Historically, the magnetic compensation behaviour in the admixed rare earth intermetallics was first reported by Williams et al. They had shown that the compensation points exist in Pr$_{1-x}$Gd$_x$Al$_2$ ($x = 0.2-0.3$) and the substitution of up to 20 atomic % of Pr by Gd results in lower magnetization values compared to the pure alloy. We have revisited the Pr$_{1-x}$Gd$_x$Al$_2$ series in the light of the results in recent years in Sm$_{1-x}$Gd$_x$Al$_2$. The Samarium ion, due to the admixture of the higher oxidation states ($3^+$, $2^+$, $1^-$) that the field-induced changes across the magnetic compensation temperature, are unique to special characteristics of Sm$^{3+}$ ions under the influence of CEF and exchange field effects. The sign change in the magneto-resistance across $T_{comp}$ is seen in Pr$_{0.83}$Gd$_{0.2}$Al$_2$, however, it is additionally accompanied by an oscillatory behaviour at lower temperatures. Further interesting result in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ is the identification of the fingerprint of the magnetic reorientation in the in-field ac-susceptibility data. To fortify the field-induced changes across the magnetic compensation temperature, we are also presenting the results in Pr$_{0.83}$Gd$_{0.17}$Al$_2$, where the $T_{comp}$ and $T_c$ are in close proximity to each other.

A series of polycrystalline Pr$_{1-x}$Gd$_x$Al$_2$ ($x = 0, 0.15, 0.17, 0.2$ and $0.25$) alloys were prepared by melting together the stoichiometric amounts of the constituent elements in a tetra arc furnace (Model: TCA 4-5, Techno Search Corp., Japan). The elemental analysis of the admixed alloys using an analyzer (JEOL JSX-3222) reassured the targeted stoichiometries. The x-ray diffraction patterns were recorded for the powdered samples using X’pert PRO x-ray diffractometer. Indexing of the x-ray patterns confirmed the cubic C15 phase in all the alloys. A small piece of Pr$_{0.83}$Gd$_{0.17}$Al$_2$ was annealed at 1000 °C for 10 days to ascertain the differences in the results in the as-grown and the annealed samples. The dc magnetization and the ac susceptibility data were recorded using Quantum Design (QD) Inc. superconducting quantum interference device (SQUID) magnetometer (Model MPMS-5). The heat capacity and the resistivity data was measured in a Physical Property Measurement System (PPMS) of QD Inc. U. S. A. The Hall resistance is measured as a function of temperature using the homemade setup for transport studies.

In Fig. 1, the field cooled cooldown (FCC) magnetization ($M_{FCC}$) curves in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ are shown in $H \sim 1$ Oe and $H = 14$ kOe. In the nominal zero field ($H \sim 1$ Oe) cooled data, the magnetic ordering temperature of this alloy is marked ($T_c \approx 64$ K). The magnetization signal is positive between $T_c$ and $T_{comp}$ ($\approx 38$ K) and...
negative below $T_{comp}$. At high fields, a minimum in the thermomagnetic curve is observed at $T^* \approx 39$ K.

FIG. 1: Field cooled cooldown magnetization ($M_{FCC}$) in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ alloy. The $T_c (\approx 64$ K) and $T_{comp} (\approx 38$ K) are marked in the nominal zero field cooled curve. In high field (14 kOe), the occurrence of magnetic turnaround results in a minimum at $T^* \approx 39$ K.

FIG. 2: (Color online) (a) Specific heat ($C_p/T$ versus $T$) data in $H = 1$ Oe, 20 kOe and 50 kOe and (b) the temperature dependence of the ac-susceptibility in $H = 1$ Oe, 0.5 kOe and 50 kOe in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ alloy. In panel (b), $T^* (\approx 39$ K) in $H = 50$ kOe is marked.

Figure 2 shows a collation of the temperature dependences of (a) the specific heat in $H = 1$ Oe, 20 kOe and 50 kOe and (b) the ac susceptibility responses in $H = 1$ Oe, 500 Oe, 50 kOe in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ alloy. In Fig. 2 (a), the nominal zero field specific heat data shows a rise in the specific heat starting at $T_c (\approx 64$ K), followed by a broad hump. The broad peak closer to $T_c$ gets suppressed as the magnetic field is progressively enhanced, and a relatively sharp field-induced peak develops at $T^*$, whose height scales with the applied magnetic field (all data not shown). In the ac-susceptibility data in the nominal zero field (cf. Fig. 2(b)), a broad peak can be seen at the magnetic transition ($T_c$ is marked at the rising edge of the peak). At high fields, this gets collapsed and an additional peak at lower temperature surfaces up at $T^*$.

FIG. 3: (Color online) The electrical resistivity as a function of temperature in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ alloy in nominal zero field and 50 kOe. An inset in the top left corner shows the Hall resistance ($R_H$) as a function of $T$ in $H = 10$ kOe. Insets, (a) and (b) in bottom right show the normalized magnetoresistance ($\Delta R(H) = (R(H) - R(0))/R(0)$) in (a) 1 kOe and (b) 50 kOe.

Figure 3 shows the electrical resistivity as a function of temperature in Pr$_{0.8}$Gd$_{0.2}$Al$_2$ alloy. The nominal zero field and 50 kOe data are plotted together for comparison. The sharp drop in the nominal zero field electrical resistance data occurs at $T_c (\approx 64$ K). This feature broadens out in 50 kOe and lies below the $R(T)$ curve in the nominal zero field. The inset panels (a) and (b) in Fig. 3 show the magnetoresistance values calculated from the electrical resistance data recorded in 1 kOe and 50 kOe. The normalized magnetoresistance in 1 kOe ($\Delta R_1 = [R(1 \text{ kOe}) - R(0)]/R(0)$) displays negative values below $T_c$ and then crosses over to the positive values at $\approx 40$ K. Below this temperature, the magnetoresistance changes sign two more times exhibiting an oscillatory character. The percent change in magnetoresistance in 1 kOe is within 0.5 %. In 50 kOe, the magnetoresistance retains the oscillatory variation and it is an order of magnitude higher. Inset in the top left corner of Fig. 3 shows the Hall resistance as a function of temperature in $H = 10$ kOe. The sign change in Hall resistance ($R_H$) can be observed at 38 K.

Figure 4(a) shows the $M_{FCC}$ in nominal zero field in Pr$_{0.85}$Gd$_{0.17}$Al$_2$ and Pr$_{0.75}$Gd$_{0.25}$Al$_2$. The $T_c$ values are marked at $\approx 55$ K and $\approx 72$ K, respectively. Both the alloys display the magnetic compensation behavior,
their $T_{\text{comp}}$ values are marked at 49 K and 33 K, respectively. Note that the $T_{\text{comp}}$ and $T_c$ are well separated in Pr$_{0.75}$Gd$_{0.25}$Al$_2$, while in Pr$_{0.83}$Gd$_{0.17}$Al$_2$ these differ only by 6 K. An inset in Fig. 4 (a) shows $M_{\text{FCC}}(T)$ response in Pr$_{0.83}$Gd$_{0.15}$Al$_2$ at $H = 55$ and 100 Oe. The zero crossover in lower field of 50 Oe and turnaround in 100 Oe can be seen at 50 K, $T_c$ of the sample is marked at 53 K.

(b) displays the in-field ($H = 50$ kOe) ac-susceptibility peak in Pr$_{0.83}$Gd$_{0.17}$Al$_2$ alloy at $\approx 48$ K. It matches with the turnaround temperature $T^*$ of the thermomagnetic curve in 50 kOe in this alloy. (Data not shown here).

In the Pr$_{1-x}$Gd$_x$Al$_2$ series with $x = 0.15, 0.17, 0.2$ and 0.25, the contribution to magnetization signals of the Pr$^{3+}$ and Gd$^{3+}$ moments are phase reversed and get compensated at specific temperatures ($T_{\text{comp}}$) below $T_c$. A comparison of the responses in $H \approx 10$ kOe in this series shows that the magnetization in Pr$_{0.83}$Gd$_{0.12}$Al$_2$ remains closest to the zero value at 5 K. The increase in the doping concentration of Gd$^{3+}$ ions in Pr$_{2-x}$Al$_2$ increases $T_c$ and decreases $T_{\text{comp}}$ values in the admixed series. At high fields, the magnetic compensation between the Pr$^{3+}$ and Gd$^{3+}$ manifests as a minimum at $T^*$ due to reorientation of these antiferromagnetically coupled entities. We have probed the magnetic reorientation process using the ac susceptibility measurements at high fields. At $H = 50$ kOe in Pr$_{0.83}$Gd$_{0.12}$Al$_2$ (Fig. 2(b)), the ac peak emerges close to the $T^*$ and can be identified with the reorientation of Pr and Gd moments with respect to the external field direction. In Pr$_{0.83}$Gd$_{0.17}$Al$_2$ the presence of the external magnetic field restricts the rare earth moments to respond to ac-fields and the two-peak structure in the ac-susceptibility shown in Fig. 4(b) disappears. However, the magnetic reorientation driven by the realignment of the rare earth moments produces ac-response and an additional peak emerges centred around $T^*$ (see the inset in Fig. 4(b)).

The positions of the peaks in the ac-responses (cf. Fig. 4) show the effect of the Gd$^{3+}$ substitution on the magnetic ordering process. The magnitude of the ac-susceptibility peak reduces by two orders with the 15 % substitution of Gd$^{3+}$ ions in pure PrAl$_2$. A possible mechanism could be the reduced magnetization of

---

**FIG. 4:** (Color online) (a) The $M_{\text{FCC}}(T)$ in nominal zero field in Pr$_{0.83}$Gd$_{0.17}$Al$_2$ and Pr$_{0.75}$Gd$_{0.25}$Al$_2$ alloys. The $T_{\text{comp}}$ and $T_c$ for both the alloys are marked. An inset in Fig. 4 (a) shows the $M_{\text{FCC}}(T)$ in Pr$_{0.83}$Gd$_{0.15}$Al$_2$ in 55 Oe and 100 Oe. $T_{\text{comp}}$ and $T_c$ values in Pr$_{0.83}$Gd$_{0.15}$Al$_2$ are 50 K and 53 K, respectively. (b) The nominal zero field cooled in-phase ac-susceptibility in alloys with $x = 0.15, 0.17$, and 0.25. Note the two peak ac-response in the Pr$_{0.83}$Gd$_{0.17}$Al$_2$. An inset in panel (b) shows the ac-susceptibility peak in 50 kOe (the corresponding $T_{\text{comp}}$ is marked) in Pr$_{0.83}$Gd$_{0.17}$Al$_2$.

---

**FIG. 5:** (Color online) (a) The temperature dependence of the specific heat in nominal zero field and 40 kOe in Pr$_{0.83}$Gd$_{0.17}$Al$_2$. Inset (a) in Fig. 5 shows a portion of $M_{\text{FCC}}(T)$ in $x = 0.17$ in $H = 50$ kOe. Inset (b) shows the ‘difference specific heat’, $\Delta C_p(T)/T = (C_p(T) - C_p(0))/T$, in Pr$_{0.83}$Gd$_{0.17}$Al$_2$ and Pr$_{0.83}$Gd$_{0.2}$Al$_2$.

---

In the Pr$_{1-x}$Gd$_x$Al$_2$ series with $x = 0.15, 0.17, 0.2$ and 0.25, the contribution to magnetization signals of the Pr$^{3+}$ and Gd$^{3+}$ moments are phase reversed and get compensated at specific temperatures ($T_{\text{comp}}$) below $T_c$. A comparison of the responses in $H \approx 10$ kOe in this series shows that the magnetization in Pr$_{0.83}$Gd$_{0.12}$Al$_2$ remains closest to the zero value at 5 K. The increase in the doping concentration of Gd$^{3+}$ ions in Pr$_{2-x}$Al$_2$ increases $T_c$ and decreases $T_{\text{comp}}$ values in the admixed series. At high fields, the magnetic compensation between the Pr$^{3+}$ and Gd$^{3+}$ manifests as a minimum at $T^*$ due to reorientation of these antiferromagnetically coupled entities. We have probed the magnetic reorientation process using the ac susceptibility measurements at high fields. At $H = 50$ kOe in Pr$_{0.83}$Gd$_{0.12}$Al$_2$ (Fig. 2(b)), the ac peak emerges close to the $T^*$ and can be identified with the reorientation of Pr and Gd moments with respect to the external field direction. In Pr$_{0.83}$Gd$_{0.17}$Al$_2$ the presence of the external magnetic field restricts the rare earth moments to respond to ac-fields and the two-peak structure in the ac-susceptibility shown in Fig. 4(b) disappears. However, the magnetic reorientation driven by the realignment of the rare earth moments produces ac-response and an additional peak emerges centred around $T^*$ (see the inset in Fig. 4(b)).

The positions of the peaks in the ac-responses (cf. Fig. 4) show the effect of the Gd$^{3+}$ substitution on the magnetic ordering process. The magnitude of the ac-susceptibility peak reduces by two orders with the 15 % substitution of Gd$^{3+}$ ions in pure PrAl$_2$. A possible mechanism could be the reduced magnetization of
the domains because of the antiferromagnetic coupling between the Gd\(^{3+}\) and Pr\(^{3+}\) moments. Pr\(_{0.85}\)Gd\(_{0.15}\)Al\(_2\) alloy appears to be on the verge of nucleating domains which has the dominance of Gd\(^{3+}\) moments (note the shoulder before the rising ac-peak in Fig. 4(b)), while a sharp peak near the same position has emerged in the ac-response in Pr\(_{0.85}\)Gd\(_{0.17}\)Al\(_2\). The sharp ac-peak in Pr\(_{0.85}\)Gd\(_{0.17}\)Al\(_2\) could be attributed to the freezing in of the domains in which magnetization from Gd\(^{3+}\) ions dominates close to the magnetic ordering temperature, followed by the ac-response of all the domains realigning during the magnetic compensation process. With 20-25 atomic \% substitution of the Gd\(^{3+}\) ions in PrAl\(_2\), the ac-response is completely dominated by the Gd moments and the dynamics of all the antiferromagnetically coupled Pr\(^{3+}\) moments also slows down right at the \(T_c\).

The magnetic reorientation process also leaves an imprint in the temperature dependence of the specific heat in Pr\(_{0.8}\)Gd\(_{0.2}\)Al\(_2\). In nominal zero field (Fig. 2(a)) the magnetic transition at \(T_c\) is captured, however, at high fields, an additional peak surfaces up at \(T^* \approx 39\) K. The emergence of the specific heat peak at \(T^*\) indicates that the magnetic reorientation has an attribute of the pseudo-phase transition in these alloys.

The temperature dependence of the specific heat in Pr\(_{0.85}\)Gd\(_{0.17}\)Al\(_2\) is shown in the Fig. 5. Inset (a) in Fig. 5 shows a portion of the thermomagnetic curve at high fields (50 kOe) which does not have the usual minimum close to \(T_{comp}\). The peak in the ‘difference specific heat’ data in \(H = 20\) kOe close to \(T_c\) in Pr\(_{0.85}\)Gd\(_{0.17}\)Al\(_2\) appears to be sharp (see inset (b) in Fig. 5). This sharpness can be compared with the peak in its high field ac-susceptibility data (see inset in Fig. 4(b)). These two sharp features support the notion of field-induced phase transition at compensation temperature, as advocated by Chen et al.\(^{17}\) in the Sm\(_{0.98}\)Gd\(_{0.02}\)Al\(_2\) alloy, where the magnetic compensation phenomenon was attributed to the special properties of the Sm\(^{3+}\) ions\(^{18}\). Chen et al.\(^{17}\) also observed the sign change (-ve to +ve) in the magnetoresistance across \(T_{comp}\) in Sm\(_{0.98}\)Gd\(_{0.02}\)Al\(_2\) alloy. We do not observe this correlation in Pr\(_{1-x}\)Gd\(_x\)Al\(_2\) series. However, the sign change in the Hall resistance correlates to the results in Sm\(_{0.98}\)Gd\(_{0.02}\)Al\(_2\) alloy. The spin-disorder resistivity which freezes at \(T_c\), again appears to get alive while cooling the sample in the presence of the high external magnetic field. This can occur if the spin-orbit configuration continues to undergo a rearrangement, which is the case in these alloys during in-field cooling. We believe that the oscillatory nature of the magnetoresistance (see Insets (a) and (b) in Fig. 3) is a generic feature in the admixed rare earth intermetallics showing the compensation behavior. It is fruitful to recall here that oscillatory magneto-resistance response also stands reported in a single crystal of Nd\(_{0.75}\)Ho\(_{0.25}\)Al\(_2\)\(^{18}\).

To summarize, the magnetic compensation behaviour in the Pr\(_{1-x}\)Gd\(_x\)Al\(_2\) series has been studied in the contemporary context. The fingerprint of magnetic turnaround across \(T_{comp}\) is identified in the in-field ac-susceptibility data. The temperature dependence of the in-field specific heat supports the notion of field-induced phase transition across the \(T_{comp}\) and it corroborates the earlier observation in the single crystal Nd\(_{0.75}\)Ho\(_{0.25}\)Al\(_2\)\(^{18}\). A curious oscillatory behavior of the magnetoresistance as a function of temperature is observed in the Pr\(_{0.8}\)Gd\(_{0.2}\)Al\(_2\) alloy. The change in sign of the Hall voltage across \(T_{comp}\) is also an important observation. It should be of interest to explore the temperature dependences of the 4f-spin and 4f-orbital contributions of Pr\(^{3+}\) and 4f-spin contribution of Gd\(^{3+}\) via x-ray magnetic circular dichroism measurements in single crystals of Pr\(_{1-x}\)Gd\(_x\)Al\(_2\) alloys.

We thank S. K. Dhar and P. L. Paulose for their association in the initial phase of this work. We also acknowledge D. D. Buddhikot for his help in some of the measurements.

---

1. H. J. Williams, J. H. Wernick, E. A. Nesbitt, R. C. Sherwood, J. Phys. Soc. of Jpn. 17-B1, 91 (1962).
2. H. Adachi and H. Ino, Nature 401, 148 (1999).
3. H. Adachi, H. Ino, H. Miwa, Phys. Rev. B 59, 11445 (1999).
4. H. Adachi, H. Ino, H. Miwa, Phys. Rev. B 56, 349 (1997).
5. H. Adachi, H. Kawata, H. Hashimoto, Y. Sato, I. Matsumoto, Y. Tanaka, Phys. Rev. Lett. 87, 127202 (2001).
6. J. W. Taylor et al. Phys. Rev. B 66, 161319(R) (2002).
7. X. H. Chen, K. Q. Wang, P. H. Hor, Y. Y. Xue, C. W. Chu, Phys. Rev. B 72, 054436 (2005).
8. Z. H. Wu et al., J. Phys. D: Appl. Phys. 38, 3567 (2005).
9. S. Qiao et al., J. Electron Spectrosc. Relat. Phenom. 144-147, 759 (2005).
10. A. K. Grover, D. Rambabu, S. K. Dhar, S. K. Malik, R. Vijayaraghavan, G. Hilbcher, H. Kirchmayer, Proc. DAE Nuclear Physics and Solid State Physics Symposium (India) 26C, 252 (1983).
11. A. K. Grover and S. K. Dhar, in Experimental and Theoretical Aspects of Valence Fluctuations, edited by L. C. Gupta and S. K. Malik (Plenum, New York, 1987), p. 481.
12. A. K. Grover et al., in Proc. DAE SSPS, Solid State Physics (India) 52, 27 (2007).
13. V. C. Rakhecha et al., ibid, 51, 949 (2006).
14. P. D. Kulkarni, A. Thamizhavel, P. L. Paulose, D. D. Buddhikot, A. K. Nigam, S. Ramakrishnan and A. K. Grover, ibid, 53, 1125 (2008).
15. K. H. J. Buschow and A. M. van Diepen, Phys. Rev. B 8, 5134 (1973).
16. S. K. Malik and R. Vijayaraghavan, Pramana-J. Phys. 3, 122 (1974).
17. S. K. Malik, R. Vijayaraghavan, S. K. Garg and R. J. Rimpmeester, Pure Appl. Chem. 40, 223 (1974).
18. P. D. Kulkarni, A. Thamizhavel, V. C. Rakhecha, A. K. Nigam, P. L. Paulose, S. Ramakrishnan and A. K. Grover, Europhys. Lett. 86, 47003 (2009).