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Magnetoresistance studies on RPd₂Si (R = Tb, Dy, Lu) compounds

R Rawat¹, Pallavi Kushwaha¹ and I Das²

¹ UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore-452001, India

² Saha Institute of Nuclear Physics, Experimental Condensed Matter Physics Division, 1/AF Bidhannagar, Kolkata-700064, India

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Abstract

Magnetoresistance studies on RPd_2Si (R = Tb, Dy and Lu) compounds show large negative magnetoresistance (MR) in TbPd₂Si and DyPd₂Si near the magnetic ordering temperature. Positive MR at low temperature in the ferromagnetic Dy compound is shown to arise from the orbital contribution (the Lorentz force effect). As a consequence, a deviation from the linear relation between MR and isothermal entropy change (i.e. the magnetocaloric effect) is observed. In the case of the Tb compound, anomalous magnetoresistance behavior is observed at 3 K, where the resistivity is found to be different before and after magnetic field cycling. These results suggest complex magnetic behavior in TbPd₂Si.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The ternary rare earth series RPd₂Si shows interesting magnetic behavior at low temperatures. These compounds show competing antiferro- and ferromagnetic interactions, which result in different ground states for these compounds [1]. Gd and Tb compounds order antiferromagnetically around 13.5 and 21 K respectively, even though the Curie temperatures are positive for both of them, whereas the DyPd₂Si compound shows a ferromagnetic order below 9 K. Both Gd and Tb compounds show a metamagnetic transition with the application of magnetic field [1]. Besides this, a large magnetocaloric effect has been observed near the magnetic transition temperature in Gd and Dy compounds [2, 3]. The correlation between magnetoresistance (MR) and the magnetocaloric effect (MCE) near the magnetic transition has been shown in some rare earth intermetallics [4, 5]. In these studies it has been shown that the compounds with large MCE also show large MR, though the reverse is not always true. Subsequently, a proportional relation between MR and MCE has been shown in a variety of metallic ferromagnets [6]. Therefore, we expect large magnetoresistance in these compounds.

In this paper, MR studies on RPd_2Si (R = Tb, Dy, Lu) compounds are presented. All the compounds show large MR. In the case of ferromagnetic DyPd₂Si, the Lorentz force effect was found to be responsible for positive MR. It is shown that

this effect gives rise to a deviation from the linear relation of MR with MCE. Besides the large positive MR, the Tb compound shows anomalous thermomagnetic irreversibilities below 9 K, indicating complex magnetic behavior in this system.

2. Experimental details

The polycrystalline RPd₂Si (R = Tb, Dy and Lu) compounds used in the present study were from the same ingots used in earlier studies [2, 3]. All the compounds were found to be single phase and crystallize in a Fe₃B-type orthorhombic structure. The resistivity and magnetoresistance measurements were carried out by the standard linear four probe method up to 8 T magnetic field on a rectangular shape sample (typical size 15 mm × 2 mm × 1.5 mm). For in-field measurements magnetic field is applied parallel to the current direction. Magnetization measurements were performed using the vibrating sample magnetometer option of a physical property measurement system (PPMS) from Quantum Design, USA.

3. Results and discussion

Figure 1 shows the resistivity behavior of all the compounds in the presence of zero and 8 T magnetic field. The absolute value of resistivity can have large uncertainty due to the brittle



Figure 1. Resistivity as a function of temperature at 0 and 8 T magnetic field for the compounds (a) TbPd₂Si and (b) DyPd₂Si. The bottom inset shows the temperature dependence of magnetoresistance for 8 T magnetic field for respective compounds. The top inset in (a) shows the temperature dependence of the magnetization measured in 500 Oe magnetic field during warming for zero field cooled TbPd₂Si.

nature of samples. Resistivity in the absence of magnetic field shows two transitions for the Tb compound and one transition for the Dy compound. The magnetization measurement shown in the top left inset for the Tb compound also indicates two antiferromagnetic transitions. The transition temperatures are found to be 19 and 9 K for TbPd₂Si and 9 K for DyPd₂Si. These transition temperatures are in agreement with earlier heat capacity [3] and magnetization studies [1]. The resistivity in the presence of 8 T magnetic field shows large negative magnetoresistance near the magnetic transition for both compounds. However, in the case of the ferromagnetic Dy compound the 8 T resistivity is found to be higher than zero field resistivity at low temperatures. Generally, positive magnetoresistance is unexpected in ferromagnetic compounds. To find out the origin of the magnetoresistance behavior, isothermal magnetoresistance measurements were carried out at different temperatures.

The longitudinal magnetoresistance (MR) as a function of magnetic field at various constant temperatures is shown in figure 2 for all the compounds. The MR is defined as $\Delta \rho / \rho = \{\rho(H) - \rho(0)\}/\rho(0)$, where $\rho(0)$ is the resistivity in zero field and $\rho(H)$ is the resistivity in the presence of magnetic field *H*.



0

Figure 2. (a)–(c) Magnetic field dependence of magnetoresistance (MR) for the compound RPd₂Si (R = Tb, Dy and Lu), respectively. The metamagnetic transition for TbPd₂Si and positive MR for DyPd₂Si are highlighted. The inset in (b) shows the magnetic contribution to MR for DyPd₂Si, obtained by subtracting MR of LuPd₂Si from DyPd₂Si. The inset in (c) shows Kohler's plot for LuPd₂Si.

In the paramagnetic regime the magnetoresistance shows $-H^2$ dependence, the magnitude of which increases close to the magnetic transition temperature. Below the magnetic ordering temperature the MR at low field remains negligible for small field values for TbPd₂Si. These observations are in accordance with the antiferromagnetic nature of this compound, where MR is expected to be negligible and positive [7]. However with a further increase in magnetic field, MR decreases rapidly and shows large negative values and finally tends to saturate at higher magnetic field. The antiferromagnetic-like behavior at

[a]

low field and ferromagnetic-like behavior at high field indicates a field induced metamagnetic transition in this compound. The observed metamagnetic transition is in accordance with earlier studies in this compound [1, 3]. Besides this, at low temperature, the slope of the MR curve is slightly positive above 4 T applied magnetic field (see 3, 6 and 10 K data in figure 2(a)).

In the case of the Dy compound the MR behavior is typical of a ferromagnet around 10 K, which is close to the ferromagnetic transition in this compound. However, at low temperature and high field, the magnitude of the negative MR decreases and finally becomes positive, as shown for 3 and 6 K data in figure 2(b). The magnitude of positive MR increases with decreasing temperature and it varies almost linearly above 1 T magnetic field. The positive MR in metallic ferromagnets generally arises at low field due to anisotropic magnetoresistance or domain rearrangement. However, the magnitude of such a contribution remains very small. The positive MR can also arise due to the Lorentz force effect as observed in a metallic system. This positive contribution to MR dominates at low temperature and high magnetic field, where $\omega_{\rm c}\tau$ becomes large ($\omega_{\rm c}$ is the cyclotron frequency and τ is the mean free path of the charge carriers). In this case the positive MR is generally found to be of the order of a few per cent in high purity metals, whereas it is more than 20% at 3 K in DyPd₂Si. To differentiate the Lorentz force contribution and magnetic contribution to MR, we measured the MR in nonmagnetic LuPd₂Si, which is shown in figure 2(c). It shows positive MR at low temperature and its magnitude is comparable to DyPd₂Si. The magnitude of MR correlates with the resistivity of the sample, which is explicitly demonstrated in the inset of figure 2(c). Here MR is plotted as a function of $H/\rho(T)$ (Kohler's plot where $H/\rho(T) \propto \omega_c \tau$) and shows that all the isothermal MR curves merge almost to a single curve in this plot. It shows that the observed positive MR arises from a single scattering mechanism, which can be attributed to the orbital effect (the Lorentz force effect) in this case. Therefore, MR at low temperature in magnetic compounds has two contributions: (i) a positive contribution arising from the Lorentz force effect and (ii) a negative contribution due to suppression of magnetic scattering. At low temperature and high magnetic field the Lorentz force effect dominates, whereas magnetic scattering becomes negligible. This shows that the Fermi surface is highly anisotropic in these compounds and also explains the positive slope in TbPd₂Si at low temperature and high field. Since the residual resistivity of DyPd₂Si is almost equal to LuPd₂Si, the orbital contribution to MR in DyPd₂Si can be considered to be equal to that of LuPd₂Si. When we separate this contribution from the MR of $DyPd_2Si$, as shown in the inset of figure 2(b) for 3 and 6 K data, the positive contribution to MR almost vanishes in the entire field range. This is expected from a MCE study on this compound [3, 6].

In the case of Tb compounds anomalous MR behavior is observed below the 9 K transition. The transition is accompanied by thermal hysteresis. At 3 K (see figure 3), the resistivity before and after the application of magnetic field is different in this compound. After field cycling, the



Figure 3. Magnetic field dependence of magnetoresistance for the compound TbPd₂Si at 3 K. Solid circles represent the $0 \rightarrow 8$ T (virgin) curve, open triangles represent the +8 T $\rightarrow -8$ T curve and solid triangles represent the -8 T $\rightarrow +8$ T curve. The large difference between zero field resistivity before and after the application of magnetic field is highlighted in the right inset. The left inset shows irreversibility and metamagnetic transitions at low field in the magnetic field dependence of the magnetization.

resistivity is about 16% higher than its initial value at zero field. Some systems do show an open hysteresis loop where the high field magnetic state persists down to zero field at low temperature [8-11]. However, in the present case the high field state is of low resistance compared to the initial zero field state, whereas after field cycling the final zero field state has higher resistance. Thus the possibility of the presence of the high field state at zero field is ruled out. It appears that the free energy of this system has several minima of comparable energy corresponding to different magnetic states. Depending on the path followed in H-T space the system can be trapped in different magnetic states. Therefore, the zero field magnetic state after magnetic field cycling could be different from its initial state. M versus H data shown in the left inset of the same figure also suggest different magnetic states at low field after field cycling. It shows almost negligible M at low field values and metamagnetic transition with increasing magnetic field (virgin curve). During the field decreasing cycle it shows irreversibility at low field values persisting down to zero field. The difference between zero field resistivity before and after magnetic field cycling decreases with increasing temperature. This may be attributed to increased thermal fluctuations that can transform the system into its ground state configuration within the measurement timescale. This has been observed in MR measurements at various temperatures. However, to verify these speculations detailed MR measurements are required.

4. Conclusions

To conclude, we have observed large magnetoresistance in TbPd₂Si and DyPd₂Si compounds. The magnetoresistance at low temperature is dominated by magnetic scattering along with the Lorentz force effect. This suggests a complex Fermi surface in these compounds. Due to the Lorentz force effect MR behavior does not correlate with MCE even in ferromagnetic DyPd₂Si. These studies also show anomalous thermomagnetic irreversibilities and metamagnetic transition in the Tb compound.

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