu_B$/Mn in 120 kOe (close to the theoretical value of $5\mu_B$ for Mn$^{2+}$ ion). From the values obtained for $\mu_{eff}$ and $\mu_s$ we conclude that the 3d state of the Mn ions in (LaPd$_3$)$_8$Mn has a localized character in contrast to the usual band like character of Mn 3d state in the intermetallics.

It may be noticed that though $\Theta_p$ obtained from the high temperature Curie-Weiss
behavior of the susceptibility data is positive, indicative of ferromagnetic correlations, the peak in $\chi$ at $T_1$ and the nature of isothermal magnetization suggest antiferromagnetic ordering. The data presented in Fig. 4 and 5 shows that the antiferromagnetic state is not robust. We speculate that the ferromagnetic and antiferromagnetic interactions are competing in this compound. It is worthy of mention here that the Mn sublattice in $(\text{CePd}_3)_8\text{Mn}$ orders ferromagnetically (see below) and our preliminary investigations reveal ferromagnetic ground state in $(\text{LaPd}_{2.75}\text{Rh}_{0.25})_8\text{Mn}$ and $(\text{LaPd}_{2.75}\text{Ag}_{0.25})_8\text{Mn}$. Thus it appears that the nature of magnetic ordering is rather sensitively dependent on slight variations in lattice parameter, conduction electron concentration, etc.

It is interesting to note here that the metamagnetic transitions observed in $(\text{LaPd}_3)_8\text{Mn}$ is qualitatively similar to that observed in bilayer itinerant metamagnet $\text{Sr}_3\text{Ru}_2\text{O}_7$ where the quantum critical fluctuations associated with the metamagnetism at very low temperatures has been shown to give rise to a non-Fermi liquid behavior in magnetotransport\supercite{18,19}. Therefore, it will be very interesting and pertinent to investigate transport and magnetic properties of $(\text{LaPd}_3)_8\text{Mn}$ at very low temperatures and in fields close to the ‘critical field’ $\sim 8.25$ kOe: the field at which the $T_2$-peak in magnetization disappears below 1.8 K or the isothermal magnetization exhibits metamagnetic transition at 1.8 K. We present here our preliminary results on the magnetotransport measurements of $(\text{LaPd}_3)_8\text{Mn}$ down to 1.5 K.

The zero-field resistivity, $\rho(H=0, T)$, data of $(\text{LaPd}_3)_8\text{Mn}$ between 1.5–300 K are shown in the inset of Fig. 6. A rather low residual resistivity of $\sim 7\mu\Omega\text{cm}$ and a moderately good residual resistivity ratio of about 4.5 is observed. At high temperatures $\rho(0, T)$ decreases almost linearly with temperature down to 100 K. The magnetic transition at $T_1$ is clearly reflected by a distinct change in the slope around 48 K below which $\rho(0, T)$ drops precipitously due to the loss of spin-disorder scattering. Around $T_2 = 23$ K the zero-field resistivity is smooth with no discernible anomaly. The magnetoresistivity (MR) measured at few selected fields (Fig. 6) shows interesting features, which correlate nicely with the changes in magnetization in applied fields. The MR, measured in applied fields of 7 and 10 kOe, is positive between the temperatures corresponding to $T_1$ and $T_2$-peaks in the low field magnetization, in accordance with the positive MR typically observed in the antiferromagnetic state. However, at lower temperatures the MR changes sign and exhibits a field
dependent peak, the peak position $T_2$ coinciding with that in the DC magnetization at the same value of the applied field. The low temperature peak in MR vanishes for fields above $\sim 8.25$ kOe within the temperature range of our measurements, thus mimicking the corresponding behavior seen in the DC magnetization. Thus, while the zero-field resistivity is apparently featureless at $T_2 = 23$ K, the in-field resistivity changes distinctly at (field dependent) $T_2$ pointing at the bulk nature of changes taking place at $T_2$.

The in-field resistivity measured at selected fields are plotted in Fig. 7a as a function of $T_2$. The zero-field resistivity and the resistivity in applied fields greater than the ‘critical field’ (namely 8.5, 10 and 15 kOe) follows a $T^2$ variation between 1.5 and 10 K ($T^2$ behavior in zero-field persists up to nearly 15 K). The data in 10 and 15 kOe are nearly coincident with the 8.5 kOe data and are, therefore, not shown in the figure. Evidently, for applied fields less than the ‘critical field’ (namely 5, 7.8 and 8 kOe) a similar quadratic dependence of resistivity is not observed at low temperatures. However, at temperatures above the field dependent $T_2$-peak the data nearly coincides with the data taken in 8.5 kOe and thus follow a $T^2$ behavior. MR at a fixed temperature of 1.6 K was measured up to 60 kOe and is shown in Fig. 7b. MR drops precipitously to $-10\%$ at $\sim 8.25$ kOe , correlating with the metamagnetic transition (Fig. 5). In Sr$_2$Ru$_3$O$_7$ also the metamagnetism is similarly observed in the MR. In higher fields the MR becomes less negative attaining a value of $-3\%$ at 60 kOe. It would be interesting to study the thermal variation of the magnetoresistivity in magnetic field near the ‘critical field’ at very low temperatures for possible non-Fermi liquid behavior.

The heat capacity, $C$, of (LaPd$_3$)$_8$Mn, shown in Fig. 8 increases monotonically with temperature up to 60 K. No discernible anomaly is seen at T1 and T2 where the Mn sub-lattice undergoes transitions. Recalling that the heat capacity at a bulk magnetic transition for spin $S = 1/2$ shows a peak of 25 J/mol K in the mean-field approximation, the lack of anomaly at first appears puzzling. However, it should be noticed that in (LaPd$_3$)$_8$Mn the Mn ions are just about 3 molar percent. Therefore, the total heat capacity at high temperatures is predominantly due to the phonons. The background lattice heat capacity masks the anomaly expected at the magnetic transitions near T1 and T2. The heat capacity of iso-structural non-magnetic analogue (LaPd$_3$)$_8$Ga is also shown in Fig. 8. Assuming that the conduction electron density of states and the phonon contributions in
(LaPd$_3$)$_8$Mn are exactly the same as in iso-structural (LaPd$_3$)$_8$Ga, we have extracted the excess magnetic contribution, $C_{mag}$, by subtracting the two heat capacities, which is shown in the upper inset of Fig. 8. The relatively poor quality of $C_{mag}$ is easy to understand. Typically, our heat capacity set up has accuracy to within 3–4%. Subtracting the heat capacities of the two compounds will give large statistical errors ($\sim 10–20$ J/mol K) at high temperatures ($C = 250$ J/mol K at $\sim 50$ K). A peak in $C_{mag}$ centered at 48 K ($= T_1$) is indeed seen though no apparent anomaly is observed at $T_2$ as in the zero-field resistivity. In the low temperature region ($3 < T < 6$ K), $C/T$ vs. $T^2$ of (LaPd$_3$)$_8$Mn shows linear behavior (see lower inset of Fig. 8) with least square fitted values of $\gamma = 57.8 \pm 1.6$ mJ/mol K$^2$ and $\beta = 6.75 \pm 0.1$ mJ/mol K$^4$, where $\gamma$ and $\beta$ have their usual meaning. For antiferromagnets the spin wave contribution varies as $T^3$ and should not therefore affect the value of $\gamma$. For the non-magnetic analogue (LaPd$_3$)$_8$Ga $\gamma = 2.3 \pm 2.1$ mJ/ mol K$^2$ which is comparable to 2.21 mJ/mol K$^2$ in (LaPd$_3$)$_8$In reported by Cho et al. The data between 3 and 10 K were least square fitted by including additional $T^4$ and $T^6$ terms in the expression for $C/T$. The values of $\gamma$ thus obtained are 61.2 ± 5.3 and 5.4 ± 3.3 mJ/mol K$^2$ for Mn and Ga compounds respectively. Within the limits of error the two sets of values are comparable for each compound. The value of $\gamma$ for (LaPd$_3$)$_8$Mn is an order of magnitude larger than the corresponding value for (LaPd$_3$)$_8$Ga and shows that there is an enhancement in the electronic density of states at the Fermi level in the Mn compound.

C. LaPd$_3$Mnx ($x = 0.03, 0.06$)

It was inferred above from the powder x-ray diffraction pattern of that the Mn ions in LaPd$_3$Mnx ($x = 0.03$ and 0.06) are randomly distributed over the 1b sites in LaPd$_3$ lattice. A random distribution of Mn-ions coupled via RKKY exchange will give rise to a spin-glass type of freezing of the Mn moments as observed, for example, in the canonical spin-glasses like Cu–Mn, Au–Fe, etc. Indeed, our data show convincingly the spin glass state for $x = 0.03$ and 0.06 in LaPd$_3$Mnx. Fig. 9a shows the susceptibility of LaPd$_3$Mn$_{0.03}$ measured in an applied field of 50 Oe both in ZFC and FC mode. In the ZFC mode the susceptibility exhibits a peak at 2.8 K below which large thermomagnetic irreversibility is observed between ZFC and FC data. These features are typically associated with a
spin-glass freezing of the magnetic moments. This is further corroborated by the frequency
dependent peak position of the ac-susceptibility measured at 1, 9 and 999 Hz as shown
in Fig. 9b. The peak in the AC-χ shifts to higher temperatures with the increase in the
frequency of the AC excitation. The corresponding data for LaPd$_3$Mn$_{0.06}$ are shown in Fig.
10a and 10b. A similar behavior is seen but with a freezing temperature (T$_f$) of 5.5 K. The
ratio $\Delta T_f / T_f [\Delta (\log \omega)]$, a quantitative measure of the frequency-dependent peak shift, for
$x = 0.03$ and 0.06 is 0.010 and 0.0098 respectively, which compares well with the values
for the canonical spin glasses (0.005 and 0.01 for Cu–Mn and Au–Fe, respectively). This
ratio offers a good criterion for distinguishing a canonical spin glass from a spin-glass-like
material, superparamagnets for example, where it is often larger by about an order of
magnitude. The inverse molar susceptibility of LaPd$_3$Mn$_{0.03}$ and LaPd$_3$Mn$_{0.06}$ measured
up to 300 K is shown in Fig. 11. As above, the data are least squares fitted to the modified
Curie-Weiss expression in the range 100 to 300 K. The values of the best-fit parameters
are: $\mu_{\text{eff}} = 5.85 \mu_B$/Mn and 5.70 $\mu_B$/Mn, $\Theta_p = 15.8$ K and 32.3 K and $\chi_0 = -0.0002$
emu/mol and $-0.0002$ emu/mol respectively. $\mu_{\text{eff}}$ in both the alloys is comparable to
the theoretical value of 5.92 $\mu_B$ for Mn$^{2+}$ (S = 5/2) ion. Thus the magnetic moments of
the Mn-ions in these two alloys freeze randomly at T$_f$ due to the random RKKY interaction.

In canonical spin-glasses like Cu–Mn the magnetic contribution to the heat capacity,
C$_{\text{mag}}$, exhibits a broad maximum above T$_f$ and gradually falls off at increasing tempera-
tures. Fig. 12a and 12b shows the heat capacity of LaPd$_3$Mn$_x$ ($x = 0.03, 0.06$). The heat capacity of non-magnetic reference LaPd$_3$ is also displayed in the figure. The
insets show C$_{\text{mag}}$ obtained by subtracting the heat capacity of LaPd$_3$ from LaPd$_3$Mn$_x$
alloys. Extraction of C$_{\text{mag}}$ form the total heat capacity is an arduous task particularly
at high temperatures where the errors associated with the absolute values of the total
heat capacity may exceed the magnetic contribution from the Mn-moments. For $x = 0.03$ a broad maximum of height 120 mJ/mol K in C$_{\text{mag}}$ is observed at 4.5 K giving
additional evidence of a spin glass freezing of Mn moments in LaPd$_3$Mn$_{0.03}$. For $x = 0.06$, C$_{\text{mag}}$ has large scatter above 15 K nonetheless indicates a broad peak centered at 16 K.
D. (CePd₃)₈Mn

CePd₃ is an archetypal valence fluctuating compound with enhanced Pauli-paramagnetic ground state \(\chi \sim 10^{-3}\) emu/mol and \(\gamma = 50\) mJ/mol K² as \(T \to 0\) K\(^2\).\(^{21,22,23,24}\) Fig. 13a shows the ZFC and the FC magnetization of (CePd₃)₈Mn in an applied field of 1 kOe. The magnetization increases sharply below 50 K like in a ferromagnetic transition and exhibits a broad peak at 20 K in the ZFC mode. There is a pronounced difference between the ZFC and FC magnetization below the peak temperature. The field dependence of magnetization at 2.8 K, shown in the inset of Fig. 13c, is characterized by both hysteresis and a high coercive field of 6 kOe. Néel has shown that the magnetization \(M(T)\) of a ferromagnet below its Curie temperature, \(T_c\), in applied field \(h\) much smaller than the coercive field \(h_c(T)\) is approximately given by the relation \(M(T) \approx m_s(T) h^2 / h_c^2(T)\), where \(m_s(T)\) is the saturation magnetization.\(^{25}\) Therefore, if below \(T_c\) the anisotropy or the coercive field builds up at a rate much faster than \(m_s(T)\) then \(M(T)\) should show a peak at some temperature. We believe the broad peak at 20 K in ZFC data arises due to the appreciable coercivity/anisotropy in this ferromagnetic material. The FC data in such cases typically show a ferromagnetic saturation below the peak but we find that in (CePd₃)₈Mn the FC magnetization decreases by about 5% below 12 K; the reasons for this are not clear to us. The susceptibility of (CePd₃)₈Mn measured in an applied field of 5 kOe is shown in Fig. 13b. The data above 75 K can be satisfactorily fitted to the modified Curie-Weiss law. The best-fit parameters obtained from such a fitting are: \(\chi_0 = 0.002\) emu/f.u., \(\mu_{\text{eff}}^{\text{observed}} = 7.15\) \(\mu_B/\text{f.u.}\) and \(\theta_p = +38.7\) K. The positive value of \(\theta_p\) indicates ferromagnetic interactions. Indeed the Mn sublattice in (CePd₃)₈Mn orders ferromagnetically around 32 K (refer to Arott plot analysis and heat capacity section below). To deduce the effective magnetic moment per Ce ion it is reasonable to assume that the valence of Mn in (CePd₃)₈Mn is same as observed in the isomorphous (LaPd₃)₈Mn. We further assume that in the paramagnetic regime the contribution of Ce and Mn to the observed effective magnetic moment is simply additive and, therefore, it can be expressed as: \(\mu_{\text{eff}}^{\text{observed}} = \sqrt{8(\mu_{\text{eff}}^{\text{Ce}})^2 + (\mu_{\text{eff}}^{\text{Mn}})^2}\), where \(\mu_{\text{eff}}^{\text{Ce}}\) and \(\mu_{\text{eff}}^{\text{Mn}}\) are effective magnetic moments per Ce and Mn ions respectively in (CePd₃)₈Mn. Since in (LaPd₃)₈Mn the effective magnetic moment per Mn ion is 5.9\(\mu_B\), using this relation we get \(\mu_{\text{eff}}^{\text{Ce}} = 2.42\mu_B\) per Ce ion, which is close to the theoretical value of 2.54 \(\mu_B\) for a free Ce\(^{3+}\) ion. Indicating that the valence of Ce in (CePd₃)₈Mn is close to +3.
Fig. 14a shows $[M(H)]_T$ at various temperatures. In Fig. 14b we have replotted the data as $H/M$ vs. $M^2$ at $T = 20, 30, 40$ and $45$ K. Such plots, known as Arrott plots, are useful in determining the Curie temperature of a ferromagnet. For a ferromagnet the Arrott plot at $T_c$ should pass through the origin while the plots for $T < T_C$ will cut the horizontal axis at a non-zero value, which is the square of the spontaneous magnetization of the ferromagnet ($M_s$). The plots for $T > T_C$ do not intercept the horizontal axis, since the spontaneous magnetization is zero. Following the standard procedure (see for example Ref. 26), the high-field (higher than 5 kOe) data for each temperature is fitted to a fourth order polynomial in $M^2$ which is extrapolated to determine $M_s^2$. We find $M_s$ at 40 K is zero while it has a non-zero value at 30 K, indicating that the ferromagnetic ordering temperature of $(\text{CePd}_3)_8\text{Mn}$ lies between these two temperatures. Isothermal magnetization data at few more temperatures between 30 and 40 K are required to locate $T_C$ exactly. The magnetization at 2.8 K does not saturate even in 120 kOe and its field variation suggests a component varying almost linearly with the field. We believe that the linear component arises from the contribution due to the Ce ions which are in the paramagnetic state down to, at least, 1.5 K (see below, heat capacity and resistivity data). We thus attribute the ferromagnetic transition in $(\text{CePd}_3)_8\text{Mn}$ to the Mn sublattice. The magnetization at 2.8 K in 120 kOe is $6.4 \mu_B$/f.u. and exceeds the corresponding value in $(\text{LaPd}_3)_8\text{Mn}$ substantially due to the (paramagnetic) contribution from the Ce sub-lattice.

The resistivity ($\rho$) of $(\text{CePd}_3)_8\text{Mn}$ (Fig. 15) is about four times larger than that of $(\text{LaPd}_3)_8\text{Mn}$ at 300 K and its thermal variation is qualitatively similar to that seen in the dense Kondo lattice compounds. The relatively higher $\rho$ in $(\text{CePd}_3)_8\text{Mn}$ arises from the incoherent Kondo spin disorder scattering. The peak at 16 K is not due to any phase transition as both the heat capacity (see below) and magnetization data do not show any anomaly at that temperature, but it is due to the transition from the incoherent to coherent Kondo scattering regime as seen, for example, in archetypal Kondo lattice compounds CeAl$_3$, CeCu$_6$ and also in Kondo lattice antiferromagnet $(\text{CePd}_3)_8\text{M}$ (M = Ga, Ge and Al). The resistivity does not apparently exhibit any anomaly at $T_C \sim 35$ K because its large negative temperature coefficient masks the decrease in resistivity occurring due to the ordering of Mn ions (3 mol-percent). It is interesting to recall that the
resistivity of CePd$_3$ also shows a negative temperature coefficient and peaks around 130 K.$^{22}$ Qualitatively, the peak temperature varies inversely with the characteristic Kondo/spin fluctuation temperature. Therefore, there is a significant reduction of the Kondo temperature in (CePd$_3$)$_8$Mn. However, compared to Kondo-lattices (CePd$_3$)$_8$M (M = Ga, Ge, In, Sn, Sb Bi and Al)$^{7,9,10,11}$ which all order antiferromagnetically between 1.5 and 8 K, the RKKY interaction is not strong enough to induce magnetic ordering in (CePd$_3$)$_8$Mn.

The heat capacity of (CePd$_3$)$_8$Mn though comparable to that of the La analogue is however larger in the entire temperature range of 1.5 K to 60 K. This is understandable as the predominant (at higher temperatures) lattice contribution will be nearly similar in the two compounds (the atomic masses of La and Ce differ by less than 1 %) but there will be an additional 4f-derived contribution, $C_{4f}$ due to the Ce-ions in (CePd$_3$)$_8$Mn. The magnetic contribution to the heat capacity, $C_{mag}$ (which is predominantly $C_{4f}$ considering that the Mn concentration is low), was derived by subtracting the heat capacity of (LaPd$_3$)$_8$Ga from (CePd$_3$)$_8$Mn. $C_{mag}$ displayed in Fig. 16a exhibit a peak at $\sim 32$ K which we attribute to the ferromagnetic ordering of the Mn sub-lattice. The peak position of $C_{mag}$ is in agreement with the estimate of the Curie-temperature derived from Arrott plots. Inset of Fig. 16 shows $C_{mag}/T$ vs. $T^2$ below 5 K. A linear extrapolation to $T = 0$ K gives an intercept of 2.2 J/mol K$^2$ (275 mJ/Ce-mol K$^2$). In the case of Ce compounds the screening of Ce magnetic moments by the Kondo interaction results in an enhanced coefficient of the electronic specific heat $\gamma$. The large value of $C_{mag}/T$ at $T = 0$ K shows that (CePd$_3$)$_8$Mn is a dense Kondo lattice. We may recall that $\gamma$ of CePd$_3$ is $\sim 50$ mJ/mol K$^2$. The hybridization of Ce 4f-states with the conduction electrons is reduced in the ternary compound thereby leading to a narrow band density of states and an enhanced effective mass. There is no signature of the magnetic ordering of Ce ions in (CePd$_3$)$_8$Mn unlike (CePd$_3$)$_8$M (M = Ga, Ge, In, Sn, Sb Bi and Al)$^{7,8,12}$ where the heat capacity exhibits huge peaks between 1.5–10 K.

E. CePd$_3$Mnx (x = 0.02, 0.06)

The ZFC and FC magnetization data of CePd$_3$Mn$_{0.06}$ in 100 Oe and the AC susceptibility measured at 9, 99 and 999 Hz are displayed in Fig. 17a and 17b respectively. A peak in
the DC magnetization at 3.9 K with a pronounced thermomagnetic irreversibility below the peak temperature and a frequency dependent peak shift in the AC susceptibility confirms the occurrence of a spin-glass freezing in CePd$_3$Mn$_{0.06}$ at 3.9 K. Since there are two magnetic entities (Ce and Mn) the magnetization data do not distinguish which one of these is getting frozen at 3.9 K. To clarify the situation we measured the heat capacity of CePd$_3$Mn$_{0.06}$ down to 1.5 K as shown in the inset of Fig. 18. The heat capacity data are featureless (with the exception of a minor anomaly at 6.2 K due to the presence of cerium oxide in the sample). The magnetic contribution, $C_{\text{mag}}$, is obtained as $C_{\text{mag}}(T) = [C(\text{CePd}_3\text{Mn}_{0.06})(T) - C(\text{LaPd}_3)(T)]$ and is shown in the main figure (Fig. 18). A broad peak in $C_{\text{mag}}$ centered at $T \sim 2T_f$, with a peak height of $\sim 1$ J/mol K corroborates the spin glass freezing. A rough estimate of the magnetic entropy released up to 20 K yields $S_{\text{mag}} \sim 1.9$ J/mol K. This value is nearly 1/3 of what one would expect from the spin-glass freezing of one mol of Ce ions. These two features: absence of a broad peak in the C and $S_{\text{mag}} \sim 1.9$ J/mol K rule out the spin-glass freezing of the Ce ions at 3.9 K. To conclude, in CePd$_3$Mn$_{0.06}$ the Mn ions freeze in a spin-glass state below 3.9 K, while the Ce ions remain paramagnetic down to the lowest temperature measurements.

The resistivity of CePd$_3$Mn$_{0.02}$ and CePd$_3$Mn$_{0.06}$ is shown in Fig. 15. For $x = 0.02$ the resistivity is qualitatively similar to that of CePd$_3$, the peak having shifted from 130 K in CePd$_3$ to $\sim 100$ K indicating that the addition of Mn at this level does not influence the electronic state of Ce-ions to any considerable extent. But for $x = 0.06$ which further expands the lattice, the resistivity shows a negative $d\rho/dT$ down to the lowest temperature of 1.5 K. For single ion Kondo effect the magnetic part of the resistivity saturates as $T^2$ at low temperatures for $T << T_K$, where $T_K$ is the Kondo temperature. In CePd$_3$Ga$_{0.05}$, for example, the $T^2$ dependence extends up to $T = 20$ K. However our low temperature data do not follow a similar behavior. For example between 1.7 - 4 K a least square fitting of the data to the expression $\rho = \rho_0 - AT^n$ gives $\rho_0 = 344.9$ $\mu$Ω cm, A =0.065$\mu$Ω cm K$^{-2}$ and $n = 1.7$. Absence of $T^2$ behavior in the low temperature resistivity of anomalous f-electron systems has been interpreted to arise from a non-Fermi liquid ground state. Since the low temperature resistivity in CePd$_3$Mn$_{0.06}$ includes the contribution arising from the spin glass freezing of the Mn moments at 3.9 K we cannot infer conclusively a non-Fermi liquid ground state in this alloy.
IV. SUMMARY

We have synthesized and studied the magnetic, thermodynamic and magnetotransport properties of two new interstitial ‘dilute-Mn’ compounds (LaPd₃)₈Mn and (CePd₃)₈Mn. Interstitial alloys CePd₃Mnx (x = 0.02, 0.06) and LaPd₃Mnx (x = 0.03, 0.06) were also studied to bring out the differences in the properties of the compounds and the alloys. The Mn ions form a regular sub-lattice in the compounds and undergo long range magnetic ordering, while they are randomly distributed in the alloys and show spin glass behavior. The peculiarity of the antiferromagnetic state in (LaPd₃)₈Mn is reflected in the occurrence of two peaks (at 48 and 23 K) in its low field magnetization and an unusually strong field-induced shift of the lower peak (T2) towards T = 0 K at relatively low fields. Concomitantly, a metamagnetic transition is seen in the magnetization in the ordered state, which is also clearly reflected in the magnetotransport data. The magnetic ordering due to Mn-sub-lattice is ferromagnetic in (CePd₃)₈Mn, though the Ce sub-lattice shows dense Kondo lattice behavior and remains paramagnetic at least down to 1.5 K. It would be interesting to probe the magnetic behavior of (LaPd₃)₈Mn in the vicinity of the metamagnetic field at very low temperatures.

Acknowledgments

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Figure Captions:

Fig. 1. The observed (cross) and fitted (solid line) powder x-ray diffraction pattern of (LaPd$_3$)$_8$Mn. The vertical bars indicate the position of the Bragg peaks.

Fig. 2. The inverse molar susceptibilities $\chi^{-1}$ and $(\chi - \chi_0)^{-1}$ of (LaPd$_3$)$_8$Mn as a function of temperature. $\chi_0$ is estimated by fitting the susceptibility data to the modified Curie-Weiss expression (refer text for details). The line is the best-fit of the data to the modified Curie-Weiss expression.

Fig. 3 The magnetic susceptibility of (LaPd$_3$)$_8$Mn as a function of temperature: (a) DC susceptibility ($\chi$) in zero-field-cooled (ZFC) and field-cooled (FC) modes in an applied field of 100 Oe. (b) AC susceptibility ($\chi'$) at 1, 9 and 99 Hz.

Fig. 4. The magnetization (M/H) of (LaPd$_3$)$_8$Mn as a function of temperature for a number of fixed applied fields. The dotted line (guide to the eyes) trace the locus of the T2-peak which shift towards lower temperatures with increase in the applied magnetic field.

Fig. 5. The magnetization isotherms of (LaPd$_3$)$_8$Mn as a function of applied magnetic field at several temperatures. Inset: data plotted up to 120 kOe at 2K.

Fig. 6. The magnetoresistance (MR) of (LaPd$_3$)$_8$Mn is shown as a function of temperature for several fixed applied fields. The arrows (pointing upward) mark the position of T2-peak which shift towards lower temperatures with increasing applied fields. Inset: zero-field resistivity as a function of temperature up to 300 K.

Fig. 7. (a) The in-field resistivity ($\rho$) of (LaPd$_3$)$_8$Mn as a function of $T^2$ for several fixed applied fields. Lines indicate a quadratic behavior of $\rho$ in T. (b) The magnetoresistance (MR) of (LaPd$_3$)$_8$Mn as a function of applied magnetic field at 1.6 K.

Fig. 8. The heat capacity (C) of (LaPd$_3$)$_8$Mn and its non-magnetic analogue (LaPd$_3$)$_8$Ga shown as a function of temperature. Upper inset: magnetic part ($C_{mag}$) of the heat capacity in (LaPd$_3$)$_8$Mn. Lower inset: $C/T$ plotted against $T^2$ for the two compounds. The lines are linear extrapolation of the data to $T = 0$ K.
Fig. 9. The magnetic susceptibility of LaPd$_3$Mn$_{0.03}$ as a function of temperature: (a) DC susceptibility ($\chi$) in zero-field-cooled (ZFC) and field-cooled (FC) modes in an applied field of 100 Oe. (b) AC susceptibility ($\chi'$) at 1, 9 and 999 Hz. The continuous line is guide to the eyes.

Fig. 10. The magnetic susceptibility of LaPd$_3$Mn$_{0.06}$ plotted as a function of temperature: (a) DC susceptibility ($\chi$) in zero-field-cooled (ZFC) and field-cooled (FC) modes in an applied field of 50 Oe. (b) AC susceptibility ($\chi'$) at 1, 9, 99 and 999 Hz. The solid lines are guide to the eyes.

Fig. 11. The inverse molar susceptibilities: $\chi^{-1}$ and $(\chi - \chi_0)^{-1}$ of LaPd$_3$Mn$_{0.03}$ and LaPd$_3$Mn$_{0.06}$ as a function of temperature. $\chi_0$ is estimated by fitting the susceptibility data to the modified Curie-Weiss expression (refer text for details). The solid lines are the best-fit of the data to the modified Curie-Weiss expression.

Fig. 12. (a) The heat capacity (C) of LaPd$_3$Mn$_{0.03}$ plotted as a function of temperature. Inset: The magnetic part in the heat capacity of LaPd$_3$Mn$_{0.03}$ ($C_{mag}$). (b) C and $C_{mag}$ (inset) in LaPd$_3$Mn$_{0.06}$. C of the non-magnetic analogue LaPd$_3$ is also shown both a and b.

Fig. 13. (a) The magnetization (M/H) of (CePd$_3$)$_8$Mn in an applied field of 1 kOe as a function of temperature in zero-field-cooled (ZFC) and field-cooled (FC) mode. Inset: an exaggerated view of the low temperature maximum in M/H. (b) The inverse molar susceptibility in an applied field of 5 kOe as a function of temperature. The solid line is best-fit of the data to the modified Curie-Weiss expression (c) The hysteresis loop in the isothermal magnetization (M) of (CePd$_3$)$_8$Mn measured at 2.8 K as a function of applied magnetic field.

Fig. 14. The magnetization isotherms of (CePd$_3$)$_8$Mn at several temperatures between 20 and 75 K are shown as a function of temperature. (b) Arrot plots at 20, 30, 40 and 45 K in (CePd$_3$)$_8$Mn. The continuous lines are polynomial (in $M^2$) fits to the high field data (please see text for details).

Fig. 15. The resistivity of (CePd$_3$)$_8$Mn and CePd$_3$Mn$_x$ ($x = 0.02, 0.06$) as a function of temperature. Inset: an enlarged view of the same in low temperatures.
Fig. 16. (a) The magnetic part (C_{mag}) in the heat capacity of (CePd_3)_8Mn is shown as a function of temperature between 15 and 45 K. Inset: heat capacity (C) of (CePd_3)_8Mn is shown between 1.5K and 50 K. (b) C/T against T^2 at low temperatures. The line is linear extrapolation of the data to T = 0 K.

Fig. 17. The magnetic susceptibility of CePd_3Mn_{0.06} as a function of temperature: (a) DC susceptibility (χ) in zero-field-cooled (ZFC) and field-cooled (FC) modes in an applied field of 100 Oe. (b) AC susceptibility (χ') at 9, 99 and 999 Hz. The solid line is guide to the eyes.

Fig. 18. The magnetic part, C_{mag}, (left axis) and the total heat capacity, C, (right axis) of CePd_3Mn_{0.03} plotted as a function of temperature.
(LaPd$_3)_8$Mn

Fig. 2 surjeet et al.
(LaPd$_3$)$_8$Mn

(a)

(b)

$\chi$ (emu/mol)

$\chi'$ (arb. units)

H = 100 Oe

T (K)

T1

T2

amplitude = 2 Oe

0 10 20 30 40 50 60

0 0.1 0.2 0.3 0.4 0.5

fig.3 surjeet et al.
This figure "Graph4.PNG" is available in "PNG" format from:

http://arxiv.org/ps/cond-mat/0304038v1
This figure "Graph5.PNG" is available in "PNG" format from:

http://arxiv.org/ps/cond-mat/0304038v1
fig. 6 surjeet et al.
fig. 7 surjeet et al.
\( C \text{ (J/mol K)} \)

\( T \text{ (K)} \)

\( (\text{LaPd}_3)_8 \text{Mn} \)

\( (\text{LaPd}_3)_8 \text{Ga} \)

fig.8. surjeet et al.
LaPd$_3$Mn$_{0.03}$

**Figure 9.** Surjeet et al.
fig. 10 surjeet et al.
LaPd₃Mn₀.₀₃

\begin{align*}
\chi^{-1} & \quad (\chi-\chi_0)^{-1} \\
\chi^{-1} & \quad (\chi-\chi_0)^{-1}
\end{align*}

LaPd₃Mn₀.₀₆

\begin{align*}
\chi^{-1} & \quad (\chi-\chi_0)^{-1} \\
\chi^{-1} & \quad (\chi-\chi_0)^{-1}
\end{align*}

fig.11. surjeet et al.
LaPd$_3$Mn$_{0.03}$  LaPd$_3$

LaPd$_3$Mn$_{0.06}$  LaPd$_3$

fig. 12 surjeet et al.
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This figure "Graph14.PNG" is available in "PNG" format from:

http://arxiv.org/ps/cond-mat/0304038v1
(LaPd$_3$)$_8$Mn

Satellite Peaks

Intensity (arb. units)

fig. 1 surjeet et al.
\[ \rho(T) \] (\( \mu \Omega \) cm) vs. \( T(K) \)

- \( \Delta \): \((\text{CePd}_3)_8\text{Mn}\)
- \( \Box \): \(\text{CePd}_3\text{Mn}_{0.06}\)
- \( \Diamond \): \(\text{CePd}_3\text{Mn}_{0.03}\)

fig. 15 surjeet et al.
\[(\text{CePd}_3)_8\text{Mn}\]

- Fig. 16: Surjeet et al.

\[C_{\text{mag}} \text{ (J/mol K)}\]

\[\gamma (\text{CePd}_3)_8\text{Mn}\]

\[\frac{C}{T} \text{ (J/mol-K)}\]

\[T (\text{K})\]

\[T^2 (\text{K}^2)\]
fig. 17 surjeet et al.
fig. 18 surjeet et al.