Exploring the Magnetic Anisotropy of $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$

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The magnetism in the series of compounds $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ (x = 0 to 1) was studied using magnetisation and susceptibility. The ordering temperatures are found to increase almost linearly with x, while the saturation magnetisation in moderate fields have a minimum at x ~ 0.25. The Gd moments couple antiferromagnetically with the Nd/Pr.

1. Introduction

The RNi system (R = light rare-earth elements) is a convenient test bed for the study of the interplay of exchange interactions and the crystal electric field effect (CEF) in determining magnetic properties and structures. On initial inspection, such systems would appear relatively straightforward, a Cr-B type orthorhombic crystal structure with only one rare earth site and non-magnetic nickel. Nevertheless, there are some interesting magnetic behaviours. NdNi and PrNi both order ferromagnetically at low temperature but with mutually perpendicular easy axes [1]. Furthermore, such magnetic anisotropy is maintained in the mixed $Pr_{0.5}Nd_{0.5}Ni$ compound. That is, there are two magnetic sublattices, Nd and Pr, aligned at 90° to each other in low applied magnetic fields and at low temperatures [2]. Other studies have revealed that the addition of S-State Gd ions to make compounds of the form $Nd_{1-x}Gd_xNi$ or $Pr_{1-x}Gd_xNi$ result in ferrimagnets, wherein the Gd and Pr/Nd ions are antiferromagnetically coupled [3,4]. (Excluding the end point compounds x = 0 and 1 which are simple ferromagnets of course.) This paper outlines a preliminary study of both these effects embodied in a single system, namely the series $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$. Here the main interest is what configuration the Gd may adopt in the low x region. There is some potential, with the particular values of x, for the Gd spins to be frustrated.

2. Experimental Details

Poly-crystalline specimens of $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$, with x= 0, 0.25, 0.6, 1, were formed by melting together high purity constituents using an argon arc furnace. The ingots were flipped and remelted at least five times to ensure an uniform result. The phase integrity of the resulting alloys was verified with powder x-ray diffraction. The magnetic ordering (Curie) temperatures were determined using a Vibrating Sample Magnetometer (VSM), while quantitative magnetisation & susceptibility were measured with a SQUID magnetometer (MPMS).

3. Results

3.1 Transition Temperatures

The magnetic transition temperatures, T_c, were determined from the VSM data of figure 1. Estimated T_c's, listed in Table 1, vary almost linearly with x. These values were confirmed by a second set of magnetisation versus temperature measurements carried out with the MPMS in an applied field of 1.0 T. This MPMS data was then used to produce inverse susceptibility ($1/\chi$) vs. T plots and the Curie-Weiss law was applied to the high temperature, straight line portion to derive values of the paramagnetic Curie Temperature, θ_p, and the effective moment M_{eff}. Figure 2 is an example of this data for the case x = 0.25. The theoretical values for the effective moment (M_{theo}) were derived assuming a proportional average of free ion values and are in reasonable agreement with the observations. The trend of θ_p versus x is similar to that of T_c for our polycrystalline samples. Note that for single crystals of $Nd_{1-x}Gd_xNi$ and $Pr_{1-x}Gd_xNi$ a minimum in θ_p was seen along the axes of easy magnetisation near x ~ 0.25 [3,4]. This effect is a clear indication of antiferromagnetic coupling between the Gd and Pr/Nd. Here with polycrystalline material we need to examine the saturation magnetisation to show this feature.

Table 1. Parameters for $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ polycrystals with various x.

x	T_c (K)	θ_p (K)	M_{eff} (μ_B)	M_{theo} (μ_B)	M_s (μ_B/ion) (1 T)	M_s (μ_B/ion) (2 T)
0	20	6	3.72	3.60	1.2	1.3
0.25	31	22	4.68	4.69	0.30	0.48
0.6	45	37	6.73	6.56	3.1	3.2
1	75	75	8.52	7.94	7.0	7.2

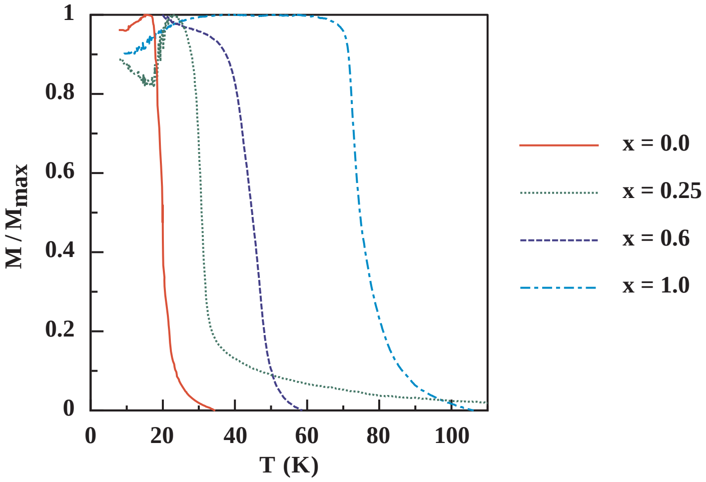

Fig. 1. Magnetisation data recorded with a VSM for the various alloy compositions (x). The vertical scale has been normalised, the applied field was 0.1 T and the derived transition temperatures are listed in Table 1.

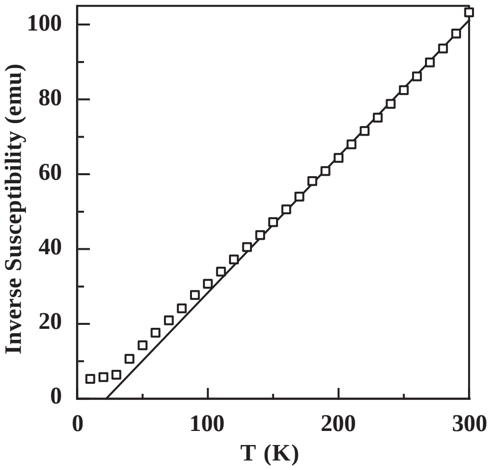

Fig. 2. Inverse susceptibility of $(Pr_{0.5}Nd_{0.5})_{0.75}Gd_{0.25}Ni$ from 5 to 300 K in an applied field of 1 T. The high temperature region of the data is fitted according to the Curie-Weiss law, yielding $\theta_P = 22$ K and $P_{eff} = 4.68\ \mu_B$.

3.2. Magnetisation in the Ordered Phase

Low temperature magnetisation data for $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ are shown in figure 3. The saturation (ordered) moment M_s is taken as the values at 2 T (following [3,4] enough to remove domains but minimise moment rotation). Figure 4 is a compilation of M_{eff} and M_s values. While the effective (paramagnetic) moment M_{eff} scales with x, the saturation (ordered) moment M_s shows a minimum ~ $x = 0.25$ reflecting the AF coupling of Gd and Pr/Nd moments. Indeed $x = 0.25$ is close to the point of no net moment reflected in little observed spontaneous magnetisation in this case. The choice of 2 T as the field to compare M_s, while fine for single crystal,s is not optimal here in a polycrystal. Therefore M_s values at 1 T are also shown and the trend at $x = 0.25$ is towards zero as the field is lowered.

In figure 5 the M_s for $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ are compared to the easy axis single crystal results for $Nd_{1-x}Gd_xNi$ and $Pr_{1-x}Gd_xNi$. The form of all three data sets is similar. (Note our GdNi has $M_s = 7.2\ \mu_B$ consistent with the literature [5] while the values for other compounds from [3,4] are slightly low by admission of the authors). Finally looking again at figure 4, it is noted that M_s versus x for $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ is modelled well using a simple linear variation of average moment plus AF coupling of the Gd and the Pr/Nd, as used by [3,4], but with additional allowance for the distribution of directions (polycrystal) at low x. That is, at low x, the Pr and Nd moments are tied to their crystal field determined easy axes and the Gd lies antiparallel to either of these, while beyond the minimum near $x = 0.25$, where the total Gd moment is the larger, the trend is for the Gd moment to track the applied field and the Pr/Nd to lie antiparallel (to the Gd).

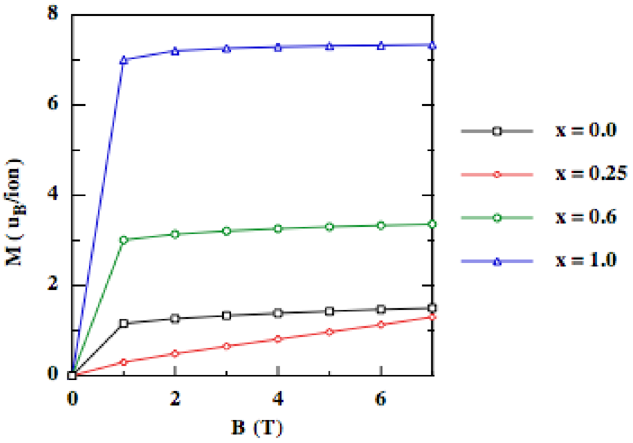

Fig. 3. Magnetisation data for the various values of x at 5 K and up to an applied field of 7 T.

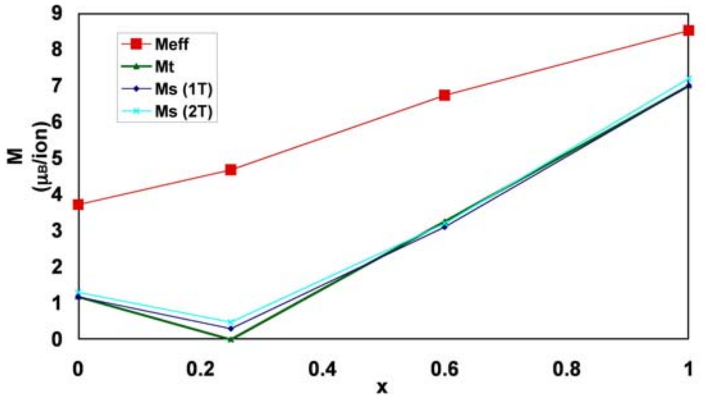

Fig. 4. Effective magnetic moment M_{eff}, saturation magnetic moment M_s and modelled (total) saturation moment M_t, against x.

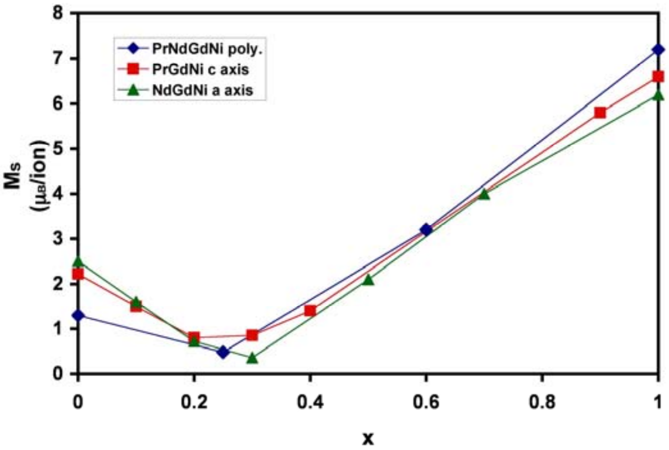

Fig. 5. Comparison of the saturation magnetic moment M_s (2T) against x for single crystal $Pr_{1-x}Gd_xNi$ and $Nd_{1-x}Gd_xNi$ [3,4] with $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$.

4. Discussion

This study of $(Pr_{0.5}Nd_{0.5})_{1-x}Gd_xNi$ confirms earlier results of the presence of antiferromagnetic coupling between Gd and the lighter rare earths in RNi. The data confirms the robustness of the magnetic anisotropies and indications of two magnetic sublattices at low x (< 0.25). However with the limited range of x and polycrystal materials alone, the exact positioning of the Gd in this regime is not revealed specifically, only that it must lie antiparallel to either the Pr or Nd. A single crystal study is planned to determine the exact configuration of the Gd moments at low x.

Acknowledgments

The authors wish to thank B. Saensunon for assisting with sample preparation and G.A. Stewart for discussions.

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