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Magnetic anomalies in Gd_2PdSi_3

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Abstract. The results of ac and dc magnetic susceptibility, thermopower and Hall effect measurements of a compound, Gd_2PdSi_3, establish that this compound orders magnetically below $T_N = 20$ K. Though the ordering appears to be of an antiferromagnetic-type, the paramagnetic Curie temperature is positive with the magnitude being nearly the same as that of T_N, suggestive of the existence of ferromagnetic correlations. The thermopower at 300 K is large, apparently due to Pd $4d$ electrons, decreasing monotonically with temperature. There is a change in the sign of Hall constant well below T_N. Also considering the observation of Kondo-like characteristics above 21 K earlier by us, the overall thermal, transport and magnetic behaviour of this compound is interesting.

Keywords. Gd_2PdSi_3; magnetic susceptibility; thermopower; Hall effect.

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Though the Gd compounds have been traditionally believed to be uninteresting from the magnetism point of view due to the well-localised nature of $4f$ orbital, we have recently brought out new features [1] in the thermal and transport behaviour of some Gd alloys, which bear significant relevance to the current trends in Kondo lattice, non-Fermi liquid and giant magnetoresistance anomalies. In particular, during the course of investigation of a new class of compounds of the type, R_2PdSi_3 [2–5], crystallizing in an AlB_2-derived structure [6, 7], we noticed [3] Kondo-lattice-like characteristics in the heat-capacity C and electrical resistivity (ρ) behaviour as well as large negative magnetoresistance in the vicinity of magnetic ordering temperature in Gd_2PdSi_3. In order to characterize this compound further, we carried out ac and dc magnetic susceptibility (χ) measurements on the alloys, $Gd_{2-x}Y_xPdSi_3$, as well as Hall effect and thermopower measurements on Gd_2PdSi_3, the results of which are reported here.

The polycrystalline samples, $Gd_{2-x}Y_xPdSi_3$ ($x = 0.0$, 0.4, 1.0, 1.6), employed in this study are the same as those used in ref. 3. The dc magnetic susceptibility measurements were carried out in the temperature interval 2–300 K in the presence of a magnetic field of 2 kOe; in addition, zero-field-cooled (ZFC) and field-cooled (FC) measurements (H = 100 Oe) were performed below 70 K for all the samples. Ac χ measurements (2–100 K) were also performed employing an ac field of 0.8 Oe at different frequencies, 9, 90 and 890 Hz. The Hall effect measurements were also performed by a conventional

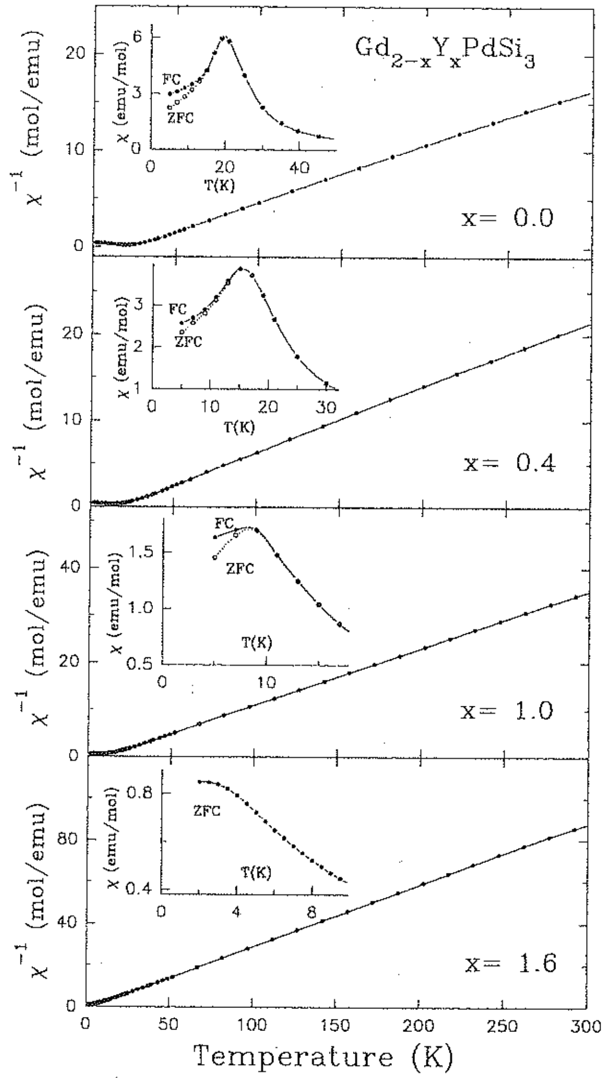

Figure 1. Inverse susceptibility (χ) as a function of temperature for $Gd_{2-x}Y_xPdSi_3$ ($x = 0.0$, 0.4, 1.0 and 1.6). A line through the linear region is drawn. The insets show the low temperature χ data, field-cooled and zero-field-cooled, in the presence of H = 100 Oe and the lines through the data points serve as guides to the eyes.

dc probe method employing a magnetic field of 10 kOe and the thermopower was measured by differential method using Au-Fe(0.07%)-chromel thermocouples.

Inverse susceptibility as a function of temperature for all the alloys is shown in figure 1. The plot is found to be practically linear above 30 K and the value of the effective moment ($\mu_{eff} = 8.2\mu_B$) obtained from this plot is found to be marginally higher than that expected for trivalent Gd, and could be arising from polarisation of the conduction band. There is a distinct peak in χ for $x = 0.0, 0.4$ and 1.0 at 19, 15 and 9 K respectively; also

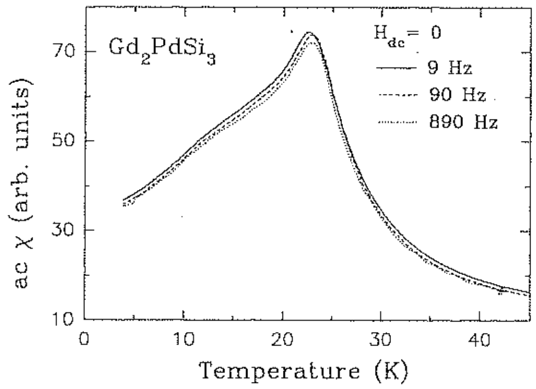

Figure 2. AC susceptibilty of Gd_2PdSi_3 at three different frequencies.

there is a tendency for χ to level off below 3 K for $x = 1.6$; these features indicate the onset of long range antiferromagnetic order at the respective temperatures. It should be noted that the values of the paramagnetic Curie temperature (θ_p) are found to be nearly the same (19, 14.5, 10 and 2.5 K respectively) as the T_N determined from the peak position (figure 1, insets); however, it is interesting that the sign of θ_p is positive, in contrast to the negative sign expected for antiferromagnets, thereby suggesting the existence of ferromagnetic correlations. The FC and ZFC data are more revealing (figure 1). For $x = 0.0$, ZFC and FC data are practically the same above 15 K, while the difference can be distinctly seen to set in below 15 K, but not at the peak; however, for $x = 0.4$ and 1.0, the difference sets in close to the peak itself. This may be related to the fact that there are two magnetic transitions for the $x = 0.0$ alloy, one at 21 K (from the minority Gd ions at the 2*b* site) and the other at 15 K (for the majority Gd ions at the 6*h* site) as shown by Mössbauer measurements (ref. 3); the ZFC-FC difference may arise from the Gd ions at 6*h* site ordering at 15 K. In order to address the question whether the ZFC-FC behaviour (which is often seen even in some antiferromagnets [8]) arises from spin-glass behaviour, we also performed ac χ measurements at various frequencies (data shown in figure 2 only for the parent compound) and we do not find any upward shift of the peak with increasing frequency, as expected for the spin-glass phenomena. The present data thus do not appear to show evidence for spin-glass freezing, definitely not above 21 K. In contrast to this situation, the compound U_2PdSi_3 has been shown to exhibit spin-glass behaviour [9]. The shoulder around 15 K in ac χ data may be related to the magnetic ordering from 6*h* site.

The results of thermopower measurements are shown in figure 3(a) both for Gd_2PdSi_3 and Lu_2PdSi_3. It is to be noted that the thermopower of the Gd sample is large at 300 K, which is true even for Lu_2PdSi_3, and is comparable to the magnitude known for the Pd metal [10], thereby indicating large Pd 4*d* density of states at the Fermi level and consequent spin fluctuations similar to that of YCo_2 [11]. The values are larger (about $-10\mu V/K$) for the Gd sample, compared to the Lu sample. S varies (decreases) linearly down to about 20 K (as expected). There is a tendency to a constant value for a narrow temperature range below 20 K. Since this shoulder appears in the non-magnetic compound as well, the feature due to magnetic ordering is not clearly separable from the phonon

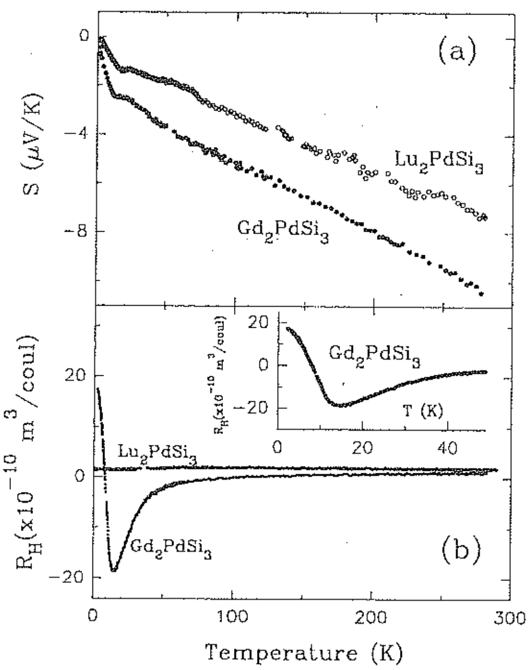

Figure 3. (**a**) The thermopower and (**b**) the Hall coefficient (R_H) of Gd_2PdSi_3 and Lu_2PdSi_3 as a function of temperature. The low temperature data of R_H are shown in an expanded form for the Gd sample in the inset.

drag effect. Thus the magnetic ordering below 21 K does not have any noticeable effect on S in the Gd alloy. In contrast to this, the Hall coefficient (R_H) of the Gd sample is completely different from that of the Lu sample (figure 3b). While the R_H is a small positive number for Lu sample at all temperatures, it is negative above 8 K for the Gd_2PdSi_3 with a distinct peak in the vicinity of magnetic ordering. We, however, find that the peak occurs only near 15 K (figure 3b, inset), without any noticeable feature at 20 K and thus the 20 K transition is not tracked by Hall measurements. A noteworthy finding is that there is a sign reversal below 8 K and it is the same temperature at which the resistivity also shows an anomaly [3]; possibly band structure effects due to magnetic superzone-boundary effects are responsible for this anomaly. In the paramagnetic region (above 21 K), the R_H data were fitted to an expression $R_H = R_0 + A/(T - \theta_p)$ (where A is a constant) and this fit clearly showed that the anomalous Hall term dominates over the entire temperature range above 21 K. Therefore, it is difficult for us to extract the carrier concentration from this data to explore whether there are any changes as one approaches T_N.

All these results provide evidence to the fact [3] that Gd_2PdSi_3 exhibits magnetic ordering below 20 K, with an additional transition at 15 K. This implies that the resistivity minimum observed around 45 K in this compound has some physical significance as discussed in ref. 3. Thus, this compound appears to be one of the interesting compounds considering Kondo-lattice-like characteristics in resistivity and heat capacity, large negative magnetoresistance in the vicinity of magnetic ordering temperature [3], antiferromagnetism with a positive θ_p, possible magnetic superzone boundary gaps resulting in negative temperature coefficient of ρ and sign reversal of Hall coefficient in the ordered state and large thermopower around 300 K.

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