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LETTER TO THE EDITOR

Magnetic relaxation behaviour in $Gd_{2-x}Y_xPdSi_3$ alloys

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Abstract. The results of time-dependent remanent magnetization (M_{IRM}) behaviour (magnetic relaxation studies) in polycrystalline $Gd_{2-x}Y_xPdSi_3$ alloys in a magnetically ordered state are reported. We observed that M_{IRM} undergoes slow relaxation with time. We have also measured ac susceptibility as a function of frequency; however, we did not find any upward shift of the peak-temperature with an increase of frequency, which suggests that these alloys are not spin-glasses. Measurements on single-crystalline Gd_2PdSi_3 show that the magnetic relaxation is an intrinsic property of the materials.

During the investigation of a new class of ternary compounds [1–5] of the type R_2PdSi_3, crystallizing in a AlB_2-derived hexagonal structure [6,7], we noted some interesting features, uncharacteristic of Gd alloys, for the compound Gd_2PdSi_3. This compound exhibits a minimum in temperature-dependent electrical resistivity (ρ) at around 45 K, well above the Néel temperature ($T_N = 21$ K), which is suppressed by the application of a moderate magnetic field (H); there is a pronounced enhancement of heat capacity (C) over a wide temperature range above T_N [2]. These properties mimic the behaviour of Ce- or U-based Kondo lattices with a magnetic ground state. The magnitude of (negative) magnetoresistance keeps increasing with decreasing temperature below 70 K, attaining large values at low temperatures, a finding of interest to the field of giant magnetoresistance. Through these results, we have emphasized the need to explore the existence of a novel magnetic precursor effect (e.g. formation of magnetic polarons) as a cause of resistivity minimum and giant magnetoresistance phenomena [8]. Other interesting findings [5] are: the *positive* sign of the paramagnetic Curie temperature with the same magnitude as T_N; a large thermopower at 300 K; and a change in the sign of the Hall constant well below T_N. In view of the observation of such interesting features, we have subjected this compound and its derivatives to further magnetic characterization. In this article, we pay special attention to the magnetic relaxation behaviour (isothermal magnetization, M_{IRM}, as a function of time, t) of these alloys in the magnetically ordered state.

The polycrystalline samples, $Gd_{2-x}Y_xPdSi_3$ ($x = 0.0$, 0.4, 1.0 and 1.6), employed in this investigation are the same as those used in previous studies [2, 5]; these samples were prepared by arc melting followed by annealing at 750 °C for one week in evacuated sealed quartz tubes. We performed magnetic relaxation measurements at low temperatures, employing a SQUID magnetometer made by Quantum Design. For this purpose, we cooled the sample in zero-field from 100 K (far above T_N) to 5 K (or 2 K), then switched on a magnetic field (H) of 5 kOe for 5 min; the magnetization was then measured as a function of time after the field was switched off by quenching the field (with the magnet reset option of the SQUID magnetometer). We

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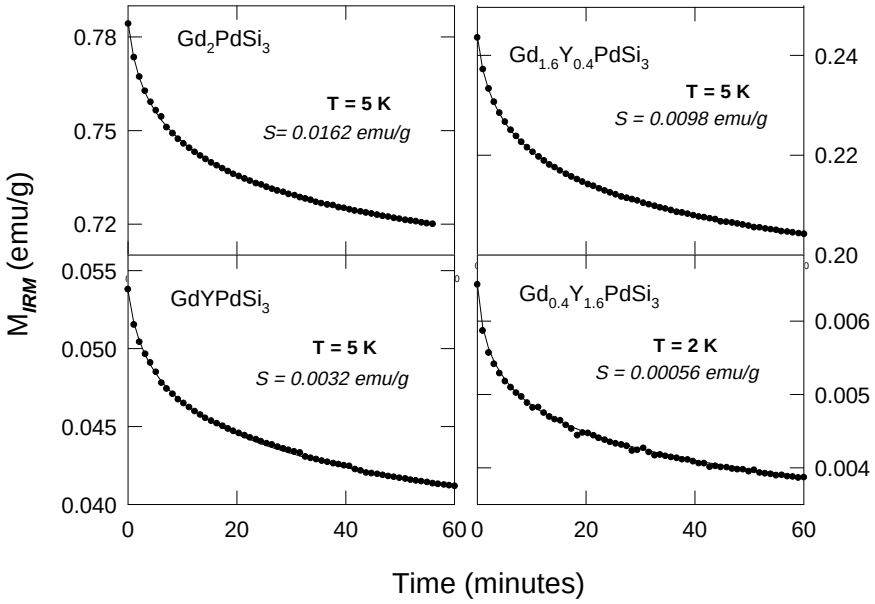

Figure 1. Time dependence of isothermal magnetization, $M_{\rm IRM}$, of polycrystalline samples of the $Gd_{2-x}Y_xPdSi_3$ series measured in the magnetically ordered state. The continuous lines are the fits to the function $M_{\rm IRM}(t) = M_0 - S\ \ln(1 + t/t_0)$. The values of S are given in the plots.

also performed magnetic hysteresis loop studies to determine the shape of the demagnetization curves and the values of the remanent magnetization. We made similar measurements on single crystals of Gd_2PdSi_3, prepared using a tetra-arc furnace, to explore the role of sample perfection or grain boundaries. Using a Lakeshore Magnetometer (Model 7225), ac susceptibility (χ) measurements were performed on the polycrystals in the vicinity of $T_{\rm N}$ at various frequencies.

We show the $M_{\rm IRM}$ behaviour as a function of t, recorded as described above, in figure 1; the measurements were performed at 5 K for all the compositions, except for $x = 1.6$, in which case the data were collected at 2 K in order that the temperature be below $T_{\rm N}$. At this point, it may be mentioned that the isothermal magnetization at these temperatures (not shown here) increases nearly linearly with H up to approximately 10 kOe, beyond which there is a tendency to saturate at higher fields (measured up to 40 kOe) with a small remanence and coercivity (vide infra). The main finding in the $M_{\rm IRM}$ data is that, immediately after the field is switched off, the value is significantly large and it falls off slowly with time. After waiting for one hour, the value drops from the initial zero-field values by about 8, 15, 26 and 35% for $x = 0.0$, 0.4, 1.0 and 1.6, respectively. The plot of $M_{\rm IRM}$ versus t could be fitted to a logarithmic function, $M_{\rm IRM}(t) = M_0 - S\ln(1 + t/t_0)$. The value of the (viscosity) coefficient S systematically decreases with decreasing Gd concentration. The coefficient t_0 depends on the measuring conditions of the magnetometer and has only limited physical relevance. At this juncture, we would like to mention that even if the data were collected in the presence of a small residual magnetic field after reducing the current through the magnet leads to zero (without utilizing the magnet reset option), the behaviour is qualitatively the same, with the values of S remaining nearly constant (within 5%); there is, however, a change in the values of M_0. Needless to say, there is no such magnetic relaxation behaviour above respective $T_{\rm N}$ values, and the values of $M_{\rm IRM}$ drop to zero within seconds after the field is quenched. In order to explore whether the above-discussed magnetic relaxation behaviour is intrinsic to the

material, the measurements were performed on the single crystals of Gd_2PdSi_3 at selected temperatures in the magnetically ordered state. We did indeed observe magnetic relaxation on the single crystals also. The values of the initial remanent magnetization M_0 and the viscosity S are given in table 1 for two different crystallographic directions at different temperatures. The remanent magnetization of the polycrystalline Gd_2PdSi_3 sample at 5 K is close to the mean value of the results for the single crystal, and the values of the coefficient S differ by less than a factor of two.

Table 1. Dependence of the magnetic relaxation of a Gd_2PdSi_3 single crystal on field direction and temperature fitted to $M_{\mathrm{IRM}}(t) = M_0 - S\ln(1 + t/t_0)$.

	$H \parallel$ [001]		$H \parallel$[100]	
T (K)	M_0 (emu g^{-1})	S (emu g^{-1})	M_0 (emu g^{-1})	S (emu g^{-1})
5	0.39	0.0087	0.92	0.013
10	0.19	0.0059	0.50	0.012
15	0.03	0.0016	0.04	0.0021

We have also performed ac susceptibility measurements at various frequencies on the polycrystalline samples, in order to explore possible spin-glass freezing [9]. The results are shown in figure 2. The peak positions correspond to bulk magnetic ordering. It is to be noted that there is no upward shift of the peak with increasing frequency, in contrast to the behaviour of spin-glasses. It is emphasized that there is no difference between the field-cooled and zero-field-cooled χ values below 21 K in single crystals [10]. Therefore, the magnetic

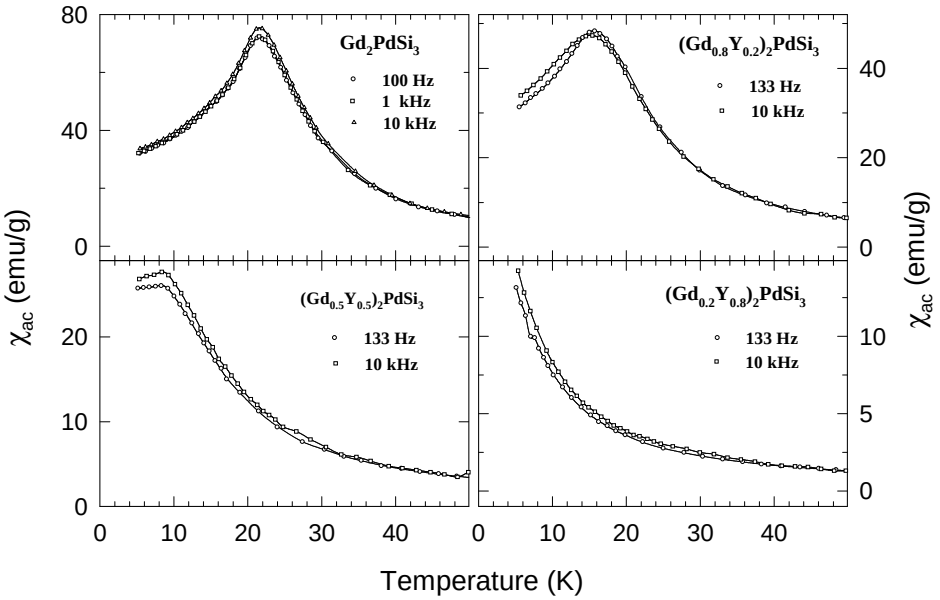

Figure 2. The ac susceptibility data at different frequencies below 50 K for the $Gd_{2-x}Y_xPdSi_3$ alloys. The peak positions represent the onset of long-range magnetic ordering. The peak is below 4 K for $x = 1.6$. The lines through the data points are guides to the eyes.

relaxation behaviour described above does not have its origin in spin-glass freezing. In general, if the magnitude of the applied magnetic field for magnetic relaxation studies is much larger than the coercive field [9, 11], the observed magnetic relaxation is usually attributed to spin-glass phenomena, considering that the relaxation behaviour is also typical of ferromagnetic materials [12–15]. For this purpose, we have also obtained the values of the coercive field from the carefully measured hysteresis loops, and the values are found to be 125 Oe (at 5 K), 52 Oe (at 5 K), 16 Oe (at 5 K) and 7 Oe (at 2 K), respectively, for the various sample compositions, and thus the field employed is far above the coercive field values. Thus, the present results are somewhat intriguing, warning of the need to be careful when interpreting the magnetic relaxation data in terms of spin-glass behaviour. It may be added that, for a number of ferromagnetic materials, Barbier [16] reported an increase of S with increasing coercivity, and we observed similar correlations in the present series of alloys as well.

Summarizing, the polycrystalline $Gd_{2-x}Y_xPdSi_3$ samples as well as single crystals of Gd_2PdSi_3 show a logarithmic time dependence of M_{IRM}, the origin of which does not lie in spin-glass behaviour. The remanent magnetization and the coefficient S for the logarithmic magnetic relaxation decrease with increasing Y content (see figure 1) or increasing temperature (see table 1). Finally, it is interesting to note [17] that, unlike the magnetic relaxation behaviour of Gd_2PdSi_3, polycrystalline samples of Eu_2PdSi_3 do not show any such relaxation in the magnetically ordered state.

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