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Pressure dependent Magnetic, AC Susceptibility and Electrical Properties of Nd$_7$Pd$_3$

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Abstract

Effects of pressure on magnetization and isothermal entropy change, AC susceptibility, and resistivity for Nd$_7$Pd$_3$ have been studied. Nd$_7$Pd$_3$ shows three magnetic transitions $T_t$ (=15 K), $T_C$ (=34 K), and $T_N$ (=38 K) at a pressure of zero bar. $T_C$ and $T_N$ are very sensitive with pressure and rate of change of $T_C$ and $T_N$ with respect to pressure, i.e., $dT_C/dp$ and $dT_N/dp$ are -1.906 K/kbar and -0.313 K/kbar respectively, whereas $T_t$ is insensitive to pressure change. It has also been observed that isothermal entropy change is also sensitive with pressure. Below 15 K, an indication of spin glass nature in ac susceptibility data is seen. Time dependent magnetization at low temperature also supports the spin glass nature but at high temperature it indicates the complex nature. Electrical resistivity data has been fitted to relation $\rho=\rho_0+AT^2$ in temperature range 12-30 K and $\rho=\rho_0+BT-CT^3$ in temperature range 40-220 K. M-H isotherm confirms the AFM nature of Nd$_7$Pd$_3$, confirmed further by Arrott’s plot.
Introduction

The rare earth intermetallic compounds R₇M₃ (where R = rare earth metal and M = transition metal) have hexagonal Th₇Fe₃-type structure (space group: P6₃mc) [1]. R in this structure occupies three non-equivalent sites (6c, 12d and 2b) whereas M resides at the site 12b [2]. The compounds of this series have multiple magnetic transitions except Gd₇M₃ [5]. As an example, Nd₇Rh₃ has two magnetic phase transitions at 34K and 9.1K [3], Sm₇Rh₃ has two transitions at 54 K and 25 K [4] and Gd₇Rh₃ behaves as an antiferromagnet below 150 K. In the same way, Nd₇Pd₃ behaves as an antiferromagnet to paramagnet at around 38 K [6], and a ferromagnet to antiferromagnet at a temperature of 34 K [6, 7].

Unlike, Nd₇Rh₃ which has the property to co-exist in antiferromagnetic and ‘super-cooled’ ferromagnetic phases after the reduction of magnetic field to zero, and still does not show spin-glass anomalies [8], Nd₇Pd₃ shows spin-glass nature at low temperature but does not co-exist in antiferromagnetic and ‘super-cooled’ nature [3]. In contrast to Gd₇Pd₃, which has spin dependent momentum density [9], Nd₇Pd₃ does not show this property, but Nd₇Pd₃ also has an interesting property that the FM→AFM (FM stands for ferromagnetism, AFM stands for antiferromagnetism), and AFM→PM (PM stands for paramagnetism) transition property of Nd₇Pd₃ is sensitive to pressure. Nd₇Pd₃ shows strong pressure effect as that is found in Ce₇Ni₃, where there is a transition from a magnetic state to a non-magnetic state takes place at low temperatures by applying weak pressure [6, 7]. Rare earth based intermetallic compounds (R₇M₃) have applications in magnetostriction, magnetoresistance and magnetocaloric effect [10, 11]. Magnetocaloric effect (MCE), which manifests as the heating or cooling of magnetic materials due to a varying magnetic field, has attracted considerable attention. The origin of MCE in many materials has been explained and its practical use to achieve low temperatures is being anticipated. Giant magnetocaloric effect exhibited by many rare earth (R)-transition metal (M) intermetallic compounds render them as potential refrigerants for magnetic refrigerators [10-11]. Large value of MCE spreads over a wide temperature range is considered as one of the most important requirements of a practical magnetic refrigerant system [10-11].

In this present work, we calculated the pressure effect on temperature dependent magnetization, and also on isothermal entropy change. It was shown by Sengupta et. al., [10] that in Nd₇Pd₃ there is an occurrence of kinetic hindrance, which leads to phase coexistence (supercooled ferromagnetic antiferromagnetic). They showed from the AC susceptibility data, that such a coexisting phase is different from spin. Nd₇Pd₃ shows spin glass so we have performed AC susceptibility and temperature dependent normalized magnetization data to confirm the spin glass nature in our sample. In the present work, we have also calculated the M-H isotherms and Arrott’s
plot to study the magnetic structure of Nd₇Pd₃. Electrical resistivity measurements were also performed to study the metallic nature of this compound.

**Experimental Details**

The sample was prepared in an argon atmosphere using arc melting furnace. The starting materials were 99.8% purity. The ingots were checked by X-ray diffraction after annealing at 873K for a week and found to be single phase (using the fullProf software) of Fe₇Th₃-type. The magnetization has been measured on different applied pressures (P) using a Cu–Be clamp type cell, which can work up to 12 kbar, attached to a SQUID magnetometer whereas ac susceptibility and transport in PPMS in the temperature range of 2–300 K. For the measurement of magnetization in a zero-field-cooled (ZFC) state, the sample was cooled from the paramagnetic state in a zero applied field, and the magnetization was measured while warming the sample. MCE has been calculated using M-H isotherms near transition temperature. Before measurement at each temperature the specimen was zero-field cooled from 60 K.

**Results and Discussion**

Figure 1 shows the room temperature powder X-ray diffraction pattern and Rietveld refinement of Nd₇Pd₃. All peaks can be indexed as the hexagonal Fe₇Th₃-type cell in the space group=P6₃mc (a=10.1369(8) Å, b=10.1369(8) Å, c=6.38354(6) Å and γ=120°). The sample is found to contain a small (2-3%) impurity of Nd₃Pd₂. The low values of R_p and R_B show that the pattern fitting is accurate [12] (shown in figure 1). The Bragg peaks of Nd₇Pd₃ (green line) and Nd₃Pd₂ (violet line) have also been shown in figure 1.
Figure 1: The observed (lines) and calculated (open circles) powder diffraction pattern of Nd₇Pd₃ after the completion of Rietveld refinement. The calculated positions of the Bragg peaks are shown as vertical bars [(Nd₇Pd₃ (green line) and Nd₃Pd₂ (violet line)] just below the plots of the observed and calculated intensities. The differences between the observed and calculated intensities are shown at the bottom of the plot.

Figure 2 shows the temperature dependence of magnetization (M-T) in various applied pressure under field-cooled (FC), and zero-field-cooled (ZFC) modes, at 500 Oe. Figure 2(a) shows the M-T plot for FC condition. dM/dT v/s T plot clearly indicates three magnetic transitions at 15 K(=Tᵣ), 34 K(=T_C) and 38 K(=T_N) as reported by Hideoki et al. [6]. While cooling down the sample, we get our first transition, around 38 K (at 0 bar) which is a paramagnetic (PM) → antiferomagnetic (AFM) transition [6] and next transition at around T_C = 34 K (at 0 bar) is AFM to ferromagnetic (FM). Below this at 34 K, Nd₇Pd₃ shows ferromagnetism, and around Tᵣ of 15 K Nd₇Pd₃ shows another transition which is referred to as a glassy nature [6] which is due to magnetic frustration. As it is visible from figure 2(a), that as pressure is increased from 0 bar to 5 kbar (keeping H constant of 500 Oe), T_C decreases from 34 K to 15 K, and at high pressure of 9 kbar, T_N is dominating over T_C and Tᵣ. So we can say that at 9 kbar, there is only PM → AFM transition, and there is no FM state [6]. Figure 2(a) clearly reveals that, as pressure is increased from 0 bar to 9 kbar, T_C and T_N shift towards a lower value, this is shown graphically by figures 2(c) and 2(d), that with increasing pressure, T_C and T_N decrease by the rate of dT_C/dp = -1.906 K/kbar and dT_N/dp = -0.313 K/kbar [6]. We have not plotted dTᵣ/dp as Tᵣ is insensitive w.r.t pressure. Figure 2(b) shows the difference between ZFC and FC mode and this difference is decreasing with increasing pressure. We also observed with increasing pressure, difference is reduced, the reason being at high pressure T_N dominates over T_C and Tᵣ.
A question arises, whether the magnetic state attained after reversing the field to path zero is purely ferromagnetic in character. Figure 3(a) shows the isothermal magnetization behaviour of Nd$_7$Pd$_3$ at 28 K, 36 K, 38 K, 40 K, and 50 K. It can be noted from figure 3(a) that, at low field, there is some magnetization in Nd$_7$Pd$_3$, and this magnetization attains a saturation moment of 1.8 $\mu_B$/Nd$^{3+}$[7]. This property of attaining maximum saturation at low magnetizing field indicates the ferromagnetic nature of Nd$_7$Pd$_3$ which is in agreement with figure 2(a). At high temperatures, there is a linear dependence of M on H, which accounts for the paramagnetic behaviour of Nd$_7$Pd$_3$ nature after the Neel temperature. Figure 3(a) clearly reveals that as we go from 36 K to 38 K and then to 40 K, we are getting an off-axis M-H plots, and off-axis plot is a characteristic of antiferromagnetic materials, so we concluded that Nd$_7$Pd$_3$ is moving towards antiferromagnetic behaviour from ferromagnetic behaviour. To know the magnetic phase transition, we plot magnetization isotherms in the form of Arrott plot.

Figure 3: (a) M vs. H isotherms and (b) Arrott plots of Nd$_7$Pd$_3$ compound near ordering temperature. Before measurements at each temperature the specimen was zero-field cooled from 60 K.
Figure 3(b) shows the Arrott plots of Nd₇Pd₃. Arrott plots follow Landau expansion [13], where the mean free energy is given by

\[ F(M) = \frac{aM^2}{2} + \frac{bM^4}{4} + \ldots - \mu_0 MH \ldots \ldots (1) \]

Here, a and b are temperature dependent coefficients. However, Arrott plots can be used to estimate T_N in antiferromagnetic materials, and T_C in FM materials [14, 15]. In the case of ferromagnetic and paramagnetic substances, b is positive, while a is positive for paramagnetic and negative for ferromagnetic materials. Banerjee [16] suggested that all Arrott plots with positive slope give second order magnetic phase transition while negative slope gives first order magnetic phase transition. A close view of figure 3(b) shows negative slope at low temperatures which indicates the first order magnetic phase transition at low temperature. We can see from this Arrott plot that as temperature is increased, we are getting antiferromagnetic behaviour. Also, at low temperatures we found that, Arrott plots are displaced to the left, which indicates the ferromagnetic behaviour. An S-shaped Arrott plots indicates FOT, which is depicted from 3(b).

Figure 4: Temperature variation of magnetization on different field at (a) 0 bar, (b) 2 kbar, (c) 5 kbar and (d) 9 kbar.

Figure 4 shows the temperature variation of magnetization at different fields from 2 KOe to 50 KOe, with a gap of 2 KOe between every two successive fields, in ZFC mode. In figure 4(a) it has been found that at a pressure of 0 bar, we get a kink at around 15K, and at 34K, this matches with the date shown in figure 2(a) at 0 bar. Then as similar to figure 2(a), we get a downfall of magnetization.
at around 38 K (=T_N). Figure 4(a) also reveals that as magnetic field is increased, T_N is increased. This same pattern is shown in figure 4(b), 4(c) and 4(d), and as pressure is increased, T_N is decreased.

Figure 4(b) shows temperature variation of magnetization on the same fields from 2 kOe to 50 kOe, but with a pressure of 2 kbar. So we can see here again T_i, T_C and T_N of 12 K, 28 K and 32 K, this data also coincides with magnetization v/s temperature data of figure 2(a). Similar are the results for figure 4(c) and 4(d). We also note a pattern here that as pressure increases from 0 bar to 9 Kbar, the magnetization decreases in value. At 9 Kbar, the FM state is again absent as can be seen from figure 4(d). Figure 4(d) also shows a huge broadening of the curves with increasing magnetic field, this is due to crystal anisotropy of Nd_7Pd_3.

Figure 4(b) shows temperature variation of magnetization on the same fields from 2 kOe to 50 kOe, but with a pressure of 2 kbar. So we can see here again T_i, T_C and T_N of 12 K, 28 K and 32 K, this data also coincides with magnetization v/s temperature data of figure 2(a). Similar are the results for figure 4(c) and 4(d). We also note a pattern here that as pressure increases from 0 bar to 9 Kbar, the magnetization decreases in value. At 9 Kbar, the FM state is again absent as can be seen from figure 4(d). Figure 4(d) also shows a huge broadening of the curves with increasing magnetic field, this is due to crystal anisotropy of Nd_7Pd_3.

Based on the magnetization isotherms measured near Curie temperature, the change in entropy of each sample near their ordering temperature was calculated using the integrated Maxwell relation [10-11]

\[
\Delta S_M(T_{i+1}, H) = \int_{T_i}^{T_{i+1}} \left( \frac{\partial M}{\partial T} \right)_H dH \approx \frac{1}{\Delta T} \left[ M(T_{i+1}, H_f) - M(T_i, H_f) \right] dH
\]

(2)

Where \( T_{\text{av}} = \frac{(T_{i+1} + T_i)}{2} \) means average temperature and \( \Delta T = T_{i+1} - T_i \) means temperature difference between two magnetization isotherms measured at \( T_{i+1} \) and \( T_i \) with the magnetic field \( H_i \) to \( H_f \). Figure 5 shows the MCE variation in Nd_7Pd_3 compound under various pressures. It can be seen from the figure that, with increase in pressure, the peak in the \( \Delta S_M \) vs. T plot moves towards low temperatures. We note that Nd_7Pd_3 exhibits reversible magnetocaloric effect. In all the four graphs we can easily see
that the entropy change is negative (positive MCE). The maximum values of $\Delta S_M$ ($\Delta S_{M_{\text{Max}}}$) are 3.8 J/kg K, 3.5 J/kg K, 3.1 J/kg K and 1.4 J/kg K at pressures of 0 bar, 2 Kbar, 5 Kbar and 9 Kbar respectively. At 0 bar, $\Delta S_{M_{\text{Max}}}$ is at $T_N$. Also, as the pressure is increased from 0 bar to 9 Kbar, the temperature at which we get $\Delta S_{M_{\text{Max}}}$ gets reduced from 38 K to 28 K. A common pattern in all the four graphs of figure 5 is that, the magnitude of $\Delta S_M$ increases, as the value of applied field is increased from 10 KOe to 50 KOe, and as the pressure is increased from 0 bar to 9 Kbar, we get $\Delta S_{M_{\text{Max}}}$ at a lower temperature. To understand the magnetic structure, more studies like neutron diffraction, are required.

The AC magnetic susceptibility is magnetic susceptibility which we get by the application of AC magnetic field. AC susceptibility is written as

$$\chi_{ac} = \chi' - i\chi'' \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots
Figure 6: Temperature variation of the (a) real and (b) imaginary component of the ac magnetic susceptibility of Nd$_7$Pd$_3$ in a 50e AC magnetic field at various frequencies.

The glassy nature as stated in figure 6(b) is analyzed using empirical Vogel–Fulcher (VF) power law and is shown in figure 7, which shows the variation of spin freezing temperature $T_f$ with relaxation time $T_0$. The AC susceptibility fits well with power law

$$T_f = T_0 + [(E_a / \kappa_B) / 100] \times \frac{1}{v}$$  \hspace{1cm} (4)

where, $T_0$ is given by 1.54K, $E_a$ is activation energy, $\kappa_B$ is Boltzman constant, and $E_a / \kappa_B = 14.95K$. As can be seen from figure 7, $T_f$ increase linearly with an increase with the logarithmic plot of $\omega_0/\omega$, where $\omega_0$ is the attempt frequency ($\omega_0 = \frac{1}{T_0}$) and $\omega$ is the operating frequency.

Figure 7: Frequency dependent temperature data fitted using the empirical Vogel-Fulcher law below $T_f$. 
Figure 8, shows normalized magnetization, M as a function of time at various temperatures in ZFC mode. \( M_0 \) is the magnetization at \( t = 0 \). All the measured values of M are normalized w.r.t \( M_0 \).

Figure 8(b) shows, normalized magnetization with time as varying function, at 3 K, and it has been found that at 3 K, the magnetization is explained using combination of two laws named as power law and stretched exponential law, given by

\[
\frac{M(t)}{M_0} = P_1 \times (1 - 2 \times t^\gamma) + P_2 \times \exp\left(\frac{-t}{\tau}\right) \quad (5)
\]

here, \( P_1 \) and \( P_2 \) are temperature dependent weight factors for both the power law and stretched exponential respectively. The stretched law is a characteristic associated with spin glass nature, so stretched law in equation 5 indicates the glassy nature of Nd\(_7\)Pd\(_3\) at low temperature. But as there is also a contribution of power law in equation 5, which signifies that glassy nature has not completely formed.

Figure 8(c) shows that at 25K, normalized magnetization, M, is very well fitted with the power law

\[
\frac{M}{M(0)} = -1 + 2t^\gamma \quad \ldots (6)
\]

In figure 8(c) the red solid line is fit to the data using power law [18]. As at 25K, normalized magnetization obeys power law, which tells about the metastability across the FOT, similar to solidification of melt when melt is undercooled. We are not able to fit in equation (6) or (7), the other temperatures. It is a clear indication of complex magnetic structure at high temperature.
Figure 8: (a) Normalized magnetization vs time (t) plot for Nd$_7$Pd$_3$ compound measured at different temperature an applied field of 10 kOe in the virgin H cycle. For each H, $M(0)$ is the value of the magnetization recorded when relaxation measurement were started. (b) At 3K data fitted with equation $M / M(0) = P_1(1 + 2t^2) + P_2 \exp(-t/\tau)$ (c) At 25 K data fitted with equation $M / M(0) = -1 + 2t'$. 

To get a better insight into the magnetic state of the compound, measurements on electrical resistivity were carried out. Figure 9 shows the electrical resistivity of Nd$_7$Pd$_3$ as a function of temperature. From 9(a), we found that the residual resistivity of the sample is about 0.01 m$\mu$-cm. Nd$_7$Pd$_3$ showing metallic character. There is a change in slope of the resistivity near all the magnetic transition as given in figure 2(a). Figure 9(a) shows three transitions. It can be found that at low temperature (i.e. temperature below $T_C$) resistivity follows the equation $\rho=\rho_0+AT^2$ and at high temperatures (i.e. temperature above $T_N$) resistivity follows $\rho=\rho_0+BT^3$, where $\rho_0$ is residual resistivity [9] given by 8.67E-6$\Omega$-cm at low temperatures and 6.91E-5$\Omega$-cm at high temperature respectively. A, B and C are temperature independent constants [9, 19]. From the electrical resistivity, we can conclude that Nd$_7$Pd$_3$ is metallic in nature over the entire temperature range, and because of the loss of spin disorder scattering due to spontaneous magnetic ordering, there is a drop in resistivity at $T_C=34$ K.

The second term in $\rho=\rho_0+AT^2$ is proportional to $T^2$ and represents the lattice spin waves. For figure 9(b), A=5.8E-8 $\Omega$-cm/K$^2$ and suggests that electron spin-wave scattering is dominant factor that helps to determine electrical resistivity in the temperature range of 12-32 K. Similarly, in high temperature range, $\rho=\rho_0+BT^3$, B=4.58E-7 $\Omega$-cm/K and C=2.37E-12 $\Omega$-cm/K$^2$ indicates the phonon contribution and also indicates that s-d scattering [9] is important in high temperature range i.e. above 40 K. Figure 9(b) and 9(c), are plotted, to determine the dominant contributions in the electrical resistivity data [20].
Figure 9: (a) Temperature dependent of electrical resistivity of Nd7Pd3 compound. Electrical resistivity data fitted to relation (b) $\rho=\rho_0+AT^2$ in temperature range 12-30 K (b) $\rho=\rho_0+BT-CT^3$ in temperature range 40-220 K.

Conclusions

Electrical resistivity, AC susceptibility, MCE and magnetization measurements were performed on Nd7Pd3 showing three magnetic transitions, first at around 15 K, then second at around 34 K and the third is around 38 K where, 34 K and 38 K are the Curie temperature and Neel temperature of Nd7Pd3 respectively. For a field of 10 kOe, it showed a MCE of 3.8 J/kg K, at 0 bar. The calculated values of $T_C$ and $T_N$ match with M-H isotherm, AC susceptibility and temperature dependent electrical resistivity as well. Spin glass nature has also been confirmed by temperature dependent AC susceptibility, normalized time dependent magnetization and Vogel-Fulcher law. The $\chi''$ v/s $T$ plot tells that at $T_C$ there is a strong peak, but at higher temperatures, $\chi''$ is nearly equal to zero, but it has a non-zero value below $T_C$. This is a characteristic of spin glass transition. Electrical resistivity of Nd7Pd3 as a function of temperature tells that Nd7Pd3 has metallic nature over the entire temperature range, and because there is a loss of spin disorder scattering, there is a drop in resistivity at $T_C=34$ K. M-H isotherm and Arrot’s plots show the AFM nature of Nd7Pd3, which is in accordance with M-T...
data in FC mode. M-T data at various pressures and at constant field of 50KOe confirms the magnetic
anisotropy of Nd7Pd3. This data has been verified by MCE plots. Large refrigerating cooling power,
large isothermal entropy change, soft magnetic behaviour and wide operating temperature range make
it an attractive candidate as magnetic refrigerant in low temperature region.

Acknowledgement
One of the authors (Pramod Kumar) thanks DST, Govt. of India for proving financial support for this
work. We would like to thank Dr. Alexandre Carvalho and Prof. Gamma for helping us in high
pressure measurements.

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