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Magnetic Properties of $URu_{1-x}Pd_xGe$ Solid Solutions

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The magnetic properties of the solid solutions $URu_{1-x}Pd_xGe$ with $x = 0.1 - 0.9$, crystallizing in the orthorhombic TiNiSi-type structure, were investigated by ac magnetic susceptibility and dc magnetization measurements. The experimental data reveal that there are three different regions of magnetic behaviour. A nonmagnetic ground state occurs for $x < 0.3$, however, as the Pd concentration is increased, the antiferromagnetism is stabilized for $x = 0.35 - 0.8$, and two successive transitions, antiferro- and ferromagnetic, emerge in the $x = 0.9$ composition.

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1. Introduction

The ternary UTM compounds, where T = d-electron transition metals and M = p-electron metalloids, show a wide variety of magnetic behaviour, depending on the T and M ions involved. It was established that hybridization between the $5f$- and conduction electrons is the main factor determining the physical properties of UTM at low temperatures [1]. In fact, the hybridization has to fulfill a dual role; both destroy the localized U moments and mediate the exchange interaction between magnetic moments. A series of compounds URuGe–URhGe–UPdGe may exemplify the important role of the hybridization, since with increasing the d-electrons numbers there occurs a change from a nonmagnetic state in URuGe through a ferromagnetic/superconducting in URhGe to a ferro/antiferromagnetic one in UPdGe. Interestingly, recent works on the solid solutions $URu_{1-x}Rh_xGe$ [2, 3] have shown that before the ferromagnetism appears, the system exhibits non-Fermi liquid (nFl) behaviour around the magnetic quantum critical point (QCP) at $x \approx 0.62$. Moreover, we have pointed out that spin-fluctuations, as a precursor, take place before the system gets nearer QCP [4].

Since, similar evolution in the ground state might be expected for $URu_{1-x}Pd_xGe$, we have studied the alloys. In this contribution, we report the results of x-ray and ac-magnetic susceptibility measurements in this system.

2. Experimental details

Polycrystalline samples of the $URu_{1-x}Pd_xGe$ solid solutions with $0.1 \leq x \leq 0.9$ were synthesized by arc-melting the appropriate amounts of U (2N8), Ru (4N) Pd (3N) and Ge (5N) under a Ti-gettered pure argon atmosphere. The samples were remelted several times to improve homogeneity. Since annealing of samples at 800 °C for 9 days had worsened their quality, we present here the data obtained on unnealed samples. Mass losses during sample preparation were less than 0.5%. Powder x-ray analysis revealed that all synthesized samples crystallize in the same crystal (TiNiSi-type) structure (space group Pnma) as that of border compounds URuGe and UPdGe. The room temperature lattice parameters were determined by a least-squares fit to the positions of the observed Bragg peaks. It is found that upon the substitution of Ru by Pd the unit cell volume increases by $\sim 6.6\%$.

Measurements of ac-susceptibility were performed in the temperature range 2–70 K using an Oxford Instrument susceptometer. AC driving field amplitude of 10 Oe and frequency of 1 kHz were applied. DC magnetization measurements were made in fields up to 5.5 T using a Quantum Design SQUID magnetometer.

3. Results and discussion

In Fig. 1a we show the temperature dependence of real $\chi'(T)$ and imaginary $\chi''(T)$ parts of the ac-susceptibility for $x = 0.2$–0.32. It can be seen that the amplitude of the susceptibility increases with x, indicating the development of the magnetic correlation between uranium ions.

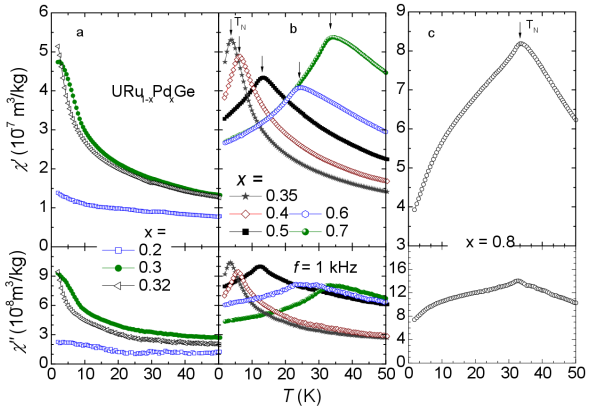

Fig. 1. Temperature dependence of real and imaginary components of the ac-magnetic susceptibility of solid solutions $URu_{1-x}Pd_xGe$ for $x = 0.2$–0.8.

With further increase of the Pd content up to $x = 0.8$, a sharp maximum appears in both $\chi'(T)$ and imaginary $\chi''(T)$ curves, announcing magnetic phase transition in these alloys. The position of the maximum increases with x, from 3 K at $x = 0.35$ and attains maximum value of 34 K at $x = 0.7$ (see Fig. 1b). Because isothermal

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magnetization at 2 K depends weakly on applied field (not shown here), we may interpret the ac-susceptibility anomaly, as due to an antiferromagnetic ordering of the uranium moments at T_N. However we may add, that an ordinary antiferromagnet should not provide any contribution in mean field phase transition to the $\chi''(T)$ part, thus the observed loss maximum may indicate the existence of a ferromagnetic component in the antiferromagnetic state. Figure 1c presents the ac-susceptibility data for $x = 0.8$. The $\chi'(T)$ and $\chi''(T)$ curves show a pronounced maximum at 32 K and a knee around 10 K. The amplitude of both the ac-susceptibility parts acquires large values, i.e, about 8×10^{-7} m^3/kg at the maximum. Clearly, the $x = 0.8$ sample exhibits different behaviour to those of $x = 0.35$–0.7. The most likely way to explain this is that the ferromagnetic component in this composition becomes more serious.

Looking at the ac susceptibility data for $x = 0.9$, shown in Fig. 2a, a pronounced maximum at 30 K in the $\chi'(T)$ curve is observed. This anomaly may signal a ferromagnetic order in the alloy, contrary to the antiferromagnetic one in the compositions considered above. The field dependence of the magnetization at selected temperatures, shown in Fig. 2b, may support the ferromagnetic order at $T_C = 30$ K. However, for $T < 20$ K, $M(H)$ shows a jump at a critical field H_{cr}, which suggests that a spin-flop in the metamagnetic transition happened in antiferromagnets. Obviously, H_{cr} is shifted down as temperature increases. For $H < H_{cr}$, a tendency to saturation of the magnetization is observed. At a field of 5.5 T, M at 2 K achieves a value of $0.7\mu_B$. As magnetic field strength reduces, the composition shows hysteresis and at 0 T there remains still a high remnant magnetization, pointing to a large magnetocrystalline anisotropy of the material.

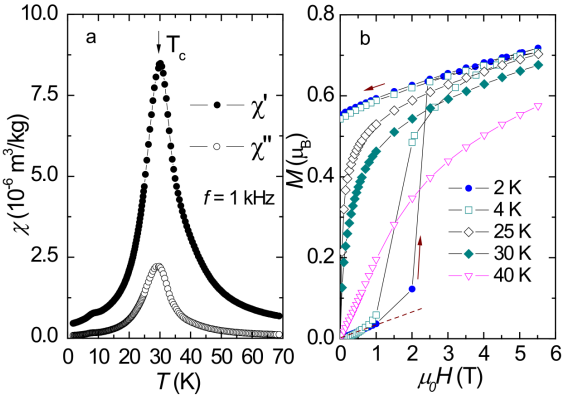

Fig. 2. a) ac-susceptibility vs temperature and b) isothermal dc-magnetization of $x = 0.9$ at several temperatures.

Some more information on the magnetic ground state can be attained if one compares the ac-susceptibility (Fig. 2a) with the fixed field data (Fig. 3a). Usually, T_C is defined as the inflection point of the $M(T)$ measured in a low magnetic field. The T_C value determined by this method agrees well with the ac-susceptibility maximum. Apparently, T_C increases with increasing applied magnetic field, and that matches the behaviour of typical ferromagnets. Likewise, if considering the temperature derivative of the magnetization dM/dT, one distinguishes another magnetic phase transition T_N, which is ascertained as the maximum of dM/dT. This anomaly shifts down to lower temperatures as field increases (see Fig. 3b), and perhaps originates from an antiferromagnetic ordering. Neutron diffraction experiments will be needed to prove the suggestion.

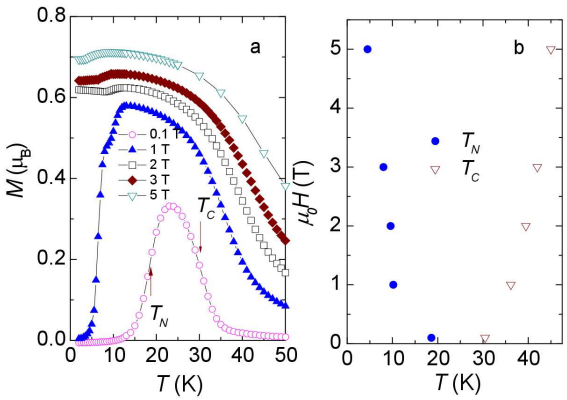

Fig. 3. a) Isofield magnetization vs temperature and b) the $H - T$ magnetic phase diagram for $x = 0.9$.

4. Conclusions

We have shown that the $URu_{1-x}Pd_xGe$ solid solutions crystallize in the orthorhombic TiNiSi-type structure. The ac-magnetic susceptibility and dc magnetization measurements allow us to stress the following points: a) a nonmagnetic-magnetic crossover occurs around $x \sim 0.32$; b) with increasing the Pd concentration up to $x = 0.8$, the antiferromagnetic order with increasing ferromagnetic components sustains; c) the $x = 0.9$ composition undergoes two successive magnetic phase transitions at $T_N = 18$ K and $T_C = 30$ K. Our experimental results suggest that further investigations are needed, as regards the spin configurations, as well as in relation with the issue of eventual emergence of nFL/QCP in this system as a consequence of possible competing Kondo and exchange interactions.

Acknowledgments

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