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Electronic properties of UPdSn diluted by Lu

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Abstract

Lu substitutions for U (up to 30%) in UPdSn lead to a linear decrease of the lattice volume. After the initial decrease, the magnetic ordering temperature approximates to 25 K. The U-effective moment weakly decreases, whereas the paramagnetic Curie temperature becomes progressively more negative. The γ-coefficient (5 mJ mol^{-1} K^{-2} for UPdSn) dramatically increases to 78 mJ mol^{-1} K^{-2} in $U_{0.85}Lu_{0.15}$PdSn proving that the low γ-value in the parent compound is an accidental feature of the 5f band and not a fingerprint of the 5f localization. © 1998 Elsevier Science S.A.

Keywords: Lu; Lattice volume; UPdSn

1. Introduction

After a critical screening, the extended search for a UTX compound with real 5f localization yields one candidate, namely UPdSn. This compound is an antiferromagnet with T_N=40 K which undergoes another magnetic phase transition at 27 K. On the basis of the magnitude of effective moment (3.3 μ_B/U), ordered moment ($\approx$2 μ_B/U) and a relatively large magnetic entropy estimated as $1.5\times R \ln 2$ [1], it can be definitely classified as a local 5f moment system.

The low value of the electronic specific heat γ=5 mJ mol^{-1} K^{-2} (similar value as recorded for UPd_3) can be attributed to the 5f states removed from the Fermi surface. Some uncertainty about the 5f localization remains even after electronic structure calculations [2] and photoelectron spectroscopy experiments [3,4], which have revealed a fingerprint of the 5f states at the Fermi level.

Most recently, the electronic structure calculations performed for the realistic magnetic structure [5] have demonstrated that, assuming the itineracy of the 5f states, a low value of γ=7.5 mJ mol^{-1} K^{-2} can be obtained as a result of a deep minimum of the density of electronic states at the Fermi energy. To learn more about the nature of the 5f states in UPdSn, we undertake currently a dilution study. Here we report on the initial phase of the study of the $(U_{1-x}Lu_x)$PdSn system.

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2. Experiment

As-cast samples prepared by arc melting of stoichiometric amounts of pure elements show a single-phase character up to x=0.25. The sample with x=0.30 shows already some spurious phases. The magnetic susceptibility between 1.6 and 300 K was studied using a SQUID and a Faraday balance magnetometer in fields up to 5 T. The specific heat measurements were performed by means of a semi-adiabatic method. The electrical resistivity was measured on spark-cut bars using an a.c. four-probe method.

3. Results

The concentration dependence of lattice parameters within the homogeneity range seen in Fig. 1 shows a weak monotonous contraction of the hexagonal lattice (structure type GaGeLi, which is the ordered ternary variant of the $CaIn_2$-type structure) preserving a constant c/a ratio. The decreasing unit cell volume is somewhat surprising taking in mind a considerably larger atomic radius of Lu (174 pm) compared to U (156 pm), but it is a matter of fact that, in analogous intermetallic compounds, the lattice parameters of uranium and heavy rare earth are similar.

Fig. 2 displays results of magnetic susceptibility measurements performed on randomly oriented powder samples. All $\chi(T)$ data can be in the paramagnetic range fitted by a modified Curie–Weiss law: $\chi(T)=C/(T-\Theta_p)+\chi_0$. As seen in Fig. 3, the value of the effective moment per

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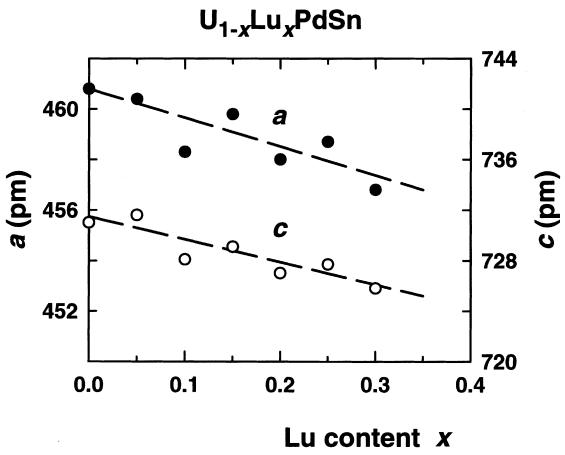

Fig. 1. Concentration variations of the lattice parameters of the GaGeLi structure type

1 U atom μ_{eff} decreases weakly with increasing Lu concentration, whereas the paramagnetic Curie temperature Θ_p becomes progressively more negative. One has to keep in mind that the data are affected by magnetic anisotropy (*c*-axis is the hard magnetization direction in UPdSn), which leads to μ_{eff} values somewhat smaller than obtained for a single crystal with the field along the easy axis. The values of the temperature-independent term χ_0 remains of the order of 3×10^{-9} m^3 mol^{-1} for all the concentrations studied. Inspection of the critical region around the magnetic phase transition shows a substantial change of the

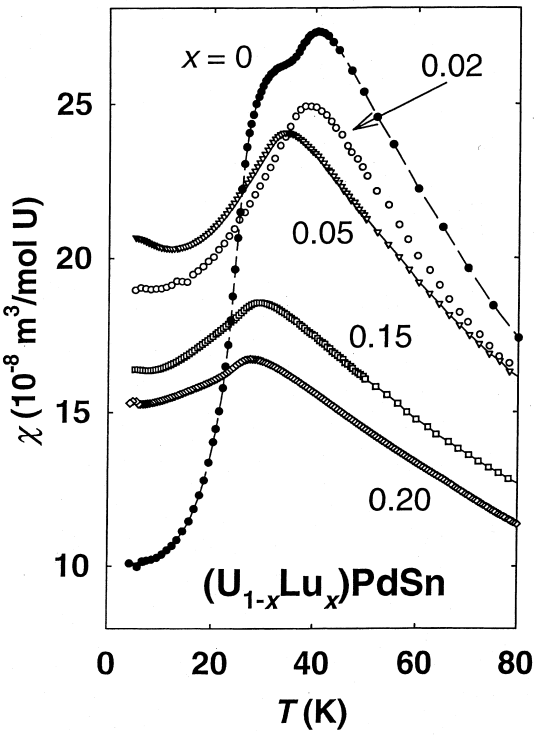

Fig. 2. Detail of the temperature dependence of magnetic susceptibility $\chi(T)$ measured in the field of 2 T for various Lu concentrations. Data compared with earlier data obtained on UPdSn polycrystal.

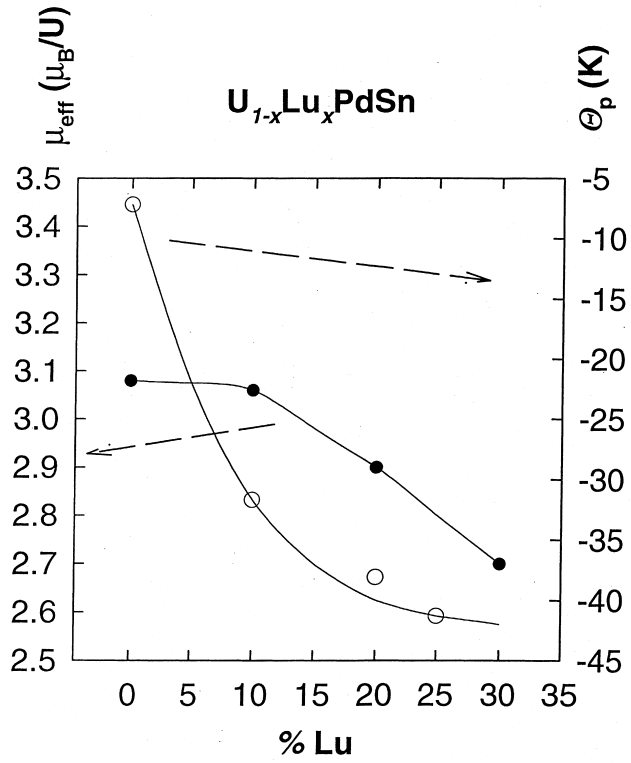

Fig. 3. Concentration dependence of effective moment μ_{eff} per U atom and paramagnetic Curie temperature Θ_p, obtained by fitting susceptibilities in the paramagnetic range to a modified Curie–Weiss law.

behaviour induced already by a 2% Lu substitution. The lower-temperature magnetic phase transition, which represents an onset of a new order parameter in the pure UPdSn [6,7], disappears completely in the Lu-substituted sample, and the low temperature susceptibility becomes substantially higher compared to the parent compound. The Néel temperature T_N decreases systematically with increasing Lu content, but for higher concentrations this tendency saturates, as seen in Fig. 4. Although we cannot be sure without a neutron diffraction experiment that the low-temperature magnetic phase from UPdSn is suppressed with Lu substitutions, the shape of the maximum on the $\chi(T)$ curve, which is more or less preserved throughout, makes the impression that the high-temperature antiferromagnetic phase of UPdSn is spread down to lowest temperatures in all Lu-substituted samples.

UPdSn has a distinct metamagnetic behaviour. A spin-flop transition is induced in fields above 3 T applied along the basal plane, and this transition is well seen also in a polycrystal. For 2% Lu a trace of this metamagnetic behaviour can be still observed (Fig. 5), but the overall effect is much weaker, and for higher Lu concentrations no metamagnetic transition could be seen in fields up to 5 T.

Temperature dependence of the specific heat has been studied for the sample $U_{0.85}Lu_{0.15}PdSn$ only. The comparison with original data on UPdSn is displayed in Figs. 6 and 7. The temperature of the anomaly related to the

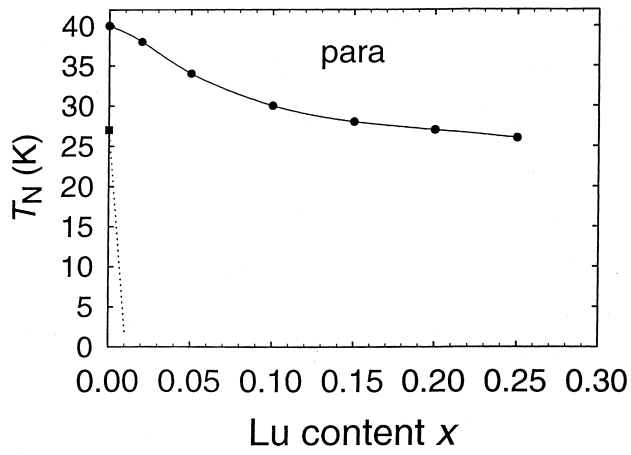

Fig. 4. Tentative magnetic phase diagram of the $(U_{1-x}Lu_x)$PdSn system. The dotted line represents schematically the fast suppression of the low-temperature magnetic structure of UPdSn. The temperatures of the order–disorder magnetic phase transitions were determined from maxima in $\chi(T)$.

antiferromagnetic phase transition is found at $T=28$ K, i.e. it coincides with the temperature of the maximum in $\chi(T)$. A remarkable effect is the enhancement of the γ-coefficient from 5 mJ mol^{-1} K^{-2} in pure UPdSn to 78 mJ mol^{-1} K^{-2} in $U_{0.85}Lu_{0.15}$PdSn (i.e. 92 mJ mol^{-1} U K^{-2}). This dramatic development of the γ-value corroborates the assumption that the very low γ-value in UPdSn is not due to the 5f localization, but comes out as a result of a more or less accidental narrow minimum in the density of itinerant 5f states at E_F. Then any modification of the DOS curve has a good chance to induce a dramatic γ enhancement.

The low-temperature detail of the specific heat data in the C/T vs. T^2 representation shows that for pure UPdSn a

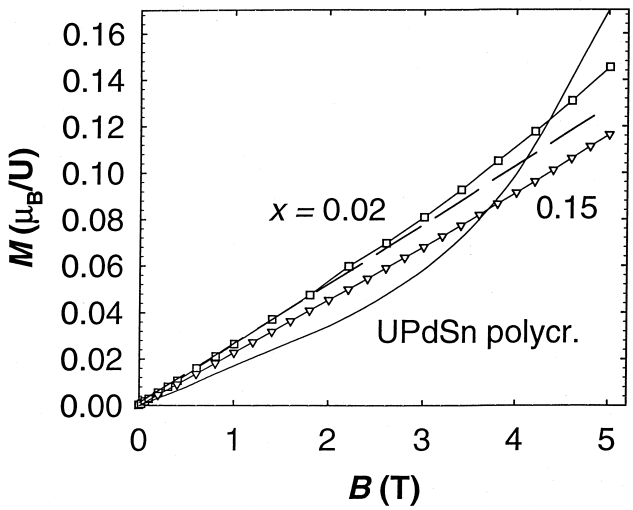

Fig. 5. Field dependence of magnetization at $T=4.2$ K of $U_{0.98}Lu_{0.02}$PdSn (squares) and $U_{0.85}Lu_{0.15}$PdSn (triangles) compared with earlier data obtained on polycrystalline UPdSn (full line). The dashed line represents the initial slope of $M(B)$ for $U_{0.98}Lu_{0.02}$PdSn.

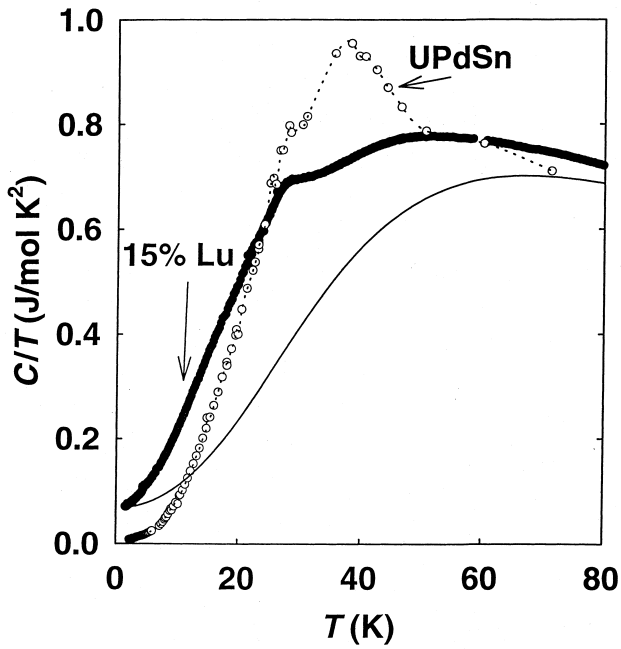

Fig. 6. Temperature dependence of the specific heat in the C/T vs. T representation for $U_{0.85}Lu_{0.15}$PdSn (full symbols) compared with earlier data on UPdSn (empty symbols). The full line represents the Debye curve adjusted for real γ in $U_{0.85}Lu_{0.15}$PdSn, representing a possible non-magnetic background specific heat.

straight line is followed only up to about 6 K. At higher T, an additional exponential term arising from collective excitations of the magnon type starts to be noticeable. The width of the gap $\Delta=48$ K [8,9], understood as the anisotropy gap in the magnon spectrum, corresponds to the anisotropy energy obtained from analysis of the magnetic

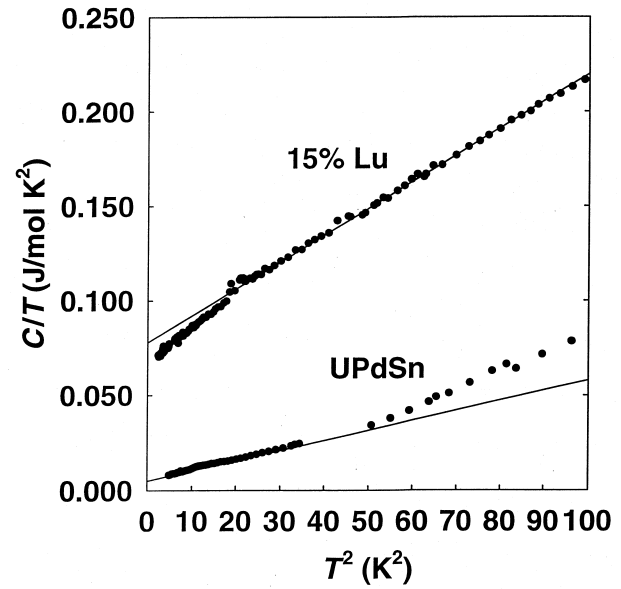

Fig. 7. Temperature dependence of the specific heat in the C/T vs. T^2 representation for $U_{0.85}Lu_{0.15}$PdSn compared with earlier data on UPdSn. Full lines represent linear extrapolation to $T\rightarrow 0$ used for the evaluation of the γ-coefficient.

susceptibility for the field along different crystallographic directions in the paramagnetic range. In the case of $U_{0.85}Lu_{0.15}PdSn$, the C/T vs. T^2 plot is linear between approx. 4 and 12 K. Above 12 K it starts to turn down already, which is probably due to the fact that the Debye temperature is somewhat lower than in UPdSn, as seen from the higher slope of the linear part. Thus the range in which the Debye function is approximated by a line is shorter. We have currently no clear explanation for the downturn at very low temperatures in the $U_{0.85}Lu_{0.15}PdSn$ data.

The electrical resistivity of UPdSn shows a broad knee around T=40–50 K, followed by a saturation at the high-temperature side. Absence of any sharp anomaly at T_N can be explained tentatively as due to the magnetic structure, in which one component of U moments remains probably disordered in the intermediate range 25–40 K. The room-temperature resistivity value exceeding 600 μΩ cm serves a clear indication of presence of the 5f states at E_F, and their hybridization with conduction electron states. The absolute resistivity values even increase with Lu substitution for U. We should note that the intrinsic absolute values need not be as high as those shown in the Fig. 8 beause they are seriously affected by a number of internal cracks in samples. However, a qualitative analysis shows that the dominant anomaly in $\rho(T)$ becomes more associated with T_N. This fact can mean that the magnetic structure of the Lu-substituted samples need not be completely identical with the high-temperature magnetic structure of UPdSn, and that the magnetic phase diagram sketched above should be taken with caution.

The resistivity drop below T_N is gradually reduced with increasing Lu concentration, and the high-temperature slope of the $\rho(T)$ curve becomes negative. Finally, for 25% Lu the negative $d\rho/dT$ is maintained over whole temperature range studied. Both features are observed typically in the case of incoherent strong f scatterers at E_F.

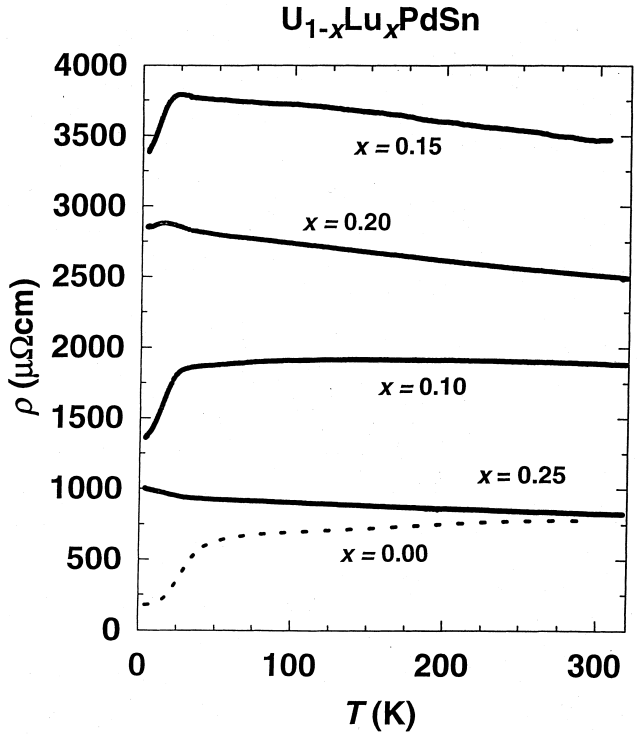

Fig. 8. Temperature dependencies of electrical resistivity $\rho(T)$ for various Lu concentrations. Data compared with earlier data obtained on UPdSn polycrystal (dashed line).

4. Discussion and conclusions

The impact of Lu substitution for U in UPdSn should be seen in two levels. In the first level we observe a dramatic change of the γ-coefficient and significant variations of magnetic properties, like the stronger anisotropy seen in suppression of the exponential term in specific heat (Δ probably strongly increases) and absence of metamagnetism in Lu-substituted materials. Also, the evolution of the Θ_p value (becoming more negative), reflecting the character of exchange interactions, shows a general strengthening of the antiferromagnetic coupling. The μ_{eff} value as the main 5f ion characteristic also somewhat decreases. All these changes (except for loss of the lower-temperature magnetic phase transition in UPdSn) proceed in a gradual way, nothing resembles any qualitative change expected at a possible 5f localization–delocalization transition.

When assessing a possible development of the 5f electron delocalization, we can expect that the 5f states of U ions become more delocalized and their magnetic moments diminish with proceeding Lu substitution. This would point to a stabilizing effect of the 5f–5f interaction (albeit mediated by non-f states) on the magnetic moment. A supporting argument is also a rather strong reduction of the lattice parameters, which can be related to a reduced U atomic radius resulting from the washing out of the 5f electron density within the U atomic spheres.

Acknowledgements

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