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Transport properties in the $URu_{1-x}Pd_xGa$ system

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Abstract. The effect of substitutions in the $URu_{1-x}Pd_xGa$ system has been investigated by means of electrical resistivity and magnetoresistivity measurements. The substitutions of Pd for Ru have been found to (i) suppress a Kondo behaviour at high temperatures, (ii) produce magnetic order and (iii) induce a Kondo-like minimum in the electrical resistivity at low temperatures. We analysed the low-temperature part of the electrical resistivity by considering electron scattering by impurities, by spin-wave excitations and originating from a Kondo effect. Samples with $0 \leqslant x \leqslant 0.3$ exhibit incoherent Kondo behaviour above about 100 K, while for the $0.4 \leqslant x \leqslant 0.8$ compositions a broad maximum appears in the resistivity around 150–180 K, presumably due to a combination of the Kondo and crystal-field effects. The magnetoresistivity results support the earlier proposal derived from the magnetic studies of the $URu_{1-x}Pd_xGa$ system in which a magnetic phase transition from paramagnetic to ferromagnetic and finally to an antiferromagnetic/ferrimagnetic state takes place. A marked difference was found in the magnetoresistivity data for the $x = 0.2$, 0.6 and 0.8 samples, reflecting a variety of magnetic structures which appear in these compounds.

1. Introduction

Recently, we investigated solid solutions of $URu_{1-x}Pd_xGa$ by means of x-ray powder diffraction, dc magnetic susceptibility and magnetization measurements [1]. All the investigated compounds, including the end-members of the series URuGa and UPdGa which were studied earlier [2–6], crystallize in the hexagonal Fe_2P-type structure (space group $P\bar{6}2m$). For the sake of clarity the magnetic phase diagram obtained in [1] for the $URu_{1-x}Pd_xGa$ compounds is reproduced in figure 1. As seen, the substitution of Ru by Pd changes the nonmagnetic behaviour of URuGa to a magnetic ordered state for $x \geqslant 0.1$. This state is probably ferromagnetic and with increasing Pd content its Curie temperature T_C increases, going through a maximum at 78 K near $x = 0.5$, and then falls to a value of 55 K at $x = 0.8$. For $x \geqslant 0.9$ an antiferromagnetic state AF1 sets in for values of T_N indicated in figure 1, but at still lower temperatures another transition is believed to take place to an antiferromagnetic state denoted as AF2. Some evidence for a ferrimagnetic state has also been indicated in the vicinity of $x = 0.8$.

The main aim of the present investigation is to study the electrical resistivity for samples with x ranging between 0.1 and 0.8. In this concentration range, the T_C–x dependence (see figure 1) resembles the well known competition between the Kondo and RKKY interactions in the system. Hence, we discuss the experimental results in terms of Kondo behaviour. The electrical resistivity data indicate that the Kondo effect exists in the investigated

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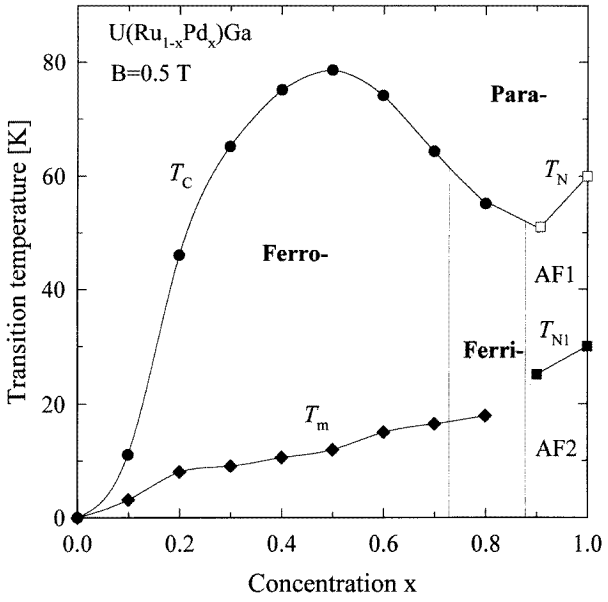

Figure 1. Magnetic phase diagram for the $URu_{1-x}Pd_xGa$ system as determined by magnetization measurements [1].

compositions and it is shown that it is an essential ingredient in understanding the magnetic behaviour of the $URu_{1-x}Pd_xGa$ system. It is also found that the magnetoresistivity data correlate well with the magnetization data obtained in our previous work [1], thus helping to clarify the nature of the magnetic ground state in the studied compositions.

2. Experimental details

The samples were synthesized by arc melting stoichiometric quantities of the constituents of the following purities: U and Ga, 99.9 wt%; Ru and Pd, 99.9999 wt%. Mass losses after melting of less than 0.1% were recorded. Samples were wrapped in tantalum foil, sealed in a quartz tube under vacuum and annealed for two weeks at 650 °C. The samples used in the present study were cut from the same ingots as the samples used for the magnetic susceptibility and magnetization measurements [1].

The electrical resistivity was measured in the temperature range of 1.5–287 K using a standard dc four-probe measuring technique. Resistivity data were collected during both cooling and heating of the samples. Longitudinal magnetoresistivity data, $\Delta\rho/\rho = \{\rho(B,T)-\rho(0,T)\}/\rho(0,T)$, were recorded in two different ways: isotherms were recorded at selected temperatures below 120 K in magnetic fields up to $B = 8$ T on zero-field cooled samples (ZFC) and isofield curves were recorded during cooling from 100 to 1.5 K at a fixed field of 8 T (FC). The samples had typical dimensions $0.6 \times 0.6 \times 5.0$ mm^3 and the uncertainty in the geometrical factor determination was of the order of 5%. A constant-current source supplied 100 mA to the specimens and the sensitivity in voltage detection was 5 μV. In the magnetoresistivity experiments, the sample temperatures were held constant to within 5 mK during isothermal measurements and the magnetic field fixed to within 2 mT during isofield measurements.

3. Results

3.1. Electrical resistivity

Some transport characteristics of the alloys studied in this work are summarized in table 1. One observes that the resistivity values at room temperature of the solid solutions of $URu_{1-x}Pd_xGa$ are appreciably lower than those of the parent compounds URuGa [4] and UPdGa [5]. Such a behaviour seems surprising since additional atomic disorder scattering should be associated with the solid solutions and this should enlarge the resistivity. One reason for the initial decrease in resistivity on Pd substitution on Ru sites may be that the resistivity of URuGa at high temperatures is dominated by incoherent Kondo scattering in the uranium sublattice. The disordered alloys ($x = 0.1$ and 0.2) suppress this type of scattering, thus leading to lower resistivity values.

Table 1. Some transport characteristics of samples of the $URu_{1-x}Pd_xGa$ system with $x = 0.1$–0.8. T_C indicates the magnetic ordering temperature determined from both magnetization and resistivity measurements. T_{min} and T_{max} are characteristic temperatures, where the resistivity exhibits a minimum or a maximum, respectively. A, C and Δ are the parameters obtained by fitting the resistivity data in the temperature range 1.5–25 K using equations (1)–(3). $\rho(287)$ and $\rho(1.5)$ denote the values of resistivity at room temperature and at 1.5 K. The data for URuGa and UPdGa are taken from [4] and [5].

x	T_C^a (K)	T_C^b (K)	T_{min} (K)	T_{max} (K)	A ($\mu\Omega$ cm K^2)	C (K)	Δ (K)	$\rho(287)$ ($\mu\Omega$ cm)	$\frac{\rho(1.5)}{\rho(287)}$
0 [4]								798	1.12
0.1	11.0	9.5						398	1.23
0.2	46.0	46.5	12	48	0.03	−0.63	33	226	1.00
0.3	65.0	65.7	7	71	0.06	−1.14	29	315	0.81
0.4	74.8	68.4	6	150	0.03	−1.07	6	374	0.78
0.5	78.4	77.5		153	0.06		6	333	0.75
0.6	74.2	67.5		158	0.04		37	292	0.91
0.7	64.3	65.2		145	0.04			205	0.73
0.8	55.0	57.9		226	0.02		10	222	0.87
1.0 [5]								1040	0.71[c]

[a] Curie temperature, T_C, determined from magnetization measurements [1].
[b] Curie temperature, T_C, determined from resistivity measurements.
[c] $\rho(4.2)/\rho(287)$.

The temperature dependences of the electrical resistivity, $\rho(T)$, of alloys with $x = 0.1$, 0.2 and 0.3 are presented in figure 2. For the sake of comparison, we also include in this figure the results of $\rho(T)$ for URuGa [4]. As the temperature is lowered from room temperature down to approximately 50 K, a Kondo-like increase of resistivity is apparent for all the compositions. Here, the Kondo impurity description is qualitatively valid since the experimental data obey a Kondo temperature dependence [7]

$$\rho_K(T) = C \ln T \tag{1}$$

with $C = -18.5$, -24.5, -9.8 and -9.5 $\mu\Omega$ cm K^{-1} for $x = 0.0$, 0.1, 0.2 and 0.3, respectively (see the solid lines in figure 2).

Previously, we have shown that ρ of URuGa increases upon cooling as $C \ln T$ and below 7 K follows an activated form $\rho(T) \approx \exp(E_a/2k_BT)$ with $E_a = 1.9$ meV [4]. This result is in disagreement with that obtained earlier by Havela *et al* [3], who reported valence fluctuation behaviour for this compound. Possibly the samples measured by ourselves [4]

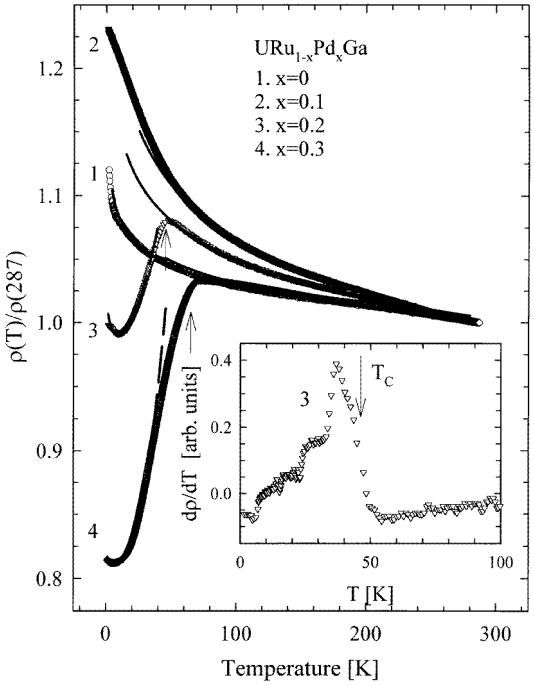

Figure 2. Normalized electrical resistivity, $\rho(T)/\rho(287\ \mathrm{K})$, versus temperature for $URu_{1-x}Pd_xGa$ samples with $0.1 \leqslant x \leqslant 0.3$. The arrows indicate the magnetic ordering temperature, T_C. The temperature dependence of the resistivity derivative, $d\rho(T)/dT$, shown in the inset is used to determine the value of T_C as described in the text. The solid and dashed lines are different fits to the experimental data. The data of URuGa are taken from [4].

and by Havela *et al* [3] exhibit some small differences in purity and stoichiometry, which cause a shift in the magnetic and transport properties from a Kondo-like behaviour (our data [4]) towards fluctuating valence character (results of Havela *et al* [3]).

The resistivity of the $x = 0.1$ sample starts to deviate from the Kondo behaviour at $T = 75$ K, increasing faster at lower temperatures (figure 2). We do not observe any distinct anomaly near $T_C = 11$ K, indicated previously by the magnetic measurements [1], but there is a slight change in slope of $\rho(T)$ near this temperature. Clear evidence of a magnetic transition is found in the $x = 0.2$ sample. At $T \approx 50$ K the resistivity of this sample reaches its maximum and then decreases due to establishment of long-range magnetic order as the temperature is lowered. The behaviour for the $x = 0.3$ sample is similar to that of the $x = 0.2$ one with the appearance of a maximum in $\rho(T)$.

Measurements of $\rho(T)$ near T_C may in principle be used to obtain the value of T_C and of the critical exponents and amplitudes that describe the magnetic critical behaviour near the phase transition [8]. Close enough to T_C, a $t^{-\alpha}$ Fischer–Langer [9] relationship is expected for ferromagnetic metals, with $t = |T - T_C|$ and α the critical exponent that describes the specific heat anomaly near T_C. This result is obtained by assuming the dominance of short-ranged fluctuations near T_C. One then expects experimentally a monotonic increasing resistivity with temperature through T_C and which is characterized by a divergence in $d\rho(T)/dT$ at T_C. The de Gennes–Friedel description in which long-range fluctuations dominate near T_C on the other hand leads to an upward pointing cusp in $\rho(T)$

at T_C [10]. While many ferromagnetic metals experimentally exhibit the Fischer–Langer behaviour, $\rho(T)$ and $d\rho(T)/dT$ of some materials behave in a more complex manner. Thus evidence of the existence of a cusp in $\rho(T)$ near T_C is found for the rare-earth Laves phase compounds $GdNi_2$ and $GdPt_2$ [11, 12]. Furthermore T_C values deduced from maxima in $d\rho(T)/dT$ for such GdT_2 compounds (T = Ni, Rh, Pt, Co, Al) were in all cases found to be consistently lower than those observed from magnetization data [12]. Our $\rho(T)$ curves for $x = 0.2$–0.8 exhibit the effect of long-range magnetic order below T_C, while above T_C compounds with $x \leqslant 0.3$ are characterized by a Kondo-like resistivity. The superposition of these contributions and of the electron–phonon interaction (of unknown magnitude) evidently yields $\rho(T)$ curves that exhibit a maximum near T_C for $x = 0.2$ and 0.3. The peak in $d\rho(T)/dT$ shown in the inset to figure 2 for the $x = 0.2$ sample does not agree well with a $t^{-\alpha}$ description and furthermore maxima in $d\rho(T)/dT$ for all our samples were consistently observed at lower temperatures than values of T_C previously found from our magnetization studies [1]. As an operational procedure and motivated by the preceding considerations we obtain values for T_C for our samples from the temperature at which the resistivity derivative shows an inflection point. Fair agreement between these values of T_C and those determined between from magnetization measurements has been found (table 1).

An interesting feature of the $\rho(T)$ curves at low temperatures is the occurrence of a resistivity minimum at $T_{min} \approx 12$ K and 7 K, for $x = 0.2$ and 0.3, respectively. This unusual feature has previously been observed, e.g. for the U-substituted alloys (U, M)Ru_2Si_2, where M is a nonmagnetic metal such as Th [13], Y or Sc [14]. The authors of [13] have explained the observed behaviour as being an impurity Kondo-hole effect, a concept that has already been investigated theoretically by Sollie and Schlottmann [15]. On the other hand, Cornut and Coqblin [16] have developed a model for Ce–intermetallic Kondo systems in which the effect of the crystalline electric field (CEF) is explicitly considered. In their model a peak in $\rho(T)$ is obtained at a temperature corresponding to the overall splitting temperature, Δ_{CF}, expressed in K. At both higher or lower temperatures compared to Δ_{CF}, ρ exhibits a logarithmic temperature dependence. The low-temperature logarithmic behaviour arises due to the Kondo scattering from the crystal-field ground state only, while the high-temperature one corresponds to the Kondo scattering from the excited crystal-field levels. Thus, in terms of this theory the transport properties of a number of Ce- or Yb-based compounds, such as $CeCu_2Si_2$ [17] and $YbAg_4Au$ [18], have successfully been interpreted. If the mechanism which is responsible for the minimum in $\rho(T)$ at low temperatures is due to the Kondo-hole effect, it should lead to a logarithmic temperature dependence of $\rho(T)$. Assuming the phonon scattering contribution to be negligible at low temperatures, we analysed the temperature dependence of the resistivity as the sum of the following terms:

$$\rho(T) = \rho_0 + \rho_K(T) + \rho_m(T). \tag{2}$$

The first term is the temperature independent residual resistivity, $\rho_K(T)$ denotes the Kondo-like contribution, as discussed above, and the last term $\rho_m(T)$ represents the scattering of conduction electrons by spin-wave excitations. With the assumption [19] that

$$\rho_m(T) = AT^2 \exp(-\Delta/T) \tag{3}$$

we obtained the best fit to the data in the temperature range 1.5 K $\leqslant T \leqslant$ 25 K (see the broken line for $URu_{0.8}Pd_{0.2}Ga$ and $URu_{0.7}Pd_{0.3}Ga$ in figure 2). In equation (3), Δ is the gap in the magnon spectrum. It is noted that whereas the resistivity curves in figures 2–4 for different compositions were all normalized to the values of ρ at 287 K for ease of representation and comparison, the fit parameters in table 1 correspond to the measured values of resistivity (in $\mu\Omega$ cm) for each sample.

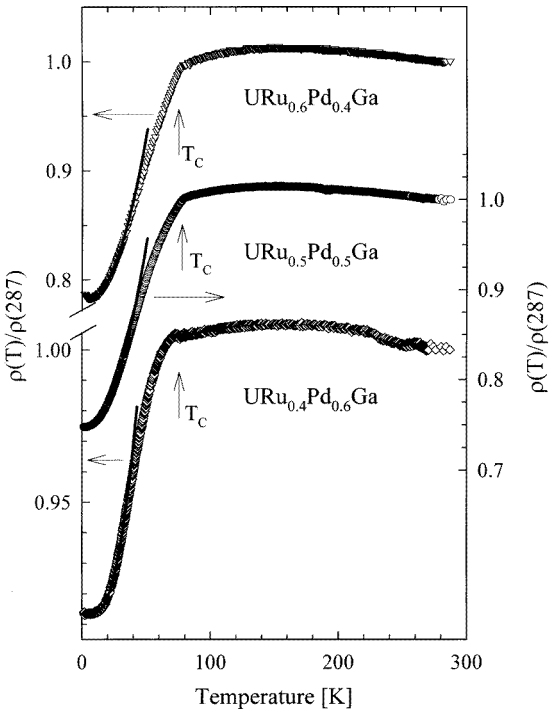

Figure 3. Normalized electrical resistivity, $\rho(T)/\rho(287\ \mathrm{K})$, versus temperature for $URu_{0.6}Pd_{0.4}Ga$, $URu_{0.5}Pd_{0.5}Ga$ and $URu_{0.4}Pd_{0.6}Ga$. The magnetic ordering temperature T_C is indicated by arrows. The solid lines are fits to the experimental data.

For the alloys with $x = 0.4$, 0.5 and 0.6, we have found a broad maximum of the resistivity at $T_{max} \approx 150$ K (figure 3). This maximum is not very sensitive to changes in the number of conduction electrons in the system. In contrast to the $x = 0.2$ and 0.3 samples, we believe that for the alloys in figure 3 the maximum does not relate to magnetic ordering effects, but rather arises due to a crystal-field effect. According to Cornut and Coqblin [16], T_{max} is approximately equal to the value of the overall crystal field splitting Δ_{CF}.

Upon cooling the resistivity of the $x = 0.4$, 0.5 and 0.6 samples shows a sharp knee characteristic of a magnetic phase transition. At temperatures well below T_C, i.e. $T < 0.5\ T_C$, we found in all three alloys a simple quadratic temperature dependence of the resistivity. However, at $T < 25$ K, one again observes the dependence expressed by equation (3). In figure 3, only the $x = 0.4$ sample shows a low-temperature Kondo-like resistivity minimum like that of $x = 0.2$ or $x = 0.3$. Thus, the dependence $\rho_K(T) = C \ln T$ can be applied for this composition as well. It is clear that for $x > 0.4$, the minimum in $\rho(T)$ indicative of the Kondo scattering becomes less distinct (see also figure 4 for $x = 0.7$ and 0.8).

The inset of figure 4 displays the high-temperature behaviour of the resistivity of the $x = 0.7$ and 0.8 alloys. It is interesting to note that the resistivity of the $x = 0.7$ alloy also shows a broad maximum at 150 K, as for the compositions $x = 0.4$, 0.5 and 0.6, whereas in the case of $x = 0.8$ this maximum is shifted to a higher temperature, i.e. to about 220 K. For UPdGa, on the other hand, the appearance of a minimum at 150 K in $\rho(T)$ is observed [5].

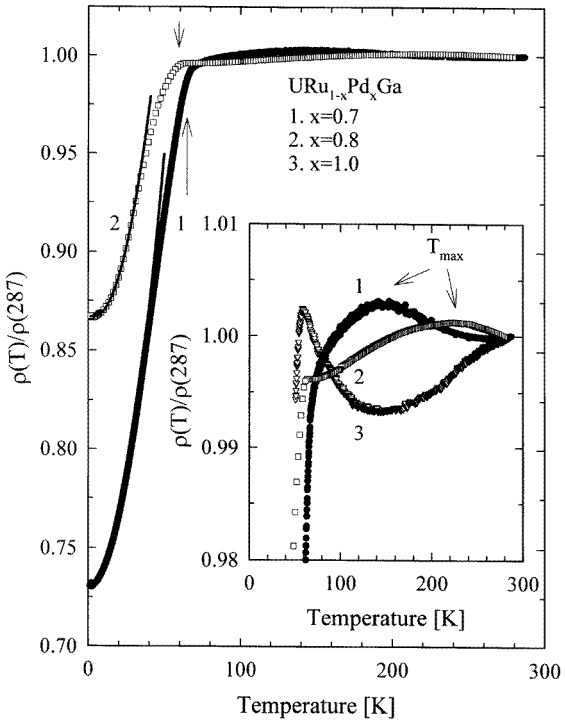

Figure 4. Normalized electrical resistivity, $\rho(T)/\rho(287\ \mathrm{K})$, versus temperature for $URu_{1-x}Pd_xGa$ with $x = 0.7$ and 0.8. The arrows indicate the magnetic ordering temperatures, T_C. The solid lines are fits to the experimental data. The inset shows the high-temperature part of the resistivity of these compositions. The data of UPdGa are taken from [5].

Evidence of magnetic phase transitions is also found for compositions $x = 0.7$ and 0.8 (figure 4). At low temperatures the resistivity of the former is proportional to T^2, while that of the latter is described by equation (3).

The values of T_C of all investigated compositions determined from the temperature derivatives are given in table 1 and compared with those determined from magnetic studies [1]. The present results confirm our earlier observation that the ordering temperature of the samples first increases with increasing x and then goes through its maximum at $x = 0.5$. However, the transition at T_m, observed in the course of the magnetic measurements [1], has not been visible in the $\rho(T)$ curves of all the investigated compounds.

3.2. Magnetoresistivity

In figure 5 we compare the temperature dependences of the electrical resistivity measured in a magnetic field $B = 8$ T with that taken in $B = 0$ for the $x = 0.2$, 0.6 and 0.8 compositions of the $URu_{1-x}Pd_xGa$ system. The field was applied along the length of the samples, i.e. in the direction of the current. It is indicated in this figure that an application of the magnetic field leads to a considerable reduction of the resistivity. At temperatures below T_C, the negative magnetoresistivity is related to a ferromagnetic ordering of the samples. The dominant scattering mechanism, i.e. the scattering of conduction electrons by spin-wave excitations, still occurs, and equation (3) is followed at low temperatures. The magnetic

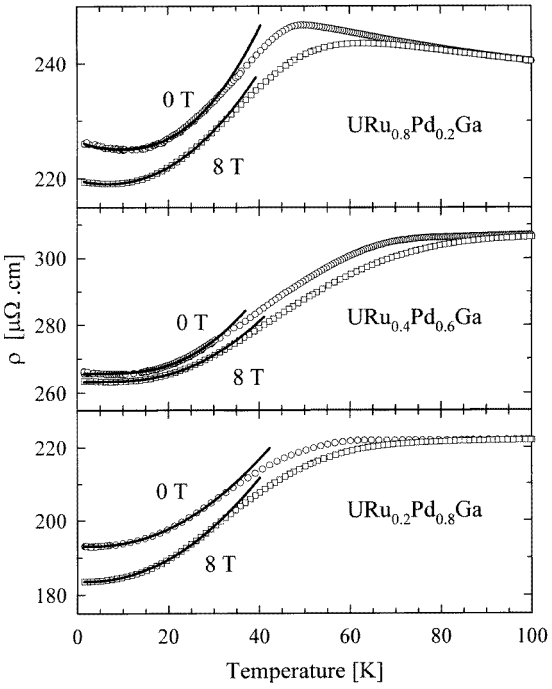

Figure 5. The effect of a magnetic field on the temperature dependence of the electrical resistivity, for the alloys $URu_{0.8}Pd_{0.2}Ga$, $URu_{0.4}Pd_{0.6}Ga$ and $URu_{0.2}Pd_{0.8}Ga$, is shown in comparison to the zero-field data. The field is applied parallel to the direction of current flow.

fields have only an influence on the value of the gap Δ: it becomes somewhat smaller than the zero-field value. These changes are from 33, 37 and 10 K to 17, 31 and 7 K at $B = 8$ T, for $x = 0.2$, 0.6 and 0.8, respectively. Therefore, these results demonstrate that the energy gap in the magnon spectrum can be altered by applying the magnetic field. This finding also explains the decrease in the resistivity value at the ordering temperature, $\rho(T = T_C)$, when a magnetic field is applied on a given sample, because the magnitude of the gap is closely related to the $\rho(T = T_C)$ value. The magnetoresistivity is also negative in the paramagnetic state and this might be discussed in terms of short-range ferromagnetic correlations between uranium atoms in the presence of the magnetic field. However, we have already shown in section 3.1 that the uranium atoms in these compounds above T_C qualitatively exhibit Kondo behaviour. For that reason, the negative magnetoresistivity in the paramagnetic state is rather described by incoherent Kondo scattering, as predicted theoretically by the authors of [20] and [21].

The temperature dependences of magnetoresistivity, henceforth indicated as $\Delta\rho(T)/\rho$, are plotted for the $x = 0.2$, 0.6 and 0.8 samples in figure 6. The results pertain to measurements at 8 T and we note differences between results obtained from isothermal measurements on ZFC samples (closed symbols) and isofield runs on FC samples (open symbols). These differences are especially pronounced for $T < 20$ K for the $x = 0.8$ sample. Above 20 K the results obtained from the two types of measurement are in fair agreement.

Considering the isofield measurements (FC samples) it is observed that there are qualitative differences in the data for the three compositions, reflecting a variety in their

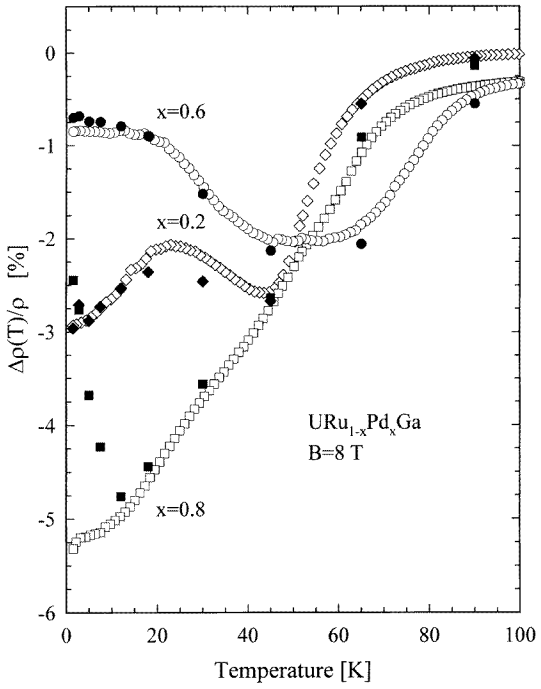

Figure 6. Longitudinal magnetoresistivity, $\Delta\rho(T)/\rho$, as a function of temperature in the alloys $URu_{0.8}Pd_{0.2}Ga$, $URu_{0.4}Pd_{0.6}Ga$ and $URu_{0.2}Pd_{0.8}Ga$. The open points are the FC data while the solid points are the ZFC ones.

magnetic behaviour. In the case of $x = 0.2$, the temperature dependence of $\Delta\rho(T)/\rho$ shows initially an increase, goes through a maximum at about 25 K and then through a minimum at about 45 K. The peak behaviour at the former temperature may be associated with a spin reorientation; from a complex antiferromagnetic/ferrimagnetic state at low temperatures to a ferromagnetic one at higher temperatures [1]. The latter state has a phase transition to the paramagnetic state at about 45 K, where the minimum in the $\Delta\rho(T)/\rho$ curve for this sample is observed. This is due to the fact that the critical fluctuations are significantly suppressed by an applied magnetic field at T_C.

The low-temperature magnetoresistivity of $URu_{0.4}Pd_{0.6}Ga$ is rather small. At 1.5 K and at 8 T, $\Delta\rho/\rho$ is only 0.7–0.8%. The most interesting feature is that $\Delta\rho(T)/\rho$ attains a broad minimum at about 50 K. The magnetic phase transition temperature T_C of this alloy was determined to be about 70 K (table 1) and, hence, the relation between this shallow minimum and the magnetic ordering at T_C is not clear in this case. The insensitivity of the position of this minimum to temperature suggests that the magnetic moments in the temperature range of 35–70 K might not be able to align in some preferred direction. Such a behaviour may correspond to the situation in this temperature range of the existence of mixed antiferromagnetic and ferromagnetic states or a spin-glass state.

For $x = 0.8$, both the magnetic susceptibility and electrical resistivity measurements indicate a magnetic phase transition below $T_C = 55$ K. Figure 6 shows that the application of a magnetic field of 8 T smears out the anomaly at T_C. Below 15 K, $\Delta\rho(T)/\rho$ exhibits a large difference between ZFC and FC cycles. Such a hysteretic behaviour has been found in the isostructural compound UPdIn [22], for which ferrimagnetic ordering with a squared-up modulation along the c-axis $(++-+-)$ was observed below 8.5 K [23].

The field dependences of the magnetoresistivity, indicated as $\Delta\rho(B)/\rho$, are plotted in figures 7, 8 and 9 for various isotherms of the ZFC $URu_{1-x}Pd_xGa$ samples ($x = 0.2$, 0.6 and 0.8, respectively). It is noted that for all isotherms depicted in figures 7–9 the particular sample was heated to 100 K and then cooled to the required measuring temperature in order to establish a demagnetized reference state for $B = 0$, before the $\Delta\rho(B)/\rho$ curve was taken. Open symbols refer to increasing field sweeps and closed symbols to the subsequent decreasing field sweeps.

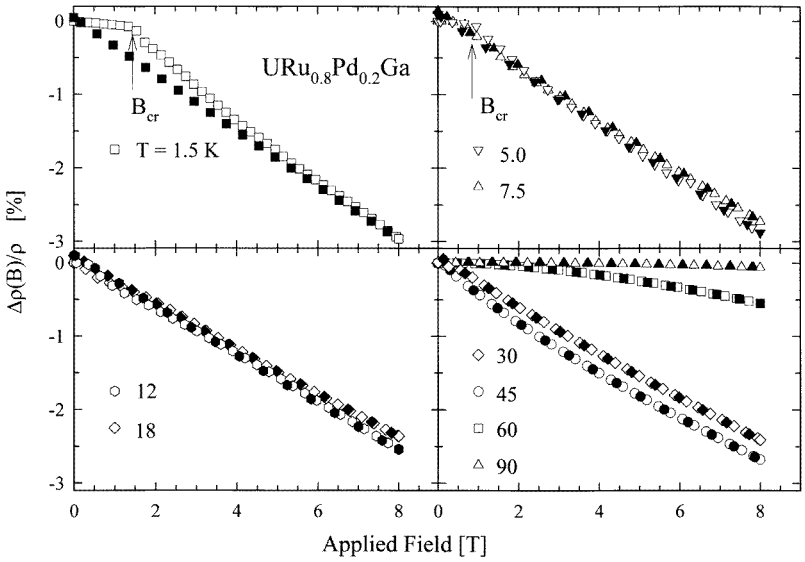

Figure 7. Longitudinal magnetoresistivity, $\Delta\rho(B)/\rho$, of $URu_{0.8}Pd_{0.2}Ga$ as a function of magnetic field. Each isotherm was taken after following a demagnetization procedure as described in the text. Open symbols refer to increasing field sweeps and the closed symbols refer to decreasing field sweeps.

Characteristic features for $\Delta\rho(B)/\rho$ for the $URu_{0.8}Pd_{0.2}Ga$ composition in figure 7 are considered in the temperature regimes $T \leqslant 7.5$ K, 12 K $\leqslant T \leqslant 45$ K and for higher-temperature isotherms of 65 and 90 K. For the isotherms with $T \leqslant 7.5$ K, $\Delta\rho(B)/\rho$ depends linearly on B up to a critical field, B_{cr}, above which this function decreases more rapidly. The small negative change of $\Delta\rho(B)/\rho$ in low fields is a signature of an antiferromagnetic state as has been reported for UCu_2Ge_2 [24] and $PrCu_{0.5}Ni_{1.5}Si_2$ [25]. Therefore, B_{cr} could be considered as the field at which a metamagnetic transition occurs. The value of B_{cr} decreases with increasing temperature from 1.5 T at 1.5 K to 0.7 T at 7.5 K. Above B_{cr}, $\Delta\rho(B)/\rho$ is found to be proportional to B^n with $1 < n < 2$, while in higher fields it is again nearly linear in B but with a considerably larger slope than that for $B < B_{cr}$. This behaviour arises because any fluctuations are always suppressed by the field. Yamada and Takada [26] and Ueda [27] have calculated the magnetoresistivity by considering the electron-spin scattering in magnetic materials. The authors indicate that $\Delta\rho(B)/\rho$ should be negative and in small magnetic fields linear in B. In the temperature regime 12 K $\leqslant T \leqslant 45$ K, the $\Delta\rho(B)/\rho$ curves do not show evidence of a metamagnetic transition (figure 7). This seems to indicate that the antiferromagnetic state probably exists only below 12 K. It is observed for the 12 K and 18 K isotherms that $\Delta\rho(B)/\rho$ varies as B^n with $1 < n < 2$ in low fields. According to the theory reported in [28], such a power-law behaviour of $\Delta\rho(B)/\rho$

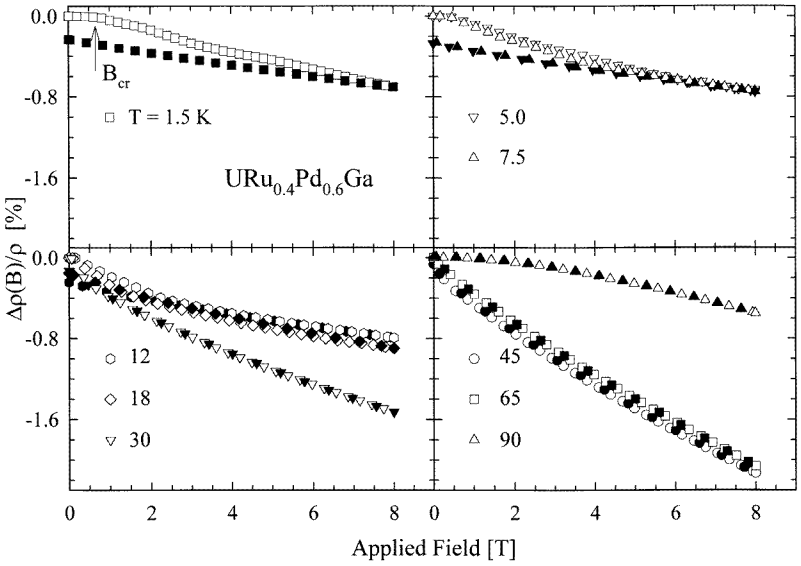

Figure 8. Longitudinal magnetoresistivity, $\Delta\rho(B)/\rho$, of $URu_{0.4}Pd_{0.6}Ga$ as a function of magnetic field. Each isotherm was taken after following a demagnetization procedure as described in the text. Open symbols refer to increasing field sweeps and the closed symbols refer to decreasing field sweeps.

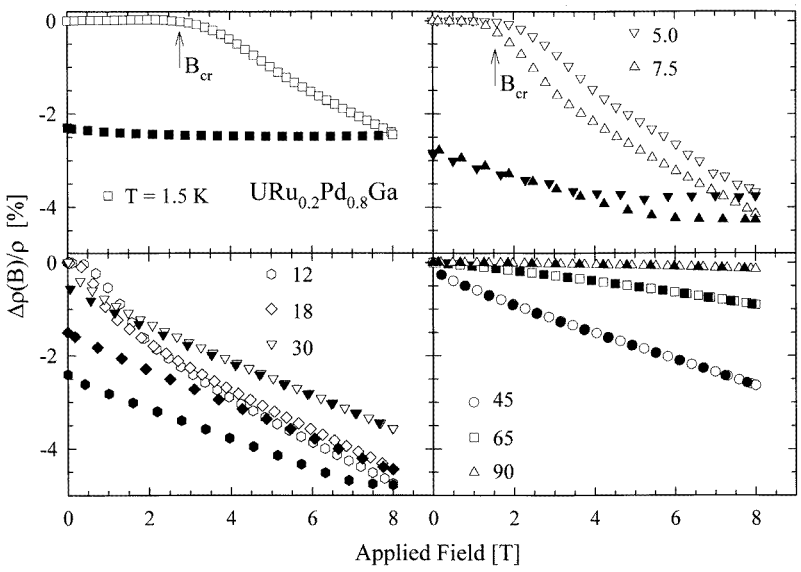

Figure 9. Longitudinal magnetoresistivity, $\Delta\rho(B)/\rho$, of $URu_{0.2}Pd_{0.8}Ga$ as a function of magnetic field. Each isotherm was taken after following a demagnetization procedure as described in the text. Open symbols refer to increasing field sweeps and the closed symbols refer to decreasing field sweeps.

is expected for random magnetic systems, where the coexistence of both ferromagnetic and antiferromagnetic states may take place. No hysteresis is observed for the $T = 45$ K

isotherm. For the $T = 65$ and 90 K isotherms, i.e. in the paramagnetic state, a negative magnetoresistivity has been observed which has a nearly quadratic dependence on B. This dependence is typical for an incoherent Kondo system [20, 21].

The above observations lead us to the conclusion that in the Kondo compound $URu_{0.8}Pd_{0.2}Ga$ a ferromagnetic order appears at $T_C \approx 45$ K and it persists down to about 18 K. Between 12 and 18 K, the ferromagnetic and antiferromagnetic components probably coexist, while below 12 K the ground state is probably antiferromagnetic.

For $URu_{0.4}Pd_{0.6}Ga$ (figure 8), $\Delta\rho(B)/\rho$ taken at 1.5 K decreases with increasing field and goes through an inflection point at about 1 T which is associated with a metamagnetic transition. In decreasing fields, $\Delta\rho(B)/\rho$ shows a large hysteresis. This hysteretic behaviour is observed in this alloy up to temperatures of 18 K.

The behaviour of $\Delta\rho(B)/\rho$ of $URu_{0.4}Pd_{0.6}Ga$ in the temperature range of 30–90 K is qualitatively like that of $URu_{0.8}Pd_{0.2}Ga$ taken in the range 45–90 K, except that the ordering temperature T_C is found to be higher for the former compound. We found also that the value of $\Delta\rho(B)/\rho$ at 90 K for this compound has the same magnitude as that of $URu_{0.8}Pd_{0.2}Ga$ measured at 65 K, suggesting that the Kondo effect in $URu_{0.4}Pd_{0.6}Ga$ is much stronger.

Figure 9 shows the field dependence of the magnetoresistivity for $URu_{0.2}Pd_{0.8}Ga$. The curves taken at 65 and 90 K correspond to the paramagnetic state of the compound. The data collected at 45 K indicate the existence of a ferromagnetic state at this temperature. Below 30 K, the $\Delta\rho(B)/\rho$ curves exhibit a strong hysteretic effect with a large remanence, as in the case of the magnetization curves (see reference [1]). Even as the magnetic field is decreased to zero, there remains a large value of the magnetoresistivity. Compared with the data of $x = 0.2$ and 0.6, one can see that the metamagnetic transition in $\Delta\rho(B)/\rho$ occurs here at higher values of both the magnetic field and temperature. This observation suggests that the antiferromagnetic state for $x = 0.8$ is more stable. Moreover, it is clear that the temperature below which $\Delta\rho(B)/\rho$ shows both hysteresis and a metamagnetic-like transition is Pd-concentration dependent.

4. Conclusions

The electrical resistivity and magnetoresistivity results on the $URu_{1-x}Pd_xGa$ system with $x = 0.1$–0.8 are summarized as follows.

(i) The $\rho(T)$ curve of each of the investigated compounds shows an anomaly at the magnetic ordering temperature, T_C, the values of which agree with those determined from magnetic measurements. We have not observed any anomaly in $\rho(T)$ caused by the lower-temperature transition at T_m.

(ii) The low-temperature behaviour of $\rho(T)$ is governed mainly by electron scattering on spin-wave excitations and characterized by a gap in the magnon spectrum. The value of this gap is decreased by an applied magnetic field. For $0.2 \leqslant x \leqslant 0.4$, we observed a minimum in the low-temperature resistivity, presumably due to a Kondo effect.

(iii) The $\rho(T)$ behaviour at high temperature may be discussed in terms of the Kondo effect for $x \leqslant 0.4$ and by the Kondo effect with accompanying crystal-field effects for the remaining compositions.

(iv) The magnetoresistivity for the $x = 0.2$, 0.6 and 0.8 samples was found to be negative in the paramagnetic state, confirming that the transport is governed by an incoherent Kondo effect.

(v) The magnetoresistivity measurements also reveal that the $URu_{1-x}Pd_xGa$ system ($0.1 < x < 0.8$) undergoes a transition to a ferromagnetic state at T_C.

(vi) The appearance of a metamagnetic-like transition and hysteresis in the $\Delta\rho/\rho$ curves at low temperatures may be attributed to the existence of an antiferromagnetic/ferrimagnetic state.

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References

[1] Tran V H, Troć R, Du Plessis P de V, Andre G and Bourée F 1997 *Phys. Rev.* B accepted for publication
[2] Sechovský V, Havela L, de Boer F R, Franse J J M, Veenhuizen P A, Sebek J, Stehno J and Andreev A V 1986 *Physica* B **142** 283
[3] Havela L, Hrebik J, Zeleny M and Andreev A V 1985 *Acta Phys. Pol.* A **68** 493
[4] Tran V H and Troć R 1997 *Acta Phys. Pol.* at press
[5] Tran V H and Troć R 1991 *J. Less-Common Met.* **175** 267
[6] Tran V H, Troć R, Bourée F, Roisnel T and André G 1995 *J. Magn. Magn. Mater.* **140–144** 1377
[7] Kondo J 1964 *Prog. Theor. Phys.* **32** 37
[8] Alexander S, Helman J S and Balberg I 1976 *Phys. Rev.* B **13** 304
[9] Fischer M E and Langer J S 1968 *Phys. Rev. Lett.* **20** 665
[10] de Gennes P G and Friedel J 1958 *J. Phys. Chem. Solids* **4** 71
[11] Kawatra M P, Skalski S, Mydosh J A and Budnick J I 1969 *Phys. Rev. Lett.* **23** 83
[12] Kawatra M P, Mydosh J A and Budnick J I 1970 *Phys. Rev.* B **2** 665
[13] Lopez de la Torre A, Visani P, Dalichaouch Y, Lee B W and Maple M B 1992 *Physica* B **179** 208
[14] Park J-G 1994 *J. Phys.: Condens. Matter* **6** 3403
[15] Sollie R and Schlottmann P 1991 *J. Appl. Phys.* **69** 5478
[16] Cornut B and Coqblin B 1972 *Phys. Rev.* B **5** 4541
[17] Lee W H and Shelton R N 1988 *Solid State Commun.* **68** 443
[18] Bauer E *et al* 1994 *Phys. Rev.* B **50** 9300
[19] Volkenshtein N V, Dyakina V P and Startsev V E 1973 *Phys. Status Solidi* b **57** 9
[20] Schlottmann P 1983 *Z. Phys.* B **51** 223
[21] Kawakami N and Okiji A 1986 *J. Phys. Soc. Japan* **55** 2114
[22] Nakotte H, Brück E, de Boer F R, Riemersma A J, Havela L and Sechovský V 1992 *Physica* B **179** 269
Sechovský V, Havela L, Nakotte H, Prokeš K, Brück E and de Boer F R 1995 *Physica* B **206/207** 501
[23] Fujii H, Kawanaka H, Takabatake T, Sugiura E, Sugiyama K and Date M 1990 *J. Magn. Magn. Mater.* **87** 235
Sugiura E, Sugiyama K, Kawanaka H, Takabatake T, Fujii H and Date M 1990 *J. Magn. Magn. Mater.* **90/91** 65
[24] Nigam A K, Roy S B and Girish Chandra 1994 *Phys. Rev.* B **49** 1127
[25] Das I and Sampathkumaran E V 1994 *Phys. Rev.* B **49** 3972
[26] Yamada H and Takada S 1973 *Prog. Theor. Phys.* **49** 1401
[27] Ueda K 1976 *Solid State Commun.* **19** 965
[28] Nigam A K and Majumdar A K 1983 *Phys. Rev.* B **27** 495, and references therein