Structural, magnetic and transport properties of the U_2TGa_3 compounds (T = Ru, Rh, Ir, Pd and Pt)

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Abstract. New ternary uranium intermetallics with the general formula U_2TGa_3 (T = Ru, Rh, Pd, Ir and Pt) have been prepared and found to crystallize in the orthorhombic $CeCu_2$-type structure. We report the results of magnetic susceptibility and electrical resistivity measurements for these compounds. It is found that U_2RuGa_3, U_2RhGa_3 and U_2IrGa_3 are ferromagnetic compounds with Curie temperatures of 73, 60 and 72 K, respectively. The compound U_2PdGa_3 is an antiferromagnet with a Néel temperature of 33 K, while U_2PtGa_3 probably undergoes two consecutive magnetic phase transitions at 80 and at 30 K to ferromagnetic and antiferromagnetic states. In all of the compounds investigated, the temperature dependence of the electrical resistivity shows features that could be ascribed to Kondo systems. The results are discussed and compared with those for the UTGa counterparts.

1. Introduction

The uranium equiatomic UTM intermetallics (T = d-electron transition metal and M = p-electron metalloid) exhibit unusual properties at low temperatures, for example, the Kondo effect [1], heavy-fermion behaviour [2], semiconductivity [3], and various magnetic ground states with complicated magnetic structures [4–7]. The variety of physical properties observed in these intermetallics is either due to a direct 5f–5f overlap and/or due to a hybridization between the 5f electrons and the spd electrons of the ligands (referred to hereafter as 5f–ligand hybridization). In order to obtain a better knowledge of the 5f–spd hybridization effect on the magnetic behaviour of the UTM series, a suitable substitution for T or M components in a given compound was used as a means of continuously varying the relevant parameters. A good example is provided by the studies of the $U(T'_{1-x}T''_x)M$ [8–11] or $UTM'_{1-x}M''_x$ [12–13] alloys (T' and T'' denote d-electron transition metals, and M' and M'' represent p-electron elements). Of course, we can also change the strength of 5f–spd hybridization by substituting M atoms for a proportion of the T atoms, hoping that their crystal structures will be unchanged or at least that the phase will have a derivative structure type. In this way, one obtains new series of compounds, broadening the possibilities of making a comparative study.

An example of such a substitution is the U_2TSi_3 compositions [14–15], since one regards these phases as derivatives of the UTSi series obtained by a replacement of half of the T atoms with Si atoms. It has been reported that the intermetallics U_2TSi_3, where T denotes 3d- or 5d-electron transition metals, crystallize mainly in the hexagonal AlB_2-type structure and show spin-glass behaviour or ferromagnetic properties [14–16].

We have prepared and characterized the new series of compounds U_2TGa_3. Among them only U_2CuGa_3 crystallizes in the hexagonal Lu_2CoGa_3-type form and does not show magnetic order at low temperatures. Magnetic data for this compound have been reported in [17]. Detailed studies on the compound U_2AuGa_3 are in progress and will be presented in a forthcoming paper. Our attempts to prepare the compounds containing T = Fe, Co and Ni were unsuccessful, since these samples were multi-phased.

In the following, we report structural, magnetic and electrical resistivity data for compositions with T = Ru, Rh, Pd, Ir, Pt. The observed properties are discussed and compared to those reported for the corresponding gallide 1:1:1 compounds [18–20].

2. Experimental details

Samples of U_2TGa_3 were synthesized by arc melting of the constituent elements of at least 99.9% purity in the stoichiometric ratio. The weight loss during melting was negligible. All materials were subsequently annealed in evacuated quartz tubes at 650 °C for one week. The samples were checked by x-ray diffraction analysis. In some compounds, the patterns show a few additional lines that correspond to about 5% of UGa_3 impurity. The presence of the antiferromagnet UGa_3, which has an ordering temperature $T_N = 70$ K [21], does not influence the observed behaviour of the compounds investigated (see below).

Magnetization studies were performed using a SQUID magnetometer (Quantum Design type MPMS-5) between 4.2 and 300 K in applied magnetic fields up to 5.5 T. Due to the irregular shapes of the specimens, a demagnetizing factor was not taken into account in data analysis. Measurement of the ac magnetic susceptibility was performed on U_2PtGa_3 using a SHE SHORE susceptometer at a frequency of 125 Hz and applying an ac field of 20 Oe. The electrical resistivity was measured by a dc four-probe technique in which the uncertainty in the geometrical factor was of the order of 5%.

3. Results

3.1. Crystal structure

Powder x-ray patterns of the U_2TGa_3 samples indicate that the diffraction peaks could be indexed in an orthorhombic system of the $CeCu_2$-type crystal structure (space group *Imma*). In the unit cell of this structure type the uranium is on the cerium sites (4e), and the transition metal and gallium atoms are arranged on the copper (8h) sites. The lattice parameters at room temperature for the U_2TGa_3 compounds are listed in table 1. One observes a decrease in the unit-cell volume when going from Ru- to Pd-based compounds. This effect suggests an increase in the hybridization strength between 5f and sp electrons only, because the 5f–d hybridization is rather decreased by increasing the number of 4d electrons.

From the crystallographic point of view, the existence of the U_2TGa_3 compounds, which have a structure different from that of the corresponding UTGa gallides, is interesting. When half of the T atoms in UTGa are replaced by Ga atoms, the number of p electrons increases while that of d electrons decreases. We believe that the change in concentration of valence electrons of a given compound may be the reason for the structural change from the Fe_2P type (for UTGa) to the $CeCu_2$-type (for U_2TGa_3). This finding is corroborated by the observed structural change for the UTM series, since the hexagonal Fe_2P-type structure exists for the UTM compounds with M = Al and Ga [20, 22] having an electronic configuration p^1 while the orthorhombic $CeCu_2$/TiNiSi-type structure appears for M = Si and Ge [23, 24], which have one p electron more than Al and Ga. Both of the crystal

Table 1. Crystallographic and magnetic characteristics of the U$_2$TGa$_3$ compounds, where T = Ru, Rh, Pd, Ir and Pt.

	U$_2$RuGa$_3$	U$_2$RhGa$_3$	U$_2$PdGa$_3$	U$_2$IrGa$_3$	U$_2$PtGa$_3$
a (Å)	4.652	4.385	4.372	4.343	4.381
b (Å)	7.037	6.964	7.017	6.927	7.033
c (Å)	7.886	7.742	7.691	7.694	7.703
Magnetic order	F	F	AF	F?/F	AF/F
T_C/T_N (K)	73	60	33	25/72	30/80
μ_s (μ_B/U atom)	0.9	0.4	—	0.6	—
μ_{eff} (μ_B/U atom)	2.12	2.20	2.58	1.85	2.14
Θ_p (K)	73	61	8	72	45
χ_0 (10^{-3} emu/U atom)	1.0	1.1	0.6	1.2	1.2

structure types are stable for most of the T metals in the UTM compounds, suggesting less influence of d electrons on a crystal change.

We mention here that Hovestreydt *et al* [25] have already discussed in detail the occurrence of different types of crystal structure in the case of equiatomic ternary RTM compounds (R = rare earth).

3.2. Magnetic properties

In figure 1 we show the temperature dependence of the magnetization (left-hand side) and the reciprocal magnetic susceptibility (right-hand side) for U$_2$RuGa$_3$, U$_2$RhGa$_3$ and U$_2$IrGa$_3$. The significant aspect of the data is that the magnetization σ for all of those samples, measured in a low field ($B = 0.01$ T), presents a huge increase with decreasing temperature. The appearance of an inflection point in the $\sigma(T)$-curves suggests the onset of a ferromagnetic transition. Choosing the Curie temperature as the minimum in $d\sigma(T)/dT$ we have obtained $T_C = 73, 60$ and 72 K for compounds containing Ru, Rh and Ir, respectively. In the case of T = Ir we have found, moreover, another minimum of $d\sigma(T)/dT$ at a temperature of 25 K. The origin of this anomaly is unclear, but may possibly be associated with a reorientation of the magnetic moments of a presumed non-collinear magnetic structure.

The paramagnetic susceptibility for all three of the compounds deviates strongly from a linear Curie–Weiss dependence. This behaviour may be expected for a system in which the contribution of a temperature-independent susceptibility χ_0, is significant. Whether this originates from the contribution of a band structure or perhaps from the anisotropy is unclear. We have fitted the experimental data for $\chi(T)$ to a modified Curie–Weiss (MCW) law:

$$\chi = \chi_0 + N_A\mu_{eff}^2/3k_B(T - \Theta_p). \tag{1}$$

The values determined for the temperature-independent term, χ_0, the paramagnetic Curie temperature, Θ_p, and the effective paramagnetic moment, μ_{eff}, are listed in table 1. The magnetic susceptibility χ_0 appears to be the same for all compounds and amounts to about
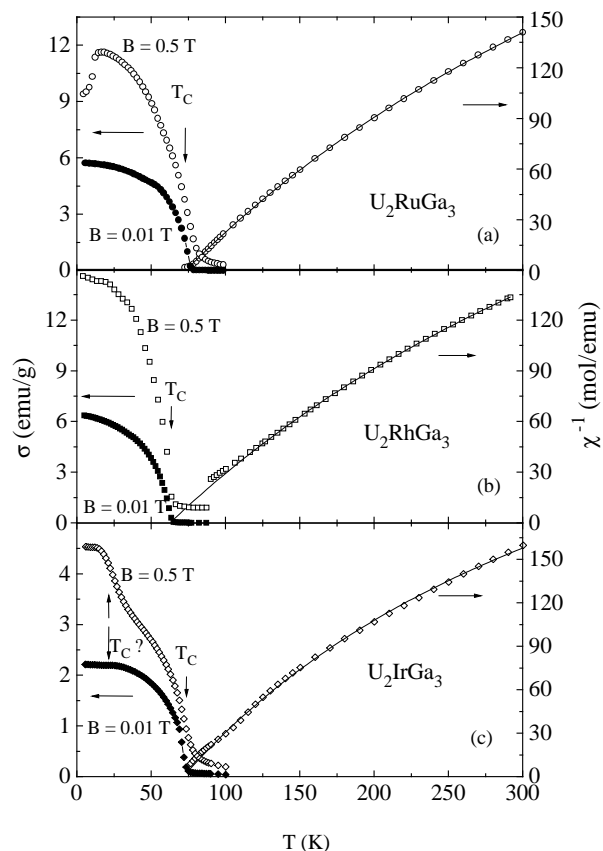
Figure 1. The temperature dependence of the magnetization (left-hand scale) and reciprocal susceptibility (right-hand scale) for (a) U_2RuGa_3, (b) U_2RhGa_3 and (c) U_2IrGa_3. The solid lines represent the MCW fits with the parameters shown in table 1. The arrows show the Curie temperatures.

1.0×10^{-3} emu/U atom. We find positive Θ_p-values of 73, 61 and 72 K for Ru-, Rh-, and Ir-based compounds, respectively. These values are comparable to the corresponding T_C-values and therefore indicate the existence of ferromagnetic interactions in these compounds. Considering the effective paramagnetic moment, we observe that in all cases the values of μ_{eff} are small, but comparable to those of 1:1:1 compounds [26]. Note that the μ_{eff}-values derived from the MCW analysis of the temperature dependence of χ along the easy direction of a compound are always higher than that of polycrystalline samples, e.g. for URhAl [27]. However, they are much less than the moments calculated from the Russel–Saunders coupling (2.54 μ_B for $5f^1$, 3.58 μ_B for $5f^2$ and 3.62 μ_B for $5f^3$).

The magnetization curves for U_2TGa_3 (T = Ru, Rh, Ir) obtained at $T = 4.2$ K are given in figure 2. As is evident from this figure, all samples order ferromagnetically at low temperatures and exhibit considerable remanence, suggesting the presence of a strong magnetic anisotropy. For both the Rh and the Ir compounds the magnetization in fields of 5.5 T appears to be still far from saturation. The spontaneous ferromagnetic moment, μ_s, is obtained from μ versus B^{-1} plots and extrapolating to $1/B = 0$. We obtain $\mu_s = 0.9, 0.4$ and 0.6 μ_B/U atom for the T = Ru, Rh and Ir compounds, respectively. The low value of
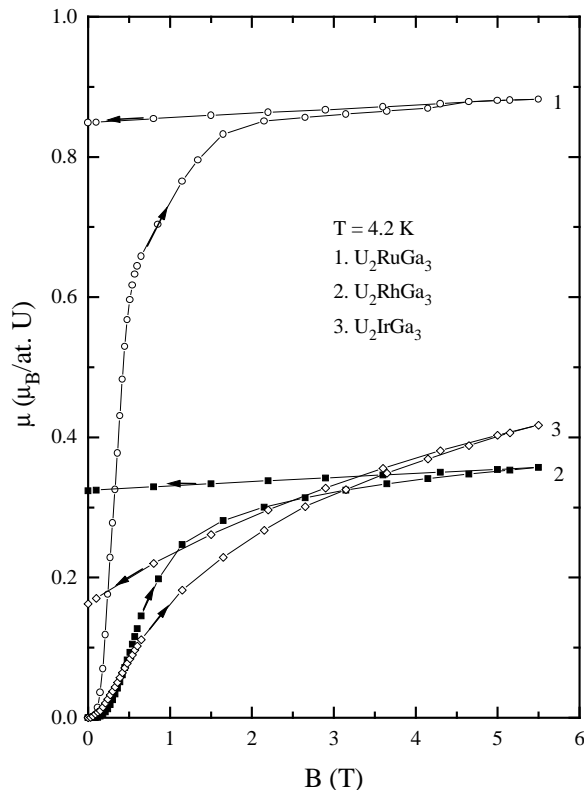
Figure 2. The magnetization as a function of the magnetic field at 4.2 K for U$_2$RuGa$_3$, U$_2$RhGa$_3$ and U$_2$IrGa$_3$.

the magnetization points to an itinerant character of the 5f electrons in these compounds.

The results of dc magnetic susceptibility measurements obtained for compounds of T = Pd and Pt are shown in figures 3(a) and 3(b), respectively. The magnetization of U$_2$PdGa$_3$ shows a peak at about 33 K. This maximum is suggestive of an antiferromagnetic (AF) phase transition in this compound. Above $T = 50$ K, the susceptibility behaviour closely adheres to a Curie–Weiss law. From the fit of the experimental data for this compound to equation (1) we find a small χ_0-value of 0.6×10^{-3} emu/U atom and a positive paramagnetic Curie temperature, $\Theta_p = 8$ K. The latter value indicates that the exchange interactions between the nearest uranium moments have a ferromagnetic characteristic. For U$_2$PdGa$_3$, the effective moment μ_{eff} reaches a large value of 2.58 μ_B/U atom, thus illustrating the increasing trend in the μ_{eff}-value with increasing number of 4d electrons (see table 1). This may be an indication of a more localized character of the 5f electrons in the Pd-based compound.

The temperature dependence of the reciprocal magnetic susceptibility of U$_2$PtGa$_3$ (figure 3(b)) demonstrates clearly non-Curie–Weiss behaviour. From the MCW fit, in the temperature range 120–300 K, an effective paramagnetic moment of 2.14 μ_B/U atom is obtained which is lower than that of U$_2$PdGa$_3$. The value of Θ_p is 45 K and χ_0 is 1.2×10^{-3} emu mol^{-1}. At low temperatures, the magnetic behaviour of U$_2$PtGa$_3$ appears very similar to that of U$_2$TGa$_3$ (T = Ru, Rh and Ir), since the magnetization shows a sharp rise below about 100 K. If one tries to relate this behaviour to the development of
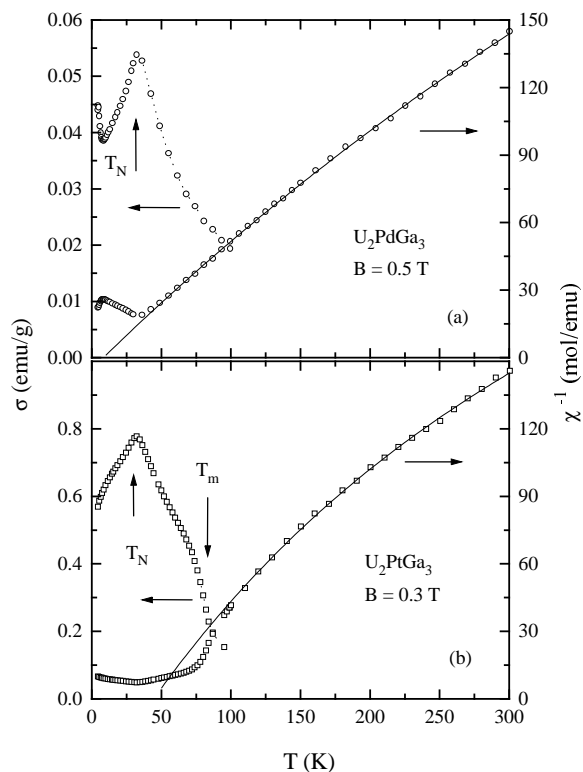
Figure 3. The temperature dependence of the magnetization (left-hand scale) and reciprocal susceptibility (right-hand scale) for (a) U_2PdGa_3, and (b) U_2PtGa_3. The solid lines represent the MCW fits with the parameters shown in table 1. The arrows indicate the magnetic ordering temperatures.

ferromagnetic ordering, then the transition temperature as an inflection point on the $\sigma(T)$-curve occurs at $T_m = 80$ K. However, the $\sigma(T)$-value (see figure 3(b)) is smaller than that of a typical ferromagnet and we suspect that the transition at 80 K may instead be correlated with the presence of weak ferromagnetism.

The magnetization curves of U_2PtGa_3 obtained at 4.2 and 50 K are displayed in figure 4. As expected, the magnetization curve obtained at $T = 50$ K shows downward curvature, resembling the behaviour of a weak ferromagnet. On the other hand the magnetization curve obtained at $T = 4.2$ K is characterized by an upward curvature and followed by a metamagnetic-like transition at about 0.8 T. Above 3 T, the magnetization shows again an upward increase suggesting the onset of a field-induced transition to another magnetic state. This behaviour rather indicates an antiferromagnetic ground state in U_2PtGa_3. Therefore, the difference of the high-temperature magnetization from that at 4.2 K demonstrates the possibility of the existence of two different magnetic phase transitions in U_2PtGa_3 below 80 K and below 30 K (see figure 3(b)).

In accordance with the dc magnetic measurements, the temperature dependence of the ac magnetic susceptibility $\chi_{ac}(T)$ (figure 5) reveals a sharp peak centred at 80 K, followed by a small peak in $\chi_{ac}(T)$ at 30 K. The χ_{ac}-peak at 80 K could be ascribed to the development of a ferromagnetic component and the low-temperature anomaly confirms that U_2PtGa_3 undergoes a transition to another magnetic state. It is clear that the transition at $T = 30$ K
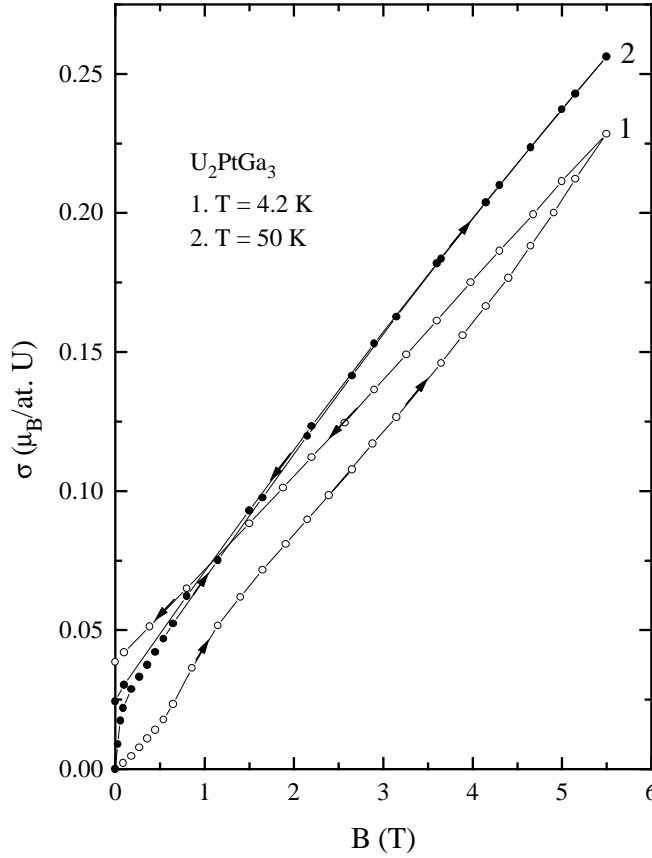
Figure 4. Magnetization curves for U$_2$PtGa$_3$ measured at 4.2 and 50 K.

does not affect the behaviour of $\chi''(T)$, and this is evidence for an antiferromagnetic transition. Here it is noted that there is a resemblance between the magnetic behaviour of U$_2$PtGa$_3$ with that of UCu$_2$Ge$_2$. The latter compound orders ferromagnetically below 105 K and antiferromagnetically at about 45 K [28–30], and the nature of those transitions has been discussed by McAlister *et al* [31] in terms of the ANNNI model [32] and also by Chakravarti *et al* [33] in terms of the Moriya and Usami model [34].

3.3. Electrical properties

The temperature dependence of the normalized electrical resistivity, $\rho(T)/\rho(\text{RT})$, of U$_2$RuGa$_3$, U$_2$RhGa$_3$ and U$_2$IrGa$_3$ is shown in figure 6. In the high-temperature range (>200 K) all of the samples show a similar temperature dependence of ρ, characterized by a negative logarithmic contribution, $-\ln T$, to the total resistivity. This contribution is expected to derive from from the Kondo effect. Using the Kondo formula [35]

$$\rho_{mag}(T) = -C \ln T \tag{2}$$

we analysed the temperature dependence of the resistivity using the following equation:

$$\rho(T) = \rho_0 + \rho_{ph}(T) - C \ln T \tag{3}$$
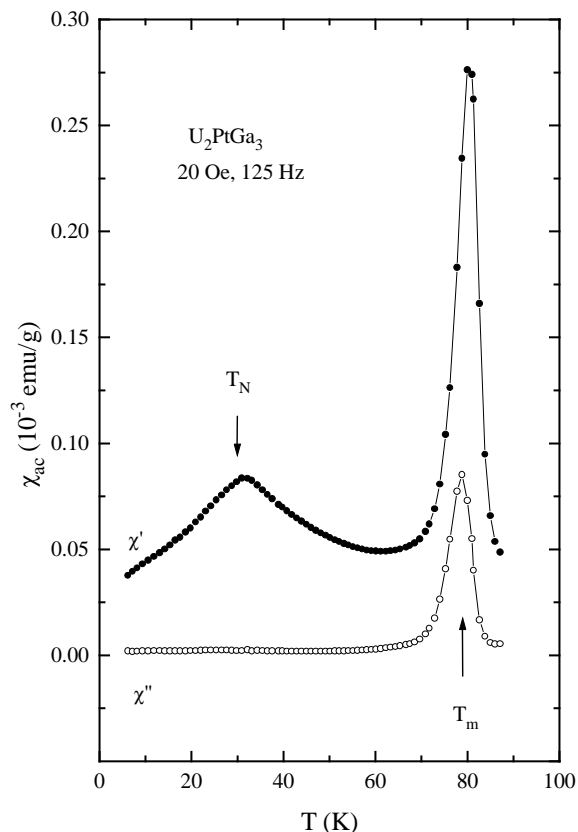
Figure 5. The temperature dependence of the real and imaginary parts, χ' and χ'', of the ac magnetic susceptibility of U_2PtGa_3 measured in a magnetic ac field of 20 Oe and at a frequency of 125 Hz. The arrows indicate the magnetic ordering temperatures.

Table 2. The electrical resistivity characteristics of the U_2TGa_3 compounds, where T = Ru, Rh, Pd, Ir and Pt. (b and C are parameters of equation (3), and ρ_{300} is the room temperature resistivity.)

	b ($\mu\Omega$ cm K^{-1})	C ($\mu\Omega$ cm K^{-1})	ρ_{300} ($\mu\Omega$ cm)
U_2RuGa_3	0.02	6.8	216
U_2RhGa_3	0.04	24.3	228
U_2PdGa_3	0.07	6.2	127
U_2IrGa_3	0.05	24.6	235
U_2PtGa_3	—	—	220

where ρ_0 is the residual resistivity due to impurities as well as due to the effect of cracks and other inhomogeneities of the samples. The second term $\rho_{ph}(T)$ describes the process of scattering with thermally excited phonons and we assumed a linear temperature dependence, bT, for this term when fitting our experimental data to equation (3). The results are shown in figure 6 as solid lines and the corresponding values of the parameters b and C are given in table 2.
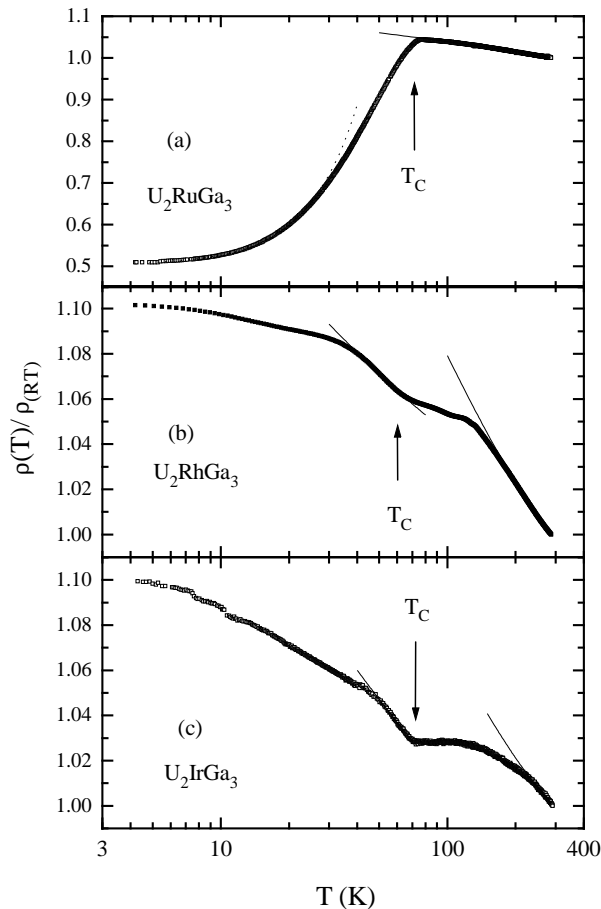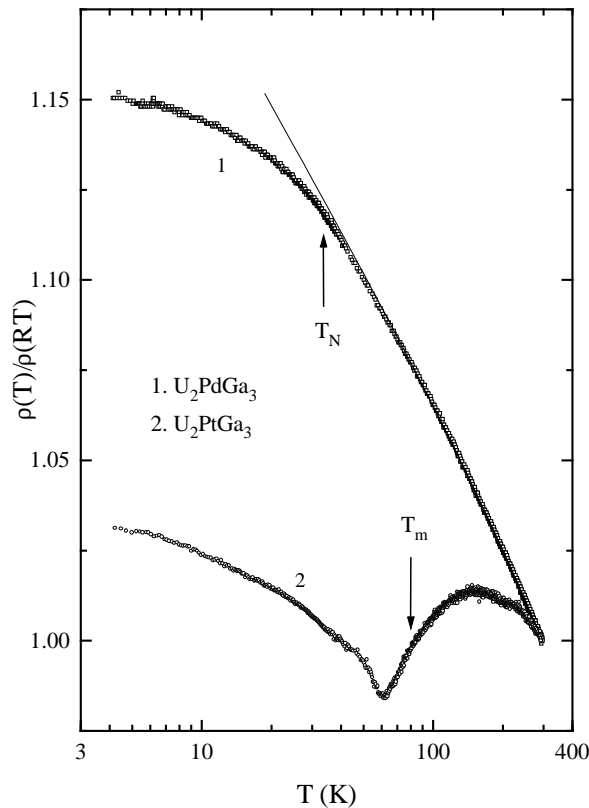
Figure 6. The temperature dependence of the electrical resistivity, ρ versus $\log T$, for the compounds (a) U$_2$RuGa$_3$, (b) U$_2$RhGa$_3$ and (c) U$_2$IrGa$_3$. The solid lines illustrate the Kondo-like behaviour. The dotted line represents the fit to a T^2-law. The arrows indicate the Curie temperatures.

At low temperatures, the U$_2$RuGa$_3$ compound shows a sharp drop of ρ just below T_C (figure 6(a)). This is due to the decrease in the spin-disorder resistivity, resulting from the magnetic ordering. In the range 4.2–25 K, $\rho(T)$ for this compound approximately follows a T^2-law, the T^2-coefficient being 0.06 $\mu\Omega$ cm K^{-2}. The low-temperature behaviour is characteristic of spin fluctuations. It is worth noting that the overall temperature dependence of U$_2$RuGa$_3$ is reminiscent of that of the ferromagnetic Kondo system UTe [36], which has a $(-\ln T)$-dependence over a wide temperature range.

For the Rh- and Ir-based compounds, the electrical resistivity behaviours shown in figures 6(b) and 6(c), respectively, are rather similar. As seen from these figures, a distinct anomaly in $\rho(T)$ appears around their Curie temperatures, below which ρ still increases with further decreasing temperature. Thus, $\rho(T)$ for both compounds has two temperature regimes with $(-\ln T)$-contributions. This behaviour is consistent with the theory given by Cornut and Coqblin [37] for the case of combined interactions of the Kondo effect and crystal-field splitting. Following this theory, the observed broad maximum at around 110 K

in the $\rho(T)$-curve of U_2IrGa_3 may act as a measure of the overall crystal-field splitting. In other words, we may speculate that U_2IrGa_3 could be a uranium analogue to many Ce- or Yb-based compounds, such as $CeAl_2$ [38], $CeCuSi_2$ [39] and YCu_4Au [40], where the influence of the crystalline field on the Kondo effect is believed to be considerable.

Another prominent feature of the resistivity behaviour of the compounds U_2RhGa_3 and U_2IrGa_3 is the tendency to saturation at temperatures below 5 K. This may be due to the incoherent Kondo effect, but further detailed experiments at lower temperatures are needed to establish this.

Figure 7. The temperature dependence of the electrical resistivity, ρ versus $\log T$, for U_2PdGa_3 and U_2PtGa_3. The solid line illustrates the Kondo-like behaviour found for U_2PdGa_3. The arrows indicate the magnetic ordering temperatures.

The electrical resistivity $\rho(T)$ of U_2PdGa_3 (figure 7) shows a Kondo-like behaviour with a $(-\ln T)$-increase on cooling down to 80 K. The fit of experimental data to equation (3) is shown in figure 7. The onset of magnetic order in this compound is hardly detectable from a weak anomaly associated with the $\rho(T)$-curve at $T = 34$ K. The electrical resistivity behaviour of U_2PtGa_3 is atypical for an ordered material. At high temperatures, ρ shows a broad maximum at around 150 K. Below T_m the resistivity decreases drastically with decreasing temperature, and goes through a deep minimum at $T = 60$ K. However, no anomaly is detected in $\rho(T)$ at around $T_N = 30$ K. The lack of such an anomaly needs an explanation based on detailed investigation including both neutron diffraction and specific heat measurements on samples of better quality. Nevertheless, we recall that the

already mentioned UCu$_2$Ge$_2$ material also does not show a resistivity anomaly at its Néel temperature [31]. The electrical resistivity of U$_2$PtGa$_3$ and URh$_2$Ge$_2$ [41] may be compared: the latter compound displays a flat maximum at about 100 K and a shallow minimum at 15 K and it is apparent that the anomaly observed in the $\chi(T)$-curves near 8 K for this material does not affect its ρ-behaviour [41–43].

At present, one should bear in mind that the large residual disorder scattering can mask magnetic signatures expected in the resistivity of Pd- and Pt-containing compounds. This often occurs for a compound with significant crystallographic disorder and may happen for all of the U$_2$TGa$_3$ compositions; a random distribution of T and Ga at the 8h sites of the CeCu$_2$-type unit cell might account for large residual resistivity and hence for the low RRR value (ratio of the residual to the room temperature resistivity).

4. Discussion

From the results presented above, it can be concluded that there is a correlation between the filling of the d band and the observed magnetic ordering. For compounds with less populated d states of the transition metal, i.e., Ru, Rh and Ir, the magnetic ground state is ferromagnetic, whereas compounds with almost filled d bands like Pd and Pt favour an antiferromagnetic ordering. This correlation is found for most of the UTM series, for which experimental and theoretical studies [26, 44] demonstrate that the 5f–ligand hybridization strength increases as one moves from the right to the left in a d-transition-metal row and/or one moves from group IIIA to group IVA elements. Therefore the mechanism of 5f–ligand hybridization [45], which has been used for interpreting the magnetic ordering trend in the UTM family, might also be applied for the U$_2$TGa$_3$ series.

Comparing the magnetic behaviour of the two series UTGa and U$_2$TGa$_3$, one observes that the magnetic state of the latter series develops more in the direction of decreased 5f–d hybridization, due to the decrease of d-electron numbers. In fact URuGa was found to be paramagnetic down to 20 mK [18], while for the corresponding U$_2$RuGa$_3$ compound we have observed the highest spontaneous moment and the highest T_C-value of the whole U$_2$TGa$_3$ series studied. In turn, the UPtGa compound exhibits a ferromagnetic phase transition at about 70 K [19, 20] and the U$_2$PtGa$_3$ compound shows anomalies at 30 and 80 K. Moreover, unlike for UTGa, where one observes the influence of spin fluctuations on the electrical resistivity, the U$_2$TGa$_3$ compounds demonstrate instead the existence of a Kondo-like behaviour. This difference in transport behaviour may be linked to enhancement of 5f–sp hybridization in the case of U$_2$TGa$_3$. Therefore, the increase in the number of conduction electrons in compounds plays a major role in structural change as well as in development of the magnetic ground state.

5. Conclusion

Structural, magnetic and electrical resistivity studies have been carried out on a new series of uranium intermetallics U$_2$TGa$_3$, where T = Ru, Rh, Pd, Ir and Pt. These compounds were found to crystallize in the orthorhombic CeCu$_2$-type structure, and to order magnetically at low temperatures. The development of magnetic states is found to be similar to that of the equiatomic 1:1:1 compounds, except that the 5f electrons in U$_2$TGa$_3$ have a more localized character than their UTGa counterparts. The observed electrical resistivity behaviour can be described in terms of the Kondo effect. The determination of the magnetic structure and other microscopic studies of the U$_2$TGa$_3$ series of compounds are necessary.

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