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On the magnetic and electrical behaviour of UPdSb

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Abstract

The ternary compound UPdSb, crystallizing with a hexagonal structure of the $CaIn_2$-type (s.g. $P6_3/mmc$), was studied by means of magnetic susceptibility, magnetization, electrical resistivity, magnetoresistivity and thermoelectric power measurements. The results revealed a strongly anisotropic nature of the magnetic characteristics with a ferromagnetic ordering below $T_C = 77$ K. The electrical behaviour of UPdSb is characteristic of moderately disordered metals showing a rather high magnitude of resistivity in the entire temperature range, little temperature variation with a negative slope in the paramagnetic region and relatively small values of the Seebeck coefficient. Two other mechanisms that bring about a negative temperature coefficient of the resistivity in similar systems, i.e. the Kondo effect or formation of a small energy gap near the Fermi level, are critically discussed.

1. Introduction

During the last decade uranium-based compounds with the overall composition UTM, where T is a d-electron transition metal and M stands for a p-electron element, have attracted much attention for their large variety of physical behaviours, related to extreme sensitivity to the degree of hybridization of uranium 5f electrons with s, p and/or d electrons of neighbouring atoms [1]. The UTM phases crystallize with several different crystal structures, of which the hexagonal ZrNiAl and $CaIn_2$ types, the orthorhombic TiNiSi type and the cubic MgAgAs type are represented most frequently. In these intermetallics long-range magnetic ordering that usually sets in at low temperatures often coexists with semiconductor-like or half-metallic electrical conductivity. Several UTM compounds are also known which show features of heavy fermion systems. Of the materials containing palladium, such a behaviour was reported, for example, for UPdIn; this compound exhibits antiferromagnetic ordering below $T_N = 20$ K with a ferromagnetic component below 7 K [2]. Other examples are magnetically ordered Kondo lattices: UPdGa (two subsequent antiferromagnetic-like transitions at 30 and 62 K [3]), UPdSi

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(antiferromagnetic-like transitions at 27 and 33 K [4]) and UPdGe (antiferromagnetic transition at 50 K and a ferromagnetic one at 28 K [5, 6]). In turn, UPdSn exhibits an antiferromagnetic ordering below 29 K [7].

In the course of our systematic studies on the magnetic, electrical and thermal behaviour of (Ln/An)PdSb intermetallics, where Ln/An stands for an lanthanoid/actinoid [8, 9], we have recently focused our attention on the uranium-based material. Previous study of UPdSb [7, 10] revealed ferromagnetic ordering with $T_C = 65$ K and quite a large electrical resistivity that is nearly temperature independent in the paramagnetic state. In this paper we communicate the results of a detailed reinvestigation of the magnetic and transport properties of this compound. To the best of our knowledge, the magnetoresistivity and thermoelectric power data are reported here for the first time.

2. Experimental details

A polycrystalline sample of UPdSb was synthesized by arc-melting stoichiometric amounts of the constituents U (99.8 wt%), Pd (99.999 wt%) and Sb (99.999 wt%) in a titanium-gettered argon atmosphere. The button was remelted several times to ensure good homogeneity. No further heat treatment was applied. The quality of the sample thus obtained was checked by powder x-ray diffraction using a Siemens diffractometer with Cu Kα radiation. The material was found to be single phase.

The magnetic properties were studied between 1.7 K and room temperature and in magnetic fields up to 5 T using a Quantum Design SQUID magnetometer. The electrical resistivity was measured from 4.2 to 300 K using a four-point DC technique. Magnetoresistivity measurements were made in magnetic fields up to 8 T using a commercial AMI superconducting magnet. The thermoelectric power was measured from 6 to 300 K employing a differential method with copper as the reference material.

3. Results and discussion

The powder diffraction pattern of the studied sample of UPdSb was indexed within a hexagonal structure of the $CaIn_2$ type (space group $P6_3/mmc$). No foreign lines were observed. The refined lattice parameters were $a = 4.593(1)$ Å and $c = 7.221(2)$ Å, in good agreement with the values reported in the literature [7].

The temperature dependence of the inverse magnetic susceptibility of UPdSb is shown in figure 1. The sample investigated orders ferromagnetically at $T_C = 77$ K (derived as a maximum in the derivative $d\chi/dT$), which is considerably higher than the value of 65 K reported previously [7, 10]. In the paramagnetic region the $\chi(T)$ variation may be approximated by a modified Curie–Weiss law $\chi(T) = \frac{N\mu_{eff}^2}{3k_B(T-\theta_p)} + \chi_0$ with the effective magnetic moment $\mu_{eff} = 2.64\ \mu_B$, the paramagnetic Curie temperature $\theta_p = 74$ K and the temperature-independent term $\chi_0 = 5.6 \times 10^{-4}$ emu mol^{-1} (note the solid line in figure 1). The value of μ_{eff} is considerably smaller than the free-ion values for both trivalent (3.62 μ_B) and tetravalent (3.58 μ_B) uranium ions, presumably mainly due to crystal field interactions. The positive paramagnetic Curie temperature is consistent with the ferromagnetic ordering, and the value of θ_p is close to T_C. It is worth noting that the paramagnetic characteristics of UPdSb derived in this way are somewhat different from those reported in [7], namely $\mu_{eff} = 2.92\ \mu_B$ and $\theta_p = 70$ K, which were, however, determined from a simple Curie–Weiss fit in the range 80–680 K.

The magnetic field dependence of the magnetization measured at $T = 1.9$ K is displayed in the inset to figure 1. It is characterized by a wide magnetization loop with a sharp transition at a rather high critical field $B_{cr} = 1.8$ T, both features being characteristic of a strongly anisotropic

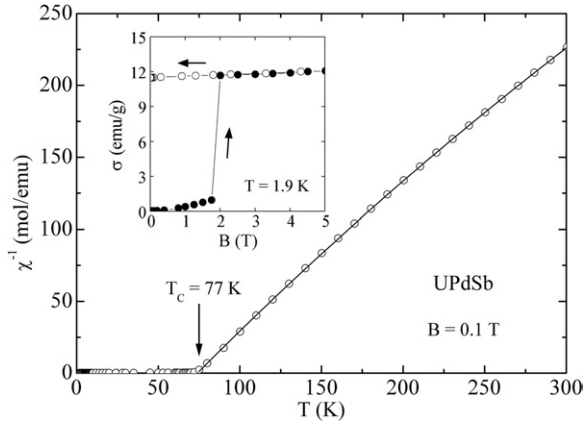

Figure 1. Temperature dependence of the inverse magnetic susceptibility of UPdSb measured in a magnetic field of 0.1 T. The solid line is a modified Curie–Weiss fit with the parameters given in the text. The arrow indicates the ferromagnetic phase transition at $T_C = 77$ K. Inset: isothermal magnetization versus magnetic field, taken at $T = 1.9$ K with increasing (full circles) and decreasing (open circles) magnetic field.

ferromagnet with a compensated domain structure and narrow Bloch walls. In stronger fields the magnetization in UPdSb saturates at a value of 12 emu g^{-1} that corresponds to a uranium magnetic moment $\mu_s = 1.0\ \mu_B$. Thus, the present results essentially confirm the previous findings by Palstra *et al* [7], who reported similar $\sigma(B)$ variation taken at 1.57 K, yet with B_{cr} being as large as 2.2 T and a still unsaturated magnetic moment of about 0.75 μ_B in a field of 4.5 T.

Figure 2 shows the temperature dependences of the magnetization taken in magnetic fields of 0.01 and 0.1 T, upon cooling the specimen in zero (ZFC) and applied (FC) magnetic field. Pronounced irreversibilities occurring in $\sigma(T)$ measured in the FC and ZFC regimes as well as the negative sign of the ZFC magnetization observed at the lowest temperatures are obvious indicators of strong domain effects, in line with the aforegiven interpretation of the $\sigma(B)$ variation.

Figure 3 shows the magnetic field dependences of the magnetization taken at several different temperatures and plotted in the form of Arrott's functions B/σ versus σ^2. The Arrott-plot analysis yields straight lines, which define the temperature variation of the spontaneous magnetization σ_s (intercepts with the abscissa axis) and the Curie temperature (isotherm crossing the origin). The so-derived magnetic ordering temperature is 77 K, in agreement with the $\sigma(T)$ data. The obtained variation of the reduced spontaneous magnetization $\sigma_s(T)/\sigma_s(0)$ versus reduced temperature T/T_C is displayed in figure 4, together with a few theoretical functions derived either from the 2D [11] or 3D [12] Ising models or within the molecular field approximation (MFA) [13]. As seen, the experimental data are situated fairly off each of the theoretical curves, yet the MFA model seems to describe the magnetic properties of UPdSb much better than the other models.

The temperature dependence of the electrical resistivity of UPdSb is shown in figure 5. Apparently, no simple metallic behaviour is observed. In the entire temperature range studied the magnitude of the resistivity is quite large, being of the order of mΩ cm (about 2.7 mΩ cm at room temperature and 1.2 mΩ cm at 4.2 K); moreover $\rho(T)$ exhibits a clear negative temperature coefficient in the paramagnetic state. The latter finding differs considerably from those of Palstra *et al* [7], who reported a tiny increase of the resistivity with rising temperature

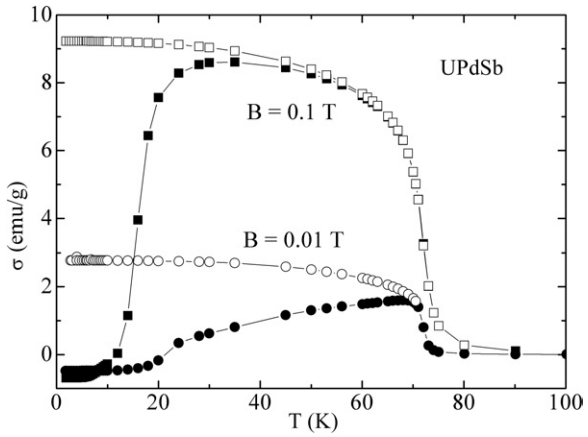

Figure 2. Temperature dependence of the magnetization of UPdSb measured in magnetic fields of 0.01 T (circles) and $B = 0.1$ T (squares). Full and open symbols denote the magnetization data measured upon cooling the specimen without and with an applied magnetic field, respectively.

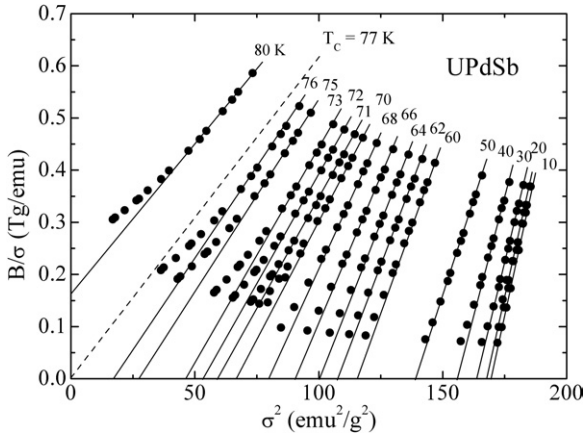

Figure 3. Arrott's plot for UPdSb. The various isotherms were taken at the temperatures given in the figure. The solid lines are respective linear approximations. The dashed line corresponds to the result expected at the Curie temperature.

above T_C. However, also in their case the resistivity was very large (about 5.2 mΩ cm at room temperature and 3.4 mΩ cm at 4.2 K).

The magnetic phase transition in UPdSb manifests itself as a pronounced kink on the $\rho(T)$ curve and a sharp peak in the temperature dependence of the derivative $\mathrm{d}\rho/\mathrm{d}T$ (see the inset to figure 5). The Curie temperature derived from these data is 72 K, i.e. it is slightly lower than that determined in the magnetic studies. In the ordered state the resistivity decreases with decreasing temperature, and below about 20 K it may be described by the formula

$$\rho(T) = \rho_0 + AT^2 \exp\left(\frac{-\Delta}{T}\right) \tag{1}$$

predicted for anisotropic ferromagnets [14]. In this equation ρ_0 stands for the residual resistivity due to scattering conduction electrons on lattice imperfections, while the second term accounts for scattering processes on ferromagnetic spin-wave excitations over the energy gap Δ in

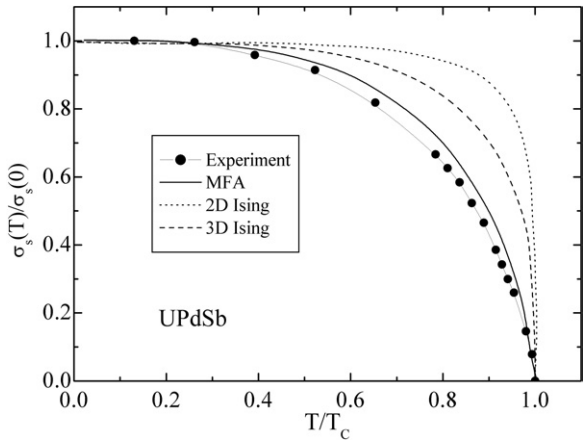

Figure 4. Reduced magnetization versus reduced temperature derived from the Arrott's plot for UPdSb (solid circles and thin line). The lines represent the theoretical functions derived within molecular field approximation (solid curve), 3D Ising model (dashed curve) and 2D Ising model (dotted curve).

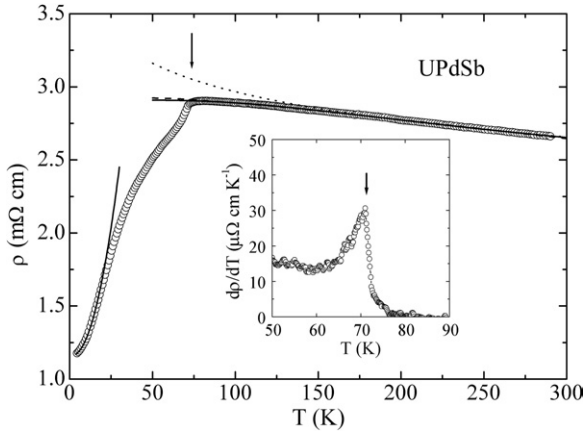

Figure 5. Temperature dependence of the electrical resistivity of UPdSb. The solid lines are least-squares fits of the experimental data to equations (1) and (4) (see the text). The dotted and the dashed lines are least-squares fits of the experimental data to equations (2) and (3), respectively. Inset: temperature derivative of the resistivity in the vicinity of T_C. The arrows indicate the ferromagnetic phase transition.

the magnon spectrum. The least-squares fit parameters are: $\rho_0 = 1.16$ mΩ cm, $A = 1.6\ \mu\Omega$ cm K^{-2} and $\Delta = 3.2$ K.

In turn, above T_C the resistivity of UPdSb gradually decreases with increasing temperature. Such a behaviour is often observed for uranium-based intermetallics and it is usually interpreted as a fingerprint of the Kondo effect. Indeed, also in the present case the $\rho(T)$ dependence may be described by the expression:

$$\rho(T) = \rho_0 + \rho_0^{\infty} + c_K \ln T \tag{2}$$

that accounts for spin-flip scattering of conduction electrons on localized magnetic moments (third term), and standard scattering on lattice defects (ρ_0) and disordered spins (ρ_0^{∞}). Fitting equation (2) to the experimental data above 170 K (note the dotted line in figure 5) yields

the parameters $\rho_0 + \rho_0^{\infty} = 4.31$ mΩ cm and $c_K = -0.29$ mΩ cm K^{-1}. If the value of the residual resistivity estimated from the low-temperature fit is accepted, then the spin disorder contribution amounts to $\rho_0^{\infty} = 3.15$ mΩ cm. Apparently, both ρ_0^{∞} and c_K are much larger than usually found for Kondo systems, which is a direct consequence of the resistivity value of UPdSb that is about an order of magnitude larger than those typical for Kondo compounds [15].

Just this large resistivity, which is rather of the magnitude characteristic of small-gap semiconductors (see e.g. [16]) may suggest that UPdSb belongs to this class of materials. Hence the experimental $\rho(T)$ data above T_C were analysed in terms of the formula

$$1/\rho(T) = \sigma_a + B \exp\left(\frac{-E_a}{2k_B T}\right) \tag{3}$$

that describes excitations of charge carriers over the energy gap E_a near the Fermi level. The result of the fit is presented in figure 5 as the dashed line, and the obtained fit parameters are as follows: $\sigma_a = 0.344$ (mΩ cm)$^{-1}$, $B = 0.129$ (mΩ cm)$^{-1}$ and $E_a = 70$ meV. The value of E_a is much smaller than expected for intrinsic semiconductors, and therefore if UPdSb is really a semiconductor then its intrinsic regime is probably not reached up to room temperature. On the other hand, one should note that at 300 K the thermally activated contribution is only a small fraction (about 10%) of the total electrical conductivity of the sample studied, which is dominated by the temperature-independent term. Therefore the above numerical results should be treated with caution, despite the quite good quality of the obtained description of $\rho(T)$.

The appearance of the energy gap in UPdSb seems to be in contradiction with a nonzero value of the electronic specific heat coefficient ($\gamma = 62$ mJ mol^{-1} K^{-2} [7, 10]). Another argument against this compound being a semiconductor is the magnitude of its thermoelectric power that does not exceed 8 μV K^{-1} (see below). Even though similar systems are known in the literature for which the semiconducting state has been established, despite them showing very small values of the Seebeck coefficient (a prominent example is UPtSn; [17]), this scenario seems highly questionable for UPdSb because of all the problems raised above.

An alternative mechanism that yields a negative temperature coefficient of resistivity is the presence in a metallic system of some atomic disorder [18]. Actually, the temperature dependence of the resistivity of UPdSb in the paramagnetic state resembles the behaviour of paramagnetic metallic glasses or amorphous ferromagnets [19, 20]. Most recently glassy electronic transport was reported for URh_2Ge_2, which is a moderately disordered heavy fermion compound [21]. The electrical behaviour of such amorphous systems was accounted for by considering quantum corrections to the normal Boltzmann conductivity, namely electronic interactions and localization effects [22, 23]. According to the approach developed in [19], the electrical conductivity of a system governed by these two scattering mechanisms is given by

$$\sigma(T) = \frac{e^2}{2\pi^2\hbar}\left[3(A^2 + B^2T^2)^{\frac{1}{2}} - 3BT + CT^{\frac{1}{2}}\right] + \sigma_0 \tag{4}$$

where

$$A = \left(\frac{1}{D\tau_{so}}\right)^{\frac{1}{2}}$$

$$B = \left(\frac{1}{4D\beta}\right)^{\frac{1}{2}}$$

and [21]

$$C = 0.7367\left(\frac{k_B}{\hbar D}\right)^{\frac{1}{2}}$$

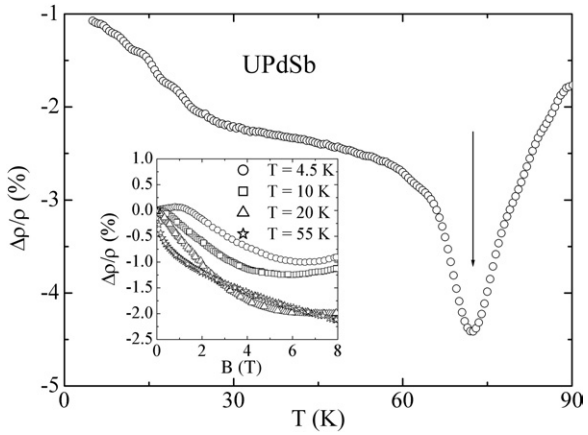

Figure 6. Temperature dependence of the transverse magnetoresistivity of UPdSb measured in an applied magnetic field of 8 T. The arrow indicates the ferromagnetic phase transition. Inset: magnetic field variations of the magnetoresistivity taken at several different temperatures.

where τ_{so} and $\tau_i = \beta T^{-2}$ are the spin–orbit and inelastic scattering times, respectively (it was assumed that τ_i is dominated by electron–phonon scattering) and D stands for the diffusion coefficient (in the definition of the parameter C it was assumed that in the weak localization regime electron–electron and electron–phonon interactions are negligible [19–21]). Fitting equation (4) to the experimental data above 80 K (see solid line in figure 5) yields the values $\sigma_0 = 0.34\ (\mathrm{m\Omega\ cm})^{-1}$, $\tau_{so} = 6.07 \times 10^{-12}$ s, $\beta = 9.9 \times 10^{-9}$ s K^2 and $D = 2.1 \times 10^{-4}$ m^2 s^{-1}. The scattering times thus obtained are similar to those characteristic of metallic glasses like the V_xSi_{1-x} alloys [19]. In turn, the diffusion coefficient is larger by about an order of magnitude than those found for V_xSi_{1-x} [19] that may reflect a difference in the density of states near the Fermi level in the two systems. These findings seem to indicate that the transport properties of UPdSb are similar to those previously observed in 3D amorphous metals, although the large value of the temperature-independent contribution σ_0 (nearly same as that derived in the 'semiconductor' scenario, i.e. the two different approaches suffer from similar problems) hampers any definitive statement on this point.

The temperature dependence of the transverse ($B \perp i$) magnetoresistivity (MR), defined as $\frac{\Delta\rho}{\rho} = \frac{\rho(B)-\rho(0)}{\rho(0)}$ and measured in an external magnetic field $B = 8$ T, is presented in figure 6. Apparently, MR is negative in the whole experimental temperature range and $\frac{\Delta\rho}{\rho}(T)$ shows a deep minimum of about -4.5% at the Curie temperature. Such a behaviour is characteristic of ferromagnets and was theoretically predicted by Yamada and Takeda [24]. According to the theory, the negative MR arises due to suppression by the magnetic field of spin fluctuations. The inset to figure 6 displays a few MR isotherms measured as a function of magnetic field at different temperatures from the ordered region. The $\frac{\Delta\rho}{\rho}(B)$ curves follow the behaviour expected for ferromagnets, except for a positive maximum in MR taken at 4.5 K in low magnetic fields. The latter feature most probably arises from the presence of a compensated domain structure, which hardly reconstructs upon applying a weak field (see discussion above).

Figure 7 shows the temperature variation of the thermoelectric power of UPdSb. At room temperature the Seebeck coefficient is small, being only about 2 μV K^{-1}. The positive sign of S might suggest that the dominant charge carriers are holes, yet a specific electronic structure near the Fermi level would also yield positive thermopower for electron-dominated transport [25]. Between 300 K and the Curie temperature the thermopower is nearly

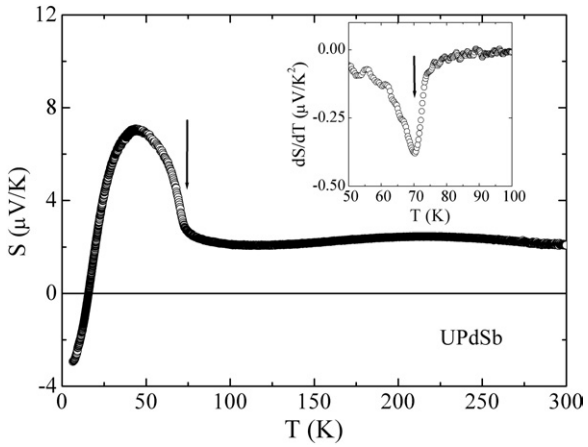

Figure 7. Temperature dependence of the thermoelectric power of UPdSb. Inset: temperature derivative of the thermopower in the vicinity of T_C. The arrows indicate the ferromagnetic phase transition.

independent of temperature. Below T_C the Seebeck coefficient starts to increase rapidly, in the vicinity of 40 K it reaches a maximum value of 7 μV K^{-1}, and then decreases abruptly changing its sign to negative at 15 K. At 0 K the thermopower is zero, thus a minimum of $S(T)$ must exist below 6 K. The inset to figure 7 shows the temperature dependence of the derivative of the thermoelectric power in the vicinity of the magnetic phase transition with a distinct minimum at T_C. A large hump of $S(T)$ below the ferromagnetic transition in UPdSb may be explained by taking into account inelastic scattering of electrons by phonons and by localized spins [26, 27].

4. Summary

UPdSb orders ferromagnetically at $T_C = 77$ K. Irreversibility in the magnetization measured in the FC and ZFC modes, negative signal observed at low temperatures in the ZFC mode, a wide magnetisation loop and a sharp transition at 1.8 T are all together indicative of a compensated narrow domain-wall structure. Such a behaviour is expected for compounds in which the anisotropy energy is much larger than the exchange energy. At the ferromagnetic phase transition distinct anomalies occur on the $\sigma(T)$, $\rho(T)$, $\frac{\Delta\rho}{\rho}(T)$ and $S(T)$ curves, and the value of T_C determined from the electrical data is slightly lower than that found by means of magnetic measurements. This finding may imply different sensitivity of the magnetic and transport characteristics to long-range magnetic ordering.

The Sommerfeld coefficient reported for UPdSb is rather large, being 62 mJ mol^{-1} K^{-2} [7, 10], and its electrical resistivity above T_C exhibits a negative temperature coefficient. For this reason one should consider the possibility that the compound studied belongs to a class of dense Kondo systems for which both an enhanced Sommerfeld coefficient and a decrease of the resistivity with increasing temperature are expected. In such a case UPdSb would be a representative of the very same group as that formed by the related UTM compounds UPdIn, UPdGa, UPdSi and UPdGe (see section 1). The latter interpretation is, however, hardly consistent with the strongly ferromagnetic properties of UPdSb, the high magnitude of its resistivity as well as with the exponential rather than logarithmic behaviour of $\rho(T)$ at high temperatures. Another possible scenario for the occurrence of $\mathrm{d}\rho/\mathrm{d}T < 0$ is the

formation of a small energy gap near the Fermi level. The analysis of the experimental data for UPdSb yielded a gap of 70 meV that seems a reasonable value for this type of compound [16]. However, the activation contribution to the total conductivity is very small compared to the temperature-independent term, the presence of the energy gap is inconsistent with the high value of the electronic specific heat, and the Seebeck coefficient exhibits rather metal-like and not semiconductor-like behaviour. On the other hand, high magnitudes of the electrical resistivity, small values of the thermopower and enhanced linear coefficients in the heat capacity are fingerprints of metallic glasses. Some degree of atomic disorder is indeed present in UPdSb because in its unit cell the Pd and Sb atoms jointly occupy a single crystallographic site. Actually, the experimental resistivity data can be reasonably well described by the theory that comprises quantum interference and electron–electron interaction effects. The values of the spin–orbit and inelastic scattering rates, extracted from the fits, are similar to those typical for metallic glasses. On this basis we tend to believe that UPdSb is not a semiconductor but rather a moderately disordered metal. Further studies, preferably optical conductivity and Hall effect measurements, would be helpful to verify the metallic character of this interesting compound.

Acknowledgments

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