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High temperature electrical resistivity in $U_{1-x}La_xPt$ compounds

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Abstract

We present electrical resistivity measurements on $U_{1-x}La_xPt$ ($x = 0$, 0.1, 0.2, 0.5, 0.8, 0.9, 1) between 2 and 1000 K. The compounds exhibiting ferromagnetic behaviour ($x \leq 0.3$) display a maximum at low temperatures and some of them a minimum at higher temperatures. For the non-ordered compounds ($x \geq 0.5$) the resistivity increases continuously with temperature. A common feature of the U-rich compounds is the existence of a negative logarithmic temperature dependence of the total resistivity above the maximum, whereas a combination of a linear term plus a logarithmic term is found for the others. Under a Kondo-like framework, a breakdown of the Mattiessen rule is suggested.

Keywords: Intermetallic; Electronic transport; Kondo effects

1. Introduction

The electrical resistivity of 'normal' metallic materials at sufficiently high temperatures is dominated by electron–phonon scattering, giving rise to a positive increment of the electrical resistivity with temperature [1]. However, some 4f (Ce, Yb) and 5f (U, Pu) compounds display opposite behaviour, and the resistivity decreases with temperature, showing a negative temperature coefficient (NTC) even up to room temperature. These compounds belong to a group of materials which has attracted much attention in recent years due to their unusual properties associated with 4f or 5f conduction band hybridisation and which are described in terms of heavy fermion, intermediate valence or Kondo lattice behaviour. In this sense, materials such as $CeCu_2Si_2$ [2], $YbCu_4Ag$ [3] and UPd_2Ga_3 [4] show a maximum at low temperatures followed by a continuous decrease of the electrical resistivity up to room temperatures. Despite this interesting anomalous behaviour, no electrical resistivity measurements above room temperature in NTC materials can be found in the literature.

UPt is a moderate heavy-fermion material ($\gamma \approx 105$ mJ/kmol) which orders ferromagnetically at $T_C = 27$ K [5,6]. Its electrical resistivity agrees with the previous general description, showing a NTC at room temperature. Its magnetic properties are strongly dependent on pressure [7], suggesting that it is close to the crossover between localised and delocalised magnetic behaviour. One controversial point is the crystal structure [8], first reported to be orthorhombic CrB-type [9], then monoclinic PdBi-type [10], whereas high resolution neutron diffraction measurements are not fully in agreement with either of these structures [11].

When we substitute La for U, the cell volume increases and continuous evolution to a 'normal' non-magnetic metal is obtained [12]. The magnetisation curves show ferromagnetic behaviour for $x < 0.5$ and paramagnetic behaviour for $x > 0.5$. The ordering temperatures derived from Arrot plots are $T_C = 18.5$ and 13.5 K for $x = 0.1$ and 0.2, respectively. If we extrapolate the variation of T_C with La concentration, non-ordered magnetic behaviour is expected for $x \geq 0.4$. However, traces of irreversibility in the field-cooled (ZC) and zero-field-cooled (ZFC) susceptibility have been found in the non-ordered compounds ($x = 0.5$ and 0.2), reminiscent of spin-glass or superparamagnetic behaviour. In this paper we present the electrical resistivity measured from 2 to 1000 K in this series of compounds.

2. Experimental

Polycrystalline $U_{1-x}La_xPt$ samples ($x = 0$, 0.1, 0.2, 0.5, 0.8, 0.9, 1) were melted in an arc furnace under argon atmosphere. They were then wrapped in tantalum foil and

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annealed under high vacuum at 900°C for 2 days. The electrical resistivity was measured using a four-probe dc technique. For the low temperature range (2–300 K) we used a standard PPMS Quantum Design apparatus, whereas for the high temperature range (260–1000 K) we developed a system in which the samples were placed in a quartz tube under high vacuum to avoid sample oxidation. The contacts were made using gold wires directly spot welded to the samples. The temperature was measured with a K-type thermocouple. Typical heating rates were 2 K/min. The estimated error in the absolute value of the electrical resistivity due to uncertainty in the sample geometry was 5%.

3. Results and analysis

The electrical resistivity for all the studied compounds is depicted in Fig. 1. For the UPt compound, the electrical resistivity strongly increases up to 27 K due to a spin disorder contribution, then passes through a wide maximum at 120 K and finally, upon heating, decreases slightly up to 770 K, where a minimum appears. The general behaviour is similar for the ordered compounds $x = 0.1$ and 0.2, which display a maximum and a NTC. For larger La concentrations ($x \geq 0.5$), the temperature coefficient becomes positive and the maximum disappears. The strong increase of the resistivity in the low temperature region decreases progressively with increasing La percentage. Finally, LaPt displays typical phonon behaviour.

In addition to the previous general description, some anomalous features are observed for $x = 0.1$ and 0.2 in the high temperature region. For the first compound, instead of a minimum, a change of slope is observed at 600 K. For the second compound, above the minimum observed at $T = 420$ K, a second minimum develops at 850 K (see arrows in Fig. 1). These extra anomalous behaviours are quite puzzling and the most likely way to explain them is the existence of a structural rearrangement at high temperatures. As commented on in the Introduction, the crystal structure of UPt is still not well resolved and extra lines in the X-ray diffraction pattern persist with La substitution [12]. In addition, we have to bear in mind that the electrical resistivity is quite sensitive to small structural changes. For a better understanding of these phenomena, DSC and X-ray measurements up to 1000 K are now in progress.

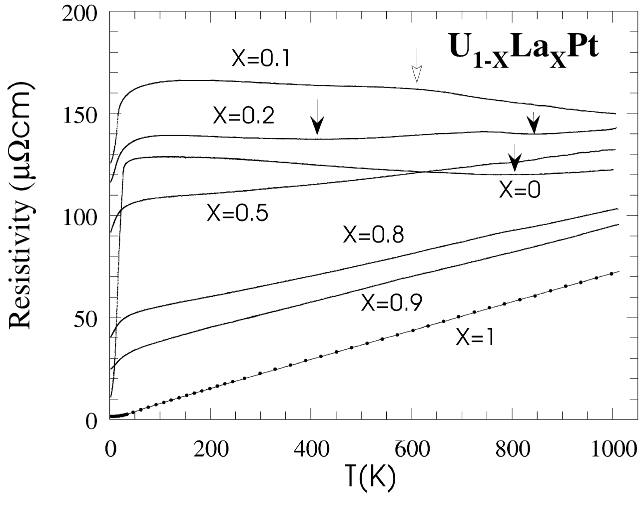

Fig. 1. Temperature dependence of the electrical resistivity for all the studied compounds between 2 and 1000 K. For the LaPt compound, the points represent some of the experimental data and the solid line the fit to the Bloch-Grüneisen law. Full arrows indicate the minima positions, and the empty arrow points to the change of slope for $x = 0.1$.

In order to analyse the electrical resistivity, the independence of the scattering processes (Mattiessen rule) is usually accepted; the total resistivity is given by

$$\rho(T) = \rho_0 + \rho_{\text{ph}}(T) + \rho_{\text{mag}}(T)$$

where ρ_0 originates from the scattering of the conduction electrons on the lattice imperfections, ρ_{ph} describes the resistivity arising from the electron–phonon interaction and ρ_{mag} is inferred from the spin-dependent scattering processes.

With respect to the evolution of the residual resistivities in $A_{1-x}B_x$ solid solution, according to the Nordheim rule the structural disorder produces a residual resistivity that is proportional to $x(1-x)$. Fig. 2 plots the experimental values of ρ_0 obtained from extrapolation of the resistivity to 0 K (points) compared with a modified Nordheim formula:

$$\rho_0(\text{UPt}) + x[\rho_0(\text{LaPt}) - \rho_0(\text{UPt})] + Cx(1-x)$$

(solid line). We note that this modification is needed in order to account for the non-zero values of the residual resistivity in the $x = 0$ and 1 compounds. Starting from

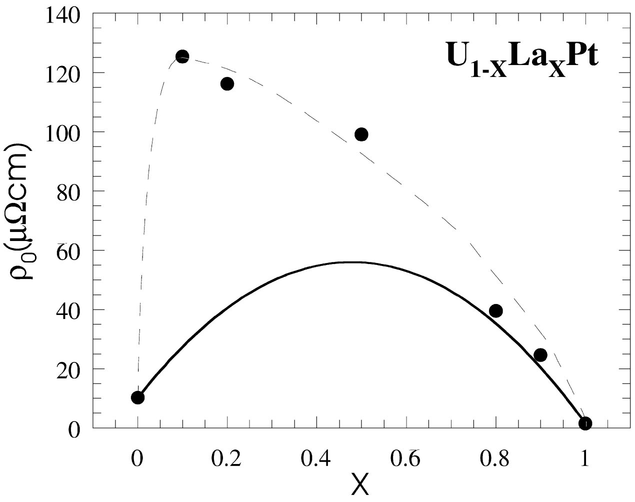

Fig. 2. Evolution of the residual resistivity with La percentage. (———) Modified Nordheim formula; see text. (- - -) A guide to the eye.

UPt, a strong increase of ρ_0 is observed for $x = 0.1$ and then a continuous decrease with increasing La substitution. This feature could be related to the existence of additional residual resistivity of magnetic origin. This residual magnetic resistivity could explain the apparent strong decrease of the spin disorder resistivity with only 10% La substitution.

The phonon contribution is usually estimated by measuring the resistivity of an isomorphous non-magnetic compound. In our series, the compound accounting for these conditions is LaPt. Its resistivity follows a T^5 dependence at low temperatures and shows linear behaviour above 150 K. The fit of the experimental values to the Bloch-Grüneisen law is represented by the solid line in Fig. 1 and yields a Debye temperature of $\theta_D = 140$ K and an electron–phonon interaction constant of $R = 31.4$ μΩ cm K^{-1}. Note the excellent quality of the fit, which is mainly due to the perfect linear behaviour followed by the experimental data up to 1000 K.

The usual way to determine the magnetic resistivity is to subtract the residual and phonon contributions. Under the hypothesis that the phonon contribution for all the compounds is similar to that of LaPt, we can calculate their magnetic resistivity using the expression

$$\rho_{\mathrm{mag}} = [\rho(T) - \rho_0]_{(\mathrm{ULa})\mathrm{Pt}} - [\rho(T) - \rho_0]_{\mathrm{LaPt}}$$

(see Fig. 3). The magnetic contributions display the same general dependence for all the compositions: a strong increase at low temperatures, followed by a maximum around 50 K and then a decrease up to 1000 K. With an increase of La percentage, the maximum becomes rounder and its resistivity value decreases, as expected due to dilution effects. The strong initial increase of the magnetic resistivity at low temperatures for the ordered compounds ($x \leq 0.2$) is mainly due to spin disorder resistivity, whereas for $x \geq 0.5$, when the long range magnetic order has disappeared, it can be related to some kind of coherence effects in a Kondo system, as we will discuss later. It is worth mentioning that the average negative slope decreases with increasing La content. Despite the nice evolution of the magnetic resistivities, we have to point out that negative magnetic resistivity values are reached at high temperatures. This indicates that the analysis performed on these compounds is a rough approach. In fact, in addition to changes in the phonon contribution in the U-based samples, the different scattering processes may not be completely independent.

Returning to the total resistivity, the existence of a minimum or a NTC is usually considered as a fingerprint of the Kondo effect. In this sense, the resistivity of many heavy-fermion compounds such as $CeAl_3$ [13] and $CeCu_6$ [14], with a maximum at low temperature and a NTC at room temperature, is usually interpreted within the Kondo lattice model. The idea is that f sites scatter electrons incoherently at high temperatures where the lattice appears as a set of Kondo impurities. The maximum of ρ_{mag} and the large decrease at lower temperatures is clear evidence for the onset of coherence and the formation of the Kondo lattice. Within this model, a negative logarithmic dependence is expected in the magnetic resistivity. Therefore, at elevated temperatures, where the phonon contribution is linear, the total resistivity should follow a simple analytical temperature dependence: $\rho = a + bT - c \ln T$, where the linear parameter (b) is not very different from that of the non-magnetic compound. This expression was used successfully to fit the resistivity of $YbCu_4Ag$ [13] between 100 and 300 K and URu_2Si_2 above room temperature [15]. Within this framework, applying this expression to our compounds, we obtain interesting behaviour. First, for the compounds showing a NTC ($x \leq 0.2$), the linear term is

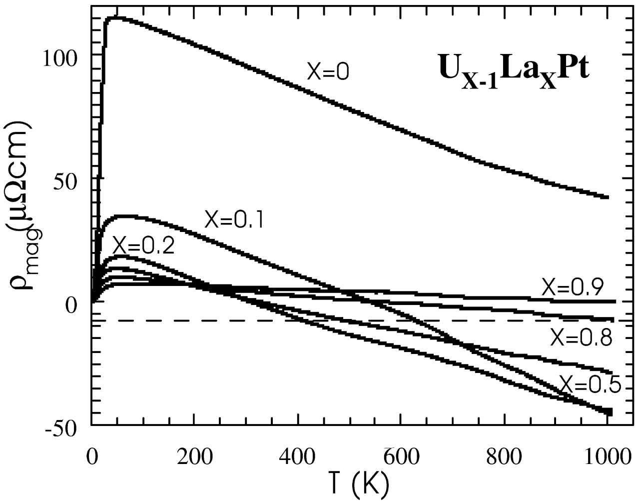

Fig. 3. Temperature dependence of the magnetic resistivity estimated as described in the text.

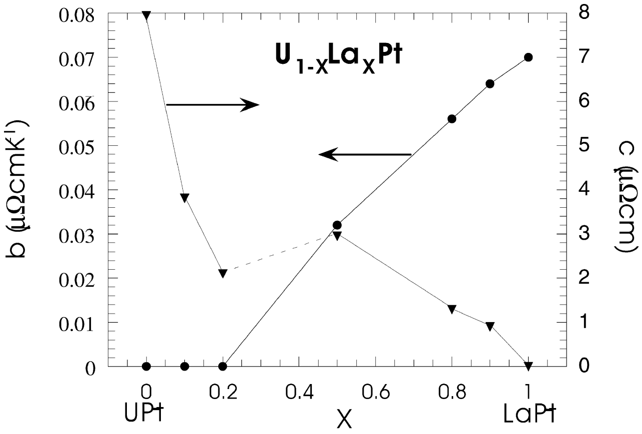

Fig. 4. Evolution of the linear coefficient, b, and the logarithmic coefficient, c, of the electrical resistivity as a function of the La percentage, x.

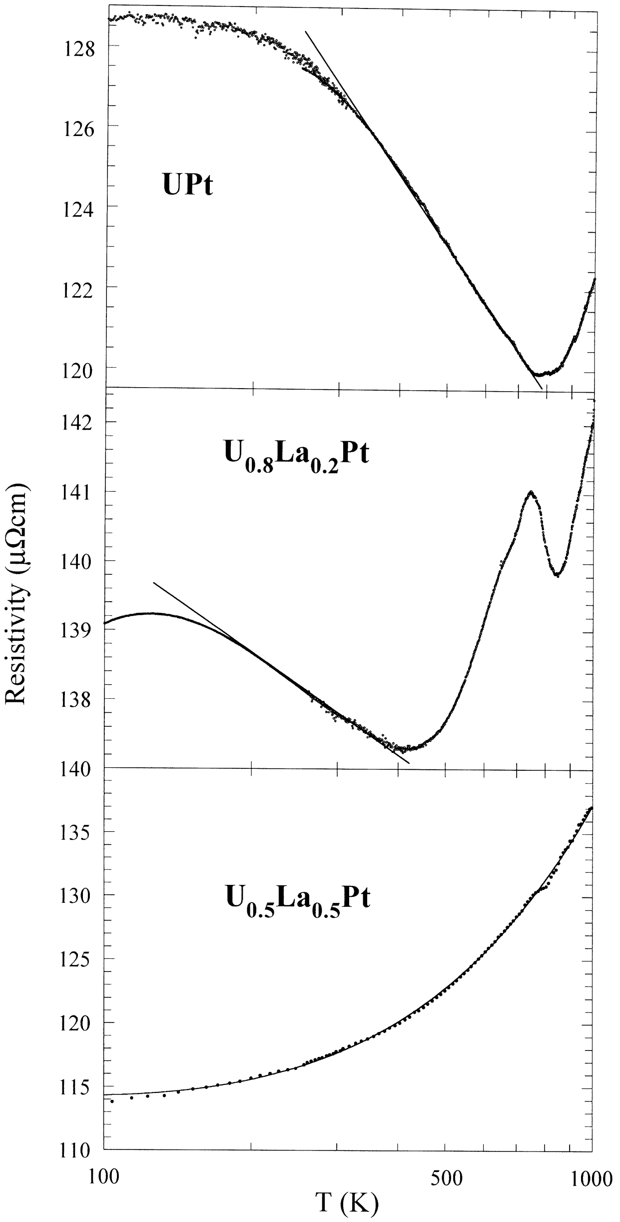

Fig. 5. Temperature dependence of the electrical resistivity on a logarithmic temperature scale above 100 K for (a) UPt, (b) $U_{0.8}La_{0.2}Pt$ and (c) $U_{0.5}La_{0.5}Pt$. (———) Fits to a $\rho = a + bT - c\,\ln(T)$ law.

zero and, therefore, the total resistivity follows a simple logarithmic dependence with a *c* parameter which decreases with increasing *x* (see Fig. 4). On the other hand, for the compositions with $x \geq 0.5$, the linear coefficient increases and the logarithmic coefficient decreases when substituting U by La, finally reaching linear behaviour for LaPt. In order to show this evolution and the temperature range of the fits, we present in Fig. 5 the resistivities of UPt, $U_{0.8}La_{0.2}Pt$ and $U_{0.5}La_{0.5}Pt$ above 100 K on a logarithmic temperature scale. In principle, the linear coefficient should not change too much because it is related to the phonon contribution; however, it changes drastically, even disappearing, for the U-rich compounds. This indicates that the hypothesis of independent scattering processes (Mattiessen rule) is clearly questionable for this system as already suggested for other U compounds [16].

In conclusion, resistivity measurements performed over a very large temperature range up to 1000 K allow us to stress the following points. (a) At high temperatures, effects corresponding to structural changes are detected, confirming the sensitivity of the resistivity with respect to minor structural modifications [17]. (b) The extended temperature range above 300 K reveals negative values for the estimated magnetic resistivities (see Fig. 3), which questions the independence of the scattering processes. This point is reinforced by the analysis of the total resistivity at high temperatures using a Kondo-type model, which leads to a zero coefficient for the linear contribution to the resistivity of the U-rich compounds.

Following these conclusions, as our experimental results suggest, a more elaborated theoretical model considering the interplay of the different contributions (Kondo, phonons, etc.) is needed, as has already been reported by various authors [16,18].

Acknowledgements

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